Universality of Liquid-Gas Mott Transitions at Finite Temperatures

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We explain, in a consistent manner, the set of seemingly conflicting experiments on the finite temperature Mott critical point, and demonstrate that the Mott transition is in the Ising universality class. We show that, even though the thermodynamic behavior of the system near such critical point is described by an Ising order parameter, the global conductivity can depend on other singular observables and, in particular, on the energy density. Finally, we show that in the presence of weak disorder the dimensionality of the system has crucial effects on the size of the critical region that is probed experimentally.

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Although band theory predicts that a system of electrons in a solid with one electron per site (unit cell) should be metallic, such a system ultimately insulates [1,2] once the local electron repulsive interactions exceeds a critical value. The onset of the insulating state, the Mott transition, arises from the relative energy cost of the on-site Coulomb repulsion U between two electrons on the same lattice site, and the kinetic energy, represented by the band width W. Then, the transition is governed solely by the ratio of U/W. At $T = 0$, it is often the case that symmetries of the microscopic system, associated with charge, orbital or spin order, may be broken in the Mott insulating state. However, at sufficiently high temperatures T, or in strongly frustrated systems, no symmetry is broken at the finite-T Mott transition. Then, the transition is characterized by paramagnetic insulating and metallic phases, whose coexistence terminates at a second-order critical point, depicted in Fig. 1(a). In this Letter, we are concerned with the universal properties of this *classical critical point* [3], as revealed by a series of apparently conflicting experiments on $(Cr_{1-x}V_x)_2O_3$ [4] and organic salts of the κ-ET [bis(ethylenedithio) tetrathiafulvalene] family [5].

Since no symmetry is broken at the finite-T Mott transition, in a strict sense there is no order parameter. Nonetheless, experimental [4,5], as well as theoretical evidence [6,7] suggests that the transition is in the Ising universality class, similar to the liquid-vapor transition. For example, Castellani *et al.* [6] constructed an effective Hamiltonian for this problem, and proposed that double occupancy plays the role of an order parameter for the Mott transition. On the insulating side, doubly occupied sites are effectively localized, but in the metal, they proliferate. A Landau-Ginzburg analysis [7] provided further evidence for a nonanalyticity in the double occupancy at a critical value of U/W that defines a Mott transition. Ising universality follows immediately because double occupancy, $\langle n_{i\uparrow} n_{i\downarrow} \rangle$, is a scalar local density field.

Experimentally, the universality of the Mott critical point is typically probed by some external parameter, such as pressure, which can tune the ratio W/U. Measurements of the conductivity, Σ, on $(Cr_{1-x}V_x)_2O_3$ [4] found that away from the critical point, the exponents defined through

$$\Delta\Sigma(t, h = 0) = \Sigma(t, h = 0) - \Sigma_c \propto |t|^{\beta_\sigma},$$
$$\Delta\Sigma(t = 0, h) \propto |h|^{1/\delta_\sigma}, \qquad \partial\Sigma(t, h)/\partial h|_{h=0} \propto |t|^{-\gamma_\sigma}, \tag{1}$$

have mean-field Ising values, $\beta_\sigma \simeq 1/2$, $\gamma_\sigma \simeq 1$, and $\delta_\sigma \simeq 3$. Here, $t = (T - T_c)/T_c$ and $h = (P - P_c)/P_c$, with (Σ_c, T_c, P_c) denoting the corresponding values at the critical endpoint. Close to the critical region, Limelette *et al.* [4] observed a drift to the critical exponents of the

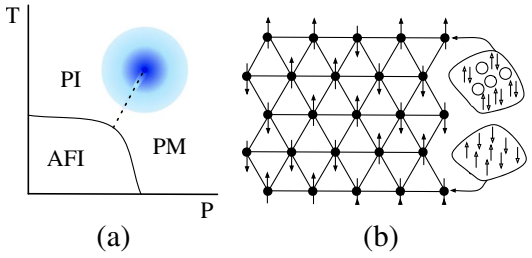

FIG. 1 (color online). (a) Typical phase diagram of Mott transitions as a function of pressure P and temperature T. At low T and P, a Néel antiferromagnetic insulator (AFI) appears; it becomes a paramagnetic insulator (PI) if T increases, or a paramagnetic metal (PM) if P increases. Dashed line: first-order transition ending at a liquid-gas critical point. Full lines: continuous phase transition to the ordered state. Colored regions: critical (dark) and mean-field (light) regimes of the critical point. (b) An Ising configuration on the triangular lattice. Up (down) spins correspond to conducting (insulating) grains of linear size of the order of the system's dephasing length.

