Continuum theories of optical phonons and polaritons in superlattices: A brief critique

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The controversy concerning the confinement of optical modes in superlattices concerns the use of electromagnetic (EM) or hydrodynamic (HD) boundary conditions and the roles of dispersion and retardation in continuum theories, and how continuum theories relate to microscopic models. In this paper we describe EM and HD continuum models and discuss recent attempts to reconcile the EM model with microscopic theory. We conclude that the lack of retardation and dispersion in the EM model results in a confusion between LO modes and polaritons. In particular, we question the attribution of a LO-type Fröhlich scalar potential to Fuchs-Kliewer interface polaritons.

I. INTRODUCTION

In recent years the study of Raman scattering in quantum wells and superlattices composed of semiconductors has revealed the existence of guided optical-phonon modes.[1–3] The confinement of optical vibrational modes is to be expected in all systems whenever the frequency bands in the adjacent materials do not overlap. The detailed description of this confinement is by no means fully understood and is currently controversial regarding the validity of continuum models. The development of a valid description in terms of a continuum model is especially important for understanding how confinement affects the electron-phonon interaction, the description of the latter in terms of microscopic models based on linear chains of atoms being impractical in terms of computational complexity. The controversy centers on the choice of boundary conditions for the longitudinally polarized optical (LO) modes, on the ability of LO modes to mix with transversely polarized optical (TO) modes, and on the importance of dispersion. Our purpose here is to attempt a clarification of some of these issues and to discuss some of the problems which attend the relating of continuum modes to microscopic modes.

It is useful to have in mind a concrete example and we take the one most studied experimentally and theoretically, namely the GaAs/AlAs quantum-well system in which the disparity of the frequency bands for optical modes is total. All microscopic models[4–6] agree that the relevant ionic displacement u must be zero at or very close to the interface. We take the GaAs slab to be of thickness L and the z direction to coincide with the normal to the plane of the layers so that $0 \leq z \leq L$ defines the quantum well. To a good approximation microscopic theory suggests that, for LO modes, the corresponding macroscopic envelope function is [Fig. 1(a)]

$$u_z \propto \sin q_z z, \quad 0 \leq z \leq L$$
$$q_z = \frac{n\pi}{L}, \quad n = 1,2,3,\ldots \quad (1)$$

For LO modes $\nabla \times \mathbf{u} = 0$, whence the in-plane displacement $\mathbf{u}_\parallel$ is of the form

$$\mathbf{u}_\parallel \propto \cos q_z z \ , \quad (2)$$

which has antinodes at the interface. The electric field associated with the ionic polarization is proportional to displacement and thus has components with the z dependence depicted in Eqs. (1) and (2). It follows that the scalar potential associated with this field is of the form

$$\phi \propto \cos q_z z \ , \quad (3)$$

which has maxima at the interfaces.

The symmetry of the potential is of importance in determining selection rules for the electron-phonon interaction whose strength is just $e\phi$. For intrasubband transitions the initial and final electron wave functions have, of course, the same parity, and so ϕ must have even parity. This means that the interaction is only with

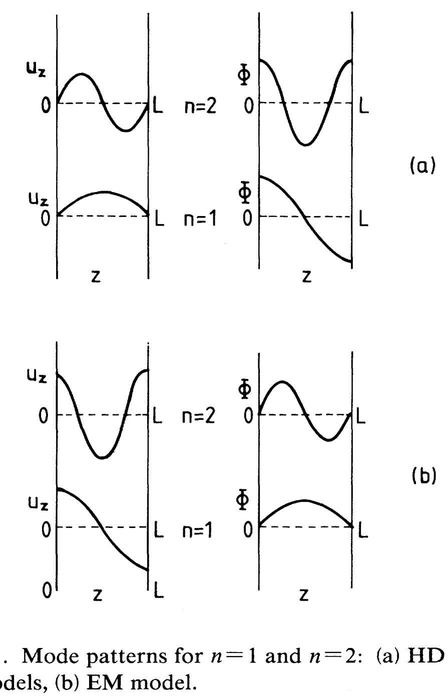

FIG. 1. Mode patterns for $n=1$ and $n=2$: (a) HD and linear chain models, (b) EM model.

modes with n even. For scattering between adjacent subbands the reverse is true, only modes with n odd contributing. The electron-phonon interaction involving intrasubband transitions has been probed by experiments on Raman scattering, and the above expectations have been confirmed.[2]

The most widely used macroscopic model is that based on the treatment of slab modes in an ionic film by Fuchs and Kliewer.[7] The characteristic features of this approach are to regard the crystal as a uniform, isotropic dielectric continuum, to neglect dispersion, and to use standard electromagnetic connection rules at the interfaces. These rules entail the continuity of the scalar potential (or, what is the same thing, of the tangential components of the electric field), of the normal components of displacement and magnetic induction, and of the tangential component of the magnetic field. For this reason we refer to this treatment as the electromagnetic (EM) model.

