Structural and magnetic properties of NiZn and Zn ferrite thin films obtained by laser ablation deposition

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Laser ablation deposition has been used to synthesize nanoscale ferrite structures. Our investigations were performed on NiZn and Zn ferrite films deposited on silicon(100) substrates. Films produced by laser ablation at room temperature were annealed at 550 °C for 1 h. Other films were deposited directly at a 550 °C substrate temperature without subsequent annealing. Complementary x-ray diffraction and superconducting quantum interference device magnetometry measurements helped identify the optimum laser ablation deposition conditions for obtaining the desired nanoferrite structures. From the hysteresis loops at 300 and 10 K we identified the paramagnetic or ferromagnetic behavior of the films. The zero field cooled–field cooled (ZFC–FC) magnetization, $M(T)$, curves yielded the value of the blocking temperature in both NiZn and Zn ferrite systems. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1854416]

I. INTRODUCTION

Spinel ferrites such as NiZn and Zn ferrites are commercially important materials because of their excellent electrical and soft magnetic properties.[1,2] They have been used for radio frequency coil, transformer cores, and rod antennas.[3] There is a particular application of NiZn ferrites requiring low porosity and controlled microstructure in the magnetic cores of read-write heads for high-speed digital tape or disk recording. For this purpose we have to understand the relationship between microstructure and magnetic properties in dense ferrite thin films.

In order to readily integrate ferrite materials into electronics devices, it is desirable to obtain them in thin-film form.[4] To this end we used laser ablation deposition as a powerful method to produce the nanoscale structures. We used postdeposition annealing at various substrate temperatures in order to obtain the desired nanostructures. Complementary x-ray diffraction and superconducting quantum interference device (SQUID) magnetometry measurements helped to determine the optimum laser ablation deposition conditions for synthesizing the nanoscale films.

II. EXPERIMENT

A $Ni_{0.5}Zn_{0.5}Fe_2O_4$ target material was prepared from Ni carbonate and α-Fe_2O_3 (Merck), homogenized for 3 h in water in a planetary mill. The powder was dried and calcinated at 850 °C for 3 h. The material was then milled for 2h in water and pressed in disks with the diameter of 27 mm. The resultant disk was sintered in air at 1100 °C for 2 h. The same procedure was followed for the $ZnFe_2O_4$ target, but the starting materials were ZnO and α-Fe_2O_3 (Merck).

Laser ablation deposition was carried out using a KrF excimer laser (Lambda Physik COMPEX 102), having a wavelength of 248 nm and a pulse width of 8 ns.[5] The laser delivered 450 mJ/pulse at a repetition rate of 10 Hz. For each laser pulse, a fluence at target of about 3 J/cm^2 was obtained. The films were deposited on Si(100) wafers (with native silica layers) and had a thickness of about 50 nm. A NiZn ferrite film was deposited with the substrate at room temperature and another complementary film was obtained depositing onto a heated substrate at 550 °C during the entire deposition process. Similarly, a Zn ferrite film was obtained at room temperature followed by subsequent annealing at 550 °C for 1 h.

We employed x-ray diffraction (XRD) and SQUID magnetometry to characterize the structure and properties of the laser ablated films. XRD measurements were performed using a Rigaku D-2013 diffractometer with Cu $K_{\alpha 1}$ radiation at $\lambda = 1.5404$ Å. The SQUID measurements yielded the 300 and 10 K hysteresis loops as well as the zero field cooled–field cooled magnetization curves for all samples obtained.

III. RESULTS AND DISCUSSION

Figure 1(a) displays the x-ray diffraction pattern of a NiZn ferrite film deposited on Si(100) substrate by laser ab-

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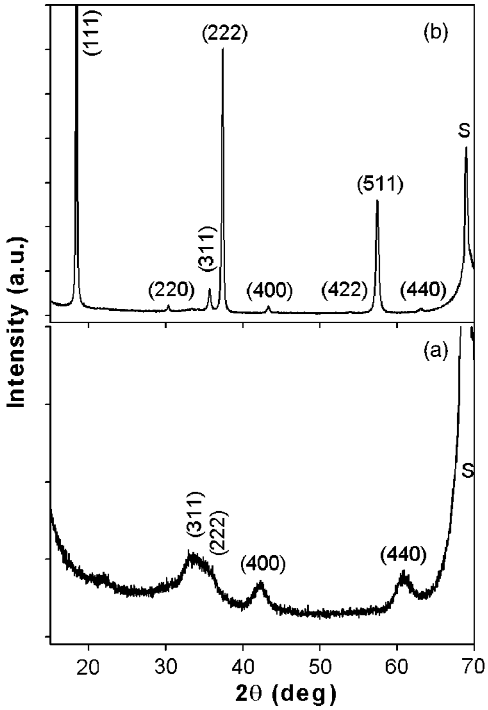

FIG. 1. X-ray diffraction pattern of the NiZn ferrite sample deposited on a Si(100) substrate at a temperature of (a) room temperature and (b) 550 °C. "S" stands for the substrate and the indices pertain to the ferrite peaks.

