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Magnetic fluctuations near the Mott transition towards a spin liquid state

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Abstract – Dynamics of magnetic moments near the Mott metal-insulator transition is investigated by a combined slave-rotor and Dynamical Mean-Field Theory solution of the Hubbard model with additional fully-frustrated random Heisenberg couplings. In the paramagnetic Mott state, the spinon decomposition allows to generate a Sachdev-Ye spin liquid in place of the collection of independent local moments that typically occurs in the absence of magnetic correlations. Cooling down into the spin-liquid phase, the onset of deviations from pure Curie behavior in the spin susceptibility is found to be correlated to the temperature scale at which the Mott transition lines experience a marked bending. We also demonstrate a weakening of the effective exchange energy upon approaching the Mott boundary from the Heisenberg limit, due to quantum fluctuations associated to zero and doubly occupied sites.

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Introduction. – The Mott metal-insulator transition, wherein electronic waves are localized by short-range electron-electron interactions (see [1] for a review), is one of the most complex phenomenon observed in strongly correlated electronic systems. Even though the appearance of a Mott gap is purely driven by the charge degrees of freedom, it is expected that magnetic fluctuations play a very crucial role in determining the true nature of this phase transition. In the paramagnetic Mott insulator, local moments are indeed well defined objects after their creation at high temperature (at a scale set by the local Coulomb interaction) and before their ultimate antiferromagnetic ordering at the Néel temperature, offering a window in which complex behavior of the spin excitations is yet to be clearly understood. Experimentally, the simplest situation in this respect occurs when the low-temperature magnetic ordering is first order, as in the case of Cr-doped V_2O_3 . Since the magnetic correlations are expected to be weak in this case, many predictions can be made from a single-site approach like the Dynamical Mean Field Theory (DMFT) [2], where local moments are described as freely fluctu-

ating from each other in the insulating state. As a consequence, the low-temperature metallic phase leads upon heating to an insulator, giving a Pomeranchuk-like effect where the entropy gain benefits to the state with magnetic degeneracy. Conversely, other classes of materials, such as the κ -organics, display a continuous magnetic transition into the Néel state, so that the localized spins will experience strong collective fluctuations. The most striking experimental consequence [3–5] lies in the progressive disappearance of the Pomeranchuk effect upon approaching the magnetic ordering temperature, so that the Mott transition lines in the pressure-temperature phase diagram *bend* at low temperature. These qualitative arguments have received recent confirmation from cluster DMFT calculations of the phase diagram of the two-dimensional Hubbard model on a frustrated lattice [6–9], that take into account nearest neighbor magnetic exchange leading to short-range singlet formation. However, the precise connection between the appearance of low-energy magnetic excitations and deviations from the phase diagram of the single-site approach has remained unclear.

Another challenging aspect of correlated fermion materials, that we wish to address here, concerns the inter-

play with disorder, that can lead to a plethora of fascinating physics, such as Anderson-Mott transitions [10] and highly frustrated spin-liquid phases [11, 12], for which extensions of Dynamical Mean Field ideas can also bring interesting light. In particular, introducing magnetic frustration (for instance due to random magnetic couplings) can have a profound influence on the nature of the Mott transition, by modifying the entropy of the magnetic Mott state, and also giving rise to slow spin dynamics [13] that can couple non-trivially to electronic excitations in the metallic phase as well [12, 14, 15]. The real nature of the Mott transition at zero temperature is believed infact to be strongly influenced by the type of magnetic correlations built by exchange interactions, so that the standard DMFT scenario [2] of a second order Brinkman-Rice transition may not apply anymore. However, due the complexity of the impurity models, which involve both fermionic and bosonic baths (see Eq. (2) below), such studies that combine Mott physics with random exchange have been rare [12]. One of our aim here is to provide a first attempt at a fully dynamical solution of this complex physical problem.

