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F-center production in alkali halide crystals by monochromatic x-ray and ultraviolet radiation

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The spectral dependence of the F-center production efficiency was observed in the hard-x-ray region at the K edge of bromine in KBr and in the near-ultraviolet part of the spectrum corresponding to the exciton region of several alkali halides. Evidence of a definite increase at the core threshold was observed as well as a significant wavelength dependence of the F-center production efficiency in the near-uv region. These studies were made possible by the development of an extremely sensitive method of detecting F centers in ultralow concentrations, which utilizes the luminescent properties of these centers.

I. INTRODUCTION

The production of defects by radiation in insulating crystals has been studied for several decades, and these investigations have produced a wealth of information on the formation of defects such as the F center. Unfortunately, the great majority of experiments on the production of defects by radiation have been performed with polychromatic radiation sources. Especially in the x-ray region, virtually nothing is known about the spectral dependence of the F-center production efficiency. It has been assumed that this efficiency is about one F center per 1000 eV of energy and that it varies slowly, if at all, throughout the spectrum. Experiments using monochromatic radiation sources have been very few due to inherent difficulties. For example, the traditional method of detecting F centers is by measuring optical absorption, yet such measurements are relatively insensitive to concentrations below 10^{15} cm^{-3}. Above 10^{15} cm^{-3}, however, saturation effects begin to set in, complicating the initial defect-formation processes. It is also true that monochromatic radiation sources are often of low flux and cannot produce large enough concentrations of F centers to be detectable by optical absorption measurements.

Because of these limitations, it was necessary to develop a method of detecting F centers in ultralow concentrations. Utilizing the luminescence properties of F centers and the effective separation of excitation and emission wavelengths, we developed such a method capable of detecting F-center concentrations several orders of magnitude smaller than are detectable by optical absorption. This has enabled us to observe the production of F-centers in a new regime: the early part of the so-called stage-I region, where it has been supposed that the production rate is constant. We have employed this technique to investigate the spectral dependence of the F-center production efficiency using relatively-low-flux monochromatic sources.

Theoretical models to describe the mechanisms by which F centers are produced were proposed thirty years ago by Seitz[1] and Varley.[2] These early proposals were soon followed by revised models, in efforts to satisfactorily explain the increasing body of experimental evidence on the production of F centers. Following suggestions of Pooley[3] and Hersh,[4] it is now generally believed that the mechanism of F-center production by energetic radiation involves a nonradiative recombination of a self-trapped exciton (STE). At low temperature the result is the formation of F and H centers with high efficiency. From which excited state of the self-trapped exciton the F-center production actually proceeds is still a matter of discussion. Several STE models employing different excited states have been proposed. For materials such as KI, RbI, NaBr, and LiCl, the temperature dependence of luminescence from the lowest triplet state of the STE is anticorrelated with F-center yield. This strongly supports the lowest triplet state as the precursor.[5,6] However, for materials such as KBr, KCl, and RbBr at low temperatures, time-resolved studies have shown that F centers are formed in the ground state within 10 ps after generation of the electron-hole pair. This is much faster than the STE luminescence decay time of about 5 ms.[7] Therefore, for these materials at low temperatures, the lowest triplet state can hardly be the precursor, and another model involving a higher excited state of the STE may be applicable.

In 1973, Kabler[8] and Itoh and Saidoh[9] suggested independently that the F center evolves by a translational motion of the X_2^- molecular ion in a $\langle 110 \rangle$ direction, following an adiabatic potential curve directly from a higher excited state of the STE. More recently, Toyozawa[10] proposed that a highly excited electronic state of the STE caused a Jahn-Teller-type instability which leads to ejection of the molecular ion along a $\langle 110 \rangle$ direction. Alternatively, Kabler and Williams[11] proposed that the molecular ion need not move precisely along a $\langle 110 \rangle$ direction, but may overcome the repulsive potential barrier by rotating in a $\langle 111 \rangle$ direction.

In all of these models, the end result is an isolated F center and an interstitial halogen atom (H center) removed from the neighborhood of the F center. In a recent work by Comins and Carragher,[12] the trap for the halogen atom has been identified as a divalent impurity-vacancy (IV) dipole or an aggregate of such dipoles. This model predicts a linear region of *F*-center production (stage I) corresponding to the stage in which singly occupied halogen traps are being created. For higher radiation doses, the model predicts a production rate approximately dependent upon the square root of radiation dose (stage II), in which a radiation-induced detrapping mechanism competes with the *F*-center stabilization process.

