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Interfacial quality and giant magnetoresistance in MBE-grown Co/Cu(111) superlattices

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We report ^{59}Co NMR measurements on three MBE-grown Co/Cu(111) multilayer films that show a progression of saturation magnetoresistance from $\Delta R/R=4\%$ to $\Delta R/R=40\%$. Our results demonstrate unambiguously that the increase in magnetoresistance is correlated with an improvement in the quality of the interfaces in these MBE-grown materials.

I. INTRODUCTION

The relationship between the magnetoresistance, magnetization, and structure of MBE-grown multilayers is still far from clear. Recent theories have proposed an enhancement of conduction electron scattering at paramagnetic interfaces, Bartlett *et al.*,[1] and loosely polarized interfacial spins, Slonczewski,[2] as possible mechanisms for creating giant magnetoresistance (GMR). The observation of GMR in sputtered Fe/Cr and Co/Cu of as much as 100% stimulated significant activity. However, all early measurements of magnetoresistance in MBE-grown Co/Cu(111) showed only a small GMR of $\Delta R/R \approx 2\%$. It was therefore suggested that the antiferromagnetic coupling associated with GMR required the (100) texture and atomically corrugated interfaces.[3]

Grieg *et al.*[4] demonstrated that GMR was possible in MBE-grown Co/Cu(111) systems by measuring a saturation magnetoresistance $\Delta R/R$ of 26% in a field of 6 T at 4.2 K for a $[\mathrm{Co}(15\ \text{Å})/\mathrm{Cu}(7\ \text{Å})]_{\times 20}$ multilayer structure. This has since been confirmed by several other workers.[5–7] The quality of the interfaces for a similar sample exhibiting $\Delta R/R=22\%$ was confirmed by ^{59}Co NMR. The NMR spectrum showed nearly all the intensity in two peaks.[8] One of the peaks was characteristic of Co in an environment surrounded by 12 nearest-neighbor Co atoms, the so-called bulk environment, and the other was associated with an environment where three of the Co nearest neighbors had been replaced by Cu atoms, i.e., the ideal interface environment for a (111) oriented multilayer (hereafter called the interface environment). We have now extended the measurements to include a sample with one of the highest reported GMR, for a MBE-grown Co/Cu multilayer, of $\Delta R/R=40\%$, and since this also has the best approximation to the ideal ^{59}Co NMR spectrum it is clear that the quality of the interface and the GMR are strongly correlated.

II. EXPERIMENT

Three Co/Cu multilayers grown at the MBE center at Leeds were examined. The composition of the films is detailed in Table I together with their saturation magnetoresistance measured at 4.2 K. The ^{59}Co NMR spectra were measured in a phase-detected, swept-frequency spin-echo spectrometer[9] with the samples in a circuit, tuned and matched to the 50Ω line. During the course of the experiments only the tuning and matching of the sample coil were changed, so any errors due to mismatches and standing waves in the instrument are identical in all the spectra measured. At each frequency the echo was integrated and measured at phase angles φ and $\varphi+\pi/2$. The phase insensitive quantity $S=\sqrt{S_\varphi^2+S_{\varphi+\pi/2}^2}$ was then determined and plotted against frequency.

In order to correct the spectrum for variations in the spin-spin relaxation time, the spin-echo decay for pulse spacings between 6 and 100 μsec was recorded and extrapolated to zero pulse spacing. This extrapolation is not trivial, particularly near the center of the bulk line, as the decay is not a simple exponential function in this region. The difference in rate of echo decay with pulse spacing at different frequencies is clearly shown in Fig. 1 and reflects the changes in spin-spin relaxation time T_2 due to the

TABLE I. Structure of multilayer films investigated.

