Absorption of infrared radiation in a small metal particle

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We calculate in the dipole approximation the cross section for the absorption of electromagnetic radiation in a metallic spherical particle. The calculation is made for low frequencies (infrared and lower), when the contribution of the eddy currents to the absorption predominates, and for relatively small particles (~ 10 nm), so that the skin effect can be neglected. No restrictions are imposed, however, on the ratio of the electron mean free path and the specimen size. The calculation shows that in the limit of large free paths the absorption cross section has a nontrivial oscillatory dependence on the radiation frequency. The possibility of observing these oscillations of the spectral dependence of the cross section is discussed.

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1. INTRODUCTION

It is known that the electromagnetic properties of small metallic particles can differ substantially from the properties of bulky metal specimens.[1] The causes of this difference, besides quantum size effects (due, e.g., to the discrete character of the electron energy spectrum[2,3]), may also be effects that can be explained classically. Thus, in particular, if the dimension a of the metal specimen is comparable with the electron mean free path Λ, or is smaller ($a \lesssim \Lambda$), the interaction of the electrons with the specimen boundary begins to influence substantially on the electron response to the external field, and therefore also on the optical properties of the specimen. Therefore at $a \lesssim \Lambda$ the optical characteristics (for example, the absorption cross section) of a metal particle should exhibit a nontrivial dependence on the ratio a/Λ. At room temperatures, in metals with good conductivity (aluminum, copper, silver and others), Λ has characteristic values 10–100 nm. On the other hand, the dimensions of the experimentally investigated particles reach 1 nm (Ref. 1), so that the situation $a \lesssim \Lambda$ seems to be quite attainable in experiment.

From the theoretical point of view, to observe the indicated size effect there is no need to resort to a consistent quantum-mechanical description of the conduction-electron system as a finite Fermi system.[3,4] All we need as a formalism capable of describing the response of the conduction electrons to the external electromagnetic field in a sample of size a at an arbitrary ratio of a and Λ (i.e., with allowance for the interaction of the electrons with the sample boundary). Such a formalism can be the standard kinetic theory of the conduction electrons in a metal.[5]

We note that the equations of macroscopic electrodynamics are applicable only in the limiting case of "massive" specimens, $a \gg \Lambda$. Therefore the known Mie theory[6] that describes the interaction of an electromagnetic wave with a metal sphere within the framework of macroscopic electrodynamics is not suitable for the description of the aforementioned size effect. It was therefore proposed in a number of papers[7,8] to use a certain prescription for extrapolating the classical results of the Mie theory to the case $a \lesssim \Lambda$ by introducing an explicit dependence of the dielectric constant ε on the quantity a/Λ, making it possible to take rough account of the influence of the specimen boundaries on the relaxation properties of the electrons [see Eq. (37) below]. In a number of cases it is possible to estimate correctly in this manner the influence of the size effect even for very small particles.[8] However, the use of such procedures is not fully corroborated and cannot take the place of a consistent kinetic calculation of the response of electrons on an external field in a specimen with finite dimensions $a \lesssim \Lambda$.

In this paper we calculate by the kinetic method the distribution function that describes the (linear) response of conduction electrons in a metal sphere of radius a to an alternating magnetic field $\mathbf{H} = \mathbf{H}_0 e^{-i\omega t}$ of a plane electromagnetic wave at an arbitrary value $a/\Lambda \lesssim 1$. The inhomogeneity of the external field $\mathbf{H}_0$ and the skin effect are not taken into account. It can be shown (see Sec. 2 below) that if the sphere is not too small, $a \gg 2$ nm, and the frequency ω lies in the infrared band (or lower), it is precisely the response to the magnetic field of the wave which makes the main contribution to the dissipation of the electromagnetic energy in the particle. From the obtained distribution function we succeeded in calculating the dependence of the cross section on the radius and on the frequency in closed analytic form. This dependence goes over, in the continuous-medium limit $a \gg \Lambda$, into the corresponding classical relation (Ref. 9, §73). In the opposite limiting case $a \ll \Lambda$ it agrees at low frequencies (in the far infrared) with the one obtained earlier (in the low frequency approximation),[10] while at high frequencies (the near infrared) it exhibits a nontrivial behavior (see Fig. 1) that can be observed experimentally in principle.

