Magnetic moments of Fe and Y in the FeBY glass forming system

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Abstract

The new Fe_{71.2}B₂₄Y_{4.8} glassy alloy has been prepared in amorphous ribbon form via melt spinning and splat cooling. The high degree of sample homogeneity and structural quality of the ribbon is revealed by the observation of narrow linewidths in the spectra of the field derivatives of the absorbed microwave power in ferromagnetic resonance experiments. Combination of vibrating sample magnetometry and element-specific X-ray magnetic circular dichroism experiments at the L_{3,2}-edges of Y revealed an induced magnetic moment of 0.065 μ_B /atom for Y antiparallel oriented to an Fe magnetic moment of 1.83 μ_B /atom.

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1. Introduction

Amorphous and nanocrystalline metallic alloys are considered nowadays as advanced materials which demonstrate superior properties in a wide range of conditions and have, therefore, a large number of important applications. They include materials with excellent static and dynamic mechanical strength, soft magnetic properties and good corrosion resistance, see for example [1–7]. It is not surprising then, that there is a strong tendency for searching for new multielement metallic systems with high glass forming ability (GFA) [1,6,7]. Recently, it was

reported that the addition of small amounts of Y in FeB results in a material with enhanced GFA [8,9]. The optimum stoichiometry was found to be Fe_{71.2}B₂₄Y_{4.8} [9]. Furthermore, this material exhibits high permeability as desired for magnetic transformer applications.

In the present work we have prepared this new material in form of ribbons via melt spinning and of splat via splat cooling. The samples were characterized combining conventional laboratory and synchrotron X-ray techniques. The materials were found to be completely or partially amorphous depending on the preparation method and the thickness. The high magnetic homogeneity and the soft magnetic nature of the amorphous samples were revealed. Finally, we have determined the magnetization as well as the induced magnetic moment at the Y sites in proximity with Fe. This is, to our knowledge, the first quantification

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of the Y-magnetic moment and its separation into spin and orbital moments.

2. Experimental details

The present work deals with the $\text{Fe}_{71.2}\text{B}_{24}\text{Y}_{4.8}$ glassy alloy. This composition has been chosen because it was reported in Ref. [9] to have the highest GFA among the other investigated alloys of the FeBY family. Samples have been prepared via melt spinning in the form of ribbons 1 mm wide and 20 μm thick. Since these ribbons are too narrow for the quantitative evaluation of the X-ray absorption (XAS) spectra, splats were also produced by a splat cooling device based on a melt sucking unit. The average thickness of these samples is near to 200 μm .

High-energy X-ray diffraction (XRD) measurements were performed at HASYLAB at DESY (Hamburg, Germany), on the experimental station PETRA 2, using monochromatic synchrotron radiation of 106 keV. The samples measured at room temperature in transmission mode were illuminated for 240 s by a well-collimated incident beam of 1 mm^2 cross-section. XRD patterns were recorded using a 2D detector (mar345 Image plate) in asymmetric mode to obtain data at high wave vector transfer (Q). The background intensity was subtracted directly from the 2D XRD pattern and the result was integrated to the 2θ -space, by using the program Fit2D [10]. Complementary XRD measurements were also performed using a standard powder diffractometer (Berthold-SEIFFERT) with Ni-filtered $\text{Cu K}\alpha_1$ radiation ($\lambda = 0.154059 \text{ nm}$).

X-ray magnetic circular dichroism (XMCD) measurements have been performed at the ID12 beam line of the European synchrotron radiation facility (E.S.R.F.) [11]. The Y $L_{3,2}$ -edges and the Fe K-edge have been recorded by means of the fluorescence detection mode and by using two type of undulators: EMU388 (for the Y L-edges) and Apple II (for the Fe K-edge). The intense photon flux provided by these types of undulators allows for a clear recording of even minor details of the XMCD spectra. For the XMCD experiment, a large magnetic field of 4 T was applied perpendicularly to the sample surface at temperatures of 300 K and 10 K. The errors in the analysis of the data are typical for XMCD determination of ferromagnetic moments in 4d and 5d transition metals with strong ‘white light’ intensities; they are of about 5% for the ratio of orbital-to-spin magnetic moment and 10% for the absolute values of the magnetic moments [12].

Complementary ferromagnetic resonance (FMR) measurements have been performed at 300 K with the help of a commercial Bruker bridge implemented at the auxiliary laboratories of the ID12 beam line. The frequency of the microwaves was 9.6 GHz. Finally, vibrating sample magnetometry (VSM) measurements have been performed at room temperature in the easy magnetization direction in order to determine the alloy saturation magnetization. The maximum applied field was 1000 kA/m.

