

**SECOND HARMONIC GENERATION INDUCED BY
SURFACE PLASMA WAVES OVER A METALLIC
SURFACE**

Thesis Submitted For the Award of the Degree of

DOCTOR OF PHILOSOPHY

in

PHYSICS

By

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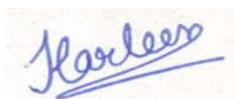
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DECLARATION

I, hereby declare that the work reported in the thesis entitled “**Second harmonic generation induced by surface plasma waves over a metallic surface**” in fulfillment of degree of **Doctor of Philosophy (Ph.D.)** is outcome of research work carried out by me under the supervision of **Dr. Vishal Thakur**, working as Professor, in the Department of Physics of Lovely Professional University, Phagwara, Punjab, India. In keeping with general practice of reporting scientific observations, due acknowledgements have been made whenever work described here has been based on findings of other investigator. This work has not been submitted in part or full to any other University or Institute for the award of any degree.



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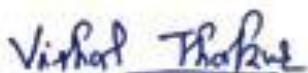
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CERTIFICATE

This is to certify that the work reported in the Ph.D. thesis entitled “**Second harmonic generation induced by surface plasma waves over a metallic surface**” submitted in fulfillment of the requirement for the reward of degree of **Doctor of Philosophy (Ph.D.)** in the Physics Department, is a research work carried out by **Harleen Kaur Dua**, 41800916, is bonafide record of her original work carried out under my supervision and that no part of thesis has been submitted for any other degree, diploma or equivalent course.



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ABSTRACT

The laser's interaction with matter has gained much importance in the past. It has become a field of interest for researchers due to its wide range of applications in the field of medical science, material science, laser based phenomenon etc. It involves the action of laser light on matter where matter can be gas, metals, dielectrics etc. The fields of laser interact with electrons and ions of matter forming plasma. These interactions are nonlinear in nature. The nonlinearity arises due to the presence of collisions and random motion of charge particles. The charge particles mainly electrons experience drift due to the electromagnetic fields of laser and give rise to many interesting phenomenon like harmonic generation which could further be used for various applications. In the present work we have investigated second harmonic generation on a metallic surface with the aim to design a model which could give an efficient and cost effective method for harmonic generation. It involves the interaction of laser with metal from glass-metal interface through Kretschmann attenuated total reflection configuration which leads to the formation of metal plasma. The electromagnetic fields of laser exert ponderomotive force on charge carriers particularly electrons leading to drift of electrons and hence their velocity changes. The electrons in the metal on interacting with laser leads to the formation of surface plasma waves (SPW). The fields of surface plasma waves are evanescent in nature, which decay rapidly away from the surface. The SPW have electromagnetic fields associated with it which could further interact with the electrons of the semiconductor, which is layered over metal. The nonlinear interactions among the SPW and electrons of semiconductor lead to second harmonic generation. The configuration chosen for the present study is Kretschmann attenuated total reflection configuration so that the maximum part of incident light gets reflected. The metals chosen for the study are Copper (Cu), Silver (Ag) and Aluminium (Al) with semiconductor Indium antimonide (n-InSb) as thin layers of the order of few nanometers. The properties of metals like reflectivity, absorbance and electron density decides the strength of surface plasma wave formed which further influences the second harmonic generation. The present model shows that metal-semiconductor interface could be a better choice for the generation of harmonics. The aim of the present work is to have a cost effective and an efficient method of harmonic

generation which could be used for medical purposes to help mankind. Four different objectives are depicted in the thesis, each underpinned by laser-plasma interaction governed by ponderomotive mechanism. The methodology and hypothesis is explained in chapter 2.

The objectives are chosen such that it covers the different aspects of second harmonic induced by surface plasma waves. The first objective (chapter 3) includes the study of second order harmonic generation induced by the SPW at the interface of metal and semiconductor with rippled metal surface and also the effect of plasma frequency on amplitude variation. The presence of density ripple amplifies nonlinearity which is advantageous for harmonic generation. By considering the mechanism to be ponderomotive, the nonlinear current density has been calculated and wave propagation studied using second order wave equation for two media. The investigation is done to study the dependence of normalized second harmonic amplitude with the normalized propagation distance at different instances of time and we showed that the SHG gives good enhancement in amplitude for Cu-InSb interface as compared to Ag-InSb and Al-InSb interfaces. The second objective (chapter 4) studies the effect of magnetic field particularly wiggler magnetic field on SHG. It has been observed that the presence of wiggler field provides phase matching conditions which helps to resonate the process of harmonic generation. Wiggler magnetic field of suitable period exerts force on electrons and oscillates them by changing their phase to make the process resonant. Under the effect of wiggler field Cu-InSb gave more enhancement in the amplitude as compared to the other two. Also, it has been observed that as the strength of field increases the normalized amplitude also increases and under optimal conditions increase is more for metal-semiconductor interface as compared to other plasma. In the third objective (chapter 5) the SHG by crossed SPW have been studied. Laser's interaction with electrons of metal lead to formation of surface plasma waves propagating at different angles with respect to z-axis. The two such SPW inclined at angles θ and $-\theta$ with respect to z-axis are considered which upon superposition give a SPW which further undergoes the interaction process with the electrons of semiconductor. The ripple density of suitable wave number is considered to compensate for the phase change. The dependence of resonant SPW generation on the angle has been observed and it was found that the

SPW which propagate parallel to z-axis could give efficient SHG and as the angle increases the efficiency of harmonic generation decreases. The surface plasma wave whose phase matches with the phase of oscillating electrons leads to further interaction with the semiconductor thereby generating second harmonic. Further, out of the three interfaces the Cu-InSb interface shows higher amplitude enhancement. The objective 4(chapter 6) investigates the effect of self focusing of laser on SHG induced by SPW. The laser with the Gaussian profile has been chosen. The beam width parameter varies periodically with propagation distance showing periodic focusing and de-focusing. Presence of the nonlinear interactions among laser fields and electrons accounts for the self focusing effect of the laser beam. It was found that the regions with low value of beam width parameter have focused SHG with high amplitude. The idea is that the focused laser beam will induce focused surface plasma waves which further show good impact on the second harmonic generated at the metal-semiconductor interface. Hence, a focused and an efficient harmonic is generated. On studying the variations among the normalized second harmonic amplitude with normalized propagation distance along with variation of beam width parameter, it was found that Cu-InSb interface gives efficient and focused output. The next chapter (chapter 7) is dedicated to summary and future scope. Our work is based on mathematical and graphical analysis using the optimized parameters from the previous research works. In our work, the results obtained show that under the given conditions, metal-semiconductor interface could give focused and efficient harmonic generation. The present analysis shows that metal-semiconductor interface in Kretschmann configuration could be a better choice for harmonic generation. Hence, it opens new options for research in future.

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LIST OF SYMBOLS

$-e$	Electric charge
m	Mass of electron
ε_m	Permittivity of metal
ε_s	Permittivity of semiconductor
n	Refractive index
c	Speed of light
Cu	Copper
Ag	Silver
Al	Aluminium
InSb	Indium antimonide
$\vec{E}$	Electric field of laser
$\vec{B}$	Magnetic field of laser
$\vec{E}_s$	Electric field of surface plasma wave
$\vec{E}_2$	Electric field of second harmonic generation
F	Ponderomotive force
ω_1	Frequency of laser
ω	Frequency of surface plasma wave
ω_p	Plasma frequency
k_1	Propagation constant of laser
k_s	Propagation constant for surface plasma wave
A	Amplitude of electric field of laser
A_0	Fundamental amplitude of laser
A_s	Amplitude of electric field of surface plasma wave
A_2	Amplitude of second harmonic generation
J	Current density

n_0	Electron density
q	Ripple wave number
z'	Normalized propagation distance
$\vec{B}_w$	Wiggler magnetic field
B_0	Amplitude of wiggler magnetic field
k_0	Wiggler wave number
τ	Laser pulse width
v_g	Group velocity of Gaussian profile
f_l	Beam width parameter of the laser pulse
r_0	Spot size of the laser at $z = 0$

ABBREVIATIONS

EM waves	Electromagnetic waves
SPP	Surface plasmon polaritons
SPW	Surface plasma waves
SPR	Surface plasmon resonance
SERS	Surface Enhanced Raman Scattering
ATR	Attenuated total reflection
SH	Second Harmonic
SHG	Second Harmonic Generation
nm	Nanometer
n-InSb	N type Indium Antimonide
Nd:YAG laser	Neodymium-doped Yttrium Aluminium Garnet Laser

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CHAPTER 1

INTRODUCTION AND OVERVIEW

1.1 Introduction and prior research

The interaction of laser and matter has given rise to many phenomenon and has become a topic of research in the past few decades. The nonlinear interaction of laser with the matter has gained much attention because of its unique applications like high harmonic generation, laser driven plasma based accelerators, sensors, photoelectron spectroscopy, etc.[1]–[5]. The electromagnetic fields of laser interact with particles of matter and accelerate them. The particles of matter can be ions in plasma or electrons in metal. Surface plasma waves (SPW) are formed when laser interact with electrons of metal. These are electromagnetic waves which propagate at the interface of two different media with different dielectric properties and conductivities. These are evanescent in nature and die with depth. The behavior of SPW is different in different media like they decay more rapidly in dielectric than in the metal. SPW over metallic surfaces have gained prominent applications in sensors, energy conversion, radiation generation, material ablation, etc. The study of SPW on metal-vacuum interface in Kretschmann attenuated total reflection configuration with metal on the glass prism has given rise to new phenomenon of frequency doubling[6]–[9]. These waves have amplitude higher than that of laser and frequency same as the laser. The high electric fields of SPW can lead to harmonic generation, wave mixing and Surface Enhanced Raman Scattering. In 1902 Wood observed some new features in light reflection measurements in metallic gratings. Then colours in metal doped glasses were observed by Maxwell Garnett. Further in 1908, theory of light scattering by spherical particles was given by Mie. In 1956 energy losses of fast moving electrons in metals were theoretically analyzed by Pines and he coined the term ‘plasmons’. In the same year term ‘polaritons’ was given by Fano representing the coupled oscillations of bound electrons with light in the transparent media. Different researchers Ritchie, Otto and Kretschmann studied behavior of surface plasmons on materials like thin films, metal films. Observations reveal that plasmon modes could exists near the surface of metals. Surface plasmon polaritons (SPP’s) for closed surface are known as

localized surface plasmon. SPP's can be excited by electrons or photons but for photons any of the agents like gratings, prisms or metal surface defects is needed as it can't be done directly. This interaction leads to production of harmonic waves [10]-[11].

The behavior of SPP in given metal-dielectric interface depends on frequency as for low frequency the dispersion relation remains same as in vacuum but for high frequency dispersion relation show changes and reaches at limiting value called plasma frequency. Propagation of SPP involves loss of energy by means of absorption or scattering. Their penetration depth is approximated by using skin depth formula, within the skin depth SPP's are sensitive to even very small disturbances and this property enables them to be used as probes to analyze the inhomogenities of surface. One more factor on which resonance of surface plasmon mainly of localized surface plasmon depends is shape of the particle[12]–[15].

Other process related to surface plasmon excitation is surface plasmon resonance (SPR) which involves analysis of extent of excitation by observing the reflected light from a prism coupler which further depends on incident angle and wavelength. The circuits based on SPR has reduced the size of components used in data processing by increasing their performance [16]–[18].

The use of metals like Ag, Ni, Au etc. for making the nano structures helps to localize the electromagnetic fields on lengths of the order of few nanometers that results in the enhancement in amplitude which can even exceed the amplitude of applied field in the vicinity of structures. Good response have been observed with metals like Ag, Au and also the metals like Co, Ti, Ni, metal oxides had been used but their efficiency of work is comparatively low[19]–[22]. At the metal-vacuum interface, the thermionic emission of electrons is caused by the energy imparted by surface plasma to the electrons. To attain the large amplitude for surface plasma wave a thin metal film can be deposited on one face of prism in attenuated total reflection Kretschmann configuration. Laser light is incident at an angle depending upon refractive index of prism and nature of metal which penetrates and interacts with electrons leading to SPW. Nature of waves generated depends upon incident frequencies and angles. The presence of external factors like density ripple on metal surface generates non linear current which can produce plasma wave of varying frequencies. Dielectric materials

with high refractive index can be considered as an alternative approach to metals due to their different properties at different frequencies [23]–[25].

In case of the semiconductors, the type of doping decides the charge carrier and the number density of charge carriers is one of the important characteristic of semiconductors. Laser's interaction with semiconductors leads to the formation of different types of plasma with different densities [26]–[28]. Kant *et al.* studied different types of plasmas under various conditions and their study concluded that the amplitude can be enhanced with the presence of density ripple. They also worked with different profiles of laser like Gaussian, Hermite Gaussian, Hermite-cosh Gaussian etc. Different factors like amplitude of laser, frequency of laser, charge carrier density etc. decides the amplitude and order of harmonic. Increase in the efficiency has been observed in the presence of agents like wiggler field, chirp pulse and density ripple [29]–[31]. Harmonic generation of third order in n-InSb and Ag showed that the enhancement in the electric field for n-InSb is more by factor of 2 than for Ag, the observation suggests that nonlinear charge density is more in semiconductors than metals. Higher harmonics can be observed at the interface of combination of metals with dielectrics where surface plasma waves generated in metals also helps in the process [32], [33].

Higher order harmonic generation is observed in both metals and semiconductors due to interaction of the charge particles particularly electrons with electromagnetic fields of light. Both materials could show nonlinear behavior that account for harmonic generation and other phenomenon. The metals with good conductive properties like Ag, Au, and Al or with good magnetic properties like Co, Ni etc. show harmonic generation upto good extent. In semiconductors charge concentration depends on doping which benefits the process. Recently, work of generation of waves in layered pattern like metal-dielectric interface, metal-metal oxide etc. have been reported and improvement in imaging techniques is the direct consequence of this work [34].

1.2 Review of literature

Chauhan *et al.* [35] studied surface plasma waves assisted second harmonic generation at metal vacuum interface using Kretschmann configuration in the presence of surface ripple and concluded that surface plasma waves excited by laser exerts ponderomotive

force on electrons and velocity so gained beats with surface ripple to produce nonlinear current which further lead to high harmonic generation.

Carletti *et al.*[36] showed that second harmonic generated from magnetic resonance in AlGaAs on aluminium oxide nanoantennas can be made resonant by varying height and radius of nanoantennas. The variation of harmonic generation with magnetic field also depends on the nature of material used. It affects the nonlinear interactions which depend on size of entities.

Fedotov *et al.*[37] demonstrated that the use of independent light sources for optical breakdown as well as for non-resonant harmonic and increase in pump intensity for laser increases the efficiency of high harmonic generation. As per their work harmonic generation takes place in the presence of strong laser field due to scattering of optical electrons by ions.

Polali *et al.*[38] studied surface plasma waves and explained that these waves are evanescent in nature. They also worked on intensity modulation methods using such fields. Their work showed the behavior of surface plasma polaritons in the medium and their dependence on different factors.

Thakur *et al.*[39], [40] have studied effect of density ripple and wiggler magnetic field on harmonic generation on materials like semiconductors, metals and showed that presence of these factors improves the efficiency of harmonic generation.

Ghaemi *et al.*[41] studied the efficiency of surface plasmons to enhance optical phenomena and found that it works better for wavelength comparable holes than for sub-wavelength holes.

Hibbins *et al.*[42] experimentally verified existence of surface plasmon mode due to localization of electric field at the interface.

Li *et al.*[43] studied second harmonic generation in a single silver nanowire monolayer molybdenum disulfide hybrid system using excited surface plasmon polaritons which are axially collimated but diverges transversally to the surface.

Pan *et al.*[44] studied second harmonic generation in silver-dielectric-gold-dielectric nanostructures forming centrosymmetric multilayered network and found that introduction of some gain media between layers compensates dissipation and leads to amplification of surface plasmon.

Pendry *et al.*[45] found that surface plasma waves for metals and for a conducting

surface perforated by holes are EM waves depending upon permittivity of plasma formed.

Kumar *et al.*[46] examined on metal vacuum interface the possibility of surface plasma waves by e-h combination in a forward biased p-n junction located near interface with thin layer of n type semiconductor between metal and p type material.

Singh and Tripathi[47] showed that when laser beams of different frequencies are made to strike metal surface at angles θ and $-\theta$ then surface plasma wave with frequency equals to their difference is formed and its frequency depends on incident frequencies and angles.

Jatav and Parashar[48] studied second harmonic generated by crossed surface plasma waves over a metal-free space interface. As per their work angle of mixing decides mode of harmonic generated and efficiency decreases for larger angles.

Vinogradov *et al.*[49] studied surface plasmon polaritons in Kretschmann configuration and found that wave incident at an angle greater than critical angle does not excite surface plasmon polaritons. If narrow beam incidents then SPPs formed due to interference of reflected plane waves are observed at the geometrical edge of beam. Huang *et al.*[50] showed that plasmonic metal-semiconductor interfaces can generate hot electrons. The fields of these electrons interact with electrons of semiconductors nonlinearly.

Bhatia *et al.*[51], [52] investigated second harmonic generation in relativistic plasma using Laguerre-Gaussian beam and found that exponential density ramp plays a vital role in focusing ability of beam and second harmonic yield. Also, they studied the influence of self focusing of Laguerre-Gaussian beam on second harmonic generation and observed enhanced amplitude in the regions of highly focused incident laser.

Kant and Thakur[53] investigated self focusing of Laguerre-Gaussian beam in relativistic plasma with exponential density ramp. Their work showed that the beam size of laser shrinks while propagating in plasma under the influence of exponential density ramp. The amplitude of harmonic generated enhances in the regions where focusing of beam occurs.

Ullah *et al.*[54] discussed different methods to tune and enhance the harmonic generation in low dimensional materials which have the potential to be used for information and communication purposes.

1.3 Motivation of the study

The interest of researchers in SPW technology is strongly motivated by the burgeoning applications in the science and industry. Many researchers have worked on metallic surfaces like metals, metal-vacuum interface as well as metal-dielectric interface but metal-semiconductor interface is a surface on which work is yet to be done related to harmonic generation. As studied by Huang *et al.* [50] metal-semiconductor interface generates energetic electrons, expectations are when these interfaces are irradiated with laser of particular wavelength may give rise to harmonic generation which can be used further.

Purpose of our research is to design a model with suitable materials and parameters which could help to improve efficiency of harmonic generation leading to better and cost-effective imaging techniques which can be used for medical purposes to help mankind.

1.4 Research objectives

The purpose of our research is to design a model with suitable materials and parameters which could help to improve efficiency of harmonic generation leading to better and cost-effective imaging techniques which can be used for medical purposes to help mankind. This will be achieved through the following objectives:

- 1) To study the second harmonic generation by surface plasma waves over a metal-semiconductor interface.
- 2) To study the effect of external magnetic fields on the efficiency of second harmonic generation by surface plasma wave.
- 3) To study the second harmonic generation by crossed surface plasma waves over a metallic surface with density ripple.
- 4) To study the effect of self-focusing on second harmonic generation by surface plasma wave.

1.5 Thesis contribution

The work presented in this thesis is based on theoretical approach to deal with the second harmonic generation induced by surface plasma waves on metallic surface. As

per our first approach the second harmonic generation is investigated on metal-semiconductor interface. The metal surface is rippled with suitable ripple wave number which helps to attain phase matching conditions. The SPW produced in metal could interact with electrons of semiconductor leading to nonlinear interactions. These nonlinear interactions results in harmonic generation. Second harmonic generation (SHG) with good amplitude enhancement is observed. Our second approach is based on harmonic generation on metal-semiconductor interface in the presence of wiggler field. The presence of field provides phase matching conditions and dependence of amplitude of SHG on wiggler field is observed. The third approach is based on the interaction of two SPW at an angle with laser's propagation direction and their influence on SHG at metal-semiconductor interface. Our fourth approach includes the investigation of the effect of self focusing of laser on SHG amplitude in the presence of rippled metal surface which helps to attain phase matching conditions.

1.6 Thesis outline

The thesis is organized as follows: the methodology is given in Chapter-2, along with the hypothesis and the theoretical consideration of basic equations used in the analysis. The configuration used for the study is Kretschmann attenuated total reflection has also been discussed in Chapter 2. The model with the detailed numerical analysis and observation of our results are discussed in Chapter-3, 4,5and 6 which includes the study of dependence of amplitude of harmonic generated on different factors like exponential density profile, wiggler magnetic field, crossed surface plasma waves and self-focusing effect of laser. Summary and future scope of the work accomplished in this thesis which includes the practical applications of the model is discussed in the Chapter-7.

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CHAPTER-2

RESEARCH METHODOLOGY

2.1 Formulation of hypothesis

The model is based on the generation of surface plasma wave by introduction of laser on glass-metal interface. The long-term goal of our research is to develop a model with suitable materials and parameters to improve efficiency of harmonic generation leading to better and cost-effective imaging techniques which can be used for medical purposes. Our specific hypothesis is that interaction of laser with metal generates surface plasma waves (SPW), which are electromagnetic waves with amplitude higher than the laser. The thin layer of n-type semiconductor is considered over metal surface in Kretschmann attenuated total reflection configuration (ATR). The interaction of SPW with electrons of n-type semiconductor which is layered over metal leads to harmonic generation. This hypothesis is based on the previous results of different experimental as well as theoretical work carried out by researchers in the field of harmonic generation. Equation for ponderomotive force is solved and nonlinear current density is derived. Using wave equation amplitude of second harmonic is calculated and solved numerically by using MATHEMATICA. The results are plotted in ORIGIN software.

Sources of data:

Theoretical results are compared with experimental and theoretical results collected by various researchers through their papers.

Research Design:

1) Analytical analysis

It involves derivation of equations for the propagation of wave under given conditions and effect of different parameters on the interaction of laser with particles of matter. It includes the study of phase matching of harmonic generated, effect of external factors and expression for normalized amplitude. The equations used for deriving relations are based on the Maxwell's Equations as well as the equations used in research work by other researchers.

2) Numerical analysis

The equations derived analytically are solved numerically using optimized values of parameters as used in previous research. The values are used to calculate normalized second harmonic amplitude under different conditions. The results obtained are plotted graphically to study the variation of normalized second harmonic amplitude with normalized propagation distance under different conditions.

Tools:

- 1) MATHEMATICA software
- 2) ORIGIN software.

2.2 Origin of ponderomotive force

When laser interacts with matter, the electromagnetic fields of laser exert force on particles of matter e.g. in metals particles are electrons, in semiconductors particles can be electrons or holes depending upon type of doping. The electric component accelerates particles and the velocity gained is calculated using equation of motion,

$$m \frac{d\vec{v}}{dt} = -e\vec{E} \quad (2.1)$$

where, $\vec{E}$ is the electric field and $\vec{v}$ is the velocity gained. The velocity $\vec{v}$ beats with magnetic field $\vec{B}$ and employ ponderomotive force $\vec{F}$ is given by

$$\vec{F} = -e(\vec{v} \times \vec{B}) \quad (2.2)$$

The same expression for the ponderomotive force has been used by many researchers [1]–[3]. Hence the ponderomotive force can be defined as a non linear force experienced by an electrically charged particle in an oscillating inhomogeneous electromagnetic field that accelerates the particles.

The current density produced due to nonlinear interactions can be calculated by using,

$$\vec{J} = ne\vec{v}' \quad (2.3)$$

where, $\vec{v}'$ is the velocity gained by the action of ponderomotive force.

2.3 Wave governing equation

The propagation of electromagnetic wave in media is governed by the wave equation based on Maxwell's equations. The introduction of dielectric constant helps to incorporate the impact of material on wave propagation. The wave equation for second harmonic wave propagating in vacuum is

$$\nabla^2 \vec{E}_2 = \frac{4\pi\partial\vec{J}}{v^2\partial t} + \frac{\partial^2 \vec{E}_2}{c^2\partial t^2} \quad (2.4)$$

where, $\vec{v}$ is the velocity with which nonlinear current propagates in media, $\vec{J}$ is the current density of linear and non-linear currents and $\vec{E}_2$ is the electric field of second harmonic generation. Here, medium is metal-semiconductor interface so to account the behavior of wave in medium the $\vec{v}$ is calculated using the concept of refractive index as $v = c/n$ where, n is the refractive index and c is the speed of light in vacuum. Also $n \propto \sqrt{\epsilon}$ and for metal-semiconductor interface,

$$n \propto \sqrt{\frac{\epsilon_m + \epsilon_s}{\epsilon_m \epsilon_s}} \quad (2.5)$$

where, ϵ_m and ϵ_s as the permittivity of metal and semiconductor respectively. So,

$$\frac{1}{v^2} = \frac{n^2}{c^2} = \frac{1}{c^2} \left(\frac{\epsilon_m + \epsilon_s}{\epsilon_m \epsilon_s} \right) \quad (2.6)$$

Hence, wave equation for propagation of second harmonic wave at metal-semiconductor interface becomes

$$\nabla^2 \vec{E}_2 = \frac{4\pi(\epsilon_m + \epsilon_s)\partial\vec{J}}{c^2(\epsilon_m \epsilon_s)\partial t} + \frac{\partial^2 \vec{E}_2}{c^2\partial t^2} \quad (2.7)$$

2.4 Role of different parameters

During our analysis we see the effect of following parameters on second harmonic generation.

2.4.1 Metal plasma:- Laser field interacts with metal and form ions creating

metal plasma. It exerts force on electrons of metal due to which electrons get drifted and undergo nonlinear interactions leading to harmonic generation. When the polarized light falls on the metal film at the interface of two media with different refractive index at certain angles, it sets the electrons to oscillations. The resonant oscillation of conduction band electrons in metal nano-particles leads to surface plasmon resonance (SPR) [4], [5] .

2.4.2 Semiconductor plasma:- Semiconductor has holes and electrons as charge carriers. In p-type semiconductor holes dominate and in n type electrons dominate. Interaction of electromagnetic fields with semiconductor involves drift of charge carriers that lead to the formation of semiconductor plasma. When a SPW with large amplitude interacts with semiconductor, it causes slight perturbation in amplitude of SPW which exerts a ponderomotive force on free electrons and holes leading to spatial modulation in free carrier density. The low value of plasma frequency of semiconductor plasma as compared to metal plasma allows easy interaction of SPW with electrons. In our work we have considered n type semiconductor Indium Antimonide, its low value of band gap energy and large carrier mobility favors excitation of localized surface plasmon resonance in it [6]–[8].

2.4.3 Surface plasma waves:- These waves arise in metals due to interaction of laser with free electrons. They generally exist at the interface of two media where permittivity changes its sign. These waves are evanescent in nature. Frequency of SPW is equivalent to frequency of incident laser but the amplitude of SPW is more than the amplitude of the incident laser. It is an electromagnetic wave with electric field component parallel to the direction of propagation and perpendicular to the surface while magnetic field component parallel to the surface and perpendicular to the direction of propagation [9]–[11].

2.4.4 Kretschmann configuration:- The configuration used for the work is Kretschmann attenuated total reflection configuration with thin layer of metal on glass prism and a thin layer of n-type semiconductor over the metal. The material of prism can be chosen so as to allow laser of suitable intensity to pass through it without any

damage. Laser light is incident on glass-metal interface from glass side at suitable angle so that maximum light gets reflected. Depending upon the nature of metal the part of light which gets absorbed will further interacts with the electrons of metal. The Kretschmann configuration is preferred for thin metal layers and for thick metal layers Otto configuration is preferred. The wave number of SPW should be matched to wave number of incident light for resonant process and the configuration like ATR helps to attain it.

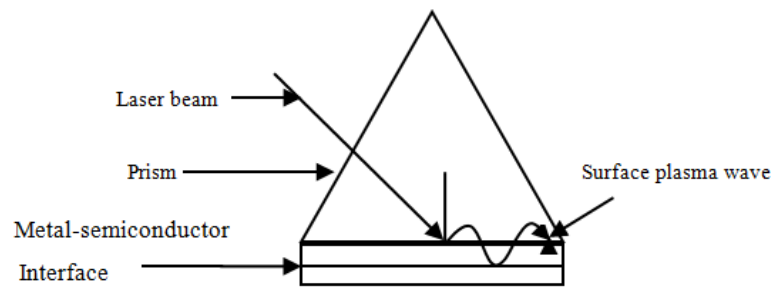


Fig.2.1. Kretschmann configuration with thin layer of a metal and a semiconductor

2.4.5 Wiggler magnetic field:- A wiggler consists of set of dipole magnets with different strengths. The resultant field produced depends on the bending of magnets in particular fashion. By adjusting the periods and gap between the magnets, required field with constant gradient can be produced. An oscillating field with suitable frequency when applied to plasma exerts force on the charge carriers and makes them oscillate with suitable phase hence, make the process of harmonic generation resonant. It provides an extra momentum to the oscillating electrons and helps to achieve phase matching. In the present work, wiggler field of low intensity 1T, 2T and 3T have been used. One such method to generate it is the edge focusing wiggler. It involves the use of permanent magnets Nd-Fe-B. By adjusting the number of periods and gap between the magnets required field with constant gradient can be produced. For our arrangement, the prism coated with metal-semiconductor can be placed in the channel between the poles to experience transverse magnetic field [12]-[13].

2.4.6 Laser profile:- An electromagnetic monochromatic beam, with amplitude envelope defined by a Gaussian function is known as Gaussian beam. There are various laser profiles such as Hermite-Cosh-Gaussian, Cosh-Gaussian, Hollow

Gaussian, Super Gaussian etc. which can be generated from Gaussian beam by varying its parameters and by using different lenses to refocus [14]–[16]. In this thesis, only Gaussian profile has been used to accomplish region to the low field region on account of the ponderomotive force. Due to which plasma electrons starts oscillating with suitable phase. The fields of the Gaussian laser beam, interacts with the electrons of plasma and oscillate them nonlinearly under the action of ponderomotive force. These nonlinear interactions accounts for the second harmonic generation.

