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MAGNETIC AND ELECTRIC TRANSPORT PROPERTIES OF Fe–Hf–O FILMS PREPARED BY SUPERSONIC PLASMA JET METHOD

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The influence of deposition conditions on the structure, magnetic properties, and electrical resistivity of Fe–Hf–O films, prepared by the supersonic plasma jet deposition technique, was investigated. Composition of the films was controlled by the nozzle composition and the working gas. It varied in the limits: 15–68 at.% of Fe, 0.5–8 at.% of Hf and 29–80 at.% of O. The films were mainly X-ray amorphous. Some of them showed a weak and broad peak near the [110] reflection of α-Fe indicating the presence of Fe-rich clusters in an amorphous matrix. Depending on the deposition parameters the magnetic properties vary from paramagnetic to ferromagnetic ones. The electrical resistivity changes from the metallic to the hopping type. In some samples a large negative magnetoresistance is observed.

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Nanogranular composites, consisting of ferromagnetic metal grains in an insulating matrix, are under the growing interest due to their prominent magnetic and magnetoresistant properties. The nanogranular Fe–Hf–O films, prepared by the reactive magnetron sputtering, exhibit good soft magnetic properties together with high resistivity in the Fe-rich region [1] and the giant magnetoresistance behavior in the Fe-poor region [2].

In this work the Fe–Hf–O films, prepared by the supersonic plasma jet method, were investigated. This method provides some advantages in comparison with the conventional reactive magnetron sputtering [3]. A simple scheme of the reactive plasma jet reactor is shown in Fig. 1. The material of the source is sputtered by the ions produced in the RF hollow cathode discharge. One RF electrode is formed by the nozzle connected to RF generator (27.12 MHz, 500 W). The hollow cathode discharge takes place at the nozzle mouth in the working gas (the mixture of Ar and H_2) coming through the nozzle into the reactor chamber. The grounded wall of the chamber and the substrate holder are the second electrode. The atoms and ions sputtered from the inner wall of the nozzle are drifted towards the substrate by the working gas leaving the nozzle with supersonic velocity. The supersonic plasma forms a well defined channel with high chemical reactivity. The hydrogen is added to the working gas in order to control the contamination of films by the residual oxygen in the reactor. The pressure inside the reactor chamber is hold constant in the range of 0.1–0.3 Torr. The composition of working gas is controlled by the flow rates of the gases. The substrate holder is cooled by water to avoid the bulk crystallization of the film during the deposition process.

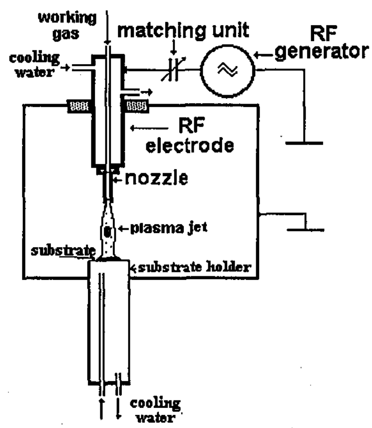

Fig. 1. RF plasma-chemical reactor with supersonic nozzle.

The composite nozzle is used to sputter Fe and Hf simultaneously. It consists of Fe and Hf rings stowed in a stainless steel or iron (99.9%) tube. To vary the composition of films the Fe/Hf filling ratio and the working gas are changed. The deposition parameters, the composition of the films, determined by the electron probe microanalysis, and the magnetic induction, evaluated from ferromagnetic resonance (FMR) measurements, are shown in Table.

The samples prepared with pulse regime of RF generator (1, 2, and 3) are fully X-ray amorphous. The other samples exhibit a weak broad peak near the [110] reflection of α-Fe superimposed on the amorphous background, which indicates the presence of small Fe-rich clusters embedded in an amorphous matrix.

The magnetic properties of the as-deposited films were investigated by ferromagnetic resonance at 35.7 GHz with magnetic field applied parallel and perpendicular to the film plane. Some typical FMR spectra are shown in Fig. 2. The films 1, 3, 5, and 8, prepared with low RF power or without H_2 in the working gas, are non-magnetic. The other samples show a variety of magnetic properties between the superparamagnetic behavior, characterized by broad resonance peaks close to the paramagnetic resonance field (sample 9 in Fig. 2), and the behavior typical of the presence of some ferromagnetic phase (sample 13 in Fig. 2). The ferromagnetic behavior is usually observed in the films deposited with the purer Ar.

The temperature dependence of electrical resistivity was measured on selected samples by the four-terminal method. Different character of conductivity was observed in different samples. The behavior varied from the one typical of amorphous metals (curve B in Fig. 3) to the non-metallic behavior with large and

TABLE

Deposition parameters, composition and magnetic induction (B) of Fe–Hf–O films.

