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EuO epitaxy by oxygen scavenging on SrTiO₃ (001): Effect of SrTiO₃ thickness and temperature

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The EuO/SrTiO₃ heterojunction is a promising combination of a ferromagnetic material and a two-dimensional electron system. We explore the deposition of Eu metal on SrTiO₃/Si pseudo-substrates, with varying SrTiO₃ (STO) thickness, under ultrahigh vacuum conditions. By varying the thickness of the STO layer (2–10 nm) and the deposition temperature (20–300 °C), we investigate the process by which oxygen is scavenged from STO by Eu. *In situ* x-ray photoelectron spectroscopy is used to investigate the electronic structure of the nominal Eu/STO/Si stack. We find that as a result of Eu deposition, epitaxial EuO is formed on thick STO (6–10 nm), leaving behind a highly oxygen-deficient SrTiO₃ layer of ~4 nm in thickness. However, if the thickness of the STO layer is comparable to or less than the scavenging depth, the crystal structure of STO is disrupted and a solid state reaction between Eu, Si, and STO occurs when the deposition is done at a high temperature (300 °C). On the other hand, at a low temperature (20 °C), only a 1–2 nm-thick EuO interlayer is grown, on top of which the Eu metal appears to be stable. This study elucidates the growth process under different conditions and provides a better understanding and control of this system. *Published by AIP Publishing.* <https://doi.org/10.1063/1.5059560>

INTRODUCTION

The ferromagnetic semiconductor EuO is a promising material for spintronic devices^{1–3} due to its very large magnetic moment (7 μ_B per Eu) and large spin-splitting of the conduction band leading to almost 100% spin polarization of carriers.^{4,5} The semiconducting behavior combined with the high magnetic moment makes it an ideal spin-filter material below its Curie temperature of 69 K.⁶ EuO has been epitaxially integrated onto graphene,⁷ silicon,^{1,8} GaN,¹ and several oxide substrates⁹ to provide ferromagnetism in these systems. In addition, EuO also exhibits large magnetoresistance, strong magneto-optical effect, and unusual transport properties.^{1,8–11} These features make EuO a fascinating oxide for fundamental research, in addition to the possibility of enabling new kinds of device applications involving spin transport.

The integration of EuO on heterostructures exhibiting a two-dimensional electron gas (2DEG) is a potentially interesting approach to explore the interaction of strong magnetism with the sheet charge, particularly for the case of the 2DEG at SrTiO₃ (STO) interfaces.^{9,12,13} By controlling the deposition of an oxide with a large negative enthalpy of formation like EuO on STO,¹⁴ one can stabilize a highly conductive layer of oxygen-deficient STO at the interface.¹⁵ This heterostructure then offers a way to combine strong ferromagnetism and a 2DEG in one system. Kormondy *et al.* have recently reported the observation of a large linear positive magnetoresistance in the 2DEG at the

EuO/SrTiO₃ interface.⁹ Lömker *et al.* also report a magnetically tunable two-dimensional electron system in the same structure.¹² Prinz *et al.* created quantum wells with ultrathin EuO layers and widened the bulk EuO bandgap from 1.19 eV to ~1.4 eV in ultrathin films by the quantum confinement effect.¹⁶

Growing thin films of high quality stoichiometric EuO is difficult because of the high stability of the competing Eu₂O₃ phase.^{6,9} Thin-film growth of EuO has been reported in several studies,^{6–9,12,17,18} and it is crucial to control the oxygen partial pressure in order to form stoichiometric EuO and prevent over-oxidation.¹⁸ Alternatively, stoichiometric EuO can be grown on certain substrates under Eu adsorption-controlled conditions.^{7,8,19} EuO integration on Si has been reported by several groups,^{6,8,18,20} and the EuO/Si interface has been predicted to be thermodynamically stable if the two materials are in contact.²¹ Lettieri *et al.* reported an epitaxial EuO film grown on Si by molecular beam epitaxy (MBE) using an SrO buffer.¹⁸ Sr deoxidation is used to remove native SiO₂^{22,23} and 5 monolayers (ML) of SrO are grown to provide a lattice-matched surface for EuO growth (0.3% mismatch). Caspers *et al.* grew polycrystalline EuO directly on HF-treated, H-passivated Si substrates by precisely controlling the Eu and oxygen flux.⁶ These authors also grew epitaxial EuO [001] on Si[001] by passivating the Si surface with a 13-Å silicon suboxide (SiO_x) buffer layer prior to EuO deposition.²⁰ However, this process also led to interfacial silicide formation. Direct EuO growth on Si was reported by Averyanov *et al.* who used sub-monolayer reconstructions of Eu on Si to create a Zintl-like template to avoid Si oxidation.⁸ There are also reports of epitaxial EuO growth on STO with buffer layers

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such as BaO or SrO to prevent over-oxidation of EuO due to oxygen out-diffusion from STO.^{24,25}

Common to all reported growth methods, EuO growth is typically performed at temperatures between 300 and 450 °C. Furthermore, controlling the oxygen pressure is always crucial to form high quality, stoichiometric EuO. Posadas *et al.* found that EuO can be grown directly on an STO substrate by Eu deposition in ultra-high vacuum (UHV) without providing any oxygen.¹⁴ It has been shown that when metals with large enthalpy of oxide formation (such as Al and Eu) are deposited on STO substrates, oxygen can be scavenged from the top STO layers and form oxides on top even without additional oxygen. The oxygen vacancies created in STO create a conductive layer on the STO side of the interface. This conductive layer in STO due to oxygen vacancies has also been reported in other studies.^{9,14,26}

