deformation potential scattering are thus found to be of the same order of magnitude. On the other hand $g^{(\text{pol})}/f = 7.4 \times 10^{-2}$ for CdS and $g^{(\text{pol})}/f = 5 \times 10^{-2}$ for GaAs. Electron-radiation interaction is two orders of magnitude stronger than the electron-phonon interaction. In the perturbation theory they should really be treated in different orders. However, we have followed the common practice of treating H_{eR} and H_{eL} in the same order of perturbation theory.

VI. CONCLUSION

Virtual quasiparticles play an important part in Raman scattering as intermediate states. The Raman tensor is found to have a pole at an incident photon energy equal to the exciton binding energy, in agreement with experimental results available at present. We believe that the Raman scattering might be used as a tool to probe quasiparticles in crystals.

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X-Ray-Induced First-Stage Coloring of NaCl[†]‡

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The initial or first-stage F-center coloring of NaCl, irradiated with x rays at room temperature, has been carefully studied. Both Korth and Harshaw samples were used. The first-stage coloring is modified by changing the experimental conditions. These include altering the dose rate; subjecting the crystals to plastic deformation before irradiation; and cycles of coloring, bleaching, and recoloring. The observed curves of F-center concentration versus dose can be resolved into one linear and three additional components. Two of these can be considered saturating exponentials. The third, which does not occur in every curve, may also be a saturating exponential, but is too weak to characterize precisely. Changing the experimental conditions alters one or two of the exponential components but leaves the others unchanged. This suggests that each component is related to a separate process and that the resolution into components is not fortuitous. The linear component is unaffected by all the above-mentioned changes in experimental conditions. The exponential component which varies slowest, as a function of irradiation time, is independent of dose rate and prior coloring and bleaching. Its saturation level is increased by plastic deformation prior to irradiation, but only after the strain has exceeded a threshold value. Beyond this threshold strain, the saturation level increases linearly with strain. The saturation level of the next slowest component is unaffected by strain. However, it is increased by a coloring-bleaching-recoloring cycle and is a function of dose rate. This dose rate dependence is given by the expression: $C_1+C_2I^2$, where C_1 and C_2 are constants and I is proportional to x-ray intensity. The saturation level of the third and fastest-changing component is always less than 10% of the total exponential contribution and is too small to study in detail. In the 185- to 350-m μ region both the absorption spectrum and the behavior under the various experimental conditions differ considerably in the two types of crystals. Also, the coloring curves seem to be strongly dependent on impurity content in a way that is also different in the two types of crystals.

INTRODUCTION

HE coloring of crystals, or more explicitly, the formation of color centers by ionizing radiation, has been studied for many years. The dependence of color-center concentration on dose or irradiation time is often called a growth curve or coloration curve. In most materials these curves are influenced by numerous factors. The most important of these appear to be the impurity content,¹⁻⁷ plastic deformation,^{1,8-12} ionizing

New York.
¹ J. H. Schulman and W. D. Compton, *Color Centers in Solids* (The MacMillan Company, New York, 1962).
² W. A. Sibley and E. Sonder, Phys. Rev. 128, 540 (1962).
³ W. A. Sibley, E. Sonder, and C. T. Butler, Phys. Rev. 136, 1527 (1964).

⁴G. A. Noble and J. J. Markham, J. Chem. Phys. 41, 1880 (1964).

radiation intensity,^{1,13–16} and the presence or absence of light that can optically bleach the centers formed.¹⁷

