Fate of Spinons at the Mott Point

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Gapless spin liquids have recently been observed in several frustrated Mott insulators, with elementary spin excitations - "spinons" - reminiscent of degenerate Fermi systems. However, their precise role at the Mott point, where charge fluctuations begin to proliferate, remains controversial and ill-understood. Here we present the simplest theoretical framework that treats the dynamics of emergent spin and charge excitations on the same footing, providing a new physical picture of the Mott metal-insulator transition at half filing. We identify a generic orthogonality mechanism leading to strong damping of spinons, arising as soon as the Mott gap closes. Our results indicates that spinons should not play a significant role within the high-temperature quantum critical regime above the Mott point - in striking agreement with all available experiments.

Introduction. The physical nature of the Mott metal to insulator transition (MIT), a phenomenon generic to strongly correlated materials, still remains the subject of much controversy and debate. In contrast to conventional critical phenomena, the relevant degrees of freedom at the MIT cannot be easily identified using an appropriate order parameter of symmetry breaking principle, although different competing orders often do arise in its vicinity. As stressed in pioneering works by Mott and Anderson, however, a fundamentally different physical mechanism has to exist, because the Mott insulating state typically persists to temperatures much higher than any conventional order.

A clear physical picture of how a sharp Mott transition can exist without any intervening order has emerged only recently, with the development of Dynamically Mean-Field Theory (DMFT) [1], which is formally exact in the limit of large coordination. Physically, it represents the limit of maximal frustration, therefore eliminating the precursors of any competing order, and retaining only purely local dynamical scattering processes. In contrast to alternative theoretical approaches describing dilute low-energy excitations, DMFT is most reliable at intermediate and high temperatures, where incoherent behavior prevails. The DMFT picture resulted in a finite temperature first-order boundary between the two phases, around which many fascinating phenomena organize themselves. Several of its most striking predictions were experimentally confirmed in a variety of systems, including organic charge-transfer salts of the κ-family, as well as various transition metal oxides. Here one can list the observation of strongly renormalized Fermi liquids [2], bad metal behavior, Ising universality near the Mott endpoint [3, 4], and even quantum critical scaling at higher temperatures [5, 6].

At lower temperatures, most Mott insulators still undergo magnetic ordering, but experimental studies of several frustrated organic materials have instead observed spin liquid behavior [7–11]. Two particular compounds have attracted a lot of attention, κ-(BEDT-
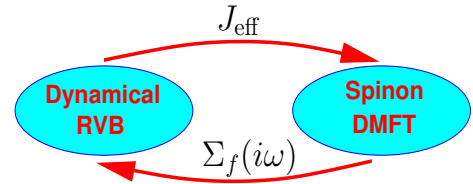

FIG. 1. (Color online) Proposed scheme to blend the static RVB approach with the DMFT, using an impurity solver based on emergent spinon degrees of freedom. The spinon self-energy $\Sigma_f(i\omega)$, usually absent from a mean-field RVB treatment, is extracted from a spinon-based DMFT framework. Using the RVB self-consistent determination of the spinon bandwidth J_{eff}, this in turn allows to incorporate feedback effects due to scattering on charge excitations, which affect the gapless spin-excitations in the Mott state.

TTF)$_2$Cu$_2$(CN)$_3$ and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$. Most remarkably, thermodynamic measurement in the Mott insulating phase of these materials have revealed behavior normally expected for metals, including displaying linear in temperature specific heat and large thermal conductivity, indicating the presence of gapless magnetic excitations.

These observations are easiest to rationalize using a time-honored idea, the resonating valence bond (RVB) theory [12], in which chargeless spin excitations embody a magnetic fluid with fermionic statistics. Despite this appealing picture, the RVB approach focuses on the zero temperature phases, and is at trouble in recovering the expected DMFT predictions upon warming up the system. One question that immediately arises concerns the compatibility of the two electronic fluids, namely the Fermi liquid describing the correlated metallic state, and the magnetic fluid characterizing such a gapless Mott insulator. In this Letter, we address this question by proposing a consistent framework that succeeds in marrying the DMFT and the RVB approaches. This allows to preserve the known high temperature phenomenology of the DMFT, while introducing the strong thermody-

