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Magnetic properties of fcc iron in Fe/fcc metal multilayers

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Abstract

Vapor-deposition technique was employed to grow the fcc iron in the Fe/Cu, Fe/Pd and Fe/Pt multilayers. The thickness, periodicity, chemical composition, microstructure, and magnetic properties of the films were characterized and measured by various methods. The experimental results indicated that, when the Fe layers were thinner than 2–3 nm, the Fe atoms could grow in the fcc structure on the polycrystalline fcc non-magnetic metal layers with a fixed thickness of 6.5–7.5 nm. The fcc Fe in the Fe/fcc metal multilayers exhibited ferromagnetic behavior, and its magnetic moment can be as high as 3.4 μ_B for Fe(1.2 nm)/Cu(7.5 nm), 3.2 μ_B for Fe(1.6 nm)/Pd(6.5 nm), and 2.1 μ_B for Fe(2.3 nm)/Pt(7.0 nm), respectively. The modification of magnetic properties of fcc Fe was attributed to a significant change in the distance among Fe atoms in the fcc lattice, resulting in a considerable changing in electron-couple, compared with that in the normal Fe-bcc structure.

Keywords: Magnetic properties; Fcc iron; Multilayers

1. Introduction

In the last 10 years, magnetic multilayered films on a nanometre scale with artificial periodicity have attracted much attention because these films may feature some anomalous magnetic properties, such as changes in magnetization as the magnetic layer thickness is reduced, appearance in some cases of a uniaxial interfacial anisotropy, and giant magnetoresistance. These phenomena are probably related to the existence of surface and interface state, i.e. the reduced coordination number and symmetry of atoms in the surface, transitional structure sublayer, interface roughness, and associated chemical disordering, etc. [1–4]. In Fe/fcc metal multilayers, the metastable fcc Fe was obtained in some systems by various deposition methods, and these films exhibited very different magnetic properties. For example, recently, Himpsel [5] and Macedo et al. [6] reported that fcc Fe grew epitaxially on Cu single crystals and formed a sharp interface. In our recent study [3], it was also found that Fe grew epitaxially on polycrystalline Cu by electron-beam vapor deposition. In Fe/Pt system, fcc Fe was obtained by Croft et al. [7] and the author's group [8]. In Fe/Pd multilayers, the metastable fcc Fe phase was also obtained and the fcc Fe exhibited ferromagnetic behavior [9]. With extensive data obtained for many systems, research interest is therefore to study the magnetic properties of fcc Fe in Fe/fcc metal multilayers, which is helpful for understanding the origin of the magnetic property of magnetic materials. We report, in this paper, the magnetic properties of fcc Fe observed in the Fe/fcc metal multilayers prepared by electron-beam vapor deposition, the correlation between the magnetic properties and the microstructure of the films, and discuss the possible mechanism responsible for the observed magnetic properties.

2. Experimental procedure

The Fe/Cu, Fe/Pd and Fe/Pt multilayered films were prepared by depositing alternately the pure constituent metal (99.99%) at rates of 0.01–0.2 nm/s

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onto a glass substrate of 0.1 mm thickness (for magnetic property study) and a NaCl single crystal with freshly cleaved surface (for microstructure analysis) in an e-gun evaporation system at a vacuum level of 5×10^{-5}–2×10^{-6} Pa. The thickness of the constituent metal, varied from 1.2 to 14 nm controlled by an in situ quartz oscillator. The total thickness of the films was controlled in the range 75–275 nm. Samples were analyzed by Transmission Electron Microscopy (TEM), Selected Area Diffraction (SAD), and X-ray diffraction to identify the structure. Rutherford Backscattering (RBS) was also employed to measure the thickness, periodicity, and chemical composition of the samples. The magnetic properties were measured with a Vibrating-sample magnetometer (VSM), with a resolution of 5×10^{-6} emu, in a magnetic field of up to 10 kOe at room temperature. The size of the VSM samples were 4 mm × 6 mm or 5 mm × 5 mm. First, a hysteresis loop of the substrate and holder was measured and the saturation magnetization (M_S) was found to be approx. 4×10^{-4} emu, which was one or two orders of magnitude lower than that of the Fe/fcc metal multilayer films. Then the hysteresis loops of the samples were measured, and the magnetization of the substrate and holder was subtracted automatically by the computer. To reduce the experimental error, measurements were made on an assembly of four similar specimens. Consequently, the magnetic moment from the substrate and holder had a negligible effect on the measured values, and the precision of the measured magnetic moment of the films was estimated to be better than 1%. After measuring the magnetic properties, the films were dissolved in 5 ml aqua regia (HNO_3:HCl = 1:3) and the Inductive Coupled Plasma Atomic Emission Spectrum (ICP) was employed to determine the Fe content in the multilayers. An average magnetic moment per Fe atom was then obtained using these data. The error involved in the ICP measurement was approx. 5%, and therefore the total error was around 6%.

