Surface polaritons in layered structures of anisotropic media

R. N. Costa Filho* and M. G. Cottam
Department of Physics, University of Western Ontario, London, Ontario, Canada N6A 3K7

E. L. Albuquerque†
Department of Physics, Harvard University, Cambridge, Massachusetts 02138

G. A. Farias*
Department of Physics, University of Antwerp, Universiteitsplein 1, B-2610 Antwerpen-Wilrijk, Belgium
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We discuss the spectra of surface polaritons which can propagate in layered structures in which one of the constituent media has an anisotropic dielectric function. Some unusual aspects of the surface-polariton spectra are emphasized for slabs and superlattices, and are shown to be strongly influenced by the orientation of the principal axis of the anisotropic material. The theory is employed to obtain numerical results for surface-phonon-polaritons in layered structures containing α-quartz. [S0163-1829(96)00528-0]

I. INTRODUCTION

Collective electromagnetic excitations (including bulk and surface modes) in slabs and superlattices have been the subject of intensive studies in the past decade or so (for reviews of this subject see, e.g., Refs. 1–3). On the experimental side, the rapid progress of crystal-growth techniques, such as molecular-beam epitaxy and metal-organic chemical vapor deposition, has made possible the fabrication of good quality specimens, and this has pushed forward the investigations of their physical properties.[3,4]

The most interesting physical effects occurring in these systems arise due to the presence of surfaces and interfaces. For example, it is well known (e.g., see Ref. 1) that an isolated dielectric-vacuum interface can support a surface excitation for appropriate values of the in-plane wave vector $\mathbf{q}_\parallel$ and frequency ω. A symmetric isotropic slab of finite thickness may typically support two such modes, one associated mainly with each surface. The two modes couple to produce odd- and even-parity combinations, split by interaction between the two surfaces. In a superlattice, on the other hand, the excitation within each layer may produce electric and magnetic fields that extend outside its boundaries. There can then be a coupling with the excitations of the other layers and, through Bloch's theorem, this can lead to a collective mode of the whole structure.

It is the aim of this paper to study collective mode of the polariton type that can propagate in layered structures, such as slabs and superlattices, containing anisotropic media. We focus our attention on *surface* polaritons in slabs and both *bulk* and *surface* polaritons in superlattices since, as we show later, they are strongly influenced by anisotropy in the dielectric function of the layer(s). The frequency-dependent dielectric function of the anisotropic medium is assumed to have a diagonal form with elements $\epsilon_{xx}(\omega)$, $\epsilon_{yy}(\omega)$, and $\epsilon_{zz}(\omega)$, relative to principal axes x, y, and z. We focus on uniaxial materials, taking one of these diagonal elements to be $\epsilon_\parallel(\omega)$ (in the direction of the optic axis) and the other two to be $\epsilon_\perp(\omega)$. To obtain a description of phonon-polaritons we assume[5]

$$\epsilon_\parallel(\omega)=\epsilon_{\parallel\infty}\prod_{i=1}^{m_\parallel}\frac{(\omega_{\parallel i}^{l^2}-\omega^2)}{(\omega_{\parallel i}^{t^2}-\omega^2)}, \tag{1}$$

in terms of the frequencies $\omega_{\parallel i}^{t}$ and $\omega_{\parallel i}^{l}$ of the TO and LO phonons, respectively. Here the index i labels the reststrahlen bands, and $m_\parallel$ is the number of these bands: there is an analogous expression for $\epsilon_\perp(\omega)$. For example, $m_\parallel=4$ and $m_\perp=8$ in the case of α-quartz.[5]

Previous calculations of surface polaritons in anisotropic media have mostly been for the single-interface geometry (see, e.g., reviews in Refs. 1, 2, and 6), while some brief studies for the two-interface geometry (i.e., slabs or films) have been reported.[6,7] The purpose of this paper is primarily to examine the role of anisotropic media in superlattices, where interesting features (compared to the one- and two-interface cases) occur due to the Bloch periodicity of the multilayer structure. Specifically, we consider the phonon-polaritons in semi-infinite binary superlattices, where one constituent material is anisotropic (e.g., α-quartz) and the other is isotropic. It is instructive first to consider the case of an anisotropic slab bounded by an isotropic medium, since this will serve to introduce the theoretical method, and will allow numerical comparison with the superlattice case.

