Magnetic tunnel junctions with ZnSe barriers

Xin Jiang, a) Alex F. Panchula, b) and Stuart S. P. Parkin c)
IBM Research Division, Almaden Research Center, San Jose, California 95120

(Received 26 August 2003; accepted 2 October 2003)

Magnetic tunnel junctions with ZnSe barriers were fabricated with a combination of magnetron sputtering, ion beam sputtering, and effusion cell evaporation. Tunneling magnetoresistance values of \(~10\%\) are observed at room temperature. The temperature and barrier thickness dependences of the junction resistance and tunneling magnetoresistance are consistent with a predominant direct tunneling mechanism when the barrier thickness is less than \(~10\text{ nm}\). © 2003 American Institute of Physics. [DOI: 10.1063/1.1630160]

Magnetic tunnel junctions (MTJs), comprised of two ferromagnetic metal electrodes separated by a thin tunnel barrier, are being widely considered for possible use as magnetic recording read heads in high density hard disk drives, and as nonvolatile storage cells in magnetic random access memories, especially because of their high tunneling magnetoresistance (TMR) values. 1–5 One of the challenges in making useful MTJ read head sensors is to reduce the junction resistance-area product to \(~1 \Omega \text{(\mu m)}^2\) to obtain sufficiently high signal-to-noise ratios for high data rate applications. To meet these requirements using conventional aluminum oxide tunnel barriers requires a very thin barrier on the order of 0.6–0.7 nm, making the barrier susceptible to pinholes which can reduce the TMR significantly. An alternative solution may be to use wide band gap semiconductors, such as ZnSe, as the tunnel barrier. With a band gap of \(~2.8\text{ eV}\), ZnSe provides a much lower barrier height compared to aluminum oxide. As a result, thicker barriers will give rise to lower resistance-area products. The lattice constant of ZnSe (5.669 Å) is almost exactly twice that of bcc Fe with only a 1.1\% mismatch. Thus, it should be possible to grow high quality ferromagnetic metal/ZnSe hybrid devices. Moreover, the chemical reactivity of Fe with ZnSe is lower than with other semiconductors such as GaAs and Si. Indeed, Fe/ZnSe hybrid structures have been prepared without a magnetic dead layer at the interface. 6 Another intriguing aspect of MTJs with ZnSe barriers is their potential application in spintronics. Ferromagnetic metals are natural candidates as spin-polarized electron sources at room temperature. Previous work has demonstrated that spins can be coherently transferred between ZnSe and other semiconductors such as GaAs. 7 Thus, the ferromagnetic metal/ZnSe structure may provide a useful system for spin injection into semiconductors.

Highly resistive ZnSe films have been grown on GaAs(001) by molecular beam epitaxy (MBE). 8 It was found that purification of the Se source played an important role in improving the film quality. Abad et al. grew Fe/ZnSe/Fe epilayers on ZnSe buffered GaAs(001). 9 Under Zn-rich conditions, the Fe films preserved their magnetic character. Marangolo et al. studied the magnetic properties of Fe films grown on ZnSe. 6 They found no reduction of the Fe magnetic moment at the Fe/ZnSe interface even after thermal annealing up to 300 °C. Although these studies suggest that it should be possible to fabricate MTJs with ZnSe barriers, there are few reports on such devices in the literature thus far. Gustavsson et al. studied the structural and transport properties of Fe/ZnSe/CoFe MTJs grown by MBE. 10 A TMR value of 16\% was observed at 10 K, which decreased with increasing temperature and vanished at temperatures above 50 K. Meanwhile, the junction resistance decreased rapidly at elevated temperatures, suggesting that thermal activation of carriers in the ZnSe barrier began to dominate transport across the tunnel junction.

In this letter, we report the fabrication of MTJs with ZnSe barriers using a combination of deposition techniques including magnetron sputtering, ion beam sputtering, and effusion cell evaporation. A \(~10\%\) TMR value was observed at room temperature. Transport measurements were conducted on MTJs with different barrier thicknesses at various temperatures to characterize the tunneling mechanism.

