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COLLISION-INDUCED MIXING IN THE $52P_{3/2}$ STATE OF RUBIDIUM AND THE $62P_{3/2}$ STATE OF CESIUM

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**COLLISION-INDUCED MIXING IN THE $5^2 P_{3/2}$ STATE
OF RUBIDIUM AND THE $6^2 P_{3/2}$ STATE OF CESIUM**

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OF RUBIDIUM AND THE $6^2P_{3/2}$ STATE OF CESIUM

Joseph Yellin

October 4, 1966

COLLISION-INDUCED MIXING IN THE $5^2P_{3/2}$ STATE
OF RUBIDIUM AND THE $6^2P_{3/2}$ STATE OF CESIUM

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ABSTRACT

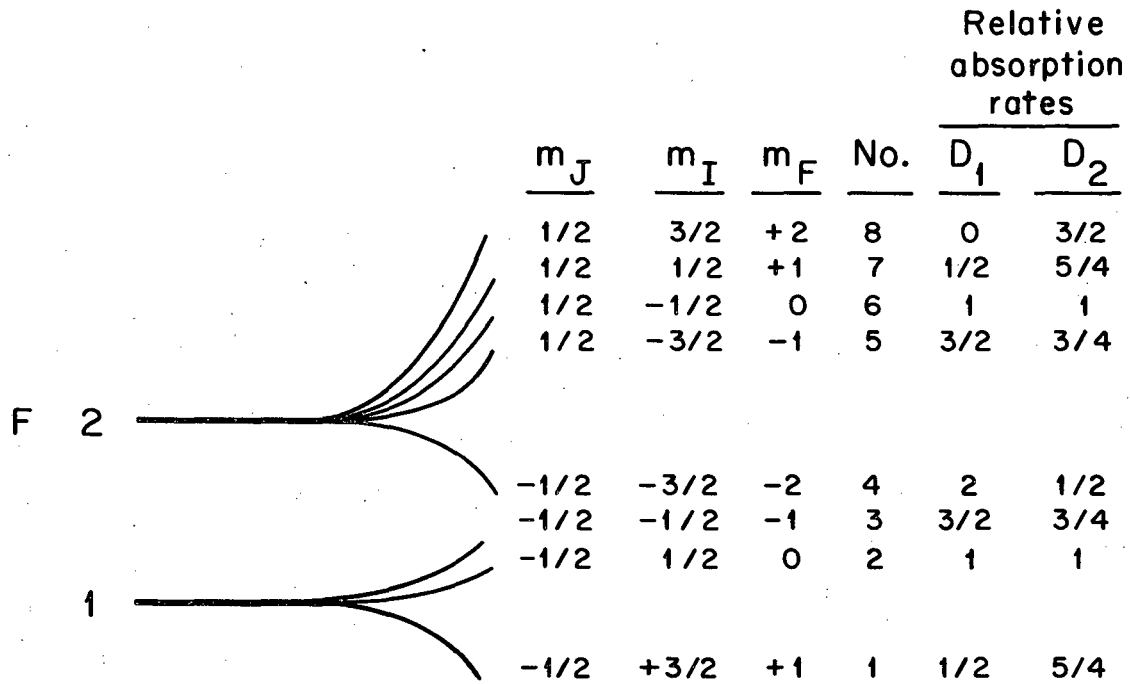
Alkali-metal vapors of rubidium and cesium have been optically pumped, with the D_2 component of the resonance radiation and the reversal of the ground-state polarization observed as a function of buffer-gas pressure. It is well known that when complete mixing occurs in the $^2P_{3/2}$ state, the polarization achieved in the ground state ($^2S_{1/2}$) is opposite to that obtained with the D_1 component of the resonance radiation when circularly polarized light is used. However, in the absence of mixing in the $^2P_{3/2}$ state, the sign of the polarization is the same for both D_1 and D_2 light. For some finite degree of mixing of the $^2P_{3/2}$ state, the equilibrium electronic polarization vanishes. From the null points observed in the D_2 pumping signal vs buffer-gas-pressure curve, we have determined relative cross sections for collisional disorientation of the $^2P_{3/2}$ state of a polarized Rb or Cs atom. For Rb^{85} we find $\sigma_{3/2}(He) = 0.44 \pm 15\%$, $\sigma_{3/2}(Ne) = 0.45 \pm 15\%$, $\sigma_{3/2}(Kr) = 1.26 \pm 15\%$, and $\sigma_{3/2}(Xe) = 1.46 \pm 15\%$; for Rb^{87} we find $\sigma_{3/2}(He) = 0.47 \pm 15\%$, $\sigma_{3/2}(Ne) = 0.46 \pm 15\%$, $\sigma_{3/2}(Kr) = 1.32 \pm 15\%$, and $\sigma_{3/2}(Xe) = 1.46 \pm 15\%$. For Cs, $\sigma_{3/2}(He) = 0.38 \pm 15\%$, $\sigma_{3/2}(Ne) = 0.42 \pm 15\%$, $\sigma_{3/2}(Kr) = 1.22 \pm 15\%$, and $\sigma_{3/2}(Xe) = 1.45 \pm 15\%$. All cross sections are relative to $\sigma_{3/2}(Ar)$.

