Kinetic theory of magnetic absorption of laser irradiation by a metallic nanoparticles

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Abstract The theory for the magnetic field energy absorption by metallic nanoparticles subjected to the irradiation by ultrashort laser pulses of different duration in the region of surface plasmon excitation is developed. For the particles of the oblate or prolate spheroidal shape there has been found the dependence of the absorbed energy on a number of factors, including a particle radius, a degree of the shape deviation from a spherical one, a pulse duration, the orientation of the magnetic field upon particle, the magnitude of carrier frequency, and the value of a shift of the carrier frequency of a laser ray from the frequency of the surface plasmon excitation in a spherical particle. An appreciable absorption grows at the length of free electron pass large compared to the particle size is established.

1.Introduction

The study of the properties of metallic nanoparticles (MN) is not only of purely scientific interest but also of practical importance in certain applications. The research of optical properties of MN has a long history. In particular, the expression for the absorption cross-section of a plane electromagnetic wave with the frequency ω by a spherical nanoparticle, whose sizes are smaller than the radiation wavelength, has been known for a long time[1]. The most general and referenced theory of the optical properties of small particles is the Mie theory [2]. This theory was developed for spherical particles in the assumption that the vector of the electric current density inside the particle, *j(r, t)*, is related to the generating field *E(r, t)* by the Ohm law. The solution of the problem to find the current density vector j(r, *t*) responsible for the energy absorption includes two stages. At the first stage, we should determine internal fields generated by wave in the nanoparticle. At the second stage, we should determine how those internal fields change the velocity distribution function of electrons, i.e. find a correction to the equilibrium Fermi distribution induced by the internal fields. The internal fields induced by wave in the nanoparticle depend on the nanoparticle shape. Therefore, we will assume that the nanoparticle has an ellipsoidal shape. It is convenient to develop a theory for this form, because the results obtained for the ellipsoidal form can be extended to a wide range of nanoparticle forms by changing the curvature radii of the ellipsoid. If the wave length λ much exceeds the nanoparticle size, then the relationship of the internal and external fields, E_0 and H_0, is known .

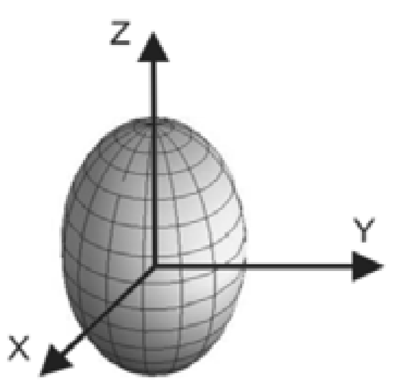
2. Theoretical model

Let a metal particle be in the field of an external electromagnetic wave (laser irradiation)

$$\begin{pmatrix} \vec{E} \\ \vec{H} \end{pmatrix} = \begin{pmatrix} \vec{E}_0 \\ \vec{H}_0 \end{pmatrix} e^{i\left(\vec{k}\vec{r}-\omega t\right)}, \qquad (1)$$

where $\vec{E}_0$ and $\vec{H}_0$ are the electric and magnetic, respectively, components of the wave field; and k is the wave vector ($k = 2\pi/\lambda$, where λ is the wave length). Then the solution of the problem to find the current density vector , responsible for the energy absorption includes two stages. At the first stage we should determine internal fields generated by wave in the nanoparticle $\vec{j}(r,t)$ i.e. At the second stage, we should determine how those internal fields change the velocity distribution function of electrons, i.e. find a correction to the equilibrium Fermi distribution induced by the internal fields. The internal fields induced by wave (1) in the nanoparticle depend on the nanoparticle shape. Therefore, we will assume that the nanoparticle has an ellipsoidal shape.

Figs. 1 and 2 schematically illustrate these two types of rotational ellipsoids.



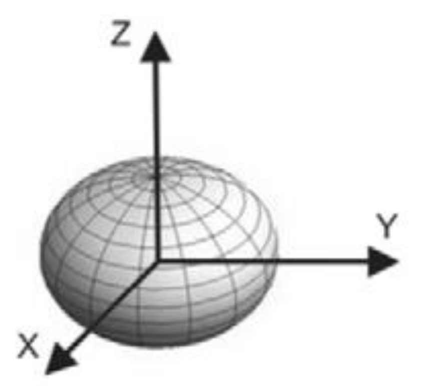


Fig.1. Elongated biaxial spheroid(a>b=c). Fig.2. Flattened biaxial spheroid(a=b>c).

