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Optical Phonon Modes in a Double Heterostructure of Polar Crystals

by

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Optical phonon modes in a double heterostructure of polar crystals

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Abstract

The equation of motion for the polarization vector for a double heterostructure of polar crystals is solved exactly within the framework of the continuum model. There exist only two types of phonon modes, the interface modes and the confined bulk modes, whose eigenvectors are obtained explicitly. Dispersion relations are derived analytically for the interface modes, while the confined bulk modes are dispersionless, a fact consistent with the model. It is also found that in Raman scattering experiments, the symmetric interface modes are predominantly longitudinal (LO) and the antisymmetric interface modes transversal (TO). In the central region of the Brillouin zone, however, they both split into two branches oscillating at LO and TO frequencies, respectively. Possible reinterpretation of various experiments are briefly discussed.

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I. Introduction

There has been great interest in recent years in the study of various vibrational modes supported by semiconductor heterostructures. The patterns of normal modes of vibrations that determine the electronic properties in such structures are evidently different from those in the bulk. The presence of interfaces necessarily alters the phonon modes, and possibly even their interaction with electrons may be modified because of the reduced dimensionality.

Experimental investigations of the spectra of possible acoustic modes in superlattices¹⁻⁴ have shown fairly-good agreement with theory. On the other hand, the longitudinal optical (LO) modes in polar crystals are much less well understood in these contexts. In all the calculations such as free-carrier absorption of light,⁵⁻⁷ scattering rates⁸ and polaronic effects,⁹ etc., the usual bulk Frölich Hamiltonian is assumed for the electron-phonon interaction in confined systems. The only requirements are that the material elastic properties match at the interfaces and that the dielectric properties are equal. More recently, the surface optical (SO) mode has been included in the treatment of polarons confined in a slab¹⁰ or near the interface in semi-infinite systems.^{11,12} For these surface and interface situations, the bulk Frölich Hamiltonian is still employed for treating the LO-phonon contribution.

On the other hand, evidences of confined modes peculiar to different types of layered structures have been noticed in various experiments. Measurements of magneto-absorption and cyclotron resonance in GaInAs/InP superlattices¹³ and GaAs/GaAlAs heterostructures¹⁴ indicate that the electron-LO-phonon interaction in these structures can be fundamentally different from that in the bulk case. In a numerical study of possible

modes of the optical phonon in layered polar crystals, it is found that phonon modes tend to be confined in each layer and that the penetration of vibrations into the adjacent layer is negligible.^{15,16} Moreover, the existence of confined phonon modes has been directly observed in a GaAs single quantum well of GaAs/GaAlAs heterostructures.¹⁷

In theoretical investigations of the vibrational modes in an ionic slab, Fuchs and Kliever¹⁸ have found the bulk LO mode with the nodes at the surfaces as well as the SO modes of different symmetries. Interface modes have been derived by Wendler¹⁹ by considering the polarization field in a double layer system, and by Lassnig²⁰ using the energy-loss method in a double heterostructure (DHS) of polar semiconductors. An alternative treatment²¹ of the DHS predicts some peculiar phonon modes that have not been borne out by observation.

In this article we present solutions for optical phonon modes in a semiconductor DHS using the continuum model of Born and Huang.²² The method of solution has been developed by various authors.^{18,19,23} Apart from the long-wavelength limit in the model, no further approximation is made throughout our calculation. Dispersion relations and eigenvectors for all the normal modes of lattice vibration are derived analytically. It is found that there exist two types of phonon modes, the interface modes and confined bulk modes.

While the existence of interface modes has been well recognized experimentally,^{24,25} their eigenvectors and dispersion relations in a DHS are solved explicitly for the first time in this paper. Our results show that either the symmetric or the antisymmetric interface modes has two branches. Their frequencies at the center of the Brillouin zone are exactly the same as those of the bulk LO and TO phonon in each material.

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Experimental evidence of confined bulk modes has recently been reported.^{17 25,26} We find that both the bulk LO and transverse optical (TO) modes are strictly confined. Further investigation on implications of such confinements is being carried out and will be reported elsewhere.

In Sec. II, we outline the procedure for deriving the equation of motion for the polarization vector. The coupled integral equations are solved for the interface modes in Sec. III and the confined bulk modes in Sec. IV. Consequences and implications of our results are discussed in Sec. V.

II. Equation of motion of the polarization field

Consider a DHS of two different polar crystals as shown in Fig. 1. A layer of material 1 with thickness a is sandwiched between two thick layers of material 2. We take the z -axis to be perpendicular to the interfaces which are located at $z = 0$ and $z = a$, respectively. Following Born and Huang,²² we start in the continuum approximation with the equation of motion for the relative displacement $u(\vec{r}, t)$ of the ion pair in material ν ($\nu = 1, 2$),

$$\mu_{\nu} \ddot{\vec{u}}_{\nu}(\vec{r}, t) = -\mu_{\nu} \omega_{0\nu}^2 \vec{u}_{\nu}(\vec{r}, t) + e^* \vec{E}(\vec{r}, t) \quad , \quad (1)$$

where μ is the reduced mass of the pair of ions, $\mu \omega_0^2$ is the short-range force constant not including Coulomb fields, $\vec{E}(\vec{r}, t)$ is the local electric field, and e^* is the effective charge of the ions. The subscript ν labels the material considered. The oscillating ions produce a polarization field $\vec{P}(\vec{r}, t)$ given by

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

where n is the number of ion pairs per unit cell and α is the polarizability. The first term in (2) represents the contribution of the ion pair when the lattice vibrates, and the second term is the electronic polarization of the ions due to the electric field associated with the optical modes. The part of the polarization produced by the electron itself as it moves through the crystal is, however, not included in our consideration, since the continuum model is not valid for such an effect.²³ The local field in (2) is related, in the long-wavelength limit, to the polarization by

