for these two regions, respectively, where $R_1/R = ((I_c+I)/2I_c)^{1/2}$. The pattern of current flow when the applied current is reduced after an excursion to $I \approx I_c$ is shown schematically in Fig. 4(b).

Introducing Eqs. (6) and (7) in Eq. (5) we have calculated $4\pi\bar{M}_z$ by numerical integration as a function of I/I_c with α as given above for various H_a. The remanent longitudinal magnetization calculated for $I=0$ after an excursion to I_c is shown versus H_a by curve B of Fig. 2. At low fields the calculated values deviate considerably from the data since the condition $\langle|B_\theta|\rangle_{\text{av}} \gg \langle|B_z-H_a|\rangle_{\text{av}}$ is not satisfied. Curve G of Fig. 3 shows the behavior of $4\pi\bar{M}_z$ calculated as I varies from $\pm I_c$ to $\mp I_c$ in a static field. We note that the calculated curve is symmetric whereas the experimental data yield asymmetric curves. In practice, the applied current can only be raised close to I_c when cycled. When this fact is taken into account in our model and appropriate expressions are derived, curves of $4\pi\bar{M}_z$ versus I/I_c exhibiting an asymmetry in qualitative agreement with the data are obtained. A discussion of this feature is beyond the scope of this preliminary report.

CONCLUSION

The longitudinal magnetization induced in cylindrical nonideal type-II superconductors by applied currents in the presence of static longitudinal fields is satisfactorily accounted for by a model which requires that (i) the net critical Lorentz force density is constant throughout the specimen and is directed radially inwards and (ii) the existing circular flux configuration is preserved consistent with the change in the applied current. From this model, paramagnetic and diamagnetic helical configurations of conduction and induced currents are thought to arise and coexist in a cylindrical wire when an applied current is cycled in the presence of a static longitudinal field. The helical circulating currents which remain after passage of the applied current and fill the bulk of the specimen can give rise to an appreciable longitudinal diamagnetic moment.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge the assistance of J. A. Szabo and Dr. D. J. Griffiths in the measurements and the collaboration of C. T. M. Chang in the calculations.

PHYSICAL REVIEW VOLUME 143, NUMBER 1 MARCH 1966

Selective Absorption of Circularly Polarized Light in Broad Bands by the Zeeman Components of Divalent Thulium in Calcium Fluoride*

C. H. ANDERSON, H. A. WEAKLIEM, AND E. S. SABISKY
RCA Laboratories, Princeton, New Jersey

(Received 22 September 1965)

The paramagnetic circular dichroism of the $4f$-$5d$ absorption bands of CaF_2:Tm^{2+} was investigated at 2.0°K and 9.0 kG. Using group theory it is shown that the bands can be separated into $E_{5/2}$ and $G_{3/2}$ character. Experimentally it was found that in this case the bands are primarily $G_{3/2}$ in character. Optical pumping experiments were performed where changes as large as $\pm 20\%$ were easily observed in the electron-paramagnetic-resonance absorption signal at X-band frequencies and 1.5°K by changing the polarization of the pump light from right to left circular. A method for optically monitoring the difference in the population of the two Zeeman components of the ground state is also reported on.

I. INTRODUCTION

AT the first Quantum Electronics Conference, Brossel made the observation that if the magnetic sublevels of the ground state of a paramagnetic ion in a solid absorbed polarized light in broad absorption bands by different amounts, one had a system very suitable for achieving large population changes in these levels by optical pumping.[1] Margerie[2] demonstrated that the four sublevels in the ground state of Cr^{3+} in ruby did have slightly different absorption coefficients for circularly polarized light in the $^4A_2$-$^4F_2$ green band. Karlov[3] with Margerie then demonstrated that the induced circular dichroism in the F center in KBr was as large as 9% in some regions of the bands and demonstrated that the spins could be inverted by optically pumping with circularly polarized light. Since then, Margerie[4] and others have made measurements on a number of other alkali halides with F centers.

There has also been a renewed interest in Faraday

---

[*] The research reported in this paper was sponsored by the Department of Defense and RCA Laboratories, Princeton, New Jersey.

