Spin-lattice relaxation time of the relaxed excited state of the F center in KI and KBr

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(Received 15 June 1978)

The spin-lattice relaxation time of the relaxed excited state (RES) of the F center in KI and KBr has been measured at 1.85 K and at magnetic fields up to 80 kG. The experimental data can be fitted with the same analytical expression as that used for the ground state, but the relaxation times are about three orders of magnitude smaller than those of the ground state. An attempt to extend to the RES the mechanisms effective in the ground state proved to be unsatisfactory.

I. INTRODUCTION

The spin-lattice relaxation time T_1 is a quantity of fundamental importance for the knowledge of the properties of paramagnetic defects in crystals. In fact from its dependence upon the magnetic field, it is possible to derive information on the interaction of the defect with the surrounding lattice. The relaxation time T_1 has been extensively studied both experimentally and theoretically for the magnetically split ground state of the F center.[1]

In order to measure the T_1 value of an isolated F center, a very low concentration of defects ($N_F \simeq 10^{16}$ cm^{-3}) is necessary to avoid spurious contributions of mutual interactions. The technique used at low temperatures, of inverting the spin populations and following at successive times the recovery of the normal ESR signal, is therefore difficult to apply because of the small number of available unpaired spins.[2]

Other methods for observing T_1 are based on the magnetic circular dichroism (MCD) and on the Faraday rotation (FR) of the F-center absorption. In fact these optical methods reflect in analogous ways, but with greater sensitivity, the population difference of the ground-state magnetic sublevels. T_1 is thus measured from the time necessary to recover the equilibrium MCD or FR signal after a quick change of the magnetic field intensity. This technique for measuring T_1 has been first used by Karlov *et al.*,[3] Romestain and Margerie,[4] Mort *et al.*,[5] and more recently by Panepucci and Mollenauer,[6] and Carvalho *et al.*[7] for the F centers in several alkali halides.

Mollenauer and Pan[8] have shown that under suitable optical-pumping conditions, it is possible to obtain a polarization of the electronic population not only in the ground state, but also in the relaxed excited state (RES) of the F center. These polarizations depend upon the spin-lattice relaxation time of the RES, $T_{1\rho}$. Following a previous convention,[9,10] we label by ρ all quantities referring to the RES. From the polarization P induced in the ground state through the optical cycle, Mollenauer and Pan[8] have derived the value of $T_{1\rho}^{-1} \leq 1500$ sec^{-1} for the F centers in KI at 1.6 K and $B = 30$ kG. Likewise, from the polarization P_ρ of the RES, it is possible to obtain an independent measurement of the same quantity.

Still another way for measuring the value of $T_{1\rho}$ was reported by the present authors[10] and based on the field dependence of the paramagnetic component of the MCD signal of the F center emission. The decrease at high fields of the saturation value of the paramagnetic signal is interpreted as due to the field dependence of $T_{1\rho}$. The values of $T_{1\rho}$ obtained with this method for KI and KBr are only approximate, but they are until now the only measurements of $T_{1\rho}$ at fields up to 80 kG and at low temperatures, $T \simeq 2$ K. At much lower fields and higher temperatures, $T > 10$ K, there are some $T_{1\rho}$ measurements obtained by Ruedin *et al.*[11] They exploited the well-known quenching of the luminescence due to pair interaction of F centers.[12]

In this paper we report more accurate $T_{1\rho}$ measurements for the F center in KI and KBr derived from the MCD absorption and emission signals. The absorption technique is similar to that used by Mollenauer and Pan[8]; the emission technique will be described below. A summary of the theory is given in Sec. II. The experimental apparatus and results are presented in Sec. III. Section IV contains a brief discussion and comments.

