Optical phonon modes at $GaAs/Al_xGa_{(1-x)}As$ semiconducting interfaces: effects of the concentration of Al

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We study long-wavelength polar optical modes at semiconductor interfaces of the GaAs/Al_xGa_(1-x)As system and take into account the influence of the Al concentration. We have considered two cases in which the interface is kept either at a variable or at a fixed electrostatic potential. The spectrum of excitations then shows existence of localized and resonant modes depending on the concentration of Al. For the case of a fixed electrostatic potential (metallized interface), the appearance of a new interface mode besides the resonant one, is found for certain ranges of the concentration x.

Keywords: Polar optical phonons; semiconducting interfaces

Se presenta el estudio de los modos de oscilación ópticos polares de longitud de onda larga en intercaras semiconductoras del sistema $GaAs/Al_xGa_{(1-x)}As$, teniendo en cuenta especialmente la influencia del valor de la concentracion de Al. Se consideran dos situaciones diferentes: en una se mantiene la intercara con el potencial electrostático variable, mientras que en la otra se considera que este potencial es fijo (intercara metalizada). El análisis del espectro de oscilaciones indica la existencia de modos localizados y de modos resonantes, dependiendo de la concentración de Al. Para el caso de la intercara metalizada, se detecta la aparición de un nuevo modo de intercara más allá del modo resonante, cuando la concentración x toma valores en un rango determinado.

Descriptores: Fonones ópticos polares; intercaras semiconductoras

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1. Introduction

During the last few years, there has been a growing interest in the study of the phonon properties in semiconducting nanostructures, mainly those concerning to long wavelength polar optical oscillations. Different approaches have been presented in order to give an appropriate phenomenological description of polar optical phonons in the long wavelength limit [1, 4]. In that scenario, several results have been reported in the cases of layered structures [5,9], quantum wires [10, 12] and quantum dots [13]. A model including the full coupling between the mechanical and electrical interactions and satisfying the mechanical and electrical boundary conditions has been put forward [2] and applied to the study of long-wavelength polar optical modes in GaAs-based heterostructures [5, 7]. The results thus obtained agree remarkably well with microscopic ab initio calculations [14, 15]. This model has also been applied recently to the study of the spectrum of the polar optical modes of GaAs surfaces and $Ga_x Al_{(1-x)} As$ interfaces [9].

In Ref. 9, localized and resonant modes were found for the case of free surfaces. When assuming a metallized surface, thus modifying the electrostatic boundary conditions, the *localized* mode is almost unaffected while the *resonant* mode disappears, thus evidencing a strong electrostatic character.

In the case of an interface, where only resonant modes exist, these modes are also affected when the interface is metallized, thus also manifesting its strong electrostatic character. This explains why a dielectric model (see, e.g., Ref. 16) successfully accounts for the interface modes found in quantum well systems. In quantum wells the modes having predominantly electrical character can be identified with the interface modes [17]. As we have seen for just one interface only one such mode is obtained. When a symmetrical quantum well is formed the two degenerate modes corresponding to the two interfaces split into two interfaces modes of opposite parities. The identification of the interface modes as predominantly electrostatic is also relevant for the theory of electron-phonon interaction in heterostructures when polar optical modes are involved. Since the measure of this interactions is $e\varphi$ a detailed analysis suggests that this interaction should be dominated by interface modes [18].

It would be interesting to study the effects of the Al concentration in the ternary compound on the polar optical modes of $Al_xGa_{(1-x)}As$ free surfaces and $GaAs/Al_xGa_{(1-x)}As$ interfaces. Because in the spirit of the *two-modes* model [19] the longitudinal and transverse thresholds will vary with the concentration x, in such a way that the longitudinal threshold of the ternary compound will cross the transverse threshold of pure GaAs, the question is if some important effect on the interface modes found in [9] can be expected. Thus, the aim of this paper is to investigate the polar optical oscillation spectrum in a $GaAs/Al_xGa_{(1-x)}As$ single heterostructure, and to determine the influence of x on it.

A brief reminder of the basic theoretical aspects and numerical values of the parameters employed in the phenomenological model are presented in Sec. 2. In Sec. 3 we present the results and conclusions coming out from our calculations.

