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Resonant tunneling in magnetoresistive Ni/NiO/Co nanowire junctions

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Magnetotransport studies performed on electrodeposited Ni/NiO/Co nanojunctions show a broad distribution of magnetoresistance values spanning from +40% to −25%, with an average of about 2%, corresponding to observations on large-area junctions. The dispersion in the results can be understood in terms of tunneling via localized states in the barrier. Calculations based on Landauer–Büttiker theory explain this behavior in terms of disorder-driven statistical variations in magnetoresistance with a finite probability of the inversion of tunnel magnetoresistance sign due to resonant tunneling. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1558657]

Magnetic tunnel junctions (MTJs) made of two ferromagnetic electrodes separated by an insulating spacer layer have aroused considerable interest due to potential applications in spin-electronic devices such as magnetic sensors and magnetic random access memories. Functioning of these devices is controlled by the phenomenon of tunneling magnetoresistance (TMR), where the tunneling current is modified when magnetizations of the two ferromagnetic layers change their relative alignment (for a review on TMR see Ref. 1). Within the simplest model, the magnitude of TMR is determined solely by the spin polarization (SP) of the density of electronic states at the Fermi energy of the two ferromagnets, P_1 and P_2,[2,3] so that

$$\mathrm{TMR} \equiv \frac{G_P - G_{\mathrm{AP}}}{G_P + G_{\mathrm{AP}}} = P_1 P_2 . \tag{1}$$

Here G_P and G_{AP} are the conductance for the parallel and antiparallel alignment of the MTJ, and we use the definition of TMR given in Ref. 3, which has the advantages of symmetry and simplicity. If both ferromagnets have the same sign of the SP, the conductance is larger when the two magnetic layers are aligned parallel. This is what is generally observed[1] and referred as the normal (positive) sign of TMR.

Recently, however, it was found that it is possible to invert the sign of the SP of tunneling electrons from Co by using a $SrTiO_3$ barrier instead of standard Al_2O_3.[4] The change in sign of the SP observed in these experiments was attributed to the effect of bonding at the ferromagnet/barrier interface that had been earlier predicted theoretically by Tsymbal and Pettifor.[5] The same mechanism was put forward to explain positive and negative values of TMR depending on the applied voltage in MTJs with Ta_2O_5 and Ta_2O_5/Al_2O_3 barriers[6] and to elucidate the inversion of TMR observed in Co-contacted multiwalled carbon nanotubes.[7]

Here we present experiments on junctions of a sufficiently small area to reveal effects driven by localized electronic states in the barrier formed due to embedded impurities or intrinsic defects. Conventional dc measurements of nanowire junctions grown by electrodeposition with a cross section ranging of 3×10^{-3}–8×10^{-3} μm^2 display two-level fluctuations of the electric current which indicate an impurity/defect-driven transport.[8] By performing measurements on a large number of samples we get access to the statistics of TMR, revealing a broad distribution of TMR values. We explain this behavior in terms of tunneling via localized states in the barrier, which under resonant conditions leads to a change in sign of the tunneling spin polarization and the inversion of TMR. Our results indicate that although disorder is detrimental for TMR in samples of large area due to averaging over a large number of defect/impurity states,[9] it can lead to a new phenomenon of resonant inversion of TMR in samples of small area.

Samples were synthesized using electrochemical techniques. Polyester track-etched membranes, 6 ± 1 μm thick, with cylindrical holes of 80 ± 20 nm in diameter, were used as templates. A gold back layer was sputtered on one side of the membranes, and was used as contacting working electrode. On the other side of the membrane, a gold contact, sputtered prior to electrodeposition, served as an indicator for interrupting the wire growth before multiple wires are connected.[10] This is a reliable method allowing a single wire to be connected. A standard electroplating Watt's bath was used to fill half of the membrane thickness with Ni (pH 3.7). Anodization of Ni was performed in 0.075 M Na_3BO_3 and 0.3 M $H_2B_4O_7$ (pH 8.4). Characterization of the dielectric layer properties has been made by means of impedance spectroscopy. The estimated thickness is found to be about 1.5 nm.[11] Mott–Schottky analysis reveals the presence of p-type impurity with concentration about N_a

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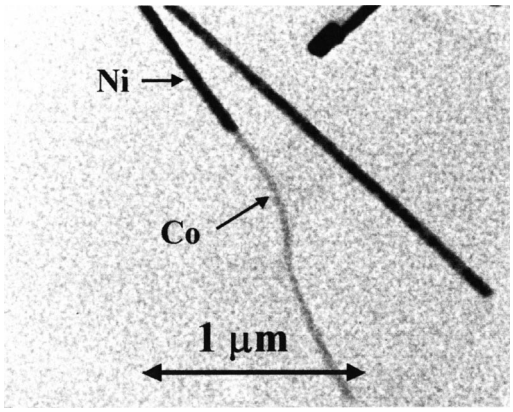

FIG. 1. Transmission electron microscope of the nanowires after dissolution of the matrix. Local x-ray fluorescence spectroscopy revealed the Ni and Co components.