The MTJs were grown on Si(001) substrates coated with 25 nm silicon oxide. The deposition system was equipped with six dc magnetron sputtering guns, five ion beam sputtering sources, and two effusion cells. The base pressure of the system was \(~10^{-9}\) Torr. Three metal shadow masks were used to form the bottom electrode, tunnel barrier, and top electrode, respectively. First, a highly conducting bottom electrode consisting of 5 nm Ta/20 nm Cu/10 nm Ta/25 nm Ir$_{22}$Mn$_{78}$/1.5 nm Co$_{70}$Fe$_{30}$/4 nm Fe was deposited using a combination of magnetron and ion beam sputtering. The Ta/Cu/Ta seed layers adhere well to silicon oxide. The magnetic moment of the bottom electrode was exchange biased via an antiferromagnetic Ir$_{22}$Mn$_{78}$ layer. As a result, the magnetic switching fields of the magnetic moments in the top and bottom electrodes are well separated and it is possible to achieve parallel or antiparallel alignment of the magnetic moments by applying suitable magnetic fields. This combination of underlayers also provides a template for growing (001) oriented bcc Fe.

A thin, 0.3-nm-thick Zn interface layer was deposited on top of the Fe layer before the growth of the ZnSe barrier. This interface layer was found to improve the growth reproducibility and the TMR of the MTJs. The addition of Zn at the ferromagnetic metal/ZnSe interface may reduce stacking fault densities in the films. It may also limit the intermixing between the ferromagnetic metal and ZnSe. Two effusion

---

a) Also at: Solid State and Photonics Laboratory, Stanford University, California, 94305.

b) Also at: Department of Applied Physics, Stanford University, California, 94305.

c) Electronic mail: parkin@almaden.ibm.com
cells, loaded with 99.999% pure ZnSe and Se, respectively, were used to grow the ZnSe tunnel barrier. Before the barrier growth, the ZnSe cell was heated at 695 °C for 3 h to purify the source material while the Se cell was not heated. The ZnSe and Se cell temperatures were kept at 746 and 100 °C, respectively, for depositing ZnSe at a rate of ~0.5 Å/s. The use of the Se cell provides a Se-rich environment, which is known to make the ZnSe films more resistive. A series of MTJs were grown with various barrier thicknesses in the same growth run. The top electrode of the MTJs consists of 15 nm Co70Fe30 capped with 5 nm Ta and 7.5 nm Ru. Again, a 0.2-nm-thick Zn interface layer was inserted between the ZnSe barrier and the Co70Fe30 layer to improve the MTJ properties. The substrates were maintained at room temperature during the entire growth. The active area of the MTJs was ~100×100 μm².

The roughness of the ZnSe barrier was analyzed with atomic force microscopy (AFM). The film shows dense grain-like features with a density of ~560 grain/μm² (Fig. 1). Large boulder-like features (bright spots in the AFM image) with a density of 1–3 boulder/μm² are also seen, which are assumed to be particulate contaminants from the laboratory environment and are excluded from the roughness calculations. The film roughness is ~0.9 nm. The roughness of ZnSe may be due to the low substrate temperature during the growth. It may be possible to obtain smoother ZnSe films with an optimized growth temperature. The chemical composition of the ZnSe films was analyzed with Rutherford backscattering (RBS), which gave a stoichiometric Zn to Se ratio of 1:1, within the experimental uncertainty of ±0.5%.

Shown in Fig. 2 is the junction resistance of a MTJ with a 6.5-nm-thick ZnSe barrier measured at room temperature. The relative change of resistance from parallel to antiparallel alignment in the major R(H) loop gives rise to a TMR value of 10.2%. The minor R(H) loop shows that the switching fields of the magnetic moments of the two electrodes are well separated. The temperature dependence of the junction resistance and TMR is summarized in Fig. 3. The junction resistance increases gradually from ~5 to ~11 kΩ as the temperature decreases from 295 to 15 K. This relatively weak temperature dependence of junction resistance is similar to that we observe in MTJs with aluminum oxide barriers and confirms that the conduction is predominantly through a direct tunneling mechanism. In contrast, Gustavsson et al. observed a much larger increase of junction resistance upon cooling, which was attributed to “freezing out” of carriers in the conduction band of the ZnSe barrier. The lack of thermally activated carriers in the ZnSe films presented here is consistent with their high resistivity. The TMR of the MTJs also increases slowly upon cooling, reaching ~20% at 15 K [Fig. 3(b)].