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

where

$$\vec{E}_l(\vec{r}, t) = \frac{4\pi}{3} \vec{P}(\vec{r}, t) \quad , \quad (3b)$$

and Γ denotes the Green tensor with components

$$\Gamma_{\alpha\beta} = \frac{1}{4\pi} \frac{\partial^2}{\partial x_{\alpha} \partial x_{\beta}} \frac{1}{|\vec{r} - \vec{r}'|} \quad . \quad (3c)$$

The equation of motion for the polarization then follows by plugging Eqs. (2) and (3) into (1):

$$\begin{aligned}
& \left[1 - \frac{4\pi}{3} \alpha_{\nu} n_{\nu} \right] \ddot{\vec{P}}(\vec{r}, t) + \left[\omega_{o\nu}^2 - 4\pi(\alpha_{\nu} n_{\nu} \omega_{o\nu}^2 + \frac{n_{\nu} e_{\nu}^{*2}}{3\mu_{\nu}}) \right] \vec{P}(\vec{r}, t) \\
& = 4\pi \alpha_{\nu} n_{\nu} \int d\vec{r}' \Gamma(\vec{r} - \vec{r}') \cdot \ddot{\vec{P}}(\vec{r}', t) \\
& + 4\pi(\alpha_{\nu} n_{\nu} \omega_{o\nu}^2 + \frac{n_{\nu} e_{\nu}^{*2}}{3\mu_{\nu}}) \int d\vec{r}' \Gamma(\vec{r} - \vec{r}') \cdot \vec{P}(\vec{r}', t) \quad (4)
\end{aligned}$$

The time-dependent part of the polarization can be separated by assuming $\vec{P}(\vec{r}, t) = \vec{P}(\vec{r}) e^{i\omega t}$ which, after substituting into (4), yields the equation for $\vec{P}(\vec{r})$,

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

where we have defined the parameters

$$\lambda_{\nu}^2 = 4\pi\omega^2 / \omega_{p\nu}^2 \quad (5b)$$

$$\lambda_{o\nu}^2 = 4\pi\omega_{o\nu}^2 / \omega_{p\nu}^2 \quad (5c)$$

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

Since the translational invariance in the z-direction is destroyed by the interfaces, we introduce the two-dimensional vectors $\vec{\kappa}$ and $\vec{\rho}$ so that $\vec{k} = (\vec{\kappa}, q)$ and $\vec{r} = (\vec{\rho}, z)$. The two-dimensional Fourier transforms can now be written as

$$\vec{P}(\vec{r}) = \left(\frac{1}{2\pi}\right)^2 \int_{-\infty}^{\infty} d\vec{\kappa} e^{i\vec{\kappa} \cdot \vec{\rho}} \vec{P}(\vec{\kappa}, z) \quad (6)$$

$$\frac{1}{r} = \frac{1}{2\pi} \int \frac{d\vec{\kappa}}{\kappa} \exp(i\vec{\kappa} \cdot \vec{\rho} - \kappa|z|) \quad (7)$$

Differentiating (7) twice with respect to the coordinates, we obtain

$$\Gamma = -\frac{1}{4\pi} \int d\vec{\kappa} e^{i\vec{\kappa} \cdot \vec{\rho} - \kappa|z|} \vec{K}\vec{K}/2\pi\kappa \quad (8)$$

where

$$\vec{K} = (\vec{\kappa}, i\theta(z)\kappa) \quad (9)$$

with the step function $\theta(z) = \pm 1$ for $z \gtrless 0$. Substituting (6) and (8) into (5a), and moving the term $-P_z$ from the right-hand side to the left, we can write after some algebra the resulting equation in a more symmetric form as

$$4\pi \begin{bmatrix} \chi_{\nu}^{-1}(\omega) & 0 & 0 \\ 0 & \chi_{\nu}^{-1}(\omega) & 0 \\ 0 & 0 & \chi_{\nu}^{-1}(\omega)\epsilon_{\nu}(\omega) \end{bmatrix} \cdot \vec{P}(\vec{\kappa}, z) = \frac{2\pi}{\kappa} \int_{-\infty}^{\infty} dz' e^{-\kappa(z-z')} \vec{K}\vec{K} \cdot \vec{P}(\vec{\kappa}, z') \quad (10)$$

where $\chi_{\nu}(\omega)$ is defined by

$$4\pi\chi_{\nu}^{-1}(\omega) = \frac{\lambda_{\nu} - \lambda_{0\nu}}{\alpha_{\nu} n_{\nu} (\lambda_{\nu} - \lambda_{0\nu}) - 1} - \frac{4\pi}{3} \quad (11)$$

It turns out that $\chi_\nu(\omega)$ is the isotropic dielectric susceptibility and is related to the dielectric function by $\chi_\nu(\omega) = \epsilon_\nu(\omega) - 1$ with

$$\epsilon_\nu(\omega) = \epsilon_{\infty\nu} \frac{\omega_{L\nu}^2 - \omega^2}{\omega_{T\nu}^2 - \omega^2} \quad (12a)$$

$$\epsilon_{\infty\nu} = 1 + 4\pi\alpha_\nu n_\nu / (1 - \frac{4\pi}{3} \alpha_\nu n_\nu) \quad (12b)$$

where we have defined the LO and TO phonon frequencies

$$\omega_{L\nu}^2 = \omega_{\text{ov}}^2 + \frac{2}{3} \omega_{p\nu}^2 / (1 + \frac{8\pi}{3} \alpha_\nu n_\nu) \quad (13a)$$

$$\omega_{T\nu}^2 = \omega_{\text{ov}}^2 - \frac{1}{3} \omega_{p\nu}^2 / (1 - \frac{4\pi}{3} \alpha_\nu n_\nu) \quad (13b)$$