In addition to the aforementioned band-gap exciton mechanisms, other possibilities come to mind for photon energies corresponding to core-edge thresholds. For example, Varley[13] proposed that *F*-center production could occur following excitation in the x-ray region through a multiple ionization of the core. He argued that if an anion in the crystal were to have more than one electron simultaneously removed from it by a process of multiple ionization, it would temporarily become a positive ion residing in a negative-ion site in the normal ionic lattice. Since the lattice ions vibrate about their mean positions, such a multiply ionized anion might be ejected by Coulombic repulsion into an interstitial position, which to a first approximation is an electrostatically neutral position with respect to the rest of the lattice. This would lead to the formation of a separated *F-H* pair through the capture of electrons by the resultant anion vacancy and the ionized interstitial halogen.

Yet another mechanism of *F*-center production at core-edge thresholds is possible in the region of energy corresponding to core excitons. Such excitations occur due to the Coulombic attraction between the excited electron and the inner-shell hole left behind. This electron-hole pair is a composite particle and, according to Kotani and Toyozawa,[14] should be self-trapped or *S*-type in nature. Again, localization occurs due to the distortion of the surrounding lattice. The self-trapped nature of the core exciton suggests the possibility of a self-trapped excitonic *F*-center production mechanism, in analogy to the STE mechanism that occurs for valence excitons.

At present, there is a controversy resulting from conflicting data as to whether the *F*-center production efficiency varies at the core-edge thresholds. Sharma and Smoluchowski[15] reported the observation of a factor of 2.5 increase in the *F*-center production efficiency in KCl for photon energies above the *K* edge of chlorine compared to photon energies below that edge. They also observed a factor of 1.1 increase in KBr for photon energies above the *K* edge of bromine compared to photon energies below the edge. Cruz-Vidal and Gomberg,[16] however, investigated the production efficiencies of the same core-edge thresholds and observed no increase in either case. There are difficulties in the experimental methods of both groups which may be responsible for the conflicting data, indicating that improved experiments need to be carried out. In the next section we outline the laser-induced luminescence method which allows the detection of very low *F*-center concentrations in the early stage I region.

II. EXPERIMENTAL TECHNIQUES

A schematic for the laser-induced luminescence method, as applied to the monochromatic x-ray experiments, is given in Fig. 1. An alkali halide crystal is freshly cleaved to an average size of about $1\times1\times0.3$ cm^3, mounted on the cold finger of a liquid-nitrogen cryostat, and then cooled to liquid-nitrogen temperature under vacuum. The *F*-center luminescence is excited by *F*-band light with the appropriate laser line of an argon-krypton ion laser or a helium-neon laser. Light from the laser is filtered by an appropriate narrow-band laser line filter *A* in Fig. 1. The laser beam is focused to about 1 mm in diameter, chopped at a frequency of 1 kHz by an electromechanical oscillator, and then refocused onto the end of an all-silica optical fiber. The other end of the optical fiber passes through the side of the cryostat and is directed onto the thin dimension of the crystal. A reference signal from the oscillator power supply is amplified and connected to the reference channel of a lock-in amplifier. The input of the lock-in comes from a tuned 1-kHz preamplifier, which amplifies the Stokes-shifted infrared *F*-center emission detected by a PbS photoconductive cell. A sharp-cut low-pass infrared filter *B* effects good separation between excitation and emission of the *F* center. A similar technique, but much less sensitive, was employed by Maki, Nagasawa, and Hirai.[17]

In the monochromatic x-ray experiments, *F* centers were produced in the sample with an x-ray beam produced by an extended x-ray-absorption fine-structure (EXAFS) spectrometer[18] mounted on a 12-kW rotating-anode x-ray machine. The focusing bent-crystal spectrometer is controlled by a minicomputer and can provide x-ray fluxes of 10^6 to 10^7 photons/sec in an energy range between 4 and 20 keV. The spectrometer, as shown in

