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Deformation twinning of a silver nanocrystal under high pressure: supplementary materials

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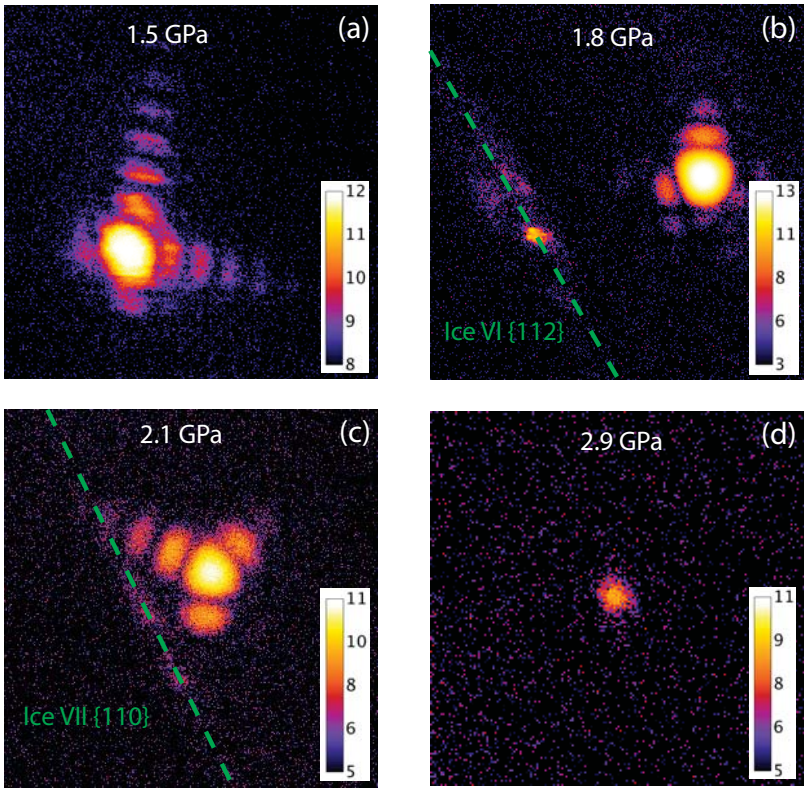
The choice of water for sealing sliver nanocubes inside the DAC was made for two main reasons: one is that the water environment serves as a good pressure transmitting medium, the other is when liquid water transforms to solid state, it holds nanocrystals' position and orientation allowing 3D Bragg CDI measurement. Molecular water has a complicated phase diagram. Liquid water transforms to Ice-VI at ∼0.9 GPa, and Ice-VI starts to transform to Ice-VII at ∼2.1 GPa. The Debye-Scherer rings created by these crystalline ice phases presented in the vicinity of silver [111] peak, when their lattice spacings were close enough. For instance, the d-spacings for Ice-VI {112} and Ice-VII {110} are 2.387 $\AA$ and 2.362 $\AA$, respectively, which are very close to the silver {111} d-spacing of 2.359 $\AA$. These two powder rings were observed during our measurement, as shown in Supplementary Figure S1 (b) and (c). Once the pressure was increased to 2.9 GPa, no crystal could be found to

