

Theory of coupled phason and sound-wave normal modes in the incommensurate phase of quartz

M. B. Walker and R. J. Gooding

Department of Physics and Scarborough College, University of Toronto, Toronto, Ontario, Canada M5S 1A7

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A phenomenological theory for coupled phasons and sound waves in the presence of viscous damping of the relative phase motion is developed for the incommensurate phase of quartz. For weak damping, five propagating normal modes occur, whereas for strong damping we find propagating nearly pure sound waves, fast relaxing nearly pure relative phase motion, and diffusive coupled sound-wave and relative phase motion. Technical points of interest are the introduction of a relative phase-displacement field, and the discussion of the rotational invariance of the free energy.

I. INTRODUCTION

It has been shown by Walker¹ that the long-wavelength Overhauser² phason excitations characteristic of an incommensurate phase are coupled to the long-wavelength sound-wave modes, and that both sound waves and phasons must be treated together in a coherent framework. In studies by Golovko and Levanyuk³ and Zeyher and Finger,⁴ which have ignored this coupling, it was shown that the nature of the phason is strongly affected by viscous damping forces which cause the mode to have a diffusive character at long wavelengths. Thus, it is evident that a study of the low-frequency excitations of incommensurate structures should include both the coupling of phasons to sound waves as well as viscous damping forces. (See *Note added in proof.*) This article develops such a theoretical model for the coupled sound-wave-phason systems, in the incommensurate phase of quartz. Another example of a coherent treatment of sound waves and phasons (which, however, neglects viscous damping forces) is provided in an article by Axe and Bak.⁵ Currat⁶ has reviewed both experimental and theoretical studies of phasons.

In the incommensurate phase of quartz, the phason results from a sliding motion of a spatially modulated optical mode. In formulating the problem of coupled sound-wave and phason motions, it is important to use coordinates which describe the motion of the modulation wave relative to that of the underlying crystal lattice, and not its motion relative to some fixed laboratory frame of reference. When this is done the kinetic energy separates into a sum of contributions representing a contribution from the motion of the underlying lattice and the modulated mode together, and one from their relative motion. Furthermore, it is the relative motion which is subjected to a viscous damping proportional to its velocity. The appropriate coordinate, which will be called the relative phase displacement field U , is introduced in Sec. II. Since U lies in the basal plane it has two components and gives rise to two vibrational modes. Thus there are a total of five coupled phason-sound-wave modes in the absence of damping.

The combined effects of phason to sound-wave coupling and viscous relaxation of phasons will be shown to have some interesting consequences. For example, in the

case of coupling of a single phason and a single sound wave, there is, at sufficiently low frequencies, only a single propagating mode which is approximately pure sound-wave motion. On the other hand, at high frequencies there is a relative motion of the incommensurate domain structure and the underlying crystal and there are propagating modes corresponding to both a phason and a sound wave; the elastic interaction between the sound wave and phason pushes their frequencies apart, however, and the low-frequency sound velocity thus lies in between the two velocities appropriate for the high-frequency region.

Another point of interest is that the high-frequency propagating modes in the incommensurate phase of quartz for the case where both the wave vector and the polarizations lie in the basal plane, are in general neither purely longitudinal nor purely transverse. This contrasts with the usual situation in crystals with hexagonal symmetry where sound waves propagating in the basal plane are either longitudinal or transverse.

The incommensurate phase of quartz is essentially a triangular Dauphiné-twin domain structure.⁷ The Dauphiné twins are the two degenerate ground states of the low-

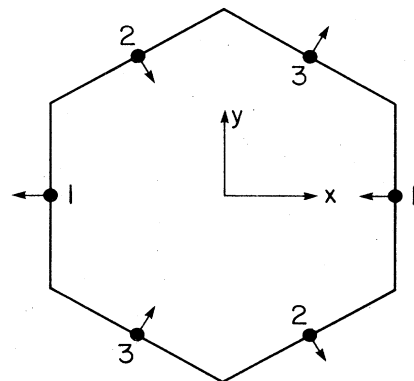


FIG. 1. Basal-plane projection of the positions of the silicon ions in the Wigner-Seitz cell of the quartz structures. The solid circles give the silicon positions in the β phase, whereas the arrows give the directions of the silicon displacements in a transition to the α phase. The direction of the x coordinate axis of Fig. 2 is also defined here.

