OPTICAL ORIENTATION OF ⁸⁵Rb AND ⁸⁷Rb ATOMS BY LIGHT OF THE D₂ LINE AND RELAXATION IN THE ²P_{3/2} STATE DUE TO COLLISIONS WITH INERT-GAS ATOMS

R. A. ZHITNIKOV, P. P. KULESHOV, A. I. OKUNEVICH, and B. N. SEVAST'YANOV

A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences

Submitted October 9, 1969

Zh. Eksp. Teor. Fiz. 58, 831-842 (March, 1970)

The dependence of optical pumping of Rb atoms on the pressure of the inert gases He, Ne, Ar, Kr and Xe is investigated. By applying a rotating magnetic field for separately observing the resonance signals from two hyperfine-structure sublevels of the Rb atom ground state, one can find the characteristic pressure of the inert gas, P'_0 , at which the signal of the hyperfine-structure sublevel with a large value of Φ (Φ is the total angular momentum) vanishes. The obtained values of P'_0 are used to calculate the disorientation cross sections in the ${}^2P_{3/2}$ excited state of the Rb atoms due to collisions with noble-gas atoms.

INTRODUCTION

MANY recent papers, both experimental^[1-4] and</sup> theoretical^[5,6], are devoted to an investigation of the optical orientation of alkali atoms by light of the D_2 line in the presence of buffer gases. Interest in optical pumping by the D_2 line is due to the fact that the optical-pumping signal reverses sign when the pressure of the buffer gas is increased. This is connected with the fact that the collisional relaxation ("mixing") in the excited ${}^{2}P_{3/2}$ state leads to inversion of the populations of the sublevels of the ground ${}^{2}S_{1/2}$ state^[1]. The inertgas pressure P_0 at which the optical-pumping signal vanishes is characteristic of the given alkali atom + inert gas combination, and can be used to obtain quantitative information concerning the collisional relaxation in the excited ${}^{2}P_{3/2}$ state. In^[2,3], this pressure P_0 of inert gases was measured for Na and Cs, and attempts were made to calculate the cross sections of the depolarizing collisions in the excited state using the measured values of P_0 .

An interesting object for investigations of this kind are the isotopes ⁸⁵Rb and ⁸⁷Rb, which have different nuclear spins I (I = $\frac{5}{2}$ for ⁸⁵Rb and I = $\frac{3}{2}$ for ⁸⁷Rb), making it possible to clarify the role of the nuclear spin in collisional relaxation. We have recently reported^[4] preliminary results of experiments on optical orientation of Rb isotopes with light of the D₂ line in the presence of inert gases.

In the present paper we used, for the excitation of resonance in the ground state of optically oriented Rb atoms, a rotating magnetic field, making it possible to consider separately the resonant signals of the optical pumping for different hfs sublevels and to investigate the depending of these signals on the inert-gas pressure.

The ground ${}^{2}S_{1/2}$ state of alkali atoms has two hfs sublevels characterized by a total angular momentum Φ ; the g factors of these sublevels have opposite signs. Therefore a rotating magnetic field induces resonant transitions only between Zeeman sublevels having the same value of Φ . The change of the direction of rotation of the field makes it possible to excite resonance in a system of sublevels having different Φ . Separate observation of the signals for two hfs sublevels of the ground state of the Rb atoms has made it possible to find the characteristic inert-gas pressures P'_0 at which the signal for the sublevel with the larger Φ vanishes. From the obtained values of P'_0 , the cross sections of the disorientation of the Rb atoms in the excited ${}^{2}P_{3/2}$ state were calculated for collisions with inert-gas atoms. It is also shown that the characteristic pressure P'_0 of the inert gas is practically independent of the spectral profile of the pump light, whereas the P_0 values measured in^[2,4] (the summary signal for both hfs of the sublevels vanishes at $P = P_0$) depends strongly on the ratio of the intensities of the hfs components in the pump light, making it difficult to use these quantities for the calculation of the cross sections of the disorienting collisions.

1. EXPERIMENTAL TECHNIQUE

To observe the optical orientation of the Rb atoms, we employed the usual method of registration of the change of the absorption of the pumping light beam at magnetic resonance in the ground state of the Rb atoms. The light source was a spherical resonant lamp, in which an electrodeless high-frequency discharge was excited. The lamp contained metallic Rb with natural concentration of the isotopes and Kr at a pressure of 2 Torr. The separation of the D₂ line from the radiation spectrum of the lamp was by means of an interference filter. The transmission of the D₁ line $(\lambda = 7946 \text{ Å})$ was less than 1% of the transmission of the D₂ line $(\lambda = 7800 \text{ Å})$.

