

# Polarizability of small metal particles at low temperatures

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The polarizability of small metal particles under various conditions is analyzed. When the discrete spacing of the levels in the small metal particles is not evident because of thermal smearing of the levels, the electric and magnetic polarizabilities can be expressed in terms of the low-frequency conductivity of the particle. The temperature dependence of the conductivity at low temperatures is determined by a quantum correction to the classical value. Two mechanisms determining the quantum correction are considered: Nyquist noise (which introduces a temperature dependence) and an external magnetic field. When the thermal smearing of the levels is less than the spacing of the levels, and the electric field is weak, the polarizability can be expressed in terms of the Green's functions of the system. When these functions are written as integrals over the  $Q$  matrix, the quantum corrections to the polarizability can be calculated.

## 1. INTRODUCTION

The properties of small metal particles are presently the subject of active experimental and theoretical research.<sup>1</sup> The magnetic and electric polarizabilities and the absorption of electromagnetic waves by these particles are being measured experimentally. Several investigators<sup>2–5</sup> are studying the absorption of electromagnetic waves and the polarizability of small metal particles theoretically.

If the average spacing  $\Delta$  between levels is much smaller than  $kT$ , the electric and magnetic polarizabilities  $\alpha_e$  and  $\alpha_m$  and the energy absorption cross section  $\sigma_E$  can be expressed in terms of the frequency  $\omega$  and the static conductivity  $\sigma$  of the material of the particle (if the frequency is not too high).<sup>6</sup>

The dependence of  $\sigma$  on external fields and the temperature in disordered conductors at low temperatures has recently been studied in detail<sup>7</sup> for samples with various effective dimensionalities,  $d_{\text{eff}} = 1, 2, 3$ . It has been found that the magnitude of the quantum corrections and their dependence on the temperature and external fields depend strongly on the dimensionality of the sample.

We are interested in the behavior of the conductivity of small metal particles which are, in a sense, “zero-dimensional” samples. In the first part of this study we derive the quantum correction to the classical (Drude) conductivity for the interference of the electron wave function in elastic scattering by impurities. We assume a uniform distribution of impurities in the sample, and we assume that the mean free path is much smaller than the size of the particle.

The magnitude of this correction is determined<sup>7</sup> by the time  $\tau_\varphi$  for phase relaxation of the electron wave function. This relaxation results from various inelastic processes: electron-electron scattering with a large energy transfer, electron-electron scattering with a small energy transfer (which may be regarded as the interaction of an electron with thermal electromagnetic fluctuations), and electrons-phonon scattering. In addition phase relaxation is caused by a magnetic field.

We consider here the phase relaxation caused by only

two mechanisms: the interaction of the electron with thermal fluctuations (Nyquist noise) and its interaction with an external magnetic field.

In Section 4 we directly calculate the correction to the polarizability of small metal particles with the help of the Kubo formula. To calculate the products of Green's functions which arise here we use the formalism in which these products are written as an integral in Grassmann variables, followed by a transformation to an integral over the  $Q$  matrix.<sup>8</sup>

Small metal particles undergo a self-cleaning,<sup>1</sup> so we are interested in the behavior of the correction to the polarizability in a hollow sphere with diffusely reflecting boundaries. It is found that the polarizability of small metal particles at the frequency  $\omega$  is given by

$$\alpha(\omega) = \alpha^{(0)}(\omega) [1 + f(\omega, \tau_\varphi)]. \quad (1)$$

Here  $\alpha^{(0)}(\omega)$  and  $\tau_\varphi$  are different for particles with and without impurities, while  $f$  is a universal function.

## 2. QUALITATIVE ANALYSIS

The correction to the conductivity for coherent effects can be calculated by examining the quantum-mechanical probability for the return of an electron.<sup>7</sup> If the scale length for inelastic scattering is long (in comparison with the mean free path  $l$  which arises from elastic scattering by impurities), the electron wave function will remain coherent for a long time. If the electron wavelength satisfies  $\lambda \ll l$ , the semiclassical approximation is valid, and the path integral in terms of which the propagation amplitude is expressed<sup>9</sup> can be used to identify the contribution from paths which run within tubes with a cross-section area on the order of  $\lambda^2$  along the classical paths of the diffusing particle. Under these conditions, the relative correction ( $\Delta\sigma/\sigma$ ) to the conductivity is proportional to the probability for a diffusing particle to return to a volume of order  $v_F \lambda^{d-1} dt$  (where  $v_F$  is the electron velocity on the Fermi surface, and  $d$  is the dimensionality of the space). Integrating over all the return times  $t$  from  $\tau$ , where  $\tau$  is the scale time for elastic scattering, to  $\tau_\varphi$ , we find