2. Theoretical and practical aspects of the model

The model to be used here is a macroscopic continuum one which assumes the ternary alloy as a three-dimensional system with translational invariance. This fact, which is common in the study of the electronic structure in those materials has also been assumed in the long wavelength phenomenological dielectric continuum model that has been widely applied to the description of the polar optical phonon modes and to the study of the electron-phonon interaction in GaAs/Al_xGa_(1-x)As heterostructures, and it is possible to mention a large number of papers devoted to its development and applications in those systems [20–26].

Let us consider a medium where $\mathbf{u}(\mathbf{r}, t)$ is the displacement and $\varphi(\mathbf{r}, t)$ is the scalar potential associated with the electric field $\mathbf{E} = -\nabla \varphi$. We have a mechanical equation of motion for \mathbf{u} which is of the form [2]

$$\rho(\omega^2 - \omega_{\rm TO}^2) \mathbf{u} + \boldsymbol{\nabla} \cdot \boldsymbol{\tau} - \alpha \nabla \varphi = 0,$$

$$\alpha^2 = \frac{\omega_{\rm TO}^2 \rho(\epsilon_0 - \epsilon_\infty)}{4\pi}.$$
 (1)

The harmonic oscillator part is contained in the first term. The second term has the nature of a dispersive mechanical term and for an isotropic medium is of the form

$$\tau_{ij} = -\rho(\beta_{\rm L}^2 - 2\beta_{\rm T}^2)\delta_{ij}\boldsymbol{\nabla}\cdot\mathbf{u} - \rho\beta_{\rm T}^2(\nabla_i u_j + \nabla_j u_i), \quad (2)$$

where $\nabla_i = \partial/\partial x_i$ and β_L and β_T are adjustable material parameters. The sign of (2) is opposite to the usual one for acoustic waves on account of the negative dispersion of the optical modes. The third term measures the effect of the coupling between the u and φ fields on the equation of motion for u. Simultaneously we have a Poisson equation for φ which reads

$$\nabla^2 \varphi = 4\pi \gamma \nabla \cdot \mathbf{u}; \quad \gamma = \alpha / \epsilon_{\infty}. \tag{3}$$

The physical meaning of the above equation is that φ is created by the polarization charge $\rho_e = \nabla \cdot \mathbf{P}$ of the polarization field given by

$$\mathbf{P} = \alpha \mathbf{u} + \frac{\epsilon_{\infty} - 1}{4\pi} \mathbf{E} = \alpha \mathbf{u} - \frac{\epsilon_{\infty} - 1}{4\pi} \nabla \varphi.$$
(4)

The term on the r.h.s. of (3) measures the effect of the coupling between the φ and **u** fields on the field equation for φ .

The non-retarded limit $(c \to \infty)$ is considered, which is fully justified for the situation under study.

For an isotropic bulk homogeneous medium it is possible to obtain independent equations for $\mathbf{u}_{\rm L}$ and $\mathbf{u}_{\rm T}$ and this yields at once the longitudinal and transverse modes with dispersion relations $\omega_{\rm L}^2 = \omega_{\rm LO}^2 - \beta_{\rm L}^2 k^2$ and $\omega_{\rm T}^2 = \omega_{\rm TO}^2 - \beta_{\rm T}^2 k^2$ respectively, where **k** is the 3D wavevector and $\omega_{\rm LO}^2 = \omega_{\rm TO}^2 (\epsilon_0/\epsilon_{\infty})$.

However, our concern is to study the matching of different media at an interface: Let discuss one interface which we take as the plane z = 0. First, we Fourier-transform in the 2D plane of the interface, so that the ω -dependent vibration amplitudes are of the form

$$\mathbf{u}(\boldsymbol{\rho}, z) = \exp(i\boldsymbol{\kappa} \cdot \boldsymbol{\rho})\mathbf{u}(z), \tag{5}$$

where κ , ρ are 2D vectors (wavevector and position, respectively).

