PHYSICAL REVIEW B VOLUME 28, NUMBER 1 1 JULY 1983

Long-wavelength phonons in incommensurate systems

W. Finger
Max-Planck-Institut für Festkörperforschung, D-7000 Stuttgart 80, Federal Republic of Germany

T. M. Rice
Institut für Theoretische Physik, Eidgenössische Technische Hochschule, CH-8093 Zürich, Switzerland
(Received 13 January 1983)

The dynamic structure factors of crystals with sublattices which are mutually incommensurate are examined. The sublattices may be individually ordered, but incommensurate, or only one may be ordered with an interpenetrating fluid of the second atomic species. At high frequencies there are two propagating longitudinal-acoustic modes arising from the separate subsystems but in the long-wavelength limit there is only one propagating longitudinal-acoustic mode. The form of the crossover from high- to low-frequency behavior and also the detailed form of the low-frequency-mode structure depends on the relative importance of impurities and the intrinsic processes which relax the relative linear momentum of the two subsystems.

I. INTRODUCTION

In an incommensurate system there are new normal modes due to the relative motion of the subsystems. A particular class of such systems consists of two interpenetrating sets of atoms which are mutually incommensurate. In this case, due to their incommensurability, the subsystems can slide past each other and these new degrees of freedom allow separate phonons in each atomic system. However, in the true long-wavelength limit one expects the system to have only one propagating set of acoustic modes corresponding to the translational symmetry of the system as a whole. Therefore, there should be two dynamical regimes and a crossover from the uncoupled phonons at high frequencies to the combined motion at low frequencies. This crossover in the dynamical behavior is the subject of this paper.

Our interest in this problem was stimulated by a comprehensive neutron scattering examination of the properties of the compound $Hg_{3-\delta}AsF_6$ by Pouget *et al.*[1] and Heilmann *et al.*[2] The latter group found that there are two sets of longitudinal-acoustic (LA) phonons whose motion is polarized along the Hg chains. In this compound the Hg atoms are arranged on two sets of chains with an average spacing which is incommensurate with the lattice of AsF_6 ions. At room temperature the Hg chains are found to be one-dimensional liquids with only short-range order though with a large coherence length. At such temperatures Heilmann *et al.*[2] found that the two LA modes remained uncoupled and propagating down to the lowest frequencies they could study. At lower temperatures there is a transition in which the two perpendicular sets of Hg chains order with each other while remaining incommensurate with the AsF_6 lattice. An elegant theory of this transition has been given by Emery and Axe.[3]

Recently Axe and Bak[4] have analyzed the unusual elastic properties of the low-temperature phase. They show that even in the ordered low-temperature phase there can be a low-frequency mode involving relative motion of the Hg and AsF_6 lattices with a correlated motion of the two sets of Hg chains. In a recent Letter[5] we discussed the crossover in the dynamics of the LA modes in the high-temperature phase of $Hg_{3-\delta}$ AsF_6. We showed that due to the exchange of phonons at points in k space where the phonons of two subsystems are degenerate in energy, there is an intrinsic coupling between the long-wavelength LA phonons of the two subsystems. This coupling leads to a crossover between two propagating LA modes at high frequency and one propagating combined LA mode and a diffusive excitation due to particle motion at low frequency. The crossover is estimated to occur on a frequency scale $\simeq 1$ MHz.[5]

In this paper we give a more detailed account of our work and extend it in several ways. First, we consider the case of two interpenetrating incommensurate lattices. In this case also there is a similar intrinsic coupling mechanism between the very-long-wavelength LA modes. Second, we consider the extrinsic coupling mechanism in the presence of impurities. We analyze the case of weak pinning following Fukuyama and Lee[6] and show that in this case the low-frequency diffusive mode is raised to a

finite-frequency pinning mode. We also discuss the combined effects of intrinsic and extrinsic coupling.

Independently of our work Brand and Bak[7] have considered the hydrodynamic equations of $Hg_{3-\delta}AsF_6$ and related systems. They also proposed that relative momentum density of the Hg and AsF_6 systems in the high-temperature phase is not conserved but has a characteristic relaxation time and this leads to the same conclusion, as we obtained from our microscopic approach, for the modes in the long-wavelength limit. Our results presented here show that a similar crossover will occur in a mode involving relative motion of the two incommensurate lattices. We therefore conclude that there are no new propagating long-wavelength modes in the low-temperature phase of $Hg_{3-\delta}AsF_6$.

Zeyher and Finger[8] recently have shown that this overdamped character of the Goldstone modes associated with incommensurability is general in such systems. They show that while selective motion may be a symmetry of the thermodynamic state of the system, the corresponding relative momentum does not commute with the Hamiltonian and so is not a conserved density in the hydrodynamic regime, leading to overdamped Goldstone modes.

The organization of the paper is as follows. Section II is an introductory section in which we describe the formalism and the basic approximations. In Sec. III we discuss incommensurate interpenetrating lattices and the intrinsic mechanisms which couple LA modes at long wavelengths. This is followed, in Sec. IV, by a consideration of impurity effects and a discussion of the extrinsic coupling mechanisms that they cause. In Sec. V we consider the case where one of the subsystems does not have long-range order and apply the results to the room-temperature phase of $Hg_{3-\delta}AsF_6$. Our conclusions are in Sec. VI.

II. SOUND-WAVE PROPAGATION IN AN INCOMMENSURATE CRYSTAL

We consider two interpenetrating three-dimensional (3D) lattices (R_a,R_b) which have the same lattice constants in the (y,z) planes but are incommensurate in the x direction. For the sake of simplicity the ions of each subsystem are assumed to be in one-to-one correspondence to the respective lattice points. The equilibrium positions $x_\mu(i)$, $\mu=a,b$, of the ions minimize the total free energy of the composite system. They will not, in general, coincide with R_a,R_b because of the interactions between the subsystems. Thermal agitation gives rise to fluctuating displacement fields $u_\mu(i)$, or, equivalently,

$$u_\mu(q)=\sum_i u_\mu(i)\exp[-iqR_\mu(i)] \ . \tag{2.1}$$

It is the purpose of this paper to elucidate the low-frequency dynamics of such crystals. To this end we study the dynamic susceptibility $\chi(q,k,\omega)$ of the displacement fields. $\chi(q,k,\omega)$ is a matrix with respect to the fields of the two lattices, and, in general, to their respective Cartesian components. Owing to the lack of discrete translational invariance in the incommensurate phase, and also because of possible imperfections, $\chi(q,k,\omega)$ is nondiagonal in the wave vectors of the fields. $\chi(q,k,\omega)$ is related to the dynamic structure factor $S(q,k,\omega)$,[9] which in the long-wavelength limit takes the form

$$S(q,\omega)\simeq 2k_BT\sum_{k,k'}F_\mu(-q,-k) \times q_\alpha[\mathrm{Im}\chi^{\alpha\beta}_{\mu\nu}(k,k',\omega)/\omega] \times q_\beta F_\nu(q,k') \ . \tag{2.2}$$

The sum also includes summation over $\mu,\nu=a,b$, as well as $\alpha,\beta=x,y,z$, and

$$F_\mu(q,k)=\sum_i\{\exp -i[qx^0_\mu(i)-kR_\mu(i)]\}/N_\mu \ . \tag{2.3}$$

N_μ denotes the number of ions in the μth subsystem. The form factor $F_\mu(q,k)$ accounts for the difference in the phase of plane waves at the equilibrium positions $x^0_\mu(i)$ and the lattice points $R_\mu(i)$; see Eq. (2.1). It is nondiagonal in the wave vectors due to the modulation in $x^0_\mu(i)$. In the limit $q\to 0$, however, the difference in phase at positions $x^0_\mu(i)$ and $R_\mu(i)$ is negligible and $F(q,k)$ becomes diagonal in q and k. The result is a simpler expression for $S(q,\omega)$:

$$S(q,\omega)\simeq 2k_BT\sum_{\mu,\nu}\sum_{\alpha,\beta}q_\alpha[\mathrm{Im}\chi^{\alpha\beta}_{\mu\nu}(q,q,\omega)/\omega]q_\beta \ . \tag{2.4}$$

Within linear-response theory we can express $\chi(q,k,\omega)$ as[10]

$$\underline{m}\underline{\chi}(q,k,\omega)\underline{N}^{-1}=-[\omega^2\underline{\mathbb{1}}-\underline{\Omega}^2-\omega\underline{\Pi}(\omega)]^{-1}(q,k) \ , \tag{2.5}$$

with

$$\underline{m}=\begin{bmatrix} m_a & 0 \\ 0 & m_b \end{bmatrix}\delta^{\alpha\beta}, \quad \underline{N}=\begin{bmatrix} N_a & 0 \\ 0 & N_b \end{bmatrix} . \tag{2.6}$$

According to Eq. (2.5), the matrix $\underline{\Omega}^2$ is proportional to the inverse of the static susceptibilities

$$\chi(q,k)=\chi(q,k,0),$$
$$\underline{\Omega}^2(q,k)=\underline{N}\underline{\chi}^{-1}(q,k)\underline{m}^{-1}\ , \tag{2.7}$$

while $\underline{\Pi}(q,k,\omega)$ denotes the self-energy matrix which determines the damping of the phonon modes. m_μ is the mass of the particles in subsystem μ.

We can get an explicit expression for $\Omega^2(q,k)$ by taking advantage of the fact that, in the high-temperature limit, $\chi(q,k)$ is related to a static displacement correlation function,[10]

$$\chi^{\alpha\beta}_{\mu\nu}(q,k)=\beta\langle u^\alpha_\mu(-q)u^\beta_\nu(q)\rangle,\quad \beta^{-1}=k_BT\ . \tag{2.8}$$

The dominant contributions to this correlation function arise from the harmonic interaction potential of the displacement fields. We can neglect the contributions from the anharmonic part of the potentials in the calculation of $\chi(q,k)$, since it will give only a small correction to the harmonic potentials, and obtain

$$(\underline{\Omega}^2)^{\alpha\beta}_{aa}(q,k)=\sum_{i,j}\nabla_\alpha\nabla_\beta\{V_{aa}(x^0_a(i)-x^0_a(j);i,j)(e^{iqR_a(i)}-e^{iqR_a(j)})(e^{-ikR_a(i)}-e^{-ikR_a(j)})/2$$
$$+V_{ab}(x^0_a(i)-x^0_b(j);i,j)\exp[i(q-k)R_a(i)]\}/M_a\ , \tag{2.9}$$

$$(\underline{\Omega}^2)^{\alpha\beta}_{ab}(q,k)=-\sum_{i,j}\nabla_\alpha\nabla_\beta V_{ab}(x^0_a(i)-x^0_b(j);i,j)\exp\{i[qR_a(i)-kR_0(j)]\}/M_b\ . \tag{2.10}$$

The remaining matrix elements of $\underline{\Omega}^2$ are obtained by interchanging a and b. $V_{\mu\nu}(x;ij)$ denotes the pair potential of ions i and j belonging, respectively, to subsystems μ and ν, and $\nabla_\alpha=\partial/\partial x_\alpha$. M_μ is the total mass of subsystem μ.

The self-energy $\Pi(q,k,\omega)$ vanishes in the harmonic approximation and we must retain the nonlinear forces between displacement fields to evaluate it. We restrict ourselves to contributions from third-order anharmonic potentials. Further, we approximate the self-energy as a product of two-phonon Green's functions and obtain[11]

$$\underline{\Pi}^{\alpha\beta}_{\mu\nu}(q,k,\omega)=iK_BT\sum_{q_1,q_2}\sum_{k_1,k_2}\phi^{\alpha\alpha_1\alpha_2}_{\mu\mu_1\mu_2}(q,q_1,q_2)\phi^{\beta\beta_1\beta_2}_{\nu\nu_1\nu_2}(-k,-k_1,-k_2)$$
$$\times\int_{-\infty}^{\infty}d\omega_1 G^{\alpha_1\beta_1}_{\mu_1\nu_1}(q_1,k_1,\omega_1)G^{\alpha_2\beta_2}_{\mu_2\nu_2}(q_2,k_2,\omega-\omega_1)/\pi M_\nu\ , \tag{2.11}$$

with phonon Green's functions

$$G(q,k,\omega)=[\chi(q,k)-\chi(q,k,\omega)]/\omega\ , \tag{2.12}$$

and third-order anharmonic coupling constants $\phi(q,k_1,k_2)$ whose explicit forms may be obtained by an appropriate generalization of the expressions given above for $\Omega^2(q,k)$, Eqs. (2.9) and (2.10). Again, the sums in Eq. (2.11) include summations over Cartesian and sublattice indices.

