Quantum phase transitions and dimensional reduction in antiferromagnets with inter-layer frustration

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For magnets with a fully frustrated inter-layer interaction, we argue that the quantum phase transitions from a paramagnetic to an antiferromagnetic ground state, driven by pressure or magnetic field, are asymptotically three-dimensional, due to interaction-generated non-frustrated inter-layer couplings. However, the relevant crossover scale is tiny, such that two-dimensional behavior occurs in an experimentally relevant low-temperature regime. In the pressure-driven case the phase transition may split, in which case an Ising symmetry related to inter-layer bond order is broken before magnetism occurs. We discuss the relation of our results to recent experiments on BaCuSi$_2$O$_6$.

In modern condensed matter physics, reduced dimensionality presents a fascinating avenue to novel effects arising from strong fluctuations. The standard realization of reduced dimensionality is via systems of chains or planes with a weak three-dimensional (3d) coupling in suitably structured materials. In such a situation, the physics is one-dimensional (1d) or two-dimensional (2d) at elevated energies (or temperatures), while it becomes 3d in the low-energy limit.

Recent experiments near magnetic quantum critical points (QCP) – both in heavy-fermion metals $^{1-3}$ and insulating dimer magnets $^{4-7}$ – have raised speculations about a rather different route to reduced dimensionality, namely through geometric frustration. The idea is that fully frustrated 3d interactions (being not necessarily weak) between 2d units effectively vanish in a well-defined low-energy limit, which in particular can be realized at a QCP. Then, it has been suggested that true 2d behavior could be observed at lowest energies.$^4$

Particularly interesting are the results on Mott-insulating quantum paramagnets, where the spin gap can be closed either by application of a magnetic field or pressure.$^{4,7}$ The field-driven quantum phase transition (QPT), from a paramagnet to an XY-ordered antiferromagnet (AF) at a field $H = H_{c1}$, belongs to the universality class of the dilute Bose gas, and the finite-temperature transition can be understood as Bose-Einstein condensation of magnons. In BaCuSi$_2$O$_6$, the transition line has been found$^8$ to follow $T_c \propto (H - H_{c1})^\psi$, with a shift exponent $\psi = 1$ characteristic of a 2d QCP, and indications for a crossover to the 3d value $\psi = 2/3$ at higher $T$. As BaCuSi$_2$O$_6$ has a body-centered tetragonal (bct) structure of Cu dimers, with a frustrated inter-layer coupling, the results have been interpreted as dimensional reduction at a QCP arising from geometric frustration.

On the theory side, an investigation$^9$ of the ordered state of a frustrated double-layer Heisenberg model using spin-wave theory concluded that inter-layer order is stabilized through an order-from-disorder mechanism, thus rendering dimensional reduction ineffective. In fact, very similar results were obtained much earlier in spin-wave studies of frustrated bct magnets.$^{9,10}$ However, the situation is different at a QCP,$^1$ as the fluctuation-generated inter-layer coupling can be expected to be proportional to the square of the order parameter and hence absent at the QCP, supporting the interpretation of dimensional reduction from frustration. However, as we show below, higher-order processes turn out to be relevant in determining the nature of the phase transitions.

The purpose of this paper is an investigation of QPT in such Mott-insulating magnets with inter-layer frustration, using a detailed symmetry analysis, field theory and bond-operator approaches. Our main results can be summarized as follows: (i) In general, geometric frustration cannot lead to dimensional reduction at asymptotically low energies, due to fluctuation-generated unfrustrated interactions. But their energy scale is tiny at quantum criticality, rendering 2d behavior observable even at very low temperatures. (ii) For the bct lattice, the ordered state breaks both the magnetic and an Ising symmetry. In the pressure-driven case, two scenarios are possible. Either there is a single transition breaking both symmetries, or the transitions is split: the Ising symmetry

![FIG. 1: (Color online) a) bct lattice of dimers. b) top view: two planes with sites shown as circles/crosses, and in-plane (inter-plane) coupling as solid (dashed) lines. c) Illustration of the $Z_2$ symmetry breaking, where the diagonal inter-plane bonds develop a spontaneous asymmetry (see text), together with the simplest lattice compatible distortion. d) $T = 0$ phase diagrams for the coupled-dimer model (1), for the pressure-driven case with and without splitting (see text) and the field-driven case.](http://example.com/fig1.png)
is broken first, which relieves the inter-layer frustration, and the subsequent magnetic transition is conventional, Fig. 1.