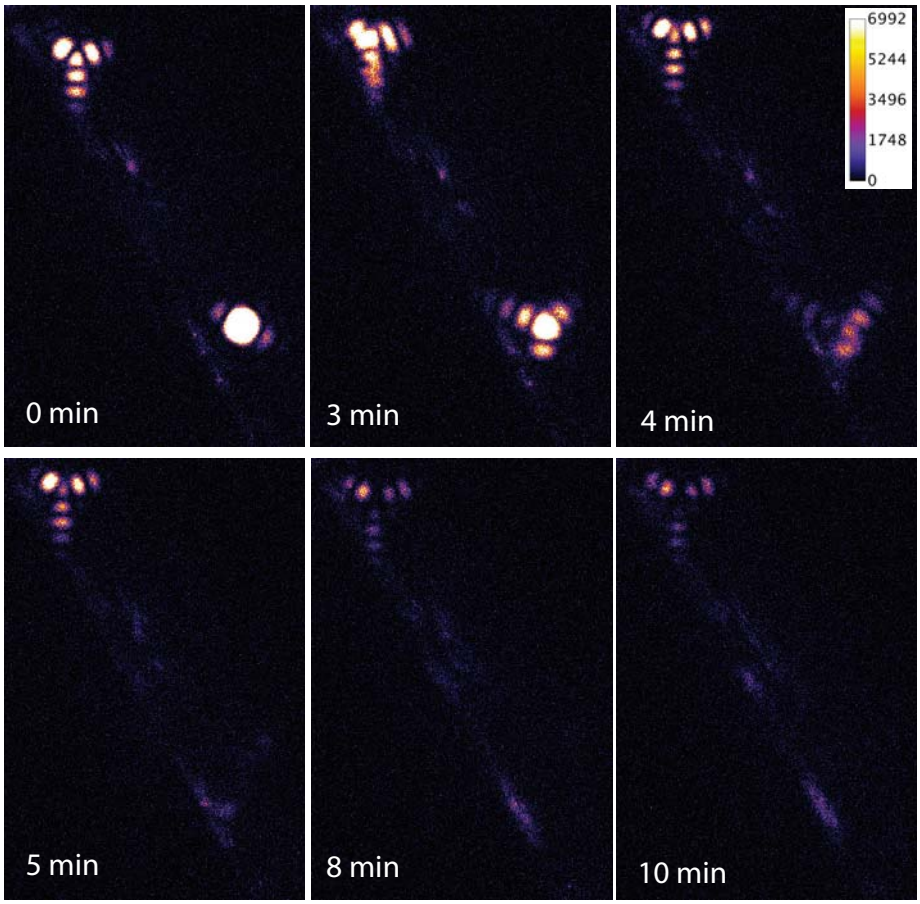
give interference fringes. This implies that the shape of silver nanocubes had become so severely deformed that no well-defined facet was remained at this pressure level, as shown in Supplementary Figure S1 (d).



Supplementary Figure S 1: Far-field diffraction patterns (on a logarithmic scale) in the vicinity of a silver (111) peak from different silver nanocubes under various pressures. At room temperature, liquid water transforms to Ice-VI at $\sim$ 0.9 GPa. Ice-VI starts to transform to Ice-VII at $\sim$ 2.1 GPa. The silver $\{111\}$ d-spacing is 2.359 $\AA$. The d-spacings for Ice-VI $\{112\}$ and Ice-VII $\{110\}$ are 2.387 $\AA$ and 2.362 $\AA$, respectively. At 2.9 GPa, no interference fringes can be found, which implies the cubic shape of the silver nanocrystals was destroyed at this pressure level.

The silver nanocrystals were not fully stabilized even after water transformed to solid state. This can be explained by the relatively low viscosity of Ice-VI phase in our measurement condition as discussed in the main content. It is noticeable that the instability behavior of crystals with slightly different orientations can differ dramatically. As shown in Supplementary Figure S2, a crystal, which was slightly off the exact Bragg condition, held its position and orientation for about 10 minutes, while another crystal, which well satisfied Bragg condition at the beginning, spun away quickly within 4 minutes. This observation

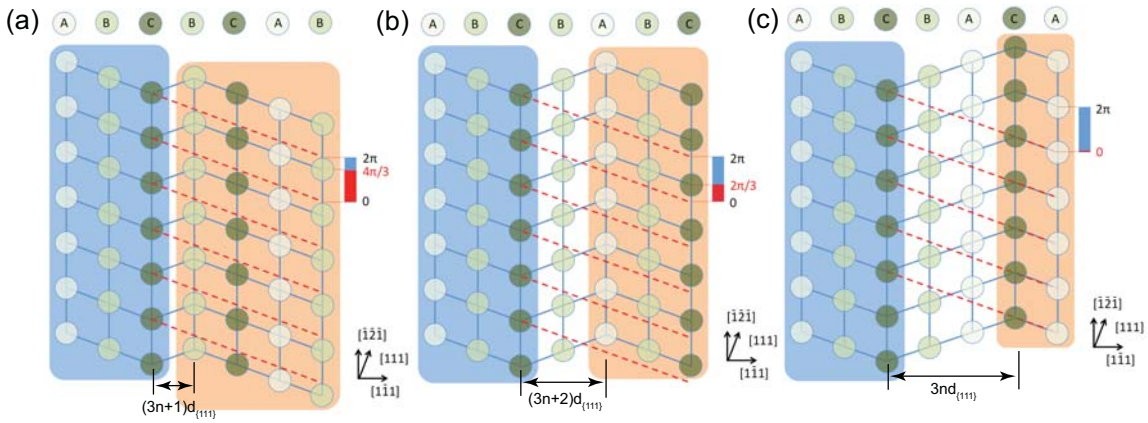
indicates that the momentum transfer introduced by Bragg diffraction was the dominant driving force on these silver nanocubes.



