Apparent Spin Polarization Decay in Cu-Dusted $Co/Al_2O_3/Co$ Tunnel Junctions

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$Co/Al_2O_3/Co$ magnetic tunnel junctions with an interfacial Cu layer have been investigated with *in situ* growth characterization and *ex situ* magnetotransport measurements. Cu interlayers grown on Co give an approximately exponential decay of the tunneling magnetoresistance with $\xi \approx 0.26$ nm while those grown on Al_2O_3 have a decay length of 0.70 nm. The difference in decay lengths can be explained by different growth morphologies, and in this way clarifies a present disagreement in the literature. For monolayer coverage of Cu, we show that the tunneling spin polarization is suppressed by at least a factor of 2 compared to Co and beyond $\approx$5 ML it becomes vanishingly small.

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Magnetic tunnel junctions (MTJ's), consisting of two ferromagnetic electrodes separated by a thin (<2 nm) insulating barrier, exhibit a large conductance difference between parallel and antiparallel alignments of the two electrodes [1]. Up to now, this effect has been understood within a simple model of Julliere [2], inspired by the ideas of Meservey and Tedrow [3], in which the tunnel current is proportional to the product of the density of states in the left and right ferromagnetic electrodes. This explanation relies, naturally, on the assumption that the electrons tunneling from the ferromagnetic electrodes are spin polarized. Within this model, the tunneling magnetoresistance (TMR), defined as the resistance change normalized by the resistance for parallel magnetizations, is given as $\Delta R/R_p = 2P_1P_2/(1 - P_1P_2)$, where P_1 and P_2 are the tunneling spin polarizations of the two electrodes. Although the Julliere model has been surprisingly successful in explaining the magnitude of the observed TMR, it, however, due to its simplicity cannot explain many observed features of TMR, such as dc bias dependence, or the decay of TMR in the presence of nonmagnetic metallic layers, so-called "dusting" layers. In the latter case, the Julliere model suggests that the presence of a nonmagnetic layer, with no intrinsic spin polarization, would lead to zero TMR. In contrast, the effect of nonmagnetic dusting layers has been investigated both theoretically [4–7] and experimentally [8–10], and in all cases a nonzero TMR is shown in the presence of nonmagnetic dusting layers, indicating an inadequacy in the traditional view of spin polarized tunneling. In this Letter, we experimentally clarify the intrinsic decay of tunneling magnetoresistance in the presence of a nonmagnetic interface layer, using Co/Cu as our model system.

In the first experiment of its kind to involve tunneling, Moodera *et al.* [10] first measured directly the spin polarization in $Al/Al_2O_3/Au/Fe$ junctions as a function of the Au interlayer thickness, finding that polarization decreases rapidly for the first 2 ML Au but decreases as $1/d$ at larger thicknesses. In the context of MTJ's, Parkin investigated TMR as a function of the thickness of a nonferromagnetic layer grown on Al_2O_3 [9]. In these experiments, a large tunneling spin polarization was surprisingly maintained over distances in excess of 10 nm, in striking contrast with the earlier experiments of Moodera for Au on Al_2O_3 [10], as well as later experiments of Sun and Freitas for Cu on Al_2O_3 [8]. As a first attempt to clarify these conflicting results, Zhang and Levy [5] have argued that the behavior of TMR in the presence of an interfacial nonmagnetic layer depends critically on the quality of the interfacial layer, with thickness fluctuations resulting in a much shorter length scale. Mathon and Umerski [7] have recently found that quantum well states in the metallic interlayer are necessary for nonzero TMR, even in the limit of coherence loss. While all work thus far predicts or observes a nonzero TMR, there is little apparent consistency in these results.

