

Rapid Note

Non-local conductivity and the effective potential in small metal particles

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Received: 3 November 1997 / Accepted: 29 January 1998

Abstract. We discuss the absorption of electromagnetic radiation in small conducting particles, in the case where the electron motion is diffusive. We refine an earlier theory in which we calculated the absorption coefficient using an effective potential describing the screened electric field. Our new theory incorporates the effects of non-locality in the electrical conductivity, and shows how the two limiting behaviours of the effective potential at high and low frequencies can be derived from a unified theory. Our results are in full agreement with our earlier calculation.

PACS. 73.23.Ps Other electronic properties of mesoscopic systems – 72.90.+y Other topics electronic transport in condensed matter – 03.65.Sq Semiclassical theories and applications

The purpose of this letter is to present a refinement of an earlier theory [1] concerning the absorption of electromagnetic radiation by small conducting particles: our earlier paper gives a brief review of the topic, and a list of references. Our theory was concerned with the case of very small particles, in which the predominant process is electric dipole absorption, and where the electron motion is diffusive; we consider only frequencies small compared to the plasma frequency.

Classically, the absorption mechanism is Ohmic heating, due to the currents which establish the polarization charges on the surface of the particle. In a quantum mechanical theory, the absorption coefficient is determined by the mean-squared matrix elements of an effective potential $\phi(\mathbf{r})$: the gradient $\nabla\phi$ is the internal electric field, which is much smaller than the externally applied field because of polarization charges on the surface of the particle [2]. The mean squared matrix element $\langle|\phi_{nm}|^2\rangle$ can be estimated semiclassically using the relation

$$\langle|\phi_{nm}|^2\rangle \sim \frac{1}{2\pi nV\hbar} \int_{-\infty}^{\infty} dt \exp(i\omega t) \langle\phi(t)\phi(0)\rangle_E \quad (1)$$

where V is the volume of the particle and n is the density of states per unit volume: the average is over states with $E_n - E_m \sim \hbar\omega$, and with both E_n and E_m close to E

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(this formula is fully explained in [3]). Our earlier paper discussed whether the frequency scale $\omega_c = D/a^2$ (where D is the diffusion constant, and a the typical dimension of the particle) plays a role in the frequency dependence of the absorption coefficient. It might be anticipated that this frequency scale would be important because it characterizes the decay of correlations of the effective potential. We argued that the effective potential is itself dependent upon the frequency, and showed that our form for the effective potential implied that ω_c plays no role in determining the absorption coefficient. A similar conclusion was also reached by Sivan and Imry [4]: their argument is compared with ours in [1].

Our expression for the effective potential was of the form

$$\phi(\mathbf{r}, \omega) = \phi_{\text{stat}}(\mathbf{r}) + i\omega\phi_{\text{dyn}}(\mathbf{r}) \quad (2)$$

where $\phi_{\text{stat}}(\mathbf{r})$ is the Thomas-Fermi potential, which binds the polarization charge which appears in response to a static electric field, and $i\omega\phi_{\text{dyn}}$ is a potential driving the currents which move the polarization charge into place. Our earlier calculation could be criticized on two grounds. Firstly, when calculating the “dynamic” potential $i\omega\phi_{\text{dyn}}(\mathbf{r})$, we used the Drude conductivity $\sigma_0 = ne^2D$, which is only strictly correct at frequencies $\omega \gg \omega_c$: at lower frequencies we should use a non-local conductivity. Secondly, the effective potential (2) is an *ad hoc* combination of its high and low frequency limits. It would be desirable to have a derivation of the effective potential which works uniformly at all frequencies. In this letter we

will present a refined version of the theory, which answers both of these criticisms: it reaches exactly the same conclusion.

Our calculation uses a formula for the non-local conductivity which was originally derived by Serota *et al.* using an argument which invokes diagrammatic theory and supersymmetric methods [5]; a more direct derivation is given in [6]. The current $\mathbf{j}(\mathbf{r}, t)$ is given by

$$\mathbf{j}(\mathbf{r}, t) = \int d\mathbf{r}' \int_{-\infty}^t dt' \tilde{\sigma}(\mathbf{r}, \mathbf{r}'; t - t') \mathbf{E}(\mathbf{r}', t') \quad (3)$$

where the components of the non-local conductivity tensor are

$$\sigma_{ij}(\mathbf{r}, \mathbf{r}'; t) = \sigma_0 [\delta(\mathbf{r} - \mathbf{r}') \delta(t) \delta_{ij} - D \partial_{r_i r_j}^2 P(\mathbf{r}, \mathbf{r}'; t)] . \quad (4)$$

