

## THE PINNING OF DISLOCATIONS BY X-IRRADIATION OF ALKALI HALIDE CRYSTALS\*

R. B. GORDON† and A. S. NOWICK‡

A study is made of the effect of X-irradiation on the room-temperature elastic modulus ( $s_{11}^{-1}$ ) of NaCl crystals. The modulus is observed to be unchanged by irradiation for well-annealed crystals, but may increase by as much as 7% when cold-worked crystals are irradiated. It is demonstrated that the modulus change on irradiation corresponds exactly to the elimination of the modulus decrease due to oscillating dislocation loops through the creation of pinning points along the dislocations. A quantitative theory is developed for the variation of modulus with X-ray dose. This theory assumes that vacancies, released within the volume of the crystal through the action of the radiation, migrate to dislocations and contribute to the formation of pinning points. The theory appears to be in good agreement with experiment and makes possible a calculation of the density of dislocations and the mean length of the free dislocation loops prior to irradiation.

### LE BLOCAGE DES DISLOCATIONS PAR IRRADIATION X DES CRISTAUX d'HALOGENURES ALCALINS

L'effet d'une irradiation X sur le module d'élasticité ( $s_{11}^{-1}$ ) de cristaux NaCl à température ambiante est étudiée. Les observations montrent que le module reste inchangé par l'irradiation pour des cristaux bien revenus, mais peut augmenter jusqu'à 7% de sa valeur quand des cristaux écrouis sont irradiés.

Il est démontré que la variation du module lors de l'irradiation correspond exactement à l'élimination de la décroissance du module due aux anneaux de dislocation oscillants, lors de la création de points d'ancrage le long des dislocations. Une théorie quantitative est développée pour la variation du module avec la dose des rayons X. Cette théorie admet que les lacunes, libérées dans le volume du cristal sous l'action de la radiation, migrent aux dislocations et contribuent à la formation de points d'ancrage.

La théorie semble être en bon accord avec l'expérience et rend possible un calcul de la densité des dislocations et la longueur moyenne des anneaux libres de dislocations avant l'irradiation.

### VERANKERUNG VON VERSETZUNGEN DURCH RÖNTGENBESTRAHLUNG VON ALKALI-HALOGENID-KRISTALLEN

Von den Autoren wurde an NaCl-Kristallen eine Untersuchung über den Einfluss von Röntgenbestrahlung auf den bei Raumtemperatur gemessenen Elastizitätsmodul ( $s_{11}^{-1}$ ) vorgenommen. Dabei wurde beobachtet, dass der Modul gut ausgeglühter Kristalle bei Bestrahlung unverändert, dass er jedoch um 7% ansteigen kann, wenn kaltbearbeitete Kristalle bestrahlt werden. Wie gezeigt wird, entspricht die Moduländerung infolge der Bestrahlung genau dem Abbau der von schwingenden Versetzungsschleifen herrührenden Modulabnahme, die dadurch rückgängig gemacht wird, dass entlang der Versetzungen Verankerungspunkte erzeugt werden. Für die Veränderung des Moduls mit der Röntgendosis wurde eine quantitative Theorie entwickelt. Dabei ist angenommen, dass Leerstellen, die durch die Bestrahlungswirkung im Kristallinnern freigeworden sind, zu den Versetzungen wandern und dort zur Bildung von Verankerungsstellen beitragen. Die Theorie scheint in guter Übereinstimmung mit den Experimenten zu stehen. Sie ermöglicht eine Berechnung der Versetzungsdichte und der mittleren freien Länge der Versetzungsschleifen vor der Bestrahlung.

## 1. INTRODUCTION

Irradiation of an alkali halide crystal with X-rays usually results in the formation of visible color within the crystal, related to the creation of certain optical absorption bands. The most prominent of these bands is the "*F* band" which is due to the presence of so-called "*F* centers" generated in the crystal during the irradiation. It is now known<sup>(1)</sup> that an

*F* center consists of an electron trapped at a negative-ion vacancy. § The number of *F* centers formed in

§ In an ionic crystal a negative-ion vacancy carries an effective positive charge. Thus, when an electron is released into the conduction band by absorption of ionizing radiation in the crystal, it may be attracted to the vacancy where it can fall into a series of bound states. Absorption of light in the *F* band occurs when this trapped electron is raised from the ground state to the first excited state. When a colored crystal is illuminated with light in the *F* band, the color slowly fades ("optical bleaching") because electrons raised to an excited state can be thermally released into the conduction band at room temperature. Other color centers may also be formed during irradiation. Among these are, for example, the V-type centers consisting of holes trapped at positive-ion vacancies or simple clusters of vacancies. Such centers, which give rise to absorption bands in the ultra-violet, will not be of primary interest here.

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† School of Mines, Columbia University, New York, N.Y.; formerly at Yale University.

‡ Hammond Metallurgical Laboratory, Yale University, New Haven, Conn.

alkali halide crystals by X-irradiation is far greater than that which can be accounted for in terms of the free vacancies originally present in the unirradiated crystals.<sup>(1)</sup> Thus, additional vacancies must be generated during the irradiation. Because a change in number and distribution of vacancies is produced by X-irradiation, it follows that a study of the change in mechanical properties resulting from irradiation with X-rays provides a unique method for obtaining information concerning the interaction between point defects and dislocations.

It has been known for many years that irradiation of alkali halides with X-rays produces marked changes in various mechanical properties of the irradiated crystals. Early experiments by Kusnetzow and Sementzow<sup>(2)</sup> and by Podaschewsky<sup>(3)</sup> have shown that the yield-strength of irradiated rock-salt is greater than the breaking strength of unirradiated specimens. Substantial increases in hardness of the alkali halides have also been observed to result from X-irradiation by Li<sup>(4)</sup> and by Westervelt.<sup>(5)</sup> These radiation-induced hardening effects are permanent; they are not reversed by optical bleaching. Recent experiments<sup>(6)</sup> have shown that for unfiltered radiation the hardness increase occurs only in the thin layer of the crystal close to the irradiated surface.

Frankl<sup>(7)</sup> discovered that irradiation has a marked effect on the internal friction properties of rock-salt, viz. that the amplitude-dependent part of the damping is almost eliminated by a brief irradiation. He found that this effects occurs throughout the entire volume of the crystals studied (in contrast to the hardness increase), and further observed that the amplitude dependence of the damping is not restored by optical bleaching of the crystal. In addition to the changes in decrement, Frankl also noted an increase in the elastic modulus (measured at low-strain amplitudes) of about 0.1% in some irradiated crystals.

According to Frankl, the observed suppression of the damping occurs because positive-ion vacancies which are freed during the irradiation, can pin down the dislocation segments responsible for the damping. (A positive-ion vacancy may be freed from a vacancy pair which dissociates when the negative-ion vacancy of the pair becomes an *F* center.) In considering this mechanism, Seitz<sup>(1)</sup> has suggested that it is more likely that the pinning is due to clusters of vacancies of both signs which condense near dislocations during irradiation at room temperature.

Unfortunately, the three mechanical properties which have been investigated in greatest detail—yield-strength, hardness, and internal friction—are relatively difficult to relate quantitatively to the dislocation

distribution. The effect of dislocations on the elastic moduli, on the other hand, is predicted by a quantitative theory. If  $N$  is the density of dislocation lines (in lines/cm<sup>2</sup> or total line-length per unit volume) which are pinned down at regular intervals to form segments of length  $l$ , then, according to Mott<sup>(8)</sup> and Friedel,<sup>(9)</sup> the movement of these segments in phase with an applied stress results in a *lowering* of the shear modulus of an isotropic material by an amount  $\Delta\mu$  given by

$$\frac{\Delta\mu}{\mu} = \frac{Nl^2}{\xi'} \quad (1)$$

where  $\xi'$  is a slowly varying function of  $l$  having a magnitude near unity. In view of the simplicity of this result, elastic-modulus measurements would appear to provide an excellent means for the investigation of the pinning of dislocations by point defects. The present investigation is directed primarily toward a detailed study of the increase in dynamic modulus which results from X-irradiation of alkali halide crystals. In some of the experiments, internal friction is also measured simultaneously with the dynamic modulus.

