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Efficient anharmonic phonon generation using a quasiperiodic lattice

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Abstract

The effects of anharmonic interactions on the phonon modes in quasiperiodic systems are studied by looking at the time-evolution Fourier spectrum. The results reveal an efficient generation of high-amplitude harmonics in Fibonacci chains. We also perform an energy-level-spacing analysis of the spectrum. Anharmonic contributions do clearly manifest by changing the level clustering behavior observed in quasiperiodic chains, and contrary to the periodic case, where the distribution is insensitive to weak anharmonic interactions. This result suggests a structural instability of the self-similar vibrational spectrum in quasiperiodic systems.

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The study of non-linear effects in materials has been subject of intense research in the last decade [1]. From the theoretical point of view there are many important questions that are still open, and at the same time many important applications have been found, for example, the generation of optical harmonics using a quasiperiodic superlattice [2]. An interesting question is how non-linearity affects the localization and the transport properties of a system [3]. For instant, it is known that non-linear interactions are essential to explain the finite thermal conductivity of a material. A common belief is that in periodic systems, where the phonons have extended wave functions, the addition of a non-linear term to the Hamiltonian leads to localization. This type of localization is called intrinsic [1] and is not produced by disorder, as in the linear Anderson Hamiltonian [4]. These localized modes are also known as discrete breathers [5], and have been observed in different physical systems ranging from electronic and magnetic solids, Josephson junctions,

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optical waveguides arrays and laser-induced photonic crystals [1]. From these considerations, it is natural to expect that when both non-linearity and disorder are present, localization is enhanced. However, the situation is much more complex since several one-dimensional models with correlated disorder exhibit delocalized states [6], which have recently been confirmed using microwave waveguides [7].

Another fascinating question is what happen in quasiperiodic systems, since they are not randomly disordered neither periodic, instead they have a long-range order. A quasiperiodic potential can be described as a Fourier series with more reciprocal basis vectors than the dimensionality of the system [8]. For example, the two-dimensional Penrose lattice can be obtained by projecting a five-dimensional hypercubic lattice into a plane. As a consequence, its reciprocal space is generated by five reciprocal basis vectors [9]. In real systems, quasiperiodicity can occur at an atomic level as in quasicrystalline metallic alloys [10], or can be found in artificial superlattices, for example, by alternating layers of GaAlAs and GaAs following a Fibonacci sequence [11,12]. Recently, it has been found that quasiperiodic multilayers are useful for efficient generation of optical second and third harmonics [2]. This kind of multilayers can be treated theoretically as one-dimensional systems [13]. However, up to our knowledge, the anharmonic effects in quasiperiodic systems have not been studied systematically. For instance, there is an experimental observation of a softening of the eigenmodes in an equivalent electric circuit [14]. In this Letter, we further pursue this problem by looking at the phonon spectrum of a quasiperiodic system with anharmonic interactions.

Let us consider the most simple quasiperiodic system, known as the Fibonacci chain (FC), in which two kinds of atoms, A and B, are arranged following the Fibonacci sequence [8], i.e., if one defines the first generation $F_1 = A$ and the second one $F_2 = BA$, the subsequent generations are given by $F_n = F_{n-1} \oplus F_{n-2}$. For instance, $F_5 = BAABABAA$. In a FC, the spring strength between atoms depends on their nature, giving two different force constants β_{AA} and $\beta_{AB} = \beta_{BA}$. Thus, the phonon dynamics of a FC, including a quartic anharmonic term in the phonon Hamiltonian, can be described by

$$m_j \frac{d^2u_j}{dt^2} = \beta_j(u_{j+1} - u_j) - \beta_{j-1}(u_j - u_{j-1}) + \eta(u_{j+1} - u_j)^3 - \eta(u_j - u_{j-1})^3, \quad (1)$$