3D Ising universality class. Mean-field behavior is also seen in NiS_2 [8].

However, similar pressure measurements [5] on the quasi-2D organic salts of the κ-ET family appear to challenge the view that the Mott transition is in the Ising universality class. In this material, Kagawa *et al.* [5] found that their data is described by the exponents $\beta_\sigma \simeq 1$, $\gamma_\sigma \simeq 1$, and $\delta_\sigma \simeq 2$, which do not seem to be consistent with the known exponents of the 2D Ising model whose exponents are [9] $\beta = \frac{1}{8}$, $\gamma = \frac{7}{4}$, and $\delta = 15$. Since the exponents obey the scaling law $\gamma_\sigma = \beta_\sigma(\delta_\sigma - 1)$, it was proposed that the Mott transition is in a new, as yet unknown universality class. The situation is further complicated by thermal expansion measurements [10] that claim to measure the heat capacity exponent α and find $0.8 < \alpha < 0.95$. This result is not only in sharp contrast to the expectation for an Ising transition (where $\alpha = 0$ for $d = 2$), it also strongly violates the scaling law $\alpha + 2\beta_\sigma + \gamma_\sigma = 2$, if one uses the exponents of Ref. [5].

In this Letter, we present a unified phenomenological description of all of these experimental facts within an Ising-type model, and resolve the issue of the universality class of the Mott transition. A complete description of these experiments requires to take into account that the conductivity depends on all possible singular observables of the associated critical system, and not just on the thermodynamic order parameter associated with the phase transition. Similar considerations were made in magnetic systems near the Curie temperature [11–13], to explain the critical exponent of the conductivity along the coexistence curve. In that case, a symmetry of the microscopic definition of the conductance prevented any coupling of the global conductivity to odd moments of the order parameter, along the coexistence line. Even though similar in spirit, the situation here is much different. Starting from an effective microscopic model near an Ising critical point, we show that: (i) the conductivity typically depends on all possible singular thermodynamic observables of the system, namely, the order parameter and energy density of the Ising model; (ii) when the coupling to the energy density dominates, there exists a large regime around the critical point, where the critical exponents for the conductivity are $(\beta_\sigma, \gamma_\sigma, \delta_\sigma) = (1, \frac{7}{8}, \frac{15}{8})$, that agree (within the error bars) with the findings of Kagawa *et al.* [5], and the corresponding mean-field exponents are $(\beta_\sigma^{MF}, \gamma_\sigma^{MF}, \delta_\sigma^{MF}) = (1, \frac{1}{2}, \frac{3}{2})$; (iii) a crossover to Ising exponents is obtained in the order parameter dominated regime as seen in Refs. [4,8]; (iv) in the presence of disorder the Mott critical point ultimately belongs to the random-field Ising model universality class, and therefore the dimensionality of the system under study is even more important for specifying its critical properties.

In order to resolve the discrepancies raised by these experiments, we consider the behavior of the conductivity of the system near the Mott critical point, assuming that it belongs to the 2D Ising universality class. Rather than starting from a microscopic picture, e.g., a Hubbard model, we consider a coarse-grained model with the correct symmetries in which the physics of the relevant transport degrees of freedom is captured. In this picture, one defines coarse-grained regions, of linear size of the order of the dephasing length l_ϕ of the system, which are either insulating or conducting. Along these lines, we consider an Ising model on a 2D lattice [cf. Fig. 1(b)]. Near the critical point, where the correlation length for density fluctuations ξ diverges, it is expected that the relevant degrees of freedom behave classically. The Ising variables S_i on each lattice site represent the fluctuating density of mobile carriers on microscopic "grains" of linear size of the order of the dephasing length l_ϕ, which are conducting ($S_i = +1$), or insulating ($S_i = -1$). The Hamiltonian is

