Carrier-assisted laser pumping of optical phonons in semiconductors under strong magnetic fields

O. A. C. Nunes
*Laboratorio de Estado Solido, Departamento de Fisica, Universidade de Brasilia, 70910 Brasilia, Distrito Federal, Brazil and Department of Theoretical Physics, Oxford University, 1 Keble Road, Oxford OX1 3NP, United Kingdom**
(Received 25 April 1983; revised manuscript received 14 November 1983)

The theory of the generation and amplification of optical phonons in semiconductors via free carriers in the presence of laser radiation is extended to take into account quantizing magnetic fields. It is found that for the laser beam polarized perpendicular to the magnetic field direction, the optical-phonon system may reach instability as the cyclotron frequency of the free carriers approaches the laser frequency.

I. INTRODUCTION

Modern laser technology has stimulated theoretical investigations on the behavior of electrons in the field of a light wave. Much work has been done on the absorption of laser waves by free carriers (FC's) in semiconductors. Here, FC absorption means light absorption by electrons (holes) accompanied by either absorption or emission of phonons (acoustic and optical) or by the interaction of the carriers with impurities. One particular study of interest is that of the generation and amplification of optical phonons in semiconductors by electrons in the laser field, which has been considered on several occasions.[1–5] It has been shown in those papers that the absorption of laser radiation by free carriers can under certain conditions give rise to a strong emission of optical phonons by electrons rather than absorption, and as a consequence the phonon system may reach instability.

In this paper we are interested in extending the theory of the generation and amplification of optical phonons in semiconductors developed previously[1–5] to take into account the presence of quantizing (strong) magnetic fields. As is well known, in the presence of a strong magnetic field, the electronic energy levels in a band are split into subbands of discrete Landau levels. Laser-cyclotron resonance absorption occurs when a photon has sufficient energy to take an electron from one Landau level to the adjacent one (transitions between other than neighboring Landau levels in semiconductors are forbidden by selection rules in semiconductors having parabolic energy bands), and this transition is accompanied by the emission or absorption of an optical phonon.[6] On the other hand, it is natural to expect that external fields, changing the spectrum and the occupation number of the electron states, will considerably influence the spectrum and damping of optical phonons. In fact, as has recently been shown,[7] the presence of a strong magnetic field can significantly change the phonon damping in a semiconductor. Therefore, this led us to consider the question of how the carrier-assisted laser generation of optical phonons in semiconductors[1–5] may be affected by the presence of a strong magnetic field. A brief description of this work has been published earlier.[8]

II. HAMILTONIAN

We consider a monochromatic beam of photons from a laser source of frequency ω_L incident on an n-type semiconducting crystal. The Hamiltonian of the system is given by

$$H = H_E + H_L + H_R + H_{EL} + H_{ER} , \tag{1}$$

where H_E is the Hamiltonian for the many-electron system, H_L is that of the phonon system, H_R is the Hamiltonian of the radiation field, H_{EL} represents the electron-phonon interaction energy, and H_{ER} is the interaction between the radiation and the electron system. Here, we have not included the direct phonon-photon coupling as we are interested only in investigating the role of the carriers in the optical-phonon pumping. We assume the crystal to be in the presence of a static magnetic field H_0, with the associated vector potential given in the Landau gauge $A_0=(-yH_0,0,0)$. We work within the effective-mass approximation and take the one-electron wave functions to be the Landau states[9] $|\alpha=n,p_x,p_z\rangle$ where n is the Landau-level quantum number. The corresponding one-electron energies are

$$E_n(p_z)=(n+\tfrac{1}{2})\hbar\omega_c+\hbar^2p_z^2/2m ,$$

where $\omega_c=eH_0/mc$ is the cyclotron frequency of the free carriers. The deformation-potential contribution to the electron-phonon interaction energy is neglected here since, in the case of polar crystals, it is generally negligible when compared with the long-range electrostatic interaction between the electrons and longitudinal-optical phonons. Therefore, the electron-phonon interaction will be described in this case by the Fröhlich Hamiltonian,[10] and in the presence of the strong magnetic field it is given by