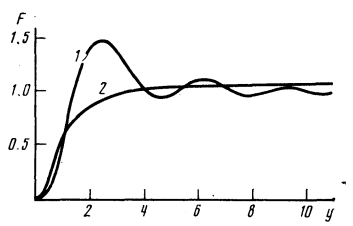

FIG. 1. Dependence of the dimensionless cross section F on the dimensionless frequency $y = a\omega/v_F$ in the absence of volume collisions ($x = a/\Lambda = 0$): 1) rigorous kinetic calculation, Eq. (29); 2) MD theory, Eq. (30).

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2. PRINCIPAL PHYSICAL ASSUMPTIONS

We consider a spherical particle of nonmagnetic metal having a radius a in the field of a plane electromagnetic wave of frequency ω. The range of admissible frequencies is determined by the condition that the contribution of the plasma resonance to the wave-energy dissipation in the medium is small:

$$\omega^2 \ll \omega_p^2 = 4\pi e^2 n/m, \tag{1}$$

where e and m are the charge and mass of the electron, n is the density of the conduction electrons, and ω_p is the plasma frequency (its characteristic value in metals is 10^{16} cm^{-1}, Ref. 11). This means in practice that ω is bounded from above by the near-infrared frequencies ($\omega \lesssim 2 \cdot 10^{15}$ sec^{-1}).

We assume that the radius a is smaller than the skin-layer depth δ (Ref. 9):

$$a < \delta, \quad \delta = \delta(\omega) = c(\omega \operatorname{Im} \sqrt{\varepsilon})^{-1}, \tag{2}$$

so that the skin effect can be neglected (c is the speed of light), and

$$\varepsilon = \varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega)$$

is the complex dielectric constant of the metal at the frequency ω). For metals in the infrared, the dependence of ε on ω is described with good accuracy by the Drude formulas (Ref. 11, pp. 321 and 324):

$$\varepsilon(\omega) = 1 + i4\pi\Sigma(\omega)/\omega, \quad \Sigma(\omega) = \Sigma_0/(1 - i\omega\tau), \tag{3}$$

where the static conductivity Σ_0 and the electron relaxation time τ are connected with the plasma frequency ω_p by the approximate relation (Ref. 11, p. 322)

$$4\pi\Sigma_0 = \omega_p^2 \tau = 4\pi e^2 n\tau/\text{cm}. \tag{4}$$

Eqs. (2)–(4) provide a simple estimate for the depth of the skin layer:

$$\delta(\omega) \approx \delta(\infty)(2/\omega\tau)^{1/2}, \quad \omega\tau \ll 1, \tag{5}$$

$$\delta(\omega) \approx \delta(\infty) = c/\omega_p, \quad \omega\tau \gg 1. \tag{6}$$

The characteristic times τ range from 10^{-14} sec at room temperature to 10^{-10} sec at low temperatures in pure samples, so that both possibilities (5) and (6) are realized in the band (1), with

$$\delta(\infty) = c/\omega_p \sim 3 \cdot 10^{-6} \text{ cm}. \tag{7}$$

It is clear therefore that under conditions (1) and (2) the radius a is certainly small compared with the wavelength λ, regardless of the frequency,

$$a \ll \lambda = 2\pi c/\omega, \tag{8}$$

so that the field of the incident wave can be regarded as homogeneous (the dipole approximation).