$$\beta H = -\frac{1}{T}\sum_{\langle ij\rangle} S_i S_j + \frac{h}{T}\sum_i S_i, \tag{2}$$

where T is the temperature, P and P_c are the pressure and the critical pressure, respectively, and $h \propto P - P_c$ plays the role of the Ising magnetic field. This model is expected to describe the physics near the critical point, where $\xi \gg l_\phi$. In this limit, all other interactions beyond nearest-neighbor are irrelevant. Near the critical point, the most singular effect of the pressure is described by a coupling to the order parameter.

To relate the order parameter fluctuations to the transport properties we will define an associated resistor network for this model, an approach that has been successfully used in other strongly correlated systems [14,15]. Let σ_C and σ_I be the local conductivities of the conducting and insulating regions, respectively. We define the bond conductance of the network model simply by adding these two conductivities in series. The bond conductance has three possible values, depending on the state of each grain, which can both be conducting, both insulating, one conducting and the other insulating. Thus, the conductance of the bond (i, j) has the form

$$\sigma_{ij} = \sigma_0[1 + g_m(S_i + S_j) + g_\epsilon S_i S_j]. \tag{3}$$

Even in this toy model, the microscopic conductivity, σ_{ij}, couples both to the order parameter, S_i, and to the energy density, $S_i S_j$, of the Ising model with naturally large couplings, g_m and g_ϵ, defined in Eq. (3). More specifically, we find that $\sigma_0 = \frac{1}{4}(\sigma_C + \sigma_I) + \frac{\sigma_C\sigma_I}{\sigma_C+\sigma_I}$, $g_m = \frac{\sigma_C-\sigma_I}{4\sigma_0}$, and $g_\epsilon = \frac{(\sigma_C-\sigma_I)^2}{4\sigma_0(\sigma_C+\sigma_I)}$. At high contrast, $\sigma_C \gg \sigma_I$, we get $g_m \simeq g_\epsilon \simeq 1$, whereas, at low contrast, $|\sigma_I - \sigma_C| \ll \sigma_C$, we get $g_\epsilon < g_m \to 0$.

The conductivity of the 2D Ising model we described is a nontrivial quantity to compute. As it was shown in the simpler case of the random resistor network (RRN) [16], networks of bonds with conductance σ_C (σ_I) chosen *randomly* with probability p and $1 - p$, the global conductivity becomes nonzero as soon as an infinite percolating and conducting cluster emerges in the system. When $\sigma_I = 0$, the critical exponent β_σ of the conductivity is nontrivially

related to the fractal properties of the incipient infinite conducting cluster. This exponent is larger than unity for random uncorrelated networks and larger than the exponent of the order parameter, because dangling bonds of the infinite cluster do not contribute to the conductivity. On the other hand, it becomes much less than unity for correlated networks, and typically very close to the exponent of the order parameter, since the infinite cluster is connected with few dangling bonds.

On the other hand, when $\sigma_I > 0$, a conducting cluster is less distinguishable from that of an insulating one, and the complex effects coming from the fractal cluster boundaries are smeared out. In the context of RRN, the percolation transition is not seen in the behavior of the conductivity, which seems to show just a crossover. If the contrast is low, $\sigma_I \simeq \sigma_C$, the actual conductivity of a single bond between sites i, j, Σ, should depend only on local observables, and we can formally expand it in powers of g_m and g_ϵ [17],

$$\Sigma = \sigma_0 + g_m\langle(S_i + S_j)\rangle + g_\epsilon\langle S_i S_j\rangle + \ldots, \qquad (4)$$

where the ellipsis represents more complex products of local spin operators (weighed by rapidly decaying functions) [17]. Near the Ising critical point, the most singular contribution of the expectation values of multispin operators in Eq. (4) is given by the expectation value of the most singular, ''primary'', operators of the Ising critical point, the order parameter m and the energy density ϵ. Thus, the most singular term of multispin operators with odd (even) number of spins is proportional to the order parameter (energy density). Therefore, within the range of convergence of this expansion,

