Quantum Well States in Spin-Dependent Tunnel Structures

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The magnetotransport behavior of magnetic tunnel junctions with a nonmagnetic interface layer has been studied. The initial effect of the added layer is to reduce the magnetoresistance effect. Also, the bias voltage dependence of the magnetoresistance becomes increasingly more asymmetric. The dependence of the magnetoresistance both on the thickness of the interface layer as well as on the bias voltage can be interpreted as signatures of the development of quantum well states.

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Ever since the first spin polarized tunneling (SPT) experiments were performed [1], this technique has richly contributed to the understanding of thin-film superconductivity and the behavior of magnetic films down to the submonolayer regime [2]. The latest development is the observation of spin-dependent tunneling between two ferromagnetic (FM) electrodes [3–6], the large junction magnetoresistance (JMR) of which has attracted much interest due to the possible application of these FM-I-FM trilayer structures (where I is the insulating tunnel barrier) as sensors and nonvolatile memory elements. From a fundamental viewpoint JMR offers the exciting possibility of studying tunneling electron spin polarizations of various FM materials at ambient conditions, and their temperature dependence [6,7], without the need for a superconducting detector and liquid helium temperatures as required by the earlier experiments.

In this Letter, the spin polarized tunneling phenomenon in the presence of an ultrathin nonmagnetic metal (NM) layer at the FM-I interface in FM-I-FM tunnel junctions is carefully explored. Our experimental studies show in some cases a negative JMR and an unexpected bias voltage dependence. Recent theoretical calculations by Vedyayev *et al.* [8] and Zhang *et al.* [9] have predicted oscillations of the JMR in FM-NM-I-NM-FM systems as a function of the thickness of the NM layer. These calculations show that the interface layer behaves as a quantum well, leading to the formation of quantum well states (QWS) when a resonance condition is fulfilled. The occurrence of a QWS at the Fermi energy results in an increase of the JMR. Moreover, for an asymmetric structure, such as FM-NM-I-FM, the sign of the JMR is predicted to oscillate as a function of the NM layer thickness. These calculations were performed for low bias voltages. We carried out experiments in a search for these quantum effects and extended the calculations to include the bias voltage dependence.

Ferromagnetic tunnel junctions were prepared by thermal evaporation through shadow masks, as described in previous publications [5]. Onto liquid nitrogen cooled glass substrates, Co bottom electrode strips were prepared, half of which were covered by six different thicknesses of Au before the 1.4 nm thick barrier Al film was deposited. After forming the oxide barrier at room temperature by glow discharge oxidation of the Al film, the top electrode of $Ni_{80}Fe_{20}$ film was deposited. Half of the resulting junctions were $Co/Au/Al_2O_3/Ni_{80}Fe_{20}$, whereas the other half were $Co/Al_2O_3/Ni_{80}Fe_{20}$ control samples. The Au thickness ranged from 0.1 to 1.2 nm, with six junctions for each Au thickness. Co and $Ni_{80}Fe_{20}$ films were grown in an applied magnetic field to obtain well defined and sufficiently different coercive fields such that clear parallel and antiparallel magnetization states of the tunnel junctions resulted. Between the 1 mm wide top and bottom electrodes, an 8 nm thick Al_2O_3 layer was deposited to cover the sides of the strips, leaving only a small junction area. (This procedure helped in the stability of the junctions and sustained bias voltages up to $|V_{dc}| \geq 2$ V.) Junction resistances (R_J) were below 25 kΩ and measured in a four-terminal geometry using an LR-700 ac resistance bridge as well as by dc techniques. The latter method was used to obtain the bias voltage dependence of tunnel current, conductance, and JMR (defined as the percentage change in R_J in an applied field with respect to its peak resistance value). The measurements were carried out at 295 and 77 K.

In Fig. 1, the junction resistance is plotted as a function of the applied magnetic field for a control junction and for junctions with an 0.3, 0.6, and 0.7 nm thick Au interface layer. Large JMR is seen for the control junction as expected. However, with 0.3 nm Au over Co, a considerable drop in JMR is observed, showing only 4.9% JMR as compared to 18.9% with no Au. But with both 0.6 and 0.7 nm Au, a dramatic change is observed; the JMR becomes negative. *This is the first case where a negative JMR has been seen in magnetic tunnel junctions* [10]. The JMR dip occurs at exactly the same field range as the peak for zero or low Au coverage. This effect is

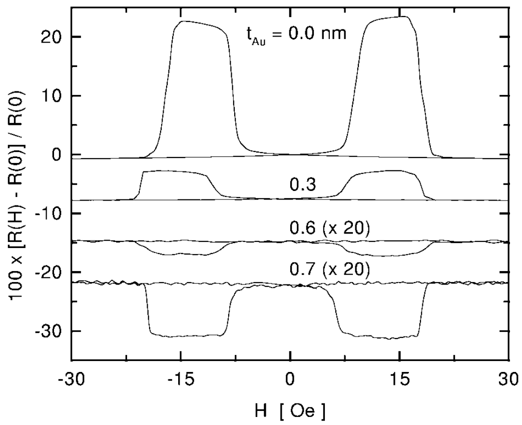

FIG. 1. The JMR at zero bias voltage and 77 K of control junction and junctions with 0.3, 0.6, and 0.7 nm thick Au interface layers. The curves are offset for clarity. The curves for 0.6 and 0.7 nm thickness are magnified 20×.

stable and reproducible even after storing the sample for 15 months in ambient conditions.

