Bose-Einstein condensation in antiferromagnets close to the saturation field

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At zero temperature and strong applied magnetic fields the ground state of an anisotropic antiferromagnet is a saturated paramagnet with fully aligned spins. We study the quantum phase transition as the field is reduced below an upper critical H_{c2} and the system enters a *XY*-antiferromagnetic phase. Using a bond operator representation we consider a model spin-1 Heisenberg antiferromagnetic with single-ion anisotropy in hypercubic lattices under strong magnetic fields. We show that the transition at H_{c2} can be interpreted as a Bose-Einstein condensation (BEC) of magnons. The theoretical results are used to analyze our magnetization versus field data in the organic compound $NiCl_2$-$4SC(NH_2)_2$ (DTN) at very low temperatures. This is the ideal BEC system to study this transition since H_{c2} is sufficiently low to be reached with static magnetic fields (as opposed to pulsed fields). The scaling of the magnetization as a function of field and temperature close to H_{c2} shows excellent agreement with the theoretical predictions. It allows us to obtain the quantum critical exponents and confirm the BEC nature of the transition at H_{c2}.

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The organic compound $NiCl_2$-$4SC(NH_2)_2$ (DTN) undergoes a field induced nonmagnetic to *XY*-antiferromagnetic transition.[1,2] This transition can be viewed as a Bose-Einstein condensation of magnons associated with the Ni spin 1 degrees of freedom. Other magnetic systems with a singlet ground state either with spin-1 Ni atoms or spin-1/2 dimers have also been shown to exhibit this transition.[3–5] At zero temperature it is driven by the magnetic field H that reduces the Zeeman energy of the $S^z=1$ state until it becomes degenerate with that of the product state of $S_i^z=0$. At this point, $H=H_{c1}$, the antiferromagnetic (AF) interactions give rise to a long range ordered phase. Experimentally, the magnetization M at very low temperatures starts to increase above the critical magnetic field H_{c1} and eventually saturates above a critical magnetic field H_{c2}.[1,2,6–9] The transition at H_{c1} has been intensively investigated, both theoretically[12,13] and experimentally,[1,2,6–9] while the one at H_{c2} is much less studied. The DTN is the ideal BEC system to investigate the latter transition since detailed magnetization curves can be obtained close to the critical field $H_{c2}=12.3$ T. In other well known BEC systems, as $BaCuSi_2O_6$ (Ref. 10) and $TlCuCl_3$,[11] the critical fields H_{c2} are 49 T and 83 T, respectively, and presently can only be reached using pulsed fields. The excellent quality of the magnetization versus field curves obtained in DTN using standard superconducting coils is essential for the scaling analysis presented here.

In this paper we study the transition at H_{c2}. A theoretical approach is more directly developed, starting from the saturated paramagnetic (PARA) phase. We consider decreasing the external magnetic field at $T=0$ to the critical value H_{c2} where the transverse components of the magnetization condense. A scaling approach for this transition has recently been proposed.[14] In this Brief Report we provide the microscopic theory for this transition. We identify its universality class as a Bose-Einstein condensation associated with a dynamic exponent $z=2$. We compare the predictions for the scaling behavior of the magnetization close to the quantum critical point (QCP) ($T=0$, $H=H_{c2}$) with experimental magnetization data on DTN and obtain an excellent agreement.

For a long time the magnetically ordered state and low energy excitations of quantum Heisenberg magnets have been studied using the spin-wave expansion (see Ref. 15 and references therein). This is usually implemented by expressing the components of the spin operators at lattice sites i in terms of canonical boson operators $b_k^\dagger$ and b_k using the Holstein-Primakoff (HP) transformation,[16] the Dyson-Maleev (DM) transformation,[17,18] or the Schwinger transformation[19] (ST). Here we use the bond-operator mean-field theory[20] (BOMFT) to study spin-1 Heisenberg AF in hypercubic lattices with single-ion anisotropy close to the quantum phase transition at the saturation field H_{c2}. It yields the phase diagram and the thermodynamic behavior of the model close to H_{c2}. The BOMFT gives an exact description of this transition for three-dimensional (3D) systems. The reason is that the effective dimension $d_{\text{eff}}=d+z=5$ associated with the QCP is larger than the upper critical dimension $d_c=4$ above which mean-field theory is exact.

