

## Kondo Effect in Magnetic Tunnel Junctions

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Tunneling magnetoresistance was found to be suppressed with decreasing temperature for magnetic tunnel junctions (MTJs) oxidized under high plasma power. A strong temperature dependence of the junction resistance was observed, along with zero-bias anomalies of dynamic resistance at low temperatures. Resistance shows a logarithmic dependence on temperature, and resistance versus temperature exhibits a scaling behavior. Our experimental data can be explained in a consistent way by the Kondo effect in the MTJs with the Kondo temperature  $T_K = 20\text{--}30$  K.

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The Kondo effect [1] is one of the most interesting problems in condensed matter physics. When the magnetic impurities are introduced into a normal metal host, measurable quantities such as resistivity become strongly temperature dependent at low temperatures. The Kondo effect is also observed in several mesoscopic systems including planar tunnel junctions [2,3], quantum dots [4], quantum nanobridges [5], etc. In *nonmagnetic* tunnel junctions the Kondo effect manifests itself as a zero-bias anomaly (ZBA) peak in the dynamic conductance [2] or dynamic resistance [3] depending on the position [6,7] of the magnetic impurity [8] in the nonmagnetic tunnel junctions. Studies of the Kondo effect both experimentally and theoretically [1,9] have focused primarily on the case of a normal metallic host. One unresolved question is if the Kondo effect can survive in a *magnetic* metallic host [10]. We attempt to answer this outstanding problem by studying the transport properties of magnetic tunnel junctions (MTJs), and provide experimental evidence for the observation of Kondo effect in the MTJs.

In this Letter we study the unusual dependence of tunneling magnetoresistance (TMR) on temperature ( $T$ ) and the oxidation time ( $t_O$ ) in MTJs ( $\text{Co}_{84}\text{Fe}_{16}/\text{Al}_2\text{O}_3/\text{Co}_{84}\text{Fe}_{16}$ ). A series of MTJs were fabricated by oxidizing the Al layer with varying  $t_O$  under rather high oxidation power (150 W), and systematic changes in the dependence of TMR on  $T$  and  $t_O$  were observed. We find (i) strongly suppressed TMR at low temperatures, more pronounced as  $t_O$  is increased, (ii) strongly  $T$  dependent peak in dynamic junction resistance ( $R$ ), and (iii) scaling behavior of  $R(T)$ . The  $R$  and TMR for usual MTJs [11,12] monotonically and gradually decrease as a function of temperature [13–16], and can be explained within the (extended) Julliere model [17] in terms of the temperature dependence of the magnetization in the ferromagnetic (FM) electrodes [18]. What is surprising is the observation [19] of a strongly  $T$  dependent

ZBA in the dynamic resistance of our MTJs, which can usually be attributed to Kondo scattering of (spin-polarized) tunneling electrons. Based on the Kondo scattering model, we provide a consistent explanation of our experimental data.

A series of MTJs were fabricated to investigate the effects of oxidation time on TMR. The  $\text{Al}_2\text{O}_3$  barrier was formed by oxidizing 1.6 nm Al layer with oxygen plasma of 150 W for  $t_O = 17, 20, 23,$  and  $26$  sec (s), respectively. The samples were heat treated by a rapid thermal annealing method [20] for 10 s at  $300^\circ\text{C}$ . Details of sample preparation for MTJs can be found in Refs. [20–22].

The  $T$  dependence of the zero-bias TMR is presented in Fig. 1 with varying oxidation time  $t_O$ . The inset shows the MR curve of several MTJs measured at 2 K. Though the TMR is reduced significantly at low temperatures with increasing  $t_O$ , the switching fields remain the same for all the MTJs. This reflects the fact that the magnetic properties

