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Dispersion of the photoanisotropy induced in an F_A(II) KCl:Li crystal

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The dichroism and birefringence photoinduced in an F_A(II) KCl:Li crystal by a linearly polarized light beam are theoretically analyzed, taking into account the off-axis position of the Li^+ ion inside the crystal. Measurements are taken between 400 and 700 nm, the crystal being at liquid-nitrogen temperature. The peculiar shapes of the resulting curves are discussed. The value of the angular shift θ of the Li^+ ion is deduced from the curves corresponding to the saturation, taking into account the wavelength of the exciting light. We found $\theta = 17° \pm 1°$ for a crystal at liquid-nitrogen temperature.

I. INTRODUCTION

Optical absorption of F centers (an electron trapped by an anion vacancy) in alkali halides arises from allowed transition of the trapped electron from a single 1s-like state to three possible 2p-like states, each one aligned along one of the three mutually orthogonal $\langle 100 \rangle$ directions of the crystal.[1] These excited states correspond to the same energy due to the cubic O_h symmetry of the center and the resulting absorption band, denoted an F band, is threefold degenerate. Small additional bands, denoted K, L_1, L_2, L_3, have been observed on the short-wavelength side of the F band. They correspond to transitions into higher excited bound (K) and conductive (L) states.

The F_A center consists of an F center perturbed by a nearest-neighbor substitutional cation impurity.[2] The local symmetry is reduced to C_{4v}. The F band is thus split into two separate bands, respectively, denoted F_{A1} and F_{A2}. The F_{A1} band characterizes the dipole transition oriented along the direction defined by the center of the vacancy and the foreign cation. The F_{A2} band, twofold degenerate, corresponds to the two transitions mutually perpendicular and also perpendicular to the first one. It is slightly shifted with respect to the undisturbed F band. The F_{A1} and F_{A2} bands are also accompanied by a series of weak absorption bands, K, L_1, L_2, and L_3, corresponding to higher energies. Lüty[2] noted an increase of K and L absorptions of F_A centers with respect to the corresponding absorptions of F centers and a slightly dichroic behavior. If K and L bands are neglected, the F_{A1} and F_{A2} bands have the same integrated absorption as the F band from which they are derived. Moreover, the F_{A1} transition has the same oscillator strength as one of the twofold-degenerate F_{A2} transitions.

Under an optical excitation with linearly polarized light belonging to the F_{A1} (F_{A2}) band, the F_A centers whose axis is parallel (perpendicular) to the light vector reorient into one of the two mutually perpendicular directions. This reorientation is done by a jump into the vacancy of one of the four anions which are both nearest neighbors of the foreign cation and next-nearest neighbors of the vacancy.

The object of the present work is first to study the dispersion of dichroism and birefringence photoinduced in F_{A1} and F_{A2} bands of an F_A-center lithium-doped KCl crystal at liquid-nitrogen temperature and, secondly, to measure the angular shift of the lithium ion inside the crystalline lattice. Section II is devoted to a brief recall of the peculiar structure of such a center. In Sec. III, we calculate the photoinduced dichroism and birefringence taking into account the wavelength of the exciting light and the amount of exposure given to the crystal. The experimental results are presented in Sec. IV and discussed in Sec. V.

II. STRUCTURE OF F_A(II) CENTERS IN KCl:Li CRYSTALS

It has been demonstrated (Refs. 3–5) that substitutional Li^+ ions in KCl occupy $\langle 111 \rangle$ off-center positions. In the case of Li^+ ions associated with F centers (F_A centers), Mieher[6] showed by ENDOR spectroscopy that the distance from the Li nucleus to the vacancy center is 3.65 Å while the distance between ions in a perfect KCl lattice is 3.14 Å. From Raman[7] and electro-optical measurements,[8] it was concluded that Li^+ ions in the KCl:F_A centers may occupy four equivalent sites lying on the $\{001\}$ planes and slightly shifted parallel to the $\langle 110 \rangle$ directions, performing a tunneling motion among them. Recently the value of this angular tilt θ has been determined by emission[9] and absorption[10] measurements.

Let us consider an F_A center whose axis is parallel to the [100] direction (Fig. 1). There are four equivalent directions for the F_{A1} transition, respectively, parallel to the lines joining the center of the vacancy and the four equivalent positions of the lithium ion. The local symmetry of the F_A center is reduced to C_S. If the direction cosines of the F_{A1} transition are given by $(\cos\theta, \sin\theta/\sqrt{2}, \sin\theta/\sqrt{2})$, the group theory assigns to one of the F_{A2} transitions to be parallel to the direction

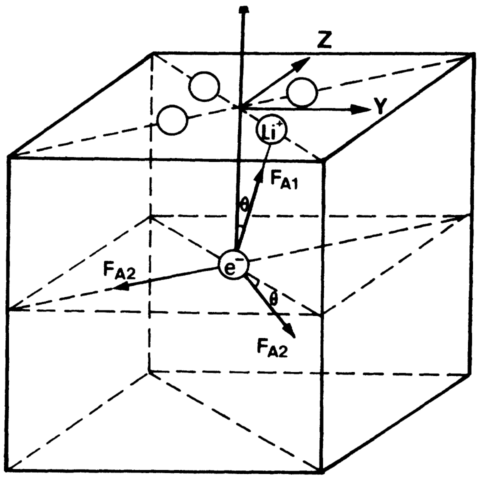

FIG. 1. Configuration of the F_A (Li) center.

