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Interface Phonons in Semiconductor Double Heterostructures

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Interface Phonons in Semiconductor Double Heterostructures

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Abstract

Within the framework of the continuum model, the equation of motion for the polarization vector in a semiconductor double heterostructure is solved exactly for the interface phonon modes. Both the eigenvectors and dispersion relations are obtained analytically. It is shown that the slab modes observed in right-angle Raman scattering in a GaAs quantum can be understood in terms of the interface modes found in this paper.

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1. Introduction

The optical phonon spectrum in semiconductor heterostructures and superlattices has been investigated extensively in recent years by Raman scattering experiments. It is by now well recognized that the reduced dimensionality gives rise to phonon modes that are fundamentally different from those in the bulk. In particular, the interface modes [1,2], confined bulk modes[3,4] and confined slab modes [5] have all been observed.

Theoretically, Fuchs and Kliewer [6] examined the optical modes of vibration in the long-wavelength limit for an isolated slab of ionic crystal. They found the surface optical (SO) modes as well as the bulk longitudinal optical (LO) modes. More recently, Wendler [7] considered a double-layer structure and derived interface modes of optical phonons, although the dispersion relations for the interface phonons in a double heterostructure (DHS) were already obtained by Lassnig [8] by means of the energy loss method.

Since the peculiar slab modes reported in Ref. 5 cannot be understood on the basis of any theory mentioned above, the continuum model of lattice vibration was not pursued further. On the other hand, there has not been any rigorous theory based on a microscopic approach except for the linear-chain model [4].

We present in this Letter the theory of interface phonons in a semiconductor DHS using the continuum model. No approximation other than the long-wavelength limit, which is intrinsic in the model, is assumed throughout our calculation. Analytic expressions for the eigenvectors and the dispersion relations are found. It is shown that the symmetric and antisymmetric interface modes each split into two branches at the center of the Brillouin zone, with their frequencies given by ω_L (the bulk longitudinal optical) and ω_T (the bulk transverse optical (TO)), respectively. Consequently, lattice vibrations with longitudinal polarization may occur at the TO frequency and vice versa. A detailed discussion of the theory including the confined bulk modes will be published elsewhere [9].

2. Theory

For a DHS with a thin layer of material 1 sandwiched between two thick layers of material 2, the equation of motion for the relative displacement $u(\vec{r},t)$ of the ion pair in material ν ($\nu = 1,2$) can be written, in the continuum approximation, as

$$\mu_\nu \ddot{\vec{u}}_\nu(\vec{r},t) = -\mu_\nu \omega^2_{o\nu} \vec{u}_\nu(\vec{r},t) + e^*_\nu \vec{E}(\vec{r},t) \quad , \tag{1}$$

where μ is the reduced mass of the pair of ions, $\mu\omega^2_o$ is the short-range force constant not including Coulomb fields, e^* is the effective charge of the ions, and $\vec{E}(\vec{r},t)$ is the local electric field. The polarization field $\vec{P}(\vec{r},t)$ produced by the oscillating ions is given by

$$\vec{P}(\vec{r},t) = n_\nu e^*_\nu \vec{u}(\vec{r},t) + n_\nu \alpha_\nu \vec{E}(\vec{r},t) \quad , \tag{2}$$

where n is the number of ion pairs per unit cell and α is the polarizability. The local field $\vec{E}$ in (2) is related, in the long-wavelength limit, to the polarization by

$$\vec{E}(\vec{r},t) = \frac{4\pi}{3} \vec{P}(\vec{r},t) + 4\pi \int d\vec{r}' \; \Gamma(\vec{r}-\vec{r}')\cdot\vec{P}(\vec{r}') \quad , \tag{3}$$

where Γ denotes the Green tensor with components

$$\Gamma_{\alpha\beta} = \frac{1}{4\pi} \frac{\partial^2}{\partial x_\alpha \partial x_\beta} \frac{1}{|\vec{r}-\vec{r}'|} \quad . \tag{4}$$

