Field-induced order-disorder transition in the quasi-one-dimensional anisotropic antiferromagnet $BaCo_2V_2O_8$

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Order-disorder transition is observed in the quasi-one-dimensional anisotropic spin chain antiferromagnet $BaCo_2V_2O_8$ in magnetic fields along the magnetic easy c axis below the Néel temperature. We construct the phase diagram in the H-T plane through magnetic and heat capacity measurements and find that the phase boundary of the order-disorder transition can be described in terms of a mean-field power law. We also suggest that quantum critical behavior from three-dimensional Néel ordering back into a one-dimensional disordered phase is observed in magnetic fields above a critical value of 4.4 T.

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Studies of one-dimensional (1D) antiferromagnetic (AF) spin chain systems have been actively done in condensed matter physics due to the discoveries of various fascinating magnetic phenomena. It is well known that an ideal 1D AF spin system does not show long-range ordering (LRO) above $T=0$ K due to strong quantum spin fluctuation, while weak interchain interaction induces three-dimensional (3D) AF-LRO at the ground state of almost all actual quasi-1D spin chain systems.[1] In fact, the 1D quantum disorder to 3D AF-LRO transition has been observed in nonmagnetic ion doped 1D spin chain systems with a spin gap at low temperatures.[2–7] On the other hand, a theoretical study suggested that 3D AF-LRO to 1D quantum disorder transition can be observed in quasi-1D spin chain systems under magnetic field sufficient to suppress interchain interaction, resulting in reentrant behavior from 3D AF-LRO into 1D quantum disordered state.[8] However, such quantum critical behavior has not been observed experimentally in any of materials so far.

In this paper, we first report on field-induced magnetic transition in a 1D spin-3/2 chain antiferromagnet $BaCo_2V_2O_8$ single crystal by means of magnetic and heat capacity measurements. Our experimental results show that an order to disorder transition occurs in $BaCo_2V_2O_8$ when a magnetic field is applied along the magnetic easy axis. We find that the phase boundary of this order-disorder transition can be described by a mean-field power law, and also suggest that the reentrant behavior from 3D Néel AF-LRO back into a 1D disordered phase is observed at a critical magnetic field of 4.4 T. In contrast to impurity effect in 1D spin gap systems, this represents an experimental observation of field-induced reentrant behavior in quasi-1D spin chain systems.

The crystal structure of $BaCo_2V_2O_8$ studied is similar to that of $SrNi_2V_2O_8$ and has tetragonal symmetry of space group I41/acd with $a=12.444(1)$ Å, $c=8.415(3)$ Å, and $Z=8$.[9] All magnetic Co^{2+} ions are equivalent in the arrays of edge-shared CoO_6 octahedra forming a screw-chain structure along the c axis. The screw chains are separated by nonmagnetic VO_4 (V^{5+}) tetrahedra and Ba^{2+} ions, resulting in a quasi-one-dimensional structural arrangement. In our search for 1D spin chain systems using a substitution of magnetic ions for Cu^{2+} in the large spin gap material $BaCu_2V_2O_8$,[10] recently we have found that $BaCo_2V_2O_8$ is a 1D spin chain antiferromagnet with Néel temperature (T_N) of ~5 K and has large magnetic anisotropy.[11]

As reported in Ref. 11, $BaCo_2V_2O_8$ single crystals ($2\times2\times6$ mm^3) were grown by a spontaneous nucleation method using high purity reagents of $BaCO_3$ (4N), $CoC_2O_4\cdot 2H_2O$ (3N), and V_2O_5 (4N) as starting materials. The orientations of crystal surfaces were confirmed using x-ray Laue backscattering analysis. dc magnetic susceptibility was measured in various applied magnetic fields at temperatures from 300 K to 2 K. Magnetizations were measured in applied fields up to 9 T at low temperatures and heat capacity was measured by a relaxation method. All of the above measurements were performed using a quantum design physical property measurement system (PPMS).

