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Dispersion, Mode-Mixing and the Electron-Phonon interaction in Nanostructures

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The electron-phonon interaction with polar optical modes in nanostructures is re-examined in the light of phonon dispersion relations and the role of the Fuchs-Kliwer (FK) mode. At an interface between adjacent polar materials the frequencies of the FK mode are drawn from the dielectric constants of the adjacent materials and are significantly smaller than the corresponding frequencies of the longitudinal optic (LO) modes at the zone centre. The requirement that all polar modes satisfy mechanical and electrical boundary conditions forces the modes to become hybrids. For a hybrid to have both FK and LO components the LO mode must have the FK frequency, which can only come about through the reduction associated with phonon dispersion relations. We illustrate the effect of phonon dispersion relations on the Fröhlich interaction by considering a simple linear-chain model of the zincblende lattice. Optical and acoustic modes become mixed towards short wavelengths in both optical and acoustic branches. A study of GaAs, InP and cubic GaN and AlN shows that the polarity of the optical branch and the acousticity of the acoustic branch are reduced by dispersion in equal measures, but the effect is relatively weak. Coupling coefficients quantifying the strengths of the interaction with electrons for optical and acoustic components of mixed modes in the optical branch show that, in most cases, the polar interaction dominates the acoustic interaction, and it is reduced from the long-wavelength result towards the zone boundary by only a few percent. The effect on the lower-frequency FK mode can be large.

1. Introduction

The interaction between electrons and the polar optical modes of crystalline nanostructure is central to the theory of electron transport in electronic devices working at and above room temperature. The standard approach assumes the Fröhlich interaction with long-wavelength modes. While this may be adequate for bulk material, it becomes questionable for nanostructures where the spectrum and structure of polar modes is affected by the presence of one or more interfaces between disparate crystalline layers. The necessity for allowable modes to satisfy mechanical and electrical boundary conditions forces, in general, the hybridization of longitudinally-polarized optical (LO), with transversely-polarized optical (TO) and electromagnetic interface (IF) modes, all at the LO frequency [1]. The difference between the LO (ω_L) and TO (ω_T) frequencies, forces the TO mode to be an interface mode. The electromagnetic dispersion governed by the large velocity of light entails that the electromagnetic component be also an interface mode. The further requirement that all components share the same frequency can be satisfied, in general, only by the property of phonon dispersion relations. It is this dependence on phonon dispersion relations that forces a re-examination of the polar interaction.

The IF mode was first introduced by Fuchs and Kliewer (FK) [2] in their model of the ionic slab, regarded as a dielectric continuum (DC). The allowed frequencies that satisfied the electrical boundary conditions in the slab exhibited a dispersion that Jones and Fuchs [3] showed was in error as a result of the neglect of mechanical boundary conditions. In the case of a single interface there are two frequencies that are determined by the condition $\varepsilon_1(\omega) + \varepsilon_2(\omega) = 0$, where $\varepsilon_{1,2}(\omega)$ are the permittivities of the adjacent materials. In neither case do these frequencies coincide with the corresponding long-wavelength frequencies of the LO modes. This is not a problem for the DC theory, but, in order for the FK mode to be a component of a hybrid, phonon dispersion relations is essential. Moreover, an analysis of the electron-phonon interaction in the case of a single heterostructure, showed that at a FK frequency the FK mode became dominant in the interaction [4].

This can be made clear by quoting the expression for the interaction potential associated with one of the adjacent layers in the single heterostructure:

$$\phi(\mathbf{q}, \mathbf{r}) = -i\alpha_0 A e^{i(q_x x - \omega t)} (\cos q_z z - \Gamma \sin q_z z - s p e^{-q_x z})$$

$$\alpha_0 = e^* / V_0 \varepsilon_\infty, \quad e^* = \omega_T^2 \mu V_0 (\varepsilon_s - \varepsilon_\infty) \quad 1$$

$$\Gamma = (q_x / q_z) p [1 - (q_x / \eta p)(p - 1)] \quad 2$$

$$p = [s(1 + R)]^{-1}$$

The factors s and R are, respectively, the field factor and the permittivity ratio associated with the IF mode:

$$s = \frac{\omega^2 - \omega_T^2}{\omega_L^2 - \omega_T^2} \quad 3$$

$$R = \varepsilon_W(\omega) / \varepsilon_B(\omega) \quad \varepsilon(\omega) = \varepsilon_\infty \left(\frac{\omega^2 - \omega_L^2}{\omega^2 - \omega_T^2} \right)$$

$$A = \frac{1}{Q} \left(\frac{\eta}{2\mu\omega} \right)^{1/2} (a_q + a_{-q}^\dagger) \quad 4$$

$$Q^2 = \frac{1}{2} (q_x^2 + q_z^2) (1 + \Gamma^2) + \frac{p q_x (2 + p)}{L} \xrightarrow{L \rightarrow \infty} \frac{1}{2} (q_x^2 + q_z^2) (1 + \Gamma^2)$$

The subscripts W and B refer to well (where the electrons are) and barrier. V_0 is the volume of the unit cell; q_x is the wave vector in the plane of the interface (isotropy is assumed), q_z the wave vector normal to the plane, ω_T and ω_L are the zone-centre frequencies of the non-polar (TO) mode, and the LO mode, μ is the reduced mass, η the exponent of the TO component, assumed to be large and L is the barrier thickness.

The permittivity associated with the LO mode is unaffected by dispersion:

$$\varepsilon(\omega) = \varepsilon_\infty \left(\frac{\omega^2 - \omega_L^2 + v_L^2 (q_x^2 + q_z^2)}{\omega^2 - \omega_T^2 + v_L^2 (q_x^2 + q_z^2)} \right) \quad 5$$

v_L is the velocity that determines the dispersion, the latter assumed to be quadratic ($\omega^2 = \omega_L^2 - v_L^2 (q_x^2 + q_z^2)$). Note that the condition $1 + R = 0$ occurs at the frequencies of the FK mode. When this applies, the potential associated with the FK mode dominates.

Given the necessity for phonon dispersion relations in order for allowable modes to exist, we need to learn how the polar interaction is affected by dispersion. A simple model of the relevant lattice dynamics reveals that an

important effect is the mixing of acoustic and optical modes, leading to a diminution of the strength of the polar interaction associated with the optical branch and a corresponding diminution of the strength associated with the acoustic branch.

2. A linear-chain model of mode-mixing

An analytic model of the 3D lattice dynamics of the diamond lattice that models the dispersion of both optical and acoustic modes is available [5], and the conversion to the zincblende lattice is straightforward. However, the model embodies the long-wavelength approximation, so it cannot be used to explore mode mixing in the general case. There have been a number of papers on the lattice dynamics, for example Jones and Fuchs [3], Rucker et al [6] and, more recently, Tutuncu and Srivastava [7]. These papers focus on the dispersion of phonon modes, but do not explicitly describe the mixing of optical and acoustic modes and its consequence for the electron-phonon interaction, which is the subject of our paper. The effect can be sufficiently well illustrated by a simple linear-chain model consisting of ions of two different types.

The coupled equations of motion are [8]:

$$\begin{aligned} \omega^2 m^A u^A + 2fu^B \cos qa - 2fu^A - 2\gamma(u^A - u^B) &= 0 \\ \omega^2 m^B u^B + 2fu^A \cos qa - 2fu^B - 2\gamma(u^B - u^A) &= 0 \end{aligned} \quad 6$$

m^A, m^B are the masses, u^A, u^B are the displacements, f is the nearest-neighbour force constant, a is the separation of the ions at equilibrium, γ is the polar force constant. We change the variables to acoustic and optical displacements according to:

$$\begin{aligned} u^A &= U + r^B u, \quad u^B = U - r^A u, \\ r^A &= m^A / M, \quad r^B = m^B / M, \quad M = m^A + m^B \end{aligned} \quad 7$$

U, u are the acoustic and polar displacements. Inserting gives, after some manipulation, two coupled equations of motions:

$$\begin{aligned} (\omega^2 - 8(f/M)\sin^2(qa/2))U + 8(f/M)r\sin^2(qa/2)u &= 0 \\ 8(f/\mu)r\sin^2(qa/2)U + [\omega^2 - 2(f + \gamma)/\mu + 8(f/M)\sin^2(qa/2)]u &= 0 \end{aligned} \quad 8$$