**Model Hamiltonian.** To be specific, we will consider a coupled-dimer system on a bct lattice (but we believe our results to be more general), with the Hamiltonian

\[
\mathcal{H} = J \sum_{in} \hat{S}_{in1} \cdot \hat{S}_{in2} + J' \sum_{\langle ij \rangle mn} \hat{S}_{inm} \cdot \hat{S}_{ijnm} + \sum_{i \Delta m \Delta m'} J_{znm}' \hat{S}_{inm} \cdot \hat{S}_{in+\Delta, n+1, m'} - \hat{H} \cdot \hat{S}_{inm} \tag{1}
\]

where \( m = 1, 2 \) labels the two spins 1/2 of each dimer, \( i, j \) are the dimer site indices in each layer, and \( n \) is the layer index. \( J \) and \( J' \) are the AF intra-dimer and in-plane inter-dimer couplings, respectively, while \( J_{znm}' \) represent the frustrated inter-layer couplings (with some specific inter-dimer structure given by the \( mm' \) dependence). The \( \sum_\Delta \) runs over four sites such that the sites \( (in) \) and \( (i+\Delta, n+1) \) are nearest neighbors in \( z \) direction. This Hamiltonian is assumed to be relevant for the material BaCuSi2O6δ (neglecting here the orthorhombic distortions in the low-temperature phase\(^{12}\)).

**Phases.** For \( J > J', |J_z| \), the zero-field ground state of \( \mathcal{H} \) is a paramagnetic singlet, with gapped triplet excitations. If \( J' \) dominates, an AF phase with in-plane ordering wavevector \( \mathbf{Q} = (\pi, \pi) \) is established, and the order in \( z \) direction is frustrated, as discussed below. \( \) For large \( |J_z| \) the in-plane order is ferromagnetic.\(^{12}\) Applying a field to the large-\( J \) quantum paramagnet leads to a Zeeman splitting of the triplet excitations. At a critical field \( H_{c1} \), the gap of the lowest mode closes, and a QPT to a gapless canted phase occurs. Upon further increasing the field, the system is driven into a fully polarized state at \( H_{c2} \). The phase diagram at \( T = 0 \) is thus similar to that of the much-studied bilayer Heisenberg model\(^{13,14}\).

**AF phase: Order from disorder.** While AF ordered planes of classical moments on the bct lattice are uncoupled, zero-point fluctuations of quantum spins lift this large degeneracy. A spin-wave calculation, for spins \( S \) with couplings \( J' \), \( J_z \) and a helical order with wavevector \( (\pi, \pi, Q_z) \), yields an inter-layer contribution to the ground-state energy of the form \( -J_z^2 S / J' \cos^2 Q_z \), favoring collinear (i.e. parallel or antiparallel) order between adjacent planes.\(^{8,9}\) In addition, higher-order terms, in a calculation for a more general ordering pattern, actually stabilize AF order between 2nd-neighbor planes.\(^{10}\) Thus, fully 3d order is stabilized within the stacks of “even” and “odd” planes, but a residual \( Z_2 \) degeneracy is left intact, corresponding to a spin inversion in every second plane – this represents a true symmetry of the AF on the bct lattice, and will be spontaneously broken in the ordered phases.

**Symmetries and magnetic order parameter.** The in-plane magnetism is unfrustrated, allowing to define an order parameter \( \bar{\vec{m}}_n(\mathbf{r}_\|) \), where \( \mathbf{r}_\| \) is the in-plane coordinate and \( n \) the layer index, with the local magnetization operator given by \( \hat{m}_n(\mathbf{r}_\|) = \exp(i\mathbf{Q} \cdot \mathbf{r}_\|) \bar{\vec{m}}_n(\mathbf{r}_\|) \). Expand-
orders in a second, subsequent transition.

Scenario (B) is appealing: From (8) it can be seen that a $\Psi$ condensate generates an unfrustrated vertical hopping of $\phi$ through the term $\lambda(\Psi)\bar{\phi}_n \cdot \bar{\phi}_{n+1}$. Thus, the $\Psi$ ordering transition removes the magnetic inter-plane frustration. In the lattice model, this can be understood as spontaneous bond order which modulates the vertical magnetic couplings within each unit cell and can easily couple to lattice distortions, Fig. 1c, i.e., $\Psi$ ordering is a structural phase transition.