Since the interface phonons propagate in the xy-plane, it is more convenient to express the polarization vector as $\vec{P} = (\vec{\pi}, P_s)$ where $\vec{\pi}$ is a two-dimensional vector defined by $\vec{\pi} = (P_\kappa, P_z)$. Thus

$$\vec{P}(\vec{\kappa}, z) = P_\kappa(\vec{\kappa}, z) \hat{\kappa} + P_z(\vec{\kappa}, z) \hat{z} + P_s(\vec{\kappa}, z) \hat{s} \quad (14)$$

where the unit vector $\hat{s}$ is defined by $\hat{s} = \hat{z} \times \hat{\kappa}$. Substituting (14) in (10), we can separate the s-component and decouple (10) into two equations:

$$\begin{bmatrix} \chi_\nu^{-1}(\omega) & 0 \\ 0 & \chi_\nu^{-1}(\omega) \epsilon_\nu(\omega) \end{bmatrix} \cdot \vec{\pi}(\vec{\kappa}, z) = \frac{1}{4\pi} \int_{-\infty}^{\infty} dz' M(z-z') \cdot \vec{\pi}(\vec{\kappa}, z') \quad (15a)$$

for the s -called p-polarization, and

$$\chi_{\nu}^{-1}(\omega) P_s(\vec{\kappa}, z) = 0 \quad (15b)$$

for the s-polarization, where M is a Hermitian matrix given by

$$M(z-z') = M^{\dagger}(z'-z) = -2\pi\kappa e^{-\kappa|z-z'|} \begin{bmatrix} 1 & i\theta(z-z') \\ i\theta(z-z') & -1 \end{bmatrix} \quad (16)$$

As we shall show in the following sections, Eq. (15a) defines an eigenvalue problem whose solutions describe the interface modes and the confined modes. Equation (15b) describes the s-polarization, which is not of concern in the present paper. The eigenvectors $\vec{\pi}(\kappa, z)$ form a complete orthonormal set. Here we just give without proof the orthonormality relation

$$\int_{-\infty}^{\infty} dz \frac{\eta_{\nu}^4(\omega_i) \eta_{\nu}^4(\omega_j)}{\omega_{p\nu}^2} \vec{\pi}_j^*(\vec{\kappa}, z) \vec{\pi}_i(\vec{\kappa}, z) = \delta_{ij} \quad (17)$$

with $\eta_{\nu}^4(\omega_i) = 1/[1 + \alpha_{\nu} n_{\nu}(\lambda_{o\nu} - \lambda_{\nu})]$. The completeness relation is given by

$$\sum_i \vec{\pi}_i^*(\kappa, z) \vec{\pi}_i(\kappa, z') = \frac{\omega_{p\nu}^2}{\eta_{\nu}^2} I \delta(z-z') \quad (18)$$

for the so-called p-polarization, and

$$\chi_{\nu}^{-1}(\omega) P_s(\vec{\kappa}, z) = 0 \quad (15b)$$

for the s-polarization, where M is a Hermitian matrix given by

$$M(z-z') = M^{\dagger}(z'-z) = -2\pi\kappa e^{-\kappa|z-z'|} \begin{bmatrix} 1 & i\theta(z-z') \\ i\theta(z-z') & -1 \end{bmatrix} \quad (16)$$

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$$\int_{-\infty}^{\infty} dz \frac{\eta_{\nu}^{\frac{1}{2}}(\omega_i) \eta_{\nu}^{\frac{1}{2}}(\omega_j)}{\omega_{p\nu}^2} \vec{\pi}_j^*(\vec{\kappa}, z) \vec{\pi}_i(\vec{\kappa}, z) = \delta_{ij} \quad (17)$$

with $\eta_{\nu}^{\frac{1}{2}}(\omega_i) = 1/[1 + \alpha_{\nu} n_{\nu}(\lambda_{o\nu} - \lambda_{\nu})]$. The completeness relation is given by

$$\sum_i \vec{\pi}_i^*(\kappa, z) \vec{\pi}_i(\kappa, z') = \frac{\omega_{p\nu}^2}{\eta_{\nu}} I \delta(z-z') \quad (18)$$

where I stands for the unit matrix. Finally, we note that the polarization vector must be real, and consequently

$$P(\vec{\kappa}, z) = P^*(-\vec{\kappa}, z) \quad (19)$$

Similarly, we have

$$\vec{\pi}(\vec{\kappa}, z) = \begin{bmatrix} -1 & 0 \\ 0 & 1 \end{bmatrix} \vec{\pi}^*(-\vec{\kappa}, z) \quad (19b)$$

III. Interface Modes

It is easier to solve the coupled integral equations (15a) by first transforming them into differential equations. This can be done by differentiating (15a) with respect to z twice and at the same time requiring

$$\det \begin{bmatrix} \chi_{\nu}^{-1}(\omega) & 0 \\ 0 & \chi_{\nu}^{-1}(\omega) \epsilon_{\nu}(\omega) \end{bmatrix} \neq 0 \quad (20)$$

The resulting equations are

$$\frac{d}{dz} P_{\kappa}(\vec{\kappa}, z) = i\kappa P_z(\vec{\kappa}, z) \quad (21a)$$

$$\frac{d^2}{dz^2} \vec{\pi}(\vec{\kappa}, z) = \kappa^2 \vec{\pi}(\vec{\kappa}, z) \quad (21b)$$

The solutions to (21) take the form

$$P_{\kappa}(\vec{\kappa}, z) = \begin{cases} iA_2 e^{\kappa z} & , \quad z < 0 \\ i(A_1 e^{\kappa z} - B_1 e^{-\kappa z}) & , \quad 0 \leq z \leq a \\ -iB_2 e^{-\kappa z} & , \quad z > a \end{cases} \quad (22a)$$

$$P_z(\vec{\kappa}, z) = \begin{cases} A_2 e^{\kappa z} & , \quad z < 0 \\ A_1 e^{\kappa z} + B_1 e^{-\kappa z} & , \quad 0 \leq z \leq a \\ B_2 e^{-\kappa z} & , \quad z > a \end{cases} \quad (22b)$$

