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Metamagnetic transition in BaVS₃: a highly correlated electron system with frustration

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Abstract

BaVS₃ has the CsCoCl₃-type structure, in which V atoms form linear chains along the *c*-axis and a triangular lattice in the *c* plane. This compound exhibits a metal–insulator transition at $T_{\text{MI}} = 70$ K and an incommensurate antiferromagnetic structure below $T_X = 30$ K. We performed high-field magnetization measurements up to 95 T using a destructive single-turn coil and found a tiny magnetization jump at around 50 T. The origin of this jump is discussed on the basis of proposed models for BaVS₃. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: BaVS₃; Strongly correlated system; Metamagnetism; Frustration

1. Introduction

BaVS₃ is a typical strongly correlated itinerant electron system, which exhibits a metal–insulator (MI) transition at $T_{\text{MI}} = 70$ K (metallic above T_{MI} and insulating below T_{MI}). The crystal structure at room temperature is the hexagonal Perovskite CsCoCl₃-type with the space group P6₃/mmc, in which V⁴⁺ ions with $S = \frac{1}{2}$ form one-dimensional (1D) chains along the *c* direction and a triangular lattice in the *c* plane. Since the intra-chain V–V distance (~ 2.8 Å) is much smaller than the intra-

chain separation (~ 6.7 Å) and nearly equal to the V metallic distance, BaVS₃ has been considered as a prototype for the 1D system. However, conductivity measurements using a single crystal have revealed that the conductivity in the *c* plane is considerable, implying inter-chain electron hopping [1]. Recent neutron scattering experiments showed that the inter-chain antiferromagnetic coupling plays important role, suggesting that the spin frustration in the *c* plane triangular lattice is crucial [2]. Furthermore, the V atom is surrounded by a nearly regular octahedron formed by S atoms, resulting in nearly degenerate t_{2g} states as the crystal field ground state. Below $T_s \sim 240$ K, it shows a small orthorhombic structural deformation, resulting in a slightly zigzag configuration of the V chains [3].

The magnetic susceptibility shows a sharp peak at T_{MI} and drops rapidly in the insulating state below T_{MI} . Although the temperature dependence

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of susceptibility looks like that of an antiferromagnet, a previous precise neutron diffraction experiment failed to detect long-range magnetic ordering [4].

Recently, we have carried out NMR and neutron scattering to understand static and dynamic properties of this compound and obtained mutually exclusive results; we observed low-frequency zero field spin-echo signals, which was interpreted as nuclear quadrupole resonance (NQR), suggesting the orbitally ordered singlet ground state [5] and magnetic Bragg peaks suggesting incommensurate antiferromagnetic spin ordering below 30 K (T_N) [6]. Thus, both the origin of MI transition and the ground-state magnetic properties of BaVS₃ are still hidden in a veil of mystery. In present paper, we report the result of a ultra-high-field magnetization measurement up to 85 T in order to obtain the information on the ground state property of BaVS₃. The high-field magnetization measurements on this compound up to 45 T were reported recently by Booth et al. [7]. Their results suggest monotonic linear increase of magnetization in this field range.

2. Experimental procedures

Polycrystalline powder samples were prepared by heating of a mixture of BaS, V and S in an evacuated quartz tube at 1223 K for 4 days. To avoid reaction of the mixture with quartz tube, carbon crucibles were used. After the prepared samples were ground, they were heated again at 923 K with excess sulfur for 3 days to ensure stoichiometry. Without the last heat treatment, sulfur-deficient samples which show ferromagnetism at low temperatures were obtained. Sample quality was tested by measuring the magnetic susceptibility by a SQUID magnetometer. We obtained the sample which exhibits almost no upturn of the susceptibility at low temperatures, indicating good quality of the sample.

The high-field magnetization was measured by using a destructive single-turn coil installed at ISSP. By means of a fast capacitor discharge, the pulse field up to 85 T was obtained in the duration time of about 10 μ s. The magnetization processes at 4.2 K and at several temperatures below T_{MI} were

measured by an induction method with well-balanced pick-up coils. Approximately, 40 mg of powder sample was used.

3. Results

Fig. 1 shows an example of traces of signals from detection coils of sample magnetization (dM/dt) and magnetic field (H) at 4.2 K. As seen in the figure, a spike-like peak was observed for each increasing and decreasing field processes although the signals near the starting point includes considerable degree of noise signals. This peak suggests a jump of the magnetization curve. The positions of the jump, H_c^{up} and H_c^{down} , are 55 and 45 T for increasing and decreasing processes, respectively, implying a hysteresis of the magnetization curve.

The measurements were performed at elevated temperatures up to 40 K. The spike-like peak was observed in this temperature range although it

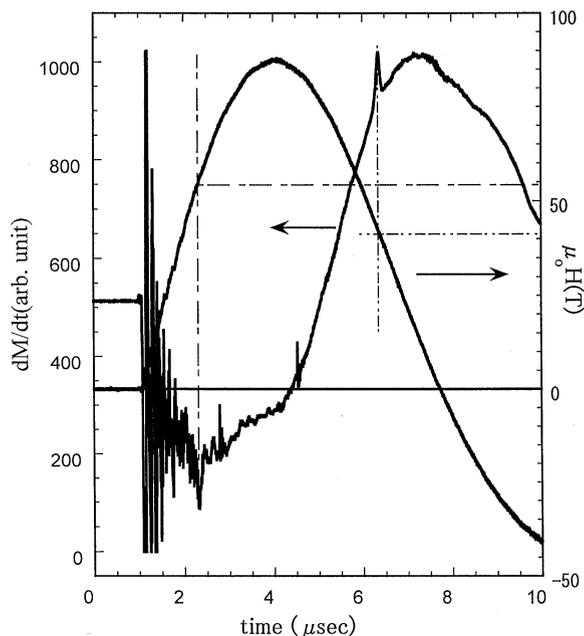


Fig. 1. Traces of signals from detection coils of sample magnetization (dM/dt) and magnet field (H) as functions of time duration at 4.2 K. Vertical lines indicate the positions of spike signals indicating magnetization jump for increasing and decreasing fields processes. Horizontal lines indicate the field strength of each spike.

