

## Surface excitations in metals: Brillouin and Raman light scattering

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The theory of inelastic light scattering in anisotropic metals by conduction electrons interacting with acoustic and optical phonons and impurities is developed. The effects of a surface are studied in detail. The Coulomb interaction of carriers is included in a self-consistent way. The scattering cross section is evaluated. The skin effect, as well as the electron-phonon interaction, modifies the electron-hole contribution. In particular, the wide relaxation continuum obtained by Zawadowski and Cardona appears with a temperature-dependent relaxation time. Sharp peaks in the cross section arise from the excitation of bulk and surface phonons. The contribution of the mixed phonon excitations, which are a superposition of longitudinal waves diminishing from the surface into the bulk and nondiminishing transverse waves, has the form of a narrow continuum. The slipping nondiminishing longitudinal phonon produces a strong nonsymmetric maximum at the threshold of the density of states.

### I. INTRODUCTION

Much interest in inelastic light scattering (ILS) in metals has been stimulated by the recent observation of this phenomenon in high-temperature superconductors.<sup>1-6</sup> This observation reveals that the present theory of ILS is very fragmentary and cannot provide the correct interpretation of experimental data.

The first theoretical paper<sup>7</sup> on ILS considered the case of clean conventional superconductors. The role of the Coulomb interaction and anisotropy was analyzed in Ref. 8. The ILS in dirty metals was studied in Ref. 9. Several attempts have been made to include the electron-phonon interaction. Phonon resonances were studied in a model in which one phonon group scatters light and another interacts with electrons.<sup>10</sup> The existence of a wide background was explained by strong electron-phonon coupling.<sup>11</sup>

According to the present theory the electron-hole contribution to ILS in a normal metal depends on two parameters:  $\tau^{-1}$  and  $v/\delta \simeq 10^{12}-10^{13} \text{ s}^{-1} \simeq 10^1-10^2 \text{ cm}^{-1}$ , where  $\tau$  is the time of relaxation,  $v$  is the Fermi velocity,  $\delta$  is the skin depth. For a clean metal and at low temperature,  $\tau^{-1} \ll v/\delta$ , the scattering cross section has the maximum at the frequency transfer  $\omega \simeq v/\delta$  and decreases as  $\omega^{-3}$  for larger frequencies. In the opposite case,  $\tau^{-1} \gg v/\delta$ , the maximum occurs at  $\omega \simeq \tau^{-1}$ , while for  $\omega > \tau^{-1}$  the cross section behaves as  $\omega^{-1}$ . An estimate based on various data for HTSC (Ref. 12) gives  $\tau^{-1} \simeq 10^2-10^3 \text{ cm}^{-1}$  at temperature  $T \simeq 100 \text{ K}$ .

In all the theoretical papers mentioned above, the metal surface was ignored, i.e., ILS being considered in the bulk metal. It is clear that this does not correspond to the real experimental situation. The distribution of the incident and scattered light is very important in the optical range. The typical size of electronic fluctuations  $v/\omega$  in this range are of the order of the skin depth. There are also other surface excitations such as phonons, plasmons, and polaritons. Thus the contribution of the sur-

face excitations must be relevant for ILS. A correct theory has to take all these effects into account.

The Green's function method has been used earlier for studying ILS in normal metals and superconductors.<sup>7-11</sup> This approach is very cumbersome when including the boundary. We develop an approach using the Boltzmann's equation with the appropriate boundary conditions.<sup>13-15</sup> This method was used before by one of us<sup>16</sup> for ILS by electron-hole pairs with plasmon excitations. The method is essentially semiclassical. It works provided that the momentum transfer is less than the Fermi momentum, and the energy transfer is less than the interband transition energy.

The role of phonons for ILS in dielectrics has been studied both theoretically and experimentally<sup>17-20</sup> (see also references cited by the authors of Refs. 18 and 19). In Refs. 20-22 the contribution of surface modes in ILS has been considered theoretically for dielectrics and semiconductors. In this case the scattering is induced by dielectric permittivity fluctuations. Our main goal, in this paper, is to how phonons affect ILS in metals.

We will focus on the surface effects here. The interaction of electrons with acoustic and optical phonons is described by a deformation potential. In a polar crystal, lattice vibrations are accompanied by a macroscopic electric field. Consequently, scattering with the polariton excitations can occur. The case of the polar crystal will be considered in a later paper. The electron-electron interaction is usually described by Poisson's equation. Within this approach one loses the surface plasmon dispersion.<sup>13</sup> The surface plasmon contribution contains the small factor  $v/c$ ,  $c$  being the light velocity, thus it is not relevant to our considerations. On the other hand, bulk plasmons exist for  $\omega > \omega_P$ , where  $\omega_P$  is the electronic plasma frequency. This frequency transfer has been considered before.<sup>13</sup>

The outline of this paper is as follows. In Sec. II, the relation between the scattering cross section and the modified density-density correlator is found. The corre-

lator is expressed in terms of the generalized susceptibility, using the fluctuation-dissipation theorem. In Sections III and IV the system of Boltzmann's equation and lattice-motion equation, with the required boundary conditions, is derived. The solution of this system of equations yields the scattering cross section. In Sections V and VI, the cross section is studied and the contributions by electron-hole pairs, bulk phonons, and surface phonons are found. The electron-hole contribution has the form of a wide relaxation continuum. The production or absorption of phonons produce strong peaks. The height of these peaks is finite, because of phonon damping (calculated self-consistently) or because of the spatial dispersion of the incident and scattered fields in the skin layer. The  $\tau$  approximation and the Coulomb interaction effects are discussed in the Appendix.

## II. EFFECTIVE HAMILTONIAN AND SCATTERING CROSS SECTION

We begin with a brief recapitulation of ILS theory. Consider a metal in the half space  $z > 0$ . The effective Hamiltonian for the inelastic electronic light scattering has the form

$$\hat{H} = \frac{e^2}{mc^2} \int d^3r \int \frac{d^3p}{(2\pi)^3} \hat{f}_p(\mathbf{r}, t) \gamma(\mathbf{p}) U(\mathbf{r}, t), \quad (1)$$

where  $\hat{f}_p(\mathbf{r}, t)$  is the electronic density fluctuation operator,  $U(\mathbf{r}, t)$  is the product of the vector potentials of the incident and scattered light (considered below as the external field),

$$A^{(i)}(\mathbf{r}, t) A^{(s)}(\mathbf{r}, t) \simeq U(\mathbf{r}, t) = U(\mathbf{k}_s, z, \omega) \exp[i(\mathbf{k}_s \mathbf{s} - \omega t)], \quad (2)$$

where  $\omega^{(i)}$  and  $\omega^{(s)}$  are the frequencies of the incident and scattered light, respectively, and  $\omega = \omega^{(i)} - \omega^{(s)}$ ,  $\mathbf{k}_s = \mathbf{k}_s^{(i)} - \mathbf{k}_s^{(s)}$ . Here, the subscript  $\mathbf{s}$  denotes the vector components along the surface. The polarization vectors  $\mathbf{e}^{(i)}$ ,  $\mathbf{e}^{(s)}$  of fields  $\mathbf{A}^{(i)}(\mathbf{r}, t)$ ,  $\mathbf{A}^{(s)}(\mathbf{r}, t)$  are included in the vertex factor,

$$\gamma(\mathbf{p}) = e_\alpha^{(i)} e_\beta^{(s)} \left( \delta_{\alpha\beta} + \frac{1}{m} \sum_n \frac{p_{fn}^\beta p_{nf}^\alpha}{\epsilon_f(\mathbf{p}) - \epsilon_n(\mathbf{p}) + \omega^{(i)}} + \frac{p_{fn}^\beta p_{nf}^\alpha}{\epsilon_f(\mathbf{p}) - \epsilon_n(\mathbf{p}) - \omega^{(s)}} \right), \quad (3)$$

representing a sum of two Feynman diagrams describing light scattering.<sup>8,23</sup> The subscript  $f$  denotes the index of the band in which carriers exist, while  $n$  denotes an arbitrary band. The vector  $\mathbf{p}_{fn}$  is the electron momentum matrix element,  $m$  is the electron mass.

The scattering cross section found using Eq. (1) is<sup>16</sup>

$$d\sigma = \left( \frac{8\pi e^2}{m\hbar\omega^{(i)}} \right)^2 \frac{\Sigma(\mathbf{k}_s, \omega)}{1 - \exp(-\omega/T)} \frac{k_z^{(s)} \omega^{(s)} d\omega^{(s)} d\Omega}{c(2\pi)^3}, \quad (4)$$

where  $\Sigma(\mathbf{k}_s, \omega)$  is defined by

$$\Sigma(\mathbf{k}_s, \omega) \propto \int_0^\infty dz dz' K_{\gamma^* \gamma}(\mathbf{k}_s, z, z', \omega) \times U^*(\mathbf{k}_s, z, \omega) U(\mathbf{k}_s, z', \omega), \quad (5)$$

where  $K_{\gamma^* \gamma}(\mathbf{k}_s, z, z', \omega)$  is the Fourier transform of the modified density-density correlation function. The proportionality coefficient is equal to the Bose factor  $[1 - \exp(-\omega/T)]^{-1}$ , which is extracted in Eq. (4). More precisely,  $K_{\gamma^* \gamma}$  is the correlation function of  $\delta \hat{n}_\gamma(\mathbf{r}, t)$ :

$$K_{\gamma^* \gamma}(\mathbf{r}, t; \mathbf{r}', t') = \langle \langle \delta \hat{n}_{\gamma^*}(\mathbf{r}, t) \delta \hat{n}_\gamma(\mathbf{r}', t') \rangle \rangle, \quad (6)$$

where  $\delta \hat{n}_\gamma(\mathbf{r}, t)$  is the modified density fluctuation,

$$\delta \hat{n}_\gamma(\mathbf{r}, t) = \int \frac{d^3p}{(2\pi)^3} \gamma(\mathbf{p}) \hat{f}_p(\mathbf{r}, t), \quad (7)$$

and where  $\langle \langle \rangle \rangle$  denotes the statistical average.

