Experimental report

11/10/2018

Proposal: 1-04-126 **Council:** 4/2017

Title: Influence Fe addition on structural and magnetic properties of Ni45Co5Mn25-xFexGa20Cu5 (x = 0, 1, 2, 4, 5, 6 and 8) Heusler alloys

Research area: Materials

This proposal is a new proposal

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Samples: Ni45Co5Mn7Fe8Ga20Cu5
Ni45Co5Mn21Fe4Ga20Cu5
Ni45Co5Mn20Fe5Ga20Cu5
Ni45Co5Mn19Fe6Ga20Cu5
Ni45Co5Mn24Fe1Ga20Cu5
Ni45Co5Mn23Fe2Ga20Cu5
Ni45Co5Mn25Ga20Cu5
Ni45Co5Mn23Ga22Cu5
Ni45Co5Mn20Fe5Ga20Cu5

Instrument	Requested days	Allocated days	From	To
D1B	0	2	30/05/2018	01/06/2018
D20	3	0		
D3	6	10	01/06/2018	11/06/2018
D10	10	0		

Abstract:

Recently, Ni-Mn-Ga based high temperature magnetic shape memory alloys have become the focus of intense research due to the observation of magnetic-field-induced strain at high temperatures, giving rise to many potential applications as contactless actuators in areas such as aerospace or automotive industries. For these compounds the martensitic structure and magnetic order are key parameters for the output strain obtained, and shown to be extremely sensitive to the composition of the alloys. We propose to carry out a neutron diffraction study in order to clarify experimentally the influence of the Fe addition on the martensitic transformation and the magnetic interactions in a series of polycrystalline and single crystalline samples of composition Ni45Co5Mn25 xFexGa20Cu5 (x=0, 1, 2, 4, 5, 6 and 8). In these alloys the effect of the Fe doping, promotes a simultaneous increase of Curie temperature and spontaneous magnetization, but a decrease of the martensitic transformation temperatures. The obtained results could be correlated with changes in the martensitic transformation and magnetic moment, key parameters for the magnetic-field-induced-martensitic-transformation.

EXPERIMENT N°: 1-04-126 **INSTRUMENTS:** D1B and D3

DATES OF EXPERIMENT 30/05/2018-11/06/2018

TITLE: Influence Fe addition on structural and magnetic properties of Ni45Co5Mn25-xFexGa20Cu5 (x = 0, 1, 2, 4, 5, 6 and 8) Heusler alloys

REPORT 19/09/2018

Shape memory alloys with a Heusler structure represent new multifunctional materials capable displaying large controllable strains through twin boundary motion in by applying temperature, stress, magnetic field or a combination of them to the alloys in the martensite (tetragona) phase. In particular, Ni-Mn-Ga alloys have shown extremely large field-induced strains up to 10% for modulated martensites and 12% for non-modulated ones at room temperature, giving rise to many potential applications as contactless actuators in areas such as aerospace, automotive industries etc. Recently, multicomponent based on Ni-Mn-Ga alloys with substitutional elements such as Cu, Co and Fe have become the focus of intense research. This is because they have shown magnetic-field-induced martensitic transformation at temperatures higher than the bas ternary alloys. Detailed studies of the magnetic moment's dependence with composition in Ni-Mn-Ga alloys, conclude that the magnetic interactions between the Mn atoms in these alloys are very sensitive to the exact site occupancy of the different atomic species, something that can be determined with high accuracy by neutron diffraction. Thus, in these new alloys, the site occupancy of the substitution atoms is an important issue to understand the alloying effect on the final properties.

The aim of the neutron measurements was to experimentally clarify the atomic ocupancy and magnetic contribution of the different atoms in a series of $Ni_{45}Co_5Mn_{25-x}Fe_xGa_{20}Cu_5$ (x = 0, 1, 2, 4, 5, 6 and 8) in powder and single crystal form.
The neutron diffraction study of the seven polycrystalline samples in powder was performed in the D1B diffractometer, using a wavelength of λ=1.28 Å. The diffraction patterns were recorded in a temperature range from 250 K (well into the martensite (tetragona) phase) to 540 K (in the austenite (cubic) paramagnetic phase), performing long measurements (30 min) at the ferromagnetic and/or paramagnetic martensite and austenite phases for each sample. These temperatures were different for each sample depending on their martensitic transformation (Tm) and Curie temperatures (Tc). The neutron diffraction patterns were fitted using FULLPROF. The results from the powder samples measured on D1B show a trend for of each substituted atoms in a specific site. In the case of Cu and Co the atoms mainly occupy the Mn and Ga sites respectively, but in the case of Fe, no preferential occupation between Ni and Mn sites is observed, as determined from Rietveld refinements.

