Polarizability and Absorption of Small Conducting Particles in a Time-Varying Electromagnetic Field

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(First draft 09/15/99)

We study small conducting particles and thin films in an oscillating longitudinal electric field. We find the charge, current, and field distribution in the particle, the polarizability and the electric dipole absorption. We account for Thomas-Fermi screening by adding a Fick’s diffusion term to Ohm’s law. Alternatively, we describe a particle as a dielectric body with a non-local dielectric constant which is derived in a microscopic linear-response theory. We show that both approaches are equivalent.

PACS: 73.23.Ps, 78.90.+t

I. INTRODUCTION

The classical theory of polarizability and absorption of small metallic particles in a time-varying electromagnetic field is a well studied and documented subject [1], [2]. The corresponding quantum theory has also been studied, starting with the paper of Gor’kov and Eliashberg [3] (GE). The quantum treatment can, in principle, address the features of (i) nonlocality of the current-field relationship, (ii) discreteness and statistics of the energy levels and (iii) self-consistent screening of the transition matrix elements. GE assumed that the spectral properties and the matrix elements were statistically independent and calculated the latter ‘semiclassically’ for both pure ($\ell > a$) and ‘dirty’ ($\ell < a$) small particles. They did not take into account the screening of the external field. This was treated later by Lushnikov, Simonov, and Maksimenko [4] along the same lines as GE.

For dirty systems in particular, the problem has been revisited recently by Blanter and Mirlin [5] (BM). They applied the formalism of the supersymmetric sigma model developed by Efetov [6] for describing the level statistics and the diffusive nature of the matrix elements of disordered wavefunctions (see also [7]). Their work confirmed the earlier result of GE for unscreened response, thereby justifying GE ansatz of statistical independence. In a later paper [5] BM incorporated screening (at zero frequency only) through an approximate formulation of the RPA in which the irreducible polarization part was restricted to first order in the diffusion term.

Mehlig and Wilkinson [8] have given a phenomenological analysis of the electric response of small dirty systems. They considered the response to lowest order in frequency postulating in advance features of the in-phase and out-of-phase parts of the current relative to the electric field, namely, that they are ohmic and diffusive, respectively. (We will see later on that this separation of the current by phase is neither necessary nor correct in detail.) In a second paper [8] they restated their earlier results, starting with the nonlocal conductivity $\sigma(r, r'; \omega)$ first derived by Serota et. al. [9]. As in the diagrammatic method of Ref. [5], the self-consistency equations for the charge/current/field distribution inside the particle are cast in integral equation form, which are cumbersome to apply in finite geometries and have not been solved.

In this paper we show that a considerable simplification in the application of the semiclassical theory in the above papers to specific geometries is achieved by modifying the classical Maxwell method for fields in dielectric and conducting bodies to include nonlocality in the electric current-electric field constitutive relation. We add to Ohm’s law in the standard Rayleigh-Drude (RD) theory a Fick’s law diffusion term, obtaining what we refer to as the Einstein transport equation. We then follow Landau and Lifshitz’s observation (see [10]) that an electrically isolated linear-response system can always be treated as a dielectric body – in this case one with a nonlocal dielectric response. However, it will be shown that even this response function can be avoided. Instead, after formulating appropriate boundary conditions, we proceed to solve Maxwell’s differential equations for the charges and fields in almost textbook-like fashion. Thereby self-consistency is achieved at all frequencies without the cumbersome integral

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equations of diagrammatic schemes. The classical treatment of dielectric objects in external fields is, after all, a self-consistent solution method for charges and their fields. The great simplicity of this approach relative to the earlier work mentioned should not distract attention from the fact that it yields without analytic complication the first exact charge/current distributions for slab and spherical geometries and that the method is obviously applicable to more complex geometries.

In Section II we discuss the Maxwell theory with the modified constitutive equation. The method of solution is presented in Section III, where it is illustrated for a slab and sphere. Section IV has the discussion of the equivalence to the earlier nonlocal integral equation first derived by microscopic theory. In Section V we compare the electric and magnetic dipole absorption.

II. THE "DIELECTRIC" EQUATIONS AND BOUNDARY CONDITIONS.

In this Section we present the modified RD macroscopic equations for a dirty metal or semiconductor which includes charge diffusion. The particle is assumed to be in a uniform quasistatic electric field \( \mathbf{E} = E_0 e^{-i\omega t} \) such as that between parallel condenser plates. This approximates the electric response to low frequency light where \( k a \ll 1 \); \( a \) is the characteristic small dimension of the particle. (The magnetic response is discussed in Sec. V.) The Maxwell equation for the longitudinal field and the continuity condition are

\[
4\pi \rho (\mathbf{r}; \omega) = \nabla \cdot \mathbf{E} (\mathbf{r}; \omega) \tag{1}
\]

\[
i\omega \rho (\mathbf{r}; \omega) = \nabla \cdot \mathbf{j} (\mathbf{r}; \omega) \tag{2}
\]

Eliminating \( \rho \), we find

\[
\nabla \cdot \left( \mathbf{j} (\mathbf{r}; \omega) - \frac{i\omega}{4\pi} \mathbf{E} (\mathbf{r}; \omega) \right) = 0
\]

so that the current density can be written as

\[
\mathbf{j} (\mathbf{r}; \omega) = -\frac{i\omega}{4\pi} \left( \mathbf{D}(\mathbf{r}; \omega) - \mathbf{E}(\mathbf{r}; \omega) \right) \tag{3}
\]

in terms of a divergenceless vector \( \mathbf{D} \)

\[
\nabla \cdot \mathbf{D}(\mathbf{r}; \omega) = 0 \tag{4}
\]

