Unusual Two-stage Dynamics of the Spin-Lattice Polaron Formation

Jan Kogoj,1 Zala Lenarčič,1 Denis Golež,1 Marcin Mierzejewski,2 Peter Prelovšek,1,3 and Janez Bonča1,3

1J. Stefan Institute, 1000 Ljubljana, Slovenia
2Institute of Physics, University of Silesia, 40-007 Katowice, Poland
3Faculty of Mathematics and Physics, University of Ljubljana, 1000 Ljubljana, Slovenia

We follow the formation of a spin-lattice polaron after a quantum quench that simulates absorption of the pump–pulse in the time–resolved experiments. We discover a two-stage relaxation where spin and lattice degrees of freedom represent an integral part of the relaxation mechanism. In the first stage the kinetic energy of the spin-lattice polaron relaxes towards its ground state value while relaxation processes via spin and phonon degrees of freedom remain roughly independent. In the second, typically much longer stage, a subsequent energy transfer between lattice and spin degrees of freedom via the charge carrier emerges. The excess local spin energy radiates away via magnon excitations.

Recent developments in the experimental techniques open unprecedented possibilities of studying the dynamics of interacting quantum systems. The time–resolved spectroscopy of solids is one of the most spectacular examples of this progress [1–7]. Solids are complex objects consisting of various subsystems with different excitations, like phonon–, spin– and charge–excitations. The real–time measurements of the relaxation processes give important information about interactions between these subsystems at various time/energy scales [3, 8–10]. However, the relaxation of various subsystems occurs at exceedingly different time–scales [11–15]. In particular, the time–resolved spectroscopy revealed a complex hierarchy of the relaxation times [4] ranging from tens of fs (e.g., for the coupling between charges and spin excitations) up to several ps (e.g., for the coupling between charges and some phonon branches). Such a broad range of relaxation times poses serious challenge for the theoretical analysis: (i) due to a large number of relevant degrees of freedom studying several subsystems is a complicated task itself (ii) some theoretical approaches (e.g. t–DMRG) are useful/applicable only in a certain time–window. Hence, various stages of the relaxations have been investigated within very different (and mostly non–overlapping) approaches [19–25]. The initial ultrafast stage has been studied within fully quantum and fully nonequilibrium approaches [17, 26, 27]. Most of the up–to-date approaches take into account charge carriers that couple to only one type of chargeless excitations. The subsequent slower stage consist in the energy flow between various chargeless subsystems and can be studied within quasiequilibrium approaches which rely on the notion of well defined temperatures [3].

In this Letter we apply a single fully nonequilibrium approach to show how the multi–stage relaxation emerges in systems of a charge carrier coupled to magnons and phonons. The initial ultrafast cooling of highly–excited charge is followed by much slower exchange of energies between the magnon and the phonon subsystem. Despite the absence of any direct coupling between magnons and phonons the latter stage of relaxation can be effectively mediated even by very dilute charge carriers.

We consider a single hole within the t–J Holstein model in one spatial dimension under the influence of a staggered field:

\[ H = H_{\text{kin}} + H_J + H_h + H_{\text{G,F}} + H_{\text{ph}}, \]

\[ H = -t_0 \sum_{i,\sigma} [\tilde{c}_{i,\sigma}^\dagger \tilde{c}_{i+1,\sigma} + \text{H.c.}] + J \sum_i S_i \cdot S_{i+1}, \quad (1) \]

\[ + \hbar \sum_i (-1)^i S_i^z + g \sum_i n_i^h (a_i^\dagger + a_i) + \omega_0 \sum_i a_i^\dagger a_i, \]

where \( t_0 \) is the nearest neighbor hopping amplitude, \( \tilde{c}_{i,\sigma} = c_{i,\sigma} (1 - c_{i-1,\sigma}^\dagger c_{i-1,\sigma}) \) is a projected fermion operator, \( J \) represents the Heisenberg exchange interaction, \( S_i \) is the spin operator and \( h \) represents the staggered magnetic field. Electron phonon coupling strength is given by \( g, a_i^\dagger (a_i) \) are phonon creation (destruction) operators at sites \( i \), and \( n_i^h = 1 - \sum_{\sigma} c_{i,\sigma}^\dagger c_{i,\sigma} \) is the hole density. \( \omega_0 \) denotes the dispersionless phonon frequency. We measure all quantities in units of \( t_0 \) and finally set \( t_0 = 1 \). The main reason for including the staggered field is to remove the spin-charge separation thus introducing the notion of the string picture, characteristic of the two-dimensional system.

