<table>
<thead>
<tr>
<th><strong>Title</strong></th>
<th>Directly observing squeezed phonon states with femtosecond x-ray diffraction</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Authors</strong></td>
<td>Johnson, Steve L.; Beaud, Paul; Vorobeva, Ekaterina; Milne, Christopher J.; Murray, Éamonn D.; Fahy, Stephen B.; Ingold, Gerhard</td>
</tr>
<tr>
<td><strong>Publication date</strong></td>
<td>2009</td>
</tr>
<tr>
<td><strong>Type of publication</strong></td>
<td>Article (peer-reviewed)</td>
</tr>
<tr>
<td><strong>Link to publisher’s version</strong></td>
<td><a href="https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.102.175503">https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.102.175503</a> - 10.1103/PhysRevLett.102.175503</td>
</tr>
<tr>
<td><strong>Rights</strong></td>
<td>© 2009, American Physical Society</td>
</tr>
<tr>
<td><strong>Download date</strong></td>
<td>2024-05-08 15:20:06</td>
</tr>
<tr>
<td><strong>Item downloaded from</strong></td>
<td><a href="https://hdl.handle.net/10468/4642">https://hdl.handle.net/10468/4642</a></td>
</tr>
</tbody>
</table>
Abstract

This document serves as a supplement to the article “Directly observing squeezed phonon number states with femtosecond x-ray diffraction” [1]. It contains additional details regarding the experimental technique and some details of the mathematical analysis of the data.
EXPERIMENTAL DETAILS

The sample under investigation is a single crystal of bismuth, cut at an angle of 54° from the (111) lattice planes toward the [211] direction. The sample was kept in a vacuum environment for all measurements, and a closed-loop He cryostat controlled the temperature for the 170 K measurement.

The data were collected in a pump-probe scheme, measuring alternately “pumped” and “unpumped” diffracted intensities to obtain the normalized diffraction change as a function of pump-probe delay time, and averaged over multiple scans. The optical pump pulses (115 fs, 800 nm, 1 kHz) hit the surface of the crystal at a grazing incidence of 10° with π-polarization. The absorbed fluence was $1.37 \pm 0.14 \text{ mJ/cm}^2$.

The femtosecond probe x-rays were produced using the electron beam slicing facility at the Swiss Light Source to generate $140 \pm 30 \text{ fs}$ duration x-ray pulses at a repetition rate of 2 kHz, synchronized to the optical pump pulses. Two grazing incidence mirrors focused the beam onto the sample, producing a beam size of 7 μm vertically and 250 μm horizontally at the position of the crystal, but with a grazing incidence angle of 0.55° with σ-polarization. As described in an earlier work [2], this small incidence angle sets the effective probe depth of the x-rays to 50 nm due to photoabsorption. Diffraction from a single multilayer mirror (Mo/B$_4$C, 25 Å period, $\gamma = 0.5$) placed just before the sample set the energy of the x-rays to 7.15 keV with a bandwidth of 1.3%. 
RECURSION RELATIONS

To solve

$$\langle (\hat{u}_j \cdot G)^2 \rangle = \sum_{k,s'} C_{k,s'} \left| \sum_s \frac{e^{j}(t) \cdot G}{\sqrt{\omega_{ks}(t)}} \left[ U_{kss'}(t) + V_{kss'}(t)^* \right] \right|^2$$  \hspace{1cm} (1)$$

for arbitrary time-dependent phonon eigenvectors $e^{j}_{ks}(t)$ and frequencies $\omega_{ks}$, we employ a recursion relation solution method modeled after the work of Kiss et al. [3] Under this scheme, we consider the time-dependence of the frequencies and eigenvectors as a series of closely spaced step-functions:

$$\omega_{ks}(t) = \begin{cases} \omega_{(0)}^{ks} & t_0 < t < t_1 \\ \omega_{(1)}^{ks} & t_1 < t < t_2 \\ \omega_{(2)}^{ks} & t_2 < t < t_3 \\ \vdots \\ \omega_{(n)}^{ks} & t_n < t < t_{n+1} \\ \vdots \end{cases}$$  \hspace{1cm} (2)$$

$$e^{j}_{ks}(t) = \begin{cases} e^{j(0)}_{ks} & t_0 < t < t_1 \\ e^{j(1)}_{ks} & t_1 < t < t_2 \\ e^{j(2)}_{ks} & t_2 < t < t_3 \\ \vdots \\ e^{j(n)}_{ks} & t_n < t < t_{n+1} \\ \vdots \end{cases}$$  \hspace{1cm} (3)$$

We then solve for the quantities $U_{kss'}(t)$ and $V_{kss'}(t)$ in equation (4) in ref. [1] over each interval.