For simplicity we assume that there is no background dielectric \( (\epsilon_B = 1) \) so that the polarization current is the mobile-charge current \( \mathbf{j} \). Then \( \mathbf{D} \) has a meaning of electric displacement field for a "dielectric" medium in which all the current is assigned to the polarization \( \mathbf{P} \), namely,

\[
\mathbf{j} (\mathbf{r}; \omega) = -i\omega \mathbf{P}(\mathbf{r}; \omega) \tag{5}
\]

so that

\[
\mathbf{D}(\mathbf{r}; \omega) = \mathbf{E}(\mathbf{r}; \omega) + 4\pi \mathbf{P}(\mathbf{r}; \omega) \tag{6}
\]

and

\[
\nabla \cdot \mathbf{P}(\mathbf{r}; \omega) = -\rho (\mathbf{r}; \omega) \tag{7}
\]

The boundary conditions reflect the facts (i) that there is diffusion so that no infinitely thin surface charge density is present and (ii) that there is no current through the sample boundary

\[
j_n (\mathbf{r}; \omega) |_{\partial} = 0 \tag{8}
\]

Consequently,

\[
D_n (\mathbf{r}; \omega) |_{\partial} = E_n (\mathbf{r}; \omega) |_{\partial} \tag{9}
\]

In the absence of surface charge, the electric field is continuous at the boundary
where $E_{\text{out}}$ is the field outside the particle (the sum of the applied field and the field of the particle). From eqs. (10) and (11) it then follows that the normal component of $D$ is also continuous at the boundary. On the other hand, the tangential component of $D$ may be discontinuous due to tangential currents at the boundary.

A constitutive equation is needed to complete the description of the medium. For this we use the "generalized Einstein transport equation"

$$j (r; \omega) = \sigma_D E (r; \omega) - D_D \nabla \rho (r; \omega)$$

where $\sigma_D$ is the Drude conductivity

$$\sigma_D = \frac{\sigma_0}{1 - i \omega \tau}$$

$\sigma_0$ and $\tau$ are the Boltzmann conductivity and the scattering time respectively and

$$D_D = \frac{D}{1 - i \omega \tau}$$

where $D$ is the static diffusion coefficient. Thus, the Einstein relation is taken to hold at $\omega \neq 0$ [10], namely

$$\frac{\sigma_D}{D_D} = \frac{\sigma_0}{D} = e^2 \frac{dn}{d\mu}$$

where $dn/d\mu$ is the thermodynamic density of states.

### III. SOLUTION FOR THE FIELD DISTRIBUTION

#### A. General Method

Combining eqs. (1), (3), and (11) leads to the following equation for the electric field:

$$\nabla^2 E (r; \omega) - \tilde{\Lambda}^2 E (r; \omega) = \frac{i \omega}{D_D} D (r; \omega)$$

where $\tilde{\Lambda}$ is the dynamical screening length given by

$$\tilde{\Lambda}^2 = -\frac{i \omega \epsilon_D}{D_D} = \Lambda^2 \left(1 - \frac{i \omega}{4\pi \sigma_D}\right)$$

and

$$\epsilon_D = 1 + \frac{4\pi i \sigma_D}{\omega}$$

$$\Lambda^2 = 4\pi e^2 \frac{dn}{d\mu}$$

are the Drude dielectric function for a conductor and the static screening length respectively.

We introduce an auxiliary field $E^h (r; \omega)$ which measures the deviation from the Rayleigh-Drude field relation, namely,

$$E^h (r; \omega) = E (r; \omega) - \frac{D (r; \omega)}{\epsilon_D}$$

It obeys the homogeneous PDE

$$\nabla^2 E^h (r; \omega) - \tilde{\Lambda}^2 E^h (r; \omega) = 0$$

and the boundary condition following from eqs. (9) and (10):
\[ E_h^n|_\partial = \left( \frac{\epsilon_D - 1}{\epsilon_D} \right) D_n|_\partial \]  
\[ E_h^i|_\partial = E_{i}^{\text{out}}|_\partial \]  

Equations (20) and (21) determine the field of the screening charge completely from the boundary values of the displacement field \( \mathbf{D} \).

Potentials can be introduced since the quasistatic fields are longitudinal:

\[ \mathbf{E} = -\nabla \phi, \quad \mathbf{D} = -\nabla \phi^d, \quad \mathbf{E}^h = -\nabla \phi^h \]  

Then from (13)

\[ \phi^h = \phi - \frac{\phi^d}{\epsilon_D} \]  

Given that the constant in \( \phi \) is determined, say, at infinity the undefined constants in \( \phi^d \) and \( \phi^h \) must compensate. We will see below that there is a natural choice for \( \phi^h \). The potentials are determined by solving the Laplace equation for \( \phi^d \) in the interior:

\[ \nabla^2 \phi^d = 0 \]  
\[ \nabla_n \phi^d|_\partial = -E_{n}^{\text{out}}|_\partial \] 

plus the auxiliary PDE for \( \phi^h \):

\[ \nabla^2 \phi^h - \Lambda^2 \phi^h = 0 \]  
\[ \nabla_n \phi^h|_\partial = \left( \frac{\epsilon_D - 1}{\epsilon_D} \right) \nabla_n \phi^d|_\partial \]  
\[ \left( \phi^h + \frac{\phi^d}{\epsilon_D} \right)|_\partial = \phi^{\text{out}}|_\partial \] 

\( \phi^h \) has the special feature of determining the charge density \( \rho \) directly. First, from eqs. (1), (4) and (23) it follows that

\[ \nabla^2 \phi^h = -4\pi \rho \]  

and hence from (26) that

\[ \rho = \frac{-\Lambda^2}{4\pi} \phi^h \]  

Note that the right hand term of eq. (26) is an arbitrary constant, not necessarily zero. The choice of zero, together with the vanishing of the total internal charge, fixes the constant in \( \phi^h \) to be such that the average value of \( \phi^h \) over the sample is zero.

