

Swift heavy ion induced radiation damage in EuPO_4 * †

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Europium phosphate (EuPO_4) is the synthetic analogue to the natural mineral monazite and is considered as a promising material for long term storage of radioactive waste because of its structural flexibility incorporate uranium and thorium without significant structure degradation due to self-irradiation. Earlier irradiation experiments with 800keV- Kr ions revealed a high radiation hardness of monazite-structure orthophosphates and low critical temperature, above which the material can no longer be amorphized [1]. In contrast to displacive radiation effects there is no information available about the response of EuPO_4 to the irradiation with swift heavy ions (SHI) where electronic energy loss processes dominate.

Single crystals of EuPO_4 were irradiated at the UNILAC facility using 2.0-GeV Au ions (energy loss 39 keV/nm). The experiments were performed at room temperature using a flux of 3×10^8 ions/cm²s and fluences between 5×10^{11} and 1×10^{13} ions/cm².

For damage analysis, the irradiated samples were investigated by Raman spectroscopy using a Horiba Jobin Yvon LabRam HR800 system with a laser of $\lambda = 473.1$ nm.

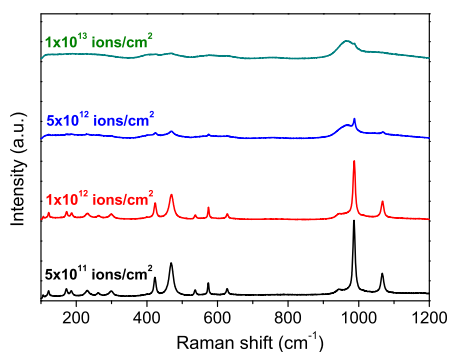


Figure 1: Raman spectra of EuPO_4 crystals exposed to 2.0 GeV Au ions of different fluences (spectra are stacked for better visualization). All spectra were recorded on spontaneously cleaved crystal planes normal to the irradiated surface at 10 μm depth.

Due to irradiation-induced stress accumulation, the crystals broke perpendicular to the irradiated surface. This created a flat normal-to-surface plane and allowed us to record Raman spectra along the full range of the ions.

Raman spectra of virgin lanthanide phosphates LnPO_4 have previously been reported, assigning the most intense peak at 987 cm^{-1} in the EuPO_4 spectrum to the symmetric stretching mode of the $(\text{PO}_4)^{3-}$ group [2]. After applying a fluence of 5×10^{11} ions/cm², no significant change in

the Raman spectrum was observed, while further irradiations result in an overall increase of the background and a strong broadening of all Raman bands (Fig. 1). At high fluences, a broad peak develops at around 960 cm^{-1} . Given by the similarity of Raman spectra of fully amorphized phosphates and amorphous phosphate glasses, we ascribe this band to the vibrations of the $(\text{PO}_4)^{3-}$ tetrahedron in the amorphous matrix. At a fluence of 1×10^{13} ions/cm², almost all bands from the original crystalline phase disappeared. The damage cross section was estimated by analyzing the band area of the $(\text{PO}_4)^{3-}$ symmetric stretching mode from the crystalline (987 cm^{-1}) and the amorphous (960 cm^{-1}) state as a function of fluence. Based on the single-impact model [3], the damage accumulation increases in the initial stage linearly with increasing fluence and saturates for track overlapping. Applying the Poisson law allows us to determine the track radius R .

$$\frac{A_c}{A_c + A_a} = \exp(-\pi R^2 \Phi) \quad (1)$$

where Φ denotes the fluence, and A_c and A_a represent the respective band area of the crystalline (987 cm^{-1}) and amorphous (960 cm^{-1}) Raman peak. Fitting the data (Fig. 2) by Eq. 1 yields a track radius of $R = 3.4 \pm 0.2$ nm which is consistent with track radii in other minerals.

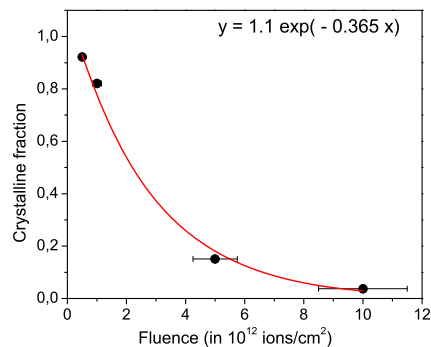


Figure 2: Crystalline sample fraction as a function of fluence. From the exponential decrease, the track radius for 2.0-GeV Au ions in EuPO_4 is deduced. The solid line corresponds to the fit equation given as inset.

References

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