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INELASTIC LIGHT SCATTERING BY ELECTRONS AND PLASMONS IN METALS

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The cross section of the inelastic light scattering by electron–hole plasma in metals is studied. The Coulomb interaction of electron excitations is taken into account selfconsistently. The system of Boltzmann's equation for electronic fluctuations and Maxwell's equations for the interaction field is solved. Raman spectra consist of the electron–hole background, diffuson and plasmon resonances. The widths of this background and resonance are determined by the electron collision rate as well as by the decay of the incident and scattered radiation in metal. The line shape depends on the screening of the electron–light interaction, i.e., on the incident radiation frequency.

1. INTRODUCTION

In recent times the inelastic light scattering has attracted considerable attention because of a puzzle of the high temperature superconductivity [1–5]. Also synchrotron sources of radiation [6, 7] with high resolution have led to an advance in experimental investigations of electronic excitations in solids and liquids. Light scattering experiments permit to obtain the detailed information on various elementary excitations: phonons [8], plasmons [9, 10] and magnons [11]. The influence of transition into superconducting state on the Raman light scattering was firstly studied theoretically in Ref. [12] and recently for the synchrotron radiation frequency [13].

The Raman spectra present a very complex picture and attention to their subtle aspects was paid only recently. In particular, lately it has been discovered that the interaction of the phonon resonances with the electron–hole continuum leads to characteristic changes in the shape of the resonance line [14–16]. The resonance line acquires the Fano resonance line shape also known as the Breit–Wigner resonance in nuclear physics. The second example, where the resonance peak has a specific shape, is the two-magnon resonance in which two magnons are involved in the electron transition through a gap [17].

The inelastic light scattering by normal metals has not yet been studied experimentally in such a detailed manner as that by superconductors. We would like to concentrate on an effect which has already been noticed [18], but not investigated in details: the influence of the spatial distribution of the incident and scattered light in a metal on the resonance line shape. Up to now in investigations of Raman scattering, the solid state is treated as a nonabsorbent substance,

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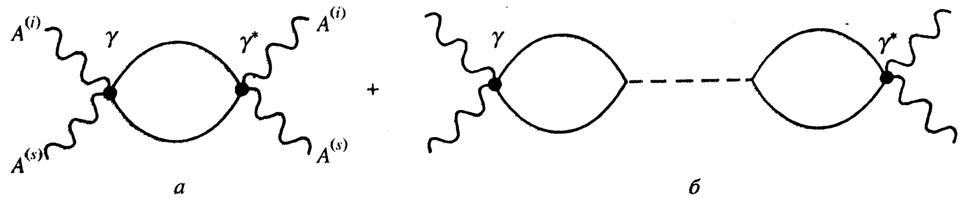

Fig. 1. Feynman diagrams depicting the two contributions to Raman scattering in metals. (*a*) The electron–hole contribution; (*b*) the contribution of electromagnetic excitation (plasmon, the dotted line) in electron–hole plasma. Here the solid line is the electron Green's function, the wavy line the incident and scattered light. The black dots are vertices describing the electron interaction with the incident and scattered light. The Coulomb interaction is represented by an empty vertex

where the incident and scattered light possess well defined wave vectors. The existence of imaginary parts of the wave vector leads to changing of the line shape. This effect should be considered as competitive to the Fano resonance.

In this paper we study the influence of the above mentioned field decay in a metal on resonance with the plasmon excitation. The interaction between plasmon resonance and the electron–hole excitations is taken into account. In Fig. 1 the effect of the electron–hole excitations is represented by a loop. The electron–hole excitations are shown in Fig. 1*a*. This diagram describes a continuum with a width depending on the collision rate. In the «dirty» limit the diagram involves a diffuson pole. The diagram 1*b*, where the electromagnetic interaction is shown by the dotted line, has a narrow plasmon pole at the electron plasma frequency ω_0. It will be shown, that the influence of the electron loop leads to asymmetry of the plasmon resonance (Fano effect). Two contributions shown in Fig. 1 should be integrated with the factor $|U(\mathbf{r},t)|^2 = |A^{(i)}A^{(s)}|^2$ having the width determined by the spatial damping of incident (i) and scattered (s) light.

The Raman cross section is expressed in terms of a linear response of the electron system with the Coulomb interaction to the external field $U(\mathbf{r},t)$. We solve selfconsistently Boltzmann's equation for electronic fluctuations and Maxwell's equations for the interaction field.

