Mott Glass in Site-Diluted $S = 1$ Antiferromagnets with Single-Ion Anisotropy

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The interplay between site dilution and quantum fluctuations in $S = 1$ Heisenberg antiferromagnets on the square lattice is investigated using quantum Monte Carlo simulations. Quantum fluctuations are tuned by a single-ion anisotropy D. In the clean limit, a sufficiently large $D > D_c = 5.65(2)J$ forces each spin into its $m_S = 0$ state, and thus destabilizes antiferromagnetic order. In the presence of site dilution, quantum fluctuations are found to destroy Néel order before the percolation threshold of the lattice is reached, if D exceeds a critical value $D^* = 2.3(2)J$. This mechanism opens up an extended quantum-disordered Mott-glass phase on the percolated lattice, characterized by a gapless spectrum and vanishing uniform susceptibility.

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Strongly interacting quantum systems on random lattices offer the possibility of realizing genuine quantum phases with unconventional properties [1]. The inhomogeneous nature of such disordered lattices can lead to the existence of strongly correlated regions with excitations of arbitrarily low energy, and yet an overall absence of long-range order. Therefore quantum "glassy" phases are possible which, at the same time, have a quantum-disordered ground state and a finite low-energy density of states, not associated with a Goldstone mode [1–3]. A large variety of physical systems potentially lends itself to realize this theoretical scenario as, e.g., ^{4}He films on disordered substrates [4], type-II superconductors with columnar defects [5], and trapped cold atoms in disordered optical potentials [6].

In this Letter we numerically demonstrate the emergence of a highly nontrivial interplay between lattice randomness and quantum fluctuations in low-dimensional antiferromagnets. Disorder can typically be introduced in antiferromagnets by dilution of the magnetic lattice with nonmagnetic ions, and an extremely high level of control on the concentration of dopants can be reached [7]. Recent experiments on the site-diluted $S = \frac{1}{2}$ square-lattice antiferromagnet have shown that the destruction of magnetic long-range order (LRO) due to doping occurs at the percolation threshold of the lattice [7], suggesting that this particular system is too far from a quantum-critical point to develop a quantum-critical regime triggered by site dilution [8,9]. In this Letter, we consider a realistic magnetic model on the square lattice in which the strength of quantum fluctuations can be tuned at will, driving the system from a classically ordered state to a quantum disordered one. Nonlinear quantum fluctuations enhanced by lattice disorder give rise to a novel quantum-disordered phase which is gapless and has a vanishing uniform susceptibility. An exact bosonic mapping of the model allows us to identify this regime with a Mott-glass phase [3] of a commensurately filled lattice gas of interacting bosons. The application of a magnetic field to such a phase drives it into a disordered Bose-glass phase [1], which maintains a gapless spectrum but acquires a finite susceptibility.

We investigate the $S = 1$ square-lattice anisotropic Heisenberg antiferromagnet (SLAHAF) with site dilution, whose Hamiltonian reads

$$\mathcal{H} = J\sum_{\langle ij\rangle} \epsilon_i\epsilon_j \boldsymbol{S}_i \cdot \boldsymbol{S}_j + D\sum_i \epsilon_i (S_i^z)^2 - hJ\sum_i \epsilon_i S_i^z. \quad (1)$$

Here $\boldsymbol{S}$ denotes $S = 1$ spin operators, and $\langle ij\rangle$ enumerates pairs of nearest-neighbor sites on the square lattice. The variables ϵ_i take the values 0 or 1 with probability p and $1 - p$, respectively, where p is the concentration of nonmagnetic dopants. We make use of the stochastic series expansion quantum Monte Carlo (SSE-QMC) method based on the operator-loop algorithm [10], which allows us to faithfully monitor the $T = 0$ physics by a β-doubling approach [8] on $L \times L$ lattices with L up to 36 sites. The results are typically averaged over at least 200 disorder realizations.

