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OPTICALLY ALIGNED Rb VAPOR
HYPERFINE STRUCTURES OF PARAMAGNETIC RESONANCE AND POPULATION DISTRIBUTION STUDY OF OPTICALLY ALIGNED Rb VAPOR

BY

Shigeru ANDO

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ABSTRACT

All Zeeman transitions ($\Delta F=0$, $\Delta m_F=\pm 1$) of ground state $^2S_{1/2}$ were well separated in the "very weak field range" on optically aligned Rb vapor. Their frequencies showed very good agreements with Breit-Rabi's formula.

The population distribution of optically aligned Rb vapor was roughly interpreted by the rate equations without considering the spin-exchange.

The competition between optical alignment and spin-exchange was confirmed on Rb^{87} by observing that which is dominant depends on the cell temperature.

Introduction

Dehmelt's optical pumping principle is featured by the observation of the circularly polarized D_1 radiation which has transmitted through the alkali-metal vapor. The paramagnetic resonance absorption of the optically aligned alkali-metal vapor had been observed and well discussed by W.E. Bell, and A.L. Bloom[1] on Na and K. They reported the separation of the resonance absorptions corresponding to $F=1$ and $F=2$ of Na. Recently, all six Zeeman transitions ($\Delta F=0$, $\Delta m_F=\pm 1$) were well separated, and the negative absorptions in $F=1$ state were observed on Na at 15 gauss by L.W. Anderson and A.T. Ramsey[2]. Two methods for observing the spin-relaxation of the optically aligned atoms have been developed, the field reversal method by Dehmelt[3] and the light interrupting method by Franzen[4]. Anderson and Ramsey[2] made precise measurement of the spin-relaxation time on Na vapor by these two methods. They assumed the competition between optical alignment and spin-exchange for explanation of the negative absorption in $F=1$ levels of Na.

This work, in succession to the previous papers[5],[6], was made for examining the agreement between the observed frequencies of Zeeman transitions ($\Delta F=0$, $\Delta m_F=\pm 1$) and the Breit-Rabi's formula in the magnetic field range from 0.5 to 20 gauss on Rb^{87} and Rb^{85} respectively. All six Zeeman transitions of Rb^{87} and ten of Rb^{85} were well resolved in the range of higher magnetic field and the discrepancies were within the

experimental error.

The intensity distribution of these Zeeman transitions ($\Delta F=0$, $\Delta m_F=\pm 1$) was observed to depend largely on the cell temperature, and explained by solving the rate equations of respective Zeeman sublevels.

The negative absorptions of paramagnetic resonance were observed for the stationary state of populations in each Zeeman sublevel corresponding to the lower F of $^2S_{1/2}$ state on both Rb^{87} and Rb^{85}. The existence of the critical temperature for negative absorption of Rb^{87} was also observed in the sense that the negative absorption took place below this temperature while the positive one above it. It seems to be an obvious evidence of the competition assumed by Anderson and Ramsey. The relation between the measured spin-relaxation time T_1 and the critical temperature will be discussed.

Experimental Apparatus

The schematic diagram of the apparatus is shown in Fig. 1. The optical system is a "Z-beam system" named by Bell and Bloom[1], in which the magnetic field is applied

