

## Optical properties of $(F_2^+)_H$ and $F$ -aggregate centers in $\text{NaCl}:\text{OH}^-$ crystals

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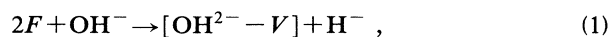
Data are presented on the optical absorption and emission of  $F$ -aggregate centers in  $\text{NaCl}:\text{OH}^-$ . In particular, we have studied the properties of the laser-active  $(F_2^+)_H$  centers. The formation of the  $(F_2^+)_H$  center is temperature dependent. One mechanism operates below and another above  $\sim 50$  K. The bleaching of the  $(F_2^+)_H$ -center emission observed under intense Nd:YAG or He-Ne laser irradiation, due to reorientation of the emitting centers, is accompanied by a process at low temperatures that is predominant at temperatures below 20 K and involves the ionization of the  $X$  or  $F_3$  centers always present in the crystal. Thus the laser-active  $(F_2^+)_H$  centers are destroyed by electron capture but are reformed by  $F$ -band light irradiation, which reverses the bleaching process.

### I. INTRODUCTION

In recent years color-center laser operation was discovered in additively colored NaCl-containing  $\text{OH}^-$  impurities.<sup>1</sup> In spite of the ease of pumping and of the excellent performance of these crystals as laser media (an excitation with a Nd:YAG laser, slope efficiency near 60%, wide-band tunability from 1.4 to 1.8  $\mu\text{m}$ , single-mode and room-temperature operations) (Refs. 2–5) some of the properties of the laser-active centers are not understood and have been the subject of several investigations.<sup>6–11</sup>

The accepted model of the center consists of a  $F_2^+$  center (two anion vacancies with a single trapped electron) associated with a  $\text{O}^{2-}$  impurity.<sup>2,6</sup> The center is usually labeled  $(F_2^+)_H$ , where the subscript  $H$  refers to the fact that the  $F_2^+$  center is close to an anionic defect on a nearby halogen-ion site.

The formation of the  $(F_2^+)_H$  centers in NaCl doped with  $\text{OH}^-$  includes the following steps: (a) additive coloration, that, besides the  $F$ -center production, is responsible for the following reaction:



where the symbol  $[\text{O}^{2-} - V]$  indicates an oxygen-vacancy defect pair, (b) room-temperature irradiation with  $F$ -band light, and (c) low-temperature (77 K) irradiation in the same spectral region.

Two different interpretations have been presented for the processes occurring in steps (b) and (c).

According to Ref. 6 (hypothesis I), step (b) produces

the  $F_2^+$  centers and step (c) allows the photoaggregation of the  $F_2^+$  center with the  $\text{O}^{2-}$  impurities producing the low-temperature stable  $(F_2^+)_H$ .

According to Refs. 2 and 10 (hypothesis II), the aggregation of  $F_2^+$  centers and oxygen impurities is completed during step (b) while step (c) involves only a reorientation of the *building blocks* of  $(F_2^+)_H$ , namely, the oxygen-vacancy defect pair  $[\text{O}^{2-} - V]$  and the  $F$  center. The reorientation at low temperatures may lead to the most stable configuration of the defects, perhaps the equilateral triangle configuration of  $\text{O}^{2-}$  and of the two anion vacancies in the  $\{111\}$  planes. A sketch of the model of the  $(F_2^+)_H$  center in this configuration is shown in Fig. 1.

Both models also describe the thermal destruction of  $(F_2^+)_H$  occurring during the warming up of the crystal in the dark above 230 K: in I the destruction of the centers is interpreted as a decomposition according to the following reaction:



while in II the disappearance of the  $(F_2^+)_H$  center is interpreted as a geometrical rearrangement of its constituents building blocks.

Both models have their shortcomings: The first interpretation is based on an unlikely motion of anion vacancies at low temperature. On the other hand, the second model assumes that the observed changes after irradiation at 77 K are configurational changes of the center, but its dichroic properties are always those of the  $\langle 110 \rangle$ -oriented  $F_2^+$  center, irrespective of the position of the  $\text{O}^{2-}$  ion.<sup>6</sup>

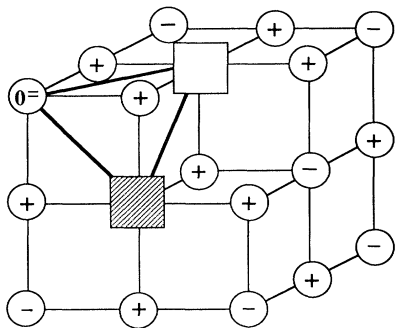


FIG. 1. Assumed configuration of ions and defects in the  $(F_2^+)_H$  center. The open square indicates the anion vacancy and the hatched square, the  $F$  center.

