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KINETIC THEORY OF ABSORPTION OF ULTRASHORT LASER PULSES BY ENSEMBLES OF METALLIC NANOPARTICLES UNDER CONDITIONS OF SURFACE PLASMON RESONANCE

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This paper presents a theory that allows one to calculate the energy absorbed by spheroidal metal nanoparticles when irradiated by ultrashort laser pulses of different duration in the region of surface plasmon resonance. Simple analytical expressions are obtained to calculate the absorption energy dependent on the frequency of carrier laser wave, on the pulse duration, and on the ratio between the axes of the ellipsoids. It is shown that at the frequency of the carrier (laser) wave, which coincides with that of the surface plasmon, the maximum absorption is observed for spherical nanoparticles. As the carrier frequency deviates from the surface plasmon one, two maxima appear in the absorption spectrum, dependent on the ratio of spheroidal axes: one corresponds to the elongated particles and the other to the flattened ones.

***Keywords**: metal nanoparticles, electron, plasmon, surface plasmon resonance, frequency, ultrashort laser pulses, energy, absorption*

INTRODUCTION

Metal nanoparticles are interesting objects from both the point of view of condensed matter physics and the practical point of view. An increase in local electric fields near nanoparticles, due to surface plasmon resonance [1], caused by laser radiation, makes them useful for use in solar energy [2], biology [3], and medicine [4]. Metal nanoparticles serve as a basis for a new direction in the development of electronics – nanoplasmonics [5]. Since the beginning of the XXI century intensive research has begun on the creation of an element base for integrated circuits on plasmons, the use of plasmons in energetics, and the creation of surface plasmon amplification (SPASER – surface plasmon amplification by stimulated emission of radiation) analogs of a laser in which plasmons are used instead of photons [6]. Even more interesting is the behavior of metal nanoparticles in the field of ultrashort laser radiation. First, the short duration of such pulses (of the order of 10^{-15} s) makes it possible to study the dynamics of electronic processes in metal nanoparticles, as well as all kinds of nonlinear optical phenomena. Second, the ultrashort laser pulse (ULP) contains almost all harmonics, including those that coincide with plasmon resonances, which make a major contribution to the absorption of light by metal nanoparticles. If the nanoparticle is spherically symmetric, it will be characterized by one plasmon resonance, and if the particle is ellipsoidal in shape, then there will be three such resonances. This feature of the absorption of ULP by metal nanoparticles will be studied in this paper. Another feature that effects the absorption is associated with the shape of nanoparticles. Earlier, it was shown that in the case of asymmetric metal nanoparticles, the optical conductivity, which is determined by the electrical absorption and the width of the plasmon resonance line, becomes a tensor value [8].

This paper considers the peculiarities of the absorption of ULP by metal nanoparticles with sizes much smaller than the free path of electrons in them. In particular, the energy absorbed by metal nanoparticles in the process of plasmon resonance was found, when the carrier frequency of ULI coincides with that of plasmon resonance, and when this frequency deviates from the resonant one.

MATERIALS AND METHODS

Let us consider the case when an ensemble of metal nanoparticles is irradiated with a laser pulse, the electric field of which is given by the following expression

$$\vec{E}(\vec{r},t)=\vec{E}_0\left[-\Gamma_0^2\left(t-\frac{\vec{k}_o\vec{r}}{\omega_0}\right)^2\right]\cos\left[\omega_0\left(t-\frac{\vec{k}_o\vec{r}}{\omega_0}\right)\right], \quad (1)$$

where ω_0 – is carrier frequency of the electromagnetic wave (laser pulse), $|\vec{k}_0|=\omega_0/c$, Γ_0 – is the value inverse to the duration of the laser pulse, $\vec{E}_0$ – is the maximum value of the electric field strength in the laser pulse.