The circularly polarized light of the D_2 line (the light flux was approximately 40 μ W/cm²) pumped Rb vapor located in an absorbing cylindrical cell (length 80 mm, diameter 40 mm). A constant magnetic field of ~1 Oe was applied parallel to the light beam. This field had a low-frequency modulation of small amplitude. The passage through resonance was effected by slow variation of the constant magnetic field. The resonance variation of the light intensity passing through the cell (optical-pumping signal) was detected with a silicon photodiode. To register the signal we used the technique of synchronous detection with recording of the derivative of the optical signal of magnetic resonance by means of an automatic plotter.

To excite resonance in the ground state of the Rb atoms, we used a rotating magnetic field. It was produced with the aid of two mutually perpendicular pairs of Helmholtz coils (diameter 260 mm), the currents in which were equal in amplitude and shifted 90° in phase. The resultant vector of the magnetic radio-frequency field (frequency 500 kHz) rotated in a plane perpendicular to the light beam. Simple switching of the capacitors connected in series with each pair of rings made it possible to reverse the direction of rotation of the magnetic-field vector (the magnitude of the vector did not change upon reversal).

The absorbing cell, placed in the center of the system of Helmholtz coils, was connected to a highvacuum setup, which made it possible to effect evacuation and a controlled admission of the inert gases. The inert-gas pressure was measured with a Pirani manometer.

2. EXPERIMENTAL RESULTS

Figure 1 shows a typical dependence of the signals obtained by optical pumping with the D_2 line on the inert-gas pressure, using ⁸⁷Rb in Ar as an example. The ordinates represent the maximum value A of the derivative of the optical-pumping signal. Positive values of A in Fig. 1 correspond to an increase of the transparency of the absorbing cell for light of the D_2 line in magnetic resonance in the ground state.

It is seen from Fig. 1 that the behavior of the signals for the sublevels with different Φ is greatly different. Attention is called first of all to the change of the sign of the signal for the sublevel with $\Phi = 2$. The sharp decrease of this signal with reversal of the sign at $P = P'_0$ is due to the strong relaxation in the excited state. As will be shown below, this relaxation near the point P'_0 leads to population inversion in the system of sublevels with $\Phi = 2$. No such inversion takes place in the system of sublevels with $\Phi = 1$, and the signal for the level with $\Phi = 1$ depends little on the pressure. The decrease of the signals in the region of very low pressures is connected with relaxation on the walls of the cell. In Fig. 1 is noted the characteristic pressure P_0 at which the total optical-pumping signal vanishes, as well as the other characteristic pressure P'' at which the amplitudes of the signals from the sublevels with different Φ are equal. The dependence of the signals on the pressure for ⁸⁵Rb is similar. For this isotope, a reversal of the signal sign is observed for the sublevel with $\Phi = 3$.

For the inert gases He and Ne, the pressure dependence of the signals is similar to that shown in Fig. 1 for Ar. For Kr and Xe, the qualitative difference compared with Ar lies in the sharper decrease of the signal amplitudes with increasing pressure, this being the consequence of appreciable relaxation in the ground state of the Rb atoms. Thus, it is known that the cross sections for the disorientation in the ground state of the Rb atoms (in cm²) are equal to 5.9 $\times 10^{-21}$ and 1.3×10^{-20} for Kr and Xe, whereas for Ne

FIG. 1. Experimental plot of the amplitude of the derivative of the signals of optical pumping of ⁸⁷Rb atoms on the argon pressure ($t=10^{\circ}$ C). The pumping is by means of circularly polarized light of the D₂ line.



and Ar these cross sections are equal to 5.2×10^{-23} and 3.7×10^{-22} ^[7]. The absolute value of the signals in Kr and Xe was much lower than in Ne and Ar. The width of the signals was independent of the type of gas and amounted to approximately 5 kHz in the pressure range from zero to 10–15 Torr. In our experiments, the width of the signals was determined by the inhomogeneity of the longitudinal field in the volume of the absorbing cell.

The inert-gas pressure P'_0 at which the signal for the sublevel with the larger Φ vanishes does not depend on the amplitude of the radio-frequency field (in a region far from saturation). When the light intensity is strongly decreased, a certain decrease of P'_0 is observed for both isotopes. Thus, when the light intensity is decreased by a factor of four, a 5% decrease of P'_0 is observed in Ne. With increasing temperature, P'_0 shifts as a rule towards higher pressures (when the temperature is increased from zero to 20°C, P' increases by 5%). However, a small maximum of P'_0 is observed for ⁸⁷Rb in Ne and Ar at 10°C (15-20% relative to the value of P'_0 at $0^{\circ}C$). Such an anomaly in the temperature dependence is apparently connected with the spin exchange between the Rb isotopes. It is clear, however, that the influence of the spin exchange should decrease with decreasing temperature, owing to the strong decrease of the pressure of the alkalimetal vapor. Therefore, to eliminate the action of the spin exchange on the value of P'_0 , we measured this quantity for as low a temperature as possible. In addition, we used a weak resonant radio-frequency field in the measurements of P'_0 (we verified that the signal is proportional to the square of the amplitude of the RF field).

