Controlling Magnetic Properties of EuS-Based Spin Valve Structures on Si(001)

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We report on the growth and magnetic characterization of the ferromagnetic insulator Europium Sulfide (EuS) on (001)-oriented Silicon substrates. The influence of the EuS film thickness on the coercive field and Curie temperature was systematically investigated with regard to a potential application of thin EuS films as spin filter tunnel barriers. In a further step, we fabricated EuS/Si(001) spin valve structures with both ferromagnetic Gd and exchange-biased CoO/Co counter electrodes. An independent magnetic switching of the EuS barrier and the ferromagnetic layers was accomplished by eliminating intermediate magnetic exchange couplings. Our results clearly demonstrate the feasibility to employ thin EuS ferromagnetic insulator films as spin filter tunnel barriers on Silicon(001) in spin valve structures for future magnetotransport experiments.

Index Terms—Europium chalcogenides, ferromagnetic insulators, spin-electronics, spin filter tunneling.

I. INTRODUCTION

O NE major challenge in the field of spintronics is the efficient generation and injection of highly spin-polarized electron currents from ferromagnetic (FM) contacts into semiconductors (SC), its manipulation, and the subsequent electrical detection using a second ferromagnetic contact [1]. This apparently simple concept forms the basis of novel spin-based electronic devices, such as spin transistors and spin-based logics. It has turned out, however, that a large conductivity mismatch at the FM/SC interface sets fundamental limits to the degree of injected spin polarization, if a ferromagnetic metal is to provide the spin-polarized carriers [2]. Subsequent studies revealed that an efficient injection of spins into semiconductors requires a high resistance interface contact, that may be realized by inserting a tunnel barrier adjacent to the semiconductor [3], [4]. Various ways to implement this concept have been proposed [5], for instance, using a ferromagnetic metal/insulating oxide contact, tailoring a dedicated Schottky barrier between a FM metal and the SC or—so far rarely investigated—making use of the spin filter effect in a ferromagnetic insulator to inject spin-polarized electrons into a semiconductor.

Spin filter tunneling is a very interesting phenomenon, both from a fundamental and technological point of view. It describes the spin-selective transport of electrons across a magnetic tunnel barrier [6]. Spin filtering was first discovered in the ferromagnetic insulator EuS, where it was shown that the exchange splitting of the conduction band creates two distinct tunnel barrier heights for spin-up and spin-down electrons [8]. Due to the exponential dependence of the tunneling current on the barrier heights, highly spin polarized charge currents may be generated that way. However, in the thickness regime typically used for tunneling, i.e., a few nanometers, the magnetism of a spin filter barrier may be suppressed [9]. The efficiency of spin filter tunneling is directly connected to the intrinsic magnetic properties of the barrier material [10]. A detailed investigation of thin ferromagnetic insulator/SC heterostructures is therefore required in order to establish highly functional spin filter tunnel contacts to a semiconductor with well-defined magnetic properties down to the monolayer regime.

The spin filter efficiency of a magnetic barrier is typically demonstrated by using one of the following experimental techniques: Either the spin polarized tunnel current is probed by a superconducting Al electrode as the spin analyzer, or a magnetoresistance (MR) measurement is performed with a FM metal electrode acting as the spin detector [11]. In the latter case, a stable antiparallel alignment between the FM metal and the EuS spin filter barrier is necessary to observe the full MR signal. This condition may be met by either choosing both FM metal and barrier with different coercive fields or by magnetically pinning the FM metal electrode by the exchange bias (EB) effect with an adjacent antiferromagnet in a spin-valve-type spin filter/semiconductor system.

In the present work, we have studied the growth and magnetism of EuS single layers and FM/EuS bilayers that may serve as efficient spin filter tunnel contacts and -spin valves to Silicon. Although EuS has a low Curie temperature of \( T_C = 16.9 \) K, the material is a well-known magnetic insulator which may serve as a model system to address the fundamental aspects associated with the spin filter approach. We have carefully investigated the thickness-dependent magnetic properties of EuS/Si(001) and of spin valves structures employing either ferromagnetic Gd or exchange-biased CoO/Co counter electrodes. The magnetic properties of the EuS/Si(001) systems were analyzed with respect to their suitability as spin filter tunnel barriers. Both FM/EuS bilayer systems yield stable antiparallel magnetic configurations, which renders them potential spin detectors in future spin filter/Si(001) transport devices.

