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Dependence of Magnetic Properties of Fe/Al Multilayers on Layer Thickness and Temperature

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Abstract: ***Magnetic properties of Fe/Al multilayer structures are studied as a function of thickness and temperature using vibrating sample magnetometer (VSM). The XRD measurements show substantial interdiffusion at the interfaces, and particularly at lower thicknesses, these multilayer structures look like a composite single layer film of Fe and Al. The corresponding magnetic measurements show a soft magnetic behaviour of all the films with an in-plane easy direction of the magnetization. However, at low temperature, 100 K, both coercivity and saturation field increases as compared to room temperature below $d_{Fe} \leq 20$ Å and is probably due to the existence of antiferromagnetic interlayer coupling between the iron layers, which coexist with the ferromagnetic interaction within each iron layer and also due to alloying at the interfaces.***

Keywords: Fe/Al multilayers, magnetic properties, Soft magnetic multilayer

1. Introduction

Studies of magnetic interactions between ferromagnetic films separated by non-magnetic metallic films have been a subject of extensive investigations from both theoretical and experimental points of view [1]. Recently, research on high performance magnetic recording heads has pointed out that multilayered structures and artificial super-lattices have revealed good potentialities as well as accurate specifications for the problem of large recording fields and high frequency response [2]. The metallic multilayers (ML) obtained by intercalating deposition of ferromagnetic and non-magnetic films show an improvement of magnetic properties, as well as high magnetic permeability, together with reduced magneto-striction and coercivity [3,4]. In fact, Fe/Ni [2], Fe/Co [5] and Fe-C/Ni-Fe [6] multilayered structures have been reported to have high magnetization and nearly zero magnetosctriction. In this respect, recently Fe/Al bilayer and ML systems have also been studied extensively because of their attractive structural and soft magnetic properties [7-9]. In the past, Fe/Al system has been treated as an ordinary ferromagnet with one transition temperature at the Curie temperature [10]. But Wu et al. [11] and recently Rao et al. [12] have observed a deviation in the magnetic properties of Fe/Al from the normal ferromagnetic behavior and found two magnetic transition temperatures.

However, these interesting properties are greatly influenced by various micro structural ML parameters such as individual layer thickness, number of bilayers and the quality of interfaces formed under different growth conditions [13,14]. Recently, many researchers have found that with decreasing Fe layer thickness, severe interdiffusion occurs at Fe/Al interfaces, resulting in the formation of various kinds of aluminides [15-17]. Consequently, the interaction of ferromagnetic layers across different intermetallic aluminides may show antiferromagnetic or ferromagnetic coupling depending on the kind of aluminides. Although a number of experimental works has been done to understand the mechanism of interlayer coupling in this system, the results are controversial and it is not yet well understood how the formation of iron aluminide in the spacer layer affects the coupling. The nature of this coupling is thought to be dependent on the structure and composition of spacer layer, and thus their measurements can provide important information about the magnetic properties of the multilayers. Therefore, in the present paper, thickness and temperature dependent magnetic properties of electron beam evaporated ultra-thin Fe/Al structures are studied as a function of Fe layer thickness, keeping Al layer thickness constant using vibrating sample magnetometer (VSM).

2. Experimental Details

In the present work, a set of MLS, each with 15 bilayers, was prepared with a constant Al thickness of 10 Å and Fe layer thickness varying from 10 Å to 40 Å in step of 10 Å, respectively, on float glass and silicon substrates, using e-beam evaporation system under UHV ($\sim 8 \times 10^{-9}$ Torr) conditions at room temperature. Deposition rate of 0.1 Å/s for both Fe and Al was controlled using quartz crystal thickness monitor. A capping layer of 20 Å of Al was also deposited on the top of each sample in order to protect the MLS from oxidation. The first layer on the substrate was of Al. The corresponding bilayers samples were also deposited under the same conditions in a single run without breaking the vacuum using the substrate masking facility.

Analysis of the structural and phase composition investigations of the MLS was carried out using GIXRD. All the GIXRD patterns recorded at an incidence angle of 0.5°. The corresponding magnetic properties were measured at 300 K and 100 K using vibrating sample magnetometer (VSM) [18].

3. Results and Discussion

3.1 Grazing incidence x-ray diffraction measurements

Magnetic properties are strongly dependent on crystal structure of the material. Thus, the crystal properties of these MLS were analyzed in order to identify the causes of the variation in magnetic properties. Figure. 1 shows the GIXRD patterns of as-deposited $[Fe/Al]_{x15}$ MLS as a function of Fe layer thickness [15]. From the recorded diffraction curves, it is clearly seen that all the deposited MLS are textured mainly along (110) direction of α-Fe. The peaks corresponding to Al were not detected in any of the recorded spectrums, indicating that the deposited ultra-thin Al layer is amorphous or nanocrystalline in nature. However, the peak corresponding to α–Fe (110) of ML samples was significantly different from that of bulk Fe. We interpret that the peak position shift is caused by the elongation of the (110) interplanar distance ‘d’ due to large internal stress in the Fe layers induced by adjacent Al layers, and their intermixing during deposition causing the formation of iron aluminide layer at the interface. Further, the intensity of (110) peak reduces substantially and FWHM increases with decreasing Fe layer thickness, particularly for $d_{Fe} \leq 20$ Å, the peak shows a broad hump around $2\theta = 43.28°$. Indeed, this is expected because the Al and Fe thickness involved are very small and may not form continuous layers that would lead to well define interfaces. The deposited structures in these cases show a single mixed layer of Fe and Al clusters.