Substituting (22) in the integral equation (15a), one obtains a set of homogeneous equations for the amplitudes A_{ν} and B_{ν} of the p-polarization. The condition for the existence of a nontrivial solution then leads to the dispersion relation

$$\frac{\epsilon_1(\omega) - \epsilon_2(\omega)}{\epsilon_1(\omega) + \epsilon_2(\omega)} = \pm e^{\kappa a} \quad (23)$$

The + and - signs on the right-hand side of (23) correspond to the symmetric and antisymmetric modes of the interface phonons, respectively. The polarization amplitudes are found to satisfy the following relations:

$$B_1^{\pm}/A_1^{\pm} = B_2^{\pm}/A_2^{\pm} = \mp e^{\kappa a} \quad (24a)$$

$$A_2^{\pm} = \frac{\chi_2(\omega)}{\chi_1(\omega)} (1 \pm e^{\kappa a}) A_1^{\pm} \quad (24b)$$

$$B_2^\pm = \frac{\chi_2(\omega)}{\chi_1(\omega)} (1 \pm e^{\kappa a}) B_1^\pm \quad (24c)$$

Equations (24) are equivalent to the boundary conditions that the wave functions have to satisfy at the interfaces. Thus the differential equations (21) yield naturally the correct boundary conditions after substituting their solutions (22) into the integral equation (15a). On the contrary, the hydrodynamic terms introduced in Ref. 23 are inconsistent with the boundary conditions. Combining Eqs. (22)-(24), we find the eigenvectors for the antisymmetric interface phonon modes to be

$$\vec{\pi}_a = \begin{cases} C_a \left[\frac{\epsilon_2(\omega)-1}{\epsilon_1(\omega)-1} \right] e^{\kappa z} (-i, -1) \sinh\left(\frac{\kappa a}{2}\right) & , \quad z < 0 \\ C_a [i \sinh(\kappa(z-\frac{a}{2})), \cosh(\kappa(z-a/2))] & , \quad 0 \leq z \leq a \\ C_a \left[\frac{\epsilon_2(\omega)-1}{\epsilon_1(\omega)-1} \right] e^{-\kappa(z-a)} (i, -1) \sinh\left(\frac{\kappa a}{2}\right) & , \quad z > a \end{cases} \quad (25a)$$

and for the symmetric modes to be

$$\vec{\pi}_s = \begin{cases} C_s \left[\frac{\epsilon_2(\omega)-1}{\epsilon_1(\omega)-1} \right] e^{\kappa z} (i, 1) \cosh\left(\frac{\kappa a}{2}\right) & , \quad z < 0 \\ C_s [i \cosh(\kappa(z-a/2)), \sinh(\kappa(z-a/2))] & , \quad 0 \leq z \leq a \\ C_s \left[\frac{\epsilon_2(\omega)-1}{\epsilon_1(\omega)-1} \right] e^{-\kappa(z-a)} (i, -1) \cosh\left(\frac{\kappa a}{2}\right) & , \quad z > a \end{cases} \quad (25b)$$

where, according to (17), the normalization constants are given by

$$C_{a,s} = \sqrt{\frac{\kappa}{\sinh(\kappa a)} \left[\frac{\eta_1}{\omega_{p1}^2} - \frac{\eta_2}{\omega_{p2}^2} \frac{\epsilon_1}{\epsilon_2} \left(\frac{\chi_2}{\chi_1} \right)^2 \right]} \quad (25c)$$

A heterostructure composed of two media with dielectric functions given by (12) always supports four distinct interface modes of vibrations, two from each medium. The dispersion relations for these modes can then be calculated explicitly from (12) and (23). The results are

$$\begin{aligned} \omega_a^{\pm} = & \left\{ \epsilon_{\infty 2} (\omega_{T1}^2 + \omega_{L2}^2) + \epsilon_{\infty 1} (\omega_{T2}^2 + \omega_{L1}^2) \coth\left(\frac{\kappa a}{2}\right) \right. \\ & \pm \left(\epsilon_{\infty 2}^2 (\omega_{T1}^2 - \omega_{L2}^2)^2 + \epsilon_{\infty 1}^2 (\omega_{T2}^2 - \omega_{L1}^2)^2 \coth^2\left(\frac{\kappa a}{2}\right) \right. \\ & + 2\epsilon_{\infty 1} \epsilon_{\infty 2} [(\omega_{T1}^2 + \omega_{L2}^2)(\omega_{T2}^2 + \omega_{L1}^2) \\ & \left. \left. - 2(\omega_{T2}^2 \omega_{L1}^2 + \omega_{L2}^2 \omega_{T1}^2)] \coth\left(\frac{\kappa a}{2}\right) \right]^{1/2} \right\}^{1/2} / (2[\epsilon_{\infty 2} + \epsilon_{\infty 1} \coth\left(\frac{\kappa a}{2}\right)])^{1/2} \quad (26a) \end{aligned}$$

$$\begin{aligned} \omega_s^{\pm} = & \left\{ \epsilon_{\infty 2} (\omega_{T1}^2 + \omega_{L2}^2) + \epsilon_{\infty 1} (\omega_{T2}^2 + \omega_{L1}^2) \tanh\left(\frac{\kappa a}{2}\right) \right. \\ & \pm \left(\epsilon_{\infty 2}^2 (\omega_{T1}^2 - \omega_{L2}^2)^2 + \epsilon_{\infty 1}^2 (\omega_{T2}^2 - \omega_{L1}^2)^2 \tanh^2\left(\frac{\kappa a}{2}\right) \right. \\ & + 2\epsilon_{\infty 1} \epsilon_{\infty 2} [(\omega_{T1}^2 + \omega_{L2}^2)(\omega_{T2}^2 + \omega_{L1}^2) \\ & \left. \left. - 2(\omega_{T2}^2 \omega_{L1}^2 + \omega_{L2}^2 \omega_{T1}^2)] \tanh\left(\frac{\kappa a}{2}\right) \right]^{1/2} \right\}^{1/2} / (2[\epsilon_{\infty 2} + \epsilon_{\infty 1} \tanh\left(\frac{\kappa a}{2}\right)])^{1/2} \quad (26b) \end{aligned}$$

It is seen from (26) that the interface phonon energies depend explicitly on the dimensionless quantity κa .