the estimate<sup>7</sup>

$$\frac{\Delta\sigma}{\sigma} \approx - \int_{\tau}^{\tau_{\varphi}} \frac{dt v_F \lambda^{d-1}}{(\mathcal{D}t)^{d/2}}, \quad (2)$$

where  $\mathcal{D}$  is the diffusion coefficient. In the case  $d \neq 3$  this expression is correct if the transverse dimension of the sample satisfies  $a \lesssim \lambda$ . If  $(\mathcal{D}\tau_{\varphi})^{1/2} \gg a \gg \lambda$ , the classical path uniformly fills the cross-sectional area of the sample over a time  $t \gg t_a \sim a^2/\mathcal{D}$ . The average distance traversed by the particle in the dimension in which it can diffuse freely is, as before,  $(\mathcal{D}t)^{1/2}$ . It can thus be assumed that the effective dimensionality of this sample decreases, and in the case  $d_{\text{eff}} = 2$  the probability which we are seeking is equal to the probability for return to the plane, multiplied by the probability for reaching a given point in the cross section,  $\lambda/a$ . In the same manner, for an arbitrary effective dimensionality we find

$$\frac{\Delta\sigma}{\sigma} \approx - \int_{\tau}^{\tau_{\varphi}} \left( \frac{\lambda}{a} \right)^{3-d_{\text{eff}}} \frac{dt v_F \lambda^{d_{\text{eff}}-1}}{(\mathcal{D}t)^{d_{\text{eff}}/2}}. \quad (3)$$

In the case of a small particle of volume  $V$  we find

$$\Delta\sigma/\sigma \approx -\lambda^2 v_F \tau_{\varphi}/V. \quad (4)$$

We see from this result that the correction depends on the time  $\tau_{\varphi}$ .

Let us find the characteristic time for phase relaxation due to thermal electromagnetic fluctuations. As an electron moves through an electric field, it acquires (or gives up) energy, so that the phase of the wave function changes. The phase difference acquired in motion in opposite directions along the path which begins at the time  $-t$  at the point  $\mathbf{r}$  and which ends up at the same point at the time  $t$  is<sup>7</sup>

$$\begin{aligned} \Delta\varphi(t) &= \frac{1}{\hbar} \int_{-t}^t [\delta\varepsilon_1(t_1) - \delta\varepsilon_2(t_1)] dt_1 \\ &= \frac{e}{\hbar} \int_{-t}^t dt_1 \int_{-t}^{t_1} dt' \mathbf{E}(t') \{ \mathbf{v}(t') + \mathbf{v}(-t') \}, \end{aligned} \quad (5)$$

where  $\mathbf{v}(t)$  is the velocity on the path, and  $\varepsilon$  is the energy of the particle. The square of the phase shift averaged over all such paths can be estimated with the help of this formula:

$$\langle (\Delta\varphi(t))^2 \rangle \sim (e\mathbf{E}_0)^2 \omega^2 t^4 a^2 / \hbar^2 \quad (6)$$

[in the case in which the field  $\mathbf{E}(t) = \mathbf{E} \cos \omega t$  is uniform, and the condition  $\omega t \ll 1$  hold]. If we instead have  $\omega t \gg 1$ , we find

$$\langle (\Delta\varphi(t))^2 \rangle \sim (e\mathbf{E}_0)^2 \mathcal{D}t / \hbar^2 \omega^2. \quad (7)$$

It can be seen from (6) and (7) that the phase shift occurs most effectively over a time  $t$  at frequencies  $\omega \sim 1/t$ . Accordingly, the phase shift over the time  $t$  from the entire spectrum of the electromagnetic fluctuations can be written as an integral over the frequency from 0 to  $1/t$ :

$$\langle (\Delta\varphi(t))^2 \rangle \sim \int_0^{1/t} d\omega \frac{e^2}{\hbar^2} \langle \mathbf{E}\mathbf{E} \rangle_{\omega} \omega^2 t^4 a^2. \quad (8)$$

