Universal phase diagram of disordered bosons from a doped quantum magnet

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Abstract – A quantum particle cannot in general diffuse through a disordered medium because of its wavelike nature, but interacting particles can escape localization by collectively percolating through the system. For bosonic particles this phenomenon corresponds to a quantum transition from a localized insulator phase —the Bose glass— to a superfluid phase, in which particles condense into an extended state. Here, we construct a universal phase diagram of disordered bosons in doped quantum magnets for which bosonic quasiparticles are represented by magnetized states (spin triplets) of the quantum spins, condensing into a magnetically ordered state. The appearance of a Bose glass leads to strong measurable signatures in the onset of superfluidity of the spin-triplet gas, exhibiting a complex crossover from low-temperature quantum percolation to a conventional condensation transition at intermediate temperatures.

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In continuous quantum phase transitions [1] quantum fluctuations drive a complete change of symmetry in the state of the system via the development of long-wavelength modes at arbitrarily low energies. This occurs in the case of strongly interacting bosons on a lattice, modeling widely different physical systems such as cold atoms loaded in optical lattices [2], Cooper pairs in Josephson-junction arrays [3], or magnetic quasiparticles in gapped quantum magnets [4]. In the presence of strong repulsion among the particles, a commensurate filling of the lattice stabilizes a gapped Mott insulating phase, which has all the symmetries of the original Hamiltonian. In contrast, doping the system away from commensurate filling drives it into a Bose-condensed phase [5], characterized by superfluidity and a gapless phonon-like mode.

The presence of disorder in the system can deeply affect its quantum critical behavior. In fact, disorder can localize the long-wavelength modes developed at the quantum phase transition of the clean system, and thus introduce a novel phase, with gapless localized excitations but in the absence of any long-range order. Indeed, it has been shown that lattice disorder for interacting bosons can localize the gas of doped particles/holes, hindering their condensation and thus giving rise to a new insulating gapless phase with incommensurate filling, known as a Bose glass [5,6]. Moreover, when the density of doped particles/holes exceeds a critical threshold, their mutual interactions destabilize energetically the localized state in favor of a delocalized superfluid state. It is clear that the nature of this transition is completely different from that of the Mott-insulator/superfluid transition in the absence of randomness.

Despite its extreme richness and generality, the phase diagram of so-called dirty-boson systems has so far been extremely elusive, both to experiments, due to the difficulty of accessing the quantum critical regime in a controlled disordered environment [7–9]; and to theory, due to the absence of well-controlled approximations in the presence of strong interactions and strong disorder [10]. Here we propose the use of doped quantum magnets to fully investigate the physics of dirty bosons, and we perform numerically exact simulations on a realistic model Hamiltonian to construct the universal phase diagram of the system. Our model of interest

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is the magnetic Hamiltonian of NiCl$_2$·4SC(NH$_2$)$_2$ (DTN) [11,12], which consists of coupled $S=1$ chains with strong single-ion anisotropy:

$$
\mathcal{H} = J_c \sum_{\langle ij \rangle_c} \epsilon_i \epsilon_j S_i \cdot S_j + J_{ab} \sum_{\langle \langle lm \rangle_{ab} \rangle} \epsilon_i \epsilon_m S_i \cdot S_m + D \sum_i \epsilon_i (S_i^z)^2 - g \mu_B H \sum_i \epsilon_i S_i^z,
$$

where $J_c > 0$ is the dominant antiferromagnetic coupling for bonds $\langle ij \rangle_c$ along the crystallographic $c$-axis, $J_{ab} > 0$ is the coupling for bonds $(lm)_{ab}$ in the transverse $a$, $b$ directions of the cubic lattice$^1$ and $D > 0$ is the single-ion anisotropy. We adopt the experimental parameters [11,12] $J_c = 2.2$ K, $J_{ab} = 0.18$ K, $D = 8.9$ K. In the following we will use reduced temperatures $t = k_B T / J_c$ and reduced magnetic fields $h = g \mu_B H / J_c$. Moreover we introduce randomness in the system in the form of site dilution via the variables $\epsilon_i$, taking values 0 and 1 with probability $x$ and $1-x$, respectively, where $x$ is the dilution concentration. Site dilution corresponds experimentally to Mg$^{2+}$ or Cd$^{2+}$ doping of the Ni$^{2+}$ ions.