The last two equations, when combined with Eq. (2.5) for $\chi(q,k,\omega)$ constitute an infinite set of nonlinear integral equations for $\chi(q,k,\omega)$. To make progress further approximations are necessary. As we wish to calculate $\chi(q,q,\omega)$, which enters Eq. (2.4), we first derive from Eq. (2.5) an exact expression for $\chi(q,\omega)=\chi(q,q,\omega)$,

$$\underline{m}\underline{\chi}(q,\omega)\underline{N}^{-1}=-[\omega^2\underline{\mathbb{1}}-\underline{\omega}^2(q)-\omega\underline{\Pi}(q,\omega)]^{-1}\ , \tag{2.13}$$

with

$$\underline{\omega}^2(q)=\Omega^2(q,q)+\underline{\Sigma}(q,0)\ , \tag{2.14}$$

$$\Pi(q,\omega)=\Pi(q,q,\omega)+[\Sigma(q,\omega)-\Sigma(q,0)]/\omega\ . \tag{2.15}$$

and

$$\Sigma(q,\omega)=\sum_{k,k'}[\Omega^2(q,k)+\omega\Pi(q,k,\omega)]$$
$$\times\{\omega^2-Q[\Omega^2+\omega\Pi(\omega)]Q\}^{-1}(k,k')$$
$$\times[\Omega(k',q)+\omega\Pi(k',q,\omega)]\ . \tag{2.16}$$

The quantities in Eq. (2.13) are still matrices with respect to the Cartesian components of the displacement fields and the subsystems. Equation (2.13) is a very convenient expression for $\chi(q,\omega)$. $\underline{\omega}^2(q)$, which is proportional to the inverse static susceptibility $\underline{\chi}(q)=\underline{\chi}(q,\omega=0)$, Eq. (2.13),

$$\underline{\omega}^2(q)=\underline{N}\underline{\chi}^{-1}(q)\underline{m}^{-1}\ , \tag{2.17}$$

is given explicitly by Eq. (2.14), while $\Pi(q,\omega)$, Eq. (2.15), denotes the effective self-energy associated with $\chi(q,\omega)$. $\Sigma(q,\omega)$ contributes to both $\omega^2(q)$ and $\Pi(q,\omega)$ because single Fourier components with wave vector q do not diagonalize the harmonic interaction potential when the incommensurability of the crystal and impurity scattering are included. The resolvent

$$\{\omega^2-Q[\Omega^2+\omega\Pi(\omega)]Q\}^{-1}$$

in Eq. (2.16) takes the dynamics of Fourier components with wave vectors $k\neq q$ into account. The matrix elements of $Q[\Omega^2+\omega\Pi(\omega)]Q$ are defined as

$$Q[\Omega^2+\omega\Pi(\omega)]Q(k,k')$$
$$=[\Omega^2+\omega\Pi(\omega)](k,k'),\quad (k,k')\neq q \qquad (2.18)$$

and vanish otherwise.

We now introduce further approximations. The rationale is to neglect off-diagonal Green's functions $G(k,k',\omega)$ and propagators

$$\{\omega^2-Q[\Omega^2+\omega\Pi(\omega)]Q\}^{1-}(k,k'),\quad k\neq k'$$

in Eqs. (2.11) and (2.16), respectively, where they constitute only a minor correction. This is justified in cases in which the interlattice potential V_{ab}, which is responsible for the off-diagonal terms, is sufficiently weak. Further simplifications result from the fact that we are mainly interested in low-frequency dynamics. In this limit $\omega\Pi(q,k,\omega)$ can be neglected in Eq. (2.16) for $\Sigma(q,\omega)$ as compared to $\Omega^2(q,k)$.

However, the calculation of

$$\{\omega^2-Q[\Omega^2+\omega\Pi(\omega)]Q\}^{-1}(k,k)$$

still requires inversion of a large matrix. This can be achieved approximately by writing $\Sigma(q,\omega)$, Eq. (2.16), as the sum of two terms, $\Sigma_u(q,\omega)$ and $\Sigma_i(q,\omega)$, respectively,

$$\Sigma(q,\omega)=\Sigma_u(q,\omega)+\Sigma_i(q,\omega)\ , \qquad (2.19)$$

where $\Sigma_u(q,\omega)$ comprises all contributions to $\Sigma(q,\omega)$ from umklapp processes for which k differs from q by reciprocal-lattice vectors G_μ, $\mu=a,b$, while $\Sigma_i(q,\omega)$ summarizes contributions from random impurity scattering for which k is arbitrary. One has to treat $\Sigma_u(q,\omega)$ and $\Sigma_i(q,\omega)$ differently.

In the calculation of $\Sigma_u(q,\omega)$ we restrict ourselves to the special case in which only a few G_μ yield appreciable contributions and we neglect off-diagonal elements in inverting $\omega^2-Q[\Omega^2+\omega\Pi(\omega)]Q$. Introducing $\chi_0(q,\omega)$, the susceptibility in the absence of V_{ab},

$$\underline{m}\underline{\chi}_0(q,\omega)\underline{N}^{-1}=-[\omega^2\underline{\mathbb{1}}-\underline{\Omega}_0^2(\underline{q})-\omega\underline{\Pi}^0(\underline{q},\omega)]^{-1}\ , \qquad (2.20)$$

which is diagonal in both wave vectors and sublattices, one ends up with the approximate result

$$\Sigma_u(q,\omega)=\sum_{\mu,G_\mu}\Omega^2(q,G_\mu+q)m\chi_0(G_\mu+q,\omega)$$
$$\times N^{-1}\Omega^2(G_\mu+q,q)\ . \qquad (2.21)$$

This expression is equivalent to treating umklapp processes within second-order perturbation theory. $\Omega_0^2(q)$ and $\Pi^0(q,\omega)$ are the values of Ω^2 and Π defined above for $V_{ab}=0$.

In the case of $\Sigma_i(q,\omega)$ a very large number of wave vectors k contribute to the right-hand side (rhs) of Eq. (2.16) and $\omega^2-Q[\Omega^2+\omega\Pi(\omega)]Q$ is a huge matrix. Since the matrix is so large neglecting the restriction imposed on the matrix by Q, Eq. (2.18), which affects only the mode with wave vector q, should have only a minor effect on its inverse. We therefore replace

$$\{\omega^2-Q[\Omega^2+\omega\Pi(\omega)]Q\}^{-1}(k,k)$$

by

$$[\omega^2-\Omega^2-\omega\Pi(\omega)]^{-1}(k,k),$$

which according to Eq. (2.5) is equal to $-\underline{m}\,\underline{\chi}(k,\omega)\underline{N}^{-1}$, i.e.,

$$\underline{\Sigma}_i(q,\omega)=-\sum_k\underline{\Omega}^2(q,k)\underline{m}\,\underline{\chi}(k,\omega)\underline{N}^{-1}\underline{\Omega}^2(k,q)\ . \qquad (2.22)$$

Thus while $\Sigma_u(q,\omega)$ can be approximately calculated from the susceptibility in the absence of interlattice potentials, $\Sigma_i(q,\omega)$ requires a self-consistent calculation of $\chi(q,\omega)$. The origin of this difference lies in the fact that, for umklapp processes, $\omega\Pi(\omega)$ in Eq. (2.16) can be safely neglected relative to Ω^2 as $(q,\omega)\to 0$, which in these terms is the frequency squared of a phonon with wave vector G. Such a frequency is large and, therefore, not much affected by a weak interlattice potential. In the case of $\Sigma_i(q,\omega)$, however, long-wavelength fluctuations in the potential of randomly distributed impurities give rise to intermediate states with small wave vectors k for which Ω^2 in Eq. (2.16) has the same order of magnitude as $\omega\Pi(\omega)$. Thus $\Sigma_i(q,\omega)$ is strongly dependent on the precise form of $\chi(k,\omega)$.

III. IDEAL INCOMMENSURATE CRYSTAL

In this section we consider pure incommensurate crystals. First, we calculate the equilibrium positions $x_\mu^0(i)$ by minimizing the potential energy $V_{\rm pot}$,

$$V_{\rm pot}=\sum_\mu\sum_{i,j}V_{\mu\mu}(x_\mu(i)-x_\mu(i);i,j)/2$$
$$+\sum_{i,j}V_{ab}(x_a(i)-x_b(j)i,j)\ . \qquad (3.1)$$

Writing $x_\mu^0(i)$ as

$$x_\mu^0=d_\mu+R_\mu(i)+u_\mu^0(i)\ , \qquad (3.2)$$

then the relative position d_μ of the sublattices, the lattice constants (which determine $\{R_\mu\}$), and the

modulation $u_\mu^0(i)$ have to be thought of as variational parameters. If $u_\mu^0(i)$ is sufficiently small compared to the lattice constants we may minimize $V_{\rm pot}$ in two steps. In the first step we minimize with respect to d_μ and R_μ not allowing for any modulation, i.e., $u_\mu^0=0$. We then obtain for $V_{\rm pot}$

$$V_{\rm pot}=\sum_\mu V_{\mu\mu}(G_\mu)N_\mu/2V_\mu + \sum_G V_{ab}(G)\exp[iG(d_a-d_b)]N_aN_b/V ,$$

with Fourier components $V_{\mu\nu}(Q)$,

$$V_{\mu\nu}(Q)=\int d^3x\,\exp(-iQx)V_{\mu\nu}(x) . \tag{3.4}$$

V_μ is the unit-cell volume of sublattice μ, while V denotes the total volume of the system. G is a reciprocal-lattice vector common to both subsystems. As the crystal has been assumed to be incommensurate along the x direction but commensurate in the y,z planes, the only reciprocal-lattice vectors common to both subsystems lie in the y,z plane of reciprocal space with $G^x=0$. This implies that $V_{\rm pot}$, Eq. (3.3), does not depend on the relative position $(d_a-d_b)^x$ of the sublattices along the direction of incommensurability. This invariance of $V_{\rm pot}$ against any change in $(d_a-d_b)^x$ which holds true even after allowing a modulation,[12] $u_\mu^0\neq 0$, is the essential reason for the peculiar dynamical properties of an ideal incommensurate crystal.

The positions found by minimizing $V_{\rm pot}$ under the condition $u^0=0$,

$$x_\mu^0(i)=d_\mu+R_\mu(i) , \tag{3.5}$$

do not represent the ground-state configuration of $V_{\rm pot}$: The force acting on the particles occupying these positions does not vanish. If, for instance, $F_a^\alpha(q)$ denotes the α component of the force on subsystem a with wave vector q,

$$F_a^\alpha(q)=\sum_i \exp[-iqR_a(i)] \times\sum_j \nabla_\alpha[V_{aa}(x_a^0(i)-x_a^0(j);i,j)/2 + V_{ab}(x_a^0(i)-x_b^0(j);i,j] . \tag{3.6}$$

Insertion of $x_\mu^0(i)$ from Eq. (3.5) yields a nonzero result,

$$F_a^\alpha(q)=-\sum_{G_{b1}} U^\alpha(G_{b1})\Delta_a(q-G_{b1})/V , \tag{3.7}$$

with the general element

$$U^{\alpha\cdots\gamma}(k)=\sum_G i(k+G)^\alpha\cdots i(k+G)^\gamma V_{ab}(k+G) \times\exp[i(k+G)(d_a-d_b)] . \tag{3.8}$$

Here $\Delta_\mu(k)$ denotes the Kronecker δ function of sublattice μ, which is 1 for $k=G_\mu$ and zero otherwise. $G_{\mu 1}$ in Eq. (3.7) is a reciprocal-lattice vector of subsystem μ along the direction of incommensurability. The general element defined in Eq. (3.8) is symmetrical in the Cartesian components $\alpha,\ldots,\gamma$. If one of these components refers to the x direction, then, as $G^x=0$, the relation

$$U^{x\beta\cdots\gamma}(k)=ik^xU^{\beta\cdots\gamma}(k) \tag{3.9}$$

follows immediately from the definition. Substituting for $V_{ab}(k+G)$ in Eq. (3.8), its definition [Eq. (3.4)], the summation over G can be performed to give

$$U^{\alpha\cdots\gamma}(k)=V_\perp\int dx_1 e^{-ik^xx_1}\sum_{R_\perp}\nabla_\alpha\cdots\nabla_\gamma V_{ab}(x_1+d_a-d_b-R_\perp)e^{ik^\perp R_\perp} , \tag{3.10}$$

with $R_\perp$ a lattice vector in the y,z plane, and $V_\perp$ the area of the unit cell in this plane. Thus the quantity defined in Eq. (3.8) denotes the Fourier component of the $\alpha\cdots\gamma$ derivative of the potential generated along an a chain by the regular array of b particles located on a single plane perpendicular to the direction of incommensurability. It is this potential which determines the coupling of the subsystems. Its Fourier components with wave vector G_{b1} determine, according to Eq. (3.7), the force on equidistant a particles.