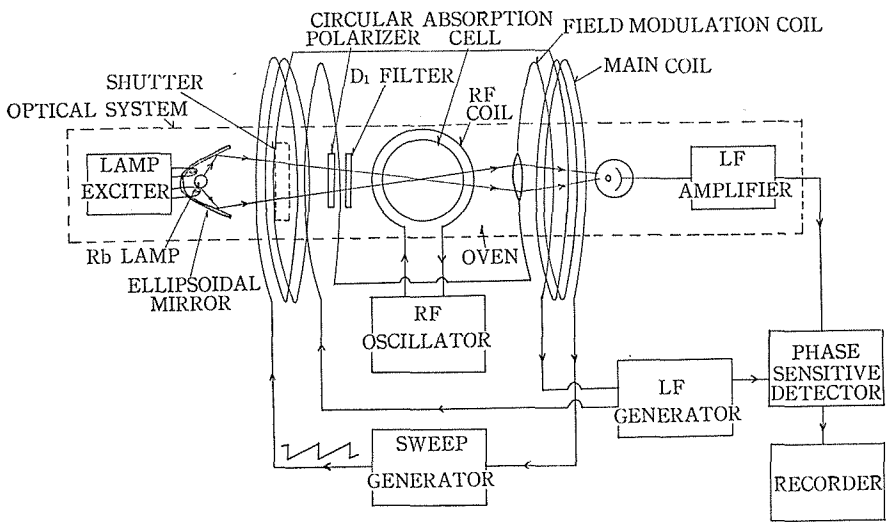


Fig. 1. Schematic diagram of the apparatus.

in the direction of the optical axis. The Rb lamp is an 1 cm diameter glass cell containing a small amount of Rb metal and 1.6 mmHg Kr gas. In order to avoid the self-absorption and to get the sharp spectral line, the Rb lamp is excited by a pair of rf coils at 100 MC/S. It emits the radiation as shown in Fig. 2 with a D_1 interference filter for the elimination of D_2 radiation. The circular polarizer employed in the experiment is a compound type[7] which is adjustable to an arbitrary wavelength. It con-

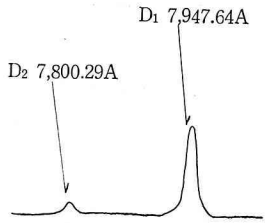


Fig. 2. Spectrum of exciting radiation.

sists of a linear polarizer and two quarter-wave retarders of 140 mμ and 210 mμ retardations respectively. By adjusting their mutual angles, the degree of circular polarization was made to be within 0.3% deflection against the analyzer rotation of 360°. As the photodetector, a series of silicon solar cells was employed for the observation of resonance absorption and a 7102 photomultiplier for the relaxation time measurement. These two photo-detectors have their sensitivity maxima near the Rb D_1 line, 7,948A. As can be seen from Fig. 3, three pairs of Helmholtz coils, two for compensating

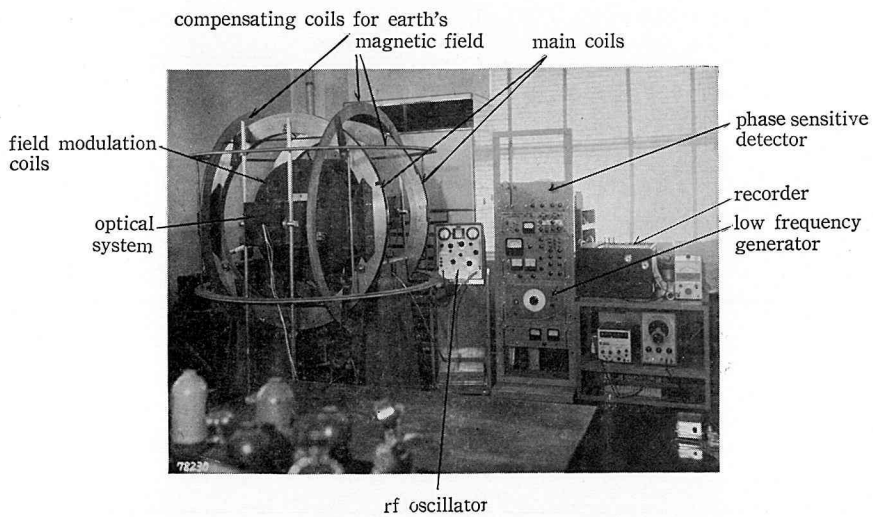


Fig. 3. Experimental apparatus.

