High field magnetism of the quasi one-dimensional anisotropic antiferromagnet BaCo$_2$V$_2$O$_8$

S Kimura$^1$, H Yashiro$^1$, M Hagiwara$^1$, K Okunishi$^2$, K Kindo$^3$, Z He$^4$, T Taniyama$^1$, and M Itoh$^4$

$^1$KYOKUGEN, Osaka University, Machikaneyama 1-3, Toyonaka, Osaka 560-8531, Japan
$^2$Department of Physics, Niigata University, Niigata 950-2181, Japan
$^3$Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8531, Japan
$^4$Materials and Structures Laboratory, Tokyo Institute of Technology, 4259 Nagatsuta, Midori Yokohama 226-8503, Japan

E-mail: kimura@mag.cqst.osaka-u.ac.jp

Abstract. We have performed high field magnetization measurements on the quasi one-dimensional (1D) antiferromagnet BaCo$_2$V$_2$O$_8$, which shows a curious field-induced order-disorder transition, in pulsed magnetic fields up to 55 T. The observed magnetization curves up to the saturation field show non-linear increases, suggesting a strong quantum fluctuation. We analyze the magnetization curves in terms of the $S = 1/2$ 1D XXZ model.

1. Introduction
Recently, quantum spin systems in high magnetic fields have attracted much attention, since they show very interesting phenomena, such as the field-induced magnetic ordering in spin gap systems or the realization of magnetization plateau. BaCo$_2$V$_2$O$_8$ is a quasi one-dimensional (1D) Co$^{2+}$ spin system, which crystallizes in the tetragonal I41/acd space group [1, 2]. In this compound, edge-shared CoO$_6$ octahedra form a screw-chain structure along the $c$-axis. The chains are separated by non-magnetic V$^{5+}$ and Ba$^{2+}$ ion, resulting in 1D structural arrangement. The temperature dependence of the magnetic susceptibility of BaCo$_2$V$_2$O$_8$ for $H$//$c$ shows a broad maximum, which is characteristic of the low dimensional antiferromagnet, around 30 K, but owing to an inevitable interchain interaction, this compound undergoes antiferromagnetic ordering at 5.4 K at zero magnetic field [2-4]. The magnetic susceptibilities exhibit large magnetic anisotropy. The recent study by He et al. revealed that this compound shows a peculiar phenomenon in magnetic field, namely the field-induced order-disorder transition [3, 4]. When the external magnetic field is applied to the $c$-axis, which is the easy axis of BaCo$_2$V$_2$O$_8$, the magnetization curve at 2 K exhibits a steep increase above $H_c$ $\approx$ 4 T [3, 4]. The heat capacity measurements showed that the magnetic ordering temperature is rapidly lowered by the external field for $H$//$c$ and no magnetic ordering is observed down to 1.8 K in the field region above $H_c$ [3, 4]. On the other hand, falling in the ordering temperature in the field for $H \perp c$ is not significant compared with that for $H$//$c$ [4]. Even at 9 T, the ordering temperature for $H \perp c$ is 4.5 K [4]. In this study, to gain deeper insights into the curious transition of BaCo$_2$V$_2$O$_8$, we have measured the high field magnetization curves up to the saturation fields by using a pulse magnet.
2. Experimental
High field magnetization measurements on a single crystal of BaCo$_2$V$_2$O$_8$ for $H//c$ and for $H\perp c$ were performed in pulsed magnetic field up to 55 T. The magnetization was measured by means of a standard pick-up coil method. The details of the pulse magnet used in the measurements were reported in the literature [5]. The single crystal used in the measurements was synthesized by a spontaneous nucleation method [6].

