# Optical phonons in mixed nonpolar-polar heterostructures

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A recently proposed continuum phenomenological model for long-wavelength optical modes in semiconductor heterostructures is employed to study mixed nonpolar-polar heterosystems. Results are presented for two systems of current interest, namely, the wide phonon band gap Si/ZnS quantum well and narrow phonon band gap Ge/GaAs quantum well.

## I. INTRODUCTION

Traditionally, nonpolar and polar structures have been considered separately (e.g., Si/Ge, and GaAs/AlAs respectively). Recently mixed nonpolar-polar systems have been developed to improve some desired physical properties. Metastable alloys of the form $(A^{III}B^{V})_{1-x}C_{2x}^{IV}$ have been studied both theoretically and experimentally. It is well known that this alloy has a specific feature in which the direct energy-band gap exhibits a much larger bowing than that of the conventional III-V alloy system. This is very promising for ''band-gap engineering'' of optoelectronic devices in the near-infrared wavelength range.

The Si/ZnS heterosystem has received a great deal of attention as current advances in epitaxy techniques have allowed the growth of heterostructures consisting of polar and nonpolar materials.[1,2] These researches were motivated by the integrability of silicon with advanced silicon microelectronic. Also, the lattice mismatch of cubic ZnS with respect to Si is only 0.3%. Molecular-beam-epitaxy growth of ZnS upon Si, and Si upon ZnS, can be easily obtained.[2]

Although a Ge/GaAs system was also considered for more than a decade,[3] theoretical investigation of this system has been slower. A close lattice match between Ge and GaAs and the other above-mentioned features are also present in these materials.

Phonons in semiconductor heterostructures have been widely studied due to their importance for many physical properties of the considered systems.[4] In polar materials, the normal modes involve coupled electrical and mechanical oscillations, and the associated long-range electric field suffices to couple different domains. In nonpolar materials the normal modes consist simply of mechanical vibrations, and only if their bulk bands overlap can modes of different domains be coupled. In this work we consider a hybrid type of heterostructure involving polar materials outside and nonpolar materials inside. We specifically consider the above-mentioned Si/ZnS and Ge/GaAs heterosystem. The reason for this selection is that different phonon properties are involved. The phonon dispersions of Ge and GaAs bulk materials overlap over a large frequency range. On the other hand, in Si/ZnS structures the mechanical vibrations characteristic of each constituent material would practically not penetrate the adjoining slabs. Strain effects are not important in both systems. This simplifies the calculations.

To describe long-wave modes, we employ a continuum phenomenological model,[5] which has proved useful in both polar[6–8] and nonpolar structures.[9] The general case of mechanical coupling between adjacent layers is considered in the calculations. In the Si/ZnS system a continuum model for optical modes considering totally confined mechanical oscillation was previously used.[10] Ge/GaAs was intensively studied (see Ref. 11 and references therein), but to the best of our knowledge no phonon calculations have been reported, though the phonons properties are more interesting than in the Si/ZnSe system.

In the two systems considered here, the nonpolar material has a phonon band gap larger than the polar one. In principle the reverse situation could be considered. After the study of the physics of the structures presented, we can predict what happens in the nonpolar/polar/nonpolar case.

In Sec. II, we describe the phenomenological model and present the main analytical results. In Sec. III, numerical results for different quantum wells are discussed. Finally some conclusions and general remarks are given.

## II. ANALYTICAL SETUP

We study the vector vibration amplitude $\mathbf{u}$, which represents the relative displacement of the two atoms in the unit cell. Other physical parameters of the medium are $\rho = \overline{M}/v_c$, the reduced mass density, where $\overline{M}$ is the reduced mass of the atoms and $v_c$ the unit-cell volume; $\omega_T$ and $\omega_L$ are the limiting longitudinal ($L$) and transversal ($T$) bulk frequencies; $\beta_T$ and $\beta_L$ are two parameters describing the dispersion of the oscillations, i.e., for bulk materials the $L$ and $T$ branches have a parabolic form $\omega_{L,T}^2(\mathbf{k}) = \omega_{L,T}^2 - \beta_{L,T}^2 k^2$ in the neighborhood of the $\Gamma$ point of the Brillouin zone. In layered structures all these quantities are piecewise dependent on the coordinates.

