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# Demystifying density functional theory for the crystal engineering community

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Organic solid-state chemists and crystal engineers increasingly turn to computational modeling to determine stability relationships between molecular crystal polymorphs, solve challenging crystal structures, predict crystal properties, and assess the potential for future polymorph discoveries. Reliable predictions, however, demand a meticulous treatment of the delicate interplay of hydrogen bonding, weak dispersion, and polarization that governs crystal packing. Over the past two decades, periodic density functional theory (DFT) has emerged as a central pillar in the successful modeling of the organic solid state (Hunnisett *et al.*, 2024; Hoja *et al.*, 2019; Firaha *et al.*, 2023; Price *et al.*, 2023). It has become a vital ingredient for accelerating solid-state discovery across pharmaceutical formulation, organic electronics, and functional materials design (Beran, 2023).

For newcomers to the field, modern computational chemistry software makes running periodic DFT calculations deceptively simple – yet obtaining reliable results requires more than blind button pushing. The choice of density functional, dispersion correction, basis set, *k*-point grid, and other model parameters can each exert a significant influence on calculated lattice energies. Resolving the sub-kilojoule-per-mole energy differences that separate competing polymorphs demands careful attention to every parameter. Fortunately, clear practical guidance can make learning to navigate these choices entirely manageable.

In their highly pedagogical contribution to the *Best Practice in Crystallography* series in *Acta Crystallographica Section C: Structural Chemistry*, van de Streek & Johnson (2026) offer an accessible road map for performing 0 K periodic DFT calculations on molecular crystals. They open with fundamental topics ranging from selecting appropriate density functionals and dispersion corrections to setting up core job parameters, making the article immediately useful to first-time users. The true highlight of the paper, however, lies in its practical wisdom found in later sections: the subtle tips and tricks that rarely make it into the published literature. The authors cover strategic choices in unit-cell representation to boost convergence, hydrogen-position normalization to accelerate calculations, and methods for quantitative crystal structure comparison. They also tackle essential real-world topics, including error assessment, uncertainty estimation, error cancellation, the limitations of common density functionals, and issues associated with complex cases such as salts, cocrystals, tautomers, and multi-component systems. Acquiring this level of practical insight would otherwise require sifting through the vast literature in the field and years of trial-and-error calculation experience. van de Streek and Johnson's guide should be essential reading for anyone entering organic crystal modeling, and even seasoned veterans will find valuable new insights within its pages (as did this author).

Looking forward, solid-state modeling and crystal structure prediction are rapidly entering an era driven by machine-learning interatomic potentials (MLIPs) (Zheng *et al.*, 2026; Gharakhanyan *et al.*, 2025; Midgley *et al.*, 2026). Trained on extensive quantum-chemical datasets, MLIPs can deliver near-DFT accuracy at a fraction of the computational cost. Yet even as these models transform the field, periodic DFT calculations will remain essential. High-level DFT calculations will continue to serve as the basis for generating high-quality training data, refining MLIP-predicted energy landscapes, and evaluating electronic or optical properties beyond an MLIP's scope. Ultimately, because MLIPs are only as robust as the DFT-included physics baked into their training datasets,

a firm grounding in periodic DFT will remain invaluable for understanding both the capabilities and the boundaries of MLIP-driven discovery.

## Conflict of interest

The author has no conflicts of interest to declare.

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