GENERATION OF SOUND BY METAL NANOCLUSTERS IN A DIELECTRIC MATRIX

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We develop the theory of the photo-acoustical effect caused by a laser action on metal nanoclusters embedded in a dielectric matrix. The energy absorbed by clusters propagates through the dielectric matrix and generates sound waves in it by the thermodeformation mechanism. The formulas for an acoustical signal are derived, and the high sensitivity of the sound wave amplitude to the shape of metal clusters, as well to such parameters of a laser irradiation as the frequency, polarization, and intensity, is revealed. The behavior of the amplitude of sound vibrations in a region of the absorption of surface plasmons is studied in detail. It is found that this amplitude at the light absorption by a discrete metal film (a system of clusters in the matrix) can exceed the corresponding amplitude for the absorption by a continuous metal film in the region of plasmon resonances by several orders of magnitude.

1. Introduction

At the irradiation of a metal nanocluster by a laser-generated light beam, hot electrons appear in the cluster. The presence of hot electrons causes the additional pressure of the electron gas on the cluster surface and induces a heat flow from the cluster to the environment. If a metal nanocluster (MN) or a system of such nanoclusters is positioned into a dielectric matrix, then both above-indicated factors, the additional pressure and the heat flow, can generate sound in the matrix. This occurs in the case where a laser-generated light beam is nonstationary (for example, a short laser pulse or a stationary laser-generated light beam modulated with a low frequency). Such optoacoustic effects in a system of MNs positioned into a transparent matrix (or on its surface) were studied in [1, 2]. In particular, a thermodeformation mechanism of sound generation in a matrix by the modulated heat flow caused by the energy transfer from hot electrons of the cluster to the dielectric matrix was considered. Work [1] presented, for the first time, the mechanism of sound generation by a modulated electron pressure which appears in MN due to a change of the electron temperature. A change of the pressure induces oscillations of the MN surface, and the oscillations of the surface, in turn, generates sound in the matrix. Later on, the same mechanism of sound generation was proposed in [3].

In addition to acoustic oscillations of the matrix, radial oscillations in MN itself are of significant interest, since a periodic change of the MN radius in the course of time leads to oscillations in the relaxation dynamics of the electron temperature (see, e.g., [4]).

In [5], a theory of the absorption of the energy of electromagnetic waves depending on the shape and size of small metal particles was constructed. The high sensitivity of the absorption to the shape of a particle and to the polarization of a wave was established.

In the present work, we intend to study optoacoustic effects related to identical MNs with the spheroidal shape. We assume that such MNs have the same orientation. This can be attained, for example, if MNs are positioned in a liquid crystal. The absorption of such a system by MNs depends on the laser radiation polarization and will manifest itself in the dependence of the acoustic signal amplitude in the matrix on the light polarization. These effects will be considered in what follows.

We note else that similar optoacoustic effects can occur also in the case where the absorbing objects are semiconducting clusters incorporated into transparent solutions [6].

2. Statement of the Problem

The generation and propagation of long-wave acoustic oscillations in a dielectric matrix are described by the equation of motion (see, e.g., [7])

$$\rho\frac{\partial^2}{\partial t^2}u_i = \sum_j \frac{\partial}{\partial x_j}\sigma_{ij}, \tag{1}$$

In (1), $\mathbf{u}_i(\mathbf{r},t)$ is a component of the displacement vector, ρ is the mass density of the matrix, σ_{ij} are components of the stress tensor, t is the time, and x_j are components of the coordinate vector $\mathbf{r}$. With regard for the temperature dependence, the components σ_{ij} have the form [7]

$$\sigma_{ij} = K\left\{\sum_\alpha u_{\alpha\alpha} - \alpha\left(T_l - T_0\right)\right\}\delta_{ij}+$$

$$+2\mu\left\{u_{ij} - \frac{1}{3}\delta_{ij}\sum_\alpha u_{\alpha\alpha}\right\}, \tag{2}$$

where K and μ are, respectively, the moduli of uniform compression and shear, and α is the constant of thermal expansion. In addition, the strain tensor in the case of small deformations can be written as

$$u_{ij} = \frac{1}{2}\left(\frac{\partial}{\partial x_i}u_j + \frac{\partial}{\partial x_j}u_i\right). \tag{3}$$

In (2), T_l is the temperature of the lattice at the given point, and T_0 is some given temperature at a remote distance from MN, at which no deformation is present. If we write the displacement vector $\mathbf{u}$ in the form of a sum of the vectors of longitudinal $\mathbf{u}_L$ and transverse $\mathbf{u}_T$ displacements,

$$\mathbf{u} = \mathbf{u}_L + \mathbf{u}_T, \qquad \nabla\times\mathbf{u}_L = 0, \qquad \nabla\mathbf{u}_T = 0, \tag{4}$$

then relations (1)–(3) yield

$$\nabla^2\mathbf{u}_L - \frac{1}{s_L^2}\frac{\partial^2}{\partial t^2}\mathbf{u}_L = \frac{3K\alpha}{3K+4\mu}\nabla T_l. \tag{5}$$

In (5),

$$s_L = \left(\frac{3K+4\mu}{3\rho}\right)^{1/2} \tag{6}$$

is the velocity of longitudinal sound. In a similar way for transverse acoustic waves, we obtain

$$\nabla^2\mathbf{u}_T - \frac{1}{s_T^2}\frac{\partial^2}{\partial t^2}\mathbf{u}_T = 0, \qquad s_T = \sqrt{\frac{\mu}{\rho}}, \tag{7}$$

where s_T is the velocity of transverse sound. As is seen, the temperature gradient does not generate transverse acoustic oscillations in a medium which is described only by two moduli K and μ.