## 2. EXPERIMENTAL METHODS

### *Modulus Measurements*

The modulus measurements are made by the dynamic method, using the three-part composite piezoelectric resonator described by Marx.<sup>(10)</sup> Two identical quartz crystals are used in this resonator, one which serves to drive the resonator in forced vibration, and the other (the gage crystal) which indicates the resulting amplitude of vibration by means of the voltage developed across its electrodes. The driving crystal is excited by an a.c. signal taken from a variable-frequency oscillator. The specimen, whose length lies in a  $\langle 100 \rangle$  direction, is thus driven in the fundamental longitudinal mode. From the measured resonant frequency of the composite resonator, the resonant frequency on the specimen alone,  $f_s$ , may be calculated. The elastic constant  $s_{11}$  of the specimen crystal is related to the frequency  $f_s$  by means of the relation

$$f_s = \frac{1}{2L_s} \sqrt{\frac{1}{\rho s_{11}}} \quad (2)$$

where  $L_s$  is the length and  $\rho$  the density of the specimen. (The quantity  $s_{11}^{-1}$  may be regarded as the appropriate elastic modulus.) Thus, the relationship between a change in  $f_s$  and the corresponding change in elastic constant is

$$2\Delta f_s/f_s = -\Delta s_{11}/s_{11}. \quad (3)$$

The advantage of using the three-part resonator is that its resonant frequency, decrement (ratio of the energy-loss per cycle to the total stored vibrational energy), and maximum strain amplitude of vibration, can all be measured simultaneously. The decrement can be obtained with equal ease regardless of whether the damping is amplitude-dependent or not. From the measured decrement of the composite resonator, the decrement  $\Delta_s$  of the specimen alone may be obtained. Furthermore, the maximum strain-amplitude in the specimen  $\epsilon_s$  may be calculated from the voltage developed across the gage crystal. All of the measurements to be described were made with specimens which had a resonant frequency very nearly equal to 85 kc, and which matched the resonant frequency of the quartz crystals to better than 5%.

The frequency of the signal generated by the variable-frequency oscillator can be determined with a precision of better than 1 part in  $10^5$  by comparison with a quartz-controlled secondary standard. This standard oscillator is in turn monitored by comparison with a reference-frequency broadcast by radio station WWV. The precision attainable in measuring the resonant frequency of the composite resonator depends on two conditions. First, the total decrement of the resonator must be small so that the maximum of the resonance response curve can be found with precision. If the decrement of the specimen is of the order of  $10^{-3}$  or less, its resonant frequency can be determined to within at least 1 part in  $10^5$  with the present equipment. Second, the composite resonator must be in thermal equilibrium. If the resonator is kept at the ambient temperature, as in the present experiments, its temperature must be measured and appropriate corrections to the observed resonant frequency made for departure of the temperature from that selected as the reference temperature. In the present case, the temperature of the resonator was measured by means of a small thermocouple mounted immediately adjacent to the specimen (the temperature coefficient of the resonant frequency of the specimen is substantially greater than that of the quartz). The practical limit of error in temperature measurement by this method is  $\pm 0.01^\circ\text{C}$ , which corresponds to an uncertainty in resonant frequency of about  $\pm 0.2$  parts in  $10^5$ .

Resonant-frequency measurements as a function of temperature were found to fall on a straight line. When the time-rate of change of temperature was less than  $0.03^\circ\text{C}/\text{min}$ , the data fell on the same straight line regardless of whether the temperature was increasing or decreasing; when the temperature was changing more rapidly, it did not. Accordingly, the

resonator was considered to be in thermal equilibrium with the thermocouple whenever the rate of change of temperature was less than  $0.03^\circ\text{C}/\text{min}$ . In order to make the necessary temperature corrections to the observed resonant frequencies, the temperature coefficient of the resonant frequency of the composite resonator was actually measured for every specimen used.

### *Specimens*

Almost all the experiments were carried out with crystals obtained from the Harshaw Chemical Company. The specimens used were free of mechanical defects and lineage structure, as revealed by the flatness of their cleavage planes. NaCl crystals were used in most of the experimental work. The spectroscopic analyses of Harshaw rock-salt made in a number of different laboratories are reported by Duerig and Markham.<sup>(11)</sup> These crystals generally carry a small amount of divalent impurity such as calcium along with traces of trivalent impurities such as iron.

The natural rock-salt used in some of the experiments was obtained from the Brush Mineral Collection, Yale University, and originated in the Stassfurt deposits. This material had a pronounced lineage structure, making it difficult to cleave into bars suitable for modulus measurements.

Bars of the various alkali halides used were supplied by Harshaw cleaved to the same cross-section as the quartz bars used to make up the composite resonator (0.22 in.<sup>2</sup>), and were further cleaved to the proper length (about 1.03 in.) to match the frequency of the quartz driver-gage assembly. It was found possible to cleave with an accuracy of about  $\pm 0.003$  in. in most cases. Final adjustment of the length was made by grinding the ends of the bars on metallographic polishing paper.

Specimens to be cold-worked were cut oversize by the correct amount and the ends coated with collodion. The plastic deformation was accomplished by compressing the bars lengthwise in a toolmaker's vise. The collodion coating prevents the ends of the bars from splitting during heavy deformation.

### *Irradiation*

The X-rays used for irradiation were obtained from a copper target Coolidge-type tube containing beryllium windows and operating at 39 kV accelerating potential and a tube current of 10 mA. Unfortunately, the power supply for the X-ray tube did not include a voltage regulator, so that normal fluctuations of the voltage taken from the mains resulted in about a 10% fluctuation in the operating conditions of the tube over periods of the order of tens of minutes.

Since the size of the X-ray beam was too small to irradiate the specimen uniformly over its length, the composite resonator was mounted so that it could be translated vertically in front of the tube. The translational motion was obtained from a cam follower running on a motor-driven cam. The cam was cut so that the resonator moved up and down with a constant velocity, its period being about 15 sec. In this way the specimen could be irradiated uniformly without removing it from the composite resonator.

The resonator assembly was mounted in a light-tight, air-tight box containing a drying agent. X-rays were admitted through an aluminum-foil window. It was demonstrated that the presence of this window in no way influenced the formation of color centers in the specimen upon irradiation.

It was not possible to make resonant-frequency measurements during the course of irradiation because of the changing capacitance between the leads to the resonator and the light-tight box during the translation of the resonator. This capacitance change caused erratic readings on the vacuum-tube voltmeter connected to the gage electrodes. It was therefore necessary to turn off the X-rays and stop the translation of the resonator before each reading. Measurements of resonant frequency showed that the irradiation slightly perturbed the thermal equilibrium of the resonator, but that equilibrium was generally restored within a minute or two after the X-rays were turned off.

In order to irradiate a specimen at low temperature it was necessary to remove it from the resonator. The specimen was surrounded on three sides by coolant by placing it in a re-entrant cavity formed on the side of the copper box containing the coolant. This box had a volume of about one quart, and the cavity projected into the box about one inch. The coolant used was either liquid air or crushed dry ice. Radiation was admitted through an aluminum-foil window placed over the mouth of the cavity, the window serving to exclude both light and moisture.

For bleaching experiments a 4.5-amp carbon arc lamp was used as the light-source. A pair of glass condenser lenses and a water-cell heat-filter were included in the optical path. For bleaching at room temperature the specimen remained attached to the composite resonator, and the aluminum window of the dry box was replaced by a lucite one. No window was used to protect the crystal in the low-temperature box during illumination at low temperature; all water-vapor condensed at the mouth of the re-entrant cavity leaving the specimen completely dry.

