Europium Silicide – a Prospective Material for Contacts with Silicon

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Metal-silicon junctions are crucial to the operation of semiconductor devices: aggressive scaling demands low-resistive metallic terminals to replace high-doped silicon in transistors. It suggests an efficient charge injection through a low Schottky barrier between a metal and Si. Tremendous efforts invested into engineering metal-silicon junctions reveal the major role of chemical bonding at the interface: premier contacts entail epitaxial integration of metal silicides with Si. Here we present epitaxially grown EuSi$_2$/Si junction characterized by RHEED, XRD, transmission electron microscopy, magnetization and transport measurements. Structural perfection leads to superb conductivity and a record-low Schottky barrier with $n$-Si while an antiferromagnetic phase invites spin-related applications. This development opens brand-new opportunities in electronics.
Additional requirements are imposed on the contact material if it is designed to be compatible with spintronic applications. Silicon spintronics is an emerging set of energy-efficient information technologies implementing spin functionality in Si\textsuperscript{12,22}. Injection of spin-polarized carriers into a semiconductor is demonstrated from magnetic semiconductors\textsuperscript{23}, half-metals\textsuperscript{24}, and metals through insulating tunnel barriers\textsuperscript{25-29} or Schottky-tunnel-barrier contacts\textsuperscript{30}. Ohmic contacts are not functional without spin pumping\textsuperscript{31}. Thus, SBH is a crucial parameter for metal/semiconductor spin injection\textsuperscript{32,33}. Although transient femtosecond spin current can be induced in a non-magnetic material\textsuperscript{34}, spin injection requires magnetic contacts. In general, any magnetic order suppresses spin scattering. Ferromagnetic silicides MnSi\textsuperscript{35} and Fe\textsubscript{2}Si\textsubscript{2}\textsuperscript{36} are effective spin injectors. On the other hand, antiferromagnetic (AFM) contacts are also functional in spintronics applications as they support spin currents and better suited for using as spin detectors than ferromagnets\textsuperscript{37}. AFM buffer layers may enhance spin transfer efficiency from a ferromagnet\textsuperscript{38}.

Here, we propose stoichiometric europium silicide EuSi\textsubscript{2} as a new multifunctional material for contacts with Si in nanoelectronics. We demonstrate that epitaxial EuSi\textsubscript{2}/Si junction is easy-to-manufacture and free of alien phases. The quality of the contact is confirmed by a number of techniques. In particular, transmission electron microscopy reveals the atomically sharp EuSi\textsubscript{2}/Si interface. The SBH of the EuSi\textsubscript{2}/n-Si junction (0.21 eV) is determined to be the lowest among all silicides suggesting its use in the SB-MOSFET technology. The AFM phase of EuSi\textsubscript{2} invites its applications in spintronics.

**Results and Discussion**

Europium (II) compounds are famous due to a wide range of electrical, magnetic and optical properties but the Eu-Si system and, in particular, EuSi\textsubscript{2}, are far from being well-studied. EuSi\textsubscript{2} crystallizes in the tetragonal α-ThSi\textsubscript{2} structure type (I41/amd space group) with lattice constants 4.304, 4.304 and 13.65 Å\textsuperscript{39}. Europium is highly reactive and its reaction with Si does not require high temperature: EuSi\textsubscript{2} appears as a common side product of EuO growth on Si surfaces\textsuperscript{40,41}. At the same time attempts to grow epitaxial films of EuSi\textsubscript{2} have been unsuccessful: resulting in nanoinclusions and/or polycrystalline films\textsuperscript{42}. Studies of the early stage of the silicide formation on Si (111) reveal that Si is the dominant diffusion species\textsuperscript{43} which is common for disilicides\textsuperscript{6}. The outcome of the reaction is probably governed by the Si diffusion rate which strongly depends on the substrate temperature. Therefore, in our studies of EuSi\textsubscript{2} growth on Si (001) substrate we pay special attention to fine tuning of temperature and Eu flux.