In the harmonic approximation the following coupled equations of motion are obtained from the Lagrangian density for polar materials, (see Ref. 6):

$$\rho(\omega^2 - \omega_T^2)\mathbf{u}(\mathbf{r}) = \alpha\nabla\phi(\mathbf{r}) + \rho\beta_L^2\nabla\nabla\cdot\mathbf{u}(\mathbf{r}) - \rho\beta_T^2\nabla\times\nabla\times\mathbf{u}(\mathbf{r}), \quad (1)$$

$$\nabla^2\phi(\mathbf{r}) = \frac{4\pi\alpha}{\epsilon_\infty}\nabla\cdot\mathbf{u}(\mathbf{r}), \quad (2)$$

 

where $\alpha=\sqrt{\omega_T^2\rho(\epsilon_0-\epsilon_\infty)/4\pi}$ is the coupling constant between mechanical oscillations and the electrostatic field $\phi$. The dispersive character of the optical phonons has been considered in the above expressions.

On Fourier transforming in two dimensions, the **r** dependence of all amplitudes involved acquires the form of a function of $z$ times $\exp i(\kappa_x x+\kappa_y y)$. For isotropic models one can choose $\kappa_x=0$ and $\kappa_y=\kappa$ without loss of generality. Following the methods used in Ref. 6, one obtains the solution space spanned by column matrices with elements $u_x$, $u_y$, $u_z$, and $\varphi$. One transverse amplitude is always decoupled from the rest. With the geometry here chosen, this is $u_x$. The remaining vibration amplitude is then sagittal.

The matching boundary conditions are straightforwardly derived from the original equations. They are the continuity of the mechanical $\mathbf{u}(z)$ and electrostatic $\phi(z)$ fields, and the normal component of the ''stress'' tensor $\boldsymbol{\sigma}$ and the displacement vector **D** at the interface.[6]

For a nonpolar material the general framework is the same, but $\alpha$ vanishes in Eqs. (1) and (2), as well as in the linearly independent solutions.[9] If this is matched to a polar medium, then in the nonpolar medium there is still a long-range electric field originating in the polar medium but decoupled from the mechanical vibraction in the nonpolar medium. Electrostatic continuity is also still required, but $\alpha$ vanishes for the nonpolar side of the matching interface. The transverse decoupled modes are of no interest for the present analysis.

The solutions for the well ($w$) (width $l$) and barrier ($b$) can be found in Ref. 6. In order to compare with the case of a nonpolar heterostructure of Ref. 9, we properly transform the mathematical expressions. In the nonpolar material well the electrostatic field is decoupled from the atomic oscillations. Using electrostatic boundary conditions, it is straightforward to write the amplitude of the part of the solution space corresponding to the external electrostatic field as a function of the amplitudes of the $L$ and $T$ solutions (compare the results presented in Refs. 6 and 9). In this way the formulas are similar to those obtained in Ref. 9. In fact, only the solution space for the barrier material has been changed.

The implications are more complicated than the simple changes in the penetration of modes in the barrier material. The secular equation for the eigenfrequencies has also been consequently modified. The influence of the magnitude of the transverse wave vector $\kappa$ is of great importance.

Looking at the solution space,[6,9] is it easy to see that the longitudinal and transverse solutions are decoupled at $\kappa=0$. The eigenvalues are independent of the electrostatic field, and we have the same secular equation as for nonpolar materials at $\kappa=0$ (see Ref. 9). Only the longitudinal amplitudes are coupled to the electric field in a simple way. For $\kappa\neq 0$ the electrostatic field is coupled to the mechanical oscillations, and this effect increases for larger values of $\kappa$.

