Chemical vapor growth and delamination of 2D honeycomb transition metal halide MX_3 nanosheets

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The 2D honeycomb structure of graphene gave an idea to the introduction of materials with congeneric pattern. Thus, transition metal halides MX_3 were highly profiled in theory, as well by means of ab initio calculations. [1] The honeycomb layers allow for strong bond anisotropy and frustration that structures could stabilize new patterns of cooperative magnetic interactions or a spin liquid state. [2] Furthermore transition metal halides are suspected to exhibit multiferroic properties or form monolayer magnets. [3, 4] One candidate to realize a Kitaev Heisenberg (KH) model is the 2D layered honeycomb magnet α-Ruthenium(III) chloride (α-$RuCl_3$) with strongly frustrated, anisotropic interactions between spin-orbit entangled J_{eff} = ½ $4d^5$ Ru^{3+} magnetic moments. [5] As alteration of physical properties on the nanoscale additionally is intended, new synthesis approaches to obtain phase pure MX_3 nanocrystals have been audited. Crystal growth of thin nanosheets succeeded via chemical vapor transport (CVT, see Fig. 1) on specific substrates for the two dimensional alpha modifications of $RuCl_3$, $MoCl_3$, $TiCl_3$, CrI_3 and $CrBr_3$ (see Fig. 2). The crystal properties are characterized by means of optical and electron microscopy, AFM, SAED, micro-RAMAN and XPS proving the desired composition, morphology, high crystallinity and phase-purity. Furthermore individualization and delamination of nanosheets on substrates down to the monolayer limit (≤ 1 nm) has been realized by means of substrate exfoliation and ultrasonication experiments in a reproducible way (see Fig. 1 & 3).

References

[1] Nicolosi, V. et al., Science, 340 (2013), 1226419-1-18
[2] Johnson, R. et al., Physical Review B, 92 (2015), 1-12
[3] Tokunaga et al., Physical Review B, 84 (2011), 2-5
[4] McGuire et al., Chemistry of Materials, 27, (2015), 612-620
[5] Kitaev, A., Annals of Physics, 321 (2006), 2–111

Figures

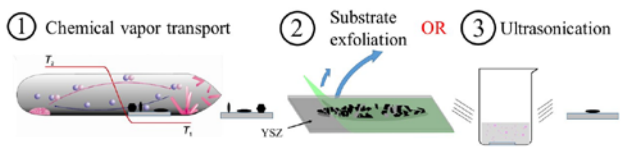

Figure 1: Concept of experimental approach

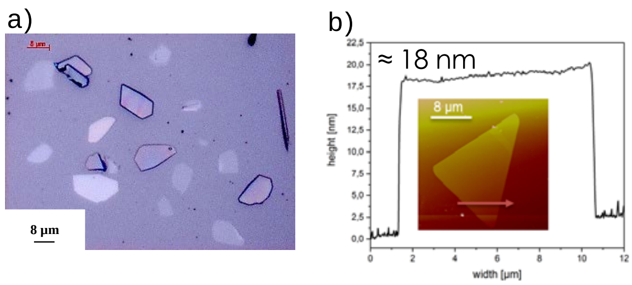

Figure 2: Structural characterization of as grown α-$RuCl_3$ nanosheets on a substrate: a) optical microscopy, b) AFM measurement of α-$RuCl_3$ nanosheet (inset: image of investigated crystal)

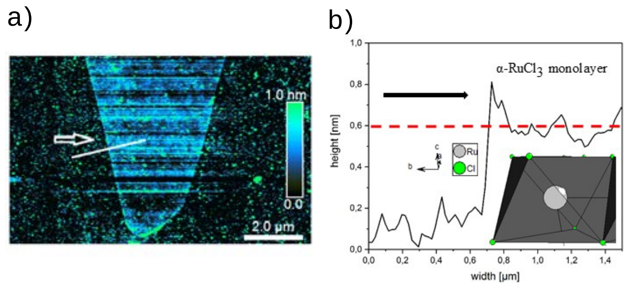

Figure 3: AFM measurement of an α-$RuCl_3$ monolayer on a substrate after ultrasonication treatment: a) investigated monolayer, b) AFM measurement (insets: α-$RuCl_3$ crystal structure and theoretical monolayer height as red line)