In zero field, a model with negative $u_1$ may follow scenario (B) for the pressure-driven transition: $\Psi$ can be understood as composed of two $\phi$ quanta, hence $m_\Psi \sim 2m_\phi$. The $u_1$ term mediates an attraction between $\phi$ and $\phi_{n+1}$. Approaching the $\phi$ ordering transition, $m_\Psi$ can become smaller than $m_\phi$, implying that $\Psi$ condenses before $\phi$. This requires a sufficiently strong $|u_1|$ (otherwise no true two-particle bound state is generated, and the transition remains in scenario (A)).

**Lattice theory.** We have studied the coupled-dimer model (1) using the bond-operator approach. Starting from a singlet product state on dimer sites $i$, $\prod |i, s\rangle$, we define bosonic operators $t^{\dagger}_{i\alpha}$, which create local triplet excitations $|i, \alpha\rangle = t^{\dagger}_i |i, s\rangle$ where $\alpha = +, -$, and $|i, +\rangle = -|i\uparrow\downarrow\rangle$, $|i, -\rangle = |i\downarrow\uparrow\rangle$ and $|i, 0\rangle = (|i\uparrow\downarrow\rangle + |i\downarrow\uparrow\rangle)/\sqrt{2}$. The Hamiltonian (1) can be re-written in triplet operators.\textsuperscript{14-16} The quadratic part reads:

$$H_2 = \sum_{\bar{q}\alpha} (A_{\bar{q}} - \alpha H) t^{\dagger}_{\bar{q}\alpha} t_{\bar{q}\alpha} + B_\parallel^2 (t_{\bar{q}\alpha} t_{-\bar{q}\alpha} + h.c.)$$

with $\alpha = -\alpha$, $A_\parallel = J + B_\parallel$, $B_\parallel = J' \cos(q_z) + \cos(q_x) + 2J_z \cos(q_z/2) \cos(q_y/2) \cos(q_x)$, where $J_z = J_{11} + J_{22} - J_{12} - J_{21}$, and the field $H$ is in $z$ direction. In addition to $H_2$ the Hamiltonian contains a quartic in-plane triplet term $H_4$: \textsuperscript{14} for $J_{4z} = J_{11} + J_{22} + J_{12} + J_{21} \neq 0$ and $J_{4z} = J_{11} - J_{22} \pm (J_{12} - J_{21}) \neq 0$ quartic and cubic inter-plane interactions, $H_4$, and $H_3$, arise, respectively.

The eigenvalues of $H_2$, $\omega_{\bar{q}} = (A_\parallel^2 - B_\parallel^2)/2 - \alpha H$, are independent of $q_z$ at in-plane wavevector $\bar{q}_z = (\pi, \pi)$. In this linearized bond-operator theory, interactions between the order-parameter fluctuations – represented by triplon quasiparticles – are ignored. The most important corrections arise from the hard-core constraint $H_U$, $\sum_i t_{\bar{q}\alpha} t_{\bar{q}\alpha} \leq 1$.\textsuperscript{14} Following the self-consistent diagrammatic approach of Ref. 14, we have calculated the renormalized triplon dispersion, focusing on how interactions lift the degenerate vertical spectrum, Fig. 3. Taking into account $H_U$ yields a dispersion along $(\pi, \pi, q_z)$ proportional to $J_z^2 \cos(2q_z)$, corresponding to the process in Fig. 2a. Further analysis shows that a similar term, but with prefactor $J_z^2$, is generated to second order in $H_4z$. (This cannot be read off from the order-parameter theory $S_\Psi$ because of the assumed local form of the quartic term.) Hence, the bandwidth along $(\pi, \pi, q_z)$ is at most of order $J_z^2$.\textsuperscript{18} (This result is not changed by $H_3z$.)

To assess the dynamics of the Ising parameter $\Psi$, we have studied bound states of two triplons in the singlet
dispersion. The dominant effect of the two-particle continuum (shaded), calculated using bond operators, for $J'/J = 0.15$, $-J_z = J'_{1z} = 0.2 J'$, $H = 0$. The inset show an energy zoom into the almost flat vertical dispersion. The dominant effect of $H$ is a Zeeman shift of $-\alpha H$ of the triplons.

channel by solving a Bethe-Salpeter equation, with a four-point vertex as input. To avoid the necessity for full self-consistency in the two-particle sector, we have added to $\mathcal{H}_4$ by hand the biquadratic $H_{\text{coll}}$ (7). The effective binding force is then given by $(J_{1z} - J_{\text{coll}})$: note that the binding from $\mathcal{H}_4$ is due to a subtle quantum effect of singlet formation, which is not contained in the order-parameter field theory, but is in fact very common for frustrated spin-1/2 systems.

