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Collective versus single-particle effects in the optical spectra of finite electronic quantum systems

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We study optical spectra of finite electronic quantum systems at frequencies smaller than the plasma frequency using a quasiclassical approach. This approach includes collective effects and enables us to analyze how the nature of the (single-particle) electron dynamics influences the optical spectra in finite electronic quantum systems. We present an analytical expression for the low-frequency absorption coefficient of electromagnetic radiation in a finite quantum system with regular ballistic electron dynamics; a two-dimensional electron gas confined to a strip of width a with specular reflection at the boundaries (our approach is not restricted to systems with regular electron dynamics; it applies equally in the case of diffusive or classically chaotic electron motion). By comparing with results of numerical computations using the random-phase approximation we show that our analytical approach provides a qualitative and quantitative understanding of the optical spectrum.

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Optical spectra of finite metallic systems have been intensively investigated for almost a century. Early approaches such as Mie's[1] are of classical nature. In Ref. 1 the absorption of electromagnetic radiation by conducting spheres is determined. It is shown that the absorption spectrum exhibits a resonance at $\omega_p/\sqrt{3}$ (where ω_p is the bulk plasma frequency) due to collective motion of the charge carriers.

In the last decades there has been a substantial amount of work on the nature of such collective resonances in metallic clusters,[2] nuclei,[3] thin films,[4–6] small metal particles,[7] and dimensionally reduced quantum systems[8–10] investigating, in particular, quantum-mechanical effects. In most of these cases, the electron dynamics is *ballistic* (the mean free path l is larger than the system size a). The majority of theoretical papers on the Mie resonance of finite metallic systems use the so-called random-phase approximation (RPA), a self-consistent, quantum-mechanical approach incorporating collective effects. The nature of the Mie resonance in finite electronic quantum systems is well understood, both qualitatively and quantitatively (for a recent review see, for instance, Ref. 2).

The emergence of the field of mesoscopic physics has fueled an increased interest in the electronic and optical properties of finite, disordered quantum systems (with *diffusive* electron dynamics, $l \ll a$) in external fields. In this context, attention has largely focused on static properties, i.e., on frequencies of the order of or smaller than $\Delta/\hbar$, where Δ is the mean level spacing of the system in question; $\Delta/\hbar$ is generally smaller than ω_p by many orders of magnitude. Mesoscopic fluctuations of the static polarizability and the capacitance, for instance, were characterized in Ref. 11, and the electron density itself in Ref. 12. In all of these cases, collective effects must be taken into account in order to adequately deal with the screening of the external field. In the static limit, a Thomas-Fermi (TF) *ansatz* is appropriate.[11,12] This was pointed out in Refs. 13–15 discussing the results of Gorkov and Eliashberg[16] on the polarizability of small metallic particles with disordered walls.

Much less is known about optical spectra in the frequency domain $\Delta/\hbar < \omega \ll \omega_p$, despite the fact that this regime is of particular interest: One expects that the spectra strongly depend on the nature of the (single-particle) electron dynamics: In *ballistic* systems with *regular* dynamics, for example, it was argued[17] that optical spectra should exhibit *resonances near multiples of* $\omega_c = \pi v_F/a$ (see also Ref. 18). Usually $\Delta/\hbar \ll \omega_c \ll \omega_p$. Moreover, according to Ref. 17, these resonances should overlap to give a *linear frequency dependence* of the absorption coefficient for $\omega \gg \omega_c$ in two-dimensional systems (classical and local electromagnetic theories predict a quadratic dependence independent of dimension). These are striking and unexpected results. They were, however, obtained within a TF approximation which is valid in the *static* limit. It must be examined to which extent *dynamic* screening effects may modify the results. Finally, many experimentally relevant systems exhibit chaotic electron dynamics. It is thus necessary to quantify how optical spectra differ from the above predictions when the electron dynamics is classically chaotic.[19]

Numerical calculations (based, for instance on the RPA) are ill-suited to answer these questions: They provide little qualitative insight in the low-frequency region and, more importantly, it is necessary to consider small systems or to make use of symmetries in order to make the numerical computations feasible. Disordered and (asymmetric) chaotic systems are very difficult to deal with. In order to understand how the classical single-particle dynamics is reflected in optical spectra of finite quantum systems, it is thus greatly desirable to have an analytical theory incorporating collective effects.