2.4.7 Intensity of incident laser:- The intensity of incident laser plays an important role in the laser plasma interactions. Intensity of laser is defined as the energy transferred by the laser per unit area. More is the intensity of laser beam, more are the photons in it hence more interactions with the particles of plasma. As a result, greater will be the free electron density which produces efficient harmonic generation. The physical reason behind this is that with the enrichment of the intensity of laser, the strength of ponderomotive force and therefore, the initial ionization of the plasma gets enhanced. Consequently, with the increase of free electron density, the nonlinearity increases and as SH generation is a nonlinear phenomenon so it get enhanced more and more. For the present thesis, the Nd:YAG laser with wavelength 1064nm and intensity $I \approx 10^{12} \text{ W/cm}^2$ has been used [17]–[19] .

2.4.8 Self-focusing of laser:-The focusing of laser in plasmas is due to the existence of nonlinear factors like collisions, relativity and ponderomotive force. The self-focusing of laser helps to reduce the factors which broaden the beam area and generate focused harmonics. We have considered the self-focusing effect along with the ponderomotive nonlinearity. When an intense beam of laser propagates through the plasma, the electrons of plasma are pushed out from the strong power region due to the ponderomotive force. It leads to the change in the density of local electrons. As a result, self-focusing of laser takes place but the laser diverges due to the diffraction. Due to the combined action of ponderomotive nonlinearity and diffraction, periodic self-focusing and defocusing arises. The focused laser could produce focused surface plasma waves on metal surface which could further generate focused SHG. The

detailed study of the self-focusing due to ponderomotive nonlinearity is discussed in the thesis [20]–[22] .

In the thesis, study of SHG has been carried out using these parameters. The wave chosen is Gaussian profile, which is made to incident on glass-metal interface. Due to nonlinear interactions harmonic generation takes place. In Chapter 3 the effect of presence of density ripple is studied, the presence of ripples enhances nonlinear interactions and resonate the process. The effect of wiggler field has been studied in chapter 4. The SHG by two surface plasma waves inclined at different angles has been studied in chapter 5. Chapter 6 includes the study of effect of focusing trend of laser on harmonic generation.

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CHAPTER 3

SECOND HARMONIC GENERATION INDUCED BY SURFACE PLASMA WAVES OVER A METAL- SEMICONDUCTOR INTERFACE.

3.1 Introduction

Surface plasma waves (SPW) have many potential applications due to which they have drawn much attention in recent past years [1]–[3]. These are the electromagnetic waves which need two media with different dielectric properties and conductivities for propagation and they propagate at the interface. The interaction of laser fields with metals form metal plasma containing electrons and ions, mainly electrons interact with the electric and magnetic fields of laser and lead to SPW which further leads to harmonic generation. Many researchers have studied propagation and applications of SPW on metal-vacuum interface and worked on different materials[4]–[10]. Harmonic generation by a laser on the materials like silver(Ag) and n-type Indium Antimonide (n-InSb) in Kretschmann ATR configuration showed that the excitation area for n-InSb is much larger than the spot size of incident laser beam and SPW decays rapidly in conductors than in dielectrics, so the enhancement in electric field is more for n-InSb than for Ag. In the visible and near infrared region with high laser inputs, noble metals like Au, Ag and Cu are the most dominant choices in the plasmonics. The presence of density ripple also affects harmonic generation in metals as the laser couples through a surface ripple that accounts for the wave number mismatch between the laser and surface plasma waves and the required ripple number decreases with the increase in frequency of laser. The choice of geometry for metal interface depends on nature of film as for thin metal layers Kretschmann is used and for thick layers Otto is preferred. In Kretschmann attenuated total reflection, the laser is incident through the glass onto the glass-metal interface with the wave number whose component along the surface matches the wave number of SPW [11]–[13]. In metal-semiconductor heterostructures, the accumulation of hot electrons facilitates

the process of harmonic generation as the excited electrons can tunnel between plasmonic entities separated by small nano-meter sized gaps generating direct current electric fields across the nano-junctions. The electron tunneling occurs in gaps of quantum regime giving rise to fields of the order of 15ns[14]. In this chapter, a model for efficient second harmonic generation by SPW on metal-semiconductor interface in Kretschmann attenuated total reflection (ATR) geometry has been presented. Study has been done for different metals like Cu, Ag and Al with semiconductor n-InSb using ponderomotive mechanism between laser fields and electrons. A thin layer of semiconductor is considered over metal surface. Surface plasma waves of metal interact with electrons of n-type semiconductor leading to second harmonic(SH) generation and a significant amplitude enhancement has been observed for Cu-InSb interface.

In section 3.2 mathematical relations of normalized second harmonic amplitude with normalized propagation distance have been derived. The graphical variation and its outcomes have been discussed in section 3.3 and section 3.4, include conclusions.

3.2. Derivation of normalized second harmonic amplitude:

Consider a thin layer of metal over a glass prism with a thin layer of semiconductor over it in attenuated total reflection Kretschmann configuration as shown in Fig.3.1. The metal surface considered here is rippled with a suitable wave number. The electron density of the film is supposed to be varying periodically along z axis. The laser light is incident on glass-metal interface leading to SPW in metal. The most of the light gets reflected but some part gets absorbed depending upon reflectivity of metal. The electromagnetic fields of laser interact with electrons of metal and produces SPW of nearly same frequency as laser but with higher amplitude. The ripple present on metal surface resonantly drives SPW and helps in phase matching as well. A thin layer of n-InSb with thickness of few nano-meters is considered above the metal surface. The electromagnetic fields of SPW at the metal-semiconductor interface interact with electrons of n-InSb, leading to SHG. InSb provides large area for excitation of electrons than the laser spot size.

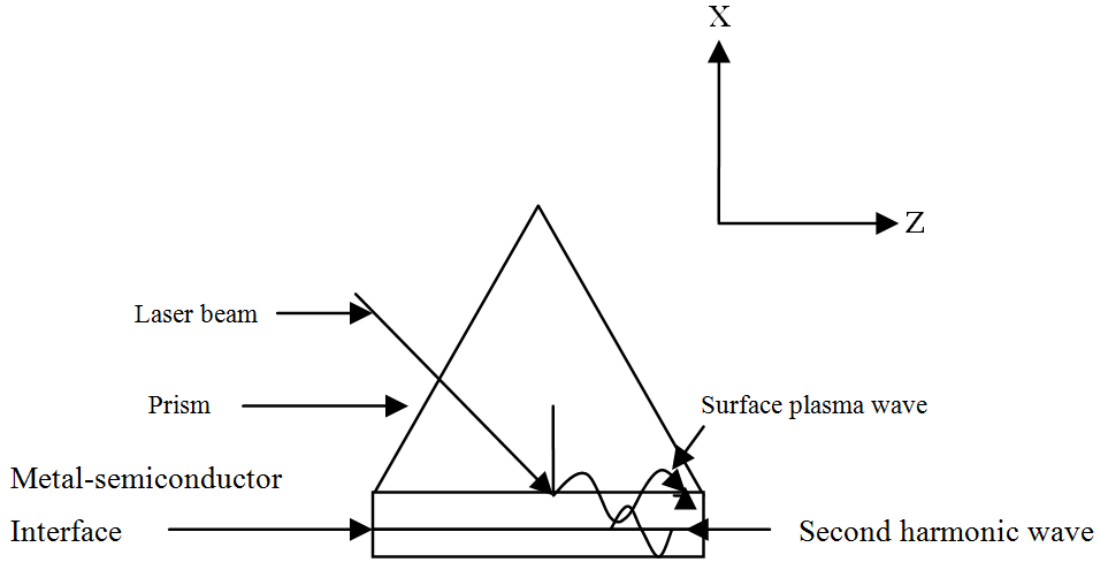


Fig.3.1. Schematic of Kretschmann configuration

Fields of laser are considered as,

$$\begin{aligned}\vec{E} &= \hat{x}A(z,t)e^{-i(\omega t - k_1 z)} \\ \vec{B} &= (\vec{k}_1 \times \vec{E}) / \omega_1\end{aligned}\quad (3.1)$$

where, A is the amplitude of electric field of laser, ω_1 is the frequency of laser and k_1 is the propagation constant. These fields interact with electrons of metal and produces surface plasma waves with electric field,

$$\vec{E}_s = \hat{x}A_s e^{-i(\omega t - k_s z)} \quad (3.2)$$

where, k_s is the propagation constant of SPW such that,

$$k_s = (\omega/c) \left(\epsilon_s \epsilon_m / (\epsilon_s + \epsilon_m) \right)^{1/2} \quad (3.3)$$

with ϵ_m and ϵ_s as the permittivity of metal and semiconductor respectively and ω is the frequency of surface plasma wave. Amplitude of surface plasma wave A_s depends on amplitude of incident laser A as $A_s = f(A)$. Following[15], A_s / A can be considered as a normalized amplitude of SPW with $A_s = pA$ where, p is a positive real number. Also, the frequency of SPW is nearly same as the frequency of incident laser so, $\omega_1 = \omega$. Now upon resonance, the oscillations of conduction electrons quickly decay to generate energetic electrons that play essential roles in prompting photons.

Acceleration of electrons is calculated using the equation of motion,

$$m \frac{d\vec{v}_1}{dt} = -e\vec{E}_s - e\vec{E} \quad (3.4)$$

$$\vec{v}_1 = \left[\int \frac{-epAe^{-i(\omega t - k_s z)} dt}{m} - \int \frac{-eAe^{-i(\omega_1 t - k_1 z)} dt}{m} \right] \hat{x} \quad (3.5)$$

As $t \rightarrow \infty, A_s, A \rightarrow 0$ so A_s and A have been considered as independent of t while integration,

$$\vec{v}_1 = \left[\frac{-epAe^{-i(\omega t - k_s z)}}{m(-i\omega)} - \frac{-eAe^{-i(\omega_1 t - k_1 z)}}{m(-i\omega_1)} \right] \hat{x} \quad (3.6)$$

The velocity $\vec{v}_1$ beats with magnetic field of laser and generates ponderomotive force $\vec{F}$, where,

$$\vec{F} = -e(\vec{v}_1 \times \vec{B})$$

due to this force velocity of electrons changes and it is taken as $\vec{v}_1'$ such that

$$\begin{aligned} \vec{v}_1' &= -\frac{e^2 k_1}{m^2 \omega_1} \left[\frac{pA^2 e^{-i[(\omega + \omega_1)t - (k_s + k_1)z]}}{2\omega_1^2} + \frac{A^2 e^{-i(2\omega_1 t - 2k_1 z)}}{2\omega_1^2} \right] \hat{z} \\ \vec{v}_1' &= -\frac{e^2 k_1}{m^2 \omega_1} \left[\frac{pA^2 e^{-i[2\omega_1 t - (k_s + k_1)z]}}{2\omega_1^2} + \frac{A^2 e^{-i(2\omega_1 t - 2k_1 z)}}{2\omega_1^2} \right] \hat{z} \end{aligned} \quad (3.7)$$

To make the process resonant metallic surface is rippled with density profile, $n_q = n_0 e^{iqz}$ where, n_0 is the electron density, q is the ripple wave number and z is the propagation distance. The nonlinear effects arise due to $\vec{v}_1'$ and the coupling of this nonlinear velocity with density ripple produces second harmonic current in the ripple region at $(2\omega, k_2 = 2k_1 + q)$ given by

$$\vec{J} = n_0 e^{iqz} e \vec{v}_1' \quad (3.8)$$

Here, q is chosen such that

$$k_2 = 2k_1 + q \quad (3.9)$$

which obey phase matching condition, which is an essential requirement for resonant

wave propagation. The field equation for second harmonic is taken as

$$\vec{E}_2 = A_2 e^{-i(2\omega_1 t - k_2 z)} \hat{x} \quad (3.10)$$

with A_2 as the amplitude and k_2 as the propagation constant of second harmonic.

The current density $\vec{J}$ due to nonlinear interactions

$$\vec{J} = -\frac{n_0 e^3 k_1}{m^2 \omega_1} \left[\frac{p A^2 e^{-i[2\omega_1 t - (k_s + k_1 + q)z]}}{2\omega_1^2} + \frac{A^2 e^{-i(2\omega_1 t - (2k_1 + q)z)}}{2\omega_1^2} \right] \hat{z} \quad (3.11)$$

The wave equation used for the calculation of amplitude of second harmonic generated is,

$$\nabla^2 \vec{E}_2 = \frac{4\pi(\epsilon_m + \epsilon_s) \partial \vec{J}}{c^2 (\epsilon_m \epsilon_s) \partial t} + \frac{\partial^2 \vec{E}_2}{c^2 \partial t^2} \quad (3.12)$$

On substituting the values from Eq.3.10 and 3.11 in Eq.3.12 we get,

$$\frac{\partial^2 (A_2 e^{-i(2\omega_1 t - k_2 z)})}{\partial z^2} = \frac{1}{c^2} \frac{\partial^2 (A_2 e^{-i(2\omega_1 t - k_2 z)})}{\partial t^2} + \frac{4\pi}{c^2} \left(\frac{\epsilon_m + \epsilon_s}{\epsilon_m \epsilon_s} \right) \frac{\partial}{\partial t} \left(-\frac{n_0 e^3 k_1}{m^2 \omega_1} \left[\frac{p A^2 e^{-i(2\omega_1 t - (k_s + k_1 + q)z)}}{2\omega_1^2} + \frac{A^2 e^{-i(2\omega_1 t - (2k_1 + q)z)}}{2\omega_1^2} \right] \right) \quad (3.13)$$

$$\frac{\partial^2 (A_2 e^{-i(2\omega_1 t - k_2 z)})}{\partial z^2} = \frac{1}{c^2} \frac{\partial^2 (A_2 e^{-i(2\omega_1 t - k_2 z)})}{\partial t^2} + \frac{4\pi}{c^2} \left(\frac{\epsilon_m + \epsilon_s}{\epsilon_m \epsilon_s} \right) \frac{\partial}{\partial t} \left(-\frac{n_0 e^3 k_1}{m^2 \omega_1} \left[\frac{p A^2 e^{-i(2\omega_1 t - (k_s + k_1 + q)z)}}{2\omega_1^2} + \frac{A^2 e^{-i(2\omega_1 t - (2k_1 + q)z)}}{2\omega_1^2} \right] \right) \quad (3.14)$$

$$\frac{\partial}{\partial z} \left(A_2 i k_2 e^{-i(2\omega_1 t - k_2 z)} + e^{-i(2\omega_1 t - k_2 z)} \frac{\partial A_2}{\partial z} \right) = \frac{1}{c^2} \frac{\partial}{\partial t} \left(-2 A_2 i \omega e^{-i(2\omega_1 t - k_2 z)} + e^{-i(2\omega_1 t - k_2 z)} \frac{\partial A_2}{\partial t} \right) + \frac{4\pi}{c^2} \left(\frac{\epsilon_m + \epsilon_s}{\epsilon_m \epsilon_s} \right) \left(-\frac{n_0 e^3 k_1}{m^2 \omega_1} \left[\frac{-2i \omega_1 p A^2 e^{-i(2\omega_1 t - (k_s + k_1 + q)z)}}{2\omega_1^2} + \frac{-2i \omega_1 A^2 e^{-i(2\omega_1 t - (2k_1 + q)z)}}{2\omega_1^2} \right] \right) \quad (3.15)$$

Using Eq.3.9 in Eq.3.15 and Here, $k_s = pk$ where p represents an integer. While solving exponential terms it will not lead to much difference in numerical values if we consider $e^{-i(\omega t - k_s z)} = e^{-i(\omega_1 t - k_1 z)}$ and $k_s + k_1 + q \approx 2k_1 + q$ (for simplification). On equating real and imaginary parts we get,

$$\frac{\partial^2 A_2}{\partial z^2} - k_2^2 A_2 = \frac{1}{c^2} \frac{\partial^2 A_2}{\partial t^2} - \frac{\omega^2 A_2}{c^2} + \frac{4\pi}{c^2} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \left(\frac{2n_0 e^2 A_2}{m\omega} + \frac{n_0 e^3 k_1 p^2 A^2}{2\omega^3 m^2} \frac{\partial A_1}{\partial t} \right) + p A_1 \frac{\partial A_1}{\partial t} + \frac{n_0 e^3 k_1 A}{\omega^3 m^2} \frac{\partial A}{\partial t} \quad (3.16)$$

$$2k_2 \frac{\partial A_2}{\partial z} + \frac{2\omega_2}{c^2} \frac{\partial A_2}{\partial t} = \frac{4\pi}{c^2} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \left(\frac{n_0 e^2}{2m\omega} \frac{\partial A_2}{\partial t} + \frac{n_0 e^3 k_1 (1+p) A^2}{\omega_1^2 m^2} \right) \quad (3.17)$$

By using,

$$\alpha = \frac{4\omega}{2c^2 k_2} + \frac{4\pi n_0 e^2}{4k_2 c^2 m\omega} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \quad \text{and} \quad \beta = \frac{4\pi}{c^2} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \frac{n_0 e^3 k_1 (1+p)}{2k_2 \omega_1^2 m^2}$$

Eq. (3.17) becomes

$$\frac{\partial A_2}{\partial z} + \alpha \frac{\partial A_2}{\partial t} = \beta A_1^2 \quad (3.18)$$

Considering Eq.3.18 and by taking the laser profile to be Gaussian as

$$A^2 = [f(z - v_g t)]^2 \quad \text{and} \quad f(z - v_g t) = A_0 e^{-\left(\frac{z - v_g t}{\tau v_g}\right)^2} \quad \text{with} \quad v_g = c \sqrt{(\varepsilon_m + \varepsilon_s) / \varepsilon_m \varepsilon_s} \quad \text{and} \quad \tau \text{ is laser pulse width.}$$

Using a new set of variables $z - v_g t = \zeta$, $z = \eta$, $\zeta_0 = \tau v_g$, $\beta' = (1 - (v_g / c)^2)$, $z' = z / \zeta_0$ and $t' = v_g t / \zeta_0$ we get,

$$\alpha \approx \frac{v_g}{c^2}, \quad \frac{\partial}{\partial z} = \frac{\partial}{\partial \zeta} + \frac{\partial}{\partial \eta} \quad \text{and} \quad \frac{\partial}{\partial t} = -v_g \frac{\partial}{\partial \zeta} \quad (3.19)$$

complimentary solution for Eq.3.18 using the above variables is

$$A_2 = \frac{-\beta A_0^2}{\beta'} f(\zeta - \beta' \eta) \quad (3.20)$$

and the particular solution is,

$$A_2 = \frac{-\beta A_0^2}{\beta'} \int \exp\left(\frac{-\zeta^2}{\zeta_0^2}\right) d\zeta, \quad (3.21)$$

So complete solution gives normalized second harmonic amplitude as,

$$\left| \frac{A_2}{A_0} \right| = \frac{4\pi}{c^2} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \frac{n_0 e^3 k_1 (1+p) A_0 \zeta_0 \sqrt{\pi}}{4k_2 \omega_1^2 m^2 \beta'} [Erf(z' - t') - Erf\{(1 - \beta')z' - t'\}] \quad (3.22)$$

3.3 Results and graphical analysis

To draw the inferences from Eq.3.22, it has been solved numerically for Nd:YAG laser with wavelength 1064nm. The absolute values of effective permittivity are taken as 57.909 for Ag, 49.58 for Cu, 106.82 for Al and 19.5 for InSb at 1064nm [16]–[18]. The thickness of metal and semiconductor has been considered of few nano-meters and doping level of n-InSb is adjusted to have $\omega_p / \omega < 1$ for semiconductor. The graphical relationship for normalized second harmonic amplitude with normalized propagation distance has been studied for three metal-semiconductor interfaces at different values of time. The other parameters of Eq.3.22 are taken as $(4\pi m_0 e^2 m^{-1})^{1/2} / \omega_1 = 0.9$, $p = 0.5$, $eA_0 / mc\omega_1 = 0.08$, $k_1 \xi_0 \sqrt{\pi} / 2\beta' = 30$ and $\beta' = 0.3$.

Fig.3.2 shows the variation of normalized second harmonic amplitude A_2/A_0 and normalized fundamental amplitude A/A_0 with normalized propagation distance z' for metals Cu, Ag and Al with thin layer of n-InSb at time $t' = 0, 2$ and 4. In Fig.3.2(a) a sharp increase in the normalized second harmonic amplitude is seen at $z' = 2$ for $t' = 0$. Increase in amplitude is more for Cu than for Ag and Al, as observed graphically under given conditions. It is due to the fact that the electromagnetic fields of laser exert ponderomotive force on electrons of metals resulting in generation of SPW. The conduction electron density for Cu is more than Ag and Al so the fields of SPW of Cu can interact more efficiently with electrons of n-type semiconductor, leading to harmonic generation with enhanced amplitude. Thakur and Kant[19] have studied variation of normalized second harmonic amplitude with normalized propagation distance in plasma and they noticed that for $\omega_p / \omega_1 > 0.5$, a sharp increase in amplitude occurs at $z' > 8$ but the present work shows that for $\omega_p / \omega_1 > 0.5$, a sharp increase in amplitude occurs at $z' < 4$. Hence, the choice of metal-semiconductor interface reduces the propagation distance for second harmonic to achieve maximum amplitude as compared to plasma. Similarly it is clear from Fig.3.2(b) and Fig.3.2(c) that amplitude of second harmonic increases with time and enhancement is more for Cu-InSb interface than for Ag-InSb and Al-InSb interfaces.

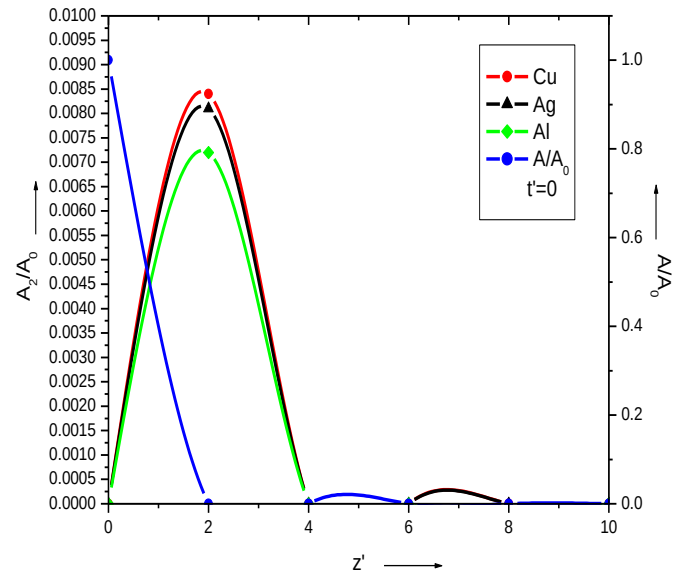


Fig.3.2(a)

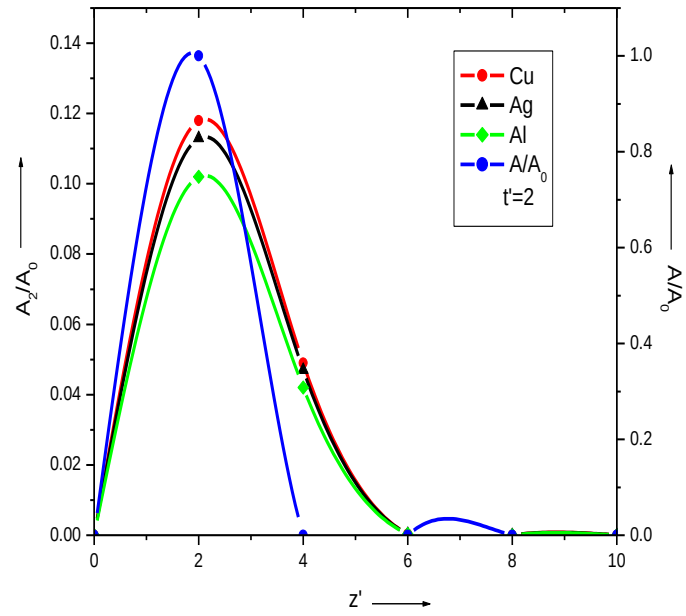


Fig.3.2(b)

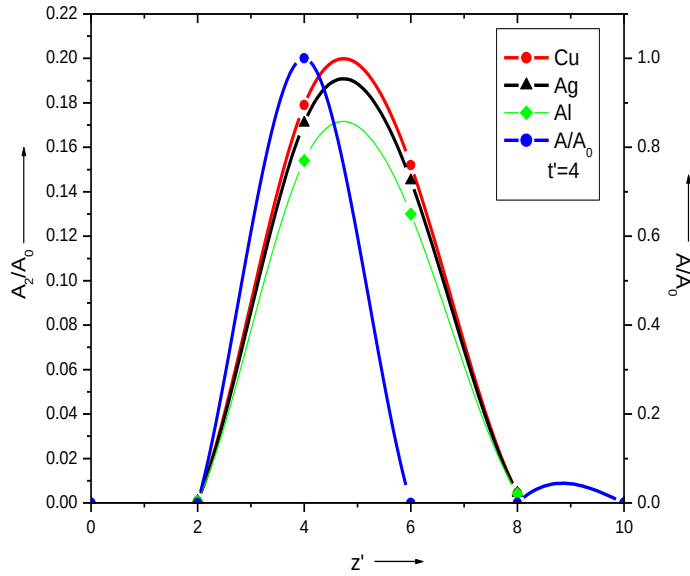


Fig.3.2(c)

Fig.3.2. Plot showing the variation of normalized second harmonic amplitude A_2/A_0 and normalized fundamental amplitude A/A_0 with normalized propagation distance z' for metals Cu, Ag and Al with layer of n-InSb.

Fig.3.3 reveals the variance of the normalized amplitude of second harmonic with propagation distance at different instants of time for Cu, Ag and Al. Values of different parameters have been taken same as taken in Fig.3.2. It is clear from Fig.3.3 that second harmonic amplitude increases with time in all the three cases but for Cu, enhancement in amplitude of SHG is found more as compared to Ag and Al. One can notice from Fig.3.3 that second harmonic amplitude increases as time passes and attain its maximum value at $z'=5$ for Cu. Takagi *et al.*[16] studied surface plasma waves in Kretschmann geometry and found that plasmon dispersion is in agreement with bulk dielectric function for Cu than Ag. Similarly in our work, we have seen better enhancement in the amplitude of second harmonic for Cu.

Fig.3.4 shows the variation of normalized second harmonic amplitude with normalized propagation distance for different values of normalized plasma frequency for Cu, Ag and Al with a thin layer of n-InSb at time $t'=2$. It is clear from the graphs that with the increase in plasma frequency, the amplitude of second harmonic increases in all the three cases but its value is maximum for Cu. Thakur and Kant[20] have studied variation of normalized second harmonic amplitude with normalized

propagation distance in plasma for different values of plasma frequency and have found increase in amplitude of second harmonic with plasma frequency. Our work shows that Cu with a thin layer of n-InSb would be a better choice for the applications based on second harmonic generation.

