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Phason Dynamics of Incommensurate Crystals

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It is shown that phase modulations (phasons) of the order parameters of incommensurate lattices are always diffusive at sufficiently large wavelengths λ. In general there is a crossover from diffusive behavior for $\lambda \gg \Lambda$ to propagating behavior for $\lambda \ll \Lambda$ where Λ is a mean free path due to nonlinear interactions. The present results explain why light- and neutron-scattering experiments probe different dynamical behavior of phasons.

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In an incommensurably modulated crystal the equilibrium positions of the atoms are modulated with a wavelength which is, at least in one direction, incommensurate with the underlying basic structure.[1] Examples include quasi one- and two-dimensional metals (for instance $NbSe_2$) as well as insulating materials [for instance $BaMnF_4$ (Ref. 2) and biphenyl[3]].

An interesting property of incommensurate crystals is the invariance of the free energy with respect to changes in the relative position of modulation and basic structure. These changes are generated by space-independent shifts of the phase of the order parameter which describes the modulation.[1] This invariance is expected to hold true as long as the modulation can be described by analytic functions.[4] The existence of an associated collective mode, the phason, with zero excitation energy follows immediately.

It has generally been argued that the phase independence of the free energy implies the existence of a gapless branch of propagating phase modulations (phasons) with a linear dispersion in the long-wavelength limit.[1] This prediction seems to agree with inelastic neutron-scattering experiments in biphenyl[3] which showed the existence of propagating phase fluctuations. However, light-scattering experiments in $BaMnF_4$ (Ref. 2) found evidence that phase fluctuations are purely diffusive, giving rise to a central peak in the spectrum of scattered light with width Dq^2. The latter result was interpreted in terms of a time-dependent Ginzburg-Landau theory which has been developed for charge-density-wave systems.[5,6]

It is the purpose of this Letter to demonstrate that both the propagating and the diffusive regimes of phase fluctuations follow quite naturally and for general reasons from the nature of the incommensurate state once the nonlinearities in the Hamiltonian are properly taken into account.

We start from the general form of the Hamiltonian for an anharmonic lattice,

$$H = \frac{1}{2}\sum_{\vec{k},j} \dot{A}(\vec{k},j)\dot{A}(-\vec{k},j) + \sum_{\nu=0}^{\infty} \frac{1}{\nu!} \sum_{\vec{k}_1,j_1;\cdots;\vec{k}_\nu,j_\nu} \Phi^{(\nu)}(-\vec{k}_1,j_1;\ldots;-\vec{k}_\nu,j_\nu)A(\vec{k}_1,j_1)\cdots A(\vec{k}_\nu,j_\nu). \quad (1)$$

$A(\vec{k},j)$ denotes the normal coordinates, j is a branch index, and $\vec{k}$ is a wave vector within the first Brillouin zone of the basic structure. $\dot{A}(\vec{k},j)$ denotes the time derivative of $A(\vec{k},j)$ and $\Phi^{(\nu)}$ are the expansion coefficients of the adiabatic potential.

We assume that the lattice transforms from the normal phase at high temperatures to an incommensurate phase at a temperature T_c and is incommensurate below T_c. This means that the expectation value of all normal coordinates is zero for $T > T_c$ and nonzero for some coordinates for $T < T_c$. In the simplest case the static modulation is caused by one primary wave vector $\vec{k}_0$ so that the expectation values $\langle A(n\vec{k}_0, j)\rangle$ are nonzero for $T < T_c$ where n is an integer.

It is convenient to introduce the fluctuations $\tilde{A}$ around the equilibrium positions of the incommensurate phase, i.e., $\tilde{A}(\vec{k}, j) = A(\vec{k}, j) - \langle A(\vec{k}, j)\rangle$. Elimination of A in favor of $\tilde{A}$ in Eq. (1) yields an expression for H in the incommensurate phase. It has the same form as the expression in Eq. (1) if A and $\Phi^{(\nu)}$ are replaced by $\tilde{A}$ and $\tilde{\Phi}^{(\nu)}$, where $\tilde{\Phi}^{(\nu)}$ are temperature-dependent expansion coefficients related to the coefficients $\Phi^{(\nu)}$ by a simple recursion formula.

