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Textures of Quantum Intrinsically Localized Modes

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Abstract

We have examined the lowest-energy members of the quantized ILMs of a generalization of the Fermi-Pasta-Ulam Hamiltonian to three-dimensions. The lowest energy ILMs are similar in form to multi-phonon bound states, except that the number of phonons is not conserved. The ILMs can be categorized as having a quasi-spin of either $S = 2$ or $S = 0$ and have other internal quantum numbers. We find that ILMs can form in three-dimensions at zero temperature, but only if the interaction exceeds a minimum value. Furthermore, as the temperature is raised, the magnitude of the minimal interaction required to stabilize the ILM is reduced. When the ILMs first form they split off from the top of the two-phonon continuum. The $S = 0$ ILMs form for lower values of the interaction than the $S = 2$ ILMs. The ILMs form preferentially for center of mass momentum $\underline{q}$ at the corner of the Brillouin zone. The tendency of ILMs to form at this momentum is traced to a confluence of van-Hove singularities in the (non-interacting) two-phonon density of states at the top of the two-phonon continuum. We have examined the ILM many-body wave functions and find that the relative coordinate part of the wave functions have symmetries associated with internal quantum numbers.

INTRODUCTION

Intrinsically Localized Modes (ILMs), or discrete breathers, are persistent oscillatory excitations of spatially homogeneous systems that have finite spatial extents and are stabilized by anharmonic interactions [1]. The ILM excitations resemble the localized vibrational modes found in the vicinity of lattice defects [2], but differ in a number of respects. Firstly, the ILMs can be centered about any site of a homogeneous lattice, whereas the defect modes are centered on impurity sites. Secondly, defect modes can exist in purely harmonic lattices, however anharmonic interactions are essential for the existence of ILMs. Early reviews of ILMs are found in refs.[3, 4]. In that ILMs have finite spatial extents and are essentially non-linear modes, they form a class of excitations which are similar to the class of soliton excitations. Soliton excitations are defined as localized excitations that have persistent shapes found in systems that are continuous, homogeneous and non-linear [5–8]. Solitons are also known to persist in discrete lattice systems [9, 10]. In fact, breather excitations (which closely resemble ILMs) were discovered to be excitations of the sine-Gordon equation [6] and the Korteweg - de Vries (KdV) equation [11]. The KdV equation was formulated to describe the solitary water waves in long shallow channels. The solitary waves of the KdV equation form the archetypical example of soliton excitations of one-dimensional non-linear systems [12]. Derrick's theorem [13] precludes solitons from occurring in two or higher-dimensional systems. Gardner *et al.* [14] and Lax [15] showed that the KdV equation is exactly integrable and, despite the non-linear nature of the equation, only has solutions that can be pictured as linear combinations of small-amplitude excitations together with a countable number of solitons. In this picture, the breather excitations can be thought of as being bound states of a pair of soliton and anti-soliton excitations, in which the oscillatory nature of the breather has its origin in the relative motion of the soliton/anti-soliton pair. For classical systems, since the state of relative motion is part of a continuum, the breather's energy spectrum is continuous. Breather excitations were also found to exist in discrete, classical, anharmonic lattices [16]. The non-existence of soliton excitations in higher-dimensional systems [13] would make it seem that breather excitations (specifically viewed as bound states of a soliton/anti-soliton pair) should also be restricted to one-dimensional systems. Despite this apparent restriction, in 1988, Sievers and Takeno [1] suggested that ILMs might also be found in higher-dimensional lattice systems. ILMs were found in two and three dimensional

classical models within the rotating wave approximation [17]. In 1997, Kiselev and Sievers [18] performed classical simulations which indicated that persistent oscillations could be expected to occur in cubic anharmonic crystals.

In 2009, Manley *et al.* performed inelastic neutron scattering experiments [19] on NaI and observed that, at elevated temperatures, the system showed a branch of excitations that cannot simply be described in terms of harmonic phonons. These authors found that at high temperatures, NaI has a branch of dispersionless excitations which exists for wave vectors $\underline{q}$ along the (1,1,1) direction close to the corner of the Brillouin zone boundary and which are polarized along the (1,1,1) direction. As seen in fig.(1), the excitations were found to have an energy of approximately 10 meV which is bounded from above by a branch of optic phonons and from below by the maximum of the branch of acoustic phonons. Manley *et al.* interpreted the anomalous excitations in terms of classical ILMs. However, unlike the ILMs observed in classical simulations which have continuous spectra, the anomalous modes have sharp discrete spectral peaks. The discrete nature of the spectrum of anomalous excitations can be taken as an indication of the quantum nature of the excitations. The need for a quantum interpretation of the reported ILMs is supported by classical simulations which indicate that the ILM excitation energies can be as large as 600 meV [20], which would preclude ILMs being seen in thermal equilibrium. The inadequacy of a classical description of the proposed ILM excitations also follows from consideration of the binding energy. In the classical theory, the binding energy is defined as the energy separation between the ILM energy and the zone-boundary phonon frequency. The value of the classical binding energy inferred from the experiment would requires a much larger value of the anharmonic interaction than is expected from standard two-body potentials [20]. Independent inelastic neutron scattering experiments were performed by Kempa *et al.* [21] which do not support the proposal that ILMs exist in NaI. Some of the uncertainty may arise from the unusual temperature dependence of the ILM features [22, 23] combined with the uncertainty in the measured temperatures.

Dashen, Hasslacher and Neveu [24, 25] showed that when the breather excitations of the continuous, exactly integrable and one-dimensional sine-Gordon field theory are quantized semi-classically, they form a hierarchy of discrete excitations. Moreover, the hierarchy of quantum breather excitations can be thought of as being formed by the hierarchy of bound states of multiples of the quantized small amplitude oscillations (phonons) [26]. Numerical

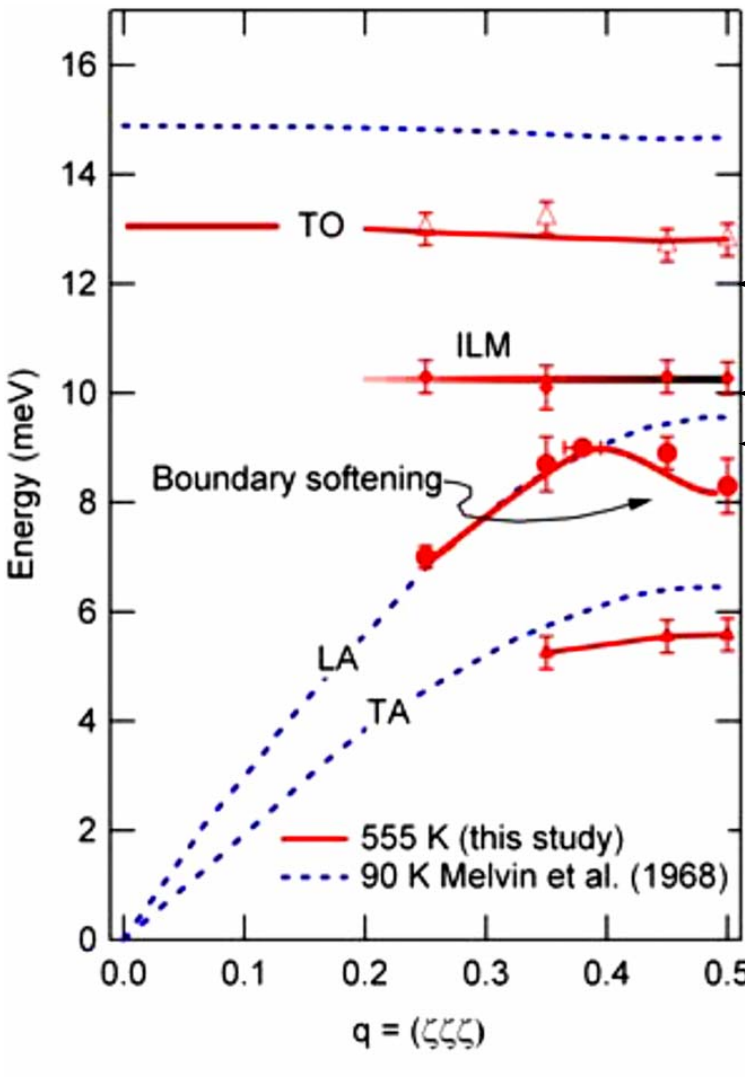

FIG. 1. (Color on line) Summary of lattice dynamics measured in NaI along the [111] direction, showing the phonon dispersion curves measured at 90 K (dashed lines) and 555 K (red lines), including a dispersionless local mode (ILM) that appears in the gap between the branches of acoustic and optic phonons. [From Manley *et al.* (2009).]

calculations on non-linear Klein-Gordon lattice models [27, 28] shows that this description of quantum breathers carries over from continuous integrable systems to non-integrable discrete lattice systems. The continuum limit of the one-dimensional Fermi-Pasta-Ulam model [29] is described by the KdV equation [30], which also exhibits breather excitations [31]. Therefore, it is not surprising that the lowest-energy excitations of the discrete quantum Fermi-Pasta-Ulam model can be described in terms of bound states of multiple numbers of phonons [32] and that the excitations have localized characters [33]. Analytic methods have been proposed

to describe the higher-energy members of this hierarchy [34]. For a diatomic Fermi-Pasta-Ulam lattice [32], the lowest-energy excitations have dispersion relations that are bounded from above by the optic phonon dispersion relation and are bounded from below by the two-acoustic phonon continuum, in agreement with the observations on NaI. The calculations of ref.[32] show that the breather excitations of the discrete one-dimensional lattice are guaranteed to exist for arbitrarily small values of the anharmonic interaction, due to the effects of the divergent van-Hove singularities in the non-interacting multi-phonon density of states. This suggests that quantized breather excitations could also exist in higher-dimensional lattices [35], if the value of the anharmonic interaction exceeds a critical value determined by the magnitude of the multi-phonon density of states.

Here, we present the generalization of the quantized Fermi-Pasta-Ulam model to higher spatial dimensions. We present the detailed calculation of the two-phonon propagator which gives us the spectrum of interacting two-phonon excitations. We find that, in addition to a continuum of scattering states, there may be branches of two-phonon resonances or bound states. Therefore, the lowest-energy breather excitations of a cubic three-dimensional crystal are consistent with the description of Dashen *et al.* [24, 25] as being the bound states of multiple numbers of phonons. We examine the ILM excitations with center of mass momenta directed along the high-symmetry lines of the Brillouin zone. We find that ILMs form preferentially at positions in the Brillouin zone where the van-Hove singularity at the upper edge of the two-phonon continuum is sharpest. A short report of our preliminary results has been published [36].

THE D=3 QUANTUM FERMI-PASTA-ULAM LATTICE

The Fermi-Pasta-Ulam lattice [29] consists of a one-dimensional array of atoms which are connected to their nearest neighbors by weakly anharmonic interactions. Fermi, Pasta and Ulam originally considered two types of anharmonicity; the α-lattice corresponded to cubic anharmonicity and the β-lattice corresponded to a quartic anharmonic interaction. The cubic anharmonic interaction makes the α-system unstable, so henceforth we shall only consider the β-lattice. The quantized Hamiltonian describing the β-Fermi-Pasta-Ulam

lattice can be written as

$$\hat{H} \;=\; \sum_{i} \; \Big[\; \frac{\hat{P}_i^2}{2\;M} \;+\; \frac{K_2}{2!} \; (\; \hat{u}_i \;-\; \hat{u}_{i+1} \;)^2 \;+\; \frac{K_4}{4!} \; (\; \tilde{u}_i \;-\; \hat{u}_{i+1} \;)^4 \; \Big] \tag{1}$$

where $\hat{P}_i$ is the momentum operator of the i-th atom and $\hat{u}_i$ is the displacement operator of the i-th atom from its equilibrium position. The constant M is the mass of the atoms, K_2 represents the strength of the harmonic component of the interaction and K_4 determines the strength of the anharmonic interaction.

In the three-dimensional generalization of the Fermi-Pasta-Ulam model, the labels i of the lattice sites are replaced by the lattice vectors $\underline{R}$ of a periodic three-dimensional crystal with a mono-atomic basis. Also, the interaction between the atom at site $\underline{R}$ only occurs with the nearest neighboring lattice sites which are located at positions denoted by $\underline{R} + \underline{\delta}$. The resulting model describes longitudinal and transverse phonon modes on the same footing. Since the Hamiltonian is to be a scalar quantity, the various terms in the Hamiltonian must be expressed as scalar products of the vector momentum and displacement operators [36]. Hence, we have

$$\begin{aligned} \hat{H} = \sum_{\underline{R}} \; \Big[\; & \frac{\hat{\underline{P}}_{\underline{R}} \cdot \hat{\underline{P}}_{\underline{R}}}{2\;M} \;+\; \frac{1}{2} \sum_{\underline{\delta}} \; \frac{K_2}{2!} \; (\; \hat{\underline{u}}_{\underline{R}} \;-\; \hat{\underline{u}}_{\underline{R}+\underline{\delta}} \;) \;.\; (\; \hat{\underline{u}}_{\underline{R}} \;-\; \hat{\underline{u}}_{\underline{R}+\underline{\delta}} \;) \\ & + \frac{1}{2} \sum_{\underline{\delta}} \frac{K_4}{4!} \; \Big(\; (\; \tilde{\underline{u}}_{\underline{R}} \;-\; \hat{\underline{u}}_{\underline{R}+\underline{\delta}} \;) \;.\; (\; \tilde{\underline{u}}_{\underline{R}} \;-\; \hat{\underline{u}}_{\underline{R}+\underline{\delta}} \;) \; \Big)^2 \; \Big] \end{aligned} \tag{2}$$

The factor of one-half occurring before the sum over $\underline{\delta}$, ensures that the interaction between a pair of atoms is only counted once. This Hamiltonian differs from the classical Hamiltonian previously investigated by Bickham and Sievers [17], in which they assumed that the anharmonic interaction only coupled the same components of the displacements. The anharmonic mixing between the different components of a vector field opens the possibility that the ILM may have a rich variety of internal structures including different spatial forms and, as we shall argue, also may have various values of an intrinsic quasi-spin.

The harmonic part of the Hamiltonian can be diagonalized. First, the vector momentum and displacement operators are expressed in terms of their spatial Fourier transforms defined by

$$\begin{aligned} \hat{\underline{P}}_{\underline{q}} &= \frac{1}{\sqrt{N}} \; \sum_{\underline{R}} \; \exp[\; i \; \underline{q} \;.\; \underline{R} \;] \; \hat{\underline{P}}_{\underline{R}} \\ \hat{\underline{u}}_{\underline{q}} &= \frac{1}{\sqrt{N}} \; \sum_{\underline{R}} \; \exp[\; i \; \underline{q} \;.\; \underline{R} \;] \; \hat{\underline{u}}_{\underline{R}} \end{aligned} \tag{3}$$

where N denotes the total number of sites in the lattice. The resulting harmonic part of the Hamiltonian, $\hat{H}_0$, can be expressed as

$$\hat{H}_0 \;=\; \sum_{\underline{q}} \; \Big[\; \frac{\hat{\underline{P}}_{\underline{q}} \cdot \hat{\underline{P}}^{\dagger}_{\underline{q}}}{2\; M} \;+\; \frac{K_2}{2!} \; \sum_{\underline{\delta}} \; \Big(\; 1 \;-\; \cos(\underline{q} \;.\; \underline{\delta}) \; \Big) \; \hat{\underline{u}}_{\underline{q}} \cdot \hat{\underline{u}}^{\dagger}_{\underline{q}} \; \Big] \tag{4}$$

where the sum over $\underline{q}$ is restricted to run over the first Brillouin zone. It should be noted that as the scalar products are invariant under rotations, the harmonic Hamiltonian is diagonal when expressed in terms of any complete set of orthonormal basis vectors $\hat{\underline{\epsilon}}_p$. The vector operators are expressed in terms of their components as

$$\begin{aligned} \hat{\underline{P}}_{\underline{q}} &= \sum_{p=1}^{p=d} \; \hat{P}_{\underline{q},p} \; \hat{\underline{\epsilon}}_p(\underline{q}) \\ \hat{\underline{u}}_{\underline{q}} &= \sum_{p=1}^{p=d} \; \hat{u}_{\underline{q},p} \; \hat{\underline{\epsilon}}_p(\underline{q}) \end{aligned} \tag{5}$$

Thus, the harmonic approximation to the Hamiltonian takes the form

$$\hat{H}_0 \;=\; \sum_{\underline{q},p} \; \Big[\; \frac{\hat{P}_{\underline{q},p} \; \hat{P}^{\dagger}_{\underline{q},p}}{2\; M} \;+\; \frac{K_2}{2!} \; \sum_{\underline{\delta}} \; \Big(\; 1 \;-\; \cos(\underline{q} \;.\; \underline{\delta}) \; \Big) \; \hat{u}_{\underline{q},p} \; \hat{u}^{\dagger}_{\underline{q},p} \; \Big] \tag{6}$$

since the polarization vectors form an orthonormal set

$$\hat{\underline{\epsilon}}_p(\underline{q}) \;.\; \hat{\underline{\epsilon}}_{p'}(\underline{q}) \;=\; \delta_{p,p'} \tag{7}$$

Therefore, the harmonic Hamiltonian is expressed as the sum of harmonic oscillators where the sum runs over all the normal mode labels $(\underline{q}, p)$. The frequencies $\omega_{\underline{q},p}$ of the harmonic modes are determined from

$$\omega^2_{\underline{q},p} \;=\; 2\; \frac{K_2}{M} \; \sum_{\underline{\delta}} \; \sin^2 \Big(\frac{\underline{q} \;.\; \underline{\delta}}{2} \Big) \tag{8}$$

and are independent of the polarization index p [17]. Henceforth, we shall set $K_2 \;=\; M\; \omega_0^2$. It is seen that, in the harmonic limit, the three-dimensional Fermi-Pasta-Ulam model describes three branches of acoustic phonon modes, which are required by Goldstone's theorem [37]. We have implicity assumed that, in our ground state, the atoms of our three-dimensional solid are located at specific positions $\underline{R}$ so that our choice of ground state is not invariant under arbitrary uniform translations although the Hamiltonian is. The harmonic part of the

Hamiltonian can be second quantized by expressing the operators for each normal mode in terms of its boson creation and annihilation operators via

$$\hat{P}_{\underline{q},p} = i \sqrt{\frac{M\hbar\omega_{\underline{q},p}}{2}} \; (\; a^{\dagger}_{-\underline{q},p} - a_{\underline{q},p} \;)$$
$$\hat{u}_{\underline{q},p} = \sqrt{\frac{\hbar}{2M\omega_{\underline{q},p}}} \; (\; a^{\dagger}_{\underline{q},p} + a_{-\underline{q},p} \;) \tag{9}$$

Thus, the harmonic Hamiltonian can be expressed as

$$\hat{H}_0 \; = \; \sum_{\underline{q},p} \; \frac{\hbar\omega_{\underline{q},p}}{2} \; (\; a^{\dagger}_{\underline{q},p} \; a_{\underline{q},p} \; + \; a_{\underline{q},p} \; a^{\dagger}_{\underline{q},p} \;) \tag{10}$$

which is diagonal in the momentum and polarization indices. Hence, the phonons have acoustic dispersion relations which are completely degenerate with respect to their polarizations. The transformational properties of the small-amplitude lattice displacements, in our case of degenerate polarizations, results in the phonon having an intrinsic quasi-spin of unity. Spin-one phonons also exist in continuum theories where there are spin-like conserved quantities [40] which in the isotropic limit [41] transform like spin .

