

Spin-Exchange Cross Section for Rb⁸⁵-Rb⁸⁷ Collisions*†

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The spin-exchange cross section for Rb⁸⁵-Rb⁸⁷ collisions has been measured to be $(1.70 \pm 0.21) \times 10^{-14}$ cm² using the techniques of optical pumping and an interferometric method for measuring the Rb vapor density. The Rb⁸⁷ constituent in a vapor of natural Rb was polarized through absorption of circularly polarized resonance radiation, and the Rb⁸⁷ polarization was partially transferred to the Rb⁸⁵ constituent through spin-exchange collisions. A phenomenological theory of the optical pumping plus the exchange polarization process which neglects nuclear spin was used to relate the cross section to the ratio of the signal strengths for the Rb⁸⁷ and Rb⁸⁵ Zeeman resonances, the relaxation time, and the Rb⁸⁷ vapor density. The density was measured by comparing spectral profiles of the resonance radiation transmitted through the optical pumping cell at room temperature with the resonance radiation transmitted at the temperature at which the signal strengths were measured. These spectral profiles were obtained through use of a scanning Fabry-Perot interferometer.

I. INTRODUCTION

THE first determination of an atomic spin-exchange cross section was accomplished by Wittke and Dicke¹ in their experiments on the hyperfine splitting in the ground state of atomic hydrogen using microwave absorption techniques. In subsequent experiments by others, optical pumping techniques² have been exploited to determine spin-exchange cross sections for several different systems. In this work, an alkali vapor contained in a Pyrex cell is polarized through absorption of circularly polarized resonance radiation, and this polarization is partially transferred to another atomic or subatomic specie in the cell through spin-exchange collisions. This phenomenon of exchange polarization was first used by Dehmelt³ to polarize an electron gas through collisions with an optically pumped Na vapor. Shortly after, exchange polarization in Rb-Cs,⁴ Na-K,⁵ and Na-Rb⁶ systems was demonstrated. Numerical estimates of spin-exchange cross sections for alkali-electron and alkali-alkali collisions were obtained from these experiments. These estimates had large uncertainties connected with them, and in effect the cross sections quoted in this early work were order of magnitude estimates. The main reason for the uncertainty in this early work was that in an optical pumping experiment, the quantity measured directly is the rate of polarization transfer from one vapor to another. This

rate depends upon the spin-exchange cross section, the relative velocities of the members of the collision, and the density of the two vapors. The difficulty lies in obtaining reliable estimates of the vapor densities.

In the present work,⁷ we have measured the spin-exchange cross section for Rb⁸⁵-Rb⁸⁷ collisions through optically pumping Rb⁸⁷ and exchange polarizing Rb⁸⁵, and by measuring the vapor density in the optical pumping cell using an interferometric technique. This technique compares the spectral profile of the Rb *D*₁ line transmitted through the cell at room temperature with the spectral profile of the same line transmitted through the cell at the temperature at which the optical pumping is performed. These profiles were obtained through use of a scanning Fabry-Perot interferometer constructed for this purpose. The interferometric technique of vapor density measurement frees this experiment from the major uncertainty involved in experiments of this type. The experimental results for Rb⁸⁵-Rb⁸⁷ collisions yield a cross section of $(1.70 \pm 0.21) \times 10^{-14}$ cm².

II. DENSITY MEASUREMENT

Two pieces of information are generally available in an optical pumping experiment to yield density estimates. These are the temperature of the optical pumping cell and the fraction of the light transmitted through the cell at optical pumping temperatures. Neither of these two pieces of information can be used to obtain reliable density estimates. The observed behavior of alkali vapor pressure below 1 mm shows a very strong functional dependence upon the temperature.⁸ A change in temperature of about 30°C is sufficient to change the pressure by an order of magnitude. Therefore, the temperature of the alkali vapor source must be known quite accurately for these data to be used. A more difficult problem in using vapor density data arises from the construction of optical pumping cells. Alkali

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¹ J. P. Wittke and R. H. Dicke, *Phys. Rev.* **103**, 620 (1956).

² For a review of optical pumping techniques see A. Kastler, *J. Opt. Soc. Am.* **47**, 460 (1957) and G. V. Skrotskii and T. G. Izyumova, *Usp. Fiz. Nauk* **73**, 423 (1961) [translation: *Soviet Phys.—Usp.* **4**, 177 (1961)].

³ H. G. Dehmelt, *Phys. Rev.* **109**, 381 (1958).

⁴ H. G. Dehmelt, *J. Phys. Radium* **19**, 866 (1958).

⁵ P. Franken, R. Sands, and J. Hobart, *Phys. Rev. Letters* **1**, 118 (1958).

⁶ R. Novick and H. E. Peters, *Phys. Rev. Letters* **1**, 120 (1958).

⁷ S. M. Jarrett, Ph.D. thesis, University of Michigan, 1962 (unpublished).