The main result of this paper is an analytical expression for the absorption coefficient for a two-dimensional electron gas confined to a strip of width a [Eqs. (14) and (15)]. This is a simple example of a two-dimensional finite electronic quantum system. It should exhibit all features discussed in Ref. 17. At the same time numerical RPA calculations are feasible and allow us to discuss the accuracy of our analytical approach.

In the following it is assumed that $\Delta/\hbar < \omega \ll \omega_p, E_F/\hbar$

where E_F is the Fermi energy. It is furthermore assumed that $\lambda, \lambda_s \gg a \gg \lambda_F$ where λ is the wave length of the external radiation, λ_s is the skin depth and λ_F is the Fermi wavelength.

We consider a closed metallic quantum system placed in an external electric field $\boldsymbol{E}_{\rm ext}$ [with a time dependence $\propto \exp(i\omega t)$]. If the wavelength is much larger than the system size a, the external field is approximately constant throughout the system and (neglecting retardation effects) can be written as the gradient of an electric potential $\varphi_{\rm ext}(\boldsymbol{r}) = E_0 x \hat{\mathbf{e}}_x$ [$\boldsymbol{r} = (x,y,z)$ is a three-dimensional coordinate vector]. As is well known, within the RPA, the electronic response of the system to $\boldsymbol{E}_{\rm ext}$ is calculated by solving a set of self-consistent equations for the effective electrical potential $\varphi(\boldsymbol{r}) = \varphi_{\rm ext}(\boldsymbol{r}) + \delta\varphi(\boldsymbol{r})$ [$\delta\varphi(\boldsymbol{r})$ is the potential due to the induced charge density $\delta\varrho(\boldsymbol{r})$]

$$\delta\varphi(\boldsymbol{r}) = \int d\boldsymbol{r}'\, G(\boldsymbol{r},\boldsymbol{r}')\,\delta\varrho(\boldsymbol{r}'), \tag{1}$$

$$\delta\varrho(\boldsymbol{r}) = -\int d\boldsymbol{r}'\, \Pi_0(\boldsymbol{r},\boldsymbol{r}';\omega)\,\varphi(\boldsymbol{r})$$

with the boundary condition that $\delta\varphi(\boldsymbol{r})$ vanishes as $|\boldsymbol{r}| \to \infty$. $G(\boldsymbol{r},\boldsymbol{r}')$ is the Green function of the Laplace equation $\Delta G(\boldsymbol{r},\boldsymbol{r}') = -\epsilon_0^{-1}\delta(\boldsymbol{r}-\boldsymbol{r}')$, ϵ_0 is the dielectric constant. $\Pi_0(\boldsymbol{r},\boldsymbol{r}';\omega)$ is the nonlocal polarizability

$$\Pi_0(\boldsymbol{r},\boldsymbol{r}';\omega) = -2e^2 \sum_{\alpha,\beta} \frac{f(\varepsilon_\alpha) - f(\varepsilon_\beta)}{\varepsilon_\alpha - \varepsilon_\beta - \hbar\omega + i\gamma}\, \psi_\alpha^*(\boldsymbol{r}')\,\psi_\beta(\boldsymbol{r}') \times \psi_\beta^*(\boldsymbol{r})\,\psi_\alpha(\boldsymbol{r}), \tag{2}$$

