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Magnetic properties of EuS spin filter tunnel contacts to silicon

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We investigate the magnetic properties of the ferromagnetic insulator EuS in view of its potential in spin-filter tunnel contacts to silicon. We prepared thin polycrystalline EuS films directly on (001) oriented Si substrates that show well-defined magnetic properties down to the monolayer regime. Addressing the question of magnetic coupling between a EuS magnetic tunnel barrier and a CoO/Co magnetic electrode, we succeeded in realizing an independent magnetic switching behavior in this spin-valve-type system. These results are important prerequisites for future spin-dependent transport experiments. © *2011 American Institute of Physics*. [doi:10.1063/1.3549609]

INTRODUCTION

Recently, magnetic insulators receive renewed attention because of their potential for application as injectors and detectors of spin-polarized carriers in semiconductor-based spintronic devices.[1] Two approaches that are commonly used for efficient spin injection into a semiconductor (SC) are diffusive transport through an ohmic contact and tunneling transport across a barrier. An efficient ohmic spin injection is greatly hampered by the conductivity mismatch between a ferromagnetic (FM) metal and a semiconductor.[2] One solution to this fundamental problem is injecting spins via a tunneling process.[3] This approach bears several variants: Tunneling from a FM metal through a nonmagnetic (NM) barrier into a SC, from a ferromagnet into a SC via a Schottky barrier and—a largely unexplored route—from a NM metal through a ferromagnetic insulator barrier into a semiconductor.

Europium chalcogenides (EuS, EuO, etc.) belong to the rare class of materials that exhibit both ferromagnetic and insulating properties. In magnetotransport, they show the interesting spin filter (SF) phenomenon which provides a unique approach to generate highly spin-polarized tunnel currents:[4] When a ferromagnetic insulator is cooled below its Curie temperature, the conduction band splits by ferromagnetic exchange. Unpolarized electrons tunneling through a SF barrier thus experience different tunnel barrier heights, and the resulting tunnel current may be highly spin-polarized.[5]

Following this approach, the final goal is to demonstrate that spin filtering is possible with a suitable combination of magnetic insulator and semiconductor. In the present work we focus on EuS/Si(001). EuS spin filter barriers have recently attracted considerable interest due to their large spin polarizing efficiency,[6,7] as well as their good structural integrability with silicon.[8] One key requirement for establishing highly functional spin-filter tunnel contacts to silicon is the ability to prepare thin EuS films on Si(001) with well-defined magnetic properties down to the thickness regime of tunnel barriers.

A main technique of demonstrating spin filtering in electrical transport relies on utilizing a ferromagnetic counter electrode as spin analyzer and probing the magnetoresistance (MR) of the ferromagnet/spin filter junction.[9] One important prerequisite for observing sizable MR signals is a well-separated magnetic switching, i.e., a defined parallel (P) and antiparallel (AP) magnetic alignment of spin filter barrier and FM electrode. With this in mind, we assert that a proper interpretation of magnetotransport measurements needs a detailed knowledge of the structural and magnetic properties of the films.

Our present work intends to shed light onto the magnetic properties in EuS/Si(001) single layers as well as FM/EuS/Si(001) bilayer systems, that may serve as spin filter tunnel contacts to silicon. First, we have carried out a detailed study of the magnetic properties of thin EuS spin filter tunnel barriers. As the ferromagnetic electrode to the EuS/Si bilayer we have chosen CoO/Co, because it has the advantage of being one of the most widely studied exchange bias (EB) systems.[10] Furthermore, the tunneling spin polarization of a Co electrode is well known from measurements on Co/Al_2O_3/Al tunnel junctions using the Tedrow–Meservey technique,[11] and thus reduces the unknown parameters in the CoO/Co/EuS/Si system. We specifically address the issue of setting different coercive fields H_C in the spin filter and FM, and avoiding a direct magnetic coupling between them. The results illustrate the capability to design functional EuS spin filter contacts on Silicon, that are of high importance for future magnetotransport experiments.

