heat for that alloy, deduced from the single-crystal critical field data of Blaugher and Hulm.<sup>30</sup> On the other hand, Nb<sub>3</sub>Sn undoubtedly has an unusually large electronic specific heat, comparable to V<sub>3</sub>Ga.<sup>31</sup>

<sup>30</sup> R. D. Blaugher and J. K. Hulm, Phys. Rev. **125**, 474 (1962). <sup>31</sup> F. J. Morin, J. P. Maita, H. J. Williams, R. Sherwood, J. H. Wernick, and J. E. Kunzler, Bull. Am. Phys. Soc. **7**, 190 (1962).

## ACKNOWLEDGMENTS

The authors wish to express their appreciation to W. E. Beck, A. C. Burgess, and P. A. Roland for their assistance in making the necessary measurements, and to D. J. Quinn for the use of his susceptibility apparatus. We would also like to thank Dr. T. R. McGuire for providing a Nb<sub>3</sub>Sn specimen for us to test.

PHYSICAL REVIEW

VOLUME 129, NUMBER 3

1 FEBRUARY 1963

# Direct Measurement of the Vacancies Produced in Sodium Chloride by Fast Reactor Neutrons\*

PAUL W. LEVY

Brookhaven National Laboratory, Upton, New York (Received 16 August 1962)

When unstrained NaCl single crystals are irradiated with gamma rays, reproducible curves of color-center, especially *F*-center, concentration vs dose can be obtained. At doses of approximately  $10^7$  R it appears that all of the negative-ion vacancies in the crystal have been converted into *F* centers and that they are stable at room temperature. When crystals which have been previously colored with gamma rays to this extent are bombarded with fast neutrons, and then the gamma-ray irradiations are resumed, there is an increase in *F* center, i.e., vacancy concentration, which is proportional to the total fast neutron flux. The number of vacancies induced by the fast neutron bombardment, determined in this way, is in agreement with the theoretical treatments of Kinchin and Pease, and Seitz and Koehler.

#### INTRODUCTION

ONE of the fundamental problems in "radiation damage" is the determination of the number of defects, e.g., vacancies, interstitials, and complex imperfections consisting of more than one vacancy or interstitial, or both, formed in a substance by a specified bombarding particle such as a 1-MeV neutron. Ideally, one would like to count the defects in a crystal before and after bombardment. This can be accomplished in many indirect ways, e.g., by measuring electrical resistivity. A direct way of determining the concentration of a specific type of defect in a crystal is by measurement of the number of color centers associated with it, providing, however, that a known fraction, or all, of the defects have been converted to color centers. Specifically, in NaCl the negative-ion vacancy concentration can be determined if a known fraction of these vacancies have been converted to F centers by electron capture. This paper will describe a direct determination of the number of vacancies produced in singlecrystal NaCl by fast neutron reactor bombardment by measuring the saturation F-center coloring.

## EXPERIMENTAL

All of the NaCl crystals used were cleaved from large crystals obtained from the Harshaw Chemical Com-

pany. The crystals for any single experiment were always taken from the same piece. None of the samples were annealed before irradiation since it was found that coloring curves obtained from untreated materials were the most reproducible. It is entirely possible that when the crystals are received they contain less strain than any of the crystals we subjected to thermal annealing. All crystals were kept in desiccators as much as possible.

For irradiation and absorption spectrum measurements, the crystals were mounted in slightly oversized plastic or aluminum frames to prevent their being strained in any way. Once optical absorption measurements and irradiations were started, the crystals were



FIG. 1. The absorption coefficient at the peak of the NaCl *F*-center absorption band vs gamma-ray dose. Usually these curves are referred to as growth or coloring curves.

<sup>\*</sup> Work performed under the auspices of the U. S. Atomic Energy Commission. Presented at the 1959 International Symposium on Color Centers in Alkali Halides, Corvallis, Oregon, September 1959 (unpublished); see also Bull. Am. Phys. Soc. 5, 184 (1960).



FIG. 2. The absorption spectrum of gamma-ray-irradiated NaCl at various total doses.

never exposed to light outside the spectrometer. During handling and irradiation they were wrapped in double layers of aluminum foil.

