

Electronic structure of EuO spin filter tunnel contacts directly on silicon

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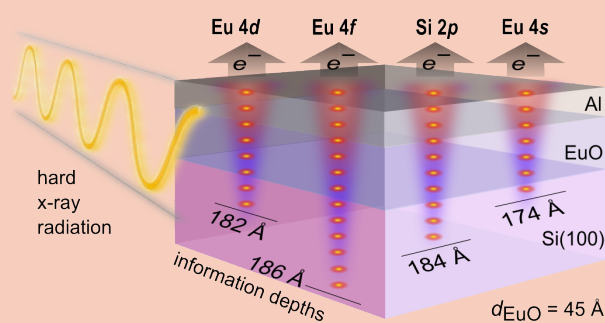
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We present an electronic structure study of a magnetic oxide/semiconductor model system, EuO on Silicon, which is dedicated for efficient spin injection and detection in silicon-based spintronics devices.

A combined electronic structure analysis of Eu core levels and valence bands using hard x-ray photoemission spectroscopy was performed to quantify the nearly ideal stoichiometry of EuO “spin filter” tunnel barriers directly on silicon, and the absence of silicon oxide at the EuO/Si interface. These results provide evidence for the successful integration of a magnetic oxide tunnel barrier silicon, paving the way for the future integration of magnetic oxides into functional spintronics devices.



Schematics of a high-energy hard x-ray photoelectron spectroscopy (HAXPES) experiment of a Al/EuO/Si heterostructure, which probes the buried EuO layer and EuO/Si interface.

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1 Introduction Accenting semiconductor electronics with spin functionality is a major thrust of current spintronics research [1]. At present, considerable efforts are being made to replace conventional ferromagnet/semiconductor (FM/SC) or FM/oxide/SC contacts—which have hitherto been underlying many key experiments [2,3]—with functional magnetic contact materials that could substantially alter the efficiency of spin injection and detection in semiconductor-based spintronics devices [4]. In particular, magnetic oxides (MO) offer the unique combination of both generating almost fully spin polarized tunnel currents via a true “spin filter” effect [5,6] and facilitating a con-

ductance matched magnetic tunnel contact to a SC [7,8]. Establishing MO tunnel contacts as efficient spin injectors and detectors bears large potential for ultimately realizing both high current transfer ratios *and* high magnetic sensitivities at the CMOS level, i. e. directly on silicon. With the goal to integrate magnetic oxide ultrathin films as efficient spin-selective tunnel barriers directly on silicon, we present a study of the electronic properties of a MO/SC model system, Europium monoxide (EuO) on Silicon, that are dedicated for the use as spin injector and spin collector contacts. EuO is predicted to be chemically stable in direct contact with silicon [9]. However, ferromagnetic EuO is

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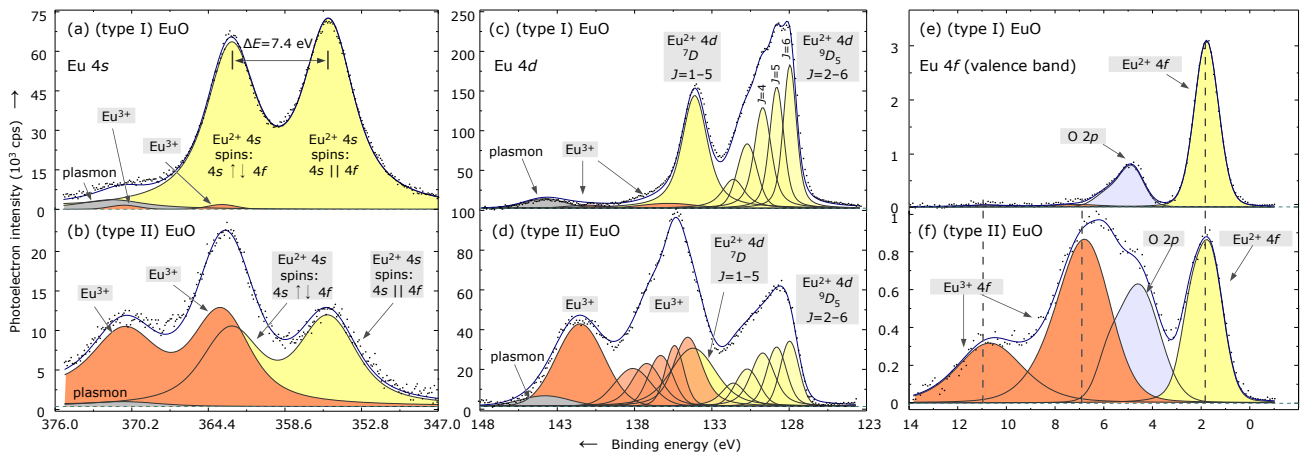


