Shape And Collision Frequency Effect On Heating Of Femtosecond Laser Pulse Irradiated Nanostructures

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Abstract - *Heating of nano particle by ultrashort pulses in the vicinity of transition from cold solid state to dense and hot plasma state is explored analytically for various nano-structures in the regime of laser intensity (I)I<<10^{15}W/cm^2, and pulse duration <100 fs). The laser irradiated nano-structured material undergoing ionization is a highly dynamical system Surface Plasmon Resonance (SPR) of which can be excited during the ionization phase, this depends on the geometry of the nano- particle as well as the often neglected dynamical collisionality of the nano-particle undergoing ionization. Solid nanostructures like nano-sphere and nano-cylinder resonate at low electron density, Hollow nanostructures like nano-tubes and nano-shell, and pointed structures like nano-elliptic cylinder have a higher SPR electron density. Absorption rate can be highly enhanced at high electron densities in hollow and pointed nanostructures. The work brings out the critical role of target geometrical parameters and laser conditions for desired applications. Collisionless nano plasma (at very low intensity I< 10^{10}W/cm^2 and very high intensity I> 10^{15}W/cm^2) support huge field enhancement. Highly collisional nano plasma formed in the intesnsity range(10^{13}W/cm^2<I<10^{15}W/cm^2) has negligible field enhancement but efficient heating at high densities.*

Key Words: **Surface Plasmon Resonance, Nano-Structures, Nano-Cylinder,Nano-Shell, Nano-tube, Ultrashort Pulses, Absorption, Collision frequency**

1. INTRODUCTION

Light interaction with nanostructure is generally studied with low power CW light source (Surface Plasmon excitation wave length and its width). Here the major interest is studying the nanostructure response to light and excitation of surface plasmon resonance. White light contains a range of wavelengths in the visible range that is used to explore and excite the surface plasmon resonance in Gold, Copper Silver nanoparticles [1]. This has applications in surface enhanced Raman scattering, Nanoparticles for diagnosis and therapy etc. There is another interest amongst laser plasma physicist where ultrashort ultrahigh intensity light (I>10^{14-15}W/cm^2)is used for plasma formation [2]. This study is majorly done for efficient particle (electron ions and neutrons) and radiation generation (X-ray generation) [2]. The intermediate regime of light nanostructure interaction when the light is intense enough to cause ionization (10^{10}<I>10^{15}/cm^2) and onset of plasma formation is rarely explored in detail [3]. At lower intensity the nano-structure (having a size between 1-100 nm)is a solid state condensed matter. Absorption in such structures proceeds through the dielectric response of the material with material dielectric constant $\varepsilon=\varepsilon_1+i\varepsilon_2$. The real and imaginary parts depends on the plasma frequency, interband contribution, damping due to electron phonon scattering and size correction for damping due to surface scattering [1]. At higher intensity much beyond the plasma formation threshold, nanostructures are used as target as they offer a high absorption and high electric field enhancement as compared to planar solid target [4]. This is desirable since the Spitzer collision frequency for plasma υ (is proportional to $T_e^{-3/2}$) which reduce considerably at high temperature and plasma becomes collisionless [5]. Therefore nanostructures which have local solid density and offer resonance at higher density are used as target. The resonance density of planar solid is n_c and for sphere and cylinder it is much higher $3n_c$ and 2 n_c respectively [4,6]. Collisional damping term is considerably small both at low (T_e<<1 eV, I< 10^{10}W/cm^2) and high temperature (T_e>100eV, I>10^{15}W/cm^2) [3,7]. Surface Plasmon Resonance is therefore sharp and SPR peak are only weakly dependent on collision frequency. In the intermediate regime the nano particle undergoes a change from cold solid state to hot plasma state and collisional frequency is highly enhanced due to substantial electron phonon scattering and reasonably strong electron ion collision frequency [7]. Moreover The material assumes a metal like transition with a simplistic Drude like dielectric constant without interband transition contribution ($\varepsilon=1-n_e/n_c/(1+i\upsilon/\omega)$). Where υ/ω is the ratio of collision frequency to incident light frequency in c.p.s. SPR is therefore expected to be broadened and the peak also is expected to be strongly dependent on damping [8]. In this paper we report the electron density of SPR of various nanostructures of different geometrical shapes. We then derive the Heating rate of ultrashort intense pulse irradiated nanostructures for different collisional damping strength. It is found that increased damping leads to the maximum heating rate shifted to a higher electron density as compared to SPR electron density without collision term. Hollow nanostructures like nano-tubes and nano-shell, and pointed structures like nano-elliptic cylinder have a higher SPR electron density. Absorption rate can be highly enhanced at high electron densities in hollow and pointed nanostructures. Thus tunability in resonance can be achieved by choice of target geometry and varying the laser intensity which governs the collisions frequency (υ) that dramatically influence the SPR in vicinity of plasma formation.

