Supporting Information - Chemically Engineered Graphene-Based 2D Organic Molecular Magnet

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Table S1. Exchange coupling field extracted from the MOKE experiments, a signature of antiferromagnetic coupling (H_{bias}).

Region	H_{bias} (Oe)
1	3305.129
2	3121.299
3	3191.634
4	3285.947
5	3621.637
6	3745.523
7	11,245.8
8	12,445.5
9	6553.333
10	2344.415

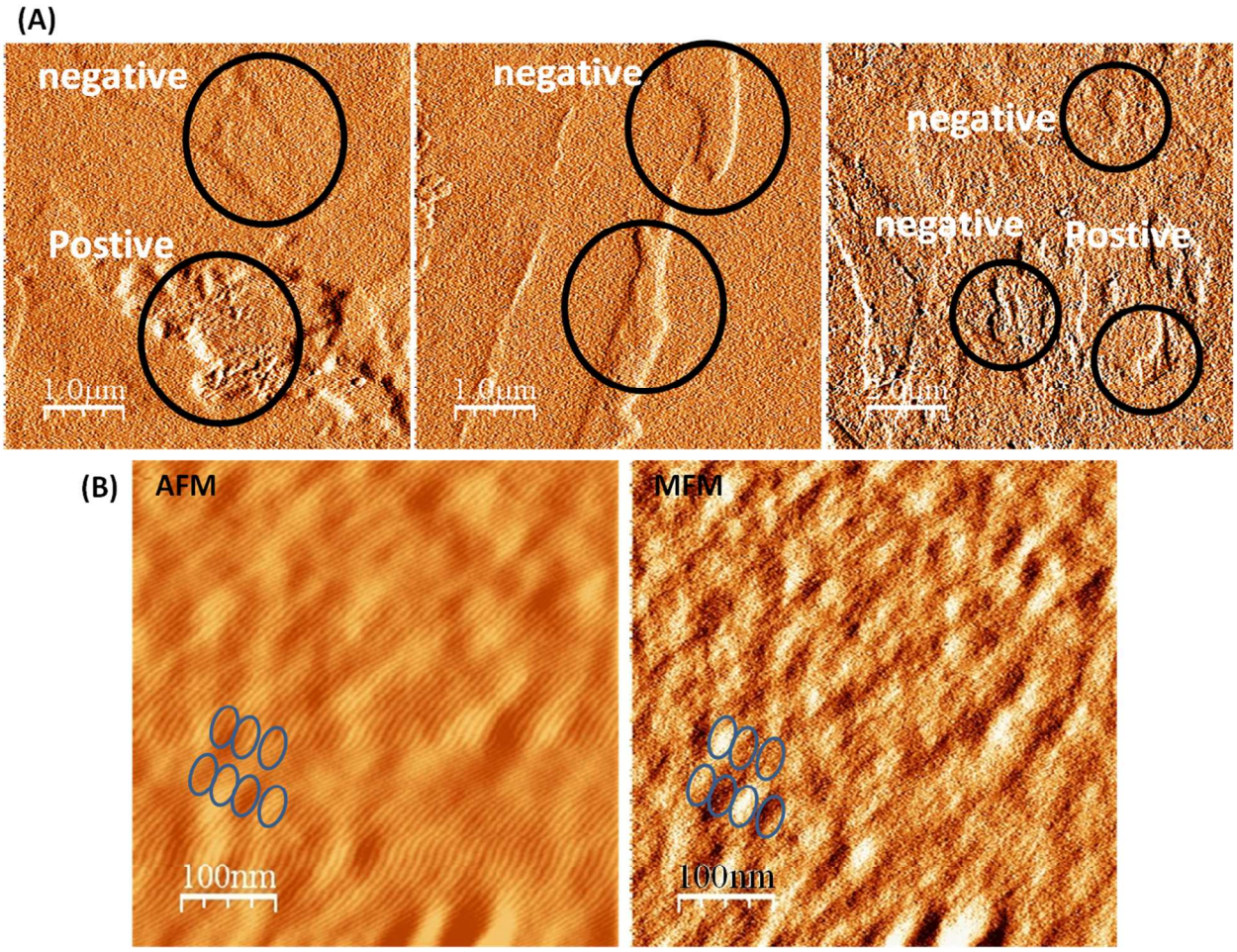

Figure S1. Macroscale MFM images of NP-EG structures. (**A**) The domain structures of magnetization pointing up and down in high-density functionalized graphene. (**B**) The mixed topology and magnetic phase of smaller domain structures of magnetization pointing up and down next to each other in functionalized graphene on a smaller scale.

