EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

ESRF

ESRF	**Experiment title:** An absorption spectroscopy study of the Invar behaviour in metastable FeNi alloys	**Experiment number**: 25-01-692
Beamline: BM-25	**Date of experiment**: from: 5 November 2008 to: 7 November 2008	**Date of report**: 28/2/2011
Shifts: 6	**Local contact(s)**: German Castro	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Pedro Gorria*, Department of Physics, University of Oviedo, Spain
Roberto Boada*, ICMA, University of Zaragoza, Spain
Jesus Chaboy*, ICMA, University of Zaragoza, Spain
Jesus A. Blanco*, University of Oviedo, Spain

Report:

For more than a century ago the invar problem [1], characterized by the anomalous physical properties of $Fe_{64}Ni_{36}$ (the most well-known one for its technical applications, being the almost vanishing thermal expansion of this material at room temperature) has been the focus of considerable activity for both experimentalists and theorists [2–5]. These anomalous properties (which include also strong pressure dependences of the Curie temperature, T_C, or the elastic bulk modulus, B) were thought to be closely linked to one another through the magnetovolume coupling and the existence of two low-spin (LS) and high-spin (HS) quantum states. This idea was partially supported by a series of experiments [6] in which the polarization dependence of forbidden-phonon modes along the ⟨001⟩ direction suggested the magnetic moment in this material was dependent on the 3d-3d near neighbour distances, and then it could be quite sensitive to pressure or stress effects. The appearance of this mode seems to be related to the invar composition, for it was not seen under identical conditions neither in pure nickel nor in the closely similar $Fe_{50}Ni_{50}$ composition. High-energy ball milling technique is nowadays widely used because it allows obtaining diverse solid materials far from the thermodynamic equilibrium and with different degrees of disorder, from amorphous alloys such as FeZr to FeNi solid solutions. Moreover, this technique can be used to induce drastic microstructural changes in a bulk alloy, such as nanostructure formation, due to severe mechanical treatment. In this way, we have investigated the effect of mechanical stress on the invar properties of $Fe_{64}Ni_{36}$. Commercial powders of $Fe_{64}Ni_{36}$ (Standard) were milled in a high-energy planetary ball mill under controlled Ar atmosphere, and further heated up to 1073 K (MS). The Figure 1 shows the XANES spectra recorded at the Fe and Ni K-edges in both Standard and MS samples together with those of bcc Fe and fcc Ni foils. Hence, the fcc crystal structure for both FeNi samples is confirmed. Moreover, while the Ni K-edge of Standard and MS samples overlaps at the near-edge region, a clear difference is detected at the Fe K-edge. This region of the absorption spectrum is extremely sensitive to the modification of the density of states (DOS), hence, the mechanical milling induces changes in the electronic state (or DOS) of the Fe atoms while the Ni ones remain unaffected.

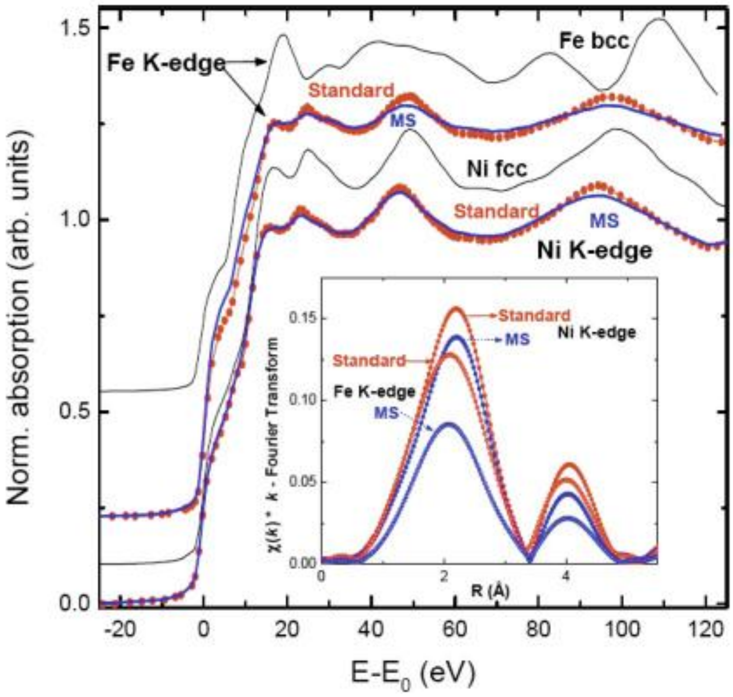

Figure 1 Comparison of the XANES spectra at the Fe and Ni K-edges for both Standard (red points) and MS (blue solid line), together with Fe and Ni foils (black lines). Inset: Fourier transforms (FT) of the EXAFS signals.

Indeed, the different effect of the milling on both Fe and Ni atoms also extends to their local structural environment. The amplitude of the EXAFS oscillations decreases and this effect is considerably greater at the Fe K-edge than at the Ni K-edge. Moreover, the modulus of the Fourier transforms (FT) of the EXAFS signals at the Ni K-edge is ~10% smaller in the MS sample than in the Standard one, while this reduction is ~40% at the Fe K-edge (see inset Fig. 4). These results indicate a more disordered local environment of the Fe atoms than that of Ni ones in the MS sample. In summary, an intrinsic physical property – the magnetic ordering temperature – in a ferromagnetic $Fe_{64}Ni_{36}$ invar alloy has been modified ($\Delta T_C \approx 70$ K) by means of severe mechanical stressing followed by a heating up to 1073 K. Neutron diffraction experiments evidence that the invar character of the sample is preserved, with a similar enlargement of the temperature range for low thermal expansion. Room temperature XRD under high pressure shows differences in the anomalous pressure dependence of the lattice parameter, with an increase of 2 GPa for the critical pressure. A more disordered local environment around Fe atoms together with the slight diffraction peak broadening, sustain an explanation based on the strain induced modification of the Fe–Fe interatomic distances, due to strong magneto-elastic coupling. To elucidate whether the enhancement of T_C in FeNi invar alloys being stressed occurs via the modification of the electronic structure, requires further investigation. **This work has been published in Phys. Status Solidi RRL 3, No. 4, 115-117 (2009)**

References

[1] C. E. Guillaume, CR Acad. Sci. 125, 235 (1897).
[2] E. F. Wassermann, J. Magn. Magn. Mater. 100, 346 (1991).
[3] M. van Schilfgaarde et al., Nature (London) 400, 46 (1999).
[4] I. A. Abrikosov et al., Phys. Rev. B 76, 014434 (2007).
[5] J. F. Valderruten et al., J. Phys.: Condens. Matter 20, 485204 (2008).
[6] P. J. Brown et al, J. Phys.: Condens. Matter 8, 1527 (1996).