namic signature of a gapless spin liquid state in the Mott phase. Akin to the impossibility of mixing oil and vinegar, we find that the Fermi liquid and the magnetic fluid are not miscible into each other in the transition region. While the insulating spin-liquid obviously cannot support mobile charge carriers, the Fermi liquid metal always shows strongly incoherent spinon excitations, due to an orthogonality catastrophe [13–15]. This observation has important consequences, because the two associated Fermi surfaces cannot be tuned into each other in a continuous way. Thus, the Mott localization into a gapless spin-liquid turns out to have first-order character even at zero temperature, similarly to the case of a spin-gapped insulator described by frozen short-range singlets. This result is in contrast with other scenarios based on the RVB picture only, in which the quasiparticles vanish continuously [16–19], but is consistent with all available experiments.

We start by describing our theoretical framework, that is sketched in Fig. 1. The main idea is to solve the DMFT equations by explicitly introducing spinon degrees of freedom, and to feedback the resulting spinon self-energy $\Sigma_f(\omega)$ into the RVB equations, thus affecting the stability of the spin liquid. To be concrete, we consider henceforth the half-filled Hubbard-Heisenberg Hamiltonian:

$$H = -t \sum_{\langle i,j \rangle \sigma} d_{i\sigma}^\dagger d_{j\sigma} + U \sum_i \left[d_{i\uparrow}^\dagger d_{i\uparrow} - \frac{1}{2} \right] \left[d_{i\downarrow}^\dagger d_{i\downarrow} - \frac{1}{2} \right]$$
$$+ J \sum_{\langle i,j \rangle} \left[\sum_{\alpha,\alpha'} d_{i\alpha}^\dagger \frac{\vec{\tau}_{\alpha,\alpha'}}{2} d_{i\alpha'} \right] \cdot \left[\sum_{\sigma,\sigma'} d_{j\sigma}^\dagger \frac{\vec{\tau}_{\sigma,\sigma'}}{2} d_{j\sigma'} \right]. \quad (1)$$

Here t is the intersite hopping, U the local Coulomb interaction, J an explicit nearest neighbor exchange, and we have denoted by $\vec{\tau}$ the set of Pauli matrices. The RVB picture results here from a decomposition of the physical electron into a chargeless spin-carrying fermion $f_{j,\sigma}^\dagger$ and a spinless charge-carrying compact boson $X_j = e^{i\theta_j}$, using on each site j the so-called slave rotor [16, 20] decomposition $d_{j,\sigma}^\dagger = f_{j,\sigma}^\dagger e^{i\theta_j}$, with $i\partial/\partial\theta_j = \sum_\sigma [f_{j\sigma}^\dagger f_{j\sigma} - \frac{1}{2}]$. This provides an effective Hamiltonian:

$$H_{\text{eff}} = \sum_{\langle i,j \rangle \sigma} f_{i\sigma}^\dagger f_{j\sigma} \left[J_{\text{eff}} - t e^{i(\theta_i - \theta_j)} \right] - \frac{U}{2} \sum_i \frac{\partial^2}{\partial \theta_i^2}, (2)$$

with $J_{\text{eff}} = -J \langle f_{i\sigma}^\dagger f_{j\sigma} \rangle$ the RVB bond parameter. This approximation of the Heisenberg term assumes the formation of a spinon Fermi surface, yet neglects a possible momentum-dependent spinon self-energy. Such non-local effects, associated to fluctuations beyond the RVB mean-field, are not expected to change the physics of the spin-liquid Mott insulator, owing to the strong stability of such zero-entropy charge-gapped state. The mean-field Hamiltonian (2) is consecutively solved within the DMFT, by employing an impurity solver that is naturally based on the spinon/rotor decomposition [20], leading to the following respective local Green's functions in Matsubara

domain:

$$G_f(i\omega_n)^{-1} = i\omega_n + \mu - \Sigma_f(i\omega_n) - \Delta_f(i\omega_n), \quad (3)$$
$$G_X(i\nu_n)^{-1} = \frac{\nu_n^2}{U} + \lambda - \Sigma_X(i\nu_n), \quad (4)$$

and self-energies in imaginary time:

$$\Sigma_f(\tau) = \Delta(\tau) G_X(\tau), \quad (5)$$
$$\Sigma_X(\tau) = \mathcal{N} \Delta(\tau) G_f(\tau), \quad (6)$$

with $\mathcal{N} = 3$ to ensure the right shape of the Mott phase diagram, and λ a Lagrange multiplier that enforces the constraint $|X_j|^2 = 1$ in average, namely $G_X(\tau = 0) = 1$. The DMFT self-consistency equations account both for the physical electron and the spinon hybridization functions, which read for the Bethe lattice: $\Delta(\tau) = t^2 G_d(\tau) = t^2 G_f(\tau) G_X(\tau)$ and $\Delta_f(\tau) = J_{\text{eff}}^2 G_f(\tau)$. The generic RVB equation, $J_{\text{eff}} = -J G_f(\langle i,j \rangle, \tau = 0)$ can now be explicited on the Bethe lattice:

$$J_{\text{eff}} = \frac{J}{2\beta} \sum_{i\omega_n} \frac{J_{\text{eff}}}{\frac{z_n^2}{2} + z_n \sqrt{\frac{z_n^2}{4} + J_{\text{eff}}^2} + J_{\text{eff}}^2}, \quad (7)$$

with $z_n = \omega_n - \text{Im}\Sigma_f(i\omega_n)$ and $\beta = 1/T$ the inverse temperature. We will set in what follows $t = 1/2$, taking the electronic half-bandwith $D = 2t = 1$ as natural energy unit. The computation of the electronic specific heat results from the internal energy per site (with N_s sites):

$$\frac{\langle H \rangle}{N_s} = \frac{2}{\beta N_s} \sum_{n,k} [\epsilon_k^d G_d(k, i\omega_n) + \epsilon_k^f G_f(k, i\omega_n)] e^{i\omega_n 0^+}$$
$$+ \frac{U}{2} D_{\uparrow\downarrow} + \frac{1}{N_s} \sum_{\langle i,j \rangle \sigma} \frac{J_{\text{eff}}^2}{J}, \quad (8)$$

with $\epsilon_k^{d/f}$ the electron and spinon dispersion relations, and $G_{d/f}(k, i\omega_n)$ their respective lattice Green's functions. For the Bethe lattice, the sum over momentum in Eq. (8) can be replaced by an energy integral over the corresponding semi-circular density of states [21]. An important term to consider here is the double occupancy $D_{\uparrow\downarrow}$, which is related to the local charge susceptibility by $D_{\uparrow\downarrow} = (1/2)\chi_c(\tau = 0)$. This quantity can be expressed from either a spinon response χ_c^f or a rotor response χ_c^X:

$$\chi_c^f(\tau) = \langle \sum_{\sigma,\sigma'} [f_{j\sigma}^\dagger(\tau) f_{j\sigma}(\tau) - \frac{1}{2}][f_{j\sigma'}^\dagger(0) f_{j\sigma'}(0) - \frac{1}{2}] \rangle,$$
$$\chi_c^X(\tau) = \langle i \frac{\partial}{\partial \theta_j}(\tau) i \frac{\partial}{\partial \theta_j}(0) \rangle. \quad (9)$$

Both expressions are equivalent only provided the constraint is dealt strictly, but in a mean-field treatment, one must use Nagaosa and Lee's composition rule [12, 21], $\chi_c(i\omega) = [(\chi_c^f)^{-1} + (\chi_c^X)^{-1}]^{-1}$.

