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Yb_2Pt_2Pb: A new quasi-two-dimensional antiferromagnet

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Abstract

We have synthesized single-crystals of Yb_2Pt_2Pb, which crystallize in the tetragonal U_2Pt_2Sn-type structure. The magnetic susceptibility χ is highly anisotropic. The $\chi_{\langle 1\,0\,0\rangle}$ for $B\|\langle 1\,0\,0\rangle$ is 30 times larger than $\chi_{\langle 0\,0\,1\rangle}$ for $B\|\langle 0\,0\,1\rangle$ at the lowest temperatures. Both $1/\chi(T)$ and $M(B)$ of Yb_2Pt_2Pb are dominated by the CEF effect. The $1/\chi_{\langle 1\,0\,0\rangle}$ above 150 K is well described by the Curie–Weiss law with $\theta = 34$ K and $\mu_{\rm eff} = 4.19\mu_B$, indicating well-localized Yb^{3+} ions at high temperatures. A broad maximum in χ is found around 3 K, just above antiferromagnetic transition temperature of 2 K. This suggests an important role for fluctuations in this system, due to reduced dimensionality or perhaps geometric frustration.

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Keywords: Tetragonal; Anisotropy; CEF; Frustration

Recently, the series of the ternary compounds with composition RA_2T_2M (RA = rare earths or actinides; T = transition metals; M = Pb, In, and Sn) has attracted much attention because of the abundance of collective phenomena in this series, such as ferromagnetic Kondo lattice, Kondo semiconductor, valence fluctuation, non-Fermi liquid, and heavy fermion. This series of compounds can be largely classified by two kinds of crystal structures, the tetragonal U_2Pt_2Sn-type (a superstructure of U_3Si_2-type) (space group $P4_2/mnm$) and the tetragonal Mo_2FeB_2-type (an ordered derivative of U_3Si_2-type) (space group P4/mbm). In these two crystal structures, *ab* planes occupied only by RA elements alternate with the *ab* planes occupied by T and M elements along *c*-axis. Further, the RA elements constitute a network of isosceles triangles, suggesting that one might expect a quasi-two-dimensional magnetic structure and anisotropic magnetic properties. However, all of the studies for this series of compounds so far have been performed on polycrystalline samples so that any anisotropic behavior has not been observed until now. Here, we present the magnetic susceptibility and magnetization along the crystal axes, $\langle 1\,0\,0\rangle$ and $\langle 0\,0\,1\rangle$ for single crystals of Yb_2Pt_2Pb.

We have synthesized single crystals of Yb_2Pt_2Pb from Pb-flux. Single crystal X-ray diffraction measurements indicate that Yb_2Pt_2Pb crystallizes in the tetragonal U_2Pt_2Sn-type structure with lattice parameters of $a = 7.7651(6)$ and $c = 7.0207(7)$ Å, which is in a good agreement with a previous report [1]. Magnetic susceptibility and magnetization were measured using a quantum design magnetic property measurement system in magnetic fields up to 7 T.

Fig. 1 shows the temperature dependence of magnetic susceptibility $\chi_{\langle 1\,0\,0\rangle}$ and $\chi_{\langle 0\,0\,1\rangle}$ for $B\|\langle 1\,0\,0\rangle$ and $B\|\langle 0\,0\,1\rangle$, respectively. The $\chi_{\langle 1\,0\,0\rangle}$ is about 30 times larger than $\chi_{\langle 0\,0\,1\rangle}$ at the lowest temperatures, indicating that the magnetic properties of Yb_2Pt_2Pb are strongly anisotropic. Considering the symmetry of the crystal structure of Yb_2Pt_2Pb, this suggests that a magnetic easy axis lies in *ab* plane, resulting in a quasi-two-dimensional magnetic property. The inverse magnetic susceptibility $1/\chi_{\langle 0\,0\,1\rangle}$ for $B\|\langle 0\,0\,1\rangle$ is not well described by Curie–Weiss law at any temperature. The nonlinear temperature dependence observed for $1/\chi_{\langle 0\,0\,1\rangle}$