II. MEASUREMENT METHODS OF $T_{1\rho}$

A. Magnetic-circular-dichroism absorption

The rate equations of the population of the RES of the F center with optical pumping have been solved in a few special cases.[8] For pumping with linearly polarized light (π), the ground-state polarization is given by

$$P=\frac{(n_+-n_-)}{(n_++n_-)}=\frac{-(T_p/T_1)\tanh\frac{1}{2}\Delta-[\tau/(\tau+2\epsilon T_{1\rho})]\tanh\frac{1}{2}\Delta_\rho}{1+T_p/T_1} \quad (1)$$

where

$$T_p^{-1}=\frac{\tau+2\epsilon T_{1\rho}}{\tau+T_{1\rho}}\,\frac{u_++u_-}{2},$$

ϵ is the spin-mixing parameter, n_+ and n_- are the populations, and u_+ and u_- the probabilities of transitions out of the $|+\frac{1}{2}\rangle$ and $|-\frac{1}{2}\rangle$ substates, τ is the radiative decay time, $\Delta=g\mu_BB/kT$, and $\Delta_\rho=g_\rho\mu_BB/kT$, where g and g_ρ are the g factors for the ground state and the RES, respectively, μ_B is the Bohr magneton, k is the Boltzmann constant, T the absolute temperature, and B is the magnetic field.

For very high pumping intensities $T_p\to 0$ and the term T_p/T_1 can be neglected. The ground-state polarization is then essentially given by

$$P=-[\tau/(\tau+2\epsilon T_{1\rho})]\tanh\tfrac{1}{2}\Delta_\rho. \quad (2)$$

The spin-lattice relaxations time $T_{1\rho}$ is much longer than the radiative lifetime τ, so that $\tau/T_{1\rho}\ll 1$. If we had neglected $\tau/T_{1\rho}$ in Eq. (2), P would have vanished and the MCD absorption signal, even at low temperatures, would only show the diamagnetic effect linear in the applied magnetic field.

We recall that the MCD signal is defined[13]

$$S=(I^+-I^-)/(I^++I^-)=C_dB+C_pP, \quad (3)$$

where $I^\pm$ are the intensities of the right- and left-circularly-polarized light ($\sigma^\pm$) transmitted through the crystal; C_d and C_p are the diamagnetic and paramagnetic constants. The deviation of the MCD signal vs the magnetic field from a straight line indicates the presence of a paramagnetic contribution due to a nonzero value of the polarization P. From the value of P and Eq. (2), it is then possible to evaluate $T_{1\rho}(B)$.

B. Magnetic-circular-dichroic emission

The MCD effect in the F-center emission has been throughly described by the present authors.[10] The rate equations of the pumping cycle allow the calculation of the polarization P_ρ of the RES. With steady optical pumping with an intense beam of linearly polarized light, assuming as before $T_p/T_1\ll 1$, one obtains

$$P_\rho=\frac{-(\tau/2\epsilon T_{1\rho})\tanh\frac{1}{2}\Delta_\rho}{1+\tau/2\epsilon T_{1\rho}}, \quad (4)$$

which coincides with the polarization of the ground-state P [see Eq. (2)].

As in the case of absorption, the polarization P_ρ is different from zero because $\tau/T_{1\rho}$ is not negligible. As a consequence the small paramagnetic contribution P_ρ produces the deviation from linearity at high magnetic fields of the diamagnetic MCD emission signal S_d. This behavior, reported previously,[10] is now completely accounted for. From the experimental values of P_ρ and Eq. (4), an independent set of values of $T_{1\rho}(B)$ can be calculated.

For pumping with steady circularly polarized light (σ^+ or σ^-), the rate equations have to be slightly changed in order to take into account the difference $\delta=\epsilon_--\epsilon_+$ in the spin-mixing parameters for the magnetic sublevels. Note that $\delta=0$ for pumping with π light. The implication of δ, introduced by Winnacker *et al.*,[14] on the rate equations will be discussed in detail in a forthcoming paper. However, the new parameter δ does not affect the values of $T_{1\rho}$. Indeed $T_{1\rho}$ is deduced as in the previous case (π pumping) from the nonlinearity of the field dependence of the MCD diamagnetic emission. On the other hand, the parameter δ changes the value of P_ρ as follows:

$$P_\rho=\frac{(\delta/2\epsilon_0)-(\tau/2\epsilon_0T_{1\rho})\tanh\frac{1}{2}\Delta_\rho}{1+\tau/2\epsilon_0T_{1\rho}}, \quad (5)$$

where δ is positive or negative depending on the pumping, σ^+ or σ^- and $\epsilon_0=\frac{1}{2}(\epsilon_-+\epsilon_+)$. This result explains the observed offset of the diamagnetic signals obtained with $\sigma^\pm$ pumping from that due to π excitation. The offset is proportional to the constant polarization $\pm\delta/2\epsilon_0$.

