Electron-spin memory and magnetic-circular-dichroic effects in the luminescence of F centers in KI, KBr, and KCl

G. Baldacchini and A. Tanga
Comitato Nazionale per l'Energia Nucleare, Centro di Frascati, Casella Postale 65, 00044 Frascati, Rome, Italy

U. M. Grassano
Istituto di Fisica dell'Università di Roma and Gruppo Nazionale di Struttura della Materia del Consiglio Nazionale delle Ricerche, Rome, Italy

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We have solved the rate equations of the optical pumping cycle of the F center in the case of two spin-mixing parameters, ϵ_+ and ϵ_-. The solutions and the experimental results for the magnetic circular dichroism of the F-center emission are in complete agreement, and have allowed for the first time the measurement of the "anisotropy" of the electron-spin memory, i.e., $(\epsilon_- - \epsilon_+)/(\epsilon_- + \epsilon_+)$. This quantity will be very useful in order to clarify the role of the various processes responsible for the loss of the spin memory in the optical cycle. Moreover, from the rate equations we have been able to find the value of the polarization in the relaxed excited state, which is essential to calculate the spin-orbit splitting in emission.

I. INTRODUCTION

In the last few years several theoretical and experimental efforts have been made in order to clarify the nature of the relaxed excited state (RES) of F centers in alkali halides. As far as experimental work is concerned a special place is reserved to magnetic-circular-dichroic (MCD) effects in luminescence, to which we have made some contribution.[1] From the knowledge of such MCD effects it is possible to calculate important parameters characterizing the RES—for instance the spin-orbit coupling. Unfortunately such a calculation requires the precise value of the electron-spin polarization in the RES, P_ρ, which can be inferred from the solution of the rate equations[2] of the optical pumping cycle of the F center. We have solved these rate equations in a previous work,[1] but we have found some discrepancies between the experimental results and the theoretical expectation.

However, in the rate equations a central role is played by the spin-mixing parameter ϵ which takes into account the slight loss of electron-spin memory exhibited by the F center during the optical cycle. Recently it has been suggested[3,4] that the choice of a unique value of ϵ cannot be justified at all for the optical transition of the F center. By keeping in mind this suggestion we have solved again the rate equations in a few special cases and we have found good agreement with the experimental results. The conclusions outlined previously[1] concerning the RES are confirmed, and in addition we give in this paper the values of relevant properties of the spin-mixing processes.

II. OPTICAL PUMPING CYCLE

A decade ago it was found[5,6] that the electron of the F center exhibits in an external magnetic field an almost complete spin memory during the optical pumping cycle, Fig. 1. In other words, the direction of the spin is slightly affected by the absorption and emission of photons, and by phonon

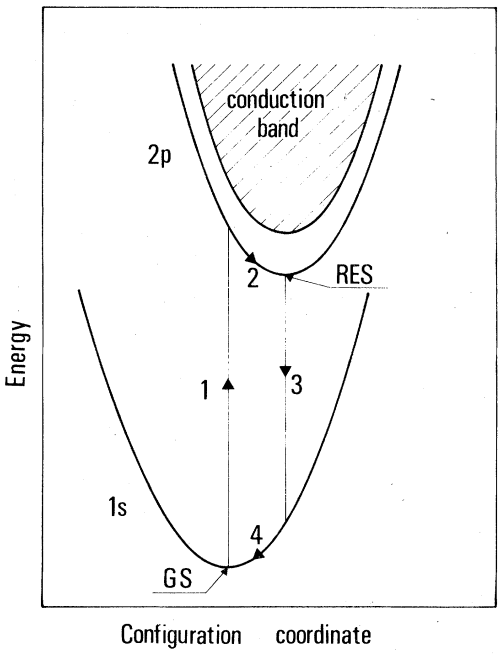

FIG. 1. Optical pumping cycle of F centers in alkali halides showing the photon (1,3) and phonon (2,4) processes.

relaxation. In order to describe completely the optical pumping cycle it was necessary to define the spin-mixing parameter ϵ, which is the fraction of spins flipped during one complete cycle. The introduction of ϵ in the rate equations[6] proved to be very useful in the description of a large variety of effects depending on the magnetic field.[2,6,7] However, by using the spin-mixing parameter introduced previously we were unable to explain some details found in the measurement of small MCD effects in the luminescence of F centers.[1,8] Independently it has been shown experimentally that the electronic polarization of the ground state (GS), P, does not follow completely the theoretical predictions made on the basis of the previous simple rate equations.[3] This last observation has been the starting point of a new critical study on the spin mixing, which brought new important results.[3,4] In the following we will review the main properties of ϵ, and we will use them to write the appropriate equations.

