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.

PROCEEDINGS OF THE INTERNATIONAL CONFERENCE ON CRYSTAL LATTICE DEFECTS, 1962, CONFERENCE
JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 18, SUPPLEMENT III, 1963

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, after irradiation, for different amount of impurities allow a discrimination between effects on the matrix itself (intrinsic effects) and on the impurity complexes (extrinsic effects).

LiF single crystals have been irradiated with Co^{60} gamma rays, x-rays and reactor neutrons. Measurements are carried out with D.C. from 10^{14} to 10^{7} Ω and A.C. for lower resistance to take care of polarization. All ionizing radiations drastically suppress the conductivity in the impurity range.

This is probably due to stabilization of the $Mg^{2+}Li$ complexes by some electronic species. For sample containing around 10^{-4} divalent impurities, the thermal annealing proceeds without any apparent kinetics as if the recovery of the irradiation effect were connected with the release of a charge carrier in one and a same process. For very pure samples one notes a slight increase of conductivity after irradiation, but the thermal annealing shows alternate random up and down kinetics. We have no explanation at all for such a behaviour.

Monocrystalline samples irradiated with thermal neutrons have been studied with respect to purity, doses and temperature of irradiation. The main effects issue from the

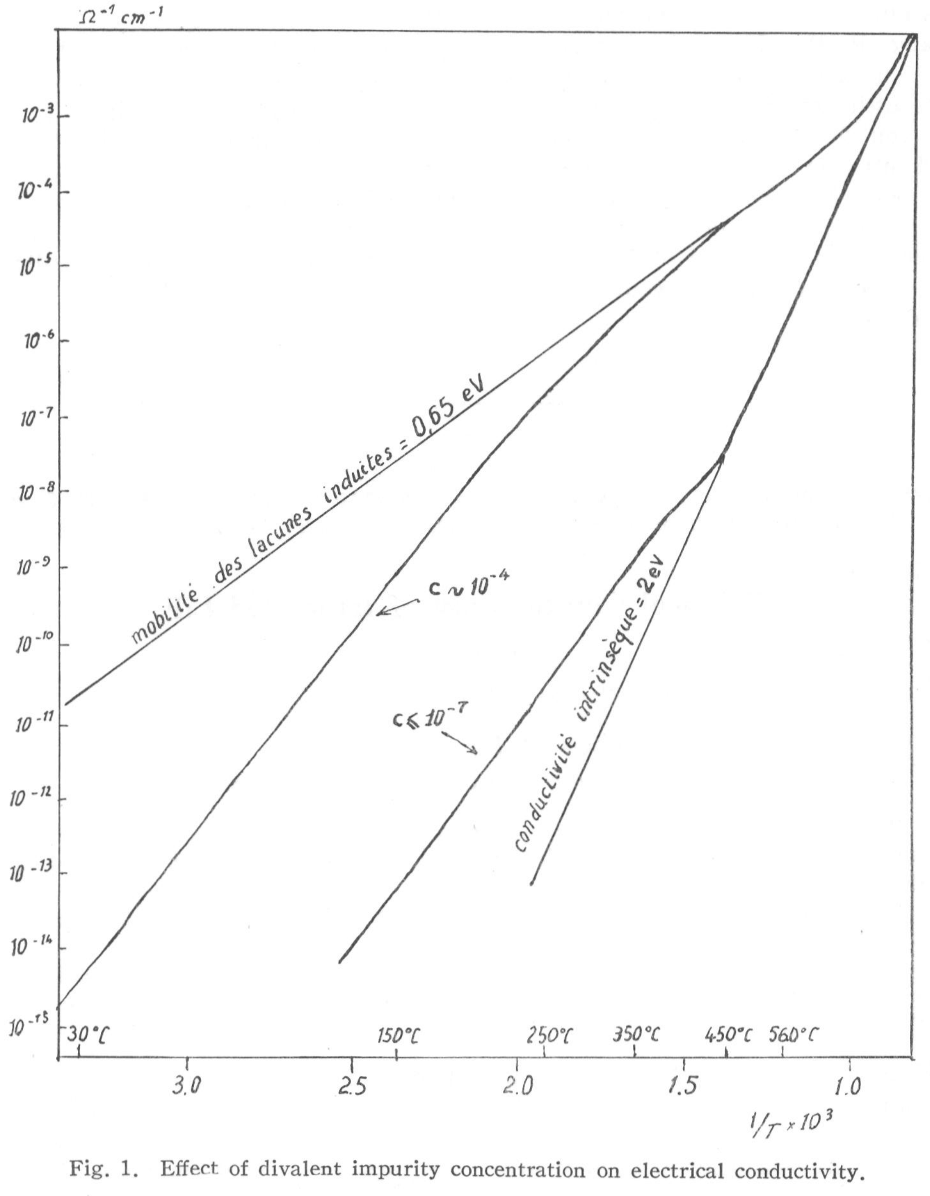

Fig. 1. Effect of divalent impurity concentration on electrical conductivity.

nuclear reaction

$$Li^6 + n \rightarrow He^4 + H^3 + 4 \text{ Mev}.$$

At low doses, ionization effects are predominant and up to 10^{14}-10^{15} nvt, the behaviour is the same as for gamma-irradiation (beside the reactor gamma spectrum, one must remember that most of the energy of the slowing down α- and tritium particles is spent in ionization processes). At higher doses a specific neutron effect shows out for pile temperature irradiations.

There is first[2] creation of a large number of positive-negative vacancy complexes obeying up to 300 °C to a quasi thermodynamical dissociation process, with a binding energy of ~0.7 ev, thus releasing positive vacancies and promoting conduction (The corresponding interstitials have apparently disappeared by formation of fluorine gas inside cavities and two dimension platelets and three dimension

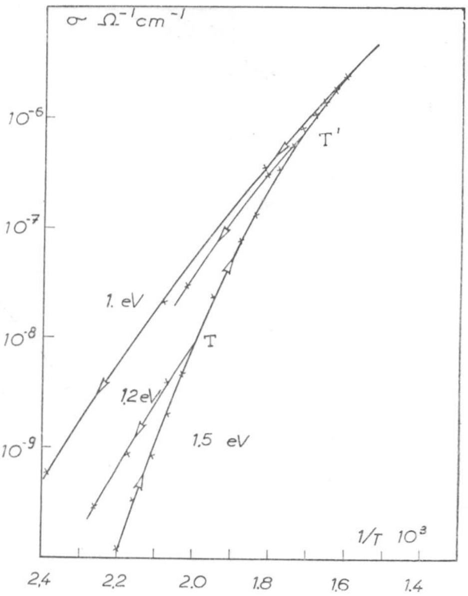

Fig. 2. Behaviour of gamma irradiated LiF.

