

Comment on “Determination of Phonon Dispersions from X-Ray Transmission Scattering: The Example of Silicon”

In a recent Letter [1], Holt *et al.* showed that x-ray thermal diffuse scattering measurements using synchrotron radiation had high enough resolution for realistic studies of lattice dynamics in solids. As an example, the force constants and phonon dispersions for crystalline silicon were determined by a least-squares fit to the measured transmission scattering intensity. It was nicely demonstrated that x-ray thermal diffuse scattering had finally become a convenient, viable tool in investigating lattice vibrations.

Central to the analysis in the Letter is an expression connecting the scattering intensity and the phonon characteristics, truncated at the first order (one-phonon scattering). It is the purpose of this Comment to point out that the formula used in the original Letter, Eq. (1) in [1], is not completely correct when the unit cell contains more than one atom, as in the diamond structure. The correct expression should contain a phase factor arising from the basis, similar to that in the structure factor.

The scattering intensity from a sample is given by

$$I(\vec{q}) = \left| \sum_m f(\vec{q}) \exp(-i\vec{q} \cdot \vec{r}_m) \right|^2, \quad (1)$$

where \vec{q} is the scattering vector, $f(\vec{q})$ the atomic scattering factor, and \vec{r}_m the atomic positions. The Bragg peaks are obtained if equilibrium positions are used, while thermal vibrations give rise to an additional diffuse intensity, the magnitude of which is related to the properties of the collective excitations, i.e., phonons in the crystal. Define the dynamical matrix in the standard way as

$$D_{nn'}^{\alpha\beta}(\vec{k}) = \frac{1}{\sqrt{\mu_n \mu_{n'}}} \sum_{\vec{R}} D_{nn'}^{\alpha\beta}(\vec{R}) \exp(-i\vec{k} \cdot \vec{R}), \quad (2)$$

where α and β are Cartesian indices, n and n' are atomic indices in a basis, \vec{R} is a lattice vector, \vec{k} is the phonon wave vector, μ is the atomic mass, and $D_{nn'}^{\alpha\beta}(\vec{R})$ is the second derivative of energy with respect to the α th component of the displacement of atom n in the unit cell at \vec{R} and the β th component of the displacement of atom n' in the unit cell at origin. The dynamical matrix thus defined is periodic in the momentum space, and one can restrict \vec{k} to be in the first Brillouin zone. If there are s atoms per unit cell, diagonalizing the $3s \times 3s$ dynamical matrix yields the phonon frequency $\omega_j(\vec{k})$ and polarization vector $\vec{e}_{n,j}(\vec{k})$ of atom n for mode $j = 1, \dots, 3s$.

By taking the thermal average of Eq. (1), one obtains the first-order diffuse intensity for an infinite crystal as

$$I_{\text{diff}}(\vec{q}) \propto \sum_{j=1}^{3s} \frac{1}{\omega_j} \coth\left(\frac{\hbar\omega_j(\vec{k})}{2k_B T}\right) |F_j(\vec{q})|^2, \quad (3)$$

where

$$F_j(\vec{q}) = \sum_{n=1}^s f_n(\vec{q}) \exp[-M_n(\vec{q}) - i\vec{q} \cdot \vec{\tau}_n] \frac{\vec{q} \cdot \vec{e}_{n,j}(\vec{k})}{\sqrt{\mu_n}}. \quad (4)$$

In Eq. (4), M_n is the Debye-Waller factor, $\vec{\tau}_n$ is the position vector for atom n in the basis, and \vec{q} and \vec{k} are related by $\vec{q} = \vec{k} + \vec{G}$, where \vec{G} is a reciprocal lattice vector. Equation (4) is similar to the structure factor term in the Bragg intensity, but with an extra inner product $\vec{q} \cdot \vec{e}_{n,j}(\vec{k})$. Compared with Eq. (1) in [1], the current expression has an additional phase factor $\exp(-i\vec{q} \cdot \vec{\tau}_n)$ and includes the polarization vector $\vec{e}_{n,j}(\vec{k})$ for all atoms n . Hence it is puzzling that an excellent fit was found in [1], which might be attributed to the large number of fitting parameters.

The general x-ray diffuse scattering intensity has been discussed in the literature [2,3]. Our expression above is consistent with these previous results, but is cast in a different form. The result in [2] was represented in terms of the “scattering matrix” which is a function of the dynamical matrix, while a different definition of the dynamical matrix was used in [3].

It is interesting to note that the intensity expression in Eqs. (3) and (4) contain a projection $\vec{q} \cdot \vec{e}_{n,j}(\vec{k})$ and that many scattering vectors \vec{q} can probe the same phonons at the reduced \vec{k} . For high symmetry directions in a crystal, for example, the [100] and [111] directions in the diamond structure, the phonons are purely longitudinal or transverse. Therefore, by properly choosing the scattering vectors in different Brillouin zones with different projection combinations, one will be able to obtain the phonon frequencies directly along these directions. These phonon frequencies can then determine the experimental planar force constants, and provide information on the three-dimensional force constants [4].

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