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LETTER TO THE EDITOR

Phonon instability in the presence of two radiation fields†

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Abstract. The rate of change in the acoustic phonon population due to scattering by conduction electrons in the presence of two laser fields is calculated. It is found that longitudinal acoustic waves propagating parallel to the direction of polarisation of the radiation fields may be amplified over a relatively narrow frequency range.

Several mechanisms are known to give rise to phonon instability in semiconductors. These mechanisms include, amongst others, the influence of a DC electric field (Hutson *et al* 1961, Spector 1966), the direct conversion of radio and microwaves into acoustic waves (Abeles 1967, Miranda 1973, Zemel and Goldstein 1973) and the influence of a strong electromagnetic wave (Ephstein 1970, Miranda 1976, 1977). Our aim in this Letter is to draw attention to the last of these possibilities in connection with experiments in which a probe and a strong pumping field are present simultaneously. This occurs when phonon instability is studied via free-carrier absorption (Fan 1956). Apart from its possible application to experimental work, we believe that this subject is of sufficient interest to warrant an independent investigation.

We have therefore considered the scattering of phonons by electrons in the presence of two laser fields. The approach closely follows that used by Ephstein (1970) and Miranda (1976, 1977). The laser beams are treated as classical plane electromagnetic waves in the dipole approximation; the electron states are described by the solution to the Schrödinger equation for an electron in the laser fields. The electron–phonon scattering is treated by first-order perturbation theory, but with retention of the laser fields to all orders. The transition probabilities are then used to write a kinetic equation for the phonon population from which the rate of damping is obtained (Harris 1969).

We start with the solution to the time-dependent Schrödinger equation for an electron in the electromagnetic field of the laser beams, namely (Seely and Harris 1973)

$$\psi(\boldsymbol{x}, t) = V^{-1/2} \exp\left(\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{x} - (\mathrm{i}/2m\hbar)\int^{t} \mathrm{d}t'[\hbar\boldsymbol{p} - (e/c)\boldsymbol{A}(t')]^2\right). \tag{1}$$

Here $\boldsymbol{p}$ is the electron wavevector such that in the absence of the radiation fields the electron energy is $\epsilon_p = \hbar^2 p^2/2m$ and

$$\boldsymbol{A}(t) = (c/\omega_1)\boldsymbol{E}_1 \cos\omega_1 t + (c/\omega_2)\boldsymbol{E}_2 \cos\omega_2 t$$

is the vector potential of lasers 1 and 2 within the dipole approximation.

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The probability amplitude for a transition from state p_1 to state p_2 due to the collision with a phonon of momentum $\hbar k$ is then given by

$$a(1 \to 2; k) = -(i/\hbar) \iint d^3x\, dt\, \psi^*_{p_2}(x, t) V_k \exp[i(k \cdot x - \omega_k t)] \psi_{p_1}(x, t) \quad (2)$$

where V_k is the electron–phonon coupling. By substituting equation (1) into (2), performing the indicated integrations (Miranda 1976, 1977), and using the well-known relation between the scattering amplitude and the T matrix (Pomerantz 1965) we obtain the transition probability per unit time $T_{\nu_1,\nu_2}(1 \to 2; k)$ for the transition from state 1 ($p_1 = p$) to state 2 ($p_2 = p + k$) due to a collision with a phonon k with absorption ($\nu_1, \nu_2 > 0$) or emission ($\nu_1, \nu_2 < 0$) of $|\nu_1|$ and $|\nu_2|$ photons:

$$T_{\nu_1,\nu_2}(1 \to 2; k) = (2\pi/\hbar) |V_k|^2 J^2_{\nu_1}(\lambda_1/\hbar\omega_1) J^2_{\nu_2}(\lambda_2/\hbar\omega_2) \times \delta(\epsilon_{p_2} - \epsilon_{p_1} - \nu_1\hbar\omega_1 - \nu_2\hbar\omega_2 - \hbar\omega_k) \quad (3)$$

where $J_{\nu_i}(\lambda_i/\hbar\omega_i)$ is the Bessel function of order ν_i and $\lambda_i = \hbar k \cdot v_i$, with $v_i = eE_i/m\omega_i$, is the field parameter. The rate of change of the number of phonons k, dN_k/dt, is given in terms of the transition probability as (Harris 1969, Miranda 1976, 1977)

$$dN_k/dt = \gamma_k N_k$$

where

$$\gamma_k = (2\pi |V_k|^2/\hbar) \sum_{\nu_1,\nu_2=-\infty}^{+\infty} \sum_p J^2_{\nu_1}(\lambda_1/\hbar\omega_1) J^2_{\nu_2}(\lambda_2/\hbar\omega_2)(f_{p+k} - f_p) \times \delta(\epsilon_{p+k} - \epsilon_p - \nu_1\hbar\omega_1 - \nu_2\hbar\omega_2 - \hbar\omega_k). \quad (4)$$

In equation (4) f_p is the electron distribution function. If $\gamma_k > 0$, the phonon population grows with time, whereas for $\gamma_k < 0$ there is damping.

