Low temperature noncollinear behavior in ultrathin Fe/Al multilayer structures

R. Brajpuriya1,2,a)
1UGC-DAE Consortium for Scientific Research, University Campus, Khandwa Road, Indore 452 001, India
2Sincrotrone Trieste S. C. p. A., S.S. 14 Km 163.5, I-34012 Trieste, Italy

(Received 10 December 2009; accepted 9 March 2010; published online 26 April 2010)

We report the low temperature noncollinear magnetic behavior of electron beam evaporated ultrathin Fe/Al multilayer (ML) structures. Investigations have been carried out with ML samples with Fe-layer thicknesses in a range of 10–40 Å and Al-layer thickness of 10 Å. The structural studies show that heavy atomic interdiffusion between Fe and Al layers occurs at the interface, resulting MLs of different complicated structures according to different sublayer thicknesses Brajpuriya, et al., [Eur. Phys. J. B 51, 131 (2006)]. The magnetic measurements show that these films are “re-entrant” systems and not ordinary ferromagnets as often assumed. The obtained results, interestingly, indicate a transition from ferromagnetic state to a low temperature disordered state where a collective frozen magnetic state with grain moments oriented randomly occurs. We interpret the observed low temperature noncollinear magnetic behavior to be due to random freezing of grain moments. © 2010 American Institute of Physics. [doi:10.1063/1.3383049]

I. INTRODUCTION

Studies of magnetic interactions between ferromagnetic films separated by nonmagnetic metallic films have been a subject of extensive investigations from both theoretical and experimental points of view. These kinds of artificial structures are expected to provide an understanding of surface magnetism and transport phenomena, such as interlayer magnetic-coupling, surface anisotropy,1,2 magneto-optical effect, and giant magnetoresistance, etc.3–4 In recent years, the existence of disordered magnetic states at low temperature, possibly due to the frustration arising as a consequence of competing exchange interactions or random anisotropy has been the focus of much attention of researchers. The presence of random interparticle interactions in the system can give rise to the appearance of a spin (cluster) glass phase of multiple metastable disordered magnetic states.5–7 On warming such a system from low temperature, the spin-glass order melts first, followed by the loss of magnetic order at Curie temperature, $T_c$. These properties were observed mostly in three-dimensional (3D) systems such as bulk crystalline and amorphous materials.8,9 On the other hand, nanostructured materials (ferrofluids,10,11 interacting nanoparticles,12 and polymers containing magnetic components13) exhibit unusual low temperature behavior suggestive of that observed in random or disordered magnetic systems due to the random distribution of anisotropy axis, interparticle interactions, and surface effects.14 However, in most cases the correlation between the magnetic behavior and the morphological origin as well as the nature of the low temperature phase for a system of interacting nanoparticles remains controversial and unclear. As the macroscopic behavior of nanostructured magnetic system is determined by the structure, size, morphology of the constituent phases, and by the type and strength of the magnetic coupling between them, it is very much important to understand how such macroscopic properties arise from the interplay of microscopic parameters to design magnetic materials for specific applications.

The fundamental magnetic and electronic properties of these structures are greatly influenced by various microstructural multilayer (ML) parameters such as the individual layer thickness, the number of bilayers, and the quality of interfaces formed under different growth conditions.15–18 The real layers always have a certain roughness because the technologies of their preparation are imperfect. For this reason, a ferromagnetic film obtained with a small amount of an evaporated substance has a discontinuous structure, i.e., separated ferromagnetic clusters appear. These structures have been studied in many works. Both nonferromagnetic metals19–21 and insulators22,23 are used as layers between the ferromagnetic layers. As a rule, the possible magnetic states of the system were not analyzed in detail in these works. Most authors consider either a narrow temperature range or fixed thicknesses of the ferromagnetic layers, etc. A comparatively complete set of works is devoted to studying the properties of the Co/Fe/Al$_2$O$_3$ discontinuous ML metal–insulator structure22,23 with the discontinuous ferromagnetic layers. The magnetic state of such a system is determined primarily by the dipole–dipole interaction between the particles within the ferromagnetic layers, whereas, the interlayer interaction is negligibly weak. The existence of various magnetic phases including ferromagnetic, superferromagnetic, and spin-glass phases were revealed in those works for various nominal thicknesses of the magnetic layer and various temperatures.

