Raman scattering in a two-dimensional electron gas: Boltzmann equation approach

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The inelastic light scattering in a two-dimensional electron gas is studied theoretically using the Boltzmann equation techniques. Electron-hole excitations produce the Raman spectrum essentially different from the one predicted for the 3D case. In the clean limit it has the form of a strong nonsymmetric resonance due to the square-root singularity at the electron-hole frequency $\omega = vk$, while in the opposite dirty limit the usual Lorentzian shape of the cross section is reestablished. The effects of electromagnetic field are considered self-consistently, and the contribution from collective plasmon modes is found. It is shown that unlike 3D metals where plasmon excitations are unobservable (because of very large required transferred frequencies), the two-dimensional electron system gives rise to a low-frequency ($\omega \propto k^{1/2}$) plasmon peak. A measurement of the width of this peak can provide data on the magnitude of the electron-scattering rate.
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Raman scattering is a powerful method for experimental studies of elementary excitations in various structures. In particular, high-T_c superconductors produce Raman spectra that remain mysterious over a broad region of frequency. Namely, the high-frequency continuum,[1,2] 2Δ peak (see, e.g., Ref. 3), two-magnon spectra, revealing strong mutual influence between antiferromagnetism and superconductivity,[4] still does not have a robust self-consistent theoretical description. Therefore, the development of the theory of Raman scattering from different excitations is still of considerable current interest.

Here we are interested in the Raman scattering from excitations of a two-dimensional (2D) normal electron system, namely, from electron-hole pairs and collective plasmon excitations. We show that Raman-scattering cross section in 2D systems differs from the spectrum of a 3D metal in two aspects. First, the scattering from electron-hole pairs becomes more singular (due to the square-root singularity in the density of states). A finite strength of the electron-hole contribution is determined by the electron-scattering rate. Second, as soon as the plasmon spectrum is gapless ($\omega \propto k^{1/2}$), a corresponding peak is located in the reasonably low-frequency (~ 10 meV) range.

Inelastic light scattering in 2D systems has been extensively used in investigations of the excitations of the Fermi sea,[5] the energy gap in the fractional quantum Hall regime,[6] exciton states,[7] spin-density, and charge-density excitations.[8] Experimental evidence of 2D plasmons in GaAs heterostructures comes from the Raman spectroscopy measurements in a magnetic field.[9] By varying the direction of magnetic field it is possible to distinguish Raman response of a 2D electron system from a contribution of a background.[10] The $k^{1/2}$ spectrum is clearly observed,[9] however, no data are available for the dependence of a line shape of the plasmon peak on the electron-scattering rate.

The standard quantum-mechanical theory of Raman scattering in electron systems applies the Green-function formalism. We use in the present paper a different approach, based on the Boltzmann equation. Such a semiclassical method is valid[11] when the characteristic scale of transferred light momentum is less than the Fermi momentum of a 2D electron gas. For the typical situation of GaAs/Al$_x$Ga$_{1-x}$As heterojunction, the concentration of carriers[6,9] is of the order of $1-6\times 10^{11}$ cm^{-2} and corresponds to the Fermi momentum value of $\sim 3-8\times 10^5$ cm^{-1}. On the other hand, the typical values of the momentum transfer are considerably lower, $\sim 0.2-1\times 10^5$ cm^{-1}. The main advantage of the kinetic approach is a possibility to include effects of electron scattering and to account for electromagnetic field in a self-consistent manner by solving simultaneously the Maxwell equation. This gives a plasmon contribution together with the electron-hole continuum. It is also possible to generalize this theory easily to the case of external magnetic field applied.

The system under study is shown in Fig. 1. Two-dimensional electron gas ($z=0$ plane) is embedded into a host material with the dielectric constant ϵ. The polarization of incident (i) and, hence, of scattered (s) light waves are assumed to be parallel to the plane $z=0$. The effective Hamiltonian describing Raman scattering from electronic fluctuations is bilinear in the vector potential of light,

$$H_{\rm eff} = \frac{e^2}{2mc^2}\int d^2s\, \delta n_\gamma(\mathbf{s},t)\mathbf{A}^2(\mathbf{s},t), \qquad (1)$$