Figure 1 shows the temperature dependences of the magnetic susceptibility of a $BaCo_2V_2O_8$ single crystal in a magnetic field of 1 T parallel ($\chi_\parallel$) and perpendicular ($\chi_\perp$) to the c axis. A broad peak in the $\chi_\parallel$ around 30 K is a clear sign of 1D short-range ordering and a rapid decrease in the susceptibility below ~5 K indicates a long-range magnetic transition. A sharp peak is also seen in the $\chi_\perp$ at ~5 K, clearly showing that the magnetic transition at ~5 K is associated with an antiferromagnetic long-range ordering. Also, a small change in the $\chi_\perp$ below 5 K indicates that the crystallographic c axis corresponds to the magnetic easy axis. A large

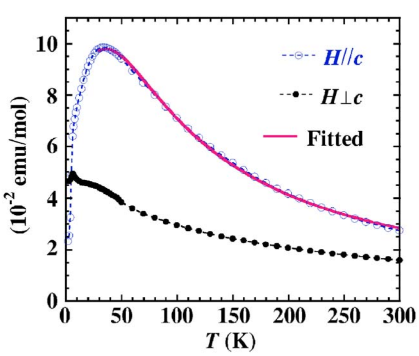

FIG. 1. (Color online) Magnetic susceptibilities in $H=1$ T parallel ($\chi_\parallel$) and perpendicular ($\chi_\perp$) to the c axis. The solid line is a fit based on 1D $S=3/2$ linear chain model.

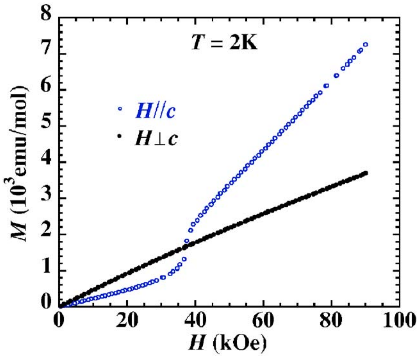

FIG. 2. (Color online) Magnetization versus applied field at 2 K.

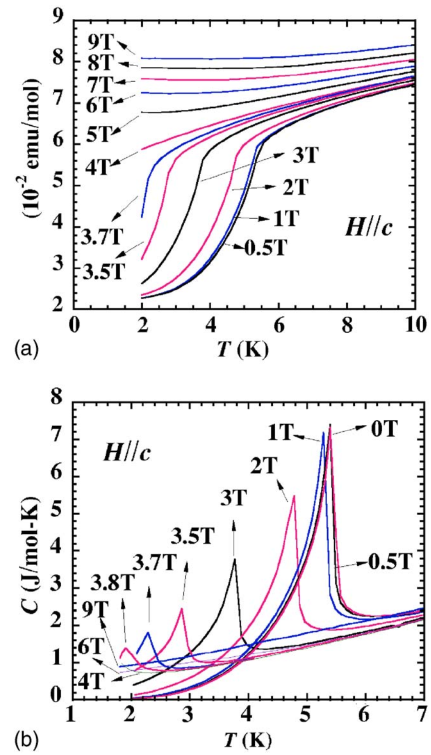

FIG. 3. (Color online) (a) Low temperature susceptibilities measured in different fields from 0.5 T to 9 T parallel to the c axis, and (b) low temperature heat capacity data measured in different fields.

difference between $\chi_{\parallel}$ and $\chi_{\perp}$, which persists even up to room temperature, is an evidence for large anisotropy in the system. In addition, $\chi_{\parallel}$ above 30 K can be well fitted using 1D spin-3/2 linear chain model[12] with $J/k_B = -5.46(0)$ K and $g = 4.37(8)$, the value of which also supports large anisotropy in the system. From these structural and magnetic data, $BaCo_2V_2O_8$ is deduced to be a quasi-1D spin chain antiferromagnet with $T_N = \sim 5$ K and has the magnetic easy axis along the c axis with large anisotropy.