The anharmonic interaction part of the Hamiltonian is written in terms of the lattices sites $\underline{R}$ and the nearest neighboring lattice sites located at $\underline{R} + \underline{\delta}$ as

$$\hat{H}_{int} = \frac{1}{2} \; \sum_{\underline{R}} \sum_{\underline{\delta}} \frac{K_4}{4!} \; \Big(\; (\; \hat{\underline{u}}_{\underline{R}} \; - \; \hat{\underline{u}}_{\underline{R}+\underline{\delta}} \;) \; . \; (\; \hat{\underline{u}}_{\underline{R}} \; - \; \hat{\underline{u}}_{\underline{R}+\underline{\delta}} \;) \Big)^2 \tag{11}$$

Using equations (3) and (5), the interaction $\hat{H}_{int}$ can be re-written as

$$\hat{H}_{int} = \frac{1}{2 \; N} \sum_{\underline{\delta}} \frac{K_4}{4!} \sum_{\underline{k}_1,\underline{k}_2,\underline{k}_3,\underline{k}_4} \Delta_{\underline{k}_1+\underline{k}_2+\underline{k}_3+\underline{k}_4} \; \exp\Big[i(\underline{k}_1 + \underline{k}_2 + \underline{k}_3 + \underline{k}_4).\frac{\underline{\delta}}{2} \Big]$$
$$\times \; 16 \; \sin(\frac{\underline{k}_1.\underline{\delta}}{2}) \sin(\frac{\underline{k}_2.\underline{\delta}}{2}) \sin(\frac{\underline{k}_3.\underline{\delta}}{2}) \sin(\frac{\underline{k}_4.\underline{\delta}}{2}) \; \sum_{p_1,p_2} \; \hat{u}_{\underline{k}_1,p_1} \hat{u}_{\underline{k}_2,p_1} \hat{u}_{\underline{k}_3,p_2} \hat{u}_{\underline{k}_4,p_2} \tag{12}$$

In this expression the function $\Delta_{\underline{k}}$ is a Kronecker delta function which conserves the total momentum, modulo a reciprocal lattice vector, and is defined by

$$\Delta_{\underline{k}} \; = \; \sum_{\underline{G}} \; \delta_{\underline{k}+\underline{G}} \tag{13}$$

where the sum runs over all the reciprocal lattice vectors $\underline{G}$. On substituting the expressions for $\hat{u}_{\underline{k},p}$ given in eqn.(9), the interaction Hamiltonian takes the second quantized form

$$\hat{H}_{int} = \frac{I_4}{4! \; N} \sum_{p_1,p_2} \; \sum_{\underline{k}_1,\underline{k}_2,\underline{k}_3,\underline{k}_4} \; \Delta\Big(\sum_{m=1}^{4} \underline{k}_m \Big)$$

$$\times \sum_{\underline{\delta}} \prod_{n=1}^{2} F^{\underline{\delta}}_{\underline{k}_n} (a^{\dagger}_{\underline{k}_n,p_1} + a_{-\underline{k}_n,p_1}) \prod_{m=3}^{4} F^{\underline{\delta}}_{\underline{k}_m} (a^{\dagger}_{\underline{k}_m,p_2} + a_{-\underline{k}_m,p_2}) \tag{14}$$

where the complex form factors $F_{\underline{k}_n.\underline{\delta}}$ are given by

$$F^{\underline{\delta}}_{\underline{k}_n} = \frac{\sin \frac{\underline{k}_n.\underline{\delta}}{2}}{\left[\sum_{\underline{\delta}'} \sin^2 \frac{\underline{k}_n.\underline{\delta}'}{2} \right]^{\frac{1}{4}}} \exp\left[i \frac{\underline{k}_n.\underline{\delta}}{2} \right] \tag{15}$$

and I_4 is the strength of the anharmonic interaction

$$I_4 = \frac{\hbar^2 K_4}{2\ M^2 \omega_0^2}. \tag{16}$$

which has units of energy.

THE TWO-PHONON PROPAGATOR

The two-phonon propagator has the form of a four by four matrix $D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k}, t : \underline{k}', 0)$, which involves the expectation value of the product of two two-phonon operators $\hat{A}^{(\alpha)}_{\sigma;\underline{q}}(\underline{k}, t)$; one product evaluated at time t and the other product at time zero. The components of the two-phonon propagator are expressed as

$$D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k}, t; \underline{k}', 0) \ = \ - \frac{i}{\hbar} \langle\ \hat{T}\ \hat{A}^{(\alpha)}_{\sigma;\underline{q}}(\underline{k}, t)\ \hat{A}^{(\beta)\dagger}_{\sigma';\underline{q}}(\underline{k}', 0)\ \rangle \tag{17}$$

where $\hat{T}$ is Wick's time-ordering operator. The two-phonon operators $\hat{A}^{(\alpha)}_{\sigma;\underline{q}}(\underline{k})$ are defined as

$$\begin{aligned}
\hat{A}^{(++)}_{\sigma;\underline{q}}(\underline{k}) &\equiv a^{\dagger}_{\frac{q}{2}+\underline{k},p_1}\ a^{\dagger}_{\frac{q}{2}-\underline{k},p_2} \\
\hat{A}^{(+-)}_{\sigma;\underline{q}}(\underline{k}) &\equiv a^{\dagger}_{\frac{q}{2}+\underline{k},p_1}\ a_{-\frac{q}{2}+\underline{k},p_2} \\
\hat{A}^{(-+)}_{\sigma;\underline{q}}(\underline{k}) &\equiv a_{-\frac{q}{2}-\underline{k},p_1}\ a^{\dagger}_{\frac{q}{2}-\underline{k},p_2} \\
\hat{A}^{(--)}_{\sigma;\underline{q}}(\underline{k}) &\equiv a_{-\frac{q}{2}-\underline{k},p_1}\ a_{-\frac{q}{2}+\underline{k},p_2}
\end{aligned} \tag{18}$$

where the ordered pair of indices (a_1, a_2), in which a_1 and a_2 are taken from the set of $\{+, -\}$, are denoted by a single index (α) that takes on four values. Likewise, the ordered pair of polarization indices (p_1, p_2) are denoted by a single index σ. To avoid ambiguity between two phonon states labeled by σ and its permutation, momentum space will be separated into two disjoint volumes, such that if the point $\underline{k}$ falls within one region, the value of $-\underline{k}$

falls within the other region. Henceforth, we adopt the convention that $\underline{k}$ is restricted to one region. The components of the propagator can be obtained from the equation of motion

$$i\,\hbar\,\frac{\partial}{\partial t}\,D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k},t;\underline{k}',0) = \delta(t)\,\langle\,[\,\hat{A}^{(\alpha)}_{\sigma;\underline{q}}(\underline{k},t)\;,\;\hat{A}^{(\beta)\dagger}_{\sigma';\underline{q}}(\underline{k}',0)\,]\,\rangle - \frac{i}{\hbar}\,\langle\,\hat{T}\,[\,\hat{A}^{(\alpha)}_{\sigma;\underline{q}}(\underline{k},t)\;,\;\hat{H}(t)\,]\,\hat{A}^{(\beta)\dagger}_{\sigma';\underline{q}}(\underline{k}',0)\,\rangle \tag{19}$$

where both terms on the right-hand side only involve equal-time commutators. The inhomogeneous term

$$[\,\hat{A}^{(\alpha)}_{\sigma;\underline{q}}(\underline{k},0)\;,\;\hat{A}^{(\beta)\dagger}_{\sigma';\underline{q}}(\underline{k}',0)\,] \;=\; [\,a^{a_1}_{a_1(\frac{q}{2}+\underline{k}),p_1}a^{a_2}_{a_2(\frac{q}{2}-\underline{k}),p_2}\;,\;a^{b_2\dagger}_{b_2(\frac{q}{2}-\underline{k}'),p'_2}a^{b_1\dagger}_{b_1(\frac{q}{2}+\underline{k}'),p'_1}\,] \tag{20}$$

can be evaluated as

$$=-\,\Delta_{\underline{k}-\underline{k}'}\,\delta^{\alpha,\beta}_{\sigma,\sigma'}\,\left[\,a_2\,\left(\frac{1}{2}+N_{a_1(\underline{q}/2+\underline{k}),p_1}\right)\;+\;a_1\,\left(\frac{1}{2}+N_{a_2(\underline{q}/2-\underline{k}),p_2}\right)\,\right] \tag{21}$$

where

$$N_{\underline{k},p}\;=<\;a^{\dagger}_{\underline{k},p}\,a_{\underline{k},p}\;> \tag{22}$$

is the average number of phonons in the mode with wave vector $\underline{k}$ and polarization p.

In the absence of the interaction, the two-phonon propagators can be found exactly. For purely harmonic interactions, the commutator with the interaction can be evaluated exactly, leading to the equations of motion

$$\left(\,i\,\hbar\,\frac{\partial}{\partial t}\;+\;a_1\,\hbar\,\omega_{a_1(\frac{q}{2}+\underline{k}),p_1}\;+\;a_2\,\hbar\,\omega_{a_2(\frac{q}{2}-\underline{k}),p_2}\,\right)\,D_{0\,\sigma,\sigma';\underline{q}}^{(\alpha),(\beta)}(\underline{k},t;\underline{k}',0) = -\,\delta(t)\,\Delta_{\underline{k}-\underline{k}'}\,\delta^{\alpha,\beta}_{\sigma,\sigma'}\,\left[\,a_2\,\left(\frac{1}{2}+N_{a_1(\underline{q}/2+\underline{k}),p_1}\right)\;+\;a_1\,\left(\frac{1}{2}+N_{a_2(\underline{q}/2-\underline{k}),p_2}\right)\,\right] \tag{23}$$

The above first-order differential equation can be reduced to an algebraic equation by Fourier transforming. The Fourier transform of the two-phonon propagator is defined by

$$D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}':\omega)\;=\;\int_{-\infty}^{\infty}\,D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k},t;\underline{k}',0)\;e^{-i\omega t}\;dt \tag{24}$$

Fourier transformation of the equation of motion results in the non-interacting propagator being given by

$$D_{0\,\sigma,\sigma';\underline{q}}^{(\alpha),(\beta)}(\underline{k};\underline{k}':\omega)\;=\;\Delta_{\underline{k}-\underline{k}'}\,\delta^{\alpha,\beta}_{\sigma,\sigma'}\,\frac{\left[\,a_2\,\left(\frac{1}{2}+N_{a_1(\underline{q}/2+\underline{k}),p_1}\right)\;+\;a_1\,\left(\frac{1}{2}+N_{a_2(\underline{q}/2-\underline{k}),p_2}\right)\,\right]}{\left(\,\hbar\,\omega\;-\;a_1\,\hbar\,\omega_{a_1(\frac{q}{2}+\underline{k}),p_1}\;-\;a_2\,\hbar\,\omega_{a_2(\frac{q}{2}-\underline{k}),p_2}\right)} \tag{25}$$

Thus, the non-interacting propagator is diagonal in the pairs of ordered indices α and β, as well as in the ordered pairs of polarization indices σ and σ'. Furthermore, the Bloch

wave vectors $\underline{k}$ and $\underline{k}'$ are conserved, modulo a reciprocal lattice vector. To emphasize the diagonal nature of the non-interacting propagator we shall introduce the notation

$$D_{0\sigma,\sigma';\underline{q}}^{(\alpha),(\beta)}(\underline{k};\underline{k}':\omega) \;=\; \Delta_{\underline{k}-\underline{k}'}\;\delta_{\sigma,\sigma'}^{\alpha,\beta}\;D_{\sigma,\underline{q}}^{(0)(\alpha)}(\underline{k}:\omega) \tag{26}$$

The interaction produces an additional term in the equation of motion, which is expressed as

$$-\;\frac{i}{\hbar}\;<\;|\;\hat{T}\;\Big[\;\hat{A}_{\sigma;\underline{q}}^{(\alpha)}(\underline{k},t)\;,\;\hat{H}_{int}(t)\;\Big]\;\hat{A}_{\sigma';\underline{q}}^{(\beta)\dagger}(\underline{k}',0)\;|\;> \tag{27}$$

The equal-time commutator involves a weighted sum over terms of the form

$$\Big[\;a^{a_1}_{a_1(\frac{q}{2}+\underline{k}),p_1}\;a^{a_2}_{a_2(\frac{q}{2}-\underline{k}),p_2}\;,\;\prod_{n=1}^{2}\;(a^{\dagger}_{\underline{k}_n,r_1}+a_{-\underline{k}_n,r_1})\;\prod_{m=3}^{4}\;(a^{\dagger}_{\underline{k}_m,r_2}+a_{-\underline{k}_m,r_2})\;\Big] \tag{28}$$

and produces terms which are fourth-order in the phonon operators. The equations of motion can be truncated by replacing the product of two of the four phonon operators by their expectation values. This step results in the Ladder Approximation, which is expected to be exact if there are only two phonons present in the system and if the number of phonons is conserved. As was shown by Agranovitch [38], if one projects the excitations of the system from the full Hilbert space onto a manifold of states with a fixed number of phonons, the excitations can be found without further approximation. However, it has also been shown [39] that a simple projection onto the space of fixed number of phonons leads to a violation of Goldstone's theorem [37]. Here we show that the phonon excitations obey Goldstone's theorem, if the space of fixed phonons is supplemented by two-phonon creation and two-phonon annihilation processes. Furthermore, the equations of motion reduce to the Ladder Approximation in an appropriately extended manifold of states.

On taking an expectation values of all possible pairs of phonon operators and using the identities

$$\begin{aligned}
&<\;a^{a}_{a(\underline{q}),p}\;(a^{\dagger}_{\underline{k},r}+a_{-\underline{k},r})\;>=\Delta_{\underline{q}+\underline{k}}\;\delta_{p,r}\;(\;N_{\underline{q},p}\;+\frac{1}{2}\;-\;\frac{a}{2}\;)\\
&<\;(a^{\dagger}_{\underline{k},r}+a_{-\underline{k},r})\;a^{a}_{a(\underline{q}),p}\;>=\Delta_{\underline{q}+\underline{k}}\;\delta_{p,r}\;(\;N_{\underline{q},p}\;+\frac{1}{2}\;+\;\frac{a}{2}\;)
\end{aligned} \tag{29}$$

one finds two types of contributions. The contributions in which the phonon occupation numbers don't depend on the center of mass momentum $\underline{q}$ produce corrections to the non-interacting phonon frequencies

$$\hbar\;\delta\omega_{\underline{q}}=\frac{5}{36}\;\Big(\frac{\omega_{\underline{q}}}{\omega_0}\Big)\;\frac{I_4}{N}\;\sum_{\underline{k}}\;\Big(\frac{\omega_{\underline{k}}}{\omega_0}\Big)\;(\;2\;N_{\underline{k}}\;+\;1\;) \tag{30}$$

The identification with a change in phonon frequencies can be seen through the effect of this truncation applied directly to the interaction. The resulting contribution can be considered as yielding a temperature-dependent renormalization of the harmonic spring constant K_2 [42] and obeys Goldstone's theorem [37]. Hence, these terms can be eliminated by re-defining the transformation of eqn.(9) which relates the lattice displacement operators to the phonon operators. This transformation produces the renormalization $\omega_{\underline{q}} = \omega_{\underline{q}}^{harm} + \delta\omega_{\underline{q}}$ in all expressions. Thus the transformation completely eliminates the anomalous terms in the one-phonon Green's function, at the same level of approximation. Henceforth, we shall ignore these frequency shifts and focus our attention on the terms where the occupation numbers depend on the center of mass momentum.