We consider the scattering light outside of the metal and normalize the cross section per incident flux. A straightforward calculation shows<sup>13</sup> that the polarization vector  $\mathbf{e}^{(s)}$  or  $(\mathbf{e}^{(i)})$  has to be replaced by

$$e_x^{(s)} = \frac{\zeta_l^{(s)} (ck_z^{(s)}/\omega^{(s)})}{\zeta_l^{(s)} + \epsilon_{xx}(\omega^{(s)}) k_x^{(s)}},$$

$$e_y^{(s)} = \frac{(k_z^{(s)} \omega^{(s)}/c)^{1/2}}{\zeta_l^{(s)} + k_x^{(s)}},$$

$$e_z^{(s)} = -k_x^{(s)} \frac{\epsilon_{xx}(\omega^{(s)})}{\epsilon_{zz}(\omega^{(s)})} \frac{(ck_z^{(s)}/\omega^{(s)})}{\zeta_l^{(s)} + \epsilon_{xx}(\omega^{(s)}) k_x^{(s)}},$$

where  $\epsilon_{ik}$  is the dielectric function of a metal and  $\zeta_l$ ,  $\zeta_t$  depend on polarizations ( $s$  or  $p$ ) of scattered (incident) light:

$$\zeta_l^{(s)} = \left[ \frac{\epsilon_{xx}(\omega^{(s)})}{\epsilon_{zz}(\omega^{(s)})} \left( \frac{\omega^{(s)2}}{c^2} \epsilon_{zz}(\omega^{(s)}) - k_x^{(s)2} \right) \right]^{1/2},$$

$$\zeta_t^{(s)} = \left[ \frac{\omega^{(s)2}}{c^2} \epsilon_{yy}(\omega^{(s)}) - k_x^{(s)2} \right]^{1/2}. \quad (8)$$

Below, we assume that (i) the transfer momentum  $\mathbf{k}_s$  is directed along the  $x$  axis, (ii) the light incidence is normal ( $k_x^{(i)} = 0$ ), and (iii) the  $x, y, z$  axes are the symmetry axes of the metal.

In order to find the Fourier transform of the correlator (6), we apply the general fluctuation-dissipation theorem:

$$K_{\gamma^* \gamma}(\mathbf{k}_s, z, z', \omega) = \frac{2}{1 - \exp(-\omega/T)} \text{Im} \alpha(\mathbf{k}_s, z, z', \omega), \quad (9)$$

where  $\alpha(\mathbf{k}_s, z, z', \omega)$  is the generalized susceptibility in the external field  $U(\mathbf{k}_s, k_z, \omega)$  (2),

$$\langle \langle \delta \hat{n}_{\gamma^*}(\mathbf{k}_s, z, \omega) \rangle \rangle = 2 \int \frac{d^3p}{(2\pi)^3} \gamma^*(\mathbf{p}) \langle \langle \hat{f}_p(\mathbf{k}_s, z, \omega) \rangle \rangle$$

$$= - \int_0^\infty dz' \alpha(\mathbf{k}_s, z, z', \omega) U(\mathbf{k}_s, z', \omega). \quad (10)$$

### III. BOLTZMANN'S EQUATION

According to Eqs. (9),(10) one needs to evaluate the electronic distribution function  $f_p(\mathbf{k}_s, z, \omega) = \langle \langle \hat{f}_p(\mathbf{k}_s, z, \omega) \rangle \rangle$ . We apply the Boltzmann equation,

$$\frac{\partial f_p(\mathbf{r}, t)}{\partial t} + \mathbf{v} \frac{\partial f_p(\mathbf{r}, t)}{\partial \mathbf{r}} + \dot{\mathbf{p}} \frac{\partial f_p(\mathbf{r}, t)}{\partial \mathbf{p}} = \hat{S}t f_p(\mathbf{r}, t), \quad (11)$$

where  $\hat{S}t f_p(\mathbf{r}, t)$  is the collision integral with both impurities and phonons.

Let us write the local electron spectrum of the deformed lattice as follows:

$$\varepsilon(\mathbf{p}, \mathbf{r}, t) = \varepsilon_0(\mathbf{p}) + \gamma(\mathbf{p})U(\mathbf{r}, t) + \lambda_{ik}(\mathbf{p})u_{ik}(\mathbf{r}, t) + \xi_i(\mathbf{p})w_i(\mathbf{r}, t), \quad (12)$$

where  $\varepsilon_0(\mathbf{p})$  is the spectrum for the undeformed lattice and the deformation tensor is

$$u_{ik} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} \right),$$

where  $u_i$  and  $w_i$  are the acoustic and optical displacements, respectively. We assume for simplicity that the unit cell contains two atoms. The electron-acoustic-phonon interaction, as well as the electron-optical-phonon interaction is described in Eq. (12) by the deformation potential.

Let us linearize Eq. (11) using

$$f_p(\mathbf{r}, t) = f_0[\varepsilon(\mathbf{p}, \mathbf{r}, t) - \mu] + \frac{df_0}{d\varepsilon} \delta f_p(\mathbf{r}, t), \quad (13)$$

where  $f_0[\varepsilon(\mathbf{p}, \mathbf{r}, t) - \mu]$  is the Fermi-Dirac local distribution function. It is significant that the collision integral is cancelled by the local-equilibrium term in (13).

We write the collision integral in the  $\tau$  approximation,

$$\hat{S}t f_p(\mathbf{r}, t) = -\frac{1}{\tau_p} \frac{df_0}{d\varepsilon} \delta f_p(\mathbf{r}, t), \quad (14)$$

where  $\tau_p$  is the time of relaxation for collisions with both impurities and phonons. In the Appendix we consider the impure metal, beyond the  $\tau$  approximation.

Defining the chemical potential by means of the charge-conservation condition,

$$\int \frac{d^3p}{(2\pi)^3} f_0(\varepsilon - \mu) = \int \frac{d^3p}{(2\pi)^3} f_0(\varepsilon_0 - \mu_0), \quad (15)$$

we obtain

$$\mu(\mathbf{r}, t) = \mu_0 + [\langle \gamma(\mathbf{p}) \rangle U(\mathbf{r}, t) + \langle \lambda_{ik}(\mathbf{p}) \rangle u_{ik}(\mathbf{r}, t) + \langle \xi_i(\mathbf{p}) \rangle w_i(\mathbf{r}, t)] / \langle 1 \rangle, \quad (16)$$

where the brackets denote the integration over the Fermi surface:

$$\langle \rangle = 2 \int (\cdot) \frac{dS_F}{(2\pi)^3 v},$$

since we assume that the electrons are degenerate. The condition (15) expresses Debye screening for frequencies

less than  $\omega_P$ . It implies the renormalization of vertices  $\gamma(\mathbf{p}), \lambda_{ik}(\mathbf{p}), \xi_i(\mathbf{p})$ , in (12):

$$\begin{aligned} \gamma(\mathbf{p}) &\rightarrow Y(\mathbf{p}) = \gamma(\mathbf{p}) - \langle \gamma(\mathbf{p}) \rangle / \langle 1 \rangle, \\ \lambda_{ik}(\mathbf{p}) &\rightarrow \Lambda_{ik}(\mathbf{p}) = \lambda_{ik}(\mathbf{p}) - \langle \lambda_{ik}(\mathbf{p}) \rangle / \langle 1 \rangle, \\ \xi_i(\mathbf{p}) &\rightarrow \Xi_i(\mathbf{p}) = \xi_i(\mathbf{p}) - \langle \xi_i(\mathbf{p}) \rangle / \langle 1 \rangle. \end{aligned} \quad (17)$$

The linearized Boltzmann equation has the form

$$\begin{aligned} \frac{\partial \delta f_p(\mathbf{r}, t)}{\partial t} + \mathbf{v} \frac{\partial \delta f_p(\mathbf{r}, t)}{\partial \mathbf{r}} + \frac{\delta f_p(\mathbf{r}, t)}{\tau_p} \\ = -\frac{\partial}{\partial t} [Y(\mathbf{p})U(\mathbf{r}, t) + \Lambda_{ik}(\mathbf{p})u_{ik}(\mathbf{r}, t) \\ + \Xi_i(\mathbf{p})w_i(\mathbf{r}, t)] - e\mathbf{v} \cdot \mathbf{E}(\mathbf{r}, t). \end{aligned} \quad (18)$$

A term proportional to  $\nabla \mu(\mathbf{r}, t)$  in Eq. (18) is included into the electric field  $\mathbf{E}(\mathbf{r}, t)$  representing the electron-electron interaction. The above equations have to be supplemented by Maxwell's equations for  $\mathbf{E}(\mathbf{r}, t)$  and the corresponding boundary conditions.<sup>13</sup> However, our calculation shows that the Coulomb interaction results only in the renormalization (17), for the frequency transfers we are interested in, namely,  $\omega \ll \omega_P$  (see Appendix). Therefore, we omit the term  $e\mathbf{v} \cdot \mathbf{E}(\mathbf{r}, t)$ .