In this Letter, we investigate the behavior of TMR in $Co/Cu/Al_2O_3/Co$ and $Co/Al_2O_3/Cu/Co$ systems with a view to elucidate any growth-related effects behind the experimental disparity, using *in situ* scanning tunneling microscopy (STM), Auger electron spectroscopy (AES), x-ray photoelectron spectroscopy (XPS), and *ex situ* magnetotransport measurements. For samples with near layer by layer growth of Cu on Co (underneath the Al_2O_3 barrier), as confirmed by scanning AES, we unambiguously demonstrate for the first time that the decay length of TMR is extremely short ($\approx$0.26 nm), such that 1 ML Cu reduces the observed TMR and tunneling spin polarization by more than 50%, and by about 5 ML Cu are nearly vanishing. In our opinion, this result has implications for the possible mechanisms behind the spin polarization decay. On the other hand, we show that the intrinsic behavior of the TMR and tunneling spin polarization decay length is obscured by nonideal clusterlike growth mechanisms when Cu is deposited on top of the Al_2O_3 barrier, and in this respect, we could clarify the experimental discrepancies in literature.

Ferromagnetic tunnel junctions were prepared by UHV dc/rf magnetron sputtering (base pressure $<5 \times 10^{-10}$ mbar) through metal contact masks onto Si(100)

substrates. *In situ* cleaning in an O_2 plasma was used to effectively remove remaining carbon and water contamination and provide an insulating layer to prevent conduction through the Si substrates. Junction structures of Si/SiO_x/Ta 3.5 nm/Co 8 nm/FeMn 10 nm/Co 5 nm/Al_2O_3/Cu d nm/Co 15 nm/Ta 2 nm ("above" Cu) and Si/SiO_x/Ta 3.5 nm/Co 8 nm/FeMn 10 nm/Co 5 nm/Cu d nm/Al_2O_3/Co 15 nm/Ta 2 nm ("below" Cu) were prepared with junction areas of 300 μm $\times$ 300 μm to 500 μm $\times$ 500 μm, with 24 junctions per sample, 6 of which contained no Cu and thus served as control junctions. The two types of junctions differed only with respect to the placement of the Cu layer, i.e., under or over the Al_2O_3 barrier. The Al_2O_3 layers were formed by plasma oxidation of 2.3 nm Al in 10^{-1} mbar O_2 for 200 sec. Postgrowth annealing at 200 °C in magnetic field for 30 min was used to promote a uniform exchange biasing direction. *In situ* XPS and *ex situ* optical techniques were used to confirm that for control junctions there was no Co oxidation and a minimal amount of leftover metallic Al; the details of these experiments will be published elsewhere [11]. *In situ* STM on homogenous Si/Ta, Si/Ta/Co, and Si/Ta/Co/Al samples indicated flat films with small grains and a mean roughness of <0.3 nm for all layers. Junction resistances and dynamic resistances (dV/dI) were measured using standard ac lock-in techniques, while TMR ($\Delta R/R_p$) was measured using both dc and ac lock-in techniques.

In Fig. 1, TMR as a function of applied field is shown for a control junction, and junctions with 0.42 nm Cu grown above and below the barrier. A clear antiparallel alignment of the Co layers is observed, confirmed by magneto-optical Kerr effect measurements in the junction area. For all junctions reported in this Letter, the lead resistance in the junction area was at least a factor of 5 smaller than the junction resistance to avoid "anomalous" TMR [12]. Control junctions (i.e., no Cu layer) for each sample had an average TMR of 22%–27% at 295 K. Current-voltage (I-V) curves fitted with the expression of Brinkman, Dynes, and Rowell [13] gave estimated barrier heights of 2–3 eV. Fitted barrier parameters as well as junction resistances showed no significant trend with Cu thickness, indicating that the role of the insulating barrier is unchanged by the addition of Cu. In Fig. 2, TMR at 295 K is plotted as a function of inserted Cu layer thickness for junctions with Cu grown below and above the Al_2O_3 layer. The TMR for each Cu thickness is normalized to the value obtained for the average of the six control junctions for that sample. Each point represents at least four different tunnel junctions, while error bars indicate the relative scatter of the TMR for each thickness. As seen in Fig. 2, there is a striking difference in the decay of the TMR vs d_{Cu} for the two types of samples. Fitting the data to an exponential decay gives length scales of 0.26 and 0.70 nm for samples with Cu below and above the Al_2O_3 layer, respectively. Since in this work both types of samples, with the Cu layer inserted above or below the Al_2O_3 barrier, were grown *identically* except for the placement of the Cu layer, some *extrinsic structural difference* must be responsible for the different decay lengths.

