Absorption of radiation by small metallic particles: a general self-consistent approach

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(June 28, 2006)

Abstract

We introduce a theory for the absorption of electromagnetic radiation by small metal particles, which generalises the random phase approximation by incorporating both electric and magnetic dipole absorption within a unified self-consistent scheme. We demonstrate the equivalence of the new approach to a superficially dissimilar perturbative approach. We show how to obtain solutions to the self-consistent equations using a classical approximation, taking into account the non-locality of the polarisability and the conductivity tensor. We discuss the nature of the self-consistent solutions for diffusive and ballistic electron dynamics.

arXiv:physics/9907036 v1   21 Jul 1999

I. INTRODUCTION

The quantum theory of absorption of radiation by small metal particles has proved to be a difficult area, partly because it has not always been realised that the internal electric field must be treated self-consistently, and partly because of confusion about how to obtain the self-consistent field. The literature is surveyed in [1-3], and the introductory sections of [4] and [5] include a brief review of the literature relevant to the present paper. This paper extends a series of works [4-8] which have considered the self-consistent potential from a semiclassical viewpoint.

First we clarify the regimes in which our results are applicable. The following frequency scales are relevant to the interaction of small metal particles with radiation. The lowest scale is given by the mean level spacing Δ, i.e., $\omega_\Delta = \Delta/\hbar$. In d dimensions $\omega_\Delta \propto a^{-d}$, where a is the characteristic size of the particle. Another typical frequency scale is given by the inverse of the typical time taken for an electron to traverse the particle. In systems with diffusive electron motion it is given by $\omega_c = D/a^2$, where D is the diffusion constant. In particles with ballistic electron motion, ω_c is given by the inverse time of flight, $\omega_c = v_F/a$, where v_F is the Fermi velocity. The highest frequency scales are the plasma frequency ω_p and the frequency $\omega_F = E_F/\hbar$ derived from the Fermi energy. In two dimensions, $\omega_p \propto a^{-1/2}$, whereas in three dimensions ω_p is comparable to the Fermi frequency, which is independent of the size of the particle. The frequency scales are therefore ordered as follows

$$\omega_\Delta \ll \omega_c \ll \omega_p \leq \omega_F \,. \tag{1}$$

Our results are relevant to frequencies satisfying $\omega \gg \omega_\Delta$ (which justifies the neglect of quantum effects), and $\omega \ll \omega_p$ (which means that screening of external fields by polarisation of the particle is significant). There are six relevant length scales in the problem, namely the wave length λ of the external radiation, the linear dimension a of the particle, the skin depth λ_s, the Thomas-Fermi screening length λ_{TF}, the Fermi wavelength λ_F and the mean free path l. In the following it will be assumed that

$$\lambda, \lambda_s \gg a \gg \lambda_{TF} \sim \lambda_F \,. \tag{2}$$

Both diffusive ($l \ll a$) and ballistic ($l \gg a$) dynamics will be discussed.

Under these conditions the absorption can be divided into electric and magnetic dipole contributions. In earlier papers these were discussed separately: the electric dipole absorption was discussed for various different situations in [4-7], and the magnetic dipole absorption case was discussed in [8]. It is desirable to have a unified description. In section II we develop a generalisation of the commonly used 'random phase approximation' [9] scheme which gives a uniform approach to both electric and magnetic dipole absorption. Most treatments of the electric dipole absorption coefficient are based upon calculating the imaginary part of the polarisability. References [4-7] used an alternative approach, namely time dependent perturbation theory in the effective potential. In section III we show that these apparently dissimilar approaches are equivalent.

We also discuss a unified approach to determining the self-consistent field: we present some new results for the ballistic case, where the bulk mean free path of the electrons is large compared to the dimensions of the particle. Section VI describes the form of the non-local polarisability, and section V discusses a general semiclassical method for determining

the self-consistent field. We show that the self-consistent potential used in [4,6], which treated electric dipole absorption ballistic systems, was not correct for $\omega \gg \omega_{\mathrm{c}}$. Section VI summarises the results, and presents an argument indicating that the results of [4,6] are nevertheless qualitatively correct.

II. CALCULATION OF THE ABSORPTION COEFFICIENT

This section will discuss the general principles underlying the calculation of the electromagnetic response.

A. Formulation of the problem

An electromagnetic wave induces currents which result in both electric and magnetic polarisation of a conducting conducting particle. In what follows we will only consider linear effects (where the polarisation is proportional to the applied field), and the externally applied field will be assumed to be uniform over the dimension of the particle. We will only be concerned with the coefficients relating dipolar moments to the externally applied field: higher moments will not be considered. The electric dipole $\mathbf{d}$ and magnetic dipole $\mathbf{m}$ of a single particle are given by

$$\mathbf{d} = \tilde{\alpha}\,\mathbf{E}_{\mathrm{ext}}, \quad \mathbf{m} = \tilde{\beta}\,\mathbf{B}_{\mathrm{ext}} + \tilde{\gamma}\,\mathbf{E}_{\mathrm{ext}} \tag{3}$$

where $\mathbf{E}_{\mathrm{ext}}$ and $\mathbf{B}_{\mathrm{ext}}$ are the externally applied electric and magnetic fields, $\tilde{\alpha}$ and $\tilde{\beta}$ are the electric and magnetic susceptibility tensors of the particle. The cross-susceptibility $\tilde{\gamma}$ is not usually included. It is absent for spherical particles and some other symmetric geometries, and when it is non-zero it vanishes in the low-frequency limit. We will ultimately give a completely general treatment, showing that the cross-susceptibility makes no contribution to the absorption, but for simplicity the cross-term will be dropped in the remainder of this introductory section. The externally applied fields are assumed to be multiplied by a factor of the form $\exp(-\mathrm{i}\omega t)$, and the polarisability tensors are understood to be functions of ω with complex valued components, because there may be a phase shift between the applied field and the response. For example, the actual value of the dipole moment at time t is taken to be $\mathbf{d}(t) = \mathrm{Re}[\mathbf{d}\exp(-\mathrm{i}\omega t)]$.

These polarisations are detectable at a macroscopic level in various ways: they alter the dielectric constant and magnetic permeability of the medium in which the particles are dispersed, and they may also be detected by observing scattering and absorption of radiation. The polarisability determines two processes which result in the attenuation of radiation, namely scattering and absorption of energy. Both of these processes can be characterised conveniently at the microscopic level by calculating the rate of loss of energy from the incident beam due to interaction with a single particle: this will be denoted by $\overline{dE/dt}$ [where $\overline{X}$ denotes the time average of $X(t)$]. The two most commonly used measures used to quantify these processes are the cross section per particle $\mathcal{S}$ and the attenuation coefficient γ. To relate the energy loss to the cross section, note that the energy density in an electromagnetic wave is $\frac{1}{2}\epsilon_0\mathbf{E}^2$: the cross section is therefore

$$\mathcal{S} = \frac{2}{\epsilon_0 c \mathbf{E}^2} \overline{\frac{dE}{dt}} \qquad (4)$$

where $\overline{X}$ denotes the time axerage of $X(t)$. The attenuation coefficient γ is defined by the expression $I = I_0 \exp(-\gamma z)$, where I is the intensity at distance z along the beam. The attenuation coefficient is given by $\gamma = \mathcal{N}\mathcal{S}$, where $\mathcal{N}$ is the particle density.

At low frequencies the real part of the polarisability approaches a constant (and the imaginary part approaches zero). It follows that at sufficiently low frequencies the scattering cross section scales as ω^4. It will be shown that in the absorption cross section typically scales as ω^2, implying that absorption is expected to be the dominant process at low frequencies.