The slight loss of spin memory is a consequence of the electron states not being pure magnetic Zeeman levels. So a reversing of the spin is expected only when such states are partially mixed. The major contribution to ϵ during the cycle represented in Fig. 1 comes from process 1, where the electron is excited from the GS, $1s$-like, to a $2p$-like state. Indeed there is a considerable spin-orbit coupling in the $2p$ state, which mixes the pure spin levels. The two radiationless processes 2 and 4 are not known in detail so it is not possible to forecast exactly their influence upon ϵ. However, it is believed that they affect very little the spin states.[2,3,9] Finally, the hyperfine coupling between the F center and the surrounding nuclear spins gives some contribution to ϵ at low magnetic fields. Such interactions is present in the GS and much more in the RES, where the electron wave function is very diffuse. For this reason hyperfine mixing is often associated with luminescence.

A correct analysis of the various causes of spin mixing shows a peculiar difference between process 1 and all the others.[3,4] Indeed, the spin mixing due to spin-orbit coupling is different for the spin-up and the spin-down electrons. So it is necessary to introduce two values ϵ_+ and ϵ_- for the two Zeeman levels. On the contrary all the other processes lead to an isotropic value for the spin-mixing parameter, i.e., $\epsilon_+ = \epsilon_- = \epsilon^*$.

III. RATE EQUATIONS

Figure 2 shows the level scheme of the optical pumping cycle of the F center in a magnetic field. We assume that the spin-orbit coupling in absorption is the main mechanism for spin mixing, so that $\epsilon_+ \neq \epsilon_-$. We have neglected all the other contributions to the spin mixing for the following reasons. First of all, we study the effects in magnetic fields high enough so that the hyperfine interaction can be neglected. Second, the contribution due to the radiationless transitions, if any, is small and isotropic and the effect of ϵ^* on the rate equations seems to mitigate the anisotropy as it was shown in some simple cases.[3,10] Moreover, by neglecting ϵ^* one avoids further complication of the rate equations, whose solutions are often difficult to obtain in analytical form.

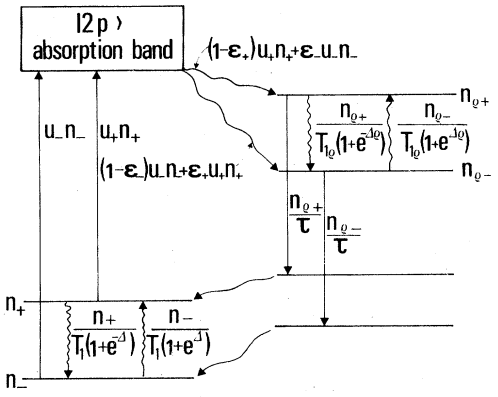

FIG. 2. Optical pumping cycle of the F center with the magnetic sublevels (shown). The figure refers to the case of anisotropic spin mixing, $\epsilon_- \neq \epsilon_+$. The Zeeman splittings are not drawn to scale.

In Fig. 2 n_+ (n_-) is the population of the $M_s = +\frac{1}{2}$ ($M_s = -\frac{1}{2}$) state, T_1 is the spin-lattice relaxation time in the GS, Δ is the energy separation of the Zeeman sublevels in units of $K_B T$, where K_B is the Boltzmann constant and T the absolute temperature, and τ is the lifetime of the RES. The corresponding quantities referring to the RES are indicated by the subscript ρ. The rate equations of the optical cycle are