III. EXPERIMENTAL APPARATUS AND RESULTS

The experimental setup for measuring the MCD signal in absorption is shown schematically in Fig. 1. A He-Ne laser (Spectra Physics Model 124 A; output power $\simeq$20 mW in the TEM_{00} mode), linearly polarized, is focused to a spot of about 100 μm yielding a pump intensity on the sample surface of the order of 100 W/cm^2.

The same beam is also used to monitor the MCD signal, and therefore its polarization is modulated

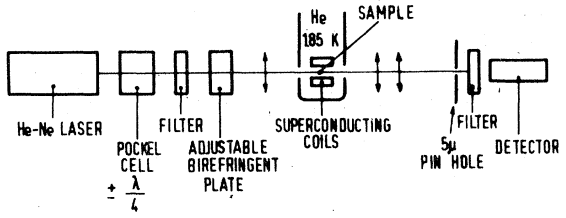

FIG. 1. Block diagram of the experimental apparatus of MCD absorption.

by a Pockel cell between right- and left-circularly-polarized light, σ^+ and σ^-, at the frequency of 20 kHz.

The time average of the polarization is therefore linear as supposed in deriving Eq. (1). An adjustable birefringent plate was used in addition to the modulation to adjust the symmetry of the modulation about the linear polarization. Because the ground-state polarization depends upon the pumping light intensity, it is important to measure signals coming only from a small area of the sample uniformly illuminated. For this reason the pumped area is imaged on a pin hole of 5 or 10 μm diameter, by one-to-one magnifying optics and the detector "sees" only this small part of the laser focal spot and not those parts that can be differently pumped. Infrared cutoff filters (Scott KG3) were used to stop infrared laser emission and F-center fluorescence. The modulated signal detected by a lock-in amplifier, were recorded together with the dc transmission.

F centers were produced by additive coloration of home-grown KI and KBr crystals. The number of F centers was approximately 3×10^{16} cm^{-3}. All measurements were performed at 1.85 K in a liquid-helium immersion cryostat equipped, with a superconducting magnet providing a field up to 80 kG.[15]

In order to calibrate the absolute value of the MCD signal, the paramagnetic signal S was measured at 1.85 K with an unfocused beam of very low intensity (≤ 50 μW/cm^2), and the saturation value at high fields was taken as the value corresponding to the saturation of the spin polarization $|P|=1$. The diamagnetic terms in these conditions are negligible. The value of the MCD signal with high-intensity pumping can thus be directly calibrated in percent of polarization (see Figs. 2 and 3).

Figure 2 shows the MCD signal for KI as a function of magnetic field. The straight line is the extrapolated diamagnetic signal, the slope coincides with the known values.[8] At low fields, the diamagnetic signal is predominant, but the deviation from the straight line at high fields shows

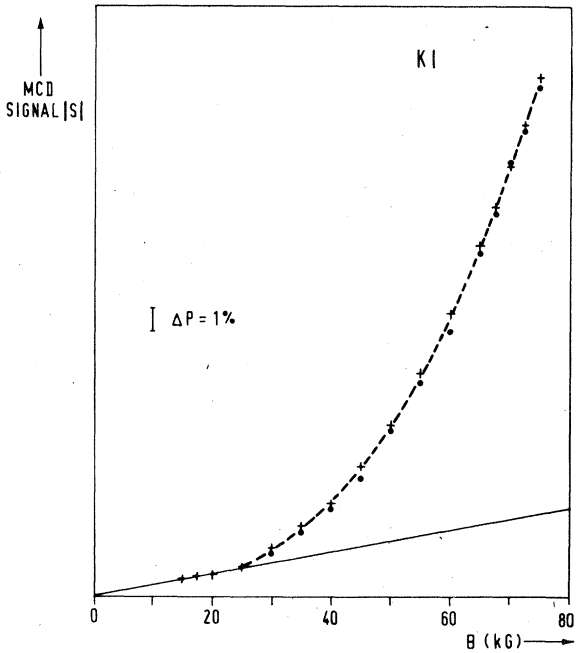

FIG. 2. Magnetic-circular-dichroic signal in KI as a function of magnetic field measured with saturating light pumping at 1.85 K. The symbols (+) and (•) refer to positive and negative magnetic fields, respectively. The straight line is an estimate of the diamagnetic signal. For calibration, the signal produced by a change of polarization $\Delta P=1\%$ is shown at the left-hand side.

the contribution to the polarization due to the $T_{1\rho}^{-1}$. No difference was noticed in the amplitude of the signal with two different pin holes of 5 and of 10 μm diameter, while larger pin holes yielded larger signals. For this reason, we believe we have quenched all contributions, due to unsaturated weakly pumped sample area, proportional to T_1^{-1}.