Table I lists the measured values of P'_0 for the Rb isotopes in inert gases. It is seen from the foregoing data that P'_0 for ${}^{85}\text{Rb}$ (${}^{85}P'_0$) is larger in all the investigated gases than P'_0 for ${}^{87}\text{Rb}$ (${}^{87}P'_0$). The error in the measurement of the pressure was 10%. In Table I is given also the ratio $q = {}^{85}\text{P}'_0/{}^{87}\text{P}'_0$. This ratio is approx-

Table I

Gas	t, °C	⁵5 _{₽₀} ′, Torr	" <i>P</i> ₀', Torr	q= ⁸⁵ P ₀ '/ ⁸⁷ P ₀ '
He Ne Ar Kr Xe	0 0 0 10	3,04 6,91 4,32 3,73 3,86	2,124,322,752,462,60	$\begin{array}{c} 1.43 \pm 0.05 \\ 1.60 \pm 0.05 \\ 1.57 \pm 0.05 \\ 1.52 \pm 0.05 \\ 1.50 \pm 0.06 \end{array}$

imately the same within the limits of errors for all the inert gases, with a certain deviation in the case of helium. The error for the ratio q was determined from the mean-squared deviations in a series of experiments.

We note also that the ratio $\rho = P''/P'_0$ was approximately constant for each isotope. For He, Ne, Ar, Kr, and Xe we obtained respectively the following values of ρ : 2.1, 2.3, 2.1, 2.1, and 1.7 for ⁸⁵Rb and 1.4, 1.5, 1.5, 1.4, and 1.3 for ⁸⁷Rb.

3. POPULATION-BALANCE EQUATIONS IN OPTICAL PUMPING BY THE D_2 LINE WITH ALLOWANCE FOR COLLISION RELAXATION IN THE EXCITED $^2P_{3/2}$ STATE

The problem of obtaining quantitative information concerning the collision relaxation in the ${}^{2}P_{3/2}$ state from the experimental values of P'_{0} presupposes the solution of the population-balance equations for optical pumping, and a comparison of the theoretical dependence of the optical-pumping signal on the inert-gas pressure with the corresponding experimental dependence.

To take into account the collision relaxation in the excited ${}^{2}P_{3/2}$ state, we use the results of Okunevich and Perel^{>[8]}. The relation obtained in that paper for the rate of change of the populations of the sublevels of the excited ${}^{2}P_{3/2}$ state in collisions with inert-gas atoms is given by

$$\left(\frac{\partial f_{Fm}}{\partial t}\right)_{\text{collision}} = -n\bar{v}s\sum_{F_{i}m_{i}}\Gamma_{Fm}^{F_{i}m_{i}}f_{F_{i}m_{i}},\tag{1}$$

where

$$\Gamma_{Fm}^{F_{i}m_{1}} = \sum_{jn} (2F+1) (2F_{1}+1) \left(\begin{array}{c} F F_{1} \ j \\ -m m_{1} n \end{array} \right)^{2} \left\{ \begin{array}{c} J \ j \ J \\ F_{1} I F \end{array} \right\}^{2} (2J+1) h_{j},$$
(2)
$$h_{1} = \sigma_{1} / n \overline{n} s \qquad (3)$$

Here f_{Fm} is the population of the sublevel of the excited ${}^{2}P_{3/2}$ state characterized by the total angular momentum F and its projection m; $J = {}^{3}\!\!/_{2}$ is the electron angular momentum, I is the spin of the nucleus, n is the concentration of the inert-gas atoms, $\overline{v} = (8kT/\pi\mu)^{1/2}$ is the average relative velocity of the alkali atom and of the inert-gas atom (μ is the reduced mass of these atoms).

According to relation (1), to each transition in the system of sublevels of the excited state there corresponds a different cross section $(s\Gamma_{Fn}^{F_1m_1})$. The quantity s (see also formula (1)) has the dimension of area, and we shall call it the reduced cross section of the collision disorientation.

The following values of h_j were obtained in^[8] by numerically solving the collision problem for the case of a Van der Waals interaction between the atoms: h_0 = 0.97, $h_1 = -0.21$, $h_2 = -0.44$, and $h_3 = -0.32$. It is shown in^[8] that the value of s is connected with the Van der Waals interaction constants for the pair comprising the inert atom and the alkali metal atom in the excited state by the simple relation

$$s = \frac{3}{5} (24\pi^4)^{1/s} \Gamma\left(\frac{9}{5}\right) \left(\frac{c_{\frac{1}{2}} - c_{\frac{3}{2}}}{\vec{v}\hbar}\right)^{\frac{1}{2}}.$$
 (4)

The difference between the Van der Waals constants

 $c_{1/2}$ and $c_{3/2}$ in formula (4) characterizes the energy difference ΔE of the terms of the quasimolecule produced upon collision:

$$\Delta E = (-c_{\frac{1}{2}}/R^{6}) - (-c_{\frac{3}{2}}/R^{6}),$$

where R is the distance between the colliding atoms.