where m_j can either be m_A or m_B, and u_j is the displacement of atom j at time t. The anharmonic interaction parameter η is supposed to be smaller than the harmonic interaction β_j and then is taken as a constant. In this Letter, we set the units of mass $m_A = m$ and for spring constant $\beta_{AA} = \beta$. Usually, to obtain the stationary solutions of non-linear equations (1), the well known rotating-wave approximation is used [15]. In a previous article, we used this method in conjunction with the perturbation theory to study the FC. However, this approximation only predicts a shift of the original frequencies [16]. Also, there was no estimation of the accuracy of the solution. To go beyond this approximation, in the present work we use a totally different approach. We solved the equations of motion (1) by using a discrete time $\Delta t = 0.1/\omega_0$ to optimize the efficiency of the simulation, where $\omega_0 = \sqrt{\beta/m}$. At each time we calculated the force, velocity and acceleration of the atoms in the chain, using the leap-frog integration method [17]. Periodic boundary conditions were used for chains of $N_s = 2584$ sites. As initial conditions, we chose zero velocity for all atoms, i.e., $v_j(0) = 0$, and a delta function at a site in the chain, for example, $u_j(0) = a\delta_{j,1}$, where a is the interatomic separation. This delta function is a superposition of all normal modes and its Fourier transform is just a flat function in the reciprocal space. As a consequence, one is sure from the beginning of exciting all modes with the same weight for periodic systems, avoiding the mode recurrence problem that was found after the pioneering work of Ulam–Fermi–Pasta [18]. Once the chain was excited with this initial condition, the system was allowed to evolve until it reached a number of time steps $N_t = 2^l$, where l is an integer that usually had the value 24, determined by the largest integer that the computer can handle. Then, we took the fast Fourier transform of the whole set of displacements $u_j(t)$

$$U(\omega) \equiv \sum_{j=1}^{N_s} \left\| \frac{1}{N_t} \sum_{t=0}^{t_{\max}} u_j(t) e^{i\omega t} \right\|, \quad (2)$$

where $t_{\max} = N_t \Delta t$.

To test the procedure, we use a periodic chain ($\beta_{AB} = \beta_{AA} = \beta$ and $m_A = m_B = m$) with only har-

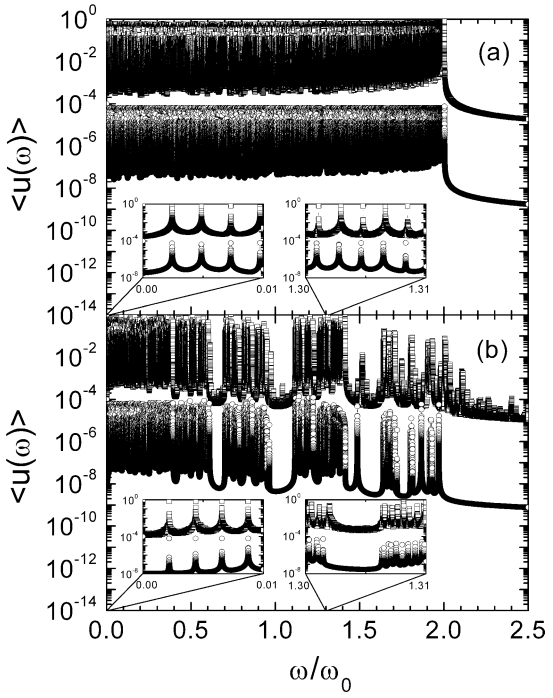

Fig. 1. Fourier spectrum of the displacements with (open squares) and without (open circles) non-linear interaction ($\eta = 0.2\beta$) for (a) a periodic chain, and (b) a Fibonacci chain with $\beta_{AB} = \beta_{AA}/2$, both containing 2584 atoms. Insets: amplifications of two regions of each spectrum. In all the figures, the harmonic data have been vertically shifted by a factor of 10^{-4}.