In the case of an elongated spheroid (Fig. 1), its two small half-axes are equal to each other (b = c), whereas in the case of a flattened spheroid (Fig. 2) its two large half-axes are equal (a = b). If you introduce the designation $R_{\parallel}$ for the large half-axis, and $R_{\perp}$ for the small half-axis of ellipsoid, then in the case of elongated spheroid $R_{\parallel} = a > R_{\perp} = b = c$ and the flattened $R_{\parallel} = c > R_{\perp} = b = a$. looking for the solutions of the equations, we obtained the following expressions

Energy of magnetic absorption by a spheroidal metal nanoparticle brings us to the next expression

$$W_m = \frac{9V}{128}\frac{ne^2 v_F R_{\perp}}{mc^2}\left\{\Phi_1^m\left(H_{\parallel}^0\right)^2 + \Phi_2^m\left(\frac{R_{\parallel}^2}{R_{\perp}^2+R_{\parallel}^2}\right)^2\left(H_{\perp}^0\right)^2\right\} \qquad (2)$$

In the case of spherical nanoparticles, $R_{\perp} = R_{\parallel} = a$ $\varepsilon_p \to 0$, $\Phi_1^m = 2/3$, $\Phi_2^m = 8/3$ we obtain the next result [7]:

$$W_m^0 = \frac{3}{64} Va \frac{ne^2 v_F}{mc^2}\left(H^0\right)^2. \qquad (3)$$

While studying the dependence of the shape of nanoparticles on the energy absorption convenient to take the ratio between the energy absorbed by a spheroidal particle W_m and the energy absorbed by a spherical particle W_m^0 with the same volume, we obtain the next expression:

$$\frac{W_m}{W_m^0} = \frac{3}{2}\left(\frac{R_{\perp}}{R_{\parallel}}\right)^{1/3}\left[\Phi_1^m \cos^2\theta + \frac{\Phi_2^m \sin^2\theta}{\left[1+\left(R_{\perp}/R_{\parallel}\right)^2\right]^2}\right] \qquad (4)$$

3. Results of computation calculation and discussion

Here we present the results of computation calculation and discuss them. It is of interest to study influence the effect of the orientation of the magnetic field vector of the incident wave $\vec{H}_0$ with respect to the spheroid axes on the process of magnetic absorption of light (laser irradiation) in metal nanoparticles. Figures 3 show the dependences of the ratio W_m/W_m^0 for several values of the angle between the $\vec{H}_0$ vector and the spheroid semiaxis.

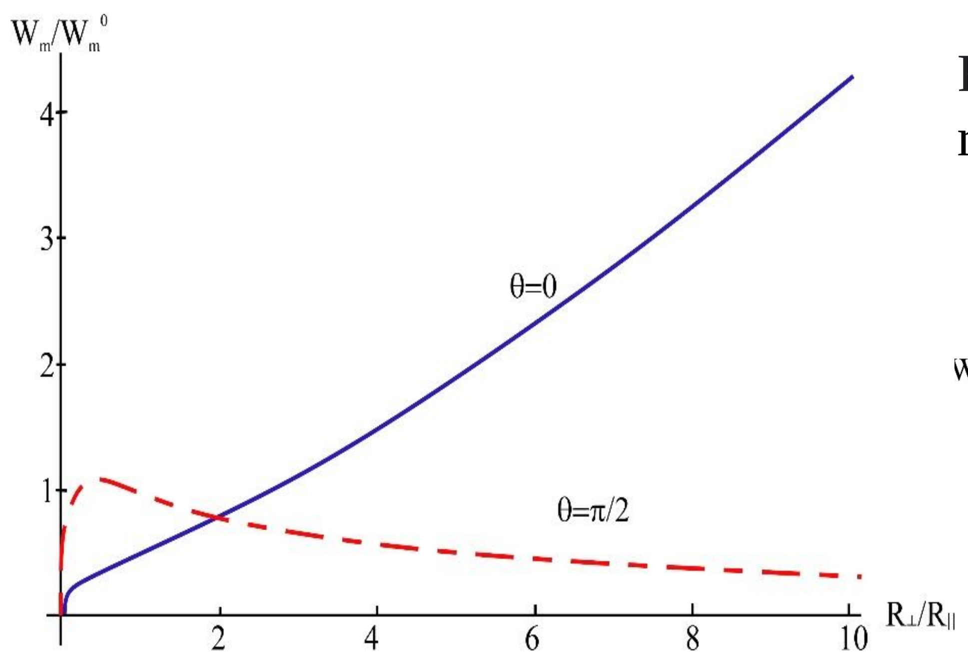


Fig.3. Dependence of the W_m/W_m^0 ratio on the ratio $R_{\perp}/R_{\parallel}$ at θ=0 and θ=π/2.

