Exchange bias in Fe/EuTe(111) bilayers

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We report on the investigation of the exchange bias effect in Fe layers on EuTe(111), an antiferromagnetic semiconductor. For this ferromagnet (FM)/semiconducting antiferromagnet (AFM) exchange bias system, we have found positive and negative exchange bias effect (EB). Fresh samples exhibit positive EB, independently of the applied cooling field, indicating antiferromagnetic coupling between the FM and the AFM layers at the Fe/EuTe(111) interface. The change in EB with time, from positive EB for fresh samples to negative EB after short time,is attributed to aging effects at the Fe/EuTe interface. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2767220]

I. INTRODUCTION

Magnetic interaction at interfaces between different layered magnetic materials is an important current topic in physics and materials science, particularly when combining ferromagnetic metals and semiconductors, due the emergence of the spintronics.[1–3] The semiconductor/metal integration in nanoelectronic devices is a current task for several applications, as well as the inclusion of controllable magnetic properties in these devices. The exchange bias effect (EB) associated with the exchange coupling between ferromagnetic (FM) and antiferromagnetic (AFM) systems is being extensively studied both due to its key role in spin valves and tunneling magnetoresistance devices, among others, and due to its interesting basic properties.[4–8] In a typical FM/AFM exchange-biased system, a positive cooling field results in the observation of a shift of the hysteresis loop toward negative field, i.e., in a negative exchange bias field (H_E). Positive exchange bias, a shift of the hysteresis loop toward a positive magnetic field when the system is cooled in positive fields, is an unusual situation. It was first observed in Fe/FeF_2 and Fe/MnF_2 bilayers,[5,9] more recently in other layered systems,[10,11] and is normally associated to antiferromagnetic exchange coupling at the interface between the magnetic layers.[5]

Nowadays, there is huge interest in diluted magnetic semiconductor and also a renewed interest in bulk magnetic semiconductors, among them GaN and the europium chalcogenides.[12–16] The exchange bias effect in semiconductor nanostructures has also been experimentally investigated, although not using semiconductors at the AFM counterpart.[17–19] EuTe, one of the europium chalcogenides, is a magnetic semiconductor extensively investigated and a type II Heisenberg antiferromagnet with a Néel temperature (T_N) of 9.6 K.[20] Below T_N, epitaxial EuTe films with a (111) orientation present AFM order with ferromagnetic (111) planes with the spins in adjacent (111) planes antiparallel to each other, and the (111) plane as the easy plane.[21] As the (111) planes of EuTe would constitute an ideal uncompensated surface and as the relevant Eu-Eu magnetic interactions are well known, ferromagnetic/EuTe seems a promising system for the study of fundamental properties related to EB, phenomenon that still needs a complete understanding. Furthermore, its NaCl structure presents an ideal configuration for the investigation of exchange and superexchange interactions.[20–22]

Although the combination of magnetic semiconductor and 3*d* metal films be of high interest, as pointed out earlier, there are only a few works on 3*d* metal/EuTe systems[23,24] and, to the best of our knowledge, no experimental investigation on the exchange bias effect in FM/AFM bilayers based on the AFM EuTe. We have investigated the exchange bias effect in Fe/EuTe(111) bilayers prepared by molecular beam epitaxy (MBE) and, depending on the time period between the preparation of the Fe/EuTe bilayer and the magnetic measurements, we have found positive or negative shifts of the magnetic hysteresis loops. The change in EB with time, from positive H_E for fresh samples to negative H_E after short time is attributed to aging effects at the Fe/EuTe interface. These results are present in detail.

II. EXPERIMENTAL PROCEDURE

Fe/EuTe bilayer samples with structure of 150 or 80 Å Fe/1200 Å EuTe(111) were grown on top of $BaF_2(111)$ substrates and covered with a 30 Å Cu protective layer. Fully relaxed EuTe layers were grown in a Riber 32P MBE system onto freshly cleaved (111) BaF_2 surfaces baked at 450 °C for 30 min immediately before the growth. This

