

## Quantized intrinsically localized modes of the Fermi–Pasta–Ulam lattice

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We have examined the quantized  $n=2$  excitation spectrum of the Fermi–Pasta–Ulam lattice. The spectrum is composed of a resonance in the two-phonon continuum and a branch of infinitely long-lived excitations which splits off from the top of the two-phonon creation continuum. We calculate the zero-temperature limit of the many-body wavefunction and show that this mode corresponds to an intrinsically localized mode (ILM). In one dimension, we find that there is no lower threshold value of the repulsive interaction that must be exceeded if the ILM is to be formed. However, the spatial extent of the ILM wavefunction rapidly increases as the interaction is decreased to zero. The dispersion relation shows that the discrete  $n=2$  ILM and the resonance hybridize as the center of mass wavevector  $q$  is increased towards the zone boundary. The many-body wavefunction shows that as the zone boundary is approached, there is a destructive resonance which occurs between pairs of sites separated by an odd number of lattice spacings. We compare our theoretical results with the recent experimental observation of a discrete ILM in NaI by Manley et al.

**Keywords:** intrinsically localized modes; discrete breathers; quantized excitations; Fermi–Pasta–Ulam model

### 1. Introduction

In 1955, Enrico Fermi, Joe Pasta and Stanislaw Ulam pioneered the use of computers in theoretical physics when they studied the equilibration of the vibrations of a finite segment of a classical, weakly anharmonic, one-dimensional lattice [1]. In particular, they numerically solved the equations of motion for the weakly anharmonic chain which had initial conditions that corresponded to the excitation of a normal mode found in the harmonic approximation. What they expected was that the anharmonicity would result in a redistribution of energy between the various approximate normal modes and that eventually, an equilibrium state would be attained. They expected that in the equilibrium state, the energy would be distributed amongst the harmonic modes in a manner consistent with the equipartition theorem. However, they were surprised when they found that although the system initially showed signs of equilibration, the sharing of energy ceased, then reversed, and that

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the initial configuration was approached on a time-scale much shorter than the Poincaré recurrence time of the linear system. (For a chain of 32, atoms the initial state was almost recovered on a time-scale associated with about 158 periods of the harmonic oscillators.) Subsequent computations by James Tuck and Mary Tsingou Menzel [2] showed that, on a longer time-scale, the system exhibited super-recurrences. This phenomenon remained a complete puzzle until Zabusky and Kruskal mapped the discrete lattice problem onto the nonlinear Korteweg–de Vries equation and showed [3], via numerical computation, that this system also exhibited a similarly unusual recurrence phenomenon. In 1895 the Korteweg–de Vries equation (KdV) was proposed [4] to describe the localized non-dispersive wave forms that had been observed in canals [5]. The waves described by the KdV equation have their forms stabilized by a balance between the nonlinear terms with the dispersion. Zabusky and Kruskal showed [3] that when two non-dispersive waves of the sort described by Korteweg and de Vries collided, they emerged after the collision with their forms and velocities intact. Zabusky and Kruskal first penned the term solitons to describe these localized and non-dispersive waves. This discovery spurred intensive analytic investigation [6,7], which soon showed that the KdV system was completely integrable. For finite discrete lattice systems, integrability implies that the number of degrees of freedom of the system is equal to the number of independent invariants. For the continuous KdV system, Miura et al. [8] found a method for constructing an infinite number of constants of motion. The existence of the infinite number of conservation laws affects the non-ergodic nature of the KdV system. The discovery of the multi-soliton solutions of the KdV equation was soon followed by the discovery of breather excitations. Breather excitations have the form of stable pulsating waves that are of finite spatial extent and are stabilized by nonlinear interactions (see Figure 1). Soliton and breather-like excitations have been found in many other integrable continuous systems [9], and solitons have also been found in integrable discrete lattice systems [10]. Approximate soliton excitations have been predicted and observed in non-integrable lattice systems [11–14].

