Electron-phonon interactions and recurrence phenomena in one-dimensional systems

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(Received 18 October 1993)

We study the time dependence of a combined system of an electron described by a tight-binding model interacting with vibrational degrees of freedom in a one-dimensional lattice. The time evolution of our coupled model is described by a nonlinear system of differential-difference equations. Apart from localized solutions (polarons) and extended (Bloch-like) solutions, we found that for a small parameter range, there exists a periodic exchange of energy between the electronic and vibrational degrees of freedom: a small part of the electronic energy is transferred to the lattice and then completely back to the electron with an impressive regularity.

The interactions between electrons and lattice vibrations are of fundamental importance in understanding the behavior of solids. Basic properties, such as electrical resistivity, depend crucially on this interaction. More complicated processes of diametrically opposite phenomenology, such as superconductivity and small polaron formation,[1] are also due to electron-phonon interaction. Traditionally, this difficult problem is treated within the framework of many-body theory and the focus is on the properties of the ground state and the low-lying excited states. Besides the inevitable approximations, the traditional approach is not easily adapted to time evolution problems, such as the behavior of a highly excited single electron in an otherwise empty band. The usual approach to this problem is based upon the assumption that the electron gradually transfers its energy to the lattice (within a few times the lattice vibration typical period), reaches the bottom of the band, and then the problem is reduced to a study of the ground state. The many-body theory has difficulties also in treating the simultaneous presence of electron-phonon interaction and disorder, especially near the phase transition point of propagating to localized eigenstates.[2] In recent years a complementary approach to the many-body theory is emerging under the generic name of nonlinear physics.[3–5] It is based on rigorous analytical treatment and numerical simulations of simple models such as the nonlinear Schrödinger (NLS) equation, nonharmonic lattice vibrations, and nonlinear magnetic Hamiltonians. This relatively new approach is perfectly suited for studying time evolution problems and for incorporating disorder. One of the models treated through this technique is the interaction of an intramolecular excitation (exciton) with the lattice vibrations[5,6] as well as the coupling of the latter to the off-diagonal electronic matrix element.[7] More recently, an extensive numerical simulation of an electron propagating in a quasiperiodic one-dimensional (1D) model and interacting with lattice vibrations was performed,[8,9] where the previously unexplored, highly excited states of the system were studied for the first time. Among other surprising findings, strong deviations from the expected thermodynamic behavior were observed.

In the present paper we report an even more striking behavior of this coupled electron-lattice vibration model. Indeed, for a small part of the parameter space, we found that the electron initially placed in a highly excited eigenstate, after transferring a small fraction of its energy to the lattice, reabsorbs the transferred energy and the entire coupled system returns (within numerical uncertainties) to its initial state. This behavior repeats itself in an almost perfect periodic way for hundreds of periods. This impressive recurrence is reminiscent of the Fermi-Pasta-Ulam[10] results and suggests that our complicated electron-lattice vibration Hamiltonian may belong to a torus involving a yet unknown integrable coupled model. The Hamiltonian describing our model is $H = H_e + H_l + H_{e\text{-}l}$, where

$$H_e = \sum_n \epsilon_n |n\rangle\langle n| - J \sum_n |n\rangle(\langle n+1| + \langle n-1|), \quad (1)$$

$$H_\ell = \frac{1}{2M}\sum_n p_n^2 + \frac{K}{2}\sum_n (u_{n+1} - u_n)^2, \quad (2)$$

$$H_{e\text{-}\ell} = \chi \sum_n |n\rangle\langle n|(u_{n+1} - u_{n-1}). \quad (3)$$

The local orbitals $|n\rangle$ centered around site n ($n = 1,...,N$) form a 1D lattice. In the present work we restrict ourselves to the periodic case; i.e., the diagonal matrix elements ϵ_n have the same value (taken to be zero). The lattice part H_ℓ describes a system of N coupled classical harmonic oscillators with mass M, displacement u_n, momentum $p_n = M\dot{u}_n$, maximum eigenfrequency $2\omega_0$, and sound velocity $c=\omega_0 a$, where $\omega_0 = \sqrt{\frac{K}{M}} \equiv t_\ell^{-1}$, a is the lattice constant, and t_ℓ is the characteristic lattice time. The electron-lattice interaction $H_{e\text{-}\ell}$ is a symmetrized deformation potential with χ being the strength of the coupling which can be characterized by the dimensionless quantity $\lambda = \frac{\chi^2}{KJ}$ (similar to the one appearing

