Dielectric Breakdown of Mott Insulators in Dynamical Mean-Field Theory

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Using nonequilibrium dynamical mean-field theory, we compute the time evolution of the current in a Mott insulator after a strong electric field is turned on. We observe the formation of a quasistationary state in which the current is almost time independent although the system is constantly excited. At moderately strong fields this state is stable for quite long times. The stationary current exhibits a threshold behavior as a function of the field, in which the threshold increases with the Coulomb interaction and vanishes as the metal-insulator transition is approached.

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Nonequilibrium phase transitions and nonlinear transport are becoming central issues in the study of strongly correlated systems. One of the most basic phenomena is the dielectric breakdown (destruction of insulating states due to strong electric fields) [1–4]. In Mott insulators, the electron motion is frozen as a result of strong repulsive interactions [5], and in equilibrium the doping of carriers into a Mott insulator leads to interesting quantum states such as high-T_c superconductivity in two-dimensional materials. Thus, it is natural to ask how nonequilibrium carriers behave when electrons in a Mott insulator start to move in response to strong electric fields. Experimentally, the physics of nonlinear transport in correlated electron systems has been studied in oxides [1] as well as in organic materials [6]. In one-dimensional Mott insulators, dielectric breakdown was observed, and it was found that the current increases with a threshold behavior [1]. The current-voltage (I-V) characteristics exhibit a strong nonlinearity with a negative differential resistivity between the weak current and large current regimes. More recently, the problem of nonlinear transport has also attracted interest in the cold atom community, where a novel realization of the Mott-insulating state has been achieved. In Ref. [7] the effect of a potential gradient was studied to probe the excitation spectrum.

A theoretical description of nonlinear transport is challenging because one needs to take into account two nonperturbative effects, electric fields and electron-electron interactions, simultaneously. In one-dimensional systems, reliable numerical techniques such as exact diagonalization and the time-dependent density matrix renormalization group are available, and a threshold behavior in the current was indeed observed [2]. A many-body Schwinger-Landau-Zener mechanism, in which doubly occupied states (doublons) and holes are pair produced by quantum tunneling, was proposed as an explanation [3]. In these studies, a direct calculation of the I-V characteristics is absent since a steady state current cannot be easily reached in a finite system. On the other hand, the current through a thin Mott-insulating layer coupled to leads at fixed temperature was computed in Ref. [8], and a strongly nonlinear I-V characteristic was found. Another totally unexplored issue is the temperature effect, where experiments suggest a relatively strong temperature dependence of the current.

The purpose of this Letter is to address these questions using dynamical mean-field theory (DMFT) [9], which is suitable for the study of high-dimensional bulk systems. Nonequilibrium DMFT [10] has been used to reveal various types of steady states and relaxation phenomena in the Falicov-Kimball model [11–13] and the Hubbard model [14]. For the current analysis one must compute the dynamics of the Hubbard model at rather strong interactions up to relatively long times. So far, this task has been prohibitively difficult for impurity solvers based on real-time quantum Monte Carlo simulations [14], but it has become accessible recently through an implementation of the self-consistent hybridization expansion within the Keldysh framework [15].

In the following we focus on the Mott-insulating phase in the half filled Hubbard model on a d-dimensional cubic lattice with lattice spacing a,

$$H = \sum_{\langle ij\rangle\sigma} V_{ij}(t) c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i \Big(n_{i\uparrow} - \frac{1}{2}\Big)\Big(n_{i\downarrow} - \frac{1}{2}\Big). \quad (1)$$

Here $c^\dagger_{i\sigma}$ ($c_{i\sigma}$) denotes the creation (annihilation) operator for an electron with spin σ at lattice site $\boldsymbol{R}_i$, U is the local Coulomb repulsion, and V_{ij} describes hopping between the sites. To study the dielectric breakdown of the Mott insulator, we initially prepare the system in thermal equilibrium at temperature $T = 1/\beta$ and apply a spatially homogeneous electric field $\boldsymbol{F}(t)$ for time $t > 0$. Using a gauge with pure vector potential $\boldsymbol{A}(t)$, i.e., $\boldsymbol{F}(t) = -\partial_t \boldsymbol{A}(t)/c$, $\boldsymbol{F}(t)$ is incorporated into Eq. (1) by means of the Peierls substitution, $V_{ij}(t) = V^0_{ij} \exp[ie(\boldsymbol{R}_j - \boldsymbol{R}_i)\boldsymbol{A}(t)/\hbar c]$. The field is chosen to point along the body diagonal $\hat{\boldsymbol{\eta}} = (1 \ldots 1)^t$ of the unit cell. It is turned on to a value F within a switching time t_0, $\boldsymbol{F}(t) = \hat{\boldsymbol{\eta}} F r(t/t_0)$, using a switching profile $r(x) = \frac{1}{2} - \frac{3}{4}\cos(\pi x) + \frac{1}{4}\cos(\pi x)^3$ for $0 \le x \le 1$. The main results turn out to be independent of the switching, and we choose $t_0 = 3$ if not stated otherwise.