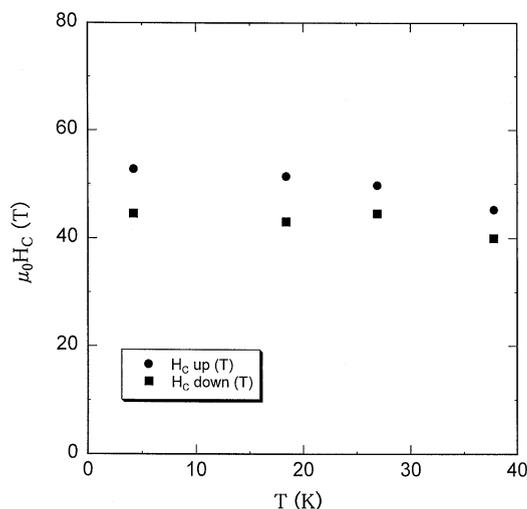


Fig. 2. Temperature dependence of the critical fields of BaVS₃ for field up and down processes.

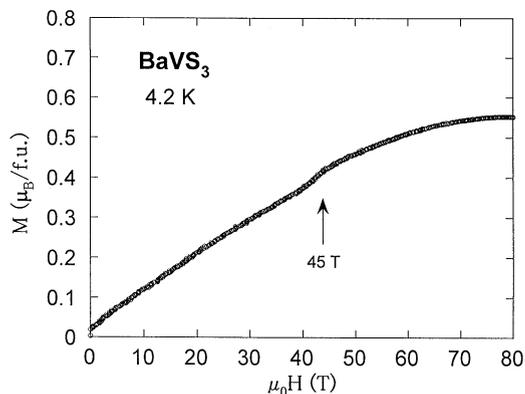


Fig. 3. Magnetization curve of BaVS₃ at 4.2 K for decreasing field. An arrow indicates the position of the magnetization jump.

becomes broader and weaker with increasing temperature. H_c for each temperature is plotted in Fig. 2. It becomes evident that H_c remains almost constant.

It is highly desired to estimate the magnitude of the magnetization jump in order to elucidate the origin of the jump. However, it is not easy to obtain a reliable magnetization value from the present data. Nevertheless, we tried to estimate the magnetization value by integrating the signal after smoothing noise signals by a computer analysis. The

absolute value was determined by comparing the integrated value with that obtained for Ni measured in the same condition. Fig. 3 shows the result of present estimation for decreasing field. The magnetization increases almost linearly up to 40 T and shows a tiny jump of about $0.1\mu_B$ at 45 T. Then, it exhibits a trend of saturation. The magnetic susceptibility estimated from the slope of the curve at low fields is somewhat larger than that obtained by a SQUID magnetometer for the same sample by the factor of 2, implying that the cancellation of applied field may not be complete.

4. Discussion

As mentioned in the first section, neither the origin of MI transition nor the ground-state magnetic properties of BaVS₃ is still well understood. As for magnetic structure, there are controversial observations conflicting each other.

NMR and NQR results suggested the orbital ordered spin singlet state [5]. Such a state may appear in the triangular lattice with orbital degeneracies and spin frustration as proposed by Pen et al. [8]. Although there is no direct calculation on the effect of applied field, it is likely that there exists some extent of a spin gap to break the singlet state. Then, we expect a much larger magnetization jump as observed in spin-Peierls systems [9,10] in conflict with the present result.

On the other hand, recent neutron diffraction measurements using a long-wavelength neutron beam ($\lambda = 4.05 \text{ \AA}$) has revealed that magnetic Bragg peaks appear at low angles below $T_X = 30 \text{ K}$, indicating long-range incommensurate antiferromagnetic spin ordering with the propagation vector of $(0.226 \ 0.226 \ 0)$ [6]. A possible magnetic structure to explain the observed magnetic peaks will be a modulated 120° structure often observed in CsCoCl₃-type frustrated X - Y spin systems. A tiny jump of magnetization was observed in such a system like CsCuCl₃ [11]. Although the exact spin structure and so the mechanism of the magnetization jump may not be the same for both systems, it is plausible that the anomaly in the magnetization curve of BaVS₃ is also characteristic of the frustrated X - Y spin system on a triangular lattice.

There remains, however, a serious problem that the magnetic Bragg peaks disappear at 30 K but the magnetization anomaly is still observed at 40 K. Low-energy inelastic neutron scattering measurements performed between 30 and 70 K have revealed that fluctuating antiferromagnetic order with almost the same propagation vector persists even in this temperature range [6] and, therefore, the system may be regarded as in a spin-liquid state, keeping the nearly the same spin correlation as in the ordered state below T_X . More precise experiments and a theory are requested to have a definite conclusion on this mysterious system.

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