II. SINGLE SLAB

Consider initially a single anisotropic slab of thickness l occupying the region $-l<z<0$, where we assume the principal axis z to be perpendicular to the surfaces. The surfaces are parallel to the Cartesian xy plane, and the slab is surrounded by the vacuum. We examine the physically interesting case of a p-polarized electromagnetic wave propagating in this slab. In general, the excitation will set up a fluctuating electromagnetic field in the vacuum regions above and below the slab. Translational invariance parallel to the surfaces ensures that all elementary excitations are characterized by a

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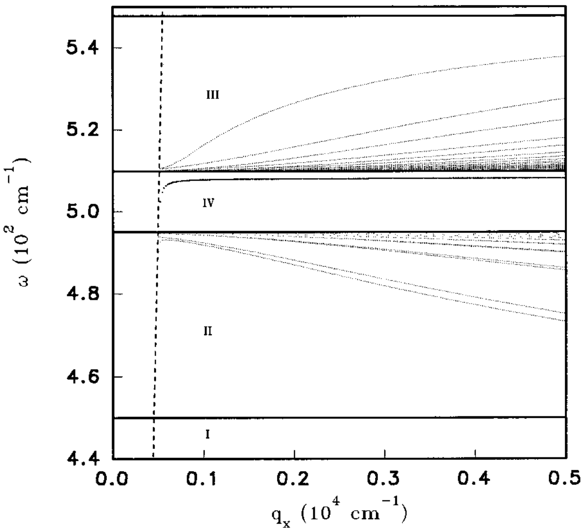

FIG. 1. Surface polaritons in a slab of α-quartz (thickness 4 μm) surrounded by vacuum. The optic axis of the crystal is oriented parallel to the x axis. Here the short-dashed line is the light line. The notation for the different regions is explained in the text.

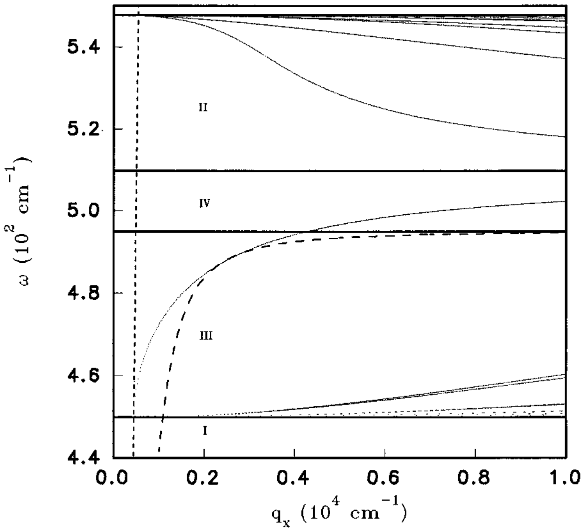

FIG. 2. As in Fig. 1, but with the optic axis in the z direction. The long-dashed line represents the curve $q_x = \sqrt{\epsilon_{zz}}\omega/c$.

two-dimensional wave vector $\mathbf{q}_\parallel$ parallel to the surfaces. We choose $\mathbf{q}_\parallel$ along the x axis, and the standard solutions of Maxwell's equations then yield [where we omit for convenience common factors of $\exp(iq_x - i\omega t)$]

$$\mathbf{E} = (E_{x_1}, 0, iq_x E_{x_1}/\alpha_1)\exp(-\alpha_1 z) \quad \text{for } z>0, \tag{2}$$

$$\begin{aligned}\mathbf{E} = \{&A \exp[-\alpha_2(z+l/2)] + B \exp[\alpha_2(z+l/2)], 0,\\ &(iq_x\epsilon_{xx}/\alpha_2\epsilon_{zz})A \exp[-\alpha_2(z+l/2)]\\ &-B\exp[\alpha_2(z+l/2)]\} \quad \text{for } -l<z<0,\end{aligned} \tag{3}$$

$$\mathbf{E} = (E_{x_3}, 0, -iq_x E_{x_3}/\alpha_1)\exp[\alpha_1(z+l)] \quad \text{for } z<-l. \tag{4}$$

Here we have denoted $\alpha_1^2 = q_x^2 - \omega^2/c^2$ and $\alpha_2^2 = (\epsilon_{xx}/\epsilon_{zz})(q_x^2 - \epsilon_{zz}\omega^2/c^2)$.