### 3. EXPERIMENTAL RESULTS

#### *General Character of the Radiation Effects*

The effect of X-irradiation is to increase the modulus and to decrease the internal friction of NaCl crystals. The magnitude of the total change in modulus or in damping produced by irradiation is sensitive to the history of the individual specimen under investigation. Regardless of magnitude, however, both these properties respond to irradiation in the manner shown in Fig. 1. Here the modulus change is reported in terms of the change  $\Delta f_s$  in the resonant frequency of the specimen, and the internal friction as the decrement  $\Delta_s$ . The resonant frequency generally decreases with increasing strain amplitude of vibration.\* The frequency measurements reported in Fig. 1 (and throughout the rest of this paper), however, are measured at *low-strain amplitudes* where frequency is independent of amplitude. Fig. 1 shows that at the beginning of irradiation the changes in frequency and decrement are very rapid, but with larger X-ray doses the rate of change decreases, and eventually "saturation" values are reached. The decrement at large strain amplitudes is decreased by irradiation to the same final value as the decrement observed at small strain amplitudes, i.e. the damping becomes independent of strain amplitude, as was observed by Frankl.

The frequency and decrement changes resulting from X-irradiation are permanent at room temperature; after irradiation to saturation, neither of these quantities shows any tendency to return to its preirradiation value. Illuminating a crystal with white light after irradiation likewise has no effect on either the frequency or the decrement, in spite of the fact that such illumination bleaches out all of the coloration in the crystal.

The effects of irradiation on both the frequency and the damping represent changes throughout the volume of the crystal (in contrast to the hardness increase<sup>(6)</sup> which occurs only in a thin layer near the surface where soft X-rays are heavily absorbed). This is evident in the case of the damping merely from the fact that irradiation on one face of the specimen destroys all of the amplitude-dependent part of the damping. That the modulus increase is a volume effect is demonstrated by irradiating a crystal on one face until the frequency change is nearly complete, and then irradiating on the reverse face. For one of the NaCl specimens (No. 4) it was found, for example, that  $\Delta f_s$  after irradiating on one face for

\* These changes are usually small compared to the change in frequency (at a given amplitude) produced by irradiation.

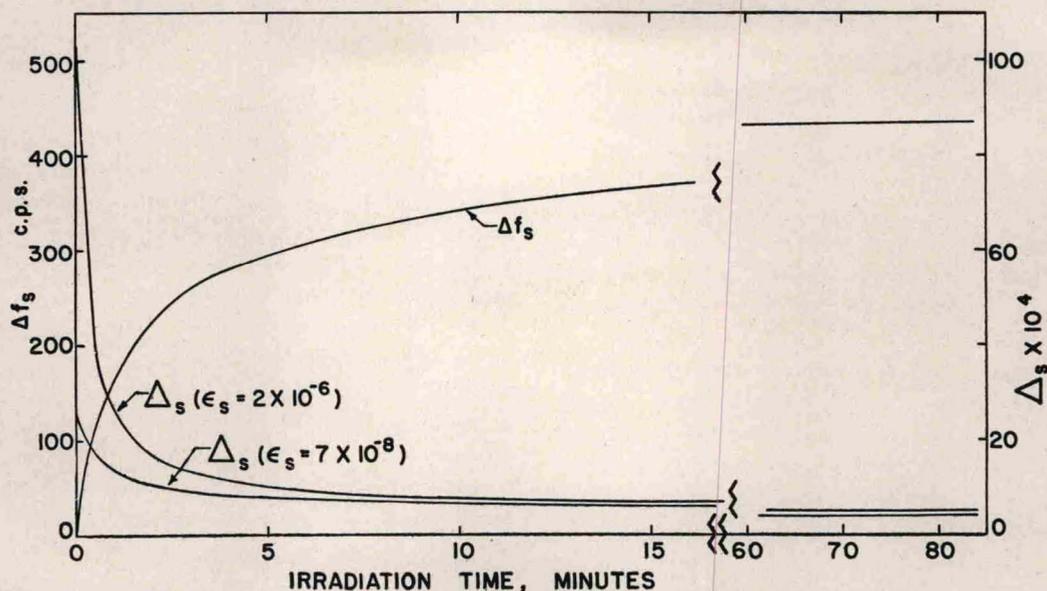


FIG. 1. Changes in resonant frequency, and in the decrement at two different strain amplitudes, of an NaCl crystal (No. 5) exposed to 39 kV X-rays. Experimental points were taken at time-intervals in geometric progression (viz. at 0.5, 1, 2, ... min) and lie on the curves shown to within the thickness of the pen line.

50 min was 175 c.p.s., while irradiation on the back face to saturation produced an additional change of only 10 c.p.s. Further irradiation with higher-energy X-rays has no additional effect on the modulus, after saturation has been achieved with lower-energy radiation.

Additional evidence on this point is supplied by an experiment in which a crystal was irradiated through a thin NaCl filter. The filter crystal, 0.030 cm thick, was interposed in the X-ray beam in front of the specimen, and the resonant frequency and decrement measured as a function of dose. The filter is thick enough to absorb a substantial fraction of the incident X-ray intensity (particularly in the long-wavelength region). Nevertheless, the modulus and damping curves so obtained are completely equivalent to those obtained without a filter. This result shows that the changes in modulus and damping are produced throughout the volume of the crystal by the more penetrating (short wavelength) X-rays.

It is known that X-irradiation increases the length<sup>(12)</sup> and decreases the density<sup>(13)</sup> of rock-salt crystals, so that some question may arise as to whether the changes in resonant frequency observed in these experiments can be wholly attributed to modulus changes. First it should be noted that, for the present radiation dosages, the density and dimensional changes are most probably confined to the layer less than 0.01 cm thick at the front of the crystal, where the majority of the incident radiant energy is absorbed,<sup>(6)</sup> whereas the resonant-frequency change resulting from irradiation

has been shown to be a volume effect. Second, the magnitude of the dimensional and density changes produced by the relatively short irradiations used in these experiments are too small to account for changes in resonant frequency of the magnitudes found here. For example, the increase in length of about 2 parts in  $10^5$  found by Sakaguchi and Suita<sup>(12)</sup> in irradiated NaCl would cause a resonant-frequency change of only 1 c.p.s. at 85 kc, which is within the experimental error of the present measurements. It is concluded that the resonant-frequency changes observed upon irradiation must be due entirely to changes in modulus.

The modulus and damping changes resulting from irradiation have been measured in specimens of NaCl from four different lots of Harshaw crystals and from one natural crystal. The nature of the radiation effects produced in all these crystals was found to be the same; only the magnitude of the effects differed from crystal to crystal.

#### Effects of Deformation and Recovery

A crystal which has been annealed at 500°C for 1 h and subsequently handled with care is characterized by a decrement which is almost amplitude-independent and by a modulus which is unaffected by X-irradiation. Curve A in Fig. 2 shows the decrement of a carefully annealed crystal as a function of strain amplitude.