- ⁵ T. M. Srinivasan and W. D. Compton, Phys. Rev. 137, A264 (1965)
- ⁶ P. M. Gruzensky, J. Chem. Phys. 43, 3807 (1965).
- ⁷ L. I. Roshchina and V. P. Klyukvina, Fiz. Tverd. Tela 7, 2262 (1965) [English transl.: Soviet Phys.—Solid State 7, 1829 (1966)].
- (1966)].
 ⁸ F. Agullo-Lopez and P. W. Levy, Bull. Am. Phys. Soc. 8, 268 (1963).
 ⁹ W. A. Sibley, Phys. Rev. 133, A1176 (1964).
 ¹⁰ R. Chang, Phys. Rev. 138, A839 (1965).
 ¹¹ W. A. Sibley, C. M. Nelson, and J. H. Crawford, Jr., Phys. Rev. 139, A1328 (1965).
 ¹² B. S. H. Royce and R. Smoluchowski, Discussions Faraday Soc. 38, 218 (1964).
 ¹³ P. G. Harrison, Phys. Rev. 131, 2505 (1963).
 ¹⁴ E. Sonder and W. A. Sibley, Phys. Rev. 129, 1578 (1963).
 ¹⁵ P. V. Mitchell, D. A. Wiegand, and R. Smoluchowski, Phys. Rev. 121, 484 (1961).
 ¹⁶ E. Abramson and M. E. Caspari, Phys. Rev. 129, 536

¹⁶ E. Abramson and M. E. Caspari, Phys. Rev. 129, 536 (1963).

¹⁷ J. W. Mathews, W. C. Mallard, and W. A. Sibley, Phys. Rev. 146, 611 (1966).

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In the alkali halides, the F-center coloration curves obtained at room temperature can, in almost every case, be divided into three stages. These are schematically shown in Fig. 1. The factors which influence the growth curves often affect the various stages in different ways. There is a modest amount of information on the factors influencing stages II and III. Some of the factors that influence stage-I F-center formation in NaCl at room temperature are described in this paper. These include the dependence on radiation intensity, i.e., dose rate, plastic deformation, and optical bleaching after prior coloring.

EXPERIMENTAL

The NaCl samples used in these measurements were cleaved from a single crystal obtained from Korth, and from two crystals obtained from Harshaw. The samples were 1×1 cm on a side and between 0.8 and 1 mm thick. After cleaving they were used without any additional treatment, such as heat treatment. Samples were colored by exposing them to x rays from a tungsten target filtered by a 1.5 mm beryllium window and 1.6 mm of aluminum. The tube was always operated at 60 kV at currents between 5 and 30 mA. With a tube current of 10 mA, the samples were subjected to approximately 900 R/min. Under these conditions the coloring, i.e., the x-ray absorption, is quite uniform as indicated by the observation that the color-center concentration did not depend on sample thickness for crystals between 0.6 and 1.6 mm. In order to compare coloring curves obtained at different intensities the absorption coefficient μ_F has been plotted against an arbitrary dose unit. The unit used is 1 min exposure at 10 mA tube current, which is roughly 900 R/min.

Optical-absorption measurements were made with a Cary 14 R spectrophotometer. Crystals were kept in the dark throughout each series of measurements except when they were intentionally bleached by exposure to light. All points on the F-center coloring curves were obtained 2 min after the irradiation was terminated. Usually the measurements used in the coloring curves were restricted to the F band. Occasionally, the entire spectrum was determined. If the F-center measurement was repeated approximately 5 min later, the same absorption was obtained. This indicates that the coloring is stable for a period longer than required to make the measurement. The samples were plastically deformed along their longest dimension by slowly compressing them by hand in a toolmakers vise. The parameter labeled percent strain is the deformation, in percent of the original length, measured after the compressing stress was released.

ANALYSIS OF THE COLORING CURVES

A typical curve of F-center concentration versus irradiation time, or total dose, is schematically shown in Fig. 1. The analysis used, in this paper, to describe



Fig. 1. The upper plot shows a schematic curve of *F*-center concentration versus dose, or irradiation time, as usually observed in NaCl or KCl. The stages shown are specified in the most commonly used way. The lower section is a schematic representation of the method used to analyze the first-stage coloring into "exponential" components. Note that the parameter μ_s is defined in the upper figure and that the exponential parameters, i.e., the *a*'s, are computed from the slopes of the μ_i .

the stage-I coloring contains several steps. First, the linear region in stage II is extrapolated to t=0. The slope of this linear part is labeled a_L . The difference μ_3 between this extrapolated linear stage and the coloration curve is then plotted, using a semilogarithmic scale, against the irradiation time. This is schematically shown in the upper curve of Fig. 1. The intercept of the extrapolation of the curve μ_3 to t=0 is called A_3 . This is equivalent to assuming that μ_3 is given by $\mu_3 = A_3$ \times {1-exp($-a_3t$)}. The constant a_3 is computed from the slope of μ_3 . This procedure is repeated using the difference between the extrapolation of μ_3 and the remainder to determine the parameters A_2 and a_2 that define the curve μ_2 , as shown in the lower part of Fig. 1. This procedure can be repeated to generate the parameters describing the remaining component μ_1 , and so forth.

