Role of impact ionization in the thermalization of photo-excited Mott insulators

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We study the influence of the pulse energy and fluence on the thermalization of photo-doped Mott insulators. If the Mott gap is smaller than the width of the Hubbard bands, the kinetic energy of individual carriers can be large enough to produce doublon-hole pairs via a process analogous to impact ionization. The thermalization dynamics, which involves an adjustment of the doublon and hole densities, thus changes as a function of the energy of the photo-doped carriers and exhibits two timescales – a fast relaxation related to impact ionization and a slower timescale associated with higher-order scattering processes. The slow dynamics depends more strongly on the gap size and the photo-doping concentration.

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I. INTRODUCTION

The photodoping of a Mott insulator provides a relatively simple way to induce and study a nonequilibrium phase transition. If a laser pulse with a frequency comparable to the gap is applied to a Mott insulator, doublon-hole pairs are produced, and these mobile carriers lead to a metallic response of the photodoped system. The changes in the optical conductivity associated with this metallization have been studied using time-resolved spectroscopy. In the pioneering work by Iwai and collaborators on a Ni-chain compound, a Drude peak in the conductivity was measured promptly after the photodoping pulse, and the metallic state was found to last for a few picoseconds.

One can distinguish at least two mechanisms which play a role in the relaxation of photo-doped carriers. On the one hand, electron-electron scattering can lead to a thermalization of the electronic subsystem at a hot “electron temperature,” and on the other hand, carriers dissipate their initially high kinetic energy through scattering with “external” degrees of freedom like spins or phonons. A large body of theoretical work on photo-doped Mott insulators has focused on the latter relaxation processes involving the scattering with spins in an antiferromagnetic background or the coupling to phonons. In this paper we assume that electron-electron scattering is the fast mechanism, so that we can study the thermalization of isolated electrons and neglect the energy loss processes which affect the dynamics only on longer times; the limits of this assumption will be discussed below.

In metals, a rapid thermalization of the electronic system is typically observed and underlies the assumption that a quasi-equilibrium picture or two temperature model can be used to describe the dynamics already at very short times after an excitation. In an insulator, the thermalization and relaxation involves an adjustment in the number of electron-hole pairs, which can be a slow process in the presence of a large gap. It was found that in a purely electronic model (paramagnetic Hubbard model with on-site repulsion $U$) the thermalization time depends exponentially on the gap size, and that even the relaxation of the distribution of photodoped carriers within the Hubbard bands can be extremely slow. The explanation for this is relatively simple. If the energy $U$ which is needed for the production of a single doublon-hole pair is substantially larger than the typical kinetic energy of a single doublon or hole, complicated multiparticle scattering processes are needed for thermalization – hence the exponential scaling with $U$. (For a similar reason, the doublon-hole recombination via emission of magnons or phonons becomes slow when the gap is large.)

The same physics can be expected to manifest itself if we consider a fixed interaction (gap size) but different pulse energies. If the gap size is smaller than the width of the Hubbard bands, one may anticipate a strong dependence of the relaxation dynamics on the kinetic energy of the photo-doped carriers. A carrier which is inserted at the upper band edge then has a sufficient kinetic energy that a two-particle scattering can lead to the creation of an additional doublon-hole pair, a process which we will call “impact ionization.” If, on the other hand, the carriers are inserted at the lower band edge, their kinetic energy is not sufficient for impact ionization, and the doublon-hole production relies on rare multiple-particle scattering events.

Since impact ionization processes can to a large extent determine the number of mobile carriers in the photo-induced metal, an understanding of this physics is crucial for possible applications of photo-induced metal-insulator transitions in ultra-fast switches, or for the efficient operation of photovoltaic devices. In this paper we focus on the paramagnetic Mott-Hubbard insulator with a relatively small gap, and study in more detail the electronic thermalization processes, with the goal of disentangling the fast impact ionization channel from slower thermalization processes. Experimentally, the two relax-
ation mechanisms can be distinguished by signatures in the time-resolved photo-emission spectrum, and possibly also by considering their fluence dependence: impact ionization processes involve single doublons or holes and the associated relaxation time should thus not depend strongly on the density of photo-doped carriers, while higher order scattering processes will become more frequent if the density of carriers increases.

