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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 $[\bar{2}\bar{1}\bar{1}]$ 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 $7 \mu\text{m}$ vertically and $250 \mu\text{m}$ 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, $\gamma = 0.5$) 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

$$\langle (\hat{\mathbf{u}}_j \cdot \mathbf{G})^2 \rangle = \sum_{\mathbf{k}, s'} C_{\mathbf{k}s'} \left| \sum_s \frac{\boldsymbol{\epsilon}_{\mathbf{k}s}^j(t) \cdot \mathbf{G}}{\sqrt{\omega_{\mathbf{k}s}(t)}} [U_{\mathbf{k}ss'}(t) + V_{\mathbf{k}ss'}(t)^*] \right|^2 \quad (1)$$

for arbitrary time-dependent phonon eigenvectors $\boldsymbol{\epsilon}_{\mathbf{k}s}^j(t)$ and frequencies $\omega_{\mathbf{k}s}$, 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:

$$\omega_{\mathbf{k}s}(t) = \begin{cases} \omega_{\mathbf{k}s}^{(0)} & t_0 < t < t_1 \\ \omega_{\mathbf{k}s}^{(1)} & t_1 < t < t_2 \\ \omega_{\mathbf{k}s}^{(2)} & t_2 < t < t_3 \\ \dots & \\ \omega_{\mathbf{k}s}^{(n)} & t_n < t < t_{n+1} \\ \dots & \end{cases} \quad (2)$$

$$\boldsymbol{\epsilon}_{\mathbf{k}s}^j(t) = \begin{cases} \boldsymbol{\epsilon}_{\mathbf{k}s}^{j(0)} & t_0 < t < t_1 \\ \boldsymbol{\epsilon}_{\mathbf{k}s}^{j(1)} & t_1 < t < t_2 \\ \boldsymbol{\epsilon}_{\mathbf{k}s}^{j(2)} & t_2 < t < t_3 \\ \dots & \\ \boldsymbol{\epsilon}_{\mathbf{k}s}^{j(n)} & t_n < t < t_{n+1} \\ \dots & \end{cases} \quad (3)$$

We then solve for the quantities $U_{\mathbf{k}ss'}(t)$ and $V_{\mathbf{k}ss'}(t)$ 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 $\hat{a}_{\mathbf{k}s}^{(n)}$ be $\hat{a}_{\mathbf{k}s}$ at a time just after t_n . We may then write $a_{\mathbf{k}s}(t) = \hat{a}_{\mathbf{k}s}^{(n)} e^{-i\omega_{\mathbf{k}s}^{(n)}(t-t_n)}$ for $t_n < t < t_{n+1}$. Thus

$$\begin{aligned} U_{\mathbf{k}ss'}(t) &= U_{\mathbf{k}ss'}^{(n)} e^{-i\omega_{\mathbf{k}s}^{(n)}(t-t_n)} \\ V_{\mathbf{k}ss'}(t) &= V_{\mathbf{k}ss'}^{(n)} e^{-i\omega_{\mathbf{k}s}^{(n)}(t-t_n)} \end{aligned} \quad \text{for } t_n < t < t_{n+1} \quad (4)$$

where we have used the condition $\omega_{-\mathbf{k}s} = \omega_{\mathbf{k}s}$.

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

$$\hat{\mathbf{u}}_j(\mathbf{R}) = \frac{1}{\sqrt{N}} \sum_{\mathbf{k},s} \sqrt{\frac{\hbar}{2\omega_{\mathbf{k}s}(t)}} (\hat{a}_{\mathbf{k}s} + \hat{a}_{-\mathbf{k}s}^\dagger) \boldsymbol{\epsilon}_{\mathbf{k}s}^j(t) e^{i\mathbf{k}\cdot\mathbf{R}} \quad (5)$$

and momentum

$$\hat{\mathbf{p}}_j(\mathbf{R}) = \frac{-i}{\sqrt{N}} \sum_{\mathbf{k},s} M_j \sqrt{\frac{\hbar\omega_{\mathbf{k}s}(t)}{2}} (\hat{a}_{\mathbf{k}s} - \hat{a}_{-\mathbf{k}s}^\dagger) \boldsymbol{\epsilon}_{\mathbf{k}s}^j(t) e^{i\mathbf{k}\cdot\mathbf{R}} \quad (6)$$

be continuous at t_{n+1} . Using the relation $\boldsymbol{\epsilon}_{-\mathbf{k}s}^j = (\boldsymbol{\epsilon}_{\mathbf{k}s}^j)^*$ and the eigenvector orthonormality condition $\sum_j M_j (\boldsymbol{\epsilon}_{\mathbf{k}s}^j)^* \cdot \boldsymbol{\epsilon}_{\mathbf{k}s'}^j = \delta_{ss'}$ we obtain the recursion relations

$$U_{\mathbf{k}ss'}^{(n+1)} = \frac{1}{2} \sum_{j,s''} M_j \left[(\boldsymbol{\epsilon}_{\mathbf{k}s}^{j(n+1)})^* \cdot \boldsymbol{\epsilon}_{\mathbf{k}s''}^{j(n)} \right] \left[A_{\mathbf{k}ss''}^{(n)} U_{\mathbf{k}s''s'}^{(n)} + B_{\mathbf{k}ss''}^{(n)} (V_{\mathbf{k}s''s'}^{(n)})^* \right] \quad (7)$$

$$V_{\mathbf{k}ss'}^{(n+1)} = \frac{1}{2} \sum_{j,s''} M_j \left[\boldsymbol{\epsilon}_{\mathbf{k}s}^{j(n+1)} \cdot (\boldsymbol{\epsilon}_{\mathbf{k}s''}^{j(n)})^* \right] \left[A_{\mathbf{k}ss''}^{(n)} V_{\mathbf{k}s''s'}^{(n)} + B_{\mathbf{k}ss''}^{(n)} (U_{\mathbf{k}s''s'}^{(n)})^* \right] \quad (8)$$

$$A_{\mathbf{k}ss''}^{(n)} = \frac{\omega_{\mathbf{k}s}^{(n+1)} + \omega_{\mathbf{k}s''}^{(n)}}{\sqrt{\omega_{\mathbf{k}s}^{(n+1)} \omega_{\mathbf{k}s''}^{(n)}}} e^{-i\omega_{\mathbf{k}s''}^{(n)}(t_{n+1}-t_n)} \quad (9)$$

$$B_{\mathbf{k}ss''}^{(n)} = \frac{\omega_{\mathbf{k}s}^{(n+1)} - \omega_{\mathbf{k}s''}^{(n)}}{\sqrt{\omega_{\mathbf{k}s}^{(n+1)} \omega_{\mathbf{k}s''}^{(n)}}} e^{i\omega_{\mathbf{k}s''}^{(n)}(t_{n+1}-t_n)} \quad (10)$$

If the crystal is in thermal equilibrium at $t = t_0$, the initial conditions are $U_{\mathbf{k}ss'}^{(0)} = \delta_{ss'}$ and $V_{\mathbf{k}ss'}^{(0)} = 0$. 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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