A standard Holstein-Primakoff spin-to-boson mapping [14] allows to recast the above Hamiltonian in the form of bosonic operators $b_i$, $b_i^\dagger$, with the local Hilbert space limited to the Fock states $|n \rangle = |0 \rangle$, $|1 \rangle$ and $|2 \rangle$ corresponding to the states $|m_S \rangle = |1 \rangle$, $|0 \rangle$ and $|1 \rangle$ of a $S=1$ spin. The resulting bosonic Hamiltonian has the form

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

Upon this transformation the $J$ couplings take the role of nearest-neighbor hopping amplitudes and repulsion potentials, while the single-ion anisotropy $D$ plays the role of on-site repulsion for the bosons, and finally the magnetic field $h$ acts as the chemical potential. The bosonic Hamiltonian has indeed the structure of a Bose-Hubbard model on a site-diluted lattice, with a constraint on particle number occupation. Compared to the standard Bose-Hubbard model, the extra $\sqrt{1 - \frac{n_i}{2}}$ terms, which correct the values of the non-zero matrix elements of the $b$, $b^\dagger$ operators to give the appropriate ones for the spin operators $S^+$, $S^z$, do not alter the fundamental $U(1)$ symmetry of the Bose-Hubbard model under the global operator rotation $b \rightarrow e^{i \phi} b$. Therefore the universal physics of the Hubbard model is completely captured by the spin model. For instance, in the clean case $x = 0$ the above model describes correctly the Mott-insulator/superfluid quantum phase transition at commensurate filling (corresponding to $h = 0$) upon changing the ratio of anisotropy (repulsion) to hopping (exchange coupling) [15].

Due to the large anisotropy $D$, the ground state of the system in zero field is well approximated by a collection of singlets $|m_S = 0 \rangle$, corresponding to a gapped Mott insulator of bosons with $n = 1$ particle per site. In the case of the pure system [11], applying a magnetic field above a critical value $h_0(1) = 1.65(1)$ allows to close the gap in the spectrum and to increase the particle density above unit filling, corresponding to the appearance of a finite magnetization along the field direction. Upon increasing the filling, the system develops a superfluid and condensate fraction, which translates into the spin stiffness of the magnetic system and into a peak at $q = (\pi, \pi, \pi)$ in the transverse static structure factor $S_\perp(q) = \frac{1}{(1 - x)^3} \sum_{ij} \exp[iq \cdot (r_i - r_j)] \langle \sum_i S_i^x S_j^x \rangle$, (3) respectively. The condensation order parameter is directly related to the spontaneous magnetization appearing in the $xy$-plane [16], $\langle b_i \rangle \approx \langle S_i^x \rangle / \sqrt{\langle S_i^z \rangle}$. Condensation (in the form of magnetic order) survives at finite temperature up to a critical temperature $t_c$, which scales with the applied field as $t_c \sim (h - h_c(0))^{\phi_0}$, where $\phi_0 = 2/3$ as correctly predicted by mean-field theory for the condensation of a diluted Bose gas [17]. The experimental verification of this prediction [16] is considered as the strongest evidence for magnetic Bose-Einstein condensation (BEC).

Increasing the field even further reduces the state of the system to that of a dilute gas of singly occupied sites (excess triplet holes), associated with $|m_S = 0 \rangle$ spin states. A high field eventually leads to full polarization of the spins at the critical value $h_c(0) = 8.69$, corresponding to a second Mott insulating state of triplets (namely the vacuum of triplet holes), and to a vanishing of the critical temperature which obey the same scaling law, $t_c \sim (h_c(0) - h)^{\phi_0}$.