We take the boundary condition for Boltzmann's equation (18) at  $z = 0$ ,

$$(f_p(\mathbf{r}, \omega))_{v_z > 0} = (f_p(\mathbf{r}, \omega))_{v_z < 0},$$

$$\varepsilon(\mathbf{p}_s, v_z > 0, \mathbf{r}, \omega) = \varepsilon(\mathbf{p}_s, v_z < 0, \mathbf{r}, \omega), \quad (19)$$

whose form describes the specular reflection of electrons. A more realistic boundary condition does not affect the final results in an essential way.<sup>24</sup>

### IV. EQUATIONS FOR PHONON DISPLACEMENTS

The phonon fields in the long-wave approximation obey the following equations

$$\begin{aligned} -\lambda_{iklm} \frac{\partial^2 u_l(\mathbf{r}, \omega)}{\partial x_k \partial x_m} - \rho \omega^2 u_i(\mathbf{r}, \omega) \\ = 2 \frac{\partial}{\partial x_k} \int \frac{d^3p}{(2\pi)^3} \lambda_{ik}(\mathbf{p}) f_p(\mathbf{r}, \omega), \end{aligned} \quad (20)$$

$$\begin{aligned} -\mu_{iklm} \frac{\partial^2 w_l(\mathbf{r}, \omega)}{\partial x_k \partial x_m} + \rho' \chi_{il} w_l(\mathbf{r}, \omega) - \rho' \omega^2 w_i(\mathbf{r}, \omega) \\ = -2 \int \frac{d^3p}{(2\pi)^3} \xi_i(\mathbf{p}) f_p(\mathbf{r}, \omega), \end{aligned} \quad (21)$$

where  $\rho$  is the metal density and  $\rho'$  is the reduced mass density. The right-hand sides of these equations represent the electron-phonon interaction. Equation (20) is known as the equation of the dynamical theory of elasticity (see the review<sup>25</sup> and references cited therein). The derivation of Eq. (21) is straightforward and will be given elsewhere.

The boundary conditions for Eqs. (20) are determined by the fact that the normal component of the stress tensor must vanish at the surface  $z = 0^+$ . They are

$$\lambda_{izlm} \frac{\partial u_l(\mathbf{r}, \omega)}{\partial x_m} + 2 \int \frac{d^3p}{(2\pi)^3} \lambda_{iz}(\mathbf{p}) f_p(\mathbf{r}, \omega) = 0, \quad (22)$$

$$\mu_{izlm} \frac{\partial w_l(\mathbf{r}, \omega)}{\partial x_m} = 0. \quad (23)$$

The last term in Eq. (22) represents the pressure on the surface inside the metal due to the electron-phonon interaction.

We use the even continuation in the  $z < 0$  half space for  $U(\mathbf{k}_s, z, \omega)$  and for the components parallel to the surfaces of the phonon displacements  $u_s(\mathbf{k}_s, z, \omega)$ ,  $w_s(\mathbf{k}_s, z, \omega)$ . For the perpendicular components  $u_z(\mathbf{k}_s, z, \omega)$ ,  $w_z(\mathbf{k}_s, z, \omega)$ , we use the odd continuation.

Taking the Fourier transform with respect to all coordinates, we obtain the solution to Boltzmann's equation (18):

$$\delta f_p(\mathbf{k}, \omega) = -\frac{\omega}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau_p^{-1}} [Y(\mathbf{p})U(\mathbf{k}, \omega) + \Lambda_{ik}(\mathbf{p})u_{ik}(\mathbf{k}, \omega) + \Xi_i(\mathbf{p})w_i(\mathbf{k}, \omega)]. \quad (24)$$

One can see that expression (24) satisfies the boundary condition (19), since

$$\delta f_p(\mathbf{k}_s, z = 0, \omega) = \int \frac{dk_z}{2\pi} \delta f_p(\mathbf{k}_s, k_z, \omega),$$

and we can change the signs of  $p_z$  and  $k_z$  conserving the value of the integral, because the vectors and tensors  $v_i$ ,  $Y(\mathbf{p})$ ,  $\Lambda_{ik}(\mathbf{p})$ ,  $\Xi_i(\mathbf{p})$  each has a definite parity.

There are singularities at  $z = 0$ , which arise in Eq. (20), (21) after the continuing into the  $z < 0$  half space and which are due to the terms containing derivatives with respect to  $z$ . Using Eqs. (13), (24), we get the Fourier transform of Eq. (20),

$$\begin{aligned} & (\lambda_{\alpha klm} k_k k_m - \rho \omega^2 \delta_{\alpha l}) u_l(\mathbf{k}, \omega) \\ &= - \left\langle \frac{\Lambda_{\alpha k}(\mathbf{p}) \Lambda_{lm}(\mathbf{p}) (\mathbf{v} \cdot \mathbf{k} - i\tau_p^{-1})}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau_p^{-1}} \right\rangle k_k k_m u_l(\mathbf{k}, \omega) \\ & \quad - i \langle Y(\mathbf{p}) \Lambda_{\alpha k}(\mathbf{p}) \rangle k_k U(\mathbf{k}, \omega) + k_\alpha C_\alpha^{\text{ac}}(\mathbf{k}_s, \omega). \end{aligned} \quad (25)$$

The first term on the right-hand side gives the renormalization of the elastic constants,

$$\lambda_{iklm} \rightarrow \lambda_{iklm} - \langle \Lambda_{ik}(\mathbf{p}) \Lambda_{lm}(\mathbf{p}) \rangle$$

and the damping (see for example, Ref. 25)

$$\Gamma_{\text{ac}}(\mathbf{k}) \simeq \frac{s^2 k}{v} \min(kl, 1). \quad (26)$$

For these estimates we use  $\Lambda(\mathbf{p}) \simeq \varepsilon_F$ ,  $\rho \simeq v p_F^4 / 3\pi^2 s^2$ , where  $s$  is on the order of the sound velocity.

The sound dispersion relation is

$$\omega = \omega_{\text{ac}}(\mathbf{k}) - i\Gamma_{\text{ac}}(\mathbf{k}), \quad (27)$$

where  $\omega_{\text{ac}}(\mathbf{k})$  is an eigenvalue of the matrix in the left-hand side of Eq. (25).

The second term on the right-hand side of Eq. (25) represents the response to the external field  $U(\mathbf{k}, \omega)$ . This simple form is obtained by ignoring the terms  $\simeq \omega/vk \simeq s/v$ . The third term on the right-hand side of Eq. (25) reveals  $\delta$ -function singularities at  $z = 0$  and represents

the surface effect. There is no sum over the Greek symbol  $\alpha$ . Note that the term containing  $k_z$  arises only for the equation with  $\alpha = z$ . The constants  $C_\alpha^{\text{ac}}(\mathbf{k}_s, \omega)$  have to be defined from the boundary condition (22). The term proportional to  $w_i(\mathbf{k}, \omega)$  is omitted in Eq. (25), since it is of the order of  $s/v$ .

Using the bulk Green's matrix  $D_{ik}^{(\text{ac}-b)}$ , which obeys the equation

$$(\lambda_{iklm} k_k k_m - \rho \omega^2 \delta_{il}) D_{ln}^{(\text{ac}-b)}(\mathbf{k}, \omega) = \delta_{in}, \quad (28)$$

we find the solutions of Eq. (25),

$$\begin{aligned} u_l(\mathbf{k}, \omega) = & \sum_{\alpha} D_{l\alpha}^{(\text{ac}-b)}(\mathbf{k}, \omega) k_\alpha C_\alpha^{\text{ac}}(\mathbf{k}_s, \omega) \\ & + D_{lk}^{(\text{ac}-b)}(\mathbf{k}, \omega) f_k^{\text{ac}}(\mathbf{k}, \omega), \end{aligned} \quad (29)$$

where the force

$$f_i^{\text{ac}}(\mathbf{k}, \omega) = -i \langle Y(\mathbf{p}) \Lambda_{ik}(\mathbf{p}) \rangle k_k U(\mathbf{k}, \omega). \quad (30)$$

We do not write the terms with the damping in Eq. (28), as we have included it in  $\omega$  through the formula (27).

Substituting Eq. (29) into the boundary condition (22), one finds

$$\begin{aligned} C_\alpha^{\text{ac}}(\mathbf{k}_s, \omega) = & D_{\alpha i}^{(\text{ac}-s)}(\mathbf{k}_s, \omega) \int \frac{dk_z}{2\pi} \left( \lambda_{izlm} \right. \\ & \left. + \left\langle \frac{\omega \Lambda_{iz} \Lambda_{lm}}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau_p^{-1}} \right\rangle \right) \\ & \times D_{lk}^{(\text{ac}-b)}(\mathbf{k}, \omega) f_k^{\text{ac}}(\mathbf{k}, \omega) k_m, \end{aligned} \quad (31)$$

where the surface Green's matrix  $D_{\alpha k}^{(\text{ac}-s)}(\mathbf{k}_s, \omega)$  obeys the equation,

$$\begin{aligned} \sum_{\alpha} D_{\alpha k}^{(\text{ac}-s)}(\mathbf{k}_s, \omega) \int \frac{dk_z}{2\pi} D_{l\alpha}^{(\text{ac}-b)}(\mathbf{k}, \omega) k_m k_\alpha e^{ik_z z} \\ \times \left( \lambda_{izlm} + \left\langle \frac{\omega \Lambda_{iz} \Lambda_{lm}}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau_p^{-1}} \right\rangle \right) = -\delta_{ik}, \end{aligned} \quad (32)$$

for  $z \rightarrow 0^+$ . The second term in the parenthesis gives the additional contribution to the surface phonon damping. The matrix Green's functions  $D_{ik}^{(b)}$ ,  $D_{ik}^{(s)}$  contain all the information about the phonon spectrum of a semi-infinite metal sample.