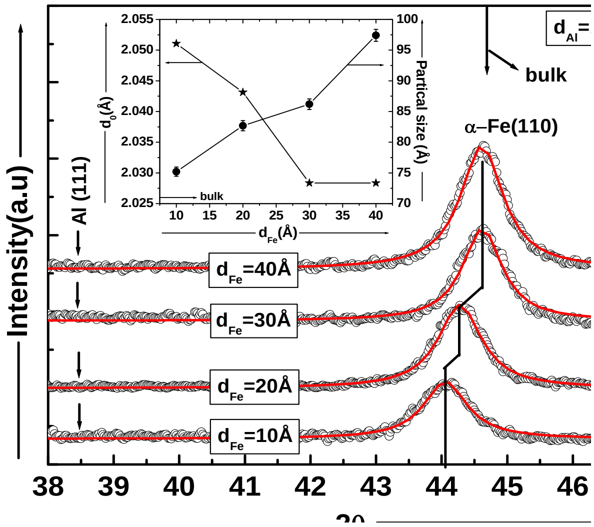

Figure 1: GIXRD patterns of the as prepared $[Fe/Al]_{x15}$ MLS as a function of Fe layer thickness. Inset shows the dependence of the average spacing ‘d’ and particle size on the Fe layer thickness.

Grain size of Fe crystallites is a critical structural parameter to modify the magnetic properties, together with the grain orientation, which controls the magnetic anisotropy. Therefore, we have determined the average particle size from the recorded GIXRD patterns using Scherrer formulism as shown in the inset of figure 1. It is found that the particle decreases substantially with decreasing d_{Fe}, indicating that ML with greater d_{Fe} have larger and more oriented α–Fe crystallites and below $d_{Fe} \leq 20$ Å appeared to be composed of nanocrystallites or amorphous Fe grains, which we assume, is due to the substantial intermixing at the interfaces, leading to a distorted Fe lattice structure. In addition to this, we have also measured the d spacing of α-Fe crystallites in these MLS as a function of d_{Fe} (shown in the same fig. 1). It is found that the d spacing decreases from 2.052 Å to 2.028 Å as the Fe layer thickness increases and this matches fairly well with bulk d spacing (d=2.026 Å) at d_{Fe}=40 Å. The variation in the d spacing as a function of d_{Fe} indicates the presence of stresses in the deposited layers. At lower Fe thicknesses it suggests a compressive stress at the interfaces, which is released as d_{Fe} increases to form a continuous layer.

3.2 Magnetic measurements

The magnetic measurements reported in the present study were carried out using a low field vibrating sample magnetometer. In all the measurements, the magnetic field was applied parallel to the surface of the film plane and hysteresis loops were recorded up to the saturation of the magnetization.

Fig. 2 shows the typical M-H loops of Fe/Al MLS as a function of d_{Fe} recorded at 300 K and 100K. It should be noted that the entire MLS show an easy saturation of the magnetization with applied magnetic field suggesting an in-plane easy direction of the magnetization. The coercivity (H_c), saturation field (H_s) and magnetization (M_s) values determined from the hysteresis loops are plotted as a function of Fe layer thickness at both the temperatures as shown in fig. 2d. The small values of H_c and H_s indicate a soft magnetic behavior of the MLS and can be explained by the weak crystalline magnetic anisotropy due to the existence of small crystal grains and negligible magnetostriction. Additionally, it has been reported in the literature that the domain wall energy, in MLS consisting of magnetic and non-magnetic layers, becomes smaller than that of the single layer film [19]. Therefore, the decrease of the domain wall energy due to the magnetostatic coupling between Fe layers could be an additional reason for improving the soft magnetic properties. Similar results are also reported by M. Senda et al. in their investigation carried out on MLS consisting of Fe and nonmagnetic layers of Al_2O_3, Cu, C, Si and Ti prepared with the sputtering technique [3]. However, some interesting changes are observed in coercivity (H_c) and saturation field (H_s) at low temperature (100 K) as the Fe layer thickness decreases from 40 Å to 10 Å as shown in fig. 2. At room temperature, both H_c and H_s decreases as the Fe layer thickness decreases and show a minimum at d_{Fe}=10 Å. However, at low temperature (100 K), both H_c and H_s increases below $d_{Fe} \leq 20$ Å as compared to room temperature. We deduce that this is caused by the exchange coupling between Fe layers and Al layers. With decreasing Fe layer thickness $d_{Fe} \leq 20$ Å, the relative number of Fe atoms that are exchanged coupled to the Al layer increase. The exchange coupling can pin the interface spins of the soft Fe layer, leading to the increase of coercivity with the decrease of Fe layer thickness at low temperature. Also, $d_{Fe} \leq 20$ Å film behave like a single domain region, where magnetization reversed takes place only by rotation of saturation magnetization (M_s) vector in accordance with the Stoner and Wonefarth model may be one of the causes for high coercivity value. Similar to H_c and H_s, M_s also decreases as Fe layer thickness decreases, but no change is observed in the value of M_s at both the temperatures.

