halides increases strongly upon decrease of the ratio S/D (where S is the space between adjacent halogen ions in a [110] direction of the normal lattice and D the diameter of the halogen atom). In going from KBr to NaBr, S/D diminishes by a factor of approximately two, while the energy needed to form an F center becomes 600 times greater. In the cesium halides, as one goes from CsCl to CsI, the corresponding S/D ratio decreases by a factor of 2.5. Consequently, one might expect a much lower colorability for CsI than for CsCl, at least at low temperatures.

ACKNOWLEDGMENTS

Microwave Modulation of Light in Paramagnetic Crystals*

N. BLOEMBERGEN, P. S. PERSHAN, AND L. R. WILCOX
Gordon McKay Laboratory, Harvard University, Cambridge, Massachusetts
(Received August 3, 1960)

The considerations of Dehmelt and several other workers about the modulation of light by radio-frequency signals in atomic vapors are extended to paramagnetic solids. It is shown that these materials, driven near a microwave resonance at low temperatures may be used both to create and to detect modulation of light at microwave frequencies. Experimental design criteria are discussed at the hand of two numerical examples, modulated circular dichroism in ruby and modulated Faraday rotation in a broad class of ionic rare earth compounds. Some possible applications of microwave modulated light are reviewed.

I. INTRODUCTION

THE modulation of light has a long history. As early as 1881 Righi[1] obtained fluctuations in an interference pattern by a periodic mechanical rotation of one in a pair of crossed Nicols. Rupp[2] produced unresolved side bands on light by operating a Kerr cell near 10^8 cps and detected their passage through a vapor cell absorbing only the central component. More recently Dehmelt[3] proposed that a precessing magnetization of atoms in alkali vapors should give a modulation of the absorption coefficient for circular polarized light propagating in the plane of the precessing component. Such a time dependent magnetization may be created by polarization of the alkali atoms (e.g., by optical pumping) and driving the resonance between two Zeeman levels with a radio-frequency magnetic field.

The successful outcome of such an experiment at 550 kc/sec on Na vapor has been reported by Bell and Bloom.[4] Bloom has announced a similar experiment on a higher frequency transition in K vapor. Series[5] has reported related experiments on modulation in Hg vapor where the precessing magnetization is induced in the excited state.

It is well known that large precessing components of magnetization can be produced by exciting the spin resonance in paramagnetic crystals at low temperatures. It is the purpose of this paper to investigate what happens to a circularly polarized light beam which passes through a crystal in which a precessing magnetization is established by an exciting microwave field.

Several optical experiments and their interplay with the spin resonances in ruby have recently been reported.[6-9] The analogy with corresponding experiments in alkali vapors has been pointed out. In paramagnetic crystals the polarization is established by thermal relaxation processes and need not be established by optical pumping. The characteristics of an experiment in ruby analogous to the Bell and Bloom experiment in Na vapor will be analyzed in Secs. II and III.

Kastler[10] has pointed out that the Faraday rotation in paramagnetic crystals could be decreased by saturation of the microwave resonance(s) in the ground-state multiplet. Opechowski[11] has given a detailed theory of the effect, which can be described simply by saying that microwave saturation changes the effective magnetic temperature. Daniels and Wesemeyer[12] have demonstrated the disappearance of optical Faraday rotation in

[1] A. Righi, J. phys. 2, 437 (1883).
[2] E. Rupp, Z. Physik 47, 72 (1927).
[3] H. G. Dehmelt, Phys. Rev. 105, 1924 (1957).
[4] W. E. Bell and A. L. Bloom, Phys. Rev. 107, 1559 (1957).
[5] G. W. Series, Proceeding of the Rochester Conference on Coherence, June, 1960 (unpublished).
[6] I. Wieder, Phys. Rev. Letters 3, 468 (1959).
[7] S. Geschwind, R. J. Collins, and A. L. Schawlow, Phys. Rev. Letters 3, 545 (1959).
[8] J. Brossel, S. Geschwind, and A. L. Schawlow, Phys. Rev. Letters 3, 548 (1959).
[9] T. H. Maiman, Phys. Rev. Letters 4, 564 (1960).
[10] A. Kastler, Compt. rend. 232, 953 (1951).
[11] W. Opechowski, Revs. Modern Phys. 25, 264 (1953).
[12] J. M. Daniels and H. Wesemeyer Can. J. Phys. 36, 405 (1958).

neodymium ethyl sulphate when the transition between the two Zeeman levels in the ground-state Kramers doublet of Nd^{+++} is saturated.

A variation of this experiment, in which the direction of the incident light is at right angles to the dc magnetic field, but parallel to the microwave magnetic field is discussed in Sec. IV. An observable microwave modulation of the plane of polarization would result.

II. THEORY OF MODULATED CIRCULAR DICHROISM IN RUBY

Consider the geometry of Fig. 1(a) in which circular polarized light is incident along the z axis, parallel to the c axis of the crystal. A homogeneous magnetic field is applied at right angles, parallel to the x axis. The crystal is placed in a microwave cavity with a strong magnetic radio-frequency component in the z direction. The incident light is assumed to contain black-body radiation in a frequency interval, which covers completely the structure of the R_1 line alone, or the structure of the R_2 line alone. The incident light will be treated classically and will be considered as a small perturbation. The Hamiltonian of the problem is

$$\mathcal{3C}=\mathcal{3C}_{cr}+\mathcal{3C}_0+\mathcal{3C}_{rf}+V_L,$$

where $\mathcal{3C}_{cr}$ is the crystalline field potential including the spin orbit coupling. It determines the splittings between the quartet ground state $^4A_2$ and the doublet states $\bar{E}(^2E)$ and $2\bar{A}(^2E)$. The notation of Sugano and
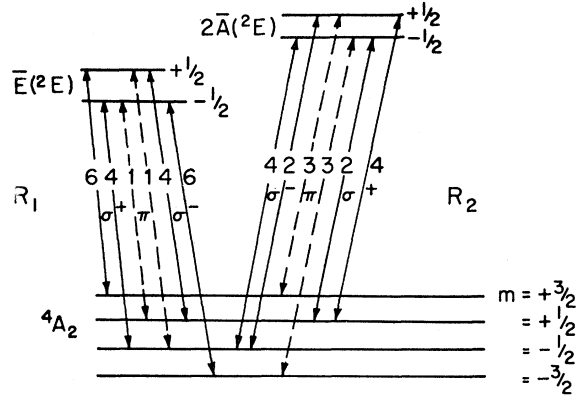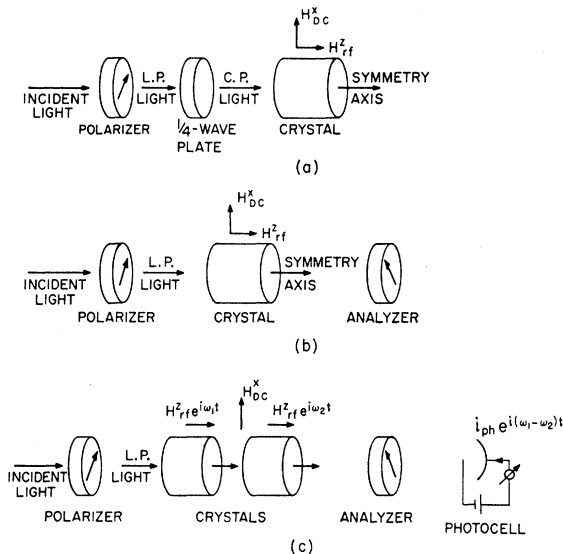
FIG. 1(a). Experimental geometry for microwave modulation of magnetic circular dichroism of light. (b). Microwave modulation of light by optical Faraday rotation of the plane of polarization or by circular dichroism. The light passes through crossed polarizer and analyzer, between which a crystal with a radiofrequency component of magnetization is inserted. (c). Heterodyne detection of microwave modulation of light. The photocurrent is modulated at an intermediate beat frequency.

