Magnetic dipole absorption of radiation in small conducting particles

Magnetic dipole absorption of radiation in small conducting particles

Michael Wilkinson†§, Bernhard Mehlig†‖ and Paul N Walker‡

† Isaac Newton Institute for Mathematical Sciences, University of Cambridge, 20, Clarkson Road, Cambridge CB3 0EH, UK
‡ Laboratoire de Physique des Solides, associé au CNRS, Université Paris-Sud, 91405 Orsay, France

Abstract. We give a theoretical treatment of magnetic dipole absorption of electromagnetic radiation in small conducting particles, at photon energies which are large compared to the single-particle level spacing, and small compared to the plasma frequency. We discuss both diffusive and ballistic electron dynamics for particles of arbitrary shape.

The conductivity becomes non-local when the frequency is smaller than the frequency ω_c characterizing the transit of electrons from one side of the particle to the other, but in the diffusive case ω_c plays no role in determining the absorption coefficient. In the ballistic case, the absorption coefficient is proportional to ω^2 for $\omega \ll \omega_c$, but is a decreasing function of ω for $\omega \gg \omega_c$.

1. Introduction

The interaction of electromagnetic radiation with small conducting particles has attracted much interest, both from experimental and theoretical perspectives. Historically, experimental work in this field has been frustrated by the difficulties associated with obtaining uniform and precisely characterized metal grains by techniques such as abrasion or precipitation. References [1–3] are general reviews of this field. Recently there has been a revival of interest in this topic, caused by new developments in both experimental techniques and theoretical perspectives. The problems of controlling the size, composition, and microscopic structure of the particles can be overcome using lithographic technologies from the field of mesoscopic physics, reviewed in [4]. Theoretical interest in this problem has been stimulated by wishing to understand the response of 'generic', 'chaotic' quantum systems to perturbations, by extending the random-matrix models for spectra: this approach was initiated in a seminal paper by Gorkov and Eliashberg [5], and more recent work on applications of random-matrix methods is reviewed in [4].

For sufficiently low frequency ω, classical electromagnetic theory suggests that the absorption scales as ω^2, and therefore dominates scattering, which scales as ω^4. The classical theory of the interaction of electromagnetic radiation with spherical particles of uniform composition was considered by Mie [6]; the theory encompasses conducting particles with

§ Permanent address: Department of Physics and Applied Physics, John Anderson Building, University of Strathclyde, Glasgow G4 0NG, UK.
‖ Permanent address: Max-Planck-Institut für Physik komplexer Systeme, Nöthnitzer Strasse 38, 01187 Dresden, Germany.

a complex dielectric constant, which is often modelled by the Drude theory [7]. If the particles are small compared to both the wavelength and the electromagnetic skin depth of the radiation, the dominant contributions to the absorption are called the electric and magnetic dipole terms [8]. In both cases the absorption is due to Joule or Ohmic heating caused by electrical currents flowing through the particle: the electric dipole term is due to currents which establish electrical polarization of the particle, and the magnetic dipole term is due to eddy currents induced by variation of the magnetic field.

At frequencies below the plasma frequency, the electric field is screened from the interior of the particle, but the magnetic field can penetrate the whole of the particle. Although magnetic effects are negligible in atomic absorption processes, they could become significant when the number of atoms in the particle is sufficiently large that most of the atoms are screened from the electric field. In fact, magnetic dipole absorption is often the dominant absorption process in suspensions of small metal particles [9]. Very few of the many theoretical papers on absorption of radiation by small particles have considered the magnetic dipole contribution; some exceptions are [1, 10] which consider magnetic dipole absorption in the context of effective-medium theories. Because it is typically the dominant contribution, it is appropriate to consider the problem of magnetic dipole absorption in some detail.

The Mie theory is restricted to spherical particles in which the dynamics of the charge carriers is diffusive, and quantum mechanical effects are ignored. In this paper we will give the first theoretical treatment of magnetic dipole absorption going beyond the Mie theory. The conducting particle is modelled as a container or arbitrary shape, which confines a gas of non-interacting fermions (which we term electrons): this is the standard model for metallic systems with high densities of charge carriers. The motion of the electrons within the boundary of the container may be either free, which we refer to as the ballistic case, or it may be impeded by elastic scatterers, which result in the electron trajectories within the particle being diffusive: these model the limiting cases of very pure and very disordered conductors. Our approach allows for arbitrary particle geometries, and we give careful consideration to the fact that the conductivity is non-local when the particles are very small: quantum mechanical effects are included using a semiclassical approach. The paper complements [11–13], which gave a comparably comprehensive treatment of electric dipole absorption.

We do not explicitly consider the structures in the absorption close to the single-particle level spacing which were originally considered by Gorkov and Eliashberg [5]. These structures are determined by repulsion between energy levels and the appropriate tool to analyse them is random-matrix theory. Their full characterization requires an estimate of a mean square matrix element, which was not given correctly in [5]. Our results for the low-frequency limit provide the correct estimate of this quantity for magnetic dipole absorption.

Sections 2 to 4 will be concerned with various aspects of the formulation of the problem, discussing respectively the relation between the absorption of radiation and correlation functions of the electron motion, the criteria for a self-consistent solution of the equations determining the electric field driving the eddy currents, and the definition and semiclassical estimation of the non-local conductivity which is required to determine the self-consistent field. Our semiclassical estimates for the non-local conductivity are closely related to expressions given by Argaman [14]. In section 5 we develop the theory for magnetic dipole absorption in particles with diffusive electron motion, assuming that the electric field is known. This calculation also yields the form of the non-local conductivity applicable to the diffusive case: in section 6 we consider the solution for the self-consistent field, and discuss results for some specific geometries. Our formula for the non-local conductivity is identical

to one given by Serota and co-workers [15, 16], who used diagrammatic techniques. Our derivation is more direct and requires fewer assumptions: we discuss this point further in section 6.

Our approach uses a semiclassical estimate described in [17], which relates mean squared matrix elements to classical correlation functions. We might expect that there should be features in the absorption spectrum which are related to the characteristic timescale for decay of classical correlations, in this case the typical time for a particle to cross the particle; the importance of this timescale was emphasized by Thouless [18], and in the case of diffusive electron motion, we will refer to the characteristic frequency scale $\omega_c = D/a^2$ as the Thouless frequency (D is the diffusion constant and a is the characteristic size of the particle). Another reason for expecting ω_c to play a role in determining the absorption coefficient is that the conductivity is non-local when ω is not large compared to ω_c. We find however that ω_c plays no role in the final expression for the absorption coefficient.

We consider the case of ballistic electron motion in section 7. We are only able to gain limited information about this case: we find that the absorption coefficient is proportional to ω^2 at frequencies small compared to $\omega_c = v_F/a$, and that it is a decreasing function of frequency for $\omega \gg \omega_c$.

Our conclusions are in many ways parallel to those for electric dipole absorption. In ballistic systems, it was found [11, 12] that the electric dipole absorption has resonances in the absorption coefficient with a frequency scale $\omega_c = a/v_F$, where v_F is the velocity at the Fermi energy. By contrast, in the case of diffusive electron motion, it was found [13] that there is no structure in the absorption coefficient at the frequency scale $\omega_c = D/a^2$.

Finally, we remark that there is a large literature concerned with the effects of time-dependent magnetic fluxes on metallic loops: when the magnetic flux varies sinusoidally, the absorption of energy by the loop is a special case of the magnetic dipole absorption which we consider here. Most of the papers on this topic are concerned with quantum-size effects analogous to those considered by Gorkov and Eliashberg [5]; two recent studies in this area are [16] and [19, 20].

2. Formulation of the problem

The absorption of radiation is usually described by an extinction coefficient $\gamma(\omega)$, which is defined as the fractional loss of intensity per unit length of sample, divided by the volume fraction F occupied by the particles. We will express our results in terms of the rate of absorption of energy $\langle dE/dt \rangle$ within a single particle. If the amplitudes of the electric and magnetic fields are E_0 are B_0 respectively, the intensity of the radiation is $I = \frac{1}{2}\epsilon_0 E_0^2 = B_0^2/2\mu_0$, and the relationship between γ and $\langle dE/dt \rangle$ is therefore

$$\gamma = \frac{2}{V\epsilon_0 c E_0^2}\left\langle \frac{dE}{dt} \right\rangle \tag{2.1}$$

where V is the volume of a single particle. In this paper we will define the absorption coefficient $\alpha(\omega)$ as the rate of absorption of energy for a single particle, divided by the electric field intensity:

$$\alpha(\omega) = \frac{1}{E_0^2}\left\langle \frac{dE}{dt} \right\rangle = \frac{1}{c^2 B_0^2}\left\langle \frac{dE}{dt} \right\rangle. \tag{2.2}$$

The normalization with respect to electric (rather than magnetic) field intensity is used to facilitate comparison with the results in [11–13].