3. Results

The as-quenched ribbons exhibit the characteristic diffuse scattering pattern for metallic glasses with a maximum at $2\theta = 3.27^\circ$ and pronounced oscillations, typical for amorphous materials, are visible up to $2\theta = 15^\circ$, as shown in Fig. 1. The large number of oscillations and the noise-free XRD pattern as compared to the regular ones recorded in conventional laboratories, are attributed to the use of photons of very high-energy (106 keV) in combination with the high photon flux provided by synchrotron sources such as the one at HASYLAB. The amorphous nature of the sample was also confirmed by the near-edge X-ray absorption fine structure (NEXAFS) spectrum recorded at the K-edge of Fe at E.S.R.F. (not shown here). This spectrum was practically structureless unlike the typical ones reported for crystalline Fe. For the gradual transition of the spectra from crystalline to amorphous Fe see as a reference, for example [13]. The splat-cooled sample, on the other hand, was a mixture of amorphous and crystalline structure, as its XRD pattern revealed. Besides the larger total thickness ($\sim 200 \mu\text{m}$) the dominantly crystalline structure, according to our experience, it might also be connected to a thin non-melted layer typical of ingots melted on the water-cooled hearth. It promotes crystal growth on solidification.

Fig. 2 shows the room temperature spectrum of the field derivative of the absorbed microwave power in FMR experiments for the $\text{Fe}_{71.2}\text{B}_{24}\text{Y}_{4.8}$ amorphous ribbon. The magnetic field was applied parallel to the easy magnetic axis of the sample, which is the direction parallel to the ribbon plane due to the dominant shape anisotropy. The position of the FMR, determined precisely by the derivative of the absorption spectrum, is $H_R = 66.4 \text{ kA/m}$.

Amorphous alloys have small magnetocrystalline anisotropy. FMR measurements (together with a precise value of the magnetization) are in principle suitable for the determination of magnetic anisotropy constants [14,15]; it would, however, go beyond the scope of this

Fig. 1. X-ray diffraction patterns recorded for an amorphous $\text{Fe}_{71.2}\text{B}_{24}\text{Y}_{4.8}$ ribbon.

Fig. 2. Ferromagnetic resonance signal recorded for the $\text{Fe}_{71.2}\text{B}_{24}\text{Y}_{4.8}$ ribbon. The small linewidth value indicates the high degree of structural and magnetic homogeneity.

short contribution. Most important for our purpose is the very narrow linewidth $\Delta H = 5$ kA/m of the FMR line. This is indicative for samples of high structural and magnetic homogeneity, as explained in detail in Ref. [16]. The linewidth values are as low as found in other high quality amorphous Fe-based alloys, Fe-whiskers and single-crystalline Fe/V superlattices [17].

In Fig. 3, the VSM magnetization curve is presented for a $\text{Fe}_{71.2}\text{B}_{24}\text{Y}_{4.8}$ amorphous ribbon. The magnetic field was applied parallel to the ribbon's plane (easy axis). One may notice that the sample saturates very easily. The saturation magnetization of the sample is about $142 \text{ Am}^2/\text{kg}$. Our result is comparable to recent magnetic measurements on FeBY bulk metallic glasses with similar stoichiometry [8]. The magnetization curve shown in Fig. 3, which is typical for soft magnetic materials, reveals a coercivity value of $H_C \sim 190 \text{ A/m}$. This value is quite close to the figures

Fig. 3. Vibrating sample magnetometry loop recorded at room temperature for the $\text{Fe}_{71.2}\text{B}_{24}\text{Y}_{4.8}$ ribbon, with the magnetic field applied parallel to the ribbon plane. The inset shows the magnification of the loop near small fields, revealing a coercivity value of $H_C = 190 \text{ A/m}$ typical for magnetostrictive soft magnetic materials.

reported for similar amorphous magnetic alloys, see for example [18].

In Fig. 4, the XAS and the XMCD spectra are plotted. They have been recorded at room temperature at the $L_{3,2}$ -edges of Y for the splat-cooled (partially crystalline) sample. Qualitatively similar data were also recorded for the amorphous ribbon. This is expected as the nearest neighbor atomic environments and, therefore, the distribution of magnetic moments is closely related. While the corrections for the self-absorption and saturation effects (necessitated by the relatively large sample thickness of the splats as compared to the penetration depth of the X-rays) can be carried out quantitatively, the background correction for the narrow ribbon samples cannot be made with the necessary accuracy. This is the reason why the absolute values (spin and orbital moments of the Y atom) are only determined for the splats; they are expected to be close to the parameters relevant for the amorphous alloy. For the XAS spectra the ratio of the L_3/L_2 was normalized to 2/1 according to Ref. [19]. The existence of finite XMCD signals reveals the presence of magnetic moments [15]. In our case it shows unambiguously that Y has acquired an induced magnetic moment. The XMCD signals are rather small, however the high photon flux and degree of polarization offered by the third generation synchrotron radiation facilities [20] allow us to record such small XMCD signals with large signal-to-noise ratio. By knowing the direction of the magnetic field and the helicity of the beam, we conclude that Y is polarized antiparallel to the magnetic moment of Fe. This behavior is consistent with the fact that Y is in the beginning of the 4d transition elements, i.e. it has an almost empty 4d electronic band. The practically noise-free signal has allowed us to trace also element-specific hysteresis curves at the Y L_3 -edge. As a relative measurement it was also possible to trace for the narrow amorphous ribbon sample. Fig. 5 depicts a signal proportional to the projection of the Y-magnetic moment along

Fig. 4. (Top) X-ray absorption and (bottom) X-ray magnetic circular dichroism spectra recorded at the $L_{3,2}$ -edges of Y in a splat-cooled $\text{Fe}_{71.2}\text{B}_{24}\text{Y}_{4.8}$ sample. The step-like functions used in the analysis of the XAS spectra are also plotted.

