ENHANCEMENT OF COERCIVITY INDUCED BY FILM MORPHOLOGY CHANGES IN Co/Cu MULTILAYERS.

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1. Introduction

Earlier studies [1,2] in giant-magnetoresistance (GMR) Co/Cu multilayers (MLs), that exhibit low-hysteresis at about 290 K, have shown a dramatic increase of hysteresis below 100 K. These results imply that, while the activation energy for magnetization reversal is reduced by a decrease of Co layer thickness [1] (t_{Co}) or by Co-Cu alloying [2], it remains large relative to thermal energy at low temperatures. So far the properties of the micromagnetic state, that reduce hysteresis in the GMR loops at room temperature, are unknown.

The aim of the present study is to investigate the micromagnetic properties that result in low-field GMR by measuring the temperature dependence of isothermal magnetic and GMR loops in three classes of $[Co(1\ nm)/Cu(2.1\ nm)]_{30}$ MLs between 5 and 300 K. Such macroscopic measurements include combined magneto-structural information. However, micromagnetic parameters such as lateral magnetic correlation length and roughness vary with temperature and are totally different [3] from the corresponding microstructural parameters which remain unaltered in the examined temperature range. A previous study [4] has shown that these three classes exhibit different GMR ratios, hysteresis, and saturation fields at room temperature as the Co-Cu layering is modified by the deposition conditions. Since Co/Cu interface roughness affects primarily the interlayer exchange coupling whereas the grain size distribution and the density of grain boundaries alter the magnetostatic energy then the temperature dependence of GMR and magnetic hysteresis loops can separate the major contribution from each one of the two structural characteristics.

2. Structural Characterization

Three classes of GMR Co(1 nm)/Cu(2.1 nm) MLs were sputter grown with different microstructures in respect to grain size and interface roughness, depending on deposition conditions [4]. Two parameters were varied to produce the three classes (named *A*, *B*, *C*) of Co/Cu MLs: (i) the surface roughness of the Si(100) substrate, affecting the mode of growth; (ii) the thermal contact of the substrate with the, water cooled, supporting table that influence internal film stress. Transmission electron

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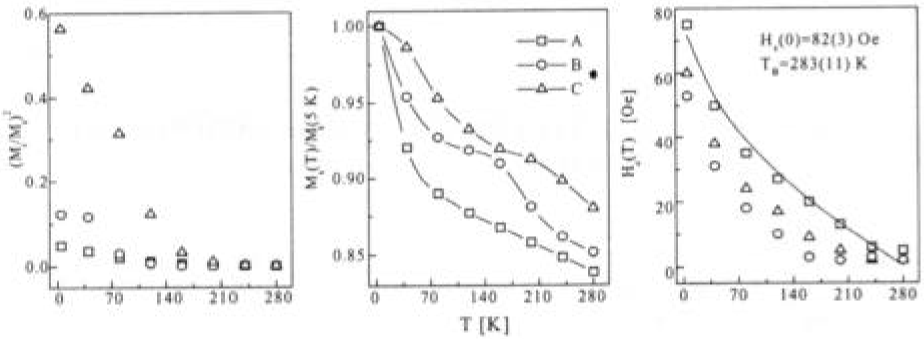

Figure 1. The plot on the right shows the temperature dependence of the coercive H_c fields, obtained from isothermal M/M_s-H loops of class *A* (squares), *B* (circles), and *C* (triangles) MLs. The solid line is the best fit using the $H_c(T)=H_c(0)[1-(T/T_B)^{1/2}]$ equation. The middle plot shows the temperature dependence of the normalized magnetization (the lines are guides to the eye) and the plot on the left shows the temperature dependence of the square of the normalized residual magnetization M_r/M_s.

microscopy (TEM) measurements [4] have revealed column-like structures with bimodal distribution of grain sizes. A 90% fraction of columnar grains with sizes more than 15 nm is observed in the sample called *A*, for sample *B* it is 70%, and in sample *C* is less than 50%. Thus a larger fraction of grains with sizes less than 10 nm appears progressively from sample *A* to *C*.

Atomic force microscopy measurements [4] on the substrate surface prior deposition and on the film surface revealed that class *A* MLs have atomically smooth substrate-film interfaces whereas both surfaces exhibit a long-range waviness with average periodicity of about 100 nm. A detailed x-ray reflectivity (XRR) study of class *B* MLs has shown [5] that both, Co/Cu interface and lateral correlation function $C(\xi)$ roughness are governing their layer morphology, while TEM measurements [4] show that class *C* MLs exhibit a large geometrical and chemical (Co-Cu mixing) roughness. Thus, increase of film roughness from class *A* to *C* MLs is an unavoidable result of the larger fraction of small grain sizes that changes the overall film morphology. However, increase of Co/Cu interface roughness weakens the magnitude of interlayer exchange coupling within each columnar structure whereas a larger fraction of small grains makes the magnetostatic contribution an important dipolar energy term in the total magnetic free energy that determines the micromagnetic state of the film.

