Cross-correlations between phonon modes in anharmonic oscillator chains: Role in heat transport

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We have computed current-current correlation functions in chains of anharmonic oscillators described by various models (FPU-β, FPU-$\alpha\beta$, ϕ^4), considering both the total current and the currents associated with individual phonon modes, which are important in view of the Green-Kubo relation for heat conductivity. Our simulations show that, contrary to the common hypothesis, there are, under some circumstances, significant correlations between neighboring modes. These cross-mode correlations are the dominant contribution to the conductivity in the low anharmonicity regime. The inverse of the timescale over which they are significant, $1/\tau_c$, is related to the anharmonicity level in a way similar to the largest Lyapunov exponent, suggesting that the two quantities are related. Cross-mode correlations exist in both anomalous and regular heat-conducting systems although we are unable to observe a transition to the independent-mode regime in the latter case.

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I. INTRODUCTION

The problem of heat transport in anharmonic chains has received much attention in the last decade or so, largely stimulated by its relevance to the growing nanotechnology industry. Many studies have already been published on this topic and there is now strong evidence that a wide variety of anharmonic oscillator chains display so-called anomalous heat conductivity, i.e., the heat flow does not follow Fourier's law.

The most striking feature of anomalous conductors is the nonintensive nature of the heat conductivity. Studies on this topic are often based on the relaxation time approximation and focus on the divergence of the relaxation time of long-wavelength modes arising from the reduced phase space available for scattering. Although this theoretical framework agrees relatively well with computations for some systems, the overall portrait is still somewhat unsatisfying. For instance, the exact conditions that yield anomalous conductivity are not yet known, and predicting in a quantitative way the heat transport properties associated with a given model is still out of reach. Conservation of momentum is thought to be an important condition for anomalous conduction, but recent studies [1,2] have sparked a new debate on the matter [2–6]. It has also been argued that, in anomalous systems, the heat conductivity $\kappa \propto N^{\alpha}$, where N is the number of oscillators in the chain and $0 < \alpha < 1$; there are reasons to believe that this exponent could be universal, with the most frequently cited values being 1/3 and 2/5, although others have been either predicted or observed [7–14]. Here, also, the situation has become less clear-cut recently, as some models seem to allow tuning α through the interaction parameters [15] or even going from anomalous to normal conduction by changing the anharmonicity [16]. More information on anomalous heat transport can be found in the reviews by Lepri *et al.* [17] and Dhar [18].

Despite the obvious connection provided by linear response theory, the relation between heat transport and the celebrated Fermi-Pasta-Ulam (FPU) problem [19] has not been much explored. Discovered in 1955, the "problem" refers to the very slow relaxation of one-dimensional anharmonic oscillator chains and other quasi-integrable classical systems. As discussed in Sec. III, the FPU Hamiltonian is a one-dimensional oscillator chain which may contain order 3 terms (FPU-α), or order 4 terms (FPU-β), or both (FPU-$\alpha\beta$), in addition to the harmonic terms. From its study, two important concepts, possibly having an impact on heat conduction, have emerged: first, Zabusky and Kruskal [20] showed that many nonlinear and dispersive systems display soliton or quasi-soliton solutions; and second, Izrailev and Chirikov [21] introduced the idea of resonance overlap as an explanation for the transition between slow and fast relaxation with increasing anharmonicity. Demonstrating that these concepts are also important for heat transport is one of the objectives of the present study.

While it is known that the FPU chain and many other models possess solutions in the form of localized disturbances, whether or not they form spontaneously and have long enough lifetimes to contribute to heat conduction is still a matter of debate. For example, Zhang *et al.* [22,23] studying the FPU-β model, found clear evidence that, in some circumstances, a soliton is formed, which absorbs most of the other excitations of the system, then goes on traveling indefinitely (under periodic boundary conditions). However, these simulations employed the Evans algorithm, in which a fictitious field imposes a heat flow, and it was found that the phenomenon only appears in the (probably unphysical) strong-field regime. On the other hand, Li *et al.* [24] and Likhachev *et al.* [25] computed the velocity of a traveling disturbance in the same model over a broad temperature range and found very good agreement with the theoretical speed of sound of renormalized phonons and less good with the velocity of Korteweg–De Vries solitons. Nonetheless, Likhachev *et al.* [25] were able to isolate three types of solitary-wave solutions from a thermalized sample by connecting it to a cold sample. Xiong *et al.* [15] found that introducing interactions with second neighbors leads to the formation of discrete breathers and it was conjectured that their interaction with phonons leads to nonuniversal behavior

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in anomalous conductivity. Given the contradictory evidence, it is certainly fair to say that, although phonons seem to be the main carriers, localized modes are quite likely to play a role in heat conduction.

A powerful way to probe the existence of localized modes, and possibly other effects, going beyond the traditional Debye-Peierls phonon gas model, is to compute phonon cross-correlations (correlations between different phonon modes). To our knowledge, there are only two such studies: first, Frizzera *et al.* [26] looked at correlations between normal modes in harmonic and anharmonic (FPU-$\alpha\beta$) chains with thermostats at both ends, presenting results mainly for momentum-momentum correlation functions $\langle P_k(t)P_{k'}(t)\rangle$. They showed that the differences in temperature profiles between harmonic and anharmonic chains arise from different cross-correlations, the diagonal correlation function ($k = k'$) leading only to a flat temperature profile. On the other hand—contrary to the methodology used in the present study and presented in the next section—these authors did not consider current-current correlations $\langle J_k(t)J_{k'}(t')\rangle$ and focused on nonequilibrium simulations only. Second, Henry and Chen [27] examined current-current correlations at equilibrium in a realistic model of a polyethylene chain. Thus, although the model is linear, it is not purely one-dimensional. Their results indicate that some initial conditions lead to the slow (anomalous) decay of correlations, which they ascribed to cross-correlations between midfrequency longitudinal acoustic modes. While this point of view is innovative, their use of a sophisticated model makes it hard to understand if this behavior follows from the linear nature of the model or from other features. Also, owing to the complexity of the model, the chains they consider are somewhat short (40 unit cells), and it is not clear if the statistical averaging was sufficient in view of the extremely slow convergence of such systems. Our objective here is to use a similar approach but, using a simpler model, try to investigate the matter in more depth and see how it relates to anomalous thermal conductivity.

The outline of this paper is the following: in Sec. II we describe the theoretical framework used to extract the correlation functions and thermal conductivities from molecular dynamics simulations. In Sec. III we present the technical details behind our numerical simulations, while our results are presented in Sec. IV. We conclude in Sec. V by summing up our results and giving some insight into how they could relate to other salient topics in heat transport theory.

II. THEORY

We study heat conductivity through equilibrium molecular dynamics simulations. This is done by computing the time-delayed correlations of the current fluctuations in a periodic system and employing the appropriate Green-Kubo relation, which in one dimension can be formulated as

$$\kappa = \frac{1}{k_B T^2 L}\int_0^\infty \langle J(0)J(t)\rangle dt, \qquad (1)$$

where κ is the heat conductivity, L is the length of the conductor, k_B is Boltzmann's constant, T is the temperature, $J(t)$ is the total heat flux, and $\langle\,\rangle$ denotes statistical averaging. Derivation of this formula can be found in advanced statistical mechanics textbooks, and general information on its use in molecular dynamics textbooks. When mass diffusion is negligible, the total flux in the chain [17] can be obtained using

$$J = \frac{a}{2m}\sum_{l=1}^{N}(p_{l+1}+p_l)F_{l+1,l}, \qquad (2)$$

where a is the lattice parameter, m is the mass of the particle, N is the number of particles, p_l is the momentum of the lth particle, and $F_{l+1,l}$ is the force on the $(l+1)$-th particle coming from the lth particle.