In the clean limit $p = 0$, and at zero magnetic field $h = 0$, the SU(2) symmetric version of this model for $D = 0$ is known exactly to show Néel LRO [11]. The presence of finite single-ion anisotropy ($D > 0$) reduces the symmetry to U(1), and antiferromagnetic ordering takes place in the xy plane. In the limit $D/J \gg 1$ the ground state becomes $|\Psi_0\rangle = \prod_i |m_S = 0\rangle_i$ to minimize the anisotropy term. Such a state has no antiferromagnetic order, since all spin-spin correlation functions are simply zero. Therefore, at a critical ratio $(D/J)_c$ a quantum phase transition [12] occurs between the xy-ordered regime and a gapped spin-liquid state with short-range correlations. Our QMC simulations of Eq. (1) with $h, p = 0$ provide an estimate of $(D/J)_c = 5.65(2)J$, obtained by linear scaling of the correlation length for the $x(y)$ spin components $\xi^{xx(yy)} \sim L$. The quantum-critical scaling of the static structure factor $S^{xx(yy)}(\pi, \pi) \sim L^{\gamma/\nu - z}$ at the above point satisfies the ex-

pected 3D XY universality class with $\gamma \approx 1.32$, $\nu \approx 0.67$, and $z = 1$ [13].

A better insight into the nature of the quantum phase transition tuned by the anisotropy is achieved by exactly mapping the spins onto bosons through the Holstein-Primakoff (HP) transformation $S_i^+ = \sqrt{1 - n_i/2}\, b_i$ and $S_i^z = 1 - n_i$ (together with a π rotation of one of the two sublattices), which gives

$$\mathcal{H} = -\sum_{\langle ij \rangle} \frac{J_{ij}}{2}\left[\sqrt{1-\frac{n_i}{2}}\, b_i b_j^\dagger \sqrt{1-\frac{n_j}{2}} + \text{H.c.}\right] + \sum_{\langle ij \rangle} J_{ij}(n_i - 1)(n_j - 1) + \sum_i \epsilon_i [D(n_i - 1)^2 - hJn_i] \tag{2}$$

Here $n_i = b_i^\dagger b_i = 0, 1, 2$ is the dynamically constrained occupation number, and $J_{ij} = J\epsilon_i\epsilon_j$ prevents bosons from hopping onto doped sites and from experiencing repulsion from those sites. It is evident that, apart from the square-root terms in the hopping Hamiltonian, the model of Eq. (2) is a Bose-Hubbard model with soft-core interactions, allowing for up to 2 particles per site. For $h = 0$ the Z_2 symmetry of the spin model translates into a particle-hole symmetry of the bosonic model, which implies that the system is exactly half-filled, $\langle n_i \rangle = 1$. Interestingly, this still holds true in the presence of site dilution, which does not alter the particle-hole symmetry of the Hamiltonian. The anisotropy term D of the spin model translates into an on-site repulsion for the HP bosons, such that the transition driven by increasing D/J can be understood as a superfluid-to-Mott-insulator (SF-MI) transition driven by the ratio between repulsion and hopping [1]. The observed $(d + 1) - XY$ universality class is expected for a commensurately filled lattice [1].

Introducing site dilution into the Hamiltonian equation (1) hence offers the intriguing opportunity of studying disorder effects on a SF-MI transition, both at commensurate ($h = 0$) and at incommensurate fillings ($h \neq 0$) [14]. In this Letter, we mainly focus on the $h = 0$ regime, and we will discuss the case of $h \neq 0$ in a forthcoming publication [16]. Moreover, the model of Eq. (1) lends itself to studying the behavior of antiferromagnetic order around the percolation threshold of the site-diluted square-lattice $p = p^* = 0.407\,253\ldots$ [17] under continuous tuning of the strength of quantum fluctuations. In the limit $D = 0$, it has been demonstrated numerically that the $S = 1$ SLHAF on a site-diluted lattice retains LRO up to the percolation threshold [18]. The system at finite p and finite D interpolates then between the $3D - XY$ transition at $p = 0$ and the percolation transition at $D = 0$.