Let us introduce a scalar potential Ψ via

$$\mathbf{u}_L = \nabla\Psi, \tag{8}$$

Then Eq. (5) yields the following equation for it:

$$\nabla^2\Psi(\mathbf{r},t) - \frac{1}{s_L^2}\frac{\partial^2}{\partial t^2}\Psi(\mathbf{r},t) = \frac{3K\alpha}{3K+4\mu}\left[T_l(\mathbf{r},t) - T_0\right]. \tag{9}$$

A partial solution of this inhomogeneous equation has the form of a retarded potential [8]

$$\Psi(\mathbf{r},t) = \frac{1}{4\pi}\frac{3K\alpha}{3K+4\mu}\int\frac{\delta T_l(\mathbf{r}, t - |\mathbf{r}-\mathbf{r}'|/s_L)}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}', \tag{10}$$

where $|\mathbf{r}-\mathbf{r}'|$ is the distance from the observation point, at which we seek a value of the potential, to the volume element $dV' = d\mathbf{r}'$. In (10), we introduced the notation

$$\delta T_l(\mathbf{r}',t) = T_l(\mathbf{r}',t) - T_0. \tag{11}$$

As for the solution of the homogeneous equation corresponding to (9), this solution describes, as was shown in [1], the sound generation by oscillations of the MN surface. Such a mechanism of sound generation becomes significant [1] under the action of short, but powerful laser pulses. Further, we will consider a situation where MN undergoes the action of a continuous laser-generated light beam, whose energy per unit volume of MN is modulated by acoustic oscillations with a low frequency ω_{ac}:

$$I = I_0(1+\cos\omega_{\text{ac}}t). \tag{12}$$

In the situation where the light intensity varies smoothly, we can neglect the sound generation caused by oscillations of the MN surface.

3. Heat Flows

In order to move further, we need to determine the distribution of temperatures $T(\mathbf{r},t)$ which determines, according to (9), an acoustic signal.

First, we consider a single MN which has the shape of an ellipsoid of rotation and is located in the dielectric matrix. Let such an MN be irradiated by a laser-generated light beam.

In the general case, the balance equations used for the determination of the electron temperature T_e of a cluster and the temperature of the metal lattice T_m can be written as a system of two differential equations (see, e.g., [9])

$$\begin{cases} C_e(T_e)\frac{\partial}{\partial t}T_e = \mathrm{div}(K_e\nabla T_e) - g(T_e - T_l) + Q, \\ C_l\frac{\partial}{\partial t}T_l = \mathrm{div}(K_l\nabla T_l) - g(T_e - T_l), \end{cases} \tag{13}$$

where C_e, C_l, and K_e, K_l are, respectively, the specific heat capacities and the coefficients of heat conduction of electrons and the lattice of MN, g is the constant of electron-phonon energy exchange, the term $g(T_e - T_l)$ characterizes the energy transferred by electrons to the lattice per unit time, and Q is the energy absorbed by electrons in unit volume of MN per unit time.

In addition to Eqs. (13) which describe heat processes in MN, it is necessary to write else the equation for the temperature of the dielectric matrix surrounding MN. This equation has the form

$$C_m\frac{\partial}{\partial t}T_m = \mathrm{div}\left(K_m\nabla T_m\right), \tag{14}$$

C_m and T_m are, respectively, the heat capacity and the coefficient of heat conduction of the matrix surrounding MN. Since it is assumed that the electrons do not leave MN (for a dielectric), only phonons transfer heat to the matrix. Therefore, the solutions of Eq. (14) and the second equation in (13) and the corresponding heat flows must be "sewed" on the boundary of MN. To this end, we could write the corresponding boundary conditions for these equations. But we choose another way instead of the solution of (13) and (14) and the mentioned procedure of "sewing." For small metal islands under consideration, their sizes are less than the free path of an electron, and the temperature distributions T_e and T_l over coordinates inside an island are not significant. Therefore, we can restict ourselves by the solution of only one equation,

$$C_m\frac{\partial}{\partial t}T_m = \mathrm{div}\left(K_m\nabla T_m\right) + G(\mathbf{r},t) \tag{15}$$

with the function

$$G(\mathbf{r},t) = \begin{cases} g(T_e - T_l), & \mathbf{r}\in V, \\ 0, & \mathbf{r}\notin V, \end{cases} \tag{16}$$

which does not depend on coordinates in the volume V of MN and is equal to zero outside it. Equation (15) describes the nonuniform heat conduction of an inhomogeneous isotropic body. It is obvious that the above-described approach is exact in the case where the quantities C_l and K_l coincide, respectively, with C_m and K_m. But if such a coincidence is absent, this approach describes the process only approximately. However, this approach is quite proper for small MNs with the almost homogeneous temperature distributions T_e and T_l inside them. In what follows, we consider that K_m is independent of the coordinates. Then the general solution of (15) has the form (see, e.g., [10, 11])

$$T_m(\mathbf{r},t) - T_0 = \frac{\varkappa_m}{\pi^{3/2}K_m}\int\limits_{-\infty}^{t} dt'\times$$

$$\times\int\limits_V \frac{G(\mathbf{r}',t')}{[4\varkappa_m(t-t')]^{3/2}}\exp\left[-\frac{|\mathbf{r}-\mathbf{r}'|^2}{4\varkappa_m(t-t')}\right]d\mathbf{r}', \tag{17}$$

where $\varkappa_m/K_m = C_m$. The integration in (17) over t' is executed from $-\infty$, because we consider that the source $G(t)$, which sets the initial conditions for the homogeneous equation, is switched-on at the time moment $t' = -\infty$, when