Crystals quenched from temperatures of the order of 500°C show a somewhat higher decrement than the annealed crystals (equivalent to Curve B, Fig. 2),

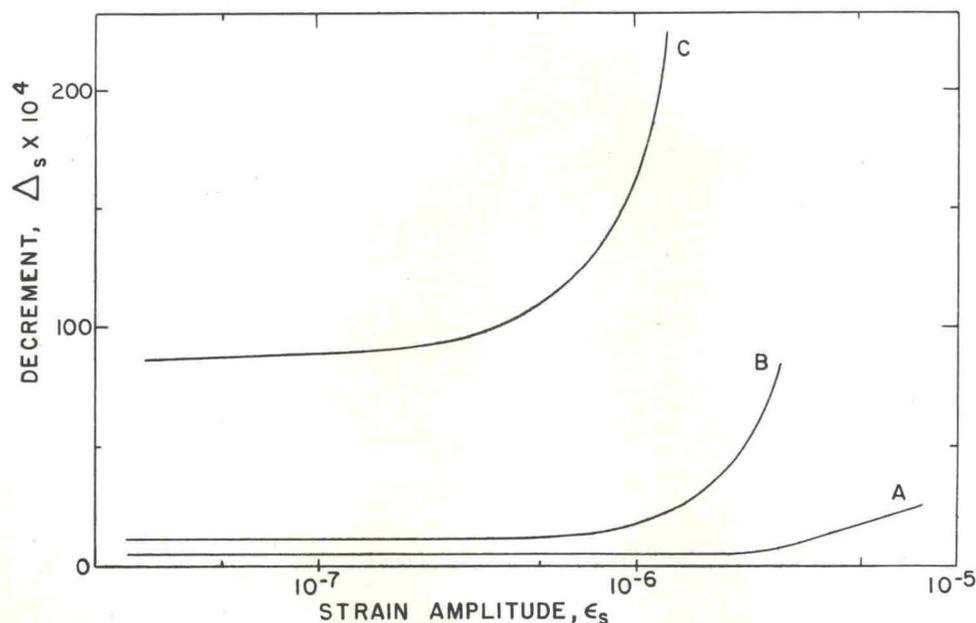


FIG. 2. Internal friction of NaCl crystals subjected to various degrees of plastic deformation. (A) fully annealed; (B) compressed 0.04%; (C) compressed 2.3%.

and a small modulus increase upon irradiation. In the as-received condition the modulus-change upon irradiation and the damping prior to irradiation are again higher than in the annealed condition. Table 1 lists some of the changes in resonant frequency produced by irradiation.

When one of the specimens used for modulus measurements is compressed in the direction of its length, slip takes place on two sets of the  $\{110\}$  planes. Both the decrement and modulus of the crystal are changed as a result of such deformation. Curve *B* in Fig. 2 shows the decrement as a function of strain amplitude observed after a carefully annealed crystal has been compressed 0.04%, while Curve *C* corresponds to this same crystal after a compression of 2.3%.

TABLE 1. Total change in resonant frequency produced in various NaCl crystals by X-irradiation

Crystal number	Treatment	Recovery time (min)	Recovery temperature (°C)	Total $\Delta f_s$ (c.p.s.)
1	annealed	—	—	0
1	quenched from 500°C	—	—	34
1	(as received)	—	—	175
8	deformed 4.07%	150	100	123
7	deformed 4.40	150	100	129
9	deformed 2.00	5760	20	212
5	deformed 2.14	180	20	434
6	deformed 4.20	1700	20	720
12	deformed 3.08	1936	20	1945
10	deformed 4.08	1440	20	2150
13	deformed 3.24	119	20	2870

It is evident that the internal friction is increased markedly by small deformations and that the strain amplitude at which the decrement becomes dependent on the amplitude of vibration is simultaneously decreased. This behavior is similar to that of deformed metal crystals.<sup>(14, 15)</sup>

The elastic modulus of the compressed crystals is lowered as a result of the deformation. This change is not conveniently observed by direct measurement, because of the dimensional changes that accompany the deformation; it becomes apparent, however, from the fact that the modulus increases spontaneously with time after deformation. Correspondingly, internal friction decreases on standing at room temperature. Similar changes in modulus and damping following plastic deformation have also been observed in deformed metals,<sup>(16)</sup> and are collectively known as the Köster effect, after their discoverer. The course of the recovery of the Köster effect is such that over a large time-interval the modulus increase is approximately linear with the logarithm of time after deformation, as is shown in Fig. 3. The decrement at large strain amplitudes decreases with time more rapidly than that at small amplitudes, while both decrement curves have the inverted *S* shape characteristic of a recovery phenomenon involving a broad spectrum of recovery times.

Exposure of the deformed crystals to X-rays increases the modulus and decreases the damping in the same way as it does in the as-received and quenched crystals, but the magnitude of the effects produced is

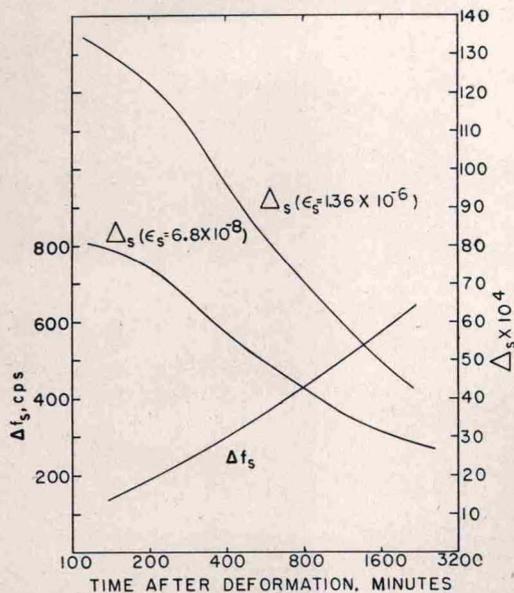


FIG. 3. Spontaneous recovery of the resonant frequency (measured from an arbitrary reference frequency) and decrement of a plastically deformed NaCl crystal (No. 6) at room temperature. The crystal was compressed 4.2% at zero time.

very much greater. The total changes in the resonant frequency of a number of crystals subjected to different deformation and recovery treatments prior to irradiation are listed in Table 1. It should be noted that the largest changes reported correspond to modulus changes of nearly 7%. It is evident from the data presented in the table that the total modulus change produced by irradiation is greater, the greater the prior deformation and the less the prior recovery time. In addition to the amount of deformation and the recovery time, the modulus change observed in any particular crystal apparently depends on other factors, such as the rate at which the crystal was deformed and/or the particular configuration of slip planes which happened to be active during the deformation (varying amounts of double slip were seen in all the crystals).

The data on crystals 7 and 8 presented in Table 1 show that recovery at 100°C prior to irradiation substantially reduces the modulus effect produced by the irradiation. It has also been shown that no modulus effect is observed when carefully annealed crystals are irradiated. These observations suggest that *irradiation can only raise the modulus in crystals where the modulus has been previously lowered by deformation and not allowed to recover fully*. In other words, the sum of the modulus increases resulting from recovery and irradiation should be the same in crystals which have been deformed the same amount, provided that care is taken to deform the crystals in exactly the same manner.

An experimental test of the above hypothesis was carried out as follows. Two crystals (Nos. 12 and 13) were carefully deformed by nearly the same amounts and then allowed to recover for different lengths of time before exposure to X-rays. The resulting resonant-frequency changes on recovery and on irradiation are shown in Fig. 4 and in Table 2. The X-irradiation was

TABLE 2. Comparison of frequency changes in two similarly deformed crystals

Crystal No.	12	13
Per cent deformation	3.08	3.24
Recovery time at 20°C (min)	2936	119
$\Delta f_s$ in recovery (c.p.s.)	1007	193
$\Delta f_s$ on irradiation (c.p.s.)	1931	2872
Total $\Delta f_s$	2938	3065
Corrected to 3.08% deformation	2938	2950

for two hours in both cases. The total changes in resonant frequency of the two crystals agree to within 4%. Even better agreement is obtained if a correction is made for the small difference in initial deformation of the two crystals, assuming a linear dependence of  $\Delta f_s$  on per cent deformation. This correction is made in the last line of Table 2 as well as in the plotted data for crystal No. 13 in Fig. 4. The excellent agreement obtained for the total modulus change of the two crystals, after correction, is undoubtedly somewhat fortuitous in view of the difficulty in deforming two different specimens in exactly the same way.

