Theory of the excitation and amplification of longitudinal-optical phonons in degenerate semiconductors under an intense laser field

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The theory of the excitation and amplification of longitudinal-optical lattice vibrations in a degenerate semiconductor in the presence of an electromagnetic wave is presented. It is shown that in the intense-field limit only multiphoton absorption processes are significant and as a result the optical-phonon population grows with time. A numerical estimate of the actual growth rate is given for a typical semiconductor like GaAs.

I. INTRODUCTION

It is well known that the electron-phonon interaction leads to the renormalization of the phonon spectrum in semiconductors and metals.[1-3] At the same time, the electron-phonon interaction may constitute a fundamental mechanism in the damping of phonons. It is therefore natural to expect that external fields that change the spectrum and occupation number of the electron states will influence the spectrum and damping of phonons. For instance, one knows that under the influence of a fairly strong dc electric field the optical-phonon damping in semiconductors may change considerably in magnitude and even in sign. The modification of the optical-phonon damping and spectrum in constant electric field was considered in Refs. 4 and 5. On the other hand, in the presence of laser radiation it is known that the interaction of such strong fields with a semiconductor not only causes the excitation of higher harmonics and amplification of phonons (acoustic[6-8] and optical[8-10]), but also changes the conductivity in these systems. In particular, the modification of the optical-phonon damping (amplification) in degenerate semiconductors in the presence of a photon field and additional presence of a strong magnetic field was considered in Ref. 10. It was pointed out in Ref. 10 that the optical-phonon amplification may in part be attributed to the single-photon absorption by free carriers accompanied by a net emission of optical phonons. If, however, the laser field is strong enough, from a quantum mechanical viewpoint, the absorption and emission processes by free carriers are of multiphoton nature.

It is the aim of this paper to look at the problem of how the free-carrier absorption of multiphoton radiation will affect the optical-phonon damping in a degenerate semiconductor. The quantum counterpart of this problem is that dealt in Ref. 10 in the absence of the strong dc magnetic field. The main difference between the present theory and the one developed in Ref. 10 is twofold. First, in the present paper we treat the electromagnetic field classically within the dipole approximation. Secondly, we will not make use of perturbation theory in the coupling of the interaction between the electrons and the laser field. It therefore follows that the present treatment is more adequate for studying the system in the strong radiation field regime.

We have accordingly considered optical phonons of frequency $\omega_{\mathbf{k}}$ interacting with free carriers in the presence of an intense laser field with frequency satisfying the inequalities

$$c/v_F \gg \omega\tau \gg 1 \quad \text{and} \quad \omega > \omega_p \ , \tag{1}$$

where τ is the electron relaxation time, ω_p is the plasma frequency, v_F is the electron Fermi velocity, and c is the speed of light. As for the optical phonons we shall confine ourselves to those for which the wave number k satisfies the condition

$$kl \gg 1 \ , \tag{2}$$

where l is the electron mean free path; such phonons constitute well-defined elementary excitations of the system. Fulfillment of conditions (1) and (2) first ensures that the electromagnetic wave penetrates well into the sample and second means that the length of this wave is far greater than the electron mean free path and wavelength of the optical-lattice vibrations. Thus the spatial dependence of the field can be neglected and we need allow only for the dependence of $\mathbf{A}$ on time ($\mathbf{A}$ is the vector potential of the laser wave). The results so obtained are also applicable in the case of $\omega < \omega_p$, provided the length of the sample is much smaller than the penetration depth of the field into the sample, namely $\delta = c(\omega_p^2 - \omega^2)^{-1/2}$.

The phonon scattering by electrons is treated using first-order perturbation theory, retaining, however, the radiation field strength to all orders. The interaction with the laser is treated exactly (i.e., an arbitrary number of laser quanta may be involved). The laser field is, however, described as a classical plane wave. The electron states are described by the solution to the Schrödinger equation for an electron in the field of a classical electromagnetic wave. The transition probabilities are then used to write a kinetic equation for the phonon population from which the damping rate is obtained. In the strong-field limit the electron-phonon collision involving multiphoton absorption becomes dominant, which in turn entails that the optical-phonon population may become unstable.

The paper is organized as follows. In Sec. II the formalism is introduced. In Sec. III the optical-phonon growth rate is calculated and in Sec. IV we given our results and conclusions.