In the limit $d = \infty$ [16] the problem is solved exactly using nonequilibrium DMFT. The DMFT self-consistency equations for finite electric field are identical for the Hubbard model and the Falicov-Kimball model, and they are detailed in Ref. [11]. The DMFT single-site problem, on the other hand, is identical to the zero field case, and we solve it by means of the self-consistent hybridization expansion [15] up to either first order [noncrossing approximation (NCA)] or second order [one-crossing approximation (OCA)]. The latter is known to be reliable for the insulating phase and the crossover regime [15,17]. We choose units of energy such that the density of states is given by $\rho(\epsilon) \propto \exp(-\epsilon^2)$, with a width (second moment) $W = \sqrt{2}/2$. Time and field are measured in units of $1/\sqrt{2}W$ and $\sqrt{2}W/ea$, respectively ($\hbar = 1$). In equilibrium, a first-order Mott transition is found in the paramagnetic phase [9], with a critical end point at $U_c \approx 3.1$ and $T_c \approx 0.02$ (determined within OCA).

After an electric field is turned on in the Mott-insulating phase, we either observe the formation of a quasistationary state with time-independent current [Fig. 1(a)] or, for very large values of F, the emergence of Bloch oscillations [$F = 10$ in Fig. 1(a)], whose period approaches $2\pi/F$ for large F after the decay of the transient behavior. In analogy to the Falicov-Kimball model [11], where Bloch oscillations are quenched by the interaction, there is no sharp separation between oscillatory and nonoscillatory regimes, but instead the current behaves irregularly at intermediate fields. In the following, we only study the quasistationary state, which will reveal the dielectric breakdown of the Mott insulator at moderately large, possibly experimentally accessible fields.

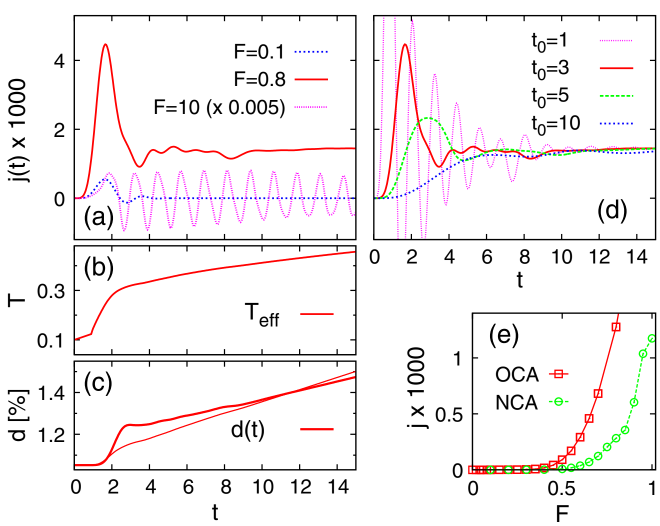

FIG. 1 (color online). Time evolution after turning on a field F in the insulator ($U = 5$, $\beta = 10$). (a) The current $j(t)$; results for $F = 10$ are multiplied with a factor 0.005 to match the scale. (b) Effective temperature T_{eff} and (c) double occupancy $d(t)$ for $F = 0.8$. The thin line shows the thermal expectation value of d at temperature $T = T_{\text{eff}}(E(t))$. (d) Current at $F = 0.8$ for various switch-on times t_0. (e) Current, averaged for $t \ge 10$, obtained using either NCA or OCA.

