Imaging Lattice dynamics in individual nanocrystals

J. N. Clark[1,8,*], L. Beitra[1], G. Xiong[1], A. Higginbotham[2], D. M. Fritz[3], H. T. Lemke[3], D. Zhu[3], M. Chollet[3], G. J. Williams[3], M. Messerschmidt[3], B. Abbey[4], R. J. Harder[5], A. M. Korsunsky[6,7], J. S. Wark[2], I. K. Robinson[1,7]

1. London Centre for Nanotechnology, University College London, London WC1E 6BT, UK.
2. Department of Physics, Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, UK.
3. Linac Coherent Light Source, SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, CA 94025, USA.
4. ARC Centre of Excellence for Coherent X-ray Science, Department of Physics, La Trobe University, Bundoora, Victoria 3086, Australia.
5. Advanced Photon Source, Argonne, IL 60439, USA.
6. Department of Engineering Science, University of Oxford, Parks Road, Oxford OX1 3PJ, UK.
7. Research Complex at Harwell, Didcot, Oxfordshire OX11 0DE, UK.
8. Stanford PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA.
**jesclark@stanford.edu*

Abstract: We report imaging of coherent acoustic phonons on the picosecond time scale within a single nanocrystal using an X-ray free-electron laser. Our results allow unprecedented comparison with predictive models and observation of the vibrational modes .

OCIS codes: 050.1940, 100.5070

1. Introduction

Insights into the behaviour of materials can be gained by observing their response during phase transitions or when they undergo lattice distortion. Laser pulses on the femtosecond time scale can be used to induce disorder with the subsequent transient processes being stroboscopically probed using ultrashort (sub picosecond) pulses of visible light [1], X-rays [2] or electrons [3] in whats described as a pump-probe experiment. Using these techniques, many fundamental phenomena have been observed, such as non-thermal melting in semiconductors, thermal melting in metals, ultrafast bond softening and phase transitions [3–5]. However, many of these studies have been confined to bulk samples or ensembles of particles and the ability to spatially resolve (that is, to image) these phenomena on nanometre length scales and femtosecond time scales has so far been challenging. Here we demonstrate three-dimensional imaging of lattice deformation in a single gold nanocrystal using the Linac Coherent Light Source (LCLS) X-ray laser. This is achieved using time-resolevd coherent diffraction imaging. The method demonstrates *ab intio* determination of the vibrational modes of a nanocrystal without *a priori* information regarding morphology or size.

2. Methods

The experiment was performed at the XPP instrument at the LCLS in the USA. Coherent X-ray free electron laser (XFEL) pulses of energy 9 keV were monochromatized before diffracting from an isolated nanocrystal. Optical pump pulses were used to generate lattice distortion, with the transient diffraction recorded using an area detector positioned at a Bragg peak (Fig. 1 A). Rocking curves were collected yielding 3D diffraction patterns which were subsequently inverted using iterative phase retrieval [6] to provide real space images of the nanocrystal and lattice deformation (Fig. 1 B).

3. Results

Show in Fig. 1 B are isosurface renderings (20% of the maximum) of the nanocrystal density as a function of optical-pump, X-ray probe delay, with the projected displacement displayed on the surface. Regions of expansion and contraction are shown in red and blue respectively. Spatial and temporal variations are evident, indicative of non-homogenous lattice deformation. Cut-planes through the center of the 60 ps delay time show the characteristic regions of expansion and contraction obtained from the experiment (left) and are compared to continuum elasticity theory (center) and molecular dynamics simulations (right).

4. Conclusion

We have been able to image in three-dimensions the lattice deformation in a single nanocrystal without any *a priori* shape or size information [7]. The technique demonstrated here will enable detailed study of nano-particles and -structures in non-equilibrium regimes, such as those introduced in a pump-probe experiment.