The absorption of radiation can be related to the imaginary parts of the polarisability tensors: we will give a careful explanation of this. Electron spin is not significant in this context, and the full Hamiltonian for the electrons is taken to be

$$H = \sum_{i=1}^{N} \frac{1}{2m}[\mathbf{p}_i - e\mathbf{A}_{\text{ext}}(\mathbf{r}_i, t)]^2 + V(\mathbf{r}_i) + \phi_{\text{ext}}(\mathbf{r}_i, t) + \frac{1}{2}\sum_{i=1}^{N}\sum_{\substack{j=1 \\ j \neq i}}^{N} \frac{e^2}{4\pi\epsilon_0|\mathbf{r}_i - \mathbf{r}_j|} \ . \qquad (5)$$

The externally applied electric and magnetic fields are considered to be spatially uniform, since the particle is small compared to the wavelength of the radiation, we ignore the spatial dependence of the electric and magnetic fields, and write:

$$\phi_{\text{ext}}(\mathbf{r}, t) = e\,\mathbf{r}.\mathbf{E}_{\text{ext}}, \quad \nabla \wedge \mathbf{A}_{\text{ext}}(\mathbf{r}, t) = \mathbf{B}_{\text{ext}}(t) \ . \qquad (6)$$

In the case where the circularly symmetric gauge

$$\mathbf{A}_{\text{ext}} = \mathbf{A}_{\text{rot}}(\mathbf{r}, t) = \tfrac{1}{2}\mathbf{B}_{\text{ext}}(t) \wedge \mathbf{r} \qquad (7)$$

is used, the full Hamiltonian contains terms coupling the system to the electric and magnetic fields, of the following form:

$$\hat{H}(t) = \hat{H}_0 + e\hat{\mathbf{X}}.\mathbf{E}_{\text{ext}}(t) + \frac{e}{2m}\hat{\mathbf{L}}.\mathbf{B}_{\text{ext}}(t) + O(\mathbf{B}_{\text{ext}}^2)$$

$$\hat{\mathbf{X}} = \sum_{i=1}^{N} \hat{\mathbf{r}}_i, \quad \hat{\mathbf{L}} = \sum_{i=1}^{N} \mathbf{r}_i \wedge \mathbf{p}_i \qquad (8)$$

where $\hat{\mathbf{X}}$ and $\hat{\mathbf{L}}$ are the total dipole operator and total angular momentum operators, and $\hat{H}_0$ is independent of time.

To facilitate the calculations we will consider ensemble averages of quantities: if the electron motion is ergodic, this is the microcanonical average, and in general the ensemble is defined by the region of phase space explored by the dynamics. Angle brackets will be used for the appropriate ensemble average. For a general choice of gauge the instantaneous rate of absorption is then

$$\left\langle \frac{dE}{dt} \right\rangle = \left\langle \frac{\partial H}{\partial t} \right\rangle = e\left\langle \sum_{i=1}^{N} \mathbf{v}_i(t).[\mathbf{E}_{\text{ext}}(t) + \mathbf{E}_{\text{ind}}(t)] \right\rangle = \int d\mathbf{r}\, \mathbf{j}(\mathbf{r}, t).[\mathbf{E}_{\text{ext}}(t) + \mathbf{E}_{\text{ind}}(\mathbf{r}, t)] \quad (9)$$

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where $\mathbf{v}_i$ is the velocity of the ith electron, $\mathbf{E}_{\text{ind}} = \mathrm{i}\omega\mathbf{A}_{\text{ext}}$ is the electric field induced by the varying magnetic field, and

$$\mathbf{j}(\mathbf{r}, t) = e\left\langle \sum_{i=1}^{N} \mathbf{v}_i(t)\, \delta[\mathbf{r} - \mathbf{r}_i(t)] \right\rangle \tag{10}$$

is the current density within the particle. In the special case where the circularly symmetric gauge is used this reduces to:

$$\left\langle \frac{dE}{dt} \right\rangle = \left\langle \frac{\partial H}{\partial t} \right\rangle = e\langle\hat{\mathbf{X}}\rangle.\dot{\mathbf{E}}_{\text{ext}}(t) + \frac{e}{2m}\langle\hat{\mathbf{L}}\rangle.\dot{\mathbf{B}}_{\text{ext}}(t) \tag{11}$$

where $\langle\hat{\mathbf{X}}\rangle$ and $\langle\hat{\mathbf{L}}\rangle$ are suitable averages of the centre of mass and angular momentum operators. It is impractical to calculate these averages from the full Hamiltonian (5), and in the next sub-section it will be shown how they may be estimated using an *effective* Hamiltonian, containing effective fields $\mathbf{A}_{\text{eff}}$ and ϕ_{eff}. At this stage we will only assume that these averages are proportional to the applied fields. These quantities $\langle\hat{\mathbf{X}}\rangle$ and $\langle\hat{\mathbf{L}}\rangle$ are related to the electric and magnetic dipole moments $\mathbf{d}$ and $\mathbf{m}$:

$$\mathbf{d} = e\langle\mathbf{X}\rangle, \quad \mathbf{m} = \frac{e}{m}\langle\mathbf{L}\rangle \ . \tag{12}$$

The rate of absorption is obtained by substituting for the time dependence of a monochromatic field using (3), and ignoring the cross term:

$$\left\langle \frac{dE}{dt} \right\rangle = -\text{Re}[\mathbf{d}\exp(-\mathrm{i}\omega t)]\,\text{Re}[\mathrm{i}\omega\,\mathbf{E}_{\text{ext}}\exp(-\mathrm{i}\omega t)]$$
$$-\tfrac{1}{2}\text{Re}[\mathbf{m}\exp(-\mathrm{i}\omega t)]\,\text{Re}[\mathrm{i}\omega\,\mathbf{B}_{\text{ext}}\exp(-\mathrm{i}\omega t)] \ . \tag{13}$$

Averaging over time gives the general form for the rate of absorption

$$\left\langle \overline{\frac{dE}{dt}} \right\rangle = \tfrac{1}{4}\omega\mathbf{E}_{\text{ext}}^{+}(\tilde{\alpha} - \tilde{\alpha}^{+})\mathbf{E}_{\text{ext}} + \tfrac{1}{8}\omega\mathbf{B}_{\text{ext}}^{+}(\tilde{\beta} - \tilde{\beta}^{+})\mathbf{B}_{\text{ext}} \ . \tag{14}$$

In the case where the polarisability tensor is isotropic, and the radiation field is plane polarised, this expression becomes

$$\left\langle \overline{\frac{dE}{dt}} \right\rangle = \tfrac{1}{2}\omega\,\text{Im}[\alpha_{ii}(\omega)]\,|\mathbf{E}_{\text{ext}}|^2 + \tfrac{1}{4}\omega\,\text{Im}[\beta_{ii}(\omega)]\,|\mathbf{B}_{\text{ext}}|^2 \tag{15}$$

We note that under the assumptions listed above, the absorption is expressed as the sum of two terms, which are naturally referred to as the electric and magnetic dipole absorption. Our final result will not neglect the magnetic dipole moment which may be induced by the electric field, but we will show that within the framework of our self-consistent approximation scheme the cross term in (3) makes no contribution to the absorption. The energy absorbed does not accumulate in the system electronic system: most of it is eventually transformed into heat by interaction with phonons.