Introducing into the kinetic equations for the populations in optical pumping^[9] the term (1), we obtain after simple transformations the following system of population-balance equations:

$$-\psi_{Fm} - z \sum_{F,m_1} \Gamma_{Fm}^{F,m_1} \psi_{F,m_1} + \sum_{\Phi\mu} k_{\Phi} A_{\Phi\mu}^{Fm} \phi_{\Phi\mu} = 0, \qquad (5)$$

$$\sum_{Fm} C^{Fm}_{\Phi\mu} \psi_{Fm} - \sum_{Fm} k_{\Phi} A^{Fm}_{\Phi\mu} \varphi_{\Phi\mu} + zr \sum_{\Phi_1\mu_1} (\varphi_{\Phi_1\mu_1} - \varphi_{\Phi\mu}) = 0.$$
 (6)

The quantity ψ_{Fm} is connected with f_{Fm} by the relation

$$\psi_{Fm} = \frac{T_p}{\tau} f_{Fm},\tag{7}$$

where τ is the lifetime in the excited ${}^{2}P_{3/2}$ state, T_{p} is the pumping time^[9] for the ground state (the quantity T_{p} is inversely proportional to the intensity of the pump light).

The matrices \hat{A} and \hat{C} , which enter in Eqs. (5) and (6), give the relative probabilities of the transitions from the ground state to the excited state and viceversa, as a result of absorption of circularly-polarized light and spontaneous decay. The matrices \hat{A} and \hat{C} are expressed in terms of the matrix elements of the dipole moment **D** as follows:

$$A_{\Phi\mu}^{Fm} = |(J_0 || D || J)|^{-2} |\langle \Phi \mu | \mathbf{e}_{\lambda_0} \mathbf{D} | Fm \rangle|^2, \tag{8}$$

$$C_{\Phi_{\mu}}^{Fm} = (2J+1) | (J_0 || D || J) |^{-2} | \langle Fm | \mathbf{D} | \Phi_{\mu} \rangle |^2.$$
(9)

In formulas (8) and (9), $J_0 = \frac{1}{2}$ and $J = \frac{3}{2}$ are the electron angular momenta of the ground and excited states; $(J_0 \parallel D \parallel J)$ is the reduced matrix element of the moment **D**; e_{λ_0} is a unit vector characterizing the polarization λ_0 of the pump light. For concreteness, we assume that pumping is by means of right-polarized light.

In Eqs. (5) and (6) we have introduced a parameter z proportional to the pressure P of the inert gas:

$$z = n\overline{v}s\tau = P\overline{v}s\tau / kT, \tag{10}$$

and a parameter r, characterizing the relaxation in the ground state:

$$r = \frac{T_p \sigma}{\tau s}$$
(11)

The quantity σ in formula (11) is the cross section for the disorientation in the ground state of the Rb atoms in collisions with the inert-gas atoms. To take into account the collision relaxation in the ground state, we use the model of "uniform" relaxation^[10]: the probabilities w of all the relaxation transitions between the sublevels of the ground state are assumed to be the same and equal to

$$w = n\overline{v}\sigma. \tag{12}$$

To take into account the spectral profile of the pump light we have introduced into Eqs. (5) and (6) the factor

$$k_{\Phi} = 1 \text{ for } \Phi = \Phi_{max}, \qquad (13)$$

$$k_{\Phi} = k \text{ for } \Phi = \Phi_{min}. \tag{14}$$

It is known that owing to the large hyperfine splitting in the ground state the spectrum of the D_2 line of alkali metals consists of two hfs components corresponding to transitions from the excited ${}^2P_{3/2}$ state into two hfs sublevels of the ground ${}^2S_{1/2}$ state, and the intensities of these components J_{Φ}_{max} and J_{Φ}_{min} are not equal. (For Rb atoms, the spectrum of the D_2 line is shown in^[11].) Since the width of these components in the emission spectrum of the lamp is usually larger than the hfs splitting in the excited state and is larger than the width of the corresponding absorption lines of the atoms in the absorbing cell, we can approximate the real spectral profile of the D_2 line by a stepwise-rectangular profile and introduce a coefficient k characterizing the ratio of the intensities of the two hfs components:

$$k = J_{\Phi_{min}} / J_{\Phi_{max}}.$$
 (15)

Equations (5) and (6) constitute a system of linear homogeneous algebraic equations of order N (N = 24 for ⁸⁷Rb and N = 36 for ⁸⁵Rb). Nonzero solutions of these equations exist if the determinant of the system is equal to zero. The populations that enter in Eqs. (5) and (6) are not, however, independent variables, since they obey the normalization condition

$$\sum_{\Phi\mu} \varphi_{\Phi\mu} = 1. \tag{16}$$

(The sum of populations of the sublevels of the excited state has been neglected, since at usual light intensities $\Sigma f_{Fm} \ll \Sigma \varphi \Phi \mu$.)

