Anomalous percolation and quantum criticality in diluted rare-earth nickelates

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A microscopic model for the diluted spin-mixed compounds (R_xY_{1-x})_2BaNiO_5 (R=magnetic rare-earth) is studied using Quantum Monte Carlo (QMC). The ordering temperature is shown to be a universal function of the impurity concentration x and the intrinsic Ni-chain correlation length. An effective model for the critical modes is derived. The possibility of a quantum critical point driven by the rare-earth-concentration and the existence of a Griffiths phase in the high dilution limit is investigated. Several possible experimental approaches to verify the results are put forward.

An increasing number of low-dimensional quantum magnets are being realized in materials artificially modified at nanometer scales. The use of layered structures, dilution and/or combination of magnetic species with different magnetic moment establish complex geometries that enhance the role of quantum correlations. The resulting competing ground states, quantum critical points, and coexistence of classical and quantum features are of great current interest in condensed matter and interdisciplinary physics. The family of mixed-spin quantum antiferromagnets (R_xY_{1-x})_2BaNiO_5 (R = magnetic rare earth) provides a unique experimental basis to discuss these issues. The x = 0 compound is an excellent realization of the 1-dimensional (1D) S = 1 Heisenberg model, thanks to a linear-chain arrangement of the magnetic Ni^{2+} ions. Neutron scattering experiments confirmed a spin liquid behavior, and revealed a Haldane gap of about 10 meV in the magnetic excitation spectrum. Inter-chain interactions, negligible for x = 0, can be controlled by substituting the intercalated non-magnetic Y^{3+} by magnetic rare earths. For x = 1 the system orders antiferromagnetically at T_N =10-50 K, depending on R. The spin dynamics is characterized by a coexistence and frequency-separation of semi-classical (spin waves) and quantum (Haldane-gap modes) excitations. By now, this phenomenon has been extensively studied experimentally, analytically, and numerically.

The focus of the present Letter, however, is the role of randomness and disorder for 0 < x < 1. Previous studies were aimed at understanding the large-x limit. Due to a finite intrinsic correlation length in Haldane spin chains (∼7 lattice units), for x ≈ 1 disorder is effectively averaged out. The problem is then merely a rescaled version of x = 1 case. For x ≤ 1/ξ_{ch} the mean distance between magnetic rare earth centers becomes comparable to ξ. To date, very little is known of this regime, beyond the fact that it should represent qualitatively new physics. Below we develop a numerical approach to this interesting problem. Our calculations predict an unusual percolation mechanism for (R_xY_{1-x})_2BaNiO_5 and related systems. We show that the ordering temperature is a universal function of xξ_{ch}. In addition, we derive an effective model for the critical modes, discussing whether the system is ordered down to zero concentration x or can support a quantum critical point.

A microscopic model for the (R_xY_{1-x})_2BaNiO_5 was proposed in Refs. [4, 7], and studied in detail for x = 1 [4]. The corresponding spin Hamiltonian includes a part that describes the individual Haldane spin chains and an additional inter-chain interaction term. The former contains no disorder and is independent of the doping level x. In contrast, the interaction term involves spin operators for the intercalated rare earth ions, and directly reflects the effect of dilution. In the present work we will consider two variations of this model:

H_1 = H_{c-c} + J \sum_{ij} S_{i,2j} S_{i+1,2j} \quad (1)

H_2 = H_{c-c} - J \sum_{ij} S_{i,2j}^2 S_{i+1,2j}^2 + \Gamma \sum_{ij} S_{i,2j}^4 \quad (2)

with H_{c-c} = J_c \sum_{ij} \epsilon_{i,2j+1} (S_{i,2j}^+ S_{i,2j+2}^- + S_{i,2j}^- S_{i,2j+2}^+)

Both Hamiltonians have a common inter-chain interaction term H_{c-c}. ε is a random variable representing the stochastic occupation of the inter-chain sites with moments s_{i,2j+1} corresponding to the rare earths with probability density p(ε) = xδ(ε - 1) + (1-x)δ(ε). As argued in Refs. [4, 7], Kramers rare earth ions in the (R_xY_{1-x})_2BaNiO_5 structure can be modeled by S = 1/2 pseudo-spins with anisotropic (Ising-type) R-Ni exchange interactions J_{ij}.

