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Sound modes in composite incommensurate crystals

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Abstract. We propose a simple phenomenological model describing composite crystals, constructed from two parallel sets of periodic inter-penetrating chains. In the harmonic approximation and neglecting thermal fluctuations we find the eigenmodes of the system. It is shown that at high frequencies there are two longitudinal sound modes with standard attenuation, while in the low frequency region there is one propagating sound mode and an over-damped phase mode. The crossover between these two regions is analyzed numerically and the dynamical structure factor is calculated. It is shown that the qualitative features of the experimentally observed spectra can be consistently described by our model.

PACS. 61.44.-n Semi-periodic solids – 63.20.Dj Phonon states and bands, normal modes, and phonon dispersion

1 Introduction

Among the various types of incommensurate systems one of the simplest kind (the so-called uniaxial composite crystal) is constructed from two sets of regular inter-penetrating incommensurate chains. The recurrent interest for these systems is related mainly with the experimental activity on several classes of intergrowth compounds which can be viewed as good physical realizations of the uniaxial composite crystal. There is a considerable literature (mainly theoretical but not only) discussing eigenmodes and related properties of composite systems (see *e.g.* the review article [1] and references therein, and the more recent papers quoted in [2,3]).

Although a number of sophisticated calculations have been published over the last 20 years [4–9], there is still a clear need for a simple (but yet non-trivial) theoretical model with predictions which can be directly tested experimentally. In previous models all branches of excitations were simultaneously examined [9], and many subtle details of the composite systems were discussed in connection with higher order commensurability effects [4], such as the hierarchical nature of gaps arising at commensurate – incommensurate phase transitions [2]. The results of these investigations, even in the long wavelength limit, are expressed in terms of many unknown parameters, and lead to many different modes, while experimentally it is not clear whether it is possible at all to observe these modes and to determine these parameters. Furthermore, although the basic theory of excitations in incommensurate and composite systems was born long ago [10–12], many questions of principle remain unsettled, and there is an impressive quantity of unexplained, partially explained or contradictory results. In part this frustrating situation is just due to the lack of a simple and tractable analytical model.

Our motivation for adding one more paper to the topic is precisely to propose such a model. We do not intend to discuss in the present paper any of the dynamical details connected with the description of discontinuity edges in the vibrational density of states nor do we deal with the many structural properties (see *e.g.* [2]) related to commensurate – incommensurate phase transitions. Our aim is much more modest and based on the fact that many robust and experimentally testable features of the excitation spectra are not sensitive to the more delicate aspects of commensurability effects.

Note that one can consider composite structures as a new state of matter like liquid or solid states. Composite structures differ from modulated crystals insofar as one can not define an average periodic structure for the full system. Unlike crystals with incommensurate density waves that are characterized by weak deviations of atoms relative to their regular positions and unlike the adsorption structures with the rigid host and flexible guest sublattices, both subsystems in composites are more or less equivalent and therefore one can not treat them perturbatively as an external potential acting on its elastic counterpart, nor can one use the classical Frenkel-Kontorova model [10] to describe phase diagrams and excitations. Note that composite incommensurate crystals are also different from quasicrystals. According to the traditional classification [13] quasicrystals refer to systems where the rotational symmetry corresponds to a forbidden

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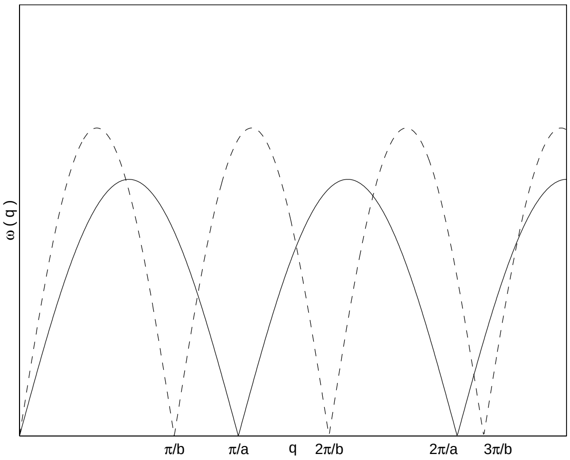

Fig. 1. Phonon spectra of two uncoupled incommensurate chains a and b.

crystal symmetry, which determines the unique ratio of incommensurate length scales that defines the structure, whereas for incommensurate crystals the rotational symmetry corresponds to an allowed crystal symmetry and many different sets of incommensurate length scales are possible.

Due to the existence of (at least) two length scales, there are twice as many hydrodynamic variables as in conventional crystals. However in quasicrystals these additional variables cannot be interpreted as real displacements of any of the sublattices, and this distinction leads to specific physical consequences.

The most interesting aspect of the physics of composites is the dynamics of their sublattices. In order to formulate the problem, let us consider first two inter-penetrating atomic chains with periods a and b and assume no coupling between them. Then, two independent sets of incommensurate (in q-space) phonon branches with dispersions $\omega_a(q) = c_a a^{-1} |\sin qa|$ and $\omega_b(q) = c_b b^{-1} |\sin qb|$ should be observed (Fig. 1). What happens with them, when the inter-chain interaction is properly taken into account? Different types of coupling are responsible for different effects.

The elastic coupling results in the renormalization of the sound velocities [8]. It leads to repulsion of the crossing branches with formation of intermediate gaps. The umklapp intersection of branches occurs however for *any* q since, because of the period incommensurability, the condition $\omega_a(Nq) \approx \omega_b(Mq)$ can always be satisfied when the integers N, M are chosen properly. Hence a rich hierarchical set of gaps in phonon spectra is expected (see *e.g.* [2]). Some general features related to breaking of analyticity due to static elastic couplings in the 1d double-chain model were reported recently in [3].

The electrostatic coupling takes place when the chains a and b are charged. It leads to nonlocal renormalization of the phonon self-energy, nontrivial retardation effects, and to the plasmon gap formation [6–8].

