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Magnetotransport and micro-x-ray absorption near-edge structure studies of glass-coated Fe–Ni–Cu microwires

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Magnetotransport and micro-x-ray absorption near-edge structure (micro-XANES) studies of glass-coated 20Fe–20Ni–60Cu microwires have been carried out on both as-cast and heat-treated samples. The micro-XANES spectroscopy data were collected at the *K* edges of Cu, Ni, and Fe with the x-ray microprobe beamline at the Center for Advanced Microstructures and Devices. Comparison of the Fe *K*-edge spectra from the microwires to standard spectra reveals that the Fe atoms in the as-cast sample are in a face-centered cubic (fcc) configuration and they remain in the fcc phase throughout the annealing processes. Giant magnetoresistance (GMR) has been observed in the microwires and it reaches $\sim 6.5\%$ at 300 K in a field of 9 T. The MR decreases to $\sim 2.5\%$ as the annealing temperature increases to 500 °C. The loss of GMR upon annealing is attributed to the growth of Fe–Ni rich magnetic particles. Significant reduction in the number of extremely small particles is obtained after annealing at 500 °C, which shows MR characteristics that are different from the as-cast and 300 °C annealed microwires. © 2000 American Institute of Physics. [S0021-8979(00)57708-1]

Recently glass-coated alloy microwires have attracted great interest since they exhibit a rich variety of magnetic properties. The diameter of the microwires is in the range of 5–20 μm and the thickness of the glass coating is around 2–10 μm . Most of the studies have focused on their soft magnetic properties. They also show a large magnetoimpedance effect although they usually do not exhibit the giant magnetoresistance (GMR) effect.^{1–3} Recently, glass-coated Fe–Ni–Cu microwires prepared by the Taylor technique have been shown to exhibit negative GMR.⁴ There is no significant difference in the magnetoresistance between $I \perp H$ and $I \parallel H$ configurations, which is characteristic of granular GMR. Both dc and low frequency ac ($f < 1$ kHz) measurements exhibit essentially the same values of magnetoresistance. The observed magnetoresistance in Fe–Ni–Cu microwires is of the same origin as that found in granular GMR materials and is distinguishable from the giant magnetoimpedance commonly seen in soft magnetic microwires. It is believed to originate from the spin dependent scattering of electrons by the magnetic nanoparticles imbedded in the nonmagnetic Cu rich matrix. This speculation, however, has yet to be experimentally verified and their magnetotransport properties need to be investigated further. In this work, structural examinations by micro-x-ray absorption near-edge structure (micro-XANES) and magnetotransport studies have been conducted on both as-cast and heat-treated samples.

The glass-coated Fe–Ni–Cu microwires were prepared using the Taylor technique described in Ref. 4. They were

extracted from the lower end of a glass tube in which a Fe–Ni–Cu alloy (Fe 20%, Ni 20%, and Cu 60%) was melted by an induction heating coil. The thickness of the glass coating is 10 μm , and the diameter of the metal core is 5 μm . The microwires were later annealed for 1 h at 300, 500, and 600 °C, respectively. The x-ray diffraction data of all samples show the three major peaks due to the (111), (200), and (220) reflections of face-centered-cubic (fcc) lattices. As will be discussed later, two fcc phases, Ni–Fe rich γ_1 and Cu rich γ_2 , co-exist near the composition Fe:Ni:Cu = 20:20:60. The lattice parameters of the two fcc phases are close and not distinguishable in our x-ray diffraction data.