temperature α phase of quartz. Figure 1 illustrates the displacements of the silicon ions in the α phase relative to their high-temperature β -phase positions.⁸ The order parameter η describing the phase transitions from the β to the incommensurate and α phases can be taken to have a magnitude equal to the magnitude of a given silicon-ion displacement and to be positive if the silicon-ion displacements are in the direction shown, and negative if these displacements are in the opposite direction. The two possible signs of η correspond to the two Dauphiné twins. In electron microscope images, one of the Dauphiné twins is black and the other is white, thus allowing electron microscope observations of Dauphiné-twin domain configurations.^{7,9,10}

The incommensurate phase of quartz is shown schematically in Fig. 2 (e.g., see Walker¹¹). There are two degenerate possibilities for the ground state corresponding to different orientations of the triangles, one shown in the upper half of Fig. 2 and the other shown in the lower half. The incommensurate phase has been observed to have a macrodomain structure, the two types of macrodomains corresponding to the two possible orientations of the triangles. In this article we assume that the crystal is composed of a single large macrodomain. It is perhaps possible to prepare such a sample by cooling through the β phase to the incommensurate-phase phase transition in an electric field directed along the c axis and thus making use of the ferroelectric properties^{12,13} of the macrodomains.

Now note that, since black regions are converted to white regions by a rotation of $2\pi/6$, the c axis is a sixfold axis of symmetry of the incommensurate phase. This is the only point-group symmetry element of the incommensurate phase. It will be shown below that the free energy describing the linear long-wavelength elastic properties of the coupled phasons and sound waves has a higher symmetry than the incommensurate phase itself, and these additional symmetries will be used to help simplify the study of the normal modes.

II. THE RELATIVE PHASE-DISPLACEMENT FIELD

The incommensurate phase can be described by the order parameter having the form

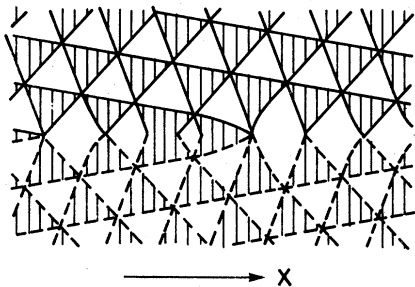


FIG. 2. Basal-plane projection of the incommensurate domain structure of quartz shows the two differently oriented macrodomains, one in the top half of the figure, and the other in the bottom half.

$$\eta(\mathbf{r}) = A \sum_{i=1}^3 \sin[\mathbf{Q}_i \cdot (\mathbf{r} - \mathbf{U}')] , \quad (1)$$

where the \mathbf{Q}_i are three wave vectors lying in the basal plane and making angles of 120° relative to one another. For the incommensurate phase of the upper half of Fig. 2, each of the three \mathbf{Q}_i 's is perpendicular to one of the three domain-wall orientations, and positive η corresponds to say black regions whereas negative η corresponds to white regions. The expression (1) with $\mathbf{U}'=0$ was obtained by Aslanyan *et al.*¹⁴

A nonzero value of the vector \mathbf{U}' in Eq. (1) corresponds to a displacement of the incommensurate domain structure by \mathbf{U}' relative to that described by Eq. (1) with $\mathbf{U}'=0$. \mathbf{U}' will be assumed to lie in the basal plane; a component of \mathbf{U}' normal to the basal plane has no physical significance as a result of its being normal to the \mathbf{Q}_i in Eq. (1). The ground state of the incommensurate phase has a continuous degeneracy resulting from the fact that the free energy is independent of \mathbf{U}' . Changing the value of \mathbf{U}' in Eq. (1) is equivalent to a certain change of the phases of the sine functions in Eq. (1) and \mathbf{U}' will be called the phase-displacement field.

The elastic properties of the crystal are described in terms of the usual elastic displacement field \mathbf{u} . A spatially uniform \mathbf{u} corresponds to a displacement of each atom in the crystal by the same vector displacement \mathbf{u} . If one starts with a crystal in which the order parameter is given by Eq. (1) with $\mathbf{U}'=0$ and then gives each atom in the crystal a basal-plane displacement \mathbf{u}_b , the new order parameter will be of the form of Eq. (1) but with $\mathbf{U}'=\mathbf{u}_b$. Therefore, we write $\mathbf{U}' - \mathbf{u}_b \equiv \mathbf{U}$, where \mathbf{u}_b is the basal-plane component of \mathbf{u} , and \mathbf{U} is called the relative phase-displacement field and represents the translation of the incommensurate domain structure relative to points fixed in the crystal lattice (these points fixed in the lattice may be thought of as the centers of mass of crystallographic unit cells, such as the one illustrated in Fig. 1).