II. MODEL AND METHOD

We investigate and quantify the effect of impact ionization by considering a Hubbard model

\[
H = \sum_{ij,\sigma} v_{ij} c_i^{\dagger} c_j + U \sum_i \left( n_{i\uparrow} - \frac{1}{2} \right) \left( n_{i\downarrow} - \frac{1}{2} \right)
\]

with on-site interaction \( U \) comparable to the bandwidth. The operators \( c_{i\sigma} \) create an electron at site \( i \) with spin \( \sigma \), and the hopping amplitude is \( v_{ij} \). The model is solved on an infinite-dimensional hypercubic lattice using nonequilibrium dynamical mean field theory (DMFT)\textsuperscript{12,13} with a strong-coupling perturbative impurity solver (non-crossing approximation, NCA).\textsuperscript{14} This lattice has a Gaussian density of states, \( \rho(\epsilon) = 1/\sqrt{\pi W} \exp(-\epsilon^2/W^2) \), and we use the width \( W \) as our unit of energy. To simulate the photo-dipole pulse, we apply a few-cycle electric field pulse of the form

\[
E(t) = E_0 e^{-(t-t_p)^2/\sigma^2} \sin(\Omega(t-t_p))
\]

with \( t_p = 6 \) and \( \sigma^2 = 6 \) in the body-diagonal of the lattice. We use a gauge without scalar potential, so that the field is given by the time derivative of the vector potential \( A \), \( E(t) = -\partial_t A(t) \). Using the Peierls substitution, the field then enters the Hamiltonian via a time-dependent shift of the dispersion, \( \epsilon_k \rightarrow \epsilon_k - A(t) \), where \( \epsilon_k \) is the Fourier transform of the hopping matrix. For details of our implementation of the nonequilibrium DMFT equations, the treatment of the electric field and the NCA impurity solver, we refer to Refs. 11 and 16.

The relaxation dynamics of photo-doped carriers depends crucially on whether or not the Mott insulator is antiferromagnetically ordered.\textsuperscript{5,18} In the present study, we restrict the calculations to the paramagnetic phase, and briefly discuss the effect of magnetic correlations in the last section. Our results hence describe the short-time electronic relaxation of photodoped Mott insulators at elevated temperature.

III. RESULTS

1. Pulse-frequency dependence

For the purpose of orientation we first plot the equilibrium spectra for different values of \( U \) and inverse temperature \( \beta = 5 \) (Fig. 1). The gap opens at \( U \approx 2.5 \) and then grows approximately linearly with \( U \). In the insulating phase, the shape of the Hubbard bands is almost independent of \( U \), and they have a width of about 3. Since an impact ionization process involves the scattering of a doublon at the upper edge of the upper band to the lower edge and a simultaneous doublon-hole excitation, it already becomes clear from the equilibrium spectra that we can only expect these processes to be relevant for interactions \( U \lesssim 4.5 \). For larger \( U \) the energy associated with scattering between states within the band is not enough to excite electrons across the gap. In the following we will thus focus on the interaction range \( 2.5 \leq U \leq 4 \).

For \( U = 2.5 \) and 3.5, the time evolution of the photodoped doublon density \( D \) after pulses with frequencies in the range \( 1.5\pi/2 \leq \Omega \leq 3.5\pi/2 \) is plotted in the top panels of Fig. 2. Here and in the following, we use the definition \( D(t) = d(t) - d(0) \), where \( d(t) \) is the time-dependent expectation value of the doublon occupancy. The amplitude \( E_0 \) of each pulse has been adjusted such that at \( t = 12 \), shortly after the pulse, the density of photodoped doublons is 0.01, and we normalize the curves by this initial density. We see that during the thermalization process, the number of doublons increases, i.e., excess kinetic energy of the photodoped carriers is transformed into interaction energy. The thermal reference value can be calculated by measuring the energy \( E_j = \int dt j(t) \cdot E(t) \) injected into the system by the pulse. Here, \( j = \sum_k n_{k\sigma} v_k(t) \) is the current, with \( n_{k\sigma}(t) = -i G_{k\sigma}^< (t, t) \) and \( v_k(t) = \partial_k \epsilon_k - A(t) \). By comparing the total energy after the pulse to that of an equilibrium system, we can compute the temperature and double occupancy which the system will reach, assuming thermalization, in the long-time limit. The thermal values of the double occupancy are indicated by the dashed horizontal lines.