Sample	$\Delta R/R$ (%)	Substrate	Structure Buffer (Å)	Multilayer (Å)
66	4	GaAs	Ge500/Co15/Cu200	Co15/Cu7×20
56	22	GaAs	Ge500/Co15/Au10	Co15/Cu7×20
159	40	Sapphire	Nb30Cu20	Co15/Cu9×20

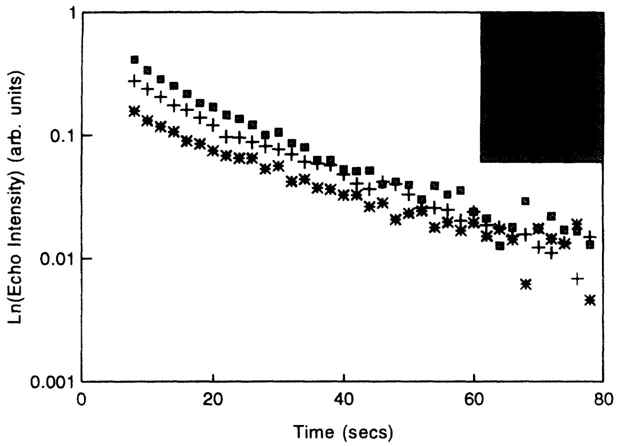

FIG. 1. Variation of the ^{59}Co spin-echo decay time as a function of frequency showing that the spin-spin relaxation time is both not constant over the frequency range investigated and not a simple exponential function.

more rapid spin diffusion near the top of the main peak. In order to correct for this difference of T_2 behavior, the T_2 curves must be measured at regular intervals and fitted to either a single or a more complex exponential function. Our measurements show that close to the peak of the bulk line an equation of the form

$$\text{Echo} = A_0 \exp(2t/T_2^a) + A_1 \exp(2t/T_2^b) \qquad (1)$$

involving both a short and long transverse relaxation time is needed, while away from the bulk peak the behavior is more nearly exponential and can be fitted to a single exponential function. However, it is essential to allow for this short component of T_2^b if the peak is to be correctly defined. Typical values for the characteristic relaxation times close to the main peak are $T_2^a = 80$ μsec and $T_2^b = 20$ μsec with an amplitude ratio $A_1/A_0 \approx 3$. It appears from our measurements that claims such as "the linewidth is reduced by an external field" may arise from the suppression of the short T_2 by the applied field, i.e., the original value of the peak had been underestimated leading to an apparent narrowing at half height in the field.

III. RESULTS AND DISCUSSION

The ^{59}Co NMR spectra for a series of three MBE-grown Co/Cu(111) samples are shown in Fig. 2. The sample shown in Fig. 2(c) simultaneously has one of the largest reported GMR, $\Delta R/R \approx 40\%$ at $T = 4.2$ K, and the nearest approximation to the spectrum of an ideal Co/Cu superlattice material. These spectra should be compared with earlier work which showed the effects of mixing at the Co/Cu interfaces.[10,11] All the present spectra were measured in the frequency range 150–245 MHz and could be fitted by three Gaussian functions [except for Fig. 2(c) where only two Gaussian distributions were required], while in the earlier work up to seven Gaussians were required.

A feature of these spectra that we particularly draw attention to is that the bulk Co line is displaced from the value found in free fcc Co by ~3 MHz, Table II. The NMR frequency of free multidomain fcc Co powder at 4.2 K is 217.4 MHz. The interior of a perfect, thin Co fcc layer would be expected to have a similar NMR frequency, although it might be displaced by lattice strain or the mixing of Co and Cu. The effect of lattice strain can

