"OFF-DIAGONAL" MAGNETIC RESONANCE

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The evolution of a magnetic moment in a system of paramagnetic particles optically oriented across a constant magnetic field $\mathbf{H}_0$ in the presence of a variable field rotating with frequency ω about $\mathbf{H}_0$ is investigated. It is shown that under these conditions a distinctive type of magnetic resonance may be realized, differing sharply from ordinary paramagnetic resonance. The resonance is characterized by the buildup of a considerable moment with a mean value across the field $\mathbf{H}_0$ that is modulated with a frequency 2ω. In the direction of the constant field, only an alternating component of the moment, with frequency ω, arises. The calculation is performed on the basis of Bloch's phenomenological equations. An experimental verification is carried out in optically oriented Cd^{111} vapor.

1. INTRODUCTION

PARAMAGNETIC resonance under ordinary conditions is described classically as the induced conical precession of a magnetic moment vector about the direction of a constant magnetic field $\mathbf{H}_0$ under the action (and with the frequency) of a rotating magnetic field $\mathbf{H}_1$ perpendicular to the constant field. Such a situation arises in the case when the reason for the appearance of magnetic polarization in a medium is the thermal relaxation of paramagnetic particles in an external polarizing field such that the direction of the stationary polarization coincides with the direction of the field. At present, there exists an optical method of magnetic-moment polarization (cf., e.g., the latest review[1]) which makes it possible to effect orientation of atomic moments in an arbitrary direction with respect to the magnetic field. In particular, a characteristic case is possible, when by means of circularly polarized light orientation of atomic spin moments is effected across the constant magnetic field. It is obvious that if the rate of orientation is slow compared with the period of precession of the moment in the field, no significant moment will build up in the system.

The position, however, is changed when variable magnetic fields are applied. An interesting situation, called parametric resonance, arises when the variable field is parallel to the constant field[2,3]. Below we consider phenomena arising in the presence of a field $\mathbf{H}_1$ perpendicular to the constant field $\mathbf{H}_0$ and rotating about it, i.e., for an arrangement of magnetic fields that is typical of ordinary paramagnetic resonance. The combination of these fields with a transverse polarization leads to the appearance of a very distinctive type of magnetic resonance, differing sharply from the ordinary type. Whereas in the ordinary case the application of the field $\mathbf{H}_1$ leads to a resonant decrease of the total magnetization and to the appearance of transverse components of the moment oscillating with the frequency ω of the field $\mathbf{H}_1$, the resonance in our case is manifested in the first place in the appearance in the system of a macroscopic moment, absent up to this point. The moment vector, the magnitude of which is conserved in time, changes its direction in a complicated way, such that its projection along the constant-field axis oscillates about zero with frequency ω whereas the transverse components, while having mean values, oscillate with frequency 2ω. Thus, the mean magnetization of the system is directed across the constant field $\mathbf{H}_0$. The conditions for the appearance of a resonance are also unusual.

Formally, the difference between the two types of resonance reduces to a difference in the type of relaxation. In the ordinary case, relaxation in the absence of a variable field leads to the establishment of longitudinal magnetization. In the case considered here, the relaxation, in which the optical orientation process is included, tends to establish in the system a transverse magnetization, which is opposed by the constant field. Since transverse magnetization is associated in the theory with the off-diagonal elements of the density matrix, this type of magnetic resonance can be conventionally called "off-diagonal."

A situation close in principle to "off-diagonal" resonance arose in the work of Dodd and Series[4], in which the modulation of the emitted radiation of excited mercury atoms was studied, for off-diagonal excitation in particular. However it was not possible to observe the above resonance in the proper sense, under the conditions of this work because of the very large width of the excited levels; for this an over-powerful variable field would be required.[1)]

2. FUNDAMENTAL RELATIONS

"Off-diagonal" resonance is most simple described by means of a system of phenomenological Bloch equations, in which an inhomogeneous term is introduced into the equation for a transverse component of the moment $\mathbf{M}$, e.g., for M_x:

$$\frac{d\mathbf{M}}{dt}=\gamma[\mathbf{MH}]-[\mathbf{i}(M_x-M_0)+\mathbf{j}M_y+\mathbf{k}M_z]\Gamma. \qquad (1)^*$$

Here γ is the gyromagnetic ratio. For simplicity and in accordance with the experimental conditions, the relaxation parameter Γ is assumed to be the same for the longitudinal and transverse components of the moment.

[1)]In this work, alignment rather than polarization of the atoms was effected, so that no magnetic moment at all arose in the system and the question would have been one of off-diagonal quadrupole resonance.

$^*[\mathbf{MH}] \equiv \mathbf{M}\times\mathbf{H}$.