$$
\begin{aligned}
\frac{dn_+}{dt} &= -u_+ n_+ + \frac{n_{\rho+}}{\tau} - \frac{1}{T_1}\left(n_+ - \frac{n_+ + n_-}{1+e^{\Delta}}\right), \\
\frac{dn_-}{dt} &= -u_- n_- + \frac{n_{\rho-}}{\tau} - \frac{1}{T_1}\left(n_- - \frac{n_+ + n_-}{1+e^{-\Delta}}\right), \\
\frac{dn_{\rho+}}{dt} &= (1-\epsilon_+)u_+ n_+ + \epsilon_- u_- n_- \\
&\quad - \frac{n_{\rho+}}{\tau} - \frac{1}{T_{1\rho}}\left(n_+ - \frac{n_{\rho+} + n_{\rho-}}{1+e^{\Delta\rho}}\right), \\
\frac{dn_{\rho-}}{dt} &= (1-\epsilon_-)u_- n_- + \epsilon_+ u_+ n_+ \\
&\quad - \frac{n_{\rho-}}{\tau} - \frac{1}{T_{1\rho}}\left(n_{\rho-} - \frac{n_{\rho+} + n_{\rho-}}{1+e^{-\Delta\rho}}\right).
\end{aligned}
\tag{1}
$$

By using the substitution $n=n_+-n_-$, $N=n_++n_-$, $n_\rho=n_{\rho+}-n_{\rho-}$, $N_\rho=n_{\rho+}+n_{\rho-}$, $u_++u_-=U$, $u_--u_+=UP_s$, where P_s is the dichroic differential absorption,[1,2] $\epsilon_++\epsilon_-=2\epsilon_0$, and $\epsilon_--\epsilon_+=\delta$, Eqs. (1) become

$$\begin{aligned}
\frac{dn}{dt}&=-\left(\frac{U}{2}+\frac{1}{T_1}\right)n+\frac{n_\rho}{\tau}+\left(\frac{U}{2}P_s-\frac{1}{T_1}\tanh\frac{\Delta}{2}\right)N,\\
\frac{dn_\rho}{dt}&=\frac{U}{2}[(1-2\epsilon_0)-\delta P_s]n-\left(1+\frac{\tau}{T_{1\rho}}\right)\frac{n_\rho}{\tau}\\
&\quad-\frac{U}{2}[(1-2\epsilon_0)P_s-\delta]N-\frac{1}{T_{1\rho}}\tanh\frac{\Delta_\rho}{2}N_\rho,\\
\frac{dN}{dt}&=-\frac{dN_\rho}{dt}=\frac{U}{2}P_sn-\frac{U}{2}N+\frac{N_\rho}{\tau}.
\end{aligned}\tag{2}$$

In order to start with more manageable equations we will make two approximations that we will keep from now on. First of all we confine our attention to the strong-pumping case, i.e., $\frac{1}{2}U\gg 1/T_1$, and so it is possible to drop the term containing the spin-lattice relaxation time T_1 in dn/dt. Second, we assume $P_sn/N\ll 1$, which is always satisfied within a few percents at worst, because $n/N\leq P_s$ and $P_s\leq 1$. As a consequence we have $N_\rho\cong NU\frac{1}{2}\tau$ from the third equation of (2) at equilibrium, $dN/dt=-dN_\rho/dt=0$. Eventually by defining the polarization of the GS, $P=n/N$, and of the RES, $P_\rho=n_\rho/N_\rho$, we obtain

$$\begin{aligned}
\frac{2}{U}\frac{dP}{dt}&=-P+P_\rho+P_s,\\
\tau\frac{dP_\rho}{dt}&=[(1-2\epsilon_0)-\delta P_s]P-\left(1+\frac{\tau}{T_{1\rho}}\right)P_\rho\\
&\quad-(1-2\epsilon_0)P_s+\delta-\frac{\tau}{T_{1\rho}}\tanh\frac{\Delta_\rho}{2},
\end{aligned}\tag{3}$$

which represents a system of two equations with two unknown quantities P and P_ρ.

IV. SOLUTION OF THE RATE EQUATIONS

We have solved the coupled differential Eqs. (3) in two cases: The stationary case where the pumping beam has a fixed polarization, circular right (σ^+), circular left (σ^-), or linear (π), and the case in which the pumping beam, constant in intensity, is modulated sinusoidally between the two states σ^+ and σ^-.

