Effect of electron-electron interaction on infrared absorption by small metallic particles

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It is shown that allowance for electron repulsion at low temperatures, which leads to neutrality of small metallic particles, decreases by unity the exponent a in the power-law dependence of the coefficient of absorption of infrared radiation K on frequency ($K \propto \omega^a$), as compared with existing theories.

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It was shown in Ref. 1 that allowance for electron-electron interaction increases infrared absorption in disordered systems and decreases its frequency dependence. In this paper, we want to point out that an analogous effect also occurs for small metallic particles with random positioning of levels. Following the papers of Kubo[2] and Gor'kov and Éliashberg,[3] infrared absorption by small particles was discussed in many papers (see the review in Ref. 4). In so doing, the main attention was focused on two problems: the statistics of levels in the particle, i.e., the form of the distribution function $\Phi(\hbar\omega)$ of the transition energy, and the difference between the external and effective fields. In this paper, we shall not consider these problems, but we shall examine

only the fact that all particles are neutral due to electron repulsion at low temperatures. This circumstance was first pointed out by Kubo,[2] but we believe that it has never been adequately included in calculations of the coefficient of absorption.

A system of small of particles can be characterized by two basic energy parameters: the average energy necessary to place an extra electron on the particle $e^2/\kappa a$ and the average distance between levels of neutral particles $\Delta = (ga^3)^{-1}$ (the average energy of excitation of the particle). Here g is the density of states of the metal at the Fermi level, a is the size of the particle, and κ is the dielectric constant of the surrounding medium. In practice, for any particle, $\Delta \ll e^2/\kappa a$. We shall first examine the range of frequencies and temperatures satisfying the inequality

$$kT \ll \hbar\omega \ll \Delta \ll \frac{e^2}{\kappa a}. \tag{1}$$

Under the conditions (1), all particles are neutral and absorption occurs by those particles whose first excited level is randomly displaced from the ground state by an amount $\hbar\omega$. The cross section for photoabsorption by the particle is usually written in the form

$$\sigma = \frac{4\pi^2 e^2}{3c} \sum_{\lambda,\lambda'} (\epsilon_{\lambda'} - \epsilon_\lambda) |R_{\lambda,\lambda'}|^2 (n_\lambda - n_{\lambda'}) \delta(\hbar\omega - \epsilon_{\lambda'} + \epsilon_\lambda), \tag{2}$$

where λ, λ' are the single-electron levels of the particle, n_λ and ϵ_λ are the population and energy of the level λ, $|R_{\lambda,\lambda'}|^2$ is the squared modulus of the interaction matrix element between the external field E and the dipole moment of the particle scaled to $(eE)^2$. (If we ignore the difference between the effective field and the external field, then R is the coordinate of the electron undergoing the transition between states λ and λ'.) In the range of frequencies being examined, the quantity $|R_{\lambda,\lambda'}|^2$ does not depend on ω.[3,5]

For averaging over the particles, the sum over λ,λ' in (2) is usually replaced by

$$\Delta^{-2} \iint d\epsilon_\lambda \, d\epsilon_{\lambda'} \, \Phi(|\epsilon_\lambda - \epsilon_{\lambda'}|),$$

where the function Φ includes the repulsion of levels (it is equal to unity if the energies of the levels are not correlated). Further, it is assumed that n_λ is a function of ϵ_λ, which decreases with increasing ϵ_λ from unity to zero, and the difference $n_\lambda - n_{\lambda'}$ is written in the form

$$n_\lambda - n_{\lambda'} = \frac{dn_\lambda}{d\epsilon_\lambda} (\epsilon_\lambda - \epsilon_{\lambda'}). \tag{3}$$

As a result, the following expression is obtained for $\langle\sigma\rangle$ (Ref. 5):

$$\langle\sigma\rangle = \sigma_0 (\hbar\omega/\Delta)^2 \Phi(\hbar\omega), \tag{4}$$

where

$$\sigma_0 = \frac{4\pi^2 e^2}{3c\hbar} \langle |R_{\lambda,\lambda'}|^2 \rangle. \tag{5}$$

The coefficient of absorption is $K = n\langle\sigma\rangle$, where n is the particle concentration. We will be interested only in the frequency dependence of K. It follows from (4) that, for example, for an orthogonal ensemble[3] for which the function $\Phi \propto \hbar\omega/\Delta$, $\hbar\omega \ll \Delta$, at the absorption coefficient $K \propto \omega^3$.

In our opinion, under the conditions (1), the reasoning leading from (2) to (4) is incorrect. We shall show below that one factor $\hbar\omega/\Delta$ in (4) must be dropped so that, for example, for an orthogonal ensemble under conditions (1) $K \propto \omega^2$, rather than ω^3.

Since particles cannot exchange electrons, the population n_λ in reality is not a general function of ϵ_λ for all particles. For example, the energy of the last filled level of a particle fluctuates from particle to particle by an amount much greater than $\hbar\omega$, and in some particles, empty levels lie much lower than filled levels of other particles. A natural requirement for a particle to contribute to absorption at frequency ω is that the difference between the energy of the first empty level and last filled level ϵ (minimum energy of excitation of the particle) would coincide with $\hbar\omega$. In this case, no physical conditions are imposed on the value of the energy of the last filled level. On the other hand, the use of Eq. (3) actually corresponds to the assumption that there exists a well-defined Fermi level, common for all particles, in the sense that the population of levels ϵ_λ is assumed to be a single-valued function of the energy distance from the Fermi level. At the same time, under conditions such that $kT \ll \hbar\omega$, it is found that absorption occurs only due to particles for which the last filled level is located in a band of width $\hbar\omega$ beneath the Fermi level. This gives rise to the second factor $\hbar\omega/\Delta$ in (4), which, in our opinion, should be dropped.

In order to average Eq. (2) over particles under conditions (1), we believe that it is necessary to retain in the sum only one pair of levels λ and λ', corresponding to the lowest excitation energy, and to replace $n_\lambda - n_{\lambda'}$ for it by unity. It is then necessary to average (2) over the magnitude of the lowest excitation energy ϵ. This gives

$$\langle\sigma\rangle = (1/\Delta)\int \sigma\ \Phi(\epsilon)\ d\epsilon = \sigma_0 \frac{\hbar\omega}{\Delta}\Phi(\hbar\omega), \tag{6}$$

i.e., a factor of $\Delta/\hbar\omega$ greater than (4).

So far we have examined a situation with the limiting low temperatures (1), which, however, is realized experimentally without any special difficulties. If $\hbar\omega \ll kT \ll \Delta$, then for absorption, as before, the particles with very low excitation energies ϵ are important. These particles can be viewed as two-level systems, i.e., instead of $n_\lambda - n_{\lambda'}$, we can write $[\exp(-\epsilon/kT) + 1]^{-1}$. Then, for $kT \gg \hbar\omega$, we obtain

$$\langle\sigma\rangle = \sigma_0 \frac{(\hbar\omega)^2}{kT\,\Delta}\Phi(\hbar\omega). \tag{7}$$

If any of the quantities kT or $\hbar\omega$ exceeds Δ, then the characteristics, which are the subject of this work, disappear and $\langle\sigma\rangle$ is given by Eq. (4). Thus, for $\hbar\omega \ll \Delta$, the temperature dependence of $\langle\sigma\rangle$ is very interesting. For $kT \gg \Delta$, the quantity $\langle\sigma\rangle$, which is determined by Eq. (4), increases according to Eq. (7) in the region $\hbar\omega \ll kT \ll \Delta$ and for $kT \ll \hbar\omega$ it saturates at the level (6).

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