Tubular Co<sup>60</sup> sources were used for all gamma-ray irradiations. The maximum temperature encountered was about 30°C, except during the reactor irradiation when the maximum temperature reached was 70°C. Reactor irradiations were made in air-cooled facilities of the BNL graphite reactor. Fast flux measurements were made with a variety of foils. By far the greatest uncertainty in the results described here is attributable to the lack of accurate reactor flux measurements.

All absorption spectrum measurements were made with a Beckman Model DU spectrophotometer except those made in connection with the annealing measurements which were made with a Beckman Model DK spectrophotometer. From time to time, tests were made to determine if any bleaching occurred during the optical absorption measurements and none was detected.

## COLORING OF NaCl BY GAMMA RAYS

When single-crystal NaCl is exposed to  $Co^{60}$  gamma rays at room temperature, a curve of *F*-center absorption coefficient vs dose, as shown in Fig. 1, is obtained unless the crystals have been mistreated, as is described below. These are referred to as coloring curves, or growth curves, and are constructed from numerous absorption spectra. All other gamma-ray irradiations, on any one crystal or group of crystals, were always made in the same gamma-ray source at approximately



FIG. 3. The decay, at room temperature, in the dark, of the F-center coloring above and below the "knee" of the curve shown in Fig. 1.



FIG. 4. Growth curves for gamma-ray-irradiated NaCl crystals before and after 30-min anneals in air, with the crystals wrapped in Al foils. Also shown is the effect of this annealing on the coloring curves of the bands at 7.20, 6.39 eV, and the band at 1.74 eV. The bands at 7.20 and 6.39 eV are near the end of the operating range of the spectrophotometer and are subject to large errors. It is not clear that the behavior of the *F* band, due to the annealing, is correlated to or modified by the behavior of these other bands.

 $2.4 \times 10^5$  R/h. A typical example of the absorption spectrum of irradiated NaCl is shown in Fig. 2. These spectra are what one would expect from earlier color center studies, i.e., they contain well-known absorption bands, except for the structureless absorption in the 3.5- to 5-eV region.

Occasionally a growth curve is obtained which differs from this example in that it follows this type of curve (Fig. 1) at low dose and then increases more rapidly than shown at higher doses.<sup>1</sup> The explanation of this abnormal increase is that the coloring is strongly dependent on strain in the crystal, and only if samples are handled with sufficient care can one obtain reproducible growth curves similar to the one given. Also, we have shown<sup>1</sup> that the growth curves can be greatly changed by heat treatment before irradiation. It is not clear whether the heat treatment followed by careful slow cooling has increased the strain or whether some other process, such as defect migration or aggregation, is the cause of the modified growth curve.

Another factor entering into the determination of reproducible coloring curves is the decay, or thermal

<sup>&</sup>lt;sup>1</sup> B. Mozer and P. W. Levy, Bull. Am. Phys. Soc. 5, 184 (1960).



FIG. 5. The absorption spectrum of a NaCl crystal before reactor irradiation, immediately after a 30-min reactor irradiation, and after gamma-ray irradiations were resumed. The  $3.6 \times 10^7$  R total dose irradiation immediately preceded, and the  $3.9 \times 10^7$  R total dose directly followed, the reactor irradiation.

bleaching, of the absorption bands after the crystal is removed from the source. This occurs even when the crystal is kept in total darkness at room temperature. The decay of the NaCl *F*-center concentration is illustrated in Fig. 3. The  $3.8 \times 10^5$  R irradiation is below the "knee" of the curve on Fig. 1, and, as is obvious from the graph, at this *F*-center concentration a large fraction of the *F* centers are unstable. The  $1.2 \times 10^7$  R irradiation is well up on the increasing portion of the curve beyond the knee and at this *F*-center concentration the color is relatively stable. Thus, to obtain reproducible growth curves at low dose it is essential to carefully correct for any decay. At high dose there is little decay and reproducible growth curves can be obtained without correction.

