MODULATION EFFECTS IN OPTICALLY ORIENTED METASTABLE He⁴

V. S. KOROL' and A. N. KOZLOV

Institute of Automation, Kirghiz Academy of Sciences; Institute of Terrestrial Magnetism, Ionosphere, and Radiowave Propagation, USSR Academy of Sciences

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The spectra of the absorption and first harmonic signals of the M_Z component of the magnetization of optically oriented He⁴ were recorded using 100% modulation (at 330 Hz) of the rf field amplitude. The spectra of the side signals were investigated as a function of the rf field amplitude H₁, modulation field, and relaxation time of the absorption cell. The experimental results were interpreted by generalizing the Primas solution to the M_Z component, which provided a satisfactory explanation of the behavior of the central and side bands at low amplitudes of H₁.

INTRODUCTION

THE method of high-frequency modulation of resonance conditions is used in NMR spectroscopy for the purpose of stabilization and measurement of the fields in magnet gaps.^[1-6] The theoretical basis of the modulation effects for the transverse M_x component of the magnetization and the corresponding mathematical calculations are given in ^[1-4]. Since the M_z component of the magnetization is not observed in classical NMR, Bloch's equation has not yet been solved for this component.

The method of optical orientation in alkali-metal vapors and in metastable helium makes it possible to record easily the effect caused by the M_Z component, using the change in light passing through an absorption cell. This problem is dealt with by Bell and Bloom.^[7] The effect of alternating magnetic fields on optically oriented atoms of alkali metals has been considered in several papers.^[8,9] In view of this, it seemed of interest to investigate the modulation effects of the M_Z component of the magnetization in the case of optically oriented metastable He⁴.

If we use small-depth modulation of one of the parameters (such as the static magnetic field H₀, the resonance frequency ω of the rf field, or the amplitude of this field H₁), we find that the spectrum of the M_Z signal contains side bands, which can be easily isolated when the condition $\Omega > \Delta \omega_h$ is satisfied (Ω is the modulation frequency and $\Delta \omega_h$ is the width of the magnetic resonance line).

According to [9-13], the appearance of the side bands can be treated as a resonance in an effective field

$H_e = \sqrt{(H_0 - \omega/\gamma)^2 + H_1^2},$

where γ is the gyromagnetic ratio. In these investigations, the mathematical description of the phenomenon is based on a double transformation of a system of Bloch's equations from the laboratory system of coordinates to coordinates rotating with the frequency of an independent oscillator ω and then to a system of coordinates rotating about the direction of H_e with a frequency Ω . However, this treatment does not provide a description of the central band of the spectrum.

We generalized Primas's solution, $^{[3]}$ making a single transformation of the system of coordinates to find the M_Z component, and the results were in full agreement

with the experimental data at low amplitudes of H_1 .

In all these cases the solution of Bloch's equations is approximate since these equations apply to ensembles containing nuclear and electron spins equal to $\frac{1}{2}$, which is not the case for optical orientation of alkali-metal atoms and metastable helium. However, the approximate solutions explain qualitatively, with certain restrictions, the physical nature of the processes.

EXPERIMENTAL DATA

Figure 1 is a block diagram of the apparatus for recording the line profiles of the absorption and first harmonic signals of oriented metastable helium atoms, by modulation of a static magnetic field $H_0 + H_m \cos \Omega t$.

The light from an electrodeless spectroscopic helium lamp 1 was passed through a polarizing system (an infrared Polaroid and a quarter-wave plate 2) and optically oriented the metastable helium atoms in a cell 4. The light transmitted by the cell was recorded by a radiation detector 6 of the FD-7K type. A discharge in the helium lamp and in the cell was excited by means of two high-frequency oscillators 7 and 8, placed at a distance of 4 m from the rest of the apparatus.

A resonance rf field H₁, amplitude-modulated to the extent of 100% at a low frequency of 330 Hz, was produced by a standard-signal generator 9 and rf coils 5, whose axes were perpendicular to the direction of the optical beam. The frequency of the standard generator was varied smoothly by a motor drive 10. Modulation coils 3, whose axes coincided with the direction of the optical beam, were supplied at a frequency $\Omega/2\pi = 50$ kHz



FIG. 1. Block diagram of liquid-helium apparatus for the observation of the fundamental absorption and first-harmonic signals of the central and side bands.

by means of a second standard signal generator 11. The radiation detector signal was measured using a highfrequency selective microvoltmeter 12 and a low-frequency spectrum analyzer 13. The isolated 330 Hz signals, produced by the selective microvoltmeter and the spectrum analyzer, were recorded alternately by an automatic recorder of the N-110 type (14).