[1] J. Brossel, *Quantum Electronics* (Columbia University Press, New York, 1960), p. 81.

[2] J. Margerie, Comp. Rend. **257**, 2634 (1963).

[3] N. V. Karlov, J. Margerie, and V. Merle D'Aubigne, J. Phys. **24**, 717 (1963).

[4] J. Margerie and R. Romestain, Comp. Rend. **258**, 4490 (1964). R. Romestain and J. Margerie, *ibid.* **258**, 2525 (1964). J. Gareyte and V. Merle D'Aubigne, *ibid.* **258**, 6393 (1964).

rotation measurements which are related to the induced circular dichroism by a Kramers-Kronig relationship. These measurements have given complementary results for the measurements of spin-orbit coupling in the *F* centers obtained from the induced circular dichroism measurements.[5] Faraday rotation and circular dichroism measurements have been made in CaF_2:Eu^{2+},[6] which has been suggested as a suitable system for studying optical pumping in solids.[7]

We wish to report here the measurement of the induced circular dichroism in CaF_2:Tm^{2+} and some preliminary measurements on optically pumping the spins. This system differs from the better understood *F* centers and CaF_2:Eu^{2+} in that the ground state of the ion is not an *S* state and there are a few hundred allowed electric dipole transitions which make up the bands. However, the divalent thulium is in a cubic environment and its 4*f*-5*d* bands start at 7000 Å, thus making it more convenient to study than the trivalent ions whose 4*f*-5*d* transitions occur in the ultraviolet. Shen[8] has made some Faraday measurements on the bands of CaF_2:Tm^{2+} but did not go into any details.

II. ENERGY LEVELS, SYMMETRIES, AND SELECTION RULES FOR CaF_2:Tm^{2+}

The energy levels of divalent thulium in calcium fluoride are shown in Fig. 1 as determined by the optical measurements of Kiss.[9] The ground state is the $E_{5/2}$ cubic crystal field component of the $^2F_{7/2}$ spin-orbit term of the $4f^{13}$ configuration. The first allowed electric dipole transitions are to the $5d\ 4f^{12}$ configuration which form a series of strong absorption bands starting at 7000 Å and going up into the uv, the center of gravity of the bands falling around 2000 Å.[10]

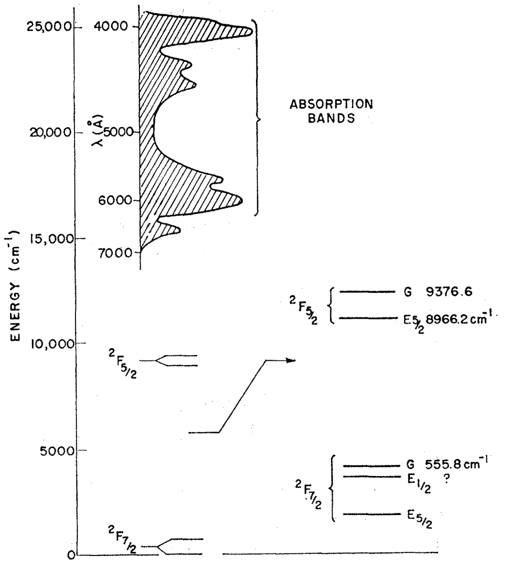

FIG. 1. Energy levels of CaF_2:Tm^{2+}.

If we assume that the eight neighboring fluorines around the thulium ion remain in a cubic structure during the transition (Franck-Condon principle), then the selection rules for the transition can be obtained by examining the symmetries of the electronic states in the point group O_h. This neglects any Jahn-Teller effects in the excited states and any simultaneous phonon emissions or absorptions. In the point group O_h the $5d\ 4f^{12}$ configuration contains the irreducible representations $152\ G_{3/2}+76E_{5/2}+75E_{1/2}$ regardless of how the 5*d* electron and the 4*f* electrons interact with each other and the crystal field. The electric dipole operator transforms as T_1 and, for right circularly polarized light $[T_1(+1)]$, the relative coupling coefficients $\langle E_{5/2}\pm 5/2|T_1(+1)|\Gamma_\mu\rangle$ are shown in Table I. From this we have for right circularly polarized light: (a) all transitions to the $E_{1/2}$ levels are forbidden, (b) only the $E_{-5/2}$ component is coupled to the $E_{5/2}$ levels, and (c) the transitions from the $E_{5/2}$ component are three times stronger to the $G_{3/2}$ levels than the corresponding transitions from the $E_{-5/2}$ component.

