Chemistry, Physics and Technology of Surface. 2023. V. 14. N 4. P. 504-511

PACS 78.67.Bf doi: 10.15407/hftp14.04.504

O.Yu. Semchuk, O.O. Havryliuk, A.A. Biliuk

KINETIC THEORY OF MAGNETIC ABSORPTION OF LASER IRRADIATION BY NANOPARTICLES

Chuiko Institute of Surface Chemistry of National Academy of Sciences of Ukraine
17 General Naumov Str., Kyiv, 03164, Ukraine, E-mail: philosoph.tb@gmail.com

When metal nanoparticles (MNPs) are illuminated with a monochromatic laser wave, the frequency of which is far from the plasmon frequency (the frequency of plasmon resonances), under certain conditions (depending on the frequency of the wave, its polarization, the size and shape of the MNPs), absorption of light by MNPs can be dominated by magnetic absorption (absorption caused by the magnetic component of the electromagnetic field of the light (laser) wave). This work is focused on studying the features of absorption caused by the influence of the magnetic component of laser radiation. This issue is rather poorly studied for MNPs of non-spherical shape. Therefore, how the shape of the particle manifests itself in its absorption of laser radiation (laser pulses) is one of the goals of our research. In this work, we will study the features of magnetic absorption of light (laser radiation) depending on the shape of the particles. In this paper, we will investigate the influence of spheroidal MNPs on this process. Calculations will be carried out using the kinetic equation method, because we will consider the case when the size of the MNP is smaller than the length of free path of the electron in the MNP. Note that the kinetic approach makes it possible to obtain correct results for the case when the size of the particle is greater than the length of the free path. For non-spherical MNPs, we have developed a theory that makes it possible to calculate the energy of magnetic absorption by a particle when it is irradiated with laser pulses. The dependence of magnetic absorption on the ratio of the radii of curvature of spheroidal MNPs and the vector of the magnetic field of an electromagnetic (laser) wave was constructed and theoretically investigated. An interesting result is the absorption of energy by a spheroidal MNP as its disco similarity increases. We now use to estimate the relative contributions of electric W_e and magnetic W_m absorption to the total absorption. For example, let us take a gold MNP's, then $\omega_p \approx 5\cdot10^{15}$ s^{-1}, $\nu \approx 10^{13}$ s^{-1}, $R = 3\cdot10^{-6}$ sm, $\omega \approx 2\cdot10^{14}$ s^{-1} (carbon dioxide laser), $\varepsilon' \approx -600$, $\varepsilon'' \approx 30$ we received the next ratio $W_e/W_m \approx 2$. We can see that for the given set of parameters magnetic absorption is twice as large as electric. Obviously, for different parameters of the particle and a different frequency range electric absorption can be either larger or smaller than magnetic absorption. Hence, when studying the dependence of optical absorption by MNP's on particle form, we must allow for both electric and magnetic absorption. For an asymmetric MNP's (for example ellipsoidal particles), apart from everything else, the ratio of the electric and magnetic contributions to absorption (as fixed frequency) is strongly dependent on the degree of particle asymmetric and wave polarization.

***Keywords**: electromagnetic wave, kinetic theory, magnetic absorption, metallic nanoparticles, electron, plasmon, frequency*

INTRODUCTION

The study of the properties of MNP's is not only of purely scientific interest but also of practical importance in certain applications. The research of optical properties of MNP's has a long history [1–4]. In particular, the expression for the absorption cross-section of a plane electromagnetic wave with the frequency ω by a spherical nanoparticle with sizes smaller than the radiation wavelength, has been known for a long time [5]. The most general and referenced theory of the optical properties of small particles is the Mie theory [6]. 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. First, we need to determine the fields generated by the incident external wave during interaction with the particle. Then we study how the electron velocity distribution function changes due to internal fields, that is, we find a correction to the equilibrium Fermi distribution. The shape of the particle affects the shape of the internal fields. Therefore, we will assume that the nanoparticle has an ellipsoidal shape. This form is convenient for the development of theories, and the results can be extended to other ellipsoidal particles by changing only the radius of curvature of the particles. 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 [5]. The goal of this study is to examine in detail the magnetic absorption of MNP's of ellipsoidal form for arbitrary orientation of the particle with respect to the direction of propagation of the incidence wave at frequencies both above and below the characteristic frequency of free passage of the electron between walls of the particle.

