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Comparison between the Effects of Neutron Irradiation at-195° C and at 100° C on LiF Crystals

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The nature of defects formed by neutron irradiation under liquid nitrogen of lithium fluoride crystals is investigated by means of x-ray experiments. Complex defects are formed: they could have been produced either directly by the bombarding particles or by clustering of point defects. The number of complex defects (but not their nature) is a function of the neutron dose as indicated by preliminary experiments. The influence of further annealing is studied: cavities are observed which resemble very much those present in crystals irradiated at 100°C.

When LiF crystals are irradiated with thermal neutrons, defects are created: vacancies, interstitials and also some compound defects. The constitution and the number of these defects vary with the neutron dose, the temperature of irradiation further annealing.

In the first stage, we have examined, by means of x-ray experiments, crystals which were irradiated in the pile without any temperature control. The irradiation temperature was of the order of magnitude of 100°C. It was found that, for a neutron dose higher than 3×10^{17} nvt, the LiF crystals contain cavities, lithium platelets of several Å thickness and, for an integrated flux higher than 7×10^{18} nvt, metallic lithium in addition[1]. To distinguish between the effects due to the two factors, *i.e.*, irradiation and temperature, in the formation of these compound defects we had to carry out new experiments on samples irradiated at low temperature. Some LiF crystals, irradiated under liquid nitrogen (3×10^{18} nvt) at the C.E.N. of Saclay[2], were obtained. Our x-ray experiments were carried out at room temperature.

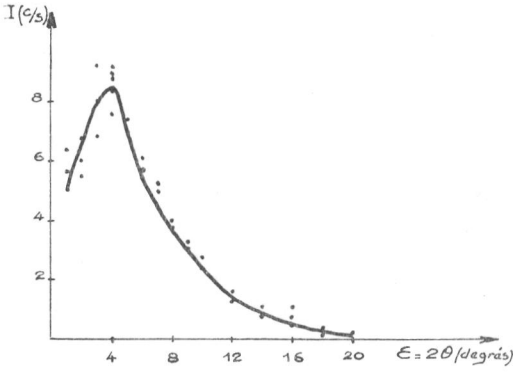

Fig. 1. Scattered intensity (room temperature).

These samples give an x-ray scattering which is isotropic around the origin of the reciprocal lattice. The scattered intensity first increases with the scattering angle ε, goes through a maximum and then falls to zero for $\varepsilon=22°$ (Fig. 1)[3]. This scattering is given by disturbed zones of the crystals: these zones seem to have a complex structure; they are formed by a cavity surrounded by a sheet, the electronic density of which is higher than the mean electronic density of the crystal[4]. From the experimental curve we deduce the diameters of the cavity and the zone itself as 3.5 and 17.5 Å respectively. These zones can be:

— directly created by irradiation at the end of the path of the α-particle and of the tritium nucleus,

— formed when the samples are brought up to room temperature: in some places the vacancies may group together and give cavities surrounded by sheets without vacancies.

From the experimental curve we deduce the number of zones as 3×10^{19}/cm^3 which is of the same order of magnitude as the number of fissions (1.3×10^{19}/cm^3). This is in good agreement with our first hypothesis. The second one brings us to a vacancy concentration of 8×10^{-3} which is in good agreement with the decrease of density (1%) observed for the same sample[5]. Thus it is impossible to choose between the two hypothesis.

When the crystals are annealed for 1 hour at increasing temperatures (in steps of 25°C), the size of the zone increases: the maximum of the scattering curve (Fig. 2) is displaced

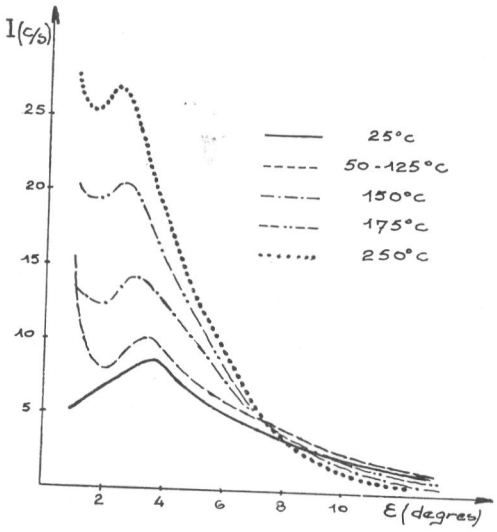

Fig. 2. Displacement of the scattering curve during the annealing of the crystal.

