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Epitaxial (001) EuO thin films have been grown on (001) Si utilizing an intermediate, epitaxial SrO buffer layer by molecular-beam epitaxy. Four-circle x-ray diffraction reveals nearly phase-pure samples. Magnetic measurements indicate that the EuO layer is ferromagnetic, with a transition temperature (68 K) close to the bulk value and a saturation magnetic moment of 4.7 Bohr magnetons per Eu atom. The magneto-optic Kerr effect observed is also comparable to bulk EuO. Such heterostructures have potential as a means to inject spin-polarized electrons into silicon for use in spintronics applications. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1593832]

Injection of spin-polarized electrons into a semiconductor serves as one of the most fundamental requirements in utilizing coherent spin phenomena in spin-based electronics. Although heterostructures and methods have been extensively developed for spin injection of carriers into III-V semiconductors,[1–6] analogous studies aimed at developing heterostructures for the injection of spin-polarized carriers from a ferromagnet into silicon are infrequent.[7,8] Given the weak spin–orbit coupling for electrons in silicon and the relatively long transverse decoherence times (T_2) observed for bound states,[9] injection and manipulation of carriers in silicon presents a system ripe with potential for spintronics applications. In this letter, we discuss the growth of a ferromagnetic/silicon heterostructure, specifically the growth of epitaxial EuO on silicon, with this application in mind.

EuO is the only known binary ferromagnetic oxide ($T_C = 69$ K)[10,11] predicted to be thermodynamically stable in contact with silicon.[12] Electrically, this semiconducting ferromagnet is extremely versatile. Oxygen-deficient EuO, for example, exhibits an insulator-to-metal transition below T_C and colossal magnetoresistive (CMR) behavior surpassing the manganite-based perovskite CMR oxides.[13,14] Its specific Faraday rotation ($\sim 5\times10^5$ °/cm at 632.8 nm) is one of the highest of any known material.[15] In addition, recent studies using spin-resolved x-ray absorption spectroscopy have shown that the conduction band of EuO (in its ferromagnetic state) is split by about 0.6 eV, causing the electrons close to the conduction band edge to be nearly 100% spin polarized.[16] This makes EuO a very attractive material for the injection or detection of spin-polarized electrons.

Despite the versatility of this compound, a relatively small body of work exists on epitaxial EuO thin films. EuO is known to grow epitaxially on CaF_2,[17] yttria-stabilized cubic zirconia (Y_2O_3–ZrO_2),[18] MgO,[19] and $SrTiO_3$ substrates.[19] Unlike most of the lanthanides, europium can be oxidized to a 2+ state and readily forms an oxide with a rock-salt structure. Consequently, the growth of EuO is expected to resemble the growth of the alkaline-earth oxides (AEOs), which have previously been demonstrated to grow epitaxially on silicon.[20–22]

All films in this study were grown by molecular-beam epitaxy (MBE) in an EPI 930 MBE chamber modified for the growth of oxides.[22] Alkaline earth (AE) metals (barium and strontium) and europium were sublimated from low-temperature effusion cells. The oxidant was molecular oxygen (99.994% pure). The deposition rate was ~1 monolayer (ML)[23] per 10–20 s. Films were grown on 2-in.-diam (001) Si.

Given the penchant for europium oxide to behave in a fashion similar to the AEOs, the submonolayer reconstructions and transition steps from the silicon to the oxide via an intermediate silicide layer described in Refs. 21 and 22 were investigated using europium. Very similar reconstructions were observed with europium metal[24] on the reconstructed silicon surface (not shown). Indeed, the bulk crystal structures of $EuSi_2$, $SrSi_2$, and $BaSi_2$ are closely related.[25] To test the ability of the europium silicide to act as a template for the epitaxial growth of an oxide, a lattice-matched (Ba,Sr)O layer was grown on it. Figure 1 shows a reflection high-energy electron diffraction (RHEED) image of 5 ML of (Ba,Sr)O grown using an interfacial europium silicide layer[24] [the growth of (Ba,Sr)O is described in Ref. 22]. This result is a demonstration of a non-AE silicide to effect the transition from silicon to epitaxial oxide.

Although, AEOs can be grown epitaxially at room temperature and cooler[22,26] (i.e., in a regime amenable to the growth of epitaxial oxides on silicon), the same is not true for epitaxial EuO. EuO films grown at room temperature on silicon with an intermediate europium or strontium silicide layer[24] were amorphous. The limiting factor appears to be the need for higher substrate temperatures to crystallize EuO.[27] Growth on (100) Y_2O_3–ZrO_2 substrates revealed

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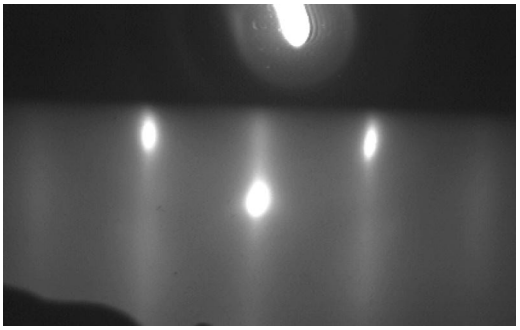

FIG. 1. RHEED image along the [110] azimuth [of both (Ba,Sr)O and the substrate] of 5 ML of (Ba,Sr)O grown at room temperature on (001) Si using an interfacial europium silicide layer.