The spectrum of fields which are uniform over the size of the particle is independent of the frequency:

$$\langle \mathbf{E}\mathbf{E} \rangle_{\omega} \sim T/\sigma V. \quad (9)$$

The phase relaxation time  $\tau_{\varphi}$  is determined by the condition

$$\langle (\Delta\varphi(\tau_{\varphi}))^2 \rangle \sim 1. \quad (10)$$

Using this condition along with (9) and (8), we can estimate the phase relaxation time corresponding to the interaction with Nyquist noise,  $\tau_N$ :

$$\tau_N^{-1} \sim e^2 T a^2 / \sigma V \hbar^2. \quad (11)$$

In a static magnetic field, there is a phase relaxation time  $\tau_H$ . In the case of a small particle, the phase relaxation time in a magnetic field can be found from estimates derived for the phase relaxation in a thin film in a longitudinal magnetic field<sup>7</sup>:

$$\tau_N^{-1} \sim \mathcal{D} (eHa/\hbar c)^2. \quad (12)$$

Since the phase relaxation times appear in the expression for the correlation functions of the propagation amplitudes in the form  $\langle A_1 A_2^* \rangle_t \propto \exp(-t/\tau_{\varphi})$ , the overall phase relaxation time is expressed in terms of all the time in the following way:

$$\tau_{\varphi}^{-1} = \tau_{ee}^{-1} + \tau_N^{-1} + \tau_H^{-1}.$$

Here  $\tau_{ee}$  is the time for pulse relaxation due to electromagnetic interactions with a large energy transfer. The inverse time  $\tau_{ee}^{-1} \sim 1/\nu V$  ( $\nu$  is the state density at the Fermi level) is also independent of the temperature.<sup>7</sup>

As a result we can write

$$\frac{\Delta\sigma}{\sigma} \sim - \frac{v_F \lambda^2}{V} \left[ \tau_{ee}^{-1} + \frac{e^2 T a^2}{\sigma V \hbar^2} + \mathcal{D} \left( \frac{eHa}{\hbar c} \right)^2 \right]^{-1}. \quad (13)$$

### 3. DERIVATION OF EXPRESSIONS FOR THE CORRECTION TO THE CONDUCTIVITY

The theory derived in Refs. 10 and 7 can be used to calculate the corrections to the conductivity without consideration of the electron interaction in the region of parameters specified above ( $T \gg \Delta$ ,  $(\mathcal{D}\tau_{\varphi})^{1/2} \gg a$ ).

The presence of a fluctuating or static electromagnetic field is manifested only in that part of the conductivity which is expressed in terms of a “cooperon,” i.e., a two-particle Green’s function in the particle-particle channel (the Cooper channel). As was shown in Ref. 10, the correction to the conductivity in the Cooper channel is

$$\Delta\sigma = - \frac{2e^2 \mathcal{D}}{\pi} \int_{-\infty}^{+\infty} C_{\eta, -\eta}^t(\mathbf{r}, \mathbf{r}) d\eta, \quad (14)$$

where the cooperon  $C$  satisfies the equation

$$\begin{aligned} \left\{ \frac{\partial}{\partial \eta} + \mathcal{D} \left[ -i\nabla - \frac{e}{c} \mathbf{A} \left( \mathbf{r}, t + \frac{\eta}{2} \right) \right. \right. \\ \left. \left. - \frac{e}{c} \mathbf{A} \left( \mathbf{r}, t - \frac{\eta}{2} \right) \right]^2 + \frac{1}{\tau_{\varphi}^0} \right\} C_{\eta, \eta'}^t(\mathbf{r}, \mathbf{r}') \\ = \delta(\mathbf{r} - \mathbf{r}') \delta(\eta - \eta'). \end{aligned} \quad (15)$$

Here  $\tau_{\varphi}^0$  is a nucleation time for the phase relaxation. This equation is to be solved along with a boundary condition at the surface of the particle,  $(\partial C/\partial \mathbf{n})_s = 0$ , where  $\mathbf{n}$  is the normal to the surface. It can be shown that the boundary condition holds automatically in an integration over paths which

do not go outside the particle.

The solution of Eq. (15) can be written as a path integral.<sup>7</sup> After an average is taken over the fluctuations of the electromagnetic field, this solution becomes

$$C_{\eta, \eta'}(\mathbf{r}, \mathbf{r}') = \int_{\substack{\mathbf{r}(\eta)=\mathbf{r} \\ \mathbf{r}(\eta')=\mathbf{r}'}} D\mathbf{r}(t_1) \exp \left\{ - \int_{\eta'}^{\eta} dt_1 \left[ \frac{\dot{\mathbf{r}}^2(t_1)}{4\mathcal{D}} + \frac{1}{\tau_\varphi^0} \right] + \frac{e^2}{2c^2} \int_{\eta'}^{\eta} dt_2 \dot{r}_\alpha(t_1) \dot{r}_\beta(t_2) \langle A_\alpha^\alpha(\mathbf{r}(t_1), t_1) A_\beta^\beta(\mathbf{r}(t_2), t_2) \rangle \right\}, \quad (16)$$

where

$$\mathbf{A}_t(\mathbf{r}, \eta) = \mathbf{A}(\mathbf{r}, t+\eta/2) + \mathbf{A}(\mathbf{r}, t-\eta/2).$$

