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Critical properties of coupled anisotropic Haldane spin chains in a magnetic field

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We use inelastic neutron scattering to measure magnetic fluctuations as a function of transverse magnetic field and temperature in a single crystal of the coupled Haldane chain compound $SrNi_2V_2O_8$ with uniaxial anisotropy. At the base temperature (2 K), spin excitations confirm a field-induced quantum phase transition from the disordered Haldane (gapped singlet) state to a gapped long-range ordered state through a quantum critical point (QCP) ($\mu_0 H_c \approx 11.5$ T). At elevated temperatures in the vicinity of the QCP, both the energy and linewidth of the lowest triplet mode scale linearly with temperature, in accordance with quantum criticality. We also demonstrate that the experimental field dependence of the triplet excitation modes at the base temperature is in agreement with the bosonic and macroscopic models, but is in contrast to the fermionic and perturbative models.

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Field-induced phenomena in quantum spin systems are of special research interest in recent years. The discovery of field-induced long-range order (LRO) [1,2], magnetization plateau [3], novel quantum states [4], realization of hidden rare symmetry [5], etc., has enriched the research in this field over the last decades. In particular, the realization of Bose-Einstein condensation (BEC) in quantum magnets gives an alternative route to understanding the physics behind many-body quantum systems [6]. Of particular interest is the gapped one-dimensional (1D) spin systems under a magnetic field. In zero field these systems have a disordered magnetic ground state even at $T = 0$ K due to strong quantum fluctuations. In general, a sufficiently strong external magnetic field can close the energy gap by suppressing the zero point fluctuations and drive the system into a new gapless phase. The low-energy physics of the gapless phase is similar to that of the intrinsically gapless $S = 1/2$ Heisenberg antiferromagnetic (AFM) chain and can be described by relativistic field theory, also known as the Tomonaga-Luttinger liquid (TLL) theory [7]. At zero temperature, the field-induced disorder-order transition is continuous [through a quantum critical point (QCP)] and can be described as a quantum phase transition (QPT). At nonzero temperature, it extends to a broad V-shaped region of quantum criticality where the physical properties show universal features independent of microscopic details [8].

In this Rapid Communication we report field-induced behaviors of the coupled spin-1 Heisenberg chain (also known as the Haldane chain) compound $SrNi_2V_2O_8$ with uniaxial anisotropy. In zero field, $SrNi_2V_2O_8$ has a disordered spin-singlet ground state and gapped triplet excitations [9]. An applied magnetic field results in a QPT to the three-dimensional (3D) ordered state with two critical fields ($\mu_0 H_c^{\perp c} \approx 12.0 \pm 0.2$ T and $\mu_0 H_c^{\parallel c} \approx 20.8 \pm 0.5$ T, respectively, at 4.2 K) [10]. Both the phase boundaries for $H \parallel c$ and $H \perp c$ follow power law behavior with exponents 0.57 and 0.43, respectively [9]. In the present study we have employed single crystal inelastic neutron scattering (INS) as a function of transverse magnetic field and temperature to map the evolution of the spin dynamics. The measured data clearly show that different dynamical behaviors belong to different magnetic phases. Our data also reveal universal scaling behaviors in the quantum critical regime in the vicinity of the QCP, where both the energy and linewidth of the lowest triplet mode scale linearly with temperature. Our detailed measurements unambiguously reveal that the field dependences of the triplet states at the base temperature are well estimated by the bosonic and macroscopic model over the full field range, but do not agree with the fermionic and perturbative models. The experimental phase diagram of $SrNi_2V_2O_8$ in the H-T plane is found to be in good agreement with the theoretical phase diagram predicted for gapped 1D systems.