monic interactions, and its spectrum ($U(\omega)$) is shown in Fig. 1(a) as open circles, where the peaks are located exactly at the same frequencies as those obtained from the diagonalization of the dynamic matrix or from the analytical dispersion relation, i.e., $\omega(k) = 2\omega_0 \sin(ka/2)$, being the wave vector $k = 2\pi n/N_s a$, with $n \in \mathbb{Z}$. Notice that there is a peak at zero frequency that can be deduced by integrating the momentum conservation and using the initial conditions

$$\sum_{j=1}^{N_s} u_j(t) = a. \tag{3}$$

Applying a Fourier transform to the previous equation, we get

$$\sum_{j=1}^{N_s} \tilde{u}_j(\omega) = a\delta(\omega), \tag{4}$$

where

$$\tilde{u}_j(\omega) \equiv \sum_{t=0}^{t_{\max}} u_j(t) e^{i\omega t}, \tag{5}$$

and

$$\delta(\omega) = \sum_{t=0}^{t_{\max}} e^{i\omega t}. \tag{6}$$

The last function becomes a Dirac delta in the limit $t_{\max} \to \infty$. In particular, for $\omega = 0$, this function gives $\delta(0) = N_t$. Considering the nature of the zero frequency mode, i.e., $\tilde{u}_j(0)$ is a positive constant due to the initial conditions, the summation of displacements for the zero frequency mode is

$$U(0) = \frac{1}{N_t} \sum_{j=1}^{N_s} \left\| \tilde{u}_j(0) \right\| = a. \tag{7}$$

In general, for the case of harmonic Hamiltonians, the height of each peak in the spectrum can be easily obtained by a decomposition of the solution in normal modes of frequency ω_α, which leads to

$$U(\omega) \equiv \sum_{\alpha=1}^{N_s} U(\omega_\alpha)\delta(\omega - \omega_\alpha), \tag{8}$$

where

$$U(\omega_\alpha) \equiv \sum_{j=1}^{N_s} \| A_{j\alpha} \| \tag{9}$$

is the height of the peak with frequency ω_α in the spectrum, and the coefficients $A_{j\alpha}$ are obtained by a projection of the initial conditions onto each normal mode. For the periodic case, our initial conditions conduct to

$$U(\omega_\alpha) = \frac{a}{N_s} \sum_{j=1}^{N_s} \left| \cos\left[2\pi\alpha(j-1)/N_s\right] \right|. \tag{10}$$

In the limit $N_s \to \infty$, the value of $U(\omega_\alpha)$ is $2a/\pi$, which is in agreement with our numerical results (open circles in Fig. 1(a)), where it is important to remark that in the data from the harmonic systems have been vertically shifted by a factor of 10^{-4} in order to improve the comparison with the non-linear cases.

For non-linear Hamiltonians, we first tested the procedure by using a three-atom chain, where the

analytical results obtained from perturbation theory [18] show a good agreement with the numerical ones. Then, we introduce a small non-linear term ($\eta = 0.2\beta$) to Eq. (1) for a periodic chain of 2584 atoms, and the spectrum remains almost equal (open squares in Fig. 1(a)). A blown-up of the acoustic region shows that the main effect is just a shift of the peaks towards higher frequencies. This shift is more evident at the middle of the spectrum, as shown in the insets of Fig. 1(a). These shifts in frequency can be predicted within the rotating-wave approximation [16]. For the case of an harmonic FC with $m_B = m$ and $\beta_{AB} = \beta/2$, open circles in Fig. 1(b), we got exactly the same eigenfrequencies than those obtained from a diagonalization of the Hamiltonian. The shape of this spectrum is known to be a multifractal set [19], and corresponds to a singular continuous spectra, with critically localized normal modes [20]. This kind of spectrum has gaps almost everywhere since the reciprocal space of a quasicrystal is a dense set. Open squares in Fig. 1(b) show the spectrum of the same FC including a small non-linear term ($\eta = 0.2\beta$). In this case, the effect is dramatic in the sense that not only the original harmonic peaks are shifted, but also new peaks appears inside the gaps of the spectrum. The intensity of these new peaks is in general two orders of magnitude larger than those found in the periodic case. Furthermore, for an anharmonic FC, the spectral limit is extended well beyond the case of a pure harmonic FC. From this analysis, it is clear that the FC is much more efficient for generating high order harmonics. A simple explanation for this phenomena is given by the structure of the reciprocal space of a quasicrystal. Quasiperiodic systems can always be described as a succession of rational approximants, this succession tends to the quasiperiodic system in the limit of an infinite unitary cell [8]. Each approximant has bands, well defined wave vectors (k), and a dispersion relation of the type $\omega_\gamma(k)$, where γ is the band index. Now, if we look to the quartic anharmonic interaction as a phonon collision, where two phonons with wavevectors k_1 and k_2 annihilates to form two new phonons with wave vectors k_3 and k_4, then, for each process the energy is conserved, $\omega_{\gamma_1}(k_1) + \omega_{\gamma_2}(k_2) = \omega_{\gamma_3}(k_3) + \omega_{\gamma_4}(k_4)$, as well as momentum, $k_1 + k_2 + k_3 + k_4 = G_{m,s}$, where for a FC $G_{m,s} = 2\pi\langle\lambda\rangle^{-1}(m + s\tau)$ is a vector in the reciprocal space, $\tau = (1+\sqrt{5})/2$, $m, s \in \mathbb{Z}$, and $\langle\lambda\rangle$ is the average lattice parameter [21]. If the lattice momentum is $G_{m,s} = 0$, the process is known as normal. Processes with $G_{m,s} \neq 0$ are responsible for thermal conductivity and are known as umklapp. In a quasicrystal, $G_{m,s}$ covers in a dense way the reciprocal space [8], thus almost all processes are umklapp. In fact, there is always a $G_{m,s}$ as close as needed and then momentum conservation is not a restriction anymore. The only remaining condition is energy conservation. As a result, three of these wave vectors can be chosen without any restriction, making the system much more efficient to generate harmonics.

