SF_6 with H_2S . When the $SF_6 + H_2S$ mixture was exposed to CO_2 laser radiation of frequency $\nu = 948$ cm⁻¹, a shift of the isotopic ratio of the residual gas towards the ³⁴S isotope was attained.

It was shown recently^[15] that v-T relaxation of CO dissolved in liquid nitrogen is strongly suppressed. It is possible that this is also true of other molecules (for example, solutions of SF₆ in liquid oxygen or nitrogen). These solutions should satisfy then all the above-mentioned requirements needed to ensure selectivity of the process when the molecules are excited with a "long" pulse of laser radiation $(\tau_p > \tau_{r-r})$, and could be used successfully for isotope separation and for other laser-chemical processes. Finally, we note that at low temperature the process can be made selective even at an appreciable energy exchange between the isotopes.^[161]

The authors consider it their pleasant duty to thank V. N. Kosipov for a useful discussion of the results, N. A. Vishnyakov for help with the experiments, and Yu. S. Leonov for his isotopic analysis of the SF_6 .

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Translated by J. G. Adashko

Observation of radiofrequency coherence during the excitation of hyperfine structure transitions in optically oriented atoms

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Radiofrequency coherence of states corresponding to the Zeeman sublevels of optically oriented cesium atoms during the excitation of hyperfine structure transitions by amplitude-modulated microwave magnetic field is reported. The coherence signals were recorded both by varying the modulation frequency and by varying the frequency of the microwave field. The signals have a different shape and behavior, depending on the intensity of the microwave magnetic field. The observed phenomena are examined theoretically, and the theoretical results are compared with experimental data.

PACS numbers: 32.60.+i, 32.30.Bv

Experiments on the optical orientation of atoms have revealed a modulation of light absorption both at the transition frequency between the Zeeman sublevels of one of the hyperfine structure levels^[1] and at the transition frequency between different hyperfine structure levels.^[2] The light intensity is then found to be modulated at the frequency of the magnetic resonance. Light modulation occurs because the magnetic resonance is accompanied by the coherent superposition of states by the optically oriented atoms, and this is manifested microscopically by the appearance of a component of the resultant magnetic moment which precesses around the constant magnetic field, or oscillates with the resonance frequency, and gives rise to the modulation of absorp-

0038-5646/77/4503-0484\$02.40



FIG. 1. Block diagram of the apparatus.

tion of light by the atoms.

In this paper, we report for the first time the modulation of light absorption at the frequency of the Zeeman transition during the excitation of transitions between the hyperfine structure levels. This excitation was achieved with amplitude-modulated microwave radiation with modulation frequency close to the frequency of the Zeeman transition. Light modulation was due to the coherent superposition of states other than those between which the resonance transitions took place. Coherence then occurred as a result of correlation between the main and the side components of the modulated perturbation of the atomic system, which excited simultaneously two hyperfine structure transitions. The observed coherent superposition of states is, in many ways, similar to other interference states of quantum-mechanical systems that have been investigated under the action of modulated perturbation.^[3]

We have investigated the coherent superposition of states with F=4, $m_F=4$ and F=4, $m_F=3$ in optically oriented ¹³³Cs atoms. The properties of the resulting resonance signals were investigated, and the experimental results were compared with theory.

1. EXPERIMENTAL METHOD

The cesium atoms were oriented optically by circularly polarized resonance radiation directed along the magnetic field. The alternating magnetic field exciting transitions between the hyperfine structure sublevels of the ground state of cesium was produced by a 3-cm microwave generator.