in superconductivity and with typical values that range from 0.1 to 1.5). If the electronic wave function is written as $\Psi_e = \sum_n c_n(t)|n\rangle$, the equations of motion for the coupled electronic-lattice system become

$$i\hbar\frac{dc_n}{dt} = \chi(u_{n+1} - u_{n-1})c_n - J(c_{n+1} + c_{n-1}), \quad (4)$$

$$M\frac{d^2u_n}{dt^2} = K(u_{n+1} + u_{n-1} - 2u_n) + \chi(|c_{n+1}|^2 - |c_{n-1}|^2). \quad (5)$$

Within the adiabatic approximation (i.e., for $Md^2u_n/dt^2 \approx 0$) this system of equations becomes a discrete version of the nonlinear Schrödinger equation. In the continuum limit it reduces to the integrable nonlinear Schrödinger equation which admits three branches of solutions: Bloch-like periodic solutions, soliton solutions, and the so-called "cnoidal wave" periodic solutions.[5,6] The adiabatic approximation is not adequate, in general, for arbitrary highly excited states. Thus, in view of the complex behavior of the discrete nonlinear Schrödinger equation and the essential complications due to the introduction of new (lattice) degrees of freedom and an associate new time scale characterizing the ionic vibrations, our model is expected to exhibit a very rich behavior and the use of numerical simulations becomes necessary. In our numerical studies, the lattice is initially at rest, undeformed, and the electron is very close to an eigenstate of H_e with energy $E_e(0) = -2$, -1, 0, or localized in a few sites around the middle with approximately the same energies as the eigenstates above. The unit of energy is J and $t_e = \hbar/J$ is the characteristic electronic time. With the choice of M and $\sqrt{\frac{M}{K}}$ as the natural units of mass and time, respectively, we are left with two parameters: (a) $\hbar$ (measured in units $J\sqrt{\frac{M}{K}}$ or equivalently the dimensionless quantity $\frac{\hbar/J}{\sqrt{M/K}} = t_e/t_\ell$). (b) χ (measured in units of $\sqrt{JK}$ or equivalently the dimensionless quantity $\frac{\chi}{\sqrt{JK}}$). Typical values for our natural units are J=1 eV, $M=Am_u$ where A is the mass number, m_u the atomic mass unit, and K=50 N/m. Periodic boundary conditions are used and the time integration is performed with the fourth-order Runge-Kutta method with a step equal to $10^{-4}t_l$. Throughout our simulations, energy is conserved to a relative accuracy of at least 10^{-5}. We examine the time development of (a) the electronic wave function, (b) the displacements and velocities of the lattice atoms, (c) the participation number $P(t) \equiv [\sum_n |c_n(t)|^4]^{-1}$, (d) the electronic energy $E_e = \langle\Psi_e|H_e|\Psi_e\rangle$, (e) the lattice energy $E_l = \frac{1}{2M}\sum_n p_n^2 + \frac{K}{2}\sum_n(u_{n+1} - u_n)^2$, and (f) the interaction energy $E_{e\text{-}l} = \chi\sum_n c_n^\dagger c_n(u_{n+1} - u_{n-1})$. In this paper, we will concentrate on the case where the electron has a small effective mass comparing to the lattice mass, i.e., $\hbar = t_e/t_\ell = 0.012\,24 \ll 1$, which is typical for most metals. This is the usual adiabatic limit. However, in very narrow-band materials the ratio t_e/t_ℓ may approach (or even exceed) unity. An extensive presentation of our results relevant to the latter case will be given elsewhere. It should be emphasized that we restrict ourselves to the low-temperature case.

For low initial energy, e.g., $E_e(0) = -2$ (which implies that the electron starts at the ground state of H_e) we recover the expected many-body behavior for a 1D system: A polaron is formed, the spatial extent of which is proportional to χ^{-2} (for weak coupling, $\chi \leq 0.5$). The shape of the localized electronic wave function is approximately the soliton-type obtained analytically within the adiabatic and the continuum approximation

$$c_n = \left(\frac{\chi^2}{2KJ}\right)^{1/2} \operatorname{sech}\left[\frac{\chi^2}{KJ}(n - n_0)\right]. \quad (6)$$

As the initial energy of the electron gets considerably higher, we do not obtain localized polarons, unless we reach extremely high and unrealistic values of the coupling constant. In fact, in all cases of small mass and highly excited electrons we found, in agreement with Refs. 8, 9, that the transfer of energy from the electron to the lattice degrees of freedom slows down and eventually it seems to stop (on the average) without the electron being thermally equalized with the lattice. This seemingly antithermodynamic behavior is associated in most cases with a very complicated time evolution involving strong short term fluctuations and long term variations. Even for relatively long times ($\sim 10^4 t_\ell$) this almost chaotic behavior continues. However, for a small part of the parameter space, we found a striking behavior which is not only antithermodynamic but very regular as well.