Confinement in the EM model is seen to occur as a result of the dielectric discontinuity between the two adjacent layers. The dielectric function is given by

$$\epsilon(\omega)=\epsilon_{\infty}\frac{\omega^2-\omega_L^2}{\omega^2-\omega_T^2}\ , \tag{4}$$

where ϵ_{∞} is the high-frequency permittivity, and ω_L and ω_T are the LO and TO angular frequencies, and it takes the value zero, because for LO modes $\omega=\omega_L$: since LO phonons in AlAs have an entirely different frequency, $\epsilon(\omega)\neq 0$ in AlAs. Because D_z, the normal component of the electric displacement, is continuous, it must be zero in AlAs, since it is zero in GaAs. Thus the electric-field component $E_z=0$ in AlAs, but there is no necessity for E_z to be continuous. Because $E_z=0$, then $\phi=0$ in AlAs, and since ϕ must be continuous,

$$\phi\propto\sin q_z z,\quad q_z=\frac{n\pi}{L},\quad n=1,2,3,\ldots \tag{5}$$

in GaAs, from whence it follows that

$$u_z\propto\cos q_z z,\quad u_{\parallel}\propto\sin q_z z\ . \tag{6}$$

These results are quite opposite to those obtained by microscopic theory [Fig. 1(b)]. They predict intrasubband scattering by odd n modes and intersubband scattering by even n modes.

There is a second macroscopic model, however, which mirrors microscopic theory more closely. This is the hydrodynamic (HD) model of Babiker.[8] Again a uniform, isotopic, dielectric continuum is assumed, but this time dispersion is fully taken into account, and indeed appears as an essential ingredient. The connection rules are the standard hydrodynamic ones. These entail the continuity of particle velocity and of mechanical pressure.

Confinement is seen to be caused by the mechanical discontinuity between the two adjacent materials. Connection across the interface is made between these $\mathbf{D}=0$ modes such that $\epsilon(\omega(q))=0$ *on both sides,* which is impossible without dispersion being taken into account. Thus, in Eq. (4) ω_L and ω_T are replaced by $\omega_L(q)$ and $\omega_T(q)$, where q is the total wave vector, which can be imaginary with a dispersion given by the complex phonon band structure (Fig. 2). Where no modes exist with $\epsilon(\omega)=0$ (as in the case for vacuum) the connection rule is still $\mathbf{D}=0$, which means, since $\epsilon(\omega)\neq 0$ in one of the media, that $\mathbf{E}=0$. With HD boundary conditions this translates to $E_z=0$ on both sides of the interface and a discontinuous scalar potential. In contrast, the EM model takes $\mathbf{E}_{\parallel}=\mathbf{0}$ on both sides, which leads to a mechanical discontinuity. For the GaAs/AlAs system the HD model predicts $u_z=0$ at the interfaces for the GaAs LO mode whether $\epsilon(\omega(q))=0$ is taken to apply to the complex branch connecting LO and LA modes in AlAs, or whether, quite simply, it is assumed that $\epsilon(\omega(q))\neq 0$ but $\mathbf{D}=\mathbf{0}$. In either case $u_z=0$, agreeing with the microscopic model [Fig. 1(a)].

We turn to the question of interface modes. The modes we have been describing are guided modes, but in principle there could be interface modes as well. These modes would have maximum amplitude at the interface and imaginary values of q_z. In the HD model the requirement that $u_z=0$ at the GaAs/AlAs interface clearly precludes the existence of LO interface modes for this system. In the EM model, however, $u_z\neq 0$, and indeed, two interface modes are predicted with frequencies which lie between ω_T and ω_L. These are often referred to as Fuchs-Kliewer modes.[7] It is obvious that these modes are not LO modes, since $\epsilon(\omega)\neq 0$. They are, in fact, polaritons[9] (Fig. 3). As such they do not have a scalar potential and cannot therefore interact with electrons via the Fröhlich interaction. To assume that they do (as one of the present authors regrettably did in a previous work[10]) is incorrect. We will discuss the relation between polaritons and LO modes later, but for the present it can be remarked that the use of EM boundary conditions in the EM model predisposes that model to confuse the two, physically distinct, types of vibration.