B. Self-consistent fields

The Hamiltonian will be approximated by an effective Hamiltonian, in which the electrons move independently. The direct interaction with the magnetic field via electron spin can also be neglected, and the effective Hamiltonian is of the form

$$\hat{H}_{\text{eff}} = \sum_{i=1}^{N} \frac{1}{2m} [\hat{\mathbf{p}}_i - e\mathbf{A}_{\text{eff}}(\mathbf{r}_i, t)]^2 + V_{\text{eff}}(\mathbf{r}_i) + \phi_{\text{eff}}(\mathbf{r}_i, t) \ . \tag{16}$$

The response of the system is determined by the interaction of the electrons with the electric field inside the particle, which is described by the effective potentials $\mathbf{A}_{\text{eff}}$ and ϕ_{eff}. The effective potentials are themselves determined by the distribution of charge within the particle. The external magnetic field is also augmented by an induced magnetic field which is produced by the action of the currents which flow in order to establish the electric polarisation. Provided the particle is sufficiently small, the induced magnetic field can be neglected, and our self-consistent theory will yield and equation for the electric field $\mathbf{E}(\mathbf{r}, \omega)$ within the particle, which is related to the time-dependent electric field as follows:

$$\mathbf{E}(\mathbf{r}, \omega) = \int_{-\infty}^{\infty} dt \, \mathbf{E}(\mathbf{r}, t) \exp(-\mathrm{i}\omega t) \ . \tag{17}$$

This field satisfies the Maxwell equations

$$\nabla . \mathbf{E} = \frac{\rho}{\epsilon_0} \ , \quad \nabla \wedge \mathbf{E} = \mathrm{i}\omega \mathbf{B}_{\text{ext}} \ . \tag{18}$$

The electric field produced by induction when the external electric field is zero will be denoted $\mathbf{E}_{\text{ind}}$. The total effective electric field is

$$\mathbf{E}(\mathbf{r}, \omega) = \mathbf{E}_{\text{ext}}(\mathbf{r}, \omega) + \mathbf{E}_{\text{ind}}(\mathbf{r}, \omega) + \nabla \phi_{\text{pol}}(\mathbf{r}, \omega) \ . \tag{19}$$

The uniform external electric field satisfies $\nabla . \mathbf{E}_{\text{ext}} = 0$, and can be derived from an external potential:

$$\mathbf{E}_{\text{ext}}(\mathbf{r}, \omega) = \nabla \phi_{\text{ext}}(\mathbf{r}, \omega) \ . \tag{20}$$

The potential ϕ_{pol} results from polarisation of the particle due to the external electric field, and is given by

$$\phi_{\text{pol}}(\mathbf{r}, \omega) = \frac{1}{4\pi\epsilon_0} \int d\mathbf{r}' \, \frac{\rho_{\text{pol}}(\mathbf{r}', \omega)}{|\mathbf{r} - \mathbf{r}'|} \tag{21}$$

where $\rho_{\text{pol}}(\mathbf{r}, \omega)$ is the charge density resulting from polarisation induced by the external electric field, but excluding any polarisation which may result from the induction field. It will be convenient to express (19) using the notation

$$|\mathbf{E}) = \nabla|\phi_{\text{ext}}) + |\mathbf{E}_{\text{ind}}) + \nabla \widehat{U}|\rho_{\text{pol}}) \tag{22}$$

where $\widehat{U}$ is an operator defined by (21), acting on the 'field vector' $|\rho_{\text{pol}})$. The dependence upon frequency will usually be shown explicitly for operators, but not for field vectors.

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The current density $\mathbf{j}(\mathbf{r}, \omega)$ flowing in the sample to build up the charge density $\rho(\mathbf{r}, \omega)$ may be assumed to be linearly related to the electric field $\mathbf{E}(\mathbf{r}, \omega)$ in the sample

$$\mathbf{j}(\mathbf{r}, \omega) = \int d\mathbf{r}' \, \Sigma(\mathbf{r}, \mathbf{r}'; \omega) \, \mathbf{E}(\mathbf{r}', \omega) \,. \tag{23}$$

In condensed notation, we write

$$|\mathbf{j}) = \widehat{\Sigma}(\omega) \, |\mathbf{E}) \tag{24}$$

where $\widehat{\Sigma}(\omega)$ is the conductivity operator. The non-local conductivity tensor $\Sigma(\mathbf{r}, \mathbf{r}', \omega)$ is related to the non-local polarisability operator $\Pi(\mathbf{r}, \mathbf{r}', \omega)$, which gives the charge density induced by a potential $\phi(\mathbf{r})$: we write

$$\rho(\mathbf{r}, \omega) = \int d\mathbf{r}' \, \Pi(\mathbf{r}, \mathbf{r}'; \omega) \, \phi(\mathbf{r}', \omega) \tag{25}$$

or in condensed notation

$$|\rho) = \widehat{\Pi}(\omega) \, |\phi) \,. \tag{26}$$

The polarisability operator $\widehat{\Pi}(\omega)$ can be related to a non-local conductivity tensor $\widehat{\Sigma}(\omega)$ by a continuity relation. Using (23) and applying the continuity equation, we find (with summation over repeated indices implied)

$$0 = \int d\mathbf{r}' \, \left[i\omega \, \Pi(\mathbf{r}, \mathbf{r}'; \omega) \phi(\mathbf{r}') - \nabla_i \Sigma_{ij}(\mathbf{r}, \mathbf{r}'; \omega) \nabla_j' \phi(\mathbf{r}') \right] \tag{27}$$

assuming that the normal component of $\widehat{\Sigma}(\omega)$ vanishes on the boundary. Upon integration by parts, after noting that the resulting equation is valid for any field $\phi(\mathbf{r}, \omega)$, we find

$$i\omega \, \widehat{\Pi}(\omega) = -\overrightarrow{\nabla} \widehat{\Sigma}(\omega) \overleftarrow{\nabla} \,. \tag{28}$$

The quantities $\widehat{\Pi}(\omega)$ and $\widehat{\Sigma}(\omega)$ enable (22) to be expressed in terms of the electric field alone, yielding a self-consistent equation. We write

$$|\mathbf{E}) = \nabla |\phi_{\text{eff}}) + |\mathbf{E}_{\text{ind}}) \tag{29}$$

where $\phi_{\text{eff}}(\mathbf{r}, \omega)$ is an effective potential. We consider the solutions for the field $\mathbf{E}_{\text{ind}}$ and the potential ϕ_{eff} separately. The charge induced by the field $\mathbf{E}_{\text{ind}}$ is

$$|\rho_{\text{ind}}) = \frac{1}{i\omega} \nabla \widehat{\Sigma}(\omega). \, |\mathbf{E}_{\text{ind}}) \,. \tag{30}$$

Applying the first of the Maxwell equations (18) and using (30) gives

$$\nabla.[|\mathbf{E}_{\text{ind}}) - i\omega \widehat{\Sigma}(\omega)|\mathbf{E}_{\text{ind}})] = 0 \tag{31}$$

which is the self-consistent equation which must be solved for the field $\mathbf{E}_{\text{ind}}$. For the effective potential, we find

$$|\phi_{\text{eff}}) = |\phi_{\text{ext}}) + \widehat{U} \widehat{\Pi}(\omega)|\phi_{\text{eff}}) \,. \tag{32}$$

This self-consistent equation is sometimes referred to as the 'random phase approximation' [9]. Equations (31) and (32) must must be solved for the self-consistent fields. We will consider semiclassical methods for solving them in section V.

C. The rate of energy absorption

The rate of energy absorption is given by (9). Averaging over time gives

$$\left\langle \overline{\frac{dE}{dt}} \right\rangle = \tfrac{1}{2} \operatorname{Re} \int d\mathbf{r}\, \mathbf{j}^*(\mathbf{r}, \omega).\mathbf{E}_{\text{ext}} + \tfrac{1}{2} \operatorname{Re} \int d\mathbf{r}\, \mathbf{j}^*(\mathbf{r}, \omega).\mathbf{E}_{\text{ind}}(\mathbf{r}, \omega) \ . \tag{33}$$

In condensed notation this will be written, by analogy with Dirac notation, as

$$\left\langle \overline{\frac{dE}{dt}} \right\rangle = \tfrac{1}{2} \operatorname{Re} (\mathbf{j}|\mathbf{E}_{\text{ext}}) + \tfrac{1}{2} \operatorname{Re} (\mathbf{j}|\mathbf{E}_{\text{ind}}) \ . \tag{34}$$