When distortions are allowed this force gives rise to a modulation of the equilibrium positions. Provided the force is sufficiently small, linear-response theory may be used to calculate $u_a^0(q)$ to lowest order in V_{ab}, and

$$u_a^{0\alpha}(q)=\sum_\beta \chi_{0a}^{\alpha\beta}(q)F_a^\beta(q) , \tag{3.11}$$

where $\chi_{0a}(q)$ is the static susceptibility of subsystem a in the absence of V_{ab} [Eq. (2.20) with $\omega=0$]. Fourier transformation then gives the modulation on each site,

$$u_a^{0\alpha}(i)=-\sum_{G_{b1}}\chi_{0a}^{\alpha\beta}(G_{b1})U^\beta(G_{b1}) \times\exp[iG_{b1}R_a(i)]/N_aV_b . \tag{3.12}$$

The expression for $u_b^0(i)$ can be derived in a similar way. Equation (3.12) includes summation over β.

With the approximate knowledge of the atomic positions we now proceed to calculate $\chi(q,\omega)$, Eq. (2.13). To start with, $\omega^2(q)$ can be calculated, according to Eqs. (2.14) and (2.21), from a knowledge of $\Omega^2(q,k)$, Eqs. (2.9) and (2.10). As the modulation $u_\mu^0(i)$ is assumed to be small compared to the lattice constants, the distance $x_\mu^0(i)-x_\mu^0(j)$ between adjacent ions i and j on sublattice μ differs only very little from $R_\mu(i)-R_\mu(j)$. We therefore neglect the modulation in calculating

$$V_{\mu\mu}(x_\mu^0(i)-x_\mu^0(j);i,j) .$$

This approximation is equivalent to treating the intrasublattice potential $V_{\mu\mu}(x)$ within the harmonic approximation, in which the force constants $\nabla_\mu\nabla_\nu V(x)$ are independent of changes in the ions' separations. However, due to the incommensurability of the sublattices, the variation in the separation $x_a^0(i)-x_b^0(j)$ of adjacent ions belonging to different subsystems is of the order of the lattice constants themselves. Thus the dependence of

$$\nabla_\alpha\nabla_\beta V_{ab}(x_a^0(i)-x_b^0(j);i,j)$$

on distance can no longer be neglected. We calculate $V_{ab}(x_a^0(j))$ up to first order in $u_\mu^0(i)$. Within this approximation the summations in Eqs. (2.9) and (2.10) can be performed explicitly to yield $\Omega^2(q,k)$ in terms of the Fourier components of the interaction potential. Below we present the diagonal elements of $\underline{\omega}^2(q)$, Eq. (2.14), to second order in V_{ab}, and the off-diagonal elements to first order:

$$(\underline{\omega}^2)_{aa}^{\alpha\beta}(q)=\omega_{0a}^2(q)+U^{\alpha\beta}(0)/m_aV_b$$
$$-\delta^{\alpha\beta}\Bigg\{\sum_{G_{b1}}[U^{\alpha\alpha\gamma}(G_{b1})\chi_{0a}^{\gamma\delta}(G_{b1})U^{\delta}(-G_{b1})+U^{\alpha\gamma}(G_{b1})\chi_{0a}^{\gamma\delta}(G_{b1}+q)U^{\delta\alpha}(-G_{b1})]/N_aV_b$$
$$+\sum_{G_{a1}}[U^{\alpha\alpha\gamma}(G_{a1})\chi_{0a}^{\gamma\delta}(G_{a1})U^{\delta}(-G_{a1})+U^{\alpha\gamma}(G_{a1}+q)\chi_{0b}^{\gamma\delta}(G_{a1}+q)$$
$$\times U^{\gamma\alpha}(-G_{a1}-q)]/N_bV_a\Bigg\}/m_aV_b , \tag{3.13}$$

$$(\underline{\omega}^2)_{ab}^{\alpha\beta}(q)=-U^{\alpha\beta}(-q)/m_bV_a . \tag{3.14}$$

$\delta^{\alpha\beta}$ denotes the Kronecker δ function and the summation over γ,δ is assumed in Eq. (3.13). The remaining matrix elements are obtained by interchanging a and b.

The matrix $(\underline{\omega}^2)_{0\mu}(q)$ on the rhs of Eq. (3.13) comprises the contributions from intralattice interaction $V_{\mu\mu}$. Diagonalization of this matrix yields the phonon spectrum of sublattice μ in the absence of V_{ab}. For $q\to 0$ the excitations correspond to sound waves, i.e., $(\underline{\omega}^2)_{0\mu}(q)$ is proportional to q^2 in this limit. The phonon modes are plane waves with wave vector q.

The remaining terms on the rhs of Eq. (3.13) arise from intersystem interaction V_{ab}. According to Eq. (2.14) two kinds of such contributions can be distinguished: those originating from $\Omega^2(q,q)$, the second, third, and fifth terms, and those from $\Sigma(q,\omega=0)$, the fourth and sixth terms. The contributions arising from $\Omega^2(q,q)$ account for the change in frequency of plane-wave phonons of lattice a due to the presence of the static potential generated by lattice b. While the second term describes this effect for the undistorted positions of the ions, the third and the fifth terms account for modulation of the equilibrium positions in the composite crystal. As $q\to 0$ these terms yield a finite contribution to $\omega_{aa}^2(0)$.

In contrast to the terms deriving from $\Omega^2(q,q)$, which arise in plane-wave phonons, the contributions due to $\Sigma(q,\omega=0)$ account for the fact that, for an incommensurate crystal, a single plane wave with wave vector q does not diagonalize the harmonic interaction potential. Instead plane waves with wave vectors differing from q by reciprocal-lattice vectors G_μ get mixed in. These admixtures, which result in distortions from the single plane-wave profile of phonon modes, change the excitation frequencies. Their contribution to $\omega^2(0)$ is finite. However, as we shall see below, there is cancellation between the terms arising from $\Omega^2(q,q)$ and $\Sigma(q,\omega=0)$.

The off-diagonal element $(\underline{\omega}^2)_{ab}(q)$, Eq. (3.14), is a consequence of the coupling between phonons on different sublattices. In the presence of long-range Coulomb interaction $(\underline{\omega}^2)_{ab}(q)$, as well as $(\underline{\omega}^2)_{\mu\mu}(q)$, shows a term which is proportional to $q_\alpha q_\beta/q^2$. Such terms are responsible for plasmonlike charge fluctuations in the spectrum of longitudinal excitations with a frequency gap at $q=0$ in both commensurate and incommensurate systems.[13,4] The transverse excitations, however, are similar to those

found in materials with short-range interactions only.[4] As we are mainly interested in the effects of incommensurability on the excitation spectra we consider in the following only the short-range part of the Coulomb interaction, i.e., assume the long-range part to be screened out by conduction electrons, as is the case, for instance, in $Hg_{3-\delta}AsF_6$. The results also apply to transverse excitations in ionic crystals.

We now discuss $\underline{\omega}^2(q)$, Eqs. (3.13) and (3.14), in the long-wavelength limit. We first consider $(\underline{\omega}^2)^{xx}_{\mu\nu}(q)$ a submatrix of $\underline{\omega}^2(q)$ which pertains to motion along the direction of incommensurability. As mentioned above $\omega_0^2(q)$ is proportional to q^2. Since there is no common reciprocal-lattice vector $G\neq 0$ with component along the direction of incommensurability, Eq. (3.9) for the interaction potential can be used to derive the q dependence of the remaining terms. From Eq. (3.13) it follows that, for $q=0$, the change in excitation energy of a sublattice phonon due to the potential generated by the other sublattice is exactly compensated for by an appropriate change in the form of the phonon mode. The only terms surviving this cancellation are terms of order q^2. Similarly, the off-diagonal element $(\omega^2)^{xx}_{ab}(q)$, Eq. (3.14), is proportional to q^2, i.e.,

$$(\underline{\omega}^2)^{xx}_{\mu\nu}(q)=\begin{bmatrix} c_a^2 & -c_{ab}^2 \\ -c_{ba}^2 & c_b^2 \end{bmatrix} q^2 , \tag{3.15}$$

with the relation $c_{ba}^2=c_{ab}^2 M_b/M_a$.

The form of $(\underline{\omega}^2)^{xx}(q)$ is in contrast to the form of the submatrices $(\underline{\omega}^2)^{\alpha\alpha}_{\mu\nu}(q)$, $\alpha=y,z$, which pertain to motion perpendicular to the direction of incommensurability. As there are common reciprocal-lattice vectors G with components in the y and z directions, $(\underline{\omega}^2)^{\alpha\alpha}_{\mu\nu}(q)$ does not vanish for $q=0$, i.e., to first order in V_{ab},

$$(\underline{\omega}^2)^{\alpha\alpha}_{\mu\nu}(0)=\begin{bmatrix} M_b & -M_a \\ -M_b & M_a \end{bmatrix} U^{\alpha\alpha}(0)/m_\alpha m_b V, \qquad \alpha=y,z . \tag{3.16}$$

As regards the coupling between displacements along different directions $(\underline{\omega}^2)^{x\alpha}_{\mu\nu}(q)$ with $\alpha=y,z$ turns out to be proportional to $q^x q^\alpha$ and thus vanishes for q in x direction. For high enough symmetry in the y,z plane and q in x direction, $(\underline{\omega}^2)^{yz}_{\mu\nu}(q)$ is zero too. Thus for wave vectors in the direction of incommensurability, motion along the principle axes is uncoupled and the 6×6 matrix $\underline{\omega}^2(q)$ decomposes into three 2×2 matrices, Eqs. (3.15) and (3.16). A similar analysis was given by Axe and Bak.[4]

The dynamics of the displacements can be derived from $\chi(q,\omega)$, Eq. (2.13). In the case of negligible self-energy $\Pi(q,\omega)$ weakly damped excitations occur at frequencies which are given by the square root of the eigenvalues of $\omega^2(q)$. Equation (3.16) for $q=0$ yields two zero-frequency modes corresponding to the rigid translation of the total system in the y,z plane. The two finite-frequency modes of Eq. (3.16) pertain to optic excitations in which the sublattices move out of phase in the y,z plane. The zero-frequency modes are part of two acoustic-phonon branches with excitation frequencies $\omega\sim q$, $q\rightarrow 0$. As the self-energy associated with a translation of the total system is of the order q^2 the damping of these acoustic excitations is negligibly small as was assumed above.

As regards motion along the direction of incommensurability Eq. (3.15) yields two eigenfrequencies with linear dispersion which are associated with the separate motion of each sublattice. Thus one has a total of four modes of the matrix $\underline{\omega}^2(q)$ with linear dispersion in this incommensurate system, instead of three acoustic modes in the commensurate case. For $q=0$ the two sublattice modes polarized in x direction become degenerate with zero excitation energy. They can be recombined to yield two other modes of zero energy: one mode corresponding to the rigid translation of the system as a whole, the other one related to an out-of-phase motion of both sublattices along the direction of incommensurability. The result that these displacement modes have zero energy is obvious for rigid displacements, and, for the out-of-phase mode, it is a consequence of the invariance of the free energy of incommensurate systems with respect to changes in the relative positions of the subsystems along the direction of incommensurability.[12]

However, for $q=0$, their self-energies differ substantially. This can be seen by noting that the self-energy $\Pi(q,\omega)$ is the Fourier transform of the space- and time-dependent correlation function of the forces acting on the modes under consideration.[10] Since rigid translations leave the Hamiltonian invariant there is no force acting on this mode and $\Pi(0,\omega)$ is zero. The out-of-phase mode, however, which shifts both subsystems relative to each other, does not leave the Hamiltonian invariant and thus generates a finite force acting on this type of motion. Although this force has no consequence for the free energy of incommensurate systems it has a nonzero correlation function and, therefore, a finite self-energy. Thus $\Pi(q,\omega)$ in Eq. (2.13) for $\chi(q,\omega)$ does not vanish in the long-wavelength limit although $\omega^2(q)$ does. Therefore, $\Pi(q,\omega)$ cannot be neglected in the calculation of $\chi(q,\omega)$ for motion along the direction of incommensurability.