I. INTRODUCTION

Optical pumping of alkali metal vapors is usually accomplished by using the D_1 ($^2P_{1/2} \rightarrow ^2S_{1/2}$) component of the resonance doublet, or, if interference filters are not available, by a mixture of D_1 and D_2 ($^2P_{3/2} \rightarrow ^2P_{1/2}$) light.¹ It has been shown by Franzen and Emslie² and by Dehmelt that large polarizations and signals are obtained when the D_1 component alone is used. Large polarizations are also obtained when the D_2 component alone is used, as was shown (for example) by Hawkins;³ however, in most experimental situations the D_2 signals are considerably smaller than the D_1 signals, due largely to the severe effect that collisional mixing of the $^2P_{3/2}$ state has on the signal. As most optical pumping experiments employ buffer gases that may disorient the excited state, it is clearly advantageous to use separated D_1 light. Thus there has been virtually no experimental investigation of the pumping process employing D_2 light. It happens though that the D_2 pumping signal is affected so dramatically by disorientation of the $^2P_{3/2}$ state that it becomes easy to observe mixing in the $^2P_{3/2}$ state resulting from collisions between the alkali atom and foreign-gas atom.

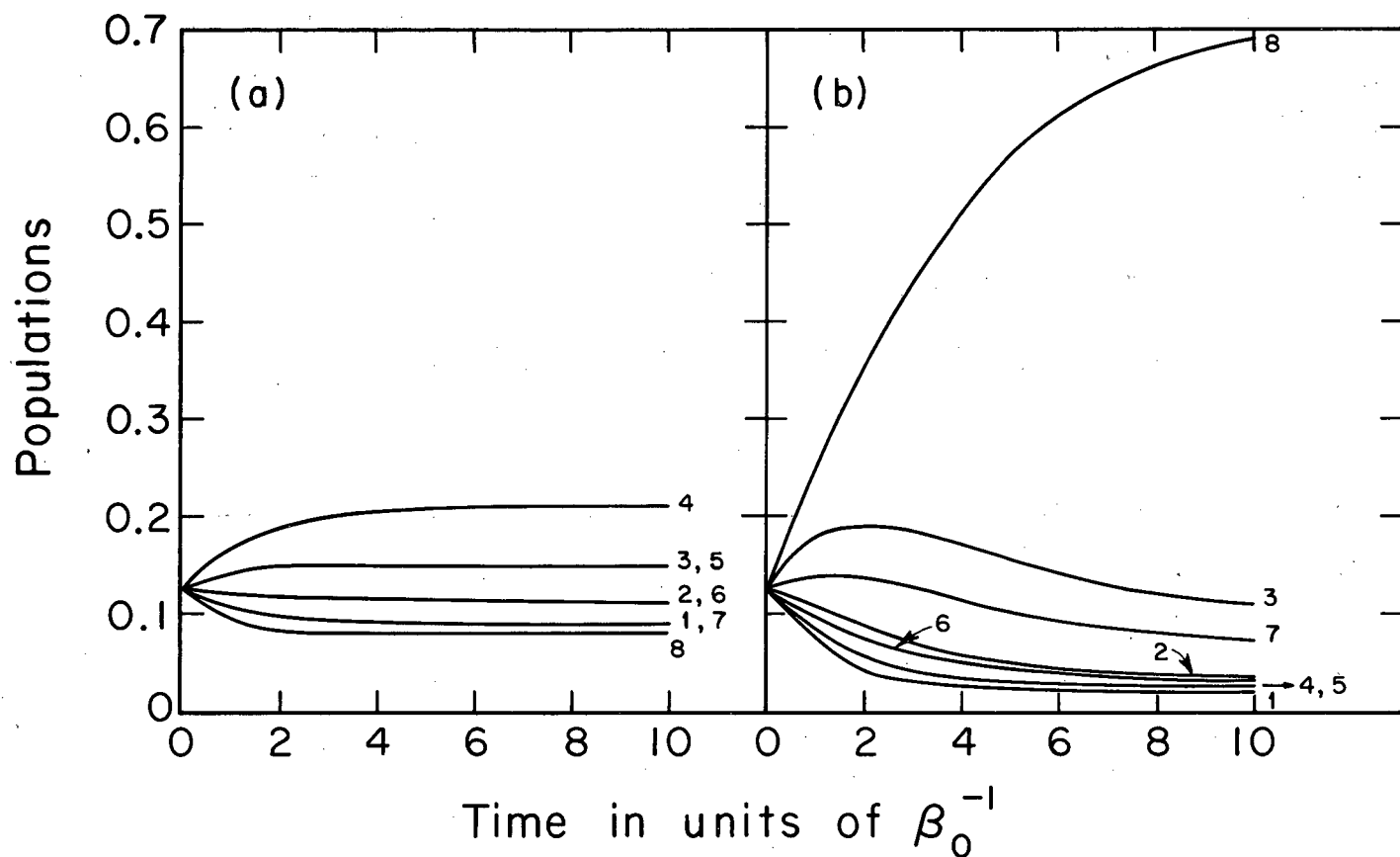
To be explicit, we restrict ourselves to an alkali with nuclear spin $I = 3/2$, and neglect for the present the ground-state relaxation. As with the case of D_1 pumping with right circularly polarized light, the alkali atoms are transferred into the level with $m_F = 2$. This level has an absorption probability which for D_2 light is $3/2$ as great as the average absorption probability for all levels, but the selection rule for absorption of a right circularly polarized photon ($\Delta m_F = +1$) and the selection