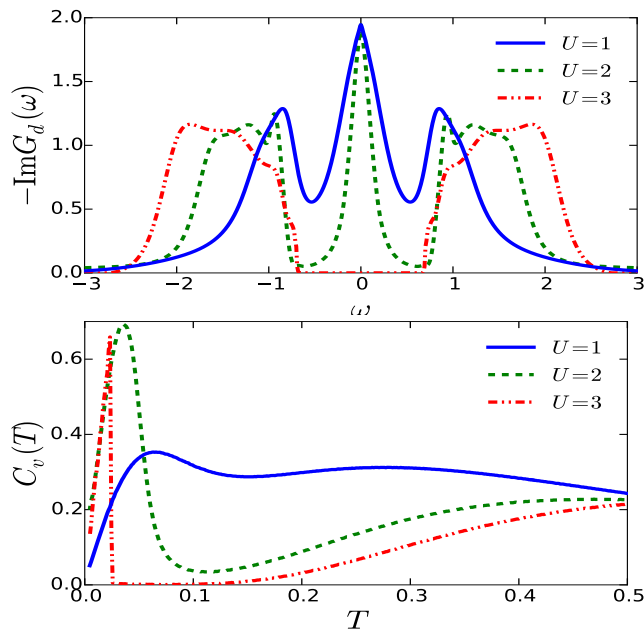The solution of the combined RVB and DMFT self-consistent scheme provides the physical density of states

FIG. 2. (Color online) Upper panel: electronic density of states across the Mott transition for $T = 0.005D$ and $J/D = 0.2$, with increasing values of the Coulomb interaction $U/D = 1, 2, 3$. Lower panel: corresponding specific heat as a function of temperature. A striking linear in temperature contribution remains in C_V at $U/D = 3$, while quasiparticles have disappeared from the density of states in the Mott phase.

and specific heat curves shown in Fig. 2. The electronic density of states shows at low temperature the expected behavior: the quasiparticle peak narrows down for increasing values of U, with strong spectral weight transfer towards Hubbard bands located at $\pm U/2$. This situation persists upon a discontinuous disappearance of the quasiparticle peak, leading to the formation of an insulating state with a large Mott gap (shown for $U = 3$). The formation of heavy quasiparticles can equally be witnessed in the specific heat (bottom panel in Fig. 2), with a strong enhancement of the $\gamma = C_V/T$ coefficient at the lowest temperature. However, in strong contrast with the usual DMFT predictions [1, 21], we find the persistence of a finite γ coefficient in the Mott phase, instead of the usually observed activated behavior for a high entropy paramagnetic insulator. This result is in agreement with the thermodynamic measurements made in several organic materials showing spin liquid behavior. Fitting from Fig. 2 the low-temperature linear slope of C_V in the Mott insulating phase, we find $\gamma = 27k_B^2/D$ and $\gamma = 14k_B^2/D$ for $J/D = 0.2$ and $J/D = 0.4$ respectively. Taking the half-bandwidth in the range $D = 200$ meV, from recent estimates on various organic systems [22, 23], we have approximately $\gamma = 80$ mJK^{-2}mol^{-1} and $\gamma = 40$ mJK^{-2}mol^{-1} for $J/D = 0.2$ and $J/D = 0.4$ (these correspond to typical values of the exchange constant in organics [24]). Our predictions are somewhat larger, but of the right magni-

tude, with the experimental value $\gamma = 20$ mJK^{-2}mol^{-1} measured both for the EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ [10] and κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ compounds [9].

We now demonstrate that this thermodynamic effect has a strong influence of the metal-insulator phase diagram. The main reason is the quenching of the entropy of the Mott insulator by the presence of a finite exchange interaction J, which typically bends the transition lines towards a stabilization of the metal upon heating. Indeed, the entropic contribution to free energy of the insulator is strongly diminished by exchange, leading to a reentrance of the first order transition lines [25–27]. We show in
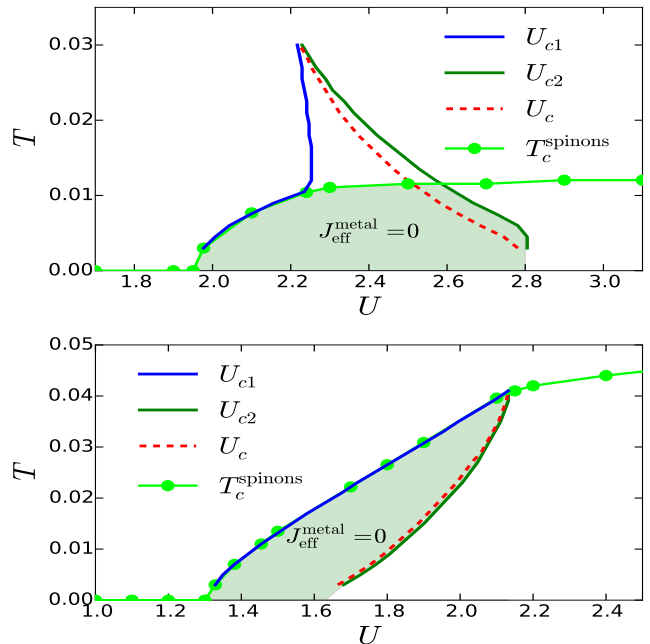

