Neutron Diffraction Study of Crystal and Magnetic Structures of Manganese Deuteride at High Pressure

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Manganese (Mn) is a prototype of antiferromagnetic itinerant-electron systems. Its crystal and magnetic structures are complex; the unit cell consists of 58 atoms on which four kinds of inequivalent magnetic moments reside to make the sum zero. In contrast, Mn hydrides, which are prepared by hydrogenation of the metal at high hydrogen pressures, have simple structures; dissolved hydrogen atoms occupay the interstitial sites of fcc, hcp or dhcp metal lattices. The former two hydrides, fcc and hcp MnH_x were confirmed to be antiferromagnetic by early neutron diffraction measuremsnts with recovered specimens at low temperarures and ambient pressures [1, 2].

We investigated the crystal and magnetic structures of MnD_x for a wide temperature range at high deuterium pressures by in situ neutron diffraction at BL11, MLF, J-PARC [3]. The fcc deuteride was prepared at 873 K and 1.3 GPa. Diffraction profiles were collected at a temperature interval of 20 K upon cooling. The fcc–hcp transition occurred at about 510 K slightly lower than the reported transition temperature. The fcc and hcp phases coexisted duering further cooling to 300 K. The pressure decreased to 0.3 GPa when the temperature reached 300 K.

A partial diffraction profile collected at 300 K and 0.3 GPa is shown in Fig. 1. The diffraction peaks from the fcc and hcp deuterides are indexed such as f111 and h100, respectively. The profile contains those from an undesired inpurity, MnO, and pressure medium, NaCl. In addition to the scattering peaks from Mn nucleus, the magnetic scattering peak (fmag 110) is clearly observed for the fcc deuteride.

The magnetic peak appeared at ~550 K in the fcc phase just above the fcc–hcp phase boudary. The peak intensity increased upon cooling to 300 K. Rietveld refinements of the observed profiles using a proposed magnetic structure [2] showed that the magnitude of the moment was ~1.8 μ_B at 300 K and an approximate Néel temperature was located at ~530 K.

For the hcp deuteride, magnetic peaks appeared at about 300 K. The temperature variation of its magnitude was measured with a recovered specimen for the temperature range 100–300 K at around 1 GPa. The magnitude of the moment was ~0.8 μ_B at 100 K in agreement with a reported value [1] and the temperature variation gave an approximate Néel temperature of ~300 K. In the both deuteride, the deuteride composition increased with decreasing temperature, showing exothermic temperature dependence.

The present study revealed that the magntitude of magnetic moments subsequently depends on the metal lattice structure. The Néel temperature of the fcc deuteride is about twice of the value of the hcp deuteride. The relationship between the magnetic and crystal structures including the deuterium composition is further subject to be interpreted in connection with the electronic band strutures.

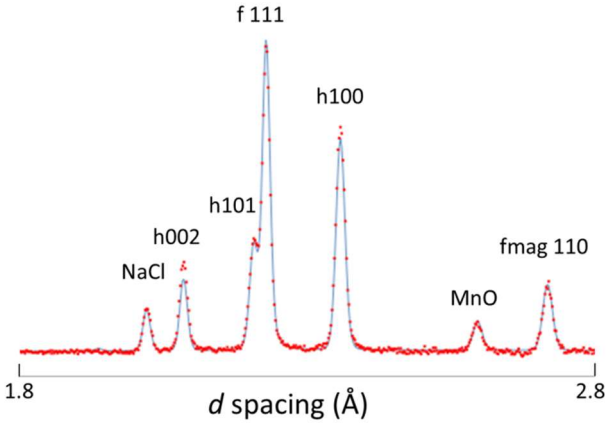

Figure 1. Neutron powder diffraction profile collected for manganese deuteide at 300 K and 0,3 GPa.

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