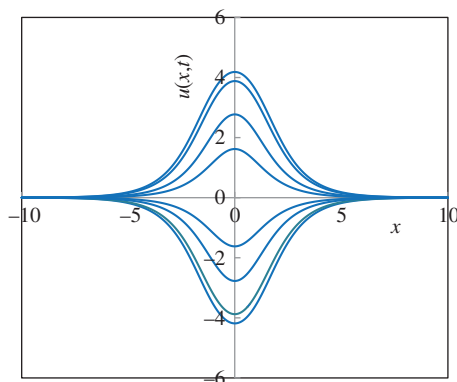


Figure 1. Snapshots showing the evolution of a breather excitation of the classical continuous sine-Gordon theory. The breather is shown in its inertial reference frame. The breather is an oscillatory excitation of finite spatial extent.

Sievers and Takeno suggested [15] that similar pulsating localized excitations should also exist in non-integrable discrete lattices and called these excitations intrinsically localized modes (ILMs). Much work has been performed on ILMs of classical discrete lattices [16–20], however, the quantum versions of the systems have received little attention [21], although there has been some early work on these types of excitations in magnetic systems [23,31] and systems with conserved numbers of bosons [24]. In classical integrable continuous systems such as the sine-Gordon system, a breather excitation can be considered as bound states of a soliton and an anti-soliton. Snapshots of a sine-Gordon breather excitation in its rest frame are shown in Figure 1, and the frequencies of the oscillations  $\omega$  are related to their spatial length-scales  $\xi$ , via

$$\omega^2 + c^2\xi^{-2} = \text{const} \quad (1)$$

where  $c$  is the speed of sound. When the internal motions of the breathers were quantized semi-classically using the Bohr–Sommerfeld quantization procedure [25,26], it became evident that the breathers form a hierarchy of excitations that can be considered as the bound states of multiples of small-amplitude wave excitations. This interpretation has been confirmed by the exact solution of the integrable quantum systems [27,28]. In recent publications [29,30], the spectra of quantized ILMs of the monatomic and diatomic Fermi–Pasta–Ulam models have been investigated using a T-matrix approach. The T-matrix approach is expected to be exact for excitations consisting of two interacting bosons in models where the number of bosons is conserved. In this note, we investigate the localized nature of the lowest members of the hierarchy of  $T=0$  excitations.

## 2. The Hamiltonian of the Fermi–Pasta–Ulam model

The Hamiltonian for the discrete quartic Fermi–Pasta–Ulam chain can be written as

$$\hat{H} = \sum_i \left[ \frac{\hat{P}_i \hat{P}_i^\dagger}{2M} + \frac{M\omega_0^2}{2} (\hat{u}_i - \hat{u}_{i+1})^2 \right] + \frac{K_4}{12} \sum_i (\hat{u}_i - \hat{u}_{i+1})^4 \quad (2)$$

in which  $\hat{u}_i$  is the displacement operator for the atom at the  $i$ -th lattice site from its nominal equilibrium position<sup>1</sup> and  $\hat{P}_i$  is the momentum operator for the corresponding atom. The first two terms in the Hamiltonian represent the approximate harmonic Hamiltonian and the third term, proportional to  $K_4$ , represents the anharmonic interaction.

The spatial Fourier transform of coordinates and momenta operators of the atoms is defined as

$$\hat{u}_q = \frac{1}{\sqrt{N}} \sum_i \exp[i q R_i] \hat{u}_i$$

$$\hat{P}_q = \frac{1}{\sqrt{N}} \sum_i \exp[i q R_i] \hat{P}_i \quad (3)$$

where  $R_i$  represents the location of the  $i$ -th lattice site. The Hamiltonian can be rewritten in terms of operators in the harmonic normal mode basis as

$$\hat{H} = \sum_q \left[ \frac{\hat{P}_q \hat{P}_q^\dagger}{2M} + M\omega_0^2(1 - \cos q)\hat{u}_q \hat{u}_q^\dagger \right] + \frac{K_4}{3N} \sum_{q,k,k'} \left( \cos \frac{q}{2} - \cos k \right) \left( \cos \frac{q}{2} - \cos k' \right) \hat{u}_{\frac{q}{2}+k} \hat{u}_{\frac{q}{2}-k} \hat{u}_{\frac{q}{2}-k'} \hat{u}_{\frac{q}{2}+k'}^\dagger. \quad (4)$$

100 Since the anharmonic interaction has a separable form, the ladder approximation for the two-particle excitations can be solved exactly.