FIG. 2. Relative transition probabilities and polarizations of the R lines in ruby according to Sugano and Tanabe.[13] The optic axis is chosen as the axis of quantization.

Tanabe[13-15] has been adopted. In this representation the crystal field is diagonal. The spin quartet will be labeled by the magnetic quantum number along the z-axis $m=+\frac{3}{2}, +\frac{1}{2}, -\frac{1}{2},$ and $-\frac{3}{2}$. In the presence of a strong magnetic field in the x direction these states are of course not proper states of the system. The Zeeman energy $\mathcal{3C}_0$ is not diagonal in this representation.

The set of states in which $\mathcal{3C}_{cr}+\mathcal{3C}_0$ is diagonal will be denoted by μ, and can be obtained from the m states by a unitary transformation U with matrix elements defined by

$$\psi_\mu=\sum_m U_{m\mu}\psi_m. \quad (1)$$

The excited states of two Kramers doublets are denoted by $2\bar{A}^\pm$ and $\bar{E}^\pm$, where $+$ and $-$ indicate the quantization with respect to the z axis. Again $\mathcal{3C}_0$ is not diagonal in this representation. The Zeeman splitting of the excited doublets will in general be different from the ground-state splittings. The perpendicular spectroscopic splitting factor of the excited states is small. It will henceforth be assumed that no two splittings are equal. A microwave field H_{rf} in the z direction will therefore be near resonance for only one transition. Its frequency will be chosen close to a resonant frequency $\omega_{\mu\mu'}$ in the ground-state quartet.

Sugano and Tanabe[13] have calculated the matrix elements of the light operator in the R_1 and R_2 doublet for three modes of polarization; σ^+ is right circular, σ^- is left circular and π is polarization parallel to the z axis. The matrix elements are simplest in the m representation with quantization along the z axis. The squares of the nonvanishing σ components are reproduced in Fig. 2.

Although all matrix elements of $\mathcal{3C}$ in the 8×8 representation can be written down explicitly, a com-

[13] S. Sugano and Y. Tanabe, J. Phys. Soc. (Japan) 13, 880 (1958).
[14] S. Sugano and Y. Tsujikawa, J. Phys. Soc. (Japan) 13, 899 (1958).
[15] Y. Tanabe and H. Kamimura, J. Phys. Soc. (Japan) 13, 394 (1958).

plete solution of the equation of motion for the density matrix,[16]

$$i\hbar\dot{\rho} = (\mathfrak{K}\rho - \rho\mathfrak{K}), \qquad (2)$$

is not feasible in the present case. Complete solutions for the case of a free atom under the simultaneous influence of a strong radio-frequency field and optical resonance radiation have been described by Cagnac, Barrat[17] and others.[18] The presence of the crystalline field and spin-orbit coupling, inhomogeneous broadening and relaxation mechanisms dictate the use of a perturbation procedure.

Consider the 4×4 submatrix $\rho_{\mu\mu'}(\mu, \mu'=1, 2, 3, 4)$ of the ground state quartet in the μ representation in which $\mathfrak{K}_{cr}+\mathfrak{K}_0$ is diagonal. A microwave field of saturating strength, but smaller than the line width, is applied near the resonant frequency of the transition $\mu=1 \rightarrow 2$.

The diagonal elements of the density matrix can be obtained from the usual relaxation rate equations for the populations with $\rho_{11}=\rho_{22}$. The problem is then reduced to a two-level spin resonance with total population $\rho_{11}+\rho_{22}$. The solution for the two by two density matrix is equivalent to the classical solution of Bloch for a precessing magnetization.[16] The off-diagonal element ρ_{12} precessing in phase with the driving microwave field can therefore be determined from the theory of Redfield.[19] The dispersive component of the magnetization does not saturate, if H_{rf} is smaller than the linewidth. The value of the real part of the susceptibility is essentially the same as the unsaturated value. Redfield gives more details (compare Fig. 6 of his paper), but for our purposes the above assumption is adequate. It is particularly good, if most of the magnetic broadening is due to unlike spins (Al^{27} nuclei). For inhomogeneous broadening, such as may be caused by a distribution of crystalline field parameters, Portis[20] arrived earlier at the same conclusions for the dispersive part of the magnetization.

Redfield's assumption about the relaxation in the precessing field and return to thermal equilibrium will be taken over, even though the presence of other levels complicates the relaxation mechanism. This is permissible because his results for the dispersive component do not depend critically on details of spin-lattice relaxation.

With microwave field $H_{rf}\cos\omega t$ in the z direction, the expectation value of the z component of the precessing magnetization is

$$\langle M_z \rangle = |M_{12}{}^z|^2\hbar^{-1}N(\rho_{11}{}^0-\rho_{22}{}^0)$$
$$\times (\omega-\omega_{12})T_2{}^2\{1+(\omega-\omega_{12})^2T_2{}^2\}^{-1}H_{rf}\cos\omega t, \quad (3)$$

for

$$\gamma^{-1}(T_1T_2)^{-\frac{1}{2}}\ll H_{rf}(\gamma T_2)^{-1}. \qquad (4)$$

The difference in populations per unit volume between levels 1 and 2 in thermal equilibrium is given by

$N(\rho_{11}{}^0-\rho_{22}{}^0)$, where N is the total number of ions per unit volume. The coefficient of $H_{rf}\cos\omega t$ is equal to the Bloch expression for the unsaturated real part of the susceptibility in a linearly polarized field.[21] There is no out-of-phase component in Eq. (3), since the absorptive component is completely saturated in the regime of Eq. (4). The values of the off-diagonal matrix elements of the density matrix and magnetic moment operator are closely related. From the expression

$$\langle M_z \rangle = (\rho_{12}M_{21}{}^z + \rho_{21}M_{12}{}^z)N,$$

and Eq. (3) it can be inferred that

$$\rho_{12} = \tfrac{1}{2}M_{12}{}^z\hbar^{-1}(\omega-\omega_{12})T_2{}^2\{1+(\omega-\omega_{12})^2T_2{}^2\}^{-1}$$
$$\times(\rho_{11}{}^0-\rho_{22}{}^0)H_{rf}\exp(i\omega t). \quad (5)$$

In the Heisenberg representation $M_{12}{}^z$ and ρ_{12} have to be multiplied by the time factor $\exp(-\omega_{12}t)$.

