

# Large-area van der Waals epitaxy of ferromagnetic $\text{Fe}_3\text{GeTe}_2$ films on graphene

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Large-area growth of magnetic 2D materials and van der Waals (vdW) heterostructures is of paramount importance for the development of ultra-compact spintronic devices [1]. Here, we report on vdW epitaxy of the ferromagnetic metal  $\text{Fe}_3\text{GeTe}_2$  – a 2D crystal with tunable properties and great potential for room temperature ferromagnetism [2] – via molecular beam epitaxy using elemental Fe, Ge, and Te evaporated from Knudsen cells, and growth temperatures around 300 °C. Epitaxial graphene on 4H-SiC(0001) fabricated by high-temperature SiC surface graphitization [3] was utilized as a substrate. Morphological and structural characterization performed with different methods including Raman spectroscopy (see Fig. 1a) and synchrotron-based grazing incidence X-ray diffraction confirmed the formation of continuous  $\text{Fe}_3\text{GeTe}_2$ /graphene heterostructure films with stable interfaces and good crystalline quality. A robust out-of-plane ferromagnetism in the  $\text{Fe}_3\text{GeTe}_2$  films, comparable to state-of-the-art flakes (e.g.,  $T_{\text{Curie}} \sim 220$  K) [2], was confirmed by X-ray magnetic circular dichroism (XMCD) and magneto-transport investigations (see Fig. 1b,c). The latter also revealed the occurrence of the quantum Hall effect in the graphene (not shown), underlining the high quality of the epitaxially grown vdW heterostructure. These results are highly relevant for further research on wafer-scale growth of vdW systems containing magnetic 2D crystals for the realization of multifunctional, atomically thin devices.

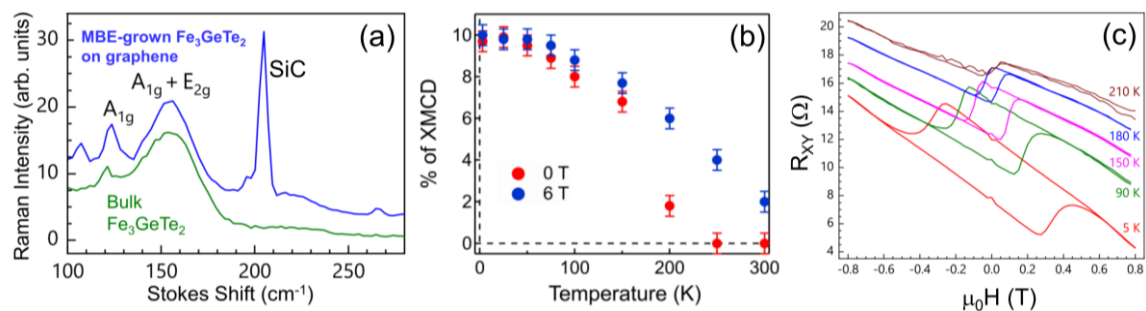
## References

[1] S.O. Valenzuela, S. Roche, Nat. Nanotechnol. **14**, 1088 (2019).

[2] Y. Deng et al., Nature **563**, 94 (2018).

[3] M. Heilmann et al., ACS Appl. Mater. Interfaces **12**, 8897 (2020).

## Figures



**Figure 1:** (a) A typical Raman spectrum for a 10 nm thick  $\text{Fe}_3\text{GeTe}_2$  MBE film showing the associated  $A_{1g}$  and  $A_{1g} + E_{2g}$  components (blue). A spectrum collected from a commercially available bulk crystal is plotted for comparison (green). (b) Temperature dependence of the Fe  $L_3$  XMCD maximum in saturation (6 T, blue) and remanence (red) for a 20 nm thick  $\text{Fe}_3\text{GeTe}_2$  film on graphene/SiC. (c) The anomalous Hall effect measured in a 1 cm<sup>2</sup> van der Pauw geometry for a 20 nm thick  $\text{Fe}_3\text{GeTe}_2$  film on graphene/SiC at different temperatures. From the magneto-transport results, a Curie temperature of about 220 K was obtained, in agreement with the XMCD analyses.