Dynamics of colloidal crystals studied by pump-probe experiments at FLASH.

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We propose time-resolved infrared (IR) pump and extreme-ultraviolet (XUV) probe diffraction experiment to investigate ultrafast structural dynamics in colloidal crystals with picosecond resolution. In our experiment, the temporal changes of Bragg peaks were analyzed and their frequency components were calculated using a Fourier analysis. Estimated oscillations in colloidal crystal were localized at a frequency range of about 4-5 GHz. Theoretical calculations of vibrations of isotropic elastic sphere based on the Lamb theory reveal 5.07 GHz eigenfrequency of the ground (breathing) mode of about 400 nm in diameter polystyrene spheres composing the colloidal crystal film used in our experiment.

Experiment

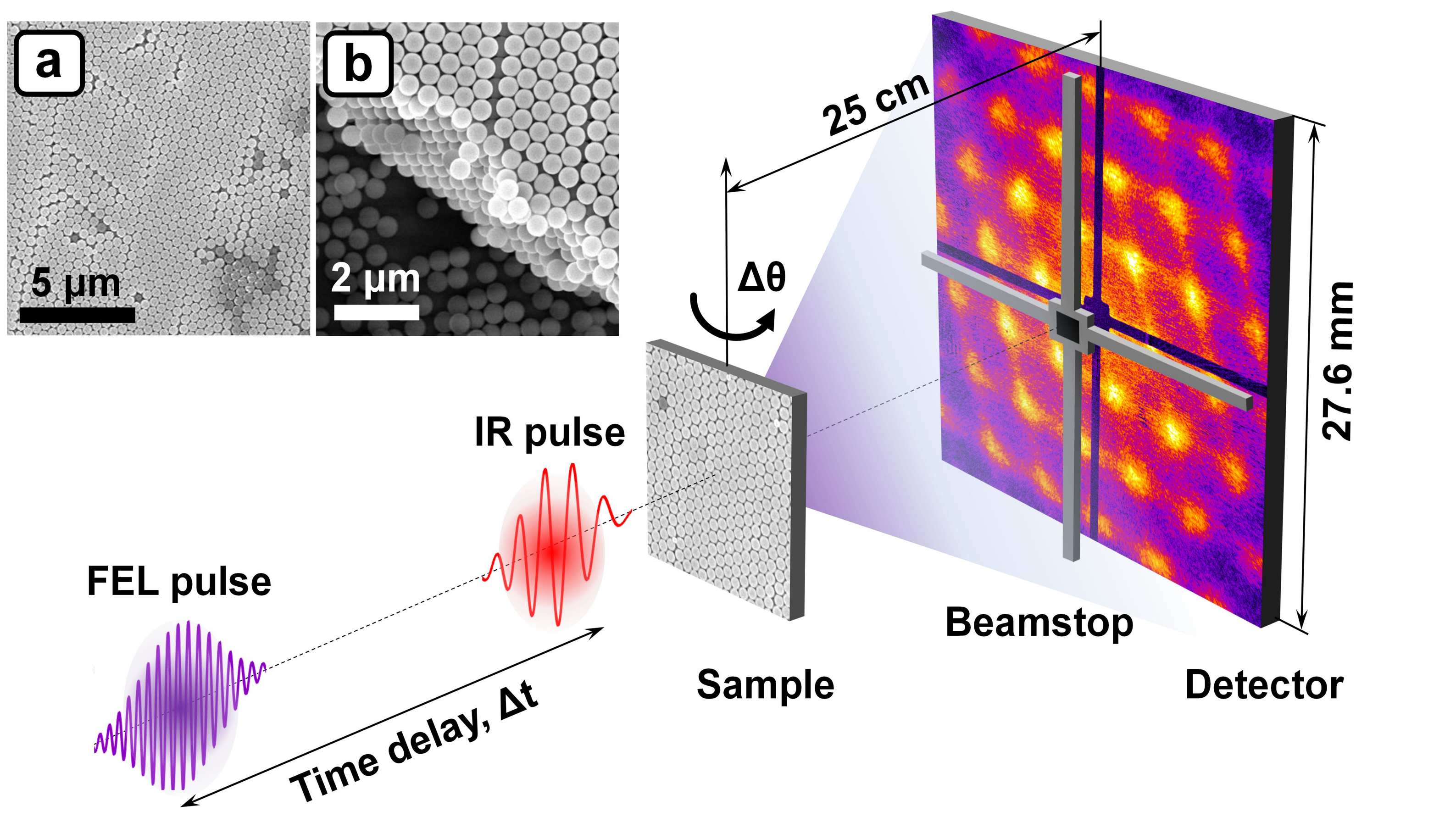

Figure 1: Schematic view of the pump-probe experiment showing infrared pump (IR pulse) and extreme-ultraviolet probe (FEL pulse) separated by the time delay Δt, the sample, and detector protected by the beamstop. Insets (a) and (b) show SEM images of colloidal crystal film used in our experiment.