In Fig. 1, we show the complete phase diagram of the system in the $p - D$ plane at $T = 0$ and $h = 0$. The boundary lines are estimated as above, using the criterion $\xi^{xx(yy)} \sim L$. This phase diagram indicates that the interplay between disorder and quantum fluctuations leads to a dramatic departure both from the SF-MI transition at $p = 0$ and from the percolation driven transition at $D = 0$. Let us start from the limit $D = 0$, and focus on the behavior of the system at the percolation threshold $p = p^*$. Here we clearly observe that, upon increasing the ratio D/J, the transition line from magnetic LRO to disorder passes through a multicritical point at $(D/J)^* = 2.3(2)$, beyond which it departs from the percolation threshold $p = p^*$ and bends toward systematically smaller p values for larger D/J. Starting from the opposite limit $p = 0$, we observe that an infinitesimal amount of disorder leads to a shift to lower values of the critical anisotropy $(D/J)_c$ that causes the destruction of LRO. Putting together these two pieces of information, we can conclude that for any finite disorder concentration p there is a nontrivial disorder-dependent value of the anisotropy $(D/J)_{c,p} < (D/J)_{c,0}$ at which nonlinear quantum fluctuations, enhanced by the reduced connectivity of the diluted lattice, are able to destroy the LRO.

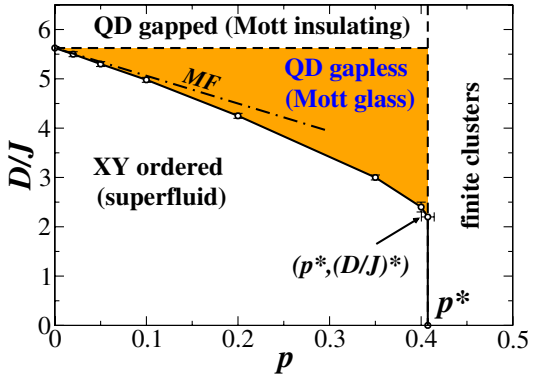

FIG. 1 (color online). Phase diagram of the site-diluted $S = 1$ SLHAF. The magnetic phases (and the corresponding bosonic ones) are indicated. QD = quantum disordered. The mean-field (MF) line corresponds to the low-dilution limit $(D/J)_{c,p} = (D/J)_{c,0}(1 - p)$.

The interplay between disorder and quantum fluctuations can be quantitatively understood in the relevant limit of $p = p^*$. At this point, the percolating nature of the largest cluster in the system crucially depends on 1D links connecting quasi-2D islands with higher local connectivity (blobs) [19]. For D/J well below $(D/J)_{c,0}$, the quasi-2D blobs have a well-defined staggered moment (see Fig. 2) which fluctuates with a characteristic frequency given by the finite-size gap of the blob if the blobs are disconnected. When the blobs are connected by links to form the percolating cluster, their local staggered moments are either long-range correlated or not, depending on the decay of correlations along the 1D links [20]. If the characteristic length l_0 of the 1D links is shorter than the correlation length ξ_{1D} for the $S = 1$ anisotropic Heisenberg model of Eq. (1) on a chain, the links are able to establish 2D LRO in the system. Otherwise the blobs are uncorrelated, and the system enters a quantum-disordered phase. An earlier detailed study [19] of the geometry of the percolating cluster reports an average link length $\langle l_0 \rangle \approx 2.7$ for site percolation on the square lattice. We have studied the correlation

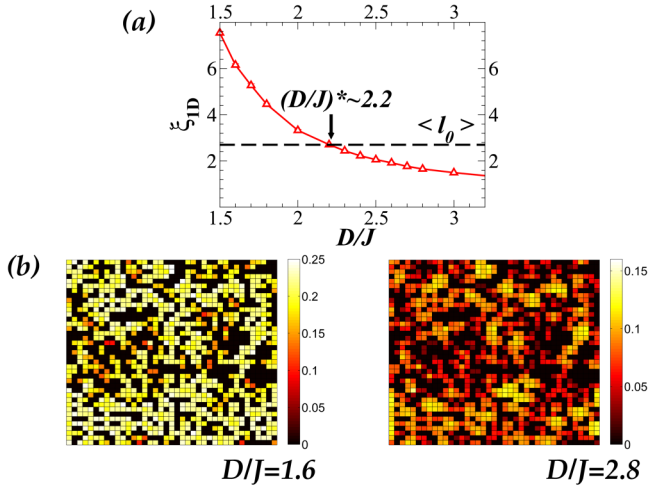

FIG. 2 (color online). (a) Ground-state correlation length of the $S = 1$ anisotropic Heisenberg antiferromagnet on the chain as a function of the anisotropy. (b) Local susceptibility $\chi_{\mathrm{loc},i} = \int_0^\beta d\tau \langle S_i^z(0) S_i^z(\tau)\rangle$ on a 40×40 lattice at the percolation threshold $p = p^*$, with anisotropy below and above $(D/J)^* = 2.2$. $\chi_{\mathrm{loc},i} = 0$ corresponds to empty sites.

