Calculation of thermal diffuse scattering

N. Wakabayashi,^{a,b} R. M. Nicklow,^c S. Katano,^b Y. Ishii,^b H. R. Child,^cH. G. Smith^c and J. A. Fernandez-Baca^c^a Department of Physics, Keio University, Yokohama 223, Japan.^b Advance Science Research Center, Japan Atomic Energy Research Institute,
Tokai, Ibaraki 319-11, Japan.^c Solid State Division, Oak Ridge National Laboratory,
Oak Ridge, TN 37831-6393, USA.

ABSTRACT

We have developed a computer program to calculate the thermal diffuse scattering (TDS) intensity distribution for single crystal specimens in a diffractometer with no energy analysis. We assumed that the phonon frequencies are approximated by those of elastic waves and that the elastic constants, density and lattice parameters of the system under study are known. The results of the calculations were compared to experimental data obtained for single crystals of Si, diamond and NiAl at the Wide Angle Neutron Diffractometer at the HFIR at Oak Ridge National Laboratory. Excellent agreement was found between the calculations and the experimental observations.

keywords: Thermal diffuse scattering, single-crystal diffraction, neutron instrumentation.

Correspondence author: Jaime A. Fernandez-Baca
Solid State Division
Oak Ridge National Laboratory
Oak Ridge, TN 37831-6393, USA
FAX: (423)576-6268
e-mail: jfn@ornl.gov

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Neutron elastic diffuse scattering is a powerful technique to study various types of static disorder in solids such as lattice distortions, precursor features to phase transformations, atomic correlations and magnetic moment distributions in alloys. Most diffractometers used in such studies do not have energy analysis capabilities and it is generally hoped that the contributions from inelastic scattering processes are small and that meaningful information about static disorder can be obtained without tedious corrections. An obvious obstacle to this approach is the strong scattering by phonons or thermal diffuse scattering (TDS), a problem especially severe in the vicinity of the Bragg reflections. In this paper we report the main features of the program WANDTDS that we developed in order to calculate the TDS intensity distribution for the Wide Angle Neutron Diffractometer (WAND)¹ at the HFIR, an instrument in which the TDS effects are appreciable.

It would be necessary to have a complete model of the lattice dynamics of the crystal under study to calculate the TDS accurately. Since this is not always possible we have simplified the problem by the use of elastic waves instead of a precise description of the phonons. This approximation should be valid only near the origin of a Brillouin zone and the calculated intensity is less accurate as the zone boundary is approached. In fact, the calculated intensity is not continuous across the boundaries, nevertheless the results show surprisingly good agreement with the measurements. The experimental set up such as the incident neutron energy, the region of the measurements in reciprocal space, and the lattice constants, density and elastic constants of the sample are inputs to the program. The region of interest is represented by wavevectors corresponding to a 100×100 -point grid in reciprocal space. For each point $\mathbf{q}$, various angles are calculated for the elastic

scattering. In addition to the elastic scattering for $\mathbf{Q}=\mathbf{G}+\mathbf{q}$ ($\mathbf{G}$ is a reciprocal lattice vector, see figure 1), inelastic scattering by phonons may occur for scattered neutron wavevectors $\mathbf{k}'$ parallel to the elastic scattering wavevector $\mathbf{k}_e$, as long as the energy and momentum conservation relations are satisfied:

$$\begin{aligned} E_0 - E' &= \frac{\hbar^2}{2m} k_0^2 - \frac{\hbar^2}{2m} k'^2 = \hbar\omega(\mathbf{q}') \\ \mathbf{k}_0 - \mathbf{k}' &= \mathbf{Q}' = \mathbf{q}' + \mathbf{G}, \end{aligned} \quad (1)$$

In these equations $\mathbf{q}'$ is the wavevector of a phonon of energy $\hbar\omega(\mathbf{q}')$, $\mathbf{k}_0$, and $\mathbf{k}'$ are the wavevectors of the incident and the inelastically scattered neutron beams, and the inelastic scattering wavevector is $\mathbf{Q}'=\mathbf{q}'+\mathbf{G}$. The phonon energies and polarization vectors are obtained by solving the eigenvalue equations for elastic waves. If the phonon energy is equal to the neutron energy change, the intensity contribution is calculated according to

$$\frac{k'}{k_0} |\mathbf{Q}' \cdot \mathbf{e}(\mathbf{q})|^2 \frac{\{n(\mathbf{q}) + \frac{1}{2} \pm \frac{1}{2}\}}{\omega(\mathbf{q})} \left\{ 1 \pm \frac{\hbar}{2E} \mathbf{k}' \cdot \nabla \omega(\mathbf{q}) \right\}^{-1} \quad (2)$$

depending on whether a phonon is created (+) or annihilated (-), $\mathbf{e}(\mathbf{q}')$ and $n(\mathbf{q}')$ are the polarization vector and the population factor of the phonon. This procedure is repeated for $E'=0$ up to some large value of energy E_m , chosen to be several times $k_B T$, and the sum of these contributions is the intensity of the inelastic processes for the nominal wavevector $\mathbf{q}$.

TDS calculations have been performed for single crystals of Si, diamond and NiAl at the

WAND and excellent agreement was found with the experimental observations. As an example figure 2 shows the actual diffraction pattern and the corresponding TDS calculation for a perfect crystal of Si. In figure 2b the results of the TDS calculation have been smeared in Q space in an attempt to simulate the instrumental resolution.

Finally we note that the inelastic contributions are extremely sensitive to incident neutron energy. In the future it may even be possible to determine the sound velocity from the TDS pattern if one can vary the incident neutron energy, this would be useful when acoustic phonon measurements cannot be made otherwise.

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References

1. The WAND is a flat-cone diffractometer designed and built by JAERI at ORNL under the US-Japan Cooperative Program on Neutron Scattering.

Figure Captions

Figure 1. Neutron scattering diagram. showing the wavevectors of the incident ($\mathbf{k}_0$), elastically scattered ($\mathbf{k}_e$) and inelastically scattered ($\mathbf{k}'$) neutron beams. $\mathbf{G}$ is a reciprocal lattice vector and $\mathbf{Q}$ and $\mathbf{q}'$ are the elastic scattering and the phonon wavevectors respectively.

Figure 2a) Two dimensional diffraction pattern of the (4 4 0) reflection of a perfect single crystal of Si at the WAND. Note that the pattern is diagonally elongated due to the instrumental resolution. The butterfly-like feature in this pattern is due to the TDS effects.

Figure 2b) Calculation of the TDS for the Bragg reflection of figure 2a.

Figure 1

FIGURE 2A

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