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LEVEL CROSSING WITH OPTICAL PUMPING IN POTASSIUM

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Level crossing in the first excited $4^2P_{3/2}$ state of potassium (^{39}K) is analysed. An improvement in measurements of level crossing signals using simultaneous optical pumping is discussed. The experimental results are demonstrated.

Introduction

A change in the angular distribution of resonance fluorescence of atoms is observed when the magnetic levels cross (*i.e.* there is a degeneracy) in an excited state. This effect was first observed in a zero field by Hanle [1] and by Colegrove *et al.* in a non-zero field [2]. The magnetic field strength at which this phenomenon of level crossing takes place depends on the hyperfine (or fine) structure. Thus, the level crossing technique has become a useful method for investigating the structure of the excited states of atoms. The accuracy of the results depends on the ratio of the distance of levels in a zero field to their natural width. In the case of Hg, Cd and Zn, which have long life times of the resonance states (longer than 10^{-7}s), narrow and well separated level crossing signals were obtained [3]–[7]. The magnetic field strengths at which these crossing occurred were exactly measured; thus, the hyperfine structure constants were determined with a high degree of accuracy.

For alkali metal atoms the situation is less favourable, because the life time of the first excited $^2P_{3/2}$ state is of the order of 10^{-8}s. The level crossing signal width then amounts to several oersteds. Moreover, the hyperfine splitting of such elements as sodium and potassium is small and thus some of the level crossing signals overlap. In addition, there are signals which are situated on the zero-field level crossing (Hanle) signal. The determination of the crossing point for even a single signal is difficult in this case. Nevertheless, a rather accurate determination of the hyperfine structure constants in Na [8] and K [9] by the level crossing method has recently been reported. The procedure was as follows: the theoretical curves giving the resonance fluorescence for a given geometry as a function of the applied magnetic field strength were computed. Both the magnetic dipole coupling constant a and electric quadrupole coupling constant b were treated as parameters. The theoretical curve

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giving the best fit to the experimental data was then used to determine the hyperfine coupling constants. In the case of potassium, considerable improvement in the accuracy was obtained. The previous results, using the atomic beam magnetic resonance method, were: $a = 5.7\pm\pm0.3$ Mc/s, $b = 2.8\pm0.8$ Mc/s [10]. The new results are: $a = 6.0\pm0.1$ Mc/s, $b = 2.9\pm0.1$ Mc/s. Thus, even without accurate knowledge of the position of the level crossing points, it was possible by analysing the global properties of the resonance fluorescence curve to get a determination of the hyperfine coupling constants.

Another, more direct approach, is to determine the position of the level crossing signals more exactly improving the experimental technique. This was done for sodium by a new technique, namely, by observing the photons emitted from the longest lived Na atoms. By this means, narrower and better resolved level crossing signals were obtained [11].

Change of the intensity ratio of the overlapping signals offers yet another approach to this problem. This can be achieved in level crossing experiment by utilizing optical pumping, which leads to a selective population of the ground state levels [12], [13]. The latter method is described here for the case of potassium.

General discussion

The first excited $4^2P_{3/2}$ state of potassium (^{39}K, $I = 3/2$) was analysed. The magnetic field values corresponding to the level crossing points were theoretically evaluated by solving the eigen-problem:

$$\mathcal{H}|\psi\rangle = E|\psi\rangle. \tag{1}$$

The Hamiltonian $\mathcal{H}$ includes two interactions: the hyperfine interaction between the nuclear and electronic angular momentum, and the interaction of both angular momenta with the applied magnetic field. The first interaction is described by the following expression

$$\mathcal{H}_{hf} = a\,\boldsymbol{I}\cdot\boldsymbol{J}+b\,\frac{(\boldsymbol{I}\cdot\boldsymbol{J})^2+3/2\,(\boldsymbol{I}\cdot\boldsymbol{J})-\boldsymbol{I}^2\boldsymbol{J}^2}{2I(2I-1)J(2J-1)} \tag{2}$$

where a is the magnetic dipole coupling constant, b is the electric quadrupole coupling constant, and I and J are the nuclear and alectronic angular momenta, respectively.