⁸ S. Dushman, *Scientific Foundations of Vacuum Technique* (John Wiley & Sons, Inc., New York, 1949), Chap. II, pp. 740-745.

metal is distilled into a Pyrex cell, covering some fraction of the cell wall. The film of alkali metal acts as a source of the vapor, but the bare glass wall of the cell acts as a sink for the alkali vapor. Therefore, the alkali vapor density at a given cell temperature depends upon how the cell is prepared, that is, on the ratio of glass wall with alkali film to bare glass wall. Knowledge of the fraction of light absorbed in the cell⁹ cannot give reliable density estimates under usual optical pumping conditions either. This information can only be used if the emission line of the lamp were identical to the absorption line of the vapor in the cell. This would be a highly unusual situation for the lamps and cells used in optical pumping experiments. The method of density measurement used in this work¹⁰ is felt to be capable of yielding results of about 10% accuracy as opposed to the order of magnitude estimates from the methods previously mentioned. This method will now be discussed.

If a parallel beam of light of wave number ν passes through a cylindrical cell of length L , the incident intensity I_0 is related to the transmitted intensity I , through an exponential law of absorption,¹¹

$$I = I_0 e^{-k_\nu L}. \quad (1)$$

This relation is a defining equation for the absorption coefficient k_ν . The integral of the absorption coefficient over the absorption line is related to the density of the vapor N through

$$\int k_\nu d\nu = (\lambda_0^2 g_2 / 8\pi c g_1 \tau) N, \quad (2)$$

where λ_0 is the wavelength of the center of the absorption line; g_1 and g_2 , the statistical weights of the ground and excited states of the transition involved in the absorption; τ , the lifetime of the excited state under spontaneous decay to the ground state; and c , the speed of light. This relation was used in conjunction with the high-frequency hyperfine component of the D_1 line of Rb^{87} , $\lambda_0 = 7948 \text{ \AA}$. The accuracy with which this relation may be applied is limited by the accuracy of the lifetime measurements. A recent determination by Stephenson¹² gives $\tau = (2.85 \pm 0.09) \times 10^{-8}$ sec. Using this value, $c = 3.00 \times 10^{10}$ cm/sec, $g_1 = g_2$ for the Rb^{87} D_1 line, and recognizing that only $\frac{3}{8}$ of the absorption is in the high-frequency hyperfine component ($^2P_{1/2}$ to $^2S_{1/2}$, $F=1$), the relation becomes

$$N = 9.07 \times 10^{12} \times \int k_\nu d\nu \text{ atoms/cm}^3 \quad (3)$$

with an uncertainty of about $\pm 3\%$.

⁹ A. C. G. Mitchell and M. W. Zemansky, *Resonance Radiation and Excited Atoms* (Cambridge University Press, London, 1961) Chap. 3, p. 118-119.

¹⁰ S. M. Jarrett and P. A. Franken (to be published).

¹¹ See Ref. 9, Chap. 3, pp. 92-97, 124-127.

¹² G. Stephenson, Proc. Phys. Soc. (London) **A64**, 458 (1951).

The integrated absorption coefficient was determined by comparison of spectral traces of the transmitted Rb^{87} D_1 high-frequency hyperfine component at room temperature (negligible Rb^{87} density) and at 90°C , the temperature at which the optical pumping experiment was performed.¹³ The traces were obtained with a scanning Fabry-Perot interferometer. The light transmitted through the optical pumping cell entered a vacuum chamber containing an etalon of thickness 1.1 cm, with 95% reflecting silver coatings. The fringe system was focused on a metal plate by a 508-mm focal length achromat. A circular hole of diameter small compared to the fringe diameter was placed at the center of the center fringe and the light passing through was monitored by a photomultiplier. The vacuum chamber was pumped down to fore pump pressure, and air slowly leaked back into the chamber through a capillary tube. This provides a time variation in the index of refraction of the medium between the Fabry-Perot plates, and thus the spectral profile of the transmitted light intensity can be obtained. For more details of this method the reader is referred to Refs. 7 and 10.

III. OPTICAL PUMPING SIGNALS

A phenomenological theory of the optical pumping plus exchange polarization process in Rb will be given in this section. A relation between the spin-exchange cross section for Rb^{85} - Rb^{87} collisions, the signal strengths encountered using the transmission monitoring technique, and the relaxation times will be derived. The spin-exchange cross section introduced in the theory will be determined through use of this relation and the measured signal strengths and relaxation times. In the present experiment, Rb^{87} is optically pumped and Rb^{85} exchange polarized. For a given magnetic field, the Zeeman resonances for the two isotopes appear at different frequencies due to their different nuclear spins. For Rb^{87} , $I = \frac{3}{2}$ and for Rb^{85} , $I = \frac{5}{2}$. If the field is sufficiently low, the individual Zeeman resonances for each isotope will be unresolved.