The particle will be considered to consist of a static potential well which traps a gas of non-interacting fermions (electrons), initially with occupation probability $f(E)$ (which would be identified with the Fermi–Dirac distribution). In a quantum mechanical calculation the rate of absorption of energy is determined by the Fermi golden rule. This states that the rate of transition under the action of a periodic perturbation with frequency ω and matrix elements ΔH_{nm}, from a state with energy E_n, to a quasi-continuum of final states with energies close to $E_m = E_n + \hbar\omega$ is

$$R = \frac{\pi}{2\hbar} g(E_m) \langle |\Delta H_{nm}|^2 \rangle_\omega \tag{2.3}$$

where $g(E_m)$ is the density of final states and $\langle |\Delta H_{nm}|^2 \rangle_\omega$ is the mean square matrix element for transitions from E_n to states close to E_m. The energy absorbed by an electron making an upward transition is $\hbar\omega$. The rate of absorption of energy is therefore

$$\left\langle \frac{\mathrm{d}E}{\mathrm{d}t} \right\rangle = \hbar\omega \int \mathrm{d}E \ g(E) R(E)[f(E) - f(E - \hbar\omega)] \sim Rg\hbar^2\omega^2 \tag{2.4}$$

where the approximate equality is applicable in the limit where $\hbar\omega$ and kT are both large compared to the mean level spacing, but small compared to other energy scales: in the right-hand expression both g and R are evaluated at the Fermi energy.

The mean square matrix element can be estimated semiclassically [14]:

$$\langle |\Delta H_{nm}|^2 \rangle_\omega = \frac{1}{2\pi\hbar g} \int_{-\infty}^{\infty} \mathrm{d}t \ \exp(\mathrm{i}\omega t) \langle \Delta H(t) \, \Delta H(0) \rangle_E \tag{2.5}$$

where $\langle \Delta H(t) \, \Delta H(0) \rangle_E$ is the microcanonical autocorrelation function of the classical observable corresponding to $\Delta \hat{H}$, evaluated at energy E. Combining this result with (2.4), the absorption coefficient can be expressed in terms of the classical autocorrelation function of the perturbation $\Delta H(\boldsymbol{r}, \boldsymbol{p})$. The resulting expression can also be written in terms of the classical change in energy of the individual electrons due to the perturbation: the change in energy of an electron following a trajectory $\boldsymbol{r}(t), \boldsymbol{p}(t)$ is

$$\Delta E(t) = \int_0^t \mathrm{d}t' \ \frac{\partial H}{\partial t'}(\boldsymbol{r}(t'), \boldsymbol{p}(t')). \tag{2.6}$$

Combining this result with (2.5), the rate of absorption of energy by the electron gas is

$$\left\langle \frac{\mathrm{d}E}{\mathrm{d}t} \right\rangle = \frac{g}{2} \frac{\mathrm{d}}{\mathrm{d}t} \left\langle \Delta E(t)^2 \right\rangle \tag{2.7}$$

where $\langle \Delta E(t)^2 \rangle$ is the variance of the change in the single-electron energies.

In our problem the perturbation is a sinusoidally varying electromagnetic field, specified by a vector potential $\boldsymbol{A}(\boldsymbol{r}) \exp(\mathrm{i}\omega t)$, and a scalar potential $\Phi(\boldsymbol{r}) \exp(\mathrm{i}\omega t)$. The component of the perturbation of the Hamiltonian which is quadratic in $\boldsymbol{A}$ can be neglected when calculating the leading-order absorption coefficient; the remaining terms are

$$\Delta \hat{H} = \frac{e}{2m_e} (\hat{\boldsymbol{p}} \cdot \boldsymbol{A} + \boldsymbol{A} \cdot \hat{\boldsymbol{p}}) + e\Phi. \tag{2.8}$$

In discussing the magnetic dipole absorption, it is given that the magnetic field is

$$\boldsymbol{B}(t) = \boldsymbol{\nabla} \times \boldsymbol{A} = \boldsymbol{B}_0 \exp[\mathrm{i}\omega t]. \tag{2.9}$$

The fluctuating magnetic field induces an electric field $\boldsymbol{E}$, which is given by

$$\boldsymbol{E} = -\frac{\partial \boldsymbol{A}}{\partial t} + \boldsymbol{\nabla}\Phi. \tag{2.10}$$

The Hamiltonian admits a set of gauge transformations $(A, \Phi) \rightarrow (A', \Phi') = (A, \Phi) + (\nabla \mu, \partial_t \mu)$ which leave the electric and magnetic fields unchanged. We will assume that the gauge has been chosen so that $\Phi = 0$. Physically, the electric field is not uniquely defined by the magnetic field, and must be determined by a self-consistent condition, which we discuss in section 3.

3. Self-consistent choice of the vector potential

3.1. Self-consistent electric field

The eddy currents induced by the fluctuating magnetic will themselves generate a magnetic field. We will consider only the case of very small particles, for which this additional magnetic field is negligible: this applies when the dimensions of the particle are small compared to the electromagnetic skin depth. This assumption is compatible with the use of semiclassical approximations, which require that the particle be large compared to the Fermi wavelength. We therefore assume that the magnetic field is simply the externally applied field, $B(t) = B_0 \exp[i\omega t] e_3$. The electric field is required to uniquely determine the perturbation of the Hamiltonian. In this section we will discuss the self-consistent calculation of the electric field.

In discussions of the Zeeman effect in atomic physics, the perturbation of the Hamiltonian representing the magnetic field is conventionally taken to be proportional to the component of the angular momentum operator along the direction of the field. It is natural to ask why a more involved procedure is used here, but is unnecessary for the Zeeman problem or for calculation of static magnetic susceptibility. At the end of this section we show that the angular momentum operator gives the correct answer in the limit where the frequency approaches zero, but not in general.

The electric field satisfies the Maxwell equations

$$\nabla \times E = \frac{\partial B}{\partial t} \qquad \nabla \cdot E = \frac{\rho}{\epsilon_0}. \tag{3.1}$$

The electric field causes a current density j to flow within the particle. We will assume that a linear response theory is valid, but in general the current may be a non-local function of the electric field: we will write

$$j(r, t) = \int dr' \int_{-\infty}^{t} dt' \, \tilde{\sigma}(r, r'; t - t') E(r', t') \tag{3.2}$$

where $\tilde{\sigma}$ is the non-local conductance tensor. We will also write this relation in the form

$$j(r, \omega) = \int dr' \, \tilde{\sigma}(r, r'; \omega) E(r', \omega) \equiv \hat{\sigma} E(r, \omega) \tag{3.3}$$

where the second equality defines an operator $\hat{\sigma}$ which maps the electric field $E(r, \omega)$ non-locally into the current field $j(r, \omega)$. For a monochromatic perturbation, the charge density is

$$\rho = -\frac{i}{\omega} \nabla \cdot j. \tag{3.4}$$

Combining these results, we find the following equation for the electric field:

$$\nabla \cdot j' = 0 \qquad j' = (\hat{\sigma} - i\omega\epsilon_0)E. \tag{3.5}$$

This equation must be supplemented by a boundary condition in order to uniquely determine the electric field. This is

$$\hat{n} \cdot j = 0 \tag{3.6}$$

where $\hat{n}$ is a unit vector normal to the boundary: this condition represents the fact that charges cannot enter or leave the sample.

3.2. Representation of the field in terms of potentials

It will be convenient to write the electric field in terms of a vector and a scalar potential:

$$E = \nabla \times \psi + \nabla\phi. \tag{3.7}$$

We will only consider in detail cases where the field $\psi(r)$ is of the form

$$\psi = \psi(x, y)e_3. \tag{3.8}$$

This form is appropriate when the conducting particle is two dimensional, lying in the plane $z = 0$, for three-dimensional particles in the form of general cylinders aligned with the z-axis, and can be extended to spheres and some other non-cylindrical geometries. Substituting (3.7), (3.8) into the Maxwell equations, we find that ψ satisfies Poisson's equation in the form

$$\nabla^2 \psi = i\omega B_0. \tag{3.9}$$

We will always choose $\psi(x, y)$ to satisfy the condition that $\psi = 0$ on the boundary. Having uniquely specified $\psi(x, y)$, equations (3.5) and (3.6) are transformed into equations determining the scalar potential ϕ.

In some cases a local, isotropic conductivity $\Sigma(\omega)$ will provide an adequate description. In this case, the condition (3.5) reduces to the requirement that $\nabla \cdot E = 0$, in which case the electric field can be written in the form

$$E = \nabla \times \psi + \nabla\phi \qquad \nabla^2\phi = 0. \tag{3.10}$$

The boundary condition corresponding to (3.6) is then satisfied by taking a solution for which $\psi = 0$ and $\phi = 0$ on the boundary: the latter condition implies that $\phi = 0$ everywhere.