If we assume $\vec{P}(\vec{r},t) = \vec{P}(\vec{r})e^{i\omega t}$, we find the equation of motion for the polarization by substituting Eqs. (2) and (3) into (1),

$$\left[\frac{\lambda_\nu - \lambda_{o\nu}}{\alpha_\nu n_\nu(\lambda_\nu - \lambda_{o\nu}) - 1} - \frac{4\pi}{3}\right] \vec{P}(\vec{r}) = 4\pi \int d\vec{r}' \; \Gamma(\vec{r}-\vec{r}')\cdot\vec{P}(\vec{r}') \quad , \tag{5}$$

where we have defined the parameters

$$\lambda_\nu^2 = 4\pi\omega^2/\omega_{p\nu}^2 \quad , \qquad \lambda_{o\nu}^2 = 4\pi\omega_{o\nu}^2/\omega_{p\nu}^2 \quad , \tag{6a,b}$$

with the ion plasma frequency $\omega_{p\nu}^2 = 4\pi n_\nu e_\nu^{*2}/\mu_\nu$.

Since translational symmetry in the z-direction is destroyed by the presence of interfaces, we introduce the two-dimensional vectors $\vec{\kappa}$ and $\vec{\rho}$ so that $\vec{k} = (\vec{\kappa},q)$ and $\vec{r} = (\vec{\rho},z)$. A two-dimensional Fourier transform of (5) then leads to the matrix equation

$$4\pi \begin{bmatrix} \chi_\nu^{-1}(\omega) & 0 & 0 \\ 0 & \chi_\nu^{-1}(\omega) & 0 \\ 0 & 0 & \chi_\nu^{-1}(\omega) \end{bmatrix} \cdot \ldots,z)$$

$$= \frac{2\pi}{\kappa} \int_{-\infty}^{\infty} dz' \; e^{-\kappa(z-z')} \; \vec{K}\vec{K}\cdot\vec{P}(\vec{\kappa},z) \tag{7}$$

in which the z-component has been left unchanged, $\vec{K} = [\vec{\kappa}, i\Theta(z)\kappa]$ where $\Theta(z)$ is the step function, and the function $\chi_\nu^{-1}(\omega)$ is defined by

$$4\pi\chi_\nu^{-1}(\omega) = \frac{\lambda_\nu - \lambda_{o\nu}}{\alpha_\nu n_\nu(\lambda_\nu - \lambda_{o\nu}) - 1} - \frac{4\pi}{3} \quad . \tag{8}$$

Here $\chi_\nu(\omega)$ is the isotropic dielectric susceptibility and is related to the dielectric function $\epsilon_\nu(\omega)$ by $\chi_\nu(\omega) = \epsilon_\nu(\omega) - 1$, where

$$\epsilon_\nu(\omega) = \epsilon_{\infty\nu}(\omega_{L\nu}^2 - \omega^2)/(\omega_{T\nu}^2 - \omega^2) \tag{9a}$$

$$\epsilon_{\infty\nu}(\omega) = 1 + 4\pi\alpha_\mu n_\nu/(1 - \frac{4\pi}{3}\alpha_\nu n_\nu) \quad , \tag{9b}$$

and the LO and TO phonon frequencies are defined by

$$\omega_{L\nu}^2 = \omega_{o\nu}^2 + \frac{2}{3}\omega_{p\nu}^2/(1 + \frac{8\pi}{3}\alpha_\nu n_\nu) \tag{10a}$$

$$\omega_{T\nu}^2 = \omega_{o\nu}^2 - \frac{1}{3}\omega_{p\nu}^2/(1 - \frac{4\pi}{3}\alpha_\nu n_\nu) \quad . \tag{10b}$$

