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Y.Z. Zhang
University of Nebraska - Lincoln

Sy_Hwang Liou
University of Nebraska-Lincoln, sliou@unl.edu

R.J. DeAngelis
University of Nebraska - Lincoln

K.W. Lee
University of Nebraska - Lincoln

C.P. Reed
University of Nebraska - Lincoln

See next page for additional authors

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Y. X. Zhang, S. H. Liou, R. J. DeAngelis, K. W. Lee, C. P. Reed, and A. Nazareth
Center for Materials Research and Analysis, University of Nebraska-Lincoln, Lincoln, Nebraska 68588-0111

Nanostructured Fe/Ag composite films were prepared by magnetron sputtering using a single target. Correlations between the microstructure and magnetic properties, in particular the effects of cluster-size distribution in the films, are discussed. The cluster-size distribution was calculated by analyzing the width and shape of the x-ray-diffraction peaks. The average cluster size in the films increased from 42 to 295 Å and from 38 to 320 Å in the systems of 41 and 27 vol % of Fe, respectively, as the substrate temperature increased from 100 to 500 °C. The maximum magnetic coercivity, as high as 900 Oe, has been observed in the film with 27 vol % of Fe, which was prepared at a substrate temperature of 400 °C.

INTRODUCTION

Finely dispersed magnetic-particle systems are of interest because of the opportunities they present for atomic engineering of materials with specific properties. Depending on the size of the particles, their magnetic properties could be changed from single-domain to multidomain behavior. The particles show superparamagnetic behavior in very-small-particle systems.

There are many methods that have been found effective for creating dispersions of small metal particles in matrices. Early reports involve the formation of gold colloids in a glass[1] and the precipitation of cobalt in dilute Co-Cu alloys.[2] More recently, numerous techniques have been reported, including the formation of nanocrystalline metals by the condensation of metal vapors onto a cold finger,[3] evaporation of metal atoms into solvent matrices,[4,5] cosputtering metal and a matrix-forming oxide,[6,7] Sol-gel techniques,[8,9] and the thermal decomposition of a metal compound with a polymer.[10,11]

In this paper, we show that the nanostructured composite material can be formed by the mixing of two immiscible constituents in the sputtering process. Under equilibrium conditions, the mutual solubility of Ag and Fe is very low in both the solid and liquid form. The solubility of Ag in solid Fe is less than 0.022 at. %. Therefore, the Fe/Ag system must form a composite material. In this type of material, the nanometer-size Fe clusters may have been precipitated out from the surrounding Ag matrix. The cluster-size distribution of these films can easily be controlled by changing the substrate temperature during the sputtering process.[12] In this paper, correlations between the microstructural and magnetic properties, in particular the effects of cluster-size distribution in the films, are discussed.

EXPERIMENTAL PROCEDURE

The Fe/Ag composite metal films were prepared by dc magnetron sputtering using a single target. The sputtering targets were made by sintering a mixture of Fe and Ag powders. The films were prepared with the substrate temperature varied from 100 to 500 °C. The sputtering gas was 4 mTorr of Ar. More detailed process conditions are described in Ref. 12. Film compositions were determined by using energy-dispersive x-ray analysis in a scanning electron microscope. The structure of the films were investigated using a Rigaku x-ray diffractometer with a Cu source. The magnetization and coercive force were measured by a commercial SQUID magnetometer.

RESULTS AND DISCUSSION

Film compositions were determined by an electron microprobe. These films have compositions near $Fe_{35}Ag_{65}$ (27 vol % of Fe) and $Fe_{50}Ag_{50}$ (41 vol % of Fe), respectively. The variation in composition of the films prepared at different substrate temperatures was less than 5 at. %.

The x-ray-diffraction patterns of Fe/Ag films with 41 vol % Fe prepared at substrate temperatures (T_s) from 100 to 500 °C are shown in Fig. 1. There are two important features in this figure: (a) the peak position shifts of the Ag(111) and Fe(110) lines with variations in T_s and (b) the decreasing linewidths of the diffraction peaks with increasing T_s. These indicate that both the d spacing of the crystal planes and the cluster size are functions of T_s. The d spacing of Ag(111), which was calculated from the 2θ