$$T_m(\mathbf{r},t)|_{t=-\infty} = T_0.$$

To determine the explicit formula for $G(\mathbf{r},t)$, we use Eq. (13). We recall that $T_l = T_m$ at $\mathbf{r}\in V$ in the case where Eq. (15) is valid. We assume again that the free path of an electron is larger than the size of MN. Then we can omit the gradient in (13) (electron temperature is invariable for the whole MN). For such sizes of a cluster, Eq. (13) yields

$$g(T_e - T_l) = Q - C_e(T_e)\frac{\partial}{\partial t}T_e. \tag{18}$$

For a stationary laser-generated light beam, the last term on the right-hand side of (18) vanishes. But, in our case, the laser beam intensity is modulated, according to (12), with a low (acoustic) frequency ω_{ac}. Therefore, we get

$$\frac{\partial}{\partial t}T_e \sim \omega_{\mathrm{ac}}T_e. \tag{19}$$

Since the heat capacity of the electron gas C_e is small (due to its degeneration) as compared with the heat capacity of the lattice, and the frequency ω_{ac} is low, we easily prove the inequality

$$Q \gg \omega_{\mathrm{ac}}C_e(T_e)T_e. \tag{20}$$

We consider that inequality (20) is valid in our case. Then, according to (16) and (18)–(20), we obtain

$$G = g(T_e - T) = Q(\mathbf{r}, t). \tag{21}$$

The energy absorbed by a cluster per unit time can be written in the form

$$VQ = cS_{\rm ab}I, \tag{22}$$

where $S_{\rm ab}$ is the absorption cross-section of the cluster. In view of (12) and (22), we can write

$$Q(\mathbf{r}, t) = Q_0(\mathbf{r})\wp(t), \tag{23}$$

where

$$\wp(t) = \frac{c}{V} S_{\rm ab} I_0 (1 + \cos\omega_{\rm ac}t), \tag{24}$$

$$Q_0(\mathbf{r}) = \begin{cases} 1, & \mathbf{r} \in V, \\ 0, & \mathbf{r} \notin V. \end{cases}$$

By substituting formula (23) in (17), we obtain

$$T(\mathbf{r}, t) = T_0 + \frac{\varkappa_m}{\pi^{3/2}K_m} \int\limits_0^\infty d\tau \frac{\wp(t-\tau)}{(4\varkappa_m\tau)^{3/2}} \times$$

$$\times \int\limits_V \exp\left[-\frac{|\mathbf{r}-\mathbf{r}'|^2}{4\varkappa_m\tau}\right] d\mathbf{r}'. \tag{25}$$

After the Fourier transformation, relation (25) becomes

$$T(\mathbf{r}, \omega) = \frac{\varkappa_m}{\pi^{3/2}K_m}\wp(\omega) \int\limits_0^\infty d\tau \frac{e^{i\omega\tau}}{(4\varkappa_m\tau)^{3/2}} \times$$

$$\times \int\limits_V \exp\left[-\frac{|\mathbf{r}-\mathbf{r}'|^2}{4\varkappa_m\tau}\right] d\mathbf{r}'. \tag{26}$$

In (26), the integral over τ can be calculated with the use of the formula [12]

$$\int\limits_0^\infty \frac{dx}{x^{3/2}} e^{-q/x} e^{ibx} = \sqrt{\frac{\pi}{q}} \exp\left(-2\sqrt{-ibq}\right). \tag{27}$$

As a result, we get

$$T(\mathbf{r}, \omega) = \frac{\wp(\omega)}{4\pi K_m} \times$$

$$\times \int\limits_V \frac{d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|} \exp\left[-(1-i)\sqrt{\frac{\omega}{2\varkappa_m}}|\mathbf{r}-\mathbf{r}'|\right], \tag{28}$$

where we used the relation $\sqrt{-i} = \frac{\sqrt{2}}{2}(1-i)$.

We now consider nanoclusters with the shape of an ellipsoid of rotation with curvature radii $R_\parallel$ (along) and $R_\perp$ (normally to the rotation axis) and with volume $V = \frac{4\pi}{3}R_\parallel R_\perp^2$. At distances from a cluster greater than its sizes, i.e.,

$$r \gg \max\left\{R_\parallel, R_\perp\right\}, \tag{29}$$

we can approximately write

$$|\mathbf{r}-\mathbf{r}'| \approx r - r'\cos\vartheta, \tag{30}$$

where ϑ is the angle between $\mathbf{r}$ and $\mathbf{r}'$. Integral (28) can be calculated in this approximation. But let us assume that, in addition to (29), the inequality

$$r'\sqrt{\frac{\omega}{2\varkappa_m}} \le \sqrt{\frac{\omega}{2\varkappa_m}} \max\left\{R_\parallel, R_\perp\right\} < 1, \tag{31}$$

holds for low frequencies $\omega \sim \omega_{\rm ac}$. Then formula (28) takes a very simple form:

$$T(\mathbf{r}, \omega) = \frac{V}{4\pi|\mathbf{r}|}\frac{\wp(\omega)}{K_m} \exp\left[-(1-i)\sqrt{\frac{\omega}{2\varkappa_m}}|\mathbf{r}|\right]. \tag{32}$$

It is seen from (32) and (24) that, at far (as compared with the sizes of MN) distances, the temperature generated in a dielectric matrix depends on the shape of MN only through the absorption cross-section $S_{\rm ab}$. But $S_{\rm ab}$, as we showed in [5], depends on the shape of MN quite significantly.

