

Femtosecond x-ray diffraction using the rotating crystal method

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Abstract. We demonstrate the rotating-crystal method in femtosecond x-ray diffraction. Structural dynamics of a photoexcited bismuth crystal is mapped in a pump-probe scheme by measuring intensity changes of many Bragg reflections simultaneously.

1 Introduction

Femtosecond x-ray diffraction provides direct insight into structural dynamics of crystalline materials on the length scale of chemical bonds and the ultrashort time scale of atomic motions. The spatial resolution R is determined by the x-ray wavelength λ and the highest measurable diffraction angle 2θ : $R = \lambda / (4\pi \sin \theta)$. Applying a pump-probe scheme with optical pump and hard x-ray probe pulses [1, 2], the temporal resolution is determined by the pump-probe cross correlation.

Until now, almost all ultrafast x-ray diffraction experiments have measured individual Bragg reflections at a time from a single crystal with a stationary orientation. Recently, we have demonstrated the powder diffraction method in a femtosecond optical pump - x-ray probe approach to measure the transient reflectivity of many Bragg reflections simultaneously [3]. This experiment revealed a by then unknown concerted transfer of electrons and protons in ammonium sulfate. A drawback of the powder method is that it cannot resolve reflections which have accidentally or systematically the same diffraction angle. In contrast, the rotation method with single crystals gives diffraction spots instead of rings, and, thus, allows for resolving reflections with equal diffraction angle. Here we present the first implementation of the rotating crystal method in femtosecond x-ray diffraction. To demonstrate its performance, we realized a rotating crystal experiment on bismuth which is presented in the following.

2 Experiment

Hard x-ray pulses of 100 fs duration and a wavelength of $\lambda = 0.154$ nm (Cu $K\alpha$) are generated by a 1 kHz laser driven plasma source which has been described elsewhere [2]. The x-ray pulses are focused onto the sample by a multilayer optics, resulting in a spot diameter of 200 μm and a photon flux of about $10^6/\text{s}$. We used a single crystal of bismuth with a cylindrical shape. The sample has a diameter of 8 mm, a thickness of 2 mm, and a surface roughness of 30 nm. The sample is excited by a 50 fs pump pulse at 800 nm and probed by a hard x-ray pulse under respective angles of grazing incidence of 7° and 1.5° . The geometry used is shown in Fig. 1 [4]. We chose the grazing incidence scheme to adapt the penetration depth of the probe to the penetration depth of the pump [5], the latter not being affected by the incidence angle. The pump pulse is π -polarized and the area density of absorbed energy has a value of $1\text{mJ}/\text{cm}^2$. To ensure that the angle of incidence remains constant while rotating the sample we aligned the rotation axis to be exactly perpendicular to the sample surface. As a reference for the fluctuations of the x-ray source we used the (111) reflection of a 60 μm thick diamond

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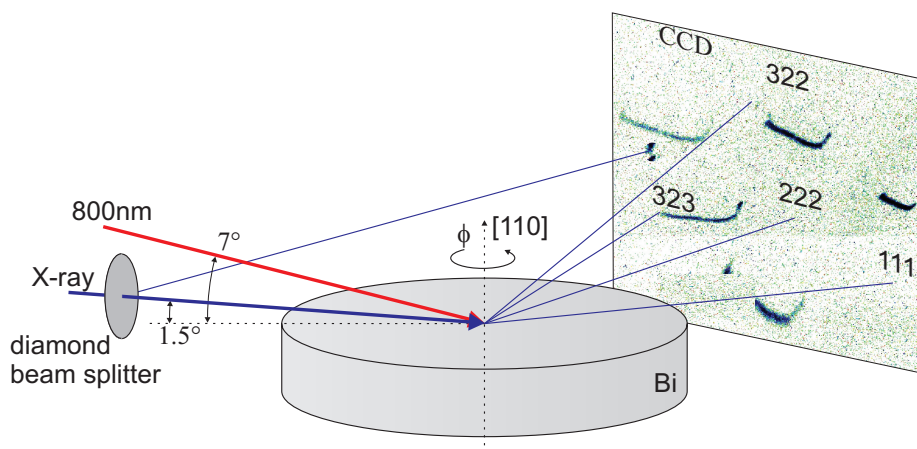


