ESRF	**Experiment title:** Short range order in magnetostrictive FeGa/MgO epitaxial layers	**Experiment number**: HC-1087
Beamline: BM30B	**Date of experiment**: from: 06/05/2015 to: 12/05/2015	**Date of report**: 08/09/2017
Shifts: 15	**Local contact(s)**: Isabelle Kieffer	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

D. Coffey, M.G. Proietti, M. Ciria, J.I Arnaudas

ICMA-CSIC-Universidad de Zaragoza, Zaragoza, SPAIN

Report:

We performed EXAFS measurements on $Fe_xGa_{(1-x)}$ epitaxial thin films grown by MBE onto MgO(001) substrates heated at different temperatures (see table I and II). The EXAFS spectra were recorded at the Fe and Ga K-edges (7112 and 10367 eV) at room temperature, in fluorescence mode. EXAFS measurements were recorded with the sample surface oriented nearly parallel (~5 deg.) to the incident beam and perpendicular to it (~85 deg.).

To perform *ab-intio* phase and amplitude calculations we generated four clusters composed by 113 and 78 atoms for the bcc and fcc symmetry respectively, with a radius of 6.1Å by the TKATOMS code. The absorber central atom was either Fe or Ga having both either Fe or Ga as scatterers atoms. The EXAFS signals for FeGa (Fe K-dge) or GaFe (Ga K-edge) systems are obtained by combining theoretical phases and amplitudes of the pure Fe and pure Ga clusters with a population factor *x*. In this way a model signal corresponding to a random distributed $Fe_{1-x}Ga_x$ alloy is obtained. We restricted the quantitative EXAFS analysis to the first peak of the FT spectrum. It corresponds, for the bcc structure, to the contribution of the $I(d_I)$ and $II(a_{par}$ and $a_{per})$ coordination shells, i.e. to the atoms at centre and at the corner of the bcc cube respectively, that due both to the small difference in distance from the central atom and to the limited R-space resolution associated to the limited spectrum k-range cannot be resolved. For the fcc structure, it corresponds to the 12 NN situated at the center of the fcc cube faces. For most of samples the FT spectra are similar to that of pure bcc Fe showing that the bcc symmetry is maintained. Nevertheless, when the substrate temperature reaches the 400ºC, the fcc symmetry becomes energetically favored and a phase change takes place as also observed by previous XRD measurements.

		Ga K-edge			Fe K-edge		
fcc sample	$T_{sub}(C)$	d_1(Ga-Fe) (Å)	%Ga	σ_1^2(Å^2)	d_1(Fe-Fe/Ga) (Å)	%Ga	σ_1^2(Å^2)
FG56	400	2.60	0.0	0.008	2.56/2.59	0.4	0.009
FG59	600	2.60	0.0	0.007	2.57/2.60	0.4	0.008
FG60	800	2.60	0.0	0.006	2.57/2.60	0.4	0.008
FG65	600	2.60	0.0	0.008	2.56/2.60	0.4	0.009

TABLE I. Best fit results for fcc films at Ga and Fe K-edges. The interatomic distance refers to Ga-Fe pairs since no Ga-Ga were observed for most samples. Statistical error are equal to 0.01 Å for d_1, 0.02 Å for a $a_{\parallel}$ and $a_{\perp}$, 0.005 for σ^2, and about ±0.1 for %Ga.

Quantitative EXAFS analysis was carried out by performing a least-squares fitting of the first FT peak contribution. on the Fourier filtered $\chi(q)$ in the range 2-12.5Å^{-1}. Simultaneous fitting of the parallel and perpendicular polarization was run for the bcc samples. For the fcc samples no difference was observed in the two polarization direction showing that the fcc lattice was

fully relaxed. The best fits results are reported in Table I and II.

For the **bcc samples** (see Table II) two kinds of fits were performed: first the chemical composition factor was defined as the same, *x*, for the I and II shell multiplying the Fe-Ga paths amplitude. Considering that the Ga is known to substitute Fe in the bcc lattice, the coordination numbers must stay equal to those of pure Fe hence the Fe-Fe paths amplitude was multiplied by (1-x). This corresponds to the hypothesis that the Ga distribution is random. A second approach was to decouple Ga concentration factor into two values, *x* for the I shell and *y* for the II one. In this way the possibility of non-random Ga distribution is allowed in the fit.