This method of analysis is equivalent to assuming that the curve of *F*-center absorption μ_F versus irradia-



FIG. 2. Typical *F*-center growth curves obtained from Harshaw and Korth NaCl crystals, both strained and unstrained, irradiated with x rays. 10 mA corresponds to approximately 900 R/min. From these curves the slope of the linear second stage can be specified to within a few percent.

tion time can be described by a function of the form

$$\mu_F = a_L t + \sum_{i}^{n} A_i (1 - e^{-a_i t}), \qquad (1)$$

where n is the number of "exponential" components. Clearly, one would choose an expression like (1) to describe the first- and second-stage coloring, on a purely empirical basis. However, arguments can be given which indicate that there is a physical basis for the use of this expression. First, this exponential-plus-linear growth has been observed both in the alkali halides^{1,18} and in other materials such as Al₂O₃.¹⁹ In the Al₂O₃ case,¹⁹ the exponential or exponential-plus-linear growth can result from a particularly simple coloring mechanism. Namely, at any time, the defect concentration is the number present before irradiation plus a contribution which increases linearly with dose, and, during any time increment, a constant fraction of the uncolored defects are converted to color centers by ionizing radiation. If the coloring mechanism involves more than one process, or if the observed absorption results from the superposition of more than one type of color center, there will be separate linear and/or exponential contributions from each. Second, the number of exponential components appears to be small. In the NaCl work described here, there is a maximum of three. A careful determination of the first-stage coloring of KCl can also

be resolved into three exponential components.²⁰ Third, some of the more plausible mechanisms proposed to explain the coloring curves lead to expressions having the same form as expression (1). Once it is realized that each term in (1) may represent a separate physical process, it is possible to state a criterion that strongly supports the use of (1) even if it does not prove its applicability. Namely, if a coloring curve consists of two or more components it should be possible, by altering the experimental conditions, to change one or more of the components and leave the others unchanged. It would be particularly useful, for determining which physical process occurs during color-center formation, to find an experimental treatment that would alter the magnitude of one component and leave all of the other constants unchanged.

EXPERIMENTAL RESULTS

A variety of coloring curves were obtained by varying the experimental conditions such as dose rate and degree of plastic deformation. It was readily determined that the curves reflected changes in experimental conditions and that they were, with infrequent exceptions, reproducible. The changes were most easily characterized in terms of the analysis given above.

To begin, all of the coloring curves obtained exhibited first- and second-stage behavior. A selection of coloring curves obtained under a variety of conditions are shown in Fig. 2. These are typical of the coloring curves ob-

¹⁸ F. Agullo-Lopez and J. L. Alvarez Rivas, Phys. Letters 18, 254 (1965).

¹⁹ P. W. Levy, Phys. Rev. 123, 1226 (1961).

²⁰ S. Kalbitzer and P. W. Levy (to be published).

tained with many alkali halides at low dose rates. The separate first and second stages are readily discernible. The second stage was usually linear over a sufficient range to determine a_L within a few percent. Also, the analysis procedure described above yielded clearly defined regions labeled μ_2 and μ_3 on Fig. 1. The least intense component μ_1 never exceeded 10% of A_L (defined in Fig. 1) and was usually much smaller. Consequently, this small μ_1 component will not be considered in the remaining discussion. It should be pointed out that μ_1 is negligible for all but the smallest values of t and does not influence the determination of any of the constants such as a_3 , A_3 , a_2 , or A_2 , etc. Thus, when μ_1 is neglected, the curve of F-center absorption μ_F versus irradiation time t, or dose, may be expressed as

$$\mu_F = A_2(1 - e^{-a_2 t}) + A_3(1 - e^{-a_3 t}) + a_L t.$$
⁽²⁾

The experimental results obtained for the different components of first stage will be discussed separately.