The other type of terms contain occupation numbers that depend on the center of mass momentum $\underline{q}$. These terms can be combined to yield

$$\begin{aligned}
&\frac{I_4}{3!\ N}\ \delta_{p_1,p_2}\ \sum_{\underline{\delta}}\ F^{\underline{\delta}\ *}_{\frac{\underline{q}}{2}+\underline{k}}\ F^{\underline{\delta}\ *}_{\frac{\underline{q}}{2}-\underline{k}}\Big[\ -a_2(N_{\frac{\underline{q}}{2}+\underline{k},p_1}+\frac{1}{2})-a_1(N_{\frac{\underline{q}}{2}-\underline{k},p_2}+\frac{1}{2})\Big]\\
&\times\sum_{\underline{k}_1}\ F^{\underline{\delta}}_{\frac{\underline{q}}{2}+\underline{k}_1}\ F^{\underline{\delta}}_{\frac{\underline{q}}{2}-\underline{k}_1}\ \sum_{\gamma,r}\ D^{(\gamma),(\beta)}_{(r,r),\sigma';\underline{q}}(\underline{k}_1;\underline{k}':\omega)\\
&+\frac{2\ I_4}{3!\ N}\ \sum_{\underline{\delta}}\ F^{\underline{\delta}\ *}_{\frac{\underline{q}}{2}+\underline{k}}\ F^{\underline{\delta}\ *}_{\frac{\underline{q}}{2}-\underline{k}}\Big[\ -a_2(N_{\frac{\underline{q}}{2}+\underline{k},p_1}+\frac{1}{2})-a_1(N_{\frac{\underline{q}}{2}-\underline{k},p_2}+\frac{1}{2})\Big]\\
&\times\sum_{\underline{k}_1}\ F^{\underline{\delta}}_{\frac{\underline{q}}{2}+\underline{k}_1}\ F^{\underline{\delta}}_{\frac{\underline{q}}{2}-\underline{k}_1}\ \sum_{\gamma}\ D^{(\gamma),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k}_1;\underline{k}':\omega)
\end{aligned}\tag{31}$$

where the sums over $\underline{k}_1$ run over both sectors of the Brillouin zone. This leads to the Fourier transformed equation of motion reducing to

$$\begin{aligned}
D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}':\omega) = \Delta_{\underline{k}-\underline{k}'}\ \delta^{\alpha,\beta}_{\sigma,\sigma'}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\\
+\frac{I_4}{3!\ N}\ \delta_{p_1,p_2}\ \sum_{\underline{\delta}}\ F^{\underline{\delta}\ *}_{\frac{\underline{q}}{2}+\underline{k}}\ F^{\underline{\delta}\ *}_{\frac{\underline{q}}{2}-\underline{k}}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\\
\times\ \sum_{\underline{k}_1}\ F^{\underline{\delta}}_{\frac{\underline{q}}{2}+\underline{k}_1}\ F^{\underline{\delta}}_{\frac{\underline{q}}{2}-\underline{k}_1}\ \sum_{\gamma,p}\ D^{(\gamma),(\beta)}_{(p,p),\sigma';\underline{q}}(\underline{k}_1;\underline{k}':\omega)\\
+\frac{2\ I_4}{3!\ N}\ \sum_{\underline{\delta}}\ F^{\underline{\delta}\ *}_{\frac{\underline{q}}{2}+\underline{k}}\ F^{\underline{\delta}\ *}_{\frac{\underline{q}}{2}-\underline{k}}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\\
\times\ \sum_{\underline{k}_1}\ F^{\underline{\delta}}_{\frac{\underline{q}}{2}+\underline{k}_1}\ F^{\underline{\delta}}_{\frac{\underline{q}}{2}-\underline{k}_1}\ \sum_{\gamma}\ D^{(\gamma),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k}_1;\underline{k}':\omega)
\end{aligned}\tag{32}$$

The above equation represents the Ladder Approximation for the two-phonon propagator which is depicted diagramatically in fig.(2). The above set of equations form a closed set

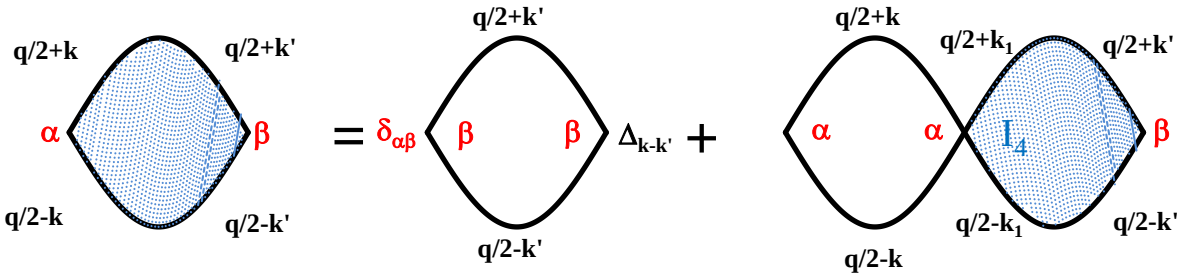

FIG. 2. (Color on line) Diagrammatic depiction of the Ladder Approximation for the two-phonon propagator. The interacting propagator is represented by the filled bubble. The unfilled bubble represents the free propagation of non-interacting phonon excitations. The vertex I_4 represents an interaction between the phonon excitations. The interaction not only scatters the phonon excitations from states with relative momentum $\underline{k}$ to states with relative momentum $\underline{k}_1$, but may also change the total number of phonons.

that can be solved by algebraic methods. The first term on the right-hand side of eqn.(32) represents the propagation of a pair of phonon excitations which do not interact. The second and third terms represent the processes in which a pair of phonons excitations interact at least once and are scattered into different momentum states. Although the initial two-phonon excitations are characterized by a specific value of α, after the first interaction process the two-phonon excitations are characterized by the different possible values of the index γ, i.e., involves states with different total numbers of (harmonic) phonons. This occurs since the interaction does not conserve the total number of phonons. In fact, the total number of phonons may change by the amounts +4, +2, 0, -2, -4 as a result of a single interaction. The Ladder Approximation for the two-phonon excitations replaces the full Hilbert space by a sub-space augmented by a space in which the total number of phonons is either increased or decreased by two.

TWO-PHONON BOUND STATES AND RESONANCES

The singularities of the two-phonon propagator yields information about the two-phonon excitations of the system. The two-phonon propagator has a dense set of simple poles associated with the non-interacting two-phonon excitations. This dense set of simple poles

yields the continuous spectrum of two-phonon excitations, where the phonons scatter from each other and then asymptotically become free. In addition to the continuum of excitations, one expects that there may be other types of singularities that represent either two-phonon resonances or two-phonon bound states. The equation for the two-phonon propagator has a non-trivial dependence on the polarization index $\sigma \equiv (p_1, p_2)$, which leads us to the expectation that the excitations will depend on σ. In particular, since phonons are vector bosons and have quasi-spin one [43], the Wigner-Eckhart theorem [44] leads one to expect that the two-phonon excitations will be characterized according to their intrinsic quasi-spin [36] which may have the values of either $S = 0$ or $S = 2$. This characterization will be deduced directly from the algebraic solution of the Ladder Approximation to the equation of motion.

In general the elements of the polarization matrix (p_1, p_2) can either be diagonal or they can be off-diagonal. Below, we shall consider the two cases separately.

x-y Polarized Excitations

In this case, where the polarizations are different $p_1 \neq p_2$, the equation of motion simplifies to

$$\begin{aligned} D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}' : \omega) = \Delta_{\underline{k}-\underline{k}'}\ \delta^{\alpha,\beta}_{\sigma,\sigma'}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k} : \omega) \\ + \frac{2\ I_4}{3\ N}\ \sum_i\ F^{i\ *}_{\frac{q}{2}+\underline{k}}\ F^{i\ *}_{\frac{q}{2}-\underline{k}}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k} : \omega) \\ \times \sum_{\underline{k}_1}\ F^{i}_{\frac{q}{2}+\underline{k}_1}\ F^{i}_{\frac{q}{2}-\underline{k}_1}\ \sum_\gamma\ D^{(\gamma),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k}_1;\underline{k}' : \omega) \end{aligned} \tag{33}$$

Hence, the off-diagonal polarization index σ is conserved. The equation can be solved by multiplying by the product $F^{j}_{\frac{q}{2}+\underline{k}}\ F^{j}_{\frac{q}{2}-\underline{k}}$, summing over α and then summing over the $\underline{k}$ values in both sectors. This procedure results in the equation

$$\Phi^{j}_{\beta,\sigma,\sigma'}(\underline{k}') \ = \ \delta_{\sigma,\sigma'}\ F^{j}_{\frac{q}{2}+\underline{k}'}\ F^{j}_{\frac{q}{2}-\underline{k}'}\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}' : \omega) \ + \ \sum_{i=1}^{3}\ \Pi^{j,i}_2(\underline{q},\omega)\ \Phi^{i}_{\beta,\sigma,\sigma'}(\underline{k}') \tag{34}$$

in which

$$\Phi^{j}_{\beta,\sigma,\sigma'}(\underline{k}') \ = \ \sum_{\underline{k},\gamma}\ F^{j}_{\frac{q}{2}+\underline{k}}\ F^{j}_{\frac{q}{2}-\underline{k}}\ D^{(\gamma),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}' : \omega) \tag{35}$$

and

$$\Pi^{j,i}_2(\underline{q},\omega) \ = \ \frac{2\ I_4}{3\ N}\ \sum_{\underline{k},\alpha}\ F^{j}_{\frac{q}{2}+\underline{k}}\ F^{j}_{\frac{q}{2}-\underline{k}}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k} : \omega)\ F^{i\ *}_{\frac{q}{2}+\underline{k}}\ F^{i\ *}_{\frac{q}{2}-\underline{k}} \tag{36}$$

The inhomogeneous term can be written in the form of a vector with components

$$\Theta^{j}_{\beta,\sigma'}(\underline{k}') \;=\; F^{j}_{\frac{q}{2}+\underline{k}'}\; F^{j}_{\frac{q}{2}-\underline{k}'}\; D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega) \tag{37}$$

The matrix equation can be solved to yield $\Phi^{j}_{\beta,\sigma}(\underline{k}')$

$$\underline{\Phi}_{\beta,\sigma,\sigma'}(\underline{k}') \;=\; \Big(\; \tilde{I} \;-\; \tilde{\Pi}_2(q,\omega)\;\Big)^{-1}\; \underline{\Theta}_{\beta,\sigma'}(\underline{k}')\; \delta_{\sigma,\sigma'} \tag{38}$$

which can then be substituted into

$$\begin{aligned} D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}':\omega) = {} & \Delta_{\underline{k}-\underline{k}'}\; \delta^{\alpha,\beta}_{\sigma,\sigma'}\; D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega) \\ & + \frac{2\; I_4}{3\; N}\; \sum_{i}\; F^{i\;*}_{\frac{q}{2}+\underline{k}}\; F^{i\;*}_{\frac{q}{2}-\underline{k}}\; D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\; \Phi^{i}_{\beta,\sigma,\sigma'}(\underline{k}') \end{aligned} \tag{39}$$

thereby yielding the two-phonon propagator as

$$\begin{aligned} D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}':\omega) = {} & \Delta_{\underline{k}-\underline{k}'}\; \delta^{\alpha,\beta}_{\sigma,\sigma'}\; D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega) \\ & + \delta_{\sigma,\sigma'}\; \sum_{i,j}\; F^{i\;*}_{\frac{q}{2}+\underline{k}}\; F^{i\;*}_{\frac{q}{2}-\underline{k}}\; D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega) \\ & \times\; \frac{2\; I_4}{3\; N}\; \Big(\; \tilde{I} \;-\; \tilde{\Pi}_2(\underline{q},\omega)\;\Big)^{-1}_{i,j} \\ & \times\; F^{j}_{\frac{q}{2}+\underline{k}'}\; F^{j}_{\frac{q}{2}-\underline{k}'}\; D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega) \end{aligned} \tag{40}$$

It should be noted that the $x-y$ polarized two-phonon propagator is diagonal in the combined polarization indices σ and σ', but $p_1 \neq p_2$.

x-x Polarized Excitations

In this case $p_1 = p_2$, so the equations of motion only involve propagators which are diagonal in the polarization indices. Due to the Kronecker delta function in the inhomogeneous term, one finds that the propagator is zero unless $p'_1 = p'_2$, so σ' must also be diagonal. On taking the trace of the equation of motion over the polarization indices σ, one obtains

$$\begin{aligned} \sum_{\sigma}\; D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}':\omega) = {} & \Delta_{\underline{k}-\underline{k}'}\; \delta^{\alpha,\beta}\; D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega) \\ & + \frac{5\; I_4}{6\; N}\; \sum_{\underline{\delta}}\; F^{\underline{\delta}\;*}_{\frac{q}{2}+\underline{k}}\; F^{\underline{\delta}\;*}_{\frac{q}{2}-\underline{k}}\; D^{(0)(\alpha)}_{\underline{q}}(\underline{k}:\omega) \\ & \times\; \sum_{\underline{k}_1}\; F^{\underline{\delta}}_{\frac{q}{2}+\underline{k}_1}\; F^{\underline{\delta}}_{\frac{q}{2}-\underline{k}_1}\; \sum_{\gamma,\sigma}\; D^{(\gamma),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k}_1;\underline{k}':\omega) \end{aligned} \tag{41}$$

where we have used the fact that, because the polarizations are degenerate, the value of $D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)$ is actually independent of σ. The sum over $\underline{\delta}$ can be written as twice the sum over three orthogonal axes

$$\sum_{\sigma} D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}':\omega) = \Delta_{\underline{k}-\underline{k}'}\ \delta^{\alpha,\beta}\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega)$$
$$+ \frac{5\ I_4}{3\ N}\ \sum_{i=1}^{3}\ F^{i\ *}_{\frac{q}{2}+\underline{k}}\ F^{i\ *}_{\frac{q}{2}-\underline{k}}\ D^{(0)(\alpha)}_{\underline{q}}(\underline{k}:\omega)$$
$$\times \sum_{\underline{k}_1} F^{i}_{\frac{q}{2}+\underline{k}_1}\ F^{i}_{\frac{q}{2}-\underline{k}_1}\ \sum_{\gamma,\sigma}\ D^{(\gamma),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k}_1;\underline{k}':\omega) \qquad (42)$$

where the form factors only depend on the i-th coordinate of the wave vectors k_i. This form of the anharmonic interaction is recognized as being the sum over three separable interactions, one for each Cartesian axis. On multiplying the equations by $F^{j}_{\frac{q}{2}+\underline{k}}\ F^{j}_{\frac{q}{2}-\underline{k}}$, summing over α and then summing over $\underline{k}$, one arrives at the equation

$$\Phi^{j}_{\beta,\sigma'}(\underline{k}') \ = \ F^{j}_{\frac{q}{2}+\underline{k}'}\ F^{j}_{\frac{q}{2}-\underline{k}'}\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega)\ +\ \sum_{i=1}^{3}\ \Pi^{j,i}_{0}(\underline{q},\omega)\ \Phi^{i}_{\beta,\sigma'}(\underline{k}') \qquad (43)$$

in which

$$\Phi^{j}_{\beta,\sigma'}(\underline{k}') \ = \ \sum_{\underline{k},\gamma,\sigma}\ F^{j}_{\frac{q}{2}+\underline{k}}\ F^{j}_{\frac{q}{2}-\underline{k}}\ D^{(\gamma),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}':\omega) \qquad (44)$$

and

$$\Pi^{j,i}_{0}(\underline{q},\omega) \ = \ \frac{5\ I_4}{3\ N}\ \sum_{\underline{k},\alpha}\ F^{j}_{\frac{q}{2}+\underline{k}}\ F^{j}_{\frac{q}{2}-\underline{k}}\ D^{(0)(\alpha)}_{\underline{q}}(\underline{k}:\omega)\ F^{i\ *}_{\frac{q}{2}+\underline{k}}\ F^{i\ *}_{\frac{q}{2}-\underline{k}} \qquad (45)$$

In the above, the sums over $\underline{k}$ run over both sectors and the sum over polarization index σ only runs over the diagonal elements. Equation(43) is recognized as a three by three matrix equation of the form

$$\Big(\ \tilde{I}\ -\ \tilde{\Pi}_0(q,\omega)\ \Big)\ \underline{\Phi}_{\beta,\sigma'}(\underline{k}') \ = \ \underline{\Theta}_{\beta,\sigma'}(\underline{k}') \qquad (46)$$

where the components of the vector $\underline{\Theta}$ are given by

$$\Theta^{j}_{\beta,\sigma'}(\underline{k}') \ = \ F^{j}_{\frac{q}{2}+\underline{k}'}\ F^{j}_{\frac{q}{2}-\underline{k}'}\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega) \qquad (47)$$

The above matrix equation can be inverted

$$\underline{\Phi}_{\beta,\sigma'}(\underline{k}') \ = \ \Big(\ \tilde{I}\ -\ \tilde{\Pi}_0(q,\omega)\ \Big)^{-1}\ \underline{\Theta}_{\beta,\sigma'}(\underline{k}') \qquad (48)$$

and substituted into eqn.(41)

$$\sum_{\sigma} D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}' : \omega) = \Delta_{\underline{k}-\underline{k}'}\ \delta^{\alpha,\beta}\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}' : \omega) + \frac{5\ I_4}{3\ N}\ \sum_{i}\ F^{i\ *}_{\frac{q}{2}+\underline{k}}\ F^{i\ *}_{\frac{q}{2}-\underline{k}}\ D^{(0)(\alpha)}_{\underline{q}}(\underline{k} : \omega)\ \Phi^{i}_{\beta,\sigma'}(\underline{k}') \tag{49}$$

to yield an expression for $\sum_{\sigma}\ D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}' : \omega)$.

