Spin polarization decay in magnetic tunnel junctions with semimetal-inserted layers

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Magnetic tunnel junctions (MTJs) were fabricated with a thin layer of semimetallic bismuth inserted between the tunnel barrier and the top ferromagnetic electrode. The tunneling magnetoresistance (TMR) was measured on a set of samples for which the thickness of the inserted layer varied from 0 to 20 nm. The TMR decreased with an exponential decay length that was found to be $\Lambda_{Bi}=4.1\ nm=0.48\ \lambda_{F,Bi}$, where $\lambda_{F,Bi}$ is the Fermi wavelength measured in comparable Bi films. This result is in remarkably good agreement with the decay length previously measured in MTJs with inserted copper layers, $\lambda_{Cu}=0.58\ \lambda_{F,Cu}$, even though the values of λ_F differ by an order of magnitude. It thereby gives a confirmation that the characteristic length scale of the tunneling density of states is the Fermi wavelength. Measurements of TMR as a function of bias voltage show a large asymmetry and the peak TMR is shifted to a nonzero value. © *2010 American Institute of Physics*. [doi:10.1063/1.3415540]

I. INTRODUCTION

The topic of spin dependent tunneling (SDT) continues to receive high attention in condensed matter physics. SDT is characterized by the tunnel magnetoresistance (TMR) measured in a magnetic tunnel junction (MTJ). The MTJ itself has become the premier magnetoelectronic device for applications.[1,2] At the same time, the study of SDT is important for a variety of other spin dependent transport topics.[3] Tunneling and SDT are extremely sensitive to the interfaces between the tunnel barrier and each metallic electrode. It follows that a natural experimental technique is to modulate one of the interfaces and then characterize the change in tunneling. For SDT, a specific example is the insertion of a thin nonmagnetic (N) layer between one of the ferromagnetic electrodes, F1 or F2, and the tunnel barrier I. Since tunneling depends on the density of states *at the interface*, this leads to a severe alteration of the interface and a dramatic effect on tunneling can be predicted.[4]

The Julliere[5] model for TMR predicts that the insertion of a thin N layer at the tunnel barrier should reduce TMR to zero because the density of states of any N material has no spin asymmetry. Although this prediction is generally accepted, several questions arise. A principal question is: what thickness of N is required in order that the density of states at the interface is that of N? This was addressed by Appelbaum and Brinkman in the context of metal/insulator/metal (M/I/M) tunneling with the conclusion that "…tunneling in M/I/M junctions should be sensitive to the density of states of the electrons within a few Fermi wavelengths (λ_F) of the interface." (Ref. 6) because λ_F is the length scale of many-body interactions. Once the length dependence of the tunneling density of states is understood and confirmed, junctions with N-layer insertions can be used to study other issues, such as interface or barrier asymmetry effects.

Because of the spin asymmetry of the density of states of ferromagnetic materials, experiments with N-inserted MTJs can determine the length scale by measuring the TMR as a function of thickness d_N of N, TMR (d_N). The measured decay length Λ then describes the range of the relevant many-body interactions. There have been numerous studies of N-inserted MTJs, but experimental results have shown some variation.[7,8] The authors of Ref. 9 performed a set of experiments on $Co/Al_2O_3/Co$ MTJs. Thin Cu layers were fabricated either underneath, or on top of, the aluminum oxide barrier. Measurements of TMR (d_N) were made for both kinds of samples. Copper layers grown under the barrier had a high structural quality (nearly layer by layer growth) and the decay length measured by the TMR data was $\Lambda_{Cu}=0.26\ nm$, equivalently $\Lambda_{Cu}=0.58\ \lambda_{F,Cu}$.

The disparity in experimental results, and in particular the observation of oscillatory TMR attributed to quantum well states in N,[10] have raised other questions. The issues of spin accumulation,[11] spin dependent scattering at the N/F interface,[12] and magnon scattering[13] have all been discussed. Only the experiments of Ref. 9 have quantitatively confirmed the prediction of Appelbaum and Brinkman.[6] The experiments reported in this paper were designed as a test of this prediction.