3. Magnetic and magnetotransport measurements

Isothermal magnetization loops were measured between 5 and 280 K with a Quantum-Design-MPMSR2 SQUID magnetometer. The temperature dependence of the normalized magnetization (M/M_s-H) loops reveals a large increase of coercivity (H_c) and saturation (H_s) fields below 120 K. The normalized $M_s(T)/M_s(5K)$, $(M_r(T)/M_s)^2$ and H_c values are plotted in figure 1 as a function of temperature for the three classes of MLs. Since the observed [4] variation in the bimodal distribution of grain sizes affects the short-range exchange, the long-range dipolar interactions and the anisotropy energy as we move from class *A* to *C* MLs, it may accounts for the different increase of H_c and remnant magnetization M_r at lower temperatures as well. The most interesting result is revealed in the temperature dependence of magnetization, which for the three classes of MLs does not follow the linear T, or $T^{3/2}$, or T^2 power laws. These laws were derived [6] for the cases of non-coupling, of FM and AF interlayer coupling, respectively, and observed [7,8] in fcc Co/Cu MLs at the first AF maximum, where the interlayer coupling term is dominant. The observed disagreement with these power laws in fig.1 is caused by the significant increase of M_s below 120 K. An enhancement of M_s induce a significant increase of the magnetostatic contributions in the long-range dipolar energy term which has not been taken into account in the minimization of the intrinsic magnetic free energy that leads to the specific power laws. Therefore, the intrinsic enhancement of M_s by decreasing temperature creates an extrinsic increase of magnetostatic dipolar interactions due to geometrical grain factors introduced by the specific microstructure. This extrinsic magnetostatic energy gives rise to a blocking temperature accompanied by significant increase of hysteresis below 120 K. Application of the same model as for magnetic nano-particles can approximate the temperature depedence of $H_c(T)$ by: $H_c(T)=H_c(0)[1-(T/T_B)^{1/2}]$, where T_B defines a blocking temperature above which domain walls can not be stabilized within the Co layers and the $H_c(T\geq T_B)=0$. This equation is used to fit the observed $H_c(T)$ values of sample *A* and the solid line in figure 1(right plot) gives an $H_c(0)=82(3)$ Oe and a $T_B=283(11)$ K. As expected, this equation can not fit the observed $H_c(T)$ values in class *B* and *C* MLs because a fraction of relatively small, FM-coupled, Co/Cu grains (not domain walls) co-exists [4] with the larger, AF-coupled, columnar structures and the above approximations are not longer valid.

The GMR measurements were performed with the four-point-probe method using a *dc* current of 10 mA. All measurements were performed by first applying the maximum positive field H parallel to current flow direction and then completing the loop. Figure 2 shows that the GMR ratios $\Delta R/R_s=(R_{max}-R_s)/Rs$, with R_{max} the maximum and R_s the minimum resistance at H_{peak} and H_s magnetic fields respectively, follow a quasilinear decrease with increasing temperature for the three classes of MLs. The large reduction of GMR effect -observed among class *A*, *B* and *C* MLs- indicates that modification of the magnetic disorder at the Co-Cu interfaces, due to changes of roughness, alters the amount of spin-dependent scattering events. Usually, such effects result [9] in strong temperature dependence of the interlayer exchange coupling strength and the corresponding H_s in the GMR loops. In accordance, figure 2 shows that the H_s exhibits a drop of more than 50% between 5 and 280 K, that evidences the strong sensitivity of the indirect coupling strength to temperature. However, the most dramatic change occurs in H_{peak} values, where the three classes of MLs exhibit a steep increase below 100 K.

The observed variation of H_s and H_{peak} in figure 2 is comparable with the strong temperature dependence observed on a scale of about 100 K in [9] Co(hcp)/Cu and [10] Co/Ru MLs. Since the H_{peak} values (figure 2) depend primarily on the magnetization reversal process then they may follow the observed temperature variation of H_c (figure 1), that is derived from the isothermal M-H loops. Thus the same equation was used to fit the H_{peak} values observed in class *A* MLs. The solid line in figure 2 is the best fit to