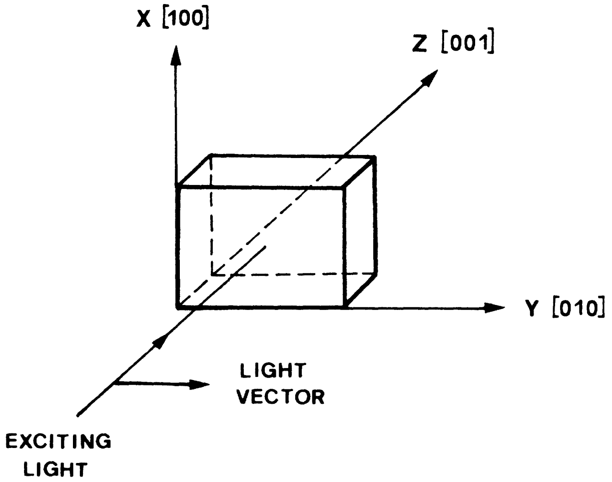

FIG. 2. Optical excitation of the crystal. The incident light is linearly polarized parallel to the [010] direction and propagates along the [001] direction. ($T = 78$ K.)

$(0,1/\sqrt{2},-1/\sqrt{2})$. The other F_{A2} transition is taken perpendicular to each of the previous ones and is thus characterized by the direction $(-\sin\theta,\cos\theta/\sqrt{2},\cos\theta/\sqrt{2})$. After Lüty,[11] the two F_{A2} transitions correspond to very slightly different $2p$-like energy levels. However, the energy difference between these two levels is much weaker than the half-width of the F_{A2} band and will be neglected in the following.

III. DETERMINATION OF THE ANISOTROPY PHOTOINDUCED BY A LINEARLY POLARIZED LIGHT VIBRATION

Initially, the crystal is isotropic and contains a random distribution of F_A(II) centers, characterized by the uniform concentration $c/3$ along each of the three $\langle 100\rangle$ directions. A reorientation process is induced by a monochromatic optical excitation propagating along the z direction [001] and linearly polarized along the y axis [010] (Fig. 2). We shall consider the general case when the wavelength of the exciting light belongs to both F_{A1} and F_{A2} bands.

A. Spatial distribution of F_A centers after excitation

The incident light brings about an optical excitation of the centers characterized by F_{A1} and F_{A2} transitions having a nonzero component along the y axis.

Let $\hat{\mathbf{y}}$ be the unit vector characterizing the light vibration incident upon the crystal. In the low-level intensity approximation, the number of photons absorbed per unit time in the F_{A1} band by a center parallel to the i axis ($i = x,y,z$) may be written as

$$N_i^{A1} = \eta_1 I(\hat{\mathbf{y}}\cdot\hat{\mathbf{r}}_{1i})^2 , \tag{1}$$

where η_1 is a constant relative to the F_{A1} band depending on the wavelength of the incident light, I is the incident light intensity, and $\hat{\mathbf{r}}_{1i}$ is the unit vector showing the direction of the transition dipole moment of the F_{A1} transition relative to the center under consideration. In the same way, the number of photons absorbed per unit time in the F_{A2} band is given by

$$N_i^{A2} = \eta_2 I[(\hat{\mathbf{y}}\cdot\hat{\mathbf{r}}_{2i})^2 + (\hat{\mathbf{y}}\cdot\hat{\mathbf{r}}'_{2i})^2] , \tag{2}$$

where η_2 is another constant, relative to the F_{A2} band and depending on the wavelength of the incident light, and $\hat{\mathbf{r}}_{2i}$ and $\hat{\mathbf{r}}'_{2i}$ are the unit vectors showing the respective directions of the F_{A2} transitions characterizing the center. The number of centers parallel to the i direction which are excited by the incident light between the times t and $t+dt$ is thus given by

$$dc_i = (N_i^{A1} + N_i^{A2})c_i dt . \tag{3}$$

Each of the excited centers has a probability P to be reoriented in one of the two perpendicular directions. Under these conditions, the time evolution of the concentrations of the centers along the crystallographic axes is described by the equations

$$\begin{aligned}
\frac{dc_x}{dt} &= -\frac{P}{2}\eta_1 I c_x \sin^2\theta + \frac{P}{2}\eta_1 I c_y \cos^2\theta + \frac{P}{4}\eta_1 c_z \sin^2\theta \\
&\quad - \frac{P}{2}\eta_2 I c_x(1+\cos^2\theta) + \frac{P}{2}\eta_2 I c_y \sin^2\theta \\
&\quad + \frac{P}{4}\eta_2 I c_z(1+\cos^2\theta) , \\
\frac{dc_y}{dt} &= -P\eta_1 I c_y \cos^2\theta + \frac{P}{4}\eta_1 I(c_x+c_z)\sin^2\theta \\
&\quad - P\eta_2 I c_y \sin^2\theta \\
&\quad + \frac{P}{4}\eta_2 I(c_x+c_z)(1+\cos^2\theta) , \\
\frac{dc_z}{dt} &= -\frac{P}{2}\eta_1 I c_z \sin^2\theta + \frac{P}{4}\eta_1 I c_x \sin^2\theta + \frac{P}{2}\eta_1 I c_y \cos^2\theta \\
&\quad - \frac{P}{2}\eta_2 I c_z(1+\cos^2\theta) + \frac{P}{4}\eta_2 I c_x(1+\cos^2\theta) \\
&\quad + \frac{P}{2}\eta_2 I c_y \sin^2\theta .
\end{aligned} \tag{4}$$