Let us now look at the limiting cases. When $a \rightarrow \infty$, $\tanh(\frac{\kappa a}{2}) = 1$ and $\coth(\frac{\kappa a}{2}) = 1$. Therefore, both (26a) and (26b) approach the same limit, given by

$$\begin{aligned}
\omega_{\infty}^{\pm} = & \left\{ \epsilon_{\infty 1} (\omega_{L1}^2 + \omega_{T2}^2) + \epsilon_{\infty 2} (\omega_{T1}^2 + \omega_{L2}^2) \right. \\
& \pm \left[\epsilon_{\infty 1}^2 (\omega_{L1}^2 - \omega_{T2}^2)^2 + \epsilon_{\infty 2}^2 (\omega_{L2}^2 - \omega_{T1}^2)^2 \right. \\
& \left. \left. + 2 \epsilon_{\infty 1} \epsilon_{\infty 2} [(\omega_{T1}^2 + \omega_{L2}^2)(\omega_{T2}^2 + \omega_{L1}^2) - 2(\omega_{T2}^2 \omega_{L1}^2 + \omega_{T1}^2 \omega_{L2}^2)] \right]^{1/2} \right\}^{1/2} \\
& \times [2(\epsilon_{\infty 1} + \epsilon_{\infty 2})]^{-1/2}, \quad (27)
\end{aligned}$$

which is identical to the result of a bilayer system with only one interface,¹⁹ as it should. In the limit $a \rightarrow 0$, the system reduces to a bulk material 2 with frequencies ω_{L2} and ω_{T2} . When $\kappa \rightarrow 0$, $\tanh(\frac{\kappa a}{2}) = 0$ and $\coth(\frac{\kappa a}{2}) \rightarrow \infty$. We then find from (26) that

$$\omega_a^{\pm} = \sqrt{[\omega_{T2}^2 + \omega_{L1}^2 \pm (\omega_{T2}^2 - \omega_{L1}^2)]/2} = \omega_{T2}, \omega_{L1} \quad (28a)$$

$$\omega_s^{\pm} = \sqrt{[\omega_{T1}^2 + \omega_{L2}^2 \pm (\omega_{T1}^2 - \omega_{L2}^2)]/2} = \omega_{T1}, \omega_{L2} \quad (28b)$$

that is, the limiting frequencies are given by the bulk LO and TO frequencies of the two materials. It may be worth mentioning yet another limit at this point. When the characteristic parameters of the two dielectrics approach each other, or when $\epsilon_1 \rightarrow \epsilon_2$, we find for a given width that the amplitudes of the interface modes of vibration diminish continuously and become zero at $\epsilon_1 = \epsilon_2$.

The interface phonon modes have no connection with bulk polarization charges because $\vec{\nabla} \cdot \vec{P} = 0$. They are accompanied by the surface charges σ at the interfaces. These charge densities can easily be determined by

calculating the difference of the polarization eigenvectors in the z -direction on both sides of the interface concerned. Thus we find from the z -components of $\vec{\pi}_{s,a}$ in (25)

$$\sigma_s = \begin{cases} |\sigma_s| & z = 0 \\ |\sigma_s| & z = a \end{cases} \quad \text{at} \quad (29a)$$

for the symmetric mode and

$$\sigma_a = \begin{cases} -|\sigma_a| & z = 0 \\ |\sigma_a| & z = a \end{cases} \quad \text{at} \quad (29b)$$

for the antisymmetric mode, where

$$|\sigma_s| = \left(\frac{\kappa}{\sinh(\kappa a)} \right)^{1/2} \frac{x_1 \sinh(\frac{\kappa a}{2}) + x_2 \cosh(\frac{\kappa a}{2})}{\left[\frac{x_1^2 \eta_1}{\omega_{p1}^2} + \frac{x_2^2 \eta_2}{\omega_{p1}^2} \coth(\frac{\kappa a}{2}) \right]^{1/2}} \quad (30a)$$

$$|\sigma_a| = \left(\frac{\kappa}{\sinh(\kappa a)} \right)^{1/2} \frac{x_1 \cosh(\frac{\kappa a}{2}) + x_2 \sinh(\frac{\kappa a}{2})}{\left[\frac{x_1^2 \eta_1}{\omega_{p1}^2} + \frac{x_2^2 \eta_2}{\omega_{p2}^2} \tanh(\frac{\kappa a}{2}) \right]^{1/2}} \quad (30b)$$

IV. Confined bulk LO and TO modes

For the LO modes, $\omega = \omega_{L\nu}$, and from (12a) we have $\epsilon_\nu(\omega_{L\nu}) = 0$ and $\epsilon_{\nu'}(\omega_{L\nu}) \neq 0$ for $\nu' \neq \nu$. Thus $\vec{D} = 0$ in layer ν , and $\vec{E} = -\vec{P}/\epsilon_0$. The differential equations satisfied by the polarization field associated with the longitudinal phonon modes follow from (15a). In layer ν we have

$$\frac{d}{dz} P_{\kappa}^L(\kappa, z) = i\kappa P_z^L(\kappa, z) \quad , \quad (31a)$$

and in layer $\nu' \neq \nu$

$$\frac{d^2}{dz^2} P_{\kappa}^L(\kappa, z) = \kappa^2 P_z^L(\kappa, z) \quad . \quad (31b)$$

