Theory of electronic Brillouin scattering in metals

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The theory of Brillouin light scattering in metals by conduction electrons which interact with acoustic phonons and impurities is developed. Effects of the surface and the Coulomb interaction of carriers are taken into account. The self-consistent electron-phonon interaction changes the electron-hole contribution—the wide relaxation continuum appears with the temperature-dependent collision frequency. The sharp peaks in the cross section arise due to excitation of the bulk longitudinal phonons, the bulk transverse phonons, and the surface Rayleigh phonons. The contribution of the bulk phonons reflected by the surface has the form of a narrow continuum with a sharp maximum for the slipping phonons.

1. The role of phonons in the inelastic light process in insulators has been studied extensively both theoretically and experimentally[1–3] (see also Refs. 2 and 3). The scattering in this case is induced by the dielectric permittivity fluctuations associated with the lattice vibrations. As was found experimentally in HTSC,[4–8] the inelastic light scattering is virtually independent of the frequency transfer in the range $\omega \simeq 10^3 - 10^4$ cm^{-1}. This background was explained by the electron-impurity interaction[9] and by the electron–phonon interaction.[10] In Ref. 11, the phonon resonances were studied in the approximation in which one phonon group scatters the light and the other interacts with the electrons. In all those papers the distribution of the incident light and scattered light in a metal was disregarded. We shall show that the distribution is especially significant.

The Green's function method used previously for studying the inelastic light scattering in normal metals and superconductors[9–12] is very cumbersome for boundary-value problems. However, the problem under study for a normal metal is essentially semiclassical because the momentum transfer is smaller than the Fermi momentum, and because the energy transfer is smaller than the interband transition frequency. We develop a new method using the Boltzmann equation with appropriate boundary conditions. This method was used in Ref. 13 for studying the Raman light scattering with the exitation of plasmons. In this paper we focus on the effect of the surface in the electron–phonon interaction, taking into account the metal anisotropy.

2. The microscopic Hamiltonian describing the inelastic light scattering has the form

$$H = \frac{e^2}{mc^2} \int d^3r \int \frac{d^3p}{(2\pi)} \hat{f}_p(\mathbf{r},t) \gamma_{\alpha\beta}(\mathbf{p}) \mathbf{A}_\alpha^{(i)}(\mathbf{r},t) \mathbf{A}_\beta^{(s)}(\mathbf{r},t), \tag{1}$$

where $\hat{f}_p(\mathbf{r},t)$ is the operator of the electronic density fluctuations, $\mathbf{A}^{(i)}(\mathbf{r},t)$ and $\mathbf{A}^{(s)}(\mathbf{r},t)$ are the vector potentials of the incident and scattered waves, and $\gamma_{\alpha\beta}(\mathbf{p})$ is the vertex factor

$$\gamma_{\alpha\beta}(\mathbf{p})=\left(\delta_{\alpha\beta}+\frac{1}{m}\sum_{n}\frac{p_{fn}^{\beta}\,p_{nf}^{\alpha}}{\epsilon_f(\mathbf{p})-\epsilon_n(\mathbf{p})+\omega^{(i)}}+\frac{p_{fn}^{\beta}\,p_{nf}^{\alpha}}{\epsilon_f(\mathbf{p})-\epsilon_n(\mathbf{p})-\omega^{(s)}}\right). \tag{2}$$

Here $\omega^{(i)}$ and $\omega^{(s)}$ are the frequencies of the incident and scattered light, f is the index of a band in which carriers exist, the sum is over all zones n, and $\mathbf{p}_{fn}$ is the electron momentum matrix element. We introduce

$$\omega=\omega^{(i)}-\omega^{(s)},\quad \mathbf{k}_s=\mathbf{k}_s^{(i)}-\mathbf{k}_s^{(s)}, \tag{3}$$

where s denotes the vector components along the surface. The cross section calculated by using Eq. (1) can be expressed in terms of the correlator

$$K_{\gamma^*\gamma}(\mathbf{r},t;\ \mathbf{r}',t')=\langle\langle\delta n_{\gamma^*}(\mathbf{r},t)\delta n_{\gamma}(\mathbf{r}',t')\rangle\rangle. \tag{4}$$