During the laser operation, the reorientation of the  $(F_2^+)_H$  center or its destruction are two possible mechanisms for the reduction of the laser output. It is important to know which is predominant in order to use a pumping light that will counteract this laser degradation.

In the present work we have investigated the optical properties of several aggregate centers that are formed during the various steps leading to the production of the  $(F_2^+)_H$  centers. Moreover, we have studied the kinetics of the  $(F_2^+)_H$  formation at different temperatures and the destruction and the recovery of this center during different types of optical excitation.

## II. EXPERIMENT

Three crystals from different sources were used in the present measurements. One crystal of NaCl doped with NaOH (500 ppm in the melt) was grown with the Kryo-

poulos method in a  $N_2$  atmosphere in the Rome-La Sapienza laboratory. A second crystal was purchased from the Crystal Growth Laboratory at the University of Utah (500 ppm doped), and a third crystal from the Crystal Growing Facility of Cornell University (200 ppm doped). The additive coloration was performed with the standard heat-pipe technique described in Ref. 12.

The measurements were performed in two laboratories (Pisa and Rome-Tor Vergata) and all relevant results were confirmed by measurements at both laboratories. All the measurements reported refer to absorption, excitation, and emission of samples attached to the cold finger of a variable temperature cryocooler. A schematic drawing of the experimental equipment used for most of the measurements is shown in Fig. 2. A slightly different apparatus with two monochromators of 25- and 10-cm focal length, respectively, was used to simultaneously measure the visible-near-IR and the IR emissions.

Irradiation with the  $F$  light was performed using a 100-W Hg high-pressure arc lamp whose light was filtered by a saturated solution of  $CuSO_4$  in water. The exciting light impinges on the sample perpendicular to the front surface. Brewster angle incidence light (such as used in the three-mirror folded laser cavity) was not used in this work. Excitation was performed with Nd:YAG (1.064  $\mu m$ ), a He-Ne (632.8 nm) laser, and monochromatized xenon or filtered Hg lamps. The emission spectra were recorded with a 25-cm focal length monochromator.

PbS, In-Ga-As, and InSb detectors or a photomultiplier with an S1 photocathode were used for monitoring the emission. The output of the detectors was normalized, at the different wavelengths with respect to the known emission of a blackbody source. Absorption measurements were performed by using a Perkin-Elmer 330 spectrophotometer.

Some of the absorption and emission spectra are

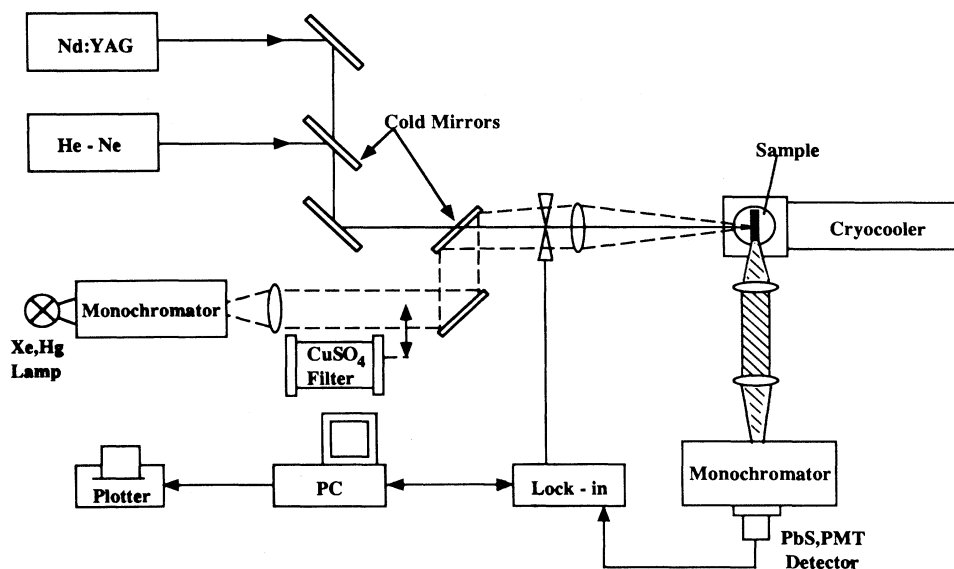


FIG. 2. Schematic diagram of the experimental apparatus.

decomposed into Gaussian and Lorentzian bands. The measured data were replotted to show intensity as a function of photon energy and, after the fitting, redrawn again to show the intensity versus wavelength.

