The ferromagnetic CoFeB has been extensively employed in various spintronics devices like magnetic tunnel junctions [1–3], spin valves [4], and spin-torque devices [5, 6] over the last decade. The electrical conduction in these devices utilising the spin degree of freedom depends on the composition, spin polarization, and crystallinity of the CoFeB layers. Therefore, a basic understanding of the electrical and magneto-transport properties of CoFeB thin films prepared under different conditions is essential for further improvement of any device based on this ferromagnetic alloy. Still, there are only a few reports on the electrical transport properties of CoFeB thin films including composites and nanotubes [7–13]. The electrical resistivity of a crystalline film is significantly lower as compared to that of an amorphous film [11, 12]. While the crystalline films show a metallic behaviour, negative temperature coefficient of resistance has been reported for the amorphous or composite films [8, 12]. Thus, the crystalline quality plays an important role in the process of electrical conduction of these films. Moreover, the thickness of these films also controls their electrical properties as the size effects become dominant at lower thicknesses. For example, Jen et al have reported an increase of resistivity in the amorphous CoFeB film on lowering of the thickness [9]. However, a thorough analysis of electrical transport in crystalline CoFeB films is lacking.

An interesting feature of CoFeB systems is the presence of resistance upturn at lower temperatures [7, 12–14]. However, there is no clear consensus on its origin due to the differences in experimental results and theoretical interpretations. Fujimori et al have reported a temperature dependent resistivity in the upturn region [7], which is expected for a variable range hopping conductivity in Coulomb gap [15]. A logarithmic temperature dependent resistivity has been observed in crystalline CoFeB film [12] and nanotube [13] as well as amorphous ribbons [14], which can originate due to one or more of these three effects; Kondo scattering [16] or Coulomb effect in granular materials [17] or tunneling model [18]. The resistance minimum in a variety of amorphous ferromagnetic alloys similar to CoFeB has been attributed to Kondo type lnT resistance [19–22] and, in some alloys, an additional minimum due to r^{1/2} resistance has been

Resistance minimum and electrical conduction mechanism in polycrystalline CoFeB thin films

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Received 10 May 2015, revised 9 September 2015
Accepted for publication 15 September 2015
Published 26 October 2015

Abstract

The temperature dependent resistance R(T) of polycrystalline ferromagnetic CoFeB thin films of varying thicknesses are analyzed considering various electrical scattering processes. We observe a resistance minimum in R(T) curves below ≃29 K, which can be explained as an effect of the intergranular Coulomb interaction in a granular system. The structural and Coulomb interaction related scattering processes contribute more as the film thickness decreases implying the role of disorder and granularity. Although the magnetic contribution to the resistance is the weakest compared to these two, it is the only thickness independent process. On the contrary, the negative coefficient of resistance can be explained by the electron interaction effect in disordered amorphous films.

Keywords: ferromagnetic alloy, thin film, spintronics material

(Some figures may appear in colour only in the online journal)
observed [23]. Moreover, in a strong ferromagnetic material like CoFeB, the role of magnon in electronic transport is quite important apart from other scattering mechanisms involving electron, phonon, lattice potentials due to structure etc. For example, a finite magnetic contribution to the resistivity due to electron-magnon scattering along with the structural contribution has been observed in amorphous ferromagnetic alloys like FeBC and FeBGe [24, 25]. All these mechanisms mostly result in various power law temperature dependence of resistivity. In order to shed some light on these aspects, we have analyzed the temperature dependent electrical transport in polycrystalline Co₄₀Fe₄₀B₂₀ thin films of varying thickness in detail. We have mainly focussed on the origin of resistance minimum observed below ≈29 K in the light of various scattering mechanisms involved in the electrical conduction.

The thin films of Co₄₀Fe₄₀B₂₀ with nominal thicknesses ranging from 5 to 40 nm have been deposited on SiO₂(300 nm)/Si(1 0 0) substrates using KrF excimer (λ ≈ 248 nm) based pulsed laser deposition technique. The growth of these films has been performed at room temperature under argon pressure of 2 × 10⁻³ mbar with a typical growth rate of 0.04 nm s⁻¹. To enhance the crystallinity, the films have been subsequently annealed at 400 °C for 1 h under high vacuum. The details of film growth have been reported previously [12]. Figure 1 shows the grazing angle x-ray diffraction scans for various CoFeB films. We have determined the actual thickness of the films from the fits to XRR data as shown in figure 1(b). The extracted thickness (t) of the films are 6, 9, 20, and 36 nm. The high angle scan profiles show the polycrystalline nature of the films (see figure 1(a)). As the film thickness increases, the intensity of (1 1 0) CoFe peak increases and it becomes much sharper as shown in figure 1(c), indicating better crystalline quality and the presence of larger grains in thicker films. While the fits to XRR data provide an estimation of interface roughness values as ≲1 nm, the surface roughness obtained from atomic force microscopy (AFM) scans is ≈1.5 nm. Moreover, the AFM images reveal the granular nature of these films as shown in figure 2(a). The grain size decreases with decreasing film thickness. A similar trend can be observed from the average grain size (L) determined by the Scherrer formula (see figure 2(b)). We have previously reported the presence of grains with similar sizes as observed by transmission electron microscopy imaging of CoFeB films [12]. This variation of grain size with film thickness has significant effects on magnetic and transport properties as we will discuss now.

