Magnetic response of YbMnO₃ single crystal

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The low ac- and dc-field magnetic susceptibilities of hexagonal YbMnO₃ single crystal have been measured along the *ab* plane and along the *c* axis (χ_{ab} , χ_c). The antiferromagnetic Néel temperature T_N is well signaled by a cusp in the temperature dependence of χ_c but no features are visible in χ_{ab} . A remarkable anisotropy of paramagnetic susceptibility and the extrapolated Curie–Weiss temperature are observed and related to the quasibidimensional magnetic structure of YbMnO₃. Below T_N magnetic irreversibility sets, reflecting the presence of a weak ferromagnetic response. Possible scenarios for the origin of the weak ferromagnetism are discussed. © 2008 American Institute of Physics. [DOI: 10.1063/1.2836403]

INTRODUCTION

YbMnO₃ belongs to the hexagonal group within the rare-earth manganites with general formula $R\text{MnO}_3$. There has been a renewed interest to $R\text{MnO}_3$ related to the magnetoresistive phenomenon in mixed valence manganites, and more recently due to the long range magnetic and polar orders, which may coexist in some of these oxides. The later phenomenon was discovered almost half a century ago by Bertaut *et al.*¹

In the hexagonal structure of YbMnO₃, each Mn³⁺ ion is surrounded by three in-plane and two apical O²⁻ ions, and exposed to a crystal field with trigonal symmetry. The magnetic order of the Mn moments is determined by antiferromagnetic in-plane Mn–O–Mn superexchange, which is much stronger than the interplane Mn–O–O–Mn exchange.¹ As a result, at T_N , Mn orders in the *ab* basal plane in a typical for frustrated antiferromagnets 120° arrangement.¹ As the temperature is further lowered below T_N , Yb moments order too.² Possible configurations of Mn and Yb moments have been suggested.^{2–4} In order to approach the mechanisms behind the intriguing properties of this highly anisotropic compound, studies on single crystals are required. Here we report the anisotropic magnetic response of YbMnO₃ single crystal to ac- and dc-magnetic fields. We will limit our current discussions to the high temperature region, where the Yb moments are in paramagnetic state.

EXPERIMENTAL

YbMnO₃ plateletlike single crystals have been grown by flux method from high temperature solution as described elsewhere.⁵ A naturally shaped crystal in the form of hexagon (thickness of 0.5 mm and mass of 0.01 g) has been cho-

sen for magnetic study. This crystal is from the same batch as the one studied by Yen *et al.*⁶ Magnetic response to dc- and ac-magnetic fields was measured by Quantum Design's superconducting quantum interference device and physical property measurement system magnetometers. The data reported here are not corrected for the demagnetizing field effect.

RESULTS AND DISCUSSION

The low dc-field cooled (FC) and zero field cooled (ZFC) susceptibilities in a field applied along the *c* axis are given (at an expanded scale) in Fig. 1. It clearly shows a peculiarity at $T_N \approx 81$ K. This peculiarity, which coincides with the one reported for a single crystal studied by Katsufuji

FIG. 1. (Color online) Temperature dependence of dc susceptibility along the *c* axis (circles) and the *ab* plane (rhombs) measured at field of 100 Oe (note that the susceptibility axis has been expanded to see the peculiarity at T_N).

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FIG. 2. (Color online) Temperature dependence of inverse dc susceptibility along the c axis (circles) and the ab plane (diamonds) measured at field of 100 Oe.

et al.,⁷ signals the onset of long range order between Mn moments. An important feature of this transition is the difference between FC and ZFC susceptibilities measured at low magnetic field, which sets right below T_N . The temperature at which the two susceptibilities split decreases with increasing the applied field and the difference between the two susceptibilities diminishes at fields higher than about 1 kOe. Such behavior is not a characteristic of an ideal antiferromagnetic structure, for which the Mn moments are supposed to compensate below T_N , but signals the existence of net magnetic moment and coercivity. Unlike the c -axis case, no any peculiarity at T_N is observed on the susceptibility measured with a field applied within the basal ab plane and the FC and ZFC susceptibilities coincide for all temperatures down to 2 K.

