# Single-crystalline Fe/Cr/Fe/MgO/Fe magnetotunnel junctions grown on GaAs(001)

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(Presented on 3 November 2005; published online 26 April 2006)

Fe/MgO/Fe/Cr/Fe tunneling structures were epitaxially grown directly on GaAs(001) without buffer layer. Interdiffusion in the Fe/GaAs interface was investigated using conversion electron Mössbauer spectroscopy. An independent magnetization switching in the Fe electrodes was achieved by pinning the magnetization of the Fe-top electrode by antiferromagnetic coupling across a Cr spacer to another Fe film. For transport measurements, optical lithography and ion etching were applied to reduce the junction area. A tunneling magnetoresistance of 10%–12% at room temperature was found. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2171939]

The injection of spin-polarized carriers from a ferromagnet (FM)-semiconductor (SC) interface is recognized as an important problem for operation of integrated FM/SC spintronics circuits.[1] Theoretical works predict that the efficiency of the spin injection from the FM into the SC can be improved for electrons created by tunneling through a barrier, since such a procces is not affected by the conductivity mismatch.[2] The experiments made on Fe/AlGaAs structures with a Schottky barrier formed in the interface showed a spin injection efficiency of 30% at room temperature.[3] The electrons can tunnel through the insulating barrier (I) intentionally grown on the SC-based substrate. In this case the tunneling electrons can be, in addition, spin filtered when another FM film is placed between the insulating barrier and the semiconductor. Its thickness has to be adjusted to fit between the inelastic mean-free path of spin-up and of spin-down electrons. In such case the carrier injection proceeds in the FM/I/FM/SC structure, therefore the spin polarization of the states available for the tunneling comes into play, i.e., the FM/I/FM exhibits a tunneling magnetoresistance (TMR). The theoretical calculations predict a 1000% TMR effect in the case of an ideal Fe/MgO/Fe junction and about 80% when a FeO layer is assumed to be formed at the Fe/MgO interface.[4] The highest experimental value of TMR at room temperature reported for the Fe/MgO/Fe magnetotunnel junction (MTJ) to the date is 180% for junctions grown on a MgO substrate.[5] Nevertheless, a similar successful attempt for Fe/MgO/Fe MTJs grown directly on semiconducting substrates without buffer layers has not been reported yet.

In this paper we report on the TMR of single-crystalline Fe/MgO/Fe tunneling structures grown on commercially available GaAs(001) substrates. An independent magnetization switching in the Fe electrodes was achieved by pinning the magnetization of one of the Fe films by antiferromagnetic coupling across a Cr spacer to another Fe film.[6,7] Such Fe/Cr/Fe system, if combined with the Fe/MgO/Fe MTJ, could remarkably improve the junction operation. First, the magnetization of one of the junction electrodes can be discretely manipulated over a broad range of magnetic field by a proper choice of the Cr spacer and Fe thicknesses in the Fe/Cr/Fe trilayer.[7] Second, spin prefiltering increases the spin polarization of the tunneling electrons that are injected into the semiconductor.

The GaAs substrates were prepared by preannealing to 520 °C, subsequent sputtering at the same temperature and finally at 590 °C (with Ar ions of energy of 500 eV).[8] The growth of Fe, Cr, and MgO was carried out at a pressure below $4\times10^{-10}$ mbars (maximum pressure after a long deposition of Fe). Magneto-optical Kerr effect (MOKE) ellipticity loops were collected in the longitudinal geometry (for 1.85 eV photon energy and incident *s*-polarized light at 49° to the sample normal) using an electromagnet with a maximum field of 30 mT. Conversion electron Mössbauer spectra (CEMS) were measured *ex situ* at room temperature with a constant acceleration spectrometer and $^{57}$Co/Rh as a

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$\gamma$-radiation source. For transport measurements, optical lithography and ion etching were applied to reduce the junction area to $5\times5$ $\mu m^2$. In order to obtain a high signal-to-noise ratio, the resistivity was measured at low bias voltage (10 mV). For the *ex situ* experiments (CEMS and transport) the samples were covered by 20 monolayers (ML) of Au in order to protect against oxidation.

