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Oscillations in the exchange coupling for (111)-oriented Co/Cu magnetic multilayers grown by molecular-beam epitaxy

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We present experimental evidence for oscillations in the strength of the exchange coupling between Co layers for (111)-oriented Co/Cu magnetic multilayers grown by molecular-beam epitaxy. The evidence comes from an analysis of the approach to saturation of the magnetization data for a series of epitaxial multilayers for which the Cu spacer thickness varies from 5 to 20 Å. We also find that, even for those samples having the maximum exchange coupling strength, only about 20% of the volume of the sample is antiferromagnetically coupled.

The properties of magnetic multilayers have been the subject of intensive research since the discovery of the giant magnetoresistance (GMR) exhibited by these systems and the associated antiferromagnetic coupling between the magnetic layers.[1–4] The exchange coupling between the magnetic layers has been shown to oscillate between antiferromagnetic (AFM) and ferromagnetic (FM) as the thickness of the nonmagnetic spacer is varied. The systems that have been studied most intensively are Fe/Cr and Co/Cu. Both have been shown to exhibit GMR of over 100%. For a long time no evidence for a GMR had been found for (111)-oriented Co/Cu multilayers grown by molecular-beam epitaxy (MBE) as a single crystal, but recently Greig *et al.*[5] reported measurements of a GMR in (111) Co/Cu grown by MBE. However, the largest recorded GMR for these MBE samples is still only 26%. The larger values of over 100% have only been seen in sputtered Co/Cu that has a (111) *texture.* Furthermore, in sputtered Co/Cu and in (100)-oriented Co/Cu grown by MBE, there have been reports of oscillations in the magnetic coupling as the magnetic spacer thickness is varied.[4,6] But there have been no observations of oscillations in the coupling of (111)-oriented Co/Cu grown by epitaxy. Because of this, some workers[4] have argued that the oscillations observed in sputtered Co/Cu with (111) texture should be attributed to (100) inclusions. However, since theory[7,8] predicts such oscillations with a period of about 10 Å, a more plausible explanation is that the lack of oscillations is a growth problem. Indeed the observation[5] of a GMR in Co/Cu multilayers grown by MBE resulted from a change in the growth technique and the use of a Au seed layer.

In this paper we report measurements showing a clear oscillation in the exchange coupling between Co layers for (111)-oriented Co/Cu grown by MBE. Moreover, from the low-field behavior of the magnetization, in those samples that exhibit a GMR, we can conclude that 80% of the sample volume is ferromagnetic. The observed GMR thus results from that 20% which is AFM coupled. Analysis of the high-field behavior of the magnetization shows evidence for such AFM coupling which oscillates as the thickness of the Cu spacer is varied. From the approach to saturation of the magnetization we observe no saturation field. However, the magnetization saturates as $(1-a/H^2)$ as one would expect in the presence of in-plane anisotropy. Indeed there has been some confusion in the literature over the "saturation" field in the Co/Cu system because the magnetoresistance and the magnetization do not appear to saturate at the same field. Of course there is no saturation field when the field is oriented away from an easy or hard axis and then the way the magnetoresistance and magnetization approach saturation will not necessarily be the same. So if one applies similar criteria to the magnetoresistance and magnetization data in deciding an effective "saturation" field, one will not necessarily get the same result. These points will be discussed in a forthcoming paper.[9]

All our Co/Cu multilayers were grown in a VG80M MBE facility in which the base pressure was 3×10^{-11} mbar. Further details of the growth conditions including reflection high-energy electron diffraction and x-ray characterization can be found in Greig *et al.*[5] along with data on the GMR for the present samples.

The Co/Cu samples studied consisted of 20 bilayers, each having a Co thickness of 15 Å and a Cu thickness that varied from 5 to 20 Å. In Figs. 1–3 we show the magnetization data for $t_{Cu}=5$, 9, and 13 Å. All the samples measured showed low-field hysteresis which confirms that there is a large degree of FM coupling in all the samples. It should be noted here that we have previously reported the magnetoresistance and magnetization for the 5 Å sample and noted that the magnetoresistance was "small"—only a few percent compared with 26% in the 7 Å sample—and the remnant magnetization was large; both facts indicate FM coupling.[5] However, we now believe that between 10 and 20% of the magnetization at higher fields shows the presence of both AFM coupling and in-plane anisotropy. For the particular case of the 5 Å sample the magnetization and magnetoresistance approach saturation at around 20 kOe, and the magne-