The theory developed is based upon a model which neglects the nuclear spin of both isotopes. That is, the optical pumping signals are assumed to depend only upon the spin polarization of the optical electrons of the Rb^{87} atoms. Spin exchange results in a change of the Rb^{87} electron polarization through Rb^{85} - Rb^{87} collisions. This model is expected to be a good approximation to the more complicated situation in which nuclear spin is considered, when the individual Zeeman resonances for each isotope are unresolved and thus detected simultaneously. Other assumptions used in this theory

¹³ An absorption cell 1.4 cm long was used in the present experiment. No absorption of Rb^{87} radiation was observed in this cell at room temperature. An absorption cell of sufficient length would of course yield measurable absorption at room temperature. A thin cell was chosen so that direct comparison of the room temperature and 90°C spectral traces could be made. The 70% absorption observed at 90°C represents a density in the cell which is about a factor of 100 higher than at room temperature.

are that the polarization achieved during the pumping process is low, in the range of a few percent, the ratio of the densities of the Rb⁸⁵ to Rb⁸⁷ vapors in the cell is given by the natural abundance in the metal, the relaxation rates in the ground state for both Rb⁸⁵ and Rb⁸⁷ are the same (this was shown to be the case experimentally), and that complete mixing in the excited state obtains due to the presence of a Ne buffer gas of 28 mm. The last assumption means that a Rb⁸⁷ atom in the excited state is randomly distributed amongst the excited state sublevels before decay to the ground state. The effect of pumping with D_1 light alone will be considered, which was the case experimentally.

Consider a cylindrical absorption cell of length L containing a vapor of natural Rb irradiated with a parallel beam of right-hand circularly polarized Rb⁸⁷ D_1 radiation (only Rb⁸⁷ will absorb). A weak dc magnetic field is applied to the cell in the direction of the incident radiation. Neglecting nuclear spin, and in low field at a temperature of about 90°C, the Rb⁸⁷ atoms are essentially equally distributed between the $^2S_{1/2}$ $M_J = +\frac{1}{2}$ and $M_J = -\frac{1}{2}$ sublevels. Only the atoms in the $M_J = -\frac{1}{2}$ sublevel can absorb the circularly polarized D_1 radiation since a $\Delta M_J = +1$ transition must be made. The rate of change of the light intensity $I(x)$ as a function of distance through the cell x is given by

$$[dI(x)/dx] = -N_- \sigma_0 I(x), \quad (4)$$

where σ_0 is the cross section for optical absorption and N_- is the number of atoms per cm³ in the $M_J = -\frac{1}{2}$ sublevel. Introducing the polarization of the Rb⁸⁷ vapor $P_1 = (N_+ - N_-)/N$, where N_+ is the number of atoms in the $M_J = +\frac{1}{2}$ state per cm³ and N the total number of Rb⁸⁷ atoms per cm³, the above becomes

$$[dI(x)/dx] = -(N\sigma_0/2)(1 - P_1)I(x). \quad (5)$$

This equation may be integrated over the length of the cell yielding

$$I = I_0 e^{-(N\sigma_0/2)L(1-\bar{P}_1)}, \quad (6)$$

where \bar{P}_1 is the average value of the polarization over the length of the cell, and I_0 is the intensity of the incident pumping radiation. The signal S is the difference in light intensity transmitted through the cell when an rf magnetic field at the Rb⁸⁷ Zeeman frequency is applied to the cell ($\bar{P}_1 = 0$) and when it is turned off. Under the assumption of low polarization ($\bar{P}_1 \leq 10^{-1}$), the signal is given by

$$S = -(N\sigma_0/2)LI_0 e^{-(N\sigma_0/2)L\bar{P}_1}. \quad (7)$$

The exchange signal S_x is the difference in the incident pumping radiation transmitted through the cell when an rf magnetic field at the Rb⁸⁵ Zeeman frequency is applied to the cell and when it is turned off. The average value of the Rb⁸⁷ polarization is different from \bar{P}_1 when this rf is applied. Let the new polarization be \bar{G}_1 . S_x is

given by

$$S_x = I_0 e^{-(N\sigma_0/2)L(1-\bar{G}_1)} - I_0 e^{-(N\sigma_0/2)L(1-\bar{P}_1)} \quad (8)$$

or

$$S_x = -(N\sigma_0/2)LI_0 e^{-(N\sigma_0/2)L(\bar{P}_1 - \bar{G}_1)} \quad (9)$$

under the assumption of low polarization. The ratio of the two signals is then

$$(S_x/S) = 1 - (\bar{G}_1/\bar{P}_1). \quad (10)$$

The average values of the Rb⁸⁷ polarization will now be calculated. First, consider the situation in which there is no interaction between the Rb⁸⁷ and Rb⁸⁵ vapors. In the absence of circularly polarized pumping radiation, the sublevels of Rb⁸⁷ are essentially equally populated at optical pumping temperatures in a field of a few gauss. Therefore, the relaxation rate for an atom going from the $M_J = +\frac{1}{2}$ to the $M_J = -\frac{1}{2}$ sublevel is equal to the rate for the reverse process. The rate equation for N_+ at a position x in the cell is given by

