Decay of polar-optical phonons in semiconductors

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In electrical transport in compound semicondutors, the polar optical phonons can be driven from equilibrium by energetic phonon emission from the electrons. These phonons decay via bulk interactions with the acoustic modes through anharmonic terms of the crystal potential and through the second-order electric moment, or second-order piezoelectric, of the lattice. The relaxation rates for these processes are calculated and applied to InSb. It is found that the second-order electric moment dominates the decay of the polar modes although the two processes are comparable in magnitude.

I. INTRODUCTION

In many electrical transport problems in semiconductors, especially at low temperatures, the energy and momentum relaxation are provided by interaction of the electrons with optical phonons. In compound semiconductors, the pertinent mode is the longitudinal polar optical phonon. The emission of these phonons is the dominant energy and momentum relaxing process for the electrons, even at low temperatures where the optical modes are not strongly excited.[1] The emitted phonons can either decay to acoustic modes or be reabsorbed by the electrons. For either of these processes, the polar phonon distribution is driven out of equilibrium due to the phonon emission by the electrons. Experimentally, it has been verified that the phonon distribution is, in fact, disturbed by the electrons.[2] The phonon distribution itself is determined by a balance between phonon emission and absorption by the electrons and by decay of the polar modes into the acoustic modes. In many applications, the decay of the polar mode into acoustic modes can be characterized by a lifetime for the phonons.

The decay process arises primarily from a three-phonon interaction in which the polar phonon is annihilated and two acoustic phonons are created in a manner which conserves both the total phonon energy and momentum. This interaction occurs through the anharmonic terms of the lattice potential energy, similarly as for nonpolar modes.[3] A phenomenological treatment of the decay of nonpolar optical phonons via these anharmonic interactions has been given by Weinrich[4] and by Klemens.[5] Calculations for the polar mode have been presented by the present author,[6] but these incorrectly incorporate the lattice polarization into the interaction. In addition to the anharmonic decay it is also possible for the polar mode to decay through coupling of the electrical polarization of the polar optical phonon to two acoustic phonons by means of the second-order piezoelectric effect.[7] In this paper, the rate of polar optical phonon decay into acoustic modes is calculated for both the anharmonic interaction and the second-order piezoelectric interaction. This rate is coupled to the concept of a phonon lifetime for the case in which it is applicable.

II. POLAR OPTICAL MODES

The polar optical mode of vibration arises in compounds and crystals due to the ionicity of the chemical bond. Even in covalently bonded materials, the compounds that lack inversion symmetry exhibit ionic bonding to some extent, charterized by their effective charge. This effective charge contributes an electric polarization for the longitudinal optical vibrational mode that is aligned along the inter-ionic axis. The polarization is given by[7,8]

$$\vec{P}=\left(\frac{\hbar}{2\gamma\omega_0}\right)^{1/2}\sum_{\vec{q}}\vec{e}_q\,(a_q e^{i\vec{q}\cdot\vec{r}}+a_q^{\dagger}e^{-i\vec{q}\cdot\vec{r}}),$$

where a_q and $a_q^{\dagger}$ are the boson annihilation and creation operators, respectively,

$$1/\gamma=(\omega_0^2/4\pi)(1/\epsilon_\infty-1/\epsilon_0)\,, \tag{2}$$

ω_0 is the angular LO polar frequency, ϵ_∞ is the high-frequency dielectric constant, and ϵ_0 is the low-frequency dielectric constant. The factor $1/\gamma$ plays the role of the square of the effective charge normalized to the ionic reduced mass, so that the actual displacement vector is given by[7]

$$\vec{u}=\left(\frac{\hbar}{2M\omega_0}\right)^{1/2}\sum_{\vec{q}}\vec{e}_q\,(a_q e^{i\vec{q}\cdot\vec{r}}+a_q^{\dagger}e^{-i\vec{q}\cdot\vec{r}})\,, \tag{3}$$

where M is the average ionic mass of the lattice atoms. This result is just that expected for the nonpolar interaction. For that reason, we expect that the anharmonic interaction will yield results that compare favorably with the phenomenological results that have been obtained previously.[4]