When care is taken to insure that strain free samples are used, NaCl growth curves show a very rapid rise during exposure to the first  $10^5$  R, the curve then rounds off to a plateau having little slope, and finally the slope of the curve increases until it reaches its final value. Later it will be seen that whenever a NaCl crystal has been treated in a way that will leave it with negative ion vacancies which *have not* trapped electrons, subsequent exposure to gamma rays caused the *F*-center intensity to rise rapidly to the plateau just as the uncolored crystal behaves when first exposed to radiation.

From the shape of the *F*-center coloring curve and from the lack of *F*-center instability at the plateau, it is reasonable to assume that once this point has been reached, all, or very nearly all, of the existing negativeion vacancies have been converted into F centers. Also, the approximate shape of these curves is given by a theoretical treatment of the growth kinetics<sup>2</sup> which predicts that the plateau is reached when all of the available vacancies are colored. Both the annealing and reactor-irradiation experiments described below show that when uncolored vacancies exist in the crystal almost all of them are converted to color centers by a dose of 10<sup>6</sup> R and that relatively little additional coloring is obtained with greater exposure. A large part or all of the slope in the coloring curves beyond the knee can be attributed to the formation of vacancies during gamma-ray irradiations. One contributation to this slope occurs because electron recoils, mostly Compton electrons, are produced which have sufficient energy to eject atoms from their normal lattice positions. Part of the observed slope may arise from evaporation of vacancies from dislocations, or other mechanisms which have been suggested but not been clearly demostrated such as photochemical decomposition, the Varley mechanism, etc. Pertinent to the experiment described here is the observation that beyond the knee the slope of the coloring curve is small; i.e., the total number of F centers formed by all of the ionization dependent <sup>2</sup> P. W. Levy, Bull. Am. Phys. Soc. 1, 136 (1956); Phys. Rev. (to be published).



FIG. 6. The gamma-ray coloring curve for the NaCl crystal whose absorption spectra, at selected points on the growth curve, are shown in Fig. 5. Also, the growth curve of a comparison crystal which received exactly the same treatment except for the reactor irradiation. Some of the absorption spectra of this comparison crystal are shown in Fig. 2.

mechanisms is small. Furthermore, as is emphasized below, even if crystals are exposed in a reactor for 2 h, they receive a total dose less than  $10^7$  R, which will produce an almost negligible increment in the *F*-center concentration. This can be estimated from the slope.

While all gamma-ray irradiations were made at room temperature, it was necessary to make the reactor irradiations at temperatures which did not exceed 70 °C. For this reason, a series of annealing experiments were performed to determine the effect of heating upon colored samples. The samples were carefully cleaved from the same boule and colored by gamma rays until the linear region was reached. Different crystals were heated for 30 min in dry air at temperatures ranging from 50 to 300 °C, and cooled by removing them, and their thin, but light-tight Al foil covering from the furnace into the room air. The absorption spectrum of each sample was then measured before gamma-ray irradiations were resumed, and, of course, after each additional irradiation.

The effect of this heat treatment on the growth curves is shown in Fig. 4. Obviously, heating to 50 and 100°C has not modified the curves to any extent and even at 150°C only a small effect is seen. Note again, that when the sample was heated sufficiently to bleach an appreciable fraction of the centers, and the gammaray recoloring was resumed, the first irradiation restored the color center concentation to a point on the new linear curve, i.e., it colored all of the existing vacancies. It appears that even at a low temperature, such as  $100^{\circ}$ C, heating has removed some of the electrons trapped in negative-ion vacancies and, in addition, as the temperature is raised, more and more of the vacancies are annealed away. From these annealing measurements one can reasonably conclude that heating the samples to  $70^{\circ}$ C during the reactor irradiation has not modified the results in any way. The possibility that reactor irradiations strain the crystals, e.g., by thermal or mechanical shock, is discussed below.