The above considerations for the acoustic phonons can be extended to optical phonons. The solution of Eq. (21) for the optical displacement is like Eqs. (28) and (29). Equations (28)–(32) can be rewritten with the substitutions:

$$\begin{aligned} \lambda_{iklm} k_k k_m - \rho \omega^2 \delta_{il} & \rightarrow \rho' \chi_{il} + \mu_{iklm} k_k k_m - \rho' \omega^2 \delta_{il}, \\ f_i^{\text{ac}} & \rightarrow f_i^{\text{op}} = - \left\langle \frac{Y(\mathbf{p}) \Xi_i(\mathbf{p}) (\mathbf{v} \cdot \mathbf{k} - i\tau_p^{-1})}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau_p^{-1}} \right\rangle U(\mathbf{k}, \omega). \end{aligned} \quad (33)$$

The optical frequencies are renormalized by the electron-

phonon interaction:

$$\chi_{ik} \rightarrow \chi_{ik} - \langle \Xi_i(\mathbf{p}) \Xi_k(\mathbf{p}) \rangle / \rho'.$$

The interaction of the optical phonons with electrons gives the phonon damping. Using the estimate  $\Xi(\mathbf{p}) \simeq \varepsilon_{FPF}$ , we obtain

$$\Gamma_{\text{op}}(\mathbf{k}) \simeq \frac{\omega_D^2}{vk} \min(kl, 1) \quad \text{for } \omega \ll vk, \quad (34)$$

$$\Gamma_{\text{op}}(\mathbf{k}) \simeq \frac{\omega^2 \tau}{\omega^2 \tau^2 + 1} \quad \text{for } \omega \gg vk. \quad (35)$$

Hence the optical-phonon dispersion  $\omega = \omega_{\text{op}}(\mathbf{k}) - i\Gamma_{\text{op}}(\mathbf{k})$  in the long-wave limit is estimated to be

$$\omega_{\text{op}}^2 = \omega_D^2 + ak^2 \simeq \chi + \frac{\mu}{\rho'} k^2, \quad (36)$$

where the constants  $\omega_D$  and  $|a| \simeq s^2$  depend on the polarization and the direction of wave propagation.

## V. BULK ELECTRON-HOLE AND PHONON EXCITATIONS

Since the distribution function (13), (24) and the fields  $w_l(\mathbf{k}, \omega)$ ,  $w_i(\mathbf{k}, \omega)$  (29) are determined, we can derive the response (10) with the help of the expression

$$\int \frac{d^3p}{(2\pi)^3} \gamma^*(\mathbf{p}) [f_p(\mathbf{k}, \omega) - f_0(\varepsilon_0 - \mu_0)].$$

Using (13) and taking only linear terms in  $\varepsilon(\mathbf{p}, \mathbf{k}, \omega) - \varepsilon_0(\mathbf{p})$ ,  $\mu(\mathbf{k}, \omega) - \mu_0$  [see Eqs. (12), (16)], we obtain [see Eqs. (5), (9)] the scattering cross section,

$$\begin{aligned} \Sigma(\mathbf{k}_s, \omega) = & -\text{Im} \int \frac{dk_z}{2\pi} U^*(\mathbf{k}, \omega) \\ & \times \left( \left\langle \frac{\omega |Y(\mathbf{p})|^2}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau_p^{-1}} \right\rangle U(\mathbf{k}, \omega) \right. \\ & - i \langle Y^*(\mathbf{p}) \Lambda_{ik}(\mathbf{p}) \rangle k_i u_k(\mathbf{k}, \omega) \\ & \left. - \langle Y^*(\mathbf{p}) \Xi_i(\mathbf{p}) \rangle w_i(\mathbf{k}, \omega) \right). \quad (37) \end{aligned}$$

The last two terms in Eq. (37) are written for most interesting range  $\omega \sim \omega_D \ll vk$ .

The expression (37) is our main result. We discuss the different terms in this formula as follows.

### A. Electron-hole excitations

The electron-hole contribution [see Fig. 1(a)] is represented by the first term in Eq. (37):

$$\Sigma_{e-h}(\mathbf{k}_s, \omega) = \left\langle |Y(\mathbf{p})|^2 \frac{\delta(\nu)}{v} \right\rangle \begin{cases} \frac{\omega}{|\zeta|^2} \left( \ln \frac{|\zeta|v}{\omega} + \pi \frac{\zeta_1}{\zeta_2} \right) & \text{for } v|\zeta| \gg \omega, \\ \frac{|\zeta|^2 v^4}{\omega^3} & \text{for } v|\zeta| < \omega. \end{cases} \quad (41)$$

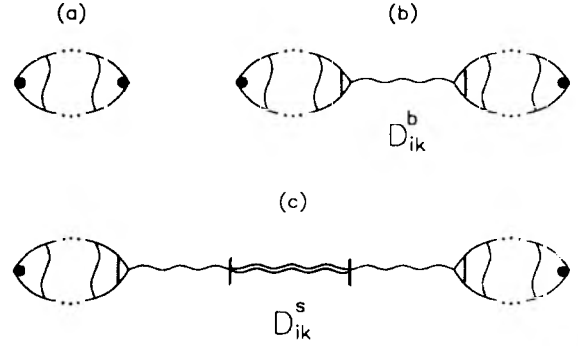


FIG. 1. Diagram representation of the correlator  $K_{\gamma^* \gamma}$ . (a) The electron-hole contribution accounting the scattering by phonons (wave lines); the black dot [vertex  $Y(\mathbf{p})$ ] shows the electron interaction with the incident and scattered light. (b) The bulk (acoustic- or optical-) phonon contribution; the electron-phonon interaction is shown by the empty vertex. The wave line corresponds to the bulk phonon Green's function ( $D_{ik}^{(\text{ac}-b)}$  or  $D_{ik}^{(\text{op}-b)}$ ). (c) The surface-phonon contribution. The surface phonon Green's function ( $D_{ik}^{(\text{ac}-s)}$  or  $D_{ik}^{(\text{op}-s)}$ ) is shown by the double wave line. The dots mean the diagram chain summed to all orders.

$$\begin{aligned} \Sigma_{e-h}(\mathbf{k}_s, \omega) = & -\text{Im} \int \frac{dk_z}{2\pi} |U(\mathbf{k}, \omega)|^2 \\ & \times \left\langle \frac{\omega |Y(\mathbf{p})|^2}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau_p^{-1}} \right\rangle, \quad (38) \end{aligned}$$

where, for normal skin-effect conditions for the incident and scattered light,<sup>13</sup>

$$U(\mathbf{k}, \omega) = \frac{2i\zeta}{\zeta^2 - k_z^2}, \quad \text{where } \zeta = \zeta_1 + i\zeta_2 = \zeta^{(i)} + \zeta^{(s)}. \quad (39)$$

Here, we omit the polarization subscript [see Eq. (8)].

In the pure limit  $l|\zeta| \gg 1$ , we can ignore  $i\tau_p^{-1}$  in the denominator of Eq. (38):

$$\Sigma_{e-h}(\mathbf{k}_s, \omega) = \pi\omega \left\langle |Y(\mathbf{p})|^2 \frac{\delta(\nu)}{v} \right\rangle \int_{k_0}^{\infty} \frac{dk_z}{2\pi} \frac{|U(\mathbf{k}, \omega)|^2}{k}, \quad (40)$$

where  $k_0$  is on the order of  $\omega/v$  and  $\nu = \mathbf{v} \cdot \mathbf{k}/vk$ . We find for the normally incident and scattered light

Such an expression was first obtained<sup>7</sup> for superconductors by applying the Green's functions method.

In the opposite limit  $l|\zeta| \ll 1$  and for isotropic  $\tau_p$ , Eq. (38) takes the form

$$\Sigma_{e-h}(\mathbf{k}_s, \omega) = \frac{\omega\tau}{(\omega\tau)^2 + 1} \langle |Y(\mathbf{p})|^2 \rangle \zeta_2^{-1}. \quad (42)$$

This contribution has the form of a wide relaxation background, it is shown in Figs. 2, 3 for the Stokes range.

