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Interface Magnetism and Spin Wave Scattering in Ferromagnet-Insulator-Ferromagnet Tunnel Junctions

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Careful tunneling studies in high quality $Co/Al_2O_3/Ni_{80}Fe_{20}$ junctions show a junction magnetoresistance (JMR) of 20.2% and 27.1% at 295 and 77 K, respectively, where the latter is in agreement with Julliere's model. The temperature dependence of the JMR can be explained by the temperature dependence of surface magnetization. The decrease of the JMR with increasing dc bias is intrinsic to ferromagnetic junctions. The strong disagreement with recent theories in the low dc bias region can be attributed to magnetic excitations in these junctions, as seen in inelastic tunneling measurements. [S0031-9007(98)05668-3]

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The spin-dependent tunneling between two ferromagnetic (FM) films across an insulator (I), successfully shown recently [1], has application potential in digital storage and magnetic sensor technologies [1–4]. The magnitude of the junction magnetoresistance (JMR) at low temperatures nearly agrees with Julliere's simple model predictions [5]. This model is based on the difference in the density of states (DOS) for the two spin directions at E_F of the itinerant electrons in the FM [6], and earlier results of spin polarized tunneling between a FM and a superconductor [7].

The JMR exhibits both a temperature (T) and a dc bias (V_{dc}) dependence [1]. These effects are surprisingly significant and depend on the quality of the junctions; the lower the JMR, the larger the temperature and the dc bias dependence. Earlier, others have reported a few percent JMR at liquid helium temperatures, whereas at room temperature it was only (0–2)%. Likewise, junctions showed a factor of 10 drop in JMR when V_{dc} was increased from 0 to 0.5 V [1,8]. There are also cases where no JMR is observed at any temperature despite having a good tunnel barrier.

Recently, theories have been proposed [9–15] to extend and make Julliere's model rigorous, in order to explain FM-I-FM tunneling and also some of the above observations. So far, they have not been very satisfactory. For example, a decrease in JMR with increasing dc bias has been predicted [12,13], but slower at low values of V_{dc} than seen experimentally.

In this Letter, we present experimental observations that show the intrinsic behavior of the ferromagnetic tunnel junctions. Our dynamic conductance (G) and inelastic electron tunneling spectroscopy (IETS) results show unique features that provide an experimental basis for a theoretical understanding of both the temperature and the dc bias dependence of the JMR.

The tunnel junctions were prepared by cryogenic evaporation through shadow masks and glow discharge oxidation, as described in previous publications [1]. Out of the 72 junctions that were prepared, there were 12 junctions with $Co_{50}Fe_{50}$ or $Ni_{80}Fe_{20}$ as the top electrode for each barrier thickness (Al film thickness ranging from 0.4 to 1.8 nm). The bottom electrode was Co for all junctions. The measured junction resistances (R_J) were in the range of 1 to 30 kΩ depending on the junction type. The junctions showed a resistance increase of less than 20% upon cooling from room temperature down to 77 K and negligible below that, comparable to $Al/Al_2O_3/Al$ reference junctions. The junctions were temperature cyclable and stable in ambient atmospheres even after six months, displaying the same characteristics. The JMR *(defined as the resistance change in a magnetic field relative to the peak value)* ranged between (14–20)% at 295 K and (24–27)% at 77 K. The best junctions (showing the highest JMR at room temperature) were $Co/Al_2O_3/Ni_{80}Fe_{20}$ with 0.8 nm of Al [16]. Several of these junctions showed a room temperature JMR of 20%. One of these best junctions was selected for detailed studies and its characteristics will be discussed in the remainder of this Letter. Similar features were seen for other good junctions as well.

The current-voltage (I-V) characteristics for the junctions were measured from room temperature down to 1 K, both for parallel and antiparallel orientations (see the section on magnetoresistance). Tunnel current was higher for the parallel than for the antiparallel orientation at all temperatures, up to 0.8 V. The I-V data were fitted to Brinkman's formula [17]. The fitted average barrier height at 295 K (1 K) for parallel orientation was 3.34 (3.59) eV, in excellent agreement with that of similarly prepared $Al/Al_2O_3/Al$ reference junctions. The corresponding barrier asymmetry and thicknesses were 0.32 (1.24) eV and 1.22 (1.19) nm, respectively. A 0.8-nm-thick Al film, as used in the barrier formation, is expected to increase to a thickness of about 1.1 nm after oxidation [18]. The fitted barrier thickness is within 10% of the expected value, thus showing good barrier uniformity. This is further substantiated by atomic force microscope observation of Al_2O_3 covered Co films, showing an rms roughness of less than 0.5 nm. All of these qualities