Analogus results are shown in Fig. 3 for KBr. In this case however the diamagnetic signals and

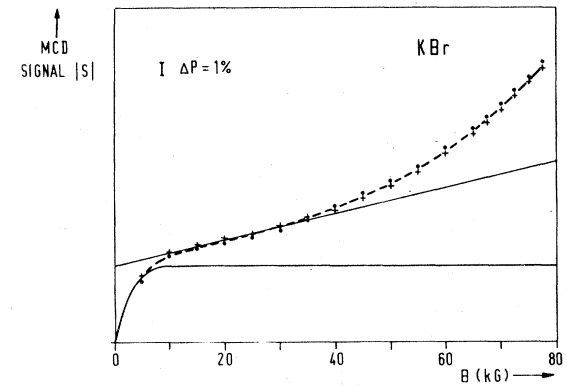

FIG. 3. Same as Fig. 2 but for KBr. For details see text.

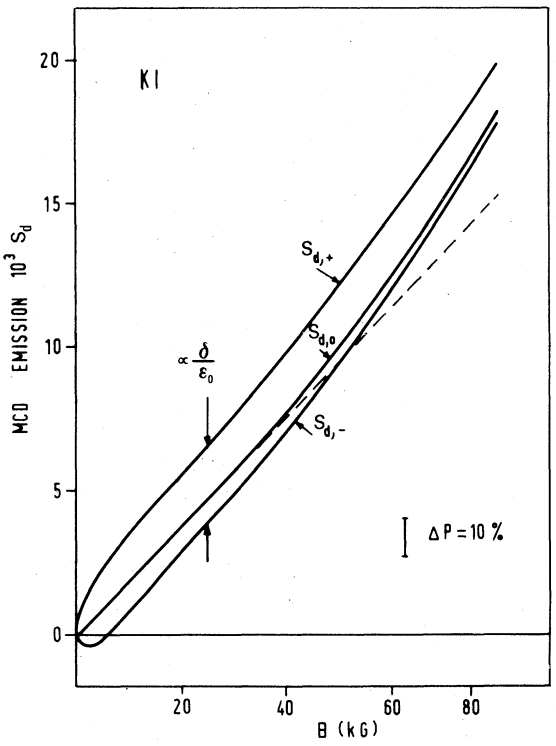

FIG. 4. Diamagnetic signals S_d of F-center emission as a function of the magnetic field for pumping with σ^+, σ^-, or π light at 1.85 K in KI. The offset $\propto \delta/\epsilon_0$ caused by the different spin-mixing parameters is also shown. The dashed line represents the pure diamagnetic signal. The change produced by a variation of polarization $\Delta P = 10\%$ is also shown.

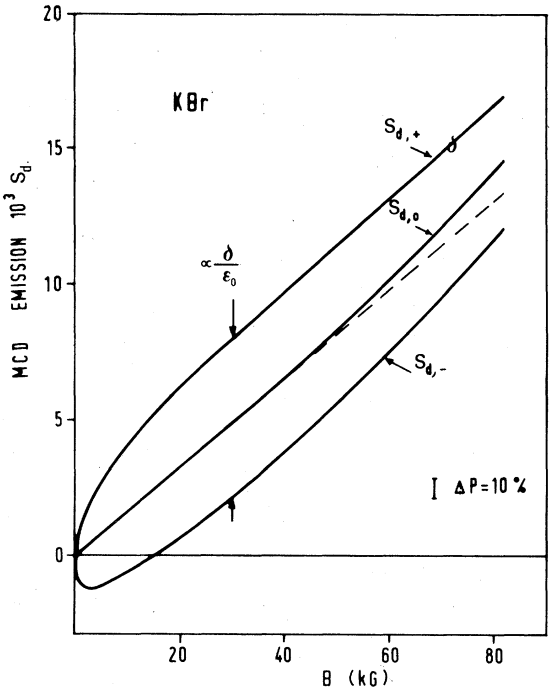

FIG. 5. Same as Fig. 4 but for KBr.

the paramagnetic contributions add up to an unexpected background consisting of a signal saturating at about 15 kG. The origin of this signal is unknown at present.