Using the electromagnetic boundary conditions at the two interfaces, the solvability condition of the resulting four homogeneous equations yields the implicit dispersion relation for the surface polaritons as

$$\exp(2\alpha_2 l) = \left(\frac{1-\alpha_1\epsilon_{xx}/\alpha_2}{1+\alpha_1\epsilon_{xx}/\alpha_2}\right)^2, \tag{5}$$

which is consistent with previous works.[6,7]

In Fig. 1 we show a typical region of the surface-phonon-polariton spectrum calculated for a film of α-quartz with its principal axis c (the optic axis) oriented parallel to the x axis. We use the dielectric function defined in Eq. (1), with parameter values as quoted in Ref. 5, and in this case $\epsilon_{xx} = \epsilon_\parallel$ and $\epsilon_{yy} = \epsilon_{zz} = \epsilon_\perp$. The thick horizontal lines correspond to optic-phonon frequencies $\omega_{\perp 4}^t \simeq 450$ cm^{-1}, $\omega_{\parallel 2}^t \simeq 495$ cm^{-1}, $\omega_{\perp 4}^l \simeq 510$ cm^{-1}, and $\omega_{\parallel 2}^l \simeq 548$ cm^{-1}. Several different regions of surface-polariton behavior can be identified between the horizontal lines as follows. Region I corresponds to $\epsilon_{zz}>0$ and $\epsilon_{xx}>0$: in this case there is no solution for either $q_x < \omega/c$ or $q_x > \epsilon_{zz}^{1/2}\omega/c$, since α_1 and α_2 are either real or pure imaginary quantities. On the other hand, for $\omega < cq_x < \epsilon_{zz}^{1/2}\omega$, α_1 is real although α_2 is pure imaginary, and there can be guided modes confined in the film. Region II has $\epsilon_{zz}<0$ and $\epsilon_{xx}>0$: here α_2 is pure imaginary whatever the value of q_x, which implies guided modes for all $q_x > \omega/c$. Region III corresponds to $\epsilon_{zz}>0$ and $\epsilon_{xx}<0$: there is no solution for $q_x < \omega/c$. For the range $\omega < cq_x < \epsilon_{zz}^{1/2}\omega$, α_2 is real and therefore the so-called virtual surface-polariton modes,[5,8] which would not occur in an isotropic medium, are allowed to propagate. These modes usually terminate at a finite value of q_x. For $q_x > \epsilon_{zz}^{1/2}\omega/c$, α_2 is pure imaginary, and guided modes can propagate in the slab. Finally, in region IV ($\epsilon_{zz}<0$ and $\epsilon_{xx}<0$), we have the propagation of real surface-polariton modes (i.e., those which are analogous to the surface polaritons in an isotropic medium) for $q_x > \omega/c$.

In addition to the deformed real surface-polariton branches and the appearance of virtual surface-polariton branches, the anisotropy gives rise to a pronounced directional dependence. This difference is clearly illustrated in Fig. 2, which is also for α-quartz but with the optic axis now in the z direction perpendicular to the surfaces ($\epsilon_{xx} = \epsilon_{yy} = \epsilon_\perp$, $\epsilon_{zz} = \epsilon_\parallel$).

III. SEMI-INFINITE SUPERLATTICE

We now turn our attention to a semi-infinite superlattice structure made up of alternating layers of two materials A and B. One of them (say medium A) is the α-quartz anisotropic material whose dielectric function is given as in Eq. (1), while medium B has a constant scalar dielectric function ϵ_B. We consider the superlattice occupying the region $z<0$ with a layer of medium A at its surface, while the vacuum fills $z>0$. Again, the Cartesian xy plane is chosen to be parallel to the surface of the layers. The nth unit cell of the superlattice is bounded by the planes $z = -(n-1)L$ and $z = -nL$, with $L = d_A + d_B$ being the size of the unit cell and d_A (d_B) denoting the thickness of the A (B) layer. The solution of Maxwell's equations in the nth unit cell in p polarization yields, with $\mathbf{q}_\parallel$ along the x axis as before,