4. Sound Generation

Having determined the distribution of temperatures in the dielectric matrix, we now turn to relation (10) which describes, according to (9), longitudinal acoustic oscillations. In (10), we carry out the Fourier transformation:

$$\Psi(\mathbf{r}, \omega) = \frac{1}{4\pi}\frac{3K\alpha}{3K+4\mu} \int \frac{d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|} T(\mathbf{r}', \omega) e^{i\omega|\mathbf{r}-\mathbf{r}'|/s_L}. \tag{33}$$

By substituting $T(\mathbf{r}', \omega)$ from (32) to the last formula, we obtain

$$\Psi(\mathbf{r}, \omega) = \frac{V}{(4\pi)^2}\frac{\wp(\omega)}{K_m}\frac{3K\alpha}{3K+4\mu} \times$$

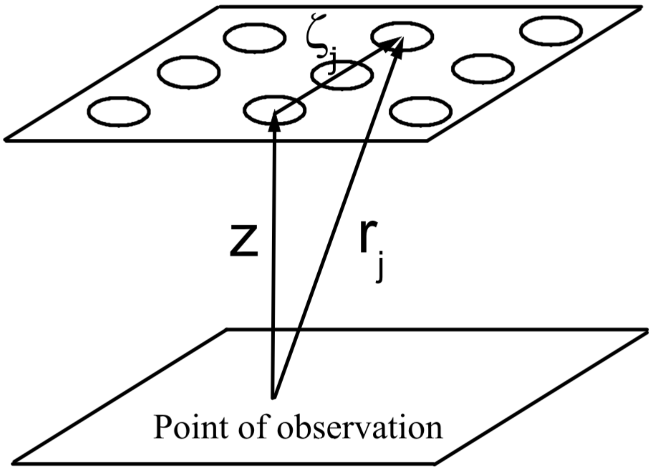

Fig. 1. Scheme of the arrangement of a point of observation and a plane of metal spheroidal clusters

$$\times \int \frac{d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'||\mathbf{r}'|} e^{-(1-i)\sqrt{\omega/(2\varkappa_m)}|\mathbf{r}'|} e^{i\omega|\mathbf{r}-\mathbf{r}'|/s_L}. \quad (34)$$

As is seen from the integrand of (34), the actual region of integration over $\mathbf{r}'$ is determined by the relation

$$|\mathbf{r}'| \leq \sqrt{2\varkappa_m/\omega} \equiv |\mathbf{r}_0|. \quad (35)$$

Therefore, at distances $r > r'$ from MN, we can set $|\mathbf{r}-\mathbf{r}'| \approx r - r'\cos\vartheta'$, where ϑ' is the angle between $\mathbf{r}$ and $\mathbf{r}'$. In this approximation, integral (34) is easily calculated, and we obtain

$$\Psi(\mathbf{r},\omega) = \frac{V}{4\pi}\frac{\alpha\varkappa_m}{K_m}\frac{K}{\rho s_L^2}\frac{\wp(\omega)}{\omega}\frac{s_L^2 + i\varkappa_m\omega}{s_L^2 + (\varkappa_m\omega/s_L)^2}\frac{e^{i\omega|\mathbf{r}|/s_L}}{|\mathbf{r}|}. \quad (36)$$

According to (24), the Fourier-component $\wp(\omega)$ reads

$$\wp(\omega) = 2\pi c\frac{S_{\rm ab}I_0}{V}\left\{\delta(\omega) + \frac{1}{2}\left[\delta(\omega-\omega_{\rm ac}) + \delta(\omega+\omega_{\rm ac})\right]\right\}. \quad (37)$$

By substituting relation (37) in (36) and by performing the inverse Fourier transformation, we have

$$\Psi(\mathbf{r},t) = \frac{c}{4\pi}\frac{I_0}{\omega_{\rm ac}}\frac{\alpha\varkappa_m}{K_m}\frac{K}{\rho s_L^2}\frac{S_{\rm ab}}{\sqrt{1+(\varkappa_m\omega_{\rm ac}/s_L^2)^2}}\times$$

$$\times\frac{\cos[\omega_{\rm ac}(t-|\mathbf{r}|/s_L)-\delta]}{|\mathbf{r}|}, \quad (38)$$

where the phase δ is determined by the relation

$\delta = \varkappa_m\omega_{\rm ac}/s_L^2$.

In (38), we omit the first term with $\delta(\omega)$, because it is not related to the sound generation. It is seen that MN with asymmetric shape generates a spherical acoustic wave at a remote distance (as compared with the sizes of MN). The asymmetry of MN absorbing a laser radiation affects (and very significantly) only the acoustic wave amplitude. This effect, i.e. the influence of the shape of MN on the acoustic effect, is especially clearly manifested in the case where we deal with a system of MNs of the same shape, size, and orientation. For a system of MNs in the dielectric matrix, formula (38) is replaced by

$$\Psi(\mathbf{r},t) = \frac{c}{4\pi}\frac{K}{\rho s_L^2}\frac{I_0}{\omega_{\rm ac}}\frac{\alpha}{\sqrt{1+(\varkappa_m\omega_{\rm ac}/s_L^2)^2}}\times$$

$$\times\sum_j\frac{\varkappa_m}{K_m}S_{\rm ab}^{(j)}\frac{\cos\left[\omega_{\rm ac}(t-|\mathbf{r}-\mathbf{r}_j|/s_L)-\delta\right]}{|\mathbf{r}-\mathbf{r}_j|}. \quad (39)$$

Relation (39) is written in the general case where the absorption cross-sections of different MNs $S_{\rm ab}^{(j)}$ and their coefficients of heat conduction and thermal diffusivity are different.

5. System of Identical Nanoclusters

We now consider a system of MNs of the same shape and size which are positioned on a single plane in the matrix (see Fig. 1).

At a distance remote as compared with that between MNs, the shape of an acoustic signal depends weakly on the specific arrangement of MNs in the plane, but it strongly depend on their orientation.

