



any uv radiation which penetrates the crystal. An enlarged ( $\times 50$ ) image of the first surface of the crystal is formed in the plane of a viewing screen with an aperture stop which passes only green light well within the region of coloration. The sample is translated between successive uv exposures to provide a part of the crystal having no prior damage. Finally, the green interrogation pulses are recorded with a fast vacuum photodiode and a Tektronix 7904 oscilloscope.

In the presentation of our data (Fig. 2), "pulse delay" refers to the time interval  $\Delta$  from the damaging pulse  $UV$  to the interrogation pulse  $G2$ , measured between their centers. Pulse  $G1$ , arriving at the crystal prior to  $UV$ , affords a record of the probe energy incident on the crystal, and  $G3$  records the defect population for a particular pulse  $UV$  as assessed at 10-nsec delay. In Fig. 2,  $D_2/D_3$  refers to the ratio of the optical density computed from pulses  $G2$  and  $G1$  to that computed from  $G3$  and  $G1$ . Mirror  $R2$  is provided with a translation device to enable variation of the delay of  $G2$ . The setting for coincident arrival of  $UV$  and  $G2$  at the sample ( $\Delta = 0$ ) is determined by caliper measurements of length and from the calculated *group* velocities in optical components<sup>7</sup>; the net uncertainty for this position corresponds to about 7 psec.

The rise time of  $D_2/D_3$  in Fig. 2 includes a substantial contribution from the nonzero pulse widths. We have developed integral expressions for  $D_2/D_3$  as a function of the interrogation pulse delay  $\Delta$ , the density  $D_3$  at 10 nsec, the maximal density  $\eta D_3$  ( $\eta$  a parameter  $\geq 1$ ), pulse-width parameters, and a measure  $\tau$  of the intrinsic rise time on some model for the coloration process itself. We write

$$D_2/D_3 = a^{-1} \ln \int_{-\infty}^{\infty} dt G(t) \exp[-\eta a F(t)].$$

Here  $a = 2.303 D_3$ ,  $G(t)$  represents the green pulse, and  $F(t)$  describes the fraction of the peak density  $\eta D_3$  which has developed by time  $t$ .  $F(t)$  is given by

$$F(t) = \int_{-\infty}^t dt' \mathfrak{F}(t-t') C(t'+\Delta),$$

where  $\mathfrak{F}(t-t')$  describes the intrinsic coloration process and  $C(t'+\Delta)$  represents the generation of carriers. For the present analysis  $G$  and  $C$  are taken to be normalized Gaussians.<sup>7</sup> For  $\mathfrak{F}(t-t')$  we have considered the exponential form  $1 - \exp[-(t-t')/\tau_e]$ , and also a step function. In either case an analytic approximation for the error function is useful.

The curve in Fig. 2 is a two-parameter variational best fit with the exponential form for  $\mathfrak{F}$ ,

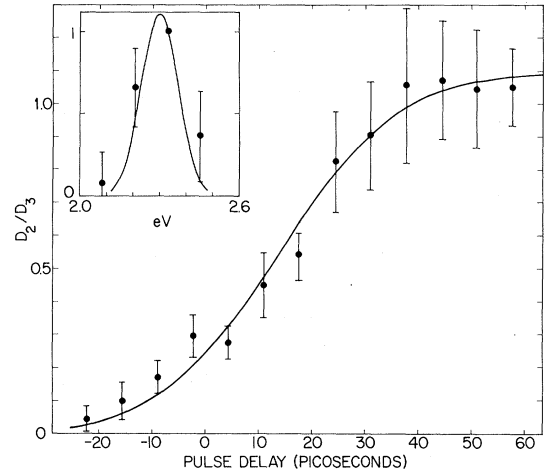


FIG. 2. For KCl at  $T \approx 25$  K, we plot fractional optical density ( $D_2/D_3$ ) at 532 nm versus delay after the 266-nm pulse. The density  $D_3$  is measured at 10 nsec. The curve is a best-fit convolution of pulse shapes with an exponential form for defect production, yielding  $\tau \approx 11$  psec. Error bars denote  $\pm 1$  standard deviation about the mean. Inset: A four-point absorption spectrum measured about 85 psec after uv excitation; the curve is the normal  $F$  band at 25 K.

yielding  $\tau_e = 11$  psec and  $\eta = 1.10$ ; the standard error is  $\sigma = 0.05$ . The pulse widths (full width at half-maximum, FWHM) employed were  $W_G = 32 \pm 7$  psec, from measurements with an Electro-Photonics streak camera, and  $W_C = 18.5$  psec.<sup>7</sup> In the part of the growth curve in which the defect population changes significantly within a pulse width, the shape retains some dependence upon the maximal density and so upon  $D_3$ . The plotted curve employs  $D_3 = 0.74$ , the average from our useful data. Within the useful range  $0.05 \leq D_3 \leq 1.5$ , the curves obtained do not depart from this curve by more than 0.1, or  $2\sigma$ . Variation of  $W_G$  produces little improvement of fit. Similar results are found for the step-function choice of  $\mathfrak{F}$ . Considering the uncertainty in the curve fitting in addition to that in determination of the position of  $R2$  for  $\Delta = 0$ , we attach an overall uncertainty of 9 psec to the rise time  $\tau$ .