Using (22),

$$\left\langle \overline{\frac{dE}{dt}} \right\rangle = \tfrac{1}{2} \operatorname{Re} (\mathbf{j}|\mathbf{E}) - \tfrac{1}{2} \operatorname{Re} (\mathbf{j}|\nabla \widehat{U}|\rho_{\text{pol}}) = \tfrac{1}{2} \operatorname{Re} (\mathbf{j}|\mathbf{E}) - \tfrac{1}{2} \operatorname{Re} \mathrm{i}\omega(\rho_{\text{pol}}|\widehat{U}|\rho_{\text{pol}}) \tag{35}$$

where $|\rho_{\text{pol}}) = \widehat{\Pi}|\phi_{\text{eff}})$; the final equality follows from an integration by parts, and use of the continuity equation. Using the fact that $\widehat{U}$ is self-adjoint, we obtain the very simple expression for the absorption

$$\left\langle \overline{\frac{dE}{dt}} \right\rangle = \tfrac{1}{2} \operatorname{Re} (\mathbf{j}|\mathbf{E}) \ . \tag{36}$$

Using the continuity equation and (22), (28) and (29),

$$(\mathbf{j}|\mathbf{E}) = \mathrm{i}\omega \, (\phi_{\text{eff}}|\widehat{\Pi}^+(\omega)|\phi_{\text{eff}}) + (\mathbf{E}_{\text{ind}}|\widehat{\Sigma}^+(\omega)|\mathbf{E}_{\text{ind}}) \ . \tag{37}$$

This gives our final expression for the absorption

$$\left\langle \overline{\frac{dE}{dt}} \right\rangle = \tfrac{1}{2} \, \omega \operatorname{Im} (\phi_{\text{eff}}|\widehat{\Pi}(\omega)|\phi_{\text{eff}}) + \tfrac{1}{2} \operatorname{Re} (\mathbf{E}_{\text{ind}}|\widehat{\Sigma}(\omega)|\mathbf{E}_{\text{ind}}) \ . \tag{38}$$

These are two independent contributions to the the rate of absorption, depending on the electric and magnetic fields respectively. It is not obvious that these are correctly identified as the electric and magnetic dipole coefficients, because the electric field may induce a charge density with non-zero angular momentum. We will now show that the first term is due solely to the electric dipole. Using the continuity equation and an integration by parts, we find

$$\operatorname{Re}(\mathbf{j}|\mathbf{E}_{\text{ext}}) = \omega \operatorname{Im}(\rho|\phi_{\text{ext}}) = \omega \operatorname{Im}[\mathbf{d}.\mathbf{E}_{\text{ext}}] \ . \tag{39}$$

The electrically induced absorption therefore depends only upon the induced dipole moment, and is independent of the magnetic moment induced by the electric field.

III. EQUIVALENCE WITH PERTURBATION THEORY

A. An alternative expression for the the absorption coefficient

In this section, we concentrate on the electric absorption. We describe an alternative approach to calculating the absorption coefficient, which was used in [4-7], and show that it

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is equivalent to the first term in (39) provided the polarisation operator $\widehat{\Pi}(\omega)$ is related in a simple way to a propagator $\widehat{P}(\omega)$. This relation will be established in section III B.

We will consider the action of the effective potential $\phi_{\text{eff}}(\mathbf{r}, t)$ on the electrons. We may use either quantum mechanical or classical perturbation theory. We will describe the quantum mechanical approach, and will use semiclassical approximations: a classical theory in which quantum mechanics only enters in choosing the Fermi-Dirac distribution for the initial distribution of electrons gives identical results. Conceptually, the simplest method for calculating the absorption in the first using the Fermi golden rule. This is expressed in terms of matrix elements ϕ_{nm} of the perturbation in the basis $|\psi_n\rangle$ formed by the eigenstates of the single-particle effective Hamiltonian, $\hat{H}_{\text{eff}}$:

$$\begin{aligned}
\hat{H}_{\text{eff}}|\psi_n\rangle &= E_n|\psi_n\rangle \\
\phi_{nm} &= \langle\psi_n|\hat{\phi}|\psi_m\rangle, \quad \hat{\phi} = \phi_{\text{eff}}(\hat{\mathbf{r}})
\end{aligned} \tag{40}$$

The Fermi golden rule states that the rate of transition from an initially occupied state to a quasi-continuum of final states, with density of states n and with energy differing by $\hbar\omega$ from the original state, is given by

$$R = \frac{\pi n e^2}{2\hbar}\langle|\phi_{nm}|^2\rangle \tag{41}$$

where the angle brackets denote an average over matrix elements $\langle\psi_n|\hat{\phi}_{\text{eff}}|\psi_m\rangle$. We will consider the case where both the temperature and the photon energy are small compared to other energy scales in the problem; generalisations are straightforward. Absorption of energy occurs due to the excitation of electrons in occupied states below the Fermi level to empty states above the Fermi level. The number of states which can be excited is $\sim n\hbar\omega$, and the energy absorbed in each transition is $\hbar\omega$: the total rate of absorption of energy is given by multiplying these factors by the transition rate R, giving

$$\overline{\frac{dE}{dt}} = \tfrac{1}{2}\pi\hbar n^2 e^2 \omega^2 \left\langle|\phi_{nm}|^2\right\rangle . \tag{42}$$

Both (39) and (42) are quadratic functions of ϕ_{eff}, but it is not immediately clear how they can be related. We will now discuss why they are equivalent.

The mean-square matrix element can be estimated from the correlation function $C_{\phi\phi}(t)$ of the effective potential

$$\langle|\phi_{nm}|^2\rangle = \frac{1}{\pi\hbar n}\text{Re}\int_0^\infty dt\, \text{e}^{\text{i}\omega t}\, C_{\phi\phi}(t) \tag{43}$$

where the correlation function is defined by

$$C_{AB}(t) = \langle A(\mathbf{r}, \mathbf{p})B(\mathbf{r}_t, \mathbf{p}_t)\rangle \equiv \frac{1}{\Omega'(E)}\int d\mathbf{r}\int d\mathbf{p}\, A(\mathbf{r}, \mathbf{p})B(\mathbf{r}_t, \mathbf{p}_t)\delta[E_{\text{F}} - H(\mathbf{r}, \mathbf{p})]$$

$$\Omega'(E) = \int d\mathbf{r}\int d\mathbf{p}\, \delta[E - H(\mathbf{r}, \mathbf{p})] \tag{44}$$

and $\mathbf{r}_t$, $\mathbf{p}_t$ are the phase space coordinates evolved under the Hamiltonian dynamics for time t, starting from the point $(\mathbf{r}, \mathbf{p})$. It will be convenient to define a propagator $P(\mathbf{r}, \mathbf{r}'; t)$ which gives the probability of reaching $\mathbf{r}'$ from $\mathbf{r}$ in time t:

$$P(\mathbf{r}, \mathbf{r}'; t) = \langle \delta(\mathbf{r}_t - \mathbf{r}') \rangle \theta(t) \tag{45}$$

where $\theta(t)$ is a step function which makes the propagator zero for negative time. The averaging will be defined in the next subsection. With this definition we have

$$C_{\phi\phi}(t) = \frac{1}{V} \int d\mathbf{r} \int d\mathbf{r}' \ P(\mathbf{r}, \mathbf{r}'; t) \, \phi(\mathbf{r}) \, \phi(\mathbf{r}') \equiv (\phi | \widehat{P}(t) | \phi) \tag{46}$$

where the operator $\widehat{P}(t)$ is defined by analogy with (23). Introducing the Fourier transform $\widehat{P}(\omega)$ of the propagator, we have

$$\begin{aligned}
\left\langle \overline{\frac{dE}{dt}} \right\rangle &= \tfrac{1}{2}\nu e^2 \omega^2 \ \mathrm{Re} \int_0^\infty dt \ \mathrm{e}^{\mathrm{i}\omega t} (\phi_{\mathrm{eff}} | \widehat{P}(t) | \phi_{\mathrm{eff}}) \\
&= \tfrac{1}{2}\nu e^2 \omega^2 \ \mathrm{Re}(\phi_{\mathrm{eff}} | \widehat{P}(\omega) | \phi_{\mathrm{eff}})
\end{aligned} \tag{47}$$

In the next section it will be shown that there is a general relation between the propagator and the polarisability operator:

$$\widehat{\Pi}(\omega) = e\nu[\widehat{I} + \mathrm{i}\omega \widehat{P}(\omega)] \tag{48}$$

where ν is the density of states per unit volume. A relation of this form has been given by Kirzhnitz [13]. We present a detailed derivation, based on Liouville's equation, below. If the potential V_{eff} appearing in (16) is constant within the conducting particle, we may write $n = \nu V$, where V is the volume of the particle. Substituting this into (39) reproduces (47), thus establishing its equivalence to (42).