The dynamical dissipative coupling originates from the internal friction between the chains. To our knowledge the dissipative dynamics of composites was studied only by the authors in references [6,7] and [9]. In the papers [6,7] the authors have taken into account dynamic resonance processes arising due to the existence of degeneracy points of the subsystem modes which allow the interchange of energy and momentum. They described these processes by including into the response matrix a finite (in the limit $q \to 0$, and $\omega \to 0$) off-diagonal contribution. For a strictly harmonic lattice the contribution vanishes identically. We propose below a microscopic realization of the dissipative dynamical coupling which is always present in real systems even in harmonic approximation. As it concerns to the paper [9] the authors took into consideration all possible types of couplings and all degrees of freedom of composite systems, and therefore it is difficult to survey their results (and all the more to compare the results with experimental data) because of the large number of hydrodynamic variables involved. Meanwhile the broadening and renormalization of phonon spectra due to the inter-chain friction can easily hide the subtle hierarchical gap structure as well as other minute effects, like the fluctuation-driven destruction of the 1d composite long-range order [5] or the static and dynamic lock-in effects [2].

It is the purpose of the present publication to provide a simple and tractable model that incorporates the dissipative dynamic coupling in composite systems to calculate the experimentally testable features of the excitation spectra. We put forward a model of a composite system, which can be used to find the eigenmode spectrum (under relatively weak assumptions), and to compute the dynamical structure factor.

The outline of this article is as follows. In Section 2 we formulate a model description of a composite incommensurate crystal which includes all the basic dynamical equations. In Sections 3 and 4 the calculations of the eigenmodes and dynamical structure factors are presented. Section 5 is devoted to a discussion and summary of our main results. The Appendix deals with a very schematic description of a particular microscopic model of a uniaxial composite system.

2 Model

The simplest composite crystal consists of alternating chains with incommensurate periods a and b that are located in the $x-y$ plane and are directed along axes parallel to x at $y = md$ and at $y = (m + 1/2)d$. In this case even the basic structure is not commensurate. Because of the local differences in environment the a chain is modulated with period b, and the b chain – with period a. Therefore the atomic positions can be given by

$$\begin{aligned}\mathbf{R}^a(n,m;t) &= md\mathbf{e}_y + na\mathbf{e}_x + \mathbf{u}^a(n,m;t)\\ &\quad + \mathbf{e}_x g_1(na - \mathit{\Delta}),\,(n,m = 0, \pm 1, ...);\\ \mathbf{R}^b(n,m;t) &= (m+1/2)d\mathbf{e}_y + nb\mathbf{e}_x + \mathbf{u}^b(n,m;t) + \mathit{\Delta}\\ &\quad + \mathbf{e}_x g_2(mb + \mathit{\Delta}),\end{aligned} \tag{1}$$

where $\mathit{\Delta}$ is a certain natural offset of one set of chains with respect to its counterpart, $\mathbf{u}^{a,b}(n,m;t)$ are the displacements of the atoms about their average equilibrium positions, $g_1(x+b) \equiv g_1(x)$ and $g_2(x+a) \equiv g_2(x)$ are modulation functions.

In fact just the definitions (1) allow us to introduce all the degrees of freedom relevant for our model of a composite system (including therefore – the main branches of excitations). If we neglect for a moment the interchain interaction (purely hypothetically since in the absence of subsystem b subsystem a would be generally unstable), we get evidently two zero – energy modes for the two chains, each one implying a rigid displacement of one chain with respect to its fixed counterpart.

Interchain interactions introduce new ingredients in this scheme. In the incommensurate state the position of the centre of mass of the atoms relative to the modulation potential can be described by a phase variable. In a rigid chain approximation the acoustic phonon mode corresponds to a global rigid displacement of the two sets of chains, while the phase mode corresponds to relative rigid displacements of the two chains in opposite directions such that the centre of mass of the composite crystal remains fixed. In a commensurate state this phase variable is pinned, thus there is one acoustical mode and one optical mode with a gap whose magnitude is determined by the pinning potential and strongly decays with the order of commensurability. In the latter case our consideration will be still valid for characteristic frequencies higher than the gap.

Considering the relative displacements to be rigid means that we neglect the above mentioned intermodulation. The interchain interaction moves the atoms on both chains away from their regularly distributed positions. However if the modulation functions g_1 and g_2 are differentiable, then the atomic displacements corresponding to the phase mode are given by the following "dressed" displacements $\tilde{\mathbf{u}}$

$$\begin{aligned}\tilde{\mathbf{u}}^a(n,m;t) &\to \mathbf{u}^a(n,m;t) + \mathbf{e}_x a g_1',\\ \tilde{\mathbf{u}}^b(n,m;t) &\to \mathbf{u}^b(n,m;t) + \mathbf{e}_x b g_2',\end{aligned} \tag{2}$$

where g_i' denote a derivative over the argument.

In order to discuss the long-wavelength, low-frequency excitations one should transform the discrete variables introduced above into continuous ones. For sound modes which consist of equal shifts of $\mathbf{R}^a(n,m;t)$ and $\mathbf{R}^b(n,m;t)$ this transformation is evident. Namely in a continuum approximation the discrete variables $\mathbf{u}^{a,b}(n,m;t)$ are substituted by continuous functions $\mathbf{u}^{a,b}(\mathbf{r},t)$. However it is not the case for the phase mode which consists of inhomogeneous (on a microscopic scale) atomic displacements (corresponding to sliding of one chain system with respect to the other). However when the modulation functions g_i are analytic, one can still use the long-wavelength description but for dressed displacements $\tilde{\mathbf{u}}$.

