Magnetic field enhanced nonlinear laser absorption by metallic surface embedded with noble-metal nanotubes

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**ABSTRACT**

A theoretical model has been developed where a p-polarised laser is irradiating a metallic surface ingrained with nanotubes at oblique incidence in the presence of a DC magnetic field. Interaction between incident laser and nanostructured metallic surface stimulates the electrons of the nanotubes, causing them to excite and resonate at the plasma frequency. Incorporating external magnetic field within the system of nanotubes significantly enhances the absorption phenomenon. The absorption coefficient has a sharp peak at surface plasma resonance , where is the plasma frequency and it shows explicit dependence on the amplitude transmission coefficient of the incident laser as well as on dimensions of the nanotubes. Increasing the magnetic field strength along with the angle of incidence is found to produce more sharp absorption peaks. Due to large surface-to-volume ratio of the nanotubes, the carried-out study has a straight relevance in various biomedical applications like DNA and immunosensors, cellular and photoacoustic imaging, and photothermal therapy, in electrochemical sensors due to their excellent catalytic properties, in optoelectronic devices like solar cells and in molecular diagnostics.

**Keywords-** Nonlinear absorption; Nanotubes; External magnetic field; Laser-metal interaction

1. **INTRODUCTION**

Plasmonics refers to the manipulation, generation, and application of plasmons along a metal-dielectric interface [1] while magnetoplasmonics, on the other hand, deals with the interaction of plasmons with magnetic field [2]. Surface plasmons (SPs), or simply plasmons are the quanta associated with plasmonics whereas surface magneto plasmons (SMPs) are associated with magnetoplasmonics.

Plasmons play an integral role in the investigation of optical properties of metals as they can easily overcome the optical-diffraction limit [3]*.* SPs have different properties compared to SMPs due to implication of magnetic field. Both SPs and SMPs are sustained by a range of metallic nanostructures, like nanodots, nanowires, nanotubes, nanocages, nanoshells, etc., [3], [4] thus providing a huge potential for the development of integrated nanophotonic devices [1], [5]. Combining the two fields have led to the evolution of several concepts like magneto-optical properties, plasmonic thermal effects on magnetization and plasmon-driven non-linear effects in magneto-optics [6]. During the last few years, various researches have been carried out in the above said fields. Plasmonics play a crucial role in nano-imaging [7], sensing applications through metamaterial with very high Q-factors [8], photovoltaic devices and near-field optical microscopy [5] along with various biomedical applications [9]. Magnetoplasmonics on the other hand find employment in sensing field via magnetoplasmonic nanowire gratings [10], magnetoplasmonic interferometers [11] and magnetoplasmonic crystals as magnetic field sensing and all-optical magnetic data storage [12].

Studying the absorption phenomenon using laser-metal interaction in one of the most important techniques regarding this purpose. Laser-metal interactions refer to the dynamic interplay between laser energy and metal materials. Laser-metal deposition [13] and ablation [14], [15], terahertz field enhancements [16], linear and nonlinear resonance absorption [17]–[20], waveguiding applications [21], [22], medical imaging [23], bio-physical studies [24], remote sensing [25], and various types of scattering like Rayleigh [26] and Compton scattering [27] are few of the many applications of laser-metal interactions. Absorption associated with smooth metal surfaces is relatively low as compared to rough metal surfaces due to the presence of the high free-electron density as they disperse light energy in the neighbouring medium [28]. Nano-structuring, which involves adding metallic nanoparticles to a material's surface to make it coarser, enhances the surface's capacity to absorb light due to multiple trapping of light. Thus, increasing the roughness of the surface via micro or nano-structuring is a viable option for the amplification of absorption phenomenon. Absorption phenomenon associated with nanostructured metal surfaces offer opportunities for enhancing optical processes, such as surface-enhanced Raman spectroscopy [29] and plasmonic photothermal therapy [30]. The interaction of electric field of the laser drives the electrons of the nanotubes to the phenomenon of surface plasmon resonance (SPR). It refers to the combined oscillations of the electrons confined between a metal and a dielectric, that allows trapping of light between the metal-dielectric interface. This further makes guiding and manipulation of light possible and efficient at nanoscale. SPR, owing to the exclusive physical, chemical, mechanical and structural properties of nanotechnology have brought about a revolutionary change in the fields of science, engineering and medicine [28]. Applying SPR to noble metal nanostructures have shown incredible growth in the past few years. Zada et al. [31] showed how incorporating SPR in noble metal nanocrystals significantly enhances the efficiency of various optoelectronic devices like photovoltaics and photodetectors. Mishra et al. [32] investigated how nanostructures made of noble metal modified magnetite is used as recyclable photocatalytic material. Hu et al. [9] studied plasmonic properties of gold nanostructures for utilization in biomedical field. They compared absorption, scattering, and extinction phenomenon for gold nanorods, nanocages, nanoshells and nanospheres. Their optical properties were thought-out for optical imaging and photothermal treatment. Graphene magnetoplasmonics has recently surfaced that shows immense potential. Kumar et al. [33] carried out theoretical formalism for THz generation via interaction of laser beams in an array of magnetised anharmonic carbon nanotubes. Titova et al. [34] generated terahertz radiation from large arrays of single-wall carbon nanotubes using femtosecond optical pulses. Kuzmin et al. [35] demonstrated graphene based magnetoplasmonics and topological effects in graphene nanostructures. Graphene nanocomposites show enhanced optical absorption in the infrared and THz region due to their improved chemical and physical properties that allows SPR. Shi et al. [36] carried out animal experiments to demonstrate photothermal therapy cancer treatment. They used graphene-based multi-functional nanocomposite as cancer theranostics as well as theragnostics.