Between steps, the time evolution of the phonon annihilation and creation operators is that of a collection of simple harmonic oscillators with constant frequencies. Let $\hat{a}_{ks}^{(n)}$ be $\hat{a}_{ks}$ at a time just after $t_n$. We may then write $a_{ks}(t) = \hat{a}_{ks}^{(n)} e^{-i\omega_{ks}^{(n)}(t-t_n)}$ for $t_n < t < t_{n+1}$. Thus

$$U_{kss'}(t) = U_{kss'}^{(n)} e^{-i\omega_{ks}^{(n)}(t-t_n)}$$

$$V_{kss'}(t) = V_{kss'}^{(n)} e^{-i\omega_{ks}^{(n)}(t-t_n)}$$  \hspace{1cm} (4)$$
where we have used the condition \( \omega_{-ks} = \omega_{ks} \).

To find \( U_{kss'}^{(n+1)} \) and \( V_{kss'}^{(n+1)} \) in terms of \( U_{kss'}^{(n)} \) and \( V_{kss'}^{(n)} \) we require that at any lattice site \( \mathbf{R} \), the atomic displacement

\[
\hat{u}_j(\mathbf{R}) = \frac{1}{\sqrt{N}} \sum_{k,s} \sqrt{\frac{\hbar}{2\omega_{ks}(t)}} (\hat{a}_{ks} + \hat{a}_{-ks} \dagger) \epsilon_k^j(t) e^{i\mathbf{k}\cdot\mathbf{R}}
\]

(5)

and momentum

\[
\hat{p}_j(\mathbf{R}) = -\frac{i}{\sqrt{N}} \sum_{k,s} M_j \sqrt{\frac{\hbar\omega_{ks}(t)}{2}} (\hat{a}_{ks} - \hat{a}_{-ks} \dagger) \epsilon_k^j(t) e^{i\mathbf{k}\cdot\mathbf{R}}
\]

(6)

be continuous at \( t_{n+1} \). Using the relation \( \epsilon_{-ks}^j = (\epsilon_{ks}^j)^\ast \) and the eigenvector orthonomality condition \( \sum_j M_j (\epsilon_k^j)^\ast \cdot \epsilon_{ks'}^j = \delta_{ss'} \) we obtain the recursion relations

\[
U_{kss'}^{(n+1)} = \frac{1}{2} \sum_{j,s''} M_j \left[ (\epsilon_{ks}^j)^\ast \cdot \epsilon_{ks''}^j \right] \left[ A_{kss''}^{(n)} U_{kss''}^{(n)} + B_{kss''}^{(n)} V_{kss''}^{(n)} \right] \tag{7}
\]

\[
V_{kss'}^{(n+1)} = \frac{1}{2} \sum_{j,s''} M_j \left[ \epsilon_{ks}^j \cdot (\epsilon_{ks''}^j)^\ast \right] \left[ A_{kss''}^{(n)} V_{kss''}^{(n)} + B_{kss''}^{(n)} (U_{kss''}^{(n)})^\ast \right] \tag{8}
\]

\[
A_{kss''}^{(n)} = \frac{\omega_{ks}^{(n+1)} + \omega_{ks''}^{(n+1)}}{\omega_{ks}^{(n+1)}} e^{-i\omega_{ks''}(t_{n+1}-t_n)}
\]

(9)

\[
B_{kss''}^{(n)} = \frac{\omega_{ks}^{(n+1)} - \omega_{ks''}^{(n+1)}}{\omega_{ks}^{(n+1)}} e^{-i\omega_{ks''}(t_{n+1}-t_n)}
\]

(10)

If the crystal is in thermal equilibrium at \( t = t_0 \), the initial conditions are \( U_{kss'}^{(0)} = \delta_{ss'} \) and \( V_{kss'}^{(0)} = 0 \). These, in combination with equations 7 and 8, allow us to solve equation 1 for the variance in atomic position at any time \( t \).

* steve.johnson@aps.org