The free energy will depend on the spatial gradients of the displacement fields \mathbf{u} and \mathbf{U} , and in order to establish the rotational invariance of the free energy the rotational significance of these gradients must be established. The rotational significance of

$$r_{\alpha\beta} = \frac{1}{2}(u_{\beta,\alpha} - u_{\alpha,\beta}) , \quad (2)$$

where

$$u_{\alpha,\beta} = \frac{\partial u_\alpha}{\partial x_\beta} , \quad (3)$$

is well known (e.g., a spatially uniform r_{zx} corresponds to a rotation of the crystal about the positive y axis by an angle r_{zx}). The rotations associated with phase strains are not quite analogous since the phase-displacement field has no z component and it is impossible to antisymmetrize $U'_{x,z}$ and $U'_{y,z}$. However, note that if $U'_{x,z}$ is spatially constant, then $U'_x = U'_{x,z}z$ plus an irrelevant constant. Equation (2) for $\eta(\mathbf{r})$ with $\mathbf{U}' = \epsilon_x U'_{x,z}z$ can be rewritten in the form

$$\eta(\mathbf{r}) = A \sum_i \sin[(\mathbf{Q}_i + \delta\mathbf{Q}_i) \cdot \mathbf{r}] , \quad (4)$$

where

$$\delta \mathbf{Q}_i = \delta \psi \times \mathbf{Q}_i, \quad \delta \psi = \epsilon_y U'_{x,z}. \quad (5)$$

Thus a constant $U'_{x,z}$ corresponds to a rotation of the incommensurate domain structure about the positive y axis by an angle $U'_{x,z}$. These and similar considerations lead us to define the relative rotational components of the strains by

$$\begin{aligned} R_{xz} &= U'_{x,z} - r_{xz}, \\ R_{yx} &= \frac{1}{2}(U'_{y,x} - U'_{x,y}) - r_{yx}, \\ R_{zy} &= -U'_{y,z} - r_{zy}. \end{aligned} \quad (6)$$

These quantities describe rotations of the incommensurate domain structure relative to the underlying crystal lattice about the positive y , z , and x axes, respectively.

Later on, the notation

$$\begin{aligned} e_{\alpha,\beta} &= \frac{1}{2}(u_{\alpha,\beta} + u_{\beta,\alpha}), \quad E_{xy} = \frac{1}{2}(U_{x,y} + U_{y,x}), \\ E_{xx} &= U_{x,x}, \quad E_{yy} = U_{y,y} \end{aligned} \quad (7)$$

will also be used.

$$\begin{aligned} F &= A_1(e_{xx} + e_{yy})^2 + A_2[(e_{xx} - e_{yy})^2 + 4e_{xy}^2] + A_3e_{zz}^2 + A_4e_{zz}(e_{xx} + e_{yy}) + A_5(e_{zx}^2 + e_{yz}^2) + A_6(E_{xx} + E_{yy})^2 \\ &+ A_7[(E_{xx} - E_{yy})^2 + 4E_{xy}^2] + A_8(E_{xx} + E_{yy})(e_{xx} + e_{yy}) + A_9[(E_{xx} - E_{yy})(e_{xx} - e_{yy}) + 4E_{xy}e_{xy}] \\ &+ A_{10}[(e_{xx} - e_{yy})E_{xy} - (E_{xx} - E_{yy})e_{xy}] + A_{11}e_{zz}(E_{xx} + E_{yy}) + A_{12}R_{yx}^2 + A_{13}(R_{xz}^2 + R_{zy}^2) + A_{14}(E_{xx} + E_{yy})R_{yx} \\ &+ A_{15}(e_{xx} + e_{yy})R_{yx} + A_{16}e_{zz}R_{yx} + A_{17}(e_{zx}R_{xz} - e_{yz}R_{zy}) + A_{18}(e_{zx}R_{zy} + e_{yz}R_{xz}). \end{aligned} \quad (9)$$

An immediately striking feature of Eq. (9) is the relatively large number of elastic constants (i.e., eighteen) required to describe this incommensurate phase. Another feature of Eq. (9) is that, because we have restricted ourselves to terms which are quadratic functions of the displacement gradients, it exhibits a higher point-group symmetry than the incommensurate phase itself. For example, the free energy of Eq. (9) is invariant not only with respect to c -axis rotations of $2\pi/6$, but to arbitrary c -axis rotations. As a consequence, the normal-mode frequencies will be independent of the orientation of the basal-plane component of the wave vector. The other important symmetry property of the free energy of Eq. (9) is its invariance with respect to a basal-plane reflection (i.e., the transformation $z \rightarrow -z$).