For the benefit of the more skeptical reader the question concerning "dressed" variables should be clarified in more detail. In fact the dressed displacements $\tilde{\mathbf{u}}$ depend on the displacements $\mathbf{u}^a$ and $\mathbf{u}^b$ as initially defined according to (1) (since the modulation functions depend on the difference $u_x^a - u_x^b$). For weak enough interactions and far enough away from commensurability, the modulation functions $g_{1,2}$ are continuous analytic functions and the energy of the system remains unchanged when the phase $u_x^a - u_x^b$ is varied. However a uniform shift $u_x^a - u_x^b$ does not indicate a uniform displacement of the atoms in both chains, since the functions g_1 and g_2 are not constants. Thus strictly speaking we should write down equations of motion separately for dressed and undressed variables. In our phenomenological approach it means a certain renormalization of all phenomenological constants entering the equations. In what follows we neglect the renormalization and will use $\mathbf{u}^{a,b}(\mathbf{r},t)$ for both (acoustical and phase) modes. In spite of this erroneous assumption (which is equivalent to $g_{1,2} \simeq$ const.) the approximation correctly identifies the important modes and characteristic scales in the problem. This sin of omission can be easily corrected when detailed information concerning the values of the parameters becomes available.

The elastic energy can be written as a quadratic form of the strain tensor $\epsilon_{ij}^\alpha = \frac{1}{2}(\partial_i u_j^\alpha + \partial_j u_i^\alpha)$:

$$\begin{aligned}&\mathcal{F} = \frac{1}{2}\lambda_{ijkl}^{\alpha\beta}\epsilon_{ij}^\alpha \epsilon_{kl}^\beta + \frac{1}{2}\sigma\left(u_y^a - u_y^b\right)^2;\\ &\alpha,\beta = a,b; \quad i,j,k,l = x,y.\end{aligned} \tag{3}$$

The inter-chain interaction is given by the relative displacements along y (the 2nd term in (3)) and also by the cross terms in the elasticity tensor λ_{ijkl}^{ab}. At the same time the free energy expansion (3) is invariant with respect to the relative shift of the chains along x because of the incommensurability of the two periods, therefore there is no terms depending explicitly on $u_x^{a,b}$ (only on gradients of the displacements, *i.e.* on $\epsilon_{ij}^{a,b}$).

As a note of caution we should remark here that there is a fundamental difference between the both kind of soft degrees of freedom in (3), *i.e.* between acoustic and phase modes. The first one is a true Goldstone mode, because it breaks the continuous translational symmetry (and therefore remains hydrodynamic and gapless even if the modulation functions are not continuous). But the latter is a pseudo-Goldstone mode and it loses its hydrodynamic character in the case of discontinuous modulation functions (since phase variable breaks only countable translational symmetry).

To complete the derivation of the dynamical equations we have to know not only the elastic energy (3) but also the dissipative function R. The internal friction produced by the sliding displacement of the chains results in a dynamical inter-chain coupling proportional to the chain velocity difference $\dot{u}_x^a - \dot{u}_x^b$ that contributes to the dissipation function:

$$R = \frac{1}{2}\Gamma\left(\dot{u}_x^a - \dot{u}_x^b\right)^2 + \frac{1}{2}\eta_{ijkl}^{\alpha\beta}\dot{\epsilon}_{ij}^\alpha \dot{\epsilon}_{kl}^\beta. \tag{4}$$

The second term in (4) corresponds to the viscous dissipation, and the coefficients $\eta_{ijkl}^{\alpha\beta}$ entering (4) have the standard meaning of viscosity tensor elements [14]. One new phenomenological coefficient Γ describes a friction proportional to the velocity difference between the two chains. A few comments to assign physical meanings to the terms in

$$D^{xx}_{\alpha\beta}(q,\omega) = \begin{pmatrix} -\rho_\alpha\omega^2 + \lambda^{aa}_{xxxx}q^2 - \mathrm{i}\Gamma\omega - \mathrm{i}\eta^{aa}_{xxxx}q^2\omega & \lambda^{ab}_{xxxx}q^2 + \mathrm{i}\Gamma\omega - \mathrm{i}\eta^{ab}_{xxxx}q^2\omega \\ \lambda^{ab}_{xxxx}q^2 + \mathrm{i}\Gamma\omega - \mathrm{i}\eta^{ab}_{xxxx}q^2\omega & -\rho_b\omega^2 + \lambda^{bb}_{xxxx}q^2 - \mathrm{i}\Gamma\omega - \mathrm{i}\eta^{bb}_{xxxx}q^2\omega \end{pmatrix}. \tag{10}$$

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equation (4) may be helpful to the reader. The first term in (4) is the energy dissipation caused by relative motion between the two set of chains. In the framework of our purely phenomenological approach we are not in a position to determine a dominant microscopic mechanism for the dissipation. A natural estimate for the order of magnitude of the friction coefficient Γ (based on the assumption that the work produced by the drag force density is completely dissipated in a viscous media) is $\Gamma \propto \eta/d^2$, where η is the characteristic viscosity. It is worth noting that dynamic dissipative, static elastic and lock-in interchain interactions can differ both quantitatively, since there is no simple relation between the coupling coefficients Γ, σ, and the elastic moduli, and qualitatively, because unlike static interactions, the dynamic dissipative coupling does not necessarily produces intermodulations of the two subsystems.

Now we are in a position to write down the equations of motion

$$\begin{aligned}\rho_{\alpha\beta}\ddot{u}_i^\alpha &= -\frac{\partial \mathcal{F}}{\partial u_i^\alpha} - \frac{\partial R}{\partial \dot{u}_i^\alpha} \\ &= \lambda^{\alpha\beta}_{ijkl}\partial_j\epsilon^\beta_{kl} + \eta^{\alpha\beta}_{ijkl}\partial_j\dot{\epsilon}^\beta_{kl} + \Gamma\delta_{ix}\left(\delta^{\alpha a} - \delta^{\alpha b}\right)\left(\dot{u}_i^a - \dot{u}_i^b\right) \\ &\quad + \sigma\delta_{iy}\left(\delta^{\alpha a} - \delta^{\alpha b}\right)\left(u_i^a - u_i^b\right),\end{aligned} \tag{5}$$

where

$$\rho_{\alpha\beta} = \begin{pmatrix} \rho_a & 0 \\ 0 & \rho_b \end{pmatrix} \tag{6}$$

is the matrix of densities. Equation (5) is the second Newton law where the forces acting on the component a are: (i) the friction between the components due to their relative motion, and (ii) the elastic forces.