For applicability of Boltzmann's equation the following conditions should be fulfilled:

$$|k| = |k^{(i)} - k^{(s)}| \ll k_F, \quad |\omega| = |\omega^{(i)} - \omega^{(s)}| \ll \varepsilon_F,$$

where k_F and ε_F are the Fermi momentum and energy[1)]. The first condition allows an analytical expression to be found for the distribution function of charge carriers. We are interested in small values of $|\omega|$ and $|\omega| \simeq \omega_0$. In the latter case the condition $e^2/\hbar v \ll 1$ should be fulfilled, where v is the Fermi velocity. A justified method is not known for an evaluation of the response function for an arbitrary $e^2/\hbar v$, but permittivity calculated by Boltzmann's equation coincides with Lindhard's expression obtained in the limit $|k| \ll k_F$ and for arbitrary values of $|\omega|$. Therefore we use the kinetic equation and write it in the τ-approximation. The τ-approximation can be well founded for elastic scattering processes [19, 20]. An attempt to include the inelastic scattering by phonons has been made in [21], where the response function was obtained by Green functions method. Strictly speaking, the collision rate τ for large frequency transfer ($|\omega| \simeq \omega_0$) depends on ω and is determined by the electron–electron collisions. The long wave part of the Coulomb interaction is taken into account in a

[1)] We put h and Boltzman's constant equal to 1 except the final expressions.

self-consistent way (Vlasov–Landau approximation; for the scattering problem, see Ref. [18]). Contributions of the local field will be disregarded.

2. INELASTIC LIGHT SCATTERING AND SUSCEPTIBILITY

We consider light scattering by a metal, which lies in the half-space $z > 0$. The Raman cross section has the form

$$d\sigma(k_x, \omega) = \left(\frac{8\pi e^2}{mc\hbar\omega^{(i)}}\right)^2 \frac{\Sigma(k_x, \omega)}{1 - \exp(-\hbar\omega/k_B T)} \frac{k_z^{(s)}\omega^{(s)}d\omega^{(s)}d\Omega^{(s)}}{c(2\pi)^3}, \tag{1}$$

where the density–density correlation function $\Sigma(k_x, \omega)$ contains the bulk and surface contributions [18]. Here we are interested in the bulk part only. Then we can use the even continuation of all the fields in the semi-space $z < 0$ and take the Fourier transform with respect to coordinates. The correlator $\Sigma(k_x, \omega)$ is expressed in terms of the generalized susceptibility $\chi(\mathbf{k}, \omega)$:

$$\Sigma(k_x, \omega) = -\int \frac{dk_z}{2\pi} |U(\mathbf{k}, \omega)|^2 \operatorname{Im} \chi(\mathbf{k}, \omega). \tag{2}$$

The generalized susceptibility $\chi(\mathbf{k}, \omega)$ in the field $U(\mathbf{k}, \omega)$ is defined as follows

$$\int \frac{2d^3p}{(2\pi)^3} \gamma^*(\mathbf{p})\ f_p(\mathbf{k}, \omega) \equiv -\chi(\mathbf{k}, \omega) U(\mathbf{k}, \omega), \tag{3}$$

where $f_p(\mathbf{r}, t)$ is the electron distribution function with a specular boundary condition at $z = 0$.

The field $U(\mathbf{r}, t)$ considered below as the external force is the product of the vector potentials of the incident $\mathbf{A}^{(i)}(\mathbf{r}, t)$ and scattered $\mathbf{A}^{(s)}(\mathbf{r}, t)$ light

$$\mathbf{A}^{(i)}(\mathbf{r}, t)\mathbf{A}^{(s)}(\mathbf{r}, t) \simeq U(\mathbf{r}, t) = U(\mathbf{k}, \omega) \exp\left[i(\mathbf{k}_s \mathbf{s} - \omega t)\right],$$

where $\mathbf{k}_s = \mathbf{k}_s^{(i)} - \mathbf{k}_s^{(s)}$. Here the subscript s denotes the vector components parallel to the surface. For the unbounded space $\mathbf{e}^{(i)}$, $\mathbf{e}^{(s)}$ are the polarization vectors of the incident and scattered fields, respectively, and they are included in the vertex function $\gamma(\mathbf{p})$. For a half space the polarization vectors $\mathbf{e}^{(i)}$, $\mathbf{e}^{(s)}$ are determined by the solution of the electrodynamic problem (for details see, Refs. [18, 20]). The vertex function $\gamma(\mathbf{p})$ contains resonance denominators appearing in the second order of the perturbation theory with respect to $\mathbf{A}^{(i)}$, $\mathbf{A}^{(s)}$. The Fourier transform of the field $U(\mathbf{r}, t)$ is given by

$$U(\mathbf{k}, \omega) = \frac{2i\zeta}{\zeta^2 - k_z^2}, \tag{4}$$

where complex $\zeta = \zeta_1 + i\zeta_2$ is the sum of the normal components of the wave vector of the incident and scattered light in the metal.