B. General relation between polarisability and the propagator

We will now relate the polarisability operator $\Pi(\mathbf{r}, \mathbf{r}'; \omega)$ to the probability propagator $P(\mathbf{r}, \mathbf{r}'; t)$, which is the probability that an electron, released at $\mathbf{r}'$ with energy equal to the Fermi energy E_{F}, will be at position $\mathbf{r}$ after time t. The discussion will be classical; a quantum mechanical derivation proceeds along similar lines.

Let the phase-space distribution be $f(\mathbf{r}, \mathbf{p}; t)$: this will, when convenient, be written $f(\alpha, t)$ where $\alpha = (\mathbf{r}, \mathbf{p})$. The Hamiltonian will be assumed to be of the form

$$H(\alpha, t) = H_0(\alpha) + X(t)H_1(\alpha) \tag{49}$$

where we will be interested in the case where $H_0 = \mathbf{p}^2/2m + V(\mathbf{r})$ and $H_1 = \phi(\mathbf{r})$. The perturbation parameter $X(t)$ is assumed to be small, so that $f(\alpha, t)$ may be expanded as a series in $X(t)$: we will be interested in the expansion as far as the first order term:

$$f(\alpha, t) = f_0(\alpha) + \int_{-\infty}^t dt' \ X(t') \, g(\alpha, t, t') + O(X^2) \ . \tag{50}$$

Substituting into the Liouville equation, $\partial_t f = \{f, H\}$, it is found that f_0 is a function of the unperturbed Hamiltonian $H_0(\alpha)$, and that the kernel $g(\alpha, t, t')$ of the first order term satisfies

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$$X(t)[g(\alpha, t, t') - \{H_1, f_0\}_\alpha] + \int_{-\infty}^t dt' \ X(t')[\partial_t g - \{H_0, g\}]_{\alpha, t, t'} = 0 \tag{51}$$

which is valid for all $X(t)$. The first term implies that

$$g(\alpha, t, t) = g(\alpha) = \{H_1, f_0\}_\alpha = \{H_1, H_0\}_\alpha \frac{\partial f_0}{\partial E}(H_0(\alpha)) \ . \tag{52}$$

The second term implies that $dg/dt = 0$, where d/dt is the total time derivative along a trajectory, so that

$$g(\alpha, t, t') = g(\alpha, t - t') = \frac{\partial f_0}{\partial E}(H_0(\alpha)) \frac{dH_1}{dt}(\alpha_{t-t'}(\alpha)) \ . \tag{53}$$

The required approximation is then

$$f(\alpha, t) = f_0(H_0(\alpha)) + \frac{\partial f_0}{\partial E}(H_0(\alpha)) \int_{-\infty}^t dt' \ X(t') \frac{dH_1}{dt}(\alpha_{t-t'}) \ . \tag{54}$$

We will use an alternative form, obtained by integration by parts

$$f(\alpha, t) = f_0(H_0(\alpha)) + X(t) \frac{\partial f_0}{\partial E}(H_0(\alpha)) H_1(\alpha)$$
$$- \frac{\partial f_0}{\partial E}(H_0(\alpha)) \int_{-\infty}^t dt' \ \dot{X}(t') \ H_1(\alpha_{t-t'}(\alpha)) \ . \tag{55}$$

We will assume that the integral converges. For ergodic systems this requires that the microcanonical average of $H_1(\alpha)$ vanishes. The density of available states in phase space is $(2\pi\hbar)^{-d}$, where d is the number of degrees of freedom. For a system of fermions, the appropriate density function is $f_0(\alpha, X) = \theta[E_F - H(\alpha, X)]/(2\pi\hbar)^d$, where $\theta(x)$ is the Fermi-Dirac distribution, which can be approximated by a downward step function when the temperature is small compared to the Fermi temperature.

Now the charge density of electrons is

$$\rho(\mathbf{r}, t) = e \int d\mathbf{p} \ f(\mathbf{r}, \mathbf{p}; t) \ . \tag{56}$$

The number density of electrons $N(E_F, \mathbf{r})$ and the density of states per unit volume at the Fermi surface $\nu(E_F, \mathbf{r})$ are

$$N(E_F, \mathbf{r}) = \frac{1}{(2\pi\hbar)^d} \int d\mathbf{p} \ \theta(H_0(\mathbf{r}, \mathbf{p}) - E_F) \ , \quad \nu(E_F, \mathbf{r}) = \frac{\partial}{\partial E_F} N(E_F, \mathbf{r}) \tag{57}$$

respectively. Also, the local average of any quantity $A(\mathbf{r}, \mathbf{p})$ for electrons at the Fermi surface is defined as

$$\langle A \rangle_{E_F, \mathbf{r}} = \int d\mathbf{p} \ A(\mathbf{r}, \mathbf{p}) \delta(H_0(\mathbf{r}, \mathbf{p}) - E_F) \Big/ \int d\mathbf{p} \ \delta(H_0(\mathbf{r}, \mathbf{p}) - E_F)$$
$$= \frac{1}{(2\pi\hbar)^d \nu(E_F, \mathbf{r})} \int d\mathbf{p} \ A(\mathbf{r}, \mathbf{p}) \delta(H_0(\mathbf{r}, \mathbf{p}) - E_F) \tag{58}$$

¿From (55) and the definition (56), we have:

11

$$
\begin{aligned}
\rho(\mathbf{r}, t) \sim\ & eN(E_\mathrm{F}, \mathbf{r}) + eX(t)\nu(E_\mathrm{F}, \mathbf{r})\phi(\mathbf{r}) \\
& - e\,\nu(E_\mathrm{F}, \mathbf{r}) \int_{-\infty}^{t} dt'\ \dot{X}(t') \int d\mathbf{r}' \int d\mathbf{p}\ \delta[E - H_0(\mathbf{r}, \mathbf{p})]\, \delta[\mathbf{r}' - \mathbf{r}_{t-t'}(\mathbf{r}, \mathbf{p})]\, \phi(\mathbf{r}') + O(X^2) \\
=\ & eN(E_\mathrm{F}, \mathbf{r}) + eX(t)\nu(E_\mathrm{F}, \mathbf{r})\phi(\mathbf{r}) \\
& - e\nu(E_\mathrm{F}, \mathbf{r}) \int_{-\infty}^{t} dt'\ \dot{X}(t') \int d\mathbf{r}'\ \langle \delta[\mathbf{r}_{t-t'}(\mathbf{r}, \mathbf{p}) - \mathbf{r}']\rangle_{E_\mathrm{F}, \mathbf{r}}\ \phi(\mathbf{r}_{t'}) + O(X^2)
\end{aligned}
\tag{59}
$$

With the definition of the propagator

$$
P(\mathbf{r}, \mathbf{r}'; t) = \theta(t)\, \langle \delta[\mathbf{r}' - \mathbf{r}_t(\mathbf{r}, \mathbf{p})]\rangle_{E_\mathrm{F}, \mathbf{r}}
\tag{60}
$$

and recalling the definition of the polarisation operator, (25), we find:

$$
\Pi(\mathbf{r}, \mathbf{r}', t - t') = e\,\theta(t - t')\nu(E_\mathrm{F}, \mathbf{r})\Big[\delta(\mathbf{r} - \mathbf{r}')\delta(t - t') + \partial_t P(\mathbf{r}, \mathbf{r}'; t - t')\Big]
\tag{61}
$$

or alternatively, in the frequency domain

$$
\Pi(\mathbf{r}, \mathbf{r}', \omega) = e\,\nu(E_\mathrm{F}, \mathbf{r})[\delta(\mathbf{r} - \mathbf{r}') + i\omega P(\mathbf{r}, \mathbf{r}'; \omega)]\ .
\tag{62}
$$

We will introduce an operator $\hat{\nu}$, which is diagonal in the position representation, so that $(\mathbf{r}|\,\hat{\nu}|\phi) = \nu(E_\mathrm{F}, \mathbf{r})\phi(\mathbf{r})$. Equation (62) may then be written in the form

$$
\widehat{\Pi}(\omega) = e\,\hat{\nu}\,[\widehat{I} + i\omega\widehat{P}(\omega)]\ .
\tag{63}
$$

There is also a relationship between the non-local conductivity $\widehat{\Sigma}(\omega)$ and the propagator $\hat{P}(\omega)$, which has previously been obtained by Serota and co-workers [10,11] (with an alternative derivation given in [8]). Their derivation was specific to the case of diffusive electron motion, whereas that given above also includes the ballistic case.

