Phonon-polariton modes in superlattices: The effect of spatial dispersion

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The equation of motion for phonons in superlattices including the retardation effect is derived. It is shown that in superlattices additional polariton modes exist due to the spatial dispersion of the optical modes of the constituent bulk materials. The reflectivity spectra of superlattices are also examined.

I. INTRODUCTION

In a polar crystal, the notion of lattice dynamics or phonons implies that the interaction between two point charges is instantaneous. The consequence is that the radiation fields emitted by the vibration of the dipoles are neglected. This is only valid when the wave vector is large compared to the photon wave vector at the optical frequency, $k_p \equiv (\epsilon_\infty)^{1/2}\omega_0/c$, where ϵ_∞ is the high-frequency dielectric constant, ω_0 is the TO-phonon frequency, and c is the speed of light. Usually, k_p is several magnitudes smaller than the size of the first Brillouin zone; therefore the omission of the retardation effect is justified for most of the cases.

The treatment of the retardation effect, first pioneered by Huang,[1] is done by incorporating the lattice dynamics and the macroscopic Maxwell equations. While it is misleading to think of phonon-polariton modes as the coupled states of the *external* photon fields and the internal phonon modes, it is nonetheless convenient to picture the lattice vibrating under the influence of the short-range forces and emitting electromagnetic fields which again interact with the lattice. The vibrations of the lattice and the radiation fields form coherent states (phonon-polariton modes) which are the true internal states of the system.

Rytov[2] and Yeh *et al.*[3] studied the light propagation in a superlattice where the dielectric constants are assumed to be frequency independent. The study of the phonon-polaritons within a microscopic frame began as early as 1972,[4] shortly after the proposal of constructing superlattices. In this work, however, the starting point is from the simplest model to calculate the optical-phonon modes and their corresponding oscillator strengths. The results are very similar to those of polyatomic materials, only qualitative in nature, and are very unrealistic. For instance, the dielectric constant from their work is isotropic which is obviously not true and the oscillator strengths of the individual modes are too close to each other. Recently there has been an increased interest in polaritons in infinite and finite superlattices.[5-7] However, these works did not address the effect of the spatial dispersion of the bulk phonon modes and the additional boundary conditions (ABC) (Ref. 8) thus originated. The dielectric theories with[9] and without[10,11] spatial dispersion effect have qualitatively explained some novel features about the superlattice vibrational modes such as the angular dependence of the long-wavelength optical phonons and the macroscopic interface modes. Though the spatial dispersion in a bulk material has very little effect on the polariton modes, the zone folding in a superlattice system makes the spatial dispersion an important effect for calculating the phonon-polariton modes. Detailed microscopic model calculations[12-14] have demonstrated that the actual optical modes in superlattices are indeed more complicated than those described by dispersionless dielectric continuum model. In the GaAs/AlAs superlattice, the optical-phonon branches of the two constituent materials do not overlap. Hence a model with parabolic phonon dispersion[9] is not a good approximation since in this model the optical-phonon branches of the two materials must overlap, resulting in nonconfined modes.

The necessity of the ABC can be most easily demonstrated in the following way. In the dielectric continuum model which neglects the retardation effect,[10] the following equations hold for the electric field ($\vec{E}$) and the electrostatic potential (Φ) in the superlattice,

$$\vec{E} = -\vec{\nabla}\Phi$$

and

$$\vec{\nabla}\cdot\epsilon\vec{\nabla}\Phi = 0 ,$$

where ϵ is the dielectric function, which has a discontinuous jump across the interfaces. If we write

$$\Phi(\vec{r}) = \phi(z)e^{-i\vec{k}_\parallel\cdot\vec{\rho}}$$

then $\phi(z)$ takes the general form in each continuous region[10]

$$\phi(z) = ae^{k_\parallel z} + be^{-k_\parallel z} + ce^{iqz} + de^{-iqz} ,$$

where q satisfies that $\epsilon(\omega, k_\parallel, q) = 0$. Note that the last two terms on the right-hand side of the above equation are due to the introduction of the spatial dispersion. Thus, we have four undetermined constants a, b, c, and d. In order to have the problem completely determined, two more boundary conditions in addition to the Maxwell boundary conditions are required. The difficulty with the

continuum model is that the ABC for the phonon modes have not yet been properly formulated. Even if these conditions were given, it would be difficult to extract an analytical formula for the dispersion relations in a superlattice system and the asymptotic angular dispersion relation would be even more difficult to obtain. The ABC have been adopted in several theoretical studies of the exciton-polariton mode.[8]

A different approach can be used to study polaritons without introducing ABC.[15] In this approach, one calculates the dielectric function $\epsilon(\vec{r},\vec{r})$ of the system including the retardation effect, then the polariton problem can be solved ABC-free. We have previously developed a simple microscopic theory based on the rigid-ion model.[14,13] Results for long-wavelength optical phonons obtained in this simple theory[14] are in good agreement with those obtained by the full-scale microscopic calculation.[13] Based on our previous work, we shall derive the equations for the superlattice phonon modes with the retardation effect included. We shall compare our results with those obtained by the dispersionless model.

