

Effect of the electron-phonon interaction on the shift and attenuation of optical phonons

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(Received 5 March 2002; published 2 July 2002)

Using the Boltzmann equation for electrons in metals, we show that the optical phonons soften and have a dispersion due to screening in agreement with the results reported recently [M. Reizer, Phys. Rev. B **61**, 40 (2000)]. Additional phonon damping and frequency shift arise when the electron-phonon interaction is properly included.

DOI: 10.1103/PhysRevB.66.020302

PACS number(s): 63.20.Dj, 63.20.Kr, 78.30.-j

Despite attracting considerable interest for half a century since the pioneering work by Fröhlich, the problem of electron-phonon interaction is still far from being solved. Migdal¹ developed a consistent many-body approach based on the Fröhlich Hamiltonian for the interaction of electrons with acoustic (sound) phonons. As Migdal showed (the Migdal theorem), the vertex corrections for acoustic phonons are small by the *adiabatic* parameter $\sqrt{m/M}$, where m and M are the electron and ion masses, respectively. The theory described correctly the electronic lifetime, renormalization of the Fermi velocity v_F , and acoustic phonon attenuation, but resulted in a strong renormalization of the sound velocity $\tilde{s} = s(1 - 2\lambda)^{1/2}$, where λ is the dimensionless coupling constant. For sufficiently strong electron-phonon coupling $\lambda \rightarrow 1/2$, the phonon frequency approached zero marking an instability point of the system. Instead, one would intuitively expect the phonon renormalization to be weak along with the adiabatic parameter.

This discrepancy was resolved by Brovman and Kagan² almost a decade later (see also Ref. 3). They demonstrated the shortcomings of the Fröhlich model that gave an anomalously large phonon renormalization. Employing the Born-Oppenheimer (adiabatic) approximation (see, e.g., Ref. 4), they found that there are two terms in the second-order perturbation theory, which compensate each other making a result small by the adiabatic parameter. Namely, when calculating the phonon self-energy function $\Pi(\omega, k)$ with help of the diagram technique, one should eliminate an adiabatic contribution of the Fröhlich model by subtracting $\Pi(\omega, k) - \Pi(0, k)$.

The interaction of electrons with optical phonons was first considered by Engelsberg and Schrieffer⁵ within Migdal's many-body approach for dispersionless phonons. They predicted a splitting of the optical phonon at finite wave numbers k into two branches. Ipatova and Subashiev⁶ calculated later on the optical phonon attenuation in the collisionless limit and pointed out that the Brovman-Kagan renormalization should be carried out for optical phonons in order to obtain correct phonon renormalization. In Ref. 7, Alexandrov and Schrieffer corrected the calculational error of Ref. 5 and argued that no splitting was found in fact. Instead, they predicted an extremely strong dispersion of optical phonons, $\omega_k = \omega_0 + \lambda v_F^2 k^2 / 3\omega_0$, due to the coupling to electrons. The large phonon dispersion is a typical result of Migdal's theory⁸ using the Fröhlich Hamiltonian. No such dispersion

has ever been observed experimentally. The usual dispersion of optical phonons in metals has the order of the sound velocity. In a recent paper, Reizer⁹ stressed the importance of screening effects which should be taken into account. References 7 and 9 are limited to the cases of both collisionless electron and phonon systems. Moreover, only the phonon renormalization was considered with no results available for the attenuation of optical phonons.

An approach different from many-body technique semiclassical approach based on the Boltzmann equation and the equations of the theory of elasticity, was developed in the papers by Akhiezer, Silin, Gurevich, Kontorovich, and many others (see Ref. 10). This approach was compared with various experiments, such as attenuation of sound waves, effects of strong magnetic fields, crystal anisotropy, sample surfaces on the sound attenuation, and so on. It can be applied to the problem of the electron-optical-phonon interaction¹¹ as well. In the present paper we develop a theory for both the attenuation and frequency shifts of optical phonons with account for effects of the Coulomb screening as well as collisions in the electron and phonon systems.

