Inelastic scattering of light by two-dimensional electrons with large momentum transfer

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The inelastic scattering of light by density fluctuations of a two-dimensional electron gas involving a large transfer of energy when retardation effects become important is investigated. Along with the plasma peak in the scattering cross section, there is a wing due to the decay of the incident photon into two photons. Like the plasma contribution to the scattering, the predicted wing is significantly enhanced under conditions of resonance with the interband transition.

In an electron plasma there exist two types of Raman scattering of light:[1)] single-particle scattering with a frequency shift of the order of kv_F, where k is the momentum transfer and v_F is the Fermi velocity (it is a question of degenerate electrons) and scattering due to collective effects. The latter are connected with the density fluctuations of the electron gas, and the corresponding frequency shift is equal to the plasmon frequency ω_p.

In an isotropic three-dimensional plasma longitudinal and transverse waves separate completely, wherefore an account of retardation effects gives only small corrections to the scattering cross section—of the order of v_F/c for the single-particle mechanism, and ω_p/ck for the collective one (c is the speed of light in the medium).

Spatial inhomogeneity or anisotropy of the plasma leads to a coupling of the longitudinal and transverse waves. In the present communication we will show that for a two-dimensional electron plasma there arises an additional contribution to the inelastic light scattering in the region of large energy transfers ω, where $\omega \geqslant ck_\parallel$, and $k_\parallel$ is the momentum transfer in the plane of the system. In this frequency region ω the light-induced excitations in the plasma are no longer quasistatic.

In an isotropic three-dimensional plasma there are purely transverse electromagnetic waves with frequency $\omega = (\omega_p^2 + (ck)^2)^{1/2}$, and also plasmons. During inelastic light scattering excitation of a plasmon is possible, but excitation of a transverse wave is kinematically forbidden.

A new interesting situation arises during scattering in a two-dimensional plasma. In this case only the parallel component of the momentum is conserved. The scattering spectrum of two-dimensional electrons, as will be shown below, contains besides the peak at the plasmon frequency a wide wing at $\omega \geqslant ck_\parallel$, corresponding to the decay of the incident photon $\omega_1 = ck_1$ into a scattered photon $\omega_2 = ck_2$ and a third photon with frequency ω and wave vector $(\mathbf{k}_\parallel, k_z)$, $k_z = (\omega^2/c^2 - k_\parallel^2)^{1/2}$. It can be easily seen that such a process is kinematically possible. (For example, in a planar waveguide of width a the dispersion of the photon has the form

$$\omega = c\left(k_\parallel{}^2 + \left(\frac{n\pi}{a}\right)^2\right)^{1/2},$$

and the corresponding curve is convex down, i.e., the spectrum is decay-like.

Let us estimate the characteristic width of the photon decay spectrum for interaction with a two-dimensional plasma. The most effective coupling of the photons with the plasmons is determined by the condition $ck_\parallel \sim \omega_p(k_\parallel)$, where

$$\omega_p = \left(\frac{2\pi e^2 N_s}{\varepsilon m_e} k_\parallel\right)^{1/2}$$

is the two-dimensional plasma frequency; N_s and m_e are the electron density and effective electron mass; and ε is the background dielectric constant. This condition determines the characteristic values $k_\parallel \sim k'$, and the corresponding width of the decay spectrum is given by

$$ck' \sim \frac{2\pi e^2 N_s}{\varepsilon m_e c}.$$

Obviously, this width competes with collisional broadening $\sim 1/\tau$, where τ is the relaxation time of the electrons. The effect which we are considering here is important when the inequality $ck' > 1/\tau$ is fulfilled, which is equivalent to the condition $2\pi\sigma_0/\varepsilon > c$, where σ_0 is the conductivity of the two-dimensional gas ($\sigma_0 = e^2 N_s \tau/m_e$).

To calculate the cross section we investigate the equilibrium fluctuations of the density ρ_s of the two-dimensional electron plasma. It is possible to calculate the density-density correlator in terms of a generalized susceptibility $\alpha_{k\parallel}(\omega)$ according to the fluctuation-dissipation theorem:[1,2]

$$\langle \rho_s{}^2 \rangle_\omega = 2\,[n(\omega) + {}^1/_2]\,\mathrm{Im}\,\alpha_{k_\parallel}(\omega),\ \hbar = 1, \tag{1}$$

where $n(\omega)$ is the Bose distribution.