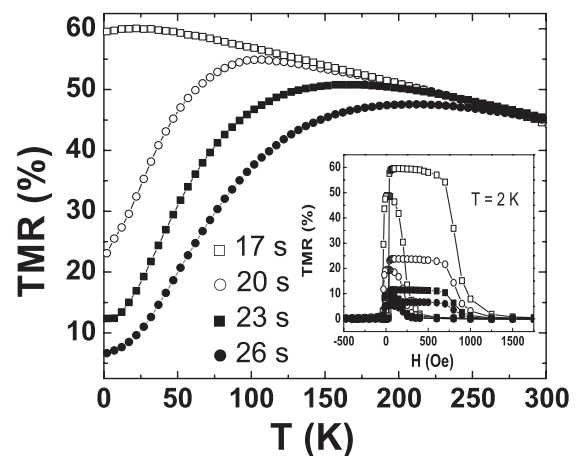


FIG. 1. Temperature dependence of TMR with oxidation time  $t_O = 17, 20, 23, 26$  s. The inset shows the TMR loop at 2 K.

of lower electrode are completely recovered through the annealing process. The MTJ with  $t_O = 17$  s (least oxidized of our MTJs) exhibits a monotonic increase of TMR with lowering  $T$ . The TMR of this least oxidized junction approaches a moderate value of 60% as  $T \rightarrow 0$  K. The  $T$  dependence of the TMR can be fitted to  $MR(T) = MR(T=0)(1 - AT^{3/2})$  with  $A = 5.1 \times 10^{-5} \text{ K}^{-3/2}$ , as expected from the magnetization in FM [13]. The TMR for the MTJs with  $t_O = 20, 23,$  and  $26$  s shows nonmonotonic dependence on  $T$ , and additionally exhibits a broad peak whose position depends on  $t_O$ . The TMR at low temperatures is reduced systematically with increasing  $t_O$ , though the TMR for all junctions approaches 45% at room temperature, regardless of the values of  $t_O$ .

The resistance of MTJs with varying  $t_O$  is plotted as a function of  $T$  in Fig. 2(a) when the magnetizations of both electrodes are parallel. The inset shows the resistance difference ( $\Delta R = R_{AP} - R_P$ ) between antiparallel (AP)

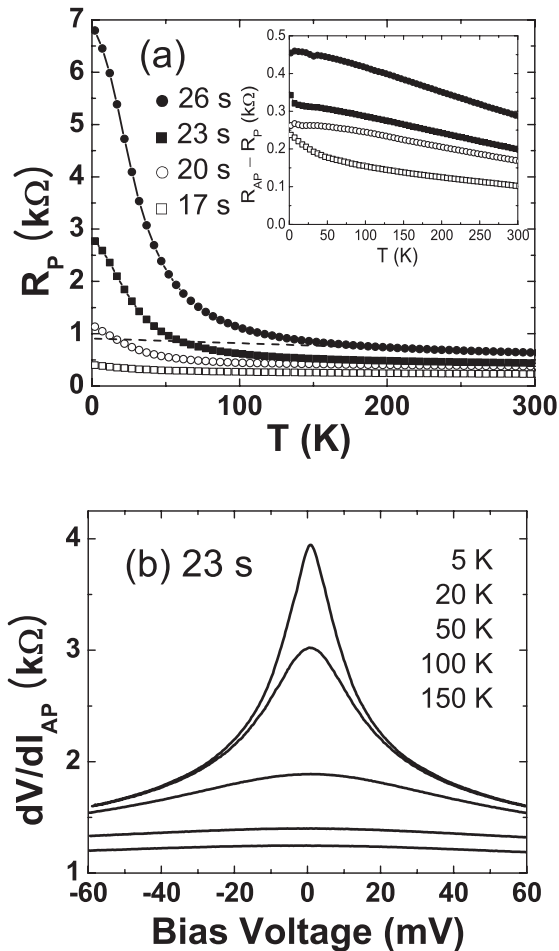


FIG. 2. (a) Temperature dependence of junction resistance  $R_P$  (parallel alignment) with  $t_O = 17, 20, 23, 26$  s. The dashed line is the temperature dependence of control MTJs [24]. The resistance difference  $R_{AP} - R_P$  is displayed in the inset. (b) Dynamic resistance (antiparallel alignment) with varying temperatures for the MTJ with  $t_O = 23$  s.

and parallel (P) magnetizations with varying  $T$ . Noticeable trends can be identified in our MTJs with changing  $t_O$ : (i) the resistance increases over the whole temperature range with increasing  $t_O$ , (ii) both  $R_P$  and  $R_{AP}$  show a sharper increase at low  $T$ 's for MTJs with longer  $t_O$ , and (iii)  $\Delta R$  is monotonic as a function of  $T$ .