Here $\langle\langle\ldots\rangle\rangle$ denotes the statistical average. The density fluctuation

$$\delta n_{\gamma}(\mathbf{r},t)=2\int\frac{d^3p}{(2\pi)^3}\,\gamma(\mathbf{p})f_p(\mathbf{r},t) \tag{5}$$

is modified by the factor $\gamma(\mathbf{p})=e_{\alpha}^{(i)}\,e_{\beta}^{(s)}\,\gamma_{\alpha\beta}(\mathbf{p})$, where the complex parameters $e_{\alpha}^{(i)}$ and $e_{\beta}^{(s)}$ are defined by matching the field in the metal to the incident field and the scattered field in the vacuum.[14]

We assume that the metal occupies the half-space $z>0$. In order to calculate the Fourier transform of the correlation function (4) with respect to the coordinates parallel to the surface $\mathbf{s}-\mathbf{s}'$ and with respect to the time $t-t'$, we use the general fluctuation-dissipation theorem:

$$K_{\gamma^*\gamma}(\mathbf{k}_s,z,z';\omega)=\frac{2}{1-\exp(-\omega/T)}\,\mathrm{Im}\ \alpha(\mathbf{k}_s,z,z';\omega), \tag{6}$$

where α is the generalized susceptibility

$$\delta n_{\gamma^*}(\mathbf{k}_s,z;\omega)=2\int\frac{d^3p}{(2\pi)^3}\,\gamma^*(\mathbf{p})f_p(\mathbf{k}_s,z;\omega)$$

$$=-\int_0^{\infty}dz'\ \alpha(\mathbf{k}_s,z,z';\omega)U(\mathbf{k}_s,z';\omega) \tag{7}$$

to the external field

$$A^{(i)}(r,t)A^{(s)}(r,t)\simeq U(\mathbf{r},t)=U(\mathbf{k}_s,z;\omega)\exp[i(\mathbf{k}_s\,\mathbf{s}-\omega t)]. \tag{8}$$

If the frequencies $\omega^{(i)}$ and $\omega^{(s)}$ are in the normal skin range, the external field will be

$$U(\mathbf{k}_s,z;\omega)=\exp(i\zeta z), \tag{9}$$

where $\zeta=\zeta_1+i\zeta_2$ is the sum of the normal components of the incident and scattered light wave vectors in the metal, which depends on their polarizations.

3. We derive the generalized susceptibility by means of the Boltzmann equation

$$\mathbf{v}\,\frac{\partial\delta f_p(\mathbf{r},\omega)}{\partial\mathbf{r}}+(-i\omega+\tau_p^{-1})\delta f_p(\mathbf{r},\omega)=i\omega\gamma(\mathbf{p})U(\mathbf{r},\omega)+i\omega\lambda_{ik}(\mathbf{p})u_{ik}(\mathbf{r},\omega)-e\mathbf{v}\mathbf{E}(\mathbf{r},\omega), \tag{10}$$

where we use the collision integral with the impurities and phonons in the τ approximation. The interaction of electrons with the external field and the accoustic phonons is taken in the form

$$\varepsilon(\mathbf{p},\mathbf{r},t)=\varepsilon_0(\mathbf{p})+\gamma(\mathbf{p})U(\mathbf{r},t)+\lambda_{ik}(\mathbf{p})u_{ik}(\mathbf{r},t), \tag{11}$$

where the last term is the deformation potential. In the typical case the phonons are in the equilibrium state. The collision integral vanishes due to the local equilibrium electronic distribution function $f_0(\varepsilon(\mathbf{p},\mathbf{r},t)-\mu)$. The Boltzmann equation (10) can be linearized by the substitution

$$f_p(\mathbf{r},t)=f_0(\varepsilon(\mathbf{p},\mathbf{r},t)-\mu)+\frac{df_0}{d\varepsilon}\,\delta f_p(\mathbf{r},t). \tag{12}$$

The chemical potential is determined by the conservation of electronic density. As a result, the vertex factors $\gamma(\mathbf{p})$ and $\lambda_{ik}(\mathbf{p})$ in (10) are renormalized:

$$\gamma(\mathbf{p})\rightarrow\gamma(\mathbf{p})-\langle\gamma(\mathbf{p})\rangle/\langle 1\rangle,\quad \lambda_{ik}(\mathbf{p})\rightarrow\lambda_{ik}(\mathbf{p})-\langle\lambda_{ik}(\mathbf{p})\rangle/\langle 1\rangle, \tag{13}$$

where the brackets denote the integration over the Fermi surface

$$\langle\ldots\rangle=2\int(\ldots)\,\frac{dS}{(2\pi)^3v}\,.$$

The electric field $\mathbf{E}(\mathbf{r},\omega)$ represents the electron–electron interaction. For the self-consistent determination of the field we use the Maxwell's equation.

