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OPTICAL PUMPING TRANSIENTS IN RUBIDIUM-87
AND APPLICATION TO DISORIENTATION CROSS SECTIONS
Richard Marrus and Joseph Yellin

June 23, 1965
Optical Pumping Transients in Rubidium-87
and Application to Disorientation Cross Sections *

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ABSTRACT

Detailed studies have been made of the light transmitted through rubidium-87 vapor during the optical pumping process, both with and without buffer gas present. The observed transients are single exponentials with no buffer gas present, and double exponentials with buffer gas present over a wide range of pumping intensities and relaxation times. These results are in excellent agreement with the predictions based on phenomenological equations in which nuclear spin is included and a single relaxation time is assumed. From a study of the amplitudes of the two exponential components of the optical-pumping transient, an effective cross section $\sigma_{\text{eff}}$ is deduced for the disorientation of rubidium-87 within the $5^2P_{1/2}$ state as a result of its collision with a buffer-gas atom. It is shown that $\sigma_{\text{eff}} = \sigma_{1/2} + \sigma_{3/2}$, where $\sigma_{1/2}$ is the cross section for disorientation within the $5^2P_{1/2}$ level, and $\sigma_{3/2}$ is the cross section for transfer from the $5^2P_{1/2}$ level to the $5^2P_{3/2}$ level by means of collisions with the buffer gas. On the basis of recently measured values for $\sigma_{3/2}$, values are deduced for $\sigma_{1/2}$. The cross sections are: $\sigma_{1/2} (\text{Rb-He}) = 1.5 (0.8) \times 10^{-17}$ cm$^2$, $\sigma_{1/2} (\text{Rb-Ne}) = 4.4 (2.2) \times 10^{-17}$ cm$^2$, and $\sigma_{1/2} (\text{Rb-Ar}) = 3.5 (1.8) \times 10^{-16}$ cm$^2$. These cross sections are deduced from a model in which the probabilities for Rb$^{87}$ to relax from any hyperfine level to any other are all equal.
This model gives a better fit with the observed transients than the assumption that the electron spin only is randomized in the $P_{1/2}$ state with the nuclear spin unaffected.

We show that accurate relaxation-time measurements can be made by measuring the time constants associated with the double exponentials as a function of light intensity, and extrapolating them to zero light intensity. Moreover, since no shutter is required, relaxation times $< 10^{-3}$ sec can be easily observed. From a study of the relaxation times as a function of buffer-gas pressure, disorientation cross sections ($\sigma_{G.S.}$) have been obtained in the ground state ($5^2S_{1/2}$).

The measured cross sections are:

$$\sigma_{G.S.} (\text{Rb-Ar}) = 3.3(1.0) \times 10^{-22} \text{ cm}^2 \quad \sigma_{G.S.} (\text{Rb-Ne}) = 1.0(0.3) \times 10^{-22} \text{ cm}^2$$

$$\sigma_{G.S.} (\text{Rb-Xe}) = 1.8(0.3) \times 10^{-19} \text{ cm}^2 \quad \sigma_{G.S.} (\text{Rb-Kr}) = 3.0(0.5) \times 10^{-20} \text{ cm}^2$$

The cross sections for krypton and xenon were measured directly. Comparison of these cross sections with those obtained by other methods is made.
I. INTRODUCTION

Although the principles that determine the shape of optical pumping transients have been discussed for a long time, there has been no serious attempt to study them experimentally. On the theoretical side, several authors have written down the rate equations governing the shape of the transients and for a particular case they have solved them numerically. Experimentally, there have been some attempts to extract information of physical interest from the pumping transients. Dehmelt made the first relaxation-time measurements based on the shape of the transients in the limit of zero light intensity, and Jarrett used the transients in connection with a measurement of the $\text{Rb}^{85}$-$\text{Rb}^{87}$ spin-exchange cross section. However, both these authors neglect nuclear spin, a doubtful assumption at best. Bloom showed that there are generally large uncertainties in relaxation times determined by the Dehmelt method.