Since our system is not coupled to a thermal bath, the energy $E = \langle H(t)\rangle$ increases at a rate $\dot{E} = F(t)j(t)$, and thus a stationary state with nonzero current cannot exist forever. However, we find that $j(t)$ remains remarkably stable even after a considerable energy increase. This fact becomes clearly evident if one looks at the effective temperature instead of the energy, i.e., the temperature $T_{\text{eff}}(E)$ after a hypothetical thermalization at energy E: For $F = 0.8$ in Fig. 1, e.g., T_{eff} increases by a factor of 1.5 during times $4 < t < 15$ [Fig. 1(b)], while $j(t)$ remains almost constant. The saturation of the current and a simultaneous increase in the double occupancy $d = \langle n_{i\uparrow}n_{i\downarrow}\rangle$ [Fig. 1(c)] indicate that the current flow causes excitations in the system that are immobile, just like the spin fluctuations in the ground state that lead to a finite double occupancy but no linear response conductivity. These excitations do not thermalize on the time scale of our simulation. Otherwise the current would increase thermally and $d(t)$ would have to match its expectation value in thermal equilibrium at temperature $T_{\text{eff}}(E(t))$ [thin line in Fig. 1(c)], which is not the case. This behavior is consistent with recent experiments on ultracold gases [18], where it was found that artificially created double occupancies in the Mott insulator relax only on exponentially long time scales.

The quasistationary current turns out to be more or less independent of how the field is turned on, and by increasing the switching time t_0 one can reduce the otherwise rather strong transient oscillations [Fig. 1(d)]. Even for slow switching, however, the transient current can be orders of magnitude larger than the stationary current. This fact is already entailed in the linear response relation $j(t) = \int_0^t ds\sigma(s)F(t-s)$, which always holds for small enough times. The transient is at least proportional to F, while the long-time limit can be exponentially small (see below). In particular, in the Mott insulator at $T = 0$ the dc conductivity $\sigma_{\text{dc}} = \int_0^\infty ds\sigma(s)$ vanishes, while the integral $\alpha = \int_0^\infty dt \int_0^t ds\sigma(s)$ over the current yields a nonzero static polarizability.

After averaging over times $t \ge 10$, the quasistationary current $\bar{j}$ at $U = 5$ shows a sharp increase around $F = 0.5$ [Fig. 1(e)], which is the hallmark of the dielectric breakdown. We note that both first-order (NCA) and second-order (OCA) implementations of the self-consistent hybridization expansion yield similar results, but in analogy to equilibrium calculations the insulating behavior is overestimated by NCA, such that the increase of the current is shifted to stronger fields. In the following we stick to the more reliable OCA as an impurity solver.

A plot of the conductance $\bar{j}/F$ on a logarithmic scale reveals a crossover from the temperature-dependent linear response conductivity $\sigma_{\mathrm{dc}}(T)$ at small F to an almost temperature-independent curve at large F [Fig. 2(a)]. These data suggest that the quasistationary current has a nonzero $T = 0$ limit, which reflects the ground state decay, or dielectric breakdown of the insulator. In analogy to the one-dimensional case [2,3], we will refer to this limiting value as the tunneling current j_{tun}. In fact, the low-temperature data in Fig. 2(a) can be fit with the same exponential law that determines the ground state decay rate in the one-dimensional Hubbard model [2,3]

$$j_{\mathrm{tun}}(F) = F\sigma_{\mathrm{tun}}^{\infty} \exp(-F_{\mathrm{th}}/F), \tag{2}$$

with a threshold F_{th} [solid line in Fig. 2(a)].

For small fields and $T > 0$, the exponentially small $j_{\mathrm{tun}}(F)$ is dominated by the linear response current $\sigma_{\mathrm{dc}}F$. Surprisingly, however, we find that thermal and tunneling current do not simply add up in the stationary state, but $\bar{\jmath}/F$ can become much smaller than σ_{dc}. The peculiar minimum at the crossover between tunneling and linear response regime in Fig. 2(a) arises because the relaxation to the stationary state becomes slower with decreasing field, such that an average of $j(t)$ in a fixed time interval still yields $\sigma_{\mathrm{dc}}F$ for $F \to 0$. A detailed analysis of the long-time behavior at intermediate F reveals that the stationary current is approached via an exponential decay $j(t) = \bar{\jmath} + \delta j \exp(-t/\tau)$, where $\bar{\jmath}$ is closer to $j_{\mathrm{tun}}(F)$ than it is to the linear response current [Fig. 2(b)]. The decay of the thermal current indicates that mobile, thermally excited carriers (and those excited during the switch on) are transformed into immobile excitations in the presence of an electric field, such that the linear response current is only visible on a time scale $\propto 1/F$. Clearly, such a behavior must be specific to a closed system, whereas in an open system the coupling to the environment constantly tends to restore the thermal state. Nevertheless, the observed relaxation phenomenon is an interesting topic for future theoretical transport investigations, and it is an open question whether it is a generic feature in closed systems.