The kinetic energy per unit volume has the form

$$T = \frac{1}{2} \sum_i \rho_i \dot{u}_i^2, \quad (10)$$

where $\rho_1 = \rho_3 = \rho_5 = \rho$ and $\rho_2 = \rho_4 = \rho^*$. Here ρ is the mass per unit volume, and ρ^* is an effective mass per unit volume associated with relative phase displacements. That $\rho_2 = \rho_4$ follows from the sixfold symmetry of the incommensurate phase. Note that the kinetic energy has this simple form because we are using \mathbf{u} and the relative displacement field \mathbf{U} as independent variables. The kinetic energy in terms of \mathbf{u} and \mathbf{U}' looks quite different and can be obtained by substituting $\mathbf{U} = \mathbf{U}' - \mathbf{u}_b$ in Eq. (10).

III. THE FREE ENERGY AND THE EQUATIONS OF MOTION

The free energy per unit volume is expanded to second order in the displacement gradients, giving

$$F = \frac{1}{2} c_{i\alpha j\beta} u_{i,\alpha} u_{j,\beta}, \quad (8)$$

where the summation convention is used, indices i, j take values 1, 2, 3, 4, 5 while $\alpha, \beta = x, y, z$, and the five-component displacement field u_i is defined by

$$(u_1, u_2, u_3, u_4, u_5) \equiv (u_x, U_x, u_y, U_y, u_z).$$

The free energy F is constructed in such a way as to be invariant with respect to sixfold rotations about the c axis (the only point-group symmetry element of the incommensurate phase). Furthermore, although the free energy must be invariant with respect to simultaneous identical rotations of the incommensurate domain structure and the underlying crystal lattice, it must depend on the relative rotational variables defined in Eq. (6).

Thus, in terms of the variables defined in Eqs. (6) and (7),

The equations of motion are

$$\frac{\partial}{\partial t} \left[\frac{\partial T}{\partial \dot{u}_i} \right] = \frac{\partial}{\partial x_\alpha} \left[\frac{\partial F}{\partial u_{i,\alpha}} \right] - \frac{\partial \Psi}{\partial u_i}, \quad (11)$$

where Ψ is a dissipation function which will be taken equal to zero for the moment. The normal-mode solutions of Eq. (11) have the form

$$u_i(\mathbf{r}, t) = u_{i0} \exp[i(\mathbf{q} \cdot \mathbf{r} - \omega t)]. \quad (12)$$

The normal-mode frequencies are determined by the eigenvalue equation

$$\omega^2 w_i = q^2 d_{ij}(\hat{\mathbf{q}}) w_j, \quad (13)$$

where $w_i = (\rho_i)^{1/2} u_i$, q is the magnitude of \mathbf{q} , and $\hat{\mathbf{q}}$ is a unit vector in the direction of \mathbf{q} . The matrix d_{ij} is given by

$$d_{ij}(\hat{\mathbf{q}}) = (\rho_i \rho_j)^{-1/2} \sum_{\alpha, \beta} c_{i\alpha j\beta} \hat{q}_\alpha \hat{q}_\beta, \quad (14)$$

where i and j are not summed over. The matrix d_{ij} is a real symmetric 5×5 matrix which can be diagonalized by an orthogonal transformation to give the frequencies of the five normal modes for a given \mathbf{q} .

The dissipation function is chosen to be

$$\Psi = \frac{1}{2} \frac{\rho^*}{\tau} \frac{\partial \mathbf{U}}{\partial t} \cdot \frac{\partial \mathbf{U}}{\partial t}. \quad (15)$$

If the elastic free energy F is set equal to zero in Eq. (11), the equation of motion for the relative phase-displacement field is

$$\frac{d^2\mathbf{U}}{dt^2} = -\frac{1}{\tau} \frac{d\mathbf{U}}{dt} \quad (16)$$

Thus, the dissipation function Ψ has been chosen so as to give a viscous damping of the motion of the incommensurate domain structure relative to that of the underlying crystal lattice. We shall be particularly interested in the effects of damping at very low frequencies, and therefore neglect the conventional¹⁵ contributions to the dissipation function quadratic in the time derivative of the displacement gradients.