3. Results and discussion
Figure 1(a) and (b) show the high field magnetization curves of BaCo$_2$V$_2$O$_8$ at 1.3 K, observed in pulsed fields for $H//c$ and for $H\perp c$, respectively. The transition field $H_c = 3.9$ T, observed for $H//c$ in this study, is consistent with the result of the previous measurements by He et al. In a high field region, peaks of field derivative of the magnetization ($dM/dH$) curves, which show a transition to the field-induced ferromagnetic phase, are observed at $H_s = 22.7$ T for $H//c$ and 40.9 T for $H\perp c$. The observed magnetization curves clearly show non-linear increases below $H_s$. In addition, as found in the $dM/dH$ curves, small anomalies are observed at $H'_c$ for both field directions. The observed critical fields are listed in Table 1. The magnetizations for both field directions increase linearly even in the field region above $H_s$. We attribute these linear magnetizations to the Van-Vleck paramagnetic contribution, which gives magnetic susceptibilities with no field dependence. Since slopes of the magnetization curve for $H//c$ below $H_c$ are almost identical with that above $H_s$, we conclude that the finite magnetization below $H_c$ at 1.3 K is also due to the Van-Vleck paramagnetism. The observed magnetization curves show large anisotropy. Similar magnetization curves were observed in the quasi 1D Ising-like antiferromagnet CsCoCl$_3$ [6-8], and its magnetization curve for the easy axis was well explained by the $S = 1/2$ 1D XXZ model, taking the next nearest neighbor interaction into account [7]. Therefore, we analyze the magnetization curves of BaCo$_2$V$_2$O$_8$ in terms of the $S = 1/2$ 1D XXZ
In our analysis, only the nearest neighbor interaction is taken into account. The magnetization curve for $H//c$ is analyzed by an exact solution for field applied to the longitudinal $z$-direction [9], and that for $H\perp c$ is compared with a theoretical calculation for the transverse field by the density-matrix renormalization-group method [10]. The Van-Vleck paramagnetic contributions, which give the linear magnetizations, are added to the above calculated magnetizations. From the analysis, the exchange parameters are estimated to be $J/k_B = 65$ K and $\epsilon = 0.46$, suggesting an Ising-like anisotropy. The obtained parameters are listed in Table 1. The magnetization curves agree with the theoretical calculations fairly well.

Table 1. Critical fields, obtained from the high field magnetization measurements on BaCo$_2$V$_2$O$_8$ at 1.3 K, and the parameters used in the analysis in terms of the $S=1/2$ 1D XXZ model.

| $H_c$(T) | $H_\perp c$(T) | $H_s$(T) | $J/k_B$(K) | $\epsilon$ | $g$ | $\chi_{VV}$(µB/T) |
|---------|----------------|-----------|-----------|------------|-----|--------------|
| $H//c$  | 3.9            | 19.5      | 22.7      | 65         | 0.46| 6.2          | 0.016 |
| $H\perp c$ | –           | 30.8      | 40.9      | 65         | 0.46| 2.95         | 0.015 |

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Finally we discuss slight discrepancies between the observed magnetization curves and the theoretically calculated ones. The magnetization above $H_c$ for $H//c$ is slightly larger than the calculated one. This is probably due to some additional interaction which enhances the field induced moment, such as the interchain interactions or the next nearest ferromagnetic intrachain interaction. The origin of the small anomaly at $H_c$, however, is not clear at the moment. For both field directions, the magnetizations at $H_c^t$, subtracted the Van-Vleck paramagnetic contribution, correspond to a half of the saturation values. To investigate the spin state in a high field region in more detail, high field/high frequency ESR measurements on BaCo$_2$V$_2$O$_8$ are now in progress.

In conclusion, we have observed the high field magnetization curves of BaCo$_2$V$_2$O$_8$ up to the saturation fields. The observed magnetization curves agree with those calculated from the $S=1/2$ 1D XXZ model fairly well. We believe that the field-induced order-disorder transition, found in BaCo$_2$V$_2$O$_8$ for $H//c$, is related to the quantum phase transition characteristic of the $S=1/2$ 1D XXZ model.

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