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Pressure-induced weak ferromagnetism in uranium dioxide, UO_2

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Abstract

The dc magnetization of insulating UO_2 under high pressure up to $\sim$1 GPa has been measured using a piston–cylinder cell. Pressure-induced weak ferromagnetism appeared at low pressure ($\sim$0.2 GPa). Both the remanent magnetization and the coercive force increase as pressure increases. This weak ferromagnetism may come from spin canting or from uncompensated moments around grain boundaries.

1. Introduction

Cubic fluorite-type uranium dioxide UO_2 is an ionically bonded insulator with localized magnetic moments of $U^{4+}(5f^2)$ ions. Under ambient pressure, UO_2 exhibits a first-order antiferromagnetic transition at $T_N \sim 30$ K accompanied by a very small lattice distortion [1–4]. The magnetic structure has been determined to be not a simple collinear structure, but a non-collinear $3\boldsymbol{k}$-structure with a dynamical Jahn–Teller distortion, on the basis of both theoretical and experimental studies [5–7]. This transition comes about as a consequence of competition between quadrupole–quadrupole (QQ) interactions and magnetic exchange interactions. Since the magnetism of UO_2 is strongly coupled with the lattice, the magnetic properties are considered to be sensitive to pressure. In order to elucidate the relation between the QQ and the exchange interactions in this system, we have performed dc magnetization measurements under hydrostatic pressure up to about 1 GPa.

2. Experimental details

A powdered sample of UO_2 was prepared by heating uranium oxide at 1273 K under an atmosphere of hydrogen. The dc magnetization measurements were carried out using a SQUID

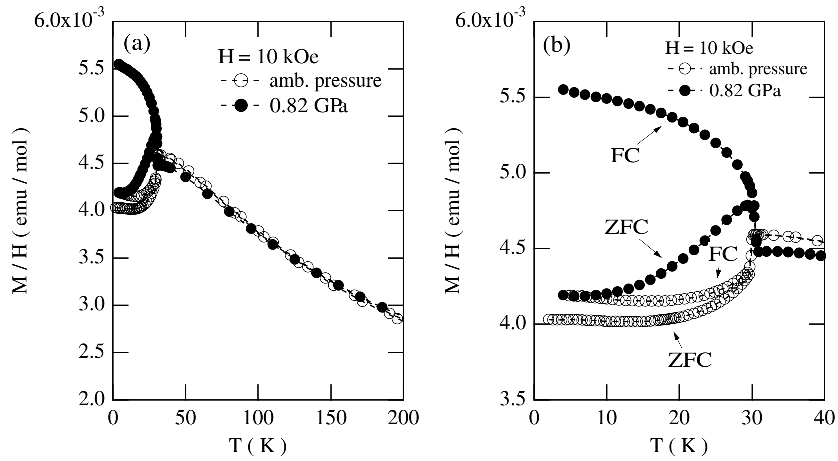

Figure 1. (a) The temperature dependence of the magnetization (M) divided by the magnetic field ($H = 10$ kOe) for UO_2 under pressure. The open circles (∘) and the closed circles (•) represent the magnetization data under ambient pressure and an external pressure of 0.82 GPa, respectively. (b) An enlargement of part (a). The ZFC and FC data are labelled with arrows.

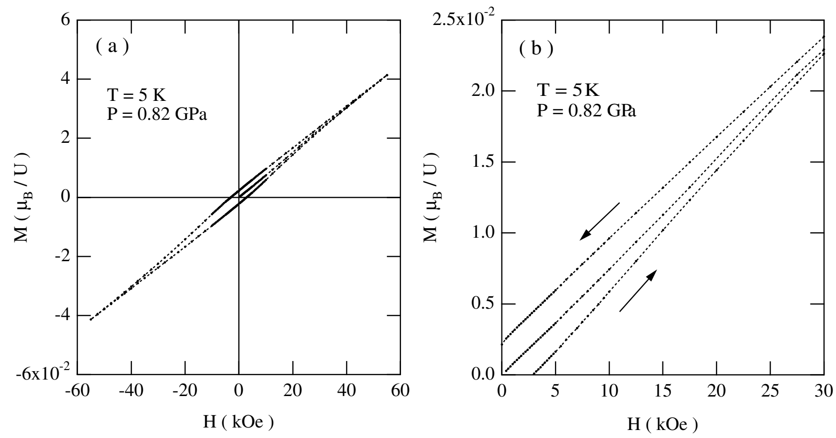

Figure 2. (a) The magnetization curve of UO_2 under the pressure of 0.82 GPa. (b) An enlargement of part (a).