To describe the propagation of interface phonons, it is more convenient to express the polarization vector as

$$\vec{P}(\vec{\kappa}, z) = (\vec{\pi}, P_s) = (P_\kappa, P_z, P_s)$$

$$= P_\kappa(\vec{\kappa}, z)\hat{\kappa} + P_z(\vec{\kappa}, z)\hat{z} + P_s(\vec{\kappa}, z)\hat{s} \quad , \tag{11}$$

where the unit vector $\hat{s}$ is defined as $\hat{s} = \hat{z} \times \hat{\kappa}$. Substituting (11) into (7), we can separate the s-component and decouple (7) into the two equations

$$\begin{pmatrix} \chi_v^{-1}(\omega) & 0 \\ 0 & \chi_v^{-1}(\omega)\epsilon_v(\omega) \end{pmatrix} \cdot \vec{\pi}(\vec{\kappa},z) = \frac{1}{4\pi}\int_{-\infty}^{\infty} dz'\ M(z-z')\cdot\vec{\pi}(\vec{\kappa},z') \qquad (12)$$

for the so-called p-polarization, and

$$\chi_\nu^{-1}(\omega)\ P_s(\vec{\kappa},z) = 0 \qquad (13)$$

for the s-polarization which is not of concern here. The matrix M in (12) is Hermitian and is given by

$$M(z-z') = M^{\dagger}(z'-z) = -\ 2\pi\kappa e^{-\kappa|z-z'|} \begin{pmatrix} 1 & i\theta(z-z') \\ i\theta(z-z') & 1 \end{pmatrix} . \qquad (14)$$

Equation (12) defines an eigenvalue problem for the normal modes of vibrations. The coupled integral equations can be more easily solved by first transforming them into coupled differential equations. Differentiating (12) twice and at the same time requiring a non-vanishing coefficient determinant, we find

$$\frac{d}{dz}\ P_\kappa(\vec{\kappa},z) = i\kappa P_z(\vec{\kappa},z) \qquad (15a)$$

$$\frac{d^2}{dz^2}\ \vec{\pi}(\vec{\kappa},z) = \kappa^2\vec{\pi}(\vec{\kappa},z) \quad . \qquad (15b)$$

The solutions to (16) take the form

$$P_{\kappa}(\vec{\kappa},z) = \begin{cases} iA_2 e^{\kappa z} , & z < 0 \\ i(A_1 e^{\kappa z} - B_1 e^{-\kappa z}) , & 0 \le z \le a \\ -iB_2 e^{-\kappa z} , & z > 0 \end{cases} \qquad (16a)$$

$$P_{z}(\vec{\kappa},z) = \begin{cases} A_2 e^{\kappa z} , & z < 0 \\ A_1 e^{\kappa z} + B_1 e^{-\kappa z} , & 0 \le z \le a , \\ B_2 e^{-\kappa z} , & z > a \end{cases} \qquad (16b)$$

where a is the thickness of material 1. When these solutions are substituted into (12), we find a set of homogeneous equations for the amplitudes A_ν and B_ν of the p-polarization. The condition that the determinant for the coefficients must not vanish leads to the dispersion relation

$$\frac{\epsilon_1(\omega) - \epsilon_2(\omega)}{\epsilon_1(\omega) + \epsilon_2(\omega)} = \pm e^{\kappa a} , \qquad (17)$$

where ϵ_1 and ϵ_2 are given by (9a). The + and - signs on the right-hand side of (17) correspond to the symmetric and antisymmetric modes of the interface phonons, respectively. Remembering (9a) and (17), it is then not difficult to find the explicit eigenvectors for the interface phonon modes from (16). The results are

$$\vec{\pi}_a = \begin{cases} C_a[\frac{\epsilon_2(\omega) - 1}{\epsilon_1(\omega) - 1}] e^{\kappa z} \sinh(\frac{\kappa a}{2})(-i, -1) , & z < 0 \\ C_a[i \sinh(\kappa(z-\frac{a}{2})), \cosh(\kappa(z-\frac{a}{2}))] , & 0 \le z \le a \\ C_a[\frac{\epsilon_2(\omega) - 1}{\epsilon_1(\omega) - 1}] e^{-\kappa(z-a)} \sinh(\frac{\kappa a}{2})(i, -1) , & z > a \end{cases} \qquad (18a)$$