On substituting the above expression for the polarization trace of the two-phonon propagator back into the equation for the xx-polarized two-phonon propagator, one obtains

$$\begin{aligned} D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}' : \omega) = {} & \Delta_{\underline{k}-\underline{k}'}\ \delta^{\alpha,\beta}_{\sigma,\sigma'}\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}' : \omega) \\ & + \frac{I_4}{3\ N}\ \sum_{i,j}\ F^{i\ *}_{\frac{q}{2}+\underline{k}}\ F^{i\ *}_{\frac{q}{2}-\underline{k}}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k} : \omega)\ \Big(\ \tilde{I}\ -\ \tilde{\Pi}_0(\underline{q},\omega)\ \Big)^{-1}_{i,j} \\ & \times\ F^{j}_{\frac{q}{2}+\underline{k}'}\ F^{j}_{\frac{q}{2}-\underline{k}'}\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}' : \omega) \\ & + \frac{2\ I_4}{3\ N}\ \sum_{i}\ F^{i\ *}_{\frac{q}{2}+\underline{k}}\ F^{i\ *}_{\frac{q}{2}-\underline{k}}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k} : \omega) \\ & \times\ \sum_{\underline{k}_1}\ F^{i}_{\frac{q}{2}+\underline{k}_1}\ F^{i}_{\frac{q}{2}-\underline{k}_1}\ \sum_{\gamma}\ D^{(\gamma),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k}_1;\underline{k}' : \omega) \end{aligned} \tag{50}$$

In the above expression, the second inhomogeneous term originates from the modes corresponding to the trace over the polarizations. This set of equations can be solved leading to the final result

$$\begin{aligned} D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}' : \omega) = {} & \Delta_{\underline{k}-\underline{k}'}\ \delta^{\alpha,\beta}_{\sigma,\sigma'}\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}' : \omega) \\ & + \sum_{i,j}\ F^{i\ *}_{\frac{q}{2}+\underline{k}}\ F^{i\ *}_{\frac{q}{2}-\underline{k}}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k} : \omega) \\ & \times\ \Big[\ \Big(\delta_{\sigma,\sigma'} - \frac{1}{3}\Big)\ \frac{2\ I_4}{3\ N}\ \Big(\ \tilde{I}\ -\ \tilde{\Pi}_2(\underline{q},\omega)\ \Big)^{-1}_{i,j} \\ & + \Big(\frac{1}{3}\Big)\ \frac{5\ I_4}{3\ N}\ \Big(\ \tilde{I}\ -\ \tilde{\Pi}_0(\underline{q},\omega)\ \Big)^{-1}_{i,j}\ \Big] \\ & \times\ F^{j}_{\frac{q}{2}+\underline{k}'}\ F^{j}_{\frac{q}{2}-\underline{k}'}\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}' : \omega) \end{aligned} \tag{51}$$

where, as before, the matrix elements of $\tilde{\Pi}_2(\underline{q},\omega)$ are given in eqn.(36). In the above, it should be noted that the joint polarizations σ and σ' are both diagonal [i.e. $\sigma = (p,p)$ and $\sigma' = (p',p')$].

EXISTENCE AND DISPERSION RELATIONS OF ILMS

The degeneracy of the phonon modes with respect to the polarization is a consequence of the space group symmetry of the vector displacement field $\underline{u}_{\underline{q}}$. The application of Noether's theorem with the discrete space group symmetry operations leads to conservation laws, such as conservation of quasi-momentum (i.e. crystal or Bloch momentum), quasi-orbital angular momentum and the existence of intrinsic quasi-spin [43]. In the above model, the phonons with the different polarizations are degenerate which leads to their having an intrinsic quasi-spin of $S = 1$ similar to the case of phonon excitations in the continuum limit description of an isotropic material [40, 41]. Hence, the phonons excitations can be expressed as linear combinations of the excitations with different polarization vectors, or equivalently as a linear combination of excitations with different values of S^z. Since the ILM excitations correspond to bound states of multiple numbers of phonons it is expected that, when the ILMs exist, they should also have integer intrinsic quasi-spins. Indeed, the lowest-energy excitations of this hierarchy have polarization indices that form a symmetric second-rank tensor [44]. Furthermore, all the excitations are degenerate since they all are determined from the solution of $\det[\ \tilde{I} - \tilde{\Pi}_2(\underline{q},\omega)\ ] = 0$, with the exception of the excitation which corresponds to the trace of the polarization matrix. This last excitation has an energy which is determined from the condition $\det[\ \tilde{I} - \tilde{\Pi}_0(\underline{q},\omega)\ ] = 0$. Thus, the five degenerate excitations corresponding to the traceless symmetric second-rank tensor may be identified as excitations with quasi-spin $S = 2$. These excitations can be expressed as linear combinations of the five eigenfunctions of S^z. The remaining non-degenerate excitation corresponds to a quasi-spin of $S = 0$. The above conclusions can also be deduced [36] by the application of the Wigner-Eckhart theorem [44]. Since $\tilde{\Pi}_2(\underline{q},\omega)$ and $\tilde{\Pi}_0(\underline{q},\omega)$ have the same structure in that they only differ by the strength of the interaction in their definitions, they can be treated simultaneously. Below, we shall examine the energy spectrum and the spatial characters as exhibited through the $\underline{q}$ dependence of these excitations along various high-symmetry lines. We shall display formulae appropriate to the $S = 2$ excitations. The corresponding formula for $S = 0$ are obtained by substituting $\tilde{\Pi}_0(\underline{q},\omega)$ for $\tilde{\Pi}_2(\underline{q},\omega)$ and replacing the explicit factor of $\frac{2}{3}$ in front of the anharmonic interaction by $\frac{5}{3}$.

Momentum Transfers (q,q,q)

The body diagonal of the Brillouin zone is a three-fold rotation axis. Thus, for momentum transfers along the body diagonal of the Brillouin zone $q_x = q_y = q_z = q$, the matrix $\Pi_2^{i,j}(\underline{q},\omega)$ is invariant under all the permutations of the summation indices k_x, k_y and k_z. Hence, $\Pi_2^{i,j}(\underline{q},\omega)$ has the symmetry

$$\Pi_2^{1,2}(\underline{q},\omega) \;=\; \Pi_2^{1,3}(\underline{q},\omega) \;=\; \Pi_2^{2,3}(\underline{q},\omega) \tag{52}$$

and

$$\Pi_2^{1,1}(\underline{q},\omega) \;=\; \Pi_2^{2,2}(\underline{q},\omega) \;=\; \Pi_2^{3,3}(\underline{q},\omega) \tag{53}$$

Therefore, the expression for the $x-y$ two-phonon propagator simplifies to

$$\begin{aligned}
D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}':\omega) = {} & \Delta_{\underline{k}-\underline{k}'}\;\delta^{\alpha,\beta}_{\sigma,\sigma'}\;D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega) \\
& + \delta_{\sigma,\sigma'}\;\frac{2\;I_3}{3\;N}\;D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\;\frac{1}{1-\Pi_2^{1,1}(\underline{q},\omega)-2\Pi_2^{1,2}(\underline{q},\omega)}\;D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega) \\
& \times\Big[\;\frac{1}{3}\;\sum_{i,j}\;F^{i\;*}_{\frac{q}{2}+\underline{k}}\;F^{i\;*}_{\frac{q}{2}-\underline{k}}\;F^{j}_{\frac{q}{2}+\underline{k}'}\;F^{j}_{\frac{q}{2}-\underline{k}'}\;\Big] \\
& + \delta_{\sigma,\sigma'}\;\frac{2\;I_3}{3\;N}\;D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\frac{1}{1-\Pi_2^{1,1}(\underline{q},\omega)+\Pi_2^{1,2}(\underline{q},\omega)}\;D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega) \\
& \times\Big[\sum_{i}\;F^{i\;*}_{\frac{q}{2}+\underline{k}}F^{i\;*}_{\frac{q}{2}-\underline{k}}F^{i}_{\frac{q}{2}+\underline{k}'}F^{i}_{\frac{q}{2}-\underline{k}'}\;-\;\frac{1}{3}\;\sum_{i,j}F^{i\;*}_{\frac{q}{2}+\underline{k}}F^{i\;*}_{\frac{q}{2}-\underline{k}}F^{j}_{\frac{q}{2}+\underline{k}'}F^{j}_{\frac{q}{2}-\underline{k}'}\Big]
\end{aligned} \tag{54}$$

The poles of the two-phonon propagator yield the two-phonon excitation spectrum of the system with momentum (q,q,q). In addition to the continuum of excitations associated with the poles of the non-interacting propagators, there are poles associated with the zeros of the denominators of the second term

$$1\;-\;\Pi_2^{1,1}(\underline{q},\omega)\;-\;2\;\Pi_2^{1,2}(\underline{q},\omega) \tag{55}$$

and the third term

$$1\;-\;\Pi_2^{1,1}(\underline{q},\omega)\;+\;\Pi_2^{1,2}(\underline{q},\omega) \tag{56}$$

The frequencies at which the denominators become zero are the frequencies of the collective excitations of the system. If the frequencies fall within the continuum of two-phonon excitations (which represents the scattering states) such that

$$\Im m\;\sum_{\underline{k},\alpha}\;D^{(0),(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\;\neq\;0 \tag{57}$$

then the collective are two-phonon resonances. Otherwise, if the frequencies are outside the continuum, so that

$$\Im m \sum_{\underline{k},\alpha} D^{(0),(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega) \;=\; 0 \tag{58}$$

then the collective excitations are two-phonon bound states. The weight of each of the two modes can be found by Taylor expanding about the frequency of each mode to find the residue. The frequency of the $S = 2$ mode originating from the divergence of the second term in eqn.(54) is given by the frequencies at which the denominator expressed in eqn.(55) vanishes. The frequencies are given by the solutions of

$$1 \;=\; \frac{2\ I_4}{3\ N} \sum_{\underline{k},\alpha} \Big[\ \frac{1}{3} \sum_{i,j} \ F^j_{\frac{q}{2}+\underline{k}} \ F^j_{\frac{q}{2}-\underline{k}} \ F^{i\ *}_{\frac{q}{2}+\underline{k}} \ F^{i\ *}_{\frac{q}{2}-\underline{k}} \ \Big] D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega) \tag{59}$$

Since the product of the form factors in the denominator of the the second term in the propagator is the same as in the numerator, which we will denote by M, the residue at the pole is given by the ratio of

$$- \ \frac{\sum_{\underline{k},\alpha} \ M \ \Big[\ D^{(0),(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega) \ \Big]^2}{\frac{\partial}{\partial\omega} \ \Big[\ \sum_{\underline{k},\alpha} \ M \ D^{(0),(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega) \ \Big]} \tag{60}$$

so one finds (at $T = 0$) that the pole has residue of strength of approximately one. It can be seen that the expression for the form factor in the denominator is similar to the factor in the numerator. Thus, any bound states associated with a zero of this denominator should represent a non-degenerate mode.

We have evaluated the right-hand side of eqn.(59) for the temperatures $\frac{k_BT}{\hbar\omega_0} = 0$, $\frac{k_BT}{\hbar\omega_0} = 2.5$ and $\frac{k_BT}{\hbar\omega_0} = 5$ with the q values (π/a), $(39\pi/40a)$, $(19\pi/20a)$, $(7\pi/8a)$, $(3\pi/4a)$ and with $\frac{I_4}{2\sqrt{6}\hbar\omega_0} = 0.145$. The theoretical results are shown in fig.(3). The figure shows the results for a range of ω near the upper edge of the two-phonon continuum. The characteristic frequencies appearing in the plots are related to the van-Hove singularities in the density of two-phonon excitations with specified center of mass momentum $\underline{q}$. The dispersion of the van-Hove singularities is shown in the upper panel of fig.(4). The relation to the van-Hove singularities is expected since the imaginary part of $\Pi^{1,1}_2(\underline{q},\omega) \ + \ 2\ \Pi^{1,2}_2(\underline{q},\omega)$ merely differs from two-phonon density of states due to the presence of the form factors and since the real and imaginary parts of the functions are related via a Hilbert transform. For each temperature, the maxima are found at frequencies at the top of the two-phonon continuum

and the maxima become more pronounced and diverge as q approaches (π/a). Furthermore, the right-hand side increases with increasing temperatures due to the presence of the Bose-Einstein distribution functions present in $D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k} : \omega)$. Therefore, one concludes from eqn.(59) that the minimal value of I_4 required to produce a non-degenerate and symmetric ILM at a fixed q value is smaller at an elevated temperature than the minimal value required to produce the ILM at $T = 0$.

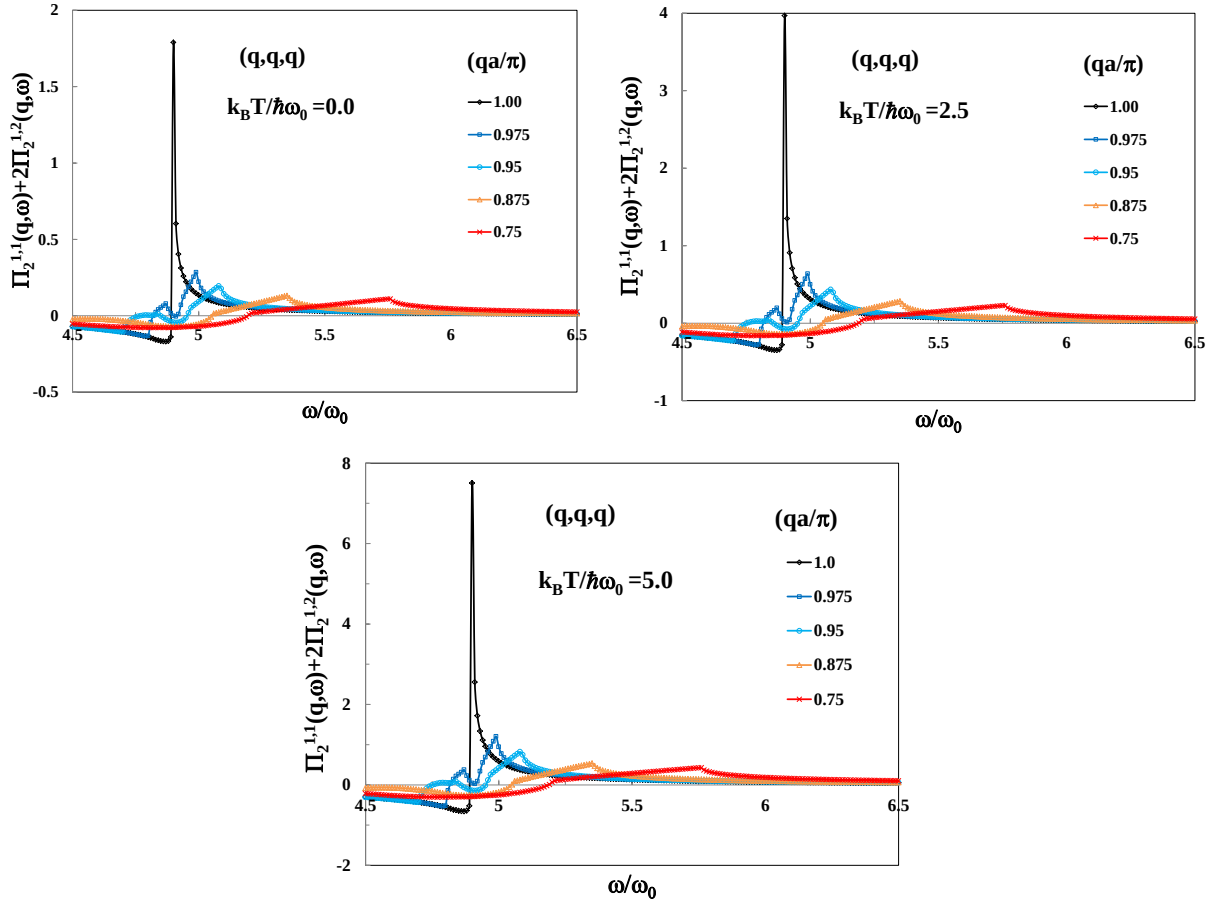

FIG. 3. (Color on line) The frequency dependence of the real part of $\Pi_2^{1,1}(\underline{q},\omega) + 2\,\Pi_2^{1,2}(\underline{q},\omega)$ for various temperatures and the $\underline{q}$ values along the body-diagonal given by (π/a), $(39\pi/40a)$, $(19\pi/20a)$, $(7\pi/8a)$, $(3\pi/4a)$. (Top Left Panel) The results evaluated at $\frac{k_BT}{\hbar\omega_0} = 0$. (Top Right Panel) The results found with $\frac{k_BT}{\hbar\omega_0} = 2.5$. (Bottom Panel) The numerical results evaluated with $\frac{k_BT}{\hbar\omega_0} = 5$.

The dispersion relation for the non-degenerate (symmetric) ILM with $S = 0$ is found from the zeros of eqn.(55) by simply replacing the Π_2's by Π_0's. The value of the interaction

I_4 chosen as $\frac{I_4}{2\sqrt{6}\ \hbar\omega_0} = 0.145$, but for this value of I_4 the ILMs that form at $T = 0$ only occur in the immediate vicinity of the point $q = (\pi/a)$ and are only slightly separated from the van-Hove singularity at the top edge of the two-phonon continuum. The top right panel in fig.(4) shows the extrema of the two-phonon continuum at which one finds the van-Hove singularities in the (non-interacting) two-phonon density of states. The continuum is bounded from above by the van-Hove singularity denoted by the full blue line, and is bounded from below by the phonon frequency (full red line). The positions of the other van-Hove singularities are denoted by the dotted lines.

At the higher temperatures of either $\frac{k_B T}{\hbar\omega_0} = 2.5$ or $\frac{k_B T}{\hbar\omega_0} = 5$, one finds that the $S = 0$ and $S = 2$ ILM dispersion relations have spread over significantly larger ranges of q. The branches of the dispersion relations are shown in the bottom left and right panels of fig.(4), for both the non-degenerate and the doubly-degenerate modes (which originate from the third term of the two-phonon propagator). The non-degenerate mode has a higher energy than the doubly-degenerate modes. Also, as q is reduced, the non-degenerate ILM approaches the upper edge of the two-phonon continuum in an almost asymptotic manner but terminates on it. Our results show that at a fixed value of q and with increasing temperature, first the $S = 0$ ILMs form and then the $S = 2$ ILMs form at a higher temperature. This is in qualitative agreement with the first set of experiments of Manley *et al.* [19] which only found ILMs at elevated temperatures.

We now examine the collective modes which originate from the third term of the two-phonon propagator given in eqn.(54). The frequencies of the (S=2) collective modes are given by the frequencies at which the expression in eqn.(56) vanishes. The frequencies are given by the solutions of

$$1 = \frac{I_4}{3\ N} \sum_{\underline{k},\alpha} \Big[\sum_i F^{i\ *}_{\frac{\underline{q}}{2}+\underline{k}} F^{i\ *}_{\frac{\underline{q}}{2}-\underline{k}} F^{i}_{\frac{\underline{q}}{2}+\underline{k}} F^{i}_{\frac{\underline{q}}{2}-\underline{k}} - \frac{1}{3} \sum_{i,j} F^{i\ *}_{\frac{\underline{q}}{2}+\underline{k}} F^{i\ *}_{\frac{\underline{q}}{2}-\underline{k}} F^{j}_{\frac{\underline{q}}{2}+\underline{k}} F^{j}_{\frac{\underline{q}}{2}-\underline{k}} \Big] D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k} : \omega) \quad (61)$$

It should be noted that the products of form factors in the denominator of the third term in the two-phonon propagator differ from the corresponding product in the numerator by a factor of two. Therefore, any mode originating from the zero of the denominator of the third term is expected to be doubly-degenerate. This is in accord with the expectations for the excitations of a simple cubic system for wave vectors on a high-symmetry axis with three-fold symmetry [45].

