Polar optical phonons at GaAs|$Al_xGa_{(1-x)}As$ interfaces: influence of the concentration of Al

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Abstract

We study long-wavelength polar optical modes at semiconductor interfaces of GaAs|$Al_xGa_{(1-x)}As$ and take into account influence of the Al concentration. We have considered two cases in which the interface is kept at unfixed and fixed electrostatic potential. The spectrum of excitation then shows existence of localized and resonant modes to depend on the concentration of Al. For the case of a fixed electrostatic potential, a splitting of the resonant mode near the transverse threshold is found for certain ranges of the concentration $x$. The existence of these modes has been obtained by computing the spectral strength through the Surface Green Function Matching (SGFM) method.

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I. 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 present in order to give an appropriate phenomenological description of polar optical phonons in the long wavelength limit [1-4]. In that scenario, there are reported some results in the cases of layered structures [5-9], quantum wires [10-12] and quantum dots [13]. In a recent paper [9] the spectrum of polar optical modes for GaAs surfaces and GaAs|Al_{0.9}Ga_{0.1}As interfaces was studied by using the theory developed in [2,5,6] and later applied to the study of those modes in GaAs-based double heterostructures, with remarkable agreement with microscopic ab-initio calculations [14,15].

In the paper mentioned above [9], for free surfaces localized and resonant modes were found. 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 the 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 the dielectric model (see e.g. [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 one obtains only one such mode. When the symmetrical quantum well is formed the two degenerate modes corresponding to the two interfaces split into the 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 $e\varphi$ the analysis suggests that this should be dominated by the role of the interface modes [20]. In our work we shall concentrate mainly on influence of concentration $x$ of Al on existence and “behavior” of polar optical localized and resonant modes at the GaAs|Al$_x$Ga$_{(1-x)}$As interface.
II. THEORETICAL AND PRACTICAL ASPECTS OF THE MODEL

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

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

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

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

$$\tau_{ij} = -\rho(\beta_L^2 - 2\beta_T^2)\delta_{ij} \nabla \cdot u - \rho\beta_T^2(\nabla_i u_j + \nabla_j u_i), \quad (2)$$

where $\nabla_i = \partial/\partial x_i$ and $\beta_L$ and $\beta_T$ are adjustable 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 $\varphi$ fields on the equation of motion for $u$. Simultaneously we have a Poisson equation for $\varphi$ which reads

$$\nabla^2 \varphi = 4\pi \gamma \nabla \cdot u; \quad \gamma = \alpha/\epsilon_\infty. \quad (3)$$

The physical meaning of the above equation is that $\varphi$ is created by the polarization charge $\rho_p = \nabla \cdot P$ of the polarization field given by:

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

The term on the r.h.s. of (3) measures the effect of the coupling between the $\varphi$ and $u$ fields on the field equation for $\varphi$. Of course we are working in the quasistatic limit ($c \to \infty$) which is fully justified for the situation under study.

For an isotropic bulk homogeneous medium it is possible to obtain independent equations for $u_L$ and $u_T$ and this yields at once the longitudinal and transverse modes with dispersion relations

$$\omega_L^2 = \omega_{LO}^2 - \beta_L^2 k^2; \quad \omega_T^2 = \omega_{TO}^2 - \beta_T^2 k^2, \quad (5)$$
where $k$ is the 3D wavevector and $\omega_{LO}^2 = \omega_{TO}^2(\epsilon_0/\epsilon_\infty)$. However, our concern is to study the matching of different media at an interface.

Let discuss one interface which we take as the plane $z = 0$. We first Fourier transform in the 2D plane of the interface, so that the $\omega$-dependent vibration amplitudes are of the form

$$u(\rho, z) = \exp(i\kappa \cdot \rho)u(z),$$

where $\kappa$, $\rho$ are 2D vectors (wavevector and position) respectively.

We proceed likewise for $\varphi$ and concentrate on $u(z)$ and $\varphi(z)$. These are $(\omega, \kappa)$-dependent quantities for which, after 2D Fourier transform, we have $(\omega, \kappa)$-dependent differential equations in the independent variable $z$. We stress that in general $u$ consists of $u_L$ and $u_T$ (see, e.g., [8]).

The matching boundary conditions were obtained in [2] and they are, in the variable $z$,

$$u_j(+0) = u_j(-0)$$
$$\varphi(+0) = \varphi(-0)$$
$$\tau_zj(+0) = \tau_zj(-0)$$
$$\epsilon_\infty \frac{d\varphi}{dz} - 4\pi \alpha u_z = \text{continuous}$$

On evaluating these expressions at $z = \pm 0$ we must account for the different values of $\beta_L$, $\beta_T$, $\epsilon_\infty$ and $\alpha$ on both sides of the interface.

We have four amplitudes ($u_x$, $u_y$, $u_z$, $\varphi$), 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

$$F \equiv \begin{bmatrix} F_M \\ F_E \end{bmatrix} \equiv \begin{bmatrix} u \\ \varphi \end{bmatrix}$$

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

$$L \cdot F = 0,$$
where $L$ is a $4 \times 4$ differential matrix which can be readily written down explicitly. Upon 2D Fourier transform $L$ depends on $\kappa$ ($\omega$-dependence understood everywhere) and contains the differential operator $d/dz$. A convenient technique for solving this problem is the Surface Green Function Matching (SGFM) method [19], which, for the case we are interested on, was applied in [9]. In the SGFM method, the key element is the surface projection of the Green function of the system $G_s$. The eigenvalues $\omega(\kappa)$ of the problem can be obtained from an procedure in which we calculate from $G_s$ the $\kappa$-resolved local density of states at $z = 0$ [19]:

$$N_s(\omega, \kappa) = -\frac{1}{\pi} \lim_{\epsilon \to \infty} \Im \text{Sp} \ G_s(\omega + i \epsilon, \kappa).$$

The eigenvalues $\omega(\kappa)$ are then the frequencies at which the peaks in the density of states $N_s$ 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.