If we fit the doublon curves in the range \( 30 \leq t \leq 60 \) to a single exponential \( a + b \exp(-t/c) \) we obtain the relaxation times \( c \) plotted in the bottom panel of Fig. 2 and the long-time values \( a \) indicated by the arrows in
FIG. 2: Relaxation after pulse excitations with different frequencies and initial inverse temperature $\beta = 5$. The amplitude of the pulses is adjusted such that the number of photo-doped doublons at $t = 12$ (shortly after the pulse) is 0.01. Top panels: time evolution of the normalized doublon density and expected thermal values (horizontal lines) for $U = 2.5$ and 3.5. Bottom panel: relaxation times ($c$) obtained by fitting $D(t)/D(t = 12)$ to the function $a + b \exp(-t/c)$ in the range $t \in [30, 60]$. The extrapolated long-time values ($a$) for $U = 3.5$ are indicated by arrows in the upper panels.

In order to show direct evidence for impact ionization we plot in Fig. 3 time resolved spectral functions $A^<(\omega, t) = \frac{1}{\pi} \text{Im} \int_{-\infty}^{\infty} dt' e^{i\omega (t' - t)} G^< (t', t)$, which are related to a time-resolved photoemission spectrum. Spectra are computed for a photo-doping pulse with $\Omega = 4\pi/2$ in the model with $U = 4$ (the initial doping concentration is 0.005). This pulse inserts the doublons at the upper edge of the upper Hubbard band. As time increases, the weight of the peak near the upper band edge decreases, while a second peak near the lower band edge starts to grow. The energy difference between the two peaks matches the energy difference between the lower peak and its symmetric counterpart in the hole distribution function (see solid black arrows). In other words, the bandwidth, gap size and pulse energy in this calculation are such that impact ionization processes can be
expected to play a role in the initial relaxation dynamics. That these processes are indeed to a large extent responsible for the doublon production follows from the lower panel, which plots the difference in the spectra at a given time and at time $t = 12$, shortly after the pulse.

If we integrate over the frequency range corresponding to positive or negative values, respectively, we find that the gain in low-energy doublons is almost exactly twice as large as the loss in high-energy doublons. The precise numbers for the ratio are 2.08 ($t = 18$), 2.05 ($t = 24$) and 2.04 ($t = 30$). The natural interpretation is that each decaying high-energy doublon produces one additional low-energy doublon via impact ionization.

2. Two-step thermalization

At least in cases such as the set-up discussed above, where the high-energy and low-energy carriers can be relatively clearly separated, one can try to reproduce the time evolution of the doublon population with a simple model that describes the decay of the high-energy doublons via impact ionization with a relaxation time $\gamma$, and the higher order scattering processes with a different associated thermalization time $\tau$. We denote the slow processes with a subscript “therm” and the fast ones with “imp” and split the total doublon number $D$ into a high and low-energy population $D_1$ and $D_2$, respectively. After thermalization, we assume that only low-energy doublons are present, and denote their number by $D_{th}$. The time evolution is then given by the equations

$$\frac{dD_1}{dt}_{\text{imp}} = -\frac{1}{\gamma}D_1,$$

$$\frac{dD_2}{dt}_{\text{imp}} = -2\left(\frac{dD_1}{dt}_{\text{imp}}\right),$$

$$\frac{dD_2}{dt}_{\text{therm}} = \frac{1}{\tau}(D_{th} - D_2).$$

The factor two in Eq. (4) accounts for the production of two low-energy-doublons per decay of a high-energy doublon in an impact ionization process. The equations governing the time evolution of the two components thus read

$$\frac{dD_1}{dt} = -\frac{1}{\gamma}D_1, \quad \frac{dD_2}{dt} = -\frac{1}{\tau}(D_{th} - D_2) + \frac{2}{\tau}D_1,$$

and the solution for the total doublon population for times $t > t_s$ becomes

$$D_{th} - D(t) = D_1(t_s)e^{-(t-t_s)/\gamma}$$

$$+ (D_{th} - D(t_s) - D_1(t_s))e^{-(t-t_s)/\tau}.$$  

Here, $t_s$ is some time after the pulse (we choose $t_s = 15$ in the following analysis), $D_{th}$ and $D(t_s)$ are known, while $D_1(t_s)$, $\gamma$ and $\tau$ must be obtained by fitting. (An upper bound for $D_1(t_s)$ can be obtained from the comparison of the time-resolved spectral function shortly after the pulse, e.g., at $t = 12$, to the thermal distribution function. This estimate yields $D_1(t_s)/D(t_s) < 0.61$ (< 0.60)

for $U = 4$, $\Omega = 4\pi/2$ and initial doping $D(t_s) = 0.005 (0.010)$.)