properties of the anisotropic Hamiltonian Eq. (1) on a chain with the SSE-QMC method, systematically estimating the one-dimensional equal-time correlation length ξ_{1D} as a function of the anisotropy D/J. According to the above argument, the loss of LRO on the percolating cluster should occur for $(D/J)^*$ such that $\xi_{1D}[(D/J)^*] \approx \langle l_0 \rangle$. This criterion leads to the estimate $(D/J)^* \approx 2.2$ (Fig. 2), which is in excellent agreement with the location of the multicritical point estimated by QMC calculations, as shown in Fig. 1. Hence we quantitatively understand the deviation of the magnetic transition from percolation in terms of a critical enhancement of local quantum fluctuations on the weak links of the percolating cluster. This is also depicted in Fig. 2(b), representing the local susceptibility on a lattice at the percolation threshold: for $(D/J) < (D/J)^*$ the local response of the system is nearly homogeneous, while for $(D/J) > (D/J)^*$ the 1D links show a strongly suppressed response due to freezing in a nearly $m_S = 0$ state.

When $p \lesssim p^*$, the links joining quasi-2D islands acquire a higher connectivity, evolving from single chains to decorated chains or ladders. At the critical value $(D/J)_{c,p}$ for $p \lesssim p^*$ the correlation length on the quasi-1D links becomes comparable with their average linear size, analogous to what happens at $p = p^*$. For weak dilution $p \ll p^*$, on the other hand, the disordered lattice can be approximately represented as a homogeneous lattice with reduced effective coordination $z_{\mathrm{eff}} = z(1 - p)$, corresponding to the Hamiltonian of Eq. (1) in which the random variables are substituted by their average $\langle \epsilon_i \rangle = 1 - p$. In this case the critical value $(D/J)_{c,p}$ should be linearly shifted by the disorder with respect to the $p = 0$ case, $(D/J)_{c,p} \approx (1 - p)(D/J)_{c,0}$, which is in very good agreement with our data for $p \lesssim 0.05$ (see Fig. 1).

The deviation of the magnetic transition from the percolation transition opens a novel quantum-disordered phase that exists on percolated lattices with connectivities arbitrarily close to the clean system as $D/J \to (D/J)_{c,0}$. This phase has remarkable unconventional properties, as evidenced by its response to a uniform magnetic field. In Fig. 3(a) we show the low-temperature uniform susceptibility $\chi_u(T \to 0)$ for representative points in the disordered phase, contrasted with the behavior in the gapped phase at $p = 0$. It is observed that $\chi_u(T \to 0)$ tends to 0, as in a phase with a singlet ground state, so that, in the bosonic language, this phase is incompressible at $T = 0$. Yet the specific temperature dependence of χ_u appears to follow an unconventional exponential law $\chi_u(T) \sim \exp[-\sqrt{\Delta/T}]$ for more than two decades of the QMC data. This is clearly different from the conventional behavior ($\sim \exp[-\Delta/T]$) expected in the presence of a finite singlet-to-triplet gap Δ. To probe this gap directly, we study the response of the system to small but finite uniform fields. Figure 3(b) shows the low-field magnetization curve of the system in the quantum-disordered phase. This is strong numerical evidence that the magnetization is finite for arbitrarily small fields, although it grows slowly with h, following the