### III. RESULTS

#### A. Optical properties of $F$ -aggregate centers

Figure 3 shows the absorption of a 2-mm thick sample of  $\text{NaCl}:\text{OH}^-$  during the various phases of the  $(F_2^+)_H$  center production. The spectra taken at room temperature (RT) after additive coloration and after a subsequent quenching from  $650^\circ\text{C}$  are shown in Fig. 3(a). The absorption spectra of the same crystal measured at liquid-nitrogen temperature (LNT) after RT irradiation with an  $F$  light, and after a further irradiation with the same spectral light at LNT, are shown in Fig. 3(b).

The main absorption bands observed at 450, 1040, and 1090 nm are due to the  $F$ ,  $F_2^+$  and  $(F_2^+)_H$  centers, respectively. The smaller absorptions at 600, 700 and 850 nm are related to  $F$ -aggregate centers and their attribution is still uncertain. The 600-nm band can be assigned to  $X$  centers, i.e., to large aggregates of  $F_3$  centers.<sup>13</sup> The  $X$ -center absorption band peaks around 600 nm, very close to the low-energy band of the  $F_3$  centers. The  $F_3^+$ - and  $F_2$ -band peaks occur at 700 and 715 nm, respectively,<sup>14</sup>

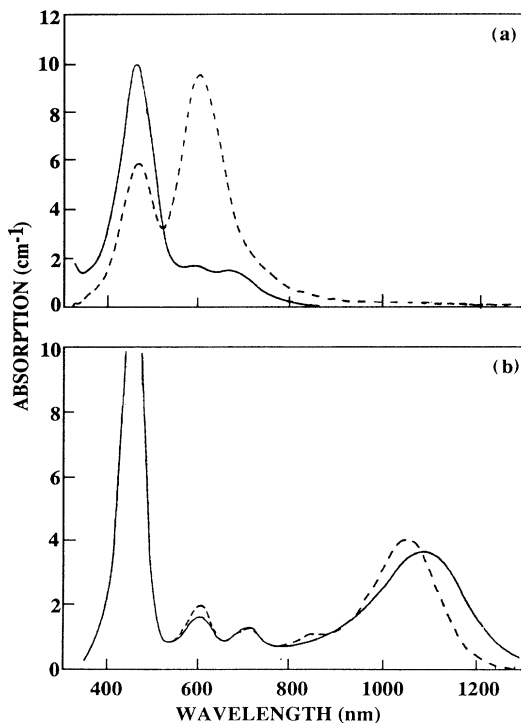


FIG. 3. Absorption spectra of a  $\text{NaCl}:\text{OH}^-$  crystal (a) at room temperature after additive coloration (dashed line) and subsequent quenching (solid line), (b) at liquid-nitrogen temperature after RT irradiation for 1 h with  $F$  light (dashed line), and after a further 5-min irradiation at LNT (solid line).

and so both centers can be responsible for the 700-nm absorption. We will return shortly to the optical properties of these centers. Eventually the 850-nm band is attributed to the  $F_4$  centers.

Often, after coloration, especially when the cooling time of the sample is not sufficiently rapid, colloids of metallic sodium develop and an absorption band appears around 550 nm.<sup>15,16</sup> Figure 4(a) shows the absorption spectrum of such samples decomposed in Gaussian curves for the  $F$  band at 450 nm and the  $X$  band at  $\sim 600$  nm and in a Lorentzian curve for the colloid band at 550 nm. The latter absorption disappears with some difficulty after repeated quenching [see Fig. 4(b)] while the  $F$  band increases and  $F_2$  and/or  $F_3^+$  appears. Keeping this in mind, we cannot completely exclude the presence of a small amount of colloids in the spectrum of Fig. 3(b).

A crystal with an absorption spectrum similar to that shown in Fig. 3(b) (solid line) has been cooled to liquid-helium temperature (LHeT) and its emission studied under various light excitations. As an example, we report in Fig. 5 the luminescence spectrum excited with the 457 nm line of an  $\text{Ar}^+$  laser. Beside the  $(F_2^+)_H$  emission band at 1560 nm, which will be dealt with in detail in the next section, a broad band appears at 1100 nm, most probably due to  $F$  and  $F_2$  centers, together with a small emission around 900 nm. The latter luminescence has been previously attributed to direct radiative deexcitation from an higher excited state of the  $(F_2^+)_H$  center.<sup>7</sup> This emission has been also observed at LNT with He-Ne laser excitation and, moreover, it seems to play an important role in the low-temperature kinetics of the  $(F_2^+)_H$  forma-