$$\Sigma = \Sigma_0(g_m, g_\epsilon) + f_m(g_m, g_\epsilon)\langle m\rangle + f_\epsilon(g_m, g_\epsilon)\langle\epsilon\rangle, \qquad (5)$$

where Σ_0, f_m, f_ϵ are nonuniversal regular polynomials in g_m and g_ϵ. Provided that the critical behavior is still controlled by the fixed point theory of the Ising model, the total conductivity should have the structure of Eq. (5). Thus, at finite contrast, Eq. (5) predicts that the actual conductivity is the sum of even and odd components, under the Ising symmetry transformation, $\Sigma = \Sigma_0 + \Sigma^{\text{even}} + \Sigma^{\text{odd}}$, and it should exhibit a *crossover* from an *energy density dominated* behavior at short distances to an *order parameter dominated* behavior at long distances. The crossover scale is controlled by the relative size of the functions f_m and f_ϵ (cf. Fig. 2). This behavior breaks down at high contrast where there is multifractal behavior (cf. inset in Fig. 2 and Ref. [18]).

We can understand the experiments of Refs. [4,5,8], if we assume that Eq. (5) applies. The results of Refs. [4,8] follow by assuming that $f_m(g_m, g_\epsilon) > f_\epsilon(g_m, g_\epsilon)$, and the conductivity scales as the order parameter. Conversely, the results of Ref. [5] follow if $f_\epsilon(g_m, g_\epsilon) \gg f_m(g_m, g_\epsilon)$, and the conductivity, for an extended regime near the critical point, scales as the energy density of the Ising model. In this case $\Delta\Sigma \propto |m|^\theta$ holds, where $\theta = (1 - \alpha)/\beta$. Then, it follows that $\beta_\sigma = \theta\beta$, $\delta_\sigma = \delta/\theta$ and $\gamma_\sigma = \gamma + \beta(1 - \theta)$. The resulting critical exponents are $(\beta_\sigma, \gamma_\sigma, \delta_\sigma) = (1, \frac{7}{8}, \frac{15}{8})$, very close to the experimental values. These exponents obey $\gamma_\sigma = \beta_\sigma(\delta_\sigma - 1)$, if $\gamma = \beta(\delta - 1)$, i.e., the conductivity exponents obey a scaling relation identical to that for the Ising exponents, in agreement with the experimental verification of this scaling in Ref. [5]. In addition, the scaling obtained by Kagawa *et al.* [5] only depends on $\beta\delta = \beta_\sigma\delta_\sigma$, as in our theory.

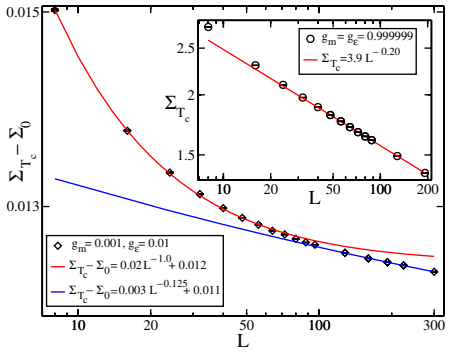

FIG. 2 (color online). Crossover behavior of the conductivity at finite contrast: the energy density (order parameter) dominates at short (long) length scales. Inset: Fractal scaling at large contrast.

In order to verify the theory presented above, we performed Monte Carlo simulations of the 2D Ising model on square and triangular lattices, using the Wolff cluster algorithm [19]. For the calculation of the conductivity, for each Ising configuration we used the Franck—Lobb algorithm [20], or explicitly solved Kirchhoff equations. As expected, we found that at the Ising critical point, for g_m, $g_\epsilon \ll 1$, the even component of the conductivity Σ^{even} scales as the energy density, while the odd component Σ^{odd} scales as the order parameter (cf. Fig. 3). As g_m, g_ϵ approach unity, a slow crossover exists to a fractal regime of the Ising clusters, which is crucial for specifying the critical exponent of the conductivity, consistent with the results of Ref. [18] (cf. inset in Fig. 2.)