If the chain is harmonic or weakly anharmonic, it can be described using its normal-mode representation. We use the transformation

$$Q_k = \frac{1}{\sqrt{N}}\sum_{l=1}^{N} q_l e^{i(2\pi k/N)l}, \qquad (3)$$

$$P_k = \frac{1}{\sqrt{N}}\sum_{l=1}^{N} p_l e^{i(2\pi k/N)l}, \qquad (4)$$

where q_l is the displacement of the lth particle from its equilibrium position. There are N such modes, each denoted by a given k value; we use $k = -\frac{N}{2}-1,\ldots,\frac{N}{2}$. For a harmonic force $F_{l+1,l} = K(q_l - q_{l+1})$, and this yields the Hamiltonian

$$H = \frac{1}{2}\sum_k\left(\frac{P_k P_k^*}{m} + m\omega_k^2 Q_k Q_k^*\right). \qquad (5)$$

Each mode can thus be associated with a totally independent phonon state of eigenfrequency

$$\omega_k = 2\sqrt{\frac{K}{m}}\left|\sin\left(\frac{\pi k}{N}\right)\right|. \qquad (6)$$

Anharmonicity will modify this Hamiltonian, lead to energy exchange between the modes, and, as shown below, result in cross-mode correlations.

As q_l and p_l are real functions, $Q_k^* = Q_{-k}$ and $P_k^* = P_{-k}$, which enables expressing Eq. (2) as

$$J = ia\sum_k P_k Q_k^* v_k \omega_k = \sum_k J_k', \qquad (7)$$

where $v_k = \frac{\partial\omega_k}{\partial k}$ is the phonon group velocity. This last equation shows that the total harmonic flux can be written as a sum of currents associated with individual phonon modes. However, in this case J_k' is a complex number which is inconvenient to handle numerically. We thus use the fact that $J_{-k}' = (J_k')^*$ to make the transformation

$$J = \frac{1}{2}\sum_k (J_k' + J_{-k}') = \sum_k \mathrm{Re}(J_k') = \sum_k J_k, \qquad (8)$$

which implies the definition

$$J_k \equiv \frac{iav_k\omega_k}{2}(P_k Q_k^* - P_k^* Q_k). \qquad (9)$$

We calculate these mode currents by fast Fourier transforming the particle positions and velocities and using Eq. (6). We also computed velocity autocorrelation functions in some systems and obtained well-defined phonon modes in all cases.

Renormalized phonon frequencies were not used, as they would not modify our (mainly qualitative) interpretation. Inserting Eq. (9) into Eq. (1) yields

$$\kappa = \frac{1}{k_B T^2 L} \sum_k \sum_p \int_0^\infty \langle J_k(0) J_p(t) \rangle dt, \quad (10)$$

showing clearly that conductivity results from phonon self-mode correlations but also from cross-mode correlations.

A very interesting point made by Henry and Chen [27] is that using the hypothesis $\langle J_k(0)J_p(t)\rangle = 0$ for $k \neq p$ (i.e., neglecting cross-correlations) leads to the well-known result

$$\kappa = \frac{1}{L} \sum_k C_k v_k^2 \tau_k, \quad (11)$$

where C_k is the heat capacity associated with mode k and τ_k is its relaxation time. This hypothesis corresponds precisely to the *stosszahlansatz*, the view that there are no significant cross-correlations, which is central to the Debye-Peierls model. As we see in the following sections, it is clearly violated in many anharmonic chain models.

We define the diagonal contribution to the current-current correlation function

$$C_d(t) = \sum_k \langle J_k(0) J_k(t) \rangle, \quad (12)$$

as well as the off-diagonal contribution

$$C_{od}(t) = \sum_k \sum_{p \neq k} \langle J_k(0) J_p(t) \rangle, \quad (13)$$

which, in practice, we compute by subtracting $C_d(t)$ from the total current-current correlation function

$$C(t) = \langle J(0) J(t) \rangle = C_d(t) + C_{od}(t). \quad (14)$$

To assess the relative importance of the cross-mode correlations we introduce the "correlation ratio," *viz.*, the ratio of the total current-current correlation function to the diagonal contribution,

$$R(t) = \frac{C_{od}(t)}{C_d(t)} = \frac{C(t)}{C_d(t)} - 1, \quad (15)$$

which will equal 0 if the *stosszahlansatz* is satisfied and be positive if off-diagonal terms have a positive contribution to the conductivity.

It should be noted that, although our analysis is only concerned with the *harmonic* current, that is, the current coming from harmonic forces, in some of our simulations the anharmonic current is large. For example, it is about five times larger than the harmonic current when $\beta = 1$ and $T = 1$. Nonetheless, it has been shown before [7] that the anharmonic current in the FPU-β model is, at least in a statistical sense, proportional to the harmonic current. Also, in all our simulations, we compared the total current computed with the full anharmonic interaction using Eq. (2), and also using $\sum_k J_k$, and observed that the proportionality relation $J(t) = C \sum_k J_k(t)$, where C is a constant, is nearly exact in all cases and at all times. Our (qualitative) conclusions about the total harmonic current therefore also apply to the anharmonic current. While this does not directly inform us on the nature of the anharmonic currents stemming from single modes, it is highly plausible that the same ratio applies. Indeed, if we denote the total (harmonic plus anharmonic) current associated with mode k as $J_k^T(t)$, in any case we must have

$$\sum_k J_k^T(t) = C \sum_k J_k(t) = \sum_k f_k(t) J_k(t), \quad (16)$$

where $f_k(t)$ are unknown functions. Because the problem is nonlinear, these functions can take any value, but the most likely scenario is certainly $f_k(t) \approx C$ for all k's. Furthermore, even if this postulate were flawed, it would not change our general conclusions concerning the existence of off-diagonal correlations.

III. NUMERICAL DETAILS

We simulate periodically replicated chains of particles interacting with their nearest neighbors through anharmonic interactions in the *NVE* ensemble, integrating the equations of motion using a standard molecular dynamics algorithm. The motion of the particles is purely one-dimensional. Most of our simulations were carried out using the FPU model:

$$H = \sum_{l=1}^{N} \frac{p_l^2}{2m} + \frac{K}{2} \sum_{l=1}^{N} (q_{l+1} - q_l)^2 + \frac{\alpha}{3} \sum_{l=1}^{N} (q_{l+1} - q_l)^3 + \frac{\beta}{4} \sum_{l=1}^{N} (q_{l+1} - q_l)^4. \quad (17)$$

This Hamiltonian has four parameters: m, the particle mass, and three interaction parameters, K, α, and β (where β is *not* the inverse temperature). As we are simulating a fictitious model, we set $m = K = 1$. The other interaction parameters, α and β, are used to obtain the desired level of anharmonicity. The lattice parameter has no effect on the dynamics except as a trivial multiplier in expressions for the heat current. For this reason we set it at a sufficiently high value to make it impossible for particles to cross, but we set it to 1 when reporting current values.

