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The Extended Hybrid Model For Optical Phonon Confinement

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Abstract

The theory of optical phonons and their confinement in heterostructures is reviewed with analogies made to the theory of acoustic phonons. The dielectric continuum (DC) model is obtained when bulk dispersion is ignored and the hybrid model is found when dispersion is included. The issue of boundary conditions is investigated and the solutions are shown with comparisons made using electron-phonon scattering.

1. Introduction

One of the main mechanisms determining key parameters of electronic and optoelectronic devices at high temperatures is optical phonon scattering. It is well known that these modes may be confined in different layers of a heterostructure [1]. The theoretical investigation of optical phonon confinement has historically been contentious. The dielectric continuum (DC) model [2,3] is the simplest method but it is known not to explain Raman spectra. The hybrid model [1] does explain the Raman scattering results and gives similar results to the DC model for the electron-phonon interaction [4]. However, this has only been applied to the GaAs/AlAs system where the *reststrahl* bands of the two materials are well separated. An extension of the hybrid theory is now timely because the systems such as GaN/AlN have overlapping reststrahl bands [5]. In this paper we review the results already obtained and extend the theory to describe these systems.

2. The equation of motion and the solution

We may write a general equation of motion for the phonon mode displacement $\mathbf{u}$ in an isotropic cubic binary polar crystal as [6]

$$\rho\ddot{\mathbf{u}} = d_1\mathbf{E} - d_2\mathbf{P} + \nabla(a\nabla.\mathbf{u}) - \nabla\times(b\nabla\times\mathbf{u}) + \mathbf{F} \tag{1}$$

The constants depend on the type of phonon we are describing, acoustic or optical. The terms involving the elastic constants a and b describe the dispersion of the modes, as will be shown. The last term is included by comparing with microscopic theory and the components of $\mathbf{F}$ have the form [1,6]

$$F_x = -\frac{\partial}{\partial x}\left[(b+b')\left(\frac{\partial u_y}{\partial y}+\frac{\partial u_z}{\partial z}\right)\right]+\frac{\partial}{\partial y}\left[(b+b')\frac{\partial u_y}{\partial x}\right]+\frac{\partial}{\partial z}\left[(b+b')\frac{\partial u_z}{\partial x}\right]. \qquad (2)$$

The constant b is the same as in the equation of motion while the remaining elastic constant b' is determined by the elastic response to rotation [1,6]. This function is zero in a bulk crystal since the material constants do not change. However, at an interface $\mathbf{F}$ is non-zero and will add terms to the boundary conditions (BCs). This term is important because, without it in the acoustic case, the correct boundary conditions from acoustic theory would not be obtained.

For acoustic modes, $\mathbf{u}$ is the displacement of the centre of mass of the unit cell and ρ is the density. Since the positive and negative ions move together in phase there is no polarisation field $\mathbf{P}$ or electric field $\mathbf{E}$, hence $\mathrm{d}_1 = \mathrm{d}_2 = 0$.

For optical phonons, $\mathbf{u}$ is the relative displacement of the ions in the unit cell and ρ is the reduced mass density of these ions. In this case a polarisation field is produced and we must include Maxwell's equations to describe the electric fields. Hence, we also have the following equations (in the non-retarded limit) [3]

$$\mathbf{P} = [\rho\omega_T^2(\epsilon_s-\epsilon_\infty)]^{1/2}\mathbf{u}+\epsilon_0(\epsilon_\infty-1)\mathbf{E},$$

$$\nabla\times\mathbf{E}=0, \qquad (3)$$

$$\nabla.\mathbf{D}=0,$$

and d_1=$\mathrm{d}_2\epsilon_0(\epsilon_s$-1), d_2=$[\rho\omega_T^2\epsilon_0^{-1}(\epsilon_s-\epsilon_\infty)]^{1/2}$ Here $\mathbf{D}$=$\epsilon_0\mathbf{E}$+$\mathbf{P}$ is the electic displacement field, ω_T is the transverse optical (TO) phonon frequency, $\epsilon_s(\epsilon_\infty)$ is the static (high frequency) dielectric constant. The longitudinal optical (LO) frequency is given by $\omega_L^2 = \omega_T^2\epsilon_s/\epsilon_\infty$.