The Hamiltonian describing the magnetic system is

$$\mathcal{H}=\frac{J}{2}\sum_{\langle i,j\rangle}(S_i^xS_j^x+S_i^yS_j^y+S_i^zS_j^z)+D\sum_i(S_i^z)^2-H\sum_i S_i^z, \quad (1)$$

where the sum is over all nearest neighbor pairs of a d-dimensional hyper-cubic lattice with N sites occupied by spins with $S=1$. $J>0$ is the AF exchange coupling, D is the single-ion anisotropy, and H the magnetic field applied in the z direction ($g\mu_B=1$). Starting from the bond-operator repre-

sentation for two spins $S=1/2$,[20] Wang and collaborators[12,13,21] obtained a representation for a spin-1 Heisenberg system with a single-ion anisotropy in terms of these operators. At zero temperature and for external magnetic fields larger than the saturation magnetic field H_{c2} the spins are fully aligned with the field. In this case the bond operator representation can be expressed as

$$S^+ = \sqrt{2}\bar{u}t_z, \quad S^- = \sqrt{2}\bar{u}t_z^\dagger, \quad S^z = 1 - t_z^\dagger t_z, \tag{2}$$

with the constraint $u_i^\dagger u_i + t_{i,z}^\dagger t_{i,z} = 1$. We have used that in this large field case the components of the spins perpendicular to the field can be projected out and those parallel *condense*, such that $u_i = u_i^\dagger = \bar{u}$. This mapping is exact for $H > H_{c2}$, where the probability of the down spin state $d^\dagger d|0\rangle$ is strictly zero at $T=0$. The magnetic ordering for $H=H_{c2}$ can be identified as a Bose-Einstein condensation of the transverse components of the spins which give rise to the collective *magnon* excitations. The operators $t_{i,z}^\dagger t_{i,z}$ describe the departure of the spins from the field direction and are associated with these excitations. Replacing Eq. (2) in Eq. (1) with $S^\pm = S^x \pm iS^y$ and changing from atomic to normal coordinates, i.e., taking $t_{i,z}^\dagger = (1/\sqrt{N})\Sigma_k e^{-ik\cdot r_i} b_k^\dagger$, we have

$$\mathcal{H}_{\rm mf} = \sum_k \omega_k b_k^\dagger b_k + E_g, \tag{3}$$

where

$$E_g = N\left(\frac{JZ}{2} + D\bar{u}^2 - H - \mu\bar{u}^2 + \mu\right) \tag{4}$$

is the ground state energy of the system. The dispersion relation of the excitations for $H > H_{c2}$ is given by

$$\omega_k = H - (D+\mu) - JZ(1 - \bar{u}^2\gamma_k), \tag{5}$$

with $\gamma_k = d^{-1}\Sigma_{\nu=1}^d \cos(\mathbf{k}\cdot\mathbf{a}_\nu)$ and Z the number of nearest neighbors. The chemical potential μ was introduced to impose the constraint condition of single occupancy. This and the parameter $\bar{u}$ are determined by solving coupled, self-consistent, saddle point equations.[13]

For fixed field H or temperature T, the thermodynamic quantities can be obtained from the internal energy U. This is given by[22,23] $U = \Sigma_k \omega_k \langle b_k^\dagger b_k\rangle + E_g$, with the Bose factor $n_k = \langle b_k^\dagger b_k\rangle = \frac{1}{2}[\coth(\beta\omega_k/2) - 1]$ and E_g given by Eq. (4). For simplicity, we impose boundary conditions on a d-dimensional hypercubic lattice with primitive lattice vectors $\mathbf{a}_\nu$ and lattice spacing $a = |\mathbf{a}_\nu| = 1$. The minus sign in front of J takes into account explicitly the antiferromagnetic nature of the exchange interactions. For hyper-cubic lattices, the minimum of the spin-wave spectrum occurs for $q = Q = (\pi/a, \pi/a, \pi/a)$ in three dimensions. Since $\omega_Q = H - (D+\mu) - JZ(\bar{u}^2+1)$, the condition $\omega_Q \equiv 0$ defines the critical field $H_{c2} = D + \mu + JZ(\bar{u}^2+1)$, below which the spin-wave energy becomes negative signaling the entrance of the system in the AF phase. Finally, writing $k = Q + q$ and expanding for small q, the spin-wave dispersion relation can be obtained as follows:

$$\omega_q = (H - H_{c2}) + \mathfrak{D}q^2, \tag{6}$$

where the spin-wave stiffness at $T=0$ is given by, $\mathfrak{D} = J\bar{u}^2$. The quantum phase transition at H_{c2} has a dynamic exponent $z=2$ due to the ferromagneticlike dispersion of the magnons, in spite of the antiferromagnetic character of the interactions.[14]