3.3. A remark on the low-frequency limit

After having described the approach used to define the correct perturbation, we will now show that any form of the electric field which has a uniform value of $\nabla \times E$ gives the correct value of the absorption coefficient in the limit $\omega \to 0$. According to (2.7), the absorption coefficient is proportional to the variance of the change in the single-particle energy. The change of the single-particle energy can be written as

$$\Delta E(t) = \int_{t_0}^{t} \mathrm{d}t' \, \frac{\partial H}{\partial t'}(r(t'), p(t')) = i\omega \int_{t_0}^{t} \mathrm{d}t' \, \frac{\mathrm{d}r}{\mathrm{d}t'} \cdot A(r(t')) \exp[i\omega t']. \tag{3.11}$$

Consider the effect of making a transformation $A \to A' = A + \nabla\varphi$ on the absorption coefficient. The change in the single-particle energy is transformed to $\Delta E'$:

$$\Delta E' = \Delta E + i\omega \int_{t_0}^{t} \mathrm{d}t' \, \exp[i\omega t'] \frac{\mathrm{d}r}{\mathrm{d}t'} \cdot \nabla\varphi$$

$$= \Delta E + i\omega \int_{t_0}^{t} \mathrm{d}\varphi[r(t')] \, \exp[i\omega t'] \equiv \Delta E + i\omega X(t, \omega). \tag{3.12}$$

If $\omega = 0$, the correction $X(t)$ introduced by the transformation is simply

$$X(t, 0) = \int_{t_0}^{t} \mathrm{d}t' \, \frac{\mathrm{d}r}{\mathrm{d}t} \cdot \nabla\varphi = \varphi[r(t)] - \varphi[r(t_0)] \tag{3.13}$$

which remains bounded as $t \to \infty$. For finite ω the correction $X(t, \omega)$ satisfies

$$\langle X^2(t, \omega) \rangle = t \int_{-\infty}^{\infty} d\tau \, \exp(i\omega\tau) \left\langle \frac{d\varphi}{dt}(\tau) \frac{d\varphi}{dt}(0) \right\rangle + O(1) \equiv \Gamma(\omega)t + O(1) \tag{3.14}$$

provided that the correlation function of $d\varphi/dt$ decays faster than τ^{-1}. Comparison with (3.13) shows that the coefficient $\Gamma(\omega)$ of the secular term approaches zero as $\omega \to 0$, implying that the gauge-dependent contribution to the absorption coefficient vanishes in the limit $\omega \to 0$.

4. Semiclassical theory for non-local conductivity

4.1. General formula

We will use a semiclassical analysis for the non-local conductivity $\tilde{\sigma}(r, r', \omega)$. We will first consider the problem in rather abstract terms: we will discuss a Hamiltonian $H(r, p, X)$, where X is a time-dependent parameter. The Hamiltonian determines the motion of particles in a gas with phase-space density $\rho(r, p, t)$. The phase-space density satisfies the Liouville equation $\partial_t \rho = \{\rho, H\}$. A solution can be written in the form

$$\rho(r, p; t) = f(H(r, p; X) - E_F)$$
$$- \dot{X} \int_{-\infty}^{t} dt' \, \frac{\partial H}{\partial X}(r(t'), p(t'); X) \frac{\partial f}{\partial E}(H(r, p; X) - E_F) + O(\dot{X}^2). \tag{4.1}$$

Formally, this is an expansion in the velocity of the perturbation, $\dot{X}$: the results will be valid for all frequencies, because the amplitude of the perturbation is infinitesimal.

The leading-order Weyl or Thomas–Fermi estimate of the density of states of a quantum system states that the density of quantum states is $(2\pi\hbar)^{-d}$ in classically accessible regions of phase space. We will therefore multiply the above solution by this factor, and take $f(E)$ to be the Fermi–Dirac function.

Our Hamiltonian, $H = (p - eA)^2/2m_e + V$ has a time-dependent vector potential, so we can write

$$\dot{X}\frac{\partial H}{\partial X} = \frac{\partial H}{\partial A} \cdot E = \frac{e}{m_e}p \cdot E. \tag{4.2}$$

The resulting current is

$$j(r, t) = \frac{e}{m_e} \int dp \, p\rho(r, p, t). \tag{4.3}$$

The current flowing in response to the electric field $E(r, t)$ is therefore

$$j_i(r, t) = \frac{e^2}{(2\pi\hbar)^d m_e^2} \sum_j \int_{-\infty}^{t} dt' \int dp \, p_i \frac{\partial f}{\partial E} P_j(r, p; t' - t) E_j(R(r, p; t' - t))$$

$$= \frac{e^2}{(2\pi\hbar)^d m_e^2} \sum_j \int_{-\infty}^{t} dt' \int dp \int dr' \, \frac{\partial f}{\partial E} p_i$$

$$\times P_j(r, p; t' - t)\delta[r' - R(r, p; t - t')]E_j(r', t') \tag{4.4}$$

where $P_i(r, p; \tau)$, $R_i(r, p, \tau)$ are the ith components of the momentum and position at time τ for a trajectory which starts at (r, p) at time $t = 0$. The components of the non-local conductivity tensor are therefore

$$\sigma_{ij}(r, r'; t) = \frac{e^2}{(2\pi\hbar)^d m_e^2}\theta(t) \int dp \, \frac{\partial f}{\partial E} p_i P_j(r, p, t)\delta[r' - R(r, p; t)]. \tag{4.5}$$

Here $\theta(t)$ is a step function: $\theta(t) = 1$ for $t \geqslant 0$ and zero otherwise. We can write this result in a simpler form:

$$\sigma_{ij}(\mathbf{r}, \mathbf{r}', t) = \frac{e^2}{(2\pi\hbar)^d m_e^2} \theta(t) \langle p_i(\mathbf{r}, 0) p_j(\mathbf{r}', t) \rangle \tag{4.6}$$

where $\langle \cdots \rangle$ denotes an average over the initial momenta, defined by (4.5). We will consider the evaluation of this quantity for diffusive motion in section 6; next we consider the case of ballistic motion.

4.2. Results specific to ballistic systems

Equation (4.5) can also be expressed as a sum over classical trajectories which travel between $\mathbf{r}$ and $\mathbf{r}'$:

$$\sigma_{ij}(\mathbf{r}, \mathbf{r}'; t) = \frac{e^2}{(2\pi\hbar)^d m_e^2} \theta(t) \sum_{\text{paths}} \left[\det\left(\frac{\partial R_k}{\partial p_l} \right) \right]^{-1} (p_{\text{init}})_j (p_{\text{fin}})_i. \tag{4.7}$$

In the low-temperature limit the term $\partial f/\partial E$ reduces to a delta function, and this expression becomes a sum of delta functions $\delta(t - \tau_j)$, where the τ_j are the times of trajectories from $\mathbf{r}$ to $\mathbf{r}'$ at the Fermi energy E_F.
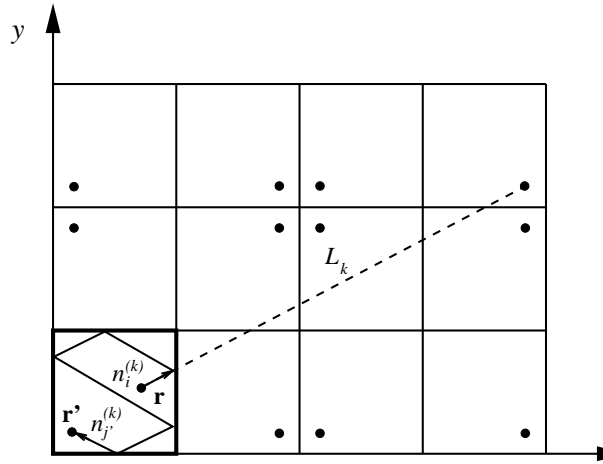

Figure 1. Illustrating the images used in discussing expression (4.11) for a square billiard. The sum over all paths from $\mathbf{r}$ to $\mathbf{r}'$ can be represented as a sum over straight lines k of length L_k connecting $\mathbf{r}$ with the image points of $\mathbf{r}'$.

It is more convenient to consider the frequency-dependent non-local conductivity: if the electric field is $\mathbf{E}(\mathbf{r}) \exp(i\omega t)$, then the current can be written in the form

$$j_i(\mathbf{r}, t) = \exp(i\omega t) \sum_j \int d\mathbf{r}' \; \sigma_{ij}(\mathbf{r}, \mathbf{r}'; \omega) E_j(\mathbf{r}'). \tag{4.8}$$

Comparing with (4.4) and (4.5), we find

$$\sigma_{ij}(\mathbf{r}, \mathbf{r}'; \omega) = \int_0^\infty d\tau \; \exp(i\omega\tau) \sigma_{ij}(\mathbf{r}, \mathbf{r}'; \tau)$$

$$= \frac{e^2}{(2\pi\hbar)^2 m_e^2} \sum_{\text{paths}} \int_0^\infty d\tau \; \exp(i\omega\tau) \int d\theta \; p_i(\theta) p_j'(\theta) \delta(\mathbf{r}' - \mathbf{R}(\mathbf{r}, \mathbf{p}; \tau)) \tag{4.9}$$

where in the second line we have specialized to the case of two dimensions, and θ is the initial angle of the trajectory. Performing the integrations, we find

$$\sigma_{ij}(\boldsymbol{r}, \boldsymbol{r}'; \omega) = \frac{e^2}{(2\pi\hbar)^2 m_e^2} \sum_{\text{paths}} \left[\det\left(\frac{\partial^2 \boldsymbol{R}}{\partial\tau\,\partial\theta} \right) \right]^{-1} p_i(\theta) p_j'(\theta) \exp(\mathrm{i}\omega\tau_k) \quad (4.10)$$

where the sum runs over all trajectories which travel from $\boldsymbol{r}$ to $\boldsymbol{r}'$ in time $\tau_k > 0$ at the Fermi energy. This general expression can be specialized in a variety of ways. We remark that it has a rather simple form for billiards with boundaries consisting of only straight edges. In this case the times τ_k are proportional to the lengths L_k of the trajectories, and because there is no focusing or de-focusing of bundles of trajectories when they bounce off the boundary, the form of the determinant is very simple: we find

$$\sigma_{ij}(\boldsymbol{r}, \boldsymbol{r}'; \omega) = \frac{e^2 p_F}{(2\pi\hbar)^2} \sum_k L_k^{-1} n_i^{(k)} n_{j'}^{(k)} \exp(\mathrm{i}\omega L_k m_e / p_F) \quad (4.11)$$

where n_i, n_j' are the components of a unit vector in respectively the initial and final directions of the trajectory. As an example, the case of a square billiard is illustrated in figure 1. The formula also gives the non-local conductivity in free space, with only the direct trajectory included.