As it is well known [14] the propagation of elastic waves in anisotropic media is a rather complicated phenomenon. The directions of polarization and the eigen-frequencies are determined by the dispersion equation which is derived for the monochromatic solutions of (5). In a general anisotropic case however none of the polarization directions correspond either purely longitudinal or purely transverse directions with respect to the direction of the wave vector. We can look for solutions of the equations of motion (5) in the form

$$u_i^\alpha = u_{i0}^\alpha \mathrm{e}^{\mathrm{i}(\mathbf{qr} - \omega t)}. \tag{7}$$

The dispersion $\omega(\mathbf{q})$ can be found by substitution of (7) into (5) and, then, by diagonalization of the corresponding dynamical equations:

$$D^{ij}_{\alpha\beta}(q,\omega)u_{i0}^\alpha = 0. \tag{8}$$

The four degrees of freedom of u_i^α ($\alpha = a, b,\ i = x, y$) correspond to the four propagating eigenmodes.

We are interesting in the case when the excited wave is propagating along $\mathbf{x}$ (other geometries will be shortly discussed in the Conclusion). Then, the equation of motion (5) can be split into a transverse part for u_y^α and a longitudinal part for u_x^α. In the transverse case, the last term in equation (5) results in the decoupling of the excitations into sound and optical modes. Thus for the polarization perpendicular to the chains we will find the usual optical and acoustical branches. Less trivial is the longitudinal case when the last term in equation (5) vanishes and the dynamical mode coupling $\Gamma\delta_{ix}(\dot{u}_i^\alpha - \dot{u}_i^\beta)$ provides the most important contribution to the properties of the propagating excitations. In the next Section we consider this case in detail.

3 Longitudinal excitation spectrum

The dynamical equation for the longitudinal eigenmode with $\mathbf{q} \parallel \mathbf{x}$ looks like

$$D^{xx}_{\alpha\beta}(q,\omega)u_{x0}^\alpha = 0 \tag{9}$$

with

See equation (10) above.

The terms with Γ and λ^{ab} provide the dynamical dissipative and elastic coupling between two sound waves propagating in chains a and b. To pick out the effect of this coupling on the phonon dispersions we neglect the intrinsic phonon attenuation that is given by terms with η.

To find the eigenmodes one should solve the characteristic equation:

$$\begin{aligned}\det D^{xx}_{\alpha\beta}(q,\omega) = \rho_\alpha\rho_b\Big[&\left(\omega^2 - c_a^2q^2 + \mathrm{i}\gamma_a\omega\right)\left(\omega^2 - c_b^2q^2\right. \\ &\left.+\mathrm{i}\gamma_b\omega\right) - \left(c_i^2q^2 + \mathrm{i}\left(\gamma_a\gamma_b\right)^{1/2}\omega\right)^2\Big] = 0\end{aligned} \tag{11}$$

where:

$$\begin{gathered}c_a^2 = \lambda^{aa}_{xxxx}/\rho_\alpha, \quad c_b^2 = \lambda^{bb}_{xxxx}/\rho_b, \quad c_i^2 = \lambda^{ab}_{xxxx}/(\rho_\alpha\rho_b)^{1/2}, \\ \gamma_a = \Gamma/\rho_\alpha, \quad \gamma_b = \Gamma/\rho_b.\end{gathered} \tag{12}$$

At small q ($q < \gamma_{a,b}/c_{a,b}$) the dynamical dissipative coupling substantially renormalizes the dispersions of the two sound modes a and b.

The first two branches correspond to the time-conjugated sound-like modes

$$\omega(q) \approx \pm c_s q - \mathrm{i}\eta_s q^2, \tag{13}$$

with effective velocity and damping:

$$c_s^2 = \frac{\gamma_a c_b^2 + \gamma_b c_a^2 - 2\left(\gamma_a\gamma_b\right)^{1/2} c_i^2}{\gamma_a + \gamma_b},$$

$$\eta_s = \frac{1}{2\left(\gamma_a + \gamma_b\right)^2} \frac{\left[\left(\gamma_a - \gamma_b\right) c_i^2 - \left(\gamma_a\gamma_b\right)^{1/2}\left(c_a^2 - c_b^2\right)\right]^2}{\gamma_a c_b^2 + \gamma_b c_a^2 - 2\left(\gamma_a\gamma_b\right)^{1/2} c_i^2}. \tag{14}$$

The third mode corresponds to the over-damped phason-like mode with pure imaginary dispersion (diffusive mode):

$$\omega \approx -\mathrm{i}\eta_{ph} q^2, \tag{15}$$

where the damping factor is given by:

$$\eta_{ph} = -\frac{c_a^2 c_b^2 - c_i^2}{\gamma_a c_b^2 + \gamma_b c_a^2 - 2\left(\gamma_a\gamma_b\right)^{1/2} c_i^2}. \tag{16}$$

In addition there is a purely relaxational fourth mode:

$$\omega = -\mathrm{i}(\gamma_a + \gamma_b) \tag{17}$$

which, however, is hidden by the third mode that relaxes much more slowly.