Eu, with very large oxide formation enthalpy and a low work function of 2.5 eV, is expected to scavenge significant amounts of oxygen from STO to form EuO, leaving behind a highly conductive oxygen-deficient SrTiO₃ layer.¹⁴ Kormondy *et al.* used this process to grow epitaxial EuO on STO single-crystal substrates at 300 °C in UHV. The spin-polarized 2DEG formed at the EuO/STO interface displayed positive linear magnetoresistance when the EuO was below its Curie temperature. From electron energy loss spectroscopy (EELS) analysis in cross-sectional scanning transmission electron microscopy, the oxygen-deficient SrTiO₃ layer was observed to be ~4 nm deep.⁹ This thickness appears to be the “oxygen scavenging depth” of Eu when deposited on STO at a temperature of 300 °C. It is expected that this depth will vary strongly with temperature since oxygen scavenging involves atomic diffusion. Therefore, it is important to investigate the effect of temperature, and also STO layer thickness, on the scavenging of oxygen in STO by Eu. In particular, it would be interesting to establish what happens when the STO thickness becomes comparable to or smaller than the oxygen scavenging depth. In other words, how much oxygen can Eu scavenge from STO before STO starts to be degraded?

EuO has a cubic rock-salt structure with a lattice constant of 5.14 Å.²⁷ The lattice matching of EuO to a cubic perovskite structure requires a 45° in-plane rotation, essentially lining up with the rock salt-like layers of the perovskite. The nominal matching is therefore between [100] direction of the perovskite and [110] direction of rock salt. Schematics of the matching interface structure are displayed in Fig. 1. In the case of EuO matching with STO ($a = 3.905$ Å), the lattice mismatch is ~7%.

In this paper, we report on the fabrication of EuO/SrTiO₃/Si heterostructures by depositing metallic Eu under UHV conditions on STO/Si pseudo-substrates with varying STO layer thickness. Different STO thicknesses allow us to place a limit on the amount of available oxygen for reaction with Eu. We investigate the oxygen scavenging mechanism by varying both the STO thickness (from 2 to 10 nm) and the temperature of Eu deposition (from 20 °C to 300 °C). Crystallinity of the growing film is monitored by *in situ* reflection high-energy electron diffraction (RHEED). We analyze the composition of the resulting structure by *in situ* x-ray photoelectron spectroscopy (XPS).

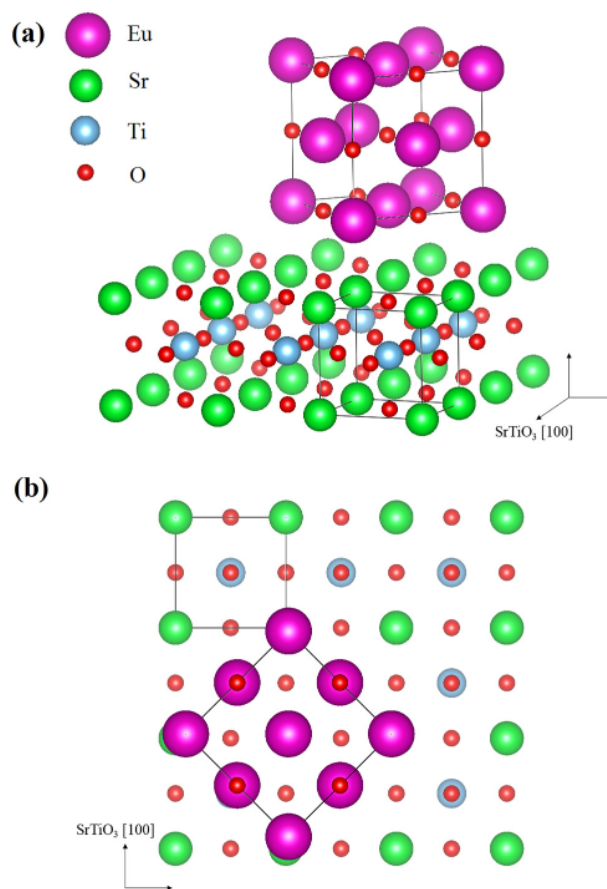


FIG. 1. Schematic of EuO/STO epitaxial arrangement: (a) side view; (b) top view.

EXPERIMENT

The deposition is performed in a customized DCA 600 MBE system with a base pressure of 6×10^{-10} Torr. The Si substrates (p-type with a resistivity of 0.1–0.2 Ω cm from University Wafer) are diced into 20×20 mm² pieces and degreased ultrasonically in acetone, isopropanol, and deionized water (18 M Ω cm) for 5 min each and then exposed to UV/ozone to remove carbon from the surface. The Si substrates are then annealed in the vacuum chamber at 700 °C for 10 min followed by the growth of a 2-nm-thick STO epitaxial film at 250 °C which is crystallized by annealing in vacuum at 500 °C.^{21,28,29} The STO films are grown such that there is no interfacial SiO₂ present by separating the oxygen gas introduction with the STO crystallization anneal.^{28–31} Additional STO layers can be grown without SiO₂ formation by depositing disordered STO at 250 °C using co-deposition under moderate oxygen pressure (~mid 10^{-7} Torr), and then annealing in UHV at 500 °C to further crystallize the STO film. It is important to prevent SiO₂ formation to ensure that the only oxygen source for scavenging by Eu is from the STO layer.

We prepared STO/Si pseudo-substrates with STO layer thicknesses of 2, 3, 4, 6, and 10 nm. For each STO layer thickness, different coverages of Eu are deposited from an effusion cell with the cell temperature fixed at 530 °C.