We proceed likewise for φ and concentrate on $\mathbf{u}(z)$ and $\varphi(z)$. These are (ω, κ) -dependent quantities for which, after 2D Fourier transform, we have (ω, κ) -dependent differential equations in the independent variable z. One interesting result arising from the model is that the coupled oscillation modes have mixed polarizations as it was discussed in Ref. 8 for the case of a double heterostructure. This means that, for $\kappa \neq 0$ the coupled modes are neither longitudinal nor transversal, but with both characters at the same time. Some modes are predominantly longitudinal and other modes are predominantly transversal. The only case in which the coupled modes have purely longitudinal or transversal character is when $\kappa = 0$.

The matching boundary conditions were obtained in Ref. 2, and for the single interface they are, in the variable z,

$$u_{j}(+0) = u_{j}(-0),$$

$$\varphi(+0) = \varphi(-0),$$

$$\tau_{zj}(+0) = \tau_{zj}(-0),$$

$$\epsilon_{\infty} \frac{d\varphi}{dz} - 4\pi\alpha u_{z} = \text{continuous.}$$
(6)

On evaluating these expressions at $z = \pm 0$ we must take into account the different values of β_L , β_T , ϵ_{∞} and α on both sides of the interface.

We have four amplitudes (u_x, u_y, u_z, φ) , four coupled second order linear differential equations and eight matching boundary conditions. As in the study of piezoelectric surface or interface waves it proves convenient to define a tetrafield,

$$\mathbf{F} \equiv \begin{bmatrix} \mathbf{F}_M \\ F_E \end{bmatrix} \equiv \begin{bmatrix} \mathbf{u} \\ \varphi \end{bmatrix}, \tag{7}$$

which has mechanical and electrical components and to condense the system (1), (3) in the form

$$\mathbf{L} \cdot \mathbf{F} = 0, \tag{8}$$

where \mathbf{L} is a 4×4 differential matrix which can be readily written down explicitly. Upon 2D Fourier transform \mathbf{L} depends on κ (ω -dependence understood everywhere) and contains the different ial operator d/dz. A convenient technique for solving this problem is the Surface Green Function Matching (SGFM) method [27], which, for the case we are interested on, was applied in Ref. 9. In the SGFM method, the key element is the surface projection of the Green function of the system \mathcal{G}_s . The eingevalues $\omega(\kappa)$ of the problem can be obtained from a procedure in which we calculate from \mathcal{G}_s the κ -resolved local density of states at z = 0 [27]:

$$\mathcal{N}_s(\omega, \kappa) = -\frac{1}{\pi} \lim_{\varepsilon \to \infty} \text{ Im Tr } \mathcal{G}_s(\omega + i\varepsilon, \kappa).$$
 (9)

The eigenvalues $\omega(\kappa)$ are then the frequencies at which the peaks in the density of states \mathcal{N}_s appear. This procedure is very convenient, especially when resonant modes can exist. Such resonant modes will appear as Lorentzian peaks superimposed on the continuum of bulk scattering states in the density of states.

Analogously to the piezoelectric case [27, 28] we shall also consider the situation in which the interface is coated by an infinitesimally thin metallic layer (metallized interface), which serves to ground that interface, thus keeping it at a constant potential while not appreciably affecting the mechanical vibrations [29, 31]. For the boundary conditions, instead of having the second equality of Eq. (7), the two independent equalities $\varphi(+0) = 0$ and $\varphi(-0) = \varphi_0$ are imposed; φ_0 being an arbitrary constant. The rules of the electrostatics are always obeyed, but the conditions at the interface are now different. Since there is always an inherently arbitrary additive constant in the electrostatic potential, the results in the end cannot depend on φ_0 . In fact, the inclusion of this new condition does not affect the dynamical analysis because it does not introduce any dependence on (ω, κ) . In this case, the projection of the Green function of the system can be obtained from the equation

$$\mathcal{G}_{s}^{-1}(\mathrm{met}) \equiv \begin{bmatrix} \mathcal{F}_{M} \cdot \mathcal{G}_{s}^{-1} \cdot \mathcal{F}_{M} & \mathcal{F}_{M} \cdot \mathcal{G}_{s}^{-1} \cdot \mathcal{F}_{E} \\ 0 & \chi_{0} \end{bmatrix}, \quad (10)$$

where \mathcal{F}_M and \mathcal{F}_E denote the mechanical and electrostatic projections of the non-metallized \mathcal{G}_s^{-1} , and χ_0 is a constant value related to the fixed potential; thus, the arbitrariness of φ_0 is transferred to it.