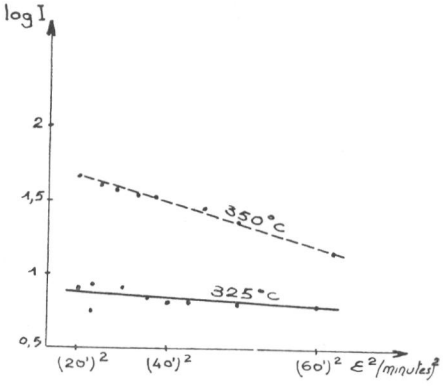

Fig. 3. Small angle scattering given by a crystal annealed for 1 hour at 325°C and 350°C.

to smaller angles. At 325°C a new phenomenon occurs: we observe a much more intense scattering. The scattered intensity has a maximum at the center of the reciprocal lattice, it increases rapidly with the temperature of annealing. At 350°C it can be well represented by the function $\exp(-K\varepsilon^2)$ (Fig. 3). The radius of gyration of the zones giving this scattering is then 28 Å. These results may be correlated to those given by samples irradiated at the pile temperature[6]: we think that cavities also appear and, from quantitative measurements, we find 10^{16} cavities/cm^3, that is to say a vacancy atomic concentration of 1.6×10^{-3}. When the annealing temperature is raised above 400°C, some anisotropy appears in the scattering: the scattered intensity becomes focused along the ⟨100⟩ directions of the reciprocal lattice and we observe the diffraction pictures given by slits parallel to the ⟨100⟩ directions of the crystal (Fig. 4). This pattern is given by cylindrical objects inside the LiF crystal: cavities or metallic lithium. Their diameter varies from 200 Å to 250 Å when the temperature is raised from 500 to 700°C.

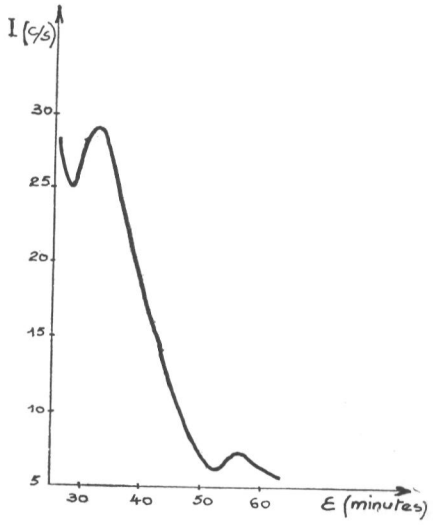

Fig. 4. Scatttered intensity along the [100] axis (crystal annealed for 5 hours at 600°C.).

So we may conclude that compound defects are also formed in low temperature neutron irradiated lithium fluoride crystals when they are brought up to room temperature. These defects are small and their size is well defined. We have also verified that heating the specimen after irradiation does not give the same result as heating it during irradiation; for instance cavities appear by heating above 325°C while they are created even at 100°C during irradiation.

References

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DISCUSSION

Bragg, R. H.: With respect to your statement, that the defect cluster size does not depend upon irradiation but upon annealing temperature. Several years ago Smallman and Willis found, in doing similar experiments to yours but at pile temperature, that there is a size *vs* irradiation dose effect, the size increasing with increasing dose. Of course they also found the size *vs* annealing temperature effect.

Lambert, M.: In previous experiments [M. Lambert and A. Guinier: C. R. Acad. Sci. Paris **244** (1957) 2791] we found, as confirmed independently by Smallman and Willis, that pile temperature neutron irradiation produces clusters widely distributed in size. Their mean size increases with increasing neutron dose.

By contrast in samples irradiated under liquid nitrogen, these clusters only appear after annealing above 300°C and their size are then homogeneous. In these specimens we find, however, at room temperature, new kinds of defects showing a complex structure. The size of these new defects seems to depend not upon neutron doses but only upon the annealing temperature.

Bragg, R. H.: If your defect clusters are very uniform in size then you can use the calculations of Malon (Acta Crystallographica) for comparison and obtain an accurate estimate of defect cluster size and shape for data as in your Fig. 4.

Do you find a lattice parameter *vs* irradiation dose having a maximum in the liquid nitrogen work as in the case of the pile temperature irradiation?

Lambert, M.: No systematic measurements of lattice parameter *vs* neutron doses have been carried out so far. All results reported in the literature (Perio and coworkers, Smallman and Willis) were obtained after pile irradiation. Actually the low temperature results would cover only limited discrete doses due to operational difficulties.

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Electrical Conductivity of Irradiated LiF

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By the study of neutron irradiation on samples of widely different impurity content, we have been able to discriminate between effects on the impurity complexes and on the LiF matrix proper itself.

Furthermore, the control of temperature of irradiation gives some insight on the defect generation itself.

It is concluded that really pure samples, by contrast with impurity compensated samples, are needed before one can get a better and quantitative description of the phenomenon.

Electrical conductivity of LiF, as any other alkali halides, is due to the mobility of positive ion vacancies[1]. In the temperature range where vacancies induced by divalent impurities overnumber the thermodynamic ones, the electrical conductivity of LiF is highly sensitive to the purity of the samples and the differences of behaviour,