We will briefly discuss the definition of the absorption coefficient, and its relation to the effective potential, before describing how we calculate the effective potential. We define the absorption coefficient $\alpha(\omega)$ as the rate of absorption of energy in a single particle, divided by the square of the amplitude of the externally applied electric field, \mathcal{E}_0 . Following [1], we use the Fermi golden rule and Pauli exclusion principle (equivalently, the Kubo formula) to calculate the rate of absorption: we have

$$\alpha(\omega) \equiv \frac{1}{\mathcal{E}_0^2} \left\langle \frac{dE}{dt} \right\rangle = \frac{\pi \hbar n^2 V^2 \omega^2}{2 \mathcal{E}_0^2} \langle |\phi_{nm}|^2 \rangle_{E_n - E_m \sim \hbar \omega} . \quad (5)$$

Given the effective potential $\phi(\mathbf{r})$, its correlation function may be written

$$\langle \phi(t) \phi(0) \rangle = \frac{1}{V} \int d\mathbf{r} \int d\mathbf{r}' \phi(\mathbf{r}) \phi(\mathbf{r}') P(\mathbf{r}, \mathbf{r}'; t) \quad (6)$$

where the diffusion propagator $P(\mathbf{r}, \mathbf{r}', t)$ satisfies $[\partial_t - D \nabla_{r_i}^2] P(\mathbf{r}, \mathbf{r}'; t) = \delta(\mathbf{r} - \mathbf{r}')$, with the Neumann boundary condition $n_i \partial_{r_i} P = 0$ (where n_i are the components of the normal to the boundary).

We now turn to the calculation of the effective potential. The externally applied electric field induces a polarization charge, with charge density $\rho(\mathbf{r})$. Following the arguments of our earlier paper [1] (see also the discussions in [7,8]), we will assume that, for frequencies small compared to the plasma frequency, the charge density is identical to the classical charge density produced by a static electric field. In three dimensional particles this is concentrated at the surface, in a layer of depth λ_s , the Thomas Fermi screening length, but in two dimensions the screening charge has a continuous distribution, with a $z^{-1/2}$ singularity, where z is the distance from the edge. Provided the frequency of the radiation is small compared to the plasma frequency, the polarization charge density may be assumed to be that given by classical electrostatics, for a constant and uniform externally applied field. For general geometry, the calculation of $\rho(\mathbf{r})$ is a difficult problem in electrostatics, but we will assume that the static polarization charge density is known.

The effective potential determines the current flowing through (4) and (5). We determine $\phi(\mathbf{r})$ by requiring that the current generates the known charge density: combining the continuity relation $i\omega\rho = \nabla \cdot \mathbf{j}$ with (3, 4), we find

$$\rho(\mathbf{r}) = -e^2 \int d\mathbf{r}' \Pi(\mathbf{r}, \mathbf{r}'; \omega) \phi(\mathbf{r}') \quad (7)$$

where

$$\Pi(\mathbf{r}, \mathbf{r}'; \omega) = n \delta(\mathbf{r} - \mathbf{r}') - i\omega n P(\mathbf{r}, \mathbf{r}'; \omega) \quad (8)$$

is a non-local polarizability.

From (7), we can see the limiting behaviours of $\phi(\mathbf{r})$ at high and low frequencies. In the low frequency limit, $\Pi(\mathbf{r}, \mathbf{r}'; \omega) \simeq n \delta(\mathbf{r} - \mathbf{r}')$ and we see that $\phi(\mathbf{r}) = (ne^2)^{-1} \rho(\mathbf{r})$. This is exactly the Thomas Fermi potential $\phi_{\text{stat}}(\mathbf{r})$ which appears in (2). In the high frequency limit, $\Pi(\mathbf{r}, \mathbf{r}'; \omega)$ approaches $-(nD/i\omega) \nabla^2 \delta(\mathbf{r} - \mathbf{r}')$. Hence, in this limit, $\phi(\mathbf{r})$ satisfies Poisson's equation in the form $\nabla^2 \phi = (i\omega/\sigma_0) \rho$: this (together with the boundary condition) defines the dynamical potential $\phi_{\text{dyn}}(\mathbf{r})$ which dominates (2) at high frequencies. Equations (7, 8) therefore provide a unified model to determine the effective potential, which gives the correct limiting behaviour at low and high frequencies.

In fact, we can also show that the effective potential (2) which we proposed in our earlier paper is the exact solution of (7). By partial integration and using the diffusion equation for the propagator, we obtain

$$[D \nabla^2 - i\omega] \rho(\mathbf{r}) = -\sigma_0 \nabla^2 \phi(\mathbf{r}) . \quad (9)$$

Using the ansatz $\phi(\mathbf{r}) = -(ne^2)^{-1} \rho(\mathbf{r}) + i\omega \phi_1(\mathbf{r})$ we obtain $\phi_1 \equiv \phi_{\text{dyn}}$: the frequency-dependent contribution to $\phi(\mathbf{r})$ is therefore precisely the dynamic potential in (2).

We are grateful to the organizers of the program 'Quantum Chaos and Disordered Systems' for inviting us to the Newton Institute. This program was supported by UK Engineering and Physical Sciences Research Council (EPSRC). The visit of BM was partially funded by the Max Planck Institute for Physics of Complex Systems, Dresden. The work of MW was also supported by a research grant from the EPSRC, reference GR/L02302.

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