The magnetic (Zeeman) interaction with a uniform field H directed along the z-axis has the form

$$\mathcal{H}_{\mathrm{zeem}} = g_J\mu_0 J_z H + g_I\mu_0 I_z H \tag{3}$$

where g_J and g_I are the electronic and the nuclear Lande g — factors, J_z and I_z are the z-components of the electronic and nuclear angular momenta, respectively, and μ_0 is the Bohr magneton. The second term in (3) will be neglected in subsequent calculations because in potassium $g_I \ll g_J$.

The crossing of levels occurs in fields for which the quantum number of the total angular momentum F ceases to be a good quantum number. The levels will be denoted by $(\mathcal{F}, m)$, where $\mathcal{F} \to F$ for $H \to 0$. The magnetic quantum number m is a constant, independent of the magnetic field strength ($m = m_F$ for $H \to 0$ and $m = m_I+m_J$ in a strong field).

The eigenfunctions $|\mathcal{F}, m\rangle$ may be written either as a linear combination of the eigenfunctions for a weak magnetic field, $|F, m_F\rangle$, or as a linear combination of the eigenfunctions for a strong field, $|I, m_I; J, m_J\rangle$, which is the approach used here. The solution of equation (1) is reduced to the solution of a set of equations for each value of the magnetic quantum number. The energies are found conventionally by solving the secular equations. With $x = g_J\mu_0 H$, we get:

for $m = 0$:

$$\begin{vmatrix} -\frac{9}{4}a+\frac{1}{4}b-\frac{3}{2}x-\mathrm{E} & \frac{3}{2}a-\frac{1}{2}b & \frac{1}{2}b & 0 \\ \frac{3}{2}a-\frac{1}{2}b & -\frac{1}{4}a+\frac{1}{4}b-\frac{1}{2}x-E & 2a & \frac{1}{2}b \\ \frac{1}{2}b & 2a & -\frac{1}{4}a+\frac{1}{4}b+\frac{1}{2}x-E & \frac{3}{2}a-\frac{1}{2}b \\ 0 & \frac{1}{2}b & \frac{3}{2}a-\frac{1}{2}b & -\frac{9}{4}a+\frac{1}{4}b+\frac{3}{2}x-E \end{vmatrix} = 0 \tag{4}$$

for $m = \pm 1$:

$$\begin{vmatrix} -\frac{3}{4}a-\frac{1}{4}b\pm\frac{3}{2}x-E & a\sqrt{3} & \frac{1}{2}b \\ a\sqrt{3} & -\frac{1}{4}a+\frac{1}{4}b\pm\frac{1}{2}x-E & a\sqrt{3} \\ \frac{1}{2}b & a\sqrt{3} & -\frac{3}{4}a-\frac{1}{4}b\mp\frac{1}{2}x-E \end{vmatrix} = 0 \tag{5}$$

for $m = \pm 2$:

$$\begin{vmatrix} \frac{3}{4}a-\frac{1}{4}b\pm\frac{1}{2}x-E & \frac{3}{2}a+\frac{1}{2}b \\ \frac{3}{2}a+\frac{1}{2}b & \frac{3}{4}a-\frac{1}{4}b\pm\frac{3}{2}x-E \end{vmatrix} = 0 \tag{6}$$

and for $m = \pm 3$:

$$\frac{9}{4}a+\frac{1}{4}b\pm\frac{3}{2}x-E=0. \tag{7}$$

The energy of the levels as a function of the x-parameter, which is proportional to the magnetic field strength H, is plotted in Fig. 1. Four level crossings with $\Delta m = 2$ are indicated by circles.