We have two problems: how to solve Eqs. (1)-(3) rigorously and what the boundary conditions are. The solution in bulk material is usually written as the sum of the longitudinal and transverse parts of the displacement [7], viz,

$$\mathbf{u}=\mathbf{u_L}+\mathbf{u_T};$$

$$\nabla\times\mathbf{u_L}=0, \quad \nabla\cdot\mathbf{u_T}=0, \qquad (4)$$

and $\mathbf{u}\infty e^{iwt}$. We now try and work towards solutions which have the form of the plane wave equation, $\nabla^2\mathbf{u}$+$q^2\mathbf{u}$=0, where $\mathbf{q}$ is the total wavevector. The additional condition obtained will be the dispersion relation.

We start by trying to find the transverse solution $\mathbf{u}_T$ by taking the curl of Eq. (1), which gives

$$\nabla \times [\rho\omega^2 \mathbf{u_T} - b\nabla \times (\nabla \times \mathbf{u_T}) - d_2\mathbf{D}] = 0. \tag{5}$$

Note the electric field $\mathbf{E}$ is always zero for this case from the Maxwell equations in Eq. (3) and the fact that there is no magnetic field. Using the identity $\nabla \times (\nabla \times \mathbf{u}) = -\nabla^2\mathbf{u} + \nabla(\nabla.\mathbf{u})$ we may obtain the desired solution if $q^2 = \rho(\omega^2 - \omega_T^2)/\mathrm{b}$, which is is the solution for TO phonons. The relations $\mathbf{E}_T$ and $\mathbf{D}_T = [\rho\omega_T^2\epsilon_0(\epsilon_s - \epsilon_\infty)]^{1/2}\mathbf{u_T}$ are also obtained. For transverse acoustic (TA) phonons $\mathrm{d}_2 = 0$ and we obtain $q^2 = \rho\omega^2/b$. Since this is the dispersion relation we identify b as the elastic constant $c_{44} = \rho\nu_T^2$ where v_T is the transverse velocity. For acoustic modes this leads to the usual linear dispersion. For optical modes the dispersion is normally negative which means we have to change the sign of c_{44} [8].

For the longitudinal solution $\mathbf{u_L}$ we take the divergence of Eq. (1), which gives

$$\nabla.[\rho\omega^2 \mathbf{u_L} + a\nabla(\nabla.\mathbf{u_L}) + (d_1 + d_2\epsilon_0)\mathbf{E}] = 0. \tag{6}$$

Note, for longitudinal modes it is not guaranteed that the displacement field $\mathbf{D}$ is zero. This does not change the method of solution and we may get the result

$$\nabla.(\nabla^2 \mathbf{u_L} + q^2\mathbf{u_L}) = 0, \tag{7}$$

with $\mathbf{a}$ identified as the elastic constant $c_{11} = \rho v_L^2$ where v_L is the longitudinal velocity. Again the sign of c_{11} is normally negative for optical phonons. Hence, we have the dispersion $\omega = v_L q$ for longitudinal acoustic (LA) modes and $\omega^2 = \omega_L^2 - v_L^2 q^2$ for LO modes.

