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PHASE DIAGRAM OF SPIN-ORBITAL MODEL

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We show that the mean-field phase diagram of the realistic spin-orbital model derived for a perovskite lattice in three dimensions consists of four different classical magnetic phases which become degenerate at orbital degeneracy. The quantum fluctuations are drastically enhanced and suppress the classical long-range order, providing a new mechanism to stabilize a quantum spin liquid near the multicritical point.

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Evidence is accumulating that orbital degrees of freedom play an essential role in doped Mott–Hubbard insulators [1]. The propagation of a hole doped into such systems is in general quite complex due to the excitonic excitations, as discussed recently for $S=1$ Haldane gap systems [2]. Here we address two important questions in the undoped systems: (i) the modification of the classical magnetic order by the proximity of orbital degeneracy, and (ii) whether a qualitatively new disordered state might be stabilized in three dimensions, similarly as in a two-dimensional (2D) model [3]. This is closely related to one of the fundamental questions discussed recently in localized magnetism: *under which circumstances does classical order collapse due to enhanced quantum fluctuations*? So far, quantum fluctuations were shown to stabilize the disordered phase in three physical situations: (i) in a bilayer Heisenberg model with strong interplane interaction [4], (ii) in a frustrated Heisenberg antiferromagnet (HAF) with long-range $J_1-J_2-J_3$ interactions [5], and (iii) in a 1/5 depleted frustrated square lattice [6]. We show below that a similar instability towards a disordered phase occurs in a natural way in the Mott–Hubbard insulators near an orbital degeneracy.

We consider the simplest undoped three-dimensional (3D) Mott–Hubbard d^9 system (Cu^{2+} ions) in the limit of large Coulomb interaction U, where the charge fluctuations have been integrated out (starting from a multiband model similar to that for high-T_c superconductors and eliminating the virtual transitions to high-energy states, $d_i^9 d_j^9 \Leftrightarrow d_i^8 d_j^{10}$). The effective Hamiltonian includes two e_g orbitals ($x^2-y^2 \sim |x\rangle$, $3z^2-1 \sim |z\rangle$), and the low-energy excitations then consist of spin-flips, as in a HAF, and of orbital excitations. Although such a physical situation has already been discussed by Kugel and Khomskii [7], the complete Hamiltonian and its phase diagram have never been investigated.

We study the resulting *spin-orbital model* with Hamiltonian $H = H_1 + H_2 + H_3$. Here H_1 describes antiferromagnetic (AF) superexchange interactions competing with orbital interactions

$$H_1 = J \sum_{\langle ij\rangle,\alpha} \left[4(S_i \cdot S_j)\left(\sigma_i^\alpha - \frac{1}{2}\right)\left(\sigma_j^\alpha - \frac{1}{2}\right) + \left(\sigma_i^\alpha + \frac{1}{2}\right)\left(\sigma_j^\alpha + \frac{1}{2}\right) - 1\right], \quad (1)$$

where $J = 4t^2/U$, and t is the hopping between $|z\rangle$ orbitals. The Hund rule exchange, $J_{\rm H}$, in the d^8 excited states favours by its very nature ferromagnetic order and gives in addition in leading order $\sim J_{\rm H}/U$,

$$H_2 = \frac{1}{2} J\eta \sum_{\langle ij\rangle,\alpha} \left[2(S_i \cdot S_j)(\sigma_i^\alpha + \sigma_j^\alpha - 1) + 4\sigma_i^\alpha \sigma_j^\alpha - \frac{1}{2}(\sigma_i^\alpha + \sigma_j^\alpha + 1)\right], \quad (2)$$

with $\eta = J_{\rm H}/J$. In Eqs. (1) and (2) S_i refers to the spin at site i, while α labels the cubic (x, y, z) axis. The orbital degrees of freedom are represented by

$$\sigma_i^x = \frac{1}{4}(-\tau_i^z + \sqrt{3}\tau_i^x), \quad \sigma_i^y = \frac{1}{4}(-\tau_i^z - \sqrt{3}\tau_i^x), \quad \sigma_i^z = \frac{1}{2}\tau_i^z, \quad (3)$$

where τ_i^α are Pauli matrices acting as orbital pseudo-spin variables on the states $|x\rangle$ and $|z\rangle$. The degeneracy of the two orbitals is lifted by a "crystal-field" term,

$$H_3 = -E_z \sum_i \sigma_i^z, \quad (4)$$

which can be associated with an uniaxial pressure along the z-axis. The superexchange interactions in H_1 are strongly anisotropic due to the anisotropy in the hopping parameters between the differently oriented e_g orbitals: along the z-direction a superexchange of $4J$ couples the $|z\rangle$ states, while there is no coupling of the $|x\rangle$ states as the superexchange involves the oxygens in the bridge positions. In contrast, the interactions within the (x, y)-planes are $9J/4$ and $J/4$ between two $|x\rangle$ and two $|z\rangle$ orbitals, respectively.

