Antiferromagnetic-paramagnetic transitions in longitudinal and transverse magnetic fields in a $SrCo_2V_2O_8$ crystal

Zhangzhen He,* Tomoyasu Taniyama, and Mitsuru Itoh
Materials and Structures Laboratory, Tokyo Institute of Technology, 4259 Nagatsuta, Midori, Yokohama 226-8503, Japan
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Magnetic and heat capacity studies of the 1D anisotropic antiferromagnet $SrCo_2V_2O_8$ demonstrate field-induced magnetic transitions in longitudinal and transverse magnetic fields. An antiferromagnetic (AF)-paramagnetic (PM) transition is observed instead of spin-flop transition in longitudinal magnetic field parallel to the magnetic easy *c*-axis. The temperature-field (*T*-*H*) phase diagram clearly shows that the AF-PM transition occurs in the same framework for both longitudinal and transverse magnetic fields in $SrCo_2V_2O_8$. This is an experimental observation of such magnetic behaviors in an anisotropic antiferromagnet, irrespective of the field directions.

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Field-induced magnetic transitions in quasi-one-dimensional (1D) antiferromagnetic (AF) spin systems have attracted much interest due to the discoveries of fascinating magnetic phenomena.[1] Field-induced disorder-to-order transition has been observed in isotropic Heisenberg spin gap systems such as $PbNi_2V_2O_8$ (Ref. 2) and $TlCuCl_3$.[3] For anisotropic antiferromagnets, however, the field-induced magnetic transitions are essentially different from those in the isotropic spin systems, depending on the directions of applied fields parallel or perpendicular to the magnetic easy axis.[4,5] It is well known that a spin-flop transition, which accompanies a jump in the magnetization, is observed in a longitudinal field to the magnetic easy axis, while an order-to-disorder transition is observed in a transverse field. Such different magnetic behaviors in the longitudinal and transverse fields have been experimentally investigated on anisotropic antiferromagnets such as $BaCu_2Si_2O_7$,[6] $K_2V_3O_8$,[7] Cs_2CoCl_4.[8] Recently, we found that an order-disorder transition occurs in the 1D anisotropic antiferromagnet $BaCo_2V_2O_8$ in a longitudinal field of 4.4 T along the magnetic easy *c*-axis.[9] However, no magnetic transition was observed in transverse fields up to 40 T,[10] indicating that different magnetic behaviors appear depending on the field direction.

$SrCo_2V_2O_8$ has the tetragonal crystal structure of space group $I41cd$ with lattice constants $a=12.267(1)$ Å, $c=8.424(1)$ Å, and $Z=8$.[11] As shown in Fig. 1, similar to $BaCo_2V_2O_8$, the most prominent structural feature is that all magnetic Co^{2+} ions are equivalent in the arrays of edge-shared CoO_6 octahedra forming a screw-chain along the *c*-axis and the screw chains are separated by non-magnetic VO_4 (V^{5+}) tetrahedra and Sr^{2+} ions, resulting in quasi-one-dimensional arrangement.

In this paper, we report the detailed magnetic and heat capacity measurements of $SrCo_2V_2O_8$ single crystals, showing field-induced magnetic transitions in both longitudinal and transverse fields to the magnetic easy *c*-axis. An antiferromagnetic (AF)-paramagnetic (PM) transition is observed in the longitudinal field instead of spin-flop transition, while an intriguing magnetic transition is also observed in a transverse magnetic field. By constructing the temperature-field (*T*-*H*) phase diagram, we find that the same magnetic transitions occur in both longitudinal and transverse fields in the quasi-1D anisotropic antiferromagnet $SrCo_2V_2O_8$, irrespective of the applied field direction.

Like $BaCo_2V_2O_8$,[12] $SrCo_2V_2O_8$ single crystals were grown by a spontaneous nucleation method using high purity reagents of $SrCO_3$ (4N), $CoC_2O_4\cdot 2H_2O$ (3N), and V_2O_5 (4N) as starting materials and the orientations of the crystal surfaces were confirmed using x-ray Laue back-scattering analysis. Magnetic susceptibilities and magnetizations were measured in various applied magnetic fields up to 9 T and heat capacity was measured using a relaxation method in a Quantum Design Physical Property Measurement System.