In addition to the electrical component, the laser pulse also has a magnetic component, which can be found from the corresponding Maxwell equation:

$$\text{rot}\ \vec{E}(\vec{r},t)=-\frac{1}{c}\frac{\partial\vec{H}(\vec{r},t)}{\partial t}. \quad (2)$$

The simplest relationship between the Fourier components of these quantities looks like:

$$\vec{H}(\vec{r},\omega)=\vec{m}\times\vec{E}(\vec{r},\omega), \quad (3)$$

where $\vec{m}=\vec{k}_0/k_0$ – unitary vector.

Now we find the Fourier component of the electric field of the laser pulse (1), which can be written as follows:

$$\vec{E}(\vec{r},\omega)=\int_{-\infty}^{\infty}\vec{E}(\vec{r},t)e^{i\omega t}dt=\vec{E}_0\frac{\sqrt{\pi}}{2\Gamma_0}\exp\left[i\vec{k}_0\vec{r}\left(\frac{\omega}{\omega_0}\right)\right]\left\{\exp\left(-\frac{(\omega-\omega_0)^2}{4\Gamma_0^2}\right)+\exp\left(-\frac{(\omega+\omega_0)^2}{4\Gamma_0^2}\right)\right\}. \quad (4)$$

In the simplest case, when $\Gamma_0\to 0$, from (4) we will obtain:

$$\vec{E}(\vec{r},\omega)\big|_{\Gamma\to 0}=\pi\vec{E}_0\exp\left\{i\vec{k}_0\vec{r}\left(\frac{\omega}{\omega_0}\right)\right\}\left[\delta(\omega-\omega_0)+\delta(\omega+\omega_0)\right]. \quad (5)$$

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 the inequality $k_0R<<1$, then 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 [8]:

$$E_{in}^j(0,\omega)=\frac{E_0^j(0,\omega)}{1+L_j\left[\varepsilon(\omega)-1\right]}, \quad (6)$$

$$E_{vr}^x=i\frac{\omega R_z^2}{c}\left[\frac{z}{R_x^2+R_z^2}H_0^y(0,\omega)-\frac{y}{R_x^2+R_y^2}H_0^z(0,\omega)\right]. \quad (7)$$

Here $\varepsilon(\omega)=\varepsilon'(\omega)+i\varepsilon''(\omega)$ – is the complex dielectric constant of the metal nanoparticle, L_j – geometric factors (depolarization coefficients), $j=x,y,z$, R_x,R_y,R_z – are the half-axes 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 [9]

$$W=\int_{-\infty}^{\infty}w(t)dt=W_e+W_m=\frac{1}{2}\text{Re}\int_{-\infty}^{\infty}dt\int_V d\vec{r}\left[\vec{j}_e(\vec{r},t)\vec{E}_{in}^*(\vec{r},t)+\vec{j}_{vr}(\vec{r},t)\vec{E}_{vr}^*(\vec{r},t)\right] \quad (8)$$

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) one. Turning to the Fourier representation, relation (8) can be rewritten as follows:

$$W=\frac{1}{4\pi}\int_{-\infty}^{\infty}d\omega\,\mathrm{Re}\int_{V}d\vec{r}\Big[\vec{j}_e(\vec{r},\omega)\vec{E}^{*}_{in}(\vec{r},\omega)+ \\ +\vec{j}_{vr}(\vec{r},\omega)\vec{E}^{*}_{vr}(\vec{r},\omega)\Big]. \tag{9}$$

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 [10]:

$$\vec{j}(\vec{r},\omega)=2e\left(\frac{m}{2\pi\hbar}\right)^{3}\int_{-\infty}^{\infty}\vec{v}f_1(\vec{r},\vec{v},\omega)d\vec{v}, \tag{10}$$

where e – is the electron charge, m – is the electron mass, $f_1(\vec{r},\vec{v},\omega)$ – 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

$$f_1(\vec{r},\vec{v},\omega)=\int_{-\infty}^{\infty}f_1(\vec{r},\vec{v},t)\exp(i\omega t)dt, \tag{11}$$

can be found as a solution of the corresponding linearized Boltzmann kinetic equation which will be present in the following form:

$$(\gamma-i\omega)f_1(\vec{r},\vec{v},\omega)+\vec{v}\frac{\partial f_1(\vec{r},\vec{v},\omega)}{\partial\vec{r}}+ \\ +e\vec{v}\Big[\vec{E}_{in}(\omega)+\vec{E}_{vr}(\vec{r},\omega)\Big]\frac{\partial f_0(\varepsilon)}{\partial\varepsilon}=0. \tag{12}$$

