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Direct epitaxial integration of the ferromagnetic semiconductor EuO with Si(1 1 1)

Dmitry V. Averyanov *, Ivan S. Sokolov, Andrey M. Tokmachev, Igor A. Karateev, Oleg A. Kondratev, Alexander N. Taldenkov, Oleg E. Parfenov, Vyacheslav G. Storchak *

National Research Center "Kurchatov Institute", Kurchatov Sq. 1, Moscow 123182, Russia

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ABSTRACT

Facing physical limits to further development, modern electronics explores alternative approaches. One of the most promising routes is offered by spintronics employing spin degree of freedom. Silicon spintronics is especially important due to the central technological role of Si. This technology requires non-equilibrium spin polarization in non-magnetic Si. The ferromagnetic semiconductor EuO has been justified as a promising candidate for electrical spin injection into Si. Here, we report the first fabrication of the EuO(1 1 1)/Si(1 1 1) structure – a magnetic material with a polar heterointerface – employing a special synthetic procedure. Structural characterization proves an atomically sharp interface and monocrystalline quality of the films while magnetic measurements match the bulk properties of EuO.

1. Introduction

Growing demands for speed, information capacity and energy efficiency call for continuous improvement of electronic devices. In this quest, traditional electronics encounters fundamental physical limitations [1]. Further progress is expected to be based on new physical principles and new materials. Spintronics, exploiting spin degree of freedom along with charge, is at the center of rapidly developed alternative electronics. Implementation of spin functionality provides new concepts of electronic devices and gives advantages in terms of the energy consumption. Spintronics is dominated by metallic materials starting from successful application of giant magnetoresistance in the development of memory devices [2]. However, spin-based elements of logic require semiconductors [3].

A special class of materials, dilute magnetic semiconductors, has been developed to match spintronic functionality with semiconductor devices. However, they are not widely applied in spintronic devices due to weakness of magnetic properties and their inhomogeneity [4,5]. A major alternative is to build spintronic devices on the base of mainstream semiconductors: within this approach only minor technological changes are necessary. As a workhorse of modern electronics silicon is, undoubtedly, a major candidate [6,7]. Silicon supports spin transport due to long spin coherence lifetime. However, nonmagnetic character of Si demands nonequilibrium spin polarization to be created, preferably by electrical injection. The use of widespread ferromagnetic contact metals as a source of spin-polarized electrons is complicated by the problem of conductivity mismatch [8]. Though significant progress has been made in spin injection through a tunnel barrier [6,7] the degree of spin polarization in Si is still insufficient for industrial applications, giving rise to approaches based on injection from a ferromagnetic semiconductor contact [9].

The ferromagnetic semiconductor EuO is probably the most promising material for such a contact. It has a simple face centered cubic (fcc) rock-salt structure. Its Heisenberg-like magnetism comes from half-filled 4*f*-states [10]. Unlike dilute magnetic semiconductors, magnetic properties of EuO are sound (7 μ_B per Eu^{2+} ion) and uniform. Due to enormous spin splitting of conduction band (~0.6 eV) it shows nearly 100% spin polarization below the Curie temperature of 69 K. The conductivity of EuO can be tuned to that of Si by doping with a variety of elements as well as oxygen vacancies. EuO exhibits a number of outstanding properties: metal-insulator transition accompanied by 13 ÷ 15 orders of magnitude change in resistivity [11], colossal magnetoresistivity effect of about 6 orders of magnitude in a magnetic field of 2 T [12], exceptionally strong magneto-optics effects [13] *etc.* These properties could provide additional functionality to spintronic devices.

* Corresponding authors.
E-mail addresses: dm_averyanov@mail.ru (D.V. Averyanov), mussr@triumf.ca (V.G. Storchak).

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Therefore, a number of attempts have been undertaken to produce a EuO/Si spin contact [14–18]. Recently, EuO has been successfully grown directly on Si(0 0 1) without any alien phases at the atomically sharp EuO/Si interface [19,20]. The applicability of this EuO/Si (0 0 1) contact to spintronics has been justified with angle-resolved photoelectron spectroscopy [21].

When the components (EuO and Si) of the heterostructure are chosen it is not very much can be done to modify the spin contact interface. A major opportunity is to change the interfacing facets. Being viewed along the (1 1 1) direction, EuO constitutes a stack of alternating oxygen and europium layers; the sequence of oppositely charged layers is a framework for formation of polar surfaces and interfaces [22]. In this capacity EuO(1 1 1) has much to offer in terms of proximity effects: EuO(1 1 1) is predicted to induce spin polarization effects in graphene [23] and transition metal dichalcogenides [24]. However, it has been synthesized only on a metal [25]. In contrast, EuS(1 1 1), a close analogue of EuO(1 1 1), is experimentally known for pronounced proximity effects when interfaced with different materials [26,27]. One can expect that EuO(1 1 1) on Si would have quite different properties than EuO (0 0 1) on Si: it might affect differences of work functions [25] and, hence, the electronic structure of the interface, relevant to spin injection. Lattice match requirement for seamless integration of EuO(1 1 1) with silicon suggests the use of Si(1 1 1).

Here, we examine the structure EuO(1 1 1)/Si(1 1 1). This magnetic material is relevant to studies of both the polar surfaces and interfaces of EuO(1 1 1), modification of the EuO/Si contact, and silicon spintronics on the Si(1 1 1) technological platform. The polar structure of the film can pose restrictions on the attainable quality of both the EuO film and its interface with Si, motivating a thorough study of the growth conditions. Below we present a successful synthetic route to the direct epitaxial integration of EuO with Si (1 1 1) accompanied by complex structural and magnetic characterization of the material.