The LO and TO modes in a medium which is isotropic and homogeneous can be kept distinct whatever the direction of propagation. In the crystal lattices of the III-V compounds, elastic anisotropy blurs this distinction except where the propagation is along a major symmetry

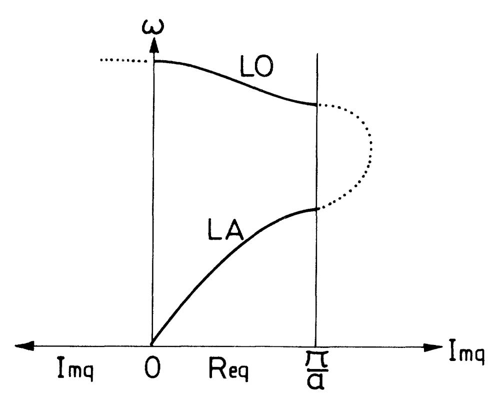

FIG. 2. Complex LO-LA phonon band structure (schematic).

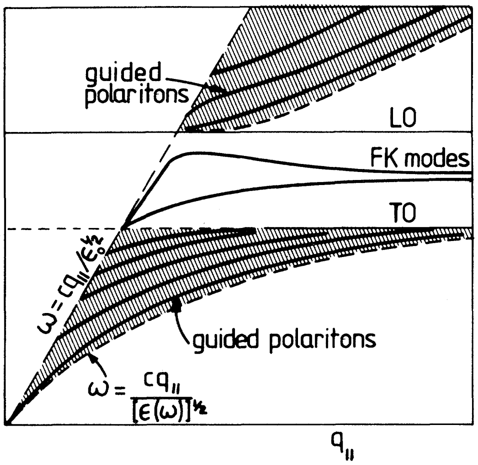

FIG. 3. Surface and guided polariton dispersion in a slab. $\epsilon(\omega)$ is the frequency-dependent dielectric constant $\epsilon(\omega) \rightarrow \epsilon_s(\omega=0)$, $\epsilon(\omega) \rightarrow \epsilon_\infty(\omega \rightarrow \infty)$. Note that the whole span of $q_{\parallel}$ depicted is small compared with typical wave vectors involved in the electron-phonon interaction.

direction, such as along the usual growth direction [100] of the layers. Isotropic continuum theories cannot describe mixed mode effects. Another defect of continuum theories is that they cannot seriously be applied to situations involving changes of vibration amplitude over distances of order of the unit-cell dimension.

Recent analyses of the relation between the EM continuum model and microscopic models have been made by Bechstedt and Gerecke[11] (BG) and by Huang and Zhu[12,13] (HZ). BG begin by adopting the nonretarded limit of Maxwell's equations, viz.,

$$\nabla \times \mathbf{E} = 0 \, , \qquad \nabla \cdot \mathbf{D} = 0 \, , \tag{7}$$

and this approximation is also implied in HZ. Both groups develop a microscopic model that allows solutions which satisfy both the elastic force equations of the lattice and the EM boundary conditions. In the case of normal incidence ($\theta=0$) this is regarded as straightforward for even modes ($n=2,4,6,\ldots$) where a "constant" added to the potential can make $\phi=0$ at the interface. For odd modes ($n=1,3,5\ldots$) this does not work. Nevertheless, HZ choose solutions which make $\phi=0$ for these modes too. In BG, ϕ is taken to be continuous, which leads for n odd to the potential being nonzero at the interfaces and z independent in alternate layers, though it is claimed that no Fröhlich interaction (and hence no Raman scattering) results from that. They note that ϕ being nonzero at the interface contradicts the EM model. In this respect the HZ solution does not contradict the HD model.

