

## Crystal structure prediction in novel nitrides: The roles of metastability and disorder

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Compared to oxides, nitrides have similar or higher cohesive energy, bond strength, and hardness. Yet, nitrides do not form minerals, and comprise a much smaller space of known compounds and crystal structures when compared to oxides. This paradox is due to the exceptionally large binding energy of the N<sub>2</sub> molecule and the resulting relative thermochemical instability of nitrides. On the other hand, nitrides have a higher propensity to form metastable structures [1]. As a part of a broader nitrides discovery effort, we have predicted the crystal structures of the previously unknown nitrides Sn<sub>2</sub>N<sub>2</sub>, Ti<sub>3</sub>N<sub>4</sub> [2] and the Zn-Mo ternaries Zn<sub>3</sub>MoN<sub>4</sub> and ZnMoN<sub>2</sub> [3], using an first principles sampling approach based on kinetically limited minimization [3]. In each of these cases, the observed crystal structures match with computational predictions only after taking into account the role of disorder. The matching structures lie up to 150 meV/at above the predicted ground state. This study highlights the need to incorporate metastability and disorder in computational crystallography.

[1] W. Sun et al., *Sci. Adv.* 2, e1600225 (2016).

[2] V.S. Bhadram et al., *Phys. Rev. Mater.* 2, 011602(R) (2018).

[3] E. Arca et al., *J. Am. Chem. Soc.* 140, 4293 (2018).