The anisotropic antiferromagnetic feature of $BaCo_2V_2O_8$ is also characterized through field-dependent magnetization measurements along the c axis below 5 K. Figure 2 displays the magnetization as a function of applied field H parallel and perpendicular to the c axis at 2 K. An abrupt increase in the magnetization is observed at around 4 T for $H \parallel c$, indicating field-induced magnetic transition, while no magnetization jump is seen for $H \perp c$ even up to 9 T. We also note that no hysteresis and remanent magnetization are seen in zero field, and the magnetization does not saturate up to 9 T.

The magnetic susceptibilities at different fields are shown in Fig. 3(a). The temperature where a rapid drop in the magnetic susceptibility (T_N) occurs shifts toward lower temperature with increasing magnetic field and the drop disappears completely when the field is larger than 4 T. These magnetic features in Figs. 2 and 3(a) are reminiscent of spin-flop transition in an anisotropic antiferromagnet as predicted by Néel.[13] However, heat capacity data shown in Fig. 3(b) clearly rule out the possibility of spin-flop transition.

The heat capacity measured at low temperatures down to 1.8 K shows a λ-like peak and the temperature showing the peak decreases rapidly with increasing magnetic field and then disappears above 1.8 K in $H = 4$ T, being in good agreement with the susceptibility data shown in Fig. 3(a). If the feature of the heat capacity is associated with a spin-flop process, there should be also another transition from the spin-flop phase to a forced ferromagnetic phase: another peak of the heat capacity should appear when the temperature is raised at $H > 4$ T. However, no other feature in the temperature-dependent heat capacity is seen up to room temperature, indicating that an order-disorder transition (AF to paramagnetic transition) likely occurs in a quasi-1D spin chain anisotropic antiferromagnet $BaCo_2V_2O_8$ single crystal, instead of spin-flop transition when magnetic field is applied along the magnetic easy c axis.

Investigations of field-induced order-disorder transition have been carried out in the last decades.[14–16] It is found that the phase boundary of field-induced order-disorder transition can be described in terms of the mean-field power law $H/H_c = (1 - T/T_c)^{\beta}$, where H_c is the critical magnetic field, T_c the critical temperature showing the order-disorder transition, and β an exponent representing the phase boundary. We depict the phase diagram in the T-H plane [Fig. 4(a)], constructing from the heat capacity data shown in Fig. 3(b). A correction of demagnetizing effect was made from M vs H (Fig. 2) according to the following equations:

$$H_{\text{in}} = H_{\text{ex}} + H_{\text{d}}, \quad (1)$$

$$H_{\text{d}} = -(N/\mu_0)I, \quad (2)$$

where H_{in} is the internal field, H_{ex} the external field, H_d the demagnetizing field, N the demagnetizing constant, and I the magnetization. It is found that the value of H_{d} is approximately 0.035 T at an external field of 4 T using $N \approx 0.1735$ for our sample ($2 \times 2 \times 6$ mm^3).[17] The data points of the phase boundary [Fig. 4(a)] can be well fitted using the equation $H = H_c(1 - T/T_N)^{\beta}$, showing the critical behavior of field-induced order-disorder (AF to paramagnetic) transition. The fit provides $T_N = 5.4$ K, $\beta = 1/3$, and the critical field H_c of 4.4 T at $T = 0$ K. We again note that no phase boundary other than the order-disorder transition is seen in this diagram, and that the AF-paramagnetic transition occurs by crossing the phase boundary instead of spin-flop transition.