$$E_{xA}^{(n)}(z)=A_{1A}^{(n)}\exp(-\alpha_A z)+A_{2A}^{(n)}\exp(\alpha_A z), \tag{6}$$

$$H_{yA}^{(n)}(z)=[-i\omega\epsilon_{xx}/\alpha_A][A_{1A}^{(n)}\exp(-\alpha_A z)-A_{2A}^{(n)}\exp(\alpha_A z)], \tag{7}$$

$$E_{xB}^{(n)}(z)=A_{1B}^{(n)}\exp(-\alpha_B z)+A_{2B}^{(n)}\exp(\alpha_B z), \tag{8}$$

$$H_{yB}^{(n)}(z)=[-i\omega\epsilon_B/\alpha_B][A_{1B}^{(n)}\exp(-\alpha_B z)-A_{2B}^{(n)}\exp(\alpha_B z)], \tag{9}$$

where we have denoted

$$\alpha_A^2=\gamma^2 q_x^2-\epsilon_{xx}\omega^2/c^2, \quad \alpha_B^2=q_x^2-\epsilon_B\omega^2/c^2 \tag{10}$$

with $\gamma=(\epsilon_{xx}/\epsilon_{zz})^{1/2}$ for the anisotropic medium.

The surface- and bulk-polariton modes of the superlattice can now be found by a straightforward transfer-matrix calculation (for full descriptions of the method see Ref. 3 or 9), generalized here to include the anisotropy in medium A. Following the procedure in Ref. 3, the transfer matrix **T** relates the coefficients of the electric field in one unit cell to those in the preceding cell according to

$$\begin{pmatrix} A_{1A}^{(n+1)} \\ A_{2A}^{(n+1)} \end{pmatrix}=\mathbf{T}\begin{pmatrix} A_{1A}^{(n)} \\ A_{2A}^{(n)} \end{pmatrix}. \tag{11}$$

The 2×2 matrix **T** can be written in product form as $\mathbf{T}=\mathbf{N}_A^{-1}\mathbf{M}_B\mathbf{N}_B^{-1}\mathbf{M}_A$, where

$$\mathbf{M}_J=\begin{pmatrix} f_J & 1/f_J \\ \xi_J f_J & -\xi_J/f_J \end{pmatrix}, \quad \mathbf{N}_J=\begin{pmatrix} 1 & 1 \\ \xi_J & -\xi_J \end{pmatrix}, \tag{12}$$

with $J=A$ or B. Here we denote $f_J=\exp(-\alpha_J d_J)$, $\xi_A=\epsilon_{xx}/\alpha_A$, and $\xi_B=\epsilon_B/\alpha_B$.

The bulk superlattice polaritons are obtained by introducing a Bloch wave number Q, giving rise to the formal dispersion relation (e.g., see Ref. 3)

$$\cos(QL)=\tfrac{1}{2}\mathrm{Tr}(\mathbf{T}). \tag{13}$$

The general dispersion relation is rather complicated, but for values of q_x where retardation effects can be ignored ($\alpha_A\simeq\gamma q_x$, $\alpha_B\simeq q_x$) the result simplifies to

$$\cos(QL)=\cosh(\gamma q_x d_A)\cosh(q_x d_B) + S(\omega)\sinh(\gamma q_x d_A)\sinh(q_x d_B), \tag{14}$$

where

$$S(\omega)=[(\gamma\epsilon_B/\epsilon_{xx})+(\gamma\epsilon_B/\epsilon_{xx})^{-1}]/2. \tag{15}$$

The calculation of the surface-polariton modes in the transfer-matrix method[3] involves formally replacing Q in Eq. (13) by $i\beta$, where β describes the decay characteristics (with $\mathrm{Re}\beta>0$ for localization). Also, another condition for β is obtained by considering the boundary conditions at $z=0$ (the interface between the vacuum and the surface layer of the anisotropic medium A). In the present case this leads to

$$T_{11}-T_{22}-\lambda T_{12}-\lambda^{-1}T_{21}=0, \tag{16}$$

where

$$\lambda=(\epsilon_{xx}\alpha_C+\alpha_A)/(\epsilon_{xx}\alpha_C-\alpha_A), \tag{17}$$