Figure 2 shows the susceptibilities of $SrCo_2V_2O_8$ measured in a magnetic field of 1000 Oe parallel ($\chi_\parallel$) and perpendicular ($\chi_\perp$) to the *c*-axis. A characteristic feature of 1D magnetism is seen in $\chi_\parallel$ with a broad maximum at around 30 K and an antiferromagnetic ordering occurs at ~5 K as shown in the inset of Fig. 2. We also note that a rapid jump is observed in both $\chi_\parallel$ and $\chi_\perp$ near 20 K associated with ferromagnetic correlation. The ferromagnetic moment is roughly estimated to be $\sim 0.05\mu_B/Co^{2+}$ in a longitudinal field of 5000 Oe, which corresponds to 1.67% of the full Co^{2+} ions ($S=3/2$) moments aligned along the *c*-axis, suggesting that the system is a canted antiferromagnet. Moreover, a large difference between $\chi_\parallel$ and $\chi_\perp$ is seen above 20 K, showing that the *c*-axis is the magnetic easy axis with

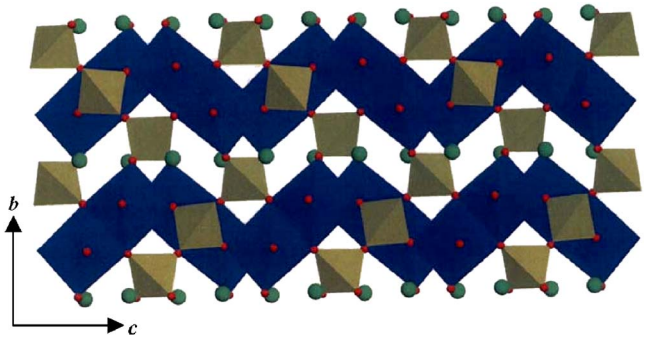

FIG. 1. (Color online) Crystal structure of $SrCo_2V_2O_8$. Octaheadra, tetraheadra, large ball and small ball represent CoO_6, VO_4, Sr, and O, respectively.

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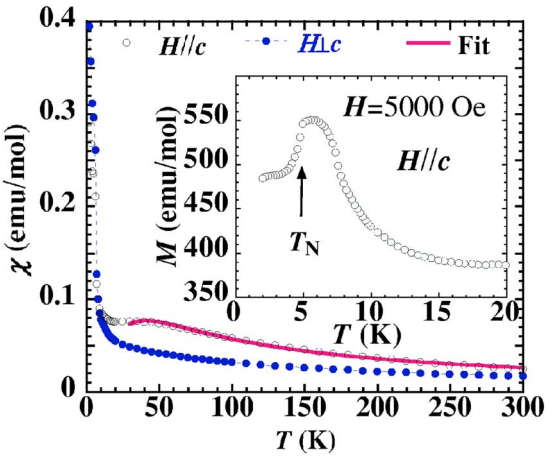

FIG. 2. (Color online) (a) Magnetic susceptibilities in $H=1000$ Oe parallel ($\chi_{\parallel}$) and perpendicular ($\chi_{\perp}$) to the c-axis. The solid line is a fit using a 1D Ising spin chain model for Co^{2+} magnetic systems (Ref. 14). The inset of Fig. 2 shows an antiferromagnetic ordering at ~5 K and the ferromagnetic ordered moment in an applied field of 5000 Oe.

large paramagnetic anisotropy. Further, $\chi_{\parallel}$ above 30 K can also be well fitted using an Ising spin chain model for Co^{2+} (d^7) magnetic systems[13] with $J/k_B=-40.6(4)$ K and $g=9.42(1)$, supporting large magnetic anisotropy in the system. Based on these structural and magnetic features, we conclude that $SrCo_2V_2O_8$ is a quasi-1D canted antiferromagnet with large magnetic anisotropy.

Figure 3 shows the magnetization (M) versus magnetic field (H) curves at low temperatures. For $H\|c$, an abrupt increase in the magnetization, which is reminiscent of spin-flop transition in an anisotropic antiferromagnet,[14] is observed at $T=2$, 3 and 4 K, while an anomaly is also observed for $H\perp c$, indicating an intriguing field-induced magnetic transition. The critical fields of the magnetic transition in $SrCo_2V_2O_8$ decrease with increasing the temperatures form 2 to 4 K for $H\|c$ and $H\perp c$. In addition, no hysteresis and remanent magnetization are seen in zero field, and the magnetization does not saturate up to 9 T. We note that the features in the magnetization of $SrCo_2V_2O_8$ are similar to that of the canted antiferromagnet $K_2V_3O_8$ displaying spin-flop transition and spin reorientation in applied fields parallel and perpendicular to the magnetic easy c-axis, respectively.[7]

