Analysis of the phenomenological models for long-wavelength polar optical modes in semiconductor layered systems

C. Trallero-Giner*
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Federal Republic of Germany

F. García-Moliner and V.R. Velasco
Instituto de Ciencia de Materiales, Consejo Superior de Investigaciones Cientificas, Serrano 123, E-28006 Madrid, Spain

M. Cardona
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Federal Republic of Germany
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The standard type of phenomenological model for this problem consists of a mechanical field equation for the vibration amplitude and—in the quasistatic limit—Poisson's equation. The critical issue concerns the matching boundary conditions. The apparent incompatibility between mechanical and electrostatic boundary conditions often appearing in the literature is analyzed and clarified. It is shown how the full solution can be obtained so that there is no incompatibility and the key features of the results, notably the symmetry pattern, agree with microscopic calculations and with Raman-scattering data.

Considerable attention is being devoted to the study of long-wavelength polar optical modes in semiconductor quantum wells and superlattices. Various theoretical models have been proposed[1–10] that represent substantially different and often diverging viewpoints and have met with varying degrees of success in the interpretation of observed experimental facts. Raman-scattering data are available for a wide range of superlattices, especially GaAs related, and show the existence of different types of modes. Some of these modes have amplitudes mainly concentrated in one of the constituent slabs, usually termed confined modes[11–13] and some with the amplitudes spreading to both constituents but tending to concentrate close to the interfaces, hence called interface modes.[14,15] The Raman experiments do not give the spatial dependence of the amplitudes, but this is surmised from the fact that the observed frequencies are forbidden in one of the constituent materials (guided or confined modes) or in both (interface modes, in this case they are induced by interface disorder). Moreover the geometry of the Raman-scattering experiment can be chosen[16] to detect modes with different symmetries. In particular, one can then experimentally establish whether the amplitude (or the potential) of the observed modes is even or odd with respect to the center of the slabs.[12] These aspects of the experimental data and the key features of the results obtained in microscopic calculations[3,4,6] should be taken as the criterion to decide whether a phenomenological model is reasonable or not, while some lack of quantitative accuracy can, to some extent, be expected and tolerated.

The central issue can be introduced by considering just one interface. The essence of a long-wavelength phenomenological model is that one uses differential calculus and then a way must be found to match the solutions at the interface. Of course it can be argued[6,10] that in principle the differential equations are not valid in the immediate vicinity of the interface, when finite changes take place over microscopic distances. Any phenomenological model of the matching problem must necessarily be an approximation, just as it is in all the existing matching calculations for long-wavelength acoustic or piezoelectric modes. Differential calculus is used to match at abrupt interfaces and the model works quite well. The same can be said, for instance, of envelope-function matching calculations for electronic states.[17]

The question raised here is not whether the standard type of phenomenological model can be *rigorously* justified. It is clear that formally it cannot. The question is whether one can start from a simple phenomenological model and obtain a solution which has all the general properties one may require and reproduces to a good approximation experimental results. In this paper we show that this can be done.

The key issue concerns the matching boundary conditions. One can approach the problem with a dielectric model[1] and impose electrostatic continuity. The pattern of vibrational amplitudes and electrostatic potential one obtains is in open disagreement with experimental evidence[11–15] and with microscopic calculations.[3,4,6–8] Moreover, one achieves electrostatic continuity at the expense of mechanical discontinuity. Alternatively, one can start from a mechanical equation of motion for the vibration amplitude[2] and then impose mechanical continuity. This yields the correct symmetry pattern for the vibration amplitudes, but it appears to produce a discontinuous electrostatic potential. The incompatibility between the separate use of these two approaches has been correctly stressed.[13,14] Our purpose is to show that both requirements—mechanical and electrostatic continuity—

can be satisfied by combining the two approaches in a proper way which accounts for the coupling between the mechanical and electrostatic fields.