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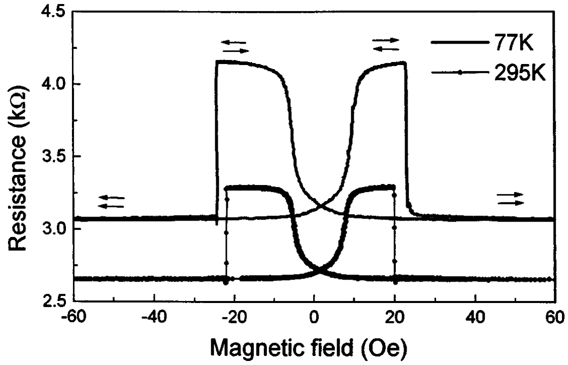

FIG. 1. Resistance vs applied magnetic field for a $Co/Al_2O_3/Ni_{80}Fe_{20}$ junction at room temperature and 77 K showing JMR values of 20.2% and 27.1%, respectively. This measurement was done using an LR-700 ac resistance bridge with an excitation ac current at 100 nA. Arrows indicate the magnetization configuration of the two FMs according to Julliere's model [1,5].

indicate that the optimum barrier with relatively clean FM interfaces is achieved. (Note that the tunneling current depends not only on the barrier properties, but also on the DOS of the electrodes and inelastic tunneling processes [19], which are not explicit in the tunneling theories. Hence, the barrier heights and the thicknesses given above are the "effective" values.)

The junction magnetoresistance curves at room temperature and 77 K are plotted in Fig. 1. They show hysteresis and a peak at low fields for either sign of the applied field. The peaks and valleys correspond to antiparallel and parallel orientations of the magnetizations (M) of the two FM electrodes, respectively [5]. The peak occurs in between the coercive fields of $Ni_{80}Fe_{20}$ (~5 Oe) and Co (~25 Oe) films. Nearly flat peaks indicate good antiparallel alignment of M, with the peak width increasing slightly with decreasing temperature. The JMR obtained from these data are 20.2% and 27.1% at room temperature and 77 K, respectively. Cooling the junction from 77 K down to 1 K caused only a marginal increase in R_J and the JMR stayed at 27.3%. The same trend was seen in all junctions, irrespective of the type of FM used. These are the best values reported for any ferromagnetic junction to date.

Dynamic conductances measured at room temperature and 1 K for parallel (upper curve) and antiparallel (bottom curve) M configurations of the electrodes are shown in Fig. 2. The curves are asymmetric with respect to zero dc bias, the parallel configuration showing more asymmetry. G increases by less than a factor of 2 between 0 and 0.75 V, indicating a high barrier. Features can also be seen in these curves between 0 and 200 mV, more apparent in the parallel case. The curves sharpened near zero bias as T decreased, more for the antiparallel orientation. This behavior is similar to the zero bias anomaly usually seen with transition metal electrodes or with impurities in the barrier [19,20]. However, in the present experiments, there were no changes in G between

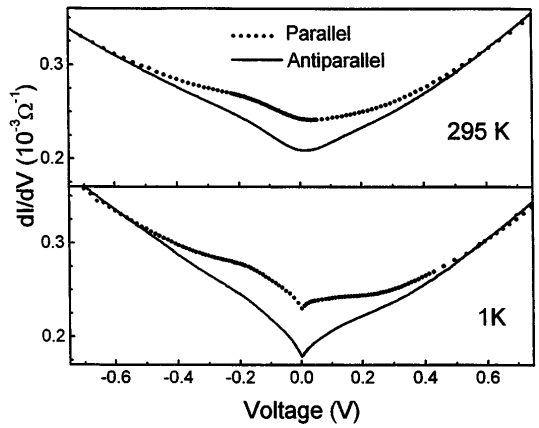

FIG. 2. Dynamic conductance at two temperatures as a function of dc bias for parallel and antiparallel orientation of magnetizations for the same junction as in Fig. 1.

4.2 and 1 K, unlike in the case with impurities or states in the tunnel barrier.

Figure 3 shows the zero field IETS spectra, i.e., the derivative of G, measured at 295, 77, and 1 K in zero field. There was little difference in these curves when measured in an applied magnetic field up to 300 Oe. Looking at the room temperature data, one can see a broad peak (dip) at about ± 100 mV, slightly better defined upon cooling down to 77 K. At 4.2 K, in addition to this peak, a sharp feature appears at 17 mV, with no change occurring between 4.2 and 1 K. Finally, a small peak