XANES spectroscopy was performed to elucidate the structure of the microwires. This technique was chosen due to its ability to provide local symmetry and coordination information in an element-specific manner. Micro-XANES measurements were performed using the x-ray microprobe at the Center for Advanced Microstructures and Devices (CAMD).⁵ The experiments were performed with the CAMD electron storage ring operating at 1.3 GeV with an average current of ~ 150 mA. The synchrotron radiation was monochromatized using Si(111) crystals in the Laboratorio Nacional de Luz Sincrotron (LNLS) double-crystal monochromator.^{6,7} The x-ray beam was focused using a Kirkpatrick–Baez focusing system⁸ with gold mirrors set at grazing angles of 6 mrad. The sample stage was scanned while imaging with the appropriate *K α* fluorescence signal to locate the focused x-ray beam (40 $\mu\text{m} \times 40 \mu\text{m}$) on the sample. XANES spectra were measured at the *K* edges of Fe (7112 eV), Ni (8333 eV), and Cu (8979 eV) in fluorescence mode by monitoring the *K α* fluorescence line with an energy

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FIG. 1. Micro-XANES spectra collected at the Fe, Ni, and Cu *K* edges. The micro-XANES spectra for the as-cast wire and for wires annealed at 300, 500, and 600 °C (collected in fluorescence) are shown, along with transmission spectra from polycrystalline Fe, Ni, and Cu standards.

dispersive germanium detector. Scans were performed over a range of -30 to -80 eV with respect to the appropriate absorption edge, with a step size of 0.5 eV and an integration time of 20 s. The total elapsed time for each XANES scan was approximately 1 h, and between two and six scans were performed on each sample. The spectra were summed and smoothed with a seven-point adjacent averaging algorithm to produce the spectra presented.

The results of the micro-XANES measurements are presented in Fig. 1. The XANES spectra can be used to identify the local structural environment based on “fingerprinting” of the spectra. The Ni and Cu *K*-edge spectra of the polycrystalline fcc Ni and Cu standards, respectively, can be used to identify the main features of the XANES spectra for these transition metals in a fcc structure. This consists of a pre-edge peak on the left shoulder of the edge (related to a $1s$ to a mixed p - d state) and a double feature at the edge (in the multiple scattering region of the x-ray absorption fine structure spectrum), and is followed by the first extended x-ray absorption fine structure oscillation ~ 50 eV past the edge. This can be contrasted to the body-centered-cubic (bcc) signature shown in the Fe *K*-edge spectrum of the Fe standard (a polycrystalline bcc Fe foil). By comparison to these standards, it can be seen that all the sample spectra reflect the fcc structure. For the Cu *K*-edge and Ni *K*-edge data, this is not unexpected. Cu atoms can be expected to be in a fcc environment, whether alloyed or segregated. Similarly, Ni atoms would be expected to be in a fcc environment.

The results at the Fe *K* edge are more interesting. If Fe atoms segregate out of the matrix, the Fe nanoclusters would be expected to have a bcc structure, which would be reflected in the XANES spectrum. However, the local coordination of the Fe atoms is clearly fcc in the as-cast and the annealed microwires. The magnetotransport behaviors of the microwires indicate the presence of magnetic nanoparticles dispersed in a nonmagnetic matrix.⁴ In the Fe–Cu system, such properties are due to the precipitation of nanoclusters of bcc Fe in the Cu rich matrix.^{9,10} The Fe *K*-edge XANES results indicate that Fe does not precipitate as bcc grains in this

system. Rather, the data suggest that the magnetic phase is a Ni–Fe alloy. This is consistent with the ternary phase diagram, which shows that fcc Ni–Fe (γ_1 phase) and fcc Cu (γ_2 phase) co-exist around composition the 20Fe–20Ni–60Cu over the temperatures at which the annealing was conducted.¹¹ The spinodal decomposition into γ_1 and γ_2 phases and related magnetic properties have been studied in bulk samples.^{12–15} Our micro-XANES study suggests that the Fe atoms remain in the fcc γ_1 phase throughout the annealing processes. It should be pointed out that Fe atoms have been found to exist in the bcc phase in an annealed 10Fe–10Ni–80Cu melt-spun ribbon as indicated in its Mössbauer spectrum.¹⁶ Although the melt-spun ribbon has a slightly different composition than our microwires, the phase diagram alone could not explain why the two have different forms of Fe.

