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Structural and magnetic properties of iron-nitride thin films deposited using a filtered cathodic vacuum arc

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Abstract

A filtered cathodic vacuum arc technique has been applied to synthesis iron-nitride thin films on silicon (100) substrate. Two approaches were carried out to introduce the nitrogen gas into the chamber to control the nitrogen content. Films with smooth surface containing α-Fe(N), α′-Fe(N), α″-$Fe_{16}N_2$, γ′-Fe_4N, ζ-Fe_2N, ε-Fe_3N and γ″-FeN phases have been obtained, respectively. The magnetic properties of the films show that small amount of nitrogen addition into the iron films increases the saturation magnetization, while excess nitrogen decreases the saturation magnetization. It is explained based on the bond-band-barrier correlation mechanism [C.Q. Sun, Prog. Mater. Sci. 48 (2003) 521] that the electronegative nitrogen changes the valence states of iron into Fe^+ or Fe dipoles with higher magnetic momentum compared with an neutral Fe atom in the bulk.

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1. Introduction

Iron–nitrogen systems of interstitial solutions (α, γ, ε), chemical compounds (γ′-Fe_4N, ζ-Fe_2N) and metastable phases (α′,α″-$Fe_{16}N_2$) show various magnetic properties [1,2]. The nitrogen-poor phases (for example, α′ and α″) are ferromagnetic stoichiometric compounds. In particular, the α″-$Fe_{16}N_2$ phase possesses giant magnetic moment [3] and hence has attracted particular interest because of the potential applications in high-density data storage. Despite the poor or even non-ferromagnetic properties, the nitrogen-rich phases have been widely studied for surface hardness, wear and corrosion resistance [4–6]. Various deposition techniques have been used to deposit Fe–N films, such as molecular beam epitaxy [7], multi-shot implantation [8], facing target sputtering [9–11] and reactive radio frequency sputtering [12]. However, these techniques require complex processes and a relatively high deposition temperature. Seldom research has been reported on the filtered cathodic vacuum arc (FCVA) technique. The FCVA technique has many advantages and it has been applied to produce quality carbon coatings as well as metal oxides and nitrides films [13,14]. The main advantage of the FCVA technique is to employ a curved magnetic field to guide the plasma generated from the cathodic vacuum arc so as to deposit film on an out-of-sight substrate. Most of the unwanted macroparticles and neutral atoms can be removed through an electromagnetic filter [13]. In this study, we deposited Fe–N films by varying the partial pressure of nitrogen in the FCVA chamber at room temperature. The nitridation effect on the microstructure, the composition and the magnetic properties of the films are systematically examined and discussed based on the recent bond-band-barrier correlation mechanism [15].

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2. Experimental details

The FCVA set-up is schematically shown in Fig. 1. The system consists of three main parts: a cathode arc source, a magnetic filtering toroidal duct and a deposition chamber. The system was pre-evacuated to 8×10^{-6} Torr by a rotary and cryo-pump system. A radial electric field was formed via the toroidal duct on the target surface. The plasma, which was steered by this field, passed through the magnetic toroidal duct with all the macro-particles and neutral atoms being filtered and reached the deposition chamber resulting in a high ionization deposition. During deposition, the substrate temperature increases from room temperature to 80–110 °C due to the bombardment of energetic ions. A cylindrical iron target (30 mm thick and 60 mm in diameter, 99.9% purity) was mounted on a water-cooled copper support as the cathode. A polished Si(100) was set on the substrate holder. The arc current was maintained at 120 A. Two approaches were used to introduce the nitrogen gas into the chamber, as shown in Fig. 1. The first approach was to introduce the nitrogen gas into the region on the top of the target, see Fig. 1(a). The arc was then ignited on the target surface in the nitrogen atmosphere. Four samples were prepared using this method, labeled as I-1, I-2, I-3 and I-4, which were deposited at various working pressure ranging from 0.7×10^{-4} to 2.0×10^{-4} Torr. The second approach was to introduce the nitrogen gas into a linear ion beam source (D.C. 2.5×20 cm) that was mounted in the deposition chamber as shown in Fig. 1(b). The working pressure ranged from 1.0×10^{-4} to 2.2×10^{-4} Torr by controlling the ratio of nitrogen to argon partial pressure (P_N/P_{Ar}). Seven samples were prepared using this approach. The detailed deposition conditions are listed in Table 1. The thickness of the films was about 80 nm.