We have grown Fe/Cr/Fe trilayers with a Cr thickness of 9 ML corresponding to antiferromagnetic coupling between the Fe layers. At low fields, the Fe films in the Fe/Cr/Fe trilayer switch simultaneously and keep their antiferromagnetic coupling. The switching field (coercivity of the trilayer) depends on the thickness relation between both Fe layers, i.e., the thickness of one of the layers ($d_1$) with respect to the thickness of the complete structure ($d_1+d_2$).[7] The lowest switching field obtained in trilayers with strongly different Fe thickness corresponds to the coercivity of the thicker layer and the highest is approached when the thickness of both Fe layers becomes similar.

To obtain best MgO barriers, they are deposited at an elevated temperature (400 °C).[9] This allows to obtain pinholes-free, atomically flat, and crystallographically well-ordered insulating layers. Another requirement for the growth of MgO barriers concerns the quality of the underlying Fe films which was improved by annealing of the Fe/Cr/Fe structure after its deposition at room temperature. The annealing results in the increased coercivity of the Fe/Cr/Fe structure which is tentatively explained by intermixing and formation of nonmagnetic Fe compounds at the Fe/GaAs interface. The nonmagnetic Fe atoms decrease the total magnetization of the bottom Fe film, which influences the relation between the magnetization of the thicker ($d_1$) Fe films with respect to the magnetization of the complete Fe/Cr/Fe structure.

The above hypothesis is confirmed by Mössbauer spectra measured for Fe films grown on GaAs(001) with a 5-ML-thick $^{57}$Fe probe layer placed just at the Fe/GaAs interface. The $^{57}$Fe layer was then completed with 10 ML of natural Fe in order to assure film continuity and a Curie temperature well above room temperature. Conversion electron Mössbauer spectrum for the as-prepared sample is plotted in Fig. 1(a). It is shown that after annealing for 20 min to 200 °C a doublet appears in the center of the spectrum [Fig. 1(b)]. Its contribution increases up to 46% with further annealing to 270 °C for another 20 min [Fig. 1(c)]. The hyperfine interaction parameters exactly correspond to room-temperature values cited for FeAs compound.[10] This determines 200 °C as a maximum temperature, which cannot be exceeded during the Fe/Cr/Fe and MgO deposition. The average magnetic hyperfine field calculated from the spectrum shown in Fig. 1(b) is 25.3 T compared to 33 T for bulk Fe. Assuming proportionality between the magnetic moment and magnetic hyperfine field, this means that the annealing at 200 °C reduces the magnetization of the 5-ML-thick $^{57}$Fe probe layer by about 23% which is equivalent to the magnetization of 1.2-ML-thick Fe layer. Such a magnetization decrease in the bottom Fe layer could be responsible for the observed increase of coercivity of the Fe/Cr/Fe structure.

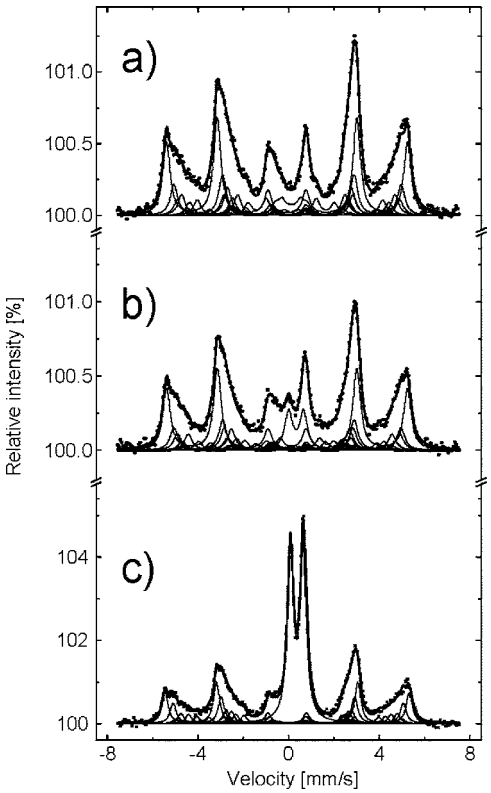


FIG. 1. Conversion electron Mössbauer spectra of 10 ML Fe/5 ML $^{57}$Fe/GaAs(001) measured at room temperature for the as-prepared sample (a), after annealing for 20 min at 200 °C (b), and at 270 °C (c).