The number of light quanta absorbed is equal to the transitions out of the ground state quartet caused by the light perturbation. Note that all terms in the Hamiltonian, except the incident light field, leave the partial trace over the ground state quartet invariant. Consider a time interval $t-t_0\ll\omega^{-1}$. In this interval the variation of the density matrix due to the presence of a microwave driving field at the frequency ω can be ignored. In such a short time interval the perturbation by light can be treated independently from the perturbation by the microwave field. The solution of the equation of motion,

$$i\hbar(\partial/\partial t)_{light}\rho = V\rho - \rho V,$$

is obtained by standard perturbation theory,

$$\rho_{\mu\mu}(t) = \left\{ \exp\left[-i\hbar^{-1}\int_{t_0}^t V(t')dt'\right] \right.$$
$$\left. \times\rho(t_0)\exp\left[+i\hbar^{-1}\int_{t_0}^t V(t'')dt''\right] \right\}_{\mu\mu}$$

$$= \rho(t_0) - i\hbar^{-1}\int_{t_0}^t dt'[V_{\mu\alpha}(t')\rho_{\alpha\mu}(t_0) - \rho_{\mu\alpha}(t_0)V_{\alpha\mu}(t')]$$

$$+ \hbar^{-2}\int_{t_0}^t\int_{t_0}^t dt'dt''\{V_{\mu\alpha}(t')\rho_{\alpha\beta}(t_0)V_{\beta\mu}(t'')\}$$

$$- \tfrac{1}{2}\hbar^{-2}\int_{t_0}^t\int_{t_0}^t dt'dt''\{V_{\mu\alpha}(t')V_{\alpha\mu'}(t'')\rho_{\mu'\mu}(t_0)$$
$$+ \rho_{\mu\mu'}(t_0)V_{\mu'\alpha}(t')V_{\alpha\mu}(t'')\}. \quad (6)$$

It is understood that summations over all excited states α and β, and over μ' are carried out. The initial time t_0 should be considered as a variable parameter. Since at t_0 all elements of the density matrix connecting with an excited level vanish, only the contribution of the last term will have to be considered. This statement is

[16] U. Fano, Revs. Modern Phys. 29, 74 (1957).
[17] B. Cagnac and J. P. Barrat, Compt. rend. 249, 534 (1959).
[18] J. P. Barrat, J. phys. radium 20, 541 and 633 (1959).
[19] A. G. Redfield, Phys. Rev. 98, 1787 (1955).
[20] A. M. Portis, Phys. Rev. 91, 1071 (1953).

[21] F. Bloch, Phys. Rev. 70, 460 (1946).

equivalent to saying that the intensity of the incident light is not capable of producing a noticeable pumping effect. The light absorption is proportional to

$$\sum_\mu \{\rho_{\mu\mu}(t) - \rho_{\mu\mu}(t_0)\}$$

$$= -\hbar^{-2} \sum_{\mu,\mu',\alpha} \int_{t_0}^t \int_{t_0}^t dt' dt'' V_{\mu\alpha}(t') V_{\alpha\mu'}(t'') \rho_{\mu'\mu}(t_0). \quad (7)$$

If the usual assumption is made that the vector potential is constant over the volume of one ion, the matrix elements of the light perturbation can be written explicitly in the Heisenberg representation of $\mathfrak{3C}_0 + \mathfrak{3C}_{cr}$,

$$V_{\mu\alpha}(t) = (e/mc)\mathbf{p}_{m\alpha} \cdot [\mathbf{A}(t) \exp(i\langle\omega_R\rangle t)$$
$$+ \mathbf{A}^*(t) \exp(-i\langle\omega_R\rangle t)] \exp(-i\omega_{\mu\alpha}t). \quad (8)$$

Here $\mathbf{p}$ is the momentum operator in the Schrödinger representation. The light wave is represented by a narrow band Gaussian random process, centered at an average absorption frequency $\langle\omega_R\rangle$ in the R_1 or R_2 line structure. The complex amplitude of the vector potential is a random function of time. Its character is described by a set of correlation functions $\langle A_i^*(t') A_j(t'') \rangle = \langle A_i^*(0) A_j(\tau) \rangle$, where i and j denote components of polarization. The correlation function becomes negligibly small for $\tau = t'' - t' > \tau_c = \Delta\omega^{-1}$, where τ_c is the correlation time and $\Delta\omega$, the bandwidth of the light. The interval of integration is chosen so that $\tau_c < t - t_0 < \omega^{-1}$. This is possible if the bandwidth of the light is large compared to the microwave frequency, i.e., if the light covers the entire structure of the absorption line.

When Eq. (8) is substituted into Eq. (7), one should also add the explicit Heisenberg time factor to $\rho_{\mu\mu'}(t_0)$. Introduction of new variables of integration $T = t' + t''$ and $\tau = t' - t''$ leads to a light absorption proportional to the time,

$$\sum_\mu [\rho_{\mu\mu}(t) - \rho_{\mu\mu}(t_0)]$$

$$= -(e^2/\hbar^2 m^2 c^2) \sum_{\mu,\mu',\alpha} \sum_{i,j^*} p_i(\mu\alpha) p_j(\alpha\mu') \rho_{\mu\mu'}(t_0)$$

$$\times \exp(-i\omega_{\mu\mu'}t) \frac{1}{2} \int_{2t_0}^{2t} \exp\{i(\omega_{\mu\alpha} + \omega_{\alpha\mu'})T\} dT$$

$$\times \int_{-\infty}^\infty \langle A_i^*(0) A_j(\tau) \rangle \exp\{i(\langle\omega_R\rangle - \tfrac{1}{2}\omega_{\mu\alpha} - \tfrac{1}{2}\omega_{\mu'\alpha})\tau\} d\tau.$$

The exponential function in the last integral can be put equal to unity, because the correlation function approaches zero before the exponent becomes appreciable, $\{\langle\omega_R\rangle - \tfrac{1}{2}(\omega_{\mu'\alpha} + \omega_{\mu'\alpha})\}\tau_c \ll 1$.

The next to last integral becomes simply,

$$\frac{1}{2} \int_{2t_0}^{2t} \exp\{i(\omega_{\mu\alpha} + \omega_{\alpha\mu'})T\} dT = (t - t_0) \exp(i\omega_{\mu\mu'}t_0),$$

since the time interval $t - t_0$ can be chosen small com-

pared to any microwave period, $(t - t_0)\omega_{\mu\mu'} \ll 1$. In this manner one obtains a rate of light absorption proportional to

$$\sum_\mu \dot\rho_{\mu\mu}(t_0) = -(e^2/\hbar^2 m^2 c^2) \sum_{\mu,\mu',\alpha} \sum_{i,j} p_i(\mu\alpha) p_j(\alpha\mu') \rho_{\mu\mu'}(t_0)$$

$$\times \int_{-\infty}^{+\infty} \langle A_i^*(0) A_j(\tau) \rangle d\tau. \quad (9)$$

The last integral is proportional to the intensity of the incident light $\mathcal{J}(\langle\omega_R\rangle)$, which has been assumed constant over the absorption band. The absorption rate derives an explicit time dependence from $\rho_{\mu\mu'}(t_0)$ which is given by Eq. (5) when $\mu = 1$, $\mu' = 2$. The light absorption rate is modulated at the microwave driving frequency ω. All Heisenberg time factors have canceled each other.

The expression (9) is invariant under a change in representation. Since the matrix elements for the momentum, or electric dipole moments, are given by Sugano and Tanabe[13] for a representation quantized along the c axis, it is convenient to make the transformation from the ground-state designation of $|\mu\rangle$ and the excited state, of $|\alpha\rangle$ to the new representation where the ground state is $|m\rangle$ and the excited state is $|a\rangle$. Equation (9) is to be rewritten as

$$\sum_\mu \dot\rho_{\mu\mu}(t_0) = \sum_m \dot\rho_{mm}(t_0)$$

$$= -(e^2/\hbar^2 m^2 c^2) \sum_{m,m',a} \sum_{i,j} p_i(ma) p_j(am') \rho_{mm'}(t_0)$$

$$\times \int_{-\infty}^\infty \langle A_i^*(0) A_j(\tau) \rangle d\tau. \quad (10)$$

The elements

$$\rho_{mm'} = \sum_{\mu,\mu'} \rho_{\mu\mu'} U_{m\mu}^* U_{m'\mu'} \quad (11)$$

derive an explicit time dependence $\exp(i\omega t)$ from $\rho_{\mu\mu'}(t_0)$, if the microwave driving field is applied at a frequency ω near $\omega_{\mu\mu'}$. The transformation matrix U has been tabulated[22] for various magnitudes and orientations of the dc magnetic field. The interest here is primarily in the case that the applied field is at right angles to the c axis and that the Zeeman splitting is large compared to the zero-field splitting. In this case the transformation matrix corresponds to the four-dimensional $(S = \tfrac{3}{2})$ representation of a rotation by 90° around the y axis,

$$U_{m\mu} = D_{m\mu}^{\tfrac{3}{2}}(0, \pi/2, 0). \quad (12)$$

Rose[23] has given a clear account of the Wigner coefficients and his notation is followed here.