2. ANALYTICAL MODEL

First we begin with assumptions of the model of nanostructure interacting with intense ultrashort pulses as a dielectric in laser field [9]. The dielectric constant is metal like ($\varepsilon=1-n_e/n_c/(1+i\upsilon/\omega)$). Where $\nu \approx 3\times10^{-6} \ln \Lambda n_e\left(\frac{Z}{T_e^{3/2}}\right)$ Z is degree of ionization, n_e is electron density T_e is electron temperature and the coulomb logarithm. $\varepsilon_1 = 1-n_e/n_c(1+(\upsilon/\omega)^2)$, and $\varepsilon_2 = \upsilon/\omega . n_e/n_c(1+(\upsilon/\omega)^2)$. For pulses <100 fs the geometry is assumed to be intact due to frozen hydrodynamic motion. Nanoparticle size << laser wavelength and laser skin depth (size is defined along the laser polarization) but larger than the Debye length. The Laplace equation is $\nabla^2 V = 0$ is solved ($n_e=n_i$) in spherical coordinate for spherical particle, cylindrical coordinates for cylindrical particle and elliptical coordinates for elliptical particle. By the continuity of tangential component of electric field and normal component of electric displacement vector the expression for Electric field inside E_{in} a nanostructure is evaluated. For hollow nanostructure the same procedure is repeated for both the inner and outer layer. For solid nanostructures (sphere, cylinder and ellipse) electric field is spatially uniform and along laser polarization. For hollow nanostructures the electric field is spatially varying spherically or azimuthally and thus the r.m.s electric field is calculated. Heating rate per unit volume of laser field driven dielectric is given by

$$\frac{\partial U}{\partial t} = \frac{\omega\varepsilon_0[\mathrm{Im}\,\varepsilon]}{2}\left|E_{in}\right|^2 \qquad (1)$$

Heating rate per unit laser intensity I ($1/2\varepsilon_0 E_0^2$) per unit laser c.p.s. ω is [Im ε]mod(E_{in}/E_0)^2 and the unit is 1/m.c.p.s. The colliional damping term is generally taken as υ/ω~1 as showm by Milchberg et al [3] for Al plasma for 10^{13}<I>10^{15}/cm^2 for 400 fs pulse in the resistivity saturation regime for 308 nm laser. Other values of damping are also taken to see its effect on SPR location and width. The lower value υ/ω=0.1 correspond to solid state cold bulk or high temperature hot plasma and υ/ω=10 corresponds to resistive saturation regime [3].

a) For nanosphere (NS)

$$\frac{\partial U}{I\omega\partial t} = \varepsilon_2\left|\frac{3}{\varepsilon+2}\right|^2 \qquad (2)$$

For collisionless plasma υ/ω=0, SPR occurs at $n_e = 3n_c$

b) For nanocylinder/nanorod (NC)

$$\frac{\partial U}{I\omega\partial t} = \varepsilon_2\left|\frac{2}{\varepsilon+1}\right|^2 \qquad (3)$$