We employ the exact diagonalization method (ED) defined over a limited functional space (EDLFS), which was successfully used to describe properties of a carrier doped into a planar ordered AFM described by the t–J model [28] and in the presence of lattice degrees of freedom, [29, 30]. The advantage of EDLFS over the standard ED follows from systematic generation of states which contain spin and phonon excitations in the vicinity of the carrier. We compute the initial state \( |\psi(t = 0)\rangle \) using the Lanczos technique and setting the initial value of the overlap integral to \( t_0 = 0 \). We then make a sudden quench by switching \( t_0 \) from 0 to 1 and time evolve the initial \( |\psi(0)\rangle \) using the time propagator with the quenched Hamiltonian. At each small time step \( \delta t \ll 1 \) we use Lanczos basis for generating the evolution \( |\psi(t)\rangle \rightarrow |\psi(t+\delta t)\rangle \) [31, 32].

In Fig. 1 we display different energies representing ex-
FIG. 1. The evolution after the \(t_0\) quench: time-dependence of the change of the kinetic energy of the hole (circles) \(\Delta E_{\text{kin}} = \langle \psi(t)|H_{\text{kin}}|\psi(t)\rangle - \langle \psi(0)|H_{\text{kin}}|\psi(0)\rangle\) (note that we plot \(-\Delta E_{\text{kin}}\)), total spin energy (triangles) \(\Delta E_{\text{spin}} = \langle \psi(t)|H_J + H_h|\psi(t)\rangle - \langle \psi(0)|H_J + H_h|\psi(0)\rangle\), phonon energy (squares) \(\Delta E_{\text{phon}} = \langle \psi(t)|H_{\text{ph}} + H_{\text{EP}}|\psi(t)\rangle - \langle \psi(0)|H_{\text{ph}} + H_{\text{EP}}|\psi(0)\rangle\) for different values of \(\lambda = g^2/2\omega_0 t_0\), \(J = 0.3\), \(h = 0.75\) and \(\omega_0 = 1.0\). Horizontal dashed lines represent absolute values of kinetic energies, calculated in the respective ground states of the quenched Hamiltonians (with \(t_0 = 0\)). Vertical arrows indicate \(\tau_{\text{form}}\).

Expectation values of different parts of Hamiltonian in Eq. 2 during the time evolution. Immediately after the quench the kinetic energy is zero, i.e. \(E_{\text{kin}} = \langle \psi(0)|H_{\text{kin}}|\psi(0)\rangle = 0\), since the initial state was prepared using \(t_0 = 0\). During the time evolution the kinetic energy decreases and approaches its respective value of the spin-lattice polaron (SLP) in the ground state at \(t_0 = 1\). Since the total energy remains a constant after the quench, the sum of all other energies must increase. Although our system is not coupled to an external electromagnetic field, its relaxation after the \(t_0\) quench is similar to the relaxation after the absorption of the electromagnetic pulse. In both cases the essential physics consists in transforming the highly elevated kinetic energy into other excitations. This claim is supported by explicit numerical simulations in Ref. [27]. In the \(\lambda = 0\) case, the decrease of \(\Delta E_{\text{kin}}\) is exactly mirrored by the increase of the total spin energy, \(\Delta E_{\text{spin}} = \Delta E_{J} + \Delta E_{h}\), see Fig. 1(a). During the spin polaron formation, manifested in the decrease of \(\Delta E_{\text{kin}}\), excess energy is absorbed by the inelastic spin degrees of freedom, directly coupled to the hole. We refer to the time scale at which the kinetic energy saturates as the spin polaron formation time \(\tau_{\text{form}}\). It is formally obtained from fitting the kinetic energy to a functional form \(\Delta E_{\text{kin}}(t) = a[1 - \exp(-t/\tau_{\text{form}})]\). The values of \(\tau_{\text{form}}\) are indicated by vertical arrows in Fig. 1.