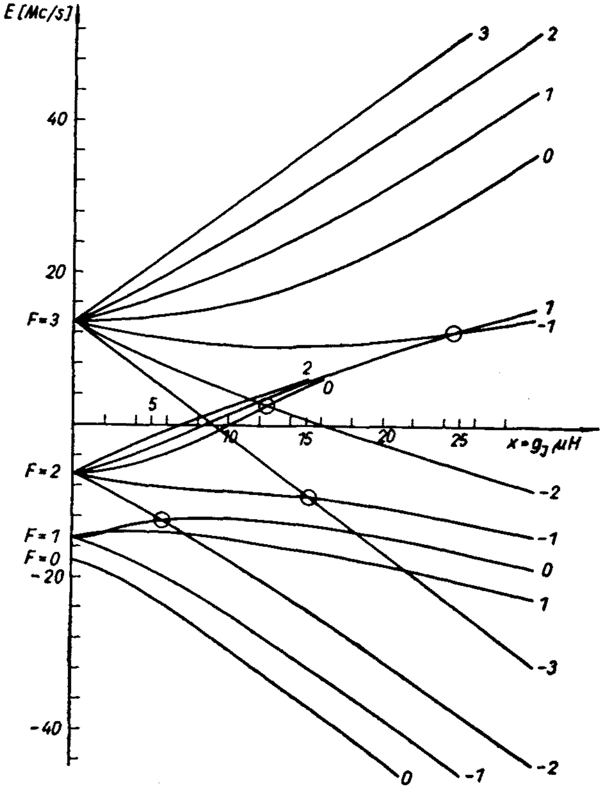

Fig. 1. Energy level diagram for the $4^2P_{3/2}$ state of ^{39}K

The relation between level crossing positions and the values of the constants a and b used in the calculation was also analysed. The calculations for $5.4 \leqslant a \leqslant 6.0$ Mc/s and $2.0 \leqslant b \leqslant 3.6$ Mc/s (for error limits given in [10]) were performed. The results are given in Figs 2, 3 and 4. The level crossing points shift toward higher magnetic fields with an increase of the magnetic dipole coupling constant a. This is due to the increase of hyperfine intervals in the zero field. The magnitude of the shift is different for each crossing; the smallest dependence is observed for the $(2, -2)(1, 0)$ crossing. On the other hand, this crossing strongly depends on the value of b. The dependence on b is quite different for each of the considered crossings. The electric quadrupole interaction irregularly "deforms" the hyperfine structure. The extremely slight dependence of the position of the $(3, -2)(2, 0)$ crossing on the value of b is to be noted.

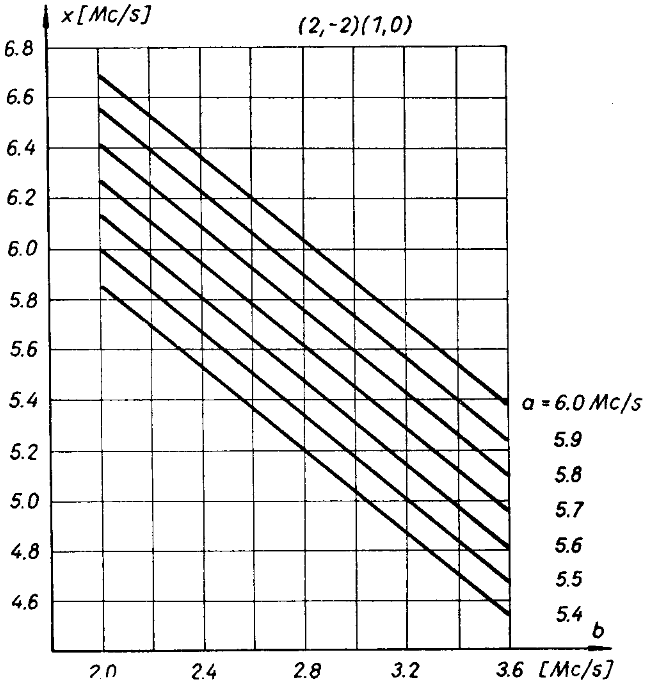

Fig. 2. The dependence of the level crossing (2,—2)(1,0) position on the values of the constants *a* and *b*

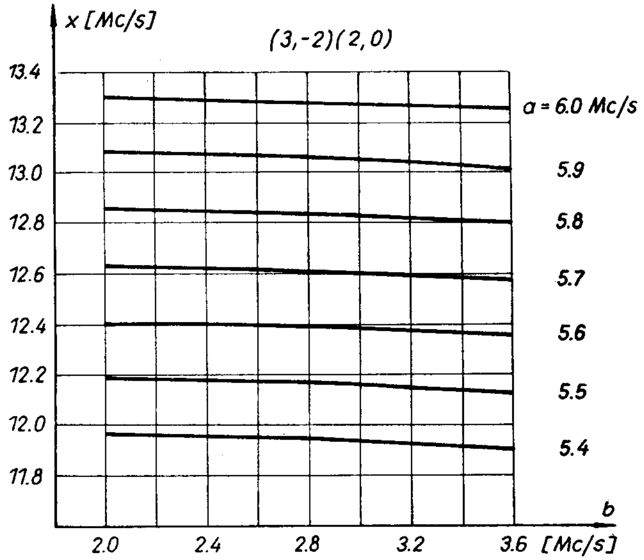

Fig. 3. The dependence of the level crossing (3, —2)(2,0) position on the values of the constants *a* and *b*