Magnetization. Close to the critical field H_{c2} the temperature-dependent magnetization should follow a power law.[24] We define the variation of uniform magnetization per volume V as $\Delta M = (M_{\rm sat} - M)/V = \Sigma_k \langle b_k^\dagger b_k\rangle$ where $M_{\rm sat}$ is the saturation magnetization. Considering the spectrum of excitations, Eq. (6), we have in the thermodynamic limit

$$\Delta M = \frac{S_d}{4\pi^d \mathfrak{D}^{d/2}} (k_B T)^{d/2} \int_y^\infty dx (x-y)^{d/2-1} \left(\coth\frac{x}{2} - 1\right), \tag{7}$$

where $x = \beta\omega_q = y + \beta^2 \mathfrak{D} q^2$, $y = \beta\delta$, S_d the solid angle and we have defined $\delta = |H - H_{c2}|$ as the distance to the QCP. At this point, we have to consider in which region of the phase diagram (see Fig. 1 of Ref. 14) we are interested. Because we want to calculate the magnetization above H_{c2} and particularly at the quantum critical trajectory, $H = H_{c2}$, $T \to 0$, we consider region II in Fig. 1 of Ref. 14 where $k_B T \gg \delta$ and consequently $y \ll 1$. Calculating the integral above in three dimensions, we obtain $\Delta M_{\rm 3D} = {\rm polylog}\left(\frac{3}{2}, e^{-y}\right) \times (k_B T)^{3/2}/(\pi^{3/2}\mathfrak{D}^{3/2})$, where ${\rm polylog}(a,z) = \Sigma_{n=1}^\infty z^n/n^a$ is the general polylogarithm function of index a at the point z. Along the quantum critical trajectory, $H = H_{c2}$, $T \to 0$, we find $\Delta M_{\rm 3D}(\delta=0) = g\mu_B \zeta(3/2)(k_B T)^{3/2}/(\pi^{3/2}\mathfrak{D}^{3/2})$, where ζ is the Riemann zeta function.

Specific heat. From the internal energy U obtained before and using the thermodynamic relation, $C_V = \partial U/\partial T$ we get

$$C_V = \frac{k_B S_d (k_B T)^{d/2}}{2\pi^d \mathfrak{D}^{d/2}} \int_y^\infty dx\, x^2 (x-y)^{d/2-1} \sinh^{-2}\left(\frac{x}{2}\right). \tag{8}$$

Again, we consider $y \ll 1$ and, along the quantum critical trajectory in 3D, we find $C_V(\delta=0) = 15 k_B(\sqrt{1+4\pi}-1) \times (k_B T)^{3/2}/(8\pi^{3/2}\mathfrak{D}^{3/2})$.

Susceptibility. Here we use the relation, $\chi = \partial M/\partial H$, where M is the magnetization. Taking the derivative of Eq. (7) with respect to H and changing variables, we have at $\delta = 0$

$$\chi = \frac{S_d}{8\pi^d \mathfrak{D}^{d/2}} (k_B T)^{d/2-1} \int_0^\infty dx\, x^{d/2-1} \sinh^{-2}\left(\frac{x}{2}\right). \tag{9}$$

For 3D the longitudinal susceptibility at the critical field is given by $\chi(\delta=0) = (k_B T)^{1/2}/[(\pi-1)\sqrt{\pi}\mathfrak{D}^{3/2}]$.

The temperature dependence of the physical properties calculated above in 3D can be easily obtained from the fact that the QCP at H_{c2} is governed by *Gaussian* exponents and the free energy has a scaling form,[24]

$$f \propto |\delta|^{2-\alpha} F(T/|\delta|^{\nu z}), \tag{10}$$

where $\delta = (H - H_{c2})$ as defined before. The Gaussian nature of the exponents in 3D is a consequence that the effective di-

mension $d_{\rm eff}=d+z=5$, which is larger than the upper critical dimension[28] $d_c=4$.

