Spin-Dependent Resonant Tunneling through Quantum-Well States in Magnetic Metallic Thin Films

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Quantum-well (QW) states in *nonmagnetic* metal films between magnetic layers are known to be important in spin-dependent transport, but QW states in *magnetic* films remains elusive. Here we identify the conditions for resonant tunneling through QW states in magnetic films and report first principles calculations of Fe/MgO/FeO/Fe/Cr and Co/MgO/Fe/Cr. We show that, at resonance, the current increases by 1 to 2 orders of magnitude. The tunneling magnetoresistance ratio is much larger than in simple spin tunnel junctions and is positive (negative) for majority- (minority-) spin resonances, with a large asymmetry between positive and negative biases. The results can serve as a basis for novel spintronic devices.

Metallic quantum-well (QW) states have long been known to play an important role in spin-dependent transport. Recently, Yuasa *et al.* [1] observed oscillations of the tunneling magnetoresistance ratio (TMR) in Co/Cu/Al-O/Ni-Fe tunnel junctions as a function of the thickness of the Cu layer, demonstrating the effect of quantum confinement of electrons in the nonmagnetic Cu layer. Most studies, however, have focused on QW states in a *nonmagnetic* layer incorporated in a magnetic multilayer structure. In these nonmagnetic layers, the electron energy dispersion is close to that of a free electron, QW states can easily be identified, and simple models can be readily applied. There exists only one report of an experimental attempt to explore spin-dependent transport through confined *magnetic* thin films. In 2002, Nagahama *et al.* [2] reported indications of oscillations in the tunneling conductance of a (100) Cr/Fe/Al-O/FeCo junction, where QW states were expected to be present in the magnetic Fe layer. The observed effect was very small, seen only in the second derivative of the current-voltage curve. Yuasa *et al.* [1] attributed the smallness of the effect to the short mean-free path and spin diffusion length in Fe. Resonant tunneling through QW states in magnetic thin films is worth exploring further, however. In addition to the intrinsic interest, resonant tunneling through magnetic layers can be switched on and off by external magnetic fields, which offers additional control in designing spintronics devices.

In this Letter, we first discuss factors that may have contributed to the smallness of the effect observed in Ref. [2]. We then identify criteria which, when met by candidate structures containing a magnetic thin film, would ensure the formation of sharp QW states in the magnetic film and result in strong resonant tunneling. We use these criteria to select specific structures for a detailed theoretical study. We report results of first principles calculations of spin-dependent transport through select structures [bcc (100) Fe/MgO/FeO/Fe/Cr and Co/MgO/Fe/Cr with epitaxial lattices]. We demonstrate the existence of sharp QW states in the thin Fe layer and that resonant tunneling occurs through these states. At resonance, the current increases by 1 to 2 orders of magnitude and the TMR increases by several orders of magnitude. The resonances are asymmetric with respect to positive or negative biases. By analyzing the wave functions of QW states, we demonstrate that the series of resonances observed in Ref. [2] are from *different* QW states that sequentially enter and leave the energy window under analysis. Finally, we show that a previously unknown QW state in the Fe minority-spin channel contributes to a large and negative TMR.

We start with a discussion of the experimental data of Ref. [2]. A short spin mean-free path and diffusion length in the Fe film is a possible cause for the smallness of the observed effect, as suggested in Ref. [1]. Indeed, it has been found [3] that in thin Fe(100) films grown epitaxially on a GaAs substrate, the majority-spin mean-free path is about 1.1 nm. The resonance effects based on majority-spin QW states would then be limited to Fe films that are thinner than about 1 nm. However, the minority-spin mean-free path is much larger, about 14 nm. If we assume that the spin diffusion length is comparable, pronounced QW effects from the minority-spin channel should be more readily observable than from the majority-spin channel in Fe(100) films of practical thicknesses. There exists another factor that can play an important role. The energy bands and their offsets in the magnetic film, the spacer layer, and the electrodes control the formation of QW states, while the Bloch-function symmetries control the transmission