Methodology. – In order to investigate the interplay of Mott physics and magnetic fluctuations in a controlled setting, we consider the standard Hubbard model supplemented with long-range fully-frustrated and random Heisenberg exchange interactions, taking frustration as a necessary prerequisite for a non-trivial spin liquid ground state [16] on the insulating side of the Mott transition:

$$H = -t \sum_{\langle ij \rangle, \sigma} d_{i\sigma}^\dagger d_{j\sigma} - \mu \sum_{i\sigma} d_{i\sigma}^\dagger d_{i\sigma} + U \sum_i d_{i\uparrow}^\dagger d_{i\uparrow} d_{i\downarrow}^\dagger d_{i\downarrow} + \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j. \quad (1)$$

Note that hopping parameters t run over adjacent sites of a yet arbitrary lattice, while the exchange terms J_{ij} connect all the local spin variables $\vec{S}_i = \sum_{\sigma, \sigma'} d_{i\sigma}^\dagger \frac{\tau_{\sigma, \sigma'}}{2} d_{i\sigma'}$ in a random fashion (amplitude and sign). Also, in Eq. (1), μ is the chemical potential and U is the local on-site repulsion, which controls the degree of electronic correlations.

This non-trivial Hamiltonian greatly simplifies in the large coordination limit, where all the quantum dynamics can be exactly encapsulated by a single-site action:

$$\mathcal{S} = \int_0^\beta d\tau \left[\sum_{\sigma} d_{\sigma}^\dagger (\partial_\tau - \mu) d_{\sigma} + U d_{\uparrow}^\dagger d_{\uparrow} d_{\downarrow}^\dagger d_{\downarrow} \right] + \int_0^\beta d\tau \int_0^\beta d\tau' \Delta(\tau - \tau') \sum_{\sigma} d_{\sigma}^\dagger(\tau) d_{\sigma}(\tau') + \int_0^\beta d\tau \int_0^\beta d\tau' Q(\tau - \tau') \vec{S}(\tau) \cdot \vec{S}(\tau'). \quad (2)$$

Here, two different baths that depend on imaginary-time τ have been introduced ($\beta = 1/T$ is the inverse temperature): i) hopping processes from the local site into the lattice (and back) result in a retarded hybridization $\Delta(\tau - \tau')$,

which corresponds to the standard DMFT single site action [2]; ii) non-local magnetic coupling exactly amounts to a retarded spin interaction $Q(\tau - \tau')$, providing friction on the local moments, hence reducing their entropy. The latter term is easily obtained through a Gaussian averaging over the disordered exchange coupling J_{ij} in Eq. (1), followed by a dynamical decoupling of a four-spin interaction, as shown in [11, 17]. For a Bethe lattice, both baths are determined self-consistently, through the rather simple set of equations:

$$\Delta(\tau) = t^2 G_d(\tau) \quad (3)$$

$$Q(\tau) = J^2 \chi(\tau). \quad (4)$$

Here, $G_d(\tau) \equiv -\langle d_{\sigma}(\tau) d_{\sigma}^\dagger(0) \rangle$ is the local d -electron single-particle Green's function, and $\chi(\tau) \equiv \langle \vec{S}(\tau) \cdot \vec{S}(0) \rangle$ the local d -electron spin susceptibility. Free d -electrons (for $U = 0$) are endowed with a semi-circular local density of states with half-width $D = 2t$, while the degree of magnetic frustration is controlled by J , the width of the probability distribution used to perform the disorder average.

We now solve the still complicated quantum impurity problem (2), by laying recourse to a method that captures all the crucial physical ingredients at play. A simple possibility is to enlarge the spin symmetry from $SU(2)$ to $SU(N)$ in the large N limit [18], from which the good emergent variables in the magnetic sector are known [11, 12] to be spin-carrying zero-charge fermions (dubbed spinons) f_{σ}^{\dagger} , with $\sigma = 1 \dots N$ (we comment below on the validity of this approach for realistic fermion species with $N = 2$). Charge is then naturally encoded through a second auxiliary field, the phase ϕ dual to the local charge, resulting in the so-called slave-rotor representation [19, 20] of the physical electron: $d_{\sigma}^{\dagger} = f_{\sigma}^{\dagger} e^{i\phi} \equiv f_{\sigma}^{\dagger} X$. Following [19], we have written the phase as a complex bosonic field X constrained as $|X(\tau)|^2 = 1$ through a Lagrange multiplier $\Lambda(\tau)$, so that the action reads (particle-hole symmetry, i.e. $\mu = -U/2$, is assumed here):

$$\mathcal{S} = \int_0^\beta d\tau \left[\frac{|\partial_\tau X|^2}{4U} + \Lambda(\tau)(|X|^2 - 1) + \sum_{\sigma} f_{\sigma}^{\dagger} \partial_\tau f_{\sigma} \right] + \int_0^\beta d\tau \int_0^\beta d\tau' \Delta(\tau - \tau') \sum_{\sigma} f_{\sigma}^{\dagger}(\tau) X(\tau) f_{\sigma}(\tau') X^{\dagger}(\tau') + \int_0^\beta d\tau \int_0^\beta d\tau' \frac{Q(\tau - \tau')}{N} \sum_{\sigma \sigma'} f_{\sigma}^{\dagger}(\tau) f_{\sigma'}(\tau) f_{\sigma'}^{\dagger}(\tau') f_{\sigma}(\tau'). \quad (5)$$