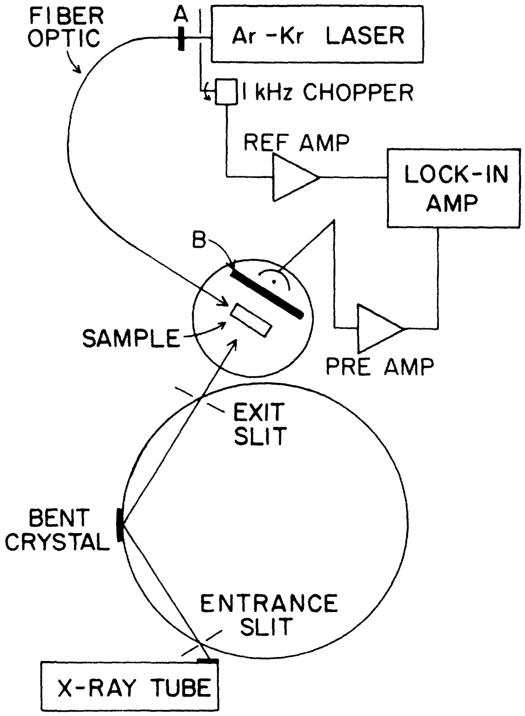

FIG. 1. Schematic of the laser-induced luminescence instrumentation installed on the EXAFS spectrometer.

Fig. 1, consisted of an entrance slit placed at the x-ray anode image, a bent Si crystal with beam flag, and an exit slit, all on a Rowland circle. The exit slit and liquid-nitrogen dewar are mounted on a single table, which is moved by a computer-controlled stepping motor, allowing the optical fiber to move as the x-ray energy is scanned and keeping the laser flux on the sample constant. The bent crystal is moved by a second stepping motor, such that the desired x-ray energy is selected and incident upon the sample. The incident x-ray flux was monitored by an ionization chamber filled with enough argon gas to block approximately 5% of the x rays. The resolution of the x-ray beam was determined to be 30 eV at full width at half maximum.

The *F*-center production efficiency experiments in the ultraviolet region of the spectrum employed the laser-induced luminescence instrumentation as shown in Fig. 1, but with the x-ray source and monochromator replaced by a 60-W deuterium lamp attached to an ultraviolet grating monochromator. Both the lamp housing and grating monochromator were fluxed with dry nitrogen to minimize oxygen absorption at short wavelengths. Utraviolet light entered the liquid-nitrogen cryostat through a Suprasil fused-silica window, and the incident photon flux was monitored by an IP28 photomultiplier tube and sodium salicylate screen.

The relative yield of *F* centers $Y_F(E)$ at a given photon energy E is defined as

$$Y_F(E)=\frac{I_F(E)}{I_{\text{ex}}(E)I_L}$$

where $I_F(E)$ is the observed luminescence signal at energy E, $I_{\text{ex}}(E)$ is the absorbed exciting x-ray or ultraviolet light intensity at energy E, and I_L the laser light intensity. In other words, the observed lock-in signal could be corrected for changes in the defect producing radiation and the exciting laser light. In practice, the intensity I_L was relatively constant.

F centers could also be produced in the volume by penetrating x rays from a conventional 100-kV x-ray tube. This was used to test the laser-induced luminescence method and for purposes of calibration. Figure 2 shows the growth of luminescence signal I_F with time for a previously uncolored crystal of KBr at 80 K irradiated with polychromatic x rays filtered by 3.2 mm of aluminum. In this case, the voltage of the tungsten-target x-ray tube was 100 kV and anode current 2 mA. The 647.1-nm output of an argon-krypton mixed-gas laser was used to excite the luminescence. Initially the luminescent signal increases linearly for two to three orders of magnitude. This is the so-called "stage-I" region. Thereafter the signal or color-center density begins to saturate in the so-called "stage-II" region. No further increase or "stage III" is observed, as previously noted for coloration of alkali halides at low or intermediate temperatures.[19,20] Presumably the saturation of permanent *F*-center density is due mainly to the exhaustion of halogen traps, for example, impurity-vacancy complexes as suggested by Comins and Carragher.[12] The experiments using monochromatic x rays and also low-intensity ultraviolet light were all carried out in the linear stage-I region.