Information concerning the amount of reorientation in the excited state is also contained in the shape of the transients. This was noted by Bender, who observed decreases in the alkali polarization at high buffer-gas pressure. In this work we are concerned with showing the following: First, the rate equations give a quantitatively accurate description of pumping transients under the experimental conditions in which they should be valid. Second, quantitative information can be obtained concerning disorientation in the excited $P_{1/2}$ state. Third, accurate relaxation times can be obtained from the pumping transients by means of an extrapolation to zero light intensity. This method, in which a shutter is not employed can be used to advantage in the range $10 \text{ msec} > T > 0.1 \text{ msec}$. The widely employed method developed by Franzen is generally limited to the range $>10 \text{ msec}$. Hence the range
over which relaxation-time measurements can be made is considerably extended. As an application of this method, we have remeasured the ground-state disorientation cross sections for Rb with the various noble gases.

II. EXPERIMENTAL METHOD

The basic apparatus is the standard one used in optical pumping experiments in which the pumping radiation is circularly polarized $D_1$ light and the transmitted light is monitored. A diagram of the apparatus is shown in Fig. 1 along with the associated electronics. The lamp was the Varian X49-609 resonance lamp with a bulb made of separated Rb isotopically enriched to 99.66%; the resonance cell also contained isotopically enriched Rb. Although the walls of the cell were coated with Paraflint, no special care was taken to achieve very long relaxation times. The cell was mounted on a vacuum system and connected to a gas-flow system capable of admitting any buffer gas to the desired pressure as measured by either a mercury or an oil manometer. The transmitted light was monitored by a solar-cell detector.

In order for the validity of the rate equations to be studied, several conditions must obtain in the resonance cell: (a) The optical path length must be long compared with the dimension of the cell; this requires extremely low densities of absorbing atoms. To assure this condition, the light absorbed by the cell was measured, and only cells with total absorption $< 5\%$ were used. Cells with absorption of more than 5% had already begun to show transient distortion which could be explained by optical thickness. (b) The time for self-spin exchange collisions between rubidium atoms ($T_{ex}$) must be long compared with $T_1$. The requirement is that $T_{ex} \gg T_1$ or $1/\sigma_{ex} \gg T_1$. Taking
$T_1 \approx 0.1 \text{ sec and } \nu = 4 \times 10^4 \text{ cm/sec, and using the value of } 10^{-14} \text{ cm}^2$

for $\sigma_{ex}$ measured for Rb$^{87}$-Rb$^{87}$ collisions by Moos and Sands, we obtain $n \ll 10^{11} / \text{cm}^3$ as the condition for neglecting self-spin exchange. This requirement is also adequate to ensure that mixing of the $^2P_{1/2}$ and $^2P_{3/2}$ states by collisions with rubidium atoms in the ground state will not occur. From total absorption measurements we estimate a density of $\approx 10^9 \text{ atoms/cm}^3$ at our operating point at room temperature.

For the emission-line width, we have assumed Jarrett's Rb$^{87}$ line profile, which was obtained from a similar lamp. Apart from the above two conditions, there are others that are important and that we will consider at the proper place in this paper.

Unfortunately, operating at room temperature at such low densities means very small signals except in the most favorable cases (long relaxation time, high light intensity, and no reorientation in the excited state). The usual technique of displaying the transient on an oscilloscope screen and photographing it would yield no quantitative information. In order for a large signal-to-noise ratio to be achieved through the accumulation of data, a multichannel pulse-height analyzer was employed (Fig. 1, block diagram). By this means an arbitrarily large signal-to-noise ratio may be obtained, although as a practical matter 1000:1 was deemed sufficient.