For collision less plasma υ/ω=0, SPR occurs at $n_e = 2n_c$

c) For nanoelliptic cylinder (NEC)with a and b as its major and minor axix and incident laser polarization is along major axis (a)

$$\frac{\partial U}{I\omega\partial t} = \varepsilon_2\left|\frac{1+a/b}{\varepsilon+a/b}\right|^2 \qquad (4)$$

For collision less plasma υ/ω=0, SPR occurs at $n_e = n_c(1+a/b)$. Note that pointed elliptical cylinders will have a high SPR electron density.

d) For nanoshell with a and b as internal and external radii.

$$\frac{\partial U}{I\omega\partial t} = \varepsilon_2\frac{9\left|(2\varepsilon+1)^2+2(a/b)^3(\varepsilon-1)^2\right|}{\left|(2\varepsilon+1)(\varepsilon+2)-2(a/b)^3(\varepsilon-1)^2\right|^2} \qquad (5)$$

For collision less plasma υ/ω=0, SPR occurs at two densities,

$$n_e/n_c(H) = (9+3\sqrt{1+8a^3/b^3})/4(1-a^3/b^3)$$

$$n_e/n_c(L) = (9-3\sqrt{1+8a^3/b^3})/4(1-a^3/b^3)$$

e) For nanotube with a and b as internal and external radii.

$$\frac{\partial U}{I\omega\partial t}=\varepsilon_2\frac{4\left|(\varepsilon+1)^2+(a/b)^2(\varepsilon-1)^2\right|}{\left|(\varepsilon+1)^2-(a/b)^2(\varepsilon-1)^2\right|^2} \quad (6)$$

For collision less plasma υ/ω=0, SPR occurs at two densities, High density n_e/n_c (H)=2/(1-a/b) and low density n_e/n_c (L)=2/(1+a/b)
To see the effect of collisional damping on the heating of various nano particles and SPR peak location the equation (2)-(6) are plotted in Fig.(1) to Fig.(5) ($\frac{\partial U}{I\omega\partial t}$ vs $\frac{n_e}{n_c}$)

3. RESULTS AND DISCUSSIONS

Ionization occurs in laser matter interaction due to multiphoton processes, electron tunneling and electron impact ionization [7,8,10,11]. These depend on the laser intensity and plasma conditions and govern the collision frequency in nano structures. A sudden increase is therefore expected in the

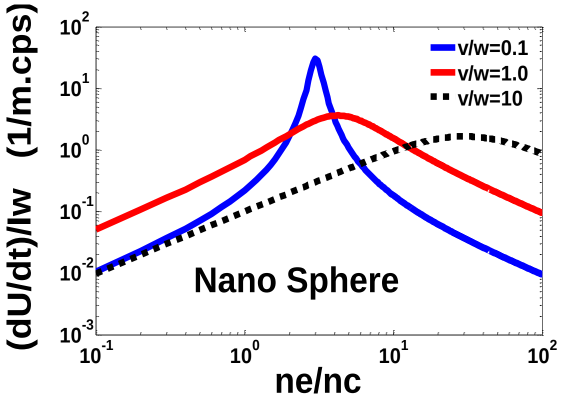

Fig-1. Heating rate per unit intensity per unit laser cycle per second as a function of electron density for nano-sphere with various damping term values υ/ω=0.1,1.0,10

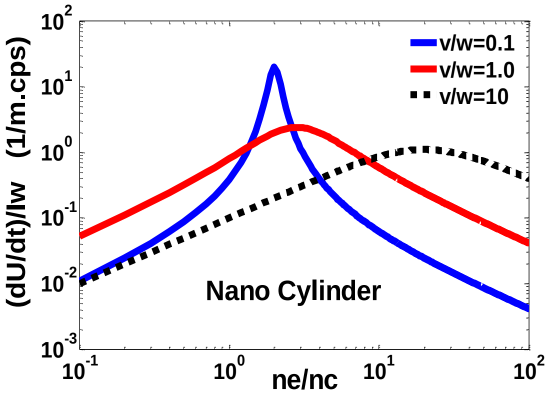

Fig-2. Heating rate per unit intensity per unit laser cycle per second as a function of electron density for nano-cylinder with various damping term values υ/ω=0.1,1.0,10

