Vol. A43 (1973) ACTA PHYSICA POLONICA No 3

OFF-DIAGONAL ELEMENT PHASES OF OPTICAL ABSORPTION MATRIX FOR ZEEMAN FINE AND HYPERFINE COMPONENTS OF MODULATED ABSORPTION IN ALKALIES WITH $I = 3/2$, 5/2 AND 7/2 *

BY S. BAUCH AND K. ROSIŃSKI

Institute of Physics, Polish Academy of Sciences, Warsaw**

(Received May 29, 1972)

The distribution of phases over the modulated Zeeman fine and hyperfine components of the radiation absorbed by the alkali vapour optically pumped and exposed to resonance magnetic field is investigated. The modulation phases of the fine components are stated to be opposite to one another. The light beam containing both fine components D_1 and D_2, the modulation phase and relative amplitude depend on the D_1-to-D_2 intensity ratio in the incident radiation and on the cell temperature. An improved modulation detection scheme for the experiment with lined resonance cell, one light beam containing D_1 and D_2 components and oblique to the static magnetic field was designed and tested. The results of the investigation can be of help in the quantitative interpretation of modulation experiments with one as well as both D_1 and D_2 fine components.

1. Introduction

Some time ago Dehmelt [1] discovered and Bell and Bloom [2] have investigated the transversal ("cross") beam modulation in the experiment of longitudinal (along a static magnetic field) optical pumping and magnetic resonance in the ground state of alkalies. Since that time the modulation phenomenon has found many important applications as a source of information on some parameters of the atomic system under study [3] and in designing magnetometers for weak fields [4]. However the phenomenon itself has not been completely investigated as yet. For instance in many cases the modulation experiment is carried out with one light beam-pumping and also modulated-oblique to the static magnetic field and with the resonance cell lined with *e. g.* paraffin [5]. In this case the pumping and detection are usually realized with the whole resonance light containing both fine components. As it is well known, pumping in the analysed case is more effective but the detection of the magnetic polarization is not.

---

* Work done under Polish Academy of Sciences Contract Nr PAN — 3.1021.

** Address: Instytut Fizyki PAN, Al. Lotników 32-46, 02-668 Warszawa, Poland.

The questions arise: do not the modulation components D_1 and D_2 compensate each other? Are the phases of the modulated fine components equal or different? Next, generalization of the problem to hyperfine components is suggested. This generalization should be considered as a problem of considerable importance because all spectral lines engaged in modulation experiments show a hyperfine structure. If there are any phase differences among the hyperfine components, the amplitude of the modulation depends on the geometry of resonance cell and on its temperature in a complex way, absorption coefficient being different for various hyperfine components.

This paper deals with the theoretical and experimental solution of the problem just stated (multiple quantum transitions being taken into account) [12]. The theoretical solution of the problem is to be found by analysing the signs of the off-diagonal optical absorption matrix elements for the Zeeman hyperfine components of the D_1 and D_2 lines.

The problem of the phase distribution over the modulated hyperfine components in the case of level structure similar to that of odd mercury isotopes, is mentioned by Cohen-Tannoudji [3]. The modulation has been reported there to disappear in the case of the excitation of broad-line (in relation to hyperfine structure) type. The analysis of the physical meaning of the quantity of radiation absorbed by the alkalies, revealed by Bouchiat [6] contains the implicit suggestion of disappearance of the modulation effect in the light beam with broad-line type distribution in relation to fine structure. Up to now such a suggestion has not been investigated explicitly nor exploited [4], [7], [8].

