doi: 10.15407/ujpe61.03.0255

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PACS 68.49.Jk, 72.10.-d, 73.20.Mf

INFLUENCE OF THE MAGNETIC DIPOLE MOMENT OF A METAL NANOELLIPSOID ON THE SCATTERING OF ELECTROMAGNETIC WAVES

The influence of the magnetic dipole moment of a non-spherical metal nanoparticle on the scattering of electromagnetic radiation by the particle has been studied in the framework of the kinetic approach. Analytical expressions for the scattering cross-section of spheroidal particles are obtained, and their dependence on the incident radiation wavelength and the nanoparticle eccentricity is analyzed. The contribution of a magnetic moment to the scattering at frequencies far from the plasmon resonance is shown to be comparable with that of the electric moment, with the ratio between the magnetic and electric moment contributions being maximum for spherical nanoparticles. The calculations are performed for an arbitrary ratio between the particle size and the free electron path, which enables our results to be compared with the Mie theory in the case where the electron scattering in the particle bulk dominates.

K e y w o r d s: electromagnetic radiation, metal nanoparticle, nanoellipsoid.

1. Introduction

In recent years, plenty of researches were devoted to the optical properties of nanoparticles. It is enough to mention some of the works dealing with various aspects of this problem, such as the influence of surface plasmons on the absorption and light scattering [1–3], nonlinear effects in the dynamics of fast electronic processes [4, 5], generation of strong ultra-violet radiation by metal nanostructures [6], optical heating of plasmon nanoparticles [7], and so forth. A detailed review of recent results, as well as theories which became classical, can be found, e.g., in works [8, 9].

If the size of a metal nanoparticle is smaller than the electromagnetic wavelength, the optical properties of such an object considerably depend on its shape. For example, the absorption and light scattering in metal nanoparticles are known to be mainly governed by plasma resonances. The number of resonances depends on the particle shape. For instance, there is one such resonance for a sphere, and three for a spheroidal particle. Moreover, if the particle size becomes smaller than the mean free path of electrons, the optical conductivity of an asymmetric particle is described by a tensor [10, 11], whose components determine the half-widths of plasma resonances. In this case, the Drude–Sommerfeld model has to be modified, because it does not take into account the tensor character of the optical conductivity, and the influence of the particle shape on plasma resonances is contained only in depolarization factors.

The authors devoted a series of works to this topic. However, the attention was mainly paid to the influence of the electric component on the scattering and light absorption. The matter is that the influence of the magnetic component of an external wave can be neglected in comparison with the electric one at plasma resonance frequencies. At the same time, it was shown in work [10] that, in the infra-red spectral interval (more exactly, at frequencies close to the frequency of a CO_2 laser), the contribution of the magnetic component to the absorption becomes comparable with that of the electric component and can be even larger, if the particle size increases.

This work is aimed at considering the influence of the field magnetic component on the electromagnetic wave scattering by metal nanoparticles in the frequency interval far from plasma resonances. In addition, the influence of the tensor character of the optical conductivity is taken into account for nonspherical (ellipsoidal) nanoparticles. Analytical expressions for the light scattering cross-section will be derived with regard for the magnetic contribution, and the dependences of the ratio between the magnetic and electric scatterings on the incident wave frequency, sizes of spheroidal nanoparticle, and spheroid eccentricity will be analyzed.

2. Formulation of the Problem

First, let us consider the general case of the problem dealing with the light scattering by a metal nanoparticle, when the particle is an ellipsoid with three different semiaxes: R_x, R_y, and R_z. Below, in order to obtain analytical expressions, we will confine the consideration to the case of an ellipsoid of revolution with $R_x = R_y \neq R_z$.

So, let an ellipsoidal metal nanoparticle be located in the field of a monochromatic electromagnetic wave

$$\begin{pmatrix}\mathbf{E}(\mathbf{r},t)\\ \mathbf{H}(\mathbf{r},t)\end{pmatrix} = \begin{pmatrix}\mathbf{E}^{(0)}\\ \mathbf{H}^{(0)}\end{pmatrix} e^{i(\mathbf{kr}-\omega t)}, \qquad (1)$$

where $\mathbf{E}$ and $\mathbf{H}$ are the electric and magnetic components, respectively, of the electromagnetic wave; ω and $\mathbf{k}$ are its frequency and wave vector; and $\mathbf{r}$ and t correspond to spatial coordinates and the time. We assume that the electromagnetic wavelength $\lambda = 2\pi/|\mathbf{k}|$ is much longer than the characteristic particle size. In this case, we may consider that the particle is in a field that is spatially uniform, but changes in time. This model makes it possible to consider only dipole modes and to neglect multipole contributions of higher orders. The electric component of the external field induces a local potential electric field $\mathbf{E}_{\text{loc}}$ inside the particle, which makes a contribution to an electric current with the density $\mathbf{j}_e$ that appears in the particle. The magnetic component of the wave induces a vortex electric field $\mathbf{E}_{\text{ed}}$ inside the metal particle, which also gives a contribution to the current with the density $\mathbf{j}_m$.

The electric and magnetic moments of metal nanoparticles induced by the external field generate a scattered wave. We will consider the scattered wave at large (in comparison with the wavelength) distances from the particle, where it can be assumed as transverse. The notations $\mathbf{E}'$ and $\mathbf{H}'$ will be used for the electric and magnetic, respectively, components of the scattered wave. The average intensity of radiation emitted by the electric and magnetic moments into the solid angle $d\Omega$ and at the distance R_0 from the particle equals [12]

$$dI_S = \frac{c}{8\pi}|[\mathbf{E}'\times\mathbf{H}']|^2 R_0^2 d\Omega = \frac{c}{8\pi}|\mathbf{H}'|^2 R_0^2 d\Omega, \qquad (2)$$

where c is the speed of light. In formula (2), we took into account that $|\mathbf{E}'| = |\mathbf{H}'|$.

