Optical modes in GaAs-based quantum wells

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This paper describes some results obtained by using a theoretical formulation of a long-wave phenomenological model for polar optical phonons in heterostructures which takes full account of (i) the coupling between the mechanical vibration $\mathbf{u}$ and the electrostatic potential φ, (ii) the fact that $\mathbf{u}$ in general has longitudinal and transverse parts, and (iii) the simultaneous satisfaction of all mechanical and electrostatic matching boundary conditions. The system of simultaneous differential equations thus established is explicitly solved without approximations. Two distinct classes of solutions are obtained. One consists of shear horizontal purely mechanical vibrations. The modes of the other class have mixed character and describe coupled electrical and mechanical vibrations. The results compare very well with experimental evidence and have the correct symmetry pattern.

Polar optical modes play an important role in electron-phonon processes such as scattering rates, polaron effects, and resonant Raman scattering[1–3] in quantum wells and superlattices. Besides microscopic calculations[3] it would be desirable to have a satisfactory phenomenological model to study the long-wave limit, to which experimental evidence is essentially circumscribed. Indeed, considerable theoretical effort has been devoted to this purpose[4–11] with substantial differences among the various proposals. The issue concerns the matching boundary conditions and, as stressed recently,[12] also the mathematical structure of the differential equations which embody the physical model.

A formulation of the basic phenomenological model from which all further developments stem,[13] which takes full account of all the couplings between electrical and mechanical excitations and accounts for all matching boundary conditions, mechanical and electrical, without *ad hoc* assumptions, has been presented elsewhere.[14] On this basis one can study the single heterojunction, where all the essential physics arise, and obtain a satisfactory understanding of the nature and spectral properties of the solution thus obtained.[12] In particular one can then identify the interface modes as essentially electrical in nature, thus explaining why interface modes can be accounted for by the dielectric models which, on the other hand, literally applied fail to explain the experimental symmetry pattern observed in quantum wells.[15] The mechanical vibration amplitude has been treated[16] in a way which is similar in spirit[14] but different in detail as it depends on whether $\Delta\omega_0$—the difference in resonant frequency at zero wavelengths for the two materials—is large or small. When it is large, for instance, the amplitude can be assumed to vanish at some chosen plane. The relation with the model of[16] and of various other models proposed in the literature is discussed in Refs. 12 and 14. Here we present some results obtained when the formulation presented in Ref. 14 and applied in Ref. 12 is used to study a quantum well.

The problem is to solve a system of four coupled differential equations for three mechanical vibration amplitudes (u_x, u_y, u_z) and one "electrical amplitude" φ, that is, the electrostatic potential, for heterostructures. We choose the geometry so that z is normal to the surfaces and $\boldsymbol{\kappa}$, the two-dimensional wave vector parallel to the surfaces, is in the y direction. One shear horizontal mode with amplitude $u_x(z)\exp(i\kappa y)$, purely transverse and mechanical, propagating in the y direction is then decoupled from the rest. Thus the problem is to study a field consisting of excitations in which the three amplitudes (u_y, u_z, φ) are coupled and the normal modes of this field have mixed electrical and mechanical character. Let $\mathbf{u}$ denote the vector (u_y, u_z). This in general consists of a longitudinal part $\mathbf{u}_L(\boldsymbol{\nabla}\times\mathbf{u}_L = 0)$ and a transverse part $\mathbf{u}_T(\boldsymbol{\nabla}\cdot\mathbf{u}_T = 0)$. The matching boundary conditions couple $\mathbf{u}_T$ to $\mathbf{u}_L$, which is coupled to φ. Thus the electrical excitation φ drives the full mechanical excitation $\mathbf{u}$ and vice versa.

A convenient way to study this problem, which allows for an easy separate identification of the electrical and mechanical spectral strength of the normal modes, is provided by the surface Green-function matching (SGFM) method, which has been described in detail elsewhere.[16] This yields the full Green function $\mathbf{G}_s$ of the matched system in terms of the bulk Green functions of the constituent media. Knowing $\mathbf{G}_s$, from this one can obtain the dispersion relations $\omega(\boldsymbol{\kappa})$ for the normal modes of the system and all spectral functions of interest. The SGFM analysis of the matching problem at one interface[12] is readily extended to the simultaneous matching at two coupled interfaces at finite distance.[17] The algebra for the present calculations was carried out by means of MATHEMATICA,[18] with which closed-form expressions are obtained for the elements of the full Green function. 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 Ref. 19. If we study the matching to GaAs then we 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.[18] The β_L and β_T parameters giving the quadratic dispersion for the bulk modes were estimated from the experimental curves of Ref. 20.