The electron distribution function $f_p(\mathbf{r}, t)$ is searched in the form

$$f_p(\mathbf{r}, t) = f_0\left(\varepsilon(\mathbf{p}, \mathbf{r}, t) - \mu\right) + \frac{df_0}{d\varepsilon_0}\, \delta f_p(\mathbf{r}, t),$$

where $f_0(\varepsilon(\mathbf{p}, \mathbf{r}, t) - \mu)$ is the Fermi–Dirac distribution function depending on the local electron spectrum

$$\varepsilon(\mathbf{p}, \mathbf{r}, t) = \varepsilon_0(\mathbf{p}) + \gamma(\mathbf{p})U(\mathbf{r}, t).$$

The nonequilibrium part of the electron distribution function is ruled by Boltzmann's equation:

$$-i(\omega - \mathbf{kv})\, \delta f_p(\mathbf{k}, \omega) = [i\omega\gamma(\mathbf{p})U(\mathbf{k}, \omega) - e\mathbf{v}\mathbf{E}(\mathbf{k}, \omega)] - \nu(\delta f_p(\mathbf{k}, \omega) - \langle\delta f_p(\mathbf{k}, \omega)\rangle), \quad (5)$$

where ν is the collision rate and angle the brackets denote averaging over the Fermi surface

$$\langle(\cdots)\rangle = \frac{1}{n_0}\int \frac{2dS}{(2\pi)^3 v}(\ldots), \quad n_0 = \int \frac{2dS}{(2\pi)^3 v},$$

n_0 is the density of electron states; we assume $T \ll \epsilon_F$.

The electric field $\mathbf{E}(\mathbf{k}, \omega)$ describes the electron–electron Coulomb interaction and is determined by the Maxwell equation

$$\mathrm{rot}\,\mathrm{rot}\,\mathbf{E}(\mathbf{r}, \omega) - \frac{\omega^2}{c^2}\mathbf{D}(\mathbf{r}, \omega) = \frac{4\pi i\omega}{c^2}\mathbf{j}(\mathbf{r}, \omega) \quad (6)$$

with

$$D_\alpha(\mathbf{k}, \omega) = \epsilon^o_{\alpha\beta}E_\beta(\mathbf{k}, \omega),$$

$$\mathbf{j}(\mathbf{k}, \omega) = e\int \frac{2d^3p}{(2\pi)^3}\mathbf{v}\,\delta f_p(\mathbf{k}, \omega), \quad (7)$$

where $\epsilon^o_{\alpha\beta}$ is the dielectric constant of the filled bands.

Substituting the solution of Boltzman's equation (5) into Eq. (7) we obtain:

$$j_\alpha(\mathbf{k}, \omega) = \sigma_{\alpha\beta}(\mathbf{k}, \omega)E_\beta(\mathbf{k}, \omega) + \Gamma^{(\gamma)}_\alpha(\mathbf{k}, \omega)U(\mathbf{k}, \omega), \quad (8)$$

where

$$\sigma_{\alpha\beta}(\mathbf{k}, \omega) = ie^2 n_0\left\langle\frac{v_\alpha\tilde{v}_\beta}{\tilde{\omega} - \mathbf{kv}}\right\rangle, \quad \Gamma^{(\gamma)}_\alpha(\mathbf{k}, \omega) = e\omega n_0\left\langle\frac{v_\alpha\tilde{\gamma}(\mathbf{p})}{\tilde{\omega} - \mathbf{kv}}\right\rangle,$$

$$\tilde{\mathbf{v}} = \mathbf{v} + \frac{i\nu\langle\mathbf{v}/(\tilde{\omega} - \mathbf{kv})\rangle}{1 - \langle i\nu/(\tilde{\omega} - \mathbf{kv})\rangle}, \qquad \tilde{\gamma}(\mathbf{p}) = \gamma(\mathbf{p}) + \frac{i\nu\langle\gamma(\mathbf{p})/(\tilde{\omega} - \mathbf{kv})\rangle}{1 - \langle i\nu/(\tilde{\omega} - \mathbf{kv})\rangle},$$

$\tilde{\omega} = \omega + i\nu$.

The solutions of Maxwell's equation (6), (7) has the form

$$E_\alpha(\mathbf{k}, \omega) = \frac{4\pi i\omega}{c^2}U(\mathbf{k}, \omega)\mathscr{D}_{\alpha\beta}(\mathbf{k}, \omega)\Gamma^{(\gamma)}_\beta(\mathbf{k}, \omega) \quad (9)$$

with the matrix

$$\mathscr{D}_{\alpha\beta}(\mathbf{k}, \omega) = \left(k^2\delta_{\alpha\beta} - k_\alpha k_\beta - \frac{\omega^2}{c^2}\epsilon_{\alpha\beta}(\mathbf{k}, \omega)\right)^{-1},$$

where $\epsilon_{\alpha\beta}(\mathbf{k},\omega)$ is related to the conductivity tensor

$$\epsilon_{\alpha\beta}(\mathbf{k},\omega) = \epsilon^{o}_{\alpha\beta} + \frac{4\pi i}{\omega}\sigma_{\alpha\beta}(\mathbf{k},\omega). \tag{10}$$

