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Temperature dependence of magnetoresistance and surface magnetization in ferromagnetic tunnel junctions

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The temperature dependence of spin-polarized tunneling is investigated between 77 and 420 K for various ferromagnetic tunnel junctions. Both the junction resistance and the magnetoresistance decrease with increasing temperature T. The experimental results are successfully described by a model that includes two current contributions. The dominant one is elastic, spin-polarized tunneling between the two ferromagnetic electrodes, each with an electron polarization P that decreases with T due to thermally excited spin waves according to $P \propto (1-\alpha T^{3/2})$, i.e., in the same way as the surface magnetization. A smaller second conductance is due to assisted, spin-independent tunneling which we find to be proportional to $T^{1.35\pm0.15}$. [S0163-1829(98)50930-7]

The observed magnetoresistance in ferromagnetic tunnel junctions (JMR) has, at low temperatures, reached nearly the optimum values[1,2] expected from Julliere's model.[3] However, even with the best junctions there is a significant decrease in JMR at room temperature (RT) as compared to values at 4.2 or 77 K. Although the strength of the temperature dependence varies, it is present irrespective of the type of tunnel barrier and ferromagnetic electrodes. No satisfactory explanation for the dependence of JMR on temperature (T) has been published so far.

Besides the JMR, it is known that the junction resistance (R_J) itself is also reduced at higher temperatures. This T dependence of R_J is, however, not only found for ferromagnetic junctions, but also for standard junctions with nonmagnetic electrodes. These latter junctions display a drop in resistance of about 25% between 4.2 K and RT, for well-formed Al_2O_3 barriers prepared by oxygen glow discharge.[4–6] This fact suggests a nonmagnetic origin of the R_J vs T behavior. With amorphous barriers such as a-Si and a-Ge, there is a substantially higher change of R_J with temperature.[7] Theoretical understanding of the temperature dependence of tunneling conductance of Al_2O_3 insulators is incomplete, whereas for the case of amorphous barriers, results have successfully been interpreted by using models based on hopping transport.[7,8]

In this paper we will address the temperature variation of JMR and R_J of ferromagnetic tunnel junctions. A natural starting point is the model proposed by Julliere,[3] which we will modify by assuming that in addition to the conductance due to direct elastic tunneling, there is a second conductance G_{SI} present. We take G_{SI} to be unpolarized and therefore not dependent on the relative orientation of the electrode magnetizations. The total conductance is thus expressed as

$$G(\theta) = G_T\{1 + P_1 P_2 \cos(\theta)\} + G_{SI}, \tag{1}$$

where θ is the angle between the magnetization directions of the two electrodes ($\theta = 0°$ or 180° for parallel or antiparallel magnetizations, respectively). P_1 and P_2 denote the effective tunneling electron spin polarizations of the ferromagnets, while G_T is the prefactor for direct elastic tunneling.

With regard to temperature dependence, there are three candidates. First of all, elastic direct tunneling varies slightly with T due to broadening of the Fermi distributions in the electrodes. Theory gives

$$G_T = G_0 CT/\sin(CT), \tag{2}$$

where G_0 is a constant and $C = 1.387\times10^{-4} d/\sqrt{\phi}$, with the barrier width (d) in Å and the barrier height (ϕ) in eV.[4] For typical barrier parameters, G_T at 300 K is only a few percent higher than at $T = 0$ K. A second factor that may vary with T is G_{SI}, in a manner determined by the responsible physical mechanism, which will be discussed later.

Third, we introduce a variation of electrode polarizations P_i with T. Conventionally, values for P are determined from a tunneling measurement at low temperature with a superconductor acting as spin analyzer.[9] Thus the polarization values are only available for $T < 1$ K. It is, however, well established that in the case of alloys, P scales approximately with the magnetic moment of the alloy as its composition is varied.[9] A logical extension of this proportionality is to adopt a polarization P that varies with T as does the magnetization.[10] Such proportionality is also found for other techniques that employ spin-polarized electrons.[11,12] Magnetization versus T has been extensively studied, and is described fairly well by thermal excitation of spin waves for T far below the Curie temperature.[12–14] This produces a term proportional to $T^{3/2}$ in the magnetization, which has been experimentally confirmed for bulk samples, ultrathin films, and surface magnetization as well. The latter is important since tunneling is an extremely surface-sensitive process.[9] For the polarization, we thus can write

$$P(T) = P_0(1 - \alpha T^{3/2}). \tag{3}$$

The material-dependent constant α is different for bulk or surface, and is generally larger for the latter due to surface exchange softening.[14] It has also been observed that both P_0 and α are very sensitive to surface contaminations.[12]

