the melting point, quenched quickly in air to room temperature, and then irradiated for one hour at 40 kv and 20 ma with a Machlett AEG50 x-ray tube with a tungsten target and beryllium window. The crystals were then uniaxially compressed in the dark at 8.1 mm/min. The resulting light emission was analyzed by using a Hilger E517 quartz spectrograph. Fifty crystals were used in obtaining the spectrum in Fig. 1. Although no sensitivity vs wavelength calibration was made on the plates, the position of the maximum was found to be the same on three quite different emulsions, namely Kodak 103-F, 103-O, and 1-N Plates.

Since the application of stress to x-ray colored crystals can result in bleaching,<sup>2-4</sup> a reasonable explanation for the observed luminescence may be that trapped electrons are freed by the motion of dislocations during compression<sup>5,6</sup> and then are annihilated by recombining with holes present in the crystal, resulting in the emission of light. If such is the case, then the nature of the luminescence would be primarily dependent on the halogen involved, in that the recombination is analogous to the electron affinity process. It is hoped that an investigation of a series of alkali halides now in progress will elucidate the mechanism.

\* This work was performed under the auspices of the U.S. Atomic Energy Commission.

<sup>1</sup> F. I. Metz *et al.*, J. Phys. Chem. **61**, 86 (1957).
<sup>2</sup> A. Smekal, Z. Ver. deut. Ing. **72**, 667 (1928).
<sup>3</sup> H. J. Schröder, Z. Physik **76**, 608 (1932).

<sup>4</sup> I. S. Jacobs, Phys. Rev. **93**, 993 (1954). <sup>5</sup> F. Seitz, Phys. Rev. **80**, 239 (1950).

<sup>6</sup> W. W. Tyler, Phys. Rev. 86, 80 (1952).

## Evidence for Interstitials in Low-**Temperature X-Ray** Irradiated LiF<sup>†</sup>

D. A. WIEGAND AND R. SMOLUCHOWSKI

Carnegie Institute of Technology, Pittsburgh, Pennsylvania (Received April 2, 1958)

**)** Y a modification of the photoelastic technique of Primak, Delbecq, and Yuster,<sup>1</sup> the volume expansion and annealing of x-ray irradiated LiF has been studied at low temperatures. One half of a thin rectangular plate is irradiated in a direction normal to the plane of the plate in such a manner as to produce electronic ionization throughout it, while the other half is shielded from the radiation. The constraint placed upon the irradiated portion of the crystal by the unirradiated portion at the boundary hampers the expansion of the irradiated part and so introduces strain into both halves of the crystal. The crystal then becomes birefringent and the strain pattern can be analyzed by the use of polarized light. In the elastic range the volume expansion is directly proportional to the angular rotation of the plane of polarization, and

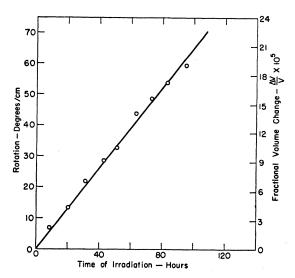


FIG. 1. Rotation of the plane of polarization and volume expansion as functions of time of x-ray irradiation at 94°K. (Crystal LiF No. 18.)

can be calculated if the elastic and photoelastic constants are also known.<sup>1,2</sup>

In Fig. 1 the volume expansion is given as a function of time of x-ray irradiation at 94°K. The crystal is seen to expand linearly in the range of  $\Delta V/V$  from roughly  $10^{-5}$  to  $10^{-4}$ . The F band was also found to increase linearly over a shorter range of exposure, i.e., up to 5.5 hours of irradiation. The optical absorption of the Fband cannot be easily measured by conventional apparatus at higher exposures. However, the time rate of expansion obtained from the F-center growth curve by assuming a volume expansion equal to the volume of one negative-ion vacancy per F center is roughly that obtained from the volume expansion curve of Fig. 1, thus indicating that F centers are produced at a constant rate throughout the whole irradiation period. In contrast to this behavior, a band at 3400 A saturates after only a short irradiation. In Fig. 2 the annealing of the volume expansion (as given by the rotation per cm of the plane of polarization), the F band, and the 3400-A band are given as the crystal is warmed from 94°K. All three quantities are seen to anneal appreciably in roughly the same temperature interval.