2. *Theoretical*

As it may be concluded on the ground of the general theoretical description of optical pumping [9] applied to the alkali metal vapour [10] the absorption of resonance light L_A depends on the atomic density matrix $\varrho_{\mu\mu'}$ of the ground state and on the optical absorption matrix A. Experimental conditions can be chosen to make ϱ independent of the excited state structure. However the absorption matrix does depend on the structure of excited state. In fact it can be presented as a sum over single elementary contributions corresponding to the Zeeman sublevels of the fine and hyperfine structures of the excited state. As a result the intensity of the absorbed radiation is found to be a sum over contributions corresponding to the excited state sublevels:

$$L_A = \sum_{\substack{\mu\mu' \\ J_eF_e}} l_{\mu\mu'}^{J_eF_e}, \text{ where } l_{\mu\mu'}^{J_eF_e} = \frac{1}{T_p} A_{\mu\mu'}^{J_eF_e} \varrho_{\mu'\mu} \text{ and } A_{\mu\mu'}^{J_eF_e} = \sum_m a_{\mu\mu'}^m,$$

$$\text{further:} \quad a_{\mu\mu'}^m = \langle\mu|\vec{e}_{\lambda_0}\vec{D}|m\rangle\,\langle m|\vec{e}^{\,*}_{\lambda_0}\vec{D}|\mu'\rangle =$$

$$= \sum C(J_gIF_g;\, \mu_J\mu-\mu_J)\,\langle\mu_J|\vec{e}_{\lambda_0}\vec{D}|m_J\rangle C(J_eIF_e;\, m_Jm-m_J)\times$$

$$\times C(J_eIF_e;\, m'_Jm-m'_J)\,\langle m'_J|\vec{e}^{\,*}_{\lambda_0}\vec{D}|\mu'_J\rangle C(J_gIF_g;\, \mu'_J\mu'-\mu'_J),$$

moreover $\mu-\mu_J = m-m_J$ and $\mu'-\mu'_J = m'-m'_J$, C means the Clebsch-Gordan coefficient, $\vec{e}_{\lambda_0}$ — polarization unit vector of modulated light beam, $\vec{D}$ — electric dipole transition operator, T_p^{-1} — a quantity proportional to the intensity of the incident light beam practically independent of the excited state structure [6]. We are interested only in the Zeeman (not hf with $\Delta F \neq 0$) modulation components with the frequency $(\mu-\mu')\omega_{rf}$ described by off-diagonal terms $A_{\mu\mu'}^{J_eF_e}\varrho_{\mu'\mu}$. Assuming the ground state magnetic quantum numbers μ and μ' constant we should observe still a few simple modulation components with the same frequency corresponding to the Zeeman fine and hyperfine sublevels of the excited state and having phases defined by the sign of $a_{\mu'\mu}^{m}$. The matrix elements $a_{\mu'\mu}^{m}$ (see Table I) have been calculated [12] for the physical situation of the alkali metal vapour being optically pumped by a σ^+-polarized light beam (Fig. 1). The quantum numbers assumed are: $J_g = 1/2$, $I = 3/2$, $5/2$ and $7/2$, $J_e = 1/2$, $3/2$. The calculations have been performed for the static magnetic fields H_0 sufficiently high to resolve well the magnetic resonance structure but leaving F_g in the ground state still a good quantum number, which occurs in a rather broad region of weak magnetic fields in view of large hyperfine splitting. H_0 being already an intermediate field for the excited state, appropriate eigenfunctions should be used [11]. It was assumed, in general, that the modulation is caused by the magnetic resonance in the ground state between the sublevels with $\mu = F_2 (F_2 > F_1)$ and $\mu' = \mu-1$ — the modulation with the radio-frequency ω_{rf}, or $\mu = F_2$ and $\mu' = \mu-2$ — the modulation with doubled radio-frequency $2\omega_{rf}$. For other pairs of μ and μ', results are similar. Circular polarization of the modulated beam was usually adopted except for $|\Delta m| = 2$, when linear polarization $\vec{j}$ was admitted. The numbers in Table I mean the relative amplitudes and the signs + and — opposite phases. In Table I the sum is also given of the amplitudes over hyperfine components for fixed fine structure component. The following facts may be easily stated on simple inspection of Table I: 1. For modulation at ω_{rf} ($|\Delta m| = 1$), the phases of hyperfine components are opposite and the sum over one fine component does not disappear, but the sum over both fine components gives zero, in agreement with the general conclusion to be drawn from paper [6]; 2. For modulation at $2\omega_{rf}$ ($|\Delta m| = 2$), the sum of the hyperfine contributions within one fine component cancels out, in agreement with the known facts (*e. g.* [13]). It is evident that this modulation may be observed, in general, only by using single hyperfine components (*e. g.* by means of a tunable dye laser), especially in the case of the component D_1 with splitting of the excited state hyperfine structure greater than the splitting of the component D_2. The modulation may be detected by circular as well as linear polarization of light.

