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Spin-polarized tunneling with Au impurity layers

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We have inserted non-magnetic impurity layers of Au into sputtered AlO_x-based magnetic tunnel junctions (F/I/F) and Meservey-Tedrow junctions (S/I/F) in order to study their effect on the tunneling magnetoresistance (TMR) and spin-polarization (TSP). Both room temperature TMR and the TSP at 250 mK decay exponentially as an interfacial Au layer is introduced between the barrier and one Co electrode, with $1/e$ decay lengths $\lambda_{\mathrm{TMR}} = 11 \pm 3$ Å and $\lambda_{\mathrm{TSP}} = 14 \pm 2$ Å. We also inserted a 1 Å thick Au layer at a variable distance from the barrier/Co interface and find that both the TMR and TSP recover to the undoped value with the shorter exponential lengthscales of $\lambda_{\mathrm{TMR}} = 7 \pm 4$ Å and $\lambda_{\mathrm{TSP}} = 6 \pm 2$ Å.

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The first report of a magnetic tunnel junction (MTJ) was made over 30 years ago by Julliere[1]. These structures consist of an ultrathin insulating barrier separating two ferromagnetic electrodes (a so-called F/I/F configuration). In the past decade there has been a huge upsurge in interest in these structures (recently reviewed by Tsymbal, Mryasov and LeClair[2]) as the improvement of fabrication techniques has lead to values of room temperature tunneling magnetoresistance (TMR) that are large enough for technological applications[3]. The magnitude of the TMR, defined as the fractional change in junction resistance $\Delta R/R$ on switching the junction from a parallel (P) to an antiparallel (AP) magnetization state, depends on the degree of tunneling spin polarization (TSP) of the F electrode materials[1]. This can be measured in an independent way by forming a junction between the ferromagnet in question and a superconductor (an S/I/F structure) by a means devised by Meservey and Tedrow[4].

The quality of the interfaces on either side of the tunneling barrier is widely considered to be paramount in these structures. Here we describe experiments where we have deliberately inserted thin layers of Au as an impurity at this point. We have introduced the Au both at the interface between a Co electrode and an AlO_x barrier, and also as a δ-layer, which we can place at a variable distance from that interface. This second case mirrors a previous study of the giant magnetoresistance (GMR) in Co/Cu/Co spin-valves[5]. There are reports of previous experiments on MTJs similar to our interface layer studies. Moodera et al. deposited Au layers onto Co electrodes before covering them with plasma-oxidized alumina, and found that the TMR almost vanished for Au layers thicker than about 1 nm, in addition to a weak quantum well oscillation[6]. Other spacer materials have also been studied, such as Cu[7–9], Cr[10], and Ru[11,12]. A variety of different effects, some pointing to the presence of quantum well states, were found[8,11]. On the other hand, we know of no comparable studies in the tunneling regime for the δ-layer experiments.

Our junctions were of the cross-strip form, deposited by dc magnetron sputtering through shadow masks, which were changed in situ to give an active area 50 μm square. The substrates were pieces of Si wafer with ~ 100 nm thermal oxide at the surface. The chamber base pressure was $\sim 2 \times 10^{-8}$ Torr, whilst the working pressure of Ar was 2.5 mTorr. Typical deposition rates, calibrated by low-angle x-ray reflectometry on test films of each material, were 2.5 Å/s for the metal layers. The barriers were formed by dc plasma oxidation in 55 mTorr of O_2 for 30 s at a power of 100 W. Two types of junctions were grown: the first were F/I/F MTJs based on the following layer stack sequence: substrate / Al (150 Å) / $Co_{68}Fe_{22}B_{10}$ (40 Å) / AlO_x (13 Å) / Co (30 Å) / $Ir_{20}Mn_{80}$ (60 Å) / Al (150 Å). The IrMn layer pins the Co through the exchange bias effect to control the switching between the P and AP states. The others were S/I/F junctions based on the stacking sequence $Al_{96}Si_4$ (40 Å) / AlO_x (13 Å) / Co (200 Å). The superconducting $Al_{96}Si_4$ layer had a critical temperature T_{C} of ~ 3 K and an in-plane critical field, measured at 1.4 K, exceeding 5 T; both quantities were determined from two point resistance measurements of a bottom electrode strip. The junctions were doped by introducing a thin layer of Au either at the barrier/Co interface, or at a point within the Co layer a distance x from that interface. It is also important to emphasize that the growth protocol used here has been demonstrated to give highly reproducible results: as series of several nominally identical undoped MTJs had TMR ratios and resistance-area products that were the same to within only a few per cent. This means that sample-to-sample variations should not mask any changes due to the introduction of Au above this level. The good tunnelling I-V properties of our junctions (not shown), along with x-ray reflectometry of comparable sheet film test samples, confirms that interfacial roughnesses are all well below 1 nm, and our barrier is smooth and pinhole-free.