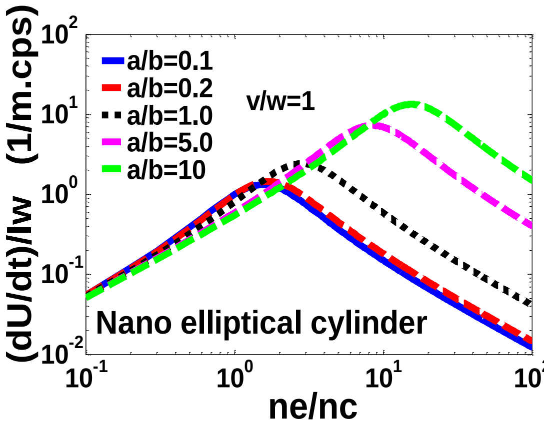

Fig-3. Heating rate per unit intensity per unit laser cycle per second as a function of electron density for nano-elliptical cylinder with various a/b values =0.1,0.2,1.0,5.0, and 10 for a fixed value of υ/ω=1

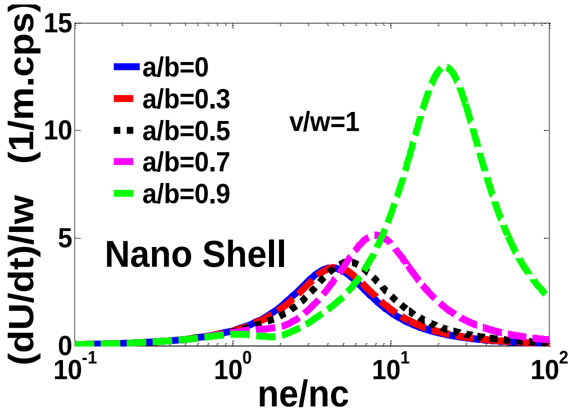

Fig-4. Heating rate per unit intensity per unit laser cycle per second as a function of electron density for nano- shell with various a/b values =0.0,0.3,0.5,0.7, and 0.9 for a fixed value of υ/ω=1

heating when ionization due to strong laser field transforms the nanoparticle from cold solid to cold plasma. Fig. (1) to (5) shows the heating rate per unit volume per unit laser intensity per unit laser frequency in c.p.s for a dielectric nano structure kept in laser field.

From Fig. (1) the electron density at which heating peaks up, occurs at higher density for stronger collisional nano plasma. For small damping the resonance is close to $3n_c$, the absorption is more than an order of magnitude high at high electron density for highly collisional plasma. Fig. (2) Also shows similar broad absorption over a wide range of densities and upshift of SPR electron density from $2n_c$ to a higher value for a larger damping term.

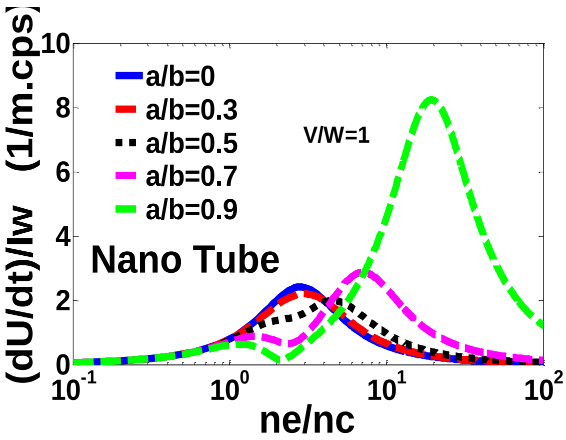

Fig-5. Heating rate per unit intensity per unit laser cycle per second as a function of electron density for nano- tube with various a/b values =0.0,0.3,0.5,0.7, and 0.9 for a fixed value of υ/ω=1

