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Nanostructure of Giant Magnetoresistance Heterogeneous Alloys $\text{Ni}_{0.20}\text{Ag}_{0.80}$ After Annealing

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Abstract. Heterogeneous alloys $\text{Ni}_{0.20}\text{Ag}_{0.80}$, presenting giant magnetoresistance properties, have been studied by Total Electron Yield X-ray absorption spectroscopy at liquid nitrogen temperature at the Ni K edge from as-deposited to annealed stages. Up to 150°C annealing, the Ni atoms are mainly in small poorly ordered agglomerates and the local Ni atomic environment is very stable and disordered. Up to 250°C annealing, some Ni atoms occupy substitutional sites in the Ag matrix and 40 to 50 % of the Ni nearest neighbors are Ag atoms. After annealing at 400°C during 10 mn, the nanostructures evolve towards larger, more compact and well ordered granules. The Ni-Ni distance varies from 0.246 to 0.248 nm as the annealing temperature increases indicating that the small agglomerates are under strain in the Ag matrix.

1. INTRODUCTION

The recent discovery of giant magnetoresistance (GMR) in magnetic heterogeneous alloys [1, 2] has added a new dimension to the GMR phenomenon studied previously in magnetic multilayers. Heterogeneous alloys are obtained by co-deposition on a substrate of two immiscible metals : one is a ferromagnetic transition metal, here Ni, and the other one is a noble metal, here Ag. According to the substrate temperature during the deposition or subsequent controlled annealing temperatures, the formation of small magnetic domains embedded in a non magnetic matrix may be favored. Previous structural and magnetic characterization of these alloys indicates that the magnetic precipitate shape evolves from a filament or dendritic shape in as-deposited samples to a more compact granular shape after annealing [3, 4, 5, 6]. It has also been demonstrated in the $x=0.20$ alloy that after deposition and for low temperature annealing, a substantial amount of Ni is in substitution in the Ag matrix. This substitutional fraction decreases substantially above 250°C. The granule size increases with the annealing temperature (from 1-2 nm at 500 K to 8-20 nm at 900 K from magnetic measurements) as does their sphericity. The alloy $\text{Ni}_{0.20}\text{Ag}_{0.80}$, which exhibits the maximum amplitude of the magnetoresistance observed for the $\text{Ni}_x\text{Ag}_{1-x}$ alloys, has been studied from as-deposited to annealed stages i) by Total Electron Yield (TEY) X-ray Absorption Spectroscopy (XAS) at 80 K at the Ni K edge at L.U.R.E. and ii) by $\theta/2\theta$ diffraction.

2. EXPERIMENT

2.1 Samples

The samples are prepared by DC magnetron sputtering of Ni and Ag on a glass substrate. This amorphous material should not induce any particular oriented crystallization of the deposited elements. The base pressure is 2.10^{-8} Torr and the Ar pressure during a deposition is 3.10^{-3} mbar. The deposition rates are first calibrated by measuring the total thickness deposited from each target in a given time (30 mn). The relative deposition rates from the two targets are then adjusted to obtain the desired alloy composition on the substrate. The substrates are maintained at liquid nitrogen temperature during deposition in order to achieve an initial degree of intermixing between the two immiscible components as high as possible. The 200 nm thick specimens are annealed for 10 mn in a furnace with a vacuum of 10^{-6} Torr at different temperatures from 100 to 400°C. These thermal treatments lead to the formation of small magnetic particles embedded in a non magnetic matrix.

2.2 Absorption spectroscopy

These absorption spectroscopy measurements have been performed on station EXAFS 1 at the L.U.R.E. synchrotron at the Ni K edge (8333 eV) using a (331) Si channel-cut monochromator. TEY low temperature measurements were performed using a device especially designed [7] to collect data on samples cooled at liquid nitrogen temperature.

2.3 X-ray diffraction measurements

Room temperature X-ray diffraction (XRD) patterns were collected with a powder diffractometer, in the θ - 2θ configuration (symmetric Bragg geometry). Data were recorded using Cu K_α radiation with a step size of 0.02° (2θ) and a counting time of 10 s. The analysis was done in the reciprocal space (magnitude of the wave vector $s = 2 \sin\theta / \lambda$).

3. RESULTS

3.1 XANES

Figure 1 shows the XANES spectra of the $\text{Ni}_{0.20}\text{Ag}_{0.80}$ alloy as-deposited and after different thermal annealings. The XANES spectrum gives information on the local geometry around the absorbing atom. It may be used qualitatively as a "fingerprint" of the crystallographic structure by comparing the features of the XANES Ni K edge spectra of these granular alloys with other spectra. For the 400°C annealed sample, the XANES spectrum presents two peaks similar to the bulk Ni one. Hence, in these alloys, Ni may adopt the fcc structure after a 400°C annealing. However, for annealing temperature lower than 400°C , the XANES spectrum exhibits only one peak in the 8340-8355 eV range. From Montano *et al.* [8], the absence of two peaks in the XANES of a microcluster, in contrast to the XANES of the bulk metal, indicates the absence of fourth- and higher-shell atoms in the cluster. Moreover, those XANES spectra look similar to the $\text{Ni}_{0.10}\text{Ag}_{0.90}$, $\text{Ni}_{0.15}\text{Ag}_{0.85}$ and bulk Ag spectra. The very limited (up to 3 neighbors) short range order of Ni in the Ni nanoagglomerates is corroborated by the substantial amount of Ni atoms in substitutional sites of the Ag matrix.