Supplementary Figure S 2: Silver (111) Bragg peak (on a logarithmic scale) drifting on and off Bragg condition at 2.1 GPa. The crystal (near bottom right) that satisfied the Bragg condition moved faster than the one (near top left), which was off the Bragg condition. This suggests that the Bragg diffraction-introduced momentum transfer dominated the driving force on these silver nanocubes.

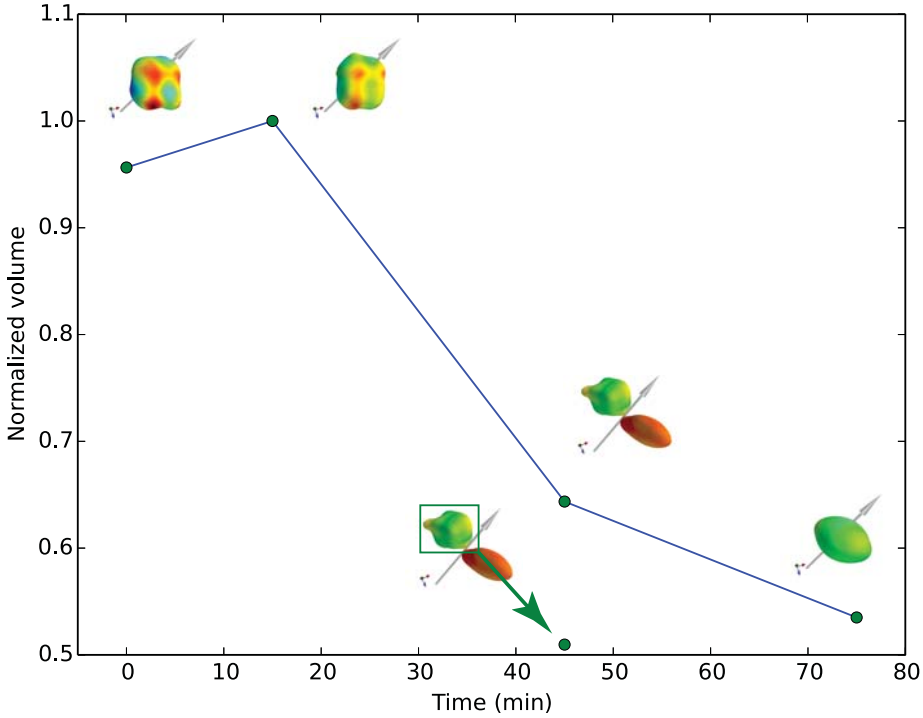
When a crystal breaks into twin domains, the crystal twin is mirror symmetric to the initial crystalline structure. The twin domain is thus not detectable at the initial Bragg condition, but leaves missing electron density in Bragg CDI reconstruction. Determined by the width of the twin domain, the phase offset between remaining crystal fragments can be estimated. Supplementary Figure S3 illustrates three offset conditions for the measured deformation twinning system along $[1\bar{1}1]$ direction. A deformation twining width of $(3n+1)\cdot d_{\{111\}}$ (a),

$(3n+2) \cdot d_{\{111\}}$ (b) and $3n \cdot d_{\{111\}}$ (c) introduce $\frac{4}{3}\pi$, $\frac{2}{3}\pi$ and $0/2\pi$ phase offsets observable in [111] direction, respectively.



Supplementary Figure S 3: Phase offsets for different deformation twinning widths. A deformation twining width of $(3n+1) \cdot d_{\{111\}}$ (a), $(3n+2) \cdot d_{\{111\}}$ (b) and $3n \cdot d_{\{111\}}$ (c) introduce $\frac{4}{3}\pi$, $\frac{2}{3}\pi$ and $0/2\pi$ phase offsets, respectively.

The volume evolution detected at the [111] Bragg peak during the continuous deformation process is plotted in Supplementary Figure S4. At the early stage of deformation, The crystal volume remained almost the same. When the deformation twinning occurred, the crystal volume was dramatically deceased to about 64%, accounting to missing section of twin domain and other disorientations during the plastic deformation process. One piece of the separated crystal fragments was further deformed and spun off the original Bragg condition, and only the other piece was observed in the last measurement.



Supplementary Figure S 4: Volume evolution of the silver nanocrsytal measured at [111] Bragg peak. The crystal volume remained almost the same at the early stage of the deformation for the first 2 measurements. When the deformation twinning occurred at the third measurement, the crystal volume was deceased to about 64%. The volume of the larger piece of the split crystal was stabilized and kept nearly unchanged in the last measurement.