The purpose of our work is threefold. (1) To derive the polariton modes for a superlattice system from macroscopic theory which can be most easily achieved. (2) To derive phonon-polariton modes from microscopic Maxwell equations for bulk and superlattice systems. (3) To include the dispersion of the bulk optical phonons and to avoid the cumbersome ABC problem.

II. DISPERSIONLESS MACROSCOPIC CONTINUUM THEORY

The discussion in this section applies to any kind of polariton. For simplicity, we assume that the constituent materials are isotropic. Throughout the paper, the growth direction of the superlattice is taken to be along the z direction. We only consider the case in which the wave vector $\vec{k}$ lies within the x-z plane ($k_y=0$). This is sufficient, since the results are independent of the azimuthal angle of $\vec{k}$ in the x-y plane. Assume that the electrostatic potential $\phi=0$ and the vector potential is $\vec{A}=(A_x,A_y,A_z)=\vec{A}_0 e^{i(k_x x+k_z z-\omega t)}$ with the Coulomb gauge $\vec{\nabla}\cdot\vec{A}=k_x A_x+k_z A_z=0$, where $\omega^2=c^2k^2/\epsilon$. Thus

$$\vec{E}=i\frac{\omega}{c}\vec{A}$$

and

$$\vec{H}=\vec{B}=i\vec{k}\times\vec{A}=i(-k_z A_y,(k^2/k_z)A_x,k_x A_y) .$$

The Maxwell boundary conditions are $\vec{E}_{\parallel}$ continuous and $\vec{H}_{\parallel}$ continuous. Hence it is required that A_x, A_y, $k_z A_y$, and $(k^2/k_z)A_x$ are continuous across the interfaces. The two components A_x, A_y are totally independent; thus they are treated separately. In what follows we shall use k_λ for the z component of the wave vector $\vec{k}$ and ϵ_λ for the dielectric constant in material λ ($\lambda=1,2$). Note that $k_\lambda=(\epsilon_\lambda\omega^2/c^2-k_x^2)^{1/2}$ are in general complex. Using the Bloch theorem we have for a Kronig-Penny model,

$$\cos(k_1d_1)\cos(k_2d_2)-\frac{1}{2}\left[r+\frac{1}{r}\right]\times\sin(k_1d_1)\sin(k_2d_2)=\cos(qd) , \quad (1)$$

where d_λ is the length of material λ within a superlattice period, $d=d_1+d_2$, and

$$r=\begin{cases}\dfrac{k_2}{k_1} & \text{for component } A_y, \text{ TE modes}\\ \dfrac{\epsilon_1}{\epsilon_2}\dfrac{k_2}{k_1} & \text{for component } A_x, \text{ TM modes.}\end{cases}$$

Since $k_p=\sqrt{\epsilon}\omega/c$ is usually small, when the wave vector $\vec{k}=(k_x,q)$ approaches zero, k_λ's are also small. After expanding the sine and cosine functions and keeping only the leading terms, we obtain the generalized Fresnel equation for the TM modes (extraordinary modes) and the dispersion relation for the TE modes (ordinary modes), with the dielectric constants given by

$$\epsilon_x=\sum_\lambda d_\lambda\epsilon_\lambda/d$$

and

$$\epsilon_z^{-1}=\sum_i d_\lambda\epsilon_\lambda^{-1} \text{ with } d=\sum_\lambda d_\lambda .$$

We shall leave the discussion until the next section.

When k_x is finite and ω is close to the phonon frequencies such that $\epsilon_\lambda\omega^2/c^2\ll k_x^2$, we have $k_i\approx ik_x$. Then, Eq. (1) for the TM modes is reduced to the following form:

$$\cosh(k_xd_1)\cosh(k_xd_2)+\frac{1}{2}\left[\frac{\epsilon_1}{\epsilon_2}+\frac{\epsilon_2}{\epsilon_1}\right]\times\sinh(k_xd_1)\sinh(k_xd_2)=\cos(qd) .$$

This equation is the heart of the dielectric macroscopic model. The angular dispersion and the interface modes can both be derived from here.

Through Eq. (1) we can easily obtain the interface modes for large k_x. From the foregoing argument, it is clear that the interface polariton modes are essentially indistinguishable from the interface phonon modes. Detailed discussions of the interface phonons can be found in Ref. 14.

III. MICROSCOPIC THEORY WITH DISPERSIVE MODEL

In the lattice-dynamics theory without retardation effect, it is assumed that the interaction between two point charges are instantaneous. This is equivalent to setting the speed of light to infinity. For polaritons (phonon-photon coupled field) the lattice dynamics is solved in conjunction with the Maxwell equations for the dielectric medium. Below we show that the polariton problem can be derived from a microscopic theory.