### III. NUMERICAL RESULTS AND DISCUSSION

The growth direction in all cases is [001]. In Table I we report the values of the input parameters used in the calculations.

We first study a wide phonon gap heterostructure: the system ZnS/Si/ZnS. In Fig. 1 we compare the dispersion relation of three wells of the same material with thicknesses 10, 20, and 40 Å. We label the modes LO or TO according to their behavior at $\kappa=0$. Here and in the following we denote the even (odd) QW modes as $LO_{2m-1}$ ($LO_{2m}$) and $TO_{2m}$ ($TO_{2m-1}$), $m=1,2,3,\ldots$. Although the modes are somewhat more dispersive than for Ge barriers (see Ref. 9), the dispersion is still fairly weak up to large values of $\kappa\approx 10^7\ \mathrm{cm}^{-1}$. In fact, the gap is smaller than in the Si/Ge case. A comparison with the total confined phonon case studied previously by Sun *et al.*[10] is also presented. In Fig. 1 we show the LO (solid up triangle) and TO (open down triangle) phonon modes at $\kappa=0$. The symbols are shifted to the right for the sake of clarity. We see that only the topmost modes in the widest wells show reasonable agreement. In most of the cases the discrepancy is very large.

TABLE I. Input parameters.

| Material | Si | Ge | GaAs | ZnS |
|---|---|---|---|---|
| $\epsilon_0$ | 11.7 | 16 | 13.2 | 8.3 |
| $\epsilon_\infty$ | $\epsilon_0$ | $\epsilon_0$ | 10.9 | 5.2 |
| $\omega_{LO}(\Gamma)$ ($\mathrm{cm}^{-1}$) | 518 | 303 | 292 | 351 |
| $\omega_{TO}(\Gamma)$ ($\mathrm{cm}^{-1}$) | $\omega_{LO}(\Gamma)$ | $\omega_{LO}(\Gamma)$ | 269 | 279 |
| $\beta_{LO}^2$ ($10^{-12}$) | 7.63 | 2.8 | 2.91 | 1.31 |
| $\beta_{TO}^2$ ($10^{-12}$) | 9.5 | 5.08 | 3.12 | $-10.67$ |
| *mismatch* to Si or Ge (%) | 0 | 0 | 0.1 | 0.4 |

For a smaller gap heterostructure we note some interesting effects. For the GaAs/Ge/GaAs well (Fig. 2), some

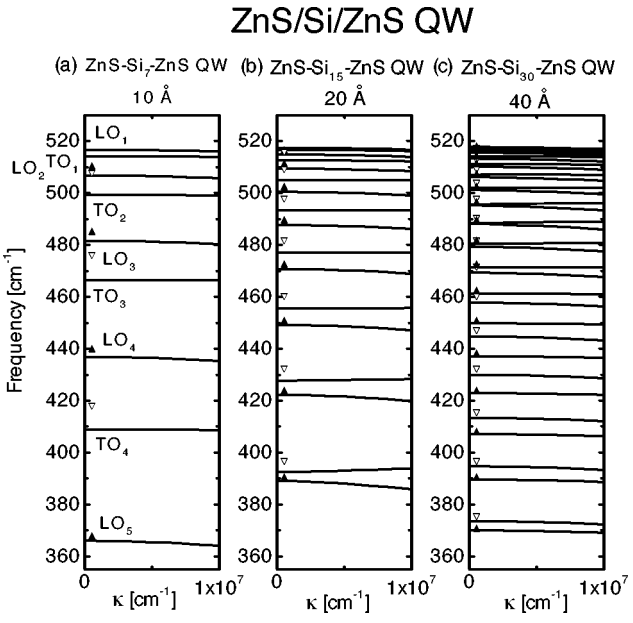


FIG. 1. Calculated phonon-dispersion curves for wide-phonon-gap heterostructures: (a) a $ZnS/Si_7/ZnS$ QW, (b) a $ZnS/Si_{15}/ZnS$ QW, and (c) a $ZnS/Si_{30}/ZnS$ QW. Phonon frequencies are in $\mathrm{cm}^{-1}$. The phonon modes are labeled according to their character at $\kappa=0$. The phonon branches are practically nondispersive. LO (solid up triangle) and TO (open down triangle) totally confined phonon modes at $\kappa=0$ are also presented. See the text for details.

