**Multi-Modal Visualization of the Optomechanical Response of Silicon Cantilevers with Ultrafast Electron Microscopy**

*Supplementary Material*

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**Data Analysis Methods**

**SAED Analysis**

The SAED patterns were loaded into analysis software, and a mask was created to remove the *000* spot. For each pattern, a 2D peak-finding method that employed sub-pixel, centroid-based fitting was used. The input parameters were baseline shape and threshold intensity value, the latter of which was chosen to include all Bragg reflections present in the image while minimizing false positives. Once each peak was fit with a 2D Gaussian function, the position, width, and area were determined for each time point.

**CBED Analysis**

In order to determine localized specimen mechanical oscillation, translation of Bragg intensity across CBED discs as a function of time delay was followed. Each CBED pattern was rotated such that the reciprocal lattice vector of the *026* spot was aligned vertically in the image. Then, a box is drawn vertically from one side of the main beam, along the reciprocal lattice vector, and past the *026* CBED disc. Each row of the box was summed, and a 3D representation (reciprocal space, intensity, time) of the temporal response was generated. In addition, the overall intensity of each box was normalized by the intensity of the center spot to correct for any photoelectron-intensity instabilities. The images were acquired in randomized time order to account for any real-time specimen motion.

**BF Image-Series Analysis: Space-Time Plot**

As the space-time plot is generated by averaging one of the spatial dimensions, the signal-to-noise ratio is improved most when the averaging axis is perpendicular to the movement of the contrast, and averaged along a direction of homogenous contrast. The image set was first corrected for specimen drift using a template-matching method. Each image was then normalized for fluctuations in photoelectron intensity. This was done by dividing each image by an average pixel intensity over a 10 x 10 vacuum region. A rectangular region of interest (ROI) was chosen such that diffraction-contrast feature movement was along the long axis of the rectangle. This involved rotation of each image in the set to determine the angle at which the highest signal-to-noise ratio of the dynamics was captured. The rectangular ROI matrix was then extracted from each image in the set, averaged along the short axis, and the resulting vectors were plotted as a function of delay for the entire image set. To generate the space-frequency plot, the time-dependent Fourier transform of the space-time plot was generated, producing a Fourier spectrum at each spatial position.

**BF Image Analysis: Frequency Maps**

Each BF image was drift-corrected using features along the specimen edge to ensure an accurate pixel-by-pixel analysis. To normalize the image intensity, every pixel in the image was divided by an average value obtained from a vacuum region of the image. This step was taken in order to correct for any fluctuations based upon real-time instabilities in photoelectron intensity. Following this, a Fourier transform of the time-dependent intensity value of each pixel was performed. Each pixel in each frame of the frequency map is the absolute value of the Fourier spectrum for that pixel at the corresponding frequency, thus yielding the oscillatory contrast response of the specimen at that location. In addition, a black outline was added to the specimen edge in post-processing to distinguish the specimen from vacuum.

**Modeling Details**

The oscillatory response of contrast features observed with all three UEM modalities rules out methodological artifacts, pointing to a coherent specimen response. In order to understand this response, a linear-elastic model of a Si wedge was created using bulk material parameters and the geometry and measurements determined with optical and electron microscopy (see Figure S3). An elastic modulus of 170 GPa was used for the <110> direction, with a density of 2329 kg/m3.

**Supplementary Figures**

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FIG. S1. UEM SAED Bragg-spot-intensity behavior across multiple experiments and timescales. (a) Temporal behavior of the and Bragg spots over the delay range of -200 to 2000 ns. Time points were obtained at 2-ns increments and acquired in random order to account for photoelectron-intensity fluctuations due to laser/tip instabilities over the course of the experiment. The inset shows a representative UEM SAED pattern (camera length = 200 mm) at -32 ns highlighting the and peaks. The intensities of the two reflections have been offset for clarity. (b) Temporal behavior of the same two Bragg peaks over a delay range of -200 to 7000 ns at 2-ns increments. Due to the number of patterns obtained during the experiment (3600), the data was acquired in delay order (*i.e.*, -200 ns, -198 ns, -196 ns, *etc.*) to capture the largest possible delay range while maintaining the 2-ns step size. The inset shows a representative UEM SAED pattern (camera length = 150 mm) at -200 ns highlighting the and peaks with blue and gold boxes, respectively. Vertical lines in both panels are drawn at delay values of 175, 440, and 700 ns to show the reproducibility of the oscillatory behavior between experiments. In order to extract an intensity value from the SAED patterns, each Bragg spot was fit with a 2D Gaussian profile. The intensity of each time point was then normalized to the mean intensity from the entire delay range and offset for clarity. The agreement between the two runs shows the replicability of the Bragg-spot intensity fluctuations despite being acquired with significantly different experimental conditions (delay step order, scan length, camera length, exposure time).