Using the solution of Boltzmann's equation (5) and Maxwell's equation (9) we obtain the generalized susceptibility (3)

$$\chi(\mathbf{k},\omega) = \omega n_0 \left\langle \frac{\gamma^*(\mathbf{p})\tilde{\gamma}(\mathbf{p})}{\tilde{\omega} - \mathbf{kv}} \right\rangle - \frac{4\pi}{c^2}\tilde{\Gamma}^{(\gamma^*)}_{\alpha}(\mathbf{k},\omega)\mathscr{D}_{\alpha\beta}(\mathbf{k},\omega)\Gamma^{(\gamma)}_{\beta}(\mathbf{k}\omega), \tag{11}$$

where

$$\tilde{\Gamma}^{(\gamma^*)}_{\alpha}(\mathbf{k},\omega) = e\omega n_0 \left\langle \frac{\tilde{v}_\alpha \gamma^*(\mathbf{p})}{\tilde{\omega} - \mathbf{kv}} \right\rangle.$$

For normal propagation of the incident and scattered waves ($\mathbf{k} = k_z = k$) the expression (11) reads

$$\chi(k,\omega) = \omega n_0 \left\langle \frac{\gamma^*(\mathbf{p})\tilde{\gamma}(\mathbf{p})}{\tilde{\omega} - kv_z} \right\rangle + \frac{4\pi}{\omega^2 \epsilon_{zz}(k,\omega)}\tilde{\Gamma}^{(\gamma^*)}_{z}(k,\omega)\Gamma^{(\gamma)}_{z}(k,\omega). \tag{12}$$

Substituting (12) into (2) we obtain

$$\Sigma(k_x = 0,\omega) = \Sigma_1(0,\omega) + \Sigma_2(0,\omega),$$

where $\Sigma_1(0,\omega)$ is the contribution of the first term in (12) related to the excitation of electron–hole pairs and $\Sigma_2(0,\omega)$ exhibits the plasmon resonance connected to $\epsilon_{zz}(k,\omega)$. These two contributions are shown in Fig. 1; the loops correspond to $\Gamma_z(k,\omega)$ and the dotted line to $\epsilon_{zz}(k,\omega)$.

3. LARGE AND LOW SPATIAL DISPERSION

Let us consider two important limiting cases.

(*i*) Large k-limit: $kv \gg |\tilde{\omega}|$.

Since $\left\langle \dfrac{v_z}{\tilde{\omega} - kv_z} \right\rangle = -\dfrac{1}{k} + i\pi\dfrac{\tilde{\omega}}{k^2}\langle \delta(\mu)/v \rangle$, with $\mu = \cos(\mathbf{k},\mathbf{v})$, we obtain the susceptibility (12) in the form

$$\chi(k,\omega) = \frac{\pi n_0 \omega}{k}\left\langle |\gamma(\mathbf{p}) - \langle \gamma(\mathbf{p})\rangle|^2 \delta(\mu)/v \right\rangle, \tag{13}$$

Eq. (13) describes the screening of light scattering by the Coulomb electron–electron interaction [22]. This means that in the frequency range $\omega \simeq v|\zeta| < \omega_0$ up to the plasma frequency ω_0 the density fluctuations are screened due to the Coulomb electron–electron interaction.

(*ii*) Low k-limit: $kv \ll |\tilde{\omega}|$.

All integrands in χ (12) we expand in power series with respect to $kv/|\tilde{\omega}|$. The second term in (12) is proportional to k^2. In the leading approximation the first term gives

$$\chi(k \to 0, \omega) = \frac{n_0\omega}{\tilde{\omega}} \left(\langle |\gamma(\mathbf{p})|^2 \rangle + \frac{i\nu}{\omega - i\nu \langle v_z^2 \rangle k^2 \tilde{\omega}^{-2}} |\langle \gamma(\mathbf{p}) \rangle|^2 \right). \quad (14)$$

We retain in the denominator (the diffuson pole) the term proportional to k^2, since it is essential at low $\omega \sim k^2 \langle v_z^2 \rangle / \nu \ll \nu$.

If we left out of considerations the frequency range near the diffuson pole, we obtain

$$\chi(k, \omega) = \chi(k = 0, \omega) + \delta\chi(k, \omega),$$

where the first term results only from the electron–hole excitations

$$-\operatorname{Im} \chi(k = 0, \omega) = n_0 \langle |\gamma(\mathbf{p}) - \langle \gamma(\mathbf{p}) \rangle|^2 \rangle \frac{\nu\omega}{\omega^2 + \nu^2}. \quad (15)$$

This frequency dependence of the cross section has been obtained in [19, 23]. The first term in Eq. (12) gives also a contribution proportional to k^2:

$$\chi_1(k, \omega) = \chi(0, \omega) + \delta\chi_1(k, \omega),$$

$$\delta\chi_1(k, \omega) = \frac{n_0\omega k^2}{\tilde{\omega}^3} \left[\langle v_z^2 |\gamma(\mathbf{p})|^2 \rangle - \frac{\nu^2}{\omega^2} \langle v_z^2 \rangle \, | \langle \gamma(\mathbf{p}) \rangle |^2 + \frac{i\nu}{\omega} (\langle \gamma^*(\mathbf{p}) \rangle \langle \gamma(\mathbf{p}) v_z^2 \rangle + \text{c.c.}) \right]. \quad (16)$$

