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Study of two-electron jumps in relaxation of Coulomb glasses

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A long-standing debate in the theory of hopping insulators concerns the role of multi-electron transitions in the dynamics of the system. The natural assumption is that as temperature is lowered, two-electron transitions will play an increasingly important role since they provide a way of tunneling through additional energy barriers which would be energetically unfavorable as successive one-electron transitions. This was disputed in [1], but later it was seen in [2]. The reason for this discrepancy is not clear and deserves further attention. One point where the two approaches diverged was in the selection and weighting of the two-electron transitions relative to one-electron transitions. We present calculations of the transition rates to second order in the tunneling matrix element, which will be used in improved numerical studies. We compare results for only one-electron jumps with results including also two-electron jumps.

1 Introduction

Systems with localized states conduct through the variable range hopping mechanism at low temperatures. The quantitative prediction for the temperature dependence of the conductivity was found by Mott and extended by Efros and Shklovskii to include the effect of Coulomb interactions. However, these theories are based on a single-particle approximation, and neglects the effect of correlated jumps of two or more electrons. At low temperatures, the system can be trapped in metastable configurations, from which it can be difficult to escape by single-electron transitions. By a two-electron transition the system can jump out of this metastable state even when the temperature is so low that the probability of making the same transition sequentially is very low because it passes through an activated intermediate state with higher energy. Thus, one would expect the importance of many-electron jumps to increase at lower temperatures. This has also been confirmed in several studies [2,3], but it has been disputed by others [1]. The first group of papers used a method which identifies the full set of low energy states of the system, and studies the possible transitions between them. Because the number of accessible states grows rapidly with increasing temperature and system size, this method is restricted to small systems and low temperatures. Tsigankov and Efros [1] used a kinetic Monte Carlo method which does not require the full set of accessible states. It can therefore handle much larger systems and higher temperatures. However, as temperature is decreased, the algorithm slows down and becomes impractical. Thus, in the published results there is a considerable gap in temperature where none of the methods have been successful, and it is therefore difficult to compare the approaches and understand the source of the discrepancy. In addition, Tsigankov and Efros [1] criticize the previous papers [3] because they use a transition rate for two-electron transitions which does not include the Coulomb interaction between the two electrons, and is therefore unphysical. Our goal is then twofold: First, we want to calculate the transition rate for two-electron transitions from first principles by extending

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to second order in the tunneling the standard calculation for one-electron transitions [4]. Second, we want to implement this transition rate in both the numerical schemes described above, but using identical systems (the same configuration of sites and the same energy disorder) and by choosing the appropriate system size and temperature study the same parameter region using both methods. In this paper we address the first of these goals, presenting the calculation of the rate for two-electron transitions. At the end some preliminary numerical results on the energy relaxation using the kinetic Monte Carlo approach are given.

## 2 Calculation of transition rates

We start from the Hamiltonian

$$H_0 = \sum_i \phi_i c_i^\dagger c_i + \sum_{i<j} V_{ij} c_i^\dagger c_j c_j^\dagger c_i$$

(1)

describing localized electrons interacting through Coulomb forces. $c_i^\dagger$ and $c_i$ are operators creating and annihilating an electron on site $i$, $\phi_i$ is the intrinsic energy of site $i$ and $V_{ij} = \frac{1}{r_{ij}}$ is the Coulomb energy. The random energies $\phi_i$ are chosen uniformly in the interval $[-W, W]$. In principle, the summation is over the full set of sites, but since we are interested in transitions of two electrons on four sites, we can restrict the summation to these four sites. The Coulomb interaction with the rest of the sites must then be included in the site energy $\phi_i$. The configurations of two electrons on four sites are described by the states

$$|a\rangle = |1100\rangle, |b\rangle = |1010\rangle, |c\rangle = |1001\rangle$$

$$|d\rangle = |0110\rangle, |e\rangle = |0101\rangle, |f\rangle = |0011\rangle$$

(2)