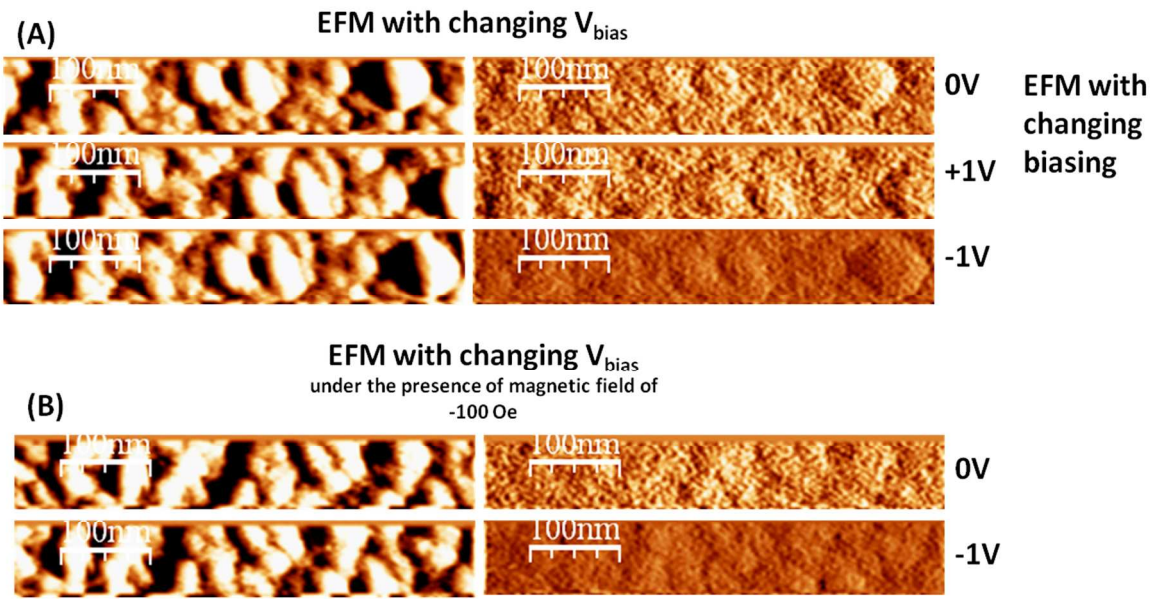

Figure S2. EFM images of functionalized graphene structures obtained by changing the voltage bias (**A**) and the magnetic field (**B**) in the same region. This behavior allows for the possibility of controlling the magnetic phase through external parameters such as the voltage bias.