In the large N limit of [19], the Green's functions for the auxiliary particles are readily obtained from Dyson equation in Matsubara frequency:

$$G_X^{-1}(i\nu_n) = \frac{\nu_n^2}{U} + \Lambda - \Sigma_X(i\nu_n) \quad (6)$$

$$G_f^{-1}(i\omega_n) = i\omega_n - \Sigma_f(i\omega_n), \quad (7)$$

where the associated self-energies read in imaginary time:

$$\Sigma_X(\tau) = N \Delta(\tau) G_f(\tau) \quad (8)$$

$$\Sigma_f(\tau) = \Delta(\tau) G_X(\tau) + Q(\tau) G_f(\tau). \quad (9)$$

Similarly, the parameter Λ in Eq. (6) is obtained by the average constraint $G_X(\tau = 0) = 1$. When re-expressed in terms of the emergent variables, the baths entering the self-consistency conditions Eqs. (3-4) can be simplified as: $\Delta(\tau) = t^2 G_d(\tau) = t^2 G_f(\tau) G_X(\tau)$ and $Q(\tau) = J^2 \chi(\tau) = J^2 G_f(\tau)^2$. Together with these self-consistent equations, the above self-energies form a non-linear set of integral equations that we solve numerically. We note that two important limits are naturally incorporated in this scheme: *i*) For $J = 0$ (standard Hubbard model), the spin bath $Q(\tau)$ can be dropped, and we recover the slave-rotor treatment [19] of the usual DMFT equations, so that a Mott metal-insulator transition occurs at a critical value of U/t ; *ii*) More interestingly at $U = \infty$, charge fluctuations are fully suppressed, as the gapped rotor propagator $G_X(\tau)$ can be discarded in Eq. (9). One then recovers the quantum spin liquid equations of Sachdev and Ye [11], where the exchange J is responsible for changing the high temperature Curie behavior of the spin susceptibility $\chi(\tau) \simeq 1/4$ into the slow temporal decay $\chi(\tau) \propto 1/|\tau|$. As a consequence, the temperature dependence of the static susceptibility $\chi(T) = \int_0^\beta d\tau \chi(\tau)$ turns from a Curie law $\chi(T) = 1/(4T)$ at $T \gg J$ into a slower logarithmic divergence $\chi(T) \propto \log(1/T)$ at $T \ll J$. This interesting absence of (spin glass) magnetic ordering at finite temperature, due to the enhanced quantum fluctuations brought by the large N fermionic representation of the spin, allows us to study the onset of magnetic fluctuations in the whole range of temperatures. Some comments are in order here on the validity of the large N limit for describing realistic single-band Heisenberg spin-liquid models at $N = 2$. Indeed, pioneer quantum Monte Carlo simulations [21] of the quantum spin glass model (in the $U = \infty$ limit) reported a very different class of solutions at finite temperature, showing a renormalized finite Curie constant, hence a still $1/T$ diverging spin susceptibility, instead of the $\log(1/T)$ result obtained by Sachdev and Ye at large- N [11]. Further studies [22–24] demonstrated however that these paramagnetic solutions were actually metastable states below the freezing temperature. The correct low temperature physics at $N = 2$ is in fact the one of a nearly quantum critical state (with tiny Edwards-Anderson order parameter $q_{EA} = 0.06$), where low-energy excitations are perfectly described by the large- N limit. In addition, we also note that the increase of quantum fluctuations occurring in our finite U Hubbard model has the tendency to enhance spin-liquid behavior, since we are far from the Heisenberg limit near the Mott boundary, as discussed previously [12, 23]. This discussion thus certainly vindicates our chosen theoretical framework for describing spin-liquid effects. Finally, we also note that most recent studies on this general topic have focused on the more challenging finite-dimension Mott transition towards a spinon liquid [20, 25–31]. Our study aims rather at including explicitly the effect of random magnetic exchange terms, which are notoriously difficult to address on

finite dimensional lattice.