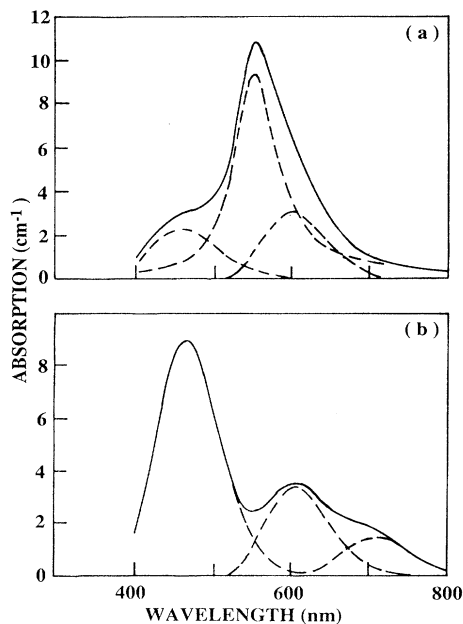


FIG. 4. Absorption spectra of a  $\text{NaCl}:\text{OH}^-$  crystal at RT after (a) additive coloration and (b) subsequent quenching. The spectral decomposition in (a) shows a band at  $\sim 550$  nm due to sodium colloids (see text for details).

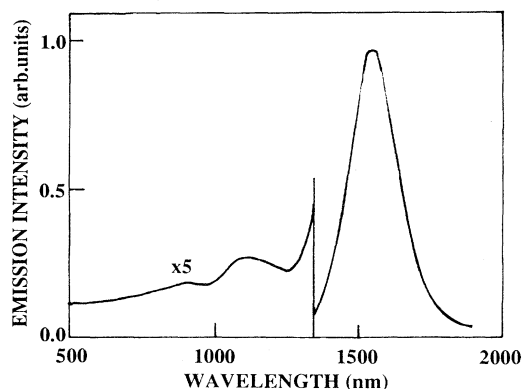


FIG. 5. Luminescence spectrum of a colored  $\text{NaCl:OH}^-$  crystal at liquid-helium temperature, excited with the 457-nm line of an Ar laser.

tion and destruction (see below).

In order to further clarify this point, we present in Figs. 6 and 7 the absorption and emission spectra taken at LNT before and after the RT irradiation. Figure 6 displays on expanded scale the absorption spectra between 500 and 800 nm. Before the RT aggregation with the  $F$  light, the absorption contains the  $X$  and  $F_3^+$  bands, after the aggregation one observes a slightly shifted  $X$  band and the disappearance of  $F_3^+$  substituted by a small  $F_2$  band. When the same experimental conditions are used, excitation with the He-Ne laser produces an emission band around 880 nm before irradiation and, after irradiation, an emission around 810 nm (see Fig. 7). Both emission bands are always present in this process (as shown in Fig. 7 by the deconvolution of the experimental curves) and changes of their intensity reflect the relative variation of the amount of the two centers. Therefore, we suppose that the 880-nm emission band is due to the  $F_3^+$  centers and the 810-nm band is related to the  $X$  absorption. These attributions are confirmed by the excitation

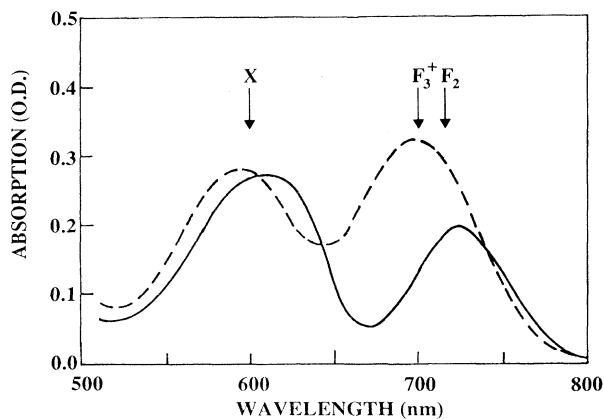


FIG. 6. Absorption spectrum of an additively colored  $\text{NaCl:OH}^-$  crystal at LNT before (dashed line) and after (solid line) irradiation with  $F$  light at room temperature.

