THE FORMATION OF DISLOCATION LOOPS AND THE OUTGROWTH OF CRYSTALLITES BY ELECTRON IRRADIATION OF THIN ALKALI HALIDE FOILS

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Résumé. — Il a été observé que des boucles de dislocation de type interstitiel et de type lacune se forment dans les lames KBr pendant l'examen au microscope électronique a transmission 100 kV. Les examens ont été faits à une température entre + 20 et — 110 °C. On a aussi observé que de petits cristaux croissent à une surface des lames quand les lames sont irradiées à — 110 °C avec des électrons.

Abstract. — Dislocation loops of interstitial and vacancy types were observed to form in kBr foils during examinations in 100 kV transmission electron microscope. The examinations were carried out in the temperature range between + 20 and — 110 °C. Small crystallites were also observed to grow on a surface of foils, when the foils were heavily irradiated at — 110 °C with electrons.

1. **Introduction.** — When a (001) foil of KCl single crystal is examined in a transmission electron microscope, four kinds of defect clusters are observed to form in the crystal by electron irradiation, as reported in my previous papers [1, 2, 3]. In these work, examinations were carried out at a temperature between room temperature and – 110 °C. And these kinds of clusters were divided into two categories. One kind of clusters formed at any temperature between them and the others formed only around room temperature. The former clusters are common to many species of alkali halides, namely, to KCl [2], KBr [4], KI [4], NaCl [5], NaBr [4] and LiF [6]. This kind of clusters were first observed by Tubbs and Forty in KCl as complex images [7], by Kawamata and Hibi in KCl [1], and by Mannami *et al.* in LiF [8] as dots. In well-resolved manner, the clusters were observed by Bethge *et al.* in NaCl [5] and by Dworschak and Waidelich in LiF [6]. By the electron images of the clusters, they supposed the clusters were prismatic dislocation loops. Afterwards, two other models were proposed for this kind of clusters with bases for reasoning. These models are alkali metal colloids [9] and aggregates of halogen molecules [4] which were proposed by Izumi and by Hobbs *et al.* respectively. These models were mainly based on the results obtained by that time in the field of color centers. However, Kawamata obtained a direct evidence to show that the clusters are prismatic dislocation loops having Burgers vectors of the type $a/2$ [011] [2]. Namely, it was observed in KCl that the clusters moved jerkily or oscillated violently in [011] *directions* under heavy irradiation and that the clusters were broken up to segments of linear defects which terminated at the foil surfaces. In general, the only linear defect in a crystal that should move easily under the action of a shear stress is a dislocation. Therefore, if a defect is shown to be both linear and readily moved by a shear stress it is almost certainly a dislocation. This test has been described above.

One of our next problems is to know whether the dislocation loops are of interstitial type or of vacancy one. The present work concerns this problem. In KBr, I found that the images of the loops made drastic changes when irradiation temperature was made lower from room temperature to – 110 °C. Namely, the loops showed small additional dislocation loops inside their periphery. As described later, this suggests that the original loops are of interstitial type and that small additional loops are of vacancy type. This agrees with Hobbs' results that the original clusters are of interstitial type [4].

It has been described above that the dislocation loops are perfect. This means that the loops consist of equal number of interstitial or vacancy of both anion and cation sub-lattices. Therefore, interstitials and vacancies must be created by electron irradiation not only in halogen sub-lattice but also in alkali metal sub-lattice. Scott and Crawford have also reached the same conclusion in a study of electric dipoles in KCl created by high 1.5 MeV electron bombardment [10]. However, the experimental results which support this conclusion are few. This paper will describe another basis of the conclusion. It is the growth of small crystallites of KBr on the surface of a KBr foil irradiated by electrons. This is concerned with the volume expansion of alkali halide which is the most striking effect of irradiation. Concerning to this effect, Hibi and Ishikawa have shown that X-irradiation of alkali halides causes the growth of separate crystallites on the surfaces of irradiated original crystals [12]. They observed the crystallites by electron microscopy using replica technique. In this work, direct observation of growth of crystallites on a surface of a KBr foil irradiated by electron were carried out.