The TMR of the MTJs was measured by a conventional four-probe dc technique at room temperature: measure-

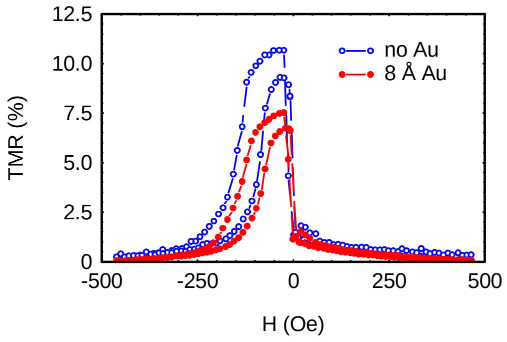

FIG. 1: (Color online) The TMR with field of a undoped MTJ, and a similar junction with 8 Å of Au at the pinned Co layer interface, measured at room temperature and a voltage bias of 10 mV.

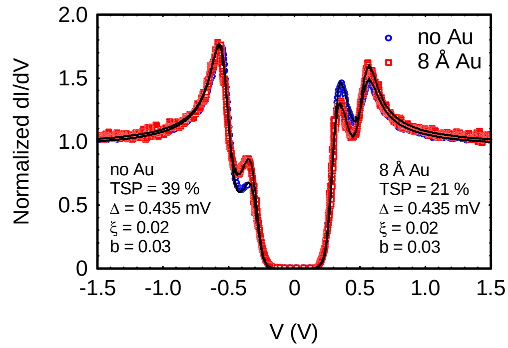

FIG. 2: (Color online) The differential conductance with bias for an undoped $Al_{96}Si_4/AlO_x/Co$ junction and a similar junction with 8 Å of Au at the barrier/Co interface, normalized to the high bias value. Simulations of the data using the AORT model are plotted as black solid lines. The model parameters for each curve are given in the plot.

ments were taken under variable bias, but we only show data here for a 10 mV applied voltage bias. All junctions were measured in their as-grown, unannealed state, to prevent any migration of the Au within the structure, for instance along grain boundaries. The differential conductance (dI/dV) as a function of applied bias V of the S/I/F junctions were measured at 250 mK in a ^{3}He refrigerator, under a constant applied field of 2 T supplied by a superconducting magnet. A separate transverse coil was used to null off any small component of field normal to the junction surface due to sample misalignment, that would otherwise give rise to vortices in the superconductor.

In Fig. 1 we show an example of some TMR data from a pair of selected F/I/F samples: an undoped one, which conforms exactly to the standard stacking sequence given previously, and one in which a Au doping layer of 8 Å thickness has been introduced at the interface between the Co pinned layer and the alumina barrier. The undoped junction has a TMR of 11%, whilst the introduction of the Au barrier has reduced the TMR to 8 %. It can be seen that there are no other significant changes to the form of the TMR loop apart from a reduction in its amplitude.