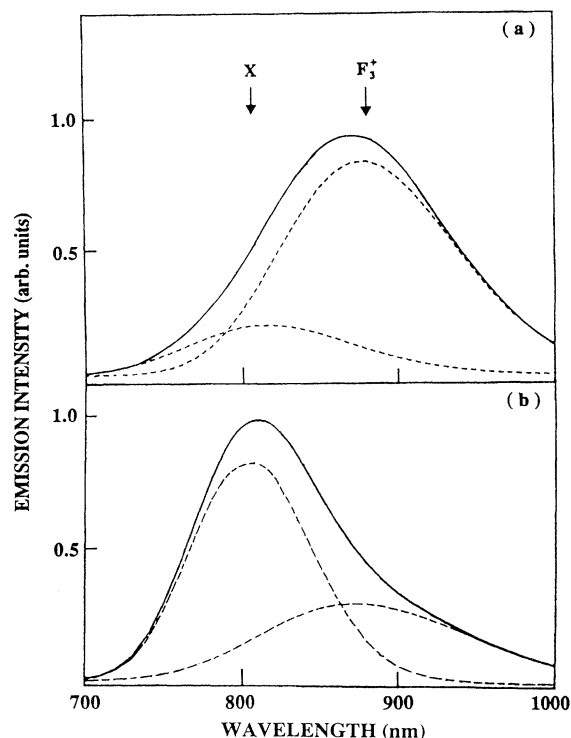


FIG. 7. LNT luminescence spectra of a colored  $\text{NaCl:OH}^-$  crystal excited by 632.8-nm He-Ne laser light, (a) before and (b) after irradiation with  $F$ -band light at room temperature. The dashed lines are a spectral decomposition into the two bands centered at 810 and 880 nm, respectively.

spectra of the two emissions and, moreover, the assignment of the 880-nm emission band to the  $F_3^+$  centers has already been reported.<sup>14</sup>

### B. Formation of the $(F_2^+)_H$ centers

The formation of the  $(F_2^+)_H$  centers has been studied by analyzing their emission band after the room-temperature  $F$ -light irradiation and by performing the low-temperature  $F$ -light irradiation at various temperatures from 10 to 120 K. The emission bands excited by a Nd:YAG laser at  $1.06 \mu\text{m}$  and observed (at different temperatures) after the RT aggregation are plotted on the left-hand side of Fig. 8. We have noticed that the peak position of the emission band and its halfwidth as measured after the room-temperature aggregation both depend on the sample and on the conversion light intensity. A computer fitting of the emission band reveals that two bands are present in OH-doped samples, with peaks at 1.48 and  $1.56 \mu\text{m}$  and halfwidths of 0.118 and 0.131 eV, respectively. The peak values almost coincide with those of the  $F_2^+$  and  $(F_2^+)_H$  centers. These data indicate that, after irradiation at room temperature, both the  $F_2^+$  and the  $(F_2^+)_H$  centers are already present in the sample. The latter centers (according to model I) can be those that statistically happen to be close to the  $\text{O}^{2-}$  impurity or, if the  $\text{O}^{2-}$  ions are already part of the center (model

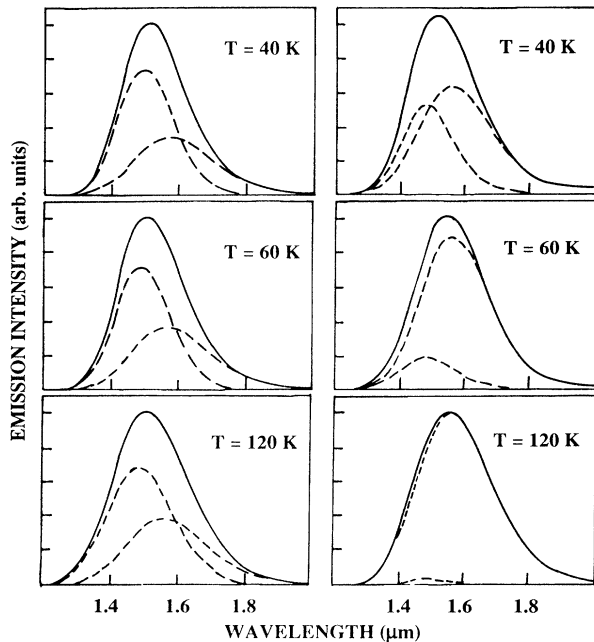


FIG. 8. Emission intensity excited by Nd:YAG laser light at three different temperatures after RT  $F$ -light irradiation (left-hand side) and after an additional 30-min  $F$ -light irradiation at the temperatures shown (right-hand side). Decomposition into the emissions from the  $F_2^+$  and  $(F_2^+)_H$  centers is shown by dashed lines.

II), some of the centers may already have the configuration showing the  $(F_2^+)_H$  emission.

The subsequent low-temperature irradiation always leads to an increase of the 1.56- $\mu\text{m}$  emission. The emission bands measured at this stage are plotted on the right-hand side of Fig. 8. After an identical  $F$ -light irradiation time ( $\sim 30$  min), the  $F_2^+ \rightarrow (F_2^+)_H$  is complete at 120 K but is still going on at 40 K.