III. ANHARMONIC DECAY

The polar optical phonons do not effectively transfer their energy to the surface, because of the extremely small value of the group velocity of these phonons. These phonons can decay, how-

ever, through a three phonon interaction involving the acoustic modes of the lattice. One of the possible three phonon interactions is due to the anharmonic terms of the crystal potential. To a normal system, a cubic term in the strain can be added to the crystal potential as[3]

$$V_{pp'} = \frac{1}{3!(N)^{1/2}} \sum_{\vec{q},\vec{q}',\vec{q}''} \vec{\xi}_q \vec{\xi}_{q'} \vec{\xi}_{q''} \vdots \underline{A} , \tag{4}$$

where N is the number of unit cells within the crystal, in Fourier transform form, and $\underline{A}$ is the third-order elastic-strain constant and is a third-rank tensor. For the two acoustic modes, $\vec{\xi}_{q'}$, $\vec{\xi}_{q''}$ are represented by

$$\vec{\xi}_q = \left(\frac{\hbar}{2M\omega_q}\right)^{1/2} \vec{e}_q (a_q e^{i\vec{q}\cdot\vec{r}} + a_q^\dagger e^{-i\vec{q}\cdot\vec{r}}) , \tag{5}$$

much as for the optical mode from Eq. (3). Retaining only the term involving the annihilation of a polar mode phonon and the creation of two acoustic modes, Eq. (4) becomes

$$V_{pp'} = \frac{1}{\sqrt{N}} \sum_{\vec{q},\vec{q}',\vec{q}''} \left(\frac{\hbar}{2M}\right)^{3/2} \left(\frac{1}{\omega_0\omega_{q'}\omega_{q''}}\right)^{1/2} \times \mathfrak{a}\, a_q a_{q'}^\dagger a_{q''}^\dagger , \tag{6}$$

where $\mathfrak{a}$ is the appropriate average over the various polarizations involved. The corresponding matrix element is then given by

$$|M_{pp'}|^2 = |\langle f|V_{pp'}|i\rangle|^2 = \frac{\hbar^3\mathfrak{a}^2}{8NM^3\omega_0 v_s^2} \sum_{\vec{q}',\vec{q}''} \left(\frac{1}{q'q''}\right) n_q(n_{q'}+1) \times (n_{q''}+1)\,\delta_{0,\vec{q}+\vec{q}'+\vec{q}''} , \tag{7}$$

where we have used the relation $\omega_{q'} = q'v_s$ for the two acoustic modes. The boson probability functions are the normal Bose-Einstein relations

$$n_q = (e^{\hbar\omega_q/k_BT} - 1)^{-1} . \tag{8}$$

The summation over $\vec{q}''$ is readily accomplished via the Kronecker δ function for the total phonon momentum.

The transition rate for decay of the polar mode phonon to the acoustic modes is given in terms of the matrix element as

$$\Gamma = \frac{2\pi}{\hbar} \sum_{\vec{q}',\vec{q}''} |M_{pp'}|^2 \delta(\hbar\omega_0 - \hbar q'v_s - \hbar q''v_s) , \tag{9}$$

or, using the above expression for $M_{pp'}$ and summing over q'',

$$\Gamma = \frac{\pi\hbar^2\mathfrak{a}^2}{4NM^3\omega_0 v_s^2} \sum_{\vec{q}'} \frac{1}{q'|\vec{q}'+\vec{q}|}\, n_q(n_{q'}+1) \times (n_{q+q'}+1)\delta(\hbar\omega_0 - \hbar q'v_s - \hbar v_s|\vec{q}'+\vec{q}|) . \tag{10}$$

In general, $\vec{q}$ is much smaller than $\vec{q}'$ so that the difference in the exponentials is small. We shall neglect this effect in the n_q terms. The summation over q' is changed into an integration as

$$\sum_{\vec{q}'} F(\vec{q}') \to \frac{V}{8\pi^3} \iiint F(q')q'^2 \sin\theta\, d\theta\, d\phi\, dq' . \tag{11}$$

The θ integration is easily carried out, yielding simply a factor of 2π. The remainder of the integral is

$$I = \frac{V}{4\pi^2} \int_0^\pi \int_0^\infty \frac{q'}{|\vec{q}'+\vec{q}|}\, \delta(\hbar\omega_0 - \hbar v_s q' - \hbar v_s|\vec{q}'+\vec{q}|)dq' \sin\theta\, d\theta . \tag{12}$$