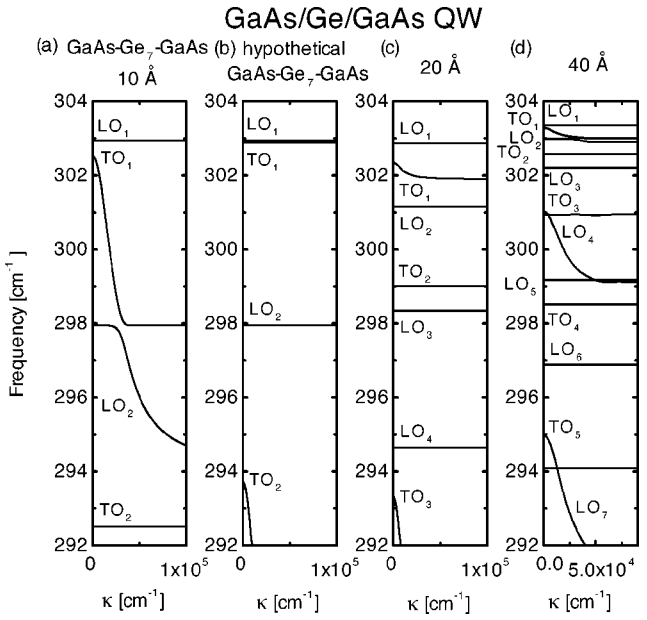


FIG. 2. Calculated phonon-dispersion curves for short-phonon-gap heterostructures: (a) a $GaAs/Ge_7/GaAs$ QW, (b) a hypothetical nonpolar $GaAs/Ge_7/GaAs$ QW, (c) a $GaAs/Ge_{15}/GaAs$ QW, and (d) a $GaAs/Ge_{30}/GaAs$ QW. Phonon frequencies are in $cm^{-1}$. The phonon modes are labeled according to their character at $\kappa=0$. The phonon branches are notably influenced by the electrostatic potential.

modes are very dispersive while others are practically nondispersive. Also, anticrossings are present in the three wells shown in the figure. These features are characteristic of the polar structures [e.g., GaAs and AlAs quantum wells (QW's) reported by Comas *et al.*[6] and Pérez-Alvarez *et al.*[7]], and are associated with the electric field involved. Comas *et al.* explained how for totally confined mechanical modes the long-range electric field is related to this characteristic dispersion of the modes.[6] Thus this significantly changes the behavior of these modes, so they are similar to the polar ones. The longest wavelength modes are the most greatly influenced. In fact, for $\kappa>10^5\ cm^{-1}$ the dispersion is practically unappreciable.

Figure 2(b) shows the $\omega$ vs $\kappa$ curves for a well with a hypothetical nonpolar barrier material, with the parameters of the GaAs $[\alpha=0,\omega_{LO}(\Gamma)=\omega_{TO}(\Gamma)]$. The dispersion is characteristic of a nonpolar structure, i.e., the electric field and not the small phonon gap is responsible for the behavior of the phonon modes in the mixed nonpolar-polar heterostructure.