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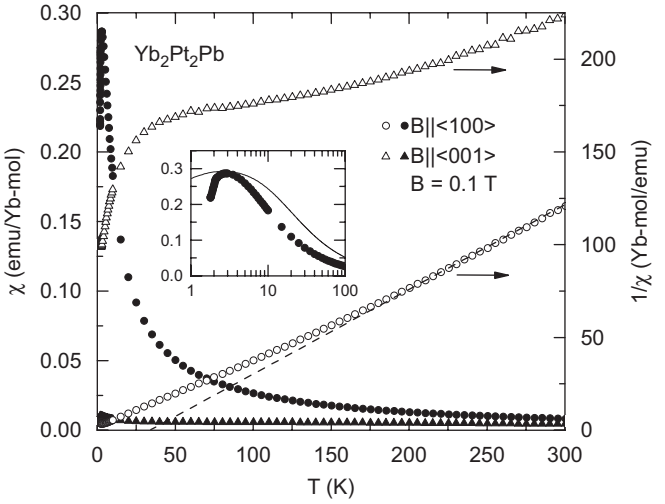

Fig. 1. The temperature dependence of $\chi_{\langle 1\,0\,0\rangle}$ (•), $\chi_{\langle 0\,0\,1\rangle}$(▲), $1/\chi_{\langle 1\,0\,0\rangle}$ (○), and $1/\chi_{\langle 0\,0\,1\rangle}$ (△) for $B\|\langle 1\,0\,0\rangle$ and $B\|\langle 0\,0\,1\rangle$, respectively. Dashed line indicates the fitting results of the Curie–Weiss law above 150 K for $1/\chi_{\langle 1\,0\,0\rangle}$. The inset shows an enlarged plot of $\chi_{\langle 1\,0\,0\rangle}$ below 100 K with log T scale. Solid line represents the susceptibility calculated for a two-dimensional spin-$\frac{1}{2}$ Heisenberg antiferromagnet on the triangular lattice, as described in the text.

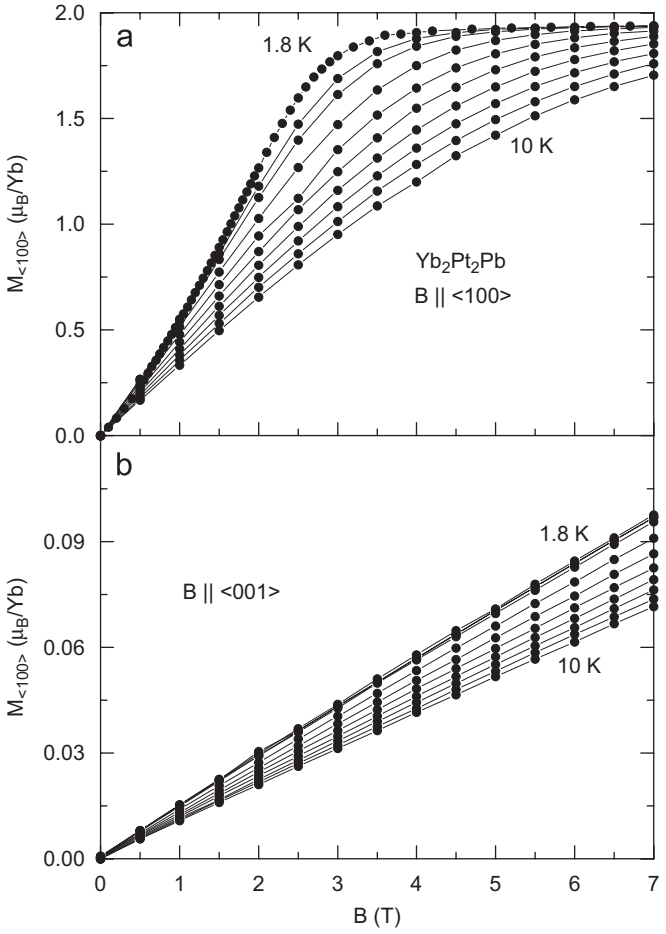

Fig. 2. The magnetic field dependence of (a) $M_{\langle 1\,0\,0\rangle}$ and (b) $M_{\langle 0\,0\,1\rangle}$ between 1.8 and 10 K for $B\|\langle 1\,0\,0\rangle$ and $B\|\langle 0\,0\,1\rangle$, respectively.

may mainly be the result of the crystalline electric fields (CEF) of the local $m2m$ symmetry of the Yb site. A prominent shoulder below 50 K is likely related to the splitting energy of ~50 K between the doublet ground state and the first excited doublet state in the CEF degeneracy with four doublets. In contrast, $1/\chi_{\langle 1\,0\,0\rangle}$ is well described above 150 K by Curie–Weiss law with a Curie temperature $\theta = 34$ K and effective moment $\mu_{\text{eff}} = 4.19\mu_{\text{B}}$, indicating well-localized Yb^{3+} ions at high temperatures.