In what follows, we will consider identical MNs of the spheroidal shape. Therefore, we assume, in order to simplify calculations, that the centers-of-mass of MNs are positioned on a square lattice (with a lattice constant a), and their rotation axes are parallel to one another. Under such an assumption, values of the vector $\mathbf{r}_j$ in formula (39) can be written in the form $\mathbf{r}_j = (n_j a, m_j a, 0)$, where n_j and m_j are integers. Then the sum over j in (39) in the polar coordinate system can be approximately replaced by the integral

$$\sum_j\frac{\cos\left[\omega_{\rm ac}\left(t-|\mathbf{r}-\mathbf{r}_j|/s_L\right)-\delta\right]}{|\mathbf{r}-\mathbf{r}_j|} \approx$$

$$\approx\frac{2\pi}{a^2}\int\limits_0^\infty\frac{\cos\left[\omega_{\rm ac}\left(t-\sqrt{z^2+\zeta^2}/s_L\right)-\delta\right]}{\sqrt{z^2+\zeta^2}}\zeta d\zeta =$$

$$= \frac{2\pi}{a^2} \int_z^\infty \cos\left[\omega_{\rm ac}\left(t - \frac{\xi}{s_L}\right) - \delta\right] d\xi =$$

$$= \frac{2\pi}{a^2} \frac{s_L}{\omega_{\rm ac}} \sin\left[\omega_{\rm ac}\left(t - \frac{z}{s_L}\right) - \delta\right]. \quad (40)$$

Here, we took the relation $|\mathbf{r} - \mathbf{r}_j| = \sqrt{z^2 + \zeta^2}$ into account and made change of the variables $\xi = \sqrt{z^2 + \zeta^2}$. To avoid a possible misunderstanding, we note that integral (40) has no exact value on the upper limit. This uncertainty can be eliminated, by considering the effect of damping of sound at remote distances from the source (this can be formally realized, by adding an imaginary term to the sound velocity s_l). By substituting relation (40) in (39) and by introducing the surface density of MNs $N_0 = 1/a^2$, we obtain

$$\Psi(\mathbf{r}, t) = \frac{1}{2} \frac{KI_0}{\rho s_L^2} \frac{\alpha \varkappa_m}{K_m} \frac{c s_L}{\omega_{\rm ac}^2} \frac{N_0 S_{\rm ab}}{\sqrt{1 + (\varkappa_m \omega_{\rm ac}/s_L^2)^2}} \times$$

$$\times \sin\left[\omega_{\rm ac}\left(t - \frac{z}{s_L}\right) - \delta\right]. \quad (41)$$

Formula (41) is similar to that for an acoustic wave in the case of a continuous film which absorbs light [11]. But the light absorption which is set by the quantity $S_{\rm ab} N_0$ in (41) is anisotropic and depends on the frequency of a laser beam in a complicated way. Respectively, the amplitude of the z-th component of the vector of a sinusoidal displacement, by (40) and (8), reads

$$A_{zL} = -\frac{1}{2} \frac{c}{\omega_{\rm ac}} \frac{\alpha \varkappa_m}{K_m} \frac{KI_0}{\rho s_L^2} \frac{N_0 S_{\rm ab}}{\sqrt{1 + (\varkappa_m \omega_{\rm ac}/s_L^2)^2}}. \quad (42)$$

It follows from relation (42) that the acoustic waves with lower frequencies $\omega_{\rm ac}$ cause greater displacement amplitudes.

Earlier [5, Eq. (83)], we obtained the formula for the energy absorbed by MN of the spheroidal shape per unit time under its irradiation by a monochromatic electromagnetic wave with frequency ω:

$$W \equiv VQ = \frac{V}{2} \sum_{j=1}^{3} \frac{\sigma_{jj} (\varepsilon_m \omega^2 / g_j)^2 |E_j^{(0)}|^2}{(\omega^2 - \omega_j^2)^2 + (4\pi L_j \sigma_{jj}/g_j)^2 \omega^2}. \quad (43)$$

In (42), σ_{jj} is the corresponding diagonal component of the tensor of high-frequency conduction, ε_m is the permittivity of the matrix, and L_j is the factor of depolarization. In addition,

$$\omega_j^2 = \frac{L_j}{g_j} \omega_{\rm pl}^2 \quad (44)$$

is the square of the frequency of a plasmon resonance, $\omega_{\rm pl}$ is the frequency of plasma oscillations of electrons, and

$$g_j = \varepsilon_m + L_j(1 - \varepsilon_m). \quad (45)$$

For media with $\varepsilon_m = 1$, we obviously have $g_j = 1$. In (43), $E_j^{(0)}$ is the j-th component of the amplitude of an electromagnetic wave which was described in [5] as

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}^{(0)} e^{i(\mathbf{kr} - \omega t)}, \quad (46)$$

where $\mathbf{k}$ is the wave vector. Formula (43) is written in the general case for a three-axis ellipsoid. In this case, MN is characterized by three plasmon resonances at frequencies $\omega_j (j = 1, 2, 3)$.

In order to study the role of an anisotropy of MN on the process of sound generation, we consider the simplest case below. Namely, let MN have the spheroidal shape. By z, we denote the rotation axis. Then we have $\sigma_{xx} = \sigma_{yy} \equiv \sigma_\perp$; $\sigma_{zz} = \sigma_\|$; $L_x = L_y \equiv L_\perp$; $L_z = L_\|$; $g_x = g_y \equiv g_\perp$; $g_z = g_\|$. Крім того, $\omega_x = \omega_y \equiv \omega_\perp$; $\omega_z = \omega_\|$. The graphic dependence of these frequencies on the degree of oblateness or elongation of MN positioned in a glass matrix with $\varepsilon_m = 7$ is shown in Fig. 2. The formulas for the factors of depolarization $L_\|$ and $L_\perp$ in (44) can be found, e.g., in [5].