In order to investigate the interplay of Coulomb repulsion U and exchange coupling J close to the Mott metal-insulator transition, one needs to analyze the relative stability of the various phases. This can be achieved from the knowledge of the total free energy (per site) which decomposes into two parts $F_{tot} = F_{imp} + F_{lat}$, respectively related to the local impurity problem (2) solved by the rotor equations (8-9) and to the original lattice model (1) via the self-consistency equations (3-4). The former contribution can be obtained straightforwardly following the derivation of the slave-rotor equation as saddle-point equations [19]:

$$\begin{aligned}
F_{imp} = & -\Lambda + \frac{N}{\beta} \sum_{\omega_n} [\ln G_f(i\omega_n) - i\omega_n G_f(i\omega_n)] \quad (10) \\
& + \frac{1}{\beta} \sum_{\omega_n} [\ln G_X(i\nu_n) - (\omega_n^2/U + \Lambda) G_X(i\nu_n)] \\
& - N \int_0^\beta d\tau [\Delta(\tau) G_X(\tau) G_f(\tau) + \frac{1}{2} Q(\tau) G_f(\tau)^2].
\end{aligned}$$

Taken as a functional of the pseudoparticle Green's functions, the previous expression generates the expected Σ_X and Σ_f self-energies Eqs. (8-9) by differentiation with respect to G_X and G_f respectively. The lattice contribution, associated to the DMFT equations, can similarly be obtained from free energy functionals [32]:

$$F_{lat} = \frac{N}{2} \int d\tau \frac{\Delta(\tau)\Delta(\tau)}{t^2} + \frac{N}{4} \int d\tau \frac{Q(\tau)Q(\tau)}{J^2}. \quad (11)$$

Minimization of F_{tot} with respect to the dynamical fields $\Delta(\tau)$ and $Q(\tau)$ reproduces indeed the DMFT self-consistency condition Eqs. (3-4).

Discussion of the results. – We are now equipped to investigate how magnetic correlations, controlled by the exchange term J , affect the Mott metal to insulator transition. To do so, we first construct the phase diagram in the (U, T) plane for fixed values of the magnetic exchange interaction J . At low temperatures this is done by monitoring the hysteresis in the local d -electron Green's function ($G_d(\tau)$ shows either slow Fermi liquid temporal decay or fast gapped behavior in the metallic and insulating states respectively). This gives rise to two boundary lines $U_{c1}(T)$ and $U_{c2}(T)$, such that a single metallic (resp. insulating) solution exists at a given temperature T for $U < U_{c1}(T)$ (resp. $U > U_{c2}(T)$). Furthermore, we determine from Eqs. (10-11) the critical line $U_c(T)$ at which the free energies of metallic and insulating branches coincide. The resulting phase diagram obtained for several values of J from $J = 0$ to $J/D = 0.5$ is displayed on Fig. 1 in the case of the spin-orbital index $N = 3$. We stress that the particular choice of $N = 3$ allows to match precisely the phase boundary of the usual single-band Hubbard model for canonical spin-1/2 fermions [19], and is helpful to make quantitative comparisons when introducing other degrees of freedom (more orbitals for example) or

new energy scales (random spin exchange as in the present work). We see from Fig. 1 that irrespective of J , there always exists an interaction range $U_{c1}(T) < U < U_{c2}(T)$ bounding a coexistence region. With increasing temperature, the transition lines always merge onto a critical point that occurs at $T_c \simeq D/30$ for $J = 0$, consistent with Refs. [2, 19]. Interestingly, Fig. 1 shows that turning on the Heisenberg interaction J depresses both T_c and U_c , leading also to a shrinking of the co-existence region. This can be understood by an energetic stabilization of the Mott phase via magnetic exchange. Turning to the zero temperature limit, one recovers by the collapse of the $U_c(T)$ and $U_{c2}(T)$ lines at $J = 0$ the continuous metal-insulator transition described by the Brinkman-Rice scenario [2, 20]. In that case, the metal is the true ground state for all $U < U_{c2}(T = 0)$. However, as seen in Fig. 1, the switching of J markedly separates the actual critical line $U_c(T)$ from the two stability lines, so that the Mott transition becomes first order at zero temperature, in agreement with recent findings on models with short-range magnetic correlations via cluster-DMFT [8].