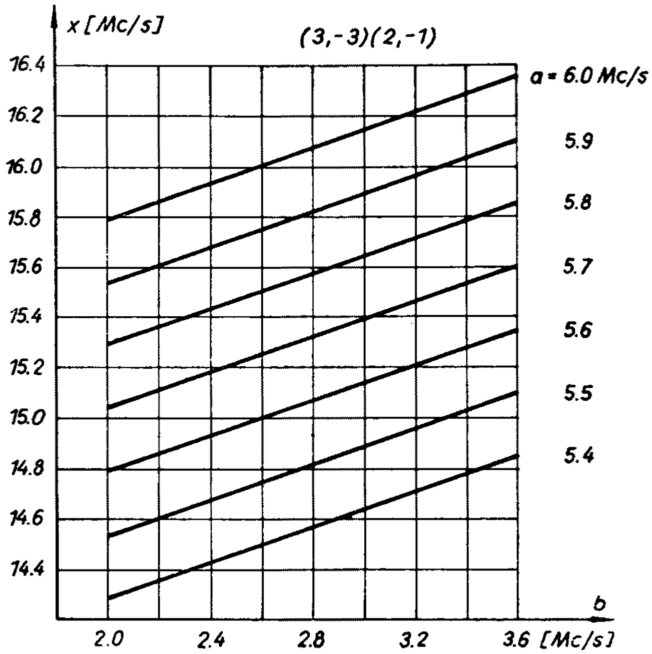

Fig. 4. The dependence of the level crossing (3, —3)(2, —1) position on the values of the constants *a* and *b*

For the (3, —3)(2, —1) crossing, the relation between the crossing position and the value of the constants *a* and *b* is particularly simple, *viz.*,

$$x_{(3,-3)(2,-1)} = \frac{5a(3a+b)}{6a+b}. \tag{8}$$

The magnetic field strengths for the level crossing points corresponding to $a = 5.7$ Mc/s, $b = 2.8$ Mc/s and $a = 6.0$ Mc/s, $b = 2.9$ Mc/s are given in Table I. The first crossing point, (2, —2)(1, 0), is situated at about 3 Oe. The life time τ of the $4^2P_{3/2}$ state of postasium is $(2.60 \pm 0.05) \times 10^{-8}$s [9]. The half width of the zero-field level crossing signal amounts to 6.5 Oe.

TABLE I

Magnetic field strength for level crossing ($\mathcal{F}$, *m*)($\mathcal{F}'$, *m'*). The hyperfine coupling constants used in the calculation

Level crossing ($\mathcal{F}$, *m*)($\mathcal{F}'$, *m'*)	H_{cross}[Oe]	
(2,−2)(1,0)	3,02 [1]	3.19 [2]
(3,−2)(2,0)	6.75 [1]	7.11 [2]
(3,−3)(2,−1)	8.21 [1]	8.63 [2]
(3,−1)(2,1)	13.21 [1]	13.86 [2]

[1] $a = 5.7$ Mc/s, $b = 2.8$ Mc/s;
[2] $a = 6.0$ Mc/s, $b = 2.9$ Mc/s.

Thus, the (2, —2)(1,0) signal is located at the slope of the Hanle signal and is relatively weak. The next two signals, (3, —2)(2, 0) and (3, —3)(2, —1), overlap and are unresolved. The predicted (3, —1)(2, 1) signal is more distant and less distinct because the corresponding levels cross under a small angle.

From the selection rules for optical transitions and the condition for coherent excitation in level crossing it follows that the contribution of crossing levels to the fluorescence is due to the absorption from only some of the magnetic levels of the ground state. In the case potassium (and other alkali metal atoms with nuclear spin $\frac{3}{2}$) the level crossing signals (2, —2)(1, 0) and (3, —2)(2, 0) are connected with the absorption from both the (2, —1) and the (1, —1) levels of the ground state. The (3, —3)(2, —1) signal is connected by absorption with the (2, —2) level only. A selective increase of the population of the ground state levels with the magnetic quantum numbers $\mu = -1$ or $\mu = -2$ should give an increase in the corresponding level crossing signals. Thus we should obtain either better separation of the examined signals from the zero-field level crossing signal or better resolution of the overlapping ones. This selective increase in population can be obtained by optical pumping.