We also considered the ϕ^4 model,

$$H = \sum_{l=1}^{N} \frac{p_l^2}{2m} + \frac{K}{2} \sum_{l=1}^{N} (q_{l+1} - q_l)^2 + \frac{\lambda}{4} \sum_{l=1}^{N} q_l^4, \quad (18)$$

where λ is another interaction parameter. Here also we set $m = K = 1$ and use λ to tune the anharmonicity. The two models are fundamentally different in that the FPU model is both energy and momentum conserving, while the ϕ^4 model is only energy conserving. Also, the fact that heat conduction is anomalous in the FPU model (at least for FPU-β) [7,9–14] while it is normal in the ϕ^4 model [28–30] is well documented.

In both cases, the parameters of the simulations include T, the average temperature of the system, which is set to 1. Scaling relations exist between α,β and T or between λ and T, but our tests indicate that keeping the interaction parameters fixed and changing the temperature does not fundamentally modify the results. The last parameter that defines the simulations is the number of particles in the chain, N. As the system is periodic, an increasing N leads to a larger number of vibration modes available to the system. In turn, as the number of modes increases, their frequencies get closer, leading to stronger interactions between them. In general, we are interested in the thermodynamic limit ($N \to \infty$) and it is therefore better to use as large an N value as practical, although comparisons between different lengths can be instructive as shown below. Unless otherwise specified, $N = 2048$ was used.

To integrate the equations of motion we use the sixth-order symplectic integrator presented in Appendix A of Lee-Dadswell *et al.* [13], with a time step of 0.3 reduced time unit (t.u.) for the cases $\beta,\alpha,\lambda < 0.4$ and a shorter time step for simulations with a higher anharmonicity, so as to ensure energy fluctuations of order 10^{-5} or better. The simulations proceed by first setting equilibrium positions and random velocities. Then a Langevin thermostat is applied for 3×10^6 t.u., followed by relaxation for another 3×10^6 t.u. using the symplectic integrator without the thermostat. After this, the actual production run begins and statistics are recorded. Statistical averages were carried out by averaging over the whole simulation time ($>3.5 \times 10^8$ t.u.) over 40–400 simulations with different initial random velocities. The mode currents J_k and their correlation functions are computed *a posteriori*.

IV. RESULTS

A. Off-diagonal correlations in the FPU-β model

We first examine the current-current correlation functions for the FPU-β model ($\alpha = 0$). Figure 1(a) shows the total correlation function $C(t)$ as well as the diagonal $C_d(t)$ and off-diagonal $C_{od}(t)$ contributions for $\beta = 0.01$, i.e., weak anharmonicity. As expected, $C_{od}(0) = 0$, reflecting the orthogonal nature of the modes. However, this quantity increases quickly and reaches a maximum after which it decays back to 0. All terms display a behavior which is consistent with a power law, with an exponent between 0 and 1. This is confirmed in Fig. 1(b), where all curves, plotted in a log-log way, are fairly linear for a moderately long time. Power-law fits were attempted on the three curves, and although the exponents depend quite sensitively on the time range considered, reasonably good fits were obtained for $\log_{10} t \in [4,5.5]$ with $C(t) \sim t^{-0.65}$, $C_d(t) \sim t^{-0.77}$, and $C_{od}(t) \sim t^{-0.58}$. Of course, if this corresponds to the asymptotic behavior of the diagonal and off-diagonal correlations, the former will decrease more rapidly than the latter, so that eventually $C(t) \approx C_{od}(t) \sim t^{-0.58}$. It is seen that the diagonal correlations are significant only for very short times in this case.

The early-time behavior is dominated by more complex relaxation processes, while the very-long-time behavior is related to a lower signal-to-noise ratio (and is therefore noisy) and the finite length of the chain. Thus, the downturn in all three curves for $\log_{10} t > 5.5$ corresponds to the exponential relaxation of the lowest frequency mode and the fluctuations result from the significant uncertainties associated with the data points. They can therefore be seen as artifacts coming from the finite size of the system and finite averaging time. Finally, Fig. 1(c) reveals that the current correlation ratio $R(t)$ rises rapidly to a value of about 3 (i.e., the off-diagonal term is three times more important than the diagonal one), then continues to rise, albeit at a slower rate. The variations at long times is again associated with noise.

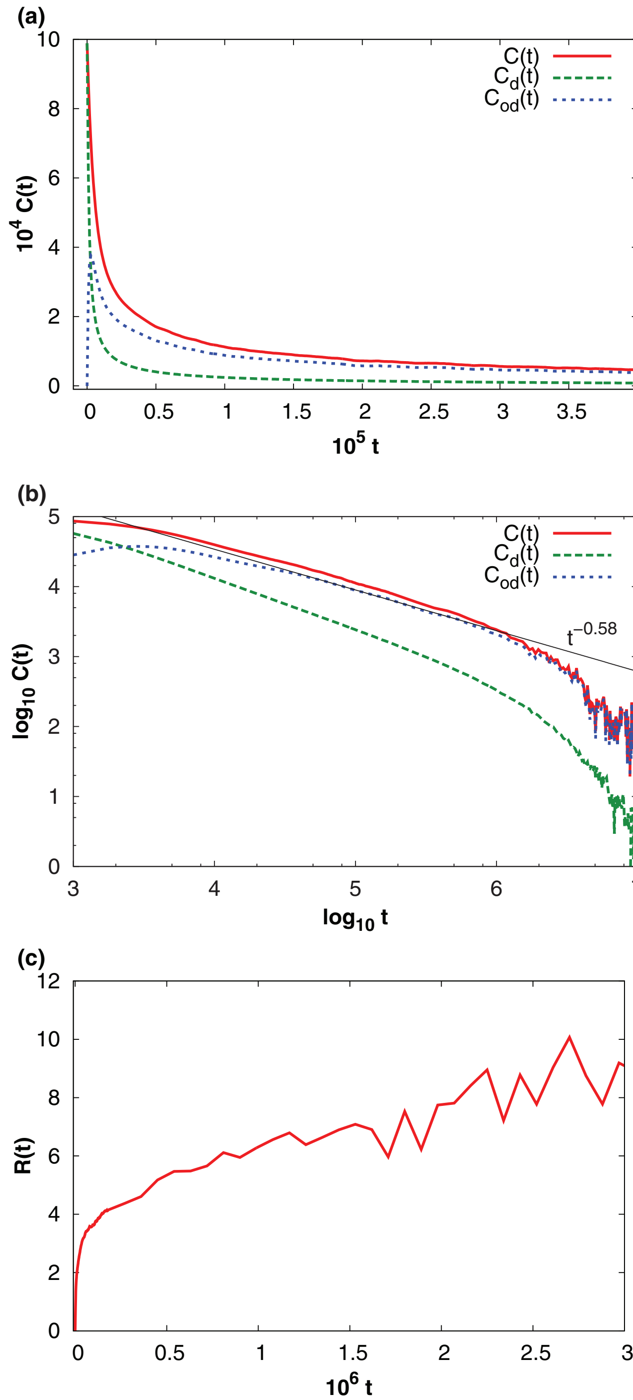

FIG. 1. (Color online) (a) Current-current correlation functions $C(t)$, $C_d(t)$, and $C_{od}(t)$ for the FPU-β model with $\beta = 0.01$. (b) Same data as (a) but in log-log form. (c) Correlation ratio $R(t)$ for the same system.