With the retardation effect included, the electric field generated by a moving ion of unit charge is written as

$$\vec{E}=-\vec{\nabla}\phi-\frac{1}{c}\frac{\partial\vec{A}}{\partial t} \tag{2}$$

with

$$\phi=\frac{e^{ik_0r}}{r} \tag{3}$$

and

$$\vec{A}=\frac{1}{c}\frac{e^{ik_0r}}{r}\frac{\partial\vec{u}}{\partial t}=-ik_0\phi\vec{u}\ , \tag{4}$$

where $k_0\equiv\omega/c$ and $\vec{u}$ is the displacement vector of the ion. The equation of motion for the ions in the superlattice is written as

$$\omega^2(\vec{k})\mathbf{M}\vec{U}=C(\vec{k})\vec{U}\ ,$$

where $\vec{M}$ is the mass matrix and $C(\vec{k})$ is the dynamic matrix which consists of two parts—the short-range (SR) interaction and the long-range Coulomb (LG) interaction:

$$C(\vec{k})=C^{\mathrm{SR}}(\vec{k})+C^{\mathrm{LG}}(\vec{k})\ .$$

The matrix elements of C^{LG} are given by

$$C^{\mathrm{LG}}_{i,j}(\vec{k},\vec{r})=Q_1Q_2\sum_{\vec{l}}[\phi_{i,j}(\vec{l}+\vec{r})+k_0^2\delta_{i,j}\phi(\vec{l}+\vec{r})]e^{i\vec{k}\cdot\vec{l}}, \tag{5}$$

where $\vec{l}$ denotes the superlattice lattice vectors and $\vec{r}$ denotes the relative atomic position vector which lies within the superlattice unit cell. Q_1 and Q_2 denote the electron transfer charges for the two ions of interest. $\phi_{i,j}$ $(i,j=x,y,z)$ denotes the (i,j)th second-order derivative of ϕ. The second term in the above equation is due to the vector potential which vanishes in the absence of retardation effect $(k_0=0)$. Rewriting Eq. (5) in the reciprocal space, we obtain

$$C^{\mathrm{LG}}_{i,j}(\vec{k},\vec{r})=\frac{Q_1Q_2}{N_pv}\sum_{\vec{\tau}}[(\tau+k)_i(\tau+k)_j\tilde{\phi}(\vec{\tau}+\vec{k})+k_0^2\delta_{i,j}\phi(\vec{k}+\vec{\tau})]e^{i(\vec{\tau}+\vec{k})\cdot\vec{r}}, \tag{6}$$

where v is the volume of the bulk unit cell, N_p is the number of bulk unit cells within the superlattice unit cell, $\vec{\tau}$ is a reciprocal-lattice vector of the superlattice, and $\tilde{\phi}$ is the Fourier transform of ϕ. We have

$$\tilde{\phi}(k)=\frac{4\pi}{k^2-k_0^2}\ . \tag{7}$$

Since for frequencies comparable with the phonon frequencies, k_0 is very small compared to the size of the Brillouin zone, it is therefore valid to drop k_0 in all the terms except the one with $\vec{\tau}=\vec{0}$. It is obvious at this stage that retardation will only affect the term with $\vec{\tau}=\vec{0}$, which contributes to the macroscopic part of the fields. For the vector-potential contributions, this is the only term that we need to keep. Note that there are additional contributions to the macroscopic part from the $\vec{\tau}\neq\vec{0}$ terms, but they are insensitive to k_0.

The above discussions demonstrate that in a three-dimensional solid, the retardation effect can be included by simply replacing k^2 with $k^2-k_0^2$ in the macroscopic part of the Coulomb matrix due to the electrostatic potential and adding the contributions from the vector potential, which is simply

$$\frac{4\pi Q_1Q_2}{N_pv}\frac{k_0^2}{k^2-k_0^2}\delta_{i,j}\ .$$

However, in practical calculations Eq. (6) does not work, because the summation over τ does not converge fast enough for the numerical evaluation. For the superlattice, a better way to evaluate the Coulomb matrix elements is to first carry out the sum over reciprocal-lattice vectors in a plane perpendicular to $\hat{\mathbf{z}}$ (the growth direction) for each fixed value of l_3, the projection of the lattice vector along the z direction, and then perform the sum over l_3. Using this procedure, we have shown in our previous paper[13] that the macroscopic part of the Coulomb matrix without the retardation effect can be written as

$$C^{\mathrm{macro}}_{i,j}=\frac{4\pi Q_1Q_2}{N_pv}\left[\frac{k_ik_j}{k^2}-\delta_{i,z}\delta_{j,z}\right]. \tag{8}$$

The macroscopic part of the Coulomb matrix due to the electrostatic potential with retardation effect is then obtained by simply replacing k^2 in Eq. (8) with $k^2-k_0^2$. Adding the contributions from the vector potential, we obtain in the long-wavelength limit $(kd\ll1)$

$$C^{\mathrm{macro}}_{i,j}=-\frac{4\pi Q_1Q_2}{N_pv}\left[\frac{k_ik_j}{k^2-k_0^2}-\delta_{iz}\delta_{jz}-\frac{k_0^2}{k^2-k_0^2}\delta_{i,j}\right]. \tag{9}$$

Introducing dimensionless variables $s_i\equiv k_i/k$ and $n\equiv k/k_0=ck/\omega$ (the refractive index), we can rewrite Eq. (9) as

$$C^{\mathrm{macro}}_{i,j}=-\frac{4\pi Q_1Q_2}{N_pv}\left[\frac{n^2}{n^2-1}s_is_j-\delta_{i,z}\delta_{j,z}-\frac{1}{n^2-1}\delta_{i,j}\right]. \tag{10}$$