The possibility of non-random or ordered Ga distribution has to be taken into account according both to previous EXAFS and XRD results[1,2] in which the existence of different phases was observed. In all previous papers the FeGa samples studied were bulk samples whereas in our case all the samples are thin epitaxial samples. The best fits results reported in Table II(**Ga K-edge**) have been obtained by the ordered distribution model since the random distribution approach systematically gave higher R-factors. %Ga in Table II stands for *y, x* is 0 for all the films studied in this work. These results show a clear tendency of Ga to undergo an anticlustering/ordering mechanism since the Ga concentration at a local scale is remarkably lower than its average value. The values expected according to D03 ordering are $x = 0$ and $y = 0$ in the I and II Ga coordination shells. Concerning interatomic distances, d_I(Ga-Fe) ranges from 2.50 to 2.53Å, and a_{par}(Ga-Fe) and a_{per}(Ga-Fe) show values ranging from 2.88 to 2.94Å. Polarized EXAFS shows that residual tensile strain is present since a_{par}(Ga-Fe)≠a_{per}(Ga-Fe) due to the lattice mismatch with the MgO substrate. At the **Fe K-edge** (not shown for lack of room) the x values are in in all cases close to 0.5. For the II shell instead, *y* is always close to 0. This result is in agreement with the non-random D03-like Ga distribution observed at the Ga K-edge.

The evidence of a D03 long-range-order should be provided by XRD but it is not the case since no (111) reflections have been observed. The EXAFS results show that Ga has a clear tendency to anticlustering, i.e. the Ga atoms tend to stay as far as possible from each other in the FeGa lattice leading to a local D03 ordering that is the kind of atomic arrangement minimizing the number of Ga-Ga pairs. Some pairs are observed in samples FG55 and FG57 in which 1 NNN atom out of 6 is Ga. For the other samples the amount of Ga-Ga pairs is lower than the 10% according to the fit error on x (Δx=0.1). This suggests the formation of a disordered or local/modified D03 phase that makes the (111) reflection intensity practically negligible but still allowing the observed weak (001) reflection.

bcc samples	(001) peak	$T_{sub}(^oC)$	d_1(Ga-Fe)(Å)	$a_{\|}$ (Å)	$a_{\perp}$(Å)	%Ga	σ_I^2(Å^2)	σ_{II}^2(Å^2)
FG23	Yes	150	2.52	2.88	2.92	0.0	0.008	0.02
FG19	No	150	2.53	2.85	2.91	0.0	0.008	0.017
FG62	No	150	2.53	2.85	2.90	0.0	0.007	0.014
FG52	No	150	2.53	2.84	2.91	0.0	0.008	0.015
FG55	Yes	150	2.51	2.87	2.94	0.15	0.008	0.025
FG57	Yes	300	2.51	2.94	2.88	0.11	0.007	0.02
FG58	Yes	500	2.53	2.88	2.88	0.0	0.006	0.014
FG63	Yes	600	2.53	2.88	2.88	0.0	0.006	0.011

TABLE II. Best fit results for bcc films at the Ga K-edge are included. The perpendicular and parallel polarization spectra were fitted simultaneously.The interatomic distance refers to Ga-Fe pairs since no Ga-Ga were observed for most samples. For samples FG55 and FG57 the low Ga-Ga pairs presence does not allow one to determine the Ga-Ga distances that are set equal to Ga-Fe. Statistical error are equal to 0.01 Å for d_1, 0.02 Å for a $a_{\|}$ and $a_{\perp}$, 0.005 for σ^2, and about ±0.1 for %Ga .

Regarding the **fcc samples** we observe the same Ga anticlustering mechanism at the Ga K-edge found for the bcc samples. No Ga atoms are observed at the Ga sites within the fit error bar on x(Δx=0.1). At the Fe K-edge, the Ga population is found to be about 0.4, that is higher than Ga concentration x=0.25. It is not far from the value expected for the fcc L12 ordered structure, compatible with these samples composition, in which 4 Ga atoms out of 12 are expected in the Fe I coordination shell and no Ga toms are foreseen in the Ga I shell. This is in agreement with the appearance of reflection (001) in complementary XRD experiments.

References

1) S. Pascarelli el at Rhys. Rev. B **77** 184406 (2008)

2) Y. Du et al Rhys. Rev. B **81** 054432 (2010)