1. The Linear Component $a_L t$

This component, which is stage II, was not intensively studied. However, the constant a_L appears to be independent of dose rate, or at most, the variation is extremely small; at least in the dose-rate range used in this work. Also, differences between cleaved and strained samples could not be detected.

2. The Slowest Exponential Component μ_3

The slowest exponential growth, i.e., the one requiring the longest irradiation time to reach saturation, is described by the parameters A_3 and a_3 . The values of A_3 varied by a factor of 2 between different blocks in the same shipment. However, A_3 was very nearly constant for samples from the same block. The intercept A_3 is independent of dose rate in the range 5 to 30 mA. Typical data indicating this is shown in Fig. 3. It is consistent with recently reported results.¹⁸ However, the slopes, i.e., the a_3 's, varied greatly from one crystal to another. Also, the values of a_3 obtained at different dose rates scatter as much as values obtained from different samples cleaved from the same block and irradiated under identical conditions. The A_3 's and a_3 's are not affected by bleaching the sample with white light and then recoloring it. In fact, the values of a_3 obtained for each sample after bleaching are exceedingly close to those obtained for the same sample before bleaching. This suggests that the variations in a_3 are due to differences between samples. In the remainder of this paper we will consider the A_3 's and a_3 's to be unaffected by changing the dose rate and coloringbleaching-recoloring cycles even though the a_3 's have considerable scatter.

Plastic deformation produces changes in both A_3 and a_3 as shown in Figs. 4 and 5. The slope a_3 appears to decrease monotonically with increasing strain. A_3 is



FIG. 3. A typical example of the exponential components observed in Korth and Harshaw NaCl. The lines, extrapolated to A_3 , represent the slowest varying component.

unaffected by small strains but increases linearly for strains exceeding a threshold value. This suggests that a small deformation does not influence the mechanisms responsible for this component. Specifically, a small amount of plastic deformation does not produce vacancies or influence whatever vacancy-producing mechanism is operating. However, the larger strains cause an



FIG. 4. The magnitude A_3 of the slowest varying component μ_3 as affected by plastic deformation prior to irradiation. All of the points in this plot were obtained from crystals colored at a dose rate of 30 mA.

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FIG. 5. The exponential parameters a_2 and a_3 as a function of strain prior to irradiation in Korth crystals. The points given were obtained from crystals at a dose rate of 30 mA.

increase in the number of vacancies, or sources of vacancies, to which this component is due. Again, if a plastically deformed sample is bleached after irradiation and it is then reirradiated the same values of A_3 and a_3 are obtained.

3. The Intermediate Exponential Component μ_2

This component is characterized by the parameters A_2 and a_2 . The effect of plastic strain, x-ray intensity, and bleaching, were investigated. First, the saturation level of μ_2 , i.e., A_2 , is not changed by plastically deforming the crystal. This is markedly different from the behavior of A_3 , which depends on strain. However, a_2 , the slope of the μ_2 component, decreased monotonically as the strain is increased. This is shown in Fig. 5 for Korth crystals. Similar plastic-deformation effects were observed in Harshaw crystals, but slightly different values of A_2 and a_2 were obtained.

The dependence of the μ_2 component on x-ray intensity is shown in Fig. 6. The intercept A_2 increases with dose rate, expressed as tube current, above 5 mA. However, again above 5 mA, it is best described as proportional to the square of the x-ray intensity *I*. This is shown in Fig. 7, where A_2 is plotted as a function of I^2 . It is apparent that both the intercept and the slope for the two types of samples are different. This dependence of A_2 on dose can be expressed as $A_2 = (\text{const})_1$ $+ (\text{const})_2I^2$. On the other hand the exponential parameter a_2 , which has somewhat less scatter than a_3 , is not dependent on dose rate and is different for Korth and Harshaw samples. Typical values are $a_2=0.36/(\text{dose unit})$ in the former and 0.46/(dose unit) in the latter.

If a sample that has been irradiated well into the linear stage is bleached with white light until all color is removed and then recolored a_2 is unchanged but A_2 is increased. This suggests that during the initial coloring, and/or during the second stage-I coloring, vacancies have been created (or rendered colorable) by irradiation.