The magnetic component of the field created by the scattered wave at large distances from the particle is given by the formula [12]

$$\mathbf{H}' = \frac{\omega^2}{c^2 R_0^2}\{[\mathbf{n}\times\mathbf{d}] + [\mathbf{n}\times[\mathbf{M}\times\mathbf{n}]]\}, \qquad (3)$$

where the unit vector $\mathbf{n}$ marks the scattering direction, and the vectors $\mathbf{d}$ and $\mathbf{M}$ are the induced electric and magnetic, respectively, dipole moments of the particle.

To find the differential scattering cross-section, the average scattered radiation intensity has to be divided by the energy flux density in the incident wave

$$I_0 = \frac{c}{8\pi}|\mathbf{H}|^2 = \frac{c}{8\pi}|\mathbf{E}|^2. \qquad (4)$$

As a result, the differential scattering cross-section equals

$$d\Sigma = \frac{dI_S}{I_0} = \frac{|\mathbf{H}'|^2}{|\mathbf{H}|^2} R_0^2 d\Omega. \qquad (5)$$

From expressions (3) and (5), it is clear that, in order to determine the differential scattering cross-section, we have to know expressions for the dipole and magnetic moments. The latter, in turn, are related to the fields $\mathbf{E}_{\text{loc}}$ and $\mathbf{E}_{\text{ed}}$ induced in the particle. Hence, to begin with, let us determine these fields.

3. Local Fields

As is known from the literature (see, e.g., [12]), the spatially uniform external electric field $\mathbf{E}^{(0)}$ induces a local potential electric field $\mathbf{E}_{\rm loc}$ in an ellipsoidal particle, which does not depend on the spatial coordinates. The local field $\mathbf{E}_{\rm loc}$ can be linearly expressed in terms of the external field $\mathbf{E}^{(0)}$, by introducing the depolarization tensor L_{ij}. In the principal axes of the tensor L_{ij}, which coincide with the principal axes of the ellipsoid, the relation between the external and local electric fields looks like [12]

$$(E_{\rm loc})_j = \frac{E_j^{(0)}}{1+L_j(\varepsilon-1)}, \tag{6}$$

where L_j are the principal values of the depolarization tensor L_{ij}, and ε is the dielectric permittivity of the metal of the particle.

In formula (6), the dielectric permittivity ε is a scalar. As was shown in work [10], for asymmetric particles smaller than the electron mean free path, the dielectric permittivity is a tensor ε_{ij}, since the high-frequency conductivity also is a tensor, σ^c_{ij}. Those two quantities are related by the well-known formula [13]

$$\varepsilon_{ij}(\mathbf{r},\omega) = \delta_{ij} + \frac{4\pi i}{\omega}\sigma^c_{ij}(\mathbf{r},\omega), \tag{7}$$

where δ_{ij} is the Kronecker symbol, and σ^c_{ij} the complex conductivity tensor. In view of the tensor character of the dielectric permittivity, expression (6) acquires the form

$$(E_{\rm loc})_j = \frac{E_j^{(0)}}{1+L_j(\varepsilon_{jj}-1)}, \tag{8}$$

where ε_{jj} stands for the diagonal component of the dielectric permittivity tensor along the axis j.

It should be noted that expressions (6) and (8) correspond to the situation where a metal nanoparticle is located in a medium with the dielectric permittivity $\varepsilon_m = 1$. The transition to the case of a medium with the dielectric permittivity $\varepsilon_m \neq 1$ is made by changing $\varepsilon \to \varepsilon/\varepsilon_m$ (or $\varepsilon_{jj} \to \varepsilon_{jj}/\varepsilon_m$ in formula (8)) [12]. Then formula (6) reads

$$(E_{\rm loc})_j = \frac{\varepsilon_m E_j^{(0)}}{\varepsilon_m + L_j(\varepsilon-\varepsilon_m)}. \tag{9}$$

It should also be noted that, in the case of an ensemble consisting of many particles, the polarization vector in a separate particle is induced not only by the external field, but also the dipoles induced by the external field in other particles belonging to the ensemble [14]. This effect will be neglected here, but it can be taken into consideration in the interaction scenario [15].

Let us proceed to the determination of a form of the vortex local field $\mathbf{E}_{\rm ed}$. This field has to satisfy Maxwell's equations

$$\mathrm{rot}\mathbf{E}_{\rm ed} = \frac{i\omega}{c}\mathbf{H}^{(0)}, \tag{10}$$

$$\mathrm{div}\mathbf{E}_{\rm ed} = 0. \tag{11}$$

As for the right-hand side of Eq. (10), we interpret the external magnetic field as a field in the particle. This approximation is valid, if the thickness of the skin layer is much larger than the characteristic size of the particle [12]

$$\delta_H = \left(\frac{\omega}{c}\mathrm{Im}\sqrt{\varepsilon}\right)^{-1} \gg R_{\rm max}, \tag{12}$$

where δ_H is the skin layer thickness, and $R_{\rm max}$ is the largest ellipsoid semiaxis.

By integrating Eq. (11) over the particle volume and using the Ostrogradskii–Gauss theorem, we obtain a condition for the vortex local field at the surface of a metal particle,

$$\mathbf{E}_{\rm ed}\cdot\mathbf{n}_S = 0, \tag{13}$$

where $\mathbf{n}_S$ is a unit vector normal to the particle surface. Equations (10), (11), and (13) completely determine the field $\mathbf{E}_{\rm ed}$. Taking inequality (12) into account, the expression on the right-hand side of Eq. (10) is constant. Therefore, the field $\mathbf{E}_{\rm ed}$ has to be a linear function of the coordinates. In the general form, this conclusion can be expressed as follows:

$$(E_{\rm ed})_j = \sum_{k=1}^{3}\alpha_{jk}x_k. \tag{14}$$

Using Eq. (11) and the boundary conditions (13), we obtain the following analytical expressions for the coefficients α_{jk}:

$$\alpha_{xy} = -\frac{i\omega}{c}\frac{R_x^2}{R_x^2+R_y^2}H_z^{(0)},\ \alpha_{yx} = \frac{i\omega}{c}\frac{R_y^2}{R_y^2+R_x^2}H_z^{(0)},$$
$$\alpha_{yz} = -\frac{i\omega}{c}\frac{R_y^2}{R_y^2+R_z^2}H_x^{(0)},\ \alpha_{zy} = \frac{i\omega}{c}\frac{R_z^2}{R_z^2+R_y^2}H_x^{(0)},$$
$$\alpha_{zx} = -\frac{i\omega}{c}\frac{R_z^2}{R_z^2+R_x^2}H_y^{(0)},\ \alpha_{xz} = \frac{i\omega}{c}\frac{R_x^2}{R_x^2+R_z^2}H_y^{(0)}, \tag{15}$$

where R_x, R_y, and R_z are the semiaxes of the ellipsoid directed along the axes x, y, and z, respectively.