3. *Experimental*

The experimental verification of some theoretical results for ^{85}Rb was based on an experiment with "crossed light beams" [2]. Rubidium vapour was pumped by the σ^+ polarized light beam incident in the direction of static magnetic field ($H_0 = 4.5 \div 22$ Oe) and the polarization S_z was created. The fine component D_1 of the pumping radiation was selected by means of a dielectric interference filter. The rubidium vapour was contained in a spherical bulb (∅ 10 cm) filled with Ar at 11 Torr, as buffer gas. Detection of

TABLE I. The values of $a^{m}_{\mu\mu'}$

Ground state					Excited state																	Radiation polarization	Remarks
J	I	F	μ	μ'	J	(F,m)															$\sum_{Fm} a^{m}_{\mu\mu'}$		
						(0,0)	(1,0)	(1,1)	(2,0)	(2,1)	(2,2)	(3,0)	(3,1)	(3,2)	(3,3)	(4,2)	(4,3)	(4,4)	(5,3)	(5,4)			
1/2	3/2	2	2	1	1/2			0,76		-0,24	0,48										1,00	$-\frac{1}{\sqrt{2}}(\vec{j}+i\vec{k})$	D_1
1/2	3/2	2	2	1	3/2			0,15		-0,25	0,50		-0,40	-1,00							-1,00		D_2
1/2	3/2	2	1	0	1/2		0,61	0,31		0,31											1,23		D_1
1/2	3/2	2	1	0	3/2	0,00	0,12	0,07		0,29		-0,73	-0,98								-1,23		D_2
1/2	5/2	3	3	2	1/2						0,85			-0,35	0,50						1,00		D_1
1/2	5/2	3	3	2	3/2						0,25			-0,42	0,65	-0,33	-1,15				-1,00		D_2
1/2	7/2	4	4	3	1/2										0,88		-0,40	0,52			1,00		D_1
1/2	7/2	4	4	3	3/2										0,29		-0,54	0,71	-0,26	-1,20	-1,00		D_2
1/2	3/2	2	2	0	1/2			0,15		-0,15											0,00		D_1
1/2	3/2	2	2	0	3/2			0,04		-0,15			0,11								0,00		D_2
1/2	3/2	1	1	-1	1/2		-0,06		0,06												0,00		D_1
1/2	3/2	1	1	-1	3/2	0,25	-0,31		0,06			0,00									0,00		D_2
1/2	3/2	2	2	0	1/2			0,30		-0,30											0,00	$\vec{j}$	D_1
1/2	3/2	2	2	0	3/2			0,08		-0,30			0,22								0,00	$\vec{j}$	D_2

the modulated fine component in the transverse parallel ("cross") beam (detection beam) with the polarization $-\frac{1}{\sqrt{2}}(\vec{j}+i\vec{k})$, was realized with photomultiplier (M12FD35, Zeiss, Jena) and by using frequency changing and lock-in detection on an intermediate frequency [14]. The phase measurement was carried out on the dispersion component of the magnetic resonance signal by means of a phase shifter plugged into the circuit of the reference tension (at an intermediate frequency). The intensity of the cross beam was weak enough not to perturb the atomic system. The conditions of the phase measurement were optimal. The phase of the modulation was measured as the filters serving to the selection of the fine components D_1 or D_2 were set into the light beam successively: a dielectric interference filter to select D_1 and a liquid filter with a neodymium salt solution, to select D_2. In all cases the useless component of the detection beam did not exceed 1%. In agreement with the theory, the phases of the D_1 and D_2 components appeared to be opposite (Fig. 2b, c).