In the case of F_A(II) KCl:Li centers, P is equal to 0.5 and is temperature independent.[2] After integration, we obtain

$$c_x(t)=c_z(t)=\frac{c}{6}[3-\gamma_1-(1-\gamma_1)e^{-t/\tau}] \tag{5a}$$

$$=\frac{c}{3}[\gamma_2+(1-\gamma_2)e^{-t/\tau}] , \tag{5b}$$

$$c_y(t)=\frac{c}{3}[\gamma_1+(1-\gamma_1)e^{-t/\tau}] , \tag{6a}$$

$$=\frac{c}{3}[3-2\gamma_2-2(1-\gamma_2)e^{-t/\tau}] , \tag{6b}$$

with

$$\gamma_1=3\frac{\eta_1\sin^2\theta+\eta_2(2-\sin^2\theta)}{\eta_1(4-3\sin^2\theta)+\eta_2(2+3\sin^2\theta)} , \tag{7}$$

$$\gamma_2=\frac{6(\eta_1\cos^2\theta+\eta_2\sin^2\theta)}{\eta_1(4-3\sin^2\theta)+\eta_2(2+3\sin^2\theta)} , \tag{8}$$

and

$$\tau^{-1}=\frac{P}{4}I[\eta_1(4-3\sin^2\theta)+\eta_2(2+3\sin^2\theta)] . \tag{9}$$

Moreover, γ_1 and γ_2 are related by

$$\gamma_1+2\gamma_2=3 . \tag{10}$$

Even if the incident light belongs to one single band (F_{A1} or F_{A2}), the reorientation is limited. In fact, for an excitation in the F_{A1} band ($\eta_2=0$), we have

$$\lim_{t\to\infty} c_y(t)=\gamma_1 c/3 , \tag{11}$$

and for an excitation in the F_{A2} band ($\eta_1=0$), we have

$$\lim_{t\to\infty} c_x(t)=\lim_{t\to\infty} c_z(t)=\gamma_2 c/3 . \tag{12}$$

B. Complex indices of the anisotropic crystal

After such an exposure, the crystal is uniaxial. The induced optic axis is taken parallel to the light vibration which induced the anisotropy. The number, per unit volume, of F_{A1} transitions associated with one of the $\langle 100\rangle$ directions, may be written as

$$C_i^{A1}=c_i(\hat{\mathbf{i}}\cdot\hat{\mathbf{r}}_{1i})^2+c_j(\hat{\mathbf{i}}\cdot\hat{\mathbf{r}}_{1j})^2+c_k(\hat{\mathbf{i}}\cdot\hat{\mathbf{r}}_{1k})^2 , \tag{13}$$

where $(i,j,k)=(x,y,z)$ and $\hat{\mathbf{i}}$ is the unit vector showing the direction of the crystallographic axis under consideration. The corresponding number C_i^{A2} of F_{A2} transitions is given by

$$C_i^{A2}=c_i[(\hat{\mathbf{i}}\cdot\hat{\mathbf{r}}_{2i})^2+(\hat{\mathbf{i}}\cdot\hat{\mathbf{r}}'_{2i})^2]+c_j[(\hat{\mathbf{i}}\cdot\hat{\mathbf{r}}_{2j})^2+(\hat{\mathbf{i}}\cdot\hat{\mathbf{r}}'_{2j})^2]$$
$$+c_k[(\hat{\mathbf{i}}\cdot\hat{\mathbf{r}}_{2k})^2+(\hat{\mathbf{i}}\cdot\hat{\mathbf{r}}'_{2k})^2] . \tag{14}$$

The complex index relative to the i crystallographic axis and corresponding to a wavelength λ is given by

$$n_i=n_c+\frac{\alpha_{A1}}{2n_c}C_i^{A1}+\frac{\alpha_{A2}}{2n_c}C_i^{A2} , \tag{15}$$

where n_c is the real refractive index of the KCl:Li crystal before the coloration and α_{A1} and α_{A2} are the complex macroscopic polarizabilities associated with the F_{A1} and F_{A2} transitions. At time $t=0$, $c_x=c_y=c_z=c/3$. It may be derived from Eqs. (13) and (14) that

$$C_i^{A1}(0)=\frac{c}{3} \quad\text{and}\quad C_i^{A2}(0)=\frac{2c}{3} , \tag{16}$$

whatever $i=(x,y,z)$ is. Consequently, the optical absorptions of the isotropic crystal of thickness d, related to each F_{Aj} band ($j=1,2$), are, respectively, given by

$$K_0^{Aj}=jkd\log_{10}e\frac{c}{3n_c}\mathrm{Im}(\alpha_{Aj}) , \tag{17}$$

where $k=2\pi/\lambda$.

