Field induced disordered-local-moment phase in site-diluted spin-gap antiferromagnets

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Abstract. Site dilution of spin-gapped antiferromagnets leads to localized free moments, which can order antiferromagnetically in two and higher dimensions. Here we show how a weak magnetic field drives this order-by-disorder state into a novel disordered-local-moment (DLM) phase, characterized by the formation of local singlets between neighboring moments and by localized moments aligned antiparallel to the field. This disordered phase is characterized by the absence of a gap, as is the case in a Bose glass. The associated field-driven quantum phase transition is consistent with the universality of a superfluid-to-Bose-glass transition. The robustness of the DLM phase and its prominent features, in particular a series of pseudo-plateaus in the magnetization curve, makes it accessible and relevant to experiments.

Valence bond solids (VBSs) in spin-gapped antiferromagnets represent some of the most fundamental examples of quantum-disordered states in condensed matter systems. The nature of such states is by now well understood theoretically and has been extensively verified experimentally. A variety of mechanisms, such as increased strength of certain bonds in the magnetic Hamiltonian [1, 2], magnetic frustration [3], and the Haldane mechanism [4], can render classical Néel order unstable towards the formation of local singlets, which arrange themselves...
A central focus of theoretical and experimental investigations has been the effect of doping on such states. In particular, it was soon realized theoretically, [9, 10] and observed experimentally [11, 12], that doping a VBS with static, non-magnetic impurities leads to the intriguing phenomenon of order-by-disorder (OBD): localized $S = 1/2$ magnetic moments appear close to the impurity sites and interact effectively via a long-range network of unfrustrated (albeit random) couplings, which decay exponentially with the inter-moment distance. These interactions, although weak, are sufficient for the local moments (LMs) to order antiferromagnetically at experimentally relevant temperatures [11]. Given the random nature of the inter-moment couplings, the OBD state induced by doping is extremely inhomogeneous, as it contains a large variety of energy scales which depend exponentially on the spatial distribution of the impurities. In this paper, we study the evolution of the OBD state upon application of a magnetic field, which represents a straightforward experimental probe of energy scales in magnetic systems. Precisely due to the large distribution of the effective couplings between the LMs, we find an amazingly rich response of the system to the applied field. The field scan reveals that the long-range order in the system is extremely tenuous for large spin gaps, and its field-driven destruction leaves behind local singlets (or spins oppositely polarized to the field) on even- (or odd-) numbered clusters of LMs, which are coupled at energies higher than the scale characteristic for Néel order (see figure 1). The local polarization fields for these LM clusters cover a continuous range, so that the magnetization process with increasing field continues also after destruction of the OBD, and the resulting DLM phase is gapless. The DLM phase has been first introduced in [13] in a site-diluted bilayer system (with the previous name ‘disordered-free-moment’); in this work, we discuss it in a planar dimer system, unveiling its microscopic nature and its striking macroscopic features. In particular the magnetization curve within this phase shows prominent features of
shows the evolution of we extrapolate a staggered magnetization magnetic order of the network of coupled LMs. In particular, for the couplings uniform susceptibility the ordered moment under application of a field. It reveals that the antiferromagnetic order is density where \( \rho \). Using stochastic series expansion (SSE) quantum Monte Carlo simulations based on the directed-loop algorithm \([1] \), we study an effective model for the network of LMs, consisting of randomly distributed \( S = 1/2 \) spins with effective couplings \( J_{ij} \).

\[
\mathcal{H} = J \sum_{i \in A} \epsilon_i \epsilon_{i+\hat{x}} S_i \cdot S_{i+\hat{x}} + J' \sum_{i \in A} \epsilon_i \epsilon_{i+\hat{y}} S_i \cdot S_{i+\hat{y}} + J' \sum_{i \in B} \sum_{\hat{d}=\hat{x},\hat{y}} \epsilon_i \epsilon_{i+\hat{d}} S_i \cdot S_{i+\hat{d}} - h \sum_i \epsilon_i S_i^z. \tag{1}
\]