In the presence of site dilution the phase diagram changes substantially. We have investigated this phase diagram via numerically exact quantum Monte Carlo technique [18] on lattice sizes as large as $16^3$, and averaging all results over $\sim 300$ disorder realizations. The critical temperatures, critical fields and critical exponents have been estimated via a careful finite-size scaling analysis of the spin structure factor, correlation length and spin stiffness [19]. The results of this extensive quantum Monte Carlo study for a dilution of $x = 15\%$ are shown in fig. 1. The most important effect of disorder on the phase diagram is that of shifting the critical fields for condensation and shrinking the condensation region with respect to the clean system, $h_{c1} \approx 1.94(1) > h_{c1}(0)$ and $h_{c2} \approx 8.52(2) < h_{c2}(0)$. This effect separates the onset of condensation from the closing of the spin gap, which still occurs at $h_{c1}(0)$ and $h_{c2}(0)$. In fact the spin gap of the doped system is the same as for the clean system.
given that the minimum spin gap in the doped system is associated with local excitations in rare regions that locally approximate the clean system, and whose size can be arbitrarily large. Hence disorder “splits” an ordinary quantum critical point into two, i.e. a spectral transition point for the closing of the gap, and a true critical point for the onset of order. Inbetween these two critical points an extended Bose-glass phase opens up, characterized by the joint absence of a spin gap and of superfluidity. In the magnetic language, this represents a gapless quantum paramagnetic phase. In the clean case of the Hamiltonian given in eq. (1) the $T=0$ quantum-critical point is Gaussian, with mean-field critical exponents and dynamical critical exponent $z=2$ [5]. In particular, the spin gap closes as $\delta \sim |h-h_{c1}^{(0)}|^\nu = |h-h_{c1}^{(0)}|$, where $\nu = 1/2$ is the mean-field correlation length exponent. In the disordered system the gap still closes in a mean-field–like fashion, but disorder completely discards the mean-field picture for the true quantum-critical points at $h_{c1}$ and $h_{c2}$, which obey the exponents of a new universality class pertinent to the superfluid–to–Bose-glass transition. The prediction of ref. [5] for the dynamical critical exponent, $z = d = 3$, is well verified by our numerical data. Moreover, we determine the correlation length exponents as $\nu = 0.7(1)$, consistent with ref. [20], and the order-parameter exponent as $\beta = 0.9(1)$ [19].

The Bose-glass state is characterized by Anderson localization [21] of the excess spin triplets (triplet holes) injected into the Mott insulating state; this leads to a magnetization profile characterized by magnetized “puddles”, located in the rare regions devoid of lattice vacancies, as shown by direct inspection of the numerical data (see fig. 2). To characterize the density profile of the localized spin triplets (triplet holes) we investigate the behavior of the magnetic participation ratio (mPR)

$$mPR = \frac{1}{(1-x)L^3} \left< \frac{\sum_i (S_i^z)^2}{\sum_i (S_i^z)^2} \right>_{\text{disorder}}$$

representing the effective fraction of the system which is magnetized, Here $\left< \ldots \right>_{\text{disorder}}$ represents the disorder average. Figure 3 show the mPR as a function of temperature for various magnetic fields. A percolating magnetization profile is characterized by a participation ratio which exceeds a fraction $p^*(x) = p_{sc} / (1 - x) = 0.366$, corresponding to the percolation threshold for the simple cubic lattice $p_{sc} = 0.312$ [22] renormalized by the

![Fig. 1](https://example.com/fig1.png)

Fig. 1: (Colour on-line) Phase diagram of coupled anisotropic $S=1$ chains in a magnetic field with 15% dilution. We plot the critical temperature $t_c$ for magnetic BEC, and the spin gap $\delta = \Delta / J_c = h_{c1}^{(0)} - h$ (for $h \leq h_{c1}^{(0)}$) and $\delta = h - h_{c2}^{(0)}$ (for $h \geq h_{c2}^{(0)}$), as a function of the applied magnetic field for zero doping ($x = 0$) and for 15% doping ($x = 0.15$). The spin gap is independent of the doping concentration. The inset shows a zoom on the region close to the lower critical field $h_{c1}$, marking the appearance of the Bose glass.