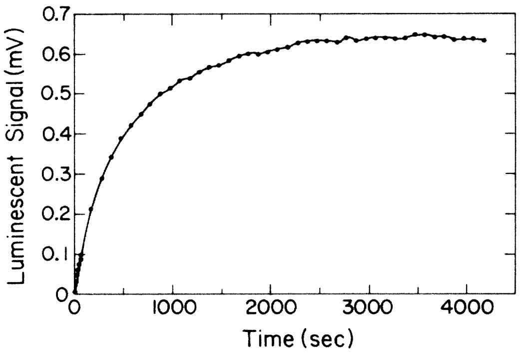

FIG. 2. Growth curve of laser-induced luminescence for a nominally pure KBr crystal as it is irradiated by polychromatic x rays from a 100-kV x-ray source.

The laser-induced luminescence method was calibrated by irradiating an uncolored sample of KBr with filtered polychromatic x rays from a 100-kV x-ray tube until the luminescent signal had increased several orders of magnitude to saturation, corresponding to a measurable color-center density. The *F*-band absorption was then measured with a spectrophotometer and the maximum optical density was determined to correspond to a final *F*-center concentration of about 2×10^{15} cm^{-3}. This yielded a calibration constant of 1×10^{6} (*F* centers)/pV before correcting for laser intensity. Assuming that the luminescent signal is proportional to the *F*-center concentration down to very low concentrations, the lowest detectable signals corresponded to an initial *F*-center concentration of about 5×10^{10} cm^{-3}. The signal corresponding to this level before x-ray exposure was usually due to roughly equal parts stray or scattered light and a residual coloration which depended upon the history of the samples used. Most work was carried out on nominally pure Harshaw crystals.

III. RESULTS AND DISCUSSION

A. Bromine *K* edge

The rotating-anode x-ray source and monochromator could also be used to take absorption spectra at various inner-level thresholds. For this purpose, a second ion chamber was mounted so as to record the transmission of x rays through a thin alkali halide sample. Figure 3 shows the optical density (proportional to absorption coefficient times thickness) of KBr in the vicinity of the bromine *K* edge at 13 475 eV. The near edge and the extended x-ray absorption fine structure (EXAFS) can be seen above threshold. These data agree with the absorption spectrum of KBr taken by Lagarde[21] using synchrotron radiation except that the prominent spike or core exciton, which occurs at the very threshold, is not resolved due to a spectral bandwidth of about 20 eV. The high-resolution synchrotron radiation data at the very edge is shown by a dotted line in Fig. 2. We now turn to observation of the spectral dependence of *F*-center production through the bromine *K* edge in KBr.

Figure 4 shows the slope of the laser-induced lumines-

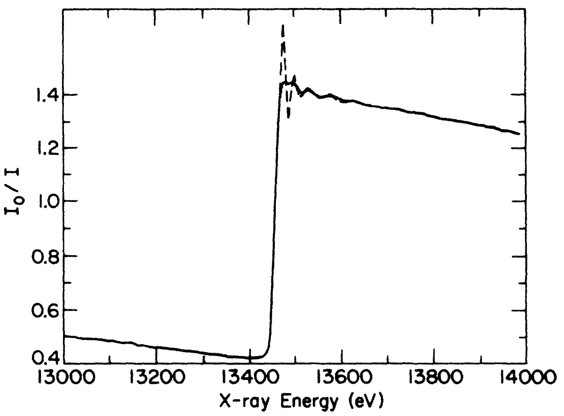

FIG. 3. X-ray absorption spectrum of KBr at room temperature as measured by the rotating anode x-ray source and bent-crystal monochromator. Spectral band width is about 20 eV. The dotted line shows the sharp structure observed by Lagarde (Ref. 21) using very-narrow-band synchrotron radiation.

cence signal *versus* time for x-ray energy between 13 000 and 14 000 eV. This figure is a composite of several runs on two different samples of nominally pure KBr, in which the separate runs have been scaled to the same general level about the edge. The curve drawn through the points represents the average spectral dependence of the *F*-center production efficiency. The final *F*-center densities achieved with the weak monochromatic x-ray beam were of the order of 10^{11} centers/cm^3, within the early stage-I region of coloration. Notice that the slope of the luminescent signal is plotted as a function of x-ray energy. Considering that the incident x-ray flux on the sample was only about 10^6 photons/sec, it is not surprising that it required approximately one hour to obtain enough data for a reasonable determination of the luminescent signal slope represented by each point. Even so, the slopes presented in Fig. 4 display a considerable amount of scattering, limiting the information on the spectral dependence of *F*-center production efficiency to gross features only.