rules for emission ($\Delta m_F = 0, \pm 1$) ensure that the atoms are trapped in the level $|F=2, m_F=2\rangle$. An atom once having arrived in this level can undergo transitions only to and from the level $|F=3, m_F=3\rangle$ of the $^2P_{3/2}$ state. Thus 100% polarization is achieved; however, if we allow the atoms to scatter out of the latter state into other sublevels of the excited state, emission will occur to levels other than $|2, 2\rangle$ in the ground state and the ground-state polarization will diminish. No amount of recycling will restore the original polarization. In fact, since the $|2, 2\rangle$ level is the most absorbing level and will therefore deplete most rapidly, whereas the $|2, -2\rangle$ level is the least absorbing, more atoms will be present in the latter and one might expect the electronic polarization to reverse in sign as we see from Fig. 1.

An actual population distribution is shown in Fig. 2. In contrast, when D_1 light is used, relaxation of the $^2P_{1/2}$ state does not change the steady-state polarization but changes only the time it takes to achieve steady state. More pumping cycles are then required to transfer the atoms into the $|2, 2\rangle$ level but once they are there, absorption ceases and the steady-state polarization is unaltered. Figure 3(a) shows the result of D_2 pumping on an alkali with nuclear spin $I = 3/2$ in the two extreme limits of (i) no mixing in the excited state and (ii) complete mixing, with uniform relaxation assumed in the ground state. This is contrasted with similar computations for D_1 pumping in Fig. 3(b). At the pressures at which nulls are observed, the ground-state relaxation is due to wall collisions and should be described well by uniform relaxation. The signal is proportional to the electronic polarization.