A block diagram of the 4pparatus is shown in Fig. 1. The pump radiation was produced by the cesium lamp 1 in which the oscillator 2 excited a high-frequency electrodeless discharge. The light emitted by this lamp was collimated by the lens 3 and then directed onto the polaroid 4 and the quarter-wave plate 5, in which it became circularly polarized before entering the cell 6. The cell walls were coated with paraffin which contained ¹³³Cs vapor at room temperature. The cell was in the form of a cylinder, 50 mm in diameter and 70 mm long. The constant magnetic field H_0 was produced by Helmholtz coils. The change in the absorption of pump radiation transmitted by the cell 6 and focusing lens 7 was recorded by the photodetector 8. The alternating magnetic field H_1 , which induced transitions between the hyperfine structure levels of the ground state of the cesium atoms was produced by the rectangular horn 9, to which the microwave power was transmitted from the 3-cm oscillator 10 along a waveguide channel. The horn was oriented relative to H_0 so that the π ($\Delta m_F = 1$) and σ (Δm_F =0) magnetic dipole transitions could also be excited. The microwave power supplied to the horn amounted to 8 mW. Amplitude-modulation of the microwave magnetic field was produced by the microwave switching diode 11, to which the output of the radiofrequency oscillator 12 was applied. The depth of the amplitude modulation could be varied between 0 and 95%. Preliminary tuning of the microwave generator 10 to the frequency of the corresponding transition between the hyperfine structure levels was achieved by observing the S_z signal on the screen of the oscillograph 16. The magnetic field H_0 was modulated by applying the output of the audiofrequency oscillator 14 to the modulating coils 13, and the signal from the photodetector 8 was applied to the narrow-band amplifier 15, the output of which was connected to the oscillograph 16. The S_z signal was also used similarly to achieve preliminary tuning of the radiofrequency oscillator 12 to the frequency of the Zeeman transition. In this case, the radiofrequency magnetic field was produced by the coils 17 which were connected to the oscillator 12. The radio and microwave frequencies were measured by an electronic, counting frequency meter 18.

The appearance of coherent superposition of states was recorded as the onset of radiofrequency modulation of the absorption of the probing circularly polarized light traveling at right-angles to the constant magnetic field H_0 in the absorption cell. This light was produced by the cesium lamp 19, which was excited by the oscillator 12. The circular polarization of light transmitted by the lens 21 was produced with the polaroid 22 and the quarter-wave plate 23. The intensity modulation of the light transmitted by the cell was recorded by the photodetector 25 on which the light was focused by the lens 24. Signals from the photodetector 25 were then amplified by the amplifier 26 which was connected either to the amplitude detector 27 or the synchronous detector 28. the reference voltage for which was produced by the radiofrequency generator 12. The output signals from 27 and 28 were recorded on a strip chart recorder 29. The signals could also be inspected visually on the screen of the oscillograph 30.

Signals due to the modulation of light resulting from the coherent superposition of states with F=4, $m_F=4$ and F=4, $m_F=3$ (see Figs. 2a) in optically oriented cesium atoms during the excitation of hyperfine structure transitions by amplitude-modulated microwave radiation were recorded in two ways. In the first method, the modulation frequency was fixed and was equal to the frequency of the Zeeman splitting of the F=4, $m_F=4$ and F=4, $m_F=3$ levels, and the frequency of the microwave generator was slowly varied in the region of the transition frequency between the F=4, $m_F=4$ and F=3, $m_F=3$ levels. In the second method, the microwave fre-



FIG. 2. a) Energy level scheme for the ground state of the cesium atom. b) A three-level quantum-mechanical system.

quency was tuned to the frequency of the $(F=4, m_F=4)$ \leftrightarrow $(F=3, m_F=3)$ transition and was held constant, whilst the frequency of modulation was varied slowly through the $(F=4, m_F=4) \leftrightarrow (F=4, m_F=3)$ transition frequency. In both cases, a light modulation signal at the modulation frequency of the microwave magnetic field was observed with the photodetector 25 when the varied frequency was equal to the transition frequency. The alternating voltage across the photodetector was amplified and recorded as described above. The signals obtained in this way and due to the coherent superposition of the $F=4, m_F=4$ and $F=4, m_F=3$ states will be referred to as the coherence signals henceforth.

2. EXPERIMENTAL RESULTS

It was immediately clear, as soon as the experiments were begun, that there was a considerable difference between the shapes of the coherence signals corresponding to the above two methods of recording. The coherence signal shown in Fig. 3a was obtained by slowly varying the frequency Ω_{32} at which the microwave magnetic field was modulated, using amplitude detection. Curves 1 and 2 in Fig. 3b are the in-phase and in-quadrature components of the same signal obtained by synchronous detection. It was found that the width of the coherence signal, obtained by varying the modulation frequency of the microwave field, was independent of the amplitude of the microwave field and of the depth of its modulation. Moreover, as the microwave power and depth of modulation were increased, the intensity of the signal was also found to increase.

All the resonance curves given in this paper were recorded under the following experimental conditions: modulation depth 95%, angle between the constant and alternating magnetic field approximately 45° .