The two Hamiltonians differ in their description of the gapped spin chains. In model (1) the chains are S=1 Heisenberg systems, as in the real material. This model is thus well suited for calculations of realistic ordering temperatures and direct comparisons with experimental data. However, model (2) allows more flexibility for a conceptual study of the problem. It incorporates quantum Ising spin chains in a transverse field (ITF) with Γ > J (paramagnetic gapped regime). The ITF model is in the same universality class as S = 1 Haldane spin chains, and shares with it many common features.
The ground state in both cases is a spin liquid with exponentially decaying correlations with the same asymptotic power-law corrections. The spectrum is gapped, and the low-lying excitations are long-lived magnon-like state with a dispersion minimum near the 1D AF zone-center. Though the symmetries of ITF and Haldane spin chains are different (the full $SU(2)$ symmetry of the Heisenberg model is reduced to $Z_2$ in the ITF), it matters little in our case. Indeed, in the both complete Hamiltonians $H^\text{ITF}$ and $H^\text{Haldane}$ symmetry is explicitly broken by the Ising nature of $J_c$. The advantage of using the ITF model for our purpose is that it allows to tune the intrinsic in-chain correlation length $\xi = \log \left( \frac{J}{\Delta} \right)$ through the entire range between the free-spin limit $\xi = 0$ to chain-criticality $\xi \to \infty$. This, in turn, enables a study of the fine interplay between the doping level $x$ and the correlation length, and the resulting unique percolation behavior.

Using an efficient Quantum Monte Carlo (QMC) cluster algorithm [1–11], we have first computed the critical temperature for a large range of transverse fields and concentrations in $\mathcal{O}$ using a periodic dilution pattern. In fig. (1a.) we plot, for different concentrations, the critical temperature $T_c$ as a function of $\xi$. A sharp crossover takes places when the temperature is of the order of the gap of the independent chain $T = \Delta_{\text{ch}} = 2(\Gamma - J)$ (dashed line) considered as a separate entity inside the whole system. It is remarkable that a purely quantum one-dimensional length scale is controlling the classical, two-dimensional transition at finite temperature. The key to understand the percolation mechanism in this system is the universal dependence of the critical temperature in the dimensionless variable $x\xi$, which is the ratio between the correlation length in the chain $\xi$ and the average distance between Nd ions $x^{-1}$. For moderate values of the transverse magnetic field different concentrations curves collapse when $x$ approaches the low dilution limit (see fig. (1b.).) This suggests that the effective degrees of freedom that become critical interact with an effective coupling that decreases with the distance between the Nd ions and the correlation length in the chains. To clarify the nature of such degrees of freedom we computed the critical exponents using the conventional finite-size analysis of the order parameter (see fig. 2). We found that the exponents are compatible with the classical 2D Ising model as well for periodic as for disordered dilution (note that the critical behavior of the randomly diluted 2D Ising model differs from the one of the pure 2D Ising model only by logarithmic corrections). This suggests that the building blocks of the effective critical model are classical Ising spins.

To express this ideas in quantitative form we have derived a low energy effective model for both Hamiltonians using a canonical transformation. The central idea underlying this effective model is that the local moments generated by the presence of the $R$ ion between the two chains are coupled due to the stiffness of the finite

FIG. 1: a.) Critical temperature $T_c$ as a function of the correlation length in the independent ITF chain $\xi$, for different concentrations of Nd$^{3+}$. The dashed line indicates the value of the gap of the independent chain ($k_B = \hbar = 1$). This energy scale separates the two completely different behaviors at $T_c = \Delta_{\text{ch}}$ showing that the 1D features of the gapped chains survive even at the critical point. b.) Universal dependence of the critical temperature on the dimensionless variable $x\xi$. At moderate transverse fields there is a sizable range where the $T_c$ for different dilutions collapse to the critical temperature of the 2D anisotropic Ising model (solid line).