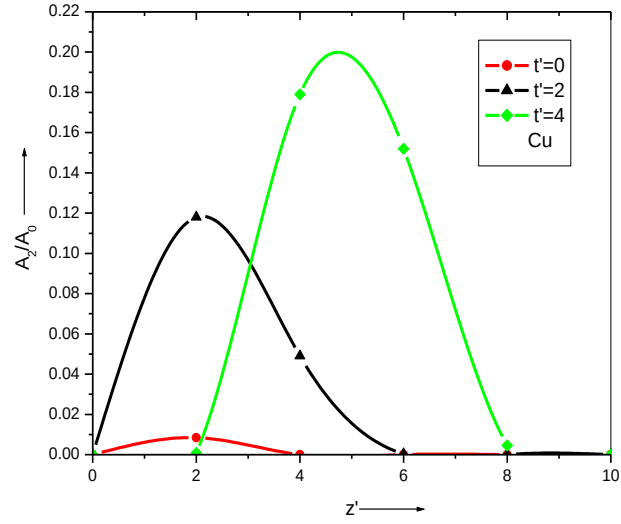


Fig.3.3(a)

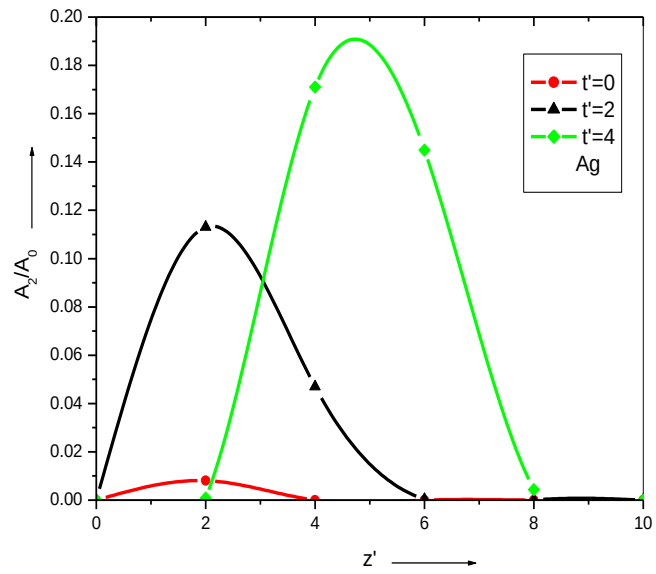


Fig.3.3(b)

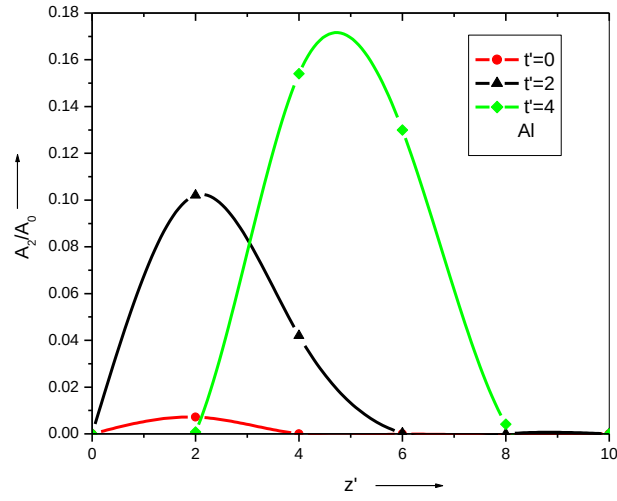


Fig.3.3(c)

Fig.3.3. Plot showing the variation of normalized second harmonic amplitude A_2/A_0 with normalized propagation distance z' for different metals like Cu in Fig.3.3(a), Ag in Fig.3.3(b) and Al in Fig.3.3(c), each with layer of n-InSb at different values of time $t' = 0, 2$ and 4

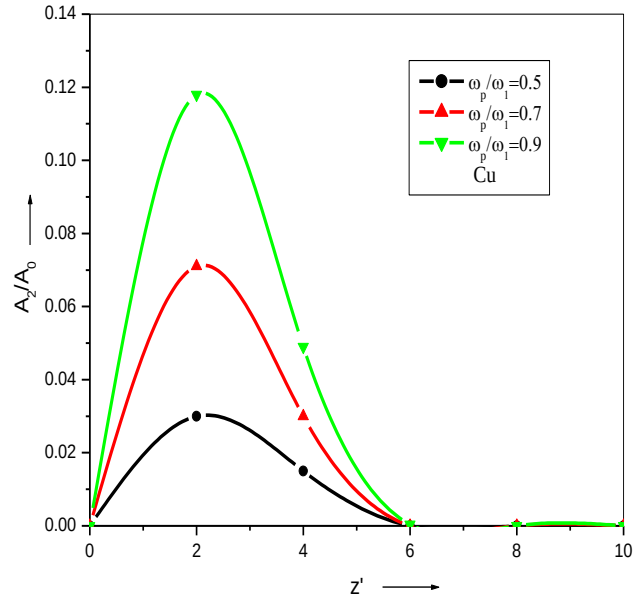


Fig.3.4(a)

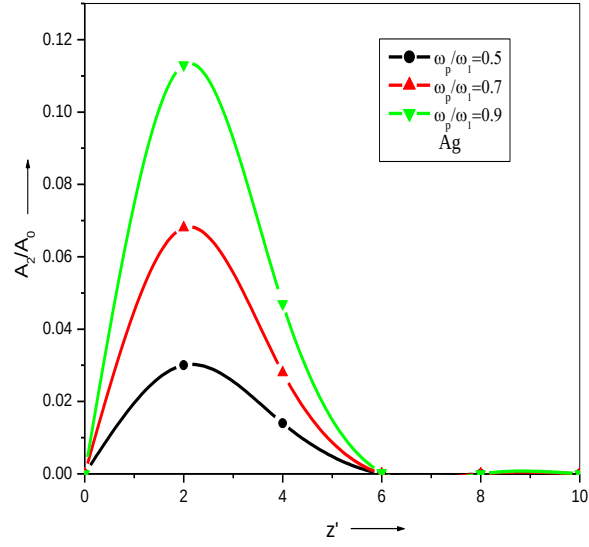


Fig.3.4(b)

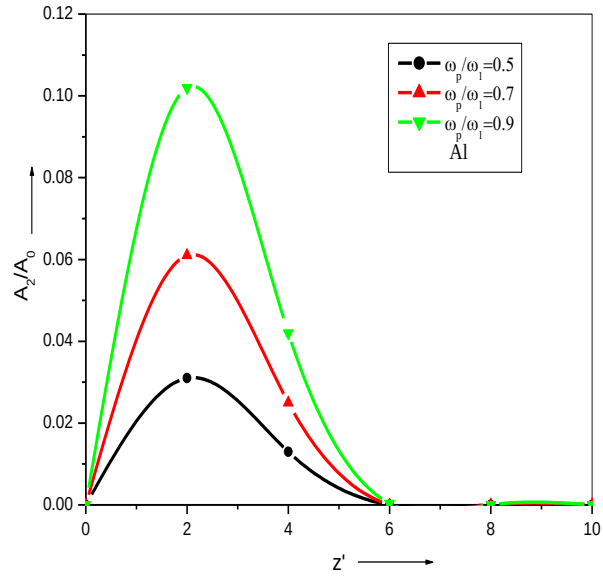


Fig.3.4(c)

Fig.3.4. Plot showing the variation of normalized second harmonic amplitude A_2/A_0 with normalized propagation distance z' for metals like Cu in Fig.3.4(a), Ag in Fig.3.4(b) and Al in Fig.3.4(c) each with layer of n-InSb for different values of normalized plasma frequency at time $t' = 2$

3.4 Conclusion

Surface plasma waves are the electromagnetic waves formed by the interaction of laser with metal surface. The evanescent fields of SPW in metals can influence

electrons of n-type semiconductor layered over metal surface via nonlinear interactions, leading to second harmonic. Amplitude enhancement for harmonic depends upon dielectric properties of material. It is observed that for metal-semiconductor interface in Kretschmann configuration the enhancement in amplitude of second harmonic is more for Cu-InSb interface than for Ag-InSb and Al-InSb interface. The second harmonic produced propagates at metal-semiconductor interface accompanied by increase in amplitude which further increases with increase in plasma frequency. It opens cost effective pathways for future applications of SPW.

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CHAPTER 4

EFFECT OF EXTERNAL MAGNETIC FIELD ON THE EFFICIENCY OF SECOND HARMONIC GENERATION BY SURFACE PLASMA WAVE

4.1 Introduction

The nonlinear interaction of laser with the matter has given rise to many applications like high harmonic generation, laser driven plasma based accelerators, sensors, photoelectron spectroscopy, etc. [1]–[10]. The interaction of laser with metal involves the action of ponderomotive force by electric and magnetic components of laser which oscillates the electrons of metal and generates surface plasma waves. SPW is an electromagnetic wave with similar frequency as the incident laser but with higher intensity. The SPW are evanescent in nature so they travel along the interface and die with depth. These waves propagate at the interface of two media with different values for relative permittivity. The photons of SPW are capable of accelerating electrons of other material layered over metal. The configuration chosen in our work is Kretschmann attenuated total reflection configuration with metal-semiconductor interface. The thin layer of InSb is considered over metal. The electromagnetic fields of SPW further undergo nonlinear interactions with electrons of InSb and leads to efficient generation of harmonic with double frequency as that of the laser. In the presence of SPW and wiggler field the electrons could travel large distances in the direction of propagation. The presence of external agents like density ripple, magnetic fields etc. help to resonate the process and increases the efficiency of harmonic generation. The wiggler field of suitable wiggler period helps to attain phase matching by providing additional momentum to the oscillating electrons. Harmonic generation on different materials and in the presence of external agents like the wiggler field has been studied by many researchers [11]–[13]. Their work showed that the presence of wiggler field improves the efficiency of harmonic generation by enhancing the amplitude due to resonant interactions among laser photons and charge particles of matter.

In the present chapter, second harmonic generation induced by surface plasma wave at the metal-semiconductor interface in the presence of a wiggler magnetic field has been

studied. The phase matching condition is fulfilled by the application of the wiggler magnetic field. Here, in section 4.2, we have obtained the expression for normalized amplitude for second harmonic generation. Results are discussed and presented in graphical form in section 4.3 and the conclusion is presented in section 4.4.

4.2 Effect of wiggler field using ponderomotive mechanism

A thin metal layer over a glass prism with a thin layer of semiconductor has been considered in Kretschmann ATR configuration as shown in Fig.4.1, where laser light is incident at an acute angle on metal glass interface leading to SPW in metal[14]. The em fields of laser interact with electrons of the metal and produce SPW of nearly the same frequency as laser but with higher amplitude. The component of the wave vector of the laser along the surface matches with the wave number of surface plasma wave, leading to efficient coupling [15]. A thin layer of n-InSb with the thickness of a few nano-meters is considered above the metal surface. The electromagnetic fields of SPW produced at the metal-semiconductor interface interact with electrons of n-InSb. The semiconductor can accumulate energetic electrons which can easily tunnel through plasmonic entities[16].

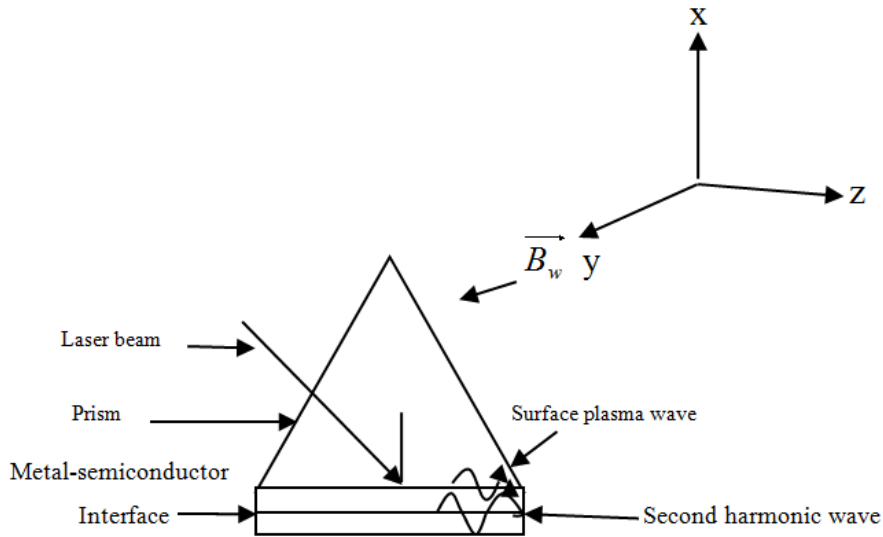


Fig.4.1. Kretschmann configuration with wiggler magnetic field

Electric and magnetic fields of laser are considered as

$$\begin{aligned}\vec{E} &= \hat{x}A(z,t)e^{-i(\omega_1 t - k_1 z)} \\ \vec{B} &= (\vec{k}_1 \times \vec{E}) / \omega_1\end{aligned}\tag{4.1}$$

where, A is the amplitude of electric field of laser, ω_1 is the frequency of laser and k_1 is the propagation constant. These fields interact with electrons of metal and produces surface plasma waves

$$\vec{E}_s = \hat{x}A_s e^{-i(\omega t - k_s z)} \quad (4.2)$$

where, k_s is the propagation constant such that,

$$k_s = (\omega/c) (\epsilon_s \epsilon_m / (\epsilon_s + \epsilon_m))^{1/2} \quad (4.3)$$

with ϵ_m and ϵ_s as the permittivity of metal and semiconductor respectively and ω is the frequency of surface plasma wave. The frequency of SPW is nearly same as the frequency of the laser, so $\omega = \omega_1$ [16]. Amplitude of surface plasma wave A_s depends on amplitude of incident laser A with $A_s = f(A)$. Following [17], A_s/A can be considered as a normalized quantity with $A_s = nA$ where, n is a positive real number. Fields of surface plasma waves interacts with electrons of semiconductor and accelerates them. Acceleration so produced is calculated using equation of motion,

$$m \frac{d\vec{v}_1}{dt} = -e\vec{E}_s - e\vec{E} \quad (4.4)$$

The velocity gained by electrons is,

$$\begin{aligned} \vec{v}_1 &= \int \left(-\frac{e}{m} \vec{E}_s - \frac{e}{m} \vec{E} \right) dt \\ v_1 &= \int \left(-\frac{e}{m} A_s e^{-i(\omega t - k_s z)} - \frac{e}{m} A e^{-i(\omega_1 t - k_1 z)} \right) dt \\ v_1 &= \int \left(-\frac{e}{m} n A e^{-i(\omega t - k_s z)} - \frac{e}{m} A e^{-i(\omega_1 t - k_1 z)} \right) dt \\ v_1 &= -\frac{e}{m} (n+1) A \int e^{-i(\omega t - k_s z)} dt \end{aligned} \quad (4.5)$$

Here, $k_s = pk$ where p represents an integer while solving exponential terms it will not lead to much difference in numerical values so for simplicity we have considered

$$e^{-i(\omega t - k_s z)} = e^{-i(\omega_1 t - k_1 z)} \quad (4.6)$$

So, using Eq.(4.5) in Eq.(4.6) we get

$$\vec{v}_1 = \left[\frac{-eAn'e^{-i(\omega t - k_s z)}}{-im\omega} \right] \hat{x} \quad (4.7)$$

Here, $n'=1+n$ which is a positive real number. Wiggler magnetic field is applied in transverse direction given by,

$$\vec{B}_w = B_0 e^{ik_0 z} \hat{y} \quad (4.8)$$

where, B_0 is the amplitude of wiggler field and k_0 is the wiggler wave number[18]. In the present work, small values of magnetic field are considered and the edge focusing method to produce magnetic field has been used. The presence of wiggler field helps to achieve the phase matching condition by providing additional momentum to the photons of second harmonic. The velocity $\vec{v}_1$ beats with wiggler magnetic field of laser and generates ponderomotive force $\vec{F}$, where,

$$\vec{F} = -e(\vec{v}_1 \times \vec{B}_w) \quad (4.9)$$

due to this force velocity of electrons changes and it is taken as $\vec{v}_1'$ such that

$$m \frac{d\vec{v}_1'}{dt} = -e \left[\frac{-eAn'e^{-i(\omega t - k_s z)}}{-im\omega} \hat{x} \times B_0 e^{ik_0 z} \hat{y} \right]$$

$$\frac{d\vec{v}_1'}{dt} = e^2 B_0 \left[\frac{An'e^{-i(\omega t - k_s z + k_0 z)}}{-im^2 \omega} \right] \hat{z}$$

$$\frac{d\vec{v}_1'}{dt} = e^2 B_0 \left[\frac{An'e^{-i(\omega t - k_s z + k_0 z)}}{-im^2 \omega} \right] \hat{z}$$

$$\vec{v}_1' = \int e^2 B_0 \left[\frac{An'e^{-i(\omega t - k_s z + k_0 z)}}{-im^2 \omega} \right] \hat{z} dt$$

$$\vec{v}_1' = \left[\frac{-e^2 n' AB_0 e^{-i(\omega t - (k_1 + k_0)z)}}{m^2 \omega^2} \right] \hat{z} \quad (4.10)$$

It will induce density perturbation at $(\omega_1, k_1 + k_0)$ in agreement with the equation of continuity,

$$n_1 = (\vec{k}_1 + \vec{k}_0) \cdot \vec{v}_1' / \omega_1 \quad (4.11)$$

To exert ponderomotive force on electrons at $(2\omega, 2k_1 + k_0)$ $\vec{v}_1'$ beats with $\vec{B}$ and imparts velocity, $\vec{v}_2^{NL}$ to electrons given as,

$$\vec{F}^{NL} = -e(\vec{v}_1 \times \vec{B}) \quad (4.12)$$

$$\begin{aligned} m \frac{d\vec{v}_2^{NL}}{dt} &= -e \left[\frac{-e^2 n' AB_0 e^{-i(\omega t - (k_1 + k_0)z)}}{m^2 \omega^2} \hat{z} \times (\vec{k}_1 \times \vec{E}) / \omega \right] \\ \frac{d\vec{v}_2^{NL}}{dt} &= e^3 n' AB_0 \left[\frac{e^{-i(\omega t - (k_1 + k_0)z)}}{m^3 \omega^2} \hat{z} \times (\vec{k}_1 \times \hat{x} A e^{-i(\omega_1 t - k_1 z)}) / \omega \right] \\ \frac{d\vec{v}_2^{NL}}{dt} &= e^3 n' AB_0 \left[\frac{e^{-i(\omega t - (k_1 + k_0)z)}}{m^3 \omega^2} \hat{z} \times (k_1 A e^{-i(\omega_1 t - k_1 z)}) \hat{y} / \omega \right] \\ \frac{d\vec{v}_2^{NL}}{dt} &= -e^3 n' A^2 B_0 \left[\frac{k_1 e^{-i(2\omega t - (2k_1 + k_0)z)}}{m^3 \omega^3} \hat{x} \right] \\ \vec{v}_2^{NL} &= \frac{-e^3 n' A^2 k_1 B_0}{m^3 \omega^3} \int e^{-i(2\omega t - (2k_1 + k_0)z)} \hat{x} dt \\ \vec{v}_2^{NL} &= \frac{-ie^3 n' A^2 k_1 B_0 e^{-i(2\omega t - (2k_1 + k_0)z)} \hat{x}}{2m^3 \omega^4} \end{aligned} \quad (4.13)$$

As the wave vector increases more than linearly with incident frequency, the momentum is not conserved, i.e. the momentum of second harmonic is more than two times the momentum of incident photon. This give rise to second harmonic nonlinear current density at metal-semiconductor interface,

$$\vec{J}_2^{NL} = -n_0 e \vec{v}_2^{NL} - n_1 e \vec{v}_1 \quad (4.14)$$

using Eq.4.7 and Eq.4.13 in Eq.4.14 we get,

$$\vec{J}_2^{NL} = -n_0 e \left[\frac{-ie^3 n' A^2 k_1 B_0 e^{-i(2\omega t - (2k_1 + k_0)z)}}{2m^3 \omega^4} \right] \hat{x} - n_1 e \left[\frac{-e A n' e^{-i(\omega t - k_1 z)}}{m(-i\omega)} \right] \hat{x} \quad (4.15)$$

The linear current density due to self consistent second harmonic field,

$$\vec{E}_2 = A_2 e^{-i(2\omega t - k_2 z)} \vec{x} \quad (4.16)$$

at $(2\omega, k_2)$ is

$$\vec{J}_2^L = n_0 e \vec{v}_2 \quad (4.17)$$

where, $\vec{v}_2$ is obtained using equation of motion,

$$m(d\vec{v}_2 / dt) = -e\vec{E}_2 \quad (4.18)$$

$$m(d\vec{v}_2 / dt) = -e A_2 e^{-i(2\omega t - k_2 z)} \hat{x}$$

$$\begin{aligned}
\frac{d\vec{v}_2}{dt} &= -\frac{e}{m} A_2 e^{-i(2\omega t - k_2 z)} \hat{x} \\
\vec{v}_2 &= -\frac{e}{m} \int A_2 e^{-i(2\omega t - k_2 z)} \hat{x} dt \\
\vec{v}_2 &= -\frac{ie}{2m\omega} A_2 e^{-i(2\omega t - k_2 z)} \hat{x}
\end{aligned} \tag{4.19}$$

So Eq. (4.17) becomes

$$\vec{J}_2^L = -i \frac{n_0 e^2}{2m\omega} A_2 e^{-i(2\omega t - k_2 z)} \hat{x} \tag{4.20}$$

Using Eq.4.15 and Eq.4.20 we get the total current density

$$\begin{aligned}
\vec{J} &= \vec{J}_2^L + \vec{J}_2^{NL} \\
\vec{J} &= -i \frac{n_0 e^2}{2m\omega} A_2 e^{-i(2\omega t - k_2 z)} \hat{x} - n_0 e \left[\frac{-ie^3 n' A^2 k_1 B_0 e^{-i(2\omega t - (2k_1 + k_0)z)}}{2m^3 \omega^4} \right] \hat{x} - n_1 e \left[\frac{-e A n' e^{-i(\omega t - k_s z)}}{m(-i\omega)} \right] \hat{x}
\end{aligned} \tag{4.21}$$

The amplitude A_2 for second harmonic is calculated by using equation,

$$\nabla^2 \vec{E}_2 = \frac{4\pi(\epsilon_m + \epsilon_s) \partial \vec{J}}{c^2(\epsilon_m \epsilon_s) \partial t} + \frac{\partial^2 \vec{E}_2}{c^2 \partial t^2} \tag{4.22}$$

Using Eq.4.16 and Eq.4.21 we get,

$$\begin{aligned}
\frac{\partial^2}{\partial z^2} (A_2 e^{-i(2\omega t - k_2 z)} \vec{x}) &= \frac{4\pi(\epsilon_m + \epsilon_s)}{c^2(\epsilon_m \epsilon_s)} \frac{\partial}{\partial t} \left(-i \frac{n_0 e^2}{2m\omega} A_2 e^{-i(2\omega t - k_2 z)} \hat{x} - n_0 e \left[\frac{-ie^3 n' A^2 k_1 B_0 e^{-i(2\omega t - (2k_1 + k_0)z)}}{2m^3 \omega^4} \right] \hat{x} \right) \\
\frac{4\pi(\epsilon_m + \epsilon_s)}{c^2(\epsilon_m \epsilon_s)} \frac{\partial}{\partial t} \left(-n_1 e \left[\frac{-e A n' e^{-i(\omega t - k_s z)}}{m(-i\omega)} \right] \hat{x} \right) &+ \frac{\partial^2}{\partial t^2} \left(\frac{A_2 e^{-i(2\omega t - k_2 z)} \hat{x}}{c^2} \right)
\end{aligned} \tag{4.23}$$

Differentiating the terms and equating real and imaginary parts we get,

By real parts

$$\frac{\partial^2 A_2}{\partial t^2} - k_2^2 A_2 = \frac{1}{c^2} \frac{\partial^2 A_2}{\partial t^2} - 4\omega^2 A_2 + \frac{4\pi(\epsilon_m + \epsilon_s)}{c^2(\epsilon_m \epsilon_s)} \left(\frac{n_0 e^2 A_2}{m} - \frac{n_0 e^4 n' B_0 k_1 A^2}{2m^3 \omega^3} - \frac{n_0 (k_1 + k_0) e^4 n' B_0 A^2}{m^3 \omega^3} \right) \tag{4.24}$$

By imaginary parts

$$2k_2 \frac{\partial A_2}{\partial z} + \left(\frac{4\omega}{c^2} + \frac{4\pi(\varepsilon_m + \varepsilon_s)n_0 e^2}{c^2(\varepsilon_m \varepsilon_s)2m\omega} \right) \frac{\partial A_2}{\partial t} = \frac{4\pi(\varepsilon_m + \varepsilon_s)}{c^2(\varepsilon_m \varepsilon_s)} \left(\frac{n_0 e^4 n' B_0 k_1 A}{2m^3 \omega^4} - \frac{n_0(k_1 + k_0)e^4 n' B_0 A}{m^3 \omega^4} \right) \frac{\partial A}{\partial t} \quad (4.25)$$

Considering Eq.4.25 for further calculations and by taking the laser profile to be

Gaussian, $A^2 = [f(z - v_{g1}t)]^2$ and $f(z - v_{g1}t) = A_0 e^{-\left(\frac{z - v_{g1}t}{\tau v_{g1}}\right)^2}$ where $v_{g1} = c\sqrt{1 - (\omega_p^2 / \omega^2)}$,

$v_{g2} = k_2(c^2 / 2\omega)\sqrt{1 - (1/4)(\omega_p^2 / \omega^2)(\varepsilon_m + \varepsilon_s) / \varepsilon_m \varepsilon_s}$ and τ is laser pulse width. Using a

new set of variables $z - v_{g1}t = \Omega$, $z = \eta$, $\Omega_0 = \tau v_{g1}$, $\beta' = (1 - v_{g1} / v_{g2})$, $z' = z\Omega_0$ and

$t' = v_{g1}t / \Omega_0$ we get,

$$\frac{\partial}{\partial z} = \frac{\partial}{\partial \Omega} + \frac{\partial}{\partial \eta} \quad (4.26)$$

$$\frac{\partial}{\partial t} = -v_{g1} \frac{\partial}{\partial \Omega} \quad (4.27)$$

Using new set of variables from Eq.4.26 and Eq.4.27 in Eq.4.25 becomes

$$\beta' \frac{\partial A_2}{\partial \Omega} + \frac{\partial A_2}{\partial \eta} = \beta A_0^2 e^{-\left(\frac{\Omega}{\Omega_0}\right)^2} 2\left(\frac{\Omega}{\Omega_0}\right) v_{g1} \quad (4.28)$$

Complimentary solution for Eq.4.28 is

$$A_2 = v_{g1} \beta A_0^2 f(\Omega - \beta' \eta) \quad (4.29)$$

particular solution for Eq.4.28 is

$$A_2 = \frac{v_{g1} \beta A_0^2}{\beta'} f(\Omega)$$

where

$$f(\Omega) = \int e^{e^{-\left(\frac{\Omega}{\Omega_0}\right)^2}} 2\left(\frac{\Omega}{\Omega_0}\right) d\Omega = e^{e^{-\left(\frac{\Omega}{\Omega_0}\right)^2}} \quad (4.30)$$

so the complete solution of Eq.4.28 using Eq.4.29 and Eq.4.30 is

$$A_2 = \frac{v_{g1} \beta A_0^2 \sqrt{\pi} \Omega_0}{2\beta'} \left[\text{Erf}\left(\frac{\Omega}{\Omega_0}\right) - \text{Erf}\left(\frac{\Omega - \beta' \eta}{\Omega_0}\right) \right] \quad (4.31)$$

$$\left| \frac{A_2}{A_0} \right| = \frac{v_{g1} \beta A_0^2 k_2 \sqrt{\pi} \Omega_0}{2\beta'} [\text{Erf}(z' - t') - \text{Erf}((1 - \beta')z' - t')] \quad (4.32)$$

using normalized second harmonic amplitude as,

$$\left| \frac{A_2}{A_0} \right| = \left\{ \left(1 - \frac{\omega_p^2}{\omega^2} \right)^{1/2} \left(\frac{a \omega_p^2}{\omega^2} \right) \left(\frac{eB_0}{m\omega} \right) \left(\frac{eA_0}{mc\omega} \right) (d + b) \left[\text{Erf}(z' - t') - \text{Erf}\{(1 - \beta')z' - t'\} \right] \right\}$$

$$\text{where, } a = \frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s}, d = \frac{k_1 \Omega_0 \sqrt{\pi} n'}{8\beta'}, \text{ and } b = \frac{(k_1 + k_0) \Omega_0 \sqrt{\pi} n'^2}{4\beta'} \quad (4.33)$$

The analysis shows the dependence of amplitude of second harmonic generated on the wiggler field. Stronger is the field applied, more is the enhancement of amplitude observed.

4.3 Results and discussion

Eq.4.33 has been solved numerically for Nd:YAG laser with wavelength 1064nm and intensity $I \approx 10^{12} \text{ W/cm}^2$. The refractive index of prism lies in the range 1.5–2. The dielectric permittivity are taken as 57.909 for Ag, 49.58 for Cu, 106.82 for Al and 19.5 for InSb at 1064nm [19]–[21]. Here, the layers of metal and semiconductor are considered to be of few nano-meters and doping level of n-InSb is adjusted to have $\omega_p / \omega < 1$ for semiconductor. The graphical relationship for normalized second harmonic amplitude with normalized propagation distance has been presented for different metal-semiconductor interfaces at different values of time and wiggler field. The other parameters of Eq.4.33 are taken as $\omega_p / \omega_1 = 0.9$, $n' = 2.5$, $eA_0 / mc\omega_1 = 0.09$, $(k_1 + k_0) \Omega_0 \sqrt{\pi} / 8\beta' = 500$, $eB_0 / m\omega = 0.006, 0.012$ and 0.018 , $k_1 \Omega_0 \sqrt{\pi} / 8\beta' = 300$ and $\beta' = 0.3$.

Fig.4.2 shows the variation of normalized second harmonic amplitude A_2/A_0 with normalized propagation distance z' for metals like Cu, Ag and Al with thin layer of n-InSb at time $t' = 4$ in the presence of wiggler magnetic field. It has been observed that as the strength of magnetic field increases, amplitude of second harmonic increases. In Fig.4.2(a), a sharp increase in amplitude has been observed at $z' = 5$ for all three metal-semiconductor interfaces at $eB_0 / m\omega = 0.006$ and increase in amplitude is more for Cu-InSb interface. The presence of wiggler field helps to attain phase matching conditions by providing additional momentum to the second harmonic photon. The electromagnetic fields of laser interact with electrons of metal and excite them. The excited electrons beat with magnetic field of laser and wiggler field, leading to

resonant SPW. These waves with high amplitude interact nonlinearly with electrons of n-InSb, leading to second harmonic. Fig.4.2(b) and Fig.4.2(c) show the variation of normalized second harmonic amplitude A_2/A_0 with normalized propagation distance z' for all three metal-semiconductor interfaces at $eB_0/m\omega = 0.012$ and $eB_0/m\omega = 0.018$ respectively. The values of other parameters for Fig.4.2(b) and Fig.4.2(c) are same as for Fig.4.2(a). As observed graphically under given conditions, increase in amplitude of SHG has been found more for Cu-InSb than for Ag-InSb and Al-InSb. The reason for this observation can be understood as the value of the conduction electron density for Cu is more as compared to Ag and Al so the fields of SPW of Cu are expected to be comparatively stronger and can efficiently interact with electrons of n-type semiconductor. As per the study by Takagi *et al.*[20] surface plasma waves in Kretschmann geometry shows good agreement with bulk dielectric function for Cu than Ag. Similarly in our work, we have seen better enhancement in the amplitude of second harmonic for Cu in the presence of wiggler field. Hashemzadeh *et al.* [22] studied SHG of hollow Gaussian beam in inhomogenous plasma and found the increase in amplitude of SHG with the increase in wiggler field. Sharma *et al.*[23] studied SHG of Cosh-Gaussian laser beam in magnetized plasma and found enhancement in efficiency in the presence of wiggler field. Similarly, in the present work good enhancement in amplitude of second harmonic has been observed in the presence of wiggler field.