Note that there is the strong dependence of  $\Sigma$  on the skin depth  $\delta \simeq 2\zeta_2^{-1}$ . In the limit  $\zeta_2 \rightarrow 0$ , the skin depth should be replaced by the sample thickness. The frequency dependence (42) is the same as for the case of electron-impurity interaction.<sup>9</sup> For high temperatures, the collision rate  $\tau^{-1}$  is determined by the electron-phonon interaction. Our results differ somewhat from those obtained in Ref. 11, where some factor violates the sum rule. If the temperature is larger than  $\omega_D/3$ , the collision integral gives<sup>26</sup>  $\tau^{-1} = 2\pi gT$  (limits of validity of this temperature behavior were found out in the numerical calculations.<sup>27</sup> Here,  $g$  is the dimensionless constant of the electron-phonon interaction (about the definition  $g$  see Ref. 28. For low temperatures  $T \ll \omega_D$ , the scattering rate  $\tau^{-1} \simeq gT^3/\omega_D^2$ .<sup>26</sup>

Our semiclassical derivation of  $\Sigma$  is valid as long as  $\omega \ll T$ . Otherwise the quantum mechanical response theory has to be used. Because of the temperature  $T$  is implicated together with  $\omega$  in sums over discrete frequencies, the obvious substitution can be made,<sup>29</sup>

$$\tau^{-1} \simeq \begin{cases} 2\pi g\omega_D & \text{for } \omega \gg (\omega_D/3, T), \\ g\omega^3/\omega_D^2 & \text{for } \omega_D/3 \gg \omega \gg T. \end{cases} \quad (43)$$

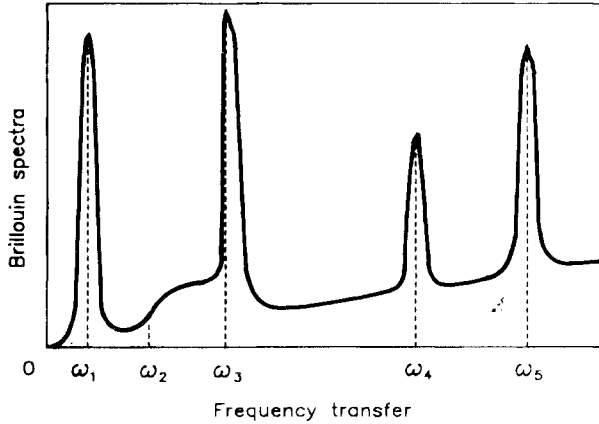


FIG. 2. The Brillouin light scattering cross section with excitation of acoustic phonons. The background is from the electron-hole contribution [Fig. 1(a)].  $\omega_1 = \omega_{ac-s}(\mathbf{k}_s)$  — the Rayleigh phonon peak;  $\omega_3 = \omega_{ac-l}(\mathbf{k}_s, k_z = 0)$  — the peak of the longitudinal phonon slipping along the metal surface.  $\omega_4 = \omega_{ac-l}(\mathbf{k}_s, k_z = \zeta_1)$ ,  $\omega_5 = \omega_{ac-l}(\mathbf{k}_s, k_z = \zeta_1)$  — the bulk transverse and longitudinal phonon peaks, respectively. The pile between  $\omega_2 = \omega_{ac-t}(\mathbf{k}_s, k_z = 0)$  and  $\omega_3$  is the contribution of the coupled transverse and longitudinal waves; the longitudinal waves decay from the boundary (like Rayleigh's wave), while the transverse waves do not decay.

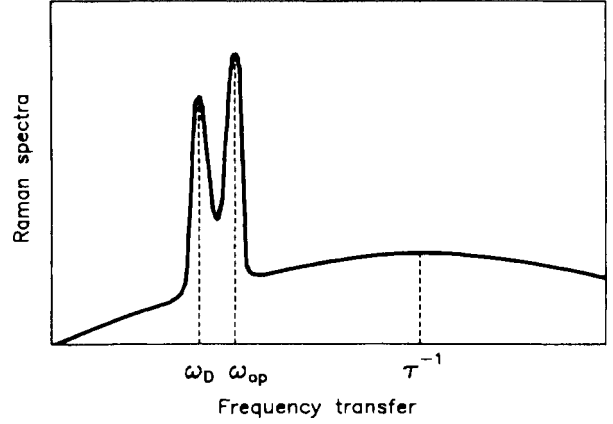


FIG. 3. The Raman scattering cross section for the normally incident and scattered light with excitation of optic phonons. The Brillouin peaks represented by Fig. 2 are omitted here. The entire electron-hole background is shown. The bulk optical phonon contribution is resolved into two peaks. One is conditioned by the singularity of phonon density of states at the threshold  $\omega_D$ . Another at  $\omega_{op} = \omega_{op-b}(\mathbf{k}_s = 0, k_z = \zeta_1)$ , is determined by the momentum and energy conservation.

## B. Acoustic bulk phonons

If we substitute the term proportional to  $f_k^{ac}(\mathbf{k}, \omega)$  from (29) into (37) for  $u_k(\mathbf{k}, \omega)$ , we obtain the acoustic bulk phonon contribution:

$$\begin{aligned} \Sigma_{ac-b}(\mathbf{k}_s, \omega) &= \text{Im} \sum_{\alpha\beta} \langle Y^*(\mathbf{p}) \Lambda_{\alpha\alpha}(\mathbf{p}) \rangle \\ &\times \langle Y(\mathbf{p}) \Lambda_{\beta\beta}(\mathbf{p}) \rangle \int \frac{dk_z}{2\pi} |U(\mathbf{k}, \omega)|^2 \\ &\times D_{\alpha\beta}^{(ac-b)}(\mathbf{k}, \omega) k_\alpha k_\beta. \end{aligned} \quad (44)$$

The bulk matrix Green's function  $D_{ik}^{(ac-b)}(\mathbf{k}, \omega)$  has poles determining the bulk phonon dispersion  $\omega_{ac-b}^{(n)}(\mathbf{k}, \omega)$ , where  $n = 1, 2, 3$ . The imaginary part in (44) comes from the damping  $\Gamma_{ac}$  (26). Using the matrix Green's function,

$$D_{ik}^{(ac-b)}(\mathbf{k}, \omega) = \frac{1}{\rho} \sum_n \frac{u_i^{(n)} u_k^{(n)*}}{\omega_{ac-b}^{(n)2}(\mathbf{k}) - (\omega + i0)^2}, \quad (45)$$

where  $u_i^{(n)}$  are the eigenvectors of the homogeneous Eq. (28), one obtains the scattering cross section for the case of the infinitesimal damping:

$$\begin{aligned} \Sigma_{ac-b}(\mathbf{k}_s, \omega) &= \text{sgn } \omega \frac{\pi}{\rho} \sum_s \int \frac{dk_z}{2\pi} \\ &\times |U(\mathbf{k}, \omega) \langle Y(\mathbf{p}) \Lambda_{ik}(\mathbf{p}) \rangle u_i^{(n)} k_k|^2 \\ &\times \delta[\omega^2 - \omega_{ac-b}^{(n)2}(\mathbf{k})]. \end{aligned}$$

Note this form has the positive sign for  $\omega > 0$  and vice versa. The tensor  $\langle Y(\mathbf{p}) \Lambda_{ik}(\mathbf{p}) \rangle$  is diagonal if the symmetry axes of a crystal are used as coordinates.

We consider the case of normally incident and scattered light in detail for the finite damping. Here, Eq. (44) gives

$$\begin{aligned} \Sigma_{ac-b}(\omega) &= |\langle Y(\mathbf{p})\Lambda_{zz}(\mathbf{p}) \rangle|^2 \text{Im} \int \frac{dk_z}{2\pi\rho} \frac{|U(\mathbf{k}, \omega)|^2 k_z^2}{s_l^2 k_z^2 - (\omega + i\Gamma_{ac})^2}, \\ & \quad (46) \end{aligned}$$

where  $s_l = (\lambda_{zzzz}/\rho)^{1/2}$  is the longitudinal sound velocity in the  $z$  direction. Note that there is no contribution from transverse phonons.

The integrand (46) has a maximum at  $|k_z| = \zeta_1$ , with width  $\zeta_2$  and a maximum at  $k_z = \pm|\omega|/s_l$ , with width  $\Gamma_{ac}/s_l$ . If  $\zeta_1 \gg \zeta_2 \gg \Gamma_{ac}/s_l$ , we integrate only the denominator, taking  $U(\mathbf{k}, \omega)$  at  $k_z = \omega/s_l$ . We get

$$\Sigma_{ac-b}(\omega) = \frac{\zeta_1 |\langle Y(\mathbf{p})\Lambda_{zz}(\mathbf{p}) \rangle|^2}{2\rho[|\omega| - s_l\zeta_1]^2 + s_l^2\zeta_2^2} \text{sgn } \omega. \quad (47)$$

This expression has the form of a sharp peak at  $|\omega| = s_l\zeta_1$ , with width  $s_l\zeta_2$ . The comparison of Eq. (47) with Eq. (38) shows that the ratio  $\Sigma_{ac-b}^{\max}/\Sigma_{e-h}^{\max} \simeq \zeta_1/\zeta_2 \gg 1$ . If  $\zeta_1 \leq \zeta_2$ , the broadening of the peak makes it impossible to observe experimentally.

If  $\zeta_2 \ll (\Gamma_{ac}/s_l, \zeta_1)$ , we integrate only  $U(\mathbf{k}, \omega)$ , taking the denominator at  $k_z = \zeta_1$ :

$$\Sigma_{ac-b}(\omega) = \frac{\zeta_1 \Gamma_{ac} |\langle Y(\mathbf{p})\Lambda_{zz}(\mathbf{p}) \rangle|^2}{2\rho\zeta_2 s_l [|\omega| - s_l\zeta_1]^2 + \Gamma_{ac}^2} \text{sgn } \omega. \quad (48)$$

The peak is sharp since the damping  $\Gamma_{ac}$  (26), which should be considered at  $k \simeq \zeta_1$ , is small. Now we find the ratio  $\Sigma_{ac-b}^{\max}/\Sigma_{e-h}^{\max} \simeq \zeta_1 s_l/\Gamma_{ac} \simeq v/s$ .

For the nonperpendicular scattering, transverse phonons are excited. The height of corresponding peaks is proportional to  $k_s^2$ . The relative height of the transverse and longitudinal peaks is  $\Sigma_{ac-t}^{\max}/\Sigma_{ac-l}^{\max} \simeq \min(k_s^2/\zeta_1^2, \zeta_1^2/k_s^2)$ . All the bulk peaks are located at  $|\omega| = \omega_{ac}^l(\mathbf{k}_s, k_z = \zeta_1)$ .