To control the state of the surface during the growth we routinely employ reflection high-energy electron diffraction (RHEED) images along the [110] azimuth of the Si substrate. The surface of the substrate is prepared by heating up to 950 °C (according to pyrometer readings) to remove the natural surface SiO\textsubscript{x} layer. The resulting surface exhibits 2 × 1 × 2 electron diffraction pattern (Fig. 1a). Then, the substrate kept at a temperature of 400 °C is exposed to a constant Eu pressure of 1.5 × 10\textsuperscript{-8} Torr coming from a Knudsen cell kept at 470 °C. It corresponds to adsorption-controlled growth (also known as MBE distillation) with an average rate 3 Å/min. It implies that the Eu flux can be varied widely without much effect on the growth outcome as long as the growth regime remains the same. RHEED images reveal a sequence of growth stages. It is common for RE silicides to self-organize and form a number of high aspect ratio nanowires when sub-monolayer amounts of metal are deposited on the surface. Also, a number of surface phases are known for RE metals on Si, including those for Eu\textsubscript{40,44}. Three of them are observed at the beginning of the growth: successive surface superstructures 1 + 2 + 2 + 1.1 × 5 + 5 × 1 and 1 + 3 + 3 × 1 are followed by formation of wide stripes on the RHEED image (Fig. 1b). Next, the stripes become thinner indicating improvement of the crystalline quality; the set of reflections gradually transforms to one that can be attributed to a single crystalline EuSi\textsubscript{2} layer (Fig. 1c). Pronounced intensity modulation along the stripes at the later stages of the growth (Fig. 1d) is a fingerprint of surface roughening. However, addition of another short high-temperature (560 °C) growth stage results in smoother surface (Fig. 1e). Films with thickness up to 560 Å have been manufactured. The lateral lattice parameter determined from the distance between streaks in the RHEED pattern is equal to 4.34 ± 0.05 Å. A capping layer of SiO\textsubscript{x} with a thickness of 200 Å is deposited on top of EuSi\textsubscript{2} to ensure its protection from the air.

Notice that our procedure is not very different from that employed in ref. 42 but the remarkable change of the outcome (single crystal instead of polycrystalline film) originates from a meticulous optimization of the growth conditions. The stability of the EuSi\textsubscript{2} structure probably plays a great role in the easy formation of the epitaxial silicide film: similar reaction of Sr with Si substrate results in polycrystalline SrSi\textsubscript{2} despite Sr being an equally active metal and very similar ionic radii of Eu(II) and Sr. Since the synthesis requires relatively mild conditions a reduced thermal budget is expected for its technological implementation. Although non-stoichiometry is often observed in rare-earth disilicides the problem is significant for hexagonal and orthorhombic structures while the tetragonal phase (like EuSi\textsubscript{2}) is characterized by a composition close to stoichiometric. As for our particular system EuSi\textsubscript{2}/Si(001), a synchrotron radiation study of EuSi2 nanoislands and polycrystalline films\textsuperscript{45} shows that the product of reaction between Eu and the Si(001) substrate has the EuSi\textsubscript{2} stoichiometry.

The epitaxial quality of the film is confirmed by X-ray diffraction (XRD) studies: Fig. 2 shows a typical 0–20 XRD scan displaying peaks (004), (008), (0012) and (0016) from EuSi\textsubscript{2}, as well as peaks from the substrate. No other phases are detected. All EuSi\textsubscript{2} peaks correspond to the same orientation with the c-axis orthogonal to the surface. EuSi\textsubscript{2} crystallites with such orientation are also observed in XRD spectra of polycrystalline EuSi\textsubscript{2}\textsuperscript{42} however accompanied by a number of other orientations. A lattice parameter of 13.633 ± 0.006 Å in the direction orthogonal to the EuSi\textsubscript{2}/Si interface is determined from the location of reflections in the 0–20 scan.

Thickness fringes are observed for the EuSi\textsubscript{2} (004) reflection (see inset in Fig. 2). This characteristic feature of x-ray diffraction is a result of the wave interference due to reflections at the interfaces, both top and bottom. Taking into account the value of the x-ray wave length (1.5418 Å), the observation of the thickness fringes is a fingerprint of sharp interfaces; otherwise the reflected waves cannot maintain the coherence and thickness fringes would not show up.
The carrier injection efficiency of the structure depends on the properties of the EuSi2/Si interface rather than on the overall quality of the film. Thus, a study of the films with transmission electron microscopy (TEM) techniques becomes indispensable. A bright field TEM image of the EuSi2 film on Si (001) is shown in Fig. 3a.
at low magnification the interface looks sharp and smooth, without any unevenness like precipitates with facets parallel to (111) Si planes. Our experiments result in a very smooth top surface of the film in strong contrast to

Figure 2. 0–2θ X-ray diffraction scan of the EuSi₂/Si junction (56 nm of the silicide). The spectrum reveals allowed peaks of EuSi₂, namely (004), (008), (0012) and (0016). Stars (*) denote peaks from the Si substrate. No extrinsic peaks are detected. Inset: thickness fringes around EuSi₂ (004) reflection for a 20 nm film.