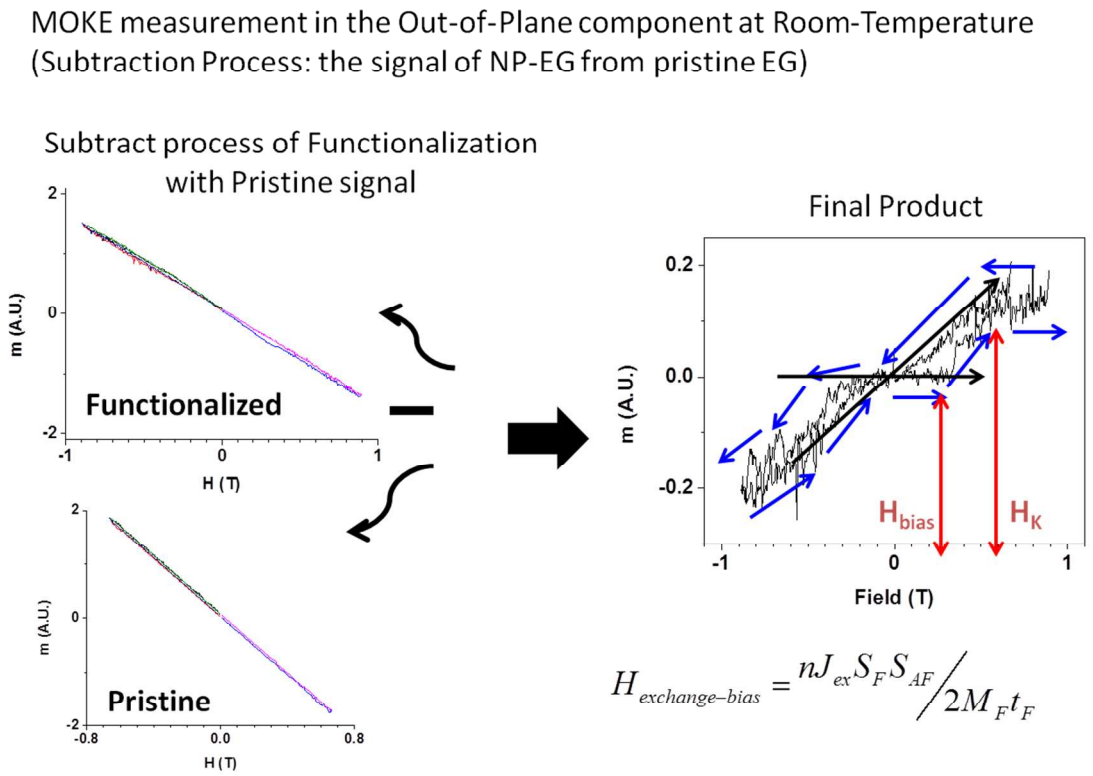

Figure S3. Subtraction of MOKE data before and after functionalization at room temperature.

Illustration of m-H loops from MOKE data such as exchange-bias and anisotropy fields.