Here \( i \) runs over the two sublattices (A and B) of a square lattice, \( \hat{x} \) and \( \hat{y} \) are the two lattice vectors, and \( \epsilon_i \) is the random dilution variable taking values 0 and 1 with probability \( p \) and \( 1-p \) respectively; \( h = g \mu_B H \) is the applied field. The couplings \( J > J' \) determine the subset of strong antiferromagnetic bonds: for \( J'/J < 0.523 [2] \), the bond anisotropy stabilizes a dimer–singlet ground state against the conventional Néel ordered state of the square-lattice antiferromagnet. All the results presented here refer to the field and doping effects deep within the dimer–singlet regime at \( J'/J = 1/4 \).

The presence of lattice vacancies induces LMs which are localized in the vicinity of the unpaired spins that have lost their \( J \)-neighbor. Perturbation theory \([10, 13, 15] \) provides an effective coupling \( J_{ij} \approx (-1)^r - \xi_0 / \epsilon_i \) \( \exp(-r/\xi_0) \) between these LMs, where \( r = |i - j| \), \( \xi_0 \) is the correlation length of the undoped system. We choose \( J_i \approx J \exp(1/\xi_0) \) in order for \( J_{ij} \) to correctly reproduce the limit \( J' \) for neighboring unpaired spins. For a deeper understanding of the Hamiltonian equation (1), it is illuminating \([16] \) to study an effective model for the network of LMs, consisting of randomly distributed \( S = 1/2 \) spins with effective couplings \( J_{ij} \).

\[
\mathcal{H}_{LM} = \frac{1}{2} \sum_{i,j} J_{ij} S_i \cdot S_j - h \sum_i S_i^z. \tag{2}
\]

We investigate the original and the effective Hamiltonian, given by equations (1) and (2), using stochastic series expansion (SSE) quantum Monte Carlo simulations based on the directed-loop algorithm \([17] \). For the original Hamiltonian equation (1) we study \( L \times L \) lattices up to \( L = 40 \) with dilution \( p = 1/8 \), whereas for the effective model equation (2) we randomly distribute spins on the same lattice sizes with a density \( p \) equal to that of the vacancies in the original model. Disorder averaging is typically performed over \( \approx 300 \) realizations. The ground-state properties are systematically obtained using a \( \beta \)-doubling approach \([18] \). Inverse temperatures up to \( \beta J = 2^{15} \) are necessary to observe the physical \( T \to 0 \) behavior. In the following, we focus our attention on the uniform magnetization per spin \( m_u = 1/N \sum_i \langle S_i^z \rangle \), where \( N \) is the total number of spins in the system considered; on the uniform susceptibility \( \chi_u/J = \partial m_u/\partial h \); on the staggered magnetization \( m_s = \sqrt{S^z(\pi, \pi)/L^2} \), where \( S^z(\pi, \pi) = 1/(2L^2) \sum_{ij} \langle S_i^z S_j^z + S_i^y S_j^y \rangle \) is the transverse static structure factor; on the correlation length \( \xi \), extracted from the \( q \)-dependent structure factor; and on the superfluid density \( \rho_s = 1/(2\beta J) \langle W^2 + W'^2 \rangle \), where \( W_i(x) \) are the winding numbers of the SSE worldlines.

