Quantum crystallography for studying dynamics and disorder in crystals

Anders Østergaard Madsen

University of Copenhagen

a.madsen@sund.ku.dk

Crystals are complicated. A thorough model of a crystal must describe not only the atomic positions, but also the atomic and molecular vibrations and possible disorder. Whereas the static models of charge densities refined against high-resolution diffraction data have been compared extensively to high-level periodic ab-initio calculations, there has been less focus on the use of ab-initio calculations in the modelling and interpretation of dynamics and disorder, in particular with respect to molecular crystals.
In this presentation, we walk through historical and recent approaches to use quantum mechanics as a set of spectacles through which we can interpret crystallographic experiments and understand the dynamics and disorder in crystals. A plethora of important bulk solid state properties depend on crystal vibrational properties and disorder. The molecular motion and disorder affect the entropy of crystals and are therefore crucial for understanding stabilities and phase changes. Mechanical properties, such as the elastic moduli of the crystal, are also intrinsically linked to the crystal lattice dynamics. In recent years, we have developed an approach to refine vibrational modes derived from periodic ab-initio calculations against X-ray diffraction data [1-2] This approach, called normal mode refinement, have been used in studies of phase transformations and polymorphic stabilities in molecular crystals [3-5].

[1] Hoser, A. A. & Madsen, A. Ø. (2016). Acta Cryst A 72, 206–214.

[2] Hoser, A. A. & Madsen, A. Ø. (2017). Acta Cryst A 73, 102–114.

[3] Hoser, A. A., Sztylko, M., Trzybiński, D. & Madsen, A. Ø. (2021). Chem. Commun.

[4] Kofoed, P. M., Hoser, A. A., Diness, F., Capelli, S. C. & Madsen, A. Ø. (2019). IUCrJ 6, 558–571.

[5] Sovago, I., Hoser, A. A. & Madsen, A. Ø. (2020). Acta Cryst A 76, 32–44.