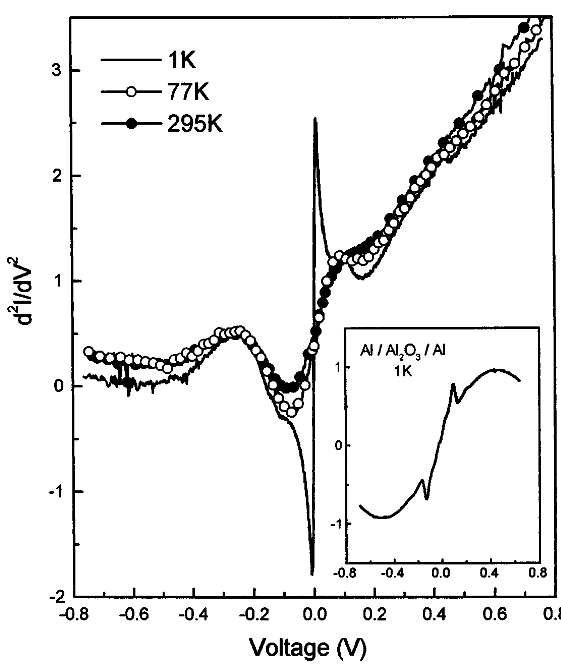

FIG. 3. IETS spectra at three temperatures for the same junction as in Fig. 1, measured at $H = 0$. Similar spectra are seen for junctions where one electrode is a FM and the other electrode is Al. The inset shows an IETS spectrum of an $Al/Al_2O_3/Al$ reference junction for comparison. Note that the features for all Al junctions were only seen at liquid He temperatures.

at 450 mV is sometimes observed. These spectroscopic features were seen in other junctions where one of the films was ferromagnetic, such as $Al/Al_2O_3/Co$, as one can expect.

For comparison, the IETS spectrum for a $Al/Al_2O_3/Al$ reference junction taken at 1 K is shown in an inset in Fig. 1. The peak at $\sim$110 mV in the IETS spectra for $Al/Al_2O_3/Al$ junctions, seen only at liquid helium temperatures, has been identified by others as due to an Al-O stretching mode [19]. The small peak sometimes visible at 450 mV has been attributed to a surface OH^- stretching mode [19]. In the IETS spectra for ferromagnetic junctions, the peak at 450 meV due to the surface OH^- stretching mode is barely visible at low temperatures. The broad peak around 100 mV is not the same as in reference junctions, in that it is also seen at room temperature. Hence, this peak is not due to the Al-O stretching mode alone. Tsui *et al.* [21] have observed a magnon peak in IETS spectra of NiO at 107 MeV. Therefore, part of the peak seen in Fig. 3 at about 100 mV and also the sharp peak at 17 mV can be interpreted as due to magnons generated in the FM electrodes.

The dc bias dependencies of the JMR at 295, 77, and 1 K are shown in Fig. 4 for both polarities of V_{dc} (defined with respect to the Co electrode). A slight dependence on the polarity is observed. As previously reported by us [1], and by others [2,3,8], the JMR decreased monotonically as V_{dc} increases, in the present case by 60% at 0.5 V. Earlier, the corresponding drop in JMR was at least a factor of 10 for "unoptimized" junctions [1,8]. Present good junctions show great improvement. The normalized data shown in Fig. 4(b) is nearly independent of temperature, even near zero dc bias, where G shows the anomaly at low temperatures.

According to the calculations of Bratkovsky [12] and Zhang *et al.* [13], the dc bias dependence of JMR arises from the influence of the applied electric field on the barrier. Increasing V_{dc} increases the overall conductance and, hence, decreases the JMR ratio. The dashed line in Fig. 4(b) is according to Bratkovsky (from Fig. 1 in Ref. [12]). The calculated JMR falls much slower than the experiment, especially at low bias. A bigger decrease in the JMR vs V_{dc} was observed for MgO barriers whose barrier height was in the range of 1 eV (as in the calculation of Zhang *et al.* [13]). Thus, it is important for applications to have a high tunnel barrier.

We propose that part of the large decrease in JMR can be attributed to the excitation of magnons, thereby randomizing the tunneling electron spins and increasing the total conductance. (The energy dependence of the spin polarization due to band structure effects may also reduce the JMR at high bias.) The peaks at 17 and 100 mV in the IETS spectra support this interpretation. In fact, there was little dependence of JMR on V_{dc} below about 15 mV, as shown by the data at 1 K [see inset in Fig. 4(a)]. This suggests a possible gap of this order in the magnon spectra.