We now study $\chi(q,\omega)$, Eq. (2.13), in the limit $q\rightarrow 0$ for displacements in the direction of incommensurability. We start by separating $\Pi(q,\omega)$ into

its real and imaginary parts, $\Pi=\Pi_1+i\Pi_2$. As $\Pi_1(q,\omega)$ is proportional to ω for $\omega\to 0$, $\omega\Pi_1(q,\omega)$ in Eq. (2.13) gives rise, in the limit $(q,\omega)\to 0$, to a mass renormalization:

$$m^*=\left[1-\lim_{\omega\to 0}\Pi_1(0,\omega)/\omega\right]m \ . \tag{3.17}$$

As the difference between the effective mass tensor m^* and m is not essential to the discussion that follows we neglect it henceforth, and obtain from Eq. (2.13) for $\chi(q,\omega)$ the expression

$$\underline{m}\underline{\chi}(q,\omega)\underline{N}^{-1}=-[\omega^2\underline{1}-\underline{\omega}^2(q)-i\omega\underline{\Pi}_2(q,\omega)]^{-1} \ , \tag{3.18}$$

which only contains $\underline{\Pi}_2(q,\omega)$. In the long-wavelength limit $\underline{\Pi}_2(q,\omega)$ can be further simplified by noting that, for $q=0$, the total linear momentum of the composite system is conserved,

$$\dot{p}_a(0)+\dot{p}_b(0)=0 \ . \tag{3.19}$$

As $\Pi_{\mu\nu}(q,\omega)$ is related to a bilinear correlation function of $\dot{p}_\mu(q)$ and $\dot{p}_\nu(q)$, it follows that, for a two-subsystem model, and given Cartesian components, all matrix elements $\Pi_{\mu\nu}(0,\omega)$ are proportional to each other and can be determined from a single quantity. This relation turns out to be

$$(\underline{\Pi}_2)^{xx}(0,0)=-\begin{pmatrix} M_b & -M_a \\ -M_b & M_a \end{pmatrix}\gamma/M \ , \tag{3.20}$$

with $M=M_a+M_b$. In Eq. (3.20) we have put ω to zero. γ denotes the relaxation rate of the relative linear momentum of the sublattices. An explicit expression for γ can be derived from the latter equation by taking advantage of Eq. (2.15) for $\underline{\Pi}(q,\omega)$, Eq. (2.21) for $\underline{\Sigma}_u(q,\omega)$, and Eq. (2.10) for $\underline{\Omega}^2(q,k)$, which we have calculated to first order in the interaction potential

$$\begin{aligned}\gamma=&-(\Pi_2)^{xx}_{aa}(0,0,0)M/M_b\\ &-\Bigg[\sum_{G_{b1}}[U^{x\gamma}(G_{b1})(\chi_{0a}(G_{b1})(\underline{\Pi}_2)^0_a(G_{b1},0)\chi_{0a}(G_{b1}))^{\gamma\delta}U^{\delta x}(-G_{b1})]/m_bN_a^2V_b\\ &+\sum_{G_{a1}}[U^{x\gamma}(G_{a1})(\chi_{0b}(G_{a1})(\Pi_2)^0_b(G_{a1},0)\chi_{0b}(G_{a1}))^{\gamma\delta}U^{\delta x}(-G_{a1})]/m_aN_b^2V_a\Bigg]M/V \ ,\end{aligned} \tag{3.21}$$

with the sums including summation over Cartesian components γ,δ.

The terms of the rhs of Eq. (3.21) express the fact that motion of one sublattice with respect to the other is damped by nonlinear forces between the particles. The first term describes this effect for a rigid relative translation. For an incommensurate system the harmonic interaction potential couples the $q=0$ plane waves of the subsystems to plane waves with wave vectors equal to a reciprocal-lattice vector of the subsystems. The damping of the latter modes gives rise to additional contributions to the damping of relative motion. They are summarized in the second and third terms on the rhs of Eq. (3.21).

Explicit expressions for $\Pi_2(q,q,0)$ in Eq. (3.21) can be derived from Eq. (2.11), which relates the self-energy to phonon Green's functions. For the sake of simplicity we replace these functions by the Green's functions in the absence of V_{ab} and, furthermore, neglect the damping of intermediate sublattice phonons and obtain

$$(\underline{\Pi}_2)^{xx}_{aa}(q,q,0)=-\pi k_BT\sum_{k_1,k_2}|\phi^{xj_1j_2}_{a\mu\nu}(q,k_1,k_2)|^2N_\mu N_\nu/m_\mu m_\nu M_a\omega_\mu^{-4}(k_1,j_1)\delta(\omega_\mu(k_1,j_1)-\omega_\nu(k_2,j_2)) \ , \tag{3.22}$$

assuming summation over sublattices μ,ν and phonon branches j_1,j_2. $\omega_\mu(k,j)$ denotes the frequency of a phonon with wave vector k in branch j of the phonon spectrum of sublattice μ. The third-order force constants with branch indices j which appear in Eq. (3.22) are related to those in Cartesian coordinates by appropriate linear combinations. Since the intrasubsystem forces vanish for $q=0$ only the interaction between the subsystems contributes to Eq. (3.22) in the long-wavelength limit. Thus the first-term on the rhs of expression (3.21) for γ comprises only contributions from V_{ab}, while only intrasubsystem forces contribute to the remaining self-energy terms appearing in Eq. (3.21).

It is important to realize that γ has a nonzero value due to interactions between harmonic phonons. Such interactions are always present in real systems even though for a strictly harmonic model γ

vanishes identically. However, as we shall see below, a nonvanishing γ strongly affects the spectrum of excitations at low frequencies and, therefore, must be taken into account for incommensurate systems. The third-order phonon process which contributes to γ according to Eq. (3.22) is a resonance process in which a phonon of one sublattice is converted into a phonon of the other sublattice which has the same energy and momentum. Such processes are allowed by momentum and energy conservation and will take place provided, of course, the phonons concerned are thermally occupied.

To get a qualitative description of the excitations we examined the poles of $\chi(q,\omega)$, Eq. (3.18), using the form of Eqs. (3.15) and (3.20) for the matrices $\omega^2(q)$ and $\Pi_2(q,\omega)$ as $(q,\omega)\to 0$. To simplify the notation we neglect in the following the off-diagonal matrix elements in $\omega^2(q)$. They change the quantitative results only slightly. For $q \gg \gamma/c_\mu$ we found the poles associated with the sublattice sound waves,

$$\omega_a(q)=\pm c_a q - i\gamma M_b/2M, \quad q \gg \gamma/c_\mu \tag{3.23}$$

and a corresponding form for $\omega_b(q)$. As far as the real part of the excitation frequencies are concerned they show the same q dependence as in the case of two noninteracting lattices. However, the damping of these modes, which in the noninteracting case is proportional to q^2, turns out, to lowest order in q, to be independent of wave vector. This is a consequence of the fact that, in the case of interacting sublattices, the linear momentum of each sublattice is no longer conserved. By lowering q, a crossover to a new dynamic regime occurs when the sublattice phonon frequencies become comparable to the phonon damping. In this regime we have only one sound-wave pole,

$$\omega_s(q)=\pm c_s q - \gamma_s q^2/2, \quad q\to 0 \tag{3.24}$$

with

$$c_s^2=(M_a c_a^2+M_b c_b^2)/M \tag{3.25}$$

and

$$\gamma_s=(c_a^2-c_s^2)(c_s^2-c_b^2)/\gamma c_s^2 . \tag{3.26}$$

This mode corresponds to a rigid translation of the entire crystal. c_s denotes the sound velocity of the composite system while the damping vanishes as q^2 for $q\to 0$. This kind of excitation is found in any crystal in the true long-wavelength limit. However, there is an excitation which is characteristic of an incommensurate crystal. It gives rise to a diffusion pole,

$$\omega_d(q)=-iDq^2, \quad q\to 0 \tag{3.27}$$

with

$$D=c_a^2 c_b^2/\gamma c_s^2 , \tag{3.28}$$

and is associated with a relative motion of the sublattices which leaves the center of mass of the total system fixed. There is an additional relaxational pole in the spectrum of $\chi(q,\omega)$,

$$\omega_\gamma(q)=-i\gamma, \quad q\to 0 \tag{3.29}$$

associated with the relative linear momentum of the subsystems. It stays finite as $q\to 0$ because the relative linear momentum is not conserved for interacting subsystems.

The origin of the diffusion pole, Eq. (3.27), can be traced back to the invariance of the free energy with respect to changes in the relative positions of the sublattices, and the fact the linear momentum associated with this motion is not conserved: The symmetry one is dealing with is a symmetry of the free energy only and not of the Hamiltonian. For $q \ll \gamma/c_s$ the relaxation rate Dq^2 of relative displacements is much smaller than the relaxation rate γ of the associated linear momentum. In this regime fluctuations in the relative positions of the sublattices obey a diffusion law. However, as q increases and Dq^2 gets the same order of magnitude as γ a crossover to a regime with oscillatory sublattice phonons occurs, Eq. (3.23). Such a behavior is not restricted to systems composed of incommensurate sublattices. A similar crossover from a diffusive to an oscillatory regime has recently been predicted to exist in the phason dynamics of incommensurably modulated crystals.[8]

In Fig. 1 we have plotted $S(q,\omega)$ vs ω for different values of q in the limit $(q,\omega)\to 0$. In this limit $S(q,\omega)$, Eq. (2.4), takes the form

$$S(q,\omega)=2k_B T(N_a N_b \gamma q^2/M m_a m_b)[(m_b-m_a)\omega^2+(m_a c_a^2-m_b c_b^2)q^2]^2 \times[(\omega^2-c_a^2 q^2)^2(\omega^2-c_b^2 q^2)^2+(\omega^2-c_s^2 q^2)^2(\omega\gamma)^2]^{-1} . \tag{3.30}$$

The plot shows the crossover from the high-frequency regime with two propagating sublattice sound waves to the low-frequency regime with only one propagating sound mode. In passing through the crossover the sublattice phonon, Eq. (3.23), with the higher frequency joins continuously to the sound wave of the composite crystal, Eq. (3.24). The maximum in $S(q,\omega)$ due to the sublattice phonon with the lower frequency disappears at a certain wave vector. For still smaller q a central peak due to the

diffusion pole [Eq. (3.27)], associated with the out-of-phase motion of the sublattices, appears. The relaxational pole, Eq. (3.29), which is related to the relative linear momentum, has only very little weight in $S(q,\omega)$.

