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Chemistry of low-dimensional materials on the atomic level

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In this study we demonstrate that the accelerated electrons in the transmission electron microscope (TEM) can act as stimulus and initiator for reactions while imaging; detailed understanding of the beam electron– specimen interactions is required. We present recent results on atomically-resolved, time-dependent in-situ TEM imaging using the chromatic- and spherical-aberration-corrected low-voltage SALVE instrument operating in the voltage range between 80kV and 20kV [1-5]. First, we elucidate the accelerating-voltage-dependent formation of defects in the case of single-layer MoS_2. The results suggest that elastic and inelastic interactions are strongly connected resulting in a twostep interaction process and density functional theory molecular dynamics shows that excitations in the electronic system can form vacancies through ballistic energy transfer at electron energies, which are much lower than the knock-on threshold for the ground state [6].
We further identify the structure of electron-beam-induced defects and their electronic properties and follow the migration paths and associated property changes in a variety of 2D TMD and TMPT crystals [6,7] and analyse in-situ structural and chemical modifications of different freestanding transition metal phosphorus tri-chalcogenides (TMPTs) from atomically resolved imaging, low-loss and high-loss EELS and 3D electron diffraction. We calculate the displacement thresholds, electronic properties, and the displacement cross-section of single vacancy S and P by ab-initio calculations which help understanding the observed structural transformations. Moreover, our results provide insight into layer-number-dependent changes in the electronic and crystallographic properties as result of lowering the dimensionality. As the TMPTs are often very oxygen-sensitive, they were prepared with the help of our newly-developed polymer-assisted sample preparation method [8,9].
We show moreover that also 2D polymers and their defects can be resolved at near-atomic level [10] which can be further increased, when imaging at 120kV.
On the fundamental base of chemistry, we show that differentiating between the bond nature by measuring the distance between two metal atoms is possible, when confined in the narrow space of a SWNT [11]. We show here the interaction between the moving matter and the carbon nanotube and its application for carrying metal atoms to a nucleation seed, realizing in situ observation of metal nucleation. we find that crystal nucleus formation occurs through a two-step nucleation mechanism [12]. Finally, we intercalate bilayer graphene in-situ by lithium, study not only the in-situ the lithiation and delithiation processes [13], as well as the formation process of the new high-density crystalline Li- phase.

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