FIG. 3. (Color online) Phase diagram for $J/D = 0.1$ (upper panel) and $J/D = 0.4$ (lower panel) as a function of Coulomb strength U/D and temperature T/D. Continuous lines denote the metal-insulator boundaries, and dashed lines indicate the true first-order transition based on the free energy. Also, the region bounded by dots show the low-temperature onset of the spinon Fermi surface in the Mott insulator. The shaded region indicates that $J_{\mathrm{eff}}^{\mathrm{metal}} = 0$ for the metallic state within the whole coexistence region. This demonstrates that spinons are only well defined quasiparticles in the low temperature Mott phase, and are never stable in the metal.

Fig. 3 two phase diagrams, for $J/D = 0.1$ and $J/D = 0.4$ respectively. The continuous lines denote the metal to insulator boundaries, $U_{c1}(T)$ and $U_{c2}(T)$ at which the insulator and metallic solution disappear respectively. For small J (upper panel), only the U_{c1} line is bent, while the whole transition region is affected for large J (lower panel), with a slight increase of the maximum critical temperature T_c due to the coupling to spinon degrees of freedom. Most importantly, the critical domain moves towards strongly reduced values of the Coulomb strength U at increasing J, so that the Brinkman-Rice transition,

associated to a diverging γ coefficient, is strongly pre-empted by spin fluctuations. This readily explains the small values of γ that were found in our calculations.

We then consider the fate of the gapless insulating spin liquid. The phase diagrams of Fig. 3 also show as dots the onset of the spinon Fermi surface, namely the low-temperature domain where the effective spinon bandwidth $J_{\text{eff}} \neq 0$. We find that this domain corresponds precisely to the region of existence of the Mott insulator (only at low temperature for small J, and at temperatures up to the critical T_c of the terminal Mott endpoint for large enough J). Said otherwise, the high entropy local moment insulator is always unstable to the formation of a low entropy spin liquid, even in the coexistence region of the MIT where the Mott gap is strongly reduced. Since the gapless spin-liquid state penetrates fully the part of the phase diagram where metal and insulator coexist, one can wonder whether the metallic state is capable of hosting non-trivial spinon excitation. We find however, for all our simulations, that the spinon bandwidth always vanishes in the metal, namely $J_{\text{eff}}^{\text{metal}} = 0$. Since the critical value $U_{c2}(T = 0)$ for the loss of the metal steadily decreases with increasing J, this shows that the Mott transition becomes intrinsically first order at zero temperature, in contrast to the case $J = 0$ (standard DMFT), where the quasiparticle weight continuously vanishes.