Under the conditions (2) and (8) the macroscopic electrodynamics (see Ref. 9, §73) leads to the following equation for the absorption cross section σ_{cl}:

$$\sigma_{cl} = \frac{12\pi a^3 \omega \varepsilon_2}{c}\left(\frac{1}{|\varepsilon|^2} + \frac{\omega^2 a^2}{90c^2}\right), \tag{9}$$

in which the two members are the first two terms [which are the principal ones under condition (8)] of the infinite expansion in multipoles in the Mie theory.[6] The first term (dipole electric $\sim |\varepsilon|^{-2}$) describes the absorption due to the currents of the electric (dipole) polarization of the particle in the external electric field of the wave. The second term (dipole magnetic) describes the absorption due to the eddy currents induced in the particle by the external magnetic field of the wave. Estimates based on Eqs. (3) and (4) show that in the considered frequency range (1), owing to the screening of the electric field in the particle ($|\varepsilon| \approx 4\pi\Sigma_0/\omega \gg 1$) in the region of applicability of Eq. (9) (i.e., at $a > \Lambda$) the dipole magnetic term is larger by several order than the dipole electric, therefore

$$\sigma_{cl} \approx \frac{2\pi a^5 \omega^2 \varepsilon_2}{15c^3} = \frac{8\pi^2 \Sigma_0 \omega^2 a^5}{15c^3(1+\omega^2\tau^2)}. \tag{10}$$

It is obvious (from continuity considerations) that by decreasing the radius a (or by increasing Λ) we should land in a (more or less narrower) region of values $a \lesssim \Lambda$ such that the macroscopic equations (9) and (10) no longer hold, but in this case the contributions of the electric-polarization currents to the absorption will remain negligibly small compared with the contribution of the eddy currents. We assume hereafter realization of just this situation, and disregard the action of the external electric field of the wave.

All the remaining physical assumptions of this paper are universal. We regard the conduction as an (almost) ideal degenerate Fermi gas and describe its response to the alternating external (magnetic) field by the Boltzmann equation in the relaxation-time and in the linear (in the external field) approximation.[5,11] It is assumed in the boundary conditions that the reflection of the electrons from the inner surface of the sphere is diffuse.[5]

3. MATHEMATICAL MODEL AND CALCULATION

Taking the foregoing assumptions into account, the absorption of the electromagnetic-wave energy in the particle is described in the following manner. The homogeneous magnetic field, $H = H_0 e^{-i\omega t}$ of the wave, which is periodic in time, induces an electric eddy field which, by virtue of the symmetry of the problem is determined from Maxwell's induction equation

$$\operatorname{rot} \mathbf{E} = -(1/c)\,\partial\mathbf{H}/\partial t \tag{11}$$

in the form

$$\mathbf{E} = \frac{1}{2c}\left[\mathbf{r} \times \frac{\partial \mathbf{H}}{\partial t}\right] = \frac{\omega}{2ic}[\mathbf{r} \times \mathbf{H}_0]e^{-i\omega t}, \tag{12}$$

where r is the radius vector (the origin 0 of the coordinates is at the center of the particle). The electric field acts on the conduction electrons in the metallic particles and produces a deviation f_1 of their distribution function f from the equilibrium Fermi function f_0:

$$f(\mathbf{r}, \mathbf{v}) = f_0(\mathscr{E}) + f_1(\mathbf{r}, \mathbf{v}), \quad \mathscr{E} = mv^2/2. \tag{13}$$

This leads ultimately to the onset of an (eddy) current

$$\mathbf{j} = e\int \mathbf{v} f \frac{2d^3(mv)}{h^3} = 2\left(\frac{m}{h}\right)^3 \int \mathbf{v} f_1 d^3 v, \tag{14}$$

and also to an energy dissipation (per unit time) $\bar{Q}$ in the volume of the particle[9]:

$$\bar{Q} = \int \overline{(\operatorname{Re}\mathbf{E}) \cdot (\operatorname{Re}\mathbf{j})}\, d^3 r = {}^1/_2 \operatorname{Re} \int \mathbf{j}\mathbf{E}^* d^3 r, \tag{15}$$

where the bar denotes time averaging and the asterisk

the complex conjugate, $\mathbf{v}$ and $\mathscr{E} = mv^2/2$ are the velocity and kinetic energy of the electron, and h is Planck's constant. In (14) we have assumed the standard normalization of the distribution function f, wherein the density of the electronic states equals $2/h^3$, and we use hereafter for the equilibrium function $f_0(\mathscr{E})$ the step-function approximation

$$f_0(\mathscr{E}) = \Theta(\mathscr{E}_F - \mathscr{E}) = \begin{cases} 1, & 0 \leqslant \mathscr{E} \leqslant \mathscr{E}_F, \\ 0, & \mathscr{E}_F < \mathscr{E}, \end{cases} \tag{16}$$

where $\mathscr{E}_F = mv_F^2/2$ is the Fermi energy.