FIG. 2: Finite-size analysis of the order parameter in the vicinity of the critical point for $J_c = 0.1$, $\Gamma = 1.25$, $x = 0.5$ and $T_c = 0.13$ in the randomly diluted system. Here the data collapse is achieved with $\beta = \frac{1}{3}$ and $\nu = 1$, the exponents of the 2D Ising model.
chains connecting them. We first study the spectrum of the local hamiltonian of the Ni-R-Ni three-spin block:
\[ h_j = J_0 S_j^z (S_{j-1}^z + S_{j+1}^z) + \Gamma (S_{j-1}^x + S_{j+1}^x). \]
The z-component of the central spin \( S^z \) representing the magnetic moment in the \( R^3 \) ion is a good quantum number. That symmetry breaks the block spectrum in two orthogonal sectors. The ground state is two-fold degenerated and each ground state belongs to distinct sectors. We label these states \(|\uparrow\rangle\) and \(|\downarrow\rangle\) that have opposite values for the expectation value of the total \( z \) component of block spin \( M^z = \pm (S^z + \bar{S}) \) where \( S = \mu (1 + n^2)^{-\frac{1}{2}} \) is the expectation value of the two outer spin magnetizations and \( \mu = J_c/2\Gamma \). There is always a finite gap \( \Delta_B = (\langle J_c/4 \rangle^2 + \Gamma^2)^{1/2} \) to the first excited state (level crossing of local states is forbidden by the symmetry). The local gap does not introduce new restrictions in the validity of our arguments because \( \Delta_B > \Delta_{ch} \) and we are interested only in the regime \( T < \Delta_{ch} \). All the formulas are valid also for the \( S=1 \) system taking \( J_c \rightarrow 2J_c \) and \( \Gamma \rightarrow 0 \)

We consider now the dominant couplings in the low dilution limit: those between two effective local moments on neighboring sites along the direction of the chains \( i, i+1 \). For model \( 2 \) this coupling is exclusively of the Ising type (there are no spin-flip process along the chains). The leading contribution to this coupling is given by the energy necessary to create a domain wall between blocks \( i \) and \( i+1 \),

\[ J_\parallel = \frac{1}{2} \langle \langle i, \psi_{i+1} | h_\parallel^z | \psi_i, \psi_{i+1} \rangle \rangle - \langle \langle i+1, \psi_{i+1} | h_\parallel^z | \psi_i, \psi_{i+1} \rangle \rangle \]

where \( h_\parallel^z = J (S_{i,j-1}^z S_{i+1,j-1}^z + S_{i,j+1}^z S_{i+1,j+1}^z) \). Using the explicit form of the block eigenstates we can proof that the effective coupling is \( J_\parallel = J S^2 + O(J^2/\Delta_B) \), i.e. the product of the energy to create a domain wall in each chain times the expectation value of the magnetization in the two outer spins of the block. In the case of \( S = 1 \) Heisenberg chains we have off-diagonal terms \( h_{xy} = J/2 (S_{i,j-1}^x S_{i+1,j-1}^y + S_{i,j+1}^x S_{i+1,j+1}^y) + \text{h.c.} \). However, the \( S_1^z \) are still good quantum numbers and this prevents spin-flipping in the effective Hamiltonian. This is the strongest argument in favor of the low-energy equivalence of models \( 1 \) and \( 2 \). An essentially identical argument can be applied in the high dilution limit to compute effective coupling between two distant local magnetic moments obtaining now \( J_\parallel = 2S^2 \rho \) where \( \rho = E(s_j = s, s_k = -s) - E(s_j = s, s_k = s) \) is the energy of the chain connecting the two blocks with spin \( s_j \) in site \( j \) and \( s_k \) in site \( k \). In other words, \( \rho \) is the energy difference between anti-parallel and parallel spins at the end of the chain and it is directly related to the static stiffness of the (finite) chain which is non-zero even in the paramagnetic case. In both chain models the asymptotic behavior is \( \rho \sim \rho_0 \exp(-|k-j|/\xi_{ch}) \).

We can test numerically the validity of this model in the periodic diluted system. The critical temperature for the 2D anisotropic Ising model is given by the expression \( \sinh \beta_c J_x \sinh \beta_c J_y = 1 \) where \( J_x = J_c S \) and \( J_y = 2J_0^2 e^{-\frac{x}{\xi}} \) and \( \beta_c = 1/T_{c,0} \). The results (solid line) are compared with the numerical simulation in Fig. \( 1 \). The agreement emerges at \( T_c << \Delta_B \) where thermal population of excited states above the doublet is strongly suppressed and the block is described by an Ising variable. The effect of the quantum fluctuations in the microscopic models is integrated out in the effective classical model and can be formally separated into two parts: the reduction of the effective magnetic moment of the Ising spins \( |\uparrow\rangle, |\downarrow\rangle \) and the exponential decay of the couplings in the direction parallel to the chains. The most prominent feature of the model in the low dilution limit is the linear behavior of \( T_c \) as a function of \( x \) at fixed \( \xi_{ch} \). This property survives in the more realistic randomly diluted case for a large range of concentrations (see Fig. \( 3 \)).