**B. Examples: The field in a sphere and slab**

1. **Sphere**

This case is presented first because it better represents the method of potentials just described. Following Ref. [1] we start with the required form for the external potential of a dipole

\[ \phi^{\text{out}}(r; \omega) = -\mathbf{E}_0 \cdot \mathbf{r} + \frac{\vec{\beta} \cdot \mathbf{r}}{r^3} \]  

and \( \vec{\beta} \) (\( \| \mathbf{E}_0 \)\) is the induced electric dipole moment of the sphere (of radius \( a \)). The interior solution of the Laplace equation (24) for \( \phi^d \) is proportional to \( E_0 \cdot \mathbf{r} \propto \cos \theta \) and, from the boundary condition (23), we get
\[ \phi^d (\mathbf{r}; \omega) = -E_0 \cdot \mathbf{r} \left( 1 + \frac{2a^3}{\alpha^3} \right) \]  

(31)

where \( \alpha = \mathcal{P}/E_0 \) is the (complex) polarizability. Likewise \( \phi^h \propto \cos \theta \). The solution of (26) is

\[ \phi^h (\mathbf{r}; \omega) \propto i_1 (r\Lambda) \cos \theta \]

\[ i_1 (z) = \frac{\cosh z}{z} - \frac{\sinh z}{z^2} \]

the latter being the spherical Bessel function of the imaginary argument. The two boundary conditions (27) determine the coefficient of the solution \( \phi^h \) together with the value of \( \alpha \), namely,

\[ \phi^h (\mathbf{r}; \omega) = -E_0 \cdot \mathbf{r} \cdot \frac{\epsilon_D - 1}{\epsilon_D + 2X} \cdot \frac{3ai_1 (r\Lambda)}{\sinh \left( a\Lambda \right) r} \]

(32)

and

\[ \alpha = a^3 \frac{(\epsilon_D - 1) X}{\epsilon_D + 2X} \]

(33)

with

\[ X = 1 - 3 \frac{\coth \left( a\Lambda \right)}{a\Lambda} + \frac{3}{(a\Lambda)^2} \]

(34)

\( X \) increases monotonically to unity as \( |a\Lambda| \) increases. This is very similar to the classical polarizability of a dielectric sphere with the dielectric constant \( \epsilon_D \) and reduces to it when the dynamical screening length is small relative to the radius, i.e. \( a\Lambda \) is large. The electric dipole absorption is (see Appendix A for a detailed discussion) is given by the imaginary part of the polarizability:

\[ Q = \frac{1}{2} \omega E_0^2 \text{Im} \alpha \]

(35)

The familiar RD absorption, namely,

\[ Q_{RD} = \frac{3\omega^2 a^3}{8\pi \sigma_0} E_0^2 \]

(36)

is obtained at low frequencies \( \omega \ll \sigma_0 \). Corrections in \( (a\Lambda)^{-1} \) are easily obtained from eq. (34) and in the lowest order

\[ Q = Q_{RD} \left( 1 - \frac{11}{2a\Lambda} \right) \]

(37)

2. Slab

Both the slab normal and the field are taken along \( z \): \( L_z \ll L_x, L_y \sim \sqrt{S} \), and \( -L_z/2 < z < L_z/2 \). The one-dimensionality permits working directly with the fields. By eqs. (10), (11) and (10) \( D (z) = \text{const} = E^\text{out} \equiv E_0 \) and

\[ \frac{d^2}{dz^2} E^h (z; \omega) - \Lambda^2 E^h (z; \omega) = 0 \]

(38)

\[ E^h (-L_z/2) = E^h (L_z/2) = \left( \frac{\epsilon_D - 1}{\epsilon_D} \right) E_0 \]

(39)
so

\[ E^h(z;\omega) = E_0 \frac{(\epsilon_D - 1) \cosh \left( \frac{z\Lambda}{2} \right)}{\cosh \frac{Lz\Lambda}{2}} \]  

(40)

And the internal electric field is

\[ E(z;\omega) = \frac{E_0}{\epsilon_D} \left( 1 + (\epsilon_D - 1) \frac{\cosh \left( \frac{z\Lambda}{2} \right)}{\cosh \frac{Lz\Lambda}{2}} \right) \]  

(41)

The polarizability and absorption given, respectively, by

\[ \alpha = \frac{L_x L_y L_z (\epsilon_D - 1)}{4\pi \epsilon_D} \left( 1 - \frac{\tanh \left( \frac{L_z \Lambda}{2} \right)}{L_z \Lambda/2} \right) \]  

(42)

and

\[ Q = Q_{RD} \left( 1 - \frac{3}{L_z \Lambda} \right) \]  

(43)

In summary, incorporating diffusion into the current field relation modifies the classical treatment of dielectric bodies by replacing the surface charge density relation \( \sigma = D_n (\epsilon - 1) / 4\pi \epsilon \) by an additional degree of freedom for the bulk charge density \( \rho(r) \). In both cases the Maxwell equations boundary conditions are all that is needed to assure self-consistency of the charge and field distribution. It is seen that the screening charge decreases exponentially with distance \( d \) from the surface as \( \exp \left( -\frac{\Lambda d}{2} \right) \) resulting in the correction to the RD absorption proportional to the fraction of the total volume occupied by the screening charge. This is a small correction for metals where \( \Lambda^{-1} \) is small but may be significant in semiconductors.

**IV. RELATION TO RESPONSE-FUNCTION FORMALISM**

In this Section we make the connection between the classical dielectric formalism in Sec. II and the quantum mechanical theories mentioned in the Introduction. As stated earlier, the dielectric method incorporates nonlocality in both the conduction and screening consistently.