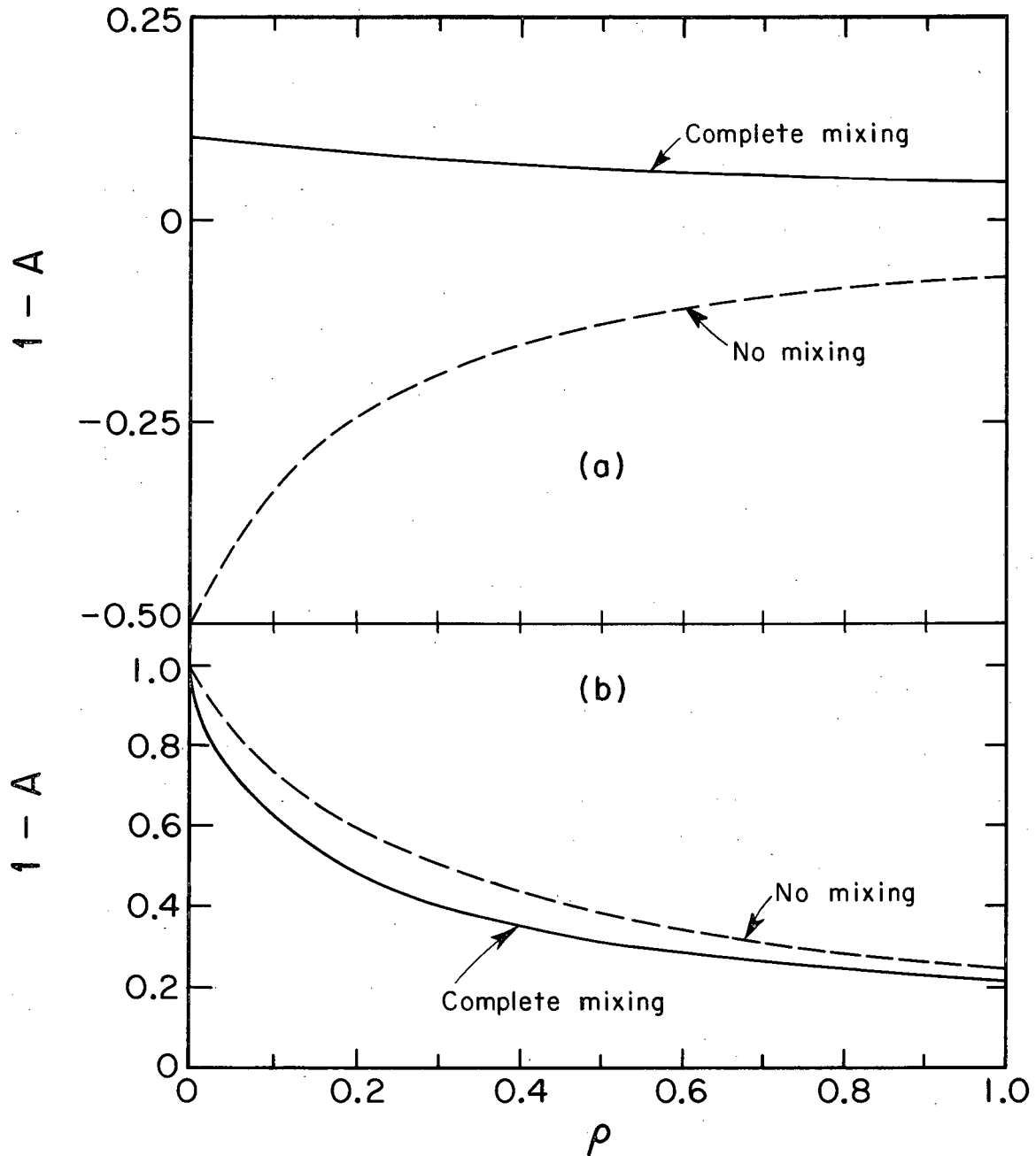
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Fig. 1. Level diagram for the $^2S_{1/2}$ ground state of an alkali with nuclear spin $I = 3/2$.