Each channel of the analyzer corresponds to a definite time after initiation of the transient, and accumulates data for an interval equal to the reciprocal of the sampling rate. The sampling rate was determined by the time-base oscillator which switched channels linearly with time, establishing a correspondence between each channel and the time; e.g., the 30th channel is always reached at the same time in the evolution of the transient. Once the transient saturated, the rf gate was
turned on and the Zeeman levels were equalized by a rf pulse whose duration is long compared with the relaxation time. When the analyzer completed its sweep, the gate was turned off and a new transient allowed to begin simultaneously with the next sweep of the analyzer. The cycle was repeated many times a minute (the number depending on the lifetime of the transient and the relaxation time) for as long as was necessary to get the required signal-to-noise ratio. The digital output of the analyzer was fed into a computer for least-squares analysis.

It was important to have enough rf power to saturate the resonance, for otherwise the initial populations would not be equalized. To determine the rf power level, we made a plot such as is shown in Fig. 2. Here the transmission of light by the polarized vapor was measured as a function of rf current for the maximum light intensity anticipated. The plot clearly shows saturation of the resonance. A simpler though adequate determination can be obtained from the rf-induced relaxation. The power level was adjusted so that the transient decays in a time $T_{rf} < T_{pumping}$.

Successive signals were almost identical, the differences being due to (a) random noise, (b) nonrandom noise, and (c) instrumental drift. The random noise obviously averages to zero whereas nonrandom noise may lead to systematic errors if it is phase locked to the sweep of the analyzer; but phase locking can usually be avoided by proper selection of the sweep rate. The chief source of drift was the light source whose intensity varied by 1 to 3% over the data-accumulation time. Only those transients were used for which the variation in light intensity during a run was $< 3\%$. 
In this section, we consider the equations that determine the shape of the optical pumping transients under the conditions described in Secs. 1 and 2. As shown there, considerable care was taken to ensure that the following conditions pertained: (a) the optical path length was long enough that the light absorbed was proportional to the number of absorbing atoms; (b) the alkali density was low enough that the effect of Rb\textsuperscript{87} - Rb\textsuperscript{87} exchange collisions could be neglected. Under these conditions, the shape of the pumping transients is determined by the rate equations.

\[
\frac{dp_i}{dt} = -p_i \sum_j b_{ij} + \sum_j b_{ij} p_j - p_i \sum_j \frac{1}{T_{ij}} + \sum_j p_j \frac{1}{T_{ji}}
\]

(1)

where \( p_i \) is the occupation probability of the \( i \)th hyperfine sublevel of the ground state; \( b_{ij} \) is the probability for an atom to be transferred from the \( i \)th to the \( j \)th sublevel by photon absorption and subsequent reemission; and \( T_{ij} \) is the relaxation time between the sublevels \( i \) and \( j \) as a result of collisions with the buffer gas and with the cell walls. For \( I = 3/2 \), these equations can be put into the form

\[
\frac{dp_i}{dt} = \frac{\beta_0}{8} \sum_{i=1}^{8} B_{ik} p_i + \frac{1}{8T}
\]

(2)

where \( \beta_0 \) is the average light-absorption probability per atom when the ground levels are equally populated, i.e., \( \beta_0 = \frac{1}{8} \sum_{k,j} b_{kj} \); and

\[
B_{ik} = \frac{8}{\beta_0} b_{ik}, \quad i \neq k, \quad B_{kk} = -\frac{8}{\beta_0} \left[ \frac{1}{T} + \sum' b_{kj} \right]
\]

where \( \sum' \) denotes a sum over all levels except the level \( k \). It is assumed that the relaxation time \( T_{ij} \) is the same for all levels \( i \) and \( j \) with \( T = 1/8 T_{ij} \). The assumption of a single relaxation time is supported by the fact that the
Franzen-type transients observed by many workers in the study of ground-state relaxation phenomena are very well fitted by single exponentials.

