Field-Induced Disorder in a Gapped Spin System with Nonmagnetic Impurities

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We study the combined effect of doping and an external magnetic field on $S = \frac{1}{2}$ dimers which are weakly coupled by three-dimensional antiferromagnetic interactions J'. We show that application of an external magnetic field H opposes the long-range Néel order which is known to result from doping with nonmagnetic impurities and drives the system back to the disordered phase if 3D interactions J' are not too large. We discuss the zero temperature phase diagram in the (H, J') plane and suggest that the reentrant behavior can be experimentally observed.

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Gapped quantum spin systems with a singlet ground state and a triplet band as lowest excitations have received much attention both experimentally and theoretically in the last decade [1]. Typical examples include $S = \frac{1}{2}$ ladders, $S = \frac{1}{2}$ dimerized chains, Haldane $(S = 1)$ spin chains, various two-dimensionally coupled systems, k and systems of $S = \frac{1}{2}$ dimers weakly coupled by three-dimensional (3D) interactions. The dimer gap characterizing two spins 1/2 with antiferromagnetic exchange interaction survives interdimer interactions and an external magnetic field up to finite critical values; however, in the presence of nonmagnetic impurities the gap in an interacting dimer system is immediately destroyed, leading to the formation of a gapless continuum of low-lying states within the gap [2–6], and, in the presence of arbitrarily small 3D interactions, to long-range Néel ordering.

Our aim in this Letter is to investigate the combined influence of doping and of an external magnetic field on the phase diagram of a typical gapped spin system, namely, an interacting 3D system of spin-$\frac{1}{2}$ dimers as realized in the materials $KCuCl_3$ [7] and $TlCuCl_3$ [8]. The pure system is described by the Hamiltonian

$$\mathcal{H} = J\sum_{\boldsymbol{r}} \boldsymbol{S}_{\boldsymbol{r},1}\cdot\boldsymbol{S}_{\boldsymbol{r},2} - H\sum_{\boldsymbol{r}\sigma} S^z_{\boldsymbol{r},\sigma=1,2} + J'\sum_{\langle \boldsymbol{r}\boldsymbol{r}'\rangle}(\boldsymbol{S}_{\boldsymbol{r},1}\cdot\boldsymbol{S}_{\boldsymbol{r}',2} + \boldsymbol{S}_{\boldsymbol{r},2}\cdot\boldsymbol{S}_{\boldsymbol{r}',1}), \quad (1)$$

where $J > 0$ and $J' > 0$ are the intradimer and interdimer exchange couplings, respectively, and H is the external magnetic field directed along the z axis. The vector $\boldsymbol{r}$ labels the dimers located at the sites of a lattice with coordination number Z, and $\langle \ldots \rangle$ denotes summation only over neighboring dimers. In (1) we have assumed that the couplings are unfrustrated so that each spin $\boldsymbol{S}_{\boldsymbol{r},\sigma}$ belongs to one of the two sublattices ($\sigma = 1$ or $\sigma = 2$).

In the absence of impurities and external field, for weak interdimer coupling $J' \ll J$ this system has a singlet ground state without any magnetic order, and a finite gap $\Delta \sim J$ to the lowest (triplet) excitation. If J' exceeds some critical value, long-range Néel order appears; this situation is realized, e.g., in NH_4CuCl_3 [9,10].

An external magnetic field closes the gap at the critical field $H = H_c = \Delta$. At $H > H_c$ the system enters a new phase that is long-range ordered when arbitrarily weak 3D coupling is present. A finite magnetization along the field direction as well as staggered order in the plane perpendicular to the field appear, the U(1) symmetry in the xy plane is spontaneously broken. This transition may be viewed as the Bose-Einstein condensation of magnons [11]. For the material $TlCuCl_3$ this field-induced ordering has been studied in detail experimentally [11–14].

The presence of nonmagnetic impurities drastically changes this picture [2,4,15,16]. Impurities generate unpaired spins, which develop an effective interaction with each other mediated by the intact dimers between them. According to the Lieb-Mattis theorem [17], the interaction between two unpaired spins is antiferromagnetic if they belong to different sublattices and ferromagnetic otherwise, so in total the unpaired spins favor antiferromagnetic ordering. At $T = 0$ long-range antiferromagnetic order develops at any arbitrarily small concentration of impurities c. Unpaired spins thus form a separate subsystem with the energy scale for its dynamics set by the average effective interaction $\tilde{J} \ll \Delta$. The excitation spectrum of the impure system is governed by the gapless continuum of the low-lying states resulting from the ordered unpaired-spin subsystem; this continuum coexists with the states above the spin gap Δ of the pure system which survive with reduced weight.