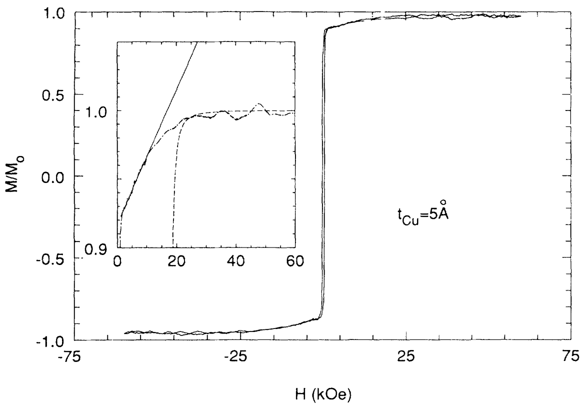

FIG. 1. The magnetization at 4.2 K vs magnetic field for the sample having $t_{Cu}=5$ Å. The inset shows a close-up of the magnetization as M/M_0 approaches unity. The fitted lines are discussed in the text.

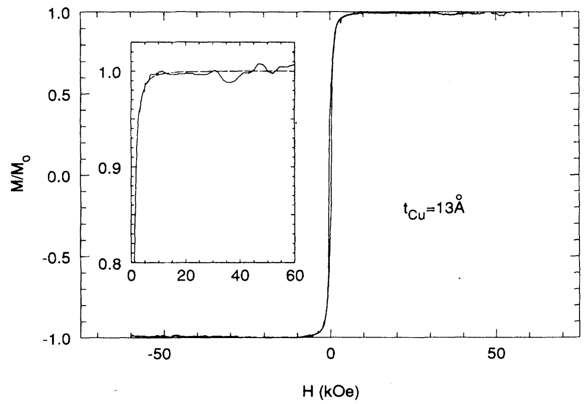

FIG. 3. The magnetization at 4.2 K vs magnetic field for the sample having $t_{Cu}=13$ Å. The inset shows a close-up of the magnetization as M/M_0 approaches unity. The fitted line is discussed in the text.

toresistance is largely independent of the orientation of the field with respect to the current; both facts indicate that the magnetoresistance is the "giant magnetoresistance" and there is AFM coupling.

In the inset to Figs. 1–3 we have plotted the magnetization at high fields. Since the samples exhibit a GMR (Ref. 5), we should expect to see evidence of AFM coupling. AFM coupling in the presence of anisotropy will lead to a linear dependence of M on H with an extrapolated saturation field of $H_{AFM}=2J/M_0$, where J is the strength of the coupling between two adjacent magnetic layers and M_0 is the saturation magnetization. However, at high fields M will not saturate. Instead the magnetization will approach saturation as $M=M_0[1-a/(H-H_{AFM})^2]$ where the coefficient a depends on the anisotropy constant and the angle between the field and some easy axis. We can see from the insets of Figs. 1–3 that if we ignore the low-field hysteresis the behavior of the magnetization is characteristic of strong AFM coupling in the presence of a weaker in-plane anisotropy. We have fitted the magnetization curve to a straight line in the region dominated by the AFM coupling between about 5 and 15 kOe. The intersection of this line with the horizontal line $M=M_0$ gives the field H_{AFM} at which the magnetization would have saturated if there were no in-plane anisotropy. In Fig. 4 we plot the resulting values of H_{AFM} for various copper thicknesses. An oscillation is clearly seen with a period of 12±2Å. This is a slightly longer period than predicted[7,8] but theoretical values of the period are for the large Cu spacer thickness limit so it is not surprising that they do not quite agree. The values of H_{AFM} are zero for the 13 Å and 16 Å samples because, for these samples, there is no region where M varies linearly with H, which suggests that these samples are completely ferromagnetically coupled. This is seen in Fig. 3 for the 13 Å sample where the fitted line is the high-field approach to saturation in the presence of in-plane anisotropy. The absence of a straight-line region at lower fields indicates the presence of FM coupling only.

At high fields we have fitted the magnetization data to

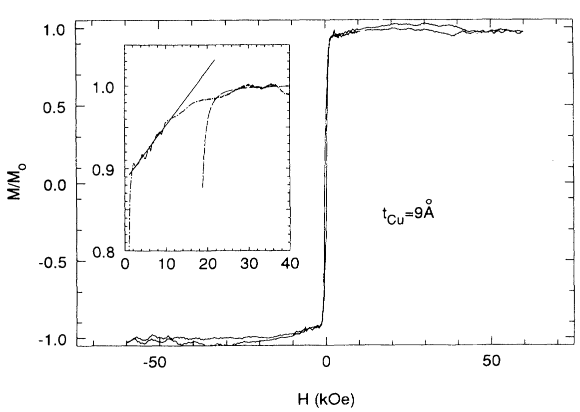

FIG. 2. The magnetization at 4.2 K vs magnetic field for the sample having $t_{Cu}=9$ Å. The inset shows a close-up of the magnetization as M/M_0 approaches unity. The fitted lines are discussed in the text.