For ease, the polar axis is taken as the q direction, so that

$$|\vec{q}'+\vec{q}| = (q'^2 + q^2 + 2qq'\cos\theta)^{1/2} . \tag{13}$$

The integration over θ involves the δ function and sets limits upon the range of q'. For example, we must have

$$\hbar\omega_0 - \hbar v_s q' - \hbar v_s(q'-q) < 0 , \qquad \hbar\omega_0 - \hbar v_s q' - \hbar v_s(q'+q) > 0 \tag{14}$$

or

$$\omega_0/2v_s - q < q' < \omega_0/2v_s + q . \tag{15}$$

Moreover, the δ function also requires

$$|\vec{q}'+\vec{q}| = \omega_0/v_s - q' , \tag{16}$$

so that the integration over θ yields the easily evaluated result

$$I = \frac{V}{4\pi^2\hbar v_s} \int_{\omega_0/2v_s - q}^{\omega_0/2v_s + q} \frac{dq'}{q} = \frac{V}{2\pi^2\hbar v_s} . \tag{17}$$

The resulting decay rate is then

$$\Gamma = \Gamma_a n_q (n_q'+1)^2 , \tag{18}$$

where

$$\Gamma_a = \hbar\mathfrak{a}^2 V/8\pi M^3 N v_s^3\omega_0 \tag{19}$$

and

$$n_q' = (e^{\hbar\omega_0/2k_BT} - 1)^{-1} . \tag{20}$$

The factor Γ_a in Eq. (19) differs from the phenomenologieal result of Weinrich[4] only in the numerical factors. These are very little different from his. However, his results do not include the temperature variation included in the boson occupation factors. There are very little data available on the third-order elastic constants of the III-V compounds, especially in the case of InSb, which we shall use as an example, due to the considerable interest in its electrical transport properties at high electric fields, where the optical phonon distribution may be disturbed. Moreover, the experiments which tend to show this disturbed distribution were in InSb.[2] The average elastic constant $\mathfrak{a}$ is an appropriate average over the third-

order elastic constants. A value of 3.7×10^{12} can be extrapolated from the data of Drabble and Brammer,[9] while similarly a value of 3.8×10^{12} is obtained from the work of Gubanov and Davydov.[10] Both values are in dyn/cm^2. These values were obtained primarily by ultrasonic waves propagating in the presence of an additional uniaxial stress. A value of 3.7×10^{12} dyn/cm^2 has been adopted for this work. The value of the other parameters used are their normal values. A result for Γ_a of 1.26×10^{10} sec^{-1} is found. The term in n_q' yields a slight modification of this at higher temperatures in addition to introducing a temperature variation of this decay rate.

IV. PIEZOELECTRIC DECAY

In addition to the anharmonic decay discussed above, the polar optical phonons can also decay into acoustic modes via the second-order piezoelectric, or second-order electric moment, interaction.[7] The potential term that is pertinent from the total crystal potential is just

$$V_{pp'} = \frac{1}{\sqrt{N}} \sum_{\vec{q},\vec{q}',\vec{q}''} \vec{E}_q \vec{\xi}_{q'} \vec{\xi}_{q''} \vdots \underline{Q} \tag{21}$$

in Fourier-transform format. The quantities $\vec{\xi}_{q'}$, $\vec{\xi}_{q''}$ are just those of the acoustic phonons from Eq. (5), while

$$\vec{E}_q = -4\pi\vec{P}_q = -4\pi\left(\frac{\hbar}{2\gamma\omega_0}\right)^{1/2}\vec{e}_q \times (a_q e^{i\vec{q}\cdot\vec{r}} + a_q^\dagger e^{-i\vec{q}\cdot\vec{r}}) . \tag{22}$$

Retaining only the terms for annihilation of one optical phonon and the creation of two acoustic phonons, the perturbing potential is just

$$V_{pp'} = \sum_{\vec{q}',\vec{q}''} \frac{2\pi\hbar}{M}\left(\frac{\hbar}{2\gamma N\omega_0\omega_{q'}\omega_{q''}}\right)^{1/2} \times Q a_q a_{q'}^\dagger a_{q''}^\dagger , \tag{23}$$

and the matrix element becomes

$$|M_{pp'}|^2 = |\langle f|V_{pp'}|i\rangle|^2 = \sum_{\vec{q}',\vec{q}''} \frac{2\pi^2\hbar^3Q^2}{M^2N\gamma\omega_0v_s^2} \times \left(\frac{1}{q'q''}\right)\delta_{0,\vec{q}+\vec{q}'-\vec{q}''} n_q(n_{q'}+1)(n_{q''}+1) . \tag{24}$$

Except for the constant factors, this matrix element has the same dependence on $\vec{q}', \vec{q}''$ as does the matrix element for the anharmonic decay. We can therefore use this term in Eq. (9), follow the same integration procedure, and obtain

$$\Gamma = \Gamma_p n_q (n_q'+1)^2 , \tag{25}$$

where

$$\Gamma_p = \frac{\hbar\, 2\pi VQ^2}{M^2N\gamma\omega_0v_s^3} = \frac{\hbar VQ^2\omega_0}{2M^2Nv_s^3}\left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0}\right) \tag{26}$$

or

$$\Gamma_P = \frac{16\pi^2M}{\gamma}\left(\frac{Q^2}{\mathcal{Q}^2}\right)\Gamma_a = 4\pi\omega_0^2\, M\left(\frac{Q^2}{\mathcal{Q}^2}\right)\left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0}\right)\Gamma_a \tag{27}$$

and n_q' is the same as that of Eq. (20).