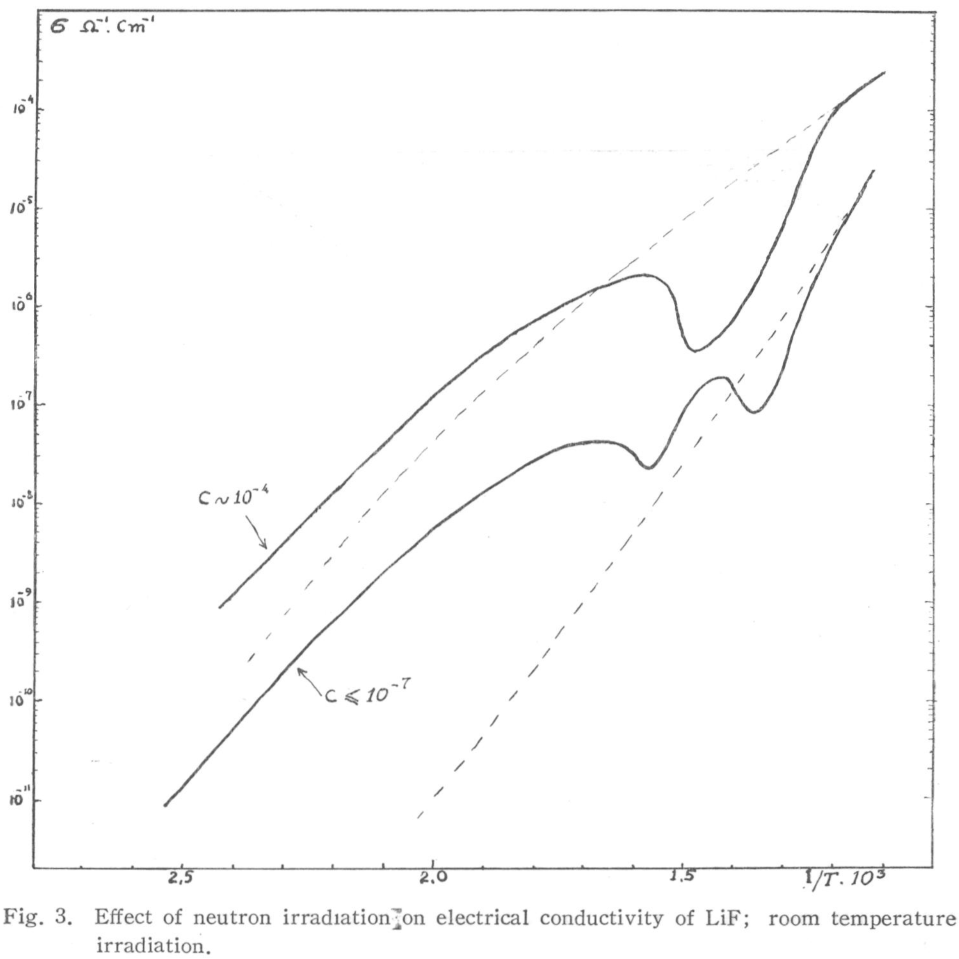

Fig. 3. Effect of neutron irradiation on electrical conductivity of LiF; room temperature irradiation.

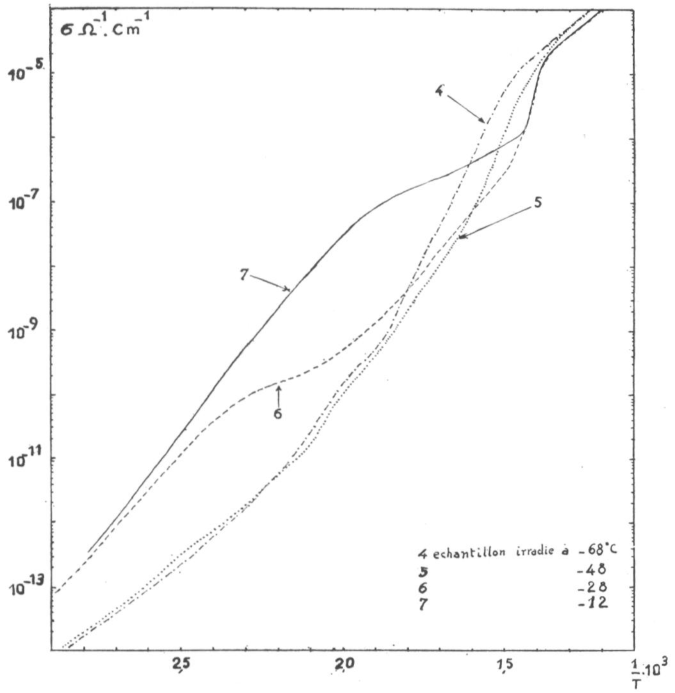

Fig. 4(a). Effect of temperature of irradiation on electrical behaviour of doped LiF.

