Tailoring few-layers van der Waals crystals through galvanostatic molecular intercalation

Daniel Tezze[1]
José M. Pereira[1], Covadonga A. García[1], Enrique A. García[1], Felix Casanova[1,3], Beatriz Martín-García[1,3], Maider Ormaza[4], Luis E. Hueso[1,3] and Marco Gobbi[2,3].

[1] CIC nanoGUNE BRTA, 20018 San Sebastian, Spain.
[2] Materials Physics Center CSIC-UPV/EHU, 20018 Donostia-San Sebastian, Spain.
[3] IKERBASQUE, Basque Foundation for Science, 48013 Bilbao, Spain.
[4] Departamento de Polímeros y Materiales Avanzados: Física, Química y Tecnología, Universidad del País Vasco, Paseo Manuel de Lardizabal 3, San Sebastián 20018, Spain.
d.tezze@nanogune.eu

Intercalation of guest species in van der Waals materials is an effective technique for customizing their properties.[1–2] The process can be achieved through different methods, that have been explored predominantly for bulk samples[3], which are impractical for the fabrication of novel nanodevices. The use of intercalated compounds in (quantum) electronic devices [4] demands controlled intercalation regimes in reduced-dimensions systems (thin flakes). This task is particularly challenging due to the complexity of the setup involved in conventional intercalation approaches. Herein, we present a paradigmatic novel route based on a galvanostatic electrochemical approach (**fig. 1a**) to directly intercalate pre-exfoliated 2H-TaS_2 flakes with cationic molecular species, revolutionizing its the state-of-the-art intercalation strategy. Our methodology yields intercalated 2H-TaS_2 flakes with unprecedented crystallinity (**fig. 1b**), crucial for integration in devices for electronics and quantum computing. Different molecular cations can be controllably intercalated, permitting to tune the on-set of superconductivity (**fig. 1c**). Moreover, we tailor the intercalation mechanism to spatially control phases within a single flake, potentially enabling van der Waals lateral heterojunctions.

References

[1] J. Zhou *et al. Adv. Mat. 33* (**2021**) 2004557
[2] J. M. Pereira *et al. Advanced Physics Research* **2023**, *2*, 2200084.
[3] H. Zhang, *et al. Science B. 65* (**2020**) 188
[4] Z. Li, *et al. Small Meth. 5* (**2021**) 2100567

Figures

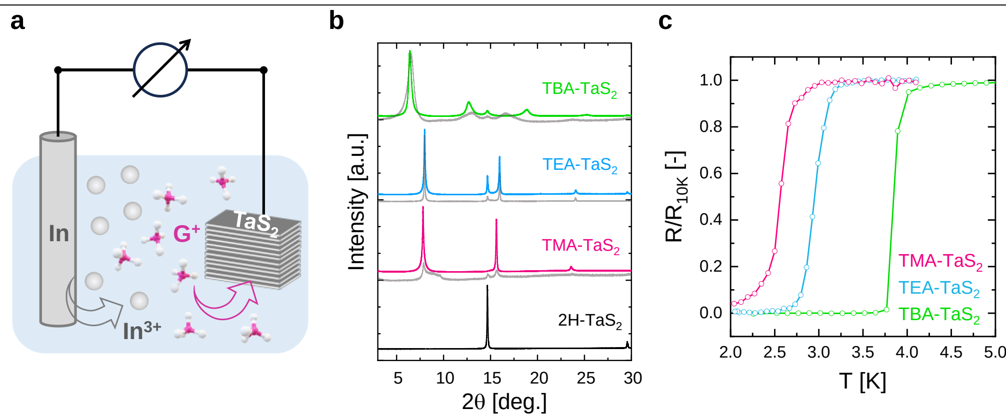

Figure 1: a) Schematic of the galvanostatic cell employed for the intercalation of 2H-TaS_2 with organic ions (G^+). **b)** XRD patterns of pristine 2H-TaS_2 and three alkylammonium intercalates (TMA_xTaS_2, TEA_xTaS_2 and TBA_xTaS_2), obtained by the galvanic approach (coloured lines) and the XRD patterns of the same intercalates using the chemical approach (grey lines). **c)** Normalized resistance measured for three alkylammonium intercalated 2H-TaS_2 thin flakes (~10L).