Owing to the circular symmetry in the x-y plane we only have to consider the case in which the wave vector lies in the x-z plane. In this case, the y-component vibration is completely decoupled from the x- and z-component vibrations, and the equation of motion for ions in the superlattice reads

$$(M_J\omega^2-C_0)\mathbf{U}(J)-[C_-\mathbf{U}(J-1)+C_+\mathbf{U}(J+1)]=\frac{4\pi}{N_pv}Q_J\begin{bmatrix}S&-S\\-S&S\end{bmatrix}\sum_{J'}Q_{J'}\mathbf{U}(J')\ , \tag{11}$$

where $\mathbf{U}$ is a four-component column vector with the first two components describing the vibrations along the x and z directions for cations and the last two components

describing those vibrations for anions. J labels the positions of bilayers (each bilayer consists of one cation and one anion atomic layer) within the superlattice period, and M_J and Q_J are the mass matrix and the electron transfer charge of the cation layer at bilayer J, respectively. C_-, C_0, and C_+ are short-range parts of the dynamic matrix (including the Coulomb matrix minus the macroscopic part), describing interaction with a bilayer to the left, the same bilayer, and a bilayer to the right, respectively. S is a 2×2 matrix given by

$$S=\begin{bmatrix} -\frac{1}{n^2-1}+\frac{n^2}{n^2-1}\sin^2\theta & \frac{n^2}{n^2-1}\sin\theta\cos\theta \\ \frac{n^2}{n^2-1}\sin\theta\cos\theta & -\frac{1}{n^2-1}-1+\frac{n^2}{n^2-1}\cos^2\theta \end{bmatrix},$$

where θ is the angle between $\vec{k}$ and $\hat{\mathbf{z}}$.

As discussed in Ref. 14, the homogeneous equation involving only the short-range forces can be easily solved. Since k is small compared with the size of the Brillouin zone, we can approximate the short-range interactions by their values at $k=0$. We denote the homogeneous solutions by

$$\mathbf{U}^{(\lambda)}_{\nu n}(J)=f^{(\lambda)}_n(J)\mathbf{P}^{(\lambda)}_{\nu n}(J),\quad \nu=T,L$$

where $f^{(\lambda)}_n(J)$ is called the "envelope function," $\lambda=1,2$ for GaAs-like and AlAs-like modes, respectively, n is the principal quantum number ($n=1,2,3,\ldots$), $\nu=T,L$ denotes the transverse and longitudinal modes, and $\mathbf{P}^{(\lambda)}_{\nu n}(J)$ is a four-component column vector describing the polarization in bilayer J. If $f^{(\lambda)}_n(J)$ is properly chosen, $\mathbf{P}^{(\lambda)}_{\nu n}(J)$ would be almost independent of J. The associated eigenfrequency is denoted by $\omega^{(\lambda)}_{\nu n}$.

For optical modes which are confined in either the GaAs or AlAs slab, it is a good approximation to write

$$f^{(\lambda)}_n(J)\approx \sin\left[\frac{nJ\pi}{N_\lambda+1}\right], \tag{12}$$

where N_λ is the total number of bilayers in material λ and J runs from 1 to N_λ. The total dipole moment in the nth mode is then proportional to

$$d^{(\lambda)}_n=\sum_J f^{(\lambda)}_n(J)=\frac{1-(-1)^n}{2}\cot\frac{n\pi}{2(N_\lambda+1)}. \tag{13}$$

Furthermore, $\mathbf{P}^{(\lambda)}_{\nu n}$ can be approximated by the polarization vector of the bulk optical mode at the zone center.

To solve Eq. (11), we expand the eigenfunction $\mathbf{U}$ in terms of the solutions to the homogeneous equation, viz.,

$$\mathbf{U}(J)=\sum_{\lambda,n,\nu} C^{(\lambda)}_{\nu n} f^{(\lambda)}_n(J)\mathbf{P}^{(\lambda)}_{\nu n}.$$

Substituting the above equation into Eq. (11) and multiplying the equation by $U^{(\lambda)\dagger}_{\nu n}$ from the left, we obtain

$$(\omega^2-\omega^{(\lambda)2}_{\nu n})C^{(\lambda)}_{\nu n}=\sum_{\lambda'}\beta_{\lambda'}d^{(\lambda')}_n\sum_{n'}d^{(\lambda')}_{n'}\sum_{\nu'}S_{\nu\nu'}C^{(\lambda')}_{\nu' n'}\vec{u}_{j,\lambda'}, \tag{14}$$

where

$$\beta_\lambda=\frac{2}{N_p(N_\lambda+1)}\frac{4\pi Q^2_\lambda}{\mu_\lambda v}=\frac{2}{N_p(N_\lambda+1)}[(\omega^{(\lambda)}_{\mathrm{LO}})^2-(\omega^{(\lambda)}_{\mathrm{TO}})^2].$$

Note that the dipole moment in a superlattice unit cell in a homogeneous mode $(\lambda,\nu n)$ is $\vec{P}^{(\lambda)}_{\nu n}=d_n\vec{p}^{(\lambda)}_\nu$, where $\vec{p}^{(\lambda)}_\nu$ denotes the dipole moment within a bulk unit cell associated with the νth branch in material λ (assumed to be $\vec{k}$ independent). Thus the total dipole strength in a polariton mode is $\vec{P}=\sum_{\lambda,\nu n}C_{\nu n}\vec{P}^{(\lambda)}_{\nu n}=\sum_{\lambda,\nu,n}d^{(\lambda)}_n C^{(\lambda)}_{\nu n}\vec{p}^{(\lambda)}_\nu$. Equation (14) now takes the form

$$\vec{P}=\begin{bmatrix} A_x & 0 \\ 0 & A_z \end{bmatrix}S\vec{P}, \tag{15}$$

where

$$A_\nu=\sum_\lambda\beta_\lambda\sum_n\frac{d^{(\lambda)2}_n}{\omega^2-\omega^{(\lambda)2}_{\nu n}},\quad \nu=x,z\ (\text{or } T,L). \tag{16}$$