It is instructive to recapitulate the results of the dynamical theory of elasticity for the renormalization of the sound velocity $\delta s = \tilde{s} - s$ and acoustic attenuation Γ in metals. For a phonon with a wave number k and frequency $\omega_k = sk$, they are given by¹⁰

$$\frac{\delta s}{s} - i \frac{\Gamma}{\omega_k} = \lambda \begin{cases} \frac{s^2}{v_F^2} - i \frac{\pi s}{2v_F} & \text{for } kv_F > |\omega_k + i\gamma| \\ \frac{\omega_k}{\omega_k + i\gamma} & \text{for } kv_F < |\omega_k + i\gamma|, \end{cases} \quad (1)$$

where γ is the electronic scattering rate, and the dimensionless coupling constant λ is proportional to the electronic density of states ν_0 at the Fermi surface (for the isotropic case $\nu_0 = m^* p_F / \pi^2$, m^* is the effective electron mass) and to the squared deformation potential ζ_{ik} . The deformation potential describes the change in the spectrum of electrons subject to lattice deformation $\varepsilon(\mathbf{p}, \mathbf{r}, t) = \varepsilon_0(\mathbf{p}) + \zeta_{ik}(\mathbf{p}) u_{ik}(\mathbf{r}, t)$, where u_{ik} is the strain tensor. Equations (1) give the correct answers in various known regimes: for the sound attenuation in the hydrodynamic limit ($\omega_k \ll \gamma$ and $k \rightarrow 0$), for the zero sound ($\omega_k \gg \gamma$ and $k \rightarrow 0$), for the Landau damping in the ballistic limit ($kv_F \gg |\omega_k + i\gamma|$). In the latter case, both the sound velocity shift and attenuation are small by the adiabatic param-

eter s/v_F contrary to the results of the Fröhlich model. Note also that Eqs. (1) show hardening of the phonon frequency due to the electron-phonon interaction in contradiction with Migdal's result.

For the case of optical phonons, two known types of the electron-phonon interaction are the deformation potential and the interaction with the electrical polarization induced by optical vibrations. We consider here for simplicity a cubic crystal with two different atoms in a unit cell. Then, there are three optical modes, and the interaction with the induced polarization has the Fröhlich form

$$\varepsilon(\mathbf{p}, \mathbf{r}, t) = \varepsilon_0(\mathbf{p}) + \zeta(\mathbf{p}) \nabla \cdot \mathbf{u}(\mathbf{r}, t), \quad (2)$$

where the scalar function $\zeta(\mathbf{p})$ of the electron momentum is the coupling with the optical displacements \mathbf{u} . In order to compare our results with previous ones, here we consider the interaction in the same form (2) as in Refs. 7–9. One can see that the principal characteristic features of the phenomenon are retained for the deformation interaction, $\delta\varepsilon = \zeta_i(\mathbf{p})u_i(\mathbf{r}, t)$, where the coupling is a vector function. The distinction is that only the longitudinal mode interacts with electrons in the case of induced polarization (2), and the interaction approaches zero in the long-wave limit. Therefore, we concentrate on the propagation of the longitudinal mode along the symmetry axis when this mode is not mixed with transverse ones. Note also that the electric field plays an important role especially when the different atoms are in the unit cell so that the dipole moment is excited under the atom vibrations.

The main point of the theory is the equation of motion in the long-wave approximation ($k \ll 1/a$) for the Fourier components of the optical-phonon displacement u_j ,

$$(\omega_k^2 - \omega^2)u_j(\mathbf{k}, \omega) = \frac{Z}{M'}E_j + \frac{ik_j}{M'N} \int \frac{2d^3p}{(2\pi)^3} \zeta(\mathbf{p}) \delta f_p(\mathbf{k}, \omega), \quad (3)$$

where E_j is the electric field associated with vibrations, N is the number of unit cells in 1 cm^3 , M' is the reduced mass of two atoms in the unit cell, and Z is the effective ionic charge. The nonperturbed phonon frequency ω_k should be considered in the absence of the electric field and without any nonadiabatic corrections. In the long-wave limit, we can roughly describe it as $\omega_k^2 = \omega_0^2 \pm s^2 k^2$ with the magnitude of s being of the order of the typical sound velocity in metals. We should mention that the optical phonons always have the so-called natural width $\Gamma^{\text{nat}} \sim \omega_0 \sqrt{m/M}$. The natural width results from decay processes into two (or more) acoustic or optical phonons, which are possible even at zero temperature. We can add this width to ω_k in the form $-i\Gamma^{\text{nat}}/2$.

The last term in Eq. (3) presents the driving force from the nonadiabatic electron system due to the deviation $\delta f_p(\mathbf{r}, t)$ from the local-equilibrium distribution function $f_0[\varepsilon(\mathbf{p}, \mathbf{r}, t) - \mu]$.