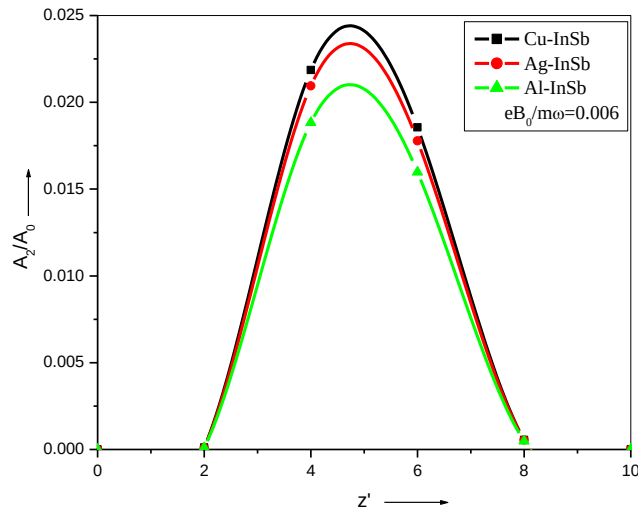


Fig.4.2(a)

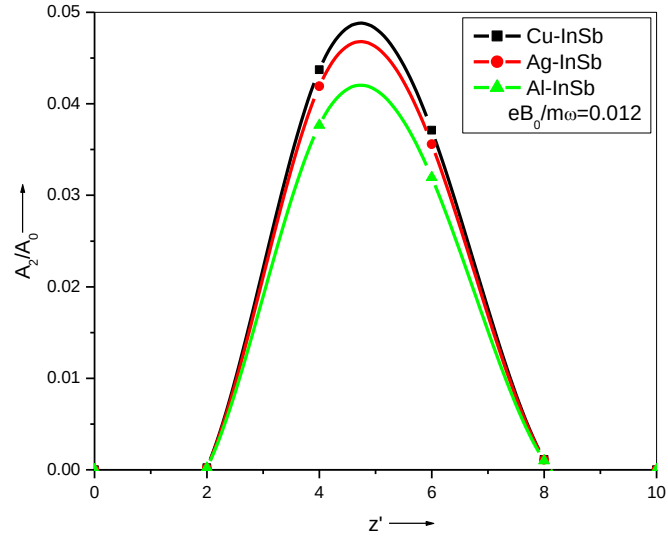


Fig.4.2(b)

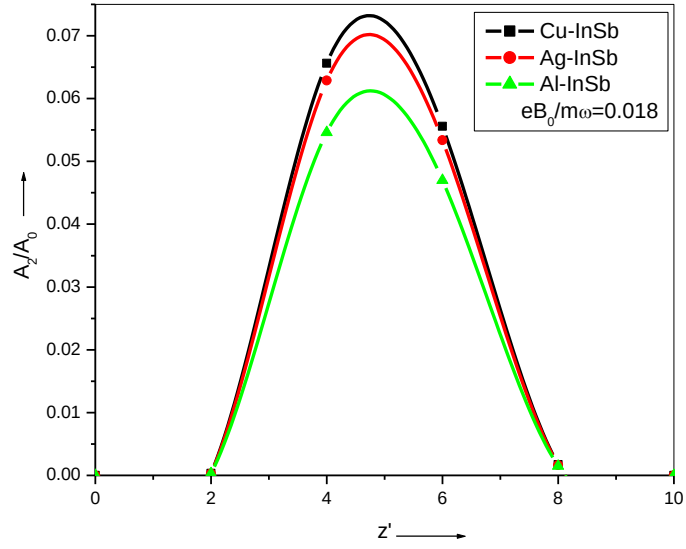


Fig.4.2(c)

Fig.4.2. Plot showing the variation of normalized second harmonic amplitude A_2/A_0 with normalized propagation distance z' for metals Cu, Ag and Al with layer of n-InSb for different values of wiggler field at time $t' = 4$

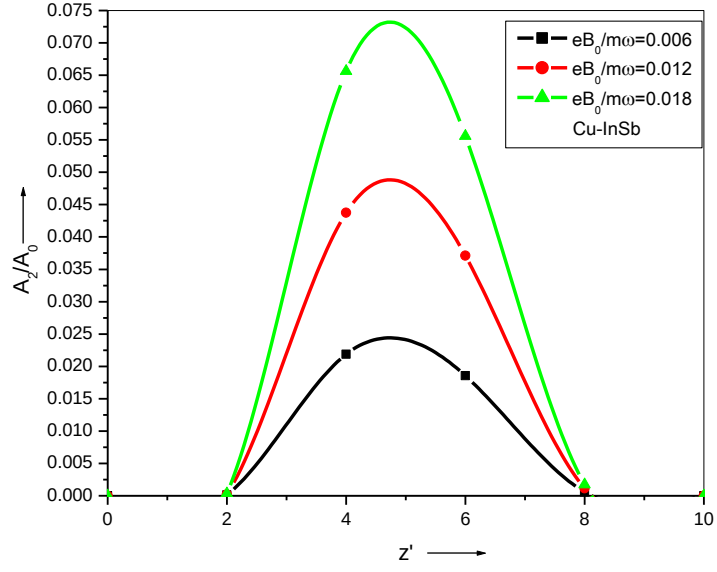


Fig.4.3

Fig.4.3. Plot showing the variation of normalized second harmonic amplitude A_2/A_0 with normalized propagation distance z' for Cu-InSb interface for different values of wiggler field at time $t' = 4$

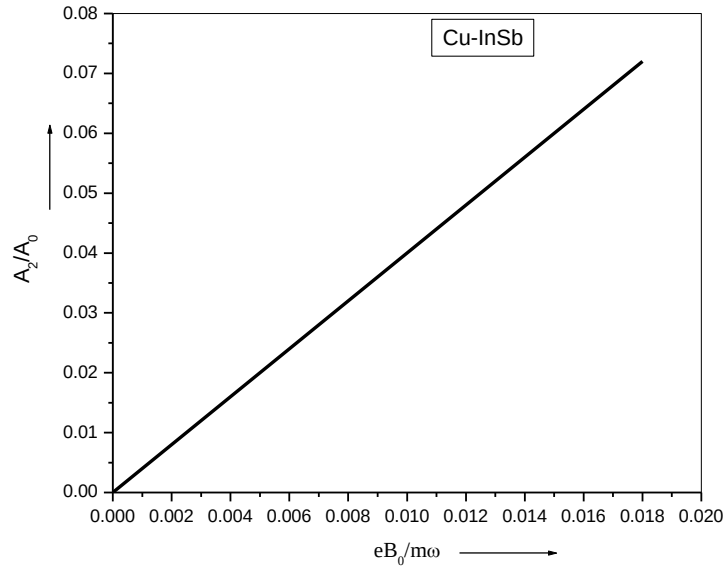


Fig.4.4

Fig.4.4. Plot showing variation of normalized amplitude of SHG for Cu-InSb interface with wiggler field strength

Fig.4.3 shows the variation of the normalized amplitude of second harmonic, A_2/A_0

with normalized propagation distance z' at different values of wiggler field with $eB_0/m\omega = 0.006, 0.012$ and 0.018 for Cu-InSb interface. The values of other parameters have been taken same as taken in Fig.4.2. It is clear from Fig.4.3 that with the slight increase in wiggler field strength, the amplitude of second harmonic enhances rapidly. In Fig.4.4 variation of normalized second harmonic amplitude with magnitude of magnetic field under resonant condition has been shown for Cu-InSb interface. It can be seen from Fig.4.4 that amplitude of harmonic increases linearly with the increase of magnetic field under resonant conditions. Aggarwal *et al.*[24] studied second harmonic generation in atomic clusters in the presence of wiggler magnetic field and they found increase in amplitude of second harmonic with the wiggler field strength. As per their work amplitude of second harmonic shows enhancement in the region where $13 < z' < 16$, for different values of wiggler frequency. In the present work, we have used similar values for wiggler frequency and have observed sharp increase in amplitude at $z' = 5$, for Cu-InSb interface. Fig.4.2 reveals that enhancement in amplitude is more for Cu-InSb as compared to Ag-InSb and Al-InSb interfaces. Our work shows that metallic surface like Cu-InSb interface would be a better choice to study the second harmonic generation and its applications.

4.4 Conclusion

The electromagnetic fields of laser interact with electrons of the metal and generate SPW, which are electromagnetic waves with higher amplitude. The SPW of different metals like Cu, Ag and Al interact nonlinearly with electrons of n-InSb, leading to second harmonic generation. The interaction has been studied in the presence of the wiggler field with different values. It has been observed that amplitude of SHG increases sharply at the Cu-InSb interface as compared to Ag-InSb and Al-InSb with the increase in wiggler field strength. The presence of the wiggler field helps in phase matching and resonate the process of second harmonic generation.

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CHAPTER 5

SECOND HARMONIC GENERATION BY CROSSED SURFACE PLASMA WAVES OVER A METALLIC SURFACE WITH DENSITY RIPPLE

5.1 Introduction

The surface plasma waves over metallic surfaces has gained much attention of researchers in the past few years due to their applications in energy conversion, radiation generation, material ablation, sensors, etc.[1-5]. SPW are electromagnetic waves with frequency nearly same as the incident light but amplitude higher than the incident light and with energy localized near the interface. Even at the modest powers, they can have high electric fields and can lead to nonlinear phenomena like harmonic generation, wave mixing, Surface Enhanced Raman Scattering(SERS), etc.[6-10]. SPW are stronger near the interface. Nonlinear mixing of oppositely traveling surface plasmons was studied by Corvi and Schaich[11], they observed that the presence of free electrons gives rise to nonlinearity. They also observed that the phase mismatch makes the process non-resonant, yet the conversion efficiency of surface plasmons is higher than that of laser. The generation of the second harmonic by the nonlinear mixing of two SPW propagating at an angle to each other over the metal-air interface was studied by Jatav and Parashar[12]. Their study concluded that the mode of excited second harmonic can be radiation mode or surface mode depending upon the angle and the electron density of the metal. Surface mode needs surface ripple for resonance and this mode is relatively more efficient. Jain *et al.* [13] studied space periodic heating of electrons by two surface plasma waves propagating at an angle to each other over the metal-vacuum interface. They observed that coupling of SPW results in the enhancement of SPW fields in the vicinity of the interface.

The presence of density ripple also affects harmonic generation significantly as harmonic yield in metals can be increased by fabricating density ripple with the required wave number over a metallic film. The coupling of laser through surface ripple accounts for the wave number matching between the laser and surface plasma

waves and the required ripple wave number decreases with the increase in the frequency of incident radiation. The choice of geometry depends on the thickness of metal, as for the thin layer of metal Kretschmann configuration is preferred. In ATR Kretschmann configuration, light is incident from glass to metal, leading to surface plasma wave. Efficient SPW is produced if the component of the wave number of the incident laser along the surface matches the wave number of SPW[14]. However, in plasma, the presence of an exponential density ramp helps to enhance the efficiency of SHG [15].

In this chapter, we present a model for efficient SHG induced by crossed SPW at a metal-semiconductor interface. SHG is studied for Cu, Ag and Al with semiconductor n-InSb using the ponderomotive mechanism between laser fields and electrons. A thin layer of semiconductor is considered over metal on glass prism in Kretschmann ATR configuration. Laser fields interact with the electrons of metal and produce SPW. Two SPW inclined at angles θ and $-\theta$ with z-axis propagate over the metal-semiconductor interface. The coupled fields of these SPW interact nonlinearly with the electrons of the semiconductor leading to SHG. The present study shows that phase matching occurs for $\theta = 0^\circ$ and for other angles phase mismatch occurs which can be compensated by the ripple wave number. Under optimized conditions, a significant enhancement in SHG amplitude has been observed for the Cu-InSb interface.

In section 5.2, mathematical relations of normalized second harmonic amplitude and propagation distance have been derived. The graphical variation and its outcomes have been discussed in section 5.3 and section 5.4, include conclusions.

5.2 Theoretical considerations

We consider the Gaussian profile of laser beam which is incident through glass prism on the metal surface, layered over the glass in Kretschmann ATR configuration, as shown in Fig.5.1(a). The electric and magnetic fields of incident laser be $\vec{E}$ and $\vec{B}$ respectively, such that

$$\begin{aligned}\vec{E} &= \hat{x}A(z,t)e^{-i(\omega_1 t - k_1 z)} \\ \vec{B} &= (\vec{k}_1 \times \vec{E})/\omega_1\end{aligned}\tag{5.1}$$

where, A is the amplitude of electric field of the laser, ω_1 is the frequency and k_1 is the

propagation constant of the laser.

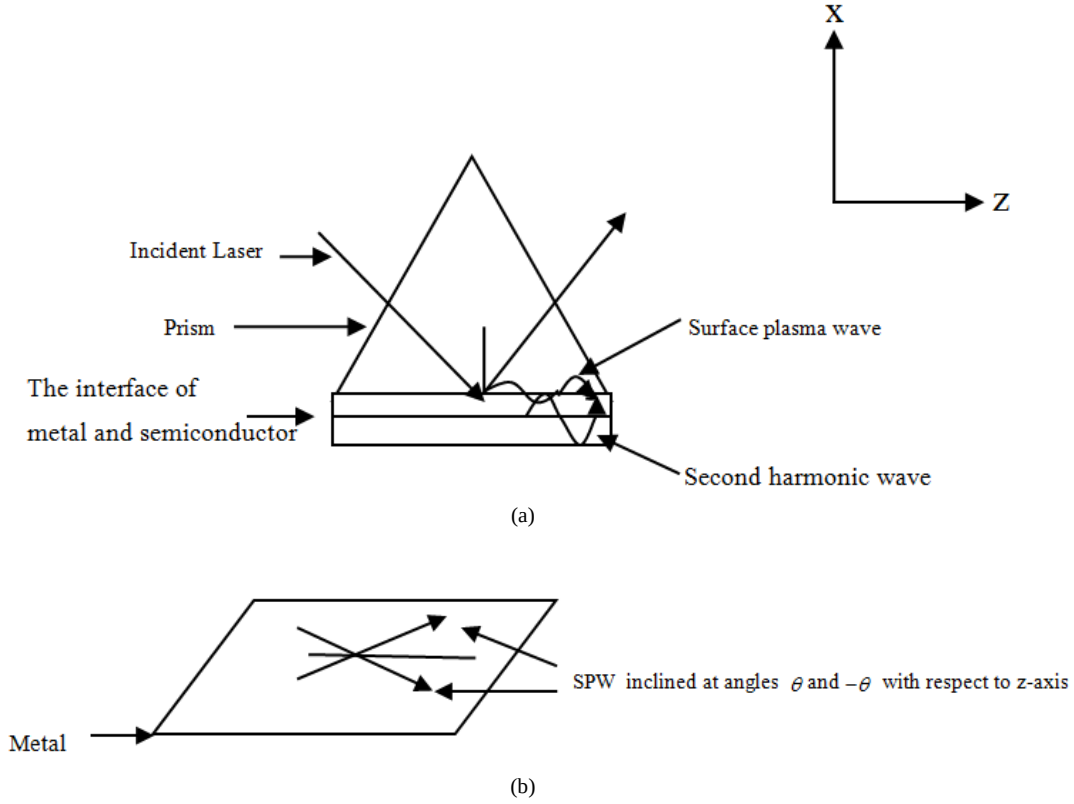


Fig.5.1 (a) Kretschmann configuration with metal and semiconductor and (b) Crossed surface plasma waves propagating on metal surface

The electromagnetic fields of laser interact with the electrons of metal and leads to formation of surface plasma waves. Two SPWs inclined at angles θ and $-\theta$ with respect to Z-axis are considered, as shown in Fig.5.1(b). The electric fields of two surface plasma waves are taken as,

$$\vec{E}_{s1} = \hat{x}A_{s1}e^{-i(\omega t - yk_s \sin\theta - zk_s \cos\theta)} \quad (5.2)$$

$$\vec{E}_{s2} = \hat{x}A_{s2}e^{-i(\omega t + yk_s \sin\theta - zk_s \cos\theta)} \quad (5.3)$$

where, $k_s = (\omega/c)(\epsilon_s \epsilon_m / (\epsilon_s + \epsilon_m))^{1/2}$ is the propagation constant of SPW with ϵ_m and ϵ_s as the permittivity of metal and semiconductor respectively, A_{s1} and A_{s2} are the amplitudes of two SPWs and ω is the frequency of surface plasma wave. The frequency of SPW and laser are nearly same[16], so $\omega_1 = \omega$. The coupled field of these two surface plasma waves is taken as $\vec{E}_s$ such that,

$$\vec{E}_s = \vec{E}_{s1} + \vec{E}_{s2} \quad (5.4)$$

The electromagnetic fields of SPW are evanescent in nature and while decaying they generate energetic electrons which further cause photons to interact with electrons of semiconductor and accelerate them. The equation of motion is,

$$m(d\vec{v} / dt) = -e\vec{E} - e\vec{E}_s, \quad (5.5)$$

Using Eq.5.1, 5.2, 5.3 and 5.4 in Eq.5.5 we get,

$$m\left(\frac{d\vec{v}}{dt}\right) = -eAe^{-i(\omega t - k_1 z)}\hat{x} - eA_{s1}e^{-i(\omega t - yk_s \sin\theta - zk_s \cos\theta)}\hat{x} - eA_{s2}e^{-i(\omega t + yk_s \sin\theta - zk_s \cos\theta)}\hat{x} \quad (5.6)$$

we obtain the oscillatory velocity $\vec{v}$ as,

$$\begin{aligned} \vec{v} &= -\frac{e}{m} \int (Ae^{-i(\omega t - k_1 z)} - A_{s1}e^{-i(\omega t - yk_s \sin\theta - zk_s \cos\theta)} - A_{s2}e^{-i(\omega t + yk_s \sin\theta - zk_s \cos\theta)})\hat{x} dt \\ \vec{v} &= \frac{-ie}{m\omega} [Ae^{-i(\omega t - k_1 z)} + A_{s1}e^{-i(\omega t - yk_s \sin\theta - zk_s \cos\theta)} + A_{s2}e^{-i(\omega t + yk_s \sin\theta - zk_s \cos\theta)}]\hat{x} \end{aligned} \quad (5.7)$$

Following Chauhan and Parashar[17], the amplitude of surface plasma wave A_s can be related to amplitude of laser as $A_s = f(A)$. According to Liu and Tripathi[18], A_s / A can be considered as a normalized parameter. Here, we have considered $A_{s1} = p' A$ and $A_{s2} = p'' A$ such that, $0 < p'$ and $p'' < 2.5$. For simplicity, we have assumed $p' = p''$. The velocity $\vec{v}$ beats with the magnetic field of laser and the velocity of electrons changes to $\vec{v}_1'$, due to ponderomotive force $\vec{F}$, where,

$$\vec{F} = -e(\vec{v} \times \vec{B}_1) \quad (5.8)$$

Using, $\vec{F} = m(d\vec{v}_1' / dt)$, we get $\vec{v}_1'$ as,

$$\begin{aligned} m\left(\frac{d\vec{v}_1'}{dt}\right) &= -e\left(\frac{-ie}{m\omega} [Ae^{-i(\omega t - k_1 z)} + A_{s1}e^{-i(\omega t - yk_s \sin\theta - zk_s \cos\theta)} + A_{s2}e^{-i(\omega t + yk_s \sin\theta - zk_s \cos\theta)}]\hat{x} \times \frac{(\vec{k}_1 \times \vec{E})}{\omega}\right) \\ m\left(\frac{d\vec{v}_1'}{dt}\right) &= -e\left(\frac{-ie}{m\omega} [Ae^{-i(\omega t - k_1 z)} + A_{s1}e^{-i(\omega t - yk_s \sin\theta - zk_s \cos\theta)} + A_{s2}e^{-i(\omega t + yk_s \sin\theta - zk_s \cos\theta)}]\hat{x} \times \frac{(k_1 Ae^{-i(\omega_1 t - k_1 z)})}{\omega}\hat{y}\right) \\ m\left(\frac{d\vec{v}_1'}{dt}\right) &= -e\left(\frac{-ie}{m\omega} [Ae^{-i(\omega t - k_1 z)} + A_{s1}e^{-i(\omega t - yk_s \sin\theta - zk_s \cos\theta)} + A_{s2}e^{-i(\omega t + yk_s \sin\theta - zk_s \cos\theta)}]\frac{(k_1 Ae^{-i(\omega_1 t - k_1 z)})}{\omega}\right)\hat{z} \end{aligned}$$

$$\begin{aligned}\vec{v}_1' &= \frac{ie^2 k_1 A}{m\omega^2} \int (e^{-i(2\omega t - 2k_1 z)} + p' e^{-i(2\omega t - yk_s \sin\theta - zk_s \cos\theta - k_1 z)} + p' e^{-i(\omega t + yk_s \sin\theta - zk_s \cos\theta - k_1 z)}) \hat{z} dt \\ \vec{v}_1' &= \frac{-e^2 A^2 k_1}{2m^2 \omega^3} [e^{-i(2\omega t - 2k_1 z)} + p' e^{-i(2\omega t - yk_s \sin\theta - zk_s \cos\theta - k_1 z)} + p' e^{-i(\omega t + yk_s \sin\theta - zk_s \cos\theta - k_1 z)}] \hat{z}\end{aligned}\quad (5.9)$$

The present model is based on ponderomotive mechanism; the accelerating force experienced by electrons could overpower the other interactions between metal ions and electrons. The density profile for rippled metallic surface is considered to be,

$$n_q = n_0 e^{iqz}$$

where, n_0 is the equilibrium electron density, q is the ripple wave number and z is the propagation distance. This velocity $\vec{v}_1'$ gives rise to nonlinear effects and it couples with density ripple to produce second harmonic current at $(2\omega, k_2 = 2k_1 + q)$ given by

$$\begin{aligned}\vec{J}_{NL} &= n_0 e^{iqz} e \vec{v}_1' \\ \vec{J}_{NL} &= \frac{-e^3 n_0 A^2 k_1}{2m^2 \omega^3} [e^{-i(2\omega t - 2k_1 z - qz)} + p' e^{-i(2\omega t - yk_s \sin\theta - zk_s \cos\theta - k_1 z - qz)} + p' e^{-i(\omega t + yk_s \sin\theta - zk_s \cos\theta - k_1 z - qz)}] \hat{z}\end{aligned}\quad (5.10)$$

Here, q is chosen such that it helps to attain phase matching condition, which is an essential requirement for resonant wave propagation with

$$q = k_2 - 2k_1 \quad (5.11)$$

For efficient coupling of laser with SPW, the wave vector of laser along the surface should be equal to the wave vector of surface plasma wave. This can be achieved if,

$$k_1 z = \pm yk_s \sin\theta - zk_s \cos\theta \quad (5.12)$$

for small angles $\sin\theta \approx \theta$ and $\cos\theta \approx 1 - \frac{\theta^2}{2}$ can be used.

The field equation for second harmonic is taken as

$$\vec{E}_2 = A_2 e^{-i(2\omega t - k_2 z)} \hat{x} \quad (5.13)$$

with A_2 as the amplitude and k_2 as the propagation constant of second harmonic. The

field $\vec{E}_2$ imparts linear velocity $\vec{v}_{2L}$ to electrons

$$m \frac{d\vec{v}_{2L}}{dt} = -e\vec{E}_2 \quad (5.14)$$

Using Eq.5.12 in Eq.5.13,

$$\begin{aligned} m \frac{d\vec{v}_{2L}}{dt} &= -eA_2 e^{-i(2\omega t - k_2 z)} \hat{x} \\ \vec{v}_{2L} &= -\frac{e}{m} A_2 \int e^{-i(2\omega t - k_2 z)} \hat{x} dt \\ \vec{v}_{2L} &= -\frac{ie}{2m\omega} A_2 e^{-i(2\omega t - k_2 z)} \hat{x} \end{aligned} \quad (5.15)$$

$\vec{v}_{2L}$ produces linear second harmonic current at $(2\omega, k_2)$ given by

$$\vec{J}_L = n_0 e \vec{v}_{2L}$$

Using Eq.5.13,

$$\vec{J}_L = -\frac{in_0 e^2}{2m\omega} A_2 e^{-i(2\omega t - k_2 z)} \hat{x} \quad (5.16)$$

The total current density is given by,

$$J = J_{NL} + J_L \quad (5.17)$$

The amplitude of second harmonic generated can be calculated by using wave equation for second harmonic,

$$\begin{aligned} \nabla^2 \vec{E}_2 &= \frac{4\pi(\epsilon_m + \epsilon_s)}{c^2(\epsilon_m \epsilon_s)} \frac{\partial \vec{J}}{\partial t} + \frac{1}{c^2} \frac{\partial^2 \vec{E}_2}{\partial t^2} \\ \frac{\partial^2 E_2}{\partial z^2} &= \frac{4\pi(\epsilon_m + \epsilon_s)}{c^2(\epsilon_m \epsilon_s)} \frac{\partial \vec{J}}{\partial t} + \frac{1}{c^2} \frac{\partial^2 E_2}{\partial t^2} \end{aligned} \quad (5.18)$$

Using Eq.5.10, 5.12, 5.14 and 5.15 in Eq.5.16

$$\begin{aligned} \frac{\partial^2 (A_2 e^{-i(2\omega t - k_2 z)})}{\partial z^2} &= \frac{4\pi(\epsilon_m + \epsilon_s)}{c^2(\epsilon_m \epsilon_s)} \frac{\partial}{\partial t} \left(\frac{-e^3 n_0 A^2 k_1}{2m^2 \omega^3} \left[e^{-i(2\omega t - 2k_1 z + qz)} + p' e^{-i(2\omega t - yk_s \sin\theta - zk_s \cos\theta - zk_1 + qz)} + \right] \right) \\ &\quad + \frac{4\pi(\epsilon_m + \epsilon_s)}{c^2(\epsilon_m \epsilon_s)} \frac{\partial}{\partial t} \left(-\frac{in_0 e^2}{2m\omega} A_2 e^{-i(2\omega t - k_2 z)} \right) + \frac{1}{c^2} \frac{\partial^2 (A_2 e^{-i(2\omega t - k_2 z)})}{\partial t^2} \end{aligned} \quad (5.19)$$

Using Eq.5.11 in Eq.5.19 we get,

$$\begin{aligned} \frac{\partial^2 (A_2 e^{-i(2\omega t - k_2 z)})}{\partial z^2} &= \frac{4\pi(\varepsilon_m + \varepsilon_s)}{c^2(\varepsilon_m \varepsilon_s)} \frac{\partial}{\partial t} \left(\frac{-e^3 n_0 A^2 k_1 (1 + 2p')}{2m^2 \omega^3} [e^{-i(2\omega t - 2k_1 z + qz)}] \right) \\ &+ \frac{\partial^2 (A_2 e^{-i(2\omega t - k_2 z)})}{c^2 \partial t^2} \end{aligned} \quad (5.20)$$

$$\begin{aligned} \frac{\partial}{\partial z} \left(A_2 i k_2 e^{-i(2\omega_1 t - k_2 z)} + e^{-i(2\omega_1 t - k_2 z)} \frac{\partial A_2}{\partial z} \right) &= \frac{1}{c^2} \frac{\partial}{\partial t} \left(-2A_2 i \omega e^{-i(2\omega_1 t - k_2 z)} + e^{-i(2\omega_1 t - k_2 z)} \frac{\partial A_2}{\partial t} \right) + \\ \frac{4\pi}{c^2} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) &\left(-\frac{n_0 e^3 k_1}{m^2 \omega_1} \left[\frac{-2i\omega_1 (1 + 2p') A^2 e^{-i[2\omega_1 t - (k_s + k_1 + q)z]}}{2\omega_1^2} + \frac{-2i\omega_1 A^2 e^{-i(2\omega_1 t - (2k_1 + q)z)}}{2\omega_1^2} \right] \right) \end{aligned} \quad (5.21)$$

Solving Eq.5.21 then using Eq.5.11, we get real and imaginary parts as

$$\begin{aligned} \frac{\partial^2 A_2}{\partial z^2} - k_2^2 A_2 &= \frac{1}{c^2} \frac{\partial^2 A_2}{\partial t^2} - \frac{\omega^2 A_2}{c^2} + \frac{4\pi}{c^2} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \left(\frac{2n_0 e^2 A_2}{m\omega} + \frac{n_0 e^3 k_1 (1 + 2p')^2 A^2}{2\omega^3 m^2} \frac{\partial A_1}{\partial t} + \right. \\ &\left. (1 + 2p') A_1 \frac{\partial A_1}{\partial t} + \frac{n_0 e^3 k_1 A}{\omega^3 m^2} \frac{\partial A}{\partial t} \right) \end{aligned} \quad (5.22)$$

$$2k_2 \frac{\partial A_2}{\partial z} + \frac{2\omega_2}{c^2} \frac{\partial A_2}{\partial t} = \frac{4\pi}{c^2} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \left(\frac{n_0 e^2}{2m\omega} \frac{\partial A_2}{\partial t} + \frac{n_0 e^3 k_1 (1 + 2p') A^2}{\omega_1^2 m^2} \right) \quad (5.23)$$

considering Eq.5.23 for further calculations and by using

$$\alpha = \frac{4\omega}{2c^2 k_2} + \frac{4\pi m_0 e^2}{4k_2 c^2 m\omega} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \quad \text{and} \quad \beta = \frac{4\pi}{c^2} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \frac{n_0 e^3 k_1 (1 + 2p')}{2k_2 \omega_1^2 m^2}$$

Eq. (5.23) becomes

$$\frac{\partial A_2}{\partial z} + \alpha \frac{\partial A_2}{\partial t} = \beta A_1^2 \quad (5.24)$$

Considering Eq.5.24 and by taking the laser profile to be Gaussian as

$$A^2 = [f(z - v_g t)]^2 \quad \text{and} \quad f(z - v_g t) = A_0 e^{-\left(\frac{z - v_g t}{\tau v_g}\right)^2} \quad \text{with} \quad v_g = c\sqrt{(\varepsilon_m + \varepsilon_s)/\varepsilon_m \varepsilon_s} \quad \text{and} \quad \tau \text{ is laser pulse width.}$$

Using a new set of variables $z - v_g t = \zeta$, $z = \eta$, $\zeta_0 = \tau v_g$, $\beta' = (1 - (v_g/c)^2)$, $z' = z/\zeta_0$ and $t' = v_g t/\zeta_0$ we get,

$$\alpha \approx \frac{v_g}{c^2}, \quad \frac{\partial}{\partial z} = \frac{\partial}{\partial \zeta} + \frac{\partial}{\partial \eta} \quad \text{and} \quad \frac{\partial}{\partial t} = -v_g \frac{\partial}{\partial \zeta} \quad (5.25)$$

complimentary solution for Eq.5.24 using the above variables is

$$A_2 = \frac{-\beta A_0^2}{\beta'} f(\zeta - \beta' \eta) \quad (5.26)$$

and the particular solution is,

$$A_2 = \frac{-\beta A_0^2}{\beta'} \int \exp\left(\frac{-\zeta^2}{\zeta_0^2}\right) d\zeta, \quad (5.27)$$

So complete solution gives normalized second harmonic amplitude as,

$$\left| \frac{A_2}{A_0} \right| = \frac{\omega_p^2}{\omega^2} \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \left(\frac{eA_0}{mc\omega} \right) (1 + 2p') \left(\frac{k_1 \xi_0 \sqrt{\pi}}{8\beta'} \right) [Erf(z' - t') - Erf\{(1 - \beta')z' - t'\}] \quad (5.28)$$

The amplitude has been derived under resonant conditions. The angle between two SPW and ripple wave number provides phase matching conditions. The dependence of amplitude on θ is determined by the relation in Eq.5.12

5.3 Results and discussion

The material of prism and the reflectivity of metal influence the angle of incidence as well as the amount of reflected light. Laser fields interact with the electrons of metal to generate SPW, which are electromagnetic waves with amplitude higher than that of the incident laser. Fig.5.2 shows the variation of normalized propagation constant (k_1/k_s) with θ , using Eq.5.12, for (a) $y=1$ and $z=1$ and for (b) $y=2$ and $z=1$. It is clear from Fig.5.2, that for $\theta=0^\circ$, (k_1/k_s) approaches to 1. The process of second harmonic generation becomes resonant, if the wave vector of the surface plasma wave matches with the wave vector of the laser. It is clear from Fig.5.2 that as the angle θ increases, phase mismatch occurs which may cause attenuation of SPW. The presence of ripple wave number q , where $q = k_2 - 2k_1$, helps to overcome this problem as it provide the additional momentum to photons of second harmonic. Jatav and Parashar[12] have studied the second harmonic generation on the metal-air interface by crossed SPWs and have found that the process is sensitive to angle between the SPWs and show periodic variation. Similarly in our work, we have observed the dependence of the SHG process on the angle between the two SPWs with periodic

variation.

