

Van der Waals epitaxy of 2D ferromagnetic Fe_{5-x}GeTe₂ films with Curie temperature above room temperature on graphene

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Abstract

Van der Waals (vdW) heterostructures combining layered ferromagnets and other two-dimensional (2D) crystals such as graphene and transition metal dichalcogenides are promising building blocks for the realization of ultra-compact devices with integrated magnetic, electronic and optical functionalities. Their implementation in various technologies depends strongly on the development of a bottom-up, scalable synthesis approach allowing to realize highly uniform heterostructures with well-defined interfaces between different 2D layered materials. It also requires that each material component of the heterostructure remains functional, which ideally includes ferromagnetic order above room temperature for 2D ferromagnets. In this contribution, we will present our recent results on van der Waals (vdW) epitaxy of the 2D itinerant ferromagnetic metal Fe_{5-x}GeTe₂ (FGT, $x \sim 0$) on single crystalline epitaxial graphene using molecular beam epitaxy. For the growth of FGT films (with thickness ranging from 10 to 15 nm), elemental Fe, Ge, and Te were co-supplied from conventional effusion cells, and a growth temperature of 300 °C was employed. As a substrate, epitaxial graphene on 4H-SiC(0001), synthesized via SiC surface graphitization, was employed. Morphological and structural characterization using methods such as atomic force microscopy, synchrotron-based grazing incidence X-ray diffraction, and scanning transmission electron microscopy (STEM) revealed that epitaxial FGT films exhibiting very good surface morphology, high crystalline quality, and a sharp interface to graphene could be realized. Interestingly, stacking faults related to the presence of single FGT layers with thicknesses exceeding those expected for the Fe₅GeTe₂ phase could be identified by STEM. We expect these to be novel FGT metastable phases with Fe composition higher than 5 and potentially enhanced magnetic properties. Temperature-dependent magneto-transport measurements and superconducting quantum interference device (SQUID) magnetometry were employed to assess the magnetic properties of the samples. Ferromagnetic order with a predominant out-of-plane magnetization was shown to persist above 350 K. Furthermore, magneto-transport also revealed that the epitaxial graphene continues to exhibit a high electronic quality. These results represent an important advance beyond non-scalable flake exfoliation and stacking methods, thus marking a crucial step toward the implementation of ferromagnetic 2D materials in practical applications.