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Interlayer magnetic coupling in single-crystalline Fe/Cr/Fe and Fe/MgO/Fe structures grown on GaAs(0 0 1)

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Abstract

Single-crystalline Fe/MgO/Fe magneto-tunnel junctions were grown epitaxially on GaAs(0 0 1). Independent magnetization switching was achieved by pinning the magnetization of one of the Fe electrodes by antiferromagnetic coupling via Cr to another Fe layer (GaAs/Fe/Cr/Fe/MgO/Fe). For the antiparallel magnetization orientation in the Fe/Cr/Fe trilayer, "reversed" minor hysteresis loops are measured with longitudinal MOKE. The shape of the loop corresponding to the top "free-Fe" electrode is determined by four-fold magnetic anisotropy with the easy-axis along [1 0 0], superimposed by a weak uniaxial anisotropy with [0 1 0] easy-axis. The loops are asymmetric due to a delicate balance between the uniaxial anisotropy and magnetic interlayer coupling between the Fe electrodes separated with a thin MgO spacer.

Keywords: Metal–insulator interfaces; Tunneling; Magnetic measurements; Iron; Magnesium oxides; Gallium arsenide; Magnetic phenomena (cyclotron resonance, phase transitions, etc.)

Single-crystalline ferromagnet/insulator/ferromagnet (FM/I/FM) magneto-tunnel structures have become a subject of increased interest in recent years. First, this is due to theoretical calculations that predict more than 70% tunneling magneto-resistance effect in the case of Fe/MgO/Fe junctions [1] and due to improved efficiency of the spin injection from the FM-spin aligner into a semiconductor for electrons injected by tunneling. Such a process is not affected by the conductivity mismatch and results in conservation of the spin polarization [2]. Secondly, magnetic exchange coupling can only be separated from other coupling mechanisms (like pinholes or "orange–peel" coupling) in the case of good quality FM/I/FM structures. Therefore, it is attractive to choose a single-crystalline insulator. In particular, for a very thin insulator layer, an antiferromagnetic (AFM) exchange coupling is predicted theoretically [3] and recently experimentally confirmed [4].

In this paper we discuss Fe/MgO/Fe tunneling structures epitaxially grown on GaAs(0 0 1). An independent magnetization switching in the Fe films is achieved by pinning the magnetization of

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one of the Fe films (called “middle”) by antiferromagnetic coupling across a Cr-spacer to another Fe film (called “bottom”). AFM-coupled Fe/Cr/Fe trilayers, epitaxially grown on GaAs(0 0 1), reflect magnetic anisotropy of the GaAs(0 0 1)/Fe system characterized by uniaxial anisotropy of the easy-axis of magnetization along the [1 1 0] direction. This allows growth of an artificial “layered antiferromagnet” exhibiting uniaxial magnetic anisotropy with minimal material and technological complexities. By combining giant magneto-resistance (of such the AFM-coupled Fe layers) and magnetic tunnel junction (Fe/MgO/Fe) elements with semiconducting materials, new magneto-electronic devices can be constructed. It has been shown recently, that the magnetic tunnel transistor with Fe/Au/Fe spin-valve base exhibits giant magneto-current [5] and thus may be useful e.g. as a room temperature source of highly spin-polarized electron current. The total structure of our samples is: GaAs(0 0 1)/Fe/Cr/Fe/MgO/Fe. The magnetization reversal in such Fe/Cr/Fe structures can be controlled by changes in the magnetic anisotropy, thickness of the Cr spacer as well as thickness of the Fe layers. At low fields, the Fe films in the Fe/Cr/Fe trilayer switch simultaneously (AFM-switching) and keep their AFM coupling. The AFM-switching field depends on the thickness relation between both Fe layers, i.e. the thickness of one of the layers with respect to the thickness of the second one. The lowest AFM-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 is the same.

