Measurements and Theoretical Calculations of Magnetic XAFS for Ni-Mn Alloys in Ordered and Disordered States

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Abstract We have measured the Mn *K*-edge magnetic EXAFS and XANES (*K*-XMCD) of $Ni_{0.75}Mn_{0.25}$ alloys prepared by various annealing times. The 2nd peaks in the Fourier transform attributed to Mn-Mn pairs in the ordered phase are greatly enhanced in comparison with those in conventional EXAFS. This result suggests that some of Ni atoms in the 2nd shell are replaced by Mn atoms, which have large magnetic moment in comparison with Ni atoms, due to heat-treatment induced atomic ordering. Relativistic theoretical calculations well explain the observed magnetic EXAFS and *K*-XMCD. We estimate that the ratio of the magnetic moments of Mn to that of Ni, which increases as annealing time. It is suggested that the magnetic moment of Mn atom increases with increase of the ordered phase by annealing.

Keywords: Ni-Mn alloy; magnetic XAFS; XMCD
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INTRODUCTION

The correlation between the structure and magnetism of 3*d* transition metal alloys has long been an important topic in condensed matter physics. The Mn based alloys have recently attracted special attention because of their potential applications in the magnetic recording technology. Ni_3Mn alloy can undergo the structural phase transition and that magnetic behavior is sensitive to their atomic arrangement [1]. Ordered Ni_3Mn is a typical ferromagnet [2] and the magnetic state in the disordered Ni_3Mn alloy is controlled by ferromagnetic interactions between Ni-Ni and Ni-Mn nearest neighbor pairs and by antiferromagnetic interactions between Mn-Mn second nearest neighbor pairs. The magnetic moment distributions were studied in disordered $Ni_{1-x}Mn_x$ (x=0.05-0.20) alloys by the polarized-neutron diffuse-scattering [3]. The magnetic moment of Mn atom decreases as the concentration of Mn, and two explanations are proposed: the ferromagnetic component of Mn atom vanishes by (1) random orientation and antiferromagnetic coupling or by (2) time averaging over up- and down-spin magnetic configurations on the Mn atom due to quantum and/or thermal fluctuation. The magnetic moment distribution is complicated and not unambiguously defined by the neutron measurement. On the other hand, magnetic XAFS is a powerful technique for the investigations of local magnetic structure in such alloys that undergo order-disorder structural transition. Miyanaga *et al.* presented Mn *K*-edge XMCD results for $Ni_{1-x}Mn_x$ (x=0.2, 0.24 and 0.25) [4] and the magnetic EXAFS for Ni_3Mn [4,5]. But the change of local magnetic structure with the atomic ordering is still unclear. In this paper, we discuss the results of experimental and theoretical magnetic XAFS of Ni_3Mn alloys in various ordered states.

EXPERIMENT AND DATA ANALYSES

The foil sample of $Ni_{0.75}Mn_{0.25}$ was prepared by polishing carefully and annealing for 50 (Exp.I), 100 (Exp.II) and 500 (Exp.III) hours at 693 K under Ar atmosphere to make certain ordered states. The saturation magnetization and long range order [1] for these samples are listed in Table 1.

The Mn *K*-edge magnetic EXAFS spectra were obtained in transmission mode at BL39XU in SPring-8 at 20K. The X-ray was monochromatized by a Si(111) double crystal monochromator and then circularly polarized by a diamond (111) X-ray phase retarder, with polarization P_c = 0.99[6]. The XMCD spectra were taken under a magnetic field of 1.2 T, which is

strong enough to saturate the magnetization of the present samples.

We perform relativistic theoretical calculation [7] of the magnetic EXAFS for binary alloy as the ordered model of Ni_3Mn including up to 4th shell atoms. To calculate the phase shifts for the magnetic EXAFS, we refer to the electronic structures obtained from FLAPW method [4].

RESULTS AND DISCUSSION
Magnetic EXAFS

Figure 1 (a) shows magnetic EXAFS $k\chi(k)$ spectra and (b) their Fourier transforms for three systems: Exp.I, II and III. The amplitudes of $k\chi(k)$ depend on the annealing time (or saturation of magnetization).