earth's magnetic field and one for producing an arbitrary field *Ho* in the direction of optical axis, were used. The pair of Helmholtz coils, 1 meter in diameter for producing field *Ho* was estimated by calculation to have the field inhomogeneity of 1×10^{-4} through 5 cm range right in the center along the coil axis. In fact the minimum half-width of the observed resonance absorption curves was about 1.1×10^{-4} *Ho* at the reduced intensity of rf field. Theoretically the limit of the half-width is of the order of 10^{-6} *Ho* in this experiment as the reciprocal of the relaxation time T_1, though it depends

on the field inhomogeneity. The direct current of *Ho* coils is supplied by a sweep generator which has the current stability within 3×10^{-5} in ten minutes. As shown in Fig. 1, for the recording of the resonance absorption, the current is swept in a sawtooth waveform by the sweep generator and the magnetic field *Ho* is modulated within the half-width of each absorption curve by a pair of field modulation coils. The frequency of the field modulation is 9.5 C/S at which the amplified photodetector signal is demodulated by the phase sensitive detector. Thus, the resonance derivative signal is recorded against the intensity of magnetic field near the resonance point at a slow speed compared with T_1. For the Franzen type measurement[4] of the spin relaxation time T_1, a photographic shutter on the radiation path is used. The shutter is reconstructed so that the radiation can be interrupted in an arbitrary interval of time and the measurement of T_1 can be made in the dark condition.

Theory and the experimental results

The theoretical frequencies of Zeeman transitions ($\Delta F=0$, $\Delta m_F=\pm1$) are calculated from Breit-Rabi's formula[8] in which the intermediate coupling of total angular momentum J and nuclear spin I is assumed. They can be written as follows,

$$\nu_{m,m-1}=-\frac{1}{h}\cdot\frac{\mu_I}{I}\cdot H\pm\frac{\Delta\nu}{2}\left\{\sqrt{1+\frac{4m}{2I+1}x+x^2}-\sqrt{1+\frac{4(m-1)}{2I+1}x+x^2}\right\},$$

where the plus sign is for the higher hyperfine quantum number F and the minus sign for the lower F. In Fig. 4, the results of the calculation, in which the values of Table I were adopted, are shown graphically. The corresponding observed

Table I

	I	μ_I	$10^{-6}\cdot\Delta\nu$
Rb^{87}	$\frac{3}{2}$	2.749	6,834.1
Rb^{85}	$\frac{5}{2}$	1.349	3,035.7

frequencies are given in the same figure. Discrepancies are within the experimental error which principally depends on the fluctuation of external magnetic field and the current instability. In the case of indoor measurements, where *Ho*=4.28, 6.43, 14.3 and 21.4 gauss, the fluctuation of external field was about 100 γ gauss in ten minutes. In the case of open space measurements, where *Ho*=0.6 gauss, the earth's magnetic field was used as *Ho* which fluctuated up to about 20 γ gauss in ten minutes.

The population distribution of $^2S_{1/2}$ Zeeman sublevels of optically aligned Rb^{87} atoms can be explained by the set of rate equations;

$$\frac{dn_{F,m}}{dt} = -AP_{F,m}n_{F,m} + \frac{1}{8}A\sum_{F,m}P_{F,m}n_{F,m} - \frac{n_{F,m} - \frac{N}{8}}{T_1},$$

where $n_{F,m}$ represents the population per unit volume at (F, m) sublevel, $N=\sum_{F,m} n_{F,m}$, $P_{F,m}$ is the probability for the transition $({}^2S_{1/2}, F, m \rightarrow {}^2P_{1/2})$ and A is the light intensity. Several assumptions are necessary for these rate equations such that the alkali-metal vapor in the cell is optically thin, that atoms excited to ${}^2P_{1/2}$ state are equally distributed to each sublevels by collision mixing and that the term related to spin-exchange collision is very small. Solving these equations under the condition of stationary state $\frac{dn_{F,m}}{dt} = 0$, the relative populations shown in Table II