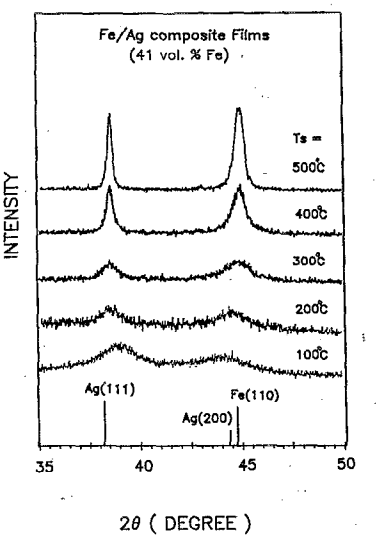

FIG. 1. X-ray θ-2θ scans of Fe/Ag composite films prepared at substrate temperatures of (a) 100 °C, (b) 200 °C, (c) 300 °C, (d) 400 °C, and (e) 500 °C. The Miller indices for elemental Fe and Ag are shown on the bottom of the figure.

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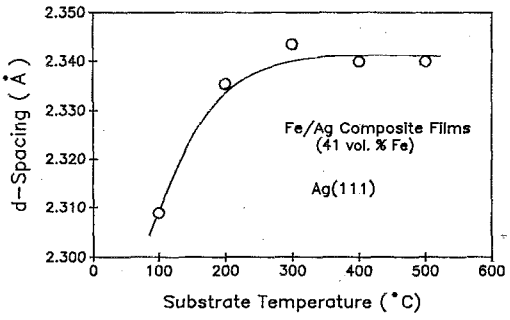

FIG. 2. d spacing of Ag(111) vs the substrate temperatures

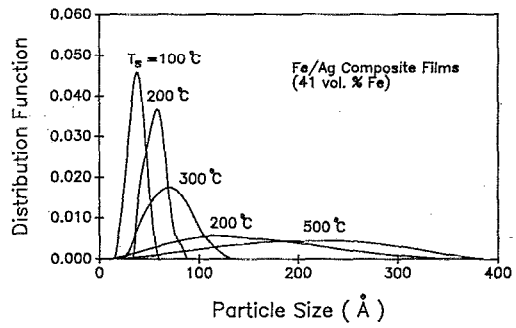

FIG. 3. Particle-size distributions of Fe/Ag composite films (41 vol % of Fe) at substrate temperatures of 100, 200, 300, 400 and 500 °C.

value of Ag(111) as a function of the deposition temperature T_s, is shown in Fig. 2. The d spacing of the Ag(111) increases from 2.310 to 2.345 Å as the substrate temperature of the films increases from 100 to 300 °C and reaches a constant of 2.345 Å for films prepared above 300 °C. The d spacing of Ag(111) prepared at a low substrate temperature (<300 °C) is smaller than that of the bulk silver element. This is expected if we assume that a small amount of iron is mixed in the silver crystal matrix. Although under equilibrium conditions, the mutual solubility of Ag and Fe is very low in both the solid and liquid form, an alloy of Fe-Ag may form under fast quenching conditions. It has been reported that Fe-Ag alloy films with up to 80 at. % Ag were made using coevaporation onto liquid-nitrogen-cooled substrates.[13,14] The amount of Fe mixed in the Ag crystal matrix in the film prepared at a 100 °C substrate temperature is estimated to be about 5% if we assume that the alloying of Fe and Ag follows Vegard's law. The films prepared at a $T_s > 300$ °C have much less alloying between the Fe and Ag as evidenced by the closeness of the d spacing of the film's Ag(111) planes with that of the bulk Ag. These results are similar to the Fe/Ag films with 27 vol % of Fe, which was reported previously.[12] We have observed in Fig. 1 that the 2θ values of the Fe(110) peak increase with increasing T_s. These also can be explained if we assume that a small amount of Ag is mixed in the Fe clusters.

As shown in Fig. 1, the linewidth of the Ag(111) peaks decrease with increasing T_s. This clearly indicates that the particle size of Ag was changed with process conditions T_s. The particle-size distribution (PSD) calculated from the shape of the Ag(111) peaks, using the theory of Warren and the computational technique described previously,[15,16] gives the results shown in Fig. 3. The average particle size increases with increasing substrate temperature, and the distribution of cluster sizes are much broader in the films prepared at higher substrate temperatures.