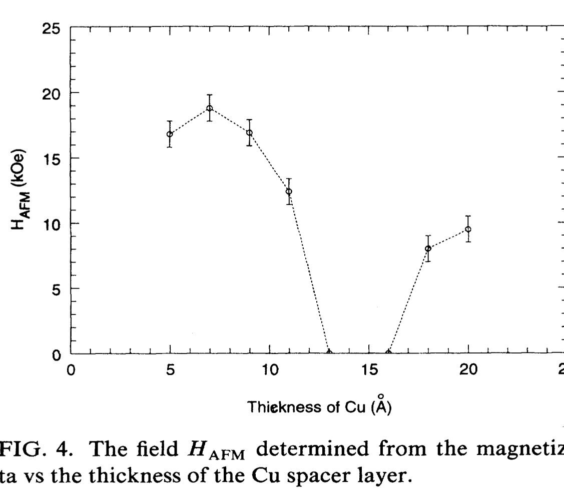

FIG. 4. The field H_{AFM} determined from the magnetization data vs the thickness of the Cu spacer layer.

$M = M_0[1 - a/(H - H_{AFM})^2]$ and determined the constant a. If we assume there is some in-plane anisotropy, the parameter a gives a measure of the in-plane anisotropy in the Co layers. Without carrying out a detailed measurement of the dependence of the anisotropy parameter a on the orientation of the sample in the applied magnetic field we cannot be sure of the exact nature of any in-plane anisotropy. The Co layers are fcc in the (111) orientation[5,10] which has threefold symmetry in the plane. fcc Co has cubic anisotropy with easy axes that give sixfold anisotropy when projected onto the (111) plane, however, we might expect this to be weak. There are other possible sources of in-plane anisotropy. For example, steps in the GaAs (110) substrate, arising from the cutting and polishing of the surface, may produce a uniaxial anisotropy. However, this does not change the overall thrust of our argument that the high-field approach to saturation is due to the presence of AFM coupling and some degree of in-plane anisotropy. As long as the field is not applied along an easy or hard axis the anisotropy constant will be given approximately by $2k/M_0 = \alpha\sqrt{a}$ where α is of order unity. We applied the field in the (112) direction of the GaAs substrate—this is the direction along which the samples easily cleave. The GaAs surface is a (110) plane and so this (112) azimuthal direction for the GaAs is not along any particular Co crystallographic axis but is 23° from one of the Co (110) directions. We have not yet investigated the angular dependence of the magnetization curves and so further measurements of the orientation dependence of a will be necessary to determine what the symmetry of any anisotorpy is.

We find that we can fit the approach to saturation in *all* the samples and get similar values of $2K/M_0 = (1.0 \pm 0.2)$ kOe. The clearest fit is seen in the 13-Å sample where there is no AFM coupling ($H_{AFM} = 0$) and the high-field expression fits the data down to relatively low fields. This value for $2K/M_0$ is quite large but is possibly enhanced due to strain from the lattice mismatch between Co and Cu as was also seen in the Co/Pd multilayers of Engel *et al.* and Heinrich *et al.*[10,11]

It should be emphasized that all the principal features of the magnetization data presented here are consistent with the GMR data we previously reported for these samples.[5] Thus, the sample ($t_{Cu} = 7$ Å) whose GMR has a peak value is also the sample which has a peak in the strength of the AFM coupling, as shown in Fig. 4. Moreover, the large decrease in magnetoresistance takes place as the magnetization increases from about 80% to 100% of saturation—precisely that part of the magnetization curve that we attribute to the 20% of the sample which is AFM coupled. Finally, a calculation[9] of the magnetoresistance of a sample having a 20% volume fraction of AFM-coupled regions embedded in a FM-coupled matrix shows that our observed value of 26% for the maximum GMR is quite reasonable for our samples. These matters are discussed in detail in a forthcoming paper.[9]

In conclusion, we have obtained evidence for AFM coupling in the magnetization of a series of (111)-oriented Co/Cu multilayers grown by MBE. The AFM coupling strength oscillates with a period of about 12 Å, as the Cu thickness is varied. The maximum volume fraction of the multilayers which is AFM coupled is only about 20%. We attribute the absence of a saturation magnetic field to the presence of in-plane anisotropy and the fact that we measure the magnetization with the field away from an easy axis. Finally, we used the data for the approach to saturation of the magnetization at high fields to obtain the value of the in-plane anisotropy constant.

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