### **REACTOR IRRADIATIONS**

From the discussion given above it is reasonable to assume that the concentration of negative ion vacancies in NaCl can be determined with reasonable accuracy only when the coloring has proceeded well beyond the knee of the F-center growth curve. Thus, to determine the number of vacancies induced by a reactor irradiation a sample is first colored with gamma rays until the stable F-center region is reached. Then it is reactor irradiated and when the radioactivity has decayed to a safe level the gamma-ray irradiations are resumed. The absorption spectra from an experiment of this type are shown in Fig. 5. It shows the absorption spectra corresponding to selected total gamma-ray doses and at the end of the seven-day "cooling off" period after the sample was removed from the reactor. The spectrum for a total dose of  $3.6 \times 10^7$  R immediately preceded,



FIG. 7. The gamma-ray-induced F-center growth for samples irradiated in the reactor 30, 60, and 120 min, and the growth curve of a comparison sample which was not reactor irradiated.

and the spectrum at a total dose of  $3.9 \times 10^7$  R immediately followed the reactor irradiation. While the sample is in the reactor and during the decay period it accumulates an additional gamma-ray dose. This added exposure may be roughly estimated and totals less than  $10^7$  R which is much smaller than the maximum dose on the coloring curves shown in Fig. 6.

To check the operation of the spectrometer, etc., and to monitor the F-center decay during the cooling off period a comparison crystal was always gamma-ray irradiated along with any sample being prepared for reactor irradiations. Thus, Fig. 6 shows the growth of the F band at 2.66 eV and the V, R, and M bands at 5.84, 2.10, and 1.70 eV in both the neutron-irradiated and comparison samples. Clearly, the comparison sample did not change during the decay period. However, the reactor irradiation has markedly increased the concentration of all the centers and the absorption in the 3.5- to 5-eV region.

The experiment described above illustrates the production of vacancies by bombarding NaCl with fast neutrons. To determine if the number of defects produced is proportional to total neutron exposure, samples were irradiated for different lengths of time. For this purpose four samples were colored by gamma rays to the linear region. Five days after the termination of the initial gamma-ray coloring absorption spectrum measurements were made to verify the stability of the coloring. Three samples were then irradiated in the reactor for 30, 60, 120 min, respectively. The fourth sample was used to demonstrate that the bleaching which occurred during the cooling off period was negligible. When the radioactivity of the crystals had sufficiently decreased, optical absorption spectrum measurements were resumed. From this data shown in Fig. 7, it can be seen that the F-center concentration has increased, even before gamma-ray irradiations were resumed.

This increase in F-center concentration is proportional to the total reactor irradiation and the longer the irradiation the closer this value is to the level reached when gamma-ray irradiations are resumed. This would be consistent with the idea that the neutron bombardment had created vacancies and the gamma-ray field in the reactor or the radioactivity in the sample produced sufficient ionization to color an appreciable fraction of the vacancies that were formed. Here again, it is apparent that the first gamma-ray irradiation after coloring is resumed, namely,  $10^6$  R, colors all of the vacancies existing at that time. In addition to the "jump" in F-center concentration, all of these crystals showed increases in the V, R, and M bands similar to



FIG. 8. Growth curves similar to those shown in Fig. 7 except that an anomalous peak occurs immediately after the gamma-ray irradiations are resumed, in the data for the reactor-irradiated samples. As discussed in the text, these peaks most likely *are not* caused by the reactor irradiations. Also, they were observed only once. If the concentration of reactor-induced vacancies is determined by extrapolating the linear part of the growth curve, as shown by the solid lines, the results are in excellent agreement with measurements made on other sets of samples.

those shown in Fig. 6. The slope of the growth curve for the sample which was not reactor irradiated and the slopes of those irradiated 30 and 60 min are nearly the same. In contrast, the slope of the sample irradiated 120 min is markedly different. One possible explanation for this is that at this defect concentration vacancyinterstitial recombination occurs, a process that may even be accelerated by ionizing radiation. Also, a reactor irradiation of this duration may have either increased the strain or annealed it enough to upset the linear growth.