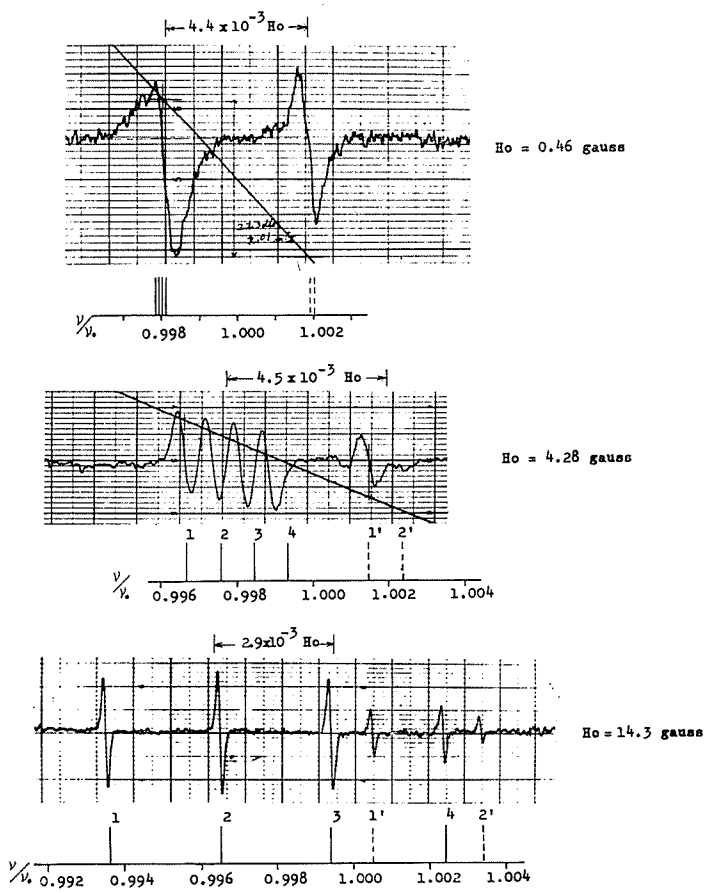


Fig. 4(a). Rb^{87} (a vacuum cell 5 cm in diameter and eicosane coated) Hyperfine structures of Zeeman transitions in the ground state of Rb both observed and calculated (below), solid lines corresponding to higher F levels and dotted lines to lower F levels.

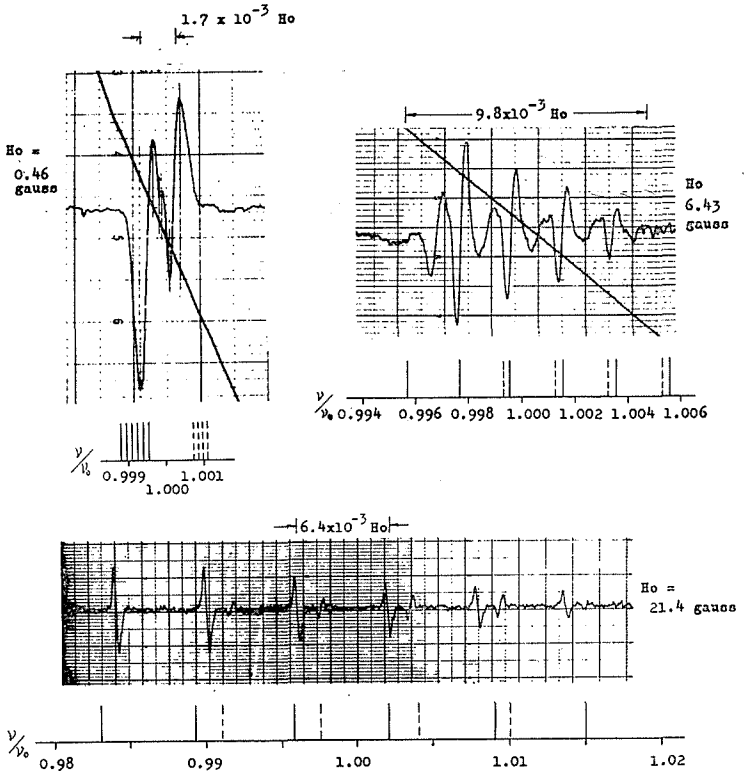


Fig. 4(b). Rb^{85} (a vacuum cell 5 cm in diameter and eicosane coated for $Ho = 0.46$ and 6.43 gauss, a 10 cm diameter cell containing 100 mm Hg Ne for $Ho = 21.4$ gauss, difference between conditions of the cells being not observed).