The apparatus used in the diamagnetic emission measurements and the results obtained have been recently described.[10] For completeness sake we report again in Figs. 4 and 5 the diamagnetic signal S_d in KI and KBr respectively derived from Figs. 3 and 4 of Ref. 10. We would like to point out that the Eqs. 4 and 5 have been obtained under the assumption $T_p/T_1 \ll 1$. This condition is now always satisfied because the largest part of the emission signal comes indeed from the most intensely pumped area of the crystal. On the contrary, in the absorption case, the weakly pumped regions gave a large (paramagnetic) signal that had to be suppressed (for example, with the use of pin holes).

IV. DISCUSSION

Values of $T_{1\rho}$ have been calculated by means of Eqs. (2), (4), and (5), from the experimental values of the MCD signals after subtraction of the diamagnetic part and, for KBr in absorption, of the saturating signal mentioned in Sec. III.

Figure 6 shows $T_{1\rho}^{-1}$ derived from the MCD absorption measurements described above. The squares represent the average of the $T_{1\rho}^{-1}$ values obtained from the MCD emission in the cases of π, σ^+, and σ^- pumping. The results are of the same order of magnitude of the data reported previously.[8,10] However, the experimental error is estimated to be here much smaller, especially at high fields.

A proper theory of the relaxation time in the RES is not presently available. However, the experimental behavior in KI and KBr suggests that the same direct processes are effective in both the ground state and the RES. For this reason we have fitted the experimental data to the following equation[6]:

$$T_{1\rho}^{-1} = (A_\rho B^3 + B_\rho B^5) \coth(g_\rho \mu_B B/2kT). \quad (6)$$

In the ground state the first relaxation process, with the B^3 dependence, is due to the hyperfine interaction of the F-center electron with the surrounding nuclei. The second term, proportional to B^5, reflects the phonon modulation of the spin-orbit coupling through the crystal field (Kronig–Van Vleck mechanism).

The parameters A_ρ and B_ρ of Eq. (6) have been determined with a least-squares-fit calculation

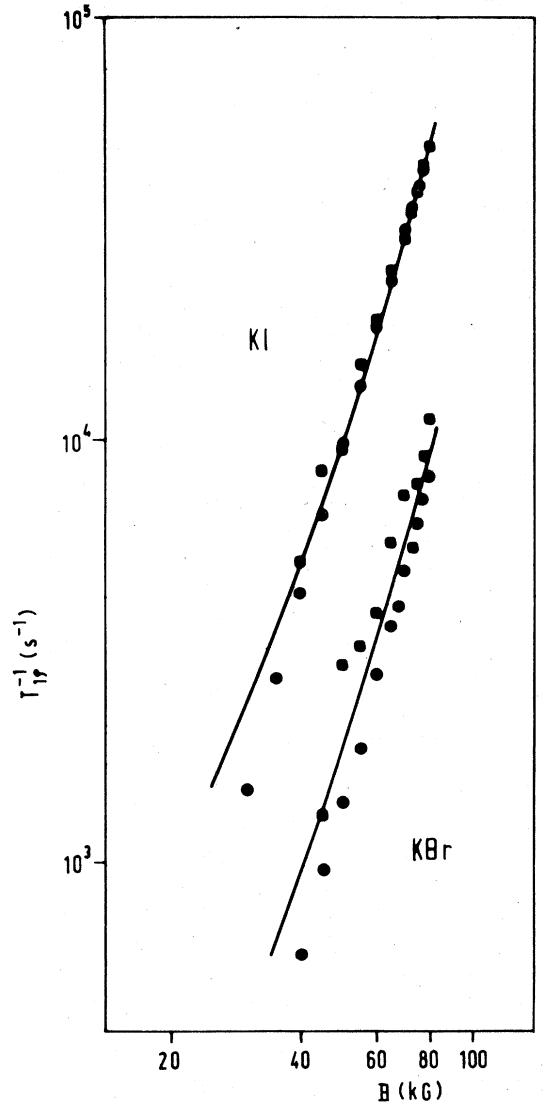

FIG. 6. Spin-lattice relaxation time $T_{1\rho}$ vs magnetic field at 1.85 K in KI and KBr. The circles represent the results obtained in absorption, while the squares refer to the emission measurements.