Optical properties of plasmonic nanostructures can be tuned by modifying their shapes and sizes. If synthesised properly, they provide excellent control over propagation and manipulation of light [37]. In the case of noble-metal nanotubes, the resonant frequency is typically associated with the excitation of localized surface plasmons, which are collective oscillations of conduction electrons within the nanotubes. The shape and size of the nanostructure, such as the diameter, length, and aspect ratio can significantly affect the resonant frequency of the localized surface plasmons. This is because the geometrical parameters determine the confinement and coupling of the electromagnetic field within the nanotubes, thereby influencing the resonant properties. Furthermore, when differently shaped nanostructures are embedded on the metal surface, the resonant interaction between the laser and the particles occurs when where is ellipticity characterizing parameter [20]. Various efforts have been made to exploit this tunable nature of nanostructures by creating various geometries of nanorods, nanoparticles, nanoshells, nanorings, etc. Shape and size of the nanostructure attributes significantly to the plasmonic modes [38]. The resonant absorptance behaviour of metallic nanoparticles with respect to the incident radiation can be tuned by varying their shapes and sizes. When the incident light frequency matches with the natural frequency of oscillation of electrons in the nanoparticles, strong absorbance is obtained at that frequency [39]. Further, adopting double nanoparticle system over single nanoparticle system exhibits much superior absorptance. The former is more efficient in trapping sunlight and has made its way to be employed in thin-film silicon solar cell for improved solar cell efficiency [40]. FDTD simulations have also been carried out to study the propagation of surface electromagnetic waves along the nanotubes as well as the interaction of light with nanoscale cylinders [41], [42].

Various nanofabrication methods for synthesising plasmonic nanostructures have been reported. Soft-interface lithography generates nanostructures over wafer-scale areas. In addition to this, electron beam lithography and photolithography are other efficient methods to produce differently shaped nanoparticles [43], [44]. Liu et al. [45] have mentioned various methods like hard template methods, anodic alumina oxide (AAO) template method and track-etched membranes for manufacturing gold nanotubes, all of which are highly efficient in synthesising gold nanotubes. Various researchers have experimentally fabricated noble-metal nanotubes having average inner core diameters lying in the range , wall thicknesses of and nanotube lengths of [38], [46], [47]. Hendren et al. [48] manufactured gold nanotubes using thin film AAO templates and polypyrrole cores with inside and outside diameters being and respectively, to study their optical properties with s- and p-polarized light at several angles of incidence.

In this paper, a theoretical analysis has been presented where absorption phenomenon of a p-polarized laser irradiating a metal surface that has been ingrained by metallic nanotubes at an oblique angle. This system is placed in midst of external static magnetic field. The laser-metal interaction stimulates the electrons of the nanotubes, causing them to excite and resonate at the plasma frequency, . In the proximity of this frequency, absorption coefficient attains its peak owing to resonant plasma oscillations. The results show that the absorption coefficient has direct dependence on transmission coefficient of the incoming laser beam and show significant enhancement when radius and length of the nanotubes are varied. The study has been executed for different incident angles and different magnetic field strengths as well. Prior research includes absorption of surface plasma wave and laser pulses on nanostructured metal surface with/without magnetic field [20], [49], [50]. Similar studies, one involving p-polarised laser on metallic nanoparticles and nanotubes [51] and the other involving Gaussian laser beam through an array of nanotubes [52], has been carried out in the absence of magnetic field. However, incorporation of static magnetic field in this study as an external stimulus acts as an enhancement parameter that significantly amplifies the absorption phenomenon. Furthermore, since the authors are interested in the dynamic response of the nanoparticles and the metal surface with the incident laser light, a pulsed laser is used as it offers high peak powers that can induce nonlinear optical effects and rapid energy deposition within a short duration. This can lead to transient changes in the absorption properties of the nanoparticles and the metal surface, which is essential for the study. Various phenomena including absorption and terahertz generation, using lasers with similar configuration, that is, an array comprising of vertically and horizontally aligned nanotubes, in the presence of magnetic field have operated with intensities of the order [53]–[55].