In the following we assume that laser 1 is the probe field and laser 2 is the strong pumping field. In the strong-field limit, $\lambda_2 \gg \hbar\omega_2$ and the argument of the Bessel function J_{ν_2} in equation (4) is large. For large values of the argument, the Bessel function is small except when the order is equal to the argument. The sum over ν_2 in equation (4) may then be written approximately as (Seely and Harris 1973, Miranda 1976, 1977)

$$\sum_{\nu_2=-\infty}^{+\infty} J^2_{\nu_2}(\lambda_2/\hbar\omega_2) \delta(\epsilon - \nu_2\hbar\omega_2) \simeq \tfrac{1}{2}[\delta(\epsilon - \lambda_2) + \delta(\epsilon + \lambda_2)]. \quad (5)$$

The factor $\frac{1}{2}$ may be verified by integrating both sides of equation (5) with respect to $\epsilon = \epsilon_{p+k} - \epsilon_p - \nu_1\hbar\omega_1 - \hbar\omega_k$. The first δ function corresponds to the absorption and the second to the emission of $\lambda_2/\hbar\omega_2$ photons. Substituting equation (5) into (4), the phonon damping then becomes

$$\gamma_k = (\pi |V_k|^2/\hbar) \sum_{\nu_1=-\infty}^{+\infty} \sum_p J^2_{\nu_1}(\lambda_1/\hbar\omega_1)(f_{p+k} - f_p)[\delta(\epsilon_{p+k} - \epsilon_p - \nu_1\hbar\omega_1 - \hbar\omega_k - \lambda_2) + \delta(\epsilon_{p+k} - \epsilon_p - \nu_1\hbar\omega_1 - \hbar\omega_k + \lambda_2)]. \quad (6)$$

Equation (6) gives the phonon damping in the presence of an intense laser with the simultaneous absorption, or emission, of $|\nu_1|$ photons of a probe field. If we restrict ourselves to one-photon transitions from the probe field, we shall retain in equation (6) only the $\nu_1 = \pm 1$ terms.

To proceed further, we now assume a classical Maxwellian distribution for the electrons. This assumption is relevant to a significant range of semiconductor materials and is valid provided the electron heating in the laser fields may be neglected. The latter, in turn, is valid if $e^2E^2/2m\omega^2 < \langle\epsilon\rangle$, where $\langle\epsilon\rangle = k_BT$ is the average energy of an electron in the absence of the radiation fields. Hence if we retain only $v_1 = \pm 1$ and perform the same calculations as those described by Seely and Harris (1973) and Miranda *et al* (1976, 1977), we finally obtain from equation (6)

$$\gamma_k = (2\pi^{1/2}n_0V|V_k|^2/\hbar m v_T^2)(v_s/v_T)J_1^2(\lambda_1/\hbar\omega_1)F(\alpha, \beta, a) \tag{7}$$

where V is the crystal volume, $\alpha = (\lambda_2 - \hbar\omega_1)/\hbar\omega_k$, $\beta = (\lambda_2 + \hbar\omega_1)/\hbar\omega_k$, $a = v_s/v_T$, v_s is the velocity of sound, $v_T = (2k_BT/m)^{1/2}$, and

$$F(\alpha, \beta, a) = 2\exp[-a^2(1+\beta^2)]\{[\beta\tanh(2\beta a^2) - 1]\cosh(2\beta a^2) + \exp[-a^2(\alpha^2 - \beta^2)][\alpha\tanh(2\alpha a^2) - 1]\cosh(2\alpha a^2)\}. \tag{8}$$

This expression for F is, in general, quite involved. A detailed analysis of it, however, indicates that it is most favourable for F to be positive when $\hbar\omega_1 \gg \lambda_2$ and $2\beta a^2 \ll 1$. Then $\beta = -\alpha = \omega_1/\omega_k$ and equation (8) reduces to

$$F \simeq 8\exp(-a^2)\exp(-x^2)(x^2 - \tfrac{1}{2}) \qquad x = \omega_1/kv_T. \tag{9}$$

It follows from equations (7) and (9) that, in contrast to the usual theories of sound amplification by a DC electric field (Hutson *et al* 1961, Spector 1966), the threshold condition for amplification is now dependent upon the value of k, instead of being the same for all values of k ($v_d > v_s$). This is seen from equation (9) which becomes positive for $x > 1/\sqrt{2}$ (or $k < \omega_1\sqrt{2}/v_T$), has a maximum at $x = \sqrt{\tfrac{3}{2}}$ and then decreases quite rapidly with increasing x. In other words, in the presence of a probe and a strong laser field, the phonon population in a relatively narrow range of values of k may become unstable, i.e. there is a selective mechanism for the excitation of high-frequency acoustic phonons, in great contrast to the usual results for sound amplification by a DC electric field.