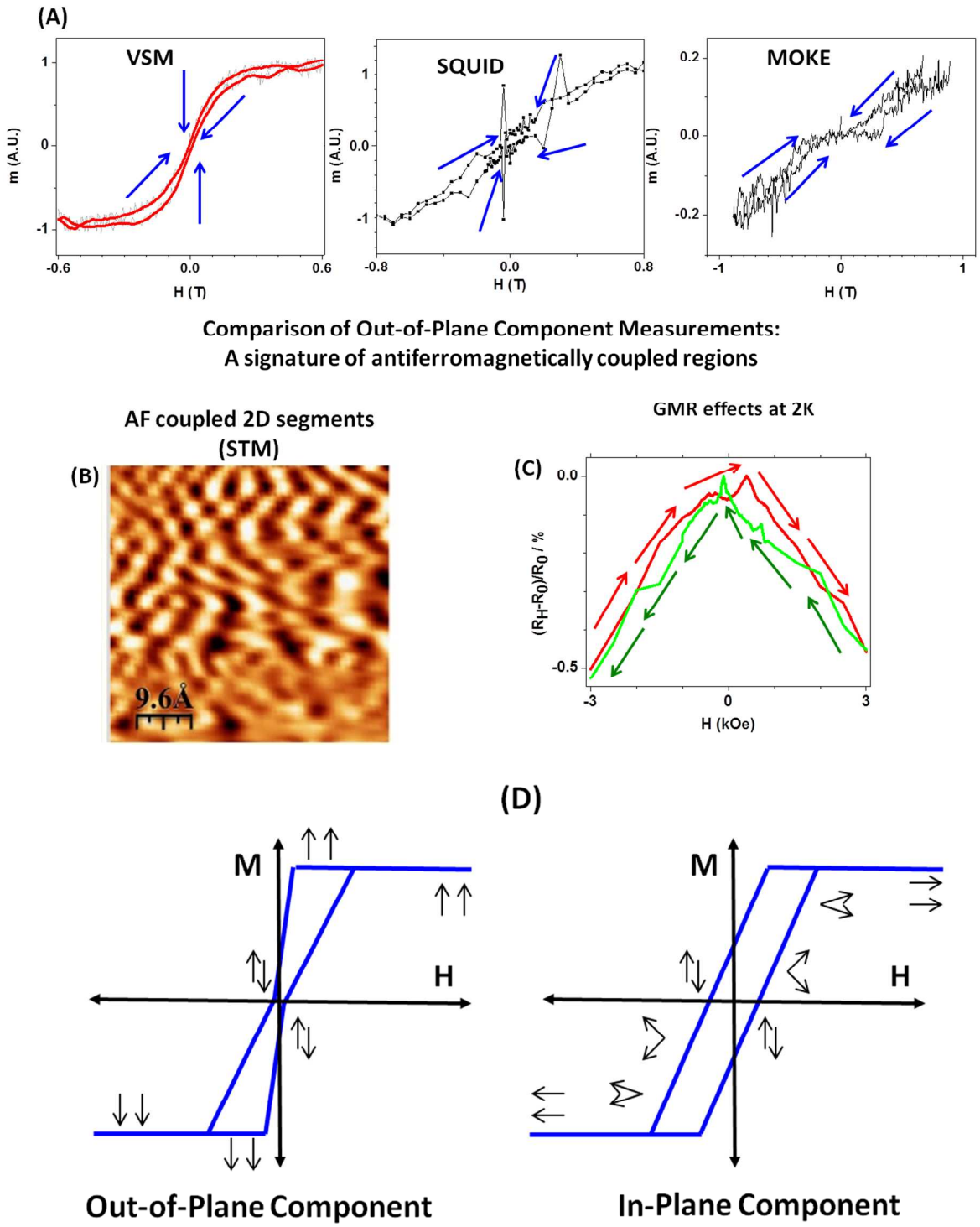

Figure S4. (A) Comparison of out-of-plane measurement data from three different types of measurements: VSM, SQUID, and MOKE. (B) STM images of antiferromagnetically coupled segments. (C) GMR effects of functionalized graphene at 2 K in the out-of-plane component.

The graphene presenting the zero field resistance is significantly different from the parallel case indicating the presence of a GMR effect. (D) Possible scenario of an experimental model of m-H loops in the in-plane and out-of-plane components.

