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EELS Study of Martensitic and Magnetic Transitions in a Polycrystalline Ni-Mn-Ga Alloy

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Ferromagnetic shape memory alloys has attracted increased attention because of its important magnetically induced strain. The Ni_2MnGa Heusler alloy undergoes a reversal martensitic transformation, from cubic to tetragonal, showing a ferromagnetic transition. The presence of ferromagnetism offers a new handle on the martensitic transformation: by applying a magnetic field there exists the possibility either of inducing a transformation between the austenite and martensite or by rearranging the variants of martensite. In this work we study changes in electronic structure of a non stoichiometric Ni_2MnGa alloy using Electron Energy Loss Spectroscopy (EELS).

Polycrystalline Ni-Mn-Ga Alloy has been prepared in an induction furnace with an argon rich environment. A heat treatment was carried out at 1000°C for 1 hour, followed by 2 days at 800°C to obtain an ordered structure. The martensitic transformation and ferromagnetic transition temperatures have been determined from calorimetric methods and found to be ~30°C and ~100°C respectively. TEM specimens of Ni-Mn-Ga alloys were placed on a Heating Specimen Holder. EELS spectra were obtained during *in situ* heating at 20, 80 and 150°C in order to acquire spectra from martensite (ferromagnetic), austenite (ferromagnetic) and austenite (non-ferromagnetic).

Electron energy loss spectra were obtained using a Gatan Parallel Electron Energy Loss Spectrometer (PEELS model 766) attached to a Philips CM-200 transmission electron microscope. Spectra were acquired in diffraction mode with 0.3 eV/ch dispersion, an aperture of 3 mm and collection semi-angle of about 10 mrad. The resolution of the spectra was determined by measuring the full width at half-maximum (FWHM) of the zero-loss peak and this was typically close to 1.8 eV, when the TEM was operated at 200 kV. The EELS spectra were corrected for dark current and readout noise. The channel to channel gain variation was minimized by normalizing the experimental spectra with independently obtained gain spectrum of the spectrometer. Next, spectra were background-subtracted by fitting the pre-edge backgrounds with a power-law function and then Fourier-Ratio deconvoluted to remove multiple scattering components.

Figures 1a and 1b show Mn and Ni L_{23} edges respectively, for 20, 80 and 150°C, where spectra have been shifted up for clarity. At first sight it is evident a change in size and shape in L_2 and L_3 white lines during the martensitic transformation (20°C – 80°C) and ferromagnetic transition (80°C – 150°C). The ratio of the intensities L_3/L_2 and the total intensity $L_{23} = L_2 + L_3$, when properly normalized, can be used to probe the number of holes in 3d bands. In order to carry out this analysis we need to isolate the white line intensities from the edge tail. Here we closely followed the method by Pearson et al [1] and the results are summarized in Table 1. Since the L_2 and L_3 edges are related to the spin-orbit coupling of 3d electrons, the integrated intensity ratios of the white lines is related not only to the distributions of the *d* electrons but also to the magnetic moment of the ions. Higher L_3/L_2 might indicate a higher local magnetic moment of the elements. We also carried out a low-loss analysis of spectra and obtained, after a Kramers-Kronig analysis, the imaginary part of the dielectric function ε_2. Fig. 3 shows a plot for ε_2*E curves for 20, 80 and 150°C. It is noted that some peaks, associated with inter-band transitions, have been enhanced during the ferromagnetic transition.

References

[1] D.H. Pearson, C.C. Ahn and B. Fultz, Phys. Rev. B **47** (1993) 8471

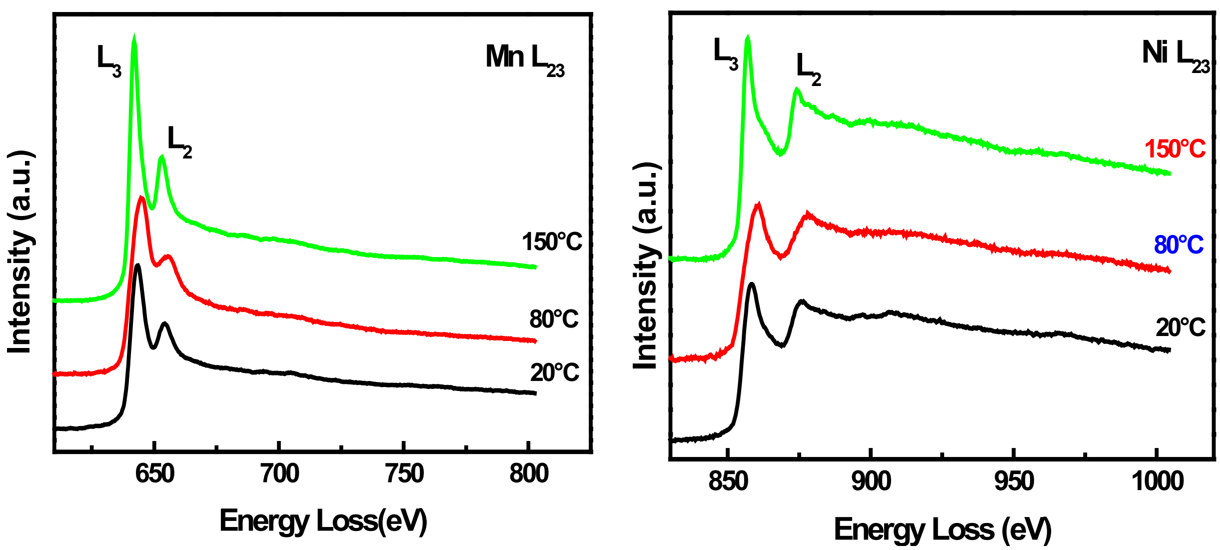

Fig.1a (left) Mn L_{23} white lines and Fig.1b (right) Ni white lines, for Martensite ferromagnetic (20°C), Austenite ferromagnetic (80°C) and Austenite paramagnetic (150°C).

TABLE 1. Normalized white lines intensities L_{23} and white lines ratio L_3/L_2

T (°C)	L_3/L_2		L_{23}	
	Mn	Ni	Mn	Ni
20	2.55	2.53	0.60	0.24
80	2.62	2.35	0.76	0.29
150	2.54	1.99	0.72	0.30

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