To gain more information whether the magnetic nature of $SrCo_2V_2O_8$ in longitudinal and transverse fields is similar to those of $K_2V_3O_8$,[7] the low-temperature magnetic susceptibilities and heat capacities are investigated in various fields. For $H\|c$, a rapid drop in the magnetic susceptibility [Fig. 4(a)], which is associated with the AF transition at the Néel temperature (T_N), shifts toward lower temperatures with increasing magnetic field and disappears completely above 4 T. Also, the heat capacity at low temperatures [Fig. 4(b)] shows a λ-like peak, and the temperature showing the peak decreases with increasing magnetic field, then the peak disappears in $H=4$ T above 2 K, which is in good agreement with the susceptibility data. For $H\perp c$, on the other hand, the peak temperature in the magnetic susceptibility gradually shifts toward lower temperatures with increasing field and

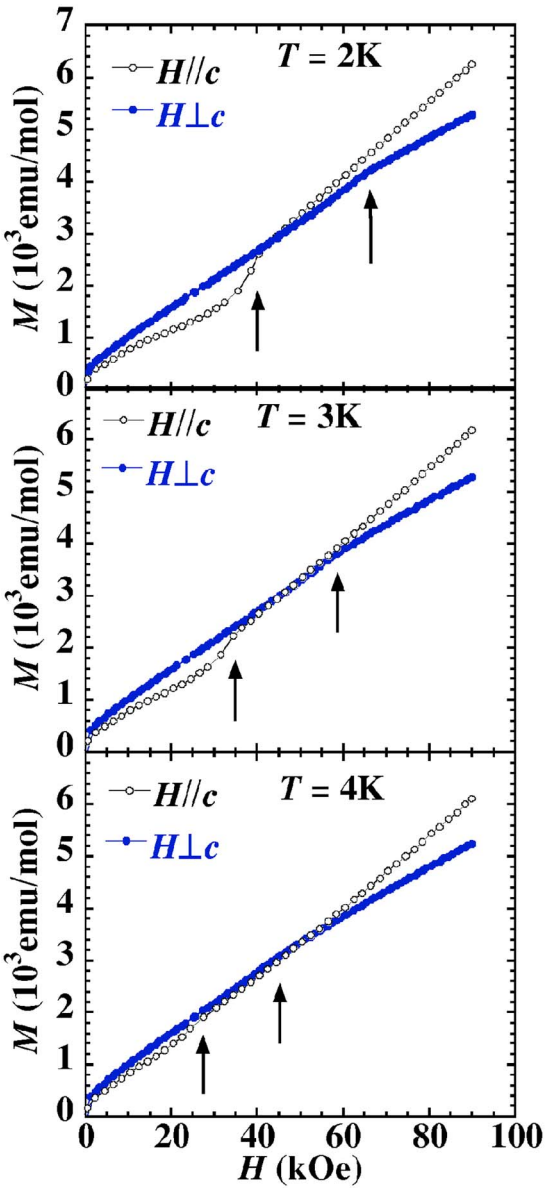

FIG. 3. (Color online) Magnetization versus applied field at different temperatures.

disappears above 7 T [Fig. 5(a)]. The heat capacity for $H\perp c$ is also compatible with the susceptibility data as shown in Fig. 5(b); a λ-like peak shifts towards lower temperature with increasing magnetic field, then the peak disappears in $H=7$ T above 2 K. We note that the features in the susceptibility and heat capacity for both $H\|c$ and $H\perp c$ are similar to each other, indicating that similar field-induced AF-PM transitions likely occur for both field orientations in $SrCo_2V_2O_8$, being different from magnetic transitions in $K_2V_3O_8$.[7]

Further evidence for the AF-PM transition similarly occurring in longitudinal and transverse fields in $SrCo_2V_2O_8$ is shown in Fig. 6. A phase diagram in the T-H plane is constructed from the heat capacity data in Fig. 4(b) and 5(b) with demagnetizing correction using the M vs H curves (Fig. 3) at each temperature: the internal field $H_{in}=H_{ex}-NM$ is used in Fig. 6, where H_{ex} denotes the external field, N de-