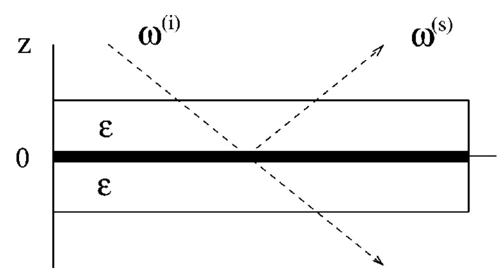

FIG. 1. Geometry of the light scattering. Thick solid line ($z=0$ plane) represents the electron gas embedded into a host material with the dielectric constant ϵ. The incident (i) light induces the in-plane 2D current, which in its turn produces a scattered (s) wave with different frequency.

where the fluctuation δn_γ expressed via the nonequilibrium partition function $\delta f_p(\mathbf{s},t)$,

$$\delta n_\gamma(\mathbf{s},t)=\int \frac{2d^2p}{(2\pi)^2}\gamma_p \delta f_p(\mathbf{s},t), \tag{2}$$

differs from the usual electronic density only by the anisotropic dimensionless factor γ_p (electron-light vertex). This factor depends on the light polarization and accounts for the virtual interband transitions. Its exact form is not essential for the following (see Ref. 14).

Varying the expression (1) over the vector potential $\mathbf{A}$, we obtain the electron current induced by the incident light with the frequency $\omega^{(i)}$ and the in-plane wave vector $\mathbf{k}_s^{(i)}$,

$$\mathbf{j}^{(i)}(\mathbf{s},t)=-\frac{e^2}{mc}\delta n_\gamma(\mathbf{s},t)\mathbf{A}^{(i)}\exp(-i\omega^{(i)}t+i\mathbf{k}_s^{(i)}\mathbf{s}). \tag{3}$$

The 2D current [Eq. (3)] produces a scattered electromagnetic wave with different frequency $\omega^{(s)}$ and wave vector $\mathbf{k}_s^{(s)}$. The solution of the corresponding nonuniform Maxwell equation is straightforward. After some simple calculations, it gives for the amplitude of light scattered into the half-space $z>0$ the expression

$$A(\omega^{(s)},\mathbf{k}_s^{(s)})=\frac{2\pi i e^2\mathbf{A}^{(i)}}{mc^2k_z^{(s)}}\delta n_\gamma(\omega,\mathbf{k}_s), \tag{4}$$

where $k_z^{(s)2}=\epsilon\omega^{(s)2}/c^2-\mathbf{k}_s^{(s)2}$; the Fourier component of the density fluctuations depends on the transferred energy $\omega=\omega^{(i)}-\omega^{(s)}$ and momentum $\mathbf{k}_s=\mathbf{k}_s^{(i)}-\mathbf{k}_s^{(s)}$.

The Raman-scattering cross section defined as the normalized energy $\langle\langle|\omega^{(s)}\mathbf{A}^{(s)}/\omega^{(i)}\mathbf{A}^{(i)}|^2\rangle\rangle$ related to the interval $d\omega^{(s)}d^2\mathbf{k}_s^{(s)}/(2\pi)^3$ has the form

$$\frac{d^2\sigma}{d\omega^{(s)}do^{(s)}}=\frac{\epsilon^{1/2}e^4}{2\pi m^2c^5}\frac{\omega^{(s)3}}{\omega^{(i)2}k_z^{(s)}}K(\omega,\mathbf{k}_s), \tag{5}$$

where $K(\omega,\mathbf{k}_s)$ is the Fourier component of the correlator of density fluctuations

$$K(\mathbf{s}-\mathbf{s}',t-t')=\langle\langle\delta n_\gamma(\mathbf{s},t)\,\delta n_\gamma(\mathbf{s}',t')\rangle\rangle.$$

One can argue that the expression (5) diverges as the direction of scattered light approaches the electron plane: $k_z^{(s)}\to 0$. In fact, this means that as soon as the ''width'' of a two-dimensional system l is assumed to be the smallest one of all of the characteristic lengths of the problem, we are restricted to the limit $k_z^{(s)}\gg l^{-1}$.

To evaluate this correlator we apply the fluctuation-dissipation theorem that expresses it via the imaginary part of the generalized response $\delta n_\gamma(\omega,\mathbf{k})$ to an arbitrary external potential $U(\omega,\mathbf{k})$ (in what follows we omit the subscript s),

$$K(\omega,\mathbf{k})=-\frac{2}{1-\exp(-\omega/T)}\,\mathrm{Im}\left(\frac{\delta n_\gamma(\omega,\mathbf{k})}{U(\omega,\mathbf{k})}\right). \tag{6}$$

The most simple way to derive the generalized response (6) is to make use of the linearized Boltzmann equation for the nonequilibrium part of the distribution function $\delta f_p=\chi_p\partial f_0/\partial\varepsilon$,[12,13]