We also measured the effects of introducing a Au interface layer on the TSP of the Co electrode by the Meservey-Tedrow method. Representative data measured on an undoped S/I/F junction, and one containing an 8 Å thick Au layer at the barrier/Co interface are shown in Fig. 2. The four peaks corresponding to the Zeeman split edges of the BCS (Bardeen-Cooper-Schrieffer) gap in the $Al_{96}Si_4$ electrode are clearly visible. These data were modeled using the theory of Alexander, Orlando, Rainer, and Tedrow (AORT)[13,14]. In this model there are four input parameters: the BCS gap Δ, the spin-orbit parameter b (which accounts for all the mechanisms that mix spin up and down without destroying the Cooper pairs), the spin-flip or depairing parameter ξ (which includes all mechanisms which break the time-reversal symmetry and hence break Cooper pairs), and the TSP itself. The fits to the data are shown in the plot as solid lines, along with the values of these parameters. Both samples are well described by similar values for Δ, b, and ξ. The TSP of our undoped sample is 39%, a typical value for a transition metal[15]. This is roughly halved, to 21 %, on the introduction of the Au interface layer.

Having shown representative data for both F/I/F and S/I/F junctions, we turn our attention to a summary of all our results, shown in Fig. 3. In panel (a) of that figure we show the data for the introduction of a Au interface layer of variable thickness between the barrier and Co electrode for both types of samples. There is a drop in TMR when the Au is first introduced, but, remarkably, an easily measurable TMR of a few % is still present for Au thicknesses exceeding 20 Å. We were able to fit our data (excluding the first point) with a phenomenological exponential decay, which returns a $1/e$ lengthscale of $\lambda_{\mathrm{TMR}} = 11 \pm 3$ Å. The TSP is seen to decay in a similar manner to the TMR. There is a rather abrupt drop in TSP on introduction of the first Å of Au from about 40 % to 30 %, but then the TSP decays more gently: again there is a finite and substantial TSP for 20 or 30 Å of Au between the barrier and the Co. Again an exponential decay fits the data (excluding the first data point), yielding $\lambda_{\mathrm{TSP}} = 14 \pm 2$ Å. No sign of quantum well oscillations is present in these data.

In Fig. 3(b) we show similar data for the case where 1 Å thick Au δ-layers have been introduced into the Co layer at a variable distance from the barrier/Co interface. When the Au is at the interface both TMR and TSP are suppressed in line with what is seen in Fig. 3(a). As the Au moves into the Co layer both the TMR and the TSP rise smoothly and rapidly towards the 39 % value for an undoped Co layer. Exponential fits yield substantially shorter $1/e$ lengthscales than those found above, $\lambda_{\mathrm{TMR}} = 6 \pm 4$ Å, whilst $\lambda_{\mathrm{TSP}} = 7 \pm 2$ Å: again,

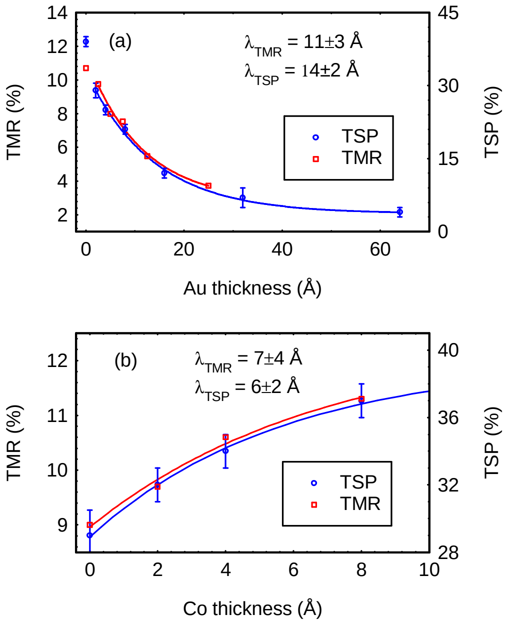

FIG. 3: (Color online) (a) The dependence of TMR and TSP on Au interface layer thickness. (b) The dependence of TMR and TSP on the distance of a 1 Å Au δ-layer from the barrier/Co interface. TMR was measured at 300 K, TSP at 250 mK. The solid lines are the results of fitting exponential decays to the data, and in each plot we report the decay lengthscales, λ, extracted from the fits.

the two fitted values are identical to within the error bar. We interpret this result as showing the lengthscale on which the Au atoms at the interface are replaced with Co: once the entire interface consists of Co the full TSP is recovered. This result is reminiscent of those obtained in GMR spin-valve systems[16,17].