One possibility is to start from a phenomenological model for the vibrational wave and then introduce *ad hoc* matching boundary conditions which force the electrostatic continuity into the analysis.[6,18] The idea is to save the simplicity of the phenomenological model, with parameters determined from a fit to bulk phonon-dispersion relations, for use in problems of practical interest like the derivation of an electron-phonon interaction Hamiltonian and the calculation of Raman-scattering efficiencies.

A different approach has been taken[5] in a study of this problem for a model in which the vibrations are totally confined by infinitely rigid barriers. The mechanical amplitudes then vanish at the boundaries but not necessarily the electrostatic potential which is continuous. This model describes well the confined modes—which are *totally* confined due to the rigid barrier condition—and gives correctly the observed symmetries, but it is limited by the approximation inherent to it and cannot describe interface modes. It is the simultaneous account of the mechanical and electrostatic field and the corresponding, and compatible, boundary conditions that makes this approach significantly different from the others. We shall now use this approach but relax the restriction to rigid barriers.

We accept from the start a phenomenological model based on an equation of motion for the vibration amplitude $\mathbf{u}$ which contains (i) spatial dispersion through terms representing mechanical forces, (ii) a natural frequency for the harmonic-oscillator–like vibrations, and (iii) the coupling to the electrostatic field $\mathbf{E}$.[19] This is the well-known equation[2,5]

$$\rho(\omega^2-\omega_{\mathrm{TO}}^2)\mathbf{u}-\boldsymbol{\nabla}(\rho v_a^2\boldsymbol{\nabla}\cdot\mathbf{u})$$

$$-\nabla\cdot(\rho v_b^2\nabla\mathbf{u})+\left(\frac{(\varepsilon_0-\varepsilon_\infty)}{4\pi}\right)^{1/2}\omega_{\mathrm{TO}}\mathbf{E}=0\ , \quad (1)$$

where ρ is the mass density, ε_0 and ε_∞ are the static and high-frequency dielectric constant, and ω_{TO} is the bulk transverse-optical frequency. The ansatz for the mechanical forces follows from general considerations[2,20] and has *the form* of the divergence of a tensor $\overleftrightarrow{\tau}$. In the case of an isotropic solid, assumed implicitly in (1),

$$\overleftrightarrow{\tau}=\rho v_a^2\nabla\cdot\mathbf{u}\ \overleftrightarrow{\mathrm{I}}+\rho v_b^2\nabla\mathbf{u}. \quad (2)$$

In the case of acoustic waves ($\omega_{\mathrm{TO}}=0$) $\overleftrightarrow{\tau}$ is literally the mechanical stress tensor, with v_a^2 and v_b^2 related to the Lamé coefficients λ, μ. In the present case v_a^2 and v_b^2 are phenomenological parameters which take into account the spatial dispersion, i.e., the k dependence of the bulk frequencies up to terms of order k^2. The essence of the phenomenological model is that (1) it is accepted as it stands, even for inhomogeneous systems where the material parameters depend on the position coordinate z normal to the interface, and this includes abrupt interfaces. Having assumed this model we want to see how the mechanical and electrostatic field can be obtained while their coupling is fully taken into account.

For this (1) must be *solved simultaneously* with Maxwell's equations which, in the quasistatic limit here considered ($c\to\infty$) or, from an experimental point of view, for wavelengths small compared with the bulk reststrahlen wavelengths, are reduced to the equations of electrostatics. In this case

$$\mathbf{E}=-\nabla\Phi \quad (3)$$

and, since there is no external free charge

$$\nabla\cdot\mathbf{D}=0\ . \quad (4)$$

However, written in this way, the coupling between the two fields is not apparent. From the general relationship

$$\mathbf{D}=\mathbf{E}+4\pi\mathbf{P} \quad (5)$$

and the particular constitutive relationship of this model[19]