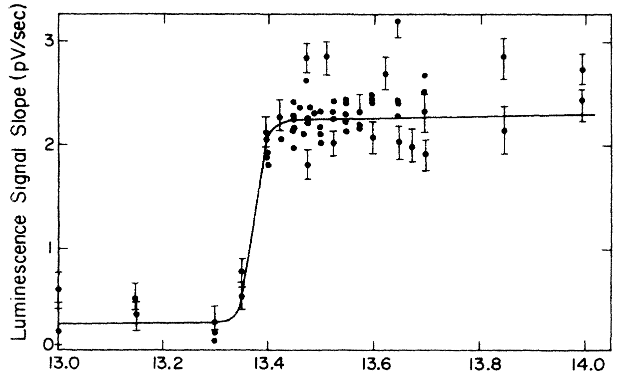

FIG. 4. *F*-center production efficiency versus x-ray energy for KBr at liquid-nitrogen temperature.

A significant increase near threshold with a step height of about eight to one is clearly evident in Fig. 4. On either side of the edge, the production efficiency is reasonably constant. Because the samples were of the order of 3 mm thick and the absorption coefficient varied from about 40 cm^{-1} below the edge to about 300 cm^{-1} above the edge, the x-ray radiation was totally absorbed by the crystal both below and above the edge in the experiment. Thus if the *F*-center production efficiency is approximately constant above and below the edge it is easy to understand the constant slopes observed even though the absorption coefficient decreases slowly in these regions.

We believe the large step height observed is clear evidence of a significant increase in *F*-center production efficiency as the x-ray energy is tuned through the *K*-edge of bromine in KBr. This is in qualitative agreement with the data of Sharma and Smoluchowski,[15] who reported an increase, but by a factor of only 1.1 for KBr and 2.5 for KCl for the chlorine *K* edge. Our result is in direct contradiction to the results published by Cruz-Vidal and Gomberg,[16] who observed no increase at all above these edges. The results of both groups are probably influenced by the fact that coloration was observed in the stage-II region, where the effect of the competing radiation-induced detrapping process is large,[12] and dependent upon x-ray intensity. Very-wide-band x-ray sources were also used.

Figure 4 shows that the average luminescent signal slope below the edge is about 0.3 pV/sec; above the edge it is about 2.3 pV/sec. Using the calibrated value of the sensitivity of the laser-induced luminescence method (1×10^6 *F* centers/pV) this yields production efficiencies of 3×10^5 (*F* centers)/sec below the edge, and 2.3×10^6 (*F* centers)/sec above the edge. The incident x-ray photon flux was about 7.5×10^5 photons/sec, therefore the *F*-center production efficiency below the edge is 0.4 *F* centers/photon and 3 *F* centers/photon above the edge. Since approximately 50% of the absorbed x radiation above the edge is reemitted as x-ray fluorescence, this efficiency corresponds to roughly 2000 eV/*F* center created above the edge. Thus above the edge, several *F* centers are created for each x-ray photon absorbed.

The observation that more than one *F* center/photon is created for energies greater than 13 475 eV, and less than one below the edge, makes it difficult to attribute the increase simply to the Varley mechanism.[2] This mechanism predicts that a single *F* center is produced for each *K*-shell ionization as a result of multiple ionization of the anion by an Auger cascade, followed by a Coulombic expulsion of the anion from its lattice site. On the other hand, our observed multiple *F*-center production above the edge is still probably due to the production of several electron-hole pairs by the Auger cascade, following ionization of the core. These electron-hole pairs recombine by the typical radiationless recombination process known to be responsible for efficient *F*-center production at much lower energies.[8–10] In fact, a detailed comparison can be made with the known vacancy cascade in krypton, a closed shell atom such as Br$^-$. An Auger cascade occurs for approximately 50% of the *K*-shell ionizations in this case, and, according to Krause,[22] this produces 6–7 electron-hole pairs per cascade event. Therefore, the observed step height of about eight corresponds with the additional production of *e-h* pairs above the edge. Also, the *efficiency* for formation of *F* centers above the edge, about 3 *F* centers/photon, agrees with the total number of electron-hole pairs produced above the edge (50% Auger cascade processes per photon resulting in 6–7 *e-h* pairs per process).