coefficient and hence the magnitude of the resonant tunneling current. This latter point was demonstrated in Refs. [4–7], where the existence of Δ_1 Bloch states near the Fermi energy is essential for predicting large TMRs in FM1/MgO/FM2 junctions, where FM1 and FM2 stand for either Fe or Co. In such systems, distinct differences between the two spin channels and any possible sharp resonances arise from the precise matching or mismatching of the Bloch wave function symmetries on both sides of the barrier. Resonant tunneling is then predicated by having a ballistic tunnel junction. From this point of view, the amorphous nature of the barrier may be the critical factor that suppressed resonant tunneling in the experiment of Ref. [2].

The above analysis leads to two criteria for the selection of candidate structures. First is the use of a crystalline spacer layer in a structure consisting of crystalline epitaxial layers. One obvious choice is MgO, which has already been used in both theoretical and experimental [8–10] work in simple spin tunnel junctions, as mentioned earlier. Lattice matching considerations automatically limit the choice of metal layers to the bcc solids of the $3d$ transition metal series and the orientation of the layers to the (100) direction. The second criterion is the suitability of the energy bands and band offsets of the thin films and electrodes. The electrode in direct contact with the thin magnetic film must be nonmagnetic while the other electrode, separated by the MgO spacer, must be magnetic. To accentuate the resonance effect through the tunneling process, the QW states must be produced from a band at the Fermi energy. The symmetry of the band must match the complex band structure in the energy gap of MgO so that the QW states have a higher transmission amplitude through the tunnel barrier than states with a different symmetry. For the bcc solids in the (100) direction with the MgO as the barrier, the relevant band has Δ_1 symmetry. The confinement effect can be produced only if in the nonmagnetic electrode the bottom of the Δ_1 band is well above the Fermi energy.

Based on the above criteria, the magnetic film and the magnetic electrode can be either bcc Fe or bcc Co. The nonmagnetic electrode must be bcc Cr. The minority-spin band structure of bcc Fe is very similar to bcc Cr, while the majority spin has a very different band structure than Cr [11]. Along the symmetry axis ΓH, the Fe majority-spin Δ_1 band crosses the Fermi energy, but there is no Δ_1 band near the Fermi energy for either the Fe minority spin or the Cr band structure. In addition to the band offsets needed for producing QW states, these energy band features provide a differentiation between the two spin channels, which we will exploit in order to achieve strong spin dependence in the resonant tunneling effect.

Using Fe as the magnetic film and the magnetic electrode, our first model system is the bcc (100) Fe/MgO/FeO/Fe/Cr junction. Here we assume that the Cr electrode is deposited first, so that the junction contains a single atomic layer of FeO at the bottom interface be-

tween Fe and MgO, which is observed experimentally [12,13] and was shown [5] to greatly impact the TMR. The structure is similar to the tunnel junctions measured in Ref. [2], except for the barrier which was Al-O. In our second model system, we use Co as the magnetic electrode, and assume that the Co electrode is deposited first, which produces bcc (100) Co/MgO/Fe/Cr. Unlike the first system, there has not been clear experimental evidence of an oxide layer between the Co electrodes and the MgO layer.

Epitaxial ultrathin films are pseudomorphic, which allows us to fix all bcc Fe, Co, and Cr lattice constant of 2.86 Å, corresponding to that of bulk Fe. The thickness of the MgO layer is fixed at eight atomic layers. The lattice structure of all the layers and the self-consistent calculations at zero bias were set up in the same manner as in Refs. [4–6]. All calculations were performed using the layer Korringa-Kohn-Rostoker implementation [14] of the local spin density approximation (LSDA) of density functional theory. Although the ground state of Cr is antiferromagnetic, for simplicity of the calculation we assumed that the bulk part of the Cr electrode is nonmagnetic. The moments of five Cr layers next to the QW layer were allowed to relax and they showed small moments with alternating signs. The self-consistent charge as a function of bias and the current-voltage curves are calculated following the procedure in Ref. [15].

