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Limiting processes for the defect accumulation under electron irradiation in KBr at 4 K

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Résumé. — Les cinétiques de croissance de tous les défauts (F, F+, H, H2, I, V) créés dans KBr par irradiation électronique ont été étudiées dans un large domaine de concentration (10^{16} \cdot 5 \times 10^{19} \text{ cm}^{-3}), à 4 K, température à laquelle aucun des défauts n’est thermiquement mobile. Les lois de croissance sont interprétées en considérant la limitation du rendement de création des nouvelles paires de Frenkel par les interstitiels stabilisés. A très haute concentration, la possibilité de recombinaison des centres H, lors de leur déplacement sous forme de crowdion avec les centres F présents, entraîne une nouvelle diminution du rendement d’accumulation des centres F. Nos résultats suggèrent, en outre, une limitation indépendante de la croissance des centres F et F+, donc des processus de formation indépendants.

Abstract. — The growth kinetic in a wide range of concentrations (10^{16} \cdot 5 \times 10^{19} \text{ cm}^{-3}) of all defects (F, F+, H, H2, I, V) created in pure KBr by electron irradiation have been studied at 4 K, temperature at which no defect is thermally mobile. The kinetic shapes are explained by considering a local action of the stabilized interstitial centers decreasing the creation yield of new Frenkel pairs. At concentration higher than 10^{19} \text{ cm}^{-3}, the decrease of the F center accumulation yield is due to the possibility of uncorrelated recombinations of the F centers and the H centers moving as dynamical crowdions. Our results suggest also an independent limitation of F and F+ center growth, thus independent formation processes.

1. Introduction. — In the alkali halides, the kinetics in the temperature range where no center is thermally mobile are of interest to study elementary phenomena of defect creation and accumulation without screening by thermal activated secondary reactions. In KBr, two kinds of Frenkel pairs, neutral F-H and charged F+I, are created. The transformation F-H → F+I which efficiency might depend on the defect concentration in the crystal [1] is not yet well understood. This paper’s purposes is to present an experimental study of the kinetics of all centers present under electron irradiation in KBr near 4 K and to analyse the kinetic shapes by simple models based on local action of centers inhibiting the creation of new Frenkel pairs. All results have been obtained using nominally pure KBr crystals purchased from the Harshaw Chemical Co. The apparatus has been described elsewhere [2]. The 20 to 60 keV electron beam having a density between 0.1 and 20 μA cm^{-2} creates defects in a thin layer (5.7 μ at 20 keV to 36.9 μ at 60 keV) at the surface of the crystal. The energy deposition rate $\dot{\varepsilon}$ lies between 10^{19} and 2 \times 10^{22} \text{ eV cm}^{-3} \text{ s}^{-1}. Varying the electron beam energy allows concentration measurements between 10^{16} and 5 \times 10^{19} \text{ cm}^{-3}, with optical densities (OD) smaller than 3 at the peak of the absorption bands. We have used results from ref. [3]

1) to determine the respective contribution of the V and H centers to the 380 nm absorption band.

2) to take into account the reaction F+ + e^- → F under irradiation. The concentration of V centers has been experimentally determined to be 6.5% of the sum of the F and F+ center concentrations. This amount corresponds to the number of F+ centers having trapped an electron, and transformed into F centers. So, to have the concentrations of centers at the equilibrium, we must substract it from the number of F centers and add it to the number of F+ centers. All our results will be corrected in this way.

2. Experimental results. — Experimental accumulation kinetics have been measured as a function of the dose for all centers. They depend only on the dose received by the crystal. No dependence on the energy deposition rate $\dot{\varepsilon}$ has been evidenced in the range allowed by the apparatus.

The main results are following.

— The total concentration of defects grows monotonically as a function of the dose.

Figure 1 shows the F and F+ growth kinetics as a function of the dose. A typical logarithmic growth is pointed out in the dose range lower than a few
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10^{22} \text{eV cm}^{-3} \text{ (zone I). For higher doses (zone II), the F center concentration grows linearly versus dose (Fig. 2), whereas the F' center concentration is always logarithmic. At concentrations higher than 10^{18} \text{F centers cm}^{-3} \text{ (zone III), a square root law is observed for the F centers as a function of the dose (Fig. 3). F' centers have not been measured in this concentration range. A simple phenomenological representation of F and F' center growth kinetics in zones I and II is given by the relation:}

\[ x = A \ln \left( \frac{d}{B} + 1 \right) + Cd \]  

where \( x \) is the defect concentration, \( d \) the dose, \( A, B \) are phenomenological parameters.

