Magnetization process in the spin-ice compound $Ho_2Ti_2O_7$

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We report the results of magnetization measurements performed on single crystals of the pyrochlore compound $Ho_2Ti_2O_7$. The low-temperature magnetization curves for the three principal directions of an applied magnetic field are found to be in nearly perfect agreement with the predictions for a nearest-neighbor spin-ice model, contrary to previous reports.

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The pyrochlore compound $Ho_2Ti_2O_7$ is considered to be a good realization of the spin-ice model, where the magnetic Ho^{3+} ions possess a strong Ising anisotropy and are constrained to point along the local $\langle 111 \rangle$ direction.[1] This assertion is most vividly corroborated by the elastic neutron-scattering patterns obtained in $Ho_2Ti_2O_7$ at sufficiently low temperature.[2] In the spin-ice model, the following values of saturation magnetization are predicted:[3–5] $M^{sat}_{H\|[100]}=\mu/\sqrt{3}$, $M^{sat}_{H\|[111]}=\mu/2$, and $M^{sat}_{H\|[110]}=\mu/\sqrt{6}$, where μ is a full magnetic moment of a Ho^{3+} ion (the corresponding spin configurations are shown in Fig. 1 of Ref. 5).

Cornelius and Gardner[6] have reported the results of magnetization and susceptibility studies on single crystals of the pyrochlore $Ho_2Ti_2O_7$. The authors claimed that the observed magnetization isotherms are qualitatively similar to that predicted by the spin-ice model. Two major contradictions of their results with the theoretical predictions are, however, obvious. The first discrepancy is that the saturation magnetic moment[7] of $5.9\mu_B$ per Ho atom is reported to be nearly independent of the orientation of magnetic field, while in the spin-ice model M_{111}, M_{110}, and M_{100} must differ. The second discrepancy is that for $H\|[110]$ the magnetization is claimed to be almost temperature independent below $T=4$ K. In addition, the reported low-field susceptibility for $H\|[110]$ is very different from the other two directions, which contradicts the cubic symmetry of the crystal.

These are rather surprising results, given that the magnetization curves of a similar spin-ice compound, $Dy_2Ti_2O_7$, do follow the theoretical predictions quite precisely.[5,8] In an attempt to clarify the situation we have remeasured the magnetization curves of $Ho_2Ti_2O_7$ for the three principal directions of an applied magnetic field, [100], [110], and [111].

Single crystals of $Ho_2Ti_2O_7$ have been grown by the floating-zone technique using an infrared image furnace.[9] By measuring the x-ray-diffraction patterns on powder samples prepared from single crystals, we confirmed the high purity of the crystals, since no impurity peaks were found. The magnetization versus field data were collected using an Oxford Instruments vibrating sample magnetometer (VSM) between 1.6 and 18 K in applied fields of up to 12 T. We have used small (5 to 25 mg) samples of various shapes.[10] The absolute accuracy of the magnetization measurements was of the order of 3%.[11] The principal axes were determined using x-ray-diffraction Laue photographs; the crystals were aligned to within an accuracy of 1–2°. Due to a significant anisotropy and the large magnetic moments involved in the system, it is extremely difficult to keep the $H\|[110]$ orientation for a sample in a high applied field. For a crystal in an unfavorable orientation, even in a moderate field of 3–4 T the magnetic forces involved are strong enough to break the crystal. Special precautions needed to be taken to ensure that the crystal does not move in an applied field.

The results of the measurements are summarized in Figs. 1 and 2. In Fig. 1 the observed magnetization is plotted against the effective magnetic field, $H=H_{ext}-nM$, n being the demagnetization factor.[10] The field dependence of the magnetic susceptibility at various temperatures obtained by differentiation of the M_{111} data is shown in Fig. 2. For the magnetic field applied along the easy-axis direction, $H\|[100]$, the magnetization curves look rather similar to those reported previously.[6] The only small difference is in the value of saturation magnetization: $(6.22\pm0.19)\mu_B$/Ho compared to $5.9\mu_B$/Ho in Ref. 6, which could be attributed to the limited experimental accuracy. For $T>2$ K, Cornelius