II. SAMPLE PREPARATION

EuS films were deposited directly onto Si(001) substrates using molecular beam epitaxy (MBE) technique at a base pressure of \( \sim 1 \times 10^{-10} \) mbar. In order to create ordered and hydrogen-passivated surfaces, the Si(001) wafer pieces
were prepared ex situ by wet chemical treatment in hydrofluoric acid (HF) solution [12]. After rapid introduction into the growth chamber, the substrates were heated in situ to 300°C for outgassing. EuS was deposited at low deposition rates of approximately 1 Å/min from a stoichiometric powder by electron-beam evaporation, with the Si(001) substrate held at 300 K. Ex situ x-ray diffraction experiments confirmed the polycrystalline structure of the EuS/Si(001) films.

Ferromagnetic metal layers, either Gd or Co, were deposited by e-beam evaporation. Ultrathin Al$_2$O$_3$ spacer layers ($d = 1$ Å) were inserted between EuS and the ferromagnetic metal by e-beam evaporation from sapphire pieces. In a successive step, CoO/Co bilayers were prepared by surface oxidizing the clean Co films with initial thickness of $d = 18$ Å by a controlled exposure to oxygen [13]. Directly after Co deposition, gaseous oxygen of 99.998% purity was introduced into the MBE apparatus. A leak valve was used to accurately control the gas flow. A distinct exposure of 560 L ($1 L = 1.33 \times 10^{-6}$ m³ s⁻¹) was set by filling the chamber with O$_2$ at a pressure of $1 \times 10^{-5}$ mbar for a duration of 75 s. Both thin film deposition and oxygen exposure were performed with the substrate held at room temperature. In order to prevent the samples from further oxidation in air, they were capped with 200 Å of Al$_2$O$_3$ for ex situ investigations. The magnetic behavior was determined by hysteresis loop and temperature-dependent magnetization measurements carried out in a Quantum Design superconducting quantum interference device (SQUID) magnetometer.

III. EXPERIMENTAL RESULTS

A. EuS on Si(001)

In an initial step, we investigate the magnetic properties of single EuS films on Si(001) with thicknesses $d = 1$–6 Å typically used for tunneling. The magnetization vs. magnetic field curve $M(H)$ of a 6 nm thick EuS layer, representative of the numerous samples that were measured, is shown in Fig. 1(a). $M(H)$ shows a strong ferromagnetic behavior with a well-defined rectangular hysteresis loop. A saturation magnetic moment of $M_S \approx 6.0 \mu_B$ per Eu$^{2+}$ ion is determined at $T = 2$ K, which is reduced by about 15% relative to the bulk value of 6.9 $\mu_B$. A reason for the slightly reduced value could be a predominantly granular structure of the polycrystalline films. A large remanent magnetization of about 96% of $M_S$ is found, and saturation is achieved for applied magnetic fields above 100 Oe. The EuS films are magnetically soft exhibiting coercivities of $H_C \approx 60$ Oe only for all EuS thicknesses $d = 2$–6 nm, as shown in the inset of Fig. 1(a), while only the thinnest 1 Å EuS film shows a reduced $H_C$. This property may enable a defined magnetic switching behavior of ultrathin EuS films in magnetic multilayer systems.

Fig. 1(b) displays magnetization versus temperature $M(T)$ of the 6 nm EuS/Si(001) sample. The Curie temperature $T_C$ slightly exceeds that of bulk EuS (16.9 K), indicating the presence of free carriers within the EuS film, that may also cause the deviation of $M(T)$ from Brillouin’s law near $T_C$ [14]. As shown in the inset of Fig. 1(b), a clear trend towards lower $T_C$ is present, particularly as the EuS film thickness decreases below $d = 3$ Å. This is a general behavior of thin ferromagnetic films and was predicted by Schiller et al. [15]. The trend is caused by the lower coordination of the Eu$^{2+}$ ions at the film interfaces, as the increasing surface-to-volume ratio leads to weaker magnetic exchange interactions. Overall, our experimental results for $T_C(d)$ yield lower values than given by the theoretical curve. This tendency is reasonable when considering the polycrystalline structure of EuS, and the influence of the adjacent layer interfaces. We conclude, that the EuS/Si(001) single layers exhibit good magnetic properties down to the monolayer regime and bulk-like behavior for thicknesses above $d \geq 3$ Å, which is an important feature for their potential use as magnetic tunnel barriers on Si(001).