RELATION BETWEEN F-CENTER COLORING AND OTHER BANDS

An effort has been made to determine if the *F*-center growth curves are influenced by the presence or absence of other centers. In particular, an effort has been made to correlate the *F*-center growth with absorption bands in the 185- or 350-m μ region where the Korth and Harshaw crystals are quite different. Prior to irradiation the Korth samples are free of absorption bands in the *V*-band region. Irradiation produces a broad band, extending from 200 to 270 m μ and one peak at 225 m μ , which is, presumably, the V_2 center.¹ Clearly, more than one band is present since a shoulder appears on the long-wavelength side of this peak. This is shown in Fig. 8. After bleaching with white light, the presumed V_2 center disappears and only a small broad band remains in the 230-240-mu region.



FIG. 6. The lines represent the next to slowest component. The differences between extrapolations of these lines and the corresponding points on the zero-dose axis are the third component. Usually this is only a few percent of the total F-center concentration.



FIG. 7. Demonstration that the parameter A_2 depends on the square of the dose rate and is given by the expression $A_2 = (\text{const})_1 + (\text{const})_2 I^2$.

It has been shown that the equilibrium F-center coloring in KCl and NaCl is dose-rate-dependent.¹³ In other words, when a sample colored at a high rate is subsequently exposed to less intense irradiation the Fcenter absorption is decreased and when a sample colored at a low rate is subsequently irradiated at a high rate the absorption increases. Figure 8 shows both the V-region and F-center coloring of a Korth crystal during a sequence of irradiations at different dose rates. These results show two things. First, the F-center results are in accord with the previous observations on NaCl and KCl.¹³ Second, the V-region absorption varies with dose rate in the same manner that the F-center concentration varies. This strongly suggests that the growth of the bands in both regions are interdependent. And, if this is the case, the observed I^2 dependence of A_2 could result from a kinetic process proportional to the product of the electron and hole-trap concentrations.

In Harshaw crystals irradiation increases the absorption throughout the V-band region; clearly identifiable peaks are not observed. Specifically, the peaks observed in Korth crystals are absent. However, in the Harshaw samples the absorption at 185 m μ decreases rapidly with irradiation until a certain value is reached. Simultaneously, a band appears to be forming at 189 m μ . After it has increased to the point that its presence cannot be doubted both it and the 185-m μ band continue to increase. However, the two bands merge into a single band. After bleaching with white light the Fcenter disappears and the band at 189 m μ increases. This is shown in Fig. 9. It would be very difficult to resolve these two bands sufficiently to describe this behavior quantitatively.

Since the F-center and ultraviolet bands had been shown to be dose-rate-dependent in Korth crystals, similar effects were searched for in Harshaw crystals. A typical sequence, showing the effect of decreasing the dose rate after initially coloring at a high rate is shown in Fig. 10. In this case the F band decreases while the

TABLE I. Modifications of the parameters in Eq. (2) by varying the experimental conditions. (1) The a_3 's scatter by approximately 25% in a nonsystematic manner. However, they are unaffected by coloring-bleaching-recoloring cycles. (2) The a_2 's differ by approximately 20% at different dose rates. (3) A_3 is unaffected by small strains but increases linearly with strains above a threshold value.

Crystal	Experimental	Parameters				
source	variable	a_L	A_3	$a_{3}(1)$	A_2	a_2
Korth	Dose rate Strain Bleaching	Ua U U	U I(3) U	U D° U	I ^b U I	U(2) D U
Harshaw	Dose rate Strain Bleaching	U U U	U I(3) U	U D U	I U I	U(2) D U
^a U=Unchanged. ^b I=Increased.			ased.	\circ D = Decreased.		

189 m μ absorption increases. The changes in the 185and 190-m μ regions appear to be related to the OH content. In NaCl the peak of the OH absorption has been reported at both 185 m μ^{21} and 190 m μ .²² Also, it has been shown that the OH band decreases as the *F* band increases and that simultaneously a band, attributed to *U* centers, develops on the long-wavelength side of the OH band.²¹ If the *F*-center formation mechanism depends on both the *U*-center and *F*-center concentrations the formation rate could vary as the square of the dose rate.