This paper is divided into four sections. Section II provides the analytical study and the mathematical formalism associated with the absorption phenomenon. Section III presents the results and discussions while conclusions are enclosed in Section IV.

1. **MATHEMATICAL FORMULATION**

Consider a free space-metal interface along the -plane, placed at , such that the metal occupies the half space and free space takes the other half space (Figure 1). An array of metallic nanotubes with length , radius and interparticle distance is implanted on the metal surface. A p-polarized laser is obliquely irradiating the metal surface at an angle , along the -plane, in the presence of static magnetic field acting along the -direction. The electric field profile of the incident laser beam, as it propagates through a cluster of nanotubes is

(1)

where is the amplitude of the incident laser beam.



**Figure 1: Schematic diagram of an obliquely incident laser irradiating a metal surface that contains an array of metallic nanotubes in the presence of static magnetic field**

After its simultaneous reflection and transmission from the metal surface, reflected and transmitted profiles of electric field is

(2)

(3)

where and are the reflection and transmission coefficients, is the propagation vector along the z-direction and is the permittivity of metal. The gross electric field within the nanotubes is

(4)

When the incident laser beam irradiates the metallic surface, the electrons of the nanotubes get displaced from their mean position with effective displacement and so they exhibit a consequent oscillatory velocity . This interaction between electrons of the nanotubes and the laser is supervised by the equation of motion

(5)

Here, is the mass of the electron, is the electronic charge, is the collisional frequency, is the plasma frequency, being the electron density of the electrons of the nanotubes and being the permittivity of free-space. Here, is the externally applied static magnetic field along the 𝑦-axis. In (5), the first term represents momentum change, the second term represents collisional damping, and the third term corresponds to force acting on the electrons of the nanotubes due to the presence of external magnetic field. Rewriting the above equation on applying , gives us expressions for electronic displacement and velocity

(6)

(7)

Using and solving , (7) gives

(8)

(9)

where is the cyclotron frequency*,*  and .

Applying continuity equation for magnetic field and boundary condition for electric field at boundary at the metal-free space interface provides us with

(10)

(11)

where is the -component of current density and is the number density of electrons within the nanotubes. Now,

(12)

Here is the number of nanotubes per unit area, is the radius the nanotubes and is the inherent electron density of the nanotubes. Solving (11) and (12) gives us:

(13)

Simplification of the above equation gives us

Further solving (13) gives us the reflection and transmission coefficients

(14)

(15)

Poynting vector defines the amount of power absorbed by the electrons of the nanotubes when struck by laser beam and is given by

(16)

where \* refers to complex conjugate.

Elaborating (16) we get

(17)

Power absorbed per unit area per unit time by the electrons of the nanotubes

(18)

where , is the areal density of the nanotubes, being inter-tube separation. Absorption coefficient is evaluated as

(19)

where is the average power of the incident laser.

Using (18) and (19) and substituting the value of , we get

(20)

1. **RESULTS AND DISCUSSION**

Analytical results are presented when a laser in obliquely incident on a metallic surface that is ingrained with a thin array of metallic nanotubes in the presence of external DC magnetic field. Results are accomplished by varying strength of external magnetic field, radii of nanotubes, angle of incidence and for different materials. We have used normalised laser amplitude which corresponds to incident laser beam intensity [55]. Figure 2 presents the plot of normalized absorption coefficient with normalized frequency for different incident angles for gold nanotubes with parameters and . Figure 3 depicts normalized absorption coefficient versus normalized frequency for different cyclotron frequencies for nanotubes with radius . Rest of the parameters are and . In figure 4, we have variation of normalized absorption coefficient with normalized frequency for nanotubes with different radii and at and , whereas figure 5 shows variation of normalized absorption coefficient with normalized frequency for three different materials, i.e., for gold, silver, and carbon. Rest of the parameters remain unchanged.

It is apparent from figure 2 that with the increment in angle of incidence, the absorption coefficient increases. The absorption coefficient is found to be at whereas it is at . Hence an obvious gain in the absorption phenomenon is seen, which in this case is about 88%. Similar results have been obtained by [20] for nanoparticles and [51] with nanotubes in addition to nanoparticles without magnetic field. However, the application of external magnetic field significantly alters the optical absorption phenomenon associated with metal nanostructures as it provides much more effective and efficient laser-metal coupling.



