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Computational modelling and simulation of polycrystalline coherent inelastic neutron scattering

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ABSTRACT

Inelastic neutron scattering is an experimental technique widely used to investigate the vibrational characteristics of materials in condensed matter research. While coherent inelastic neutron scattering is typically restricted to single-crystal samples, analysis of the complex datasets obtained on polycrystalline samples remains challenging, even for the simplest of structures. However, given the common availability of high performance computing platforms, it is becoming feasible to apply computationally intensive calculation methods that sample millions of q-points to the simulation of polycrystalline models of increasing complexity, a technique referred to as poly-CINS analysis. This approach allows the interpretation of experimental results by comparison and fitting against theoretical models. This paper describes a new high-performance implementation of the Scatter code, a modelling package developed for the General Utility Lattice Program (Gulp) for heterogenous CPU-GPU computing architectures. It provides the ability to generate theoretical poly-CINS data sets from semi-empirical and *ab-initio* models. We present the computational framework behind its implementation, applying an example of a semi-empirical model for the dynamics of a large unit-cell system, namely the two (low and ambient temperature) phases of solid C60 to illustrate the methodology and its scalability characteristics.

1. Introduction

In the field of materials characterisation, inelastic neutron scattering (INS) experiments have traditionally been restricted to either incoherent scattering from polycrystals or coherent scattering (CINS) from single crystals. The limited application of CINS to polycrystals can be attributed to the complexity of the spectra generated by the superimposition of scattering intensities over all crystalline orientations and the tendency to obscure relevant information that is available from the direct measurement of dispersion curves using a Triple Axis Spectrometer or direct geometry chopper spectrometer. However, a broad class of important materials, particularly nano-materials, are only obtainable in polycrystalline form. It is therefore of interest to investigate new methods of interpreting the coherent scattering data from such samples. The complexity and significant resource demands that arise from the application of computational modelling techniques to this problem require new approaches and the support of advanced computational infrastructure.

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Materials researchers already employ neutron scattering simulations as a means of validating and refining their models [1,2]. Nevertheless, limited research has been devoted to the systematic application of computational solutions for the modelling of polycrystalline materials in current simulation packages (such as *a*CLIMAX [3,4], RASPA [5], or PHONON [6]) that require model output from *ab initio* software, such as CASTEP [7] or VASP [8]. The SCATTER code allows the generation of poly-CINS modelling data using semi-empirical potential models (as well as output from the DFT codes mentioned) via the General Utility Lattice Program (GULP) software package [9,10], a popular lattice dynamics and simulation package in the materials science community.

The methodology behind the application of this software to systems with small unit cells such as aluminium [11] has been presented elsewhere. The present work has been motivated by the need to allow the application of SCATTER to the analysis of polycrystalline samples with large unit cells, as well as enable future real-time instrumentation applications, where simulations may be conducted alongside an inelastic neutron scattering experiment and theoretical models refined in real time as the data becomes available. Typical experimental run-times for the collection of poly-CINS data on even high-flux instruments can exceed 24 hours, as excellent statistics are required to resolve one-phonon features in experimental data. New instruments and beam-lines are under development that will provide unprecedented neutron fluxes. allowing orders of magnitude more neutrons on sample in a given time period than are currently available, even at high flux beam-lines such as those found at the Spallation Neutron Source in the Oak Ridge National Laboratory in the US and the European Spallation Source in Lund, Sweden. For this reason alone, an investment in effective performance is critical to ensure that this method is extensible to the next generation of distributed heterogeneous CPU-GPU architectures entering the market over the next decade [12].

This paper presents the current state of the implementation of high-performance poly-CINS modelling in SCATTER. It explores the deployment and scalability characteristics of the SCATTER code on a range of modern heterogeneous architectures—multicore, multinode and graphics processing units (GPUs). This capability is crucially important when the computational cost associated with large unit cell models (common for many nanomaterials) is further increased by the need for fine grain sampling of reciprocal space to create theoretical data sets that are of sufficient quality and resolution to compare against results obtained experimentally from modern spectrometers. The computational resources necessary to perform modelling on this scale at moderate cost have until recently been unavailable.

2. Background

2.1. Inelastic neutron scattering from polycrystalline materials

The purpose of an INS experiment is to determine the one-phonon scattering function $S(Q, \omega)$ which provides information about the relative position and motion of each atom in a target sample. As originally derived by Van Hove [13], this scattering function can be expressed in terms of the respective one-phonon coherent and incoherent scattering functions. These scattering functions, as presented in Eqs. (1) and (2), depend only on the interactions between the nuclei and define the corresponding cross sections [14]. The coherent component, depending on the average value of the scattering amplitude, contains all the information about the relative positions and motions of every nuclear pair. The incoherent scattering contribution depends only on the motions of each atom taken in isolation. It should be further noted that these calculations do not take account other effects including–but not limited–to multi-phonon terms, multiple scattering, instrument resolution, etc.

$$S_{coh}(Q,\omega) = \frac{1}{2N}\sum_{s}\sum_{\tau}\frac{1}{\omega_s}\left|\sum_{d}\frac{\bar{b}_d}{\sqrt{M_d}}\exp(-W_d)\exp(i\mathbf{Q}\cdot\mathbf{r}_d)(\mathbf{Q}\cdot\mathbf{e}_{ds})\right|^2 \times\left\langle n_s+\frac{1}{2}\mp\frac{1}{2}\right\rangle\delta(\omega\pm\omega_s)\delta(\mathbf{Q}\mp\mathbf{q}-\tau) \tag{1}$$

$$S_{inc}(Q,\omega) = \frac{1}{2N}\sum_{d}\left\{\overline{b_d^2}-(\overline{b_d})^2\right\}\frac{1}{2M_d}\exp(-2W_d)\times\sum_{s}\frac{(\mathbf{Q}\cdot\mathbf{e}_{ds})^2}{\omega_s}\left\langle n_s+\frac{1}{2}\mp\frac{1}{2}\right\rangle\times\delta(\omega\pm\omega_s) \tag{2}$$

In Eqs. (1) and (2), $S_{coh}(Q, \omega)$ and $S_{inc}(Q, \omega)$ are the respective coherent and incoherent scattering functions in a system of d atoms, for phonon mode s, reciprocal lattice vector τ, scattering length b_d, atomic mass M_d, Debye–Waller factor W_d, momentum transfer vector $\mathbf{Q}$, atomic position r_d, polarisation vector $\mathbf{e}_{ds}$, frequency ω, and phonon wavevector $\mathbf{q}$ with neutron energy gain/loss $\left\langle n_s+\frac{1}{2}\mp\frac{1}{2}\right\rangle\delta(\omega\pm\omega_s)$. N represents the number of nuclei in the unit cell.

The energy transfer of inelastic scattering results from the neutron energy gain or loss on interaction with the lattice. For a given momentum transfer vector $\mathbf{Q}$ representing the momentum change between incident and scattered wave vectors, and a vibrational frequency of the quantised lattice vibration (or phonon) created or annihilated by the scattering event, the frequency of the phonon is directly related to the modulus of the energy transfer between the target material and the scattered neutron, as determined by momentum and energy conservation and the principle of detailed balance [14].