Switching on EP coupling adds additional degrees of freedom, coupled to the hole. The most discernible effect of increasing \(\lambda\) is the shortening of the SLP formation time \(\tau_{\text{form}}\), qualitatively consistent with the Matthiessen’s rule. For example, at small \(\lambda = g^2/2\omega_0 = 0.025\), \(\Delta E_{\text{kin}}\) decreases slightly faster in comparison to the \(\lambda = 0\) case. Moreover, the excess energy is distributed between \(\Delta E_{\text{spin}}\) and \(\Delta E_{\text{phon}} = \Delta E_{\text{ph}} + \Delta E_{\text{EP}}\), as seen in Fig. 1(b). Following more closely the time evolution of \(\Delta E_{\text{spin}}\) we observe that \(\Delta E_{\text{spin}}\) reaches a broad maximum just above \(t \gtrsim \tau_{\text{form}}\), which is followed by a gradual decrease that is matched by a slow increase in \(\Delta E_{\text{phon}}\). During this time \(\Delta E_{\text{kin}}\) remains largely unchanged. These results are consistent with a subsequent slow redistribution of the energy from spin to lattice degrees of freedom. While there is no direct coupling between the spin and the lattice sector, such redistribution can only take place via coupling to the charge. It is thus not surprising that we obtain a much longer time scale for this energy exchange process, with a very rough estimate \(t_{\text{ex}} \gtrsim 50\).

With further increasing \(\lambda\), the SLP formation time \(\tau_{\text{form}}\) further shortens and up to \(\lambda = 0.1\) the final amount of the excess energy absorbed by phonons, \(\Delta E_{\text{phon}}\), increases. In this particular case, see Fig. 1(c), we observe no change of different parts of energies after \(\tau_{\text{form}}\). A different physical picture is seen in the case when \(\lambda \gtrsim 0.2\). In this case the \(\Delta E_{\text{kin}}\) again reaches the steady state value within the initial time \(t \sim \tau_{\text{form}}\), meanwhile \(\Delta E_{\text{phon}}\) reaches a broad maximum. However, with further increasing of time, \(t \gtrsim \tau_{\text{form}}\), we observe a subsequent energy flow, which is in this case reversed in comparison to \(\lambda = 0.025\) case, i.e. from (decreasing) \(\Delta E_{\text{phon}}\) to (increasing) \(\Delta E_{\text{spin}}\). This energy transfer from lattice to spin degrees of freedom again takes place on a much longer time scale \(t_{\text{ex}}\) in comparison with the relaxation time of the kinetic energy, i.e. \(t_{\text{ex}} \gg \tau_{\text{form}}\). The amount of the subsequent energy transfer becomes more pronounced at larger \(\lambda = 0.4\). Again, during this energy transfer the kinetic energy of the SLP remains roughly unchanged. Moreover, comparing \(\Delta E_{\text{phon}}(t)\) in the long-time limit, \(t \sim 50\), for systems with increasing \(\lambda \gtrsim 0.1\), we find that the energy absorbed by the lattice saturates as \(\lambda\) increases towards \(\lambda = 0.4\).

While the subsequent energy transfer between spin and lattice degrees of freedom clearly indicates that the emission/absorption of phonons and spin excitations represents strongly interconnected processes in the second stage of relaxation, there remains an open question concerning the interdependence of these inelastic processes in the first stage of the relaxation. To gain further insight into scattering process in the first stage of relaxation we test applicability of the Matthiessen’s rule and split the SLP formation time into two possibly independent contributions:

\[
\tau_{\text{form}}^{-1}(J, \lambda) = \tau_{\text{form}}^{-1}(J, \lambda = 0) + \bar{\tau}_{\text{form}}^{-1}(\lambda).
\]

Based on the assumption of the validity of the Matthiessen’s rule \(\bar{\tau}_{\text{form}}(\lambda)\) represents the bare phonon contribution to the SLP formation time. In the case of independent scattering processes \(\bar{\tau}_{\text{form}}(\lambda)\) should remain
$J$-independent. In Fig. 2 we present $1/\tau_{\text{form}}(\lambda)$ extracted from systems with different values of $J$. Up to $\lambda \lesssim 0.2$ the values nearly overlap, signaling that the emission of phonons and local string excitations represent nearly independent processes. For larger $\lambda = 0.3$ and 0.4, we observe a slight upward deviation of $1/\tau_{\text{form}}(\lambda)$ for systems with increasing $J$.