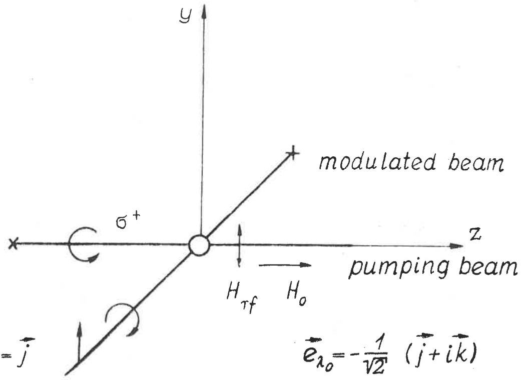

Fig. 1. The geometry and the polarization of crossed light beams

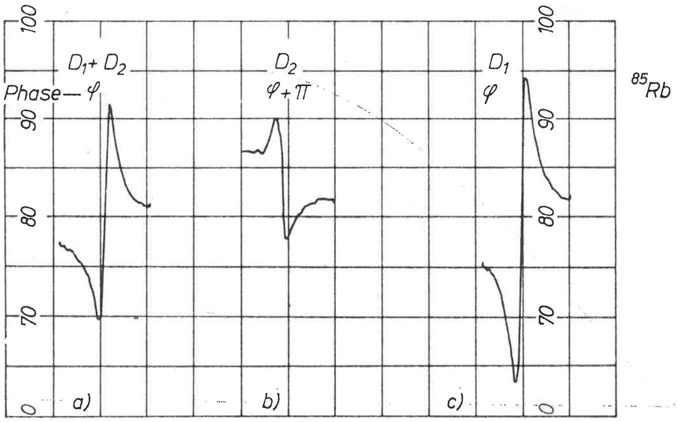

Fig. 2. The dispersion component of the magnetic resonance at 2 MHz. Phases for D_1, D_2 and D_1+D_2 are φ and $\varphi+\pi$ respectively

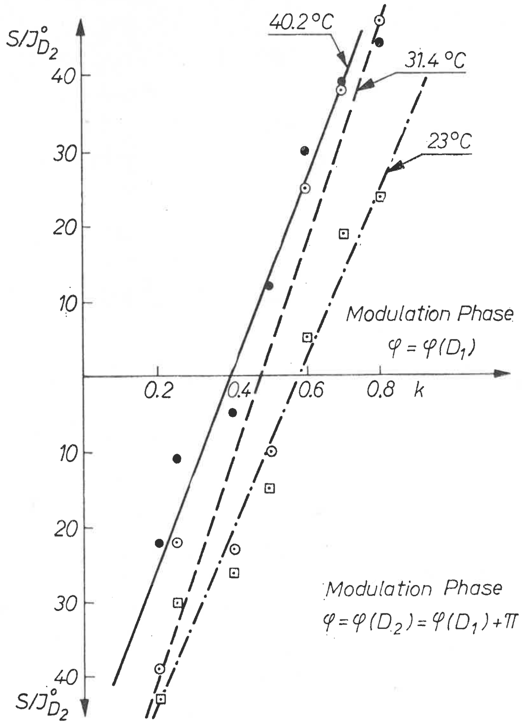

Fig. 3. $S/J^0_{D_2} \equiv \Delta S/I_{02}$ plotted as the function of $k = I_{01}/I_{02}$