II. FORMALISM

We assume a plane electromagnetic wave propagating along the z direction. The spatial dependence of the laser wave is neglected (dipole approximation). The time-dependent Schrödinger equation for a free carrier (electron or hole) in a semiconductor in the field of the laser light

$$i\hbar\frac{\partial\psi}{\partial t}(\mathbf{r},t)=\frac{1}{2m^*}\left|\frac{\hbar}{i}\nabla-\frac{e}{c}\mathbf{A}(t)\right|^2\psi(\mathbf{r},t)\ , \quad (3)$$

where

$$\mathbf{A}(t)=A_0\hat{\mathbf{e}}_x\cos(\omega t)\ , \quad (4)$$

has the solution[11] (normalized in a box of volume Ω)

$$\psi(\mathbf{r},t)=\frac{1}{\sqrt{\Omega}}\exp\left[i\mathbf{p}\cdot\mathbf{r}-\frac{i}{2m^*\hbar}\times\int^t dt'\,|\hbar\mathbf{p}-\frac{e}{c}\mathbf{A}(t')\,|^2dt'\right]\ . \quad (5)$$

Here $\mathbf{p}$ is the electron wave vector such that in the absence of the radiation field the electron energy E_p is $E_p=\hbar^2p^2/2m^*$. In (5), m^* is the effective mass of an electron.

Treating the electron-phonon interaction as the perturbation, the probability amplitude for the transition from state $i=\psi_{\mathbf{p}}(\mathbf{r},t)$ to stage $f=\psi_{\mathbf{p}'}(\mathbf{r},t)$ due to a collision with an optical phonon $\hbar\mathbf{k}$ is

$$A(i\rightarrow f)=-\frac{i}{\hbar}\int\psi^*_{\mathbf{p}'}(\mathbf{r},t)V_{\mathbf{k}}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_k t)}\psi_{\mathbf{p}}(\mathbf{r},t)d\mathbf{r}\,dt \quad (6)$$

where

$$V_k=i\frac{e}{k}\left[\frac{2\pi\hbar}{\Omega}\right]^{1/2}\omega_k^{1/2}(\epsilon_\infty^{-1}-\epsilon_0^{-1})$$

is the Fourier component of the electron—optical-phonon interaction.[10] Substituting Eq. (5) into (6) and performing the spatial integration we obtain

$$A(i\rightarrow f)=-i\frac{V_k}{\hbar}\frac{(2\pi)^3}{\Omega}\delta(p'-p-k)\int_{-\tau/2}^{\tau/2}dt\,\exp(i/\hbar)(E_{p'}-E_p-\hbar\omega_k)\exp[-i(\lambda/\hbar\omega)\sin(\omega t)]\ , \quad (7)$$

where $\lambda=e\hbar\mathbf{k}\cdot\mathbf{E}_0/m^*\omega$ is the field parameter. Using a Bessel function expansion for

$$\exp[-i(\lambda/\hbar\omega)\sin(\omega t)]\ ,$$

the time integration in Eq. (7) can be performed to yield

$$A(i\rightarrow f)=-2\pi iV_k\frac{(2\pi)^3}{\Omega}\delta(p'-p-\mathbf{k})\times\sum_{s=-\infty}^{+\infty}J_s(\lambda/\hbar\omega)\delta(E_{p'}-E_p-\hbar\omega_k-s\hbar\omega)\ . \quad (8)$$

Here $J_s(\lambda/\hbar\omega)$ is the Bessel function of order s. The scattering amplitude is now specialized to describe the absorption or emission of s photons.

From the well-known relation between the scattering amplitude and the T matrix,[12,13] we can then use Eq. (8) to obtain the transition probability per unit time $T_s(i\rightarrow f)$ for the electronic transition from state $i=\psi_{\mathbf{p}}$ to state $f=\psi_{\mathbf{p}+\mathbf{k}}$, due to a collision with an optical phonon $\mathbf{k}$ with absorption ($s>0$) or emission ($s<0$) of $|s|$ photons from the laser field. One then obtains

$$T_s(i\rightarrow f)=\frac{2\pi}{\hbar}|V_k|^2J_s^2\left[\frac{\lambda}{\hbar\omega}\right]\times\delta(E_{p+k}-E_p-\hbar\omega_k-s\hbar\omega)\ . \quad (9)$$

Since we are treating the laser field classically, the number of photons $|s|$ has the same meaning as in the theory of bounded systems under intense laser fields[13,14]—it is the number of $\hbar\omega$ such that $\Delta E=E_{p'}-E_p-\hbar\omega_k=s\hbar\omega$.

