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Soft magnetic properties, high frequency characteristics, and thermal stability of co-sputtered FeCoTiN films

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ABSTRACT

FeCoTiN films were prepared by reactive radio frequency magnetron co-sputtering technique in an argon and nitrogen mixture atmosphere. The soft magnetic properties, high frequency characteristics, and magnetic thermal stabilities of the FeCoTiN films were investigated. The FeCoTiN films deposited at an N_2/Ar ratio of 7% have good soft magnetic properties and high saturation magnetization ($\mu_0 M_s$) of about 1.9 T. The ferromagnetic resonance frequency reaches 2.2 GHz with the real part of relative permeability above 500 before rolloff for the as-deposited $(Fe_{65.4}Co_{34.6})_{94.7}Ti_{5.3}N$ films. The magnetic thermal stability of the films with higher Ti content is obviously improved. The $(Fe_{65.4}Co_{34.6})_{87.1}Ti_{12.9}N$ film even annealed at 450 °C exhibits good magnetic softness and dynamic characteristics. The ferromagnetic resonance frequency reaches 1.9 GHz with the real part of relative permeability larger than 500 before rolloff for the annealed films.

1. Introduction

Soft magnetic films with large permeability and high ferromagnetic resonance frequency have been studied intensively because of their potential applications in micromagnetic devices and electromagnetic wave absorption [1,2]. For these applications, magnetic films should possess high saturation magnetization ($\mu_0 M_s$), high anisotropy field (H_k), high resistivity (ρ), and low coercive field to increase work frequency into GHz range. The thermal stability of magnetic properties of the films is also important for the actual applications which should often go through a high temperature process over 400 °C [3]. The well-known FINEMET-type alloys have excellent soft magnetic properties but are not thermally stable above 250 °C. Though the NANOPERM and HITPERM alloys doped with Hf exhibit good soft magnetic properties up to 550 °C but are not suitable for high frequency applications in GHz because of the low resonance frequency [4,5]. FeXN (X = Ta, Hf, Ti, Al, etc.) films are considered to be promising materials for their excellent soft magnetic properties, high saturation magnetization, and better reliabilities than the FeN films [6–11]. However, the uniaxial anisotropy fields of these films are typically below 20 Oe in the as-deposited states and can be reduced in annealing at the temperature above 150 °C [12,13]. The recently developed FeCoN films have an in-plane uniaxial anisotropy field usually exceeding 30 Oe, good magnetic softness, and high $\mu_0 M_s$ above 2 T in the as-deposited states [14–16]. However, the magnetic thermal stability of the FeCoN films, especially in high frequency, has not been studied systematically so far. FeCoTaN and FeCoAlN films show good dynamic properties after annealing at 400 °C for 30 min, but their saturation magnetization and permeability are obviously low compared to FeCoN films [3]. Addition of proper elements into FeCoN alloys may improve their thermal stability with no obvious weakening of saturation magnetization and high frequency characteristics.

In our present work, we studied the effects of Ti addition into FeCoN alloy films on the soft magnetic properties, high frequency characteristics, and thermal stability of FeCoTiN films. The results show that the FeCoTiN films remain good soft magnetic properties and uniaxial anisotropy. The magnetic thermal stability of the FeCoTiN films is obviously improved with the Ti addition. The ferromagnetic resonance frequency can reach 1.9 GHz even when the films are annealed at 450 °C for 1 h. The FeCoTiN films may be a promising candidate for the high-frequency applications.

2. Experimental

A series of FeCoTiN films with thickness of about 100 nm were deposited on glass substrates by means of the reactive radio frequency magnetron co-sputtering technique in an Ar and N_2 mixture atmosphere. The samples for TEM analysis were grown onto copper grid-supported carbon films. An $Fe_{65}Co_{35}$ alloy target of 60 mm

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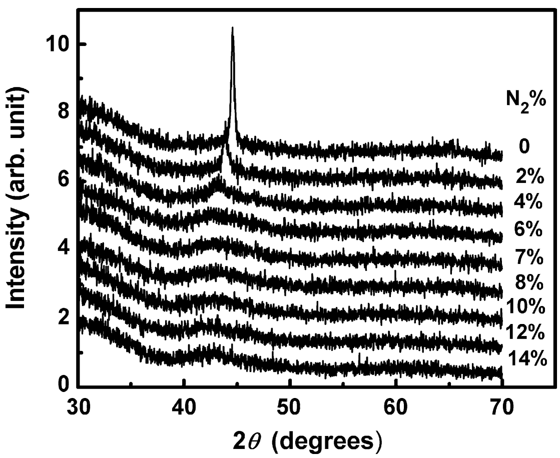

Fig. 1. The XRD patterns for the as-deposited $(Fe_{65.4}Co_{34.6})_{94.7}Ti_{5.3}N$ films deposited at different N_2/Ar gas flux percentages (N_2%).

in diameter and a Ti target of 50 mm in diameter were used as the sputtering targets. The sputtering power for the $Fe_{65}Co_{35}$ alloy target was kept at 90 W; and the sputtering power of the Ti target was adjusted from 0 to 83 W to control the Ti content in the films. The base pressure of the sputtering chamber was better than 7×10^{-5} Pa. The Ar gas flux was kept at 25 sccm during sputtering. The nitrogen content in the films was controlled by adjusting the N_2 gas flux. An external field of about 200 Oe was applied to the in-plane direction of the film to induce in-plane uniaxial anisotropy and define the easy axis.

The thermomagnetic annealing of the films were performed in a vacuum better than 5×10^{-4} Pa. A DC magnetic field of 150 Oe was applied parallel to the film plane during annealing. The direction of this field along the film plane is in the same direction as used during deposition.