For $U = 2.5$ the relaxation is well described by a single exponential. This follows already from the data in the top left panel of Fig. 2 which show that the extrapolated long-time values from an exponential fit in the range $t \in [30, 60]$ correctly predict the thermal doublon density. For $U = 3$ and $3.5$, the fitting of the data with the double-exponential decay [5] works rather well. We summarize the results of this analysis in Tab. I. One finds fast relaxation times $\gamma \sim 15$ and slow relaxation times $\tau \sim 60$ for $U = 3$, and fast (slow) relaxation rates of approximately 40–50 (250–350) for $U = 3.5$. The (relative) initial excited population decreases as the pulse frequency is lowered, in rough agreement with the time-resolved spectra. At the lowest pulse frequencies considered, the separation between high-energy and low-energy populations becomes blurred and our model fit becomes less meaningful. For $U = 4$, all relaxation times become rather long, and it is difficult to obtain reliable fits. We find $\gamma \approx 90$ and $\tau \approx 6000$ (with a large uncertainty), and an initial excited population of about $D_1(t_s)/D(t_s) \approx 0.4$, consistent with the upper bound of 0.6 estimated from the spectral function.

While one should probably not consider more than the first digit of the relaxation times and initial high-energy populations in Tab. I, our model does provide a consistent description of the doublon relaxation, and the results demonstrate that impact ionization processes play a significant role in the interaction range $3 \leq U \leq 4$. In particular, they lead to a two-step thermalization with a fast initial doublon production and an associated transfer of spectral weight from the upper to the lower band edge, followed by a much slower thermalization of the relaxed distribution. The slow timescale $\tau$ grows fast with increasing $U$, which is consistent with a previous analysis based on single exponential fits [11]. However, also the relaxation time $\gamma$ associated with the impact ionization increases with $U$, which indicates that these pro-

| $U$ | $\Omega$ | $D_{th} - D(t_s)$ | $D_1(t_s)$ | $\frac{D_1(t_s)}{D(t_s)}$ | $\gamma$ | $\tau$ |
|-----|--------|----------------|-----------|-------------------|-------|------|
| 2.5 | $\frac{3\pi}{2}$ | 0.00448 | 0.000343 | 0.034 | 7.20 | 18.8 |
| 2.5 | $\frac{3\pi}{2}$ | 0.00421 | 0.000273 | 0.027 | 7.75 | 19.0 |
| 2.5 | $\frac{3\pi}{2}$ | 0.00348 | 0.000207 | 0.021 | 9.35 | 19.6 |
| 3 | $\frac{3\pi}{2}$ | 0.00684 | 0.00129 | 0.13 | 13.4 | 60.3 |
| 3 | $\frac{3\pi}{2}$ | 0.00674 | 0.00117 | 0.12 | 15.0 | 61.4 |
| 3 | $\frac{3\pi}{2}$ | 0.00573 | 0.000810 | 0.08 | 16.5 | 64.9 |
| 3.5 | $\frac{3\pi}{2}$ | 0.00789 | 0.00359 | 0.34 | 44.0 | 376 |
| 3.5 | $\frac{3\pi}{2}$ | 0.00669 | 0.00223 | 0.22 | 48.4 | 257 |

(4 | $\frac{3\pi}{2}$ | 0.00820 | 0.00387 | 0.39 | 86.9 | 5990 )

TABLE I: Relaxation times and initial excited populations $D_1(t_s)$ extracted from fits to model [6] in the range $t \in [15, 60]$ ($t_s = 15$). The doping concentration after the pulse is $D(t_s) = 0.010$ in all cases.
cesses become less likely as the energy cost of producing a doublon-hole pair increases. This is also expected, since the kinetic energy of the photo-doped carriers is bounded, and this bound is essentially independent of $U$.

