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Critical behavior of the magnetization in the spin-gapped system NiCl2–4SC(NH2)2
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Citation: J. Appl. Phys. 105, 07D501 (2009); doi: 10.1063/1.3055265
View online: http://dx.doi.org/10.1063/1.3055265
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Critical behavior of the magnetization in the spin-gapped system \( \text{NiCl}_2-4\text{SC} (\text{NH}_2)_2 \)

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(Received 12 November 2008; received 12 September 2008; accepted 1 October 2008; published online 29 January 2009)

We report accurate magnetization measurements on the spin-gap compound \( \text{NiCl}_2-4\text{SC} (\text{NH}_2)_2 \) around the low portion of the magnetic induced phase ordering. The critical density of the magnetization at the phase boundary is analyzed in terms of a Bose–Einstein condensation (BEC) of bosonic particles, and the boson interaction strength is obtained as \( v_0=0.61 \text{ meV} \). The detailed analysis of the magnetization data across the transition leads to the conclusion for the preservation of the \( U(1) \) symmetry, as required for BEC. © 2009 American Institute of Physics. [DOI: 10.1063/1.3055265]

Magnetic-field-induced Bose–Einstein condensation (BEC) in gapped quantum magnets has attracted interest recently.1–3 In the spin-gap compound \( \text{NiCl}_2-4\text{SC} (\text{NH}_2)_2 \), known as DTN, the single ion anisotropy \( D=0.75 \text{ meV} \) is responsible for the splitting of the \( S=1 \) triplet in a singlet ground state and a doublet.4,5 The application of a magnetic field induces a long-range order, even at zero temperature. However, in the field-induced gapless phase at \( H_c \approx 2.1 \text{ T} \), the system orders antiferromagnetically with moments perpendicular to the applied field. From the field is increased further, the spins cant and a second quantum phase transition occurs at \( H_{\text{sat}} \) with the spins polarized parallel to the applied field. From the resulting phase diagram, bounded by \( H_c(0)=2.13 \text{ T} \), \( H_{\text{sat}}(0)=12.2 \text{ T} \), \( T_{\text{sat}}=1.2 \text{ K} \), and \( g=2.23 \), the excitation gap is calculated as \( \Delta=0.27 \text{ meV} \), where \( T_{\text{sat}} \) is defined as the maximum temperature of the ordered phase.4,6

The obtained parameters for DTN and their crystal symmetry enable this system to have its magnetic properties studied within the picture of BEC of spin degrees of freedom, a bosonic particle with spin \( S=1 \). As DTN, described as a quasi-one-dimensional magnet, has weak interchain interaction, \( J_{\text{perp}}=0.1J \),5 bosons are free to propagate in three dimensions—a requirement for BEC to occur at finite temperature.

Besides DTN, a few other samples were studied in the picture of BEC such as the compounds \( \text{TICl}_3 \)7–9 and \( \text{BaCuSi}_2\text{O}_6 \).10,11

The BEC is associated with bosonic particles at low temperatures which experience a condensation into one quantum state at a temperature \( T_{\text{BEC}} \). At low temperatures the critical density of bosons below which BEC is observed in a gas of weakly interacting bosons in the dilute limit depends on the temperature and mass as12,13

\[
d_c(T) = \kappa \left( \frac{k_B}{2\pi m} \right)^{3/2} m^{3/2} T^{3/2},
\]

where \( \kappa \) is a numerical constant \( \sim O(1) \), and \( m = (m,m,m) \)1/2 is the effective mass, calculated along the three axes in the anisotropic case.7,14 In other words, the power-law dependence on mass and temperature is expected to be the same as that for isotropic noninteracting bosons, while the constant differs only moderately from its value in the noninteracting, isotropic limit \( \sim (g/3)^2 \). By minimizing \( m \), we can observe BEC at relatively high temperatures without sacrificing the condition of low density. In quantum magnets, the boson mass is sufficiently small that BEC can be observed on the order of kelvins. Another matter related to the observation of BEC is the \( U(1) \) symmetry of the system, which is responsible for the conservation of the number of particles. In spin language it corresponds to the existence of a symmetry axis around the magnetic ion. In quantum magnets, BEC can be induced by applying the magnetic field along this direction.1,3,4

The phase in which the system undergoes a magnetic ordering in the picture of BEC was discussed by Nikuni et al.3 Based on the Hartree–Fock–Popov mean field analysis of a hard-core boson Hamiltonian, a constraint \( v_0 \) is introduced to take into account the short-range repulsion between bosons. This constraint is responsible in avoiding the collapse of the particles, so that the condensed bosons are finite at the transition. This quantum phase, in which the bosons become weakly interacting, has a critical field-temperature line \( H_c(T) \) at low temperatures approaching3,4,9

\[
H_c(T) - H_c(0) = AT^\phi,
\]

in which \( \phi=3/2 \) in the limit of zero temperature. For DTN, in Ref. 4 we studied this phase diagram. The results for the

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parameters were \( A = 0.70 \, T/K^{3/2} \) and \( \phi = 1.5 \). This value of the exponent is the first evidence for the identification of this phase with a BEC.

