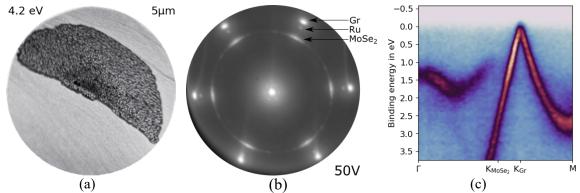
## In-situ growth characterization of 2D heterostructures: MoSe<sub>2</sub> on intercalated graphene/Ru(0001)

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The combination of graphene with other two-dimensional materials in vertically stacked 2d heterostructures gives rise to the possibility of altering the electronic properties of both materials in the attempt of tailoring new structures for the application in electronics, catalysis, or sensors. Despite the great interest, most of the investigated 2d heterostructures were realized by mechanical exfoliation and chemical vapor deposition in the millibar range, and true *in-situ* characterization of the growth process is rarely found in the literature. On *in-situ* prepared samples, Dau et al. [1] have shown that a single layer of MoSe<sub>2</sub> grown on few-layer graphene is effectively n-doped and a bandgap of 250meV is opened at the Dirac Point of graphene. To understand the growth process and its significance in terms of electronic properties, here, we have investigated the growth of singlelayer MoSe<sub>2</sub> on single-layer graphene on Ru(0001) via real-time in-situ LEEM and µLEED measurements. The graphene is grown by deposition of carbon from an ethylene precursor at elevated temperatures. After the preparation of graphene, MoSe<sub>2</sub> has been prepared via codeposition of Mo and Se at 250°C as shown in Figure 1a). When only partially covering the Ru(0001) surface with graphene, intercalation of selenium atoms through the edges of the graphene can be observed, which leads to a decoupling of the graphene and the substrate and appears to enhance the subsequent growth of MoSe<sub>2</sub> on the graphene. Rotational ordering of the MoSe<sub>2</sub> is achieved by annealing at elevated temperatures, which is strongly enhanced by the pronounced mobility of single-domain MoSe<sub>2</sub> islands that align with the high symmetry orientations of the underlying graphene (cf. Figure 1b), indicating a non-negligible interaction between the two vander-Waals materials. XPEEM and µARPES (cf. Figure 1c) prove the monolayer nature of the asgrown MoSe<sub>2</sub> as well as the free-standing character of the Se-intercalated graphene underneath. Interestingly, the charge-transfer doping between the graphene and the intercalant is effectively reversed by the charge transfer between the graphene and the MoSe<sub>2</sub>, leading to a virtually undoped, freestanding graphene inside the selenium-MoSe<sub>2</sub> sandwich.



**Figure 1**. (a) BF-LEEM image of MoSe2/Gr/Ru(0001). The lens-shaped graphene island appears dark while the MoSe<sub>2</sub> is visible as bright spots inside the island. (b)  $\mu$ LEED pattern obtained on the graphene island. The reflections of the respective materials are marked. (b)  $\mu$ ARPES of MoSe2/Gr/Ru(0001) obtained on the graphene island.

## References

[1] M. Dau, M. Gay, et al., ACS NANO 12, 2319 (2018)