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), ω is the frequency of the incident wave, t is the time of the incident wave, $\vec{r}$ is the spatial coordinate. Then the solution of the problem to find the current density vector $\vec{j}(\vec{r},t)$ responsible for the energy absorption includes two stages. At the first stage, we should determine internal fields generated by wave (1) 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 (1) in the nanoparticle depend on the nanoparticle form. In this paper the nanoparticles with ellipsoidal form will be analysed. 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 (from discoid to rod-like ones) by changing the curvature radii of the ellipsoid.

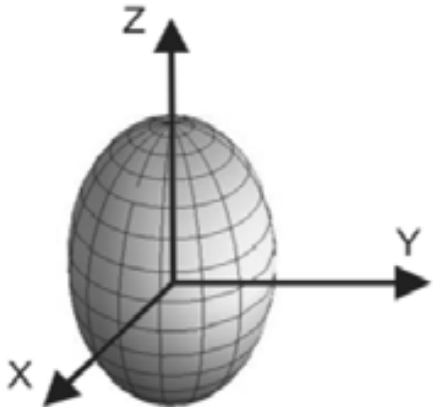

Fig. 1. Elongated biaxial spheroid (z > (x = y))

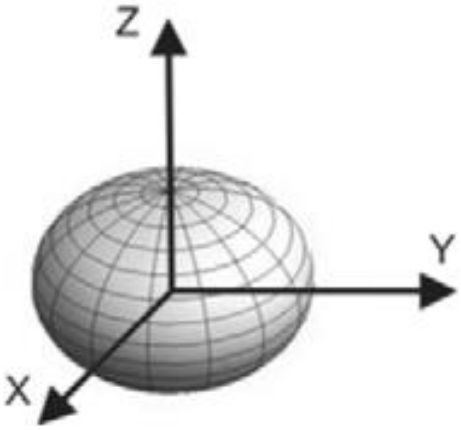

Fig. 2. Flattened biaxial spheroid (z (x = y))

From the viewpoint of practical application, nanoparticles in the form of elongated or flattened spheroids – which are formed by the rotation of an ellipse around a short or long axis – are of greatest interest. Figs. 1 and 2 schematically illustrate these two types of rotational ellipsoids.

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 $R_{\|}$ for the large half-axis, and $R_{\perp}$ for the small half-axis of ellipsoid, then in the case of elongated spheroid $R_{\|} = z > R_{\perp} = x = y$ and the flattened $R_{\|} = z < R_{\perp} = x = y$.

The electric field of the laser pulse induces inside the metal nanoparticle a potential electric field $\vec{E}_{in}(\vec{r},t)$, and a magnetic vortex electric field $\vec{E}_{vr}(\vec{r},t)$. If the characteristic size of the nanoparticle R satisfies of the inequality $k_0R << 1$ holds, then in this case the Fourier coordinate dependence of the components of the electric and magnetic fields of the laser wave can be neglected [7]. For an ellipsoidal nanoparticle,

this allows us to obtain the following expressions for internal potential $E_{in}^{j}(0,\omega)$ and vortex $E_{vr}^{j}(0,\omega)$ electric fields as [7]:

$$E_{in}^{j}(0,\omega)=\frac{E_0^{j}(0,\omega)}{1+L_j\left[\varepsilon(\omega)-1\right]}, \tag{2}$$

$$E_{vr}^{x}=i\frac{\omega R_z^2}{c}\left[\frac{z}{R_x^2+R_z^2}H_0^{y}(0,\omega)-\frac{y}{R_x^2+R_y^2}H_0^{z}(0,\omega)\right]. \tag{3}$$

Here $\varepsilon(\omega)=\varepsilon'(\omega)+i\varepsilon''(\omega)$ is the complex dielectric constant of the metal nanoparticle, L_j is the geometric factors (depolarization coefficients) [5], $j = x, y, z$, R_x, R_y, R_z is the half-axis of the ellipsoid in the x, y and z directions, respectively.