for the antisymmetric modes, and

$$\vec{\pi}_s = \begin{cases} C_s\left[\frac{\epsilon_2(\omega) - 1}{\epsilon_1(\omega) - 1}\right] e^{\kappa z} \cosh(\frac{\kappa a}{2})(i,1) , & z< 0 \\ C_s[i \cosh(\kappa(z-a/2)), \sinh(\kappa(z-\frac{a}{2}))] , & 0 \leq z \leq a \\ C_s\left[\frac{\epsilon_2(\omega) - 1}{\epsilon_1(\omega) - 1}\right] e^{-\kappa(z-a)} \cosh(\frac{\kappa a}{2})(i,-1) , & z > a \end{cases} \qquad (18b)$$

for the symmetric modes. The normalization constants turn out to be equal to each other, namely,

$$C_a = C_s = \sqrt{\frac{\kappa}{\sinh(\kappa a)} \Big/ \left[\frac{\eta_1}{\omega_{p1}^2} - \frac{\eta_2}{\omega_{p2}^2} \frac{\epsilon_1}{\epsilon_2} \left(\frac{\chi_2}{\chi_1}\right)^2\right]} , \qquad (19)$$

where

$$\eta_\nu(\omega_i) = 1/[1 + \alpha_\nu \eta_\nu(\lambda_{o\nu} - \lambda_\nu)]^2 . \qquad (20)$$

The explicit dispersion relations follow directly from (9) and (17):

$$\omega_a^{\pm} = \Big\{\epsilon_{\infty 2}(\omega_{T1}^2+\omega_{L2}^2) + \epsilon_{\infty 1}(\omega_{T2}^2+\omega_{L1}^2) \coth(\frac{\kappa a}{2})$$

$$\pm \, (\epsilon_{\infty 2}^2(\omega_{T1}^2-\omega_{L2}^2)^2 + \epsilon_{\infty 1}^2(\omega_{T2}^2-\omega_{L1}^2)^2 \coth^2(\frac{\kappa a}{2})$$

$$+ 2\epsilon_{\infty 1}\epsilon_{\infty 2}[(\omega_{T1}^2+\omega_{L2}^2)(\omega_{T2}^2+\omega_{L1}^2)$$

$$- 2(\omega_{T2}^2\omega_{L1}^2+\omega_{L2}^2\omega_{T1}^2)\coth(\tfrac{\kappa a}{2})])^{\frac{1}{2}}\Big\}^{\frac{1}{2}} \{2[\epsilon_{\infty 2}+\epsilon_{\infty 1}\coth(\tfrac{\kappa a}{2})]\}^{-\frac{1}{2}} \tag{21a}$$

$$\omega_s^{\pm} = \Big\{\epsilon_{\infty 2}(\omega_{T1}^2+\omega_{L2}^2) + \epsilon_{\infty 1}(\omega_{T2}^2+\omega_{L1}^2)\tanh(\tfrac{\kappa a}{2})$$

$$\pm (\epsilon_{\infty 2}^2(\omega_{T1}^2-\omega_{L2}^2)^2 + \epsilon_{\infty 1}^2(\omega_{T2}^2-\omega_{L1}^2)^2\tanh^2(\tfrac{\kappa a}{2})$$

$$+ 2\epsilon_{\infty 1}\epsilon_{\infty 2}[(\omega_{T1}^2+\omega_{L2}^2)(\omega_{T2}^2+\omega_{L1}^2)$$

$$- 2(\omega_{T2}^2\omega_{L1}^2+\omega_{T1}^2\omega_{L2}^2)]\tanh(\tfrac{\kappa a}{2}))^{\frac{1}{2}}\Big\}^{\frac{1}{2}}\{2[\epsilon_{\infty 2}+\epsilon_{\infty 1}\tanh(\tfrac{\kappa a}{2})]\}^{-\frac{1}{2}} . \tag{21b}$$