We report SDT in ferromagnetic metal 1/tunnel barrier/semimetal (Bi)/ferromagnetic metal 2 structures. Bismuth (Bi) is a group V semimetallic element with a highly anisotropic Fermi surface and unusual transport properties.[14,15] The Fermi wavelength of Bi is an order of magnitude longer than that of Cu. From the TMR (d_N) data, the decay length

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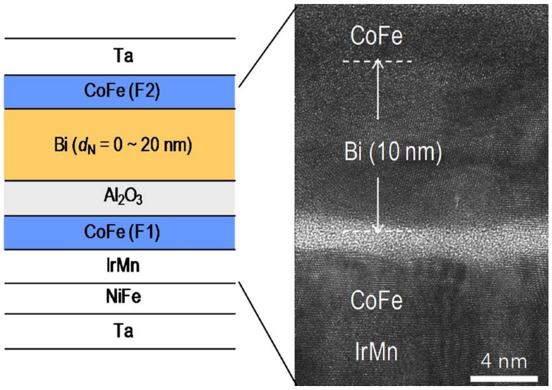

FIG. 1. (Color online) (a) Schematic cross-section view of the MTJ showing the Bi layer inserted between the Al_2O_3 barrier and top ferromagnetic electrode. (b) Cross-sectional TEM micrographs for sample with 10 nm thick Bi layer.

was found to be $\Lambda_{Bi}=4.1$ nm$=0.48$ $\lambda_{F,Bi}$. This result is in remarkably good agreement with that of Ref. 9, even though the values of λ_F differ by an order of magnitude. Furthermore, we study the symmetry of the effect of bias voltage, TMR (V_{bias}), and make the surprising discovery that the decay length Λ_{Bi} is 50% longer for one bias polarity than for the other.

II. EXPERIMENT

MTJs were prepared by a dc/rf magnetron sputtering system with a base pressure of 4.0×10^{-8} Torr on thermally oxidized Si(100) substrates. The generic structure of the Bi-inserted MTJs was [SiO_2 (substrate)/Ta(5 nm)/$Ni_{81}Fe_{19}$ (6 nm)/$Ir_{50}Mn_{50}$(8 nm)/$Co_{84}Fe_{16}$(4 nm)/Al_2O_3(1.6 nm)/Bi(d_N)/$Co_{84}Fe_{16}$(10 nm)/Ta(5 nm)], as shown in a schematic cross-section in Fig. 1(a). A magnetic field of 300 Oe was applied during deposition in order to induce an easy magnetization axis in each electrode. The Al_2O_3 tunnel barrier was formed by using an *in situ* dc plasma oxidation process (11.9 W dc/in.^{2}) in 20 mTorr O_2 after growing a 1.6 nm thick Al film in a separate chamber. The inserted Bi layer was deposited on the Al_2O_3 barrier using rf sputtering (10 W). The thickness of this interfacial layer varied from zero to 20 nm. The deposition of whole stacks was followed by a combination of photolithography, ion milling, and lift-off processes to form MTJs with lateral size 50×50 μm^2. Finally, a 150 nm thick Al cross strip was deposited after patterning by photolithography. Transport properties of MTJs were measured at room temperature by a dc 4-probe method.

For N-inserted MTJs, the morphology of an N layer grown on the tunnel barrier is of crucial importance. A discontinuous N layer permits pinhole contact between the barrier and the top ferromagnetic electrode, and the TMR can be dominated by F1/I/F2 tunneling at the pinholes.[4] Transmission electron microscopy (TEM) was used to study the cross-sectional structure of Bi-inserted MTJ stacks. Figure 1(b) presents a cross-sectional TEM image of a Bi-inserted MTJ with $d_N=10$ nm. The image confirms that the inserted Bi layer is continuous and uniform and has no pinholes. The Al_2O_3/Bi and Bi/F2 interfaces were also found to be clean and flat.