Then, we have the Boltzmann equation

$$\begin{aligned} & -i(\omega - \mathbf{k} \cdot \mathbf{v}) \delta f_p(\mathbf{k}, \omega) + \gamma[\delta f_p(\mathbf{k}, \omega) - \langle \delta f_p(\mathbf{k}, \omega) \rangle] \\ & = -[\omega \zeta(\mathbf{p}) \mathbf{k} \cdot \mathbf{u}(\mathbf{k}, \omega) + e \mathbf{v} \cdot \mathbf{E}] \frac{df_0}{d\varepsilon} \end{aligned} \quad (4)$$

in the approximation of relaxation rate γ , which holds at low temperatures when the electron-impurity interaction dominates as well as at temperatures higher than the Debye temperature when the phonon-phonon collisions can be considered as elastic. The term in the angular brackets in Eq. (4), which denote the average over the Fermi surface,

$$\langle \dots \rangle = \frac{1}{\nu_0} \int (\dots) \frac{2dS_F}{v(2\pi)^3},$$

arises from the out term in the collision integral. Notice, that the condition $\langle \zeta(\mathbf{p}) \rangle = 0$ should be fulfilled, because the number of electrons in the local-equilibrium state $f_0[\varepsilon(\mathbf{p}, \mathbf{r}, t) - \mu]$ is conserved. The validity conditions of Boltzmann's equation (4) are $k \ll p_F$ and $\omega \ll \varepsilon_F$. Let us emphasize that Eqs. (3) and (4) are nothing but the equations for the phonon and electron self-energy, respectively, of the diagram technique.

With the help of the Maxwell equations, the electric field is expressed in the terms of polarization \mathbf{P} as follows:

$$\mathbf{E} = -4\pi \mathbf{k}(\mathbf{k} \cdot \mathbf{P})/k^2, \quad (5)$$

provided that phonons are excited in the optical region $k \gg \omega/c$, where the wave vector is determined by the incident light $k \sim \omega^{(i)}/c$ and the frequency ω is of the order of the optical phonon frequency ω_0 . It is seen that the electric field is longitudinal and only the longitudinal component of polarization P_z (\mathbf{k} is taken along the z axis) plays a role being related to the phonon displacement and the electric field by the equation

$$P_z = NZu_z + \alpha E + \frac{ie}{k} \int \frac{2d^3p}{(2\pi)^3} \delta f_p(\mathbf{k}, \omega), \quad (6)$$

where the first term is caused by ionic motion, α is the polarizability of filled bands, and the last term is the carrier contribution defined by the variation of the electron density, $\rho^{(e)} = -i\mathbf{k} \cdot \mathbf{P}^{(e)}$.

Equations (3)–(6) give the complete system of our problem. The Boltzmann equation (4) has the solution in the form

$$\delta f_p(\mathbf{k}, \omega) = -\chi_p(\mathbf{k}, \omega) \frac{df_0}{d\varepsilon},$$

where

$$\chi_p(\mathbf{k}, \omega) = i[e \mathbf{v} \cdot \mathbf{E} + \omega \zeta(\mathbf{p}) \mathbf{k} \cdot \mathbf{u} + \gamma \langle \chi_p(\mathbf{k}, \omega) \rangle] / \Delta, \quad (7)$$

$$\langle \chi_p(\mathbf{k}, \omega) \rangle = i \langle [e \mathbf{v} \cdot \mathbf{E} + \omega \zeta(\mathbf{p}) \mathbf{k} \cdot \mathbf{u}] / \Delta \rangle / (1 - i \langle \gamma / \Delta \rangle),$$

and we set $\Delta = \omega - \mathbf{k} \cdot \mathbf{v} + i\gamma$.

Using this solution, we obtain the polarization (6) and rewrite the electric field (5) in terms of the longitudinal displacement u_z ,

$$\varepsilon_e(k, \omega)E = -4\pi\beta_{fd}u_z, \quad (8)$$

where we introduce the field-displacement response function

$$\beta_{fd} = NZ - e\omega\nu_0 \frac{\langle \zeta(\mathbf{p})/\Delta \rangle}{1 - i\langle \gamma/\Delta \rangle}. \quad (9)$$

The electron contribution into the dielectric function has the known form

$$\varepsilon_e(k, \omega) - \varepsilon_\infty = -\frac{4\pi e^2 \nu_0 \langle v_z/\Delta \rangle}{k(1 - i\langle \gamma/\Delta \rangle)}, \quad (10)$$

where the high-frequency permittivity $\varepsilon_\infty = 1 + 4\pi\alpha$.