The resistance at room temperature increases almost linearly with  $t_O$  ( $R_P = 229, 377, 439, 639 \text{ } \Omega$  with  $t_O = 17, 20, 23, 26$  s, respectively.) Since all our samples are fabricated with the same junction size ( $50 \times 50 \text{ } \mu\text{m}^2$ ) and initial thickness (1.6 nm) of the Al layer before oxidation, we can deduce that the differences in junction resistance stem mainly from changing  $t_O$ . Since our MTJs are prepared under a rather high oxygen plasma (150 W), the bottom FM layer is oxidized, especially for longer  $t_O$ , and the tunnel barrier becomes effectively widened. According to Simmons' model [23], a 0.1 nm increase of the barrier width can give rise to a factor of 5 increase of the junction resistance (using a typical effective barrier height of 2.5 eV). A slightly longer oxidation of the magnetic layer may be the cause of the significant increase of resistance as observed in our MTJs.

More interestingly, the longer oxidation time  $t_O$  leads to a more rapid increase of the junction resistance with lowering  $T$ . For ideal tunnel barriers, the junction resistance is expected to decrease with increasing  $T$  due to thermal excitations of tunneling electrons. This effect for typical barrier width and height of MTJ results in only a gradual change in resistance [13]. For a MTJ with  $t_O = 17$  s, the change of resistance is almost linear and about a 70% increase is observed from 300 to 2 K. This is a typical behavior of the junction resistance with  $T$  which has been observed in other group's experiments [13–16] as well. The resistance increases about 3, 6, and 11 times as the temperature is lowered from 300 to 2 K for  $t_O = 20, 23,$  and  $26$  s cases, respectively. The effectively widened tunneling barrier alone cannot explain this strong  $T$  dependence of the junction resistance. In addition to widening the effective tunneling barrier, the longer oxidation seems to produce a new scattering mechanism which becomes active at low  $T$ .

The resistance difference  $\Delta R$ , as with usual MTJs [13–16], increases slowly with decreasing  $T$ , which is similar to the  $T$  dependence of magnetization of the FM layer. This means that the spin polarization (SP) of tunneling current is not destroyed by strongly  $T$  dependent scattering mechanism. The change in  $\Delta R$  with  $t_O$  is very weakly influenced by the strongly  $T$  dependent scattering process at low  $T$ 's, but can be explained by the effectively increased width of tunneling barrier.

The dynamic resistance ( $dV/dI$ ) as a function of bias voltage with varying  $T$  is displayed in Fig. 2(b) for the MTJ with  $t_O = 23$  s. The dynamic resistance is featureless and flat near zero bias at high  $T$ 's, but develops a peak structure near zero bias at low  $T$ 's. The zero-bias anomaly in the

dynamic resistance also suggests that the additional scattering process becomes active at the low  $T$  and low bias voltage regime. Note that the strongly  $T$  dependent ZBA in *nonmagnetic* tunneling junctions [2,3] was explained by invoking the Kondo effect [6,7] due to magnetic impurities [8] near the metal-insulator interface. We note in passing that other groups [12,15,19] observed weakly  $T$  dependent ZBAs in  $dI/dV$ , which is in strong contrast with our data. A different scattering mechanism is in play in our MTJs.

The ZBA in dynamic resistance, as well as dramatic increase of the resistance, implies that an additional scattering process is turned on at low  $T$ . We can estimate the contribution (denoted as  $R^*$ ) to the junction resistance of this scattering process by subtracting the  $T$  dependence of control MTJs [24] from the raw data. For example,  $R_p^*$  for the  $t_O = 26$  s case is obtained by subtracting the dashed line from the solid circles in Fig. 2(a). Both  $R_p^*$  and  $R_{AP}^*$  are plotted in Fig. 3 with two scaling parameters  $R_0$  and  $T_K$ . Here  $R_0$  is the value of  $R^*$  extrapolated at  $T = 0$  K and  $T_K$  is defined as the temperature satisfying the relation  $R^*(T_K) = R^*(T = 0)/2$ .  $T_K$  is about the same for both P and AP configurations for a given oxidation time. The estimated  $T_K = 22, 25, 30$  K for samples with  $t_O = 20, 23, 26$  s, respectively.  $R^*$ 's with different  $t_O$  collapse into one curve showing the universal scaling behavior. The solid line is obtained from the empirical expression [25] with  $s = 0.9$ ,  $R^*(T) = R_0[T_0^2/(T^2 + T_0^2)]^s$  where  $T_0 = T_K/\sqrt{2^{1/s} - 1}$ . The  $R^*$  shows a logarithmic dependence on  $T$  in the crossover regime near  $T_K$ , and saturates below  $T_K$ . The scaling behavior and the logarithmic  $T$  dependence of the resistance are we believe strong evidence for the Kondo effect in our MTJs.

