ESRF	**Experiment title:** Nanomagnetism of graphene on a metal	**Experiment number**: HE-3397
Beamline:	**Date of experiment**: from: 17.01.2011 to: 01.02.2011	**Date of report**: 31.08.2011
Shifts: 18	**Local contact(s)**: Violetta Sessi	*Received at ESRF:*

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Report:

Using X-ray magnetic circular dichroism (XMCD), we explored the magnetism of Co and Fe clusters on graphene/Ir(111), which we relate to the cluster size and distribution by scanning tunneling microscopy (STM). We additionally conducted a preliminary investigation of the ferromagnetic/superparamagnetic transition in a Eu layer intercalated between graphene and Ir(111).

The experiment was precedented by a thorough preparation time which allowed fine preparation of the ultra-high vacuum (UHV) chambers and adaptation of the heating system to high temperature flashes of the samples, with the help of electron-beam heating. Ir(111) single crystals were prepared by repeated cycles of 1500 K flashes an Ar^+ sputtering. Graphene was prepared by chemical vapour deposition of ethylene let in the UHV chambers *via* a leak valve. Co, Fe, Ir and Pt were evaporated at room temperature by electron-beam evaporation with the help of evaporators available at ID08 and a multi-pocket evaporator provided by the Köln applicants. Eu was evaporated thanks to a Knudsen cell provided by the Köln applicants. The preparation time allowed for reliable calibration of the evaporators flux, sample temperature, with the help of *in situ* post-growth STM analysis.

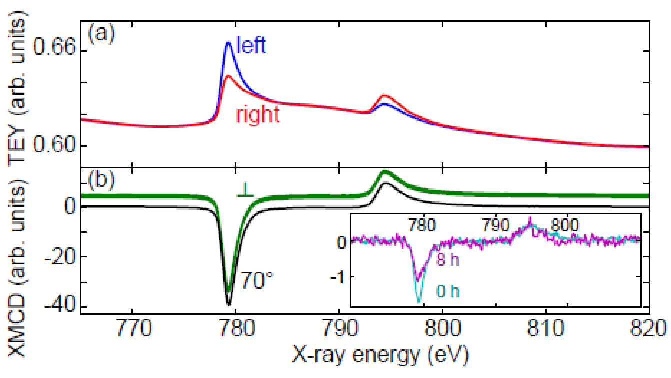

Figure 1: (a) TEY for $Pt_{13}Co_{26}$ clusters across the $L_{2,3}$ Co absorption edges, for left and right-circularly polarized X-rays in perpendicular (⊥) incidence, at 5 T and 10 K. (b) XMCD signals for ⊥ and 70° incidence (vertically shifted for clarity). Inset: XMCD signal normalized to absorption for Ir_4Co_8 clusters, at the beginning and after heigh hours of irradiation.

We systematically addressed the magnetism of Co and Fe nanoclusters self-organized or not on the 2.5 nm pitch graphene/Ir(111) moiré template. Direct evaporation of Co and Fe on graphene/Ir(111) at room temperature yields disordered cluster assemblies, while seeding of the clusters with small Pt or Ir clusters anchored to the moiré sites drives the ordered growth of PtCo, PtFe, IrCo or IrFe clusters. Eleven samples, some involving Pt and Ir seeding and some not, were studied with XMCD. We made three main findings (*Vo-Van, Schumacher, Coraux, Sessi, Fruchart, Brookes, Ohresser, Michely, accepted for publication in Appl. Phys. Lett., arXiv:1107.2571*):

– the magnetic nanoclusters have extremely weak magnetic anisotropy (Figs. 1&2), although orbital moments are slightly enhanced as compared to bulk values,
– the temperature-dependent magnetization-magnetic field (M-H) loops (Fig. 2) reveal spin blocks which size largely exceeds that of an individual cluster, pointing to inter-cluster magnetic interactions,
– facile degradation of the clusters take place upon exposure to the soft X-ray beam, resulting in a loss of magnetic moment (Fig. 1 inset).