So, taken *separately*, both doping with nonmagnetic impurities and application of an external field tend to induce ordering in a gapped spin system. In the following we will show that an external field may drive the doped system back into the disordered phase, causing reentrant behavior of the Néel order when H increases.

Dynamics of an intact dimer system.—To derive the effective interaction between two unpaired spins, we start with the dynamics of the pure dimer system using the

dimer field theory [18], a continuum version of the bond boson operator approach [13]. The quantum state is formulated as a product of dimer states of the form

$$|\psi\rangle = (1 - A^2 - B^2)^{1/2}|s\rangle + \sum_j (A_j + iB_j)|t_j\rangle, \qquad (2)$$

where $|s\rangle$ is the singlet state and $|t_j\rangle$, $j = x, y, z$ are the three triplet states of the spin dimer in the Cartesian basis. The vectors $\boldsymbol{A}$, $\boldsymbol{B}$ are related to the magnetization $\boldsymbol{M} = \langle \boldsymbol{S}_1 + \boldsymbol{S}_2 \rangle$ and staggered magnetization $\boldsymbol{L} = \langle \boldsymbol{S}_1 - \boldsymbol{S}_2 \rangle$ of the dimer as follows:

$$\boldsymbol{M} = 2(\boldsymbol{A} \times \boldsymbol{B}), \qquad \boldsymbol{L} = 2(1 - A^2 - B^2)^{1/2}\boldsymbol{A}. \qquad (3)$$

The Lagrangian density (per dimer) can be written as

$$\mathcal{L} = 2\hbar \boldsymbol{B} \cdot \frac{\partial \boldsymbol{A}}{\partial t} - E_{\mathrm{dim}} - \frac{ZJ'}{4}\{(\nabla \boldsymbol{L})^2 + (\nabla \boldsymbol{M})^2\},$$
$$E_{\mathrm{dim}} = J(A^2 + B^2) + \frac{ZJ'}{4}(\boldsymbol{M}^2 - \boldsymbol{L}^2) - HM_z, \qquad (4)$$

where we denote $(\nabla \boldsymbol{A})^2 \equiv \sum_i (\partial \boldsymbol{A}/\partial x_i)^2$ and the lattice constant is set to unity. Assuming $A, B \ll 1$, at the quadratic level one obtains the dispersion of a magnon with $S^z = \mu$ as $\varepsilon(\boldsymbol{k}) = \Delta(1 + k^2\xi^2)^{1/2} - \mu H$, where

$$\Delta = J(1 - 2\delta)^{1/2}, \quad \xi = \frac{v}{\Delta}, \quad v = J\delta^{1/2}, \quad \delta = \frac{ZJ'}{2J}. \qquad (5)$$

Here δ is the dimensionless interdimer (3D) exchange coupling and ξ is the spin correlation length *of a pure dimer system*. If the 3D interaction is small, i.e., $\delta \ll 1$, then $\xi \simeq \delta^{1/2} \ll 1$.

Interactions between unpaired spins.—The effective interaction between two unpaired spins can be estimated [2] in second-order perturbation theory,

$$J_{\mathrm{eff}} = \mathcal{K} \int d^3k \frac{(ZJ')^2}{\varepsilon(k)} e^{i\boldsymbol{k}\cdot\boldsymbol{r}} = \frac{4\pi\mathcal{K}(ZJ')^2}{rv\xi} K_1\left(\frac{r}{\xi}\right), \qquad (6)$$

where $\mathcal{K}$ is a constant of the order of unity, determined by the matrix elements of the perturbation, $\boldsymbol{r}$ is the vector connecting the two spins, K_1 is the modified Bessel function, and we omit the oscillating sign depending on whether or not the spins belong to the same sublattice. We use the $H = 0$ expression for the magnon energy since, as will become clear below, we are interested in fields which are small compared to the gap Δ. Since $\xi \ll 1$, one can write down the effective interaction as

$$J_{\mathrm{eff}}(r) = J_1 r^{-3/2} e^{-r/\xi}, \qquad (7)$$

where $J_1 = CJ\delta^{5/4}$ with $C = \sqrt{128\pi^3}\mathcal{K}$. Since J_{eff} decays very fast with r, it is sufficient to take into account interactions only between neighboring unpaired spins. For a given impurity concentration c, the distribution of distances R from an impurity to its nearest neighbor can be estimated in the continuum approximation as

$$p(R) = c \exp\left\{-\frac{4\pi c}{3}R^3\right\}, \qquad \int p(R) 4\pi R^2 dR = 1. \qquad (8)$$

