

<b>ESRF</b>	<b>Experiment title:</b> Magnetism of Eu/graphene compounds	Experiment number: HE-3741				
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## **Report:**

During our beamtime, we obtained good results on Eu monolayers intercalated under graphene/Ir(111). Two different structures form, depending on the deposited amount of Eu: one which has a  $(2\times2)$  superstructure with respect to graphene, and one which has a  $(\sqrt{3}\times\sqrt{3})R30^\circ$  superstructure with respect to graphene. We investigated both systems by XMCD in this beamtime. A publication has resulted from this beamtime [Phys. Rev. B 90, 235437 (2014)], the published results of the beamtime are contained in section V to VIII of that paper.

Briefly, we measured XMCD at the Eu M5,4 edges at the highest attainable fields of 5T and lowest attainable temperature of 10K. Our measurements revealed Eu to be in a 4f^7 configuration, with orbital momentum m\_L = 0 and spin momentum m\_S = 7/2, as concluded by use of sum rules. We have furthermore obtained field-dependent magnetization loops, i.e. M(H), for both structures. In the (2×2) superstructure, the Eu moments displayed plain paramagnetic behavior, while in the  $(\sqrt{3} \times \sqrt{3})R30^\circ$  superstructure we found superparamagnetic behavior, i.e., a large susceptibility, but no spontaneous magnetization. The  $(\sqrt{3} \times \sqrt{3})R30^\circ$  superstructure exhibited a strong anisotropy, which could be explained by the shape anisotropy alone, as the layer is atomically thin and Eu has a large magnetic moment (7 µB), which enters quadratically into the shape anisotropy energy. A magnetocrystalline contribution was concluded to be negligible due to the spherically symmetric Eu 4f^7 shell. Figure 1 and 2 and Table I summarize our results on the intercalated Eu structures.

From the data on the  $(\sqrt{3} \times \sqrt{3})R30^\circ$  structure, we concluded that it is close to ferromagnetic ordering. We reckoned that, possibly, as according to the Mermin-Wagner theorem, ferromagnetic order cannot occur in a truly two-dimensional systems without sufficient anisotropy, the spontaneous magnetization and hysteresis

may be suppressed in our system down to lower temperatures. In retrospect, more beamtime should have been spent to obtain a temperature-dependence of the magnetization loops, to clarify the situation.

Less successfully, we also spent significant effort to measure Eu monolayers on top of graphene, and "sandwich" structures, where a graphene monolayer is sandwiched between two Eu monolayers, in order to investigate the magnetic coupling through graphene. However, these structures were found to be highly sensistive to oxidation during the transfer to the magnet, as there is no protection from oxidation by the inert graphene cover, as in the intercalated structures. The samples oxidized, despite the fact that the transfer from the preparation chamber to the magnet all happened in situ and in nominal UHV pressures. The strong ferromagnetic EuO contamination led us to discard these measurements. From this beamtime, we therefore also conclude that to investigate monolayers of non-noble metals, it is absolutely necessary to perform the metal deposition onto the sample while the sample is inside the cryostat, where a low 10^-10mbar pressure is maintained by the cryopumping effect at all times.



Figure 1: (a) Normalized XAS signal at 10 K and 5T for positive ( $\mu_+$ , solid green line) and negative ( $\mu_-$ , dashed red line) helicities in normal incidence. (b) Resulting XMCD signal ( $\mu_+-\mu_-$ ). (c) Polarization-averaged XAS spectrum  $1/2^*(\mu_+ + \mu_-)$  (blue solid line) with a steplike continuum background (green dashed line).

	$ms(\mu B)$		$\chi[\mu_{\rm B}/({\rm T}\cdot{\rm atom})]$	
Sample	0°°	60°	$0^{\circ}$	60°
(2×2)	4.9	5.3	1.2	1.5
$(\sqrt{3}\times\sqrt{3})R30^{\circ}$	6.3	6.8	6.2	24

Table 1 Spin moment m\_S per Eu atom derived from the sum rules (for n\_h=7), and zero-field susceptibilities for intercalated layers of different densities. The data were taken at 10 K under a field of 5 T. X rays and magnetic field were both incident normally (0°) or grazing (60°). We estimate the relative errors to be 10%.



Figure 2: Magnetization loops for a fully intercalated  $(3\sqrt{\times}3\sqrt{)}R30^\circ$  Eu layer (squares) and a (2×2) Eu layer (circles) at 10 K for normal (half solid) and grazing (solid) incidence. Each curve is scaled to the corresponding effective spin moment per Eu atom at 5 T.