In the nonpolar material the mechanical oscillations are not coupled to the electrostatic potential. But they are coupled through the interface to the atomic vibrations in the polar semiconductor, which in turn are coupled to their own inherent electric field. In this way the electrostatic field has some influence on the mechanical oscillations of the nonpolar atoms in a mixed nonpolar-polar heterostructure. The wider nonpolar wells are less sensitive to the influence of the polar material on the other side of the interface. For this reason the phonon branches are less dispersive for wider wells (see Fig. 2). On the other hand, the electrostatic field has a transverse character. At $\kappa=0$ it is not coupled to the atomic oscillations. Its influence begins for $\kappa\neq0$ (see the discussion at the end of Sec. II), and increases as the value of $\kappa$ grows, while the mode has a sufficiently long wavelength $(\kappa=2\pi/\lambda)$ to extend farther than a single layer. For this reason the dispersive character is only appreciable in Fig. 2 for $\kappa<10^5\ cm^{-1}$.

## IV. CONCLUSIONS

We have studied the optical long-wavelength phonon modes in mixed nonpolar-polar heterostructures, employing a phenomenological model. The analytical results are cast in expressions similar to the ones of nonpolar materials[9] in order to compare both results from mathematical and physical points of view. We focused our attention on ZnS/Si/ZnS and GaAs/Ge/GaAs wells due to their importance in practical and basic research.

We have shown that an external electrostatic potential gives a polar character to the phonon modes of nonpolar Ge embedded in polar GaAs. The effect of the electric field increases from $\kappa=0$ until it reaches values of order $10^{-3}$ times the value of $\kappa$ corresponding to the Brillouin-zone boundary, and it is not appreciable for larger values of the wave vector. This effect is more important in the thinnest wells, but for sufficiently high barriers the influence of the external field is lower. This is the case for the ZnS/Si/ZnS structure, where we also show the importance of employing correct boundary conditions for the right description of all phonon modes.

Of course a structure of the nonpolar/polar/nonpolar type could also be considered. The polar character of the well material will then be predominant in the heterostructure phonon spectra, as previous physical analysis has shown.[11]

In this work we always considered ideal interfaces. Strain due to a large lattice mismatch, nonideal interfaces, and strange atomic planes at the interface could also be studied in the frame of the continuum long-wave model used here.[9,12] This describes a real situation found experimentally, e.g., when growing the Si/ZnS heterosystem an As monolayer is intercalated to satisfy the local bonding requirements.[2]

[1] M. Yokoyama, K.I. Kashiro, and S.I. Otha, J. Cryst. Growth **81**, 73 (1987).
[2] X. Zhou, S. Jiang, and W.P. Kirk, J. Appl. Phys. **82**, 2251 (1987).
[3] J. Pollman and S.T. Pantelides, Phys. Rev. B **21**, 709 (1980).
[4] B. Jusserand and M. Cardona, *in Light Scattering in Solids V*, edited by M. Cardona and G. Güntherodt (Springer-Verlag, Heidelberg, 1989).
[5] C. Trallero-Giner, F. García-Moliner, V.R. Velasco, and M. Cardona, Phys. Rev. B **45**, 11 944 (1992).
[6] F. Comas, R. Pérez-Alvarez, C. Trallero-Giner, and M. Cardona, Superlattices Microstruct. **14**, 95 (1993); R. Pérez-Alvarez, V.R. Velasco, and F. García-Moliner, Phys. Scr. **51**, 526 (1995).
[7] R. Pérez-Alvarez, F. García-Moliner, V.R. Velasco, and C. Trallero-Giner, J. Phys.: Condens. Matter **5**, 5389 (1993).
[8] C. Trallero-Giner, R. Pérez-Alvarez, and F. García-Moliner, *Long Wave Polar Modes in Semiconductor Heterostructures*

(Pergamon/Elsevier Science, London, 1998).
[9] F. de León-Pérez and R. Pérez-Alvarez, Phys. Rev. B **61**, 4820 (2000).
[10] G. Sun, Y. Lu, L. Friedman, and R.A. Soref, Phys. Rev. B **57**, 6550 (1998).
[11] J. Rufinus and G.E. Crook, Appl. Phys. B: Photophys. Laser Chem. **81**, 758 (1997).
[12] R. Pérez-Alvarez and C. Trallero-Giner, Phys. Scr. **56**, 407 (1997).