In order to determine more quantitatively the  $(F_2^+)_H$  center formation, we measured, under simultaneous pumping with a Nd:YAG laser and  $F$  light, the emission intensity in the tail of the band at 1.8  $\mu\text{m}$ , where the  $F_2^+$  emission is nearly absent. The results are reported in Fig. 9 for various temperatures. From these curves, which do not follow a simple exponential behavior, it is possible to extract a time constant  $\tau$ , defined as the time in which the emission reaches one-half of the asymptotic value. The semilogarithmic plot of  $\tau$  has been reported in Fig. 10 as a function of  $1/T$ . The values of  $\tau$  are nearly constant below  $\sim 50$  K while at higher temperatures they show a linear behavior, which indicates the presence of a temperature-activated phenomenon. Indeed, these latter data can be fitted by the equation

$$\frac{1}{\tau} = A \exp \left[ \frac{-\Delta E}{k_B T} \right] \quad (3)$$

with an activation energy  $\Delta E$  of the order of 40 meV.

The low-temperature process, with negligible activa-

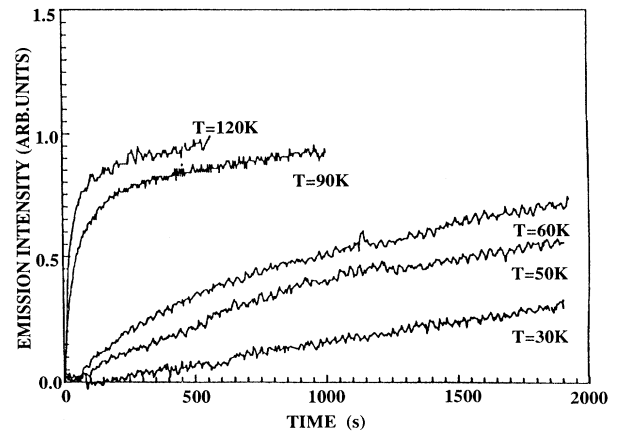


FIG. 9. Temperature dependence of the luminescence at 1.8  $\mu\text{m}$  from the  $(F_2^+)_H$  centers pumped by Nd:YAG laser light as a function of  $F$ -light irradiation time.

tion energy (if any), is much slower than the one at higher temperatures, but nevertheless the total number of the  $(F_2^+)_H$  centers formed at the end of the conversion is about the same in both cases. Since the low-temperature rate is temperature independent, we deduce that no migration or thermally activated rearrangement of vacancies or impurity ions occurs at this stage. After excitation, the  $F_2^+$  center has a small probability of tunneling among different lattice sites until it forms the  $(F_2^+)_H$  center.

From previous and preliminary measurements of the  $F_2^+ \rightarrow (F_2^+)_H$  conversion,<sup>17</sup> we estimated an activation energy below 50 K of  $\sim 5$  meV that led us to speculate about the possibility of a center migration (in agreement with hypothesis I) even at 10 K. The present and more accurate data seem to rule out this mechanism for the low-temperature conversion.

At higher temperatures ( $T > 50$  K), the conversion process displays an activation energy of  $\sim 40$  meV. A

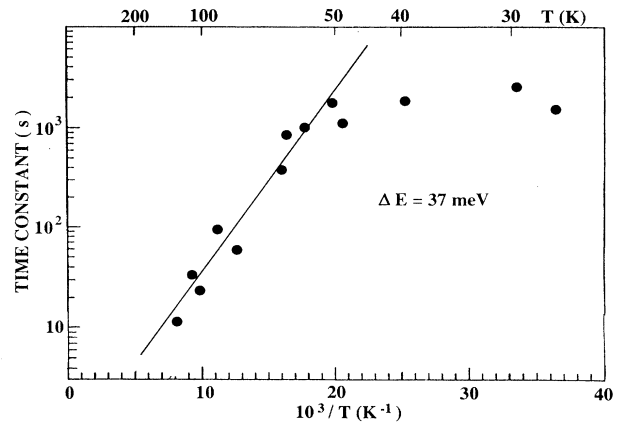


FIG. 10. Arrhenius plot showing time constant,  $\tau$ , for  $(F_2^+)_H$  production by  $F$ -light irradiation as a function of  $T^{-1}$ .

value of the same order of magnitude has been measured for the thermally activated reorientation of the  $F_2$  centers,<sup>18</sup> and we can suppose that a similar process also occurs here. Thus, the probability of  $(F_2^+)_H$ -center formation increases markedly.

With successive reorientations, e.g., by means of  $60^\circ$  jumps, the  $F_2^+$  centers may also migrate through the lattice. However, the total number of the  $(F_2^+)_H$  centers formed is nearly constant at all temperatures and then one is forced to suppose that the oxygen ion is always part of the center, since, otherwise, in the temperature region of increased mobility, the probability of forming an  $(F_2^+)_H$  center should increase.