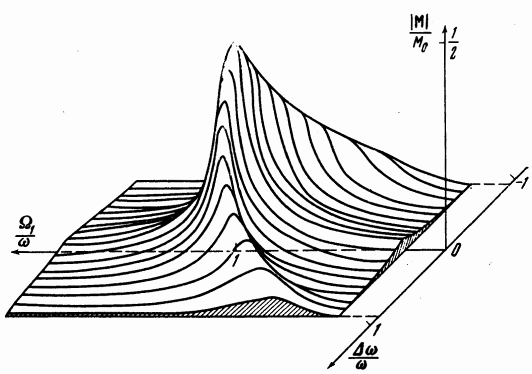

FIG. 1. Dependence of the modulus $|\mathbf{M}|$ of the magnetic moment on the amplitude H_1 of the variable field and the detuning $\Delta\omega$.

In accordance with what has been said above, the magnetic field **H** has the form

$$\mathbf{H} = \mathbf{i}H_1 \cos \omega t + \mathbf{j}H_1 \sin \omega t + \mathbf{k}H_0. \qquad (2)$$

The phenomenological orientation parameter M_0 has the meaning of the magnetization that would be established in the system along the X-axis (along which the polarizing light beam is assumed to be directed) in the absence of any magnetic fields.

The system of Eqs. (1) for a magnetic field of the form (2) admits an exact solution, which is rather cumbersome. Written out below are simplified expressions, valid for $\omega \gg \Gamma$, i.e., applicable in the region in which the magnetic splitting of the levels is appreciably greater than their width Γ and we can keep only the resonance terms:

$$M_x = \Gamma M_0 \frac{(1-\Delta\omega/\Omega)^2\Gamma - (\Omega_1/\Omega)^2[\Gamma\cos 2\omega t - (\Omega-\omega)\sin 2\omega t]}{4[\Gamma^2 + (\Omega-\omega)^2]},$$

$$M_y = \Gamma M_0 \frac{(\omega-\Omega)(1-\Delta\omega/\Omega)^2 - (\Omega_1/\Omega)^2[(\Omega-\omega)\cos 2\omega t + \Gamma\sin 2\omega t]}{4[\Gamma^2+(\Omega-\omega)^2]},$$

$$M_z = \Gamma M_0 \frac{(1-\Delta\omega/\Omega)^2[(\Omega-\omega)\sin\omega t - \Gamma\cos\omega t]\Omega_1/\Omega}{2[\Gamma^2+(\Omega-\omega)^2]} \qquad (3)$$

We have used the usual notation: $\Omega_1 = \gamma H_1$,

$$\Omega = \sqrt{\Omega_1^2 + \Delta\omega^2}, \quad \Delta\omega = \omega_0 - \omega, \quad \omega_0 = -\gamma H_0.$$

By examining the solutions (3), we can see that the modulus $|\mathbf{M}|$ of the moment

$$|\mathbf{M}| = \frac{\Gamma M_0}{2} \frac{1-\Delta\omega/\Omega}{\sqrt{\Gamma^2+(\Omega-\omega)^2}} \qquad (4)$$

does not depend on the time and increases resonantly in the neighborhood of $\Omega = \omega$. The quantity $|\mathbf{M}|$ at resonance $\Omega = \omega$ depends on the detuning $\Delta\omega$ and has a maximum at $\Delta\omega = 0$. Since $\Omega = \sqrt{\Omega_1^2 + \Delta\omega^2}$, the resonance $\Omega = \omega$ can be achieved by variation of the two variables Ω_1 and $\Delta\omega$ within the limits 0 to $|\omega|$. Figure 1 shows a three-dimensional diagram of the dependence of the modulus of the moment on Ω_1 and $\Delta\omega$.

As can be seen from (3), the moment vector describes a complicated motion in space. The tip of the vector moves along the line of intersection of a sphere of radius $|\mathbf{M}|$ with a right circular cylinder touching the sphere from within. Figure 2 shows a hodograph of the moment at the principle resonance, i.e., for $\Delta\omega = 0$ and $\Omega - \omega = 0$. For this case the solutions have their simplest form:

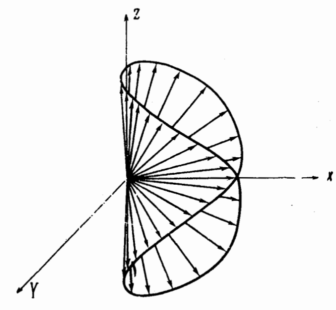

FIG. 2. Hodograph of the moment **M** at the principle resonance ($\Delta\omega = 0, \Omega - \omega = 0$).

$$M_x = \frac{M_0}{4}(1-\cos 2\omega t), \quad M_y = -\frac{M_0}{4}\sin 2\omega t,$$
$$M_z = -\frac{M_0}{2}\cos\omega t, \quad |\mathbf{M}| = M_0/2.$$

Thus, at the principle resonance the angular momentum of the system increases up to a value only half as large as the stationary value attained in the complete absence of fields.

We note a further interesting feature of "off-diagonal" resonance—the absence of saturation. The resonance contour of any component of the moment as a function of the difference $\Omega - \omega$ is Lorentzian with width Γ, irrespective of the intensity H_1 of the variable field.

3. THE EXPERIMENT

As material for the experiment we used optically oriented cadmium Cd^{111} vapor[5]. To eliminate spurious effects from the modulation of the luminescence in the variable magnetic field, the polarization was produced in the presence of a buffer gas, xenon, at pressure of a few torr.