The XMCD signal was recorded in total electron yield (TEY) under several X-ray beam incidences in order to derive the spin and orbital moment contributions with the help of sum rules. Subtracting the TEY at the L_3 edge and pre-edge value, and normalizing to the pre-edge value, yielded M-H loops, which were measured at two incidences and various temperatures. Smaller clusters (*e.g.* ~ 25 Co atoms) are found superparamagnetic at the lowest accessible temperature (~10 K).

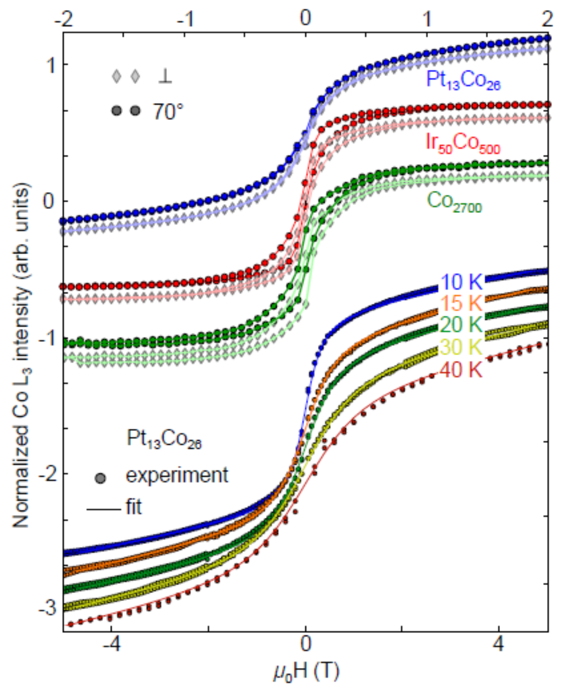

Figure 2: Normalized M-H loops (upper horizontal axis) at 10 K for different samples, for ⊥ and 70° incidence. Temperature-dependent M-H loops (lower horizontal axis) and Langevin fits for $Pt_{13}Co_{26}$ at ⊥ incidence. Curves are vertically shifted.

A last sample was devoted to a preliminary investigation of the magnetism in Eu-intercalated graphene/Ir(111). Following room-temperature evaporation of about 70 % of an Eu monolayer, mild annealing lead to intercalation of a (√3×√3) Eu layer and re-evaporation of the excess Eu. The XMCD signal and M-H loops acquired at the Eu M-edge reveal strong magnetic anisotropy, with an in-plane easy axis (Fig. 3). The Eu layer is ferromagnetic at 10 K and has barely detectable coercivity around 40 K, pointing to a blocking temperature between 10 and 50 K (further experiments are needed for finer assessment). These observations compare well with recent spin-polarized density functional theory calculations pointing to a magnetic phase transition at a few 10 K, and call for more comprehensive and systematic measurements.

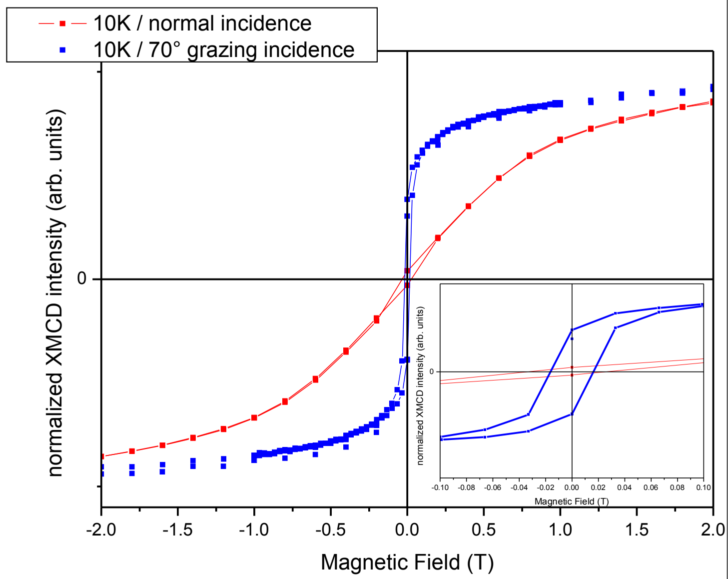

Figure 3: Normalized M-H loops for a (√3×√3) Eu layer intercalated between graphene and Ir(111), for normal and 70° incidences, at 10 K, and magnification of the low-field region (inset).