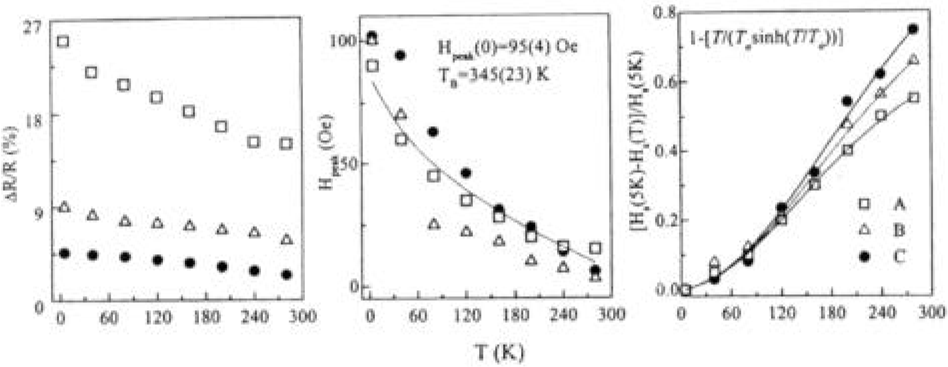

Figure 2. The temperature dependence of the GMR ratios (right) and H_{peak} (middle), obtained from the isothermal GMR loops of class *A* (squares), *B* (triangles) and *C* (solid circles) MLs, are shown. The solid line is the best fit of H_{peak} for class *A* MLs, using the $H_c(T)=H_c(0)[1-(T/T_B)^{1/2}]$ equation. The temperature dependence of the reduced saturation fields H_s, obtained from the isothermal GMR loops, are shown on the right plot for the three classes of Co/Cu MLs. The solid lines are the best fits using the equation shown in the plot.

square symbols, showing that this equation is a good approximation to H_{peak} variation as well.

The most important result is related to the temperature dependence of H_s (fig.2) as the Co-Cu layering is modified in the three classes of Co/Cu MLs. An approximation that describes successfully the temperature dependence of H_s in AF-coupled MLs assumes that the velocity of electrons υ_F at the extremal points of the spacer Fermi surface (k_s) governs the temperature dependence of the oscillatory interlayer exchange coupling strength (J_{osc}) and the one-electron model [11] predicts for the fractional decrease of H_s that: $[H_s(0)-H_s(T)]/H_s(0)\sim 1-[(T/T_0)/\sinh(T/T_0)]$, where the T_0 is a characteristic temperature. Figure 2 shows that this equation fits the fractional decrease of H_s, indicating that the Co/Cu interface roughnes governs the temperature dependence of interlayer exchange coupling in the three classes of Co/Cu MLs. Earlier studies [9,10,11] indicate that T_0 is of the order of 100 K. In agreement, figure 2 shows that this equation fits the fractional decrease of H_s for class *A* (crosses), *B* (triangles) and *C* (solid circles) MLs, using a T_0 of 84(4) K, 96(11) K, and 105(10) K respectively. In Co(hcp)/Cu MLs the existence of either magnetically dead interfacial Co regions, that could modify the thermal evolution of the potential barrier, or fully confined magnetic carries in the spacer potential-well were proposed [9] as possible explanations for the strong temperature dependence of exchange coupling. It has been argued that [9] chemical roughness changes the spin-dependent potential barrier at the interfaces and alters the character of the electronic states nearby the Fermi surface. In agreement, the obtained variation of T_0 can be associated with a larger Co-Cu intermixing (chemical roughness) at the interfaces as we move from class *A* to *C* MLs.

4. Conclusion

This study shows that the obtained differences among class *A*, *B* and *C* MLs are due to different degrees of Co-Cu intermixing at the interfaces and to different geometric factors of the grains. Both are crucial for the obtained $(T/T_0)/\sinh(T/T_0)$ dependence of interlayer coupling because an increase of the fraction of small grain sizes increases the film roughness. However, the obtained increase of T_0 as we move from class *A* to *C* MLs can be understood only if we consider it as a thermal blocking or spin-freezing energy that depends on the concentration of Co loose-spins near the interfaces, rather than as velocity of the carriers [11] at the stationery points of the spacer Fermi surface. Thus, it is the thermal activation energy, which decouples magnetically the residual Co spins at the interfaces due to Co-Cu intermixing, that causes the desired lowering of H_s at room temperature. On the other hand, it is the decrease of M_s from the magnetic decoupling of interfacial Co spins, which lowers the magnetostatic energy at grain boundaries above the T_B, that causes the softening of H_c and H_{peak} values observed in figures 1 and 2 respectivelly. Thus, the degree of H_c and H_{peak} softening scales with the density of grain boundaries (or the fraction of smaller grain sizes) present in the developed microstructure. The experimental results indicate that the spin structure of the examined Co/Cu MLs is not stable above the obtained T_0 or T_B values because the lowering of dipolar (magnetostatic) interactions [12] at grain boundaries can create a secondary short-range order state where domain walls fluctuate infinitely.

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