Structural defect creation by swift heavy ions in CaF_2 single crystals

R. Assylbayev*[1,2], A. Akilbekov[1], A. Dauletbekova[1], A. Lushchik[2], E. Shablonin[2], E. Vasil'chenko[2], M. Zdorovets[3]

[1] L.N.Gumilyov Eurasian National University, Astana 010008, Kazakhstan
[2] Institute of Physics, University of Tartu, Tartu 50411, Estonia
[3]Institute of Nuclear Physics, Astana 010008, Kazakhstan
*Corresponding author: ruslanassylbay@yandex.ru

Calcium fluoride single crystals belong to the most popular optical materials due to their transparency in a wide spectral region. Pure and doped with different cation impurities (Me^{3+}, Me^{2+}, Me^{+}) CaF_2 can be used for many applications, for instance as lenses and windows in ultraviolet laser lithography, laser media, radiation detectors and dosimeters, elements for navigation and orientation, etc. (see, e.g., [1] and references therein). High-dense irradiation (e.g., by swift heavy ions - SHIs) creates in CaF_2 different structural radiation damages including F-type aggregate centres and metal colloids. The presence of extrinsic defects (impurities) and the rise of a number of structural intrinsic defects (point defects and their aggregates) during a prolonged irradiation strongly affect the properties of CaF_2 useful for different applications. More than 95% of the energy of SHI with $E > 1$ MeV/u is spent on ionization losses providing an extremely high density of electronic excitations within cylindrical ion tracks (see [2]).

Oxygen-free single crystals of CaF_2 irradiated at room temperature by SHIs in Darmstadt (^{209}Bi, energy per nucleon 11.4 MeV/u, 3×10^{11}-10^{12} ions/cm^2, ion range R ~90 μm) or at DC-60 cyclotron in Astana (^{132}Xe, 1.75 MeV/u, 5×10^{12}-10^{14} ions/cm^2, R ~18 μm) have been the main investigation objects. The radiation-induced optical absorption (RIOA) measured from 1.5 to 10.5 eV after preheating of the ion-irradiated samples to different temperatures (up to ~1200 K) has been analyzed in order to separate different types of F-aggregates, calcium colloids as well as the defects responsible for the absorption in UV-VUV spectral region (4.5-10.5 eV). In addition, the curves of thermally stimulated luminescence were measured during linear heating (β = 2 K/s) of preliminary irradiated samples. Activation energies and frequency factors have been determined for separated peaks in ion-irradiated pure and thulium-doped CaF_2 samples.

SHI-irradiation of CaF_2 causes the appearance of a complex wide RIOA band in visible range tentatively associated with F-aggregate centres and calcium colloids [1]. The annealing of different F-type bands mainly occurs at 400–600 K. There are several RIOA bands in UV-VUV spectral region as well. It is worth noting that just high-dense ion-irradiation is responsible for such radiation damage, because some of these bands (especially RIOA band peaked at ~9.7 eV) cannot be induced by 100-keV protons or X-rays (50 keV, 20 mA, W). Similar to the case of model alkali halides (see, e.g. [3]), these RIOA bands at 5–10 eV can be tentatively ascribed to trihalide quasi-molecules or electronic excitations localized near complex radiation damages (i.e. Greek bands in alkali halides), while a detailed study of their origin still lies ahead.

The joint action of impact (causing shock waves) and nonimpact (decay and recombination of radiation-induced electronic excitations) mechanisms is considered to be responsible for the creation of novel structural defects with complex structure (including 3D defects involving many host ions) under Xe^{132} or Bi^{209} irradiation. These 3D defects serve as stoppers for dislocations thus impeding their movement to the surface and facilitating the cracking and brittle destruction of the irradiated single crystals.

Thus, the irradiation with SHIs affects the mechanical properties and causes the efficient cracking and fracturing of CaF_2 single crystals into pieces with (111) faces. These mechanical effects are especially efficient in the samples irradiated with ^{209}Bi ions with an extremely high nuclear spin of 9/2, while ^{132}Xe ions have zero nuclear spin (see also [4]). A specific contribution of ion magnetic properties to radiation damage needs further analysis.

References

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