The evaluation of the population of the ground state levels in potassium under optical pumping conditions was made by Hawkins' method [14]. The real ensemble of atoms was replaced by an idealized ensemble in which each atom absorbs the same number of photons N. Hawkins analysed optical pumping for sodium in a weak field. The expected populations of ground state levels for a given number of absorbed photons were calculated.

Similar calculations for potassium in a magnetic field of 7.5 Oe were performed (omitting, however, the fact that in the $4^2P_{3/2}$ state the levels $\mathcal{F} = 0$ and $\mathcal{F} = 1$ overlap). The field value we have chosen is intermediate with respect to the positions of the two overlapping level crossing which we have investigated by utilizing simultaneous optical pumping. Optical transition probabilities for the D_1 and D_2 resonance lines with $H = 7.5$ Oe were calculated and on this basis the probabilities of transitions from a given $(\mathcal{F}, \mu)$ level to all possible $(\mathcal{F}', \mu')$ levels as a result of the absorption and following re-emission of resonance radiation were evaluated. Equal incident D-lines intensities and uniform spectral intensity of the

TABLE II

Probabilities of transition, induced by σ^- radiation, between initial $(\mathcal{F}, \mu)$ and final $(\mathcal{F}', \mu')$ levels of the ground state of potassium at $H = 7.5$ Oe. The probabilities are normalized so that each initial state has a total probability equal unity. Only non-zero elements are indicated

$(\mathcal{F}, \mu)$ $(\mathcal{F}', \mu')$	(2,−2)	(2,−1)	(2,0)	(2,1)	(2,2)	(1,−1)	(1,01)	(1,1)
(2,−2)	1	0.343	0.166			0.250	0.093	
(2,−1)		0.371	0.156	0.157		0.287	0.084	0.038
(2,0)			0.219	0.135	0.082		0.214	0.044
(2,1)				0.224	0.157			0.132
(2,2)					0.378			
(1,−1)		0.287	0.244	0.109		0.463	0.100	0.014
(1,0)			0.215	0.243	0.112		0.508	0.051
(1,1)				0.132	0.271			0.721

source over the absorption profile of atoms were assumed. This is fulfilled for the lamp used in the experiment. Then the sum of the absorption probabilities from each ($\mathcal{F}$, μ) level is independent of $\mathcal{F}$ and μ. It was assumed that the excited atom is not subject to any disorientation process (the potassium vapour pressure was about 10^{-7} mm Hg, no buffer gas was present). After a life time τ, an atom returns to the ground state. The path of this return depends uniquely on the probabilities of optical transitions. Table II, similar to that given in [14], shows the extent to which the initial ($\mathcal{F}$, μ) level populates the final ($\mathcal{F}'$, μ') levels after σ^- absorption. Using this table, the expected populations $\sigma_{\mathcal{F},\mu;\mathcal{F},\mu}$ of an ensemble of atoms after absorption of N photons were calculated (Table III).

TABLE III

Expected values of population, $\sigma_{\mathcal{F},\mu;\mathcal{F},\mu}$, in the ground state of potassium at $H = 7.5$ Oe for ensembles, each of whose membres has scattered N photons. For $N = 0$ (no pumping) $\sigma_{\mathcal{F},\mu;\mathcal{F},\mu} = 0.125$ is assumed

N \ $\sigma_{\mathcal{F},\mu;\mathcal{F},\mu}$	2,–2; 2,–2	2,–1; 2,–1	2,0; 2,0	2,1; 2,1	2,2; 2,2	1,–1; 1,–1	1,0; 1,0	1,1; 1,1
1	0.231	0.137	0.087	0.064	0.047	0.152	0.141	0.140
2	0.344	0.135	0.068	0.040	0.018	0.154	0.119	0.122
3	0.451	0.126	0.052	0.028	0.007	0.145	0.093	0.098

A comparison of these populations with those obtained by Hawkins for a weak field demonstrates the smaller efficiency of optical pumping in stronger magnetic fields. This is due to a change in optical transition probabilities in a field of $H = 7.5$ Oe, primarily for the D_2 line. The transition probabilities for the D_1 line do not change noticeably. This is the result of the larger hyperfine splitting of the $4^2P_{1/2}$ state ($a = 28.85$ Mc/s [10]). From Table III one can conclude that a considerable increase in the (3, -3)(2, -1) level crossing signal will be obtained by optical pumping.