Figure 3. Microscopic structure of the EuSi₂/Si junction. (a) Low-magnification cross-sectional bright-field TEM image of the 56 nm EuSi₂ film on Si protected by SiOₓ viewed along the [110] zone axis of the Si substrate and showing the absence of side products. (b) Selected area electron diffraction pattern of EuSi₂ superimposed with that of Si revealing their relative orientation. (c) Medium-magnification cross-sectional bright-field TEM image of the EuSi₂/Si interface showing out-of-phase boundaries in the film. (d) High-resolution cross-sectional bright-field TEM image demonstrating atomic structure of the EuSi₂/Si interface.
The average distance between dislocations is quite close to those known in the literature and those determined in our RHEED and XRD studies. The orientational magnetic ordering temperature is the largest (along with that of GdSi$_2$) among RE silicides. To compare with magnetic field up to 7 T. A robust antiferromagnetism of EuSi$_2$ constitutes its additional functional advantage: the Néel temperature – obviously not in this case. The magnetic field $H_{\text{N}}$ is close to $H_{\text{N}}(\text{ErSi}_2) = 5 K$ and $H_{\text{N}}(\text{ErSi}_2) > 5 K$.

The transport properties of the films support the results of magnetic measurements. A sharp anomaly associated with the AFM transition is observed in the vicinity of 40 K – the resistivity decreases by an order of magnitude (Fig. 5). The shift of the anomaly in a magnetic field of 9 T is $\Delta \rho = 4.3 K$. The resistivity of a magnetic metal is $\rho(0) \approx 10 K$. The estimated effective moment per Eu ion is about 9 $\mu_B$, which is close to the effective moment 7.9 $\mu_B$ associated with Eu$^{2+}$ ions (spin $S = 7/2$). It corroborates the electron energy loss spectroscopy study which determined Eu in its silicide to be divalent. The magnetic field dependence of the sample magnetization (inset of Fig. 4) is linear for $T > 5 K$ and magnetic field up to 7 T. A robust antiferromagnetism of EuSi$_2$ constitutes its additional functional advantage: AFM is accompanied by opening a spin gap which eliminates low-lying spin excitations detrimental to spin coherence. This property may enable efficient spin transport.

Figure 4. The temperature dependence of the magnetic susceptibility of the EuSi$_2$/Si junction measured in magnetic field $H = 1 T$ applied along the surface of the 56 nm film ($\chi_{||}$, red circles) and normal to the surface of the film ($\chi_{\perp}$, blue squares). Solid black lines show Curie-Weiss law approximations of $\chi_{||}$ and $\chi_{\perp}$ above the Néel temperature. Inset: the temperature dependencies of the magnetization per Eu atom for different magnetic fields normal to the surface of the EuSi$_2$ film.

The form of the specific heat in the Ising model for 3D antiferromagnets. Crystalline defects usually suppress the anomaly at the Néel temperature. Inset: the temperature dependencies of the magnetization per Eu atom for different magnetic fields normal to the surface of the EuSi$_2$ film.

The absence of any intermediate layer at the interface is found in high-resolution (HR) electron microscopy images with different magnification (Fig. 3c, d). This is quite remarkable as an amorphous interlayer is found to occur in most metal/silicon systems. Another observation is the presence of atomic steps on the Si surface with the height varying between $a_{\text{Si}}/2$ and $a_{\text{Si}}$, where $a_{\text{Si}}$ is the Si unit cell constant. The projection of the Burgers vector on (110)Si is $a_{\text{Si}}/2[110]$. The average distance between dislocations is 20 Å – the value expected from the lattice mismatch between EuSi$_2$ and Si. Further inspection of HR images reveals out-of-phase boundaries (OPBs) in the EuSi$_2$ film as well as regions of 50–200 Å size with tiny misorientations. The formation of OPBs is associated with the steps at the interface: the shift between the adjacent regions is close to $c/4$. The density of OPBs diminishes from the EuSi$_2$/Si interface to the EuSi$_2$ surface. The misorientations come from misfit dislocations and related strains. The film homogeneity is established by TEM studies of polycrystalline EuSi$_2$ films of previous attempts. The selected area (electron) diffraction pattern (SADP), shown in Fig. 3b, certifies that EuSi$_2$ adopts a tetragonal crystal lattice with lattice parameters $a = 4.3 \text{ Å}$ and $c = 13.6 \text{ Å}$, quite close to those known in the literature and those determined in our RHEED and XRD studies. The orientation relationships derived from the SADP are:

$$\left[001\right]_{\text{Si}} \parallel \left[001\right]_{\text{EuSi}_2} ; \left(110\right)_{\text{Si}} \parallel \left(110\right)_{\text{EuSi}_2} ;$$

corroborating the RHEED and XRD data. It means that the lattice mismatch between EuSi$_2$ and Si is large, approximately 12%.