A useful way to extract information from the spectrum is to do level spacing statistics, since there is a close relationship between the nature of localization and its eigenvalue statistics [22], e.g., for a disordered metal, a Wigner distribution $P_W(s) \sim s\exp(-\pi s^2/4)$ is observed, while for a disordered insulator it becomes a Poisson's law $P_P(s) \sim \exp(-s)$, where $s = |\omega_{i+1} - \omega_i|/\Delta$ is the frequency difference of consecutive peaks in the spectrum divided by the mean level spacing Δ. The essential difference between these two distributions arises from their small s behavior. Recently, a new kind of level statistics has been found in one-dimensional quasiperiodic systems, that shows a power law behavior, revealing a level clustering mechanism [23]. To do the spectral statistics, in principle one needs to find the phonon eigenvalue spectrum, and then obtain the level spacing statistics through an unfolding process [22]. A very important fact is that the level statistics do not care about the height of each frequency peak in the Fourier spectrum. In order to include this fact, we made cutoffs at different heights in the spectra of Figs. 1(a) and (b), as shown, respectively, in Figs. 2 and 3, where the unnormalized $P(s)$ are presented to show the number of harmonics created for each level spacing. When the cutoff is high, we only get the most prominent peaks, while for a cutoff at zero all the peaks are considered. Comparing Figs. 2(a) and 3(a), it is observed that the difference between the harmonic (open circles) and anharmonic (open squares) spectra is larger in the FC than in the periodic chain, as obtained in an analysis based on the rotating-wave approximation [16]. Nevertheless, in the present analysis when a non-linear interaction is introduced in the FC, $P(s)$ grows for small s, which means a tendency towards level clustering and an efficient creation of new peaks, contrary to the results obtained from the rotating-wave approx-