The sample preparation and characterization were carried out in an ultra-high vacuum chamber equipped with molecular beam epitaxy (MBE), Auger electron spectroscopy (AES), low energy electron diffraction (LEED), scanning tunneling microscopy (STM) and in situ longitudinal magneto optic Kerr effect (MOKE). MOKE loops were collected in the longitudinal geometry by using an electromagnet with a maximum field of 30 mT. The GaAs substrates were cleaned by 500 eV Ar^+ sputtering at 590 °C until no traces of contamination were detected in AES spectra and sharp LEED patterns were observed. The cleaning procedure resulted in a Ga terminated (4×6)-like reconstruction, which is found to protect the Fe film against strong intermixing with As and Ga [6]. Fe, Cr and MgO were deposited at a rate of 1–1.5 ML/min by MBE from thoroughly outgassed high-purity rods at a pressure below 4×10^{-10} mbar.

Depending on the magnetic techniques used for characterization, the observed magnetic properties of multilayers may vary. In our case of antiferromagnetically coupled Fe layers, “reversed” minor hysteresis loops are observed with longitudinal MOKE (Fig. 1, at low fields). This means that a negative remanence is detected when the thicker bottom Fe layer is saturated along the applied field. This behavior is interpreted by depth variation of the MOKE sensitivity, which results in a smaller contribution of the thicker but deeper layer, than of the thinner middle layer, to the total MOKE signal [7]. The thinner middle Fe film is magnetized opposite to the field thus contributes with a negative ellipticity and a reversed minor loop is observed. This scenario can be verified if other magnetic transition (resulting in a positive ellipticity) occurs within the field range that can be applied in situ (±30 mT in the case of our experimental setup).

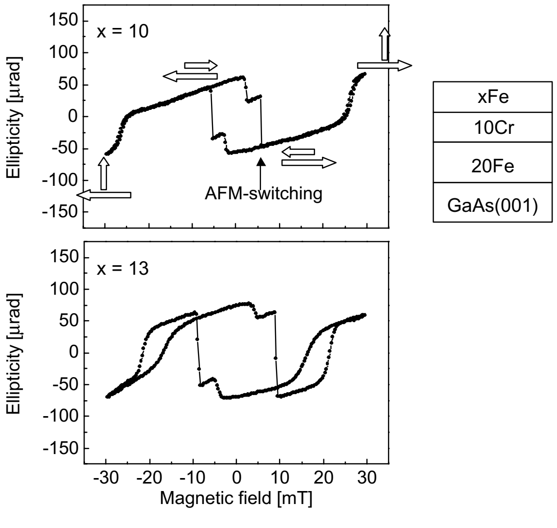

Fig. 1. MOKE loops measured at 300 K along [1 0 0] for GaAs(0 0 1)/20Fe/10Cr/*x*Fe. For this thickness of the Cr spacer, the field of 30 mT is sufficient to ravage the antiparallel orientation of magnetization in the Fe layers. Inhomogeneity (steps) of the AFM-switching field is clearly visible.

At a Cr thickness of 10 ML the AFM-switching field is relatively small and we observe jet another magnetic transition. The field is strong enough to orient the magnetization of the middle Fe film perpendicularly to the magnetization of the bottom one, i.e. to observe a spin–flop transition [8,9]. This is reflected in a complex shape of the MOKE minor loops measured in this case as it is shown in Fig. 1 for GaAs(0 0 1)/20Fe/10Cr/10–13Fe samples (the field is applied along [1 0 0]). The loops confirm that the negative ellipticity at positive fields (and vice versa) originates in magneto-optical properties of the GaAs/Fe/Cr/Fe system and in a depth sensitivity of MOKE signal [7]. Just above the AFM-switching field, the thinner top Fe layer is magnetized opposite to the field, but contributes more to the total MOKE signal than the thicker bottom Fe layer. With increasing field, the ellipticity becomes positive due to the spin–flop transition that eliminates the negative contribution of the middle Fe layer to the total MOKE signal. The positive ellipticity corresponds to the bottom Fe layer only which is magnetized along the field, but contributes less to the MOKE signal than it is expected from its thickness. It is also seen that when the thickness of the middle Fe film increases, the AFM-switching field increases following the trend mentioned above. In the case of this particular sample, the spin–flop transition does not occur if the film is magnetized along the [1 1 0] direction, at least up to the maximum field available in our experimental setup. It is also seen in Fig. 1 that the AFM-switching in the Fe/Cr/Fe structure is not homogeneous: two slightly different AFM-switching fields can be distinguished. This behavior is observed only when the bottom (thicker) Fe layer of the Fe/Cr/Fe structure was deposited in two steps: first 10 ML of Fe at 300 K and the rest 10 ML of Fe at 450 K. This kind of inhomogeneity was already seen in coercivity of the bottom Fe layer in the MOKE loops measured just after this layer was grown. Most likely this is caused by Ga/As diffusion into the Fe film during its annealing which changes the coercivity of the part of the Fe film that interfaces with the GaAs(0 0 1) substrate.