At high q: $(q > \gamma_{a,b}/c_{a,b})$ the elastic coupling leads to the effective renormalization of the velocities of the sound modes a and b. The effect of the dynamical (dissipative) coupling is manifested by a specific q -independent sound attenuation. The dispersion of these two time-conjugated sound modes reads:

$$\omega = \pm c_{12} q - \mathrm{i}\gamma_{12}. \tag{18}$$

The renormalized velocities and attenuation factor are given by:

$$2c_{12}^2 = c_a^2(1 \pm \kappa) + c_b^2(1 \mp \kappa), \tag{19}$$

$$4\gamma_{12} = \left(\sqrt{\gamma_a\left(1 \pm \kappa^{-1}\right)} - \sqrt{\gamma_b\left(1 \mp \kappa^{-1}\right)}\right)^2. \tag{20}$$

where the elastic coupling parameter κ:

$$\kappa = \sqrt{1 + \frac{4c_i^4}{\left(c_a^2 - c_b^2\right)^2}} \tag{21}$$

is larger than one (weak coupling) and smaller than $(c_a^2 + c_b^2)/\left|c_a^2 - c_b^2\right|$ (stability condition).

At higher q the intrinsic phonon attenuation $\sim -\mathrm{i}(\eta_{xxxx}^{aa,bb}/\rho_{(a,b)})q^2$ is superimposed onto the dynamical attenuation $-\mathrm{i}\gamma_{12}$, a situation which can lead to a nontrivial q-dependence of the phonon line-width.

Of course it is possible to find the eigenmodes (at least no problem at all to find the dispersion laws numerically) in the general case of arbitrary q taking into account all phenomenological parameters entering (10). However aiming for a simple model we have chosen the simplest coupling form for the displacement components on both chains, following the principle of minimal requirements. To keep our model tractable (but yet non-trivial) and to avoid tedious calculations we drop the intra-mode coupling $\lambda_{xxxx}^{ab} q^2$ that does not change the qualitative picture. As will be discussed further, the intrinsic phonon attenuation expressed by $\mathrm{i}\eta_{xxxx}^{\alpha\beta} q^2\omega$ can be also neglected in the low energy region which is relevant to this discussion. Although actually this picture is not entirely correct it correctly identifies the important modes and characteristic scales in the problem.

As a note of caution we should also keep in mind that there are four types of interactions involved, namely:

- the elastic interaction proportional to gradients of the displacements, and related to off-diagonal terms of the elasticity tensor;
- locking interaction, proportional to sublattice displacement differences (the last term in (5) with the coefficient σ);
- two dissipative couplings described by the off-diagonal terms of the viscosity tensor and by the mutual friction coefficient Γ in (5).

The most robust phenomena are related to the dissipative couplings which we have taken into account in the derivation of the dispersion laws (13–18). However the locking and elastic couplings could lead to appreciable effects, like *e.g.* a gap at $q = 0$ for the sliding mode.

4 Dynamical structure factor: scattering experiments

The above discussed distinctive features of the excitation spectrum should be observable in different kinds of scattering experiments: ultrasonic measurements, Brillouin light scattering and inelastic coherent scattering of neutrons or X-rays. The double-differential cross-section gives the probability that say a neutron with initial energy E is scattered (coherently or incoherently) into a detector subtending a solid angle $\mathrm{d}\Omega$ around a certain direction with spherical coordinates 2θ and ϕ, with a final energy E'. We note $\hbar\omega = E - E'$ ($\hbar\omega$ is the energy transfer), and $\hbar\mathbf{Q} = \hbar\mathbf{k} - \hbar\mathbf{k}'$ is the momentum transfer, where $\hbar\mathbf{k}$ and $\hbar\mathbf{k}'$, are the incoming and outgoing neutron momenta. The scattered intensity is proportional to the differential cross section for inelastic scattering:

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}\omega} = \frac{k'}{k}\frac{1}{2\pi\hbar}\int \mathrm{d}t\mathrm{e}^{\mathrm{i}\omega(t-t')} \times \left\langle \sum_{n,n',m,m',\alpha,\beta} s_\alpha s_\beta \mathrm{e}^{-\mathrm{i}\mathbf{Q}\mathbf{R}^\alpha(n,m;t)} \mathrm{e}^{\mathrm{i}\mathbf{Q}\mathbf{R}^\beta(n',m';t')} \right\rangle, \tag{22}$$

where angular brackets denote a thermal average, $s_\alpha = s_a, s_b$ are the coherent atomic scattering factors *i.e.* differential cross sections of light, sound, neutrons or X-rays for single atoms of chains a and b, and the integrand in (22) is called the dynamical structure factor $S(\mathbf{Q},\omega)$ which is accessible by scattering techniques, and obeys the so-called detailed balance condition

$$S(\mathbf{Q},\omega)=\exp\left(\frac{\hbar\omega}{T}\right)S(-\mathbf{Q},-\omega)\,,$$

if the sample is in thermal equilibrium.

According to the fluctuation dissipation theorem [16]

$$S(\mathbf{Q},\omega)=\frac{1}{1-\exp(-\hbar\omega/T)}\frac{1}{\pi}\chi''(\mathbf{Q},\omega),$$

where χ'' is the imaginary part of the generalized susceptibility.

Calculations of χ'' is a formidable task that can be done only on the basis of an appropriate microscopic model (see *e.g.* the Appendix to our paper). In a general case the dynamic susceptibility χ is a matrix with respect to the displacement fields of the two chains and to the respective Cartesian components. Owing to the lack of discrete translational invariance in the x-direction it is non-diagonal also in the wave vectors. Since the full investigation of 2d (all the more 3d) problem requires an extraordinary amounts of computational work we must defer the investigation of the full problem to the time when experimental results suitable for a quantitative comparison become available (see also the Conclusion section of the paper), and present here only the results for the wave vector parallel to the incommensurate direction (x-axis).