A similar argument can still be given if the incident light were strictly monochromatic, but the excited final states α of the ion were diffuse. In this case all transitions

[22] See, e.g., J. Weber, Revs. Modern Phys. **31**, 681 (1959), Appendix.
[23] M. E. Rose, *Elementary Theory of Angular Momentum* (John Wiley & Sons, Inc., New York, 1959), Chap. 4.

$\mu \to \alpha$ would still be excited. In ruby, however, not only the ground levels m of ^4A, but also the excited doublets (^2E) are extremely sharp. If the incident light could only excite the transition from one state $\mu \to \alpha$, then the light absorption would be proportional to $\rho_{\mu\mu}$, which is independent of time. For such narrow line excitation the preceding argument breaks down, because in a suitable time interval of integration $t - t_0 > \tau_c > \omega^{-1}$ the variation of matrix elements $\rho_{mm'}$ with the microwave precession would not be negligible.

III. EXPERIMENTAL DESIGN CONSIDERATIONS AND NUMERICAL EXAMPLE

Consider the specific case with left circular polarized light incident along the c axis, covering all transitions $\mu \to \alpha$ in the R_2 structure. The light Hamiltonian for this polarization is given by Eq. (8) with $\mathbf{A} = \mathbf{A}_0(\hat{x} + i\hat{y})$ where $\hat{x}$ and $\hat{y}$ are unit vectors in the x and y axis, respectively. The only nonvanishing matrix elements according to Fig. 2 occur for $m = m' = -\frac{1}{2}$, $(a = 2A^+ | p_+ | m = -\frac{1}{2})$ $= (m = -\frac{1}{2} | p_- | a = 2A^+) = \sqrt{2}(a = 2A^- | p_+ | m = -\frac{1}{2})$ $= \sqrt{2}(m = -\frac{1}{2} | p_- | a = 2A^-)$, where the notation $p_+ = p_x + ip_y$ and $p_- = p_x - ip_y$ has been introduced. The absorption coefficient is proportional to $\rho_{-\frac{1}{2},-\frac{1}{2}}$, which is modulated at the microwave frequency ω according to Eqs. (5), (10), and (11). Left circular polarized light in the R_1 lines can make transitions from the $m = -\frac{3}{2}$ and $m = +\frac{1}{2}$ level with relative strengths 3:2. The absorption coefficient is proportional to $3\rho_{-\frac{3}{2},-\frac{3}{2}} + 2\rho_{\frac{1}{2},\frac{1}{2}}$. In this case the relative phase of the time variation in these elements is important. Similar consideration can be given for right circular polarized light.

Consider a specific numerical example. A pink ruby crystal at 1.6°K is placed in a dc magnetic field of about 10 500 oersteds perpendicular to the c axis in the x direction, so that the microwave resonance frequency between the two lowest levels $\mu = -\frac{3}{2} \to -\frac{1}{2}$ occurs at 24 kMc/sec. Since the Zeeman energy is large compared to the zero field splitting, the magnetic quantum number μ along the x axis is nearly perfect and the transformation to the m representation is very nearly that given by Eq. (12). The ruby is placed with the c axis parallel to the axis in a right-cylindrical cavity, driven in the TE_{011} (wave meter) mode. The microwave magnetic field is predominantly along the c axis and is assumed to have an amplitude $H_{rf} = 1$ oersted. With a cavity volume of about 1 cc and an unloaded $Q_0 = 30\,000$, this corresponds to a power dissipation in the walls of 10 milliwatts. This power consumption can easily be supplied by standard klystroms and can be conveniently dissipated by liquid helium at 1.6°K. It corresponds to a pump speed of 0.35 l/sec at the cryostat. The estimate for Q_0 is considered conservative for a pure copper cavity at low temperature, where the resistivity is mainly determined by the magnetoresistance and the anomalous skin effect.

The power dissipation by the magnetic ions can always be made negligible compared to the wall losses, if a sufficiently high degree of saturation is reached. The

maximum power transferred by the spins is determined by their spin-lattice relaxation time and is roughly $(\Delta N_0)T_1^{-1}h\nu$. With $T_1 = 10^{-2}$ sec and $\Delta N_0 = 10^{18}$ (see below) in a dilute ruby sample (0.02% Cr) of 0.2 cc volume one obtains 0.7 milliwatts. It is clear from these considerations that a long spin-lattice relaxation time and a very high quality factor are favorable in an experiment to obtain the largest possible precessing component of magnetization for a given power consumption. A dilute pink specimen is chosen because it has the narrowest microwave line width of about $T_2^{-1} = 1.8 \times 10^8$ sec^{-1} according to Strandberg ($\Delta\nu = 60$ Mc/sec between points of half maximum absorption). The optical density (opacity) at the R lines is still sufficiently large for such a specimen. The optical absorption coefficient[24] is about 5 cm^{-1} for the population of the levels occurring in our example.

Since the condition (4) is satisfied by our choice of parameters, Eq. (5) may be applied. Since $\exp(-h\nu/kT) = 0.5$ with our choice of temperature and frequency, the equilibrium populations are $\rho_{11}^0 = 0.54$, $\rho_{22}^0 = 0.27$, $\rho_{33}^0 = 0.13$, $\rho_{44}^0 = 0.06$. Advantage of the Boltzmann factor is taken if the resonance between the two lowest levels is saturated, $\rho_{11}^0 - \rho_{22}^0 = 0.27$. The matrix element $(1 | M_z | 2)$ between the $\mu = -\frac{3}{2}$ and $\mu' = -\frac{1}{2}$ state is $g_\| \beta \frac{1}{2} \sqrt{3}$. Note that $g\beta h^{-1} H_{rf} T_2$ is just the ratio between the half linewidth at half maximum and the microwave amplitude, both measured in gauss. This ratio is 0.1 in this numerical example. The field or frequency is, of course, adjusted off resonance to maximize the dispersive component; $(\omega - \omega_{12})T_2 = 1$. One finds $\rho_{2i} = \rho_{12} = \frac{1}{2}\sqrt{3}\, 0.1 \times 0.26 \exp(i\omega t)$. The diagonal elements depend somewhat on the details of the relaxation mechanism but a reasonable choice for the saturated state is $\rho_{11} = \rho_{22} = 0.4$, $\rho_{33} = 0.14$, $\rho_{44} = 0.06$. Equations (11) and (12) then yield

$$\rho_{m,m=-\frac{1}{2},-\frac{1}{2}}(t_0) = \frac{1}{8}\sqrt{3}\{\rho_{12}(t_0) + \rho_{21}(t_0)\}$$
$$+ \frac{3}{8}\rho_{11} + \frac{1}{8}\rho_{22} + \frac{1}{8}\rho_{33} + \frac{3}{8}\rho_{44}, \quad (13)$$

$$\rho_{-\frac{1}{2},-\frac{1}{2}}(t) = 0.24(1 + 0.01 \cos\omega t).$$

The modulation depth is one percent. In the same manner one finds that the absorption coefficient for the right circular polarized R_2 line is proportional to

$$\rho_{+\frac{1}{2},\frac{1}{2}}(t) = 0.24(1 - 0.01 \cos\omega t).$$

It turns out that the case of circularly polarized R_1 light is less favorable, because the modulation of the $m = -\frac{3}{2}$ and $m = +\frac{1}{2}$ levels have opposite phase. The modulation depth under the same conditions is only 0.2% in this case. Similarly, driving of the $\mu = -\frac{1}{2} \to +\frac{1}{2}$ transition leads to smaller modulation depths of 0.33% for the R_2 lines; but to a larger depth of 1.33% for the R_1 line. This larger value is obtained in spite of the fact that the population $\rho_{33}^0 - \rho_{22}^0$ is only half of $\rho_{22}^0 - \rho_{11}^0$. It