Early in the course of these experiments the data obtained from one series of preliminary measurements, which are shown in Fig. 8, differed from the anticipated behavior which occurred in all the other measurements. In this one instance the F-center density, as a function of gamma-ray dose following the reactor irradiation, first increased to a maximum, then decreased, and then increased slowly with a slope comparable to the coloring curves prior to the reactor irradiation. This behavior can be regarded as the superposition of the expected steplike behavior observed in the other cases and the contribution from a temporary, transient, or metastable concentration of negative-ion vacancies (which were converted to F centers during the gamma-ray irradiation); such vacancies could have been introduced by the reactor irradiations or during this phase of the

measurements from some extraneous cause such as mishandling. It is unlikely that this additional contribution is due to the reactor irradiation since it clearly occurred only once. There is, however, just the slightest indication of a similar occurrence in the data shown in Fig. 6 in both the F center and other absorption bands. Also, the annealing measurements described above rule out the possibility this transient effect is a result of increasing the crystal temperature from roughly 25 to 70°C during the reactor irradiations. In fact, the annealing measurements suggest that this increase in temperature would decrease the F-center concentration if it affects it at all. It is well established<sup>1,3</sup> that NaCl F-center coloring or growth curves can be greatly influenced by straining the crystals prior to irradiation. Specifically strained NaCl crystals are colored much more rapidly than unstrained ones, at least in the initial coloring stages. These considerations strongly suggest that the observed transient coloring, i.e., the peak superimposed on the reactor induced step, was caused by inadvertently straining the crystals during one or more of the manipulations associated with this particular reactor irradiation. Indeed, the preliminary results obtained from unpublished experiments designed to investigate the role of straining the <sup>3</sup> P. V. Mitchell, D. A. Wiegand, and R. Smoluchowski, Phys. Rev. 117, 442 (1960).

NaCl crystals during irradiation are in accord with this explanation.

#### COMPARISON WITH THEORY

From the step in the growth curves, shown in Figs. 6 and 7 (and also from Fig. 8) the number of negativeion vacancies produced during the reactor irradiation can be computed from Smakula's formula. Also, if the neutron flux is known then the number of displacements produced per incident fast neutron can be determined. Fortunately, all of the constants appearing in Smakula's formula are known for NaCl, including the oscillator strength. An oscillator strength of f=0.9 was used but the uncertainty introduced by this is small compared to other uncertainties. The F-center absorption band was assumed to be Gaussian. Currently, there are two theoretical treatments dealing with the problem of calculating the number of displacements produced by bombarding a substance with energetic particles such as fast neutrons. One treatment is due to Kinchin and Pease<sup>4</sup> and the other to Seitz and Koehler.<sup>5</sup> Actually, these do not differ much in principle. Their similarities and dissimilarities have been discussed by Dienes and Vineyard.<sup>6</sup> In most instances, the Kinchin and Pease treatment predicts that, very roughly, 30% less defects are produced, for the same energy fast neutron, than the Seitz and Koehler theory. The first treatment is easier to handle computationally and will be the only one referred to in the remainder of this discussion.

The production of defects by fast neutrons is a twostep process. The first step is the ejection of an atom, i.e., a recoil, from its normal lattice position by an elastic collision with a neutron. (Obviously, damage can result from inelastic collisions but this results in only an insignificant effect in fast neutron bombardment of NaCl.) The interaction between fast neutrons and lattice atoms can be computed readily from the fast neutron elastic scattering cross section of Na and Cl, which averages about 3 b for each. The second step is the production of displacements by the recoiling atom as it moves through the lattice. Kinchin and Pease give two expressions applicable to fast neutron bombardment of NaCl; one is valid well below 1 MeV and the other is valid above 1 MeV. These have been used to compute the curves shown in Fig. 9 where, in order to span the region in which a valid expression is not currently available, the curves have been joined by "sketching in" what appears to be a reasonable curve to connect the two regions. These curves clearly show how the number of displaced atoms which are formed depend upon the mass of the recoiling atom.



FIG. 9. The number of displacements formed per fast neutron for the different recoiling atoms in NaCl and KCl, as a function of neutron energy and computed from the Kinchin and Pease relations using 25 eV as the threshold energy. One of these relations is valid well below and the other above 1 MeV. A single continuous curve was obtained by interpolating between the two valid regions.