The localized magnetic moments, which are necessary for observing the Kondo effect, are, we believe, formed in

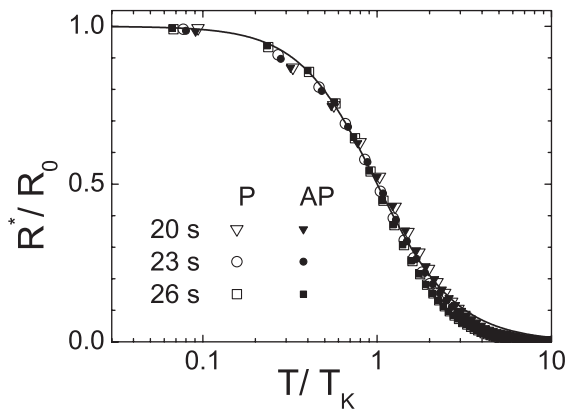


FIG. 3. Scaling behavior of the junction resistance contribution from the impurity scattering (see text). The Kondo temperature  $T_K$  is about 22, 25, and 30 K for samples oxidized 20, 23, and 26 s, respectively. The value of  $R_0$  is about 680 (680), 2218 (2250), and 5942 (5900)  $\Omega$  for the three samples with parallel (antiparallel) magnetization orientations. The solid line is the empirical curve with  $s = 0.9$  (see text).

the process of oxidizing the Al layer. Since the power of the oxygen plasma is rather high, the oxygen ions penetrate the Al layer and reach the FM CoFe electrode. We determined the concentration of atomic elements, using the x-ray photoemission spectroscopy (XPS) while milling the MTJs. The signature of excess oxygen (O) atoms at CoFe was observed just below the interface when the Al oxide layer was totally removed (no Al signal) [26].

To further investigate the nature of the scattering centers, we performed the grazing incident angle x-ray diffraction (XRD) and the x-ray reflectivity (XRR) measurements at the 2C1 beamline in Pohang Light Source on the  $\text{AlO}_x/\text{CoFe}/\text{Ta}/\text{SiO}_2/\text{Si}$  structure by controlling Al oxidation time [27]. The XRR result showed that an extra layer of 5  $\text{\AA}$  thickness is formed between  $\text{AlO}_x$  and CoFe layers during the oxidation. The grazing angle XRD further elucidated the presence of  $\text{Al}_{1+x}\text{Fe}_{1-x}\text{O}_{3+\delta}$  clusters with  $-0.3 \leq x \leq 0.2$  and  $\delta \leq 0.05$ , but the amount of the clusters turned out to be much less than one monolayer, indicating that the ferrite clusters are sparsely distributed all over the interface. The paramagnetic impurities seem to be formed within the oxidized interface region (presumably Fe ions in a mixed and disordered Fe-Al oxide matrix), consistent with work on *nonmagnetic* tunnel junctions [2,3].

The structure of ZBA is known to be very sensitive to the topology between the Kondo impurities and the tunnel junction [6,7]. When the Kondo impurities are located inside the  $\text{Al}_2\text{O}_3$  layer, the conductance is enhanced due to opening of the new Kondo resonant channel with lowering  $T$ . When the Kondo impurities form at the interface between one lead and the tunnel barrier, these Kondo impurities act as strongly  $T$  dependent scattering centers and tend to block the flow of electrons from one lead to the other. The second scenario conforms with the strong enhancement of resistance in our MTJs.