The screw chains in $SrNi_2V_2O_8$ are formed by edge-shared NiO_6 octahedra along the crystallographic c axis. All magnetic Ni^{2+} ions ($3d^8$, $S = 1$) are equivalent within a unit cell. The screw chains are separated by nonmagnetic VO_4 (V^{5+}; $3d^0$, $S = 0$) tetrahedra [9]. The dominant interaction in $SrNi_2V_2O_8$ is the nearest-neighbor intrachain interaction ($J = 8.7$ meV). The presence of multiple competing interchain interactions ($J_\perp^{\text{eff}}/J \approx 0.03$) leads to complicated low-energy excitations where the Haldane gap is replaced by three energy minima at different AFM zone centers [11], each of them split into two modes by the uniaxial anisotropy ($D = -0.32$ meV) [11].

Single crystals of $SrNi_2V_2O_8$ were grown by a traveling solvent floating zone technique [12]. INS spectra were measured on a cylindrical single crystal (diameter ≈ 6 mm and length ≈ 30 mm; mass ≈ 2.5 g) by using the cold neutron triple axis spectrometer PANDA at Heinz Maier-Leibnitz Zentrum (MLZ), Garching, Germany. A double focusing pyrolytic graphite monochromator and analyzer were used and the final energy was fixed at 5.1 meV ($k_f = 1.57$ Å^{-1}) with a cooled beryllium filter after the sample to remove higher order neutrons. The temperature and field dependent measurements were performed using a commercial Oxford 15 T vertical magnet. The zero-field temperature dependent measurements were performed using a closed cycle refrigerator.

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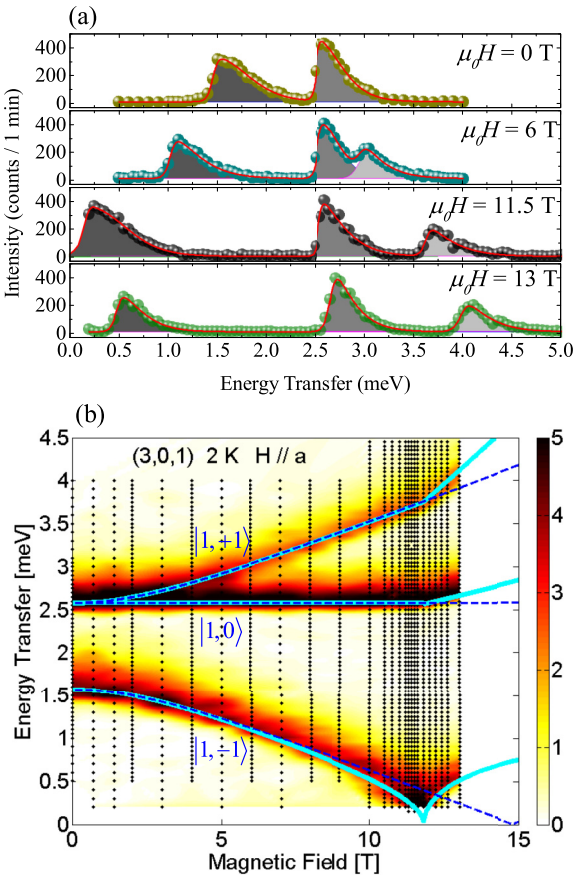

FIG. 1. (Color online) Field dependence of triplet states at 2 K. (a) Constant-Q energy scans at the AFM zone center (3,0,1) under $\mu_0 H = 0$, 6, 11.5, and 13 T at 2 K. The individual contributions of the three excitation modes are shown by the dark, medium, and light shaded regions. The thick red curves through the experimental data points are the sum of the three contributions. (b) Color map of the field dependence. The thick solid cyan lines and the thin dashed blue lines are the calculated field dependence as per the bosonic/macroscopic and perturbative/fermionic models, respectively (see text for details).