Turning now to a system with higher anharmonicity, $\beta = 0.1$, Fig. 2(a) is qualitatively similar to Fig. 1(a), though, of course, the correlations dissipate more rapidly (note the different time scale). Here, again, $C(t)$ and $C_d(t)$ reach their maximum for $t = 0$ and then decrease, while $C_{od}(t)$ starts from 0, quickly peaks, and then decreases. Examining Fig. 2(b), however, important differences are now visible. This time, only $C_d(t)$ seems to display a true power-law behavior, with

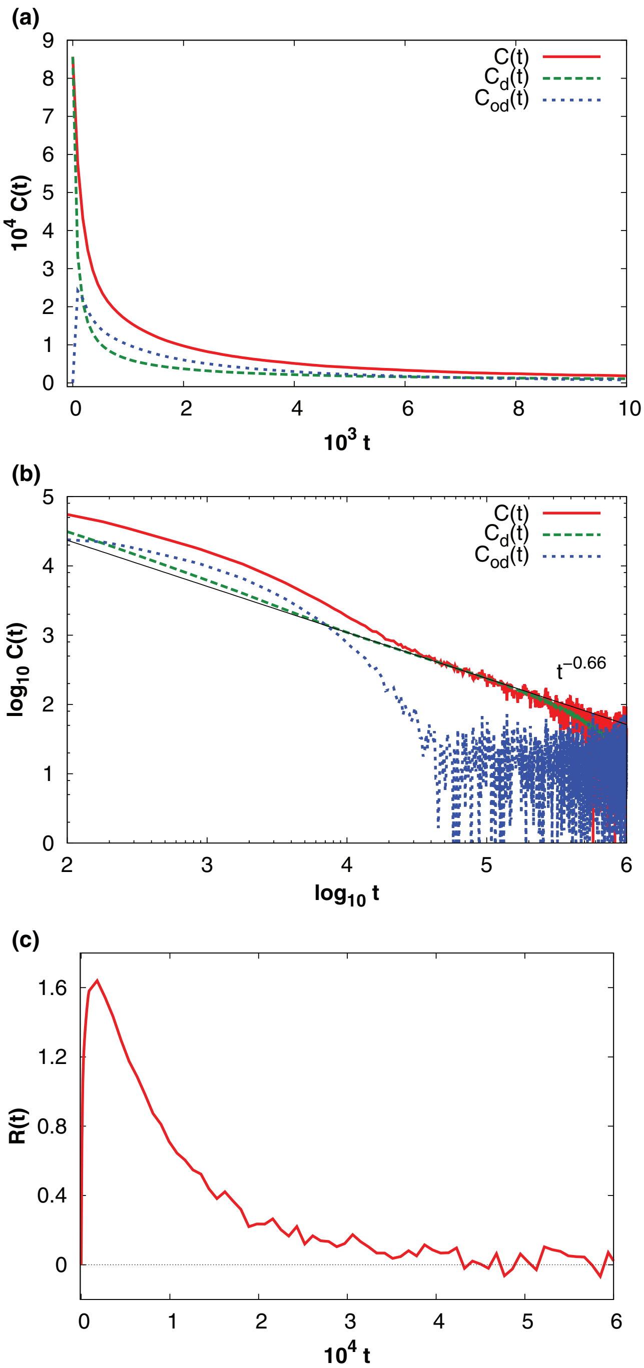

FIG. 2. (Color online) (a) Current-current correlation functions $C(t)$, $C_d(t)$, and $C_{od}(t)$ for the FPU-β model with $\beta = 0.1$. (b) Same data as (a) but in log-log form. (c) Correlation ratio $R(t)$ for the same system.

exponent -0.66. The off-diagonal term $C_{od}(t)$ is clearly decaying more rapidly than a power law but, nevertheless, more slowly than an exponential. The behavior for $\log_{10} t > 4.5$ is once more due to the low signal-to-noise ratio. Being the sum of the two previous terms, $C(t)$ goes from a behavior similar to that of $C_{od}(t)$ for short times to being roughly equal to $C_d(t)$ for long times. This can also be seen in Fig. 2(c), where it is clear that the off-diagonal terms are important only for short times and then return to 0.

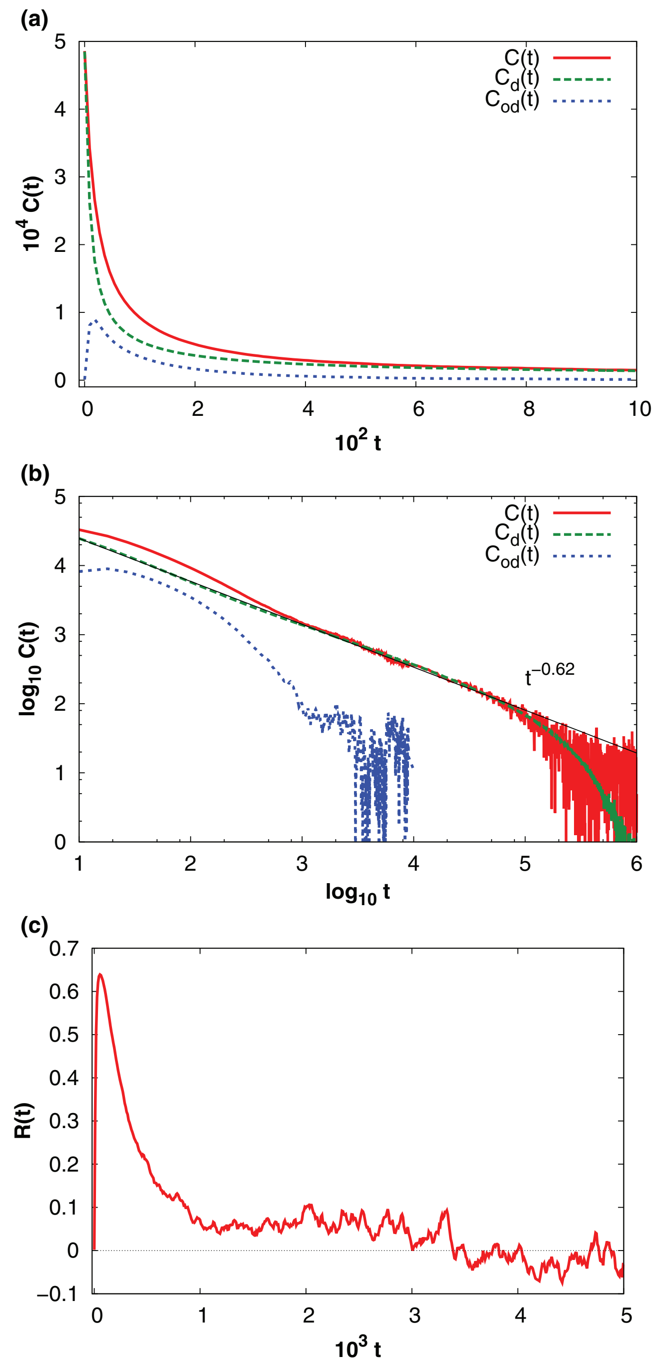

FIG. 3. (Color online) (a) Current-current correlation functions $C(t)$, $C_d(t)$, and $C_{od}(t)$ for the FPU-β model with $\beta = 1$. (b) Same data as (a) but in log-log form. $C_{od}(t)$ is shown up to $\log_{10} t = 4$ only (when noise starts to dominate), to avoid obscuring the other curves. (c) Correlation ratio $R(t)$ for the same system.