So far we have studied the excitation spectra only in the limit $(q,\omega)\rightarrow 0$. Owing to the incommensurability of the crystal, however, the excitation spectra will not be periodic in q space as in the case of commensurate crystals. The low-frequency dynamics near reciprocal-lattice vectors of incommensurate crystals, for instance, will be different from the low-frequency dynamics near $q=0$. To elucidate this point further we have calculated $S(q,\omega)$ in the limit $(q,\omega)\rightarrow(G_{b1},0)$. In the absence of intersublattice forces the phonon spectrum of sublattice μ will be the same for q and $G_\mu+q$. Thus in the noninteracting case, there are branches of excitations, the acoustic phonons of sublattice b, whose frequencies vanish as $q\rightarrow G_{b1}$, while, due to the incommensurability along the direction of G_{b1}, the phonons of sublattice a attain finite frequencies for $q\rightarrow G_{b1}$. This has the consequence that, for q close to G_{b1} and $\omega\rightarrow 0$, the dominant contributions to $S(q,\omega)$ arise from density fluctuations in the b lattice only. In analogy to Eq. (2.4), $S(q,\omega)$ takes the form

$$S(q,\omega)\simeq 2k_BTq^2\mathrm{Im}\chi_{bb}^{xx}(k,\omega)/\omega\ , \tag{3.31}$$

with $k=q-G_{b1}$. This equation relates the low-frequency limit of $S(q,\omega)$ near $q=G_{b1}$ to the corresponding limit of $\chi_{bb}(k,\omega)$ near $k=0$. It remains to be true after switching on a small intersublattice potential V_{ab}. In the latter case $\chi_{bb}(k,\omega)$ can be easily deduced from Eq. (3.18) for $\chi(k,\omega)$, $(k,\omega)\rightarrow 0$:

$$S(q,\omega)\simeq 2k_BT(M_aM_b\gamma G_{b1}^2/Mm_b^2)(\omega^2c_a^2k^2)^2[(\omega^2-c_a^2k^2)^2(\omega^2-c_b^2k^2)^2+(\omega^2-c_s^2k^2)^2(\omega\gamma)^2]^{-1}\ . \tag{3.32}$$

In Fig. 2 we have plotted $S(q,\omega)$, Eq. (3.32), in the limit $(k,\omega)\rightarrow 0$ assuming that the sound velocity of lattice b is smaller than the sound velocity of lattice a, i.e., we have $c_b<c_s<c_a$. The peak seen in Fig. 2 for $k\gtrsim\gamma/c_s$ is due to the sound wave on lattice b. The excitations of lattice a all have frequencies well outside the frequency range of Fig. 2. In lowering k a crossover with a broad structure in $S(q,\omega)$ occurs near $k\simeq\gamma/c_s$. For still smaller k, $k\ll\gamma/c_s$, the

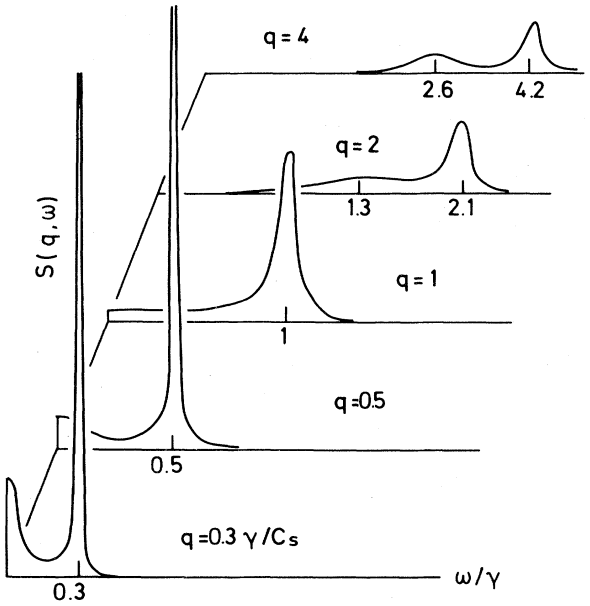

FIG. 1. Calculated dynamic structure factor $S(q,\omega)$ of a pure crystal composed of two incommensurate sublattices. For wave vectors $q\gg\gamma/c_s$ the two sublattice phonons are shown. The higher-frequency mode joins continuously to the sound wave (velocity c_s) of the composite crystal, $q\ll\gamma/c_s$. The central peak is due to the out-of-phase motion of the subsystems. The crossover is determined by the relaxation rate γ of the relative linear momentum of the subsystems.

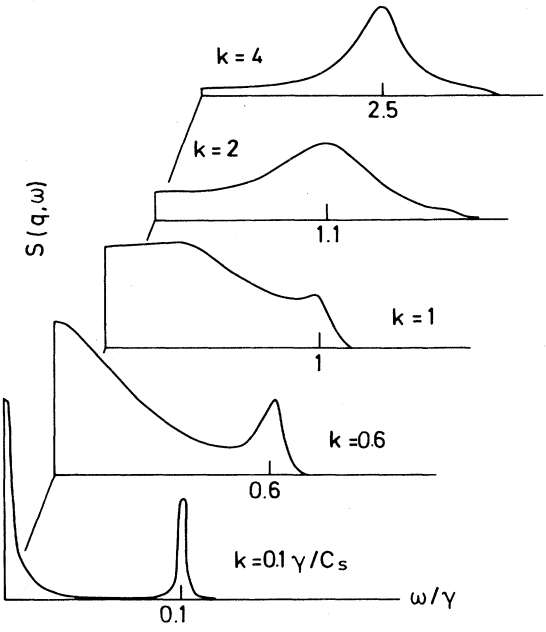

FIG. 2. $S(q,\omega)$ calculated for a pure incommensurate crystal in the limit $(q,\omega)\rightarrow(G_b,0)$, $k=q-G_b$, with G_b a reciprocal-lattice vector of the lattice with the lower sublattice sound velocity. This sublattice phonon, $k\gtrsim\gamma/c_s$, joins continuously to the central peak due to the out-of-phase motion of the sublattices, $k\ll\gamma/c_s$. The propagating mode, $k\ll\gamma/c_s$, corresponds to the sound wave (velocity c_s) of the composite crystal. γ denotes the relaxation rate of the relative momentum of the subsystems.

narrow peaks due to the diffusion mode and the sound wave of the composite crystal can be clearly distinguished at frequencies $\omega=0$ and $\omega=c_s k$, respectively. In passing through the crossover the sublattice sound wave joins continuously to the central peak associated with the out-of-phase motion of the subsystem.

We have also analyzed $S(q,\omega)$, Eq. (3.32), for $c_b > c_s > c_a$. Again we find a crossover near $k \simeq \gamma/c_s$ below which the sound wave of sublattice b evolves into the out-of-phase motion of the subsystems, with a central peak in $S(q,\omega)$ and the sound wave of the composite crystal. In this case, however, the crossover is accompanied by a narrowing of the linewidth of the sublattice sound wave which, in passing through the crossover, joins continuously to the sound wave of the composite system, contrary to the case of $c_b < c_s < c_a$ considered above (see Fig. 2).

IV. INCOMMENSURATE CRYSTAL WITH RANDOM-POTENTIAL FLUCTUATIONS

For a composite incommensurate crystal the free energy does not depend on the relative position $(d_a - d_b)^x$ of the sublattices along the direction of incommensurability. However, as we shall show below, this property is changed by the presence of random fluctuations in the interaction V_{ab} induced by impurities. The particles of the system adjust their equilibrium positions to random potential fluctuations in order to minimize the total potential energy including the impurity potential. According to Fukuyama and Lee[6] two limiting cases are of importance. In the strong-pinning case the system is able to respond to each individual impurity while in the weak-pinning case the system responds to the average potential of a large but finite number of impurities. The weak-pinning limit is best described by an "order parameter" $(d_a - d_b)^x$ which is no longer spatially constant throughout the sample as in the pure case but varies over distances of order ξ (Ref. 14), which are much larger than the mean separation of impurities.

To demonstrate the effect of random potential fluctuations on the dynamics of an incommensurate system we study a simple model in which impurities M_i are randomly distributed on sublattice b. We assume that the impurities have the same mass m_b and interaction potential V_b as all other particles of sublattice b, but their interaction potential with particles of sublattice a is $V_{ab}+\delta V$ instead of V_{ab}. Furthermore, we restrict ourselves to the weak-pinning limit. The total potential energy has the form

$$V_{\rm pot} = \sum_{\mu} \sum_{i,j} V_{\mu\mu}(x_\mu(i) - \chi_\mu(j);i,j)/2 + \sum_{i,j} [V_{ab}(x_a(i) - x_b(j);i,j) + \delta V(x_a(i) - x_b(j);i,j)\delta_{j,M_i}] \, . \tag{4.1}$$

Here the Kronecker δ function is equal to 1 when particle j is an impurity M_i and zero otherwise. The equilibrium positions of $V_{\rm pot}$ may be written as

$$x_\mu^0(i) = d_\mu(i) + R_\mu(i) \, , \tag{4.2}$$

where $d_\mu(i)$ comprises the periodic short-wavelength modulations considered in the preceding section arising from V_{ab}, as well as long-wavelength Fourier components due to δV. These long-wavelength fluctuations in $d_\mu(i)$ can be approximated as a continuous displacement field $d_\mu(x)$ with a characteristic rate of change ∇d_μ:

$$\nabla d_\mu \simeq \lambda \xi^{-1} \, , \tag{4.3}$$

where λ denotes a length of the order of a lattice constant. The contributions of $d_\mu(x)$ to $V_{\rm pot}$, Eq. (4.1), can be partitioned into two terms,

$$V_{\rm pot} = V_e + V_i \, . \tag{4.4}$$

V_e summarizes the contributions from the elastic medium defined by the first three terms on the rhs of Eq. (4.1),

$$V_e = c\xi^{-2}/2 \, , \tag{4.5}$$

with an elastic constant c which is related to the matrix elements of $\omega^2(q \to 0)$, Eq. (3.15). For distortions ∇d_μ, Eq. (4.3), along the x direction c takes the form

$$c = M c_s^2 \lambda^2 \, , \tag{4.6}$$

with c_s, Eq. (3.25), the sound-wave velocity of the composite crystal. V_i embodies the contribution to $V_{\rm pot}$ from δV,

$$V_i = \sum_{i,M_i} \delta V(x_a(i) - x_b(M_i)) \, . \tag{4.7}$$

Introducing Fourier components $\delta V(q)$, as in Eq. (3.4), V_i can be written as

$$V_i = \sum_{Q} \delta V(Q) \sum_{i,M_i} \exp\{iQ[x_a^0(i) - x_b^0(M_i)]\}/V \, , \tag{4.8}$$

with N_i the total number of impurities and $x_\mu^0(i)$ given by Eq. (4.2). To proceed further we introduce cells α of lateral dimension ξ such that $d_\mu(x)$ is approximately constant within each cell with value d_μ^α varying from cell to cell. We are thus able to per-

form the sum over the phase factors in Eq. (4.8) for each individual cell neglecting contributions from different cells, which is appropriate for a short-range potential δV:

$$V_i=\delta V(0)N_aN_i/V + \sum_{G_a}\delta V(G_a)\sum_{\alpha}\exp[iG_a(d_a^{\alpha}-d_b^{\alpha})]\rho_i^{\alpha}(G_a)/V_a \ , \tag{4.9}$$

with

$$\rho_i^{\alpha}(G_a)=\sum_{M_i}^{\alpha}\exp[iG_aX_{\alpha}-R_b(M_i)] \ , \tag{4.10}$$

and X_α denoting the position of cell α. The sum in the latter equation extends only over impurities of cell α. As the impurities are randomly distributed

$$\rho_i^{\alpha}(G_a)=(N_i^{\alpha})^{1/2}\exp[i\phi_{\alpha}(G_a)] \ , \tag{4.11}$$

where N_i^α denotes the number of impurities in cell α, and $\phi_\alpha(G_a)$ is an appropriate phase. Introducing $\phi(G_b)$ as the phase of $\delta V(G_b)$ we obtain for V_i,

$$V_i=\delta V(0)N_aN_i/V + \sum_{G_a}|\delta V(G_a)|\sum_{\alpha}(N_i^{\alpha})^{1/2}f^{\alpha}(G_a)/V_a \ , \tag{4.12}$$

with the phase factors included in $f^\alpha(G_a)$:

$$\bar{f}^{\alpha}(G_a)=\cos[\phi_i(G_a)+\phi_{\alpha}(G_a)+G_a(d_a^{\alpha}-d_b^{\alpha})] \ . \tag{4.13}$$

For the equilibrium configuration $d_a^\alpha-d_b^\alpha$ will be such that $f^\alpha(G_a)<0$ for all relevant G_a so that the contribution of the second term on the rhs of Eq. (4.12) to V_i is negative. Finally, we replace N_i and $f^\alpha(G_a)$ by their respective cell averages $\bar{N}_i^{\alpha},\bar{f}(G_a)$:

$$\bar{N}_i^{\alpha}=\xi^dN_i/V, \ \ \bar{f}(G_a)<0 \tag{4.14}$$

with $d=3$ the space dimension. As the number of cells is equal to V/ξ^d we arrive at the result

$$V_i=\delta V(0)N_aN_i/V + \xi^{-d/2}(N_i/V)^{1/2}N_a\sum_{G_a}|\delta V(G_a)|\bar{f}(G_a) \ . \tag{4.15}$$

From this equation and Eq. (4.5) it follows that, for $d<4$, $V_{\rm pot}$ [Eq. (4.4)] is negative for large ξ but positive for small ξ. Thus $V_{\rm pot}$ has its minimum value at a finite coherence length given by

$$\xi^{-2+d/2}=-dN_a(N_i/V)^{1/2} \times\sum_{G_a}|\delta V(G_a)|\bar{f}(G_a)/2c_s^2\lambda^2M \ . \tag{4.16}$$