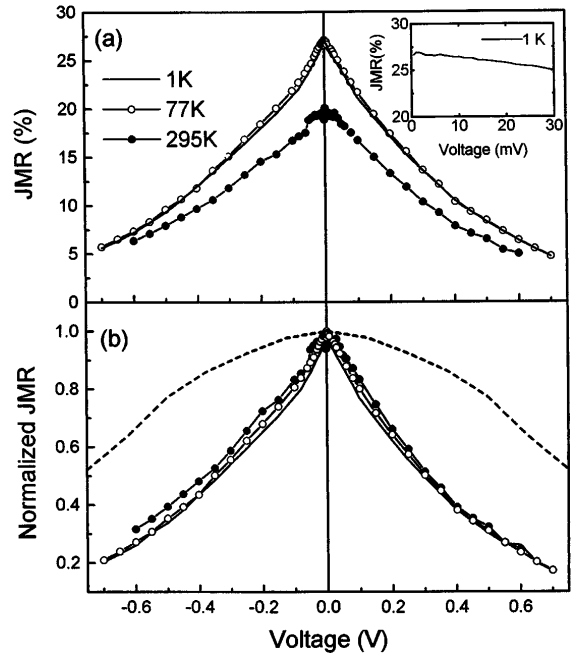

FIG. 4. JMR vs dc bias at three temperatures for the same junction as in Fig. 1. Data shown are (a) the actual percentages and (b) normalized at zero bias. The inset shows the JMR in the low bias region displaying near constancy of JMR. The dashed line in (b) is the theoretically expected variation for a Fe-Al_2O_3-Fe junction with a 3 eV barrier height (from Fig. 1 in Ref. [12]).

For a FM_1-I-FM_2 tunnel junction, Julliere's model [5], based on Stearns's theory [6], gives $JMR = 2P_1P_2/(1 + P_1P_2)$, where P_1 and P_2 are the conduction electron spin polarization measured by tunneling with a superconductor counter electrode as a spin detector [7]. These latter measurements, performed at 0.4 K and near the Fermi level, yielded $P_{Co} = 35\%$ and $P_{NiFe} = 45\%$. These give an expected JMR of 27.2% for a $Co/Al_2O_3/Ni_{80}Fe_{20}$ junction, in perfect agreement with the measured values at 77 and 1 K. However, at 295 K, a JMR of only 20.2% is observed.

It is known from spin wave theory [22] and observed experimentally [23] on clean surfaces in systems such as $FeNiB_{0.5}$, $Ni_{80}Fe_{20}$, and Fe that the temperature dependence of surface magnetization $[M_S(T)]$ follows a $T^{3/2}$ dependence: $M_S(T) = M_S(0)[1 - B_ST^{3/2}]$. The value of B_S is seen up to two or more than the bulk value, depending on the surface cleanliness [23]. In the above ferromagnets, about 15% (or higher for a contaminated surface) decrease in M_S has been observed as T increases from 77 to 300 K, with only $\sim$2% or less change below 77 K. Also, the spin polarization $P(T)$ has been shown to be proportional to M_S [23].

Tunneling electrons come from the top one to two monolayers of the FM and, hence, should reflect surface properties. This has been shown in a series of spin polarized tunneling experiments involving ultrathin FM films near the barrier and a superconducting spin detector [7]. The polarization of these tunneling electrons is, thus, expected to follow the temperature dependence of the surface magnetization. Assuming the polarization to follow a $T^{3/2}$ dependence and Julliere's model, and choosing values for B_S such that it reproduces the ratio of the experimental conductance difference (between parallel and antiparallel configuration) at 77 and 295 K, the JMR at room temperature can be evaluated. This yields an estimate for the JMR value at room temperature of 22.8%, which is in fair agreement with our experimental value of 20.2%. Below 77 K, both M_S and JMR do not change significantly. The lower value of JMR at higher T can thus be attributed to the temperature dependence of the surface magnetization of the FM films. Detailed temperature dependence of JMR will be published separately.

The data published for $NiFeB_{0.5}$ and thin films of $Ni_{80}Fe_{20}$, with even small amounts of surface contamination [23], show a stronger temperature dependence and reduced magnitude of M_S (with T_C below room temperature for $Ni_{80}Fe_{20}$). This might explain the irreproducible, low, or absent JMR at room temperature in many of the reports in the literature, whereas cooling these junctions down to low temperatures sometimes restores the JMR to nearly its expected value [8,24].

In addition to P, the factors that influence the JMR value are the following: (i) the FM/I interface cleanliness, (ii) the barrier quality, and (iii) well defined and separated H_C of the FM electrodes. It is nontrivial to completely oxidize the barrier Al film without oxidizing the FM surface of the bottom FM and also achieve a clean FM/I interface, to reach the full JMR value. Oxidation of the surface near the barrier can affect the JMR by spin scattering due to the strong paramagnetic nature of the magnetic transition metal oxides at higher temperatures. At low temperatures, in the antiferromagnetically ordered state of these oxides, the spin scattering is negligible [25], thus showing a finite JMR in many cases. Also, impurities in the barrier are detrimental to the JMR [12,26].

In conclusion, detailed tunneling studies as a function of temperature and dc bias in well-characterized, good junctions reveal some of the fundamental phenomena in FM-I-FM tunnel junctions. The temperature dependence of JMR has been attributed to the temperature dependence of the surface magnetization of the FM electrodes, also explaining the failure to observe JMR at room temperature in many earlier reports. The dc bias dependence of JMR does not agree with recent theories, especially in the low bias region where magnetic excitations play a role, as shown by the inelastic tunneling spectra.

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