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Changes in cluster magnetism and suppression of local superconductivity in amorphous FeCrB alloy irradiated by Ar^+ ions

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ABSTRACT

We show that cluster magnetism in ferromagnetic amorphous $Fe_{67}Cr_{18}B_{15}$ alloy is related to the presence of large, $D=150$–250 Å, α-*(Fe Cr)* clusters responsible for basic changes in cluster magnetism, small, $D=30$–100 Å, α-(*Fe, Cr*) and Fe_3B clusters and subcluster atomic α-(*Fe, Cr, B*) groupings, $D=10$–20 Å, in disordered intercluster medium. For initial sample and irradiated one ($\Phi=1.5\times10^{18}$ ions/cm^2) superconductivity exists in the cluster shells of metallic α-(*Fe, Cr*) phase where ferromagnetism of iron is counterbalanced by antiferromagnetism of chromium. At $\Phi=3\times10^{18}$ ions/cm^2, the internal stresses intensify and the process of iron and chromium phase separation, favorable for mesoscopic superconductivity, changes for inverse one promoting more homogeneous distribution of iron and chromium in the clusters as well as gigantic (twice as much) increase in density of the samples. As a result, in the cluster shells ferromagnetism is restored leading to the increase in magnetization of the sample and suppression of local superconductivity. For initial samples, the temperature dependence of resistivity $\rho(T)\sim T^2$ is determined by the electron scattering on quantum defects. In strongly inhomogeneous samples, after irradiation by fluence $\Phi=1.5\times10^{18}$ ions/cm^2, the transition to a dependence $\rho(T)\sim T^{1/2}$ is caused by the effects of weak localization. In more homogeneous samples, at $\Phi=3\times10^{18}$ ions/cm^2, a return to the dependence $\rho(T)\sim T^2$ is observed.

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1. Introduction

Amorphous and nanocrystalline Fe–Cr alloys were being intensively studied during last decades. Their good mechanical properties including variable hardness, wear resistance, chemical and irradiation stability are attractive properties for applications in atomic engineering [1]. These materials are also interesting because of their unique magnetic properties characterized by considerable variety and are determined not only by the concentrations of ferromagnetic iron and antiferromagnetic chromium, but are strongly dependent on their atomic order [2–4]. According to the publications, the most representative results on the interaction between magnetic moments of iron and chromium are the effects observed in layered GMR structures [5].

Recently we have shown that along with the traditional applications, ferromagnetic Fe–Cr alloys are interesting due to the effects related to local superconductivity observed at low temperatures [6,7].

Coexistence of ferromagnetism and superconductivity in the solids is a very interesting topic [8,9]. In amorphous $Fe_{67}Cr_{18}B_{15}$ alloy, superconductivity originates due to the presence of α-(*Fe, Cr*) magnetic clusters 180–230 Å in size occupying ~0.4–0.5% of total volume of the sample [6]. Local superconductivity in ferromagnetic medium can be considerably enhanced under irradiation by Ar^+ ions with the energy of 30 keV at a dose up to 1.5×10^{18} ions/cm^2. After irradiation, there is a more than 10 times increase (from 0.4–0.5% to 7–8%) in concentration of superconducting phase [7]. At the same time, after irradiation, the cluster size decreases down to 130–150 Å leading to a considerable increase in sensitivity of superconducting state to external field [7].

The ion irradiation has been chosen as the tool for initiation of non-equilibrium state of amorphous alloy stimulating phase separation in α-(*Fe, Cr*) magnetic clusters [7]. In this case, the atoms

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of iron concentrate mainly in the central parts of the clusters and the atoms of chromium are pushed aside to their periphery. Superconductivity arises in the cluster shells, where ferromagnetism of iron is compensated by antiferromagnetism of chromium. These changes in cluster magnetism are indispensable conditions for generation of superconducting clusters.

Primarily, the main objective of this work was to increase the superconducting phase concentration up to the values close to a percolation threshold by increasing the dose of ion irradiation. In case of success, such samples could possess very high sensitivity to external field. At the same time, we also expected to observe the influence of quantum confinement effects on critical temperature of superconducting transition with decrease in size of α-(*Fe, Cr*) clusters under irradiation [10,11]. However, it turned out that at irradiation of amorphous $Fe_{67}Cr_{18}B_{15}$ ribbon, the increase in total fluence of Ar^+ ions from 1.5×10^{18} ions/cm^2 to 3×10^{18} ions/cm^2 causes full suppression of local superconductivity. So, we have carried out detailed investigations of atomic structure, electrical, and magnetic properties of the samples to determine an origin of this phenomenon.

2. Experiment

The samples of $Fe_{67}Cr_{18}B_{15}$ metglass ribbon 10 mm wide and 30 μm thick prepared by a standard melt-quench technology were irradiated in an ion accelerator by a 30 keV Ar^+-ion beam up to a total dose of 3×10^{18} ions/cm^2 at temperatures $T\leq100$ °C. In this case, each side of the sample was irradiated in turn by a fluence of 1.5×10^{18} ions/cm^2. As a result, a total dose $\Phi=3\times10^{18}$ ions/cm^2 is twice as high than the irradiation dose used in Ref. [7]. The atomic order was investigated using $CrK_\alpha-$ radiation in Debye X-ray camera with vanadic absorber of $CrK_\beta-$ radiation [12,13]. The $CrK\alpha_1$ and $CrK\alpha_2$ X-ray lines are represented by one line $\lambda_{K\alpha}=(2\lambda_{K\alpha1}+\lambda_{K\alpha2})/3=2.29092$ Å. Magnetic properties of the ribbons were measured by a SQUID magnetometer. Electrical parameters of the samples were determined by the four-probe method. The electron paramagnetic resonance spectra were investigated using a Bruker EMX spectrometer at fixed frequency $f=9.25$ GHz and $T=300$ K.

