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Discovering Two-Dimensional Non-van der Waals Materials

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While two-dimensional (2D) materials are traditionally associated with bulk layered compounds bonded by weak van der Waals (vdW) interactions, the recent surprising experimental realization of *non*-vdW 2D compounds obtained from non-layered crystals [1,2] opens up a new direction in 2D systems research. These materials show several distinct differences to traditional 2D sheets as their surface was revealed to be terminated by cations rather than anions. Here, we outline several dozens of candidates of this novel materials class derived from employing data-driven research principles in conjunction with autonomous *ab initio* calculations [3,4] (Figure 1). The oxidation state of the surface cations of the 2D sheets turns out to be an enabling descriptor regarding the manufacturing of these systems as it determines their exfoliation energy: small oxidation states promote easy peel off [3]. When extending the set from oxides to sulfides and chlorides, the exfoliation energy becomes ultra low due to strong surface relaxations [4]. The materials also pass several tests validating their vibrational and dynamic stability. The candidates exhibit a wide range of appealing electronic, optical and in particular magnetic properties making these systems an attractive platform for fundamental and applied nanoscience.

References

[1] A. Puthirath Balan *et al.*, Nat. Nanotechnol. **13** (2018) 602.
[2] A. Puthirath Balan *et al.*, Chem. Mater. **30** (2018) 5923.
[3] R. Friedrich *et al.*, Nano Lett. **22** (2022) 989.
[4] T. Barnowsky *et al.*, Adv. Electron. Mater. (2023) 2201112.

Figures

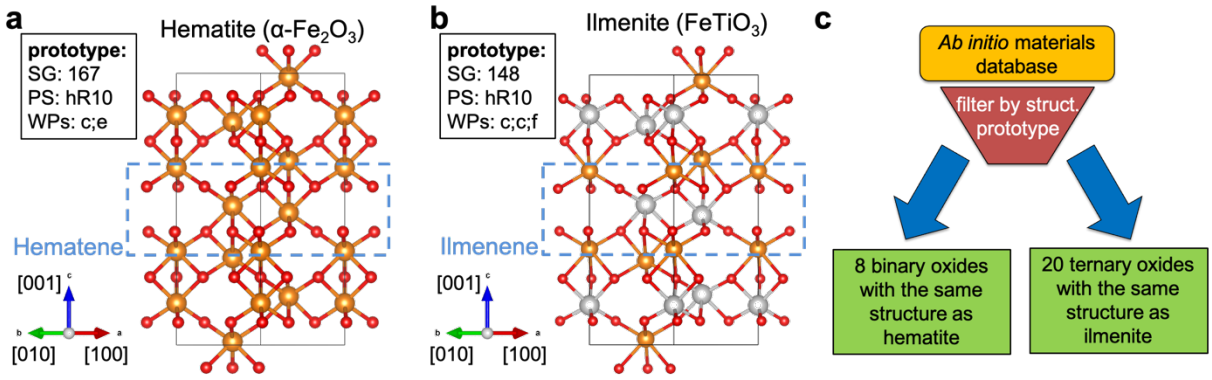

Figure 1: 2D materials from non-vdW systems. Atomic structure of (**a**) hematite (α-Fe_2O_3) and (**b**) ilmenite ($FeTiO_3$), the first non-vdW bulk materials with 2D analogues [1, 2]. For both prototypes, space group (SG), Pearson symbol (PS), and Wyckoff positions (WPs) are indicated in the respective boxes. In each case, the exfoliable [001] facet (monolayer) is indicated in the blue dashed frame leading to hematene and ilmenene 2D materials. Colors: Fe orange, O red, and Ti light gray. The black line denotes the conventional unit cell. (**c**) Schematic workflow illustrating how the candidate systems were obtained from the *ab initio* materials database using structural information as input.