At the $\bar{\Gamma}$ point, the Δ_1 band is primarily s (angular momentum $l = 0$), but there is no s component in any of the other bands. In Fig. 1, we show the s partial density of states (DOS) within the Fe film sandwiched between the MgO barrier layer and the Cr layer in bcc (100) Fe/MgO/FeO/8Fe/Cr, with an 8 ML (monolayer) thickness of the Fe film. There is a gap around the Fermi energy with the bottom of the conduction band coming from the tail of the Δ_1 band in the Cr electrode. Within this gap, there are several QW states as indicated by the sharp spikes
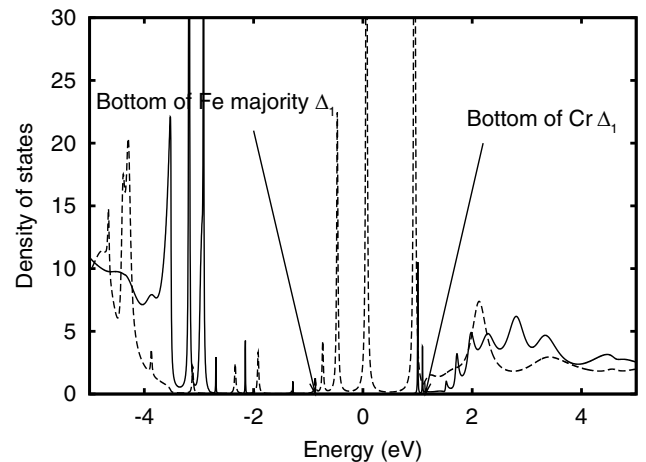

FIG. 1. s-resolved partial density of states in the eight Fe layers in the QW film of Fe/MgO/FeO/8Fe/Cr at the $\bar{\Gamma}(\mathbf{k}_\parallel = 0)$ point. Solid line, minority spin; dashed line, majority spin. The Fermi energy is at 0 eV.

in the DOS. In the majority-spin channel, five of these states are derived from the Fe Δ_1 band. The width of these peaks is equal to exactly twice the imaginary part of the energy and scales linearly as the imaginary part of the energy, indicating that the true width of these states is essentially zero, and that they are indeed QW states and not resonances. The surprise in Fig. 1 is the QW state in the minority spin just below the bottom of the Cr Δ_1 band. This is not an interface state since its wave function extends throughout the Fe film. However, this QW state is not seen in Co/MgO/9Fe/Cr, indicating that it is probably sensitive to the presence (or absence) of the FeO layer on the interface.

The thickness dependence of the majority-spin QW state energies relative to the Fermi energy is shown in Fig. 2 for QW states between 0.3 and 1.1 eV. This window is the same as the one in which Nagahama *et al.* [2] observed oscillations in conductance due to the QW states. If we compare Fig. 2 with Fig. 2 in Ref. [2], the agreement between the two figures is striking. However, the differences between the two figures are more important because they tell us what we cannot learn from experimental data alone. We note that in Ref. [2] a QW resonance is observed at each monolayer thickness. This observation led to the assumption that each series of resonances is due to the same QW state and led the authors to conjecture that the resonances are due to QW states from the $\bar{X}$ point. In contrast, in Fig. 2, the resonances do not occur for every layer thickness. Analysis of the wave functions reveals that the resonances correspond to different QW states, all from the $\bar{\Gamma}$ point. The numbers next to each data point in Fig. 2 indicate the number of nodes of the wave function. This picture tells us that, when the layer thickness increases by 1 or 2 ML, one QW state moves out of the energy window and the next QW state moves in. The actual films in Ref. [2] may consist of regions of varying thick-
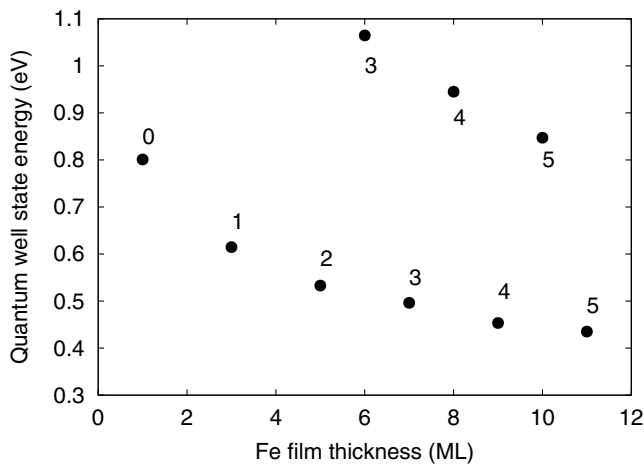
nesses, so the measurements are averaged over even and odd layers.