The impurity potential $\delta V(x)$ affects the propagation of sound. Its contribution $\delta\Omega^2(q,k)$ to the matrix $\Omega^2(q,k)$, Eqs. (2.9) and (2.10), is given as follows:

$$(\delta\underline{\Omega}^2)_{aa}^{\alpha\beta}(q,k)=\sum_{i,M_i}\nabla_{\alpha}\nabla_{\beta}\delta V(x_a^0(i)-x_b^0(M_i))\exp[i(q-k)R_a(i)]/M_a \ , \tag{4.17}$$

$$(\delta\underline{\Omega}^2)_{ab}^{\alpha\beta}(q,k)=-\sum_{i,M_i}\nabla_{\alpha}\nabla_{\beta}\delta V(x_a^0(i)-x_b^0(M_i))\exp\{i[qR_a(i)-kR_b(M_i)]\}/M_b \ , \tag{4.18}$$

with the remaining matrix elements obtained by interchanging a and b. In the following we consider only motion in the direction of incommensurability, $\alpha=\beta=x$. $\delta\Omega_{\mu\nu}^{2xx}(q,q)$ gives rise to a contribution to $\omega^2(q)$, Eq. (2.14), which is first order in δV. Neglecting higher-order contributions due to $\Sigma(q,\omega=0)$, Eqs. (2.14) and (2.16), one gets for $\omega^2(q)$, $q\to0$,

$$(\underline{\omega}^2)_{\mu\nu}^{xx}(q)=\begin{bmatrix}M_b & -M_a\\ -M_b & M_0\end{bmatrix}\omega_p^2/M+\begin{bmatrix}c_a^2 & -c_{ab}^2\\ -c_{ba}^2 & c_b^2\end{bmatrix}q^2 \ , \tag{4.19}$$

with the pinning frequency ω_p,

$$\omega_p^2=\sum_{i,M_i}\nabla_x^2\delta V(x_a^0(i)-x_b^0(M_i))M/M_aM_b \ . \tag{4.20}$$

The last term on the rhs of Eq. (4.19) represents the contribution from all forces other than δV, Eq. (3.15). This term is, to a good approximation, unaffected by the long-wavelength fluctuations in the atomic positions induced by δV. The first term on the rhs of Eq. (4.19) is a consequence of δV. To get this form we neglect the q dependence in the off-diagonal element $\delta\Omega_{ab}^{2xx}(q,q)$, Eq. (4.18), which is a good approximation when $\delta V(x)$ is a short-range potential and q is much smaller than a reciprocal-lattice vector G_μ. This is equivalent to assuming that δV has only a negligible contribution to the sound-wave velocities. The pinning frequency ω_p can be estimated from Eq. (4.20) in a way which is completely analogous to the derivation of Eq. (4.15)

for V_i. One gets a finite value for ω_p,

$$\omega_p^2 = -\xi^{-d/2}(N_i/V)^{1/2} \times \sum_{G_a} |\delta V^{xx}(G_a)| \bar{f}(G_a) MN_a/M_aM_b \ , \tag{4.21}$$

with $|\delta V^{xx}(G_a)| = G_a^2 |\delta V(G_a)|$. Assuming for the sake of simplicity that only the smallest reciprocal-lattice vectors, $|G_a| = 2\pi/\lambda_a$, contribute to ω_p^2 the sum over G_a in the latter equation can be related to ξ using Eq. (4.16),

$$\omega_p^2 = 2(2\pi c_s/\xi)^2 (M\lambda/\lambda_a)^2 |dM_aM_b \simeq (2\pi c_s/\xi)^2 \ . \tag{4.22}$$

Thus ω_p is a direct measure of the coherence length ξ. Defining $\kappa = \omega_p/c_s$ we obtain the result

$$\kappa \simeq 2\pi\xi^{-1} \ . \tag{4.23}$$

The eigenvalues of $\omega_{\mu\nu}^{2xx}(q)$, Eq. (4.19), give two regimes with different q dependence (see Fig. 3). For $q \gg \kappa$ the eigenvalues correspond to the sublattice phonon frequencies,

$$\omega_\mu(q) = c_\mu q, \quad q \gg \kappa \tag{4.24}$$

whereas, for $q \ll \kappa$, one eigenmode is the sound wave of the composite crystal,

$$\omega_s(q) = c_s q, \quad q \ll \kappa \tag{4.25}$$

while the second mode, the optical mode, attains a finite frequency,

$$\omega_0(q) = \omega_p, \quad q \to 0 \ . \tag{4.26}$$

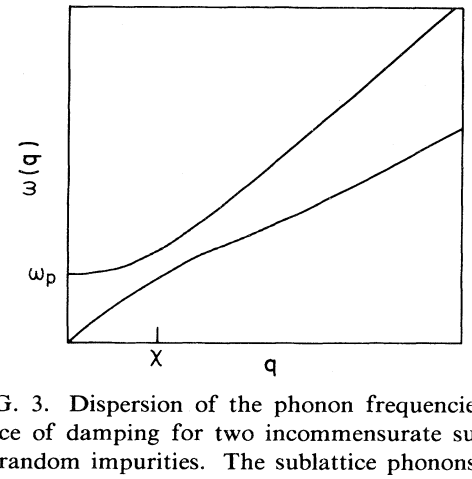

FIG. 3. Dispersion of the phonon frequencies in the absence of damping for two incommensurate sublattices with random impurities. The sublattice phonons, $q \gg \kappa$, join continuously to the sound wave of the composite crystal and the pinned out-of-phase motion, $q \ll \kappa$. ω_p denotes the pinning frequency and κ^{-1} the finite coherence length induced by the impurities.

This behavior of the eigenvalues of $\omega_{\mu\nu}^{2xx}(q)$ is qualitatively different from the case with $\kappa = 0$, which we have studied before. In the latter case only the two-sublattice phonon regime exists, i.e., Eq. (4.24) with $\kappa = 0$.

Random potential fluctuations not only contribute to $\omega^2(q)$ but also to the self-energy $\Pi(q,\omega)$, Eq. (2.15). In our analysis we considered explicitly the contribution of δV to $\Sigma_i(q,\omega)$, Eq. (2.22), which accounts for the scattering of plane-wave phonons by random potential fluctuations, but neglected unharmonic effects due to δV, which contribute to $\Pi(q,q,\omega)$. $\Omega^2(q,k)$ on the rhs of Eq. (2.22) is given by Eqs. (4.17) and (4.18) for $\delta\Omega^2(q,k)$:

$$(\delta\underline{\Omega}^2)_{\mu\nu}^{xx}(q,k) = \begin{bmatrix} M_b & -M_a \\ -M_b & M_a \end{bmatrix} h(q-k)/M \ , \quad q-k \neq 0 \tag{4.27}$$

with

$$h(q) = \sum_{i,M_i} \nabla_x^2 \delta V(x_a^0(q) - x_b^0(M_i)) \times \exp[iqR_a(i)] M/M_aM_b \ . \tag{4.28}$$

Equation (4.27) is correct provided $|q-k|$ is much smaller than a reciprocal-lattice vector. As characteristic fluctuations in the particles' positions occur only on a scale set by the coherence length ξ, $h(q)$ vanishes for $q \gg \xi^{-1}$, $h(q) \simeq 0$. To estimate $h(q)$ for $q \ll \xi^{-1}$ we take advantage of a sum rule which follows immediately from the definition of $h(q)$, Eq. (4.28),

$$\sum_q h(q)h(-q) = (M/M_aM_b)^2 N_a \times \sum_i \left[\sum_{M_i} \nabla_x^2 \delta V \times (x_a^0(i) - x_b^0(M_i)) \right]^2 . \tag{4.29}$$

The sum over impurities M_i on the rhs of this relation can be estimated from Eq. (4.20) for ω_p^2 assuming macroscopic homogeneity,

$$\sum_{M_i} \nabla_x^2 \delta V(x_a^0(i) - x_b^0(M_i)) \simeq \omega_p^2 M_aM_b/MN_a \ . \tag{4.30}$$

Substituting this relation into Eq. (4.29) one gets the simple result

$$\sum_q h(q)h(-q) \simeq \omega_p^4 \ . \tag{4.31}$$

Assuming a Gaussian form for $h(q)$,

$$h(q)=A\exp[-(q\xi)^2/2]\ , \tag{4.32}$$

Eq. (4.31) allows the determination of A,

$$A=\omega_p^2(2\pi\xi)^{d/2}V^{-1/2}\pi^{-d/4}\ . \tag{4.33}$$

With these approximations we get for $\underline{\Sigma}_i(q,\omega)$, Eq. (2.22),

$$\underline{\Sigma}_i(q,\omega)=-\sum_k \exp(-|q-k|^2\xi^2)\begin{bmatrix} M_b & -M_a \\ -M_b & M_a \end{bmatrix}\underline{m\chi}(k,\omega)\underline{N}^{-1}\begin{bmatrix} M_b & -M_a \\ -M_b & M_a \end{bmatrix}(A/M)^2\ , \tag{4.34}$$

which relates $\underline{\Sigma}_i(q,\omega)$ to $\underline{m\chi}(k,\omega)\underline{N}^{-1}$, Eq. (2.13),

$$\underline{m\chi}(k,\omega)\underline{N}^{-1}=-[\omega^2\underline{\mathbb{1}}-\underline{\omega}^2(k)-\omega\underline{\Pi}(k,\omega)]^{-1}\ . \tag{4.35}$$

$\underline{\omega}^2(k)$ is given by Eq. (4.19) while $\underline{\Pi}(k,\omega)$, Eqs. (2.15) and (2.19), separates into two terms,

$$\underline{\Pi}(k,\omega)=\underline{\Pi}_u(k,\omega)+\underline{\Pi}_i(k,\omega)\ , \tag{4.36}$$

of which $\Pi_u(k,\omega)$ comprises the contributions of forces other than δV, Eqs. (2.15) and (2.11), whereas $\Pi_i(k,\omega)$ summarizes the impurity contributions,

$$\Pi_i(k,\omega)=[\Sigma_i(k,\omega)-\Sigma_i(k,0)]/\omega\ . \tag{4.37}$$

Equations (4.34)—(4.37) allow a self-consistent calculation of $\chi(q,\omega)$. The result depends strongly on the ratio of Π_u to Π_i in Eq. (4.36). In the case in which the impurity damping dominates the intrinsic one, $\Pi_u \ll \Pi_i$, we have carried out a self-consistent calculation. Figure 4 shows $S(q,\omega)$ in the limit $(q,\omega)\to 0$. For $\omega \gg \omega_p$ the sublattice phonons, Eq. (4.24), can be clearly distinguished. With decreasing ω the linewidth of these phonons increases as well as the relative spectral weight of the sublattice phonon with the lower frequency. At the crossover $\omega \simeq \omega_p$, the self-energy $\Pi_i(q,\omega)$, which gives rise to the phonon damping, attains its maximum value, which turns out to be of order ω_p. Thus the condition $\Pi_u \ll \Pi_i$ is equivalent to $|\Pi_u(k,\omega)| \ll \omega_p$.

For $\omega \ll \omega_p$, $S(q,\omega)$ clearly shows the contribution due to weakly damped sound wave of the composite crystal, Eq. (4.25). It joins continuously to the lower-frequency sublattice phonon. This behavior differs from the pure case, $\omega_p=0$ (Fig. 1), in which the sound wave of the composite crystal joins continuously to the higher-frequency sublattice phonon. It is a consequence of the different behavior of $\omega^2(q\to 0)$ in the respective cases, Eqs. (3.15) and (4.19). For $\omega_p\neq 0$ the higher-frequency sublattice phonon joins continuously to the strongly damped optical mode, Eq. (4.26), which, however, has only little spectral weight in $S(q,\omega)$ for $q\to 0$.