To analyze the properties of ferromagnetic tunnel junctions we use two quantities, the *measured* resistance R_M for parallel magnetization of the electrodes and JMR. Using Eq. (1) these are written as

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$$\mathrm{JMR}=(2P_1P_2)/(1+P_1P_2+G_{SI}/G_T)$$

and

$$R_M=(1/G_T)/(1+P_1P_2+G_{SI}/G_T).$$

Thus, if G_{SI} is present, the JMR is always smaller than the value $(2P_1P_2)/(1+P_1P_2)$ predicted by Julliere. Moreover, notice that the ratio JMR/R_M, which is equivalent to $\Delta G=G(\theta=0)-G(\theta=180)$, does not contain G_{SI}:

$$\Delta G=2G_TP_1P_2. \tag{4}$$

As already noted, the dependence of G_T on T is small and can be calculated from Eq. (2). Therefore, a plot of ΔG vs T directly reflects the T dependence of P_1 and P_2. In addition, one can easily see from Eq. (1) that G_{SI} is given by

$$G_{SI}=\langle G\rangle-G_T, \tag{5}$$

where $\langle G\rangle$ is the conductance averaged over parallel and antiparallel magnetization. This provides a means to extract G_{SI} and its variation with T.

We will now describe in detail the measured temperature dependence of the JMR and R_M between 77 K and ≈400 K, for junctions with Al_2O_3 barriers and a variety of ferromagnetic electrodes. We show that the data can be described very well with the above model, which includes the effect of spin-wave excitations on the electron polarization, and a spin-independent contribution to the tunnel conductance. Data analysis gives P as a function of T for different ferromagnetic electrodes, and allows a complete determination of the spin-independent current.

Ferromagnetic tunnel junctions were fabricated by vacuum evaporation onto cryogenically cooled glass substrates using shadow masks as described in Ref. 1. Ferromagnetic electrodes included Co and $Ni_{80}Fe_{20}$. A glow discharge oxidized Al layer (1.2–1.6 nm) formed the Al_2O_3 tunnel barrier. In the case of symmetric junctions such as $Co/Al_2O_3/Co$ (hereafter referred to as a Co-Co junction), the top electrode was exchange biased by a 40 nm NiO film prepared by reactive evaporation.[15] Overall 30 different junctions with R_M in the range 0.4–10 kΩ and a JMR of ⩾10% at RT were studied using an ac resistance bridge (LR-700). Care was exercised to have junctions that had ferromagnetic films with well-separated coercivities, to obtain complete magnetization reversal at all T.

In Fig. 1, the temperature dependence of R_M and JMR for three representative junctions is shown. Both R_M and JMR decrease with increasing temperature up to a certain critical temperature. Above that, the junction becomes unstable and R_M may go up or down with time depending on the improvement or degradation of the barrier. We consider only the behavior below such a critical temperature, where the junctions are stable. For the junctions in Fig. 1, the temperature dependence is quite significant. For example, for the Co-Co junction, R_M decreases by 17% from 77 to 300 K; simultaneously, the JMR decreases from 21.2% to 17.6%. For both the Co-NiFe junctions the decrease of JMR is stronger than that of the Co-Co junction. Moreover, the two Co-NiFe junctions have slightly different JMR at the lowest T and a somewhat different decay with T, even though the same electrode materials are used. With respect to R_M we note that classical tunneling theory[4] [Eq. (2)] predicts a change in R_M of only 1.5%, for this junction with $d=1.3$ nm and $\phi=3.4$ eV. Therefore, a large discrepancy exists between the experimental observation and the theory based on purely direct tunneling only. This suggests the presence of an additional current mechanism that has a stronger temperature dependence.