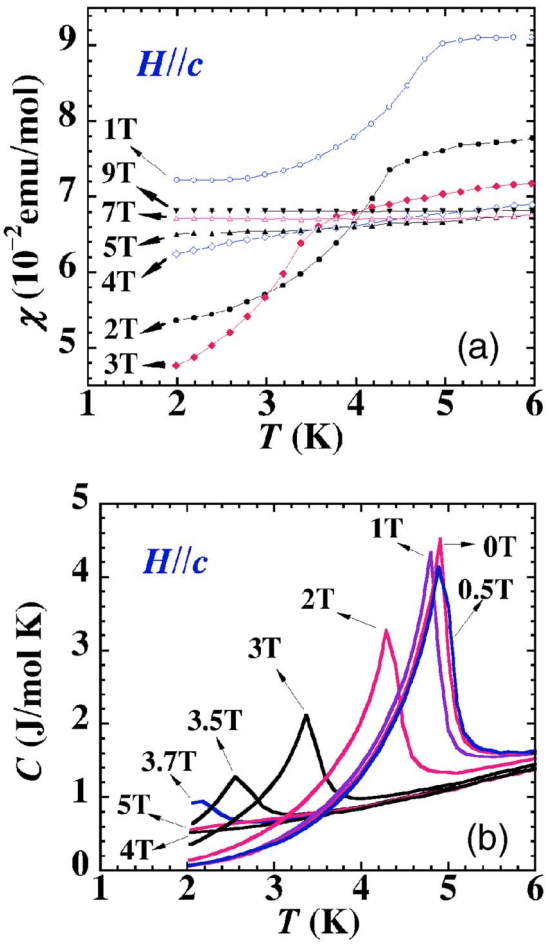

FIG. 4. (Color online) (a) Low temperature susceptibility and (b) heat capacity data measured in various fields parallel to the c-axis.

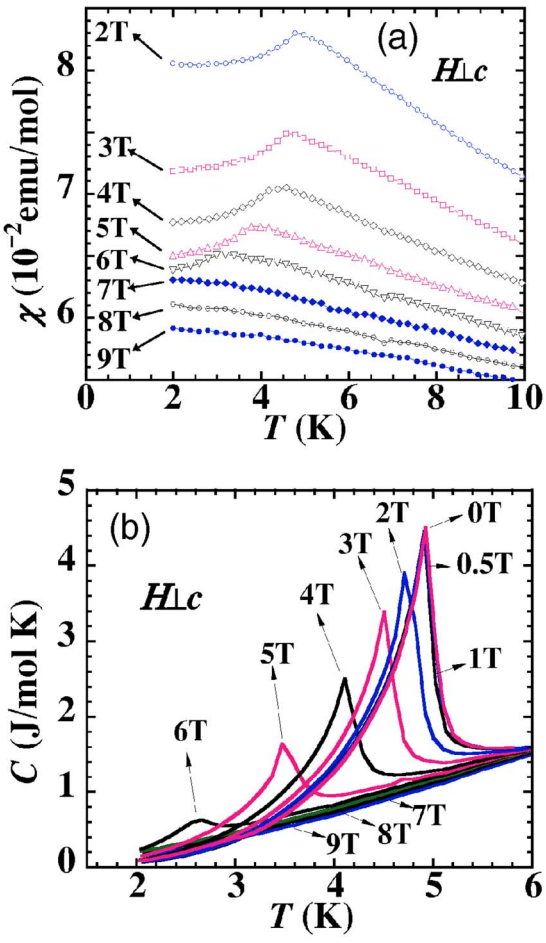

FIG. 5. (Color online) (a) Low temperature susceptibility and (b) heat capacity data measured in various fields perpendicular to the c-axis.

magnetizing constant, and M the magnetization. We find that the data points representing the phase boundary for $H \| c$ and $H \perp c$ can be well fitted using the same equation $H_{in}=H_c(1-T/T_N)^{\beta}$ based on a mean-field power law,[15,16] associated with the critical behavior of field-induced AF to PM transition. The fits (Fit1 and Fit2) provide $T_N=4.8$ K, $\beta=1/3$, and the longitudinal critical field (H_{cl}) of 4.4 T and transverse critical field (H_{ct}) of 7.6 T at $T=0$ K, respectively. Further, the temperature variation of the critical longitudinal field falls on the same curve for transverse field after multiplying a factor of $H_{ct}/H_{cl}=\sim 1.727$, clearly evidences the same magnetic behaviors of AF-FM transition in both longitudinal and transverse fields. In addition, it should also be noted that if magnetic transition is a spin-flop transition, two-phase boundaries, i.e., spin-flop transition and AF-paramagnetic transition, should be seen in the H-T plane. However, no phase boundary other than the AF-PM transition is seen in the diagram for $H \| c$ (Fig. 6), clearly ruling out the possibility of spin-flop transition in the applied fields along the magnetic easy c-axis. This finding also supports that the same magnetic transitions occur in both longitudinal and transverse fields for $SrCo_2V_2O_8$, irrespective of the field direction.