[24] F. Varsanyi, D. L. Wood and A. L. Schawlow, Phys. Rev. Letters 3, 544 (1959).

has its origin in the constructive interference of the modulation of $\rho_{-\frac{3}{2},-\frac{3}{2}}$ and $\rho_{+\frac{1}{2},+\frac{1}{2}}$ which are now in phase.

The intensity in a circularly polarized light beam is related to the amplitude of the vector potential A_0 by

$$(\mathcal{S}_R) = (2\pi \nu_L{}^2/c)\int_{-\infty}^{\infty} \langle A_0(0)A_0(\tau)\rangle d\tau. \qquad (14)$$

The absorbed light energy/cm² sec from a beam of left circular polarized R_2 light in a segment dz is equal to the decrease in beam intensity,

$$+d\mathcal{S}=Nh\nu_L\rho_{-\frac{3}{2},-\frac{3}{2}}^0 dz. \qquad (15)$$

With the use of Eqs. (10) and (13) this becomes

$$d\mathcal{S}=-\frac{|p_+|^2 e^2}{\nu_L \hbar c m^2}N\mathcal{S}\langle\nu_R\rangle 0.24(1+0.01\,\cos\omega t)dz. \qquad (16)$$

Here $e^2|p_+|^2/4\pi^2\nu_L{}^2 m^2=e^2|x+iy|^2$ is the square of the electric dipole moment operator connecting the $m=-\frac{1}{2}$ ground level with the $2A^+$ level.

It is not immediately possible to integrate Eq. (16) for large thickness z, because the spectral quality of the incident white light will change after traversing a finite thickness. In a dilute ruby with well-resolved Zeeman components a rather complex situation will result. In a darker ruby, with sufficient overlap of components the expression $N\mathcal{S}(\nu)$ may be replaced by $N\mathcal{S}g(\nu)$, where $\mathcal{S}$ is now the total light intensity and $g(\nu)$ is a normalized shape function, describing the combined spectral response of all ions. This distribution does not change with the distance z. Cross relaxation processes in the spin system will keep it constant. An absorption coefficient β for incident monochromatic light at frequency ν can then be defined,

$$\beta=-(\partial\mathcal{S}/\partial z)/\mathcal{S}$$
$$=4\pi^2\nu_L\hbar^{-1}c^{-1}e^2|x+iy|^2g(\nu)N\rho_{-\frac{1}{2}-\frac{1}{2}}(t). \qquad (17)$$

The modulation depth s is introduced by

$$\rho_{-\frac{1}{2},-\frac{1}{2}}=C(1+s\,\cos\omega t), \quad \beta=\beta_0(1+s\,\cos\omega t). \qquad (18)$$

The transmitted intensity after a thickness z is modulated according to

$$\mathcal{S}=\mathcal{S}_0\exp(-\beta_0 z)(1-s\beta_0 z\,\cos\omega t), \qquad (19)$$

valid for $s\beta_0 z\ll1$. The oscillating part is maximum for $\beta_0 z=1$ and then the modulation depth of the intensity is also s, valid for $s\ll1$. In practice the modulation depth will be considerably smaller, because the incident light will contain wavelengths for which $\beta_0 z\ll1$.

The detection of light modulation has thus far been based on the photoelectric effect which is proportional to the light intensity. For frequencies higher than 10^8 cps the photocathode has to become an integral part of the microwave circuitry. Forrester[25] and co-workers

[25] A. T. Forrester, R. A. Gudmundsen, and P. O. Johnson, Phys. Rev. **99**, 1691 (1955).

have established that the variation in the emitted photocurrent can follow light fluctuations at 10 kMc/sec. It is probable that the photocurrent from a cathode forming the wall of a suitably constructed K-band cavity could re-excite microwave power. It would, however, require considerable development and it would involve the simultaneous testing of two effects, microwave modulation of magnetic circular dichroism and of the photoelectric emission at K-band frequencies.

It is therefore proposed here to utilize the same effect for generation and detection of light modulation. Since the modulation depth is so small, it is also advantageous to use a light bridge scheme to avoid the shot effect of the unmodulated photon current. Consider therefore the case of R_2 light, linearly polarized perpendicular to the c axis as shown in Fig. 1(b). Decompose into a right and left circular wave. These waves will propagate according to $\exp(2\pi i n_L z/\lambda)$ and $\exp(2\pi i n_R z/\lambda)$, where λ is the vacuum wavelength and the complex index of refraction is

$$n_L=n_0+i(\lambda/2\pi)(\beta_0/2)(1-s\,\cos\omega t),$$
$$n_R=n_0+i(\lambda/2\pi)(\beta_0/2)(1+s\,\cos\omega t). \qquad (20)$$

After having traversed a distance z, the light will be elliptically polarized and the linear component at right angles to the original incident polarization will have an amplitude

$$|A_L|-|A_R|=2A_0\sinh(\tfrac{1}{2}\beta_0 zs\,\cos\omega t)\exp(-\tfrac{1}{2}\beta_0 z).$$

The intensity observed through a crossed polarizer and analyzer, with the dichroic ruby in between, will be, for $\beta_0 zs\ll1$,

$$\mathcal{S}_\perp=\mathcal{S}_0\exp(-\beta_0 z)(\beta_0 zs)^2\tfrac{1}{2}(1+\cos2\omega t). \qquad (21)$$

The signal is a maximum for $\beta_0 z=2$, $s\ll1$. It can be considered as the output of an autodyne or square law rectifier. This becomes evident if one considers two ruby crystals in the same orientation, but driven with different phases, as shown in Fig. 1(c). The factor $\tfrac{1}{2}(1+\cos2\omega t)$ in Eq. (21) is then replaced by $\cos\phi_1\cos\phi_2$.

The difference in phase may be created by passing the light beam twice through the same crystal with the aid of mirrors. In this way the time interval between the two light passages is measured, which is a basic observation in the determination of the velocity of light.

If the phases are different because two different microwave driving frequencies are used, $\phi_1=\omega_1 t$ and $\phi_2=\omega_2 t$, an intermediate frequency beat at $\omega_2-\omega_1$ in the photoelectric current will result, which is detectable with a conventional photomultiplier. This signal would be a more specific indication of the contemplated effect from an experimental point of view than the dc signal given by Eq. (21). In either case further differentiation against spurious effects can be obtained by audiofrequency modulation of the microwave power, frequency, or the dc magnetic field H_0. The signal is admittedly weak, since Eq. (21) for $\beta_0 z=2$ gives

$$\mathcal{S}_\perp=0.27\,\mathcal{S}_0 s^2,$$

where $\mathcal{I}_0$ is the intensity incident on the crystal in the frequency interval of the absorption line and the modulation depth $s \sim 10^{-2}$. Nevertheless the back-ground light in the crossed analyzer can be kept with some care at the same low level, and the modulation depth is of the order of unity. It then becomes a matter of detecting a beam of intensity which is a few parts in 10^5 of the incident beam. This is perfectly feasible. If necessary, the dark current in the photocell can be reduced by cooling to eliminate its associated shot noise.[26] The remaining fluctuations are then essentially those inherent in the statistics of counting individual photons.

