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Soft magnetic property and high-frequency permeability of $[Fe_{80}Ni_{20}–O/TiO_2]_n$ multilayer thin films

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Multilayer thin films with excellent magnetic property and high-frequency permeability possess a significant position in electromagnetic miniaturization devices applied in GHz range. In this work, by controlling the alternately magnetron sputtering time at room temperature, we fabricated $[Fe_{80}Ni_{20}–O/TiO_2]_n$ multilayer thin films with different TiO_2 interlayer thicknesses (t = 0.25–4 nm) and fixed $Fe_{80}Ni_{20}–O$ layer thickness without applying inducing magnetic field. The static and dynamic magnetic properties of these films were investigated and evident in-plane uniaxial magnetic anisotropy field can be adjusted in a broad range only by changing the thickness of the TiO_2 interlayer. When the t increases gradually from 0.25 to 4 nm, the resistivity and in-plane uniaxial magnetic anisotropy field increase monotonically. In particular, by controlling the thickness of each TiO_2 interlayer to an optimized value, good soft magnetic property and high-frequency performances in GHz range have been observed.

1. Introduction

As one kind of significant material, high-frequency soft magnetic films are in great demand in various electromagnetic equipments, such as microwave noise filters, electromagnetic interference suppressor, and film inductors [1–3]. The films applied in gigahertz range should possess high saturation magnetization (M_s), high permeability (μ), large ferromagnetic resonance frequency (f_r) and high resistivity (ρ) simultaneously [4,5]. However, there exist two challenges when the films are applied in the high-frequency range. According to the Snoek's limit, it's difficult to obtain high μ and high f_r at the same time, meanwhile eddy-current loss cannot be ignored due to low ρ in most metal-based nanocrystalline magnetic films [6,7]. These are bottlenecks of the soft magnetic films for applications in high-frequency gigahertz range, therefore a large amount of studies are proceeded to solve this issue. Since the f_r is proportional to $(H_k \times 4\pi M_s)^{1/2}$, and the static permeability (μ_s) is expressed as $\mu_s = 1 + 4\pi M_s/H_k$, in which H_k is magnetic anisotropy field, the value of H_k should be adjusted in a large range, in order to achieve proper permeability and ferromagnetic resonance frequency synchronously [8].

In recent years, magnetic granular films, of which the microstructure consist of magnetic metal nanoparticles such as Fe, Co or iron-based alloy embedded into a nonmagnetic insulating matrix like Al_2O_3, HfO_2, SiO_2 and Si_3N_4 have been researched thoroughly [9–12]. The magnetic property and resistivity of these films can be adjusted by changing the volume fraction or the size of the magnetic metal grains, and the optimal integrated performance can be obtained at certain composition range. However, the strong demagnetization caused by separated metal nanoparticles has an adverse effect on soft magnetic property and consequently, good magnetic properties are only received in certain narrow volume fraction of magnetic particles in the whole granular film system [13]. As some recent papers mentioned, for monolayer nanocrystalline or granular soft magnetic thin films, preferable high-frequency magnetic properties are confined to the certain film thickness about a few hundred nanometer, since the microstructure and stress of film are strongly influenced with increasing thickness [14,15]. Moreover, in order to induce in-plane magnetic anisotropy as well as improve high-frequency magnetic performance, most reports chose to apply an external inducing magnetic field, either by placing one during deposition or by applying one during post-annealing. Therefore, it is urgent to find a feasible way to avoid the deteriorations of soft magnetic properties and a suitable way to adjust in-plane magnetic anisotropy of films. Laminating the films into magnetic multilayer thin films (e.g. introducing the nonmagnetic insulating phase as interlayer) is one promising way to overcome the defects we mentioned above. Not only can the insulating phases contribute to a higher resistivity and a less eddy current loss, but also they would not have extrinsic affect on the properties of each magnetic layer. Furthermore, it would help to achieve magnetic films with a large thickness for practical application. Besides, the interlayers can restrain the grain size of ferromagnetic layers at a certain level to protect the performance of each ferromagnetic layer from extrinsic influences

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[16,17]. Based on these advantages, the preparation of multilayer thin films is a beneficial attempt for high-frequency soft magnetic films.