The superparamagnetic behavior was observed in a number of structures with a metal layer, where the Ruderman–Kittel–Kasuya–Yosida exchange interaction between the ul-
trathin ferromagnetic layers is important [e.g., Co/Cu, Co/Ag, and Fe/Si (Ref. 21)]. Of particular interest is the Fe/Al system, which has been actively studied for the last decades because it has been shown to possess excellent soft magnetic properties. Most investigations concerned the nature of the exchange interaction between the Fe layers, its connection with the magnetic and structural properties of the Al layers, and the dependence on the roughness degree of the interlayer interfaces in superlattices. Results of these investigations clearly reveal a wide variety of magnetic interactions at the interfaces that are characteristic of iron, intermetallic, and nonmagnetic phases. A decrease in the thickness of the Fe layers down to the ultrathin and discontinuous must result in the transformation of the magnetic state of the ML structures. Recently Carbucicchio et al. have investigated the magnetic properties of ultrathin Fe/Al film ML films as a function of Fe layer thickness by means of conversion electron Mössbauer spectroscopy, alternating gradient force magnetometry, and ac susceptibility and obtain very interesting results. 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), and paramagnetic compounds at the interfaces. Thus, it is interesting to see how these ML structures behave at low temperature. In the past, Fe/Al ML system has been treated as an ordinary ferromagnet with one transition temperature at the Curie temperature. However, in the present study we have observed a deviation in the magnetic properties of Fe/Al from the normal ferromagnetic behavior. The obtained results clearly indicate a magnetic phase transition from high temperature ferromagnetic state to a low temperature disordered state similar to a spin-glass phase. It is proposed that the noncollinear magnetic behavior at low temperature originates from the tendency of the freezing of random oriented magnetic moments.

**II. EXPERIMENTAL DETAILS**

In the present work, a set of ML samples, each with 15 bilayers, were prepared with a constant Al thickness of 10 Å and a Fe layer thickness varying from 10 Å to 40 Å in steps of 10 Å, respectively, on float glass substrates, using an e-beam evaporation system under UHV (~8 × 10^-9 Torr) conditions at room temperature (RT). The deposition rate of 0.1 Å/s for both Fe and Al was controlled using a quartz crystal thickness monitor. A capping layer of 25 Å of Al was deposited on the top of each sample in order to protect the MLs from oxidation. The first layer on the substrate was Al.

The microstructural and morphological investigations of the MLs were carried out using grazing incidence x-ray diffraction and atomic force microscopy techniques. The low temperature magnetic properties of Fe/Al ML structures were investigated by performing dc magnetization and magnetization versus field (M−H) hysteresis loops measurements using Quantum design superconducting quantum interference device magnetometer and vibrating sample magnetometer (VSM) as a function of temperature down to 5 K. The calibration on magnetization measurement was done by using standard Ni sample.

**III. MAGNETIC MEASUREMENTS**

To understand the nature of the magnetic behavior at low temperature, the temperature dependent magnetization (M−T) measurement was performed. In the M−T measurement, the sample was cooled in zero-field to a lower temperature [zero-field-cooled (ZFC)]. The magnetization was then recorded on heating the sample to high temperature under a constant applied field. Similarly, the sample was cooled in a constant applied field to a lower temperature and then the magnetization was recorded on heating the sample to high temperature with maintaining the same applied field [field-cooled (FC)]. Figure 1 shows the temperature dependence of $M_{ZFC}(T)$ and $M_{FC}(T)$ curves measured under 100 and 500 Oe applied field. The magnetization initially rose as the temperature increased to a certain temperature $T_m$, beyond which the magnetization decreased with further increase in temperature. The $M_{FC}(T)$ and $M_{ZFC}(T)$ dependences coincide in the high-temperature region but they begin to be different as temperature decreases. When the sample was subsequently cooled in the same field, the magnetization did not retrace the initial curve in all the temperature ranges especially below the temperature $T_m$ at which the magnetization has peaked. The transition temperature, $T_m$, is the temperature at which the magnetization peaks and below which irreversibilities set in and the system might enter a different magnetic phase. Qualitatively similar temperature dependences of the magnetization are observed for all of the samples under investigation, i.e., peak in the magnetization and irreversibilities. Every curve exhibits a peak, which shifts to higher temperature (from ~45 to ~151 K) with decreasing Fe layer thickness. Thus, the magnetic state of the samples at low temperatures is irreversible and depends on its magnetic prehistory.
As reported previously,\textsuperscript{30} the samples with sufficiently thin iron layers at high temperatures behave as superparamagnets. Such a behavior indicates that the ultrathin Fe layers, being inhomogeneous in their thickness, are separated into clusters the interaction between which is weaker than the interaction inside a cluster. The irreversible behavior of the magnetization of such a system at low temperatures can be attributed to two causes,\textsuperscript{23} first, the blockade of the magnetic moment of each cluster due to the anisotropy of individual particles and second, the appearance of the collective spin-glass state associated with a random interparticle interaction. This anomaly disappears as the temperature increases. The observed behavior indicates that the system at low temperatures can be in the spin-glass state characterized by many local energy minima with various total magnetizations.\textsuperscript{31} The transition of the system from the state with a small magnetic moment to the state with a large magnetic moment requires much higher fields than those necessary for the rotation of this magnetic moment as a whole.