Figure 4a shows the recorded coherence signal for a slow variation in the frequency Ω_{31} of the microwave magnetic field and amplitude detection. The modulation frequency Ω_{32} was held constant and close to the resonance frequency of the $(F=4, m_F=4) \leftrightarrow (F=4, m_F=3)$ transition. The curves in Fig. 4a correspond to different values of the microwave power radiated by the antenna (P is the relative microwave power, P=1 corresponds to 8 mW fed into the antenna).



FIG. 3. Coherence signals recorded by slowly varying the modulation frequency for a fixed frequency of the microwave magnetic field. The constant magnetic field was $H_0 = 0.69$ Oe. a) Signal obtained with the amplitude detector, b) in-phase (1) and in-quadrature (2) components of the signal recorded with the synchronous detector; c) theoretical shape corresponding to a).



FIG. 4. Coherence signals recorded by slowly varying the frequency of the microwave magnetic field at fixed modulation frequency. The signals were recorded with the aid of an amplitude detector. The magnetic field was $H_0=0.69$ Oe. a) Experimental curves; b), c) theoretical curves. Comparison of Figs. 3a and 4a shows that there is a sharp difference between these signals. The curves shown in Fig. 4a have a much greater width and exhibit a dip at the center. The dip disappears as the micro-wave power is reduced. Although the curves shown in Fig. 4 resemble the usual curves obtained for S_x signals at the Larmor frequency during radiofrequency saturation, ^[4] they do, in fact, exhibit different behavior. Thus, in our case, the dip can easily be reduced to zero simply by reducing the modulation depth without increasing the amplitude of the resonance microwave field.

The influence of the radiofield amplitude on the shape of the resonance curve is, in the present case, essentially different from the effect of radiofrequency saturation for the usual S_r signals corresponding to the modulation of light at the frequency of the magnetic resonance for which the dip at the center cannot be reduced to zero. The asymmetry of the resonance curves shown in Fig. 4a is, to some extent, accidental. Experiment has shown that it is due to imprecise tuning of the modulation frequency to the frequency of the $(F=4, m_F=4)$ \leftrightarrow (F=4, m_F =3) transition. When the coherence signals are recorded, instability in the magnetic field H_0 disturbs precise tuning of the modulation frequency, and the curves become asymmetric. It is clear from Fig. 4a that the two peaks on the coherence signal correspond to a considerable detuning of the microwaves from the hyperfine structure transition frequency (this detuning is about 5 kHz at P=1). It was precisely for this detuning, corresponding to the maximum signal, that the curves shown in Figs. 3a and b were obtained for P=1.

In addition to the foregoing effects, we have observed a further type of coherence signal which appears when the frequency of the microwave magnetic field is close to the frequency of the hyperfine transition (F=4, m_F $=4) - (F=3, m_F=3)$ and the modulation frequency corresponds to twice the frequency of the Zeeman transition. It is clear from Fig. 2a that, in this case, one should observe the coherent superposition of states corresponding to the F=4, $m_F=4$ and F=4, $m_F=2$ levels. To record a coherence signal of this type, we used a transverse, linearly polarized, light beam in which the polarization vector was perpendicular to the constant magnetic field. The resonance curves obtained by the above two methods for such signals are shown in Fig. 5. It is clear that the nature of the variation in the coherence signal is similar to that in Figs. 3a and 4a. It is important to note, however, that the signal amplitude in this effect is substantially smaller than in the previous case.

3. DISCUSSION OF EXPERIMENTAL RESULTS

We shall interpret the observed effects in terms of a three-level system (Fig. 2b) because the modulated microwave magnetic field acts only on three levels. The frequency separations of the levels will be denoted by ω_{21} , ω_{31} , and ω_{32} .

When a multilevel quantum-mechanical system is illuminated by electromagnetic radiation of sufficient power, the usual single-photon processes may be ac-



FIG. 5. Signals corresponding to the coherent superposition of the F=4, $m_F=4$ and F=4, $m_F=2$ states obtained with amplitude detection. The magnetic field was $H_0=0.2$ Oe. a) Signal obtained by slowly varying the frequency of the microwave magnetic field at fixed modulation frequency; b) signal obtained by slowly varying the modulation frequency at fixed frequency of the microwave magnetic field.

companied by nonlinear effects involving multiphoton absorption and emission. For example, in the level system shown in Fig. 2b, the combination harmonic at frequency $\Omega_{32} = \Omega_{31} - \Omega_{21}$ ^[5] appears when the system is illuminated at frequencies $\Omega_{31} \approx \omega_{31}$ and $\Omega_{21} \approx \omega_{21}$. We have, in fact, observed this phenomenon because the microwave radiation at frequency Ω_{31} modulated at Ω_{32} contains the main (Ω_{31}) and side $(\Omega_{21} = \Omega_{31} - \Omega_{32})$ components which are coherent with respect to each other.