If intermittent operation is used by applying, e.g., a 90° pulse to the microwave spin resonance, much larger values of s can be obtained. The equations given in this section are of course equally valid for this case. During the pulse and in the free precession immediately following the microwave signal, the light signal will be more than two orders of magnitude larger than assumed above.

A variation of these experiments is obtained when the light source itself is modulated. This can be done in ruby by magnetic resonance in the excited state. The fluorescent ruby light source should have a higher Cr concentration so that the emitted light spectrum is broadened sufficiently to cover the entire absorption spectrum of a second ruby crystal.

The magnetic resonance condition in these paramagnetic crystals at very low temperature should be corrected for shape demagnetization effects in the same manner as is customary for ferromagnetic materials.

IV. MODULATION OF FARADAY ROTATION IN CRYSTALS WITH RARE EARTH IONS

The dispersive companion effect to circular dichroism is Faraday rotation of the plane of polarization of linear polarized light. Condon[27] has reviewed this relationship for optically active molecules, but it holds equally well for paramagnetic rotation. ϕ and ψ denote two vectors in the direction of the incident light. The magnitude of ϕ corresponds to the angle of rotation per unit length and the magnitude of ψ corresponds to the ellipticity per unit length, acquired by incident linear polarized light,

$$\phi + i\psi = 8\pi^2 \nu_L^2 N h^{-1} e^{-1} \epsilon^{-\frac{1}{2}}$$
$$\times \sum_{fg} \frac{i\mathbf{p}_{fg} \times \mathbf{p}_{gf}}{\nu_L^2 - \nu_{gf}^2 + 2\pi i \nu_L \Gamma_{gf}} \rho_{ff}. \quad (22)$$

Here $\mathbf{p}_{gf}$ is the matrix element of the electric dipole moment operator between a state f, occupied with a probability ρ_{ff} and an optically excited state g, separated by an energy interval $h\nu_{gf}$. The formula is valid for substances which are optically isotropic with dielectric constant ϵ in the absence of a magnetic field, if the rotation per wavelength and the ellipticity are small,

$|\phi|\lambda \ll 1$ and $|\psi| \ll 1$. It contains the example of the preceding section as a special case. Take $\nu = \nu_{fg} = \nu_{ma}$ and consider only the z component of one term. Note the relationship,

$$2i\{(p_x)_{ma}(p_y)_{am} - (p_y)_{ma}(p_x)_{am}\}$$
$$= (p_x - ip_y)_{ma}(p_x + ip_y)_{am}$$
$$\quad - (p_x + ip_y)_{ma}(p_x - ip_y)_{am}$$
$$= e^2\{|(x - iy)_{ma}|^2 - |(x + iy)_{ma}|^2\}. \quad (23)$$

The inverse linewidth Γ^{-1} is equal to $\pi^2 g_{max}(\nu)$ and the relationship $|\psi| = \frac{1}{4}(\beta_L - \beta_R)$ is readily established by comparison of Eqs. (17), (22), and (23).

The interest is now centered on transparent regions of the crystal, where the term $2\pi i \nu_L \Gamma_{fg}$ in the denominator is negligible compared to $\nu_L^2 - \nu_{fg}^2$. Equation (22) then reduces to a well-known expression first given by Kramers.[28] One might think that the effects off-resonance should be even smaller than the result of the preceding section. In that case, however, the optical matrix element is very small and is due to higher order admixtures of the spin-orbit coupling. In that way a dependence of the spin orientation in the ground state (spin-only) quartet could be obtained.

If the orbital angular momentum is not quenched, much larger effects which are still appreciable in the transparent regions of the crystal result. For the present purpose the interest will therefore be centered on crystals containing ions of the rare earth group in "non-S" states, in which microwave magnetic resonances are observable.

Consider first the case of free ions. The occupied states f are characterized by principal quantum number n, total angular momentum j. The summation over f reduces to a summation over the magnetic quantum number m. The excited states g are characterized by $n'j'm'$. The frequency ν_{fg} can be assumed to be independent of m and m', $\nu_{gf} = \nu_{n'j'} - \nu_{nj}$, since the Zeeman splittings are small compared to the optical frequencies. The dependence of the numerator on m and m' is, according to the Wigner-Eckart theorem and Eq.(23), contained in a term with the Clebsch-Gordan coefficients[29] $\{C^2(j1j'; m, m' = m+1) - C^2(j1j'; m, m' = m-1)\} \propto m$.

The paramagnetic Faraday rotation around the z axis in a $2j+1$-manifold of given total angular momentum is therefore proportional to the z-component of magnetization, and can be expressed as a partial trace,

$$\phi_z \propto \sum_m m\rho_{mm} = \mathrm{Tr}_m\{M_z\rho\} = \langle M_z \rangle. \quad (24)$$

Consider next the influence of a crystalline field. Following the argument of Van Vleck,[30] one assumes that no other j' states are admixed to the ground-state multiplet. The crystal field splitting is assumed to be

[26] J. M. W. Milatz and N. Bloembergen, Physica **9**, 449 (1946).
[27] E. U. Condon, Revs. Modern Phys. **9**, 432 (1937).

[28] H. A. Kramers, Proc. Acad. Sci. (Amsterdam) **33**, 959 (1930); also *Collected Papers*, p. 522. A factor 4π appears to be missing from his Eq. (34).
[29] Compare reference 23, p. 46 and pp. 85 ff.
[30] J. H. Van Vleck and M. H. Hebb, Phys. Rev. **46**, 17 (1934).

small compared to the separation between the ground-state multiplet and excited states. The new ground-state multiplet can be denoted by $nj\mu$, the new excited states by $n'\zeta'\mu$. The new ground-state multiplet is connected to the old one by the unitary transformation

$$\psi_\mu = \sum_m \delta_{nj:n'j'} U_{\mu m} \psi_m.$$

Furthermore, it is assumed that the variation in the frequency denominators due to the crystalline field is negligible,

$$\nu_L{}^2 - \nu^2(n'\zeta'\mu'; nj\mu) = \nu_L{}^2 - \nu^2(n'j', nj).$$

With these assumptions the trace relationship (24) remains valid on the application of a crystal field.

This argument of Van Vleck remains *a fortiori* valid if a microwave resonance field is applied near the frequency $\nu_{\mu\mu'}$, causing transitions between the states μ and μ' in the ground state multiplet. The unitary transformation associated with the microwave resonance is described by a two-by-two matrix, $U_{\mu\mu',\mu''\mu'''}$ and is similar to the transformation of a spin $S = \frac{1}{2}$ quantized along H_0, to a rotating coordinate system and subsequent quantization along the direction of the effective field.

The trace relationship (24) still holds. The modulation of the plane of polarization is proportional to the modulation of magnetization along the direction of the incident light. This latter quantity is, however, given by Eq. (3) with $\mu = 1$, $\mu' = 2$. If the dc paramagnetic Faraday rotation is known, its modulation can be calculated,

$$\phi_z = (\langle M_z \rangle / M_s)\phi_{\text{sat}}. \qquad (25)$$

Here M_s is the saturation magnetization and ϕ_{sat} is the saturation value of the rotatory power.