A. Pumping with fixed polarization

The polarization of the GS and the RES are given by

$$P=\frac{\left(1+\frac{\tau}{2\epsilon_0T_{1\rho}}\right)P_s+\frac{\delta}{2\epsilon_0}-\frac{\tau}{2\epsilon_0T_{1\rho}}\tanh\frac{\Delta_\rho}{2}}{1+\frac{\tau}{2\epsilon_0T_{1\rho}}+\frac{\delta}{2\epsilon_0}P_s},\tag{4}$$

$$P_\rho=\frac{\frac{\delta}{2\epsilon_0}(1-P_s^2)-\frac{\tau}{2\epsilon_0T_{1\rho}}\tanh\frac{\Delta_\rho}{2}}{1+\frac{\tau}{2\epsilon_0T_{1\rho}}+\frac{\delta}{2\epsilon_0}P_s}.\tag{5}$$

Equation (4) reduces to Eq. (9b) of Ref. 3 if we put $\tau/2\epsilon_0T_{1\rho}\ll 1$ as is the case for magnetic fields less than $\simeq 30$ kG in KI and KBr.[1,11,12] Besides, $\delta/2\epsilon_0<1$, as we will show later in the paper, and $P_s\ll 1$, so the terms $\delta P_s/2\epsilon_0$ and P_s can be neglected. In this case Eqs. (4) and (5) simplify to

$$P=P_s+\delta/2\epsilon_0,\tag{6}$$

$$P_\rho=\delta/2\epsilon.\tag{7}$$

It is obvious that the term $\delta/2\epsilon_0$, which can be opposite in sign with respect to P_s, has a big impact on both P and P_ρ. Indeed, they are quite different from the old solution[2,6] for which $\delta=0$, i.e., $\epsilon_+=\epsilon_-$. Note that both P_s and δ vanish if the pumping light is linearly polarized.

B. Pumping with modulated polarization

If the pump beam is sinusoidally modulated between σ_+ and σ_- we have to make the two following substitutions in Eqs. (3):

$$P_s\to P_s\sin\omega t,\quad \delta\to\delta\sin\omega t\tag{8}$$

where $\omega/2\pi$ is the frequency of the modulation. The solutions of Eqs. (3) in the case of modulated pumping can be simplified by dropping terms in δ^2 and P_s^2. Besides, we did not take into consideration terms oscillating at higher harmonics. In conclusion the amplitude of the polarization can be expressed as the sum of two vectors as in the following:

$$\begin{aligned}
P&=|\overline{P}(1)+\overline{P}(2)|,\\
P&=|\overline{P}_\rho(1)+\overline{P}_\rho(2)|,
\end{aligned}\tag{9}$$

where $P(1)$ and $P_\rho(1)$ represent the old solutions[1,2] for which $\delta=0$, and $P(2)$ and $P_\rho(2)$ contain the contribution of δ. In order to calculate the sums in Eqs. (9) the phase shifts $\varphi=\varphi(1)-\varphi(2)$ and $\varphi_\rho=\varphi_\rho(1)-\varphi_\rho(2)$ between the two vectors have to be known. The final result for P is given by

$$P(1)=P_s\frac{\frac{1}{2}U(\omega^2\tau^2+4\epsilon_0^2)^{1/2}}{(\tau^2\omega^4+\omega^2+\epsilon_0^2U^2)^{1/2}},\tag{10a}$$

$$P(2)=\frac{\delta}{2\epsilon_0}\frac{\epsilon_0U}{(\tau^2\omega^4+\omega^2+\epsilon^2U^2)^{1/2}},\tag{10b}$$

$$\varphi(1)=\arctan\left(\frac{(1-2\epsilon_0)\omega^2\tau+2\epsilon_0^2U}{\omega(2\epsilon_0+\tau^2\omega^2)}\right);\quad\left(-\frac{\pi}{2}--\pi\right),\tag{10c}$$

$$\varphi(2)=\arctan\left(\frac{\epsilon_0U-\tau\omega^2}{\omega}\right);\quad\left(-\frac{\pi}{2}--\frac{3}{2}\pi\right)\tag{10d}$$

and for P_ρ by

$$P_\rho(1) = (1-2\epsilon_0)P_s \frac{\omega}{(\tau^2\omega^4+\omega^2+\epsilon_0^2U^2)^{1/2}}, \tag{11a}$$

