Structure, Electronic, and Magnetic Properties of Non-van der Waals Two-Dimensional Materials from Data-driven Design

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While two-dimensional (2D) materials are traditionally derived from bulk layered compounds bonded by weak van der Waals (vdW) forces, the recent surprising experimental realization of non-vdW 2D compounds obtained from non-layered crystals [1,2] foreshadows a new direction in 2D systems research. To elucidate their structure and properties, electron microscopy and first-principles calculations are indispensable tools. Contributing to the predictive design of these novel nanoscale compounds, here, we present several dozens of candidates derived from applying data-driven research methodologies in conjunction with autonomous first-principles calculations [3,4]. We find that the oxidation state of the surface cations of the 2D sheets as well as accounting for strong surface relaxations upon exfoliation are crucial factors determining their stabilization.The candidates exhibit a wide range of appealing electronic, optical and in particular magnetic properties owing to the (magnetic) cations at the surface of the sheets. Despite of several ferromagnetic candidates, even for the antiferromagnetic representatives, the surface spin polarizations are diverse ranging from moderate to large values modulated in addition by ferromagnetic and antiferromagnetic in-plane coupling [3]. These features can be accessed by experimental techniques such as (spin-polarized) scanning tunnelling microscopy (STM). At the same time, chemical tuning by surface passivation provides a valuable handle to further control the magnetic properties of these novel 2D compounds [5] thus rendering them an attractive platform for fundamental and applied nanoscience.

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