Using the above-presented notation and taking the relations

$$W = S_{\rm ab} \left(\frac{c}{8\pi} \sqrt{\varepsilon_m} |\mathbf{E}^{(0)}|^2\right), \quad (47)$$

and (43) into account, we obtain the following formula for the absorption cross-section:

$$S_{\rm ab} = 4\pi V \frac{\varepsilon_m^{3/2}}{c} \omega^4 \left\{ \frac{(\sigma_\| / g_\|^2) \cos^2\theta}{(\omega^2 - \omega_\|^2)^2 + (4\pi L_\| \sigma_\| / g_\|)^2 \omega^2} + \right.$$

$$\left. + \frac{(\sigma_\perp / g_\perp^2) \sin^2\theta}{(\omega^2 - \omega_\perp^2)^2 + (4\pi L_\perp \sigma_\perp / g_\perp)^2 \omega^2} \right\}, \quad (48)$$

Here, V is the volume of MN, θ is the angle between the rotation axis of the spheroid and the unit vector of the polarization of an electromagnetic wave, and components of the tensor of conduction at frequencies $\omega \gg \nu$ (ν is the frequency of electron collisions) are

$$\sigma_{\binom{\|}{\perp}}(\omega) = \frac{9}{32\pi} \left(\frac{\omega_{\rm pl}}{\omega}\right)^2 \frac{\upsilon_F}{R_\perp} \binom{\eta(e_s)}{\rho(e_s)}, \quad (49)$$

where the functions $\eta(e_s)$ and $\rho(e_s)$ dependent of the spheroid eccentricity e_s can be found in the explicit analytic form, for example, in [5]. For MN of the spherical

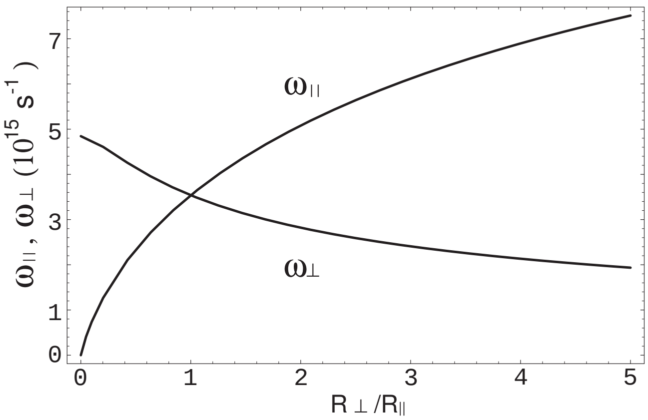

Fig. 2. Plasmon frequencies $\omega_\parallel$ and $\omega_\perp$ of dipole oscillations of electrons, respectively, along (continuous curve) and across (dashed curve) the rotation axis of a spheroid versus the degree of oblateness or elongation of a gold spheroidal nanocluster

shape, $\eta(0) = \rho(0) \equiv 2/3$. The formula similar to (48) also describes well the energy absorption by a metal particle in the case where the width of incident laser pulses is large [13].

Formula (48) determines both the frequency and the polarization dependence of the absorption. The factors of depolarization $L_\parallel$ and $L_\perp$ define the dependence of the absorption on the shape of MN and, according to (44), determine also the positions of plasma resonances. For a spherical MN, $L_\parallel = L_\perp \equiv 1/3$.

6. Discussion of Results

Using (43), we consider that the main contribution to the absorption cross-section is given by plasma resonances. The half-width of these resonances for the polarizations along and across the rotation axis of the spheroid is given by the formula

$$\gamma_{\left(\begin{smallmatrix}\parallel\\ \perp\end{smallmatrix}\right)}(\omega) = 2\pi L_{\left(\begin{smallmatrix}\parallel\\ \perp\end{smallmatrix}\right)}\sigma_{\left(\begin{smallmatrix}\parallel\\ \perp\end{smallmatrix}\right)}(\omega). \tag{50}$$

This half-width is a significant physical characteristic, because it reflects the type of interactions in the system.

Above we obtained formula (41) for a scalar potential which defines an acoustic displacement according to (8). In this case, we considered that the laser radiation is absorbed by a system of identical spheroidal MNs, whose rotation axes are parallel to one another and lie in the same plane. The heat flow from these MNs generates sound in the dielectric matrix. If a continuous metal film absorbing the laser radiation would be on the surface of the dielectric matrix instead of a system of MNs, we would obtain a formula for the scalar potential Ψ similar to (41). But the former will include the quantity corresponding to the absorption of a continuous film instead of the product $N_0S_{\rm ab}$ which determines the share of a laser-generated light beam absorbed by MN. By definition, the share of the energy absorbed by a continuous metal film under condition of the light transmission tending to zero is equal to

$$\eta \approx \frac{I - I_R}{I} = 1 - R, \tag{51}$$

where I_R is the intensity of a reflected laser beam, and R is the share of the energy of a reflected radiation. Formula (51) requires also the assumption that the film thickness exceeds the skin depth. Otherwise, the share of the absorbed energy would be higher.

As is known, at the normal incidence of light incoming from vacuum, we have

$$R = \frac{(n-1)^2 + \kappa^2}{(n+1)^2 + \kappa^2}, \tag{52}$$

where n and κ are defined by the relation

$$\sqrt{\epsilon(\omega)} = n + i\kappa, \tag{53}$$

and the permittivity of a metal takes the form

$$\epsilon(\omega) = \epsilon'(\omega) + i\epsilon''(\omega) \simeq 1 - \left(\frac{\omega_{\rm pl}}{\omega}\right)^2 + i\frac{\nu}{\omega}\left(\frac{\omega_{\rm pl}}{\omega}\right)^2. \tag{54}$$

In (54), ν is the frequency of electron-phonon collisions. In this case, it is assumed that the frequency ω belongs to the interval

$$\nu < \omega < \omega_{\rm pl}. \tag{55}$$

Let us use formulas (51)–(54), inequality (55), and the assumption that[1] $\kappa \gg n$. Then, for a continuous metal film, we obtain

$$\eta \approx 2\frac{\nu}{\omega_{\rm pl}}. \tag{56}$$

The evaluation for a film fabricated, for example, from gold gives $\eta \approx 0.006$.