We consider a GaAs well of width $d = 20$ Å with $Al_{0.9}Ga_{0.1}As$ outside. Figure 1 shows the dispersion relations for the GaAs eigenmodes. Since all the modes are different eigenmodes of the same eigenvalue problem we can label them by a discrete label $m = 1, 2, \ldots$. We adscribe $m = 1$ to the highest mode, so m grows on going downwards in frequency. We have concentrated mainly in the range of κ from zero to values of order 10^7 cm^{-1}, which represent a very small fraction of the Brillouin zone and yet span the range of physical interest. We stress that the labeling of the eigenmodes by increasing m on moving downwards holds for all κ, either before or after the crossovers seen in the figure. The frequency eigenvalues are in fairly good agreement with experimental data,[15] especially in their spacings, with only a small systematic difference in the absolute value, which could be easily accounted for by a small inaccuracy in the determination of the well width.

Before discussing the details of the crossover of modes 1 and 2 in Fig. 1, it is interesting to see the physical nature of the normal modes in this range and we note that for $\kappa < \kappa_c$ (the crossover value of κ) the first mode is the more dispersive, while for $\kappa > \kappa_c$ the first mode is almost flat and the more dispersive branch has become the mode $m = 2$. The clearest way to characterize these modes is to separate out from the local density of states the spectral strengths corresponding to the electrical and mechanical excitations. In the present calculation this is done by separating out the contribution of the fourth diagonal element of the Green function $\mathbf{G}_s$. Figure 2 shows the results for the first two modes in the neighborhood of κ_c. Electrical and mechanical spectral strengths are given for $\kappa = 1.1 \times 10^6$ $cm^{-1} < \kappa_c$ and $\kappa = 1.4 \times 10^6$ $cm^{-1} > \kappa_c$. For $\kappa < \kappa_c$ [Fig. 2(a)] the electrical spectral strength is practically all in the first mode, while the mechanical spectral strength [Fig. 2(b)] is practically all in the second mode. Thus the first mode for very low values of κ consists mostly of an electrical excitation, while the second mode is mostly mechanical in origin, although both have, qualitatively speaking, a mixed character. This is transferred between the two modes for $\kappa > \kappa_c$, where it is now the first mode that is mostly mechanical [Fig. 2(d)] while the second mode is mostly electrical [Fig. 2(c)].

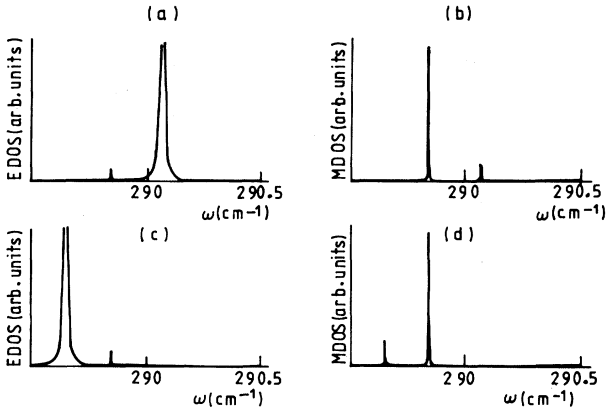

FIG. 2. (a) and (c) Electrical and (b) and (d) mechanical spectral strengths for the upper and lower modes in the immediate neighborhood of the crossover between modes $m = 1$ and $m = 2$. (a) and (b) $\kappa = 1.1 \times 10^6$ cm^{-1} and (c) and (d) $\kappa = 1.4 \times 10^6$ cm^{-1}. Spectral strengths in arbitrary units and ω in cm^{-1}.

Figure 3 shows the spatial dependence of u_z and φ for the first three modes when $\kappa = 2 \times 10^6$ $cm^{-1} > \kappa_c$. There is a nonvanishing amplitude u_y, which is never-

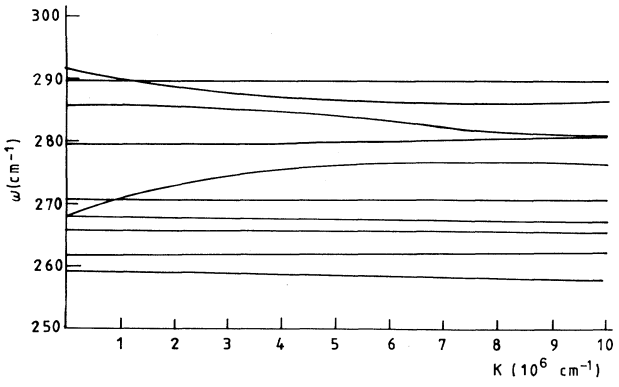

FIG. 1. Dispersion relations for the GaAs eigenmodes (in cm^{-1}) and varying κ (in 10^6 cm^{-1}). Note the splitting of the degeneracy of the modes $m = 6$ and 7 for $\kappa \neq 0$.