We have also studied the opposite limit, $\Pi_u < \Pi_i$, in which the intrinsic damping is much larger than the impurity damping. This is equivalent to assuming $|\Pi_u(k,\omega)| > \omega_p$. In this case the self-consistent character of Eqs. (4.34)—(4.37), which comes into play through $\Pi_i(k,\omega)$, is negligible. As in the pure case we replace in the limit $(q,\omega)\to 0$, $\Pi(q,\omega)$ by $i\Pi_2(0,0)$, with the latter quantity given by Eq. (3.20). With these approximations we get for $\chi(q,\omega)$,

$$\underline{m\chi}(q,\omega)\underline{N}^{-1}=-[\omega^2\underline{\mathbb{1}}-\underline{\omega}^2(q)-i\omega\underline{\Pi}_2(q,\omega)]^{-1}\ , \tag{4.38}$$

which looks the same as the corresponding expression, Eq. (3.18), in the absence of impurities. However, in the present case $\omega^2(q)$ is given by Eq. (4.19) which, through $\omega_p\neq 0$, reflects the pinning by im-

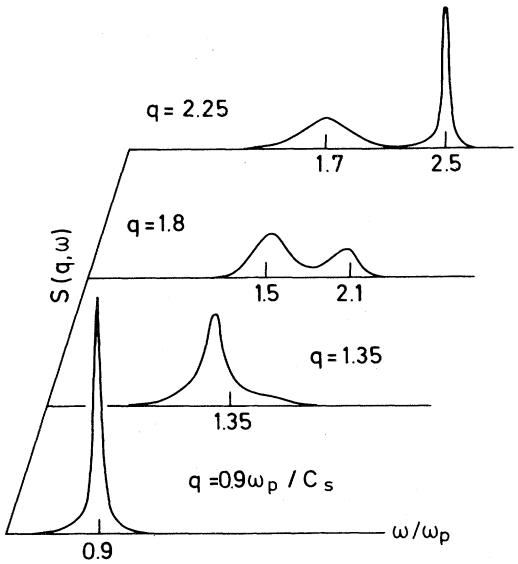

FIG. 4. $S(q,\omega)$ calculated for an incommensurate two-sublattice model with random impurities when impurity scattering dominates intrinsic mechanisms of momentum relaxation. The sublattice phonon, $q \gg \omega_p/c_s$, with the lower frequency joins continuously to the sound wave (velocity c_s) of the composite crystal, $q \ll \omega_p/c_s$. The pinned out-of-phase mode of the subsystems with pinning frequency ω_p has no spectral weight in $S(q,\omega)$ for $q\to 0$.

purities. For $q=0$, the poles of $\underline{m}\underline{\chi}(q,\omega)\underline{N}^{-1}$ are as follows:

$$\omega_s^2(0)=0\ , \tag{4.39}$$

$$\omega_d(0)=-i\omega_p^2/\gamma\ , \tag{4.40}$$

$$\omega_T(0)=-i(\gamma-\omega_p^2/\gamma)\ . \tag{4.41}$$

The corresponding poles in the pure case are given by Eqs. (3.24), (3.27), and (3.29). The zero-frequency mode pertains to the sound wave of the composite crystal. Impurity pinning of the sublattices leads to a relaxational pole, Eq. (4.40), for the relative motion of the sublattices which, in contrast to the pure case, Eq. (3.27), attains a finite value in the limit $q\to 0$. Thus for incommensurate crystals, the relative motion of the sublattices is strongly influenced by random interaction potential fluctuations. Equation (4.41) is related to the relaxation of relative linear momentum.

The pinning of relative sublattice motion, however, is only effective for $q<\omega_p/c_s$. This can be seen by studying $S(q,\omega)$. The same approximations which lead to Eq. (3.30) for $S(q,\omega)$ in the pure case now give the expression

$$S(q,\omega)=2k_BT(N_aN_b\gamma q^2/Mm_am_b)[(m_b-m_a)\omega^2+(m_ac_a^2-m_bc_b^2)q^2]^2$$
$$\times\{[(\omega^2-c_a^2q^2)(\omega^2-c_b^2q^2)-(\omega^2-c_s^2q^2)\omega_p^2]^2+(\omega^2-c_s^2q^2)^2(\omega\gamma)^2\}^{-1}\ , \tag{4.42}$$

which differs from Eq. (3.30) only by the term proportional to ω_p^2 in the denominator. For $q\gg\omega_p/c_s$ the consequences of ω_p on $S(q,\omega)$ are negligibly small. As for $\omega_p=0$ we observe the crossover at $q\simeq\gamma/c_s$ from the sublattice phonon regime to the regime with the sound wave of the composite crystal as the only propagating mode.

In Fig. 5 we have plotted $S(q,\omega)$ vs ω for $q\leq\gamma/c_s$. For intermediate values of q, $\omega_p\ll c_sq\ll\gamma$, the central peak and contributions from the sound wave of the composite crystal are shown. Lowering q the maximum of the central peak increases while its width decreases as in the pure case. However, due to $\omega_p\neq 0$, the central peak attains a finite maximum for $q\simeq\omega_p/c_s$ and, instead of increasing further, decreases for still smaller wave vectors. For $q\ll\omega_p/c_s$ the only contributions to $S(q,\omega)$ arise from the sound wave of the composite crystal.

A strong narrowing of the sound-wave linewidth occurs for $q\ll\omega_p/c_s$. In the intermediate regime, $\omega_p\ll c_sq\ll\gamma$, the damping of sound waves of the composite system takes the form γ_sq^2 with γ_s given by Eq. (3.26). The damping has the same order of magnitude as the relaxation rate Dq^2 of relative displacements, Eq. (3.28). In the intermediate dynamical regime, γ_s [Eq. (3.26)] is determined by γ, the self-energy at zero wave vector, Eq. (3.20). There are minor corrections to this form of sound-wave damping arising from the wave-vector dependence of the self-energies. But these contributions are negligible for $\omega_p\ll c_sq\ll\gamma$. For $q\simeq\omega_p/c_s$, however, the analysis of $\chi(q,\omega)$, Eq. (4.38), shows that, with the relative displacement mode attaining a finite relaxation rate, the contribution to sound-wave damping from the self-energy at zero wave vector decreases. Finally, for $q\to 0$, the q-dependent parts of the self-energy, which we have neglected so far, take over the sound-wave damping. For $q\ll\omega_p/c_s$ the damping takes the form γ_sq^2 with

$$\gamma_s=i\lim_{q\to 0}[M_a(\Pi_{aa}^{xx}(q,0)+\Pi_{ba}^{xx}(q,0))$$
$$+M_b(\Pi_{bb}^{xx}(q,0)+\Pi_{ab}^{xx}(q,0))]/Mq^2\ . \tag{4.43}$$

Note that the self-energy at zero wave vector, Eq. (3.20), which we considered so far, does not contribute to γ_s in Eq. (4.43). Thus to get a nonzero value for γ_s in the dynamical regime $q\ll\omega_p/c_s$ one has to keep track of the q^2 terms in $\Pi(q,\omega)$.

V. SOUND PROPAGATION IN A SOLID-LIQUID SYSTEM

The results obtained in the previous sections refer to systems composed of two incommensurate sublattices. However, there are substances in which one of the subsystems has a fluidlike character. $Hg_{3-\delta}AsF_6$ above 120 K is such a system. It consists of a body-centered-tetragonal lattice of AsF_6 ions and two nonintersecting sets of Hg chains running in the a and b directions, respectively. At room temperature the Hg chains act as one-dimensional (1D) fluids.

To elucidate sound-wave propagation in such systems we have studied in detail a model consisting of a 3D lattice b and a periodic array a of chains along the x direction. The a particles are assumed to behave as a fluid along the chains. There is a close analogy of this model and the system composed of two incommensurate sublattices in the absence of impurity pinning. In both cases the free energy is invariant with respect to arbitrary changes in the relative position of the subsystems along a direction defined either by the direction of incommensurabili-

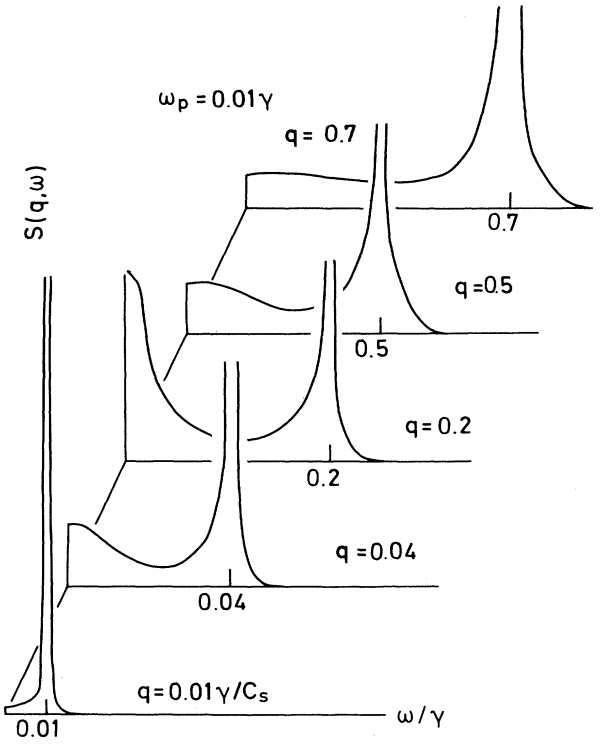

FIG. 5. $S(q,\omega)$ calculated for an incommensurate crystal with random impurities, and $q<\gamma/c_s$, when the relaxation rate γ of the relative linear momentum exceeds the pinning frequency ω_p. The propagating mode corresponds to the sound wave of the composite crystal (sound velocity c_s). The central peak due to the out-of-phase motion of the sublattices has maximal intensity for $q\simeq\omega_p/c_s$, and loses its spectral weight in $S(q,\omega)$, $q\to 0$, because of impurity pinning.

ty or the direction of the 1D fluids. Furthermore, for both systems this symmetry of the free energy is not a symmetry of the Hamiltonian. It is, therefore, not surprising that sound-wave propagation turns out to be very similar in both systems.

In the liquid-solid model under consideration the dynamical variables relevant for sound propagation are the particle density $\rho_a(q)$ of the liquidlike subsystem and the displacement field $u_b(q)$ of the 3D lattice. As we restrict ourselves to longitudinal sound waves propagating in chain direction, a convenient set of variables consists of $\rho_a(q)$ and $-iqu_b(q)$ with $q=q_x$. The latter dynamical variable is, to lowest order in the displacements, proportional to density fluctuations $\rho_b(q)$ in the 3D lattice.

The quantity of interest is the dynamical structure factor $S(q,\omega)$ of the composite system. To lowest order in the displacement field $S(q,\omega)$ takes the form[9]

$$S(q,\omega)=2k_BT\,\mathrm{Im}\sum_{\mu,\nu}\chi_{\mu\nu}(q,\omega)/\omega\ , \tag{5.1}$$

where $\chi(q,\omega)$ denotes the matrix of dynamic susceptibilities associated with the set of variables at hand. The corresponding expression for the case of two solid subsystems was given in Eq. (2.4). A convenient expression for $\chi(q,\omega)$ is[9]

$$\underline{m}\,\underline{\chi}(q,\omega)\underline{N}^{-1}=-q^2[\omega^2\underline{\mathbb{1}}-\underline{\omega}^2(q)-\omega\underline{\Pi}(q,\omega)]^{-1}\ , \tag{5.2}$$

with the matrices $\underline{m}$ and $\underline{N}^{-1}$ given in Eq. (2.6). This form for $\underline{\chi}(q,\omega)$ replaces Eq. (2.13). The matrix $\underline{\omega}^2(q)$ is related by Eq. (5.2) to the inverse static susceptibility $\underline{\chi}^{-1}(q)$,

$$\underline{\omega}^2(q)=q^2\underline{N}\underline{\chi}^{-1}(q)\underline{m}^{-1}\ . \tag{5.3}$$

As $\underline{\chi}^{-1}(q)$ is given by a second-order derivative of an appropriate free-energy functional, its asymptotic form in the long-wavelength limit can be deduced from the behavior of the free energy.[8] In the absence of intersubsystem interaction and long-range Coulomb forces, $\chi^{-1}(q=0)$ attains a finite value related to the subsystem compressibilities. As for the composite system the fluidlike character of one subsystem still guarantees invariance of the free energy with respect to changes in the relative position of the subsystems along the direction of the 1D fluids. This invariance of the free energy gives rise to a finite matrix $\chi^{-1}(q=0)$ even for the composite system. The derivation of this result is closely analogous to the derivation given by Zeyher and Finger[8] for incommensurably modulated crystals. Thus in the large-wavelength limit, the matrix $\omega^2(q)$, Eq. (5.3), is proportional to q^2 and takes the form of Eq. (3.15).

