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THEORY OF ELECTRONIC BRILLOUIN SCATTERING IN METALS

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The theory of the Brillouin light scattering in metals by conduction electrons interacting with acoustic phonons and impurities is developed. Effects of the surface and the Coulomb interaction of carriers are taken into consideration. The self-consistent electron-phonon interaction changes the electron-hole contribution - the wide relaxation continuum is appeared with the temperature dependent collision frequency.The sharp peaks in the cross section are arised due to exitation 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 the sharp maximum for the slipping phonons.

1. The role of phonons for the inelastic light process in dielectrics is well studied both theoretically and experimentally [1-3] (see also references in [2,3]). The scattering in this case is induced by the dielectric permittivity fluctuations connected with the lattice vibrations. As it was found experimentally in HTSC [4-8], the inelastic light scattering depends almost not all on the frequency transfer in the range $\omega \simeq 10^3 - 10^4$ cm^{-1}. This background was explained by the elecron-impurity interaction [9] and by the electron-phonon interaction [10]. In the paper [11] the phonon resonances was studied in the approximation in which one phonon group scatteres the light and another interacts with electrons. In all these papers the distribution of the incident and scattered light in metal is not considered. We shall see the distribution is especially significant.

The Green's functions method used previously for studying the inelasatic light scattering in normal metals and superconductors [9-12] is very cumbrous for boundary problems. However, the problem under study for a normal metal is essentially semiclassical, because the momentum transfer is smaller than the Fermi momentum and the energy transfer is smaller than the interband transition frequency. We develop a new method using the Boltzmann equation with appropriate boundary conditions. It was used in [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:

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

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 vector potentials of incident end scattered waves, $\gamma_{\alpha\beta}(\mathbf{p})$ is the vertex factor

$$\gamma_{\alpha\beta}(\mathbf{p}) = \left(\delta_{\alpha\beta} + \frac{1}{m}\sum_n \frac{p^{\beta}_{fn}p^{\alpha}_{nf}}{\epsilon_f(\mathbf{p}) - \epsilon_n(\mathbf{p}) + \omega^{(i)}} + \frac{p^{\beta}_{fn}p^{\alpha}_{nf}}{\epsilon_f(\mathbf{p}) - \epsilon_n(\mathbf{p}) - \omega^{(s)}}\right), \qquad (2)$$

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

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

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

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

here $<< ... >>$ 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 $e_\alpha^{(i)}, e_\beta^{(s)}$ are defined by matching of the field in metal to the incident and scattered field in 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 to the time $t - t'$ we apply 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

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

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

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

where $\zeta = \zeta_1 + i\zeta_2$ is the sum of the normal components of the incident and scattered light wave vectors in metal and 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 both impurities and phonons in τ-approximation. The interaction of electrons with the exernal 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 situation the phonons are in an equilibrium state. The collision integral is vanished by the local equilibrium electronic distribution function $f_0(\varepsilon(\mathbf{p},\mathbf{r},t)-\mu)$. The Boltzmann equation (10) has been linearised 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 renormalised:

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

where the brackets denote the integration over the Fermi surface:

$$<\ldots>=2\int(\ldots)\frac{dS}{(2\pi)^3v}.$$

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

The accoustic phonon field obeys the elastic equation:

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

where ρ is the metal density. The last term describes the elecron response on phonons [15].

We apply the specular boundary condition for Boltzmann's equation (10). Conservation of 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 the vanishing of the normal stress tensor components :

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

To solve the above equations we use the even continuation into the $z<0$ half-space for $U(\mathbf{k}_s,z,\omega)$, for the parallel to the surface components of the electric field $E_s(\mathbf{k}_s,z,\omega)$ 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 apply the odd continuation. Hence we can use the Fourier transform with respect to all coordinates, and find the solution of Boltzmann's equation.

The singularities at $z=0$ arised 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 equations respect to z coordinate. This surface contribution have to be determined from the boundary conditions. In the Maxwell equation similar terms result the surface plasmon contribution [13]. They are omitted here.