The generalized susceptibility is found as the response function to a perturbation of the form

$$\hat{V} = -e \int \hat{\rho}_s(\mathbf{r}) f(\mathbf{r}, t)\, d^3\mathbf{r}, \tag{2}$$

where e is the charge of the electron, f is the generalized force (in the present case a scalar field), $\mathbf{r} = (\mathbf{x}, z)$, $\mathbf{x}$ is the position vector in the plane of the system, z is the perpendicular coordinate, and the electrons are located at $z = 0$. The density perturbation in the Fourier representation with respect to the time t and the position vector $\mathbf{x}$ is then

$$\rho(k_\parallel, \omega) = \alpha_{k_\parallel}(\omega)\, e f(\mathbf{k}_\parallel, \omega)\, \delta(z). \tag{3}$$

The material equations give the connection between the current density and the field:

$$\mathbf{j}(\mathbf{k}_\parallel, \omega) = \sigma[\mathbf{E}(\mathbf{k}_\parallel, \omega) + i\mathbf{k}_\parallel f(\mathbf{k}_\parallel, \omega)]\,\delta(z), \tag{4}$$

where σ is the conductivity, $\mathbf{E}(\mathbf{k}_\parallel, \omega)$ is the induced electric field parallel to the plasma layer at $z = 0$, which is found from the Maxwell equations for the potentials φ and $\mathbf{A}$. In

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these equations the charge and current densities are proportional to $\delta(z)$. Satisfying the corresponding boundary matching conditions, we find (at $z=0$)

$$\mathbf{E}(\mathbf{k}_\parallel,\omega)=-i\mathbf{k}_\parallel\varphi+\frac{i\omega}{c_0}\mathbf{A}_\parallel, \tag{5}$$

$$\varphi=\frac{2\pi}{\varepsilon R}e\rho(\mathbf{k}_\parallel,\omega),\quad \mathbf{A}=\frac{2\pi}{c_0R}\mathbf{j}(\mathbf{k}_\parallel,\omega),\quad R=\left(k_\parallel^2-\frac{\omega^2}{c^2}\right)^{1/2}, \tag{6}$$

where $c=c_0/\varepsilon^{1/2}$ and c_0 is the speed of light in a vacuum.

Using the continuity equation, we obtain the response function in the limit $\omega\gg k_\parallel v_F$, which corresponds to a "cold" plasma:

$$\alpha_{k_\parallel}(\omega)=\frac{i\sigma k_\parallel^2}{e^2(\omega+2\pi i\sigma R/\varepsilon)},\quad \sigma=\frac{e^2N_s\tau}{m_e(1-i\omega\tau)}. \tag{7}$$

It is now clear that in the absence of a three-dimensional case, where the generalized susceptibility as a function of ω has only a simple pole at the plasma frequency, in the situation under consideration in addition to the plasma pole there is also a branch cut, beginning at $\omega=ck_\parallel$, where $R=0$. The additional contribution to the Raman scattering cross section, which was mentioned in the Introduction, is due specifically to this branch cut.

From Eqs. (1) and (7) we obtain the density correlator:

$$\langle\rho^2\rangle_\omega=2\left[n(\omega)+\frac{1}{2}\right]F(k_\parallel,\omega),$$

$$F(k_\parallel,\omega)=\frac{\omega_p^2k_\parallel}{2\pi e^2}\left[\frac{\omega\nu}{\omega^2\nu^2+\{\omega^2-\omega_p^2[1-\omega^2/(ck_\parallel)^2]^{1/2}\}^2}\times\theta((ck_\parallel)^2-\omega^2)\right.$$
$$\left.+\frac{\omega\nu+\omega_p^2(\omega^2/(ck_\parallel)^2-1)^{1/2}\operatorname{sign}\omega}{\omega^4+\{\omega\nu+\omega_p^2[\omega^2/(ck_\parallel)^2-1]^{1/2}\operatorname{sign}\omega\}^2}\theta(\omega^2-(ck_\parallel)^2)\right]. \tag{8}$$

Here $\nu=1/\tau$, $\theta(x)=1$ for $x>0$, and $\theta(x)=0$ for $x<0$.