Table 1. Site occupancy of the sample Fe1 resulted from the Rietveld refinement.

Atomic site	Coordinates	Element (Occ)
Site_Mn	(0,0,0)	Mn(0.78), Cu(0.22)
Site_Ga	(1/2,1/2,1/2)	Ga(0.78), Co(0.15), Mn(0.07)
Site_Ni	(1/4,1/4,1/4), (3/4,3/4,3/4)	Ni(1.79), Mn(0.11), Fe(0.06), Co(0.04)

Figure 1 shows a good agreement between the experimental results and the Rietveld refinement for the sample Fe1 measured at 540K (paramagnetic austenite). The structure presents an Fm-3m symmetry with cell parameters a=b=c=5.0867nm.

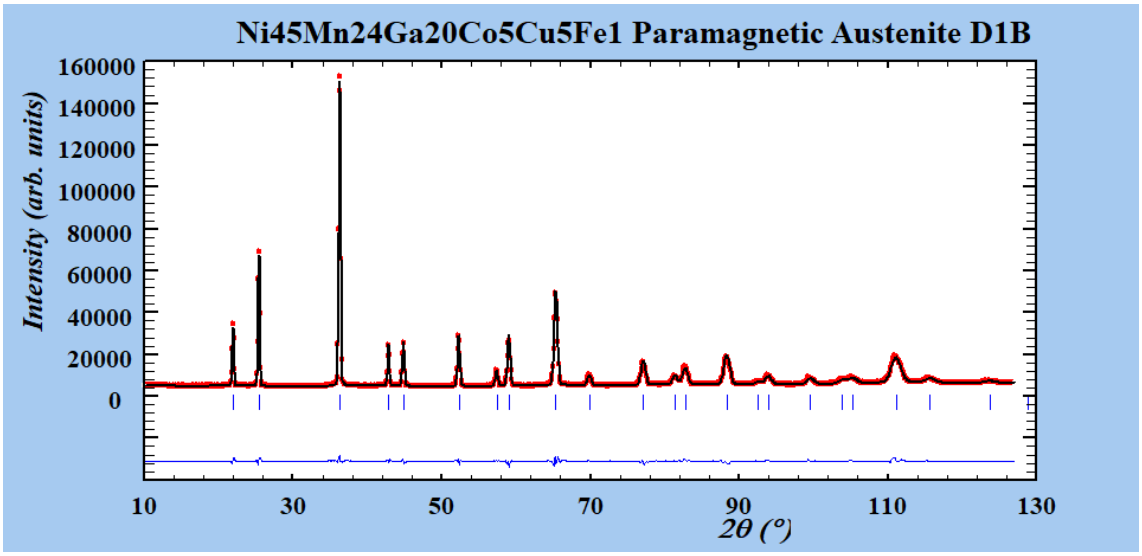

Figure 1. Rietveld refinement for the sample with x = 1

For the experiment at D3, a rectangular prism ≈3x3x4 mm of the samples SCFe4 and SCFe5 with nominal compositions $Ni_{45}Co_5Mn_{21}Fe_4Ga_{20}Cu_5$ and $Ni_{45}Co_5Mn_{20}Fe_5Ga_{20}Cu_5$ respectively; were cut from larger single crystals and heat & stress treated to obtain a single martensite variant state.

After a series of measurements on the single variant martensite state (300 K), the samples were heated up to 400 K, into the ferromagnetic austenite phase, and up to 540 K into the paramagnetic austenite phase. A new series of measurements were performed at both 400 and 540 K working at a wavelength of λ = 0.825 Å. A magnetic field of 7T was applied during the measurements.

Exhaustive work will be done to obtain the magnetic contribution of the atoms in the different sites, using the results obtained on D9 for the same samples during experiment 5-15-623, to refine the site occupancies in the single crystals.