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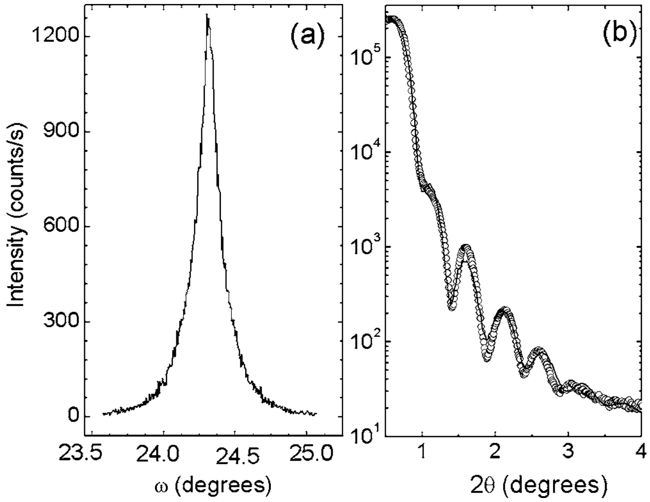

FIG. 1. (a) Rocking curve of the (222) Bragg reflection of the 1200 Å thick EuTe(111) epilayer. (b) Low-angle x-ray reflectivity data (open circles) for a 150 Å Fe/1200 Å EuTe(111) bilayer (sample 2), along with the best least-squares fit (solid line).

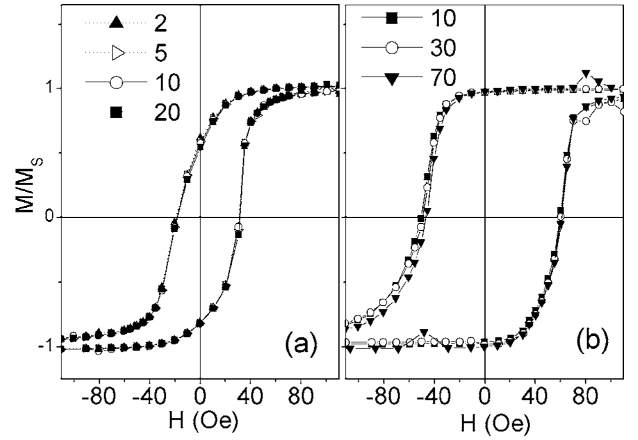

FIG. 2. SQUID magnetization loops at 2 K for two fresh 150 Å Fe/EuTe samples obtained for cooling fields (H_{FC}) applied in-plane and ranging from 2 to 20 kOe for (a) sample 1 and (b) from 2 to 70 kOe for sample 2. The values of the different H_{FC}s, in kOe, are indicated in (a) and (b).

method prevents the buildup of high thermal strains in the sample on cooling to cryogenic temperatures. For the preparation of the EuTe films, two individual cells were used as Eu and Te_2 beam source and the growth rate was kept constant at ~1.3 Å/s. The EuTe(111) epitaxial layers were grown at 175 °C. The growth conditions have to be precisely controlled in order to obtain high quality and smooth EuTe epitaxial films grown in a two-dimensional layer-by-layer mode,[25] since the EuTe(111) surface has a very strong tendency to a three-dimensional growth mode.[26] After the EuTe growth, the samples were transferred to a second MBE system and the Fe and the protective Cu layers were grown at room temperature. The pressure in the ultrahigh vacuum chamber was in the low 10^{-9} mbar range during the depositions of Fe and Cu. The growth rate was 5.0 Å/min for Fe and Cu. Prior to Fe deposition, the surface stoichiometry of the EuTe film was controlled *in situ* by x-ray photoelectron spectroscopy. Evaluation of the Eu and Te 3*d* peaks confirms an Eu:Te ratio equal to 1.0 for the as prepared EuTe/BaF_2(111) surfaces. Structural characterization was performed by high-angle x-ray diffraction (XRD) and grazing-incidence x-ray reflectivity (GIXR). The magnetic and EB properties were investigated by superconducting quantum interference device (SQUID) magnetometry at temperatures from 2 to 300 K and after cooling through T_N of the AFM EuTe layer in magnetic fields (H_{FC}) ranging from 2 to 70 kOe. Hysteresis loops were measured in applied magnetic fields within ±2 kOe. The magnetic fields, including H_{FC}, were always applied in the sample plane, along a ⟨110⟩ direction.

III. RESULTS AND DISCUSSION

The good structural quality of the EuTe epilayers on BaF_2(111)[26] is illustrated in Fig. 1(a) by the x-ray diffraction rocking curve of the (222) Bragg reflection of a 1200 Å EuTe film, where a full width at half maximum equal to 0.197° is observed. For the Fe/EuTe(111) bilayers, XRD analysis indicates that the Fe layers are polycrystalline. Such a polycrystalline structure is probably induced by an initial island-like growth, as is typical for metal deposition on insulator or semiconductor substrate,[27] and was already reported for Ni and also for Fe growth on EuTe.[23,24] As determined by GIXR [Fig. 1(b)], the typical roughness at the Fe/EuTe interface is around 1.0 nm.