The problem is thus reduced to finding the deviation f_1 of the distribution function from the equilibrium value f_0 produced by the vortical field (12). In the approximation linear in the external field, the function f_1 satisfies the kinetic equation[5,11]

$$-i\omega f_1 + \mathbf{v}\frac{\partial f_1}{\partial \mathbf{r}} + e\mathbf{v}\mathbf{E}\frac{\partial f_0}{\partial \mathscr{E}} = -\frac{f_1}{\tau}, \tag{17}$$

where the proposed stationary dependence on the time is $f_1 \sim e^{-i\omega t}$, and the collision integral is calculated in the relaxation-time approximation:

$$(df_1/dt)_{\text{coll}} = -f_1/\tau.$$

To determine uniquely the function f_1 we must specify for it a boundary condition on the spherical surface of the boundary. We choose this condition to be the diffuse-reflection condition[5]:

$$f_1(\mathbf{r}, \mathbf{v}) = 0 \quad \text{at} \quad \begin{cases} |r| = a, \\ \mathbf{rv} < 0. \end{cases} \tag{18}$$

Solving (17) by the method of characteristics,[12] we obtain

$$f_1 = A(e^{-\nu t'} - 1)/\nu, \quad t' \geqslant 0, \tag{19}$$

where

$$\nu = 1/\tau - i\omega, \quad A = e\mathbf{v}\mathbf{E}\,\partial f_0/\partial \mathscr{E} = \frac{e\omega}{2ic}\left(\frac{\partial f_0}{\partial \mathscr{E}}\right)[\mathbf{v}\times\mathbf{r}]\mathbf{H}_0 e^{-i\omega t}, \tag{20}$$

and $\mathbf{v}$ and A are constant along the trajectory (characteristic), while the parameter t' has the meaning of the time of electron motion along the trajectory from the boundary to the point $\mathbf{r}$ with velocity $\mathbf{v}$, and is defined as a function of $\mathbf{r}$ and $\mathbf{v}$ by the equation

$$t' = \{\mathbf{rv} + [(\mathbf{rv})^2 + (a^2 - r^2)v^2]^{1/2}\}/v^2. \tag{21}$$

Relations (19), (20), and (21) determine completely the solution f_1 of Eq. (17) with boundary condition (18), enabling us to calculate the current (14) and the average dissipated power (15). In the calculation of the integrals (14) and (15) it is convenient to change to spherical coordinates both in coordinate space (r, θ, and φ, with the polar axis $z \parallel \mathbf{H}_0$), and in velocity space (v, α, β, with polar axis v_r). The field (12) has in spherical coordinates only a φ component

$$\mathbf{E} = E_\varphi \mathbf{e}_\varphi, \quad E_\varphi = \frac{i\omega}{2c} r H_0 \sin\theta e^{-i\omega t}. \tag{22}$$

Accordingly the current (14) has also only a φ component (the current lines are closed circles with centers on the z axis on a plane perpendicular to the z axis):

$$j_\varphi = \frac{2m^3e^2E_\varphi}{h^3\nu}\int d^3v\,\delta(\mathscr{E} - \mathscr{E}_F)(1 - e^{-\nu t'})v_\varphi^2$$
$$= E_\varphi\left(\frac{ne^2a}{mv_F}\right)\cdot\frac{3}{4}\int_0^\pi d\alpha\,\frac{1 - e^{-z\eta}}{z}\sin^3\alpha; \tag{23}$$

$$n = 2(m/h)^3\int f_0\,d^3v = 2(m/h)^3 4\pi v_F^3/3,$$

$$z = \nu a/v_F = a/v_F\tau - ia\omega/v_F = x - iy, \tag{24}$$

$$v_F t'/a = \eta = \xi\cos\alpha + [1 - \xi^2\sin^2\alpha]^{1/2}, \quad \xi = r/a. \tag{25}$$