Modes propagating at an angle θ to the superlattice axis become mixed to some degree, but for LO modes ϕ vanishes at the interface as a consequence of the EM boundary conditions in both treatments. Both treatments also discover Fuchs-Kliewer (FK) interface modes to which they ascribe a scalar potential. HZ find that the fundamental LO mode ($n=1$) exists only for $\theta=0$ and converts into the upper FK interface mode for $\theta>0$. They argue that this means that the $n=2$ mode should be taken to be the microscopic counterpart of the $n=1$ mode of the EM model, since in the latter the FK interface corresponds to an $n=0$ mode. When they compare the $n=1$ EM mode with the $n=2$ microscopic mode they find a discrepancy in the ionic displacement, which they eliminate (largely) by adding short-range components. They then conclude that the EM model, thus modified, is a good description of confined and interface LO modes. BG are less happy with the EM model and emphasize that the lack of dispersion in that model is a serious deficiency.

The models of BG and HZ, though differing in some aspects, show a substantial amount of agreement. Nevertheless, there appear to be several elements on which both models are based that are open to criticism. These concern (i) dispersion, (ii) the relations between LO modes and polaritons, and (iii) the criteria for boundary conditions. In what follows we discuss these aspects and arrive at the conclusion that all models of LO modes based on EM boundary conditions are fundamentally flawed and that the HD model is the best continuum model that we have so far.

II. DISPERSION

As remarked by both HZ and BG the neglect of dispersion in continuum models leads to the situation where all LO modes are degenerate, and similarly TO modes, whence any linear combination can be taken. More fundamentally, without dispersion it is impossible to construct a quantum theory of vibrational fields. Assuming an isotropic homogeneous continuum which is dispersive we can define the allowed eigenvalues for the system (which in general includes interfaces) as a whole and distinguish longitudinally polarized from transversely polarized modes without ambiguity. In such a model there is no mode mixing—an allowed LO mode with frequency $\omega_L(q)$ is an allowed mode for the system as a whole. Without the dependence on wave vector (dispersion) it is impossible to match the frequencies of LO modes across an interface. We saw that in the EM model matching could only be carried out between a LO mode and a polariton, which meant mixing modes. With dispersion this is naturally avoided.

Taking dispersion into account modifies the permittivity function as follows:

$$\epsilon(\omega) = \epsilon_\infty(q) \frac{\omega^2 - \omega_L^2(q)}{\omega^2 - \omega_T^2(q)} \, . \tag{8}$$

For long wavelengths we can assume that $\epsilon_\infty(q) = \epsilon_\infty(0)$, and, to a good approximation,

$$\omega_L^2 = \omega_{LO}^2 - v_L^2 q^2 \, , \qquad \omega_T^2 = \omega_{TO}^2 - v_T^2 q^2 \, , \tag{9}$$

where ω_{LO} and ω_{TO} are the zone-center frequencies of the

LO and TO modes, and v_L and v_T are the corresponding acoustic velocities. For a frequency to obtain throughout the system it may be necessary that q be complex. In general it is essential to know the complex as well as the real band structure for the phonons (Fig. 2). With dispersion it is now possible for $\epsilon(\omega)=0$ everywhere, and thus LO modes keep their character when crossing an interface. (However, the magnitude of the imaginary wave vector must be regarded as unrestricted for this to be generally true.) We note that the HD model contains all of these features, whereas the EM model does not.

III. OPTICAL PHONONS AND POLARITONS

In a polar material ionic vibrations can generate electric fields and affect the dielectric function $\epsilon(\omega)$, which in turn affects the propagation of EM waves. The allowed vibrations are described by Maxwell's equations plus the equations describing mechanical stress and strain in the medium. The equation which indicates what modes of vibration are possible in the absence of free charge is that of Gauss:

$$\nabla\cdot\mathbf{D}=0 , \tag{10}$$

where $\mathbf{D}$ is the electric displacement. For a traveling wave of wave vector $\mathbf{q}$, Eq. (10) reduces to

$$\mathbf{q}\cdot\mathbf{D}=0 , \tag{11}$$

and so, with finite q, two categories of solution are allowed, namely,

$$\begin{aligned} &\mathbf{D}=0 \quad \text{(longitudinal)} , \\ &\mathbf{D}\neq 0 \quad \text{(transverse)} . \end{aligned} \tag{12}$$

Now $\mathbf{D}=\epsilon_0\mathbf{E}+\mathbf{P}$, where ϵ_0 is the vacuum permittivity and $\mathbf{P}$ is the polarization. For long wavelengths the latter is related to the longitudinal ionic displacement $\mathbf{u}$ via $\mathbf{P}=e^*\mathbf{u}/V_0$, where e^* is the effective ionic charge and V_0 is the volume of the unit cell. Thus $\mathbf{D}=0$ implies that $\mathbf{E}=-e^*\mathbf{u}/\epsilon_0 V_0$ corresponding to the field of a LO mode, attached to which will be a scalar potential ϕ, whose gradient is the negative of $\mathbf{E}$. The energy of an electron in this potential is given by $e\phi$, which is the basis of the Fröhlich interaction.