### C. Optical bulk phonons

The contribution of the optical bulk phonons to the cross section is contained in the last term in Eq. (37),

$$\begin{aligned} \Sigma_{op-b}(\mathbf{k}_s, \omega) &= \text{Im} \langle Y^*(\mathbf{p})\Xi_i(\mathbf{p}) \rangle \\ & \quad \times \langle Y(\mathbf{p})\Xi_i(\mathbf{p}) \rangle \int \frac{dk_z}{2\pi} |U(\mathbf{k}, \omega)|^2 \\ & \quad \times D_{ik}^{(op-b)}(\mathbf{k}, \omega). \end{aligned} \quad (49)$$

First, we consider the case when the vector  $\langle Y(\mathbf{p})\Xi_i(\mathbf{p}) \rangle$  lies in  $z$  direction. Equation (49) gives for normally incident and scattered light

$$\begin{aligned} \Sigma_{op-b}(\omega) &= |\langle Y(\mathbf{p})\Xi_z(\mathbf{p}) \rangle|^2 \text{Im} \int \frac{dk_z}{2\pi\rho'} \\ & \quad \times \frac{|U(\mathbf{k}, \omega)|^2}{ak_z^2 + \omega_D^2 - (\omega + i\Gamma_{op})^2}. \end{aligned} \quad (50)$$

The denominator has a minimum at  $k_z = k_{\min}$ , with width  $\Delta k_z$ ,

$$k_{\min}^2 = (\omega^2 - \omega_D^2)/a,$$

$$\Delta k_z = |\omega\Gamma_{op}/[a(\omega^2 - \omega_D^2)]^{1/2}|, \quad (51)$$

where  $\Gamma_{op}$  is given by Eq. (34), upon substituting  $k = \zeta_1$ .

Let us consider two limiting cases for the typical situation,  $\zeta_1 \gg \zeta_2$ . Below we take  $\omega > 0$  in order to write the formulas in the clearest way; for  $\omega < 0$ , one substitutes  $\omega \rightarrow |\omega|$  and changes the sign of  $\Sigma$ .

(i) The damping is relatively large,  $\Delta k_z \gg \zeta_2$ . Then we integrate only  $|U|^2$ , taking the denominator at  $k_z = \zeta_2$ :

$$\Sigma_{op-b}(\omega) = \frac{\Gamma_{op} |\langle Y(\mathbf{p})\Xi_z(\mathbf{p}) \rangle|^2}{2\rho'\zeta_2\omega_D \{[\omega - (\omega_D^2 + a\zeta_1^2)^{1/2}]^2 + \Gamma_{op}^2\}}. \quad (52)$$

We see that the peak is at  $\omega = (\omega_D^2 + a\zeta_1^2)^{1/2}$ . Our estimate of the peak height gives  $\Sigma_{op-b}^{\max}/\Sigma_{e-h}^{\max} \simeq \omega_D/\Gamma_{op}$ .

(ii) The damping is small,  $\Delta k_z \ll \zeta_2$ . Integrating only the denominator and taking  $|U|^2$  at  $k_z = k_{\min}$ , one finds

$$\begin{aligned} \Sigma_{op-b}(\omega) &= \frac{|\langle Y(\mathbf{p})\Xi_z(\mathbf{p}) \rangle|^2}{2a\rho'(2\omega_D)^{1/2} [(k_{\min} - \zeta_1)^2 + \zeta_2^2]} \\ & \quad \times \text{Re} \left( \frac{a}{\omega - \omega_D + i\Gamma_{op}} \right)^{1/2}. \end{aligned} \quad (53)$$

Note that

$$\begin{aligned} \text{Re} \left( \frac{a}{\omega - \omega_D + i\Gamma} \right)^{1/2} \\ = |a|^{1/2} \left( \frac{(\omega - \omega_D) \text{sgn } a + [(\omega - \omega_D)^2 + \Gamma^2]^{1/2}}{(\omega - \omega_D)^2 + \Gamma^2} \right)^{1/2}. \end{aligned}$$

The expression (53) has two peaks (see Fig. 3). One is controlled by the singularity of the phonon density of states at the threshold  $\omega = \omega_D$ . Its relative height is  $\Sigma_{op-b}^{\max}/\Sigma_{e-h}^{\max} \simeq \zeta_2 p_F^{1/2} v^{1/2}/\zeta_1^{3/2} s^{1/2}$ . The other, at  $\omega = \omega_{op}(k_s = 0, k_z = \zeta_1)$ , is determined by momentum and energy conservation. Here,  $\Sigma_{op-b}^{\max}/\Sigma_{e-h}^{\max} \simeq p_F^2/\zeta_1\zeta_2$ . The frequency interval between the peaks is  $a\zeta_1^2/2\omega_D$ , whereas their width is  $a\zeta_1\zeta_2/2\omega_D$ . We see the peaks are resolved for the above situation.

If  $\zeta_1 \leq \zeta_2$ , it is easy to see that there is only one peak at the frequency  $\omega_D$  determined by the singularity of the phonon density of states. This result is obvious, and we do not derive it to save space. For the nonperpendicular scattering for which we still have  $\langle Y(\mathbf{p})\Xi_s(\mathbf{p}) \rangle = 0$ , the transverse peaks exist also; the relative height  $\Sigma_{op-t}^{\max}/\Sigma_{op-l}^{\max} \simeq \min(k_s^2/k_z^2, 1)$ . If the vector  $\langle Y(\mathbf{p})\Xi_i(\mathbf{p}) \rangle$  lies in  $s$  direction, the transverse peaks exist only for normal scattering. For inclined scattering, the ratio is  $\Sigma_{op-t}^{\max}/\Sigma_{op-l}^{\max} \simeq \min(k_s^2/k_z^2, 1)$ . For the general case, both the longitudinal and transverse peaks exist with approximately equal heights.

## VI. SURFACE PHONON EXCITATIONS

The first term in the right-hand side of (29) represents the surface phonon contribution. Upon the substitution into (37), we get [Fig. 1(c)],

$$\Sigma_s(\mathbf{k}_s, \omega) = \text{Im} \sum_{\alpha\beta} D_{\alpha z}^s(\mathbf{k}_s, \omega) \lambda_{zz\beta\beta} I_\alpha^*(\mathbf{k}_s, \omega) I_\beta(\mathbf{k}_s, \omega), \quad (54)$$

where for the acoustic phonons

$$I_\alpha^{\text{ac}}(\mathbf{k}_s, \omega) = \sum_{\gamma} \langle Y(\mathbf{p}) \Lambda_{\gamma\gamma}(\mathbf{p}) \rangle \times \int \frac{dk_z}{2\pi} U(\mathbf{k}, \omega) D_{\gamma\alpha}^{\text{(ac-b)}}(\mathbf{k}, \omega) k_\gamma k_\alpha, \quad (55)$$

and for the optical phonons,

$$I_\alpha^{\text{op}}(\mathbf{k}_s, \omega) = \sum_{\gamma} \langle Y(\mathbf{p}) \Xi_{\gamma}(\mathbf{p}) \rangle \times \int \frac{dk_z}{2\pi} U(\mathbf{k}, \omega) D_{\gamma\alpha}^{\text{(op-b)}}(\mathbf{k}, \omega) k_\gamma. \quad (56)$$

The surface matrix Green's functions  $D_{ik}^{\text{(ac-s)}}(\mathbf{k}_s, \omega)$  and  $D_{ik}^{\text{(op-s)}}(\mathbf{k}_s, \omega)$  have poles determining the surface acoustic-phonon (Rayleigh's) spectrum  $\omega = \omega_{\text{ac-s}}(\mathbf{k}_s)$  and the surface optical-phonon spectrum  $\omega = \omega_{\text{op-s}}(\mathbf{k}_s)$ .

### A. Acoustic surface phonons

The scattering cross section (54) is expressed in terms of  $I_\alpha^{\text{ac}}(\mathbf{k}_s, \omega)$  and  $D_{ik}^{\text{(ac-s)}}(\mathbf{k}_s, \omega)$ . We now have the challenging task of finding their explicit form. But it is easy to solve a problem for the important isotropic case, in which the tensor of elastic constants

$$\lambda_{iklm} = \rho s_t^2 (\delta_{il} \delta_{km} + \delta_{im} \delta_{kl}) + \rho (s_l^2 - 2s_t^2) \delta_{ik} \delta_{lm}, \quad (57)$$

where  $s_t$  is the transverse sound velocity. Substituting (57) into the equation defining the bulk Green's matrix (28), we get

$$D_{ik}^{\text{(ac-b)}} = \frac{1}{\rho s_t^2 s_l^2 (k_z^2 + \kappa_l^2) (k_z^2 + \kappa_t^2)} \times \begin{pmatrix} s_l^2 k_z^2 + s_t^2 \kappa_t^2 & -(s_l^2 - s_t^2) k_s k_z \\ -(s_l^2 - s_t^2) k_s k_z & s_t^2 k_z^2 + s_l^2 \kappa_l^2 \end{pmatrix}, \quad (58)$$

here we denote  $\kappa_{l,t}^2 = k_s^2 - \omega^2/s_{l,t}^2$ .

With the help of (32), we find after the intricate calculation

$$D_{\alpha z}^{\text{(ac-s)}} \lambda_{zz\beta\beta} = -\frac{2\omega^2 \kappa_l \rho}{s_t^4 [(k_s^2 + \kappa_t^2)^2 - 4k_s^2 \kappa_t \kappa_l]} \times \begin{pmatrix} (s_l^2 - 2s_t^2)^2 (s_l^2 - 2s_t^2) s_l^2 \\ (s_l^2 - 2s_t^2) s_l^2 & s_l^4 \end{pmatrix}. \quad (59)$$

Note this matrix is symmetric, therefore the cross section (4) is positively defined.