The corresponding refractive index variations induced in each band by the coloration may be written as

$$\Delta n_0^{Aj}=j\frac{c}{6n_c}\mathrm{Re}(\alpha_{Aj}) . \tag{18}$$

The photoinduced dichroism D and birefringence Φ are, respectively, defined by

$$D=2kd\,\mathrm{Im}(n_x-n_y)\log_{10}e \tag{19a}$$

and

$$\Phi=kd\,\mathrm{Re}(n_x-n_y) . \tag{19b}$$

After calculations, D and Φ are finally given by

$$D=(-1)^p\tfrac{3}{4}p(K_0^{A2}-2K_0^{A1})(1-\Gamma_p)(1-e^{-t/\tau}) , \tag{20}$$

$$\Phi=(-1)^p\tfrac{3}{4}pkd(\Delta n_0^{A2}-2\Delta n_0^{A1})(1-\Gamma_p)(1-e^{-t/\tau}) , \tag{21}$$

where $p=(1,2)$ and

$$\Gamma_p=\tfrac{1}{2}[3\sin^2\theta+\gamma_p(2-3\sin^2\theta)] . \tag{22}$$

C. Phenomena at saturation: measurement of angle θ

At saturation, the dichroisms induced in each band ($j=1,2$) and the birefringence are, respectively, given by

$$D_{ps}^{Aj}(\lambda)=(-1)^{p+j}\frac{3}{2}\frac{p}{j}(1-\Gamma_p)K_0^{Aj}(\lambda) , \tag{23}$$

$$\Phi_{ps}(\lambda)=(-1)^p\tfrac{3}{4}pkd(1-\Gamma_p)[\Delta n_0^{A2}(\lambda)-2\Delta n_0^{A1}(\lambda)] . \tag{24}$$

Consequently, whatever are the reading wavelengths λ, λ', and λ'', we have, considering an exciting wavelength of the F_{A1} band ($p=1$, $\eta_2=0$) on the one hand, and an exciting wavelength of the F_{A2} band ($p=2$, $\eta_1=0$) on the other hand,

$$\frac{D_{2s}^{A1}(\lambda)}{D_{1s}^{A1}(\lambda)}=\frac{D_{2s}^{A2}(\lambda')}{D_{1s}^{A2}(\lambda')}=\frac{\Phi_{2s}(\lambda'')}{\Phi_{1s}(\lambda'')}$$
$$=\frac{2(1-\Gamma_2)}{1-\Gamma_1}$$
$$=R=\frac{4-3\sin^2\theta}{2+3\sin^2\theta} . \tag{25}$$

The angle θ is thus given by

$$\theta=\sin^{-1}\left[\left[\frac{2}{3}\frac{2-R}{R+1}\right]^{1/2}\right] . \qquad (26)$$

IV. MEASUREMENT OF PHOTOINDUCED ANISOTROPY

A. Manufacture of the crystal

The crystal was grown by the Bridgman-Stockbarger method[12] from a melt of KCl and LiCl (1 mol. %) in a silica tube. Color centers (F centers) were generated by electrolysis between a pointed cathode and a flat anode at high temperature (640 °C) with an electric field of 40 V cm^{-1}. At this temperature, the coloring is completed in about 30 min. Then, the crystal was properly quenched at room temperature. Optical transformation of the F centers into F_A centers was performed at room temperature with a diffuse white-light beam.

B. Excitation of the crystal

An isotropic plane-parallel plate, at liquid-nitrogen temperature, is illuminated in normal incidence by a parallel beam of monochromatic light, propagating along the z axis and linearly polarized parallel to the y axis (Fig. 2). To induce the anisotropy, we used four laser wavelengths: 496.5, 514.5, 568.2, and 632.8 nm. In each case, we illuminated the crystal with successive exposures until saturation.

C. Measurement of dichroism and birefringence

To achieve these measurements, we used a well-known technique. The plane-parallel sample, at liquid-nitrogen temperature, is mounted between two polarizers P and A (Fig. 3) which are, respectively, inclined one at β and the other at $\pi/4$ on the y axis. They are illuminated by a parallel beam of white light derived from a xenon lamp, type XBO. The lens L_2 focuses the beam emerging from the analyzer on to the entrance slit of a Czerny-Turner spectroscope. A multichannel analyzer, constituted of 1024 photodiodes, measures the irradiance $I(\lambda,\beta)$ of the spectrum lying in the exit plane of the spectroscope, as a function of the wavelength, between 400 and 700 nm. The values of $D(\lambda)$ and $\Phi(\lambda)$ are derived from $I(\lambda,\beta)$ for various values of the angle β.

D. Experimental results

The crystal is 2 mm thick and contains about 3×10^{16} F_A centers/cm^3. It is cut along the crystallographic axes and optically polished. The curve of Fig. 4 represents the optical absorption of this crystal between 400 and 700 nm. This experimental curve is generated by four Gaussian absorption bands L_1, K, F_{A2}, and F_{A1} numerically computed and, respectively, centered at 350, 490, 550, and 628 nm.