The first term in the expression for F corresponds to plasmons with retardation taken into account. In the collisionless approximation ($\nu\to0$) it changes over into $\delta(\omega^2-\Omega_p^2)$, where

$$\Omega_p^2=-\frac{\omega_p^4}{2(ck_\parallel)^2}+\left[\frac{\omega_p^8}{4(ck_\parallel)^4}+\omega_p^4\right]^{1/2} \tag{8a}$$

is the dispersion law of two-dimensional plasmons.

The second component, which is different from zero for $\omega\geqslant ck_\parallel$ is due to the photons which are in thermal equilibrium with the system of electrons. We explain this in the following way. From the Green's function of the photon in the dielectric (with dielectric constant ε) we can obtain an expression for the field correlators:[2]

$$\langle E_iE_j\rangle_{\omega,\mathbf{k}}=\frac{\omega^2}{c^2}2\left[n(\omega)+\frac{1}{2}\right]\operatorname{Im}\frac{4\pi}{\omega^2/c^2-k^2+i0}\left(\delta_{ij}-\frac{c^2k_ik_j}{\omega^2}\right). \tag{9}$$

Let a gas of two-dimensional electrons with conductivity σ be present in a dielectric at $z=0$. In the region of weak coupling between the plasma waves and the electromagnetic waves $\omega_p\ll ck_\parallel$ (i.e., without account of self-consistent effects) it is possible then to find the current correlator ($j_\parallel=\sigma E_\parallel$) and then from the continuity equation obtain the density correlator:

$$\langle\rho_s^2\rangle_\omega=\frac{k_{\parallel i}k_{\parallel j}}{e^2\omega^2}\langle j_{\parallel i}j_{\parallel j}\rangle|_{z=0}=\frac{\sigma\sigma^*}{e^2\omega^2}k_{\parallel i}k_{\parallel j}\langle E_{\parallel i}E_{\parallel j}\rangle|_{z=0}. \tag{10}$$

Here for simplicity σ does not take account of collisions: $\sigma=ie^2N_s/m_e\omega$. Integrating Eq. (9) over k_z, we find the correlator $\langle E_{\parallel i}E_{\parallel j}\rangle$ at $z=0$ and we obtain

$$\langle\rho_s^2\rangle_\omega=2\left[n(\omega)+\frac{1}{2}\right]\frac{\omega_p^4k_\parallel}{2\pi e^2}\frac{[\omega^2/(ck_\parallel)^2-1]^{1/2}}{\omega^4}\times\operatorname{sign}\omega\,\theta(\omega^2-(ck_\parallel)^2), \tag{11}$$

which is exactly equal to the limit of Eq. (8) for $\nu\to0$ and $\omega_p\ll ck_\parallel$ in the region $\omega\geqslant ck_\parallel$.

In the case of strong collisions ($\sigma_0\ll c$) it is possible to neglect the terms in formula (8) that contain radicals, after which the θ-functions "join together," and all that remains, as expected, is the plasma contribution.

The light scattering cross section, as is known,[3,4] is determined by the density-density correlator. Therefore for the case of a two-dimensional plasma we can at once write

$$\frac{d\sigma}{d\Omega\,d\omega}=\frac{1}{\pi}\frac{\omega_1}{\omega_2}\left(\frac{e^2}{m_ec_0^2}\right)^2[n(\omega)+1](\mathbf{e}_{1,\parallel}\mathbf{e}_{2,\parallel})^2F(k_\parallel,\omega). \tag{12}$$

Here the indices 1 and 2 denote the incident and scattered waves, respectively, $\mathbf{e}_\parallel$ is the projection of the polarization vector on the plane $z=0$. Thus, besides the peak at the plasma frequency, the light scattering spectrum in the limit $\nu\to0$ contains a wide wing beginning at $\omega=ck_\parallel$ and the described photon decay.

Let us analyze expression (12) in the collisionless limit ($\nu\to0$). First let us consider the kinematics of light scattering by a two-dimensional system in backscattering geometry for the Stokes region of the spectrum ($\omega>0$).