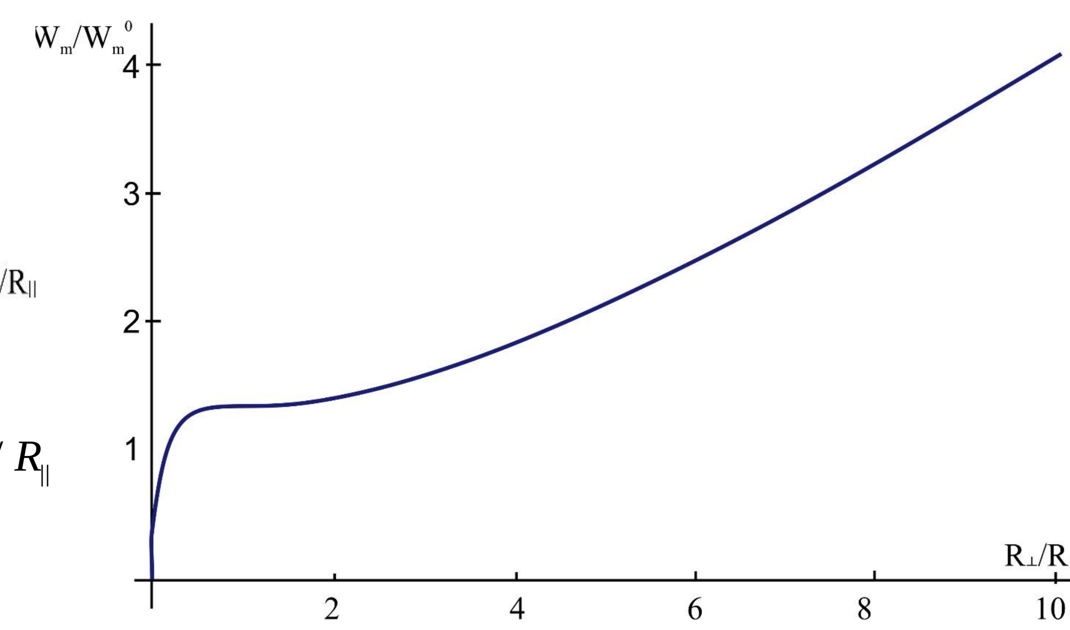


Fig.4. The dependence of the average ratio $\langle W_m/W_m^0\rangle$ on the spheroidal ratio $R_{\perp}/R_{\parallel}$.

4. Conclusion

The kinetic theory of the magnetic light absorption by a metal nanoparticle with regard to the influence of a nanoparticle shape on this process is constructed. In particular, the effect of spheroidal nanoparticles is an alyzed. It is convenient to develop the theory for the ellipsoidal form, because the results obtained for this form can be extended to a wide range of nanoparticle forms (from discoid to rod-like ones), The curvature radii of the ellipsoid are varied in this case by means of only the ratio between them, , because the nanoparticle volume remains constant. The main results obtained in this work are as follows.

1.An expression for the ratio between the energies absorbed by the spheroidal and spherical particles (with the same volumes) is derived, which is suitable for applications. 2. An expression is obtained for the calculation of the averaged value of the ratio between the magnetic absorption by the spheroidal and spherical metal nanoparticles in terms of the ratio between the curvature radii of the spheroid $R_{\perp}/R_{\parallel}$. 3.·Also contains the results of computational experiments. The most interesting result of the latter is the growth of the energy absorption by the spheroidal nanoparticle with the growth of $R_{\perp}/R_{\parallel}$.(its disk-like character) at an arbitrary value of the angle $\theta \in \{[0,\pi/2) \cup (\pi/2,\pi]\}$. The only value, at which the curve $W_m(R_{\perp}/R_{\parallel},\theta)/W_m^0$ asymptotically approaches zero, is $\theta = \pi/2$. 4. The magnetic absorption in the frequency range(4), where it can be substantial

On the other hand, the magnetic absorption in the frequency range (4), where it can be substantial does not depend on the frequency at all.

References

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