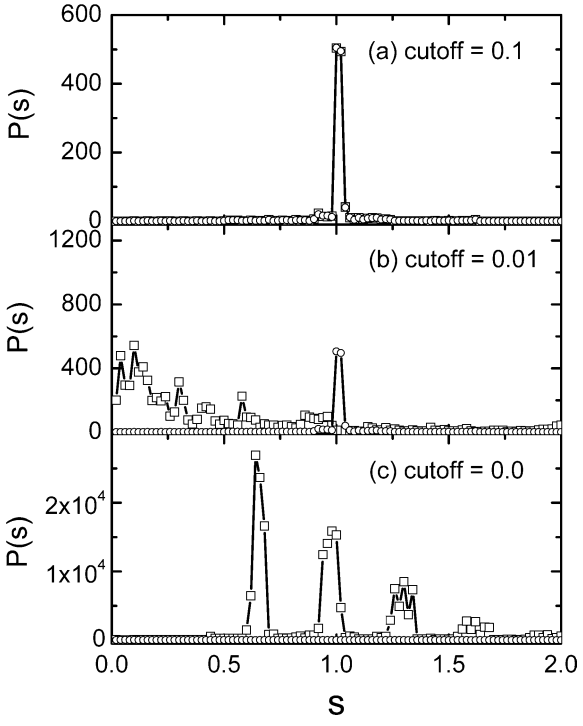

Fig. 2. Level-spacing distribution $[P(s)]$ with unfolding for the same periodic chain as in Fig. 1, using cutoffs in the displacement spectrum of (a) 0.1, (b) 0.01, and (c) 0.0. The open circles correspond to non-linear interaction $\eta = 0$ and open squares to $\eta = 0.2\beta$.

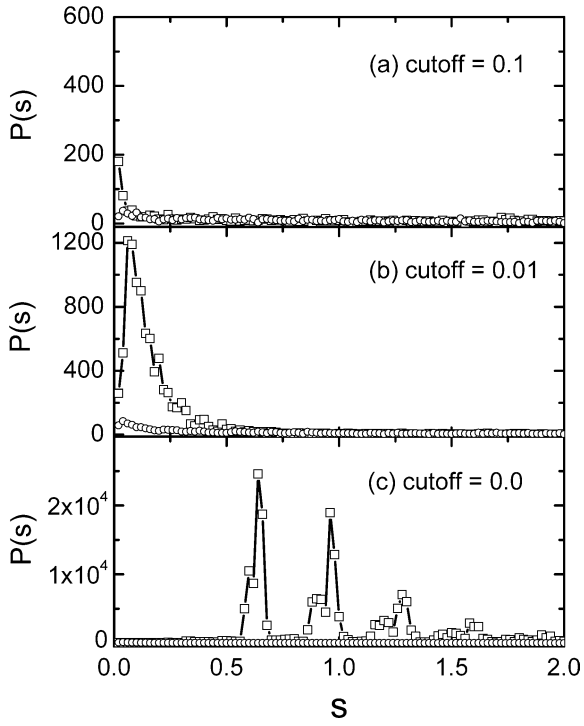

Fig. 3. Level-spacing distribution $[P(s)]$ with unfolding for the same FC as in Fig. 1, using cutoffs in the displacement spectrum of (a) 0.1, (b) 0.01, and (c) 0.0. The open circles correspond to non-linear interaction $\eta = 0$ and open squares to $\eta = 0.2\beta$.

imation, where the total number of resonance frequencies remains the same since they are only shifted from their original positions. When the cutoff is decreased to 0.01, Figs. 2(b) and 3(b), more anharmonic modes with high amplitude are observed in the FC, which supports the hypothesis of an efficient generation of harmonics. Finally, Figs. 2(c) and 3(c) show a similar $P(s)$, where a series of peaks separated by a regular distance are obtained. This happens because the number of peaks is of the same order of magnitude than the grid points of the frequency and then, the peaks are separated by few grids spacings. In our case, the count of peaks is almost 2.1×10^5 peaks and the partition of time produces a discretization in ω of size $\Delta\omega = 2\pi/(N_t \Delta t)$, i.e., there are 6.6×10^5 frequency partitions in the interval $[0, 2.5\omega_0]$. One would expect that this feature could be removed by increasing N_t. To test this idea, we have performed a calculation using a $N_t = 2^{25}$ and surprisingly the results remain essentially unchanged. This fact could be due to the fractal nature of the attractors in both periodic and quasiperiodic systems when anharmonic interactions are present [18].

In summary, we have studied the anharmonic vibrational modes in quasiperiodic systems beyond the rotating wave approximation. The results show an efficient generation of harmonics with high amplitude in the FC compared with the periodic case. The level spacing statistics reveals that the self-similar vibrational spectrum of the FC is unstable upon anharmonic interactions. This fact could be important in the understanding of the mode softening in quasiperiodic electric circuits [14], and the lack of clear observations of self-similar spectra in real quasicrystals [8].

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