Also, note that it is the total number of displaced atoms formed by each type of recoil which has been computed. In order to compare the computed vacancy concentrations with the experimental measurements, in which only negative-ion vacancy concentrations are determined, it is assumed that one-half of the total number of vacancies formed by each type of recoil are negative-ion vacancies. In addition, these curves are strongly dependent on the "threshold displacement energy," the minimum energy which must be transferred to a lattice atom to eject it from its normal position. Inasmuch as a direct measurement of the threshold energy for NaCl is not available, 25 eV has been used for both kinds of atoms.6

To ascertain the number of displaced atoms resulting from a given irradiation, an accurate measurement of the neutron flux is essential. This requires a determina-



FIG. 10. A comparison of the concentration of defects induced by reactor irradiation, and determined from the growth curves. with the theoretical treatment of Kinchin and Pease. Most of the uncertainty in the calculated curve results from a lack of an accurate measurement of the fast neutron flux and spectrum. The circle was computed from the data shown on Fig. 6 and the triangles from Fig. 7.

<sup>&</sup>lt;sup>4</sup> G. H. Kinchin and R. S. Pease, *Reports on Progress in Physics* (The Physical Society, London, 1955), Vol. 18, p. 1. <sup>5</sup> F. Seitz and J. S. Koehler, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956),

Vol. 2.

<sup>&</sup>lt;sup>6</sup> G. J. Dienes and G. H. Vineyard, Radiation Effects in Solids (Interscience Publishers, Inc., New York, 1957).

tion of both the total flux and spectral distribution of the fast neutrons. From Fig. 9 it can be seen that the number of displacements produced by neutrons above 1 MeV is relatively independent of the neutron energy and below 1 MeV the number of displacements is roughly proportional to neutron energy. Thus, if the average energy of the neutrons is well above 1 MeV an accurate knowledge of the spectral distribution is less important than it would be if the average neutron energy is at or below 1 MeV.

One can assume that the average neutron energy is well above 1 MeV when the samples being irradiated are close to fuel elements and relatively little moderating, or degrading of the neutrons, is occurring. Ideally, it is better to irradiate samples inside of hollow tubes, or similar geometric configurations of fissionable materials which capture all or a large fraction of the incident slow neutron creating a flux of fission spectrum neutrons in the tube interior. Unfortunately, such an arrangement, usually called a converter, was not available for these experiments. In addition, precise flux spectrum measurements could not be obtained for the irradiation facilities actually used. The flux measurements that were made in these facilities indicate that a substantial fraction of the fast neutrons were above 1 MeV and thus it is reasonable to assume an average fast neutron energy of 1 MeV and this value was used for all computations. From these considerations it is apparent that some uncertainty has been introduced in the computation of the total number of vacancies found because the flux spectrum could not be precisely determined. However, since the average energy of the fast neutrons was greater than 1 MeV (unless there are unknown errors) and because of the way the defect formation depends on neutron energy, as shown in Fig. 9, this error should not exceed 20 to 30%.

All of the results which are included here giving the direct measurement of the number of displacements produced in NaCl by reactor neutrons are contained in Fig. 10. In this figure the number of displacements experimentally determined from the data in Fig. 6 is given by the open circle and the results of the data shown on Fig. 7 are shown by the triangles. The number of displacements expected was calculated from the treatment of Kinchin and Pease. Also indicated is the error introduced because of uncertainties in the neutron flux and spectrum measurements. Obviously, the measurements are in reasonable agreement with the calculated values.

In computing the number of negative-ion vacancies formed, the contribution from centers containing more than one vacancy has been ignored. That the reactor irradiation has produced some of these multiple defects is clearly evident from Fig. 6 which shows there is a marked increase in the concentration of V, R, and Mbands. Just how many vacancies are involved in these multiple defects is difficult to ascertain. If the oscillator strength of any of them happens to be small, say 0.1

(none are known at present), they could include as many vacancies as are contained in F centers. It is not surprising that neutron bombardment should create multiple defects. The displacements from any one recoil will cluster along its path and some of these should aggregate into V, R, M centers, etc., and perhaps some centers of a type produced only in this manner.