$$(dN_+/dt) = -N_+R + N_-R + \frac{1}{2}N_- \sigma_0 I(x), \quad (11)$$

where R is the relaxation rate and the factor of $\frac{1}{2}$ in the last term comes from the assumption of complete mixing in the excited state. With the introduction of spin exchange, atoms of Rb⁸⁷ and Rb⁸⁵ can exchange their electronic spin orientations during collision. The cross section for the process Q_x is the spin-exchange cross section to be experimentally determined. Only those collisions in which the Rb⁸⁵ and Rb⁸⁷ are initially in opposite spin states can give rise to changes in the populations of the Rb⁸⁵ and Rb⁸⁷ ground states' sublevels. The rate equations for Rb⁸⁷ and for Rb⁸⁵ with spin exchange are

$$(dN_+/dt) = -N_+R + N_-R + \frac{1}{2}N_- \sigma_0 I(x) + N_- n_+ v_r Q_x - N_+ n_- v_r Q_x, \quad (12)$$

$$(dn_+/dt) = -n_+R + n_-R + n_- N_+ v_r Q_x - n_+ N_- v_r Q_x, \quad (13)$$

where n_+ and n_- are the populations of the $M_J = +\frac{1}{2}$ and $M_J = -\frac{1}{2}$ ground-state sublevels of Rb⁸⁵ and v_r is the relative velocity of Rb⁸⁵ and Rb⁸⁷ atoms. The terms involving spin exchange in the rate equations neglect the velocity distribution (and thus relative energy distribution) of the members of the collision. The simplification is made because of the unknown energy dependence of Q_x . The v_r used will be the root mean square relative velocity.¹⁴ All atoms of Rb⁸⁵ will be taken to have the same velocity, i.e., that velocity which will give the correct total energy of the Rb⁸⁵ vapor at a temperature T and likewise for the Rb⁸⁷ atoms. Let the Rb⁸⁵ polarization be defined by $P_2 = (n_+ - n_-)/n$ where n is the Rb⁸⁵ density. Substituting P_1 , P_2 , and $a = n/N$, the ratio of Rb⁸⁵ to Rb⁸⁷ vapor densities, the

¹⁴ The choice of the root mean square velocity is somewhat arbitrary. Others might be inclined to choose the mean velocity. The present choice was made to be consistent with the early work of Dehmelt (Ref. 3). No consistent convention appears in the literature, however.

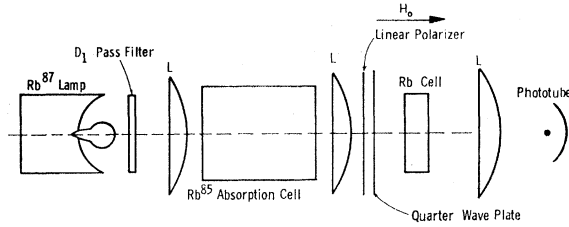


FIG. 1. Experimental arrangement for making optical pumping signal measurements. A weak dc magnetic field $H_0=3.6$ G is applied to the Rb cell parallel to the pumping light axis. Only the Rb D_1 radiation capable of being absorbed by Rb 87 reaches the cell.

rate equations become

$$\begin{aligned} (dP_1/dt) = & [\sigma_0 I(x)/2] - \{[\sigma_0 I(x)/2] + 2R\} P_1 \\ & - aNv_r Q_x P_1 + aNv_r Q_x P_2 \end{aligned} \quad (14)$$

$$(dP_2/dt) = -2RP_2 - Nv_r Q_x P_2 + Nv_r Q_x P_1. \quad (15)$$

To calculate S_x/S , we need the steady-state average values of the polarization of Rb 87 when P_2 , the Rb 85 polarization, is at its steady-state value and when $P_2=0$ (equal population of the ground-state sublevels of Rb 85 is forced by application of an rf magnetic field at the Rb 85 frequency). The former is \bar{P}_1 and the latter is \bar{G}_1 . In order to calculate these, the light distribution $I(x)$ must be known. $I(x)$ itself depends upon the Rb 87 polarization. However, a first-order solution for the polarization may be obtained through substitution of the zero-order light distribution, i.e., the light distribution in the absence of polarization. This is valid under the assumption of low polarization. The polarizations are:

$$\bar{G}_1 = \frac{1}{L} \int_0^L \frac{(\sigma_0 I_0/2) e^{-N\sigma_0 x/2}}{\frac{\sigma_0 I_0}{2} e^{-N\sigma_0 x/2} + 2R + aNv_r Q_x} dx \quad (16)$$

$$\bar{P}_1 = \frac{1}{L} \int_0^L \frac{(\sigma_0 I_0/2) e^{-N\sigma_0 x/2}}{\frac{\sigma_0 I_0}{2} e^{-N\sigma_0 x/2} + 2R + \frac{Nv_r Q_x (2aR)}{Nv_r Q_x + 2R}} dx. \quad (17)$$