3. Results and discussion

3.1. Structure characterization

Results of low-angle X-ray diffraction and RBS confirmed the artificial periodicity of all the multilayered films, and the periodicities obtained, respectively from low-angle X-ray diffraction, agreed well with the RBS. For example, Fig. 1 shows a low-angle X-ray diffraction pattern for an [Fe(3 nm)/Pd(6.5 nm)]14 multilayers taken with Cu Kα radiation. From this figure, the second-, third-, fourth- and fifth order diffraction peaks of the Fe/Pd bilayers can be observed. According to these data, the periodicity of the multilayers is approx. 9.8 nm, confirming the

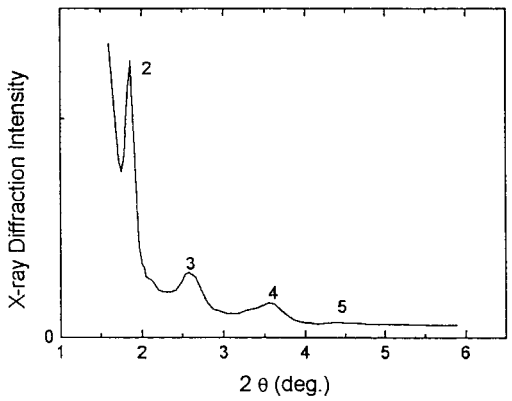

Fig. 1. Low-angle X-ray diffraction pattern of an [Fe(3 nm)/Pd(6.5 nm)]$_{14}$ multilayers.

deposited metal thickness. Fig. 2 shows a RBS spectrum of the [Fe(8.0 nm)/Pt(7.0 nm)]10 multilayers. This spectrum was obtained with 2.023 MeV He^+ ions, and the laboratory backscattering angle was 165°. In order to resolve individual layers by the detector at this energy, which presented a resolution of approx. 10 nm, the sample was tilted by 60°. From this figure, one can see that both iron and platinum spectra consist of ten peaks, corresponding to ten Fe/Pt bilayers in the multilayers. The total thickness of the sample is estimated to be approx. 150 nm by resolving the RBS spectrum, which agrees with the nominal thickness.

The microstructure of the films was examined by means of transmission electron microscopy (TEM), selected area electron diffraction (SAD), and X-ray diffraction. Table 1 shows the crystal structure of the constituent metals in the multilayers. From this table, one can see that for the Fe/Cu multilayers, when $t_{Fe} \leq 1.5$ nm and $t_{Cu} = 7.5$ nm, the metastable fcc Fe was obtained, which grew epitaxially on polycrystalline fcc Cu, as the difference of the atom radius between iron and copper is only approx. 3% [3]. The lattice parameter of fcc Fe was approx. 0.360 ± 0.005

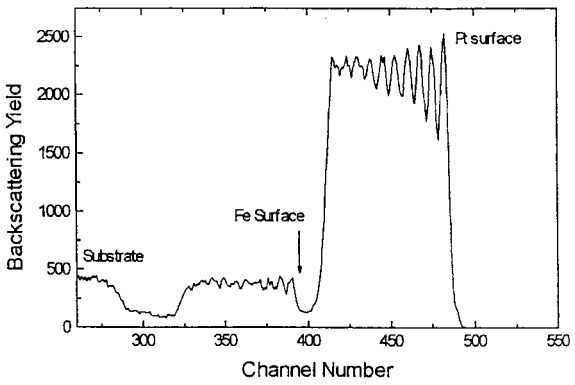

Fig. 2. The RBS spectrum of the [Fe(8.0 nm)/Pt(7.0 nm)]$_{10}$ multilayers.