In the following, the proposed model is successfully applied to the experimental data. In Fig. 2, we show $\Delta G(T)/$

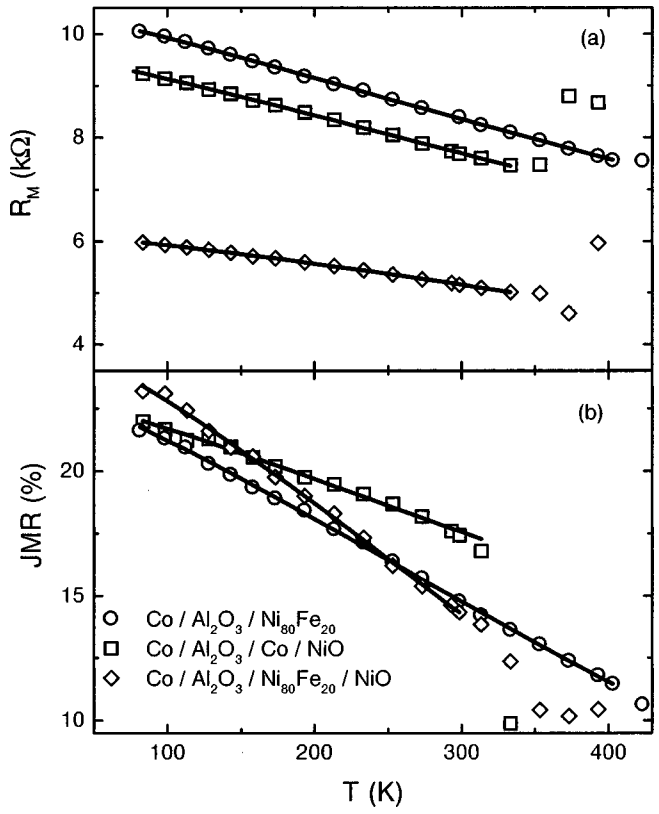

FIG. 1. Temperature dependence of junction resistance (a) and magnetoresistance (b) for three junctions: $Co/Al_2O_3/NiFe$ (circles), $Co/Al_2O_3/Co/NiO$ (squares), and $Co/Al_2O_3/NiFe/NiO$ (diamonds). The solid lines represent the theoretical fits.

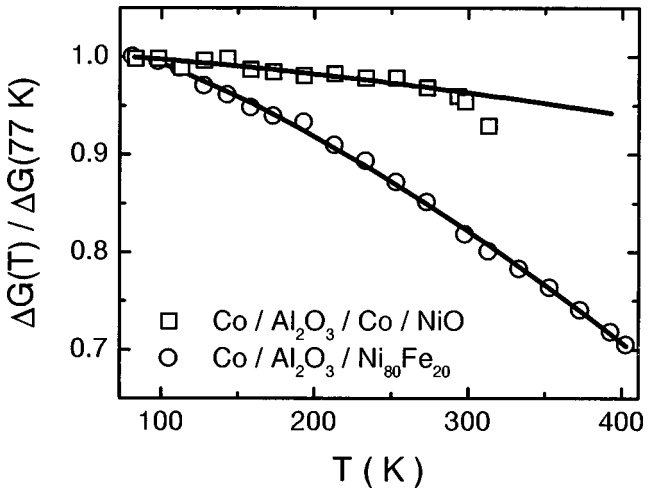

FIG. 2. Temperature dependence of the normalized ΔG for two representative ferromagnetic junctions. The solid lines are the fits to the theory based on thermal spin-wave excitations.