Introducing

$$\hat{\mathbf{S}}_{i,j}=-S_{i,j}-\delta_{i,z}\delta_{j,z}=(n^2-1)\delta_{i,j}-\frac{n^2}{n^2-1}s_i,s_j, \tag{17}$$

we can rewrite Eq. (15) as

$$\begin{bmatrix} P_x \\ P_z+A_zP_z \end{bmatrix}=-\begin{bmatrix} A_x & 0 \\ 0 & A_z \end{bmatrix}\hat{\mathbf{S}}\vec{P},$$

which again yields

$$\vec{P}=-\begin{bmatrix} A_x & 0 \\ 0 & \frac{A_z}{1+A_z} \end{bmatrix}\hat{\mathbf{S}}\vec{P}\equiv 4\pi\chi\hat{\mathbf{S}}\vec{P}. \tag{18}$$

Anologue to the bulk case, we see that (see, for example, Ref. 16)

$$4\pi\hat{\mathbf{S}}\vec{P}=\vec{E} \tag{19}$$

is the total macroscopic electric field. It is then clear that

$$\chi=-\frac{1}{4\pi}\begin{bmatrix} A_x & 0 \\ 0 & A_z/(1+A_z) \end{bmatrix} \tag{20}$$

is the susceptibility of the lattice. Thus the dielectric constant is

$$\epsilon=1+4\pi\chi=\begin{bmatrix}1-A_x & 0\\ 0 & 1/(1+A_z)\end{bmatrix} . \tag{21}$$

With the use of Eqs. (17) and (21), Eq. (18) can be put into the bra-ket form

$$(n^{-2}\epsilon-1)|P\rangle+|s\rangle\langle s|P\rangle=0 ,$$

where a bra (ket) is a two-dimensional column (row) vector. The generalized Fresnel equation (see the Appendix for derivation) gives for the extraordinary polariton modes

$$\frac{\sin^2\theta}{\frac{1}{n^2}-\frac{1}{\epsilon_x}}+\frac{\cos^2\theta}{\frac{1}{n^2}-\frac{1}{\epsilon_x}}=0 , \tag{22}$$

which is easily transformed to the following form:

$$\epsilon_x\epsilon_z-n^2(\epsilon_x\sin^2\theta+\epsilon_z\cos^2\theta)=0 .$$

When $\theta=0$ the above equation also gives the dispersion for ordinary polariton modes, viz., $\epsilon_x-n^2=0$.

IV. CORRECTIONS DUE TO THE ELECTRONIC POLARIZABILITY

The derivation so far is based on the rigid-ion model which does not treat the electronic polarizability properly. This model may give good dispersion relations for phonon modes, but the results for polaritons will be erroneous even for bulk systems. For if the electronic polarizability is ignored, the high-frequency dielectric constants will be unity so that the slope of the polariton dispersion at higher frequencies will be unity instead of $1/(\epsilon_\infty)^{1/2}$. Furthermore, the coupling region will also be shifted to a smaller wave-vector region. This problem can be remedied with the dipole model. In this model it is assumed that the electron clouds are massless. For bulk material we have

$$(M\omega^2-H_0)\vec{P}_I=-\frac{Q^2}{v}\vec{E}_l \tag{23}$$

and

$$\vec{P}=\vec{P}_I+\vec{P}_e=\vec{P}_I+\alpha\vec{E}_l , \tag{24}$$

where H_0 is the short-range interaction matrix, M and Q are mass and charge matrices, subscripts I and e stand for ion and electron, respectively, α is the electronic polarizability, and $\vec{E}_l=\vec{E}_e+\vec{E}_I$ is the net local electric field due electrons and ions. We next write

$$\vec{E}_l=V(\vec{k})\vec{P}_l+V(\vec{0})\vec{P}_e+\vec{E} , \tag{25}$$

where $V(\vec{k})$ is the short-range part of the Coulomb matrix and $\vec{E}$ is the macroscopic electric field. Here we use $V(\vec{0})$ for the electronic term, because we can view the electronic polarization as a continuum field. At $\vec{k}=0$, $V_0=V(\vec{0})=4\pi/3$, $-4\pi/3$ for the transverse and longitudinal parts, respectively. Equations (24) and (25) lead to

$$(1-\alpha V_0)\vec{P}_e=\alpha\vec{E}+\alpha V\vec{P}_I .$$

Hence we have

$$\left[M\omega^2-H_0+\frac{Q^2}{v(1-\alpha V_0)}V\right]\vec{P}_I=-\frac{Q^2}{v(1-\alpha V_0)}\vec{E} . \tag{26}$$