We have performed the same steps that were undertaken in the investigation of the

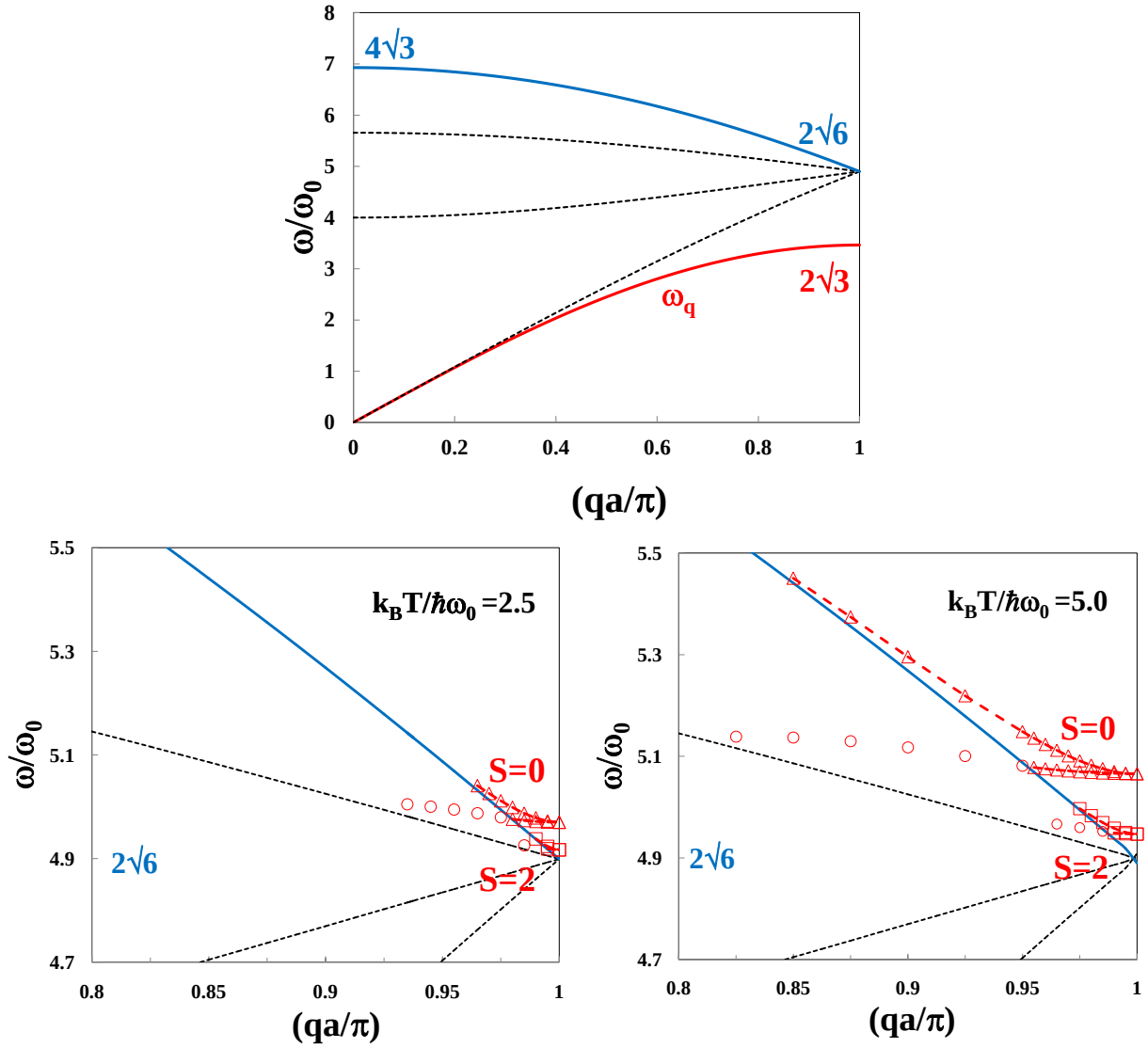

FIG. 4. (Color on line) The positions of the van-Hove singularities for $\underline{q}$ values along the body diagonal of the Brillouin zone (q, q, q) (Top Panel). The lower two panels show the dispersion relations of the ILMs along the body diagonal of the Brillouin zone for the non-degenerate mode. The dispersion relations for $S = 0$ are marked by open triangles while the $S = 2$ dispersion relations are denoted by open squares. The branches of non-degenerate modes have higher energies than the doubly-degenerate modes. (Bottom Left Panel) The dispersion relations calculated at $\frac{k_B T}{\hbar\omega_0} = 2.5$. (Bottom Right Panel). The dispersion relations evaluated with $\frac{k_B T}{\hbar\omega_0} = 5$.

degenerate mode described by eqn.(61). The real part of $\Pi_2^{1,1}(\underline{q}, \omega) - \Pi_2^{1,2}(\underline{q}, \omega)$ has been evaluated at the same set of temperatures and q values as in fig.(3), i.e. $\frac{k_B T}{\hbar\omega_0} = 0$, $\frac{k_B T}{\hbar\omega_0} = 2.5$,

and $\frac{k_BT}{\hbar\omega_0} = 5$. Figure(5) shows the results for various q values given by (π/a), $(39\pi/40a)$, $(19\pi/20a)$, $(7\pi/8a)$, $(3\pi/4a)$. It should be noted that the van-Hove singularity at the top of the two-phonon continuum does not yield a peak, since the combination of form factors vanish at the position of the highest van-Hove singularity. Hence, the top most peak is generally at an energy below the upper edge of the continuum. It can be seen that the magnitude of the function increases as the temperature increases. Furthermore, the peak approaches the upper edge of the continuum and diverges when q approaches (π/a).

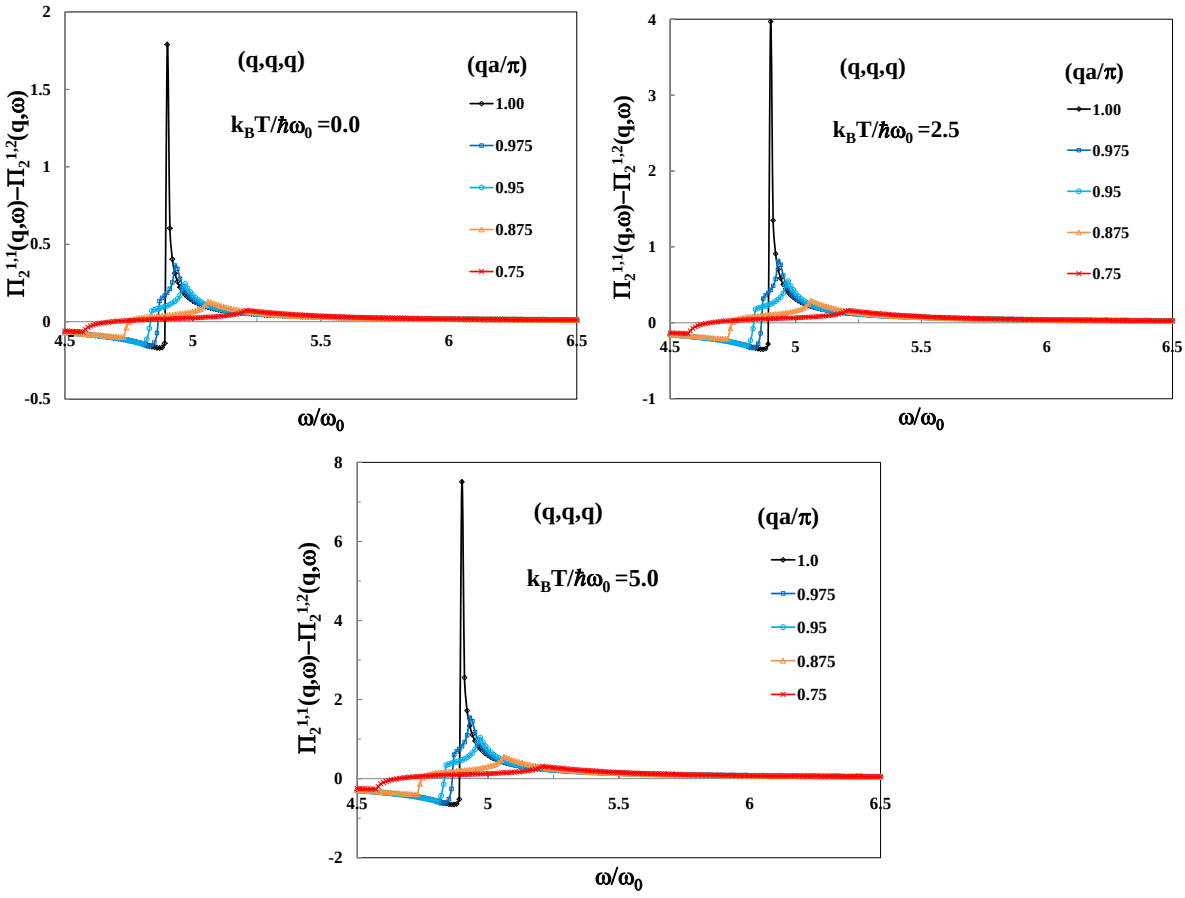

FIG. 5. (Color on line) The ω dependence of the real part of $\Pi_2^{1,1}(\underline{q},\omega) - \Pi_2^{1,2}(\underline{q},\omega)$ for $\underline{q}$ along (q,q,q) with q values given by (π/a), $(39\pi/40a)$, $(19\pi/20a)$, $(7\pi/8a)$, $(3\pi/4a)$. (Top Left Panel) The real part evaluated at $\frac{k_BT}{\hbar\omega_0} = 0$. (Top Right Panel) The real part of the correlation function evaluated at the temperature $\frac{k_BT}{\hbar\omega_0} = 2.5$. (Bottom Panel) The real part evaluated with $\frac{k_BT}{\hbar\omega_0} = 5$.

The ILM dispersion relation for the degenerate mode with $\underline{q}$ along the (q,q,q) direction has been evaluated with the same value of the interaction strength $\frac{I_4}{2\sqrt{6}\hbar\omega_0} = 0.145$ and for the same set of temperatures as in fig.(3). At zero temperature, the branches of $S = 0$

and $S = 2$ ILMs are ignorable since they only occur for an insignificant range of q values. In contrast to the case of zero temperature, for the temperatures given by $\frac{k_B T}{\hbar\omega_0} = 2.5$ and $\frac{k_B T}{\hbar\omega_0} = 5$ the branches of $S = 0$ and $S = 2$ ILMs are spread over significant ranges of q. The results are shown in fig.(4). Generally, the doubly-degenerate modes have lower energies than the non-degenerate modes. In contrast to the dispersion of the non-degenerate mode, the dispersion relation for the doubly-degenerate ILM enters into the two-phonon continuum. For q values where the dispersion relation extends inside the continuum, the ILM exists as a narrow resonance.

The branch of degenerate ILMs has the same energy as the non-degenerate branch at the point R where $q = (\pi/a)$ since the off-diagonal matrix elements of $\Pi_S^{i,j}(\underline{q}, \omega)$ vanish at that point. The vanishing of the off-diagonal matrix elements and the three-fold degeneracy at R is expected since the point R is invariant under the entire cubic symmetry group, as is the Γ point. The branch of doubly-degenerate modes separates from the non-degenerate mode and drops to lower energies when q moves away from (π/a). As shall be seen when we examine the ILM wave functions, in the continuum limit the non-degenerate ILM has a symmetric wave function $(x^2 + y^2 + z^2)$ without nodes but the doubly-degenerate modes have nodes in their wave functions. These textures of the wave functions are dictated by specific combinations of the form factors. The existence of the nodes implies that the non-degenerate modes have higher kinetic energies and, therefore, have smaller "binding energies" as measured from the upper boundary of the two-phonon continuum. Thus, the existence of nodes accounts for the difference in the energies of the two types of ILMs.

Our results for both the degenerate and non-degenerate modes, show that the ILMs form preferentially for $\underline{q}$ values near the corner of the Brillouin zone, and they split off from the upper edge of the two-phonon continuum. The reason for ILMs to preferentially form near this $\underline{q}$ value can be traced back to the convergence of several van-Hove singularities in the (non-interacting) two-phonon density of states at the upper-edge of the two-phonon continuum. This confluence occurs when the center of mass momentum $\underline{q}$ approaches the zone boundary. For the quantized lattice, the binding energy is defined as the energy splitting of the ILM from the top of the two-phonon continuum. This is to be contrasted with the definition used in classical theories where the "binding energy" is defined to be the difference in the energies of the ILM and the largest acoustic phonon. Hence, we see that when an quantum ILM first forms, it has a sizeable energy separation from the largest acoustic phonon

frequency. Plus, as $\underline{q}$ moves away from the corner of the Brillouin zone, the binding energies of the ILMs decrease and ILMs' energies approach the upper boundary of the two-phonon continuum. However, only the degenerate ILMs continue beyond the boundary where they exist as resonances.

Momentum Transfers (q,q,0)

The axis $q_z = 0$, $q_x = q_y = q$ is a two-fold rotation axis. Thus, for momentum transfers along the $(q, q, 0)$ direction, the symmetry of the function $\Pi_2^{i,j}(\underline{q}, \omega)$ is reduced from the (q, q, q) case considered previously. The symmetry of $\Pi_2^{i,j}(\underline{q}, \omega)$ results in

$$\Pi_2^{1,2}(\underline{q}, \omega) \; \neq \; \Pi_2^{1,3}(\underline{q}, \omega) \; = \; \Pi_2^{2,3}(\underline{q}, \omega) \tag{62}$$

and

$$\Pi_2^{1,1}(\underline{q}, \omega) \; = \; \Pi_2^{2,2}(\underline{q}, \omega) \; \neq \; \Pi_2^{3,3}(\underline{q}, \omega) \tag{63}$$

Due to this symmetry, the inverse matrix becomes

$$\begin{aligned}
\Big(\; \tilde{I} \; - \; \tilde{\Pi}_2(\underline{q}, \omega) \; \Big)^{-1} = \frac{1}{2} \; & \frac{1}{1 \; - \; \Pi_2^{1,1}(\underline{q}, \omega) \; + \; \Pi_2^{1,2}(\underline{q}, \omega)} \; \begin{pmatrix} 1 & -1 & 0 \\ -1 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \\
& + \frac{1}{2} \; \frac{1}{Det_2(\underline{q}, \omega)} \\
& \times \begin{pmatrix} 1 - \Pi_2^{3,3}(\underline{q}, \omega) & 1 - \Pi_2^{3,3}(\underline{q}, \omega) & 2 \; \Pi_2^{1,3}(\underline{q}, \omega) \\ 1 - \Pi_2^{3,3}(\underline{q}, \omega) & 1 - \Pi_2^{3,3}(\underline{q}, \omega) & 2 \; \Pi_2^{1,3}(\underline{q}, \omega) \\ 2 \; \Pi_2^{3,1}(\underline{q}, \omega) & 2 \; \Pi_2^{3,1}(\underline{q}, \omega) & 2\Big(1 - \Pi_2^{1,1}(\underline{q}, \omega) - \Pi_2^{1,2}(\underline{q}, \omega)\Big) \end{pmatrix}
\end{aligned} \tag{64}$$

where

$$Det_2(\underline{q} : \omega) \; = \; \Big(\; 1 \; - \; \Pi_2^{3,3}(\underline{q}, \omega) \; \Big) \; \Big(\; 1 \; - \; \Pi_2^{1,1}(\underline{q}, \omega) \; - \; \Pi_2^{1,2}(\underline{q}, \omega) \; \Big) \; - \; 2 \; \Big| \; \Pi_2^{1,3}(\underline{q}, \omega) \; \Big|^2 \tag{65}$$