where the occupation numbers refer to positions 1,2,3 and 4, respectively. We consider phonon assisted tunneling from the initial state $|a\rangle$ to the final state $|f\rangle$. This is achieved by taking the tunneling term as a perturbation to the bare Hamiltonian $H_0$ and finding the transition rate between the perturbed states due to phonons. The tunneling Hamiltonian is

$$\Delta H = \sum_{i<j} t_{ij} c_i^\dagger c_j + \text{h.c.}$$

(3)

where the tunneling amplitude $t_{ij} = e^{-r_{ij}/a}$ depends exponentially on the distance $r_{ij}$ between the sites $i$ and $j$ and $a$ is the localization radius. The initial state is to second order in the tunneling

$$|\tilde{a}\rangle = |a\rangle + \frac{t_{23} t_{14}}{E_a - E_b} |b\rangle + \frac{t_{24} t_{13}}{E_a - E_c} |c\rangle + \frac{t_{13} t_{14}}{E_a - E_d} |d\rangle + \frac{t_{14} t_{23}}{E_a - E_e} |e\rangle$$

$$+ \frac{1}{E_a - E_f} \left[ \frac{t_{23} t_{14}}{E_a - E_b} + \frac{t_{24} t_{13}}{E_a - E_c} + \frac{t_{13} t_{24}}{E_a - E_d} + \frac{t_{14} t_{23}}{E_a - E_e} \right] |f\rangle.$$  

(4)

Here we have kept only terms which contribute to the phonon assisted transitions, additional second order terms which only would give higher order terms in the transition rate have been dropped. $E_\alpha$ refers to the energy of configuration $|\alpha\rangle$. A similar expression is found for the final state.

Now we want to calculate the transition rate between these perturbed states resulting from the phonon interaction:

$$H_{\text{e-ph}} = \sum_q \sum_i c_i^\dagger c_i \left( e^{-iqr} \gamma_q b_q + \text{h.c.} \right)$$

(5)

where $b_q$ is the phonon annihilation operator and $\gamma_q$ is a numerical factor depending on the exact phonon interaction. The calculation is a direct generalization of the one found in [4] for one-electron transitions.
assume that the electron on an impurity is described by a hydrogen-like wavefunction with a localization
radius $a$ and that $qa \ll 1$ where $q$ is the wavevector of the phonon, so that

$$\langle i | e^{-iqr_i} | i \rangle = \left(1 + \left(\frac{qa}{2}\right)^2\right)^{-2} \approx 1$$

(6)

where $|i\rangle$ describes an electron on impurity $i$. We further assume that $qr_{ij} \gg 1$, which allows us to replace factors of the form $\cos qr_{ij}$ by 0 when integrating over the directions of $q$. The full squared matrix element is then

$$\langle \tilde{\alpha} | H_{e-ph} | \tilde{f} \rangle^2 = \frac{8\pi}{s^3} \gamma^2 (\Delta E) N(\Delta E) |t_{13}t_{24}|^2 (E_f - E_d + a - e_c)^2$$

$$\times \left[ \frac{1}{(E_a - E_c)^2(E_f - E_d)^2} + \frac{1}{(E_a - E_d)^2(E_f - E_c)^2} \right]$$

$$+ \frac{8\pi}{s^3} \gamma^2 (\Delta E) N(\Delta E) |t_{23}t_{14}|^2 (E_f - E_b + a - e_c)^2$$

$$\times \left[ \frac{1}{(E_a - E_b)^2(E_f - E_c)^2} + \frac{1}{(E_a - E_c)^2(E_f - E_b)^2} \right]$$

$$+ \frac{8\pi}{s^3} \gamma^2 (\Delta E) N(\Delta E) t_{13}t_{24}t_{23}t_{14} (E_f - E_d + a - e_c) (E_f - E_b + a - e_c)$$

$$\times \left[ \frac{1}{(E_a - E_b)(E_f - E_c)} + \frac{1}{(E_a - E_c)(E_f - E_b)} \right] \times \left[ \frac{1}{(E_a - E_b)(E_f - E_c)} + \frac{1}{(E_a - E_c)(E_f - E_b)} \right]$$