The negative JMR is observed at 295 and 77 K for Au coverage of 0.6–0.8 nm. Beyond ~0.9 nm Au, the JMR is immeasurably small (JMR $< 0.01\%$) and remains so up to 1.2 nm Au, the highest Au thickness studied. The JMR as a function of Au thickness is shown in Fig. 2. The big drop in JMR as a function of the metal thickness in the range shown here was also observed for other elements including Cu, Cr, Pd, and Ag. One of the junctions with 0.4 nm Cu at the interface also showed a negative JMR of 1.7%. Otherwise, only positive or no JMR was seen.

In general, as reported before [6,11], for a FM-I-FM junction the JMR decreases monotonically as a function of bias voltage (see $t_{\mathrm{Au}} = 0.0$ nm data in Fig. 3). For the present control junctions, the JMR decreased to a few percent at $|V_{\mathrm{dc}}| \sim 0.7$ V and in some cases the junctions withstood $|V_{\mathrm{dc}}| \geq 1.8$ V bias, still showing a measurable JMR. Asymmetry in the bias dependence of the JMR was always observed, as well as in the *I-V* data, as is generally the case for dissimilar electrode materials.

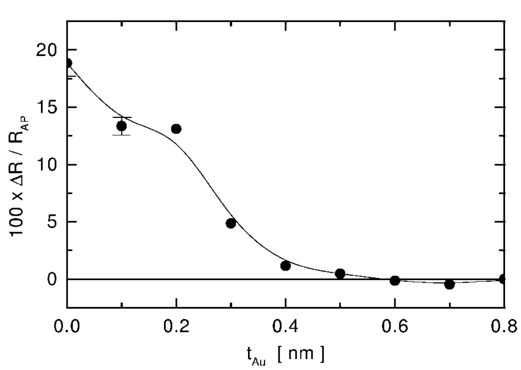

FIG. 2. Dependence of the JMR at zero bias voltage and 77 K on the thickness of Au interface layer. Error bars indicate junction to junction scatter of the JMR values.

These *I-V* measurements yielded average barrier heights above 2.5 eV and thicknesses in the range of 1.1–1.3 nm for the junctions, by fitting to Brinkman's formula [12].

The bias voltage dependencies of the JMR for various junctions with increasing Au at the interface are also shown in Fig. 3. An asymmetrically decreasing JMR with bias voltage is found for Au thickness up to 0.4 nm, see Fig. 3(a). But for Au coverage of 0.5 nm and beyond, the bias voltage dependence is dramatically different; the JMR changes sign and the shape changes [see Fig. 3(b)]. For instance, with 0.5 nm Au, the JMR slightly increases initially before showing a decrease with positive bias (Co film positive), whereas for the reverse bias it is negative and nearly constant beyond about -0.3 V. For 0.6 and 0.7 nm Au, the JMR is negative even at $V = 0$ V. With positive bias it becomes less negative and changes sign again between 0.2 and 0.4 V for these two thicknesses. There is a broad peak in the JMR between 0.5 and 0.7 V and it crosses the axis once again at higher voltages. With reverse bias, the JMR continues to be negative, with a broad dip between -0.2 and -0.4 V. The position of the peak or dip is independent of the temperature between 295 and 77 K, even though the magnitude of the JMR changes. A very small ($<0.05\%$) negative JMR was observed for 0.8 nm Au.

A number of phenomena related to the Co/Au interface might influence the JMR. As a first possible explanation, Moodera *et al.* [13], using a superconducting Al film as a spin detector, have reported direct measurements of the tunneling electron spin polarization for Fe/Au/Al_2O_3/Al