The manner in which the modulus changes with X-ray dosage has been demonstrated thus far only in Fig. 1. It is now of interest to compare such modulus vs. dose curves for crystals subjected to different prior treatments. For this purpose it is convenient to define the *modulus defect*,  $\Phi$ , as

$$\Phi = \frac{M^\infty - M}{M^\infty} = 2 \frac{f_s^\infty - f_s}{f_s^\infty} \quad (4)$$

where  $M^\infty$  and  $f_s^\infty$  are the "saturation values" of the modulus  $M = s_{11}^{-1}$ , and of the resonant frequency  $f_s$ , of the specimen attained after prolonged irradiation. The quantity  $\Phi_0$  will represent the modulus defect at the start of irradiation. The relation between the modulus vs. dose curves for specimens deformed different amounts is shown in Fig. 5, where the relative modulus defect,  $\Phi/\Phi_0$ , is plotted against the logarithm of the irradiation time. It is seen that during the major part of the modulus change these curves are straight lines; also, within experimental error, curves for the different crystals differ from one another only

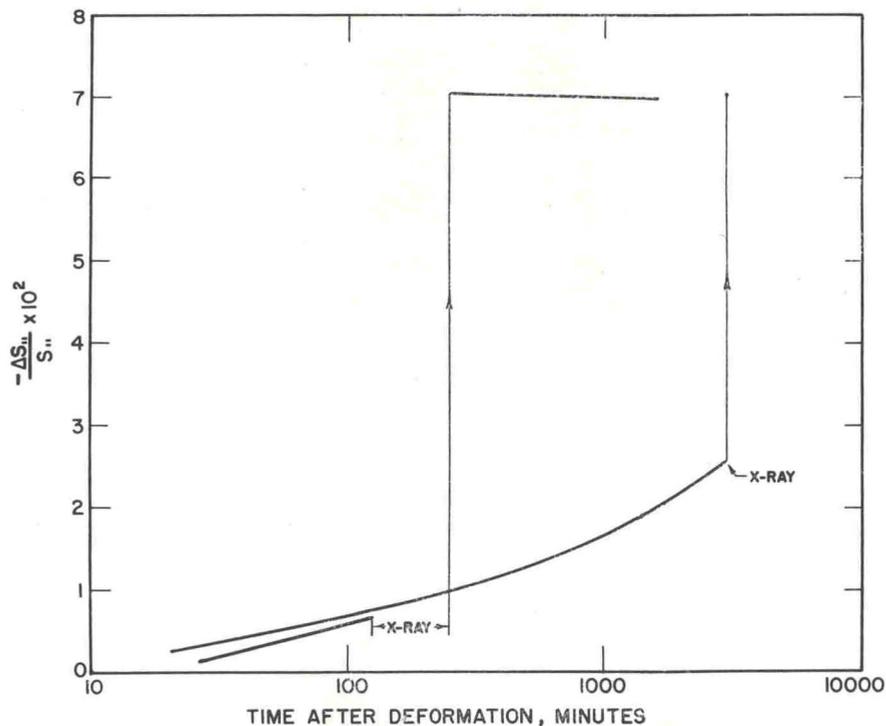


FIG. 4. The effect of spontaneous recovery followed by X-irradiation on the elastic constant of two crystals (Nos. 12 and 13). Specimens were subjected to X-irradiation for 2 h at the times indicated.

in such a way that they can be shifted into coincidence by a translation along the log-time axis.

#### Effect of Irradiation Temperature

Crystal No. 9 was irradiated at a temperature of 42°C instead of room temperature. As is evident from Fig. 5, irradiation at this slightly elevated temperature does not change the form of the modulus defect vs. dose curve.

Measurements at room temperature were also made on crystals irradiated at low temperatures. In view of the

fact that Frankl<sup>(7)</sup> reported a decrease in damping for crystals irradiated at a low temperature and warmed in the dark, but not for crystals irradiated and optically bleached at low temperature, it seemed desirable to carry out similar experiments on the modulus changes. Accordingly, some of the crystals were subjected to strong illumination after low-temperature irradiation, while the others were warmed up to room temperature in the dark. The results of these experiments are given in Table 3. In this Table,  $\Delta f_s$  (obs) is the total change in resonant frequency

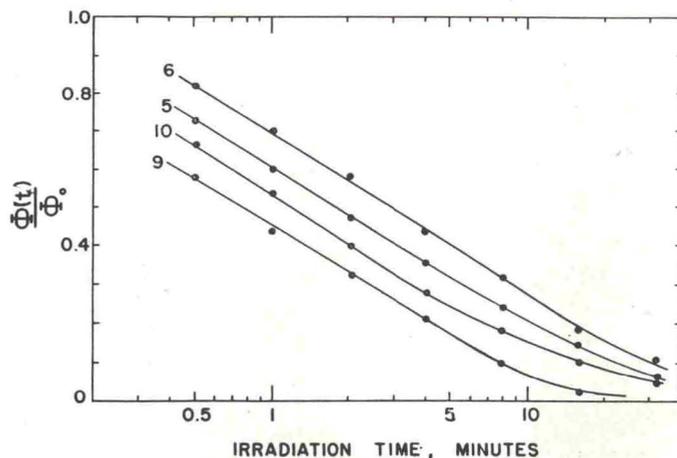


FIG. 5. Relative modulus defect of deformed NaCl crystals subjected to X-irradiation. Each curve is numbered with the identification number of the crystal measured. Data for crystal No. 4 superimposes on that for crystal No. 9.

TABLE 3. Change in resonant frequency, measured at room temperature, resulting from 1-h irradiations at various temperatures. (All crystals deformed approximately 1.5% and allowed to recover about 800 min before irradiation)

Crystal No.	Irradiation temperature (°K)	Illumination	$\Delta f_s$ (obs)	$\Delta f_s$ (corr)
18	298	none	235	241
19	180	none	250	256
20	180	at 180°K	403	310
21	78	none	261	266
22	78	at 78°K	187	214
23	78	during warm-up	45	48

upon irradiation, obtained by direct measurement; the  $\Delta f_s$  (corr) values are obtained by correcting the observed values to a standard deformation of 1.50%, under the assumption of a direct proportionality between the total frequency change and the amount of deformation. This correction is generally quite small.

In view of the difficulty in obtaining exactly the same modulus change in two different crystals, even for the same per cent deformation, it cannot be claimed that there are any significant differences between the modulus changes observed for crystals Nos. 18–22. It should be noted that, in contrast to the results reported by Frankl, there is no substantial difference between a crystal irradiated at liquid-nitrogen temperature and then warmed up in the dark (No. 21) and one irradiated and strongly illuminated at liquid-nitrogen temperature before warming (No. 22). The smaller modulus change in specimen No. 23 must, however, be regarded as significantly different. This crystal was irradiated in the usual way, but during illumination with the arc lamp the water-cell heat-filter was removed and the liquid air in the low-temperature box allowed to boil away. In this way the crystal was slowly warmed up to a temperature somewhat less than room temperature over the course of an hour while it was simultaneously subjected to strong illumination.

It was observed that very much less color is produced by the same X-ray dose when a crystal is irradiated at 78°K instead of at 180°K or room temperature. Furthermore, the color produced at either 180°K or 78°K cannot be bleached appreciably at the temperature of irradiation even under prolonged exposure to strong illumination. Among the crystals listed in Table 3, bleaching was effected only on crystal No. 23.

#### *Other Alkali Halides*

In addition to the work on NaCl, modulus and damping measurements were also made during

irradiation of KCl and LiF crystals. The observed response of the modulus and damping of KCl to X-irradiation was similar in all respects to that of NaCl. The decrement of plastically deformed LiF, however, is substantially less than that of NaCl and KCl, while the response of both its modulus and damping to X-irradiation is much slower. The resulting damping change is also small, as is to be expected in view of the fact that plastic deformation produces only a small increase in damping. Nevertheless, the magnitude of the modulus change upon irradiation seems to be comparable with that observed in both NaCl and KCl.

#### 4. DISCUSSION

The modulus measurements on cold-worked crystals show clearly that the modulus increase produced by irradiating NaCl crystals with X-rays corresponds exactly to the elimination of the modulus decrease (Köster effect) produced by plastic deformation. The Köster effect is known to be due to the presence of mobile dislocation loops produced during deformation,<sup>(7)</sup> i.e. to the effect described by equation (1). The radiation-induced modulus increase must then result from the restraint of the motion of these dislocation loops, either through regrouping of the loops in the dislocation network into more stable configurations or by the pinning down of the loops at a finite number of points by the products of irradiation. Since X-irradiation does not result in any appreciable gross heating of the crystal structure, it is difficult to see how the irradiation can produce a regrouping of dislocations. Consequently the modulus increase may be most reasonably ascribed to the formation of pinning points along dislocation loops; it may be that these pinning points are vacancies or clusters of vacancies located along the dislocations.

