Magnetic Properties of NdFe$_{0.9}$Mn$_{0.1}$O$_3$

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In our paper we study effect of Mn for Fe substitution on magnetic properties of NdFe$_{x-1}$Mn$_x$O$_3$ compounds for $x = 0$ and 0.1, which have been grown by the OFZ technique. The Néel temperature decreases from $T_{N1} = 691$ K to $T_{N1} = 621$ K, and the anomaly in AC susceptibility, related to spin reorientation, vanishes with Mn substitution. Low temperature heat capacity measurement for sample with $x = 0$ revealed that substitution of Mn for Fe shifts a Schottky-type anomaly at $T_{sh}$ to higher temperatures. Another anomaly is generated by doping at $T_{max} = 11$ K. The anomaly is smeared out by magnetic field, confirming its magnetic origin.

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Magnetic properties of NdFe$_3$O$_4$ are mostly determined by three magnetic interactions (Fe-Fe, Fe-Nd and Nd-Nd), which are present in this material. Magnetic ordering of Fe$^{3+}$ ions creates a canted antiferromagnetic ordering below the Néel temperature at about $T_{N1} = 690$ K [1]. Upon cooling the magnetic moments of Fe$^{3+}$ exhibit reorientation from the $a$-axis to the $c$-axis in the spin reorientation region (103–165 K) [2]. Low temperature heat capacity measurements revealed a Schottky anomaly at about 2 K and a sharp maximum at $T_{N2} = 1.05$ K [3]. Neutron diffraction measurements confirmed magnetic ordering for Nd-Fe sublattice below 1 K and long-range ordering due to Nd-Nd interaction below 0.4 K [4, 5]. In our paper we study the effect of Mn substitution for Fe in NdFe$_3$O$_4$.

NdFe$_{x-1}$Mn$_x$O$_3$ ingots with $x = 0$ and 0.1 have been grown by the optical floating zone (OFZ) technique in the four mirrors furnace. The X-ray powder diffraction pattern and EDX analysis taken from the top and the end of ingots revealed that samples are single phase materials. Both samples adopt orthorhombic crystal structure (space group $Pbmm$) with lattice parameters $a = 5.5889(2)$ nm, $b = 7.7619(3)$ nm, $c = 5.4521(2)$ nm for $x = 0$ and $a = 5.6011(9)$ nm, $b = 7.748(8)$ nm, $c = 5.4483(9)$ nm for $x = 0.1$, respectively. Magnetization and AC susceptibility were measured on a SQUID magnetometer on MPMS in temperatures range from 2 to 720 K and magnetic flux density up to 5 T. Measurements of heat capacity were performed on PPMS in temperature range from 2 K to 200 K and in magnetic fields up to 3 T.

Transition from paramagnetic to canted antiferromagnetic state is accompanied with sharp peak in $\mu(T)$ curves (see inset of Fig. 1), which can be attributed to Hopkinson effect. Such a peak is a very characteristic feature shown by a number of ferromagnetic materials. In our case this peak indicates that ferromagnetic component can be strong. The Néel temperature decreases from 691 K to 621 K with Mn doping. The spin reorientation in the sample with $x = 0.1$ is indicated by steep decrease of magnetization in the range between 150 K and 125 K (Fig. 1), but the relatively flat curve down to 15 K is unusual in comparison with NdFe$_3$O$_4$. The rise of magnetization below 15 K indicates increasing Nd-Fe magnetic interactions which start to develop at higher temperature in comparison with NdFe$_3$O$_4$.

The spin reorientation is not accompanied by any anomaly in the AC in phase susceptibility of NdFe$_{0.9}$Mn$_{0.1}$O$_3$ (Fig. 2.) or out of phase susceptibility. Frequency dependent anomaly in the AC susceptibili-
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Fig. 2. In phase AC susceptibility of \( \text{NdFe}_{0.9}\text{Mn}_{0.1}\text{O}_3 \); inset shows frequency dependence of the in phase AC susceptibility for \( \text{NdFeO}_3 \).

Fig. 3. Hysteresis loops for \( \text{NdFe}_{0.9}\text{Mn}_{0.1}\text{O}_3 \) above the spin reorientation transition.

Fig. 4. Hysteresis loops for \( \text{NdFe}_{0.9}\text{Mn}_{0.1}\text{O}_3 \) below the spin reorientation transition.

Fig. 5. Temperature dependence of heat capacity shows a Shottky-type of anomaly at \( T_{N2} \) and another anomaly at \( T_{max} \).

Substitution of Mn for Fe shifts the Shottky-type anomaly from \( T_{sh} = 2 \text{ K} \) for \( \text{NdFeO}_3 \) [3] to higher temperatures, reaching value \( T_{sh} = 2.76 \text{ K} \) for \( \text{NdFe}_{0.9}\text{Mn}_{0.1}\text{O}_3 \). Another anomaly at about \( T_{max} = 11 \text{ K} \) is generated by doping. The anomaly is smeared out by magnetic field, confirming magnetic origin of the anomaly (Fig. 5).

In conclusion, the substitution of Mn for Fe shifts the Shottky-type anomaly to higher temperatures and reduces Fe-Fe magnetic interactions leading to a decrease of \( T_{N1} \). Anomalies in AC susceptibility and heat capacity data at \( T_{max} \) indicate new magnetic interactions in \( \text{NdFe}_{x-1}\text{Mn}_x\text{O}_3 \).

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References