**A. Response formalism**

We start with the nonlocal response function for the electric current vs. the electric field in the form first given in Ref. [10] which was derived from a field theoretical formalism for diffusive motion, namely,

\[ j_\alpha (r;\omega) = \int \sigma_{\alpha\beta} (r, r';\omega) E_{\beta} (r';\omega) d r' \]  

(44)

where, for both \( r \) and \( r' \) within the sample,

\[ \sigma_{\alpha\beta} (r, r';\omega) = \sigma_D \left( \delta_{\alpha\beta} \delta(r - r') - \nabla_\alpha \nabla'_\beta d(r, r';\omega) \right) \]  

(45)

and \( d \) is the diffusion propagator ("diffuson") satisfying the equation

\[ \nabla^2 d(r, r';\omega) = -\delta(r - r') - \frac{i\omega}{D_D} d(r, r';\omega) \]  

(46)

and the boundary condition

\[ \nabla_n d(r, r';\omega) \vert_\partial = \nabla'_n d(r, r';\omega) \vert_{\partial'} = 0 \]  

(47)
Outside the sample $d$ vanishes.

We note here two features of eqs. (44)-(47): Firstly, the conductivity tensor $\sigma$ with components $\sigma_{\alpha\beta}$ is longitudinal, producing longitudinal current $j$ when $E$ is longitudinal ($= -\nabla \phi$). (Furthermore, $j_n = 0$). Secondly, the coefficient with $D_D$ on the right in eq. (46) is consistent with the appearance of $\sigma_D$ in eq. (45) and both are necessary. With them, eqs. (45) and (46) are the real-space equivalent of the longitudinal conductivity, eq. (3.145) of Ref. [10], in an unbound homogeneous medium, namely,

$$\sigma_L(q, \omega) = \frac{ie^2}{m} \left[ \frac{\omega}{\tau} + \frac{s^2 q^2}{\omega} \right]^{-1}$$

$$\sigma_0 = \frac{ne^2}{m}$$

(48)

where $\sigma_0 = ne^2/\tau m$ and $s$ is the fermi-liquid sound velocity

$$s^2 = \frac{n d\mu}{m \text{dn}} \frac{ne^2 D_0}{\sigma_0}$$

(49)

In $q$-space, eqs. (45), (46) are

$$\sigma(q, \omega) = \sigma_D \left( 1 - q^2 d(q, \omega) \right)$$

$$d(q, \omega) = \left( q^2 - \frac{i\omega}{D_D} \right)^{-1}$$

(50)

and combine to yield eq. (48).

To connect with Sec. II we next show that the constitutive relation (44) is equivalent to the Einstein transport eq. (11). We need some variants of eq. (44). Inserting $E = -\nabla \phi$ in eq. (44), and integrating by parts gives

$$j(r; \omega) = i\omega e^2 \frac{dn}{d\mu} \nabla \int d(r, r'; \omega) \phi(r'; \omega) \, dr'$$

(51)

Combining the continuity equation (2) with eq. (46) gives the nonlocal relation for the charge density

$$\rho(r; \omega) = \int \Pi(r, r'; \omega) \phi(r'; \omega) \, dr'$$

(52)

where

$$\Pi(r, r'; \omega) = e^2 \frac{dn}{d\mu} \left( -\delta(r - r') - \frac{i\omega}{D_D} d(r, r'; \omega) \right)$$

$$= e^2 \frac{dn}{d\mu} \nabla^2 d(r, r'; \omega)$$

(53)

(54)

Equations (52) and (53) give the alternative form

$$D_D \rho(r; \omega) = -\sigma_D \phi(r; \omega) - i\omega e^2 \frac{dn}{d\mu} \int d(r, r'; \omega) \phi(r'; \omega) \, dr'$$

(55)

the gradient of which, combined with (51), yields the differential (quasilocal) constitutive ansatz (11) in Sec. II.

The field equation (15) is similarly verified by adding Poisson’s equation to eq. (55), namely,

$$\left( \nabla^2 - \Lambda^2 \right) \phi(r; \omega) = \frac{i\omega \Lambda^2}{D_D} \int d(r, r'; \omega) \phi(r'; \omega) \, dr'$$

(56)

This integro-differential form is typically how self-consistency appears in the microscopic theories mentioned earlier. But taking the gradient of (54) (and using eqs. (51) and (3)) again gives a quasilocal equation which rearranges to (13). Thus the dielectric method of Sec. II and eqs. (44) etc. are equivalent and the latter can be compared with the previous work.

In Ref. [3] BM derive the unscreened polarizability of a small particle which, in the present notation, is given by
Here, subscript "0" means unscreened and $S\left(\frac{\omega}{\Delta}\right)$ is the level density correlation function with $\omega = \varepsilon - \varepsilon'$ and $\Delta$ is the average level spacing. In the semiclassical limit $\omega \gg \Delta$, $S = 1$. (BM took $\alpha_0$ to be isotropic, so their eq. (18) has $(1/3) (r - r')^2$, $\vec{d}$ is $d$ less the zero-mode term $\propto V^{-1}$ (see eq. (68)) so that $\int \vec{d}d = 0$.

The unscreened polarizability here result from replacing $\phi(r)$ in eq. (52) by $\phi_0(r) = -r \cdot \mathbf{E}_0$, with $x$ measured from the "center of mass", so $\int x_\alpha d\mathbf{r} = 0$. The induced electric dipole is

$$P_{\alpha \alpha} = \int x_\alpha \rho_0(r, \omega) \, d\mathbf{r} = \left\{- \int \int x_\alpha x'_\beta \Pi(r, r'; \omega) \, d\mathbf{r}' \right\} \mathbf{E}_{0\beta}$$

which is easily shown to agree with eq. (57) in the semiclassical limit.

BM extended their work via the RPA towards a self-consistent theory; but their calculation were complicated and limited to lower order in the bubble diagrams, with the result not being self-consistent.

