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A new magnetic multilayer system: Iron-bismuth

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A new magnetic multilayer system: Iron-bismuth

F. Z. Cui, Y. D. Fan, and Y. Wang

Department of Materials Science and Engineering, Tsinghua University, Beijing 100084, People's Republic of China

A. M. Vredenberg

FOM Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

H. J. G. Draaisma

Department of Physics, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands

R. Xu

Solid State Physics Laboratory, University of Groningen, 9718 EP Groningen, The Netherlands

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Magnetic Fe-Bi multilayers have been, for the first time, synthesized by using electron-beam evaporation at 140 K. The relationships between film structure and magnetic properties were investigated by means of measurements of magnetization, x-ray diffraction, electron diffraction, and Mössbauer spectra. Films are ferromagnetic or paramagnetic, depending on Fe layer thickness, t_{Fe} . Films with t_{Fe} of about 1 nm exhibit perpendicular magnetic anisotropy while these with the thicker t_{Fe} have in-plane magnetism. Temperature dependence of magnetization in the Fe-Bi multilayers was also studied in the temperature range of 77–600 K.

I. INTRODUCTION

Artificially multilayered films in which one of the constituent elements is ferromagnetic in its bulk form are in interest both in magnetic materials and in magnetism theory. Among the various combinations of constituent elements in magnetic multilayers, mutually insoluble metals have attracted much attention in recent years,^{1–4} since sharper interfaces can be formed. Recently, den Broeder *et al.*⁵ experimentally found that perpendicular magnetic anisotropy of Co-Au multilayers is due to sharpening of the interfaces.

The Fe-Bi system exhibits complete immiscibility in the condensed state as shown in the binary phase diagram of Bi-Fe.⁶ Fe is a typical ferromagnet, while Bi is the most diamagnetic metal,⁷ special magnetism in Fe-Bi multilayers may be expected. Also, until this study there have been no magnetic Fe-Bi multilayers reported, although metastable Fe-Bi alloy films⁸ and proximity effect in sandwich Fe-Bi films⁹ were investigated previously. It is thus of our interest to investigate this new system of magnetic multilayers. In the present paper we describe the structure and magnetic behavior of the Fe-Bi multilayers.

II. EXPERIMENTAL PROCEDURES

The Fe-Bi multilayers were prepared by sequential deposition of Fe and Bi using dual electron-beam evaporation sources and a shutter to interrupt the particle flux at preset times. The base vacuum was of less than 5×10^{-6} Pa. The deposition rate was kept constant at a fixed value in the range of 0.05–0.1 nm/s, as monitored by a quartz resonator. During deposition, silicon wafer or NaCl substrates of the films were cooled by liquid nitrogen. The

actual temperature was detected as about 140 K. The total thickness of the multilayers were more than 150 nm and about 60 nm for silicon and NaCl substrates, respectively.

Their periodic structure was checked by x-ray diffractometry (XRD) with the scattering vector perpendicular to the film plane. $\text{CuK}\alpha$ radiation was used. Some samples with NaCl substrate were analyzed by transmission electron microscopy (TEM). The thickness ratios of Fe and Bi were checked by Rutherford backscattering spectroscopy. The thickness of the individual Fe layer, t_{Fe} , and that of Bi, t_{Bi} , were derived from the thickness ratio mentioned above and the modulation period D determined by XRD. Magnetic measurements were made by using a vibrating sample magnetometer (VSM) with an applied field of up to 1300 kA/m, both parallel and perpendicular to the film plane.

The ^{57}Fe conversion electron Mössbauer spectra (CEMS) were recorded at room temperature using a constant acceleration spectrometer with a source of 5-mCi ^{57}Co in Rh matrix. A proportional counter with a 4% CH_4 in He gas flow was used to detect the 7.3-keV K -shell conversion electrons. The voltage was around 1300 V. Velocity scale was calibrated from a standard α -Fe spectrum. All spectra were analyzed by least square fitting.