In this work, we investigated the fabrication and high-frequency properties of $[Fe_{80}Ni_{20}–O/TiO_2]_n$ multilayer thin films, and chose $Fe_{80}Ni_{20}$ alloy as the magnetic phase and TiO_2 as nonmagnetic insulating phase of multilayer thin films after weighing many considerations. The ferromagnetic layers of multilayer thin films should have excellent soft magnetic performances including high saturation magnetization and low coercivity, therefore, it is necessary to use a suitable magnetic alloy as matrix of ferromagnetic layers. It is well known that the magnetic moments of Fe–Ni alloy obey Slater–Pauli curve [18], so the M_s of the Fe–Ni alloy with lower composition of Ni ($C_{Ni} \leqslant 20\%$) is slightly larger than that of Fe ($M_s = 2.15$ T), but the M_s decreases obviously with higher composition of Ni (e.g. permalloy and invar). Besides, Fe–Ni alloy with proper composition of Ni is beneficial for decreasing the magnetocrystalline anisotropy and improving the soft magnetic property. Therefore, after considering these factors, we used the $Fe_{80}Ni_{20}$ alloy as the matrix of ferromagnetic layers in multilayer thin films. Moreover, the multilayer films for high-frequency applications might be magnetic core in electromagnetic devices like film inductors operating in GHz range, so the multilayer films should have high resistivity and high permittivity to prevent electromagnetic interference and achieve electromagnetic matching. TiO_2 not only has high resistance but also has high relative permittivity ($\varepsilon \sim 110$), which is far more than those of other insulating oxide phases (e.g. Al_2O_3, ZnO, SiO_2) [19]. So the multilayer films with TiO_2 interlayer can adjust resistivity and permittivity in high-frequency simultaneously, and be beneficial for electromagnetic matching in electromagnetic devices operating in GHz range. Base on our previous research about $Fe_{80}Ni_{20}$–O films to achieve the in-plane uniaxial magnetic anisotropy (IPUMA) in a simple and effective way by adding a very low dose of oxygen [20], we demonstrate that the IPUMA of the $[Fe_{80}Ni_{20}–O/TiO_2]_n$ multilayer thin films is only related to the thickness (t) of nonmagnetic insulating TiO_2 interlayer. Therefore, adjustable ferromagnetic resonance frequency can be obtained in a broad range with high saturation magnetization, high resistivity and larger permeability simultaneously.

2. Experiments details

$[Fe_{80}Ni_{20}–O/TiO_2]_n$ multilayer thin films were deposited on (1 0 0)-oriented silicon substrates and glass slides by direct current (DC) and radio frequency (RF) magnetron sputtering at room temperature. The background pressure of the chamber was evacuated below 4×10^{-4} Pa. In the whole sputtering process, the flow rate of Ar gas was kept at 20 standard-state cubic centimeter per minute (SCCM), and the pressure of the chamber was maintained at 0.4 Pa during the deposition. The relative O_2 flow ratio, $R(O_2)$ = [O_2 flow rate]/[Ar flow rate + O_2 flow rate] (in%), was fixed in 1%. In order to obtain $[Fe_{80}Ni_{20}–O/TiO_2]_n$ multilayer thin films, we conducted pre-sputtering to remove the oxide on the surface of targets for a long time with a lower power, and then the depositions were conducted using a power of 75 W for $Fe_{80}Ni_{20}$ alloy target (purity 99.99%) and 100 W for Ti target (purity 99.99%) with a substrate rotation speed of 24 rpm. We sputtered ferromagnetic layer and nonmagnetic insulating interlayer alternately, and the thickness of each monolayer was controlled by adjusting sputtering time using the switched baffles. The total thickness of the multilayer thin films was controlled at about 120 nm, and the thickness of ferromagnetic $Fe_{80}Ni_{20}$–O single layer was well controlled in 5 nm and the thickness of nonmagnetic TiO_2 single layer was changed from 0.25 to 4 nm by adjusting the sputtering time. Our previous work had demonstrated this method could effectively obtain films with obvious multilayer structure [21], and, in this work, multilayer thin films with well defined layer stacking periodicity were studied by several experimental characterizations. The X-ray diffraction (XRD, PANalytical B.V.) was used to analyze the crystal structure and phases of the multilayer thin films. Scanning electron microscopy (SEM, LEO 1530FE) working at 20 kV accelerating voltage was performed to carry on the surface morphology investigation. The static magnetic properties were characterized by a vibrating sample magnetometer (VSM, Lake Shore 7404). The microwave permeability was measured at frequencies from 400 MHz to 5 GHz by using a vector network analyzer and with a shorted microstrip transmission-line perturbation method.

