



	Experiment title: Elemental magnetic moments and magnetic order in the Fe₅GeTe₂ van der Waals magnet doped with cobalt	Experiment number: HC 5028
Beamline: ID32	Date of experiment: from: 20/09/2022 to: 25/09/2022	Date of report: 6/9/2023
Shifts: 18	Local contact(s): Roberto SANT	<i>Received at ESRF:</i>
Names and affiliations of applicants (* indicates experimentalists): F. Bonell*, J. Courtin*, M. Jamet Univ. Grenoble Alpes/CNRS/CEA IRIG-SPINTEC, 17 rue des Martyrs 38054 GRENOBLE cedex 9 R. Sant* ESRF, 71 avenue des Martyrs CS 40220 FR - 38043 GRENOBLE cedex 9		

Report:

(Fe_{1-x}Co_x)₅GeTe₂ (FCGT) is a high temperature van der Waals magnet synthesized only recently (2020) and so far exclusively studied in bulk form. Our objective was to employ XMCD as an element-sensitive magnetic probe to measure the individual Fe and Co magnetic moments, to clarify the ground state magnetic configuration in films with various compositions and to verify whether the material retains ferromagnetic ordering in the two-dimensional limit.

We studied FCGT monolayers (ML) grown at SPINTEC on Ge(111), with $x = 0, 0.15, 0.25$ and 0.5 . Since the samples had to be transferred in air, the films were capped with a 3-nm-thick CaF₂ layer. We systematically measured the XAS and MCD at the Fe and Co L-edges and at the Te M-edge. The measurements were performed with total electron yield (TEY) detection at 4 K, at an X-ray beam incidence of 30° from the surface, and under a magnetic field of ±1 T in order to saturate the magnetization along the beam. As shown in Fig. 1a, the fluorine present in the CaF₂ capping layer significantly contributed to the XAS signal (the F K-edge is located few eV below the Fe L-edge and is strongly structured). In order to correct the corresponding contribution to the Fe L-edge base line, we measured the XAS of a Ge(111)/CaF₂ (3 nm) reference sample under the same conditions. At the Co L-edge, our reference was a Ge(111)/Fe₅GeTe₂ (1 ML)/CaF₂ (3 nm) multilayer, in order to suppress the Fe L-edge fine structure in addition to one of the F K-edge. These reference XAS signals are represented as dashed grey lines in Fig. 1a. Correction of the raw data from these base lines yields the spectra in Fig. 1b. The sharp XAS line shapes, the significant MCD signal and the absence of multiplet structures suggest a posteriori that the interaction between CaF₂ and FCGT is weak or inexistent.

The ratio of XAS peak areas at the Co and Fe edges was found to be proportional to $x/(1-x)$. This confirms the good substitution of Fe with Co in the whole investigated composition range. The MCD signals were significant (~30% of the XAS at the Fe L₃), suggesting ferromagnetic order in the FCGT monolayers. The average Fe and Co magnetic moments were found to be ferromagnetically coupled. A small dichroic signal was also measured at the Te M-edge. The observed trends with varying x are expected to be indicative of the specific location and bonding of Co atoms in the cell. A theoretical investigation is under progress.

Last, we measured MCD field loops at the Fe L₃ energy in both normal and grazing incidence. A clear hysteresis and significant magnetic anisotropy was evidenced, confirming ferromagnetic ordering in monolayers. The largest anisotropy was observed for $x = 0.15$, which correlates with the largest Curie temperature ~200 K. These data establish FCGT as a (record-)high-temperature van der Waals two-dimensional ferromagnet.