Finally, the rate of change of the number of optical phonons of wave numbers k, dN_k/dt, is then given in terms of the transition probability T_s as[10]

$$\frac{dN_k}{dt}=\gamma_kN_k\ , \quad (10)$$

where

$$\gamma_k=\frac{2\pi}{\hbar}|V_k|^2\sum_{s=-\infty}^{+\infty}\sum_{\mathbf{p}}J_s^2\left[\frac{\lambda}{\hbar\omega}\right](f_{p+k}-f_p)\times\delta(E_{p+k}-E_p-\hbar\omega_k-s\hbar\omega)\ . \quad (11)$$

In Eq. (11), f_p is the Fermi distribution function. If $\gamma_k>0$ the optical-phonon population grows with time, whereas for $\gamma_k<0$ we have damping. Actually the criterion for the onset of the instability is somewhat more involved. In our discussion so far we have completely neglected other mechanisms such as multiphonon processes which may interact with the phonons and lead to a finite phonon lifetime even in the absence of the electron-phonon interaction. We may take them into account by introducing a phenomenological phonon decay rate $\nu(k)$ due to processes other than phonon emission and absorption by electrons. The actual criterion for the optical-phonon instability for a particular wave vector k is therefore

$$\gamma(k)-\nu(k)>0 . \tag{12}$$

III. GROWTH RATE

In this section we shall calculate $\gamma(k)$ in the limit of strong radiation field (the low-field regime is essentially that of Refs. 9 and 10) and show that under certain circumstances $\gamma(k)$ may be positive and larger than $\nu(k)$.

In the strong-field limit, $\lambda \gg \hbar\omega$ and the argument of the Bessel function in Eq. (11) is larger. Of course $\lambda \gg \hbar\omega$ depends upon the direction of **k**. However, since we shall be mainly interested in **k** parallel to the field amplitude $\mathbf{E}_0$, the condition $\lambda \gg \hbar\omega$ is essentially $|\mathbf{E}_0|$ large. For large values of argument, the Bessel function is small except when the order is equal to the argument. The sum over s in Eq. (11) may then be written approximately as

$$\sum_{s=-\infty}^{+\infty} J_s^2(\lambda/\hbar\omega)\delta(E-s\hbar\omega)=\tfrac{1}{2}[\delta(E-\lambda)+(E+\lambda)] . \tag{13}$$

The factor $\frac{1}{2}$ may be verified by integrating both sides of Eq. (13) with respect to $E=E_{p+k}-E_p-\hbar\omega_k$. The first δ function corresponds to the emission and the second to the absorption of $\lambda/\hbar\omega$ photons. In other words, for the intense-field case only multiphoton processes are significant and the electron-phonon collision takes place with the absorption or emission of $\lambda/\hbar\omega \gg 1$ photons. Substituting Eq. (13) in Eq. (11), the optical-phonon excitation rate becomes

$$\gamma(k)=\frac{\pi|V|^2}{\hbar}\sum_{\mathbf{p}}(f_{p+k}-f_p)[\delta(E_{p+k}-E_p-\hbar\omega_k-\lambda)+\delta(E_{p+k}-E_p-\hbar\omega_k+\lambda)] . \tag{14}$$

If we further assume that $\lambda \gg E_F$ (E_F is the Fermi energy) we may neglect the contribution of the processes in which photons are emitted compared to the contribution of processes in which photons are absorbed. This means that in the case $\lambda \gg E_F$ the contribution of the first δ function may be neglected so that Eq. (14) reduces to

$$\gamma(k)=\frac{\Omega|V_k|^2}{4\hbar\pi}\int_0^\infty dp\,p^2\int_{-1}^{+1}dx\,[f(E_p+\hbar\omega_k-\lambda)-f(E_p)]\delta(E_{p+k}-E_p-\hbar\omega_k+\lambda) , \tag{15}$$

after transforming the sum over **p** into an integral; here x denotes the cosine of the angle between **p** and **k** where the polar axis for the **p** integration was taken along the direction of **k**.