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Fig. 2. Population distribution of an alkali with nuclear spin $I = 3/2$, optically pumped with right circularly polarized D_2 light. (a) Complete mixing in the $2P_{3/2}$ state; (b) no mixing in the $2P_{3/2}$ state. A relaxation parameter $\rho = 0.1$ is assumed and corresponds, for example, to a pumping time of 1 msec and a ground-state relaxation time of 10 msec. See Ref. 2.



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Fig. 3. Optical pumping signal for an alkali with nuclear spin $I = 3/2$. (a) D_2 light; (b) D_1 light. The signal is $1 - A$ where A is the equilibrium absorption.

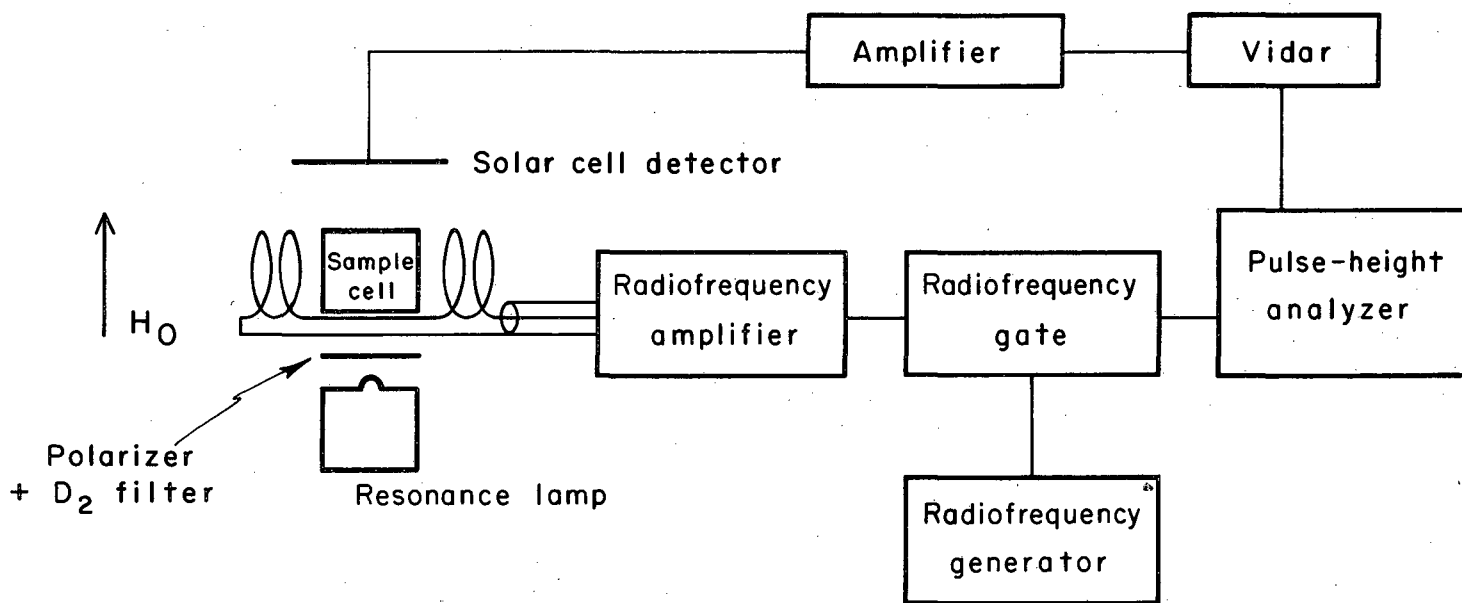
The reversal of the electronic polarization forms the basis of the experiment described below.

II. EXPERIMENTAL METHOD

The basic apparatus is the standard one used in optical pumping experiments in which the pumping radiation is circularly polarized D_2 light and the transmitted light is monitored. A diagram of the apparatus along with the associated electronics is shown in Fig. 4. We used a Varian X49-609 resonance lamp⁴ with the bulbs made of isotopically enriched Rb^{87} (99.16%), Rb^{85} (99.54%), and Cs.⁵ The D_2 signals were readily observable on a scope in a wall-coated cell or a cell with several mm of Hg buffer gas; but a pulse-height analyzer (PHA) had to be employed to observe signals near the null point where the polarization is very small. The PHA was operated in time mode as a phase-sensitive amplifier with typical integration times of 3 minutes. Operation of the PHA as a phase-sensitive amplifier has previously been described.⁶ A solar-cell detector monitored the transmitted light.