Upon substituting the polarizations in $S_x/S = 1 - \bar{G}_1/\bar{P}_1$ and performing the integrations the following relation is obtained for low polarization.

$$\begin{aligned} (Nv_r Q_x/R) = & [(1+a)/a][S_x/S/(1-S_x/S)] \\ & + \left[\left(\frac{1+a}{a} \right)^2 \left(\frac{S_x/S}{1-S_x/S} \right)^2 + \frac{4}{a} \left(\frac{S_x/S}{1-S_x/S} \right) \right]^{\frac{1}{2}}. \end{aligned} \quad (18)$$

Using this relation, the ratio of $Nv_r Q_x$ to R can be determined by a measurement of the ratio of exchange to direct optical pumping signals and the known vapor density ratio. N can be measured by the interferometric

technique described, and v_r can be calculated using a measurement of the cell temperature.

R must be determined in order to obtain Q_x . This may be accomplished by observing the time dependence of the signal at a reduced temperature. Since the exchange rate $Nv_r Q_x$, is directly proportional to the density, by reducing the density to low enough value, $Nv_r Q_x$ may be neglected in comparison to R . Since the signal S is directly proportional to the Rb 87 polarization P_1 , for low polarization, R may be determined. Neglecting the exchange terms,

$$(dP_1/dt) = [\sigma_0 I(x)/2] - \{[\sigma_0 I(x)/2] + 2R\} P_1. \quad (19)$$

With rf on at the Rb 87 frequency, $P_1=0$. With the initial condition of $P_1=0$, the solution is

$$P_1 = P_0(1 - e^{-t/T})$$

$$P_0 = \frac{[\sigma_0 I(x)/2]}{[\sigma_0 I(x)/2] + 2R}$$

$$1/T = [\sigma_0 I(x)/2] + 2R. \quad (20)$$

The pumping rate $[\sigma_0 I(x)/2]$ at a given position in the cell is directly proportional to the incident intensity I_0 . By observing the time dependence of the signal at two values of incident light intensity, e.g., I_0 and $I_0/2$, R may be determined. With this additional information Q_x may be determined.

IV. EXPERIMENTAL ARRANGEMENT

Two separate types of measurements were involved in determining the cross section. These were measurements of the strengths and time dependence of optical pumping signals and spectroscopic measurements to determine the vapor density. Standard circuitry and techniques were used in both sets of measurements and will be described briefly.

Figure 1 shows the experimental arrangement for making the optical pumping signal measurements. An interference filter isolated the D_1 line from a Varian spectral lamp using an enriched Rb 87 bulb. The D_1 line was prefiltered before entering the optical pumping cell containing natural Rb. A filter cell containing 99% pure Rb 85 was used to absorb the Rb 87 light capable of being absorbed by the Rb 85 in the optical pumping cell as well as that light emitted by any Rb 85 poisoning in the lamp bulb. Polaroid linear polarizer and quarter wave plate rendered the light circularly polarized before entering the optical pumping cell. The transmitted light was monitored by an RCA 917 phototube. A pulsed rf technique was used to induce the Zeeman resonances. Phase sensitive detection was used in the measurements of the signals which were displayed on a chart recorder. Wide band detection was used in studying the time dependence of the signal to determine R . The signal was displayed on a CRO and photographed. Ten exposures were taken per frame to average the noise.

The density was measured using the interferometric technique. The Rb⁸⁵ filter cell, the linear polarizer, and the quarter wave plate were removed from the experimental arrangement of Fig. 1. A mirror (not shown) which deflected the transmitted light beam to the phototube was also removed which allowed the transmitted light to enter the scanning Fabry-Perot interferometer. The light beam was mechanically chopped at 150 cps. The center fringe of the interference pattern was monitored by a liquid air cooled RCA 7102 photomultiplier as air was slowly leaked back into the evacuated chamber containing the Fabry-Perot etalon. Phase sensitive detection was used and a permanent record of the spectral profiles of the Rb⁸⁷ D_1 line obtained by displaying the signal on a chart recorder.

V. RESULTS

Preliminary to the measurement of Q_x , studies were made of the feasibility of optically pumping the Rb⁸⁷

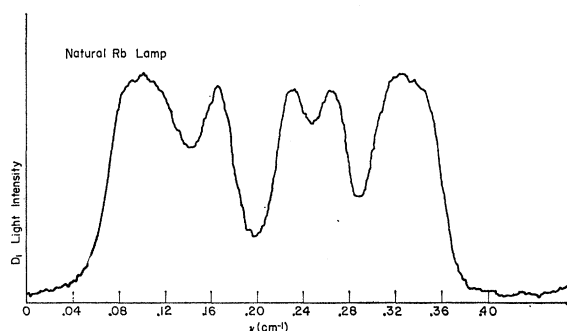


FIG. 2. The Rb D_1 line emitted by a Varian spectral lamp containing natural Rb metal. The peaks on the extremities of the line are due to the two Rb⁸⁷ hyperfine components. The center is due mainly to the two Rb⁸⁵ hyperfine components.