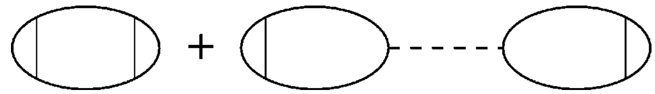

FIG. 2. Diagrammatic representation of the density-density correlation function (10). The solid lines represent electron propagators, the dashed line stands for the electromagnetic Green function, and the empty vertex for the effective electron-light interaction.

$$-i(\omega-\mathbf{kv}+i\tau^{-1})\chi_p(\mathbf{k},\omega)=i\omega\gamma_pU(\mathbf{k},\omega)-e\mathbf{vE}(\mathbf{k},\omega), \tag{7}$$

where $f_0(\varepsilon)$ is the local-equilibrium Fermi-Dirac partition function. The second term on the right-hand side of Eq. (7) accounts for the fluctuating electromagnetic field $\mathbf{E}$. It satisfies the Maxwell equation with the nonequilibrium electric current determined from Eq. (7),

$$\mathrm{rot\,rot}\,\mathbf{E}(z,\mathbf{s},\omega)-\frac{\epsilon\omega^2}{c^2}\mathbf{E}(z,\mathbf{s},\omega)=-\frac{4\pi ie\omega}{c^2}\delta(z)\times\langle\mathbf{v}\chi_p(\mathbf{s},\omega)\rangle. \tag{8}$$

Here the angular brackets denote the integral over the Fermi line

$$\langle\cdots\rangle=\int\frac{2dp_F}{v(2\pi)^2}(\cdots).$$

We are interested in the solution of Eq. (8) at $z=0$. The straightforward derivation gives for the Fourier component of the electric field

$$\mathbf{E}(z=0,\mathbf{k},\omega)=\frac{2\pi ie}{\epsilon\omega}\sqrt{k^2-\epsilon\omega^2/c^2}\langle\mathbf{v}\chi_p(\mathbf{k},\omega)\rangle. \tag{9}$$

Substituting the solution (9) into the Boltzmann equation (7) one gets the integral equation for the electronic density fluctuation $\chi_p(\mathbf{k},\omega)$. Such an equation has a simple solution, which after the substitution into Eq. (2) and then into the fluctuation-dissipation theorem (6), gives the Raman cross section. Finally, we obtain (see Fig. 2)

$$K(\mathbf{k},\omega)\propto-\mathrm{Im}\left\langle\frac{\omega\gamma_p^2}{\omega-\mathbf{kv}+i\tau^{-1}}\right\rangle+\mathrm{Im}\,F_\alpha(\mathbf{k},\omega)D_{\alpha\beta}(\mathbf{k},\omega)F_\beta(\mathbf{k},\omega), \tag{10}$$

where the proportionality coefficient (Bose factor) is omitted, see Eq. (6); $D(\mathbf{k},\omega)$ is the two-dimensional electromagnetic Green function,

$$D^{-1}_{\alpha\beta}(\mathbf{k},\omega)=\frac{1}{\omega}\left\langle\frac{v_\alpha v_\beta}{\omega-\mathbf{kv}+i\tau^{-1}}\right\rangle-\frac{\epsilon\delta_{\alpha\beta}}{2\pi e^2\sqrt{k^2-\epsilon\omega^2/c^2}}, \tag{11}$$

and $F_\alpha(\mathbf{k},\omega)$ is the oscillator strength,

$$F_\alpha(\mathbf{k},\omega)=\left\langle\frac{v_\alpha\gamma_p}{\omega-\mathbf{kv}+i\tau^{-1}}\right\rangle. \tag{12}$$

We devote the rest of this paper to the discussion of different terms in the Raman cross section [Eq. (10)].

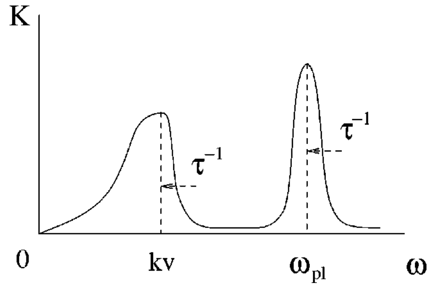

FIG. 3. Raman-scattering cross section. The nonsymmetric resonance is located at $\omega = kv$. Its width at $\omega > kv$ as well as its height are determined by the finite value of the scattering rate τ^{-1}. The second symmetric resonance corresponds to excitation of plasmon.

