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## **Topochemistry and Delamination of Layered Topological Insulators**

M. Lê Anh, M. Ruck

Technische Universität Dresden, Bergstraße 66, Dresden, Germany mai.le\_anh@tu-dresden.de

Topological insulators (TI) gained high interest due to their protected electronic surface states that are sheltered against scattering due to the specific symmetry (topology) of the electronic band-structure. This manifests in almost dissipation-free electron transport. Thus, TIs are envisioned as promising candidates in the field of nanoelectronics and in quantum computing.<sup>[1,2]</sup> For technological application thin layers are favored. And this is precisely where our research deploys when it comes to the synthesis and delamination of 3D layered TI structures to thin 2D TI sheets. The majority of TIs are 3D structures in which layers are stacked in an ordered manner, promoted by van-der-Waals, e.g. in Bi<sub>2</sub>Tel<sup>[3]</sup>, or by ionic interactions, e.g. in Bi<sub>14</sub>Rh<sub>3</sub>I<sub>9</sub><sup>[4]</sup> (Figure 1 left). The latter one consists of <sup>2</sup><sub>∞</sub>[(Bi<sub>4</sub>Rh)<sub>3</sub>I]<sup>2+</sup> intermetallic nets that are separated by alternating (Bi<sub>2</sub>I<sub>8</sub>)<sup>2–</sup> anionic spacers. Indeed, the real challenge, but at the same time the feasible benefit, was the layer charge. Respectively, we introduced two strategies for the wet-chemical delamination of Bi<sub>14</sub>Rh<sub>3</sub>I<sub>9</sub>: (A) The *reductive* approach removes the layer charge and decreases the coupling of adjacent intermetallic layers by reaction with *n*-butyllithium in *n*-hexane or 1-dodecanthiol in oleylamine. (B) In the *substitutive* approach the anionic spacer is exchanged for a larger organic zwitterion, e.g. betaine in a deep eutectic solvent (DES) with adipic acid or in suitable solvents, to increase the distance between the intermetallic nets progressively. Both strategies led to a great expansion of the layered structures. Moreover, flakes with about 10 layers thickness and areas up to 100  $\mu$ m<sup>2</sup> of Bi<sub>14</sub>Rh<sub>3</sub>I<sub>9</sub> as well as of Bi<sub>2</sub>TeI were isolated (Figure 1 right).

## References

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## **Figures**



**Figure 1:** Left to right: Crystal structures of  $Bi_2TeI$  and  $Bi_{14}Rh_3I_9$ ; SEM and TEM images of  $Bi_{14}Rh_3I_9$  crystals after the reductive approach with *n*-butyllithium in *n*-hexane resulting in swollen crystals and irregular sheets and after the substitutive approach with betaine in dimethylformamide leading to a shifting of sheets and irregular exfoliated sheets.