Considering only fluctuations which are homogeneous over the dimensions of the particle, we may assume

$$\langle A_\alpha A_\beta \rangle_\omega = \frac{2T}{\sigma V} \frac{c^2}{\omega^2} \delta_{\alpha\beta},$$

and we can write the expression as

$$C_{\eta, \eta'}(\mathbf{r}, \mathbf{r}') = \int_{\substack{\mathbf{r}(\eta)=\mathbf{r} \\ \mathbf{r}(\eta')=\mathbf{r}'}} D\mathbf{r}(t_1) \exp \left\{ - \int_{\eta'}^{\eta} dt_1 \left[ \frac{\dot{\mathbf{r}}^2(t_1)}{4\mathcal{D}} + \frac{1}{\tau_\varphi^0} \right] + \frac{e^2 T}{2\sigma V} \int_{\eta'}^{\eta} dt_2 \dot{r}_i(t_1) \dot{r}_i(t_2) (|t_1+t_2| + |t_1-t_2|) \right\}. \quad (17)$$

After some further manipulations, we find the expression

$$C_{\eta, -\eta}(\mathbf{r}, \mathbf{r}') = \int_{\substack{\mathbf{r}(\eta)=\mathbf{r} \\ \mathbf{r}(-\eta)=\mathbf{r}'}} D\mathbf{r}(t_1) \exp \left\{ - \int_{-\eta}^{\eta} dt_1 \left[ \frac{\dot{\mathbf{r}}^2(t_1)}{4\mathcal{D}} + \frac{1}{\tau_\varphi^0} \right] - \frac{e^2 T}{2\sigma V} (\mathbf{r}' - \mathbf{r})^2 + \frac{e^2 T}{2\sigma V} [\mathbf{r}(t_1) - \mathbf{r}(-t_1)]^2 \right\}. \quad (18)$$

Since the correction to the conductivity is expressed in terms of the cooperon at coincident points, the term  $e^2 T (\mathbf{r}' - \mathbf{r})^2 / 2\sigma V$  is zero. The remaining integral can be rewritten in the following form, by virtue of the small quantity  $e^2 T a^2 / 2\sigma V$ :

$$C_{-\eta, \eta}(\mathbf{r}, \mathbf{r}') = \int_{\substack{\mathbf{r}(\eta)=\mathbf{r} \\ \mathbf{r}(-\eta)=\mathbf{r}'}} D\mathbf{r}(t) \exp \left\{ - \int_{-\eta}^{\eta} dt \left[ \frac{\dot{\mathbf{r}}^2(t)}{4\mathcal{D}} + \frac{1}{\tau_\varphi^0} \right] \right\} \times \left\{ 1 - \int_{-\eta}^{\eta} \frac{e^2 T}{2\sigma V} [\mathbf{r}(t) - \mathbf{r}(-t)]^2 \right\}. \quad (19)$$

After a long time  $\eta$  (long in comparison with the time for diffusion through a sphere,  $a^2/\mathcal{D}$ , the paths which are important in the integral are those with a length considerably greater than the dimensions of the sphere. Accordingly, over a time  $\eta$  the path fills the sample uniformly, and we find

$$C_{-\eta, \eta}(\mathbf{r}, \mathbf{r}') = \frac{1}{V} \exp \left( - \frac{2\eta}{\tau_\varphi^0} - \frac{e^2 T}{\sigma V} \langle \mathbf{r}^2 \rangle 2\eta \right), \quad (20)$$

where

$$\langle \mathbf{r}^2 \rangle = \frac{3}{4\pi R^3} \int d\mathbf{r} = \frac{3}{5} R^2.$$

We then find

$$\int_{-\infty}^{+\infty} C_{-\eta, \eta}(\mathbf{r}, \mathbf{r}') d\eta = \frac{1}{V} \left[ \frac{1}{\tau_\varphi^0} + \frac{3e^2 T R^2}{5\sigma V} \right]^{-1}. \quad (21)$$

The correction to the conductivity then becomes

$$\Delta\sigma = -\frac{2e^2 \mathcal{D}}{\pi} \frac{1}{V} \left[ \frac{1}{\tau_\varphi^0} + \frac{3e^2 T R^2}{5\sigma V} \right]^{-1}. \quad (22)$$