Table II

	m_F	$P_{F,m}$ for $(^2S_{1/2} \rightarrow {}^2P_{1/2})$	relative population	product of Δn and transition probability ($F=0$, $\Delta m_F=-1$)				*
				$ApT_1=0.70$	$ApT_1=0.53$	$ApT_1=0.30$	$ApT_1=0.115$	
$F=2$	2	0	1	1.00	1.00	1.00	1.00	1
	1	p	$\frac{1}{ApT_1+1}$	0.63	0.74	0.95	1.22	2
	0	$2p$	$\frac{1}{2ApT_1+1}$	0.35	0.51	0.66	1.02	3
	−1	$3p$	$\frac{1}{3ApT_1+1}$	0.14	0.20	0.32	0.57	4
$^2S_{1/2}$	−2	$4p$	$\frac{1}{4ApT_1+1}$					
				0.21	0.25	0.32	0.41	1′
$F=1$	−1	p	$\frac{1}{ApT_1+1}$	0.12	0.17	0.22	0.34	2′
	0	$2p$	$\frac{1}{2ApT_1+1}$					
	1	$3p$	$\frac{1}{3ApT_1+1}$					

numbers noted in the * column are refered to Fig. 4

were obtained in the case of Rb^{87}.

It is supposed that the observed intensity of the resonance absorption corresponding to Zeeman transition is proportional to the product of the population difference Δn and the probability of Zeeman transition ($\Delta F=0$, $\Delta m_F=\pm 1$), because $\partial I/\partial(\Delta n)$, where I is the intensity of the transmitted radiation, can be written as follows,

$$\frac{\partial I}{\partial(\Delta n)} \propto (P_i - P_{i-1}),$$

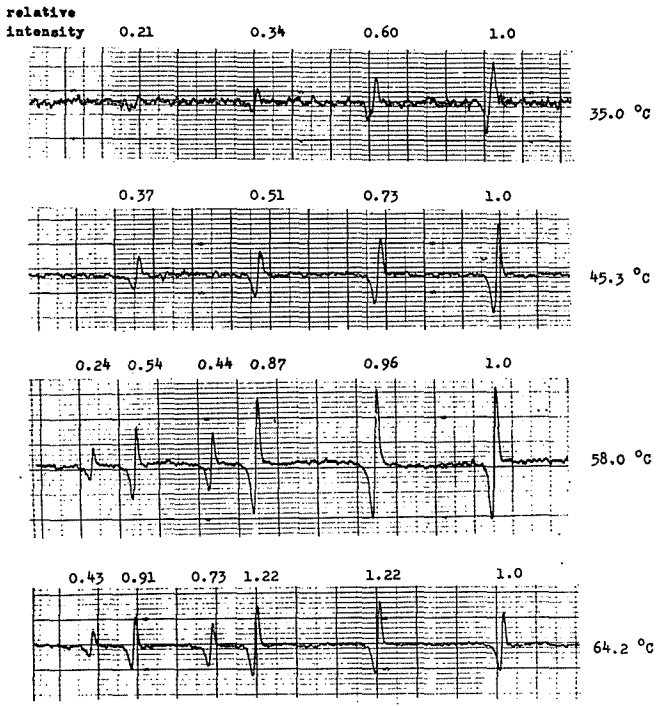
where $(P_i - P_{i-1})$ is constant for the transition ($^2S_{1/2} \rightarrow {}^2P_{1/2}$). In Table II, the values of these products are tabulated against the values of ApT_1, where p is the normalization factor for the transition probability. Fig. 5 shows the observed intensity distributions of resonance derivatives near the frequency of 10 MC/S, in the cases of Rb^{87} and Rb^{85}. In the data, the cell temperature is taken for the parameter, and the total population N varies so far as ten times with the temperature change of about 20 degree Kelvin. In general, p decreases when such an extreme increase in N takes place. Thus, it is possible to make the parameter ApT_1 in Table II correspond to the cell temperature indicated in Fig. 5. From comparison of table II with Fig. 5, although there are slight discrepancies, it is likely that the rate equations are able to explain the population distributions of the optically aligned atoms. Another evidence is given by Fig. 6, which shows the reversal of the distribution of resonance absorptions by that of the selection rule.