SUMMARY OF FACTORS AFFECTING THE STAGE-I COLORING

To reiterate, the first-stage coloring curves obtained by exposing NaCl to x-ray irradiation were analyzed by assuming they are composed of a linear part, i.e., the second stage, plus three additional nonlinear components. Two of these components are accurately described by the expression $A\{1-\exp(-at)\}$. The third component is too small to conclusively characterize, but it may be describable by the same expression. The



FIG. 8. The absorption of Korth crystals in the F- and V-band regions after x-ray irradiation at different dose rates.

H. W. Etzel and D. A. Patterson, Phys. Rev. 112, 1112 (1958).
 J. Rolfe, Phys. Rev. Letters 1, 56 (1958).



FIG. 9. The absorptions of Harshaw crystals, in the $185-200-\mu$ m region, as a function of x-ray dose. Shown are curves for "as cleaved" samples and for crystals that had been colored and bleached. The numbers refer to the total dose in units of minutes at 10 mA.

linear part was unaffected or, at most, was changed very little by changing the experimental conditions.

Three experimental variations of the coloring conditions were applied to the samples. The observed changes in coloring behavior are summarized in Table I and additional data are given below:

Dose rate: The parameters a_2 , a_3 , and A_3 remain constant as the dose rate is changed. However, A_2 does depend on the dose rate. This dependence is described by the expression $A_2 = (\text{const})_1 + (\text{const})_2 I^2$, where I is proportional to the x-ray intensity; actually it is the tube current and was varied between 5 and 30 mA.

Optical bleaching: When a colored sample is bleached with white light until all absorption bands have been removed, and then recolored, a_2 , A_3 , a_3 remain constant but A_2 increases.

Plastic deformation: When samples are colored after being strained, and the parameters compared with those obtained from unstrained samples, A_2 is unchanged, a_2 and a_3 both decrease, but the ratio a_2/a_3 remains nearly constant. However, A_3 changes in a somewhat surprising manner. It is unchanged by small strains, but above a certain threshold strain it increases linearly with increasing strain.

It would be interesting to determine if any correlations exist between the parameters describing the three components μ_1 , μ_2 , and μ_3 . Of the three, μ_1 is too small to be considered. Only one correlation having any degree of certainty can be found. This is described immediately above and states that the ratio a_2/a_3 is unaffected by plastic deformation.

DISCUSSION

The data, reported above, describe the first-stage coloring of NaCl in samples from two sources subjected to three different experimental conditions: (1) as cleaved but colored at a variety of x-ray intensities, (2) plastically deformed before coloring, and (3) colored, bleached,

and then recolored. The results obtained by changing these conditions are summarized in Table I. The crystals from the different sources differ in impurity content. Spectrochemical analysis indicates that the Korth material contained Cu, Ti, Fe, and Al impurities, whereas the same analytical procedures did not detect any impurities in the Harshaw crystals. Furthermore, the two materials differed greatly in far ultraviolet absorption both before and after coloring. In the 185- to 200-m μ region, the Korth material is unchanged by irradiation. In contrast, in Harshaw crystals the "OH" band is decreased by radiation and an adjacent band. presumed to be a U band, increases. In the V-band region, the Korth material forms two or more individual bands during irradiation while the Harshaw material shows a small absorption increase not resolvable into bands. The relation between the far-ultraviolet coloring and F-center formation will be considered below.

There is an important point to be emphasized regarding the methods of analysis. The resolution of the data into a linear second stage and a first stage consisting of three "exponential" components would have to be considered fortuitous if it were not for the following: Namely, variations which were introduced by changing the experimental conditions affected only certain constants. In two cases, when the dose rate was varied or the samples bleached and recolored, only one of the parameters changed and the other parameters remained constant. In the other case, when strained and unstrained samples are compared, three parameters changed but two remained constant. Specifically, A_2 is dependent on the dose rate and is altered by the bleaching sequence while a_2 , a_3 , and A_3 all remain unchanged. It was noted above that a_3 appears to fluctuate in a nonsystematic manner with dose rate. Also, the a_3 's varied greatly between crystals from the same block. However, in the bleaching and recoloring experiments the value of a_3 , for each crystal, obtained before and



FIG. 10. The absorption of Harshaw crystals in the F-band and U-band regions after the x-ray irradiation at different dose rates.

after bleaching, was almost exactly the same. This observation supports the conclusion that the coloringbleaching-recoloring cycle leaves all of the constants unchanged except A_2 .