The thicknesses of the films were measured with a Dektak-8 Stylus profilometer. The chemical composition of the Fe, Co, and Ti elements in the films was determined by an IRIS Advantage atomic emission spectrometer. The structure of the films was analyzed by X-ray diffraction (XRD) at room temperature on a Rigaku D/Max-2400 X-ray diffractometer with Cu Kα radiation and by transmission electron microscope (TEM) observations on a Philips EM-400T electron microscope using working voltage of 100 kV. The static magnetic properties of the films were measured by a Lake Shore 7304 vibrating sample magnetometer (VSM) at room temperature. The electrical resistivity was measured by a conventional four-point probe method at room temperature. The complex permeability spectra of the films were measured in 0.1–5 GHz frequency range using shorted microstrip transmission-line perturbation method [17,18] at room temperature with an Agilent PNA E8363B network analyzer.

3. Results and discussions

3.1. Structure and properties of as-deposited films

The FeCoTiN films with different Ti contents and N_2/Ar gas flux ratios (N_2%) were prepared and characterized. The atomic ratio of Fe:Co in all the films is about 65.4:34.6. The XRD patterns for the as-deposited $(Fe_{65.4}Co_{34.6})_{94.7}Ti_{5.3}N$ films with different N_2% are shown in Fig. 1. In the diffraction patterns of the films with N_2% from 0 to 4%, only the diffraction peak for the body-centered cubic α-Fe(CoTiN) (1 1 0) is present, implying that the (1 1 0) plane is preferred in the films. The (1 1 0) diffraction peak width becomes broader with increasing N_2%, indicating that the grain size in the films decreases with increasing N_2%. Meanwhile, the (1 1 0) diffraction peak clearly shifts to lower angles with increasing N_2%, indicating that the lattice expands with increasing N content. The 2θ value of the (1 1 0) diffraction peak of the $(Fe_{65.4}Co_{34.6})_{94.7}Ti_{5.3}$ film without N is 44.58°, which is slightly smaller than that of FeCo alloy. Hence the introduction of Ti also brings about the lattice expansion of the FeCo alloy. However, the diffraction peak almost disappears when N_2% is higher than 4%. This indicates that these films may become amorphous. The selected area electron diffraction (SAED) patterns in Fig. 2 confirm the formation of amorphous structure. These results show that the formation of amorphous structure is more favorable in the FeCoTiN films than the FeCoN films [14,19].

The variations of saturation magnetization, in-plane uniaxial anisotropy, and coercivity of the as-deposited $(Fe_{65.4}Co_{34.6})_{94.7}Ti_{5.3}N$ films with N_2% are shown in Fig. 3. With N_2% increasing from 0 to 14%, $\mu_0 M_s$ of the films decrease from 2.23 to 1.25 T. The films deposited with N_2% less than 4% have higher $\mu_0 M_s$ than 2.1 T but relatively high coercivity and no obvious in-plane magnetic anisotropy. With the increase of N_2%, both the hard axes coercivity (H_{ch}) and the easy axes coercivity (H_{ce}) of the films gradually decrease to a low value below 3 Oe. The hysteresis loops of the films with N_2% of 7% show the good magnetic softness and typical in-plane uniaxial anisotropy (Fig. 4). The decrease of $\mu_0 M_s$ and coercivity should be attributed to the variations of the composition and structure in the films. The amorphization and probable formation of the nitride clusters in the films with the increase of N_2% may be the main reason for the decreasing $\mu_0 M_s$. The magnetic exchange interaction in the amorphous or nanocrystalline structure contributes to the soft

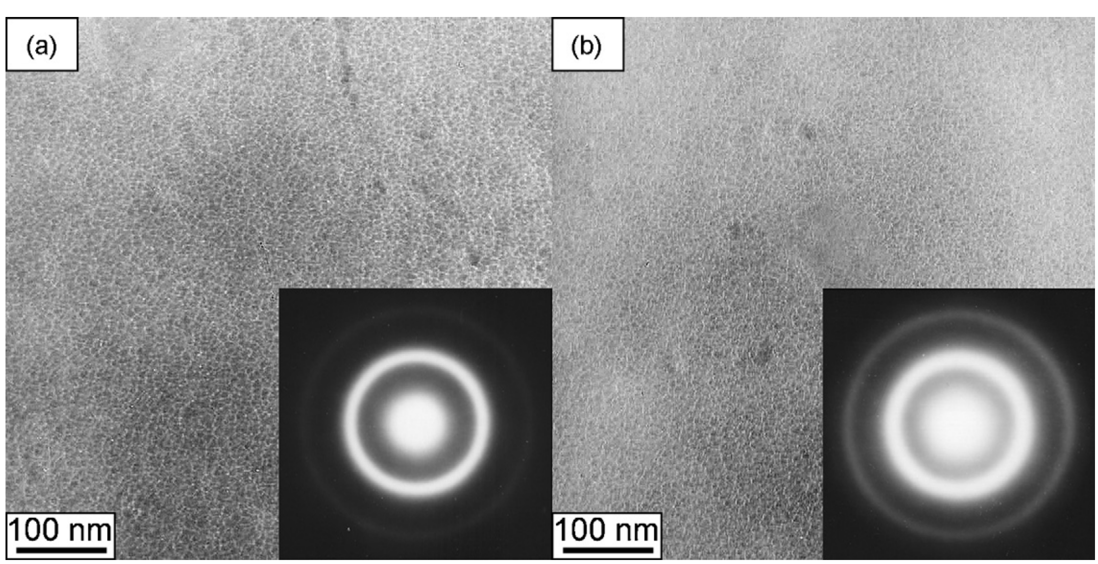

Fig. 2. The TEM bright field image and SAED pattern of the as-deposited films with 5.3% Ti (a) and 12.9% Ti (b).