We shall gain additional insight into this unusual relaxation dynamics by computing time-dependent change of the hole-spin and hole-phonon number correlation functions defined as:

$$C_s(t, j) = \sum_i (-1)^{i+j} \left[ \langle \psi(t)|n_i^b S_{i+j}^z|\psi(t) \rangle - \langle \psi_G|n_i^b S_{i+j}^z|\psi_G \rangle \right]$$  \hspace{1cm} (3)

$$C_{\text{ph}}(t, j) = \sum_i \left[ \langle \psi(t)|n_i^h n_{i+j}^{\text{ph}}|\psi(t) \rangle - \langle \psi_G|n_i^h n_{i+j}^{\text{ph}}|\psi_G \rangle \right],$$  \hspace{1cm} (4)

where $n_i^\text{ph} = a_i^+ a_i$ is the phonon number operator and $|\psi_G\rangle$ is the ground state wavefunction of the quenched Hamiltonian.

In Fig. 3(a) we present the density plot of the hole-spin correlation function representing the distribution of spin excitations relative to the hole position at $j = 0$ for the case of $\lambda = 0.0$. At short times, $t \lesssim \tau_{\text{form}}$, we observe a fast expansion of the front line of $C_s(t, j)$ with a well defined initial velocity approximately equal the maximal group velocity of the free particle, $v_{\text{free}} \sim 2$, indicated by a full straight line. The expansion at later times $t \gtrsim \tau_{\text{form}}$ slows down. The peak values of $C_s(t, j)$ separate from the hole position at $j = 0$ and move away with a rather well defined velocity that approximately matches the maximal magnon velocity $v_{\text{mag}} = J + h - \sqrt{h(h+2J)}$, as indicated by the dashed line.

The emerging physical picture is consistent with a two-stage spin polaron formation process. In the first stage the hole travels with the velocity not exceeding the free particle one, $v_{\text{free}} \sim 2$, and emits its excess energy by creating local spin excitations. This stage is completed in a very short time, roughly given by $\tau_{\text{form}}$, as the kinetic energy of the hole approaches the kinetic energy of the spin polaron in its ground state. An excited spin polaron is thus formed. At this point the polaron is surrounded by the excess amount of local spin excitations. In the second stage the dissipation emerges through a process where the excess spin energy is radiated away via magnon excitations while $\Delta E_{\text{kin}}$ remains nearly constant.

This effect survives as well at finite values of $\lambda$, see Figs. 3(b-f). In this case, as already seen from Fig. 1, a part of the excess kinetic energy is absorbed by the phonon subsystem, which renders less available energy for spin excitations. The most discernible effect of increasing $\lambda$ on the spin subsystem is thus the overall decrease of $C_s(t, j)$. Due to the absence of direct magnon-phonon coupling, $v_{\text{mag}}$ remains unchanged. The other noticeable effect of increasing $\lambda$ is the shortening time of the initial fast expansion of $C_s(t, j)$ with $v_{\text{free}}$.

In Fig. 4 we show the density plot of the hole-phonon number correlation function. As in the previous case, we observe a fast expansion of $C_{\text{ph}}(t, j)$ for short times, $t \lesssim \tau_{\text{form}}$ with a well defined velocity given by $v_{\text{free}}$. At longer times, $t \gtrsim \tau_{\text{form}}$, two distinct effects are observed:

FIG. 2. $1/\tau_{\text{form}}(\lambda)$ obtained from Eq. 2 using $J = 0, 0.3$ and 0.4 with $h = 0.7J$. In the case of $J = 0$ we obtain $\tau_{\text{form}}^{-1}(J = 0, \lambda = 0) = 0$ since in this case the hole behaves as a free particle which leads to an infinite relaxation time. Results for $J = 0$ were then obtained from $\tau_{\text{form}}(\lambda) = \tau_{\text{form}}(J = 0, \lambda)$.

FIG. 3. Hole-spin correlation function $C_s(t, j)$ for $J = 0.3$, $h = 0.7J$ and different values of $\lambda$. Full line indicates maximal free electron velocity $v_{\text{free}} = 2$ and the dashed line represents the magnon velocity $v_{\text{mag}} = J + h - \sqrt{h(h+2J)}$. The expansion at later times $t \gtrsim \tau_{\text{form}}$ results in a full straight line. The expansion at later times $t \gtrsim \tau_{\text{form}}$ results in a full straight line.
FIG. 4. Hole-phonon number correlation function $C_{ph}(t, j)$ (density plots (a-e) and on-site value (f)) the rest of parameters are the same as in Fig. 3. Dashed and dotted lines connect points of selected constant values of $C_{ph}(t, j)$. Full line indicates maximal free electron velocity $v_{ave} = 2$ and open circles present $j_{ave}(t)$ as explained in the text.