In the case of an external magnetic field, the correction can be found in a similar way. The time for phase relaxation due to the magnetic field is

$$\tau_H^{-1} = \frac{8}{5} \mathcal{D} e^2 H^2 R^2 / \hbar^2 c^2. \quad (23)$$

The combined expression for  $\Delta\sigma$  then becomes

$$\Delta\sigma = -\frac{2e^2 \mathcal{D}}{\pi} \frac{1}{V} \left[ \frac{1}{\nu V} + \frac{8\mathcal{D} e^2 H^2 R^2}{5\hbar^2 c^2} + \frac{3e^2 T R^2}{5\sigma V \hbar^2} \right]^{-1}. \quad (24)$$

#### 4. DERIVATION OF AN EXPRESSION FOR THE POLARIZABILITY

If the thermal smearing of the levels is less than  $\Delta$ , and the strength and frequency of the external field satisfy the conditions

$$E \ll \Delta/e a, \quad \omega \ll v_F/a,$$

the interaction of the small metal particle with the field can be treated by perturbation theory for a quantum-mechanical system<sup>2</sup> with an interaction operator

$$U = -\mathbf{E} \hat{d}, \quad (25)$$

where  $\hat{d}$  is the dipole momentum operator. Using the Kubo formula, we can write an expression for the susceptibility in the form<sup>2</sup>

$$\chi(\omega) = -\frac{e^2}{3} \sum_{kl} \frac{n_k - n_l}{\varepsilon_k - \varepsilon_l + \omega + i\delta} \langle k | \mathbf{r} | l \rangle \langle l | \mathbf{r} | k \rangle. \quad (26)$$

We rewrite this expression by means of retarded and advanced Green's functions:

$$\chi(\omega) = -\frac{e^2}{3} \frac{1}{2\pi i} \int d\mathbf{r} d\mathbf{r}' \int d\varepsilon [n(\varepsilon) - n(\varepsilon - \omega)] G_{\varepsilon+\omega}^R(\mathbf{r}, \mathbf{r}') X [G_{\varepsilon}^R(\mathbf{r}', \mathbf{r}) - G_{\varepsilon}^A(\mathbf{r}', \mathbf{r})] \mathbf{r} \mathbf{r}'. \quad (27)$$

Here

$$G_{\varepsilon}^{R(A)} = \sum_k \frac{\psi_k(\mathbf{r}) \psi_k^*(\mathbf{r}')}{\varepsilon - \varepsilon_k \pm i\delta}, \quad \hat{H} \psi_k(\mathbf{r}) = \varepsilon_k \psi_k(\mathbf{r}), \quad \hat{H} = -\frac{\hbar^2 \Delta}{2m} + U(\mathbf{r}),$$

where  $U(\mathbf{r})$  is the random field of impurities. Here  $\langle U \rangle = 0$ , and

$$\langle U(\mathbf{r}) U(\mathbf{r}') \rangle = (2\pi v \tau)^{-1} \delta(\mathbf{r} - \mathbf{r}').$$

An average must be taken over the random field  $U(\mathbf{r})$ ; then the following characteristic term appears in the expression for the susceptibility:

$$\int [ \langle G_{\varepsilon+\omega}^R(\mathbf{r}, \mathbf{r}') G_{\varepsilon}^R(\mathbf{r}', \mathbf{r}) \rangle - \langle G_{\varepsilon+\omega}^R(\mathbf{r}, \mathbf{r}') G_{\varepsilon}^A(\mathbf{r}', \mathbf{r}) \rangle ] \mathbf{r} \mathbf{r}' d\mathbf{r} d\mathbf{r}'.$$

Expectation values of this type can<sup>8</sup> be rewritten in terms of an integral in Grassmann variables and then trans-

formed to an integral over the  $Q$  matrix. The product  $\langle G_{\epsilon+\omega}^R G_{\epsilon}^A \rangle$  is important here, while the product  $\langle G_{\epsilon+\omega}^R G_{\epsilon}^R \rangle$  can be omitted.

The expression in which we are interested is

$$\begin{aligned} & \int \langle G_{\epsilon+\omega}^R(\mathbf{r}, \mathbf{r}') G_{\epsilon}^A(\mathbf{r}', \mathbf{r}) \rangle \mathbf{r} \mathbf{r}' d\mathbf{r} d\mathbf{r}' \\ &= \int \int DQ d\mathbf{r} d\mathbf{r}' e^{-F} \text{Sp}[(1-\Lambda)(1-\tau_s)Q(\mathbf{r})] \\ & \quad \times (1+\Lambda)(1+\tau_s)Q(\mathbf{r}') \left| \mathbf{r} \mathbf{r}' \left( 16N^2 \int DQ e^{-F} \right)^{-1} \right|_{N=0}, \\ F = & F_0 + \frac{\pi v}{4} \int \left\{ \mathcal{D} \text{Sp}(\nabla Q)^2 + 2i\omega \text{Sp}(\Lambda Q) + \frac{\mathcal{D}}{\tau_\varphi} \text{Sp}[\tau_s, Q]^2 \right\} d\mathbf{r}. \end{aligned} \quad (28)$$