To explain the line shape and the behavior of the coherence signals, we must solve the equation for the density matrix in the case of the three-level system interacting with an amplitude-modulated alternating magnetic field. When only the resonance terms are retained in this equation, it becomes identical with the set of equations for a three-level system interacting with two highfrequency fields of frequency $\Omega_{31} \sim \omega_{31}$ and $\Omega_{21} \sim \omega_{21}$. The solution of this set of equations is given, for example, in Fain's book^[6] in the case where the amplitude of one of the fields is much greater than that of the other. In fact, the following inequality should be satisfied:

$$(\mu_{a_{12}}H_{a_{13}})^2 \gg (\mu_{a_{12}}H_{a_{12}})^2, \tag{1}$$

where $\mu_{a_{ij}}$ is the projection of the matrix element of the magnetic dipole moment and $H_{a_{ij}}$ is the amplitude of the resonance magnetic field acting during the i - j transition.

The inequality given by (1) may be regarded as satisfied in our experiments because the matrix element of the magnetic moment for the $(F=3, m_F=3) \rightarrow (F=4, m_F=4)$ transition, excited by the main component of the alternating magnetic field, is greater than the matrix elements for the $(F=3, m_F=3) \rightarrow (F=4, m_F=3)$ and $(F=3), m_F=3) \rightarrow (F=4, m_F=2)$ transitions, excited by the side component. Moreover, the intensity of the main component is greater than that of the side components. In fact, when the modulation depth is 100%, and the alternating magnetic field is at 45° to the constant field, we may write

$$(\mu_{a_{12}}H_{a_{13}})^2/(\mu_{a_{12}}H_{a_{12}})^2 = [(I+1/2+m_F)(I+3/2+m_F)]/[(I+1/2)^2-m_F^2] = 8,$$

where $m_F = 3$, I is the nuclear spin which, in the case of cesium, is 7/2, and the expressions for the matrix elements are taken from the book by Condon and Shortley.^[7]

When (1) is satisfied, the solution for the element σ_{32}

of the density matrix in which we are interested is given by $^{[6]}$

$$\sigma_{32} = A_{12}A_{31} \{ [i(\Omega_{21} - \omega_{21}) + \tau_{12}^{-1}] \\ \times [-i(\Omega_{31} - \omega_{31}) + \tau_{31}^{-1}]^{-1} (\sigma_{11} - \sigma_{33}) + (\sigma_{11} - \sigma_{22}) \} \\ \times \{ [i(\Omega_{21} - \omega_{21}) + \tau_{12}^{-1}] [-i(\Omega_{32} - \omega_{32}) + \tau_{32}^{-1}] \\ + |A_{13}|^2 \}^{-1} \exp(-i\Omega_{32}t) = \rho_{32} \exp(-i\Omega_{32}t),$$
(2)

where

 $\Omega_{32} = \Omega_{31} - \Omega_{21},$

In the above expressions, τ_{ij} is the characteristic time for the attenuation of the nondiagonal elements of the density matrix (transverse relaxation time), i, j = 1, 2, 3; $A_{ij} = \mu_{a_{ij}} H_{a_{ij}}/\hbar$,

$$\begin{split} \sigma_{11} - \sigma_{33} &= \frac{(\sigma_{11}^{\circ} - \sigma_{33}^{\circ}) \left[1 + (\Omega_{31} - \omega_{31})^2 \tau_{31}^2 \right]}{1 + (\Omega_{31} - \omega_{31})^2 \tau_{31}^2 + 4|A_{13}|^2 T \tau_{31}} \\ \sigma_{11} - \sigma_{22} &= (\sigma_{11}^{\circ} - \sigma_{22}^{\circ}) - \frac{2(\sigma_{11}^{\circ} - \sigma_{33}^{\circ}) |A_{13}|^2 \tau_{31} T}{1 + (\Omega_{31} - \omega_{31})^2 \tau_{31}^2 + 4|A_{13}|^2 T \tau_{31}} \end{split}$$

T is the characteristic lifetime of levels 1 and 3, and $\sigma_{11}^0 - \sigma_{22}^0$, $\sigma_{11}^0 - \sigma_{33}^0$ are the initial population differences due to optical pumping $(\sigma_{33}^0 > \sigma_{22}^0 > \sigma_{11}^0)$.