e is the electron charge, ε_α and $\psi_\alpha(\boldsymbol{r})$ are the single-particle eigenvalues and eigenfunctions of the undisturbed system, they are usually calculated with in a Hartree-Fock or a local-density approximation. $f(\varepsilon) = \Theta(E_F - \varepsilon)$, and $\gamma > 0$ is smaller than Δ. Within the RPA, the absorption coefficient (proportional to the energy dissipation per unit time) may be written as[3]

$$\alpha(\omega) = \frac{\hbar\omega}{2E_0^2} \mathrm{Im}\, d(\omega), \tag{3}$$

where

$$d(\omega) = \int d\boldsymbol{r}\, d\boldsymbol{r}'\, \delta\varrho^*(\boldsymbol{r})\,\Pi_0(\boldsymbol{r},\boldsymbol{r}';\omega)\,\varphi(\boldsymbol{r}') \tag{4}$$

is the (complex) dipole moment and the asterisk denotes complex conjugation.

In the following we derive an explicit analytical expression for the absorption coefficient $\alpha(\omega)$ of a finite electronic quantum system in an external electric field, valid in the frequency range $\Delta/\hbar < \omega \ll \omega_p$. We make use of the approximations suggested in Ref. 20. First, according to Fermi's golden rule, the absorption coefficient is

$$\alpha(\omega) \simeq \frac{\pi\hbar^2 e^2 \omega^2}{2\Delta^2 E_0^2} \left| \langle \psi_\alpha | \varphi | \psi_\beta \rangle \right|_{\varepsilon_\alpha - \varepsilon_\beta \simeq \omega}^{\varepsilon_\alpha \simeq E_F}. \tag{5}$$

Second, the matrix elements of φ are evaluated within a semiclassical approximation.[18,21–24]. Third, φ itself is determined within a quasiclassical approximation: According to Eq. (1), the effective electric potential is given by (in symbolic notation)

$$\varphi = -\Pi_0^{-1}\delta\varrho. \tag{6}$$

In order to determine $\delta\varrho$, Eqs. (1) are usually solved numerically, using a real-space discretization[25] or by expanding in a suitable basis set. An approximate analytical solution may be obtained by noting that for $\omega \ll \omega_p$, $||G\Pi_0|| \gg 1$. In other words, $\delta\varrho$ is well approximated by the classical charge density $\delta\varrho_{\rm cl}$ of the metallic system subjected to an external potential $\varphi_{\rm ext}$,

$$\Delta\varphi_{\rm cl} = -\delta\varrho_{\rm cl}/\epsilon_0 \tag{7}$$

with $\varphi_{\rm cl}(\boldsymbol{r}) \to \varphi_{\rm ext}(\boldsymbol{r})$ as $|\boldsymbol{r}| \to \infty$ and $\varphi_{\rm cl} = 0$ within the system (in the classical limit, the external field is thus screened out completely). Equation (7) may be solved for $\delta\varrho_{\rm cl}$ using standard methods.[26] Fourth, Π_0 is determined within a quasiclassical approximation[27,20]

$$\Pi_0(\boldsymbol{r},\boldsymbol{r}';\omega) = e^2\nu_d[\,\delta(\boldsymbol{r}-\boldsymbol{r}') + i\omega P^{(d)}(\boldsymbol{r},\boldsymbol{r}';\omega)], \tag{8}$$

where ν_d is the density of states per unit volume in d dimensions and $P^{(d)}(\boldsymbol{r},\boldsymbol{r}';\omega)$ is the Fourier transform of the classical propagator $P^{(d)}(\boldsymbol{r},\boldsymbol{r}';t)$. In ballistic systems it is written as a sum over classical paths p from $\boldsymbol{r}$ to $\boldsymbol{r}'$

$$P^{(d)}(\boldsymbol{r},\boldsymbol{r}';\omega) = \sum_{\text{cl. paths } p} \left| \det\left[\frac{\partial(\boldsymbol{r}')}{\partial(\tau_p, \boldsymbol{n}_p)} \right] \right|^{-1} \exp(i\omega\tau_p). \tag{9}$$

Here τ_p is the time taken from $\boldsymbol{r}$ to $\boldsymbol{r}'$ along the path p, and $\boldsymbol{n}_p$ is a unit vector describing the direction of the initial velocity. The case of diffusive dynamics was discussed in Ref. 28.