The internal fields $\vec{E}_{in}(\vec{r},t)$ and $\vec{E}_{vr}(\vec{r},t)$ induce the corresponding currents $\vec{j}_{in}(\vec{r},t)$ and $\vec{j}_{vr}(\vec{r},t)$. Therefore, the total energy absorbed by the metal nanoparticle will be equal to [5]

$$W=\int_{-\infty}^{\infty}w(t)dt=W_e+W_m=$$

$$=\frac{1}{2}\mathrm{Re}\int_{-\infty}^{\infty}dt\int_{V}d\vec{r}\left[\vec{j}_e(\vec{r},t)\vec{E}_{in}^{*}(\vec{r},t)+\vec{j}_{vr}(\vec{r},t)\vec{E}_{vr}^{*}(\vec{r},t)\right] \tag{4}$$

where V is the volume of the nanoparticle, $w(t)$ is the power absorbed by the nanoparticle.

The absorption caused by the field $\vec{E}_{in}(\vec{r},t)$ is called electric (W_e), and the absorption caused by the field $\vec{E}_{vr}(\vec{r},t)$ is called magnetic (W_m). Turning to the Fourier representation, relation (8) can be rewritten as follows:

$$W=\frac{1}{4\pi}\int_{-\infty}^{\infty}d\omega\,\mathrm{Re}\int_{V}d\vec{r}\left[\vec{j}_e(\vec{r},\omega)\vec{E}_{in}^{*}(\vec{r},\omega)+\right.$$

$$\left.+\vec{j}_{vr}(\vec{r},\omega)\vec{E}_{vr}^{*}(\vec{r},\omega)\right]. \tag{5}$$

We will consider only the absorption associated with the action of the magnetic field of the incident electromagnetic wave, i.e., the so-called magnetic absorption. In this case Eg. (5) will be rewritten as follows:

$$W_m=\frac{1}{4\pi}\int_{-\infty}^{\infty}d\omega\,\mathrm{Re}\int_{V}d\vec{r}\,\vec{j}_{vr}(\vec{r},\omega)\vec{E}_{vr}^{*}(\vec{r},\omega). \tag{6}$$

In the general case, the current $\vec{j}(\vec{r},\omega)$ at point $\vec{r}$ inside the nanoparticle caused by the internal fields $\vec{E}_{in}(0,\omega)$ and $\vec{E}_{vr}(\vec{r},\omega)$, can be written as follows [7]:

$$\vec{j}(\vec{r},\omega)=2e\left(\frac{m}{2\pi\hbar}\right)^3\int_{-\infty}^{\infty}\vec{v}f_1(\vec{r},\vec{v},\omega)d\vec{v}, \tag{7}$$

where e is the charge of the electron, m is the mass of the electron, $f_1(\vec{r},\vec{v},\omega)$ is the Fourier component of the nonequilibrium electron distribution function in the nanoparticle which serves as an additive to the equilibrium Fermi distribution function $f_0(\varepsilon)$. The Fourier component of the nonequilibrium electron distribution function which given as

$$f_1(\vec{r},\vec{v},\omega)=\int_{-\infty}^{\infty}f_1(\vec{r},\vec{v},t)\exp(i\omega t)dt, \tag{8}$$

can be found as a solution of the corresponding linearized Boltzmann kinetic equation [8], which will be present at the following form:

$$(\gamma-i\omega)f_1(\vec{r},\vec{v},\omega)+\vec{v}\frac{\partial f_1(\vec{r},\vec{v},\omega)}{\partial\vec{r}}+$$

$$+e\vec{v}\left[\vec{E}_{in}(\omega)+\vec{E}_{vr}(\vec{r},\omega)\right]\frac{\partial f_0(\varepsilon)}{\partial\varepsilon}=0. \tag{9}$$

Here γ is the collisions frequency of electrons in the particles bulk.

Equation (9) must be supplemented by appropriate boundary conditions. As such, we choose the condition of diffuse reflection of electrons from the inner surfer of the nanoparticle

$$f_1(\vec{r},\vec{v},\omega)\big|_S=0,\quad v_n<0. \tag{10}$$

In (10), v_n is the normal to the surface S component of the electron velocity. The substantiation of such boundary conditions can be found, in particular, in [7]. Equation (9) is easiest to solve by moving to new variables:

$$x_i'=x_i\frac{R}{R_i},\quad v_j'=v_j\frac{R}{R_i}, \tag{11}$$

in which the ellipsoid takes the form of a sphere equal in volume to a radius $R=\sqrt[3]{R_{\parallel}R_{\perp}^2}$. Solved the partial differential equation (9) by the

method of characteristics, we can obtain a solution for the function $f_1(\vec{r},\vec{v})$ in the form [7]

$$f_1(\vec{r},\vec{v}) = -e\frac{\partial f_0}{\partial \varepsilon}\left[\vec{v}\vec{E}_{in} + \sum_{j=1}^{3}\alpha_{ij}v_i\left(\frac{x'_j R_j}{R} + \right.\right.$$

$$\left.\left. + v\frac{\partial}{\partial(\gamma - i\omega)}\right)\right]\left(\frac{1-\exp\left[-(\gamma - i\omega)t'\right]}{\gamma - i\omega}\right). \qquad (12)$$