EuS thin films were grown on (001) oriented Silicon wafer pieces under ultrahigh vacuum (UHV) conditions at a base pressure in the low 10^{-10} range. Prior to introduction into the growth chamber, the Si(001) substrates were cleaned *ex situ* by wet chemical etching in hydrofluoric acid (HF) solution. In this way, ordered and hydrogen-passivated surfaces were obtained.[12] The substrates were heated *in situ* to 300°C for outgassing. During deposition, the substrate was held at room temperature (RT). EuS was evaporated from an electron-beam heated molybdenum crucible containing stoichiometric EuS powder. The growth proceeded at a deposition rate of ~ 1 A/min, which was monitored by a water-cooled microbalance.

As magnetic counter electrodes on top of EuS we prepare thin Co/CoO bilayers following an in situ preparation procedure.[13] In a first step clean thin Co films with an initial

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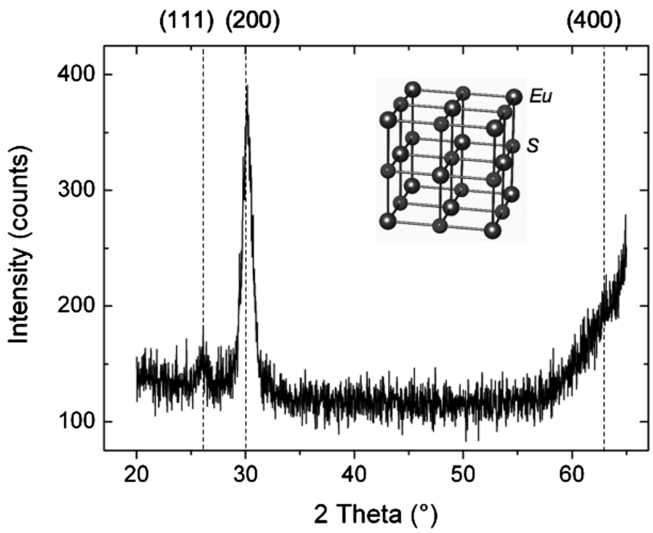

FIG. 1. X-ray diffraction (XRD) from a 15 nm thick EuS layer grown on Si(001) at room temperature (RT). The silicon substrate was tilted by 2° in order to attenuate the strong silicon (400) peak. Diffraction peaks from EuS, which crystallizes in a fcc cubic rock salt structure (see inset), are indicated.

thickness of $d = 18$ nm were deposited by e-beam evaporation and subsequently surface oxidized by a controlled exposure to oxygen at room temperature. A leak valve was used to accurately control the gas flow. A distinct oxygen exposure of 560 L (1 L $= 1.33 \times 10^{-6}$ mbar s) was set by filling the chamber with high purity O_2 gas at a pressure of 1×10^{-5} mbar. In order to prevent the samples from further oxidation in air, they were capped with 20 nm Al_2O_3 for ex situ investigations.

We start by investigating the crystalline structure of EuS/Si(001) films by x-ray diffraction (XRD). High angle $\Theta - 2\Theta$ scans collected for a typical 15 nm thick EuS film are shown in Fig. 1. The XRD spectrum indicates polycrystalline EuS, which naturally crystallizes in a face-centered-cubic (fcc) rock salt structure (inset Fig. 1). We observe the (111), (200), and (400) peaks of EuS, which match that of the EuS powder reference data. Growth of epitaxial bulk EuS on Si has only been reported for substrate temperatures above 900°C.[14]

In a next step, we address the magnetic properties of single EuS films grow on Si(001) in the thickness regime appropriate for tunnel barriers. All measurements employed a quantum design superconducting quantum interference device (SQUID) magnetometer. Figure 2(a) displays *M(H)* magnetization hysteresis loops of 1−6 nm thick EuS layers recorded at 2 K. All films with $d > 2$ nm show clear ferromagnetic behavior with a net magnetic moment of up to $\sim(6.0\pm0.4)\mu_B$ for 6 nm thick films, taking into account a EuS volume uncertainty of about 5%. M_S is reduced relative to the bulk value by approximately 15%. This observation can be assigned to the granular structure of the polycrystalline EuS films as inferred from the XRD results. A large remanent magnetization of $\sim$96% of the value predicted from the spin quantum number $S = 7/2$ is found for films $\geq$4 nm, and the films saturate in applied magnetic fields above 75 Oe. The low coercivities H_c of about 60 Oe observed for all film thicknesses with $d > 2$ nm is consistent with other reports.[15]