A second comparison is with the papers by Mehlig and Wilkinson [8] on absorption in small particles, particularly the second paper in which they derive their phenomenological formalism using eqs. (44)-(46), but with static values $\sigma_0$ and $D_0$. Their principal result is an equation of mixed form (their eq. (9) with sign change), namely

$$(D \nabla^2 + i\omega) \rho = -\sigma \nabla^2 \phi_{MW}$$

Here $\phi_{MW}$ is an effective potential, taken to be the sum of the diffusive part $\phi_{\text{static}}$ plus an ohmic part $\phi_{\text{dynamic}}$ with separate and pre designed frequency dependencies. Eq. (59) was intended as the basis for a self-consistent theory. An equation like eq. (59) derives from the operation of $d^{-1}$, or $\nabla^2 + i\omega/D$, on eq. (58), giving

$$\left(\nabla^2 - \bar{\Lambda}^2\right) \nabla^2 \phi = 0 = \left(\nabla^2 - \bar{\Lambda}^2\right) \rho$$

The right hand of (58) is the time-dependent Fermi-Thomas-Debye screening equation. If $\bar{\Lambda}^2$ is separated in its parts (eq. (18)) and $\rho$ is replaced by $-\nabla^2 \phi/4\pi$ in the first part, the form (58) results - with the differences mentioned above.

Boundary conditions on $\phi$ must be added for a complete specification of the solution of the 4th order PDE on the left or, correspondingly, boundary conditions on $\rho$ on the right. The form of the latter is already provided in Sec. III by the boundary conditions (23) for $\phi^b$.

The low-frequency absorption cross-section can be obtained by expanding eq. (22) in powers of $\omega$. The result is that, to $O(\omega^2)$, the absorption is given in terms of static quantities only and no integral equation is needed. An identical expression, for a special case of sphere, was given by Maksimenko et. al. in Ref. [1]. Our derivation shows that the region of validity is $\omega < \omega_T$. The details are in Appendix A.

B. The nonlocal dielectric function

The solution method in Sec. III used the auxiliary potential $\phi^b$ to bypass the need for the non-local dielectric function. Nonetheless, the properties of the latter are of some interest since $\epsilon$ gives an alternative approach to calculating the fields and currents. The relevant dielectric tensor $\epsilon (\mathbf{r}, \mathbf{r'}; \omega)$ is derived from the definitions (31), (32); and the constitutive equation is provided by eq. (44), giving

$$D_\alpha (\mathbf{r}; \omega) = \int \epsilon_{\alpha \beta} (\mathbf{r}, \mathbf{r'}; \omega) \mathbf{E}_\beta (\mathbf{r'}; \omega) \, d\mathbf{r'}$$

with

$$\epsilon_{\alpha \beta} (\mathbf{r}, \mathbf{r'}; \omega) = \delta_{\alpha \beta} \delta (\mathbf{r} - \mathbf{r'}) + \frac{4\pi i}{\omega} \sigma_{\alpha \beta} (\mathbf{r}, \mathbf{r'}; \omega)$$

$$= \epsilon_D (\omega) \delta_{\alpha \beta} \delta (\mathbf{r} - \mathbf{r'}) - \frac{4\pi i}{\omega} \nabla_\alpha \nabla'_\beta d (\mathbf{r}, \mathbf{r'}; \omega)$$

The properties of $\epsilon$ discussed above eqs. (44)-(46) imply that $\epsilon$ is a longitudinal tensor (which also guaranties the boundary condition (5)).
The eigenfunction expansion of $\psi$ is a natural basis for calculations. It is derived from the corresponding eigenfunctions of $d$, namely, those of the Laplacian which obey the Neumann boundary conditions: eq. (62) can be written as

$$\nabla^2 \Omega_m (r) + \lambda_m^2 \Omega_m (r) = 0$$
$$\nabla_n \Omega_m (r)|_\beta = 0$$  \hspace{1cm} (64)

These form a complete orthogonal set, with real and positive $\lambda^2_m$, except for the $m = 0$, ”zero mode”

$$\Omega_0 = V^{-1/2}, \lambda_0 = 0$$  \hspace{1cm} (66)

The eigenfunction representation of $d$ is

$$d(r, r'; \omega) = \sum_{m=0}^\infty \frac{\Omega_m (r) \Omega_m (r')}{\lambda^2_m - i\omega/D}$$
$$= \bar{d}(r, r'; \omega) + \frac{i\omega}{VD}$$  \hspace{1cm} (67)

where $\bar{d}$ is the sum without $m = 0$. It satisfies

$$\nabla^2 \bar{d}(r, r'; \omega) = -\delta(r - r') + \frac{1}{V} - \frac{i\omega}{D} \bar{d}(r, r'; \omega)$$  \hspace{1cm} (68)

and is the Fourier transform of the time-dependent diffusion of Ref. [3].

The orthonormal vector eigenfunctions are

$$v_m (r) = \lambda^{-1}_m \nabla \Omega_m (r); m \neq 0$$
$$\int v_m (r) \cdot v_n (r) \, dr = \delta_{mn}; \hat{n} \cdot v_m|_\beta = 0$$  \hspace{1cm} (70)

are complete as discussed below for the expansion of longitudinal tensor operators.

Let $A(r) = A_L(r) + A_T(r)$, longitudinal and transverse parts. The eigenfunction sum

$$\delta^L_{\alpha\beta} (r, r') = \sum_{m \neq 0} v_{m\alpha} (r) v_{m\beta} (r')$$  \hspace{1cm} (72)

is the longitudinal $\delta$-function in the sense that

$$\int \delta^L_{\alpha\beta} (r, r') \, A_\beta (r') \, dr = A_{L\alpha} (r')$$

The action of $\delta^L_{\alpha\beta} (r, r')$ on a longitudinal electric field is equivalent to $\delta_{\alpha\beta} \delta(r - r')$. This leads to the representation

$$\epsilon_{\alpha\beta} (r, r'; \omega) = \sum_{m \neq 0} \epsilon_{m} (\omega) v_{m\alpha} (r) v_{m\beta} (r')$$  \hspace{1cm} (73)

where

$$\epsilon_{m} (\omega) = \frac{D \lambda^2_m - i\omega\epsilon_D}{D \lambda^2_m - i\omega} = 1 + \Lambda^2 \frac{1}{\lambda^2_m - \frac{i\omega}{D}}$$  \hspace{1cm} (74)