Another fundamental quantity useful for determining the phase diagram is the magnetization \( M \) as a function of temperature \( T \) and magnetic field \( H \). The critical points of the induced phase boundary \((T_c, H_c)\) can be experimentally determined from curves of \( M(T) \) at fixed \( H \), which show a cusplike minimum at \( T_c \), or from \( M(H) \) curves, which show an inflection point at \( H_c \).\(^{3,7,9,11}\)

The applied magnetic field is responsible for driving the system to the BEC phase, providing the creation of bosons at \( H_c, T_c \). The number of created bosons per magnetic ion is given by the density of magnetization \( n(T) = M(T)/M_{\text{sat}} \), where \( M \) is the longitudinal magnetization and \( M_{\text{sat}} \) is its saturated value.\(^3\) At the transition the magnetization acquires its saturated value, there is one created boson per unit site.\(^3\)

In this model the low field boundary of the phase diagram \( H \) vs \( T \) can be expressed as a function of \( n_c \) by\(^3,9\)

\[
H_c(T) - H_c(0) = \left[ 2v_0/g\mu_B \right] n_c(T).
\]

At low temperatures the critical field \( H_c \) in Eq. (2) becomes \( T^{3/2} \) dependent since the density in Eq. (3) may be expressed by \( n \propto T^{3/2} \). The \( 3/2 \) exponent is the low temperature limiting behavior of the boson distribution function, which is only valid in the limit of \( T \ll J \). Since the power law results from the dependence of the critical density of bosons at low \( T \), it is significant to study the phase boundary \( H_c \) directly as a function of \( n_c \), which should be valid over a large temperature range.

The purpose of this work is to investigate, using magnetization data, the characteristics of the low field boundary \( H_c(T) \) of DTN where field-induced ordering appears. As a consequence, we can associate the density of bosons with the interactions \( v_0 \) among them.

The phase diagram of DTN was theoretically studied using different approaches in the BEC picture.\(^{15-17}\) The space group of DTN is \( I4 \), with two molecules in the unit cell. The \( S=1 \) \( \text{Ni}^{3+} \) atoms form two interpenetrating tetragonal lattices.\(^{18}\)

The magnetization was measured using a vibrating sample magnetometer in a \( ^3\)He cryostat, placed in a superconductor magnet. The field was applied along the tetragonal axis of the sample, which is a necessary condition to induce BEC. The critical density of magnetization was determined by analyzing magnetization curves measured up to 5 T for several temperatures. These experiments were performed at the High Field Magnetic Laboratory facility of the University of Sao Paulo.\(^5\)

In Fig. 1 plots of magnetization data are shown for different temperatures between 0.5 and 1.0 K. The magnetization is almost zero in the gapped phase and starts to increase rapidly above the field \( H_c(T) \), at which long-range order takes place. The transition \( H_c \) was determined from the peak in \( d^2M/dH^2 \), which corresponds to the inflection point in \( dM/dH \),\(^3\) as shown in Fig. 2. This inflection becomes less pronounced with increasing temperature. The slope reduction of \( dM/dH \) above the critical field \( H_c \) may be a consequence of quantum fluctuations to the uppermost spin level, which can be calculated using quantum Monte Carlo techniques.\(^5\)

The critical field \( H_c \) as a function of \( n_c \), plotted in Fig. 3(a), follows the linear relation of Eq. (3). The interaction strength constant \( v_0 \) in Eq. (3) is obtained from the derivative of the fitted curve \( dH_c/dn_c = 9.52 = 2v_0/g\mu_B \) as \( v_0 = 0.61 \, \text{meV} \). The zero temperature transition field is obtained as \( H_c(0) = 2.15 \, \text{T} \), in close agreement with Ref. 4, showing the validity of the linear behavior in the whole temperature range. Using the value of \( A = 0.70 \, T/K^{3/2} \), as obtained from the phase diagram at low temperature in Ref. 4,
and $B=9.52$, we can use Eqs. (2) and (3) simultaneously to obtain the derivative $dnc/dT^{3/2}=A/B=0.073$, which is plotted in Fig. 3(b) as the straight line. The continuity of this line at low temperature, with the experimental points at high temperature, is a robust indication of the coherence of the theory used to join results from different experimental data.

An analysis of the derivative $dM/dH$ in Fig. 2 shows a sharp kink at $H_c$. In a recent report, the possibility of a Dzyaloshinskii–Moriya (DM) interaction in DTN was proposed to explain the extra lines in electron spin resonance experiments. This anisotropic interaction between corner-center coupling spins at sites $i,j$ in the body-centered tetragonal lattice of DTN is given by $-d\cdot(S_i\times S_j)$, where $d$ is a specific vector coefficient. If a DM interaction exists with the vector $d$ pointing away from the tetragonal axis, which would break the $U(1)$ symmetry, then the phase transition at $H_c$ in the magnetization would be broadened due to a non-uniform field distribution in the sample. On the other hand, if a DM interaction exists with the vector $d$ pointing along the tetragonal axis, then the $U(1)$ symmetry required for BEC is preserved. Traces of $M$ vs $H$ are the best for analyzing the configuration of the vector $d$ in DTN. As seen in Fig. 2, we observe a unique and sharp transition at $H_c$. Thus we conclude from $M(H)$ data that the energy scale of any $U(1)$ symmetry-breaking interactions is negligible at the measured temperatures.

In summary, we study the magnetization of DTN near the critical field where the excitation gap closes and a BEC of spin degrees of freedom is supposed to occur. Our analysis allows us to calculate the boson interaction strength $v_0=0.61$ meV, whose small value is compatible with a dilute boson system. We also address a question about the existence of $U(1)$ symmetry in this system, showing that it is preserved. These results are in accord with the picture of BEC for the ordered induced phase in DTN.

This work was supported in part by the Brazilian agencies CNPq and FAPESP. LANL is supported by U.S. DOE under Contract No. W-7405-ENG-36. NHMFL is supported by the DOE, the NSF, and the state of Florida.


![Graph showing critical transition field and critical density of magnetization](image_url)