The negative absorptions of the paramagnetic resonance of optically aligned atomic vapor were originally observed at $F=1$ Zeeman sublevels of $^2S_{1/2}$ on Na by Anderson and Ramsey[2]. In their experiments, two types of observation were made. One is that the negative absorptions were observed at a stationary state by sweeping the field Ho very slowly compared with T_1. The other is that the negative absorptions could be observed only at the rapid sweep of Ho compared with T_1. In order to explain these results, they solved the rate equations containing spin-exchange term in the case of low radiation intensity. As a result the population difference δ between $F=1$ Zeeman sublevels was:

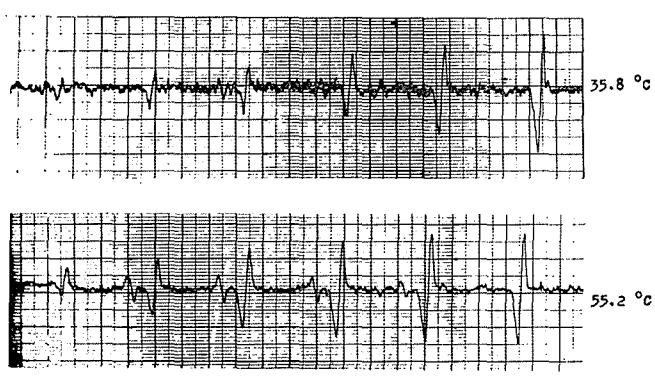
$$\delta = \frac{\dfrac{32}{T_1^2} + \dfrac{5.4}{T_1 T_2} - \dfrac{3.8}{T_2^2}}{\left(\dfrac{8}{T_1} + \dfrac{2.78}{T_2}\right)\left(\dfrac{32}{T_1^2} + \dfrac{16.5}{T_1 T_2}\right)} A,$$

where $T_2 = \dfrac{1}{(Nv\sigma)_{av.}}$, v is the relative velocity between atoms and σ is the cross section for the spin-exchange collision. The time T_2 can be measured as the sweep time corresponding to the half-width of negative absorption in the case of rapid sweep.

In the present experiment, both T_1 and T_2 were observed to decrease with the increase of the cell temperature. T_1 depends largely on the conditions of cell making



(a) Data on Rb^{87} (a vacuum cell 5 cm in diameter and eicosane coated, Ho=14.3 gauss)



(b) Data on Rb^{85} (a 10 cm diameter cell containing 100 mm Hg Ne, Ho=21.4 gauss)

Fig. 5. Intensity distributions of resonance derivatives at various values of cell temperature.