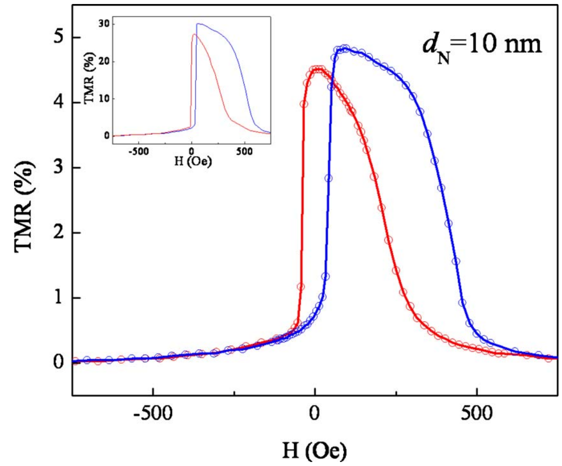

FIG. 2. (Color online) TMR curve measured at room temperature for an FM/I/Bi/FM tunnel junction with $d_N=10$ nm. Inset: TMR curve for a representative control junction with no Bi layer.

III. RESULTS AND DISCUSSION

An example of room temperature tunneling data is presented in Fig. 2 for the MTJ with $d_N=10$ nm, showing TMR=4.8%. The inset shows data for a representative control junction with no inserted Bi layer ($d_N=0$ nm). The measured value, TMR=29% before annealing, indicates a high quality Al_2O_3 barrier having SDT efficiency comparable with the control junctions in the study of Ref. 9 (TMR =22% to 27%). The TMR improved after annealing, reaching a postanneal value TMR of 47% and further confirming that high quality MTJs were achieved with our fabrication procedure. Since the morphology of inserted Bi layers is affected by annealing, all measurements were performed with as-grown MTJs.

The dependence of TMR on the thickness of the inserted Bi layer for thicknesses $d_N=0$ to 20 nm is presented in Fig. 3. The data were fit to a simple exponential

$$\mathrm{TMR}(d_N) = A \exp\left(-\frac{d_N}{\Lambda_{Bi}}\right) \quad (1)$$

where A is a free-fitting parameter for the amplitude. The least-squares fitting procedure gives $A=29.3\%\pm1.2\%$, the

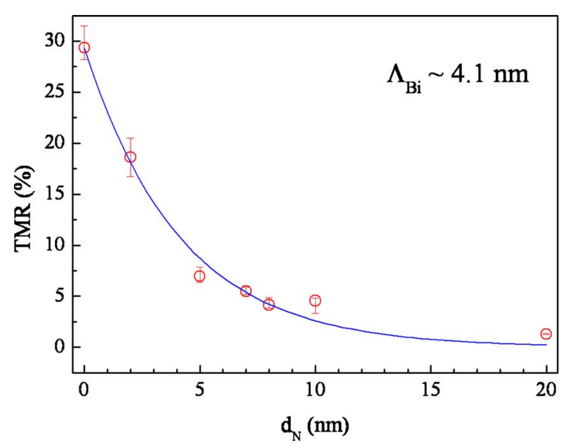

FIG. 3. (Color online) The variation in TMR as a function of thickness of the Bi-inserted layer, TMR (d_N). Open circles are average values and error bars represent distributed values for several junctions with same Bi thickness. Solid line is a fit to the equation in the text.

same as the experimentally observed value for $d_N=0$. The single remaining fitting parameter was found to be $\Lambda_{Bi}=4.1\pm0.3$ nm.

Semimetallic Bi has a complicated and highly anisotropic Fermi surface, and the electronic transport properties of Bi are uniquely different from those of common metals.[14,15] Pockets of the Fermi surface have electrons with low density and small effective mass and the Fermi wavelength can be roughly 40 nm in bulk samples[16] or epitaxially grown films.[17] Thin Bi films are typically semiconducting.[18] However, band-bending at an interface with another material may result in additional interface states. Near the interface (and for sufficiently thin films), the carrier density may increase, and the Fermi wavelength decrease, by an order of magnitude.[18] To estimate the Fermi wavelength in our Bi films, measurements of the conductivity and Hall coefficient were performed on Bi films grown under identical conditions and of comparable quality. Using the experimentally determined value of carrier density, $n=1.36\times10^{19}$ cm^{-3}, the Fermi wave number was found to be $k_F=7.4\times10^6$ cm^{-1}. It follows that $\lambda_{F,Bi}=2\pi/k_F=8.5$ nm, and we note that the fitted decay length can be written as $\Lambda_{Bi}=4.1$ nm$=0.48$ $\lambda_{F,Bi}$. This result is in remarkably good agreement with data for inserted copper layers, $\Lambda_{Cu}=0.58$ $\lambda_{F,Cu}$,[9] even though the values of Λ and λ_F differ by an order of magnitude for the two materials. Our results provide a confirmation of the prediction of Ref. 6, and support the assertion that the range of many-body interactions that determine the tunneling density of states is the order of the Fermi wavelength. It could also be confirmed that the exponential function is well-fitted to the data as in Fig. 3.