Approximating $f(E_p)$ by a step function and performing the integration in Eq. (15) one finally obtains

$$\gamma(k)=\frac{(m^*)^2e^2\omega_k(\epsilon_\infty^{-1}-\epsilon_0^{-1})}{2k^3\hbar^3}\left[\frac{e\mathbf{k}\cdot\mathbf{E}_0}{m^*\omega}-\omega_k\right] , \tag{16}$$

provided $\lambda \gg E_F$.

IV. RESULTS AND CONCLUSIONS

Equation (16) is the quantity of interest to us. It then follows from (16) that for a particular optical-phonon wave vector the necessary condition for the onset of the optical-phonon instability is just the Cerenkov condition $\mathbf{k}\cdot\mathbf{V}_0>\omega_k$, where $\mathbf{V}_0=e\mathbf{E}_0/m^*\omega$. The above result is formally analogous to the one for the electron-phonon system in the presence of a dc electric field. The difference lies in the fact that in the latter case $\mathbf{V}_0=e\mathbf{E}_0/m^*\omega$ is replaced by the drift velocity $\mathbf{V}_d$ as imposed by the static field.[4,5]

The condition $\lambda \gg E_F$ restricts the present theory[15] only to short-wavelength optical phonons. To see this let us consider in more detail the condition $\lambda \gg E_F$. For the sake of simplicity we shall consider only optical phonon propagating parallel to $\mathbf{E}_0$. For a given k the condition $\lambda=E_F$ defines a critical field strength E_c, namely

$$E_c=m^*\omega E_F\lambda\!\!\!^-/e\hbar , \tag{17}$$

$\lambda\!\!\!^-$ being the optical-phonon wavelength. If follows that the critical laser intensity such that $\lambda=E_F$ will be given by

$$I_c=\frac{c}{8\pi}\left[\frac{m^*\omega E_F\lambda\!\!\!^-}{e\hbar}\right]^2 .$$

To get a numerical estimate of the value of I_c let us consider the following values for the physical parameters of an n-type semiconductor such as GaAs: $m^*=0.068m_0$, $\epsilon_\infty=10.9$, $\epsilon_0=12.9$, $\omega_k\approx\omega_0=5.6\times10^{13}$ sec^{-1} (dispersion is neglected), $E_F\simeq10^{-14}$ ergs. Hence using a Nd:glass laser for which $\omega\approx2.0\times10^{15}$ sec^{-1}, it can be seen that I_c becomes

$$I_c=2.0\times10^{23}\lambda\!\!\!^{-2}\ \mathrm{W/cm^2} .$$

Hence for $\lambda\!\!\!^-=10^{-7}$ cm, I_c is of the order of 10^9 W/cm^2. For $\lambda\!\!\!^-=10^{-6}$ cm the power needed is 2 orders of magnitude higher which starts to impose some experimental difficulties as, for instance, damage to the crystal. Therefore, restricting ourselves to small values of $\lambda\!\!\!^-$ and using, for instance, a Nd:glass laser with a power density of the order of 10^9 W/cm^2, together with the values of the physical parameters given above for n-type GaAs, we estimate $\gamma(k)$ to be of the order of 10^{11} sec^{-1} for $\hbar=10^{-7}$ cm. One notices that under the above conditions $v_0=eE_0/m^*\omega$ is of the order of 10^7 cm sec^{-1}, which is greater than $v_{\mathrm{ph}}=\omega_k/k\approx4\times10^6$ cm sec^{-1}.

As mentioned before the actual condition for a net amplification of optical phonon requires the phonon growth rate to be greater than the loss $\nu(k)$, which is of the order of[16] $10^{10}-10^{11}$ sec^{-1}. Hence since the amplification factor $\gamma(k)$ may be of the order of or greater than $\nu(k)$, one might hope to achieve a net amplification of short-wavelength optical phonons when the semiconducting crystal is under an intense laser light. This is of particular interest since the conventional amplification of optical phonons via a dc electric field[4,5] favors phonons of relatively small k's (long wavelengths), i.e., $k=10^5$ cm^{-1}.