Figures 3(a)–(d) display the room temperature magnetic hysteresis loops of all films measured for both in-plane and out-of-plane fields. Clearly, we observe low saturation fields and a high squareness ratio for in-plane loops. While the in-plane squareness is greater than 0.7, the out-of-plane squareness is less than 0.05. These observations indicate that the magnetic easy axis is in the film plane. The saturation magnetization of CoFeB can be determined from the linear fit to the thickness dependent saturation magnet moment (Mₛ). The extracted value of saturation magnetization comes out to be 1580 emu cc⁻¹. Furthermore, we observe a magnetic dead layer of 0.8 nm, which is close to the values of dead layer observed in other CoFeB interfaces [3]. Figure 3(f) presents the coercivity (Hettings) of the easy axis loop as a function of d, which clearly shows the enhancement of Hₐ with increasing d. Similar behaviour observed in various ferromagnetic alloys has been attributed to the grain size effect [26–28]. With decreasing grain size, the effective anisotropy constant reduces, which leads to an increase in the effective exchange range [28]. We have also observed such reduction in anisotropy with decreasing thickness (or decreasing grain size) [29]. Under such scenario, the exchange-coupling between the grains is enhanced and, thus, the Hₐ reduces with decreasing thickness.

Now, we present the electrical transport study of our films (see figure 4(a)). The temperature dependent four-probe sheet resistance Rₛ(T) shows the metallic behaviour for all CoFeB films. The resistance decreases with lowering of the temperature up to a certain temperature, known as resistance minimum temperature (Tmin), and then it starts increasing down to 5 K. The Tmin for the 36 nm thick film is 26 K and it gradually increases with decreasing thickness (see figure 4(b)). The thickness dependence of sheet conductance Gₛ = 1/Rₛ at T = 273 K shows a linear thickness dependence as shown in figure 4(c). The slope of the linear fit yields a resistivity of (33 ± 2) μΩ cm, which is close to previously reported values for several micron thick amorphous CoFeB ribbons [14]. However, one can observe a finite intercept of the linear fit on the temperature axis, which suggests the presence of an electrically less conducting dead layer. The existence of such dead layers has been reported in thin films of manganite [30] and Heusler alloy [31]. This dead layer can form on the surface and at film-substrate interface due to surface oxidation, roughness effect, and boron segregation at the interface [9, 12]. Alternatively, the presence of larger number of grain boundaries in thinner films can also result in such behavior. The thickness of the dead layer is given by the intercept on the thickness axis, which comes out as tₛ ≈ 2.5 nm.
Coming back to $R_T$ data, we have fitted these using the following expressions. For the low temperature ($5 \text{ K} \leq T \leq 100 \text{ K}$) regime, the resistance is expressed as:

$$R_T = a_0 + a_2 T^2 + \beta \ln T,$$

and, for high temperature ($100 \text{ K} \leq T \leq 273 \text{ K}$) regime,

$$R_T = b_0 + b_1 T + b_2 T^2,$$

where $a_0$, $a_2$, $\beta$, $b_0$, $b_1$, and $b_2$ are the constants. The temperature independent terms ($a_0$ and $b_0$) represent the residual sheet resistance $R_0(0 \text{ K})$. Using these values, the $t_d$ again comes out as $\approx 2.5 \text{ nm}$ (see figure 4(c)), which implies that the dead layer remains fixed in whole temperature range. The $T^2$ term in above expressions can arise due to the effects like electron-magnon scattering and electron-lattice scattering. The first scattering process is due to the coherent scattering of electrons by long-wavelength magnons in a ferromagnet, which introduces a magnetic contribution ($\rho_{\text{mag}}/t$) to $R_T$. On the other hand, the incoherent scattering can introduce a $T^{3/2}$ term in an amorphous ferromagnet while such process is absent in a crystalline ferromagnet [24]. Therefore, we do not observe a $T^{3/2}$ dependency in our fits. The latter process involves the scattering of conduction electrons from the lattice potential of a transition-metal system, which results in a structural contribution ($\rho_{\text{str}}/t$) to $R_T$ [32, 33]. While $\rho_{\text{mag}}$ varies as $T^2$ for all temperatures, $\rho_{\text{str}}$ shows a transition from $T^2$ to linear $T$ dependence as the temperature increases. The last term in equation (1) can arise due to two effects. Firstly, the logarithm dependence of resistance is a characteristic feature of Kondo scattering observed in dilute magnetic metals, quantum dots, and heavy electron systems [16]. The Kondo behavior can be explained as the interaction of conduction electrons with...
localized spins of magnetic impurities. Such effect can also be observed in ferromagnetic materials, where the effective field distribution for a magnetic spin has a long tail extended below zero field and, thus, some of the spins participate in spin flipping scattering process [34]. One should note that this effective field model is only valid for amorphous systems while, in crystalline ferromagnet, distinct field lines are observed in place of a field distribution [35]. Also, the absence of resistance saturation (or tendency for saturation) in many reports does not provide a solid evidence of Kondo effect [14, 21, 22]. Thus, for our metallic crystalline ferromagnetic CoFeB thin films, the presence of Kondo effect well below its Curie temperature ($T_c$) is highly unlikely. We will provide further support to our conjecture later on.