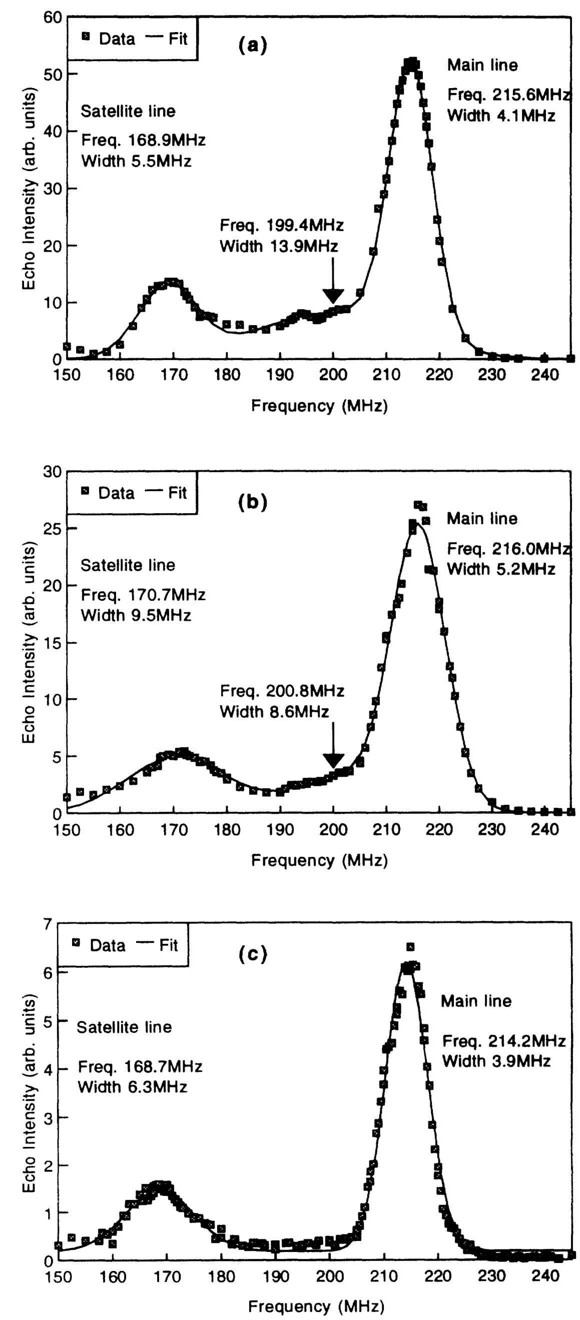

FIG. 2. ^{59}Co NMR spectra corrected for ω^2 and frequency dependence of spin-spin relaxation time for a series of Co/Cu(111) multilayers (a) $\Delta R/R = 4\%$, (b) $\Delta R/R = 22\%$, (c) $\Delta R/R = 40\%$.

TABLE II. The frequency of the main line shown in Fig. 2 for the samples listed in Table I. The shift in the NMR main line is expressed solely in terms of pure volume lattice expansion and equivalent hydrostatic pressure. (The situation must be more complicated since it would be expected that the expansion of the Co lattice in the plane would be accompanied by a contraction normal to the plane.) The final entry shows the effect of expanding the Co lattice spacing to match that of the Cu lattice.

Sample	$\Delta R/R$ (%)	Main line freq. (MHz)	Equivalent pressure (kbar)	Equivalent lattice expansion $\Delta V/V$ (%)
66	4	215.6	-16	0.9
56	22	216.0	-12	0.7
159	40	214.2	-29	1.5
Co$\Longrightarrow$Cu		204.9	-113	5.9

be estimated from high-pressure measurements on fcc Co powder which show that the NMR frequency is increased by 0.106 ± 0.01 MHz/kbar. The Co lattice will be extensively strained in the plane because the lattice constant of Cu is greater than that of Co and will presumably be contracted normal to the interface. Taking the lattice constants for Co and Cu to be 3.54 and 3.61 Å, respectively, it follows that a volume expansion $\Delta V/V=5.9\%$ would occur if the Co lattice were uniformly expanded in three dimensions. This would correspond to a negative pressure of 113 kbar and a frequency shift of -11.9 MHz. In fact, a feature of all three ^{59}Co spectra shown in Fig. 2 is that the main Co line is always at a frequency below the 217.4 MHz of fcc Co powder and is lowest (214.2 MHz) for the material on a Nb substrate that has the largest GMR. Table II illustrates the effect of assuming that the shift can be explained in terms of pure volume strain, assuming that the bulk modulus of hcp Co (= 1.9 Mbar) is similar to that of fcc Co. The shift in the NMR main line is expressed solely in terms of pure volume lattice expansion and equivalent hydrostatic pressure. (The situation must be more complicated since it would be expected that the expansion of the Co lattice in the plane would be accompanied by a contraction normal to the plane.) The final entry shows the effect of expanding the Co lattice spacing, in three dimensions, to match that of the Cu lattice.