We close this section by remarking that in the limit $\omega \to \infty$ the non-local conductivity becomes a very rapidly varying function of $\boldsymbol{r}'$, except for when the path length of the trajectory is very short. Unless the electric field is a rapidly varying function of position, the dominant contribution to (4.4) comes from the region where $\boldsymbol{r}'$ is close to $\boldsymbol{r}$, implying that a local conductivity $\Sigma_{ij}(\boldsymbol{r}, \omega)$ will give an adequate description. When $\boldsymbol{r}'$ is close to $\boldsymbol{r}$, the non-local conductivity can be approximated by (4.11), with only the direct trajectory included. The local conductivity is then obtained as follows:

$$\Sigma_{ij}(\omega) = \frac{e^2 p_F}{(2\pi\hbar)^2} \int \mathrm{d}\boldsymbol{R}\, \frac{1}{R} \exp(\mathrm{i}\omega m_e R / p_F) n_i n_j' = \frac{\mathrm{i}e^2 p_F^2}{2\pi\hbar^2 m_e \omega} \delta_{ij} = \frac{\mathrm{i}N e^2}{m_e \omega} \delta_{ij} \quad (4.12)$$

where N is the electron density per unit area. This result is precisely the same as the high-frequency limit of the Drude formula for the conductivity.

5. The absorption coefficient for diffusive electrons

5.1. Preliminary comments

In the present section we calculate the absorption coefficient $\alpha(\omega)$ for systems with diffusive electron motion, using (2.6) and (2.7), assuming that the self-consistent electric field is known. We show that the absorption coefficient can be written as a sum of two terms. The first term describes a classical bulk contribution. The second term introduces boundary contributions which could modify the absorption coefficient at frequencies below $\omega_c = D/a^2$.

We begin by briefly discussing the classical expression for the absorption coefficient: it is natural to compare the final answer with this result. The rate of absorption of energy is given by integrating the rate of Joule heating $\boldsymbol{j} \cdot \boldsymbol{E}$ over the volume of the particle. The current density $\boldsymbol{j}$ is proportional to the local electric field: $\boldsymbol{j} = \Sigma_0 \boldsymbol{E} = \mathrm{i}\omega\Sigma_0 \boldsymbol{A}$ where Σ_0 is the bulk conductivity of the metal. We therefore have

$$\alpha(\omega) = \frac{1}{2\Sigma_0 E_0^2} \int \mathrm{d}\boldsymbol{r}\, |\boldsymbol{j}|^2 = \frac{\Sigma_0 \omega^2}{2E_0^2} \int \mathrm{d}\boldsymbol{r}\, \boldsymbol{A}^2 = \frac{ne^2 D\omega^2}{2c^2 B_0^2} \int \mathrm{d}\boldsymbol{r}\, \boldsymbol{A}^2 \quad (5.1)$$

where n is the density of states per unit volume, $n = dN/dE = g/V$. The boundary condition for the electric field is determined by the fact that the current must be tangential to the boundary. Unless there is a constant biasing magnetic field present, E is aligned with j. This implies that $E \cdot \hat{n} = 0$ at all points on the boundary (where $\hat{n}$ is a unit vector normal to the boundary).

Equation (2.5) suggests that the absorption coefficient might exhibit deviations from classical behaviour at frequencies small compared to the Thouless frequency $\omega_c = a^2/D$, which is the inverse of the time taken for an electron to diffuse across the sample. In the following, we give a semiclassical treatment of the absorption coefficient with diffusive electron motion. There are two distinct but related issues which must be addressed here. Firstly, for a given electric field $E(r)$, does the absorption coefficient exhibit any structures at the Thouless energy? Secondly, is the self-consistent solution for the electric field different above and below the Thouless frequency? In this section we consider the first of these issues. In section 6 we will show how one of the results below can be used to determine the non-local conductance, and consider the determination of the self-consistent field in greater detail.

5.2. Calculation of the energy absorbed

Our calculation is based upon (2.6) and (2.7). Because the instantaneous velocity is not well defined for a diffusive trajectory, we will divide the trajectory of the electron into finite segments, in which the electron travels from r_n to r_{n+1} with a uniform velocity, in a fixed time increment δt. The r_n are chosen from an ensemble of random walks confined within the boundary of the particle. The change in the single-electron energy is

$$\Delta E(t) = \text{Re}\left[i e\omega \int_0^t dt' \ \exp(i\omega t) A(r) \cdot \frac{dr}{dt'}\right] = \text{Re}\left[i e\omega \sum_{n=0}^{\mathcal{N}-1} \exp(i\omega t_n) \bar{A}(r_n) \cdot \delta r_n\right] \quad (5.2)$$

where $t = \mathcal{N} \delta t$, $t_n = (n + \frac{1}{2}) \delta t$, $\delta r_n = r_{n+1} - r_n$, and the quantity $\bar{A}$ is defined by the requirement that each term in the sum equals the contribution to the integral from the corresponding link in the random walk. For diffusive motion with fixed diffusion constant, we must take $\langle \delta r^2 \rangle \sim \delta t$, so the error in each step must be $O(\delta r^3)$ to achieve a convergent estimate of the integral. The sum (5.2) can be approximated as follows:

$$\Delta E(t) = \text{Re}\left[i e\omega \sum_{n=0}^{\mathcal{N}-1} \exp(i\omega t_n) A[\tfrac{1}{2}(r_n + r_{n+1})] \cdot (r_{n+1} - r_n)\right] + O(\mathcal{N} \delta r^3) \quad (5.3)$$

and the error term vanishes in the limit $\delta t \to 0$. We remark that equation (5.2) is a stochastic integral, and evaluation of the integrand of the mid-point of the step is equivalent to using the Stratonovich definition of the integral [21]. The variance of (5.3) is

$$\langle \Delta E(t)^2 \rangle = e^2 \omega^2 \sum_{i,j=0}^{\mathcal{N}-1} \exp[i\omega(t_i - t_j)]\langle A[\tfrac{1}{2}(r_i + r_{i+1})] \cdot \delta r_i$$
$$\times A[\tfrac{1}{2}(r_j + r_{j+1})] \cdot \delta r_j \rangle + O(\delta t)$$
$$\sim e^2 \omega^2 \mathcal{N} \sum_{n=-\infty}^{\infty} \exp(i\omega t_n) C_n \quad (5.4)$$

where

$$C_n = \langle A[\tfrac{1}{2}(r_0 + r_1)] \cdot \delta r_0 \ A[\tfrac{1}{2}(r_n + r_{n+1})] \cdot \delta r_n \rangle. \quad (5.5)$$

The absorption coefficient is therefore proportional to the Fourier transform of a correlation function $C(t)$:

$$\alpha(\omega) = \frac{ge^2\omega^2}{2\,\delta t\,B_0^2}\sum_{n=-\infty}^{\infty}\exp(i\omega t_n)C_n \equiv \frac{ge^2\omega^2}{2B_0^2}\int_{-\infty}^{\infty}\mathrm{d}t\,\exp(i\omega t)C(t) \qquad (5.6)$$

where $C(n\,\delta t)\equiv C_n/(\delta t)^2$. The correlation function in (5.5) is an average over diffusing trajectories of the electrons; it can be expressed in terms of the propagator $\mathcal{P}(r,r';t)$ which gives the probability density for reaching r' from initial position r at time t: for times large compared to δt this satisfies the diffusion equation $[\partial_t - D\nabla_r^2]\mathcal{P}=0$. The correlation function (5.5) involves trajectories which visit four different positions, $r_0 = r - \frac{1}{2}\delta r$, $r_1 = r + \frac{1}{2}\delta r$, $r_n = r' - \frac{1}{2}\delta r'$ and $r_{n+1}=r'+\frac{1}{2}\delta r'$, at times 0, δt, $t-\delta t$ and t: three propagators are required to give the probability of for a path visiting these four positions, and the average runs over all four positions. Assuming summation over repeated indices, the correlation function is

$$C(t) = \frac{1}{V\delta t^2}\int\mathrm{d}r\int\mathrm{d}r'\int\mathrm{d}\delta r\int\mathrm{d}\delta r'\;A_i(r+\tfrac{1}{2}\delta r)A_j(r'+\tfrac{1}{2}\delta r')\,\delta r_i\,\delta r'_j$$
$$\times\,\mathcal{P}(r+\delta r,r';t-\delta t)\mathcal{P}(r,r+\delta r;\delta t)\mathcal{P}(r',r'+\delta r';\delta t). \qquad (5.7)$$