Formulas (14) and (15) give explicit analytic expressions for the vortex electric field in the particle. It is easy to verify that, in the spherical case, we obtain the expression

$$\mathbf{E}_{\rm ed} = \frac{\omega}{2ic}[\mathbf{r} \times \mathbf{H}^{(0)}]. \tag{16}$$

From whence, one can see that the vortex local field is perpendicular to the external magnetic field in the case of spherical particle.

Having determined the local fields that arise in a metal nanoparticle under the action of an external electromagnetic field [see formulas (6), (8), (14), and (15)], it is possible to calculate the electric and magnetic dipole moments induced in the particle.

4. Electric and Magnetic Dipole Moments

The electric and magnetic dipole moments that arise under the influence of an external electromagnetic field are related to the current density $\mathbf{j}(\mathbf{r}, t)$ by the following relations [12]:

$$\frac{\partial}{\partial t}\mathbf{d}(t) = \int\limits_V d^3r'\mathbf{j}(\mathbf{r}', t), \tag{17}$$

$$\mathbf{M}(t) = \frac{1}{2c}\int\limits_V d^3r'[\mathbf{r}' \times \mathbf{j}], \tag{18}$$

where $\mathbf{d}(t)$ and $\mathbf{M}(t)$ are the electric and magnetic, respectively, dipole moments, and the integration is carried out over the nanoparticle volume V. The current density $\mathbf{j}(\mathbf{r}, t)$, in turn, can be determined, if we know the distribution of electrons in the nanoparticle over their velocities. More precisely, we should find a nonequilibrium correction to the Fermi function, which describes the action of the local fields $\mathbf{E}_{\rm loc}$ and $\mathbf{E}_{\rm ed}$ in the particle.

In the linear approximation, the distribution function of electron velocities can be written in the form

$$f(\mathbf{r}, \mathbf{v}, t) = f_0(\varepsilon) + f_1(\mathbf{r}, \mathbf{v})e^{-i\omega t}, \tag{19}$$

where $f_0(\varepsilon)$ is the Fermi distribution function, and $f_1(\mathbf{r}, \mathbf{v})$ is the nonequilibrium term in the linear approximation. In this case, the Boltzmann kinetic equation looks like

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

$$+ e(\mathbf{E}_{\rm loc} + \mathbf{E}_{\rm ed})\mathbf{v}\frac{\partial f_0}{\partial \varepsilon} = 0, \tag{20}$$

and the collision integral is calculated in the relaxation-time approximation,

$$\left(\frac{\partial f_1}{\partial t}\right)_{\rm col} = -\frac{f_1}{\tau}, \quad \tau = \frac{1}{\nu}. \tag{21}$$

Equation (20) should be supplemented with boundary conditions at the nanoparticle surface. Both the diffusion and specular boundary conditions are considered in the literature (see, e.g., work [16]). For nonplanar boundaries (in our case, the boundary is described by the equation of ellipsoid), we may confine the model to the diffusion conditions at the particle surface:

$$f_1(\mathbf{r}, \mathbf{v})|_S = 0, \ v_n < 0, \tag{22}$$

where v_n is the velocity component normal to the particle surface.

We will not dwell in detail here on the solution procedure for Eq. (20) with the boundary conditions (22). The corresponding details can be found, e.g., in work [17]. We would like only to note that it is convenient to find a solution after changing to a new coordinate system

$$x'_i = \gamma_i x_i, \ v'_i = \gamma_i v_i, \ \gamma_i = \frac{R}{R_i}, \ R = (R_x R_y R_z)^{1/3}. \tag{23}$$

Then the solution of Eq. (20) with the boundary conditions (22) looks like

$$f_1(\mathbf{r}, \mathbf{v}) = -e\frac{\partial f_0}{\partial \varepsilon}\Bigg\{\mathbf{v}\mathbf{E}_{\rm loc} + $$

$$+ \sum_{i,j=1}^{3} \alpha_{ij} v_i \left(\frac{x'_j}{\gamma_j} + v_j \frac{\partial}{\partial \tilde{\nu}}\right)\Bigg\}\frac{1 - e^{-\tilde{\nu}t'}}{\tilde{\nu}}, \tag{24}$$

where the notation $\tilde{\nu} = \nu - i\omega$ is introduced, and

$$t'(\mathbf{r}', \mathbf{v}') = \frac{1}{v'^2}\left[\mathbf{r}'\mathbf{v}' + \sqrt{(\mathbf{R}^2 - \mathbf{r}'^2)\mathbf{v}'^2 + (\mathbf{r}'\mathbf{v}')^2}\right] \tag{25}$$

is the characteristic of Eq. (20).

Now, on the basis of the distribution function (24), we can calculate the electric current density, by using the formula

$$\mathbf{j}(\mathbf{r}, \omega) = 2e\left(\frac{m}{2\pi\hbar}\right)^3 \iiint \mathbf{v} f_1(\mathbf{r}, \mathbf{v}) d^3v, \tag{26}$$

where e and m are the electron charge and mass, respectively, and $\hbar$ Planck's constant. Hence, expressions (17), (18), and (24)–(26) completely determine the electric and magnetic dipole moments that arise in the metal nanoparticle under the action of an external electromagnetic field. Nevertheless, the further calculations are convenient to be carried out in terms of the polarizability, κ_{ij}, and magnetic susceptibility, χ_{ij}, tensors. They are introduced by the relations