$$P_\rho(2) = \frac{\delta}{2\epsilon_0} \frac{(4\epsilon_0^2\omega^2+\epsilon_0^2U^2)^{1/2}}{(\tau^2\omega^4+\omega^2+\epsilon_0^2U^2)^{1/2}}, \tag{11b}$$

$$\varphi_\rho(2) = \arctan\left(\frac{-\omega}{\epsilon_0U-\tau\omega^2}\right); \quad (\pi-0), \tag{11c}$$

$$\varphi_\rho(2) = \arctan\left(\frac{\epsilon_0U+\omega^2 2/U}{[(1-2\epsilon_0)+(2/U)\tau\omega^2]\omega}\right); \quad \left(-\frac{\pi}{2}--\pi\right). \tag{11d}$$

On the right-hand sides (in parentheses) we give the ranges of variation of the phases when the modulation frequency ω changes from 0 to ∞.

In order to explain the features of the solutions we have sketched them in a qualitative manner, which, however, retains the main properties, in Figs. 3(a) and 3(b).

We consider the case $\delta/2\epsilon_0 < P_s$. Keeping in mind that one has to take into account the phase shifts to have the total polarization, we can make the following observations. The polarization of the GS has a significant value only at very low frequency of modulation, $\omega < \epsilon_0 U$ ($\epsilon_0 U \leq 10^4$ at the maximum reasonable pumping levels). Indeed at higher frequencies the polarization is lowered by the factor $\frac{1}{2}U\tau$ which is at most $\sim 10^{-2}$. However, for $\omega < \epsilon_0 U$ there is a contribution to P due to the anisotropy of the spin-mixing parameter, $P(2) \cong \delta/2\epsilon_0$, which can have the same sign of P_s or the opposite.

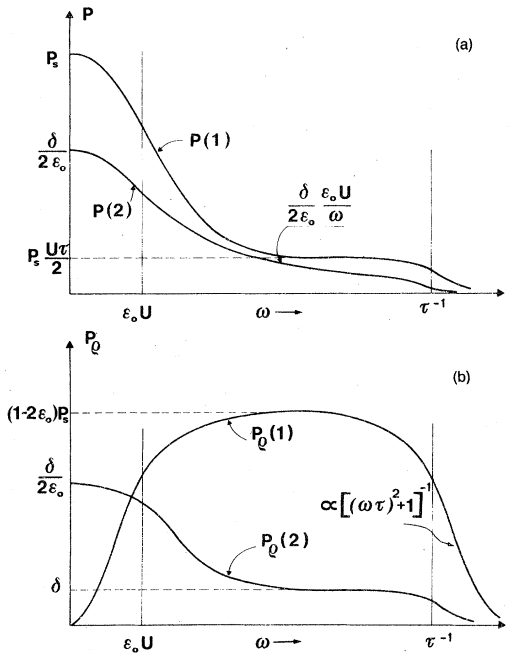

FIG. 3. Qualitative behavior of the solutions for P (a) and P_ρ (b) vs the frequency of modulation. The frequency scale is not linear and only the main features are reported along with some significant parameters. See text for details.

In the RES the situation is quite different. While $P_\rho(1)$ takes its broad maximum value at intermediate frequencies, $\epsilon_0 U < \omega < \tau^{-1}$, $P_\rho(2)$ gives its main contribution only at low frequencies, $\omega < \epsilon_0 U$ (note that $\delta < \delta/2\epsilon_0$ because $2\epsilon_0 < 1$). In this case the two effects, the dichroism in absorption, P_s, and the anisotropy of the spin memory, $\delta/2\epsilon_0$, are quite well separated in frequency.

As a last observation we wish to point out that the polarizations P and P_ρ as defined in Eqs. (9) coincide with the stationary solution (6) and (7) when $\omega \to 0$. Indeed this must be the case because the modulation pumping reduces to the stationary one when the frequency of modulation approaches zero.

V. EXPERIMENTAL RESULTS AND DISCUSSION

Some years ago we have reported extensive experimental measurements both on the diamagnetic and the paramagnetic effects of the F-center emission in KCl, KBr, and KI.[1,8] At that time we were really puzzled by some results that we could not understand in the light of the then known theory.

