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Enhanced magnetocaloric properties of FeZr amorphous films by C ion implantation

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ABSTRACT

For a high magnetic refrigeration efficiency, a large change of magnetic entropy ($-\Delta S_M$) associated with a high relative cooling power (RCP) are highly desirable. Here, the ion implantation is used to enhance both parameters in FeZr amorphous films. A considerable enhancement of both ($-\Delta S_M$) and RCP, from 0.66 to 1.01 J/kg K and 84.5 to 155.5 J/kg respectively, for the as-grown sample compared to the C-implanted sample is obtained for a magnetic field change of $\mu_0\Delta H = 1.5$ T. The increase of ($-\Delta S_M$) and RCP values, are attributed to the increase of the magnetization and the chemical inhomogeneity, respectively upon C implantation.

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1. Introduction

The magnetocaloric effect refers to the change in temperature of a magnetic material as a response to a magnetic field [1]. Since a long time, the magnetocaloric effect has been used for magnetic refrigeration in low-temperature physics. At present, materials exhibiting large magnetocaloric effects close to room temperature are being sought of in order to be able to replace conventional vapor compression refrigerants by magnetic refrigeration [2–4]. The magnetic refrigeration is a desirable technology due to its high efficiency, and ecological compatibility [5,6]. In order to make the magnetic refrigeration effective, large adiabatic temperature (ΔT_{ad}) and magnetic entropy ($-\Delta S_M$) changes are required [7]. A broad temperature distribution of ($-\Delta S_M$) curve is also advantageous [8,9] which should enhance the amount of heat that a magnetocaloric material can absorb during a refrigeration cycle. For this purpose, some groups have reported the increase of the working temperature region of ($-\Delta S_M$) by employing composite or two-phase compounds as working substance [10–13]. Amorphous ferromagnets can be very soft, which is essential if one aims reducing hysteresis losses. In addition it is possible to precisely tune the Curie temperature (T_C) of e.g. amorphous iron rich $Fe_{93}Zr_7$ (FeZr), by doping it with light elements, to a value around room temperature [14,15]. Ion implantation is a useful technique to accurately dope amorphous thin films. In addition, it induces a concentration gradient of the implanted element along the film thickness, which should induce a broadening of the temperature distribution of ($-\Delta S_M$). To the best of our knowledge, the use of ion-implantation as a way to obtain a broad temperature distribution of ($-\Delta S_M$) has never been reported. In this study, C ion implantation is used to enhance Tc, ($-\Delta S_M$), and RCP in a $(FeZr)C_{0.11}$ amorphous film.

2. Experimental details

FeZr amorphous films were grown in an ultrahigh vacuum system using dc magnetron sputtering. The base pressure of the chamber was below 9.10^{-10} mbar and the operating pressure of Ar gas, with a purity of 99.9999%, was 2.7×10^{-3} mbar during growth. Prior to the growth, the Si(001) substrate with the size of ($10 \times 10 \times 0.5$ mm^3) and a native oxide layer on the surface was annealed at 500 °C for 30 min in order to desorb contaminants. To facilitate an amorphous growth of the FeZr layer, we used a 5-nm-thick amorphous $Al_{70}Zr_{30}$ seeding layer, deposited from a compound target with a purity of 99.95%. The use of an $Al_{70}Zr_{30}$ seeding layer facilitates wetting, which allows having a highly amorphous character of FeZr films with absence of nanocrystallites [16]. A 40-nm-thick FeZr layer was deposited at room-temperature using a target of $Fe_{93}Zr_7$. To protect the FeZr against oxidation, a 6-nm-thick $Al_{70}Zr_{30}$ capping layer was used. The implantation of C

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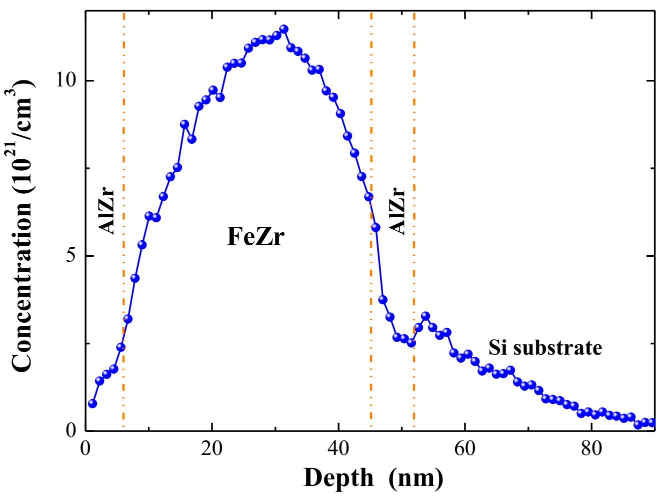