A strong anisotropy is also found between the magnetizations $M_{\langle 1\,0\,0\rangle}$ and $M_{\langle 0\,0\,1\rangle}$ for $B\|\langle 1\,0\,0\rangle$ and $B\|\langle 0\,0\,1\rangle$, respectively, as shown in Fig. 2. With increasing magnetic fields from 0 T, $M_{\langle 1\,0\,0\rangle}$ gradually increases up to 3 T and saturates to $2\mu_{\text{B}}$ above 4 T after showing a shoulder around 3.5 T, while $M_{\langle 0\,0\,1\rangle}$ exhibits only linear behavior and is only $0.1\mu_{\text{B}}$ even at 7 T. On the other hand, the saturation moments of $M_{\langle 1\,0\,0\rangle}$ are much smaller than the fully degenerate value of $4.0\mu_{\text{B}}$, but is consistent with that found in Yb-compounds with a doublet ground state caused by the tetragonal CEF.

The magnetic properties of Yb_2Pt_2Pb at low temperatures are quite remarkable. A sharp drop in $\chi_{\langle 1\,0\,0\rangle}$ is found at 2.0 K as shown in the inset of Fig. 1, indicating an antiferromagnetic transition ($T_{\text{N}} = 2$ K). For $\chi_{\langle 0\,0\,1\rangle}$, the suppression of magnetic susceptibility associated with magnetic ordering at T_{N} is much weaker than that in $\chi_{\langle 1\,0\,0\rangle}$ (not shown here). Most remarkably, we find that there is a broad maximum in $\chi_{\langle 1\,0\,0\rangle}$, centered around 3 K, indicating that magnetic order emerges from a strongly fluctuating state. We believe that both the two-dimensionality and the geometric frustration of the localized Yb moments may be responsible for these anomalous fluctuations.

Recently, a similar broad maximum in the temperature dependence of χ was found above the ordering temperature of some organic compounds with two-dimensional and triangular networks of spin $\frac{1}{2}$ moments [3]. The maximum in both the susceptibility and the heat capacity was subsequently described by a theoretical expression derived from a high temperature series expansion for a spin-$\frac{1}{2}$ Heisenberg antiferromagnet on a triangular lattice [2]. We have carried out a similar comparison, and the results are presented in the inset of Fig. 1. The best fit to our experimental data was achieved using angular momentum $J = 3/2$ and an antiferromagnetic in-plane exchange interaction of 7 K. The agreement between the data and the theoretical expression is quite reasonable, although the theoretical expression clearly overestimates the role of fluctuations. Specifically, the crystal structure of Yb_2Pt_2Pb insures that there are two different exchange couplings among the Yb moments in *ab* plane; so frustration in the triangular lattices will necessarily be somewhat relieved, relative to the theoretical expression. Second, we note that because of crystal electric fields, the true angular momentum state is unknown, although the deduced value of $J = \frac{3}{2}$ is reasonable given the crystal field scheme implied by the susceptibility and magnetization measurements. We expect

the theoretical expression to become increasingly inaccurate as the temperature approaches ~50 K, which is the first crystal field excitation. Finally, the theoretical expression ignores exchange coupling between the Yb atoms in neighboring *ab* planes, which ultimately lead to three-dimensional magnetic order. It is noted that, according to experimental observation for those organic compounds with the two- dimensional triangular lattice, the deviation from the regular triangular lattice induces enhancing T_N in the presence of interlayer couplings [3]. The primary success of the theoretical expression is to match the maximum in the experimental susceptibility, assuming an average in-plane exchange interaction of ~7 K.

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