3. Experimental results

We show that the most surprising and interesting is the recurrent character of changes in the sample properties with the ion irradiation dose increase. By-turn, the changes in cluster magnetism due to ferromagnetism reinforcement in the cluster shells result in a catastrophic influence on local superconductivity: after irradiation by a fluence $\Phi=3\times10^{18}$ ions/cm^2, superconductivity collapses.

3.1. Structure and density

The investigations of X-ray diffraction patterns of $Fe_{67}Cr_{18}B_{15}$ samples in initial state and after irradiation show that for all samples the basic diffraction maximum is split into two maxima (Fig. 1) evidencing a tendency to the concentration phase separation of metallic phase, i.e. α-*(Fe, Cr)*→α-*Fe*+α-*Cr* with spatial redistribution of iron and chromium atoms in the clusters of α-phase. There are two basic phases in amorphous $Fe_{67}Cr_{18}B_{15}$ alloy: metallic, α-*(Fe, Cr)*, and boride, Fe_3B, ones. Chemical activity of boron is very high, therefore boron tends to saturate all its valence bonds and to form Fe_3B clusters representing the stablest phase for amorphous state of the investigated ribbons. Consequently, 45% of iron out of its full 67% concentration in amorphous alloy

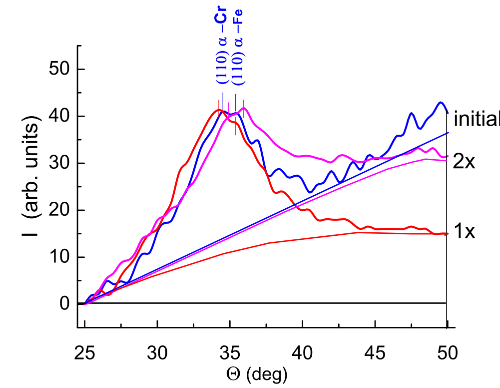

Fig. 1. Changes in the form of diffraction maximum for the samples of amorphous $Fe_{67}Cr_{18}B_{15}$ alloy in initial state and after irradiation by Ar^+ ions at fluences $\Phi=1.5\times10^{18}$ ions/cm^2 (1*x*) and $\Phi=3.0\times10^{18}$ ions/cm^2 (2*x*).

tends to be a part of boride phase. The limiting ratio for concentrations of chromium and iron in metallic phase is equal to 18/22=0.818. The pushing of chromium into peripheral areas of the clusters forms a shell, in which the conditions favorable for local superconductivity are created, where ferromagnetism of iron is compensated by antiferromagnetism of chromium [7]. Iron concentrates predominately in the central parts of the clusters. If the size of such grouping exceeds the superparamagnetic limit (for pure iron it equals 50 Å [14]), it gives the contribution to ferromagnetism.

The diffusive maxima observed above halo correspond to the positions of the (*110*) lines for α-*Cr* and α-*Fe* (Fig. 1). It is evident that the fragments of planes of the (*110*) type are the basic structural elements in the clusters of metallic phase. These fragments of the most densely packed and easily built up planes with low indices are formed still in the melt and grow up during the time of melt-quench, $\tau\sim10^{-6}$ s, at preparation of amorphous ribbon.

After irradiation with $\Phi=1.5\times10^{18}$ ions/cm^2, both diffusive maxima, (*110*) α-*Cr* and (*110*) α-*Fe*, characterizing metallic phase, are displaced towards lower angles (Fig. 1). For better understanding, one can characterize the changes in structure not by those in the angles of diffraction maxima (θ), but using the changes in corresponding interplanar spacings (d) determining a value of the deformations (ε) as $\varepsilon=(d_{irrad}\text{-}d_{initial})/d_{initial}$, where d_{irrad} and $d_{initial}$ are interplanar spacing between the (*110*) α-*Cr* or (*110*) α-*Fe* planes for irradiated and initial states of the samples, respectively. The values of deformation (ε) are shown in the table. As can be seen in Fig. 1 and the table, the angular displacement of the (*110*) α-*Fe* maximum is weak ($\varepsilon_{\alpha\text{-}Fe}=+0.3\%$) and for the (*110*) α-*Cr* one it is more considerable ($\varepsilon_{\alpha\text{-}Cr}=+\ 0.9\%$) implying the essential increase in interplanar spacings in the fragments of planes, which form cluster shells with chromium pushed out at irradiation. The stretched planes present in the clusters of metallic phase result from structural disordering of the samples at diffusion of chromium from the central parts of the clusters to their shells during ion irradiation.

On the contrary, after irradiation by a dose $\Phi=3\times10^{18}$ ions/cm^2, the diffusive maxima are displaced towards higher angles showing the appearance of compressed planes in the irradiated ribbon. Unlike the previous case, a displacement of the (*110*) α-*Fe* maximum is larger ($\varepsilon_{\alpha\text{-}Fe}=-1.2\%$) than for the (*110*) α-*Cr* maximum ($\varepsilon_{\alpha\text{-}Cr}=-1\%$).

The occurrence of compressing residual stresses after irradiation of amorphous ribbon by high ion fluence has made us to measure the density of the samples. These measurements have shown that the density of irradiated samples increases twice as much, from 3.6 g/cm^3 to 7.4 g/cm^3, verging towards the density of

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