Since we have studied $GaAs/Al_xGa_{(1-x)}As$ systems, some words about the fitting procedure employed to estimate the input parameters are in order. The mass density and the background dielectric constants were obtained from a linear interpolation of the value for the pure materials-AlAs and GaAs-according to the expressions [32]

$$\rho(x) = 5.36 - 1.60x, \quad (c.g.s.);$$

$$\epsilon_0(x) = 13.18 - 3.12x, \quad (e.s.u.);$$

$$\epsilon_{\infty}(x) = 10.89 - 2.73x, \quad (e.s.u.). \quad (11)$$

However, this type of interpolation would not work for ω_{LO} , ω_{TO} , β_L and β_T . It has been strongly argued on the basis of experimental evidence [19] that the ternary compound

 $Al_xGa_{(1-x)}As$ can be described in the *two-mode model*. We shall adopt this viewpoint. It then follows that if we study the matching to GaAs we must assign to the ternary alloy the values of the frequencies for the LO and TO modes found experimentally for the GaAs-like modes in this alloy [19]

$$\omega_{\text{LO.GaAs}}(x) = 292.37 - 52.83x + 14.44x^2,$$

$$\omega_{\text{TO.GaAs}}(x) = 268.50 - 5.16x - 9.36x^2.$$
(12)

(Here and henceforth ω is always given in cm⁻¹).

As it is discussed in Refs. 33 and 34 $\beta_{\rm L}$ and $\beta_{\rm T}$ are given by $\beta_{\rm L}^2 = (\Lambda_1 + \Lambda_2)/\rho$ and $\beta_{\rm T}^2 = \Lambda_1/\rho$, where ρ is the mass density of the bulk material and Λ_1 and Λ_2 are two independent phenomenological parameters describing a fourth rank tensor associated to the elastic properties of the medium. This tensor appears in the term corresponding to the internal stresses in the Lagrangian density of the system; from which Eqs. (1)–(3) are derived [4, 5]. Λ_1 and Λ_2 are obtained from the curvature of the phonon dispersion relation near $\kappa = 0$ [2, 8, 33].

The β_L and β_T parameters are not usually reported in the literature and they are not known for the different types of modes in alloys. We have estimated their values for the pure materials (x = 0, x = 1) from the experimental curves of Ref. 19.

We have also made the following assumption: For very low (high) concentrations of Al, that is for $x \approx 0(x \approx 1)$, we take dispersion laws with $\beta = 0$ for the AlAs(GaAs)-like modes. This assumption relies on the fact that for these situations the atoms in question are isolated and their phonon branches must be flat. For a given concentration x, we perform a linear interpolation between the values for x = 0 and x = 1. With ω in cm⁻¹, β is dimensionless and we obtain

$$\beta_{\rm L,GaAs}^2(x) = 2.91(1-x)10^{-12},$$

$$\beta_{\rm T,GaAs}^2(x) = 3.12(1-x)10^{-12}.$$
 (13)

It is obvious that this interpolation procedure is not the only one which can be conceived. As it was reported in Ref. 9, the values of these parameters do not significantly modify the results coming out from our calculations.

3. Results and conclusions

We discuss next the results obtained with the model above considered. A small imaginary part equal to 10^{-6} cm⁻¹ was added to ω , in order to adscribe the local modes to very narrow peaks which are shown instead of the ideal δ -functions. Repeating the calculations for different values of x in the GaAs/Al_xGa_(1-x)As system, we have observed the concentration of Al to have certain influence on the interface modes previously obtained in Ref. 9: the resonant interface mode and the localized interface mode. Even when both kinds of



FIGURE 1. LDOS for the resonant mode in a non-metallized interface for x from 0.9 to 0.5. Curves, from *left* to *right*, correspond to decreasing values of Al concentration with step $\Delta x = 0.05$.

modes have, at the same time, electrical and mechanical nature, the so-called resonant mode bears a very strong electrostatic origin. It is actually a resonance with the bulk GaAs continuous. As it is graphically shown in Refs. 9 and 35, if the physical nature of the spectral strength is analysed by separately evaluating the contributions of the mechanical and electrical terms \mathcal{F}_M and \mathcal{F}_E , it can be seen that the interface resonant mode is in fact an electrical mode.