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

Equation (12) 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

$$f_1(\vec{r},\vec{v},\omega)\big|_S=0,\quad v_n<0. \tag{13}$$

In (13), 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 [11]. Equation (12) is easiest to solve by moving to new variables

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

in which the ellipsoid takes the form equal in volume to a sphere with a radius $R=\sqrt[3]{R_{\|}R_{\perp}^{2}}$.

Dependent on the frequency of the laser wave, its polarization and the shape of the nanoparticles, the dominant mechanism of energy absorption in a nanoparticle can be both electrical and magnetic absorption [11]. On the other hand, dependent on how much the frequency of the incident laser wave differs from that of the plasmon resonance, individual or collective absorption mechanisms may be prevalent. In this paper, we consider the absorption of ultrashort laser pulses in the region of plasmon resonance. Thus, we will take into account the collective (plasmon) absorption mechanism, which is part of the electrical absorption. Therefore, magnetic absorption can be neglected by placing $\vec{E}_{vr}(\vec{r},\omega)=0$. In this case, the solution of equation (12) can be found as follows:

$$f_1(\vec{r},v',\omega)=-e\vec{v}'\vec{E}_{in}\frac{1-\exp\Big[-(\nu-i\omega)t'(\vec{r}',v')\Big]}{\gamma-i\omega}\frac{\partial f_0}{\partial\varepsilon}. \tag{14}$$

Within the zero approximation for a small parameter (kT/ε_F) in (14) $\frac{\partial f_0}{\partial\varepsilon}$ can be replaced by $-\delta(\varepsilon-\varepsilon_F)$; here ε_F – is Fermi energy. In (14) a notation is introduced:

$$t'(\vec{r}',\vec{v}')=\frac{1}{v'^{2}}\left[\vec{r}'\vec{v}'+\sqrt{\left(R^2-r'^{2}\right)v'^{2}+\left(\vec{r}'\vec{v}'\right)^{2}}\right]. \tag{15}$$

Now let us consider the energy absorption of laser radiation by metal nanoparticles under conditions of plasmon resonance. In this case, the predominant mechanism of energy absorption will be electrical absorption, because under conditions of plasmon resonance there is a collective mechanism of energy absorption. Thus, substituting (10) and (14) in (9), we can find the following expression for the total energy

absorbed by the metal particle under conditions of plasmon resonance:

$$W_e = \frac{V}{4\pi}\sum_{j=1}^{3}\int_{-\infty}^{\infty}\frac{\omega^4\sigma_{jj}(\omega)\left|\vec{E}_j^{(0)}(0,\omega)\right|^2}{\left(\omega^2-\omega_j^2\right)^2+\left[4\pi L_j\sigma_{jj}(\omega)\omega\right]^2}d\omega. \qquad (16)$$

If the plasmon attenuation is small ($\gamma(\omega)=4\pi L_j\sigma_{jj}(\omega)\to 0$), then in (16) we can use the following representation for the δ-function [12]:

$$\delta(x)=\frac{1}{\pi}\lim\nolimits_{\alpha\to 0}\frac{\alpha}{\alpha^2+x^2}. \qquad (17)$$

For our case $\alpha=4\pi L_j\sigma_{jj}(\omega)\omega,\quad x^2=\left(\omega^2-\omega_j^2\right)$. Having carried out in (16) integration on frequency by means of δ – function (17) we will obtain for the energy absorbed by a metal nanoparticle, the following expression

$$W_e \approx \frac{V}{64}\left(\frac{\omega_p}{\Gamma_0}\right)^2\sum_{j=1}^{3}\left|E_0^j\left(\exp\left[\frac{\left(\sqrt{L_j}\omega_p-\omega_0\right)^2}{4\Gamma_0^2}\right]+\exp\left[\frac{\left(\sqrt{L_j}\omega_p+\omega_0\right)^2}{4\Gamma_0^2}\right]\right)\right|^2. \qquad (18)$$