Fig. 1. Simulated change of the carbon concentration profile across a complete stack of Si(001)/$Al_{73}Zr_{27}$ (5 nm)/$Fe_{93}Zr_7$ (40 nm)/$Al_{73}Zr_{27}$ (6 nm). The simulation was carried out by the SRIM software[17] and using an ion energy of 18 keV and an implanted dose of 4.75×10^{16} ion/cm^2, which corresponds to an average normalized amount of carbon within the FeZr layer of 0.11.

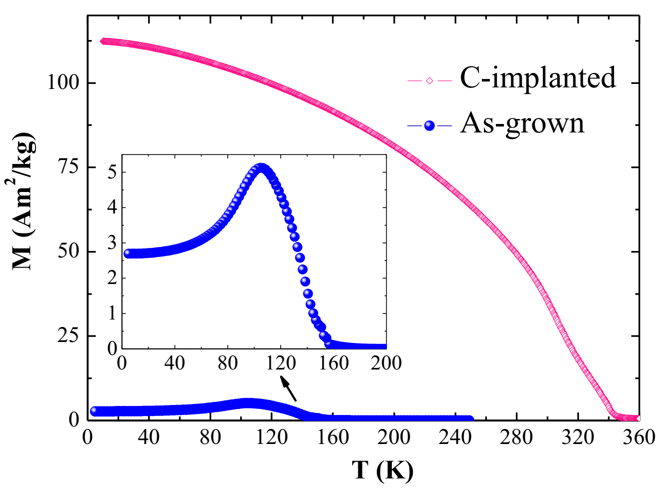

Fig. 2. Zero-field-cooled (ZFC) magnetization as a function of temperature of the C-implanted and as-grown FeZr amorphous films measured in an applied magnetic field of 0.5 mT. The inset highlights the curve for the as-grown FeZr film.

was performed ex-situ at room temperature with a Danfysik ion implanter using a mass-separated $^{12}C^+$ beam extracted from a CH_4 plasma source. The ion energy was 18 keV, and the ion dose was 4.75×10^{16} ion/cm^2, yielding films with an average amount of implanted ions normalized to the number of atoms in the FeZr layer of about 0.11, i.e. $(FeZr)C_{0.11}$. The implantation dose defines the number of C ions per unit area, which hit the sample. The implanting beam was scanned homogeneously across the entire surface of the sample and the beam current was automatically integrated during implantation to reach the desired dose with an absolute error of less than 2%. For more information on the implantation procedure see Refs. [17,18]. The magnetic measurements were recorded between 10 and 390 K using a superconducting quantum interference device (SQUID) magnetometer. The magnetic field was applied parallel to the sample surface.

3. Results and discussion

First, the composition aspect of the studied samples is discussed. Fig. 1 displays the simulated carbon depth profile through the sample obtained using the *stopping and range of ions in matter* (SRIM) software [19]. As can be observed in Fig. 1, the carbon concentration within the 40-nm-thick FeZr layer presents a distribution with a maximum at around 15 nm. One can notice that the majority of the carbon ions are inserted within the FeZr layer and merely few percents are introduced in the $Al_{70}Zr_{30}$ layers and the Si(001) substrate. The conservation of the amorphous nature of the $(FeZr)C_{0.11}$ sample was confirmed by EXAFS measurements on the Fe K-edge and AFM images confirm that only the top 1-nm of the 6-nm-thick $Al_{70}Zr_{30}$ capping layer is damaged by the ion implantation [14].

The temperature dependence of magnetization for the as-grown and C-implanted FeZr samples, measured in an applied field of 0.5 mT on heating from 10 K after cooling in zero-field, are shown in Fig. 2. As can be observed both samples exhibit a ferromagnetic behavior and the T_C increases upon C implantation. One can notice that the magnetic transition for the implanted sample is not so sharp. This is due to the fact that the implanted sample is not chemically homogenous and exhibits a gradient in the carbon composition, which prevents a sharp transition. We note that the magnetization of the as-grown sample decreases at low temperature, which is due to the non-collinear ferromagnetism of the as-grown sample. The magnetic interactions in the as-

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