Fig. 1 : XANES spectra for the $\text{Ni}_{0.20}\text{Ag}_{0.80}$ heterogeneous alloys as-deposited and annealed at 100, 150, 250 and 400°C compared with bulk Ni, bulk Ag, $\text{Ni}_{0.10}\text{Ag}_{0.90}$ and $\text{Ni}_{0.15}\text{Ag}_{0.85}$ corresponding spectra.

Fig. 2 : FT of the EXAFS signal of the $\text{Ni}_{0.20}\text{Ag}_{0.80}$ samples as-deposited and annealed at 100, 150, 250 and 400°C compared with the bulk Ni FT and the as-deposited $\text{Ni}_{0.10}\text{Ag}_{0.90}$ FT.

3.2 EXAFS analysis

The edge energy E_0 has been determined at the inflexion point in the edge region. The pre-edge region has been fitted with a first-order polynomial and the post-edge region by two three-order polynomials. Data are then Fourier transformed (Fig. 2) using a k^3 weighting and a gaussian window extending from 40 to 650 eV. The values reported on the x-axis of the FT graphs are raw distances from Fourier Transforms of the EXAFS signal, i.e. they are not corrected by phase shifts. The FT main peak corresponds to the Ni-Ni nearest neighbor (NN) contribution. A shoulder at the large R region of the main peak is visible on the FTs of the samples as-deposited and annealed up to 250°C and is situated at the same position than the main FT peak of the $\text{Ni}_{0.10}\text{Ag}_{0.90}$ sample. It corresponds to Ni atoms in substitution in the Ag matrix. Up to 250°C , the local Ni atomic environment is stable and a very short range order exists. At that stage, there is probably an amorphous or ill ordered structure within Ni agglomerates. The Ni-Ni radial distribution function indicates better ordering as the annealing

temperature increases. After a 400°C annealing, the three FT peaks, in the 0.3-0.5 nm range, indicate that a fcc Ni structure has formed confirming the XANES results.

The EXAFS spectra have been fitted using the EXCURV92 spherical wave $X\alpha$ model [9] by assuming two backscattering contributions Ni and Ag in the first FT peak (cf. Table I). The coordination number error is estimated to 10 %, the absolute distance error is 0.002 nm. The distance error for a relative comparison between the different sample results is only 0.001 nm. The Debye-Waller factor uncertainty is about $0.1 \cdot 10^{-4} \text{ nm}^2$. The Ni-Ni (resp. Ag-Ag) theoretical file validity has been checked with a transmission EXAFS spectrum of a fcc Ni (resp. Ag) metallic foil. After a 150°C annealing, the increase of the Ni-Ni NN distance up to the bulk Ni-Ni distance 0.248 nm suggests that small agglomerates are under strain in the Ag matrix. The coordination number indicates that the increasing annealing temperature favors Ni agglomeration.

Table : Fit results of the $\text{Ni}_{0.20}\text{Ag}_{0.80}$ samples as-deposited and annealed at different temperatures.

Sample	N	R (nm)	$2\sigma^2 (10^{-4} \text{ nm}^2)$
as-dep. Ni-Ni	4.7	0.247	1.4
Ni-Ag	4.5	0.275	2.4
100°C Ni-Ni	4.7	0.246	1.6
Ni-Ag	4.8	0.274	2.4
150°C Ni-Ni	5.5	0.248	1.5
Ni-Ag	3.9	0.276	2.4
250°C Ni-Ni	5.8	0.248	1.2
Ni-Ag	3.8	0.279	1.9
400°C Ni-Ni	11.3	0.248	0.9
Bulk Ni	12.0	0.248	0.8
Bulk Ag	12.0	0.287	0.7

Fig. 3 : X-ray diffraction spectra of the $\text{Ni}_{0.20}\text{Ag}_{0.80}$ samples as a function of annealing temperatures.

3.3 X-ray diffraction analysis

Figure 3 shows the XRD patterns limited to the reciprocal lattice area near the dense plane for the $\text{Ni}_{0.20}\text{Ag}_{0.80}$ alloy as-deposited and after a 250 and 400°C annealings. These patterns display a peak located near the theoretical position of the Ag (111) peak observed for the pure material. Hence, an Ag-rich face-centered cubic phase exists in this sample from the as-deposited to the 400°C annealed stages. Deduced from Vegard's law, the atomic concentration in nickel in the Ag-rich phase decreases as the annealing temperature increases. The atomic concentration in nickel outside the Ag-rich phase can be estimated to 90 % after a 400°C annealing. This is in good agreement with the EXAFS results as the Ni-Ag coordination number decreases as the annealing temperature increases and as the Ni-Ni NN coordination number is estimated to be slightly smaller than 12 after a 400°C annealing.

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