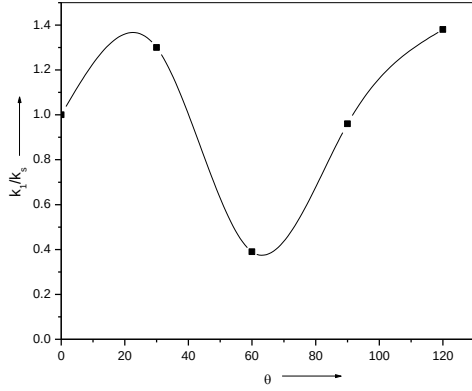


Fig.5.2(a)

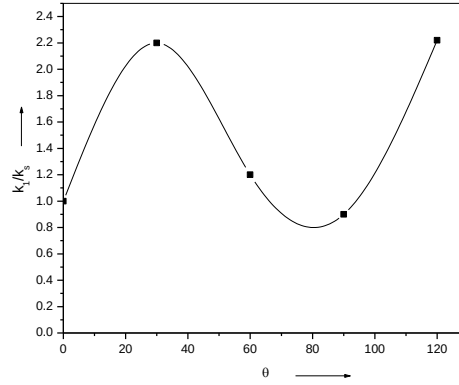


Fig.5.2(b)

Fig.5.2 Plot showing the variation of normalized propagation constant k_1 / k_s with angle θ between two SPWs, Fig.2(a) for $y=1$ and $z=1$ and Fig.2(b) for $y = 2$ and $z = 1$

Eq.5.28 has been solved numerically for Nd:YAG laser with wavelength $1.064\mu\text{m}$ and the other parameters are taken as $\omega_p / \omega = 0.85$, $p'=1$, $eA_0 / mc\omega = 0.01$, $k_1 \xi_0 \sqrt{\pi} / 8\beta' = 5$ and $\beta' = 0.3$. The absolute values of effective permittivity at $1.064\mu\text{m}$ are taken as 49.58 for Cu, 57.909 for Ag, 106.82 for Al and 19.5 for InSb [19]–[21]. Metal and semiconductor layers have been considered of few nano-meters and doping level of n-InSb is adjusted to have $\omega_p / \omega < 1$ for semiconductor. Fig.5.3 shows the variation of normalized second harmonic amplitude A_2 / A_0 with normalized propagation distance z' for metals Cu, Ag and Al with thin layer of n-InSb at different time $t' = 0, 3$ and 6. Fig.5.3(a) depicts the variation of A_2 / A_0 with z' at $t' = 0$ and a sharp increase in the normalized second harmonic amplitude is seen at $z' = 2$. Increase in amplitude of SHG is observed more for Cu-InSb than that of Ag-InSb and Al-InSb, under phase matching conditions. This is due to the reason that Cu has less reflectivity than Ag and Al, so the fields of SPW for Cu can interact more efficiently with the electrons of n-type semiconductor and can generate second harmonic with enhanced amplitude. Thakur and Kant[22] have studied SHG in plasma with exponential density profile under wiggler magnetic field and they noticed the sharp increase in amplitude at $z' > 8$ for $\omega_p / \omega_1 > 0.5$, but the present work shows that the

sharp increase in amplitude of second harmonic induced by crossed SPW at metal-semiconductor interface occurs at $z' < 8$ for $\omega_p / \omega_1 > 0.5$ in the absence of external agent like wiggler field. Hence, the propagation distance for second harmonic decreases for the metal-semiconductor interface as compared to the plasma. Similarly, Fig.5.3(b) and (c) show the increase in amplitude of second harmonic with time and enhancement in SHG amplitude is more for Cu-InSb interface than for Ag-InSb and Al-InSb interfaces.

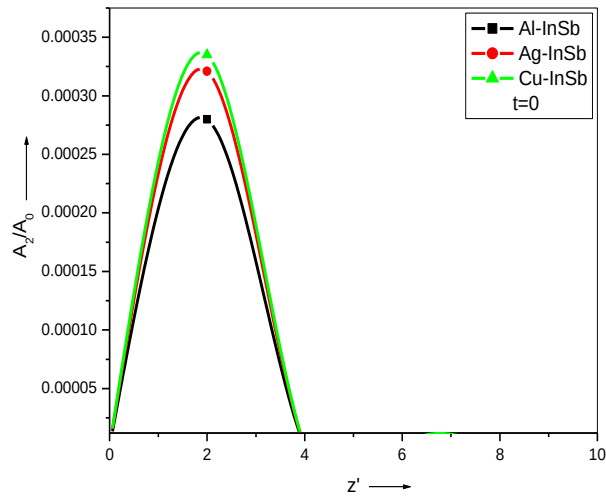


Fig.5.3(a)

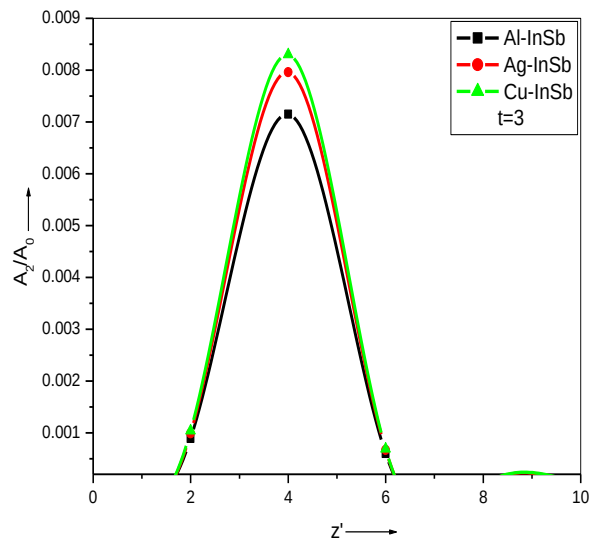


Fig.5.3(b)

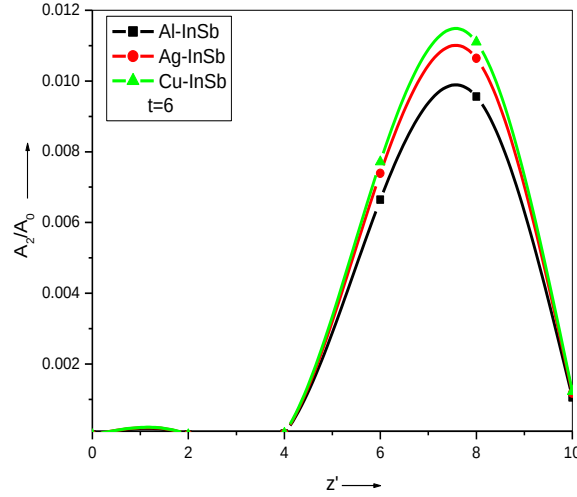


Fig.5.3(c)

Fig.5.3. The variation of normalized second harmonic amplitude A_2/A_0 with normalized propagation distance z' for Cu, Ag and Al with layer of n-InSb at different values of time. (a) at $t'=0$, (b) at $t'=3$ and (c) at $t'=6$.

5.4 Conclusions

The interaction of laser with electrons of metals leads to surface plasma waves. Two SPW inclined at angles θ and $-\theta$ with the z -axis are propagating over metal-semiconductor interface, on coupling they give rise to electromagnetic fields capable of prompting photons which can further interact with electrons of n -type semiconductor, layered over metal. The metal surface is rippled to achieve phase matching conditions. Two SPW with $\theta = 0^\circ$ results in the efficient second harmonic generation and with the change in angle phase mismatch occurs which is compensated by the ripple wave number q . It is observed that at resonance the enhancement in SHG has been found more for the Cu-InSb than that of Ag-InSb and Al-InSb.

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CHAPTER 6

EFFECT OF SELF-FOCUSING ON SECOND HARMONIC GENERATION BY SURFACE PLASMA WAVE

6.1 Introduction

Surface plasma waves (SPW) formed on the surface of a metal due to interaction between laser and electrons has gained many applications in the field of biology, technology and industry [1-9]. These waves are guided electromagnetic waves propagating along the interface of a metal and a dielectric. The fields of laser and electrons interact nonlinearly which results in the self-focusing of the wave. The self-focusing of laser helps to improve the efficiency of harmonic generation. The smaller phase velocity of axial portion of SPW than the non-axial phase results in self-focusing [10]-[11]. The presence of an exponential density ramp enhances the self-focusing of the laser which in turn strengthens the SHG in plasma. The density ripple couples with the laser beam and helps in phase matching which increases the efficiency of SHG [12]-[14]. The presence of factors like magnetic field, focusing of the pump laser and increase in the collision frequency supports self-focusing of laser. The effect of self-focusing of laser with different laser profiles like Cosh-Gaussian, Laguerre-Gaussian, Hermite Gaussian etc. under different conditions of plasma have also been studied. In general, it has been observed that the self-focussed laser beam leads to efficient harmonic generation [15]-[20].

The parameters like density profile and intensity play a significant role in the self-focusing of the laser beam. Self-focusing of an intense laser beam is due to relativistic mass and ponderomotive effects. The increase in plasma density leads to stronger self-focusing and the beam width parameter attains a minimum value [21]-[22].

This chapter includes the investigation of the effect of the self-focusing of laser on SHG induced by SPW on the metal-semiconductor interface. These interactions are governed by the ponderomotive mechanism. The presence of nonlinearity accounts for the self-focusing of the laser beam which in turn influences the efficiency of SHG. Section 6.2 and 6.3 includes derivations for equations of beam width parameter and

second harmonic amplitude respectively, results and graphical discussion is included in section 6.4. The conclusion is presented in section 6.5.

6.2 Equation for beam width parameter

We considered the metal-semiconductor interface in Kretschmann ATR configuration as shown in Fig.6.1, with nanometer thick layers of metal and semiconductor. Metals considered for analysis are Cu, Ag and Al with semiconductor n-InSb. Light incident on glass-metal interface produces SPW on metal surface, which is rippled to achieve phase matching condition for a resonant harmonic generation. The electromagnetic fields of SPW can penetrate through the plasmonic entities of semiconductors and can interact with the electrons.

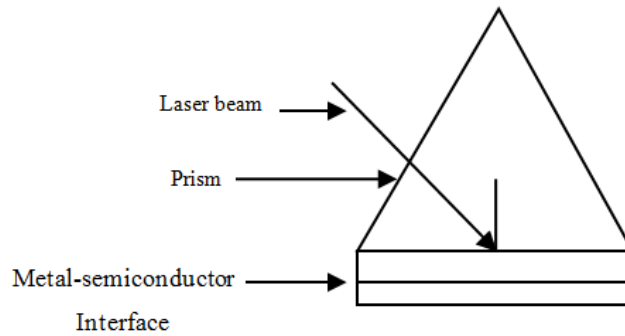


Fig.6.1. Kretschmann configuration with thin layer of a metal and a semiconductor

Laser with Gaussian profile is considered with electromagnetic fields as,

$$\vec{E} = \hat{x} A e^{-i(\omega_1 t - k_1 z)} \quad (6.1)$$

$$\vec{B} = (\vec{k}_1 \times \vec{E}) / \omega_1 \quad (6.2)$$

where, ω_1 and k_1 are frequency and propagation constant of the laser respectively and A is the amplitude of electric field of the laser with,

$$A = (A_{i0} / f_l(z)) e^{\frac{-r^2}{r_0^2 f_l^2}} \quad (6.3)$$

where, A_{i0} is constant amplitude of the laser pulse, f_l is the beam width parameter of the laser pulse and r_0 are the spot size of the laser at $z = 0$. These fields interact with

electrons of metal and produces surface plasma waves with electric field,

$$\vec{E}_s = \hat{x}A_s e^{-i(\omega t - k_s z)} \quad (6.4)$$

where, k_s is the propagation constant of SPW such that,

$$k_s = (\omega/c) \left(\epsilon_s \epsilon_m / (\epsilon_s + \epsilon_m) \right)^{1/2} \quad (6.5)$$

with ϵ_m and ϵ_s as the permittivity of metal and semiconductor respectively and ω is the frequency of surface plasma wave. Amplitude of surface plasma wave A_s depends on amplitude of incident laser A as $A_s = f(A)$ and A_s / A can be considered as a normalized parameter with $A_s = jA$ where, j is a positive real number. Also, frequency of SPW is same as frequency of laser so, $\omega_1 = \omega$ [23]. The electrons of semiconductor get accelerated due to the electric fields of SPW, their velocity can be calculated using equation of motion

$$m(d\vec{v}_1/dt) = -e\vec{E}_s - e\vec{E} \quad (6.6)$$

and it comes out to be,

$$\begin{aligned} v_1 &= \int \left(-\frac{e}{m} jA e^{-i(\omega t - k_s z)} - \frac{e}{m} A e^{-i(\omega_1 t - k_1 z)} \right) dt \\ v_1 &= \left[\frac{-ejA e^{-i(\omega t - k_s z)}}{m(-i\omega)} - \frac{-eA e^{-i(\omega_1 t - k_1 z)}}{m(-i\omega_1)} \right] \end{aligned} \quad (6.7)$$

The magnetic field of laser beats with the velocity of electrons and exerts ponderomotive force $\vec{F} = -e(\vec{v}_1 \times \vec{B})$ which imparts an oscillatory velocity to electrons as,

$$\vec{v} = -\frac{e^2 k_l}{m^2} \left[\frac{jA_l^2 e^{-i[(2\omega_l)t - (k_s + k_l)z]}}{2\omega_l^3} + \frac{A_l^2 e^{-i(2\omega_l t - 2k_l z)}}{2\omega_l^3} \right] \hat{z} \quad (6.8)$$

Metal surface is rippled with density profile, $n_q = n_0 e^{iqz}$ where, n_0 is the equilibrium electron density, q is the ripple wave number and z is the propagation distance. The non linear velocity $\vec{v}$ couples with density ripple and produce second harmonic current given by at $(2\omega_l, 2k_l + q)$

$$\vec{J}_{NL} = n_0 e^{iqz} e\vec{v} \quad (6.9)$$

Here, q is chosen such that $k_2 = 2k_1 + q$ and it obeys phase matching condition. The field equation for second harmonic is taken as,

$$\vec{E}_2 = A_2 e^{-i(2\omega_1 t - k_2 z)} \hat{x} \quad (6.10)$$

where, A_2 is the amplitude of the SHG. The linear current density due to self-consistent second harmonic field $\vec{E}_2$ at $(2\omega_1, k_2)$ is

$$\vec{J}_{2L} = n_0 e \vec{v}_{2L} \quad (6.11)$$

where, $\vec{v}_{2L}$ is obtained using equation of motion

$$m(d\vec{v}_{2L}/dt) = -e\vec{E}_2$$

$$\vec{v}_{2L} = -\frac{ie}{2m\omega} A_2 e^{-i(2\omega_1 t - k_2 z)} \hat{x} \quad (6.12)$$

So, current density can be calculated by taking,

$$J = J_{NL} + J_{2L} \quad (6.13)$$

using Eq.6.8, 6.9, 6.11, 6.12 in Eq.6.13 we get,

$$J = -\frac{n_0 e^3 k_l}{m^2} \left[\frac{j A_l^2 e^{-i[(2\omega_1)t - (k_s + k_l + q)z]}}{2\omega_l^3} + \frac{A_l^2 e^{-i(2\omega_1 t - (2k_1 + q)z)}}{2\omega_l^3} \right] - \frac{ie}{2m\omega} A_2 e^{-i(2\omega_1 t - k_2 z)} \quad (6.14)$$

The wave equation governing the propagation of laser beam can be taken as,

$$\frac{\partial^2 E_1}{\partial z^2} + \frac{\partial^2 E_1}{\partial r^2} + \frac{\partial E_1}{r \partial r} + \frac{\omega^2 (\epsilon_m + \epsilon_s) E_1}{c^2 (\epsilon_m \epsilon_s)} = 0 \quad (6.15)$$

Eq.6.15 has been solved numerically to study the variation of beam width parameter with propagation distance using Eq.6.1 and Eq.6.3. The relation of beam width parameter with normalized propagation distance is obtained by equating coefficients as,

$$\frac{\partial^2 E_1}{\partial z^2} = A_{l0} e^{-\frac{r^2 f_l^2}{r_0^2}} \left[\frac{4r^4}{f_l^7 r_0^4} \left(\frac{\partial f_l}{\partial z} \right)^2 + \frac{2r^2}{f_l^4 r_0^2} \left(\frac{\partial^2 f_l}{\partial z^2} \right) - \frac{10r^2}{f_l^5 r_0^2} \left(\frac{\partial f_l}{\partial z} \right)^2 - \frac{1}{f_l^2} \frac{\partial^2 f_l}{\partial z^2} + \frac{2}{f_l^3} \left(\frac{\partial f_l}{\partial z} \right)^2 \right] \quad (6.16)$$

$$\frac{\partial^2 E_1}{\partial r^2} = \frac{4r^2 A_{l0} e^{-\frac{r^2 f_l^2}{r_0^2}}}{f_l^5 r_0^4} - \frac{2 A_{l0} e^{-\frac{r^2 f_l^2}{r_0^2}}}{f_l^3 r_0^2} \quad (6.17)$$

$$\frac{\partial E_1}{\partial r} = -\frac{2rA_{10}e^{-\frac{r^2 f_l^2}{r_0^2}}}{f_l^3 r_0^2} \quad (6.18)$$

$$\frac{\omega^2(\varepsilon_m + \varepsilon_s)E_1}{c^2(\varepsilon_m \varepsilon_s)} = \left(\frac{\omega^2}{c^2}\right) \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s}\right) \frac{A_{10}e^{-\frac{r^2 f_l^2}{r_0^2}}}{f_1} \quad (6.19)$$

$$\begin{aligned} A_{10}e^{-\frac{r^2 f_l^2}{r_0^2}} & \left[\frac{4r^4}{f_l^7 r_0^4} \left(\frac{\partial f_l}{\partial z}\right)^2 + \frac{2r^2}{f_l^4 r_0^2} \left(\frac{\partial^2 f_l}{\partial z^2}\right) - \frac{10r^2}{f_l^5 r_0^2} \left(\frac{\partial f_l}{\partial z}\right)^2 - \frac{1}{f_l^2} \frac{\partial^2 f_l}{\partial z^2} + \frac{2}{f_l^3} \left(\frac{\partial f_l}{\partial z}\right)^2 \right] + \frac{4r^2 A_{10}e^{-\frac{r^2 f_l^2}{r_0^2}}}{f_l^5 r_0^4} \\ & - \frac{4A_{10}e^{-\frac{r^2 f_l^2}{r_0^2}}}{f_l^3 r_0^2} + \left(\frac{\omega^2}{c^2}\right) \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s}\right) \frac{A_{10}e^{-\frac{r^2 f_l^2}{r_0^2}}}{f_1} = 0 \end{aligned} \quad (6.20)$$

on comparing the coefficients of r^2 of Eq.6.20 we get,

$$\begin{aligned} \frac{d^2 f_l}{dz'^2} \left(\frac{2r^2}{r_0^2 f_l^4} + \frac{-1}{f_l^2} \right) + \left(\frac{df_l}{dz'} \right)^2 \left(\frac{2}{f_l^3} + \frac{4r^4}{r_0^4 f_l^7} - \frac{10r^2}{r_0^2 f_l^5} \right) &= \frac{k_l^2 \xi_0^2}{f_l} + \frac{4r^2 \xi_0^2}{f_l^5 r_0^4} - \frac{4\xi_0^2}{r_0^2 f_l^3} \\ &- \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \frac{\xi_0^2 \omega_l^2}{c^2 f_l} \end{aligned} \quad (6.21)$$

where, $z' = z / \xi_0$ and ξ_0 is a normalizing parameter. The terms on L.H.S of Eq.6.21 signifies the effect of ponderomotive mechanism. First three terms on R.H.S give the impact of laser propagation on laser spot size and beam width parameter. Last term on R.H.S includes the effect of material on beam width parameter.

6.3 Equation for second harmonic amplitude

The amplitude of second harmonic generated can be calculated by using wave equation obtained by using Maxwell's relations,

$$\nabla^2 \vec{E}_2 = \frac{4\pi(\varepsilon_m + \varepsilon_s)\partial \vec{J}}{c^2(\varepsilon_m \varepsilon_s)\partial t} + \frac{\partial^2 \vec{E}_2}{c^2 \partial t^2} \quad (6.22)$$

Using Eq.6.10 and Eq.6.14 in Eq.6.22 the relation for normalized second harmonic amplitude with normalized propagation distance is obtained by equating real terms as,

$$\frac{\partial^2(A_2)}{\partial z'^2} + \frac{\omega_l^2 \xi_0^2}{c^2}(A_2) + \frac{\omega_p^2 \xi_0^2 (\varepsilon_m + \varepsilon_s)}{c^2 (\varepsilon_m \varepsilon_s)}(A_2) + \left(\frac{\omega_p^2}{\omega_l^2} \right) \left(\frac{\varepsilon_m + \varepsilon_s}{\varepsilon_m \varepsilon_s} \right) \left(\frac{e A_l^2}{m c \omega_l} \right) \left(\frac{k_1 \xi_0^2 \omega_l}{c} \right) (1 + j) = 0 \quad (6.23)$$

where, variables used are $A_l = (A_{l0} / f_l(z)) e^{\frac{-r^2}{r_0^2 f_l^2}}$, $\omega_p^2 = 4\pi n_0 e^2 / m$ and $z' = z / \xi_0$.

Eq.6.28 gives relation of amplitude of second harmonic generated with beam width parameter of laser. It clearly shows that amplitude of output wave is dependent on focusing and defocusing nature of laser.

6.4. Results and graphical analysis

Eq.6.21 and Eq.6.23 have been solved numerically for Nd:YAG laser having frequency $2.8 \times 10^{14} \text{ s}^{-1}$ and intensity $I \approx 10^{12} \text{ W/cm}^2$. The values for the permittivity of the metals and the semiconductor are taken as $\varepsilon_m = 49.58$ for Cu, 57.909 for Ag, 106.82 for Al and $\varepsilon_s = 19.5$ for InSb [24]–[26]. The doping of n-InSb is adjusted such that, it acts like an under-dense plasma for the electromagnetic SPW. Eq.6.21 has been analyzed graphically to find the relationship between beam width parameter f_l and normalized propagation distance z' with other parameters taken as $\xi_0 / r_0 = 0.002$, $k_l \xi_0 = 0.5$, $\omega_l \xi_0 / c = 0.8$, $\omega_p \xi_0 / c = 0.5$ and $r / r_0 = 0.3$. Fig. 6.2 reveals that the beam width parameter attains minimum value at $z' = 4$ and the Cu-InSb has lesser value for f_l as compared to Ag-InSb and Al-InSb. This can be explained as that the laser incident on metal from glass side interacts with electrons to produce SPW. These are electromagnetic waves which propagate at the interface of different media. Since, amplitude of SPW depends on the amplitude of laser so it is expected that focusing of laser beam will have impact on focusing of SPW. Also, the reflectivity of Ag and Al is more than the Cu so fields of laser could interrelate better with the electrons of Cu leading to impactful SPW which results in focused SHG at the Cu-InSb interface. The electromagnetic fields of SPW interact nonlinearly with the electrons of n-InSb resulting in SHG. The focusing and de-focusing of SHG arise due to nonlinear interactions accompanied by a decrease in diffraction effects. The self-focusing action of laser beam enhances the intensity of SPW produced in metal which on further

interaction with electrons of InSb generates harmonic with amplitude efficiently enhanced in the region of high focusing. The beam width parameter of laser attains minimum value in the region of high focusing. Since SPW are also electromagnetic waves with frequency similar to laser, so it is expected that in the regions of focusing of laser SPW also gets focused. As the amplitude of SPW is generally higher than the laser, so it is expected that intensity of SPW further gets enhanced in the regions of high focusing which leads to efficient second harmonic generation.

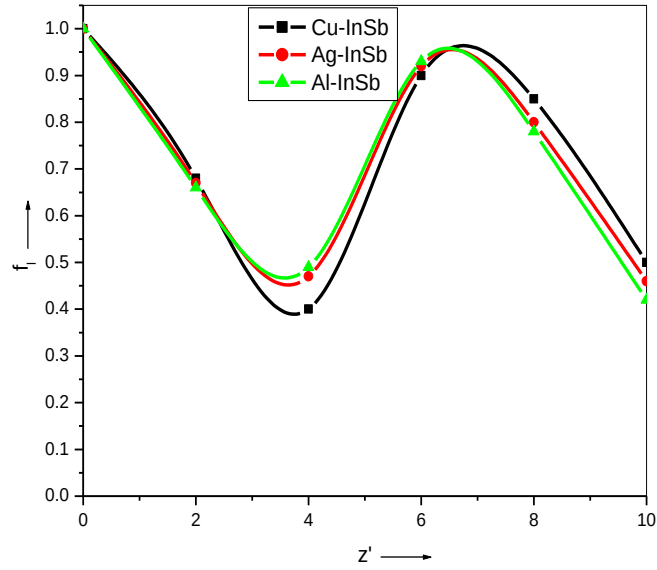


Fig.6.2

Fig.6.2. Plot for the variation of beam width parameter with normalized propagation distance for Cu, Ag and Al with n-InSb.

Eq.6.23 has also been solved graphically using Eq.6.2 with $\omega_p / \omega_l = 0.5$ and 0.8 , $j = 1$ and $eA_{l0} / mc\omega_l = 0.02$. One can observe from Fig.6.3 that the SHG gets focused and also shows significant enhancement in amplitude at $z'=4$. The enhancement in amplitude is more for the Cu-InSb interface as compared to Ag-InSb and Al-InSb. The increase in amplitude is more pronounced at higher values of plasma frequency. The self-focusing of laser has been studied by Thakur and Kant[27] in plasma under exponential density ramp in the presence of a wiggler field. Their investigation shows that the beam width parameter of pump laser shows periodic variation and attains minimum value at $z'=8$ where second harmonic amplitude show significant enhancement. The present investigation has been done for Cu-InSb, Ag-InSb and Al-

InSb interfaces with the rippled metal surface in the absence of the wiggler field. We have also observed that the beam width parameter of laser shows periodic variation but attains minimum value at $z'=4$, as shown in Fig.6.2. One can observe from Fig.6.3 that amplitude of SHG gives maxima at $z'=4$. Hence, the use of metal-semiconductor interface for SHG has reduced the propagation distance of efficient SHG. Our work put forwards the metal-semiconductor interface as a better choice for SHG. The harmonic generated by self-focused laser improves the efficiency of process and can be useful in designing better imaging techniques useful in medical sciences.

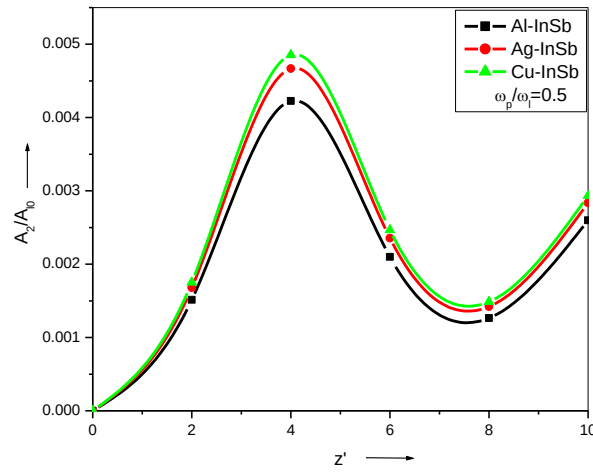


Fig.6.3(a)

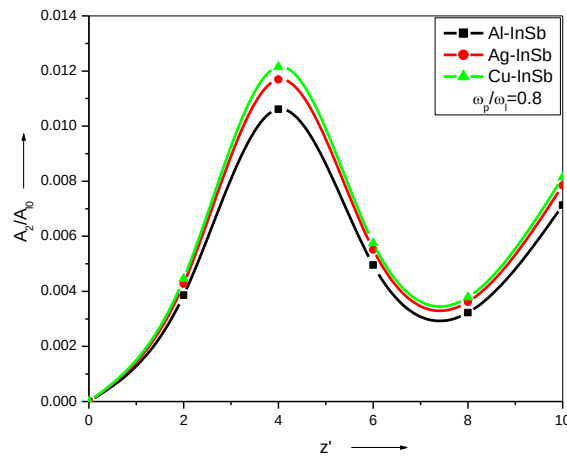


Fig.6.3(b)

Fig.6.3 Plot showing the variation of normalized second harmonic amplitude with normalized propagation distance at (a) $\omega_p / \omega_l = 0.5$ and (b) $\omega_p / \omega_l = 0.8$

6.5 Conclusion

The effect of self-focusing of laser on SHG induced by SPW on Cu-InSb, Ag-InSb and Al-InSb interfaces has been investigated. The metals considered are Cu, Ag, Al with n-InSb in Kretschmann ATR configuration. SPW produced by the interaction of laser with metal interacts nonlinearly with electrons of the semiconductor leading to SHG. Due to nonlinear effects, laser beam shows periodic focusing and defocusing but the enhancement in the amplitude of SHG is found to be more in the regions of minimum beam width parameter. Under resonance, the Cu-InSb interface gives the maximum increase in amplitude along with a focused beam. The present work shows that the metal-semiconductor interface could be a better choice for SHG and its applications.