III. RESULTS AND DISCUSSION

A. Structure

The presence of a multilayered periodic structure was confirmed in the films composed of t_{Fe} down to 0.5 nm using the low-angle XRD analysis. The first- and second-order reflections usually are obvious in XRD spectra. The high-angle multilayer reflections were located around the 003 and 102 reflections of the rhombohedral Bi layers. In

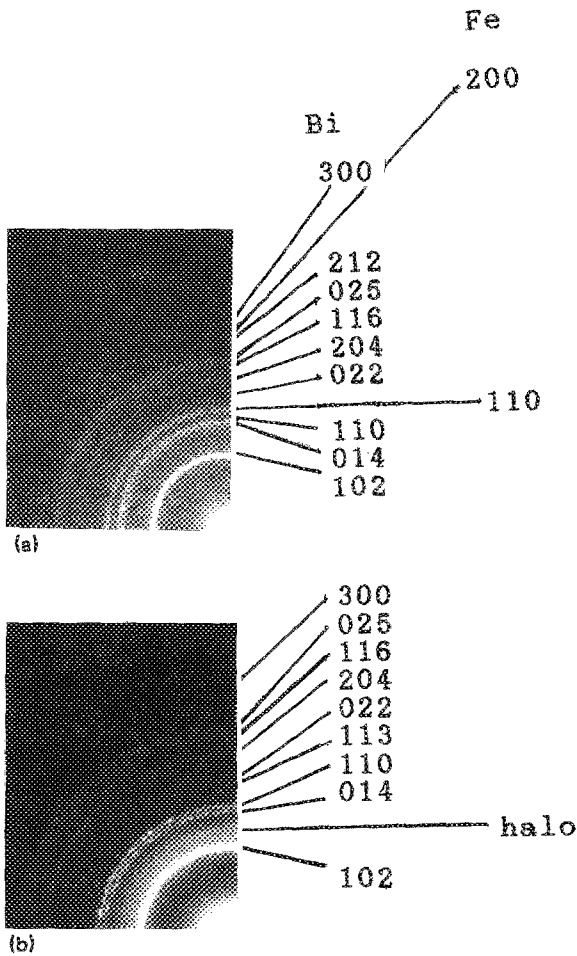


FIG. 1. Electron diffraction patterns of (a) film of t_{Fe} of 4 nm and (b) film with t_{Fe} of 1 nm. A halo at an interatomic spacing around 0.25 nm appears in pattern (b).

our XRD spectra, signal from Fe layers was always very weak relative to that from Bi.

In order to observe the dependence of the structure of Fe layers on t_{Fe} , TEM analysis was performed. It was found that the structure of Fe layer is of BCC when t_{Fe} is more than 1.5 nm, and the structure is amorphous if t_{Fe} is in the range of 0.5–1.5 nm. Figure 1 gives the electron diffraction patterns of plan-view TEM for (a) sample of $t_{\text{Fe}} = 4$ nm and (b) film of $t_{\text{Fe}} = 1$ nm. It is seen that in the case of (b), the rings belonging to pure bcc Fe are not visible, meanwhile a halo with atomic spacing around 0.25 nm appears. From the diffraction data and atomic radius of Fe ($r_{\text{Fe}} = 0.126$ nm), a disordered structure of the Fe layers may be inferred.

B. Magnetic behavior

The magnetization curves were measured for Fe-Bi multilayers with various t_{Fe} ranging from 0.5 to 5 nm, and t_{Bi} of more than 3 nm. The values of the saturation magnetization $4\pi M_s$, taken per unit volume Fe, are given in Fig. 2. It is seen that for t_{Fe} less than 2.5 nm, M_s becomes increasingly lower than the value for pure α -Fe ($4\pi M_s = 2.15$ T). For the film with t_{Fe} of 0.5 nm, which corresponds to two atomic layers of Fe, a nonmagnetic