$$\mathbf{P}=\left(\frac{\rho(\varepsilon_0-\varepsilon_\infty)}{4\pi}\right)^{1/2}\omega_{\mathrm{TO}}\mathbf{u}+\frac{(\varepsilon_\infty-1)}{4\pi}\mathbf{E}, \quad (6)$$

we make the coupling of the mechanical and electrical fields explicitly appear in the electrostatic field equation by noting that the source of Φ is the polarization charge, so that

$$\nabla^2\Phi=4\pi\boldsymbol{\nabla}\cdot\mathbf{P} \quad (7)$$

which, using the above expressions and the Lyddane-Sachs-Teller relations, yields

$$\nabla^2\Phi=\left[4\pi\rho\left(\varepsilon_\infty^{-1}-\varepsilon_0^{-1}\right)\right]^{1/2}\omega_{\mathrm{LO}}\nabla\cdot\mathbf{u}\ . \quad (8)$$

Furthermore, substituting from (3) in (1)

$$\rho\left(\omega^2-\omega_{\mathrm{TO}}^2\right)\mathbf{u}-\nabla\left(\rho v_a^2\nabla\cdot\mathbf{u}\right)$$

$$-\nabla\cdot(\rho v_b^2\nabla\mathbf{u})-\left(\frac{(\varepsilon_0-\varepsilon_\infty)}{4\pi}\right)^{1/2}\omega_{\mathrm{TO}}\nabla\Phi=0\ . \quad (9)$$

The coupling between the $\mathbf{u}$ and Φ is now explicitly written into the field equations.

We stress that $\mathbf{u}$ in general consists of a longitudinal and a transverse field

$$\mathbf{u}=\mathbf{u}_L+\mathbf{u}_T \quad (10)$$

with $\boldsymbol{\nabla}\times\mathbf{u}_L=0$ and $\boldsymbol{\nabla}\cdot\mathbf{u}_T=0$. Only the $\mathbf{u}_L$ part of $\mathbf{u}$ appears explicitly in (8), but this $\mathbf{u}_L$ is affected *in general* by the coupling to $\mathbf{u}_T$. This is another aspect which is often obscure in the literature. In a bulk homogeneous medium described by this model the longitudinal and transverse modes decouple and one easily obtains from (8) and (9) independent dispersion relations for the bulk LO and TO modes. It is the boundary conditions that, in general, couple the longitudinal and transverse parts, as is common experience with problems involving matching between *isotropic* media. It suffices to study ordinary Rayleigh surface waves[20] to see this. In some cases one can decouple them, notably with some geom-

etry specifically designed for a Raman-scattering experiment which detects modes vibrating in some symmetry direction and with $\vec{\kappa} = \mathbf{0}$, $\vec{\kappa}$ being the two-dimensional (2D) wave vector parallel to the surface. In other situations one can make an *approximation*, for example, in the study of electron-phonon scattering, which involves only the $\mathbf{u}_L$ part of $\mathbf{u}$ and is such that only very low wave vectors have an appreciable weight.[4]

However, the issues raised here can be clarified without need to decouple LO and TO modes. What we have in (9) is a field equation for the total $\mathbf{u}$ given that the $\mathbf{u}_L$ part of it is coupled to Φ, while (8) says that Φ is the potential created by the polarization charge

$$\rho_{\rm pol} = -\left(\frac{\rho}{4\pi}\left(\varepsilon_\infty^{-1} - \varepsilon_0^{-1}\right)\right)^{1/2} \omega_{\rm LO} \nabla \cdot \mathbf{u} \,, \tag{11}$$

where $\mathbf{u}$ is the solution of the mechanical field equation (9). Seen from the point of view of electron-phonon coupling this is the potential that the external test charge of an electron sees in a system where the vibration $\mathbf{u}$ produces a polarization charge $\rho_{\rm pol}$ given by (11). Hence, it is very important to obtain a full solution which simultaneously describes both $\mathbf{u}$ and Φ correctly which, according to the laws of electrostatics, requires continuity of Φ everywhere. This is guaranteed by the phenomenological model embodied in (8) and (9). The moment we accept differential equations to describe the amplitudes $\mathbf{u}$ and Φ these must be continuous at the matching interface:

$$\mathbf{u}(z = +0) = \mathbf{u}(z = -0); \quad \Phi(z = +0) = \Phi(z = -0). \tag{12}$$