From (15) we obtain now the average dissipated power $\overline{Q}$ and, dividing by the average flux density $cH_0^2/8\pi$ in the wave, we obtain the absorption cross section σ:

$$\sigma = {}^1/_2(8\pi/cH_0^2)\int \mathrm{Re}(j_\varphi E_\varphi^*)\,d^3r = \sigma_0 F(x, y),$$

$$\sigma_0 = \pi^2 ne^2 a^4 v_F/2mc^3, \tag{26}$$

$$F(x, y) = 4y^2\,\mathrm{Re}\int_0^1 \xi^4\,d\xi\int_0^\pi d\alpha\,\frac{1 - e^{-z\eta}}{z}\sin^3\alpha.$$

The integral in (26) can be calculated explicitly. To this end we make the change $\alpha \to \eta$ in the integration variable in accord with (25):

$$\int_0^\pi d\alpha\,\frac{1 - e^{-z\eta}}{z}\sin^3\alpha = \int_{1-\xi}^{1+\xi} d\eta\,\frac{1 - e^{-z\eta}}{z}\left[1 - \frac{(\eta^2 + \xi^2 - 1)^2}{4\xi^2\eta^2}\right]\frac{(\eta^2 - \xi^2 + 1)}{2\xi\eta^2}.$$

We next change the order of integration:

$$\int_0^1 d\xi\int_{1-\xi}^{1+\xi} d\eta(\ldots) = \int_0^2 d\eta\int_{|\eta-1|}^1 d\xi(\ldots),$$

so that the integral (26) takes the form

$$F(x, y) = \frac{y^2}{2}\,\mathrm{Re}\left\{\int_0^2 \frac{d\eta}{\eta^4}\,\frac{1 - e^{-z\eta}}{z}\int_{|\eta-1|}^1 \xi\,d\xi[4\xi^2\eta^2 - (\eta^2 + \xi^2 - 1)^2](\eta^2 - \xi^2 + 1)\right\}.$$

The calculation of the inner integral, while cumbersome, entails no fundamental difficulties and yields

$$\int_{|\eta-1|}^1 \xi\,d\xi[4\xi^2\eta^2 - (\xi^2 + \eta^2 - 1)^2](\eta^2 - \xi^2 + 1) = \frac{\eta^4}{4}(4 - \eta^2)^2,$$

after which the outer integral is easily calculated, and we ultimately obtain the function F in the form

$$F(x, y) = 4y^2\,\mathrm{Re}\left\{\frac{8}{15p} - \frac{1}{p^2} + \frac{4}{p^4} - \frac{24}{p^6} + 8\left(\frac{1}{p^4} + \frac{3}{p^5} + \frac{3}{p^6}\right)e^{-p}\right\}, \tag{27}$$

where $p = 2z = 2(x - iy)$. (It is not advantageous to separate the real part in explicit form, since it is more convenient to carry out the numerical calculations in complex form.)

4. DISCUSSION OF RESULTS

The first term in the curly brackets corresponds to the classical result (10):

$$\sigma_{\text{cl}} = \sigma_0 F_{\text{cl}}(x, y), \quad F_{\text{cl}}(x, y) = 4y^2\,\mathrm{Re}\left(\frac{8}{15p}\right) = \frac{16xy^2}{15(x^2 + y^2)}. \tag{28}$$

The sum of all the remaining terms describes the con-

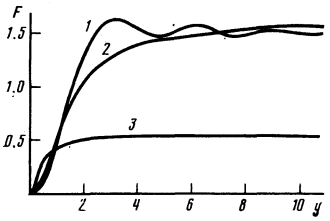

FIG. 2. Dimensionless cross section F vs. the dimensionless frequency $y = a\omega/v_F$ at $x = a/\Lambda = 0.5$: 1) rigorous kinetic calculation, Eq. (27); 2) MD theory, Eq. (30); 3) classical theory, Eq. (28).