component in a vapor of natural Rb. The first study was to examine qualitatively the dependence of both the Rb⁸⁷ and Rb⁸⁵ signals on temperature or density. In a vapor of natural Rb, Rb⁸⁵ has 2.59 times the density of Rb⁸⁷. If the direct optical signals for both isotopes were being observed, the Rb⁸⁵ signal maximum would be attained at a lower temperature than the Rb⁸⁷, due to its higher concentration in the vapor. While the Rb⁸⁷ signal is approaching its maximum, the Rb⁸⁵ signal will decline in strength as less Rb⁸⁵ light is transmitted through the cell due to the higher density of Rb⁸⁵. Thus, qualitatively, a decreasing ratio of Rb⁸⁵ to Rb⁸⁷ signal strengths would be expected as the temperature of the cell is increased. If an Rb⁸⁵ exchange signal is in fact being detected, the opposite would be expected since the Rb⁸⁵ signal depends upon the number of Rb⁸⁵-Rb⁸⁷ collisions in the vapor, which increases with increasing density (or temperature). With the pure Rb⁸⁵ filter cell in place in the apparatus, the latter situation was encountered. Upon removal of this cell, the former

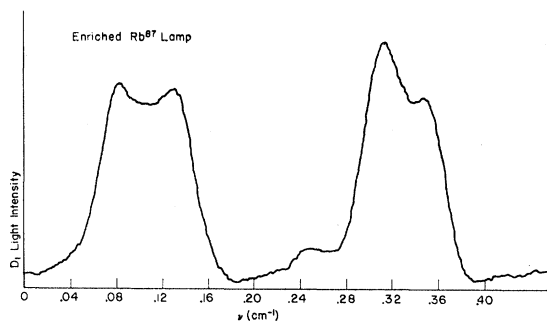


FIG. 3. The Rb D_1 line emitted by a Varian spectral lamp containing enriched Rb⁸⁷ metal. The component on the left arises from transitions to the $F=2$ sublevel of the ground state and the component on the right from transitions to the $F=1$ sublevel.

situation was encountered. A more direct way of showing that Rb⁸⁷ alone was optically pumped was to examine the pumping light with the scanning Fabry-Perot interferometer. Figures 2, 3, and 4 show the results of this investigation. Figure 2 is the D_1 line in emission from a Varian spectral lamp using a bulb containing natural Rb. The complex structure arises from the hyperfine structure of both isotopes. Figure 3 shows the D_1 line using an enriched Rb⁸⁷ bulb. A poisoning of Rb⁸⁵ is apparent next to the high-frequency Rb⁸⁷ hyperfine component. Also, Rb⁸⁵ is capable of absorbing the high-frequency side of the low-frequency Rb⁸⁷ hyperfine component. Figure 4 shows the action of the Rb⁸⁵ filter cell. The Rb⁸⁵ poisoning is gone and the high-frequency side of the low-frequency Rb⁸⁷ hyperfine component is attenuated. This measurement indicates that the light intensity available for direct pumping of Rb⁸⁵ and Rb⁸⁷ would provide a ratio of these direct pumping signals no larger than 1:225 (the signal is proportional to the square of the incident light intensity). The ratio observed under these pumping conditions was actually 1:9. Therefore, direct optical pumping cannot be responsible for the relatively large Rb⁸⁵ signals measured. It is felt that the contribution of direct pumping is negligible.

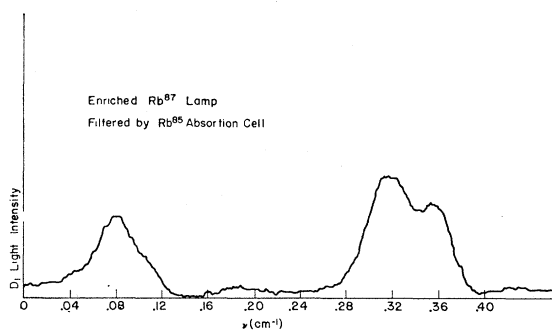


FIG. 4. The Rb D_1 line emitted by a Varian spectral lamp containing enriched Rb⁸⁷ metal after being filtered through a cell containing Rb⁸⁵ vapor. The component on the left arises from transitions to the $F=2$ sublevel of the ground state and the component on the right from transitions to the $F=1$ sublevel.

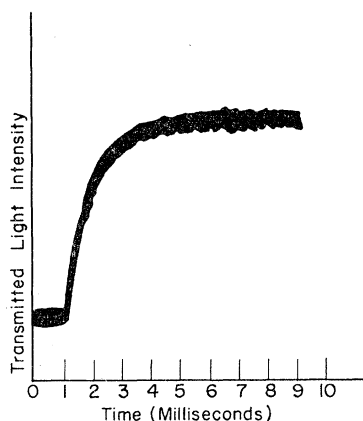


FIG. 5. A reproduction of a photograph showing the time dependence of the transmitted light intensity (the optical pumping signal). Pumping action of the light begins at about 1 msec.