Let us introduce new dynamical variables $P(\vec{q})$ by means of

$$P(\vec{q}) = \sum_{n,j} in\langle A(n\vec{k}_0, j)\rangle \tilde{A}(n\vec{k}_0 + \vec{q}, j). \quad (2)$$

$P(\vec{q})$ describes a modulation with wave vector $\vec{q}$ of the phase of the order parameter in a linear approximation.[1] We prefer to use $P(\vec{q})$ instead of the phase itself because $P(\vec{q})$ is connected with the original dynamical variables by a linear transformation which simplifies the dynamics greatly. The dynamical susceptibility $\chi(\vec{q}, z)$ associated with $P(\vec{q})$ satisfies the following equation[7]:

$$[z^2 - \Delta^2/\chi(\vec{q}) - z\Pi(\vec{q}, z)]\chi(\vec{q}, z) = -\Delta^2 \quad (3)$$

with

$$\Delta^2 = \sum_{n,j} n^2 |\langle A(n\vec{k}_0, j)\rangle|^2, \quad (4)$$

and

$$\Pi(\vec{q}, z) = \Delta^{-2} \subset QL^2P(\vec{q}) | (z - QLQ)^{-1} QL^2P(\vec{q}) \supset. \quad (5)$$

$\chi(\vec{q})$ is the static susceptibility $\chi(\vec{q}, z = 0)$. $\subset A | B \supset$ denotes Mori's scalar product of the two dynamical variables A and B. L is the Liouville operator and Ω projects out the components parallel to $P(\vec{q})$ and $LP(\vec{q})$.

The excitations of the system are determined by the poles of $\chi(\vec{q}, z)$ in the complex z plane. The second term in the square brackets of Eq. (3) is equal to the square of the oscillation frequency of phase modulations in the absence of irreversible processes. This term can be obtained from free-energy considerations. $\Pi(\vec{q}, z)$ is a complex self-energy. Its real part describes a frequency-dependent shift of the oscillation frequency, its imaginary part the damping. In the following we are interested in the low-frequency, long-wavelength behavior of the phase dynamics. It is therefore sufficient to consider the leading terms of $\chi(\vec{q})$ and $\Pi(\vec{q}, z)$ for small $\vec{q}$ and z.

The inverse of $\chi(\vec{q})$ is given by a second functional derivative of the free energy $F[P]$:

$$\chi^{-1}(\vec{q}) = \frac{\delta^2 F[P]}{\delta P(\vec{q})\, \delta P^*(\vec{q})}\bigg|_{P=0}. \quad (6)$$

$F[P]$ depends only on the single normal coordinate $P(\vec{q})$. It can be obtained from the free energy $F[A]$,[1] which is a functional of the normal coordinates $A(n\vec{k}_0 + \vec{q}, j)$, by summing $\exp(-F[A]/k_B T)$ over all variables except $P(\vec{q})$. This, in general, is a very difficult problem. We therefore restrict ourselves in this Letter to a mean-field approximation, i.e., we replace $F[P]$ by $F[A]$, putting

$$\tilde{A}(n\vec{k}_0 + \vec{q}, j) = -inP(\vec{q})\langle A(n\vec{k}_0, j)\rangle^*/\Delta^2. \quad (7)$$

Equation (7) is obtained from Eq. (2) by inversion. It gives the mean value of A for a given value of P provided the normal coordinates other than P have zero values. Thus fluctuations in the normal coordinates other than P are neglected in this approximation.