Turning to the transversely polarized solutions we first note that the dielectric function is given by Eq. (8). Since in general $\mathbf{D}=\epsilon(\omega)\mathbf{E}$, the solution $\mathbf{D}=0$, $\mathbf{E}\neq 0$, means that $\omega=\omega_L(q)$, as it should. For all frequencies $\omega\neq\omega_T(q)$ or $\omega_L(q)$, $\epsilon(\omega)$ is finite and both $\mathbf{D}$ and $\mathbf{E}$ exist as finite transverse vectors. These modes are the polaritons, of mixed photon-TO character, which become increasingly phononlike as ω approaches $\omega_T(q)$.

A glance at the other equations of Maxwell will illuminate those modes. $\mathbf{D}=0$ implies, in the absence of a current, $\mathbf{H}=0$, where $\mathbf{H}$ is the magnetic field, and so

$$\nabla\times\mathbf{E}=0 . \tag{13}$$

This is only true for LO modes. For polaritons both $\mathbf{D}$ and $\mathbf{E}$ are finite and so $\mathbf{H}$ is also finite. In fact, polaritons are just simply EM waves with a dispersion relation given by[9]

$$\omega^2=\frac{c^2}{\epsilon(\omega)}q^2 \tag{14}$$

and

$$\nabla\times\mathbf{E}=-\mu_0\frac{\partial\mathbf{H}}{\partial t} , \tag{15}$$

where c is the velocity of light *in vacuo*. Note that in the frequency region $\omega_T<\omega<\omega_L$ the permittivity is negative and consequently q^2 must be negative, implying attenuation in the medium. This is the reststrahlen region. It is also the region in which lie the Fuchs-Kliewer interface modes. The latter are thus nothing but surface polaritons and they can interact with electrons through the usual $\mathbf{A}\cdot\mathbf{p}$ interaction ($\mathbf{A}$ is the vector potential and $\mathbf{p}$ is the momentum of the particle). That is true also of the guided polaritons. Guided polaritons, being EM waves, do not possess a scalar potential and they therefore cannot interact strongly with electrons. However, polaritons with frequencies near $\omega_T(q)$ may interact with electrons via a deformation potential. It is clear as far as the limiting case of TO modes are concerned the only interaction possible is via a deformation potential. For electrons in a central conduction-band valley this would be ruled out, but for holes it is not.[14] One might expect polaritons to interact with electrons solely through the magnetic process, but with holes through both the magnetic and deformation-potential processes.

It is unfortunate that the electron-phonon interaction based on the dielectric continuum model with interface and guided Fuchs-Kliewer modes continues to be founded on a scalar-potential interaction, as though the modes which were described were LO modes and not polaritons.[15] A clue to how this error could have been made is suggested by Eq. (15). For wave vectors large compared with those of visible light it is often assumed that so-called retardation effects can be ignored and thus the right-hand side of Eq. (15) is put to zero as in Eq. (7). But then it looks like Eq. (13), which could be held to describe a longitudinally polarized mode. If this mistake is made it is easy to assume the existence of a Fröhlich potential associated with a polariton, but this naturally would be incorrect.

IV. BOUNDARY CONDITIONS

Boundary conditions ensure the continuity of the flow of energy and momentum. In order to identify the correct boundary conditions for each of the three types of mode it is necessary to define these correctly.

We note first of all that neither the LO nor pure TO modes [polaritons with $\omega=\omega_T(q)$] possess EM energy, only mechanical. The energy density in a traveling wave is given by

$$U=\bar{\rho}\dot{u}^2 , \tag{16}$$

where $\bar{\rho}$ is the reduced mass density. The flux intensity is thus multiplied by the group velocity $v_g=\nabla_{\mathbf{q}}\omega$:

$$\mathbf{S}=\bar{\rho}\dot{u}^2\nabla_{\mathbf{q}}\omega . \tag{17}$$

From Eq. (9) $\nabla_{\mathbf{q}}\omega=-v_L^2\mathbf{q}/\omega$, and so, focusing on the LO mode,

$$\mathbf{S}_L = -\frac{\bar{\rho}\dot{u}^2 v_L^2}{\omega}\mathbf{q} = \bar{\rho} v_L^2 \dot{\mathbf{u}}(\nabla\cdot\mathbf{u}) \; . \tag{18}$$