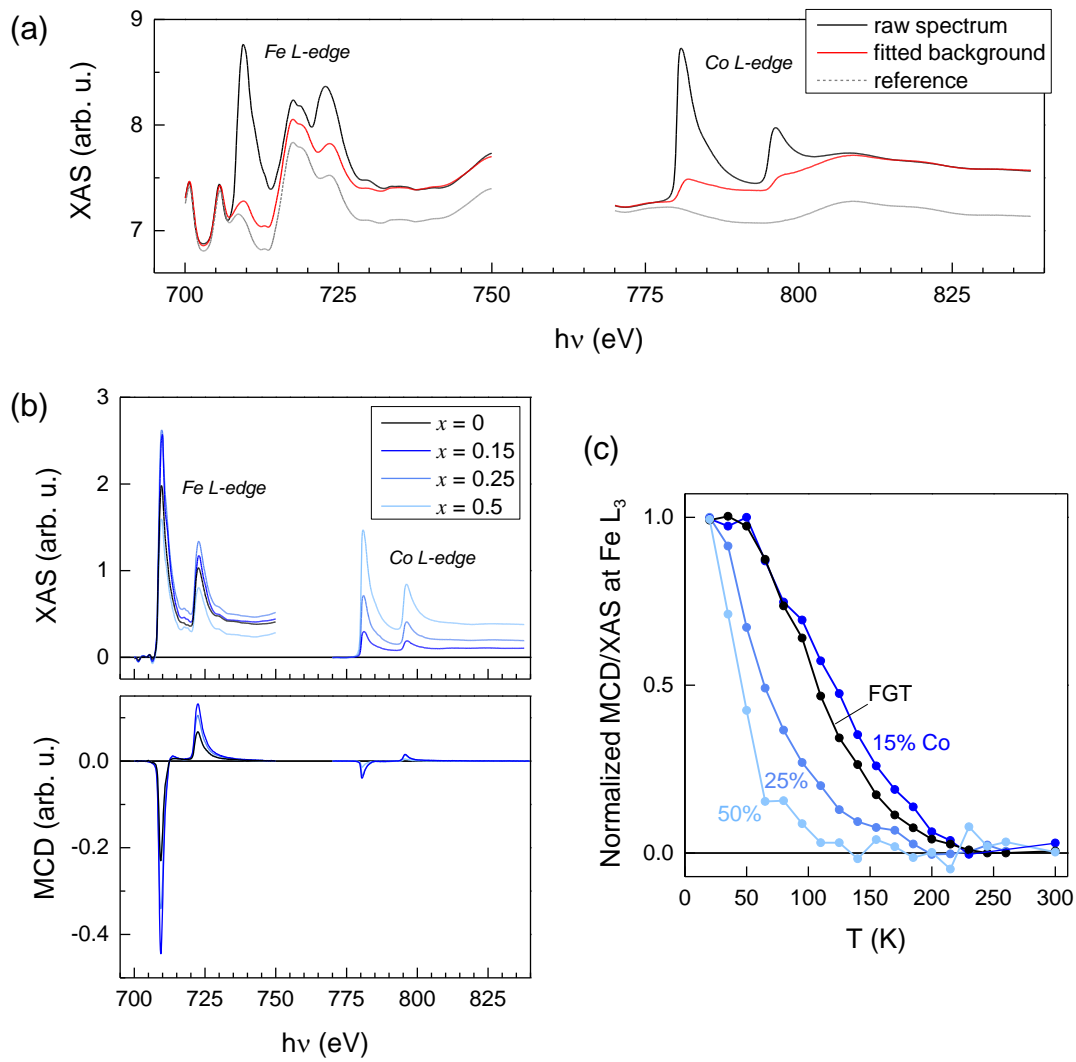


Fig.1 (a) Raw XAS spectra of $Ge(111)/(Fe_{0.5}Co_{0.5})_5GeTe_2$ (1 ML)/ CaF_2 (3 nm) (black solid line). The background (red line) is obtained by fitting the data in the pre- and post-edge regions with the sum of a CaF_2 reference XAS (grey dotted line) and a double-step function with steps located at L_3 and L_2 edges. (b) Background-corrected spectra for various Co contents x . (c) Temperature dependence of the MCD at Fe L_3 close to remanence (0.05 T) for various Co contents.