The acoustic phonon field obeys the elastic equation

$$-\lambda_{iklm}\,\frac{\partial^2u_l(\mathbf{r},\omega)}{\partial x_k\,\partial x_m}-\rho\omega^2\,u_i(\mathbf{r},\omega)=2\,\frac{\partial}{\partial x_k}\int\frac{d^3p}{(2\pi)^3}\,\lambda_{ik}(\mathbf{p})f_p(\mathbf{r},\omega), \tag{14}$$

where ρ is the density of the metal. The last term describes the electron response to phonons.[15]

We apply the specular boundary condition for Boltzmann's equation (10). Conservation of the tangential components of the electric and magnetic fields implies the boundary conditions at the surface $z=0$ for Maxwell's equations. The boundary condition for the elastic equation (14) means that the normal stress tensor components vanish:

$$\lambda_{izlm}\,\frac{\partial u_l(\mathbf{r},\omega)}{\partial x_m}=0\quad\text{for}\ z=0^+. \tag{15}$$

To solve the above equations, we use the even continuation to the $z<0$ half-space for $U(\mathbf{k}_s,z,\omega)$ for the components of the electric field $E_s(\mathbf{k}_s,z,\omega)$ parallel to the surface and for the elastic displacement $u_s(\mathbf{k}_s,z,\omega)$. For the perpendicular components

$E_z(\mathbf{k}_s,z,\omega)$ and $u_z(\mathbf{k}_s,z,\omega)$ we use the odd continuation. Hence we can use the Fourier transform with respect to all coordinates, and we find the solution of Boltzmann's equation.

The singularities at $z=0$ arise in the equations describing the phonons, (14), and in Maxwell's equations after the continuation. Thus the additional terms appear in the Fourier transforms of the equations with respect to the z coordinate. This surface contribution must be determined from the boundary conditions. In the Maxwell's equation such terms give the surface plasmon contribution.[13] These terms are omitted here.

The Fourier transform of (14) gives

$$(\lambda_{\alpha klm}\, k_k\, k_m-\rho\omega^2\,\delta_{\alpha l})u_l(\mathbf{k},\omega)=f_\alpha(\mathbf{k},\omega)+k_\alpha\, C_\alpha(\mathbf{k}_s,\omega), \tag{16}$$

where

$$f_i(\mathbf{k},\omega)=-i\langle\gamma(\mathbf{p})\lambda_{ik}(\mathbf{p})\rangle k_k\, U(\mathbf{k},\omega)-\omega\left\langle\frac{\lambda_{ik}\,\lambda_{lm}}{-\mathbf{vk}+i\tau_p^{-1}}\right\rangle k_k\, k_m u_l(\mathbf{k},\omega). \tag{17}$$

For the sound frequency range $|\omega|\simeq\omega_D\ll vk$, where ω_D is the Debye frequency. Only the main contribution with respect to ω/vk was kept in the first term on the right side in (17). The main contribution is real in the second term and renormalizes the sound velocity. Therefore, the next-order term of the series expansion is retained in it. It is imaginary and gives the sound damping.[15]

The last term in (16), where there is no sum over the Greek symbol α, appears as a result of the singularities at $z=0$. The quantities $C_\alpha(\mathbf{k}_s,\omega)$, which do not depend on k_z, are determined by the condition (15):

$$C_\alpha(\mathbf{k}_s,\omega)=D^s_{\alpha i}(\mathbf{k}_s,\omega)\lambda_{izlm}\int\frac{dk_z}{2\pi}\,D^b_{lk}(\mathbf{k},\omega)f_k(\mathbf{k},\omega)k_m, \tag{18}$$

where $D^b_{ik}(\mathbf{k},\omega)$ is the bulk Green's matrix for Eq. (16). The surface Green's matrix $D^s_{ik}(\mathbf{k}_s,\omega)$ for the boundary condition (15) is determined by the equation

$$\sum_\alpha D^s_{\alpha k}(\mathbf{k}_s,\omega)\lambda_{izlm}\int\frac{dk_z}{2\pi}\,D^b_{l\alpha}(\mathbf{k},\omega)k_m\,k_\alpha\,e^{ik_z z}=-\delta_{ik}\quad\text{for } z\to 0^+. \tag{19}$$

Thus we obtain the solution of the elastic equations (14) and (16). Using the solution of the Boltzmann equation (10), we find the generalized susceptibility (7) and we calculate the cross section.