In the range  $|\omega| < s_t k_s$ , both  $\kappa_t$  and  $\kappa_l$  are real. The

transverse and longitudinal waves are coupled together by the boundary conditions [this is shown in Fig. 1(c) by the vertical lines] into Rayleigh's wave. The dispersion of the surface waves<sup>30</sup> is determined by the pole of the matrix  $D_{ik}^{\text{(ac-s)}}(\mathbf{k}_s, \omega)$  shown by the double wave line on Fig. 1(c). The bulk phonon matrix Green's functions, integrated over  $k_z$  in Eqs. (54) and (55), are shown by single wave lines. Note that the integration over  $k_z$  is not relevant to the surface phonon line. The imaginary part in Eq. (59) comes from the damping  $\Gamma_{\text{ac}}$  in the denominator of Eq. (59), where  $\omega \rightarrow \omega + i\Gamma_{\text{ac}}(\mathbf{k}_s)$ . Thus, the scattering cross section has the form of a sharp peak at  $|\omega| = s_R k_s$ , corresponding to Rayleigh's phonons. The exact expression for  $I_i^{\text{ac}}(\mathbf{k}_s, \omega)$  (55) is cumbersome, but it depends almost not at all on  $\zeta$ ,

$$I_i^{\text{ac}}(\mathbf{k}_s, \omega) \simeq \langle Y(\mathbf{p}) \Lambda(\mathbf{p}) \rangle / \rho s^2. \quad (60)$$

Substituting Eqs. (59), (60) into Eq. (54) and extracting the imaginary part, we obtain near the pole

$$\Sigma_{\text{ac-s}}(\mathbf{k}_s, \omega) \simeq \frac{\Gamma_{\text{ac}} |\langle Y(\mathbf{p}) \Lambda(\mathbf{p}) \rangle|^2}{\rho s \{ [|\omega| - \omega_{\text{ac-s}}(\mathbf{k}_s)]^2 + \Gamma_{\text{ac}}^2 \}}. \quad (61)$$

The formula is exact as long as  $\lambda_{iklm}$  has the isotropic form (57). Estimating the expression (61), we get  $\Sigma_{\text{ac-s}}^{\text{max}} / \Sigma_{e-h}^{\text{max}} \simeq s \zeta_2 / \Gamma_{\text{ac}}$ .

In the domain  $s_t k_s < \omega < s_l k_s$ , the imaginary part appears because  $\kappa_t$  is imaginary ( $\kappa_l$  is still real). In other words, transverse phonons propagate into the bulk. The longitudinal phonons are still decay from the surface. The contribution of such quasisurface excitations (they are called sometimes as "mixed" modes) results in the narrow continuum, which at  $\omega \geq s_t k_s$  has the following frequency dependence (see Fig. 2),

$$\Sigma_{\text{ac-s}}(\mathbf{k}_s, \omega) \simeq \frac{|\langle Y(\mathbf{p}) \Lambda(\mathbf{p}) \rangle|^2}{\rho s^2 k_s^2} \left( \frac{\omega^2}{s_t^2} - k_s^2 \right)^{1/2}. \quad (62)$$

In the range  $s_l k_s < \omega$ , both  $\kappa_t$  and  $\kappa_l$  are imaginary: the transverse and longitudinal phonons propagate into the bulk. The longitudinal phonons interact with electrons more efficiently than the transverse ones. The integral (55) has the square root singularity at  $\omega = s_l k_s$ :

$$I_s^{\text{ac}}(\mathbf{k}_s, \omega) \simeq \langle Y(\mathbf{p}) \Lambda(\mathbf{p}) \rangle \frac{i k_s^2}{\rho s_l^2 \zeta} \left( k_s^2 - \frac{\omega^2}{s_l^2} \right)^{-1/2}.$$

Taking into account  $D_{ik}^{\text{(ac-s)}}$  and  $\Gamma_{\text{ac}}$  given by Eqs. (59), (26), we obtain

$$\Sigma_{\text{ac-s}}(\mathbf{k}_s, \omega) \simeq \frac{k_s^{3/2} |\langle Y(\mathbf{p}) \Lambda(\mathbf{p}) \rangle|^2}{\rho s^{3/2} |\zeta|^2} \times \left( \frac{|\omega| - s_l k_s + [ (|\omega| - s_l k_s)^2 + \Gamma_{\text{ac}}^2 ]^{1/2}}{(|\omega| - s_l k_s)^2 + \Gamma_{\text{ac}}^2} \right)^{1/2}. \quad (63)$$

This nonsymmetric peak results from the slipping longitudinal phonons,  $k_z \rightarrow 0$ . The estimate of the ratio is

$\Sigma_{ac-s}^{\max}/\Sigma_{e-h}^{\max} \simeq (v/s)^{1/2} k_s \zeta_2 / |\zeta|^2$ . A similar peak ("longitudinal resonance") has appeared in the theory of ILS in dielectrics.<sup>22</sup>

### B. Optical surface phonons

We consider the optical surface contribution (56) for the most interesting case when the vector  $\langle Y(\mathbf{p})\Xi_i(\mathbf{p}) \rangle$  is in  $z$  direction. The evaluation is similar to the case of acoustic surface phonons. The optical-phonon matrix Green's function  $D_{ik}^{\text{op}-s}(\mathbf{k}_s, \omega)$  has the same form as Eq. (59), with  $\kappa_{i,t}^2 = k_s^2 - (\omega^2 - \omega_D^2)/a_{l,t}$ , where  $a_l = \mu_{zzzz}/\rho'$  and  $a_t = \mu_{szzs}/\rho'$ . The pole of  $D_{ik}^{\text{op}-s}(\mathbf{k}_s, \omega)$  gives the optical surface phonon spectrum  $\omega = \omega_{\text{op}-s}(\mathbf{k}_s)$ , which consist of two branches (on the contrary to the acoustic Rayleigh spectrum) and will be considered in detail elsewhere.<sup>32</sup> The surface optical phonons have been studied in Ref. 31 for an isotropic dielectric.

The optical surface peak has the form:

$$\Sigma_{\text{op}-s}(\mathbf{k}_s, \omega) \simeq \frac{\Gamma_{\text{op}} |\langle Y(\mathbf{p})\Xi_s(\mathbf{p}) \rangle|^2}{\rho' |a|^{1/2} \max(k_s^2, |\zeta|^2) \{[\omega - \omega_{\text{op}-s}(\mathbf{k}_s)]^2 + \Gamma_{\text{op}}^2\}} \quad (64)$$

In the interval  $\omega_D^2 + a_t k_s^2 < \omega^2 < \omega_D^2 + a_l k_s^2$ , there are mixed optical modes which contribute in the cross section the following continuum:

$$\Sigma_{\text{op}-s}(\mathbf{k}_s, \omega) \simeq \frac{|\langle Y(\mathbf{p})\Xi_s(\mathbf{p}) \rangle|^2}{\rho' |a|^{3/2} k_s^2 \max(k_s^2, |\zeta|^2)} (\omega^2 - \omega_D^2 - a_t k_s^2)^{1/2}.$$

The optical longitudinal resonance has the shape,

$$\Sigma_{\text{op}-s}(\mathbf{k}_s, \omega) \simeq \frac{|\langle Y(\mathbf{p})\Xi_s(\mathbf{p}) \rangle|^2}{\rho' k_s^{1/2} |a|^{3/4} |\zeta|^2} \times \left( \frac{\delta\omega \operatorname{sgn} a_l + [\delta\omega^2 + \Gamma_{\text{op}}^2]^{1/2}}{\delta\omega^2 + \Gamma_{\text{op}}^2} \right)^{1/2}, \quad (65)$$

where

$$\delta\omega = \omega - (\omega_D^2 + a_l k_s^2)^{1/2}.$$

This peak shape is asymmetric. For  $a_l > 0$ , the cross section decreases rapidly in the range  $\delta\omega \sim \Gamma_{\text{op}}$ , if  $\delta\omega < 0$ . At the opposite side of the peak, where  $\delta\omega > 0$ , the cross section decreases slowly, as  $(\delta\omega)^{-1/2}$ . One can see that the longitudinal resonance is absent, if the vector  $\langle Y(\mathbf{p})\Xi_i(\mathbf{p}) \rangle$  lies in  $z$  direction.

## VII. SUMMARY AND CONCLUSIONS

In this paper, we evaluate the semiclassical response of a semi-infinite metal to an arbitrary external field  $U(\mathbf{r}, t)$ . The existence of lattice vibrations is taken into account and leads to nontrivial effects. Our method is based on the straightforward solution to the boundary problem.

It is applied to ILS by a normal metal. We have considered the effect of the electron-phonon interaction in the inelastic light scattering. The interaction manifests itself in three different contributions.

First, the electron-phonon interaction modifies [Fig. 1(a)] the scattering by the electron-hole pairs in the same manner as for conductivity. This contribution may be called the relaxation background and is observed in the frequency transfer range  $\omega \sim \tau^{-1} = 2\pi g\omega_D \sim 10^3 \text{ cm}^{-1}$  [see Eqs. (42), (43)].