For $\vec{q} = 0$ we have the following expansion for $F[P]$:

$$F[P] = \sum_{\nu=0} F^{(\nu)} P^\nu, \quad (8)$$

with $P = P(0)$. Within the above approximation the coefficient $F^{(2)}$ is given by

$$F^{(2)} = -\frac{1}{\Delta^2} \sum_{\nu} \sum_{n_1 j_1 \ldots n_\nu j_\nu} f^{(\nu)}(n_1 j_1, \ldots, n_\nu j_\nu)\Delta(n_1\vec{k}_0 + \ldots + n_\nu\vec{k}_0)(n_1 n_2 + n_1 n_3 + \ldots + n_{\nu-1} n_\nu) \times \langle A(n_1\vec{k}_0, j_1)\rangle \cdots \langle A(n_\nu\vec{k}_0, j_\nu)\rangle. \quad (9)$$

$f^{(\nu)}$ is the Taylor expansion coefficient of $F[A]$ of νth order. Since this coefficient refers to the unmodulated structure it obeys the quasimomentum selection rule which has been written separately by means of the Kronecker function Δ. $\Delta(\vec{k})$ is equal to 1 if $\vec{k}$ is a reciprocal-lattice vector and zero

otherwise. By exploitation of the Kronecker function, Eq. (9) can be rewritten as

$$F^{(2)} = \frac{1}{2\Delta^2} \sum_{\nu=0}^{\infty} \sum_{n_1 j_1 \ldots n_\nu j_\nu} f^{(\nu)}(n_1 j_1, \ldots, n_\nu j_\nu) \times \Delta(n_1 \vec{k}_0 + \ldots + n_\nu \vec{k}_0) \Big(\sum_{\mu=1}^{\nu} n_\mu^2 \Big) \langle A(n_1 \vec{k}_0, j_1) \rangle \cdots \langle A(n_\nu \vec{k}_0, j_\nu) \rangle . \quad (10)$$

Let us consider the following fluctuation α around $\langle A(n\vec{k}_0, j)\rangle$:

$$\tilde{A}(n\vec{k}_0, j) = (n^2/2\Delta^2) \langle A(n\vec{k}_0, j)\rangle \alpha . \quad (11)$$

Inserting Eq. (11) into $F[A]$, one recognizes that the right-hand side of Eq. (10) represents just the first-order change in $F[A]$ due to α. However, this change has to vanish because F is stationary with respect to all first-order changes so that $F^{(2)} = 0$. Consequently we can write without loss of generality

$$\chi^{-1}(\vec{q}) = \sum_{\alpha=1}^{3} \frac{c_\alpha^2 q_\alpha^2}{\Delta^2} . \quad (12)$$

c_α has the meaning of a phase velocity for phase modulations propagating along the direction α in the absence of irreversible processes; see Eq. (3). The above derivation shows that the free energy does not depend on the linearized phase variable $P(0)$ up to second order in $P(0)$ [the higher orders in $P(0)$ do not vanish in general, as an extension of the above argument shows].

For sufficiently long wavelengths and low frequencies the dynamics of the phase modulations is determined by the hydrodynamic limit of $\Pi(\vec{q}, z)$, i.e., by $\lim_{z\to 0}\lim_{\vec{q}\to 0} \Pi(\vec{q}, z) = -i\gamma$ where γ is positive. Strictly speaking the existence of this limit is only guaranteed if all hydrodynamic variables (i.e., also the energy and momentum density and three displacement variables) are considered explicitly. To keep things simple we have omitted the other hydrodynamic variables from our discussion which is a good approximation if we are only interested in the leading contribution to γ.