4. The scattering cross section has the form

$$d\sigma=\left(\frac{8\pi e^2}{mc\hbar\omega^{(i)}}\right)^2\frac{\Sigma(\mathbf{k}_s,\omega)}{1-\exp(-\omega/T)}\,\frac{k_z^{(s)}\,\omega^{(s)}d\omega^{(s)}d\Omega}{c(2\pi)^3}, \tag{20}$$

where $\Sigma(\mathbf{k}_s,\omega)$ contains the electron–hole (e–h), the phonon bulk (ph–b), and the phonon surface (ph–s) contributions.

The electron–hole contribution (the background in Fig. 1, where only the Stokes range is shown) has the form

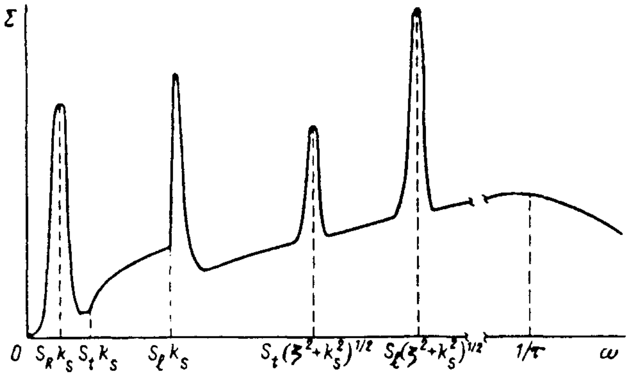

FIG. 1.

$$\Sigma_{\mathrm{c-h}}(\mathbf{k}_s,\omega)=-\mathrm{Im}\int\frac{dk_z}{2\pi}\,|U(\mathbf{k},\omega)|^2\left\langle\frac{\omega|\gamma(\mathbf{p})|^2}{\omega-\mathbf{vk}+i\tau_p^{-1}}\right\rangle. \tag{21}$$

For an isotropic τ and in the limiting case $l|\zeta|\ll 1$ $(l=v\tau)$, this expression takes the form

$$\Sigma_{\mathrm{c-h}}(\mathbf{k}_s,\omega)=\frac{\omega\tau}{(\omega\tau)^2+1}\langle|\gamma(\mathbf{p})|^2\rangle\zeta_2^{-1}. \tag{22}$$

Note that $\gamma(\mathbf{p})$ is renormalized, (13). The frequency dependence $\Sigma_{\mathrm{c-h}}(\mathbf{k}_s,\omega)$ was obtained in Ref. 9 for the electron-impurity interaction. For a higher temperature τ^{-1} is determined by the electron–phonon interaction. If the frequency transfer (or temperature) is larger than $\omega_D/3$, we have $\tau^{-1}=2\pi g\omega_D$ (or $2\pi gT$). Here g is a dimensionless constant of the electron–phonon interaction (concerning the coefficient 3 and the definition g see Ref. 16). For a low temperature and a low frequency $T,|\omega|\ll\omega_D$, the scattering rate $\tau^{-1}\simeq g\omega_D^{-2}\max(|\omega|^3,T^3)$. These results concerning the electron–phonon interaction differ slightly from those obtained in Ref. 10, where the factor violates the sum rule.

The bulk phonon contribution is

$$\Sigma_{\mathrm{ph-b}}(\mathbf{k}_s,\omega)=-\mathrm{Im}\sum_{\alpha\beta}\langle\gamma^*(\mathbf{p})\lambda_{\alpha\alpha}(\mathbf{p})\rangle$$
$$\times\langle\gamma(\mathbf{p})\lambda_{\beta\beta}(\mathbf{p})\rangle\int\frac{dk_z}{2\pi}\,|U(\mathbf{k},\omega)|^2\,D^b_{\alpha\beta}(\mathbf{k},\omega)k_\alpha k_\beta,$$

where it is assumed that the coordinate axes are the symmetry axes of a crystal in which the tensor $\langle\gamma(\mathbf{p})\lambda_{ik}(\mathbf{p})\rangle$ has a diagonal form. The Green's matrix $D^b_{\alpha\beta}(\mathbf{k},\omega)$ introduced above has poles which determine the bulk phonon dispersion.