Analysis of the temperature dependence of the conductance and TMR reveals that the spin polarization through the barrier is about 30.5% and that there is a significant spin-independent contribution to the tunneling. This contribution, which accounts for ~25% of the conductance at room
temperature is most likely due to defect mediated tunneling conduction via a single trap state. This conductance, which is assumed to have a strong temperature dependence, accounts for the large increase (over a factor of 2) in TMR with decreasing temperature.

The thickness dependence of junction resistance in parallel alignment at room temperature is presented in Fig. 4(a). Rather thick barriers (>5 nm) were needed in order to obtain measurable junction resistance. By contrast, similar MTJs with aluminum oxide tunnel barriers exhibit measurable resistance for barriers only 2–3 nm thick. For ZnSe barriers with thicknesses in the range from 5 to 10 nm, the junction resistance increases exponentially with barrier thickness, as expected for tunneling transport. The solid line in Fig. 4(a) is a fit assuming an exponential thickness dependence, which indicates that the junction resistance increases by an order of magnitude for every additional 0.8 nm ZnSe. In contrast, for typical MTJs with aluminum oxide barriers, the junction resistance increases by an order of magnitude every ~0.25 nm. In the WKB approximation, the logarithm of the junction resistance is linearly dependent on barrier thickness with a slope proportional to $\sqrt{m^* \phi}$, where $m^*$ and $\phi$ are the electron effective mass and the tunnel barrier height, respectively. ZnSe has a small barrier height and a small electron effective mass. This may account for the relatively slow increase of junction resistance with barrier thickness as well as the rather thick barriers needed to obtain suitably resistive junctions for junctions with areas of ~10,000 $\mu$m$^2$.

For ZnSe barriers thicker than 10 nm, the rate of increase of junction resistance with barrier thickness is reduced. This implies that the tunneling process is dominated by hopping or tunneling via defect states in the ZnSe barrier. Electrons transmitted through such defect states can no longer preserve their spin orientation. Consequently, the TMR is expected to diminish, as observed. As shown in Fig. 4(b), for thin barriers, the TMR value is roughly constant. For thicker barriers, the TMR goes to zero.

A simple model can be used to account for the thickness dependence of the TMR. In the parallel alignment, the total junction conductance can be written as $G_T = G_{T,P} + G_{H}$, where $G_{T,P}$ and $G_{H}$ are the conductance through the direct tunneling mechanism and hopping mechanism, respectively. Similarly, the total junction conductance in the antiparallel alignment is $G_{AP} = G_{T,AP} + G_{H}$. The measured TMR is therefore given by

$$\text{TMR} = \frac{G_T - G_{AP}}{G_{AP}} = \frac{T_0}{1 + G_H / G_{T,AP}}$$

where $T_0 = (G_{T,P} - G_{T,AP}) / G_{T,AP}$. We assume an exponential increase of $G_H / G_{T,AP}$ with barrier thickness in the form of $A \exp(Bl)$, where $A$ and $B$ are constants and $t$ is the barrier thickness. In addition, we assume that $T_0$ is independent of the barrier thickness. The solid line in Fig. 4(b) is the result from using this model, where the fitting parameters are: $T_0 = 9\%$, $A = 3 \times 10^{-8}$, $B = 1.7$ nm$^{-1}$. The calculation agrees with the experimental data qualitatively.

In conclusion, we have demonstrated the fabrication of MTJs with ZnSe barriers. A TMR value of 10% was observed at room temperature which increases to ~20% at 15 K. The temperature and barrier thickness dependences of the junction resistance and TMR are consistent with a predominant direct tunneling transport mechanism when the barrier is thinner than ~10 nm. For thicker barriers, the measurements indicate that a defect mediated tunneling mechanism begins to contribute to the junction conductance, which results in a decreased TMR. These results suggest that ZnSe barriers have potential for providing low resistance-area product magnetic tunnel junctions.

The authors thank Ronald F. Marks and Andrew Kellock for AFM and RBS measurements. The authors thank DARPA for their partial support of this work.