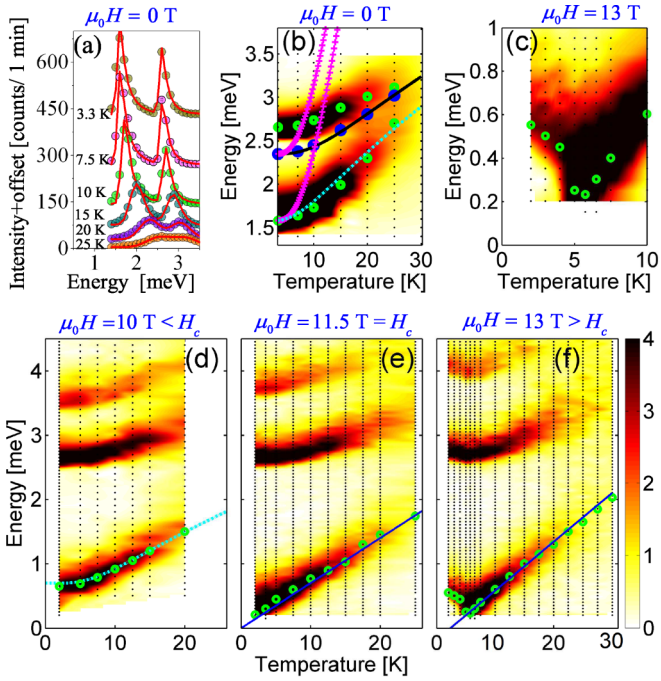

FIG. 2. (Color online) Temperature dependence of the triplet states at the AFM zone center (3,0,1). (a) Constant-Q scans at different temperatures (symbols) in zero field. The data sets are shifted vertically for clarity. Solid red lines are fits to the data, as described in the text. Color maps of the temperature dependence under (b) 0 T, (d) 10 T, (e) 11.5 T, and (c) and (f) 13 T. The green circular points are the fitted peak positions. The blue points in (b) are the mean values of the triplet sublevels expected without anisotropy. The cyan dashed-dotted and solid black lines are the fits to the lowest triplet level and the mean values, respectively, by Eq. (1). The temperature dependence of the energy gap as per the nonlinear sigma model (NLσM) [22] is shown by the pink crosses in (b). The dashed line in (d) is the fitted curve as per Eq. (1). The solid curves in (e) and (f) are the linear fits. (c) An enlarged view of the 13 T data over low temperatures and low energies.

The low-energy excitation spectra (constant-Q scans) of $SrNi_2V_2O_8$ at an AFM zone center (3,0,1) are shown in Fig. 1(a) for transverse magnetic fields 0, 6, 11.5, and 13 T ($H \parallel a$; perpendicular to the easy axis along the c axis). First, the system is characterized in zero field [top panel of Fig. 1(a)]. In agreement with previous studies [10], two distinct branches of gapped excitations (magnons) are found at 1.56 ± 0.1 and 2.58 ± 0.1 meV, respectively. The two peaks correspond to the splitting of the triplet states into a longitudinally polarized low-energy singlet and a high-energy transverse-polarized doublet by the uniaxial single-ion anisotropy [11]. The contribution of the individual modes was estimated by the fits to the line shapes that took account of the instrumental resolution, the energy gap, and the local curvature of the mode dispersion. For the field and temperature dependent measurements, the AFM zone center (3,0,1) was selected where the intensities of both modes are almost equal.

All the data collected at different fields, including those plotted in Fig. 1(a), are combined in a 2D color plot in Fig. 1(b). With the application of magnetic field, the triplet states split further into three distinct branches ($|1,-1\rangle$, $|1,0\rangle$, $|1,+1\rangle$ states) due to the Zeeman effect. With increasing magnetic field, the lowest singlet mode ($|1,-1\rangle$) shifts to the lower energy and becomes gapless at the critical field $\mu_0 H_c \approx 11.5$ T, resulting in a QPT. The modes $|1,-1\rangle$ and $|1,+1\rangle$ show nonlinear field dependence, while $|1,0\rangle$ is field independent. Upon further increasing magnetic field above H_c, an energy gap reappears and the lowest-energy mode shows a parabolic type field dependence. Both the higher-energy modes show an upturn above H_c.