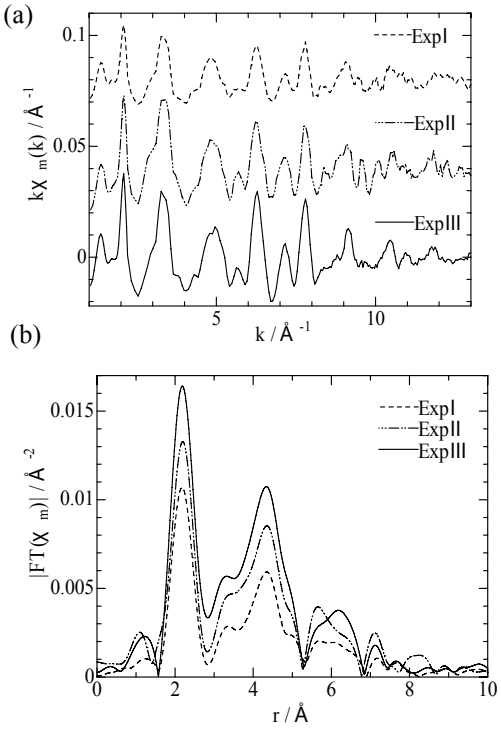

FIGURE 1. (a) Mn *K*-edge magnetic EXAFS $k\chi(k)$ spectra and (b) their Fourier transform for Exp.I, II and III.

All the peak heights in the Fourier transform depend on the annealing time. Relative intensity of the second peak attributed to Mn atoms is prominent in comparison with conventional EXAFS [5]. This indicates that Ni atoms are replaced with Mn atoms in the second coordination shell, which have large magnetic moment, by appropriate annealing.

To discuss local magnetic structures in more detail, we perform theoretical calculations of magnetic EXAFS in the relativistic framework [7]. Figure 2 shows the comparison (a) magnetic EXAFS $k\chi(k)$ and (b) their Fourier transforms between the calculated and Exp.III for the ordered model.

Since the present calculation is done in the single scattering approximation, the agreement is not perfect but that of the first and second nearest neighbor is satisfactory. We can evaluate the ratio of the magnetic moment on Mn and Ni atoms using the ratio of the first and second peak intensities [5]. The evaluated ratios of the magnetic moment of Mn and Ni are summarized in Table 1.

TABLE 1. Sample magnetization, long range order (LRO) [1] and ratio of the magnetic moment for Mn and Ni [5].

Ni_3Mn	Anneal	Magnetization	LRO[1]	Ratio of Mn/Ni[5]
Exp.I	50 hr	50.0 emu/g	0.44	1.6
Exp.II	100 hr	56.4 emu/g	0.54	2.3
Exp.III	500 hr	85.9 emu/g	0.78	2.7

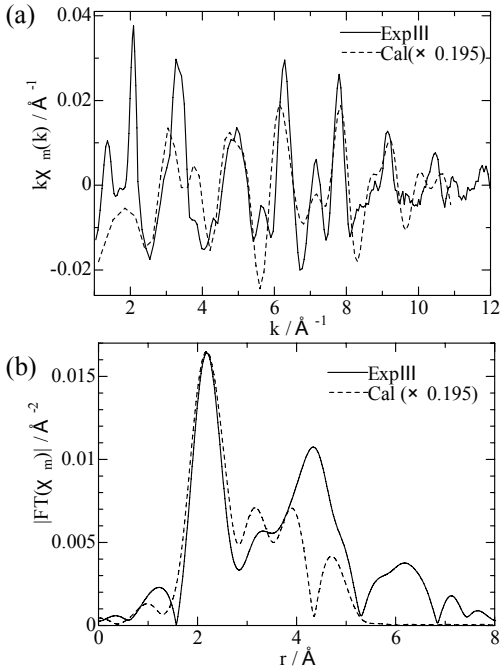

FIGURE 2. (a) Measured (Exp.III) and the calculated magnetic EXAFS $k\chi(k)$ (Exp.III) and (b) their Fourier transforms.

This result indicates that the magnetic moment ratio Mn/Ni slightly increases with the sample magnetization (or LRO) but we cannot determine whether the magnetic moment of Mn atom increases or that of Ni deceases with the magnetization.

K-edge XMCD

To answer the problem discussed in previous section, we analyze *K*-edge XMCD (magnetic XANES). Figure 3 shows the Mn *K*-XMCD for Exp.I, II and III, normalized by edge jump of conventional XAS. The larger peak intensities are observed for the longer annealing time without peak shifts.

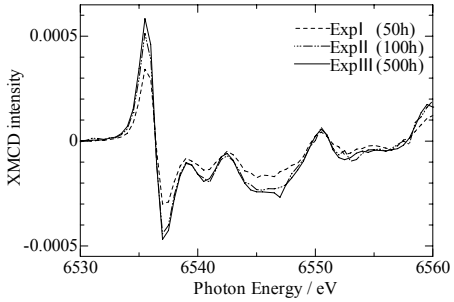

FIGURE 3. Mn *K*-XMCD spectra for Exp.I, II, and III.