Before proceeding, however, it is worthwhile to summarise the assumptions on which these results are based: (i) the electromagnetic waves should penetrate well into the sample, under the condition of applicability of the dipole approximation, which means that ω_1 and ω_2 should be such that $\omega_i\tau > 1$ and $\omega_i/\omega_p > 1$ (τ is the electron relaxation time) and (ii) assuming $\boldsymbol{k}$ is parallel to $\boldsymbol{E}_1$ and $\boldsymbol{E}_2$, equations (7) and (9) are valid provided $\omega_2 \ll kv_2 \ll \omega_1$, $k \gg 2v_s\omega_1/v_T^2$ and $v_i < v_T$. It can be shown that for a sample of InSb 1 mm × 1 mm in area, at $T = 50$ K and with a carrier concentration n_0 of about 10^{15} cm^{-3}, the above conditions are simultaneously satisfied for longitudinal acoustic waves in the |110| direction ($v_s = 3{\cdot}77 \times 10^5$ cm s^{-1}), if a CO_2 laser ($\lambda = 10$ μm) is used as laser 1 and an ICN laser ($\lambda = 538$ μm) with 115 mW of power well focused over the sample provides the pumping field. Under these circumstances ($\omega_2/\omega_1 = 0{\cdot}02$ and $v_2/v_T = 0{\cdot}1$), γ_k is negative for $k \gtrsim 10^7$ cm^{-1}, vanishes at $k = 8{\cdot}1 \times 10^6$ cm^{-1}, reaches a maximum at $k = 4{\cdot}7 \times 10^6$ cm^{-1} and finally becomes negligible for $k \lesssim 2 \times 10^6$ cm^{-1}.

Actually, the criterion for the onset of the instability is somewhat more involved. In our discussions so far we have completely ignored the phonon–phonon interaction which leads to a finite phonon lifetime. The actual criterion for the phonon instability, for a particular $\boldsymbol{k}$, is therefore

$$\gamma_k - \eta_k > 0$$

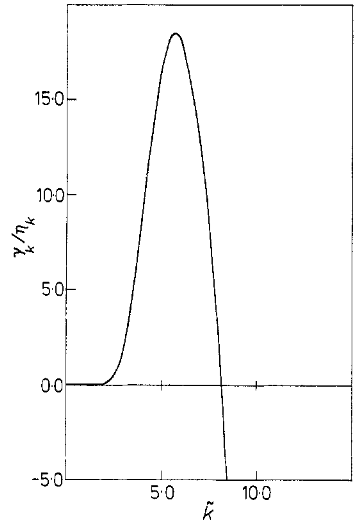

Figure 1. γ_k/η_k plotted against $\tilde{k} = k \times 10^{-6}$ for longitudinal acoustic waves propagating in the |110| direction for a sample of InSb with the following values of the physical parameters: $n_0 = 10^{15}\,\text{cm}^{-3}$; $v_s = 3{\cdot}77 \times 10^5\,\text{cm}\,\text{s}^{-1}$; $T = 50\,\text{K}$; $\gamma = 3$; laser 1 is a CO_2 laser ($\lambda = 10\,\mu\text{m}$); laser 2 is an ICN laser ($\lambda = 538\,\mu\text{m}$).

where η_k is the phonon decay rate due to anharmonic interaction. When the three-phonon interaction is the dominant anharmonic term, η_k is given by (Pomerantz 1965, Shiren 1965)

$$\eta_k = \pi^3\gamma^2\omega_k(k_B T)^4/120\rho v_s^5\hbar^3 \tag{10}$$

where γ ($\simeq 3$) is the ratio of an appropriate third-order elastic constant to a second-order constant (Shiren 1965) and ρ is the crystal density. Hence, combining equations (7), (9) and (10), assuming the hellium model (Pines 1963: $|V_k|^2 = 2\pi e^2\hbar\omega_k/Vk_D^2$; $k_D = \omega_p/v_T$) and using the above values for the physical parameters of InSb, we get

$$\gamma_k/\eta_k = (1{\cdot}57 \times 10^3)J_1^2(0{\cdot}09\tilde{k})\exp[-(5{\cdot}77/\tilde{k})^2][(5{\cdot}77/\tilde{k})^2 - \tfrac{1}{2}] \tag{11}$$

for longitundinal acoustic waves in the [110] direction with $T = 50\,\text{K}$ and $E_1 = 2{\cdot}5 \times 10^4\,\text{V}\,\text{cm}^{-1}$. Here $\tilde{k}$ is the value of the phonon wavevector in units of $10^6\,\text{cm}^{-1}$. Figure 1 shows γ_k/η_k plotted against $\tilde{k}$ for InSb with the parameters given above. As can be seen from this plot the unstable phonons are restricted to a relatively narrow range of values of k in the the vicinity of $5 \times 10^6\,\text{cm}^{-1}$.

In conclusion, it should be emphasized that our model contains a number of simplifying assumptions. Nevertheless, some essential conclusions can be drawn. Amongst them, the present mechanism has the ability of exciting high-frequency acoustic phonons propagating essentially in the direction of the polarisation of the laser fields ($\boldsymbol{E}_1$ and $\boldsymbol{E}_2$ are assumed to be parallel). For $\boldsymbol{k}$ not parallel to $\boldsymbol{E}_i$ the Bessel function in equation (11) becomes very small, which leads to damping rather than a growth of the phonon population. Secondly, the excited phonons are restricted to a relatively narrow range of values of k in the vicinity of $k \sim \omega_1/v_T$.

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