Replacing one of the equations (5) or (6) by the normalization equation (16) and solving the obtained system of equations for different values of the dimensionless parameters z, k, and r, we can find the populations of the sublevels of the ground state in the case of optical pumping by light of the D_2 line in the presence of an inert gas. We denote the populations obtained in this manner by $\varphi_{\Phi\mu}^{0}$ for the ground state and $f_{\rm Fm}^{0}$ for the excited state.

The observed optical-pumping signal is equal to the change in the absorption of light at resonance in the ground state compared with the absorption of light in the absence of resonance.

To find the populations f_{Fm}^{res} of the sublevels of the excited state and $\varphi_{\Phi\mu}^{res}$ of the ground state in the presence of a resonant RF field, it is necessary to take into account in the population balance equations the transitions induced by such a field. Let us assume that a weak longitudinal magnetic field H is applied parallel to the pump light beam. We assume also that the resonances produced by magnetic field H₁, the vector of which rotates in a plane perpendicular to the light beam. In the case when the width of the resonance line is determined mainly by the inhomogeneity of the longitudinal magnetic field (this was the case in our experiments), the probability of the transition per unit time $R_{\Phi\mu}^{\Phi\mu}$ from the sublevel Φ , μ to the sublevel Φ , μ' under the influence of the RF field H₁ can be written in the form^[12]

$$R_{\Phi\mu}^{\Phi\mu'} = H_1^2 Q(H) \Delta(\Phi) P_{\Phi\mu}^{\Phi\mu'}, \qquad (17)$$

$$P_{\Phi\mu}^{\Phi\mu''} = \delta(\mu, \mu' - 1) [(\Phi - \mu) (\Phi + \mu + 1)] + \delta(\mu, \mu' + 1) [(\Phi + \mu) (\Phi - \mu + 1)].$$
(18)

In formula (17), Q(H) is a function of the longitudinal magnetic field H, proportional to the form function of the resonance line. The symbol $\Delta(\Phi)$ is determined by the relation

$$\Delta(\Phi) = \begin{cases} \text{for } \Phi = \Phi_{\max} \text{ and a right-polarized RF field} \\ \text{for } \Phi = \Phi_{\min} \text{ and a left-polarized RF field (19)} \\ \text{for all remaining cases.} \end{cases}$$

Relation (19) follows from the fact that the g-factor of the two hfs levels of the Rb atoms have opposite signs, namely

$$g_{\Phi_{max}} > 0, \quad g_{\Phi_{min}} < 0$$

Let us assume that the resonant field H_1 is small. Then the population changes due to this field

$$f_{Fm}^{(2)} = f_{Fm}^{\text{res}} - f_{Fm}^{0}, \quad \varphi_{\Phi\mu}^{(2)} = \varphi_{\Phi\mu}^{\text{res}} - \varphi_{\Phi\mu}^{0},$$
 (20)

are also small quantities (of the order of H_1^2). Supplementing the balance equations by a term that takes into account the RF transitions, and making the substitution (20), we obtain for the quantities $\beta_{\rm Fm}$ and $\alpha \Phi \mu$, which are connected with $f_{\rm Fm}^{(2)}$ and $\varphi_{\Phi \mu}^{(2)}$ by the relations

$$\beta_{Fm} = \frac{1}{\tau Q H_1^2} f_{Fm}^{(2)}, \quad \alpha_{\Phi\mu} = \frac{1}{T_p Q H_1^2} \varphi_{\Phi\mu}^{(2)}$$
(21)

(we retain only terms of order H_1^2) the following system of equations:

$$-\beta_{Fm} - z \sum_{F_i m_1} \Gamma_{Fm}^{F_i m_1} \beta_{F_i m_1} + \sum_{\Phi \mu} k_{\Phi} A_{\Phi \mu}^{Fm} \alpha_{\Phi \mu} = 0, \qquad (22)$$

$$\sum_{Fm} C_{\Phi\mu}^{Fm} \beta_{Fm} - \sum_{Fm} k_{\Phi} A_{\Phi\mu}^{Fm} \alpha_{\Phi\mu} + zr \sum_{\Phi,\mu_1} (\alpha_{\Phi,\mu_1} - \alpha_{\Phi\mu})$$
$$= \Delta(\Phi) \sum_{\mu'} P_{\Phi\mu}^{\Phi\mu'} (\phi_{\Phi\mu}^0 - \phi_{\Phi\mu'}^0).$$
(23)