$=10^{25}$ m^{-3}, somewhat lower then previously reported.[12] The top ferromagnet was made by Co electrodeposition in a nonaqueous bath, avoiding the dissolution of the oxide film. Details of the procedure were published elsewhere.[8,11] The transmission electron microscopy image of nanowires after dissolution of the membrane revealed the presence of Ni and Co (Fig. 1).

Samples with smaller resistance than 50 kΩ and larger than 10 MΩ were discarded in order to avoid shorts and nonreproducible measurements. Electrical properties of the Ni/NiO/Co nanowires were investigated at low temperatures (1.6–5 K), using dc measurements. We made over 200 samples, from which more than 60 were fulfilling the required resistance range conditions. The observed distribution of TMR (Fig. 2) are presented on two scales. One is the TMR defined according to Eq. (1), and the other is the commonly used magnetoresistance (MR) ratio $(R_{\mathrm{AP}}-R_P)/R_P$, where R_P and R_{AP} are the resistance for the parallel and antiparallel alignment, respectively. As is evident from Fig. 2(a), the measured distribution is very broad spanning the TMR values from +0.2 to −0.1 (from +40% to −25%). These results are very different from those obtained on large area Ni/NiO/Co MTJs,[13,14] which showed small positive TMR values of 2% or less.

Figure 3 displays magnetoresistance curves measured for the samples displaying the largest positive (a), small positive (b), and largest negative (c) values of TMR. The sharpness of the magnetoresistance curves with resistance changes occurring within a few Oe, confirms unambiguously that a single wire dominates in our measurements. If two or more wires are measured in parallel, we expect to observe several steps in the MR curve, corresponding to different magnetic switching fields of different wires.[14] Multiple jumps in the MR curve might however be hindered by the measurement noise for samples with low TMR values (less than 0.01). This is indicated in the histogram of Fig. 2(a) by the unshaded bar.

The observation of a telegraph noise at large bias (50 mV or more, corresponding to currents of 100 nA or more), due to trapping and untrapping of single electrons at an impurity site,[8] reveals that the current flows though a limited area. The presented measurements at low bias and low temperatures did not show fluctuations larger than a few percent for our dc measurements (at rates between 1 mHz and 10 Hz). We can estimate that a single junction contains several tens of impurities from our impedance spectroscopy measurements at room temperature. The intrinsically rough sur-

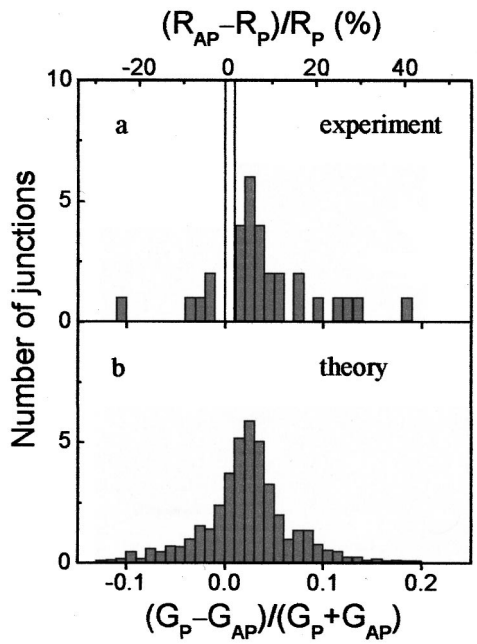

FIG. 2. (a) Experimental distribution of TMR values in magnetic Ni/NiO/Co nanojunctions measured at 4.2 K. The vertical scale is cut at $N=10$ (at the highest peak $N=33$). The unshaded bar indicates a possible contribution from samples with multiple junctions. (b) Calculated normalized distribution of TMR values for $\gamma=4\beta$ and $\delta=0.015\beta$.

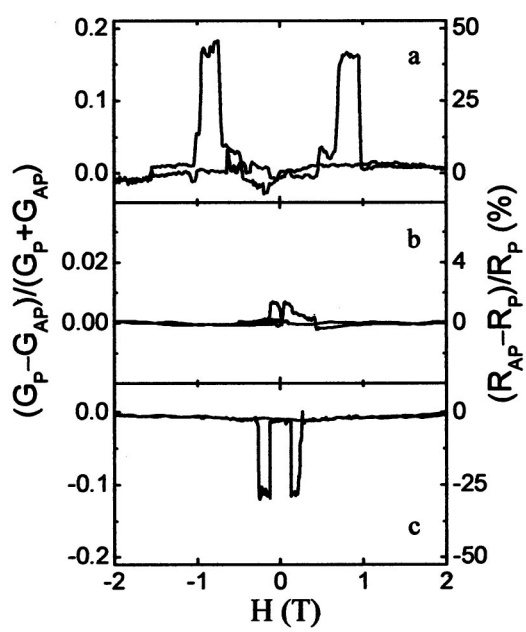

FIG. 3. Magnetoresistance curves measured at 1.6 K, showing the highest magnitude of TMR observed (a), corresponding to a TMR ratio of 0.17 (40% using the standard definition of TMR), a small magnitude TMR (b) with a scale magnified by a factor 5, and the largest negative magnitude of TMR (c), corresponding to a TMR ratio of −0.11 (−25% using the standard definition of TMR).

