Supporting Information

2D ferromagnetism in europium/graphene bilayer

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Experimental methods

a. Synthesis

The samples of Eu/graphene bilayer are synthesized in the Riber Compact 12 system for molecular beam epitaxy. The synthesis is carried out in ultrahigh vacuum – base pressure in the growth chamber does not exceed 10^{-10} Torr – maintained by a system of an ion pump, a sublimation pump, a cryopump, and cryopanels cooled by liquid N_2. The substrates are large-area graphene sheets wet transferred to SiO_2/Si(001) by Graphenea – more than 98% of the substrate surface is covered with graphene; the grain size is up to 20 μm; the graphene conductivity is of *p*-type with a carrier mobility ~10^3 cm^2 V^{-1} s^{-1}. 4N Eu, 4N Al, and 4N SiO_x are supplied from Knudsen cell effusion sources. The intensity of molecular beams is controlled with a Bayard-Alpert ionization gauge fitted at the substrate site. The substrate temperature is measured with the PhotriX ML-AAPX/090 infrared pyrometer operating at a wavelength 0.9 μm.

The first step of synthesis is vacuum-assisted thermal annealing of the substrate at 600 °C for 10 min. Then, the Eu effusion cell is heated to 535 °C which corresponds to a Eu flow of $4.5 \cdot 10^{-8}$ Torr. During the synthesis, graphene/SiO_2/Si(001) is kept at 20 °C, to prevent Eu intercalation. The optimized time of Eu deposition onto graphene is about 20 s. To protect the samples from air, two options are used for capping at room temperature: (i) polycrystalline Al layer or (ii) amorphous SiO_x layer. In both cases, the capping layer is grown exceedingly thick (around 200 nm).

b. Structural characterization

The surface of the sample – graphene or Eu/graphene bilayer – is controlled *in situ* with a reflection high-energy (15 keV) electron diffractometer furnished with the kSA 400 Analytical RHEED system. The microstructure of the films is studied *ex situ* with high-resolution electron microscopy. The cross-sectional specimens are prepared in the Helios NanoLab 600i scanning electron microscope/focused ion beam (FIB) dual beam system. First, a 2 μm Pt layer is deposited on top of the capping layer; then, a cross-section 2 μm × 5 μm × 5 μm is cut employing FIB milling with 30 keV Ga^+ ions; it is further thinned and cleaned till electron transparency with 5 keV and 2 keV Ga^+ ions, respectively. The cross-sections are studied with the 300 kV TEM/STEM Titan 80-300 microscope equipped with bright-field and high-angle annular dark-field detectors. The images are processed with the Digital Micrograph and Tecnai Imaging and Analysis software.

c. Magnetism and electron transport

The magnetic properties of the samples are determined with the MPMS XL-7 Superconducting Quantum Interference Device. The samples are mounted in plastic straws; accuracy of the sample orientation with respect to the applied magnetic field is better than 2°. The measurements are carried out employing the reciprocating sample option. The ferromagnetic moments of the Eu/graphene bilayer are determined in two ways: (i) subtraction of the substrate background (measured separately); (ii) subtraction of contributions linear in magnetic field. The latter approach is a two-step procedure – determination of the magnetic susceptibility χ by measuring the magnetic moment χH^* at a high field H^*; subtraction according to $M_{FM}(H) = M(H) - \chi H$. Both approaches produce similar results proving the correctness and self-consistency of the subtraction.

The Hall effect and magnetoresistance in Eu/graphene bilayer are studied with the Lake Shore 9709A Hall effect measurement system. The four-terminal sensing measurements are carried out employing square samples with a lateral dimension about 5 mm. An image of the device employed and schemes for R_{xx} and R_{xy} measurements are presented in Fig. S2. Fabrication of electrical contacts to the films requires special care because contact of Eu/graphene bilayer with air should be avoided. We deposit metal on each terminal *ex situ*, after the film is capped with SiO_x. To this purpose, we employ an Ag-Sn-Ga alloy which melts around room temperature and exhibits very good adhesion properties. The capping is removed mechanically under the melted alloy which prevents contact of the material with air. The quality of the contacts is tested by measuring I-V characteristic curves.