The dependence of A_2 on bleaching and dose rate is consistent with the recently reported^{23,24} formation of vacancies during state-I coloring in KCl. Also, it would appear that two different vacancy-formation mechanisms contribute to stage-I coloring. This follows from the observation that A_2 and A_3 behave differently under the different experimental conditions and that a_2 is 7 to 9 times a_3 .

The separation of the first-stage coloring into three components appears to be consistent with both the first-stage coloring results and annealing studies on KCl irradiated at room temperature.^{19,25} It was shown that the first-stage coloring could be separated into three exponential components. Also, the annealing was describable by bimolecular kinetics and could be separated into three distinct stages. Each stage could be characterized by a well-defined activation energy and pre-exponential factor.

It can be assumed, in accordance with the spectroscopic data, that the Korth material contains more and/or different impurities than the Harshaw material. From this assumption one could conclude that the dependence of A_2 on the square of the dose (I^2), shown in Fig. 7, is related to impurity content. This follows from the observation that $(const)_2$ in the expression A_2 $= (\text{const})_1 + (\text{const})_2 I^2$ is five or six times larger for Korth than for Harshaw crystals.

In addition to the discussion above, which was primarily concerned with bleaching and impurities, some conclusions can be made about the strain-related effects. First, plastic deformation does not introduce additional exponential components. Second, the constants A_3 and A_2 , which can be regarded as the magnitude of the exponential components, are not immediately related to dislocation motion. A_2 is not affected at all by deformation and A_3 is altered only by strains well above the minimum strains known to increase the dislocation

concentration.²⁶ Furthermore, the behavior of A_3 , shown in Fig. 4, is similar to the dependence of M-center formation on strain.²⁷ These observations suggest that the first-stage strain-related processes occur only after a certain minimum strain is exceeded. It is conceivable that prior to irradiation, the crystals have been strained to the extent that a considerable amount of strainrelated processes have already occurred. This might happen during crystal growth or cleaving, or both. Thus, the effects produced by purposeful straining do not occur until the applied strain exceeds the observed minima.

The absorption-spectrum measurements indicate two things. First, that the F-center coloring is related to the centers responsible for far-ultraviolet absorption. And second, they support the contention that the coloring mechanisms are impurity related. The Harshaw and Korth material have different ultraviolet absorption spectra, and change in different ways during irradiation. In Korth crystals, the F and V bands decrease or increase simultaneously as the dose rate is changed. In Harshaw crystals, the U band decreases and the F band increases as the dose rate is changed.

The current data suggest an interesting division between electronic and mechanical effects. The parameter A_2 is altered during bleaching and recoloring, and is also dose-rate-dependent. Both of these processes may be regarded as electronic. In contrast, the other constants, a_2 , a_3 , and A_3 , (a_3 could conceivably be an exception) are modified by plastic deformation but not by the "electronic" processes. Thus, one could speculate that A_3 is dependent on some mechanical property and that A_2 is controlled by an electronic process.

The preceding discussion has been purposely restricted to a few comments on the process of resolving the first-stage coloring into separate components. This work is being extended to include coloring at liquidnitrogen temperature²⁸ and at temperatures between 78 and 300°K. Consequently, the possibility that the various components can be attributed to one or more mechanisms will not be discussed here.

²³ F. Frölich and P. Grau, Phys. Status Solidi 12, 421 (1965). ²⁴ A. Bleckmann and K. Thommen, Z. Physik **191**, (1966)

²⁵ S. Kalbitzer and P. W. Levy, Bull. Am. Phys. Soc. 9, 262 (1964), (to be published).

²⁶ R. W. Davidge and P. L. Pratt, Phys. Status Solidi 6, 759 (1964).

²⁷ F. Agullo-Lopez and P. W. Levy (to be published).

²⁸ P. W. Levy and J. L. Alvarez Rivas, Bull. Am. Phys. Soc. 12, 410 (1967).