As regards the self-energy $\Pi(q,\omega)$ in Eq. (5.2) conservation of total linear momentum of the system, Eq. (3.19), ensures validity of Eq. (3.20) for the $(q,\omega)\to 0$ limit of $\Pi(q,\omega)$ with a nonzero value of γ due to the relaxation of relative linear momentum. Thus Eq. (5.2) for $\chi(q,\omega)$ reduces to the form appropriate for incommensurate sublattices, Eq. (3.18). As in the latter systems we expect two dynamic regimes for sound propagation in solid-liquid systems: a high-frequency regime with propagating subsystem sound waves, and a low-frequency regime with only one propagating sound mode associated with in-phase motion of the subsystems and a diffusive out-of-phase motion of the subsystems[5] (see Fig. 1). γ separates the frequency regimes.

In examining the processes which contribute to γ we restrict ourselves to contributions from particle motion along the direction of the chains. The analysis given below can be easily extended to include contributions from motion perpendicular to

the chains. The force acting on the liquid subsystem, $\dot{p}_a(0)=-\dot{p}_b(0)$, can be expressed as

$$\dot{p}_a^x(0)=\left[-\sum_{G_{b1}} U^x(G_{b1})\rho_a(-G_{b1}) + \sum_k U^{xx}(k)\rho_a(-k)u_b^x(k)+\cdots\right]\Big/V , \tag{5.4}$$

with G_{b1} a reciprocal-lattice vector of the solid subsystem along the chain direction, and $U(k)$ defined in Eq. (3.8). The first term on the rhs of this equation denotes the force on the liquid generated by the particles of the 3D lattice at their equilibrium positions, while the second term accounts for thermal fluctuations about the equilibrium positions. The dots in Eq. (5.4) indicate contributions of higher order in the displacement field.

In lowest-order perturbation theory each term on the rhs of Eq. (5.4) gives rise to a corresponding term in the self-energy associated with $p_a(0)$, i.e., we have $\gamma=\gamma_1+\gamma_2+\cdots$. γ_1 has its origins in the first term on the rhs of Eq. (5.4),

$$\gamma_1=\sum_{G_{b1}} |U^x(G_{b1})|^2 S^0{}_a(G_{b1},0)M/2M_aM_bV_b^2 , \tag{5.5}$$

with $S_a^0(q,\omega)$ the dynamic structure factor of the liquid in the absence of V_{ab}. This contribution is due to the fact that the static potential generated by the 3D lattice induces in the liquid a coupling between density fluctuations whose wave vectors differ by reciprocal-lattice vectors G_{b1} of the 3D lattice. The damping of density fluctuations thus is coupled to the $q=0$ mode and gives rise to γ_1. This follows immediately by relating $S_a^0(q,\omega)$ in Eq. (5.5) to the self-energy $\Pi_a^0(q,\omega)$ of density fluctuations,[9]

$$\gamma_1=-\sum_{G_{b1}} |U(G_{b1})|^2\Pi_{2a}^0(G_{b1},0) \times[\chi_{0a}(G_{b1})]^2M/m_bN_a^2N_bV_b^2 , \tag{5.6}$$

where $\chi_{0a}(q)$ is proportional to the static structure factor $S_a^0(q)$ of the liquid, $\chi_{0a}(q)=S_a^0(q)/k_BT$. This form of γ shows a close analogy to the second term on the rhs of Eq. (3.21) for γ in the case of incommensurate lattices. The origin of the latter term is the same as that of γ_1. The third term of Eq. (3.21), however, has no analogy in liquid-solid systems since, due to the lack of equilibrium positions along the chains, there are no reciprocal-lattice vectors G_{a1} of the liquid subsystem along the chain direction and the static potential set up by the liquid has only zero wave vector (modulo G_b).

The second term on the rhs of Eq. (5.4) gives rise to γ_2,

$$\gamma_2=k_BT\int d^3k\,|U^x(k)|^2 \times\int_{-\infty}^{\infty} d\omega\, G_a^0(k,\omega) \times G_b^0(-k,\omega)M/16\pi^4M_aM_bV , \tag{5.7}$$

with $G_\mu^0(q,\omega)$ the Green's functions associated with $\rho_a(q)$, $\mu=a$, and $-iqu_b(q)$, $\mu=b$, in the absence of V_{ab}. They are defined in terms of susceptibilities by Eq. (2.12). To progress further we neglect effects of damping on $G_\mu^0(k,\omega)$ and obtain

$$\gamma_2=k_BT\int d^3k\,|U^{xxx}(k)|^2\omega^{-4}(k) \times\delta(\omega_b(k)-\omega_a(k))M/(4\pi m_am_b)^2 . \tag{5.8}$$

Here $\omega_b(k)$ denotes a phonon frequency of the 3D lattice while $\omega_a(k)$ is the frequency of a density fluctuation in the liquid. Comparison of this result with Eq. (3.22) shows that γ_2 corresponds to the first term on the rhs of Eq. (3.21) for γ in the case of incommensurate sublattices.

The contributions to γ_2 arise from subsystem excitations which, for $V_{ab}=0$, have the same momentum and energy. Coupling of these degenerate modes by V_{ab} leads to exchange of energy and momentum and thus contributes to sound-wave damping in the subsystems. Though some reconstruction of the excitation spectrum takes place near points of degeneracies this does not change the argument above since the coupling between sound waves and reconstructed modes still exists. The application of the above results to $Hg_{3-\delta}AsF_6$ at $T>120$ K requires some modifications of the model because of the more complex structure of this substance which consists of two different sets of 1D liquids rather than one set as assumed in the model. However, if we restrict ourselves to longitudinal sound waves polarized in the direction of one set of Hg chains, the a chains for instance, the AsF_6 lattice and the Hg chains in b direction are commensurate as far as displacements in the a direction are concerned. Thus the out-of-phase motion of the latter systems attains a finite excitation frequency in the long-wavelength limit and only the in-phase motion corresponding to sound waves in the subsystem, composed of an AsF_6 lattice and b chains, are effective in coupling to sound waves along the a chains. The dynamic susceptibility of these two sound modes has the form of Eq. (5.2) with the matrices m and N^{-1} given in Eq. (2.6). m_a denotes the mass of a Hg ion while m_b is an effective mass associated with the in-phase

motion of the b chains and the AsH_6 lattice,

$$m_b^{-1}=(m_a^{-1}M_a+m_c^{-1}M_c)/M_b \ .$$

m_c is the mass of an AsF_6 ion and M_c denotes the total mass of the AsF_6 system, while M_a is the mass of the b chains, i.e., half the total Hg mass of $Hg_{3-\delta}AsF_6$, and $M_b=M_a+M_c$. N_μ in Eq. (2.6) is given by $M_\mu m_\mu^{-1}$.

With these modifications the results of the two-subsystem model studied above can be applied to $Hg_{3-\delta}AsF_6$ at $T>120$ K. The subsystem sound waves have been measured by inelastic neutron scattering.[2] At room temperature the respective sound velocities turn out to be $c_a=23.8$ meV Å and $c_b=14.0$ meV Å. For the long-wavelength limit the theory predicts the existence of sound waves of the composite crystal with a longitudinal sound velocity $c_s=18.3$ meV Å, Eq. (3.25), and a diffusion mode due to out-of-phase motion of the a chains and the system composed of b chains and an AsF_6 lattice.[5]

The sound waves of the composite crystal exist only below a frequency determined by the damping γ of motion along the a chains, Eqs. (5.5) and (5.8). Numerical estimates of γ, Eq. (5.5), for $Hg_{3-\delta}AsF_6$ show that umklapp process contributing to γ_1 are less efficient in coupling the subsystem motion than dynamic resonance processes which contribute to γ_2, Eq. (5.8). γ_1 turns out to be an order of magnitude smaller than γ_2.

In the dynamic resonance processes, which give rise to damping on the a chains, all modes of the b chains and AsF_6 lattice may take part, such as, for instance, in-phase motion of the b chains and an AsF_6 lattice along the direction of a chains, as well as density fluctuations on the b chains. The contributions of these modes to γ_2 is given by Eq. (5.8) with $U^{xxx}(k)$ replaced by an appropriate form. We made a numerical estimate of γ_2 using the experimental excitation spectra of $Hg_{3-\delta}AsF_6$,[2] and assuming the short-range interaction between Hg and AsF_6 ions to be Coulombic with charges $e(\mathrm{Hg})=+(3-\delta)^{-1}e$ and $e(AsF_6)=-e$, respectively. The estimate we obtained for γ_2 at $T=300$ K was of the order of 1 MHz.[5] This is a small frequency as far as neutron scattering experiments are concerned but well within the range of ultrasonic measurements. A more precise estimate of γ has to include the exact form of the screening of the Coulomb interaction in $Hg_{3-\delta}AsF_6$ by the conduction electrons. The precise form of the screening, however, is not known.

Contrary to the case of incommensurate lattices, random fluctuations in the interaction between a fluid and a lattice turn out to be ineffective in pinning the relative motion of the subsystems. The random potential is generally believed to be averaged out by thermal motion of the fluid. This requires a thermal coherence length ξ_T of the 1D fluids,[3] which is much smaller than the characteristic length ξ_i of changes in the fluid's density due to random potentials. At low temperatures, however, with increasing coherence in the particles' positions, ξ_T may exceed ξ_i. The system is then best described as a disordered solid and impurity pinning is anticipated.

VI. CONCLUSIONS

In this work we started from a microscopic Hamiltonian and derived the form of the coupling between the long-wavelength acoustic modes of a system with two incommensurate atomic subsystems. There are both intrinsic and extrinsic mechanisms which are important at very long wavelengths. These lead to only one propagating LA mode in this limit corresponding to the combined motion of the system. The characteristic frequency which governs the crossover to high-frequency behavior, with two uncoupled propagating modes in the subsystems, can be quite low, e.g., we estimate it as 1 MHz in $Hg_{3-\delta}AsF_6$ for the intrinsic coupling mechanisms. Our results are an example of the general phenomenon of the overdamping of the phase modes, or relative motion modes in incommensurate systems, in the long-wavelength limit discussed by Zeyher and Finger.[8]

ACKNOWLEDGMENTS

Stimulating discussions with R. Zeyher on the hydrodynamics of incommensurate systems are thankfully acknowledged by one of us (W.F.). The support of the Alexander von Humboldt Foundation and the hospitality of the Max-Planck-Institut für Festkörperforschung is gratefully acknowledged by T.M.R. during the beginning period of this work.

[1] J. P. Pouget, G. Shirane, J. M. Hastings, A. J. Heeger, N. D. Miro, and A. G. MacDiarmid, Phys. Rev. B 18, 3645 (1978).

[2] I. U. Heilmann, J. D. Axe, J. M. Hastings, and G. Shirane, A. J. Heeger, and A. G. MacDiarmid, Phys. Rev. B 20, 751 (1979).

[3] V. J. Emery and J. D. Axe, Phys. Rev. Lett. 40, 1507 (1978).

[4]J. D. Axe and P. Bak, Phys. Rev. B 26, 4963 (1982).
[5]W. Finger and T. M. Rice, Phys. Rev. Lett. 49, 468 (1982).
[6]H. Fukuyama and P. A. Lee, Phys. Rev. B 17, 535 (1978).
[7]H. Brand and P. Bak, Phys. Rev. A 27, 1062 (1983).
[8]R. Zeyher and W. Finger, Phys. Rev. Lett. 49, 1833 (1982).
[9]See, e.g., D. Forster, *Hydrodynamic Fluctuations, Broken Symmetry, and Correlation Functions* (Benjamin, Reading, Mass., 1975).
[10]W. Götze and K. H. Michel, in *Dynamical Properties of Solids,* edited by G. K. Horton and A. A. Maradudin (North-Holland, Amsterdam, 1974), Vol. 1, p. 499.
[11]See, e.g., S. Nakajima, Y. Toyozawa, and R. Abe, *The Physics of Elementary Excitations* (Springer, Berlin, 1980).
[12]S. C. Ying, Phys. Rev. B 3, 4160 (1971); G. Theodorou and T. M. Rice, *ibid.* 18, 2840 (1978).
[13]G. Theodorou, Solid State Commun. 33, 561 (1980).
[14]Y. Imry and S.-k. Ma, Phys. Rev. Lett. 35, 1399 (1975); L. J. Sham and B. R. Patton, Phys. Rev. B 13, 3151 (1976).