TABLE I. Normalized coupling coefficients. $\langle E_{5/2}\pm\frac{5}{2}|T_1 1|\Gamma\mu\rangle$ for the group O_h.

	$E_{1/2}$		$E_{5/2}$		$G_{3/2}$			
	$\frac{1}{2}$	$-\frac{1}{2}$	$\frac{5}{2}$	$-\frac{5}{2}$	$\frac{3}{2}$	$\frac{1}{2}$	$-\frac{1}{2}$	$-\frac{3}{2}$
$E_{5/2}$	0	0	0	0	0	0	-1	0
$E_{-5/2}$	0	0	$-\sqrt{\frac{2}{3}}$	0	0	0	0	$\sqrt{\frac{1}{3}}$

Formally, the CaF_2:Tm^{2+} is similar to the *F* center case, since in this notation the $^2S_{1/2}$ ground state of the *F* center is an $E_{1/2}$ level and the band is split by the spin-orbit coupling into the levels $^2P_{1/2}$ and $^2P_{3/2}$ which are $E_{1/2}$ and $G_{3/2}$ levels. The selection rules for these levels are similar to those given above for Tm^{2+} when the $\frac{5}{2}$ and $\frac{1}{2}$ notations are interchanged. The latter however, has only two states which make up the band and the absolute absorption coefficients for these $E_{1/2}$ and $G_{3/2}$ states are related. The CaF_2:Tm^{2+} has many more states and no such clear relationship exists between the absorption coefficients for the different states.

III. ABSORPTION COEFFICIENTS

The ground state splits in a magnetic field with a *g* value of +3.453 and the 100% Tm^{169} isotope with a spin of $\frac{1}{2}$ introduces a hyperfine splitting in the ground state of 1.098 Gc.[11] Above a few hundred gauss the

---

[5] F. Luty and J. Mort, Phys. Rev. Letters **12**, 45 (1964). J. Mort, F. Luty, and F. C. Brown, Phys. Rev. **137**, A566 (1965).

[6] Y. R. Shen and N. Bloembergen, Phys. Rev. **133**, A515 (1964); B. P. Zakharchenya and A. Va. Ryskin, Opt. Spectry. (USSR) **14**, 163 (1963).

[7] G. W. Series and M. J. Taylor, J. Phys. Radium **19**, 901 (1958).

[8] Y. R. Shen, Phys. Rev. **134**, A661 (1964).

[9] Z. J. Kiss, Phys. Rev. **127**, 718 (1962).

[10] D. S. McClure and Z. J. Kiss, J. Chem. Phys. **39**, 3251 (1963); G. H. Dieke and H. M. Crosswhite, Appl. Optics **2**, 675 (1963).

[11] W. Hayes and J. W. Twidell, J. Chem. Phys. **35**, 1521 (1961).

effects of the hyperfine interaction can be completely ignored. Modest magnetic fields (<10 kG) will split the ground and excited states by at most a few wave numbers; however, the bands are usually a couple of hundred wave numbers broad. Thus, to first order we ignore these shifts and calculate the paramagnetic contribution to the induced circular dichroism.