**Figure 2: Normalised absorption coefficient vs normalised frequency for different incident**

**angles of the laser beam for an array of gold nanotubes**

Figure 3 shows gradual increase in the absorption coefficient with increase in the magnetic field strength. The static magnetic field, along with the incoming laser beam excites the electrons of the nanotubes, thereby leading them to demonstrate resonant plasma oscillations. The drift velocity of the said electrons is modified which further transform the current density. This has direct influence on the transmission coefficient and thus reforms the absorption coefficient. In this case, the surge in the absorption coefficient can be seen as it rises from to as the cyclotron frequency ranges from . The peaks obtained when magnetic field is applied are finer and more enhanced with significant increase in associated absorption. Varying the dimensions of nanotubes also have serious impact on the absorption phenomenon which can be seen from (20). Increasing the radius and length of the nanotubes increases the volume density of the electrons in the nanotubes, which results in greater number of electrons available for the incident laser beam to excite.



**Figure 3: Plot showing variation of normalised absorption coefficient vs normalised frequency for different cyclotron frequencies for gold nanotubes**

Figure 4 presents variation of normalized absorption coefficient with normalized frequency for different radii and lengths of nanotubes. Also, variation of absorption coefficient is shown for different materials while keeping the dimensions and rest of the parameters same in figure 5. Carbon shows most promising results as compared to silver and gold which are noble metals. As a results, many studies are being carried out using graphene and carbon nanotubes, either in the absence or presence of magnetic field.



**Figure 4: Variation of normalised absorption coefficient with normalised frequency for different radii of nanotubes ( for gold nanotubes**



**Figure 5: Normalised absorption coefficient (F) vs normalised frequency for an array of nanotubes of different materials**

State of polarization and the type of nano-structuring also contributes to the absorption phenomenon. Various studies have proved that p-polarised laser beam yields better absorption results as compared to its counterpart, s-polarised laser beam. Also, the reflection coefficient for s-polarised beam is much greater than its absorption coefficient, unlike for p-polarised beam, where the absorption coefficient associated is much greater than its reflection coefficient. Hence, using p-polarised laser beam aids our present study. Moreover, the presence of roughness via nanostructures on the metal surface amplifies the total effective surface area, which magnifies the surface plasmon resonant effects. Nanotubes show more promising results as compared to nanosheres as the former has large surface area as compared to latter. Gold nanotubes have a wide application in medical field. It is extensively used as biosensors such as DNA sensor, immunosensors, enzyme sensor, etc., photothermal therapy (PPT) for cancer treatment, and photoacoustic imaging because of its innocuous properties [45], [56], [57]. Gold nanoparticles have also been incorporated in inorganic solar cells to enhance their power conversion efficiency [58].

Recent progress has been made to generate terahertz (THz) radiation using nanostructures. Various researchers have employed a similar configuration to achieve tunable THz radiation. Vij et al. [59] proposed a study to generate THz radiation where two plane-polarised lasers with different frequencies were obliquely incident on an array of vertically aligned carbon nanotubes in the presence of an external wiggler magnetic field. Similar analysis was carried out by Kumar et al. [33] for an array of vertically aligned anharmonic carbon nanotubes in the presence of static magnetic field.

1. **Conclusions**

Here we explored the absorption phenomenon associated with p-polarised laser obliquely irradiating a metal surface topped with nanotubes in the presence of a static external magnetic field. When laser beam strikes the metal surface, coupling between the electric field of the laser and magnetic field alters the transmission and reflection coefficients, thereby enhancing the absorption phenomenon. Unlike s-polarised laser beam, a p-polarised laser has larger absorption coefficient, signifying better absorption and less reflectance. The proposed formalism shows that the absorption of incident laser beam on metal nanotubes is found to increase with increase in the angle of incidence and the cyclotron frequency, i.e., magnetic field strength. Increasing the surface roughness by employing nanostructures also aids in the absorption process. Embedding nanotubes on the metal surface augments absorption of light by incorporating local surface plasmon resonance effects. The applied magnetic field enhances the absorption phenomenon by exciting the electrons of the nanostructure and causing them to illustrate surface plasma resonance. Thus, the absorption phenomenon is found to have strong dependency on external magnetic field. Furthermore, it is found that the absorption coefficient has direct dependency on the transmission coefficient of the laser beam. The dimensions of the nanotubes also play a significant role in the study as the radii and the length of the nanotubes can be varied accordingly to get efficient absorption.

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