Imaging microstructure in crystals using coherent X-ray diffraction

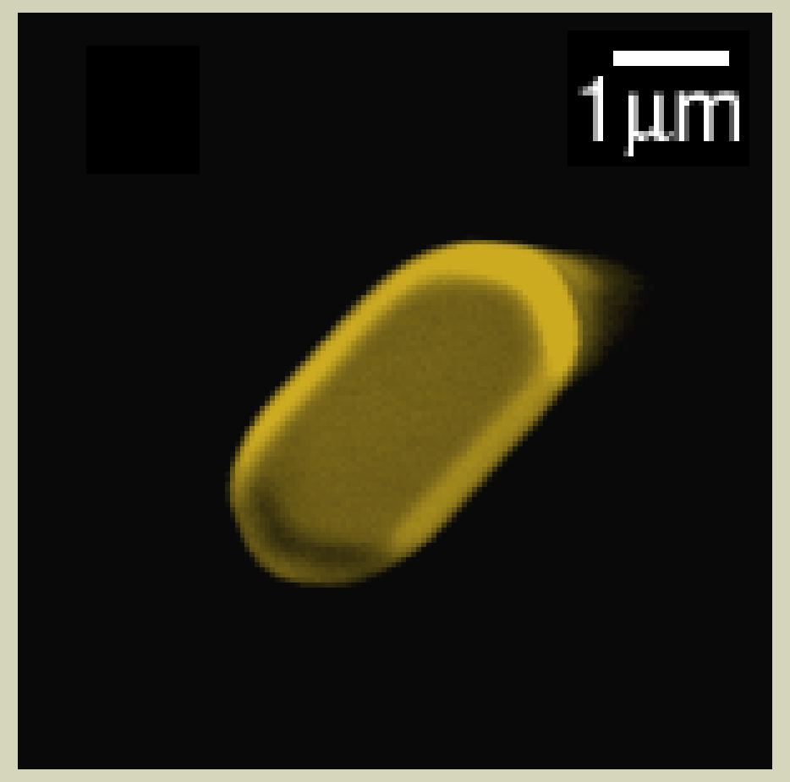
G.J. Williams[†], M.A. Pfeifer, I.A. Vartanyants, and I.K. Robinson
Dept. of Physics, University of Illinois at Urbana-Champaign

Introduction

Imaging microstructure in small crystals remains a difficult task due in part to a lack of 3D techniques for imaging these structures. Here, we demonstrate the potential to image the 3D density of a crystal by means of a coherent X-ray diffraction (CXD) experiment and subsequent phasing of the CXD pattern.