By making a mean-field approximation, one finds four classical phases (Fig. 1) with a two-sublattice long-range order (LRO), which have their counterparts in a 2D model [3]: (i) At large positive E_z, the spins are in $|x\rangle$ orbitals, the (x, y) planes decouple, and we find a planar antiferromagnet (AFxx) well known from the cuprate superconductors. (ii) At large negative E_z, the spins occupy $|z\rangle$ states, and the spin system is a 3D anisotropic antiferromagnet (AFzz). AFxx and AFzz are degenerate along the line $E_z = 0$. (iii) At large $J_{\rm H}/U$, next to the AFzz phase, a mixed-orbital (MOI) phase is found, given at each site by $|i\sigma\rangle = \cos\theta_i |x\sigma\rangle + \sin\theta_i |z\sigma\rangle$, with the sign of θ_i changing between the two sublattices. The spins are ferromagnetic (FM) within (x, y) planes, and antiferromagnetic (AF) along the z-axis. We note that a special case of the MOI phase, with the orbital

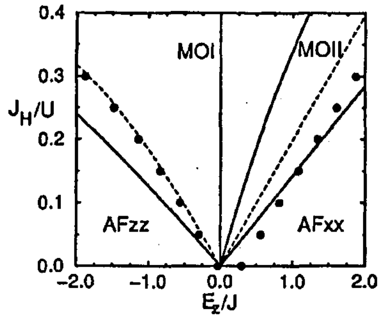

Fig. 1. Mean-field phase diagram (solid lines), soft modes at the $X = (\pi, 0, 0)$ point (dashed lines), and the instabilities of the LRO in AFxx and AFzz phases (filled circles), as functions of E_z/J and J_H/U.

staggering within the (x, y) planes like $x^2 - z^2, y^2 - z^2, x^2 - z^2, \ldots$ ($\cos\theta_i = -1/2$), was proposed by Kugel and Khomskii [7] to explain the planar FM order found in $KCuF_3$. (iv) At large J_H/U and $E_z > 0$, another mixed-orbital (MOII) phase is found, with the same orbital alternation, but AF spin order in the (x, y) planes and FM order along the z-axis. All four phases are degenerate at the multicritical point $M = (E_z, J_H) = (0, 0)$, where the spins and/or orbitals may be rotated freely. The best classical state is either a 3D antiferromagnet with a completely frustrated orbital sector [consider $\langle S_i S_{i+\delta}\rangle = -1/4$ in Eq. (1)], or a disordered spin system which gives the same energy per site ($-3J$) due to the orbital sector. We note that the degeneracy lines of the different classical states in Fig. 1 are qualitatively similar to the frustrated Heisenberg models [5, 6].

The collective excitations in each phase are straightforwardly calculated using a random phase approximation (RPA) in the Green function technique [8]. The number of modes doubles in comparison with the HAF, as the acoustic spin modes are accompanied by optical modes which correspond to excitations in the orbital sector. When the M point is approached, both longitudinal and transverse orbital modes soften. As expected, the transverse modes are more important as they provide the main contribution to the renormalization of ground state energy and magnetic LRO parameter. We studied in detail their behaviour in both AF phases with pure orbital character. In the AFxx phase the transverse orbital mode softens along $k = (\pi, 0, k_z)$ (with the lattice constant $a = 1$) and equivalent lines in the Brillouin zone (BZ), regardless how these critical lines are approached. In the AFzz phase the orbital mode softens along $k = (k_x, 0, 0)$ and equivalent lines. Thus, one finds dispersionless modes at the M point along particular lines in the BZ. As we will show elsewhere [9], the modes exhibit a *quadratic* dependence on $k_\perp$ when these lines are approached in the perpendicular direction which causes a logarithmic divergence of the quantum corrections to the order parameter at the M point. It may be expected that the quantum corrections diverge in a similar way in the MOI and MOII phases, as indicated by the softening of the modes.

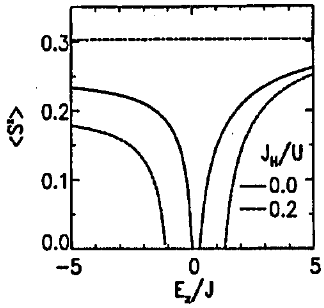

Fig. 2. Renormalized AF LRO parameter $\langle S^z\rangle$ in the AFxx (right) and AFzz (left) phase for two representative values of $J_{\rm H}/U$. The LSW value for a 2D HAF is marked by the dashed-dotted line.

The quantum corrections $\langle\delta S^z\rangle$ to the order parameter in both AFxx and AFzz phases are considerably larger than in a 2D HAF in a broad parameter regime (see Fig. 2). Similarly as in the 2D model [3], LRO is destroyed by the quantum fluctuations when the renormalized $\langle S^z\rangle$ vanishes, and a region where none of the AF phases is stable opens up in Fig. 1. Our preliminary results indicate that a disordered phase with a spin gap stabilizes instead near the M point [9]. Thus, we conclude that *orbital degeneracy* provides a new mechanism to stabilize a quantum spin liquid in three dimensions.

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