Cylindrical absorption cells 4.5 cm in diameter and 7 cm long constructed of Pyrex with optical windows were used throughout. The cells were connected to a gas delivery and vacuum system, and the buffer-gas pressure was measured with an oil manometer. Some of the cells were wall coated with Paraflint to permit observation of the D_2 signal at zero buffer-gas pressure; however, it was discovered by accident that if the liquid nitrogen trap were allowed to go dry for about 48 hours, a signal could be observed on a scope in the evacuated cell. This signal was attributed to Octoil creeping from the diffusion pump into the cell. Thereafter when it became desirable to observe signals at zero buffer-gas pressure, the liquid nitrogen was blown out of the trap. No attempt was made to measure the relaxation times in Octoil-coated cells. The cells were baked with a torch and a pressure $< 10^{-7}$ mm of Hg obtained before the alkali distilled into the cells. The Rb and Cs cells were maintained at room temperature ($22^\circ C$). Rubidium in natural abundance was used for the Rb experiments. At $22^\circ C$ the density of Rb in the cell as determined from total absorption was $\approx 10^9/cm^3$ so that complications due to Rb^{87} - Rb^{85} spin exchange were negligible. The spin-exchange cross section of Rb^{87} - Rb^{85} as measured by Jarrett⁷ and by Moos and Sands⁸ is $\approx 2 \times 10^{-14} cm^2$, so that the spin-exchange time $\tau_{ex} \approx 1$ sec is much longer than all other relaxation times. Because there is a partial overlap between the Rb^{87} and Rb^{85} emission lines,⁹ both isotopes were pumped by the enriched lamps; however, as the nuclear spin of the two isotopes is different, the rf field H_1 used to destroy the polarization acts on only one isotope and the long spin-exchange time ensures there is no appreciable coupling of the isotopes.

The D_2 pumping signals were generated in rapid succession ($\approx 10/sec$) at a particular value of buffer-gas pressure and for a fixed length of time, then stored in the PHA. A linear gate chopped the output of an rf generator tuned to the Larmor precession frequency of the particular alkali investigated; the output of the gate was amplified and fed into the rf loop about the absorption



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Fig. 4. Block diagram of apparatus

cell. The gate was in the nonconducting mode for 40 msec, and in the conducting mode for 60 msec. When the gate conducts, the polarization of the alkali vapor is destroyed, provided the splitting of the ground-state Zeeman levels is less than the line width of the rf resonance. In our experiment the line width due to magnetic-field inhomogeneity was ≈ 10 kHz, whereas the splitting in the field $H_0 = 2.7$ G was < 4 kHz. The signal is the difference between the light intensity reaching the detector with the rf off and the light intensity reaching the detector with the rf on. This difference is plotted against buffer-gas pressure.

III. DATA ANALYSIS AND RESULTS

Figures 5 through 7 show D_2 pumping signals vs buffer-gas pressure for each alkali and illustrate clearly the reversal of the alkali polarization. To interpret the data, we neglect for the moment the ground-state relaxation and make the reasonable assumption that when a null is observed in the signal the degree of disorientation in the $^2P_{3/2}$ state of a particular alkali is the same irrespective of the buffer gas used, provided only that the light intensity is fixed. Then if we observe a null for alkali X and two buffer gases A and B at buffer-gas densities n_A and n_B , respectively, we may write for the $^2P_{3/2}$ state relaxation rate