2. Material and methods

2.1. Synthesis

EuO/Si(1 1 1) films are grown by molecular beam epitaxy in a Riber Compact 12 system supporting ultra-high vacuum through a system of pumps (Gamma Vacuum Titan Ion pump, cryopump Cryo-Torr 8, a titanium sublimation pump) and cryopanels cooled by liquid nitrogen. The residual pressure is always less than 10^{-10} Torr. Eu of 4 N quality and SiO_x for capping are supplied from Knudsen cell effusion sources. O_2 flux (6N) is regulated by a system based on a mass flow controller and Baratron manometer. The temperatures are measured with thermocouples while the absolute temperature of Si(1 1 1) is calibrated with a PhotriX ML-AAPX/090 infrared pyrometer. All molecular beams at the substrate site are determined with a Bayard-Alpert ionization gauge. The Si(1 1 1) substrates are high-ohmic (>5 kOhm·cm) compensated wafers with miscut angle not exceeding 0.5 deg. The growth of EuO is carried out by depositing Eu ($4.4{\cdot}10^{-8}$ Torr) in a flux of oxygen ($6{\cdot}10^{-9}$ Torr). At these conditions, the growth rate is estimated to be 6.7 Å/min.

2.2. Characterization

The structural quality of the surface is controlled in situ with reflection high-energy electron diffractometer (RHEED) furnished with the kSA 400 analytical RHEED system. Cross-sections for electron microscopy studies are prepared in a Helios scanning electron microscope/focus ion beam (FIB) dual beam system. Films are covered with 2 μm of Pt. FIB milling with Ga^+ ions results in 2 μm-thick cross-sections; Ga^+ ion beams of lower energy are used for thinning and reaching electron transparency. The microscopic structure is determined with Titan 80–300 operating at 300 kV. The images are analyzed with Digital Micrograph and Tecnai Imaging and Analysis software. X-ray diffraction (XRD) θ-2θ scans are recorded with a Rigaku SmartLab 9 kW diffractometer using CuK_α X-ray source. Magnetic properties of the films are determined with MPMS XL-7 Superconducting Quantum Interference Device using the reciprocating sample option. The samples are mounted in plastic straws orienting the films with respect to the external magnetic field with accuracy better than 2°. Transport properties of the heterostructures are studied with a Lake Shore 9709A measurement system. Ohmic contacts to the films are made by depositing an Ag-Ga-Sn alloy onto terminals.

3. Choice of the synthesis route

Interfacing EuO with silicon is not a problem which can be solved by standard means. EuO on different substrates is usually grown employing the so-called adsorption-controlled or distillation mode. In this mode, a larger than stoichiometric flux of Eu atoms is used but excessive Eu atoms are re-evaporated by keeping the temperature high. It allows avoiding formation of higher Eu oxides – Eu_3O_4 and Eu_2O_3. This technique is successfully used to grow EuO on oxides like MgO [28], $YAlO_3$ [29] and YSZ [30]. However, its direct transfer to the Si substrate is complicated beyond a significant lattice mismatch of 5.6%. Although EuO is predicted to be thermodynamically stable in contact with EuO [31], Si has a tendency to form a variety of undesirable phases in reactions with oxygen and Eu: the former can produce silicon oxide while the latter can result in Eu silicides – most probably, tetragonal $EuSi_2$ [32] but formation of islands of layered trigonal $EuSi_2$ [33] is also possible taking into account the structural equivalence between Si layers in this compound and the Si(1 1 1) surface.

A solution to the problem of MBE growth of oxides on Si(0 0 1) without damaging the oxide/Si interface has been proposed by McKee et al. [34]. The main reason for formation of alien phases at the interface is that each Si atom on a clean reconstructed 2 × 1 Si(0 0 1) surface (standard reconstruction) has one unsaturated valence and, as a consequence, it is highly reactive. So, saturation of these dangling bonds with alkaline-earth metals brings the required protection to the Si surface. The standard recipe is to deposit half a monolayer (ML) of metal atoms, resulting in the formation of a surface phase 1 × 2; in this way each free valence of Si surface is accounted for. Such an approach has been used to integrate EuO with silicon via a buffer layer of SrO [35].

However, attempts of direct epitaxial integration of EuO with Si, without any buffer layer, using the 1 × 2 surface phase are unsuccessful [17,20]. The protection provided by this silicide is insufficient at high temperature required to synthesize EuO in the distillation process. Major efforts have been invested into direct epitaxial integration of EuO with Si by trying to improve the protection of the Si(0 0 1) surface. A number of methods has been proposed such as SiO_x passivation in an ML regime [16], Eu coating with a thickness of several (1 ÷ 3) MLs, hydrogen passivation of the Si(0 0 1) surface, and a combination of the latter two [18]. However, a breakthrough came from the use of another surface phase of Eu on Si – a 1 × 5 reconstruction [19,20]. It has a higher metal coverage with respect to the 1 × 2 reconstruction and provides a better surface protection. However, this is only part of the solution. EuO synthesis on Si(0 0 1) remains delicate: the substrate temperature is kept as low as possible (about 340 °C), and only slight increase of the Eu flux over stoichiometry is permitted.

Our aim is to find a recipe for successful direct epitaxy of EuO on Si(1 1 1) built upon our experience with integration of EuO with Si

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