The second term in Eq. (12) has the resonance form:

$$\chi_2(k, \omega) = \frac{n_0 \omega_0^2 \varepsilon_{zz}^0 k^2}{\langle v_z^2 \rangle \tilde{\omega}^4 \varepsilon_{zz}(k, \omega)} \left\langle v_z^2 \left(\gamma(\mathbf{p}) + \frac{i\nu}{\omega} \langle \gamma(\mathbf{p}) \rangle \right) \right\rangle \left\langle v_z^2 \left(\gamma^*(\mathbf{p}) + \frac{i\nu}{\omega} \langle \gamma^*(\mathbf{p}) \rangle \right) \right\rangle, \quad (17)$$

where

$$\tilde{\omega}^3 \omega \varepsilon_{zz}(k, \omega) = \varepsilon_{zz}^{(0)} \left(\tilde{\omega}^3 \omega - \omega_0^2 \tilde{\omega}^2 - \omega_0^2 u^2 k^2 - i\nu\omega_0^2 \langle v_z^2 \rangle k^2 / \omega \right),$$

the electron plasma frequency $\omega_0^2 = 4\pi e^2 n_0 \langle v_z^2 \rangle / \varepsilon_{zz}^0$, and the dispersion parameter $u^2 = \langle v_z^4 \rangle / \langle v_z^2 \rangle$.

The equation $\varepsilon_{zz}(k, \omega) = 0$ gives frequency and damping (including its dispersion) of plasmon $\omega = \omega_{pl}(k) - i\Gamma(k)$, where

$$\omega_{pl}^2(k) = \omega_0^2 + u^2 k^2, \quad \Gamma(k) = \nu \left[1 + \left(u^2 - \langle v_z^2 \rangle \right) k^2 / \omega_0^2 \right] / 2.$$

Below we will omit the small dispersion of damping.

The terms proportional to k^2 (16) and (17) can be written as follows

$$\delta\chi(k, \omega) = \frac{n_0 | \langle v_z^2 \gamma(\mathbf{p}) \rangle |^2 k^2}{\langle v_z^2 \rangle \tilde{\omega}} \left[\frac{C_1 \omega}{\tilde{\omega}^2} + \frac{\omega\omega_0^2 - \nu^2 \tilde{\omega} C_3 + 2i\nu\omega\tilde{\omega} C_2}{\Delta} \right], \quad (18)$$

where $\Delta = \tilde{\omega}^3 \omega - \tilde{\omega}^2 \omega_0^2 - \omega_0^2 u^2 k^2$,

$$C_1 = \frac{\langle v_z^2 \rangle \langle v_z^2 |\gamma(\mathbf{p})|^2 \rangle}{|\langle v_z^2 \gamma(\mathbf{p}) \rangle|^2}, \quad C_2 = \frac{\langle v_z^2 \rangle (\langle \gamma^*(\mathbf{p}) \rangle \langle v_z^2 \gamma(\mathbf{p}) \rangle + c.c.)}{2 |\langle v_z^2 \gamma(\mathbf{p}) \rangle|^2}, \quad C_3 = \frac{\langle v_z^2 \rangle^2 |\langle \gamma(\mathbf{p}) \rangle|^2}{|\langle v_z^2 \gamma(\mathbf{p}) \rangle|^2}.$$

Now we are in a position to calculate the cross section.

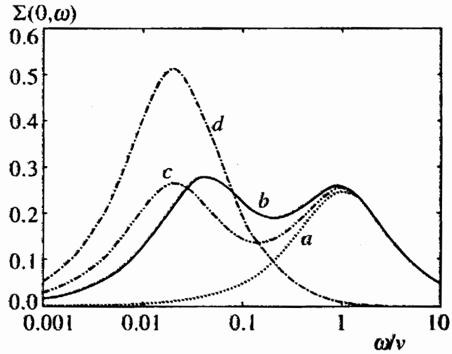

Fig. 2. Diffuson resonance and relaxation maximum at different values of parameters given in the text. In the curve (*d*) the relaxation maximum is absent for the complete screening. (*b*), (*c*) The screening is partial, the mean free length for (*b*) is larger than for (*c*). The curve (*a*) represents the Raman spectrum without the diffuson resonance (see, Eq. (23))

4. LIGHT SCATTERING CROSS SECTION

In the «clean» limit $|\omega + i\nu| \ll v|\zeta|$ one obtains the cross section by substituting Eq. (13) into (2). For a more interesting case, $\zeta_1 \gg \zeta_2$, we get

$$\Sigma(k_x = 0, \omega) = \pi n_0 \omega \left\langle |\gamma(\mathbf{p}) - \langle\gamma(\mathbf{p})\rangle|^2 \delta(\mu)/v \right\rangle \left(\frac{4}{\pi\zeta_1^2} \ln \frac{v\zeta_1}{\omega} + \frac{1}{\zeta_1\zeta_2} \right). \tag{19}$$

In this case $\Sigma(0,\omega)$ has a maximum at $\omega \simeq v\zeta_1$.