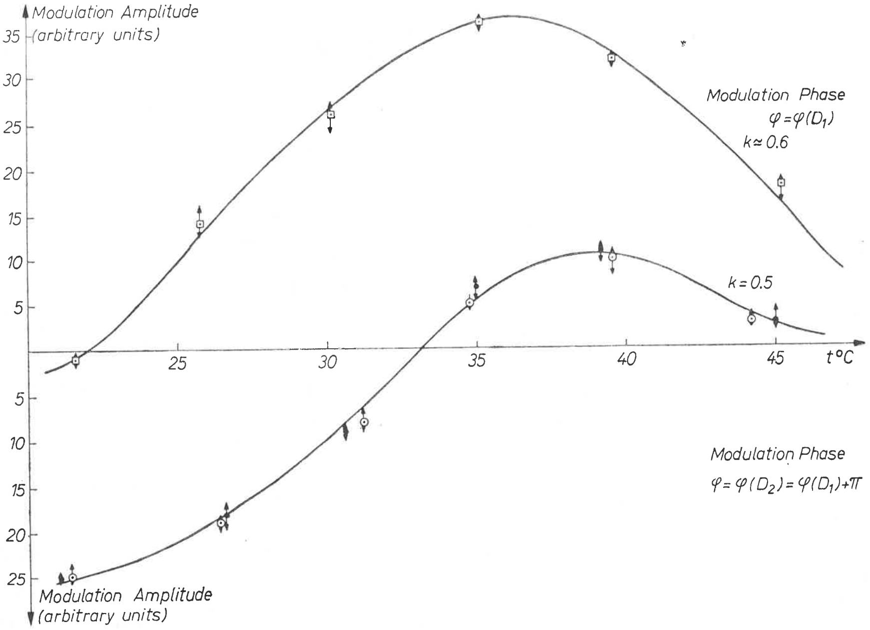

Fig. 4. Modulation amplitude plotted as the function of temperature

The next experiment was carried out with both components acting simultaneously. Though in the incident radiation the intensity of the D_2 component was greater than D_1 (after measurements with a monochromator), the modulation with the phase of the D_1 component was stated (Fig. 2a). This result must be considered to be a direct evidence of greater absorption of the D_2 component leading to the dominance of the D_1 component in the light emerging from the resonance cell. As the signal is the difference between the signal due to the D_1 and D_2 alone, the resulting signal has the phase of the D_1 component. The possible, in general, influence of the photomultiplier sensitivity distribution need not be taken into account because the S1 type photocatode sensitivity curve is rather flat in the spectral region (7800–7950 Å) under interest.

In order to investigate in some detail the relations occurring here, another modulation experiment was performed. In the incident beam the D_1-to-D_2 intensity ratio was changed. To do this the advantage was taken of the peak transmission of metal interference filter being dependent on the incidence angle. Of course a filter of appropriate wave length must be taken. It may be easily shown that for a simple fine component i the maximum value S_i^m of the modulation signal in the resonance is proportional, at constant temperature, to the incident light beam intensity I_{0i} and to the magnetic polarization of atomic system $\langle S_z \rangle$. Thus the relation holds: $S_i^m = c_i I_{0i} \langle S_z \rangle$, where c_i and $\langle S_z \rangle$ are, in general, functions of the temperature. The opposite signs of the D_1 and D_2 components of the modulation taken into account, the following formula holds for the measured signal ΔS: $\Delta S = S_1 - S_2 = I_{02} \langle S_z \rangle (c_1 - c_2 k)$, where $k = I_{01}/I_{02}$.

It follows that $\Delta S/I_{02} = \langle S_z \rangle (c_1 - c_2 k)$. Hence keeping the temperature constant, we expect $\Delta S/I_{02}$ to be a linear function of the D_1-to-D_2 intensity ratio k. k and I_{02} were measured with the aid of a monochromator. The experimental results being in rather good agreement with the above-given formula, are plotted in Fig. 3. The effect of the stronger D_2 absorption is revealed in the lack of modulation for $k = k_0 < 1$ but not for $k_0 = 1$. k_0 being the value of k for the case of lack of the modulation (phase inversion). On the ground of the experimental data $c_i \langle S_z \rangle$ in arbitrary units and the c_1 to c_2 ratio as function of the temperature can be measured. In a different experiment the temperature was changed, k being kept constant. It is evident that the relation for $\Delta S/I_{02}$ just given must hold here too, but k is a constant now. A characteristic feature of this experiment is the occurrence of the phase inversion of the modulation corresponding to a definite temperature. The inversion temperature decreases, when $k \to 1$ (Fig. 4) and in the same time the corresponding curves become more and more flat in the region of the inversion point. It may happen that there may be a lack of modulation in a quite broad, in practice, region of temperature. It is to be noticed that on the effect under study another one is superimposed, namely the well-known effect of the dependence of the polarization signal $\langle S_z \rangle$ on temperature. The polarization signal disappears in a high enough and low enough temperature reaching its maximum at an intermediate temperature.