The parameter $t' = \left(1/v'^2\right)\left[\vec{r}'\vec{v}' - \right.$ $\left. - \sqrt{\left(R^2 - r'^2\right)v'^2 + \left(\vec{r}'\vec{v}'\right)^2}\right]$ characterises the position of electron along the trajectory $\vec{r}' = \vec{v}'t' + \vec{R}$. The radius vector $\vec{R}$ gives the position of the point on the surface at which the trajectory begins (at $t' = 0$). We recall that after the deformation (14) the ellipsoidal form of the particle becomes spherical with radius *R*.

We will assume that the main mechanism of electron scattering in metal nanoparticles is electron scattering on the surface of the nanoparticle. In this case, it is interesting to consider the case of high-frequency $\omega >> \omega_s$ magnetic absorption $\omega_s = v_F/2R$ is the frequency of electron oscillations between the walls of the nanoparticle), because the vortex electric field $\vec{E}_{vr}$ is proportional to frequency and therefore the relative role of magnetic absorption increases with increasing frequency.

The substitution of Eq. (12) into Eq. (6) and (7) for the energy of magnetic absorption by a spheroidal metal nanoparticle brings us to the next expression [7]:

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

$$\left. + \Phi_2^m\left(\frac{R_\parallel^2}{R_\perp^2 + R_\parallel^2}\right)^2\left(H_\perp^0\right)^2\right\}. \qquad (13)$$

Here, v_F is the Fermi velocity, and $H_\parallel^0$ and $H_\perp^0$ are the amplitudes of the magnetic field components of the electromagnetic wave ($H_\perp = \sqrt{H_x^2 + H_y^2}$ as well as $H^0 = E^0$) that are parallel $(\parallel)$ and orthogonal $(\perp)$, respectively, to the axis of the ellipsoidal. Here Φ_1^m and Φ_2^m - some functions of the eccentricity of the ellipsoid $\varepsilon_p^2 = \left|1 - R_\perp^2/R_\parallel^2\right|$ [7, 9]. The function Φ_1^m looks like this [9]:

$$\Phi_1^m = \frac{1}{2}\int_0^{\pi/2}\partial\tau\sin^3\tau\sqrt{\sin^2\tau + (R_\perp/R_\parallel)^2\cos^2\tau} \qquad (14)$$

where τ is the angle between the axis of rotation of the spheroid and the direction of the electron's velocity.

As for the function Φ_2^m, for the two extreme cases of strongly elongated $(R_\perp < R_\perp)$ and strongly flattened $(R_\perp < R_\perp)$ ellipsoids of rotation, it has the following form [7]:

$$\Phi_2^m = \begin{cases} -\frac{1}{4\varepsilon_p^2}\left(1 - 8\varepsilon_p^2 + 4\varepsilon_p^4\right)\sqrt{1-\varepsilon_p^2} + \frac{1}{4\varepsilon_p^3}\left(1 + 2\varepsilon_p^2\right)\arcsin\varepsilon_p, & R_\perp/R_\parallel < 1, \\ \frac{1}{4\varepsilon_p^2}\left(1 + 8\varepsilon_p^2 + 4\varepsilon_p^4\right)\sqrt{1+\varepsilon_p^2} - \frac{1}{4\varepsilon_p^3}\left(1 - 2\varepsilon_p^2\right)\ln\left(\varepsilon_p + \sqrt{1+\varepsilon_p^2}\right), & R_\perp/R_\parallel > 1. \end{cases} \qquad (15)$$