In the case of longitudinal phonons it can be written in the following form (we dropped here unessential factors, since we are not interested in the absolute value of $S(\mathbf{Q},\omega)$ or $\chi''(\mathbf{Q},\omega)$):

$$S(Q,\omega)\propto \mathrm{Re}\sum_{n,n',\alpha,\beta}s_\alpha s_\beta \mathrm{e}^{\mathrm{i}Q(\alpha n-\beta n')}\times Q^2\frac{T}{\omega}\mathrm{Im}\chi^{xx}_{\alpha\beta}(n,n',\omega). \tag{23}$$

To study the structure factor we need to know the equilibrium positions of the atoms, the amplitude of oscillations, and the dispersion relation. Thus the generalized susceptibility $\chi^{xx}_{\alpha\beta}$, which is the response function, is proportional to the inverse dynamical matrix

$$\chi^{xx}_{\alpha\beta}(n,n',\omega)\sim D^{xx}_{\alpha\beta}(n,n',\omega)^{-1}. \tag{24}$$

We discuss here the cases when the momentum transfer $\mathbf{Q}$ (parallel to the x-axis) lies close to the Bragg peaks of the composite crystal $\mathbf{Q}=\mathbf{Q}^{m,n}_{a,b}+\mathbf{q}$ with

$$Q^{m,n}_{a,b}=\frac{2\pi}{a}m+\frac{2\pi}{b}n, \tag{25}$$

where m,n are integer numbers. Expression (25) implies that each observed Bragg peak can be classified as follows: main reflections of the a subsystem ($m\neq 0$, $n=0$); main reflections of the b subsystem ($m=0$, $n\neq 0$); common reflections to a and b subsystems ($m=0$, $n=0$); pure satellite reflections ($m\neq 0$, $n\neq 0$). With regard to the general program we do not have the ambition to find the dynamical structure factor in the whole region of parameters. In fact such a general formulation would not be tractable and in any case it would have little meaning in view of the qualitative nature of our model. For this reason and only for illustration purposes we consider a few particular cases.

Consider first the forward scattering when the momentum transfer $\mathbf{Q}=\mathbf{q}$ is small (acoustic case). This regime corresponds to Brillouin (light and X-ray) scattering. On the basis of (23) the structure factor $S(Q,\omega)$ is reduced to (aiming at a qualitative description of the dynamics we drop the amplitude factor $(k'/\pi k)\exp[-2W_{\mathbf{q}}]\left(1-\exp(\hbar\omega/T)\right)^{-1}$, where $W_{\mathbf{q}}$ is the Debye-Waller factor):

$$S(q,\omega)\propto\sum_{\alpha\beta}\left(q^2/\omega\right)s_\alpha s_\beta \mathrm{Im}D^{xx}_{\alpha\beta}(q,\omega)^{-1} \tag{26}$$

where $D^{xx}_{\alpha\beta}(q,\omega)$ is given by (10).

As mentioned earlier in order to pick out the effects of the dynamical dissipative coupling we neglect the elastic coupling λ^{ab} and assume that both channels a and b are equally-participating in the scattering process ($s_a=s_b=s$). Then $S(q,\omega)$ can be written in a simple form:

$$S(q,\omega)\propto \frac{s^2(\gamma_a+\gamma_b)(\rho_a-\rho_b)^2q^2[\omega^2-c_-^2q^2]^2}{(\omega^2-c_a^2q^2)^2(\omega^2-c_b^2q^2)^2+(\gamma_a+\gamma_b)\omega^2(\omega^2-c_+^2q^2)^2}, \tag{27}$$

with:

$$c_\pm^2=\frac{\left(\rho_a c_a^2\pm\rho_b c_b^2\right)}{\rho_a\pm\rho_b}\,. \tag{28}$$

The structure factor in the so-called pseudo acoustic regime (when $\mathbf{Q}$ is close to a nonzero Bragg peak) is different from the acoustic structure factor (27). It is worth noting also that this feature is different from the case of phonons in conventional crystals.

Consider *e.g.* the pseudo acoustic region close to the Bragg peak $Q_{1a}=\pi/a$ (Fig. 1). Because of the large energy separation, the modes a and b are decoupled and the structure factor for mode a is written as:

$$S_a(\pi/a+q,\omega)\propto(Q_{1a}/\omega)s_a^2D^{xx}_{aa}(\pi/a+q,\omega)^{-1}. \tag{29}$$

The matrix element $D^{xx}_{aa}(\pi/a+q,\omega)^{-1}$ coincides with $D^{xx}_{aa}(q,\omega)^{-1}$ (it corresponds to the umklapp coupling processes). On one side the "a" phonon with $Q=\pi/a+q$ is decoupled from the "b" phonon with $Q=\pi/a+q$ because of the large energy separation. At the same time the "a" phonon with $Q=\pi/a+q$ is equivalent to the "a" phonon with q, that in turn, is strongly coupled with the "b" phonon with $Q=q$.

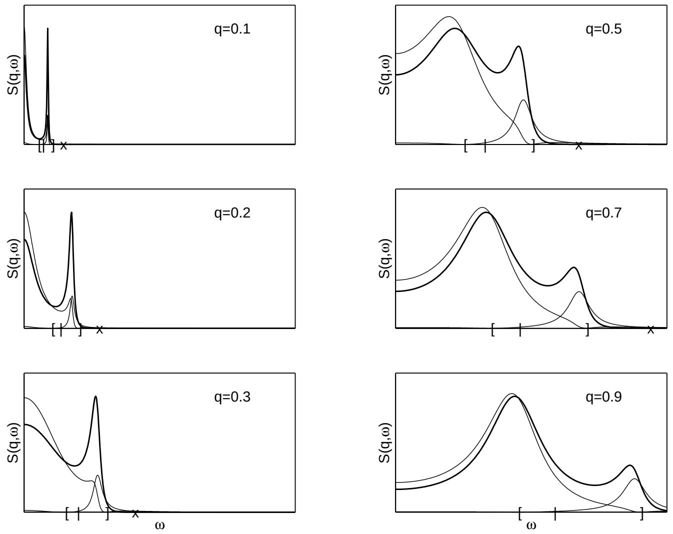

Fig. 2. The structure factor of a uniaxial composite system. Bold lines correspond to $S(q,\omega)$, normal lines correspond to $S(Q_a^m+q,\omega)$ and $S(Q_b^m+q,\omega)$. Ticks: "[" and "]" correspond to c_aq and c_bq; ticks "|" and "x" correspond to c_+q and c_-q. Parameters: $\rho_a=1$, $\rho_b=2$, $c_a=1$, $c_b=2$, $\gamma_a+\gamma_b=0.45$.