A particular case of rather great practical importance is the experiment with one light beam oblique to the static magnetic field H_0 and with coated resonance cell. As it is well known, optical pumping is more effective, when both fine components D_1 and D_2 are used together. However, the detection of the Zeeman polarization in the absorbed light beam becomes less effective, if possible at all. In the real experimental conditions there is almost always a finite polarization signal due to the difference in absorption of the D_1 and D_2 components.

Concerning the detection of the modulation, the results of this paper point out that a compensation will always be present of both modulation components. The result of the compensation depends on the D_1-to-D_2 intensity ratio in the incident light beam and on the resonance cell parameters such as its temperature and length. It is easily seen now that insertion of a filter selecting the D_1 component into the beam emerging from the resonance cell, should increase the signal. Of course gain in the signal-to-noise ratio assured in this manner will depend on many features of the arrangement. In a preliminary experiment of this type realized with Cs, an increase of the signal-to-noise ratio equal to 4 was stated after the component D_1 had been selected in the detection light beam. In a more thorough investigation an analysis would be needed of effects of the D_2 component reflected back to the resonance cell by an interference filter[1]. It seems that the reflected D_2 component should rather increase the pumping effect, its angular momentum being probably unchanged by reflection. Experiments with oblique light beam are continued.

The authors would like to thank Professor T. Skaliński for his interest shown in the course of this work and Mrs K. Krajewska-Kolwas for her help in some measurements.

REFERENCES

[1] H. G. Dehmelt, *Phys. Rev.*, **105**, 1924 (1957).

[2] W. E. Bell, A. L. Bloom, *Phys. Rev.*, **107**, 1559 (1957).

[3] C. Cohen-Tannoudji, *Ann. Phys.* (France), **7**, 469 (1962).

[4] P. A. Grivet, L. Malnar, *Measurement of Weak Magnetic Fields by Magnetic Resonance*, in: *Adv. in Electronics and Electron Physics*, Academic Press Inc., New York 1967.

[5] Takahiro Kubo, *Japan J. Appl. Phys.*, **9**, 1521 (1970).

[6] M. A. Bouchiat, *J. Phys.* (France), **26**, 415 (1965).

[7] L. N. Novikov, *Priborostroyenie i Tekhnika Eksperimenta*, Nr 4, 121 (1966).

[8] Takahiro Kubo, to be published in *Appl. Optics*, July 1972.

[9] J. P. Barrat, C. Cohen-Tannoudji, *J. Phys. Rad.*, **22**, 329, 443 (1961).

[10] K. Rosiński, *Acta Phys. Polon.*, **31**, 107 (1967).

[11] J. Kapelewski, K. Rosiński, *Acta Phys. Polon.*, **28**, 177 (1965).

[12] K. Rosiński, S. Bauch, *Bull. Acad. Polon. Sci. Ser. Sci. Math. Astron. Phys.*, **18**, 625 (1970); K. Rosiński, *Magnetic Resonance and Related Phenomena*, Proc. XVI Congress AMPERE, Bucharest 1970, p. 420.

[13] W. Robotta, A. Steudel, H. Walter, *Optical Pumping and Atomic Line Shape*, Proc. Conference OPaLS, Warsaw 1969, p. 369.

[14] K. Rosiński, *Acta Phys. Polon.*, **31**, 173 (1967).

[15] J. Brossel, in: *Optique et Electronique Quantiques*, New York 1964, p. 189.

---

[1] Private remark of Dr Rozwadowski.