The distribution of effective exchange couplings $\tilde{J}$ between the neighboring unpaired spins is given by

$$P(\tilde{J}) = \sum_{\boldsymbol{R}} p(R)\delta[\tilde{J} - J_{\mathrm{eff}}(R)].$$

Replacing the sum by an integral, one obtains

$$P(\tilde{J}) = \Theta(J_1 e^{-1/\xi} - \tilde{J}) \frac{4\pi c\xi}{\tilde{J}} R_1^2 \exp\{-4\pi c R_1^3/3\}, \qquad (9)$$

where $R_1 \simeq \xi \ln(J_1/\tilde{J})$ and Θ is the Heaviside function; note that $P(\tilde{J})$ has an upper cutoff [19]. The maximum of $P(\tilde{J})$ occurs at $\tilde{J} = \tilde{J}_{\mathrm{peak}} \simeq J_1 \exp\{-(4\pi c\xi^3)^{-1/2}\}$, and the average of $\tilde{J}$ is

$$\langle \tilde{J} \rangle = \int JP(J)dJ \simeq 4\pi c\xi J_1 e^{-1/\xi} \gg \tilde{J}_{\mathrm{peak}}. \qquad (10)$$

The interacting system of unpaired spins and intact dimers.—We go on to study the static properties of the impure system in a magnetic field in order to obtain the qualitative form of the $T = 0$ phase diagram in (H, J') space. We use a mean-field approximation (MFA) for treating the interaction between the dimers and unpaired spins, and minimize the total energy $E = E_{\mathrm{dim}} + E_{\mathrm{unp}} + E_{\mathrm{int}}$. Here E_{dim} is the energy of the intact dimers, Eq. (4), E_{unp} is the energy of the unpaired-spin subsystem, and E_{int} is the interaction between those two subsystems. We first derive from MFA the static properties for fixed $\tilde{J}$, and the average over $\tilde{J}$ is performed afterwards. The unpaired-spin subsystem is ordered, and we describe it as a two-sublattice antiferromagnet. Denoting the angle between its sublattice magnetization and the z axis as θ and the average unpaired-spin length as s ($s < \frac{1}{2}$ because of the quantum spin reduction), we write the interaction energy (per dimer) as

$$E_{\mathrm{int}} = cZJ's(M_z \cos\theta - L_x \sin\theta), \qquad (11)$$

where the x axis is chosen along the direction of the staggered order. In the self-energy E_{unp} of the unpaired-spin subsystem we include, for each unpaired spin, the interaction with nearest unpaired neighbors only; this gives for the energy per dimer (hence the factor $2c$ in front)

$$E_{\mathrm{unp}} = 2c\left(-\frac{1}{2}\tilde{J}_S s^2 \sin^2\theta - Hs\cos\theta\right). \qquad (12)$$

Here $\tilde{J}_S$ is the sum of interactions over the first "coordination sphere" in a fictitious random lattice formed by unpaired spins. While this lattice is a 3D one, $\tilde{J}_S$ is on average dominated by $\tilde{J}$ for the nearest neighbor due to the fast decay of $J_{\mathrm{eff}} \sim e^{-r/\xi}$: given there is a nearest neighbor at distance $R \sim c^{-1/3}$, the other neighbors' contribution $4\pi R^2 \xi c\tilde{J}$ to $\tilde{J}_S$ is negligible, so $\tilde{J}_S \simeq \tilde{J}$.

It is convenient to parametrize the vectors $\boldsymbol{A}$ and $\boldsymbol{B}$ as follows:

$$\boldsymbol{A} = \{\sin\alpha\cos\gamma, 0, 0\}, \qquad \boldsymbol{B} = \{0, \sin\alpha\sin\gamma, 0\}.$$

It is easy to see that $\alpha = 0$, $\theta = 0$ is always an extremum of E describing a state with a fully polarized unpaired-spin subsystem and a nonmagnetic (singlet) dimer subsystem. A simple analysis yields the following equation for the stability boundary of this solution:

$$(H^2 - J^2)(H - \tilde{J}s) + cs(ZJ')^2(J + csH - cs^2\tilde{J}) + ZJ'(H - \tilde{J}s)(J - 2csH) = 0. \quad (13)$$