Critical line. The critical line that separates the polarized PARA state from the AF phase with nonzero staggered magnetization can be written on the neighborhood of the QCP as, $T_N(H)\propto|H_{c2}-H|^{\psi}$. Theories for a 3D Bose gas[5,25] and mean-field treatment[6] give a universal value, $\psi=2/3$. Scaling theory shows that although $d_{\rm eff}>d$ in three dimensions, the magnon-magnon interaction is dangerously irrelevant and must be considered.[24,26] Then we expect that the quartic corrections to the mean-field result Eq. (3) will now be important. Within the HP representation for the spin operators, the mean-field Hamiltonian including the dynamical spin wave interactions is given by

$$\mathcal{H}'_{\rm mf}=\sum_k \omega_k b_k^\dagger b_k+\sum_{k,k'}\left(\frac{JZ}{2}+D\right)b_k^\dagger b_k b_{k'}^\dagger b_{k'}+E_g, \quad (11)$$

where $(JZ/2+D)$ works as an effective repulsion between the magnons. The dispersion ω_k is given by Eq. (5) and E_g by Eq. (4). We decouple the spin-wave interaction as $b_k^\dagger b_k b_{k'}^\dagger b_{k'}\approx b_k^\dagger b_k\langle b_k^\dagger b_k\rangle$. Thus trivially we obtain the internal energy as

$$U'=\sum_q \omega'_q\langle b_q^\dagger b_q\rangle+E_g, \quad (12)$$

where we have already considered the proximity to the QCP $(H=H_{c2}, T=0)$. The spectrum of excitations taking into account magnon-magnon interactions is

$$\omega'_q=(H-H_{c2})+\left(\frac{JZ}{2}+D\right)\langle b_k^\dagger b_k\rangle+\mathcal{D}q^2. \quad (13)$$

We set up an equation for the critical line within the mean-field approximation where the effect of magnon-magnon fluctuations are included in a self-consistent manner. The critical temperature $T_N(H)$ is determined by the condition $\delta(H,T_N)=0$, where

$$\delta(H,T_N)=(H-H_{c2})+\left(\frac{JZ}{2}+D\right)\frac{S_d}{4\pi^2}\left(\frac{k_BT_N}{\mathcal{D}}\right)^{d/2}\int_0^\infty dx x^{d/2-1}\left(\coth\frac{x}{2}-1\right). \quad (14)$$

For 2D the integral above diverges as expected from general arguments.[27] For 3D we get $k_BT_N=[\zeta(3/2)(JZ/2+D)]^{-2/3}\pi\mathcal{D}(H_{c2}-H)^{2/3}$. Notice that the effective magnon-magnon coupling strength $(JZ/2+D)$ determines the transition temperature, despite the Gaussian exponents, as expected from the *dangerously* irrelevant nature of the magnon-magnon interactions. If we write the equation for the critical line, $\delta(H,T)=0$, in the form $H_{c2}(T)=H_{c2}(0)-v_0T^{1/\psi}$, with v_0 related to the spin-wave interaction, we identify the *shift exponent*, $\psi=z/(d+z-2)=2/3$, in agreement with the renormalization group (RG) result.[26] The temperature dependence of δ arising from the spin-wave interactions can modify the temperature dependence of ΔM, C_V and χ at $H=H_{c2}$. In the limit $T\to0$ we can easily see that the purely Gaussian results for ΔM and C_V calculated above are dominant. However, for the longitudinal susceptibility the spin-wave interactions modify the purely Gaussian result. In this case, it is straightforward to show that for, $H=H_{c2}$, $T\to0$, the dominant is $\chi\propto T^{1/4}$, instead of $\chi\propto T^{1/2}$ calculated before.

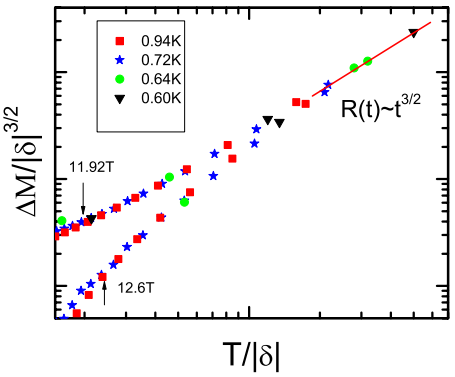

FIG. 1. (Color online) Scaling plot in logarithmic scales of the magnetization data for the compound DTN obtained for fields up to 17 T and temperatures $T=0.60$, 0.64, 0.72, and 0.94 K. The line shows the asymptotic behavior of the scaling function $R(t)\sim t^{3/2}$ in Eq. (15), for $H\to H_{c2}$. The arrows indicate the region of validity of the scaling.