Finally, we examine a highly anharmonic system, $\beta = 1$. The results, shown in Figs. 3(a)–3(c), are similar to those for $\beta = 0.1$ except that the off-diagonal correlations are at all times less important than the diagonal correlations and decay very rapidly. For this case, the asymptotic form of the current-current correlation functions $C(t) \approx C_d(t) \propto t^{-0.62}$.

Therefore, the overall picture that emerges is this: for low anharmonicity, the relative magnitude of off-diagonal terms increases continuously with time, as illustrated in Fig. 1(c). The rate of this increase grows with anharmonicity from (exactly) 0 for the harmonic system to a maximum for $\beta \sim 0.01$, owing to the fact that more scattering between phonon modes leads to more correlations. (We have done calculations for many values of β.) In this regime, $C_{od}(t) > C_d(t)$, except for very short times, in clear violation of the *stosszahlansatz*. This is the case shown in Fig. 1. Increase in the anharmonicity level eventually leads to a point (the correlation threshold) where scattering starts to destroy cross-mode correlations more rapidly than they are created. In this regime $C_{od}(t)$ is significant only for a short period at early times and $C_d(t)$ rapidly becomes the dominant term, in agreement with the *stosszahlansatz*. Further increase in anharmonicity only brings about fewer cross-mode correlations and their more rapid decay. Whether there really is a transition as a function of anharmonicity is an important (and difficult) question, to which we return in Sec. IV D. For now, suffice to say that $\beta = 0.02$ is the lowest anharmoncity level at which we are able to observe $C_{od}(t)$ to decay more rapidly than $C_d(t)$. However, it cannot be excluded that $C_{od}(t)$ would decay on much longer time scales for lower β values.

As for the exponents associated with the decay of the correlation functions, they play an important role in the theory of anomalous heat conduction because $\kappa \propto N^{\alpha}$ implies, through Eq. (10), that the $t \to \infty$ limit in the total current correlation function is $\langle J(0)J(t)\rangle \sim t^{\alpha-1}$. In this regard, the results are peculiar, as they imply that, whether the system is in the regime where off-diagonal correlations are dominant or in the regime where they are negligible, the N dependence of the conductivity is similar. However, the exponents are notoriously hard to obtain precisely, and, the chains used in this study being only moderately long, it is possible that a different behavior could be seen in longer chains and/or on longer time scales. Therefore, it is impossible to confirm whether they are in fact identical or whether they coincidentally display very close values. In any case, this demonstrates that theories making use of the (incorrect) *stosszahlansatz* can yield fairly good results (but for the wrong reasons). Also, conductivity is often computed for a fairly high anharmonicity, as convergence is easier to obtain in this case.

B. Dependence on the number of oscillators

As indicated in Sec. III, the equilibrium method for computing the heat conductivity is used to characterize the system in the thermodynamic limit $N \to \infty$. However, simulations can only be undertaken with a finite number of particles and this is why periodic boundary conditions are employed, enabling current fluctuations in the system and making the method usable. On the other hand, in the thermodynamic limit, the allowed wave vectors are continuous while they are discrete (and equally spaced) in a periodic system, the obvious assumption being that if they become close enough, the dynamics should be similar. One can nonetheless wonder whether this has an impact when considering interactions between modes. Thus, it is instructive to examine the influence of the number of particles on the correlation functions.

The correlation functions for the FPU-β model with $\beta = 0.01$ and $N = 256$, 2048, and 16 384 are shown in Fig. 4(a). As implied by Eq. (10), current fluctuations are expected to grow linearly with N; thus, to ease comparison, the correlation functions were scaled by a factor $\frac{1}{N}$. The results reveal two seemingly opposite trends. First, the total correlation function $C(t)$ seems to scale with N almost exactly, which was expected. On the other hand, the diagonal term in the correlation $C_d(t)$, and consequently $C_{od}(t)$ as well, decays at a different rate. This implies that, although the off-diagonal correlations are much more significant in long chains, the diagonal correlations exactly counteract this, leading to zero net effect. However, for $\beta = 0.1$, Fig. 4(b) indicates that above the correlation threshold the three chains exhibit nearly the same behavior except for the fact that, as expected, finite-size effects are present at shorter time scales for the shortest chain. Our hypothesis, which is motivated below, is that correlations span a finite width in frequency space and that the sparser frequency spectrum associated with short chains leads to less off-diagonal correlations.

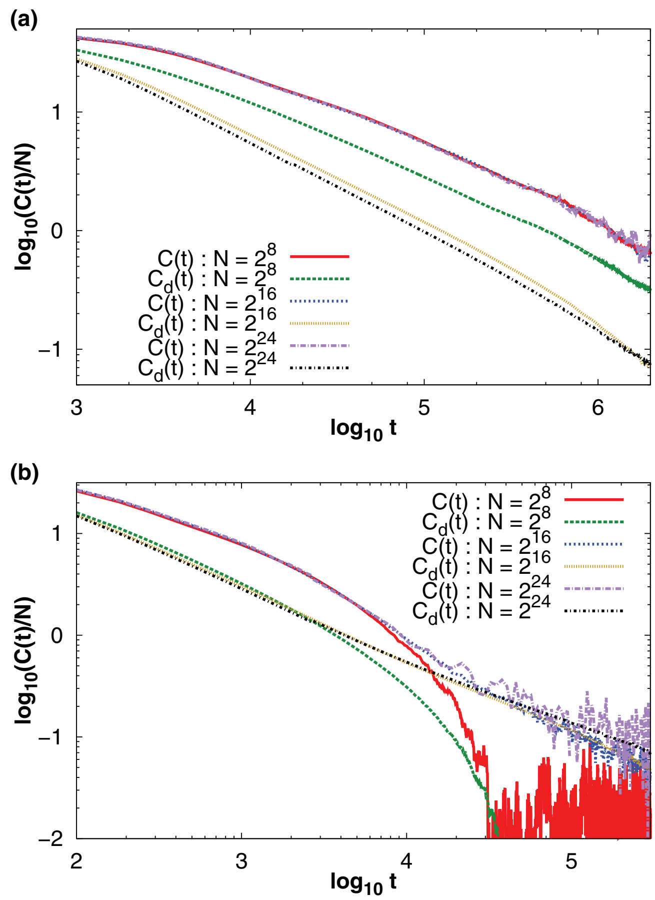

FIG. 4. (Color online) Current-current correlation functions $C(t)$ and $C_d(t)$ scaled by $1/N$ for the FPU-β model with $N = 256$, 2048, and 16 384 and (a) $\beta = 0.01$ and (b) $\beta = 0.1$.