The solution to (31b) has the same form as (22) with the coefficients determined by the boundary conditions for the field vectors. In layer ν , $\vec{D} = 0$, and $\vec{E} = -\vec{P}/\epsilon_0$. From the continuity of $\vec{D}$ across the interface, we have $\vec{D} = 0$ in layer ν' . Since $\epsilon_{\nu'} \neq 0$, we must have $\vec{E} = 0$ and $\vec{P} = 0$. Therefore (31b) has a solution that is identically zero everywhere in the layer ν' . The boundary conditions in layer ν can be found from (15a) for $\epsilon_{\nu} = 0$ and $\chi_n = -1$. After some algebra, we find from the coupled equations that

$$\vec{P}^L(a) = \vec{P}^L(0) = 0 \quad . \quad (32)$$

Hence, Eq. (31a) is satisfied in layer 1 by the eigenvectors

$$\vec{\pi}_m^L = \begin{cases} 0 & z < 0 \\ C_m^L [i \sin(\frac{m\pi}{a}z), \frac{m\pi}{a\kappa} \cos(\frac{m\pi}{a}z)] & 0 \leq z \leq a \\ 0 & z > a \end{cases} \quad , \quad (33a)$$

where m is an integer, and in layer 2 by

$$\vec{\pi}_q^L = \begin{cases} C_q^L [i \sin(qz), \frac{q}{\kappa} \cos(qz)] & , \quad z < 0 \\ 0 & 0 \leq z \leq a \\ C_q^L [i \sin q(z-a), \frac{q}{\kappa} \cos q(z-a)] & , \quad z > a \end{cases} \quad (33b)$$

where q is real, with the corresponding eigenfrequencies ω_{L1} and ω_{L2} , respectively. These modes are highly degenerate vibrations. We remark that the dispersionless nature of the confined bulk modes is consistent with the long-wavelength limit which is implied in the continuum model. The bulk optical photons have constant energy as $k \sim 0$.

Equations (33) clearly show that the longitudinal optical phonons are completely confined by the interfaces. In the central layer (labeled 1), the confinement leads to the quantization $q = m\pi/a$, where $m = 0, \pm 1, \pm 2, \dots$, while in the semi-infinite side layers (labeled 2), the wave number q remains continuous. The state vectors are normalized according to (17) with the normalization constants given by

$$C_m^L = \frac{\omega_{L1}}{\sqrt{\eta_1}} \sqrt{\frac{2}{a}} \frac{\kappa}{\sqrt{\kappa^2 + m^2 \pi^2 / a^2}} \quad (34a)$$

$$C_q^L = \frac{\omega_{L2}}{\sqrt{\eta_2}} \sqrt{\frac{1}{\pi}} \frac{\kappa}{\sqrt{\kappa^2 + q^2}} \quad (34b)$$

The confined LO phonons are related to both bulk polarization charges and interface polarization charges. The former can be found from $\rho = \vec{\nabla} \cdot \vec{P}$ and the latter follows from the boundary conditions at the interfaces. The results are

$$\rho_q^L = C_q (\kappa + q^2/\kappa) \sin(qz) \quad (35b)$$

$$\sigma_m^L = C_m \frac{m\pi}{a\kappa} \begin{cases} (-1) & \text{at } z = 0 \\ (-1)^m & \text{at } z = a \end{cases} \quad (36a)$$

$$\sigma_q^L = \begin{cases} C_q \frac{q}{\kappa} & z = 0 \\ -C_q \frac{q}{\kappa} & z = a \end{cases} \quad \text{at} \quad (36b)$$

We now turn our attention to the TO phonons for which $\omega = \omega_{T\nu}$. Equation (12a) then implies that $\chi_{\nu}^{-1}(\omega_{T\nu}) = 0$ and $\chi_{\nu'}^{-1}(\omega_{T\nu}) \neq 0$ for $\nu' \neq \nu$. Hence $\vec{E} = 0$ and $\vec{D} = \vec{P}$ in layer ν . The same consideration and procedures as described above for the LO phonons lead to eigenfrequencies ω_{T1} and ω_{T2} with corresponding eigenvectors

$$\vec{\pi}_m^T = \begin{cases} 0 & z < 0 \\ C_m^T [i \frac{m\pi}{a\kappa} \cos(\frac{m\pi}{a}z), \sin(\frac{m\pi}{a}z)] & 0 \leq z \leq a \\ 0 & z > a \end{cases} \quad (37a)$$

where m is an integer, and

$$\vec{\pi}_q^T = \begin{cases} C_q^T [i \frac{q}{\kappa} \cos(qz), \sin(qz)] & z < 0 \\ 0 & 0 \leq z \leq a \\ C_q^T [i \frac{q}{\kappa} \cos q(z-a), \sin q(z-a)] & z > a \end{cases} \quad (37b)$$

where q is a real number. We see from (37) that the TO phonons are also strictly confined by the presence of interfaces. Once more, these

eigenvectors are normalized according to (17), and the normalization constants are given by the same expressions as (34) except for the replacement of $\omega_{L\nu}$ by $\omega_{T\nu}$. The confined TO modes are, however, not associated with any polarization charge, neither bulk nor surface charge.

The s-polarization modes are given by the solution of (15b) with $\chi_{\nu}^{-1}(\omega) = 0$. This implies that the s-polarization modes exist only when the eigenfrequencies are those of the transverse optical phonons in either medium. Since these modes are completely decoupled from the other vibrational modes, they are not involved in the interaction with electrons and hence will not be discussed further.

V. Discussion

We have shown that there exist two types of phonon modes in a double heterostructure consisting of two semiconducting materials, the interface phonons and the confined bulk phonons. The interface modes may be either symmetric or antisymmetric with respect to the center of the system. They are dispersive in nature, and their frequencies for given materials depend solely upon the dimensionless quantity κa . In the center 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.