The experimental scheme is given in Fig. 3. Polarization of the cadmium vapor in the quartz cell was

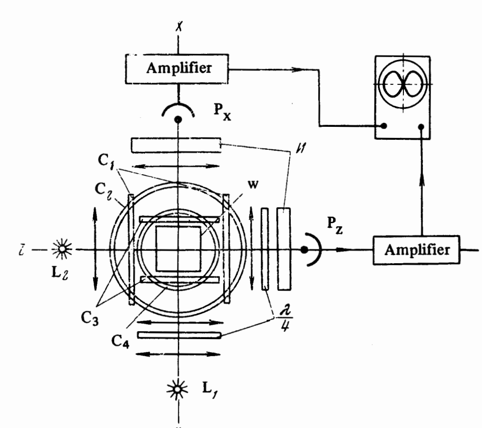

FIG. 3. Scheme of the experiment. W—Working cell with Cd^{111} vapor; C_1, C_2—Helmholtz coils to produce the constant field $\mathbf{H}_0$; C_3, C_4—Helmholtz coils to produce the variable field $\mathbf{H}_1$; L_1, L_2—lamps with Cd^{114}; P_X, P_Z—photocells; $\lambda/4$—circular polarizer; I—interference filter

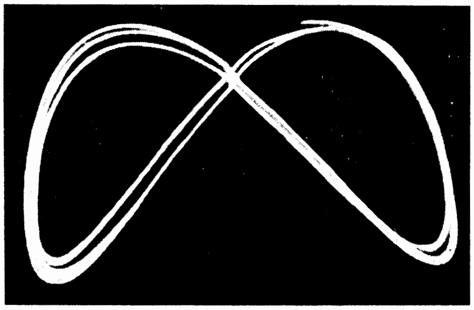

FIG. 4. Oscillogram of the variable components of the "off-diagonal" resonance signal. The X signal is fed to the vertically deflecting plates and the Z signal to the horizontally deflecting plates.

effected by circularly polarized 3261 Å light from a lamp with the isotope Cd^{114}, and this led to the selective excitation of the short-wavelength component of the hyperfine doublet. The light was directed at a right angle to the constant magnetic field H_0, the intensity of which was varied in a range about the horizontal component of the earth's field (the vertical component was compensated). The field H_1 was created by two orthogonal pairs of Helmholtz coils, which were supplied from an audio-frequency oscillator with a phase difference of 90°. An unpolarized beam of radiation of the same wavelength from a second lamp with Cd^{114} was passed through the cell along the direction of the field H_0. After passing through the cell and a circular analyzer, this beam was detected by the photocell P_z, the signal from which was proportional to the component M_z.

Under these conditions with an approximate choice of the field intensities H_1 and H_0, a clear resonance was detected, manifested in the onset of modulation at frequencies 2ω and ω in the beams along the X- and Z-axes. Figure 4 shows an oscillogram plotted with the X signal fed to the vertically deflecting plates of the oscillograph

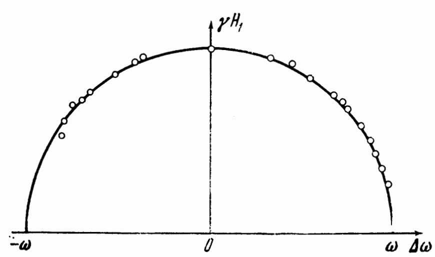

FIG. 5. Experimentally determined positions of the resonance in the coordinates Ω_1 and $\Delta\omega$. For comparison, the line $\Omega_1^2 + \Delta\omega^2 = \omega^2$ is shown.

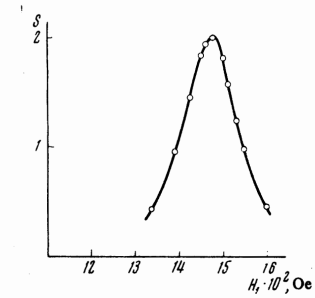

FIG. 6. Dependence of the amplitude of the X-beam modulation with frequency 2ω on the field-intensity H_1 for $\Delta\omega = 0$.

and the Z signal fed to the horizontally deflecting plates. The phases of the signals were not monitored.

For the X beam, a series of curves was then plotted for its amplitude as a function of the detuning $\Delta\omega$ for different H_1. Figure 5 shows the position of the resonances (the maxima of the modulation amplitude) in the coordinates $\Delta\omega$, Ω_1. It can be seen that, in agreement with the calculation, the experimental points are well positioned on the semicircle $\Delta\omega^2 + \Omega_1^2 = \omega^2$.

The dependence of the amplitude of the X-beam modulation with frequency 2ω on the field-intensity H_1 at zero detuning $\Delta\omega = 0$ is shown in Fig. 6.

As we should have expected, the resonance was found to be extremely narrow, with half-width ~7 Hz, which corresponds to the relaxation time for cadmium nuclei in the experimental conditions. We remark that at resonance the field-intensity H_1 was ~0.15 Oe, which under ordinary paramagnetic resonance conditions would have led to a line-broadening of up to 150 Hz.

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