We discuss the cases $t=0$ and $t\neq 0$ separately. At $t=0$, we have $\delta r = \delta r'$, and the correlation function is easily evaluated, giving a result which is $O(\delta t)$:

$$C_0 = \langle(A\cdot\delta r)^2\rangle = \frac{2D\,\delta t}{V}\int\mathrm{d}r\;A(r)^2. \qquad (5.8)$$

Here we have used $\langle(A\cdot\delta r)^2\rangle \sim \frac{1}{2}\langle A^2\rangle\langle\delta r^2\rangle = D\,\delta t\,\langle A^2\rangle$. This result implies that there is a contribution to $C(t)$ of the form $(D/V)\,\delta(t)$.
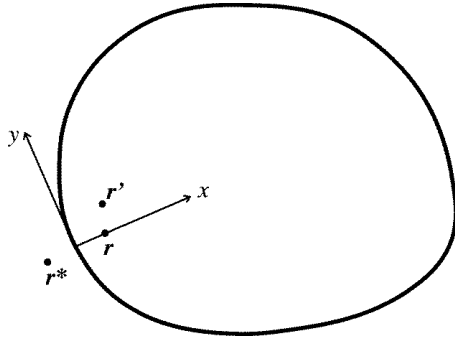

Figure 2. Illustrating the vectors and coordinate system used in the discussion of the construction of the short-time propagator.

The case $t\neq 0$ requires a more delicate treatment. We can expand (5.7) in both the short time interval δt and in the short step δr. It will turn out that the leading-order contribution is $O(\delta t^2)$, as opposed to the case for $t=0$. Because the motion is diffusive, we have $\delta r^2 \sim \delta t$, so terms up to quartic in δr must be retained:

$$C(t) \sim \frac{1}{V\delta t}\int\mathrm{d}r\int\mathrm{d}r'\int\mathrm{d}\delta r\int\mathrm{d}\delta r'\;[A_i + \tfrac{1}{2}\partial_{r_k}A_i\,\delta r_k + \tfrac{1}{8}\partial^2_{r_k r_l}A_i\,\delta r_k\,\delta r_l]$$
$$\times\,[A'_j + \tfrac{1}{2}\partial_{r'_k}A'_j\,\delta r'_k + \tfrac{1}{8}\partial^2_{r'_k r'_l}A'_j\,\delta r'_k\,\delta r'_l]\,\delta r_i\,\delta r'_j$$
$$\times\,[\mathcal{P}(r,r';t) - \partial_t\mathcal{P}(r,r';t)\,\delta t + \partial_{r_k}\mathcal{P}(r,r';t)\,\delta r_k$$
$$+\,\tfrac{1}{2}\partial^2_{r_k r_l}\mathcal{P}(r,r';t)\,\delta r_k\,\delta r_l]\mathcal{P}(r,r+\delta r;\delta t)\mathcal{P}(r',r'+\delta r';\delta t). \qquad (5.9)$$

The terms containing $\partial_t \mathcal{P}(r, r'; t)$ can be dropped when there are more than two factors of δr. The integrals over products of the δr can now be separated out to give

$$
\begin{aligned}
C(t) = \frac{1}{V\delta t^2} \int dr \int dr' \Big\{ & A_i A_j [\mathcal{P} - \partial_t \mathcal{P}\, \delta t] \langle \delta r_i \rangle \langle \delta r'_j \rangle \\
& + [\tfrac{1}{2} A'_j\, \partial_{r_k} A_i\, \mathcal{P} + \tfrac{1}{2} A'_j\, \partial_{r_k}(\mathcal{P} A_i) - \tfrac{1}{2} A'_j\, \partial_{r_k} A_i\, \mathcal{P}] \langle \delta r_i\, \delta r_k \rangle \langle \delta r'_j \rangle \\
& + [\tfrac{1}{2} A_i\, \partial_{r_k} A'_j\, \mathcal{P}] \langle \delta r'_j\, \delta r'_k \rangle \langle \delta r_i \rangle \\
& + [\tfrac{1}{8} A_i\, \partial^2_{r_k r_l} A'_j\, \mathcal{P} + \tfrac{1}{2} A_i A_j\, \partial^2_{r_k r_l} \mathcal{P}] \langle \delta r_i\, \delta r_k\, \delta r_l \rangle \langle \delta r'_j \rangle \\
& + [\tfrac{1}{2} A_i\, \partial^2_{r_k r_l} A'_j\, \mathcal{P}] \langle \delta r'_j\, \delta r'_k\, \delta r'_l \rangle \langle \delta r_i \rangle \\
& + [\tfrac{1}{4} \partial_{r_l} A_j\, \partial_{r_k} A_i\, \mathcal{P} + \tfrac{1}{2} A_i\, \partial_{r_l} A_j\, \partial_{r_k} \mathcal{P}] \langle \delta r_i\, \delta r_k \rangle \langle \delta r'_l\, \delta r'_j \rangle \Big\}
\end{aligned}
\tag{5.10}
$$

where $\langle \delta r \rangle = \int d\delta r\, \delta r\, \mathcal{P}(r, r + \delta r; \delta t)$. Now consider the form of these integrals when δt is sufficiently small. The propagator $\mathcal{P}(r, r + \delta r; \delta t)$ is small unless δr is small. When r is not close to the boundary, this propagator can be approximated by a function of the distance travelled, $\mathcal{P}_0(|\delta r|, \delta t)$: because the steps are assumed to be independent, the variance $\langle \delta r^2 \rangle$ averaged over this distribution can be identified with $2 d D\, \delta t$ (where d is the dimensionality of space). When r is close to the boundary, a solution satisfying the boundary condition $n \cdot \nabla \mathcal{P} = 0$ is constructed by the method of images. We denote the image of the source point r by r^*. The diffusion propagator is then

$$
\mathcal{P}(r, r + \delta r; \delta t) \sim \mathcal{P}_0(|\delta r|, \delta t) + \mathcal{P}_0(|\delta r + r - r^*|, \delta t).
\tag{5.11}
$$

Since δt is small compared to the time taken to traverse the particle, the second term only contributes for points r close to the surface, which can thus be considered locally flat. We then introduce a local coordinate system arranged so that the nearest boundary point defines the origin. In two dimensions, the surface tangent is given by the line $x = 0$, and the normal by $y = 0$. The point r lies at $(x, 0)$, and $r^* = (-x, 0)$ (figure 2).

The average $\langle \delta r_i \rangle$ vanishes unless r is close to the boundary, in which case the mean displacement is inwards, and its projection in the direction perpendicular to the surface is

$$
\langle \delta x \rangle_x = \int_0^\infty dx'\ (x' - x)[f(x' - x) + f(x' + x)]
\tag{5.12}
$$

where $f(x)$ is the projection of the distribution $\mathcal{P}_0(|\delta r|)$ onto the x-axis:

$$
f(x) = \int dr\ \mathcal{P}_0(|\delta r|, \delta t)\delta(x - \delta r_x)
\tag{5.13}
$$

where δr_x is the x-component of δr; this satisfies

$$
\int_{-\infty}^\infty dx\ x^2 f(x) = 2D\, \delta t.
\tag{5.14}
$$

Equations (5.12) and (5.14) show that the mean inward displacement $\langle \delta x \rangle_x$ is of typical magnitude $\sqrt{D\, \delta t}$, in a layer of depth $\sqrt{D\, \delta t}$ next to the boundary, and negligible elsewhere. The weight w of these inward displacements is clearly $\sim D\, \delta t$. We define

$$
w \equiv \lim_{L \to \infty} \int_0^L dx\ \langle \delta x \rangle_x.
\tag{5.15}
$$

We evaluate w by substituting (5.12), then making a change of variables $X = x' + x$, $X' = x' - x$. The integral is written as the sum of two integrals, one over the domain

