An all-order phonon approach to thermal diffuse scattering

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In scattering experiments, phonons manifest themselves as Thermal Diffuse Scattering (TDS). Recent research has focused on phonons due to their role in various properties. This includes interactions like phonon-electron coupling in superconductivity and phonon-spin coupling, which finds application in spintronics, among other significant interactions. Understanding phonons and detecting phonon anomalies in materials is crucial in determining the material properties and their phase stabilities. Phonons are particularly important in elastic properties, with applications spanning from geology, where they aid in discerning planetary composition and temperature through the analysis of seismological wave propagation, to research in quantum phase transitions exploring their formation in crystals and interaction with other quasi-particles [1]. However, regular TDS, predominantly from acoustic phonon modes, often masks anomalies in phonons. While software exists for one or two-phonon processes, higher-order contributions are significant in certain systems. We are currently developing software capable of describing these higher-order contributions.

TDS arises from the scattering of X-rays by phonons and is present in all crystals. It contains valuable insights into phonon dispersion relations and the crystal elastic properties. Moreover, in many crystals static displacements give rise to intensity around the Bragg peaks which is very similar in shape to TDS, and thus can also be efficiently modelled using a similar approach. Currently, there are several programs available for modelling TDS, such as TDS2EL [2] and AB2TDS [3], however they have some limitations. In particular, TDS2EL allows to extract elastic constants from the TDS, but it can only model limited regions near Bragg peaks, while AB2TDS can cover all reciprocal space, but it utilizes single or two-phonon scattering approximations. In this work we propose a method to derive joint atomic displacement parameters which are used in conjunction with YELL [4] to model and fit real-space 3D-ΔPDF TDS signals efficiently. This approach avoids the need for large real-space models and offers good computational efficiency. Joint atomic displacement parameters are calculated based on the crystal's dynamical matrix, which can be derived using various methods, including universal potentials [5], DFT-based [6] approaches, or, in an approximated fashion, from the elastic constants of the material.

Our objective is to develop user-friendly software capable of describing TDS and TDS-like static disorder. Our approach offers faster computation compared to existing software and will be a valuable tool in the suite of 3D-ΔPDF software. It will facilitate the extraction of elastic constants from diverse materials and has the potential to be extended to extracting the portion of dynamical matrix responsible for high-amplitude soft phonons.

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