In Figs. 1 and 2, the spectral strength for $\kappa = 4 \times 10^6$ cm⁻¹ is presented through the LDOS. In Fig.1, the different curves correspond, from left to right, to values of x running down from 0.9 to 0.5 with step $\Delta x = 0.05$. Unlike the case of the free GaAs surface (GaAs for z < 0, and vacuum for z > 0) also discussed in [9], no localized phonon mode with frequency beyond the maximun frequency of the GaAs crystal is observed to come out from the numerical calculation, while a peak corresponding to a resonant mode is present. In the case of the surface this resonant mode is no other than the Fuchs-Kliewer one [36], and in case of the interface it has the same electrostatic origin. For higher values of x, both sides of the interface become mechanically very different media, hence the only coupling between then is the electrostatic one. It is shown that the frequency of the resonant mode slowly increases with decreasing x and the peak, at last, disappears for values below $x \simeq 0.5$, its frequency lying between the longitudinal and the transversal GaAs bulk thresholds, which, according to Eqs. (12) are $\omega_{LO} = 292.37$, and $\omega_{TO} = 268.5$, respectively. This fact resembles the situation of the "interfacelike modes" appearing in the double heterostructure [8]. Calculations for different values of κ show the same behavior with x. The only difference is that the frequency of the peaks slowly decreases for decreasing κ .

At the same time, from Fig. 2, curves from left to right correspond to values of x decreasing from 0.2 to 0, with $\Delta x = 0.05$. In this case, no resonant mode is present; but a feature -not precisely a well-defined peak- is seen in the LDOS for frequencies below the bulk transversal optical phonons frequency. This corresponds to the so-called localized interface mode. This kind of modes appears mostly due



FIGURE 2. LDOS for the localized transverse-like oscillation mode in a non-metallized interface for x from 0.2 to 0. Curves, from *left* to *right*, correspond to decreasing values of Al concentration with $\Delta x = 0.05$. For x = 0 this mode becomes the well-known GaAs bulk transverse phonon.

to the mechanical difference between the two media. Unlike the resonant mode, whose mechanical amplitude decreases extremely strongly on the Al_xGa_(1-x)As part of the structure, this locallized mode has a stronger mechanical character, because both sides are not that mechanically different. It is also present for higher values of x; but it was not shown in Fig. 1 because its relative weight in the LDOS does not allow to discern it in the graphics. It is seen that the position of the localized mode in the transverse threshold moves, as $x \to 0$, toward a peak corresponding to the frequency of the transverse oscillations of GaAs bulk crystal. Of course, the localized mode disappears when x = 0. In the same limit, a similar behavior; i.e., the appearance of a peak corresponding to the bulk GaAs longitudinal optical phonons frequency can be observed, as it is expected, for in that case we only have the bulk GaAs crystal, and not an interface. We omit to show this second peak. For different values of κ the change in the peak position is the same described above.

For the metallized interface, we can choose a particular value of κ ; for instance $\kappa = 2 \times 10^6 \text{ cm}^{-1}$ to represent the situation, and we choose a range of frequency values just around the mode we are interested on. In that case, for values of Al concentration below 0.525, no special feature in the LDOS can be seen. But when the value of x is slightly increased, an interesting effect is observed (Fig. 3): For a very narrow range $0.526 \ge x \ge 0.529$, there always appears some feature in the LDOS below the corresponding value of $\omega_{LO}(\kappa)$ of GaAs for the x given. At the same time, there appears a second feature above that frequency (Fig. 3a) whose height increases, and when $x \simeq 0.52818$ it becomes higher than the left peak and constitutes a broad peak in the LDOS (Fig. 3b). For x around 0.52822, this peak clearly splits into two well resolved peaks (Fig. 3c), the left being smaller than the right one. Both peaks move, respectively, to lower and higher values of the frequency as x increases (Fig. 3d). The height of the right peak becomes bigger, while for a value of Al concentration such as 0.52830, the left peak is much more smaller, and for $x \simeq 0.529$ no left peak appears above the