FIG. S2. SAED intensity behavior for in-family planes. Temporal behavior of planes in the {111} family as a function of delay. Time points were obtained at 2-ns increments and acquired in random order to account for photoelectron-intensity fluctuations due to laser/cathode instabilities over the course of the experiment. The intensities of each reflection have been normalized to the average intensity over the scan and offset for clarity.



FIG. S3. Orientation-dependent spectral response. (a) Time-domain Fourier spectrum of the peak intensity for eight low-order Bragg spots. Vertical lines denote frequencies selected for generation of the bubble plots shown in (b). The inset shows an SAED pattern with colored squares to denote the Bragg spot from which the Fourier spectra originate. (b) Bubble plots of Fourier amplitudes obtained from (a). Bubble size corresponds to relative Fourier amplitude. The scale (unfilled bubbles) in lower-right of the upper-left frame corresponds to relative Fourier amplitudes of 1.0, 0.75, 0.5, and 0.25 moving from largest to smallest, respectively.



FIG. S4. UEM CBED-disc intensity as a function of delay for a near-edge cantilever location. (a) Representative UEM CBED pattern obtained within the region outlined with the green circle in Figure 4 at -200 ns. The specimen was tilted 17° off the [011] zone-axis, and the beam converged to an angle of 5 mrad (2α). Patterns were acquired in random-time order in order to remove capture-order (real-time) artifacts. Discs are labeled according to their corresponding space-time plots shown in the following panels. (b) Space-time plot of Laue-disc intensity for the disc as a function of delay. The position axis for all space-time plots is parallel to the reciprocal lattice vector, though the zero-position value is arbitrarily assigned by the chosen spatial integration window. Thus, a change in position as a function of delay denotes movement of intensity across the Laue disc, either closer to or farther from the center spot. (c) Space-time plot of Laue-disc intensity for the disc a function of delay. (d) Space-time plot of Laue-disc intensity for the disc a function of delay. All three space-time plots show qualitatively similar behavior over the delay range, though the disc shows contrast modulation that is symmetric to that of the other discs. That is, when the intensity of the and spot moves toward the center spot, the spot moves away from the center spot. This dynamic-contrast behavior is indicative of specimen tilting, not drift or heating of the lattice. The primary frequency is 1.3 MHz, of which three to four periods can be seen in (b-d).

**Video Captions**

VIDEO S1. UEM SAED dynamics. The specimen was oriented close to the [011] zone-axis. A 40-µm selected-area aperture and a 200-µm condenser aperture were used. The patterns were acquired with a 10-kHz repetition rate and a 5-s integration time per frame. The video illustrates dynamics at 2-ns steps spanning -200 to 7000 ns (3600 total frames) and plays at 30 frames per second (fps).

VIDEO S2. UEM CBED dynamics. The specimen was tilted 17° off the [011] zone-axis, and the beam converged at an angle of 5 mrad (2α). The patterns were acquired with a 10-kHz repetition rate and a 2-s integration time per frame. The video illustrates dynamics at 2-ns steps spanning -50 to 2000 ns (1025 total frames) and plays at 30 fps. The images were acquired with a random delay order to account for real-time artifacts.

VIDEO S3. UEM BF imaging dynamics.The UEM images were acquired with a 10-kHz repetition rate and a 3-s integration time per frame. The video illustrates dynamics at 2-ns steps spanning -50 to 2000 ns (1025 total frames) and plays at 30 fps. The image series was corrected for specimen drift. Scale bar = 1 µm.

VIDEO S4. Frequency localization of dynamic contrast.Frames were generated by following the BF analysis described in the data analysis methods. The frames from Video S1 were used for this analysis. Briefly, a Fourier transform of the time-dependent intensity value of each pixel was performed. Each pixel in each frame of the frequency map is the absolute value of the Fourier spectrum for that pixel at the given frequency, thus yielding the oscillatory contrast response of the specimen at that location. At each frequency value of the Fourier spectrum, a 2D image was generated and placed in order of increasing frequency.