The most interesting feature of these results is how long the exponential decay lengths in Fig. 3(a) are when compared to those in Fig. 3(b) and to most of the results cited in the introduction. One possibility is that the Au grows in islands and the data simply represent the fraction of the barrier which is still in contact with some Co. We have performed some atomic force microscopy (AFM) to try to gain information about the Au layer. Fig. 4 (a) shows an AFM image of 7 Å of Au grown on an AlO_x barrier layer, part of an incomplete MTJ stack. Small grains $\sim$ 10 nm in diameter are visible, which are likely to be made of Au. Having performed an image flooding analysis, we can say that only 11 % of the image area lies more than the nominal 7 Å thickness below the average image height. Hence, if this topography were covered with a Co layer, we would expect that Co would be in direct contact with the barrier only in this area fraction, and we ought to observe no more 11 % of the TMR or TSP of a junction without Au. Nevertheless, we can see from Fig. 3 that the observed values are rather higher than this.

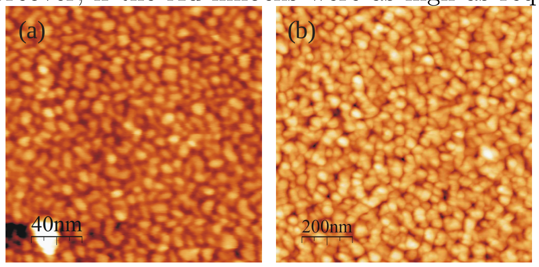

FIG. 4: (Color online) Atomic force micrograph of (a) a Au (7 Å) / AlO_x / SiO_2 sample ($\sigma_{\rm rms}$ = 7.9 Å), and (b) a completed MTJ stack including 7 Å Au ($\sigma_{\rm rms}$ = 11.5 Å).

Moreover, if the Au hillocks were as high as required by to explain the reduction in TMR and TSP using this island-growth hypothesis, the huge additional roughness for the thicker Au layers, a few nm in amplitude, would lead to a substantial increase in the orange-peel coupling field for the free layer,[18], whereas we see no observable change in Fig. 1. Moreover, this enormous roughness should also be present at the top interface of the Co layer, with the IrMn, which should give rise to a drastic reduction in exchange bias, which is also absent in our experiment. Such rough layers might also be expected to have a very much higher coercivity - again this is not observed in the TMR loops. Fig. 4(b) shows an AFM image of a completed MTJ stack including 7 Å of Au, which will give some idea of the roughness in these layers, as conformal roughness is generally accumulated through the stack in sputtered samples: the rms roughness $\sigma_{\rm rms}$ is barely more than $\sim$ 1 nm.

Hence, a substantial excess TMR and TSP remains to be explained for the case of the Au interfacial layers. An interesting possibility is that our results are due to some spin-polarization in the Au. This could arise either by a proximity effect due to the contact with the Co, or by spin injection when a current is driven by a voltage bias. This will give rise to a weak spin-polarization at the barrier which is detected by the tunnel junction.

To summarize, our measurements show that a substantial TSP can penetrate a few nanometers of Au from an interface with Co, measured both directly at 250 mK and by TMR measurements at room temperature. On the other hand both the TMR and TSP recover from their suppression by the Au δ-layer much more quickly when it is withdrawn into the Co, with the $1/e$ lengthscales < 1 nm. It seems that the TMR and TSP observed for the thicker Au layers cannot be explained entirely by discontinuities in the Au layer. Further modelling and characterization to investigate this point is ongoing at the time of writing, as well as experiments using other dopant species.

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