The TMR of MTJs is known to show a strong dependence on bias voltage. Asymmetries in TMR (V_{bias}) data are ubiquitous but the TMR peak is usually located at $V_{bias}=0$. We have studied the bias dependence of our Bi-inserted junctions and found that the TMR peak is shifted away from zero. Figure 4(a) shows the bias dependence, TMR (V_{bias}), for the control and Bi-inserted samples. The control sample shows a small asymmetry and the asymmetry is more pronounced for the Bi-inserted samples. To clarify the difference, the TMR of the $d_N=10$ nm sample is compared with the control sample and normalized with respect to the value at $V_{bias}=0$ in Fig. 4(b). While the asymmetry is obviously greater, it is furthermore seen that the peak value of TMR in the Bi-inserted sample is shifted to a positive bias value, $V_{bias}\sim50$ mV. For further analysis, the measured *I-V* curves and calculated dI/dV data (for the parallel magnetization configuration) were fits to the Simmons[19] and Brinkman[20] formulas. From these fits, the effective barrier height and average barrier thickness for the control junction were determined to be 3.54 eV and 1.2 nm, respectively. Fits to the asymmetric barrier model of Brinkman give us the barrier asymmetry parameter $\Delta\Phi$, defined as the difference between barrier heights on the two sides. In contrast to the control junction case, with $\Delta\Phi=0.42$ eV, $\Delta\Phi$ for the Bi-inserted junction ($d_N=10$ nm) is much larger $\Delta\Phi=1.66$ eV. This large barrier asymmetry may cause the strongly asymmetric bias dependence of TMR as well as the shift in peak value.[21] To provide a more clear presentation of these asymmetries,