$$d_i(\omega) = \sum_{i,j=1}^{3} \kappa_{ij}(\omega) E_j^{(0)}, \qquad (27)$$

$$M_i(\omega) = \sum_{i,j=1}^{3} \chi_{ij}(\omega) H_j^{(0)}. \qquad (28)$$

Similar expressions for the tensors introduced above can be found by calculating the Fourier transforms of Eqs. (17) and (18) and comparing them with expressions (27) and (28). The formula for the polarizability tensor κ_{ij} was obtained in previous works (see work [10]). Therefore, we will not reproduce the derivation completely. Instead, we present only the final expression, which is necessary for further calculations:

$$\kappa_{ii} = iV \frac{\langle \sigma_{ii}^c \rangle}{\omega + i4\pi L_{ii} \langle \sigma_{ii}^c \rangle}, \qquad (29)$$

where V is the volume of a metal nanoparticle, and $\langle \sigma_{ii}^c \rangle$ are the diagonal components of the complex conductivity tensor averaged over the particle volume. The expression for this tensor can be found from Ohm's law in the differential form

$$j_i(\mathbf{r},\omega) = \sum_{i,j=1}^{3} \langle \sigma_{ij}^c(\omega) \rangle (E_{\rm loc})_j, \qquad (30)$$

by using relations (24)–(26).

We omit the details of calculations, which can be found, e.g., in works [10, 11]. The final expression for $\left\langle \sigma_{ij}^c(\omega) \right\rangle$ looks like

$$\langle \sigma_{ij}^c(\omega) \rangle = \left(\frac{m}{2\pi\hbar} \right)^3 \frac{2e^2}{\tilde{\nu}} \int \frac{d^3 r'}{V} \int d^3 v \, v_i v_j \times$$
$$\times \, \delta(\varepsilon - \varepsilon_{\rm F}) \left(1 - e^{-\tilde{\nu} t'} \right), \qquad (31)$$

where ε_F is the Fermi energy. It is easy to see that, in the case $i \neq j$, the integration over the whole velocity space gives zero. Therefore, the complex conductivity tensor $\left\langle \sigma_{ij}^c(\omega) \right\rangle$ and, as a consequence, the polarizability tensor $\kappa_{ij}(\omega)$ are diagonal.

In order to find the tensor of magnetic susceptibility $\chi_{ij}(\omega)$, let us substitute expression (26) for the current density into formula (18) and use the expression for the distribution function $f_1(\mathbf{r}, \mathbf{v})$. We obtain

$$M_i = \frac{e^2}{c} \left(\frac{m}{2\pi\hbar} \right)^3 \frac{1}{\tilde{\nu}} \int d^3 v \, \delta(\varepsilon - \varepsilon_{\rm F}) \int d^3 r' \left(1 - e^{-\tilde{\nu} t'} \right) \times$$
$$\times \sum_{j,k,l,m} \varepsilon_{ijk} \alpha_{lm} \frac{x'_j x'_m v'_k v'_l}{\gamma_j \gamma_k \gamma_l \gamma_m}. \qquad (32)$$

It should be noted that expression (24) for the distribution function f_1 contains two terms which are absent from Eq. (32). However, it is easy to see that one of them (with $\mathbf{v}\mathbf{E}_{\rm loc}$) vanishes at the integration over the spatial coordinates, and the other one (with $\alpha_{ij} v_i v_j \frac{\partial}{\partial \tilde{\nu}}$) at the integration over the whole velocity space.

From Eq. (32), taking expressions (11A), (13A), and (7A) (see Appendix) into account and making the corresponding changes in the notations, we obtain

$$M_i = \frac{\pi R^5 e^2}{2\tilde{\nu} c} \left(\frac{m}{2\pi\hbar} \right)^3 \int d^3 v \, \delta(\varepsilon - \varepsilon_{\rm F}) \times$$
$$\times \sum_{j,k,l,m} \left[\frac{\varepsilon_{ijk} \alpha_{lj} v'_k v'_l}{\gamma_j^2 \gamma_k \gamma_l} \Psi_1(q) + \frac{\varepsilon_{ijk} \alpha_{lm} v'_j v'_k v'_l v'_m}{\gamma_j \gamma_k \gamma_l \gamma_m} \Psi_2(q) \right], \qquad (33)$$

where the following notations are introduced:

$$\Psi_1(q) = \frac{8}{15} - \frac{1}{q} + \frac{4}{q^3} - \frac{24}{q^5} + \frac{8}{q^2} \left(1 + \frac{3}{q^2} + \frac{3}{q^3} \right) e^{-q}, \qquad (34)$$

$$\Psi_2(q) = -\frac{1}{q} + \frac{32}{3q^2} - \frac{36}{q^3} + \frac{200}{q^5} - \frac{8}{q^2} \left(1 + \frac{8}{q} + \frac{25}{q^2} + \frac{25}{q^3} \right) e^{-q}, \qquad (35)$$

$$q = \frac{2\tilde{\nu} R}{v'}. \qquad (36)$$

The second term in Eq. (33) evidently disappears at the summation over j and k. In the first term, when integrating over the whole velocity space, only the terms with $k = l$ survive. Therefore, for the magnetic moment components, taking the coefficients α_{ij} into account, we obtain

$$\mathbf{M} = \frac{\pi R^3 e^2}{2c^2}\frac{i\omega}{\tilde{\nu}}\left(\frac{m}{2\pi\hbar}\right)^3 \int d^3v\,\delta(\varepsilon-\varepsilon_{\rm F})\Psi_1(q)\times \times \begin{bmatrix} \frac{R_y^2R_z^2}{R_y^2+R_z^2}(v_y^2+v_z^2)H_x^{(0)} \\ \frac{R_z^2R_x^2}{R_z^2+R_x^2}(v_z^2+v_x^2)H_y^{(0)} \\ \frac{R_x^2R_y^2}{R_x^2+R_y^2}(v_x^2+v_y^2)H_z^{(0)} \end{bmatrix}. \quad (37)$$

The integration over the velocities in Eq. (37) can be done analytically, only if the particle is spherical. In this case, from Eq. (37), we obtain

$$\mathbf{M} = \frac{2\pi^2 R^5 e^2 v_{\rm F}^3}{3mc^2}\frac{i\omega}{\tilde{\nu}}\left(\frac{m}{2\pi\hbar}\right)^3 \Psi_1\left(\frac{2\tilde{\nu}R}{v_{\rm F}}\right)\mathbf{H}^{(0)}. \quad (38)$$

This formula coincides with the result obtained in work [18].