A second feature of the spectra shown in Fig. 2 is that the bulk peak for the sample with the largest GMR is both the narrowest and most symmetrical that has been reported. This indicates that the strain is reasonably uniform throughout the sample as suggested by de Gronckel *et al.*[10] although it should be noted that the Gaussian width is still approximately five times the value found in free fcc Co powder.

The origin of the partially resolved line near 200 MHz in Figs. 2(a) and 2(b) is due to Co atoms in a nonideal environment with one or two Cu nearest neighbors. By integrating the intensity of this line and comparing it to the intensities of both the main and interface lines it is found that this line has a significantly greater area than could be accounted for if it arose due to the small buffer layer of bcc Co in these samples. It therefore seems more appropriate to ascribe this line to interfacial mixing and associate it with environments where one or two Co atoms have been replaced by Cu atoms. The frequency is, however, close to that attributed to bcc Co on Fe.[12]

The three distributions all show a clear satellite line at ~ 170 MHz from the Co/Cu interfaces. The frequency of this is again lower than would be predicted for three Cu nearest neighbors in bulk Co/Cu alloys. In the alloys[13] the lines are rather poorly defined, and the same quantitative analysis undertaken in Table II is not possible, but the ^{59}Co NMR frequency appears to decrease by approximately -16 MHz/Cu nearest neighbor. The decrease in frequency for the multilayers corresponds to -15.3 MHz/Cu nearest neighbor. The width of the satellite peaks is similar for the samples with the largest and smallest GMR ($\Delta R/R=4\%$ and $\Delta R/R=40\%$), but significantly greater for the intermediate sample with $\Delta R/R=22\%$. The satellite peak frequency also shows an increase for the film. This suggests that this sample has a modified strain regime at the interfaces.

Integrating the areas under the main and satellite peaks enables the ratio of the number of atoms in the interface environment to the number of atoms in the bulk environment to be determined. Table III shows the results of integrating the areas under the peaks for the three Co/Cu(111) multilayers. These show that the area ratios are close to the ideal values for 15 Å layers of Co, the discrepancies possibly being due to errors in determining the Co layer thickness.

TABLE III. Comparison of ratio of number of atoms in a bulk environment to the number of atoms in an interface environment. The ideal values were obtained by assuming the Co layers were exactly 15 Å thick with a fcc(111) orientation giving a plane spacing of 2.05 Å.

Sample	$\Delta R/R$ (%)	Multilayer (Å)	Bulk/interface ratio Ideal	Measured
66	4	Co15/Cu7×20	2.66	2.80
56	22	Co15/Cu7×20	2.66	2.71
159	40	Co15/Cu9×20	2.66	2.91

IV. CONCLUSIONS

We have demonstrated that Co/Cu(111) multilayers grown by MBE show an increase in magnetoresistance as the interfaces become more abrupt on an atomic scale. The ^{59}Co line at 200 MHz, attributed to interfacial defects, effectively vanishes for the sample exhibiting $\Delta R/R=40\%$. In addition the NMR line of the main peak narrows, suggesting that no significant strain gradient exists within the Co layers. Modeling the shift in main line frequency entirely in terms of volume changes associated with the lattice mismatch between Co and Cu suggests that the Co lattice only undergoes a small expansion.

ACKNOWLEDGMENT

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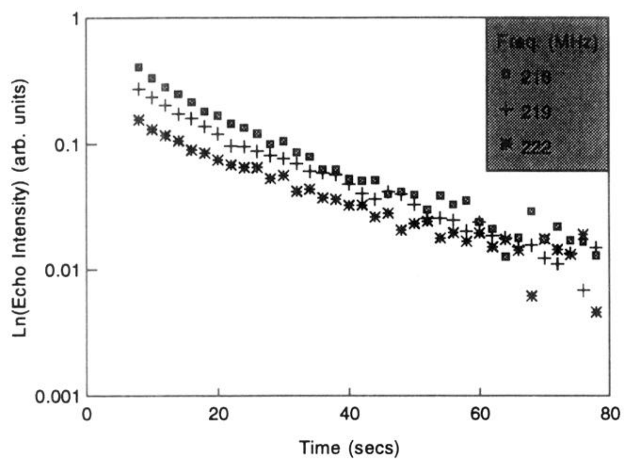

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