and are reported in Table I together with the corresponding values for the ground state.[6] The best fits of Eq. (6) are plotted as full lines in Fig. 6. The spin-lattice relaxation times are a few orders of magnitude shorter for the RES than for the ground state, and consequently, both A_ρ and B_ρ are likewise larger.

We have tried to extend to the RES the theory given by Mollenauer and Panepucci for the hyperfine term (B^3) in the ground state.[6] Two possible wave functions for the RES have been taken into account: one $|2s\rangle$-like and one $|2p\rangle$-like. The parameters of these functions were chosen according to the ENDOR results in the RES.[16,17]

TABLE I. Values of the parameters A_ρ and B_ρ obtained by fitting the experimental points to Eq. (6). The analogous quantities A and B, relative to the ground state are taken from Ref. 6.

	A ($sec^{-1}G^{-3}$)	A_ρ ($sec^{-1}G^{-3}$)	B ($sec^{-1}G^{-5}$)	B_ρ ($sec^{-1}G^{-5}$)
KI	9.71×10^{-14}	5.7×10^{-11}	10.0×10^{-23}	6.0×10^{-21}
KBr	1.05×10^{-14}	1.2×10^{-11}	1.68×10^{-23}	1.0×10^{-21}

In both cases the calculated values of A_ρ are of the same order of magnitude of those of the ground state A, and therefore much smaller than the experimental ones. For this reason we conclude that the hyperfine interaction cannot be the main mechanism responsible for such short relaxation times. We wish to point out, however, that another process could be relevant for the spin relaxation in the RES. Indeed because of the extension of the wave function over many lattice distances, the dipole-dipole interaction may play a role much more effective than in the ground state.[6]

For the coefficient of the B^5 term the extension to the RES of the formula given for the Kronig–Van Vleck mechanism in the ground state[6] has been made using the spin-orbit splitting λ and the energy separation E_ρ between the RES and the next excited state in the relaxed configuration. Both quantities have been measured from the magnetic-circular-dichroic effects in the luminescence.[10,18] The constant B_ρ can thus be written:

$$B_\rho = \frac{3}{4\pi^2\hbar^4}\frac{e^4}{\rho d^2}\left(\frac{g_\rho\mu_B}{v}\right)^5\frac{\lambda^2}{E_\rho^4}, \tag{7}$$

where e is the electronic charge, ρ the crystal density, d the interionic distance, and v the velocity of sound. The calculated values of B_ρ are reported in Table II. In the same table the ratios $B_{\rho\text{theor}}/B_{\rho\text{expt}}$ and[6] $B_{\text{theor}}/B_{\text{expt}}$ are given for comparison. The agreement between the experimental and theoretical values is not very good in either the ground state or the RES. However, the formula given by Panepucci and Mollenauer[6] is a very crude extension of an expression calculated by Kronig for a different system. Until a proper calculation for the F center is lacking, we can only state that the previous mechanism of relaxation gives only approximately the proper dependence upon the spin-orbit splitting λ and the energy separation E_ρ.

Recently a theory[7,19] has been developed in which the phonon modulation of the g factor is the mechanism responsible for the relaxation in the ground state. This is a consequence of the fact that the

TABLE II. Values of the parameter B_ρ computed from the expression (7) and in comparison with B_ρ experimental. The parameters given in the table were used in the calculation. Also the ratio $B_{\text{theor}}/B_{\text{expt}}$ in the ground state (Ref. 6) is reported.