The temperature dependence of the triplet states in zero field is depicted in Figs. 2(a) and 2(b). With increasing temperature, the energies of the triplet states increase monotonically and show an activation type behavior. The inelastic peaks broaden progressively with increasing temperature and could not be observed above ~30 K. The temperature dependences of the triplet states under magnetic fields of 10, 11.5, and 13 T (below, at, and above the H_c) are depicted in Figs. 2(d)–2(f), respectively. At $\mu_0 H = 10$ T ($H < H_c$), all the states show nonlinear temperature dependence similar to the zero-field case. At the critical field $\mu_0 H_c = 11.5$ T, the energy of the

lowest mode increases linearly with increasing temperature, whereas the two higher-energy modes retain the nonlinear behavior. At $\mu_0 H = 13$ T (in the 3D ordered state), the energy of the lowest mode first decreases rapidly with increasing temperature and becomes zero at ≈5.5 K (corresponding to T_N) [Fig. 2(c)], then show a linear temperature dependence above T_N. The higher two modes decrease first with increasing temperature and then show a nonlinear behavior at higher temperatures.

The field dependence of the triplet states at the base temperature (2 K) was compared to various theoretical models. Several theoretical approaches have been made to explain the field dependence of the Haldane chains in the presence of single-ion anisotropy. First, a macroscopic field theory method (*bosonic model*) was proposed by Affleck [13,14] based on the O(3) nonlinear sigma model (NLσM). In this model, a three-component bosonic field was introduced with a quartic potential and was treated in the Gaussian approximation. The anisotropy was introduced by assuming three different masses (three different gap values Δ_α, $\alpha = 1,2,3$) for the three-field components. The second theory was proposed by Tsvelik [15] (*fermionic model*) where the spin-1 Heisenberg chain is described in terms of a field theory of three right- and left-moving Majorana fermions. This model was successfully used to describe the field dependencies of the triplet states below the H_c of the Haldane chain compound $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ (NENP), which has planar anisotropy. The third theory, proposed by Golinelli *et al.* [16], uses a perturbation calculation for the singlet phase (*perturbative model*). The perturbative model predicts similar field dependences of the triplet states and exactly the same critical field values as the fermionic model. Another macroscopic theory (*macroscopic model*), considering a general case of a spin-gap magnet with the field-induced 3D ordering, was developed by Farutin *et al.* [17]. The field dependences of the triplet sublevels and the critical fields predicted by this theory are identical to the bosonic model.

The field dependences of the triplet states are calculated by both the bosonic/macroscopic model (thick solid cyan lines) [17] and fermionic/perturbative model (the dashed blue lines) [16] and compared with the present experimental data in Fig. 1(b). For these calculations, we fixed the zero-field gap values to 1.57 and 2.58 meV and used the g value of 2.24 [18]. It is clearly evident that the bosonic/macroscopic model reproduces the observed behavior well all over the whole field range. However, the fermionic/perturbative model significantly deviates from the data near the H_c and results in a different critical field of $\mu_0 H_c = \sqrt{\Delta_1 \Delta_2}/g\mu_B \approx 15$ T. It is also worth mentioning that, in contrast to our experimental data, the fermionic model does not predict any change of slope at H_c for the upper two triplet modes (Fig. 1). The models are, however, indistinguishable in the low field region ($\mu_0 H < 7$ T). In agreement with the present observations, the field dependences of the isostructural compound $PbNi_2V_2O_8$ were also described by the bosonic/macroscopic model [19]. On the other hand, for most of the studied Haldane chain compounds, which have planar anisotropy, the fermionic model estimates the gap values more accurately [20,21].