IV. NORMAL MODES FOR \mathbf{q} PARALLEL TO THE C AXIS

For \mathbf{q} parallel to the c axis the matrix d_{ij} can be shown by symmetry arguments to have the form

$$\begin{pmatrix} a & b & 0 & d & 0 \\ b & c & -d & 0 & 0 \\ 0 & -d & a & b & 0 \\ d & 0 & b & c & 0 \\ 0 & 0 & 0 & 0 & e \end{pmatrix} \quad (17)$$

If desired, the parameters in this matrix can be found in terms of the elastic constants A_3 , A_5 , A_{13} , A_{17} , and A_{18} .

The variable $w_5 = \rho^{1/2}u_z$ is decoupled from the others. This variable is associated with a longitudinal sound-wave mode of velocity $v_l = e^{1/2}$. The remaining variables w_1 , w_2 , w_3 , and w_4 are coupled together and give rise to two pairs of doubly degenerate transverse modes, the two distinct velocities being

$$v_{\pm} = \frac{1}{2} \{ (a+c) \pm [(a-c)^2 + 4(b^2+d^2)]^{1/2} \} \quad (18)$$

If d [which is proportional to A_{18} of Eq. (9)] were equal to zero the polarization directions \mathbf{u} and \mathbf{U} for a given mode would be parallel, but since d is not, in general, zero, this is not, in general, the case. The twofold degeneracy for each transverse mode results from the fact that, given a transverse mode, a rotation of $\pi/2$ about the c axis produces another linearly independent transverse mode having the same frequency.

Now add the damping of the relative coordinate \mathbf{U} to the problem by combining Eqs. (11)–(15) and (17). The longitudinal mode is unchanged, but the transverse-mode frequencies are found to be given by the solutions of

$$(p^2 - v_+^2)(p^2 - v_-^2) + ipB(p^2 - v^2) = 0, \quad (19)$$

where

$$p = \omega/q, \quad B = (q\tau)^{-1}, \quad v = \sqrt{a}. \quad (20)$$

Note that

$$v_+ > v > v_- \quad (21)$$

The four solutions of Eq. (19) can easily be found in the short-wavelength or weak-damping limit $v'q\tau \gg 1$ and in the long-wavelength or strong-damping limit $v'q\tau \ll 1$

(here v' is the order of magnitude of v_+ , v_- , or v). In the short-wavelength case the four frequencies are

$$\omega = \pm v_+ q - \frac{i}{2\tau} \left(\frac{v_+^2 - v^2}{v_+^2 - v_-^2} \right), \quad (22)$$

$$\omega = \pm v_- q - \frac{i}{2\tau} \left(\frac{v_-^2 - v^2}{v_+^2 - v_-^2} \right). \quad (23)$$

In the long-wavelength case the four frequencies are

$$\omega = \pm vq - i \left[\frac{(v_+^2 - v^2)(v^2 - v_-^2)\tau}{2v^2} \right] q^2, \quad (24)$$

$$\omega = -i \left[\frac{v_+^2 v_-^2 \tau}{v^2} \right] q^2, \quad (25)$$

$$\omega = -i/\tau. \quad (26)$$

In the short-wavelength case one finds modes propagating with the same velocities as obtained in Eq. (18) in the absence of damping. In the long-wavelength case one obtains a single (twofold degenerate) propagating mode with the interesting feature that its velocity v lies in between the velocities of the two high-frequency modes. At low frequencies there is also a diffusive mode [Eq. (25)] and a solution representing the rapid relaxation of the relative coordinate \mathbf{U} [e.g., see Eq. (16)].

An examination of the eigenvectors in the long-wavelength limit shows that for the strongly damped mode [Eq. (26)] $w \sim (kv\tau)^2 W$, for the propagating mode [Eq. (24)] $w \sim (kv\tau)^{-1} W$, and for the diffusive mode $w \sim W$. [Here $\mathbf{w} \equiv \rho^{1/2}\mathbf{u}$ and $\mathbf{W} \equiv (\rho^*)^{1/2}\mathbf{U}$.] Thus for $kv\tau \ll 1$, the strongly damped mode is approximately pure relative phase motion, the propagating mode is approximately pure sound-wave motion, and the diffusive mode involves coupled relative phase and sound-wave motion.

V. NORMAL MODES FOR \mathbf{q} IN THE BASAL PLANE

Because the free energy of Eq. (8) is invariant with respect to arbitrary rotations about the c axis, the normal-mode frequencies are independent of the basal-plane orientation of \mathbf{q} . We arbitrarily take \mathbf{q} to be in the x direction.