The transverse component of the magnetic moment of the system of atoms which corresponds to coherence between the F=4, $m_F=4$ and F=4, $m_F=3$ levels has the following components:

$$M_x \sim \operatorname{Re}(\sigma_{32}), \quad M_y \sim \operatorname{Im}(\sigma_{32}),$$
 (3)

and the in-phase and in-quadrature components of the recorded signal have the following form in the case of synchronous detection:

$$I_{\text{synch}} \sim \operatorname{Re} \rho_{32}, \quad I_{gu} \sim \operatorname{Im} \rho_{32}.$$
 (4)

In the case of amplitude-detection, the signal is

$$I_{ampl} \sim [(\text{Re } \rho_{32})^2 + (\text{Im } \rho_{32})^2]^{\frac{1}{2}}.$$
 (5)

Let us begin by considering the behavior of the function ρ_{32} for two special cases (we shall assume that $\tau_{12} = \tau_{31} = \tau$).

The first case is:

$$\Omega_{21} - \omega_{21} \approx \Omega_{31} - \omega_{31} \gg \tau^{-1}, \quad (\tau \tau_{32})^{-1} \gg |A_{13}|^2,$$

which corresponds to large detuning in a sufficiently small microwave field at the frequency of the 1 - 3 transition (Fig. 2b). We note that the condition $(\tau \tau_{32})^{-1} \gg |A_{13}|^2$ does not mean the absence of saturation of the 1 - 3 transition since $T \gg \tau$ for the cells with paraffincoated walls, which we used in our experiment.

In the above case, Eq. (2) transforms to

$$\rho_{32} = -A_{12}A_{31} \frac{i(\Omega_{21} - \omega_{21})^{-1}(\sigma_{33} - \sigma_{22})}{\tau_{32}^{-1} - i(\Omega_{32} - \omega_{32})}.$$
(6)

This describes the behavior of the coherence signal as the modulation frequency is varied in the case where the

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frequency of the microwave magnetic field is substantially detuned from the frequency of the hyperfine structure transition, which corresponds to the condition for the observation of this type of signal in our experiment (Sec. 2).

The characteristic feature of (6) is that the population difference $\sigma_{33} - \sigma_{22}$ in the numerator depends only on the microwave magnetic field acting on the 3 - 1 transition, whereas the denominator does not depend on this field. The radiofrequency broadening is therefore zero in this case, and this is confirmed experimentally (Sec. 2).

The second case is:

$$\Omega_{31} - \omega_{31} = \Omega_{21} - \omega_{21} = 0$$

and this corresponds to precise resonance (bottom of the dip in Fig. 4a). In this case,

$$\rho_{32} = A_{12}A_{31} \frac{2\sigma_{11} - (\sigma_{33} + \sigma_{22})}{\tau^{-1}\tau_{32}^{-1} + |A_{13}|^2}.$$
(7)

The numerator in (7) tends to zero as the microwave power increases. It is exactly zero when $\sigma_{33} - \sigma_{11}$ = $-(\sigma_{22} - \sigma_{11})$, i.e., after the initial population difference on the 2 - 1 transition is inverted. Using the expression for $\sigma_{11} - \sigma_{33}$ and $\sigma_{11} - \sigma_{22}$, we can readily show with the aid of (2) that the numerator in (7) vanishes when

$$|A_{13}|^{2} = \frac{\sigma_{33}^{\circ} - \sigma_{11}^{\circ} + \sigma_{22}^{\circ} - \sigma_{11}^{\circ}}{2T\tau[\sigma_{33}^{\circ} - \sigma_{11}^{\circ} - 2(\sigma_{22}^{\circ} - \sigma_{11}^{\circ})]},$$
(8)

which is possible only for $\sigma_{33}^0 - \sigma_{11}^0 > 2(\sigma_{22}^0 - \sigma_{11}^0)$. The above essential difference between this effect and the radiofrequency saturation in ordinary magnetic resonance is thus confirmed and is explained through the analysis of (7).