Experimental arrangement

The resonance cell was cylindrical, about 50 mm in diameter and 70 mm long. It was prepared by heating for several hours and simultaneous evacuation to a pressure of about 10^{-6} mm Hg. Next a small amount of potassium (of natural isotopic composition) was destilled into it and then the cell walls were covered with paraffin. The cell was put into a thermostatic chamber. The temperature was usually lover than 45°C and was kept constant.

An electrodeless discharge potassium vapour lamp was used as the light source. The emitted D lines had approximately equal intensities.

The steady magnetic field was created by a set of Helmholtz coils. The axis of the coils was parallel to the horizontal component of the Earth's magnetic field. The vertical component of the Earth's field was compensated by an additional pair of coils.

The Helmholtz coils were supplied by a storage battery. Most of the measurements were made as a function of the magnetic field strength. In order to provide a slow, continuous and approximately linear in time sweep of the magnetic field, an auxiliary supply source was employed. The magnetic field was calibrated by means of magnetic resonance in the ground state of potassium.

The resonance fluorescence in a direction perpendicular to the exciting light beam was detected by a photomultiplier (EMI, type 9558 B). The modulated photo-electric signal was fed to a phase-sensitive detector and recorded.

The preliminary measurements were made in the experimental set-up which contained a rotating polarizer [15]. The exciting light beam, linearly polarized, was directed perpendicularly to the magnetic field. The scattered light was detected along the field through the rotating polarizer. In this geometry, and with both D lines present in the light beams, simultaneous optical pumping could not be used. It would be possible only with careful spectral resolution of the pumping and scattered light beams, for instance, optical pumping peformed with the D_1 line and resonance fluorescence observed through the D_2-line filter. Interference filters with narrow spectral bands must be utilized.

The experimental arrangement used in subsequent investigations was as follows (Fig. 5). Two light beams directed along the magnetic field were used. The first light beam was linearly polarized and was described by the polarization vector $\boldsymbol{e}_{\lambda_0} = \boldsymbol{i}, \boldsymbol{j}, \boldsymbol{j} \pm \boldsymbol{i}$. The second light beam, circularly polarized, was the pumping beam. Both D-lines of the potassium lamps were used (only Schott's RG-8 filters were applied). The resonance fluorescence was observed perpendicularly to the field through the linear polarizer. The polarization vector of the scattered light was given by $\boldsymbol{e}_{\lambda} = \boldsymbol{j}$. The magnetic field was modulated and phase-sensitive detection was used. The derivative of the resonance fluorescence signal as a function of the magnetic field strength was recorded.

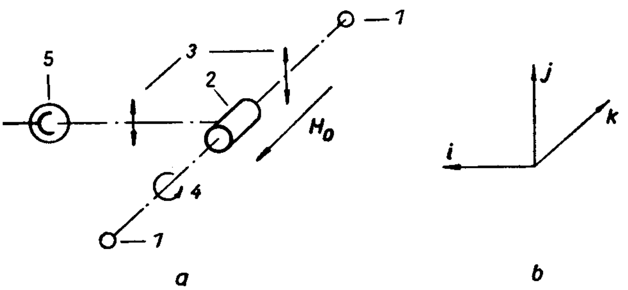

Fig. 5*a*. Apparatus for the study of level crossing with simultaneous optical pumping (lenses and filters have not been drawn). *1* – *K*-lamps, *2* – resonance cell, *3* – linear polarizer, *4* – circular polarizer, *5* – photomultiplier, *b*. orientation of unit vectors corresponding to Fig. 5*a*

Experimental results

The record of the potassium resonance fluorescence *versus* magnetic field strength is shown in Fig. 6. The record was obtained with the first mentioned experimental set-up (with the rotating polarizer). For this geometry and detection method (without field modulation) the Lorentzian signal was obtained. On the zero-field level crossing signal an addi-

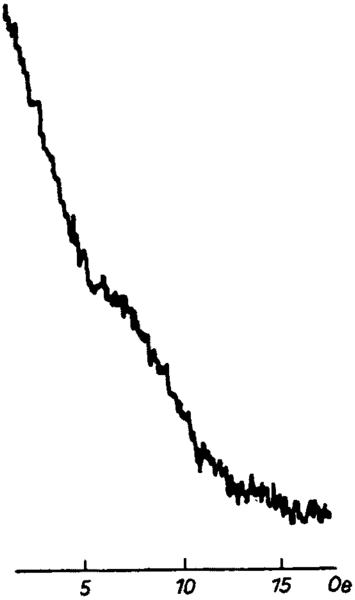

Fig. 6. Resonance fluorescence signal in potassium. Detection with a rotating polarizer