Our measured data are consistent with a distribution of the Kondo impurities in between the bottom FM metal and aluminum oxide layers. Since the Kondo impurities are located next to the one FM electrode, their coupling to the second FM lead is weak due to the oxide barrier and can be neglected compared with the coupling to the first FM. This means that the strongly  $T$  dependent part  $R^*$  is insensitive to the magnetization direction of the second FM as observed in Fig. 2(a). The quenched TMR [28] at low temperatures can now be understood by noting that the Kondo contribution ( $R^*$ ) to  $R$  is dominant at low  $T$ 's and is insensitive to the magnetization configuration. The Kondo temperature  $T_K$  is determined by the coupling of the Kondo impurity to the bottom FM electrode and is also insensitive to the magnetization direction of the second FM electrode. Hence the almost same  $T_K$ 's for P and AP configurations (from the scaling analysis in Fig. 3) are consistent with our model picture. With increased  $t_O$  more oxygen can penetrate the Al layer and oxidize Fe and Co, such that the

Kondo scattering centers become more concentrated. Since the scattering rate of tunneling electrons is proportional to the concentration of Kondo impurities (when the interimpurity interaction is neglected), the resistance is higher for the MTJs with longer  $t_0$ . When the Zeeman splitting due to applied  $B$  field is comparable to or larger than  $T_K$ , the ZBA peak in  $dI/dV$  will split into two peaks [29]. We observed no splitting of ZBA peak in our MTJs up to  $B = 9$  T at 2 K. Higher field (30–45 T) is required to observe the expected splitting because of the large  $T_K$  (20–30 K) in our MTJs. In conventional tunnel junctions [3], splitting of the ZBA peak in dynamic conductance (small  $T_K$ ) [2] was observed while no splitting in resistance (large  $T_K$ ) [3] as in our case.

The Kondo effect is destroyed due to the splitting of spin states in a quantum dot (QD) by the exchange field, when the QD is coupled to the two FMs [10]. The Kondo effect can survive only when two FMs are aligned antiparallel and are coupled to the QD symmetrically. Only in this case the hybridizations with two FMs of spin-up ( $\Gamma_\uparrow$ ) and spin-down ( $\Gamma_\downarrow$ ) states of an impurity are equal. In our model picture the magnetic impurity is coupled to only one FM lead so that seemingly  $\Gamma_\uparrow \neq \Gamma_\downarrow$ . According to the recent study of the surface density of states for Co and Fe [30], the Fermi surface is dominated by the  $d$ -band, and the  $s(d)$ -band in Co has the positive (negative) SP, respectively. When the Al oxide layer is sandwiched between two Co or Fe metals, the  $s$ -band electrons relatively easily tunnel through the barrier, while the  $d$  electrons are strongly suppressed in the barrier. The  $s$  electrons are mainly responsible for the tunneling leading to the positive SP [30]. Because of the different nature of wave functions of  $s$  and  $d$  electrons in the barrier and their opposite SP (similar to the antiparallel alignment of two FMs in QD), the total hybridizations with  $s$  and  $d$  bands for spin-up and spin-down states may be in a fine balance for our MTJs ( $\Gamma_\uparrow \approx \Gamma_\downarrow$ ) so that the Kondo effect is observed in our MTJs. Above all, our transport data (the universal scaling behavior of  $R^*(T)$  and a strong  $T$  dependent ZBA in  $dI/dV$ ) are the strong evidence for the Kondo effect.

In summary, we observed the Kondo effect in magnetic tunnel junctions, caused by the magnetic impurities (Fe ions in Al-Fe oxide matrix) formed during the Al oxidation. The resistance is strongly enhanced at low temperatures and shows the universal scaling behavior. The dynamic resistance exhibits a zero-bias anomaly peak. These are attributed to Kondo scattering of tunneling electrons by the magnetic impurities at the electrode-barrier interface. At temperatures below  $T_K$ , the junction resistance is dominated by the Kondo scattering contribution such that the TMR is strongly suppressed. Since the magnetic impurities are inactive at room temperature (above  $T_K$ ), the TMR approaches the same value regardless of the oxidation time, which is determined by the spin polarizations of the ferromagnetic electrodes.

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