Figures 3c and 4b and c show the calculated shapes of the coherence signal, based on (5), for the two cases of detection. The first corresponds to the slow variation of the modulation frequency at constant microwave field frequency (Fig. 3c). The detuning of the frequency of the microwave magnetic field from resonance was, in this case, taken to be $\Omega_{31} - \omega_{31} = 2\pi \times 5$ kHz. Figure 4b shows curves characterizing the coherence signal for the second method of detection in which the frequency of the microwave magnetic field, Ω_{31} , was varied, whereas the modulation frequency was fixed and equal to the resonance frequency, $\Omega_{32} = \omega_{32}$ (Fig. 4b corresponds to P =1, 0.54, and 0.06, in relative units). Figure 4c shows the results for the same method of detection at fixed detuning $(\Omega_{32} - \omega_{32})/2\pi = 115$ Hz. The curves in Fig. 4c correspond to microwave power P=1, 0.54, and 0.06, respectively.

The calculations were carried out for the following values of the parameters: $(\sigma_{22}^0 - \sigma_{11}^0)/(\sigma_{33}^0 - \sigma_{11}^0) = 0.48$; $H_{\rm max} = 2 \times 10^{-4}$ Oe (this corresponds to P = 1); $\tau_{32} = 620$ μ sec; $\tau = 156 \ \mu$ sec; $T = 4 \$ msec. These parameters were chosen on the basis of the following considerations. We measured the relaxation times T_1 and T_2 corresponding to magnetic resonance on the Zeeman sublevels of the cesium atoms. The relaxation time τ_{32} was assumed equal to T_2 , and the relaxation time T equal to T_1 .

$$\frac{1}{T_2} = \frac{1}{T_2} + \frac{1}{T_2 \sinh}$$
(9)

where T_{21}^{-1} and T_{2inh}^{-1} characterize the contributions of light relaxation and magnetic-field inhomogeneity to the transverse relaxation time (the intrinsic relaxation time T_{2int} was not taken into account because $T_{2int} \gg T_{21}$, T_{2inh}). The quantity τ was calculated from the formula

$$\frac{1}{\tau} = \frac{1}{T_{2l}} + \frac{7}{T_{2inh}} \qquad (10)$$

The field H_{max} (maximum value of the amplitude of the alternating magnetic field in the cell) was estimated from measured microwave power. Although the Doppler effect for the hyperfine structure transitions was not suppressed under our experimental conditions, the size of the working cell with its paraffin coating was greater than the wavelength of the incident radiation, ^[8] and so the influence of this effect on the width and shape of the coherence lines was not taken into account in the calculation. This was justified as follows.

1. The coherence signal recorded by slowly varying the modulation frequency (Fig. 3) should not, in principle, depend on the Doppler broadening. In fact, the Doppler effect produces the same shift for the frequencies ω_{31} and ω_{21} , so that the resonance conditions $\Omega_{31} - \Omega_{21} \approx \omega_{32}$ are satisfied for all the atoms simultaneously and independently of their velocity.

2. The influence of Doppler broadening on the shape of coherence signals recorded by varying the frequency of the microwave magnetic field at constant modulation frequency should be appreciable only for curves whose width is small in comparison with the Doppler broadening. Since the calculated Doppler width under our experimental conditions is 9 kHz, the distortion of the line shape is expected only at low microwave power, i.e., small P in Figs. 4b and c. The dip in the center is absent under these conditions and, therefore, the Doppler effect should produce simply a line broadening. Another possible phenomenon is a change in the shape of the dip at the center of the lines corresponding to high microwave power.

We have thus observed for the first time the coherent superposition of states corresponding to the Zeeman sublevels of optically oriented atoms during the excitation of hyperfine structure transitions by amplitudemodulated microwave magnetic field.

We have observed coherent signals both for magnetic sublevels with $\Delta m_F = 1$ (F = 4, $m_F = 4$ and F = 4, $m_F = 3$) and with $\Delta m_F = 2$ (F = 4, $m_F = 4$ and F = 4, $m_F = 2$).

We have used two methods for recording these signals, namely, variation of the frequency of the microwave field and variation of the modulation frequency. Essentially different experimental curves were obtained in these two cases. The influence of different experimental conditions on the shapes of these curves has been explained.

The phenomenon has been considered theoretically, and the results of these theoretical calculations are in agreement with experimental data.

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Translated by S. Chomet