Now, we consider the equation of motion (3) using the solution of the Boltzmann equation (7). The term proportional to u_z of the driving force can be included in the phonon frequency,

$$\tilde{\omega}^2 = \omega_k^2 + \frac{\nu_0 \omega k^2}{M'N} \left(\left\langle \frac{\zeta^2(\mathbf{p})}{\Delta} \right\rangle + \frac{i\gamma \langle \zeta(\mathbf{p})/\Delta \rangle^2}{1 - i\langle \gamma/\Delta \rangle} \right), \quad (11)$$

so that Eq. (3) reads

$$(\tilde{\omega}^2 - \omega^2)u_z = \tilde{Z}E/M', \quad (12)$$

where the renormalized ionic charge

$$\tilde{Z} = Z - \frac{e\nu_0 k}{N} \left(\left\langle \frac{\zeta(\mathbf{p})v_z}{\Delta} \right\rangle + i\gamma \frac{\langle v_z/\Delta \rangle \langle \zeta(\mathbf{p})/\Delta \rangle}{1 - i\langle \gamma/\Delta \rangle} \right).$$

Using the condition $\langle \zeta(\mathbf{p}) \rangle = 0$ we obtain $\tilde{Z} = \beta_{fd}/N$.

Then, we can express the displacement u_z from Eq. (12) in terms of E and, substituting this value into Eq. (8), obtain the dielectric function of the electron-ion system,

$$\varepsilon(k, \omega) = \varepsilon^{(e)}(k, \omega) + 4\pi N \tilde{Z}^2 / M' (\tilde{\omega}^2 - \omega^2). \quad (13)$$

The frequency of the longitudinal mode is defined by the condition $\varepsilon(k, \omega) = 0$, i.e.,

$$\omega^2 = \tilde{\omega}^2 + 4\pi N \tilde{Z}^2 / M' \varepsilon_e(k, \omega). \quad (14)$$

In the absence of free electrons, the density of states $\nu_0 = 0$ and Eq. (14) gives for the LO mode $\omega_{LO}^2 = \omega_k^2 + \omega_{pi}^2$, where $\omega_{pi}^2 = 4\pi NZ^2 / M' \varepsilon_\infty$ is the squared ion-plasma frequency of the order of ω_0^2 . For the TO mode, when the electric field $E = 0$, we obtain $\omega_{TO}^2 = \omega_k^2$.

Free electrons in metals make the large contribution into the dielectric function [see, Eq. (10)]. Expanding in powers of k we have in the zero order

$$\varepsilon^{(e)}(k, \omega) - \varepsilon_\infty = -\varepsilon_\infty \omega_{pe}^2 / \omega(\omega + i\gamma), \quad (15)$$

which corresponds simply to the Drude conductivity with the electron-plasma frequency

$$\omega_{pe}^2 = \frac{e^2}{3\pi^2 \varepsilon_\infty} \int v dS_F.$$

For large $kv_F > |\omega + i\gamma|$, the electron contribution (10) describes the Debye screening,

$$\varepsilon^{(e)}(k, \omega) - \varepsilon_\infty = \varepsilon_\infty \frac{k_0^2}{k^2} (1 + i\pi\omega/2kv_F),$$

where a term of the order of ω/kv is kept and the Debye parameter $k_0^2 = 4\pi e^2 \nu_0 / \varepsilon_\infty$.

Therefore, we can solve Eq. (14) for $\omega \ll \omega_{pe}$, using the iteration procedure to a first approximation. The solution is complex valued, and its real and imaginary parts give the LO phonon frequency and width

$$\omega_{LO}^2 - i\omega_{LO}\Gamma = \omega_k^2 + \frac{k^2 \omega_k \nu_0}{M'N} \left\langle \frac{\zeta^2(\mathbf{p})}{\Delta} \right\rangle + \frac{\omega_{pi}^2 \varepsilon_\infty}{\varepsilon^{(e)}(k, \omega_k)}. \quad (16)$$

In the case of small $kv_F < |\omega_k + i\gamma|$, expanding in k powers, we obtain

$$\omega_{LO}^2 - i\omega_{LO}\Gamma = \omega_k^2 - \omega_k(\omega_k + i\gamma) \frac{\omega_{pi}^2}{\omega_{pe}^2} + \frac{\lambda \omega_k s^2}{\omega_k + i\gamma} k^2, \quad (17)$$

where the dimensionless coupling constant $\lambda = \langle \zeta^2(\mathbf{p}) \rangle \nu_0 / \rho s^2$ contains the factor $a p_{FM}^* / m$ and the metal density ρ .