$$H_{EL}=\sum_{\alpha,\alpha',\vec{q}}(V_{EL}\langle\alpha'|e^{i\vec{q}\cdot\vec{r}}|\alpha\rangle C_{\alpha'}^{\dagger}C_{\alpha}b_{\vec{q}}^{\dagger}+\text{c.c.}) , \tag{2}$$

where

$$V_{EL}=i\frac{e}{q}\left[\frac{2\pi\hbar}{v}\right]^{1/2}\omega_0^{1/2}(\epsilon_\infty^{-1}-\epsilon_0^{-1})^{1/2}$$

and α,α' are the Landau states. The interaction of the electrons with the radiation field is given by

$$H_{ER}=\sum_{\alpha,\alpha',\vec{k}}\{V_{ER}\langle\alpha'|e^{i\vec{k}\cdot\vec{r}}\vec{e}_k\cdot[\vec{p}-(e/c)\vec{A}_0]|\alpha\rangle \times C^\dagger_{\alpha'}C_\alpha a_{\vec{k}}+\text{c.c.}\}\ , \tag{3}$$

where

$$V_{ER}=(1/m)(2\pi e^2\hbar/v\omega_L)^{1/2}\ ,$$

$\vec{p}=(\hbar/i)\vec{\nabla}$, $\vec{e}_k$ is the unit polarization vector of a photon $\vec{k}$ propagating parallel to the magnetic field and polarized along the x direction, and v is the volume of the crystal. Also in Eqs. (2) and (3), ω_0 is the LO-phonon frequency (dispersion has been neglected), and C_α, $b_{\vec{q}}$, and $a_{\vec{k}}$ are second-quantization operators for electrons, phonons, and photons, respectively.

III. OPTICAL-PHONON EXCITATION RATES

In this section we present the quantum-mechanical formulation of the excitation of optical phonons by the laser field via the free carriers in the presence of the strong magnetic field. This is very much similar to the photon conversion processes by charged particles encountered in plasma physics. In lowest order of perturbation theory,[11] the carrier-assisted photon-phonon conversion processes would be analogous to the Compton scattering of quantum field theory as shown in Fig. 1. The excitation rate $\gamma(\vec{q})$ may be calculated from the rate of change of the phonon population $N(\vec{q})$, $dN(\vec{q})/dt$, where $dN(\vec{q})/dt$ is given by the sum of the phonon probabilities for transitions in which phonons are emitted minus a similar sum of the probabilities for transition in which phonons are absorbed, averaged over the initial states, and summed over the final states. The emission or absorption of a LO phonon is accompanied by the absorption of a photon. The transition probabilities may be calculated by the usual methods of time-dependent perturbation theory[11] in terms of the second-order matrix element as

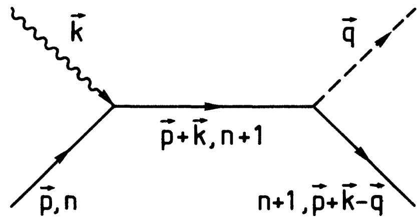

FIG. 1. Photon-phonon "Compton" scattering. The electron states ($\vec{p}$) are represented by solid lines, the phonons ($\vec{q}$) are represented by dashed lines, and the photons ($\vec{k}$) are represented by wavy lines. Here, n and $n+1$ are neighboring Landau levels.

$$W_i=\frac{2\pi}{\hbar}\sum_f|\langle f|M|i\rangle|^2\delta(E_f-E_i-\hbar\omega_L-\hbar\omega_0)\ , \tag{4}$$

where $\langle f|M|i\rangle$ is the second-order matrix element for this interaction, namely,

$$\langle f|M|i\rangle=\sum_I\left[\frac{\langle f|H_{ER}|I\rangle\langle I|H_{EL}|i\rangle}{E_i-E_I-\hbar\omega_0}+\frac{\langle I|H_{ER}|i\rangle\langle f|H_{EL}|I\rangle}{E_i-E_I+\hbar\omega_L}\right]. \tag{5}$$

