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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 $[2\bar{1}\bar{1}]$ 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 $1.37 \pm 0.14\,\mathrm{mJ/cm^2}$.

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/B_4C, 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

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

for arbitrary time-dependent phonon eigenvectors $\boldsymbol{\epsilon}^j_{\mathbf{k}s}(t)$ 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:

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

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

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 $\hat{a}^{(n)}_{\mathbf{k}s}$ be $\hat{a}_{\mathbf{k}s}$ at a time just after t_n. We may then write $a_{\mathbf{k}s}(t) = \hat{a}^{(n)}_{\mathbf{k}s} e^{-i\omega^{(n)}_{\mathbf{k}s}(t-t_n)}$ for $t_n < t < t_{n+1}$. Thus

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

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

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

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

and momentum

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

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

$$U^{(n+1)}_{\mathbf{k}ss'} = \frac{1}{2} \sum_{j,s''} M_j \left[\left(\boldsymbol{\epsilon}^{j(n+1)}_{\mathbf{k}s}\right)^* \cdot \boldsymbol{\epsilon}^{j(n)}_{\mathbf{k}s''} \right] \left[A^{(n)}_{\mathbf{k}ss''} U^{(n)}_{\mathbf{k}s''s'} + B^{(n)}_{\mathbf{k}ss''} \left(V^{(n)}_{\mathbf{k}s''s'} \right)^* \right] \tag{7}$$

$$V^{(n+1)}_{\mathbf{k}ss'} = \frac{1}{2} \sum_{j,s''} M_j \left[\boldsymbol{\epsilon}^{j(n+1)}_{\mathbf{k}s} \cdot \left(\boldsymbol{\epsilon}^{j(n)}_{\mathbf{k}s''}\right)^* \right] \left[A^{(n)}_{\mathbf{k}ss''} V^{(n)}_{\mathbf{k}s''s'} + B^{(n)}_{\mathbf{k}ss''} \left(U^{(n)}_{\mathbf{k}s''s'} \right)^* \right] \tag{8}$$

$$A^{(n)}_{\mathbf{k}ss''} = \frac{\omega^{(n+1)}_{\mathbf{k}s} + \omega^{(n)}_{\mathbf{k}s''}}{\sqrt{\omega^{(n+1)}_{\mathbf{k}s}\omega^{(n)}_{\mathbf{k}s''}}} e^{-i\omega^{(n)}_{\mathbf{k}s''}(t_{n+1}-t_n)} \tag{9}$$

$$B^{(n)}_{\mathbf{k}ss''} = \frac{\omega^{(n+1)}_{\mathbf{k}s} - \omega^{(n)}_{\mathbf{k}s''}}{\sqrt{\omega^{(n+1)}_{\mathbf{k}s}\omega^{(n)}_{\mathbf{k}s''}}} e^{i\omega^{(n)}_{\mathbf{k}s''}(t_{n+1}-t_n)} \tag{10}$$

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