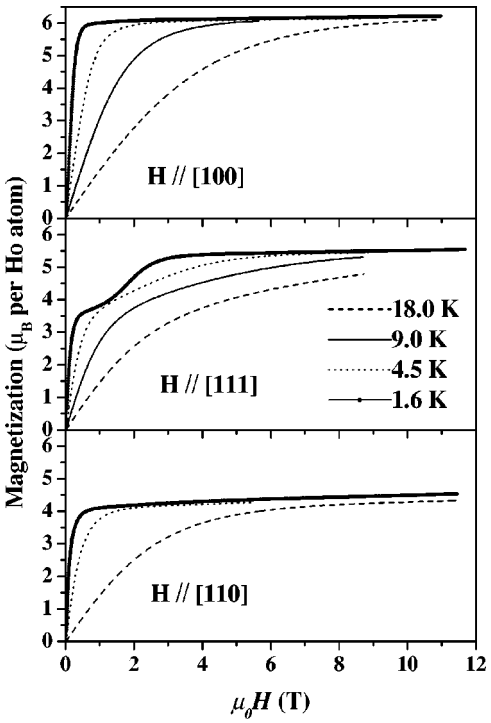

FIG. 1. Field dependence of the magnetization of $Ho_2Ti_2O_7$ single crystals at different temperatures.

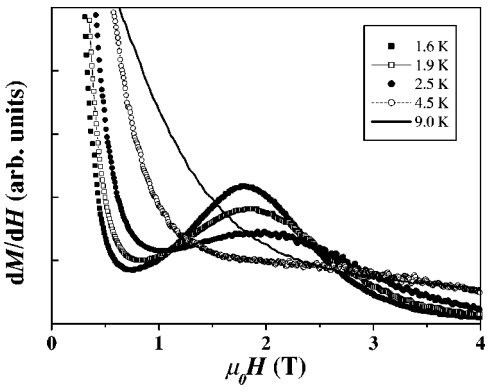

FIG. 2. Temperature evolution of the field dependence of the magnetic susceptibility for $H\|[111]$ in a single crystal of $Ho_2Ti_2O_7$.

and Gardner[6] obtained identical results for M_{111} and M_{100}, while our measurements revealed significantly different values for different orientations at all temperatures. At sufficiently low temperature, $T<2$ K, we observed a plateau in M_{111} at $3.6\mu_B$/Ho in agreement with Ref. 6. The obtained value of saturation magnetization, $M^{sat}_{H\|[111]}$, is, however, rather different: $(5.54\pm0.17)\mu_B$ per Ho atom. The saturation is reached in a field of about 3 to 4 T compared to 7 to 8 T reported previously.[6] Despite the metastable character of the plateau, no significant hysteresis in the magnetization was detected at any temperature. Note that the observed ratio $M^{sat}_{H\|[111]}/M^{sat}_{H\|[100]}=0.89\pm0.05$ agrees well with the theoretical ratio $\sqrt{3}/2\approx0.866$.

Our results for a hard-axis magnetization, M_{110}, are in the sharpest contrast with the data reported by Cornelius and Gardner.[6] The low-field magnetization ($H<0.3$ T) is found to be temperature dependent and similar to that for the other two orientations of magnetic field. With increasing field, the magnetization grows rapidly and reaches a saturation in a field of about 1.5 T. The value of the saturation magnetization for $H\|[110]$ is $(4.43\pm0.13)\mu_B$ per Ho atom. Again, the observed ratio $M^{sat}_{H\|[110]}/M^{sat}_{H\|[100]}=0.73\pm0.04$ complies with the spin-ice theory that predicts $1/\sqrt{2}\approx0.707$.

From the values of the saturation magnetization for all three orientations and from the value of M_{111} on its plateau, the full magnetic moment on the Ho site is $\mu=(10.9\pm0.3)\mu_B$. This value is consistent with the free ion moment for the $^5I_8$ state of Ho^{3+}, $\mu=10.6\mu_B$, but is also close to the value expected for an $m_J=\pm8$ doublet ground state, $\mu=10.0\mu_B$ (Ref. 12).

The lowest temperature available for our measurements, $T=1.6$ K, is still relatively high on the scale of the exchange interactions in $Ho_2Ti_2O_7$. As a result, the magnetic susceptibility remains positive on the plateau in the M vs H data and does not drop to zero (see Fig. 2). Perhaps for the same reason, we were unable to see the multiple plateaus in magnetization predicted by the Monte Carlo calculations[4,13] and observed in the spin-ice compound, $Dy_2Ti_2O_7$ (Ref. 14).

To conclude, the observed field dependence of magnetization for a single crystal of $Ho_2Ti_2O_7$ matches well the predictions for the spin-ice nearest-neighbor model contrary to an earlier report.[6]

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