magnetometer. Hydrostatic pressure was applied by utilizing a Cu–Be piston–cylinder cell with Daphne oil (7373) as a pressure-transmitting medium. The pressure value was determined by measuring the superconducting transition temperature of lead.

3. Results

Figure 1 shows the temperature dependence of the magnetization (M) divided by magnetic field ($H = 10$ kOe) under ambient pressure and an external pressure of 0.82 GPa. The respective data were collected after zero-field cooling (ZFC) and field cooling (FC), where the applied field was 10 kOe. Under ambient pressure, the magnetization obeys a Curie–Weiss law from near $T_N = 30.5$ K to room temperature with an effective moment of 3.1 μ_B/U^{4+}

and a Weiss temperature of -210 K. The magnetization abruptly reduces just below T_N, and shows little difference between ZFC and FC sequences; this may be a result of the non-collinear magnetic structure. The magnetization data under 0.82 GPa in the paramagnetic region ($T_N < T < 300$ K) are in agreement with those under ambient pressure. A very small deviation above ~200 K comes from solidification of the pressure-medium oil. Under the pressure of 0.82 GPa, the magnetization after both ZFC and FC increases just below $T_{N1} = 30.8$ K. The ZFC data under 0.82 GPa begin to decrease at $T_{N2} = 29.4$ K as temperature decreases below T_{N1}. These transition temperatures (T_{N1} and T_{N2}) depend only slightly on the applied pressure up to ~1 GPa. On the other hand, the FC data rise as for a ferromagnetic substance. This irreversibility between the ZFC and FC sequences becomes larger as the applied pressure increases. Figure 2 shows a magnetization curve of UO_2 under 0.82 GPa in the field range of -55 to +55 kOe. There is a small hysteresis where, as shown in figure 2(b), the remanent magnetization (M_R) and coercive force (H_C) are determined as 0.0021 μ_B/U^{4+} and 2.9 kOe, respectively. These results mean that there is a weak ferromagnetic moment induced by pressure. Although omitted on account of space limitations, detailed pressure experiments show that both M_R and H_C increase with applied pressure. Both become large abruptly around 0.2 GPa. More detailed results of magnetization measurements under pressure will be published elsewhere.

This pressure-induced weak ferromagnetism probably comes from canting of the magnetic moments. That is, since the spin-canted $3\boldsymbol{k}$-structure is realized even under ambient pressure, a slight change of canting angle may occur readily under applied pressure. Of course, a different scenario can also be considered wherein local moments, which are not cancelled by bulk antiferromagnetism near grain boundaries, would behave as weakly ferromagnetic. In order to clarify the origin of the weak ferromagnetism, a study of the microscopic magnetism of UO_2 under high pressure is currently under way, by means of nuclear resonance measurements.

References

[1] Frazer B C *et al* 1965 *Phys. Rev.* A **140** 1448
Willis B T and Taylor R I 1965 *Phys. Lett.* **17** 188

[2] Pirie J D and Smith T 1970 *Phys. Status Solidi* **41** 221

[3] Khattak G D 1983 *Phys. Status Solidi* A **75** 317

[4] Brandt O G and Walker C T 1967 *Phys. Rev. Lett.* **18** 11

[5] Many theoretical works have been carried out on this system. For example, see the reference
Giannozzi P and Erdös P 1987 *J. Magn. Magn. Mater.* **67** 75

[6] Also, many neutron scattering experiments have been carried out. For example, see the reference
Caciuffo R *et al* 1999 *Phys. Rev.* B **59** 13892

[7] Ikushima K *et al* 2001 *Phys. Rev.* B **63** 104404