In Fig. 1, we show this remarkable behavior. The data were obtained for the case where the electron is placed initially very close to an eigenstate with E_e=-1, the coupling constant is χ =1.5, and N=300. (Similar results were obtained for N in the vicinity of 300.) An impressive recurrence phenomenon is exhibited. The electron transfers a small fraction of its energy to the lattice; this transferred energy spreads over the vibrational degrees of freedom and then regroups and returns to the electron for each recurrence period, thus driving the system repeatedly back to its initial electronic state with the lattice undistorted and at rest. In Fig. 1(a) we show this time variation of the total lattice energy; it exhibits periodically placed spikes and between the spikes the lattice energy becomes zero. We have followed this time evolution for more than 100 periods without any visible deterioration of this regular pattern. The interaction energy follows an almost identical pattern with the maximum value being 0.0026 (instead of 0.0041 for the lattice energy). In Fig. 1(b), the time evolution of the electron energy is shown; at the position of each spike the electron transfers 0.0067 units of energy to the lattice and to the interaction energy but it recovers all this energy between spikes. Even the participation number which is a very sensitive function of any fluctuation in the wave function returns to its initial value [$P(0)$=300] with impressive regularity and accuracy [Fig. 1(c)]. The numerical solution clearly indicates that this periodic transfer of energy back and forth from the electronic modes to the lattice modes corresponds to periodic modulation and demodulation of the wave function. In other words,

the electronic wave function is not a simple Bloch state anymore, but in addition to the "carrier" wave number corresponding to this eigenstate, a wave envelope develops. When it reaches maximum modulation $|E_e - E_e(0)|$, $|E_{e\text{-}\ell}|$ and $|E_\ell|$ reach their maximum values and when it demodulates, the energies return to their initial values. The recurrence time (period) T_r for this specific example shown in Fig. 1 is $T_r = 60t_\ell$. This remarkable behavior is robust for small changes in the parameters and in the initial conditions. It survives in the region $1.35 < \chi < 1.58$, although the period depends strongly on χ ranging from $\sim 350t_\ell$ to $45t_\ell$ as χ increases within the above range. Beyond the upper limit ($\chi \simeq 2$) an intermediate type[8,9] of behavior is approached with an apparent chaotic time evolution. This recurrence has been observed for some other values of N and $\hbar$ (even though not as persistent as for $N \simeq 300$ and rarely for $\hbar$ outside the range of $0.007 < \hbar < 0.019$) but only for initial states close to the electronic eigenstate with $E_e \simeq -1$. It also seems to be persistent and stable (we run simulations for $\sim 10^4 t_\ell$ and small random perturbations did not destroy it). In Figs. 2 and 3, we show how the recurrence period (T_r) and the maximum lattice energy ($E_{\ell,\max}$) depend on the coupling (χ) and the electron mass ($\hbar$), respectively, for the region of the parameter space in which recurrence appeared. The strong dependence of T_r on the coupling constant shows that the phenomenon is due to the nonlinearity of our model and excludes linear-resonance type of explanations. As we approach the lower quasicritical value of χ ($\chi \simeq 1.35$), the recurrence period becomes extremely long and the maximum value of E_ℓ and $E_{e\text{-}\ell}$ extremely low; thus, the recurrence tends to disappear and the system seems to remain in its initial state. For values of χ close to the upper quasicritical value ($\chi \approx 1.57$) the pattern after a few regular periods shows a variation in the value of the period followed by fluctuations in the maximum values (of E_ℓ and $E_{e\text{-}\ell}$) and eventually an apparently chaotic behavior is reached with considerably larger values of E_ℓ and $E_{e\text{-}\ell}$. The dependence of the phenomenon on $\hbar$ (i.e., on the electron mass) shows two distinct regimes (Fig. 3): For $\hbar \geq 0.0115$ the behaviors of T_r and $E_{\ell,\max}$ are as in Fig. 2; i.e., T_r decreases as $\hbar$ increases, while $E_{\ell,\max}$ increases with $\hbar$. Also, the recurrence disappears for $\hbar \approx 0.019$ in a very similar way as for $\chi \approx 1.57$ described previously. However, for $\hbar \leq 0.0115$ the dependences of T_r and $E_{\ell,\max}$ are diametrically opposite: T_r increases and $E_{\ell,\max}$ decreases with increasing $\hbar$. Moreover, in this region, as opposed to the region $\hbar \geq 0.0115$, the interaction energy $E_{e\text{-}\ell}$ is negative and about equal in size to the lattice energy, so the electron energy changes very little even at the spikes. We have no explanation for this peculiar behavior. These results are remarkable not only because thermalization does not occur in the sense of eventual irreversible equipartition of energy among all different degrees of freedom, but because of the striking periodic regularity of returning to the initial state in spite of the relative complexity of our Hamiltonian. This behavior strongly suggests that our model is close to (i.e., belongs to the same torus as) an integrable system. Further support of this idea is provided by the fact that we were able to approximate our numerical results with solutions that consist of products of elliptic and trigonometric functions. The elliptic func-