The absorption coefficient for right circularly polarized light directed along the magnetic field is given by

$$\alpha^{+}(\nu)=\frac{1}{2\cosh(g\beta H/2kT)}\times[\alpha_{+5/2}(\nu)e^{-g\beta H/2kT}+\alpha_{-5/2}(\nu)e^{+g\beta H/2kT}] \quad (1)$$

and for left circularly polarized light directed along the field, which from time reversal symmetry is the same as right circular directed against the field direction, we have

$$\alpha^{-}(\nu)=\frac{1}{2\cosh(g\beta H/2kT)}\times[\alpha_{+5/2}(\nu)e^{+g\beta H/2kT}+\alpha_{-5/2}(\nu)e^{-g\beta H/2kT}], \quad (2)$$

where $\alpha_{+5/2}(\nu)$ is the absorption coefficient for right circularly polarized light with all the ions in the $+\frac{5}{2}$ level and $\alpha_{-5/2}$ is the corresponding absorption coefficient with all the ions in the $-\frac{5}{2}$ level. The difference in the absorption coefficients (or circular dichroism) is given by

$$\alpha^{+}(\nu)-\alpha^{-}(\nu)=[\alpha_{+5/2}(\nu)-\alpha_{-5/2}(\nu)]\tanh(g\beta H/2kT) \quad (3)$$

and the average absorption coefficient is given by

$$\frac{\alpha^{+}(\nu)+\alpha^{-}(\nu)}{2}=\frac{\alpha_{+5/2}(\nu)+\alpha_{-5/2}(\nu)}{2}. \quad (4)$$

If the division of the bands into $E_{5/2}$ and G character is possible using the selection rules above, then we can say

$$\alpha_{+5/2}(\nu)=\alpha_{G}(\nu), \quad (5)$$

and

$$\alpha_{-5/2}(\nu)=\tfrac{1}{3}\alpha_{G}(\nu)+\alpha_{E_{5/2}}(\nu). \quad (6)$$

IV. OPTICAL ABSORPTION EXPERIMENTS

A liquid-helium Dewar with good quality, strain free optical windows was made to fit inside the sample compartment of a Cary 14 dual beam spectrophotometer. A small superconducting solenoid $1\frac{1}{2}$ in. long with a $\frac{1}{2}$-in. bore was put horizontally in the liquid helium at the sample position; a field of 12 kG was easily obtained. The light in the two beams was linearly polarized using a Glan polarizer and then made circular with "Polaroid" plastic quarter wave plates. The plastic quarter wave plate was calibrated against a quartz Soliel compensator and was found to be satisfactory over the whole visible spectrum. To check that the windows did not affect the results, runs were made using Polaroid HNCP 37 circular polarizers, where the polarizer in the sample beam was mounted in the Dewar before the crystal. The results were similar in the two cases. However these circular polarizers absorb strongly near 3900 Å and so were not used. The procedure we used was to reverse the field direction and not change the polarizers for the two runs, thus eliminating the problem of resetting the optics accurately.

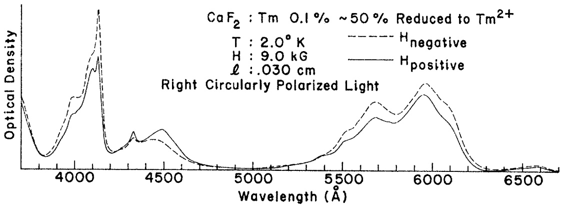

FIG. 2. Optical density curves for CaF_2:Tm^{2+} using right circularly polarized light and a magnetic field of 9 kG directed along the light path (*H* positive) and opposite to the light path (*H* negative).

A CaF_2 crystal doped with 0.1% thulium was used in which about 50% of the thulium was reduced (by the calcium baking technique[12]) to the divalent state as judged by spin resonance measurements and optical measurements on the sharp *f-f* transitions. Figure 2 shows the optical density curves for the absorption of right circularly polarized light directed along the field direction ($H+$) and against it ($H-$). [Optical density equals $0.434\,\alpha(\nu)l$.] The crystal was 0.30 mm thick, the temperature was 2.05±0.05°K, and the field was ±9.0 kG.