The different length scales observed can have a number of possible growth-related origins. The presence of Cu oxides are one obvious possibility, as are the formation of Cu-Al intermetallic compounds. In order to confirm that only pure metallic Cu was present, *in situ* XPS was performed on homogenous samples of Si/Ta 5 nm/Co 5 nm/Al_2O_3, Si/Ta 5 nm/Co 5 nm/Cu 0.5 nm/Al_2O_3, and Si/Ta 5 nm/Co 5 nm/Al_2O_3/Cu 0.5 nm, as well as plasma oxidized Cu (Si/Ta 5 nm/Co 5 nm/Al_2O_3/CuO_x) and thick Cu reference samples. Figure 3(a) shows the Cu $2p_{1/2}$ and Cu $2p_{3/2}$ lines for samples with Cu above and

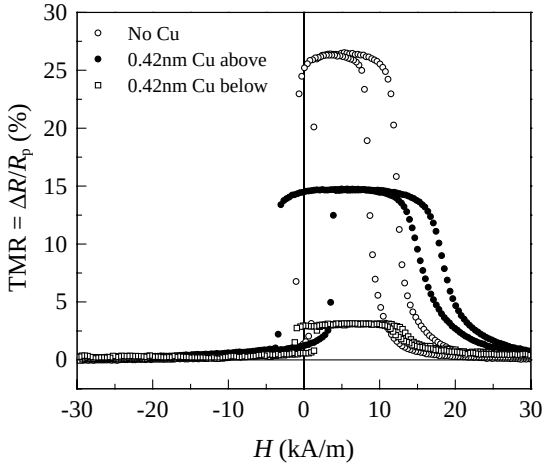

FIG. 1. Tunneling magnetoresistance (TMR) as a function of applied field for junctions with no Cu (open circles), 0.42 nm Cu above the Al_2O_3 barrier (solid circles), and 0.42 nm Cu below the Al_2O_3 barrier (open squares).

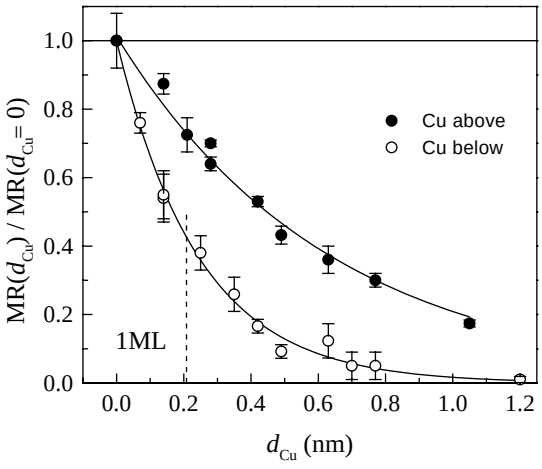

FIG. 2. Normalized TMR as a function of Cu interlayer thickness above (solid circles) and below (open circles) the Al_2O_3 barrier. The solid lines are fits to an exponential decay, while the dashed line indicates 1 ML Cu for a (111) orientation.

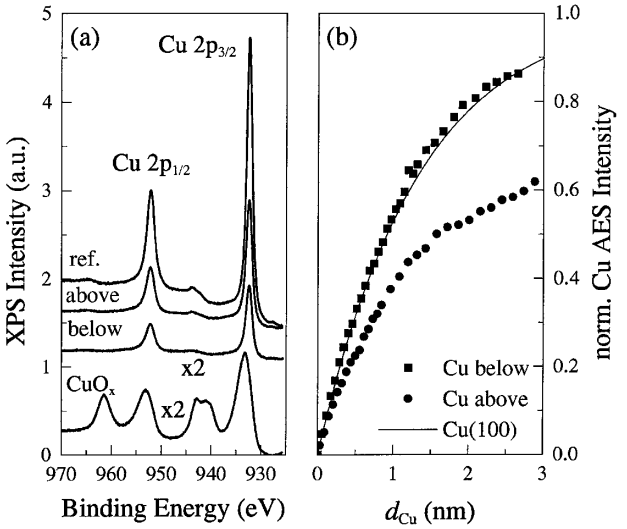

FIG. 3. (a) Cu $2p_{3/2}$ and $2p_{1/2}$ lines for samples with 0.5 nm Cu below Al_2O_3, 0.5 nm Cu above Al_2O_3, a 6 nm Cu reference film, and a 0.5 nm plasma oxidized Cu film. Other lines are explained in the text. (b) Normalized Cu AES intensity for Cu wedges grown above (circles) and below (squares) the Al_2O_3 barrier. Solid line is the result for a Cu(100) single crystalline wedge (see text).