### C. Bleaching and reorientation of $(F_2^+)_H$ centers

The performance of the  $(F_2^+)_H$ -center laser deteriorates in a few minutes if a restoring  $F$  light is not illuminating the crystal together with the Nd:YAG pumping light. The degradation of the laser has been explained by a reorientational process that decreases the number of active centers.<sup>6</sup> The excitation to a higher excited state by the  $F$ -band light is able to reorient the centers and to restore the lasing action.

The orientational bleaching should not be, however, the only reason for the laser fading because, with the geometry usually employed with these lasers (crystal at Brewster angle with two  $\langle 100 \rangle$  crystal axes in the polarization plane of the pumping beam), no  $\langle 110 \rangle$  direction is perpendicular to the pumping polarization and therefore no complete alignment of the centers along a nonlasing direction is possible.

The decrease of the emission under polarized Nd:YAG laser pumping is greatly enhanced if a He-Ne laser is simultaneously exciting the sample.<sup>6</sup> This fact has been explained as due to a two-step process: with the absorption of photons from the two lasers, the center is excited towards an upper excited state where the reorientation probability is higher. This mechanism is probably effective at temperatures around 77 K, but other processes are certainly present at lower temperatures where they predominate over the slow reorientation process. We performed several experiments to study the kinetics of the  $(F_2^+)_H$ -center formation and destruction, and we observe two distinct phenomena: a slow one due to the reorientation process and a fast one that we have ascribed to *electronic* processes.

We summarize these experiments in Fig. 11 where the emission intensity at  $1.56 \mu\text{m}$  [proportional to the number of the  $(F_2^+)_H$  centers] is sketched as a function of the time under different irradiation conditions. The time scale reflects the power of the light sources used, namely,  $\approx 1 \text{ W}$ ,  $\approx 20 \text{ mW}$ , and  $\approx 100 \text{ mW}$  for the Nd:YAG laser, He-Ne laser, and  $F$  light, respectively. At the beginning the  $(F_2^+)_H$  centers are almost equally distributed between the  $\langle 110 \rangle$  and  $\langle 1\bar{1}0 \rangle$  directions. Subsequently, the sample was irradiated with the Nd:YAG and He-Ne lasers both polarized along the  $\langle 110 \rangle$  direction. At 10 K the number of centers with axes parallel to the pumps polarizations decreases but no related increase of the number of centers with perpendicular  $\langle 1\bar{1}0 \rangle$  orientations is ob-

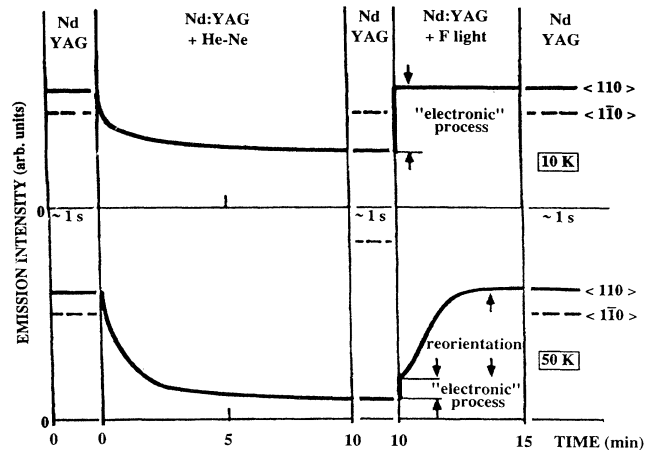


FIG. 11. Emission intensity at  $1.56 \mu\text{m}$  from the  $(F_2^+)_H$  centers in a  $\text{NaCl:OH}^-$  crystal undergoing pumping by polarized light (see text for details).

served. We are therefore observing the destruction of centers that are immediately reformed as soon as the  $F$  light is turned on. At 50 K, two processes occur during the bleaching. Part of the centers lost in the  $\langle 110 \rangle$  direction are found along the  $\langle 1\bar{1}0 \rangle$  direction (reorientation), part, as before, has disappeared (electronic processes). The unpolarized  $F$ -light irradiation immediately reforms the destroyed centers, but at a slower rate than the centers reorient themselves. At the end of the process, the initial situation is completely restored.

At the same time, the decrease at low temperatures (10 K) of the  $(F_2^+)_H$  emission is accompanied by an increase of the emission band around 880 nm. The  $F$ -band light that restores the  $(F_2^+)_H$ -center emission quenches the 880-nm band. The anticorrelated behavior of these emissions under switching the  $F$  light on and off is shown in Fig. 12.

