Swift Heavy Ion-Induced Amorphization of $CaZrO_3$ Perovskite*

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Perovskite structures (ABO_3) are an important class of ceramics with a large variety of derivative structure-types (e.g., cubic, hexagonal, and orthorhombic). Owing to this structural flexibility, perovskites have very different physical and chemical properties leading to numerous technological applications. Radiation damage in perovskites is of interest due to their potential as actinide waste forms.

Powder $CaZrO_3$ perovskite was irradiated with 940-MeV Au ions at the UNILAC M2-beamline [1]. The irradiations were performed in steps of 1×10^{12} ions/cm^2 up to a fluence of 8×10^{12} ions/cm^2 with two additional exposures at 1×10^{13} ions/cm^2 and 1.5×10^{13} ions/cm^2. During beam interruptions, the structure of the powder material was analysed by *in situ* X-ray diffraction (XRD) measurements. The instrument is a standard 4-circle X-ray diffractometer (Cu-K_α) operating in vacuum and equipped with a position-sensitive detector. Within the maximum X-ray range of about 10 µm, the electronic energy loss (dE/dx) of the Au ions was with ~36 keV/nm nearly constant, and the nuclear dE/dx was well below 0.5% of the total energy loss (SRIM 2008). Samples from irradiations at two different fluences were additionally investigated by transmission electron microscopy (TEM).

Ion-induced amorphization is evidenced in the XRD patterns by a decrease in diffraction intensity and an increase in diffuse scattering (Fig. 1a,b). The crystalline and amorphous contributions were quantified by peak-fitting the diffraction profiles at each irradiation step yielding position, FWHM, intensity, and area of the diffraction maxima. With increasing fluence, the normalized area of the 101 and 020 peaks (~22°) decreases quickly (Fig. 1c: filled circles). Concurrently, the normalized area of the broad amorphous peak (~30°) grows linearly in the initial stage and finally reaches saturation at higher fluences (Fig. 1c: filled squares). This behaviour can be understood in terms of the direct-impact model, where amorphization is described by the accumulation of individual amorphous tracks. At larger fluences, tracks begin to overlap, and the increase/decrease in amorphous/crystalline fraction becomes sub-linear. The amorphization cross-section per incident ion, σ, was determined by fits to the XRD data (Fig. 1c), and a track diameter d was extracted by assuming cylindrical track geometry ($\sigma = \pi\cdot(d/2)^2$). Based on the average of several diffraction maxima (including the broad amorphous peak), individual tracks have a calculated diameter of 6.0 ± 0.6 nm. This is in good agreement with independent TEM measurements yielding a track diameter of 6.7 ± 0.4 nm.

With accumulating radiation, the 2θ positions of all diffraction maxima showed a complex behaviour, indicating contributions from several processes. The experimentally determined FWHM of the main diffraction maxima increased significantly with ion fluence. Based on a Williamson-Hall plot, strain-induced broadening is the dominant process with microstrains approaching the elastic limit [1].

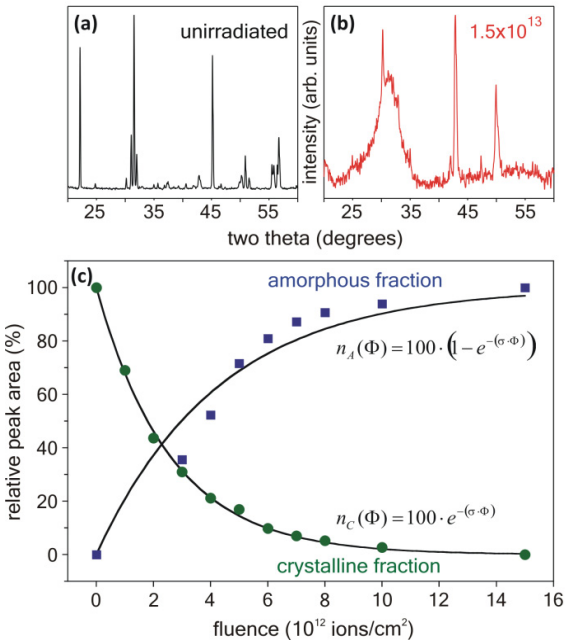

Figure 1: XRD patterns of $CaZrO_3$ perovskite (a) before and (b) after irradiation with 940-MeV Au ions, indicating full amorphization at 1.5×10^{13} ions/cm^2 (remaining peaks are due to copper sample holder and Si substrate). (c) Relative peak area, of amorphous, n_A, (diffuse peak at ~30°) and crystalline, n_C, (peak at ~22°) fraction as a function of increasing fluence. The curves are fits to the data by the given Eqs. and provide the amorphization cross-sections, σ, and track diameter d.

References

[1] M. Lang, F.X. Zhang, W.X. Li, D. Severin, M. Bender, S. Klaumünzer, C. Trautmann, R.C. Ewing, Nucl. Instr. Meth. B (2012) in press, doi:10.10/16/j.nimb.2011.12.028.

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