In the «dirty» limit $\nu \gg v|\zeta|$, there are two regions: the low and large frequency transfers. For the case of low frequency transfer $|\omega| \le \nu$, we obtain the cross section by substituting (14) into (2). If $\zeta_1 \gg \zeta_2$, we can integrate only $|U|^2$, taking the smooth function $\operatorname{Im}\chi(k,\omega)$ at $k = \zeta_1$:

$$\Sigma(k \to 0, |\omega| \le \nu) = \frac{n_0 \nu \omega}{\zeta_2(\omega^2 + \nu^2)} \left(\langle|\gamma(\mathbf{p}|^2\rangle - \frac{\omega^2 - \zeta_1^2\langle v_z^2\rangle}{\omega^2 + (\zeta_1^2\langle v_z^2\rangle/\nu)^2} |\langle\gamma(\mathbf{p})\rangle|^2 \right). \tag{20}$$

This expression has two maxima (see Fig. 2). One at $|\omega| \simeq \zeta_1^2\langle v_z^2\rangle/\nu \ll \nu$ describes the diffuson excitations. Another at $|\omega| = \nu$ is resulted from relaxation processes in the electron system. The relaxation maximum is absent (curve (*d*)), if $\langle|\gamma(\mathbf{p})|^2\rangle = |\langle\gamma(\mathbf{p})\rangle|^2$. We define this case as the complete screening limit. For $|\omega| \gg \zeta_1\langle v_z^2\rangle^{1/2}$ Eq. (20) transforms (see, curve (*a*)) to Eq. (23) obtained first by Zawadowski and Cardona [23]. The following sets of parameters ($w = |\langle\gamma(\mathbf{p})\rangle|^2/\langle|\gamma(\mathbf{p})|^2\rangle, v_0 = \langle v_z^2\rangle\zeta_1^2/\nu^2$) is used in Fig. 2: (*a*) $w = 0.5$, $v_0 = 0$; (*b*) $w = 0.5$, $v_0 = 0.04$; (*c*) $w = 0.5$, $v_0 = 0.02$; (*d*) $w = 1.0$, $v_0 = 0.02$. The units of $\Sigma(0,\omega)$ are $n_0\langle|\gamma(\mathbf{p})|^2\rangle/\zeta_2$.

For the large frequency transfer $|\omega| > \nu$, we find the cross section using (15), (18). One can obtain a simple form in the limit $u^2\zeta_1^2 \ll \nu\omega_0$, where the spatial dispersion of plasmon can be disregarded. Making use of the integrals

$$\int \frac{dk_z}{2\pi} |U(\mathbf{k},\omega)|^2 = \frac{1}{\zeta_2}, \quad \int \frac{dk_z}{2\pi} |U(\mathbf{k},\omega)|^2 k_z^2 = \frac{\zeta_1^2}{\zeta_2}, \tag{21}$$

we obtain

$$\Sigma(0,\omega) = \Sigma_1(0,\omega) + \delta\Sigma(0,\omega), \tag{22}$$

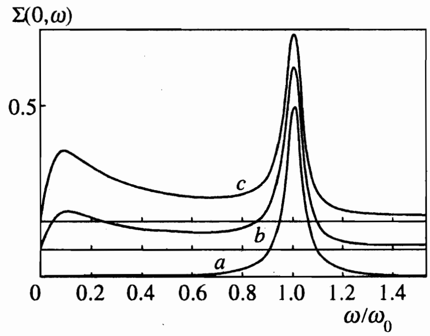

Fig. 3. Theoretical prediction of Raman spectra for normally incident and scattered light. The background is the electron–hole continuum with large collision rate. The resonance corresponds to the excitation of plasmon with low spatial dispersion. The Coulomb screening of electron density fluctuations is complete for the curve (*a*), and partial for (*c*). The curves (*b*) and (*c*) are shifted along the abscissa. The parameters are defined in the text

$$\Sigma_1(0,\omega) = n_0\langle|\gamma(\mathbf{p}) - \langle\gamma(\mathbf{p})\rangle|^2\rangle \frac{\nu\omega}{\zeta_2(\omega^2+\nu^2)}, \tag{23}$$

$$\delta\Sigma(0,\omega) = \frac{n_0|\langle\gamma(\mathbf{p})v_z^2\rangle|^2\zeta_1^2\nu\omega}{\langle v_z^2\rangle\zeta_2(\omega^2+\nu^2)^3}\left((3\omega^2-\nu^2)C_1 + \frac{\omega_0^2\left[4\omega^2(\omega^2-\nu^2) - \omega_0^2(3\omega^2-\nu^2)\right]}{(\omega^2-\omega_0^2)^2+\omega^2\nu^2} - \right.$$
$$\left. - \frac{(\omega^2+\nu^2)\left[\nu^2(3\omega^2-2\omega_0^2-\nu^2)C_3 + 2((\omega^2-\nu^2)(\omega^2-\omega_0^2) - 2\omega^2\nu^2)\right]C_2}{(\omega^2-\omega_0^2)^2+\omega^2\nu^2}\right). \tag{24}$$