The harmonic part of the Hamiltonian can be second quantized and diagonalized by the substitutions

$$\hat{P}_q = i \left( \frac{M\hbar\omega_q}{2} \right)^{\frac{1}{2}} (a_{-q}^\dagger - a_q) \quad (5)$$

$$\hat{u}_q = \left( \frac{\hbar}{2M\omega_q} \right)^{\frac{1}{2}} (a_q^\dagger + a_{-q})$$

105 where, respectively,  $a_q^\dagger$  and  $a_q$  are the boson creation and annihilation operators and where the phonon dispersion relation is given by

$$\omega_q^2 = 4\omega_0^2 \sin^2 \frac{q}{2}. \quad (6)$$

This dispersion relation describes a branch of collective bosonic excitations with frequencies that tend to zero in the limit  $q \rightarrow 0$ , as guaranteed by Goldstone's theorem. Thus, the Hamiltonian of the  $\beta$  Fermi–Pasta–Ulam lattice can be written in the second quantized form

$$\hat{H} = \sum_q \frac{\hbar\omega_q}{2} (a_q^\dagger a_q + a_q a_q^\dagger) + \hat{H}_{\text{int}} \quad (7)$$

110 where  $\hat{H}_{\text{int}}$  is the interaction Hamiltonian. The interaction has a separable form and is given by

$$\hat{H}_{\text{int}} = \frac{U_4}{12N} \sum_{k_1, k_2, k_3, k_4} \Delta_{k_1+k_2+k_3+k_4} \prod_{j=1}^4 \{F_{k_j} (a_{k_j}^\dagger + a_{-k_j})\} \quad (8)$$

where the interaction strength  $U_4$  is given by

$$U_4 = \left( \frac{K_4 \hbar^2}{M^2 \omega_0^2} \right) \quad (9)$$

and the form factor  $F_k$  is given by

$$F_k = \frac{\sin \frac{k_j}{2}}{\sqrt{|\sin \frac{k_j}{2}|}}. \quad (10)$$

115 Since the Hamiltonian does not commute with the phonon number operator, the energy eigenstates do not correspond to a fixed number of phonons. One expects that

in the absence of a phase transition, the ground state  $|\Phi_0\rangle$  should adiabatically evolve from the vacuum state  $|0\rangle$  as the strength of the anharmonic interaction is increased. Due to the quartic form of the interaction, one expects that the ground state will have non-zero components which correspond to the presence of multiples of two (harmonic) phonons.

### 3. The $n=2$ ILM excitations

As shown previously [29,30], the  $T=0$  spectra of  $n=2$  excitations consists of a continuum of weakly interacting two-phonon creation and annihilation excitations with center of mass momentum  $q$ , and a discrete branch of ILMs, which are pushed above the top edge of the continuum. The branch of ILMs is marked by the filled blue markers and is shown in Figure 2 for  $U_4/\hbar\omega_0 = 1$  (circles), 2 (squares) and 4 (triangles). The discrete nature of the quantized mode is consistent with the sharpness of the anomalous zone boundary mode observed in NaI via inelastic neutron scattering measurements [32]. The excitation energy of the anomalous mode is intermediate between the acoustic and optic modes which is also in agreement with our theory [29,30]. The continuum of excitations exhibits a branch of resonances just above the line  $\omega = 2\omega_{\frac{q}{2}}$ . The branch of resonances is marked by the filled red circles. The branches of resonances and the infinitely long-lived ILMs hybridize as  $q$  approaches the zone boundary  $q = \pi$ , see Figure 2. To demonstrate the localized nature of the ILM, we look for approximate eigenstates with momentum  $q$  that satisfy the eigenvalue equation