The Fourier transform of (14) gives

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

where

$$f_i(\mathbf{k},\omega)=-i<\gamma(\mathbf{p})\lambda_{ik}(\mathbf{p})>k_kU(\mathbf{k},\omega)-\omega<\frac{\lambda_{ik}\lambda_{lm}}{-\mathbf{v}\mathbf{k}+i\tau_p^{-1}}>k_kk_mu_l(\mathbf{k},\omega). \tag{17}$$

For the sound frequency range $|\omega| \simeq \omega_D << vk$, 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 of (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 simbol α, is appeared 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 the equation (16). The surface Green's matrix $D^s_{ik}(\mathbf{k}_s, \omega)$ for the boundary condition (15) is determined by the equation:

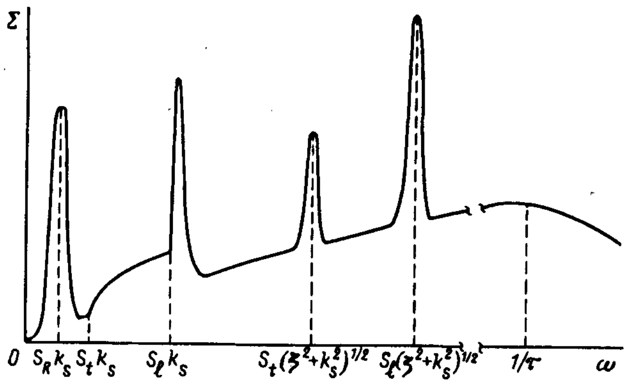
$$\sum_\alpha D^s_{\alpha k}(\mathbf{k}_s, \omega)\lambda_{izlm} \int \frac{dk_z}{2\pi} D^b_{l\alpha}(\mathbf{k}, \omega) k_m k_\alpha e^{ik_z z} = -\delta_{ik} \quad \text{for} \quad z \to 0^+. \tag{19}$$

Thus we obtain the solution of the elastic equation (14),(16). By using the solution of the Boltzmann equation (10) one can find the generalised susceptibility (7) and 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 electone-hole $(e - h)$, phonon bulk $(ph - b)$ and phonon surface $(ph - s)$ contributions.



The electrone-hole contribution (the background in Fig., where the Stokes range is shown only) has the form

$$\Sigma_{e-h}(\mathbf{k}_s, \omega) = -\mathrm{Im} \int \frac{dk_z}{2\pi} |U(\mathbf{k}, \omega)|^2 < \frac{\omega \ |\gamma(\mathbf{p})|^2}{\omega - \mathbf{v}\mathbf{k} + i\tau_p^{-1}} >, \tag{21}$$

which for the isotropic τ and in the limiting case $l|\zeta| << 1$ ($l = v\tau$) takes the form

$$\Sigma_{e-h}(\mathbf{k}_s, \omega) = \frac{\omega\tau}{(\omega\tau)^2 + 1} < |\gamma(\mathbf{p})|^2 > \zeta_2^{-1}. \quad (22)$$

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

The bulk phonon contribution is

$$\Sigma_{ph-b}(\mathbf{k}_s, \omega) = -Im \sum_{\alpha\beta} < \gamma^*(\mathbf{p})\lambda_{\alpha\alpha}(\mathbf{p}) >< \gamma(\mathbf{p})\lambda_{\beta\beta}(\mathbf{p}) > \int \frac{dk_z}{2\pi} |U(\mathbf{k}, \omega)|^2 D^b_{\alpha\beta}(\mathbf{k}, \omega) k_\alpha k_\beta, \quad (23)$$

where we assume that the coordinate axises are the symmetry axises of a cristall, in which the tensor $< \gamma(\mathbf{p})\lambda_{ik}(\mathbf{p}) >$ has a diagonal form. The Green's matrix $D^b_{\alpha\beta}(\mathbf{k}, \omega)$ introduced above has poles determining the bulk phonon dispersion.

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

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

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

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

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

The surface contribution is

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

with

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

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

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

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

$$\Gamma \simeq \tau\omega^2 \quad \text{for } kl << 1 \qquad \text{and} \qquad \Gamma \simeq \frac{s}{v}|\omega| \quad \text{for } kl >> 1.$$

Beside of the Rayleigh pole there are the imaginary part in (25) in the range $|\omega| > s_t k_s$, where the bulk transverse phonons can exist. Here the imaginary part has a form of a narrow continuum. For the range $|\omega| > s_l k_s$ the bulk longitudinal phonons are exist too. The slipping longitudinal phonons give a nonsymmetric (Fano like) resonance. The shape of the resonance has the form:

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

The resonant phenomena discussed here are at low frequency transfer. The relaxation continuum is in wide freguency range.

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