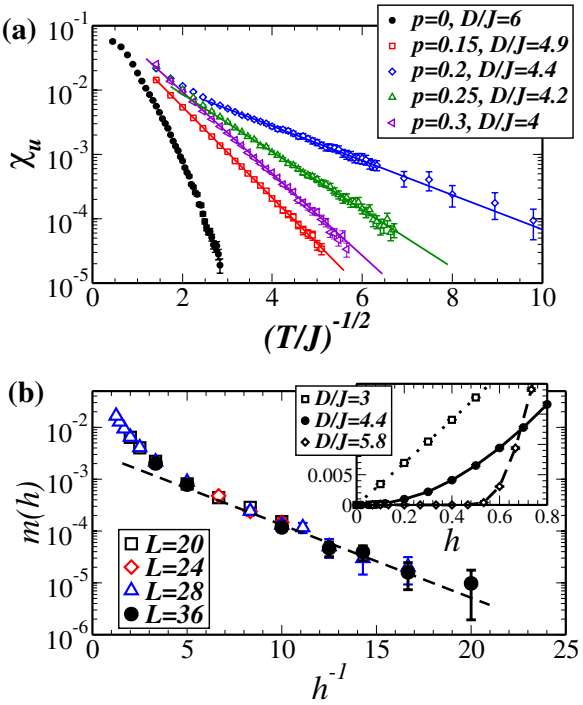

FIG. 3 (color online). (a) Uniform susceptibility for various points in the Mott-glass phase, contrasted with the case of a point in the gapped phase for $p = 0$ (system size: $L = 36$). (b) Uniform magnetization upon application of a weak field in the Mott-glass phase at $p = 0.2$, $D/J = 4.4$. In the inset: different magnetization curves starting from the XY ordered phase ($D/J = 3$), Mott-glass phase ($D/J = 4.4$), and QD gapped phase ($D/J = 5.8$). For $D/J = 3$ we have plotted $m/2$, and, for $D/J = 5.8$, h is rescaled by a factor of 3. Here $L = 28$ and $p = 0.2$.

unconventional exponential behavior $m(h) \sim \exp[-A/h]$ over approximately two decades of the QMC data. The magnetization clearly shows that the singlet-to-triplet gap vanishes even at $h = 0$, and this observation is still compatible with a vanishing uniform susceptibility for $h = 0$ because $\chi_u = dm/dh \sim h^{-2} \exp[-A/h] \to 0$ for $h \to 0$, but it is nonzero for any finite h. The magnetization curve in the novel disordered phase contrasts with the linear behavior of $m(h)$ when starting from the XY ordered phase and with the gapped behavior when starting from the Mott insulating phase (inset of Fig. 3). The exponential T dependence of χ_u and h dependence of m can be fully captured by a simple effective model in which the response to a weak field is given by independent clean regions of the percolating cluster, whose exponentially rare statistics reflects itself in the exponentially small response functions [15,16]. The gapless nature of the spectrum is a property of the entire novel disordered phase for $(D/J)_{c,p} \leq D/J \leq (D/J)_{c,0}$. In fact, for any p one can always find an arbitrarily large (albeit rare) clean region which locally approximates the behavior of the clean square lattice and which has consequently local excitations at arbitrarily low energy as long as $D/J \leq (D/J)_{c,0}$.

To summarize, we have seen evidence of an incompressible, yet gapless phase of the system. The amorphous nature of this disordered phase and its rich low-energy dynamics make it akin to glassy phases, and at the same time its incompressibility, along with the commensurate filling of the lattice, bears a strong resemblance to a Mott insulating phase. Hence it can be called a Mott glass, a name that has been used in the recent literature for dirty-boson models at commensurate filling [3] in analogy with a phase of disordered fermions having the same thermodynamic and spectral signatures [2]. Moreover, for any finite h the ground-state phase acquires a finite susceptibility, namely, the Mott-glass phase is driven into a Bose glass [1] which is disordered, gapless, and compressible [16].