$$\hat{H}|\Psi_q\rangle = E_q|\Psi_q\rangle. \tag{11}$$

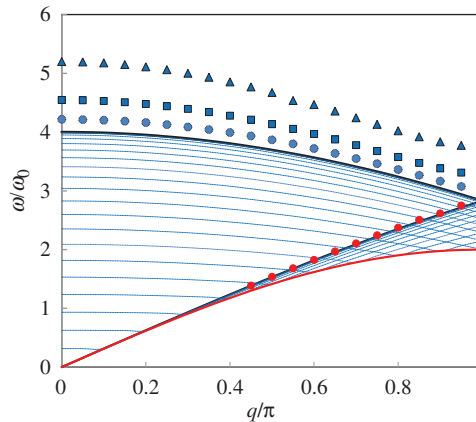


Figure 2. The  $(\omega, q)$  phase space which shows the extent of the continuum of two-phonon creation (shaded area). The lower edge of the continuum is marked by the one-phonon dispersion relation and is shown by the red line. The dispersion relation for the  $n=2$  resonance follows the band extremum at  $\omega = 2\omega_{\frac{q}{2}}$  (red circles). The top edge of the two-phonon continuum is located at  $\omega = 2\omega_{\frac{\pi+q}{2}}$ . The anharmonic interaction pushes the  $n=2$  ILM (filled markers) to energies above the top of the continuum.

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In the limit  $T \rightarrow 0$ , the approximate eigenstates simplify and have the form<sup>2</sup>

$$|\Psi_q\rangle = \sum_k (C_{k,q}^{(++)} a_{\frac{q}{2}+k}^\dagger a_{\frac{q}{2}-k}^\dagger + C_{k,q}^{(--)}) a_{-\frac{q}{2}-k} a_{-\frac{q}{2}+k} |\Phi_0\rangle \quad (12)$$

where the coefficients  $C_{k,q}^{(++)}$  and  $C_{k,q}^{(--)}$  have still to be determined and the summation over  $k$  is restricted to positive values. The state  $|\Phi_0\rangle$  is the ground state which satisfies

$$\hat{H}|\Phi_0\rangle = E_0|\Phi_0\rangle. \quad (13)$$

The unknown coefficients are determined by taking matrix elements of the eigenvalue equation for  $|\Psi_q\rangle$  with the states

$$a_{\frac{q}{2}+k}^\dagger a_{\frac{q}{2}-k}^\dagger |\Phi_0\rangle \quad (14)$$

and

$$a_{-\frac{q}{2}-k} a_{-\frac{q}{2}+k} |\Phi_0\rangle. \quad (15)$$

After commuting the Hamiltonian with the pair of operators, and renormalizing the phonon frequencies to lowest order [31] via

$$\omega_q \rightarrow \omega_q \left[ 1 + \left( \frac{U_4}{2\hbar\omega_0} \right) \frac{1}{N} \sum_k \left| \sin \frac{k}{2} \right| (1 + 2N_k) \right] \quad (16)$$

this procedure leads to the set of equations

$$(E_q - E_0 - \hbar(\omega_{\frac{q}{2}+k} + \omega_{\frac{q}{2}-k})) C_{k,q}^{(++)} = \langle \Phi_0 | [a_{\frac{q}{2}+k} a_{\frac{q}{2}-k} \hat{H}_{\text{int}}] | \Psi_q \rangle \quad (17)$$

and

$$(E_q - E_0 + \hbar(\omega_{\frac{q}{2}+k} + \omega_{\frac{q}{2}-k})) C_{k,q}^{(--)}) = \langle \Phi_0 | [a_{-\frac{q}{2}-k}^\dagger a_{-\frac{q}{2}+k}^\dagger \hat{H}_{\text{int}}] | \Psi_q \rangle. \quad (18)$$