Thus for the phonon-polariton modes the effective charge is

$$Q'=\frac{Q}{(1-\alpha V_0)^{1/2}}$$

which has different values for the vibration along the two different directions. The optical-phonon frequencies at the zone center are

$$\omega_{\rm TO}^2=\omega_0^2-\frac{4\pi}{3}\frac{Q^2}{\mu v\left[1-\frac{4\pi}{3}\alpha\right]}$$

and

$$\omega_{\rm LO}^2=\omega_0^2+\frac{8\pi}{3}\frac{Q^2}{\mu v\left[1+\frac{8\pi}{3}\alpha\right]} .$$

This is also the result of a shell model for the bulk materials. The ion charge can be obtained from the following relations:

$$\frac{4\pi Q^2}{v\mu}=\left[1+\frac{8\pi}{3}\alpha\right]\left[1-\frac{4\pi}{3}\alpha\right]\Delta\omega^2, \quad \frac{4\pi}{3}\alpha=\frac{\epsilon_\infty-1}{\epsilon_\infty+2} .$$

Hence for each mode vn we have

$$\gamma_{vn}=-\frac{4\pi\alpha}{1-\alpha V(\vec{0})}+\frac{4\pi[1-\alpha V(\vec{0})+\alpha V_{vn}]Q^2}{(1-\alpha V_0)^2\mu v}\frac{1}{\omega^2-\omega_{vn}^2} , \tag{27}$$

where V_{vn} is the expectation value of V in mode vn. Note that the index λ has been omitted in the above equations.

In Eq. (16) the following replacement should be made:

$$\frac{4\pi Q^2}{v\mu(\omega^2-\omega_{vn}^2)}\rightarrow\gamma_{vn} .$$

The dielectric functions for the two symmetry directions are

$$\epsilon_x=1-A_x=\sum_{\lambda,n}S_n^{(\lambda)}\epsilon_{n,x}=\epsilon_{\infty,x}-\sum_{\lambda,n}\frac{R_{Tn}^{(\lambda)}}{\omega^2-\omega_{Tn}^{(\lambda)^2}} ,$$

$$\frac{1}{\epsilon_z}=1+A_z=\sum_{\lambda,n}S_n^{(\lambda)}\frac{1}{\epsilon_{n,z}}=\frac{1}{\epsilon_{\infty,z}}+\sum_{\lambda,n}\frac{R_{Ln}^{(\lambda)}}{\omega^2-\omega_{Ln}^{(\lambda)^2}} ,$$

where $S_n^{(\lambda)}=2d_n^{(\lambda)^2}/N_p(N_\lambda+1)$ and

$$R_{vn}=\Delta\omega^2(1-4\pi\alpha/3+V_{vn}\alpha), \quad v=L,T$$

is the oscillator strength for the mode vn (omitting index λ),

$$\epsilon_{\infty,x}=\sum_{\lambda}\frac{N_\lambda}{N_p}\epsilon_{\infty,\lambda}\ ,$$

and

$$\frac{1}{\epsilon_{\infty,z}}=\sum_{\lambda}\frac{N_\lambda}{N_p}\frac{1}{\epsilon_{\infty,\lambda}}\ .$$

Phenomenologically, $\epsilon_{\infty,\nu}$, $\omega_{\nu n}$, and $R_{\nu n}$ can be adjusted to fit the experimental data.

When the retardation effect is neglected, we have

$$(1-A_x)(1+A_z)\sin^2\theta+\cos^2\theta=0\ ,$$

which is just the result we have obtained in a previous paper[14] for the angular dependence of the optical phonons in superlattices.

V. RESULTS

A. Dispersion curves

We solve the generalized Fresnel equation using the standard numerical method (see Ref. 16). The polariton dispersion curves for a (5,5) GaAs/AlAs superlattice for wave vectors along the z direction ($\theta=0$) are plotted in Fig. 1 along with the results obtained from the dispersionless macroscopic model (dashed lines). Several features in this figure deserve mention. First, the microscopic calculations with spatial dispersion effect give rise to more polariton branches than those obtained by the dispersionless model. Second, there are three polariton branches which have substantial dispersion, corresponding to coupled modes derived from the photon, the GaAs-like principal ($n=1$) TO phonon, and the AlAs-like principal ($n=1$) TO phonon. The dispersions of these polariton branches obtained in both models are similar. The remaining polariton branches are either pure LO phonons or coupled modes derived predominantly from the $n\neq1$ TO phonons, which are nearly dispersionless. This can be explained as follows. We see that without the macroscopic field, each individual mode can be viewed as an independent oscillator whose dipole strength is approximately proportional to d_n. (This can be clarified by comparing the equation above with the cases with polyatomic unit cells[16] where each dipole can be physically identified.) Thus the main contribution is originated from the principal modes ($n=1$) which have the largest dipole strengths. If we ignore the dipole strengths of the $n\neq1$ modes, then the microscopic model becomes qualitatively equivalent to the dispersionless macroscopic model. Third, the frequencies of the polariton modes at $k=0$ are given by

$$\omega^2(1-A_x)\prod_{\lambda,n}(\omega^2-\omega_{Ln}^{(\lambda)^2})=0\ .$$

They are no longer angular dependent (as opposed to the cases where the retardation effect is neglected). Fourth, the angular dispersion of the optical phonons are given by the polariton modes in the limit $ck=\infty$. This is of course not in contradiction with the definition of the long-wavelength optical phonons since when the phonons and photons are well decoupled the wave vector k is still substantially smaller than the size of the Brillouin zone in