A condition on momentum is equivalent to a condition on the force acting. Thus it is necessary for mechanical stability that the pressure associated with the optical vibration be continuous. For the LO mode the optical strain can be written

$$s = \nabla\cdot\mathbf{u} \; , \tag{19}$$

from which a pressure π can be defined via an elastic modulus c_L:

$$\pi = c_L(\nabla\cdot\mathbf{u}) \; , \tag{20}$$

where we define the modulus in terms of the velocity of sound as $c_L = \bar{\rho} v_L^2$, whence

$$\pi = \bar{\rho} v_L^2(\nabla\cdot\mathbf{u}) \; . \tag{21}$$

The continuity of $\mathbf{S}_z$ and π boils down to the continuity of u_z and $\bar{\rho} v_L^2(\nabla\cdot\mathbf{u})$, which are just the HD boundary conditions used by Babiker.[8] A similar argument leads to HD conditions for purely TO modes. It seems that the use of EM boundary conditions for LO modes is not warranted on these grounds.

Polaritons have both EM and mechanical energy and both must be taken into account. We defer this to a future publication and treat polaritons as though they were pure EM waves. The flux vector is then

$$\mathbf{S} = \mathbf{E}\times\mathbf{H} \; , \tag{22}$$

whose continuity in itself provides for the continuity of the tangential components of **E** and **H**, i.e., the EM boundary conditions. Thus the use of EM boundary conditions for the Fuchs-Kliewer interface modes and for guided polaritons is justified only provided the mechanical energy component can be neglected.

V. DISCUSSION

From the foregoing discussion it seems clear that the use of EM boundary conditions cannot be justified on the grounds of energy conservation (except for very-long-wavelength polaritons). Since without the continuity of energy flow no system is stable, it follows that the EM model has to be abandoned as a description of confined LO modes. There seems therefore no point in attempting to reconcile microscopic theories with that model, nor, moreover, is there any point for microscopic theories of LO modes to have EM boundary conditions built into their structure.

It also emerges that dispersion is essential in a continuum model if confusion of mode types is to be avoided. Thus LO and TO modes retain their individual characters across a boundary and do not mix. Even in a real crystal individual characters are retained whatever the direction of propagation, though polarizations may not. Thus a LO mode will retain its scalar potential whatever transverse components may enter as a consequence of crystal anisotropy. Its conversion to a surface polariton for propagation directions away from the superlattice axis, as suggested by HZ, appears, on these grounds, to be unphysical.

The attribution of a scalar potential to surface polaritons goes back to the original treatment by Fuchs and Kliewer. There can be no doubt that the FK interface modes are, indeed, polaritons. As such they are basically transverse modes satisfying Eqs. (14) and (15), and they consequently do not possess a scalar potential. Under certain circumstances true LO interface modes can exist.[8] For example, in the $Al_xGa_{1-x}As$/GaAs system with x not too large the mismatch between the elastic properties of the two media is small enough for u_z to be appreciable such that a guided mode in GaAs penetrates into the $Al_xGa_{1-x}As$ layer. An interface mode is then possible for finite $\mathbf{q}_{\parallel}$, and being a true LO mode it will interact strongly with electrons via its scalar potential.[16] Clearly, such an interface mode has to be distinguished from the FK interface modes, which will also be present.

In conclusion, it seems that the HD model is the best continuum model available at the present time. Its principal virtue is that it incorporates energy conservation, which is a basic requirement. The connection rules used in the HD model are, however, not above criticism, as Akera and Ando[17] point out, and are not the only ones which conserve mechanical energy, but they are the simplest. Of course, as a continuum model it is open to the criticism, voiced by Deans and Inkson[18] against all continuum models, that it is ill equipped to describe the real vibrations of a superlattice, especially a short-period superlattice. But what concerns many people about the HD model is that it allows the scalar potential to be discontinuous. Electrostatic potentials are certainly not allowed to be discontinuous because test charges would have "schizophrenic" energies. It is, however, unclear that the same criterion ought to be applied to the scalar potential of a quantum field when the latter acts solely as a coupling between it and the electron field. The discontinuity means that an electron in GaAs where ϕ is finite would interact with a GaAs LO phonon, whereas an electron in AlAs, where $\phi=0$, would not. This does not seem so terrible.

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