$X \leqslant L$, $|X'| \leqslant X$, which vanishes because of a symmetry, and another integral which involves only $f(X')$ in the limit $L \to \infty$: we find $w = D\,\delta t$, which implies that

$$\int \mathrm{d}\boldsymbol{r}\ F_i(\boldsymbol{r})\langle \delta r_i \rangle = -D\,\delta t \int \mathrm{d}s_i\ F_i \tag{5.16}$$

for any vector field $\boldsymbol{F}$, where $\mathrm{d}s_i$ are the components of a vector element of the surface. When evaluating the integrals over $\langle \delta r_i\,\delta r_j \rangle$ we can approximate these terms by $2D\,\delta t\,\delta_{ij}$, because the second term in (5.12) is significant only in a narrow layer of width $\sqrt{D\,\delta t}$. The terms containing averages of δr^3 make no contribution at order δt^2. Retaining only the leading-order terms, we find the following contribution for $t \neq 0$:

$$
\begin{aligned}
C(t) = {} & \frac{D^2}{V} \int \mathrm{d}\boldsymbol{r} \int \mathrm{d}\boldsymbol{r}'\ \partial_{r_i} A_i(\boldsymbol{r})\,\partial_{r'_j} A_j(\boldsymbol{r}')\,\mathcal{P}(\boldsymbol{r},\boldsymbol{r}';t) \\
& - 2\frac{D^2}{V} \int \mathrm{d}s_i \int \mathrm{d}\boldsymbol{r}'\ A_i(\boldsymbol{r})\,\partial_{r'_j} A_j(\boldsymbol{r}')\,\mathcal{P}(\boldsymbol{r},\boldsymbol{r}';t) \\
& + \frac{D^2}{V} \int \mathrm{d}s_i \int \mathrm{d}s'_j\ A_i(\boldsymbol{s})A_j(\boldsymbol{s}')\mathcal{P}(\boldsymbol{s},\boldsymbol{s}';t).
\end{aligned} \tag{5.17}
$$

After integrating by parts, and adding the delta-function contribution from $t = 0$, we find

$$C(t) = D\,\delta(t) \int \mathrm{d}\boldsymbol{r}\ A_i A_i - \frac{D^2}{V} \int \mathrm{d}\boldsymbol{r} \int \mathrm{d}\boldsymbol{r}'\ A_i A_j\,\partial^2_{r_i r'_j}\mathcal{P}(\boldsymbol{r},\boldsymbol{r}';t). \tag{5.18}$$

Before Fourier transforming this expression to determine the absorption coefficient, we will introduce a convenient expression for the propagator:

$$\mathcal{P}(\boldsymbol{r},\boldsymbol{r}';t) = \theta(t) \sum_{\alpha} \chi_\alpha(\boldsymbol{r})\chi_\alpha(\boldsymbol{r}') \exp(-Dk_\alpha^2 t) \tag{5.19}$$

where the $\chi_\alpha(\boldsymbol{r})$ are solutions of the Helmholtz equation $[\Delta + k_\alpha^2]\chi_\alpha = 0$, satisfying the Neumann boundary condition $n_i\,\partial_{r_i}\chi_\alpha = 0$. Using this result, we find the following expression for the absorption coefficient:

$$\alpha(\omega) = K\omega^2 \left[\int \mathrm{d}\boldsymbol{r}\ A^2 - \sum_\alpha \frac{D^2 k_\alpha^2}{(Dk_\alpha^2)^2 + \omega^2} \left| \int \mathrm{d}\boldsymbol{r}\ A_i\,\partial_{r_i}\chi_\alpha \right|^2 \right] \tag{5.20}$$

where

$$K = \frac{ge^2 D}{B_0^2} = \frac{\Sigma_0}{B_0^2}. \tag{5.21}$$

Equation (5.20) is the main result of this section. As pointed out above, it consists of two terms. The first term is just the classical result derived at the beginning of this section. The second term is written as a sum over eigenmodes of the diffusion propagator. It incorporates boundary effects, as will be seen in the next section.

5.3. The low-frequency limit

We conclude this section with two remarks concerning the low-frequency limit. First, we show that the result (5.20) fulfils a condition discussed at the end of section 3, where we noted that the low-frequency limit of the absorption coefficient must be invariant under adding any gradient to the vector potential. To see this, consider the effect of the following transformation:

$$A_i \to A_i + \partial_{r_i}\varphi. \tag{5.22}$$

In the limit $\omega \to 0$ the absorption coefficient is determined by the integral of the correlation function. The addition of the term $\partial_{r_i}\varphi$ produces two new terms in the delta function contribution in (5.18), one an integral containing $A_i \partial_{r_i}\varphi$, the other containing $\partial_{r_i}\varphi \partial_{r_i}\varphi$. Corresponding additional terms appear in the double space integral in (5.18). Consider the first of these terms, linear in $\partial_{r_i}\varphi$. In order for these terms to make no contribution to the absorption coefficient, it is sufficient to show that the following two integrals are equal:

$$I = D \int_{-\infty}^{\infty} dt \int dr \int dr' \ A_i \, \partial_{r_j}\varphi \, \partial^2_{r_i, r'_j} \mathcal{P} \tag{5.23}$$

$$I' = 2 \int dr \ A_i \, \partial_{r_i}\varphi. \tag{5.24}$$

We will use the result

$$\delta(r - r') = \sum_{\alpha} \chi_{\alpha}(r)\chi_{\alpha}(r'). \tag{5.25}$$

Using (5.20), we find

$$I = 2 \sum_{\alpha} \frac{1}{k_{\alpha}^2} \int dr \int dr' \ A_i \, \partial_{r_j}\varphi \, \partial_{r_i}\chi_{\alpha}(r) \, \partial_{r_j}\chi_{\alpha}(r')$$

$$= 2 \int dr \int dr' \ A_i\varphi(r') \, \partial_{r_i}\chi_{\alpha}(r) \, \chi_{\alpha}(r')$$

$$= 2 \int dr \int dr' \ A_i\varphi(r') \, \partial_{r_i}\delta(r - r') = I'. \tag{5.26}$$

This shows that terms involving $A_i \partial_{r_i}\varphi$ cancel and do not contribute to the absorption in the limit $\omega \to 0$. Since this result applies for any vector field A_i, we can replace A_i by $\partial_{r_i}\varphi$ and deduce immediately that the terms quadratic in $\partial_{r_i}\varphi$ also cancel.

Second, we comment on the form of the absorption coefficient in the limits $\omega \gg \omega_c$ and $\omega \ll \omega_c$. The vector potential can always be written as a sum of the curl of a divergenceless field, and a gradient:

$$i\omega A = a + \nabla\varphi = \nabla \times \psi + \nabla\varphi \qquad \nabla \cdot \psi = 0 \tag{5.27}$$

with the field a chosen so that it is tangential to the boundary ($\hat{n} \cdot a = 0$). In the case $\omega \gg \omega_c$, the absorption coefficient is determined by the delta-function contribution, and we have

$$\lim_{\omega/\omega_c \to \infty} \alpha(\omega) = K\omega^2 \int dr \ (a^2 + \nabla\varphi^2) \tag{5.28}$$

with K given by (5.21). In the case $\omega \ll \omega_c$, on the other hand, the calculation that we described above shows that the potential φ makes no contribution to the absorption coefficient, and that

$$\lim_{\omega/\omega_c \to 0} \alpha(\omega) = K\omega^2 \int dr \ a^2. \tag{5.29}$$

We showed in section 3 that if the vector potential is written in the form (5.27), then the potential φ is zero when the conductivity is isotropic and local. Inspection of (5.20) shows that the boundary contribution to the correlation function vanishes when φ vanishes. In more general cases, comparison of (5.28) and (5.29) shows that if the electric field is independent of frequency, the absorption coefficient is reduced, relative to its classical value, at frequencies below the Thouless frequency ω_c.

6. Self-consistent electric field: the diffusive case

6.1. Non-local conductance

We can deduce the non-local conductivity from the results of the previous section in two ways. We could use (4.6) as the definition of the conductivity, and evaluate it by setting $A(r) = \delta(r - R)e_i$ in (5.5), so that

$$\sigma_{ij}(R, R'; t) = \frac{e^2}{(2\pi\hbar)^d \delta t^2}\theta(t)\langle \delta(R - r)\,\delta r_i\,\delta(R' - r')\,\delta r'_j\rangle. \tag{6.1}$$

Alternatively, if we write the absorption coefficient in terms of the non-local conductivity in the form

$$\alpha(\omega) = \frac{1}{2E_0^2}\int dr \int dr' \int d\tau\ \exp(i\omega\tau)\sigma_{ij}(r, r', \tau)E_i(r)E_j(r') \tag{6.2}$$

the kernel $\sigma_{ij}(r, r'; t)$ is deduced from (5.18). By either route we find, in the frequency domain,

$$\sigma(r, r'; \omega) = \Sigma_0\Big[\delta_{ij}\delta(r - r') - D\,\partial_{r_i}\,\partial_{r'_j}\mathcal{P}(r, r'; \omega)\Big] \tag{6.3}$$

where Σ_0 is the bulk conductivity, and $\mathcal{P}(r, r'; \omega)$ is the Fourier transform of the propagator, given by (5.19).