In our experiment, a fraction  $5 \times 10^{-5}$  of the 532-nm energy reaches the sample. In the absence of the  $\times 10^{-2}$  attenuator ( $F1$  in Fig. 1), definite effects of saturation are evident. There may be residual effects of this kind even with  $F1$  in place. The value  $\eta - 1 = 0.10$  is believed to be an indication of depletion of the ground-state  $F$ -center population by the pulse  $G2$ , but in principle it encompasses annealing effects as well.

The decay of absorption is not monitored be-

yond 10 nsec. Low-temperature decay characteristics at later times are probably similar to those observed after electron-pulse excitation.<sup>2,8</sup> In that work there was found nonexponential decay with principal components having  $\tau \leq 20 \mu\text{sec}$ , leaving a stable  $F$  band  $\approx 10\%$  of the peak.<sup>2</sup> Less detailed measurements performed with the sample at room temperature yield a growth curve very similar to that observed at low temperature, but the absorption decays to about 50% of its peak value within 8 nsec. The decay is attributed to pairwise annealing of  $F$  and  $H$  centers.

We have made a comparison of the spectral profile of early absorption with the normal  $F$  band, using, in addition to the 532-nm second harmonic, three other wavelengths derived by stimulated Raman scattering in benzene (inset, Fig. 2). Interrogation at 532 nm was performed 70 psec after the uv pulse. The Raman-shifted pulses may be delayed an additional 30 psec, but the absorption has little time dependence for  $\Delta > 40$  psec. We have plotted the absorption at each Raman wavelength relative to that at 532 nm. The solid curve is a Gaussian with FWHM and peak appropriate to the KCl  $F$  band at 25 K, the estimated local temperature (see below). The fit is sufficiently close to identify the absorption with  $F$  centers. Laser pulse heating cannot account for the evident broadening, since a temperature of about 110 K would be required. The broadening may well be due to perturbing effects of  $H$  centers very near some of the  $F$  centers, in the population of transient pairs. In the electron-pulse experiments, there was found a background absorption due to self-trapped excitons.<sup>2</sup>

Coloration-depth profiles produced in a thick crystal by pulses of varying power are consistent with absorption of uv energy by a two-quantum process. Transmission measurements show that about 30% of the incident uv pulse energy is deposited in our 0.8-mm-thick sample. In the first 0.4 mm of the sample, where the coloration is most dense, an energy density of approximately  $1 \text{ J/cm}^3$  is deposited in a typical shot. For an equilibrium sample temperature of 10 K, we estimate that the local temperature rises only to about 25 K. Because of divergence of the focused beam as well as pulse depletion, the profile of coloration versus depth is nonuniform. The average  $F$ -center density produced in the first 0.4 mm of the sample in our low-temperature work is estimated<sup>9</sup> to be  $10^{17} \text{ cm}^{-3}$ , and a similar optical density at room temperature required about  $2 \times 10^{17} \text{ cm}^{-3}$ .

We have characterized the ambient vacancy concentration in our pure KCl<sup>10</sup> by studying the first coloration stage under room-temperature x irradiation, following Mitchell, Wiegand, and Smoluchowski.<sup>11</sup> The first stage saturates at about  $1.8 \times 10^{16} \text{ cm}^{-3}$ . Furthermore, at low temperature it has been demonstrated that  $F$  centers formed by charge transfer to vacancies must pass through the first excited state, delaying the 532-nm ground-state absorption by the 0.6- $\mu\text{sec}$  excited-state lifetime.<sup>12</sup> Thus the absorption observed in the present experiment reflects the creation of new lattice defects rather than the transfer of electrons onto preexisting vacancies.