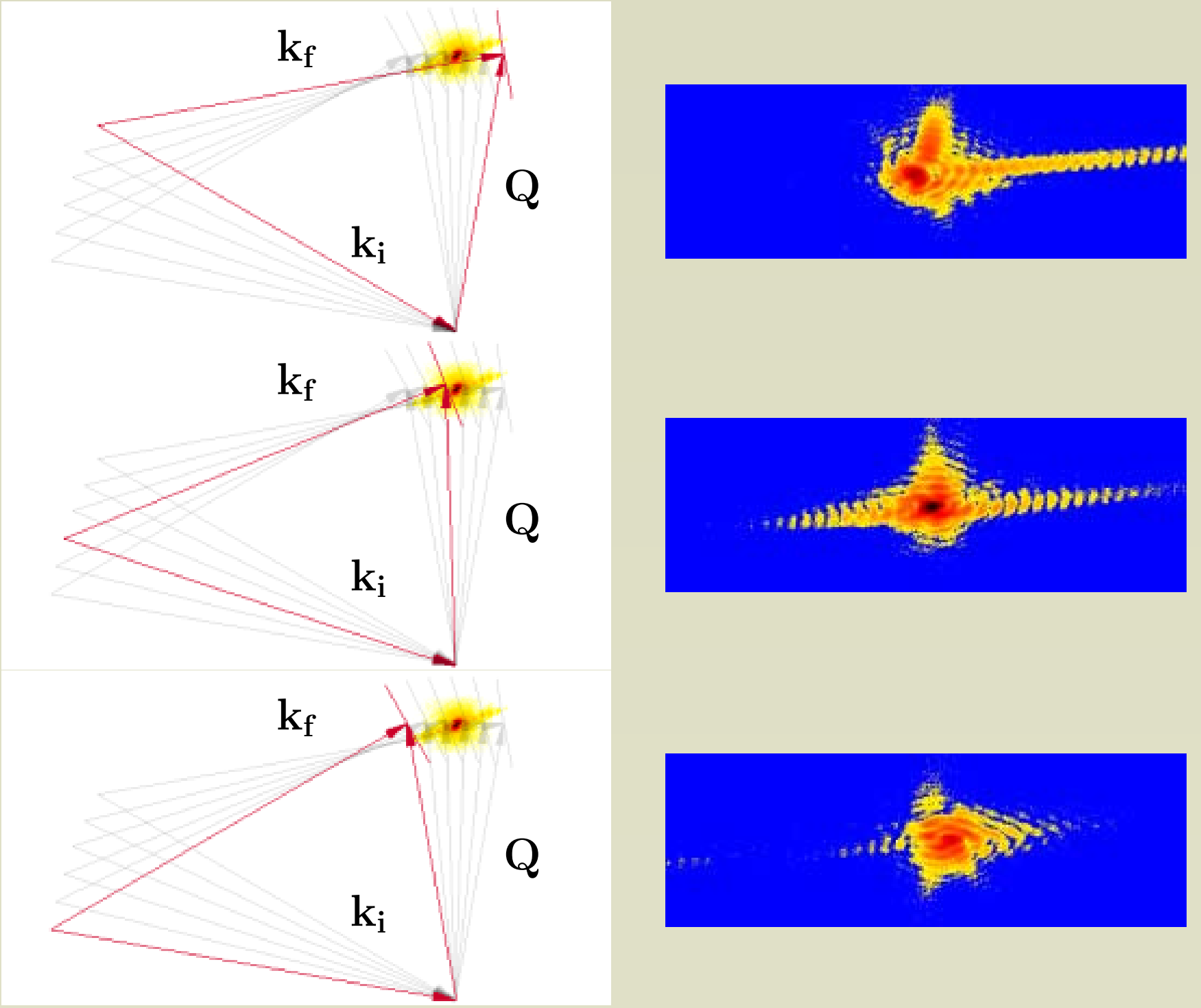
Theory and Sample Preparation

In the kinematical limit, the diffracted intensity is well understood to be the Fourier transform (FT) of the diffracting density, of which we may measure the magnitude of the amplitude only. Thus, to recover the shape of the crystal, we must measure the intensity distribution near a Bragg point and recover the lost phase information computationally. To prepare crystals smaller than the coherence volume of the X-ray beam, we evaporated Au onto Si(100) wafers, whose native oxide was left intact. We then placed the sample in an *in situ* heating cell and held the sample temperature above 950° for tens of hours to produce small crystals similar to the one shown right. The Pb samples were created analogously in a UHV chamber.



SEM image of a Au crystal.

