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## **Report:**

## Summary

The original aim of the experiment is to study the details of the laser-induced amorphization in tellurium with optical pump, X-ray probe measurements. This goal has been achieved partially: we have observed partial amorphization of the sample and have been able to follow the time dependence of the re-crystallization. However, because only a small percentage of the sample was amorphized, the diffraction patterns were dominated by the crystalline peaks and the tails of these peaks. As a consequence, we are not able to obtain detailed structural information on the amorphous phase. Therefore, follow-up experiments with a different setup are necessary to achieve the original goal of the experiment.

## **Results**

A schematic diagram of the experimental setup is shown in Figure 1, as was described also in the proposal. We use an optical pump, grazing incidence X-ray probe setup to study the amorphization of Te by optical laser. The sample is kept at low temperatures using a liquid nitrogen cryostat.



Figure 1: Experimental setup. The sample is excited by an 800 nm laser at normal incidence. X-rays with 18 keV photon energy comes in with about 0.3 degree grazing angle. The WAXS pattern is collected by a detector.

A typical scan is performed in the following way: First, we collect several X-ray diffraction patterns of the cold (crystalline) sample. Then, we deliver 10 laser shots onto the sample. This is because the degree of amorphization was observed to increase significantly with the number of laser shots, up to about 10 shots. The same phenomenon was also observed by Cheng et al. [1]. After the laser shots, we collect X-ray diffraction patterns at regular intervals, typically every 5 s. We observe that the sample re-crystallizes gradually and nearly returns to its original state after sufficient wait time.

Figure 2 shows more details on the recrystallization process using wide-angle X-ray diffraction. Panel A shows the azimuthally integrated diffraction data, I(Q). From dark red to dark blue, the curves show increasing delay after the laser shots when the X-ray diffraction pattern is taken. The curves almost overlap because the differences between them are very small. These differences become clearer when the diffraction pattern from the cold crystalline sample is subtracted, as shown in panel B. In particular, in the diffraction pattern immediately after the laser shots (dark red), there is a clear increase of a broad feature spanning 1 to 3 Å<sup>-1</sup>, as well as a clear decrease in the crystalline peak slightly below 2 Å<sup>-1</sup>. This indicates that the sample is partially amorphized by the laser shots. With increasing delay time (from red to blue in the plot), these two features gradually decrease in amplitude, and the I(Q) curve recovers its original value.

We also analyze quantitatively the evolution as a function of time, as shown in panels C and D. The former shows the change in the intensity of the crystalline peak (slightly below 2 Å<sup>-1</sup>) and the latter the change in the intensity of the amorphous feature, both with respect to the cold diffraction data. These time-dependence data can be well described by an exponential decay function fit, as indicated by the red dashed lines. The extracted decay time constant,  $\tau_0$ , is about 90 s.

We note that similar re-crystallization results have been observed by Cheng et al. [1] using laser reflectivity measurements, although the time constant  $\tau_0$  is slightly shorter. Our structural measurement results confirm the interpretation of Cheng et al. [1].



Figure 2: Partial amorphization and re-crystallization of Te at 253 K. (A) Azimuthally integrated diffraction intensity, I(Q); from dark red to dark blue, the curves show increasing delay up to ~300 s after the laser shot. The curves almost all overlap. (B) Same as A but subtracting I(Q) of the cold (crystalline) data taken before the laser shot. With increasing delay, the amorphous feature decreases and the crystalline peak recovers. (C) Change in the Te peak intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot. (D) Change in the amorphous intensity compared with the cold shot.

However, because the amorphization is only partial and the crystalline peaks are intense, as can be seen in Fig. 2A-B, the extracted amorphous signal is strongly influenced by the tails of the crystalline peaks, especially at high Q. This problem could not be resolved by changing the fluence of the 800 nm laser. Therefore, we have not been able to obtain detailed structural information on the amorphous phase. Future experiments with a different setup design are necessary to achieve the original goal.

References

[1] Y. Cheng et al., *Phys. Rev. B* 98, 134112 (2018), doi: <u>10.1103/PhysRevB.98.134112</u>