At the fixed angles φ_1 and φ_2 respectively for the wave vectors of the incident and scattered light relative to the normal to the plane of the two-dimensional plasma (we assume that $\mathbf{k}_1$ and $\mathbf{k}_2$ and the normal lie in one plane) $k_\parallel$ is a function of ω. According to the laws of conservation of parallel momentum and energy we have

$$k_\parallel=k_1(\sin\varphi_1-\sin\varphi_2)+(\sin\varphi_2)\,\omega/c.$$

This expression must be substituted into Eq. (12) in order to obtain the scattering cross section as a function of ω. Decay of the photon ω_1 is allowed for $\omega\geqslant\omega_0$, where ω_0 is found from the equation $\omega_0=ck_\parallel(\omega_0)$:

$$\omega_0=\begin{cases}\omega_1\dfrac{\sin\varphi_1-\sin\varphi_2}{1-\sin\varphi_2}, & \varphi_1\geqslant\varphi_2,\\[2ex] \omega_1\dfrac{\sin\varphi_2-\sin\varphi_1}{1+\sin\varphi_2}, & \varphi_1\leqslant\varphi_2.\end{cases} \tag{13}$$

Thus, the region of values of ω where photon decay is allowed occupies the interval $\omega_0\leqslant\omega\leqslant\omega_1$; the condition $\omega\geqslant ck_\parallel(\omega)$ is of course satisfied here.

The condition of small energy transfer $\omega_0\ll\omega_1$, which is traditional for the presently available experiments, is realized at angles close to specular ($|\sin(\varphi_1-\varphi_2)|\ll1$). The scattering cross section in this case behaves at large frequencies ($\omega\to\omega_1$) like ω^{-1}, but near the threshold it grows according to the law $[(\omega-\omega_0)^{1/2}+\text{const}]$.

The ratio of the total intensities of scattering by the photons $I_{\rm ph}$ and the plasmons $I_{\rm pl}$ is obtained from Eq. (12). At not too close angles φ_1 and φ_2 (when $\omega_p(q) < cq$), where $q \sim k_1 |\sin\varphi_1 - \sin\varphi_2|$ is the characteristic momentum transfer, we have

$$\frac{I_{\rm ph}}{I_{\rm pl}} \sim \left(\frac{\omega_p(q)}{cq}\right)^3 \ln\frac{k_1}{q}. \tag{14}$$

In the opposite limit ($\omega_p(q) > cq$) the intensity of the wing is greater than the intensity of the plasma peak:

$$I_{\rm ph}/I_{\rm pl} = [\omega_p(q)/cq]^2.$$

Note that this case requires for its realization quite large dimensions L of the interaction region of the light beam with the electrons. Thus, for GaAs–GaAlAs structures at $N_s = 3\cdot 10^{11}$ cm^{-2} it is necessary that $L > 1$ cm [then the obvious condition $qL > 1$ coincides with the condition $\omega_p(q) > cq$].

In the mirror channel ($\varphi_1 = \varphi_2 = \varphi$) scattering by a plasmon is kinematically forbidden, but decay of a photon is allowed, starting with zero energy transfer, since $\omega_0 = 0$. The cross section at $\varphi_1 = \varphi_2$ is described by the expression [see Eq. (8)]

$$S \propto [n(\omega)+1]\frac{\omega}{\omega^2+\omega'^2}, \quad \omega' = \frac{2\pi e^2 N_s}{\varepsilon m_e c}\cos\varphi. \tag{15}$$

Thus, in the collisionless limit, which we are considering here, the scattering line width ω' is determined only by photon decay.[2)]

The quantity $\omega' \sim \omega_p^2/ck_\parallel$ in the more realistic case $\omega_p \ll ck_\parallel$ is small ($\omega' \ll \omega_p$). In the region of small energy transfer collisional single-particle scattering is a competing mechanism. The width of the corresponding spectral interval is of the order of ν, and if the condition $2\pi\sigma_0/\varepsilon \gg c$ is satisfied we obtain $\omega' \gg \nu$.

The condition $2\pi\sigma_0/\varepsilon > c$ (the two-dimensional conductivity is greater than the speed of light), as is well known,[5,6] defines a regime in which relaxation processes in the two-dimensional plasma can no longer be considered as quasistatic, i.e., it is necessary to take retardation effects into account (vortex fields, etc.).

Thus, in the region of frequencies ω much smaller than the plasmon frequency, the effect considered here is accessible for observation even in the mirror channel in the case of samples with sufficiently high electron mobility.