The curves of dichroism corresponding to successive exposure times for each exciting wavelength are all of characteristic shape. We show here the curves obtained with the 568.2-nm [Fig. 5(a)] and 632.8-nm [Fig. 6(a)] excitations. There is always a zero of dichroism and two extrema for three wavelengths independent of the exciting wavelength and exposure times. The extrema are centered at the maxima $\lambda_2=550$ nm and $\lambda_1=628$ nm of the absorption bands F_{A2} and F_{A1}. The cancellation of dichroism is for $\lambda_D=590$ nm.

Concerning the birefringence, the corresponding curves are represented in Figs. 5(b) and 6(b). Their respective shapes are similar for the various exciting wavelengths and the different exposures. The birefringence of the crystal passes twice through zero for values λ_{B2} and λ_{B1} of the wavelength, respectively, situated in the neighborhood of the peaks of dichroism. Φ is maximum for a wavelength λ_{BM} very close to that which cancels the dichroism. λ_{BM} is slightly varying with the exposure time and suffers a shift in the region of short wavelengths when it is increasing.

To obtain θ, we measured, for each exciting wavelength, the saturated dichroisms D_s^{A1}, D_s^{A2} and the birefringence Φ_s at the wavelengths corresponding to

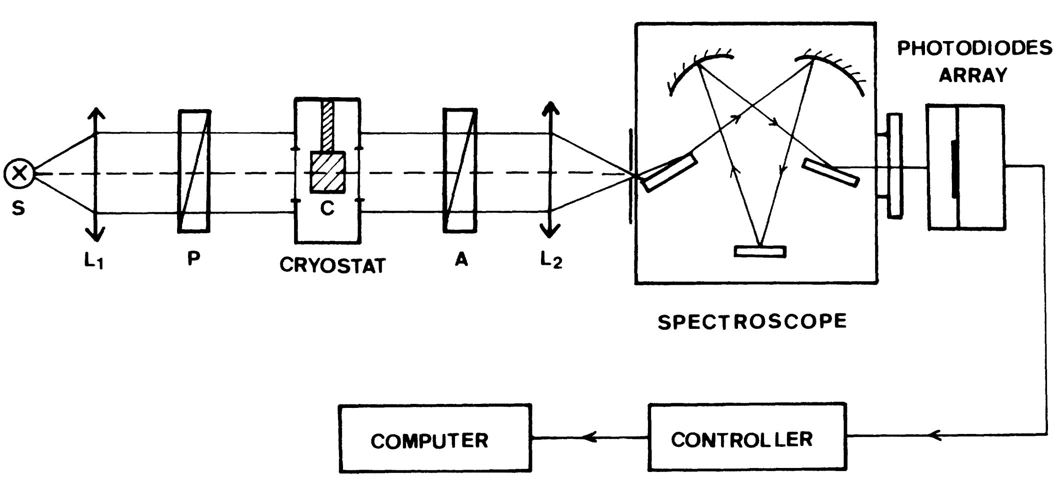

FIG. 3. Measurement of photoinduced anisotropy. The analyzer A is inclined at $\pi/4$ on the optic axis of C. The polarizer P is rotating around the axis of the setup. ($T=78$ K.)

their respective maxima. Among the exciting wavelengths, only one (632.8 nm) belongs to the F_{A1} band. We denote by $D_{2s}^{Aj}(\lambda_j)$ $(j=1,2)$ and $\Phi_{2s}(\lambda_{\mathrm{BM}})$ the photoinduced anisotropy resulting from the excitation by the three other ones. The experimental results are represented on Table I for the three exciting wavelengths of F_{A2} band.

V. DISCUSSION

A. Dichroism

As shown by Eqs. (20), the variations of the photoinduced dichroism versus the wavelength are only dependent on the term $K_0^{A2}-2K_0^{A1}$. This term has been computed from the two decomposed Gaussian curves. The resulting curve, represented in Fig. 7, shows the same characteristic wavelengths λ_1, λ_2, and λ_D as the experimental curves. However, the last ones exhibit a very weak dichroism in the K band, which has been neglected in Sec. III.

It may be easily deduced from Eq. (20) that whatever the exciting wavelength and the exposure times are, we have

$$\frac{D^{A1}(\lambda,t)}{D^{A2}(\lambda',t)}=-2\frac{K_0^{A1}(\lambda)}{K_0^{A2}(\lambda')} . \tag{27}$$

Assuming that the centers are damped and noninteracting oscillators, it may be demonstrated from the Smakula relation[13] relative to a Gaussian absorption band that

$$\frac{D^{A1}(\lambda_1,t)}{D^{A2}(\lambda_2,t)}=-\frac{H_2}{H_1} , \tag{28}$$

where H_1 and H_2 are the full widths at half maximum of the F_{A1} and F_{A2} bands, respectively. This ratio is temperature independent. The experimental results obtained at liquid-helium temperature by Fritz, Lüty, and Rausch[14] give

$$H_2/H_1=1.58 . \tag{29}$$

From the curves of Fig. 4, we have

$$2\frac{K_0^{A1}(\lambda_1)}{K_0^{A2}(\lambda_2)}=1.55 \text{ and } \frac{H_2}{H_1}=1.7 . \tag{30}$$