We emphasize that the assumptions and approximations outlined above apply *independently of the nature of the electron dynamics*. In the following we show by comparison with quantum-mechanical RPA calculations that our approach provides a qualitative and quantitative description of the absorption spectrum in the frequency range $\Delta/\hbar < \omega \ll \omega_p$.

Two-dimensional strip. We consider a two-dimensional electron gas confined to a strip in the x-y plane surrounded by vacuum,[29] subject to a time-dependent electric field $\boldsymbol{E}_{\rm ext}$ directed along the negative x axis [compare Fig. 1(a)]. The width of the strip (along the x axis) is a, its length L (along the y axis), with $L \gg a$. Within the system, the electrons move ballistically, and they are specularly reflected at the boundaries at $x = \pm a/2$.

We write $\delta\varrho(\boldsymbol{r}) = \delta\sigma(x,y)\,\delta(z)$. For $L \gg a$, the surface-charge density $\delta\sigma$ depends on x only and the resolvent G is written as

$$G = \frac{1}{\epsilon_0} \int \frac{dq}{2\pi} \frac{1}{2|q|} e^{iq(x-x')} e^{-|q||z-z'|}. \tag{10}$$

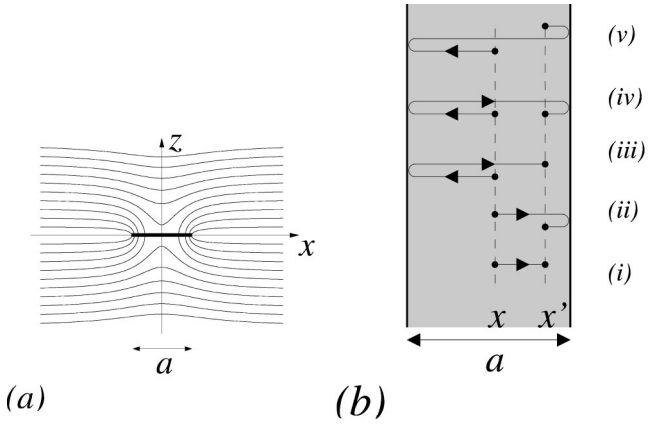

FIG. 1. (a) Electric field lines (in the x-z plane) for an infinitely long metallic strip of width a (in the x-y plane, oriented along the y axis) placed in a constant external electric field $\boldsymbol{E}_{\rm ext}=-E_0\hat{\mathbf{e}}_x$. (b) Classical paths from x to x' contributing to $\Lambda_0(x,x';\omega)$.

With

$$\frac{1}{L}\int dy dy' \Pi_0(\boldsymbol{r};\boldsymbol{r}';\omega)=\Lambda_0(x,x';\omega)\,\delta(z)\,\delta(z'), \quad (11)$$

the RPA equations (1) are reduced to a set of one-dimensional equations for $\varphi(x,z=0)$ with the kernel $\Lambda_0(x,x';\omega)$. We model the confinement in the x direction by introducing hard-wall boundary conditions. This is adequate in the range of parameters considered below and simplifies the quasiclassical analysis. We solve the resulting self-consistent equations numerically using a real-space discretization and obtain the absorption coefficient from Eq. (3).