One of these unexplained results was the offset of the diamagnetic signal obtained with σ^+ or σ^- polarization of the pumping beam with respect to that produced by π polarization. This offset, which we called S_c, has been displayed in Figs. 3 and 4 of Ref. 1, and more clearly in Figs. 4 and 5 of Ref. 12, where the diamagnetic signals have been normalized to the luminescence intensity. It is evident at this point that the constant signal S_c is originated by the polarization of the RES given by formula (7), i.e., $S_c \propto P_\rho = \delta/2\epsilon_0$. For magnetic fields higher than 30 kG solution (7) is replaced by (5), where the relaxation time in the RES, $T_{1\rho}$, plays an important role. In a previous paper[12] we used the formula (5) together with the measured nonlinearity of the diamagnetic effect above 30 kG to obtain $T_{1\rho}$ in KBr and KI.

The anisotropy of the spin memory can also explain the behavior of the paramagnetic effect ΔS_p, especially at low frequencies of modulation, $\omega \to 0$. Indeed we have been able to fit the old experimental data (Figs. 9–11 of Ref. 1) by using Eqs. (9) and (11) containing the new term $P_\rho(2)$ [see Eq. (11b) and Fig. 3(b)]. The results of the fitting obtained by a least-square computer program are reported in Figs. 4–6, where the theoretical curves are plotted together with the experimental points

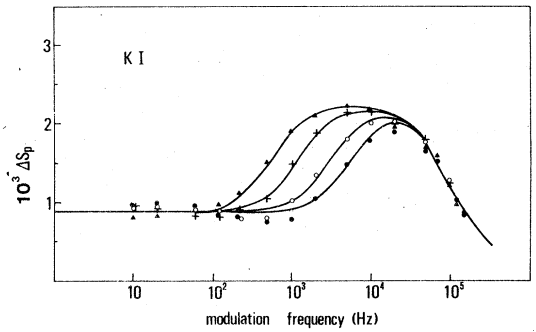

FIG. 4. Paramagnetic effect ΔS_p at 20 kG as a function of the modulation frequency, $\omega/2\pi$, in KI. Pumping power on the crystal surface, $W_0 = 7.5$ mW for the black circles, $\sim\frac{1}{2}W_0$ for empty circles, $\sim\frac{1}{4}W_0$ for crosses, and $\sim\frac{1}{10}W_0$ for triangles. The continuous curves represent the best fit of solutions on the experimental points.

for KI, KBr, and KCl, respectively. In Table I we report the curve-fitting parameters. The P_s values have been taken from the literature.[2] The pump rate U has been calculated by measuring the pumping power impinging on the samples. Because the value of U is only approximately known, there is an uncertainty in the numbers reported in the table, which reflects on the values of the spin-mixing parameter ϵ_0. Indeed from the fit we essentially derive the product $\epsilon_0 U$; see formulas (11) and Fig. 3(b). The values of U reported in the table represent the maximum power used. The various fitting curves on each of Figs. 4–6 are obtained with the same set of parameters by changing only the value of U according to the levels of pumping power used. The good general agreement of the fit is a strong proof of the fitness of the rate Eqs. (1) to explain the optical cycle of the F center.

Leaving out the anisotropy of the spin memory, $\delta/2\epsilon_0$, which represents a new result, all the other parameters in Table I almost coincide with those found previously.[1] Furthermore, the existence of a spin-mixing anisotropy does not substantially

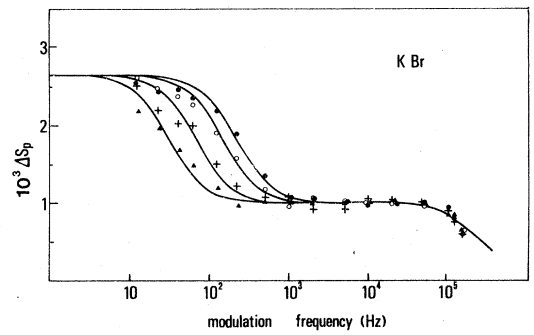

FIG. 5. Same as Fig. 4 for KBr.