In the limit of long wavelengths we can identify:

$$\omega_T^2 = 2f/\mu, \quad \omega_L^2 = 2(f + \gamma)/\mu$$

where $r = (r^A - r^B)/2$. The magnitude of the mass factor, r , is clearly a vital quantity in mode-mixing. Solutions of the coupled equations exist provided that the determinant of the coefficients vanishes, a condition that defines the dispersion:

$$\left(\frac{\omega_L}{\omega_T} \right)^2 = \frac{1}{2} \left[\left(\frac{\omega_L}{\omega_T} \right)^4 \pm \left(\left(\frac{\omega_L}{\omega_T} \right)^4 - 16 \frac{\mu}{M} \sin^2(qa/2) \left\{ \left(\frac{\omega_L}{\omega_T} \right)^2 - 4 \left(\frac{\mu}{M} + r^2 \right) \sin^2(qa/2) \right\} \right)^{1/2} \right] \quad 9$$

Once the dispersion is determined, the mixing of the modes can be quantified. In the optical branch:

$$\frac{U}{u} \equiv \lambda_{op} = \frac{-4(\mu/M)r\sin^2(qa/2)}{(\omega/\omega_T)^2 - 4(\mu/M)\sin^2(qa/2)} \quad 10$$

In the acoustic branch:

$$\frac{u}{U} \equiv \lambda_{ac} = \frac{-4r\sin^2(qa/2)}{(\omega/\omega_T)^2 - (\omega_L/\omega_T)^2 + 4(\mu/M)\sin^2(qa/2)} \quad 11$$

The appearance of an acoustic element in the optical branch reduces the polarity. A measure of this reduction can be estimated noting that mode-mixing can have no effect on the energy of the mixed mode, which is proportional to μu_0^2 , where μu_0^2 is associated with the energy of the mode at long wavelengths.

Thus:

$$MU^2 + \mu u^2 = \mu u_0^2 \quad 12$$

The square of the optical displacement is a measure of the scattering potential associated with polarity, thus:

$$(M\lambda_{op}^2 + \mu)u^2 = \mu u_0^2$$

$$\left(\frac{u}{u_0}\right)^2 \equiv p_{op} = \left(\frac{\mu/M}{\mu/M + \lambda_{op}^2}\right) \quad 13$$

Taking the square of the acoustic displacement as a measure of the scattering potential associated with the deformation, we have:

$$\left(\frac{U}{U_0}\right)^2 \equiv p_{ac} = \left(\frac{1}{1 + (\mu/M)\lambda_{ac}^2}\right) \quad 14$$

It can be shown that $p_{op} = p_{ac}$: the drop in polarity of the optic mode equals the drop in acousticity of the acoustic mode, a symmetry we might have intuitively expected.

To illustrate actual rates, we focus on the optical branch. The scattering rate, W is given by the Fermi Golden Rule:

$$W = \frac{2\pi}{\eta} \int \left| \langle f | H_{ep} | i \rangle \right|^2 \delta(E_f - E_i) dN_f \quad 15$$

Where E_i, E_f are the initial and final electron energies respectively, $\langle f |, |i \rangle$, are the final and initial eigenstates of the electron, H_{ep} is the Hamiltonian for the electron-phonon interaction and the integral is performed over the density of final states. For optical and acoustic modes with the same frequency and wave vector, the respective rates are quantified by the following coupling coefficients depicting power:

$$\text{Opticalmode} \quad C_{op}^2(\omega, q) = \frac{e^2 \omega_L^2}{V_0 \omega q^2} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_s} \right) \quad 16$$

$$\text{Acousticmode} \quad C_{ac}^2(\omega, q) = \frac{\Xi^2 q^2}{M\omega}$$

V_0 is the volume of the unit cell; $\omega_L, \epsilon_\infty, \epsilon_s$ are, respectively, the zone-centre LO frequency, the high-frequency permittivity and the static permittivity; Ξ is the deformation potential. Due to mode mixing, the optical component is reduced by the factor p_{op} . The equivalent factor for the acoustic mode is obtained from

$$U^2 = \lambda_{op}^2 u^2 = \lambda_{op}^2 p_{op} u_0^2 = \lambda_{op}^2 p_{op} (M/\mu) U_0^2 \quad 17$$