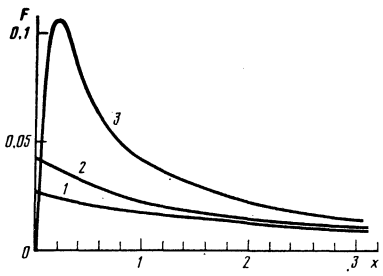

FIG. 3. Dependence of the dimensionless cross section on the dimensionless mean free path $x=a/\Lambda$ in the low frequency region ($y=a\omega/v_F=0.2$) calculated from various equations: 1) rigorous kinetic calculation, Eq. (27); 2) MD theory, Eq. (30); 3) classical theory, Eq. (28).

tribution of the kinetic effects due to the diffuse reflection of the electrons from the spherical surface of the specimen. In the limit $x = a/\Lambda \gg 1$ this contribution of the surface collisions decreases like x^{-2} and turns out to be small compared with the contribution (28) of the volume collisions, which decreases like x^{-1}. Thus, in the case of small mean free paths $\Lambda \ll a$ our result goes over into the classical result (28). However, even at $\Lambda \sim a$ ($x \sim 1$) and more so in the opposite limiting case of large mean free paths $\Lambda \gg a$ ($x \ll 1$) the contribution of the surface collisions to the absorption cross sections becomes quite appreciable and even predominant (see Figs. 1–3), since $F_{cl}(0,y) = 0$, whereas the exact kinetic relation (27) has a finite limit as $x \to 0$:

$$F(0,y)=1+\frac{1}{y^2}+\frac{3}{2y^4}+\left(\frac{2}{y^2}-\frac{3}{2y^4}\right)\cos 2y-\frac{3\sin 2y}{y^3}. \quad (29)$$

There is a known method of extrapolating the classical formulas (9) and (10) to the case when the dimension a of the specimen is comparable with or smaller than the mean free path Λ. In this method the frequency of the volume collisions is replaced by the combined frequency of the volume and surface collisions[7,8]:

$$1/\tau \to 1/\tau + v_F/a.$$

Introduction of this substitution into the Drude formulas (3) and (4) and the use of the so modified expression for $\varepsilon(\omega)$ in the classical formulas (9) and (10) leads to the so called "modified Drude theory" (MD).[13] Transition to the MD theory is equivalent to the substitution $x \to x + 1$ ($\Lambda^{-1} \to \Lambda^{-1} + a^{-1}$) in (28):

$$\sigma_{MD}=\sigma_0 F_{MD}(x,y), \quad F_{MD}(x,y)=\frac{16}{15}\frac{(x+1)y^2}{(x+1)^2+y^2}. \quad (30)$$

The values of F_{MD} are of the same order as F (see Figs. 1–3), thereby confirming the validity of the MD approximation [at least under the condition (18) of purely diffuse reflection from the boundary]. We note that in the limit $x \ll 1$ at low frequencies ($y \ll 1$) we have $F = (5/8)F_{MD}$, which coincides with the result of Ref. 10. However, the exact relation (27) has a distinguishing feature missing in principle from either the classical (28) or from the MD theory (30). At $x \ll 1$ ($a \ll \Lambda$, low temperatures, pure specimens) the frequency dependence of the cross section at high frequencies ($\omega > v_F/a$, $y > 1$) takes the form of damped oscillations (see Figs. 1 and 2). For particles of radius $a \sim 10$ nm these oscillations lie in the near infrared ($1 < \lambda < 10$ μm). If the dipole magnetic contribution to the absorption remains dominant in the near *IR* band (the frequency of the plasma resonance is far enough), these oscillations can be observed in experiment. The experimental data of Ref. 14 (see Fig. 4) seem to point to the presence of such oscillations in precisely the near infrared, but a reliable interpretation of these data is made difficult by their low accuracy and by the polydisperse character of the specimens.

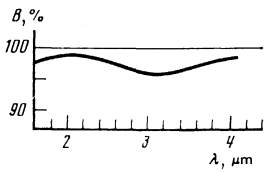

FIG. 4. Transparency B (%) of specimen of ultradisperse gold particles ($a=17$ Å) vs. the wavelength (μm) in the near infrared (from the data of Ref. 14).

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