The inverse operator $\epsilon^{-1}$ is similarly expanded, with coefficients

$$(\epsilon^{-1})_m = \epsilon^{-1}_{m\beta} = 1 - \Lambda^2 \frac{1}{\lambda^2_m + \Lambda^2}$$  \hspace{1cm} (75)

Eq. (75) may be useful for calculating the electrical field within ellipsoidal samples [1]. For these cases the interior $D$-field is constant, $D_o = a_{\alpha\beta} E_{0\beta} \ (\text{sum on } \beta)$, where $a_{\alpha\beta}$ are depolarization factors. For a slab, $a_{\alpha\beta} = \delta_{\alpha\beta}$ and for a sphere $a_{\alpha\beta} = \delta_{\alpha\beta} \left(1 + 2p/\epsilon_D a^3 \right)$. Then the interior electric field is
\[ E_\alpha (\mathbf{r}; \omega) = \sum_m A_m (E_0; \omega) v_{m\alpha} (\mathbf{r}) \] (76)

where

\[ A_m = \frac{1}{\epsilon_m (\omega)} \mathbf{J}_m \cdot \mathbf{D} = \frac{1}{\epsilon_m (\omega)} \mathbf{J}_m \cdot \vec{\alpha} \cdot E_0 \] (77)

and

\[ \mathbf{J}_m = \int v_m (\mathbf{r}) \, d\mathbf{r} \] (78)

The frequency dependence resides entirely in the coefficients \( \epsilon_m (\omega) \). Substituting \( v_{m\alpha} \) by \( \Omega_m \) gives the potential distribution

\[ \phi (\mathbf{r}; \omega) = \sum_m A_m (E_0; \omega) \Omega_m (\mathbf{r}) \] (79)

Eqs. (77) and (79) are not simple to evaluate except for a slab where \( \epsilon_m \) and \( v_m \) have simple form. The calculation is given in Appendix B. The solution (41) is reproduced. We have not carried out the sum analytically for a sphere and ellipsoid.

V. ESTIMATES: DEGENERATE-ELECTRON METAL VS. NONDEGENERATE SEMICONDUCTOR

A. Screening lengths

Static screening length is \( \Lambda^{-1} \), where for metals and semiconductors respectively,

\[
\Lambda_{\text{met}}^2 = \frac{6\pi n_{\text{met}} e^2}{E_F}, \quad \text{Thomas-Fermi}
\]

\[
\Lambda_{\text{sc}}^2 = \frac{4\pi n_{\text{sc}} e^2}{k_B T \epsilon_0}, \quad \text{Debye (\epsilon_0 - dielectric constant)}
\]

and

\[
\frac{\Lambda_{\text{sc}}^{-1}}{\Lambda_{\text{met}}^{-1}} = \left( \frac{n_{\text{met}}}{n_{\text{sc}}} \right) \frac{3 k_B T \epsilon_0}{2 E_F \epsilon_0} \right)^{1/2}
\]

with \( n_{\text{met}} \sim 10^{22} - 10^{23}\text{cm}^{-3}, n_{\text{sc}} \sim 10^{18}, \epsilon_0 \sim 10 \) screening length in semiconductor is \( \sim (10 - 100) \times \) screening length in metal. For a metal,

\[
\Lambda_{\text{met}}^{-1} \sim 0.1\text{nm}
\]

is much less than mean free path \( \ell \sim 10\text{nm} \) typical for dirty metal particles. For a semiconductor,

\[
\Lambda_{\text{sc}}^{-1} \sim (3 - 10)\text{nm}
\]

It is evident that the correction \( \delta = 5.5/\Lambda \alpha \) in eq. (37) will be too small to observe in metal particles. For a 50nm particle

\[
\delta_{\text{met}} \sim 10^{-2}
\]

On the other hand, for a semiconductor particle of the same radius

\[
\delta_{\text{met}} \sim 3 - 1
\]
B. Magnetic dipole absorption

The dipolar absorption of light is the sum of electric and magnetic parts

\[ Q_E = \frac{1}{2} \omega E_0^2 \operatorname{Im} \alpha_E \quad \text{and} \quad Q_H = \frac{1}{2} \omega E_0^2 \operatorname{Im} \alpha_H \]

In the electric case the field is screened by the surface layer (neglecting diffusion) by a factor \( \sim \omega/\sigma_0 \), while, in the magnetic case the currents penetrate to the skin effect depth \( \sqrt{2\pi\sigma_0\omega/c} \), which is greater than the particle size. Taking the expressions from Ref. [1] §§ 72-73, we get for a sphere the ratio

\[ \frac{Q_H}{Q_E} = \frac{8\pi^2 a^2 \sigma_0^2 H_0^2}{45 \omega^2 E_0^2} \]

In free space the last factor is one. For the metallic particle \((a \sim 50nm)\)

\[ \frac{Q_H}{Q_E} \sim 40 \]

but for a semiconductor

\[ \frac{Q_H}{Q_E} \sim \left( \frac{\sigma_{sc}}{\sigma_{met}} \right)^2 \times \text{above} \]

which can be much smaller than one.

VI. CONCLUSIONS

The classical Maxwell dielectric theory based on the "generalized Einstein" constitutive equation [1] reproduces the results of the previous semiclassical treatments of the response of a small "dirty" conductor to an oscillating electric field in the limit of the continuous energy spectrum. When the discreteness of energy levels can no longer be neglected, the quantum theory becomes important. Both classical and semiclassical treatments require for their validity that the mean free path \( \ell \) be small in comparison with the particle dimension, in the latter case because the matrix elements between impurity wave functions are treated as the excitation of the diffusion propagators. In Sec. IV the diffusion model, exclusive of level statistics, was shown to be equivalent to the transport equation (11).

