Néel transition in (Mg,Fe)O: A possible change of magnetic structure

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ABSTRACT

The Néel temperature ($T_N$) of (Mg$_{1-x}$Fe$_x$)O and its compositional dependence have been studied with a superconducting quantum interference device (SQUID) in the temperature range 5–300 K at constant magnetic field of 1000 Oe. We determined the magnetic phase diagram at ambient pressure. As the $T_N$ constantly increased with iron content, we found a kink in the paramagnetic-antiferromagnetic boundary at around $X = 0.5$, which suggests a possible change of the magnetic structure in the antiferromagnetic state. We propose that the magnetic boundary ends at $X = 0.12(1)$ and 0 K. Our results show that the pressure of the rhombohedral distortions of (Mg,Fe)O can be very sensitive to Fe content if the structure distortion is correlated to magnetic ordering.

Keywords: Néel transition, SQUID, (Mg,Fe)O, magnetic structure, low temperature

INTRODUCTION

Wüstite (FeO) is an important component in the Earth’s lower mantle as an iron-rich end-member of the major oxide, (Mg,Fe)O. It transforms to a rhombohedral structure from the B1 (rock salt) structure above 16 GPa at 300 K (Zou et al. 1980) and the Néel transition is also known at about 200 K and ambient pressure. Okamoto et al. (1967) reported that the pressure dependence of the $T_N$ is 6.5 K/GPa below 0.5 GPa. A linear extrapolation of the Néel transition indicates that at ambient temperature the transition would occur at about 16 GPa, which coincides with the condition of the observed rhombohedral distortion (Zou et al. 1980). The magnetic transition was also observed at similar physical conditions in high-pressure Mössbauer spectroscopy (MS) (Nasu 1994). Therefore, the rhombohedral distortion in FeO has been interpreted as the result of an increase of the superexchange interaction of magnetic ions in the antiferromagnetic ordering (Smart and Greenwald 1951).

At higher pressure, wüstite transforms to a B8 (NiAs) structure at 74 GPa and 900 K (Fei and Mao 1994). A possible metallic state in the B8 phase, which may be related to the magnetic structure (Cohen et al. 1997; Fang et al. 1999), has been the subject of several studies with implications in geophysics due to its extreme importance in understanding the properties and composition of the Earth’s core (Fei and Mao 1994; Kondo et al. 2004). At higher pressure, the $T_N$ was observed to decrease with increasing pressure between 90 and 120 GPa (Badro et al. 1999). These $P-T$ conditions are located below the proposed structure boundary (Fei and Mao 1994). In general, the antiferromagnetic effect increases with increasing pressure due to decreasing Fe-Fe distance. This implies that the volume reduction occurs by structural distortion. The study of the magnetic properties of wüstite is important for understanding the origin of the structural transformation.

The structural distortion is also influenced by composition. Periclase (MgO), the other end-member of (Mg,Fe)O, has a B1 structure at ambient condition and forms a complete solid solution with wüstite. Periclase is nonmagnetic and shows no structural transition at least to 227 GPa and room temperature (Duffy et al. 1995). However, there are apparent contradictions among different experimental studies performed on intermediate (Mg,Fe)O compositions. (Mg$_{0.9}$Fe$_{0.1}$)O and (Mg$_{0.85}$Fe$_{0.15}$)O dissociated into magnesium-rich and iron-rich oxide components at 86 GPa and 1000 K in an externally heated diamond anvil cell (DAC) experiment (Dubrovinsky et al. 2000). On the other hand, the compositions (Mg$_{0.39}$Fe$_{0.61}$)O, (Mg$_{0.28}$Fe$_{0.72}$)O, (Mg$_{0.28}$Fe$_{0.72}$)O, (Mg$_{0.10}$Fe$_{0.90}$)O, and (Mg$_{0.05}$Fe$_{0.95}$)O, did not reproduce the phenomena in laser-heating experiments in the DAC (Lin et al. 2002; Kondo et al. 2004). The addition of periclase component to wüstite should stabilize the B1 structure at much higher pressures and temperatures. For example, (Mg$_{0.25}$Fe$_{0.75}$)O did not distort to a rhombohedral structure below 60 GPa at room temperature (Lin et al. 2002). A combined in situ study of X-ray powder diffraction (XRD), MS, and X-ray absorption near edge structure (XANES) spectroscopy for (Mg$_{0.85}$Fe$_{0.15}$)O showed the structural transformation at 35(1) GPa and room temperature (Kantor et al. 2006). No magnetic splitting was observed in the Mössbauer spectra of the high-pressure phase at least to 56 GPa, indicating that rhombohedral distortion does not involve magnetic ordering. Consequently, Kantor et al. (2006) concluded that the transition from the B1 to the rhombohedral structure is accompanied by a rapid $C_{44}$ mode softening (Sumino et al. 1980) and a decrease in sound velocities, and that it is not induced by magnetic ordering.

The relationship between the magnetic change, composition, hydrostaticity, and structure of (Mg,Fe)O is still a debated issue. In this study, we have investigated the compositional dependence of the $T_N$ to study the origin of the rhombohedral distortion in (Mg,Fe)O at high pressure.

EXPERIMENTAL METHODS

Eight (Mg$_{1-x}$Fe$_x$)O compositions were synthesized from oxide mixtures of Fe$_2$O$_3$ and MgO (Wako Chemical Co. Ltd.) with various Mg/Fe molar ratios ($X = 0.20, 0.31, 0.40, 0.50, 0.60, 0.70, 0.80, 0.95$). The compositions were sintered in...