Measurements of S_x/S at 90°C yield a result of (0.107 ± 0.001) . These measurements were made at low light levels and, therefore, low polarization corresponding to the assumptions in the signal calculations. Figure 5 shows a photograph used in measuring R . Figure 6 shows a plot of the $\ln(1 - P_1/P_0)$ versus time for incident I_0 and $I_0/2$. These data were obtained at 50°C . Four points were plotted for each trace and the slopes determined by a least-squares fit. Measurements were also made on Rb^{87} at 60°C and the direct optical pumping signal for Rb^{85} at 57°C . All experimental values of R determined from these measurements agreed to within 5%. It was concluded that to within this accuracy, R for Rb^{85} and Rb^{87} were equal. The results yield $R = (413 \pm 21) \text{ sec}^{-1}$.

Measurements on the spectral traces of the transmitted $\text{Rb}^{87} D_1$ high-frequency hyperfine component at room temperature and at 90°C yield a density $N = (3.33 \pm 0.37) \times 10^{11} \text{ atoms/cm}^3$. At 90°C , the peak absorption was about 70%. The half-width of the absorption line was measured to be 0.039 cm^{-1} . This is

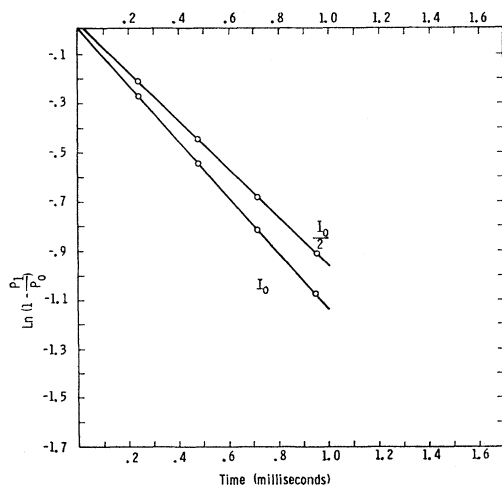


FIG. 6. Graph used to determine relaxation time. Two straight lines shown are for two different (I_0 and $I_0/2$) values of the pumping light intensity.

about twice the Doppler width of the absorption line. However, it must be noted that the hyperfine line actually contains two components in the intensity ratio of 5 to 1 separated by 0.025 cm^{-1} . Also, the absorption occurred in an atmosphere of 28 mm of Ne. Apparently no information is available on broadening of the Rb resonance lines in Ne, but Ch'en¹⁵ has measured the broadening of Rb lines in argon. The result for argon indicates a half-width due to pressure broadening of 0.023 cm^{-1} . Under the assumption that Ne produces a similar width, the present half-width measurement seems reasonable. Other structures in emission were measured to have a half-width of 0.030 cm^{-1} , showing that the width observed is not predominantly instrumental. The theoretical resolving limit was calculated to be about 0.008 cm^{-1} . An estimated value for the resolving limit was taken to be about 0.019 cm^{-1} , halfway between the limits of 0.008 cm^{-1} and 0.030 cm^{-1} . Calculations of finite resolution corrections to integrated

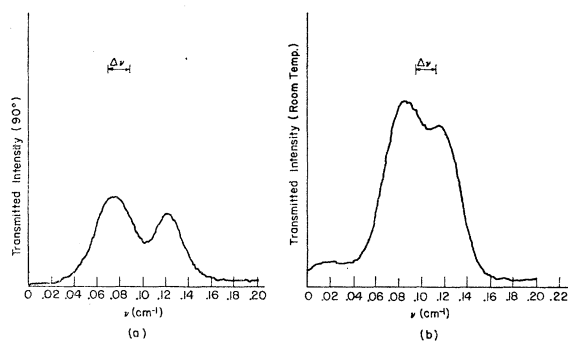


FIG. 7. Spectral profiles of high-frequency hyperfine component of the $\text{Rb}^{87} D_1$ line transmitted through the optical pumping cell at (a) 90°C and (b) room temperature. $\Delta\nu$ is approximate resolving limit of the Fabry-Perot interferometer. These traces were used to determine Rb^{87} vapor density.

absorption coefficient measurements reported by Kostkowski and Bass¹⁶ indicate a correction of +3% with an uncertainty of $\pm 2\%$ due to the uncertainty in the width and shape of the bandpass function for the interferometer. This correction was applied to the raw data. Other sources of error were the uncertainty in the actual zero of light on the spectral traces, signal to noise considerations, and the uncertainty in the lifetime measurements. Figure 7 shows an example of traces which are compared to yield the integrated absorption coefficient.

Other numerical data needed to calculate the cross section are $a = 2.59$ obtained from the natural abundance percentages for Rb^{85} and Rb^{87} , 72.15% and 27.85%,¹⁷ respectively, and $v_r = (4.59 \pm 0.05) \times 10^4$

¹⁵ S. Y. Ch'en, Phys. Rev. 58, 1051 (1940).