Second, the light scattering by electrons may involve the excitation or absorption of a bulk phonon [Fig. 1(b)]. The Brillouin-Mandelstam scattering with the excitation of acoustic bulk phonons is attended with the low frequencies transfer  $\omega \sim sk \sim 0.1 \text{ K} \sim 5 \text{ GHz}$ , since the momentum transfer  $k$  of light in the optical range is small. The scattering is observable only when the skin depth is rather large. The Raman scattering with the excitation of the optical bulk phonons is associated with the frequency transfer on the order of the Debye frequency  $10^2 \text{ cm}^{-1}$ . In the case of small phonon damping, the corresponding peaks may be decomposed into two maxima (Fig. 3). One is controlled by the singularity of phonon density of states at the threshold and another is determined by the momentum and energy conservation. The width of bulk acoustic- and optical-phonon resonances [see Eqs. (47) and (48) and (52) and (53)] is determined in competition between  $\Gamma$  and  $s\zeta_2$ , where the phonon damping  $\Gamma$  and the field decrement  $\zeta_2$  are given by Eqs. (26), (34), and (39).

Third, the excitation of surface acoustic and optical phonons is possible [Fig. 1(c)]. The width of surface resonances [see Eqs. (61), (64)] does not exceed the width of bulk resonances, because it is defined by the phonon damping and does not depend on the field decrement. In addition there is a narrow continuum in ILS from a special type of the mixed phonons, which are a superposition of the longitudinal wave with an amplitude diminishing from the boundary into the bulk (similar to the Rayleigh wave) and the nondiminishing transverse wave. This continuum is accompanied by the nonsymmetric longitudinal resonance Eqs. (63), (65).

Let us note that the peak shapes are independent of temperature, in agreement with experimental results.<sup>1-6</sup> But their widths are studied insufficiently in the experiment and the attribution to bulk or surface excitations is undetermined.

The theory of Raman light scattering in polar metals will be published soon in the future. Here, the electric field plays a more important role. There are two ways to affect the scattering. First, the bulk optical phonon dispersion is modified. It has an immediate impact on the surface optical-phonon dispersion. Second, a different type of surface excitations appears—the so called Fuchs-Kliwer surface polaritons.<sup>33</sup> The peaks corresponding to excitation and absorption of polaritons come in the cross section.

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### APPENDIX: RAMAN SCATTERING IN IMPURE METAL WITH COULOMB INTERACTION

Let us show that the above result (37) may be obtained by accounting the electron-electron interactions and the electron-impurity interaction beyond the  $\tau$  approximation. Here, all the surface effects are omitted as well as the electron-phonon coupling for simplicity sake.

We search for the solution of Boltzmann's equation,

$$f_p(\mathbf{k}, \omega) = f_0[\varepsilon(\mathbf{p}, \mathbf{k}, \omega) - \mu_0] + \frac{df_0}{d\varepsilon} \delta f_p(\mathbf{k}, \omega), \quad (\text{A1})$$

depending on the unperturbed chemical potential  $\mu_0$  and the local energy,

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

The linearized Boltzmann's equation with help of (A1) is

$$-i(\omega - \mathbf{v} \cdot \mathbf{k})\delta f_p(\mathbf{k}, \omega) = i\omega\gamma(\mathbf{p})U(\mathbf{k}, \omega) + ie\mathbf{v} \cdot \mathbf{k}\phi(\mathbf{k}, \omega) + \hat{S}t f_p(\mathbf{k}, \omega), \quad (\text{A2})$$

where the electron-impurity collision integral,

$$\begin{aligned} \hat{S}t f_p(\mathbf{k}, \omega) \\ = 2 \int \frac{dS'_F}{(2\pi)^3 v} w(\mathbf{p}, \mathbf{p}') [\delta f_{p'}(\mathbf{k}, \omega) - \delta f_p(\mathbf{k}, \omega)]. \end{aligned} \quad (\text{A3})$$

We have the Poisson equation,

$$\epsilon_{ik} k_i k_k \phi(\mathbf{k}, \omega) = 8\pi e \int \frac{d^3 p}{(2\pi)^3} f_p(\mathbf{k}, \omega), \quad (\text{A4})$$

for the potential  $\phi(\mathbf{k}, \omega)$  of the Coulomb electron interaction, where  $\epsilon_{ik}$  are the dielectric constants of the lattice.

Because of the Debye screening, the electron-impurity interaction in metals can be described by a short-range potential. Hence the scattering amplitude is isotropic  $w(\mathbf{p}, \mathbf{p}') = w_0$ . Then one rewrites Eq. (A2),

$$\begin{aligned} \delta f_p(\mathbf{k}, \omega) = & -\frac{1}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau^{-1}} \\ & \times \left( \omega\gamma(\mathbf{p})U(\mathbf{k}, \omega) + e\mathbf{v} \cdot \mathbf{k}\phi(\mathbf{k}, \omega) \right. \\ & \left. - i\tau^{-1} \frac{\langle \delta f_p(\mathbf{k}, \omega) \rangle}{\langle 1 \rangle} \right), \end{aligned} \quad (\text{A5})$$

where the scattering rate is  $\tau^{-1} = \langle w_0 \rangle$ . Integrating both sides of Eq. (A5) over the Fermi surface, we find

$$\begin{aligned} \frac{\langle \delta f_p(\mathbf{k}, \omega) \rangle}{\langle 1 \rangle} = & - \left\langle \left\langle \frac{\omega\gamma(\mathbf{p})}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau^{-1}} \right\rangle U(\mathbf{k}, \omega) \right. \\ & \left. + \left\langle \frac{\mathbf{v} \cdot \mathbf{k}}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau^{-1}} \right\rangle e\phi(\mathbf{k}, \omega) \right) \\ & \times \left( \left\langle \left\langle \frac{\omega - \mathbf{v} \cdot \mathbf{k}}{\omega - \mathbf{v} \cdot \mathbf{k} + i\tau^{-1}} \right\rangle \right\rangle^{-1} \right). \end{aligned} \quad (\text{A6})$$

Let us consider two limiting cases.

(a) Small momentum transfer,  $|\omega + i\tau^{-1}| \gg vk$ . Then we omit all the terms proportional to  $\mathbf{v} \cdot \mathbf{k}$ . In this lowest order, we lose the diffuson pole in Eq. (A6) (see Ref. 16). Equation (A6) gives  $\langle \delta f_p(\mathbf{k}, \omega) \rangle = -\langle \gamma(\mathbf{p}) \rangle U(\mathbf{k}, \omega)$ , and we obtain

$$\delta f_p(\mathbf{k}, \omega) = -\frac{U(\mathbf{k}, \omega)}{\omega + i\tau^{-1}} \left( \omega\gamma(\mathbf{p}) + i\tau^{-1} \frac{\langle \gamma(\mathbf{p}) \rangle}{\langle 1 \rangle} \right). \quad (\text{A7})$$

Substituting (A7) into (10), we get our result (37) for the small momentum transfer limit.

(b) Large momentum transfer,  $|\omega + i\tau^{-1}| \ll vk$ . In this case the last term in (A5) is negligible, and  $\tau^{-1}$  determines only the bypass round the pole. The solution of Poisson's equation after the substitution (A1), (A5) into (A4) gives

$$\phi(\mathbf{k}, \omega) = \frac{4\pi e U(\mathbf{k}, \omega)}{D(\mathbf{k}, \omega)} \left\langle \frac{\mathbf{v} \cdot \mathbf{k} \gamma(\mathbf{p})}{\omega - \mathbf{v} \cdot \mathbf{k} + i0} \right\rangle, \quad (\text{A8})$$

where

$$D(\mathbf{k}, \omega) = \epsilon_{ik} k_i k_k - 4\pi e^2 \left\langle \frac{\mathbf{v} \cdot \mathbf{k}}{\omega - \mathbf{v} \cdot \mathbf{k} + i0} \right\rangle. \quad (\text{A9})$$

Using Eqs. (A1), (A5), and (A8) we get the response (10):

$$\begin{aligned} \frac{\delta n_\gamma(\mathbf{k}, \omega)}{U(\mathbf{k}, \omega)} = & \left\langle \frac{\mathbf{v} \cdot \mathbf{k} |\gamma(\mathbf{p})|^2}{\omega - \mathbf{v} \cdot \mathbf{k} + i0} \right\rangle \\ & + \left\langle \frac{\mathbf{v} \cdot \mathbf{k} \gamma^*(\mathbf{p})}{\omega - \mathbf{v} \cdot \mathbf{k} + i0} \right\rangle \left\langle \frac{\mathbf{v} \cdot \mathbf{k} \gamma(\mathbf{p})}{\omega - \mathbf{v} \cdot \mathbf{k} + i0} \right\rangle \\ & \times \left( \left\langle \left\langle \frac{\mathbf{v} \cdot \mathbf{k}}{\omega - \mathbf{v} \cdot \mathbf{k} + i0} \right\rangle \right\rangle^{-1} \right). \end{aligned} \quad (\text{A10})$$

Expanding in powers of  $\omega/vk$ ,

$$\left\langle \frac{\mathbf{v} \cdot \mathbf{k} \gamma(\mathbf{p})}{\omega - \mathbf{v} \cdot \mathbf{k} + i0} \right\rangle = -\langle \gamma(\mathbf{p}) \rangle - \frac{i\pi\omega}{k} \left\langle \gamma(\mathbf{p}) \frac{\delta(\nu)}{v} \right\rangle, \quad (\text{A11})$$

one can rewrite the expression (A10):

$$\frac{\delta n_\gamma(\mathbf{k}, \omega)}{U(\mathbf{k}, \omega)} = -\langle |Y(\mathbf{p})|^2 \rangle - \frac{i\pi\omega}{k} \left\langle |Y(\mathbf{p})|^2 \frac{\delta(\nu)}{v} \right\rangle, \quad (\text{A12})$$

where the vertex  $\gamma(\mathbf{p})$  is renormalized according to Eqs. (17). Hence we obtain Eq. (40).

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