Scaling analysis of the magnetization. We start from the free energy density, which close to the zero temperature quantum phase transition has the scaling form given by Eq. (10). The zero temperature critical exponents α, ν and the dynamic exponent z are related to the dimensionality of the system d by the quantum hyperscaling relation, $2-\alpha=\nu(d+z)$.[28] In general for $d_{\rm eff}=d+z>4$, i.e., above the upper critical dimension $d_c=4$, the exponents associated with the QCP at $\delta=0$ take Gaussian values, and in particular the correlation length exponent, $\nu=1/2$. That this is the case in the present theory can be immediately verified writing the thermodynamic functions in a scaling form and identifying the relevant exponents. Furthermore, Eq. (6) yields the dynamic exponent $z=2$. Using the relation $\Delta M\propto\partial f/\partial H$ we get

$$\Delta M\propto|\delta|^{1-\alpha}R(T/|\delta|^{\nu z}), \quad (15)$$

where $\Delta M=M_{\rm sat}(T,H_{\rm sat})-M(T,H)$ and $M_{\rm sat}$ is measured at the highest fields, $H_{\rm sat}\gtrsim15$ T. Using the hyperscaling relation for 3D we obtain $1-\alpha=\nu(3+z)-1=(1/2)(3+2)-1=3/2$.

Figure 1 shows the scaling plot of the magnetization for the compound DTN in fields up to 17 T and for several temperatures. The magnetization data was obtained using a vibrating sample magnetometer adapted to be used in a ^{3}He cryostat. The external magnetic field, produced by a superconducting coil, was aligned with the tetragonal axis of the sample, necessary condition to induce BEC. As shown in the figure the experimental data collapses in a good scaling plot when using the critical exponents appropriate for three dimensions. It can also be seen in Fig. 1 that for $H\to H_{c2}$, the scaling function $R(t\to\infty)\sim t^{3/2}$, such that, in this limit $\Delta M_{\rm 3D}\propto T^{3/2}$ in agreement with the theory. The lower (upper)

branch in Fig. 1 corresponds to data for higher (lower) fields than the critical magnetic field $H_{c2}(0)=12.3$ T. This critical field obtained from a criterion of best data collapse is in very good agreement with that found by Paduan-Filho *et al.*[2] using numerical differentiation of the magnetization data. We point out that a good scaling of the data is observed for magnetic fields sufficiently close to the critical field, i.e., for $11.92\ \mathrm{T} < H_{c2} < 12.6\ \mathrm{T}$.

Summary. In spite of the large literature on the subject of BEC of magnons, the phase diagram and the thermodynamic properties around the upper critical magnetic field H_{c2}, have not yet been completely examined. Since, in general, high magnetic fields are necessary to reach H_{c2}, experimental results and consequently theoretical work are much more scarce in this region of the phase diagram. As we pointed out, the organic compound $NiCl_2$-$4SC(NH_2)_2$ (DTN) is ideal for this kind of studies since detailed magnetization curves can be obtained for very low temperatures close to H_{c2}. With this motivation we introduced a BOMFT approximation to study theoretically the upper field transition. We have obtained the dominant low temperature behavior of the magnetization ($\Delta M \propto T^{3/2}$), specific heat ($C_V \propto T^{3/2}$), and susceptibility ($\chi \propto T^{1/4}$) at the quantum critical trajectory and determined the shift exponent of the Neel line. We pointed out that, although the magnon-magnon interactions are irrelevant in the RG sense close to the QCP, they should be taken into account and determine the temperature dependence of the critical line and that of the susceptibility along the quantum critical trajectory. Our mean-field approach is justified since the effective dimension for the transition at the QCP ($H=H_{c2}$, $T=0$) is above the upper critical dimension. Finally using the theoretical prediction we obtained for the scaling form of the field and temperature dependent magnetization close to $H=H_{c2}$, $T=0$, we performed a scaling analysis of our magnetization data for DTN. The very good agreement between the theoretical and experimental results provides unequivocal evidence that the transition at H_{c2} is a BEC of magnons.

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