In the case of spherical nanoparticles, $R_\perp = R_\parallel = a$, $\varepsilon_p \to 0$, $\Phi_1^m = 2/3$, $\Phi_2^m = 8/3$. Using those parameters from the Eq. (13), we obtain the next result [7]:

$$W_m^0 = \frac{3}{64}Va\frac{ne^2 v_F}{mc^2}\left(H^0\right)^2. \qquad (16)$$

While studying the dependence of the form of nanoparticles on the energy absorption it is 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. From the (13) and (15) we obtain the next expression:

$$\frac{W_m}{W_m^0}=\frac{3}{2}\left(\frac{R_\perp}{R_\parallel}\right)^{1/3}\left[\Phi_1^m\cos^2\theta+\frac{\Phi_2^m\sin^2\theta}{\left[1+\left(R_\perp/R_\parallel\right)^2\right]^2}\right] \qquad (17)$$

(θ is the angle between the vector $\vec{H}^0$ and the spheroid axis).

Note that we consider the frequency interval

$$\gamma << \omega << \omega_l \ \left(l=\perp,\parallel\right) \qquad (18)$$

For typical metals, the frequency $\gamma \sim 10^{13}$ s^{-1}. The inequality $\nu \ll \omega$ allows us to neglect the bulk scattering of electrons. The inequality $\omega << \left(\omega_\perp, \omega_\parallel\right)$ means that we are far from plasma resonances, and the electric absorption does not "hide" the magnetic one.

Now let's return to electrical absorption. From relation (5) for electromagnetic absorption, we obtain the following expression:

$$W_e=\frac{1}{4\pi}\int_{-\infty}^{\infty}d\omega\,\mathrm{Re}\int_V d\vec{r}\left[\vec{j}_e\left(\vec{r},\omega\right)\vec{E}_{in}^{*}\left(\vec{r},\omega\right)\right]. \qquad (19)$$

Next, following the method outlined above, with the linearized Boltzmann kinetic equation, we find the Fourier component of the no equilibrium electron distribution function due to the action of the electric field of an electromagnetic (laser) wave:

$$f_1\left(\vec{r},\vec{v}\right)=-e\frac{\partial f_0}{\partial\varepsilon}\left[\vec{v}\vec{E}_{in}+\sum_{j,i=1}^{3}\alpha_{ij}v_i\left(\frac{x_j'R_j}{R}+\nu\frac{\partial}{\partial\left(\gamma-i\omega\right)}\right)\right]\left(\frac{1-\exp\left[-\left(\gamma-i\omega\right)t'\right]}{\gamma-i\omega}\right); \qquad (20)$$

t' can be formally considered as the "time" of the electron's movement along a characteristic trajectory, α_{ij} is the constant of the expansion of the intensity of the generated electric magnetic vortex electric field into the Fourier series [7]

Further calculations were carried out according to the methodology given in the paper [9]. This work considered the case when the frequency of a monochromatic electromagnetic wave is far from the frequency of plasmon resonances. In this case, depending on the size and shape of the MNP, as on well as the frequency and polarization of the wave, magnetic absorption will be dominant. For example, for gold particles with a size of 6 μm, the magnetic absorption will be twice the electric absorption at the frequency $2\cdot10^{14}$ s^{-1}. Therefore, the final formula takes the form:

$$W_e=V\frac{9}{16}\frac{ne^2}{m\omega^2}\frac{v_F}{R_\perp}\left[\varphi_\perp\left|E_\perp^0\right|^2+\varphi_\parallel\left|E_\parallel^0\right|^2\right], \qquad (21)$$

where $\varphi_\perp$ and $\varphi_\parallel$ is the function of the $\varepsilon_p^2=\left|1-R_\perp^2/R_\parallel^2\right|$:

$$\varphi_\perp=\frac{1}{2}\begin{cases}\frac{1}{2}\left(1+\frac{1}{2e_p^2}\right)\sqrt{1-e_p^2}+\frac{1}{e_p^2}\left(1-\frac{1}{4e_p^2}\right)\arcsin e_p, & R_\perp<R_\parallel\\ \frac{1}{2}\left(1-\frac{1}{2e_p^2}\right)\sqrt{1+e_p^2}+\frac{1}{e_p}\left(1+\frac{1}{4e_p^2}\right)\ln\left(\sqrt{1+e_p^2}+e_p\right), & R_\perp>R_\parallel\end{cases} \qquad (22)$$

$$\varphi_\parallel=\begin{cases}\frac{1}{2}\left(1-\frac{1}{2e_p^2}\right)\sqrt{1-e_p^2}+\frac{1}{4e_p^3}\arcsin e_p, & R_\perp<R_\parallel\\ \frac{1}{2}\left(1+\frac{1}{2e_p^2}\right)\sqrt{1+e_p^2}+\frac{1}{4e_p^2}\ln\left(\sqrt{1+e_p^2}+e_p\right), & R_\perp>R_\parallel\end{cases} \qquad (23)$$