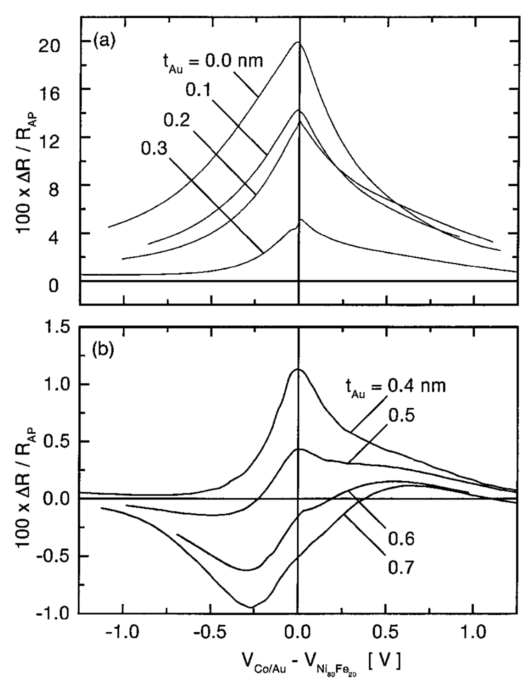

FIG. 3. Dependence of the JMR on bias voltage for increasing thicknesses of the Au interface layer at $T = 77$ K: (a) $t_{\mathrm{NM}} \leq$ 0.3 nm and (b) $t_{\mathrm{NM}} \geq 0.4$ nm.

junctions. The polarization showed a steep decrease with Au coverage, from a value of 44% without Au to ~3% for about 0.4 nm of Au, and continued to drop with further increase of Au thickness. A similar reduction of the Co polarization with 0.4 nm added Au is not inconsistent with the present experiments in the Au thickness range of 0–0.4 nm. However, the polarization results cannot explain the negative JMR and distinctive bias voltage dependence of the present results. As similar behavior is found for several elements, also band structure effects (beyond the effect of QWS formation, see below) do not seem the main explanation.

In the remainder of this Letter, we will show that QWS can qualitatively explain the features of the observed effects. Results from numerical calculations are presented, based on a model first proposed by Slonczewski [14], including recent extensions [15]. Within the model, the transmission probabilities for electrons are calculated by solving the Schrödinger equation for free, noninteracting electrons in a potential energy profile as defined in Fig. 4. Within the constraints of the model assumptions the calculation is exact. Although this simplified model is notoriously inaccurate in predicting JMR values, it correctly predicts the key observations.

The configuration used in the calculations, see Fig. 4, consists of two identical ferromagnetic metal layers (FM_1 and FM_2), the barrier layer (I), and a nonmagnetic metal layer (NM) located between I and FM_2. For both FM layers the Fermi energy is chosen to correspond with that of the well-known spin-split free-electron-like itinerant d-electron bands of Fe [16], i.e., $E_{F,FMi} = 2.62 + (-)1.96$ eV for the majority (minority) spin electrons. A typical symmetric barrier height and barrier width are chosen, $\phi_{bar} = 2.5$ eV and $t_{bar} = 1.5$ nm, respectively. The electronic parameters of the NM metal of thickness t_{NM} are chosen to correspond to those of Au [17], i.e., $E_{F,NM} = 5.51$ eV. All electrons are assumed to have free electron mass and the calculations are performed for zero temperature. In analogy with the experiments, the sign of the bias voltage is defined as that of the FM_2/NM electrode with the FM_1 electrode as common.

Figure 5 shows the calculated JMR for increasing t_{NM}. Some qualitative observations can be made, independent of the details of the chosen parameters [8,9]. The JMR rapidly oscillates between positive and negative values due to QWS which develop when the round trip phase accumulation equals 2π. The period equals $\pi/k_{F,NM}$, with $k_{F,NM}$ the Fermi wave vector inside the NM layer. The envelope of the short period oscillations approaches a nonzero value (different from the value without NM layer) in a damped oscillatory way for thicknesses $t_{NM} >$ 50 nm. The persistence of the oscillations up to very large thicknesses results from the strong forward filtering effect of the barrier (only perpendicularly incident electrons contribute to the conductance). We note that the breaking of phase coherence in a thick interface layer results in quenching of the JMR [8,18].

As in the experiment t_{NM} changes in monolayer steps only, Fig. 5 also shows intersections for several monolayer thicknesses $t_{ML,NM}$. This aliasing effect [19] results in oscillations with a much longer period. For a monolayer thickness of 0.2355 nm, corresponding to the perpendicular bulk lattice parameter of Au⟨111⟩, the JMR initially increases. However, with a suitably chosen ratio between the monolayer thickness and the Fermi wavelength inside the NM layer, which within certain limits may be realized in thin films, the model calculations can reproduce the experimentally observed initial fast decrease of the JMR in a surprisingly quantitative way.