Here, E_i and E_f are the initial and final energies of the electron, and $\hbar\omega_L$ and $\hbar\omega_0$ are the photon and phonon energies, respectively. The sum is over all intermediate states I of the carrier. To evaluate $\langle f|M|i\rangle$ we need to know the overlaps $\langle\alpha'|e^{i\vec{q}\cdot\vec{r}}|\alpha\rangle$ and

$$\langle\alpha'|e^{i\vec{k}\cdot\vec{r}}\vec{e}_k\cdot[\vec{p}-(e/c)\vec{A}_0]|\alpha\rangle\ .$$

With the use of the Landau wave functions[9] it can be seen that[7,12]

$$\langle\alpha'|e^{i\vec{q}\cdot\vec{r}}|\alpha\rangle=\delta_{p'_x,p_x+q_x}\delta_{p'_z,p_z+q_z}\lambda_{n',n} \tag{6}$$

with

$$\lambda_{n',n}=\delta_{n',n}e^{-\rho/2}L_n(\rho)+\Theta(n'-n)\left[\frac{n!}{n'!}\right]^{1/2}e^{i(n'-n)\phi}e^{-\rho/2}\rho^{(n'-n)/2}L_n^{n'-n}(\rho)+\Theta(n-n')\left[\frac{n'!}{n!}\right]^{1/2}e^{i(n-n')\phi}e^{-\rho/2}\rho^{(n-n')/2}L_{n'}^{n-n'}(\rho)\ . \tag{7}$$

For a photon propagating along the z axis and polarized parallel to the x direction we obtain

$$\langle\alpha'|e^{i\vec{k}\cdot\vec{r}}\vec{e}_k\cdot\left[\vec{p}-\frac{e}{c}\vec{A}_0\right]|\alpha\rangle=\delta_{p'_x,p_x}\delta_{p'_z,p_z+k}\left[\frac{m\hbar\omega_c}{2}\right]^{1/2}[(n+1)^{1/2}\delta_{n',n+1}-n^{1/2}\delta_{n',n-1}]\ . \tag{8}$$

Here, $L_n(\rho)$ are the associated Laguerre polynomials, Θ is the unit step function,

$$\Theta(n-n')=\begin{cases}1 & \text{if } n>n'\ ,\\ 0 & \text{if } n\le n'\ ,\end{cases}$$

$$\rho=\hbar q_\perp/2m\omega_c\ ,\quad \phi=\tan^{-1}(q_y/q_x)\ ,$$

and $q_\perp$ is the component of the phonon wave vector $\vec{q}$ perpendicular to the magnetic field.

The expression for $\lambda_{n',n}$ in (7) is very complicated for arbitrary $\vec{q}$ because, in general, both the $n\to n$ and $n\to n'$ $(n\neq n')$ transitions are possible. However, because of the selection rules for semiconductors having parabolic energy bands,[13] only transitions between neighboring Landau levels ($n'=n\pm1$) are allowed. For simplicity, we shall now consider only upward $n'=n+1$ electron transitions, which is valid if we assume here the ultraquantum limit in which the carrier concentration is such that only the $n=0$ Landau level is fully occupied. Under the foregoing considerations, Eq. (7) reduces to

$$\lambda_{n',n}=\left[\frac{n!}{n'!}\right]^{1/2} e^{i(n'-n)\phi}e^{-\rho/2}\rho^{(n'-n)/2}L_n^{n'-n}(\rho)\ , \quad n'>n\ . \tag{9}$$