below the barrier, as well as the reference Cu and CuO_x spectra. The presence of Cu in an oxidized state can be observed by the appearance of satellite peaks at slightly higher binding energies than the two primary peaks, as well as a slight shift toward higher binding energy of the primary peaks. For Cu above or below the Al_2O_3 barrier, no detectable presence of the oxide satellite peaks or binding energy shifts caused by alloying were found. The energy difference between the $2p_{1/2}$ and $2p_{3/2}$ binding energies of 19.8 eV is in agreement with Cu reference spectra, further illustrating that only metallic Cu is present in either case. In addition, no oxidized Co was detected, and almost no metallic Al (<1 ML) was observed for these samples [11].

Scanning AES is a well-known technique for investigating the growth mode of thin layers [14]. Measuring the AES intensity as a function of sample thickness, comparative growth studies may be unambiguously performed. In order to compare the growth of Cu on Co vs Al_2O_3, we have performed scanning AES on wedge shaped samples of Si/Ta 5 nm/Co 5 nm/Cu wedge 0–3 nm and Si/Ta 5 nm/Co 5 nm/Al_2O_3/Cu wedge 0–3 nm. In Fig. 3(b) the Auger Cu (920 eV) relative intensities for the two wedge samples are shown as a function of nominal Cu thickness. For Cu grown on Co (*below* Al_2O_3), the observed Auger intensity gives a characteristic AES decay length of $\lambda = 1.31$ nm. In order to unambiguously determine the intrinsic mean-free path *under the same experimental conditions*, a flat, epitaxial Cu(100) wedge was grown on Co(100), which was grown on a Cu(100) single crystal (see, e.g., [15]). Scanning AES measurements on this wedge gave a mean-free path of $\lambda = 1.32$ nm (line in Fig. 3b), in agreement with the reported values of 6–8 ML for ~920 eV electrons [14]. From this, we can conclude that Cu on Co grows in a nearly layer by layer manner for our sputtered samples. For Cu grown on Al_2O_3, however, the growth mode is strikingly different from growth on Co (see again Fig. 3b), similar to island type growth [14]. From the initial portion of the curve, we obtain a fitted decay length of $\lambda \approx 2$ nm, further indicating that Cu does *not* grow in a planar manner on Al_2O_3. In addition, even for 3 nm Cu grown on Al_2O_3 the Auger signal from the underlying Al and O were clearly visible even at 3.0 nm, while for Cu on Co, the Co signal was unobservable beyond 2 nm.

Focusing on the case of Cu on Co, it is determined for this case that we are nearly observing the intrinsic behavior, which we regard as an *upper limit* for the intrinsic length scale. This now allows us to estimate the tunneling spin polarization with one monolayer of Cu coverage on Co. For our (111) textured films, 1 ML Cu has an estimated thickness of 0.208 nm; for this thickness the observed TMR is about 0.45 of its initial value, and by approximately two monolayers, the observed TMR is <0.2 of its initial value. Using the Julliere [2] model, we may estimate an "effective tunneling spin polarization" as well. With $P_{\mathrm{Co}} = 34\%$ at 295 K (obtained from control Co-Co junctions), this gives an apparent tunneling spin polarization of 16%–17% for 1 ML Cu coverage, and approximately 5% at 2 ML Cu coverage. We note, however, that if the same analysis were misleadingly applied to samples with Cu grown on Al_2O_3, the resulting spin polarization would be more than a factor of 3 higher. Sun and Freitas have recently [8] proposed a model of TMR decrease with Cu thickness based on a partially covered ferromagnetic electrode. For the case of Cu grown on Al_2O_3, any uncovered regions where the tunneling is via Co only will dominate the magnetoconductance and obscure the intrinsic behavior of spins tunneling via the Cu layer. Further, the scanning AES measurements for this type of sample indicates that beyond ≈1.5 nm Cu [see Fig. 3(b)], coverage increases much more slowly as growth proceeds in a three dimensional manner. In this regime the TMR will decrease more slowly than for thinner Cu layers, and may also misleadingly lead to a rather long length scale, or a transition between two types of apparent behavior as observed by Sun and Freitas [8]. This perhaps also applies to the case of Au on Al_2O_3, where such a transitional regime in the polarization behavior as a function of thickness beyond 1.5 nm was observed by Moodera *et al.* [10].