2. **Experimental.** — The single crystals of KCl, KBr, KI and NaCl using in the present work were obtained from the Harshaw Chemical Company. Thin foils were prepared by a method reported in previous papers [1, 2, 3] for transmission electron microscopy. Namely, crystals were cleaved in (001) planes into plates 0.1 to 0.3 mm thick and thinned by dissolution in water. In this way, thin foils nearly parallel to a (001) plane were obtained. The foils were examined at a temperature between – 110 and + 20 °C using 100 keV incident electrons in a HU-11 B electron microscope equipped with a liquid-nitrogen cooled stage. The beam current density ranged from 0.1 mA/cm^2 to 1 A/cm^2 on a specimen and the diameter of the beam was about 20 μm at a specimen.

3. **Results and discussion.** — 3.1 GROWTH OF DISLOCATION LOOPS OF INTERSTITIAL TYPE AND VACANCY TYPE. — Figure 1 is an electron micrograph of defect clusters produced in KBr by electron irradiation during examination at 23 °C. The beam current density is low and about 1 mA/cm^2 at the specimen. The defect clusters are seen as nearly rectangular loops.

With decreasing irradiation temperature, these loops were observed to decrease in size and to increase in number. In addition, damage patterns began to make drastic changes at about 0 °C. These changes are shown in figure 2 which is a micrograph of a specimen irradiated at – 110 °C. We find that each of loops has a small additional loop inside it. The outer and inner loops are denoted by I and V in figure 2.

The next experiment was carried out as follows. First, large I-loops were introduced into a specimen by irradiation at 20 °C and then the specimen was slowly cooled to – 110 °C and re-irradiated. By this re-irradiation small V-loops were generated at the inner periphery of the large I-loops, as shown in the micrograph of figure 3. In figure 3, small V-loops are seen as dots.

Figure 4 is an electron micrograph of KI irradiated at – 110 °C. Small V-dots are seen at the inner periphery of I-loops. Similar phenomena were also observed in KCl and NaCl but they are not so clear.

During examinations, I- and V-loops were observed to move jerkily or oscillate in < 011 > directions as observed in KCl [2]. The movements of loops became violent, when irradiation temperature was low, when

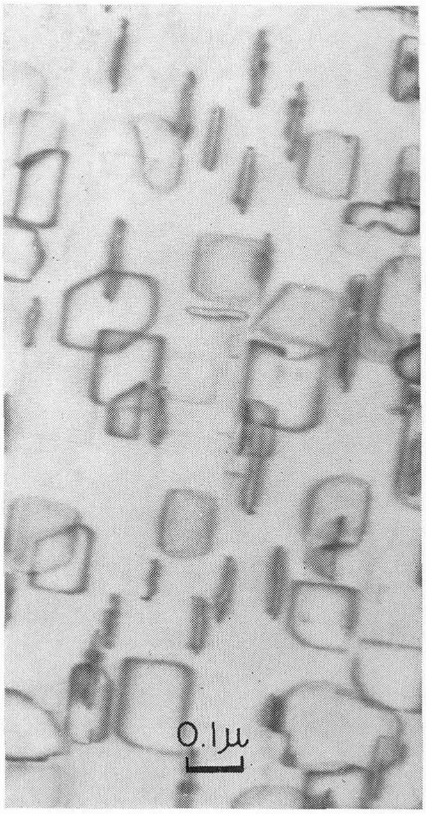

FIG. 1. — Interstitial dislocation loops in a KBr foil irradiated at 23 °C with 100 keV electrons at 1 mA/cm^2.

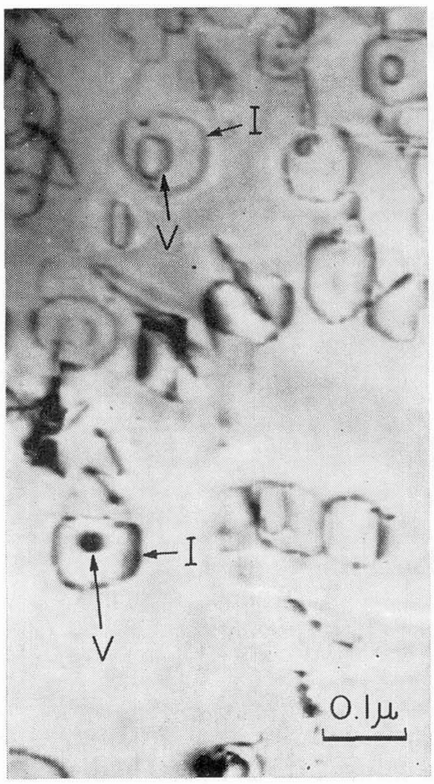

FIG. 2. — Combined interstitial-vacancy dislocation loops in a KBr foil irradiated at — 110 °C with 100 keV electron at about 0.1 mA/cm^2.