Results show that the crystal still contains F centers. Table II shows the values of $-D^{A1}(\lambda_1)/D^{A2}(\lambda_2)$ calculated for each exciting wavelength, by averaging the experimental data corresponding to all of the exposures. The uncertainty on the value of $-D^{A1}(\lambda_1)/D^{A2}(\lambda_2)$ is

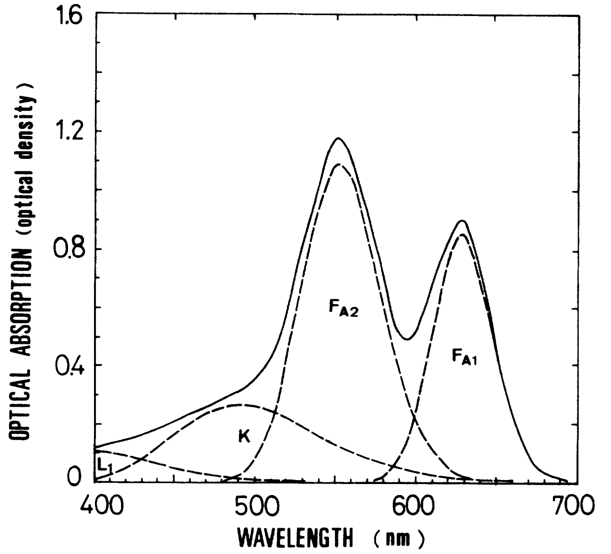

FIG. 4. Solid curve shows the optical absorption of the F_A(II) KCl:Li crystal before irradiation. The numerical decomposition gives rise to four Gaussian curves in dashed line. ($T=78$ K.)

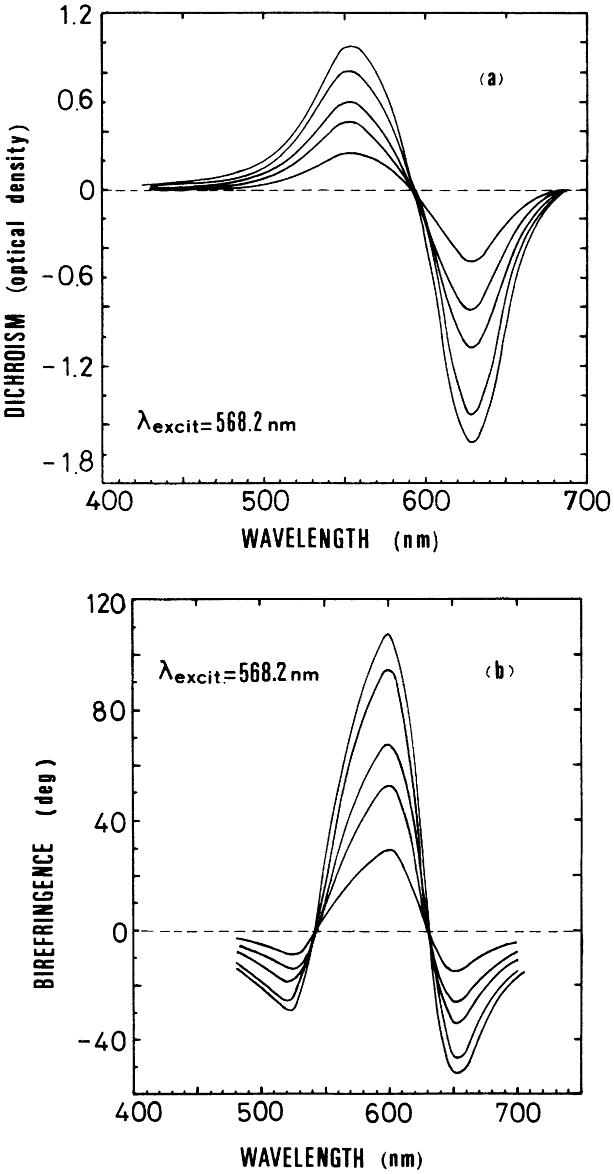

FIG. 5. Anisotropy photoinduced by the 568.2-nm radiation and corresponding to increasing exposures: (a) dichroism, (b) birefringence.

smaller as the exciting wavelength is far from the maximum of the K band.

B. Birefringence

In the same way as the dichroism, the dispersion of the photoinduced birefringence is only a function of the term $k(\Delta n_0^{A2}-2\Delta n_0^{A1})$. The refractive index variations, induced in each absorption band (F_{A1} and F_{A2}) by the coloration of the KCl:Li crystal, are induced from the corresponding optical absorptions via the Kramers-Kronig relations. The calculations have been done by Dexter[15] and

$$\Delta n_0^{Ai}=-cf_i\frac{\hbar^2e^2}{m\epsilon_0}\frac{(n_c^2+2)^2}{9n_c}\times\frac{\exp[-(E-E_i)^2/W_i^2]}{W_i(E+E_i)}\int_0^{(E-E_i)/W_i}e^{t^2}dt\ , \tag{31}$$

where $i=(1,2)$, f_i is the oscillator strength of the transition, $\hbar$, e, m, and ϵ_0 have their usual meanings, E is the photon energy, and