Fig. (3) Shows heating rate of nano elliptical cylinder with various ellipticity Electric field is perpendicular to cylinder axis. It is seen when a/b increase the absorption peak occurs at progressively high density. For a fixed value of damping term υ/ω=1the heating rate may become more than two magnitude higher at higher electron density for pointed structures oriented parallel to applied field.

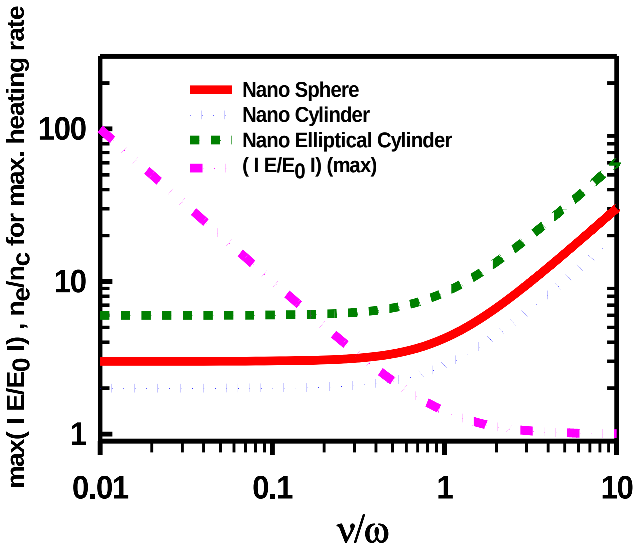

Fig-6. Maximum Field enhancement vs collision frequency in a solid nanostructure is independent of its shape shown by pink dash dot line. Electron density for maximum heating rate vs υ/ω for nano shhre , cylinder and ellipse.

Another class of nanostructures are hollow in nature having degree of hollowness a/b i.e. inner to outer radii ratio. Fig.(4) shows heating rate in a nano shell as a function of electron density for various degree of hollowness and a fixed collision frequency of υ/ω=1. For a nanoshell the two resonances are noted the low density resonance is inconsequentially small compared to the high density resonance. Shells with higher degree of hollowness show SPR at higher density, collisions further broaden the high density SPR. Collision also lead to peaking of absorption rate at a higher density as compared to the value predicted for a collisionless plasma. Similarly Fig.(5) shows heating rate in a nano tube as a function of electron density for various degree of hollowness and a fixed collision frequency of υ/ω=1. For nanotubes also the two resonances are noted the absorption in the vicinity of low density resonance is quite small compared to the high density resonance. Tubes with higher degree of hollowness show SPR at higher density, collisions broaden the high density SPR. The damping term due to electron ion collisions also lead to peaking of absorption rate at a higher density than the electron density for SPR predicted for a collisionless plasma with $\varepsilon=1-n_e/n_c$. If a high density solid state nano plasma matter is to be heated the nanostructure may be tailored to have desirable hollowness or should be pointed like ellipsoid or elliptical cylinder. Thus one may conclude that the solid nanoparticle get converted to hot plasma at a lower incident intensity since their SPR density is high as compared to the SPR density of planar solid formed plasma. In addition more than an order of magnitude higher heating rates is noted at high density in hollow nanostructures implying ablation threshold in such structures is expected to be further lower in comparison to solid nano-structures.

