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MeV ion irradiation of Co/Cu multilayers

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We examine the effect of MeV ion-beam irradiation on the giant magnetoresistance and related interlayer magnetic coupling in sputtered Co/Cu multilayers. At ion doses higher than $10^{13}/\text{cm}^2$, the resistivity of the multilayers increases noticeably, well beyond that measured for pure copper or cobalt films. This increase in resistivity of the multilayers is tentatively ascribed to ion-beam-induced interface disorder. With increasing dose, the magnetic interlayer coupling passes systematically from a mainly antiferromagnetic (AF) coupling to a ferromagnetic one and, in parallel, the giant magnetoresistance (GMR) is progressively destroyed. A linear relationship between the GMR and the volume fraction of AF coupled regions is observed up to an ion dose of $2\times10^{14}/\text{cm}^2$. [S0021-8979(97)71308-2]

INTRODUCTION

Since the discovery of giant magnetoresistance (GMR) in magnetic multilayers, the effects of interfacial roughness on the interlayer magnetic coupling and magnetoresistance (MR) have been of great interest. In Co/Cu multilayers, on the one hand, spin-dependent electron scattering at interfaces was revealed through planar doping.[1] On the other hand, by roughening interfaces with codeposition, Suzuki *et al.*[2] reported that the MR was weakened by interface mixing and it was thus proposed that the scattering centers causing the GMR were in the Co layers. This lack of consistency may partly stem from the difficulty of modifying and controlling the interface structure in a systematic manner. In particular, most *in situ* techniques aimed at modifying interface, for example, varying deposition conditions, are expected to have a significant influence on the growth of the subsequent layers. Further complexities could be involved due to the accumulation of interface roughness from layer to layer, which leads to a gradient of interface roughness across a superlattice.

In this context, *ex situ* approaches of interface modification might have the advantage of avoiding these complexities and should be helpful in clarifying the role of interfaces. In this paper, we report that interface disorder is produced in sputtered Co/Cu multilayers by ion-beam irradiation, with only minor grain growth and texture change being evidenced up to a dose of 1×10^{15} ions/cm^2. Nevertheless, this beam-induced disorder is found to have a large effect on both the antiferromagnetic (AF) interlayer coupling and the magnetotransport properties of the multilayers.

EXPERIMENTAL METHODS

Multilayers with the configuration Cu(50 Å)/[Co(16 Å)/Cu(20 Å)]$_{30}$/Cu(30 Å) were prepared by rf magnetron sputtering onto glass (Corning 7059) substrates, at a sputtering pressure of 6 mTorr of argon gas, starting from a base pressure before sputtering below 1×10^{-7} Torr. The deposition rates, which had been calibrated with a quartz-crystal monitor, were about 2 Å/s for copper and 1 Å/s for cobalt. Nominal thicknesses were confirmed by DEKTAK and low-angle x-ray reflectivity measurements. Sample magnetization was measured at room temperature using a vibrating sample magnetometer. Transport measurements (resistivity and magnetoresistance) were carried out using a high-resolution ac bridge, also at room temperature. The structures of the samples were characterized by low- and high-angle x-ray diffraction using Cu $K\alpha$ radiation with the scattering vector perpendicular to the film surface.

Ion-beam irradiation experiments were performed in a vacuum of 10^{-7} Torr with 1 MeV Si^+ ions using the Université de Montréal Tandem accelerator. To avoid heating effects during irradiation, the samples were placed in thermal contact with a copper block kept at the liquid–nitrogen temperature (77 K) and the beam current was maintained below 50 nA/cm^2. Irradiation doses ranged from 10^{12} to 10^{15} ions/cm^2. The energy loss of the 1 MeV Si^+ ions in the 1000 Å thick samples amounts to roughly 200 keV; only a very small fraction (<0.1%) of the implanted ions come to rest in the multilayer, the rest being transmitted or backscattered.[3]