It is interesting to consider whether the absence of percolation threshold is a robust feature against the presence of weak spin-flip processes between the central spin \( R \) and its neighbors in the perpendicular direction that belong...
to the chains, $h_{XY}^{XY} = J_{xy}^{xy} / 2 s_i^x (S_{i+1}^y + S_{i}^y) + h.c.$.
A finite value for $J_{xy}^{xy}$ breaks the conservation law for $s_i^z$ and establishes quantum coherence between $|\uparrow\rangle$ and $|\downarrow\rangle$.

In leading order, it has the same effect of a transverse magnetic field acting on the spin blocks. If $J_{xy}^{xy} \ll \Delta_L$ we can compute within second order perturbation theory the matrix element $\Gamma_{eff} = \langle \downarrow | H_{xy} | \uparrow \rangle = J_{xy}^{xy} / 4 \Delta_L$. This $\Gamma_{eff}$ should not be confused with the original transversal field $\Gamma$ in the chains. The effective model at $T=0$ is now a 2D ITF in that has a quantum critical point (QCP) for $\Gamma = 3.044(2)$ [12]. In the original model this translates into a QCP governed by the concentration and therefore a finite percolation threshold, $x_c^{-1} = \xi_{ch} \log(\Gamma_{eff}/\alpha \rho_0)$ where $\alpha$ is a numerical constant. Due to the fact that the random dilution affects only inter-chain sites a percolating cluster of magnetic moments exists with probability one for any finite value of $x$. But unlike the homogeneously diluted $S = 1/2$ 2D Heisenberg systems [13], it can be can be disordered by pure quantum fluctuations.

In Fig. 3, we show the generic zero temperature phase diagram that we expect for the randomly diluted system: Since the critical value for the effective matrix element $\Gamma_{eff}$ vanishes exponentially for $x \rightarrow 0$, strongly diluted ($x \ll 1$) experimental systems are most probably in the paramagnetic phase (PM). Nevertheless, since the system is disordered and close to a quantum phase transition at $\Gamma_{eff}(x)$, one expects the region $\Gamma_{eff} \ll \Gamma_c$ to be a Griffiths-McCoy phase [14, 15, 16], in which various connected spins $\Delta$ appear with an exponentially small probability $p_\Delta = \exp(-\Lambda N)$ ($p$ being the probability for an occupied site), but first order perturbation theory tells us that for small transverse field strengths the gap of this cluster is also exponentially small in the number of connected spins $\Delta \sim \exp(-\sigma N)$, leading to an exponentially large tunneling time. This implies, at zero temperature, a power law behavior for the autocorrelation function $\langle (\sigma_i^z(t) \sigma_j^z(0)) \rangle_{\omega} \sim t^{-\lambda/\sigma}$, and thus an algebraic singularity of, for instance, the local zero-frequency susceptibility $\chi(\omega = 0) \sim T^{-1+\lambda/\sigma}$. These Griffiths-McCoy singularities are generic for a randomly diluted transverse Ising system (actually generic for any kind of disorder). In our case the dilution is such that no isolated finite clusters exist, nevertheless parts of the system are more strongly coupled than others simply because there are more occupied inter-chain sites. Generic disorder, like random ferromagnetic bonds, also leads to Griffiths-McCoy singularities in non-diluted transverse Ising systems, as was demonstrated numerically [10], hence we also expect them to be present in the system we were considering here.

In conclusion we would like to stress the experimental implications of our theoretical findings. Our central predictions, particularly the universal scaling of the ordering temperature, should be straightforward to verify on $(R_x Y_{1-x})_2 Ba NiO_3$ powder samples by bulk magnetic and calorimetric measurements or by neutron diffraction. Conversely, the intrinsic chain correlation length can be effectively measured through measuring the $x$-dependence of the ordering temperature. The finer effects discussed above, including those associated with strong disorder effects and algebraic singularities, can be directly probed in more involved single-crystal neutron scattering experiments.

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