Let us now look at the limiting cases. When $\kappa a \to \infty$, $\tanh(\frac{\kappa a}{2}) = 1$ and $\coth(\frac{\kappa a}{2}) = 1$. Therefore, both (21a) and (21b) approach the same limit which is identical to the result of a bilayer system with only one interface [7], as it should be. The limit $\kappa a \to 0$ yields, of course, the bulk material 2 with frequencies ω_{T2} and ω_{L2}. When $\kappa a \to 0$, $\tanh(\frac{\kappa a}{2}) = 0$ and $\coth(\frac{\kappa a}{2}) \to \infty$. We then find from (21) that

$$\omega_a^{\pm} = \sqrt{[\omega_{T2}^2+\omega_{L1}^2 \pm (\omega_{T2}^2-\omega_{L1}^2)]/2} = \omega_{T2},\omega_{L1} \tag{22a}$$

$$\omega_s^{\pm} = \sqrt{[\omega_{T1}^2+\omega_{L2}^2 \pm (\omega_{T1}^2-\omega_{L2}^2)]/2} = \omega_{T1},\omega_{L2} , \tag{22b}$$

Thus in the central region of the Brillouin zone, these modes have the same frequencies as those of the bulk LO and TO phonons in each material. For this reason, we shall refer to them as LO-like and TO-like interface phonons.

3. Computational Example

As an example, we take GaAs as material 1 and AℓAs as material 2 for our DHS. The dispersion relations calculated from (21) are plotted in Fig. 1.

Many experiments have been performed for this particular type of layered structure. In particular, an experiment of right-angle Raman scattering in a GaAs quantum well [5] reveals novel selection rules for polarization of the incident and scattered light. It is observed that the transverse vibration occurs at the bulk LO frequency of GaAs, while the longitudinal vibration occurs at the bulk TO frequency of GaAs. We show below that this surprising phenomenon can be understood in terms of Raman scattering from interface phonons.

In Raman scattering experiments, the phonon energy is given by the Raman shift, while its momentum is determined by the photon momentum transferred. Under ordinary conditions, the scattering is observed only near the Brillouin zone center. In fact, the experimental parameters involved in Ref. 3 imply κ ~ 0.1. Therefore, the polarization vector $\vec{\pi}_a$ is dominated by its z-component $P_z^a = C_a \cosh[x(z - a/2)]$ according to (18a), or the antisymmetric interface phonon is predominantly transverse. It follows then from (22a) that the transverse wave in the central layer oscillates at the LO frequency of bulk GaAs. Similarly, we find from (18b) and (22b) that the symmetric interface phonon is predominantly longitudinal and oscillates at the bulk TO frequency of GaAS. A detailed account of this and other experiments will be discussed in forthcoming publications.

In addition to the Raman scattering experiments mentioned above, cyclotron resonance measurements of the electron-phonon interaction have shown pinning phenomena at frequencies below ω_L. In the measurement of the 1s-2p transition energy of hydrogenic impurity confined in a quantum well [10], the pinning is observed at a frequency about 40 cm^{-1} below ω_L, which is 20 cm^{-1} below ω_T. This surprising result has not yet been accounted for theoretically, although the existence of propagating LO modes has been suggested as a possible origin of zone-folding effects [10]. Our preliminary results of interface phonon modes in a superlattice indicate that it is possible to reproduce experimental data with perhaps some modification of the Frölich coupling constant. More careful study is necessary, however, before any definite conclusion can be made.