Figure 1 Hard x-ray photoemission spectra from (a), (b) Eu 4s core levels and (c), (d) Eu 4d core levels, as well as (e), (f) Eu 4f valence bands, recorded at a photon excitation energy of 4.2 keV in normal emission geometry. A quantitative peak analysis yields a relative fraction of Eu^{3+} cations of only $(3.1 \pm 1.5)\%$ for (type I) stoichiometric EuO and of $(60.2 \pm 4.8)\%$ for (type II) O-rich $\text{Eu}_{1-x}\text{O}_{1+x}$.

very difficult to synthesize due to its instantaneous reactivity towards higher, antiferromagnetic oxides, i. e. Eu_2O_3 , under oxygen atmosphere. Furthermore, excess oxygen may readily oxidize the silicon surface, which can lead to a loss of injected and detected tunneling spin polarization. We succeeded in preparing high-quality EuO/Si(001) heterostructures with bulk-like magnetic properties [10]. We used hard x-ray photoelectron spectroscopy (HAXPES) to probe the electronic structure of EuO tunnel barriers and the EuO/Si interface. Due to its large information depth—exceeding 10 nm at photon energies of 10 keV [11]—HAXPES is a perfectly suited technique to probe the chemical quality of buried layers and interfaces.

2 Experimental Details Stoichiometric EuO (type I) and O-rich $\text{Eu}_{1-x}\text{O}_{1+x}$ (type II) ultrathin films were prepared directly on Si(001) without any buffer layer using molecular beam epitaxy under ultrahigh vacuum conditions ($p_{\text{base}} \leq 10^{-10}$ mbar). Prior to deposition, Si(001) substrates were etched in diluted hydrofluoric acid in order to remove the native SiO_2 layer and prepare a hydrogen terminated H-Si(1 \times 1) surface, which was confirmed by low-energy electron diffraction. 40 Å thick EuO films were synthesized by evaporating pure Eu metal (99.98%) in a molecular O_2 atmosphere, employing a Europium distillation process at $T_S = 350^\circ\text{C}$ with a persistently controlled O_2 partial pressure during the entire EuO synthesis [12, 13]. In this way, polycrystalline EuO with the different stoichiometries (type I) and (type II) was generated by reactive MBE. Finally, a 40 Å thick Al capping layer was deposited to prevent further EuO oxidation.

HAXPES experiments were carried out at the undulator beamline P09 at PETRA III (DESY Hamburg) using a photon excitation energy of 4.2 keV and multichannel hemispherical electron energy analyzer (SPECS Phoibos 225 HV). The spectra were recorded at room temperature in

normal emission geometry with an energy resolution of 0.5 eV. The binding energy scale was calibrated to the metallic Fermi edge of an Au foil in contact with the sample. A Shirley-type background was subtracted from the raw data to correct inelastic photoelectron scattering [11].

3 Results and Discussion

3.1 Core-level and valence band HAXPES By studying photoemission spectroscopy from both core and valence levels, we obtain a comprehensive set of information on the electronic structure of EuO/Si(001). Fig. 1 (a)–(f) shows the Eu 4s and Eu 4d core levels, as well as the Eu 4f valence bands for (type I) stoichiometric EuO and (type II) O-rich $\text{Eu}_{1-x}\text{O}_{1+x}$ on Si. Eu cations in stoichiometric, ferromagnetic EuO are divalent, whereas Eu^{3+} antiferromagnetic contributions are expected in O-rich EuO. A quantitative peak analysis of the divalent and trivalent spectral contributions in the Eu 4s, 4d and 4f spectra is employed to extract the EuO chemical composition.

In a first step, we assign the individual spectral features observed in the Eu core level and valence spectra. Fig. 1 (a) and (b) shows the Eu 4s core level spectra for both (type I) and (type II) EuO compounds. The prominent double-peak structure is caused by coupling of the 4s core level with the localized Eu 4f state, which leads to an exchange splitting $\Delta E = 7.4$ eV of the 4s inner shell. For (type I) EuO, the 4s double-peak is assigned to divalent Eu^{2+} spectral contributions, whereas an additional, overlapping trivalent Eu^{3+} double-peak feature appears in Fig. 1(b), that is chemically shifted by 8.1 eV towards higher binding energy.

The Eu 4d core level spectra are depicted in Fig. 1(c) and (d). They show a complex multiplet structure distributed in a wide energy range due to the strong 4d–4f exchange interaction and much weaker 4d spin-orbit splitting. Therefore, the 4d spectra cannot be separated into their $4d_{3/2}$ and $4d_{5/2}$ components. We can attribute the two main

spectral contributions of the $4d$ spectra to a $J = L - S$ multiplet splitting caused by $4d-4f$ interaction, and assign the peaks to the 7D and 9D multiplets, respectively. The fine structure of the 7D final state is not resolved, whereas the $J = 2-6$ components in the 9D state is easily identified. Once more, we clearly observe a mainly divalent Eu^{2+} valency in (type I) EuO, but significant spectral contributions from Eu^{3+} cations in (type II) O-EuO.