The photon flux at the center of the absorption line is taken as a constant independent of the levels being pumped. The most accurate measurements bearing on this point are the line-profile measurements taken by Jarrett on the Varian lamp with an Rb$^{87}$ bulb. His measurements indicate that the light intensity from each of the hyperfine components are nearly equal. Hence we believe no serious error is made when the flux is taken to be the same for the two hyperfine states. These equations (2) have as solutions the form

$$p_i = \sum_{j=1}^{g} A_j e^{-t/\tau_j} + C_1$$

where the reciprocals of the time constants $\tau_j^{-1}$ are determined as eigenvalues from the matrix $B_{ik}$. For the extreme cases of no mixing in the excited state and complete mixing in the excited state, these matrices have been written down by Franzen and Emslie. Of interest to us here is the intermediate case when the excited-state mixing is incomplete. Two processes are important. First, collisions with buffer-gas atoms can cause transfer of excited alkali atoms from the $P_{3/2}$ to the $P_{1/2}$ state (characterized by cross section $\sigma_{3/2}$). Second, collisions with buffer-gas atoms can cause mixing among the $P_{1/2}$ substates ($\sigma_{1/2}$). We assume that the probability for an atom to go from one magnetic substate to any other as the result of a buffer-gas collision is independent of the substate. With this assumption we can define an average probability $\alpha$ for an atom in an excited sublevel to be transferred to
another excited sublevel as a result of collisions with buffer-gas atoms.

We obtain \( f n(1 - \alpha) = n \overline{v} T_{1/2} \sigma_{\text{eff}} \), where \( \sigma_{\text{eff}} = \sigma_{1/2} + \sigma_{3/2} \). \( n \) is the density of buffer-gas atoms, \( \overline{v} \) is the average relative velocity, and \( T_{1/2} \) is the lifetime of \( P_{1/2} \). In terms of \( \alpha \), the matrix \( B_{ik} \) that describes the situation is simply \( (1 - \alpha)B_{ik}^{\text{nm}} + \alpha B_{ik}^{\text{cm}} \), where \( B_{ik}^{\text{nm}} \) is the matrix for no mixing and \( B_{ik}^{\text{cm}} \) is the matrix for complete mixing. It follows that the time constant \( \tau^i \) is given by

\[
\frac{1}{\tau^i} = (1 - \alpha) \frac{1}{\tau_{\text{nm}}} + \alpha \frac{1}{\tau_{\text{cm}}}
\]

where the subscripts refer to no mixing and complete mixing. Cases other than the one in which the mixing probability in the excited state is independent of substate are of interest.

The solution of Eq. (1) and the determination of the parameters \( A_i, C_j, \) and \( \tau_j \) were done on the IBM 7090 with the initial condition of equal population in all the magnetic sublevels. We defer until Sec. IV a discussion of these results, however.

IV. DATA ANALYSIS

A. Relaxation-Time Measurement

If we are to adapt the foregoing equations to the observed transients, we must introduce values for the three parameters—the ground-state relaxation time \( T \), the average light-absorption probability \( \beta_0 \), and the probability for depolarization in the excited state \( \alpha \). The ground-state relaxation time can be easily determined after noting that in the limit \( \beta_0 \to 0 \), the time constants \( \tau_j \) all approach the value \( T \). It is also easy to see that the time constants are inversely proportional to \( \beta_0 \) and therefore to the light intensity. Hence our procedure was to observe the
transients at several different values of the light intensity. The light intensity was varied by putting flat glass plates (each of about 15% attenuation) between the lamp and the resonance cell. The relative light intensity was then measured by means of the dc output of the solar cell. Next the transients were resolved into their exponential time constants, and for a particular cell the inverse time constants were plotted vs the light intensity. The resultant straight lines were then extrapolated to zero intensity. In Fig. 3, we show the results for two different bulbs with no buffer gas but each with a wall coating of Paraflint. The wall coating of the two bulbs differs in quality, and the relaxation times are considerably different. For both bulbs the pumping transients were single exponentials to a high degree of accuracy for all light intensities. There was no trace of a second component (see Fig. 6).