The reappearance of the energy gap above H_c indicates long-range anisotropic magnetic ordering. This is consistent with our previous magnetization and specific heat study which revealed 3D magnetic ordering above H_c [10]. The field-induced 3D ordered state for $H > H_c$ is further evident from the temperature dependence of the triplet states in the present study, where with increasing temperature the energy gap decreases and becomes zero at T_N. These results are in contrast to the theoretical prediction of the absence of long-range order and gapless excitations of the TLL state applicable to uncoupled Haldane chains without anisotropy. The three well-defined excitation modes in the ordered state ($H > H_c$) are in contrast to two Goldstone modes in conventional long-range ordered states. Therefore, at least one of the three modes must be polarized along the ordered moment direction (longitudinal magnon). Thus, quantum-mechanical effects remain significantly important even in the ordered state, which may be due to small ordered moment values.

The effects of temperature on the triplet states are discussed below. In zero field, at low temperatures ($T \to 0$), the gapped magnon excitations in the Haldane chains are long lived since mutual collisions between them are rare due to their exponentially small density. However, at elevated temperatures, the population of magnons is significant and hence interactions between them become important. The interactions result into a reduction of magnon lifetimes and a renormalization of their energies. The quantum NLσM predicts activation behavior of the Haldane gap with temperature [22]. This model agrees reasonably well with the experimental temperature dependence of the Haldane gaps in $CsNiCl_3$ and NENP at low temperatures ($T < \Delta$) [23,24]. However, this model is clearly inconsistent with our experimental data [pink crosses in Fig. 2(b)] over the full temperature range. Such inconsistencies were also reported for the Haldane chain compound $Ni(C_3H_{10}N_2)_2N_3(ClO_4)$ (NINAZ) [25]. The temperature dependence of NINAZ was successfully modeled by an empirical modification of the NLσM expression by replacing the prefactor $2\pi\Delta(0)$ by a refinable variable α [25],

$$\Delta(T) = \Delta(0) + \sqrt{\alpha T}\exp\left(\frac{-\Delta(0)}{T}\right), \qquad (1)$$

where the $\Delta(0)$ is the Haldane gap at $T = 0$ K. Equation (1) reproduces the experimental data [Fig. 2(b)] well. The best fitting parameters are $\alpha = 0.14$ (meV/K)^2 and $\Delta(0) = 1.58$ meV for the lowest mode, and $\alpha = 0.105$ (meV/K)^2 and $\Delta(0) = 2.35$ meV for the mean energy mode [$\Delta_{\text{mean}} = (0.57\Delta_+ + 1.41\Delta_-)/1.98$], respectively. Both α values are significantly smaller than that of the respective theoretical values of $\alpha = 2\pi\Delta(0) \approx 9.9$ [22] and ≈14.76 (meV/K)^2, respectively. At 10 T ($H < H_c$), the temperature dependence is also reproduced by Eq. (1) [Fig. 2(d)] with $\Delta(0) = 0.7$ meV and $\alpha = 0.14$ (zero-field value).

In contrast to the activation type behavior for $H < H_c$, the striking linear temperature dependence of the lowest triplet mode at and above the H_c [Figs. 2(e) and 2(f)] is observed. At the critical field $\mu_0 H_c = 11.5$ T, the lowest triplet mode follows a linear temperature dependence with a slope ∼0.07 meV/K [Fig. 2(e)]. The linear temperature dependence ($dE/dT \sim 0.076$ meV/K) is also found for $\mu_0 H = 13$ T ($H > H_c$) above $T_N \approx 5.5$ K [Fig. 2(f)]. These results reveal that the energy of the lowest mode scales linearly with the temperature [$E(T) \approx k_B T$] in the vicinity of the QCP (H_c).