It should be noted that expressions (37) and (38) have a general value in the meaning that they are applicable irrespective of whether the particle size is larger or smaller than the electron mean free path. It is easy to demonstrate that, in the case where the particle size exceeds the electron mean free path, the expressions for the magnetic moment can be simplified. For this purpose, we note that the dimensionless quantity q together with the magnetic moment vector $\mathbf{M}$ depend on the relations among the frequencies ν, ν_s, and ω, where the frequency

$$\nu_s = \frac{v_{\rm F}}{2R} \quad (39)$$

has a sense of the frequency of collisions of an electron with particle walls, whereas the frequency ν is a quantity reciprocal to the relaxation time and has a sense of the frequency of collisions of an electron in the particle bulk. If $\nu \gg \nu_s$, the electron scattering in the particle bulk dominates; whereas, in the inverse case $\nu \ll \nu_s$, the scattering of electrons owing to their collisions with the particle surface plays the key role. In the limiting case of bulk scattering, expression (38) transforms into the known result [12]

$$\mathbf{M} = \frac{(kR)^2}{30}R^3\mathbf{H}^{(0)}. \quad (40)$$

The formula for the magnetic susceptibility tensor χ_{ij} can be derived from expressions (28) and (37). For the polarizability κ_{ij}, it can be done from relations (29) and (31). We note that the integration over the spatial coordinates in formula (31) can be executed, as was made for the magnetic moment. As a result, for the conductivity tensor, we obtain

$$\langle\sigma_{ij}^c(\omega)\rangle = \frac{3e^2}{2\tilde{\nu}}\left(\frac{m}{2\pi\hbar}\right)^3 \int d^3v\delta(\varepsilon-\varepsilon_{\rm F})\,v_iv_j\Psi(q), \quad (41)$$

where

$$\Psi(q) = \frac{4}{3} - \frac{2}{q} + \frac{4}{q^3} - \frac{4}{q^2}\left(1+\frac{1}{q}\right)e^{-q}. \quad (42)$$

In the spherical case, the calculation of the integral in Eq. (41) brings about the formula

$$\langle\sigma_{ij}^c(\omega)\rangle 0 = \frac{3}{16\pi}\frac{\omega_{\rm pl}^2}{\tilde{\nu}}\Psi(q)|_{v=v_{\rm F}}\delta_{ij}, \quad (43)$$

where the notations for the plasma frequency,

$$\omega_{\rm pl} = \sqrt{\frac{4\pi ne^2}{m}}, \quad (44)$$

and free charge carrier concentration,

$$n = \frac{8\pi}{3}\left(\frac{m}{2\pi\hbar}\right)^3, \quad (45)$$

are introduced. It is easy to see that, in the case $q \gg$ $\gg 1$, Eq. (43) leads to the known Drude formula for the high-frequency optical conductivity

$$\langle\sigma_{ij}^c(\omega)\rangle = \frac{1}{4\pi}\frac{\omega_{\rm pl}^2}{\nu - i\omega}\delta_{ij}. \quad (46)$$

Expression (46) is applicable, if the bulk electron scattering dominates over the surface one, i.e. when $\nu \gg \nu_s$.

5. Scattering Cross-Section

As was indicated above, the differential scattering cross-section is determined by the ratio between the average (over the period) intensity of a scattered wave at the distance R_0 from the nanoparticle into the solid angle $d\Omega$ to the energy density in the incident flux, i.e.

$$d\Sigma = \frac{\omega^4}{c^4}\frac{1}{|\mathbf{H}^{(0)}|^2}\Big\{|[\mathbf{n}\times\mathbf{d}]|^2 + |[\mathbf{n}\times[\mathbf{M}\times\mathbf{n}]]|^2 + + 2{\rm Re}\left([\mathbf{n}\times\mathbf{d}]\cdot[\mathbf{n}\times[\mathbf{M}\times\mathbf{n}]]\right)\Big\}d\Omega. \quad (47)$$

Expression (47) describes the cross-section of light scattering by a single metal nanoparticle. Although the experiments, in which the interaction of light with a single nanoparticle is studied, are already available nowadays, an ensemble of particles is a much more convenient object for the analysis. Therefore, let us consider an ensemble of metal nanoparticles with the same volume and shape. Let the distance between the particles in the ensemble be much larger than their size. In this case, the interaction between the particles can be neglected. All expressions obtained till now are applicable to the general case of three-axial ellipsoid. Below, to simplify calculations, we choose the particle shape as an ellipsoid of revolution (a spheroid), for which $R_x = R_y = R_\perp$ and $R_z = R_\parallel$.

Let us introduce a unit vector $\mathbf{q}_0$ directed along the spheroid rotation axis. The vectors $\mathbf{E}^{(0)}$ and $\mathbf{H}^{(0)}$ describing the external electromagnetic wave can be decomposed into the components parallel and perpendicular to the rotation axis, as is shown in Fig. 1:

$$\mathbf{E}^{(0)} = \mathbf{E}_\parallel^{(0)} + \mathbf{E}_\perp^{(0)} = (\mathbf{E}^{(0)}\cdot\mathbf{q}_0)\mathbf{q}_0 + \mathbf{E}_\perp^{(0)}, \tag{48}$$

$$\mathbf{H}^{(0)} = \mathbf{H}_\parallel^{(0)} + \mathbf{H}_\perp^{(0)} = (\mathbf{H}^{(0)}\cdot\mathbf{q}_0)\mathbf{q}_0 + \mathbf{H}_\perp^{(0)}. \tag{49}$$

Expressions (27) and (28) give us the components of the electric and magnetic moments along the field components of the incident wave in the spheroidal case:

$$\mathbf{d} = \kappa_\parallel\mathbf{E}_\parallel^{(0)} + \kappa_\perp\mathbf{E}_\perp^{(0)}, \tag{50}$$

$$\mathbf{M} = \chi_\parallel\mathbf{H}_\parallel^{(0)} + \chi_\perp\mathbf{H}_\perp^{(0)}. \tag{51}$$

Here, $\kappa_\parallel$, $\kappa_\perp$ and $\chi_\parallel$, $\chi_\perp$ are the components of the polarizability and magnetic susceptibility, respectively, tensors along the rotation axis ($\parallel$) of the spheroid and perpendicularly to it ($\perp$).