To analyze the *K*-XMCD spectra, we carried out relativistic full multiple scattering calculation [7]. Figure 4 shows the comparison between the measured *K*-XMCD (Exp.III, 500h) and the calculated one. The magnetic moments of Mn and Ni are referred in the literature [4]. The agreement is satisfactory.

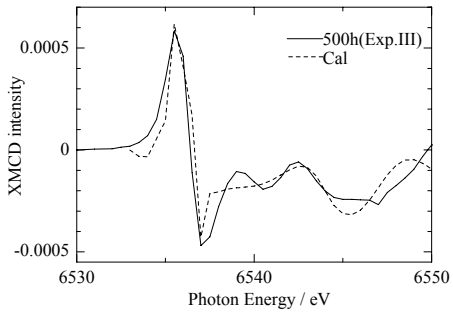

FIGURE 4. Comparison between the measured *K*-XMCD (500h, Exp.III) and the calculated one.

Figure 5 shows the calculated Mn *K*-XMCD for different magnetic moments [μ_B] on Ni atom (that on Mn is fixed to 3.2μ_B). In Fig. 5 the main positive peak around 6535eV is quite sensitive to the magnetic moments on Ni atom. The moment 0.6μ_B on Ni gives a negative peak in that region, so that we can say that μ_B (Ni)(>0.8) does not change for the annealing condition. Figure 6 shows the same result for different magnetic moments on Mn atom (that on Ni is fixed to 0.8μ_B). In this case the main peak position is not so sensitive to the magnetic moments on Mn atom in comparison with Ni, whereas a peak around 6550eV shifts depending on μ_B (Mn). Figure 3 shows, however, no peak shift, which suggests that μ_B (Mn) does not show so large change by the annealing. Combining with the magnetic EXAFS results discussed in previous section, it is suggested that the magnetic moment of Mn increases slightly with increase of the volume of ordered phase by thermal annealing, although the possibility of decreasing the Ni magnetic moment is low.

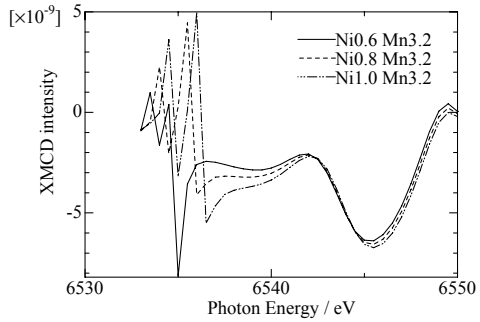

FIGURE 5. Calculated Mn *K*-XMCD with variation of the magnetic moments [μ_B] of Ni atom.

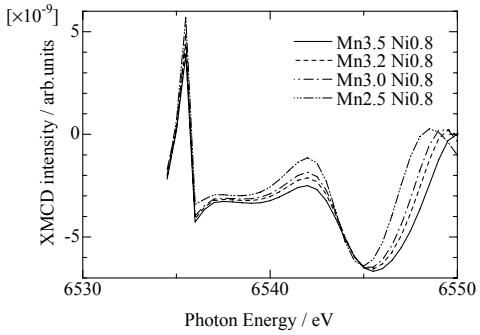

FIGURE 6. Calculated Mn *K*-XMCD with variation of the magnetic moments [μ_B] of Mn atom.

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REFERENCES

1. T. Okazaki, *Jpn. J. Appl. Phys*., **34**, 1537-1541 (1995).
2. A. Paoletti and F.P. Ricci, *J. Appl. Phys.,* **34**, 1571-1572 (1963).
3. J.W. Cable, and H.R. Child, *Phys. Rev. B*, **10**, 4607-4615 (1974).
4. T. Miyanaga, T. Okazaki, R. Maruko, K. Takegahara, S. Nagamatsu, T. Fujikawa, H. Kon, Y. Sakisaka*, J. Synchrotron Rad*., **10**, 113- 119 (2003).
5. T. Miyanaga, T. Okazaki, R. Maruko, S. Nagamatsu, T. Fujikawa, Y. Sakisaka, *Physica Scripta*, **T115**, 632-634 (2005).
6. M. Suzuki, N. Kawamura, M. Mizumaki, A. Urata, H. Maruyama, S. Goto, T. Ishikawa*., J. Synchrotron Rad*., **6**, 190-192 (1999).
7. T. Fujikawa and S. Nagamatsu, *J. Elect. Spectroscopy and Rel. Phen.* **129**, 55- 69 (2003).