**Dominant in-plane cleavage direction of CrPS4 monolayer**

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In-plane cleavage directions of 2D crystals are displayed and often well-defined in their flakes exfoliated by the widely-used scotch-tape method. Here, we investigate the correlation between dominant in-plane cleavage direction and elastic properties in different directions. CrPS4 flakes show a preferential in-plane cleavage direction of 67.5˚, corresponding to <110> direction. To explain it, we calculated directional dependence of Young’s modulus and fracture energy using first-principles density functional theory calculations. We found that fracture energy is directly relevant to the in-plane cleavage direction of CrPS4. Our study can provide a facile approach to figure out the direction of 2D crystals without complex characterization process, which is valuable for material processing of 2D materials.

1. Joe, M. et al., J. Phys. Condens. Matter **29**, 405801 (2017).