Now we will compare the contribution of electric and magnetic absorption. For simplicity, let's do it for spherical MNP's. For MNP's, the dimensions of which are both larger and smaller than the free path length of the electron in the MNP's, the total energy of electric and magnetic absorption, as can be seen from (13) and (21), for spherical MNP's will be equal to.

$$W = \frac{9}{8\pi} V \omega \varepsilon'' \left[\frac{1}{\left(2+\varepsilon'\right)^2} + \frac{\omega^2 R^2}{90c^2} \right] \left| \vec{E}_0 \right|^2, \qquad (24)$$

where ε' and ε'' are , respectively, the real and imaginary parts of the dielectric constant, R is the particle radius and c is the speed of light. The first time in Eq. (24) describes electric absorption (W_e) and the second term magnetic absorption (W_m). If the particles is larger than mean free party, i.e., bulk scattering is dominant, the expression for the dielectric constant of the metal has standard form

$$\varepsilon_0 = \varepsilon' + i\varepsilon'' = 1 - \frac{\omega_p^2}{\omega^2 + \nu^2} + i \frac{\nu \omega_p^2}{\omega\left(\omega^2 + \nu^2\right)}, \qquad (25)$$

where ω_p is the plasma frequency, and ν is the collision rate.

We now use Eq. (24) and (25) to estimate the relative contributions of electric and magnetic absorption to the total absorption. For example, let us take a gold MNP's, then $\omega_p \approx 5 \cdot 10^{15}$ s^{-1}, $\nu \approx 10^{13}$ s^{-1}, $R = 3 \cdot 10^{-6}$ sm, $\omega \approx 2 \cdot 10^{14}$ s^{-1} (carbon dioxide laser), $\varepsilon' \approx -600$, $\varepsilon'' \approx 30$ from (24) we received the next ratio

$$\frac{W_e}{W_m} \simeq \frac{1}{90} \left(\frac{\omega R}{c} \right) \left| \varepsilon_0 \right|^2 \approx 2. \qquad (26)$$

From (25) we can see that for the given set of parameters magnetic absorption is twice as large as electric. Obviously, for different parameters of the particle and a different frequency range electric absorption can be either larger or smaller than magnetic absorption. Hence, when studying the dependence of optical absorption by MNP's on particle form, we must allow for both electric and magnetic absorption. For an asymmetric MNP's (for example ellipsoidal particles), apart from everything else, the ratio of the electric and magnetic contributions to absorption (as fixed frequency) is strongly dependent on the degree of particle asymmetric and wave polarization.

RESULTS OF COMPUTATION CALCULATION AND DISCUSSION

Here we present the results of computation calculation and discuss them. In the calculations, equation (17) was used for Fig. 3. Equation (27) was used for Fig. 4. The equation (18) was the frequency range. They were performed in Wolfram Mathematics with a step of 0.1, with subsequent construction of graphs in Origin. 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. Fig. 3 show the dependences of the ratio W_m / W_m^0 for several values of the angle between the vector $\vec{H}^0$ and the spheroid semiaxis.

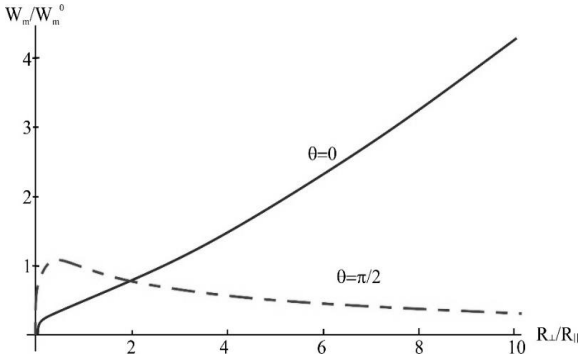

Fig. 3. Dependence of the ratio W_m / W_m^0 on the ratio $R_\perp / R_\parallel$ at θ=0 and θ=π/2