RESULTS AND DISCUSSION

To illustrate the effect of particle shape on the nature of the absorption of pulsed laser radiation, we limit ourselves to considering nanoparticles in the form of ellipsoids of rotation (spheroids). The geometric factors L included in (18) for nanoparticles having the shape of an elongated ($R_\perp < R_\parallel$) or flattened ($R_\perp > R_\parallel$) spheroid ($R_\perp$ and $R_\parallel$ – half-axis along and across the axis of rotation of the spheroid) have the form [13]

$$L_\perp = L_x = L_y = \frac{1}{2}\left(1-L_\parallel\right), \qquad (19)$$

$$L_\parallel = L_z = \begin{cases}\dfrac{1-\varepsilon_p^2}{\varepsilon_p^2}\left[\dfrac{1}{2\varepsilon_p}\ln\left(\dfrac{1+\varepsilon_p}{1-\varepsilon_p}\right)-1\right], & \text{elongated spheroid, } R_\perp < R_\parallel,\\ \dfrac{1+\varepsilon_p^2}{\varepsilon_p^3}\left[\varepsilon_p-\operatorname{arctg}\varepsilon_p\right], & \text{flattened spheroid, } R_\perp > R_\parallel,\end{cases} \qquad (20)$$

where $\varepsilon_p^2=\left|1-R_\perp^2/R_\parallel^2\right|$.

Now in (18) we can proceed to the value of S_e using the $S_e=\left(4\pi/V\left|\vec{E}_0\right|^2\right)W_e$ ratio and obtain an expression for the ratio of the energy absorbed per unit volume of the metal nanoparticle to the energy of the incident laser wave

$$S_e = \frac{4\pi}{V\left|\vec{E}_0\right|^2}W_e = \frac{\pi}{16\left|\vec{E}_0\right|^2}\left(\frac{\omega_p}{\Gamma_0}\right)^2\sum_{j=1}^{3}\left|\vec{E}_0^{\,j}\left(\exp\left[\frac{\left(\sqrt{L_j}\omega_p-\omega_0\right)^2}{4\Gamma_0^2}\right]+\exp\left[\frac{\left(\sqrt{L_j}\omega_p+\omega_0\right)^2}{4\Gamma_0^2}\right]\right)\right|^2. \qquad (21)$$

We confine ourselves to considering metal nanoparticles of spheroidal shape and choose for convenience such a polarization of the electric field of the incident laser wave at which it would be possible to excite simultaneously plasmon oscillations both along and across the axis of rotation of the spheroid. If component $\vec{E}_0^{\parallel}$ is considered to be directed along and $\vec{E}_0^{\perp}$ – is transverse to the axis of rotation of the spheroid, then in the general case such polarization will be

$$E_0^{\parallel}=\left|\vec{E}_0\right|\cos\theta,\quad E_0^{\perp}=\left|\vec{E}_0\right|\sin\theta, \qquad (22)$$

where θ – is the angle between the axis of rotation of the spheroid and the vector of the electric field strength of the incident laser wave $\vec{E}_0$.

Substituting (22) into (21), we obtain:

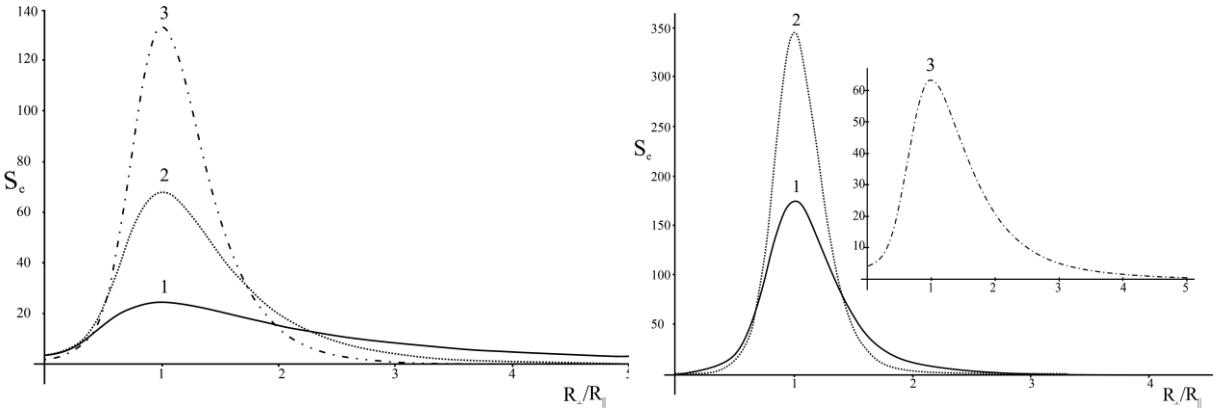
$$S_e = \frac{\pi}{8}\left(\frac{\omega_p}{2\Gamma_0}\right)^2 \left| \begin{array}{l} \cos\theta\left(\exp\left[\frac{\left(\sqrt{L_{\|}}\omega_p-\omega_0\right)^2}{4\Gamma_0^2}\right]+\exp\left[\frac{\left(\sqrt{L_{\|}}\omega_p+\omega_0\right)^2}{4\Gamma_0^2}\right]\right)+ \\ \sin\theta\left(\exp\left[\frac{\left(\sqrt{L_{\perp}}\omega_p-\omega_0\right)^2}{4\Gamma_0^2}\right]+\exp\left[\frac{\left(\sqrt{L_{\perp}}\omega_p+\omega_0\right)^2}{4\Gamma_0^2}\right]\right) \end{array} \right|^2. \quad (23)$$

Now evaluate the parameters included in formula (23). Let's choose $\omega_0 = \Omega = \omega_p/\sqrt{3}, \theta = \frac{\pi}{4}$. Graphical dependences for S_e from $R_\perp/R_\|$ for Na ($\omega_p = 5.65\cdot10^{15}$s^{-1}) and Ag ($\omega_p = 13.8\cdot10^{15}$s^{-1}) nanoparticles at different laser pulse durations are shown in Fig. 1.

From Fig. 1, in particular, it follows that better absorbed laser pulses with a longer duration. The maximum absorption is observed at $R_\perp/R_\| = 1$, *i.e.* for spherical particles.

Now we calculate the value of S_e for the case when the carrier frequency of the laser pulse ω_0 differs from the plasmon resonance frequency in the spherical nanoparticle $\Omega = \omega_p/\sqrt{3}$. We choose $\omega_0 = 3.0\cdot10^{15}$ s^{-1}. Fig. 2 shows the results of numerical calculations of the energy of S_e, absorbed by the metal nanoparticle Na, at different durations of the laser pulse as dependent on the shape of the nanoparticle.

From the analysis of Fig. 2 it follows that as soon as the carrier frequency ω_0 deviates from the frequency of the surface plasmon in the spherical nanoparticle $\Omega = \omega_p/\sqrt{3}$, the peak energy absorption, dependent on the ratio of $R_\perp/R_\|$ is split into two. The first maximum corresponds to the elongated ($R_\perp/R_\| < 1$), and the second – to the flattened ($R_\perp/R_\| > 1$) ellipsoids.



a *b*

Fig. 1. Dependence of S_e energy absorbed by Na (*a*) and Ag (*b*) nanoparticles on the ratio between spheroid half-axes $R_\perp/R_\|$ for different values of laser pulse duration Γ_0: *1* – $1.085\cdot10^{15}$ s^{-1}, *2* – $0.6512\cdot10^{15}$s^{-1}, *3* – $0.465\cdot10^{15}$ s^{-1} (for Ag, high graph)