   This form for the non-local conductivity was originally given in reference [16] (and in the DC limit in reference [15]). The argument in these earlier papers involves the diagrammatic analysis of disorder-averaged perturbation theory, and in the case of [16] it appeals to a sigma model formalism. We believe that our derivation is more direct, and also more compelling. Our derivation considers the effect of the surface explicitly, whereas it is not clear from the diagrammatic analysis that there are not additional contributions which arise from integrating fields over the surface of the sample. Our derivation also deals explicitly with the fact that the trajectories are discontinuous, and we show explicitly how the correct evaluation is related to the Stratonovich definition of the integral over the trajectory.

6.2. Self-consistent solution

The self-consistent electric field has to be chosen to satisfy (3.5). First we remark that in the case of diffusive electron motion, the term containing ϵ_0 is negligible, and can be dropped. Estimating the magnitude of $\hat{\sigma}$ by the bulk conductance $\Sigma_0 = ne^2 D$, and noting that the bulk plasma frequency scale is $\omega_p^2 \sim Ne^2/m_e\epsilon_0$, we see that this term is negligible provided that $\omega \gg \omega_s$, where ω_s is the elastic scattering rate. This is consistent with the assumption that the electron motion is diffusive.

   We therefore wish to determine an electric field for which $\nabla\cdot(\hat{\sigma}E) = 0$. Using (6.3) this satisfies

$$\partial_{r_i}\int dr'\,\Big[\delta(r - r')E_i(r') - D\,\partial_{r_i}\,\partial_{r'_j}\mathcal{P}(r, r'; \omega)E_j(r')\Big] = 0. \tag{6.4}$$

Using the diffusion equation we find

$$i\omega\Big[\int dr'\,\mathcal{P}(r, r'; \omega)\,\partial_{r'_j}E_j(r') - \int ds'_j\,\mathcal{P}(r, r'; \omega)E_j(r')\Big] = 0. \tag{6.5}$$

This equation has a solution where $\nabla\cdot E = 0$ everywhere, with E tangential to the boundary. The classical solution for an isotropic local conductance satisfies these conditions.

6.3. Calculation of the absorption coefficient

We can now use the solution of the form (3.7), (3.8), with $\phi = 0$, and with ψ tangential to the boundary. Referring to (5.20) we observe that, after integrating by parts, the integrals are seen to vanish because the field A is divergenceless and is tangential at the boundary. It follows that the summation in (5.20) vanishes, and that the absorption coefficient is given by the classical expression (5.1), at all relevant frequencies.

We conclude by describing a useful approach to calculating the field ψ, satisfying Poisson's equation (3.9). The solution can be obtained from a Green's function $G(r, r')$ satisfying $\nabla^2 G = -\delta(r - r')$. A suitable Green's function is

$$G(r, r') = \sum_n \frac{\xi_n(r)\xi_n(r')}{k_n^2} \tag{6.6}$$

where the $\xi_n(r)$ and k_n^2 are eigenfunctions and eigenvalues of the Helmholtz equation, $[\nabla^2 + k_n^2]\xi_n = 0$, solved with the Dirichlet boundary condition $\xi_n(r) = 0$. The field $\psi(r)$ is then obtained by applying this Green's function to the source term $i\omega B_0$ appearing in (3.9). In two dimensions, the absorption coefficient can then be written in terms of the ξ_n as follows:

$$\alpha(\omega) = \frac{\Sigma_0}{2E_0^2} \int dr\, |\nabla\psi|^2 = -\frac{\Sigma_0}{2E_0^2} \int dr\, \psi\, \nabla^2\psi^* = i\omega \frac{\Sigma_0 B_0}{2(E_0)^2} \int dr\, \psi. \tag{6.7}$$

Using the Green's function (6.6) to obtain ψ, we have

$$\alpha(\omega) = \frac{\Sigma_0 \omega^2}{2c^2} \sum_n \frac{1}{k_n^2} \left| \int dr\, \xi_n(r) \right|^2. \tag{6.8}$$

This is a very general expression for the classical magnetic dipole absorption coefficient in particles with diffusive electron motion, expressed in terms of solutions of the two-dimensional Helmholtz equation. As it stands, equation (6.8) is valid for two-dimensional particles.

We note that the eigenfunctions $\xi_n(r)$ obey Dirichlet boundary conditions and that the absorption coefficient for a given geometry can be significantly reduced by applying cuts orthogonal to the boundary: the main contribution to the sum in (6.8) comes from the ground state and the low-lying states and the corresponding eigenvalues are increased by applying cuts. This behaviour is expected since such cuts inhibit the flow of eddy currents which causes the absorption.

6.4. Some examples for simple geometries

In this section we summarize our results for specific geometries, namely discs, squares, and spheres (the result for the latter is well known [1, 8], and is included to establish connections with earlier work). For two-dimensional discs of radius a, we have $\psi = i\omega B_0 r^2/4$. Using (3.10) and (5.1) we obtain

$$\alpha(\omega) = \frac{\pi}{16} \frac{\Sigma_0 \omega^2 a^4}{c^2}. \tag{6.9}$$

This result is easily shown to be consistent with (6.8), using the fact that the k_n are defined by $J_0(k_n a) = 0$ and that $\sum_n k_n^{-4} = a^4/32$. For squares of side-length a, we obtain

$$\alpha(\omega) = \frac{32}{\pi^6} \frac{\Sigma_0 \omega^2 a^4}{c^2} \sum_{\substack{m,n>0}}^{odd} \frac{1}{m^2 n^2} \frac{1}{m^2 + n^2}. \tag{6.10}$$

The sum can be evaluated numerically and gives $0.528\ldots$. Finally, for spheres of radius a, equation (6.8) is to be modified as follows:

$$\alpha(\omega) = \frac{\Sigma_0\omega^2}{2c^2} \int_{-a}^{a} \mathrm{d}z \sum_n \frac{1}{k_n^2} \left| \int \mathrm{d}r \; \xi_n(r) \right|^2 \tag{6.11}$$

where the eigenvalues are defined by $J_0(k_n r_\perp) = 0$ and $r_\perp^2 = a^2 - z^2$. We obtain

$$\alpha(\omega) = \frac{\pi}{15} \frac{\Sigma_0\omega^2 a^5}{c^2}. \tag{6.12}$$

Equation (6.12) reproduces the well-known absorption coefficient for metallic spheres [1]. In [1], this result is compared to the absorption coefficient for electric dipole absorption, which has a different size dependence, $\sim a^3$. In two dimensions, on the other hand, the size dependence is the same for magnetic and electric dipole absorption. The latter coefficient was calculated in [13]:

$$\alpha_{\mathrm{el}}(\omega) = \frac{34}{9\pi} \frac{\epsilon_0^2 a^4 \omega^2}{\Sigma_0}. \tag{6.13}$$

7. Ballistic electron motion

7.1. General remarks

In the case where the electron motion is ballistic, the electric field must be determined by the non-local conductivity, and equations (3.5) and (3.6) must be solved to determine the electric field. Fortunately, in both the high- and low-frequency limits, there are considerations which simplify the discussion.

In the low-frequency limit, $\omega \ll \omega_c$, we showed in section 3 that only $\nabla \times E$ is relevant, and that we can use any electric field for which the circulation is spatially uniform. It follows from (2.3)–(2.5) that the absorption coefficient is proportional to ω^2 at low frequencies.

In the high-frequency limit, the conductivity tensor (4.10) acts over a range $R \sim v_F/\omega$, and the conductivity becomes effectively local, with value $\Sigma(\omega) = \mathrm{i}Ne^2/m_e\omega$. The non-local self-consistency condition for the electric field then reduces to the same requirements as for the diffusive case: the electric field is tangential to the boundary, and is derived from a field ψ which satisfies (3.9).

From (2.5), it is clear that the high-frequency behaviour is determined by discontinuities in derivatives of the correlation function. The correlation function of the smooth perturbation $f(t) = \dot{r} \cdot E$ for motion in a billiard has discontinuous derivatives due to the change of direction when the particle collides with the boundary. In the neighbourhood of a collision with the boundary at $t = 0$, the perturbation takes the form

$$f(t) = (\dot{r}' + \dot{r}'') \cdot E[(\dot{r}' + \dot{r}'')t]\Theta(-t) + (\dot{r}' - \dot{r}'') \cdot E[(\dot{r}' - \dot{r}'')t]\Theta(t) \tag{7.1}$$

where $\dot{r}'$ and $\dot{r}''$ are respectively tangential and normal components of the velocity at the instant before the collision, and $\Theta(t)$ is the step function. Taylor expanding $E(r)$ we find (with repeated indices summed over)

$$\begin{aligned} f(t) \sim \dot{r}_i' E_i + \dot{r}_i'' E_i \{\Theta(-t) - \Theta(t)\} + (\dot{r}_i' \dot{r}_j' + \dot{r}_i'' \dot{r}_j'')\, \partial_{r_j} E_i\, t \\ + (\dot{r}_i' \dot{r}_j'' + \dot{r}_i'' \dot{r}_j')\, \partial_{r_j} E_i\, t \{\Theta(-t) - \Theta(t)\}. \end{aligned} \tag{7.2}$$

For a general electric field, $f(t)$ has discontinuities in t of magnitude $2\dot{r}_i'' E_i$ on collision with the boundary, but for electric fields tangential to the boundary, the discontinuities are in the first derivative, and are of magnitude $2(\dot{r}_i' \dot{r}_j'' + \dot{r}_i'' \dot{r}_j')\, \partial_{r_j} E_i$.