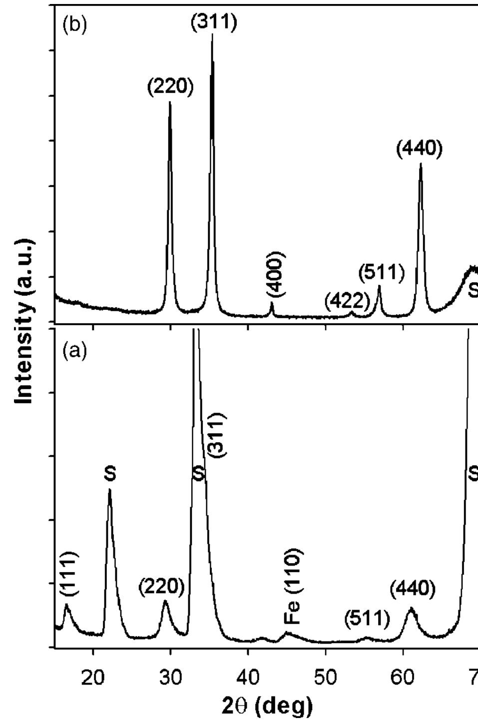

FIG. 2. X-ray diffraction spectra of the Zn ferrite film (a) deposited at room temperature and (b) deposited and subsequently annealed at 550 °C for 1 h.

lation deposition, with the substrate kept at room temperature. Figure 1(b) shows the x-ray diffraction spectrum of the NiZn ferrite deposited on the Si substrate maintained at a temperature of 550 °C during the deposition process. It can be seen by comparison that the heated substrate yielded a better crystalline structure for the ferrite film. The x-ray studies indicate that a polycrystalline ferrite film is formed for select values of the deposition parameters. Determination of the particle size from the XRD linewidth, using the Scherer formula, yields a value of about 30 nm for the grain size in the nanostructured films, as compared to a film thickness of about 50 nm.

Figure 2(a) illustrates the x-ray diffraction pattern of the Zn ferrite film deposited on a substrate at room temperature, and Fig. 2(b) shows the x-ray diffraction spectrum of the same film after annealing at 550 °C for 1 h. Both spectra confirm a crystalline structure in the nanoferrite films, but the annealed sample has sharper lines and is better crystallized compared to the as-deposited film. As effect of Ni additions to the Zn ferrite structure, the XRD patterns are different for the two systems considered.

Figures 3(a) and 3(b) show the zero field cooled–field cooled (ZFC–FC) magnetization curves as a function of temperature, for the Zn and NiZn ferrite films, respectively, recorded in a magnetic field of 500 Oe. The films with post-deposition annealing were selected for these measurements, due to their good crystalline structure. The blocking temperatures obtained are 30 K for the Zn ferrite thin film and 240 K for the NiZn ferrite system. These temperatures define a limit for the onset of the superparamagnetic behavior in these structures and confirm the well-known behavior of the Zn ferrite to be superparamagnetic at room temperature.

Figure 4(a) presents the 10 K hysteresis loop of the NiZn ferrite film, which displays a closed loop with a reduced coercivity relative to the bulk value. The annealed sample was selected for these measurements. Figure 4(b) shows the 300 K hysteresis loop of the NiZn nanoferrite

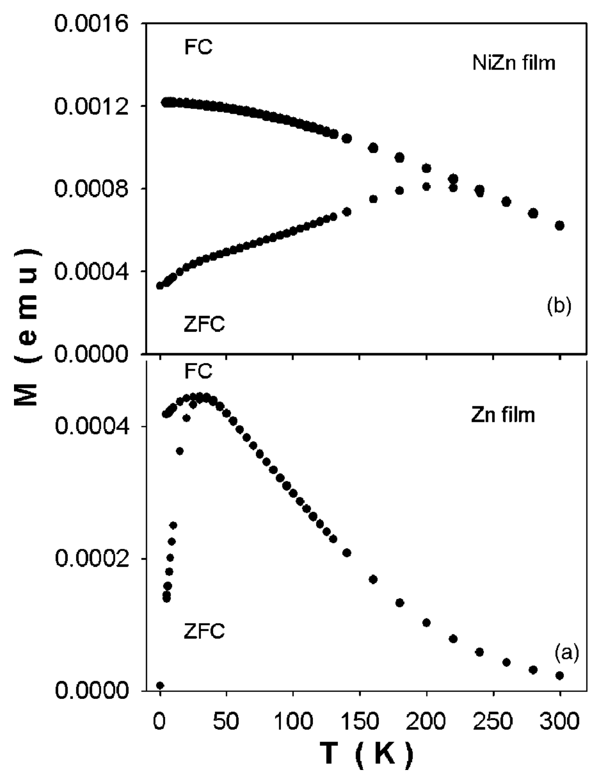

FIG. 3. Zero field cooled–field cooled magnetization curves for the (a) Zn ferrite film and (b) NiZn ferrite film recorded in a magnetic field of 500 Oe.

