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Hierarchical optimization for neutron scattering problems

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ABSTRACT

We present a scalable optimization method for neutron scattering problems that determines confidence regions of simulation parameters in lattice dynamics models used to fit neutron scattering data for crystalline solids. The method uses physics-based hierarchical dimension reduction in both the computational simulation domain and the parameter space. We demonstrate for silicon that after a few iterations the method converges to parameters values (interatomic force-constants) computed with density functional theory simulations.

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1. Introduction

Neutron scattering is a powerful technique for the experimental measurement of the atomic structure and dynamics of condensed matter [3,1,2]. It has been widely used in crystallography, physics, physical chemistry, biophysics, and materials research. Neutron scattering experiments probe the sample's scattering function, $S(\mathbf{Q},\omega)$, also called dynamical structure factor [3,1,2]. In the case of nuclear scattering (neutrons can also be scattered by magnetic spins), $S(\mathbf{Q},\omega)$ contains information about the spatial arrangements of atoms inside the sample, as well as the types of motions followed by the atoms [4,5]. In most cases of crystalline matter, the atoms are arranged periodically in space with a repeated unit cell, and the atoms undergo small oscillations in time around their equilibrium positions, with a motion that can be expanded in a set of harmonic oscillators (so-called phonons). The interatomic distances are of the order of Angstroems and the vibrational frequencies for the atomic oscillations are of the order of terahertz. These collective oscillations of atoms on the crystalline lattice have a scattering function, $S(\mathbf{Q},\omega)$, whose support is confined to the so-called phonon dispersion surface in four dimensional $(\mathbf{Q},\omega)$ space, where $\mathbf{Q}$ denotes the three dimensions of momentum space and ω denotes the energy transfer between neutron and sample. What can be experimentally measured in a neutron scattering experiment is the scattered intensity which is directly proportional to the scattering function, S, convolved with the response of the experimental apparatus (resolution function, R), and detailed in the next section.

The scattered intensity convolution is computationally expensive in four dimensional $(\mathbf{Q},\omega)$ space, and mathematical research has made some relatively recent strides in convolution integration that can be adapted by neutron scientists to build fast and accurate approximations to neutron scattering functions. These advancements include analytic evaluation of Gaus-

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sian by Gaussian convolutions [6,7], fast algorithms for inverse transforms of sums of Gaussians [8], fast Gaussian transform [9,10], compressed sensing of kernels [11], and limited memory block Krylov singular value decomposition (LMSVD) [12]. There exists a strong need today to develop mathematical methods that can determine the optimal set of force-constants parameters $\{\Phi\}$, which best reproduces the neutron intensity measurements, and we denote $S_{\{\Phi\}}$ to be the scattering function related to the force-constants parameters $\{\Phi\}$. Such methods would have a broad impact maximizing scientific returns from neutron scattering experiments. However, constructing a computational efficient method to extract $\{\Phi\}$ from the neutron scattering intensity is difficult because the parameter space has a large dimension, $\text{card}\{\Phi\} \sim 50$, and because of noise in the experimental data, as well as often incomplete information about the resolution function R. The focus of this research is on scalable methods for this high dimensional parameter space.

In this paper, we consider the neutron scattering problem as an inverse optimization problem and focus on inelastic neutron scattering from phonons in harmonic crystalline materials. Under the framework of this type of scattering problem, there are several difficulties that make conventional optimization methods difficult. First of all, this is a high dimensional optimization problem, thus the computational cost is very high. Also, there are multiple local optima in the problem and global optimization algorithm is needed. In addition, the scattering intensity measurement is a large data set and comparing theoretical results with the entire intensity data is expensive. To deal with these difficulties, we develop a scalable hierarchical optimization algorithm which is composed by two parts: a physics-based model reduction method that decomposes a high dimensional optimization problem into several lower dimensional sub-optimization problems; a hierarchical searching algorithm which is a stochastic global optimization method using only a portion of the intensity measurement.