	λ (meV)	E_ρ (meV)	$B_{\rho\ \text{theor}}$ ($\text{sec}^{-1}\ \text{G}^{-5}$)	$B_{\rho\ \text{theor}}/B_{\rho\ \text{expt}}$	$B_{\text{theor}}/B_{\text{expt}}$
KI	0.6	60	3.5×10^{-19}	62	24
KBr	0.6	70	2.2×10^{-19}	220	24.4

g shift $\Delta g = g - g_e$ depends on the position of the ions around the vacancy.[20] The g shift comes from the orthogonalization of the F-center wavefunction to the crystal ion-core orbitals. This new process has the same dependence on magnetic field and temperature as the Kronig–Van Vleck process, but the following expression for B_ρ:

$$B_\rho \cong 3.3\times10^{-3}(g^3\mu_B^5/\hbar^4)(\Delta g)^2(1/\rho v^5). \quad (8)$$

The extension of this mechanism to the RES is possible because the g shift in the RES is originated mainly by the same effect as in the ground state.[21] Indeed starting from an expression valid for the ground state,[22] we can write for the relaxed configuration $\Delta g_\rho \propto ZA/E_\rho$, where Z is the nuclear charge of the ion, A is the so-called amplification factor and $E_\rho \simeq 0.1$ eV. In Table III we have normalized the g shifts to that of KI, and the values obtained are compared to the experimental ones. The extraordinary agreement is clearly fortuitous in view of the many approximations made. Nevertheless it strongly supports the explanation given above for the origin of the big negative g shifts.

The vibronic mixing between the $|2s\rangle$ and the $|2p\rangle$ excited states has been recently taken as the only source of the large g shifts.[23] However in this framework the spin-orbit coupling λ must be positive, while it is expected[21] negative as in the unrelaxed configuration, and the calculated values of the polarization P_ρ are surprisingly smaller that those obtained from the rate equations of the optical cycle.[10,24] Furthermore considering that important experimental features in KI cannot be fitted at all, the above hypothesis appears hardly acceptable. In conclusion while not excluding a small vibronic contribution, it seems more reasonable in the light of the present knowledge to ascribe the large g shifts to the interaction of the RES wave function with the ions surrounding the F center.

We used formula (8) to calculate both B and B_ρ, and the results are given in Table IV together with the experimental ones. The agreement seems to be much better than by using the Kramer–Van Vleck theory, see Table II. However, as shown by Carvalho *et al.*,[7] this theory does not work equally well for all the F centers in various crystals.

In conclusion we can say that the state of knowledge on the relaxation processes in the RES is still far away from being satisfactory. A simple extension of the mechanism active in the ground state for the cubic dependence on the magnetic field is not enough to explain the experimental data. On the other hand, two theories for the fifth-power dependence on the magnetic field seem to be capable in principle to fit the experimental results. It is our belief that more experimental data have to be obtained and proper theories have to be developed in order to clarify the whole situation. However, we wish to point out that every improvement in our knowledge of the relaxation processes in the RES requires a better knowledge of the RES itself.

TABLE III. g shifts in the RES are calculated from $\Delta g \propto ZA/E_\rho$ and normalized to that of KI. The values of Z are also given. The experimental results are shown in the last column. The two theoretical values for KF refer to the choice of the negative or positive ions in the calculation of Δg.

	A	Z	$\Delta g/\Delta g_{\text{KI}}$ (theor)	$\Delta g/\Delta g_{\text{KI}}$ (expt)
KI	7500	53	1	1
KBr	4000	35	0.35	0.37
KCl	1500	17	0.065	0.064
KF	350×650	9×19	0.010×0.030	0.020

TABLE IV. B and B_ρ calculated with the formula (8). A comparison with the experimental values is also given.

	B_{theor} ($\text{sec}^{-1}\ \text{G}^{-5}$)	$B_{\text{theor}}/B_{\text{expt}}$	$B_{\rho\ \text{theor}}$ ($\text{sec}^{-1}\ \text{G}^{-5}$)	$B_{\rho\ \text{theor}}/B_{\rho\ \text{expt}}$
KI	3.1×10^{-23}	0.31	3.1×10^{-21}	0.5
KBr	4.7×10^{-24}	0.28	2.3×10^{-22}	0.23

ACKNOWLEDGMENTS

The authors are indebted to Dr. M. Altarelli for helpful discussions and to Dr. A. De Angelis for the loan of the He-Ne laser. Two of us (U. M. G. and A. T.) are grateful to the Laboratories of Comitato Nazionale per l'Energia Nucleare Frascati for the hospitality during the completion of this work.

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