Consider for example a crystal of neodymium ethylsulphate with linearly polarized light incident along the axis of symmetry, which is taken in the z direction. A dc magnetic field is applied along the x axis of about 5500 gauss which splits the ground state Kramers doublet by 0.5 cm^{-1}, corresponding to a resonant frequency $\nu_0 = 15$ kMc/sec.

The crystal is placed along the axis of a cylindrical cavity driven in the TE_{011} mode. The microwave field is along the z axis with an amplitude of 1 gauss. The geometry is therefore quite similar to that used in the preceding example, Fig. 1(b).

The saturation value of the optical paramagnetic Faraday rotation has been determined by Becquerel, de Haas, and van den Handel[31] as 114°/mm, for a saturation magnetization $\frac{1}{2}g_{11}\beta N$. If the crystal is kept at 1.6°K, $\rho_{11}{}^0 - \rho_{22}{}^0 = \tanh(h\nu_0/2kT) = 0.22$.

In the undiluted salt there is no resolved structure for H_0 perpendicular to the axis of symmetry. The half line width between half maximum power points ΔH is about

150 oersteds.[32] This width, which may be equated roughly to $(g_\perp \beta h^{-1} T_2)^{-1}$, is due to the magnetic interactions between Nd^{+++} ions.

The anisotropic g factor has also been measured by Bleaney,[33] $g_{11} = 3.53$ and $g_\perp = 2.07$. Note the advantage of the large g_{11}, when one substitutes for $M_{12}{}^z$ in Eq. (3). One finds for $(\omega - \omega_{12})T_2 = 1$,

$$\langle M_z \rangle = \frac{1}{4}\frac{g_{11}}{g_\perp}\frac{H_{\text{rf}}}{\Delta H}(\rho_{11}{}^0 - \rho_{22}{}^0)M_s \cos\omega t. \qquad (26)$$

With Eq. (25) one finds for the modulation amplitude ϕ_M of the plane of polarization, after passage through a 5-mm thick crystal, about 20 minutes of arc. If a magnetically dilute ethylsulphate crystal is taken (3% Nd, 97% La), the line width ΔH is about 7.5 oersted and caused mostly by the local field from the protons. One gains a factor 20 in inverse width. At the same time one loses a factor 30 in saturation magnetization. One has to multiply also by a factor 0.8, representing the abundance of the even isotopes. The odd isotopes have a resolved hfs and do not contribute to the precessing magnetization. The rotation amplitude would be 12' of arc in 5 mm thickness. Nevertheless, a dilute specimen is preferable because the microwave resonance is more clearly defined, the power dissipation per unit volume is much reduced and the ratio of modulated to dc rotary power is increased.

In dilute (3% Ce, 97% La) ethylsulphate with $g_{11} = 0.95$, $g_\perp = 2.18$ and a saturation rotation 430°/mm, Eqs. (25) and (26) yield about the same value for ϕ_M under the same conditions.[34,35] In either case a microwave field of one oersted fulfills the condition (4). The spin-lattice relaxation time in the Nd salt has recently been measured by observation of the recovery of dc Faraday rotation after saturation. Daniels[36] finds $T_1 \sim 10^{-2}$ sec. The relaxation time in Ce salts drops exponentially at low temperature. The process takes place via excited doublets.[37] Although the relaxation time in the ethylsulphate is not known accurately, it is expected to become longer than a millisecond below 2°K. The power absorbed by the spins from the microwave field is of the order of a milliwatt/cc, and there is no absorption of light in this case.

The expected rotation of about a quarter of a degree is just near the limit of resolution of a pair of crossed Nicol prisms. Let $\langle \phi_0 \rangle$ and $\langle \phi_0{}^2 \rangle$ denote the mean and mean square deviation from perfectly crossed polarizers.

[31] J. Becquerel, W. J. deHaas, and J. van den Handel, Physica **5**, 753 (1938).

[32] B. Bleaney, R. J. Elliott, and H. E. D. Scovil, Proc. Phys. Soc. (London) **A64**, 933 (1951).

[33] B. Bleaney, H. E. D. Scovil, and R. S. Trenam, Proc. Roy. Soc. (London) **A223**, 15 (1954).

[34] K. D. Bowers and J. Owen, *Reports on Progress in Physics* (The Physical Society, London, 1955), Vol. 18, p. 364.

[35] J. Becquerel, W. J. deHaas, and J. van den Handel, Physica **5**, 857 (1938).

[36] J. M. Daniels and K. E. Riethoff, Can. J. Phys. **38**, 604 (1960).

[37] C. B. P. Finn, R. Orbach and W. P. Wolf, Proceedings of the Seventh International Conference on Low-Temperature Physics, Toronto, 1960 (unpublished).

The light field after passage through polarizer, modulating crystal and analyzer is represented by

$$A_0 \cos\omega_L t \sin[\phi_M \cos\omega t + \langle\phi_0\rangle]$$
$$\approx A_0 \cos\omega_L t(\phi_M \cos\omega t + \langle\phi_0\rangle), \quad (27)$$

since $\phi_M \approx 0.5 \times 10^{-2} \ll 1$ and $\langle\phi_0\rangle \ll 1$.

The intensity of the signal in terms of the incident intensity $\mathcal{J}_0$ is

$$\mathcal{J} = \mathcal{J}_0 \phi_M^2 \times \tfrac{1}{2}(1+\cos 2\omega t)$$
$$+ 2\mathcal{J}_0 \phi_M \langle\phi_0\rangle \cos\omega t + \mathcal{J}_0\langle\phi_0^2\rangle. \quad (28)$$

The modulation can be detected in the same manner as discussed in the preceding section. It can also be detected by spectral analysis. If the incident light consists of a sharp spectral line with width $\Delta\omega$ smaller than the microwave frequency ω, side bands at $\omega_L \pm \omega$ can be resolved. The intensity in each side band is $\tfrac{1}{4}\phi_M^2\mathcal{J}_0 \sim 3 \times 10^{-6}\mathcal{J}_0$. This should be compared with the intensity of the central component $\mathcal{J}_0\langle\phi_0^2\rangle$ which is of the order of $10^{-5}\mathcal{J}_0$ for crossed Nicols. A spectrometer of moderate resolution (0.1 cm^{-1}) should suffice. A resonant vapor absorption cell would also be suitable to eliminate the central carrier. The resonance D light from an alkali vapor discharge may, e.g., be passed through an absorption cell containing the same alkali vapor. The first term on the right-hand side in the expression (28) for the intensity arises from beats of the side bands with themselves and each other. The second term represents beats of the side bands with the central carrier. It would be detectable as modulation of the photoelectric effect. This effect assumes a maximum value for $\langle\phi_0\rangle = 45°$, as can be verified immediately from the left-hand side of Eq. (27) when the approximation $\langle\phi_0\rangle \ll 1$ is not made.

The analogy of the side bands discussed here with Raman lines is only superficial. There is no definite phase relationship between the Raman lines and the incident light, as the incident and scattered light and the molecular vibrations have random phases. In our case the coherent microwave precession of the spins produces a well defined phase relationship between each light component in the central line and the side bands.

An important advantage of the Faraday rotation is the absence of light absorption, in contrast to the absorptive nature of circular dichroism. The incident light may cover a broad frequency interval. Considerably larger total light intensities can be handled. The thickness of the active crystal is not limited by absorption.

In noncubic crystals the aperture is severely limited, because of rotation of the plane of polarization by the anisotropic dielectric tensor. The interference with the Faraday rotation has been described by Kramers[28] and Becquerel.[38] For this reason the incident light has been taken parallel to the optic axis in our examples. For a light ray making a small angle θ with the optical symmetry axis, the angle ϕ of the plane of polarization will be changed to $-\phi$ by birefringence in a distance

[38] J. Becquerel, J. phys. radium **9**, 337 (1928).