RESULTS AND DISCUSSIONS

For ion-beam doses up to 10^{13} ions/cm^2, no changes in resistivity, MR, or magnetization are observed. Figure 1 indicates that the saturation resistivity (ρ_s) of a multilayer increases noticeably as a function of total ion dose up to 10^{15} ions/cm^2; also shown are the corresponding variations of the resistivities of 1000 Å Cu and Co films. Here we concentrate on ρ_s to eliminate the GMR contribution. For these doses, the resistivities of both pure films are nearly unchanged upon irradiation which rules out the possibility that the increase in resistivity of the multilayer is associated with bulk defects produced by the ion beam. For our multilayers, the dpa (displacements per atom) at a dose of 10^{14} ions/cm^2 is as small as 0.06, so that the effect on the bulk resistivity can be well neglected for all but the highest doses. Since in these metallic multilayers, the electron mean free path is comparable to the layer thickness, their resistivities can provide valuable information on the interface electron scattering and interfacial structures. We therefore suggest that the present large

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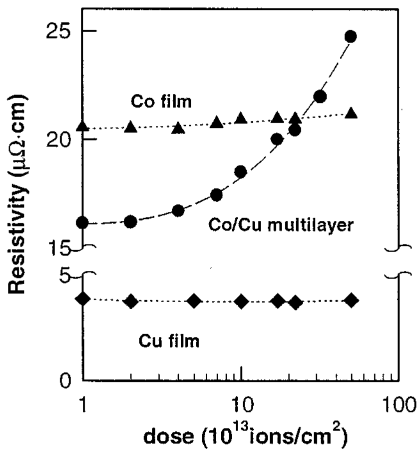

FIG. 1. Resistivities of a [Co(15 Å)/Cu(20 Å)]$_{30}$ multilayer as well as of 1000 Å Cu and Co films as functions of the ion dose. The dashed line is a guide to the eye.

increase of resistivity in the Co/Cu multilayers is connected with interface disorder induced by ion-beam irradiation. In particular, ion-beam mixing through ballistic effects has been confirmed in many bilayers/multilayers systems, including systems with large positive heats of mixing.[4] Moreover, we have also obtained direct evidence of ion-beam-induced interface disorder in our samples through low-angle x-ray reflectivity measurements.[5]

Variations of the magnetic properties of the same multilayer are also observed as a function of ion dose. Figure 2 details the increase in the remanence ratio (M_r/M_s) (where M_r and M_s are the remanence and saturation magnetization, respectively) and the decrease in saturation magnetic field (H_s/H_{so}). These parameters are usually related to the degree of AF coupling between magnetic layers. Initially, most of the layers are AF coupled, although about 30% of the sample is still found to be ferromagnetically coupled. This incomplete AF coupling has been found to be characteristic of Co/Cu multilayers sputtered onto copper buffer layers and

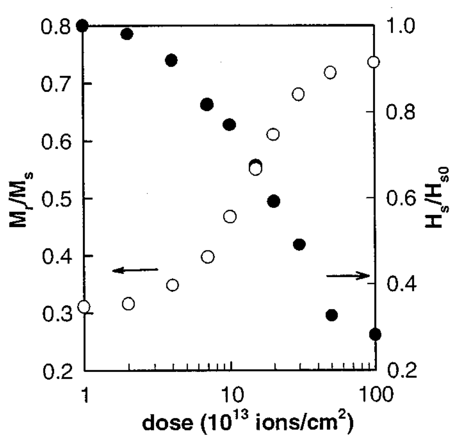

FIG. 2. The remanent magnetization ratio (M_r/M_s), left scale, and the normalized saturation magnetic field (H_s/H_{so}), right scale, for a [Co(15 Å)/Cu(20 Å)]$_{30}$ multilayer as functions of the ion dose. H_{so} is the saturation field of the as-deposited multilayer.

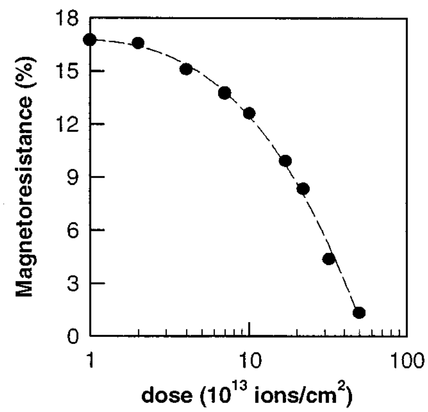

FIG. 3. MR ratio of a [Co(15 Å)/Cu(20 Å)]$_{30}$ multilayer as a function of the ion dose. The dashed line is a guide to the eye.