If we now make use of the properties of the Laguerre polynomials,[14] Eq. (9) can be drastically simplified in the case when $\rho\ll1$ ($\rho=\hbar q_\perp^2/m\omega_c$). Under these conditions ($\rho\ll1$),

$$L_n^r(\rho)\approx\frac{(n+r)!}{n!r!}-\frac{(n+r)!}{(n-1)!(r+1)!}\rho\ .$$

With the use of these results for $L_n^r(\rho)$ and taking into account the ultraquantum limit ($n=0$), Eq. (9) reduces to

$$\lambda_{1,0}\simeq e^{i\phi}\rho^{1/2}\ . \tag{10}$$

The condition $\rho\ll1$ can easily be obtained by increasing the magnetic field strength ($\rho\propto1/H_0$) for a fixed phonon propagation direction. Hence, using Eqs. (5), (6), (8), and (10) we can write $\langle f\,|M|\,i\rangle$ as

$$\langle f\,|\,M\,|\,i\rangle=V_{EL}V_{ER}(m\hbar\omega_c/2)^{1/2}e^{i\phi}\rho^{1/2}\mathscr{E}\ ,$$

$$\mathscr{E}=\frac{1}{E_{0,p_z}-E_{1,p_z+k}+\hbar\omega_L}+\frac{1}{E_{0,p_z}-E_{1,p_z-q_z}-\hbar\omega_0}=\frac{\hbar^2kq_z/m}{(\hbar\omega_L-\hbar\omega_c-\hbar^2p_zk/m)^2}\ . \tag{11}$$

Having obtained $\langle f\,|M|\,i\rangle$, we can now look at the rate of change of the optical-phonon population due to the carrier-assisted process (Fig. 1),

$$(\hbar\omega_L)_{\vec{k}}+e_{\alpha=0;p_x,p_z}\to(\hbar\omega_0)_{\vec{q}}+e_{\alpha'=1;p_x-q_x,p_z+k-q_z}\ , \tag{12}$$

where an electron (hole) in an initial state $|\alpha=0,\ p_x,\ p_z\rangle$ absorbs a photon $\vec{k}=k\hat{z}$ and collides with an optical phonon $\vec{q}$, leaving in a final state $|\alpha'=1,p_x-q_x,\ p_z+k-q_z\rangle$, namely,

$$\frac{dN_q}{dt}=\gamma_qN_q\ , \tag{13}$$

with

$$\gamma_q=\frac{2\pi}{\hbar}\sum_{p_x,p_z}|\langle f\,|M|\,i\rangle|^2[N_L(N_q+1)f_\alpha(1-f_{\alpha'})-(N_L+1)N_qf_{\alpha'}(1-f_\alpha)]\times\delta(E_{\alpha'}-E_\alpha+\hbar\omega_0-\hbar\omega_L)\ . \tag{14}$$

Here, f_α is the Fermi distribution function and N_L is the number of photons with wave vector $\vec{k}$. Assuming further that N_q and N_L are large compared with unity, one obtains, from (14),

$$\gamma_q=\frac{2\pi}{\hbar}N_L\sum_{p_x,p_z}|\langle f\,|M|\,i\rangle|^2(f_\alpha-f_{\alpha'})\times\delta(E_\alpha-E_{\alpha'}+\hbar\omega_L-\hbar\omega_0)\ . \tag{15}$$

It follows immediately from (15) that if the photon-phonon system is at resonance ($\omega_L=\omega_0$), $E_{\alpha'}=E_\alpha$, and $\gamma_q=0$. In other words, for $\omega_L=\omega_0$ there is an exchange of energy between the bosonic systems having the carriers as spectators; the carrier-assisted phonon excitation is 0. On the other hand, if ω_L is near ω_0 but not necessarily at resonance (as is the case considered here), the carrier final energy differs slightly from its initial energy and the following approximation for the carrier-distribution function is assumed:

$$f(E_{\alpha'})=f(E_\alpha)+\hbar(\omega_L-\omega_0)\frac{\partial f(E_\alpha)}{\partial E_\alpha}\ . \tag{16}$$

Inserting this result into (15), we obtain the following for the optical-phonon excitation rate:

$$\gamma_q=2\pi N_L(\omega_0-\omega_L)\sum_{p_x,p_z}|\langle f\,|M|\,i\rangle|^2\frac{\partial f(E_\alpha)}{\partial E_\alpha}\times\delta(E_\alpha-E_{\alpha'}+\hbar\omega_L-\hbar\omega_0)\ . \tag{17}$$