The corresponding quasiclassical approximation for $\alpha(\omega)$ is obtained as described above: The classical surface-charge density is determined by solving Eq. (7) in elliptic cylinder coordinates: $\delta\sigma_{\rm cl}(x)=2\epsilon_0 E_0(a^2/4-x^2)^{-1/2}\Theta(|x|-a/2)$. The corresponding classical field lines are shown in Fig. 1(a). According to Eqs. (8),(9) the one-dimensional kernel $\Lambda_0(x,x';\omega)$ is given by a sum over classical paths as shown in Fig. 1(b) which may be summed by Poisson summation. Using Eq. (6) one obtains for the effective electric potential

$$\varphi(x)=\sum_{\mu>0}\varphi_\mu\cos[\mu\pi(x/a+1/2)], \quad (12)$$

$$\varphi_\mu=\frac{2\pi\epsilon_0 E_0}{e^2\nu_2}\frac{\sqrt{\omega^2-\omega_c^2\mu^2}}{\omega-\sqrt{\omega^2-\omega_c^2\mu^2}}\sin\left(\frac{\mu\pi}{2}\right)J_1\left(\frac{\mu\pi}{2}\right).$$

In the limit of $\omega\to 0$, $\varphi(x)$ tends to the linearized TF potential $\varphi_{\rm stat}(x,z=0)=(\epsilon_0 q_s)^{-1}\delta\sigma(x)$ where $q_S=e^2\nu_2/\epsilon_0$ is the two-dimensional TF screening vector. In the limit of large frequencies ($\omega\gg\omega_c$), $\varphi(x)$ tends to $\varphi_{\rm dyn}$ in the bulk of the sample, a dynamical contribution corresponding to a current building up the screening charges. It obeys $\partial^2\varphi_{\rm dyn}/\partial x^2=-(m_e\omega^2/e^2)\delta\sigma/\sigma_0$ where m_e is the electron mass and σ_0 is the areal charge density of the electrons. One may argue that

$$\varphi\simeq\varphi_{\rm stat}+\varphi_{\rm dyn}. \quad (13)$$

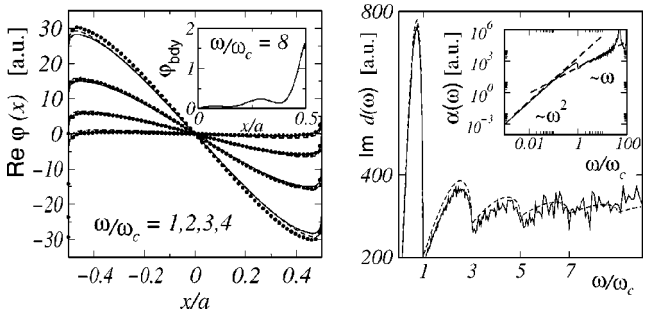

FIG. 2. Left: $\varphi(x)$ for a strip of width $a=10^3$ [a.u.] with $r_s=1$: quantum-mechanical results (•), analytical results according to Eq. (12) (—) and Eq. (13) (– – –). The inset shows the correction term $\varphi_{\rm bdy}(x)$. Right: Shows $\mathrm{Im}\,d(\omega)$ (for $a=10^4$ [a.u.] and $r_s=1$) as a function of ω: RPA result (—) and Eq. (14) (– – –). The inset shows $\alpha(\omega)$ as a function of ω: RPA result (—) and Eq. (15) (– – –). As usual $r_s=r_0/a_0$ where a_0 is the Bohr radius and r_0 is the length scale defined in terms of area per electron.

The intuitive ansatz (13) is justified by observing that the difference between Eqs. (13) and (12) is small except for x in a boundary layer of width $\delta x\propto v_F/\omega$. This difference is thus termed boundary contribution, $\varphi_{\rm bdy}$.