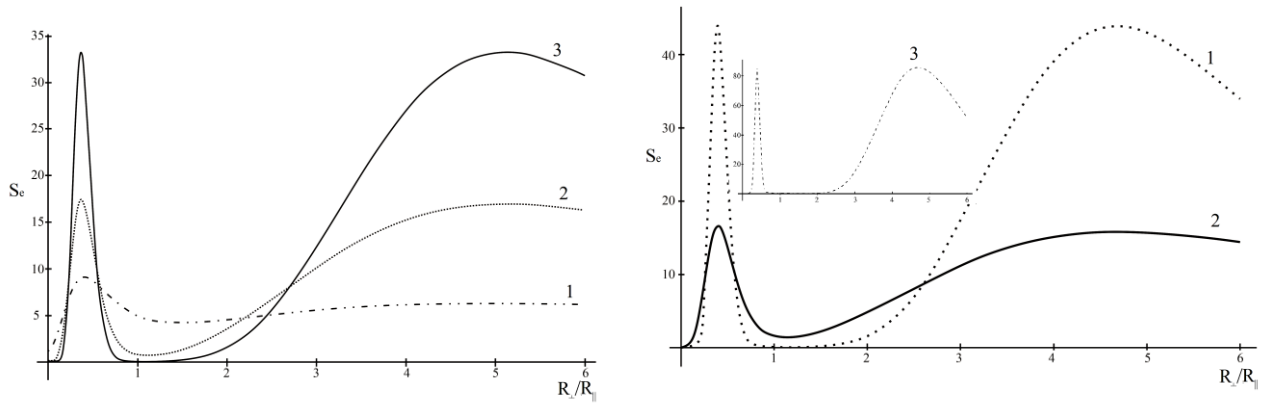


Fig. 2. Dependence of the energy of S_e, absorbed by the nanoparticle Na (*a*) and Ag (*b*), on the frequency $\omega_0 = 3.0 \cdot 10^{15} s^{-1}$, on the ratio between the half-axes $R_\perp / R_\parallel$ of the spheroid for different values of the laser pulse duration Γ_0 *1* – $1.085 \cdot 10^{15}$ s^{-1}, *2* – $0.6512 \cdot 10^{15}$ s^{-1}, *3* – $0.465 \cdot 10^{15}$ s^{-1}

CONCLUSION

The kinetic theory of absorption of ULP by metal nanoparticles of ellipsoidal shape is considered. Simple analytical expressions are obtain that allow one calculating the absorption energy as dependent on the frequency of the carrier laser wave, the pulse duration and the ratio between the axes of ellipsoids (particle shape).

At the carrier wave frequency, which coincides with the surface plasmon frequency, the maximum absorption is observed for spherical nanoparticles. As the carrier frequency deviates from the surface plasmon frequency, two maxima appear in the absorption spectrum, which depend on the ratio of the spheroidal axes: one corresponds to the elongated particles and the other to the flattened ones. As the frequency deviates from the resonant one, the peak of absorbed energy first decreases in absolute value, then splits into two, and finally stabilizes for both elongated and flattened spheroidal nanoparticles.

Кінетична теорія поглинання ультракоротких лазерних імпульсів ансамблями металевих наночастинок в умовах поверхневого плазмонного резонансу

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У цій роботі представлена теорія, яка дозволяє розрахувати енергію, поглинуту сфероїдними металевими наночастинками при опроміненні ультракороткими лазерними імпульсами різної тривалості в області поверхневого плазмонного резонансу. Отримано прості аналітичні вирази, що дозволяють розрахувати енергію поглинання в залежності від частоти несучої лазерної хвилі, тривалості імпульсу та співвідношення між осями еліпсоїдів. Показано, що на частоті несучої (лазерної) хвилі, яка збігається з частотою поверхневого плазмона, максимальне поглинання спостерігається для сферичних наночастинок. При відхиленні несучої частоти від частоти поверхневого плазмона в спектрі поглинання з'являються два максимуми залежно від співвідношення осей сферичної форми: один відповідає видовженим частинкам, а інший – сплощеним.

***Ключові слова**: металеві наночастинки, електрон, плазмон, поверхневий плазмонний резонанс, частота, ультракороткі лазерні імпульси, енергія, поглинання*

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