Because the basal plane is a plane of reflection symmetry of the free energy, the coordinate $w_5 = \sqrt{\rho}u_z$ is not coupled to the remaining coordinates. This coordinate thus yields an undamped [in the approximation of Eq. (15)] propagating transverse mode with dispersion relation $\omega = \pm v'_l q$.

The remaining four coordinates are all coupled to one another and in the absence of damping give modes propagating with four distinct velocities since there is no symmetry to cause degeneracies. It is necessary to solve a quartic equation to obtain expressions for these frequencies as a function of the elastic constants. Furthermore, the polarization vectors \mathbf{u}_0 and \mathbf{U}_0 associated with these modes, while lying in the basal plane, will, in general, be neither parallel nor perpendicular to \mathbf{q} .

There is one special somewhat artificial case for which

analytic results for the frequencies of the modes polarized in the basal plane can be obtained. If A_{10} , A_{14} , and A_{15} in Eq. (9) are negligibly small, the remaining terms in Eq. (9) which are nonzero for \mathbf{q} in the basal plane are invariant with respect to a reflection in any plane containing the c axis. When this additional symmetry is present, the matrix d_{ij} has the simple block-diagonal form

$$\begin{pmatrix} a & b & 0 & 0 & 0 \\ b & c & 0 & 0 & 0 \\ 0 & 0 & d & e & 0 \\ 0 & 0 & e & f & 0 \\ 0 & 0 & 0 & 0 & g \end{pmatrix}. \quad (27)$$

The 2×2 matrix in the upper left-hand corner couples u_x and U_x and determines the velocities v_{l+} and v_{l-} of two longitudinal modes. When damping is included, there is, in the long-wavelength limit, a single propagating longitudinal mode with a velocity $v_l = \sqrt{a}$ such that $v_{l+} > v_l > v_{l-}$. A complete description of the frequencies of the modes is given by Eqs. (22)–(26) of the preceding section with v_+ , v , and v_- replaced by v_{l+} , v_l , and v_{l-} , respectively.

Similarly, the central 2×2 matrix in Eq. (27) couples u_y and U_y and determines the short-wavelength velocities v_{t+} , v_{t-} of two propagating transverse modes. The frequencies of these modes in the presence of damping are given by Eqs. (22) to (26) with v_{t+} , $v_t = \sqrt{d}$, and v_{t-} replacing v_+ , v , and v_- , respectively.

Finally, it should be emphasized that the analytical results just described for \mathbf{q} in the basal plane are for the special case $A_{10} = A_{14} = A_{15} = 0$. Whether or not this is a good approximation can be determined by experimentally finding the polarization of the modes.

VI. CONCLUSIONS

A phenomenological model determining the coupled sound-wave phason modes in the incommensurate phase of quartz was developed in terms of the usual sound-wave

displacement field \mathbf{u} and a relative phase-displacement field \mathbf{U} . In the absence of damping the coupled system has five normal modes for which the frequency varies linearly with the magnitude of the wave vector.

For the case of \mathbf{q} parallel to the c axis, analytical results are easily obtained. There is one longitudinal mode (with both \mathbf{u} and \mathbf{U} parallel to \mathbf{q}) and there are two doubly degenerate pairs of transverse modes (with both \mathbf{u} and \mathbf{U} perpendicular to \mathbf{q}). Viscous damping of the relative phase-displacement field \mathbf{U} strongly affects the character of the transverse modes at long wavelength. Whereas at relatively short wavelengths there are two doubly degenerate propagating transverse modes with two distinct velocities, v_+ and v_- , at long wavelengths, there is only one doubly degenerate propagating mode; this mode has a velocity v such that $v_+ > v > v_-$ and is approximately pure sound-wave motion. There is also a doubly degenerate diffusive mode which involves coupled sound-wave and relative phase motion, and a fast relaxing mode which is approximately pure relative phase motion.

The case of \mathbf{q} lying in the basal plane is more difficult to analyze analytically in that for modes polarized in the basal plane an equation of eighth order in the frequency must be solved in the general case, and if damping is neglected, a quartic equation must be solved. An interesting result for this case is that modes polarized in the basal plane are, in general, neither purely longitudinal nor purely transverse, although they may be approximately so if certain elastic constants are sufficiently small; if these elastic constants are neglected completely, analytical results similar to the case for \mathbf{q} parallel to the c axis are obtained.

Note added in proof. Other relevant studies include Refs. 16–18.

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