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CHAPTER 7

SUMMARY AND FUTURE SCOPE

7.1 Summary

In the study presented in this thesis, the new methods to generate second harmonic generation have been given. The materials used are metal and semiconductor. The thin layers of metal and semiconductor are considered over glass prism in Kretschmann attenuated total reflection configuration. The study has been done in the presence of different factors like density ripple and wiggler magnetic field. The SHG by crossed surface plasma waves and effect of self-focusing of laser has also been studied. The laser is incident on metal-glass interface obliquely to experience maximum reflection but depending on the reflectivity of metal the electromagnetic fields of laser could interact with electrons of metal. The interactions are nonlinear in nature and could give rise to surface plasma waves. The SPW are electromagnetic waves with frequency nearly same as laser but amplitude is higher than the laser. The fields of SPW could influence electrons of n-type semiconductor and can lead to harmonic generation. The harmonic generated by this method efficient, focused and show good enhancement in amplitude. The present work suggests that the metal-semiconductor interface could be a better choice for studying harmonic generation. The efficient harmonic generated by this method can be used for imaging techniques, spectroscopy, as probes for studying structures etc. Different applications involving the use of harmonic generation could use the waves generated by the given method. The purpose of present work is to design an efficient and cost effective model for harmonic generation which could be used for different applications. The proposed method of harmonic generation open up new opportunities:-

7.1.1 Plasma Physics

The given model generates an efficient, focused and an intense SHG which could be used further for studying laser plasma interactions in different media. It gives a way to the new methods of harmonic generation using materials with different dielectric properties.

7.1.2 Medical Science

The harmonics generated by laser plasma interaction with different materials are now an integral part of medical sciences. The SHG produced by this method can be used for medical purposes like diagnosis of tumors, cancer treatments etc. The harmonic generated is resonant and has good propagation length. It can also serve as an amplified harmonic for bio-sensors.

7.1.3 Material Science

The applications of harmonic generation in phase transition studies and structural analysis can be done efficiently by the waves produced by this method.

7.2 Future Scope

In terms of future scope, the advent of SHG on metal-semiconductor interface induced by SPW is opening up new opportunities in nonlinear physics. One of the most profound features is that the second harmonic generated by this method can serve as a better option for its applications. Many researchers have worked on metal-vacuum interface in Kretschmann configuration as well as in different plasmas like collisional plasma, collisionless plasma, and clustered gas plasma but present study opens up new options for research in future. Present work can be used in medical sciences for new methods of imaging techniques. Present model opens new pathways for experimental research as well.

List of Publications

Papers published in SCI Journal

1. H. K. Dua, N. Kant, and V. Thakur, "Second Harmonic Generation Induced by a Surface Plasma Wave on a Metallic Surface in the Presence of a Wiggler Magnetic Field," *Braz. J. Phys.*, vol. 52, no. 2, pp. 1–6, 2022.
2. H. K. Dua, N. Kant, and V. Thakur, "Surface plasma wave induced second-harmonic generation on a metal–semiconductor interface: effect of self-focusing of a laser," *Appl. Opt.*, vol. 61, no. 16, pp. 4731–4734, 2022.

Papers accepted by SCI Journal

1. Second harmonic generation induced by crossed surface plasma waves at metal-semiconductor interface by Non linear Optics quantum optics

Papers communicated to SCI Journal

1. Frequency doubling by surface plasma waves at three metal-semiconductor interfaces in Iranian Journal of Physics

Papers published in UGC care listed Journal

1. H. K. Dua, "Enhancement in Second Harmonic Generated by ultra-fast Lasers in Plasma by wiggler magnetic field," *J. Gujarat Res. Soc.*, vol. 21, no. 8, pp. 1514–1519, 2019.
2. H. K. Dua and V. Thakur, "Harmonic generation in metals and semiconductors-A review," *JETIR* vol. 6, no.1, pp. 1-3, 2019.

Papers published in International Conferences

1. H. K. Dua, N. Kant, and V. Thakur, "Second Harmonic Generation by ultra-fast Lasers in Plasma.," in *Journal of Physics: Conference Series*, 2020, vol. 1531, no. 1, pp. 012008.
2. H. K. Dua, N. Kant, and V. Thakur, "Effect of wiggler magnetic field on harmonic generation at metallic surface," in *Journal of Physics: Conference Series*, 2022, vol. 2267, no. 1, pp. 012025.

List of Conferences

Sr. No	Name of Conference	Date	Paper presented
1	International Conference on “Recent Advances in Fundamental and Applied Sciences”	November 5-6,2019	Second Harmonic Generation By Ultra fast lasers in Plasma.
2	International Conference on “Recent Advances in Fundamental and Applied Sciences”	June 25-26,2021	Effect of Wiggler Magnetic Field on Harmonic Generation at metallic Surface
3	International Conference on “Recent Advances in Fundamental and Applied Sciences”	March 24-25,2023	Frequency doubling on a metallic surface by Hermite-Cosh Gaussian beam

List of Workshops

Sr. No	Name of the Workshop	Date
1	One week National Workshop on “Research Methodology” organized by GSSDGS Khalsa College Patiala.	22-27 February,2021
2	National Faculty Development Workshop on “Research Methodology” organized by GCG Ludhiana.	15-20 March,2021

COPY OF PUBLICATIONS



Second Harmonic Generation Induced by a Surface Plasma Wave on a Metallic Surface in the Presence of a Wiggler Magnetic Field

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Abstract

Under the influence of a wiggler magnetic field, the phenomenon of second harmonic generation at the metal–semiconductor interface, induced by a surface plasma wave (SPW), has been investigated. Metals like Cu, Ag, and Al, each with a thin layer of n-InSb over it, are considered for our study. Laser light is incident on metal layered on glass prism in attenuated total reflection Kretschmann configuration (ATR), which generates SPW. The SPW further interacts nonlinearly with the electrons of the n-type semiconductor which is layered over the metal, leading to second harmonic generation (SHG). The presence of an external wiggler magnetic field makes the process resonant and helps in phase matching. Relatively more enhancement in the amplitude of the second harmonic is observed for Cu-InSb as compared to Ag-InSb and Al-InSb. Numerical analysis shows that the enhancement in the amplitude of SHG increases with the intensity of the wiggler magnetic field.

Keywords Surface plasma waves · Wiggler field · Semiconductor · Second harmonic generation

1 Introduction

Surface plasma waves (SPW) are electromagnetic waves that are formed by the interaction of lasers with metals. These waves propagate along the interface of two different media and have amplitude larger than the amplitude of the laser [1, 2]. The nonlinear interaction of laser with the matter has gained much attention in the past few decades because of its unique applications like high harmonic generation, laser-driven plasma-based accelerators, sensors, photoelectron spectroscopy, etc. [3–12]. Harmonic generation on different materials and in the presence of external agents like the wiggler field has been studied by many researchers. Jha et al. [13] studied SHG with an intense laser beam in magnetized plasma and found high conversion efficiency in the presence of the transverse magnetic field. Rajput et al. [14] investigated the generation of third harmonic by a short pulse laser in plasma and observed that the presence of the wiggler magnetic field makes the generation process resonant accompanied by the increase in the efficiency of harmonic generation. Vinay et al. [15] investigated propagation of a laser

pulse through plasma and found that the laser interacts nonlinearly with electrons exerting a ponderomotive force which in turn makes them oscillates. It results in harmonic generation. Abedi-Varaki [16] investigated the acceleration of electrons of metal by SPW in the presence of a helical magnetostatic wiggler and observed that under the effect of both SPW and wiggler field, the electrons can travel more distance in the direction of propagation of the laser. Vij et al. [17] studied the production of terahertz (THz) radiation by carbon nanotubes in the presence of a wiggler magnetic field and observed that the presence of the wiggler field enhances the efficiency of the generation of THz radiation. Abedi-Varaki and Jafari [18] studied the generation of THz radiations by mixing two Cosh-Gaussian laser beams in the presence of a wiggler magnetic field and observed the increase in efficiency with the increase in wiggler frequency. Ghimire et al. [19] had investigated harmonic generation in solids and reported that terahertz or infrared fields are suitable for metals and semiconductors due to their small bandgap. The presence of external factors like the wiggler magnetic field helps to make the process resonant. Singh et al. [20] studied the acceleration of electrons by a laser pulse in the presence of a magnetic wiggler, in vacuum and plasma. Their work showed that the suitably tapered magnetic wiggler can maintain electron-laser resonance conditions for a longer time, and electrons can gain sufficient energy.

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In the present manuscript, second harmonic generation induced by surface plasma wave at the metal–semiconductor interface in the presence of a wiggler magnetic field has been studied. The phase matching condition is fulfilled by the application of the wiggler magnetic field. Here, in Sect. 2, we have obtained the expression for normalized amplitude for second harmonic generation. Results are discussed and presented in graphical form in Sect. 3, and the conclusion is presented in Sect. 4.

2 Theoretical Considerations

A thin metal layer over a glass prism with a thin layer of semiconductor has been considered in Kretschmann ATR configuration, as shown in Fig. 1, where laser light is incident at an acute angle on a metal–glass interface leading to SPW in the metal [21]. The EM fields of the laser interact with electrons of the metal and produce SPW of nearly the same frequency as that of the laser but with higher amplitude. The component of the wave vector of the laser along the surface matches with the wave number of surface plasma wave, leading to efficient coupling [22]. A thin layer of n-InSb with the thickness of a few nanometers is considered above the metal surface. The electromagnetic fields of SPW produced at the metal–semiconductor interface interact with electrons of n-InSb. The semiconductor can accumulate energetic electrons which can easily tunnel through plasmonic entities [23].

Electric and magnetic fields of laser are considered as $\vec{E} = \hat{x}A(z, t)e^{-i(\omega t - k_1 z)}$ and $\vec{B} = (\vec{k}_1 \times \vec{E})/\omega$ where A is the amplitude of the laser field, ω is the frequency of the laser, and k_1 is the propagation constant. These fields interact with electrons of metal and produces surface plasma waves $\vec{E}_s = \hat{x}A_s e^{-i(\omega_s t - k_s z)}$ where k_s is the propagation constant

such that $k_s = (\omega_s/c)(\epsilon_s \epsilon_m / (\epsilon_s + \epsilon_m))^{1/2}$ with ϵ_m and ϵ_s as the permittivity of metal and semiconductor, respectively, and ω_s is the frequency of the surface plasma wave. The frequency of SPW is nearly same as the frequency of the laser, so $\omega_s = \omega$ [16]. The amplitude of surface plasma wave A_s depends on the amplitude of incident laser A with $A_s = f(A)$. Following [24], A_s/A can be considered as a normalized quantity with $A_s = nA$, where n is a positive real number. Fields of surface plasma waves interact with electrons of the semiconductor and accelerate them. Acceleration so produced is calculated using the equation of motion:

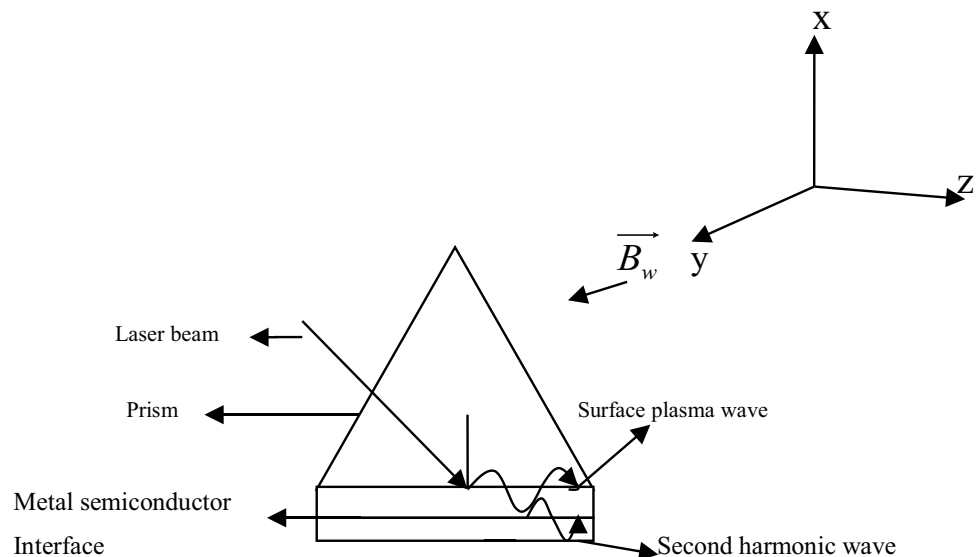
$$m \frac{d\vec{v}_1}{dt} = -e\vec{E}_s - e\vec{E} \quad (1)$$

The velocity gained by electrons is,

$$\vec{v}_1 = \left[\frac{-eAn' e^{-i(\omega t - k_s z)}}{m(-i\omega)} \right] \hat{x} \quad (2)$$

As the wave vector increases more than linearly with incident frequency, the momentum is not conserved, i.e., the momentum of second harmonic is more than two times the momentum of incident photon. Here, $n' = 1 + n$ which is a positive real number. Wiggler magnetic field is applied in transverse direction, given by $\vec{B}_w = B_0 e^{ik_0 z} \hat{y}$ where B_0 is the amplitude of wiggler field, and k_0 is the wiggler wave number [25]. The presence of wiggler field helps to achieve the phase matching condition by providing additional momentum to the photons of second harmonic. The velocity $\vec{v}_1$ beats with the wiggler magnetic field of the laser and generates ponderomotive force $\vec{F}$, where $\vec{F} = -e(\vec{v}_1 \times \vec{B}_w)$. Due to this force the velocity of electrons changes, and it is taken as $\vec{v}_1'$ such that

Fig. 1 Kretschmann configuration with thin layer of a metal and a semiconductor



$$\vec{v}_1 = \left[\frac{-e^2 n' A B_0 e^{-i(\omega t - (k_1 + k_0)z)}}{m^2 \omega^2} \right] \hat{z} \quad (3)$$

It will induce density perturbation at $(\omega_1, k_1 + k_0)$ in agreement with the equation of continuity:

$$n_1 = (\vec{k}_1 + \vec{k}_0) n_0 \vec{v}_1 / \omega_1 \quad (4)$$

To exert ponderomotive force on electrons at $(2\omega, 2k_1 + k_0)$, $\vec{v}_1$ beats with $\vec{B}_1$ and imparts velocity $\vec{v}_2^{NL}$ to electrons, given as

$$\vec{v}_2^{NL} = \left[\frac{-ie^3 n' A^2 k_1 B_0 e^{-i(2\omega t - (2k_1 + k_0)z)}}{2m^3 \omega^4} \right] \hat{x} \quad (5)$$

This gives rise to second harmonic nonlinear current density at metal-semiconductor interface,

$$\vec{J}_2^{NL} = -n_0 e \vec{v}_2^{NL} - n_1 e \vec{v}_1 \quad (6)$$

The linear current density due to self-consistent second harmonic field $\vec{E}_2 = A_2 e^{-i(2\omega t - k_2 z)} \hat{x}$ at $(2\omega, k_2)$ is $\vec{J}_2^L = n_0 e \vec{v}_2$ where $\vec{v}_2$ is obtained using equation of motion $m(d\vec{v}_2/dt) = -e\vec{E}_2$.

The amplitude A_2 for second harmonic is calculated by using equation:

$$\nabla^2 \vec{E}_2 = \frac{4\pi(\epsilon_m + \epsilon_s)\partial \vec{J}}{c^2(\epsilon_m \epsilon_s)\partial t} + \frac{\partial^2 \vec{E}_2}{c^2 \partial t^2} \quad (7)$$

We take the laser profile to be Gaussian as $A^2 = [F(z - v_{g1}t)]^2$ and $F(z - v_{g1}t) = A_0 e^{-\left(\frac{z - v_{g1}t}{v_{g1}\tau}\right)^2}$ with $v_{g1} = c\sqrt{1 - (\omega_p^2/\omega^2)}$, $v_{g2} = k_2(c^2/2\omega)\sqrt{1 - (1/4)(\omega_p^2/\omega^2)(\epsilon_m + \epsilon_s)/\epsilon_m \epsilon_s}$, and τ is laser pulse width. Using a new set of variables $z - v_{g1}t = \Omega z = \eta, \Omega_0 = \tau v_{g1}, \beta' = (1 - v_{g1}/v_{g2}), z' = z\Omega_0$, and $t' = v_{g1}t/\Omega_0$, we get normalized second harmonic amplitude as:

$$\left| \frac{A_2}{A_0} \right| = \left\{ \left(1 - \frac{\omega_p^2}{\omega^2} \right)^{1/2} \left(\frac{a \omega_p^2}{\omega^2} \right) \left(\frac{eB_0}{m\omega} \right) \left(\frac{eA_0}{mc\omega} \right) \right. \\ \left. (d + b) [Erf(z' - t') - Erf\{(1 - \beta')z' - t'\}] \right\} \quad (8)$$

where $a = \frac{\epsilon_m + \epsilon_s}{\epsilon_m \epsilon_s}$, $d = \frac{k_1 \Omega_0 \sqrt{\pi} n'}{8\beta'}$ and $b = \frac{(k_1 + k_0) \Omega_0 \sqrt{\pi} n'^2}{4\beta'}$.

3 Results and Discussion

Equation (8) has been solved numerically for a Nd:YAG laser with wavelength 1064 nm and intensity $I \approx 10^{12}$ W/cm². The refractive index of the prism lies in the range 1.5 – 2. The dielectric permittivity is taken as 57.909 for Ag, 49.58 for Cu, 106.82 for Al, and 19.5 for InSb at 1064 nm [26–28].

Here, the layers of metal and semiconductor are considered to be of few nanometers, and the doping level of n-InSb is adjusted to have $\omega_p/\omega < 1$ for the semiconductor. The graphical relationship for normalized second harmonic amplitude with normalized propagation distance has been presented for different metal–semiconductor interfaces at different values of time and wiggler field. The other parameters of Eq. (8) are taken as $\omega_p/\omega_1 = 0.9$, $n' = 2.5$, $eA_0/mc\omega_1 = 0.09$, $(k_1 + k_0)\Omega_0\sqrt{\pi}/8\beta' = 500$, $eB_0/m\omega = 0.006, 0.012$ and 0.018 , $k_1\Omega_0\sqrt{\pi}/8\beta' = 300$, and $\beta' = 0.3$.

Figure 2 shows the variation of normalized second harmonic amplitude A_2/A_0 with normalized propagation distance z' for metals like Cu, Ag, and Al with thin layer of n-InSb, at time $t' = 4$, in the presence of a wiggler magnetic field. It has been observed that as the strength of magnetic field increases, the amplitude of second harmonic increases. In Fig. 2a, a sharp increase in amplitude has been observed at $z' = 5$ for all three metal–semiconductor interfaces at $eB_0/m\omega = 0.006$, with larger increase in amplitude in the case of the Cu-InSb interface. The presence of the wiggler field helps to attain phase-matching conditions by providing additional momentum to the second harmonic photon. The electromagnetic fields of laser interact with electrons of metal and excite them. The excited electrons beat with the laser magnetic field and the wiggler field, leading to resonant SPW. These waves with high amplitude interact nonlinearly with electrons of n-InSb, leading to second harmonic. Figure 2b and c show the variation of normalized second harmonic amplitude A_2/A_0 with normalized propagation distance z' for all three metal–semiconductor interfaces, at $eB_0/m\omega = 0.012$ and $eB_0/m\omega = 0.018$, respectively. The values of other parameters for Fig. 2b and c are the same as for Fig. 2a. As observed graphically under given conditions, increase in amplitude of SHG has been found more significant for Cu-InSb than for Ag-InSb and Al-InSb. The reason for this observation can be understood as the value of the density of conduction electrons in Cu is larger than in Ag and Al, therefore the fields of SPW in Cu are expected to be comparatively stronger and can efficiently interact with electrons of n-type semiconductor. As per the study by Takagi et al. [27], surface plasma waves in Kretschmann geometry show better agreement with bulk dielectric function for Cu than for Ag. Similarly, in our work, we have seen better enhancement in the amplitude of second harmonic for Cu. Hashemzadeh et al. [29] studied SHG of hollow Gaussian beam in inhomogeneous plasma and found the increase in amplitude of SHG with the increase in wiggler field. Sharma et al. [30] studied SHG of cosh-Gaussian laser beam in magnetized plasma and found enhancement in efficiency in the presence of wiggler field. Similarly, in the present work, good enhancement in amplitude of second harmonic has been observed in the presence of wiggler field.

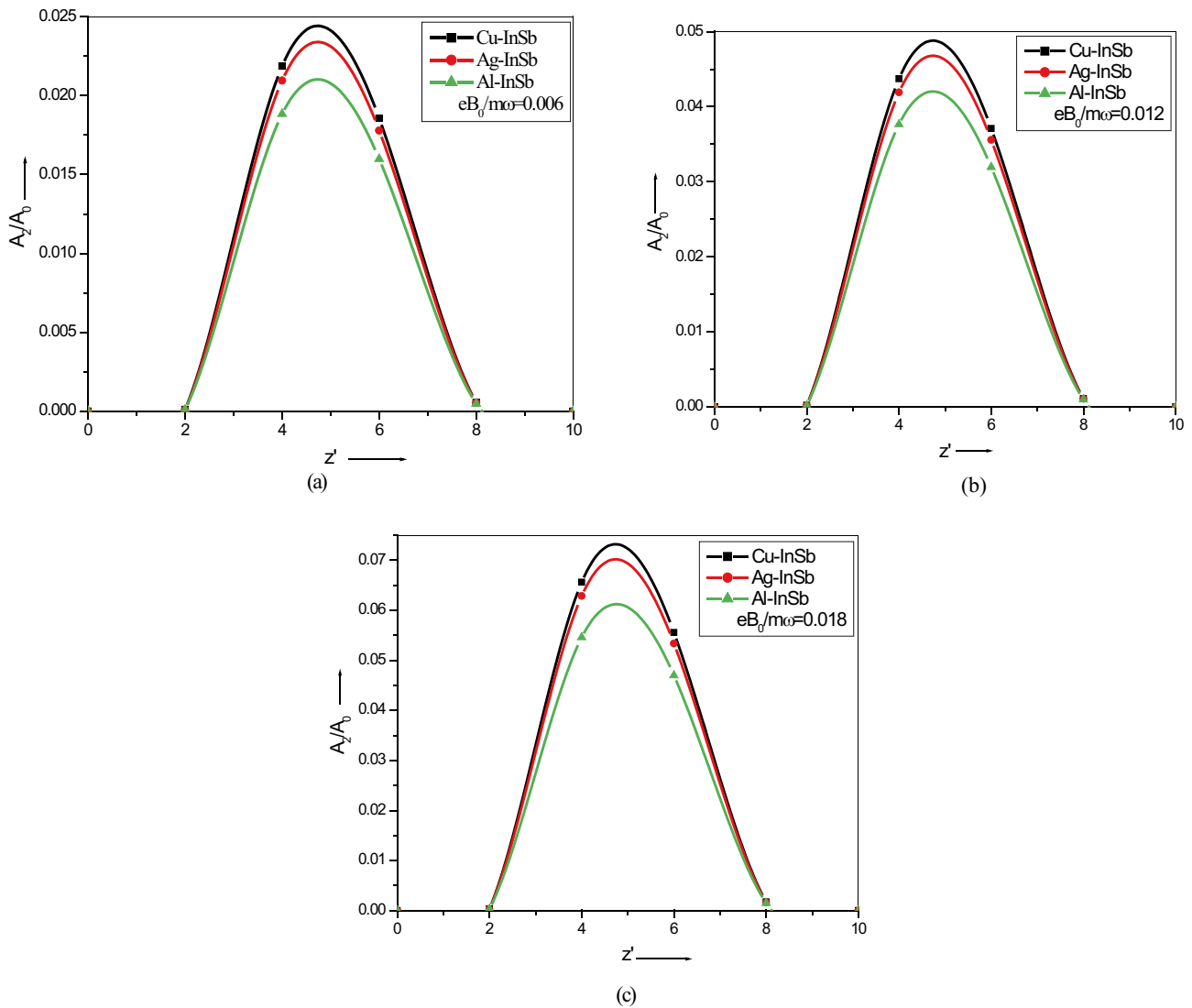
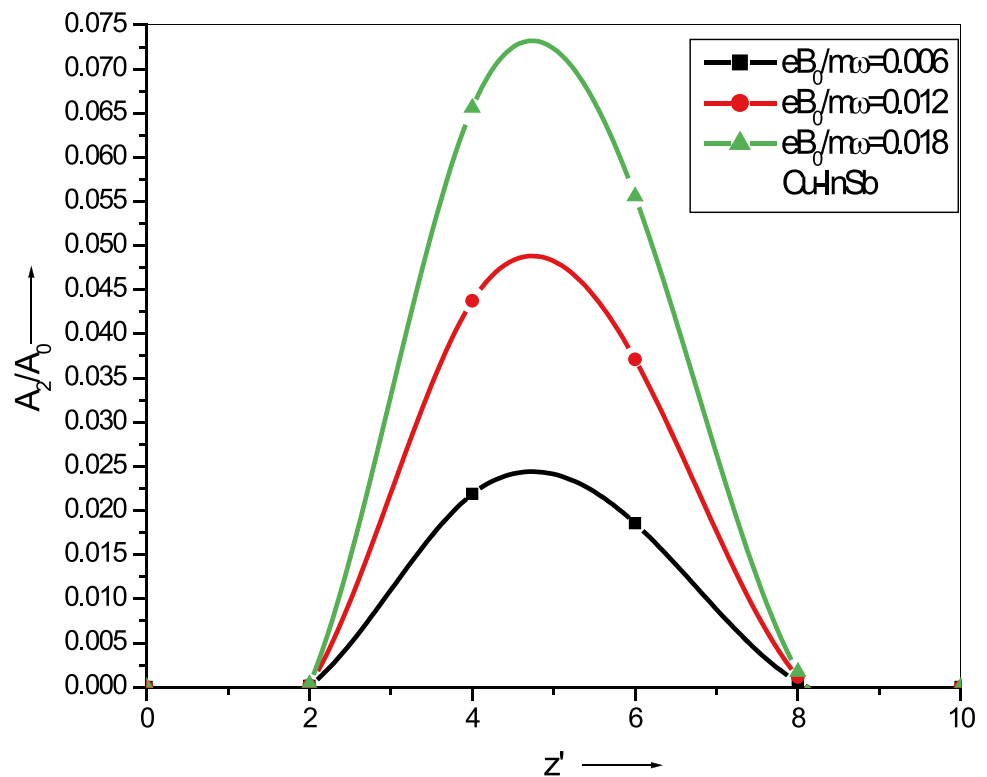


Fig. 2 Variation of normalized second harmonic amplitude A_2 / A_0 with normalized propagation distance z' , for metals Cu, Ag, and Al with layer of n-InSb, for different values of wiggler field $eB_0/m\omega$ at time $t' = 4$

Figure 3 shows the variation of the normalized amplitude of second harmonic, A_2 / A_0 , with normalized propagation distance z' , at different values of wiggler field with $eB_0/m\omega = 0.006, 0.012$ and 0.018 , for Cu-InSb interface. The values of other parameters have been taken the same as those in Fig. 2. It is clear from Fig. 3 that with a slight increase in wiggler field strength, the amplitude of second harmonic enhances rapidly. Aggarwal et al. [31] studied second harmonic generation in atomic clusters in the presence of wiggler magnetic field, and they found increase in amplitude of second harmonic with

the wiggler field strength. In their work the amplitude of second harmonic shows enhancement in the region where $13 < z' < 16$, for different values of wiggler frequency. In the present work, we have used the same values for wiggler frequency and have observed sharp increase in amplitude at $z' = 5$, for the Cu-InSb interface. Figure 2 reveals that the enhancement in amplitude is larger for Cu-InSb, as compared to Ag-InSb and Al-InSb interfaces. Our work shows that a metallic surface like the Cu-InSb interface would be a better choice to study the second harmonic generation and its applications.

Fig. 3 Variation of normalized second harmonic amplitude A_2/A_0 with normalized propagation distance z' , for Cu-InSb interface, for different values of wiggler field at time $t' = 4$



4 Conclusion

The electromagnetic fields of laser interact with electrons of the metal and generate SPW, which are electromagnetic waves with higher amplitude. The SPW of different metals like Cu, Ag, and Al interact nonlinearly with electrons of n-InSb, leading to second harmonic generation. The interaction has been studied in the presence of a wiggler field with different amplitudes. It has been observed that the amplitude increases sharply at the Cu-InSb interface, as compared to Ag-InSb and Al-InSb. The enhancement in second harmonic amplitude has been observed with the increase in wiggler field strength. The presence of the wiggler field helps in phase matching and resonates with the process of second harmonic generation.