The shape of the MOKE loops is more complex after the structure is completed by further deposition of MgO and Fe layers on the top of the GaAs(0 0 1)/Fe/Cr/Fe stack. This is shown in Fig. 2 for the GaAs(0 0 1)/20Fe/10Cr/13Fe/10MgO/10–20Fe samples. Qualitatively, the shape of the loop can be understood by combining the loop measured for the GaAs(0 0 1)/20Fe/10Cr/13Fe sample (Fig. 1) with the loop of a single 10 (or 20) ML thick Fe film characterized by a small coercivity. For MgO-spacer thickness of about 13 ML, the minor loop corresponding to the top "free-Fe" layer is rectangular and almost symmetric with respect to the zero-field axis. With reduced thickness of the MgO insulating layer, the loop shifts towards positive fields. At MgO thicknesses below 5 ML, the shift approaches the value of 8mT suggesting a clear ferromagnetic coupling between the middle and top Fe films. Taking into account that the middle Fe layer is magnetized opposite to the applied field, the shift of the loop of the top Fe layer to positive fields means that parallel magnetization orientation is preferred over antiparallel. The shift is almost the same when the field is applied along the [1 1 0] direction. This ferromag-

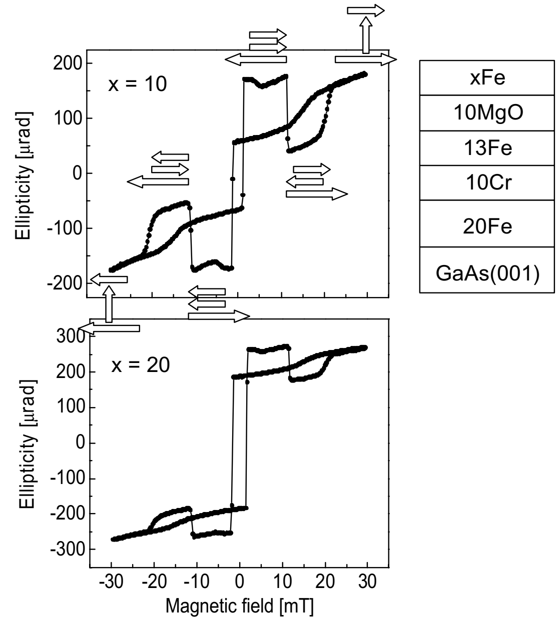

Fig. 2. MOKE loops measured at 300 K for GaAs(0 0 1)/20Fe/10Cr/13Fe/13MgO/*x*Fe along the [1 0 0] direction. Complex shape of the loops corresponds to the magnetization reversal in the 20Fe/10Cr/Fe trilayer (shown in Fig. 1) superimposed by the low-coercivity loop of the top "free" Fe layer (contributing more to the total loop with increasing *x*).

netic coupling between the middle and top Fe films across the MgO spacer layer is also seen in the AFM-switching field which is larger than the value measured before the top MgO/Fe sequence was deposited. This effect is illustrated for the sample in which the "spin–flop" transition does not occur within the applied field range (Fig. 3, before and after the top MgO/Fe deposition, respectively). Such an increase of the AFM-switching field is reasonable assuming that the top "free-Fe" layer is not perfectly separated from the Fe/Cr/Fe structure and a significant ferromagnetic coupling across the MgO layer exists [8]. Due to this coupling, the top layer participates to the magnetic moment of the middle Fe layer. The AFM-coupled structures reverse their magnetization in an external magnetic field due to the torque, which is proportional to the net magnetic moment. The coupling between the Fe/Cr/Fe structure and the top "free-Fe" layer causes the net magnetic moment to decrease with respect to that of the Fe/Cr/Fe structure. Consequently, higher fields are required to reverse the magnetization of the structure (see Fig. 3). Below 3 ML of MgO, the coupling between the top Fe films is strong enough to reverse the magnetization of both Fe films together. The coupling causes the middle and the top Fe layers to behave like a single thick Fe layer. This is immediately seen from the loop because the "thick Fe layer" (thicker than the bottom Fe layer) is magnetized along the field and the minor MOKE loop is not "reversed" [7]. The critical thickness of the MgO layer at which the magnetizations of the Fe films switch separately depends mainly on the quality of the Fe/MgO interface. Nevertheless, we were not able to reach MgO thicknesses of the order of 1–2 ML without direct ferromagnetic coupling existing between the Fe layers. This is contrary to the observation of Faure-Vincent et al. [4] who were able to detect an antiferromagnetic coupling between two Fe films separated with the MgO spacer thinner than 3 ML, however in a much better defined system.