The frequency dependence of the cross section (22) is shown in Fig. 3 for various values of parameters: $C_1 = C_3 = 1$, $u^2\zeta_1^2/\omega_0^2 = 0.04$, $\nu/\omega_0 = 0.08$, and (*a*) $C_2 = 1$, $x = 0$, (*b*) $C_2 = 2$, $x = 0.2$, (*c*) $C_2 = 2$, $x = 0.4$, where $x \equiv u^2\langle|\gamma(\mathbf{p}) - \langle\gamma(\mathbf{p})\rangle|^2\rangle\langle v_z^2\rangle/|\langle\gamma(\mathbf{p})v_z^2\rangle|^2$. The units of $\Sigma(0,\omega)$ are $n_0|\langle\gamma(\mathbf{p})v_z^2\rangle|^2/\zeta_2\langle v_z^2\rangle u^2$.

There is a wide background in the range $\nu < |\omega| < \omega_0$ even for a weak **p**-dependence $\gamma(\mathbf{p})$ when

$$\langle|\gamma(\mathbf{p}) - \langle\gamma(\mathbf{p})\rangle|^2\rangle \simeq 0.2|\langle\gamma(\mathbf{p})\rangle|^2.$$

The resonance line shape is asymmetric if the coefficient C_2 differs markedly from unity: the resonance curve drops more rapidly at the side $|\omega| > \omega_0$. Let us note that the parameters $\gamma(\mathbf{p})$, ζ_1 and ζ_2 depend on frequencies of the incident and scattered light, and ν is a function of ω. All these dependencies can be disregarded near the plasmon resonance, but they modify the form of the background.

To take into account the spatial dispersion of plasmons we calculate the integral

$$\int \frac{dk_z}{2\pi}|U(\mathbf{k},\omega)|^2\frac{k_z^2}{k_z^2-k_p^2} = \frac{\zeta_1}{2\zeta_2}\left(\frac{\zeta_1 - k_p\,\mathrm{sign}\omega}{\zeta_1^2 - k_p^2 + 2i\zeta_1\zeta_2} + (k_p,\zeta_2 \to -k_p,-\zeta_2)\right),$$

where the terms of the order ζ_2^2/ζ_1^2 are omitted and $k_p^2 = (a+ib)/u^2\omega_0^2$,

$$a = (\omega^2-\nu^2)(\omega^2-\omega_0^2) - 2\nu^2\omega^2, \quad b = \nu\omega\left[2(\omega^2-\omega_0^2) + \omega^2 - \nu^2\right].$$

Using (2) and (18) we obtain instead of (24) the contribution

$$\delta\Sigma(0,\omega) = \frac{n_0|\langle\gamma(\mathbf{p})v_z^2\rangle|^2\zeta_1}{2\langle v_z^2\rangle\zeta_2}\left[\frac{2\omega\nu(3\omega^2-\nu^2)\zeta_1C_1}{(\omega^2+\nu^2)^3} + \left(\left(\frac{\omega^2\omega_0^2}{\omega^2+\nu^2} - \nu^2C_3\right)\mathscr{L}_2(k_p,\zeta_2) + \right.\right.$$
$$\left.\left. + \nu\omega\left(\frac{\omega_0^2}{\omega^2+\nu^2} - 2C_2\right)\mathscr{L}_1(k_p,\zeta_2) + (k_p,\zeta_2 \to -k_p,-\zeta_2)\right)\right], \tag{25}$$

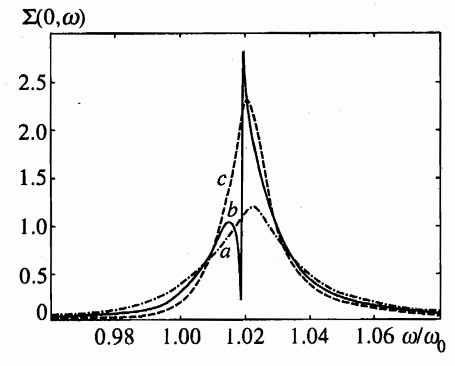

Fig. 4. Plasmon resonance in light scattering for crossover of damping (curve (*b*) $\nu/\omega_0 = 0.0115$); (*a*) plasmon damping is larger than the field damping ($\nu/\omega_0 = 0.02$); (*c*) $\nu/\omega_0 = 0.006$; ν is the electron collision rate, ω_0 — the plasma frequency

where

$$\mathscr{L}_1(k_p,\zeta_2) = \left[(\zeta_1 - k_1 \operatorname{sign}\omega)\left(a - u^2\omega_0^2\zeta_1^2\right) - \left(b - 2u^2\omega_0^2\zeta_1\zeta_2\right) k_2 \operatorname{sign}\omega\right]/\Delta_1,$$

$$\mathscr{L}_2(k_p,\zeta_2) = \left[(\zeta_1 - k_1 \operatorname{sign}\omega)\left(b - 2u^2\omega_0^2\zeta_1\zeta_2\right) + \left(a - u^2\omega_0^2\zeta_1^2\right) k_2 \operatorname{sign}\omega\right]/\Delta_1,$$

$$\Delta_1 = \left(a - u^2\omega_0^2\zeta_1^2\right)^2 + \left(b - 2u^2\omega_0^2\zeta_1\zeta_2\right)^2, \qquad (26)$$