Using the expressions in Eqs. (3) and (4) we can find the absorption coefficients $\alpha_{\pm5/2}(\nu)$ from the data in Fig. 2 knowing T, H, and g. These are given in Fig. 3, in terms of the optical density. Since $D_{+5/2}(\nu)>D_{-5/2}(\nu)$ for most frequencies it is readily observed that the bands are primarily G in character and in fact in a number of regions the ratio

$$\frac{D_{+5/2}-D_{-5/2}}{D_{+5/2}+D_{-5/2}}$$

is greater than 0.4 and so approaches closely the maxi-

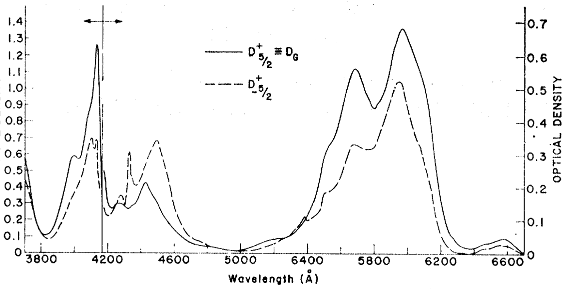

FIG. 3. Optical density curves for all the ions in the $\pm\frac{5}{2}$ magnetic sublevels of CaF_2:Tm^{2+} as deduced from Fig. 2 using Eqs. (3) and (4).

---

[12] Z. J. Kiss and P. N. Yocom, J. Chem. Phys. 41, 1511 (1964).

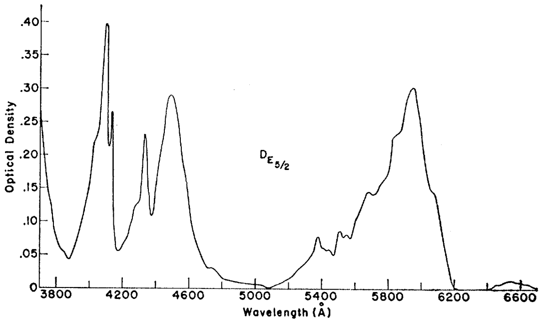

FIG. 4. Optical density curve for the $E_{5/2}$ part of the bands as deduced from Fig. 3 using Eq. (6).

mum of 0.5 for pure G character. This dominance might be expected since there are twice as many G levels as $E_{5/2}$ levels in the band. Assuming that the division of the G and $E_{5/2}$ character is valid, the curve $D_{+5/2}$ (ν) displays the G part of the bands directly, and $D_{-5/2}$ (ν) $-\frac{1}{3}D_{+5/2}$ (ν) as shown in Fig. 4 shows the $E_{5/2}$ part of the band. Inspection of these results gives the impression that the division is not perfect but good.

V. COMPARISON WITH THE FARADAY EFFECT MEASUREMENTS

The difference $D_{+5/2}$ $(\nu)-D_{-5/2}$ (ν) is plotted in Fig. 5 along with a replot of Shen's Faraday rotation measurements.[8] These are related through the Kramers-Kronig relationship

$$\theta(\nu)=\frac{l}{2\pi}P\int_0^\infty \frac{\nu'[\alpha_{5/2}(\nu')-\alpha_{-5/2}(\nu')]}{\nu'^2-\nu^2}d\nu'\left[\tanh\frac{g\beta H}{2kT}\right],$$

where P stands for the Cauchy principle value of the integral. Qualitatively the Faraday rotation curve does appear to follow as the dispersion of the circular dichroism. The indirect knowledge about the bands which the Faraday effect gives compared to the induced circular dichroism results suggests that the latter are more suitable for general investigations into the structure of such broad absorption bands.

VI. FIELD DEPENDENCE OF THE CIRCULAR DICHROISM

If a beam of unpolarized light falls on a crystal of CaF_2:Tm^{2+} directed along a magnetic field, the right and left circularly polarized components will be differentially absorbed. Assuming that the intensity of the light is low enough that it does not change the spin population in the sample, the percent of circular polarization in the beam when it leaves the crystal is given by

$$\rho=\tanh\{(l/2)[\alpha_{+5/2}(\nu)-\alpha_{-5/2}(\nu)]\tanh(g\beta H/2kT_S)\},$$

where T_S is now the effective temperature of the spins.

To detect the circularly polarized component of the light beam we used the experimental arrangement shown in Fig. 6. The beam of light to be analyzed is passed through a quarter wave plate rotating at ω and then a fixed linear "Polaroid sheet" HN-22 before being detected by a PbS detector or a photodiode. The output of the detector is then sent into a PAR lock-in detector tuned at 2 ω. It is easy to show that in such an arrangement the second harmonic detects only the circularly polarized component and the fourth harmonic can be used to detect the linearly polarized component.