Thus, for a continuous metal film on the surface of a dielectric matrix, we would obtain relation (41) for Ψ, in which $2\nu/\omega_{\rm pl}$ would stand instead of $N_0S_{\rm ab}$. In view of the above discussion, it is expedient to normalize the amplitude of acoustic oscillations (42) by an analogous amplitude characteristic of a continuous metal film $A_{\rm film}$. The ratio of these amplitudes is as follows:

$$A = \frac{A_{zL}}{A_{\rm film}} = \frac{\omega_{\rm pl}}{2\nu}N_0S_{\rm ab}. \tag{57}$$

[1] For example, $n = 0.26$ and $\kappa = 2.16$ for gold [14].

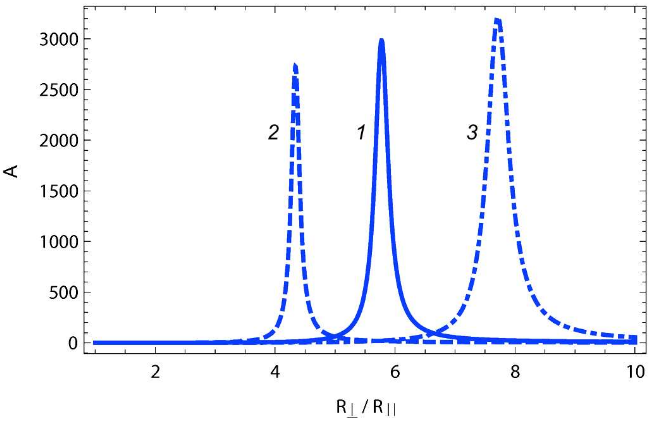

Fig. 3. Amplitude of sound oscillations of the matrix versus the degree of oblateness or elongation of a spheroidal MN with a volume equal to the volume of a sphere with the radius $R = \sqrt[3]{R_\perp^2 R_\parallel} =$ 200 Å at the frequencies of the plasmon resonance $\Omega - 0.1\Omega$ (dashed curve), Ω (continuous curve), and $\Omega + 0.1\Omega$ (dash-dotted curve) and at the incidence angle of a laser beam $\theta = \pi/4$

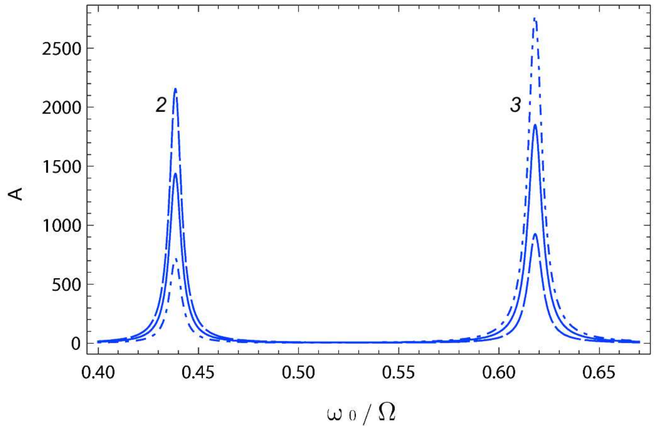

Fig. 4. Amplitude of acoustic oscillations of the matrix versus the frequency of a laser wave for various angles θ of its incidence on an oblate MN ($R_\perp/R_\parallel = 1.5$): $\pi/4$ (continuous curve); $\pi/3$ (dashed curve), and $\pi/6$ (dash-dotted curve). The volume of a spheroidal MN corresponds to the volume of a sphere with the radius $R = 200$ Å. The distance between MNs $a = 2000$ Å

In Fig. 3, we present the dependence of the ratio of sound amplitudes (57) at $\theta = \pi/4$ on the degree of oblateness or elongation of a spheroidal MN at the plasmon resonance frequency $\omega = \omega_{\rm pl}/\sqrt{3} \equiv \Omega$ characteristic of a particle of the spherical shape and at two other frequencies which are not much higher or lower than Ω. Here and below, the calculations of (57) are carried out with the use of formulas (42) and (48) for a gold particle in the glass matrix ($\varepsilon_m = 7$) with the following values of parameters: $\nu_{0^\circ{\rm C}} \simeq 3.39 \times 10^{13}$ s^{-1} [14] $n_e \simeq 5.9 \times 10^{22}$ cm^{-3} [15], $a = 2000$ Å. The other parameters were calculated by the formulas

$$\omega_{\rm pl} = \sqrt{4\pi n_e e^2/m}, \qquad \upsilon_F = \frac{2\pi\hbar}{m}\left(n_e\frac{3}{8\pi}\right)^{1/3}, \tag{58}$$

where e and m are, respectively, the charge and mass of an electron, and n_e is the concentration of electrons.

Curve *1* corresponds to the plasmon resonance which arise in a spherical MN positioned in the medium with the permittivity ε_m. For materials of the matrix with lower permittivities, the resonance shifts to the side lower ratios $R_\perp R_\parallel$ and approaches $R_\perp/R_\parallel = 1$ as $\varepsilon_m \to 1$. The most intense sound signal is observed, obviously, at the plasmon frequencies. We have already established in out previous studies that the shape of MN is closely related to frequencies, at which it absorbs in the resonance manner [16]. The frequencies $\omega < \Omega$ (curve *2*) and $\omega > \Omega$ (curve *3*) correspond, respectively, to plasmon oscillations of electrons across and along the spheroid axis (see Fig. 2). Comparing curves *1–3*, we obtain that, for more and more oblate MNs, the characteristic resonance frequencies shift to the short-wavelength side of the spectrum. In this case, the half-width of the resonance curve increases proportionally to the ratio $R_\perp/R_\parallel$.