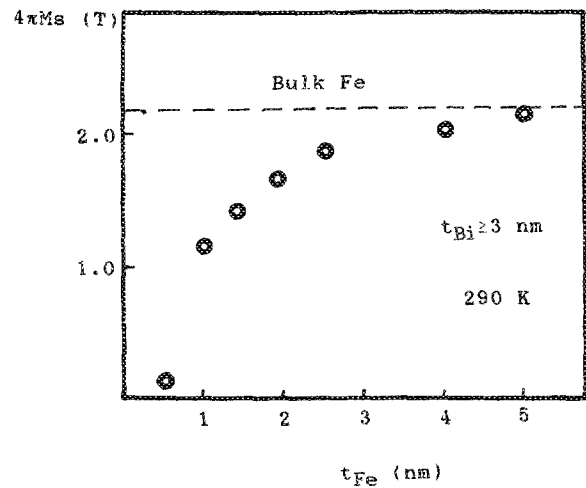


FIG. 2. Average saturation magnetization per unit Fe volume vs t_{Fe} .

state occurs. This phenomenon is called magnetic dead layer,¹⁰ which is still an interesting subject.¹¹ Our result suggests that there is an antiferromagnetic coupling of Fe atoms via Bi atoms.

Apart from a thickness dependence of M_s , as deduced from the magnetization curves, a perpendicular magnetic anisotropy in films with t_{Fe} of 1 nm was observed as shown in Fig. 3(b). For the samples with t_{Fe} more than 1.5 nm the easy axis of magnetization shifts into the film plane. Figure 3(a) shows such an example. The total anisotropy energy per unit volume Fe, K_u , was estimated to be 5.5×10^5 J/m³ in the case of Fig. 3(b), from the area between the parallel and perpendicular magnetization curves. For this sample, the shape anisotropy energy, $2\pi M_s^2$, is 4.7×10^5 J/m³, which is smaller than K_u . Thus, a state of magnetization perpendicular to the film surface is built up.

The CEMS for samples of multilayers are given in Fig. 4. The spectrum with t_{Fe} of 4 nm can be resolved into a magnetically split six-line spectrum corresponding to hyperfine field of about 330 kOe, which indicates that the structure of the Fe layer for most part is bcc. However, the

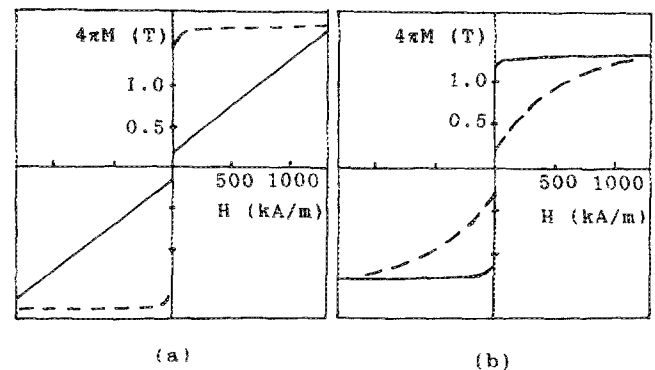


FIG. 3. Magnetization curves measured in fields parallel (dotted line) and perpendicular (solid line) to the film plane for Fe-Bi multilayers of (a) $t_{\text{Fe}} = 2.5$ nm and (b) $t_{\text{Fe}} = 1$ nm.