Magnetization loops at 2 K for two 150 Å Fe/EuTe(111) samples after cooling in in-plane magnetic fields (H_{FC}) ranging from 2 to 20 kOe for sample 1 and from 2 to 70 kOe for sample 2, always set at 20 K, as obtained within few days after the preparation of the samples ("fresh samples"), are shown in the Fig. 2. The value of the exchange bias field (H_E) at 2 K, +10±2 Oe for sample 1 and +13±2 Oe for sample 2, is always positive and independent of the cooling field, as recently observed for FM/FM bilayers.[10] The observed positive H_E can be understood as consequence of an antiferromagnetic coupling between the Fe spins and the Eu spins at the Fe/EuTe(111) interface in fresh samples,[5] which could take place via direct or indirect exchange interaction depending on the composition of the topmost atomic layer of the AF. The dependence of H_E and H_C with the temperature was also investigated, as shown in Fig. 3 for magnetization loops of samples 1 and 2, below and above T_N of the EuTe. The coercivity (H_C) of the Fe films increases with decreasing temperature (from 15±2 Oe at 20 K to 47±2 Oe at 2 K for sample 2, for example), evidencing also the exchange coupling between the ferromagnetic Fe with the antiferromagnetic EuTe layers. The observed H_E values indicate that the interface energy of the Fe/EuTe(111) system is relatively small. However, this is somewhat expected given the single crystal nature of the AFM layer.[28,29]

Surprisingly, the Fe/EuTe(111) bilayers present a fast aging concerning the magnetic behavior. SQUID magnetization hysteresis loops repeated few (~3) weeks after the first measurements, after keeping the samples stored at room temperature (RT) in a dry vacuum excicator, revealed negative exchange bias of −30 Oe at 2 K [Fig. 4(a)]. As shown in Fig. 4(b), the negative H_E vanishes at the T_N of the EuTe, as expected for a FM/AFM exchange biased system. The reversal in signal of the exchange bias field in the Fe/EuTe(111)

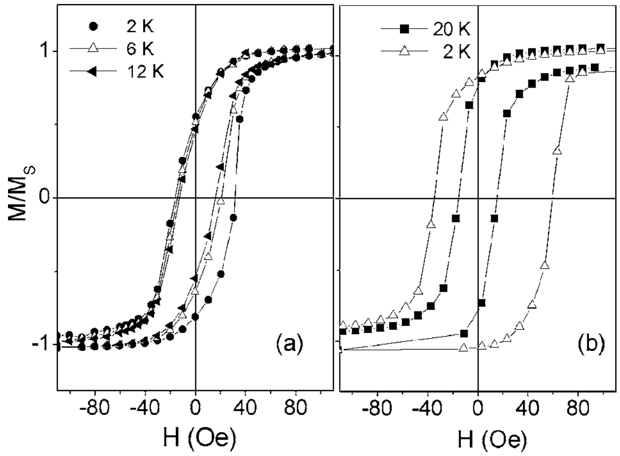

FIG. 3. SQUID magnetization loops at different temperatures for two fresh 150 Å Fe/EuTe bilayers: (a) sample 1 and (b) sample 2.

bilayers is evident from the comparison of the measurements for sample 2 at 2 K and cooling field of ~3.0 kOe, shown in Figs. 3(b) and 4(a), where a change in H_E from +13 Oe for the fresh sample to −30 Oe after aging at RT can be observed. The changes in the form of the loops [more rounded for the fresh sample, Fig. 3(b)] could be an indication of different magnetization reversal behavior, but could also be attributed to some alteration in the morphology of the sample with time. For a sample with a thinner Fe layer (sample 3, 80 Å Fe/1200 Å EuTe), a H_E of −142 Oe was observed under equivalent conditions and after similar aging time, as shown in Fig. 5.