Since the bulk frequencies are determined by the positions of the zeroes and poles of the dielectric functions as can be seen from (12), different compositions of the double heterostructure can result in different frequency combinations. However, only three distinct combinations as shown in Fig. 2 are possible, where we have assumed $\omega_{T2} > \omega_{T1}$ without loss of generality. It is observed that as the width of the central layer

increases, the four interface modes become two degenerate modes. In the limit of large a , these modes have the same frequencies as those in a bilayer heterostructure. In case (a), the degenerate modes are material-like, while in the other two cases they are LO-like and TO-like. Experimentally, only case (a) has been observed thus far. It is therefore interesting to carry out experiments on samples with $\omega_{L1}, \omega_{L2} > \omega_{T1}, \omega_{T2}$, such as GaAs/Ga_{0.3}Al_{0.7}As (GaAs-type) and InP/AlSb.

It should also be of great interest to note that the peculiar mode observed in the 90° Raman scattering experiment²⁷ may be understood, as has been pointed out in a recent communication,²⁸ in terms of the interface modes derived in Sec. III. In other words, the novel slab modes reported in Ref. 27 are in fact the interface modes. A detailed analysis of this experiment will be published elsewhere, and here we give only a qualitative account. Since Raman scattering experiments involve only phonons of very small κ , the dominant component of the polarization vector $\vec{\pi}_a$ is P_z according to (25a). Therefore the antisymmetric interface modes are predominantly TO modes. In the central layer, this TO mode oscillates at the LO frequency of GaAs, in agreement with the experimental result in the right-angle scattering configuration. Similarly, (25b) shows that the polarization $\vec{\pi}_s$ has a dominant P_κ component, or the symmetric interface mode in the central layer is predominantly longitudinal and oscillates at the bulk TO frequency of GaAs.

In addition to the Raman scattering experiments, the interesting pinning phenomenon has been reported in recent measurements of cyclotron resonance. That the electron interacts with optical phonons at the bulk TO frequency has been observed in the measurements of the magnetopolaron frequency in semiconductor quantum wells,²⁹⁻³¹ and it has been attributed to

the classical dielectric effect.²⁹ This is essentially a polariton effect rather than a polaronic one. When the 1s-2p transition energy of a hydrogenic impurity atom in a GaAs quantum well is measured in strong magnetic fields,³¹ the pinning is found at a frequency about 40 cm^{-1} below ω_L ($\sim 20 \text{ cm}^{-1}$ below ω_T). To our knowledge, there is no theory up to the present time that can account for this result.³² The existence of traveling LO phonons and the zone-folding effect has been suggested as a possible source of this phenomenon.³¹ We have solved the interface phonon modes in a superlattice, and our preliminary results indicate that probably the interface modes are responsible for this strange pinning phenomenon. More careful study is necessary, however, before any definite conclusion can be made. Work along this direction is also underway and will be discussed in forthcoming publications.

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Figure Captions

1. Geometry of the double heterostructure.
2. Dispersion relations of the interface modes in the double heterostructure for different compositions: (a) GaAs/AlAs for which $\omega_{L2} > \omega_{T2} > \omega_{L1} > \omega_{T1}$, (b) GaAs/Al_{0.3}Ga_{0.7}As (GaAs-type) for which $\omega_{L2} > \omega_{L1} > \omega_{T2} > \omega_{T1}$, (c) InP/AlSb for which $\omega_{L1} > \omega_{L2} > \omega_{T2} > \omega_{T1}$.