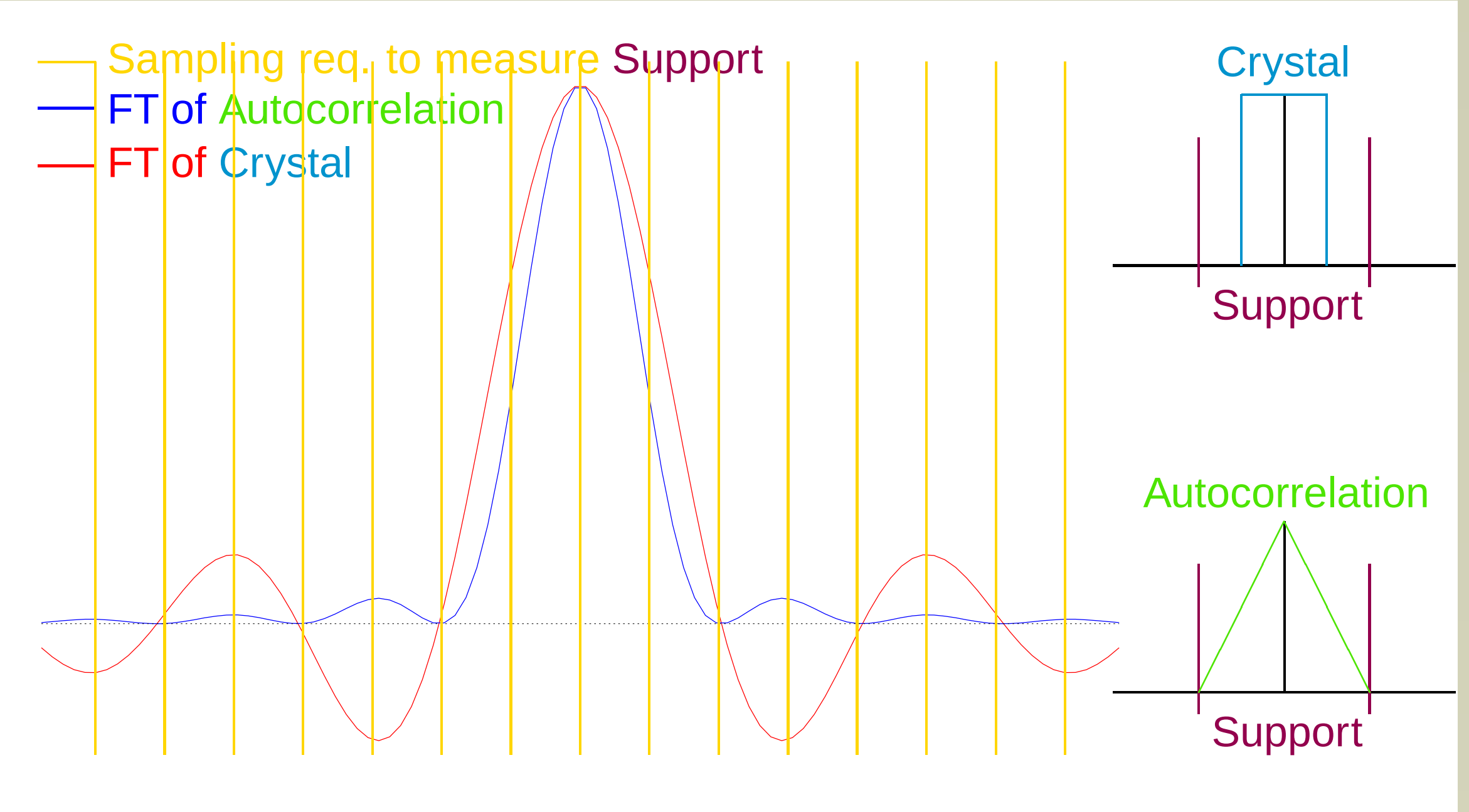
Data Collection



Data is collected by rocking the sample in 0.002° increments acquiring 2D slices through a 3D CXD pattern

The coherence of the X-ray beam is determined in the transverse directions to be 13μm $\times$ 60μm, by the source size, and 0.5μm in the longitudinal, by a Si(111) DCM. Since the Au crystals have off-specular $\{111\}$ reflections randomly oriented about the shared specular (111) reflection, we may use diffraction to select a single crystal. Then, by rocking the sample very slightly, we may capture 2D slices through the 3D CXD pattern as shown to the left.