The second possible explanation is the Coulomb interaction in a granular metal, where a logarithmic temperature dependent conductivity can be observed in metallic regime with dimensionless tunneling conductance $g \gg 1$ [17, 36]. To verify if this scenario is valid in our case, we have determined $g$. In the temperature range of $5 \, \text{K} \leq T \leq 100 \, \text{K}$, the maximum relative change in resistance, i.e. $|R_c(T)/R_c(10 \, \text{K})| - 1$ as-deposited and as-annealed 40 nm films. The curves are fitted to the expression: $R_c(T) = a_0 + a_1 T^{1/2} + a_2 T^2 + \beta \ln T$. The fitting parameters $a_{1/2}$ and $-\beta$ are plotted in panel (b).

Comparing this expression with the logarithmic term in equation (1), we have $\beta = -(\hbar/2\pi e^2)(L/d^2) g^{-2}$. Figure 5 shows the estimated values of $g$ for $d = 2$ and 3 using the fitting parameter $\beta$. Clearly, $g \gg 1$ for all the films, which indicates that the conduction is not a tunneling process through an
increases with decreasing contribution coming due to inter-granular and \(2^\text{nd} \), where \(\dots\). Figure 7(a) shows that values are quite close to the values obtained \(\frac{\text{uni}}{2}\). The 100 K, comes out as \(\approx 9.0\), which indicates the e–e interaction is still a highly dominant process for the 300 °C annealed film. Olivier et al have reported the presence of both \(\text{ln} T \) and \(T^{1/2}\) dependent resistance in FeCrB metallic glasses, where the \(T^{1/2}\) term is responsible for the resistance minimum in some cases [23]. Thus, the presence of negative TCR in all three samples (albeit in different temperature regimes) has two different origins; i.e. e–e interactions in a disordered amorphous film and Coulomb effect in the granular polycrystalline film.

In order to find out the significance of quadratic temperature dependent resistance, we have converted the coefficients \(a_2 \) and \(b_2 \) in the form of Resistivities, i.e. \(a_2' = a_2\sqrt{\text{mag}}\) and \(b_2' = b_2\sqrt{\text{mag}}\). The \(b_2' T^2\) term represents the \(\rho_{\text{mag}}\) while the \(a_2' T^2\) term is a combination of \(\rho_{\text{mag}}\) and \(\rho_{\text{str}}\). Figure 7(a) shows that \(b_2'\) (or \(\rho_{\text{mag}}\)) remains almost constant irrespective of \(t\). Such behaviour implies that all films are magnetically homogeneous, which can also be confirmed from magnetic hysteresis loop measurements shown in figure 3. Moreover, the disorder present in thinner films only introduce a minor correction to the magnetic \(T^2\) term and, thus, do not significantly alter \(\rho_{\text{mag}}\) [24]. The \(b_2'\) values are quite close to the values obtained for other ferromagnetic metals (2.2–3.2 × \(10^{-11}\) Ω cm K\(^{-2}\) for Fe and Co) and alloys (0.98 × \(10^{-11}\) Ω cm K\(^{-2}\) for Fe_{90}B_{20}) [24, 40, 41]. The \(\rho_{\text{mag}}\) is proportional to \((J_{sd}/D)^2\), where \(J_{sd}\) is the \(s–d\) exchange integral and \(D\) is the spin-wave stiffness constant [24]. We have estimated the values of \(D\) from temperature dependent magnetization \(M(T)\) data as shown in the inset of figure 7(a). The \(M(T)\) in the low temperature regime is given by Bloch law as follows: \(M(T) = M(0)(1 – BT^{3/2})\), where \(B = [\zeta(3/2) g_e \mu_B M(0)] / (k_B / 4\pi D)^{3/2}\) [42]. Here, \(\zeta(3/2) = 2.612\) is the Riemann \(\zeta\) function, \(g_e = 2\) is the