As the nominal thickness of the evaporated iron layer increases, interactions inside the magnetic layer increases. This increase results in the decrease in the spin-glass transition temperature. As pointed out by the authors of Refs. 13, 14, and 32, the spin-glass phenomena observed in those works are primarily associated with such isolated iron clusters whose structure and concentration are determined by the sample sputtering technology and are difficult to control. The authors of Refs. 13, 14, and 32 also pointed out that the directions of the anisotropy in a polycrystalline iron film are different for different crystallites. Owing to this circumstance, the ferromagnetic layers in the ground state may be divided into isolated domains, and the interlayer exchange interaction is of a pinning-potential character in this case. As a result, such Fe layers can also make a contribution to the splitting between the $M_{FE}(T)$ and $M_{ZFC}(T)$ curves and other phenomena inherent in spin glasses.

However, this irreversible behavior and the peak in the magnetization are not the properties of a typical ferromagnetic material-like iron. To see whether we have a paramagnetic state above the transition temperature, $T_m$, in the Fe/Al MLs, we measured the magnetic hysteresis loop at RT (see Fig. 2). Clearly, from the figure, the state is not a paramagnetic one but rather a ferromagneticleike state with the coercivity of about 11 Oe in in-plane direction. To confirm that the observed $M(T)$ behavior is due to the ML nature of the sample and is no way connected to anomalous behavior of the iron film itself, $M(T)$ data were taken on a Fe (50 Å) single layer film taken at 100 Oe field. Magnetization for the Fe film varied monotonically with $T$ and found no peak in the magnetization as well as irreversible behavior. Similar kind of behavior is also observed by Vitali et al.\textsuperscript{33} when measured the magnetization on 30 and 200 Å single layer iron film. Thus the observed behavior of $M(T)$ seems to be a property of Fe/Al ML system and existence of the interlayer coupling between the iron layers which coexist with the ferromagnetic interaction with in each iron layers is causing the peak in the magnetization curve. This is similar in principle to what happens in spin-glass materials. In these materials, the competition between ferromagnetic and antiferromagnetic interaction causes the spin-glass ordering that is responsible for the peak in the magnetization and irreversibilities. However, at a relatively high applied magnetic field of 500 Oe the peak disappears and the separation between ZFC and FC curves decreases (nearly overlap) as shown in Fig. 1(d) for [Fe(40 Å)/Al(10 Å)]\textsubscript{X15} MLs. This indicates that $T_m$ does not represent a transition from ferromagnetic to paramagnetism. The author thinks that $T_m$ represents a transition from a ferromagnetic state to a low temperature disordered state where a collective frozen magnetic state with grain moments oriented randomly occurs. Such systems are known as “re-entrant” systems.

Further, it is interesting to see which theoretical expression describes temperature dependence of magnetization. At low temperature, for small values of $H$, the magnetization $M(T)$ is well described in terms of the spin wave approximation,\textsuperscript{34,35} namely

$$M(T) \equiv M(0)(1 - BT^{3/2}),$$

(1)

where, $M(0)$ is the saturation magnetization at 0 K and corresponds to complete alignment of moments and $B$ is the Bloch coefficient related to spin wave excitation. To check this temperature dependence of $M$, magnetization data was taken on [Fe(40 Å)/Al(10 Å)]\textsubscript{X15} MLs between RT and 150 K. Equation (1) is valid for a 3D ferromagnet. If iron layers in a ML sample are decoupled, hence, representing quasi-two-dimensional magnetic structure, then the above relation may not be valid. If there is a coupling between the iron layers, however, small it may be, a MLs is likely to behave as a 3D magnetic solid and the above formula may be applied. Assuming that is the case, a plot of $M$ versus $T^{3/2}$ is shown in Fig. 3, where $M$ values used are the one obtained under the FC condition. The plot clearly shows a linear behavior between $M$ and $T^{3/2}$ as expected from Eq. (1). The applicability of Eq. (1) also suggests that the iron films in these MLs are not decoupled from each other but seem to be magnetically coupled to each other. However, this temperature dependence is in contrast to the Stoner–Wohlfarth model,\textsuperscript{36} which is a $T^2$ dependence. Since the $T^{3/2}$ depen-
dence gives a slightly better fit, \( M(0) \) and the coefficient \( B \) are determined using a fit to Eq. (1). The fit gave \( M(0) = 1.42 \times 10^3 \) emu/cc and \( B = 7.66 \times 10^{-6} \) K\(^{-3/2} \).