In obtaining (17) we used the fact that $H_\parallel^0 = H^0 \cos\theta, H_\perp^0 = H^0 \sin\theta$ (θ is the angle between the vector $\vec{H}^0$ and the spheroid axis). Therefore, we assume that spheroidal metal nanoparticles are chaotically oriented in the dielectric matrix we must averaging over orientations of the spheroid axis Eq. (17). After this we assume

$$\left\langle \frac{W_m}{W_m^0} \right\rangle = \frac{4}{3} \left(\frac{R_\perp}{R_\parallel} \right)^{1/3} \left[\Phi_1^m + \frac{\Phi_2^m}{\left[1 + \left(R_\perp / R_\parallel \right)^2 \right]^2} \right]. \qquad (27)$$

Fig. 4 shows dependence of the average ratio $\left\langle W_m / W_m^0 \right\rangle$ average value in parentheses on the spheroidal semiaxes ratio $R_\perp / R_\parallel$.

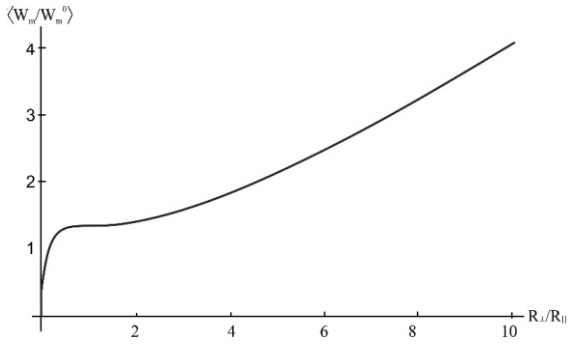

Fig. 4. The dependence of the average ratio $\left\langle W_m / W_m^0 \right\rangle$ on the spheroidal ratio $R_\perp / R_\parallel$

The Fig. 4 exhibits the dependence of the average ratio $\left\langle W_m / W_m^0 \right\rangle$ on the spheroidal semiaxes ratio $R_\perp / R_\parallel$. From Fig. 4, the following conclusion can be drawn. The more oblate the spheroidal metal nanoparticle, the larger is its averaged magnetic absorption. This growth has a smooth character at $R_\perp / R_\parallel > 0.5$.

From the analysis of Fig. 3 it follows that depending on the angle θ between the vector and the spheroid axis, the nature of the magnetic absorption changes. In particular, the maximum growth of magnetic absorption by a spheroidal metal nanoparticle is observed at two values of the angle θ: 0 and π. In addition, if a very oblate (almost flat) particle is oriented normally to the magnetic field vector (θ = 0 or π), then the effect of its interaction with this field is maximum. But if the same particle (almost flat) is oriented along the magnetic field vector ($\theta = \pi/2$), then the effect of its interaction with this field is small.

CONCLUSION

In this paper, the kinetic theory of the magnetic light absorption by a metal nanoparticle with regard to the influence of a nanoparticle form on this process is constructed. In particular, the effect of spheroidal nanoparticles is analyzed. 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, $R_\perp / R_\parallel$, because the nanoparticle volume remains constant. The main results obtained in this work are as follows.

1. An expression (17) 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 (27) 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. The paper 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 $\theta \in \left\{ [0, \pi/2) \cup (\pi/2, \pi] \right\}$. The only value, at which the curve $W_m \left(R_\perp / R_\parallel, \theta \right) / W_m^0$ asymptotically approaches zero, is $\theta = \pi/2$.
4. The magnetic absorption in the frequency range Eq. (18), where it can be substantial. On the other hand, the magnetic absorption in the frequency range, where it can be substantial does not depend on the frequency at all.
5. The results of the work can be used to study solar cells with metal nanoparticles incorporated into the photosensitive layer. Also for the study of energy conversion processes in these elements.

Кінетична теорія магнітного поглинання лазерного опромінювання металевими наночастинками

О.Ю. Семчук, О.О. Гаврилюк, А.А. Білюк

Інститут хімії поверхні ім. О.О. Чуйка Національної академії наук України
вул. Генерала Наумова, 17, Київ, 03164, Україна, philosoph.tb@gmail.com