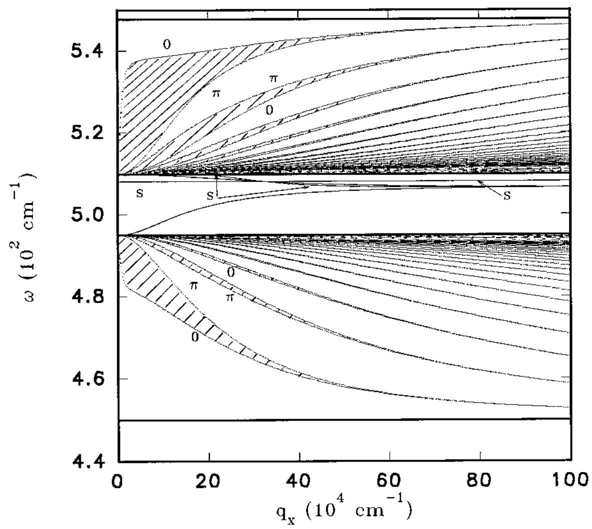

FIG. 3. Bulk and surface polaritons in a semi-infinite binary superlattice where one of the constituents (A) is α-quartz, with its optic axis parallel to the x axis. Here $d_A=100$ nm, $d_B=50$ nm, and $\epsilon_B=1.5$.

with $\alpha_C^2=q_x^2-\omega^2/c^2$. Equations (16) and (13), with $Q\rightarrow i\beta$, represent implicit equations which can be solved to obtain β and/or ω for the surface modes.

Suppose that ϵ_B is positive, so that the optically active medium is the anisotropic one (medium A), taken to be α-quartz. Figure 3 shows the bulk- and surface-polariton frequencies plotted against q_x for the superlattice for the case where the optic axis of the α-quartz is parallel to the x axis. The bulk modes of the superlattice occur as bands with edges corresponding to the lines $QL=0$ and $QL=\pi$. For relatively small q_x the bulk bands are wide and are shown shaded, while for large q_x they are narrow. The surface modes of the superlattice occur only in region IV (defined as in Sec. II) in this case and are labeled by S. For completeness, in Figs. 4 and 5 we also show the corresponding cases for superlattices where the optic axis of α-quartz is oriented parallel to the y

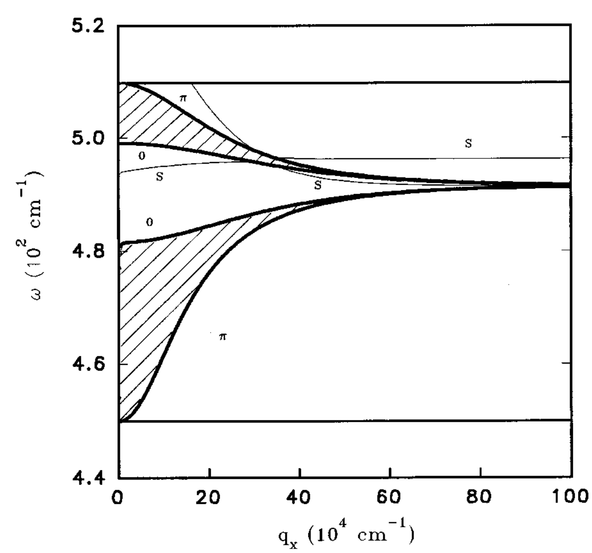

FIG. 4. As in Fig. 3, but with the optic axis parallel to the y axis.

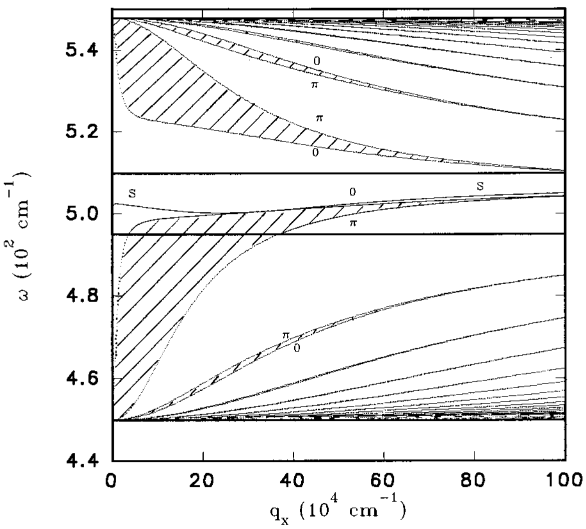

FIG. 5. As in Fig. 3, but with the optic axis parallel to the z axis.