![Fig. 2](https://example.com/fig2.png)

Fig. 2: (Colour on-line) Local magnetization profile (corresponding to the excess spin-triplet bosons) of coupled $S=1$ chains, eq. (1) on a $16^3$ lattice with 15% dilution, obtained via quantum Monte Carlo simulations. The local magnetization is indicated by the size of the dots. Four representative points in the $(h, t)$-plane are shown $(h, t)$ coordinates are indicated below each figure), for the following bosonic phases: 1) a Bose glass with localized incoherent excess spin quasiparticles; 2) an inhomogeneous BEC phase, with long-range coherence due to weak links between localizes states; 3) a normal Bose gas, with incoherent spin quasiparticles which are spread over a percolating region; 4) a BEC phase, with long-range coherence of spin quasiparticles homogeneously spread over a percolating region. In addition to these phases, the Mott insulator (not shown), is characterized by the absence of excess spin quasiparticles. Among the phenomena connecting the various phases, quantum percolation and condensation are sharp transitions, while thermal percolation is a broad crossover. The pictures shown here refer to the regime $h \sim h_{c1}$. An analogous succession of phases, mirrored along the magnetic field axis, is observed for $h \sim h_{c2}$.
dilution. Below this threshold, the magnetization profile is composed of spatially disconnected puddles, which are phase-incoherent, resulting in the absence of long-range order of the transverse spin components. This remains true for a significant portion of the finite-temperature region above the zero-temperature Bose glass, despite the strong thermal activation of spin triplets/holes. We term this temperature region as thermal Bose glass. In such a region the magnetization $m(t, h) = \sum_i \langle S^z_i \rangle / [(1 - x)L^3]$ can be well reproduced by a model of disconnected clusters with sizes obeying the statistics of rare clean regions in a site-diluted cubic lattice [19].

Increasing the temperature above the characteristic value $t_{\text{mPR}=p^*}$ at which the magnetic participation ratio reaches the condition $m_{\text{PR}} = p^*$, one enters into a thermal percolation crossover region, marking the upper bound of the thermal Bose glass. Above this region the spin triplets/holes, albeit incoherent, are delocalized over the entire lattice. Thermal percolation is marked by a substantial change in the statistics of the local magnetization $\langle S^z_i \rangle$, corresponding to the local excess population of triplets/holes. Indeed one can easily show that $m_{\text{PR}} = [1 + (\sigma/m)^2]^{-1}$, where $m$ is the average of the $\langle S^z_i \rangle$ distribution (namely the total magnetization) and $\sigma$ its standard deviation. In particular the link between $m_{\text{PR}}$ and the local magnetization distribution suggests a second criterion for thermal percolation. Considering the ratio $R = m/\sigma$, we take $R = 1$ (corresponding to $m_{\text{PR}} = 1/2$) as the crossover value above which the fluctuations are dominated by the mean. This crossover marks the passage from a highly inhomogeneous magnetization profile to a homogeneous one. Hence in the following we adopt the two criteria ($m_{\text{PR}} = p^*$ and $m_{\text{PR}} = 1/2$) to mark the thermal percolation crossover. Most importantly, in the absence of spontaneous ordering transverse to the field, the $\langle S^z_i \rangle$ distribution is probed by the lineshape of nuclear magnetic resonance [23], which means that thermal percolation can be experimentally detected.