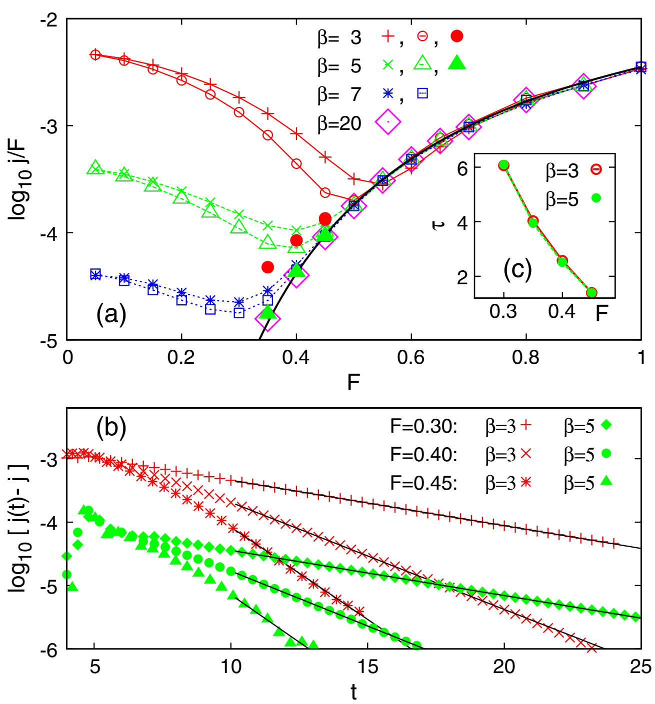

FIG. 2 (color online). (a) Time-averaged current, $\bar{\jmath}/F$, for $U = 5$. Crosslike and open symbols correspond to a time average for $8 < t < 10$ and $10 < t < 12$, respectively. Filled symbols result from an extrapolation to $t = \infty$ [see (b)]. The solid black line is obtained by fitting the data for $\beta = 20$ with Eq. (2) [$F_{\mathrm{th}} = 1.92$, $\sigma_{\mathrm{tun}}^{\infty} = 0.066$]. (b) Long-time evolution of the current for $U = 5$, analyzed by a nonlinear least-squares fit $j(t) = \bar{\jmath} + \delta j \exp(-t/\tau)$ (solid lines); the value $\bar{\jmath}$ is shown by the filled symbols in (a). (c) The relaxation time τ.

Figure 3 summarizes the main numerical results of this Letter by comparing the current in the tunneling regime ($T \downarrow 0$) and in the linear response regime ($F \to 0$, $t \lesssim 1/F$). For $U \lesssim 3$, the linear response conductivity σ_{dc} increases with decreasing temperature, while it becomes exponentially small for $U \gtrsim 3$,

$$\sigma_{\mathrm{dc}} \sim \exp(-\Delta/T), \tag{3}$$

thus signaling the metal-insulator transition at $U \approx 3$ [Figs. 3(a) and 3(b)]. (Strictly speaking, the metal-insulator transition displayed in Fig. 3 is only a crossover because temperatures are larger than T_c.) On the other hand, by fitting $\bar{\jmath}/F$ for $\beta = 20$ with Eq. (2), we obtain the threshold F_{th} as a function of the interaction [Figs. 3(c) and 3(d)]. Deep in the insulating phase, both Δ and F_{th} increase linearly with U, while they vanish as the metal-insulator transition is approached. Because the metal-insulator transition is first order in DMFT [9], observables in the insulating phase are expected to display nonanalytic behavior not at the actual transition U_{c2} at $T = 0$ but at the spinodal point U_{c1} where the insulator at $T = 0$ disappears. However, the precise behavior of Δ and F_{th} at the transition is difficult to obtain within the current approach: As F_{th} decreases, a fit of $\bar{\jmath}/F$ with Eq. (2) becomes increasingly difficult because the stationary state is eventually no longer reached within the numerically accessible times for $F < F_{\mathrm{th}}$ due to the slow saturation of $j(t)$ for small F [cf. Fig. 2(c)].