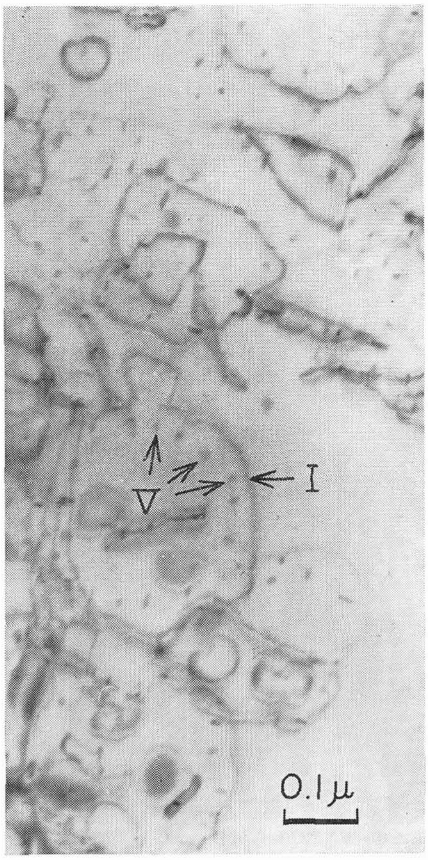

FIG. 3. — A row of small vacancy dislocation loops along the inner periphery of a large interstitial dislocation loop in an electron-irradiated KBr foil. First, the interstitial loops were produced by irradiation at 20 °C and then the vacancy loops, at — 110 °C, at 0.1 mA/cm² in both cases.

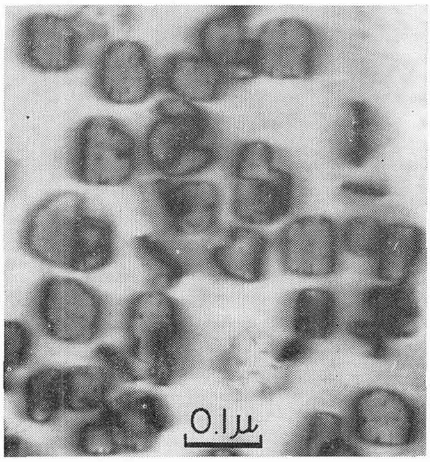

FIG. 4. — Large interstitial dislocation loops and small vacancy dislocation loops inside the large loops in a KI foil irradiated at — 110 °C by 100 keV electrons at 0.1 mA/cm².

beam current density was high, and when the size of the loops was small. At — 110 °C, these small V-loops readily moved inside the large I-loops or passed out of the foil, even under light irradiation. Therefore, the specimens must have been irradiated very carefully at very low beam current densities in order to take the micrograph of small V-loops.

These movements of the defect clusters are consistent with prismatic dislocation loops moving conservatively on their glide cylinder defined their perimeter and Burgers vector of the type $a/2$ [011].

Combined images of large and small loops had already been observed by Urban during the irradiation of nickel foils with 650 keV electrons in a high-voltage electron microscope [11]. His conclusion is that the outer loops are interstitial dislocation loops and inner loops are vacancy ones. The preferential growth of interstitial loops takes place because the elastic-size interaction of a dislocation with an interstitial is stronger than with a vacancy. For the growth of vacancy loops, the spacial separation of interstitials and vacancies by elastic field around an edge dislocation is necessary. The inner periphery of interstitial loop is the compression side. The density of vacancies increase at this side. Therefore, this side is the most favourable site for the nucleation of every type of vacancy agglomerates. It is thought that the present observation in KBr is explained by the same way. This explanation agrees with Hobbs' results that defect clusters created in KI are of interstitial type [4].