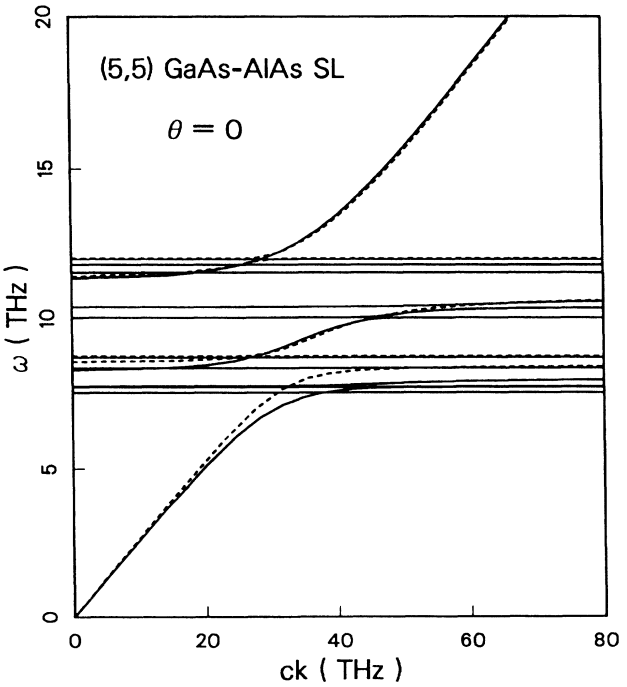

FIG. 1. Polariton dispersion curves for a (5,5) GaAs/AlAs superlattice along the growth (z) direction.

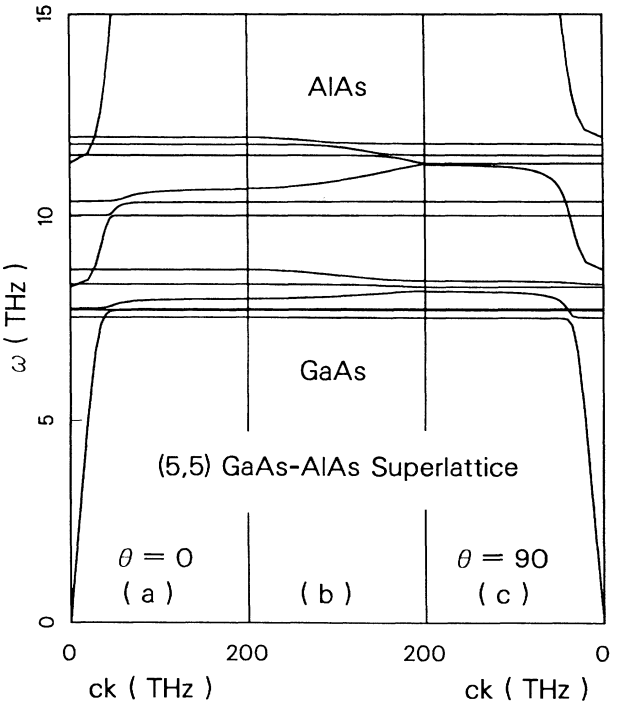

FIG. 2. Polariton dispersion curves for a (5,5) GaAs/AlAs superlattice: (a) $\theta=0°$, $ck=0$–80 THz; (b) $\theta=0°$–$90°$, $ck=\infty$; (c) $\theta=90°$, $ck=80$–0 THz.

a typical superlattice with period less than a few hundred angstroms.

To demonstrate the relation between the polariton dispersion and the angular dispersion of long-wavelength optical-phonon modes, we plot in Fig. 2(a) the polariton dispersion curves at $\theta=0°$ with ck varying from 0 to 200 THz, in Fig. 2(b) the angular dispersion curves for long-wavelength ($k\approx 0$) optical phonons (without the retardation effect, i.e., $ck=\infty$) with θ varying from 0° to 90°, and in Fig. 2(c) the polariton dispersion curves at $\theta=90°$ with ck varying from 200 to 0 THz for the (5,5) superlattice. We point out here that if the high-frequency dielectric constants of the two materials are not much different, the effect of the electronic polarization is not significant as far as the mechanical phonon modes are concerned. It is easy to see from Eq. (26) that if the dielectric constants in the two materials are the same and if we let $V_{\nu n}=V(\vec{0})$ then the formulas for the phonon dispersion are exactly the same with or without including the electronic polarization.