The fast change in EB from positive to negative H_E could be due to interdiffusion effects between Fe and EuTe taking place at RT. To address possible intermixing at the Fe/EuTe interface, a sample with a 15 Å thick ^{57}Fe probe layer at the Fe/EuTe interface was prepared and investigated by conversion electron Mössbauer spectroscopy. Apart from the typical six line component due to α-Fe, the spectrum (not shown) also revealed the presence of a nonmagnetic compo-

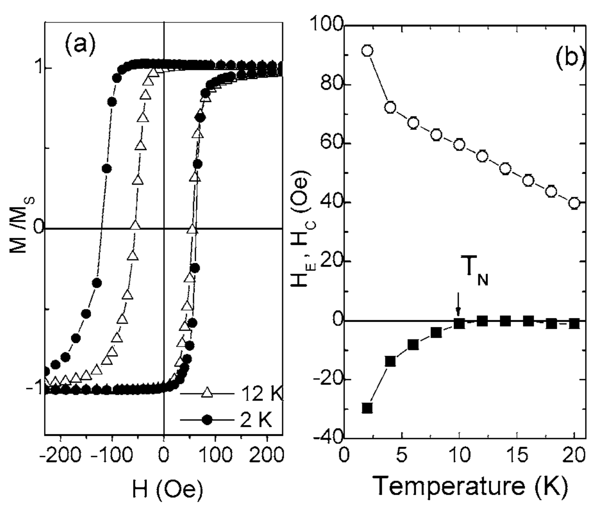

FIG. 4. (a) SQUID magnetization loops of sample 2 at 2 K (solid circles) and 12 K (open triangles), after ~3 weeks stored at RT and in vacuum ("aged"); (b) evolution of the values of the exchange bias field (H_E, solid squares) and coercivity (H_C, open circles) with temperature. The arrow indicates the Néel temperature of the EuTe.

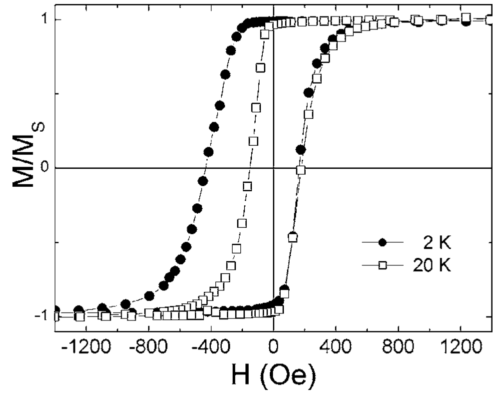

FIG. 5. SQUID magnetization loops for an aged 80 Å Fe/EuTe(111) bilayer (sample 3) at 2 (a) and 20 K (b), measured with a cooling field of 3 kOe applied in-plane.

nent (a doublet) that suggests the formation of Fe-Te bonds at the Fe/EuTe interface. A chemical reaction at the Fe-EuTe interface would break the magnetic order at the very first layers of the EuTe(111), affecting the coupling between the Fe film and the antiferromagnetic layer.

Although chemical effects at the interfaces are often observed in different FM/AFM exchange-biased systems, like bilayers with metals,[30] oxides,[31] or fluorides[32] as the AFM counterpart, and also in model systems for the integration of ferromagnetic materials with semiconductors, like Fe on GaAs and related semiconductor substrates,[33] for these systems there is no indication of alteration of the magnetic behavior within short time after preparation due to aging at room temperature. The Fe/semiconductor system studied here has been shown to be highly sensitive to aging effects at room temperature and the change of sign of the interface coupling seems to be caused by the interface interdiffusion. Moreover, the increase of H_E after aging can be also a consequence of the diminution of the effective thickness of the FM layer due to the intermixing. Nevertheless, the influence of other effects, like an eventual dilution of the AFM layer, as observed by Miltényi *et al.* for Co/CoO layers,[29] could not be evaluated in our experiments up to now and cannot be ruled out.

IV. CONCLUSION

In conclusion, for Fe/EuTe(111) bilayers, an exchange-biased system with a semiconductor as an antiferromagnet, we have observed positive exchange bias effect independently of the applied cooling field, suggesting antiferromagnetic coupling between the Fe atoms at the interface and the uncompensated EuTe(111) surface. Fast relaxation from positive to negative exchange bias within short time (days, and keeping the samples in vacuum at room temperature) seems to be due to interdiffusion at the Fe/EuTe interface. Although much more research on this FM/semiconducting AFM system is still needed, chemical reactivity and formation of compounds at the interfaces between semiconductors and metals, as it seems to be the case for Fe-EuTe, is a common phenomenon, and perhaps all-semiconductor structures may be needed for full capability spintronics devices.

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