Figure 2(b) displays the $M(T)$ characteristics for 1−6 nm thick EuS films on Si(001) taken at an applied field of 50 Oe, which show a clear trend toward lower T_C with decreasing film thickness. The T_C of thicker EuS films approach the bulk value of 16.9 K, whereas thinner EuS films ($d \leq 3$ nm) show significantly reduced T_C. This observation was predicted as a general behavior in low-dimensional 4*f* FMs by Schiller *et al*. [see inset Fig. 2(b)].[16] It is primarily caused by weakened exchange interactions between the magnetic Eu^{2+} ions. In particular, the increasing atomic surface-to-volume ratio for the thinner films as well as the dislocation of grain boundaries cause the reduced magnetic exchange.[17] From the magnetic measurements we conclude, that our thin EuS films on Si(001) exhibit good magnetic properties down to the monolayer regime and bulklike behavior for thicknesses above $d = 3$ nm, which is an important feature for the future use as magnetic tunnel barriers on Silicon.

Now we focus on the central issue that has to be carefully considered in FM/spin filter tunnel contacts, i.e. the magnetic coupling of magnetic barrier and electrode via exchange interactions. As a consequence, they may not switch independently in an applied magnetic field. The existence of a stable antiparallel (AP) magnetic alignment between the EuS barrier and the FM electrode must therefore be confirmed prior to any magnetotransport measurement. The magnetic switching behavior of CoO/Co/EuS multilayers was once again studied by SQUID magnetometry.

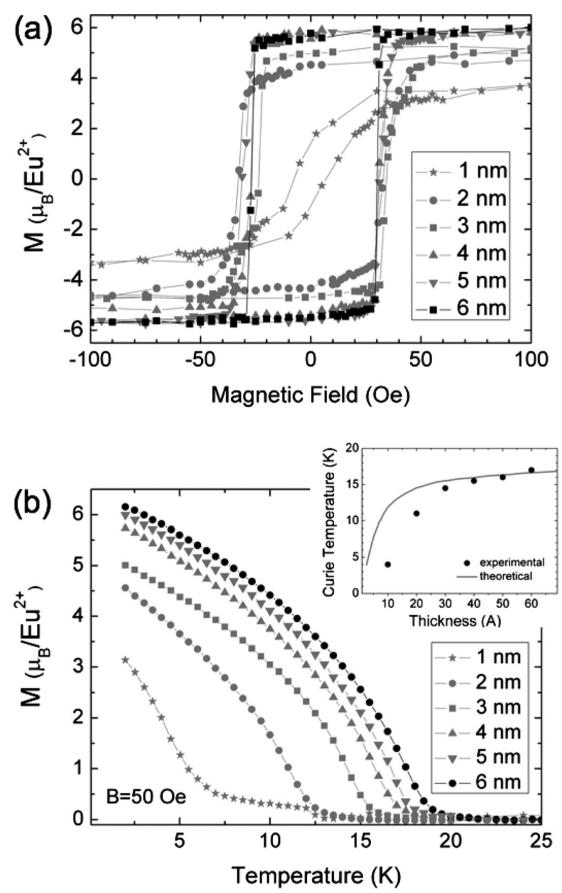

FIG. 2. (a) Hysteresis loops of 1−6 nm thick EuS films on Si(001) at $T = 2$ K. (b) Dependence of T_C on the EuS thickness ($B = 50$ Oe). The inset shows the T_C dependence compared with theoretical predictions by Schiller *et al*. (Ref. 16).