Equation (16) relates the optical-phonon growth rate to the photon and carrier distributions. In particular, if γ_q is positive the phonon population is amplified at the expense of the laser field, whereas if $\gamma_q<0$ it is damped. Of course, a net growth of the phonon population is only achieved provided γ_q is greater than the lattice losses η_q which is typically[15] of the order of $10^{10}-10^{11}$ sec^{-1}. We now assume the electron gas to be degenerate, i.e., $k_BT\ll E_F$. In this case we may approximate $f(E_\alpha)$ by[16]

$$f(E_\alpha)=\begin{cases}1 & \text{for } E_\alpha<E_F\ ,\\ 0 & \text{for } E_\alpha>E_F\ ,\end{cases}$$

which is equivalent to saying that the perturbations in the electron gas caused by temperature do not considerably alter the carrier distribution. One obtains

$$\gamma_q=2\pi N_L(\omega-\omega_0)\sum_{p_x,p_z}|\langle f\,|M|\,i\rangle|^2\delta(E_\alpha-E_F)\times\delta(E_\alpha-E_{\alpha'}+\hbar\omega_L-\hbar\omega_0)\ . \tag{18}$$

The sums in (18) can now be performed easily with the help of the δ functions. Hence, upon taking the degeneracy in p_x, given by $m\omega_cL^2/2\pi\hbar$, γ_q is finally given by

$$\gamma_q=\frac{q_z^2q_\perp^2}{|k-q_z|}\frac{\pi e^4\omega_L\omega_0(N_L/V)(\epsilon_\infty^{-1}-\epsilon_0^{-1})(E_F-\hbar^2p_0^2/2m)}{2m^2\omega_c\hbar q^2c^2(\omega_L-\omega_c)^4}\times(\omega_L-\omega_0)\ , \tag{19}$$

provided that

$$|p_0| \leq p_F \,, \tag{20}$$

where

$$p_0 = \frac{m}{\hbar q_z}\left[\omega_L - \omega_0 - \frac{\hbar}{2m}(k - q_z)^2\right] . \tag{21}$$

In Eq. (19), $q_\perp = q \sin\theta$ where θ is the angle that $\vec{q}$ makes with the magnetic field direction.

IV. DISCUSSION

Equation (19) is the expression for the optical-phonon excitation rate via the free carriers in the presence of laser and strong magnetic fields. It indicates that the optical-phonon population grows with time if $\omega_L > \omega_0$. Physically, this means that a pumping field cannot generate optical phonons of frequency higher than its own frequency. The growth rate is no longer isotropic, being maximum for optical phonons propagating in a direction such that $q_\perp = q_z = q/\sqrt{2}$. Furthermore, upon taking into account conditions (20) and (21) indicated above, and the fact that in intraband absorption processes we always have $q \gg k$ (k is the photon wave number), the excited optical phonons are restricted to a relatively narrow band of q values in the interval $k \ll q \leq 2p_F$, p_F being the electron wave number at the Fermi level. The upper limit is set by considerations of momentum conservation. Since in an intraband process the photon gives insufficient momentum to the electron for the energy it provides to leave the electron in an allowed state, an electron can only emit an optical phonon whose maximum momentum is twice that of the electron. Since the maximum momentum of the electron is p_F, phonons with $q > 2p_F$ do not interact directly with electrons. By the time an instability is obtained when the condition $\gamma_q \geq \eta_q$ is satisfied (η_q are the linear losses), the phonon population grows with time and saturates because of the nonlinear relaxation mechanisms which should be effective in stabilizing these amplified optical phonons.[17] This ultimately limits the phonon population. As mentioned before, the condition $\gamma_q \geq \eta_q$ determines the laser threshold intensity for an actual optical-phonon amplification. To obtain an order-of-magnitude estimate of the critical value of N_L/V (photon density) we assume the following values for the physical parameters of an n-type InSb sample illuminated by a CO_2 laser ($\lambda = 10.8$ μm) in the magnetic field of 60 kG: $m = 10^{-29}$ g, $\epsilon_\infty = 14.06$, $\epsilon_0 = 18.0$, $\omega_0 = 3.8\times 10^{13}$ sec^{-1}, $n_0 = 10^{16}$ cm^{-3}, and $E_F = \hbar\omega_c$. Hence, assuming that η_q is typically 10^{10}–10^{11} sec^{-1}, and considering the case where the phonons are propagating in the direction $\theta = \pi/4$ for which $q_z = q_\perp = q/\sqrt{2}$, one obtains, for the photon density, $N_L/V = 3.0\times 10^{18}$ cm^{-3} with $q = 3\times 10^5$ cm^{-1} and $\omega_c = 1.0\times 10^{14}$ sec^{-1}. This, in turn, entails a laser intensity of about 10^9 W/cm^2, which is well within the experimental capabilities.