An attempt will now be made to calculate the shape of the curves of modulus vs. radiation dose with the aid of a number of simplifying assumptions. Let  $\phi$  be the fractional decrease in modulus (i.e. the modulus defect) in any given element of volume of the crystal due to the oscillating dislocation loops. Correspondingly,  $\Phi$  will designate the modulus defect of the crystal, resulting from the dislocations. (This definition is then consistent with equation 4.) Let  $P$  represent the total number of pinning points per unit volume created by irradiation,  $n$  be the number of pinning points per unit length of dislocation line (assumed for simplicity to be equally spaced),  $N$  be the total length of dislocation line per unit volume, and  $x$  represent depth below the irradiated surface. Since the number of pinning points is, presumably,

a function of both dose (measured by the time of irradiation,  $t$ ) and depth, we write  $P(t,x)$ . The linear density of pinning points as a function of dose and depth is

$$n(t,x) = n_0 + P(t,x)/N \quad (5)$$

where  $n_0$  is the number of pinning points per unit length of dislocation line present before irradiation. Presumably the number  $n_0$  represents, primarily, points where three dislocation lines intersect. Since  $n$  varies with both  $t$  and  $x$ , the modulus defect  $\phi$  of an element of volume of the crystal will depend on both  $t$  and  $x$ . The modulus defect for the whole crystal,  $\Phi(t)$ , observed at any time  $t$  will then be the average of the modulus defect in each layer of the crystal, providing that  $\phi \ll 1$  everywhere. Thus

$$\Phi(t) = \frac{1}{b} \int_0^b \phi(t,x) dx \quad (6)$$

where  $b$  is the thickness of the crystal in the direction of the incident radiation.

In order to express  $\phi(t,x)$  in terms of  $n(t,x)$  we may make use of the theory of the decrease in modulus due to dislocation loops devised by Mott and Friedel. Since  $n = l^{-1}$ , equation (1) becomes

$$\Delta\mu/\mu = N/n^2\xi'$$

which, as discussed in the appendix, may be written as

$$\phi = -\frac{\Delta s_{11}}{s_{11}} = \frac{N}{n^2\xi} \quad (7)$$

where  $\xi$  is a slowly-varying function of  $n$  whose value is about 2.5. Substituting equations (5) and (7) into (6), we obtain,

$$\Phi(t) = \frac{N}{b\xi} \int_0^b \frac{dx}{(n_0 + P(t,x)/N)^2} \quad (8)$$

if  $\xi$  is taken as essentially constant. Correspondingly,  $\Phi_0$ , the value of  $\Phi$  prior to irradiation, is given by

$$\Phi_0 = N/\xi n_0^2. \quad (8a)$$

To carry the calculation further requires the introduction of more detailed assumptions about  $P(t,x)$ . The following two assumptions will be considered.

*Assumption I:* The number of pinning points created by the irradiation is assumed to be proportional to the number of  $F$  centers created. Now, it is known from optical measurements<sup>(6)</sup> that, at all depths in the crystal, the number of  $F$  centers per unit volume increases nearly linearly with irradiation time in the range of X-ray dosage required to bring the

modulus increase very close to saturation.\* Thus it is possible to introduce the simplification that

$$P(t,x) = f(x) \cdot t \quad (9)$$

where  $f(x)$  gives the dependence of the rate of coloring on depth. It was also observed in the optical measurements<sup>(6)</sup> that the initial rate of formation of  $F$  centers is essentially the same in annealed crystals as in crystals deformed various amounts before irradiation. Consequently,  $f(x)$  may be taken to be essentially the same in all the crystals which are being considered here. Equation (8) combined with (8a) then becomes

$$\frac{\Phi(t)}{\Phi_0} = \frac{1}{b} \int_0^b \frac{dx}{(1 + tf(x)/Nn_0)^2} \quad (10)$$

Thus the relative modulus defect,  $\Phi(t)/\Phi_0$ , depends on irradiation time and on the previous history of the crystal only through variable  $t/Nn_0$ . It is therefore anticipated that curves of  $\Phi/\Phi_0$  vs. log (irradiation time) for different crystals should differ from each other only by a translation along the log-time axis (i.e. by a change in time scale). As shown in Fig. 5, this prediction is indeed verified within experimental error<sup>†</sup> for crystals subjected to varying amounts of deformation and recovery.

If the constant  $C$  is arbitrarily defined as the change in time scale required to bring all of the  $\Phi/\Phi_0$  vs. log-time curves into coincidence with the curve for crystal No. 6, the quantity  $C$  will be proportional to  $(Nn_0)^{-1}$ . Utilizing this result and equation (8a), we can evaluate quantities proportional to  $N$  and  $n_0$  for each crystal in terms of the known quantities  $C$  and  $\Phi_0$ . Thus,

$$\left. \begin{aligned} N &\approx \Phi_0^{1/3} C^{-2/3} \\ l_0 = n_0^{-1} &\approx (C\Phi_0)^{1/3} \end{aligned} \right\} \quad (11)$$

Values of these quantities are given in Table 4 under the heading "Assumption I" for a number of crystals having different mechanical histories.

*Assumption II:* The fact that optical measurements show an initial rate of formation of  $F$  centers which is essentially independent of the state of deformation or anneal of the specimen implies that  $F$  centers are not formed at dislocations during irradiation<sup>‡</sup>. The

\* In the optical measurements (reference 6) direct irradiation was used, in contrast to the present experiments in which the specimen was translated in front of the X-ray beam. Consequently a 10-min irradiation in the present experiments is equivalent to only 2 min in the optical experiments.

† It should be noted that the experimental error in determining  $\Phi(t)$  is largest when  $\Phi(t)$  approaches zero, because of the difficulty in determining the exact saturation value of the modulus.

‡ The interpretation of these optical data is presented in greater detail elsewhere.<sup>(6)</sup>

modulus increase resulting from X-irradiation is supposed to arise, however, from the pinning of dislocations by the action of the absorbed radiation. It is therefore conceivable that the modulus increase is due to *direct* interaction between the products of the irradiation (excitons and/or free electrons and holes) and dislocations, even though *F* centers are not primarily formed in this way. If this were the case, the density of pinning points *P* created by the irradiation would not be proportional to the number of *F* centers (as in Assumption I) but rather to the density of dislocation lines *N*. We may therefore state, for Assumption II, that equation (9) still applies, except that *f(x)* is no longer independent of the histories of the various specimens, but rather that it is proportional to *N*. The result obtained under Assumption I, that the curves of  $\Phi(t)/\Phi_0$  vs. log (irradiation time) for crystals given different prior treatments are displaced from each other by translation along the log-time axis, is therefore still valid under Assumption II. However, the relative shift *C* of the curves is now proportional to  $n_0^{-1}$  and independent of *N* (since the proportionality of *f(x)* to *N* cancels this quantity out of equation 10). Under these conditions, we obtain for Assumption II, in place of equations (11),

$$\left. \begin{aligned} N &\approx \Phi_0 C^{-2} \\ l_0 &= n_0^{-1} \approx C \end{aligned} \right\} \quad (12)$$

Values of the quantity  $\Phi_0 C^{-2}$  for various crystals are given in Table 4, under the heading "Assumption II," while the quantity *C* itself is already listed in an earlier column.