The annealing of the volume expansion between 110° and 160°K cannot be due to diffusion of single positiveor negative-ion vacancies since the activation energies are much too high. The probability for single jumps of vacancies or interstitials may be high enough, however, to account for this annealing, for instance in terms of close pair vacancy-interstitial recombination. Diffusion of vacancy pairs, interstitials, or crowdions are other possibilities to be considered. The data of Fig. 2 indicate that while  $\Delta V/V$  anneals by roughly a factor of  $\frac{3}{4}$ , the F band anneals by only  $\frac{1}{3}$ . In addition, measurements of the growth of the F band at 94°K after the crystal has been irradiated at 94°K, warmed to 160°K, and

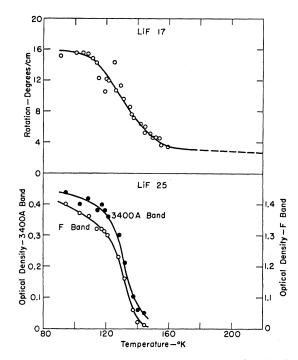


FIG. 2. Annealing of volume expansion of LiF No. 17 and optical absorption bands of LiF No. 25 with rise in temperature after irradiation at 94°K.

recooled to 94°K indicate that the density of negativeion vacancies is not decreased but that the F band anneals due to loss of electrons, probably to holes from the 3400-A band which has recently been associated with the molecule-ion center by the Argonne group.<sup>3</sup> It therefore seems that the annealing cannot be due to any processes involving negative-ion vacancies such as halide vacancy-interstitial recombination or diffusion of vacancy pairs. Since positive-ion vacancies do not diffuse, the annealing must be of necessity involve interstitials if it is assumed that only vacancies and interstitials can be generated by the x-ray irradiation. It is then reasonable to suppose that the interstitials are generated by a mechanism such as that suggested by Varley.<sup>4</sup> This conclusion applies to proton irradiation, since in this case also the ionization causes most of the lattice defects. The annealing of  $\Delta V/V$  may then be due to alkali vacancy-interstitial recombination or to processes involving only interstitials such as clustering.

A number of holes equal to the number of electrons trapped in negative-ion vacancies must be trapped elsewhere in the crystal. In addition to impurities, positive-ion vacancies and interstitial halides are possible hole traps. However, while the magnetic resonance spectrum to be expected from a hole trapped at a positive-ion vacancy has yet to be observed, Känzig and Woodruff<sup>5,6</sup> have observed a paramagnetic center, the H center, which could be associated with an interstitial halide in a crowdion configuration. This

center has been found to anneal somewhere between 80° and 130°K, or roughly in the same temperature range as the volume expansion.<sup>3</sup> It is therefore possible that the annealing of  $\Delta V/V$  occurs because the interstitial halide in a crowdion configuration moves to a body-centered interstitial position. Measurements are now being made to determine if in KCl, as in LiF, the volume expansion anneals out appreciably at the same temperature as the H center.

† Research supported by a U. S. Atomic Energy Commission contract.

<sup>1</sup> Primak, Delbecq, and Yuster, Phys. Rev. 98, 1708 (1955). <sup>2</sup> A more complete discussion of the methods and results will be published shortly.

 <sup>3</sup> W. Känzig (private communication).
<sup>4</sup> J. H. O. Varley, Nature 174, 886 (1954).
<sup>5</sup> W. Känzig and T. O. Woodruff, Phys. Rev. 109, 220 (1958). <sup>6</sup> W. Känzig and T. O. Woodruff, Bull. Am. Phys. Soc. Ser. II,

3, 61 (1958).

## Possible Explanation for Mosaic-Like **Bragg Reflection in Quartz** under Strain\*

## DWIGHT W. BERREMAN

Department of Physics, University of Oregon, Eugene, Oregon (Received March 31, 1958)

T is a well-known fact that a quartz crystal under strain tends to reflect x-rays, especially hard x-rays, as if the crystal were of mosaic structure, while it becomes very nearly ideally perfect when the strain is small.1-3 This effect may be due to some sort of reversible dislocations in the crystal. The reversibility of any such dislocations would have to be such as to allow for the extremely perfect elasticity of quartz in all types of strain from static flexure to ultrahigh-frequency oscillations.

The available evidence does not seem to preclude an alternative explanation based on very small accidental curvatures in Bragg planes, which avoids the necessity of postulating any dislocations within the crystal. Suppose the reflecting Bragg planes in a crystal have, locally, a radius of curvature R in the plane of the incident and reflected x-ray propagation vectors. Then a characteristic length S can be defined as that distance, parallel to the Bragg planes, such that the sagitta of an arc of length S in the curved Bragg planes is half the Bragg spacing, d. The relation between S, R, and d is  $S^2 = 4Rd$  (Fig. 1).

The length S so chosen is approximately the length over which coherent interference can occur from parallel curved crystal planes. Thus, if S is somewhat less than the primary extinction distance,4,5 the crystal will reflect as if it consisted of many independent "thin" layers, that is, like a mosaic crystal. In that case, reflectivity is independent of S. On the other hand, if S is somewhat larger than the primary extinction distance, and if the total crystal thickness is also larger,