In order to obtain more information hysteresis loops (\( M-H \) curves) were also recorded using VSM at RT and 100 K. For these measurements, the low magnetic field was applied parallel to the surface of the film plane and hysteresis loops were recorded up to the saturation of the magnetization. Figure 4 shows the typical \( M-H \) loops of \([\text{Fe}(d_{\text{Fe}})/\text{Al}(10 \text{ Å})]_{15} \) MLs having \( d_{\text{Fe}} = 30 \) Å and 10 Å, respectively. It should be noted that both MLs show low coercivity \( (H_c) \) and 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 at both temperature are tabulated in Table I. The small values of \( H_s \) and \( H_c \) indicate a soft magnetic behavior of the ML films and are thought to be caused by the weaker crystalline magnetic anisotropy due to the existence of small crystal grains in Fe layers. It has been reported in the literature that the domain wall energy, in the ML films and are thought to be caused by the existence of small crystal grains in Fe layers. It has been reported in the literature that the domain wall energy, in the ML films and are thought to be caused by the existence of small crystal grains in Fe layers.

However, some interesting changes are observed in coercivity \( (H_c) \) as well as saturation field \( (H_s) \) values at low temperature (i.e., 100 K) as the Fe layer thickness decreases from 40 to 10 Å as shown in Table I. At RT, both \( H_c \) and \( H_s \) decreases as the Fe layer thickness decreases and show a minimum at \( d_{\text{Fe}} = 10 \) Å. However, at low temperature, i.e., at 100 K, both \( H_c \) and \( H_s \) increases below \( d_{\text{Fe}} \leq 20 \) Å as compared to RT. The author expects that this be caused by the exchange coupling between Fe and Al layers. As Fe layer thickness decreases below \( d_{\text{Fe}} \leq 20 \) Å, the relative number of Fe atoms that are exchange coupled to the Al layer increase. The exchange coupling can pin the interface spins of the soft Fe layer, leading to the increase in coercivity and saturation field with the decrease in Fe layer thickness at low temperature. Similar to \( H_c \) and \( H_s \), \( M_s \) also decreases as Fe layer thickness decreases but not much change is observed in the values of \( M_s \) at both temperatures. However, \( M_s \) values obtained for these MLs are much lower than that of the bulk Fe. The saturated magnetization of iron films, prepared in UHV, is reported to be the same as that of bulk iron if the film thickness is equal or higher than 25 Å. Therefore, much lower values of \( M \) for these MLs may be due to the following possibilities, namely, (1) the iron film may have a small percentage of its thickness as “magnetically dead” thereby making the effective thickness of the iron film smaller than 25 Å, which may have lower magnetization value than that of bulk iron and (2) interdiffusion of iron and aluminum atoms at the interface and/or formation of nonmagnetic compounds at the interfaces.

### Table I. Magnetic parameters at 300 K (RT) and 100 K.

<table>
<thead>
<tr>
<th></th>
<th>~( H_s ) (Oe)</th>
<th>300 K</th>
<th>100 K</th>
<th>~( H_s ) (Oe)</th>
<th>300 K</th>
<th>100 K</th>
<th>~( M_s ) (×10^3 emu/cc)</th>
<th>300 K</th>
<th>100 K</th>
<th>~( M_s ) (×10^3 emu/cc)</th>
<th>300 K</th>
<th>100 K</th>
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</thead>
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<tr>
<td>MLs-1</td>
<td>5.9</td>
<td>8.3</td>
<td></td>
<td>8.1</td>
<td>14.7</td>
<td></td>
<td>0.67</td>
<td>0.63</td>
<td></td>
<td>0.75</td>
<td>0.75</td>
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</tr>
<tr>
<td>MLs-2</td>
<td>6.5</td>
<td>12.5</td>
<td></td>
<td>26.9</td>
<td>33.8</td>
<td></td>
<td>0.38</td>
<td>0.54</td>
<td></td>
<td>0.87</td>
<td>0.87</td>
<td></td>
</tr>
<tr>
<td>MLs-3</td>
<td>10.2</td>
<td>10.4</td>
<td></td>
<td>39.8</td>
<td>41.2</td>
<td></td>
<td>0.62</td>
<td>0.55</td>
<td></td>
<td>1.23</td>
<td>1.23</td>
<td></td>
</tr>
<tr>
<td>MLs-4</td>
<td>10.7</td>
<td>11.2</td>
<td></td>
<td>30.2</td>
<td>30.8</td>
<td></td>
<td>0.81</td>
<td>0.83</td>
<td></td>
<td>1.35</td>
<td>1.35</td>
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</tr>
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</table>
IV. DISCUSSION