It is perhaps not surprising that we see no evidence for a spike near threshold in the observed *F*-center production efficiency. The resolution of our apparatus is just not sufficient to resolve the influence of the peak in the spectrum as narrow as reported by Lagarde.[21] Therefore, we are not able to make any determination of the presence of entirely new channels of defect formation related to the core exciton from the present experiments. On the other hand, an intense well monochromated synchrotron source might allow the observation of additional spectral detail.

B. Near-ultraviolet region

The efficiency of *F*-center production in nominally pure KBr, KI, and RbBr was studied as a function of photon energy in the near ultraviolet region of the spectrum between 180 and 300 nm. Figure 5 shows the *F*-center production efficiency (slope of luminescent signal) versus wavelength for a previously colored sample of nominally pure KBr. The solid circles represent the first spectrum taken on the sample that day and the open circles represent the second spectrum taken immediately after the first.

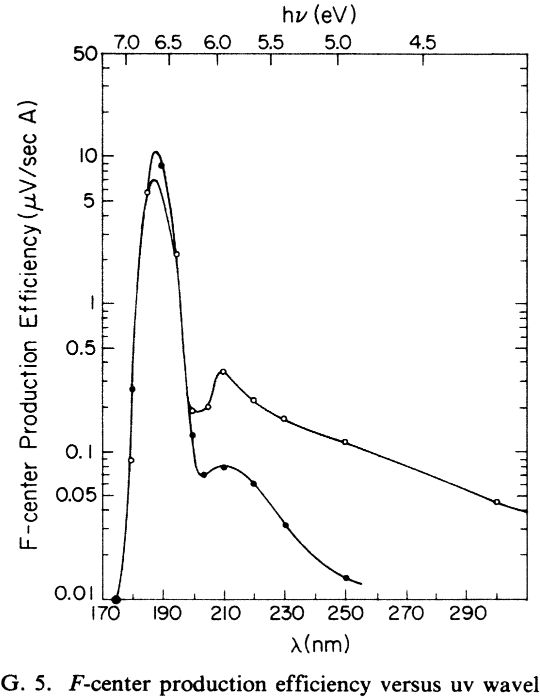

FIG. 5. *F*-center production efficiency versus uv wavelength for a previously colored sample of KBr at liquid-nitrogen temperature. Note that a log scale is used on the vertical axis. The lower curve is the first spectrum taken. The upper curve (open circles) was taken immediately after the first and shows an increase due to α centers associated with laser bleaching as discussed in the text.

The most prominent feature of the spectra shown in Fig. 5 is the large peak at about 188 nm. This feature decreases rapidly as the uv photon energy is increased, and is accompanied by a much smaller peak on its long wavelength side at about 210 nm. The 188-nm peak agrees well with the maximum in the exciton absorption tail predicted by the model of Kristianpoller and Israeli and observed in their thermoluminescence measurements.[23] The maximum does not occur at the peak of the first exciton at 183 nm,[24] but rather on its long-wavelength tail, due to the effect of the high absorbance and reflectance at the exciton peak. Such high absorption causes the uv radiation that is absorbed to be effective in only a very thin layer at the surface. Even though our crystals were freshly cleaved before mounting in the cryostat, impurities quickly form during the mounting procedure, and they might provide an alternative channel of deexcitation for electrons and holes. Another possibility is that saturation occurs due to exhaustion of halogen traps at high color center density close to the surface.