In Fig. 3 we show the calculated *I-V* curves of the Fe/MgO/FeO/8Fe/Cr (a) and Co/MgO/9Fe/Cr (b) tunnel junctions for both spin channels with the moment in the Fe film aligned parallel or antiparallel to the Fe (or Co) electrode. A positive bias is defined to make the Fermi energy of the Cr electrode higher than that of the ferromagnetic electrode on the other side of the tunnel barrier. For positive biases, electrons are injected into the QW states, and states above the Fermi energy are important. Likewise, for negative biases the QW states below the Fermi energy are important. The majority-spin current shows a staircase feature where the sharp jumps in the current occur at bias voltages that are precisely equal to the energies of the QW states. Between these resonances, the current remains nearly constant, indicating that nearly all of the majority-spin current flows through the QW states. The first QW resonance for Fe/MgO/FeO/8Fe/Cr at positive biases occurs at 0.068 V. At this voltage, the current increases by more than an order of magnitude. The first resonance for Co/MgO/9Fe/Cr is at 0.238 V, where the current shoots up by 2 orders of
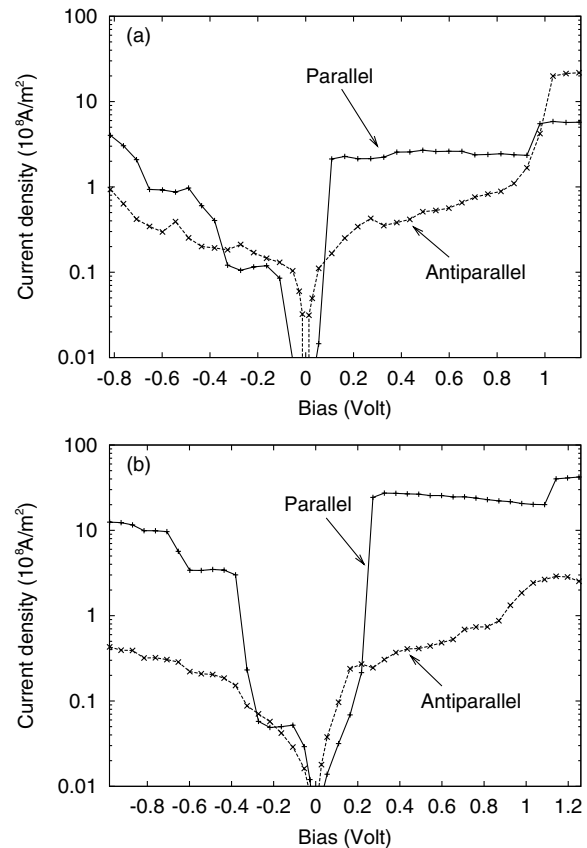

FIG. 3. Tunneling current as a function of bias voltage. (a) Fe/MgO/FeO/8Fe/Cr; (b) Co/MgO/9Fe/Cr. Solid lines are for parallel alignment of the moments in the Fe film with the magnetic electrode. Dashed lines are for antiparallel alignment.