In Fig. 4 we show the results for three cells containing buffer gas. The shape of the pumping transients was found to be very precisely double exponentials for all values of the light intensity. As detailed in Sec. IV. B, the transients were separated into their exponential components by computer and the resulting inverse time constants were plotted vs the light intensity. That both exponentials of these cells converge to the same value at zero intensity is predicted by the rate equations and was always observed.

An inspection of the graphs shows that this method gives quite reliable results for relaxation-time measurements in the range $10 \text{ msec} > T > 1 \text{ msec}$. The Franzen method requires shutters whose opening and closing times are fast compared to $T$ and cannot be conveniently applied in this range. In Fig. 5 we show the pumping transient for $\text{Rb}^{87}$ vapor in a cell with no buffer gas and no wall coating. The time constant (0.6 msec) of the exponential is very close to the relaxation time.
From this value it is possible for one to deduce an average number of wall collisions for disorientation to take place; on the average, about 1.4 collisions with the wall are needed. The deviation from 1 might possible arise from a partial wall coating of diffusion-pump oil back-streaming into the resonance cell. In any case, it is apparent that the assumption of "zero polarization at the walls" is a good one, since on the average an atom bounces many times after the first bounce before traveling many-mean-free paths away.

B. Determination of $\beta_0$ for Cells with No Buffer Gas

For cells containing no buffer gas there is no relaxation in the excited state and the only parameters are $\beta_0$ and $T$. The relaxation time may be determined as in Sec. IV.A. To see if the theory is valid for this case, we have chosen to proceed in the following way. Although we were prepared to make multiexponential fits to the observed data, it turns out, remarkably enough, that the data can be fitted very precisely by a single exponential. The transients for a number of cells corresponding to several relaxation times and different light intensities were fitted to single exponentials. For a particular cell, we then chose a value of $\beta_0$ that made the theory give a best fit for the data corresponding to an arbitrary value of the light intensity. For any other setting of the light intensity, the value of $\beta_0$ will then be determined by means of the voltage output of the solar cell. Thus for a given cell, the relaxation time is determined as described in Sec. IV.A, and $\beta_0$ is determined by the data at a single light intensity. Unfortunately, for two different cells it is not correct to say that the same voltage from the solar cell corresponds to the same $\beta_0$. The quantity $\beta_0$ is a measure of the photon flux through the vapor. Reflections on the faces of different cells can be considerably different.
Since it is clearly desirable to calibrate the lamp for all cells with one measurement, we have attempted to minimize this difference by using similar construction and flat faces for all the cells. In Fig. 6 we compare the experimental and theoretical transients seen in two cells with different relaxation times; these are the two cells with relaxation times of 42 msec and 250 msec, respectively, from which the data for Fig. 3 were obtained. Each of the transients was taken at a different light intensity. The points are the output of the analyzer and the lines through them are the theoretical fits obtained from the rate equations. The observed fits are excellent, and both the theoretical and experimental transients are very precisely single exponentials. We emphasize that all nine theoretical lines are obtained from only one adjustable parameter, the value of $\beta_0$ chosen to fit one of the curves. The relaxation times are not parameters, since they are determined from the plot in Fig. 3. The theoretical transients were obtained on the IBM 7090 computer by solving the matrix $B_{ij}$ for the eigenvalues and finding the coefficient that multiplies the corresponding exponential term in the solution under the assumption of equal populations of the sublevels at $t = 0$.