The shape of the loop of the top "free-Fe" layer is determined by the 4-fold magnetic anisotropy with the easy-axis along [1 0 0], which is, however, not equivalent to the [0 1 0] axis (Fig. 4). This causes the loops of a double-step character with zero net "magnetization" within a well defined range of the applied field. The vanishing MOKE ellipticity measured for a single ferromagnetic Fe film means that the magnetization of this top layer does not contribute to the MOKE response detected in longitudinal geometry, i.e. its magnetization is oriented perpendicular to the field. Magnetization reversal proceeds through intermediate spin alignment along the [0 1 0] direction suggesting that magnetization along [0 1 0] is energetically more favorable. When the field is applied along the [0 1 0] direction, the loops are always of a single-step character. This kind of behavior was observed previously for several systems, e.g. in the case of Fe films grown on vicinal Ag(0 0 1) [10]. A final verification of the character of the process should come from transversal MOKE analysis which is in progress [11]. Such additional uniaxial magnetic anisotropy (with easy-axis along [0 1 0]) usually originates in details of the growth conditions; however, an influence of the interlayer coupling cannot be excluded. Thus we tried to saturate the bottom Fe/Cr/Fe structure along [1 0 0] and measure minor MOKE loop along [0 1 0], and vice versa, saturate along [0 1 0] and

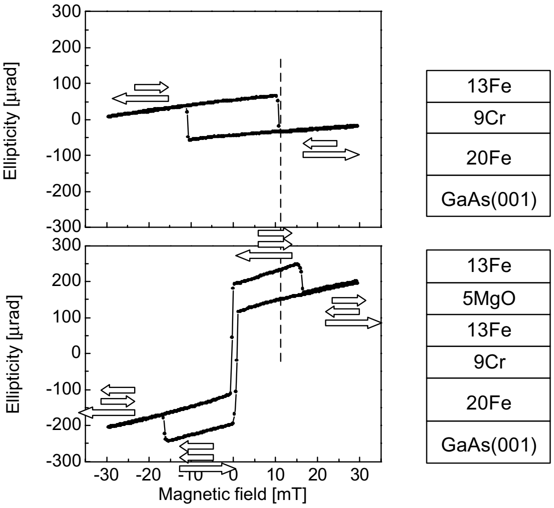

Fig. 3. MOKE loops measured at 300 K along the [1 1 0] direction for GaAs(0 0 1)/20Fe/9Cr/13Fe sample (upper loop) and after the structure is completed with 5MgO/13Fe (lower loop). The AFM-switching field is increased after the MgO/Fe sequence deposition (as it is indicated by the broken line) due to the ferromagnetic coupling between the Fe/Cr/Fe and top "free-Fe" layers across the MgO spacer which is only 5 ML thick.