A straightforward calculation gives

$$S_a\left(\frac{\pi}{a}+q,\omega\right)\propto \frac{Q_{1a}^2(\gamma_a+\gamma_b)\rho_a^2\left(\omega^2-c_a^2q^2\right)^2}{\left(\omega^2-c_a^2q^2\right)^2\left(\omega^2-c_b^2q^2\right)^2+\left(\gamma_a+\gamma_b\right)\omega^2\left(\omega^2-c_+^2q^2\right)^2}. \tag{30}$$

The results of the calculations of $S(\mathbf{q},\omega)$ for different values of ω scanning the structure factor at a fixed value of q are shown in Figure 2. The picture presented above leads to the following qualitative predictions:

- The maximum in $S(\mathbf{q},\omega)$ due to the sublattice phonon with the lower frequency disappears at a certain wavevector.
- Both $S(q,\omega)$ and $S(n\pi/a+q,\omega)$, $S(m\pi/b+q,\omega)$ have asymmetric profiles with "antiresonance" points where they vanish (as in the case of the Fano resonance, when the interference of a discrete state with a continuum gives rise to characteristically asymmetric peaks in excitation spectra);
- Although $S(n\pi/a+q,\omega)$, $S(m\pi/b+q,\omega)$ correspond to "decoupled" phonons, they show traces of mixing between the two phonon branches because of the umklapp process;
- At low q we always have a double peak structure with a central peak and a sound-like side peak.

5 Conclusion

Let us sum up the results of our paper. We propose a simple phenomenological model describing a composite crystal, constructed from two sets of periodic inter-penetrating chains having a common direction (either commensurate or incommensurate). We intentionally restrict ourselves to the simplest case taking into account mainly the dissipative coupling of the two subsystems, since theoretical modelling of composite crystals is often hampered by lack of detailed knowledge concerning the parameters entering more complete and more sophisticated models than the present one.

As incommensurate composites do not possess translational symmetry, one cannot define a Brillouin zone. In zero order approximation if both sublattices are independent, there are two phonon modes (one for each sublattice) but only one sound velocity (in the long-wavelength limit). Coupling between sublattices leads (again in the hydrodynamic, *i.e.* $q\to 0$ limit) to one acoustic mode for the composite, and one sliding mode, as found above. In this limit the acoustic mode is associated with a global translation of the composite, while the sliding mode describes the relative "dressed" translation of the two sublattices.

We find the eigenmodes of the system and calculate the dynamical structure factor $S(\mathbf{Q},\omega)$. We show that at high frequencies there are two longitudinal sound modes with wave vector independent attenuation, while in the low frequency region there is one propagating sound mode and two over-damped phase modes. The structure factor has a specific profile resembling the Fano resonance. We anticipate that these effects will be observable, and that understanding the physical mechanism will be essential to predict the behaviour of the composite materials.

However a number of remarks related to our results are in order. First, the separation of high and low frequency regions depends on the natural scale $q_c\propto\gamma/c$ (see Eqs. (13–18)), and the large q or ω limit corresponds to the uncoupled case, when both eigenvectors are those of acoustic modes propagating more or less independently, while in the opposite limit the chains are tightly coupled. Of course in real materials the critical wave vector q_c could be so large that the coupled limit is adequate over the whole accessible range of wave vectors.

Second, in fact we considered a composite chain system embedded into two dimensional space. Thus one should distinguish between modes propagating along the x and y directions. For the modes propagating along x we found in the low q limit two diffusive and one pair of degenerate sound like modes. In addition there are transverse modes which are: two time-conjugated optical ones (*i.e.* with a gap) and two time-conjugated acoustic modes (sound like). For the propagation along y the situation is inverse: transverse modes give two diffusive modes and two time-conjugated sound like modes, while longitudinal modes are two time-conjugated optical modes and two time-conjugated acoustic modes.

Note also that it is to be expected that the forces coupling the two chains are weak for relative displacements along the chain direction but of normal strength for displacements perpendicular to the chains. It implies that even the smallest wave vectors parallel to the x-axis probed *e.g.* by neutrons (see below our short survey of experimental data) may fall within the large q uncoupled regime, whereas all wave vectors in the orthogonal directions are in the normal coupled regime.

Third, we assumed that atomic motions are not related to charge fluctuations (therefore we neglected Coulomb forces). As a result of Coulomb interactions additional $q = 0$ gaps (plasmon gaps) will open, and in addition long-range Coulomb interactions will lead to a renormalization of the short-range elastic constants. The detailed discussion of all these problems is beyond the scope of our paper.

It is worth to compare the predictions of the present model, with results of [6,7] where the authors also stressed the relevance of dynamical couplings. Qualitatively (especially in the low frequency region) our model leads to the same three longitudinal modes as predicted in [6,7]. However we presented a much simpler derivation which offers a deeper insight into the physical structure of the problem. Besides we should stress that we determined as well the dispersion laws for the transverse modes and for the high-frequency region we found an unusual q-independent contribution to the sound modes attenuation (due to our dynamical coupling) superimposed on the conventional ($\propto q^2$) line-width. Note also that for our model there are no universal inequalities between sound mode velocities c_a, c_b, c_s as reported in [6,7].

In spite of the current research activity in the field of composite crystals, there are still few experimental results to which model predictions can be compared. Furthermore, the generic aspects of composite crystals which this and other models aim to describe, are often masked in practice by other effects, specific to each particular compound under study. One of the first composite systems studied with respect to dynamics, is the $\mathrm{Hg}_{3-\delta}\mathrm{AsF}_6$ "mercury-chain" compound, which turned out to be a rather complex and unique system with two orthogonal sets of Hg-chains, showing 1d liquid-like behaviour at high temperature ($T > T_c = 120$ K) [17]. Below T_c, the long-range phase-correlation between parallel Hg-chains is established through indirect interchain interactions, mediated by the orthogonal set of Hg-chains (which order simultaneously). Hence the compound is not a good 1d-composite model system.