Here  $\mathcal{D} = v_F^2 \tau / 3$ ;  $\Lambda$  is a  $2N \times 2N$  diagonal matrix with the elements  $\lambda_n = 1(n \leq N)$  and  $\lambda_n = -1(n > N)$ ; and  $Q$  is a  $2N \times 2N$  matrix with the elements

$$Q_{nm} = \begin{pmatrix} D_{nm} & \Delta_{nm} \\ -\Delta_{nm} & D_{nm} \end{pmatrix}, \quad D_{nm} = D_{mn}, \quad \Delta_{nm} = -\Delta_{mn}.$$

The last term in the expression for the free energy  $F$  corrects for electron phase relaxation processes;  $\tau_\varphi$  is the phase relaxation time which appears in the equation for the cooperon.

The susceptibility uncorrected for the cooperon was calculated in Ref. 2; it can also be derived in the Gaussian approximation in our formalism. In this case the matrix  $Q$  is written in the form

$$Q = \Lambda e^W, \quad W = \begin{pmatrix} 0 & B \\ -B^+ & 0 \end{pmatrix},$$

where  $B$  is an arbitrary real-quaternion matrix, and the expansion of the free energy is carried out up to second order in  $B$ . The elements  $\Delta_{nm}$  are assumed to be uniform over the sample, while the  $D_{nm}(\mathbf{r})$  are expanded in the eigenfunctions of the Laplacian  $\varphi_k(\mathbf{r})$  with the boundary condition  $\mathbf{n}\nabla\varphi_k = 0$ , where  $\mathbf{n}$  is the normal to the surface (the “confinement” condition).

To calculate the correction containing the cooperon, we must expand  $Q$  up to fourth order in  $B$  and also expand the exponential function in (28). The leading contribution comes from the following term, which appears in the expression for the free energy:

$$\frac{\partial D(\mathbf{r})}{\partial \mathbf{r}} \Delta \frac{\partial D^*(\mathbf{r})}{\partial \mathbf{r}} \Delta^*.$$

This result means that the correction in first order in the cooperon arises from a matrix element, not from the level correlation function. [We recall that  $\chi(\omega)$  can be written in the form<sup>2</sup>

$$\chi(\omega) = \chi_0 - \frac{e^2 \omega^2}{3} v^2 \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} d\epsilon_1 d\epsilon_2 \frac{n_1 - n_2}{\epsilon_1 - \epsilon_2} \frac{(\mathbf{r}_{12} \cdot \mathbf{r}_{21}) R(|\epsilon_1 - \epsilon_2|)}{(\epsilon_1 - \epsilon_2)^2 - (\omega + i\delta)^2}, \quad (29)$$

where  $\chi_0$  is the static susceptibility, and  $R(\omega)$  is the Dyson correlation function of levels, which was reproduced in Ref. 11 by a formalism like that used in Ref. 8;  $Q$  was treated as a matrix which was uniform over the sample.] Integrating the

coefficient of the exponential function in (28) with a Gaussian weight in  $B$ , we find the following expression for the correction to the susceptibility:

$$\begin{aligned} \Delta \chi &= \chi^0(\omega) \frac{1}{3\pi v} \langle \Delta \Delta^* \rangle, \\ \chi^0(\omega) &= \chi_0 + \frac{1}{3} \cdot \frac{48}{175} \frac{e^2}{2\pi v} \frac{R^4}{\mathcal{D}} v^2 \omega^2 \int_{-\infty}^{+\infty} \frac{R(|\epsilon|) d\epsilon}{\epsilon^2 - (\omega + i\delta)^2}, \\ \chi_0 &= 4e^2 R^5 m p_F / 15\pi. \end{aligned} \quad (30)$$

The particle polarizability is expressed in terms of  $\chi$  as follows<sup>12</sup>:  $\alpha = \chi / (1 + \frac{4}{3}\pi\chi)$ . If there are no impurities in the sample, we could carry out an analogous derivation for  $\langle G_{\epsilon+\omega}^R G_{\epsilon}^A \rangle$ , assuming the surface to be rough, and taking an average over the roughness. The expression for the correction should remain of the same form as in (30). For this case,  $\chi^0(\omega)$  was calculated in Ref. 2; it was found to be

$$\chi^0(\omega) = \chi_0 + \frac{1}{3} \cdot \frac{139}{150} \frac{e^2}{2\pi v} \frac{R^3}{v_F} v^2 \omega^2 \int_{-\infty}^{\infty} \frac{R(|\epsilon|) d\epsilon}{\epsilon^2 - (\omega + i\delta)^2}. \quad (31)$$