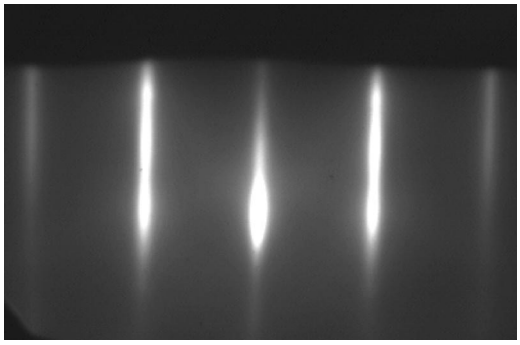

FIG. 2. RHEED image along the [110] azimuth of EuO after the growth of 660 Å of EuO on a 5-ML buffer layer of SrO on (001) Si on which an interfacial strontium silicide layer was grown.

that substrate temperatures of ~400–450 °C in an oxygen background pressure of $(1-2)\times10^{-8}$ Torr were necessary for epitaxial growth at the growth rates used in this study.[28] The direct growth of EuO on europium or strontium silicide[24] on silicon under these conditions, however, did not yield epitaxial EuO films, presumably due to oxidation of the underlying silicon.

One solution to address this issue is to use a silicon-compatible buffer layer that can be deposited epitaxially at low temperature to move away from the silicon surface to help minimize unwanted silicon oxidation. Given the ability to grow epitaxial SrO and BaO on (001) Si at room temperature[21,22] and the similarities in lattice constant and structure among these materials,[29,30] an AEO buffer layer is an obvious choice. A number of different buffer layers and growth schemes were attempted to grow an optimized EuO layer on an AEO buffer layer. The buffer layers were grown using the process outlined in Ref. 22 for the growth of AEOs, and all the EuO layers were grown at ~400–450 °C in $1-2\times10^{-8}$ Torr O_2 [i.e., the minimum temperature and background pressure found to be successful for the growth of epitaxial EuO on single-crystal (Y_2O_3–ZrO_2) substrates in our system].[28] The oxygen pressure during growth is important as europium can exist in oxidation states ranging from Eu^{2+} to Eu^{3+} and correspondingly assumes structures other than rock salt, that is, Eu_2O_3 and Eu_3O_4.[31]

Due to the excellent lattice match between EuO and SrO (0.3% mismatch) compared to BaO (7.2% mismatch), we restricted our study to the use of SrO as a buffer layer. Epitaxial growth of EuO on top of this stack (SrO on europium or strontium silicide[24] on silicon) was found to require elevated growth temperatures (~400–450 °C) for the growth rates used. At these temperatures, we observed that the SrO buffer layer began to degrade during heating in ultrahigh vacuum above a critical temperature. The degradation, which was manifested by a reduction in RHEED intensity, was presumably due to a reaction between SrO and silicon, resulting in the formation of strontium silicate [analogous to the reaction between (Ba,Sr)O and silicon].[22] With this in mind, the thickness of the SrO buffer layer was varied. The structural quality of the resulting epitaxial EuO films, based on rocking curve full width at half-maximum (FWHM) from x-ray diffraction of the 002 EuO peak, was highest for SrO buffer layers with thicknesses $\geq$5 ML. The epitaxial EuO film discussed in detail below was grown on a 5-ML-thick SrO buffer layer.

Figure 2 shows a RHEED image at the end of the growth of a 660 Å thick EuO film on a 5-ML buffer layer of SrO on strontium silicide[24] on silicon. Figure 3(a) shows a θ–2θ scan of this same film. The FWHM in 2θ and ω of the 002 EuO reflections are 0.30° and 0.93°, respectively. (Because of its sensitivity to air and water, the EuO film was capped with 1300 Å of aluminum before removal from the MBE chamber.) The ϕ scan of the 202 EuO reflection (FWHM in ϕ of 1.2°) shown in Fig. 3(b) corroborates that the EuO is epitaxial and oriented with a cube-on-cube orientation relationship, i.e., (001) EuO‖(001) Si and [100] EuO‖[100] Si.