- The experiment was carried out at BL3 at FLASH at DESY.
- Pump (IR) pulses are generated by a Ti:sapphire laser operating at 800 nm with a pulse duration of about 100 fs.
- Probe (FEL) pulses are the XUV 100 fs pulses (8 nm) from FLASH operated in a single-bunch mode.
- Time delay was varied from 0 to 1000 ps in 50 and 100 ps steps.
- The sample was an 11 layers thick colloidal crystal consisting of polystyrene spheres of about 400 nm in diameter (see insets in Fig. 1).
- The sample can be rotated around the vertical axis (Δθ).
- Diffraction patterns (see Fig. 2) were recorded on a CCD detector protected from the direct beam by a cross-shaped beamstop.
- Two series of the pump-probe experiments were carried out in a transmission geometry with different rotation angles of the sample: Δθ = 0° and Δθ = 35°.
- At each time delay 10 diffraction patterns were recorded in non-destructive regime.

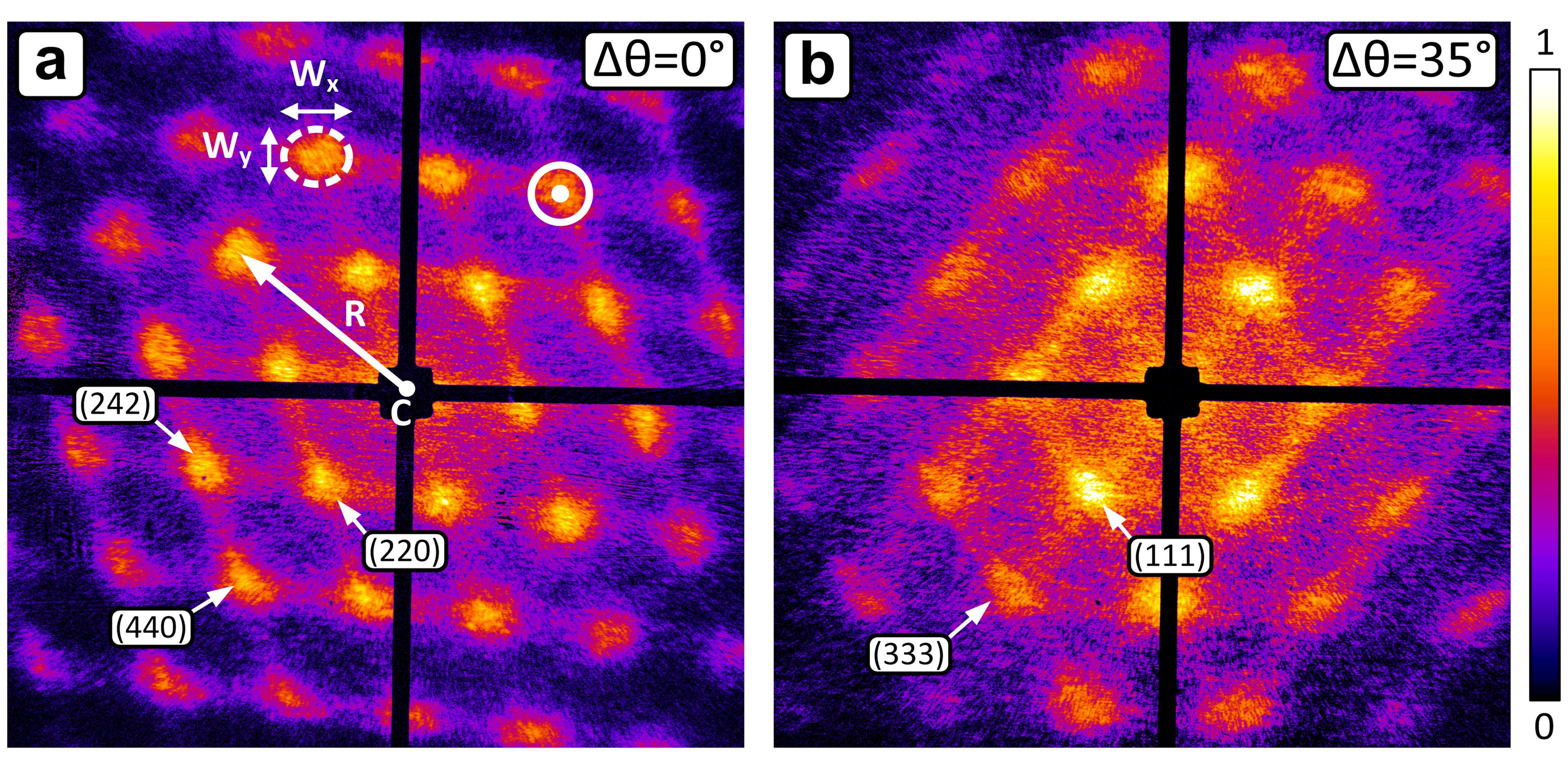

Figure 2: Selected single-shot diffraction patterns on a logarithmic scale measured at different azimuthal angle orientations (a) Δθ=0º, (b) Δθ=35º.