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The resistivity of EuSi$_2$ changes from 37 $\mu$Ohm·cm at room temperature to 2.1 $\mu$Ohm·cm at 2 K. The residual resistivity is significantly smaller than that observed for ErSi$_{2.50}$ or ErSi$_{1.751}$ thin films. Hall effect measurements of our films determine the electron concentration about $10^{22}$ cm$^{-3}$. The Hall mobility increases from 5 cm$^2$/V·s at room temperature to 500 cm$^2$/V·s at 1.5 K. These properties make EuSi$_2$ a very attractive metallic junction material.

In Si-based nanoelectronics the resistivity of a silicide is coupled with another major characteristic, consumption of Si in its reaction with a metal. Low silicon consumption is a most important technological requirement constraining applications of metal silicides to ultra-shallow junctions and silicon-on-insulator films. Low sheet resistance requires the silicide thickness to be maximized but correspondingly increased Si consumption leads to local junction penetration. It is a major factor that hinders applications of CoSi$_2$ and elevates NiSi among other mid-gap silicides. Si consumption is characterized by the ratio of the resulting silicide thickness to the thickness of consumed Si, required to be as large as possible. Typical values are close to 1: 1.10 for TiSi$_2$, 0.97 for CoSi$_2$, 1.20 for NiSi and 1.27 for stoichiometric ErSi$_2$. The same parameter calculated for EuSi$_2$ is very large (1.58) making this material highly attractive for nanoscale applications. However, EuSi$_2$ is not designed to compete with transition-metal low-resistivity silicides; instead, it is suggested as a prospective material for the SB-MOSFET technology (see below).

Injection of carriers into Si from a metal contact is governed by the SBH. The Si substrate in our experiments exhibits resistivity 3.8 kOhm·cm, electron concentration $1.5 \times 10^{12}$ cm$^{-3}$ and Hall mobility 1100 cm$^2$/V·s$^{-1}$. The current-voltage characteristic of the SBH for the grown EuSi$_2$/n-Si contact is distinctly asymmetric in the temperature region between 160 K and 300 K (Fig. 6). With a small forward bias the I–V curve is exponential. Defects at the metal/Si interface effectively increase the SBH and hinder the injection$^4$. The I–V curve for the EuSi$_2$/n-Si contact follows the ideal (not influenced by surface states) classical thermionic theory$^{52}$, yet another indication of the quality of the interface.
\[ I = I_s \cdot \exp\left(\frac{qV}{kT}\right) - 1, \]

where \( I_s \) is the saturation current, \( T \) – the absolute temperature, \( q \) – the electron charge, \( A \) is the Richardson constant, \( S \) is the contact area and \( \Phi_b \) is the SBH. According to our data the SBH of the EuSi\(_2\)/n-Si junction is 0.21 ± 0.01 eV, significantly smaller than the values known for other RE silicides\(^{16}\). The record-low Schottky barrier height constitutes a major advantage of EuSi\(_2\) over other silicides in competition for employment as contact material in electronics.

In summary, taking into account technological advantages of metal silicides and their full compatibility with Si technology we propose europium disilicide as a prospective junction material. In the course of our work we optimized conditions for manufacturing EuSi\(_2\)/Si contacts. Epitaxial films are grown by reaction of Eu with silicon substrate. The synthesis is robust, easy to implement and what is most important is free from unwanted side products. Moreover, electron microscopy shows that the EuSi\(_2\)/Si interface is atomically abrupt despite a significant lattice mismatch.

Apart from the superb structural quality of the EuSi\(_2\)/Si interface and EuSi\(_2\) film, europium silicide exhibits a combination of properties which respond to demands of modern electronics: At low temperature EuSi\(_2\) becomes antiferromagnetic which may assist applications employing spin-related phenomena. Rather low resistivity and very low Si consumption are among other advantages of the material. Most importantly, the EuSi\(_2\)/n-Si junction exhibits the lowest among silicides Schottky barrier height. Overall, EuSi\(_2\) is the most promising material for the SB-MOSFET technology.