All expressions, which are required to find the differential scattering cross-section (47), were determined above. We are interested in the case where the wave is not scattered by a single particle, but by an ensemble of chaotically oriented spheroidal metal nanoparticles. Every spheroid in this ensemble has its own vector $\mathbf{q}_0$ directed along the rotation axis. Therefore, in order to find the cross-section of scattering by such an ensemble, expression (47) has to be averaged over all possible directions of the vector $\mathbf{q}_0$. As a result of the averaging over the directions of nanospheroid orientations, we obtain the following expression for the differential scattering cross-section:

$$\langle d\Sigma\rangle_{\mathbf{q}_0} = \frac{\omega^4}{15c^4}\frac{1}{|\mathbf{H}^{(0)}|^2}\Bigg\{2|\mathbf{E}^{(0)}|^2|\kappa_\perp-\kappa_\parallel|^2 + \frac{1}{2}\left|[\mathbf{n}\times\mathbf{E}^{(0)}]\right|^2\left(3|2\kappa_\perp+\kappa_\parallel|^2+2|\kappa_\perp|^2+|\kappa_\parallel|^2\right) + 2|\mathbf{H}^{(0)}|^2|\chi_\perp-\chi_\parallel|^2 + \frac{1}{2}\left|[\mathbf{n}\times\mathbf{H}^{(0)}]\right|^2\left(3|2\chi_\perp+\chi_\parallel|^2 + 2|\chi_\perp|^2+|\chi_\parallel|^2\right)+2\mathrm{Re}\Big(5\mathbf{E}^{(0)}[\mathbf{H}^{(0)}\times\mathbf{n}](\kappa_\parallel\chi_\perp^*+\kappa_\perp\chi_\parallel^*) + 7[\mathbf{n}\times\mathbf{E}^{(0)}]\cdot[\mathbf{n}\times\mathbf{H}^{(0)}]\,\kappa_\perp\chi_\perp^*\Big)\Bigg\}d\Omega. \tag{52}$$

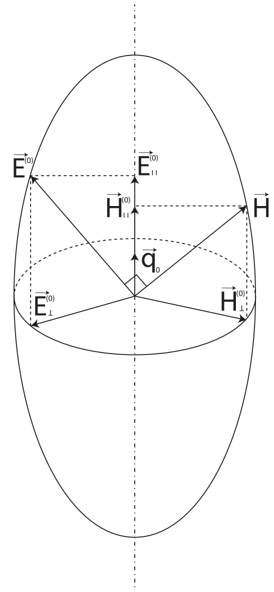

Fig. 1. Decomposition of the vectors $\mathbf{E}^{(0)}$ and $\mathbf{H}^{(0)}$ taking the symmetry of the problem into account

The analysis of this expression is complicated, because the angle between the direction of the observation vector $\mathbf{n}$ and the electric component of an incident wave $\mathbf{E}^{(0)}$, on the one hand, and the angle between $\mathbf{n}$ and $\mathbf{H}^{(0)}$, on the other hand, are not connected with each other. Therefore, in order to avoid the analysis of the dependence of the differential scattering cross-section on those angles, let us integrate expression (52) over all scattering directions. In other words, let us determine the total scattering cross-section. The result takes the following form:

$$\langle\Sigma\rangle_{\mathbf{q}_0} = \frac{2\pi\omega^4}{15c^4}\Bigg\{4|\kappa_\perp-\kappa_\parallel|^2 + \frac{1}{3}\left(3|2\kappa_\perp+\kappa_\parallel|^2 + 2|\kappa_\perp|^2+|\kappa_\parallel|^2\right) + 4|\chi_\perp-\chi_\parallel|^2 + \frac{1}{3}\left(3|2\chi_\perp+\chi_\parallel|^2 + 2|\chi_\perp|^2+|\chi_\parallel|^2\right)\Bigg\}. \tag{53}$$

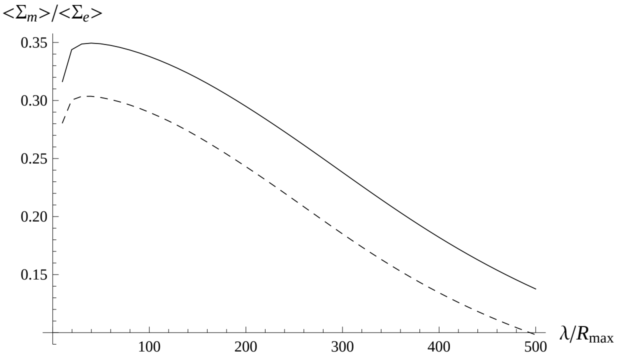

Fig. 2. Dependences of the ratio between the cross-sections of magnetic and electric scatterings, $\langle\Sigma_m\rangle/\langle\Sigma_e\rangle$ on the incident wavelength λ normalized by the length $R_{\max}$ of the larger semiaxis of a spheroid for a copper nanoparticle with $R = 20$ nm and various ratios $R_\perp/R_\parallel = 1.5$ (solid curve) and 0.67 (dashed curve)

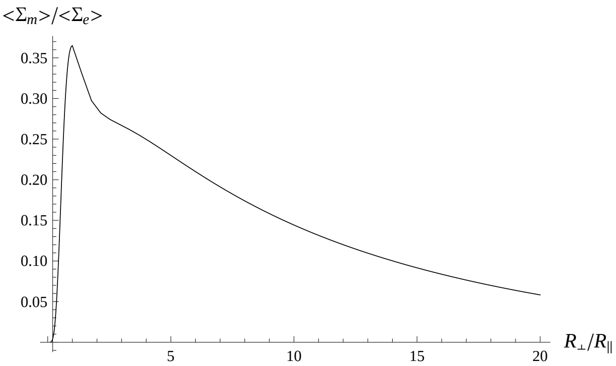

Fig. 3. Dependence of the ratio $\langle\Sigma_m\rangle/\langle\Sigma_e\rangle$ between the cross-sections of magnetic and electric scatterings on the ratio $R_\perp/R_\parallel$ between the spheroid semiaxes for a copper nanoparticle with $R = 20$ nm at the frequency $\omega = 2 \times 10^{15}$ s^{-1} ($\lambda \approx 942.5$ nm)