We Fourier transform in 2D, thus introducing a dependence on the transverse $\vec{\kappa}$, and are left with differential equations in the position variable z. We now integrate (9) from $-\eta$ to $+\eta$ and let $\eta \to 0$. Since Φ is continuous this yields matching boundary conditions involving the *first derivatives* of the components of $\mathbf{u}$. If we write the force per unit volume in the form of the divergence of the tensor written down in (2), then these conditions take the form

$$\tau_{zj}(z = +0) = \tau_{zj}(z = -0), \quad (j = x, y, z). \tag{13}$$

In the case of acoustic waves this expresses the continuity of the forces per unit area transmitted across the surface. In the present problem one can interpret this in the same way, always within the frame of the approximation, embodied in the model. The z component of (13) reads

$$\rho\left(v_a^2 + v_b^2\right) \left.\frac{\partial u_z}{\partial z}\right|_{z=+0} = \rho\left(v_a^2 + v_b^2\right) \left.\frac{\partial u_z}{\partial z}\right|_{z=-0} \tag{14}$$

which one can in the same spirit interpret as the continuity of the hydrostatic pressure[2] although the present analysis shows that this does not require the decoupling of $\mathbf{u}_L$ and $\mathbf{u}_T$ in the approximation one may choose to call hydrodynamic. All that matters is that given (9) one obtains mathematically the three continuity conditions of (13). The explicit form of these is readily obtained from (9). Note that Eq. (14) can be mapped onto the Bastard boundary condition[17] by replacing the $\rho\left(v_a^2 + v_b^2\right)$ by $1/m$, m being the effective mass.

It is also easily seen by using (6) that integration of (8) from $-\eta$ to $+\eta$, with $\eta \to 0$, yields

$$D_z(z = +0) = D_z(z = -0), \tag{15}$$

as expected from general principles of electrostatics. What we stress is that the matching boundary conditions (13) and (15) follow mathematically and unambiguously once the phenomenological model has been accepted.

The issue initially raised has been formally settled. In (8) and (9) we have a system of four second-order differential equations. We need eight matching boundary conditions. These are (12), (13), and (15). This defines an eigenvalue problem whose solution is uniquely determined by the four differential equations and the eight independent matching boundary conditions. The physical interpretation of these conditions and the approximations embodied in the phenomenological model are a separate issue. Mathematically the problem is uniquely and correctly specified while the coupling between the $\mathbf{u}$ and $\mathbf{E}$ fields is evident and explicit in the analysis.

The confusion arises when one decouples $\mathbf{u}_L$ and $\mathbf{u}_T$ and the two field equations. If $\mathbf{u}$ is only $\mathbf{u}_L$, then $\mathbf{P}$ (6) is only $\mathbf{P}_L$, so $\mathbf{D}$ is only $\mathbf{D}_L$ and its curl vanishes. Since its divergence also vanishes (4), then $\mathbf{D}$ vanishes, which yields

$$\mathbf{E} = -\left[4\pi\rho\left(\varepsilon_\infty^{-1} - \varepsilon_0^{-1}\right)\right]^{1/2} \omega_{\rm LO}\mathbf{u} \,. \tag{16}$$

Substituting this in (9) one obtains a field equation for $\mathbf{u}$ only. The other field equation is often taken to be (4) which involves the dispersive dielectric functions. However, the two field equations are apparently decoupled and this has led to their independent use as optional alternatives. The contradiction arises because the $\mathbf{E}$ and $\mathbf{u}$ fields are really coupled: they must always fulfill (16). This is what causes the apparent incompatibility of boundary conditions: if one chooses to work from the mechanical equation, imposes mechanical continuity, and uses (16) and (3) to obtain Φ by inspection, then Φ is discontinuous. The point is that the correct way to solve the full problem is not to do this but to determine Φ by solving (8) with correct electrostatic matching boundary conditions even when one studies a purely longitudinal field. The physical implications of this for the electron-phonon interaction has just been stressed.