For the $d_2$Fe/9 ML Cr/$d_1$Fe trilayer on GaAs(001), another change of magnetization takes place at higher fields, i.e., above the coercivity of the Fe/Cr/Fe structure. This change corresponds to more or less perpendicular orientation of the magnetization of the thinner $d_2$Fe films with respect to the magnetization of the thicker $d_1$Fe films, which is due to the interplay between magnetic anisotropy and interlayer magnetic coupling.[7] As reported previously, for the 15 ML Fe/9 ML Cr/20 ML Fe/GaAs(001) structure this kind of magnetization change occurs at a field of about 60 mT.[7] The field value at which the magnetization changes, as well as the whole field dependence of magnetization, $M(H)$, can be modeled by minimizing the total magnetic energy of the Fe/Cr/Fe system including anisotropy and both bilinear and biquadratic interlayer couplings. The calculations were performed assuming the same anisotropy constants for the bottom $d_1$Fe layer as those of the single Fe film of the same thickness grown on GaAs(001) (Ref. 11) [Fig. 2]. For the 15 ML Fe/9 ML Cr/20 ML Fe/GaAs(001) the magnetization changes abruptly (spin-flop) at the field of about 60 mT in agreement with the value measured for the same structure. Obviously, the anisotropy constants of the bottom Fe layer depend on its thickness. Much thicker Fe/Cr/Fe bottom electrodes (i.e., thicker Fe layers) are technically required in order to avoid the situation, when the Fe/Cr/Fe bottom electrode of the Fe/MgO/Fe/Cr/Fe junction would be fully etched. According to Ref. 11, above the thickness of 20 ML of Fe grown on GaAs(001), the fourfold anisotropy dominates over the uniaxial anisotropy. For the bottom layer thickness $d_1=50$ ML, we expect a fourfold anisotropy con-

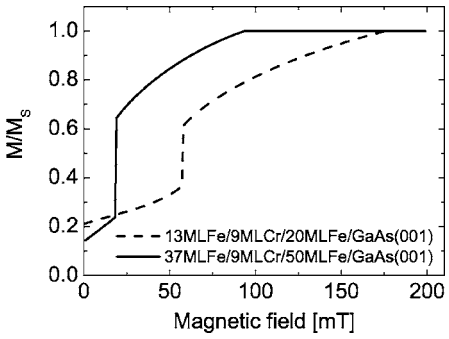


FIG. 2. Magnetization normalized to the saturation magnetization calculated for the Fe/Cr/Fe trilayer system assuming (i) fourfold anisotropy with the easy axis of magnetization along [100] in both Fe layers, (ii) additional uniaxial anisotropy with easy axis along [110] only in the bottom Fe layer, and (iii) bilinear and biquadratic exchange coupling between the Fe layers.

stant three times larger than the uniaxial anisotropy constant.[11] Then, the $M(H)$ calculations performed for the 37 ML Fe/9 ML Cr/50 ML Fe/GaAs(001) structure predicts the change of magnetization of the Fe/Cr/Fe structure at a field of about 20 mT.