IV. PARTICULAR FORMS FOR THE POLARISABILITY

A. Spatially homogeneous, ballistic system

For a spatially homogeneous system, $\Pi(\mathbf{r}, \mathbf{r}'; \omega)$ is a function of $\mathbf{r} - \mathbf{r}'$, and is conveniently represented by its Fourier transform, $\Pi(\mathbf{q}, \omega)$: in d dimensions

$$
\widehat{\Pi}(\omega) = \frac{V}{(2\pi)^d} \int d\mathbf{q}\ |\chi_\mathbf{q})\, \Pi(\mathbf{q}, \omega)\, (\chi_\mathbf{q}|\ , \quad (\mathbf{r}|\chi_\mathbf{q}) = \frac{1}{\sqrt{V}}e^{i\mathbf{q}\cdot\mathbf{r}}
\tag{64}
$$

where V is the volume of the system. In the case where the electron motion is ballistic, the propagator is, for $d = 3$,

$$
\begin{aligned}
P(\mathbf{r}, \mathbf{r}'; t) &= \frac{1}{4\pi R^2}\delta(R - v_\mathrm{F}t)\ , \quad R = |\mathbf{r} - \mathbf{r}'|\ , \\
P(\mathbf{r}, \mathbf{r}'; \omega) &= \frac{1}{4\pi v_\mathrm{F} R^2}\, e^{i\omega R/v_\mathrm{F}}
\end{aligned}
\tag{65}
$$

and the Fourier transform representation of the polarisability is

$$\Pi(q,\omega) = \nu e \left(1 - \frac{1}{2\lambda} \log \left| \frac{\lambda + 1}{\lambda - 1} \right| + i\theta(\lambda - 1)\frac{\pi}{2\lambda} \right) , \quad \lambda = \frac{qv_{\mathrm{F}}}{\omega} , \tag{66}$$

which is the semiclassical limit of Eqs. (12.48a,b) in [9]. In two dimensions, $\Pi(q,\omega)$ is given by

$$\Pi(q,\omega) = e\nu \begin{cases} 1 + i(\lambda - 1)^{-1/2} & \text{for } \lambda > 1 \\[2mm] 1 - (1 - \lambda^2)^{-1/2} & \text{for } \lambda < 1 . \end{cases} \tag{67}$$

B. Low and high frequency limits

In the low frequency limit, it is immediately clear from (63) that the induced charge density is $\rho(\mathbf{r}) = \nu e[\phi(\mathbf{r}) - \langle \phi \rangle]$ where $\langle \phi \rangle$ is the space average of ϕ over the particle. We shall be only be concerned with cases where $\langle \phi \rangle$ vanishes, so that we may write

$$\Pi(\mathbf{r}, \mathbf{r}'; \omega) \sim \nu\, e\, \delta(\mathbf{r} - \mathbf{r}') , \quad \omega \ll \omega_{\mathrm{c}} . \tag{68}$$

For sufficiently high frequencies, and sufficiently far from the boundary of the particle, the conductivity is local, with value $\sigma(\omega)$:

$$\Sigma_{ij}(\mathbf{r}, \mathbf{r}'; \omega) = \delta_{ij}\, \delta(\mathbf{r} - \mathbf{r}')\, \sigma(\omega) \tag{69}$$

and the bulk conductivity $\sigma(\omega)$ may, in the case of diffusive electron motion, be approximated by the Drude formula

$$\sigma(\omega) = \frac{\nu e^2 D}{1 + i\omega\tau} , \quad \tau = \frac{m}{N}\frac{\partial N}{\partial E}D \tag{70}$$

where D is the diffusion constant, and m the electron effective mass. In the case of ballistic electron motion, the bulk conductivity is determined purely by the inertia of the electrons, and is non-dissipative:

$$\sigma(\omega) = \frac{Ne^2}{im\omega} . \tag{71}$$

When the non-local conductivity can be approximated by (69), the non-local polarisability takes the simple form

$$\Pi(\mathbf{r}, \mathbf{r}'; \omega) = \frac{i\sigma(\omega)}{\omega}\nabla_i\nabla_i'\delta(\mathbf{r} - \mathbf{r}')$$
$$= -\frac{i\sigma(\omega)}{\omega}\nabla^2\delta(\mathbf{r} - \mathbf{r}') . \tag{72}$$

This approximation is expected to be valid when $\omega \gg \omega_{\mathrm{c}}$, and when both $\mathbf{r}$ and $\mathbf{r}'$ are much greater than a distance Λ from the boundary: in the ballistic case $\Lambda = v_{\mathrm{F}}/\omega$, and in the diffusive case $\Lambda = \sqrt{D/\omega}$. The same conclusion can also be reached by considering the expressions (66), (68) in the limit $\lambda \to 0$: for $d = 3$ we find that $\Pi(q,\omega) \sim \frac{1}{3}\nu e\lambda^2 = \nu e v_{\mathrm{F}}^2 q^2/3\omega^2$, which is equivalent to the Fourier transform of (72) when the conductivity is given by (71).

C. Polarisability close to a boundary

Next we consider the polarisation charge close to the boundary of the particle. Here we are concerned with the high frequency case, $\omega \gg \omega_c$. In the low frequency case (68) gives an adequate approximation, but our discussion of the high frequency case assumed that the conductivity could be approximated as that of a homogeneous system. Another reason for discussing the boundary separately is that we expect that the polarisation charge density may have a singularity there.

We may assume that for $\omega \gg \omega_c$ the polarisability operator is short ranged. A smooth boundary may therefore be approximated locally by a flat surface, $z = 0$ in some local Cartesian coordinates. The polarisability is given by (53), and we approximate the propagator from $\mathbf{r}' = (x', y', z')$ to $\mathbf{r} = (x, y, z)$ by the sum of a direct contribution and a contribution originating from an image source at $\mathbf{r}'_{\mathrm{im}} = (x', y', -z')$, so that

$$\rho(\mathbf{r}) = \nu e \left[\phi(\mathbf{r}) + \mathrm{i}\omega \int d\mathbf{r}' \, [P(\mathbf{r}, \mathbf{r}'; \omega) + P(\mathbf{r}, \mathbf{r}'_{\mathrm{im}}; \omega)] \phi(\mathbf{r}') \right] . \tag{73}$$

The charge charge density is concentrated in a narrow layer at the surface, and may typically be approximated by writing

$$\rho(\mathbf{r}) = \rho_s(z) K(\mathbf{S}) \tag{74}$$

where $\mathbf{S}$ labels points on the boundary, and z is a coordinate normal to the boundary. In this case, the potential in the neighbourhood of the boundary is of the form $\phi(\mathbf{r}) = \phi_s(z) K(\mathbf{S})$, where $\phi_s(z)$ satisfies

$$\rho_s(z) = \nu e \left[\phi_s(z) + \frac{1}{\Lambda} \int_0^\infty dz' \, [G((z - z')/\Lambda)) + G((z + z')/\Lambda)] \phi_s(z') \right] \tag{75}$$

where $\Lambda = v_{\mathrm{F}}/\omega$, and the function $G(x)$ is easily related to the Fourier transform of $\Pi(q, \omega)$.