B. Gd/EuS/Si(001) bilayers

In the next step, we focus on a central issue that has to be carefully considered when extending the study towards ferromagnet/spin filter bilayers, i.e., that the ferromagnetic barrier and metal electrode may couple via magnetic exchange interactions, if they are in direct contact with each other. As a consequence, they may not switch independently under an applied magnetic field. We therefore aimed at establishing a stable parallel (P) and antiparallel (AP) magnetic alignment between the EuS barrier and the Gd electrode with regard to prospective magnetotransport measurements.

The magnetic hysteresis loop for a Gd (15 Å)/EuS (3 Å) bilayer sample was measured at 2 K and 30 K, as shown in
In order to eliminate the coupling between the spin filter barrier and ferromagnetic electrode, and attain a stable antiparallel magnetic state, we inserted an ultrathin AlO_x spacer between the EuS and Gd layers. This insulating film has to be continuous in order to magnetically decouple the system, but also sufficiently thin to allow for direct tunneling. The impact of a 1 nm thick AlO_x layer is demonstrated in Fig. 2(b): Two distinct switching events are observed at 2 K, one at low field corresponding to the EuS magnetization reversal, and one at higher fields corresponding to the Gd magnetization reversal. At 2 K, the EuS layer switched at approximately 40 Oe, while the Gd magnetization reversal takes place over a relatively broad range, starting at ~600 Oe and being fully reversed at ~1200 Oe, which makes the hysteresis strongly curved. In contrast to the EuS/Gd bilayers, the magnetic contribution of the EuS in the Gd/AlO_x/EuS is clearly visible and the magnetization loop may be decomposed into two independent contributions corresponding to EuS and Gd. From the 30 K curve, which resembles that of the Gd/EuS bilayer sample, it can be inferred that the switching event at the larger magnetic field must be due to Gd contributions, while EuS has a much lower coercive field. The presence of two distinct magnetization reversal events in Gd(15 nm)/AlO_x(1 nm)/EuS(3 nm) samples therefore clearly indicates that the magnetic coupling is weakened by the insulating AlO_x spacer and the Gd electrode and EuS barrier may be aligned parallel or antiparallel in a magnetotransport experiment. Our results are in good agreement with recent experiments, where an optimized magnetic decoupling between two magnetic spin filter films was accomplished for a 0.6 Å thick AlO_x spacer layer [7]. Regarding magnetotransport experiments, the ultrathin AlO_x barrier is expected to monotonically increase the total junction impedance with barrier thickness. The barrier heights for EuS and AlO_x have been determined as 0.8 eV and 1.5 eV, respectively [16]. Previous studies show a negligible influence of the AlO_x spacer layer on spin-dependent transport in the present thickness regime, since direct tunneling is dominating electron transport and only low probability of spin flip scattering inside the AlO_x barrier is expected [7].

C. CoO/Co/EuS/Si(001) spin valves

In the following we investigate a further spin-valve type system. The magnetic switching behavior of CoO/Co/EuS multilayers was once again studied by SQUID magnetometry. 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 suggests that the Co electrode was blocked by exchange coupling with the adjacent EuS layer. Thus, both magnetic layers are aligned parallel under any applied magnetic field. In the present case, we eliminate the coupling and attain an antiparallel magnetic state by combining two approaches. 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. Shown in Fig. 3 is the resulting M(H) curve of a
in developing EuS/Si(001) spin valve structures with both ferromagnetic Gd and exchange-biased CoO/Co counter electrodes. An independent magnetic switching of the EuS barrier and ferromagnetic layers was accomplished by eliminating intermediate magnetic exchange coupling. Our results clearly demonstrate the feasibility to incorporate thin EuS ferromagnetic insulator films as spin filter tunnel barriers in spin valve structures on Silicon(001). Future magnetotransport studies on these samples are of major interest, as they may lead to fundamental insights into the spin-dependent transport across spin filter/silicon interfaces.

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REFERENCES