can be ascribed to interface roughness within the films.[6] This point has been confirmed recently by Kudrnovský *et al.*[7] through a first-principle calculation, which concludes that a small amount of interface imperfection (roughness or interdiffusion) may dramatically suppress AF interlayer coupling. In a somewhat naive picture, interface roughness gives rise to local fluctuations of the spacer (Cu) layer thickness. As magnetic coupling is extremely sensitive to this thickness, interface roughness may result in partial ferromagnetic coupling.[6] Taking $(1-M_r/M_s)$ as an estimate of the volume fraction of AF coupled regions[6] and H_s as indicative of the AF coupling strength, the current results suggest that both the net AF coupled fraction and the AF coupling strength are systematically reduced by irradiation.

High-angle x-ray diffraction after each irradiation reveals little change in crystallographic texture. The as-deposited sample is textured principally in the fcc (111) direction with a relatively weak fcc (200) component. Ion irradiation leads to subtle reductions in the linewidths of both (111) and (200) peaks, indicating minor grain growth and a possible slight increase in the number of (200)-oriented grains. However, these crystallographic changes cannot explain the increasing resistivity and the decreasing AF coupling with increasing ion dose: first, larger grain size should decrease, rather than increase, the bulk resistivity; second, previous studies show that, in sputtered Co/Cu multilayers, the AF coupling is weakened as the grains are textured in the (111) direction, and that the presence of (200)-oriented grains enhances AF coupling rather than suppresses it.[8]

Figure 3 shows that the GMR falls rapidly with ion dose. Given the fact that the GMR is interface related, it is a clear indication that these ion doses produce significant interface modification. In Fe/Cr multilayers, Kelly *et al.*[9] found that ion irradiation led to an increase in the GMR in spite of a significantly reduced AF coupling. Such behavior was explained in terms of the enhanced spin-dependent electron scattering at the Fe/Cr interfaces. In contrast, we observe no increase in the MR upon irradiation of our Co/Cu at any dosage level. It is thus difficult for Co/Cu multilayers to separate the relative importance for the GMR of interfacial spin-dependent scattering and interlayer magnetic coupling.

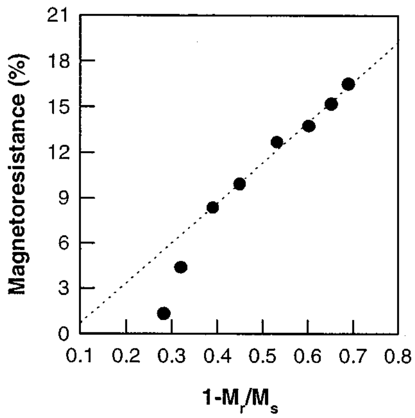

FIG. 4. MR ratio vs $(1-M_r/M_s)$ for the multilayer of Figs. 2 and 3. The dashed line is a linear fit for the data with $(1-M_r/M_s)$ larger than 0.4.

For the samples discussed here, it is interesting to note that the decrease of MR occurs at similar ion doses as the suppression of AF magnetic coupling, as is evident from Figs. 2 and 3, and suggests that the MR and the AF coupling are directly connected. To quantify this point, the MR ratio is drawn as a function of the remanence ratio in Fig. 4. For values of $(1-M_r/M_s)$ from 40% to 70%, a nearly linear relationship is obtained. We obtain here in a *single* sample the same relationship which other groups[10,11] have observed for a series of different samples. This result has also been recently deduced theoretically from a calculation based on an effective medium approximation.[11] Consequently, we conclude that the variation of the MR ratio by irradiation up to a dose of 2×10^{14} ions/cm^2 can be understood to a large extent by a reduced fraction of AF coupled domains rather than by enhanced interface scattering. At higher doses, the origin of the more rapid variation of the MR is still unclear. One possible explanation is that, at these doses, strong mixing of Co and Cu atoms occurs near the interfaces giving rise to strong spin-independent scattering within the intermixed regions, a senario which is consistent with the rapid increase in multilayer resistivity as seen in Fig. 1.

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