**Methods**

**Synthesis.** The samples are grown in Riber Compact 12 system for molecular beam epitaxy furnished with UHV system comprising Gamma Vacuum Titan Ion Pump, cryopump Cryo-Torr 8 (Brooks CTI Cryogenics), titanium sublimation pump and cryopanels cooled down by liquid nitrogen. The base pressure is less than 10\(^{-10}\) Torr. 4N Eu and capping material SiO are supplied from Knudsen cell effusion sources. The temperature of the substrate is controlled with PhotriX ML-AAPIX/090 infrared pyrometer (LumaSense Technologies) operating at the 0.9\(\mu\)m wavelength. Molecular beam intensity is measured with Bayard-Alpert ionization gauge fitted at the substrate site. The substrates are high-ohmic compensated Si (001) wafers with miscut angles not exceeding 0.5\(^\circ\).

**Transmission Electron Microscopy.** The samples are prepared for analytical TEM/STEM are prepared with 2 different techniques. One is a standard procedure comprising mechanical polishing of cross-sections down to a thickness of 20–25\(\mu\)m followed by ion milling with Ar\(^+\) using Gatan 691 PIPS at an accelerating voltage of 3 keV until perforation; the final milling is carried out with 0.1 keV Ar\(^+\) ions. The other procedure employs Helios (FEI) scanning electron microscope (SEM)/Focus Ion Beam (FIB) dual beam system equipped with gas injectors for C and Pt deposition and a micromanipulator (Omniprobe). A 2\(\mu\)m Pt layer is deposited on the surface of the sample. FIB milling (30 keV Ga\(^+\) ions) results in 2\(\mu\)m thick cross-sections of approximately 8 × 5\(\mu\)m\(^2\) area. Electron transparency is achieved by further thinning and final cleaning with 5 keV and 2 keV Ga\(^+\) ion beams, respectively. The cross-sections are covered by thin C layers to prevent oxidation in the Helios chamber before breaking the vacuum. The specimens are studied with a TEM/STEM Titan 80–300 (FEI) operating at 300kV. The microscope is equipped with a spherical aberration (C\(_s\)) corrector, a HAADF detector, an atmospheric pressure ionization (API) source, and an acceleration energy filter (GIF). The images are acquired using the Digital Micrograph (Gatan) and Tecnai Imaging and Analysis (FEI) software.

**Characterization.** The surface of the films is controlled in situ with reflection high-energy electron diffraction fitted with kSA 400 Analytical RHEED System (k-Space Associates, Inc.). X-ray diffraction data are obtained with Bruker D8 Advance spectrometer (CuK\(_\alpha\), X-ray source). Magnetization measurements of the films are carried out with MPMS XL-7 SQUID magnetometer (Quantum Design) using reciprocating sample option (RSO). The samples are mounted in plastic straws orienting the surface of the films parallel or perpendicular to the external magnetic field with the accuracy better than 5\(^\circ\). The diamagnetic moment of the Si substrate exceeds the magnetic moment of thin EuSi\(_2\) films; its subtraction from the signal generates a systematic error of about 10% in the value of magnetization. The demagnetization field is not taken into account. Transport measurements of resistivity and Hall effect in EuSi\(_2\) and I–V characteristics of the EuSi\(_2\)/Si junction are carried out by the four-terminal sensing method using Lake Shore 9709A Hall effect measurement system.

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**Acknowledgements**

The work is partially supported by NRC “Kurchatov Institute”, Russian Foundation for Basic Research through grants 16-07-00204 and 16-29-03027, and Russian Science Foundation through grant 14-19-00662.
Author Contributions
D.V.A. and V.G.S. synthesized the EuSi₂ films. C.G.K., I.A.K. and A.L.V. carried out T.E.M. experiments. G.V.P. performed X-ray studies. A.N.T. carried out magnetization measurements. O.E.P. performed transport experiments. A.M.T., E.F.L. and V.G.S. carried out the analysis. A.M.T. and V.G.S. wrote the paper with contributions from D.V.A., A.L.V., A.N.T. and O.E.P. All authors reviewed the manuscript.

Additional Information
Competing financial interests: The authors declare no competing financial interests.

How to cite this article: Averyanov, D. V. et al. Europium Silicide – a Prospective Material for Contacts with Silicon. Sci. Rep. 6, 25980; doi: 10.1038/srep25980 (2016).

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