If the kinetics of the F' center and the complementary I center are exactly proportional this is not the case for the kinetics of the F center and the H center. H centers are known to undergo a strong attractive interaction to form H2 and more complex aggregates [4]. So, the complementary behavior of the H, H2 and F centers must be considered simultaneously. The H centers saturate at a value close to 10^{18} \text{cm}^{-3} and the H2 center concentration versus F center concentration shows a quadratic growth in zone I. This curve is explained in the same way as the quadratic relation found by Itoh and Saidoh for the H and H2 centers [5].

Taking into account the quadratic relation

\[ n(H)/n(F) = 1 - 2k n(F), \]

as shown in figure 4.

We can determine the factor \( k \), in good agreement with [5]. At higher F center concentration, more complex aggregates are formed.
3. Model of limiting processes for the defect accumulation. — A model for very low temperature growth kinetics of defects must take into account two experimental evidences:

1) no center is thermally mobile [6],
2) the Frenkel pairs are correlated.

The first point indicates that one must consider local properties to explain the decrease of the Frenkel pair creation efficiency. A mathematical representation for it is the assumption of a forbidden volume, where the Frenkel pair cannot be formed, around some types of centers [7]. The second point can be deduced from first order recombination kinetics of neutral Frenkel pairs observed both after a pulsed irradiation in KBr at 8 K [8] and during thermal recombination stages [9]. Our experimental results in zones I and II are well fitted (Fig. 2) with the following hypothesis.

a) The I-center is the main responsible center for the forbidden volume. (i) The perturbation caused by an I center in its neighborhood is known to be more important than the one caused by the other centers [10-13]. (ii) The inhibition mechanism is due to the strain field around the I center causing the recombination of a newly formed Frenkel pair, on the basis of analogies with other cases of lowering the pair creation efficiency by large dopants such as $I^-$ [14] or $Rb^+$ [15] in KBr.

b) The recombination occurs after the charge transfer from F to the H center resulting in $F^+$ and I centers [8, 16]: the F and $F^+$ center kinetics have to be described independently.

So, using the arguments of [7], we can write:

$$n(F^+) = \frac{1}{v} \ln (R^+ vd + 1)$$

(5)

and

$$n(F) = \frac{R}{R^+ v} \ln (R^+ vd + 1) + R_0 d \quad R = R' - R_0$$

(6)

with $v$: forbidden volume associated to the I center, $R^+$ and $R'$: the initial formation rate of $F^+$ and F centers respectively, and $R_0$: the residual formation rate for F centers in the forbidden volume.

The F center kinetics in zone II show that there is a residual formation rate in the forbidden volume. The 1/2-power law observed in zone III is consistent with this assumption, if we expect in zone III an important role of the decorrelated recombinations of Frenkel pairs, as in the case of temperatures where only the interstitials are thermally mobile (e.g. LiF at room temperature, KBr at 77 K) [17-19], due to the range of the dynamic crowding in the lattice when an interstitial is formed. So, following [17-19], we must have

$$n(F) \approx \left( 2 R_0 \frac{\sigma_T n(T)}{\sigma_F} \right)^{1/2} \left( \frac{\sigma_T}{\sigma_F} \right)$$

(7)

where $\sigma_T$, $\sigma_F$ are respectively the capture cross section of the interstitial by an F center and a interstitial aggregate (number n(T)). From data given in zone II and III and the assumption that the concentration n(T) is about $10^{18}$ cm$^{-3}$, i.e. that all H centers at saturation are nucleation germs, we find $\sigma_T/\sigma_F \approx 5$ which is not very different from the ratio of the geometrical sections of $H_2$ and F centers.

DISCUSSION

Question. — N. ITOH.

Have you tried to obtain the growth curve at 20 K, where the I center is substantially unstable. Such an experiment would prove the validity of your model of the inhibition of the F-H creation in the vicinity of the I center.

Reply. — A. NOUAILHAT.

Yes, but not exactly in this manner: we compare the vacancy creation rate at 4 K for the same amount of vacancy concentration in KBr irradiated in 4 K or with a previous irradiation at 77 K. They are different in the second case, the creation rate is about the same as for the original crystal. This confirms the role of I centers.

Question. — F. AGULLO-LOPEZ.

You are assuming that all defects are thermally stable. However, I am wondering whether defect mobility could occur under irradiation and lead to defect reactions and annihilation.

Reply. — A. NOUAILHAT.

No; I have no evidence of mobility of defects occurring under irradiation at 4 K.

References

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