The magnetoresistance of the microwires was measured in a Quantum Design physical property measurement system (PPMS) system up to a maximum magnetic field of 9 T. Magnetoresistance $MR = \Delta R/R = (R_{(H)} - R_{(0)})/R_{(0)}$, where $R_{(H)}$ and $R_{(0)}$ are resistances at an applied field H and $H = 0$, respectively, were measured in fields both perpendicular and parallel to the wire orientations. There is no significant orientation dependence in any of the samples. Figure 2 shows the room temperature MR of the as-cast, 300, and 500 °C annealed microwires when the field was applied perpendicular to the wires. Data sets were not obtained for the microwires annealed at 600 °C due to the formation of severe cracks upon annealing. At room temperature and at 9 T the MR is about 6.5% for the as-cast sample, and it decreases slightly upon annealing at 300 °C for 1 h. The loss of GMR upon annealing is attributed to the growth of γ_1 phase magnetic Ni–Fe grains. It has been shown that grains of both γ_1 and γ_2 phases undergo progressive coarsening upon annealing at increasingly higher temperatures and longer durations.¹⁵ The grain growth upon annealing has been demonstrated in our microwires in the magnetic susceptibility versus temperature plots where the blocking temperature increases with the annealing. The blocking temperature, which

FIG. 2. Room temperature magnetoresistance of as-cast and of annealed at 300 and 500 °C, respectively, glass-coated Fe–Ni–Cu microwires.

is proportional to particle volume, is less than 50 and about 130 K, respectively, for the as-cast and annealed (at 300 °C) microwires. The grain growth effectively reduces the interface region between γ_1 and γ_2 phases which is mainly responsible for the GMR. This is further illustrated by the MR data of the microwire annealed at 500 °C, where the MR is substantially reduced to about 2.5%. The data also suggest that, although the grain growth starts as low as 300 °C, rapid coarsening occurs at a higher temperature, somewhere between 300 and 500 °C. An interesting feature of the MR data taken from the microwires annealed at 500 °C is that, in addition to the much reduced magnitude, MR exhibits a large positive curvature (Fig. 2, low field region), which is absent from the as-cast and 300 °C annealed samples. The lack of any sign of saturation in MR in very high fields has been attributed to the extremely small magnetic particles typically present in granular materials; see, for example, Ref. 17. This is the situation for the as-cast and 300 °C annealed microwires. Upon heating to 500 °C, grain growth reduces the number of very fine particles, and the high field MR decreases significantly. Even in this sample the magnitude of MR increases almost linearly with the field in the high field region, which suggests that significant particle size distribution still exists in the 500 °C annealed microwires.¹⁷

Chen *et al.*¹⁸ reported a MR study on 20Fe–20Ni–60Cu and found that the MR of the sample annealed at 450 °C for 3 min was higher than the as-quenched sample. Upon further annealing for a longer time (30 min at 450 °C) or at a higher temperature (30 min 650 °C), the MR decreases again. The initial increase in MR after short annealing may be due to decomposition into the γ_1 and γ_2 phases from some single

phase region that was retained as a result of the quenching. The annealing conducted in our study was of 1 h duration and thus may not reveal any possible decomposition (if any single phase region remains in the as-cast samples) that might occur after a shorter annealing time. In addition, the starting materials and quenching conditions were different. Our starting samples were obtained by quenching from melt to room temperature directly as opposed to quenching from the solid single phase region.¹⁸ It should also be mentioned that the MR in Chen *et al.*'s report 20Fe–20Ni–60Cu was attributed to interface scattering where both the particles and matrix are ferromagnetic.

In summary, 20Fe–20Ni–60Cu microwires were studied with micro-XANES spectroscopy, which shows fcc symmetry of the Fe atoms in the as-cast and annealed samples. The Fe exists mainly in the Fe–Ni rich fcc γ_1 phase, which grows in particle size upon annealing. The magnetoresistance of the annealed samples shows a corresponding decrease in magnitude as the magnetic particles grow. A significant reduction in the number of small particles is obtained after annealing at 500 °C, which shows MR characteristics that are different from the as-cast and 300 °C annealed microwires.

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