FIGURE 3. LDOS for a metallized interface showing: (a) the appearance of the resonant mode; (b) the peak corresponding to that mode (a different scale is used in order to clearly show the existence of the peak); (c) the splitting of the resonant mode; (d) the separation of the two peaks as x increases and (e) the dissappearance of the left peak and the prevailing of the other one, close to the GaAs bulk transversal frequency, as reported in Ref. 9.

 $\omega_{\rm LO}(\kappa)$ of GaAs, and the right peak remains and locates now above the value of $\omega_{\rm TO}$ in the GaAs bulk (Fig. 3e). It must be remarked that, while this process occurs, the right peak is always located to the left of the bulk transversal frequency of the GaAs modes, until it crosses that value of ω . If $x \ge 0.529$, a situation very similar to that reported in Ref. [9] is ob-



served; i.e., the resonant mode is always very close to the transverse threshold. The same picture can be seen for other values of κ , but with a slightly different range for x. It is clear that these concentration variations and frequency shifts are too small to be observed experimentally, but it could be possible to find a system consisting of materials with stronger ionic character such as II-VI semiconductors for which the situation would be better for an experimental detection.

Figures. 4a and 4b show the appearance and dissappearance of the second resonant mode for $\kappa = 4 \times 10^6 \text{ cm}^{-1}$. In both figures, the three-dimensional plot allows to observe the combined effect of the variation of ω and x.

It is possible to identify the remaining peak with the resonant mode above described. The left-disappearing-peak corresponds to another-new-resonant mode, and the reason for its origin has only been the modification of the electrical boundary conditions when metallizing the interface. This constitutes an additional evidence of the predominantly electrical character of this kind of modes.

The main conclusion of this work is that in a $GaAs/Al_xGa_{(1-x)}As$ single heterostructure, the type of interface modes appearing depends on the value of the concentration of Al in the barrier. Higher values of x give rise to resonant interface modes which bear a strong electrostatic character. This modes would be responsible for an appreciable electron-phonon interaction. On the other hand, small values



FIGURE 4. a) LDOS for a metallized GaAs/Al_xGa_(1-x)As interface showing the splitting of the resonant mode below the transversal threshold ($\kappa = 4 \times 10^6 \text{ cm}^{-1}$). b) The continuation of LDOS from Fig. 4a for higher values of x showing the disappearance of the left peak ($\kappa = 4 \times 10^6 \text{ cm}^{-1}$).

of x allow for interface localized modes close to the bulk GaAs transverse threshold. Although the optical phonon modes in the framework of the macroscopic continuum model that has been used here do not exhibit pure LO or pure TO polarizations, the contribution of the localized modes to the electron-phonon interaction is expected to be small even when they have some longitudinal character due to the polarization mixing.

The idea of a very thin metallic layer at the interface between two semiconducting materials would seem rather difficult to accept as a real physical system. Nonetheless, the possibility of the growing of ultrathin metallic or semiconducting epilayers is already a fact. What we intend to discuss is the important effect that the change in the electrostatic boundary conditions has upon the polar optical oscillations at the interface between crystalline ionic systems. According to the results we are presenting here, it seems that the analysis of this problem deserves some attention. For instance, it is interesting to study the change in the scalar potential φ under the situation above described and, consequently, its effect on the electron-phonon interaction, that could be more important than the predicted frequency shifts. This work is already in progress.

Recently, an envelope-function theory for phonons in heterostructures [4] shows interesting results for the InAs/GaSb system. Even when it appears that the study of multipleinterfaces heterostructures is more attractive, the consideration of a single one still gives the possibility of a deeper understanding of the effect of interfaces on the long-wavelength oscillations, so it would be desirable to study, for instance, the presence of the mechanical interface modes which occurs in that system.

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