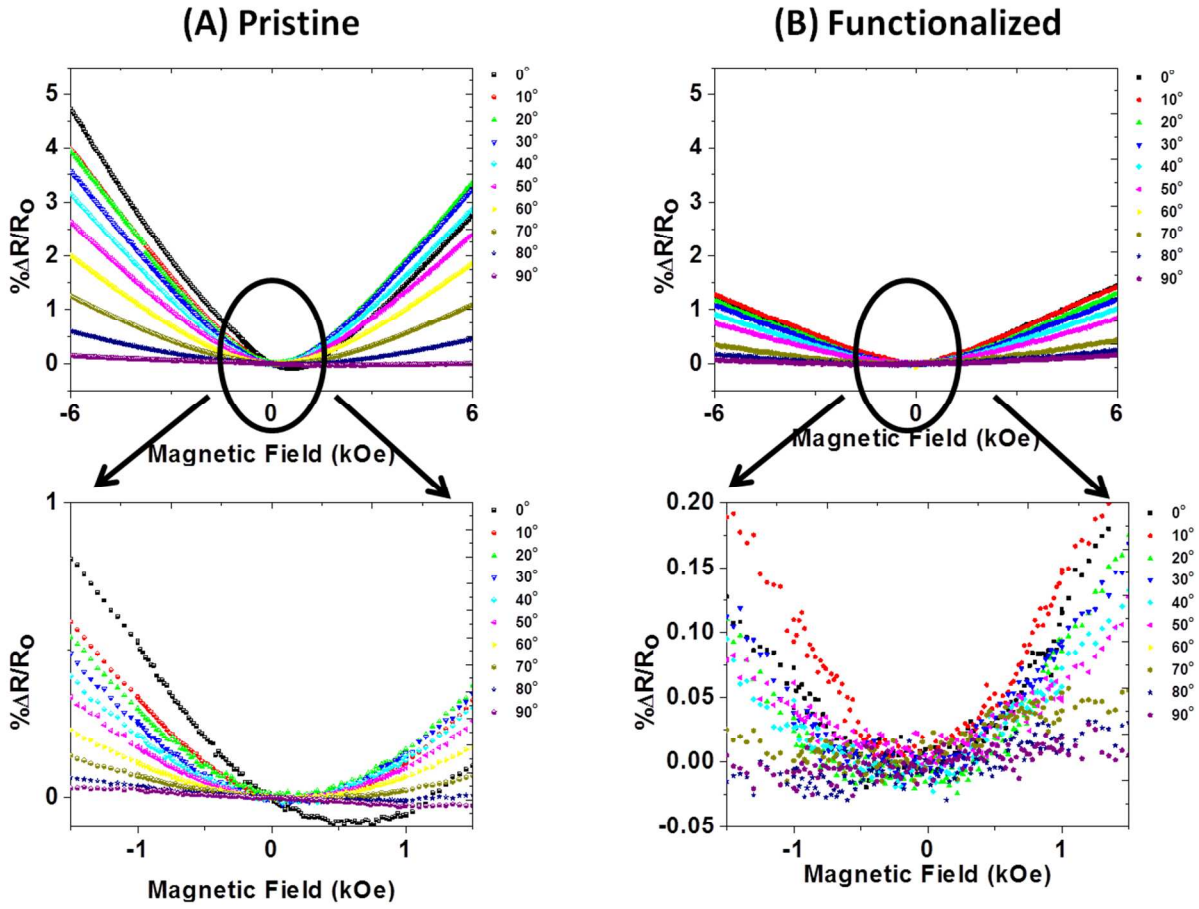

Figure S5. Rotation measurements of positive-MR at angles from out-of-plane to in-plane. The fit to the MR is slightly asymmetry before functionalization and symmetric after functionalization. This is a signature of non-uniformity, such as the rotational stacking default based on a change in scattering mechanism. This type of transition from asymmetry to symmetry was observed *via* EFM and Raman spectroscopy. Proper control of the positive MR in

functionalized graphene could lead the control of future magnetoelectric devices through external forces such as an electric field.