C. Determination of $\beta_0$ and $\alpha$ for Cells with Buffer Gas

The value of $\beta_0$ remains approximately constant so long as $(\Delta \nu)_{\text{emission}} \gg (\Delta \nu)_{\text{absorption}}$, so that the value of $\beta_0$ is the same as that with no buffer gas. With buffer gas the width of the emission line is about eight times that of the Doppler-broadened absorption line, but the ratio $(\Delta \nu)_{\text{emission}}/(\Delta \nu)_{\text{absorption}}$ becomes rapidly less favorable as buffer gas is added to the absorption cell; thus $\beta_0$ must be corrected for pressure broadening. For the higher pressures ($> 3$ cm)
the Lorentz broadening is already appreciably larger than the Doppler width so that we assume a pure Lorentzian absorption line. The half width of the Rb resonance line \((P_{1/2})\) has been measured by Shang-Yi Ch' en, who finds that for each cm of He (25° C) the absorption-line half width increases by 0.0072 cm\(^{-1}\), whereas for each cm of Ar (25° C) the increase is 0.0075 cm\(^{-1}\). For the emission line we also assume a Lorentzian with a width of 0.09 cm\(^{-1}\). This is the width measured by Jarrett for a Varian lamp similar to ours. We have then for \(\beta_0\)

\[
\beta_0 \propto \int_{-\infty}^{\infty} \frac{1}{(v - v_0)^2 + \left(\frac{\Delta v_{\text{emission}}}{2}\right)^2} \cdot \frac{1}{(v - v_0)^2 + \left(\frac{\Delta v_{\text{L}}}{2}\right)^2} \, dv. \quad (5)
\]

The measured amplitude ratio \(A_2/A_1\) and the corrected values of \(\rho = 1/\beta_0 T\) are now used in conjunction with data such as that shown in Fig. 7 to determine the values of \(\alpha\) corresponding to a given pressure. In Fig. 8 are plotted the theoretical amplitude ratios as a function of \(\rho\) and \(\alpha\); these data were obtained by solving the signal equations for different values of \(\alpha\) and \(\rho\), averaging the exact solutions over the sampling interval, and then fitting the averaged signals to two exponentials by the method of least squares. The theoretical signals were calculated in units of \(1/\beta_0\); therefore, in order to average the signals over the actual sampling intervals, it was necessary to know the value of \(\beta_0\). For example, if \(\beta_0\) was measured as 0.5 msec\(^{-1}\), then each unit of \(1/\beta_0\) equals 2 msec. Thus if the multichannel analyzer was sampling the transient in 1-msec intervals, the theoretical curves were averaged over one-half unit (of \(1/\beta_0\)). In practice, this would mean that the signal was evaluated every one-tenth unit (0.2 msec) and that each group of five points was averaged in succession before the signals...
were fitted to two exponentials. By so averaging the theoretical signals, we duplicated the condition under which the transients were sampled.

V. RESULTS

A. Shape of the Pumping Transients

Figure 6 shows that both theoretically and experimentally the pumping transients are singly exponential in the no-buffer-gas situation. It is surprising that the theoretical pumping transients in the no-buffer-gas case are singly exponential when it is clear that the general solution is multiply exponential. There seems to be no deeper reason for this than it is a mathematical accident that the general solution can be well represented by a single exponential, at least for the range of values we have investigated.

Figure 7 shows a sequence of curves taken at different buffer-gas pressures in the same cell. The appearance of a second exponential is observed and it is seen that the theoretical and experimental shapes agree well. For high buffer-gas pressures, the values of $\beta_0$ were corrected for pressure-broadening effects as described in Sec. IV. All of the measurements were made in a single cell mounted directly on the vacuum system so that no errors were introduced in the value of $\beta_0$. The appearance of a second exponential was observed in all cells with the addition of buffer gas and is interpreted by us as indicating mixing in the excited state.

B. Excited-State Disorientation Cross Sections

At different buffer-gas pressures, different values of the mixing probability $\alpha$ pertain. One of the features of the doubly exponential transients is that the ratio of the time constants is roughly independent
of the gas pressure. A more sensitive test of the value of \( \alpha \) is the ratio of the amplitudes at zero time. This ratio was used to determine \( \alpha \) and the associated error. We showed previously that

\[
\alpha = 1 - \exp\left( -\frac{n\nu}{T} \frac{1}{2} \sigma_{\text{eff}} \right),
\]

where

\[
\sigma_{\text{eff}} = \sigma_{1/2} + \sigma_{3/2}.
\]