C. Structure of the off-diagonal correlations

The hypothesis put forward in the previous section, *viz*., that correlations span mostly neighboring modes, can be tested by examining how any specific mode becomes correlated with others. To do this we again consider the FPU-β chains with $N = 256$ and $N = 2048$ oscillators. We introduce the

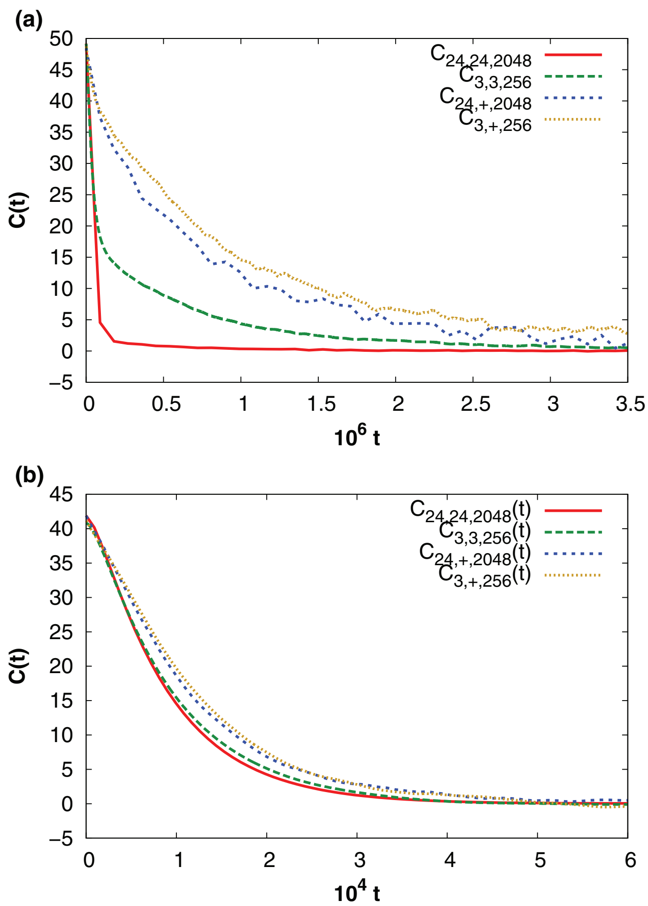

FIG. 5. (Color online) Current-current correlation functions $C_{24,24,2048}(t)$, $C_{3,3,256}(t)$, $C_{24,+,2048}(t)$, and $C_{3,+,256}(t)$ for the FPU-β model with (a) $\beta = 0.01$ and (b) $\beta = 0.1$. In the first case the $C_{k,k,N}(t)$ curves are found to be different, while the $C_{k,+,N}(t)$ curves are similar; in the second case all curves are relatively similar.

shorthand notation $J_+(t) \equiv \sum_{k>0} J_k(t)$ and

$$C_{k,p,N}(t) = \langle J_k(0) J_p(t) \rangle_N, \tag{19}$$

the subscript N indicating that the average is taken for a system of N oscillators. Figure 5 shows the correlation functions for modes of the same frequency in the two chains under ($\beta = 0.01$) [Fig. 5(a)] and over ($\beta = 0.1$) [Fig. 5(b)], the correlation threshold. In every case, the autocorrelation function $C_{k,k,N}(0)$ is equal to $C_{k,+,N}(0)$ [our definition for J_k implies $J_k = J_{-k}$, and thus taking $\langle J_k(0) J_+(t) \rangle$ instead of $\langle J_k(0) J(t) \rangle$ avoids double-counting], meaning that off-diagonal correlations are on average 0. Comparing the autocorrelations $C_{24,24,2048}(t)$ and $C_{3,3,256}(t)$ in Fig. 5(a), a much faster decay for the longest chain is found. On the other hand, the correlations with all the modes $C_{24,+,2048}(t)$ and $C_{3,+,256}(t)$ are much more similar. This observation supports the idea that the initial excitation of mode k quickly spreads to neighboring modes over a given energy range, then slowly decays. For the shortest chain, as mode frequencies are farther apart, a larger fraction of the excitation stays associated with mode k. Figure 5(b), for $\beta = 0.1$ (above the threshold), confirms that, in this case, off-diagonal correlations have much less impact. In both cases, the relaxation of the short chain is slightly slower.

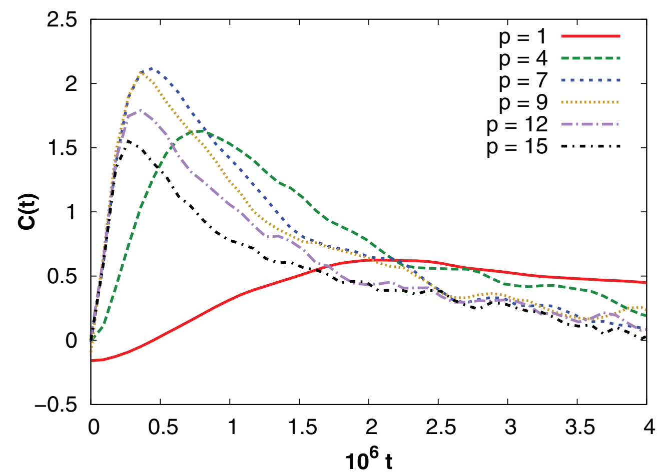

FIG. 6. (Color online) Current-current correlation functions $C_{8,p,2048}(t)$ for the FPU-β model with $\beta = 0.01$ and various values of p. Higher values of p lead to an earlier peak, and values closest to 8 lead to a higher maximum.

We can also look at exactly which modes get correlated with a specific mode. In Fig. 6, correlation functions of type $C_{k,p,2048}(t)$ are shown for $k = 8$, $\beta = 0.01$, and various values of p. Other choices of k produce similar results, although correlations decay more rapidly or more slowly when k is lower or higher, as expected. It can be seen that correlations are maximum for p values closest to k and that the peak correlations monotonously decrease when the difference between k and p increases. Nonetheless, the integrated correlations are often more important for lower p modes because, although the corresponding correlation functions tend to peak later and at lower values, they also decay slower.

The same kind of behavior is also exhibited in simulations in which energy relaxation from the first few modes (the original FPU problem) is studied (see, e.g., [31]). Indeed, energy quickly spreads to all modes up to a given frequency, which depends on the energy density but not on N, after which relaxation to higher frequency modes occurs, but on much longer time scales. Thus, for longer chains more individual modes are involved in this initial packet, just as our results indicate.

D. Relation to the FPU problem

We now put our findings in the broader perspective of the relaxation properties of the FPU model. Specifically, it is known that the FPU-β model displays a transition from weak to strong stochasticity (see, for example, Ref. [32]), although it is conjectured that equilibrium is always reached if the system is allowed to evolve for a sufficiently long time. It thus seems natural to link the crossover we observe in the level of off-diagonal correlations to the transition in the stochasticity level. In particular, one could imagine that these correlations inhibit the full expression of randomness in the evolution of the system and are associated with the weak-stochasticity regime.