The real efficacy of the dielectric method is, of course, for the quantitative treatment of screening which, in both the nonlocal classical and quantum formalisms, involves having to solve integral equations in 3D geometries. The nonlocality is taken into account by the auxiliary field \( E_h \) which represents the distributions of charge (see eq. (29)). Solving for the fields is not significantly more complicated than for bodies with local dielectric constants.

An unanswered question in the above considerations is the validity of the screening description at short distances. There are three lengths of interest: the mean free path \( \ell \), the screening distance \( \Lambda^{-1} \) and the particle size \( a \). In metallic particles \((\ell \ll a)\) typically \( \Lambda^{-1} \) is much less than \( \ell \) and the transport equation (11) has questionable physical meaning on the scale of \( \Lambda^{-1} \). Nonetheless, in the limit \( \Lambda^{-1} \to 0 \) where all charge is on the surface, the classical Rayleigh-Drude theory of metallic particle is obtained. This suggests that the correction for finite screening in eqs. (13) and (37) is still qualitatively correct. The same criticism applies to the quantum mechanical descriptions in Refs. [2], [3] and [4] as well as to Pines-Nozieres equation (18). The theory here may provide a more consistent picture for semiconductor particles where screening lengths are much larger.

It is interesting that, despite the above mentioned physical limitation, the mean free path itself does not appear explicitly – only scattering time \( \tau \) in the Drude form \( 1 - i\omega\tau \). Nor does the Thouless frequency \( \omega_T \) appear explicitly. It does determine the applicability of the low-frequency absorption in Appendix A.

Finally we conjecture that in the extreme quantum region when the levels are discrete and \( \omega \) and/or \( T < \Delta \), use of the "Golden Rule" and impurity-averaged spectral quantities like \( S(\omega/\Delta) \) is fundamentally wrong. We will return to this point in later work.
APPENDIX A: LOW-FREQUENCY ABSORPTION

First, we review expressions for absorption of energy from an applied electric field. The local absorption rate is

\[ q(r; \omega) = \frac{1}{2} \text{Re} \left\{ j(r; \omega) \cdot E^*(r; \omega) \right\} \quad (A1) \]

The integrated absorption rate

\[ Q(\omega) = \int q(r; \omega) \, dr \quad (A2) \]

can be rewritten for a longitudinal electric field by applying Gauss' theorem and the continuity equation,

\[ Q(\omega) = \frac{\omega}{2} \text{Im} \left\{ \int \rho(r; \omega) \phi^*(r; \omega) \, dr \right\} \quad (A3) \]

The symmetry of this form is used in the derivation of eq. (A11) below.

A less symmetric form for \( Q(\omega) \) in which \( \phi(r) \) is replaced by \( \phi_0(r) \), the applied field in the absence of the sample, is familiar from thermodynamic arguments (Ref. [1]) for quasistatic fields. We remark that replacing \( \phi \) by \( \phi_0 \) is valid at all frequencies because Coulomb forces are conservative and do no net work in a cycle. We digress briefly to prove this (probably well-known) statement, namely that the induced field

\[ \phi_{\text{ind}} = \phi - \phi_0 \]

does no work. Because the system is isolated electrically, the total induced charge \( \rho_{\text{ind}} \) is zero and \( \phi_{\text{ind}} \sim r^{-2} \) at large distances. (An initial charge distribution \( \rho_0 \) does not contribute to \( Q \) and is ignored.) So \( \rho_{\text{ind}} = \rho \) and

\[ \nabla^2 \phi = \nabla^2 \phi_{\text{ind}} = 4\pi \rho_{\text{ind}} \quad (A4) \]

with different conditions at "infinity." To see that

\[ \Delta Q = \int \text{Im} \left\{ \rho_{\text{ind}} \phi_{\text{ind}} \right\} = 0 \quad (A5) \]

integrate by parts over all space. The surface term goes to zero at large \( r \), and the new integrand \( \text{Im} \left\{ |\nabla \phi_{\text{ind}}|^2 \right\} \) vanishes.

For the small systems considered here \( \phi_0(r) = -E_0 \cdot r \) so that

\[ Q(\omega) = \frac{\omega}{2} \text{Im} \left\{ \vec{P} \cdot E_0^* \right\} \quad (A6) \]

where \( \vec{P} \) is the induced moment \( \vec{P} = \int r \rho_{\text{ind}}(r; \omega) \, dr \), which is \( \int P(r; \omega) \, dr \) for the dielectric medium (eq. (7)). For linear response, \( \vec{P} = \alpha E_0 \), \( (A6) \) becomes

\[ Q(\omega) = \frac{\omega}{2} |E_0|^2 \text{Im} \left\{ \alpha(\omega) \right\} \quad (A7) \]

To lowest order in \( \omega \), \( Q \propto \omega^2 \). A symmetrical form of the coefficient of \( \omega^2 \) can be derived from eq. \( (A3) \) by expanding \( \rho \) and \( \phi \) to first order in \( \omega \). Write

\[ \rho = \rho_{\text{st}} + \omega \rho_1, \phi = \phi_{\text{st}} + \omega \phi_1 \quad (A8) \]

Now \( \phi_{\text{st}} = \phi^h \) because \( \epsilon_D(0) = \infty \), so that

\[ \rho_{\text{st}} = -\frac{\Lambda^2}{4\pi} \phi_{\text{st}} \quad (A9) \]

The product of the static terms is real so it gives zero in \( (A3) \), as expected, and leaves

\[ Q = \frac{\omega^2}{2} \text{Im} \left\{ \int (\rho_1 \phi^*_{\text{st}} - \rho_{\text{st}} \phi^*_1) \, dr \right\} \quad (A10) \]
To \( O (\omega) \) eqs. (52)-(54) give

\[
\rho_1 = \int [\Pi_{st} (r, r') \phi_1 (r') + \Pi_1 (r, r') \phi_{st} (r')] \, dr'
\]

(A11)

where

\[
\Pi_{st} (r, r') = \Pi (r, r'; 0) = -i e^2 \frac{d}{d\mu} \delta (r - r')
\]