The basic quantity in γ is the fluctuating force $QL^2P(0)$ which acts on the $\vec{q}=0$ phase modulation. Using Eq. (2) and the Hamiltonian of Eq. (1) rewritten in terms of the fluctuating variables $\tilde{A}(\vec{k}, j)$ we obtain from cubic anharmonicity

$$QL^2P(0) = \sum_{\substack{\vec{k}_2, j_2; \vec{k}_3, j_3 \\ n, j}} (in/2) \langle A(n\vec{k}_0, j)\rangle \tilde{\Phi}^{(3)}(-n\vec{k}_0, j; -\vec{k}_2, j_2; -\vec{k}_3, j_3) A(\vec{k}_2, j_2) A(\vec{k}_3, j_3) . \quad (13)$$

In calculating Eq. (13) we used the fact that there is no term $\sim \tilde{\Phi}^{(2)}$: It can be shown that $P(0)$ is an eigenfunction of $\tilde{\Phi}^{(2)}$ so that $L^2P(0)$ is proportional to $P(0)$ and $QL^2P(0)$ is therefore zero. Equation (13) shows that the fluctuating forces acting on $P(0)$ vanish in the harmonic limit but are nonzero if anharmonicity is taken into account. Physically this means that there are fluctuating forces acting on the order parameter if its phase is shifted rigidly. The divergence of χ for $\vec{q}\to 0$ tells us that the reversible part of the work necessary to shift the phase is zero. However, the irreversible part associated with this phase shift is nonzero, i.e., a change in the phase is associated with friction. This behavior is quite different from the case of acoustical phonons in the limit $\vec{q}\to 0$: There the corresponding eigenvectors describe rigid translations of the whole solid and experience no friction, so that the fluctuating forces vanish for $\vec{q}\to 0$.

The constant γ can be evaluated by inserting Eq. (13) into Eq. (5) and by approximating the intermediate states by the eigenfunctions of $\tilde{\Phi}^{(2)}$. Since the resulting expressions are complicated we will limit ourselves to a simple case: We assume that the intermediate states can be approximated by the phonons of the high-temperature phase (i.e., the reconstruction of the excitation spectrum near $\vec{q} \sim \pm \vec{k}_0$ can be neglected). The expression for γ becomes in the classical limit

$$\gamma = \frac{\pi T}{4\Delta^2} \sum_{\substack{nj, n'j' \\ \vec{k}_1 j_1, \vec{k}_2 j_2}} nn' \langle A(n\vec{k}_0, j)\rangle^* \langle A(n'\vec{k}_0, j')\rangle \frac{\tilde{\Phi}^{(3)*}(n\vec{k}_0, j; -\vec{k}_1, j_1; -\vec{k}_2, j_2) \tilde{\Phi}^{(3)}(n'\vec{k}_0, j'; -\vec{k}_1, j_1; -\vec{k}_2, j_2)}{\omega^4(\vec{k}_1, j_1)} \times \delta(\omega(\vec{k}_1, j_1) - \omega(\vec{k}_2, j_2)) . \quad (14)$$

It shows that γ is indeed nonzero for $T \neq 0$ and that only difference processes contribute. γ varies continuously with temperature near T_c. The critical temperature dependence due to Δ^2 in the prefactor is canceled out by the temperature dependence of the two expectation values of A. If the plane-wave limit for the order parameter applies, expression (14) is identical with the $z \to 0$ limit of the self-energy of the soft phonon. We therefore expect that γ is of the order of 1 cm^{-1} for most incommensurate crystals at temperatures of order T_c.

In the classical limit Eq. (3) yields the following dynamic structure factor:

$$S(\vec{q}, \omega) = \frac{2T\gamma\Delta^2}{(\omega^2 - \sum_\alpha c_\alpha^2 q_\alpha^2)^2 + \omega^2\gamma^2}. \quad (15)$$

The poles of the denominator of S are at $\omega = \frac{1}{2}[-i\gamma \pm (-\gamma^2 + 4\sum_\alpha c_\alpha^2 q_\alpha^2)^{1/2}]$. One therefore has to distinguish between two cases:

(a) $c_\alpha q_\alpha \ll \gamma/2$. The position of the poles is then approximately given by $-i\gamma$ and $-i\sum_\alpha c_\alpha^2 q_\alpha^2/\gamma$. An analysis of Eq. (15) shows that the relaxation pole $-i\gamma$ has only a small weight in S so that

$$S(\vec{q}, \omega) \sim \frac{2T\Delta^2}{\gamma} \frac{1}{\omega^2 + (\sum_\alpha c_\alpha^2 q_\alpha^2/\gamma)^2}. \quad (16)$$

Physically one obtains the following picture: If fluctuations in the phase of the order parameter with wavelengths larger than $\Lambda = 2c_\alpha/\gamma$ are induced the order parameter will reach its equilibrium value again via diffusion processes which are due to internal anharmonic interactions.