Let us consider $\Sigma_{\mathrm{ph-b}}(\mathbf{k}_s,\omega)$ in the typical case

$$\frac{s}{v}\,\zeta_1\min(\zeta_1,l,1)\ll\zeta_2<\zeta_1$$

for normal incidence and normal scattering. The integral (23) gives

$$\Sigma_{\text{ph-b}}(\omega)=\frac{\pi\zeta_1|<\gamma(\mathbf{p})\lambda_{zz}(\mathbf{p})>|^2}{2\rho[(|\omega|-s_l\zeta_1)^2+s_l^2\ \zeta_2^2]}\ \text{sign}\ \omega, \tag{24}$$

where s_l is the longitudinal sound velocity. This expression has the form of a peak at $|\omega|=s_l\zeta_1$, whose width is $s_l\zeta_2$. In the opposite case, $\zeta_1<\zeta_2$, the peak broadening prevents its observation. A comparison of (22) and (24) shows that the ratio $\Sigma_{\text{ph-b}}^{\max}/\Sigma_{\text{e-h}}^{\max}\simeq\zeta_1/\zeta_2$. The nonperpendicular incidence and scattering also include the transverse phonon peaks whose height is proportional to k_s^2 and is on the order of the longitudinal phonon peak. The peaks are located at $|\omega|=\omega_{l,t}(\mathbf{k}_s,k_z=\zeta_1)$.

The surface contribution is

$$\Sigma_{\text{ph-s}}(\mathbf{k}_s,\omega)=-\text{Im}\sum_{\alpha\beta}D^s_{\alpha z}(\mathbf{k}_s,\omega)\lambda_{zz\beta\beta}\,I^*_\alpha(\mathbf{k}_s,\omega)I_\beta(\mathbf{k}_s,\omega) \tag{25}$$

with

$$I_\alpha(\mathbf{k}_s,\omega)=\sum_\gamma\ <\gamma(\mathbf{p})\lambda_{\gamma\gamma}(\mathbf{p})>\int\frac{dk_z}{2\pi}\,U(\mathbf{k},\omega)D^b_{\gamma\alpha}(\mathbf{k},\omega)k_\gamma\,k_\alpha. \tag{26}$$

The surface Green's matrix $D^s_{ik}(\mathbf{k}_s,\omega)$ has a pole which is defined by the Rayleigh dispersion of the phonons, $|\omega|=\omega_R(\mathbf{k}_s)$. There is a corresponding peak in the cross section whose shape is

$$\Sigma^{(1)}_{\text{ph-s}}(\mathbf{k}_s,\omega)\simeq\frac{\Gamma|<\gamma(\mathbf{p})\lambda_{zz}(\mathbf{p})>|^2}{\rho s([|\omega|-\omega_R(\mathbf{k}_s)]^2+\Gamma^2)}\ \text{sign}\ \omega. \tag{27}$$

Here and in (28) the coefficients are given for the isotropic case and for $|\zeta|\gg k_s$. The sound damping Γ obtained from the last term in (17) is

$$\Gamma\simeq\tau\omega^2\quad\text{for }kl\ll1\quad\text{and}\quad\Gamma\simeq\frac{s}{v}|\omega|\quad\text{for }kl\gg1.$$

In addition to the Rayleigh pole, there is the imaginary part in (25) in the range $|\omega|>s_tk_s$, where the bulk transverse phonons can exist. Here the imaginary part has the shape of a narrow continuum. In the range $|\omega|>s_l\,k_s$ the bulk longitudinal phonons also exist. The slipping longitudinal phonons produce a nonsymmetric (Fano-like) resonance, whose shape has the form

$$\Sigma^{(2)}_{\text{ph-s}}(\mathbf{k}_s,\omega)\simeq\frac{k_s^2|<\gamma(\mathbf{p})\lambda_{zz}(\mathbf{p})>|^2}{\rho s_l^2\ \zeta_1^2}\left(\frac{s_l\,\Gamma/k_s}{(|\omega|-s_l\,k_s)^2+\Gamma^2}\right)^{1/2}\text{sign}\ \omega,\ |\omega|>s_lk_s. \tag{28}$$

The resonant phenomena discussed here occur at low-frequency transfer. The relaxation continuum is in the wide frequency range.

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