The results obtained from the magnetization measurements suggest that the low temperature magnetization behavior is dominated by the spin dependent scattering, which could be systematically controlled by the application of external magnetic field. Moreover, it indicates that by decreasing the temperature below $T_m$, the soft ferromagnetic system observed at higher temperature evolves towards a noncollinear magnetic structure. For temperature above $T_m$, the random oriented spins are magnetized along with the adjacent grains by contact with their molecular field and the whole system is expected to behave as a soft ferromagnet with lower coercivity. On decreasing the temperature below $T_m$, the random oriented spins start to freeze toward the ground state reducing the ferromagnetic exchange coupling between grains, which is possibly a main source for enhancement in coercivity.

The present experimental results can be interpreted from the generalized random anisotropy model (RAM), as discussed previously for other nanocrystalline systems. According to the RAM for two phase system, the ferromagnetic exchange interaction between two adjacent grains ($\xi_{ij}$) and exchange correlation length through the intergrain boundaries ($\xi_g$) are defined as $\xi_{ij} = \alpha A$ (where $A$ is exchange coupling coefficient ($A = 10^{-11}$ J m$^{-1}$) and $\alpha$ is the factor ranging between 0 and 1) and $\xi_g = \sqrt{K/\gamma A}$ (where $K$ is experimental anisotropy constant of about 3 $\times 10^{-11}$ J m$^{-3}$ at RT and $\gamma$ is a parameter varying between 0 and 1 and closely related to the exchange correlation length of the amorphous matrix), respectively. The observed ferromagnetic behavior at RT could be explained from the above relations, since the average crystallite size ($d \approx 7-9$ nm) is smaller than the $\xi_g$ ($>15$ nm), if $\gamma=1$ together with the assumption that the grain boundary thickness ($\delta$) is smaller than the exchange correlation length of the interface ($\xi_g$). In addition, the condition $\xi_g > \delta$ implies that the spins at the grain boundary are oriented in the direction imposed by the surrounding environment and hence there exists a ferromagnetic exchange interaction between crystallites and the whole system is ferromagnetic and soft. By decreasing the temperature, (i) the $\xi_{ij}$ decreases, (ii) the anisotropy at the boundary grows large, and (iii) the links holding the boundary spins aligned to the magnetization vectors of the surrounding crystallites are progressively severed. As a result, the grains are uncoupled but they interact strongly through dipole-dipole interactions, which can be either ferromagnetic or antiferromagnetic. The consequent mixing of these interactions which compete with local anisotropy is responsible for the random freezing of the grain moments, which leads to a noncollinear magnetic structure at lower temperature. The freezing of grain moments entails that the whole system may act as a spin-glasslike system, which is fully coherent with the present experimental results.

V. CONCLUSIONS

The low temperature noncollinear magnetic behavior of the evaporated Fe/Al ML structures, investigated by the dc magnetization, has been reported. All the films show a soft ferromagnetic behavior at RT and are mainly attributed to weaker crystalline magnetic anisotropy owing to the presence of small crystal grains in Fe layers. These films appear to have a re-entrant phase where the system enters a spin-glasslike phase from a ferromagnetic phase upon cooling, which were interpreted from a generalized RAM. We interpret the observed low temperature noncollinear magnetic behavior to be due to random freezing of grain moments. Interestingly, from theoretical point of view the present system provides an opportunity for studying frustration associated with interactions at experimentally accessible temperature and provides a system in which there is random anisotropy along with these competing (dipolar) exchange interactions.

ACKNOWLEDGMENTS

The author would like to acknowledge CSIR, UGC, Dr. D. S. Kothari PDF, India and ICTP-TRIL Program, Italy for research fellowship. Acknowledgments are also given to A. Benerjee, S. B. Roy, and T. Shripathi for magnetic measurements and discussion. This work is dedicated to the memory of Dr. S. M. Chaudhari who untimely passed away during this work.