On approximating the expectation value of the commutators involving the interaction, we find that the coefficients are given by the coupled equations

$$C_{k,q}^{(++)} = \frac{2U_4 (1 + N_{\frac{q}{2}+k} + N_{\frac{q}{2}-k}) F_{\frac{q}{2}+k} F_{\frac{q}{2}-k}}{N (E_q - E_0 - \hbar(\omega_{\frac{q}{2}+k} + \omega_{\frac{q}{2}-k}))} \sum_{\pm, k'} F_{\frac{q}{2}+k'}^* F_{\frac{q}{2}-k'}^* C_{k,q}^{(\pm\pm)} \quad (19)$$

and

$$C_{k,q}^{(--)}) = -\frac{2U_4 (1 + N_{\frac{q}{2}+k} + N_{\frac{q}{2}-k}) F_{\frac{q}{2}+k} F_{\frac{q}{2}-k}}{N (E_q - E_0 + \hbar(\omega_{\frac{q}{2}+k} + \omega_{\frac{q}{2}-k}))} \sum_{\pm, k'} F_{\frac{q}{2}+k'}^* F_{\frac{q}{2}-k'}^* C_{k,q}^{(\pm\pm)}. \quad (20)$$

In the above expressions, the quantities  $N_q$  are the temperature-dependent Bose-Einstein distribution functions. We have neglected the temperature-independent corrections to the phonon modes since they generate corrections proportional to

$$-\frac{\partial}{\partial\omega} \left[ \left( \frac{U_4}{N} \right)^2 \sum_{k_1, k_2} \frac{|F_q|^2 |F_{k_1}|^2 |F_{k_2}|^2 |F_{q-k_1-k_2}|^2}{(\omega + \omega_{k_1} + \omega_{k_2} + \omega_{k_1+k_2-q})} \right] \Big|_{\omega=\omega_q}. \quad (21)$$

155 The above correction terms were neglected since they are recognized as partially canceling with similar terms originating from the renormalizations of the phonon energies due to processes in which the total boson occupation number changes by two. Consistency requires that the excitation energy  $E_q - E_0$  be given by the solution of the equation

$$1 = \frac{U_4}{N} \sum_k |F_{\frac{q}{2}+k}|^2 |F_{\frac{q}{2}-k}|^2 \left[ \frac{(1 + N_{\frac{q}{2}+k} + N_{\frac{q}{2}-k})}{E_q - E_0 - \hbar(\omega_{\frac{q}{2}+k} + \omega_{\frac{q}{2}-k})} - \frac{(1 + N_{\frac{q}{2}+k} + N_{\frac{q}{2}-k})}{E_q - E_0 + \hbar(\omega_{\frac{q}{2}+k} + \omega_{\frac{q}{2}-k})} \right] \quad (22)$$

160 where now the summation runs over all the positive and negative values of  $k$ . The above equation always has solutions for repulsive interactions  $U_4$ , no matter how small the interaction is. The existence of ILMs at arbitrarily small interaction strengths is associated with the one-dimensional van Hove singularities of the lattice and is not expected to hold in higher dimensional systems. The  $T=0$  two-particle field operator for the state can be written as the product of a center of mass wavefunction and the relative wave operator

$$\hat{\Psi}_q(r_i, r_j) = \frac{1}{\sqrt{N}} \exp \left[ iq \left( \frac{r_i + r_j}{2} \right) \right] (\hat{\psi}_q^+(r_i - r_j) + \hat{\psi}_q^-(r_i - r_j)) \quad (23)$$

where the two parts represent the two-phonon creation (the large component) and two-phonon annihilation (the small component) contributions respectively. The two components of the unnormalized relative wavefunction have opposite phases and are given by

$$\psi_q^+(r_i - r_j) = \frac{1}{N} \sum_k \frac{\cos k(r_i - r_j) F_{\frac{q}{2}+k} F_{\frac{q}{2}-k}}{E_q - E_0 - \hbar(\omega_{\frac{q}{2}+k} + \omega_{\frac{q}{2}-k})} \quad (24)$$