In the absence of a magnetic field, the effective couplings \( J_{ij} \) give rise to long-range magnetic order of the network of coupled LMs. In particular, for \( J'/J = 1/4 \) and \( p = 1/8 \) we extrapolate a staggered magnetization \( m_s = 0.032(3) \) in the thermodynamic limit. Although the couplings \( J_{ij} \) range between 0 and \( J' \), the average coupling strength responsible for the long-range order turns out to be much smaller than \( J' \) \([13] \). Figure 2 shows the evolution of the ordered moment under application of a field. It reveals that the antiferromagnetic order is

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already destroyed at a field $h \ll J'$, namely at $h = h_{DLM} = 0.007(1) J$. Yet, a striking feature of this disordered phase is that the destruction of long-range order is not accompanied by the full polarization of the LMs, as it would ordinarily happen in a homogeneous antiferromagnet. At the critical field $h_{DLM}$ the uniform magnetization is found to be $m_u = 0.0208(6)$, much less than the value $m_u = pS = 1/16$ corresponding to fully polarized LMs, which is attained at a much larger field $h_{plateau}/J \approx 0.5$. Consequently the DLM phase, appearing between $h_{DLM}$ and $h_{plateau}$, is highly unconventional, retaining a finite uniform susceptibility and a gapless spectrum. For $h > h_{plateau}$ the saturation of the magnetization of LMs leads to the full restoration of a gapped disordered phase due to the field $15$.

As shown in figure 2, the main features of the field dependence $m_u$ and $m_s$ for $h < h_{plateau}$ in the doped coupled-dimer model of equation (1) are very well reproduced by the effective model equation (2), for which we take $\xi_0 = 1$ as found by simulations at $h = 0$ and $p = 0$. In particular, the fundamental property of a DLM phase with $m_s = 0$ and $\chi_u > 0$ is confirmed in the effective model. This reveals that the LMs are essentially the only degrees of freedom responding to a field $h < h_{plateau}$ in the doped coupled-dimer system.
The novel QPT between the OBD and the DLM phase is studied using finite-size scaling analysis of the correlation length, \( \xi = L F_\xi[L^{1/\nu} \delta h] \), of the superfluid density \( \rho_s = L^{d-2+\nu} F_{\rho_s}[L^{1/\nu} \delta h] \), and of the staggered magnetization \( m_s = L^{-\beta/\nu} F_{m_s}[L^{1/\nu} \delta h] \), where \( \delta h = h - h_{DLM} \), as shown in figure 3. This allows us to extract the critical exponents \( z, \nu \) and \( \beta \). For the original Hamiltonian equation (1) we find \( z = 2.0(1), \nu = 1.0(1) \) and \( \beta = 0.9(1) \). These estimates are also confirmed in the effective LM model. The above exponents are fully consistent with those of the 2\( d \) superfluid-to-Bose-glass (SF–BG) QPT previously studied in diluted bilayers [13], and with the exponents found at the BG–SF transition for higher fields for the model equation (1) [19]. In particular \( z \) is in agreement with the general theoretical prediction \( z = d [20] \), and \( \nu \) satisfies the fundamental Harris criterion \( \nu \geq 2/d \). Altogether the present results and those of [13, 19] point towards a general SF-BG universality in \( d = 2 \) for order–disorder transitions at which the uniform susceptibility \( \chi_u \) remains finite, corresponding to the absence of a gap.