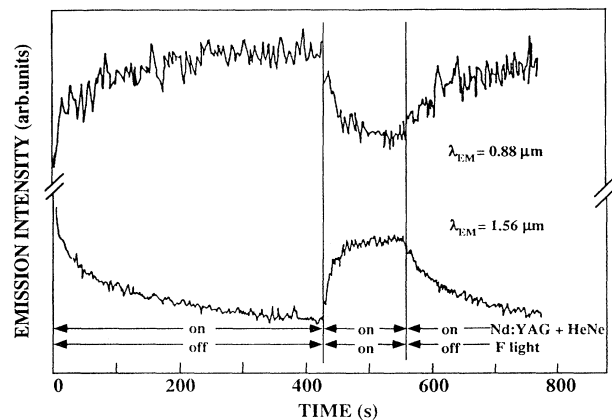


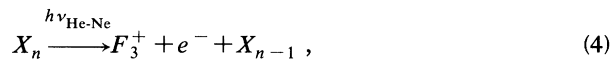
FIG. 12. Emission intensities at 10 K from the  $(F_2^+)_H$  center ( $\lambda=1.56 \mu\text{m}$ ) and from the  $F_3^+$  center ( $\lambda=0.88 \mu\text{m}$ ) under Nd:YAG and He-Ne laser excitation as the  $F$  light is switched on and off. The intensity variation is  $\sim 50\%$  of the maximum intensity.

#### IV. DISCUSSION

The experimental results described in the previous section can be explained by the following mechanisms. After coloration and quenching, the crystal contains some  $F_3^+$  centers, together with large  $F$ -center aggregates. This is clearly evidenced by the absorption bands at 700 nm ( $F_3^+$ ) and 600 nm ( $X$ ), as shown by the dashed line in Fig. 6. After the aggregation, only the  $F_3^+$  centers are destroyed with the formation of the  $F_2$  and  $F_2^+$  centers (Fig. 6, solid line, and Ref. 6).

The He-Ne laser irradiation ionizes some of the  $X$  centers and the released free electrons can be captured by the  $(F_2^+)_H$  centers, thus destroying the laser-active centers. Formation of  $F_3^+$  centers due to ionization of  $X$  centers has been observed in NaCl:OH<sup>-</sup> crystals.<sup>13</sup> This fact supports the hypothesis that the  $X$  centers (more exactly  $X_n$  centers) are weakly bound aggregates of the  $F_3$  centers. The presence of the  $F_3^+$  centers in these conversion processes is confirmed by the observation of the  $F_3^+$ -center luminescence around 880 nm.

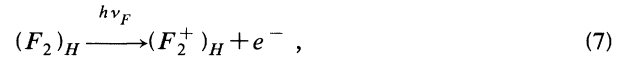
The following reactions are assumed:



As a consequence of the above reactions, the He-Ne and Nd:YAG lights produce a decrease of the 1.56- $\mu\text{m}$  ( $(F_2^+)_H$ ) emission and a simultaneous increase of the  $F_3^+$  emission excited by the He-Ne laser light.

Only the simultaneous presence of light from the two lasers induces this process. Thus, we assume that electron capture by the  $(F_2^+)_H$  centers can occur only in the  $(F_2^+)_H^*$  (excited state). Indeed, the center as a whole is neutral, but in the excited state the attractive potential of the negative-ion vacancy might not be completely shielded by the electron trapped on the center.

The switching of  $F$ -band light on reverses reactions (4)–(6). The  $(F_2)_H$  centers are ionized and the  $F_3^+$  are neutralized to  $F_3$  by electron capture according to the following reactions:



The previous kinetic reactions give a plausible explanation of the results of Fig. 12, where the different time constants displayed by the anticorrelated emissions are a consequence of different  $F$ -light and He-Ne laser light pumping powers.

#### V. CONCLUSIONS

We have shown that the formation of the laser-active  $(F_2^+)_H$  centers in NaCl:OH<sup>-</sup> under  $F$ -light irradiation above 50 K is a thermally activated process, with an activation energy of about 40 meV. At lower temperatures, the formation rate is lower and temperature independent. Our measurements seem to indicate that the oxygen impurity is close to the  $F_2^+$  center after the RT irradiation. The experimental results also indicate the existence of additional complex processes involving the various color centers in these crystals.

Indeed, we have obtained evidence indicating that the large  $F$ -center aggregates ( $X$  centers) play an important role in the formation and in the bleaching of the  $(F_2^+)_H$  centers. In addition to demonstrating that the bleaching depends on the orientation of the centers, we have demonstrated an optically stimulated conversion between the  $(F_2)_H$  and  $(F_2^+)_H$  centers that is accompanied by a related transformation between the  $F_3^+$  and  $F_3$  centers. The presence of the  $F_3$  centers, in the neutral or in the ionized state, is important for both the bleaching and recovery of the laser-active  $(F_2^+)_H$  centers in NaCl:OH<sup>-</sup>.

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