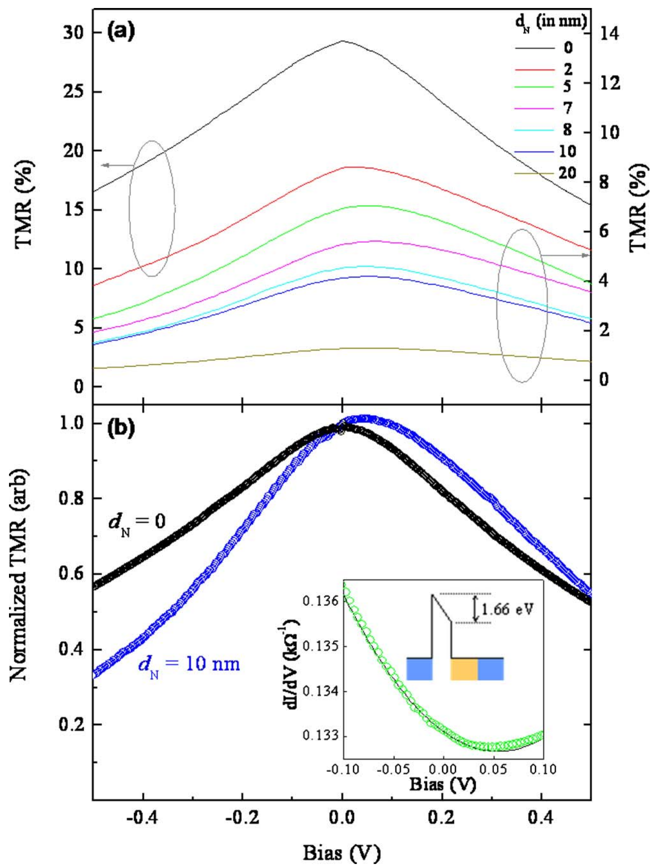

FIG. 4. (Color online) (a) The dependence of TMR with bias voltage, TMR (V_{bias}), at room temperature, for samples with Bi insertion layers of thickness 0, 2, 5, 7, 8, 10, and 20 nm. (b) TMR (V_{bias}) for the control junction (black symbols) and for $d_N=10$ nm (colored symbols). The latter is normalized to the zero bias value of the former. Inset: bias dependence of dI/dV (green circles) for $d_N=10$ nm and the fit to the Brinkman model (Ref. 6) (black line).

the data of Fig. 4(a) are fit to Eq. (1) for a range of bias voltages. The amplitude of the control sample at a given bias voltage is used for the value of A at each bias value. The resulting plot, Λ_{Bi} (V_{bias}), is shown in Fig. 5. The decay length is seen to vary by about 50%, from a minimum of 3.1 nm ($V_{bias}=-500$ mV) to a maximum of 4.7 nm ($V_{bias}=+250$ mV). This is believed to be the observation of bias dependent decay lengths with a strong asymmetry. Although

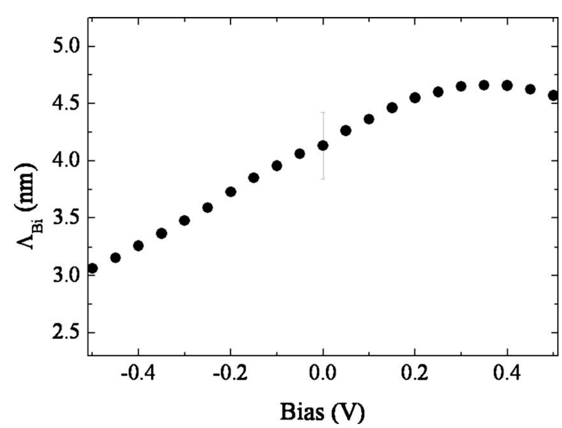

FIG. 5. Asymmetries in TMR (V_{bias}) are presented by plotting the dependence of decay length Λ_{Bi} with bias voltage, Λ_{Bi} (V_{bias}).

the detailed explanation is not clear, the authors considered that one of the possible reasons for the observed bias dependent decay lengths is the magnon excitations at F/*I* interface.[22,23] For the case of reverse bias (see Fig. 5 inset), the hot electrons with energy higher than the Fermi level tunnel through the barrier and reach to the F1. Then these hot electrons may cause magnon excitations while losing energy. This results in the flipping of electron spins, which decreases the decay length. By contrast, tunneling electrons from F1 to Bi (under the forward bias) lose their energy while passing through the Bi layer. Consequently, the electrons arrive at F2 generate less magnon excitations and exhibit longer decay length under the forward bias.

IV. CONCLUSIONS

In conclusion, a systematic study of SDT in MTJs, $Co_{84}Fe_{16}$ (4 nm)/Al_2O_3(1.6 nm)/Bi(d_N)/$Co_{84}Fe_{16}$(10 nm), having an inserted interfacial layer of Bi has been presented. The thickness d_N of the Bi layer was varied from 0 to 20 nm. TEM confirmed that the interfaces in the Bi-inserted MTJ stack are high quality and that all layers are continuous and free of pinholes. An exponential fit to TMR (d_N) at zero bias gives $\Lambda_{Bi}=4.1$ nm$=0.48$ $\lambda_{F,Bi}$. This result is in good agreement with data from similar experiments using inserted copper layers, $\Lambda_{Cu}=0.58$ $\lambda_{F,Cu}$,[9] even though the values of Λ and λ_F differ by an order of magnitude for the two materials. It therefore gives a confirmation to the prediction that the range of many-body interactions that determine the density of states is the order of the Fermi wavelength.[6] The dependence of TMR on bias voltage also was studied. Large asymmetries in TMR (V_{bias}), which include a shift in the peak TMR to a nonzero bias value, were presented as Λ_{Bi} (V_{bias}). The decay length Λ_{Bi} is 50% longer for one bias polarity bias than for the other.

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