We move on to the analysis of the Eu $4f$ valence bands in Fig. 1 (e) and (f). The pronounced peak located at 1.8 eV below E_F displays the Eu^{2+} $4f$ final state multiplet. Spectral contributions up to 4.5 eV binding energy are attributed to valence bands of overlapping Eu^{2+} $4f$ and O $2p$ states. The chemically shifted Eu^{3+} $4f$ multiplet structure in (type II) O-rich EuO is located between 5 and 13 eV.

Finally, we performed a quantitative peak analysis by fitting the spectral contributions with convoluted Gaussian-Lorentzian curves. The result of the fitting procedure is shown by the solid lines in Fig. 1(a)–(f), which match the experimental data points very well. From the integrated spectral intensities of the divalent Eu^{2+} and trivalent Eu^{3+} components, we derive a relative fraction of Eu^{3+} cations of only $(3.1 \pm 1.5)\%$ for (type I) stoichiometric EuO and of $(60.2 \pm 4.8)\%$ for (type II) O-rich $\text{Eu}_{1-x}\text{O}_{1+x}$.

3.2 Chemical state of the EuO/silicon interface

In a next step, we probe the local chemistry and bonding at the EuO/silicon transport interface. Photoemission from the Si $2p$ core level was recorded in normal (0°) and off-normal (45°) electron emission geometry, using a photon energy of $h\nu = 4.2$ keV. In this way, the information depth of the escaping photoelectrons is substantially varied between ~ 184 Å and ~ 132 Å, respectively, which allows one to distinguish spectral contributions from bulk and interface-like electronic states of the buried Si substrate.

For (type I) stoichiometric EuO/Si(100), we observe a well-resolved Si $2p$ doublet structure for both electron emission geometries in Fig. 2 (a) and (b), which is indicative for an integral Si^0 valency both in the bulk and interface regions of the substrate. Photoemission contributions from other Si $2p$ valences are absent in these spectra, which confirms the chemical stability of the EuO/Si(001)

interface as predicted by thermodynamic calculations [9]. In (type II) O-rich EuO/Si(100), in contrast, besides the sharp Si^0 $2p$ peak a broader feature can be observed on the high binding energy side in Fig. 2 (c) and (d). This feature is attributed to the emission from Si^{4+} $2p$ states, which indicates the presence of SiO_2 . The chemical shift of $\Delta E = (3.8 \pm 0.1)$ eV compares very well with literature [14]. The Si^{4+} $2p$ spectral intensity is enhanced for off-normal emission, which confirms that the SiO_2 signal mainly originates from the EuO/Si interface.

We finally conclude, that the chemical state of the EuO/Si(001) interface directly correlates with the specific EuO growth conditions at elevated substrate temperatures. In particular, any oxygen excess during EuO synthesis not only leads to the formation of antiferromagnetic Eu_2O_3 , but also promotes an oxidation of the EuO/Si interface. Only if the Eu distillation process and specific range of oxygen supply is precisely matched during synthesis, high quality EuO thin films can be grown directly on Si without interface oxidation, and thus can be integrated on silicon as efficient “spin filter” tunnel injector and collector contacts.

4 Summary We presented an electronic structure study of a magnetic oxide/semiconductor model system, EuO on Silicon. We succeeded in stabilizing ultrathin EuO films on Si(001), and confirmed their nearly ideal stoichiometry using core-level and valence band hard x-ray photoemission spectroscopy. Moreover, we identified a chemically stable EuO/Si interface, with no signs of silicon oxide formation. With the aim to establish this novel class of spin injector/collector contacts on silicon, high-quality EuO magnetic oxide tunnel barriers will be integrated into silicon-based transport devices in the near future.

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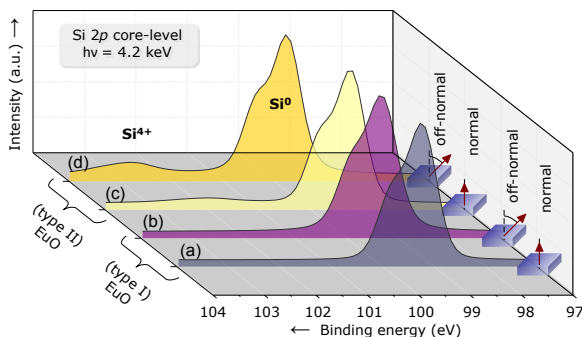


Figure 2 Si $2p$ core level photoemission spectra for (type I) stoichiometric EuO and (type II) O-rich EuO, recorded at 4.2 keV in normal (0°) and off-normal (45°) electron emission geometry.

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