For sufficiently strong positive $(J_{1z} - J_{\text{coll}})$ we find a bound state below the two-particle continuum, whose wavefunction changes sign under 90-degree in-plane rotations of the internal coordinate, consistent with (3) and Fig. 1c. Close to the pressure-driven phase transition the bound state (with dispersion minimum at total momentum $Q = 0$) is pulled below the single-particle gap, due to the weak vertical triplon dispersion. Clearly, a condensation of this bound state corresponds to the $Z_2$ symmetry breaking advocated above. This existence of the bound state depends on microscopic details, and a comprehensive numerical analysis is difficult due to finite-size effects. (For the parameter values of Fig. 3, our calculations indicate no bound state.)

**Pressure vs. field tuning.** So far, most considerations were for $H = 0$. In finite field, the spin symmetry is reduced to $U(1)$, and the expressions in (2,4,5,8) are modified accordingly. However, the symmetry analysis in the paramagnetic phase, leading to (4,5), remains valid. The most important difference is in the bound-state behavior: Close to $H_{c1}$ a possible singlet bound state involves at least one high-energy triplet and will never condense. Then, scenario (A) always applies, with a 3d QPT which breaks the $U(1) \times Z_2$ symmetry and obeys mean-field exponents.

**Discussion.** We conclude that dimensional reduction in quantum-critical frustrated bct magnets does not occur at lowest energies, due to interaction effects. However, the crossover scale below which 3d behavior is established (which is given by the vertical dispersion) is tiny, $E_z \propto J'/J^3$ (instead of $J_z$ as in an unfrustrated system). Above $E_z$, the shift exponent $\psi$ will take its 2d value. In the pressure-driven case, depending on microscopic parameters the ordering transition may be split, then bond order occurs before magnetic order.

**Relevance to BaCuSi$_2$O$_6$**. Recent neutron scattering hints on the presence of multiple triplon excitations in zero field, which indicate an enlarged unit cell with inequivalent dimers. Assuming that the lattice modulation occurs along the $c$ axis, one arrives at a qualitatively distinct scenario for “dimensional reduction”: The condensate established at $H_{c1}$ is strongly inhomogeneous in $z$ direction, and hence effectively 2d – note that frustration is not a required ingredient in this scenario. (This conclusion is supported by the fact that the measured vertical triplon dispersions are tiny for all in-plane wavevectors, at variance with the results in Fig. 3.)

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1. H. v. Löhneysen, A. Rosch, M. Vojta, and P. Wölfle, Rev. Mod. Phys. **79**, 1015 (2007).
2. O. Stockert et al., Phys. Rev. Lett. **80**, 5627 (1998).
3. O. Trovarelli et al., Phys. Rev. Lett. **85**, 626 (2000).
4. S. E. Sebastian et al., Nature **441**, 617 (2006).
5. M. Jaime et al., Phys. Rev. Lett. **93**, 087203 (2004).
6. Ch. Rüegg et al., Phys. Rev. Lett. **93**, 257201 (2004).
7. Ch. Rüegg et al., Nature **423**, 62 (2003).
8. M. Maltseva and P. Coleman, Phys. Rev. B **72**, 174415 (2005).
9. E. Rastelli, S. Sedazzari, and A. Tassi, J. Phys. Cond. Matter **2**, 8935 (1990).
10. T. Yildirim, A. B. Harris, and E. F. Shender, Phys. Rev. B **53**, 6455 (1996).
11. C. D. Batista et al., Phys. Rev. Lett. **98**, 257201 (2007).
12. E. C. Samulon et al., Phys. Rev. B **73**, 100407(R) (2006).
13. A. W. Sandvik and D. J. Scalapino, Phys. Rev. Lett. **72**, 2777 (1994).
14. V. N. Kotov et al., Phys. Rev. Lett. **80**, 5790 (1998).
15. S. Sachdev and R. N. Bhatt, Phys. Rev. B **41**, 9323 (1990).
16. M. Matsumoto, B. Normand, T. M. Rice, and M. Sigrist, Phys. Rev. Lett. **89**, 077203 (2002).
17. O. P. Sushkov and V. N. Kotov, Phys. Rev. Lett. **81**, 1941 (1998).
18. O. Rösch and M. Vojta, unpublished.
19. N. Read and S. Sachdev, Phys. Rev. Lett. **62**, 1694 (1989).
20. Ch. Rüegg et al., Phys. Rev. Lett. **98**, 017202 (2007).
21. Ch. Rüegg, private communication.