Another solution is the saturated state with all spins fully polarized, which corresponds to $\theta = 0$, $\alpha = \pi/2$, $\gamma = \pi/4$. It becomes unstable if the field drops below the value

$$H_s = J + ZJ'(1 + cs). \quad (14)$$

At fixed J' Eq. (13) has two solutions for H. Up to the second order in J', $\tilde{J}$ and to the first order in c these lower and upper critical fields are given by

$$H_{c1} \simeq \tilde{J}s + cs(ZJ')^2/J, \quad (15)$$

$$H_{c2} \simeq J - ZJ'(1/2 - cs) - (ZJ')^2/(4J). \quad (16)$$

For $H_{c1} < H < H_{c2}$ the extremum at $\alpha = \theta = 0$ is the stable solution. Note that H_{c2} is in fact the critical field of a pure material determined by the intradimer coupling J, with only small corrections from impurities. In the vicinity of H_{c2} the behavior is similar to that in the pure limit (see Sect. 1.6 of [1]): $\alpha \simeq [(H - H_{c2})/(3J\delta)]^{1/2}$, $\theta \simeq \sqrt{2}\delta\alpha$, and $\gamma \simeq \pi/4 - \delta$. The lower critical field H_{c1} marks a new energy scale related to the impurities, below H_{c1} the intact dimer parameters α, γ, and the orientation θ of unpaired spins are obtained as

$$\theta \simeq [2(1 - H/H_{c1})]^{1/2}, \qquad \alpha \simeq 2\delta cs\theta\cos\gamma, \qquad \gamma \simeq (\tilde{J} - cZJ')s/2, \quad (17)$$

Thus, the following physical picture emerges: At zero external field, impurities order due to their effective interaction and induce a staggered order also in the system of intact dimers, $\alpha \neq 0$. Application of the external field leads to canting of all spins in the field direction. Whereas this canting is weak for the intact dimers, it is much stronger in the weakly interacting subsystem of unpaired spins: θ moves from $\pi/2$ towards 0 with increasing field. Thus the effective staggered field which the dimer subsystem feels from the unpaired spins is diminished. At $H = H_{c1}$ the unpaired spins become saturated ($\theta = 0$), and simultaneously the staggered order in the dimer subsystem vanishes ($\alpha = 0$). The system enters a disordered phase with dimers in the singlet state and unpaired spins fully magnetized, which extends up to another critical field H_{c2}. With increase of J', those two critical fields coalesce and the disordered phase disappears. The resulting phase diagram is shown schematically in Fig. 1.

The main feature of the above picture, the reentrant behavior of the order, is due to the existence of two different energy scales and thus is independent of the MFA, while the true critical exponent in Eq. (17) will of course differ from the MFA value $\frac{1}{2}$.

As a by-product, the effect of the impurities on the high-energy spectrum can be obtained: Replace E_{dim} in the Lagrangian (4) by the full energy E and assume that θ is frozen at its equilibrium value. This amounts to taking into account the static part of the unpaired-spin subsystem, whereas its dynamics, occurring at the much lower energy scale $\tilde{J}$, is neglected. As a result the triplet gap Δ splits due to the emergence of the staggered order in x direction. At $H = 0$, the gaps are related to the order parameter $A_0 \propto m_{\mathrm{st}}$ as follows:

$$\Delta_{y,z} = (\Delta^2 + 4v^2A_0^2)^{1/2}, \quad \Delta_x = (\Delta^2 + 12v^2A_0^2)^{1/2}. \quad (18)$$

For $A_0 \ll 1$, the square of the order parameter determining the intensity of the Bragg peak is approximately linearly related to the gap energy. This proportionality between the change in the gap energy and the intensity of the Bragg peak was observed experimentally in $TlCuCl_3$ doped with Mg [20] by varying the temperature. Our calculation is for $T = 0$, but we expect that this relation continues to be valid for finite temperature.

Averaging over the impurity distribution.—As a final step we discuss the influence of the statistical character of the effective interaction $\tilde{J}$ between unpaired spins on the staggered magnetization. For a fixed homogeneous $\tilde{J}$, in the vicinity of H_{c1} given by Eq. (15) the total staggered magnetization per dimer vanishes as a square root [21]:

$$m_{\mathrm{st}} \simeq cs(1 + ZJ'/J)[8(1 - H/H_{c1})]^{1/2}, \quad (19)$$

where the term proportional to J' describes the contribution of intact dimers. The physical critical field $H_1 = \max H_{c1}$ at which the staggered magnetization m_{st} completely disappears is determined by the maximum value for $\tilde{J}$ (the cutoff of the distribution), which yields