The rest of this paper is organized as follows. In the next section, we give a brief description to the neutron scattering optimization problem. In Section 3, we introduce our optimization algorithm to solve the neutron scattering problem. In Section 4, we show an example to indicate the effectiveness of our algorithm. Finally, Section 5 contains conclusions and directions for the future research.

2. Neutron scattering optimization problem

In the neutron scattering optimization problem, we aim to determine a model scattering function S which provides details about the physical properties of the studied material, given the experimental neutron intensity $I(\mathbf{Q}, \omega)$. The intensity measured $I(\mathbf{Q}, \omega)$ spans a four dimensional domain in Q_x, Q_y, Q_z, and ω, where $\mathbf{Q}$ denotes the three dimensional neutron wave vector transfer, probing spatial periodicities in the sample, and ω is the energy transfer between neutron and sample. Such 4D datasets are efficiently collected with state-of-the-art experiments on single-crystalline samples with time-of-flight spectrometers [14–16], such as the ARCS, CNCS, or HYSPEC spectrometers at the Spallation Neutron Source at Oak Ridge National Laboratory [17,18]. The experimental resolution function, R, which contains characteristics of the instrument and is independent of the sample material, can play a crucial role in detailed interpretation and understanding of the results, and must be convoluted with the scattering function.

The mathematical and computational challenges of neutron scattering center around the recovery of the scattering function S, based on an experimentally-measured intensity $I = S * R$. Since recovery of S from I is a non-linear problem, we use an inverse algorithm, minimizing the deviation between a model intensity $I_{\{\Phi\}}$ and the measured intensity I:

$$\min_{\sigma_S} \left\{ \left\| I_{\{\Phi\}} - I \right\|_{L^2}^2 \right\}, \tag{1}$$

where $\{\Phi\}$ is a set of optimizable parameters specifying the physics of the sample and σ_S is the set of scattering functions related to $\{\Phi\}$.

In this research, we focus on the scientifically important problem of using neutron scattering experiments to extract information about the microscopic dynamics of atoms in materials. In this scientific case, the parameters $\{\Phi\}$ are used to describe the spring stiffnesses (force-constants) between atoms in the sample, which control the atomic vibrations (or "phonons"). However, determining these force-constants $\{\Phi\}$ from the neutron scattering intensity is a challenging problem because of the large number of unknown force-constants, and because of the four dimensional convolution of S with the instrumental resolution R.

The model intensity $I_{\{\Phi\}}$ is obtained by building a model of the scattering function for phonons in the crystal, $S_{\{\Phi\}}$, from a set of force-constant parameters $\{\Phi\}$, and convoluting it with the instrument resolution:

$$I_{\{\Phi\}}(\mathbf{Q}, \omega) = S_{\{\Phi\}}(\mathbf{Q}, \omega) * R(\mathbf{Q}, \omega). \tag{2}$$

Thus, we solve for $S_{\{\Phi\}}$ via optimization of the force-constants $\{\Phi\}$ in the lattice dynamics model, which specifies $S_{\{\Phi\}}$. Assuming a potential energy that is quadratic in the atomic positions $\boldsymbol{r}$, we construct S from the set of interatomic force-constants, $\Phi_{i,j}$, between pairs of atoms $(\boldsymbol{r}_i, \boldsymbol{r}_j)$. Fourier transformation of these position-dependent force-constants, $\Phi_{i,j}(\boldsymbol{r}_j - \boldsymbol{r}_i)$, leads to the construction of a wavevector-dependent dynamical matrix, $\mathcal{D}$ (hermitian, positive definite). The eigenvalues of $\mathcal{D}$ are the squares of atomic vibration frequencies, ω^2, corresponding to different modes of vibrations (phonons), and the corresponding eigenvectors provide the spatial pattern of atomic vibration in each mode ("polarization vectors", $\boldsymbol{\epsilon}$) [4].