Fig. 1

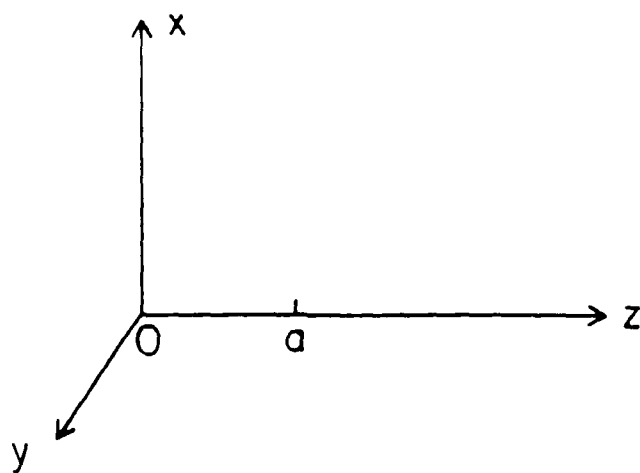
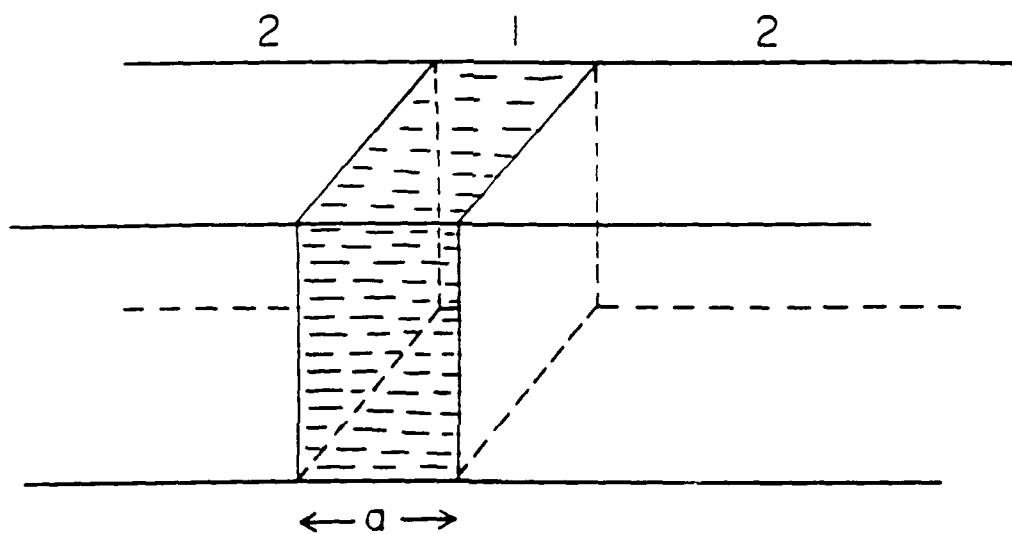
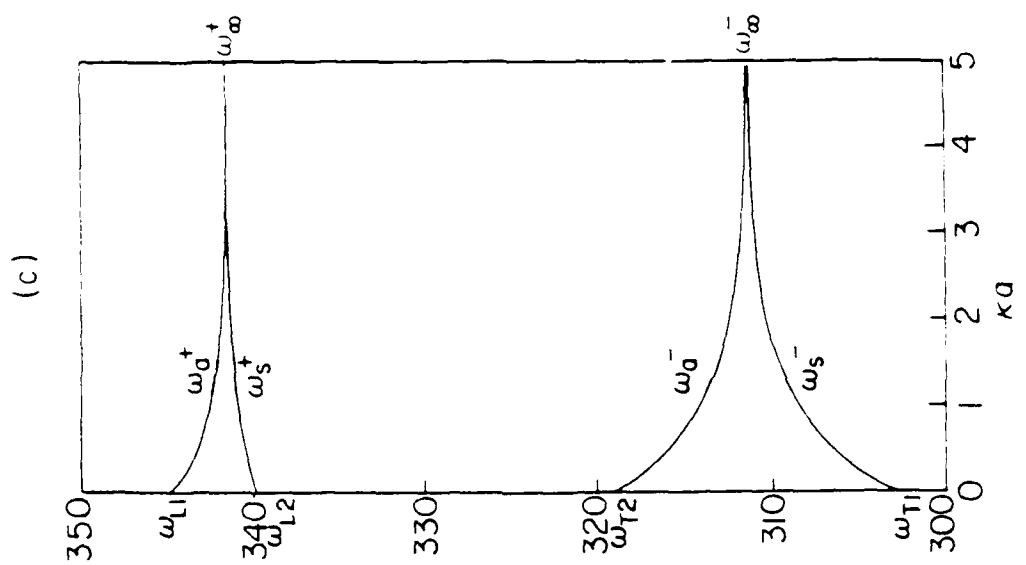
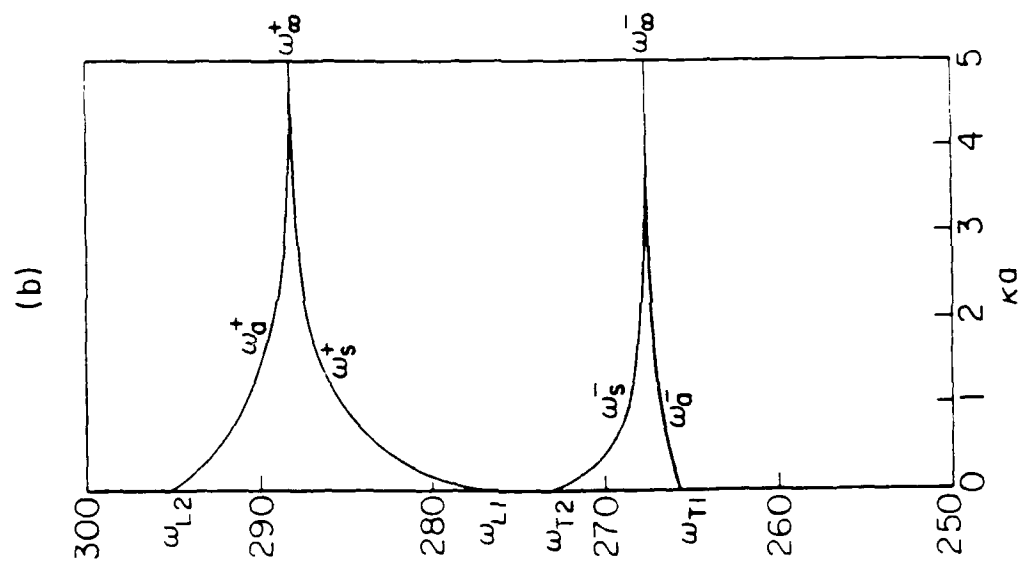
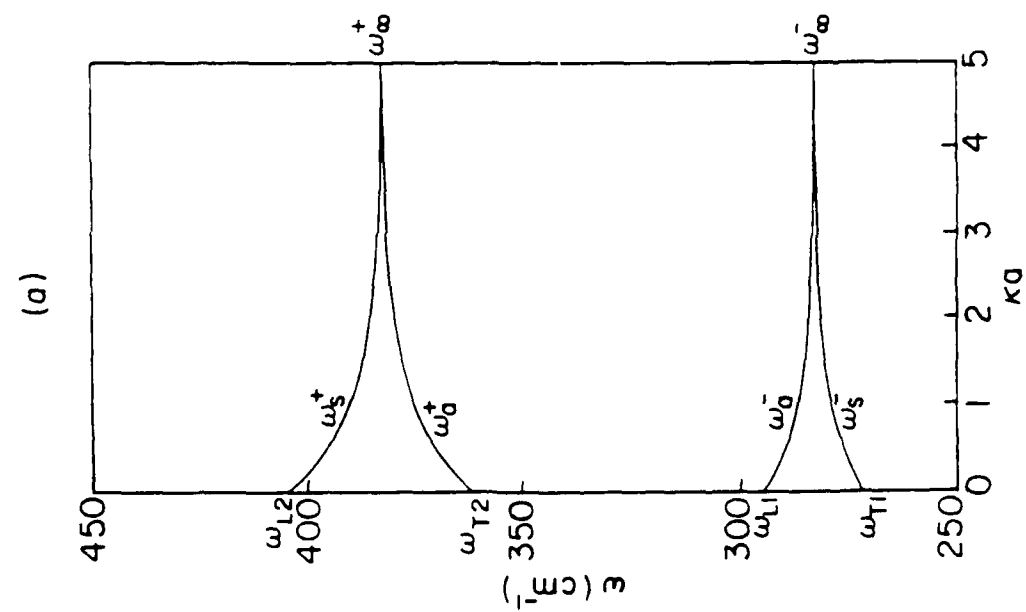


Fig. 2



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