FIG. 2. The thickness dependence of the majority-spin QW state energies for Fe/MgO/FeO/nFe/Cr for $n = 1, \ldots, 11$. The numbers next to the data points indicate the number of nodes in the wave function.
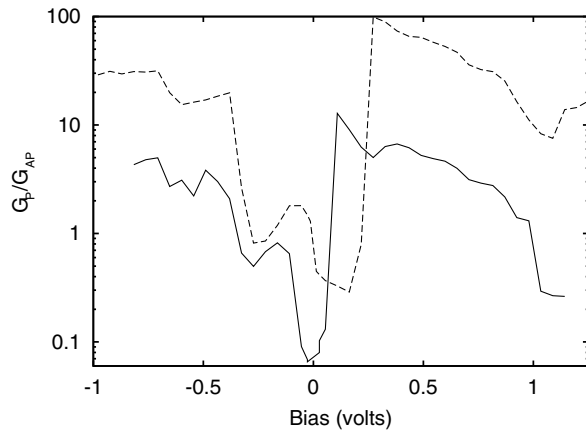
FIG. 4. Conductance ratio as a function of bias voltage for Fe/MgO/FeO/8Fe/Cr (solid line) and Co/MgO/9Fe/Cr (dashed line).

magnitude. The jumps in the current for negative biases at the same voltage values are smaller. At low biases, there are no dramatic jumps in the current for antiparallel alignment of the moments (the moment of the Fe film is aligned oppositely with respect to that of the Fe electrode). Thus, the resonance effect can be easily switched off by applying a small magnetic field. For Fe/MgO/FeO/8Fe/Cr, the antiparallel current becomes larger by almost an order of magnitude than the parallel current when the positive bias approaches 1 V. At this voltage, the QW state in the Fe minority-spin channel enters the transport window, resulting in a large negative TMR, as we will see below. Such an effect is not seen for Co/MgO/9Fe/Cr.

In Fig. 4 we show the conductance ratio G_P/G_{AP}, where G_P is the parallel conductance and G_{AP} is the antiparallel conductance, as a function of the bias voltage. At very small voltages the TMR is sharply negative ($G_P/G_{AP} < 1$). For Fe/MgO/FeO/8Fe/Cr the ratio G_{AP}/G_P approaches 1600% near zero bias. Because one of the electrodes is Cr, which does not have the Δ_1 state near the Fermi energy, the majority-spin current is suppressed at small biases when the QW states are outside the bias window producing the negative TMR. The TMR reaches peaks at the voltages when each additional QW state starts to contribute to the current. The effect is particularly large for positive biases. For Fe/MgO/FeO/8Fe/Cr, the maximum TMR of 1200% at the first QW resonance at positive biases is an order of magnitude larger than the TMR [15] obtained at this voltage for a similar junction but with Fe electrodes on both sides, whereby the QW states were absent. At higher bias, the TMR declines with voltage because the resonant contribution from each QW state remains relatively constant while the antiparallel current increases with the voltage. For Fe/MgO/FeO/8Fe/Cr, the minority-spin QW resonance appears near 1 V and the TMR turns negative again at this bias. Above 1 V the ratio G_{AP}/G_P is 380%.

To summarize, our first principles calculations predict a significant QW resonance effect in a class of tunnel-ing junctions including Fe/MgO/FeO/Fe/Cr and Co/MgO/Fe/Cr. We predict several-fold increases in tunneling current and the TMR due to resonant tunneling through the metallic QW states in the Fe film. Both majority- and minority-spin QW states can contribute to resonant tunneling. The former contributes positively to the TMR, while the latter contributes negatively. We also predict a large asymmetry in the *I-V* curve between positive and negative biases. These properties make these junctions excellent candidates for novel spintronics devices that may combine the features of diodes, rectifiers, field effect transistors, and TMR sensors.

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