B. Reflectivity

The reflectivity spectrum of a solid can be influenced by the structure of the surface and the spatial dispersion.[17] In the discussions below, we shall ignore the effects due to the reconstruction and the roughness of the surface. For incident light with polarization along the direction ν ($=x,z$), the reflectivity can be obtained through the following relation:

$$R=\left|\frac{1-[\epsilon_\nu(\omega)]^{1/2}}{1+[\epsilon_\nu(\omega)]^{1/2}}\right|^2 .$$

Note that for incident light propagating along the growth (z) direction, only the x polarization is possible, whereas for incident light propagating in the plane (perpendicular to the growth direction), both the x and z polarizations are possible. For damped phonon-polariton modes,

$$\epsilon_x=\epsilon_{x,\infty}-\sum_{\lambda,n}\frac{R_{T,n}^{(\lambda)}}{\omega^2-\omega_{Tn}^{(\lambda)2}+i2\Gamma_{n,x}^{(\lambda)}\omega} ,$$

$$\frac{1}{\epsilon_z}=\frac{1}{\epsilon_{x,\infty}}+\sum_{\lambda,n}\frac{R_{Ln}^{(\lambda)}}{\omega^2-\omega_{Ln}^{(\lambda)2}+i2\Gamma_{n,z}^{(\lambda)}\omega} ,$$

where $\Gamma_{n,\nu}^{(\lambda)}$ are the damping parameters for the corresponding modes.

The numerical results are plotted in Figs. 3 and 4 for incident light polarized along the x and z directions, respectively. The damping factors used are 0.01 THz. It is found that when the broadening is large the main contribution comes from the principal modes independent of the size of the superlattice. Only when the broadening is sufficiently small can the contributions from other modes be observable.

VI. SUMMARY

In summary, we have shown that the equation of motion for polaritons can be obtained from microscopic theory. When the spatial dispersion effect is included, the number of phonon modes are proportional to the size of the period.

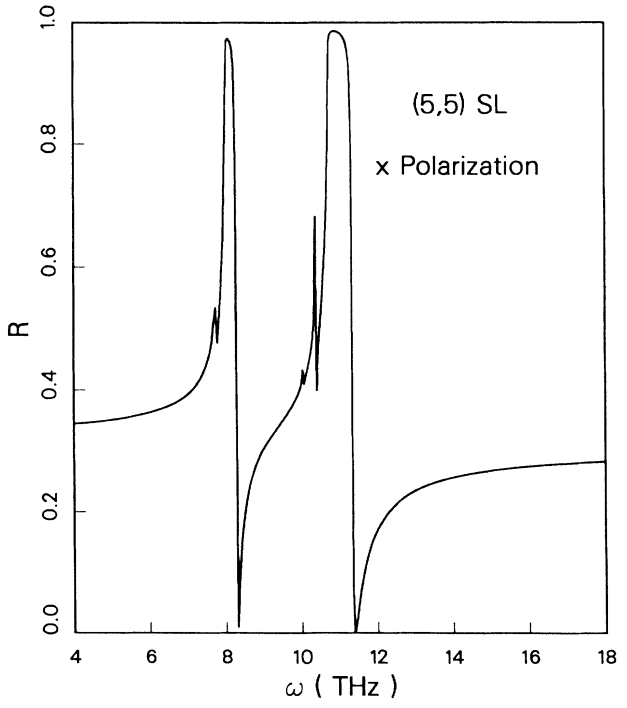

FIG. 3. Reflectivity spectrum of a (5,5) GaAs/AlAs superlattice for incident light polarized along the in-plane (x) direction.

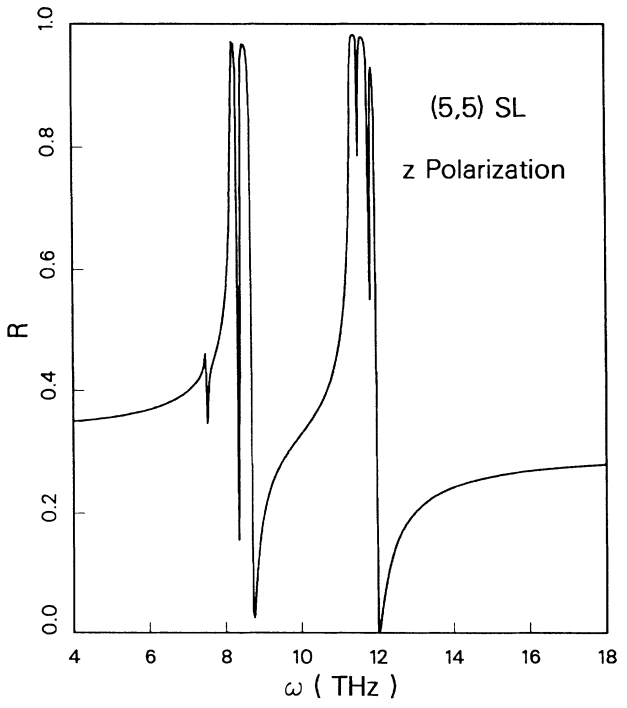

FIG. 4. Reflectivity spectrum of a (5,5) GaAs/AlAs superlattice for incident light polarized along the growth (z) direction.

ACKNOWLEDGMENTS

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APPENDIX

Consider the following type of equation:

$$A|x\rangle+|s\rangle\langle s|x\rangle=0 .$$

Letting $B=A^{-1}$, we obtain

$$|x\rangle+B|s\rangle\langle s|x\rangle=0 ,$$

which is then transformed to the form

$$\langle s|x\rangle+\langle s|B|s\rangle\langle s|x\rangle=0 .$$

The nontrivial solution is

$$1+\langle s|B|s\rangle=0 .$$

If $|s\rangle$ is normalized we have

$$\langle s|(1+B)|s\rangle=0 .$$

If B is diagonal we then have

$$\sum_i (1+B_i)s_i^2=0 .$$

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