An instructive way to illustrate the two-step relaxation is to plot the doublon production rate $(d/dt)D(t)$ as a function of the deviation of the doublon density from the thermal value, $D_{th} - D(t)$. In this case, our model predicts a crossover from a small linear slope (corresponding to the slow long-time thermalization process) to a steeper slope (corresponding to the impact ionization processes). Indeed, for $U \gtrsim 3$, the data sets for different pulse energies fall roughly onto a single curve which describes such a crossover. For the data sets corresponding to the highest pulse frequencies, we plot the fits to model (6) by dashed lines. These fits also roughly reproduce the relaxation for the other pulse frequencies, which shows that the model provides a consistent description of the thermalization process.

In the lower panel of Fig. 4 we show the time-evolution for $U = 3.5$, $\Omega = \frac{3\pi}{2}$, as predicted by the model (parameters from Tab. II). One can clearly see the two-step relaxation to the thermal value (dashed line), with a rapid initial increase of the doublon density, linked to impact ionization, followed by a much slower thermalization.

We also note that in this set-up (small-gap insulator excited by a pulse which inserts the doublons at the upper edge of the Hubbard band), the thermalization leads to almost a factor of two increase in the number of doublons (see also the third column of Tab. II).

3. Fluence dependence

The impact ionization processes can be distinguished from the slower thermalization processes also by analyzing the dependence of the relaxation times on the photo-doping concentration, or fluence. Since impact ionization involves only a single doublon or hole, we expect a weak fluence dependence of the fast relaxation time $\gamma$. On the other hand, the higher-order scattering processes involving several doublons and/or holes should exhibit a stronger dependence on the photo-doping concentration, so that we expect an increase in the slow relaxation time $\tau$ as the pulse amplitude is decreased.

We analyze the fluence dependence of the relaxation for $U = 3$, $\Omega = 3.5\pi/2$ and pulse amplitudes ranging from 0.25 to 6. The doping concentrations in the thermalized state and at $t = 15$, shortly after the pulse, are given in Tab. II. For small pulse amplitude, the number of photo-doped carriers grows proportional to the square of the pulse amplitude, as expected. The thermalization in this regime leads to more than a doubling of the mobile carriers. For pulse amplitudes $\gtrsim 2$, the number of carriers grows more slowly than the power of the field pulse, and also the relative increase of the doublon population associated with thermalization is lower. To avoid complications due to strongly non-linear absorption processes, we do not consider higher amplitudes.

The top panel of Fig. 4 shows the corresponding time evolution of the doublon concentration, normalized at $t = 15$. The results for amplitudes smaller than 2 all collapse onto a single curve. This shows that in the ini-

![FIG. 4: Top panel: doublon production rate plotted as a function of $D_{th} - D(t)$ for different pulse energies and indicated values of $U$. Fits of model (6) to the curves corresponding to the highest pulse energy are shown by the dashed lines. Bottom panel: Time evolution of the normalized doublon population as predicted from the fit to model (6) for $U = 3.5$, $\Omega = 3.5\pi/2$ ($t_s = 15$).]

| Fluence $t_s$ | $D(t_s)$ | $D_{th}$ | $\beta_{th}$ | $\gamma$ | $\tau$ |
|---------------|----------|---------|-------------|---------|------|
| 0.25          | 0.000108 | 0.000236| 4.884       | 19.9    | 214  |
| 0.5           | 0.000429 | 0.000917| 4.593       | 19.5    | 194  |
| 1             | 0.00167  | 0.00334 | 3.879       | 18.3    | 147  |
| 2             | 0.00593  | 0.0105  | 2.854       | 15.6    | 85.0 |
| 6             | 0.0165   | 0.0252  | 1.996       | 11.2    | 46.1 |

TABLE II: Relaxation times and initial populations extracted from fits to model (6) in the range $t \in [15, 60]$ for $U = 3$, $\Omega = 3.5\pi/2$, $t_s = 15$ and indicated pulse amplitudes.
In the antiferromagnetic phase, the model for $D(t)$ should therefore include an additional relaxation time $\kappa$ associated with the cooling of the population $D_1$, and since this cooling conserves the number of doublons (there are no excitations across the gap associated with these spin-flip or phonon scatterings), the decrease in the population $D_1$ should lead to a corresponding increase in the relaxed population $D_2$. Equation (3) would thus have to be modified as $(dD_1/dt)_{\text{imp+cool}} = (-1/\gamma - 1/\kappa)D_1$, and Eq. (4) as $(dD_2/dt)_{\text{imp+cool}} = (2/\gamma + 1/\kappa)D_2$. The efficient dissipation of kinetic energy and the associated rapid decrease in the high-energy population in an antiferromagnetic system is expected to have a significant effect on the thermalization dynamics in small gap Mott insulators. It reduces the effectiveness of the impact ionization.