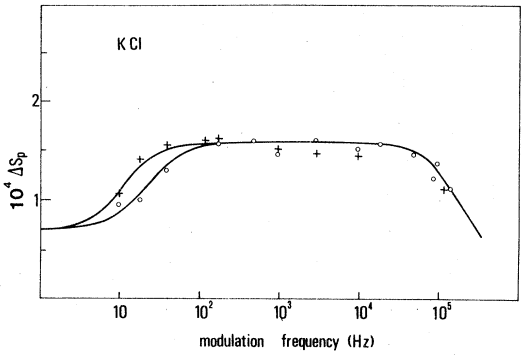

FIG. 6. Same as Fig. 4 for KCl. Pumping power on the crystal surface, $W_0 = 12.5$ mW for circles and $\sim\frac{1}{2}W_0$ for crosses.

change our knowledge of the properties of the RES.[1,13] But let us clarify better this point. As stressed in the Introduction, there is a strong interest in knowing the numerical values of the polarization in the RES, P_ρ. From Eqs. (11), with the parameters of Table I, we have calculated the values of P_ρ at intermediate frequencies of modulation, $\epsilon_0 U < \omega < \tau^{-1}$. Table II reports these values along with those of $\delta = \epsilon_- - \epsilon_+$ at the wavelength of the pumping radiation. The numbers in parentheses are obtained by taking the values of ϵ_0 given by Mollenauer *et al.*[2] At this point we can calculate the spin-orbit splitting $|\lambda|$ in the RES. The values (meV) are 1.0 (0.7) for KI, 0.6 (0.7) for KBr, and 0.4 (0.4) for KCl. The numbers in parentheses are obtained by taking the values in parentheses for P_ρ in Table II. These new values of $|\lambda|$ are almost the same as those given previously,[1] and as a consequence the conclusions drawn there remain substantially the same.

Recently Imanaka *et al.*[10] solved the rate equations of the optical cycle in the stationary case, by taking into consideration the various processes (see Chap. II) leading to the spin mixing. Moreover, they measured S_c in KBr pumping at the same our wavelength and they found $S_c = 1.4 \times 10^{-3}$ against our value[1] $S_c = 3 \times 10^{-3}$. Because it was found[8] that S_c decreases in KBr at high concentration of F centers, we suspect that the discrepancy could be due to different sample coloration.

TABLE I. Values of the parameters used for the fitting of the experimental data in Figs. 4–6. See text for details.

	P_s	U_{max} (sec^{-1})	ϵ_0	$\delta/2\epsilon_0$	τ (μsec)
KI	0.4	15×10^4	0.3	0.05	2.5
KBr	0.15	7.0×10^4	0.02	0.34	1.3
KCl	0.05	3.5×10^4	0.005	0.02	1.0

TABLE II. Anisotropy of the spin-mixing parameter $\delta = \epsilon_- - \epsilon_+$ is given at the wavelength of the pumping radiation λ_p, along with the calculated values of the polarization P_ρ. The numbers in parentheses are calculated using the values of ϵ_0 given by Molleanuer *et al.*[2]

	λ_p (Å)	$\delta = \epsilon_- - \epsilon_+$	P_ρ
KI	6328	0.030 (0.025)	0.13 (0.18)
KBr	6328	0.014 (0.027)	0.13 (0.11)
KCl	5145	0.0002 (0.0004)	0.05 (0.05)

The experimental value of $\delta/2\epsilon_0$ compares fairly well with the only calculation made in KBr.[4] Indeed Mauser *et al.* found $\delta/2\epsilon_0 = 0.3$ (value extracted from Fig. 1 of their work), against our $\delta/2\epsilon_0 = 0.34$. But the agreement is completely at odds when one tries to compare the single values of the two spin-mixing parameters, ϵ_+ or ϵ_-. However the calculated values of ϵ_+ and ϵ_- in Ref. 4 are not very reliable if taken singly.[14]

In conclusion we would like to stress the importance of the measurements of the anisotropy of the spin memory, i.e., $\delta/2\epsilon_0 = (\epsilon_- - \epsilon_+)/(\epsilon_- + \epsilon_+)$. First of all, its insertion in the optical cycle of the F center by means of the rate Eqs. (1) explains satisfactorily the experimental results.[1,3] Second, we hope that from its knowledge it will be possible to clarify much better the role of the various processes which contribute to the final value of the spin-mixing parameter.

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