Thus a logarithmic plot of \( \alpha \) vs buffer-gas pressure should yield a straight line whose slope is a measure of \( \sigma_{\text{eff}} \). The plot so obtained for each of the three buffer gases is shown in Fig. 9 and the values deduced for \( \sigma_{\text{eff}} \) are given in Table I. The values measured for \( \sigma_{\text{eff}} \) are decidedly smaller than the values obtained for \( \sigma_{3/2} \) by Jordan and Franken\(^{12}\) in sodium-buffer-gas collisions. Recently Beahn et al. determined the cross section \( \sigma_{3/2} \) for rubidium collisions with buffer gas by measuring the ratios of \( D_2/D_4 \) radiation in the scattered light.\(^{13}\) Their results are consistent with our small values for \( \sigma_{\text{eff}} \) and enable us to deduce values for \( \sigma_{1/2} \) from our cross sections. These are given also in Table I.

One of the assumptions made in this study is that there is equal probability for scattering into any of the hyperfine levels. A reasonable alternative mechanism is that during collision the nuclear and electronic moments are decoupled and the nuclear state is unaffected. Such a mechanism is made plausible by the fact that the interaction energy is about 1/40 eV, which is considerably larger than the hyperfine splitting. Accordingly we have analyzed our data on the assumption that in the complete-mixing case only the electron spin is randomized. Our results indicate that electron-spin randomization should produce only very slight changes in the shape of the transients from the no-buffer gas
Hence, we believe that the mechanism for mixing in the excited $P_{1/2}$ state must involve randomization of the nuclear spin as well.

C. Ground-State Disorientation Cross Sections

By now the disorientation cross sections in the ground state have been measured by many workers. However, all of them employed the Franzen technique for measuring relaxation times, and there is some disagreement among the values. In addition, there are no direct measurements of the xenon and krypton cross sections. It seemed desirable, therefore, for us to remeasure these values with our technique for measuring the relaxation times.

Our method differs from the usual method also in that our cell walls were coated with Paraffin; hence the usual condition of zero polarization at the walls does not apply. However, the relaxation time can still be written as

$$\frac{1}{T_1} = gD_0 \frac{N_0}{N} + \sigma_{G.S.} N \overline{N},$$

where $g$ is a geometrical factor, $D_0$ is the diffusion coefficient, and $N_0$ is the density at atmospheric pressure. Therefore, a plot of $N/T_1$ vs $N^2$ should yield a straight line whose slope is a measure of $\sigma_{G.S.}$. Such a plot for the various buffer gases is shown in Fig. 10. Note that our xenon and krypton measurements were made with undiluted gases. This is made possible by our ability to measure short relaxation times. No attempts were made to determine the diffusion coefficients because of the wall coatings used here.

In Table II we exhibit the results of our cross-section measurements and compare them with those of others. Our argon and neon cross sections agree with those of Franzen, but our xenon and krypton
transients are about five times theirs. However, Franzen's results were based on extrapolation to zero light intensities without any attempt made to separate the exponential components. Hence we believe our results to be more reliable.
FOOTNOTES AND REFERENCES

*Work performed under the auspices of the U. S. Atomic Energy Commission.

9. Transfer of excited Rb$^{87}$ atoms from the $P_{1/2}$ to the $P_{3/2}$ state resulting from collisions with ground-state alkali atoms appears to have very large cross sections, judging by the work of Chapman et al. But even cross sections as large as those reported for potassium ($\approx 10^{-11}$ cm$^2$) contribute negligibly to excited-state relaxation because of the low density of ground-state atoms. See G. D. Chapman, L. Krause, and I. H. Brockman, Can. J. Phys. 42, 535 (1964).
11. The width of a densitometer trace of the lamp profile, obtained by John Conway of the Lawrence Radiation Laboratory, was similar to that obtained by Jarrett.

Table I. Cross sections for mixing the P states of Na and Rb.