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Surface plasma wave induced second-harmonic generation on a metal–semiconductor interface: effect of self-focusing of a laser

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The effect of the self-focusing of a laser on second-harmonic generation (SHG) induced by the surface plasma waves (SPW) over the metal–semiconductor interface has been investigated. The nonlinear interactions of a laser with metal generate SPW. These high-amplitude waves could penetrate through plasmonic entities of metal and can interact with electrons of a semiconductor leading to SHG. The behavior of the beam width parameter for different metals has been analyzed. One may notice that the beam width parameter shows periodic variation with the distance, but the amplitude shows maxima in the regions of the low beam width parameter. The focusing accompanied by the increase in SHG amplitude is found to be better for the Cu–InSb interface as compared to the Ag–InSb and Al–InSb ones. The present work puts forward the metal–semiconductor interface as a good choice for SHG. © 2022 Optica Publishing Group

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1. INTRODUCTION

The advancement in laser technology has led to various non-linear phenomena, out of which laser–plasma interaction has gained much importance in the past few decades. The formation of surface plasma waves (SPW) on the surface of a metal due to interaction between laser and electrons has gained many applications in the field of biology, technology, and industry [1–9]. These waves are guided electromagnetic waves propagating along the interface of a metal and a dielectric. They decay more rapidly in metals as compared to the dielectric. The nonlinear interactions between laser fields and electrons result in the self-focusing of the wave, which has been studied by many researchers and found that the self-focusing of laser helps to improve the efficiency of harmonic generation. Kumar [10] has studied self-focusing of SPW using the ponderomotive mechanism and has observed that the axial portion of SPW has a smaller phase velocity than the non-axial phase, resulting in self-focusing. Thakur and Kant [11–13] investigated second-harmonic generation (SHG) in plasma under exponential density ramp and revealed that self-focusing of pump laser leads to efficient SHG. The presence of an exponential density ramp strengthens the self-focusing of the laser which in turn enhances the SHG in plasma. As per the work of Singh *et al.* [14], the application of density ripple in plasma gives resonant SHG. The density ripple couples with the laser beam and helps in phase matching, which increases the efficiency of SHG. Niknam *et al.* [15] studied the self-focusing properties of an intense electromagnetic beam in underdense plasma and found

that the presence of magnetic field and increase in the collision frequency supports self-focusing of the laser. Parashar [16] studied the self-focusing effect of laser on the third-harmonic generation in clustered gas and found that the efficiency of harmonic generation is sensitive to the focusing of the pump laser. Sharma *et al.* [17] examined the influence of self-focusing on the third-harmonic generation using an intense Gaussian laser. They used the ponderomotive mechanism and found that the efficiency of harmonic generation is more in the regions where self-focusing dominates. Hashemzadeh [18] investigated the self-focusing characteristics of Gaussian laser beam propagating in collisional plasma by considering the ponderomotive non-linearity and inhomogeneities in density and temperature. The work by Nanda *et al.* [19] concluded that the combined role of magnetic field and density ramp reduces the spot size of the laser beam and leads to efficient self-focusing.

The parameters like density profile and intensity play a significant role in the self-focusing of the laser beam. Self-focusing of an intense laser beam is due to relativistic mass and ponderomotive effects. The increase in plasma density leads to stronger self-focusing and the beam width parameter attains a minimum value [20,21]. Wani and Kant [22] investigated relativistic self-focusing of Hermite–cosine–Gaussian laser beam in collisionless plasma under Wentzel–Kramers–Brillouin and paraxial approximations. Their work reveals that lower values of intensity and decentered parameter results in a more focused beam. The effect of self-focusing of laser with different laser profiles like Cosh–Gaussian, Laguerre–Gaussian, Hermite–Gaussian,

etc., under different conditions of plasma have also been studied. In general, it has been observed that the self-focused laser beam leads to efficient harmonic generation [23–26].

Here, we investigate the effect of the self-focusing of laser on SHG induced by SPW on the metal–semiconductor interface. The nonlinear interactions of electromagnetic fields of SPW with the electrons of n-type semiconductor lead to SHG. These interactions are governed by the ponderomotive mechanism. The presence of nonlinearity accounts for the self-focusing of the laser beam which in turn influences the efficiency of SHG. Section 2 includes theoretical considerations, and results and discussion are included in Section 3. The conclusion is presented in Section 4.

2. THEORETICAL CONSIDERATIONS

We considered the metal–semiconductor interface in the Kretschmann attenuated total reflection (ATR) configuration as shown in Fig. 1, with nanometer thick layers of metal and semiconductor. Metals considered for analysis are Cu, Ag, and Al with semiconductor n-InSb. Light incident on the glass–metal interface produces SPW on a metal surface, which is rippled to achieve the phase-matching condition for a resonant harmonic generation. The electromagnetic fields of SPW can penetrate through the plasmonic entities of semiconductors and can interact with the electrons.

Laser with Gaussian profile is considered with electromagnetic fields as

$$\vec{E}_l = \hat{x} A_l e^{-i(\omega_l t - k_l z)} \quad \text{and} \quad \vec{B}_l = (\vec{k}_l \times \vec{E}_l) / \omega_l, \quad (1)$$

where ω_l and k_l are frequency and propagation constant of the laser, respectively, and A_l is the amplitude of the electric field of the laser with

$$A_l = (A_{l0} / f_l(z)) e^{-\frac{r^2}{2f_l^2}}, \quad (2)$$

where A_{l0} is constant amplitude of the laser pulse, f_l is the beam width parameter of the laser pulse, and r_0 is the spot size of the laser at $z = 0$. These fields interact with electrons of metal and produces surface plasma waves with electric field $\vec{E}_s = \hat{x} A_s e^{-i(\omega t - k_s z)}$, where k_s is the propagation constant of SPW such that $k_s = (\omega/c)(\epsilon_s \epsilon_m / (\epsilon_s + \epsilon_m))^{1/2}$ with ϵ_m and ϵ_s as the permittivity of metal and semiconductor, respectively, and ω is the frequency of surface plasma wave. Amplitude of surface plasma wave A_s depends on the amplitude of incident laser A_l , as $A_s = f(A_l)$ and A_s/A_l can be considered as a normalized parameter with $A_s = j A_l$, where j is a positive

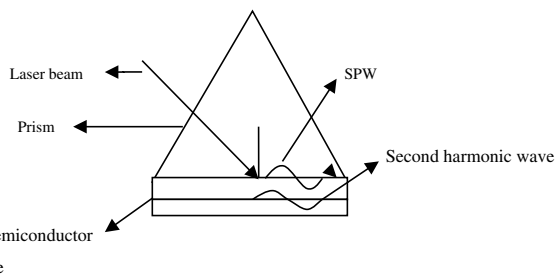


Fig. 1. Kretschmann configuration with thin layer of a metal and a semiconductor.

real number. Also, the frequency of SPW is the same as the frequency of the laser, so $\omega_l = \omega$ [27]. The electrons of semiconductor get accelerated due to the electric fields of SPW, and their velocity can be calculated using equation of motion $m(d\vec{v}_1/dt) = -e\vec{E}_s - e\vec{E}_l$, and it comes out to be

$$\begin{aligned} \vec{v}_1 &= \left[\frac{-e A_s e^{-i(\omega t - k_s z)}}{m(-i\omega)} - \frac{-e A_l e^{-i(\omega_l t - k_l z)}}{m(-i\omega_l)} \right] \hat{x} \\ \vec{v}_1 &= \left[\frac{-e j A_l e^{-i(\omega t - k_s z)}}{m(-i\omega)} - \frac{-e A_l e^{-i(\omega_l t - k_l z)}}{m(-i\omega_l)} \right] \hat{x}. \end{aligned} \quad (3)$$

The magnetic field of laser beats with the velocity of electrons and exerts ponderomotive force $\vec{F} = -e(\vec{v}_1 \times \vec{B}_l)$, which imparts an oscillatory velocity to electrons as

$$\vec{v} = -\frac{e^2 k_l}{m^2} \left[\frac{j A_l^2 e^{-i[(2\omega_l)t - (k_s + k_l)z]}}{2\omega_l^3} + \frac{A_l^2 e^{-i(2\omega_l t - 2k_l z)}}{2\omega_l^3} \right] \hat{z}. \quad (4)$$

Metal surface is rippled with density profile $n_q = n_0 e^{iqz}$, where n_0 is the equilibrium electron density, q is the ripple wave number, and z is the propagation distance. The nonlinear velocity $\vec{v}$ couples with density ripple and produces second-harmonic current given by $\vec{J}_{NL} = n_0 e^{iqz} e \vec{v}$ at $(2\omega_l, 2k_l + q)$. Here, q is chosen such that $k_2 = 2k_l + q$, and it obeys the phase-matching condition. The field equation for the second harmonic is taken as

$$\vec{E}_2 = A_2 e^{-i(2\omega_l t - k_2 z)} \hat{x}, \quad (5)$$

where A_2 is the amplitude of the SHG. The linear current density due to self-consistent second-harmonic field $\vec{E}_2$ at $(2\omega_l, k_2)$ is $\vec{J}_{2L} = n_0 e \vec{v}_2$, where $\vec{v}_2$ is obtained using equation of motion $m(d\vec{v}_2/dt) = -e\vec{E}_2$. So, current density can be calculated by taking

$$\vec{J} = \vec{J}_{NL} + \vec{J}_{2L}. \quad (6)$$

The wave equation governing the propagation of laser beam can be taken as

$$\frac{\partial^2 \vec{E}_l}{\partial z^2} + \frac{\partial^2 \vec{E}_l}{\partial r^2} + \frac{\partial \vec{E}_l}{r \partial r} + \frac{\omega^2 (\epsilon_m + \epsilon_s) \vec{E}_l}{c^2 (\epsilon_m \epsilon_s)} = 0. \quad (7)$$

Equation (7) has been solved numerically to study the variation of beam width parameter with propagation distance using Eqs. (1) and (2). The relation of beam width parameter with normalized propagation distance is obtained as

$$\begin{aligned} \frac{d^2 f_l}{dz'^2} \left(\frac{2r^2}{r_0^2 f_l^4} + \frac{-1}{f_l^2} \right) + \left(\frac{df_l}{dz'} \right)^2 \left(\frac{2}{f_l^3} + \frac{4r^4}{r_0^4 f_l^7} - \frac{10r^2}{r_0^2 f_l^5} \right) \\ = \frac{k_l^2 \xi_0^2}{f_l} + \frac{4r^2 \xi_0^2}{f_l^5 r_0^4} - \frac{4\xi_0^2}{r_0^2 f_l^3} - \left(\frac{\epsilon_m + \epsilon_s}{\epsilon_m \epsilon_s} \right) \frac{\xi_0^2 \omega_l^2}{c^2 f_l}, \end{aligned} \quad (8)$$

where $z' = z/\xi_0$ and ξ_0 is a normalizing parameter. The terms on the left-hand side (L.H.S) of Eq. (8) signifies the effect of the ponderomotive mechanism. The first three terms on the right-hand side (R.H.S) give the impact of laser propagation on laser

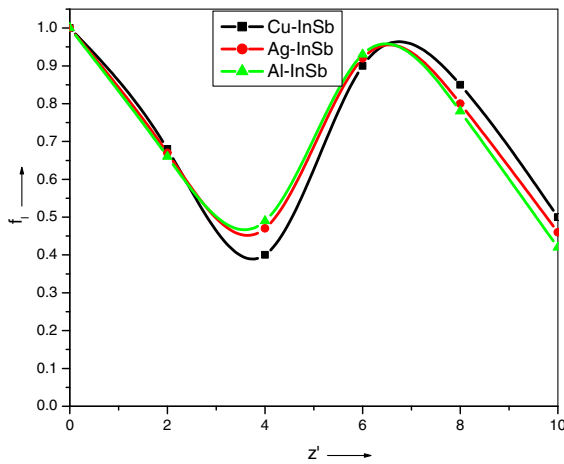


Fig. 2. Variation of beam width parameter with normalized propagation distance for Cu, Ag, and Al with n-InSb.

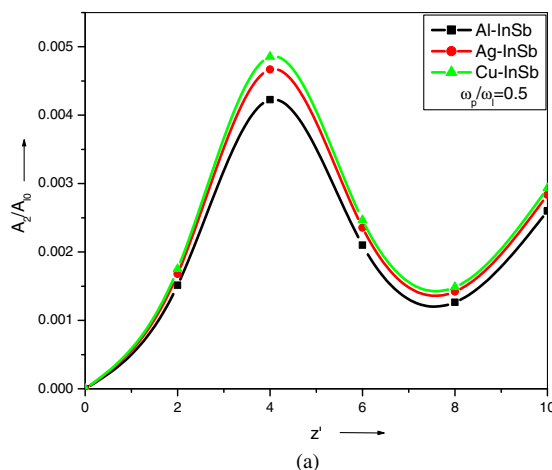
spot size and beam width parameter. The last term on the R.H.S includes the effect of material on the beam width parameter. The amplitude of second harmonic generated can be calculated by using the wave equation obtained by using Maxwell's relations

$$\nabla^2 \vec{E}_2 = \frac{4\pi(\epsilon_m + \epsilon_s)}{c^2(\epsilon_m \epsilon_s)} \frac{\partial \vec{J}}{\partial t} + \frac{\partial^2 \vec{E}_2}{c^2 \partial t^2}. \quad (9)$$

Using Eqs. (5) and (6) in Eq. (9), the relation for normalized second-harmonic amplitude with normalized propagation distance is obtained as

$$\frac{\partial^2(A_2)}{\partial z'^2} + \frac{\omega_l^2 \xi_0^2}{c^2}(A_2) + \frac{\omega_p^2 \xi_0^2(\epsilon_m + \epsilon_s)}{c^2(\epsilon_m \epsilon_s)}(A_2) + \left(\frac{\omega_p^2}{\omega_l^2}\right) \left(\frac{\epsilon_m + \epsilon_s}{\epsilon_m \epsilon_s}\right) \left(\frac{e A_l^2}{m c \omega_l}\right) \left(\frac{k_1 \xi_0^2 \omega_l}{c}\right) (1 + j) = 0, \quad (10)$$

where variables used are $A_l = (A_{l0}/f_l(z))e^{\frac{-r^2}{2f_l^2}}$, $\omega_p^2 = 4\pi n_0 e^2/m$, and $z' = z/\xi_0$.



3. RESULTS AND DISCUSSION

Equations (8) and (10) have been solved numerically for Nd:YAG laser having frequency $2.8 \times 10^{14} \text{ s}^{-1}$ and intensity $I \approx 10^{12} \text{ W/cm}^2$. The values for the permittivity of the metals and the semiconductor are taken as $\epsilon_m = 49.58$, for Cu, 57.909 for Ag, 106.82 for Al and $\epsilon_s = 19.5$ for InSb [28–30]. The doping of n-InSb is adjusted such that it acts like an underdense plasma for the electromagnetic SPW. Equation (8) has been analyzed graphically to find the relationship between beam width parameter f_l and normalized propagation distance z' with other parameters taken as $\xi_0/r_0 = 0.002$, $k_l \xi_0 = 0.5$, $\omega_l \xi_0/c = 0.8$, $\omega_p \xi_0/c = 0.5$ and $r/r_0 = 0.3$. Figure 2 reveals that the beam width parameter attains minimum value at $z' = 4$ and the Cu–InSb has lesser value for f_l as compared to Ag–InSb and Al–InSb. This can be explained as that the laser incident on metal from the glass side interacts with electrons to produce SPW. These are electromagnetic waves, which propagate at the interface of different media. Since the amplitude of SPW depends on the amplitude of the laser [31], it is expected that the focusing of the laser beam will have an impact on focusing of SPW. Also, the reflectivity of Ag and Al is more than the Cu, so fields of lasers could interrelate better with the electrons of Cu, leading to impactful SPW, which results in focused SHG at the Cu–InSb interface. The electromagnetic fields of SPW interact nonlinearly with the electrons of n-InSb, resulting in SHG. The focusing and defocusing of SHG arise due to nonlinear interactions accompanied by a decrease in diffraction effects.

Equation (10) has also been solved graphically using Eq. (2) with $\omega_p/\omega_l = 0.5$ and 0.8 , $j = 1$, and $e A_{l0}/m c \omega_l = 0.02$. One can observe from Fig. 3 that the SHG gets focused and also shows significant enhancement in amplitude at $z' = 4$. The enhancement in amplitude is more for the Cu–InSb interface as compared to Ag–InSb and Al–InSb. The increase in amplitude is more pronounced at higher values of plasma frequency. The self-focusing of laser has been studied by Thakur and Kant [32] in plasma under the exponential density ramp in the presence of a wiggler field. Their investigation shows that the beam width parameter of the pump laser shows periodic variation and attains minimum value at $z' = 8$, where second-harmonic amplitude show significant enhancement. The present investigation has

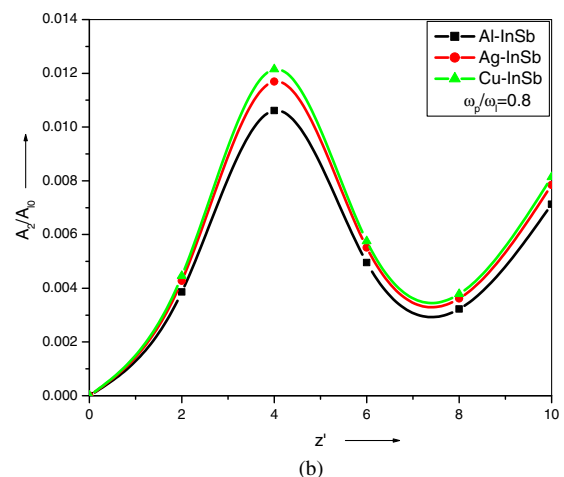


Fig. 3. Variation of normalized second-harmonic amplitude with normalized propagation distance at (a) $\omega_p/\omega_l = 0.5$ and (b) $\omega_p/\omega_l = 0.8$.

been done for Cu–InSb, Ag–InSb, and Al–InSb interfaces with the rippled metal surface in the absence of the wiggler field. We have also observed that the beam width parameter of the laser shows periodic variation but attains minimum value at $z' = 4$, as shown in Fig. 2. One can observe from Fig. 3 that amplitude of SHG gives maxima at $z' = 4$. Hence, the use of a metal–semiconductor interface for SHG has reduced the propagation distance of efficient SHG. Our work puts forward the metal–semiconductor interface as a better choice for SHG.

4. CONCLUSION

The effect of self-focusing of laser on SHG induced by SPW on Cu–InSb, Ag–InSb, and Al–InSb interfaces has been investigated. The metals considered are Cu, Ag, Al with n-InSb in a Kretschmann ATR configuration. SPW produced by the interaction of laser with metal interacts nonlinearly with electrons of the semiconductor leading to SHG. Due to nonlinear effects, a laser beam shows periodic focusing and defocusing, but the enhancement in the amplitude of SHG is found to be more in the regions of minimum beam width parameter. Under resonance, the Cu–InSb interface gives the maximum increase in amplitude along with a focused beam. The present work shows that the metal–semiconductor interface could be a better choice for SHG and its applications.

Disclosures. The authors declare no conflict of interests.

Data availability. The data underlying the results presented in this manuscript may be obtained from the authors after making a reasonable request.

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Second Harmonic Generation by ultra-fast Lasers in Plasma.

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Abstract

The interaction of laser with plasma gives rise to several novel relativistic nonlinear optical effects. Higher harmonic generation is an important nonlinear effect. High power ultrafast lasers uses chirped pulse amplification technique to reduce duration of pulse and hence produces pulses of extremely high power densities which in turn oscillates electrons in the medium. The velocity of oscillating electron beats with applied frequency giving rise to higher order harmonics. This paper presents the idea of generation of second harmonic waves with the ultra-fast lasers in plasma , phase matching conditions of second harmonic generated by self modulation under paraxial approximations and its application in imaging microscopy.

Key words : Ultra fast Lasers , Pulse amplification .

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1.Introduction

Plasma which is an ionic form of matter interacts with laser to give rise to different phenomenons. Introduction of Laser in plasma oscillates electrons and oscillating frequencies beats with laser frequency to give rise to different harmonics like second ,third harmonics etc. . The electrons recolliding with the parents ions are responsible for higher harmonic generation[1]. Permanent changes in the properties of materials have also been observed due to laser's interactions with materials.[2] In the presence of laser field , the non linearity of the medium enables the incident field frequencies to interact with each other in medium and produces fields with different frequencies.[3] High intensity short pulse lasers leads to minimum deposition of thermal energy in materials and give more precise outputs. Pulse energy is one of the factors that decide size of a laser.Reduction in size leads to production of high densities. Frequency mixing of ultra fast pulse laser's fundamental and second harmonic fields generates electric current whose magnitude and directions depends upon phase between two fields. The time scale of current obtained depends on the duration of laser pulse. For ultra fast laser terahertz frequencies can be generated.[4] The presence of wiggler magnetic field also enhances the amplitude of second harmonic.[5] SHG can be used to obtain highly sensitive and high resolution information of the collagen in the body. SHG microscopy on collagen fibers is done to examine arterial fibrillation.[6]

The technique of amplification of short pulse involving chirping and stretching of pulses to be amplified to saturated value then restoring to original width by using optical compressor . The intensity within the amplifier has to stay below a critical level at which non linear effects become significant.[7] Femtosecond fibre CPA systems like Ytterbium doped fibre chirp CPA system delivers as short as 800fs pulses with mJ



level pulse energies at average power of more than 100W at repetition rates of 200 KHz . A stretcher compressor unit consisting of dielectric diffraction gratings produces pulse with least distortions.[8] The other technique called Optical Parametric Chirped Pulse Amplification OPCPA can generate ultra fast laser beams by employing periodically poled crystals as amplifying media which produces phase controlled optical cycle pulses.[9] Self phase modulation along with self-focusing effects arises due to change in refractive index of material with laser intensity.[10] Harmonic generation and electron acceleration by lasers in plasma have also been studied under different parameters like chirp pulse, magnetic field giving similar conclusions.[11-15] The interaction of ultra fast lasers with high intense beams in plasma leads to better self focusing of beam which in turn leads to increase in oscillating frequency of electrons. This interaction generates second harmonic waves with high intensity as well as higher harmonics but with low intensities. The dispersion and non linear effects can be reduced by using thin crystals. The reduction in dispersive effects leads to the condition $n(2\omega) = n(\omega)$, which implies $k(2\omega) = k(\omega)$ and $\lambda_\omega = 2\lambda_{2\omega}$. If phase mismatch occurs it SH generated may go out of phase.[16]

In this paper section 2 contains derivation for amplitude of second harmonic in normalized form , section 3 contains results and discussions, section 4 explains its application in imaging microscopy and conclusion is presented in section 5.

2.Theoretical considerations

Consider the short pulse laser through n type semiconductor in the presence of wiggler magnetic field. The electromagnetic waves can be represented as

$$\vec{E}_1 = \hat{x}A_1(z,t)e^{-i(\omega_1 t - k_1 z)}, \quad (1)$$

$$\vec{B}_1 = \frac{\vec{k}_1 \times \vec{E}_1}{\omega_1}, \quad (2)$$

$$\vec{B}_w = \hat{y}B_0 e^{ik_o z} \quad (3)$$

B_w is Transverse wiggler magnetic field , A_1 is the amplitude of fundamental laser pulse Laser pulse interacts with semiconductor non linearly and produce second harmonic. Its electric vector can be expressed as

$$\vec{E}_2 = \hat{x}A_2(z,t)e^{-i(\omega_2 t - k_2 z)}, \quad \text{where } \omega_2 = 2\omega_1 \quad (4)$$

Linear dispersion relation followed by fundamental and 2nd harmonic is

$$k^2 = \frac{\omega^2}{c^2} \left(\epsilon_L - \frac{\omega_p^2}{\omega^2} \right) \quad (5)$$

where ϵ_L is lattice permittivity .

and $\omega_p^2 = 4\pi n_o e^2 / m$ is called plasma frequency.

Here $k_2 > 2k_1$, the difference is delivered to second harmonic by wiggler magnetic field when

$$k_2 = 2k_1 + k_o$$

$$k_o = k_2 - 2k_1$$

$$k_o = \frac{3\omega_p^2}{4\omega_1 c \sqrt{\epsilon_L}}$$
(6)

Using eq (1) and (2)

$$\text{velocity } v_1 = \frac{eE_1 x}{mi(\omega_1 + i\nu)}$$
(7)

It beats with magnetic field of eq (3) to exert force F_1 and velocity v_1' is produced at $(\omega_1, k_1 + k_o)$

$$v_1' = \frac{-e^2 A_1 B_o e^{-i(\omega_1 t - (k_1 + k_o)z)}}{2m^2 \omega_1 (\omega_1 + i\nu)} x$$
(8)

Now this velocity beats with magnetic field of eq (2) to give second harmonic and provides oscillatory

velocity v_2 at $(2\omega_1, 2k_1 + k_o)$

$$v_2 = \frac{-e^3 A_1^2 B_o k_1 e^{-i(2\omega_1 t - (2k_1 + k_o)z)}}{16m^3 \omega_1^3 (\omega_1 + i\nu)} x$$
(9)

Now velocity in eq (9) beats with wiggler magnetic field in eq. (3) to produce a transverse pondermotive

force at $(2\omega_1, 2k_1 + k_o)$

$$v_2^{NL} = \frac{-e^4 A_1^2 B_o^2 k_1 e^{-i(2\omega_1 t - (2k_1 + k_o)z)}}{128m^4 \omega_1^4 (\omega_1 + i\nu)} x$$
(10)

$$v_2^{L} = \frac{-eE_2}{m i(2\omega_1 + i\nu)} x$$
(11)

Second harmonic current density

$$J_2^L = \frac{-n_o e^2 E_2}{mi(2\omega_1 + i\nu)} x$$
(12)

$$J_2^{NL} = \frac{-n_o e^5 A_1^2 B_o^2 k_1 e^{-i(2\omega_1 t - (2k_1 + k_o)z)}}{128m^4 \omega_1^4 (\omega_1 + i\nu)} x$$
(13)

Now wave equation for Second harmonic for electric field of eq. (4)

$$\nabla^2 E_2 = \frac{4\pi \partial J_2}{\epsilon_L c^2 \partial t} + \frac{\partial^2 E_2}{c^2 \partial t^2}$$
(14)

On solving eq(14) using eq. (4),(12),(13)

$$\frac{\partial A_2}{\partial z} + \frac{2\omega_1}{k_2 c^2} \frac{\partial A_2}{\partial t} = \frac{4\pi n_o e^5 B_o^2 k_1 A_1^2}{128 \varepsilon_L c^2 k_2 m^4 \omega_1^3 (\omega_1 + i\nu)} \quad (15)$$

$$\frac{\partial A_2}{\partial z} + \alpha \frac{\partial A_2}{\partial t} = \beta A_1^2 \quad (16)$$

where

$$A_1^2 = [F(z - v_{g1}t)]^2$$

$$v_{g1} = c \sqrt{\varepsilon_L - \frac{\omega_p^2}{\omega_1^2}}$$

$$v_{g2} = c \sqrt{\varepsilon_L - \frac{\omega_p^2}{4\omega_1^2}}, \quad \alpha = \frac{2\omega_1}{k_2 c^2}, \quad \beta = \frac{4\pi n_o e^5 B_o^2 k_1}{128 \varepsilon_L k_2 c^2 m^4 \omega_1^3 (\omega_1 + i\nu)} \quad (17)$$

using eq(17) eq(16) can be written as

$$\frac{\partial A_2}{\partial z} + \frac{1}{v_{g2}} \frac{\partial A_2}{\partial t} = \beta [F(z - v_{g1}t)]^2 \quad (18)$$

Now considering temporal profile of laser pulse to be Gaussian

$$F(z - v_{g1}t) = A_o e^{-\frac{(z - v_{g1}t)^2}{\tau^2 v_{g1}^2}} \quad (19)$$

Laser pulse length is τ

Normalized amplitude of second harmonics

$$\left| \frac{A_2}{A_o} \right| = \left[\frac{1}{\frac{4\omega_1^2}{c^2} (\varepsilon_L - \frac{\omega_p^2}{4\omega_1^2})} \right]^{1/2} \frac{4\pi n_o e^5 B_o^2 k_1}{128 \varepsilon_L c^2 m^4 \omega_1^3 (\omega_1 + i\nu)} \frac{\zeta_o A_o \sqrt{\pi}}{2\beta'} [erf(z' - t') - erf((1 - \beta')z' - t')] \quad (20)$$

3. Results and discussion

In this paper semiconductor plasma has been considered. In semiconductors charge carriers electrons and holes are created in pairs. Mobility of holes contributes to the absorption process, which behaves like electrons. Here the generation of second harmonic and the variation of normalized amplitude A_2 / A_0 (eq(20)) with normalized distance z' under the effect of different parameters have been studied. For typical case n type Ge semiconductor with a doping level of $n_o \approx 10^{20} \text{ cm}^{-3}$ is irradiated with CO₂ laser with $\omega_1 = 1.8 \times 10^{14} \text{ rad/s}$, with collisional frequency of $\nu = 10^{14} \text{ Hz}$. Figure 1 shows the variation of normalized amplitude A_2 / A_0 with normalized distance z' at $\xi_0 k_1 \sqrt{\pi} / 2\beta' = 0.5$, $eA_0 / m\omega_1 c = 9$, $eB_0 / m\omega_1 = 0.8$ for $t' = 2$, $\omega_p / \omega_1 = 0.4, 0.6, 0.8$ and Figure 2 shows variation at $t' = 4$ and it has been observed that as time increases, the increase in plasma frequency enhances the wave amplitude producing more focused and sharp second harmonic which can lead to better imaging outputs.