The first term represents the scattering from the electron-hole pairs. For the estimates we suppose the Fermi line to be isotropic, i.e.,

$$K_{e\text{-}h}(k,\omega) \propto -\frac{m\gamma^2}{\pi}\mathrm{Im}\frac{\omega}{\sqrt{(\omega+i\tau^{-1})^2-k^2v^2}}. \tag{13}$$

In the dirty limit $kv\tau \ll 1$ (the so-called zero-momentum transfer limit), one can neglect v^2k^2 in the denominator. The cross section then takes the same well-known Lorentzian form as in the 3D case,[15] $\sim \omega\tau/(\omega^2\tau^2+1)$.

In the clean limit $kv\tau \gg 1$, the expression (13) has a square-root singularity at $\omega = kv$ rather than a steplike one (as in the 3D case). It results in the strong nonsymmetric resonance (see Fig. 3); the finite height of this resonance is controlled by the scattering rate τ^{-1}. For anisotropic Fermi line, the resonance location is defined by the maximum value of electron velocity along the momentum transfer $\omega = \mathbf{kv}_{\max}$.

The second term in Eq. (10) represents effects of Coulomb interaction and collective electron excitations, namely, 2D plasmons. This term is important in the clean limit. At low transferred frequencies $\omega \ll kv$, it results in the screening of the isotropic scattering channel through the renormalization of the electron-light vertex $\gamma_p \rightarrow \gamma_p - \langle \gamma_p \rangle / \langle 1 \rangle$, similar to the usual 3D case.[12,13]

At high transferred frequencies $\omega \gg kv$, the second term in Eq. (10) gives a 2D plasmon peak located at the plasmon frequency $\omega_{pl}(k)$, which is determined by the dispersion equation

$$\omega^2 = \frac{2\pi e^2}{\epsilon}\langle v_k^2 \rangle \sqrt{k^2 - \epsilon\omega^2/c^2}, \tag{14}$$

where v_k means the component of electron velocity in the k direction. The typical momenta transfer k are of the order of light momenta. Hence from the formula (14) one can see that $\omega \gg vk$ proving the initial assumption was correct. We can also omit the term $\epsilon\omega^2/c^2$ (this term cares for the finite plasmon velocity) in Eq. (14) in comparison with the term k^2 due to the fact that $c^2 k \gg v^2 p_F$ for typical values of k. Indeed, this means that it is enough to use the Poisson equation for electromagnetic fluctuations instead of the Maxwell equation (8). The only difference occurs at very small transferred momenta where the Poisson equation gives the infinite plasmon velocity in the limit $k \rightarrow 0$.

The formula (14) is valid only if the plasmon wavelength becomes large compared to the layer thickness, $k \gg l^{-1}$. If this condition is violated, the three-dimensional problem has to be solved with boundary conditions satisfied on both sides of the layer. Its solution gives the expression

$$\omega_{pl}^2(k) = \frac{4\pi e^2}{\epsilon}\sqrt{\langle v_k^2 \rangle \langle v_z^2 \rangle}\ \tanh\left(\sqrt{\frac{\langle v_k^2 \rangle}{\langle v_z^2 \rangle}}\frac{kl}{2}\right),$$

where the electron velocity along the perpendicular direction v_z appears. Note that the angular brackets now denote the integral over the three-dimensional Fermi surface. When $l \rightarrow 0$, this expression reduces to Eq. (14) and for $l \rightarrow \infty$, it gives the frequency of ordinary 3D plasmon.

Near the plasmon resonance the Raman cross section has the symmetric Lorentzian line shape

$$K_{pl}(k,\omega) \propto \frac{m\gamma^2}{8\pi\omega}\frac{k^2 v^2 \tau^{-1}}{[\omega_{pl}(k)-\omega]^2 + \tau^{-2}/4}. \tag{15}$$

The relative height of two resonances (13) and (15) is $K_{e\text{-}h}/K_{pl} \sim k^{3/2} v^{3/2} \tau^{1/2}/\omega_{pl}$ and can be either more or less than unity depending on the momentum transfer k and scattering rate τ^{-1}.

In conclusion, we have calculated the Raman-scattering intensity from two-dimensional electronic fluctuations. The main distinctive features from a usual three-dimensional metal are the more singular electron-hole contribution (13) and low-frequency plasmon resonance (15). The electronic Raman scattering in a 2D system in a transverse magnetic field can be considered studied the same Boltzmann equation technique as it has been done for the 3D electron system.[16]

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