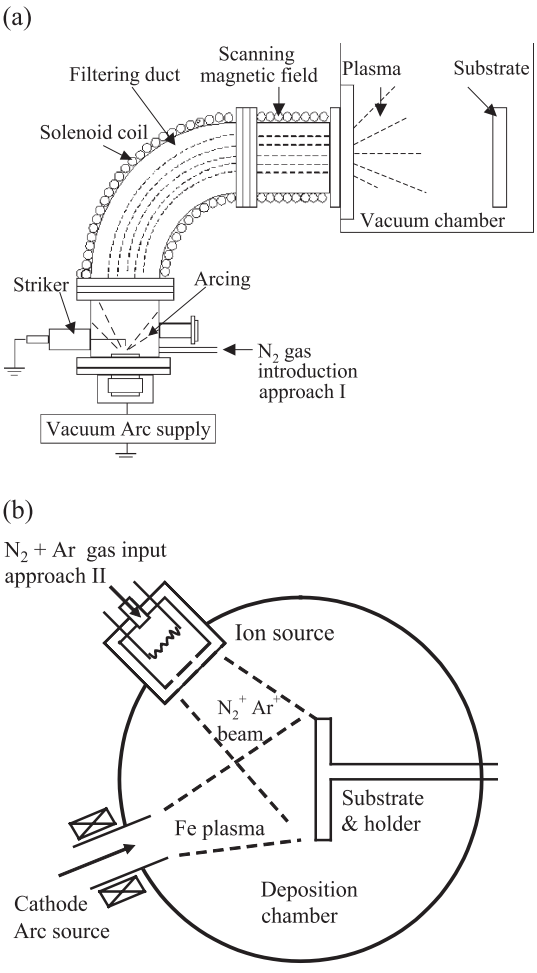

Fig. 1. Schematically illustration of the FCVA deposition system and the two approaches of nitrogen gas introduction (a) into the plasma region in the cathode source and (b) into a linear ion beam source that mounted on the deposition chamber mechanically.

Table 1
Deposition conditions

Gas input	Sample no.	Working pressure (10^{-4} Torr)	P_N/P_{Ar}
I	I-1	0.7	100:0
	I-2	1.0	100:0
	I-3	1.5	100:0
	I-4	2.0	100:0
II	II-1	1.0	10:7
	II-2	1.2	20:7
	II-3	1.4	30:7
	II-4	1.6	40:7
	II-5	1.8	50:7
	II-6	2.1	80:7
	II-7	2.3	100:7

The crystal structure of the films was characterized by Siemens D5005 X-ray diffraction (XRD) with a glancing incident mode (angle of 1°) and CuKα radiation (40 kV, 40 mA). Surface morphology and roughness of the films were characterized by atomic force microscopy (tapping mode of Dimension 3000 Scanning Probe Microscope, Digital Instruments). The binding energy and chemical composition were characterized using X-ray photoelectron spectroscopy (XPS) with a Kratos-Axis spectrometer with Dual-Anode Al Kα (1486.6 eV) X-ray radiation (15 kV and 10 mA) and hemispherical electron energy analyzer. To remove surface contamination from the films, Ar ion bombardment was carried out for 8 min using a differential pumping ion gun (Kratos MacroBeam) with an accelerating voltage of 4 keV and Ar gas pressure of 1×10^{-7} Torr. The magnetic properties were characterized using a vibrating sample magnetometer (VSM, Oxford).

3. Experimental results

3.1. Deposition rate and surface morphology

The deposition rates for the two approaches are quite different. Using approach I, the deposition rate decreases from 39 to 16 nm/min with the increase in P_N from 0.7×10^{-4} to 2.0×10^{-4} Torr. In approach II, however, the deposition rate is much lower, ranging from 6.4 to 0.35 nm/min, and it decreases with the decrease in P_N/P_{Ar} from