$k_1 + ik_2 = k_p$ with

$$k_1 = \left(\sqrt{a^2+b^2} + a\right)^{1/2}/\sqrt{2}u\omega_0, \quad k_2 = \left(\sqrt{a^2+b^2} - a\right)^{1/2} \operatorname{sign}\omega/\sqrt{2}u\omega_0.$$

For $u^2\zeta_1^2 \ll \nu\omega$ Eq. (25) coincides with (24). Expression (25) is valid if $|\omega + i\nu| \gg v\zeta_1$.

The position of a resonance is given by the condition $a = u^2\omega_0^2\zeta_1^2$. It means that a plasmon is excited with the wave vector equal to the double wave vector of the incident light (we consider the back scattering geometry). The width of the resonance is determined by competition between intrinsic plasmon damping b and decay of light ζ_2. Usually the condition $2u^2\omega_0^2\zeta_1\zeta_2 \ll |b|$ holds, where the width of plasmon resonance is connected with the electron collision rate only (curve (*a*) in Fig. 4).

The resonance line has a critical behaviour ((*b*) in Fig. 4) when $b \simeq 2u^2\omega_0^2\zeta_1\zeta_2$ (see (26)). In the case (*c*) the resonance width is conditioned by decay of the light ζ_2. The parameter x and the units of $\Sigma(0,\omega)$ are defined as for Fig. 3 (see text after (24)), $C_1 = C_3 = 1$, $C_2 = 2$, $u^2\zeta_1^2/\omega_0^2 = 0.04$, $x = 0.2$, $2\zeta_2/\zeta_1 = 0.3$.

5. CONCLUSIONS

In this paper we considered the effect of the electron collision rate and incident field decay on inelastic light scattering in metals. Our method is based on the straightforward solution of Bolztmann's equation for electronic fluctuations and Maxwell's equation for the electron–electron interaction field. If the incident radiation frequency is not so large, the momentum dependence of the electron–light interaction $\gamma(\mathbf{p})$ has to be taken into consideration. For the scattering geometry A_{1g} (incident and scattered light are polarized along x-axis and propagate along z-axis)

$$\gamma(\mathbf{p}) = e_x^{(i)} e_x^{(s)} \left[1 + \frac{1}{m} \sum_n \left(\frac{|p_{fn}^x|^2}{\epsilon_f(\mathbf{p}) - \epsilon_n(\mathbf{p}) + \omega^{(i)}} + \frac{|p_{fn}^x|^2}{\epsilon_f(\mathbf{p}) - \epsilon_n(\mathbf{p}) - \omega^{(s)}} \right) \right], \qquad (27)$$

where the subscript f denotes the index of the band in which the carriers exist, the transitions take place into any band n, p_{fn}^x is the electron momentum matrix element, m is the electron mass, and for the semi-infinite metal [18] $e_x^{(i)} = \left(1 + \sqrt{\epsilon_{xx}(\omega^{(i)})}\right)^{-1}$.

The electron–light interaction is screened by the Coulomb interaction as well as by electron–electron collisions. The smooth part of the Raman spectra (background) is determined by the screened electron–light interaction. This part exists due to $\mathbf{p}$-dependent second term in bracket (27). The screening is not effective for the diffuson and plasmon resonances. For complete screening there are only the plasmon resonance with symmetric line shape and the asymmetric diffuson maximum. In an intermediate case, the wide electron–hole background appears and the plasmon resonance has the asymmetric line shape. For low collision rate, the width of the resonance depends on the decay of incident radiation. If the collision rate is comparable with the decay of incident radiation, the plasmon resonance curve has non trivial form. Let us emphasize, that the diffuson maxima and the plasmon resonance are located in very different parts of spectrum. The collision rate $\nu \approx 10^3$ cm^{-1}, according to the estimation given in Ref. [16] for YBaCuO in the normal state and for the optic range of the incident radiation. Then the diffusion maximum has to be observed at $\omega \approx 10$ cm^{-1}.

In a layered system having a cylindrical Fermi surface with its axis perpendicular to the surface of the sample, the plasmon peak resonance does not appear for incident light normal to the surface, since $v_z = 0$. However, if the incident light falls at the surface an angle different from zero, the plasmon resonance peak should be observed.

In order to observe the peculiarity of plasmon resonance one needs a source of radiation with frequency comparable to the interband electron energy and a good resolution.

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