Most of the published measurements of the number of defects produced by radiation damage indicate that the number of defects formed is one-half to one-tenth that predicted by theory (see Dienes and Vineyard<sup>6</sup>). Almost all of these results were obtained from heavily irradiated samples containing large defect concentrations. This difference between theory and experiment is most often explained by assuming that considerable recombination of defects occurred during or immediately after irradiation or that there was appreciable thermal annealing during irradiation. In contrast, the experimental results given here are unusual since they agree with the theoretical predictions. While these measurements may be entirely reliable, for completeness one should consider the possibility that some extraneous effect has "crept in" and that the observed agreement is merely fortuitous. Specifically, is the number of defects produced in NaCl by fast neutrons actually less than predicted by theory and less than measured in these experiments? In other words, did some process occur other than the production of defects by recoiling lattice atoms which caused an additional increase in the vacancy concentration? In analyzing the data given above, it has been assumed that all of the change in F-center concentration, which occurs as a result of reactor irradiation, resulted from the damage process. Actually, there are several processes which conceivably could occur that would invalidate this assumption.

First, the alkali halide color center literature contains numerous references suggesting, but not definitely establishing, that the F-center absorption band may actually consist of the superposition of two or even three bands. Each of these additional bands would be caused by defects other than F centers. Obviously, if this superposition of bands actually occurs, attributing all of the observed optical absorption to F centers will result in an overestimate of the number of negative-ion vacancies. Secondly, during irradiation vacancies could be produced by processes other than the normal radiation damage process. Already discussed is the manner in which mechanically straining crystals, either purposely or inadvertently, prior to gamma-ray irradiation produces large increases in F-center concentration when the crystals are subsequently exposed to radiation. In addition to the possibility that reactor irradiation has strained the samples in a way related to macroscopic strain, each radiation damage event probably produces a microscopic strain region. In passing through the lattice an energetic recoil will temporarily create a region of high temperature usually called a thermal spike. After the temperature pulse has subsided these regions may retain considerable strain or stress also called spikes. (No attempt will be made to describe in detail the various kinds of spikes which have been proposed, nor to give references to the literature on spikes.) Each spike may form dislocations which in turn produces additional vacancies that become F centers when the gamma-ray irradiations are resumed. Besides the two mechanisms described above, there is an additional one which apparently has not been seriously considered. Dislocations will exist in all but the most carefully annealed crystals, and they will be static as far as dislocation motion is concerned. Most dislocations will be pinned at various points by a variety of different kinds of pinning points. For example, any Frank-Read sources present will be inoperative because of pinning, pileup, or lack of driving stress. In other words, the dislocation system will be in a static equilibrium situation. During irradiation this equilibrium can be disturbed. In particular, any of the various spikes associated with each damage event can unpin the existing dislocation. For example, Frank-Read sources which were inoperative because of pileup could give rise to considerable dislocation motion. In addition, the strains associated with spikes can be relieved by the motion of existing or even new dislocations. Obviously, if any of these indirect radiation-damage dislocations-coupled processes occur, they can create many vacancies in addition to those created in the usual radiation damage process. Thus, while the measurements described in this paper

indicate that the number of vacancies produced by reactor neutron bombardment of NaCl is substantially in agreement with the theoretical predictions, there is a possibility that this agreement is fortuitous.

#### DISPLACEMENT MEASUREMENTS IN KCl

Results similar to those described above have been obtained with single crystals of KCl. However, it is not clear that they can be used to obtain a quantitative measurement of the displacements formed by neutron bombardment. The *F*-center coloring is less stable, the shape of the coloring curve appears to be modified by reactor irradiation, and the rapid coloring of existing vacancies apparently does not occur. However, results obtained so far clearly show the production of vacancies by fast neutrons.

#### CONCLUSION

The F-center coloring of NaCl single crystals has been utilized to make a direct measurement of the number of negative-ion vacancies produced by fast neutron bombardment. The number of vacancies found is in accord with the theories of Kinchin and Pease, and Seitz and Koehler. The error in these experiments is approximately 50% and almost all of it is the result of uncertainties in the fast neutron flux and spectrum measurements. It appears that these color center measurements can be made with considerable improvement in accuracy, and attempts to accomplish this are underway.