$$W_i=\frac{H_i}{2\sqrt{\ln 2}}\ . \tag{32}$$

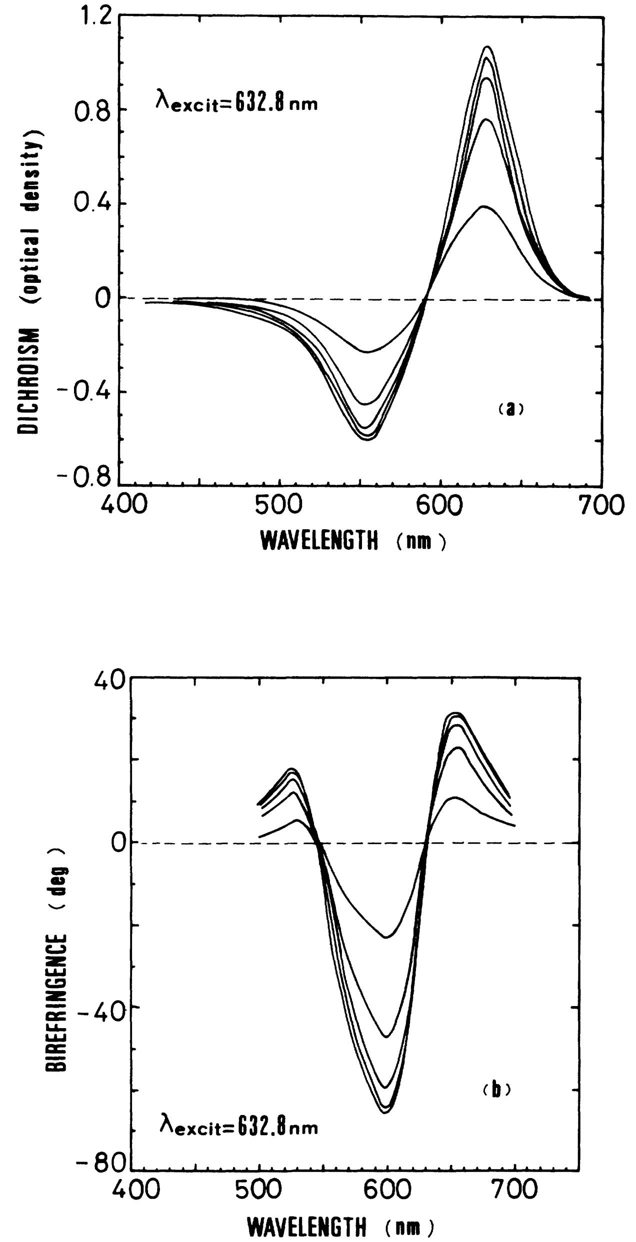

FIG. 6. Anisotropy photoinduced by the 632.8-nm radiation and corresponding to increasing exposures: (a) dichroism, (b) birefringence.

TABLE I. Values of angle θ deduced from the ratios of the maximum anisotropies (dichroism and birefringence) induced by a radiation of the F_{A2} band and the wavelength 632.8 nm of the F_{A1} band.

	$\lambda_{\text{excit}}=$ 496.5 nm	$\lambda_{\text{excit}}=$ 514.5 nm	$\lambda_{\text{excit}}=$ 568.2 nm
$\dfrac{D_{2s}^{A1}(\lambda_1)}{D_{1s}^{A1}(\lambda_1)}$	1.16	1.39	1.61
θ	30°	24°	18°
$\dfrac{D_{2s}^{A2}(\lambda_2)}{D_{1s}^{A2}(\lambda_2)}$	1.11	1.33	1.71
θ	32°	26°	16°
$\dfrac{\Phi_{2s}(\lambda_{\text{BM}})}{\Phi_{1s}(\lambda_{\text{BM}})}$	1.16	1.42	1.64
θ	30°	24°	17°

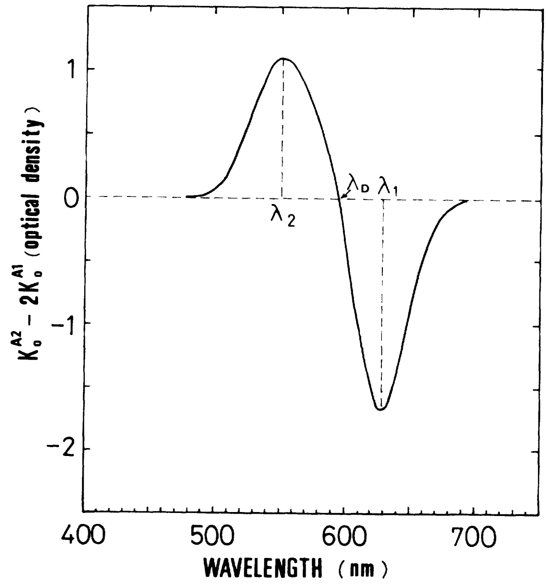

FIG. 7. Variations of $K_0^{A2}-2K_0^{A1}$ vs the wavelength.

TABLE II. Values of $-D^{A1}(\lambda_1)/D^{A2}(\lambda_2)$ for each exciting wavelength.