ΔG(77 K) vs T for two representative junctions, each with a 1.4 nm Al layer oxidized to form the barrier. The plot shows that ΔG decreases considerably as T increases. For the $Co/Al_2O_3/Ni_{80}Fe_{20}$ junction, ΔG decreases by approximately 30% as T increases from 77 K to about 400 K, indicating a substantial reduction of P. We remark here that the behavior of ΔG cannot be attributed to the T dependence of G_T [Eq. (2)] since it has the wrong sign. The solid lines in Fig. 2 are fits obtained by using Eqs. (2)–(4), where in Eq. (2) experimentally determined barrier parameters (based on Ref. 4) are used. The fitted curves agree well with the experimental data, except for a few data points at high T. These deviations are due to irreversible changes of the barrier as already mentioned, and, in Co-Co junctions, also due to the loss of the exchange biasing field.[15]

For the Co-Co junction in Fig. 2, ΔG shows a much weaker decay as compared to the other junctions having $Ni_{80}Fe_{20}$ as one electrode. From data for over 10 junctions, we obtain $\alpha_{Co}=1–6\times10^{-6}\,K^{-3/2}$ and $\alpha_{NiFe}=3–5\times10^{-5}\,K^{-3/2}$. Thus, the spin-wave-related reduction of P is approximately an order of magnitude larger for $Ni_{80}Fe_{20}$. This result agrees with the rough trend that spin-wave excitations are less effective in reducing the magnetization for ferromagnets with a higher Curie temperature ($T_C=1360$ K for Co while 850 K for $Ni_{80}Fe_{20}$). The obtained values for α are comparable to those derived from the magnetization measurements. For instance, α for bulk $Ni_{80}Fe_{20}$ is $1.23\times10^{-5}\,K^{-3/2}$.[13] Since the present tunneling experiment probes the electron polarization at the ferromagnet's surface, we are dealing with the T dependence of the surface magnetization, which can have an α more than twice as large as the corresponding bulk value.[12,14] The experiments also show that α for the same material may take different values, which depends largely on the junction interface quality. Higher contamination at the interface can lead to higher α, resulting in a considerable decrease of P with increasing T. This might partially explain many of the previous results on ferromagnetic tunnel junctions.[16] Valuable insight into these phenomena is expected to be obtained from temperature-dependent measurements in ferromagnetic tunnel junctions, complementing other methods for determining surface magnetic properties, while at the same time providing input for theoretical work aimed at relating the tunneling spin polarization to intrinsic properties of the ferromagnetic materials.

Next we consider the spin-independent conductance G_{SI}, which can now completely be determined[17] from Eq. (5). The obtained G_{SI} as a function of T, together with its ratio to the total conductance (G), is illustrated in Figs. 3(a) and 3(b), respectively, for two Co-Co junctions. It is seen that G_{SI} increases monotonically as T is raised. The experimental data were fitted to a power-law $G_{SI}(T)\propto T^{\gamma}$, as given by the solid lines, yielding $\gamma=1.33$. However, some uncertainty in the values of P_0 limits the overall precision with which γ can be determined.[17] Using available data on a number of junctions gives a γ of 1.35 ± 0.15, with $P_0=34\pm2\%$ for Co and $P_0=42\pm3\%$ for $Ni_{80}Fe_{20}$. We conclude that G_{SI} rises much faster with T than the (spin-polarized) direct tunneling does [such that the ratio G_{SI}/G increases as T goes up, see Fig. 3(b)]. This explains the unusually strong reduction of the overall junction resistance, while it causes the JMR to go down with T even faster than due to spin-wave excitations alone. This has probably played a role in many of the earlier results, where a sizable JMR was observed *only* at 4.2 K, and essentially no effect (<2%) was reported at RT.[16]