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 formulae [20]:

$$\rho(x) = 5.36 - 1.60 x; \ (\text{c.g.s.})$$
$$\epsilon_0(x) = 13.18 - 3.12 x; \ (\text{e.s.u.})$$
$$\epsilon_\infty(x) = 10.89 - 2.73 x; \ (\text{e.s.u.})$$

However, this type of interpolation would not work for $\omega_{LO}$, $\omega_{TO}$, $\beta_L$ and $\beta_T$. It has been strongly argued on the basis of experimental evidence [20] 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 [21]

$$\omega_{LO,GaAs}(x) = 292.37 - 52.83 x + 14.44 x^2;$$
$$\omega_{TO,GaAs}(x) = 268.50 - 5.16 x - 9.36 x^2$$

(12)
Here and henceforth $\omega$ is always given in cm$^{-1}$.

The $\beta_L$ and $\beta_T$ 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. [21].

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 $\omega$ in cm$^{-1}$, $\beta$ is dimensionless and we obtain

$$\beta^2_{L,GaAs}(x) = 2.91 \ (1 - x) \ 10^{-12},$$
$$\beta^2_{T,GaAs}(x) = 3.12 \ (1 - x) \ 10^{-12} \ (13)$$

### III. RESULTS AND CONCLUSIONS

Now we are going to discuss the results obtained by the model considered above. A small imaginary part, equal $10^{-6}$ cm$^{-1}$, was added to $\omega$, so one can ascribe the local modes to very narrow peaks, which show in the calculations instead of the ideal $\delta$-functions. Repeating the calculations for different values of $x$ in $GaAs|Al_{(x)}Ga_{(1-x)}As$ we have observed the concentration of $Al$ to have influence on “behavior” of localized and resonant modes obtained in [9].

We are going to start with the study of the $GaAs|Al_{(x)}Ga_{(1-x)}As$ non-metallized interface. In our figures we present the spectral strength for value of $\kappa = 4 \times 10^6$ cm$^{-1}$. It is seen that no localized mode is present, while the peak corresponding to the resonant mode is present ($\omega \approx 282.6$ cm$^{-1}$). In Fig.1 we can see that the frequency of the resonant mode slowly increases with decreasing $x$ and the peak, at last, disappears for values below $x \approx 0.5$. At the same time, from Fig.2 it can be 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. A similar behavior can be observed for the longitudinal one, thus we omit to show it.
We shall pass now to study the case when the interface is metallized. Here, a very interesting effect can be seen (Fig.3). As it was previously reported [9], in this case the resonant mode moves toward the transverse threshold and is very close to it. But when studying the dependence of its position with Al concentration it appears that, for a very narrow range $0.5292 \geq x \geq 0.5308$ (with $\kappa = 4 \times 10^6 \text{cm}^{-1}$), the corresponding peak in the LDOS splits into two very well resolved peaks located on both sides of the transverse threshold. When the concentration is raised, the left peak disappears (Figs.4a,4b) and the remaining one locates near $\omega_{TO}$ as was obtained in [9]. We do not know any previous experimental or theoretical report on that kind of splitting, which can also be seen for other values of $\kappa$, but with a slightly different range for $x$ (Fig.5). The present calculation does not allow us to give a precise explanation for such an effect. We assume that, regarding the strong electrostatic character of this mode, for those values of $x$, the potential energy at the interface modifies in a way that some kind of “potential well” is present, leading to a situation analogous to the case of a quantum well, where the two degenerate modes corresponding to the two interfaces split into the two interface modes of opposite parities. Of course, here we do not have two interfaces, but only one, and only one of those modes splits. The “confinement” for the oscillations would come from the shape of the potential energy distribution at the interface. Nevertheless, the exact explanation can only be obtained by means of a detailed analysis of that distribution. According to the results we are presenting here it seems that the study of this problem deserves some attention and will be considered elsewhere.

Recently, a new envelope-function theory for phonons in heterostructures [4] shows interesting results for the $\text{InAs}|\text{GaSb}$ system. Even when it appears that the study of multiple-interfaces heterostructures is more attractive looking forward possible applications, 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 occurred in those systems.

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IV. FIGURE CAPTIONS

Fig.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.

Fig.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.

Fig.3: LDOS showing the splitting of the resonant mode at the transverse threshold in a metallized $GaAs|Al(x)Ga_{(1-x)}As$ interface for $\kappa = 4 \times 10^6 \text{cm}^{-1}$.

Fig.4(a): LDOS for a metallized interface. It is seen how one of the peaks (the left one in Fig.3) resulting from the splitting decreases its height with increasing $x$.

Fig.4(b): The continuation of LDOS from Fig.3 for higher values of $x$ showing the disappearance of the left peak ($\kappa = 4 \times 10^6 \text{cm}^{-1}$).

Fig.5: Splitting in the LDOS for a metallized $GaAs|Al(x)Ga_{(1-x)}As$ interface for $\kappa = 2 \times 10^6 \text{cm}^{-1}$.