To test this idea, we measure the stochasticity level by computing the largest Lyapunov exponent of the system, using standard procedures [33], as a function of β. The data are reported in Fig. 7 and generally agree with established results

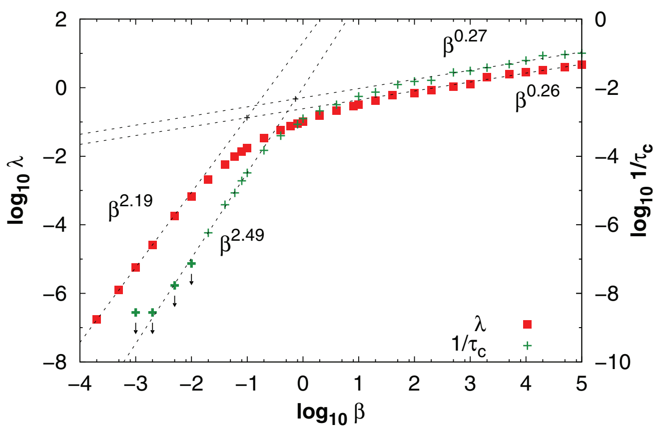

FIG. 7. (Color online) Lyapunov exponent λ and inverse correlation time $1/\tau_c$ as a function of β for the FPU-β model with $T = 1$ and $N = 2048$. Straight lines correspond to fits to the low- and high-anharmonicity data points. The transition is at $\beta = 0.099$ for λ and at $\beta = 0.73$ for $1/\tau_c$. Arrows indicate that the associated data are upper bounds on $1/\tau_c$, as transitions were not observed for these simulations.

[32]. Indeed, we observe two distinct power laws, with exponent 2.19 in the low-anharmonicity limit (the reported values are generally ∼2) and exponent 0.26 in the high-anharmonicity limit (1/4 was expected). The observed transition, defined as the point at which the two fits to the data cross, is located at $\beta = 0.099$. In the same figure, we also report the inverse of the correlation time, τ_c, defined as the time (after the initial transient) for which $C_{od}(t)$ has decreased to $0.9C(t)$. As explained previously, for $\beta < 0.02$ the transition times were never reached in our simulations, and therefore we can only plot upper bounds on $1/\tau_c$ (cf. arrows pointing downward). The two curves show strong similarities. First, the inverse correlation time also displays a transition between two power-law regimes which, in the high-anharmonicity limit, has nearly the same exponent as the largest Lyapunov exponent (0.27 vs 0.26). The low-anharmonicity regime has a slightly sharper slope (2.49 vs 2.19) and the transition is observed at a higher anharmonicity ($\beta = 0.73$ vs 0.099). The difference between the values at which the two regimes cross is real but much of the ranges over which the two transitions take place overlap. Also, a change in the somewhat arbitrary definition of τ_c could shift the transition in either direction. In any case, we observe that the existence of long-lasting off-diagonal correlations is associated with significant inhibition of the expression of chaos in the system. Although we cannot establish an obvious causal relationship between them, the similarities are nevertheless strong enough that, we conjecture, the two properties are related. In this case, the presence of localized modes is a likely explanation for both the correlations between modes and the reduced stochasticity level. And consequently, there exists a connection between the two most important aspects of the FPU problem: the existence of localized modes and the stochasticity threshold.

E. Cross-correlations in the FPU-$\alpha\beta$ and ϕ^4 models

We now examine two models in order to assess how some of the conclusions in the previous sections apply to

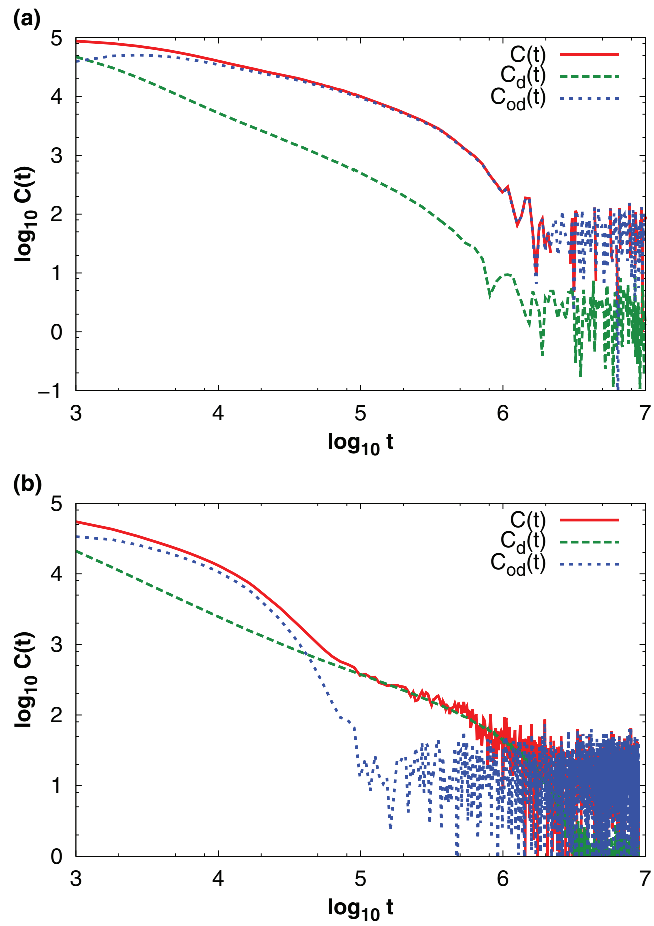

FIG. 8. (Color online) Current-current correlation functions $C(t)$, $C_d(t)$, and $C_{od}(t)$ for the FPU-$\alpha\beta$ model with (a) $\alpha = \beta = 0.01$ and (b) $\alpha = \beta = 0.1$.

one-dimensional systems in general. To this effect, we consider one anomalous asymmetric model and one regular symmetric model. For the former we chose the FPU-$\alpha\beta$ model with $\alpha = \beta$, obviously similar to the FPU-β model, although it is asymmetric. There have been recent claims that it is in fact a normal heat conductor [2,6] or, at least, that its asymptotic anomalous behavior can only be seen on much longer time scales [3–5]. Our results seem to agree with the latter view and might explain why it is so. Figure 8 shows, once more, the main correlation functions $C(t)$, $C_d(t)$, and $C_{od}(t)$ for $\alpha = \beta = 0.01$ and $\alpha = \beta = 0.1$. These are similar to what we observed for the FPU-β model in that, below a given anharmonicity threshold, the off-diagonal term $C_{od}(t)$ is the most important, while above the threshold, the off-diagonal terms decay more rapidly than the diagonal ones, and for this reason, the latter dominate at longer times. In fact, cross-mode correlations, which tend to contribute non-power-law terms to $C(t)$, seem to be significantly more important for this anharmonic model. Preliminary tests on the Lennard-Jones model either with first-neighbor interactions only or with first- and second-neighbor interactions (also asymmetric anomalous models) lead to even more cross-mode correlations. Regarding whether or not the FPU-$\alpha\beta$ is anomalous, our data confirm that $C(t)$ follows a power law for a sufficiently long time, indicating anomalous behavior. The fast relaxation of $C_{od}(t)$ in Fig. 8(b) could,