In order to solve Eq. (7), we introduce a potential function Φ, $\mathbf{u} \propto \nabla\Phi$. This gives the equation

$$\nabla^2(\nabla^2 + q^2)\Phi = 0, \tag{8}$$

and we see that the solution of this equation is a linear combination of the solutions of two equations: $(\nabla^2 + q^2)\Phi = 0$ and $\nabla^2\Phi = 0$. We identify these solutions, respectively, as longitudinal modes and interface (IF) modes. For LO modes we obtain the relations $\mathbf{E_L} = -[\rho\omega_T^2(\epsilon_s - \epsilon_\infty)/\epsilon_0\epsilon_\infty^2]^{1/2}\mathbf{u_L}$ and $\mathbf{D_L} = 0$

For the IF modes, $\nabla^2\Phi = 0$ implies that the displacement satisfies both $\nabla \times \mathbf{u} = 0$ and $\nabla . \mathbf{u} = 0$. Note, for acoustic modes this is not a solution of Eq. (1) and, hence, the interface mode part should not be included in this case. The IF mode solution is possible for optical modes and we obtain the relations $\mathbf{E} = -[\rho\omega_T^2(\epsilon_s - \epsilon_\infty)/\epsilon_0\epsilon_\infty^2]^{1/2}(\omega^2 - \omega_T^2)/(\omega_L^2 - \omega_T^2)\ \mathbf{u}$ and $\mathbf{D} = \epsilon(\omega)\mathbf{E}$ where $\epsilon(\omega) = \epsilon_\infty(\omega^2 - \omega_T^2)/(\omega_L^2 - \omega_T^2)$. However, the solution will contain exponential terms. In bulk media these will not converge at infinity so again they are not a solution in this case. For a heterostructure this solution is possible because different exponential terms can be taken in different parts of the structure. These modes are due

to the presence of interfaces and this is why they are called interface modes. The total displacement is the sum of $\mathbf{u}=\mathbf{u}_L+\mathbf{u}_T+\mathbf{u}_I$, if we have a heterostructure.

We now have our solution of Eq. (1) for both acoustic and optical phonons apart from one amplitude. If we have a heterostructure the boundary conditions will connect the solutions in each region again leaving one amplitude. This amplitude must be defined by a quantisation condition. The reader is directed to [1,3] for extensive details. The result is

$$\hat{H} = \frac{\rho}{4}\left[\int |\dot{\hat{u}}|^2 d^3r - \omega^2 \int [\hat{u}|^2 d^3r\right] = \sum_q \hbar\omega \left(a_q^\dagger a_q + \frac{1}{2}\right). \tag{9}$$

We now need to investigate what the boundary conditions should be.

3. Boundary conditions

The mechanical boundary conditions (BCs) are obtained by integrating Eq. (1) across an interface and ensuring the continuity of the equation. If we assume a planar interface perpendicular to the z axis and that the wave propagates in the x direction in the plane (we may do this without loss of generality), we obtain the mechanical BCs as continuity of

$$u_x \quad and \quad u_z, \tag{10}$$

$$a\nabla.\mathbf{u} - (b+b')\frac{\partial u_x}{\partial x} and b\frac{\partial u_x}{\partial z} + b'\frac{\partial u_z}{\partial x}. \tag{11}$$

We also have the usual electrostatic BCs of E_x and D_z continuous in the case of optical phonons from continuity of Eq. (3).

As mentioned previously, the elastic constant b′ is determined by the elastic response to rotation. For acoustic displacements rotational strains vanish and, hence, b′ = b. [6] If this is included in Eq. (11) the usual acoustic BCs are obtained. Note, that this is only true because **F** is included in Eq. (1). For optical displacements, rotational strains need not vanish; indeed it is not known how b′ is related to b [6]. The elastic constant in this case can only be determined from microscopic theory [6] and some progress has recently been made [9]. One suggestion was that b' = -b [6] which is the other extreme from the acoustic BCs, optical BCs, in analogy to electromagnetic waves which require a material with purely rotationally elastic properties. Note, this solution is no better than acoustic BCs but it is another simple set of mechanical BCs to use.

For the GaAs/AlAs system this does not present a problem; the frequencies of the materials are separated and one may ignore the modes in one material while the modes in the other material are propogating [4]. In effect, this ignores the BCs in Eq. (11) and sets $\mathbf{u} = 0$ at the boundaries. For the GaN/AlN system, however, the *reststrahl* bands do overlap and this requires that the modes be present in both materials necessitating the need for the second set of boundary conditions in Eq. (11). This is the extension of the hybrid model.