The phase relaxation time in an empty sphere in a magnetic field is  $\tau_H^{-1} = e^2 v_F H^2 R^3 / 12$  (this value can be derived if we know the temperature of the superconductivity transition in small particles, which was calculated in Ref. 13). The time for the phase relaxation due to electromagnetic fluctuations, on the other hand, remains the same as before. This assertion can be proved in the following way. We write an expression for the cooperon, using Feynman integrals for the propagation functions:

$$C(\mathbf{r}_1, \mathbf{r}_2, -\eta, \eta)$$

$$\begin{aligned} &= \int_{\substack{\mathbf{x}_1(\eta)=\mathbf{r}_2 \\ \mathbf{x}_1(-\eta)=\mathbf{r}_1}} D\mathbf{x}_1(t_1) \exp \left\{ i \int_{-\eta}^{\eta} dt_1 \left[ \frac{m}{2} \dot{\mathbf{x}}_1^2 - e\varphi(\mathbf{x}_1, t_1) \right. \right. \\ &\quad \left. \left. + \frac{e}{c} \mathbf{A}(\mathbf{x}_1, t_1) \dot{\mathbf{x}}_1 \right] \right\} \int_{\substack{\mathbf{x}_2(\eta)=\mathbf{r}_1 \\ \mathbf{x}_2(-\eta)=\mathbf{r}_2}} D\mathbf{x}_2(t_2) \exp \left\{ -i \int_{-\eta}^{\eta} dt_2 \left[ \frac{m}{2} \dot{\mathbf{x}}_2^2 \right. \right. \\ &\quad \left. \left. - e\varphi(\mathbf{x}_2, t_2) + \frac{e}{c} \mathbf{A}(\mathbf{x}_2, t_2) \dot{\mathbf{x}}_2 \right] \right\} \\ &= \int_{\substack{\mathbf{x}_{1,3}(\eta)=\mathbf{r}_2 \\ \mathbf{x}_{1,3}(-\eta)=\mathbf{r}_1}} D\mathbf{x}_1 D\mathbf{x}_2 \exp \left( i \int_{-\eta}^{\eta} L dt \right), \end{aligned} \quad (32)$$

where  $\mathbf{x}_2(-t) = \mathbf{x}_3(t)$ , and

$$\begin{aligned} L &= \frac{m}{2} (\dot{\mathbf{x}}_1^2 - \dot{\mathbf{x}}_3^2) - e[\varphi(\mathbf{x}_1, t) - \varphi(\mathbf{x}_3, -t)] \\ &\quad + \frac{e}{c} [\mathbf{A}(\mathbf{x}_1, t) \dot{\mathbf{x}}_1 + \mathbf{A}(\mathbf{x}_3, -t) \dot{\mathbf{x}}_3]. \end{aligned}$$

We now transform to the new coordinates  $\mathbf{x}_{1,3} = \mathbf{x} \pm \mathbf{y}/2$ . In this case, the paths with small  $\mathbf{y}(t)$  are important in the integral over the paths  $\mathbf{x}(t), \mathbf{y}(t)$ . In the Lagrangian, we carry out an expansion in the small  $\mathbf{y}$ :

$$\begin{aligned}
L = & m \ddot{\mathbf{x}} \cdot \dot{\mathbf{y}} - e[\varphi(\mathbf{x}, t) - \varphi(\mathbf{x}, -t)] - \frac{e}{2} [\nabla \varphi(\mathbf{x}, t) + \nabla \varphi(\mathbf{x}, -t)] \cdot \mathbf{y} \\
& + \frac{e}{c} [\mathbf{A}(\mathbf{x}, t) + \mathbf{A}(\mathbf{x}, -t)] \cdot \mathbf{x} + \frac{e}{2c} [\mathbf{A}(\mathbf{x}, t) - \mathbf{A}(\mathbf{x}, -t)] \cdot \dot{\mathbf{y}} \\
& + \frac{e}{2c} \dot{\mathbf{x}}_\alpha \left[ \frac{\partial A^\alpha(\mathbf{x}, -t)}{\partial x_\beta} - \frac{\partial A^\beta(\mathbf{x}, -t)}{\partial x_\alpha} \right. \\
& \quad \left. + \frac{\partial A^\beta(\mathbf{x}, t)}{\partial x_\alpha} - \frac{\partial A^\alpha(\mathbf{x}, t)}{\partial x_\beta} \right] y^\beta. \tag{33}
\end{aligned}$$

