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First-Principles Molecular Crystal Structure Prediction: The Importance of Collective van der Waals Interactions and Free Energies JOHANNES HOJA, Physics and Materials Science Research Unit, University of Luxembourg, HSIN-YU KO, ROBERTO CAR, Department of Chemistry, Princeton University, ROBERT A. DISTASIO JR., Department of Chemistry and Chemical Biology, Cornell University, ALEXANDRE TKATCHENKO, Physics and Materials Science Research Unit, University of Luxembourg — For meaningful crystal structure predictions (CSP) of practically relevant molecular crystals, it is imperative to calculate relative lattice energies with an accuracy of about 1 kJ/mol since it is necessary to correctly rank the stabilities of numerous low-energy polymorphs. Accurate results can be obtained by utilizing density-functional theory (DFT) supplemented by a high-level van der Waals (vdW) model, such as the many-body dispersion (MBD) method. Since relative stabilities have to be determined at finite temperatures, vibrational free energies have to be considered as well. The importance of an accurate treatment of vdW interactions and free energies is discussed in terms of the systems studied in the most recent CSP blind test [Reilly et al., *Acta Cryst. B* 72, 439 (2016)], with a particular focus on a challenging case of a highly polymorphic system. It is shown that collective many-body vdW interactions completely change the relative stabilities of the studied polymorphs in comparison to a pairwise approach. Furthermore, the inclusion of thermal fluctuations also leads to a significant improvement of the relative stabilities. Overall, the CSP approach based on DFT+MBD provides remarkably accurate structures and stability rankings for relevant molecular crystals.

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