Apart from the crossover region at $U < 4$, however, we do observe the behavior described by Eqs. (2) and (3) over a wide range of temperatures and fields. Although our setup involves a closed system in which energy is conserved, we expect that these DMFT results resemble the I-V characteristics of a real Mott-insulating material for large and small fields, respectively. While the quasistationary state cannot exist forever in our setup, it can indeed be stable in a real solid provided that the excess energy is either passed to the lattice or flowing to the boundaries where it is absorbed by the leads. This would lead to a steady entropy increase of the environment, while the entropy of the system itself can only be evaluated after

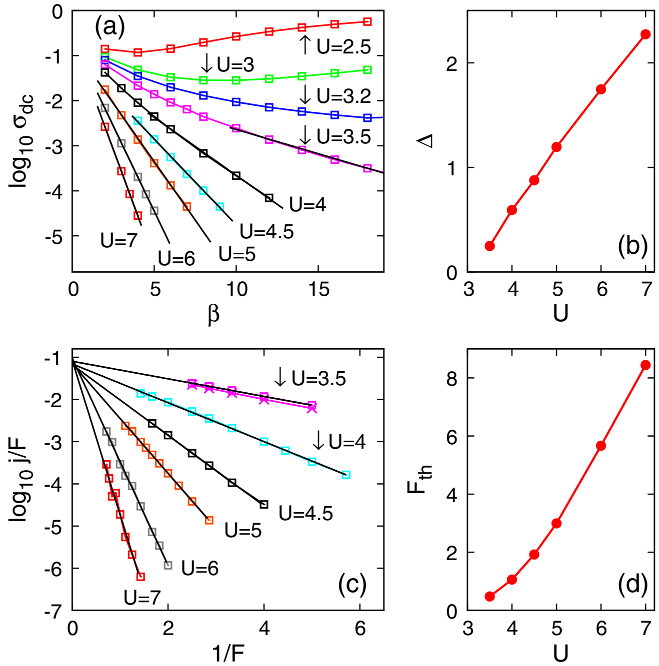

FIG. 3 (color online). (a) Linear response conductivity, obtained by extrapolating $\bar{j}/F$ to $F = 0$. Thin solid lines correspond to fits with Eq. (3); the resulting gap $\Delta(U)$ is shown in (b). (c) Conductance $\bar{j}/F$ for the stationary current at $\beta = 20$. For $U = 3.5$, there is a still a slight drift of the current at the largest times (see text), and we plot time averages for $8 < t < 10$ (crosses) and $12 < t < 14$ (open symbols). Solid lines are linear fits according to Eq. (2). (d) The threshold field $F_{\text{th}}(U)$ resulting from the fits in (c).

the field is switched off and the system is allowed to relax to a thermal equilibrium state. Since the tunneling current turns out to be essentially independent of the excitation of the system (cf. Fig. 1), differences between the quasistationary current in our setup and the stationary current in experiments are expected to be of the order of the linear response current (3), which is negligible compared to Eq. (2) for large enough fields. For small fields or high temperatures, on the other hand, the open system should recover the linear response behavior which is lost in the closed system at long times.

We find that the ratio F_{th}/Δ is only weakly dependent on the interaction or the bandwidth in the insulator, and it should thus give the correct order of magnitude for the breakdown field in real Mott insulators as well. Note that this value, i.e., $F_{\text{th}} \approx 2 - 3\Delta/ea$, is much larger than the temperature-dependent threshold which is obtained in the experiments of Ref. [1] in connection with a negative differential resistance (for $SrCuO_3$, e.g., $F_{\text{th}} \approx 10^{-4}\Delta/ea$ at $T = 190$ K). The threshold behavior in these one-dimensional materials must thus be of a different origin, and indeed collective excitations were proposed in Ref. [1], as the temperature dependence of the experimental threshold is similar to what is expected for charge-ordered materials. The larger threshold found in our analysis, which may be achieved in experiments on thin layers of insulating material between metallic leads, should be observed in paramagnetic Mott insulators when other sources of destabilizing the insulator are not present.

In conclusion, we have investigated the dielectric breakdown of a Mott insulator in the Hubbard model by computing the current in a strong electric field F. Our main result is the formation of a quasistationary nonequilibrium state with time-independent current, which may be called a field-induced metal. In the limit of small temperature, the stationary current resembles the exponential law [Eq. (2)] for the ground state decay rate in a one-dimensional Hubbard model due to many-body Landau-Zener tunneling [2,3]. Its value becomes exponentially small below a threshold field which vanishes at the metal-insulator transition.

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