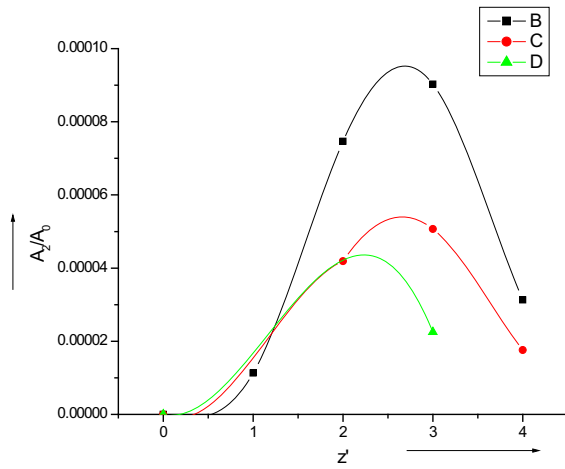


Figure 1 shows variation of normalized amplitude A_2 / A_0 with z' at different values of ω_p / ω_1 at $t'=2$.

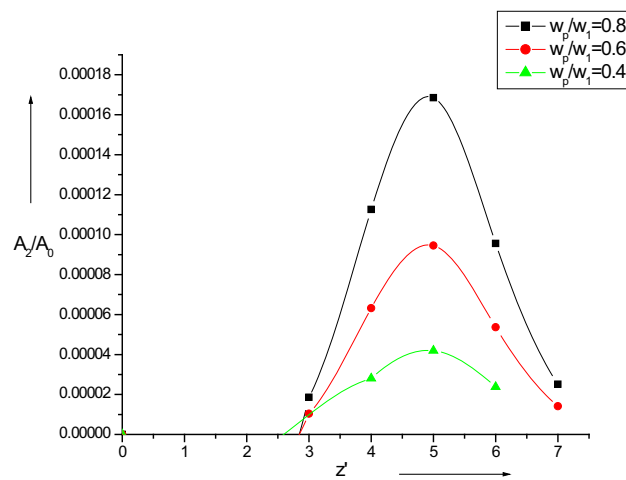


Figure 2 shows variation of normalized amplitude A_2 / A_0 with z' at different values of ω_p / ω_1 at $t'=4$.

4. Second harmonic waves in imaging microscopy

Second harmonic imaging microscopy is a powerful tool in laser scanning systems with high resolution, high contrast and three dimensional studies of live cells and tissues.

SHG has been used to study collagen fibers which showed that collagen fibrils are regularly packed as polycrystalline lattice. The study of sclera fibrils revealed their structure as inhomogenous tube like structures comprising thin hard shells[6].

The use of SHG in imaging microscopy has helped to study those structures which earlier were thought to be studied by electron microscopy. The use of ultra fast lasers can even provide more advantage in terms of clarity of image and to study the events which last for picoseconds to femtoseconds. Another advantage it provides is the reduced amount of collateral damage.

5. Conclusion

The interaction of ultra fast lasers with matter generates higher harmonics with low dispersion and better self focusing. Due to low pump power required by these lasers, their conversion efficiency is better. Increased amplitude of incident beam leads to better focused second harmonic at very low dispersion rates. The SHG by ultra fast lasers provides advantage to imaging microscopy in terms of better resolution and study of events lasting from picoseconds to femtoseconds. The future aspect of the use of SHG by ultra fast lasers may include the study of tumour formation in chronic diseases which may be proved beneficial in their treatment.

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Effect of wiggler magnetic field on harmonic generation at metallic surface

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Abstract. Second harmonic generation induced by surface plasma wave at the metal-semiconductor interface has been investigated. A thin layer of metals like Cu and Al have been considered in attenuated total reflection configuration(ATR) with a thin layer of semiconductor n-InSb over it. The study has been done in the presence of a wiggler magnetic field. The field provides additional momentum to the photons which help to attain phase matching conditions, leading to a resonant process of harmonic generation. Numerical analysis under given conditions show that Cu-InSb is the better choice for the harmonic generation as compared to Al-InSb.

Keywords: Wiggler magnetic field, second harmonic generation

1. Introduction

The interaction of laser with the matter has gained the interest of many researchers and has many applications like harmonic generation, surface plasmon resonance, sensors, accelerators, etc.[1–4]. The interaction of laser with metals gives rise to another type of electromagnetic wave called surface plasma wave with amplitude higher than that of the laser. These waves are localized near the interface and their intensity decreases with depth. SPW can be generated in metal by using either the Otto configuration or the Kretschmann configuration. For a thin layer of metals, the Kretschmann configuration is preferred in which, a thin layer of metal is considered over glass prism and the laser is incident from the glass-metal interface. The electromagnetic fields of laser interact with electrons of metals by exerting ponderomotive force, leading to SPW[5–8]. In metal-semiconductor heterostructures, the oscillations of conduction electrons decay upon surface plasmon resonance and generate energetic electrons for further excitation of photons[9]. Chauhan and Parashar.[10] theoretically studied second harmonic generation assisted by SPW over a metal-vacuum interface using Kretschmann attenuated total reflection (ATR) configuration and their work showed that the presence of external factors like grating structure or density ripple helps to enhance the harmonic generation efficiency. Sharma *et.al.*[11] studied the second harmonic generation of a cosh-Gaussian laser beam in magnetized plasma in the presence of a wiggler magnetic field and concluded that the presence of a wiggler field helps to enhance the efficiency of harmonic generation by providing additional momentum to the second harmonic photon which helps in phase matching and makes the process resonant. Tsang[12] investigated second harmonic and third harmonic generation using ATR configuration for Ag and found



that better results are shown by femtosecond lasers of intensity greater than $1 \times 10^{11} \text{ Wcm}^{-2}$. Abedi *et.al.*[13] studied the generation of terahertz (THz) in wiggler assisted collisional magnetized plasma and found an increase in the amplitude of THz radiation with the increase in wiggler frequency.

In the present paper, a model for the second harmonic generation has been presented using the metal-semiconductor interface in Kretschmann ATR configuration with metals like Cu and Al, each with a thin layer of n-InSb over it. The harmonic generation has been studied in the presence of a wiggler field. Laser is incident at the glass-metal interface which produces SPWs in metal. The electromagnetic fields of SPW interact nonlinearly with electrons of the semiconductor by exerting ponderomotive force and generate the second harmonic. The presence of wiggler magnetic field help to resonate the process by providing additional momentum to second harmonic photons which further helps to attain phase matching conditions. The present work shows that the Cu-InSb interface gives the second harmonic more efficiently than the Al-InSb interface.

In this paper, section 2 contains the derivation of mathematical relations of normalized second harmonic amplitude with propagation distance. Section 3 includes the graphical variation and its outcomes and section 4, includes conclusions.

2. Theoretical considerations

In the present work, laser beam with Gaussian profile has been considered which is incident through glass electric and magnetic fields of incident laser be $\vec{E}_1$ and $\vec{B}_1$ respectively, such that as $\vec{E}_1 = \hat{x}F(z,t)\exp(-i(\omega_1 t - k_1 z))$ and $\vec{B}_1 = (\vec{k}_1 \times \vec{E}_1)/\omega_1$ where, A is the amplitude of electric field of laser, ω_1 is the frequency and k_1 is the propagation constant of laser. These fields interact with electrons of metal and produces surface plasma waves $\vec{E}_s = \hat{x}F_s \exp(-i(\omega t - k_s z))$ where, k_s is the propagation constant such that, $k_s = (\omega/c)(\epsilon_s \epsilon_m / (\epsilon_s + \epsilon_m))^{1/2}$ with ϵ_m and ϵ_s as the permittivity of metal and semiconductor respectively and ω is the frequency of surface plasma wave.

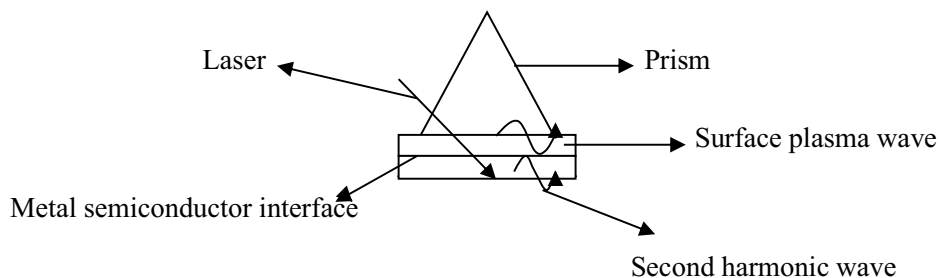


Figure1. Kretschmann configuration with thin layer of a metal and a semiconductor.

Wiggler magnetic field is applied in transverse direction given by, $\vec{B}_w = B_0 \exp(ik_0 z) \hat{y}$ where, B_0 is the amplitude of wiggler field and k_0 is the wiggler wave number. According to [14], F_s / F can be considered as a normalized quantity with $F_s = NF$ where, N is a positive real number.

Using equation of motion,

$$m \frac{d\vec{v}_1}{dt} = -e\vec{E}_s - e\vec{E} \quad (1)$$

The velocity gained by electrons is,

$$\vec{v}_1 = \left[\frac{-eF(1+N)\exp(-i(\omega t - k_s z))}{m(-i\omega)} \right] \hat{x} \quad (2)$$

The velocity $\vec{v}_1$ beats with wiggler magnetic field of laser and generates ponderomotive force $\vec{f}$, where, $\vec{f} = -e(\vec{v}_1 \times \vec{B}_w)$ due to this force velocity of electrons changes and it is taken as $\vec{v}_1'$ such that, it will induce density perturbation at $(\omega_1, k_1 + k_0)$ in agreement with equation of continuity,

$$n_1 = (\vec{k}_1 + \vec{k}_0) \cdot \vec{v}_1' / \omega_1 \quad (3)$$

Now $\vec{v}_1'$ beats with $\vec{B}_1$ and imparts velocity $\vec{v}_2^{NL}$ to electrons given as,

$$\vec{v}_2^{NL} = \left[\frac{-ie^3(1+N)F^2 k_1 B_0 \exp[-i(2\omega t - (2k_1 + k_0)z)]}{2m^3 \omega^4} \right] \hat{x} \quad (4)$$

This give rise to second harmonic nonlinear current density at metal semiconductor interface,

$$\vec{J}_2^{NL} = -n_0 e \vec{v}_2^{NL} - n_1 e \vec{v}_1 / 2 \quad (5)$$

The linear current density due to self consistent second harmonic field $\vec{E}_2 = F_2 \exp(-i(2\omega t - k_2 z)) \hat{x}$ at $(2\omega, k_2)$ is $\vec{J}_2^L = n_0 e \vec{v}_2$ where, $\vec{v}_2$ is obtained using equation of motion $m(d\vec{v}_2 / dt) = -e\vec{E}_2$.

The amplitude A_2' for second harmonic is calculated by using equation,

$$\nabla^2 \vec{E}_2 = \frac{4\pi(\epsilon_m + \epsilon_s)\partial \vec{J}}{c^2(\epsilon_m \epsilon_s)\partial t} + \frac{\partial^2 \vec{E}_2}{c^2 \partial t^2} \quad (6)$$

Laser profile has been taken Gaussian as $F^2 = F_0^2 \exp\left[-\left(\frac{z - v_{g1}t}{\tau v_{g1}}\right)^2\right]$, with $v_{g1} = c\sqrt{1 - (\omega_p^2/\omega^2)}$ and τ as laser pulse width. Using a new set of variables $z - v_{g1}t = \chi$, $z = \eta$, $\chi_0 = \tau v_{g1}$, $\beta' = (1 - v_{g1}/v_{g2})$, $\xi = z/\chi_0$ and $t' = v_{g1}t/\chi_0$ we get normalized second harmonic amplitude as,

$$\left|\frac{F_2}{F_0}\right| = \left\{ \sqrt{\left(1 - \frac{\omega_p^2}{\omega^2}\right)} \left(\frac{\epsilon_m + \epsilon_s}{\epsilon_m \epsilon_s}\right) \left(\frac{\omega_p^2}{\omega^2}\right) \left(\frac{eB_0}{m\omega}\right) \left(\frac{eF_0}{mc\omega}\right) \times \left[\frac{k_1 \chi_0 \sqrt{\pi} (1 + N)}{8\beta'} + \frac{(k_1 + k_0) \chi_0 \sqrt{\pi} (1 + N)^2}{4\beta'} \right] \times \left[\text{Erf}(\xi - t') - \text{Erf}\{(1 - \beta')\xi - t'\} \right] \right\} \quad (7)$$

3. Results and discussion

Eq.(7) has been solved numerically for Nd:YAG laser with $\omega = 1.7 \times 10^{15} \text{ rad/s}$. Values for effective permittivity are taken as 49.58 for Cu, 106.82 for Al and 19.5 for InSb. The other parameters of Eq.(8) are taken as $\omega_p/\omega_1 = 0.9$, $eB_0/m\omega = 0.02$ and 0.04 , $n' = 2.5$, $k_1 \chi_0 \sqrt{\pi}/8\beta' = 0.2$, $\beta' = 0.3$, $eF_0/mc\omega_1 = 9$ and $(k_1 + k_0) \chi_0 \sqrt{\pi}/8\beta' = 0.23$.

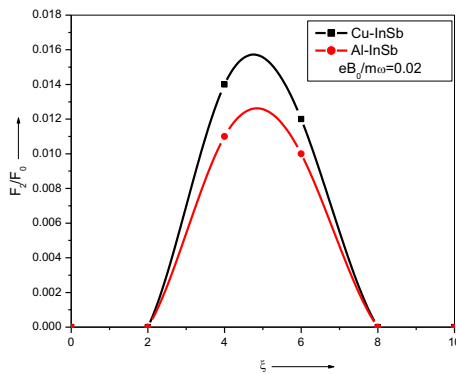


Figure 2(a)

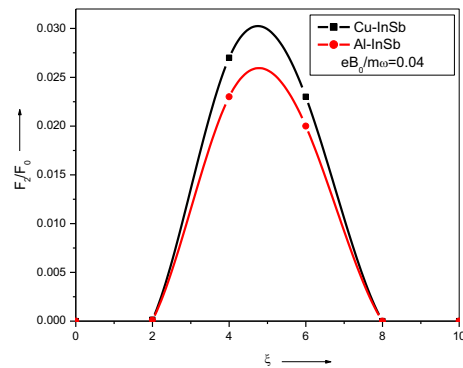


Figure 2(b)

Figure 2. Variation of normalized second harmonic amplitude F_2/F_0 with normalized propagation distance ξ for metals Cu, Ag and Al with layer of n-InSb for different values of wiggler field at time $t'=4$.

Figure 2 shows the variation of normalized second harmonic amplitude F_2/F_0 with normalized propagation distance ξ for metals like Cu and Al with a thin layer of n-InSb at time $t'=4$ in the presence of a wiggler magnetic field. It has been observed that as the strength of the magnetic field increases, the amplitude of the second harmonic increases. It is clear from Figure 2(a) that at wiggler frequency $eB_0/m\omega = 0.02$, amplitude of second harmonic at Cu-InSb interface shows more enhancement than Al-InSb. In Figure 2(b) the value of wiggler frequency has been taken as $eB_0/m\omega = 0.04$ and it has been observed that amplitude also shows enhancement by nearly 50%. Aggarwal *et al.* [15] studied SHG of the laser beam in the plasma of atomic clusters and found enhancement in the efficiency in the presence of wiggler field. Similarly, the present work shows that the amplitude of the second harmonic increases in the presence of a wiggler field.

4. Conclusion

The interaction of laser with metal surface gives rise to SPW, which are electromagnetic waves with amplitude higher than that of the laser. These waves propagate along the surface and can interact nonlinearly with electrons of n-type semiconductor, layered over the metal surface. The electromagnetic fields of SPW interact more efficiently with the electrons of semiconductor in the presence of a wiggler magnetic field. Under given conditions, good enhancement in the amplitude of the second harmonic has been observed for the Cu-InSb interface as compared to the Al-InSb interface.

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Harmonic generation in metals and semiconductors- A review

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Abstract

Harmonic generation in different materials has been a topic of interest for many researchers from many decades. Different materials have been explored under different conditions. Laser interacts with particles of material and accelerates them. Velocity of particles beats with the fields of laser and show different phenomena; one of them is generation of higher harmonics. This paper contains review on harmonic generation in metals and semiconductors.

Key words: Non-linear interactions, surface plasmons, charge concentration.

A Review Report

The interaction of em fields of laser with matter creates plasma. The charge carriers of matter get accelerated producing non linear effects. Type of plasma formed depends on nature of material and parameters of laser used. In metals free electrons form plasma upon interaction with em fields. Electrons beat with electric and magnetic fields and leads to non linear current density producing non linear effects like generation of harmonic waves with frequency higher than the frequency of incident light. In metals interaction of photons and electrons also gets influenced by electron-electron and electron-ion interactions. In metals harmonic generation from bulk arises due to polarization of magnetic dipole source or two types of surface polarizations which include parallel and perpendicular modes. Out of these, the contributions of bulk and parallel surface depend only on bulk properties and they are not surface sensitive. Hence main contribution to non linear phenomena in metals is due to surface effects. However presence of adsorbents over metal surface may reduce contribution of normal surface polarizations then bulk and parallel surface contributions dominate.[1] Timbrell *et. al.* showed that for metals surface contribution to non linear power generation is much more than bulk contributions.[2] The non linear behavior arises because of interaction of photons with atoms of metallic surface. Metal clusters which are made of several hundreds of atoms supports plasmon excitations.[3] In metals interaction of photons with surface

plasmons lead to polaritons and corresponding waves are called surface plasmon polaritons (SPP). In past few years SPP based structures have gained great importance due to its properties and applications in optics, Surface enhanced Raman spectroscopy (SERS), data storage etc. Electrons or photons can excite SPPs but in case of photons presence of other agents like prisms or gratings or defects on metal surface is needed. Such interactions lead to production of harmonic waves. Surface plasmon resonance based circuit has overcome the limitations related to size of circuits to be used in data processing nano devices with high performance.[4] By using nano structures specifically made out of metals like Ag, Ni, Au etc. external em fields can be localized on length scales of just a few nanometers, resulting in greatly enhanced field amplitudes that can exceed those of the external field by orders of magnitude in the vicinity of nanostructures. Metals like Ag, Au have been studied and give good response but work on metals like Co, Ti, Ni, metal oxides has also been done in past few years.[5] For metal vacuum interface, surface plasma wave of large amplitude heats up the electrons and gets absorbed, this increase in temperature causes thermionic emission of electrons. To attain large amplitude required for surface plasma waves can be attained by depositing a metal film on face of prism in attenuated total reflection configuration.[6] Harmonics generated from magnetic resonance in Al-GaAs on aluminium oxide nanoantennas can be made resonant by varying height and radius of nanoantennas and irregularities in metal surfaces behave equivalently to nanostructures.[7] Nano particles made of high refractive index dielectric materials have been proposed as an alternative to metals, driven by their low losses and presence of magnetic response inspite of being non magnetic materials. They suffer large losses and absence of magnetic response at optical frequencies.[8] Nature of waves generated depends on angle of incidence also as if laser beams of different frequencies are incident on metal surface at angles θ and $-\theta$ then surface plasma wave with frequency equals to their difference is formed and frequency of surface wave depends upon incident frequencies and angels.[9] Presence of density ripple of suitable wave number on metal surface produces non linear current which give rise to plasma wave of different frequencies.[10]

In semiconductors charge carriers are electrons or holes depending type of doping. Number density is one of the important characteristics. Interaction of laser with semiconductors forms different types of plasma like under dense plasma, over dense plasma etc. Kant *et. al.* have studied such plasmas under different conditions and concluded presence of density ripple enhances wave amplitude to remarkable extent. Their study also includes

impact of different laser profiles like Gaussian profile, Hermite Gaussian and Hermite-cosh Gaussian.[11,12] The electric and magnetic fields of laser drifts electrons creating non linear current density and leading to production of different harmonics. The amplitude and type of harmonic is decided by frequency of incident laser, charge carrier concentration. Other agents like presence of density ripple, wiggler field, chirp pulse also enhances efficiency of production and amplitude of wave generated. Thakur *et al.* have observed the effect of chirp, wiggler field with density ripple and has found good enhancement in harmonic amplitude.[13–16] For third harmonic generation in n-InSb and Ag enhancement in the electric field in case of n-InSb by order of 2 has been observed than for Ag due to high non linear density in semiconductors than metals.[17] Combination of metals with dielectrics also lead to formation of higher harmonics at the interface, surface plasma waves of metals help in this phenomena.[8] Charge density is one of the important deciding factors for the order of harmonic generation, highly doped semiconductors respond satisfactorily for the same.

Both metals and semiconductor show high harmonic generation due to interaction of em fields with charge particles particularly electrons. Both show non linear behavior upon interaction which leads to non linear current density producing harmonic waves as well as other non linear phenomena. Metals which are generally good conductors like Ag, Au, and Al or with good magnetic properties like Co, Ni etc. show harmonic generation upto good extent. In case of semiconductors charge concentration depends on doping which facilitates this process. Recent work includes generation of waves in layered pattern like metal-dielectric interface, metal-metal oxide etc. Advancements in this field will lead to improvement in imaging techniques.

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Enhancement in Second Harmonic Generated by ultra-fast Lasers in Plasma by wiggler magnetic field

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ABSTRACT

Nonlinear optical effects which arise due to laser's interaction with plasma includes higher harmonic generation. High density beams of ultra fast lasers provides oscillations to charge carriers which in turn interacts with magnetic field giving rise to various effects. This paper studies influence of applied wiggler magnetic field on amplitude of second harmonic generated for semiconductor plasma.

KEY WORDS

Ultra fast Lasers ,wiggler magnetic field.

1.INTRODUCTION

Plasma exists in different forms like gaseous plasma , semiconductor plasma ,metal plasma etc. The constituent particles are mainly charged particles like ions, electrons etc. [1, 2] Introduction of Laser in plasma oscillates electrons and these oscillating electron's frequency interacts with laser frequency to give rise to different phenomenons including generation of different harmonics [3]. The electrons which recollides with the parents ions generally give rise to higher harmonics. Along with laser field , the non linearity of the medium plays an important role in producing fields with different frequencies [4]. High intensity beams of ultra fast lasers give more precise outputs in terms of focused waves [5]. Wiggler magnetic field also enhances amplitude of second harmonic by providing additional momentum. This makes the process resonant and increases efficiency of the harmonic generation [6]. The use of ultra fast lasers and thin crystals reduces the dispersive effects which leads to the condition $n(2\omega) = n(\omega)$, which

implies $k(2\omega) = k(\omega)$ and $\lambda_{\omega} = 2\lambda_{2\omega}$. If phase mismatch occurs then SH generated may go out of phase [7]. This paper studies the impact of wiggler magnetic field on a semiconductor plasma.

In this paper section 2 contains derivation for amplitude of second harmonic in normalized form , section 3 contains results and discussions, section 4 explains its application in imaging microscopy and conclusion is given in section 5.

2. THEORETICAL CONSIDERATIONS

The incident laser beam is represented as

$$\vec{E}_1 = \hat{x}A_1(z,t)e^{-i(\omega_1 t - k_1 z)}, \quad (1)$$

$$\vec{B}_1 = \frac{\vec{k}_1 \times \vec{E}_1}{\omega_1}, \quad (2)$$

$$\vec{B}_W = \hat{y}B_0 e^{ik_0 z} \quad (3)$$

$\vec{B}_W$ is transverse wiggler magnetic field , A_1 is amplitude of fundamental laser.

Second harmonic generated due to non-linear interaction is represented as

$$\vec{E}_2 = \hat{x}A_2(z,t)e^{-i(\omega_2 t - k_2 z)}, \text{ with} \quad (4)$$

$$\omega_2 = 2\omega_1$$

Linear dispersion relation followed by fundamental and 2nd harmonic is

$$k^2 \approx \frac{\omega^2}{c^2}(\epsilon_L - \frac{\omega_p^2}{\omega^2}) \quad (5)$$

where ϵ_L is lattice permittivity .

and $\omega_p^2 = 4\pi n_0 e^2 / m$ is called plasma frequency.

Here $k_2 > 2k_1$, the difference is delivered to second harmonic by wiggler magnetic field

when

$$k_2 = 2k_1 + k_o$$

$$k_o = k_2 - 2k_1$$

$$k_o = \frac{3\omega_p^2}{4\omega_1 c \sqrt{\epsilon_L}} \quad (6)$$

Using eq (1) and (2)

$$\text{velocity } v_1 = \frac{eE_1 \hat{x}}{mi(\omega_1 + i\nu)} \quad (7)$$

It beats with magnetic field of eq (3) and then with magnetic field of eq (2) to give second

harmonic and provides oscillatory velocity v_2 at $(2\omega_1, 2k_1 + k_o)$

$$v_2 = \frac{-e^3 A_1^2 B_o k_1 e^{-i(2\omega_1 t - (2k_1 + k_o)z)} \hat{x}}{16m^3 \omega_1^3 (\omega_1 + i\nu)} \quad (8)$$

Now velocity in eq (8) beats with wiggler magnetic field in eq. (3) to produce a transverse

pondermotive force at $(2\omega_1, 2k_1 + k_o)$

$$v_2^{NL} = \frac{-e^4 A_1^2 B_o^2 k_1 e^{-i(2\omega_1 t - (2k_1 + k_o)z)} \hat{x}}{128m^4 \omega_1^4 (\omega_1 + i\nu)} \quad (9)$$

$$v_2^L = \frac{-eE_2}{m i(2\omega_1 + i\nu)} \hat{x} \quad (10)$$

Now wave equation for Second harmonic for electric field of eq. (4)

$$\nabla^2 E_2 = \frac{4\pi\partial J_2}{\epsilon_L c^2 \partial t} + \frac{\partial^2 E_2}{c^2 \partial t^2} \quad (11)$$

On solving

$$\frac{\partial A_2}{\partial z} + \frac{2\omega_1}{k_2 c^2} \frac{\partial A_2}{\partial t} = \frac{4\pi m_o e^5 B_o^2 k_1 A_1^2}{128 \epsilon_L c^2 k_2 m^4 \omega_1^3 (\omega_1 + i\nu)} \quad (12)$$

$$A_2 = \left[\frac{1}{\frac{4\omega_1^2}{c^2} (\epsilon_L - \frac{\omega_p^2}{4\omega_1^2})} \right]^{1/2} \frac{4\pi m_o e^5 B_o^2 k_1}{128 \epsilon_L c^2 m^4 \omega_1^3 (\omega_1 + i\nu)} \frac{\zeta_o A_o^2 \sqrt{\pi}}{2\beta'} [erf(z'-t') - erf((1-\beta')z'-t')] \quad (13)$$

3. RESULTS AND DISCUSSION

In this paper semiconductor plasma has been considered. For typical case n type Ge semiconductor with a doping level of $n_o \approx 10^{20} \text{ cm}^{-3}$ is irradiated with Nd:YAG laser with $\omega_1 = 2.8 \times 10^{14} \text{ rad/s}$, with collisional frequency of $\nu = 10^{14} \text{ Hz}$. Figure 1 shows the variation of normalized amplitude A_2/A_0 with normalized distance z' at $\xi_o k_1 \sqrt{\pi}/2\beta' = 0.5, eA_o/m\omega_1 c = 9, eB_o/m\omega_1 = 0.6, 0.8, 1$ for $t'=0, \omega_p/\omega_1 = 0.8$ and Figure 1 shows variation at $t'=2$ and Figure 2 exhibits variation of the amplitude with position at $t'=4$. From both figures it has been observed that as time increases, increase in the magnitude of wiggler magnetic field at particular plasma frequency enhances the wave amplitude producing more focused and sharp second harmonic which can lead to better imaging outputs.

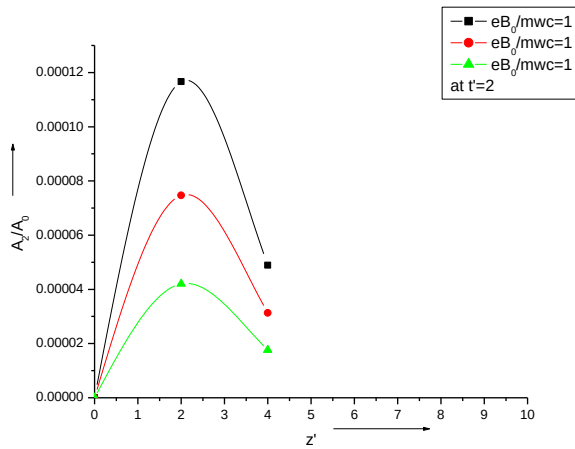


Figure 1 shows variation of normalized amplitude A_2 / A_0 with z' at different values of ω_p / ω_1 at $t'=2$.

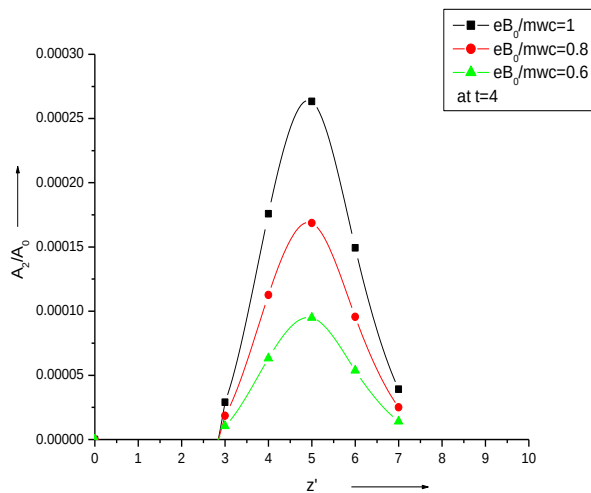


Figure 2 displays variation of the normalized amplitude A_2 / A_0 with z' at different values of ω_p / ω_1 at $t'=4$.

5. CONCLUSION

Laser's interaction with matter like plasma generates higher harmonics due to interaction between applied field and electrons. The application of wiggler magnetic field enhances their amplitude and makes output waves more focused with time. The increase in amplitude of applied wiggler magnetic field at particular time further increases the amplitude of output wave which in turn makes higher harmonic generation process more efficient. This may lead to better imaging and may prove itself useful for medical purposes.

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