The electric field enhancement in solid nanostructures is $\left|\frac{E}{E_0}\right| = \frac{3}{|\varepsilon+2|}, \frac{2}{|\varepsilon+1|}, \frac{(a/b+1)}{|\varepsilon+a/b|}$ for nano sphere cylinder and elliptical cylinder. The electric field maximizes at $3n_c$, $2n_c$ and $(a/b+1)n_c$. The density at which field maximization occurs is independent of υ/ω. However the magnitude of maximum electric field enhancement at these densities depends on υ/ω and is given by the following equation.

$$\left|\frac{E}{E_0}\right|_{\max} = \sqrt{1+\frac{1}{(\nu/\omega)^2}} \qquad (7)$$

It is noteworthy that for these solid nanostructures the value of maximum field enhancement is independent of the shape. Fig. (6) shows the maximum field enhancement as a function of collision frequency using equation (7) (pink dash dot curve) The enhancement is huge when υ/ω is small and for large values of collision frequency the field amplification is close to one that is negligible. Maximum electric field enhancement occurs at density independent of collision frequency but the value of the field enhancement depends on the collision frequency shown in (Pink dash dot line). Clearly collision less plasma shows a high field enhancement and negligible enhancement is seen for highly collisional plasma (Note maximum field enhancement is independent of the geometrical shape of solid nano structures like spherical (NS) cylindrical (NC)or elliptical (NEC). As shown the density at which maximum heating occurs is a strong function of collision frequency. For a collisionless plasma its close to $3n_c$ and 2 n_c for nano spheres and nano cylinder but density for maximum heating drastically increase for higher damping present in highly collisional plasma accessible to the resistive saturation regime (10^{13}W/cm^2<I<10^{15}W/cm^2) [3]. Using equation (2),(3),(4), the density at which maximum heating occurs that is the SPR occurs can be found analytically given by the following equations (8),(9) and (10).

$$\text{NS } n_e / n_c \;{}_{\max heating} = 3\sqrt{1+(\nu/\omega)^2} \quad (8)$$

$$NC \;\; n_e / n_c \;{}_{\max heating} = 2\sqrt{1+(\nu/\omega)^2} \quad (9)$$

Fig. 6 shows graphically the density for maximum heating at SPR condition as function of collision frequency. Maximum

$$NEC \;\; n_e / n_c \;{}_{\max heating} = (1+a/b)\sqrt{1+(\nu/\omega)^2} \quad (10)$$

heating occurs at progressively higher densities for highly collisional plasma. Pointed structure has further high heating rates at high density [12]. So for practical applications like laser based field electron emission where high electric fields are desirable the laser heated matter should be either cold solid or hot plasma since they are practically collisionless. On the other hand if heating or energy coupling is desirable in matter that is dense and warm laser intensity should be such that equally efficient electron phonon collision and electron ion scattering must be present.

CONCLUSIONS

The SPR of nanoparticles with CW laser/ light is tuned by varying the incident light wavelength whereas for intense laser SPR tuning is done by varying electron density during the ionization. The following conclusions can be made on the basis of analysis done in this paper of ultrashort pulse irradiated nanostructure in the vicinity of plasma formation:

1. SPR peaks at enhanced electron density for hollow and pointed nanostructures. Absorption is broadened over a wide range of electron densities.
2. SPR peak of absorption or heating depend strongly on collision frequency (υ/ω is quite high around the plasma formation threshold).
3. Multiple resonance in hollow nano-structure, enhanced resonance density and strong collision frequency dependence of SPR must be taken in to account for moderate intensities present in the foot of intense pulses I>>10^{14}W/cm^2 as it affects the absorption dynamics for the subsequent coming high intensity around the peak of laser pulse. Laser heated hollow and pointed nanostructures can be efficient field emitters as they can be heated very efficiently at high density.
4. The critical role of target geometry like shape, size and degree of hollowness , as well as laser parameters is identified. Collision less nano plasma (at very low intensity I< 10^{10}W/cm^2 and very high intensity I> 10^{15}W/cm^2) support huge field enhancement. Highly collisional nano plasma formed in the intensity range(10^{13}W/cm^2<I<10^{15}W/cm^2) has negligible field enhancement but efficient heating at high densities.

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