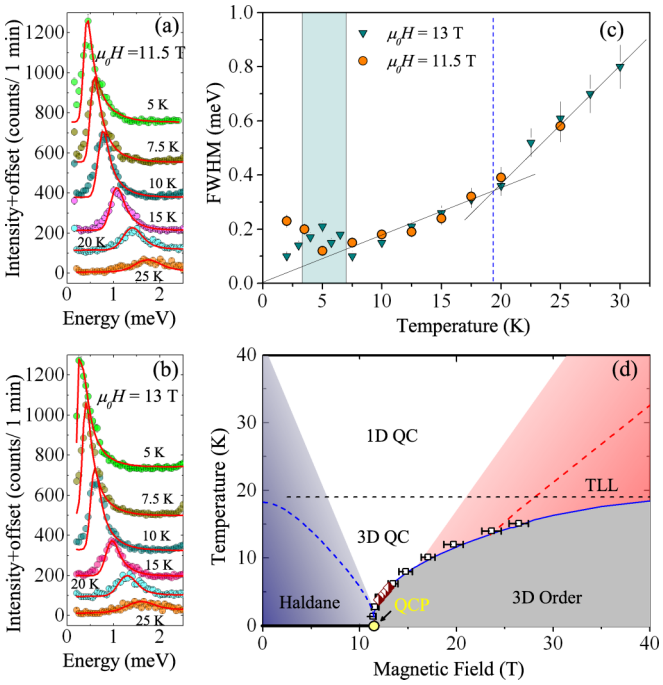

FIG. 3. (Color online) Constant-Q scans over the lowest excitation mode at different temperatures (symbols) under (a) 11.5 and (b) 13 T. Solid red lines are fits to the data, as described in the text. The data sets are shifted vertically for clarity. (c) The linewidth [full width at half maximum (FWHM)] of the lowest excitation mode as a function of temperature at the critical field 11.5 and at 13 T. The shaded region represents the transition region between the 3D ordered state and the QC state. (d) The experimental H-T phase diagram for the Haldane chain compound $SrNi_2V_2O_8$. The solid and dashes curves represent the ordered-disorder phase boundary and the crossover regions, respectively.

Therefore, the intrinsic energy scale of the system is energy over temperature (E/T, the universal scaling parameter) rather than these two quantities separately, which is a signature of a quantum critical (QC) state [26,27]. The scaling behaviors are further investigated by extracting the linewidths (Γ) from the measured excitation spectra. The linewidth was also found to scale with temperature ($\Gamma = \alpha T$) [Fig. 3(c)], revealing that the critical damping is an essential property of the quantum critical excitations [27]. The change of slope ($\alpha = d\Gamma/dT$) around 19 K is due to a crossover from the 3D QC state to the 1D QC state. This also implies that the weakly coupled spin chains feature well-separated temperature scales relevant for the physics in 3D and 1D, as predicted by a recent theory [28].

The experimental phase diagram is shown in Fig. 3(d), and reproduces the theoretically predicted phases for quasi-1D gapped spin systems [28]. The experimental points from the present study are shown by the half-solid diamonds. Open square data points are from our previous high-field magnetization study [10]. Between the disordered Haldane and the field-induced long-range ordered states, the dominant behavior is quantum critical (E/T) scaling. When the temperature is larger than the interchain interactions ($k_BT > J_{3D}$), the system behaves as a set of independent rather than coupled chains and features a different rate of critical slowing down, resulting in separate temperature scales for 1D QC and 3D QC states. The expected phase boundary for the TLL phase [$T_c \sim 0.76(H - H_c)$] [29] is shown by the red dashed line.

In summary, our single crystal INS investigation of the coupled Haldane chain compound $SrNi_2V_2O_8$ with uniaxial anisotropy at 2 K reveals a field-induced QPT with a QCP at $\mu_0 H_c \sim 11.5$ T. The field dependences of the triplet states are in quantitative agreement with the bosonic and macroscopic models, but are inconsistent with the fermionic and perturbative models. Detailed field and temperature dependent measurements reveal quantum critical behaviors in the vicinity of the QCP. Universal scaling is found in the QC regime, where both the energy and linewidths of the excitations scale linearly with temperature. Further, the temperature dependent spin dynamics in the QC regime reveals a crossover between 3D QC and 1D QC states at ~19 K.

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