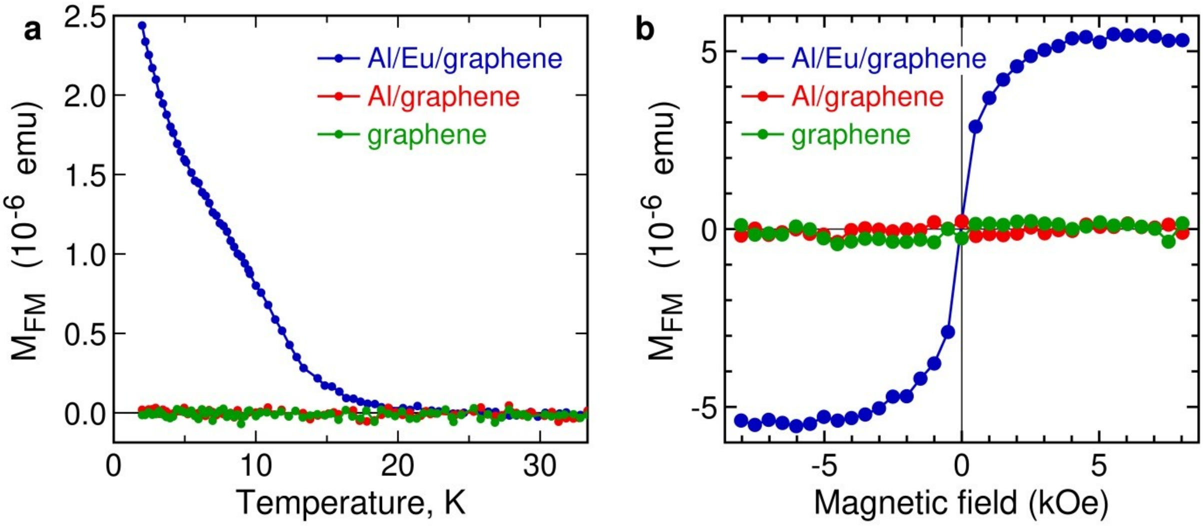

Fig. S1. (a) Temperature dependence of the ferromagnetic moment in Al/Eu/graphene (blue) as compared with Al/graphene (red) and graphene (green), all on SiO_2/Si; the measurements are carried out in an in-plane magnetic field 200 Oe. (b) In-plane magnetic field dependence of the magnetic moment in Al/Eu/graphene (blue), Al/graphene (red) and graphene (green) on SiO_2/Si at 2 K.

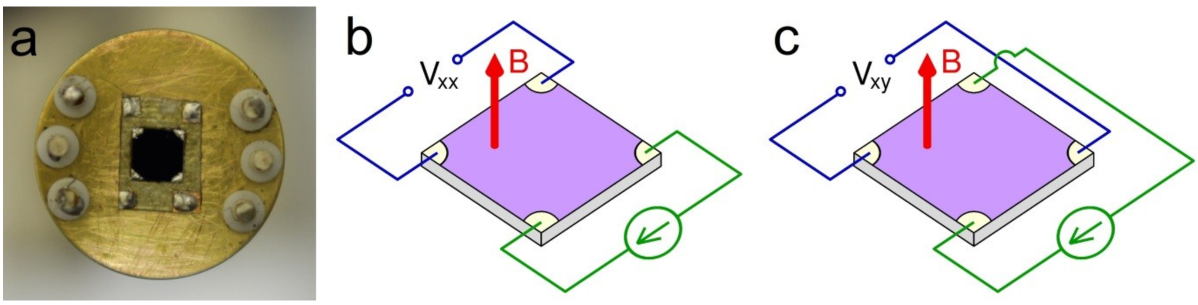

Fig. S2. (a) Image of the device implementing van der Pauw measurements of R_{xx} and R_{xy}. (b) and (c) Electrical schemes for van der Pauw measurements of the longitudinal magnetoresistance $R_{xx}(B)$ (b) and the Hall magnetoresistance $R_{xy}(B)$ (c) in an out-of-plane magnetic field (the values are produced by averaging over a set of contact permutations).