We could further reduce the number of BCs to just the electrostatic BCs if we ignore bulk dispersion. In this case the frequencies of each mode cannot overlap and the solution separates into the separate LO, TO and IF solutions. This leads to the dielectric continuum (DC) model [2,3] which could be derived purely from electrostatic theory if the dielectric functions of the materials are known. The hybrid model can therefore be regarded as an extension of the DC theory including bulk dispersion of the modes and, what is more important, the additional mechanical BCs.

For this paper we will assume both sets of mechanical BCs and also the limit of the dielectric continuum model and compare the differences. We will now find the solution for a quantum well and calculate the electron-phonon scattering rates for a general system of materials.

4. Results

We assume a quantum well of material 1 (GaN) between material 2 (AlN) with interfaces at z = ±a/2 (see Figure 1). For the most general solution, we start by assuming that all of the LO and TO modes are evanescent and decay from the interfaces in the z direction. The frequency of the mode is ω and must be the same for each individual part of the solution, i.e.

$$\omega = \left[\omega_{L1,2}^2 - \nu_{L1,2}^2(q^2 - k_{L1,2}^2)\right]^{1/2} = \left[\omega_{T1,2}^2 - \nu_{T1,2}^2(q^2 - k_{T1,2}^2)\right]^{1/2}, \qquad (12)$$

where q is the in-plane wavevector (which we assume is in the x direction) and $k_{L1,2}$ ($k_{T1,2}$) is the LO (TO) mode wavevector perpendicular to the plane. With this solution no part is propogating in the z direction. When the frequency of the mode is covered by the bulk dispersion of a part of the solution, the perpendicular wavevector of that part becomes imaginary and travelling waves are obtained as the solution for that part.

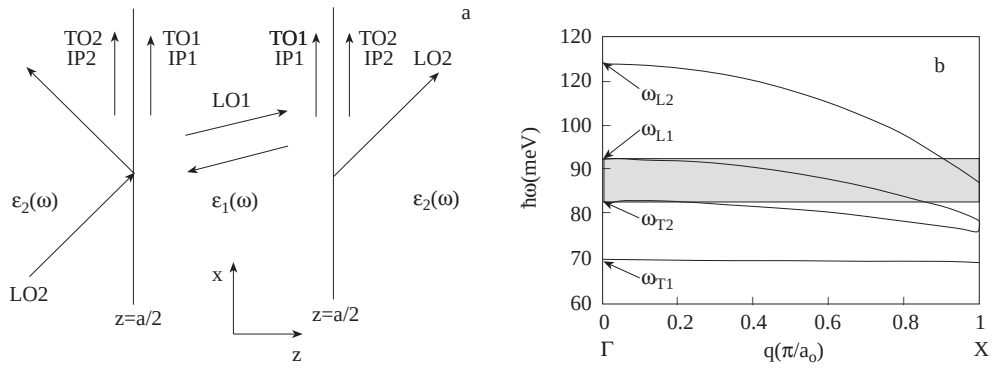

Figure 1. (a) A schematic of the structure and an example of the solution with LO2 and LO1 modes propagating. (b) A parabolic apporximation to the bulk phonons of GaN/AlN. The shaded area is where the reststrahl bands overlap.