The inverse susceptibility (Fig. 2) along the hexagonal c axis obeys Curie–Weiss law at temperatures above 150 K. When the field is applied within the basal ab plane it deviates from the linear dependence until temperatures as high as room temperature. Linear fit of the data along the c axis yields asymptotic Weiss temperature $\theta = -170$ K in a good agreement with the c -axis value obtained by Sugie *et al.*² but smaller than the value for polycrystalline samples^{8,9} or the one measured in single crystals assembly.¹⁰ From the Curie constant $C_g = 2.1 \times 10^{-2}$ K emu/g an effective paramagnetic moment $\mu_{\text{eff}} = 6.8 \mu_B$ is estimated. The later experimental value is close to the theoretical value of $\mu_{\text{eff}} = (\mu_{\text{Mn}}^2 + \mu_{\text{Yb}}^2)^{1/2} = 6.7 \mu_B$ calculated from the Yb^{3+} free ion ($4.54 \mu_B$) and the spin-only value for the Mn^{3+} ion ($4.90 \mu_B$). Assuming that at high enough temperatures the slope of the inverse susceptibility within the ab plane be equal to the slope along the c axis¹¹ (equal Curie constants), one could speculate that θ , determined from ab -plane data above room temperature, should be at least -230 K. Assuming that the value of θ is a measure of the exchange interaction, this would imply effectively stronger intraplane than interplane antiferromagnetic interactions. The observed anisotropy in θ could account for the difference between the θ values along the c axis of single crystals and those reported for polycrystalline samples. In the later case the θ value is averaged over the crystallographic

FIG. 3. (Color online) Temperature dependence of the real χ' and imaginary χ'' components of the ac susceptibility measured at ac field of 10 Oe applied along the c axis at different frequencies.

directions. We also note that for both crystallographic orientations the ratio between $|\theta|$ and T_N is smaller than the value of about 10, reported for RMnO_3 with nonmagnetic rare earth.⁷

As seen from Fig. 2, the inverse susceptibility in both crystallographic orientations does not approach the temperature of magnetic order in the way expected for an antiferromagnet. Instead of having an upturn when approaching T_N from above a downturn, reminiscent of a ferrimagnet is observed. It is worth to note that well above T_N the susceptibility is field independent (at least up to 5 kOe). However, the c -axis susceptibility below T_N strongly depends on the applied magnetic field (data not shown), implying that a net magnetization and hysteresis exist in magnetically ordered state.

The above observations are in agreement with the measured ac-susceptibility response. As seen from Fig. 3, both the real and the imaginary components of ac-susceptibility along the c axis exhibit frequency dependent maxima just below $T_N = 81$ K (T_N is unambiguously identified by the onset of magnetic losses). No any peculiarity at T_N is observed on the ac-susceptibility measured with field applied within the basal ab plane. This behavior is reminiscent to the one observed of LnMnO_3 single crystal, which exhibits weak ferromagnetism attributed to canting of the antiferromagnetic moments due to antisymmetric Dzyaloshinskii–Moria interaction.¹²

As it has been mentioned before, the presence of weak ferromagnetic component along the c axis is supported by the hysteresis observed between FC and ZFC susceptibility curves measured at low fields, as well as by the presence of coercivity up to T_N . The coercivity and the magnetic moment at 10 K, i.e., above the ordering temperature of Yb, are 0.5 kOe and $0.03 \mu_B/\text{f.u.}$ (at 30 K they are 0.25 kOe and $0.01 \mu_B/\text{f.u.}$ respectively). No coercivity is present above 81 K, suggesting that the Curie temperature of the observed weak ferromagnetism coincides with the Neel temperature of the triangular lattice, T_N . From the value of magnetic moment measured along the c axis at 8 K and assuming that this

moment results from canting of Mn^{3+} moments, one could estimate a canting angle of about 0.35° from the ab plane.

The estimated canting angle is typical for weak ferromagnetism originating from antisymmetric Dzyaloshinskii–Moria exchange of the type $D(\mathbf{S}_i \times \mathbf{S}_j)$. Symmetry considerations permit such exchange and, in fact, weak ferromagnetism along the c axis has been already reported for YMnO_3 in the classical paper by Bertaut *et al.*¹ However, one should not exclude two more possible reasons for the observed weak magnetic moment along the c axis: crystal field interactions of the type DS_z^2 or possible nonstoichiometry leading to mixture of Mn^{3+} and Mn^{4+} ions. It is worth to mention that the presence of magnetic rare earths does not seem to be at the origin of the intriguing ZFC-FC hysteresis, as we have observed it in YMnO_3 (Ref. 13) thin films and TbMnO_3 thin films¹⁴ in both hexagonal and orthorhombic phases.

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