Our results make possible determination of the efficiency for production of Frenkel defects by recombination of electrons and holes in KCl. For two-quantum absorption, approximately 60 eV of energy deposition is required per  $F$  center formed (at either temperature); i.e., about 15% of electron-hole recombinations lead to creation of a Frenkel defect. This agrees with a value 17% ( $\pm 8\%$  uncertainty on each efficiency) based on electron-pulse studies of coloration,<sup>2,5</sup> using an estimate of 25 eV required per carrier pair created by energetic electrons.<sup>13</sup> Still higher efficiency can be inferred for KBr.<sup>1,2,5</sup>

In conclusion, within about 11 psec after excitation the system relaxes by carrier thermalization through  $\sim 0.8 \text{ eV}$ , and by exciton formation and nonradiative transitions through  $\sim 1 \text{ eV}$ , culminating with ejection of  $\text{Cl}^0$  to form an  $F$  center in its ground state. There is little time for relaxation across large electronic gaps. Since the first four levels of the self-trapped exciton are separated typically by more than five phonons, this suggests that halide motion leading to defect formation starts from fairly high-lying electronic levels of the STE. On the other hand, a certain compactness of the electron orbital is required in the most recently proposed mechanisms.<sup>4-6</sup> The electron is viewed as settling into the developing vacancy, providing stabilization as the  $\text{Cl}_2^-$  molecular ion translates *as a unit* along a [110] direction. More than one STE state may have the requisite symmetry and compactness to initiate this odd-parity relaxation. It is likely that the intermediate states of the system are not well described in the adiabatic approximation.

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<sup>7</sup>Because of group-velocity mismatch in the redoubler, the emergent pulse UV contains components with 0- to 13-psec transit delay relative to G2. This necessitates a correction of about -7 psec to the  $\Delta$  values

from caliper measurements. Also it contributes to the pulse widths for UV and for carrier generation. In our analysis with Gaussian line shapes, we have taken  $W_{UV} = (\frac{1}{2}W_G^2 + 13^2)^{1/2}$ , and  $W_C = 2^{-1/2}W_{UV}$ .

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## Effect of Transverse Magnetic Fields on dc Josephson Current\*

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We have observed diffraction patterns in critical dc Josephson tunnel current versus magnetic field applied perpendicular to the junction plane. The patterns can be explained by assuming that the perpendicular field penetrates the edge of each film and channels through the junction parallel to the films. An "edge penetration depth,"  $\lambda_{\perp}$ , gives a quantitative description of the patterns and their dependence on the angle of field direction. We have found  $\lambda_{\perp}(T) \propto (1 - T/T_c)^{-1/2}$ .

If a magnetic field is applied parallel to the plane of a Josephson tunnel junction, the zero-voltage critical current ( $I_c$ ) as a function of magnetic field ( $H$ ) will exhibit a diffractionlike pattern.<sup>1,2</sup> Existing theory<sup>2-4</sup> shows that  $I_c$  is given by

$$I_c = I_J \left| \frac{\sin(\pi H L d / \Phi_0)}{\pi H L d / \Phi_0} \right|, \quad (1)$$

where  $I_J$  is the Josephson zero-voltage critical current in zero magnetic field,  $L$  is the dimension of the junction perpendicular to  $H$ ,  $d = 2\lambda_{\parallel} + l \approx 2\lambda_{\parallel}$  with  $\lambda_{\parallel}$  the surface penetration depth and  $l$  the oxide thickness, and  $\Phi_0$  is the magnetic flux quantum equal to  $2 \times 10^{-7}$  G cm<sup>2</sup>. The critical current goes to zero whenever the junction contains an integral number of flux quanta, i.e., for the  $n$ th zero the magnetic field satisfies

$$H_n = n\Phi_0 / Ld, \quad n = 1, 2, \dots \quad (2)$$

According to this theory, if the magnetic field is applied at an angle  $\theta$  with respect to the plane of

the junction (see inset of Fig. 1) the magnetic field necessary to produce the  $n$ th minimum of

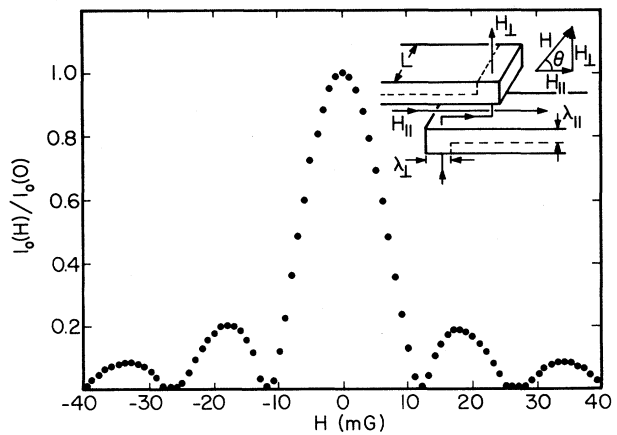


FIG. 1. Typical diffraction pattern for  $\theta = 90^\circ$  at  $T = 2.50$  K. Junction dimensions are  $0.74 \text{ mm} \times 0.35 \text{ mm}$ . The inset shows the magnetic-field orientation relative to the junction.