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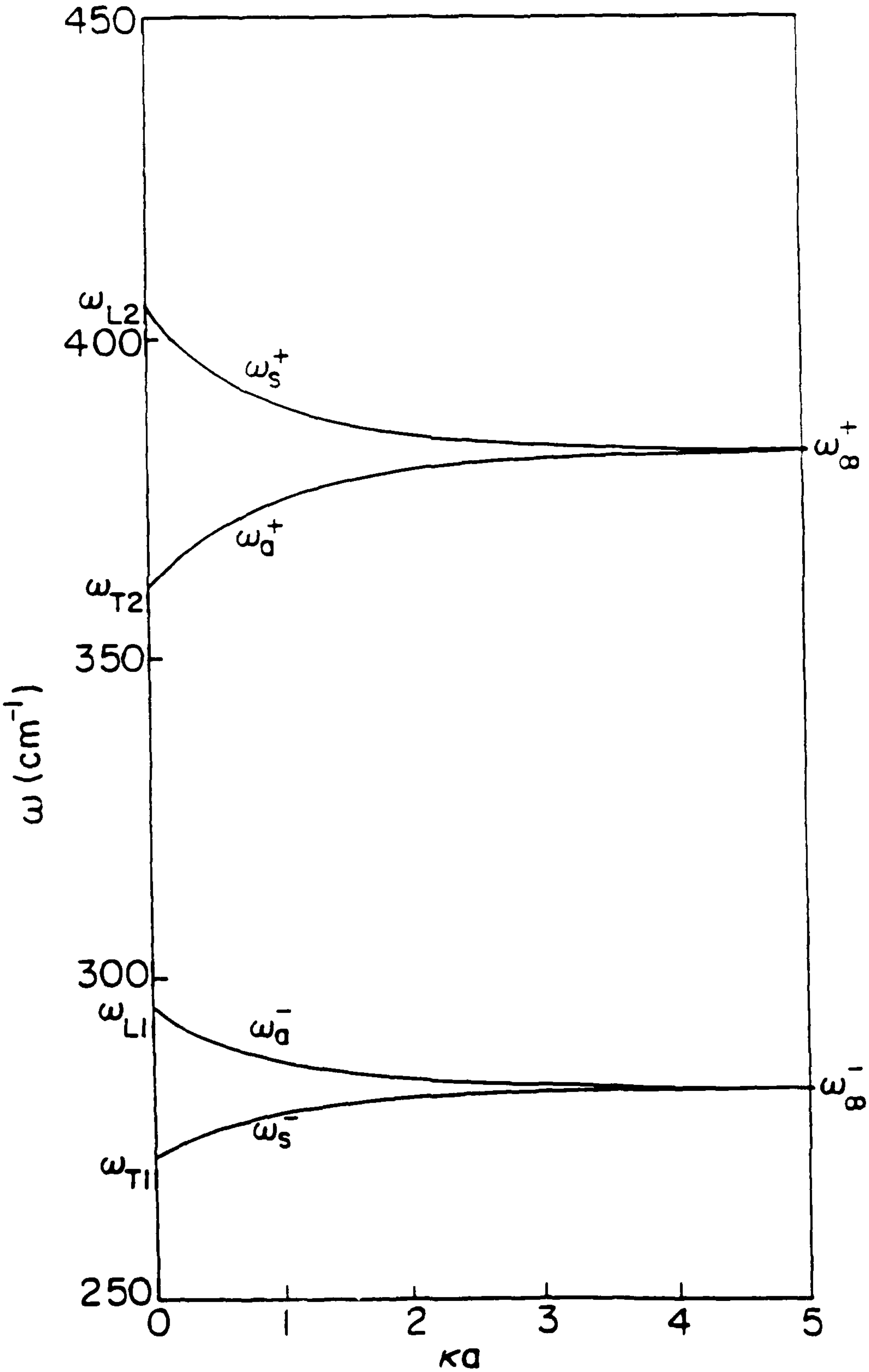
FIGURE CAPTION

1. Dispersion relations for the interface modes in GaAs/AℓAs double heterostructure for which $\omega_{L2} > \omega_{T2} > \omega_{L1} > \omega_{T1}$.

Fig. 1



450
400
350
300
250
ω_{L2}
ω_{T2}
ω_{L1}
ω_{T1}
ω_s^+
ω_a^+
ω_a^-
ω_s^-
ω_∞^+
ω_∞^-
$\omega\ (cm^{-1})$
0
1
2
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