References [4,8] report 3D mean-field Ising behavior and a small critical region in $(Cr_{1-x}V_x)_2O_3$ and NiS_2 under pressure, respectively, in contrast to the extended critical region with 2D Ising exponents of Ref. [5]. We can understand these experiments by considering the effects of quenched disorder on an Ising critical point. The difference between a quasi-2D and a 3D material is a strongly anisotropic Ising interaction along the direction perpendicular to the planes. Disorder that locally favors the localized over the delocalized state or vice versa, corresponds to a *random magnetic field*, and couples to the order parameter of the Ising transition. This induces density fluctuations. The relevant model for this discussion is the anisotropic 3D random-field Ising model (RFIM),

$$H = -J_{xy}\sum_{\{ij\}_{xy}} S_i S_j - J_z\sum_{\{kl\}_z} S_k S_l + \sum_i h_i S_i, \qquad (7)$$

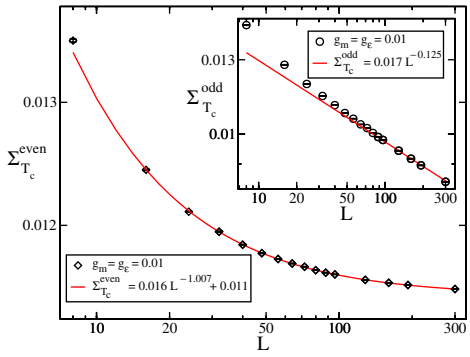

FIG. 3 (color online). Monte Carlo data which verify the expected behavior of the conductivity when g_m, $g\epsilon \ll 1$. Σ^{even} scales as the energy density (see text). Inset: Σ^{odd} scales as the order parameter.

where h_i is a random field with variance Δ. For $d = 3$, there is a continuous phase transition in the 3D random-field Ising model (3DRFIM) universality class [21] for any anisotropy J_{xy}/J_z, an irrelevant operator at the 3D RFIM fixed point. However, for large anisotropy and weak disorder, relevant to the quasi-2D organics, there is a large dimensional crossover regime from 2D RFIM behavior, with an exponentially long correlation length, to the narrower 3D RFIM criticality [22]. What changes between the 3D isotropic materials and the quasi-2D organics is not the universality class, but where the planar correlations become critical. For weak disorder $\Delta \ll J_{xy}$ and strong anisotropy $J_z/J_{xy} \ll 1$, the planes are essentially decoupled and 2D-RFIM behavior holds with $\xi_{xy} \gg 1$ in a large region away from the transition point. When $J_z \simeq J_{xy}$, the critical region is narrow, and controlled by the 3D RFIM fixed point.

With regards to the thermal expansion measurements that claim to measure the heat capacity exponent α [10], we argue that the volume change is proportional to the Ising order parameter of the Mott transition, i.e., $\Delta l \propto m$, yielding $l^{-1} dl/dT \propto t^{\beta-1}$. The thermal expansion diverges with exponent $1 - \beta = 0.875$, consistent with Ref. [10] where it is found in the range 0.8–0.95.

Some major predictions can be drawn from our picture. First, all thermodynamic observables near the Mott critical point should have Ising critical exponents. Second, regarding the critical behavior in quasi-2D organic salts [5], in the clean system, the conductivity along the coexistence line should have the same critical exponent ($\beta_\sigma = 1$) in both mean-field and true-critical regimes. This means that the conductivity jump $\Delta\Sigma_J \equiv \Sigma(T, h = 0^+) - \Sigma(T, h = 0^-)$ along the coexistence line, which should be proportional to the order parameter, should have distinct mean-field and critical regimes, where $\beta_{\Delta\Sigma_J} = 1/2$ and $\beta_{\Delta\Sigma_J} = 1/8$ respectively. Finally, the first-order Mott transition should be broadened by disorder, thereby rounding the conductance jump to a continuous transition [23,24], with effects of glassiness and spatial inhomogeneity [15,25,26].

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