and z axes, respectively. These figures demonstrate how strongly dependent the polariton spectra are on the orientation of the anisotropy axis to the z axis (normal to the layers) and the x axis (direction of $q_{\parallel}$). When the optic axis is parallel to the y axis (in Fig. 4) the spectrum is more similar to that for a superlattice composed of isotropic media (e.g., see Ref. 3), since $\gamma=1$ in this case. In Figs. 6 and 7 the frequencies of the bulk modes are plotted against QL/π for the cases of the optic axis parallel to the x and z axes, respectively. Results are shown in both figures for two different values of q_x. For $q_x=5\times10^4$ cm^{-1} the bulk bands are wide, as mentioned previously, and so the corresponding curves in Figs. 6 and 7 show appreciable dispersion, whereas for $q_x=50\times10^4$ cm^{-1} the bulk bands are narrow and the curves are relatively flat.

Finally, we note that the above numerical examples for superlattices correspond to cases where the effects of retardation are relatively small, since typically $q_x \gg \omega/c$. In Fig. 8, however, we show an example of bulk- and surface-polariton frequencies plotted against q_x (for a fixed value of QL) in the region $q_x \leqslant 1\times10^4$ cm^{-1}, where the retardation effects on the modes are important. As one can infer, a comparison of this figure with Fig. 5 (where retardation effects were not considered) shows that only the surface mode (S) is sensitive to retardation effects near the light line. Indeed, it is clear now that this mode does not have a flat behavior, but that it bends toward the light line, for the range of $q_x=0.05\times10^4$ cm^{-1} (where it merges) to $q_x=0.2\times10^4$ cm^{-1}. The bulk bands, on the other hand, are largely unaffected by the retardation effects, as we might expect for this kind of polariton mode.

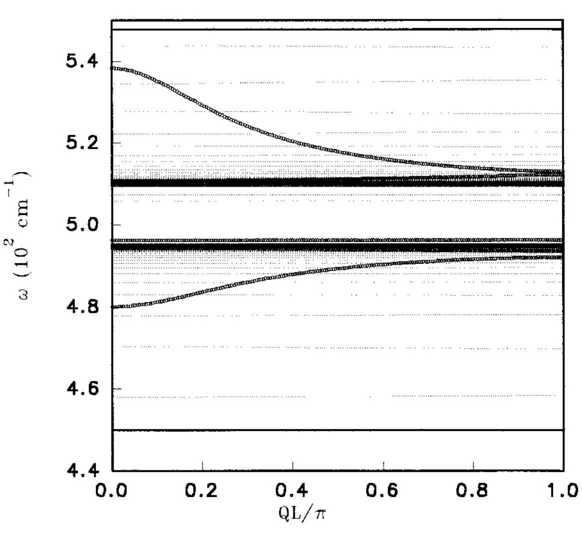

FIG. 6. Bulk polariton frequencies plotted against QL/π for a binary superlattice with the optic axis of α-quartz parallel to the x axis. We have taken two values of q_x: 5×10^4 cm^{-1} (○○○) and 50×10^4 cm^{-1} (⋯). Other parameters are as in Fig. 3.

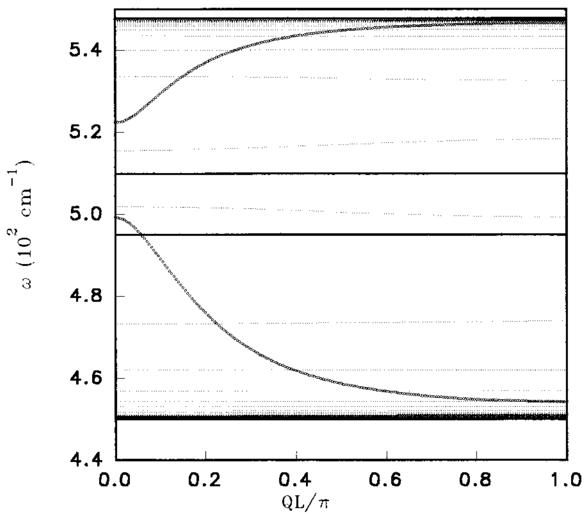

FIG. 7. As in Fig. 6, but with the optic axis in the z direction.