При освітленні металевих наночастинок (МНЧ) монохроматичною лазерної хвилею, частота якої далека від плазмонної частоти (частоти плазмонних резонансів), при певних умовах (залежно від частоти хвилі, її поляризації, розміру та форми МНЧ), поглинанні світла МНЧ може домінувати магнітне поглинання (поглинання, спричинене магнітною складовою електромагнітного поля світлової (лазерної) хвилі). Ця робота сконцентрована на вивченні особливостей поглинання, зумовленого впливом магнітної компоненти лазерного випромінювання. Це питання досить маловивчене для МНЧ несферичної форми. Тому те, як проявляється форма частинки в поглинанні нею лазерного випромінювання (лазерних імпульсів) є однією з цілей нашого дослідження. У цій роботі вивчатимемо особливості магнітного поглинання світла (лазерного випромінювання) в залежності від форми частинок. В роботі ми досліджуватимемо вплив на цей процес МНЧ сфероїдальної форми. Розрахунки проводитимемо методом кінетичного рівняння, тому що розглядатимемо випадок, коли розмір МНЧ менший від довжини вільного пробігу електрона в МНЧ. Зауважимо, що кінетичний підхід дає змогу отримати правильні результати для випадку, коли розмір частинки більший за довжину вільного пробігу. Для МНЧ несферичної форми нами розроблено теорію, яка має змогу обчислити енергію магнітного поглинання частинками при її опроміненні лазерними імпульсами. Побудовано і теоретично досліджено залежність магнітного поглинання від відношення радіусів кривизни сфероїдальних МНЧ і вектором магнітного поля електромагнітної (лазерної) хвилі. Цікавим результатом є поглинання енергії сфероїдальною МНЧ у міру зростання її дископодібності. Тепер ми використовуємо для оцінки відносного внеску електричного та магнітного поглинання в загальне поглинання. Наприклад, візьмемо золоті МНЧ $\omega_p \approx 5\cdot10^{15}\ \mathrm{s}^{-1}$, $\nu \approx 10^{13}\ \mathrm{s}^{-1}$, $R = 3\cdot10^{-6}$ sm, $\omega \approx 2\cdot10^{14}\ \mathrm{s}^{-1}$ *(вуглекислотний лазер),* $\varepsilon' \approx -600$, $\varepsilon'' \approx$. *Ми отримали наступне співвідношення* $W_e/W_m \approx 2$. *Ми бачимо, що для даного набору параметрів магнітне поглинання вдвічі більше, ніж електричне. Очевидно, що для різних параметрів частинки і різного діапазону частот електричне поглинання може бути як більшим, так і меншим, ніж магнітне. Отже, вивчаючи залежність оптичного поглинання МНЧ від форми частинок, ми повинні враховувати як електричне, так і магнітне поглинання. Для асиметричних МНЧ (наприклад, еліпсоїдальних частинок), окрім усього іншого, співвідношення електричних і магнітних внесків у поглинання (як фіксована частота) сильно залежить від ступеня асиметричності частинок і хвильової поляризації.*

Ключові слова*: магнітне поглинання, металеві наночастинки, електрон, плазмон, частота, енергія, поглинання*

REFERENCES

1. Boren C.F., Huffman D.R. *Absorption and Scattering of Light by Small Particles*. (New York City: Wiley, 1983).
2. van de Huls H.C. *Light Scattering by Small Particles*. (New York City: Wiley, 1957).
3. Kreibig U., Vollmer M. *Optical Properties of Metal Clusters*. (Berlin: Springer, 1995).
4. Venger E.F., Goncharenko A.V., Dmitruk M.L. *Optics of Small Particles and Disperse Media*. (Kyiv: Naukova Dumka, 1999). [in Ukrainian].
5. Landau L.D., Lifshitz E.M. *Electrodynamics of Continuous Media*. (New York: Oxford Pergamon Press, 1984).
6. Mie G. Beiträge zur Optik trüber Medien, speziell kolloidaler Metallösungen. *Annalen der Physik*. 1908. **25**: 377.
7. Tomchuk P.M., Tomchuk B.P. Optical absorption by small metal particles. *J. Exp. Theor. Phys.* 1997. **85**: 360.
8. Dykman I.N., Tomchuk P.M. *Phenomena of transfer and fluctuations in semiconductors*. (Kyiv: Naukova Dumka, 1982). [in Ukrainian].
9. Semchuk O.Yu., Havriliuk O.O., Biliuk A.A. Kinetic theory of surface plasmon resonance in metal nanoparticles. *Surface*. 2020. **12(27)**: 3. [in Ukrainian].

Received 02.06.2023, accepted 27.11.2023