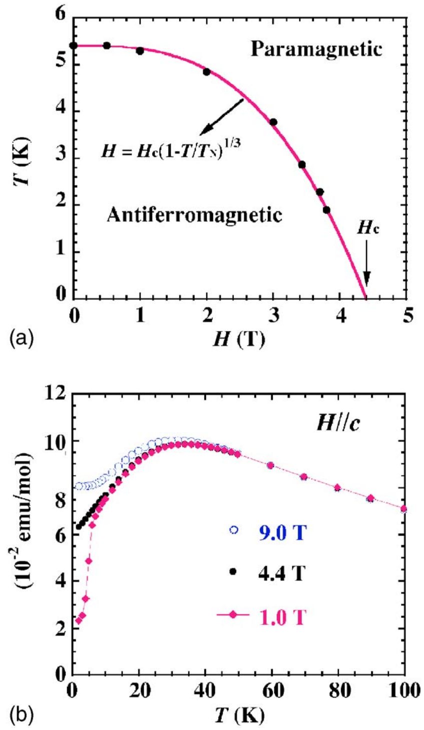

FIG. 4. (Color online) (a) Phase diagram of $BaCo_2V_2O_8$ in the field (H) -temperature (T) plane. Solid line is a fit of $H = H_c(1-T/T_N)^{1/3}$ with $T_N = 5.4$ K and $H_c = 4.4$ T. (b) The c-axis susceptibilities measured at three different fields (1, 4.4, and 9 T) showing a magnetic transition at ~5 K with almost no change of the broad peak feature at ~30 K.

Further evidence for the collapsing behavior of AF-LRO above $T=0$ K is seen in the temperature-dependent magnetic susceptibilities $\chi_{\parallel}$ at three different applied fields, $H = 1$ T ($H < H_c$), 4.4 T (H_c), and 9 T ($H > H_c$) [Fig. 4(b)]. The temperature of the broad peak in the susceptibility does not change under applied fields even up to 9 T, while the susceptibility below 5 K increases with increasing field. It is also noticed that the temperature profile of the susceptibility at 9 T is similar to those of quasi-1D spin gap systems, supporting our description that the 3D Néel AF-LRO is destroyed above $T=0$ K and 1D quantum spin chain character recovers with increasing field above 4.4 T.

In general, magnetic systems in the vicinity of the critical point are characterized by quantum fluctuation as well as thermal fluctuation over various length scales. At high temperatures, the thermal fluctuation is predominant compared with the quantum fluctuation, while quantum fluctuation, in particular in the low-lying states, dominates thermal fluctuation near $T=0$ K and strongly influences the critical behavior. In fact, an ideal 1D spin system does not show LRO above $T=0$ K due to strong quantum spin fluctuation and a quasi-1D spin chain system exhibits 3D LRO at the ground state since weak interchain interaction dominates the quantum spin fluctuation.[1] Our experimental results clearly show that the quasi-1D spin chain system $BaCo_2V_2O_8$ does not display AF-LRO above $T=0$ K under applied fields above 4.4 T parallel to the magnetic easy c axis, indicating the reentrant behavior from 3D AF-LRO back into a 1D quantum disordered phase. Therefore, the quantum critical behavior of 3D Néel LRO back into a 1D quantum disordered phase is indeed realized in the quasi-1D spin chain system $BaCo_2V_2O_8$, presumably due to quantum fluctuation induced by external magnetic field.

Recently, Mikeska, *et al.* theoretically suggested that external magnetic field can induce a reentrant behavior in a gapped spin system with nonmagnetic impurities.[8] In fact, nonmagnetic impurity effect on the gapped spin systems such as 1D double spin chain system $KCuCl_3$ (Ref. 18) leads to a 3D Néel LRO at low temperatures mainly due to interchain (interdimer) interaction.[19] The field-induced reentrant phenomena in the gapped spin systems with nonmagnetic impurities seems similar to that in 1D spin chain system $BaCo_2V_2O_8$, and therefore we suggest that this mechanism could give a possible explanation for the fact that an external magnetic field suppresses interchain (3D) interaction and drives the system back into the disordered phase.

In summary, we have observed a field-induced order-disorder transition in the quasi-1D spin chain system $BaCo_2V_2O_8$ instead of spin-flop transition under external field along the magnetic easy c axis. We have found that the phase boundary of the order-disorder transition can be described in terms of the mean-field power law, $H = H_c(1 - T/T_N)^{\beta}$ with a critical field $H_c = 4.4$ T. We also indicate that the field-induced quantum critical behavior from 3D Néel order back into a 1D quantum disordered phase is observed at magnetic fields above 4.4 T. We envisage that this study of the order-disorder transition and quantum critical behavior in the quasi-1D anisotropic antiferromagnet $BaCo_2V_2O_8$ will stimulate further theoretical and experimental studies of quasi-1D spin chain systems.

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