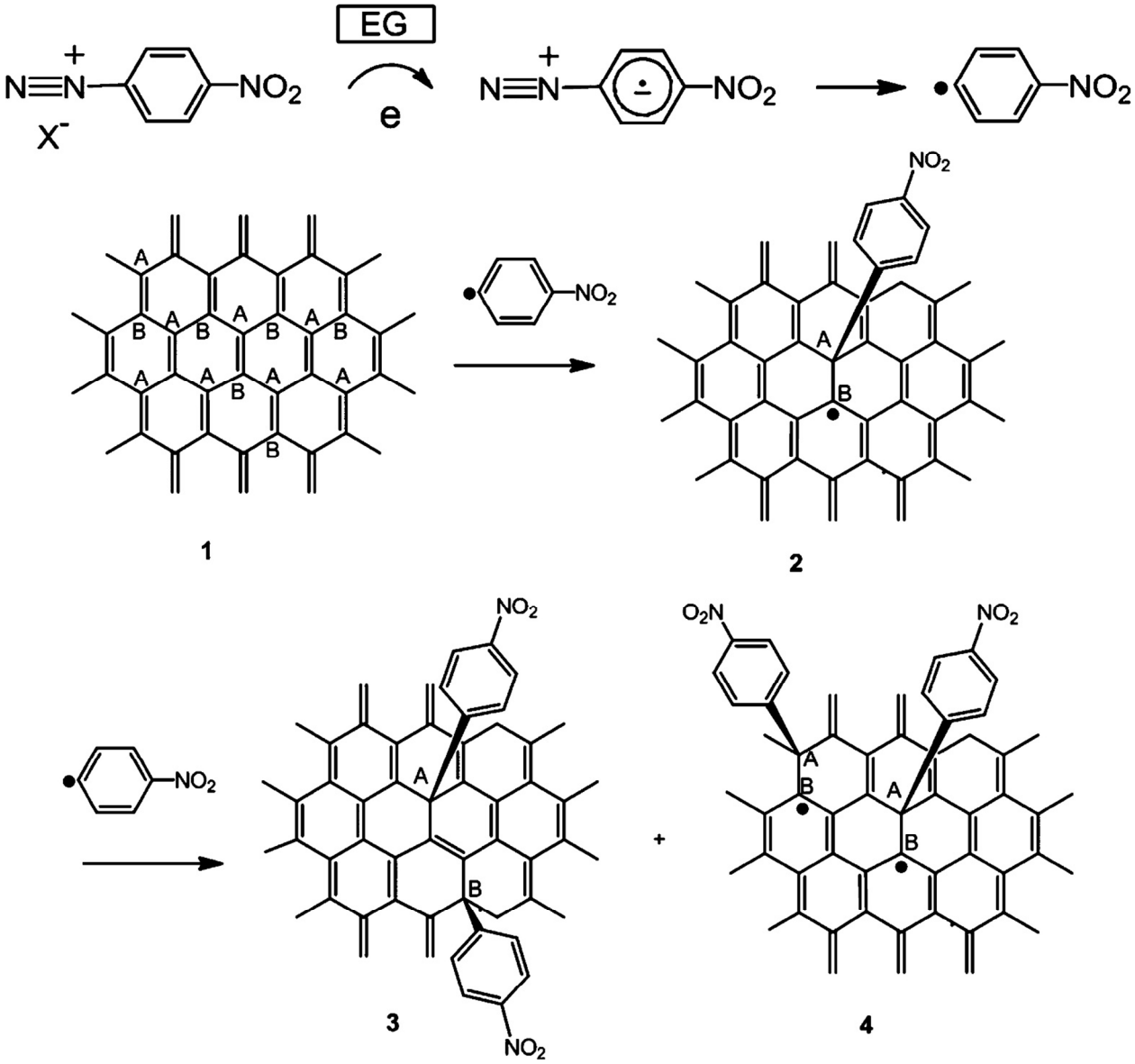

Figure S6. Schematic diagrams of the chemically functionalized nanoribbons: structures of the NP-functionalized epitaxial graphene products formed by spontaneous electron transfer from EG to p-nitrobenzene diazonium salt. The structure of the products 3 is expected to be antiferromagnetic whereas structure 4 would yield a ferromagnetic interaction between the spins.

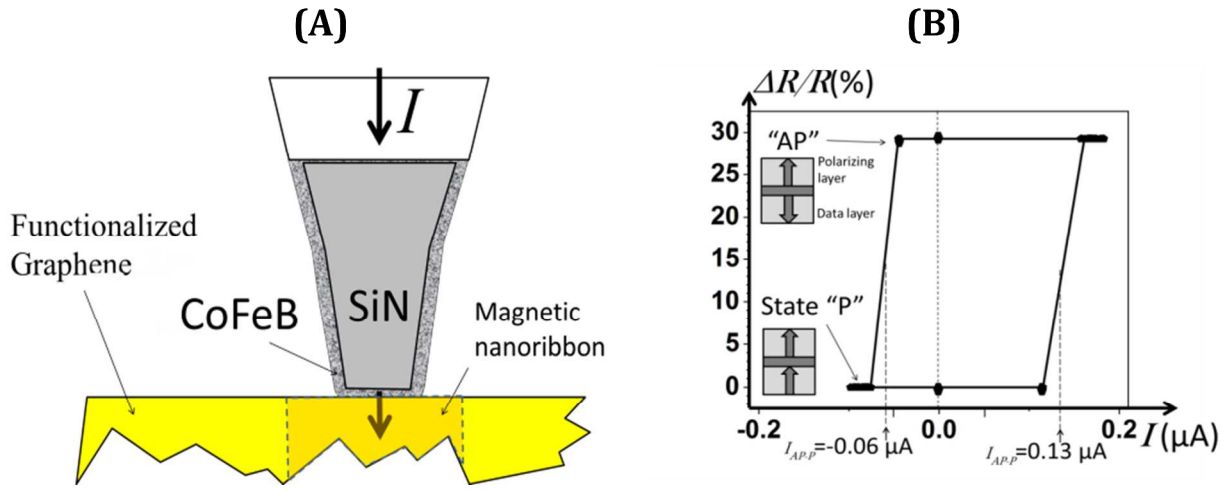

Figure S7. (A) A possible schematic illustration of a 3D nano-MTJ between the metal tip and a chemically isolated nanoribbon to enable STT-based energy-efficient information switching for further experiment. (B) I-R plot for Ta/CoFeB/MgO/CoFeB/Ta structures with a copper substrate. These results would be useful in applications for CINs with the advantages of a long spin relaxation time and extremely low energy under the control of an external electric field.