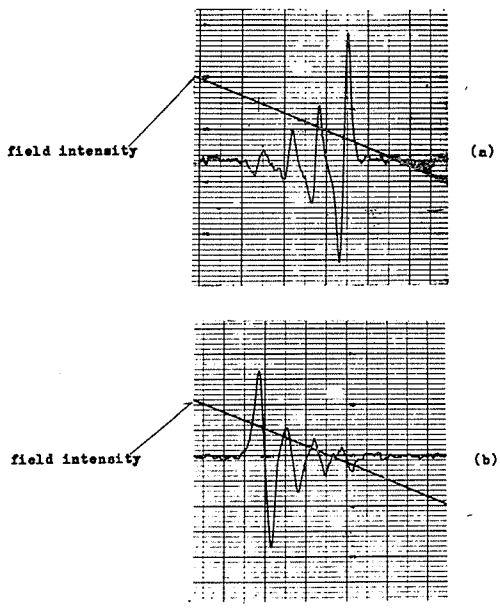


Fig. 6. Reversal of intensity distribution of hyperfine structures in $^2S_{1/2}$, $F=1$ state of Rb^{87} with the reversal of field direction, the correspondences to the selection rules being shown by (a) to $\Delta m_F=+1$ and (b) to $\Delta m_F=-1$ (a vacuum cell 5 cm in diameter and eicosane coated, $Ho=4.28$ gauss).

such as impurities, wall coating and so on. In the case of Rb^{87}, as described in the earlier part of the paper, the existence of the critical temperature of negative absorption was observed for both vacuum and buffer gas contained cells. Taking $\delta=0$ in the above equation, $T_1=T_2/3.7$ was derived. In the case of Fig. 7, where the critical temperature was 45°C and $T_1=30$ m sec., $T_2=8$ m sec. was obtained. This result shows a rough agreement with the direct measurement of T_2. In the case of Fig. 7, where rf field intensity is very large so that the resonance absorptions in $F=2$ state are saturated, the population differences corresponding to the negative absorptions in $F=1$ are supposed to be very small compared with $F=2$ state.

Conclusion

The hyperfine structures of Zeeman transitions of the ground state of Rb^{87} and Rb^{85} were observed to show good agreement with Breit-Rabi's formula in the magnetic field range from 0.5 to 20 gauss.

The population distributions of optically aligned Rb vapor are roughly inter-

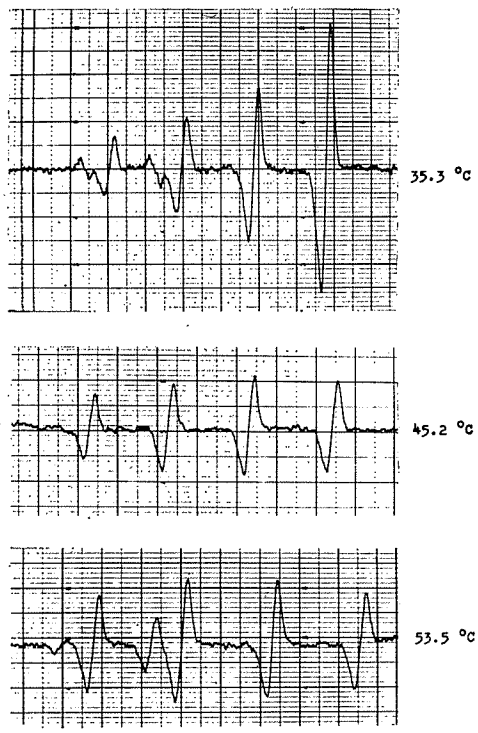


Fig. 7. Critical temperature for negative absorption on Rb^{87} (a vacuum cell 10 cm in diameter and eicosane coated, $Ho=$ 14.3 gauss).

preted by the rate equations without consideration of the spin-exchange. In order to explain the population distributions in the lower F state, it is necessary to consider the spin-exchange. The theory of the competition between optical alignment and spin-exchange originated by Anderson and Ramsey was confirmed by the existence of the critical temperature of the negative absorption.

There are, however, many unsolved problems. The rate equations should be solved under more complete conditions, such as comparatively high intensity of D_1 radiation and consideration of the spin-exchange term, the differences in related characteristics between buffer gas filled cell and vacuum cell should be clarified, the values of T_1 of Rb^{87} and Rb^{85} should be observed individually, and the cross section of the spin-exchange between Rb^{87} and Rb^{85} should be measured.

Acknowledgements

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