Kristianpoller and Israeli[23] reported that the thermoluminescent peak formed by irradiating in the exciton tail displayed a linear dependence on irradiation dose. This suggests that the 188-nm peak is not due to the exhaustion of halogen traps near the surface, otherwise a nonlinear dose dependence should have been observed. We observed a linear dose dependence in the *F*-center production efficiency at the maximum production wavelength for over an order of magnitude. The shift of the *F*-center production efficiency maximum to longer wavelengths is not totally explainable by the rapidly changing reflectance, either. According to Philipp and Ehrenreich, the reflectance of KBr changes from about 4% in the visible to a maximum of about 30% in the vicinity of the first exciton peak.[25] This means that the changing reflectance could only account for, at best, a factor of 1.5 difference in the *F*-center production at these wavelengths. We observed, however, a factor of about 6 difference in the *F*-center production maximum compared to the production at the exciton peak. Thus, the number of *F* centers produced in the crystal at the exciton peak is less than that for uv photons in the tail of the exciton peak, most likely due to a surface contaminant effect. This spectrum is, to our knowledge, the first direct observation of a maximum in the measured wavelength dependence of the *F*-center production efficiency in the near-uv region.

An estimate of the absolute *F*-center production efficiency in the 188-nm peak was obtained in the following way: Figure 5 gives a peak efficiency of approximately 10 μV/sec/A at 188 nm, corresponding to a luminescent signal of 660 pV/sec. Using the calibrated value of 1×10^6 (*F* centers)/pV for the luminescent signal, we calculate that approximately 7×10^8 (*F* centers)/sec were created. We calculated the monochromatic deuterium lamp flux to be about 10^{11} photons/sec in the region of 190 nm. This yields a production efficiency of about 0.007 (*F* centers)/photon or about 1000 eV/*F* center, which is the generally accepted average expenditure of energy for the creation of an *F* center at low temperatures.

A second, smaller peak can be seen in the first spectrum of Fig. 5 at about 210 nm, approximately two orders of magnitude smaller than the peak at 188 nm. In the second spectrum, an increase in the 210-nm peak is observed, such that it is smaller than the 188-nm peak by a factor of 30. This peak corresponds to *F*-center production due to irradiation into the vicinity of the alpha band peak at 203 nm. The observed *F*-center production in the α band is confirmation that an analogous excitonic mechanism is responsible for defect creation in the vicinity of a preexisting vacancy, as previously indicated by TL measurements.[23] This can be viewed as *F*-center production by radiationless recombination of excitons perturbed by vacancies. The fact that we observe an increase in *F*-center production in the α band in the second run is explained by the phenomenon of laser bleaching. In the first run an additional population of *F* centers was created by the near uv light. During the experiment, the sample was continually illuminated by *F*-band light from the laser, which converted a significant fraction of the newly created *F* centers to F' and α centers. This caused a net increase in the α center concentration in the crystal, and the second run gives evidence of this by the increased *F*-center production at the α band.

We next investigated *F*-center production in the near uv in a sample of nominally pure KI. This was of interest because the first exciton peak in KI is at a much longer wavelength, allowing us to probe the production efficiency on both the low- and high-energy side of the first exciton peak.

The solid line and points in Fig. 6 show the *F*-center production efficiency in KI for near uv photons in the range between 180 and 300 nm. Also shown by a dashed line is the well-known optical density of KI at 77 K.[26] Note that there is no appreciable defect production for wavelengths above 250 nm, but below this, four peaks are evident. The main peak is at 218 nm, followed by one at 208 nm and one at 193 nm. On the long-wavelength side of the main peak, a smaller one appears at about 236 nm. The minimum between the two largest maxima occurs exactly at 213 nm, the exact position of the exciton peak! This is however, precisely as expected according to the model of Kristianpoller and Israeli.[23] Because of the high absorbance and reflectance at the exciton peak, one would expect to see maxima in the *F*-center production efficiency on either side of the exciton peak, as observed.

The smallest peak at about 236 nm is two orders of magnitude smaller than the largest peak and lies in the region of the α-band peak in KI at 238 nm. This production maximum again gives evidence of *F*-center production in KI by excitons perturbed by vacancies. The highest energy peak observed at 193 nm is also two orders of magnitude smaller than the largest peak. It lies at a relative minimum in the transition region between the excitonic part of the absorption spectrum and the band-to-band region. The minimum between it and the 208-nm peak occurs at the local maximum observed in the absorption spectrum at 200 nm. In this part of the spectrum, the absorbance and reflectance are very high and so one would expect an "inverse" production curve when compared to the absorption curve in this region. Such a phenomenon was observed in the excitation spectrum of certain glow peaks in KBr,[27] and was attributed to the variation of the penetration depth of the radiation. Our observations in the transition part of the spectrum support this concept.