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faces of the nanowires[11] make a current flow through an area significantly smaller than the wire section likely.

An interesting phenomenon, which follows from our measurements, is the inversion of TMR observed on a number of samples [see Fig. 2(a)]. In order to elucidate this effect, we consider a simple one-dimensional picture of tunneling via an impurity state in the barrier. The conductance per spin as a function of energy E is given by[15]

$$G=\frac{4e^2}{h}\frac{\Gamma_1\Gamma_2}{(E-E_r)^2+(\Gamma_1+\Gamma_2)^2}, \tag{2}$$

where E_r is the energy of the resonant state and Γ_1 and Γ_2 are leak rates of an electron from the impurity state to the left and right electrodes. We assume for simplicity that the latter are proportional to the densities of states of the electrodes, ρ_1 and ρ_2, at the left and right interfaces, so that $\Gamma_1\propto\rho_1\exp[-2\kappa x]$ and $\Gamma_2\propto\rho_2\exp[-2\kappa(d-x)]$, where κ is the decay constant and x is the position of the impurity within the barrier of thickness d. Off resonance, when $|E-E_r|\gg\Gamma_1+\Gamma_2$, the latter assumption implies that the spin conductance is given by $G\propto\rho_1\rho_2$. When tunneling occurs between ferromagnetic electrodes this leads to TMR, which is given by Eq. (1) with $P_{1,2}=(\rho_{1,2}^{\uparrow}-\rho_{1,2}^{\downarrow})/(\rho_{1,2}^{\uparrow}+\rho_{1,2}^{\downarrow})$. At resonance, when $E-E_r=0$, the situation is different. Assuming for simplicity an asymmetric position of impurity we obtain from Eq. (2) that $G\propto\rho_2/\rho_1$, if $x<d/2$ and hence $\Gamma_1\gg\Gamma_2$, and we obtain that $G\propto\rho_1/\rho_2$, if $x>d/2$ and hence $\Gamma_1\ll\Gamma_2$. In both cases, the conductance is inversely proportional to the density of states of one of the ferromagnets that results in the sign inversion

$$\mathrm{TMR}=-P_1P_2. \tag{3}$$

We see, therefore, that the resonant tunneling leads to the inversion of TMR, which originates from the spin-dependent leak rates that invert the effective SP of the one of the ferromagnetic electrodes.

The occurrence of the normal and inverse TMR is controlled by statistical properties of disorder configurations in the nanojunctions. In order to study these properties in more detail we have performed calculations of TMR using the Landauer–Büttiker theory[16] including inelastic scattering.[17] We used a single-band tight-binding model within a simple cubic geometry. The on-site atomic energies of the barrier atoms are set equal to 7β, where β is the hopping integral, which provides no states at the Fermi energy, $E_F=0$, for the perfect structure. Disorder is introduced as a random variation of the on-site atomic energies with a uniform distribution of width γ. This disorder broadens the conduction band creating localized states within the band gap of the insulator. The influence of the electrodes is taken into account using spin-dependent self energies $\Sigma_{1,2}^{\uparrow,\downarrow}$, which are parameterized to the densities of states of the electrodes, $\Sigma_{1,2}^{\uparrow,\downarrow}=-i\pi\beta^2\rho_{1,2}^{\uparrow,\downarrow}$, in the spirit of the model used in Ref. 18. This allows introducing the spin polarizations of the electrodes, which in the calculations are taken to be $P=P_1=P_2=0.6$, a representative value characterizing Co and Ni ferromagnets.[19] Inelastic scattering is introduced by connecting each atomic site of the structure to ''scattering'' electrodes that serve as phase-breaking scatterers.[17] In the calculations the self energies of the scattering electrodes Σ_S are parameterized so that $\Sigma_S=i\delta$, where δ is a parameter.

With increasing disorder parameter γ the distribution broadens, resulting in the inversion of TMR when the localized states start to appear at the Fermi energy. Inelastic scattering narrows the distribution shifting the histogram maximum toward zero and making the distribution more symmetrical. The distribution of TMR, which provides the best fit to the experimental data of Fig. 2(a), is shown in the histogram of Fig. 2(b). This fit gives an estimate for the energy dispersion of defect states, and the magnitude of inelastic scattering. We note that, as is evident from the histograms, the median value of the distribution is a small positive value. This is consistent with experiments performed on large-area Ni/NiO/Co samples that demonstrate small TMR values less than 2%,[13,14] of the same order of magnitude as the average value for our nanojunctions.

In conclusion, we have shown that studies performed on electrodeposited Ni/NiO/Co nanojunctions reveal the important role of localized states in the barrier, which can invert TMR. This phenomenon is explained in terms of disorder-driven statistical variations in TMR with a finite probability of inversion due to resonant tunneling. Our results demonstrate that the specifics of atomic arrangement in magnetic nanojunctions have a considerable impact on spin-dependent transport.

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