IV. CONCLUSIONS

We have presented calculations for the bulk and surface polaritons in multilayer structures, particularly semi-infinite

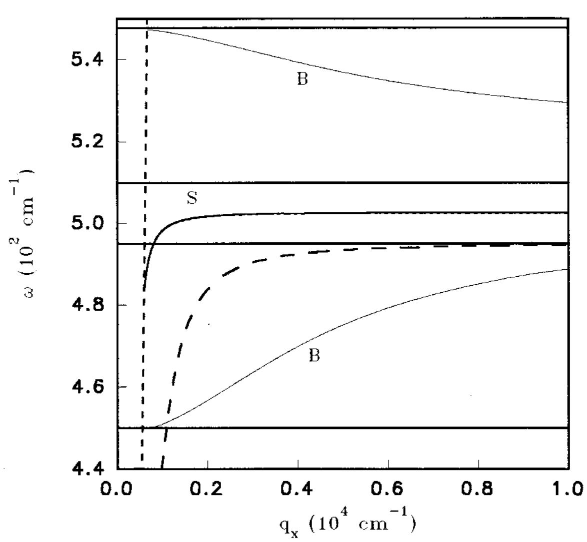

FIG. 8. Bulk (B) and surface (S) polaritons for a binary superlattice, as in Fig. 5, but now we focus on the region where retardation effects are important ($q_x \leqslant 1\times10^4$ cm^{-1}), taking $QL/\pi=0.02$ for the bulk modes.

binary superlattices in which one of the materials has an anisotropic dielectric function, thereby generalizing earlier work on isotropic superlattices and on anisotropic slabs. Using α-quartz as an example, the effects of anisotropy on the phonon-polariton spectra were shown to be important, and dependent on the orientation of the optic axis of α-quartz relative to the direction of the in-plane propagation $q_{\parallel}$ and the normal to the layers. This was the case even in the wave-vector regime where retardation effects become small.

It would be of interest to have experimental data to test our predicted anisotropy dependence of the polariton spectra in superlattices. Structures composed of layers of α-quartz, alternating with an isotropic inactive material, would be suitable. Appropriate experimental techniques could use Raman scattering or attenuated total reflection, or the recent advances in far-infrared Fourier-transform spectroscopy.[10]

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*Permanent address: Departamento de Física, Universidade Federal do Ceará, 60455-760, Fortaleza (Ce), Brazil.

†Permanent address: Departamento de Física, Universidade Federal do Rio Grande do Norte, 59072-970, Natal (RN) Brazil.

[1] M. G. Cottam and D. R. Tilley, *Introduction to Surface and Superlattice Excitations* (Cambridge University Press, Cambridge, 1989).

[2] *Interfaces, Quantum Wells and Superlattices*, edited by C. R. Leavens and R. Taylor (Plenum, New York, 1988).

[3] E. L. Albuquerque and M. G. Cottam, Phys. Rep. **233**, 67 (1993).

[4] *Molecular Beam Epitaxy and Heterostructures*, edited by L. L. Chang and K. Ploog (Plenum, New York, 1985).

[5] H. J. Falge and A. Otto, Phys. Status Solidi B **56**, 523 (1973).

[6] G. Borstel and H. J. Falge, in *Electromagnetic Surface Modes*, edited by A. D. Boardman (Wiley, New York, 1982).

[7] G. Borstel, Z. Naturforsch. **29a**, 1306 (1974).

[8] A. Harstein, E. Burstein, J. J. Brian, and R. F. Wallis, Solid State Commun. **12**, 1083 (1973).

[9] G. A. Farias, M. M. Auto, and E. L. Albuquerque, Phys. Rev. B **38**, 12 540 (1988).

[10] T. Dumelow, D. E. Brown, and T. J. Parker, Proc. SPIE **2104**, 633 (1993).