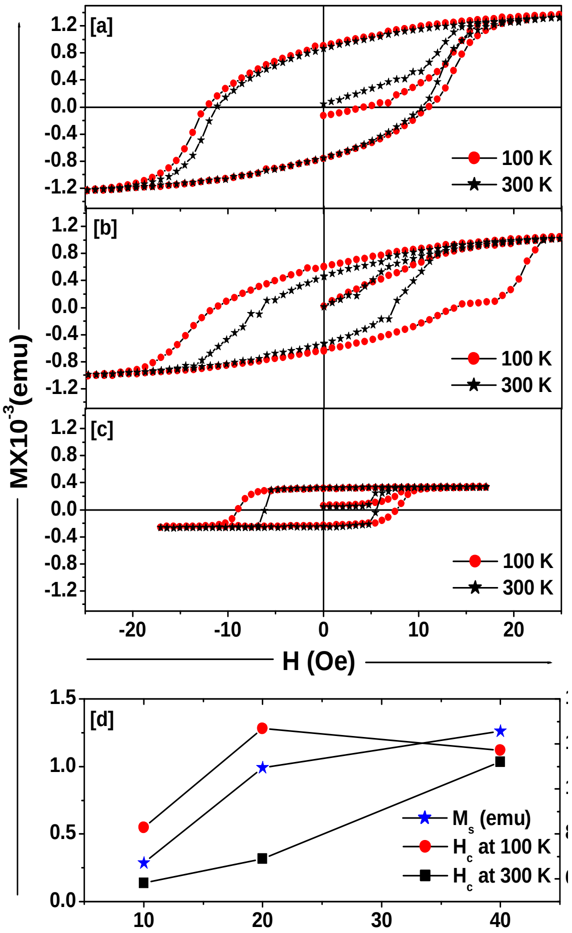

Figure 2: Hysteresis loops of the as prepared a) [Fe (40 Å)/Al (10 Å)]$_{x15}$, b) [Fe (20 Å)/Al (10 Å)]$_{x15}$ and c) [Fe (10 Å)/Al (10 Å)]$_{x15}$ MLS and d) dependence of the H_c and M_s values on the Fe layer thickness at 300 K and 100 K.

The M_s values obtained for these MLS are much lower than that of the bulk Fe, suggesting the formation of a nonmagnetic FeAl intermetallic layer at the interface which is also reflected from our structural studies.

Similarly, M. Carbucicchio et al. has also investigated the magnetic properties of e-beam evaporated Fe/Al ultrathin film multilayer as a function of Fe layer thickness by means of conversion electron Mossbauer spectroscopy, alternating gradient force magnetometry and AC suceptometry. They found that with decreasing Fe layer thickness; Al diffusion causes a progressive loss of periodicity, giving rise to the formation of iron-aluminum solid solution, Fe (Al)$_{ss}$ and intermetallic compounds at the interfaces and also the magnetic behaviour progressively evolves from ferromagnetic to super-paramagnetic [20]. However, the present results are different from the above-mentioned case. The present results do not show a super-paramagnetic behaviour as superparamagnet has no hysteresis, i.e. both the remanent magnetization and coercivity are zero and also the M (H) loops taken at different temperatures should superimpose in a M vs. H/T plot, in contrast to the present experimental findings. Therefore, the observed drastic changes in the hysteresis loops can be better explained as follows: (i) As Fe layer thickness is reduced below a critical value the deposited structure does not form a MLS at all and the resulting deposited MLS, as indicated by the structural studies, resembles a composite single layer film consisting of Fe and Al nano size clusters. (ii) The decrease of the grain size and pure ferromagnetic Fe content corresponds to an increase of the surrounding paramagnetic compounds at the interfaces and (iii) probably due to the existence of antiferromagnetic interlayer coupling between the iron layers, which coexist with the ferromagnetic interaction within each iron layer. However, as per the best of our knowledge, this type of behavior has not been reported earlier. A better understanding of this behaviour requires detailed analysis of the system based on further experimental observations.

4. Conclusions

The paper presents thickness and temperature dependent magnetic properties of ultra-thin Fe/Al structures as a function of Fe layer thickness. The structural studies on these MLS show substantial intermixing at the interfaces and the deposited structure resembles a composite single layer film consisting of Fe and Al clusters at lower Fe layer thicknesses. The observed changes in the magnetic behavior at low temperature, particularly below $d_{Fe} \leq 20$ Å, is probably due to the existence of antiferromagnetic interlayer coupling and the alloying at the interfaces.

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