Sample #	Nozzle Fe:Hf ratio	RF power [W]	Ar flow rate [sccm]	Ar purity %	H_2 flow rate [sccm]	Pressure [Torr]	Composition at.%			B [T]
							Fe	Hf	O	
1[a]	3:1	30[c]	400	99.99	70.4	0.23	15	5	80	0
2	7.7:1	30[c]	400	99.99	70.4	0.25	35	1	64	0.30
3	7.7:1	80[c]	400	99.99	70.4	0.30	35.5	2.5	62	0
4	7.7:1	100	400	99.99	70.4	0.25	37	2	61	0.12
5	7.7:1	50	400	99.99	70.4	0.25	37.5	0.5	62	0
6	7.7:1	150	400	99.99	70.4	0.25	53	3	44	0.29
7	7.7:1	150	400	99.99	35.2	0.24	53	2	45	0.64
8	7.7:1	150	400	99.99	0	0.23	55	3	42	0
9	5:1	150	400	99.99	70.4	0.25	36.5	5.5	58	0.17
10	5:1	150	400	99.99	35.2	0.25	39	5	56	0.17
11	5:1	150	600	99.99	70.4	0.33	35	6	59	0.08
12[b]	5:1	150	400	99.99	70.4	0.25	33	2	65	0.31
13[b]	5:1	150	400	99.999	70.4	0.26	68	3	29	1.07
14[b]	5:2	150	400	99.999	70.4	0.24	46	8	46	
15[b]	5:2	150	400	99.999	70.4	0.24	49	7	44	

[a]substrate not cooled, [b]nozzle in Fe tube, [c]pulse

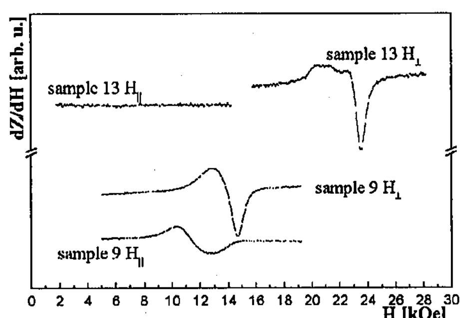

Fig. 2

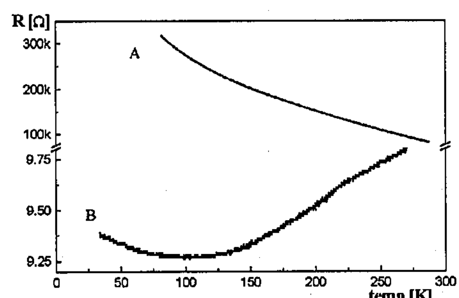

Fig. 3

Fig. 2. Typical resonance curves measured at microwave frequency of 35.7 GHz with magnetic field parallel ($H_{\|}$) and perpendicular ($H_{\perp}$) to the film plane.

Fig. 3. Temperature dependencies of electrical resistance for the typical metallic (B) and non-metallic (A) Fe–Hf–O films. A — sample 9, B — sample 7.

temperature dependent resistance (curve A). The metallic type of conductivity is usually observed in the films with low oxygen content. The temperature dependence of resistivity in the non-metallic films can be well fitted by the $\ln\rho \sim T^{-1/2}$ dependence (below 100 K), typical of the hopping type of conductivity in granular composite materials [4].

On selected samples the magnetoresistance at room temperature and 77 K was measured with magnetic field up to 6 kOe applied in the film plane. The largest magnetoresistance was found in the samples exhibiting the non-metallic conductivity. The typical giant magnetoresistance (GMR) effect, observed for sample 9, is shown in Fig. 4. The relative change of resistivity ($\Delta\rho/\rho = 2.1\%$ at 77 K) is comparable with the value of 4.6% reported for the sputter-deposited film $Fe_{44}Hf_{20}O_{36}$ [5]. As can be seen, the effect substantially decreases for room temperature in contradiction to the results of Hayakawa et al. [2]. In our case, however, the sample is not fully saturated in the field range used, which may be explained by smaller magnetic moments of the superparamagnetic clusters.

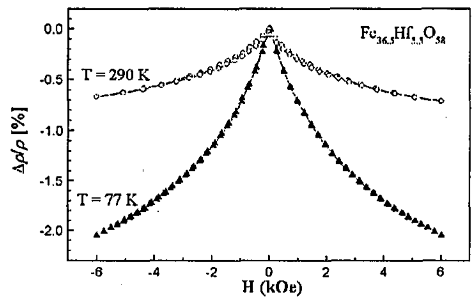

Fig. 4. Magnetoresistance curves of the sample 9 measured at 290 K and 77 K in magnetic field parallel to the film plane.

In conclusion, it has been shown that the supersonic plasma jet method can be used to deposit composite nanogranular magnetic films. The magnetic and conductivity properties can be controlled by the composition of the nozzle and the deposition parameters. The Fe–Hf–O films with high oxygen content exhibit the non-metallic character of conductivity and show a large magnetoresistance effect.

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