There is very little data on the value of the second-order electric moment, especially in InSb. However, its relative importance can be inferred from the calculations of Flytzanis[11] for the relative effects of two phonon Raman scattering. Such an extrapolation gives the factor

$$(\mathcal{Q}/Q)^2 \sim 1.43\times10^4 \tag{28}$$

and

$$\Gamma_P = 1.5\,\Gamma_a = 1.9\times10^{10}\ \text{sec}^{-1} . \tag{29}$$

The combined decay rate then becomes

$$\Gamma = \Gamma_0(n_q'+1)^2 n_q , \tag{30}$$

where

$$\Gamma_0 = \Gamma_a + \Gamma_P \approx 3.16\times10^{10}\ \text{sec}^{-1} \tag{31}$$

for the values given above.

V. LIFETIME

The lifetime of the excess polar optical phonons is related not only to the rate at which they can decay into two acoustic phonons, but also to the rate at which they are generated by decay of the acoustic modes.[12] Thus, the continuity equation for the optical phonons may be written

$$\frac{dn_q}{dt} = -(\Gamma - G) , \tag{32}$$

where Γ is given by Eq. (30) and G is the rate at which the polar modes are generated by three-phonon processes. The matrix element in G is the same as that for Γ since the interactions are the same from considerations of detailed balance, but the operators are now $a_q^\dagger a_{q'}a_{q''}$ rather than the $a_q a_{q'}^\dagger a_{q''}^\dagger$ that were used in Eq. (6). The resulting change is in the phonon occupation numbers only, and we may write the generation rate of optical phonons due to the three-phonon process as

$$G = \Gamma_0(n_q+1)(n_q')^2 , \tag{33}$$

so that

$$\frac{dn_q}{dt} = -\Gamma_0[n_q(n_q'+1)^2 - (n_q+1)(n_q')^2] . \tag{34}$$

It is easily shown that, for n_q and n_q' given by Eqs. (8) and (20), respectively, the term in square brackets on the right-hand side of Eq. (34) vanishes in equilibrium. A steady-state phonon density therefore is maintained when the lattice is in

thermal equilibrium. For deviation from equilibrium, we write

$$n_q = n_{q,0} + \Delta n_q \ , \tag{35}$$

and then

$$\frac{d(\Delta n_q)}{dt} = -\Gamma_0 \Delta n_q (1 + 2n_q') \ . \tag{36}$$

The phonon lifetime is defined from

$$1/\tau_p = -(\Gamma - G)/\Delta n_q \ , \tag{37}$$

so that

$$1/\tau_p = \Gamma_0 (1 + 2n_q') \ . \tag{38}$$

The factor in parentheses involves a weak temperature dependence upon the lifetime of the polar modes. For example, for the numbers discussed above, τ_p is 3.16×10^{-11} sec at 4.2 °K, 2.29×10^{-11} sec at 77 °K, and 7.26×10^{-12} sec at 300 °K, where the values are seen to be mainly set by Γ_0. In this temperature range, τ_p changes only by about a factor of 4 for the entire range.

VI. DISCUSSION

The values of the polar optical-phonon lifetime that are computed here are, in general, somewhat larger than that obtained from measurements of the linewidth of Raman scattering in InSb. However, surface phonons play a role in Raman scattering,[13] and probably mask the role of the bulk phonon interactions. For the polar optical phonon, the dominant relaxation process appears to be via the second-order electric moment rather than the anharmonic interaction, although the two processes are of comparable magnitudes. The value of the phonon lifetime is considerably smaller than that required to explain the transport observations,[2] by two orders of magnitude, and the dominant time constant in those experiments is probably the electron energy relaxation time. The calculations for the disturbance of the phonon distribution by the hot electrons[6] depends upon the value of τ_p and probably largely overestimates the disturbance of the phonons if the considerably shorter lifetime calculated here is utilized.

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