The experimentally measured Kerr ellipticity hysteresis loop for the complete 16 ML Fe/15 ML MgO/37 ML Fe/9 ML Cr/50 ML Fe/GaAs(001) MTJ structure is shown in Fig. 3(a). The shape of the loop can be explained as a superposition of the reversed minor loop for the 37 ML Fe/9 ML Cr/50 ML Fe structure and a loop with a small coercivity. The magnetization of the Fe/Cr/Fe structure changes at a field of about 20 mT. This is exactly the value which was calculated for this structure as shown in Fig. 2. The effect of decreasing ellipticity with increasing field is of magnetooptical origin.[12] The small coercivity loop corresponds to the top Fe electrode

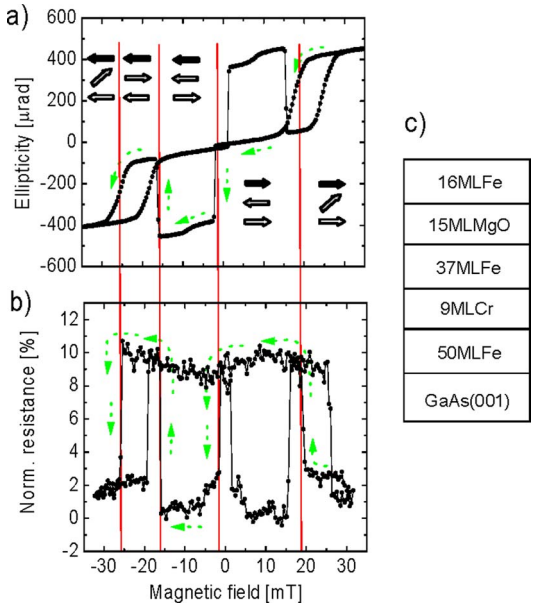


FIG. 3. (a) MOKE loop for 16 ML Fe/15 ML MgO/37 ML Fe/9 ML Cr/50 ML Fe/GaAs(001) structure measured at room temperature along the [110] direction. The arrows correspond to the magnetization of the respective Fe films. (b) Corresponding magnetoresistance measurement of the $5\times5$ $\mu$m$^2$ junction performed at room temperature with a 10 mV bias voltage, in the external magnetic field applied along [110]. A schematic diagram of the complete sample is drawn on the right (c).

which behaves like a "free" Fe layer if the MgO insulating layer is thick enough. For the electric transport measurements, the junction's area had to be reduced due to existence of pinholes in the MgO barrier.[13] The pinholes play the role of "conducting bridges" inside the insulator, which cause short circuits between the Fe electrodes of the MTJ. In order to decrease the probability of having the pinholes in the junction, the junction area was reduced by optical lithography and ion etching to patches of size $5\times5$ $\mu$m$^2$. Figure 3(b) shows a result of the transport measurement at room temperature in the magnetic field applied along the [110] direction. After a careful analysis of the relative magnetization orientation of the Fe electrodes in the Fe/MgO/Fe junction, it becomes obvious that the resistance changes follow exactly the changes of magnetization orientation of the Fe electrodes [Fig. 3(a)]. The observed tunneling magnetoresistance of 10% is much lower than that predicted theoretically and achieved for Fe/MgO/Fe junctions grown on MgO.[5] However, it was rather difficult to obtain a good crystallographic order and an atomically flat surface of Fe film grown on GaAs as well as of MgO layer. We suggest that this is due to the limited temperature that can be applied during the growth providing no interdiffusion in the Fe/GaAs interface. Most likely, the imperfections in the MgO insulating layer are responsible for the reduced TMR effect.

In conclusion we report on a TMR effect of 10% in the Fe/MgO/Fe system grown directly on GaAs(001). The obtained value is much lower than this predicted theoretically for Fe/MgO/Fe junctions and that achieved experimentally growing the Fe/MgO/Fe junctions on MgO substrate. In the case of the MTJ grown on GaAs(001), it was difficult to obtain a good crystallographic order and atomically flat films due to the limited temperature providing no interdiffusion between GaAs and overlaying Fe. Also the spin polarization of one or both Fe electrodes could be reduced due to FeO formation at the interface.

The authors want to thank Dr. H. Brückl for the transport measurements performed in the Department of Physics, University of Bielefeld, Germany.

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