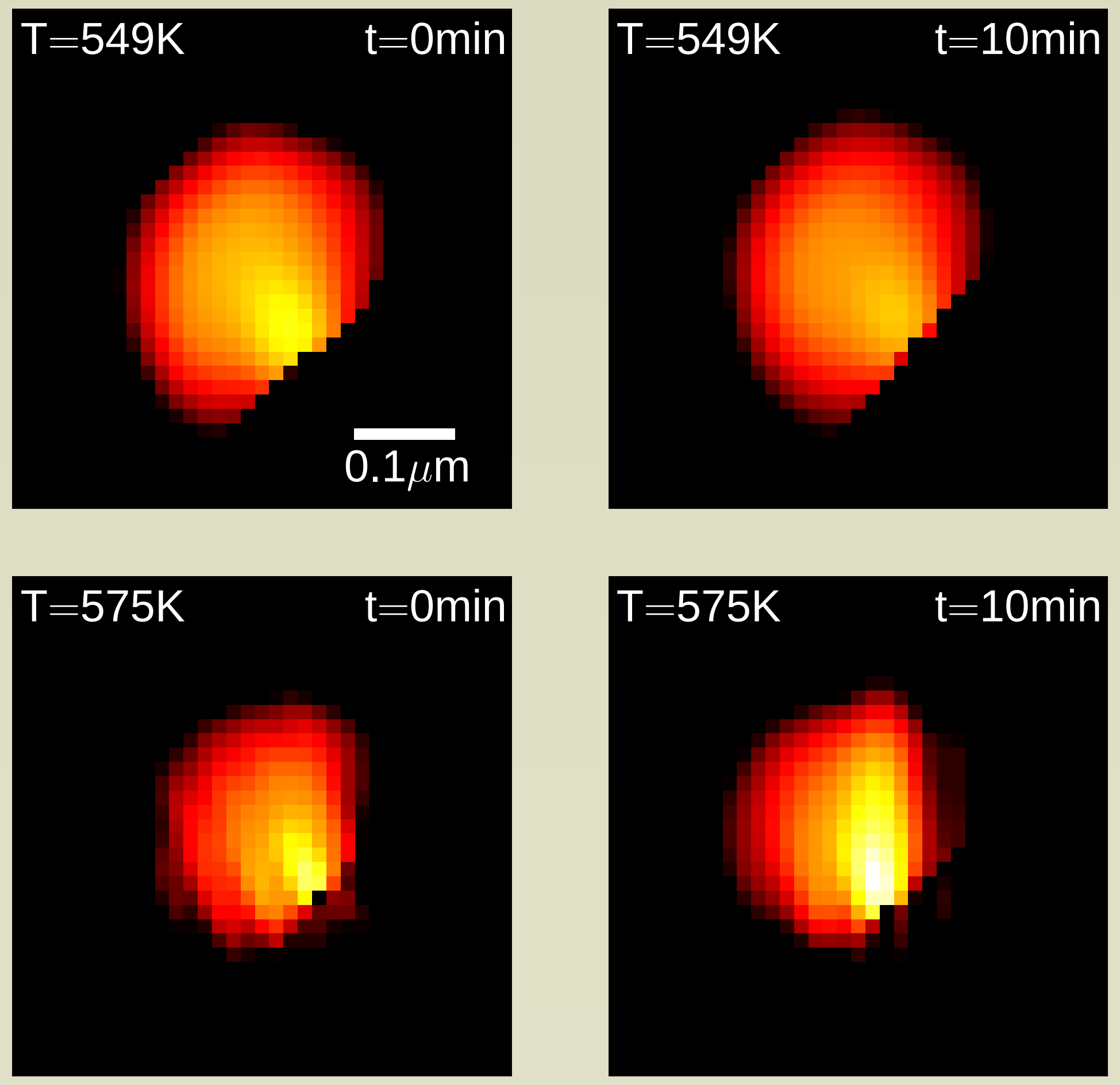
Oversampling and Phasing



The intensity, the FT of the autocorrelation, must be oversampled by a factor of 2 so that the FT of the crystal is sampled at the Nyquist frequency.

Since we measure intensity, we have lost the phase information that we need to invert the measured pattern directly into the 3D density. Since diffraction is continuous in reciprocal space, we can sample the CXD pattern more than twice as finely as the Nyquist rate–*i.e.*, *oversampling by a factor of 2*–and attempt to iteratively phase the measured CXD pattern.