In the case of large k , expanding in $|\omega_k + i\gamma|/kv_F$, we obtain

$$\omega_{LO}^2 - i\omega_{LO}\Gamma = \omega_k^2 - i \frac{\pi \omega_k s^2 k}{2v_F} \left(\lambda + \frac{\omega_{pi}^2}{s^2 k_0^2} \right) + \frac{\omega_{pi}^2}{k_0^2} k^2, \quad (18)$$

where the coupling constant λ is defined, when the asymptotic value of the integral is calculated: $\nu_0 \langle \zeta^2(\mathbf{p})/\Delta \rangle / M'N = -i\pi s^2 \lambda / 2kv_F$. Note, that the value of λ in Eq. (18), as well as in Eq. (17), vanishes for the case of isotropy due to the condition $\langle \zeta(\mathbf{p}) \rangle = 0$.

Hence, the squared frequency of the longitudinal optical mode is essentially less (by the factor ω_{pi}^2) than for insulators, due to screening of the electric field by free electrons. The additional phonon softening, width, and dispersion in Eq. (17) involve the adiabatic parameter $(\omega_0/\omega_{pe})^2 \sim m/M$, and they are small compared with $\Gamma^{\text{nat}} \sim \omega_0 \sqrt{m/M}$. In the region where Eq. (18) is valid, we see the additional k -dependent width (terms in the parentheses), which is comparable to Γ^{nat} . Here, the λ term conditioned by the electron-phonon interaction is similar to the damping of the acoustic phonons [see the first formula in Eqs. (1)]. Now we omit the shift containing the small factor $(s/v_F)^2$. The second term in the parentheses as well as the last term in Eq. (18), giving the k dispersion, are induced by screening and λ are independent. Since $\omega_{pi}^2/k_0^2 \approx s^2$, this dispersion has a typical value for the phonon branches.

Let us rewrite in our notations the respective results of Ref. 9, Eqs. (9) and (7), retaining only main terms:

$$\omega_{LO}^2 = \omega_k^2 - \frac{\omega_{pi}^2 (\omega_{pi}^2 + \omega_k^2)}{\omega_{pe}^2 (1 - 2\lambda \ln 2)}, \quad kv_F < \omega_k, \quad (19)$$

$$\omega_{\text{LO}}^2 = \omega_k^2 + \frac{\omega_{pi}^2}{3\omega_{pe}^2} (kv_F)^2, \quad kv_F > \omega_k. \quad (20)$$

Comparing with Eqs. (17) and (18) we see that the k dispersion coincides practically. Next, we agree that the contribution ω_{pi}^2 vanishes from the frequency of the LO mode. The softening and damping due to both the electron-phonon scattering (γ) and the phonon decay processes (Γ^{nat}) were ignored in Ref. 9. Concerning the electron-phonon interaction λ , the reason of disagreement was discussed in the beginning of the paper: this is a shortcoming of the diagram technique based on the Fröhlich model. But the most essential difference is the k -dependent width in Eq. (18), which is missed in Eq. (20).

In conclusion, let us make several remarks. The case of the large k -values (18) is most interesting because the electron-phonon and electrodynamic contributions into the phonon width (first and second terms in the parentheses, re-

spectively) can compete. The result depends on the Debye screening and the wave vector k . In Raman experiments, the parameter $kv_F/|\omega_0 + i\gamma| \sim \omega^{(i)}v_F/c\omega_0 \approx 0.3$ if $\omega^{(i)} \approx 10^4$ K and $\omega_0 \approx 10^2$ K, and for metals $v_F \approx 10^8$ cm/s. Therefore, the high incident-light frequency or neutron experiments are desirable. It is more simply to observe the electrodynamic effect in semiconductors, where k_0 is smaller. We have an example of such experiments in works in Ref. 12, where the metal-insulator transition was observed in the GaN crystal under pressure. For the conducting phase, the longitudinal mode softens and obtains the additional damping in comparison with the insulator state. Then, using Eq. (17), we calculate¹³ the collision rate γ which is consistent with the value obtained from the conductivity.

The author thanks E. G. Mishchenko for fruitful discussions. The work was partially supported by the RFBR (Project No. 01-02-16211).

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