A choice between the two assumptions just discussed may be made on the basis of a comparison of numbers proportional to *N* and *l*<sub>0</sub> calculated in terms of these assumptions. For example, when the quantities  $\Phi_0^{1/3} C^{-2/3}$  for different crystals are compared, one finds that interpretation of the data in terms of Assumption I leads to the conclusion that *N* increases roughly in proportion to the amount of deformation and decreases somewhat as recovery is

allowed to proceed. The values of  $(C\Phi_0)^{1/3}$  show that *l*<sub>0</sub> is very nearly the same in all cases except one, viz. that of crystal No. 10. This crystal was evidently either deformed at a substantially different rate or else was very different in its history prior to deformation. Its large decrement ( $15 \times 10^{-3}$  as against a value of  $4 \times 10^{-3}$  for crystal No. 6) as well as its large  $\Phi_0$ -value are consistent with the large value of *l*<sub>0</sub>.

On the other hand, interpretation of the data in terms of Assumption II leads to numerical values which are difficult to reconcile with the prior treatments received by the various crystals. A comparison of  $\Phi_0 C^{-2}$  for crystals Nos. 5 and 9 shows that *N* is required to decrease by a factor greater than 10 during recovery from 180 min to 5760 min for specimens deformed very nearly the same amount. It is difficult to reconcile such a large decrease in *N* as the result of this recovery with the fact that such recovery does not greatly affect other properties of the material (such as X-ray line broadening) which are sensitive to dislocation density. The large difference in values of  $\Phi_0 C^{-2}$  for crystals No. 6 and 10 is also difficult to explain, since the deformations and recovery times of these two crystals are very nearly the same. Finally, according to Assumption II, *N* would vary much faster than linearly with the amount of deformation. This result also does not seem reasonable. In general, then, it is seen that Assumption I leads to the most satisfactory agreement between the calculated numbers proportional to *N* and *l*<sub>0</sub> and the histories of the specimens.

Evaluation of the integral in equation (10) requires knowledge of the form of *f(x)*. In terms of Assumption I, *f(x)* should have the same form as the variation in the density of *F* centers with depth below the irradiated surface. This variation has been determined experimentally and found to be roughly proportional to the rate of absorption of X-ray energy at each depth in the crystal.<sup>(18)</sup> The functional form of this depth dependence is rather complex, but under the conditions of irradiation used in the present experiments,

TABLE 4. Analysis of the modulus changes in cold-worked NaCl crystals

Crystal No.	Def. %	Room temperature recovery (min)	$\Phi_0 \times 10^2$	<i>C</i>	Assumption I		Assumption II	$N\beta^{2/3}$ ( $\text{cm}^{-2} \times 10^{-8}$ )	$l_0\beta^{-1/3}$ ( $\text{cm} \times 10^6$ )
					$\Phi_0^{1/3} C^{-2/3}$	$(C\Phi_0)^{1/3}$	$\Phi_0/C^2 \times 10^3$		
6	4.20	1700	1.68	1.00	0.258	0.256	16.8	32.0	3.61
10	4.08	1440	5.02	2.47	0.201	0.505	8.22	25.0	7.04
5	2.14	180	1.01	1.65	0.154	0.255	3.68	19.2	3.61
9	2.00	5760	0.495	3.79	0.068	0.265	0.343	8.5	3.75
4	(as received)		0.413	3.79	0.067	0.250	0.287	8.3	3.53

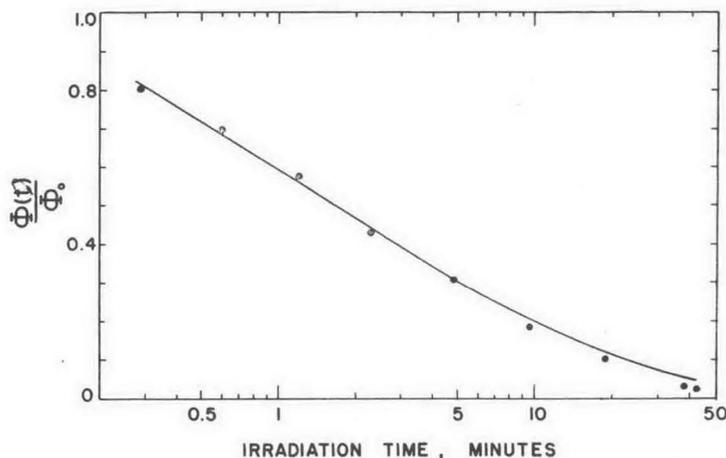


FIG. 6. Comparison of calculated and observed relative modulus defect for crystal No. 6. Curve, calculated. Points, experimental.

may be roughly approximated over a substantial range of depths by an exponential. Hence we write

$$f(x) \cong Ae^{-\gamma x} \quad (13)$$

where  $\gamma$  is the effective absorption coefficient of X-rays in the crystal. Making this substitution in equation (10), the integration may be carried out to obtain

$$\frac{\Phi(t)}{\Phi_0} = \frac{1}{\gamma b} \left\{ \frac{1}{1 + Kt} - \frac{1}{1 + Kte^{-\gamma b}} + \ln \frac{1 + Kte^{-\gamma b}}{(1 + Kt)e^{-\gamma b}} \right\} \quad (14)$$

where  $K = A/Nn_0$ . The expression on the right-hand side of equation (14) has the property that, in a plot of  $\Phi(t)/\Phi_0$  vs.  $\log$  (irradiation time), the factor  $\gamma$  determines the shape of the curve while  $K$  only determines its position along the log-time axis. The best fit of equation (14) to the data of Fig. 5 is obtained with  $\gamma = 9.5 \text{ cm}^{-1}$ . Fig. 6 shows the theoretical expression fitted to data for crystal No. 6, using this value of  $\gamma$  and  $K = 1.76 \text{ min}^{-1}$ . Values of  $K$  for the other crystals are proportional to the values of  $C$  given in Table 4.

The value of  $\gamma$  obtained by means of this analysis agrees, within experimental error, with a rough value obtained from optical measurements on similarly irradiated crystals. It is also equal to the absorption coefficient of NaCl for X-rays of wavelength  $0.55 \text{ \AA}$ . This wavelength falls close to that for maximum intensity in the continuous spectrum of the X-rays used.

Assumption I may be expressed by the statement that  $\beta F$  centers are formed for each pinning point created by irradiation. Then, according to the assumptions of equations (9) and (13), the number of  $F$  centers generated per unit volume per unit time at

depth  $x$  is  $Be^{-\gamma x}$ , where  $B = \beta A$ . The quantity  $B$  may be obtained from optical data on the rate of formation of  $F$  centers at a given depth in a rock-salt crystal irradiated in the same way as the modulus specimens. From such data<sup>(6)</sup>  $B$  is found to be  $1.5 \times 10^{15} \text{ cm}^{-3} \text{ sec}^{-1}$ . The definition of  $K$  and equation (8a) provide two equations in  $N$  and  $n_0$ , which when solved for these quantities give

$$\left. \begin{aligned} N &= (B^2 \xi \Phi_0 / \beta^2 K^2)^{1/3} \\ l_0 &= n_0^{-1} = (B \xi K \Phi_0 / B)^{1/3} \end{aligned} \right\} \quad (15)$$

Since all the quantities on the right-hand sides of the above equations are known, except  $\beta$ , the quantities  $N\beta^{2/3}$  and  $l_0\beta^{-1/3}$  may be determined from the experimental results. Values for these quantities are displayed in the final columns of Table 4, calculated with  $\xi = 2.5$  (see Appendix).

Comparison of the values of  $N\beta^{2/3}$  and  $l_0\beta^{-1/3}$  presented in Table 4 for crystals Nos. 5 and 9 suggests that the annealing out of the modulus defect produced by cold-working results from a decrease in  $N$ , and that  $l_0$  remains about constant. This result is in contrast to the suggestion previously advanced,<sup>(17)</sup> according to which this recovery effect is the result of rearrangement of the dislocation network generated by deformation into a more stable configuration, i.e. that recovery results from a decrease in  $l$  rather than a decrease in  $N$ . Further experiments along these lines would be most desirable.