In Fig. 6 the calculated bias voltage dependence is shown for two monolayer thicknesses. A fast drop of the JMR is obtained with increasing NM layer thickness. Without the NM layer at the interface, the bias voltage

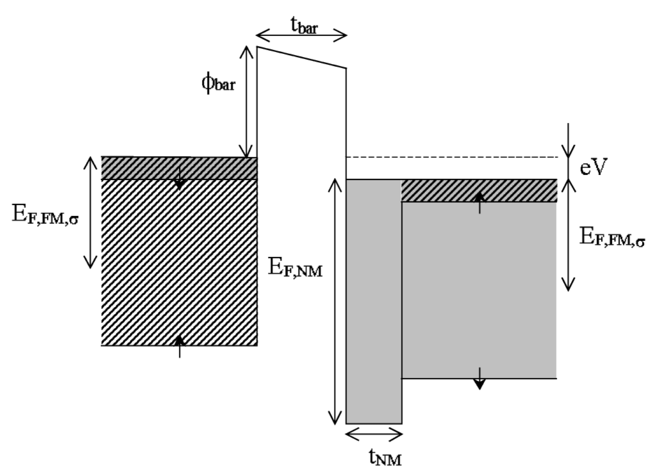

FIG. 4. The potential energy landscape of the tunnel junctions with an additional nonmagnetic metal layer on one side of the barrier in the antiparallel state. The sign of the bias voltage is defined with the right electrode being positive.

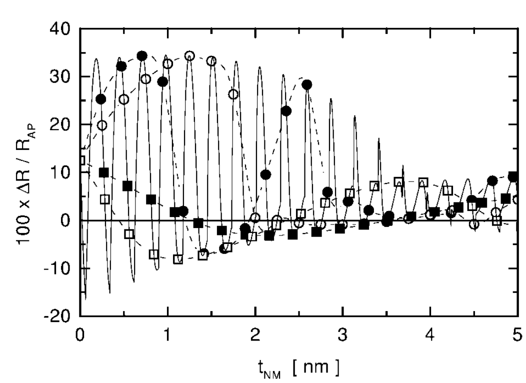

FIG. 5. Calculated nonmagnetic metal layer thickness dependence of the JMR at zero bias voltage, using $E_{F,FMi} = 2.62 \pm 1.96$ eV, $E_{F,NM} = 5.51$ eV, $\phi_{bar} = 2.5$ eV, and $t_{bar} = 1.5$ nm. The continuous variation is intersected at full monolayer coverages (dashed lines): (•) $t_{ML,NM} = 0.2355$ nm (Au⟨111⟩), (∘) 0.25 nm, (■) 0.27 nm, and (□) 0.28 nm.

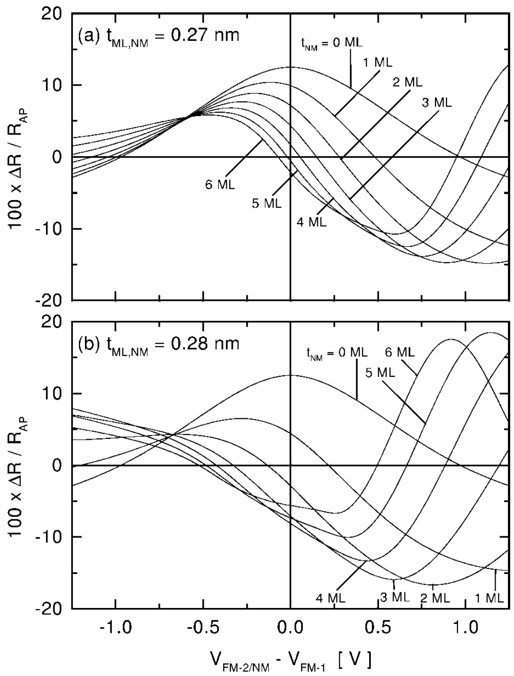

FIG. 6. Calculated bias voltage dependence of the JMR using the same parameters as in Fig. 5: (a) $t_{\mathrm{ML,NM}} = 0.27$ nm and (b) 0.28 nm.

dependence is symmetric around zero bias voltage and similar to previous model calculations [20]. When t_{NM} increases, an asymmetry develops which describes several of the key features observed in the experimental data, such as the (multiple) zero crossings. Even the voltage scale at which the features occur is reproduced surprisingly well. Also the effect of the bias voltage can be interpreted in terms of QWS, i.e., changes in the round trip phase accumulation. However, most remarkably, the sign of the bias voltage seems to be inverted, which is still not understood. One possibility is the formation of the compound Al_2-Au, called "purple plague," during deposition. During the oxidation of this compound, the Au might remain on top of the forming oxide as a surfactant, effectively reversing the measuring geometry.

In conclusion, we have shown that the addition of a nonmagnetic metal interface layer causes a sharp reduction of the junction magnetoresistance, which can even become negative, and an unusual bias voltage dependence. Apart from the sign of the applied bias voltage, surprising good quantitative agreement could be obtained in simple model calculations, using realistic parameters. This study shows that the properties of magnetic tunnel junctions are not solely determined by the properties of the interfaces between the insulator and the electrodes. In a next stage, electrodes may be engineered such that a stronger spin filtering is obtained, for example, by using a FM/NM/FM trilayer electrode, with suitably chosen layer thicknesses.

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