(7)

where $s$ is the speed of sound, $\Delta E = E_f - E_a$ and $N(\Delta E) = 1/(e^{\Delta E/kT} - 1)$ is the equilibrium phonon density. We note that in the first term, the combination

$$E_f - E_d + a - e_c = V_{12} - V_{23} + V_{34} - V_{14}.$$  

(8)

When the distance between the pairs 13 and 24 is much larger than the distances within each pair, this corresponds to the dipole-dipole interaction between the pairs. A similar statement applies to the combination $E_f - E_b + a - e_c$ in the second term.

Regarding the energy dependence, there are several points:

- Terms like $E_f - E_d + a - e_c$: these describe the dipole interaction between the pairs, and are important.

- The terms in the square brackets: The divergence of these terms at certain points reflect the limitation of the perturbation theory rather than any physical effect. Therefore we want to limit it and replace it by 1 when it is larger than 1. The size of $E_a - E_b$ is of the order of the disorder $W$ which is of order 1. Therefore the terms in the brackets are usually large, and also temperature independent, so we propose to set them equal to 1. We believe that this is of no physical consequence, and will not affect the results qualitatively.

- The factors $\gamma^2 (\Delta E) N(\Delta E)$: These depend on the particular model of phonon interaction and appear also in the transition rate of one-electron transitions. In accordance with [5] we replace it by $\min(1, e^{-\Delta E/kT})$.
We therefore arrive at the simplified expression for the transition rate

$$\Gamma_{af} = \frac{1}{\tau_0} \min(1, e^{-\Delta E/T}) \left[ |t_{13}t_{24}|^2 (E_f - E_d + E_a - E_c)^2 ight. \\
+ |t_{23}t_{14}|^2 (E_f - E_b + E_a - E_c)^2 \\
+ t_{13}t_{24}t_{23}t_{14} (E_f - E_d + E_a - E_c)(E_f - E_b + E_a - E_c) \right]$$

(9)

where $\tau_0$ is a constant which we take as our unit of time. Note that from the definition of $t_{ij}$ and Eq. (8), the expression in the square brackets depends only on the distances between the impurities and not on the energies. We note that $\Gamma_{af}$ cancels in the absence of interactions. It also reduces to the expression in [1] when the transition involves two short jumps very far apart and represents an improvement for compact excitations, which we believe are more important.

![Energy relaxation as function of time at three different temperatures: (a) 0.01, (b) 0.005, and (c) 0.001. Thin curves include only one-electron transitions while thick curves include one- and two-electron transitions. The different curves correspond to different random sequences in the Monte Carlo algorithm.](image-url)
3 Preliminary numerical results

The transition rate (9) was implemented together with one-electron transitions in a Monte Carlo algorithm similar to the one introduced by Tsigankov and Efros [1]. A random transition, involving one or two electrons is chosen, weighted by the distance dependent part of the transition rate. Then the jump is accepted or rejected based on the energy dependent part of the transition rate. We use a two-dimensional square lattice of $100 \times 100$ sites with half filling. The localization radius $a$ is equal to one lattice spacing and we have used $W = 1$ in units of the nearest neighbor Coulomb interaction. We show here some preliminary results on the relaxation of the energy. The system is initialized in some random configuration (the same in all the simulations shown here, the disorder is also identical in all cases), and the time evolution of the total energy is followed as a function of time. Fig. 1 compares the energy relaxation with and without two-electron transitions at three different temperatures. We see that as expected there is no clear difference at the highest temperature ($T = 0.01$, note however that this is already below the temperature considered in [1]). At lower temperatures it is clear that the simulations including two-electron transitions on average relax faster.

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