For the quantum well we choose the solution:

$$\begin{aligned}
\mathbf{u_{L2}} &= \left[qA_L e^{-k_{L2}z} + qBe^{k_{L2}z}, 0, ik_{L2}A_L e^{-k_{L2}z} - ik_{L2}Be^{k_{L2}z}\right] e^{iqx}, \\
\mathbf{u_{T2}} &= \left[k_{T2}A_T e^{-k_{T2}z} + k_{T2}Ce^{k_{T2}z}, 0, iqA_T e^{-k_{T2}z} - iqCe^{k_{T2}z}\right] e^{iqx}, z \leq -a/2; \quad (13) \\
\mathbf{u_{I2}} &= \left[qDe^{qz}, 0, -iqDe^{qz}\right] e^{iqx},
\end{aligned}$$

$$\begin{aligned}
\mathbf{u_{L2}} &= \left[qEe^{-k_{L2}z}, 0, ik_{L2}Ee^{-k_{L2}z}\right] e^{iqx}, \\
\mathbf{u_{T2}} &= \left[k_{T2}Fe^{-k_{T2}z}, 0, iqFe^{-k_{T2}z}\right] e^{iqx}, \qquad z \geq -a/2; \quad (14) \\
\mathbf{u_{I2}} &= \left[qGe^{-qz}, 0, iqGe^{-gz}\right] e^{iqx},
\end{aligned}$$

$$\begin{aligned}
\mathbf{u_{L1}} &= \left[qHe^{-k_{L1}z} + qIe^{k_{L1}z}, 0, ik_{L1}He^{-k_{L1}z} - ik_{L1}Ie^{k_{L1}z}\right] e^{iqx}, \\
\mathbf{u_{T1}} &= \left[k_{T1}Je^{-k_{T1}z} + k_{T1}Ke^{k_{T1}z}, 0, iqJe^{-k_{T1}z} - iqke^{k_{T1}z}\right] e^{iqx}, \quad [z] \leq a/2 \quad (15) \\
\mathbf{u_{I1}} &= \left[qLe^{-qz} + qMe^{qz}, 0, iqLe^{-qz} - iqMe^{qz}\right] e^{iqx},
\end{aligned}$$

We then apply the electrostatic BCs and the mechanical BCs in Eqs. (10) and (11) and solve the twelve equations for different frequency ranges. If the LO2 (TO2) mode can propagate $\mathrm{A}_T = 0$ ($\mathrm{A}_L = 0$), k_{L2} (k_{T2}) becomes imaginary and the other constants are found in terms of A_L (A_T). This case is a mode from material 2 impinging on the interface and exciting the other modes (see Figure 1(a)). The normalisation condition defines the last constant as if it were a bulk mode in material 2 [4]. If no mode in material 2 can propagate, $\mathrm{A}_L = \mathrm{A}_T = 0$ and a dispersion relation is obtained [4]. These are the modes in material 1 which are mainly confined. Here the normalisation condition must be carried out to obtain the final constant. These solutions are used for the GaN/AlN system (see Figure 1(b)).

We may now take two limits. If we separate the frequencies of the two materials, we find that we can ignore the modes in material 1 (2) when the frequencies are in the range of the reststrahl band of material 2 (1) [4]. However, we must still keep an electric field due to the interface phonons in all regions. This solution converges to the hybrid solution for the GaAs/AlAs system [4] if the BCs of the form in Eq. (11) are used. These BCs then become redundant. If we reduce the bulk dispersion of all the modes, the solutions separate and we find that the mechanical BCs in Eqs. (10) and (11) become redundant; this is the DC model result.

To compare the different solutions, we calculate the elecron-phonon interaction assuming that the well has infinite barriers. We calculate the intrasubband emission rate in the ground state with initial electron energy $2\hbar\,\omega_{L1}$, which will involve symmetric modes, and the intersubband emission rate from the first excited state to the ground state with zero initial electron energy, which will involve the antisymmetric modes both with T = 300K. These results are shown in Figures 2(a) and 2(b), respectively, for the three sets of BCs (the DC model and the hybrid model with acoustic and optical mechanical BCs) for the GaN/AlN system. It is very difficult to distinguish the curves which is a consequence

of the weak dispersion in these systems and the fact that we have summed over all the phonon modes.

