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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%.
RECURSION RELATIONS

To solve
\[
\left\langle \left( \hat{u}_j \cdot \mathbf{G} \right)^2 \right\rangle = \sum_{k, s'} C_{ks'} \left| \sum_s \hat{e}^j_s(t) \cdot \frac{\mathbf{G}}{\sqrt{\omega_{ks}(t)}} \left[ U_{kss'}(t) + V_{kss'}(t)^* \right] \right|^2
\]
(1)
for arbitrary time-dependent phonon eigenvectors \( \hat{e}^j_{ks}(t) \) 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:

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

\[
\hat{e}^j_{ks}(t) = \begin{cases} 
\hat{e}^{j(0)}_{ks} & t_0 < t < t_1 \\
\hat{e}^{j(1)}_{ks} & t_1 < t < t_2 \\
\hat{e}^{j(2)}_{ks} & t_2 < t < t_3 \\
\vdots \\
\hat{e}^{j(n)}_{ks} & t_n < t < t_{n+1} \\
\vdots 
\end{cases}
\]
(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 \( \hat{a}^{(n)}_{ks} \) be \( \hat{a}_{ks} \) at a time just after \( t_n \). We may then write \( a_{ks}(t) = \hat{a}^{(n)}_{ks} e^{-i\omega^{(n)}_{ks}(t-t_n)} \) for \( t_n < t < t_{n+1} \). Thus

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

\[
V_{kss'}(t) = V^{(n)}_{kss'} e^{-i\omega^{(n)}_{ks}(t-t_n)} \quad \text{for} \quad t_n < t < t_{n+1}
\]
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

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

and momentum

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

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

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

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

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

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

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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