The occurrence of a bosonic Mott-glass phase in the site-diluted $S = 1$ antiferromagnet with single-ion anisotropy is well understood within the bosonic mapping of Eq. (2) [3]. In particular we wish to stress that the Hamiltonian of Eq. (1), both in its clean and disordered form, is a reliable description of the magnetic degrees of freedom in strongly anisotropic $S = 1$ antiferromagnetic insulators with and without doping. In fact, the gapped quantum-disordered state induced by the large anisotropy term is realized in a variety of Ni-based compounds, such as $Ni(C_5H_5NO)_6 \times (NO_3)_2$ [21], $[Ni(C_5H_5NO)_6](ClO_4)_2$ [22], and the more recently investigated $NiCl_2 \cdot 4SC(NH_2)_2$ [23]. These systems have in general a three-dimensional magnetic lattice, but the results we have shown for the 2D case can be generalized straightforwardly to 3D. Moreover one can imagine tuning the D/J ratio experimentally by applying hydrostatic pressure to the crystals.

The Z_2 symmetry of the magnetic Hamiltonian in zero field for all these systems translates into particle-hole symmetry and commensurate (unit) filling of the soft-core bosons on any lattice site, regardless the geometry of the lattice itself, namely, also on random percolating clusters. This ingredient, which is crucial for the existence of the Mott glass, appears hard to achieve in systems of real bosons, as, e.g., in bosons in optical lattices, where the introduction of disorder will invariably change the particle occupation near lattice defects.

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[1] M. P. A. Fisher *et al.*, Phys. Rev. B **40**, 546 (1989).
[2] E. Orignac *et al.*, Phys. Rev. Lett. **83**, 2378 (1999); T. Giamarchi, P. Le Doussal, and E. Orignac, Phys. Rev. B **64**, 245119 (2001).
[3] N. Prokof'ev and B. Svistunov, Phys. Rev. Lett. **92**, 015703 (2004); E. Altman *et al.*, *ibid.* **93**, 150402 (2004); K. G. Balabanyan *et al.*, *ibid.* **95**, 055701 (2005).
[4] G. A. Csáthy *et al.*, Phys. Rev. Lett. **91**, 235301 (2003).
[5] G. Blatter *et al.*, Rev. Mod. Phys. **66**, 1125 (1994).
[6] J. Lye *et al.*, Phys. Rev. Lett. **95**, 070401 (2005); D. Clément *et al.*, *ibid.* **95**, 170409 (2005); C. Fort *et al.*, *ibid.* **95**, 170410 (2005); T. Schulte *et al.*, *ibid.* **95**, 170411 (2005).
[7] O. P. Vajk *et al.*, Science **295**, 1691 (2002).
[8] A. W. Sandvik, Phys. Rev. B **66**, 024418 (2002).
[9] O. P. Vajk and M. Greven, Phys. Rev. Lett. **89**, 177202 (2002); A. W. Sandvik, *ibid.* **89**, 177201 (2002).
[10] O. F. Syljuåsen, Phys. Rev. E **67**, 046701 (2003).
[11] E. J. Neves and J. F. Perez, Phys. Lett. A **114**, 331 (1986).
[12] H.-T. Wang and Y. Wang, Phys. Rev. B **71**, 104429 (2005).
[13] A. Pelissetto and E. Vicari, Phys. Rep. **368**, 549 (2002).
[14] At variance with other spin-gap systems based, e.g., on weakly coupled dimers or Haldane chains (see Ref. [15] and references therein), doping does not produce $S = \frac{1}{2}$ moments close to the impurity sites, so that the bosonic mapping of Eq. (2) is exact for any doping.
[15] T. Roscilde, Phys. Rev. B **74**, 144418 (2006).
[16] T. Roscilde and S. Haas (to be published).
[17] M. E. J. Newman and R. M. Ziff, Phys. Rev. Lett. **85**, 4104 (2000).
[18] K. Kato *et al.*, Phys. Rev. Lett. **84**, 4204 (2000).
[19] A. Coniglio, J. Phys. A **15**, 3829 (1982); H. J. Herrmann and H. E. Stanley, Phys. Rev. Lett. **53**, 1121 (1984).
[20] N. Bray-Ali *et al.*, Phys. Rev. B **73**, 064417 (2006).
[21] R. L. Carlin *et al.*, J. Phys. C **12**, 293 (1979).
[22] K. M. Diederix *et al.*, Phys. Lett. A **60**, 247 (1977).
[23] A. Paduan-Filho *et al.*, Phys. Rev. B **69**, 020405 (2004); V. Zapf *et al.*, Phys. Rev. Lett. **96**, 077204 (2006).