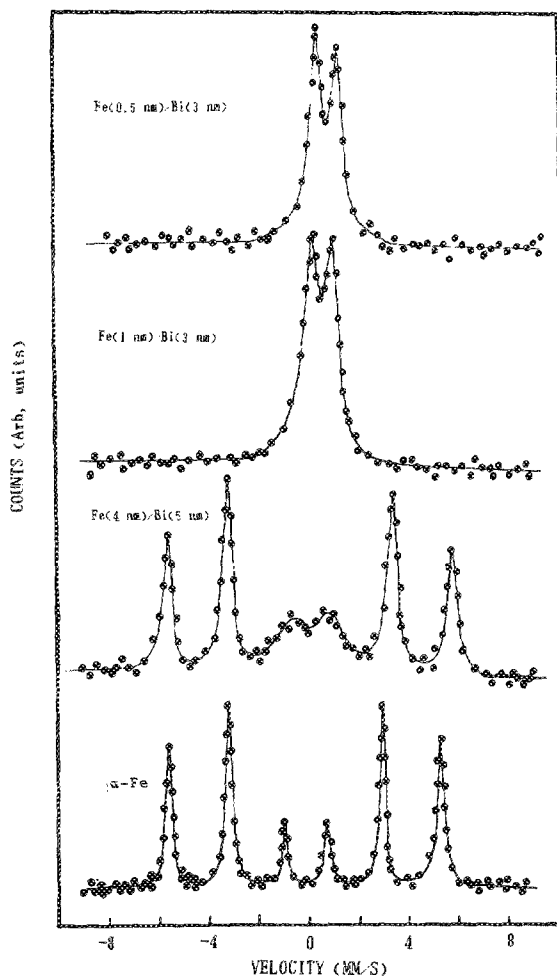


FIG. 4. Mössbauer spectra of several Fe-Bi multilayers and α -Fe.

linewidths of the middle two peaks are much larger than that obtained for standard α -Fe. This is probably due to a lower intensity doublet near zero velocity which shows a small part of Fe in a nonmagnetic environment. No remarkable information of interfacial diffusion in this sample can be obtained. The relative intensities of six lines are about 3:4:1:1:4:3, indicating that the direction of the magnetization is in parallel to the film plane. These results agree well with the above measurement of VSM, XRD and TEM. The spectra with a central doublet for t_{Fe} of both 0.5 and 1 nm are entirely nonmagnetic at room temperature. In the case of t_{Fe} of 0.5 nm, Mössbauer spectrum also confirm the VSM findings, i.e., the Fe layers in between Bi layers have no local magnetic moment. However, in the case of a sample with t_{Fe} of 1 nm, the Mössbauer spectrum indicates nonmagnetic, while VSM gave $4\pi M_s$ approaching to 1.2 T. This comparison strongly implies that at room temperature the magnetic state is that of superparamagnetism.

The temperature dependence of magnetization was studied in a temperature range of 77–600 K. Figure 5 shows a representative case. It is the temperature dependence magnetization in the films with t_{Fe} of 2 nm. Every

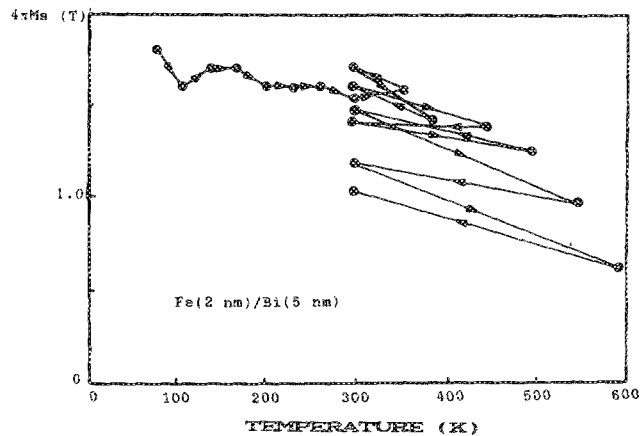


FIG. 5. Temperature dependence of magnetization in Fe-Bi multilayer with $t_{Fe} = 2$ nm and $t_{Bi} = 5$ nm.

datum in the figure was obtained after keeping sample at the given temperature of 0.5 h. The arrows indicate the order of the magnetizations versus temperature. It is noted that on annealing at 350 and 400 K the sample showed a little increase of magnetic moment. This probably is due to interface sharpening through a “uphill” diffusion⁵ of both Fe and Bi. Magnetization after heat treatment at temperature higher than 550 K is steeply decreased. This is understandable if one considers the low melting point of Bi (547 K).

IV. CONCLUSION

Magnetic Fe-Bi multilayers have been studied for the first time. Structural measurements indicate that artificially periodic structure can be made for films of t_{Fe} down to 0.5 nm by electron-beam evaporation at substrate temperature of 140 K. The structure of Fe layers transforms from bcc into amorphous below t_{Fe} of about 1.5 nm. At room temperature, saturation magnetization of the multilayers per unit volume Fe does not exceed that of pure α -Fe and decreases as t_{Fe} does. A perpendicular interface anisotropy is present for the films with t_{Fe} of about 1 nm, while for thicker t_{Fe} , the preferred magnetization direction is in the film plane.

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