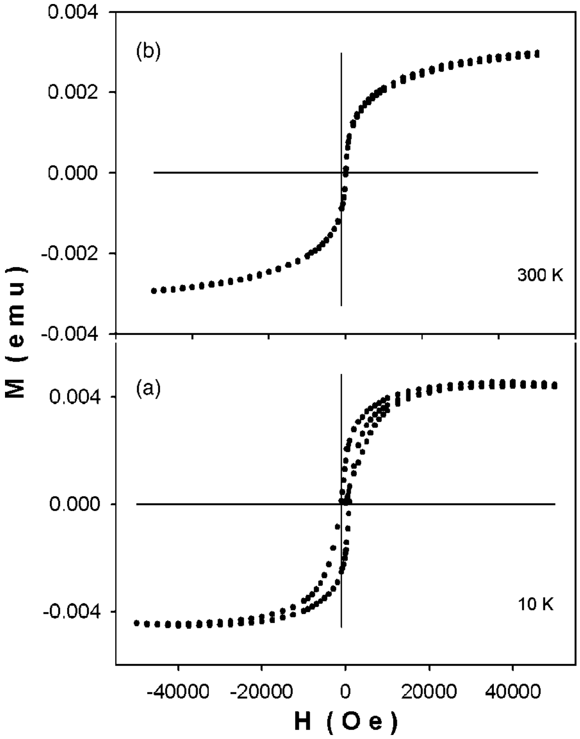

FIG. 4. Hysteresis loops for the NiZn ferrite films recorded at (a) 10 K and (b) 300 K.

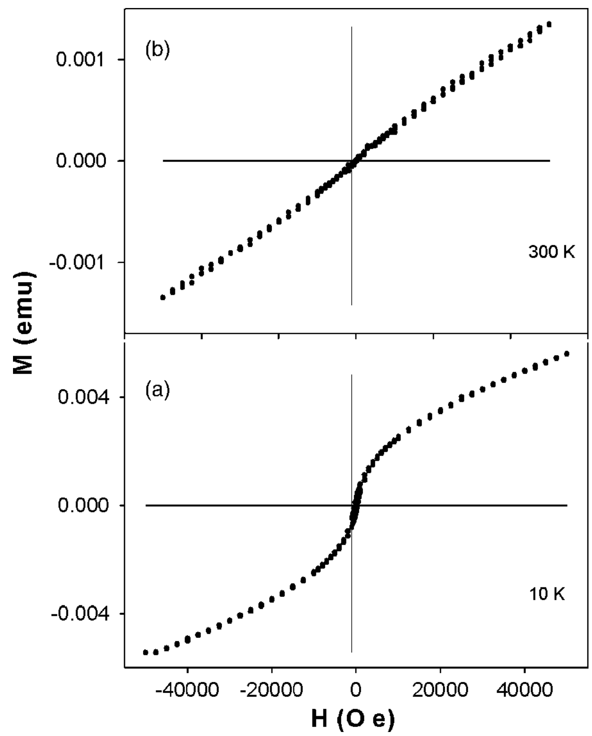

FIG. 5. Hysteresis loops for the Zn ferrite films obtained at (a) 10 K and (b) 300 K.

film, which indicates a superparamagnetic behavior of this system. This result is consistent with atomic force microscopy images, which show island-like growth patterns for the annealed system. A similar superparamagnetic behavior was obtained from the hysteresis loops of Zn ferrite both at 10 and 300 K in Fig. 5, which correlates well with the low blocking temperature for this compound.

By studying various deposition and annealing conditions, it appears that the best crystalline structure and magnetic responses occur for samples deposited at elevated substrate temperatures, followed closely by those with postdeposition annealing.

IV. CONCLUSIONS

We obtained the x-ray diffraction patterns, hysteresis loops, and magnetization curves for a series of NiZn and Zn nanoferrite films obtained by laser ablation deposition on Si substrates. Comparison of bulk and film magnetic properties shows that the magnetic properties of the films are in many respects similar to those of the bulk,[4,6] which makes the laser ablation deposited ferrite films prime candidates for thin-film high-frequency microwave device applications. In particular, the saturation magnetization is very similar to the bulk value, but the coercivity values are slightly different.

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