The amplitudes of the rf field H_1 and of the modulation field H_m were determined by the parametric resonance method described by Novikov.^[13] The modulation coils were calibrated using apparatus for the observation of the magnetic resonance signal of optically oriented cesium atoms in a field of 0.14 Oe; the magnetic resonance frequency in this case was $\Omega/2\pi = 50$ kHz.

We shall designate the absorption signal and its harmonics using two subscripts: $S_{Z(\mu, n)}$, where μ is the serial number of the side band and n is the serial number of the harmonic. For example, the central absorption signal will be denoted by $S_{Z(0,0)}$.

Figure 2a shows the absorption signals of the side bands $\mu = \pm 2$, ± 1 and of the central signal for a constant modulation depth, which were recorded when the rf field H₁ was varied from 27 to 228 Γ ($\Gamma = 10^{-5}$ Oe). It is evident from Fig. 2a that the central absorption signal saturated rapidly; the saturation of the side lines occurred at much higher values of H₁.

Figure 2b shows the dependences of the absorption signal intensities on the modulation coefficient $\beta = \gamma H_m/\Omega$. The rf field, used to obtain the results shown, was relatively weak in order to avoid appreciable saturation. It was clear that in H_1 = const the width of the central signal decreased while the widths of the side bands increased.

The traces obtained at the modulation frequency Ω (Figs. 3a and 3b) were of considerable interest; as far as the authors are aware these are the first such records obtained. The traces shown in Fig. 3a were re-

corded in a constant modulation field but for various values of H₁. It follows from these traces that the field H₁ affected strongly the width and intensity of the side lines. The first harmonic of Ω at the position of the central absorption line was completely absent. Figure 3b shows traces of the recorded line profile of the first harmonic of Ω in a constant rf field H₁ when β was varied. When the modulation coefficient was increased, the signal rapidly reached saturation.

Comparison of the traces of the absorption lines and of the first harmonic indicated that the first harmonic of the side lines was already saturated at low values of $\beta = 0.1-0.2$, while the signal of the fundamental harmonic became saturated much later. A slight shift of the centers of the side lines, in the direction of the central absorption line, was observed when H₁ was increased.

We investigated the dependence of the intensity of the signal of the first harmonic of Ω on the effective longitudinal relaxation time τ_1 , which is shown in Fig. 4. We assumed that the optimum time was equal to the relaxation time of an absorption cell with $\tau_1 \approx 0.4$ msec. We found that use of absorption cells with lower values of τ_1 reduced the absorption signal as well as the first harmonic of the side line but the rate of reduction of the first-harmonic signal (line 1) was half the rate of reduction of the absorption signal (line 2). We should point out that when the deviation, $\Delta \omega$, of the frequency from the resonance value was reduced in a constant field H₁, the first-harmonic signal increased.

THEORY AND COMPARISON WITH EXPERIMENT

The theoretical interpretation of the dependences obtained will be based on the solution of Bloch's equations for the case of modulation of resonance conditions, described by Primas.^[3] Primas's solution will be gen-





FIG. 2. a) Traces of the absoption signals of the central $(\mu = 0)$ and side $(\mu = \pm 1, \pm 2)$ bands as a function of the rf field intensity H_1 ; $\tilde{S}_2(0, \pm 1; \pm 2) =$ $f(\Delta \omega, H_1), H_m = 1000 \Gamma$, $\gamma H_m/\Omega = 0.56, \Omega = 50 \text{ kHz}$ (He⁴). b) Traces of the line profile for a constant amplitude of H_1 and various values of the modulation field H_m ; $\tilde{S}_2(0, \pm 1; \pm 2) = f(\Delta \omega, H_m), H_1 = 54 \Gamma$; $\Omega = 50 \text{ kc}$ (He⁴).



FIG. 3. Traces of the profiles of the first-harmonics of the side bands: a) at a constant amplitude of H_m and various values of H₁, $\tilde{S}_{z}(\pm 1, 1) = f(\Delta \omega, H_1)$, H_m = 333 Γ , $\gamma H_m/\Omega = 0.17$, $\Omega =$ 50 kc(He⁴); b) at a constant amplitude of H₁ and various values of H_m, $\tilde{S}_{z}(\pm 1, 1) = f(\Delta \omega, H_m)$, H H₁ = 80 Γ , $\Omega =$ 50 kc (He⁴).

where

$$A = \cos n\Omega t + n\Omega \tau_1 \sin n\Omega t, B = \sin n\Omega t - n\Omega \tau_1 \cos n\Omega t,$$