Table 1
The crystal structure of the constituent metals in the multilayers

Specimen	Crystal structure	Lattice parameter of fcc Fe (nm)	Growth model
Fe/Cu (d_{Cu} = 7.5 nm, $d_{Fe} \leq 1.5$ nm)	fcc Fe + fcc Cu	0.360 ± 0.05	Epitaxial growth
Fe/Cu (d_{Cu} = 7.5 nm, $d_{Fe} > 2.5$ nm)	bcc Fe + fcc Cu	–	
Fe/Pd (d_{Pd} = 6.5 nm, $d_{Fe} \leq 6.5$ nm)	fcc Fe + fcc Pd	0.360 ± 0.05	Metastable phase
Fe/Pd (d_{Pd} = 6.5 nm, $d_{Fe} > 7.0$ nm)	bcc Fe + fcc Pd	–	
Fe/Pt (d_{Cu} = 7.0 nm, $d_{Fe} \leq 3.4$ nm)	fcc Fe + fcc Pt	0.389 ± 0.02	Epitaxial growth
Fe/Pt (d_{Cu} = 7.0 nm, $d_{Fe} > 5.6$ nm)	bcc Fe + fcc Pt	–	

nm. While $t_{Fe} > 2.5$ nm, the films consist of polycrystalline bcc Fe and fcc Cu, and the grain size of the films is approx. 1–5 nm.

For Fe/Pd multilayers, the experimental condition to obtain the metastable fcc Fe phase is $t_{Fe} \leq 6.5$ nm and $t_{Pd} = 6.5$ nm. Under such condition, the SAD patterns of the films consist of two sets of sharp diffraction rings from two fcc phases, i.e one is the fcc Pd phase and the other is a metastable Fe phase also with an fcc structure [9]. Their lattice parameters were approx. 0.389 ± 0.005 nm and 0.360 ± 0.005 nm, respectively. While $t_{Fe} > 7.0$ nm, the diffraction lines from metastable fcc Fe disappeared, and the films consist of bcc Fe and fcc Pd phases. The lattice parameter of the bcc phase was approx. 0.286 ± 0.005 nm, which was the same as that of the bulk bcc Fe.

For Fe/Pt multilayers, the metastable fcc Fe was also obtained when $t_{Fe} \leq 3.4$ nm and $t_{Pt} = 7.0$ nm [8]. However, the fcc Fe in Fe/Pt films was different from that in the Fe/Cu and Fe/Pt pairs. The lattice parameters of the new metastable fcc Fe phase in the Fe/Pt system was approx. 0.389 ± 0.002 nm, which is greater than that observed in Fe/Cu and Fe/Pd films. From the SAD patterns of Fe/Pt films, it was found that when $t_{Fe} \leq 3.4$ nm, there was only one fcc phase. According to the RBS and low-angle X-ray diffraction results, the films had a good periodic structure, and the lattice parameter of the observed fcc phase was about the same as that of pure platinum, i.e. 0.392 nm. It could therefore be thought that, under our experimental conditions, the growth of Fe atoms in an fcc structure on thick Pt layers is responsible for observing only one fcc structure. While $t_{Fe} >$ 5.6 nm, the Fe can not grow in an fcc structure on the Pt layer because of the internal stress caused by the large mismatch between Fe and Pt (approx. 8%), and the films consist of bcc Fe and fcc Pt.