170 and

$$\psi_q^-(r_i - r_j) = -\frac{1}{N} \sum_k \frac{\cos k(r_i - r_j) F_{\frac{q}{2}+k} F_{\frac{q}{2}-k}}{E_q - E_0 + \hbar(\omega_{\frac{q}{2}+k} + \omega_{\frac{q}{2}-k})}. \quad (25)$$

Both the components of the ILM wavefunctions are symmetric under the interchange of  $r_i$  and  $r_j$ , which is as expected for excitations that satisfy Bose-Einstein statistics. The normalized wavefunctions are shown in Figures 3 and 4. It should be recognized that the wavefunctions are only defined at the lattice sites but to help with visualization, the markers have been joined by continuous lines. It is seen that for most  $q$  values, the ILM state changes sign on alternate lattice sites, and has a decaying envelope. The alternating phase is a consequence of the ILMs being primarily composed of states from the upper edge of the two-phonon creation continuum for which  $k = \pi$ . The localization length  $\xi$  which governs the decay of the envelope is determined by the energy separation between the ILM and the top of the two-phonon continuum, i.e.

$$\xi^{-2} \propto E_q - E_0 - 2\hbar\omega_{\frac{2\pi+q}{2}}. \quad (26)$$

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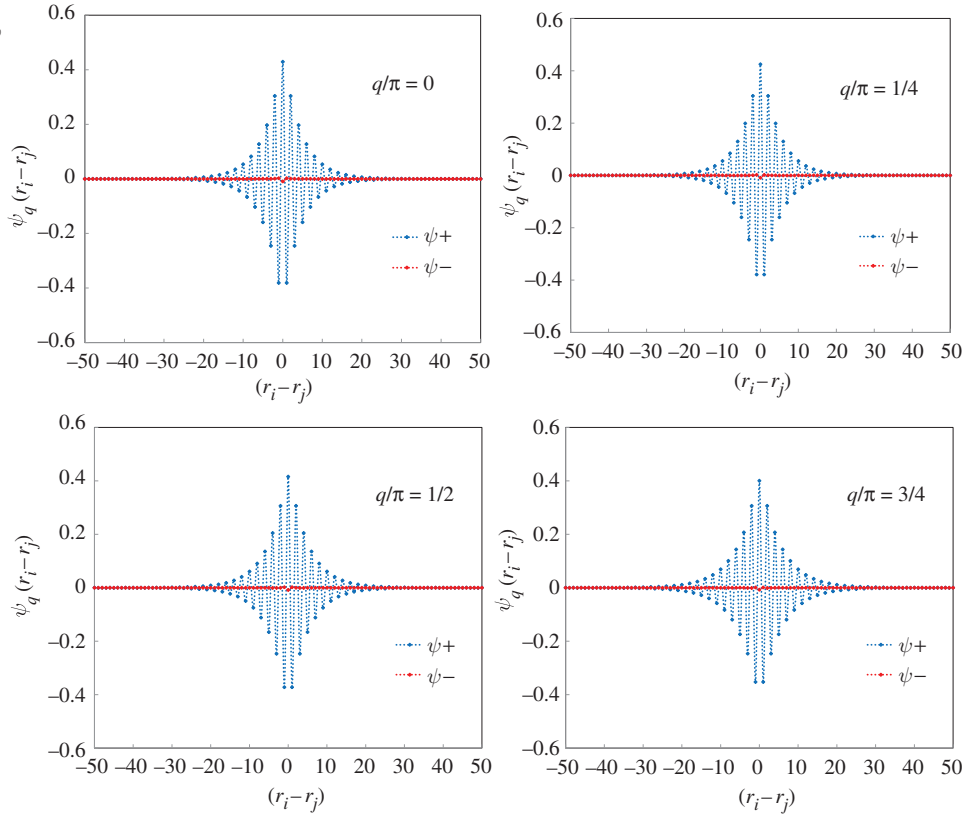


Figure 3. The two-photon creation  $\psi_q^+(r_i - r_j)$  and two-photon annihilation  $\psi_q^-(r_i - r_j)$  components of the ILM wavefunction, evaluated for  $U_4/\hbar\omega_0 = 0.25$ , are shown for the various values of  $q$  marked in each panel.