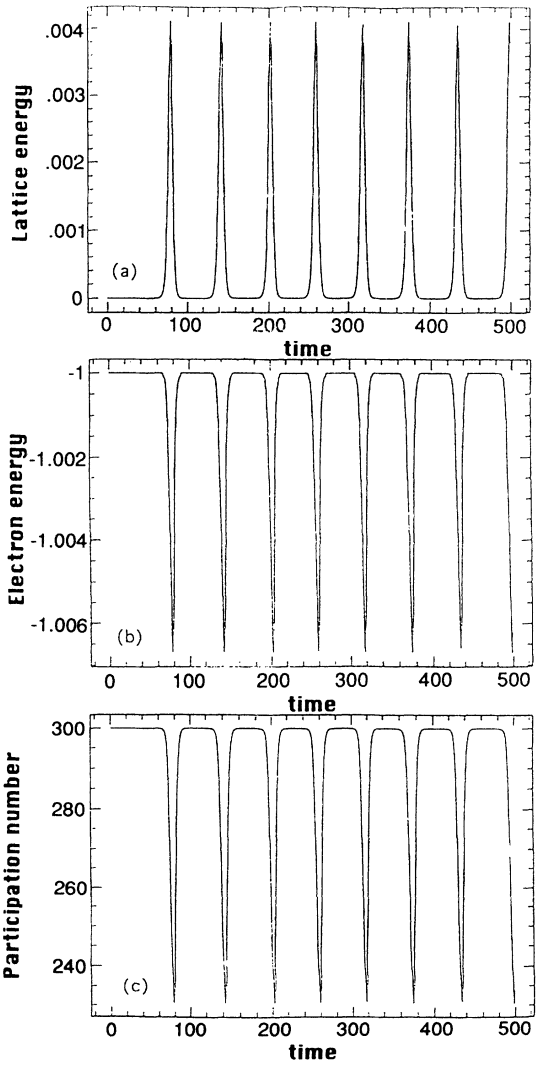

FIG. 1. The lattice energy E_ℓ (a), the electron energy E_e (b), and the participation number (c) versus time for a system with initial energy $E_e(0) = -1$, the electron-phonon coupling $\chi = 1.5$, $N = 300$, and $\hbar = 0.012\,24$. The unit of time is t_ℓ, length is the lattice spacing, and energy is J.

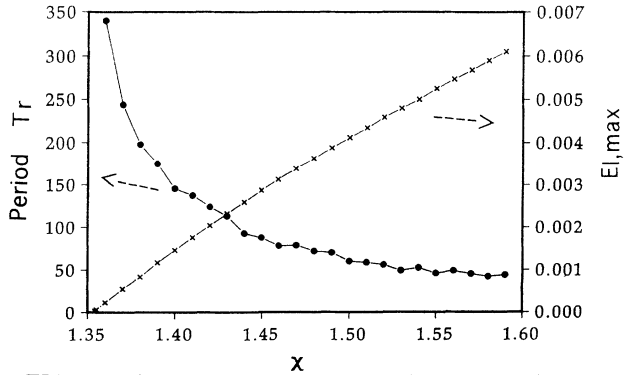

FIG. 2. The recurrence period T_r (solid circles) and the maximum lattice energy amplitude $E_{\ell,\max}$ (×'s) as a function of the electron-phonon coupling χ for a system with $N = 300$ and $\hbar = 0.01224$.