Figure 2(a) shows $\varphi(x)$ according to Eqs. (12) and (13) compared with the results of a numerical RPA calculation. One observes excellent agreement (and $\varphi_{\rm bdy}$ is small except at the boundary). Our results show that for larger frequencies ($\omega>\omega_c$), the dynamical potential $\varphi_{\rm dyn}$ makes a significant contribution to φ and dynamical screening effects cannot be neglected. For the absorption coefficient we obtain using Eqs. (5) and (12)

$$\alpha(\omega)\simeq\frac{\pi^2\hbar\epsilon_0^2 a}{e^2\nu_2}\frac{\omega^2}{\omega_c^2}\sum_{\mu>[\omega/\omega_c]}^{\rm odd}\frac{\sqrt{\mu^2\omega_c^2-\omega^2}}{\mu^2}J_1^2(\mu\pi/2) \quad (14)$$

with limiting forms

$$\alpha(\omega)=\begin{cases}\pi^2 C\hbar\epsilon_0^2 a\omega^2/(e^2\nu_2\omega_c) & \text{for } \omega\ll\omega_c,\\ \pi\hbar\epsilon_0^2 a\omega/(4e^2\nu_2) & \text{for } \omega\gg\omega_c\end{cases} \quad (15)$$

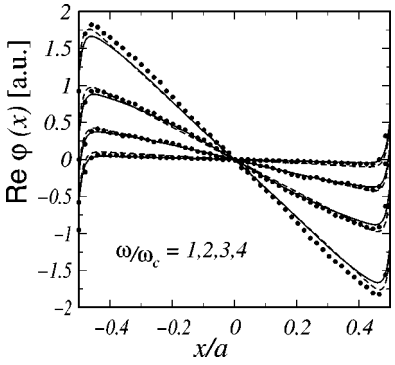

FIG. 3. Shows $\varphi(x)$ for a thin film of width $a=50$ [a.u.] with $r_s=1$: RPA results (•), analytical results according to Eq. (6) (—) and using $\varphi\simeq\varphi_{\rm stat}+\varphi_{\rm dyn}$ (– – –).

and $C \simeq 0.12$. In Fig. 2(b), we show quantum-mechanical RPA results in comparison with Eqs. (14) and (15) and observe excellent agreement. We observe prominent resonances in the absorption coefficient near odd multiples of ω_c, due to single-particle cyclotron orbits (electrons moving in phase with the external field). This establishes that the single-particle resonances conjectured in Ref. 17 exist within the RPA. Their positions, strengths and shapes are very well described by Eq. (14). Equation (15) and the inset of Fig. 2(b) show that in the limit of large frequencies ($\omega \gg \omega_c$), the absorption coefficient is linear in ω. In the opposite limit ($\omega \ll \omega_c$) where the TF approach is adequate, it is quadratic. We conclude that the quasiclassical approximation described above, for the parameters considered here, provides a quantitative description of the optical properties.

Three-dimensional thin film. To conclude we discuss φ for a thin film[30] of width a in the y-z plane subject to an external potential $\varphi_{\rm ext}(\boldsymbol{r}) = E_0 x \hat{\mathbf{e}}_x$. The classical charge density is concentrated at the boundary, $\delta\varrho_{\rm cl}(x) = \pm\, \delta\sigma\, \delta(x \pm a/2)$. The corresponding static and dynamical contributions to $\varphi(x)$ are singular; we thus use the TF charge density[6] instead of $\delta\varrho_{\rm cl}$ (appropriate in the limit of small k_s corresponding to high electron densities): $\delta\varrho_{\rm TF}(x) = k_s \epsilon_0 E_0 \sinh(k_s x)/\cosh(k_s a/2)$. Here $k_s^2 = e^2 \nu_3/\epsilon_0$ is the three-dimensional TF screening vector. The RPA equations are easily solved within a real-space discretization. Our results for $\varphi(x)$ are shown in Fig. 3, and compared to results of the analytical approach using Eqs. (5)–(9).

We have also calculated the absorption coefficient for $\Delta/\hbar < \omega \ll \omega_p$ within the RPA. The analytical approach must be used with caution in the case of the film since it requires that φ be smooth on the scale of λ_F. It turns out that $\alpha(\omega)$ is, to a good approximation, quadratic in ω as opposed to the two-dimensional case. As in $d=2$ dimensions we observe resonances near odd multiples of ω_c (not shown).

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