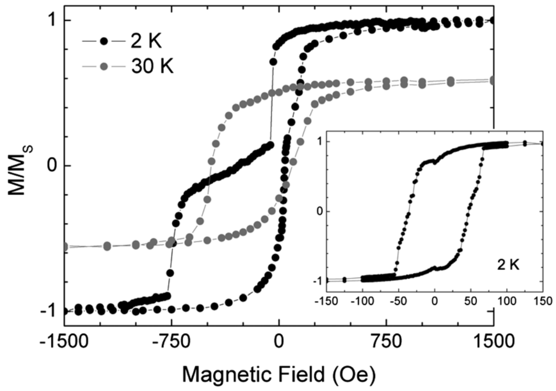

FIG. 3. Magnetic hysteresis loops *M(H)* taken at T = 2 and 30 K for CoO/Co/AlO$_x$ (1 nm)/EuS(4 nm) on Si(001). The initial Co thickness was 18 nm, that was exposed to an oxygen dose of ~560 L. Inset: *M(H)* of a Co(18 nm)/EuS(3 nm)/Si(001) system taken at T = 2 K.

Shown in the inset of Fig. 3 is a hysteresis cycle measured at 2 K, that corresponds to a typical Co/EuS(3 nm)/Si sample. The magnetization curve for this system did not show independent switching, but rather resulted in one single hysteresis loop with a coercive field of ~100 Oe. This result was expected considering the direct contact of both FM films, and indicates that the Co electrode has been blocked via an exchange coupling at the EuS interface. Thus, in the present system both magnetic layers are aligned parallel regardless of the applied magnetic field. In order to eliminate the coupling and to attain an antiparallel magnetic state, we inserted (i) an ultrathin AlO$_x$ layer in between the Co and EuS layers and (ii) pinned the Co electrode by an antiferromagnetic CoO layer. This insulating spacer has to be sufficiently thin to allow for tunneling, but thick enough to magnetically decouple both FMs. The impact of this approach is clearly demonstrated in Fig. 3: The *M(H)* curve of a CoO/Co/ AlO$_x$ (1 nm)/EuS(4 nm)/Si sample, taken at 2 and 30 K, exhibits a clear shift of the hysteresis loop from zero field, indicative of a considerable exchange bias effect in the CoO/Co bilayer. Moreover, the *M(H)* curve at 2 K contains two gradual inflection points at ±50 Oe, which correspond to a superimposed magnetic contribution of EuS. These findings suggests that no direct magnetic exchange is present between the EuS and Co layers and a magnetic decoupling is achieved by inserting the ultrathin AlO$_x$ spacer.

In order to quantify the exchange bias effect in the CoO/Co electrode in more detail, we analyzed the coercivities for decreasing and increasing external fields, H_{CA} and H_{CP}, that are −720 and +30 Oe, respectively. Using the formula $H_E = |H_{CA} + H_{CP}|/2$ the effect can be quantitatively described by an exchange bias field H_E of ~375 Oe. In order to estimate the oxide thickness in CoO/Co, we compared the total magnetic moment to a similar sample, but with a clean 18 nm thick Co layer (not shown here). The magnetic moment of CoO/Co has been reduced about 13% by the formation of the oxide surface layer. From this, it can be inferred that approximately 20 Å of the Co layer have been oxidized by an in situ oxygen exposure of ~560 L. This result is in excellent agreement with similar studies of the in situ oxidation of Co thin films.[13] The *in situ* preparation of CoO/Co/AlO$_x$/EuS multilayers on Si(001) thus resulted in a strong exchange biasing, and a spin valvelike system with defined AP and P magnetic states is at hand, opening the perspective for future MR measurements.

In summary, we have successfully prepared thin films of the magnetic insulator EuS on Si(001) substrates. The EuS films are polycrystalline and their magnetic properties show bulklike behavior for thicknesses above 3 nm, which is an important prerequisite for utilizing them as efficient magnetic tunnel barriers. We succeeded in developing CoO/Co/EuS bilayers with well defined P and AP states by inserting an ultrathin AlO$_x$ spacer layer. Further magnetotransport studies on EuS/Si(001) are therefore of undeniable interest, as they may lead to a promising development of spin filter tunnel contacts to silicon.

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