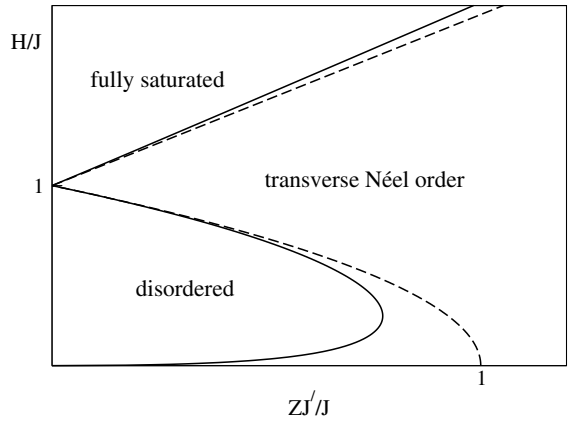

FIG. 1. Schematic mean-field $T = 0$ phase diagram of the model (1). The dashed line corresponds to the pure system and the solid line to a finite concentration of impurities. The boundaries of the disordered and saturated phases are determined by (13) and (14), respectively.

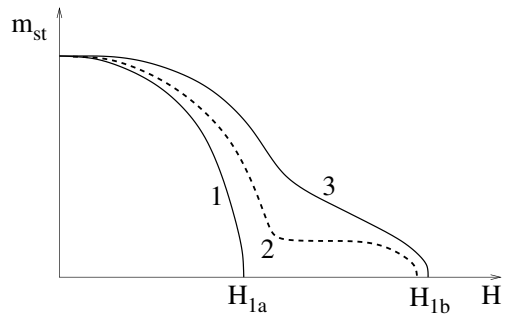

FIG. 2. Schematic field dependence of the staggered magnetization. Curves 1, 2, and 3 correspond to weak $\delta \ll \delta_1$, intermediate $\delta_1 \ll \delta \ll \delta_2$, and strong $\delta \gg \delta_2$ interdimer coupling, respectively; here $\delta = \frac{ZJ'}{2J}$ and $\delta_{1,2}$ as in (21), (22).

$$H_1 \simeq sJ(C\delta^{5/4}e^{-1/\sqrt{\delta}} + 4c\delta^2). \quad (20)$$

However, in order to describe the behavior of $m_{\rm st}$ between $H = 0$ and $H = H_1$ we have to consider its variation due to the random spatial distribution of $\tilde{J}$. Three regimes can be identified: for very weak interdimer coupling

$$\delta \ll \delta_1, \qquad \text{where } \delta_1^{3/4}\exp\{\delta_1^{-1/2}\} = C/(4c), \quad (21)$$

the second term in (15) is always leading, the effect of averaging is negligible, $m_{\rm st}$ is nearly homogeneous and varies as a pure square root, $m_{\rm st} \propto \sqrt{H_{1a} - H}$, $H_{1a} = 4cs\delta^2 J$. For stronger J' the inequality (21) is violated and for $\delta \gg \delta_1$ the first term in (15) dominates, yielding the critical field $H_1 \simeq H_{1b} = sJ_1 e^{-1/\xi}$. The behavior of $m_{\rm st}$ for $H < H_1$ depends again on the interdimer interaction strength: If $\langle\tilde{J}\rangle \ll c(ZJ')^2/J$, which corresponds to the intermediate coupling

$$\delta_1 \ll \delta \ll \delta_2, \qquad \text{with } \delta_2^{1/4}\exp\{\delta_2^{-1/2}\} = \pi C, \quad (22)$$

then the main contribution to $m_{\rm st}$ has an overall behavior $\propto \sqrt{H_{1a} - H}$, and in addition there is a weak shoulder extending to a higher field $H = H_{1b}$. The shoulder results from the high exchange energy tail of the distribution and reflects the inhomogeneity of the system in this limit: small islands with unsaturated unpaired spins survive in the field range $H_{1a} < H < H_{1b}$ due to the random distribution of impurities. The strength of the shoulder grows with increasing J' and finally, for $\delta \gg \delta_2$, merges with the main part, as sketched in Fig. 2. An estimate for $c = 0.01$ yields $\delta_1 \sim 8 \times 10^{-3}$ and $\delta_2 \sim 0.02$.

Summary.—We have shown that in a system of weakly coupled spin dimers doped with nonmagnetic impurities, the application of an external field H may lead to a reentrant behavior of long-range antiferromagnetic order when the field increases. Essentially, this result comes from the fact that the distribution of couplings between unpaired spins has an upper cutoff, setting a new energy scale which corresponds to the lower critical field H_{c1} and is much smaller than the intradimer exchange J determining the upper critical field H_{c2}. Those two energy scales converge with the increase of interdimer coupling J', so the reentrant behavior is more likely to be observed in materials with a sufficiently small ratio of J'/J. Therefore, among known materials, $KCuCl_3$ [7] is the most promising candidate for an experimental investigation of this phenomenon.

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