The strengths of the optical and acoustic components are then:

$$C_{op}^{*2} = p_{op} C_{op}^2$$

$$C_{ac}^{*2} = \lambda_{op}^2 p_{op} (M/\mu) C_{ac}^2 \quad 18$$

3. Mixed modes in semiconductors

We have examined the mixing of modes in a few semiconductors, specifically GaAs, InP, GaN and AlN. Data for GaAs, InP and cubic GaN/AlN are

from [9], [10], [11] respectively. In the linear-chain model the distance apart of the ions was taken to be $a_0/4$, where a_0 is the lattice constant. Results are shown in Figs. 1-4. The dispersion curves are in rough agreement with observation. Mode-mixing is significant only for InP, GaN and AlN. The long-wavelength approximation, as regards the electron-phonon interaction, is excellent for GaAs throughout the zone (Fig. 6). Even for InP, GaN and AlN the drop in polarity towards the zone boundary is weak.

The polar optical phonon hybrid that satisfies electrical and mechanical boundary conditions at the interface consist of LO, TO and EM interface mode components. All must have the same frequency. Under conditions when the EM interface mode becomes a FK mode, dominating the interaction with electrons, the LO component is required to have the same frequency, which it can have via dispersion, but this requires the hybrid to have a large wavevector. This affects the interaction strength and, moreover, introduces mixed-mode effects.

In a bulk model of the electron – phonon interaction, the effect of mode mixing is negligible, because energy and momentum conservation limits the phonons to relatively small wave vectors in the usual case. In a nanostructure, where both mechanical and electrical boundary conditions apply, a FK mode, hybridized with an LO mode, can have a frequency that puts it near the zone boundary, or even outside the Reststrahlen bands. The frequencies of the FK modes in AlN/GaN, derived from standard properties, are 105meV and 73.9meV (1.24 and 1.06, in units of the relevant TO frequency). Both lie outside the linear-chain bands. Relative to the experimentally-determined bands quoted by Bennett et al [12] (Fig.5), only the lower frequency lies outside the GaN band. In this case only the AlN frequency FK mode would provide scattering in the GaN.

Our results are based on an extremely simple model. A full account of the relevant lattice dynamics would be desirable. As regards the electron-phonon interaction, an exact account would not only have to take into account the role of dispersion and of mode-mixing, but also of the effect of elastic anisotropy which mixes transverse and longitudinal polarizations. Both polar and deformation potentials will be active in both branches, and be dependent, like the frequency, on wave vector, making the necessary summations over wave vector more computer intensive. Fortunately, phonon dispersion relations is relatively weak along with the associated mode-mixing, and elastic anisotropy can be accounted for by using appropriate averages for the elastic constants. In practice, the polar interaction with electrons is affected by mode mixing only near the zone boundary.

We conclude that a knowledge of LO dispersion in a system is essential in the determination of the strength of the electron-phonon interaction in nanostructures. Assuming that the dispersion is quadratic in wave vector, we need only the LO frequencies at the Γ and X points to provide the breadth of the Reststrahlen band.

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Figure 1 Dispersion for GaAs (blue, dotted), GaN (red - solid), InP (black – dash-dot) & AlN (green-dash).

Figure 2 Ratio of displacements: u/U for the acoustic branch, U/u for the optic branch GaAs (blue, dotted), GaN (red - solid), InP (black – dash-dot) & AlN (green-dash).

Figure 3 Drop in the polarity of the optic branch, and the equal drop in acousticity in the acoustic branch GaAs (blue, dotted), GaN (red - solid), InP (black – dash-dot) & AlN (green-dash).

Figure 4 Coupling coefficients of mixed modes. The upper curves are for the optical component, the lower curves for the acoustic component GaAs (blue, dotted), GaN (red - solid), InP (black – dash-dot) & AlN (green-dash).

Fig.5 Dispersion of optical modes ($\Gamma-X$) for AlN and GaN [10]. The phonon energies of the two Fuchs-Kliwer modes are 105 and 73.9 meV.