In conclusion, in this paper we have discussed the conditions of excitation and amplification of optical phonons in degenerate semiconductors under quantizing magnetic fields by free carriers in the presence of laser radiation. It was found that for the laser beam polarized perpendicular to the magnetic field, the optical-phonon amplification can be obtained through the electronic transitions between neighboring Landau levels accompanied by either emission or absorption of optical phonons. Furthermore, the excited phonons are restricted to a relatively narrow band of phonon wave numbers. For the laser beam polarized along the dc magnetic field, on the other hand, the dependence of the free-carrier absorption as well as of the amplification coefficient on the applied magnetic field only appears when the quantization of the electronic energy levels in the magnetic field becomes important, which occurs when the separation between adjacent Landau levels, $\hbar\omega_c$, is greater than either the collisional broadening $\hbar/\tau$ or the thermal broadening $k_B T$ of these Landau levels. This problem will be considered in a forthcoming paper.

ACKNOWLEDGMENTS

The author would like to acknowledge the support provided by a grant from the Brazilian National Research Council (Conselho National de Pesquisas). I also wish to thank Professor L. C. M. Miranda of Centro Tecnologico Aeronautica, to whom I am indebted for many stimulating discussions during his stay at Universidade de Brasilia.

---

*Present address.

[1]E. M. Epshtein, Fiz. Tekh. Poluprovodn. 11, 421 (1977) [Sov. Phys.—Semicond. 11, 243 (1977)].

[2]L. V. Gurevich and D. A. Parshin, Zh. Eksp. Teor. Fiz. 72, 1589 (1977) [Sov. Phys.—JETP 45, 834 (1977)].

[3]F. T. Vas'ko, Fiz. Tverd. Tela (Leningrad) 19, 3279 (1977) [Sov. Phys.—Solid State 19, 1916 (1977)].

[4]V. A. Yakovlev, Fiz. Tekh. Poluprovodn. 12, 1615 (1978) [Sov. Phys.—Semicond. 12, 954 (1978)].

[5]O. A. C. Nunes, Solid State Commun. 42, 451 (1982).

[6]E. R. Generazio and H. N. Spector, Phys. Rev. B 20, 5162 (1979).

[7]Y. Lee and N. Tzoar, Phys. Rev. 178, 1303 (1969).

[8]O. A. C. Nunes, Phys. Lett. 88A, 53 (1982).

[9]L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Pergamon, Oxford, 1958), p. 474.

[10]H. Fröhlich, Adv. Phys. 3, 325 (1954).

[11]D. Pines, *The Many Body Problem* (Benjamin, New York, 1962).

[12]M. A. F. Gomes and L. C. M. Miranda, Phys. Rev. B 12, 3788 (1975).

[13]B. Lax and J. Mavroides, in *Solid State Physics,* edited by F. Seitz and D. Turnbull (Academic, New York, 1962), Vol. 11, p. 261.

[14]See, e.g., *Bateman Manuscript Project,* edited by H. Erdelyi (McGraw-Hill, New York, 1953), Vol. 1, p. 39.

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

[16]C. K. N. Patel and E. D. Shaw, Phys. Rev. B 3, 1279 (1971).

[17]O. A. C. Nunes, Phys. Status Solidi B 112, K131 (1982).