There is an approximate sum-rule [10] which should still apply here. We have a complete, orthogonal set with the BCs we have taken in all three cases. Although it cannot be shown easily in a mathematical context, this should imply that the results be similar [11]. Also, the DC model is only the zero bulk dispersion limit of the hybrid model and the bulk dispersion of the optical phonons in semiconductors is weak. The full result (and the hybrid model for the GaAs/AlAs system) also has resonant points in the dispersion where the DC phonon modes would be [3]. Hence, the correct mechanical BCs are not important in this case.

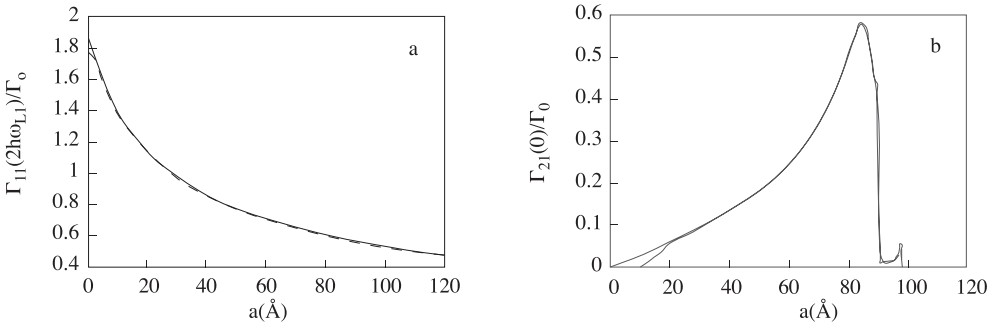

Figure 2. The (a) intrasubband and (b) intersubband electron-optical phonon emission rates. The solid curve is the DC result, the dashed curve the hybrid model using acoustic BCs and the dotted curve using optical BCs.

Figure 3 shows an example of the parallel displacement and electric potential for a propagating TO mode in material 2 and a propagating LO mode in material 1. There is also a resonance with the interface mode around this frequency. Note, the TO mode does not have an electric potential associated with it.

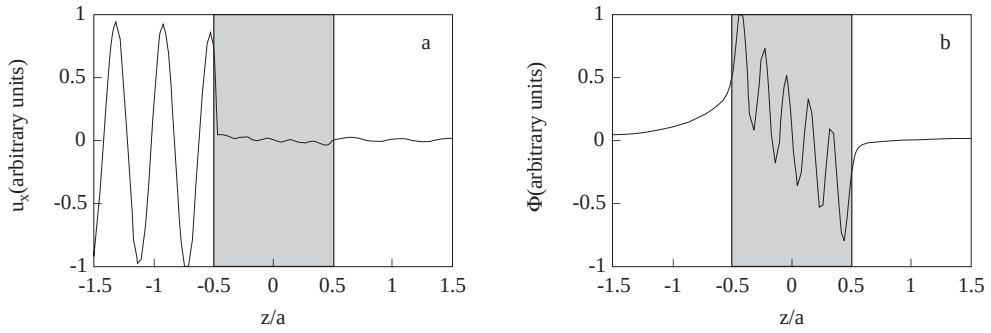

Figure 3. The (a) parallel relative displacement u_x and (b) electric potential Φ for a mode of energy $\hbar\omega = 81$meV using the hybrid model with acoustic boundary conditions.

Although the scattering rates may not be different this does not mean that the modes are the same. Work is continuing to find the dispersion of the modes for a superlattice to compare and contrast the long wavelength frequencies obtainable in Raman scattering for different mechanical BCs. It is at this wavevector range where the mechanical BCs are expected to be important.

5. Conclusions

The optical phonons of heterostructures have been derived with analogy to acoustic theory. The boundary conditions have been derived and attention should be drawn to the extra elastic constant b' which can only be obtained from microscopic theory. It has been shown that, as far as the evaluation of total electron-phonon scattering rates are concerned, the simple dielectric continuum model, which only includes electrostatic boundary conditions, is valid.

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