In the DLM phase, the magnetization curves of the original Hamiltonian equation (1) and the effective model equation (2) show a dramatic feature: beside the large plateau appearing at \( h \geq h_{\text{plateau}} \), one observes the presence of apparent intermediate plateaus at around 3/4 and 95% of the saturation magnetization. A detailed study of the temperature-dependent susceptibility in this field region reveals that these features are actually PPs, which retain an extremely small slope (figure 2). For both \( \mathcal{H} \) and \( \mathcal{H}_{LM} \) the first PP extends up to \( h \approx 0.7 J' \); a second PP markedly appears around \( h \approx 1.2 J' \) for \( \mathcal{H} \) (it is rounded off for \( \mathcal{H}_{LM} \) [19]); and, for both models, the true saturation plateau is only attained at \( h \approx 2 J' \). Despite their gaplessness, the PPs are seen to be resistant to thermal effects, up to temperatures \( T \sim J/100 \) [19]. These fundamental features can be understood within the picture of strongly interacting clusters of LMs in the DLM phase. As shown in figure 1, the zero-field OBD phase is essentially inhomogeneous due to the random nature of the couplings. A majority of LMs are spaced from each other by an average distance \( \langle r \rangle = p^{-1/d} \), large in the small dilution limit, and interact via weak average couplings \( \langle J_{\text{eff}} \rangle \sim p J' \) [14]. However, fluctuations in the spatial distribution of the impurities also lead to small clusters of LMs located on neighboring sites, and thus interacting with much stronger couplings \( J' \). If antiferromagnetically coupled in even-numbered clusters, the strongly interacting LMs participate only marginally in the OBD state of the system, and have...
a significant singlet component in their ground state wavefunction. This is directly revealed in a histogram of the bond energies (figure 4(a)) $E_b = J_b \langle S_{1,b} \cdot S_{2,b} \rangle$ where $J_b = J, J'$ and $(1, b), (2, b)$ are the two neighboring lattice sites participating in the bond b. Beside the peak at $E_b \approx -3J/4$, corresponding to singlets on intact dimers, a further peak at $E_b \approx -3J'/4$ is observed, corresponding to LM dimer singlets, as well as a peak at $E_b \approx -J'/2$ corresponding to LM trimers.

Applying a magnetic field $h \gtrsim \langle J_{\text{eff}} \rangle$ clearly has the effect of destroying the long-range order of the LMs, but, as sketched in figure 1, at the same time the LM singlets are left intact, while odd-numbered clusters are only partially polarized and their sites with higher coordination exhibit a spin polarized opposite to the field (e.g. the central site of a trimer). This has the fundamental consequence that the antiferromagnetic order disappears but the LMs are far from saturation. This is clearly seen in the histogram of the local magnetic moments $\langle S^z_i \rangle$ of the unpaired spins only (figure 4(b)): for small fields, a double-peak structure appears with a peak at $\langle S^z_i \rangle = S$ corresponding to fully polarized LMs, and a strong quantum peak at $\langle S^z_i \rangle = 0$.

**Figure 4.** Histogram of bond energies (a) and local magnetizations for unpaired spins (b).
corresponding to LM singlets. The large tails for $0 < \langle S_i^z \rangle < S$ and for $\langle S_i^z \rangle < 0$ come instead from LMs in odd-numbered clusters. In fact, we can resolve two more peaks at $\langle S_i^z \rangle = 1/3$ and $\langle S_i^z \rangle = -1/6$ at larger fields, which correspond to partially polarized spins in LM trimers. We do not find any significant dependence of peaks at $\langle S_i^z \rangle = 0$ and $\langle S_i^z \rangle = 1/3$ on lattice size in the DLM phase, revealing that the statistics of LM dimers and trimers is fully captured by the system sizes under investigation. LM clusters have widely different local gaps to full polarization, both due to their geometric structure (dimers, trimers, quadrumers, etc), and to the local field they experience from the other LMs. Yet the distribution of rare LM clusters clearly assigns dominant statistical weight to the dimers, and this simple geometric fact is the reason for the appearance of the first PP: the magnetization process nearly stops until the local gap of the dominant LM dimers is overcome at $h \lesssim J'$. Nonetheless, the slope remains finite because the LM dimers have a distribution of local gaps. The magnetization value and the field location of the PP can be quantitatively related to the statistics of LMs clustered in dimers [19]. Analogously, one can quantitatively associate the second plateau with the statistics of the LM trimers [19]. Higher-order plateaus associated with larger local polarization fields should be expected, but they cannot be resolved within the given numerical accuracy.