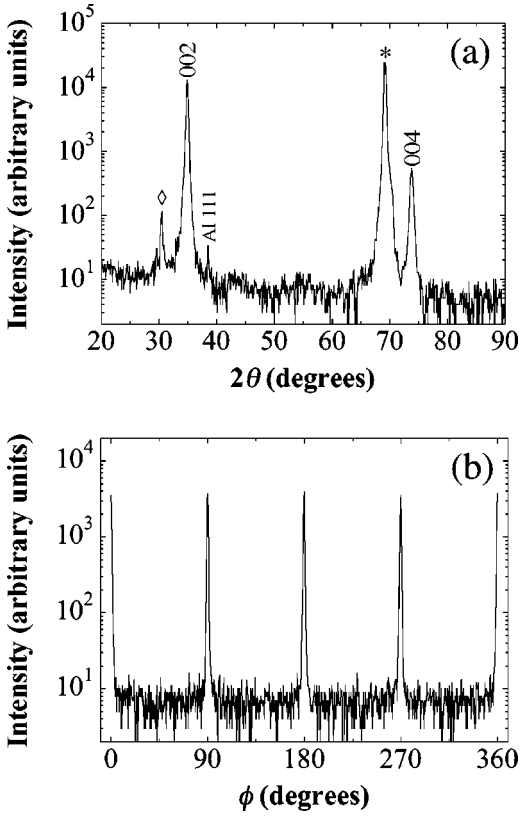

FIG. 3. (a) θ–2θ x-ray diffraction scan and (b) ϕ scan of the 202 EuO reflections of the same film whose RHEED image is shown in Fig. 2. $\phi=0°$ corresponds to the in-plane component of the diffraction vector aligned parallel to the [100] direction of the silicon substrate. From these scans, the in-plane and out-of-plane lattice parameters of the EuO film are $a=5.14\pm0.03$ Å and $c=5.13\pm0.01$ Å, respectively. The intense 002ℓ peaks from EuO are labeled. The 004 Si substrate peak is labeled as (*). The peak marked ◇ is from the 320 reflection of Eu_3O_4.

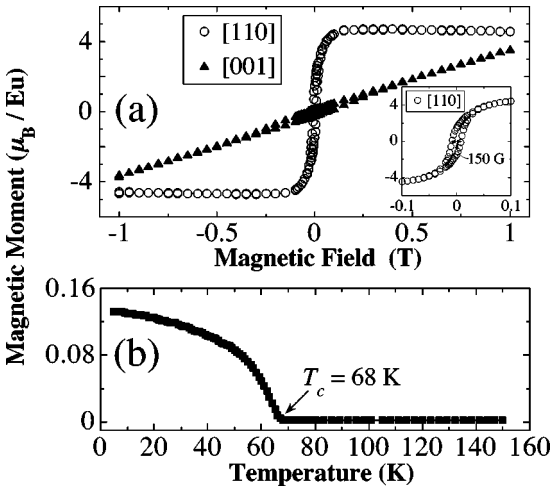

FIG. 4. (a) In-plane **B**‖[110] and out-of-plane **B**‖[001] hysteresis loops taken at $T=10$ K. Inset depicts zoomed in [110] data showing remanence of ~1.5 μB per Eu atom and coercivity of ~150 G. (b) Zero-field warming curve with the sample oriented along the [110] direction reveals a Curie temperature of ~68 K.

Magnetic measurements were performed on the same sample using a commercial superconducting quantum interference device magnetometer. Figure 4(a) shows hysteresis scans taken at 10 K both in-plane with **B** parallel to [110] and out-of-plane with **B** parallel to [001]. The [110] scan reveals that the film is ferromagnetic with a coercivity of ~75 G, remanence of ~1.5 Bohr magnetons (μ_B) per Eu atom, and saturation magnetic moment of ~4.7 μ_B per Eu atom. Strong shape anisotropy is present in this sample due to its thin-film nature and can clearly be seen in the [001] field scan, which shows that the sample is not fully saturated even at 1 T, as well as negligible hysteresis. The zero-field warming curve with the sample oriented along [110], shown in Fig. 4(b), shows that the remanence decreases to zero at approximately 68 K, which is near the bulk value of T_C for EuO.[10,11] Similar data taken for samples oriented with **B**‖[100] indicated no significant in-plane anisotropy.

EuO has one of the largest magneto-optical responses ever reported.[15] The origin of this response comes from the large exchange splitting in the ferromagnetic state. Polar magneto-optical Kerr effect measurements of the same 660-Å-thick EuO film are shown in Fig. 5. The polar Kerr rotation saturates at ~0.15° near 2 T, consistent with magnetization measurements. The saturation value of the Kerr rotation is smaller than but comparable to other values reported,[15,17] and may be attributable to interference effects at the Al/EuO and EuO/Si interfaces.

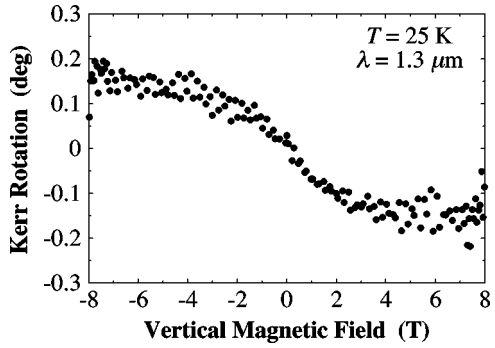

FIG. 5. Polar Kerr response of the same film whose RHEED image is shown in Fig. 2, measured at $T=25$ K and $\lambda=1.3$ μm.

The epitaxial integration of EuO with silicon via a thin intermediate epitaxial SrO buffer layer is demonstrated. The SrO layer can be made sufficiently thin for this insulator (with 5.3-eV band gap at low temperature)[30] to function as a tunnel barrier. The efficacy of epitaxial EuO/SrO/Si heterostructures for the injection of spin-polarized electrons into silicon remains to be determined.

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