 $\frac{\gamma H_m}{\Omega}$

n n9

0.17

0.28

0.37

0.56

$$K = \gamma^2 H_1^2 \tau_1 \tau_2 \sum_{\alpha = -\infty}^{\infty} J_{\alpha}^2 / [1 + (\Delta \omega + \alpha \Omega)^2 \tau_2^2]$$

 m_0/N_0 is the rate of orientation; N_0 is the number of active atoms; P_Z is the probability of orientation of atoms along the z direction; τ_2 is the effective transverse relaxation time; $J_{\mu+n}$, J_{μ} , J_{∞} are Bessel's functions of the first kind with an argument $\beta = \gamma H_m/\Omega$.

The expression (4) is obtained on the assumption that the time-dependent terms in $M_{\rm Z}$ can be omitted. However, this does not reduce appreciably the value of $M_{\rm Z}$ or $S_{\rm Z}.$

Under the experimental conditions, we can isolate a single term in the product of two series of Eq. (4) and this term has definite values of the serial number of the side band μ and the serial number of the harmonic n. To achieve this, experimental conditions are established under which a given term in the series has the strongest intensity. Then the series in α is replaced by a single term for which $\alpha = \mu$ and $(1 + K)^{-1}$ can be placed under the double summation sign. Taking these considerations into account, we shall write the expressions for the fundamental and first harmonics of the central signal and the first side bands, replacing, for the sake of simplicity, the constant term $M_0P_Zm_0/N_0$ with C, so that only the variable components depending on the detuning $\Delta \omega$ are retained:

the absorption signal of the central band ($\mu = 0, n = 0$) is

$$\tilde{S}_{z(0,0)} = C \frac{\gamma^2 H_1 \cdot \tau_1 \tau_2 J_0^2}{1 + (\Delta \omega \tau_2)^2 + \gamma^2 H_1^2 \cdot \tau_1 \tau_2 J_0^2};$$
(5)

the signal of the first harmonic of the central band $(\mu = 0, n = 1)$ is

 $\widetilde{S}_{z}^{(1,1)}, \widetilde{S}_{z}^{(-1,0)},$ $\overset{\mu V}{\overset{\eta V}}}}{\overset{\eta V}{\overset{\eta V}{$

eralized to find the M_Z component using Bloch's equations in a rotating system of coordinates:

$$\vec{F} + [1/\tau_2 + j\Delta\omega(t)]F = -\gamma H_1 M_z, \qquad (1)$$

$$\dot{M}_z + (M_z - M_0) / \tau_1 = \gamma H_1 V;$$
 (2)

Here, $\Delta \omega(t) = \Delta \omega + \Delta \omega_m \cos \Omega t$, $\mathbf{F} = \mathbf{V} + \mathbf{jU}$.

The solution of Eqs. (1) and (2) is sought in the form of series

$$F = \sum_{n = -\infty}^{\infty} F_n \exp(jn\Omega t) = \sum_{n = -\infty}^{\infty} (V_n + jU_n) \exp(jn\Omega t),$$
$$M_z = \sum_{n = -\infty}^{\infty} M_{z_n} \exp(jn\Omega t).$$

Integration of Eq. (2) gives the expression

$$M_{z} = M_{0} + \gamma H_{1} \tau_{1} \sum_{n=-\infty}^{\infty} \left[V_{n} \frac{\exp(jn\Omega t)}{1 + jn\Omega\tau_{1}} + V_{-n} \frac{\exp(jn\Omega t)}{1 - jn\Omega\tau_{1}} \right].$$
(3)

Using the expression, obtained by Bell and Bloom,^[7] for the "longitudinal signal" proportional to the M_Z component of the magnetization, we find that our relationship for the modulation under resonance conditions is of the form

$$S_{z} = N_{0}P_{z} - M_{0}P_{z} \frac{M_{0}}{N_{0}} \left\{ 1 - \frac{\gamma^{2}H_{1}^{2}\tau_{1}\tau_{2}}{1+K} \sum_{\mu=-\infty}^{\infty} \sum_{n=0}^{\infty} J_{\mu} \frac{(J_{\mu+n} + J_{\mu-n})A + j(J_{\mu+n} - J_{\mu-n})B}{[1 + (\Delta\omega + \mu\Omega)^{2}\tau_{2}^{2}][1 + (n\Omega\tau_{1})^{2}]} \right\}$$
(4)

$$S_{z(0, 1)} = C \frac{2j J_0 J_1 \gamma^2 H_1^2 \tau_1^2 \tau_2 \Omega \cos \Omega t}{(1 + \Omega^2 \tau_1^2) [1 + (\Delta \omega \tau_3)^2 + \gamma^2 H_1^2 \tau_1 \tau_2 J_0^2]};$$
(6)