We finally show that spinons are dramatically repelled from the Fermi liquid because of the generic occurence of an orthogonality catastrophe in the spinon self-energy $\Sigma_f(\omega)$ for a Fermi liquid state. The reason is deeply rooted in the spinon/rotor decomposition, and its associated constraint $Q = i\partial/\partial\theta_j - \sum_\sigma [f_{j\sigma}^\dagger f_{j\sigma} - \frac{1}{2}] = 0$. While the spectral function of the physical electron $d_{j\sigma}^\dagger = f_{j\sigma}^\dagger e^{i\theta_j}$ connects excitations within the physical subspace $Q = 0$ only, the spinon density of states corresponds to processes that link the physical subspace $Q = 0$ to an unphysical subspace $Q = 1$ containing one extra auxiliary slave-particle. Thus, the spinon density of states involves matrix elements of the type $|\langle \Phi^{(1)} | f_{j\sigma}^\dagger | \Phi^{(0)} \rangle|^2$, where $|\Phi^{(Q)}\rangle$ are wavefunctions living in different Q-subspace, which thus experience in a metal a singular x-ray edge. Due to this mechanism, the spinon self-energy acquires an anomalous frequency dependence $\Sigma_f(\omega) \propto \omega^\alpha$ at low-energy (typically $\alpha < 1/2$). This crucial property is well obeyed by construction in our impurity solver [20], and can be shown from an exact numerical solution of quantum impurity problems [13, 14], as well as from $1/N$ fluctuations [28] around the condensed slave boson mean-field theory (upon which the static RVB picture is based). The calculated spinon self-energy is shown in Fig. 4 for the metallic and insulating phases. The anomalously large spinon scattering rate is clearly seen for $U/D = 1$, while a regular behavior is found for $U/D = 3$. Spinon are thus very incoherent in the metallic state, insomuch as to fully destroys their Fermi surface, as we discuss now.
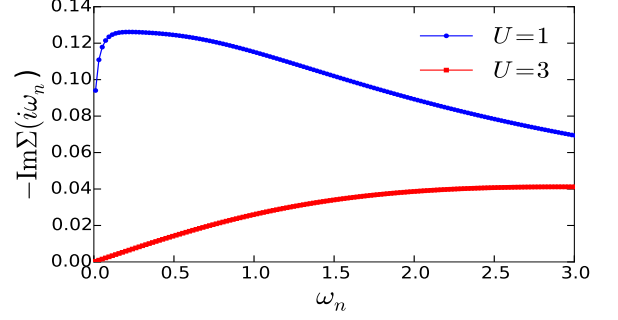

FIG. 4. (Color online) Frequency-dependent spinon self-energy $\Sigma_f(i\omega_n)$ at temperature $T = 0.005D$ for $U/D = 1$ (metal) and $U/D = 3$ (insulator). The strong enhancement in the metal is due to an x-ray edge effect, which is a hallmark the spinon spectral density.

The spinon orthogonality catastrophe is indeed the key physical mechanism leading to the instability of the spinon Fermi surface in the whole metallic phase. The absence of spinons in the metal, $J_{\text{eff}}^{\text{metal}} = 0$, can be formulated as a simple inequality from the linearized version of the RVB equation (7):

$$\frac{1}{J} > \frac{1}{2\beta} \sum_{i\omega_n} \frac{1}{[\omega_n - \text{Im}\Sigma_f(i\omega_n)]^2}. \tag{10}$$

Using the anomalous spinon self-energy $\Sigma_f(i\omega) = -iC_f|\omega|^\alpha \text{Sign}(\omega)$ at low energy, and evaluating the Matsubara sum at low temperatures, we find the inequality for stability of the a true Fermi liquid:

$$\frac{1}{J} > \int_0^\infty \frac{d\omega}{\pi} \frac{1}{[\omega + C_f\omega^\alpha]^2} \tag{11}$$

Since $\alpha < 1/2$, the infrared divergence in the integral is cut off and the inequality is fulfilled for small J. In contrast, the Mott insulator is always unstable to the spin-liquid because the integral diverges for $\Sigma_f = 0$. These analytic arguments are in complete agreement with our numerical findings.

In conclusion, we have examined the role of gapless spin excitations, characterizing frustrated Mott insulators, in the vicinity of the Mott metal-insulator transition at half-filling. Our result indicate that the spin-liquid arises simultaneously with Mott gap opening, suggesting that local magnetic moments generically tend to form a zero-entropy gapless state in absence of magnetic ordering. However, a spinon Fermi surface cannot be stabilized upon closing of the Mott gap, due to an orthogonality catastrophe we identified with emerging charge fluctuations. This mechanism leads to the conclusion that the Mott transition associated with the loss of quasiparticles at U_{c2} is inherently of first-order type, even at zero temperature and for a zero entropy Mott insulator. From this perspective, we advocate a scenario where behavior

consistent with quantum criticality (QC) has purely local character and emerges only at high temperatures, above the metal-insulator coexistence region, consistent with experiments. Our result indicate, thus, that spin liquid correlations do not play a significant role within this high temperature QC region, which marks the closing of the Mott gap. The physical picture we propose is dramatically different from the perspective provided from alternative theories [16–19], which postulate the dominance of spin-liquid excitation in the entire QC regime surrounding the Mott point.

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