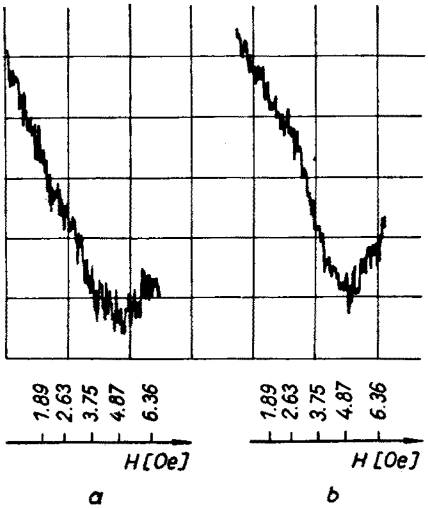

Fig. 7. The level crossing (2,−2)(1,0) signal and the Hanle signal. Polarization vector of the incident light $e_{\lambda_0} = j+i$. Peak to peak modulation of the magnetic field 0.3 Oe. *a* – with σ^+ optical pumping, *b* – without optical pumping

tional signal at about 7 Oe is visible and probably there is a trace of another signal at about 13 Oe.

In the set-up with field modulation two level crossing signals were observed, (2, −2) (1, 0), and overlapping (3, −2)(2, 0) and (3, −3)(2, −1). The first one (2, −2)(1, 0), at about 3 Oe, could be observed for a small amplitude of the modulating field. In Fig. 7*b* it is seen as a change in the slope of zero-field signal. Since the polarization vector of the exciting light was $\boldsymbol{e}_{\lambda_0} = \boldsymbol{i}+\boldsymbol{j}$, the registered signal is a derivative of a dispersion-shaped signal. In Fig. 7*a* the same zero-field level crossing curve with simultaneous σ^+ optical pumping is shown. The (2, —2)(1, 0) signal disappears here, because now the ground state levels with magnetic quantum number $\mu = -1$ are depopulated. As seen from Table III, the populations of the (2, —1) and (1, —1) ground state levels do not change noticeably with σ^- optical pumping. For small pumping light intensity (N small), a slight increase of population is obtained. For large pumping beam intensity, the populations of the previously mentioned levels decrease and are comparable with thoe corresponding to conditions without optical pumping. Thus σ^- optical pumping does not give an improvement in the (2, —2)(1, 0) level crossing signal.

An increase in the (3, —3)(2, —1) signal, which is associated with absorption from the (2, —2) level of the ground state, may be attained with optical pumping. This increase, however, is smaller than expected from the data in Table III, since the calculated populations are given only for a pumping beam and in the experiment a linearly polarized light beam is also present. This second light beam contributes a relaxation factor to the system. Its relaxation action is shown in Fig. 8. The magnetic resonance spectrum in the ground state of potassium with σ^- optical pumping is seen in Fig. 8*a*. Changes in the π component of the resonance