It should also be noted that the localization length  $\xi$  only has a slight dependence on the value of the center of mass momentum. This is unlike the situation for the breather excitations of the classical continuous sine-Gordon theory, where the exact solution shows that the length of the breather excitation undergoes a Lorentz contraction, for which  $c$  is the speed of sound. For the Fermi–Pasta–Ulam model which is not Lorentz covariant, the difference can be traced to the dominant spatial scale being set by the anharmonicity. In Figure 4 it is seen that the magnitude of the two-photon annihilation component increases as  $U_4$  increases. The vanishing of the two-photon annihilation component in the limit  $U_4 \rightarrow 0$  is consistent with the conservation of the number of phonons in the harmonic limit. Inspection of the wavefunction shown in Figure 5 shows that for  $q = \pi$ , the breather ILM wavefunction is zero for pairs of sites separated by an odd number of lattice spacings. This is a result of the hybridization of the resonance mode (which is composed primarily from states with  $k \approx 0$ ) and the ILM for  $q$  values near the zone boundary. At the zone boundary, the hybridization results in the destructive interference on alternate lattice sites.

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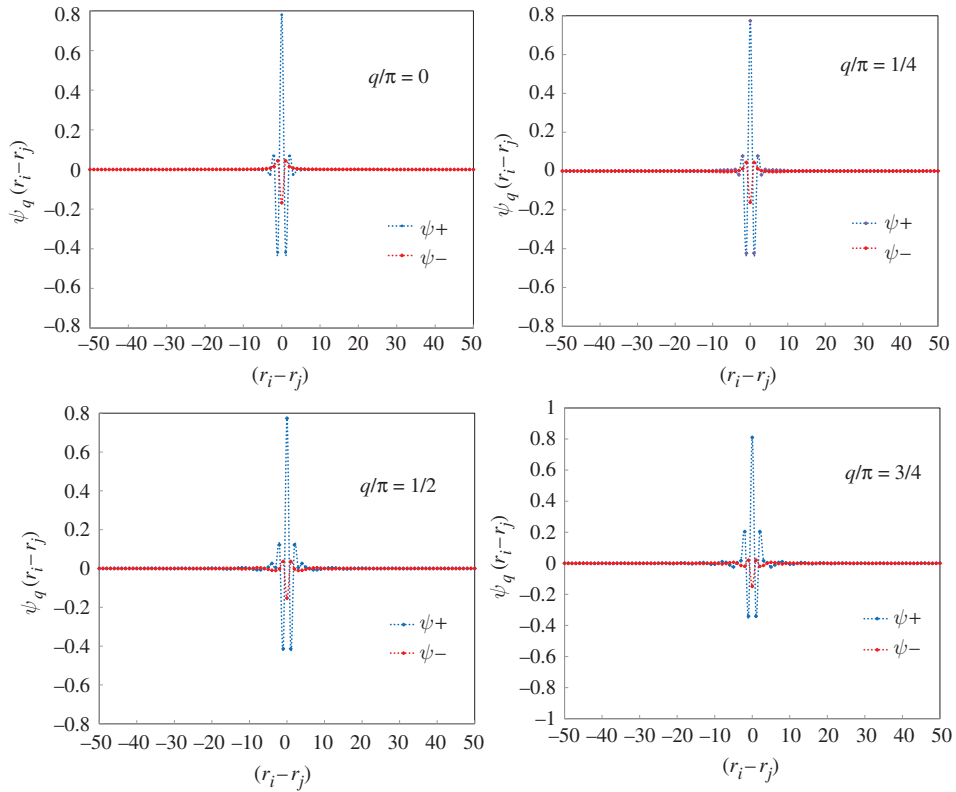