The alkane-urea compounds [18] appear a priori as better model systems because of their strongly uniaxial hexagonal structure. However, the structural transition at $T_c \simeq 130$ K, marking the onset of ferroelastic domains, precludes a detailed inelastic neutron or Brillouin study at low temperatures, where mode-broadening due to molecular disorder and anharmonic effects could be suppressed. The report by Schmicker *et al.* [19] of Brillouin scattering evidence for a propagating phase mode, has clearly been refuted by subsequent measurements by Ollivier *et al.* [20]. Instead, these latter authors report observing essentially normal acoustic dispersions and a quasielastic line polarized in the chain direction, which they ascribe to an overdamped, pinned phase mode. In the limit when the mode damping coefficient f exceeds the pinning frequency ω_g ($\omega_g \ll f$), the phase-mode response would become quasielastic with a width $\gamma_{Q.E.} = \omega_g^2/f$. It is indeed quite plausible that, in the q-range covered by light scattering, neither ω_g nor f would show much q-dependence, in which case one would not expect much q-dependence for the quasielastic linewidth $\gamma_{Q.E}$, in agreement with the results in reference [20]. It conforms as well with our theoretical predictions presented in Section 3. The fact that $\gamma_{Q.E}$ is also found to be T-independent between 130 K and 300 K, is much more puzzling since, over such a wide T-range, one would expect ω_g to soften and Γ to increase with T, leading to a significantly narrower quasielastic line at room-T, in clear contradiction with the measurements. Hence, the assignment of the observed quasielastic line as a collective excitation must be taken as tentative only. On the other hand inelastic neutron scattering data on the $n = 19$ alkane-urea compound [21], show only one longitudinal acoustic (LA) mode in the chain direction (instead of two as would be expected in the high-q limit), with an anomalous q-independent linewidth. The non-observation of the collective modes associated with the alkane sublattice can be ascribed to the strong orientational and translational disorder of the alkane molecules [22,23] above T_c.

Another interesting system is the aperiodic layered crystal $\mathrm{Bi_2Sr_2CaCu_2O_{8+\delta}}$, which was recently suggested to belong to the class of composite (rather than modulated) incommensurate systems [24,25]. The mode structure observed by Etrillard *et al.* [25] is consistent with the presence of two independent LA branches along the incommensurate direction, as expected in the q-range of the neutron experiment. The relative strength of the two branches is however not understood, nor is the absence of a similar sublattice decoupling for the TA branches polarised in the incommensurate direction.

It is obvious from the above remarks that the experimental situation still remains very open. To a large extent, this is due to the difficulty in identifying suitable model systems and in growing single-crystal specimens of appropriate size and quality for light and neutron scattering experiments. We hope that this work stimulates further efforts in this field.

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Appendix

The equilibrium positions of the atoms in the incommensurate direction (x) could be written in a more abridged form than (1), namely

$$x_n^a(t) = na + u_x^a + g_1\left(na + u_x^a - u_x^b\right);$$

$$x_m^b(t) = mb + u_x^b + g_2\left(mb + u_x^b - u_x^a\right),$$

with $g_1(x+b) \equiv g_1(x)$ and $g_2(x+a) \equiv g_2(x)$. Both functions g_1 and g_2 can be found from a static microscopic model.

The energy of interaction within the chains is written as usually, the dynamical coupling in the dissipation function can be written as:

$$R = \frac{1}{2}\Gamma \sum_{mn} \left(\dot{u}_n^a - \dot{u}_m^b\right)^2 f(ma - nb),$$

where $f(x)$ can be modelled as:

$$f(x) = \exp\left[-\frac{x^2}{\lambda^2}\right].$$

Suppose we have a relatively long-range coupling $\lambda \gg a, b$. In the case similar to that considered in the main text of the paper the equations of motion (after Fourier transformation over t) read

$$\begin{aligned}-\omega^2 \rho_a u_n^a &= \lambda_a (u_{n+1}^a + u_{n-1}^a - 2u_n^a)\\ &\quad + \mathrm{i}\Gamma \sum_m f(ma - nb) u_m^b;\end{aligned}$$

$$\begin{aligned}-\omega^2 \rho_b u_n^b &= \lambda_b \left(u_{n+1}^b + u_{n-1}^b - 2u_n^b\right)\\ &\quad + \mathrm{i}\Gamma \sum_m f(ma - nb) u_m^a,\end{aligned}$$

or, after space Fourier transformation in ma, nb: $u_n^a = u_{n0}^a \exp \mathrm{i}(\omega t - qna)$, $u_n^a = u_{n0}^b \exp \mathrm{i}(\omega t - qnb)$

$$\begin{aligned}-\omega^2 \rho_a u_{n0}^a &= -\lambda_a \sin^2 \frac{qa}{2} u_{n0}^a\\ &\quad + \mathrm{i}\Gamma \sum_m \mathrm{e}^{-(ma-nb)^2/\lambda^2} \mathrm{e}^{\mathrm{i}q(ma-nb)} u_{m0}^b;\end{aligned}$$

$$\begin{aligned}-\omega^2 \rho_b u_{n0}^b &= -\lambda_a \sin^2 \frac{qb}{2} u_{n0}^b\\ &\quad + \mathrm{i}\Gamma \sum_m \mathrm{e}^{-(ma-nb)^2/\lambda^2} \mathrm{e}^{\mathrm{i}q(ma-nb)} u_{m0}^a.\end{aligned}$$

These equations give $\chi_{\alpha\beta}^{xx}(n, m, \omega)$ which we need for the analysis of the neutron scattering.

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