To increase the scattering cross section, resonance amplification is used, in which the frequency of the exciting light is near the width of the forbidden band. Let us consider as an example the resonance with the spin-orbit decoupled band $\omega_1 \approx E_g = E_0 + \Delta_0$. The dispersion law of the electrons now differs from the standard $p^2/2m_e$ since it is necessary to take into account the two energy bands. It is well known (see, e.g., Ref. 7) that in this case the scattering is determined not by the density fluctuations, but by fluctuations of the quantities which are the coefficients in the expansion of the light-matter interaction Hamiltonian in powers of the vector potential of the light wave. In the resonance case under consideration the question is one of fluctuations of the quantity[8]

$$N = \int \Psi^+ e^{i\mathbf{kr}} \gamma(\mathbf{p}) \Psi \, d^3\mathbf{r},$$

where

$$\gamma(\mathbf{p}) = \frac{|P_{CV}{}^2|}{m_0{}^2}\frac{m_e}{3}\frac{\mathbf{e}_1\mathbf{e}_2}{E_C(\mathbf{p}+\mathbf{k}_\parallel) - E_V(\mathbf{p}+\mathbf{k}_\parallel-\mathbf{k}_1) - \omega_1}. \tag{16}$$

The Kane model was used to obtain this formula: P_{CV} is the interband matrix element of the momentum; the indices C and V label the conduction band and the valence band $E_0 + \Delta_0$, respectively; and m_0 is the mass of the free electron.

In order to avoid cumbersome calculations, let us restrict the discussion to the simplest case: $k_1 v_F \ll \Delta(p_F)$, where the resonance detuning $\Delta(p) = E_g + p^2/2\mu - \omega_1$, and μ is the reduced mass of the electron and the hole. This condition corresponds to the region outside the so-called "strong" resonance (see Ref. 8). In addition, we assume that $\omega_p \ll ck_\parallel$, which is the case in the usual Raman-scattering experiments (the momentum transfer is not too small). Then it is possible to neglect self-consistent effects in the calculation of the intensity of the high-frequency wing $\omega \gg ck_\parallel$. Using the usual approach to find the correction to the density matrix of the electrons interacting with the free electromagnetic field, it is possible to express the quantity $\langle N(t)N^+(0)\rangle_\omega$ in terms of the field correlator (9). As a result we obtain the scattering cross section in the collisionless approximation:

$$\frac{d\sigma}{d\Omega\, d\omega} = R^2(\omega_1)\frac{(\mathbf{e}_1\mathbf{e}_2)^2}{\pi}\frac{\omega_1}{\omega_2}\left(\frac{e^2}{m_0 c_0{}^2}\right)^2 [n(\omega)+1] \times \frac{N_s k_\parallel{}^2}{\varepsilon m_e}\frac{\omega_p{}^2}{\omega^4}\left[\frac{\omega^2}{(ck_\parallel)^2} - 1\right]^{1/2} \theta(\omega^2 - (ck_\parallel)^2), \tag{17}$$

where the amplification factor is $R(\omega_1) = |P_{CV}{}^2|/3m_0\Delta(p_F)$.

To summarize, in the present article we have predicted a new (to our knowledge) mechanism of inelastic light scattering by free electrons, consisting of the decay of the initial photon into two. The effect is specific for spatially inhomogeneous systems (one of the limiting cases is a two-dimensional electron gas), in which the electromagnetic field in the plasma cannot be separated into longitudinal and transverse modes. Under these conditions retardation effects become important, and they lead in particular to the appearance of an additional contribution to the Raman scattering in the region of relatively large energy deficits $\omega \gg ck_\parallel$. In this case, the above-implemented "cold" plasma condition $\omega \gg k_\parallel v_F$ is satisfied, but in the situation with resonant amplification the formulas which we have obtained correspond to the quite typical experimental situation of not too small detunings $\Delta \gg k_1 v_F$. Of course, it would be best to try to detect the effect found here in samples with high conductivity ($\sigma_0 \gg c$). Then the electron plasma can be assumed to be collisionless. Note, however, that an account of collisions, as can be seen from the general formula (8), does not qualitatively change the results.

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1) In this paper we will ignore spin effects.

2) The authors are grateful to V. I. Fal'ko for having pointed out in the review that the quantity ω' also gives the relativistic limit for the line width of the cyclotron resonance in a two-dimensional gas.

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