$d=\lambda\epsilon_\perp/\theta^2(\epsilon_{11}-\epsilon_\perp)$ where λ is the wavelength in the crystal for light moving parallel to the axis. The aperture of the light beam and the thickness of the crystal are therefore limited by the relation

$$d\theta^2 \ll \lambda\epsilon_\perp/(\epsilon_{11}-\epsilon_\perp).$$

Since the relative anisotropy of the refraction index in the ethylsulphates is about 0.5% or $(\epsilon_{11}-\epsilon_\perp)/\epsilon_\perp = 10^{-2}$, the apex angle θ is limited to about 3° for a 5-mm thick crystal. The thickness of the crystal is of course also limited by the phase of the microwave field. It should not be more than half a guide wavelength thick.

A crystal with cubic symmetry and an isotropic index of refraction would offer an important advantage. The alignment of the crystal with respect to light beam and magnetic field would be much less critical and larger apertures could be used. A CaF_2 crystal with rare earth ions as impurities is an example of such a material. It is not necessary that the individual ions are at sites of cubic symmetry. Microwave resonances from cubic and noncubic sites have been reported.[39-41]

V. OTHER METHODS AND APPLICATIONS

Although the calculated effects are small, they should be detectable in the laboratory and could be used in a determination of the velocity of light.[42] The light beam would be reflected from the mirror and pass the same paramagnetic crystal a second time. As the distance between mirror and crystal is varied, the modulation depth of the light, for, e.g., intensity in the side bands, would pass alternatively through maxima and zero. The crystal would act like a very fast shutter, about two orders of magnitude faster than the fastest Kerr cell, for a small fraction of the light. This small fraction can readily be identified by the modulation and crossed polarizer techniques described in this paper.

The magnitude of the effect can be enhanced considerably in several ways over the numerical results obtained in the preceding section. The magnitude of the precessing magnetization, and therefore ϕ_M, can be enhanced by an order of magnitude in microwave pulses. A light source could be pulsed at the same time, so that the product $\mathcal{J}_0\phi_M$ or $\mathcal{J}_0\phi_M^2$ in Eq. (28) is increased by factor 10^3 or more. The rotation per unit length also increases generally toward the shorter optical wavelengths. The efficiency of the photoelectric detector increases at the same time. Incident light from the near infrared to the near ultraviolet can readily be used in these experiments. Significant increases in the modulation depth can be obtained according to Eq. (22) in a wavelength interval on either side of narrow optical

[39] J. M. Baker, B. Bleaney, and W. Hayes, Proc. Roy. Soc. (London) **B247**, 141 (1958).

[40] J. M. Baker, W. Hayes, and D. A. Jones, Proc. Phys. Soc. (London) **73**, 943 (1959).

[41] M. Dvir and W. Low, Proc. Phys. Soc. (London) **75**, 136 (1960).

[42] E. Bergstrand, *Encyclopedia of Physics* (Springer-Verlag, Berlin, Germany, 1956), Vol. 24, p. 1.

absorption lines in the spectrum of the rare earth ion. No Faraday rotation experiments appear to have been made in the vicinity of such absorption lines.

In the absence of significant light absorption the thickness of the crystal may be increased. As in the case with other magnetic resonance applications, extension to higher frequencies into the millimeter range is straightforward if sufficient microwave driving power is available. The increased value of $h\nu/kT$ will also enhance the modulation angle ϕ_M. If the dimension of the crystal becomes comparable to or larger than the microwave wavelength, a traveling wave geometry is indicated in which the light wave and microwave travel with the same phase velocity. It is at this time a matter of speculation whether the modulated photoemission current and frequency multiplication effects inherent in Eqs. (25)–(28) can be used to advantage at these high frequencies. Phase control of microwave signals at different points in space by means of a modulated light beam is another possibility.

The corresponding effect in ferromagnetic materials would offer the advantage of a large magnetization at a more convenient operating temperature. In particular the exchange narrowed resonance in yttrium iron garnet (YIG) appears attractive. Dillon[43] has briefly mentioned the possibility of a combination of microwave and optical phenomena in garnets. Nevertheless, the maximum obtainable effect in YIG is rather smaller than in the paramagnetic crystals cited above. The transverse rotating component of magnetization assumes a maximum value $(\Delta H_k M_s/4\pi)^{\frac{1}{2}}$ for a critical microwave field strength $H_{\text{crit,rf}} = \Delta H (\Delta H_k/4\pi M_s)^{\frac{1}{2}}$, at which other spin waves with finite wavenumber become excited through nonlinear coupling effects.[44] In these expressions M_s is the saturation magnetization, ΔH_k the true line width due to finite lifetime, ΔH is observed width, which includes inhomogeneity broadening. The maximum ratio of transverse to saturation magnetization, $\langle M_z \rangle/M_s$ in Eq. (25), is about 0.05 at a microwave field strength of a few tenths of an oersted in carefully polished garnet. Although larger ratios exist in principle in ferromagnetic materials with larger damping ΔH_k, the microwave power required for maximum precession angle is prohibitive in such cases.

The dc Faraday rotation in YIG is 200°/mm at 7000 A. Since the absorption coefficient is 1300 cm^{-1} at this wavelength, the maximum useable thickness is only 0.02 mm. Nothing would be gained by going to other wavelengths, since the rotary power and absorption coefficient are roughly proportional. It follows from Clogston's theory[45] of ferromagnetic rotary power and the pseudo-vector transformation property of magnetic rotary power[28] that Eq. (25) remains valid in this ferromagnetic case. The plane of polarization would be modulated by $0.02 \times 200 \times 0.05 = 12$ minutes of arc. Although this effect is of the same order of magnitude as for the ethylsulphates, it would be much harder to detect in the presence of magnetostriction causing optical birefringence and of circular dichroism. The possibilities for enhancement discussed above for paramagnetic crystals do not exist in this case.

Ultrasonic vibrations with concomitant variations in the optical index of refraction by acoustical strain have been used extensively in quartz at lower frequencies.[42] Extension of ultrasonic techniques to the microwave frequency range has recently been achieved. It has not yet been possible[46] to set up a microwave sound field in a well-defined mode over a rather large volume containing many acoustical wavelengths. This would be a prerequisite for optical modulation by this technique, which also requires very low temperatures because of ultrasonic attenuation.

It may therefore be concluded that an experimental investigation of microwave modulation of light in paramagnetic crystals is well warranted, especially in cubic crystals containing rare earth ions. Such a program is currently under way in our laboratory.

Note added in proof. The solid state Kerr effect[47] which occurs in dihydrogenphosphates may be modulated at microwave frequencies. The displacements of the hydrogen atoms in this crystal can follow rapid variations in an electric field. It turns out accidentally that a given energy density of electromagnetic microwave radiation produces about the same rotation per unit length of the plane of polarization in dihydrogenphosphate at room temperature as in the paramagnetic crystals at liquid helium temperature.

[43] J. F. Dillon, J. Appl. Phys. **29**, 539 (1958). See footnote on p. 541.

[44] See, e.g., H. Suhl, J. Phys. Chem. Solids **1**, 207 (1957); J. J. Green, thesis, Harvard University, 1959 (unpublished).

[45] A. M. Clogston, J. phys. radium **20**, 151 (1959).

[46] E. H. Jacobsen, *Proceeding of Conference on Quantum Electronics* (Columbia University Press, New York, 1960), p. 468.

[47] R. O. Carpenter, J. Opt. Soc. Am. **40**, 225 (1950).