In conclusion, we have considered in this paper an electron-phonon system in the field of an intense electromagnetic wave. It was found that in the limit $\lambda \gg E_F$ only the electron-phonon collisions accompanied by multiphoton absorption are significant, and, as a result, the optical phonon population may grow with time when the field strength E_0 exceeds certain threshold value given by Eqs. (12) and (17). Furthermore, it was also shown that the present theory is restricted to short-wavelength optical phonons. The reason why the present mechanism is limited to high k is essentially due to the condition $\lambda \gg E_F$ which is only satisfied, from a practical point of view, for high k.

Physically, the optical-phonon instability in the field of a strong electromagnetic wave may be understood by assuming that the effect of the intense field is to give a drift velocity $v_0=eE_0/m^*\omega$ to the charge carriers. Hence when v_0 exceeds the phase velocity of the optical phonons, the phonon population in the Cerenkov cone may, in principle, grow with time. The actual observation of the population growth is achieved only if $\gamma(k)$ is greater than $\nu(k)$. A numerical estimate of $\gamma(k)-\nu(k)$ based upon the values of the physical parameters of the GaAs suggests to us that using a Nd:glass laser with a power density of about 10^9 W/cm^2 one might, in principle, observe a net amplification of optical phonons.

We finish by pointing out that our model contains a number of simplified assumptions. Nevertheless, some essential conclusions can be drawn therefrom. Amongst them, the present mechanism has the ability of exciting optical phonons of short wavelength (high values of k) propagating perpendicular to the z axis (i.e., parallel to $\mathbf{E}_0$). This is of particular interest since the conventional methods (e.g., drifting carriers[4,5]) favors amplification along the z axis. The amplification of short-wavelength optical phonons considered here can manifest itself experimentally in an increase in the absorption of the laser wave by the semiconducting sample.

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[1] A. B. Migdal, Zh. Eksp. Teor. Fiz. **34**, 1438 (1958) [Sov. Phys.—JETP **7**, 996 (1958)].

[2] V. L. Bouch-Bruevich, Fiz. Tverd. Tela (Leningrad) 2, 1857 (1960) [Sov. Phys.—Solid State **2**, 1678 (1961)].

[3] V. L. Gurevich and V. D. Kagan, Fiz. Tverd. Tela (Leningrad) **4**, 2441 (1962) [Sov. Phys.—Solid State **4**, 1788 (1963)].

[4] I. A. Chaban and A. A. Chadan, Fiz. Tverd. Tela (Leningrad) **6**, 2411 (1964) [Sov. Phys.—Solid State **6**, 1913 (1964)].

[5] I. Yokota, Phys. Lett. **10**, 27 (1964).

[6] E. M. Epshtein, Pis'ma Zh. Eksp. Teor. Fiz. **13**, 511 (1971).

[7] L. C. M. Miranda, J. Phys. C **9**, 2971 (1976).

[8] O. A. C. Nunes, J. Appl. Phys. **56**, 2694 (1984).

[9] E. M. Epshtein, Fiz. Tekh. Poluprovdn. **11**, 42 (1977) [Sov. Phys.—Semicond. **11**, 243 (1977)].

[10] O. A. C. Nunes, Phys. Rev. B **29**, 5679 (1984).

[11] V. I. Mel'nikov, Pis'ma Zh. Eksp. Teor. Fiz. **9**, 204 (1969) [JETP Lett. **9**, 120 (1969)].

[12] P. Roman, *Advanced Quantum Mechanics* (Addison-Wesley, Reading, 1965), p. 285.

[13] H. Reiss, Phys. Rev. A **1**, 803 (1970).

[14] H. Reiss, Phys. Rev. D **4**, 3533 (1971).

[15] In a general case, the photon-emission processes by electrons are also taken into account in the calculation of the optical-phonon growth rate and it can be seen by integrating Eq. (14) that these processes give a contribution to Eq. (16) by subtracting from it an extra term $A[(e\mathbf{k}\cdot\mathbf{E}_0/m^*\omega)+\omega_k]$, where A is the coefficient appearing in Eq. (16). In this case we will only have damping of optical phonons provided γ_k is always negative whatever the k values.

[16] S. Frota-Pessoa and R. Luzzi, Phys. Rev. B **13**, 5420 (1976).