If we choose MN of a certain shape (oblate or elongated) and vary the carrier frequency of a laser, then formulas (42) and (48) yield that two sound waves (doublet) appear in MN of the oblate *spheroidal* shape in correspondence to two plasmon resonances observed in MN of such a shape (Fig. 4). The relative height of peaks in the doublet can be controlled by varying the incidence angle of an electromagnetic wave.

First, we take the incidence angle of the electromagnetic wave relative to the rotation axis of the spheroid to be equal to $\theta = \pi/4$. That is, this angle is such that plasmon (dipole) oscillations of electrons across and along the rotation axis of the spheroid can be excited to the same extent. In this case, we observe (curve *1*) that the less intense maximum located at lower frequencies corresponds to the plasmon resonance which arises in a spheroidal MN at the frequency $\omega_\perp$ across the rotation axis of the spheroid, and the more intense maximum at higher frequencies corresponds to the plasmon resonance at the frequency $\omega_\parallel$ along the rotation axis of the spheroid. For MN of the elongated shape, on the contrary, the less intense maximum would be at higher frequencies and would correspond to the frequency $\omega_\perp$. The different intensities of the peaks is caused by the corresponding frequency dependence of the factors of depolarization which are present in (48). As the incidence angle increases from $\pi/4$ to $\pi/3$ (curve *2*), the intensity of the sound amplitude peak increases at the frequency $\omega_\perp$ and decreases at the frequency $\omega_\parallel$. On the contrary, the incidence angle decreases from $\pi/4$ to $\pi/6$ (curve *3*), the intensity of the sound amplitude peak decreases at the frequency $\omega_\perp$ and increases at the frequency $\omega_\parallel$. It is obvious that, at the angle $\theta = 0^0$, the peak at the frequency $\omega_\perp$ disappears in correspondence with (48), and the intensity of the peak at the frequency $\omega_\parallel$ becomes maximum. Otherwise, at an angle of $\pi/2$, the peak at the frequency $\omega_\parallel$ disappears, and the peak at the frequency $\omega_\perp$ remains maximum.

By concluding, it is worth noting that, at the above-chosen sizes of MNs and distances between them, all MNs cover only 3% of the area, in which the centers-of-mass of MNs are positioned. In this case, the amplitude of sound waves generated by MNs exceeds the amplitude of waves which can arise under the same conditions in a continuous metal film on the surface of the dielectric matrix by several orders (in our case, by three orders). Such significant difference between optoacoustic properties of a discrete (island) metal film and a continuous metal film is related to the fact that the light absorption by discrete films reaches a maximum in the frequency range of plasmon resonances, whereas this frequency range corresponds to the region of the almost complete reflection of light for a continuous metal film.

7. Conclusions

We have developed the theory of acoustooptic phenomena for metal nanoclusters incorporated in a dielectric matrix which allows one to determine the amplitude of acoustic oscillations of the matrix at various polarizations of the incident electromagnetic wave. The obtained analytic formulas allow one to evaluate the heat flows between MNs and the matrix, to determine the temperature of the matrix at any time moment and at any distance from MN, and to find the dependence of the amplitude of an acoustic signal on elastic constants of the medium, intensity of a laser beam, and the cross-section of absorption by MN.

The case where the frequency of a laser beam is close to plasmon frequencies of a spheroidal MN is studied in detail. We have obtained the dependence of the amplitude of acoustic oscillations of the matrix on the degree of oblateness or elongation of a metal spheroidal cluster on the frequencies of a plasmon resonance.

As a function of the frequency of a laser beam, the amplitude of an acoustic wave in a spheroidal MN has two maxima with different intensities, as distinct from spherical MNs, where a single maximum is observed. This is caused by resonances which arise at the excitation by a laser at the frequencies of oscillations of a plasmon along and across the rotation axis of a spheroid. By the distance between the doublet peaks, we can estimate the degree of oblateness or elongation of MN. The intensity of the doublet peaks can be controlled by the variation of the incidence angle of a laser beam relative to the rotation axis of a spheroid.

We have revealed a significant difference of optoacoustic properties of discrete and continuous metal films on the surface of a transparent dielectric matrix in the region of plasmon resonances.

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ГЕНЕРАЦІЯ ЗВУКУ МЕТАЛЕВИМИ НАНОКЛАСТЕРАМИ В ДІЕЛЕКТРИЧНІЙ МАТРИЦІ

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Р е з ю м е

Побудовано теорію фотоакустичного ефекту, зумовленого дією лазерного опромінення на металеві нанокластери, інкорпоровані в діелектричну матрицю. Поглинута кластерами енергія поширюється у вигляді тепла в діелектричній матриці і генерує в ній згідно з термодеформаційним механізмом звукові хвилі. У роботі отримано формули для акустичного сигналу і виявлено високу чутливість амплітуди звукової хвилі до форми металевих кластерів, а також таких параметрів лазерного випромінювання, як частота, поляризація, інтенсивність. Детально досліджено поведінку амплітуди звукових коливань в області збудження поверхневих плазмонів. Знайдено, що ця амплітуда при поглинанні світла дискретною металевою плівкою (системою кластерів у матриці) в області плазмонних резонансів може на кілька порядків перевищувати відповідну амплітуду при поглинанні суцільною металевою плівкою.