If $f(t)$ has discontinuities in its nth derivative, the Fourier transform of its correlation function decays as $\omega^{-2(n+1)}$ as $\omega \to \infty$. The absorption coefficient is obtained from this Fourier transform by multiplying by a factor which contains ω^2. In the case of a general field we therefore expect the absorption coefficient to approach a constant for $\omega \gg \omega_c$, whereas for a tangential field we expect that $\alpha(\omega) \sim \omega^{-2}$ for $\omega \gg \omega_c$. We know that the field $\boldsymbol{E}(\boldsymbol{r}, \omega)$ approaches the tangential form as $\omega \to \infty$, but we have no information about how rapidly this limit is approached. We can only say that the absorption coefficient must decrease for $\omega \gg \omega_c$, and that it is unlikely to decrease faster than ω^{-2}.

7.2. An example: the square billiard

It is instructive to discuss an example: we consider the absorption coefficient for a square billiard with ballistic electron motion, with two different, frequency-independent, choices for the electric field $\boldsymbol{E}(\boldsymbol{r})$, both satisfying (3.7), (3.9). First we calculate the absorption coefficient assuming that the field is tangential to the boundary, and then consider the case where it is circularly symmetric (which corresponds to taking the angular momentum operator $\hat{L}_z$ as the perturbation. The results will illustrate the application of (2.5), and will verify two of the conclusions from the arguments above: we find that the absorption coefficients agree in the low-frequency limit, and that at high frequencies the absorption scales as ω^{-2} for the tangential field, but as ω^0 for the radially symmetric field.

In terms of the perturbation

$$\Delta H(\boldsymbol{r}, \boldsymbol{p}) = \frac{e}{m_e} \boldsymbol{p} \cdot \boldsymbol{A}(\boldsymbol{r}) \tag{7.3}$$

the absorption coefficient is given by

$$\alpha(\omega) = \frac{\omega^2}{2} g(E_F) \, \text{Re} \int_0^\infty \mathrm{d}t \, \mathrm{e}^{\mathrm{i}\omega t} \Big\langle \Delta H(\boldsymbol{r}_t, \boldsymbol{p}_t) \, \Delta H(\boldsymbol{r}, \boldsymbol{p}) \Big\rangle. \tag{7.4}$$

The perturbation (7.3) is determined by the choice of the vector potential $\boldsymbol{A}(\boldsymbol{r})$. We will first assume that $\boldsymbol{A}(\boldsymbol{r})$ is tangential to the boundary of the particle. Accordingly we take $\phi = 0$ in equation (3.7). The field $\psi(x, y)$ in equation (3.8) is determined from equation (3.9) which is most conveniently solved using the Green's function (6.6). For a square of side a we obtain for the perturbation

$$\Delta H(\boldsymbol{r}, \boldsymbol{p}) = \frac{e}{m_e} B_0 a^2 \left(\frac{2}{\pi} \right)^4 \Bigg[p_x \sum_{mn}^{\text{odd}} \frac{n\pi}{a} \frac{\sin(m\pi x/a) \cos(n\pi y/a)}{mn(m^2 + n^2)} $$
$$+ p_y \sum_{mn}^{\text{odd}} \frac{m\pi}{a} \frac{\cos(m\pi x/a) \sin(n\pi y/a)}{mn(m^2 + n^2)} \Bigg]. \tag{7.5}$$

Since motion in the square is integrable, the autocorrelation function of the perturbation in (7.4) is calculated as an average over tori:

$$\Big\langle \Delta H(\boldsymbol{r}_t, \boldsymbol{p}_t) \, \Delta H(\boldsymbol{r}, \boldsymbol{p}) \Big\rangle = \int \frac{\mathrm{d}^2\theta}{(2\pi)^2} \, \mathrm{d}\mu(\boldsymbol{I}) \, \Delta H(\boldsymbol{I}, \boldsymbol{\theta}) \, \Delta H(\boldsymbol{I}, \boldsymbol{\theta} + \boldsymbol{\omega}(\boldsymbol{I})t) \tag{7.6}$$

where $\mathrm{d}\mu(\boldsymbol{I}) = g(E_F)^{-1} \delta[E_F - H(\boldsymbol{I})]$ averages over the tori. $\boldsymbol{I}$ and $\boldsymbol{\theta}$ are the action and angle variables characterizing the motion in the square; $\boldsymbol{\omega}(\boldsymbol{I})$ are the respective frequencies. Equation (7.6) is easily evaluated [22]. The result is of the form

$$\alpha(\omega) = \frac{8}{\pi^8} \frac{m_e e^2 \omega^2 a^5 v_F}{c^2 \hbar^2} f(\omega/\omega_c) \tag{7.7}$$

where $f(z)$ is an energy-independent scaling function and $\omega_c = v_F/a$. For large frequencies, $f(z) \sim z^{-4}$ and hence $\alpha(\omega) \sim \omega^{-2}$. For small frequencies, on the other hand, one obtains

$$\alpha(\omega) = \frac{8}{\pi^8} \frac{m_e e^2 \omega^2 a^5 v_F}{c^2 \hbar^2} \sum_{m,n>0}^{\text{odd}} \frac{1}{m^2 n^2} \frac{1}{(m^2+n^2)^{3/2}}. \tag{7.8}$$

As remarked in section 7.1, the absorption coefficient is proportional to ω^2 for small frequencies.

In order to verify explicitly that the low-frequency absorption does not depend on the boundary conditions for the electric field, as discussed in sections 3.3 and 7.1, the above calculation can be repeated using a vector potential in the symmetric gauge:

$$\boldsymbol{A} = \frac{B_0}{2}(-y, x, 0). \tag{7.9}$$

We note that this choice of the vector potential does not satisfy tangential boundary conditions. The corresponding perturbation is

$$\Delta H(\boldsymbol{r}, \boldsymbol{p}) = \frac{e}{m_e} \boldsymbol{p} \cdot \boldsymbol{A} = \frac{eB_0}{2m_e} L_z. \tag{7.10}$$

For small frequencies we find again the result (7.8), thus verifying explicitly that the boundary conditions do not influence the low-frequency absorption. For high frequencies, on the other hand, we find $\alpha(\omega) \sim \omega^0$, as predicted in the previous section.

Acknowledgments

References

[1] Carr G L, Perkowitz S and Tanner D B 1985 *Infrared and Millimeter Waves* vol 13, ed K J Button (New York: Academic) p 169
[2] Perenboom J A A J, Wyder P and Meier F 1981 *Phys. Rep.* **78** 173
[3] Halperin W P 1986 *Rev. Mod. Phys.* **58** 533
[4] Akkermans E, Montambaux G, Pichard J-L and Zinn-Justin J (ed) 1995 *Mesoscopic Quantum Physics (Les Houches Session LXI)* (Amsterdam: North-Holland)
[5] Gorkov L P and Eliashberg G M 1965 *Zh. Eksp. Teor. Fiz.* **48** 1407 (Engl. Transl. 1965 *Sov. Phys.–JETP* **21** 940)
[6] Mie G 1908 *Ann. Phys., Lpz.* **25** 377
[7] Ashcroft N W and Mermin N D 1976 *Solid State Physics* (Philadelphia, PA: Saunders College)
[8] Landau L D and Lifshitz E M 1960 *Electrodynamics of Continuous Media (Landau and Lifshitz Course of Theoretical Physics 8)* (Oxford: Pergamon)
[9] Tanner D B, Sievers A J and Buhrman R A 1975 *Phys. Rev.* B **11** 1330
[10] Stroud D and Pan F P 1978 *Phys. Rev.* B **17** 1602
[11] Austin E J and Wilkinson M 1993 *J. Phys.: Condens. Matter* **5** 8461
[12] Wilkinson M and Austin E J 1994 *J. Phys.: Condens. Matter* **6** 4153
[13] Mehlig B and Wilkinson M 1997 *J. Phys.: Condens. Matter* **9** 3277
[14] Argaman N 1993 *Phys. Rev.* B **47** 4440
[15] Kane C L, Serota R A and Lee P A 1988 *Phys. Rev.* B **37** 6701

[16] Serota R A, Yu J and Kim Y H 1990 *Phys. Rev.* B **42** 9724
[17] Wilkinson M 1987 *J. Phys. A: Math. Gen.* **20** 2415
[18] Thouless D J 1974 *Phys. Rep.* **13** 93
[19] Kamenev A and Gefen Y 1995 *Int. J. Mod. Phys.* B **9** 751
[20] Reulet B, Ramin M, Bouchiat H and Mailly D 1996 *Phys. Rev. Lett.* **75** 124
[21] van Kampen N G 1981 *Stochastic Processes in Physics and Chemistry* (Amsterdam: North-Holland)
[22] Mehlig B 1997 *Phys. Rev.* B **55** R10 193