It should be noted that the two matrices on the right-hand side have elements (i, j) which are simply related to the transpose of the matrix formed from the derivatives of the respective denominators with respect to $\Pi^{i,j}(\underline{q}, \omega)$. Thus, the two-phonon propagator reduces to the

form

$$D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}':\omega) = \Delta_{\underline{k}-\underline{k}'}\ \delta^{\alpha,\beta}_{\sigma,\sigma'}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)$$
$$+\ \delta_{\sigma,\sigma'}\ \frac{2\ I_3}{3\ N}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\frac{1}{1-\Pi^{1,1}_2(\underline{q},\omega)+\Pi^{1,2}_2(\underline{q},\omega)}\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega)$$
$$\times\ \Big[\ \frac{1}{2}\ (\ F^{x*}_{\frac{q}{2}+\underline{k}}F^{x\ *}_{\frac{q}{2}-\underline{k}}\ -\ F^{y*}_{\frac{q}{2}+\underline{k}}F^{y\ *}_{\frac{q}{2}-\underline{k}}\ )\ (\ F^{x}_{\frac{q}{2}+\underline{k}'}F^{x}_{\frac{q}{2}-\underline{k}'}\ -\ F^{y}_{\frac{q}{2}+\underline{k}'}F^{y}_{\frac{q}{2}-\underline{k}'}\ )\ \Big]$$
$$+\ \delta_{\sigma,\sigma'}\ \frac{2\ I_3}{3\ N}\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\ \Big(\ \frac{M_2(\underline{q},\omega)}{Det_2(\underline{q},\omega)}\ \Big)\ D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega) \qquad (66)$$

where

$$M_2(\underline{q},\omega) = F^{z\ *}_{\frac{q}{2}+\underline{k}}F^{z\ *}_{\frac{q}{2}-\underline{k}}\ \Big(\ 1-\Pi^{1,1}_2(\underline{q},\omega)-\Pi^{1,2}_2(\underline{q},\omega)\ \Big)\ F^{z}_{\frac{q}{2}+\underline{k}'}F^{z}_{\frac{q}{2}-\underline{k}'}$$
$$+\ (\ F^{x\ *}_{\frac{q}{2}+\underline{k}}F^{x\ *}_{\frac{q}{2}-\underline{k}}+F^{y\ *}_{\frac{q}{2}+\underline{k}}F^{y\ *}_{\frac{q}{2}-\underline{k}}\ )\ \frac{1}{2}\ \Big(\ 1-\Pi^{3,3}_2(\underline{q},\omega)\ \Big)$$
$$\times\ (\ F^{x}_{\frac{q}{2}+\underline{k}'}F^{x}_{\frac{q}{2}-\underline{k}'}+F^{y}_{\frac{q}{2}+\underline{k}'}F^{y}_{\frac{q}{2}-\underline{k}'}\ )$$
$$+\ (F^{x\ *}_{\frac{q}{2}+\underline{k}}F^{x\ *}_{\frac{q}{2}-\underline{k}}+F^{y\ *}_{\frac{q}{2}+\underline{k}}F^{y\ *}_{\frac{q}{2}-\underline{k}})\ \Pi^{1,3}_2(\underline{q},\omega)\ (F^{z}_{\frac{q}{2}+\underline{k}'}F^{z}_{\frac{q}{2}-\underline{k}'})$$
$$+\ (F^{z\ *}_{\frac{q}{2}+\underline{k}}F^{z\ *}_{\frac{q}{2}-\underline{k}})\ \Pi^{3,1}_2(\underline{q},\omega)^*\ (F^{x}_{\frac{q}{2}+\underline{k}'}F^{x}_{\frac{q}{2}-\underline{k}'}+F^{y}_{\frac{q}{2}+\underline{k}'}F^{y}_{\frac{q}{2}-\underline{k}'}) \qquad (67)$$

Since the denominator of the second term in the two-phonon propagator

$$1\ -\ \Pi^{1,1}_2(\underline{q},\omega)\ +\ \Pi^{1,2}_2(\underline{q},\omega) \qquad (68)$$

can be re-written as

$$1-\frac{2\ I_4}{3\ N}\ \sum_{\underline{k},\alpha}\ \Big[\ \frac{1}{2}\ (\ F^{x\ *}_{\frac{q}{2}+\underline{k}}F^{x\ *}_{\frac{q}{2}-\underline{k}}\ -\ F^{y\ *}_{\frac{q}{2}+\underline{k}}F^{y\ *}_{\frac{q}{2}-\underline{k}}\ )\ (\ F^{x}_{\frac{q}{2}+\underline{k}}F^{x}_{\frac{q}{2}-\underline{k}}\ -\ F^{y}_{\frac{q}{2}+\underline{k}}F^{y}_{\frac{q}{2}-\underline{k}}\ )\ \Big]\ D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega) \qquad (69)$$

it can be seen that the expression for the form factor in the denominator is similar to the factor in the numerator. Therefore, any bound state associated with a zero of this denominator would represent a non-degenerate mode. The denominator of the third term [$Det_2(\underline{q}:\omega)$] may produce either zero, one or two modes.

Similar to our consideration of the (q,q,q) high-symmetry line, we have evaluated the right-hand side of eqn.(69) for the same temperatures and the same set of q values (π/a), $(39\pi/40a)$, $(19\pi/20a)$, $(14\pi/16a)$, $(6\pi/8a)$ used previously. The temperature dependence is similar to that found for the (q,q,q) direction. The peaks of $\Pi^{1,1}_2(\underline{q},\omega)\ -\ \Pi^{1,2}_2(\underline{q},\omega)$ located near the upper edge of the continuum are shown in the bottom panel of fig.(6). It can be seen that the peaks are non-dispersive, are generally within the continuum and increases slowly as q approaches (π/a). However, the peaks are considerably lower and broader compared

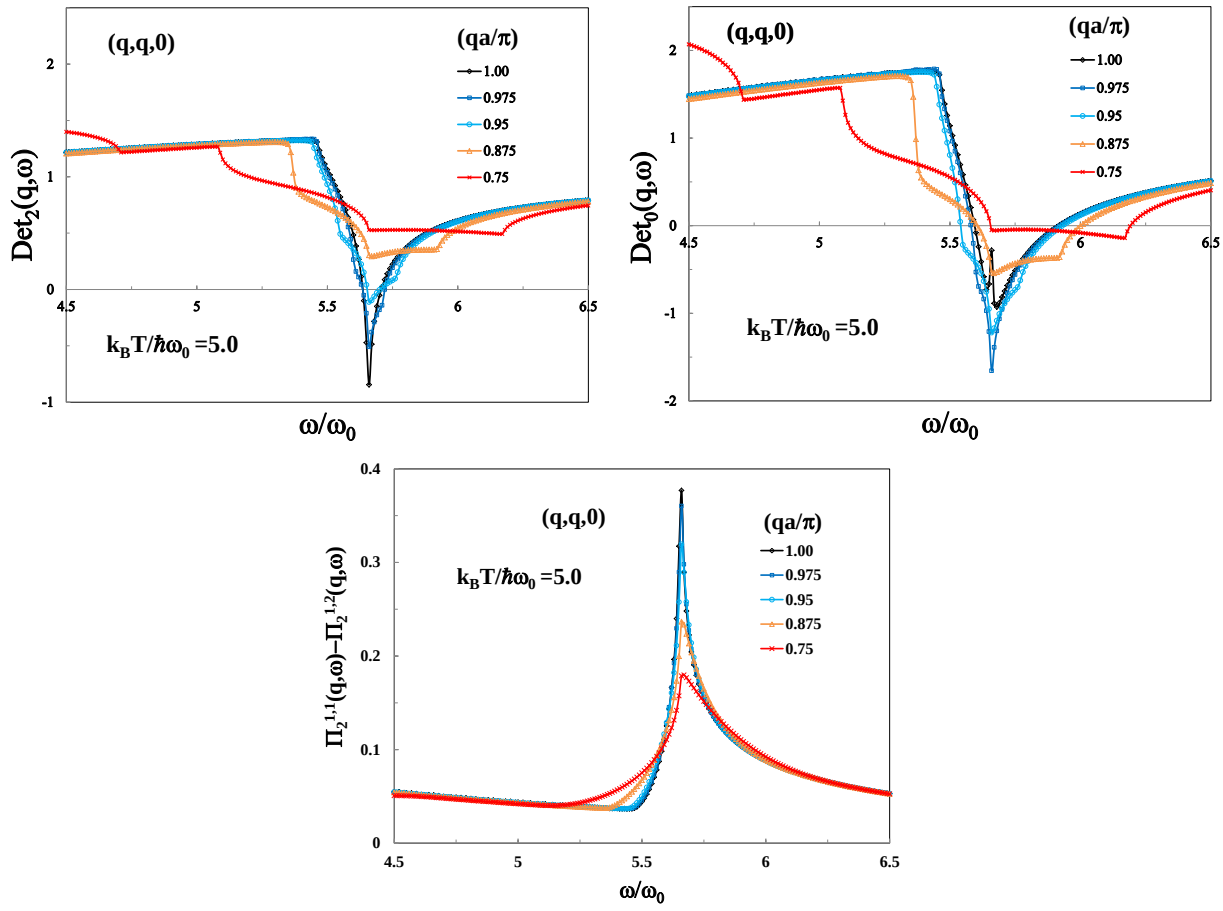

FIG. 6. (Color on line) The ω-dependence of functions in the denominators of the various parts of the two-phonon propagators for various $\underline{q}$ along $(q,q,0)$ with q values given by (π/a), $(39\pi/40a)$, $(19\pi/20a)$, $(7\pi/8a)$, $(3\pi/4a)$ and $\frac{k_BT}{\hbar\omega_0}=5$. The imaginary parts of $\Pi_S^{i,j}(\underline{q},\omega)$ have been discarded. (Top Left Panel). The frequency dependence of the denominator $(Det_2(\underline{q},\omega))$ for $S=2$. (Top Right Panel) The frequency dependence of the $S=0$ denominator $Det_0(\underline{q},\omega)$. (Bottom Panel). The frequency-dependence of the real part of $\Pi_2^{1,1}(\underline{q},\omega) - \Pi_2^{1,2}(\underline{q},\omega)$.

with the case when $\underline{q}$ is directed along the body-diagonal. For the value of the interaction strength $\frac{I_4}{2\sqrt{6}\hbar\omega_0}=0.145$, we find that (x^2-y^2) ILMs do not form at the highest temperature $\frac{k_BT}{\hbar\omega_0}=5$. The relatively small value of the maximum of $\Pi_2^{1,1}(\underline{q},\omega) - \Pi_2^{1,2}(\underline{q},\omega)$ at the top of the two-phonon continuum is attributable to the positions of the van-Hove singularities, shown in fig.(7). Although the van-Hove singularities do coalesce at the Brillouin zone boundary, they coalesce in two groups of four and so the degeneracy at the upper edge of the two-phonon continuum is significantly reduced for the line $(q,q,0)$ as compared to the

higher-symmetry line (q,q,q). The frequency dependencies of the $Det_S(\underline{q},\omega)$ are shown in the upper two panels of fig.(6). At the M point where $q=(\pi/a)$, the off-diagonal matrix elements $\Pi_S^{i,j}(\underline{q},\omega)$ vanish, hence $Det_S(\underline{q},\omega)$ factorizes. Therefore at the point $q=(\pi/a)$, the possible ILMs with quasi-spin S are restricted to the pair of degenerate (x^2+y^2) and (x^2-y^2) modes and a non-degenerate z^2 mode [45]. For other values of q, the off-diagonal matrix elements on the line Σ are non-zero and so the degeneracy of the (x^2-y^2) mode and the mode which may evolve from the (x^2+y^2) mode would be lifted. At our highest temperature and with the chosen value of the interaction strength, only the ILM exist which develops from the $\Pi_S^{z,z}(\underline{q},\omega)$ mode at $q=(\pi/a)$. The ILM dispersion relations are shown in fig.(7). Other branches of ILMs do exist in our model at low temperatures if the interaction

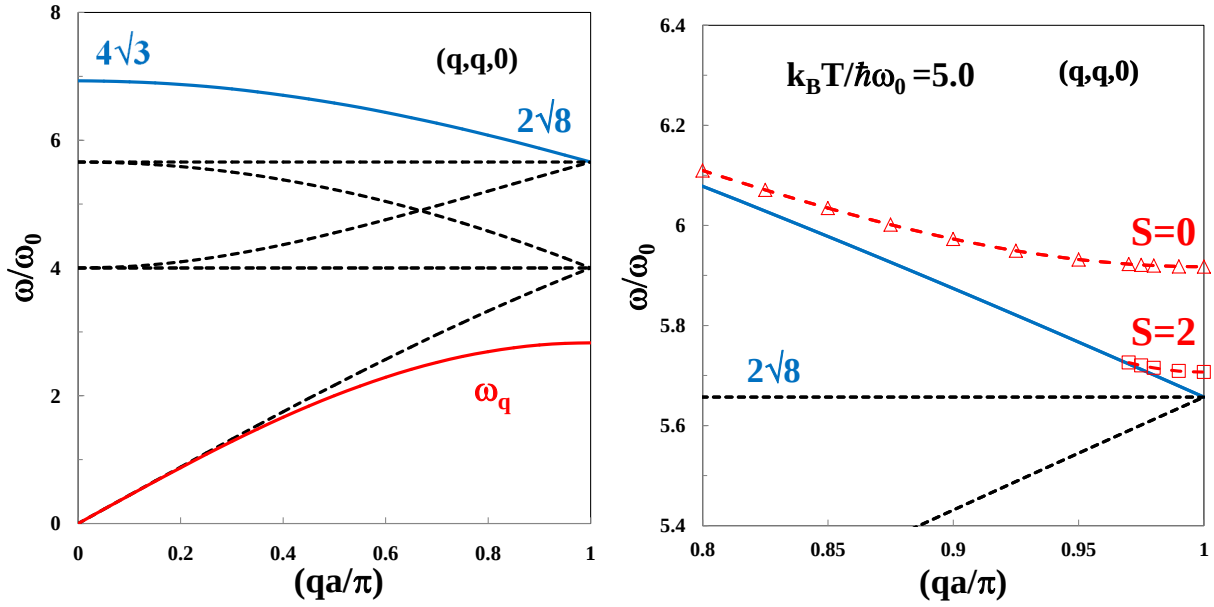

FIG. 7. (Color on line) (Left Panel) The (ω,q) phase space for the $(q,q,0)$ direction. The continuum is bounded from above by the van-Hove singularity denoted by the full blue line, and is bounded from below by the phonon frequency (full red line). The positions of the other van-Hove singularities are denoted by dotted lines. (Right Panel) The dispersion relation for the mixed character ILMs.

strength is significantly higher, but values of the interaction strengths that are larger than the Debye frequency should be regarded as unphysical.

Momentum Transfers (q,0,0)

For momentum transfers $(q,0,0)$ the symmetry of the function $\Pi_2^{i,j}(\underline{q},\omega)$ is reduced from the case (q,q,q) considered earlier, so we have

$$\Pi_2^{1,2}(\underline{q},\omega) \;=\; \Pi_2^{1,3}(\underline{q},\omega) \;\neq\; \Pi_2^{2,3}(\underline{q},\omega) \tag{70}$$

and

$$\Pi_2^{1,1}(\underline{q},\omega) \;\neq\; \Pi_2^{2,2}(\underline{q},\omega) \;=\; \Pi_2^{3,3}(\underline{q},\omega) \tag{71}$$

This symmetry is similar to the symmetry found for $(q,q,0)$. Therefore, the form of the two-phonon propagator along $(q,0,0)$ when expressed in terms of $\Pi_2^{i,j}(\underline{q},\omega)$ reduces to the form found for $(q,q,0)$, except that the indices x and z are interchanged. However, the functions $\Pi_2^{i,j}(\underline{q},\omega)$ are expected to have quite different frequency dependencies when evaluated along the two high-symmetry axes, as can be seen from the respective two-phonon density of states.

Due to the reduced symmetry, the two-phonon propagator reduces to the form

$$\begin{aligned}
D^{(\alpha),(\beta)}_{\sigma,\sigma';\underline{q}}(\underline{k};\underline{k}':\omega) = \Delta_{\underline{k}-\underline{k}'}\;\delta^{\alpha,\beta}_{\sigma,\sigma'}\;D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega) \\
+\;\delta_{\sigma,\sigma'}\;\frac{2\;I_3}{3\;N}\;D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\frac{1}{1-\Pi_2^{3,3}(\underline{q},\omega)+\Pi_2^{3,2}(\underline{q},\omega)}\;D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega) \\
\times\;\Big[\;\frac{1}{2}\;\big(\;F^{y*}_{\frac{q}{2}+\underline{k}}F^{y\;*}_{\frac{q}{2}-\underline{k}}\;-\;F^{z*}_{\frac{q}{2}+\underline{k}}F^{z\;*}_{\frac{q}{2}-\underline{k}}\;\big)\;\big(\;F^{y}_{\frac{q}{2}+\underline{k}'}F^{y}_{\frac{q}{2}-\underline{k}'}\;-\;F^{z}_{\frac{q}{2}+\underline{k}'}F^{z}_{\frac{q}{2}-\underline{k}'}\;\big)\;\Big] \\
+\;\delta_{\sigma,\sigma'}\;\frac{2\;I_3}{3\;N}\;D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega)\;\Big(\;\frac{M_2(\underline{q},\omega)}{Det_2(\underline{q},\omega)}\;\Big)\;D^{(0)(\beta)}_{\sigma',\underline{q}}(\underline{k}':\omega)
\end{aligned} \tag{72}$$

where, for this case, the determinant has the new form of

$$Det_2(\underline{q}:\omega) \;=\; \Big(\;1\;-\;\Pi_2^{1,1}(\underline{q},\omega)\;\Big)\;\Big(\;1\;-\;\Pi_2^{3,3}(\underline{q},\omega)\;-\;\Pi_2^{3,2}(\underline{q},\omega)\;\Big)\;-\;2\;\Big|\;\Pi_2^{3,1}(\underline{q},\omega)\;\Big|^2 \tag{73}$$

and the matrix element also has a different form which is given by

$$\begin{aligned}
M_2(\underline{q},\omega) = F^{x\;*}_{\frac{q}{2}+\underline{k}}F^{x\;*}_{\frac{q}{2}-\underline{k}}\;\Big(\;1-\Pi_2^{3,3}(\underline{q},\omega)-\Pi_2^{3,2}(\underline{q},\omega)\;\Big)\;F^{x}_{\frac{q}{2}+\underline{k}'}F^{x}_{\frac{q}{2}-\underline{k}'} \\
+\;(\;F^{y\;*}_{\frac{q}{2}+\underline{k}}F^{y\;*}_{\frac{q}{2}-\underline{k}}+F^{z\;*}_{\frac{q}{2}+\underline{k}}F^{z\;*}_{\frac{q}{2}-\underline{k}}\;)\;\frac{1}{2}\;\Big(\;1-\Pi_2^{1,1}(\underline{q},\omega)\;\Big) \\
\times\;(\;F^{y}_{\frac{q}{2}+\underline{k}'}F^{y}_{\frac{q}{2}-\underline{k}'}+F^{z}_{\frac{q}{2}+\underline{k}'}F^{z}_{\frac{q}{2}-\underline{k}'}\;) \\
+\;(F^{y\;*}_{\frac{q}{2}+\underline{k}}F^{y\;*}_{\frac{q}{2}-\underline{k}}+F^{z\;*}_{\frac{q}{2}+\underline{k}}F^{z\;*}_{\frac{q}{2}-\underline{k}})\;\Pi_2^{3,1}(\underline{q},\omega)\;(F^{x}_{\frac{q}{2}+\underline{k}'}F^{x}_{\frac{q}{2}-\underline{k}'}) \\
+\;(F^{x\;*}_{\frac{q}{2}+\underline{k}}F^{x\;*}_{\frac{q}{2}-\underline{k}})\;\Pi_2^{1,3}(\underline{q},\omega)\;(F^{y}_{\frac{q}{2}+\underline{k}'}F^{y}_{\frac{q}{2}-\underline{k}'}+F^{z}_{\frac{q}{2}+\underline{k}'}F^{z}_{\frac{q}{2}-\underline{k}'})
\end{aligned} \tag{74}$$

Since the denominator of the second term in the two-phonon propagator

$$1 \; - \; \Pi_2^{3,3}(\underline{q},\omega) \; + \; \Pi_2^{3,2}(\underline{q},\omega) \tag{75}$$

can be rewritten as

$$1 - \frac{2}{3}\frac{I_4}{N} \sum_{\underline{k},\alpha} \Big[\frac{1}{2} \left(F^{y\,*}_{\frac{q}{2}+\underline{k}} F^{y\,*}_{\frac{q}{2}-\underline{k}} - F^{z\,*}_{\frac{q}{2}+\underline{k}} F^{z\,*}_{\frac{q}{2}-\underline{k}} \right) \left(F^{y}_{\frac{q}{2}+\underline{k}} F^{y}_{\frac{q}{2}-\underline{k}} - F^{z}_{\frac{q}{2}+\underline{k}} F^{z}_{\frac{q}{2}-\underline{k}} \right) \Big] D^{(0)(\alpha)}_{\sigma,\underline{q}}(\underline{k}:\omega) \tag{76}$$

it can be seen that the expression for the form factor in the denominator is similar to the factor in the numerator. Thus, any bound states associated with a zero of this denominator should represent a non-degenerate mode. Likewise, the vanishing of the denominator of the third term $Det_2(\underline{q}:\omega)$ may lead to either, two, one or zero modes. If two modes are formed, they are expected to be non-degenerate.