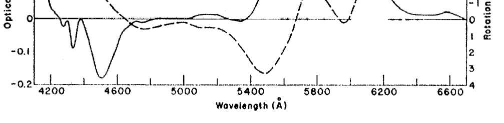

FIG. 5. Circular dichroism as found from Fig. 2 and the Faraday rotation as given by Shen. (Ref. 8).

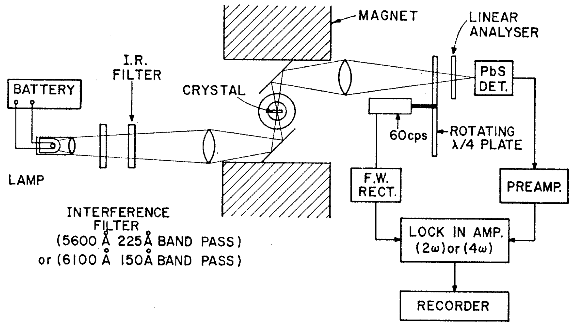

FIG. 6. Experimental arrangement to detect the circularly polarized component of a beam of light induced by passing through a CaF_2:Tm^{2+} crystal in a magnetic field.

Using this detector we measured the amount of circularly polarized light in the band 6025 to 6175 Å induced by being passed through a crystal 2 mm thick and at 4.2°K as a function of magnetic field. Since we operated in a region where the arguments of the hyperbolic tangents were small, ρ was found to be fairly linear with field as shown in Fig. 7.

This technique forms a rather convenient and sensitive way to monitor the spin temperature in this crystal. The Faraday effect has also been used for this purpose.[13]

VII. OPTICAL PUMPING OF THE THULIUM SPINS

The experimental arrangement for optically pumping the spins is shown in Fig. 8. The lamp was a 2.5 kW compact mercury arc. The net effect of the optics was to focus at most a flux of 10 W per cm^2 of visible light in the area of the crystal. The linear polarizer (Polaroid

---

[13] W. S. C. Chang and J. Q. Burgess, Appl. Optics **1**, 329 (1962). N. Bloembergen, P. S. Pershan, and L. R. Wilcox, Phys. Rev. **120**, 2014 (1960).

HN-38) was placed in the region where the beam was broad to eliminate extreme heating effects associated with the 60% absorption which takes place in it (leaving 4 W/cm^2 maximum flux at the crystal). The quarter wave plate was positioned just before the Dewar in such a way that it could be rotated or removed. Effects of the mirror inducing some circular polarization could be minimized by having the incident linear polarization in the plane of the incident ray.

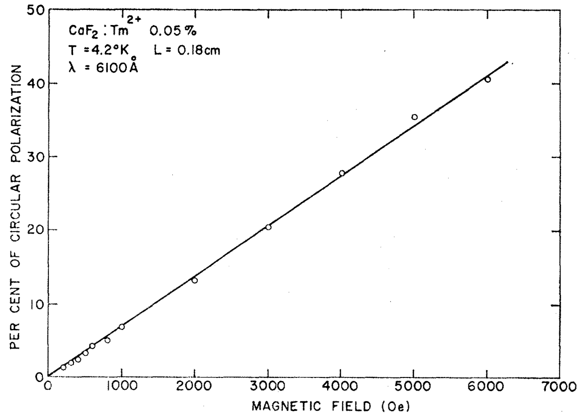

FIG. 7. Percent of circular polarization induced in a beam of light passing through a 0.18-cm crystal of 0.05% CaF$_2$:Tm^{2+} at 4.2°K in the direction of a magnetic field as a function of field strength.