Fig. 5. Hard-axis magnetization curve for Y recorded with the help of the element-specific X-ray magnetic circular dichroism technique. The reversed path as compared to typical magnetization curves indicates the antiparallel orientation of the Y-magnetic moment with respect to the Fe one.

the direction of the applied field. The curve was recorded with the magnetic field applied in the direction normal to the film plane for the ribbon prepared from the ingot. This is a typical hard-axis magnetization curve with saturation field of about 1 MA/m. The reversed path of the curve indicates the negative sign of the Y-magnetic moment with respect to the Fe one.

In order to determine the magnetic moment of Y and separate it into spin and orbital contributions we have applied the sum-rule analysis [21,22] to the spectra of Fig. 4. Moreover, in our analysis, we considered the temperature dependence of the XMCD signals or equivalently, that of the element-specific magnetizations between 10 and 300 K, which is of the order of 10%. For the ‘white line’ intensities, one usually subtracts the continuum from the XAS. The continuum is represented by a step-function, as a first approximation, and this recipe has been successfully applied for 3d ferromagnetic elemental metals and the transition metals of the 5d series for which the ‘white line’ intensities were large [12], like for the Y case shown in Fig. 4. Our analysis provides for the total magnetic moment of Y the value $\mu_{\text{tot}} = 0.065 \mu_{\text{B}}/\text{atom} \pm 10\%$. The ratio of orbital-to-spin magnetic moment is $\mu_{\text{L}}/\mu_{\text{S}} = -0.064 \pm 5\%$, with the negative sign indicating antiparallel orientation of spin and orbital magnetic moments of Y, a result consistent with the third rule of Hund for an element with less than half-filled 4d states.

4. Discussion

The induced magnetic moment of Y is relatively small. It, however, has definitely a non-zero value, unlike a recent report for the magnetic moment of Y in YCo_2 films [19]. Whereas the local electronic structure of Y should evidently be different in YCo_2 and in amorphous $\text{Fe}_{71.2}\text{B}_{24}\text{Y}_{4.8}$

alloy due to the different atomic surroundings, the qualitatively different Y response to the neighboring magnetic moments is quite remarkable. In that case the zero magnetic moment of Y had been attributed to an almost empty 4d electronic band [19]. We believe that the very small, but finite, induced magnetic moment of Y determined, to our knowledge, for the first time in this work by XMCD, is made possible by the technical evolution of the third generation synchrotron radiation sources. One may notice that this evolution has already allowed for the determination of induced magnetic moments even smaller than the one of Y, such as the ones of Ta in CoCrPtTa alloys [23] and of Au in Co/Au multilayers [24]. Taking into account the small proportion of the Y to Fe atoms ($\sim 6.7\%$) and the ratio of the Y to Fe magnetic moments ($\sim 3\text{--}4\%$), it is safely concluded that the magnetic contribution of the induced magnetic moment of Y to the total magnetization of the FeBY samples should be only of about 0.2–0.3%.

Following the analysis described in Ref. [25], we combined the XMCD information for the Y-moment with the VSM information for the total magnetization. Furthermore, we considered the 10% decrease of the magnetization between 10 and 300 K. This way, we determined the magnetic moment of Fe to be $1.83 \mu_{\text{B}}/\text{atom} \pm 5\%$. This value is somewhat decreased as compared to the bulk-Fe moment [18]. This is evidently caused by the d–d (Fe–Y) and p–d hybridization (Fe–B) of the Fe d-electrons with the orbitals of the surrounding non-magnetic atoms. In contrast to the earlier concept of charge transfer (which is not confirmed by direct techniques as photoelectron spectroscopy) it is now understood that the change of magnetization is dominantly caused by the hybridization of the electron states. The influence of the nearest neighbor atoms is best described as the admixture of the external atomic orbitals, i.e. by the hybridization of the d-electron states (which dominate the magnetic moment) with the neighboring atom d and p-electron states [26].

5. Summary

In this work, the new $\text{Fe}_{71.2}\text{B}_{24}\text{Y}_{4.8}$ glassy alloy has been investigated in the form of ribbons and sput-cooled samples with a combination of synchrotron X-ray and conventional laboratory techniques. X-ray diffraction experiments at HASYLAB have shown that the ribbons are amorphous. This observation is further supported by the X-ray absorption fine structure spectrum recorded at the K-edge of Fe at E.S.R.F. Ferromagnetic resonance measurements carried out on the ribbons have revealed a high structural and magnetic homogeneity of the samples. Vibrating sample magnetometry in combination with X-ray magnetic circular dichroism measurements at the $L_{3,2}$ -edges of Y at E.S.R.F. have allowed for the determination of Fe and induced Y-magnetic moments; the latter ones were quantitatively determined, to our knowledge, for the first time.

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