We have examined the condition necessary for ILMs to exist for $\underline{q}$ vectors along the $(q,0,0)$ direction. The frequencies of an ILM should given by the solution of either eqn.(75) or the vanishing of the determinant given in eqn.(73). The results of the right-hand side

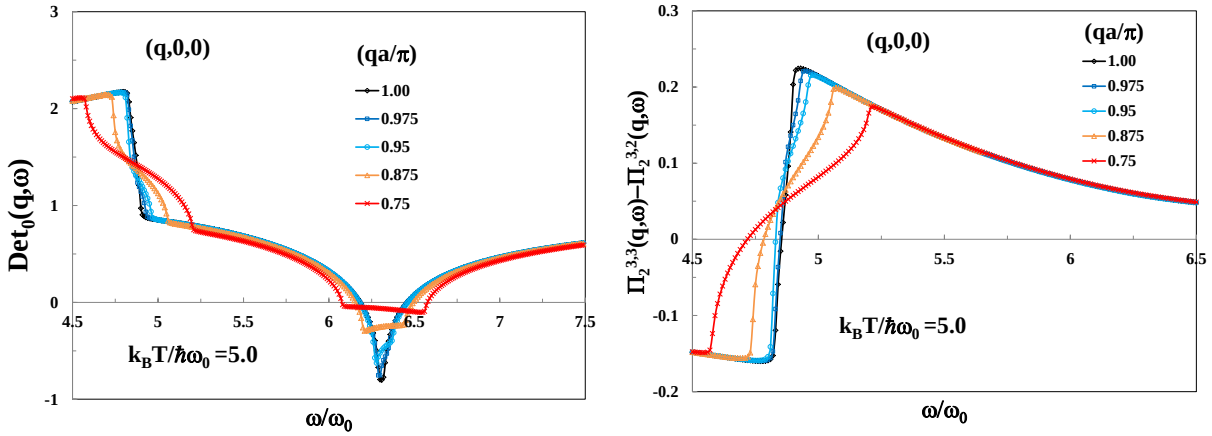

FIG. 8. (Color on line) The frequency dependence of the denominators of the two-phonon propagator (discarding all imaginary parts) evaluated on the line $(q,0,0)$ with the q values given by (π/a), $(39\pi/40a)$, $(19\pi/20a)$, $(7\pi/8a)$, $(3\pi/4a)$ for $\frac{k_BT}{\hbar\omega_0} = 5$. (Left Panel) The frequency dependence of $Det_0(\underline{q},\omega)$ for the $S=0$ excitations in which all imaginary parts of $\Pi_0^{i,j}(q,\omega)$ have been neglected. (Right Panel) The frequency dependence of the real part of $\Pi_2^{3,3}(\underline{q},\omega) - \Pi_2^{3,2}(\underline{q},\omega)$.

of eqn.(75) are shown in fig.(8) for the same q values as considered previously. As can be

seen, the maxima have very small values when compared with the maxima found for the the high-symmetry lines (q,q,q) and $(q,q,0)$. This implies that a much stronger non-linear interaction or higher temperatures would be needed to stabilize a branch of (y^2-z^2) ILMs. The denominator $Det_0(q,\omega)$ is also shown in the left panel of fig.(8) which shows that only one branch of $S=0$ ILMs form for this direction with the temperatures and strength of I_4 chosen. The reduction in number of branches of ILMs is attributed to the reduction in the number of van-Hove singularities that coalesce at $q=(\pi/a)$. Since $\Pi_S^{3,1}(\underline{q},\omega)$ vanishes at $q=(\pi/a)$, the determinant factorizes and it can be recognized that at $q=(\pi/a)$ the mode has (y^2+z^2) character. The ILM dispersion relation and the positions of the van-Hove singularities in the (ω,q) phase space are shown in fig.(9).

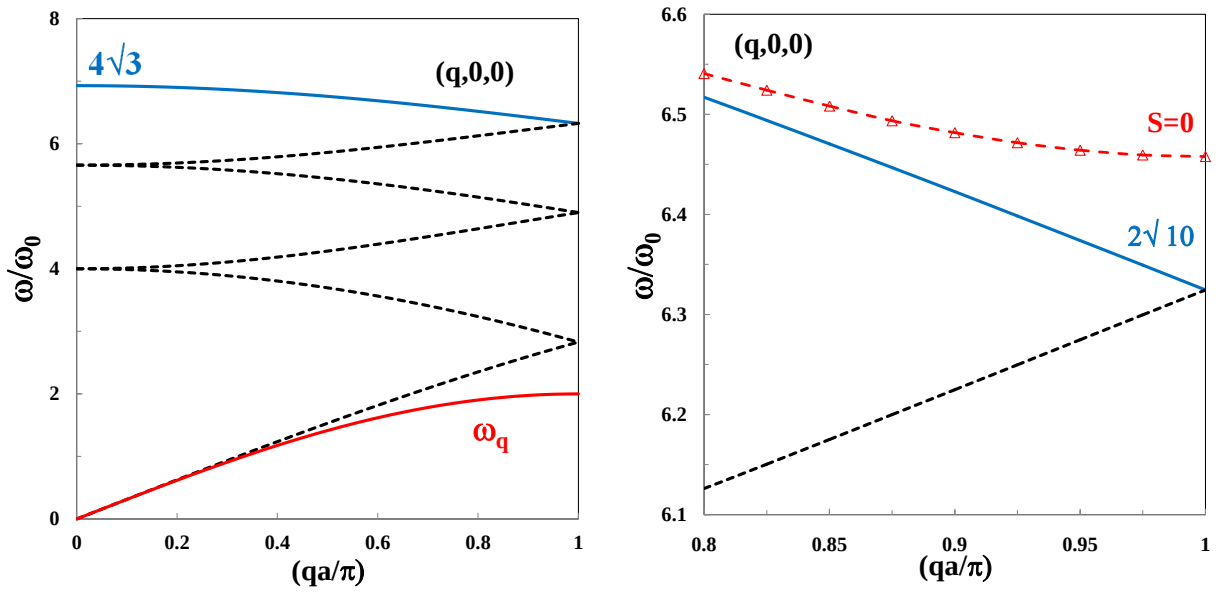

FIG. 9. (Color on line) (Left Panel) The (ω,q) phase space for the $(q,0,0)$ direction. (Right Panel) The dispersion relation for the $S=0$ ILM.

THE ILM WAVE FUNCTIONS

Quantized ILMs are described as many-body states $\mid \Psi_{\underline{q},\sigma} >$, so their wave functions are linear superpositions of wave functions with different numbers of excitations. Here, we shall determine the wave functions corresponding to the ILMs found in the Ladder Approximation.

The ground state $\mid \Phi_0 \; >$ satisfies the eigenvalue equation

$$\hat{H} \mid \Phi_0 \; > = \; E_0 \mid \Phi_0 \; > \tag{77}$$

where E_0 is the ground state energy. The ILM state $\mid \Psi_{\underline{q},\sigma} \; >$ also satisfies the eigenvalue equation

$$\hat{H} \mid \Psi_{\underline{q},\sigma} \; > = \; E_{\underline{q},\sigma} \mid \Psi_{\underline{q},\sigma} \; > \tag{78}$$

where $E_{\underline{q},\sigma}$ is the energy of the ILM state.

The ILMs found in the Ladder Approximation are described as excitations based on the ground state, but are linear superpositions which includes different numbers of phonon excitations

$$\mid \Psi_{\underline{q},\sigma} \; > = \; \sum_{\alpha,\underline{k}} \; C^{(\alpha)}_{\underline{q},\sigma}(\underline{k}) \; a^{a_1}_{a_1(\frac{q}{2}+\underline{k}),p_1} \; a^{a_2}_{a_2(\frac{q}{2}-\underline{k}),p_2} \mid \Phi_0 \; > \tag{79}$$

where $\alpha \equiv (a_1, a_2)$ and $\sigma \equiv (p_1, p_2)$. The complex coefficients $C^{(\alpha)}_{\underline{q},\sigma}(\underline{k})$ can be found by projecting the ILM state onto the components with fixed numbers of (harmonic) phonons

$$C^{(\alpha)}_{\underline{q},\sigma}(\underline{k}) \; = \; < \; \Phi_0 \mid a^{a_2 \; \dagger}_{a_2(\frac{q}{2}-\underline{k}),p_2} \; a^{a_1 \; \dagger}_{a_1(\frac{q}{2}+\underline{k}),p_1} \mid \Psi_{\underline{q},\sigma} \; > \tag{80}$$

By taking the appropriate matrix elements of the eigenvalue equations, one can show that

$$\begin{aligned} & \Big(\; E_{\underline{q},\sigma} \; - \; E_0 \; - \; a_1 \; \hbar\omega_{\frac{q}{2}+\underline{k}} \; - \; a_2 \; \hbar\omega_{\frac{q}{2}-\underline{k}} \; \Big) \; C^{(\alpha)}_{\underline{q},\sigma}(\underline{k}) \\ & = \; < \; \Phi_0 \mid [\; a^{a_2 \; \dagger}_{a_2(\frac{q}{2}-\underline{k}),p_2} \; a^{a_1 \; \dagger}_{a_1(\frac{q}{2}+\underline{k}),p_1} \; , \; \hat{H}_{int} \;] \mid \Psi_{\underline{q},\sigma} \; > \end{aligned} \tag{81}$$

The expectation of the commutator can be evaluated in the Ladder Approximation, to yield (for $p_1 \neq p_2$, i.e. $S = 2$)

$$C^{(\alpha)}_{\underline{q},\sigma}(\underline{k}) \; = \; \frac{2 \; I_4}{3 \; N} \; \sum_i \; F^i_{\frac{q}{2}+\underline{k}} F^i_{\frac{q}{2}-\underline{k}} \; D^{(0),(\alpha)}_{\underline{q},\sigma}(\underline{k} : \omega) \; \sum_{\underline{k}',\gamma} \; F^{i \; *}_{\frac{q}{2}+\underline{k}'} F^{i \; *}_{\frac{q}{2}-\underline{k}'} C^{(\gamma)}_{\underline{q},\sigma}(\underline{k}') \tag{82}$$

where ω is the excited energy which is fixed by

$$\hbar \; \omega \; = \; E_{\underline{q},\sigma} \; - \; E_0 \tag{83}$$

On defining the components of a vector $\Phi^{i \; *}_{\underline{q},\sigma}$ via

$$\Phi^{i \; *}_{\underline{q},\sigma} \; = \; \sum_{\underline{k}',\gamma} \; F^{i \; *}_{\frac{q}{2}+\underline{k}'} F^{i \; *}_{\frac{q}{2}-\underline{k}'} C^{(\gamma)}_{\underline{q},\sigma}(\underline{k}') \tag{84}$$

one can find that the self-consistency condition is given by the homogeneous matrix equation

$$\left[\tilde{I} \; - \; \tilde{\Pi}_2^*(\underline{q},\omega) \right] \underline{\Phi}^*_{\underline{q},\sigma} \; = \; 0 \tag{85}$$

For the above equation to have a solution, the determinant of the matrix must vanish. Therefore, the excitation energy of the ILM must satisfy exactly the same condition found from considerations of the two-phonon propagator. Since the matrix equation is homogeneous, the eigenvector $\underline{\Phi}^*_{\underline{q},\sigma}$ is only defined up to a normalization and an arbitrary phase factor. Once the eigenvector has been obtained, the un-normalized expansion coefficients are given by

$$C^{(\alpha)}_{\underline{q},\sigma}(\underline{k}) \; = \; \frac{2}{3} \frac{I_4}{N} \sum_i \; F^i_{\frac{\underline{q}}{2}+\underline{k}} F^i_{\frac{\underline{q}}{2}-\underline{k}} \; D^{(0),(\alpha)}_{\underline{q},\sigma}(\underline{k}:\omega) \; \Phi^{i\;*}_{\underline{q},\sigma} \tag{86}$$

Since the center of mass wave function is normalized to unity, the many-body relative wave function is subject to the normalization condition

$$\sum_{\underline{k},\alpha} \; \mid C^{(\alpha)}_{\underline{q},\sigma}(\underline{k}) \mid^2 \; = \; 1 \tag{87}$$

A measure of the reasonableness of the Ladder Approximation is given by the ratio of $C^{--}_{\underline{q},\sigma}$ to $C^{++}_{\underline{q},\sigma}$. The coefficient $C^{--}_{\underline{q},\sigma}$ can be considered as a measure of the fluctuations in the total number of phonons and when it vanishes the Ladder Approximation reduces to the T-matrix approximation which becomes exact.

Since ILMs preferentially form near the high-symmetry point (π,π,π), we have examined the relative coordinate part of the two-phonon wave function for momentum transfers on the high-symmetry line (q,q,q). For $\underline{q}$ on this high-symmetry line, we found that the ILMs consist of a non-degenerate and a doubly-degenerate mode.

The excitation energy of the doubly-degenerate excitation is given by the solution of

$$1 \; - \; \Pi_2^{1,1}(\underline{q},\omega) \; + \; \Pi_2^{1,2}(\underline{q},\omega) \; = \; 0 \tag{88}$$

These excitations correspond to linear combinations of the two eigenvectors

$$\begin{pmatrix} \Phi^{1\;*}_{\underline{q},\sigma} \\ \Phi^{2\;*}_{\underline{q},\sigma} \\ \Phi^{3\;*}_{\underline{q},\sigma} \end{pmatrix} \; = \; \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -1 \\ 0 \end{pmatrix} \tag{89}$$

and

$$\begin{pmatrix} \Phi^{1\;*}_{\underline{q},\sigma} \\ \Phi^{2\;*}_{\underline{q},\sigma} \\ \Phi^{3\;*}_{\underline{q},\sigma} \end{pmatrix} \; = \; \frac{1}{\sqrt{6}} \begin{pmatrix} 1 \\ 1 \\ -2 \end{pmatrix} \tag{90}$$

Thus, the un-normalized expansion coefficients for the two degenerate excitations are given by

$$C^{(\alpha)}_{\underline{q},\sigma}(\underline{k}) \;=\; \frac{2\ I_4}{3\ N}\ D^{(0),(\alpha)}_{\underline{q},\sigma}(\underline{k}:\omega)\ \frac{1}{\sqrt{2}}\ \Big(\ F^x_{\frac{\underline{q}}{2}+\underline{k}}F^x_{\frac{\underline{q}}{2}-\underline{k}}\ -\ F^y_{\frac{\underline{q}}{2}+\underline{k}}F^y_{\frac{\underline{q}}{2}-\underline{k}}\ \Big) \tag{91}$$

and

$$C^{(\alpha)}_{\underline{q},\sigma}(\underline{k}) \;=\; \frac{2\ I_4}{3\ N}\ D^{(0),(\alpha)}_{\underline{q},\sigma}(\underline{k}:\omega)\ \frac{1}{\sqrt{6}}\ \Big(\ F^x_{\frac{\underline{q}}{2}+\underline{k}}F^x_{\frac{\underline{q}}{2}-\underline{k}}\ +\ F^y_{\frac{\underline{q}}{2}+\underline{k}}F^y_{\frac{\underline{q}}{2}-\underline{k}}\ -\ 2\ F^z_{\frac{\underline{q}}{2}+\underline{k}}F^z_{\frac{\underline{q}}{2}-\underline{k}}\ \Big). \tag{92}$$

The non-degenerate ILM with $S = 2$ has an excitation energy ω which is determined from

$$1\ -\ \Pi^{1,1}_2(\underline{q},\omega)\ -\ 2\ \Pi^{1,2}_2(\underline{q},\omega)\ =\ 0 \tag{93}$$

