Properties of Electron-Beam-Modified Low-Dimensional Inorganic and Organic Materials

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Low-dimensional materials exhibit properties, which often differ strongly from those of the bulk counterparts and offer unique opportunities for new and miniaturized electronic and optical devices [1]. In situ electron microscopy allows to functionalize the thin material while imaging. Here we present recent results using our unique chromatic and spherical aberration-corrected SALVE instrument both in imaging and spectroscopy [2].

We first discuss the formation of defects in two-dimensional inorganic and organic crystals. For transition metal di-chalcogenides (TMDs) we show that the formation of vacancies is possible at electron voltages nearly half of the knock-on threshold and quantify the damage [3,4]. Further, we analyse in-situ structural and chemical modifications of different freestanding transition metal phosphorus tri-chalcogenides (TMPTs). The atomically resolved analysis of freestanding few-layer TMPTs is difficult, and rarely reported due to their susceptibility to oxidation. Here we present our work on the effect of electron irradiation in freestanding few-layer TMPTs at TEM acceleration voltages between 30 and 80 kV. Our first-principle calculations predict that the electron beam predominantly removes sulphur from the sample, which is confirmed by combined EELS and EDX experiments. In addition, we conduct in-situ experiments to study annealing and electron-beam-induced structure transformations of few-layer $MnPS_3$. We observe the formation of new phases with the net formula $MnS_{(1-x)}P_x$ and show that suitable dose rates allow their controlled growth embedded in the antiferromagnetic host. Complementary ab-initio calculations prove the stability of the new phases and predict their magnetic and electronic properties. As the TMPTs are often very oxygen-sensitive, they were prepared with the help of a newly-developed polymer-assisted sample preparation method [5].
We also present studies on the structure of two-dimensional polymer crystals and show that an accelerating voltage of 120kV is optimal to resolve the structure on the molecular level [6]. Furthermore, we present in-situ studies of a miniaturized electrochemical cell, where reversibly single-crystalline bilayer graphene is lithiated and delithiated in controlled manner using an electrochemical gate confined to a device protrusion [7]. Moreover, we report on the nucleation mechanism of the formed crystalline lithium phase, which we now understand on the level of the single atoms. We show that differentiating between the bond nature between two metal atoms is now possible [8].

References

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