

Estimation of thermal diffuse scattering from *ab-initio* lattice-dynamical models

*Monika Kovacic*¹, *Anders Ø. Madsen*¹

1. Department of Chemistry, University of Copenhagen, Universitetsparken 5, Copenhagen, 2100, Denmark

Thermal diffuse scattering (TDS) originates from scattering with the collective lattice vibrations (phonons) within a crystal and, thus, its effect increases with the temperature-dependent population of phonon modes. The continuously distributed TDS has its dominant contributions near the reciprocal lattice vectors giving rise to larger Bragg peak intensities and, in the modeling stage, smaller Debye-Waller (DW) factors.

Typically measurements for charge density studies are conducted at 100 K, where it is not known to what extent TDS affects the data. This has consequences for the correct deconvolution of thermal motion from the static charge density. The common strategy is to either ignore the presence of TDS or to reduce the TDS by cooling the crystals to the lowest obtainable temperatures.

The latter can be subject to experimental complications, especially, when applying external perturbations to the system in the case of pressure or excited state studies, for instance, or in the study of solid state phase transformations that are often affected by thermal effects. Nevertheless, in many cases, there remain a considerable difference between the DW factors obtained from X-ray and neutron scattering experiments {Blessing:1995vk}, suggesting the importance of TDS corrections to the scattering intensities.

Fortunately, the advances within computational power provide a strategy for estimating the TDS contribution on larger molecular systems by deriving the lattice-dynamics from *ab-initio* calculations. We pursue this in the present work using pyrazinamide, a polymorphic anti-tuberculosis drug, as our model system. Additionally, we contrast the *ab-initio* calculations to inelastic X-ray and neutron scattering studies, respectively.