The average particle size versus T_s of Fe/Ag with a 27 and 41 volume fraction of Fe is shown in Fig. 4. These two systems are quite similar; the average particle size in both systems increase with increasing T_s. As the substrate temperature increases from 100 to 500 °C, the average Ag cluster size in the films increases from 42 to 295 Å and from 38 to 320 Å in the systems of 41 and 27 vol % of Fe, respectively. As seen in Fig. 1, the linewidth of the Fe(110) peaks also decreases with increasing T_s, which behaves similarly to the Ag(111) peaks. Because of the overlap of the Ag(200) and Fe(110) peaks, we cannot make a clear conclusion here. However, one would expect that the particle size of the Fe cluster would increase with increasing T_s.

The magnetic properties of these films were studied by a commercial SQUID magnetometer. The magnetic coercivity (H_c) measured at 25 °C of films with 27 and 41 vol % of Fe versus the substrate temperatures is shown in Figs. 5(a) and 5(b), respectively. The H_c's increase with increasing T_s and reach a maximum for the films prepared at 400 °C substrate temperature. As shown in Fig. 5(a), H_c as high as 900 Oe has been achieved. It is known that the coercivity of fine particles has a striking dependence on their size.[17] As the particle size is reduced, it is typically found that the coercivity increases, goes through a maximum, and then tends toward zero. These magnetic behaviors are due to the mechanism of the magnetization which depends on the particle size. At large sizes, magnetization is changed by domain-wall motion which takes much less energy. The result is small coercivity. Below a critical diameter D_s, which depends on the alloys, the particles become single domains, and in this size range the coercivity reaches a maximum. In this size range, their magnetization is changed by spin rotation, which usually requires a larger energy than domain-wall motion. Below a critical diameter D_s, the coercivity decreases again because of thermal effects

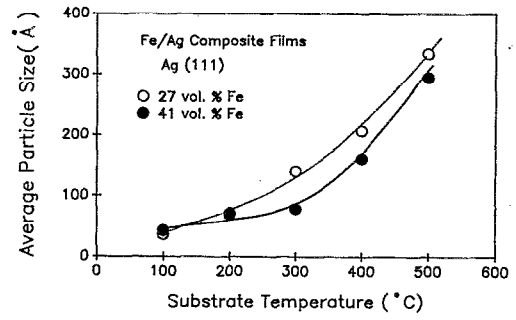

FIG. 4. Average particle size in Fe/Ag composite films (27 and 41 vol % of Fe) vs substrate temperature.

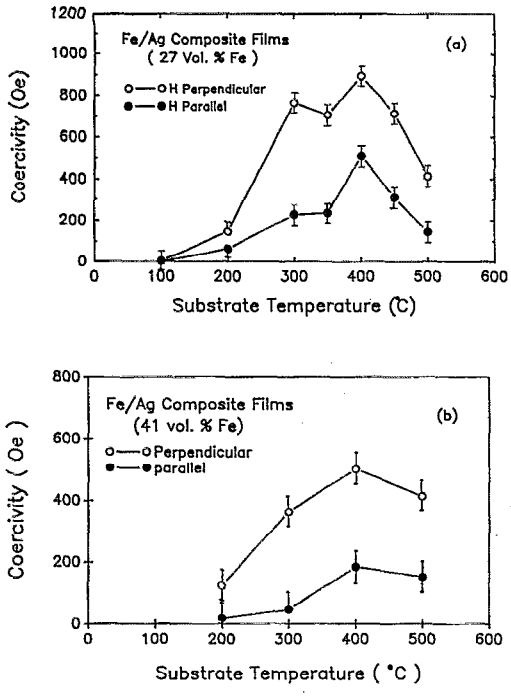

FIG. 5. Magnetic coercivities at 25 °C of Fe/Ag composite films containing (a) 27 vol % of Fe and (b) 41 vol % of Fe deposited at various substrate temperatures. (Data measured when the applied field was both perpendicular and parallel to the film surface are shown.)

which will cause the spontaneous rotation of spin. Such particles are called superparamagnetic. The behavior of H_c found in Fe/Ag composite films, are shown in Fig. 5, can be correlated with their particle sizes. Because the H_c of multidomain iron particles is only a few Oe, the large H_c found in Fe/Ag films indicates that the size of Fe clusters approaches a single domain. The critical diameter for iron particles is about 200 Å, which is in good agreement with the reported by Luborsky.[17]

In summary, we have synthesized Fe/Ag composite metal films with nanometer sizes of Fe and Ag clusters. The ability to control the size of clusters has been demonstrated. The much-enhanced magnetic coercivity was found to be related to the particle size.

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