Two comments should be made concerning the result obtained. First, while integrating over the coordinates of the vector $\mathbf{n}$, the cross term including the product of the fields $\mathbf{E}^{(0)}$ and $\mathbf{H}^{(0)}$ vanishes. Second, although the dependences of the total scattering cross-section $\langle\Sigma\rangle_{\mathbf{q}_0}$ on $\kappa_{\perp,\parallel}(\omega)$ and $\chi_{\perp,\parallel}(\omega)$ are functionally identical, the frequency dependences of the electric and magnetic responses are different, because $\kappa_{\perp,\parallel}(\omega)$ and $\chi_{\perp,\parallel}(\omega)$ differently depend on the frequency.

Our task consists in comparing the contributions of the electric and magnetic components to the scattering cross-section at frequencies far from the frequencies of plasma resonances. For this purpose, let us numerically calculate the ratio between those two components,

$$\frac{\langle\Sigma_m\rangle}{\langle\Sigma_e\rangle} = \frac{4|\chi_\perp - \chi_\parallel|^2 + \frac{1}{3}\left(3|2\chi_\perp + \chi_\parallel|^2 + 2|\chi_\perp|^2 + |\chi_\parallel|^2\right)}{4|\kappa_\perp - \kappa_\parallel|^2 + \frac{1}{3}\left(3|2\kappa_\perp + \kappa_\parallel|^2 + 2|\kappa_\perp|^2 + |\kappa_\parallel|^2\right)}. \tag{54}$$

In Fig. 2, the dependence of expression (54) on the incident wavelength divided by the length of the longest spheroid axis is shown. The wavelength normalization was done, because the developed theory is valid only if the particle size is much smaller than the wavelength, and, hence, the shorter waves cannot be considered. From Fig. 2, one can see that, at wavelengths far from the plasma resonances, the magnetic component of the scattering is of the same order as the electric one and, therefore cannot be neglected.

Figure 3 demonstrates the dependence of expression (54) on the ratio between the spheroid axes $R_\perp/R_\parallel$. As is seen, there is a peak at a point, where $R_\perp = R_\parallel$. This means that the contribution of the magnetic component is maximum, if the particle is a sphere.

6. Conclusions

To summarize, the contribution of the magnetic dipole scattering to the scattering cross-section of an electromagnetic wave by the ensemble of chaotically oriented spheroidal nanoparticles has been studied for an arbitrary relation between the particle size and the electron mean free path. It is shown that, at frequencies far from those of plasma resonances, the cross-sections of magnetic and electric scatterings are of the same order. On the contrary, at frequencies close to plasma resonances, the magnetic component can be neglected in comparison with the electric one. In addition, the influence of the nanoparticle shape on the ratio between the cross-sections of electric and magnetic scatterings is analyzed. The magnetic scattering contribution is shown to be maximum if the nanoparticle has spherical shape. The analytical expression for the magnetic moment of a metal nanoellipsoid is derived for the first time within the kinetic approach. The formula transforms into the classical result for a sphere, if the mean free path of electrons is much longer than the particle size.

APPENDIX

Let us calculate the magnetic moment of a metal nanoparticle given by the formula:

$$M_i = \frac{e^2}{c}\left(\frac{m}{2\pi\hbar}\right)^3\frac{1}{\tilde\nu}\int d^3v\,\delta(\varepsilon-\varepsilon_{\rm F})\int d^3r'\left(1-e^{-\tilde\nu t'}\right)\times$$
$$\times\sum_{j,k,l,m}\varepsilon_{ijk}\alpha_{lm}\frac{x'_j x'_m v'_k v'_l}{\gamma_j\gamma_k\gamma_l\gamma_m}. \qquad (1A)$$

First, let us integrate over the spatial coordinates $\mathbf{r}'$. For this purpose, we orient the axis z' along the vector $\mathbf{v}$. In this case, the characteristic does not depend on the angle φ', and we should calculate the integral

$$\int_0^{2\pi} d\varphi' x'_j x'_m = \int_0^{2\pi} d\varphi'(\mathbf{e}_j\cdot\mathbf{r}')(\mathbf{e}_m\cdot\mathbf{r}'). \qquad (2A)$$

Here, we introduced the unit vectors in the coordinate system associated with the ellipsoid semiaxes:

$$\mathbf{e}_j = (\sin\psi_j\cos\varphi_j, \sin\psi_j\sin\varphi_j, \cos\psi_j), \qquad (3A)$$

where ψ_j is the angle between the vectors $\mathbf{e}_j$ and $\mathbf{v}'$, and φ_j is the polar angle in a plane perpendicular to the vector $\mathbf{v}'$. Additionally introducing the angle θ' between the vectors $\mathbf{r}'$ and $\mathbf{v}$, as well as the polar angle φ' in a plane perpendicular to the vector $\mathbf{v}$, we can expand expression (2A) in the form

$$\int_0^{2\pi} d\varphi' x'_j x'_m = 2\pi r'^2\Big\{\cos\psi_j\cos\psi_m + \sin^2\theta'\times$$
$$\times\left[\frac{1}{2}\sin\psi_j\sin\psi_m\cos(\varphi_j-\varphi_m) - \cos\psi_j\cos\psi_m\right]\Big\}. \qquad (4A)$$

From Eq. (4A) with regard for the relations

$$\sin\psi_j\sin\psi_m\cos(\varphi_j-\varphi_m) + \cos\psi_j\cos\psi_m = \mathbf{e}_j\cdot\mathbf{e}_m = \delta_{jm}, \qquad (5A)$$

$$\cos\psi_j = \frac{\mathbf{e}_j\cdot\mathbf{v}'}{v'^2}, \cos\psi_m = \frac{\mathbf{e}_m\cdot\mathbf{v}'}{v'^2}, \qquad (6A)$$

we obtain finally

$$\int_0^{2\pi} d\varphi' x'_j x'_m = 2\pi r'^2\left\{\frac{v'_j v'_m}{v'^2} + \frac{1}{2}\sin^2\theta'\left[\delta_{jm} - 3\frac{v'_j v'_m}{v'^2}\right]\right\}. \qquad (7A)$$