The main objection one can make to the procedure just followed is the assumption of isotropy in (1). Cubic crystals, for instance, are known to be anisotropic to second order in the components of $\mathbf{k}$. It is possible to generalize (2) so as to include this anisotropy. This can be easily done by replacing (1) by

$$\rho\left(\omega^2 - \omega_{\rm TO}^2\right)\mathbf{u} - \nabla \cdot \overleftrightarrow{\tau} - \left(\frac{(\varepsilon_0 - \varepsilon_\infty)}{4\pi}\right)^{1/2} \omega_{\rm TO} \nabla\Phi = 0, \tag{17}$$

where for the tensor $\overleftrightarrow{\tau}$ we use instead of (2) (in matrix notation)

$$\overleftrightarrow{\tau} = \rho \begin{pmatrix} v_a^2 \nabla \cdot \mathbf{u} & (v_c^2 - v_a^2)\frac{\partial u_y}{\partial y} & (v_c^2 - v_a^2)\frac{\partial u_z}{\partial z} \\ (v_c^2 - v_a^2)\frac{\partial u_x}{\partial x} & v_a^2 \nabla \cdot \mathbf{u} & (v_c^2 - v_a^2)\frac{\partial u_z}{\partial z} \\ (v_c^2 - v_a^2)\frac{\partial u_x}{\partial x} & (v_c^2 - v_a^2)\frac{\partial u_y}{\partial y} & v_a^2 \nabla \cdot \mathbf{u} \end{pmatrix} + \rho v_b^2 \nabla \mathbf{u}. \quad (18)$$

In this case the v_a^2, v_b^2, v_c^2 parameters are obtained fitting the corresponding bulk phonon-dispersion relation [$v_c = v_a$ leads back to the isotropic case of Eq. (2)].

The mechanical vibration amplitude has been treated[21] in a way which is similar in spirit to the analysis proposed here, although it differs in detail. A dynamical matrix is written down inspired in the form of the effective Hamiltonian for envelope-function calculations of electronic structures. Due to crystal symmetry the mathematical form is the same as we obtain from mechanical considerations. The parameters of this dynamical matrix are found by fitting to the results of a microscopic calculation based on a rigid-ion model, which gives a correct description of the optical vibrations. The discussion is then centered on the equation of motion for the mechanical vibration amplitude and the corresponding matching boundary conditions, for which two extreme cases are studied. These correspond to $\Delta\omega_0$—the difference in resonant frequency at zero wave vector—being either much larger or much smaller than the bulk bandwidths. In the first case the amplitude is assumed to vanish at some chosen plane. In the second case, after some plausible approximations a kind of continuity equation is derived which expresses energy conservation and this is used to establish matching boundary conditions.

Although this is close in spirit to our present analysis, it differs in two important respects, namely (i) Our model does not depend on whether $\Delta\omega_0$ is large or small and thus can be directly used for $GaAs/Al_xGa_{1-x}As$ systems with arbitrary x; (ii) we discuss explicitly the differential equation of Φ, its coupling to the mechanical differential equation, and the compatibility between the matching boundary conditions for both fields. This also allows for an explicit study of the electron-phonon interaction Hamiltonian in which one can easily see how to introduce screening due to the electron gas if one has a model for its dielectric function. This is a very important issue for transport calculations in populated quantum wells with external modulation doping.