The determinant of the system of equations (22) and (23) coincides with the determinant of the system (5) and (6), and consequently vanishes. To find the quantities $\alpha \Phi \mu$ and $\beta_{\rm Fm}$ it is necessary to replace one of the equations (22) or (23) by the "normalization" equation

$$\sum_{\Phi\mu} \alpha_{\Phi\mu} = 0. \tag{24}$$

The change of the light absorption at resonance (the absorption signal) ΔI is equal to

$$\Delta I = -\frac{N}{T} \sum_{\substack{\boldsymbol{\Phi} \boldsymbol{\mu} \\ Fm}} k_{\boldsymbol{\Phi}} A_{\boldsymbol{\Phi} \boldsymbol{\mu}}^{Fm} \boldsymbol{\varphi}_{\boldsymbol{\Phi} \boldsymbol{\mu}}^{(2)}, \qquad (25)$$

where N is the concentration of the considered Rb isotope. In our experiments we registered as the opticalpump signal A the derivative of the absorption signal with respect to the magnetic field H, i.e.,

 $A = \partial(\Delta I)/\partial H$. Taking into account the definition (19) of the quantity $\Delta(\Phi)$, Eqs. (22) and (23) constitute a compact way of writing down the system of equations for the two opposite directions of rotation of the resonant RF field.

We denote the solution of Eqs. (22) and (23) (with allowance for (24)) in the case of a right-polarized field by $\beta_{\rm Fm}^{\rm max}$ and $\alpha_{\Phi\mu}^{\rm max}$ (such a field induces resonant transitions between sublevels with $\Phi = \Phi_{\rm max}$), and in the case of a left-polarized field by $\beta_{\rm Fm}^{\rm min}$ and $\alpha_{\Phi\mu}^{\rm min}$. Using this notation, and also (21), we obtain for the signals from the sublevels with different Φ the following expressions:

$$A_{\Phi_{max}} = -N \frac{\partial Q}{\partial H} H_1^2 \sum_{Fm} k_{\Phi} A_{\Phi\mu}^{Fm} \alpha_{\Phi\mu}^{max} , \qquad (26)$$

$$A_{\Phi_{min}} = -N \frac{\partial Q}{\partial H} H_1^2 \sum_{\substack{Fm \\ \Phi_\mu}} k_{\Phi} A_{\Phi\mu}^{Fm} a_{\Phi\mu}^{min} .$$
 (27)

It is seen from formulas (26) and (27) that the optical-pumping signals are proportional to H_1^2 . This property is the direct consequence of the assumed smallness of the amplitude of H_1 of the RD field, and can be used for an experimental verification of the satisfaction of the condition of smallness of the RF field.

4. CALCULATION OF THE REDUCED CROSS SECTION s FROM THE OBTAINED EXPERIMENTAL DATA

At a large pump-light intensity and weak relaxation in the ground state, it is possible to neglect in Eqs. (6) and (23) the terms containing the parameter r, for in this case $r \ll 1$.

Equations (5) and (6) with allowance for (16) were solved for the isotopes ⁸⁵Rb and ⁸⁷Rb with an electronic computer at r = 0 and different values of k and z. (By way of an example, Fig. 2 shows the calculated dependences of the sublevel populations of the ground state of ⁸⁷Rb on the parameter z. It is seen from Fig. 2 that with increasing z population inversion takes place in the system of sublevels with $\Phi = 2$, and no inversion takes place in the system of sublevels with $\Phi = 1.$)

The obtained solutions $\varphi_{\Phi\mu}^0$ were used to solve Eqs. (22) and (23) with allowance for (24) at the same values of the parameters, and to calculate the reduced theoretical signals $A(NH_1^2\partial Q/\partial H)^{-1}$ by means of formu-



FIG. 2. Dependence of the populations $\varphi_{\Phi\mu}^{\circ}$ of the Zeeman sublevels of the ground ${}^{2}S_{1/2}$ state of the 87 Rb atoms on the value of the parameter z at k = 0.8 and r = 0, in the case of pumping with right-polarized light of the D₂ line.

FIG. 3. Dependence of the quantities z'_0 and z_0 for ⁸⁵ Rb and ⁸⁷ Rb on the ratio of the intensities of the hfs components in the pump light, k (without allowance for the relaxation in the ground ² S_{1/2} state).

las (26) and (27). As a result, we determined the dependence of the optical-pumping signals on z at several values of k and obtained the values of the parameter $z = z'_0$, at which the signal from the sublevel with $\Phi = \Phi_{\rm mx}$ vanishes. It turned out that z'_0 is practically independent of k, its values being ${}^{85}z'_0 = 3.96$ and ${}^{87}z'_0 = 2.49$ for ${}^{85}\text{Rb}$ and ${}^{87}\text{Rb}$, respectively (see Fig. 3).