Substituting this result into Eq. (1A), we obtain two terms,

$$I_1 = \int_0^{\pi} d\theta'\sin\theta'\int_0^R dr' r'^4\left(1-e^{-\tilde\nu t'}\right), \qquad (8A)$$

$$I_2 = \int_0^{\pi} d\theta'\sin^3\theta'\int_0^R dr' r'^4\left(1-e^{-\tilde\nu t'}\right). \qquad (9A)$$

which are to be integrated over θ' and r'. They can be calculated, if we make the substitutions

$$\eta = \frac{r'}{R}, \quad \xi = \frac{v't'}{R}. \qquad (10A)$$

Then, for I_1, we obtain

$$I_1 = R^5\int_0^1 d\eta\eta^4\int_{1-\eta}^{1+\eta} d\xi\frac{\xi^2-\eta^2+1}{2\xi^2\eta}\left[1-\exp\left(-\frac{\tilde\nu R}{v'}\xi\right)\right] =$$
$$= \frac{1}{2}R^5\int_0^2\frac{d\xi}{\xi^2}\left[1-\exp\left(-\frac{\tilde\nu R}{v'}\xi\right)\right]\int_{|\xi-1|}^1 d\eta\eta^3(\xi^2-\eta^2+1) =$$
$$= R^5\left[\frac{2}{5} - \frac{1}{q} + \frac{8}{3q^2} - \frac{6}{q^3} + \frac{32}{q^5} - \frac{2}{q^2}\left(1+\frac{5}{q}+\frac{16}{q^2}+\frac{16}{q^3}\right)e^{-q}\right], \qquad (11A)$$

where

$$q = \frac{2\tilde\nu R}{v'}. \qquad (12A)$$

Analogously, for I_2, we obtain

$$I_2 = R^5\left[\frac{4}{15} - \frac{1}{2q} + \frac{2}{q^3} - \frac{12}{q^5} + \frac{4}{q^2}\left(\frac{1}{q}+\frac{3}{q^2}+\frac{3}{q^3}\right)e^{-q}\right]. \qquad (13A)$$

1. B. Hourahine and F. Papoff, Opt. Express **21**, 17 (2013).
2. Y.H. Fu, A.I. Kuznetsov, A.E. Miroshnichenko, Y.F. Yu, and B. Luk'yanchuk, Nature Comm. **4**, 1527 (2013).
3. P. Liu, Ju. Liu, Ji. Liu, X. Zhao, J. Xie, and Y. Wang, Opt. Commun. **284**, 1076 (2011).
4. R. Sato, M. Ohnuma, K. Oyoshi, and Y. Takeda, Phys. Rev. B **90**, 125417 (2014).
5. H. Baida, D. Mongin, D. Christofilos, G. Bachelier, A. Crut, P. Maioli, N. Del Fatti, and F. Vallée, Phys. Rev. Lett. **107**, 057402 (2011).
6. M. Sivis, M. Duwe, B. Abel, and C. Ropers, Nature Phys. **9**, 304 (2013).
7. P. Berto, M.S.A. Mohamed, H. Rigneault, and G. Baffou, Phys. Rev. B **90**, 035439 (2014).
8. G.V. Hartland, Chem. Rev. **111**, 3858 (2011).
9. V.V. Klimov, *Nanoplasmonics* (Nauka, Moscow, 2012) (in Russian).
10. P.M. Tomchuk and N.I. Grigorchuk, Phys. Rev. B **73**, 155423 (2006).
11. N.I. Grigorchuk and P.M. Tomchuk, Phys. Rev. B **84**, 085448 (2011).
12. L.D. Landau and E.M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Press, New York, 1984).
13. A.F. Alexandrov, L.S. Bogdankevich, and A.A. Rukhadze, *Principles of Plasma Electrodynamics* (Springer, Berlin, 1984).
14. H.J. Trodahl, Phys. Rev. B **19**, 1316 (1979).
15. E.D. Belotsky, S.P. Luk'yanets, and P.M. Tomchuk, Zh. Èksp. Teor. Fiz. **101**, 163 (1992).
16. S.V. Berezkina, I.A. Kuznetsova, and A.A. Yushkanov, Zh. Tekhn. Fiz. **74**, 67 (2004).
17. A.G. Lesskis, V.E. Pasternak, and A.A. Yushkanov, Zh. Èksp. Teor. Fiz. **83**, 310 (1982).

18. I.A. Kuznetsova, M.E. Lebedev, and A.A. Yushkanov, Pis'ma Zh. Tekhn. Fiz. **40**, 8 (2014).

Received 09.09.14.
Translated from Ukrainian by O.I. Voitenko

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ВПЛИВ МАГНІТНОЇ СКЛАДОВОЇ ПОЛЯ НА РОЗСІЯННЯ ЕЛЕКТРОМАГНІТНОЇ ХВИЛІ МЕТАЛЕВИМ НАНОЕЛІПСОЇДОМ

Р е з ю м е

У рамках кінетичного підходу досліджено вплив магнітного дипольного моменту асиметричної металевої наночастинки на розсіяння електромагнітного випромінювання. Для частинок сфероїдальної форми отримані аналітичні вирази для перерізу розсіяння та проаналізована їхня залежність від довжини хвилі падаючого випромінювання і ексцентриситету сфероїда. Показано, що в діапазоні частот, далеких від плазмових резонансів, внесок магнітного моменту в розсіяння одного порядку величини з електричним, причому відношення магнітного розсіяння до електричного стає максимальним, коли частинка має форму сфери. Крім того, всі розрахунки виконані для довільного співвідношення між розміром частинки і довжиною вільного пробігу електрона, що дає можливість порівняти результати з теорією Мі у випадку домінуючої ролі розсіяння електронів в об'ємі наночастинки.