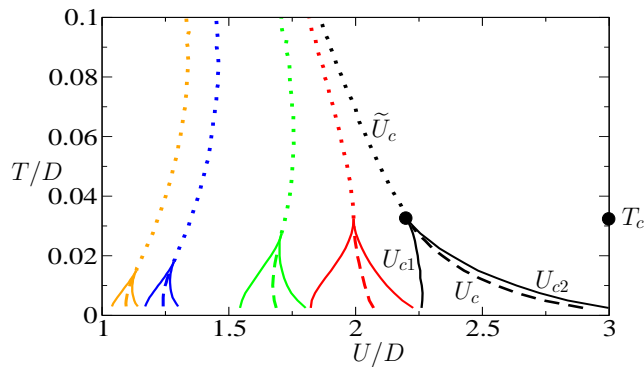


Fig. 1: Metal-insulator transition phase diagram for several values of $J = 0, 0.1, 0.2, 0.4, 0.5$ (right to left). Labels of the various critical lines, as defined in the text, are explicated for $J = 0$. The absence of data for $T/D < 0.003$ is due to the simulations performed at finite temperature using Matsubara Green's functions, but the explored temperature range covers all the interesting physical regimes.

Next, we turn our attention to the orientation of the transition lines as the interaction between the local-moments is increased. At $J = 0$, the $U_c(T)$ line is such that heating the correlated metal always leads to the Mott insulator, a Pomeranchuk-type effect that relates to the high entropy of the insulating phase. This feature persists even *above* the transition temperature: by determining at fixed $T > T_c$ the crossover interaction $\tilde{U}_c(T)$ at which the d -electron Green's function (taken e.g. at $\tau = \beta/2$) displays an inflection point, one distinguishes between a bad metal (i.e. an increasing resistivity with increasing temperature with non quadratic behavior) and a bad insulator (i.e. a non-gapped decreasing resistivity with increasing temperature), see [33–36]. The continuity of the

critical $U_c(T)$ line (determined from the free energy at $T < T_c$) into the crossover line $\tilde{U}_c(T)$ (determined from the Green's function at $T > T_c$) serves as a vindication of our methodology. We also note that such a crossover at $T > T_c$ is seen experimentally in the magnetic and transport measurement of κ -BEDT salts [3, 4]. Now, as J is increased one expects the entropy of the paramagnetic Mott state to be reduced by magnetic exchange, thus weakening the Pomeranchuk lineshape. This is indeed seen in Fig. 1 by a progressive leftward bending of the critical lines at increasing J , and consistent with results of Ref. [7–9]. A particularity of our model is the re-entrance of the Pomeranchuk effect at the lowest observable temperature, which we could attribute to the reduced (with respect to $\log(2)$ of free moments) yet non-zero entropy of the insulating spin liquid state [11, 12]. We have checked also that increasing orbital degeneracy with larger N values tends to weaken all magnetic effects, recovering the standard $J = 0$ phase diagram.

A key point of our study concerns the connection between the exchange induced modifications of the phase diagram (which can be observed e.g. in transport experiments) and the anomalous spin-liquid dynamics. The spin susceptibility $\chi(\tau)$ in the absence of exchange $J = 0$ reduces trivially to a pure Curie law in the whole Mott state $U > U_{c2}$, as checked in Fig. 2. At finite J , Curie

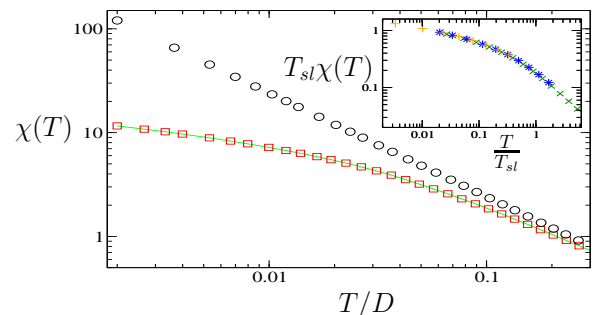


Fig. 2: Spin susceptibility $\chi(T)$ versus temperature for $J = 0$ and large U (circles, corresponding to uncoupled magnetic moments in the Mott phase), and for $J = 0.2$ and $U/D = 1.8$ (squares, corresponding to a spin-liquid Mott phase). A $1/T$ Curie behavior applies to the whole $J = 0$ curve (free moments), while a spin-liquid susceptibility Ansatz $\chi(T) = (1/4T_{sl}) \ln(1 + T_{sl}/T)$ describes the case of finite $J = 0.2$ exchange (full line). The inset demonstrates scaling of our numerical data for three different values of the exchange interaction $J = 0.2, 0.4, 0.8$ in the spin-liquid Mott phase.