Integrating by parts, we find

$$\begin{aligned}
\exp \left( i \int_{-\eta}^{\eta} L dt \right) = & \exp \left\{ i \int_{-\eta}^{\eta} \left( -m \ddot{\mathbf{x}} - \frac{e}{2} [\nabla \varphi(\mathbf{x}, t) + \nabla \varphi(\mathbf{x}, -t)] \right. \right. \\
& \left. \left. - \frac{e}{2c} [\dot{\mathbf{A}}(\mathbf{x}, t) + \dot{\mathbf{A}}(\mathbf{x}, -t)] - \frac{e}{2c} [\dot{\mathbf{x}}, \mathbf{H}(t) - \mathbf{H}(-t)] \right) y dt \right\} \\
& \times \exp \left\{ i \int_{-\eta}^{\eta} \frac{e}{c} [\mathbf{A}(t) + \mathbf{A}(-t)] \dot{\mathbf{x}}(t) dt \right\}. \tag{34}
\end{aligned}$$

After an integration of  $\mathbf{y}$  we find

$$\begin{aligned}
C_{-\eta, \eta}(\mathbf{r}, \mathbf{r}) = & \int_{\mathbf{x}(-\eta) = \mathbf{r}}^{\mathbf{x}(\eta) = \mathbf{r}} D\mathbf{x}(t) \delta \left( m \ddot{\mathbf{x}} - e \mathbf{E} - \frac{e}{2c} [\dot{\mathbf{x}}, \mathbf{H}(t) - \mathbf{H}(-t)] \right) \\
& \times \exp \left\{ i \int_{-\eta}^{\eta} \frac{e}{c} [\mathbf{A}(t) + \mathbf{A}(-t)] \dot{\mathbf{x}}(t) dt \right\}. \tag{35}
\end{aligned}$$

This expression must be averaged over  $\mathbf{A}(t)$ , and then we must take an integral over the classical paths, which obey the equation specified by the  $\delta$ -function. At large values of  $\eta$ , the only important consideration for the calculation of  $\tau_N^{-1}$  is that after a long time the paths fill the sample uniformly, for both a sphere containing impurities and an empty sphere, and the values of  $\tau_N^{-1}$  are the same in the two cases.

## 5. EXPERIMENTAL OBSERVATION

The contribution from Nyquist noise can apparently be determined most simply in the quantum correction to the polarizability which we have calculated. If  $\tau_H^{-1} \gg \tau_{ee}^{-1} + \tau_N^{-1}$ , i.e., if the magnetic field is strong, we can write

$$\Delta \alpha \sim -(\tau_{ee}^{-1} + \tau_N^{-1}) / \tau_H^{-2}. \tag{36}$$

The term  $\tau_N^{-1} / \tau_H^{-2}$  is proportional to  $T/H^4$  and can easily be distinguished from the other temperature-dependent corrections to the conductivity because of the dependence on the magnetic field.

It should be noted that in an experimental observation of the absorption of electromagnetic waves by a small metal particle one should observe an effect as the temperature is lowered, since the electron temperature exceeds the phonon

temperature.<sup>14</sup> In a metal at a low temperature, the electron-electron collision times are far shorter than that for electron-phonon scattering, so that a quasiequilibrium distribution is established in the electron system with an electron temperature which is generally different from the phonon temperature. When the phonon temperature  $T_{ph}$  is zero, the electron temperature  $T_e$  will be finite at a finite absorption power, and the absorption cross section should correspond to this finite temperature. This limiting temperature can be estimated by using the expression for the characteristic time ( $\tau_e$ ) of the electron energy relaxation due to phonon emission during diffusion in a field of impurities.<sup>15</sup> At the lowest temperatures, this expression is

$$\tau_e = \frac{\pi}{16\alpha\mu} \frac{u^2}{\mathcal{D}} \frac{\Theta^3}{(\epsilon - \mu)^4}. \tag{37}$$

Here  $\alpha$  is the dimensionless constant of the electron-phonon interaction,  $\mu$  is the chemical potential,  $\Theta$  is the Debye temperature, and  $u$  is the sound velocity. Equating the absorbed energy to the energy transferred to phonons by electrons, we find the following expression for the limiting electron temperature:

$$T_e \approx [\Theta^5 R^2 \omega^2 \sigma_0 V |E|^2 / 10\pi^3 \tau \alpha c^2 \mu^2 N]^{1/6}. \tag{38}$$

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