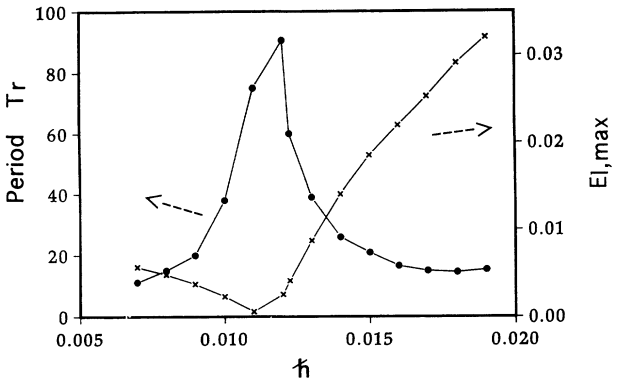

FIG. 3. The recurrence period T_r (solid circles) and the maximum lattice energy amplitude $E_{\ell,\max}$ ($\times$'s) as a function of the electron mass ($\hbar$) for a system with N=300 and χ=1.5.

tions (with time-dependent arguments only) are the envelope functions. The form of approximate solutions for the electronic wave function and the lattice displacements reproduces the various energies and participation number profiles. More specifically we have

$$\text{Re } c_n = \frac{1}{\sqrt{N}} \cos\left(\frac{\pi}{3}n - \omega t\right) + C_1 cn(\lambda t|k) \cos\left(\frac{2\pi}{N}n + \frac{\pi}{3}n + \omega' t + \omega t\right), \quad (7a)$$

$$u_n = C_2 cn(\lambda t|k) \cos\left(\frac{2\pi}{N}n + \frac{2\pi}{3}n + \omega' t + \varphi\right), \quad (7b)$$

where $\hbar\omega = E_e(0) = -1, \pi/3$ is the initial Bloch wave vector [corresponding to $E_e(0) = -1$], C_1 and C_2, the modulation amplitudes, are related to $E_{\ell,\max}$, λ is related to the recurrence period, T_r, k (the modulus) is almost unity, $\omega' = 2\pi/T'$, $T' \approx 2.64 t_\ell$, and φ is a phase, which depends on the parameters. The intermediate frequency ω' seems to be independent of the parameters χ and $\hbar$. The above expressions, although they fit rather well the numerical data, are not exact solutions of the equations of motion (4) and (5) for any value of the parameters and the initial conditions. However, these expressions have a similar form (except that the argument of the elliptic function depends on time only and not on $\xi = ut - x$ as in the integrable case) as a class of exact solutions of the integrable model of the continuous NLS equation[5] and some integrable version of the discrete NLS equation.[11] The appearance of these solutions is related to the instability of the initial Bloch eigenstate. A similar recurrence resulting from a Benjamin-Feir instability has been observed both numerically and experimentally for the NLS equation.[12–14] However, the fact that recurrence emerges in our model, which in addition to the electronic degree of freedom fully includes the vibrational degrees of freedom, indicates that this phenomenon might have a more general physical significance. It also appears that the initial state giving rise to the recurrence of our model does not necessarily satisfy the conditions obtained from linear and nonlinear stability analysis for the NLS.[4,15] It is possible that for this small region (or perhaps for other small regions as well) of the parameter space our system exhibits local integrability or it is very close to being integrable.

In conclusion, we found that a highly excited electron exhibits a very remarkable time evolution for a region of the parameter space of our relatively complex model. Not only equipartition of energy is not reached, but in this region of parameter and initial condition space a striking periodic recurrence behavior is exhibited, driven by the instability of the initial Bloch state but returning to it with a remarkable regularity. This recurrence can be described approximately by "cnoidal" wave solutions, suggesting a torus of almost integrability interpretation of our results.

The authors would like to thank Dr. D. Turner for his help with the parallelization of the computer programs. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-ENG-82. This work was supported by the Director of Energy Research, Office of Basic Energy Sciences, NATO Grant No. RG769/87 and NSF Grant No. INT-9117356, and CEC Grant No. SCC*.CT90- 0020.

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