behavior is seen only at high temperatures such that $T \gg J$, while departure from the independent local moment picture is observed by a slower logarithmic divergence at zero temperature. A crossover scale T_{sl} characterizing the onset of the spin liquid state can be accurately determined from a fit of $\chi(T)$ by the appealing form $\chi(T) = 1/(4T_{sl}) \ln(1 + T_{sl}/T)$ which follows the expected

high and low temperature regimes (the agreement of this Ansatz with our data is excellent, see inset of Fig. 2 where scaling is demonstrated). This simple Ansatz allows us to extract precisely, for a given value of J , the U -dependence of the spin-spin correlation scale T_{sl} , see Fig. 3, obtained for several J values. We note first that T_{sl} saturates at

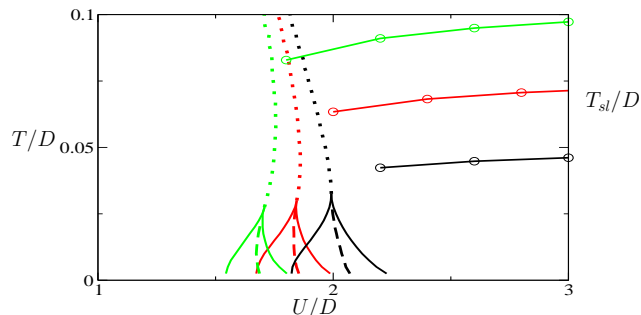


Fig. 3: Superposed plot of the U -dependent spin-liquid onset temperature T_{sl} for several $J = 0.1, 0.15, 0.2$ (circles, bottom to top) with the corresponding metal-insulator phase diagrams (right to left). The merging of T_{sl} with the temperature scale where the crossover lines bend demonstrate the crucial role of the magnetic fluctuations in the destruction of the Pomeranchuk effect.

very large U values to (half) the bare magnetic exchange constant J , an expected result since only spin dynamics plays a role upon freezing of the charge excitations, leaving J as the only relevant scale. More interesting is the net decrease of T_{sl} when U is diminished towards the critical Mott boundary. This feature signals a weakening of the spin correlations in the vicinity of the Mott transition, brought by the finite probability of double and zero occupation of the d state (valence fluctuations in the particle-hole symmetric regime). Quite intriguing is also that the spin susceptibility continues to display the characteristic signature of the spin liquid even in a regime where the ground state is metallic, close to the coexistence region, as seen by the fact that T_{sl} remains finite near the Mott boundary (note that it is difficult to determine T_{sl} precisely in the metallic phase, due to the existence of multiple energy scales). This result is reminiscent of a previous study by Parcollet and Georges [12], where the doping-induced Mott transition (at $U = \infty$) within the fully random Heisenberg model was considered. These authors showed that the magnetic crossover scale T_{sl} persists at small doping, leading to a slush state of incoherent metal with spin liquid correlations, similar to what we observe here at half-filling in the intermediate Coulomb interaction regime. Our last and perhaps most important prediction is the existence of a clear link between the spin liquid coherence scale T_{sl} and the temperature where the bending of the metal-insulator crossover lines takes place, as manifest from Fig. 3. This crucial observation indeed shows that magnetic exchange is responsible for the quenching of the Pomeranchuk effect and the bending of the Mott transition lines, a result previously anticipated on physi-

cal grounds.

Conclusion. – We have investigated the role played by fluctuating local moments coupled via frustrated long-range interactions near the Mott boundary. The metal-insulator transition was found to be modified by the Heisenberg exchange in favor of the insulating state and even to turn discontinuous at zero temperature. Studying the magnetic susceptibility, we were able to connect the bending of the critical lines to the appearance of collective spin correlations, with reduced effective exchange. These effects should be generic to other microscopic models and experimental systems near the Mott transition. One interesting open question is whether the quantum critical behavior of the large dimension Mott transition [37] persists to a quenching of magnetic moments induced by exchange interactions. Recent development of efficient bosonic impurity solvers [38–40] should allow such further explorations of this rich physics.

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