(A12)

and

\[
\Pi_1 (r, r') = -\frac{i e^2}{D} \frac{d}{d\mu} d (r, r'; 0)
\]

(A13)

The term linear in \( \phi_1 \) in the brackets of eq. (A11) is real so that the absorption, to order \( \omega^2 \), is determined by \( \omega = 0 \) quantities

\[
\frac{\omega^2}{2} A \int \int \phi_{st}^*(r) d (r, r'; 0) \phi_{st} (r') \, dr' \, dr
\]

(A14)

where

\[
A = \frac{e^2}{D} \frac{dn}{d\mu} = \frac{1}{\sigma_0} \left( \frac{e^2}{D} \frac{dn}{d\mu} \right)^2 = \frac{1}{\sigma_0} \left( \frac{A^4}{4\pi^2} \right)^2
\]

(A15)

From the eigenfunction expansion of \( d (r, r'; \omega) \), eq. (68), we see that \( d (\omega) \approx d (0) \) as long as \( \omega < D \lambda_1^2 \sim \omega_T \), the Thouless frequency. The Drude frequency correction \( \omega_T \omega \) is determined by \( \sigma_0 \) and \( D \) is negligible at these frequencies since \( \omega_T \omega \approx (r / L)^2 \). The latter must be small in order for the constitutive equation (11) to be meaningful physically.

We have checked (A14) for the spherical case (eq. (33)). From eq. (32)

\[
\phi_{st} (r) = (-E_0 \cdot r) f (r)
\]

(A16)

\[
f (r) = \frac{3a}{r} \csc (a \Lambda) i_1 (r \Lambda)
\]

(A17)

\( d (r, r'; 0) \) is seen from eqs. (66), (67) be the Coulomb Green function with Neumann boundary conditions

\[
d (r, r'; 0) = \frac{1}{D} \left[ \frac{1}{4\pi |r - r'|} + \frac{1}{2V} \left( \frac{r^2}{3} - a^2 \right) + \sum_{l=1}^{\infty} \frac{(l + 1) (rr')^l}{4\pi \sqrt{a^{2l+1}}} P_l (\cos \theta) \right]
\]

(A18)

where \( P_l (\cos \theta) \) is the Lagrange polynomial and \( \theta \) is the angle between \( r \) and \( r' \). Only the \( l = 1 \) component

\[
d_1 (r, r'; 0) = \frac{1}{D} \left( \frac{1}{4\pi r_+ z} + \frac{rr'}{2\pi a^2} \right) \cos \theta
\]

(A19)

(where \( r_+ \) and \( r_- \) denote the lesser and greater, respectively, of \( r \) and \( r' \)) contributes to the integral of eq. (A14). The result

\[
Q = \frac{3a^2 \omega^2}{8\pi \sigma_0} |E_0|^2 F (\Lambda a)
\]

(A20)

\[
= Q_{RD} F (\Lambda a)
\]

(A21)

where

\[
F (z) = 1 - \frac{6}{z} \left( \coth z - \frac{1}{z} \right) + \frac{6}{z^2} \left( \coth z - \frac{1}{z} \right)^2 + \frac{1}{2z} \coth z - \frac{1}{2} \left( \coth^2 z - 1 \right)
\]

(A22)

agrees with the term \( \text{Im} \left\{ \alpha \right\} \) (33) which is linear in \( \omega \).

Finally, we compare (A14) with the expression for absorption obtained by Maksimenko et. al., in Ref. 1 who extended the semiclassical method of GE [3] to take screening into account. The unscreened dipolar energy \( -e r \cdot E_0 \) is replaced by a "screened" dipole \( -e R \cdot E_0 \). The latter is found within the RPA in which the irreducible polarization
part has the same form as polarization itself, namely, having matrix elements between disordered states. The solution of the RPA integral equation, after an average over states at the Fermi surface and restricting to spherical geometry, has the same form as eqs. (A16), (A17):

\[
\phi_{st}(r) = -R(r) \cdot E_0 \quad \text{(A23)}
\]

\[
R(r) = rf(r) \quad \text{(A24)}
\]

The agreement reflects the correspondence between the self-consistency of the RPA and of the Maxwell dielectric theory. (The resulting formula for \(Q\) in Ref. [4] contains the level correlation function \(S(\omega/\Delta)\) and reduces to the present form when \(S = 1\).)

**APPENDIX B: DIELECTRIC TENSOR IN A SLAB**

The eigenfunctions and eigenvalues of \(d(r, r'; \omega)\) are corresponding to eqs. (64) and (65),

\[
\Omega(z, z'; \omega) = \sqrt{\frac{2}{L}} \cos \left( \frac{m\pi (z + L/2)}{L} \right), -\frac{L}{2} \leq z \leq \frac{L}{2} \quad \text{(B1)}
\]

and

\[
\lambda^2 - \frac{i}{D_D} \omega = \left( \frac{m\pi}{L} \right)^2 - \frac{i}{D_D} \frac{\omega}{D_D} \quad \text{(B2)}
\]

The vector eigenfunctions are

\[
v_m(z) = \hat{z} \sqrt{\frac{2}{L}} \sin \left( \frac{m\pi}{L} \left( z + \frac{L}{2} \right) \right) \quad \text{(B3)}
\]

We treat the case in Sec. III where \(E_0 \parallel \hat{z}\) so the vector notation \(\hat{z}\) can be dropped. Then \(D = E_0\) and the prescription in eq. (74) gives

\[
\frac{E(z; \omega)}{E_0} = \frac{4}{\pi} \sum_{n} \frac{1}{2n+1} \left( 1 - \Lambda^2 \frac{1}{(2n+1) \frac{\pi}{L})^2 + \Lambda^2} \right)^{-1} \sin \left( \frac{m\pi (z + L/2)}{L} \right) \quad \text{(B4)}
\]

The summation is easily done and yields the solution (41).

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