Ionic 2D Materials based on Alkali Halides

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A key requirement in the field of 2D materials is the development of new systems with a higher bandgap than those currently available, which is currently around 6 eV. Furthermore, these materials should enable the easy, reproducible introduction of impurities and defects while maintaining crystallinity and stability, thereby enabling their use in photonic devices and other applications. In this project, the challenge was to obtain 2D flakes from a 3D ionic starting material with a cubic rock salt structure (e.g., alkali halides such as NaCl, NaF, KCl, KF, or CsF). They will have physical properties that differ substantially from those of currently known 2D materials. For example, there are a few large bandgap 2D materials, almost exclusively h-BN layers. The h-BN bulk system has a band gap of around 4 eV, and its monolayer has a band gap of 6 eV, while the employed alkali halides in this project have bulk band gaps of 10.0 eV (CsF), 10.9 eV (KF), 11.7 eV (NaF), 8.5 eV (KCl), and 8.6 eV (NaCl). Furthermore, h-BN poses a serious challenge for incorporating numerous defects in a stable and reproducible manner, limiting its potential applications in devices such as single-photon emitters. The crucial point is how it is possible to exfoliate an isotropic 3D material with ionic interactions in three spatial directions.[1,2] The intercalation of bromine molecules in CsF was corroborated, many years ago, by X-ray single-crystal diffraction.[1,3, see Figure 1a] The intercalated structure is analogous to that of other conventional 2D materials; for example, graphene also with bromine molecules. This intercalation into the 3D structure of the alkali halide has two effects: one is to generate an anisotropic 2D structure with alternating monoatomic CsF layers and Br_2 molecular layers. The second important effect is that, now, for the exfoliation of the CsF·Br_2 compound, only the non-ionic F···Br-Br interactions must be broken. Preliminary DFT calculations indicate that such contact energies are of the same order of magnitude as other non-covalent interactions of common 2D materials. The intercalation of such 2D materials has been performed by the intercalation of Br_2 and IBr molecules (see Fig 1b-d) combined with different exfoliation techniques (sonication-centrifugation, ball milling, microwaves..). The characterization of the 2D flakes has been performed using Atomic Force Microscopy. The ultimate goal of this project is to exploit the advantages of this new family of 2D systems, such as single-photon emission and superfluorescence, since alkali halides are textbook model systems for introducing defects and dopants.

References

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Figures

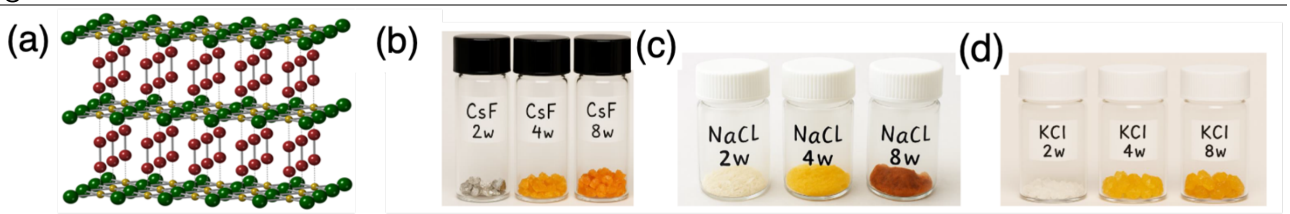

Figure 1: (a) X-ray single-crystal diffraction of CsF·Br_2 compound. (b-d) Intercalated samples of different alkali halides after some weeks of reaction with bromine,