D. Diffusive electron motion

In the diffusive case, it is possible to write a useful eigenfunction expansion for the linear response functions: for $t > 0$ the propagator $P(\mathbf{r}, \mathbf{r}'; t)$ satisfies the diffusion equation $\partial_t P = D\nabla^2 P$, or $[\mathrm{i}\omega - D\nabla^2]P(\mathbf{r}, \mathbf{r}'; \omega) = -\delta(\mathbf{r} - \mathbf{r}')$, and satisfies the Neumann boundary condition. It can be expressed in terms of a set of eigenfunctions $\chi_n(\mathbf{r})$ of the Helmholtz equation $(\nabla^2 + k_n^2)\chi_n(\mathbf{r}) = 0$, satisfying the same boundary condition: $\hat{\mathbf{n}}.\nabla\chi_n = 0$, where $\hat{\mathbf{n}}$ is a normal vector on the boundary of the particle. The propagator can then be written

$$\widehat{P}(\omega) = \sum_n \frac{1}{\mathrm{i}\omega - Dk_n^2} |\chi_n)(\chi_n| . \tag{76}$$

Expansions for other linear response functions are easily obtained in the same form. For example, if the density of states per unit volume ν is independent of $\mathbf{r}$, (54) implies that the polarisability can be written in this form, with the coefficient of the operator $|\chi_n)(\chi_n|$ given by $\nu e D k_n^2/(Dk_n^2 - \mathrm{i}\omega)$.

V. THE SELF-CONSISTENT FIELD

A. Approximate equations for the self-consistent fields

Here we discuss how the solution of the self-consistent equations can be greatly simplified by the use of 'semiclassical' approximations. We consider the electric dipole absorption first.

Calculation of the electric dipole absorption coefficient via either (39) or (47) requires the self-consistent fields $\phi_{\text{eff}}(\mathbf{r}, \omega)$, which is given by equation (32):

$$|\phi_{\text{ext}}) = [\hat{I} - \hat{U}\hat{\Pi}(\omega)]|\phi_{\text{eff}}) \ . \tag{77}$$

Formally, solution of this equation requires calculation of the inverse of $\hat{I} - \hat{U}\hat{\Pi}(\omega)$: this could be done explicitly in a numerical calculation by expanding in a suitable basis set. We will aim instead for an approximate analytic solution. For frequencies small compared to the plasma frequency ω_{p}, the external electric field is 'screened' by polarisation charges, so that the internal field is much smaller than the externally applied field. The key physical intuition is that the external electric field is almost exactly cancelled by the electric field due to the induced charge density $\rho(\mathbf{r})$. Let $\rho_{\text{cl}}(\mathbf{r})$ be the charge density induced on the particle by a static external field, according to classical electrodynamics: this charge density gives an induced electric field which precisely cancels the externally applied field inside the particle. For frequencies small compared to the plasma frequency, the induced charge density is well approximated by $\rho_{\text{cl}}(\mathbf{r})$: we will assume that

$$|\rho) = |\rho_{\text{cl}}) + O(\omega/\omega_{\text{p}}) + O(a/a_0) \tag{78}$$

where a is the characteristic dimension of the particle, and a_0 is the Bohr radius. The classical charge distribution formally satisfies an equation analogous to (77), in which the term representing the internal field $|\phi_{\text{eff}})$ is set equal to zero:

$$|\phi_{\text{ext}}) + \hat{U}|\rho_{\text{cl}}) = 0 \ . \tag{79}$$

We will denote our approximation to the effective potential $|\phi_{\text{eff}})$ by $|\phi)$: it is the potential which generates the polarisation charge $|\rho_{\text{cl}})$, and is given by

$$|\rho_{\text{cl}}) = \hat{\Pi}(\omega)|\phi) \tag{80}$$

or equivalently by $|\phi_{\text{ext}}) + \hat{U}\hat{\Pi}(\omega)|\phi) = 0$. Comparing with (77), it is clear that this solution $|\phi)$ is a good approximation to $|\phi_{\text{eff}})$ provided $||\hat{U}\hat{\Pi}(\omega)|| \gg 1$, where $||\hat{X}||$ is an appropriate norm of the operator $\hat{X}$. To estimate this norm, we consider the effect of an arbitrary potential ϕ: at zero frequency, the induced charge density is $\hat{\Pi}\phi = e\nu\phi$, and for a particle of characteristic dimension a in d dimensions, the induced charge may be approximated by a dipole formed by charges of magnitude $Q \sim \rho a^d$, with separation a: this results in an electrical potential of magnitude $\phi' \sim eQ/(\epsilon_0 a)$. In three dimensions this leads to the following estimate for $||\hat{U}\hat{\Pi}|| \sim \phi'/\phi$

$$||\hat{U}\hat{\Pi}|| \sim \frac{\omega_{\text{p}}^2}{\omega^2} \tag{81}$$

where ω_p is the three dimensional bulk plasma frequency, $\omega_\mathrm{p} = [Ne^2/(4\pi\epsilon_0 m)]^{1/2}$, N being the electron density. For frequencies $\omega \gg \omega_\mathrm{c}$, a similar argument gives

$$||\hat{U}\hat{\Pi}(\omega \gg \omega_\mathrm{c})|| \sim \frac{\omega_\mathrm{p}^2}{\omega\omega_\mathrm{c}} \ . \tag{82}$$

In the case of ballistic dynamics $\omega_\mathrm{c} = v_\mathrm{F}/a$, equations (81) and (82) are of the order of $\sigma_0/(\epsilon_0\omega_\mathrm{c})$ and $\sigma_0/(\epsilon_0\omega)$ respectively.

We can, in principle, determine improved approximations to the exact solution of (78) from the solution of (80):

$$\begin{aligned}|\phi_\mathrm{eff}) &= -[\hat{I} - \hat{U}\hat{\Pi}(\omega)]^{-1}\hat{U}\hat{\Pi}(\omega)\,|\phi) \\ &= |\phi) - [\hat{I} - \hat{U}\hat{\Pi}(\omega)]^{-1}\,|\phi) \ . \end{aligned} \tag{83}$$

Equations (81) and (82) show that the correction term in (83) is small. Having shown that $|\phi_\mathrm{eff}) \sim |\phi)$, we discuss how to estimate the solution $|\phi)$ of (80) in Secs. VB and VC.

Finally we consider the semiclassical solution for the magnetically induced field, $\mathbf{E}_\mathrm{ind}$ which satisfies (31):

$$\nabla.[\hat{I} - \mathrm{i}\omega\epsilon_0\widehat{\Sigma}(\omega)]\,|\mathbf{E}_\mathrm{ind}) = 0 \ . \tag{84}$$

At low frequencies we can estimate the conductivity by $\Sigma \sim Ne^2/m\omega_\mathrm{s}$ where ω_s is the scattering frequency; at high frequencies ω is replaced by the frequency ω. In the low frequency limit we therefore estimate

$$\frac{1}{\epsilon_0\omega}||\widehat{\Sigma}(\omega)|| \sim \frac{\omega_\mathrm{p}^2}{\omega\omega_\mathrm{s}} \ . \tag{85}$$

In the frequency range that we are concerned with, the term involving the identity operator in (84) is therefore negligible; the same conclusion applies when $\omega \gg \omega_\mathrm{s}$. We can therefore find an approximate solution to (84) by requiring that the induced charge density is zero, i.e.

$$\nabla.\widehat{\Sigma}(\omega)|\mathbf{E}_\mathrm{ind}) = 0 \ . \tag{86}$$

This justifies the neglect of a cross-term in (3) with an electric dipole induced by the magnetic field. A solution to (86) may be determined by choosing an initial approximation $\mathbf{E}'_\mathrm{ind}$ which satisfies $\nabla\wedge\mathbf{E}'_\mathrm{ind} = \mathrm{i}\omega\mathbf{B}_\mathrm{ext}$. A polarisation charge ρ'_ind would be generated from this field. An additional field which is the gradient of a potential $\chi(\mathbf{r})$ is added, such that $\mathbf{E}_\mathrm{ind} = \mathbf{E}'_\mathrm{ind}+\nabla\chi$. The condition upon χ for (86) to be satisfied is

$$|\rho'_\mathrm{ind}) = \widehat{\Pi}(\omega)\,|\chi) \ . \tag{87}$$

This equation is analogous to (80).

