

Radiation response in fluorite-related systems with dual spatial length-scales

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Fluorite-related materials are encountered in nuclear applications as advanced fuels and inert matrix for minor actinides. Several of these compounds display great resistance to radiation-induced amorphization. In these systems, there seem to be specific differences in the detailed response to radiation because of the competition between cation and anion spatial correlations describing their organization.

Under irradiation, a sharp decrease of the characteristic spatial correlation length describing the O vacancy order is usually observed. But once radiation is switched off, the disordered fluorite phase is suddenly quenched to a broken-symmetry phase where domains with spatial correlations of the vacancies form and coarsen with time as the system tries to reach a local equilibrium on larger and larger scales. This process of return to equilibrium takes place maintaining a much longer correlation length of the cation sublattice, thus preserving a long-range crystallinity, so that the system behaves effectively as a paracrystal.

We believe the small correlation length of the anion sublattice can affect the energy dissipation process in the material during the irradiation: the fluctuation-dissipation theorem establishes a relation between equilibrium correlation functions and linear response functions; excess energy can be dissipated because of the work done by the fluctuating viscous stresses at domains walls in resisting elastic strain fluctuations, in a way akin to the familiar processes of fluid dynamics, so that the energy dissipation is most effective at the smallest turbulence scales. The nonequilibrium dynamics of these open systems seems effective in dissipating the energy of the irradiation process and this can provide a universal pathway for searching extreme radiation resistance materials.