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EPAPS for “Directly observing squeezed phonon number states with femtosecond x-ray diffraction”

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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 ± 0.14 mJ/cm².

The femtosecond probe x-rays were produced using the electron beam slicing facility at the Swiss Light Source to generate 140 ± 30 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/B₄C, 25 Å period, γ = 0.5) placed just before the sample set the energy of the x-rays to 7.15 keV with a bandwidth of 1.3%.
To solve
\[ \langle (\hat{u}_j \cdot \mathbf{G})^2 \rangle = \sum_{k,s'} C_{ks'} \left| \sum_s \frac{\epsilon_{ks}(t) \cdot \mathbf{G}}{\sqrt{\omega_{ks}(t)}} \right|^2 (U_{kss'}(t) + V_{kss'}(t)^*) \] (1)
for arbitrary time-dependent phonon eigenvectors \( \epsilon_{ks}(t) \) 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:

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

\[ \epsilon_{ks}(t) = \begin{cases} 
\epsilon_{ks}^{(0)} & t_0 < t < t_1 \\
\epsilon_{ks}^{(1)} & t_1 < t < t_2 \\
\epsilon_{ks}^{(2)} & t_2 < t < t_3 \\
\vdots & \vdots \\
\epsilon_{ks}^{(n)} & t_n < t < t_{n+1} \\
\vdots & \vdots 
\end{cases} 
\] (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

\[ U_{kss'}(t) = U_{kss'}^{(n)} e^{-i\omega_{ks}^{(n)}(t-t_n)} \] for \( t_n < t < t_{n+1} \) (4)

\[ V_{kss'}(t) = V_{kss'}^{(n)} e^{-i\omega_{ks}^{(n)}(t-t_n)} \]
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 $R$, the atomic displacement

$$\hat{u}_j(R) = \frac{1}{\sqrt{N}} \sum_{k,s} \sqrt{\frac{\hbar}{2\omega_{ks}(t)}} (\hat{a}_{ks} + \hat{a}_{-ks}^\dagger) \epsilon_k(t) e^{ikR}$$  \hspace{1cm} (5)$$

and momentum

$$\hat{p}_j(R) = -\frac{i}{\sqrt{N}} \sum_{k,s} M_j \sqrt{\frac{\hbar \omega_{ks}(t)}{2}} (\hat{a}_{ks} - \hat{a}_{-ks}^\dagger) \epsilon_k(t) e^{ikR}$$  \hspace{1cm} (6)$$

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

$$U_{kss'}^{(n+1)} = \frac{1}{2} \sum_{j,j''} M_j \left[ (\epsilon_k^j)^* \epsilon_k^{j''} \right] \left[ A_{kss''}^{(n)} U_{kss'''}^{(n)} + B_{kss''}^{(n)} (V_{kss'''}^{(n)})^* \right]$$  \hspace{1cm} (7)$$

$$V_{kss'}^{(n+1)} = \frac{1}{2} \sum_{j,j''} M_j \left[ \epsilon_k^j \cdot (\epsilon_k^{j''})^* \right] \left[ A_{kss''}^{(n)} V_{kss'''}^{(n)} + B_{kss''}^{(n)} (U_{kss'''}^{(n)})^* \right]$$  \hspace{1cm} (8)$$

$$A_{kss''}^{(n)} = \frac{\omega_{kss'}^{(n+1)} + \omega_{kss''}^{(n)}}{\omega_{kss'}^{(n+1)} - \omega_{kss''}^{(n)}} e^{-i \omega_{kss'}^{(n)} (t_{n+1} - t_n)}$$  \hspace{1cm} (9)$$

$$B_{kss''}^{(n)} = \frac{\omega_{kss'}^{(n+1)} - \omega_{kss''}^{(n)}}{\omega_{kss'}^{(n+1)} - \omega_{kss''}^{(n)}} e^{-i \omega_{kss''}^{(n)} (t_{n+1} - t_n)}$$  \hspace{1cm} (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 $t$. 

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