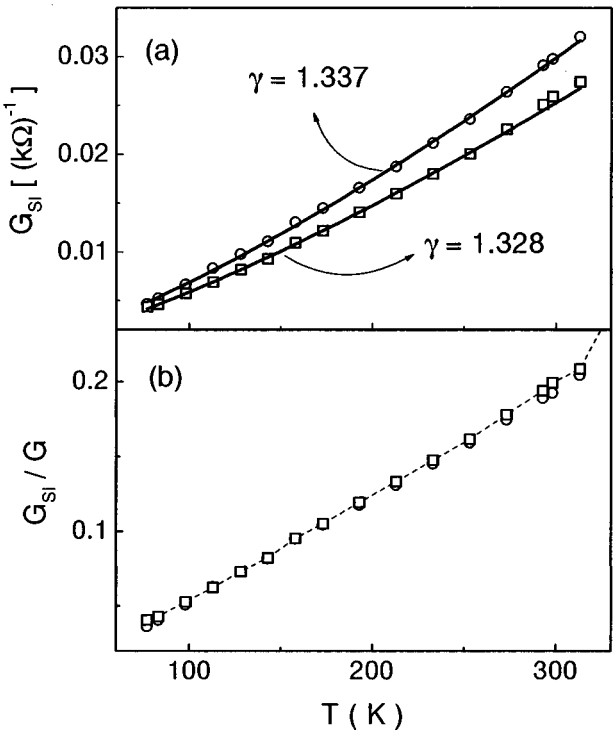

FIG. 3. Temperature dependence of the spin-independent conductance G_{SI} (a) and its relative contribution to the total conductance (b) for two $Co/Al_2O_3/Co/NiO$ junctions. The solid line in Fig. 3(a) is a fit to a power-law T^{γ}.

Several physical mechanisms may be responsible for the spin-independent contribution. First of all, imperfections in the Al_2O_3 barrier may be present in sufficient density to provide a noticeable hopping conductance through the associated localized states. Hopping is known to dominate transport through amorphous Ge or Si barriers[7] and, if responsible for G_{SI} in the present case, it suggests a somewhat amorphous character of the Al_2O_3 insulator. Theoretical work[8] shows that hopping through chains of N localized states should have a power law dependence on T, the exponent being $\gamma(N)=N-[2/(N+1)]$. The temperature dependence originates from phonon emission or absorption at the transition from the first to the next localized state along the chain, to overcome the energy difference between the two levels. Elastic tunneling is, however, assumed between the first and last state on the one hand, and the electrodes on the other hand. For $N=1$ this leads to $\gamma=0$, but for $N=2$ we have $\gamma=4/3$, surprisingly close to our experimental finding.

Yet, we stress that other processes have to be considered. For instance, some ions of the electrodes may be misplaced in the Al_2O_3, producing states near the barrier interfaces. Alternatively, nonuniformity in the Al layer possibly leaves pinholes and/or causes oxidation of the bottom electrode. Magnetic oxides often have poorer insulating properties, or are semiconducting. For the resulting extra conductance, the expected temperature dependence is not well known.

The above-mentioned mechanisms for G_{SI} do not rely on the ferromagnetic nature of the electrodes. This explains why

junctions with nonmagnetic electrodes[4–6] also display a 15 to 25% drop in resistance between 4.2 and 300 K. In contrast, the emission or absorption of spin waves by tunneling electrons, as previously proposed[1,5] and recently calculated by Zhang *et al.*[18] in an attempt to explain the bias and temperature dependence of JMR, can only explain the variation of resistance for *magnetic* tunnel junctions. Moreover, spin-wave-assisted tunneling is an inelastic, higher order process with an associated tunneling matrix element that is smaller than that for direct elastic tunneling.[18] In our first order description presented here, the tunnel current is simply drawn from a reservoir of electrons whose polarization decreases with T, as it follows the (surface) magnetization.

In conclusion, both junction resistance and magnetoresistance of ferromagnetic tunnel junctions were found to decrease with increasing T. The results are successfully described by a simple model that contains two contributions to the tunnel conductance. The dominant one is due to direct elastic tunneling, with the tunneling electron polarization P decreasing as $1-\alpha T^{3/2}$. The existence of a second, spin-independent conductance was invoked to explain the variation of the junction resistance with T, while it also adds to the reduction of JMR with T. Temperature-dependent measurements in ferromagnetic tunnel junctions can thus be used to study the surface properties of ferromagnetic materials.

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[17] Extraction of G_{SI} requires G_T to be known [see Eq. (5)], which can be determined from Eq. (4) if the polarizations P_i at $T\approx 0$ are known. These can in principle be obtained from the JMR value at T low enough such that G_{SI} can be neglected. In the present work, the lowest T attained was 81 K, at which the above condition is not entirely fulfilled. This introduces a small uncertainty in G_T and consequently also in G_{SI}.

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