Determination of phonon dispersions from x-ray transmission scattering: the example of silicon

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Introduction

Phonons are the fundamental quanta of lattice vibration in a solid. They play a critical role in phenomena such as superconductivity and many types of phase transitions, and they are the basis for the acoustic, thermal, elastic, and infrared properties of solids [1]. This report demonstrates a new approach to phonon studies based on x-ray intensity patterns produced by scattering from thermally populated phonons using silicon as a test case. A least-squares analysis of these patterns in terms of lattice dynamics yields dispersions for all six phonon branches in excellent agreement with neutron scattering results. The fast data acquisition rate, simplicity of the experiment, and minimal requirement of sample volume make this method attractive for a wide range of applications in materials research.

Methods and Materials

Our experiment was performed at the undulator beamline of sector 33 UNI-CAT (University, Industry, and National Laboratory Collaborative Access Team) at the Advanced Photon Source (APS). A transmission Laue geometry was employed, in which a 28 keV beam was sent at normal incidence through commercial Si wafers with a thickness of 0.5 mm. An image plate positioned behind the sample was used to record the images with an exposure time of ~10 s each. The data shown below were taken with the sample in air.

Results

Figures 1(a) and 1(b) are experimental pictures of Si(111) and Si(100), respectively. Figures 1(c) and 1(d) are the corresponding model calculations to be discussed below. By virtue of the wavelength selected, the Bragg condition is never satisfied over the entire area of detection. Thus, none of the bright spots are caused by crystal diffraction. One can readily see the symmetry of the pattern. The picture is three-fold symmetric for the (111) sample, and four-fold symmetric for the (100) sample.

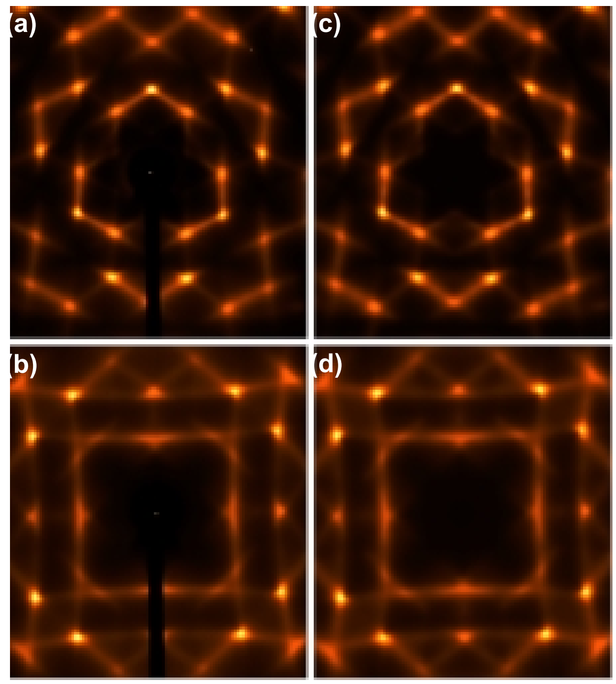

Figure 1: (a) and (b) are experimental pictures of Si(111) and Si(100), respectively; (c) and (d) are corresponding model calculation.

The theoretical pictures and phonon dispersions are derived from a force-constant formalism of the lattice dynamics, also known as the Born-von Karman model [2–3]. The intensity of scattering by an unpolarized incident x-ray at a given momentum transfer $\boldsymbol{q}$ is given by a sum over the contributions from the six phonon branches [4]:

$$I_0 \propto \sum_{j=1}^{6} \frac{\left| \sum_{s=1}^{2} f \cdot e^{-M} \cdot \mathbf{q} \cdot \hat{\mathbf{e}}_{j,s}(\mathbf{q}) \right|^2}{\omega_j} \coth\left(\frac{\hbar\omega_j}{2k_B T} \right) \qquad (1)$$

In equation (1), for a silicon atom s in the unit cell, *f* is the atomic scattering factor, *M* is the Debye-Wa.ller factor [5], ω is the phonon frequency, $\hat{\mathbf{e}}_{j,s}$ is the polarization vector of the phonon mode j for basis atom s, k_B is the Boltzmann constant, *T* is the sample temperature (300 K), and *j* is the index for the six phonon branches.

The calculated intensity in each pixel, on a logarithmic scale, is given by equation (2):

$$I_{theory} = D \bullet \log \{[\sin^2\phi + \cos^2\phi \bullet \cos^2(2\theta)](A \bullet I_0 + B)\cos(2\theta) + C\} \qquad (2)$$

In this equation, ϕ is the azimuthal angle between the plane of polarization of the incident beam and the scattering plane, and 2θ is the scattering angle. The expression within the brackets containing these angles accounts for the linear polarization of the incident beam. The quantity *A* is an intensity factor, *B* represents a background from higher order and defect scattering from the sample, *C* represents a uniform background, and *D* is an overall scaling factor related to the image plate response function.

Equations (1) and (2) are used to generate theoretical patterns, which are compared to the experimental ones. The shadows of the beam stop and post are excluded from this comparison by using a mask function. A least-squares algorithm is employed for a simultaneous pixel-by-pixel fit to both experimental pictures shown in Figure 1. The best fits are shown in Figure 1, and the corresponding phonon dispersion curves are shown as solid curves in Figure 2. The dotted curves in Figure 2 can be regarded as a *k*-space interpolation of the neutron data. The agreement with available neutron data [6–7], presented in Figure 2 as circles, is excellent, thus validating the present method as a technique for determination of phonon dispersions.

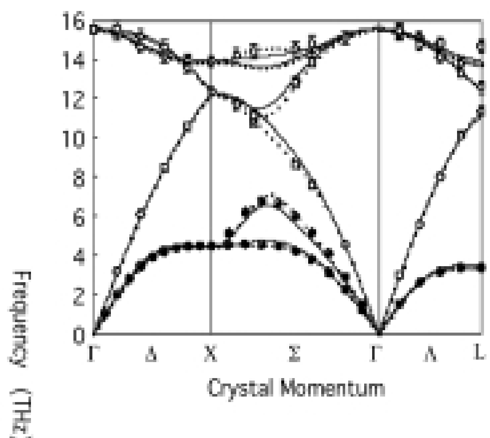

Figure 2: Phonon dispersion curves.

Discussion

Many areas of experimental condensed matter physics would benefit from an alternative to neutron scattering or inelastic x-ray scattering to establish phonon properties. This work establishes x-ray phonon scattering patterns as a powerful technique for measuring phonon properties. Entire sets of dispersion curves can be derived based on data taken in seconds. This is demonstrated using Si as a test case; a simultaneous fit of the (100) and (111) patterns yields phonon dispersion curves as accurate as neutron scattering results.

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