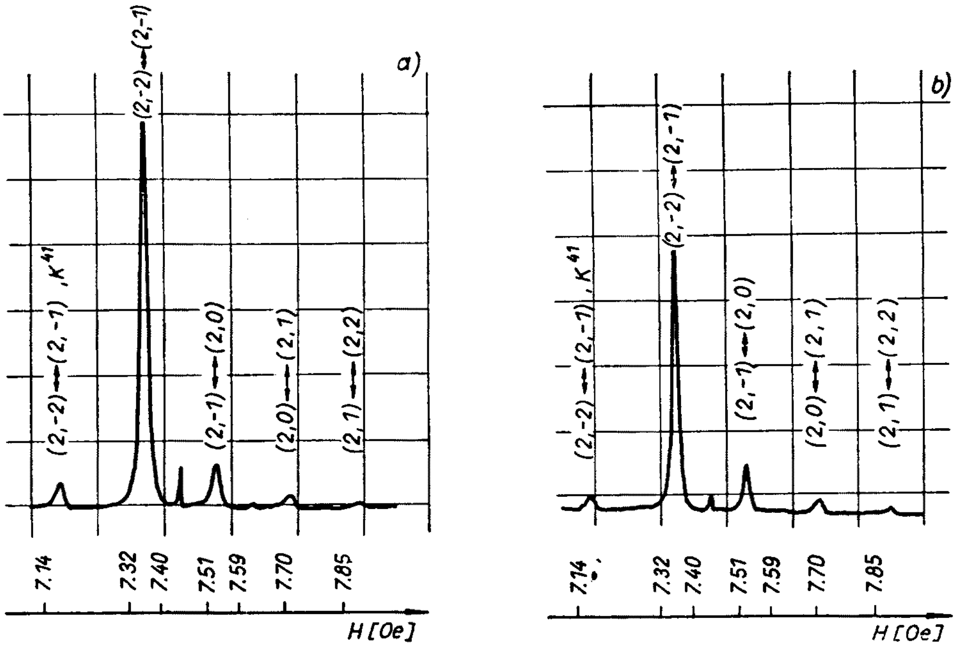

Fig. 8. Magnetic resonance spectrum in the ground state of potassium. Radio frequency 5.333 Mc/s. *a* – spectrum with only the σ^- optical pumping light beam, *b* – spectrum with both the pumping beam and linearly polarized light beam present

fluorescence were directly registered by a recording galvanometer. Only a pumping light beam is present. In Fig. 8*b*, the magnetic resonance spectrum for both of the mentioned light beams is recorded. The magnetic resonance (2, —2) ↔ (2, —1) decreases to about 2/3 of its maximum value without the second light beam. Thus, theoretically, for the best results the intensity of the light beam which excites the crossing levels should be as small as possible, whereas the intensity of the pumping beam should be as large as possible. In practice, we must consider the fact that the signal-to-noise ratio, rather than the absolute magnitude of the signal, is important. It is to be noted that the noise which arose from the pumping light beam was also registered.

The second observed level crossing signal was that of the superposition of the (3, —2) (2, 0) and the (3, —3)(2, —1) signals. (Fig. 9*a*). Simultaneous optical pumping causes an increase in the signal by a factor amounting to 2.5 (Fig. 9*b*). At the same time a shift of

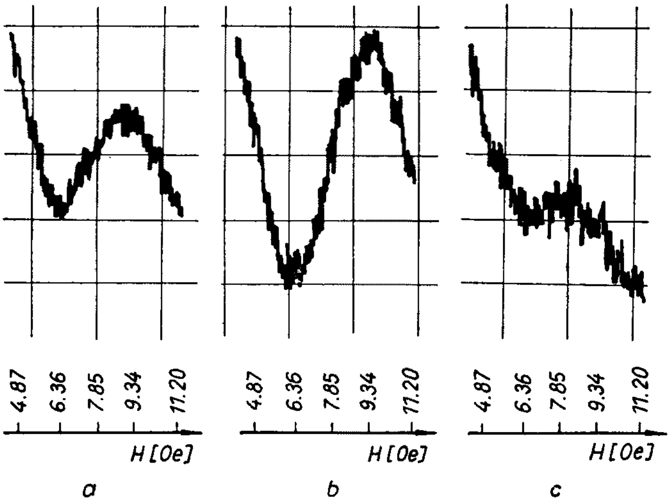

Fig. 9. Signal of the level crossings (3,—2)(2,0) and (3,—3)(2,—1). Polarization vector of the incident light $e_{\lambda_0} = j$; *a* – without optical pumping (equal populations of the ground state levels), *b* – with σ^- optical pumping, *c* – with σ^+ optical pumping

the signal towards higher magnetic field strength is observed. This effect is due to the greater contribution of the (3, —3)(2, —1) crossing signal. The shifted signal is situated at $H = 7.91 \pm 0.1$ Oe. The measurements for four different polarizations of the incident light were performed (either dispersion-shaped curves or derivative dispersion-shaped curves were registered).

In Fig. 9*c* the signal with simultaneous optical pumping σ^+ is also shown. In this case the population of the (2, —2) level of the ground state diminishes more than the population of the (2, —1) and the (1, —1) levels. In the superposition of signals the contribution of the (3, —2)(2, 0) crossing predominates. However the overall signal decreases and the signal-to-noise ratio is very poor.

The signals of level crossing obtained with optical pumping were not, however, the "pure" signals, corresponding to a single crossing. Thus direct determination of hyperfine coupling constants was not possible.

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