Data Analysis

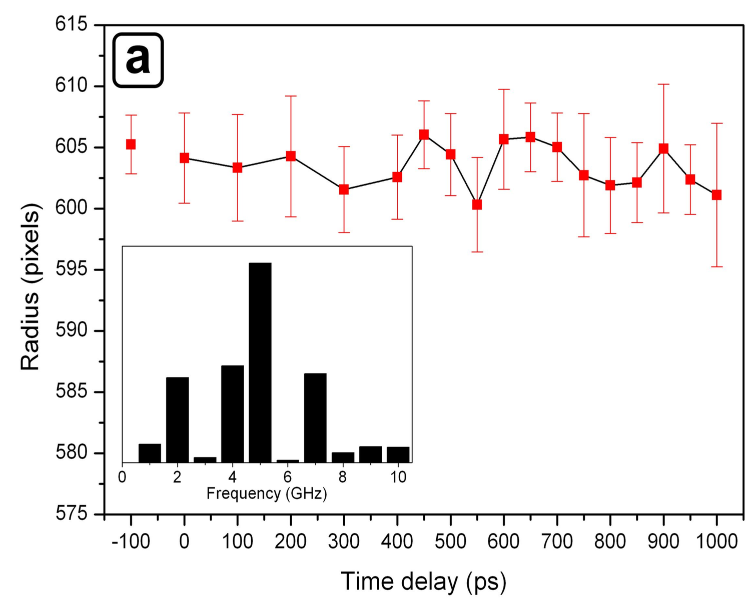

Figure 3: (a) Peak to center distance and (b) horizontal FWHM of the 440 Bragg peak shown in figure 2(a) for different time delay. Insets show the power spectrum of the Fourier transform of data.

We have analyzed each single-shot diffraction pattern in the following way:

- The center of each diffraction pattern and the relative peak to center distances were determined for the sets of 220, 242 and 440 Bragg peaks.
- For each Bragg peak the size (FWHM) was calculated in the vertical (W_y) and horizontal (W_x) directions (see Fig. 2a).
- We calculated the ratio of the integrated intensity within the center of peak to the integrated intensity in a ring with radius of 35 pixels (see Fig. 2a).
- For each time delay, all parameters were averaged over 10 diffraction patterns and their standard deviations were calculated (see Fig. 3a,b).
- To estimate characteristic frequencies probed in the experiment, we calculated Fourier transform of time-dependent parameters for all Bragg peaks and averaged their power spectrums (see Fig. 4).

All power spectrums show an increasing at the frequency region of 4-5 GHz.

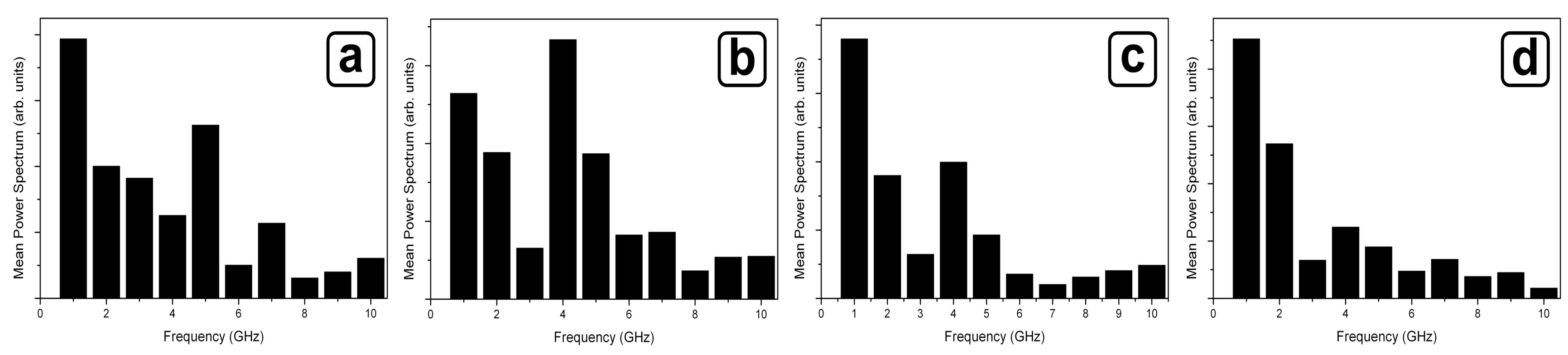

Figure 4: Averaged frequency components of (a) peak to center distances, (b) horizontal and (c) vertical beam sizes, and (d) ratios of peak to background intensity.

We also performed theoretical simulations based on the theory of vibrations of an isotropic elastic sphere and found the eigenfrequency of breathing mode 5.07 GHz, which corresponds to the period of vibrations of 197 ps.

Summary and outlook

In summary, we measured diffraction patterns from colloidal crystal film which was pumped by a short IR Laser pulses and probed by FEL, while changing the time delay between pulses at ps resolution.

Dynamics were studied by the analysis of Bragg peaks and Fourier transform of such parameters as the peak to center distances, horizontal and vertical peak sizes and peak to background ratios.

Proposed pump-probe experiments combined with the femtosecond x-ray coherent diffractive imaging technique has the potential to visualize the ultrafast motions in colloidal crystals with nanometer spatial resolution at the femtosecond time scale.