Fig. 1. Femtosecond grazing-incidence rotating-crystal diffraction setup showing the 800 nm pump, x-ray probe and diffracted beams. The diamond beam splitter is a diamond single crystal, which is cut parallel to the (111) lattice plane and aligned for the (111) reflection. The CCD screen shows the five reflections which were measured in the femtosecond experiment.

crystal in the incident x-ray beam. This x-ray beamsplitter reflects approximately 5% of the incoming x-ray intensity.

To enhance the signal-to-noise ratio we oscillate the sample within a 15° range and slow down the angular velocity at positions where reflections occur. All reflections, including the reflection from the diamond crystal are measured simultaneously and integrated on a deep depletion CCD.

3 Results and Discussion

Averaging over a large number of individual pump-probe scans gives the transients shown in Fig 2. The diffracted x-ray intensity is plotted as a function of pump-probe delay for the (222), (111), (322) and (323) diffraction peaks. The data for the (222) and (111) reflection are in good agreement with the literature data [5]. Note, that the oscillatory behavior of the (111) reflection is clearly visible even though the intensity changes are of the order of $\approx 5\%$ only. This demonstrates the high time resolution and the high sensitivity of our method. In the third and fourth panel of Fig 2 we show the first measurement of the (322) and (323) transient reflection [4], the (322) reflection shows an oscillation with identical frequency and phase as those of the (111) reflection.

4 Conclusion

We presented the first implementation of the rotating crystal method in femtosecond x-ray diffraction. We would like to emphasize that in the rotation method, as well as in the powder method, a large number of reflections are measured simultaneously under the same conditions, e.g., pump intensity, spatial overlap and temporal overlap. In contrast to powder-diffraction, the rotation method allows for a separation of different reflections which have the same diffraction angle.

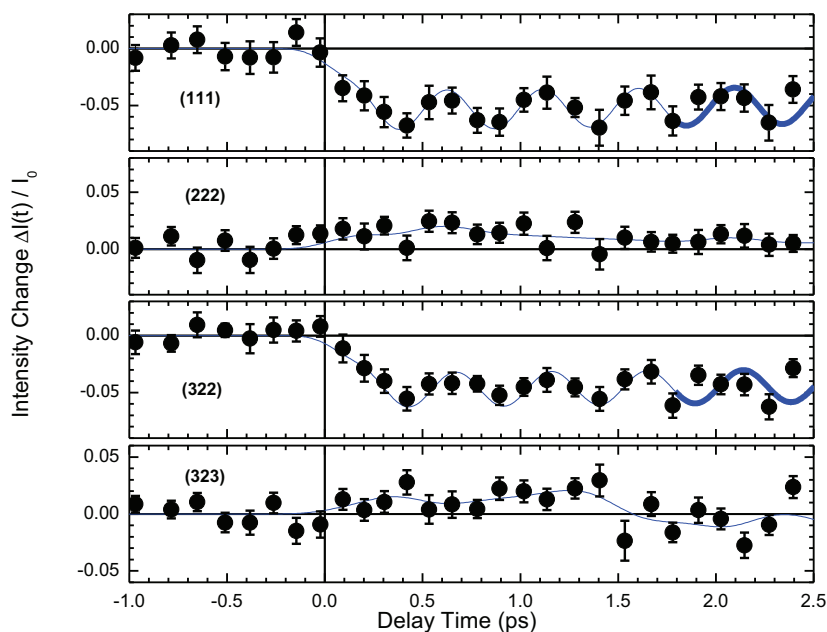


Fig. 2. Reflectivity change of particular reflections measured with the rotating-crystal experiment. The solid lines are guides to the eye.

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