3.2 GROWTH OF SMALL CRYSTALLITES ON THE SURFACES OF A FOIL. — Figure 5 is an electron micrograph of a KBr foil irradiated at — 110 °C. The beam current

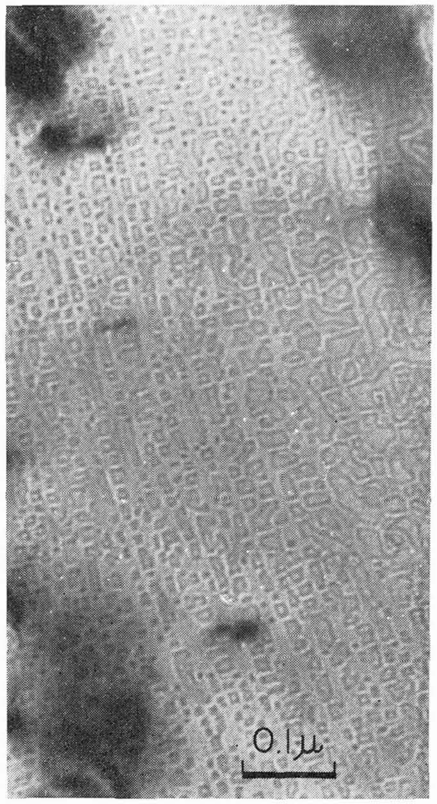

FIG. 5. — Small crystallites on the surface of a KBr foil irradiated at — 110 °C by 100 keV electrons at 1 mA/cm².

density was 1 mA/cm^2. The growth of small crystallites are seen all over the area in addition to the strong-contrast images due to dislocation loops. These crystallites are rectangular in their images and the edge of the crystallites are oriented in [001] directions of the original crystal. Another striking feature is that the crystallites form along the steps of the surface of the foil. The micrograph of figure 6 clearly shows steps decorated with small crystallites. This means that the crystallites grew on the surface. In addition, the

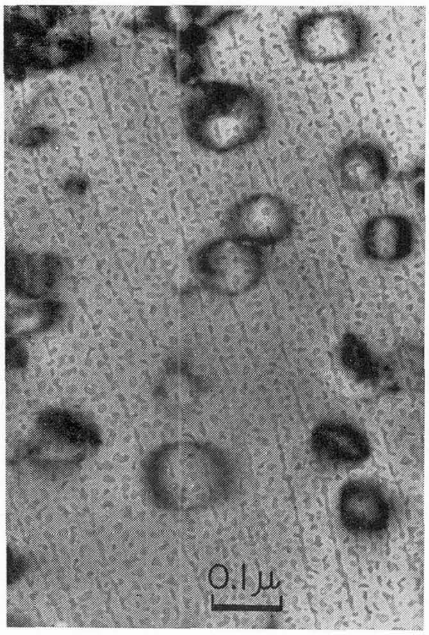

FIG. 6. — Steps decorated with small crystallites on the surface of a KBr foil irradiated at — 110 °C by 100 keV electrons at 1 mA/cm^2.

crystallites yielded no distinct indication in electron diffraction patterns in spite of the growth of many crystallites. Therefore, it is thought that the crystallites are the results of outgrowths of the original crystal. Namely, the crystallites consist of KBr and are coherent to the original crystal.

On the other hand, in figures 5 and 6, it appears that small crystallites form only at one surface of the foils. Overlapped images of crystallites formed at upper and lower surfaces have not been observed throughout this work. In the initial stage of irradiation, two series of steps belonging to upper and lower surfaces could be seen, as shown in figure 7. However, after irradiation, only one series of steps decorated with small crystallites were observed, as shown in figure 6. This aspect of the problem has not yet been completely investigated. But it is supposed that crystallites did

FIG. 7. — Two series of steps on surface of a KBr foil.

not grow at the lower surface because it was contaminated during irradiation.

4. **Conclusion.** — Dislocation loops of interstitial type and of vacancy type were observed to form in KCl, KBr, KI and NaCl which were irradiated at – 110 °C by electrons. In addition, the outgrowth of small crystallites were also observed on a surface of an irradisted KBr foil. Interstitial dislocation loops and crystallites are agglomerates of an equal number of interstitials of alkali metal and halogen sub-lattices and vacancy dislocation loops are agglomerates of an equal number of vacancies of both sub-lattices. Therefore, interstitials and vacancies must be created in pairs by electron irradiation not only in halogen sub-lattice but also in alkali metal sub-lattice.

In the absence of dislocations, either grown-in dislocations or dislocation loops produced by irradiation, production of point defects takes place in both sub-lattices, because small crystallites were observed to grow in the crystal foils lacking dislocations.

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