(i) a decrease of $C_{ph}(t, j)$ at smaller distances from the hole position, $j \lesssim 6$, as highlighted by a dashed lines in Fig. 4 and (ii) a further expansion of $C_{ph}(t, j)$ with rather well defined velocity at larger distances for $j \gtrsim 6$, emphasized by dotted lines. Since Einstein phonons possess zero group velocity, there must exist an alternative mechanism for the explanation of the observed velocity. Within a semi-classical picture the highly excited hole first slows down by creating local spin and phonon excitations thus forming an excited SLP with its kinetic energy $E_{kin}(t)$ that remains above its equilibrium ground state value, $E_{kin}(t) = \Delta_{kin}(t) + E_{kin,G}$, see also Fig. 1. The existence of the finite $\Delta_{kin}(t)$ is not solely due to finite-size effects but primarily due to a finite energy gap in the system. For this reason the relaxing SLP does not approach its exact ground state since it can not emit arbitrarily small energy quantum. Due to the finite value of $\Delta_{kin}(t)$, the average group velocity of the excited SLP $v_{ave}$ remains finite. The excited SLP in the semi-classical sense moves away from its otherwise localized phonon excitations, which in turn causes the observed expansion of $C_{ph}(t, j)$. To test this idea we estimated the averaged SLP group velocity from:

$$v_{ave} = \sqrt{\frac{1}{\pi} \int_{-\pi/2}^{\pi/2} dk \left( \frac{4E(k)}{\pi k} \right)^2},$$

where $E(k)$ is the equilibrium SLP dispersion relation computed using the quenched Hamiltonian while the average is taken over the whole AFM Brillouin zone. Note that the ground state of the SLP has $k = \pi/2$. Circles in Fig. 4 represent shifted distances $j_{ave} = tv_{ave} + j_0$. With increasing $\lambda$, $v_{ave}$ decreases since the quasiparticle band becomes narrower. All this is well reflected by $j_{ave}(t)$ in Fig. 4(a) through (e), matching rather well the expansion of $C_{ph}(t, j)$.

The subsequent decrease of the number of phonons observed in $C_{ph}(t, j)$ in the vicinity of the hole position, i.e. for $j \lesssim 6$ for $\lambda \geq 0.1$, reveals an intricate thermalization process in which the excited lattice subsystem transfers some of its excess energy to the excited SLP, which consequently thermalizes via magnon emission. The effect becomes more pronounced with increasing $\lambda$. Taking into consideration the sum-rule $\sum_j C_{ph}(t, j) = \Delta E_{ph}(t)/\omega_0$, we find this behavior consistent with the decrease of $\Delta E_{ph}(t)$ and its transfer to $\Delta E_{spin}(t)$, as seen in Fig. 1. The $C_{ph}(t, 0)$, presented in Fig. 4(f), in the regime of $\lambda \geq 0.2$ clearly shows a decrease of the average on-site phonon number towards the respective ground state value on the time-scale $t_{ex}$. The thermalization of the excess of local phonon excitations and the energy transfer between the phonon and the spin sub-system occurs on the same time-scale.

In summary, the SLP relaxes in two stages. In the first stage the highly exited hole lowers its kinetic energy by emitting local spin and phonon excitations and forms an excited SLP with a non-zero average group velocity and with the kinetic energy close to its equilibrium value. There is no essential departure from the Mattiessen’s rule in this stage hence phonons and spin excitations act as rather independent relaxation mechanisms for highly excited charges. Our results indicate that the experimentally observed times of the primary relaxation ($\sim 10 fs$) may be explained either within purely magnetic interactions or in a scenario where both phonons and spins couple to charge carriers. In the second stage of relaxation a subsequent energy transfer between phonon and spin degrees of freedom, mediated by the excited SLP, takes place. Simultaneously, SLP thermalizes by emitting magnons. Thermalization of locally excited phonons is realized in part via the energy transfer from phonons to magnons mediated by the SLP as well as by the spread of the excited phonon cloud that occurs due to a non-zero average group velocity of the SLP. The second stage relaxation time may be much longer than the first stage one.

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