The ratio of the values of z'_0 for the two Rb isotopes, $q = {}^{85}z'_0/{}^{87}z'_0 = 1.59$, can be compared directly with the corresponding ratio of the experimentally measured P'_0 . From the comparison in Table I we see that within the limits of measurement errors there is good agreement between the theoretical and experimental values for all gases, with a certain deviation in the case of He.

For comparison, Fig. 3 shows also the dependence of another characteristic value of the parameter $z = z_0$ on k. At $z = z_0$, the summary signal

$$A_{\rm c} = A_{\Phi_{max}} + A_{\Phi_{min}}$$

vanishes.

The dependence of z_0 on k is quite strong, as can be readily understood from the very definition of z_0 and k. The corresponding experimentally measured values of P_0 should also depend strongly on k, making it difficult to use P_0 for a determination of the cross section s.

Using the calculated values of z_0' and the values of P'_0 given in Table I, and also $\tau = 27.0 \pm 0.5$ nsec^[13], we obtained with the aid of (10) the reduced cross sections of s for all the investigated inert gases. Table II lists the values of s calculated for $T = 300^{\circ}K$ using the temperature dependence of the cross section s, namely s ~ $T^{-1/5}$ (see Formula (4)). It is seen from Table II that within the limits of the experimental error the values of s for the two isotopes of Rb coincide. This indicates that the assumption made in^[8], namely that the collisions of the Rb atoms in the ${}^{2}P_{3/2}$ state with the inert-gas atoms lead to reorientation only of the angular momentum of the electron shell J, but not the angular momentum of the nucleus I, is correct. Table II gives also the cross sections s obtained as a result of a theoretical estimate in^[8]. We see that there is good agreement between theory and experiment for all gases except He.

We recall that we are using in the determination of z'_0 the values of h_j [Eq. (3)] calculated in^[8] for a Van der Waals interaction of the colliding atoms (an estimate of the value of s was made also for this interaction). Therefore the large difference between the values of s calculated from experimental data and the theoretical value in the case of He casts doubts on the validity of the calculations made above for this gas. The discrepancy in the case of He is explained apparently by the fact that for this gas the Van der Waals interaction is not predominant, owing to the small

Table II. Values of $s(A^2)$ for T = 300° K

	He	Ne	Ar	Kr	Xe
⁸³ Rb ⁸⁷ Rb Theoreti- cal value	109 98 57	99 100 98	206 203 193	297 283 250	324 305 315

polarizability of this gas. For the remaining inert gases, the assumed Van der Waals interaction is apparently valid. This conclusion is confirmed in the main also by the experimental data on the shift of the spectral lines of Rb in inert gases^[14].

Using the mean values of the cross sections s for the two isotopes and formula (4), we obtain for the Rb atoms and for the inert gases the following values of the difference between the Van der Waals constants $(c_{1/2} - c_{3/2})$ in atomic units: He - (<134), Ne - 60, Ar - 280, Kr - 530, Xe - 600.

We note that the balance equations (5), (6), and (22), (23) do not depend on the type of the insert gases. In the absence of relaxation in the ground state, only the parameter z depends on the nature of the inert gas. Therefore the dependence of the experimental signals on the parameter z should be the same for those inert gases in which it is possible to neglect relaxation in the ground state, and should coincide, apart from a factor, with the corresponding theoretical dependence.

Figure 4 shows the dependences of the theoretical and experimental optical-pumping signals on the parameter z for ⁸⁷Rb. To convert from P to z, we used the relation $z = z'_0 P/P'_0$. The theoretical signals were tied-in in amplitude with the experimental ones at the point z = 12 using the signal for the sublevel with $\Phi = 2$. It should be noted that the ratio of the amplitudes of the signals for the levels with different Φ depends strongly on k. To construct the theoretical signals shown in Fig. 4, we used the fact that the ratio $\rho = P''/P'_0$, measured in the experiment, was approximately 1.5 for ⁸⁷Rb in all the inert gases. The corresponding theoretical value of ρ is obtained at $k \approx 0.8$. (For ⁸⁵Rb we have $\rho_{exp} = 2.1$ and respectively $k \approx 0.8$.)

It is seen from Fig. 4 that the theoretical and experimental dependences agree well for Ne and Ar (the



FIG. 4

FIG. 5

FIG. 4. Dependence of the experimental optical-pumping signals of the ⁸⁷ Rb atoms on the value of the parameter z in light inert gases (He, Ne, and Ar). Solid curves—theoretical, calculated with k = 0.8 and without allowance for relaxation in the ground ² S_{1/2} state. For $\Phi = 2$: $\Delta =$ He, $\times -$ Ne, $\bigcirc -$ Ar; for $\Phi = 1$: $\oplus -$ He, $\diamondsuit -$ Ne, $\square -$ Ar.