![Figure 7](image-url)
gyromagnetic ratio, and \( \rho_{\text{B}} \) is the Bohr magneton. Using these expressions, the \( D \) comes out as 88 and 185 meV Å\(^{-2}\) for the 9 and 36 nm thick films, respectively, which are comparable to the reported values for various FeB based alloys [42]. Since the \( \rho_{\text{mag}} \) (and thereby \( |J_{\text{sd}}|D^2 \)) is thickness independent, \( |J_{\text{sd}}| \) (\( t = 9 \) nm) \(< |J_{\text{sd}}| \) (\( t = 36 \) nm). Here, we want to point out that the values of \( |J_{\text{sd}}| \) are negative. In a Kondo picture, \( \beta t \propto J_{\text{sd}} \), which implies that \(-\beta t \) (\( t = 9 \) nm) \(< -\beta t \) (\( t = 36 \) nm). But an opposite trend observed in our case (see figure 5) discards the Kondo effect as the explanation of the resistance minimum. On the other hand, the low temperature \( T^2 \) coefficient \( \alpha_2 \) is much larger than \( \alpha_2 \) and increases with reducing \( t \), which suggests an additional contribution apart from thickness independent \( \alpha_2 \). This extra contribution corresponds to \( \rho_{\text{an}} \) and it gets enhanced in thinner films due to the presence of more structural disorders and grain boundaries. Comparing these two effects, we observe a substantial magnetic contribution although \( \rho_{\text{an}} \) is at least 3.5 times more dominating in comparison to \( \rho_{\text{mag}} \).

Apart from the \( T^2 \) term, the \( R_{\text{sd}}(T) \) expressions involve two other important terms; i.e. \( \beta \ln T \) (in low \( T \) regime) and \( b_1 T \) (in high \( T \) regime). We have defined the relative weight of these terms with respect to corresponding \( T^2 \) terms as follows: \( W(\beta,a_2) = \int \beta \ln T dT / \int a_2 T^2 dT \) and \( W(b_1,b_2) = \int b_1 T dT / \int b_2 T^2 dT \) with the integrations performed using the limits \( 5 K \leq T \leq 100 \) K and \( 100 K \leq T \leq 273 \) K, respectively. Figure 7(b) shows these relative weights as a function of \( t \). Clearly, the Coulomb effect plays a dominating role as compared to the \( T^2 \) term at lower temperatures as \( W(\beta,a_2) > 1 \). Moreover, the increase of \( W(\beta,a_2) \) with decreasing thickness suggests that this effect becomes rather important as the grain size reduces and the grain boundary contribution increases. In the high temperature regime, \( \rho_{\text{an}} \sim T \) and the \( W(b_1,b_2) \) basically represents the relative weight of \( \rho_{\text{an}} \) with respect to \( \rho_{\text{mag}} \). Thus, the dominating effect of linear \( T \) term and its enhancement with decreasing \( t \) are well expected as explained before. While the electron–phonon (e–p) scattering also shows a linear \( T \) dependency above Bloch–Grunenisen temperature (\( T_{\text{BG}} \)), it should reduce with increasing grain boundary and, thus, with decreasing thickness [43]. This behaviour is opposite to the trend seen in figure 7(b). Moreover, the e–p scattering in low \( T \) regime (\( T \ll T_{\text{BG}} \)) results in a \( T^5 \) dependent resistance term. We have not observed any improvement to the fits by additional introduction of this term in equation (1). Therefore, one can safely assume that the electron–phonon scattering does not play an important role in electronic conduction.

In conclusion, we have investigated pulsed laser deposited polycrystalline CoFeB films of varying thickness. The analysis of transport measurements on CoFeB thin films demonstrates the effect of granularity and disorder on the structural, magnetic, and Coulomb interaction related scattering processes. The resistance minimum is related to the grains (and the grain boundaries) present in the film. While the magnetic contribution to resistance remains independent of the film thickness, the structural contribution and inter-granular Coulomb effect increases with decreasing thickness. In amorphous films, the electrical conduction is mainly dominated by the electron interaction effects in disordered systems. Our comprehensive study of electronic transport in CoFeB films shows that the electrical conduction in thinner films will be affected by granularity (or Coulomb charging effects). Therefore, better performance of spintronics devices can be achieved either by reducing device size to the order of grain size or by developing a better fabrication technique for thin films with large grains.

Acknowledgments

We thank R C Budhani for his valuable comments, K K Maurya for XRD measurements, and V Toutam for AFM measurements. We acknowledge Council of Scientific and Industrial Research (CSIR) & Department of Science and Technology (DST) for financial support.

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