With the light off we have an electron paramagnetic resonance (EPR) signal given by

$$I_0=(Nh\nu/2kT)$$

and a relaxation time to this equilibrium value of $\tau_0=1/2r$, where r is the relaxation rate for the spins to leave one state and go to the other. With the light shining on an optically thin crystal but containing no circular polarization component we have a different equilibrium value given by

$$S_0=\frac{N_i(rh\nu/kT)}{2r+p}+\frac{N_n h\nu}{2kT}=I_0\left[\frac{\tau N_i}{\tau_0 N}+\frac{N_n}{N}\right],$$

where N_i is the number of ions being illuminated and N_n is the number of those not illuminated because of some obstruction to the light. p is the effective optical pump rate for ions to go from one spin level to the other and $1/\tau=2r+p$ is the rate at which the EPR signal relaxes with the light on. If the light is right or left circularly polarized (±) the EPR signals are

$$S_\pm=N_i\frac{[(rh\nu/kT)\pm\Delta p]}{2r+p}+N_n\frac{h\nu}{2kT}$$

$$=I_0\left[\frac{\tau}{\tau_0}\left(1\pm\frac{kT}{h\nu}\frac{\Delta p}{r}\right)\frac{N_i}{r}+\frac{N_n}{N}\right],$$

where Δp is the effective differential pumping rate of the spins. If the initial beam is linearly polarized the addition of the quarter wave plate does not change $1/\tau=2r+p$, since the total pumping rate p is not changed.

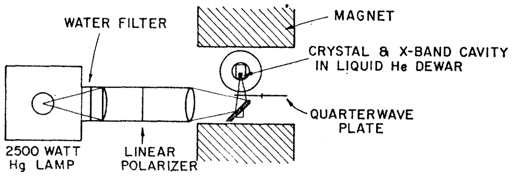

FIG. 8. Experimental arrangement for optically pumping the spins.

The first experiments were performed on a 0.01% CaF$_2$:Tm^{2+} crystal placed against the side wall of an X-band cavity operating in the TE$_{101}$ mode. To be able to optically pump the crystal, many slots were cut into the wall parallel to the current flow to affect the cavity Q as little as possible. We have previously used the same one to observe the optically excited state in CaF$_2$:Dy^{2+}.[14] Figure 9 shows a time display of the EPR signal. At one point the light was turned on (unpolarized) and the signal decayed to its new equilibrium value at a rate of $2r+p$, and then the light was turned off and the signal recovered to its initial value at the slower rate $2r$. From Fig. 9 one finds that the pump rate was 3.2 sec^{-1}, the relaxation rate was 0.62 sec^{-1}, and 50% of the crystal was effectively illuminated. This spin relaxation time is much longer than that measured by Huang,[15] but he used a higher concentrated CaF$_2$:Tm^{2+} sample.

Figure 10 shows similar plots for the case when the light is turned on: (a) linearly polarized, (b) right circularly polarized and directed along the field, and (c) left circularly polarized. From these results, we find that although the addition of the quarter wave plate could change the EPR signal (of the illuminated spins) by ±22%, the effective differential pumping rate was

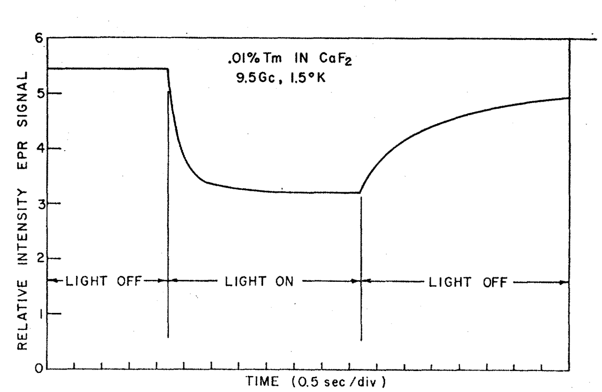

FIG. 9. Time dependence of EPR signal using unpolarized light.

---

[14] E. S. Sabisky and C. H. Anderson, Phys. Rev. Letters **13**, 754 (1964).

[15] Chao-Yuan Huang, Phys. Rev. **139**, A241 (1965).