(b) $c_\alpha q_\alpha \gg \gamma/2$. The position of the poles is then approximately at $\omega = -i\gamma/2 \pm (\sum_\alpha c_\alpha^2 q_\alpha^2)^{1/2}$. This means that the phase responds to perturbations with wavelengths smaller than Λ by carrying out damped oscillations around its equilibrium value.

The transition between diffusive and oscillating response occurs for $q \sim 1/\Lambda$. If we take for c_α typical values for sound velocities and the above estimate for γ, Λ is of the order of 100 Å. Thus we would expect that light scattering experiments would probe mainly the diffusive regime whereas inelastic neutron scattering experiments would probe mainly the propagating regime.

Finally we would like to make a comment on the relationship between a continuous broken symmetry and Goldstone modes.[8,9] Usually it is said that a continuous symmetry is broken if the ground state of the system (or more generally the density matrix) is not invariant under the corresponding symmetry operations whereas the total Hamiltonian H is. This implies that the generators of those symmetry operations which restore the full symmetry commute with H and give rise to conservation laws. For instance the averaged one-particle density in a solid breaks the continuous translational symmetry. The total momentum operator $\vec{P}$ generates infinitesimal rigid translations which restore the full translational symmetry. $\vec{P}$ commutes with H and is therefore a conserved quantity. The continuous broken symmetry and the conservation of the toral momentum are interrelated and as a result there exist propagating sound waves. In our case the generator of infinitesimal shifts in the phase of the order parameter does not commute with H. This follows for instance from the explicit dependence of H on the phase $P(0)$ once the anharmonicity is taken into account. As a result a continuous symmetry of the free energy but not of the Hamiltonian is broken. Phasons are therefore not propagating Goldstone modes but are always diffusive for sufficiently long wavelengths.

We think that the general conclusions outlined above are in agreement with the available experimental data. Phasons have been seen as propagating excitations in biphenyl[3] in inelastic neutron-scattering experiments where the momentum transfer is large. On the other hand many attempts to observe phasons in the form of Brillouin doublets by light-scattering experiments were unsuccessful.[10] Recent quasielastic light-scattering data in the incommensurate phase of $BaMnF_4$ (Ref. 2) show instead a central peak (different from the Rayleigh peak caused by entropy fluctuations) with a $\vec{q}$ dependence as predicted by Eq. (16). Taking the observed sound velocity of transverse phonons as a first approximation for c_α we find that the measured widths of the central peak (Fig. 6 of Ref. 2) correspond to the values $\gamma \sim 1.7$ and 2.5 cm^{-1}, which are in the same range as the value estimated above.

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Superfluidity of a Spin-$\frac{1}{2}$ Bose Fluid: Spin-Polarized Hydrogen

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It is shown here that superfluids (such as ^{3}He-A and Bose-condensed spin-polarized hydrogen) whose order parameters transform like an angular momentum eigenstate $|j, m \neq 0\rangle$ will have similar superfluidity—similar Josephson and vorticity equations. In particular, if spin-polarized hydrogen condenses into the b state as currently believed, it will not have a stable superflow. Nonuniform magnetic fields can also induce in it a persistent current similar to that of ^{3}He-A.