This eigenvalue corresponds to the eigenvector

$$\begin{pmatrix} \Phi^{1\ *}_{\underline{q},\sigma} \\ \Phi^{2\ *}_{\underline{q},\sigma} \\ \Phi^{3\ *}_{\underline{q},\sigma} \end{pmatrix} \;=\; \frac{1}{\sqrt{3}}\ \begin{pmatrix} 1 \\ 1 \\ 1 \end{pmatrix} \tag{94}$$

and so the un-normalized components of the $(x^2+y^2+z^2)$ wavefunction are given by

$$C^{(\alpha)}_{\underline{q},\sigma}(\underline{k}) \;=\; \frac{2\ I_4}{3\ N}\ D^{(0),(\alpha)}_{\underline{q},\sigma}(\underline{k}:\omega)\ \frac{1}{\sqrt{3}}\ \Big(\ F^x_{\frac{\underline{q}}{2}+\underline{k}}F^x_{\frac{\underline{q}}{2}-\underline{k}}\ +\ F^y_{\frac{\underline{q}}{2}+\underline{k}}F^y_{\frac{\underline{q}}{2}-\underline{k}}\ +\ F^z_{\frac{\underline{q}}{2}+\underline{k}}F^z_{\frac{\underline{q}}{2}-\underline{k}}\ \Big) \tag{95}$$

We have evaluated the relative coordinate, two-phonon creation part of the $S = 0$ ILM wave function given in eqn.(95) corresponding to the non-degenerate symmetric $(x^2+y^2+z^2)$ mode. Figure(10) shows the results for $T = 0$ and for the value of I_4 given by $\frac{I_4}{2\sqrt{6}\hbar\omega_0} = 0.7458$. We have chosen a very large value of the interaction strength in order to illustrate the spatial dependence of the wave functions compactly. Even with this very large value of the interaction, the ratio of $C^{--}_{\underline{q},\sigma}$ to $C^{++}_{\underline{q},\sigma}$ is only of the order of $\sim 3\times 10^{-2}$, indicating that the resonant part of the anharmonic interaction still dominates over the non-resonant part, so the Ladder Approximation is still expected to lead to reasonable results. The left plot indicates the wave function at $\underline{q} = (\pi/a)(1,1,1)$. Similarly the right plot shows the wave function at $\underline{q} = (38\pi/40a)(1,1,1)$. It is clearly seen that the wave function of this non-degenerate mode is symmetric under rotations of $\frac{\pi}{4}$. Furthermore, the amplitude of the relative wave function has an envelope that decreases exponentially with distance from the center. The localization length is of the order of a lattice spacing, since we have chosen a very large interaction strength and since the localization length decreases as the interaction strength is increased. The wave function with $\underline{q} = (\pi/a)(1,1,1)$ has a faster decay compared

to $\underline{q} = (38\pi/40a)(1,1,1)$. We relate the q-dependence of the decay to the q-dependence of the binding energy, since the localization length can be shown to go to infinity when the binding energy approaches zero. For $\underline{q} = (\pi/a)(1,1,1)$, it is seen that the wave function goes to zero at every alternate site, which is due to a complete destructive interference originating from $\underline{k}$ values close to pairs of the degenerate van-Hove singularities. The destructive interference is no longer complete when $\underline{q}$ is moved away from (π/a). The reason for the incomplete interference is that the van-Hove singularities have their degeneracies lifted, therefore, the amplitudes of $C^{\alpha}_{\underline{q},\sigma}(\underline{k})$ are no longer equal at the $\underline{k}$ positions of the singularities.

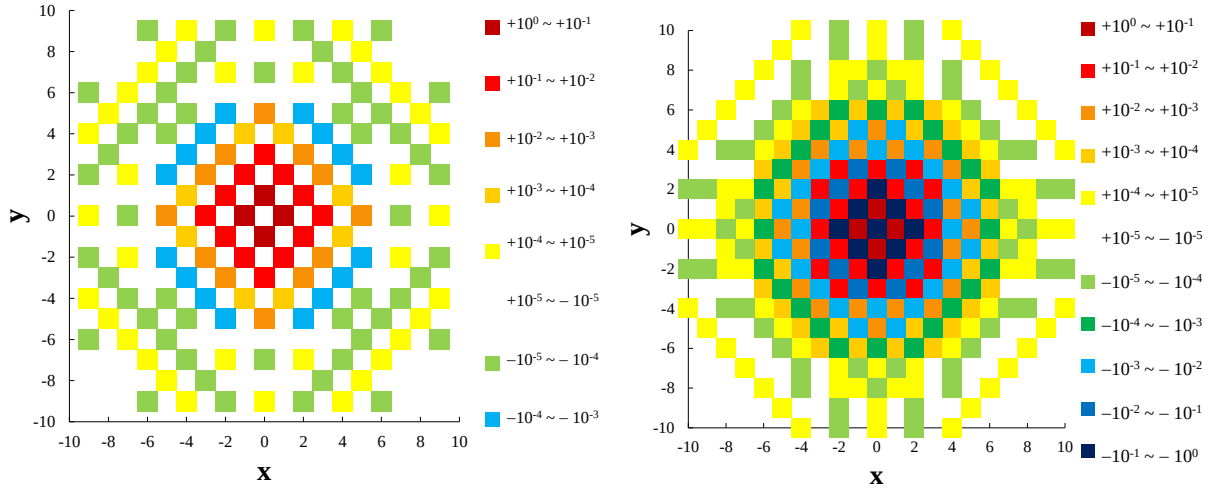

FIG. 10. (Color on line) A contour plot of the (un-normalized) relative two-phonon creation part of the symmetric $(x^2+y^2+z^2)$ ILM wave function ${\Psi_{\underline{q}}}^{++}(R)$ corresponding to eqn.(95) plotted at lattice sites in the plane $R_z = 0$ for $\underline{q} = (\pi/a)(1,1,1)$ (Left) and for $\underline{q} = (38\pi/40a)(1,1,1)$ (Right).

We have evaluated the wave functions for the degenerate pair of $S = 0$ excitations corresponding to the $S = 0$ counterparts of eqn.(91) and eqn.(92). Figure(11) shows the two-phonon creation part of the $S = 0$ ILM's relative coordinate wave function ${\Psi_{\underline{q}}}^{++}(R)$ corresponding to the antisymmetric $(x^2 - y^2)$ mode given in eqn.(91) for $T = 0$ and a large interaction strength $\frac{I_4}{2\sqrt{6}\hbar\omega_0} = 0.7458$. The wavefunctions are plotted in the (x, y) plane. The left plot shows the wavefunction for $\underline{q} = (\pi/a)(1,1,1)$ and similarly the right plot shows the wave function at a smaller center of mass momentum $\underline{q} = (38\pi/40a)(1,1,1)$. It is seen that this mode is antisymmetric under the interchange $(x \leftrightarrow y)$ and, therefore, has a line of nodes in the (x, y) plane. Furthermore, one sees that the wave function has an envelope which decays more slowly when q is moved away from (π/a). Also, one sees that the inter-

ference at the alternate sites becomes incomplete for $q \neq (\pi/a)$, similar to the results for the non-degenerate mode.

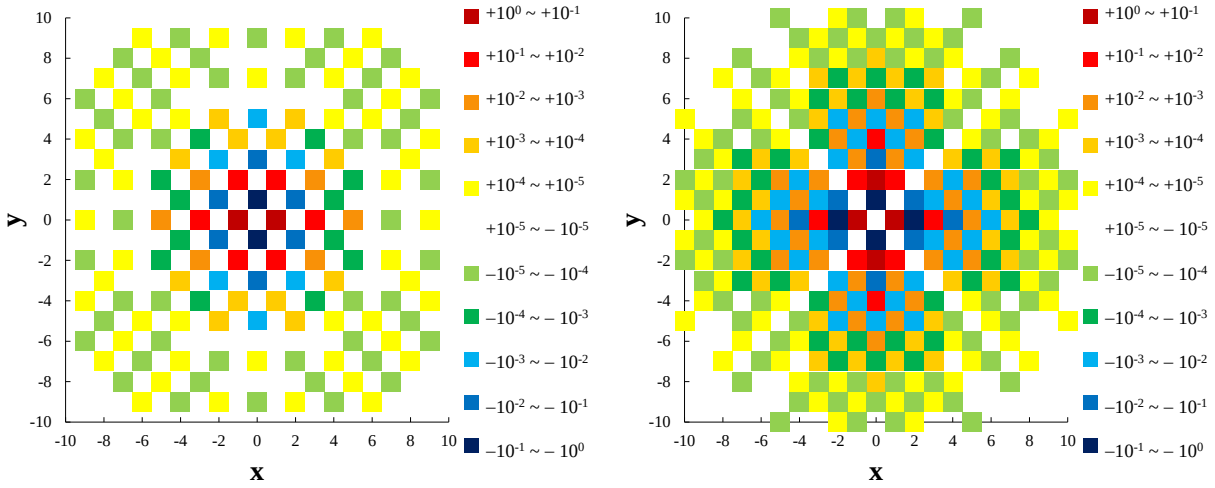

FIG. 11. (Color on line) A contour plot of the (un-normalized) relative two-phonon creation part of the $S = 0$ $(x^2 - y^2)$ ILM wave function ${\Psi_{\underline{q}}}^{++}(R)$ corresponding to eqn.(91) plotted at lattice sites in the plane $R_z = 0$ for $\underline{q} = (\pi/a)(1, 1, 1)$ (Left) and for $\underline{q} = (38\pi/40a)(1, 1, 1)$ (Right).

The degenerate partner $S = 0$ $(x^2 + y^2 - 2z^2)$ wavefunction [corresponding to eqn.(92)] is plotted in the (x, y) plane in fig.(12) for $T = 0$ and $\frac{I_4}{2\sqrt{6}\hbar\omega_0} = 0.7458$. The left plot shows the ILM wavefunction ${\Psi_{\underline{q}}}^{++}(R)$ for $\underline{q} = (\pi/a)(1, 1, 1)$ and the right plot shows the wave function for $\underline{q} = (38\pi/40a)(1, 1, 1)$. The $(x^2 + y^2 - 2z^2)$ wave function at $q = (\pi/a)$, when plotted in the (x, y) plane is identical to the $(x^2 + y^2 + z^2)$ wave function at $q = (\pi/a)$. It is seen that the wave function is symmetric under rotations through $\frac{\pi}{2}$ around the z-axis. The only point in the (x, y) plane where the nodes are commensurate with the underlying lattice is the point $x = y = z = 0$, so the $(x^2 + y^2 - 2z^2)$ wavefunction is always identically zero at the origin. In addition to a discrete "ring" of commensurate points found on a plane of constant z, the nodes of the $(x^2 + y^2 - 2z^2)$ wavefunction can be easily seen by plotting the wave function in the plane $x - y = 0$. Since this is simply related to the projection of the $(x^2 - y^2)$ wave function on to the (x, y) plane, we shall not plot that here. The wave function has an exponentially decaying envelope which, as explained previously, has a q-dependent localization length that is governed by the binding energy. Likewise, the degree of destructive interference is q-dependent and is governed by the relative energies of the van-Hove singularities.

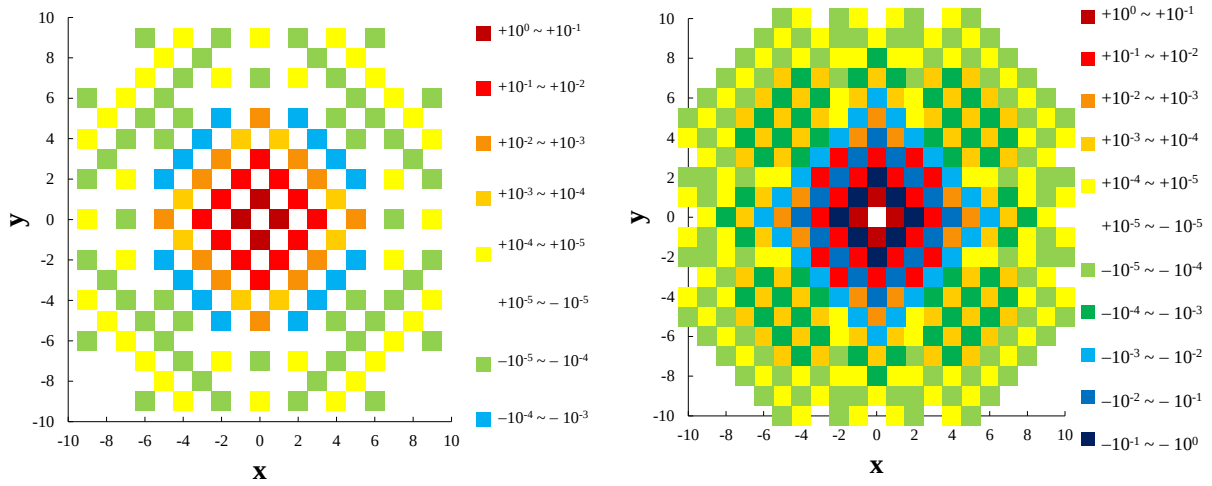

FIG. 12. (Color on line) A contour plot of the (un-normalized) relative two-phonon creation part of the $S = 0$ $(x^2 + y^2 - 2z^2)$ ILM wave function $\Psi_{\underline{q}}{}^{++}(R)$ corresponding to eqn.(92) plotted at lattice sites in the plane $R_z = 0$ for $\underline{q} = (\pi/a)(1, 1, 1)$ (Left) and for $\underline{q} = (38\pi/40a)(1, 1, 1)$ (Right).

DISCUSSION

We have examined the lowest-energy members of the quantized ILMs of a generalization of the Fermi-Pasta-Ulam Hamiltonian to three-dimensions. In the generalized Fermi-Pasta-Ulam model, the acoustic phonons are degenerate with respect to the polarization index so the phonons can be regarded as quasi-spin one excitations. The lowest energy ILMs are similar in form to multi-phonon bound states [38], except that the number of phonons is not conserved. A simple projection of the low-energy excitations on to manifold of states with a specific number of phonons leads to a violation of Goldstone's theorem [39]. In order to avoid this violation, we found it is necessary to include states in which the total number of phonons is allowed to either increase or decrease by two. We calculate the lowest members of the ILM excitations using the Ladder Approximation. The Ladder Approximation augments the space of a fixed number of phonons and, in this respect, is consistent with our treatment of the Goldstone modes. The ILMs can be categorized as having a quasi-spin of either $S = 2$ or $S = 0$ and have other internal quantum numbers. We find that ILMs can form in three-dimensions at zero temperature, but only if the interaction exceeds a minimum value. We find that, as the temperature is raised, the magnitude of the minimal interaction required to stabilize the ILM is reduced. When the ILMs first form they split off from the

top of the two-phonon continuum. The $S = 0$ ILMs form for lower values of the interaction than the $S = 2$ ILMs. The ILMs form preferentially for center of mass momentum $\underline{q}$ at the corner of the Brillouin zone. The tendency of ILMs to form at this momentum is traced to a confluence of van-Hove singularities in the (non-interacting) two-phonon density of states at the top of the two-phonon continuum for this $\underline{q}$ value. We have examined the ILM wave functions and find that the relative coordinate part of the wave functions have symmetries that are dictated by the internal quantum numbers. The (degenerate) asymmetric ILMs wave functions exhibit nodes which have the effect of increasing their kinetic energies relative to the (non-degenerate) symmetric $(x^2+y^2+z^2)$ ILM and, thus, the nodes are responsible for the splitting between the branches of the ILM dispersion relations. This splitting vanishes at $q = (\pi/a)$ due to a destructive interference in the wave function which originates from the $\underline{k}$ vectors associated with pairs of van-Hove singularities. As a result, for this particular value of the center of mass momentum q, the symmetric and antisymmetric modes vanish at every alternate site thereby obfuscating the nodes and eliminating the splitting between the branches.

The results show a number of similarities with the early experimental results of Manley *et al.*. First, NaI has a rock salt structure and the ILMs are only found in the vicinity of $\underline{q} = (\pi/a)(1, 1, 1)$, whereas the theory for a simple cubic lattice shows that the ILMs prefer to form at the same $\underline{q}$ value. Experimentally, it was found that the ILMs only occur at elevated temperatures, i.e. the ILMs were observed at $T = 555$ K [19] but were not observed at 100 K [46]. This finding is consistent with our theory, if it is assumed that at low temperatures the interaction is too weak to produce ILM but, due to the presence of a Bose-Einstein function, is enhanced above the minimum value at $T = 555$ K. Experimentally, the ILM's energy is found to be in the gap between the optic and acoustic modes. As seen in fig.(1), the energy separation between the ILM and the longitudinal or transverse acoustic modes is already sizeable [20]. The magnitude of this separation is too large to be accounted for by classical theories using standard two-body potentials [20]. Furthermore, the localization length of 1.2 nm inferred from the measurements of Manley *et al.* would not be compatible with a large binding energy. In our quantum theory, the existence of such a large energy separation does not require an unusually large anharmonicity, since the binding energy of the ILM should be measured from the top edge of the two-phonon continuum and not the acoustic phonon energy as found in classical theories [17, 18, 20]. Also, the spatial extent of

the ILM inferred from the measurements is consistent with that expected from our theory with $\frac{I_4}{2\sqrt{6}\hbar\omega_0} \sim 0.1$. The discrepancies between the first measurements of Manley *et al.* and our theory are that our theory predicts that the ILM is dispersive whereas the measurements show a flat branch which extends over a sizeable q range ($\sim \frac{1}{3}$ of the Brillouin zone). One may speculate that the flatness of the observed dispersion relation is an indication that the measurements are sensitive to the asymmetric ILMs, which have a higher intensity due to the degeneracy and for which the dispersion relations are flatter. However, the large q range over which the ILM peak persists would have to be interpreted as indicating the branch of long-lived ILM extends within the two-phonon continuum as a broadened resonance. This interpretation in terms of a broadening of the feature could be consistent with the observed loss of intensity that occurs as q is varied towards the zone center. It should be noted that these speculations are based on the assumption that the features of this calculation are robust and would survive in a more realistic description of the lattice dynamics, which includes the optic modes, lifts the degeneracy between the phonons with different polarizations and also includes cubic anharmonicity. A more serious challenge to theory is presented by recent experiments [22, 23] which suggest that the ILMs are highly sensitive to small changes in temperature and may act cooperatively. The experiments suggest that small temperature changes produce symmetry-breaking dynamical structures [22]. Furthermore, the experiments indicate that the ILMs form randomly stacked two-dimensional structures [23], in contrast to the dilute gas of localized ILMs discussed here.

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