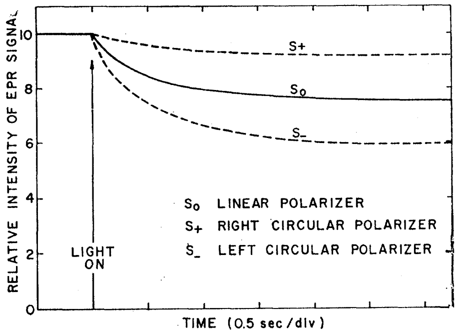

FIG. 10. Time dependence of EPR signal using polarized light. 0.01% Tm^{2+} in CaF_2, 9.5 Gc/sec, 1.5°K.

only 5% of the total pump rate. By using a set of band cutoff filters we were able to show that most of the effective pumping came from the 5770 and 5790 Å Hg lines as would be expected from the structure of the absorption bands in CaF_2. Figure 3 shows that we should expect a differential pumping efficiency of at least 15% for this wavelength, the measured efficiency is a factor of 3 lower for some unknown reason.

VIII. Tm^{2+} IN OTHER HOSTS

Since the strong 5770–5790 Å lines of mercury occur in a region where the difference between the absorption cross sections is somewhat less than average, it was thought that different hosts might shift the Tm^{2+} bands slightly into a more favorable region. In particular, in the crystals SrF_2 and BaF_2 the optical data of Kiss[16] indicate that the thulium bands (relative to those in CaF_2) shift about 100 and 250 Å, respectively, toward the blue in these hosts. This would put the 5460-Å Hg line in a very favorable position and move the 5770–5790-Å lines into a possible less efficient region and would probably have to be filtered out.

Crystals of SrF_2:Tm were obtained and reduced such that they gave a dark-green color. By optical pumping in an arrangement identical to that used for the CaF_2 experiments we achieved about the same effect. Later, we found that the green color was due to bands not identified with the thulium and, hence, hindred the pumping.

Some BaF_2:Tm crystals have also been obtained, but have so far not been satisfactorily reduced to the divalent state.

IX. DISCUSSION AND CONCLUSIONS

It should be noted that there is a distinct difference between the results for CaF_2:Tm^{2+} and the *F* center, in that divalent thulium has a large net differential absorption in the bands whereas the *F* center has an average effect of zero. We attribute this difference to the fact that the ground state of the *F* center is orbitally nondegenerate, whereas because of a large spin-orbit coupling and a relatively weak crystal field interaction, the ground state of CaF_2:Tm^{2+} has a large orbital moment.

Spin-orbit coupling is important in producing appreciable selective absorption because the electric dipole moment operator associated with the incident radiation field acts only on the orbital part of the wave functions, leaving the spin part unaffected. When the crystal field dominates over the spin-orbit coupling, the wave functions are, to first order, simple products of orbital and spin functions. Since the selection rule is then $\Delta S = \Delta m_s = 0$, there is no net spin change accompanying the absorption and thus no net change in the magnetization, which is primarily determined by the spins in this case. However, to second order the spin-orbit coupling can mix a small amount of orbital motion into the magnetic sublevels of the ground or excited states and thus lead to selective absorption. Physically we might say that the spin-orbit coupling provides a handle for the light to turn over the spins. The results for both ruby and the *F* center suggest that the bands are split and rigidly shifted by the spin-orbit coupling.[17] This is reflected in the *F* center work where the dichroism measurements are used to determine the magnitude of the spin-orbit parameter of the excited state.

When the spin-orbit coupling dominates as it does in the ground state of CaF_2:Tm^{2+} the Zeeman sublevels are no longer pure spin states but have appreciable orbital moments. Thus the magnetization can be changed quite directly by circularly polarized light. Conditions similar to those in CaF_2:Tm^{2+} also hold in most of the other paramagnetic divalent rare earth ions and so these ions should also show large selective absorption. Measurements on these are being made at present. The trivalent ions will also show similar effects but as previously mentioned, their strong 4*f* 5*d* bands occur primarily in the ultraviolet.

ACKNOWLEDGMENTS

We wish to thank D. S. McClure for some interesting discussions at the beginning of this work, and acknowledge the technical assistance of C. C. Neil.

---

[16] Z. J. Kiss (private communication).

[17] See Refs. 2, 3, 4, and also C. H. Henry, S. F. Schnatterly, and C. P. Slichter, Phys. Rev. 137, A583 (1965).