FIG. 5. Dependence of the experimental optical-pumping signals of the ⁸⁵ Rb atoms in Xe on the value of the parameter z. Solid and dashed curves—corresponding theoretical relations calculated with allowance for relaxation in the ground 2 S_{1/2} state at k = 0.8 and r = 8 × 10⁻³ (solid curve) and r = 3 × 10⁻³ (dashed).

discrepancy between the experimental signals for $\Phi = 2$ and the corresponding theoretical signals at small z is due to relaxation on the walls of the cell). In the case of He, there is a discrepancy (at small values of z) as a result of the fact that the values of h_j used by us cannot be employed for this gas. The agreement between theory and experiment confirms the assumption that relaxation in the ground state can be neglected in the case of Ne and Ar (and possibly in the case of He).

In the case of Kr and Xe, the relaxation in the ground state can apparently not be neglected, since a sharp qualitative difference was observed between the pressure dependences of the optical-pumping signals for these gases and for the light inert gases. In order to estimate the influence of the relaxation, we solved Eqs. (5), (6) and (22), (23) with allowance for (16), (24)at several values of the parameter r. It turned out that the amplitude of the signals decreases very strongly with increasing r, whereas z'_0 depends little on r. For example, for ⁸⁵Rb (at k = 0.8) we have obtained values of z'_0 equal to 3.93, 3.72, 3.42, and 3.36 respectively at r equal to 10^{-4} , 10^{-3} , 10^{-2} , and 10^{-1} , whereas at r = 0 we have $z'_0 = 3.96$. Using the experimentally measured value of the light flux of the D_2 line, 40 μ W/cm², and approximating the spectral profile of the D_2 line with a rectangle of width 0.1 cm⁻¹, we obtained for T_p a value of 0.2 msec. The values of σ and s in formula (11) were taken from^[7] and from Table II. According to formula (11), the values of the parameter r for Ne, Ar, Kr, and Xe are 4×10^{-5} , 1×10^{-4} , 2×10^{-3} , and 3×10^{-3} , respectively. Thus, in the case of Ne and Ar it is indeed possible to neglect the relaxation in the ground state $(z'_0 \text{ changes by less than})$ 1% when the relaxation is taken into account).

To estimate the value of the parameter r in the case of Kr and Xe, we likewise used a direct comparison of the theoretical and experimental signals at several values of the parameter r. Agreement between the theoretical and experimental dependences on the parameter z (for ⁸⁵Rb) is obtained at $r = 10^{-3}$ for Kr and $r = 8 \times 10^{-3}$ for Xe. Figure 5 shows by way of an example the theoretical and experimental curves for ⁸⁵Rb in Xe. The amplitude tie-in was effected at the minimum point for the signal from $\Phi = 2$. For comparison, Fig. 5 shows the theoretical curves for two values of the parameter r, 8×10^{-3} and 3×10^{-3} .

Bearing in mind the values of the parameter r obtained as a result of such a double estimate, we can conclude that the values of s listed in Table II are apparently overestimated by $\sim 7\%$ in the case of Kr and by $\sim 12\%$ in the case of Xe.

For a more rigorous account of the relaxation in the ground state in the calculation of the cross sections s, it is necessary first of all to use a more adequate model of relaxation in the ground state than the uniform-relaxation model employed by us, and also to measure directly the pump time T_p .

We are grateful to V. I. Perel' for consultation on the theoretical aspects of the present work.

¹J. Fricke and J. Haas, Z. Naturf. 21a, 1319 (1966).

²M. Elbel and F. Naumann, Z. Phys. 204, 501 (1967).

³J. Fricke, J. Haas, E. Luscher, and F. A. Franz, Phys. Rev. **163**, 45 (1967).

⁴R. A. Zhitnikov, P. P. Kuleshov, and A. I. Okunevich, Phys. Lett. 29A, 239 (1969).

⁵ F. A. Franz and J. R. Franz, Phys. Rev. 148, 82 (1966).

- ⁶ P. Violino, Nuovo Cimento 54B, 61 (1968).
- ⁷W. Franzen, Phys. Rev. 115, 850 (1959).
- ⁸A. I. Okunevich and V. I. Perel', Zh. Eksp. Teor.
- Fiz. 58, 666 (1970) [Sov. Phys.-JETP 31, 356 (1970)].
- ⁹J. P. Barrat and C. Cohen-Tannoudji, J. Phys. Rad. 22, 329, 443 (1961).

¹⁰W. Franzen and A. G. Emslie, Phys. Rev. 108, 1453 (1957).

- ¹¹W. Bell, A. Bloom, and J. Lynch, Rev. Sci. Instr. 32, 688 (1961).
- 32, 688 (1961). ¹²A. Lösche, Nuclear Induction (Russ. transl.), IIL, 1963.
 - ¹³J. K. Link, J. Opt. Soc. Am. 56, 1195 (1966).
- ¹⁴S. Chen and M. Takeo, Revs. Modern Phys. 29, 20 (1957)].
- Translated by J. G. Adashko

103