We now discuss why the AF-PM transition occurs instead

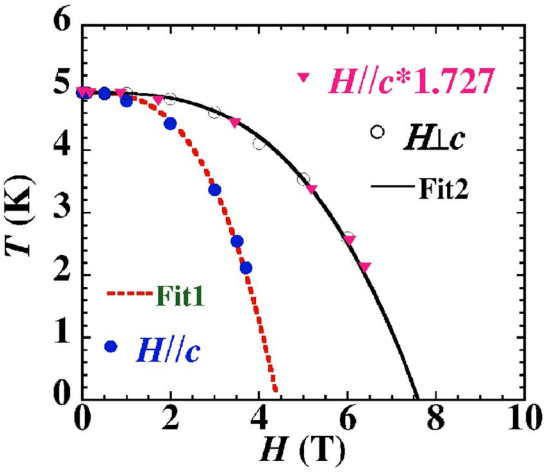

FIG. 6. (Color online) Phase diagram of $SrCo_2V_2O_8$ in the field (H)-temperature (T) plane. Solid lines and dashed lines are fits with $H=H_c(1-T/T_N)^{1/3}$. The triangles denote the data points for fields parallel to the c-axis where the critical fields are multiplied by a factor of 1.727 at each temperature.

of spin-flop transition in a longitudinal field parallel to the magnetic easy *c*-axis on the basis of a simple uniaxial two-sublattice mean-field model. It is well known that spin moments in an anisotropic antiferromagnet tend to flip towards a direction perpendicular to the field direction to gain a magnetic energy of $0.5(\chi_{\perp}\text{-}\chi_{\parallel})H^2$ when magnetic field is applied along the magnetic easy axis. If the magnetic anisotropy is not strong enough, the spin-flop transition appears at a critical field H_{SF} as the gain of magnetic energy can overcompensate the anisotropy energy loss due to the deviation of spin moments from the preferred spin orientation. With a further increase in magnetic field, the moments rotate to the direction of magnetic easy axis gradually until the critical field H_c is reached: the spin moments are parallel to the easy axis and the forced ferromagnetic (paramagnetic) phase is entered. The critical fields of H_{SF} and H_c are related to the antiferromagnetic exchange field H_{AF} and the anisotropy field H_A as follows: $H_{SF}=(2H_{AF}H_A-H_A^2)^{1/2}$ and $H_c=2H_{AF}-H_A$.[17] Therefore, an increase in the magnetic anisotropy increases the spin-flop field H_{SF}, while H_c is reduced; for $H_A=H_{AF}$, the critical fields H_{SF} and H_c become equal. Since $SrCo_2V_2O_8$ have large magnetic anisotropy, spin-flop field could be greater than the AF-PM transition field, leading to the fact that no spin-flop transition occurs and the system enters the paramagnetic phase directly. In other words, sufficiently large anisotropy in $SrCo_2V_2O_8$ causes the same magnetic transitions in longitudinal and transverse fields, irrespective of the applied field direction.

In summary, we have observed field-induced magnetic transitions in the quasi-1D spin chain system $SrCo_2V_2O_8$ under longitudinal and transverse fields by means of magnetic susceptibility and heat capacity measurements. We find that antiferromagnetic-paramagnetic transition is observed instead of spin-flop transition in the longitudinal field to magnetic easy *c*-axis, which is similar to that in the transverse field. Further, the temperature-field (*T*-*H*) phase diagram ensures that the antiferromagnetic-paramagnetic transitions occur in the same framework for both longitudinal and transverse fields. These interesting magnetic transitions in $SrCo_2V_2O_8$, irrespective of the applied field direction, are likely due to its large anisotropy. We envisage that the experimental observation of field-induced magnetic transition in $SrCo_2V_2O_8$ will stimulate further theoretical and experimental studies of quasi-1D anisotropic spin chain systems.

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*Present address: Institute for Solid State Physics, University of Tokyo, Kashiwanoha 5-1-5, Kashiwa, Chiba 277-8581, Japan

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