IV. DISCUSSION AND CONCLUSION

In this study, we considered the thermalization dynamics after a photo-doping pulse in a purely electronic model without magnetic order. Our data analysis based on Eqs. (3)–(5) assumed that the high-energy doublon population $D_1$ decays only via impact ionization processes. This assumption is reasonable if the timescale $\gamma$ associated with impact ionization is much shorter than the thermalization timescale $\tau$, and if other mechanisms such as the cooling of hot carriers through scattering with phonons or magnons can be neglected. For a bandwidth $W$ of the order of 1 eV, corresponding to a unit of time of 0.66 fs, the fast relaxation times in Tab. I are of the order of 5-70 fs. In a Mott insulator with strong electron-phonon coupling, the cooling rate associated with electron-phonon scattering can be of the same order of magnitude. Furthermore, the lattice excitations can modify the accessible electronic states and thus have a profound influence on the doublon life-time and the thermalization pathway. Such effects are obviously not captured by the Hubbard model considered in this study, so that our results are only valid if the electron-phonon coupling is weak enough that it does not affect the dynamics on the abovementioned timescales.

However, even in a purely electronic system, additional relaxation processes may come into play. At low temperature, in the magnetically ordered phase, spin-flip scattering provides a particularly efficient dissipation channel, which can lead to a fast redistribution of spectral weight within the Hubbard bands. Exact diagonalization based studies of the motion of a single carrier in an antiferromagnetic background suggest that the excess kinetic energy of a photo-doped carrier is transferred to the spin background within a few hopping times and also recent DMFT studies of photodoped antiferromagnetic Mott insulators revealed a very fast cooling of the photo-carriers. In the antiferromagnetic phase, the model for $D(t)$ is due to impact ionization.

FIG. 5: Time evolution of the doublon concentration for $U = 3$, $\Omega = 3.5\pi/2$ and different pulse amplitudes. Top panel: Normalized doublon population. Bottom panel: Relaxation of the doublon concentration to the thermal value. The curves for amplitude < 6 are multiplied by an arbitrary factor, to enable a better comparison of the long-time behavior. Dashed lines are fits to model on the time-interval $t \in [15, 60]$. 

The estimated value of $\gamma \approx 20 \ll \tau$ implies that the initial fast increase of the doublon population evident in the upper panel of Fig. 5 is due to impact ionization.
The main finding of this study is that in situations where the gap-size is smaller than the width of the Hubbard bands, the kinetic energy of the photo-doped particles can be large enough that impact ionization processes play an important role in the initial relaxation. In fact, for the largest interactions considered ($U = 3.5 - 4$), the doublon-hole production on the computationally accessible timescales is almost entirely due to impact ionization processes. We have demonstrated this by analyzing the time-resolved photoemission spectrum, and by extracting the impact-ionization and thermalization timescales from fits to a model with two exponentials, which was found to provide a rather good description of the time evolution of the doublon density. These timescales depend on the gap size, with the slow timescale (related to higher order scattering processes) growing much more rapidly with gap size than the fast one (related to impact ionization), while the pulse frequency mainly affects the relative population of high-energy carriers which can trigger impact ionizations. The two timescales also exhibit a different dependence on the pulse amplitude (or density of photo-doped carriers), with the impact ionization process becoming insensitive to the doping concentration in the small-doping regime, while the slow timescale grows rapidly with decreasing fluence.

The impact ionization process could be relevant in Mott solar cell applications, because it can lead to an approximate doubling of carriers on a timescale which may be much shorter than the timescale associated with the dissipation of energy to phonons. This could provide a mechanism to harvest the energy of photons in a broader frequency range. However, the impact ionization processes are efficient only in Mott insulators with a small gap relative to the width of the Hubbard bands. Whether or not these processes can contribute significantly to the power produced by Mott solar cells such as the LaVO$_3$ hetero-structure proposed in Ref. 19 remains an open question. To address this issue one would have to consider a realistic set-up, and also study the diffusion of the photo-doped carriers to the leads, the effect of the spin background, and the coupling to phonons.

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