<table>
<thead>
<tr>
<th>Buffer gas</th>
<th>Na (×10^{-16} cm^2)</th>
<th>Rb (×10^{-17} cm^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>σ^{3/2}</td>
<td>σ^{3/2, 1/2}^a</td>
</tr>
<tr>
<td>He</td>
<td>23^b</td>
<td>1.0^c</td>
</tr>
<tr>
<td>Ne</td>
<td>23.5^b</td>
<td></td>
</tr>
<tr>
<td>Ar</td>
<td>34^b</td>
<td></td>
</tr>
<tr>
<td></td>
<td>56^c</td>
<td>126^d</td>
</tr>
</tbody>
</table>

a. The cross section σ^{3/2} refers to the transfer of the alkali from the P_{3/2} state to the P_{1/2} state as the result of a buffer-gas collision.


c. See Ref. 13

Table II. Ground-state disorientation cross sections for Rb-noble-gas collisions.

<table>
<thead>
<tr>
<th>Buffer gas</th>
<th>Rb$^{87}$ (25° C)</th>
<th>Rb$^{87}$ (44° C)</th>
<th>Rb (50° C)</th>
<th>Rb (67° C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>1.0×10$^{-22}$ cm$^2$</td>
<td>1.6×10$^{-22}$ cm$^2$</td>
<td>6.2×10$^{-25}$ cm$^2$</td>
<td>3.3×10$^{-25}$ cm$^2$</td>
</tr>
<tr>
<td>Ne</td>
<td>3.3×10$^{-22}$ cm$^2$</td>
<td>9×10$^{-22}$ cm$^2$</td>
<td>5.2×10$^{-23}$ cm$^2$</td>
<td>3.3×10$^{-24}$ cm$^2$</td>
</tr>
<tr>
<td>Ar</td>
<td>2.0×10$^{-20}$ cm$^2$</td>
<td>1.3×10$^{-20}$ cm$^2$</td>
<td>3.7×10$^{-22}$ cm$^2$</td>
<td>1.1×10$^{-22}$ cm$^2$</td>
</tr>
<tr>
<td>Kr</td>
<td>1.8×10$^{-19}$ cm$^2$</td>
<td>5.9×10$^{-21}$ cm$^2$</td>
<td>4.2×10$^{-22}$ cm$^2$</td>
<td>7.3×10$^{-21}$ cm$^2$</td>
</tr>
</tbody>
</table>

d. See Ref. 6.
FIGURE CAPTIONS

Fig. 1. Block diagram of apparatus.

Fig. 2. Determination of rf level needed to saturate resonance.

Fig. 3. Determination of relaxation time for Rb$^{87}$ in two wall-coated cells with no buffer gas. The relaxation time is the reciprocal of the zero light intensity intercept.

Fig. 4. Determination of relaxation time for Rb$^{87}$ in three wall-coated cells containing buffer gas. The two lines for each gas correspond to the two exponential components of the pumping transient.

Fig. 5. Observed pumping transient in a cell containing no buffer gas and no wall coating.

Fig. 6. Observed pumping transients for each of the points shown in Fig. 3. The points are experimental and the lines are the predictions of the theory. Both are single exponential.

Fig. 7. Observed pumping transients in an Rb$^{87}$ resonance cell as the buffer gas pressure is varied.

Fig. 8. Theoretical amplitude ratios as a function of $\rho$ and $\alpha$.

Fig. 9. Plot of mixing probability $\alpha$ vs buffer gas pressure for the three observed gases.

Fig. 10. Determination of $\sigma_{G.S.}$ from a plot of $N/T_1$ vs $N^2$. 
Fig. 1
Fig. 2
Fig. 3

- $\beta_0 = 0.001 \ I$
- $T_1 = 42 \text{ msec}$

- $\beta_0 = 0.00072 \ I$
- $T_1 = 250 \text{ msec}$
Fig. 4
Fig. 5
Fig. 6

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Fig. 7

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Fig. 8
Fig. 9
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