¹⁶ H. J. Kostkowski and A. M. Bass, J. Opt. Soc. Am. 46, 1060 (1956).

¹⁷ Handbook of Chemistry and Physics (Chemical Rubber Publishing Company, Cleveland, 1960), 41st ed., p. 463.

cm/sec. The uncertainty given for v_r is due to an allowed $\pm 10^\circ\text{C}$ uncertainty in the cell temperature as measured by a mercury thermometer placed in the oven containing the optical pumping cell.

The data of this experiment yields a spin-exchange cross section for Rb⁸⁵-Rb⁸⁷ collisions of $(1.70 \pm 0.21) \times 10^{-14}$ cm². The early work of Franken, Sands, and Hobart⁵ and Novick and Peters⁶ yielded cross sections of 5×10^{-14} cm² for Na-K collisions and 2×10^{-14} cm² for Na-Rb⁸⁵ collisions. These results were considered to be reliable within a factor of 3 of the quoted values. The uncertainty was primarily due to unreliable density estimates. With the use in the present work of the interferometric technique of density measurement, estimated to be reliable to $\pm 11\%$, this type of uncertainty was largely overcome. The total error quoted for the cross section is closely related to a 70% confidence interval. The result presented is in reasonable agreement with measurements of Rb⁸⁵-Rb⁸⁵ and Rb⁸⁷-Rb⁸⁷ spin-exchange collision cross sections performed

by Moos and Sands¹⁸ in this laboratory using an electron paramagnetic resonance technique.

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¹⁸ T. Stark and R. H. Sands have kindly informed us by private communication of recent results for both isotopes of Rb based on the Ph.D. thesis of H. W. Moos at the University of Michigan, 1961 (to be published).

Interaction of Optical and Infrared Radiation with Metastable Hydrogen Atoms

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This paper is an extension of a previous one and discusses the theory of the quenching of the metastable 2S state of atomic hydrogen by means of optical radiation, for example by the light from a ruby laser. The case discussed is that for which the incident intensity is sufficiently weak for the usual quantum-electrodynamical perturbation theory to be valid. A procedure developed by Schwartz and others is used to carry out the sum over intermediate states without explicit enumeration. The results are given for a range of incident wavelengths from 5000 Å to 50 μ. For unpolarized light from a ruby laser (6934 Å), the total cross section for quenching is found to be $\sigma_Q = 1.27 \times 10^{-22}$ cm². The cross section for coherent scattering has also been calculated for the same range of wavelengths; for ruby laser light, the total cross section for scattering is found to be $\sigma_S = 1.03 \times 10^{-23}$ cm².

1. INTRODUCTION

IN a previous paper,¹ hereinafter referred to as I, an approximate calculation was reported for the quenching of the atomic hydrogen 2S state by means of the light from a ruby laser. It was concluded that, (a) the process can be analyzed by means of the usual quantum-electrodynamical perturbation theory² provided that the peak electric-field strength in the laser beam is less than about 10^7 V/cm, and that if this is the case, then, (b) the process consists mainly of virtual excitation to the 3P state followed by spontaneous decay to the 1S ground state.

In this paper the treatment is restricted to the weak-field case but all possible intermediate states, including those in the continuum, are taken into account.

¹ W. Zernik, Phys. Rev. **132**, 320 (1963).

² See, for example, W. Heitler, *The Quantum Theory of Radiation* (Clarendon Press, Oxford, 1954), 3rd ed.

In addition, numerical results for the quenching cross section are given for a range of incident wavelengths from 5000 Å to 50 μ, essentially covering the range of currently available laser frequencies. The maximum intensity of the incident radiation for which the results are valid is a function of the frequency, as explained in I. In particular, the theory breaks down completely in the neighborhood of the Balmer frequencies.

2. PERTURBATION THEORY RESULTS

As indicated in I, second-order perturbation theory yields a cross section for quenching given by:

$$\frac{d\sigma}{d\Omega} = r_0^2 \left\{ k_1^3 k_0 \left| \sum_i \frac{(\mathbf{e}_0 \cdot \mathbf{r}_{i0})(\mathbf{e}_1 \cdot \mathbf{r}_{fi})}{E_0 - E_i + k_0} + \frac{(\mathbf{e}_1 \cdot \mathbf{r}_{i0})(\mathbf{e}_0 \cdot \mathbf{r}_{fi})}{E_0 - E_i - k_1} \right|^2 \right. \\ \left. + k_2^3 k_0 \left| \sum_i \frac{(\mathbf{e}_0 \cdot \mathbf{r}_{i0})(\mathbf{e}_2 \cdot \mathbf{r}_{fi})}{E_0 - E_i - k_0} + \frac{(\mathbf{e}_2 \cdot \mathbf{r}_{i0})(\mathbf{e}_0 \cdot \mathbf{r}_{fi})}{E_0 - E_i - k_2} \right|^2 \right\}. \quad (1)$$