Values for  $\beta$ , the number of  $F$  centers per pinning point, which fall in the range from 1 to 100, would appear to be reasonable. For crystal No. 6 (deformed 4%)  $N$  values of  $3 \times 10^9$  and  $1 \times 10^8 \text{ cm}^{-2}$  and  $l_0$  values of 100 and 500 atomic distances correspond to  $\beta = 1$  and 100 respectively. A comparison of these results with Harper's estimates of dislocation densities

in lightly cold-worked iron,<sup>(19)</sup> favors values of  $\beta$  near unity; on the other hand, to obtain a reasonable value for  $N$  for the as-received crystal (No. 4, Table 4) would require a relatively high value for  $\beta$ .

Since Assumption II above has been ruled out, the results of the preceding analysis support the hypothesis already developed on the basis of optical data,<sup>(6)</sup> viz. that vacancies are not generated at dislocations during short-time X-irradiation, except in a layer of the specimen less than 0.01 cm thick adjacent to the irradiated surface (the region of "slow-type" coloring). This thin layer near the surface does not appreciably influence the observed modulus defect which derives from the entire volume of the crystal, as demonstrated in these experiments.

If vacancies which produce the pinning of dislocations during irradiation are not generated at dislocations, what then is their source? According to Frankl and to Seitz,<sup>(1)</sup> positive-ion vacancies, liberated from vacancy pairs (or larger aggregates) when the negative-ion vacancy of a pair becomes an  $F$  center, may migrate to the dislocation lines, where they may act as pinning points either individually or after condensation into clusters. It appears that the jump-frequency of positive-ion vacancies in NaCl at room temperature is great enough to allow vacancies liberated in this way to reach dislocations.\*

Since the above mechanism of dislocation pinning involves vacancy diffusion, no modulus change is to be expected when irradiation and modulus measurement are *both* conducted at low temperatures. If, as in the present experiments, a crystal is irradiated at low temperature and warmed up without first bleaching out the color centers, the usual modulus change may still be observed at room temperature. This is due to the fact that dissociation of vacancy pairs will have occurred during irradiation at the low temperature, leaving the positive-ion vacancies free to migrate to dislocations during the warm-up period. This explanation accounts for the results of the low-temperature experiments on crystals Nos. 19 and 21 (Table 3). Since there is no appreciable bleaching of  $F$  centers formed at 78°K or 180°K during illumination at the temperature of irradiation, it is no surprise that the same modulus change is obtained even when the specimens are illuminated before warm-up (crystals Nos. 20 and 22). Only in the case of illumination *during* slow warm-up (crystal No. 23) is the room-temperature modulus change substantially reduced. This result may be explained

if it is assumed that during warm-up the bleaching process takes place before the positive-ion vacancies become mobile. When an electron is released from a negative-ion vacancy by the action of light quanta, this vacancy regains its net charge and is then electrostatically attracted to the positive-ion vacancy with which it was originally associated. Thus, as soon as the specimen reaches the temperature range in which vacancy mobility begins, recombination will take place. The fact that there is a small change in modulus in crystal No. 23 shows that recombination is not complete, i.e. some vacancies escape and produce pinning. On the other hand, when warm-up is in the dark, there is no electrostatic attraction, so that positive-ion vacancies are free to migrate when the temperature becomes sufficiently high.

The decrease in damping due to irradiation seems to be due to the same cause as the increase in elastic modulus, i.e. to dislocation pinning. In view of the fact that substantial bleaching does not take place at liquid-nitrogen temperature, it is difficult to see why the damping was not suppressed in Frankl's experiment in which a specimen was first irradiated and illuminated at 78°K, then warmed to room temperature. Since Frankl illuminated his crystal with an ordinary incandescent lamp placed close to the specimen,<sup>(20)</sup> it is suggested that the light-source employed by Frankl warmed the crystal up to the range of temperatures where a substantial amount of bleaching could take place.

## 5. CONCLUSIONS

1. Pinning points on dislocations are created during X-irradiation of an alkali halide crystal at room temperature through the release of vacancies within the volume of the crystal and the migration of these vacancies to dislocations.

2. Relatively few pinning points are produced in a crystal irradiated at 78°K and slowly warmed up under strong bleaching illumination. Under these conditions vacancies of opposite sign, which were originally dissociated through the action of the radiation, may recombine.

3. The dislocation density calculated from curves of modulus vs. irradiation time is of the order of  $10^8$ – $10^9$  cm<sup>-2</sup> for a crystal deformed 4%; the corresponding mean length of free dislocation segment is of the order of 100 atom distances. Upon recovery at room temperature, there appears to be a substantial decrease in dislocation density.

4. The pinning of dislocations by vacancies is by no means restricted to ionic crystals. Recent experiments show that excess vacancies produced in metals by

\* Positive-ion vacancies have a significantly lower activation energy for migration than negative-ion vacancies in most alkali halides.<sup>(1)</sup>

neutron irradiation<sup>(21)</sup> and by quenching from elevated temperatures<sup>(22)</sup> also result in dislocation pinning.

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#### Appendix

The factor  $\xi'$  which appears in equation (1) is given by<sup>(9)</sup>

$$\xi' = \frac{3}{\zeta\pi} \ln \left( \frac{r_1}{r_0} \right)$$

where  $\zeta = 1$  for a screw dislocation and  $\zeta = 1 + \nu$  for an edge dislocation,  $\nu$  being Poisson's ratio. This result applies to an isotropic medium. The quantities  $r_0$  and  $r_1$  are the radii of the inner and outer surfaces, respectively, of the doubly connected region used in calculating the stress field around a dislocation line. Since in the case of a short segment of dislocation of length  $l$  the calculation of the stress field should only be carried out to a distance of about  $l$  from the dislocation line,  $r_1$  may be taken as approximately equal to  $l$ . The value to be taken for  $r_0$  is somewhat more uncertain, but according to Cottrell<sup>(23)</sup> should be chosen to be several times the Burgers vector, i.e. about  $10^{-7}$  cm. In a real crystal the dislocation network will be made up of segments having partly a screw character and partly an edge character; nevertheless,  $\zeta$  will still be of the order of unity, since  $\nu \cong 0.3$ . Because it is but a slowly varying function of  $l$ ,  $\xi'$  is taken to be a constant in the present calculations. In the specimens being considered,  $l$  has values ranging between  $10^{-5}$  and  $10^{-6}$ , so that  $\xi'$  will be approximately equal to 3.

The elastic modulus which is measured in the present case is  $s_{11}^{-1}$ , whereas the shear modulus  $\mu$  on the  $\{110\}$  slip planes is given by

$$\mu^{-1} = \frac{1}{2}s_{44} + s_{11} - s_{12}$$

It is this shear modulus which properly belongs in equation (1) if an attempt is to be made to introduce

anisotropy. Even though it is known that the motion of dislocation loops does not affect the cubical compressibility  $3(s_{11} + 2s_{12})$ , there is not enough information about the effect of dislocation motion on the compliance constant  $s_{44}$  to make possible a calculation of the relation between a change in  $\mu$  and a change in the measured compliance  $s_{11}$ . Detailed analysis of the atomic forces which surround the dislocation loops would be required to obtain this information.

Instead, it seems desirable to adhere to an isotropic approximation. The measured modulus  $s_{11}^{-1}$  corresponds to Young's modulus,  $Y$ , and

$$\frac{\Delta Y}{Y} = \left( 1 - \frac{\mu}{\mu + 3k} \right) \frac{\Delta \mu}{\mu}$$

where  $k$  is the bulk modulus. Thus, by analogy to equation (1),

$$-\frac{\Delta s_{11}}{s_{11}} = \frac{Nl^2}{\xi}$$

where

$$\xi = \left( 1 - \frac{\mu}{\mu + 3k} \right) \xi'$$

Using the measured values of  $s_{11}$ ,  $s_{12}$ , and  $s_{44}$  to obtain values for  $\mu$  and  $k$ , it is found that  $\xi \cong 2.5$ .

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