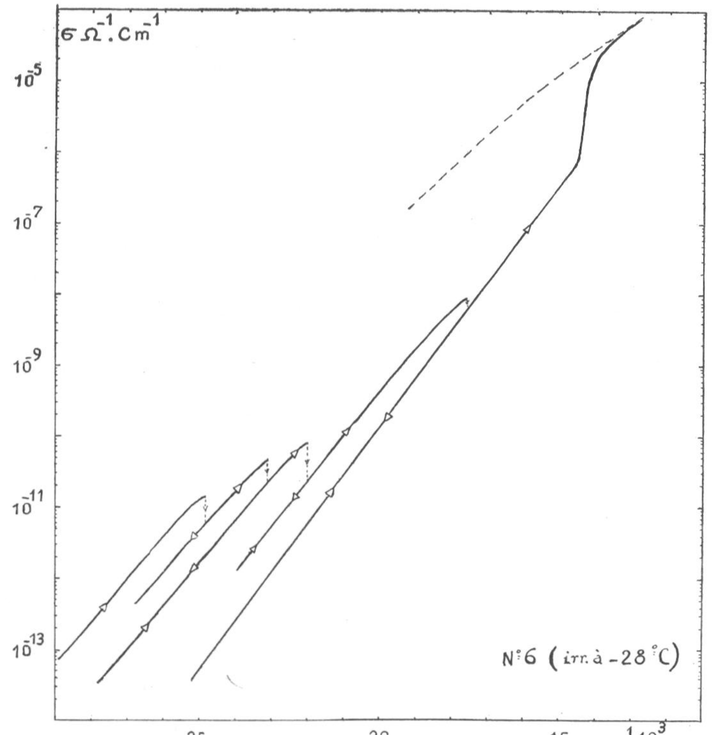

Fig. 4(b). Electrical conductivity of "room temperature" irradiated LiF, after annealing out of "intrinsic" extra carriers.

aggregates of lithium metal[3]). The amount of such centers reaches $\sim 10^{-4}$ for doses around 10^{18} nvt; the conductivity of naturally doped samples is then of the same order of magnitude as before irradiation, whereas for high purity samples, it increases by a factor of nearly 10^3. These centers disappear between 300° and 380°C, probably when negative ion vacancies start to show appreciable mobility. These results are confirmed by dielectric losses experiments[4]).

After thermal regression in this temperature range, pure samples show only intrinsic conductivity, whereas normal samples display a new, well defined, single activation energy (1.35 ev) conductivity domain. If one assumes that the carriers are still positive ion vacancies and their mobility has not been modified by irradiation, this conductivity should be ascribed to a complex associating the Mg^{2+} impurity, a Li vacancy and something else with a very high binding energy: $(1.35-0.65)\times 2 \simeq 1.4$ ev. This defect is associated with heavy colouring of the crystal: bleaching, recovery of conductivity and thermoluminescence emission are associated in the last stage of rapid annealing above 400°C.

The defects responsible for the two effects on the LiF matrix itself and on the divalent impurity complexes are not primary defects of the nuclear reaction or the subsequent slowing down of energetic particles. This is born out by low temperature irradiation experiments which show the influence of some intermediate thermal-activated diffusion processes. Several samples have been simultaneously neutron irradiated at temperature ranging from −195 to −10°C. All samples irradiated below −60°C show the same behaviour reminiscent of gamma-irradiation[5]). The trapping of Mg^{2+} leading both to the 1.35 ev conductivity domain and the last rapid annealing stage above 400°C begins to show only for irradiations above −50°C and there is no hint to creation of negative-positive vacancy pairs at temperature lower than −40°C.

Since optical and thermoluminescence experiments point out to the overwhelming importance of impurities, even in the field of colour centers, the possibility of discriminating between intrinsic and extrinsic effects through the choice of a proper irradiation temperature should be of a great help in further interpretation. However little progress is expected until purer samples become available and the doping is accurately controlled.

References

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