The observation of different length scales in the present experiments for differently grown Cu interlayers may explain the disagreement between experimental results in literature. The length scales observed by Sun and Freitas [8] for Cu grown on Al_2O_3 agree quite well with those reported here, while those reported by Parkin [9] are much longer. The latter case may simply be due to the observed

clusterlike growth of Cu on Al_2O_3, higher roughness, or intermixing. Calculations by Bogicevic and Jennison [16] of metal adsorption on ultrathin Al_2O_3 films indicate that Cu is initially wetting on Al_2O_3, but upon subsequent deposition forms clusters. This is consistent with the present AES and TMR results, as well as the TMR data of Sun and Freitas where the Cu was also grown on Al_2O_3.

Zhang and Levy [5] find theoretically that for uniform nonmagnetic layers a rather long (3–10 nm) length scale is expected arising from coherent electron transmission, but that for nonmagnetic layers with thickness fluctuations, only a few monolayers are required to completely quench the TMR. Note that this length scale should not be confused with bulk mean-free paths or spin diffusion lengths. However, in the present case of well grown Cu on Co, still an extremely short length scale is observed. Furthermore, the monotonic decrease of the TMR, vanishing by 1.2 nm Cu, lacks the oscillatory features expected from most simplistic free-electron calculations as well [4,6]. We therefore speculate that the assumption of strict $k_{||}$ conservation or electron transmission with fixed phase may not be quite realistic even for well grown structures. In the absence of coherent transmission, it is not unexpected, based on the calculations of Zhang and Levy, that only a few monolayers are required to completely destroy the tunneling spin polarization. We further conjecture that diffuse scattering at the Co/Cu or Cu/Al_2O_3 interfaces could be responsible for the destruction of $k_{||}$ conservation, making coherent transmission perhaps extremely difficult to observe in these structures, as also pointed out by Mathon and Umerski [7]. By similar reasoning, one might anticipate that quantum well effects will not be observable. Although this is true for the present case, Moodera *et al.* [17] reported very recently oscillatory features in a quite analogous junction with an Au layer at the interface. Given the fact that in this case also a comparable extremely short decay length is observed, this disparity between Cu and Au is rather puzzling and deserves further study.

It is also noteworthy that the TMR length scale observed for Cu on Co, 0.26 nm, is rather close to 1 ML for Cu (0.208 nm). The fact that the TMR is highly sensitive to even one monolayer of Cu may be consistent with tunneling dominated via surface states, or an sp-d exchange interaction between the Co local moments and the Cu tunneling electrons. The latter speculation can also be related to experiments and theoretical work [3] on ultrathin ferromagnetic metals backed with various nonmagnetic metals, where it was proposed that the hybridization of s-p electrons of the backing metal with d electrons in the ferromagnetic layer determined the onset of ferromagnetism, similar to the suppression of impurity magnetism in a nonmagnetic host.

In summary, we find that the intrinsic decay of tunneling magnetoresistance in the presence of a nonmagnetic interface layer can be masked by extrinsic growth-related phenomena, by which an ongoing dispute in recent literature has been explicated. Tunnel junctions with differently grown nonmagnetic layers were used to illustrate this, characterized with *in situ* XPS, Auger, STM, and *ex situ* magnetotransport measurements. For well grown Cu interlayers, we show that only a few monolayers are required to destroy tunneling spin polarization.

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