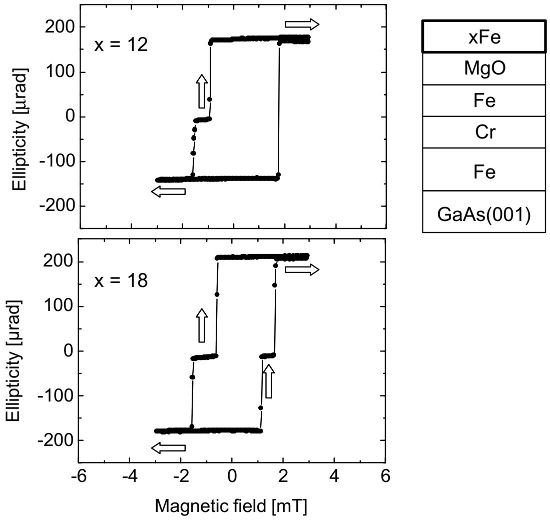

Fig. 4. MOKE minor loops measured at 300 K for the top "free-Fe" layer along the [1 0 0] direction. With increasing thickness of the Fe layer the loop is more symmetric. This is due to the uniaxial anisotropy with the easy-axis of magnetization along [0 1 0] direction that dominates over the interlayer coupling.

measure along [1 0 0]. We found that character of the loop depends only on the direction along which the field is applied for MOKE measurements, with no influence of the previous magnetization history. These results show that the "double-step" character of the loops is of a growth-induced character rather than the anisotropy of the magnetic coupling. Covering of the top "free-Fe" layer with a protective layer of Au (at 300 K) does not change magnetic anisotropy of the Fe layer. Nevertheless, annealing the whole structure at 450 K results in the "single-step" minor loop or causes a weaker "double-step" character. This can be related to changes of the in-plane surface anisotropy of bcc-Fe that depend on temperature at which the fcc-Au-coating is performed [12].

The top "free-Fe" layer becomes ferromagnetic at 300 K only after more than 7 ML of Fe is deposited. This is due to the three-dimensional growth of Fe on MgO(0 0 1) reported before by many groups (e.g. [13]). However, in the case that the Fe layer is grown on a perfect surface of the MgO-single-crystal, the onset of ferromagnetic order at RT is reported to persist at the Fe thickness below 6 ML [13]. One has to remember that in our experiments Fe is grown on an MgO thin film whose quality is determined by the quality of the underlying Fe layer. It is difficult to improve flatness of the Fe surface due to a limitation in the temperature that can be applied for growth or annealing. Temperatures above 500 K result in a strong intermixing between the GaAs substrate and Fe, which changes dramatically the magnetic properties of the system [14].

When the top "free-Fe" layer is only 8–9 ML thick and the field is varied from negative to positive values, the magnetization reverses directly from $[\bar{1}\,0\,0]$ to [1 0 0] orientation (Fig. 4). When the field is varied from positive to negative values, the magnetization switches from [1 0 0], through intermediate orientation along [0 1 0], to the final alignment along $[\bar{1}\,0\,0]$. This is due to a delicate balance between the 4-fold anisotropy, uniaxial anisotropy with [0 1 0] easy-axis and magnetic interlayer coupling. The magnetization switches to the [0 1 0] direction more easily if the applied field direction supports parallel orientation of magnetizations of both the Fe electrodes. The parallel orientation is preferred due to the weak ferromagnetic coupling between the Fe layers across the MgO spacer. More energy is required to align the magnetization antiparallel to the bottom Fe electrode of the junction. In this case, the magnetization switches directly to the [1 0 0] direction. With increasing film thickness, the four-fold anisotropy is more pronounced and the intermediate magnetization alignment along the [0 1 0] direction persists both for positive and negative fields (Fig. 4). The results are explained under the single domain assumption, which is justified by almost rectangular shape of the hysteresis loops we measured.

To conclude we stress the fact that we are able to grow single-crystalline Fe/Cr/Fe/MgO/Fe tunneling structures on top of atomically flat GaAs(0 0 1) substrates. Manipulation of the magnetization orientation in the Fe layers separated with the MgO spacer is realized by tuning the magnetic anisotropy with respect to the indirect interlayer magnetic coupling across the Cr spacer and relative thickness of the Fe layers. For MgO thickness of less than 6 ML, it is difficult to restrict magnetic coupling between the Fe layers to the exchange coupling only. The magnetization reversal in the top "free-Fe" layer along [1 0 0] proceeds through the intermediate magnetization orientation along the [0 1 0] direction. Typically "double-stepped" MOKE loops detected in this case become asymmetric for very thin Fe films due to the delicate balance between the uniaxial magnetic anisotropy and the interlayer coupling. The minimum MgO layer thickness, at which the Fe layers can be reversed separately, is about 2.5 ML.

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