Table 2
The magnetic properties of the fcc Fe phase in various systems

Specimen	Magnetic moment (μ_B)	Hc(Oe)
Fe(1.5 nm)/Cu(7.5 nm)	3.44 ± 0.28	19
Fe(1.2 nm)/Pd(6.5 nm)	2.77 ± 0.17	21
Fe(1.6 nm)/Pd(6.5 nm)	3.27 ± 0.20	18
Fe(3.0 nm)/Pd(6.5 nm)	3.04 ± 0.19	19
Fe(4.3 nm)/Pd(6.5 nm)	2.77 ± 0.17	20
Fe(6.5 nm)/Pd(6.5 nm)	2.85 ± 0.18	18
Fe(1.2 nm)/Pt(7.0 nm)	1.85 ± 0.12	14
Fe(2.3 nm)/Pt(7.0 nm)	2.12 ± 0.13	12
Fe(3.4 nm)/Pt(7.0 nm)	2.08 ± 0.13	9

3.2. Magnetic properties

The VSM results indicated that all the fcc phases in Fe/Cu, Fe/Pd and Fe/Pt multilayers exhibit ferromagnetic behavior and show an in-plane easy axis of magnetization. For example, Fig. 3 shows three typical hysteresis loops of Fe(1.5 nm)/Cu(7.5 nm), Fe(1.6 nm)/Pd(6.5 nm) and Fe(3.4 nm)/Pt(7.0 nm) in a magnetic field of 5 kOe, respectively. Table 2 shows the magnetic properties of the fcc Fe phase in various systems. From the table, one can see that with the exception of the Fe/Pt multilayers, the magnetic moment per Fe atom in the Fe/Cu and Fe/Pd films was obviously higher than that of the bulk bcc Fe (2.15 μ_B), and that the enhancement of the magnetic moment increased with decreasing Fe layer thickness, reaching a maximum value of 3.27 μ_B for Fe(1.6 nm)/Pd(6.5 nm) films and 3.44 μ_B for Fe(1.5

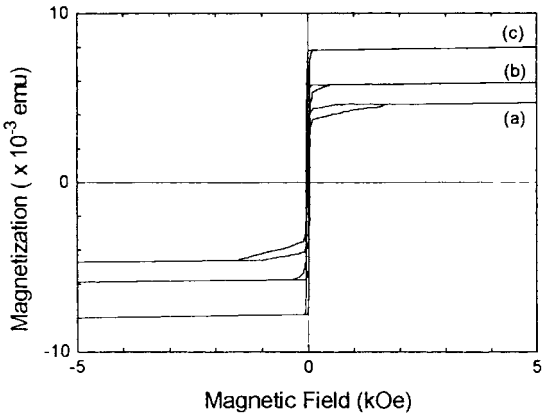

Fig. 3. Three typical hysteresis loops of (a) [Fe(1.5 nm)/Cu(7.5 nm)]$_{11}$; (b) [Fe(1.6 nm)/Pd(6.5 nm)]$_{15}$; and (c) [Fe(3.4 nm)/Pt(7.0 nm)]$_{14}$ multilayers in a magnetic field of 5 kOe, respectively. To reduce the measuring error, four identical specimens were put together in one measurement to obtain the hysteresis loops.

nm)/Cu(7.5 nm) films, respectively, i.e. approx. 1.5 times that of the bulk Fe.

Based on an all-electron total energy local spin density approach, Freeman et al. [10–12] predicted that there would be a significant enhancement in two-dimensional magnetism at the surfaces and interfaces in the transition metals grown on noble metals, e.g. Fe in a thin-film form with fcc structure can exhibit ferromagnetic behavior, in contrast to its bulk fcc phase, which is non-magnetic. The magnetic moment of the Fe atom, in comparison to its value of 2.15 μ_B in the bcc bulk, could be up to 2.98 μ_B for the topmost Fe overlayer and the clean Fe(001) surface. The first-principles all-electron linearized augmented plane wave method investigations in Fe/Cu(001) superlattices by Zhou et al. [13], also predicted that the magnetic moment of the interface Fe layer is stabilized at the high spin state of the fcc crystals. Tight-binding calculations of the magnetic surface, interface and multilayers by Krompiewski et al. [2,14,15] gave a similar prediction. These calculations can therefore explain the observed magnetic properties of fcc Fe, i.e exhibiting ferromagnetic behavior.