λ (nm)	$-\frac{D^{A1}(\lambda_1)}{D^{A2}(\lambda_2)}$
496.5	1.68±0.25
514.5	1.80±0.13
568.2	1.76±0.10
632.8	1.71±0.05

We computed Δn_0^{A1} and Δn_0^{A2}, taking for n_c the value 1.49 which is the refractive index of a pure KCl crystal. We assigned to f_1 and f_2 the respective values 0.33 and 0.66, neglecting the areas under the *K* and *L* bands. The results are represented in Fig. 8 (Δn_0^{A1} and Δn_0^{A2}) as a function of the wavelength. The quantity $k(\Delta n_0^{A2} - 2\Delta n_0^{A1})$ is shown in Fig. 9.

It may be seen in Eqs. (21) that the variations of the refractive index coupled with each band have always opposite signs. They partially compensate each other on the short-wavelength side of the F_{A2} band and long-wavelength side of the F_{A1} band. They are additive between the two absorption maxima λ_2 and λ_1 and the birefringence is maximum for a wavelength situated near that which cancels the dichroism.

C. Angle θ

The values obtained for θ (Table I) are strongly dependent on the exciting wavelength, as already noted in a previous paper.[10] Such a dispersion may be imputed to the *K* band which has been completely neglected in our theory. This is the reason why we consider only the values of θ obtained with the exciting wavelengths 568.2 and 632.8 nm. Consequently

$$\theta = 17° \pm 1° \,. \tag{33}$$

This value is in good agreement with those given in Refs. 10 and 16 for a crystal at liquid-nitrogen temperature.

VI. CONCLUSION

We presented in this paper the dispersion curves of dichroism and birefringence photoinduced in an F_A(II) KCl:Li crystal by a beam of linearly polarized light. These curves show that the variations of the photoinduced anisotropy versus the probe wavelength are only dependent on the optical properties of the crystal (optical absorption and refractive index) before the optical excitation.

At saturation, the photoinduced birefringence cannot be neglected for some probe wavelengths of the spectrum, situated in the neighborhood of λ_D (590 nm at liquid-nitrogen temperature) between the two absorption maxima. For these wavelengths, the dichroism of the sample is practically negligible and the medium may be considered as a pure phase medium.

It has been shown, moreover, that the angle θ may be deduced from the dichroism or birefringence induced at saturation by a wavelength of the F_{A2} band on the one hand, and a wavelength of the F_{A1} band on the other hand. Such a method is applicable even if the crystal still contains *F* centers since it only concerns the photoinduced anisotropy. Consequently, the measurements are as precise in the F_{A2} band as in the F_{A1} band.

In a next paper, the part of the *K* band in the reorientation process will be investigated. In fact, it cannot be completely neglected as shown here by the experimental results obtained with the 496.5- and 514.5-nm exciting radiations.

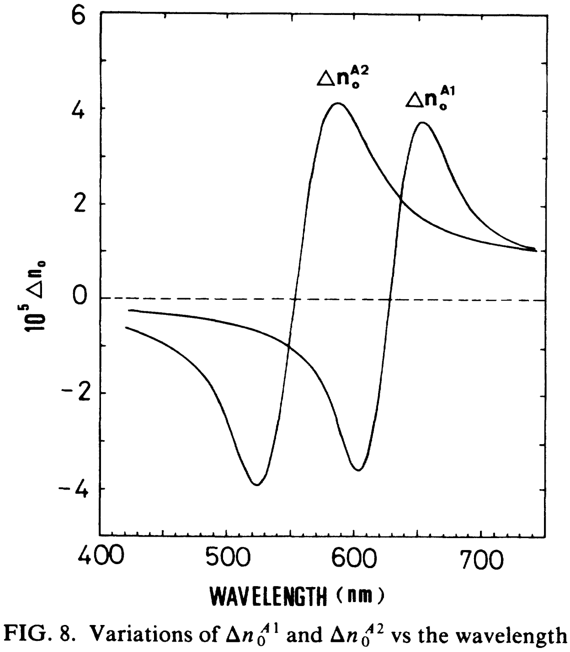

FIG. 8. Variations of Δn_0^{A1} and Δn_0^{A2} vs the wavelength.

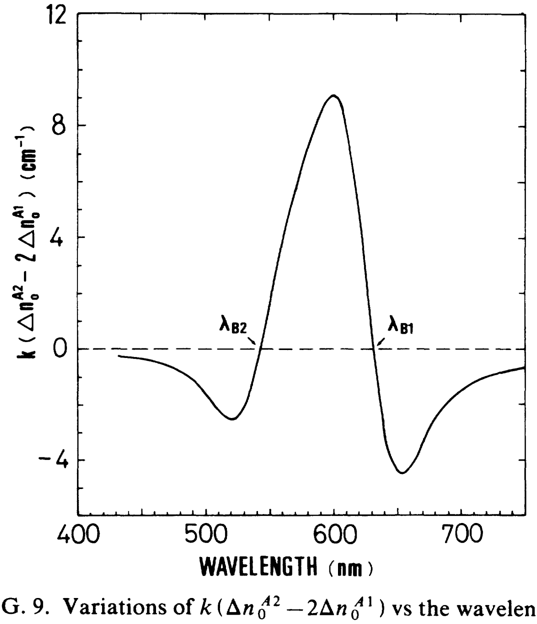

FIG. 9. Variations of $k(\Delta n_0^{A2} - 2\Delta n_0^{A1})$ vs the wavelength.

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