the absorption signal of the first side band (μ = -1, n = 0) is

$$S_{z(-1,0)} = C \frac{\gamma^2 H_1^2 \tau_1 \tau_2 J_1^2}{1 + (\Delta \omega - \Omega)^2 \tau_2^2 + \gamma^2 H_1^2 \tau_1 \tau_2 J_1^2};$$
(7)

the signal of the first harmonic of the first side band ($\mu = -1$, n = 1) is

$$S_{z(-1.1)} = C\gamma^2 H_1^2 \tau_1 \tau_2 \Omega J_1 \frac{(J_0 + J_2) \sin \Omega t - j(J_0 - J_2) \cos \Omega t}{(1 + \Omega^2 \tau_1^2) [1 + (\Delta \omega - \Omega)^2 \tau_2^2 + \gamma^2 H_1^2 \tau_1 \tau_2 J_1^2]} . (8)$$

In the derivation of these formulas, we have included terms with the factor $\Omega \tau_1 \gg 1$ in the expressions for A and B of Eq. (4). In our case, assuming $\tau_1 = \tau_2 \approx 10^{-3} - 10^{-4}$ sec and $\Omega = 2\pi \times 5 \times 10^4$, we find that $\Omega \tau_1 \ge 10^2$.

When the modulation coefficient β is increased, the lifting of the saturation of the central line (Fig. 2b) and of the saturation of the side bands is due to the presence of different Bessel's functions J_0 and J_1 in the term $\gamma^2 H_1^2 \tau_1 \tau_2 J_{\mu}$ in Eqs. (5) and (7). When the value of β is increased, J_0 decreases but J_1 increases. The dependence $S_{Z} = f(H_{1}, \Delta \omega)$ for $\beta = const$ (Fig. 2a) shows that the saturation of the central and side bands depends strongly on H₁. The first harmonic of the first side band should be approximately two orders of magnitude smaller, i.e., $\tau_1\Omega$ weaker than the intensity of the absorption signal, which is in agreement with the experimental data. As mentioned earlier, the saturation of the first harmonic of the first side bands $S_{Z(\pm 1,1)}$, observed when the modulation field is increased, is more rapid than the saturation of the central absorption line.

When $\beta \le 0.5$, the Bessel's functions can be reduced to $J_0 = 1$, $J_1 = \beta/2$, $J_2 = 0$. In this case, the expression for the first harmonic of the side band is given by

$$S_{z(-1,1)} = C \frac{j\gamma^2 H_1^2 \tau_2 \exp(j\Omega t)\beta}{\tau_1 \Omega \left[1 + (\Delta \omega - \Omega)^2 \tau_2^2 + \gamma^2 H_1^2 \tau_1 \tau_2 \beta^2\right]}.$$
 (9)

The presence of the term $\tau_1\Omega$ in the denominator explains qualitatively the different behavior of the experimentally recorded central absorption signal and of the first harmonic of the side band (Fig. 4) when considered as a function of τ_1 .

CONCLUSIONS

1. Experiments carried out on metastable helium are generally, in agreement with the theory of resonance in an effective field. When the rf field H_1 is increased, a shift of the side bands toward the central absorption line is observed and this shift follows from the definition of the rf field modulation frequency

$$\Omega = \pm [\Delta \omega^2 + (\gamma H_1)^2]^{\frac{1}{2}}.$$

The phases of the first-harmonic signals of the upper and lower side bands differ by 180° .

2 The expressions for S_Z in the case $\Delta\omega/\gamma\approx 10H_1,$ found by a single transformation to a rotating system of coordinates, explain satisfactorily such phenomena as the lifting of the saturation of the central absorption line, as well as the increase of the first-harmonic signals of the first side bands when H₁ and τ_1 are varied.

3. The first harmonic of the first side band can be attributed to an interaction of the optical beam with the transverse moment of the forced precession of spins M_{Xe} about the direction of the effective field H_e . An increase of the M_{Xe} signal is proportional to an increase of the angle θ between the direction of the field H_0 and the effective field H_e . The rotation of H_e can be compared with the rotation of the optical axis of a singlebeam magnetometer about the direction of H_0 during observation of the signal S_X .

4. These experiments suggest the feasibility of a self oscillation magnetometer intended primarily for the measurement of alternating magnetic fields in a wide range of frequencies (up to 2–3 kHz), in which the feedback is provided by the low-frequency channel Ω .

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