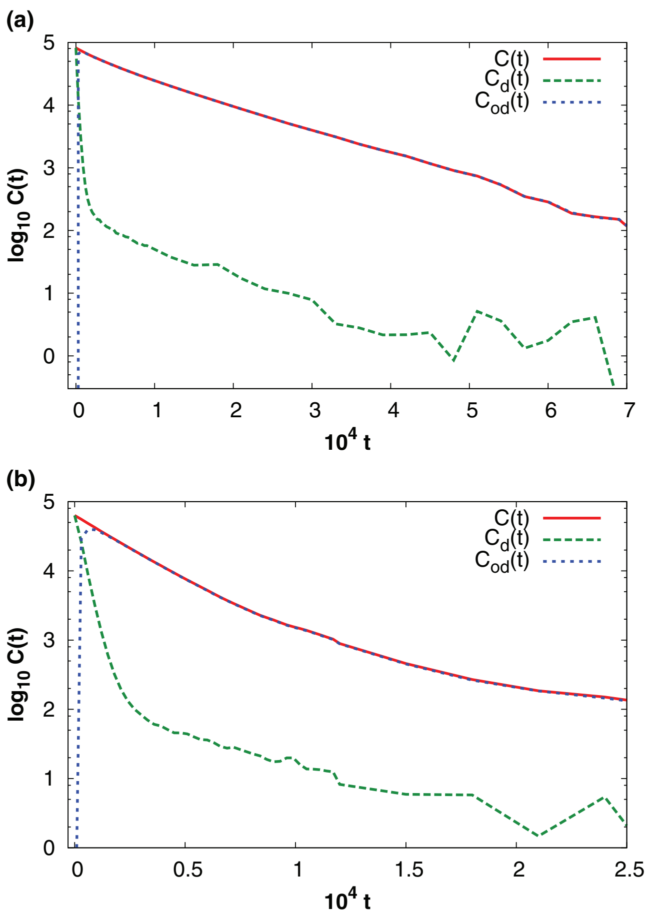

FIG. 9. (Color online) Current-current correlation functions $C(t)$, $C_d(t)$, and $C_{od}(t)$ [note that $C(t)$ and $C_{od}(t)$ are superimposed for almost all times] for the ϕ^4 model with (a) $\lambda = 0.01$ and (b) $\lambda = 0.1$. Note that only the y axis is in log form in both cases.

however, lead one to believe that, if one looks only at data for $\log_{10} t < 5$, the integral of $C(t)$ is finite. Thus, the plateau observed in plots of JN vs N (for example, see Fig. 2 in Ref. [2]) or the changes of slope in plots of κ vs N in Ref. [14] could in fact be a signature of significant off-diagonal correlations.

This naturally leads to questions relating these correlations to anomalous conductivity. To address this point, we computed cross-mode correlations in a second model, the ϕ^4 model, which is symmetric and widely known to be regular. The correlation functions for $\lambda = 0.01$ and $\lambda = 0.1$ are shown in Fig. 9; as relaxation is now expected to be exponential, the data are presented in a log-linear scale. Indeed, $C(t)$ is fairly linear for both values of the anharmonicity parameter. In both cases, $C_d(t)$ decreases to negligible values in a very short time and quite generally $C(t) \approx C_{od}(t)$. We were not able to observe a threshold at which the relative contribution of $C_{od}(t)$ drops for this model, likely because, in contrast to the anomalous models, all correlations are now short-term (exponential decay) so that the diagonal correlations cannot "outlive" the off-diagonal terms. In any case, it is clear that the existence of cross-mode correlations is not a condition for it to display anomalous conductivity.

Another interesting point here concerns the nature of the relaxation of $C_d(t)$, which happens in an initial very fast step, followed by a slower one, with a relaxation time similar to that of $C(t)$ or $C_{od}(t)$. Again, this might be explained by the fact that current fluctuations in a given mode quickly spread out to modes of similar frequencies, after which this "semilocal" excitation is attenuated.

V. CONCLUSION

By computing the correlations between different phonon modes in anharmonic oscillator chains, we have shown that the *stosszahlansatz*—the hypothesis according to which the phonon modes are independent—is incorrect in the low-anharmonicity regime and in general for short-term correlations. Considering principally the FPU-β model, but also the FPU-$\alpha\beta$ model, we found that, below a certain anharmonicity threshold, phonon modes can become correlated and remain so for long times. While modes are, on average, independent, ($\langle J_k(0) J_{p\neq k}(0)\rangle = 0$), a fluctuation of the current associated with a given mode quickly spreads to the neighboring ones, and the mode packet thus created then decays in a manner similar to that of the original individual phonon mode. This explains the success, at least on a qualitative level, of theoretical explanations of the phenomenon based on the relaxation-time approximation, even though it is fundamentally incorrect. Although the diagonal and off-diagonal contributions of the total current-current correlation function generally follow different trends, their sum typically corresponds to a power law ($C(t) \sim t^{-\gamma}$) for long times but the computed exponents, $\gamma \sim 0.58$–0.66, cannot lend unambiguous support to either the $\kappa \propto N^{2/5}$ or the $\kappa \propto N^{1/3}$ hypothesis.

We have also shown that the inverse of the off-diagonal correlation time $1/\tau_c$ behaves like the largest Lyapunov exponent of the system, as both quantities exhibit two scaling regimes as a function of β, with similar exponents and transition points within an order of magnitude of each other.

However, it must be noted that the phenomenon described here does *not* explain or cause anomalous conductivity. Indeed, we found that the spreading of the current across neighboring modes also happens in the ϕ^4 chain, a model known to display regular heat conductivity. From this point of view, our results are in contradiction with the conclusions of Henry and Chen [27], although their model is in some ways fundamentally different from the Hamiltonians we studied: even though correlations between modes are significant in oscillator chains, we have found no evidence that they are either sufficient or necessary for a system to exhibit anomalous heat conduction. This does not mean that they are meaningless, however, as the study of asymmetric models illustrates how they can help shape the relationship between the length and the conductance of a device. A better understanding of these correlations could thus help tailor the heat transport properties of nanostructures.

A technical sidenote is in order here: in all cases the fluctuations associated with $C_d(t)$, the diagonal correlation function, are much less than that associated with $C(t)$, the total current-current correlation function. Much focus has been put on the asymptotic form of $C(t)$, which, at least in the high-anharmonicity regime, seems to equal that of $C_d(t)$. Even though substantial computing costs are associated with the calculation of the individual $J_k(t)$ for every mode, it might still be possible, using appropriate sampling, to probe long time scales more efficiently by studying $C_d(t)$ rather than $C(t)$.

Finally, some questions remain unanswered. First, it would be interesting to examine how our findings apply to nonequilibrium conditions. The equilibrium method for computing thermal conductivity is associated with linear response theory and the existence of a local thermodynamic equilibrium. Knowing that the models studied can display nonperturbative localized solutions, its applicability is still not fully established. Nonetheless, the aforementioned comparisons of our results for the FPU-$\alpha\beta$ model with those from nonequilibrium simulations, as well as similar comparisons in previous studies, are encouraging. It would, however, be interesting to examine nonequilibrium systems to try to find similar effects. As they stand, the existence of these cross-correlations and their form might imply that they are associated with localized disturbances. It is therefore possible that these off-diagonal correlations are a signature of soliton waves or breathers, an hypothesis that seems to be supported by a recent study [34] indicating that the lifetime of solitons in FPU-β lattices is maximum for a moderate anharmonicity level. These points will be assessed in future work.

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