Figure 4. The two-photon creation  $\psi_q^+(r_i - r_j)$  and two-photon annihilation  $\psi_q^-(r_i - r_j)$  components of the ILM wavefunction, evaluated for  $U_4/\hbar\omega_0 = 4.0$  are shown for various values of  $q$ .

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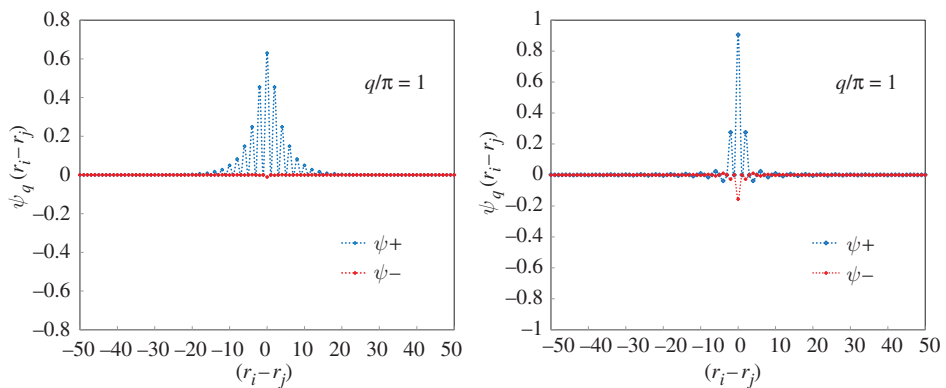


Figure 5. The two-photon creation  $\psi_q^+(r_i - r_j)$  and two-photon annihilation  $\psi_q^-(r_i - r_j)$  components of the ILM wavefunction, evaluated at the Brillouin zone boundary  $q = \pi$  are shown for  $U_4/\hbar\omega_0 = 0.25$  (left panel) and  $U_4/\hbar\omega_0 = 4.0$  (right panel).

#### 4. Discussion

In summary, we have shown that the quantized Fermi–Pasta–Ulam lattice supports intrinsically nonlinear excitations that exist as either discrete long-lived modes or resonances. By contrast, the ILMs of the classical lattice are expected to form continua. The discrete spectrum of the quantized ILM is consistent with experimental observations on NaI [32]. Although similar anomalous excitations have been reported [33–36] in the vibrational spectra of  $\alpha$ -uranium, the temperature dependence of the excitations is not consistent with expectations of a dilute gas of ILMs. An alternate description of the excitations in  $\alpha$ -uranium has been proposed [37,38]. Here we found that the quantized excitations split from the top of the two-phonon creation continuum, no matter how small the repulsive interaction is. The excitation energy of the quantized ILM shows dispersion indicating that they are mobile excitations [39], whereas it is believed that classical ILMs are immobile [40]. For infinitesimal strength of the anharmonic interaction, the localization length of the ILM is of the order of the size of the system but decreases rapidly as the strength of the interaction increases. As  $q$  approaches the Brillouin zone boundary, the resonance and the breather hybridize and subsequently exchange identity as  $q$  is further increased into the next Brillouin zone. The degeneracy is manifested in the many-body wavefunction by a destructive interference on alternate sites of the lattice.

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#### Notes

1. The ground state is degenerate under the continuous transformation  $u_n \rightarrow u_n + \delta$ , and therefore the assumed spontaneous symmetry breaking leads to the occurrence of Goldstone modes.
2. At finite temperatures, this ansatz should be generalized to include terms which involve a product of one creation and one annihilation operator. The ansatz reproduces the exact solutions for the two-boson excitations of models in which the total boson occupation number is a conserved quantity.

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