3. Results and discussion

The X-ray photoelectron spectroscopy (XPS) was used to analyze the phase type of the oxidized interlayer in multilayer film samples. The XPS spectrum of Ti 2p for the multilayer thin film with t = 3 nm (Fig. 1) shows two obvious peaks located at 458.4 eV and 464.4 eV, corresponding to Ti 2p peaks of TiO_2 [22]. This indicates that the interlayer of multilayer thin films was oxidized into TiO_2 insulating interlayer at the $R(O_2)$ = 1% during deposition.

As shown in the XRD patterns of the $[Fe_{80}Ni_{20}–O/TiO_2]_n$ multilayer thin films with t varying from 0.5 to 4 nm (Fig. 2), only (1 1 0) and (2 0 0) diffraction peaks of bcc $Fe_{80}Ni_{20}$ phase can be observed, whereas the diffraction peaks of the insulating TiO_2 phase cannot be observed for all films. In addition, we have fabricated a TiO_2 film over 30 nm thickness under the same sputtering condition, but there were no diffraction peaks of TiO_2 either. So, we infer that the TiO_2 interlayer exists as a form of amorphous morphology under this sputtering condition from 0.5 nm to 4 nm. Meanwhile, when the t gradually increases from 0.5 to 4 nm, the width of $Fe_{80}Ni_{20}$ diffraction peaks get wider and the peaks height get lower, which reflects a tendency of grain refinement in the ferromagnetic layer. According to Scherrer equation, the estimated grain sizes are 18.2, 14.8, 9.6, 7.6, 7.1, and 7.3 nm for t = 0.5, 1, 1.5, 2, 3, and 4 nm, respectively. Therefore, due to the gradually thickening of TiO_2 interlayer, the continuous growths of the $Fe_{80}Ni_{20}$–O layers are interrupted, and the $Fe_{80}Ni_{20}$–O grain size is noticeably reduced.

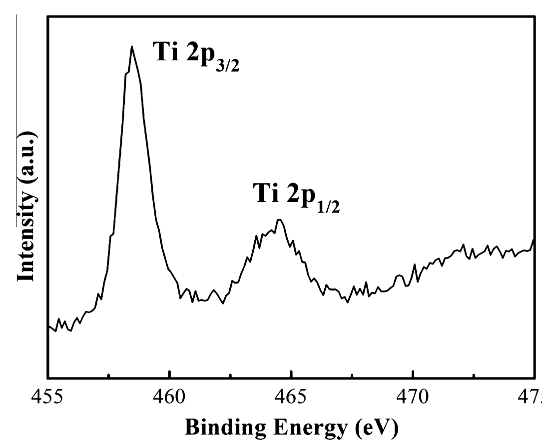

Fig. 1. XPS spectrum of Ti 2p for the multilayer thin film with t = 3 nm.

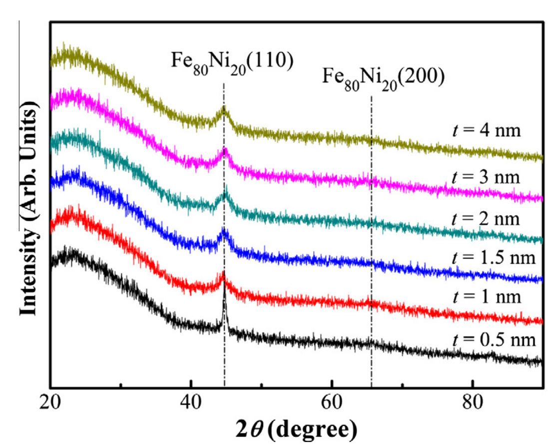

Fig. 2. XRD patterns of the $[Fe_{80}Ni_{20}–O/TiO_2]_n$ multilayer thin films with t varying from 0.5 to 4 nm.