B. Solutions in the ballistic case

We will discuss approximate solutions of (80) valid in the limits $\omega \ll \omega_c$ and $\omega \gg \omega_c$. The first of these represents the static potential required to hold the classical charge distribution in place in the zero-frequency limit: it will be written $|\phi_{\text{stat}})$, and its form is immediately apparent from (63):

$$|\phi_{\text{stat}}) = \frac{1}{e\nu}|\rho_{\text{cl}}) . \tag{88}$$

This is simply a linearised Thomas-Fermi approximation [12]. The semiclassical approximations underlying this expression assume that the potentials are slowly varying on the scale of the Fermi wavelength. This is questionable when the charge density has a singularity on the surface of the particle: we will return to this later.

In the limit $\omega \gg \omega_c$, we found [equation (72)] that the polarisability may be approximated by $\hat{\Pi} \sim -(i\sigma(\omega)/\omega)\nabla_{\mathbf{r}}^2\delta(\mathbf{r} - \mathbf{r}')$, for $\mathbf{r}$ and $\mathbf{r}'$ not too close to the boundary. For points not too close to the boundary, or where the charge density is non-analytic, we can approximate the solution of (80) by a 'dynamic' potential, which is of the form $|\phi_{\text{dyn}}) = \lambda|\psi)$, where $\nabla^2\psi(\mathbf{r}) = \rho_{\text{cl}}(\mathbf{r})$, and $\hat{n}.\nabla\psi(\mathbf{r}) = 0$. Substituting these forms into (80) we find that $\lambda = -i\omega/\sigma(\omega)$:

$$|\phi_{\text{dyn}}) = -\frac{i\omega}{\sigma(\omega)}|\psi) , \quad \nabla^2|\psi) = |\rho_{\text{cl}}) . \tag{89}$$

An interpretation of the dynamic potential is that it moves the polarisation charge into place.

Close to the boundary (89) is not necessarily a good approximation to the effective potential. One reason is that the approximations underlying (72) fail, and the polarisation must be described by (73) or (75). Another reason is that the charge density $|\rho_{\text{cl}})$ has a singularity there. In the notation of (74), the projected charge density $\rho_s(z)$ is concentrated on the boundary in the three dimensional case [4] so that $\rho_s(z) \sim \delta(z)$, and in the two dimensional case it diverges on the boundary, such that $\rho_s(z) \sim z^{-1/2}$ for $z > 0$ [6]. The form of (75) indicates that the potential $\phi_s(z)$ also has the same type of singularity as the charge density at the boundary.

To summarise, the following picture emerges. For low frequencies, $\omega \ll \omega_c$ the potential is approximately $\phi_{\text{stat}}(\mathbf{r})$. At high frequencies, $\omega \gg \omega_c$, the potential is well approximated by $\phi_{\text{dyn}}(\mathbf{r})$ within the interior of the particle. In the vicinity of the boundary, the potential has a dominant divergent contribution, which is well approximated by $\phi_{\text{stat}}(\mathbf{r})$.

C. Diffusive case

In the diffusive case, equation (80) can be solved exactly, using the representation of the propagator in the form (76). Expanding the potential $|\phi)$ in terms of the functions $|\chi_n)$ leads to the expression

$$|\phi) = \frac{1}{e\nu}\sum_n(\chi_n|\rho_{\text{cl}})\Big[1 - \frac{i\omega}{Dk_n^2}\Big]|\chi_n) = |\phi_{\text{stat}}) + |\phi_{\text{dyn}}) \tag{90}$$

Note that in the diffusive case the potential is precisely equal to the sum of the static and dynamic contributions [7].

VI. DISCUSSION: CALCULATION OF THE ABSORPTION COEFFICIENT

Once an adequate approximation for the effective potential has been obtained, the electric absorption coefficient is obtained from (47): the absorption coefficient is proportional to $\omega^2(\phi|\hat{P}(\omega)|\phi)$. Previous papers [4-8] have discussed methods for the evaluation of the absorption coefficient using equation (47) in some detail, for specific cases. This paper has presented a general approach to the determining of the effective potential, and some remarks on applying this to calculating the absorption coefficient may be useful.

When the electron motion is diffusive, the absorption is very easily evaluated using (90) and (76). It is found that the coefficient is proportional to ω^2, and that (at least within the framework of the approximations used in section V) the frequency scale ω_c plays no role. The absorption coefficient can be shown to be exactly equal to the classical value in this case [7,5].

The case of ballistic electron motion is more difficult. It might be expected that $C_{\phi\phi}(\omega) = (\phi|\hat{P}(\omega)|\phi)$ approaches a non-zero limit as $\omega \to 0$, implying that the absorption coefficient is proportional to ω^2 for low frequencies. This expectation is correct for cases where the electron motion is ergodic (the most important cases being diffusive electron motion, and the ballistic case with a rough boundary). In the case of integrable electron motion, which can be realised experimentally if the boundary appears smooth on the scale of the Fermi wavelength, $C_{\phi\phi}(\omega)$ typically approaches zero as $\omega \to 0$ in a non-analytic fashion. In the important special case of particles with circular symmetry, $C_{\phi\phi}(\omega)$ is zero for $\omega < \omega_c$, where ω_c is the frequency of a glancing circular orbit. Thus the electron dynamics plays an important role in determining the low frequency absorption.

The high frequency absorption, by contrast, is determined by the nature of the singularities of the function $f(t) = \phi(\mathbf{r_t})$, which can result from singularities in the motion $\mathbf{r}(t)$, or in the potential $\phi(\mathbf{r})$. We will discuss the ballistic case. The dominant contribution comes from the singularities of $\phi(\mathbf{r})$ in the neighbourhood of the boundary. The absorption coefficient was calculated in [4] and [6] for the three dimensional and two dimensional cases respectively, assuming that the effective potential is equal to $\phi_{\text{stat}}(\mathbf{r})$. In three dimensions, this potential is a delta function singularity concentrated on the boundary, and in two dimensions it diverges as $z^{-1/2}$, where z is the normal distance from the boundary. These forms for the potential imply that $(\phi|\hat{P}(\omega)|\phi) \sim \omega^0$ in three dimensions and $\sim \omega^{-1}$ in two dimensions. In [4] and [6] it was predicted that in the case of particles with circular symmetry, the absorption coefficient shows a sequence of resonances superposed on a regular contribution increasing as ω^2 and ω for three and two dimensions respectively.

The more sophisticated approach introduced in this paper indicates that the potential $|\phi)$ should satisfy (80), whereas the potential used in [4], [6] was simply the static potential, satisfying $|\rho_{\text{cl}}) = \hat{\Pi}(0)|\phi_{\text{stat}})$. We must discuss the extent to which this refinement will change the results. In section V B we argued that the singularities of the effective potential at the boundary are the same as those of the 'static' potential. We can therefore hypothesise that the more refined theory would make a quantitative rather than qualitative difference to the results. This issue will be addressed in a subsequent paper, which will consider the inversion of (75) to determine the potential $\phi_s(z)$ from the charge density $\rho_s(z)$, and its use to estimate the high frequency absorption coefficient.

ACKNOWLEDGMENTS

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