Given the above analysis, it is to be expected that the transition to triplet/hole condensation, induced by the applied field, will be very different when coming from the thermal Bose glass or when coming from the normal Bose gas. In the former case, indeed, the triplet/hole gas undergoes percolation of quantum coherence, and hence condensation, via tunneling of the triplet/hole bosons between the magnetization puddles; this clearly represents a finite-temperature quantum percolation phenomenon. In the latter case, in contrast, condensation occurs on the magnetized backbone of the diluted lattice, akin to field-induced condensation in a clean system.

As shown in fig. 4, this crossover from low-temperature quantum percolation to more conventional condensation on a random network has a striking signature in the central feature of magnetic BEC, namely the scaling of the critical temperature close to the lower critical field. The Bose-glass nature of the disordered phase at low temperature leads to the breakdown of the scaling prediction stemming from mean-field theory of BEC in a dilute Bose gas. Numerically we find that the onset of $T_c$ follows $T_c \propto |h - h_{c1}|^{\phi}$ with $\phi = 1.16(5)$, in violation of the mean-field prediction [19]. But, even more remarkably, above a characteristic temperature range the $t_{c}(h)$ curve shows a clear crossover to a different curvature, consistent this time with the mean-field exponent $\phi_0 = 2/3$. We observe that this crossover corresponds to the dividing region between the two different condensation regimes, namely from a thermal Bose glass and from a normal Bose gas. Indeed, above the crossover region spatial percolation of the magnetization profile occurs already.
before condensation, and hence condensation acquires a conventional character, while disorder only plays the role of effectively renormalizing the Hamiltonian parameters of the clean system. It is important to stress that the disorder-induced crossover in the scaling behavior is not accompanied by a change in the universality class of the transition: indeed, according to the Harris criterion [24] disorder is not pertinent for the 3D XY condensation transition of the clean system, for which $\nu \approx 0.67 \geq 2/D = 2/3$ [25].

In conclusion, we have shown that a realistic disordered quantum spin model, corresponding to the magnetic Hamiltonian of DTN [11] under non-magnetic doping, can elucidate the universal features of the physics of strongly correlated bosons on a disordered lattice in the grand-canonical ensemble. A variety of measurement tools, such as magnetometry and NMR, can reveal the central features of Anderson localization of triplet bosons, leading to inhomogeneous magnetization profiles, and to thermal and/or quantum percolation thereof. These results clearly demonstrate the potential of model quantum magnets as a valuable testbed for theories of complex Bose systems with strong interactions and disorder, and more generally for theories of the fundamental interplay between quantum critical fluctuations and geometric randomness.

In the specific context of disordered bosons, quantum magnets have distinct experimental features which make them complementary or even competitive with respect to atomic Bose fluids. Indeed, most promising experimental steps toward the controlled realization of the Bose-glass phase are being undertaken in the context of ultracold bosons trapped in disordered optical lattices [26], which are nonetheless strongly affected by finite-size effects and cannot currently access the details of the quantum critical regime; and in the context of $^3$He in nanoporous media [27,28], in which the instability towards solidification currently prevents a clear observation of a Bose-glass-to-superfluid transition at low temperatures. Quantum magnets such as DTN have the advantage of fully exhibiting quantum critical properties at experimentally accessible temperatures [11,29], and of exhibiting a large Bose-glass regime in parameter space under realistic doping, as shown in this paper. Moreover they offer a complementary view to atomic systems, because they realize a strongly interacting Bose fluid in the grand-canonical ensemble (while atomic systems are typically at constant particle number). Finally, other systems have been recently demonstrated to host Bose fluids of quasiparticles under continuous pumping, such as magnons in $^3$He [30] or exciton-polaritons in semiconductor quantum wells [31]; these Bose fluids can reach the condensation regime out of equilibrium, and the investigation of disorder effects in that context is certainly intriguing [32], but its relationship to equilibrium “dirty-boson” systems remains to be clarified. In this respect, another advantage of the Bose fluid realized in antiferromagnetic insulators is that it is composed of gapless excitations whose density at equilibrium is continuously controlled by the magnetic field, without any need of pumping energy into the system.

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