From the above data a clear picture of the DLM phase emerges: in this phase, a majority of the LMs are polarized, but antiparallel spins exist, corresponding to rare LM clusters. Upon a spin-to-hardcore-boson transformation $S^+ = b$, $S^- = b^\dagger$ and $S^z = 1/2 - b^\dagger b$ (where $\{b, b^\dagger\} = 1$ on the same site), these antiparallel spins take the nature of bosonic spin-down quasiparticles (↓-QPs); their Hamiltonian can be derived from the effective Hamiltonian of the LMs, equation (2), in the form

$$\mathcal{H}_{\text{Bose}} = \frac{1}{2} \sum_{i,j} \left[ \frac{J_{ij}}{2} \left( b_i b_j^\dagger + \text{h.c.} \right) + J_{ij} n_i n_j \right] - \mu \sum_i n_i, \quad (3)$$

where $n_i = b_i^\dagger b_i$ and the chemical potential reads $\mu = -h + \sum_{ij} J_{ij}$. At low fields in the OBD phase the ↓-QPs form a superfluid condensate, with order parameter $m_s = \langle \langle b_i \rangle + \langle b_i^\dagger \rangle \rangle / 2$; this condensate is progressively depleted by the applied field (acting as a negative chemical potential), up to a critical concentration at which the remaining ↓-QPs undergo localization around the strongly interacting LM clusters of dimers and trimers, giving rise to a characteristic Bose-glass state [20]. The gapless nature of this phase is due to the quasi-degeneracy of some of these localized ↓-QP states with the local vacuum (corresponding to full polarization of a LM cluster).

Throughout the paper we have presented results for a selected $J'/J$ ratio and for a single $p$ value. With this choice of parameters, we find a DLM phase over a very broad field range, $\sim J'/2$. Therefore, variations of $J'$ which are small compared to this scale cannot significantly affect the picture we have discussed. As for the stability upon changing $p$, we observe that all features of the phase diagram are expected to display a smooth power-law dependence on $p$, with easily predictable effects. The width of the OBD phase is expected to linearly depend on $p$, given that $h_{\text{DLM}}$ reflects the average energy scale of the effective interactions between LMs, so that $h_{\text{DLM}} \sim \langle J_{\text{eff}} \rangle \sim p$. The upper bound on the DLM phase is on the other hand essentially independent of $p$, and it is attained by saturating the magnetization of the largest possible LM clusters, which are in practice segments of spin chains with a saturation field $\sim 2J'$, as found in the example studied in the paper. The width of the PPs is $\sim J'$, being related to the gap of LM clusters (renormalized by the effective field created by the polarized LM), so it is only
weakly dependent on $p$. The height of the PPs on the other hand is directly proportional to the probability of the LM dimers, trimers, etc, and hence the distance of the PPs from the saturation plateau scales as $p^2$, $p^3$ etc, respectively.

It is evident from the above results that the DLM phase is relevant for experiments on site-diluted spin-gapped antiferromagnets which display an OBD phase in zero field. The fundamental condition for the observation of the DLM phase is that the spin gap of the pure system be much larger than the maximum energy scale of the LM interaction (average inter-dimer coupling in weakly coupled dimer systems, inter-chain coupling in Haldane chains). This condition is necessary to ensure that the physics of the field response of the LMs is well separated in energy from that of the field-induced ordered state (for a detailed discussion, see [13]). Candidate spin-gap compounds include, among others: Sr$_2$Cu(BO$_3$)$_2$ [21], in which dimers are arranged in a quasi-2D structure similar to the one investigated in our paper; CaV$_4$O$_9$ [22], in which spins are arranged in a 1/5-depleted square lattice, and whose behavior under doping (but in zero magnetic field) has been investigated in [23]. Joint magnetometry and neutron scattering measurements at relatively low fields should be sufficient to fully pinpoint this phase by demonstrating the absence of spontaneous order and the finite susceptibility down to zero temperature. Furthermore, NMR measurements can show the rich structure of the distribution of local magnetic moments, similar to the histogram of figure 4. The low-field location of the DLM phase and its strong physical signatures in the magnetic observables make it the most accessible novel disordered phase in quantum magnets with lattice randomness.

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