From the experimental results, it can also be found, even though Fe can grow with an fcc structure in various systems, the magnetic properties of fcc Fe changes with the lattice parameter of the fcc Fe phase, which depends on the other metals in the multilayers. The maximum magnetic moment per Fe atom with fcc structure was 3.44 μ_B for Fe(1.5 nm)/Cu(7.5 nm) films, 3.27 μ_B for Fe(1.6 nm)/Pd(6.5 nm) films and 2.12 μ_B for Fe(2.3 nm)/Pt(7.0 nm) films. The magnetic moment of fcc Fe in the Fe/Cu and Fe/Pd multilayers is significantly higher than that in the Fe/Pt multilayers. The main reason for the observed difference in magnetism of fcc Fe phases in the Fe/Cu, Fe/Pd and Fe/Pt multilayers may be explained in the following way. For the Fe/Cu system, the interface between the magnetic layer and non-magnetic layer could be considered to be an ideal situation which was used in theoretical investigation, as evidenced by the epitaxial growth of the metastable fcc Fe on a thick Cu layer, and the lattice parameter of fcc Fe was around 0.360 nm. Consequently, the Fe thin film with fcc structure in the Fe/Cu system have a higher magnetic moment than the bcc Fe.

As for the Fe/Pd multilayers, even though an fcc Fe with a lattice parameter of 0.360 ± 0.005 nm was also formed at the Fe/Pd interface, Fe could not grow epitaxially on Pd because of the radius difference. An ideal interface situation used in theoretical investigation could not be formed, because Fe and Pd have a large solid solubility, which resulted in a slightly lower magnetic moment of fcc Fe than that in the Fe/Cu films. It is generally assumed that a partial enhancement of the magnetic moment in the Pd/magnetic metal(Fe, Co) multilayers is due to the polarization of the Pd atoms [16–18], because Pd is a strong paramagnetic and a small addition of magnetic elements will induce a magnetic moment at the Pd sites (approx. 0.36 μ_B) [19]. Therefore the magnetic moment of the Pd atom was probably partly responsible, besides the enhanced magnetic moment of the fcc Fe phase, for the observed magnetic enhancement in the Fe/Pd multilayers.

For Fe/Pt multilayers, the fcc Fe had a lattice parameter of 0.389 ± 0.002 nm, which was larger than that in the Fe/Cu and Fe/Pd films. Moruzzi et al. [20] calculated the total energy and magnetization of the bulk fcc iron, and pointed out that the magnetism is related to the Wigner–Seitz radius, which contained the same volume as that of an atom in the actual lattice. The Wigner–Seitz cell volume of the fcc Fe in the Fe/Pt films is 20–30% greater than that in the Fe/Cu and Fe/Pd films, and this results in a smaller magnetic moment per Fe atom in the Fe/Pt films. Besides, the atomic exchange force decreases with increasing atomic distance, hence, a large lattice parameter will also result in a reduction of electron spin density and lower the magnetic moment. This may give a possible explanation for the observed magnetic properties of fcc Fe in the Fe/Pt multilayers.

4. Conclusion

In summary, we have shown that fcc iron in the Fe/Cu, Fe/Pd and Fe/Pt multilayers exhibits ferromagnetic behavior, and its magnetic moment was enhanced considerably in the Fe/Cu and Fe/Pd multilayers because of the epitaxial growth of the thin Fe layer on Cu and an fcc metastable phase ($a = 0.360 \pm 0.005$ nm) on Pd. The maximum magnetic moments per Fe atom in an fcc structure in the Fe/Cu Fe/Pd and Fe/Pt films were 3.44, 3.27 and 2.12 μ_B respectively, which is probably correlated with the lattice parameter of metastable fcc phase iron, i.e. a large lattice parameter results in a reduction of the magnetic moments in thin films.

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