A similar behavior of the *F*-center production efficiency in the near uv was also found in nominally pure RbBr. Figure 7 shows the production efficiency for RbBr be-

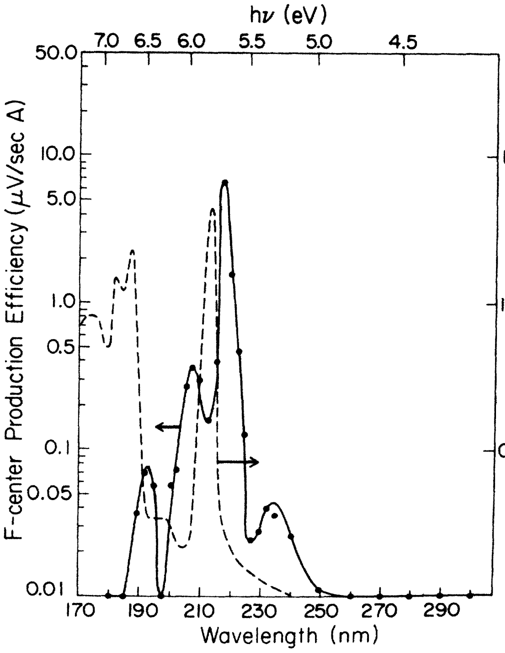

FIG. 6. *F*-center production efficiency versus uv wavelength for KI at liquid-nitrogen temperature. The dashed line shows the optical density of a thin film of KI measured by Eby, Teegarden, and Dutton at 77 K (Ref. 26).

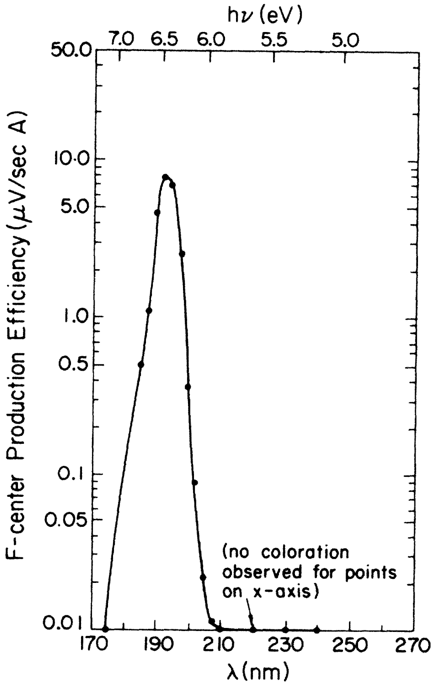

FIG. 7. *F*-center production efficiency versus uv wavelength for RbBr at liquid-nitrogen temperature.

tween 170 and 280 nm. No production was observed for wavelengths greater than 210 nm, but a large single peak was observed at 194 nm. The first exciton peak is at 188 nm, at which wavelength the production efficiency has already decreased by a factor of 20. Thus, this material also shows an F-center production peak in the exciton tail, with a sharply reduced production at the exciton peak itself.

There is no evidence of additional production at the α band in RbBr at 205 nm. Although the F-center production at this wavelength is still measurable, it is two orders of magnitude smaller than the production peak. It may be that the efficiency of production in the α band is smaller in RbBr than for KBr and KI, and therefore does not make a measurable contribution to the total F production at this region of the spectrum.

IV. CONCLUSIONS

We have investigated the spectral dependence of the efficiency of formation of F centers in diverse parts of the electromagnetic spectrum. For this purpose we have employed an extremely sensitive detection method which enables us to observe ultralow concentrations of F centers in alkali-halide crystals. This has allowed us to study a heretofore uninvestigated region of defect production, the early part of the so-called stage-I region, for concentrations of F centers as low as 10^{10} cm^{-3}. Using this method, we have measured the F-center production efficiency in the hard x-ray region at the K edge of bromine in KBr and discovered a definite increase at threshold. The existence and size of the increase at threshold is consistent with the core hole Auger cascade mechanism producing several electron-hole pairs which then recombine forming F centers by the well-known self-trapped exciton process. In the ultraviolet, maxima in the F-center production efficiency were observed in the long-wavelength tail of the first exciton peak and even in the region of the α band, providing the first direct measurements of the F-center production efficiency in this region.

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