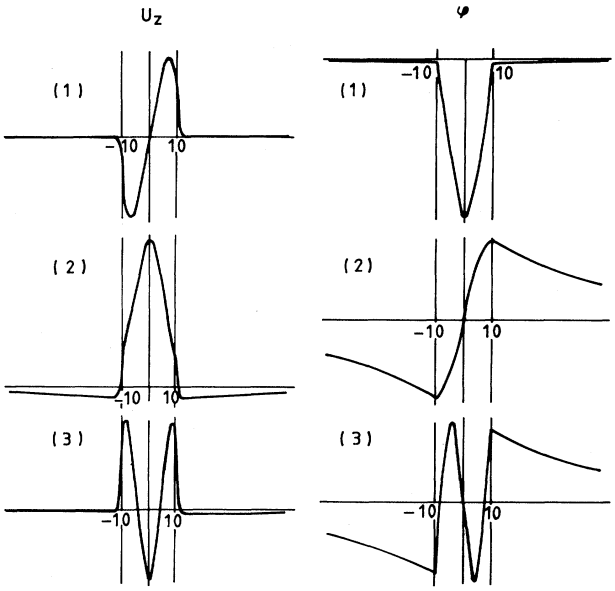

FIG. 3. Spatial dependence of the mechanical (u_z, left-hand side) and electrical (φ, right-hand side) amplitudes for $\kappa = 2 \times 10^6$ cm^{-1}, modes $m = 1, 2, 3$. The mode number is indicated in brackets and the abscissa z is in angstroms.

theless much smaller and is not displayed. These modes are no longer strictly longitudinal, as expected for $\kappa \neq 0$ when matching at interfaces is involved. However, explicit evaluation shows that $|\nabla \times \mathbf{u}|$ is still much smaller than $|\nabla \cdot \mathbf{u}|$, so these modes are still in practice quasi-L for this low value of κ.

Figures 2 and 3 show the transfer of character which takes place at the crossover of the first two modes. Both the parity of φ and the predominance of electrical or mechanical character are interchanged. If E/O denotes even/odd z dependence, then the parity sequence for φ_1 to φ_5 is O,E,O,E,O when $\kappa < \kappa_c$ and it changes to E,O,O,E,O when $\kappa > \kappa_c$.

Returning now to Fig. 1 we note that starting from $\kappa = 0$ there are two very dispersive modes which eventually cut across the rest of the spectrum. The lower one ($m = 6$) originates the second crossover with mode $m = 5$ for low κ. On looking at the entire system of eigenmode branches there are two, for every κ, which have predominantly electrical character. In the upper part of the spectrum this is $m = 1$ for $\kappa < \kappa_c$ and $m = 2$ for $\kappa > \kappa_c$, as has just been discussed and illustrated in Fig. 2. A similar situation holds about the lowest crossover, between modes 5 and 6, among which the electrical and mechanical spectral strengths are also transferred in the manner of Fig. 2.

It is in order to remark that while the physical system under study—the quantum well—is symmetric, the differential system under study is not invariant under the reflection $z \to -z$. The vibration amplitude u_x is decoupled from the other amplitudes, thus only u_z, u_y, and φ are related. It follows from the field equations that $u_y(z)$ and $\varphi(z)$ have always the parity opposite to that of $u_z(z)$, a fact which also shows in Fig. 2. Thus we can still compare the parities of different modes on the understanding that this refers only to one of the amplitudes. More specifically, we shall refer only to the parity of $\varphi(z)$. In fact, for $\kappa \neq 0$ the L and T polarizations are all in principle coupled in all the normal mode solutions. Consequently, mode mixing also takes place between modes 1 and 2, on one hand, and 5 and 6, on the other hand. The transfer of character which takes place at these crossovers occurs very sharply and requires working with careful accuracy in these frequency ranges. In practice the situation is significantly different in the crossover between modes 5 and 6, due to the fact that at $\kappa = 0$ mode 5 is totally L and mode 6 is totally T. In the crossover a strong mixing of the L and T polarizations takes place, verified in our results by evaluation of $|\nabla \times \mathbf{u}|$ and $|\nabla \cdot \mathbf{u}|$, which are then comparable. On moving backwards from κ_c towards $\kappa \to 0$ we find that the mixture of L and T character is still significant, so that even below κ_c mode 5 soon ceases to be quasi-L and mode 6 to be quasi-T.

Finally, the model also yields the transverse modes $m = 7,8,9,10$. These are strictly T for all κ, as they have only the amplitude u_x, which is factored out in the full 4×4 determinant. These modes are purely mechanical and correspond to the shear horizontal wave one always encounters in the theory of elastic surface waves.[17] The parity sequence of $u_x(z)$ for these modes is $E,O,E,\ldots$.

Thus, inasmuch as one accepts a phenomenological model to study long waves, we have obtained a satisfactory solution which satisfies all simultaneous matching boundary conditions without conflict and appears to compare well with experimental evidence and provides a simple basis for the study of electron-phonon interaction in heterostructures.

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