Finally it is interesting to consider the particular case of a GaAs well with AlAs barriers using the method just presented. In this case the frequency difference is very large and it is a good approximation to assume infinitely rigid barriers. That is $\mathbf{u}(z = \pm d/2) = 0$ and $\mathbf{u} \equiv 0$ for $|z| > d/2$, d being the well width.

We consider the case of (001) interfaces and vibrations in the (001) direction with $\vec{\kappa} = 0$, where the transverse part $\mathbf{u}_T \equiv 0$ for all $z \in (-d/2, d/2)$, we only need to study a longitudinal field. From the vanishing of $\nabla \times \mathbf{u}$ one can reduce the mechanical field equation to one differential equation for u_z with the amplitude vanishing for $|z| \geq d/2$. Equation (8) now reads

$$\frac{d^2\Phi}{dz^2} = \left[4\pi\rho\left(\varepsilon_\infty^{-1} - \varepsilon_0^{-1}\right)\right]^{1/2} \omega_{LO} \frac{du_z}{dz} \quad (19)$$

with Φ continuous. The condition (15) is in this case automatically satisfied. The full solutions of Eqs. (9) and (19) are[5]

$$u_z(z) = A_n \sin\left(\frac{n\pi}{d}(z + d/2)\right); \quad n = 1, 2, \ldots \quad (20)$$

and

$$\Phi_n(z) = C_n \begin{cases} [1 - (-1)^n], & z < -d/2 \\ 2\cos\left[\left(z + \frac{d}{2}\right)\frac{n\pi}{d}\right] - [1 + (-1)^n], & |z| < d/2 \\ [(-1)^n - 1], & z > d/2, \end{cases} \quad (21)$$

where

$$C_n = A_n \frac{\left[4\pi\rho\left(\varepsilon_\infty^{-1} - \varepsilon_0^{-1}\right)\right]^{1/2} \omega_{LO}}{(n\pi/d)} \quad (22)$$

and A_n is a normalization constant. These results agree rather well with those of microscopic calculations[3,4,6,8] and there is no incompatibility. The solution yields a continuous electrostatic potential and the symmetry pattern agrees with experimental evidence. Raman-scattering data[12,14,15] detect LO modes with Φ odd—corresponding to n odd in (21)—when the geometry of the experiment is $\bar{Z}(Y, X)Z$ and Φ even—n even—when it is $\bar{Z}(X, X)Z$ and this pattern is what one finds from (21) and from microscopic calculations.

Studies based on the so-called dielectric model use the boundary condition (15) and impose the vanishing of Φ for $|z| \geq d/2$. This yields

$$\Phi(z) \sim \begin{cases} \cos\left(\frac{n\pi}{d}z\right); & n = 1, 3, \ldots \\ \sin\left(\frac{n\pi}{d}z\right); & n = 2, 4, \ldots, \end{cases} \quad (23)$$

which gives the opposite symmetry pattern. This happens at the expense of mechanical discontinuity. On the contrary, calculations based on a decoupled field equation for $\mathbf{u}$ and the vanishing of E_z for $|z| \geq d/2$ yield the correct vibration amplitudes but then Φ is discontinuous. An *ad hoc* way out of this dilemma can be found[18] in which Φ is forced to be continuous by adding in each layer a term independent of z and dependent of n. These constants are exactly the terms involving $(-1)^n$ contained in (21), which is the solution naturally obtained without need to resort to any artifact when the mechanical and electrostatic field equations are correctly coupled and solved.

This clarifies the issue raised at the beginning. The mechanical field equation is only a model and hence an approximation, but it works quite well for the vibration amplitudes, while Φ must be obtained from (8). This provides the basis for obtaining in a rather simple way an electron-phonon-interaction Hamiltonian based on a reasonable model which works rather well in the cases in which it has been tested and leading to solutions which meet all general requirements with no incompatibility between the mechanical and electrostatic matching boundary conditions. Further work on this problem is currently in progress.

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*Permanent address: Department of Theoretical Physics, Havana University, Havana, Cuba.

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