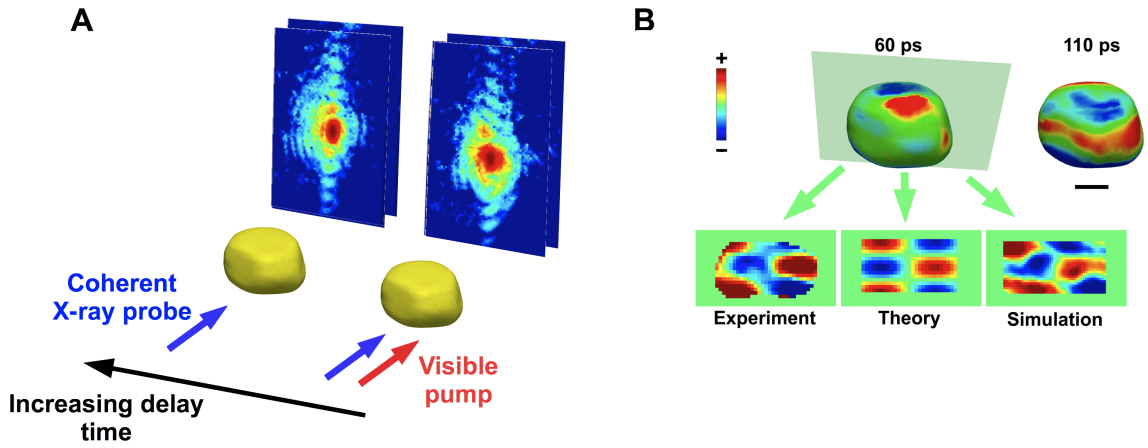

Fig. 1. a) Schematic of the experimental setup. A nanocrystal is perturbed using an optical pump pulse before a coherent X-ray pulse from an X-ray free electron laser is diffracted onto an area detector positioned at a Bragg peak. Rocking curves are collected to obtain three-dimensional data and the time-delay between pump and probe is changed to provide temporal data. b) Phase retrieval is performed on the three-dimsneiosnal data sets, yielding images of the density and projected displacement of the nanocrystal. The internal deformations obtained (experiment) can be compared to theory (theory) and molecular dynamics simulations (simulation). The scale bar is 100 nm.

References

1. C. V. Shank, R. Yen, and C. Hirlimann, "Femtosecond-time-resolved surface structural dynamics of optically excited silicon," Phys. Rev. Lett. **51**, 900–902 (1983).
2. C. Rose-Petruck, R. Jimenez, T. Guo, A. Cavalleri, C. W. Siders, F. Rksi, J. A. Squier, B. C. Walker, K. R. Wilson, and C. P. J. Barty, "Picosecond-milliangstrom lattice dynamics measured by ultrafast x-ray diffraction," Nature **398**, 310–312 (1999).
3. B. J. Siwick, J. R. Dwyer, R. E. Jordan, and R. J. D. Miller, "An atomic-level view of melting using femtosecond electron diffraction," Science **302**, 1382–1385 (2003).
4. C. W. Siders, A. Cavalleri, K. Sokolowski-Tinten, C. Tóth, T. Guo, M. Kammler, M. H. v. Hoegen, K. R. Wilson, D. v. d. Linde, and C. P. J. Barty, "Detection of nonthermal melting by ultrafast x-ray diffraction," Science **286**, 1340–1342 (1999).
5. A. M. Lindenberg, I. Kang, S. L. Johnson, T. Missalla, P. A. Heimann, Z. Chang, J. Larsson, P. H. Bucksbaum, H. C. Kapteyn, H. A. Padmore, R. W. Lee, J. S. Wark, and R. W. Falcone, "Time-resolved x-ray diffraction from coherent phonons during a laser-induced phase transition," Phys. Rev. Lett. **84**, 111–114 (2000).
6. J. R. Fienup, "Phase retrieval algorithms - a comparison," Appl. Opt. **21**, 2758–2769 (1982).
7. J. N. Clark, L. Beitra, G. Xiong, A. Higginbotham, D. M. Fritz, H. T. Lemke, D. Zhu, M. Chollet, G. J. Williams, M. Messerschmidt, B. Abbey, R. J. Harder, A. M. Korsunsky, J. S. Wark, and I. K. Robinson, "Ultrafast three-dimensional imaging of lattice dynamics in individual gold nanocrystals," Science **341**, 56–59 (2013).