2D Results–Pb Crystals

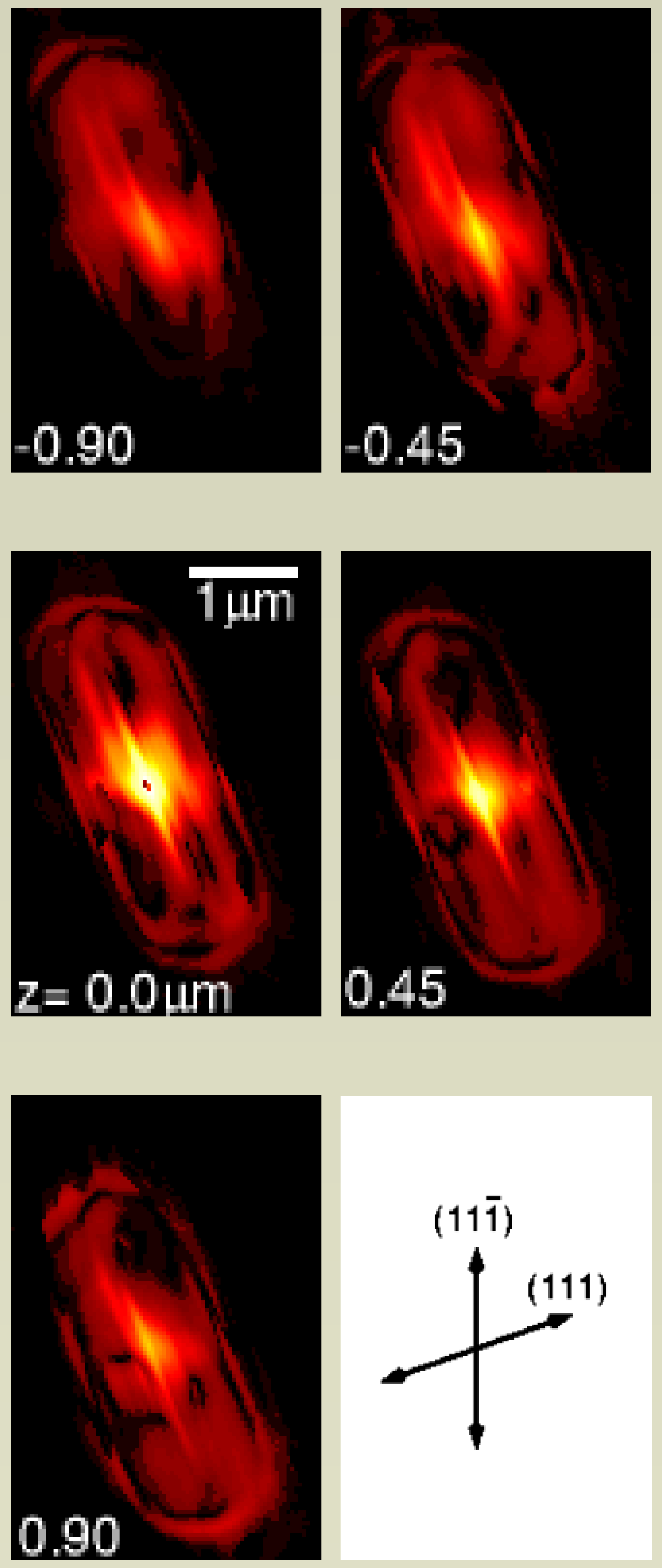


2D projections of the crystal density result from the phasing of a 2D CXD pattern. At 549K and 575K, the sample is seen to change with time.

Results from the 2D version of the CXD experiment on Pb samples–prepared analogously to the Au samples–are shown above. In this case, the reconstructed density is the projection of the 3D density onto a plane. One can clearly see the change of the particle's shape with both increasing temperature and annealing at high temperature. In the highest temperature case, we speculate that the crystal is rotating, since the diffraction disappeared soon after collection was completed. The Pb experiment was performed in a UHV system to prevent contamination.

3D Results–Au Crystals

2D slices through the reconstructed 3D density of a Au crystal are shown right. We can see that the size and shape of the reconstructed density is well within the distribution identified by SEM. The very high, pyramidal region in the center of the reconstructed density is believed to be an effect of the partial coherence of the incoming beam. Specifically, a small inclusion in a Be window 6m upstream should cause a perturbation of the illumination function and thereby give rise to an artifact of this size in the reconstruction. There is also a small 'hole' in the particle that is likely due to a slight misalignment of that region relative to the rest of the diffracting density. Finally, there appear to be narrow bands of no density in the (111) direction. These bands are at the limit our our present resolution, 50nm, but could be caused by twinning within the crystal. Future work includes characterization of the partial coherence of the incident X-ray beam and alteration of the iterative fitting to account for the presence of strain within the crystal.



2D slices through the 3D reconstructed density. Density contrast is seen in the (111) direction.

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Acknowledgments

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