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The recent stabilization of atomic spin-polarized hydrogen[1] (SPH) has generated much hope in the observation of Bose-Einstein condensation in this system. A hydrogen (H) atom can be regarded as a boson because it contains two fermions. Recently, Siggia and Ruckenstein[2] pointed out that SPH, when condensed, will behave like a spin-$\frac{1}{2}$ Bose superfluid. This is because at low temperatures, only the lowest two hyperfine states, usually refered to as a and b, are important. They correspond to the alignment and misalignment of the proton spin with the external field, modified by the hyperfine interaction. The electron spin is basically held fixed by the external field because it has a magnetic moment (μ_e) 10^3 times larger than the proton's (μ_p). More recently, Statt and Berlinsky[3] pointed out that because of molecular recombination effects, condensation is mostly likely to occur in the b state.

The purpose of this paper is to show that, no matter which hyperfine state SPH condenses into, the flow properties of this prospective $S=\frac{1}{2}$ Bose superfluid will be very similar to those of ^{3}He-A, a p-wave BCS superfluid well known for its peculiar superfluidity. In fact, we shall see that all superfluids whose order parameter transforms like an angular momentum eigenstate $|j, m \neq 0\rangle$ will have similar flow properties.

^{3}He-A consists of Cooper pairs which are orbital angular momentum eigenstates $|L=1, \vec{L}\cdot\hat{l}=1\rangle$ along a certain direction $\hat{l}$. The pair wave function is $Y_{11}(\vec{\rho}) \propto \hat{\varphi}\cdot\vec{\rho}$, where $\hat{\varphi}=\hat{\varphi}_1+i\hat{\varphi}_2$, $\hat{\varphi}_1\cdot\hat{\varphi}_2=0$, $\hat{l}=\hat{\varphi}_1\times\hat{\varphi}_2$, and $\vec{\rho}$ is the relative displacement of the helium atoms in the pair. The triad structure of the orbital order parameter $\hat{\varphi}$ is the cause of the peculiar flow properties of ^{3}He-A. For example, in simply connected containers (because of the surface boundary condition on $\hat{l}$), ^{3}He-A will carry a persistent current whose magnitude is of the order of a few vortices.[4] On the other hand, superflows (with larger winding numbers) in multiply connected containers are not intrinsically stable.[5] All these are very different from the behavior of the more familiar superfluids such as ^{4}He and superconductors, whose order parameters are scalars.

That SPH can be treated as a $S=\frac{1}{2}$ Bose gas can be formulated as follows. The Hamiltonian of SPH is $H=T+Z+V$, where T is the kinetic energy, V is the interaction between the H atoms, and Z is the sum of the Zeeman energy and the hyperfine interaction (g),

$$Z=\int d^3x\,\hat{\eta}^\dagger[\tfrac{1}{2}(\mu_e\vec{\sigma}-\mu_p\vec{\tau})\cdot\vec{H}_{\rm ext}+\tfrac{1}{4}g\vec{\tau}\cdot\vec{\sigma}]\hat{\eta}.$$

Here, $\vec{\tau}$ ($\vec{\sigma}$) is the proton (electron) Pauli matrix, $\vec{H}_{\rm ext}(\vec{r})$ is the external field, and $\hat{\eta}=(\hat{\eta}_{\mu i})$ is the hydrogen Bose operator with electron (proton) spin index μ (i). It is straightforward to show that the matrix $U=\exp[i\vec{\tau}\times\vec{\sigma}\cdot\hat{n}\epsilon/4]$, with $\epsilon=\cot^{-1}[(\mu_p+\mu_e)H_{\rm ext}/g]$ and $\hat{n}=\hat{H}_{\rm ext}$, diagonalizes Z locally, so that

$$Z=\int d^3x\,\hat{\eta}^\dagger U^\dagger[\tfrac{1}{2}(\bar{\mu}_e\vec{\sigma}-\bar{\mu}_p\vec{\tau})\cdot\vec{H}_{\rm ext}+\tfrac{1}{4}g(\vec{\tau}\cdot\hat{n})(\vec{\sigma}\cdot\hat{n})]U\hat{\eta},$$