$$\sigma(X - A) n_A \bar{v}(X - A) = \sigma(X - B) n_B \bar{v}(X - B),$$

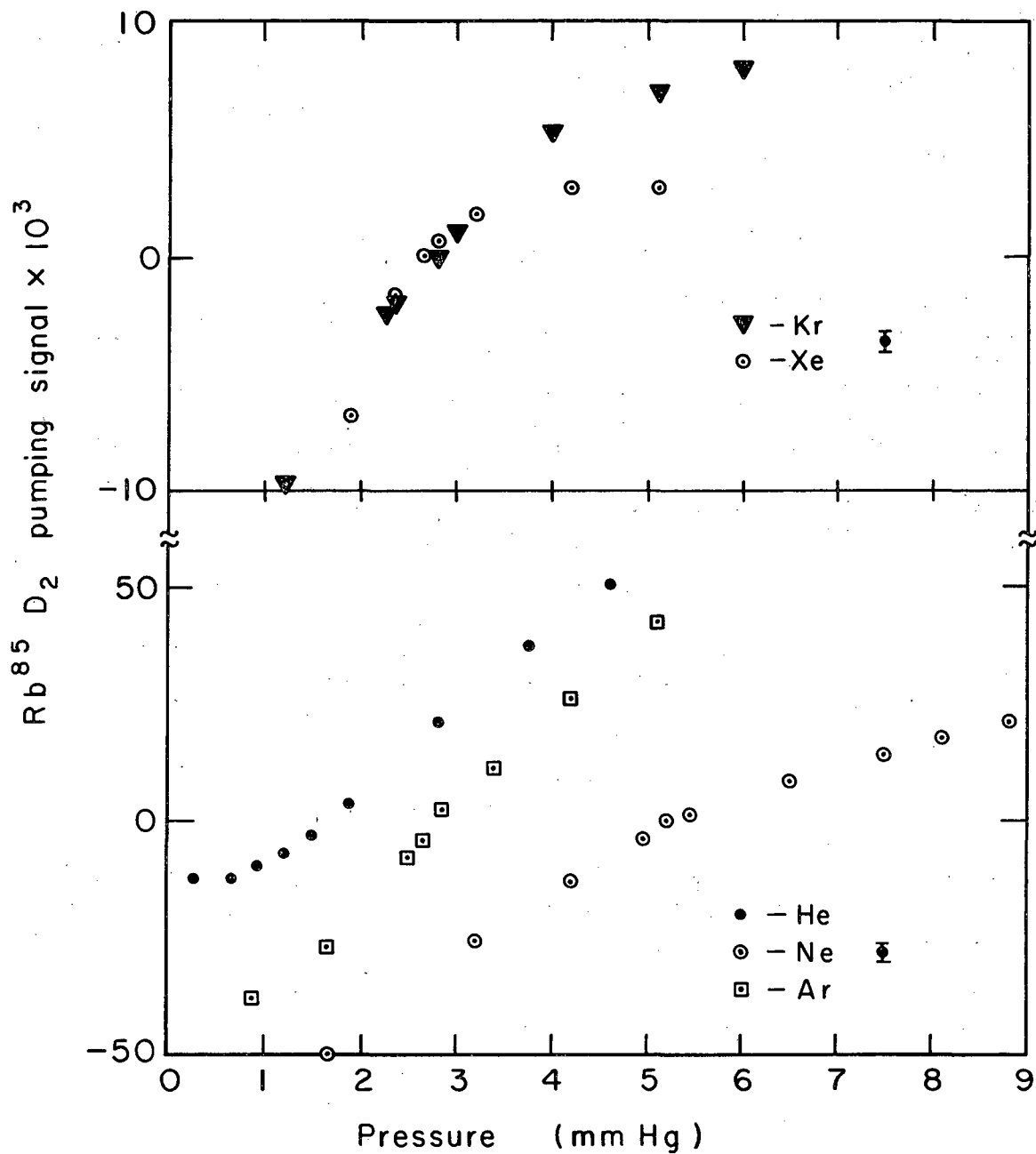
where σ is an average cross section for collisional disorientation, and \bar{v} the mean relative velocity for the colliding pair. Thus

$$\frac{\sigma(X - A)}{\sigma(X - B)} = \frac{n_B \bar{v}(X - B)}{n_A \bar{v}(X - A)}$$

In general, although the ground-state relaxation cannot invert the populations, the position of the null points may depend also on the ground-state relaxation time T_1 , unless $T_1 \gg \tau_{\text{pumping}}$. This condition was satisfied for the buffer gases neon, argon, and krypton and to a lesser extent for helium and xenon. We can estimate T_1 for the former from the diffusion theory of Franzen,¹⁰ from which we have

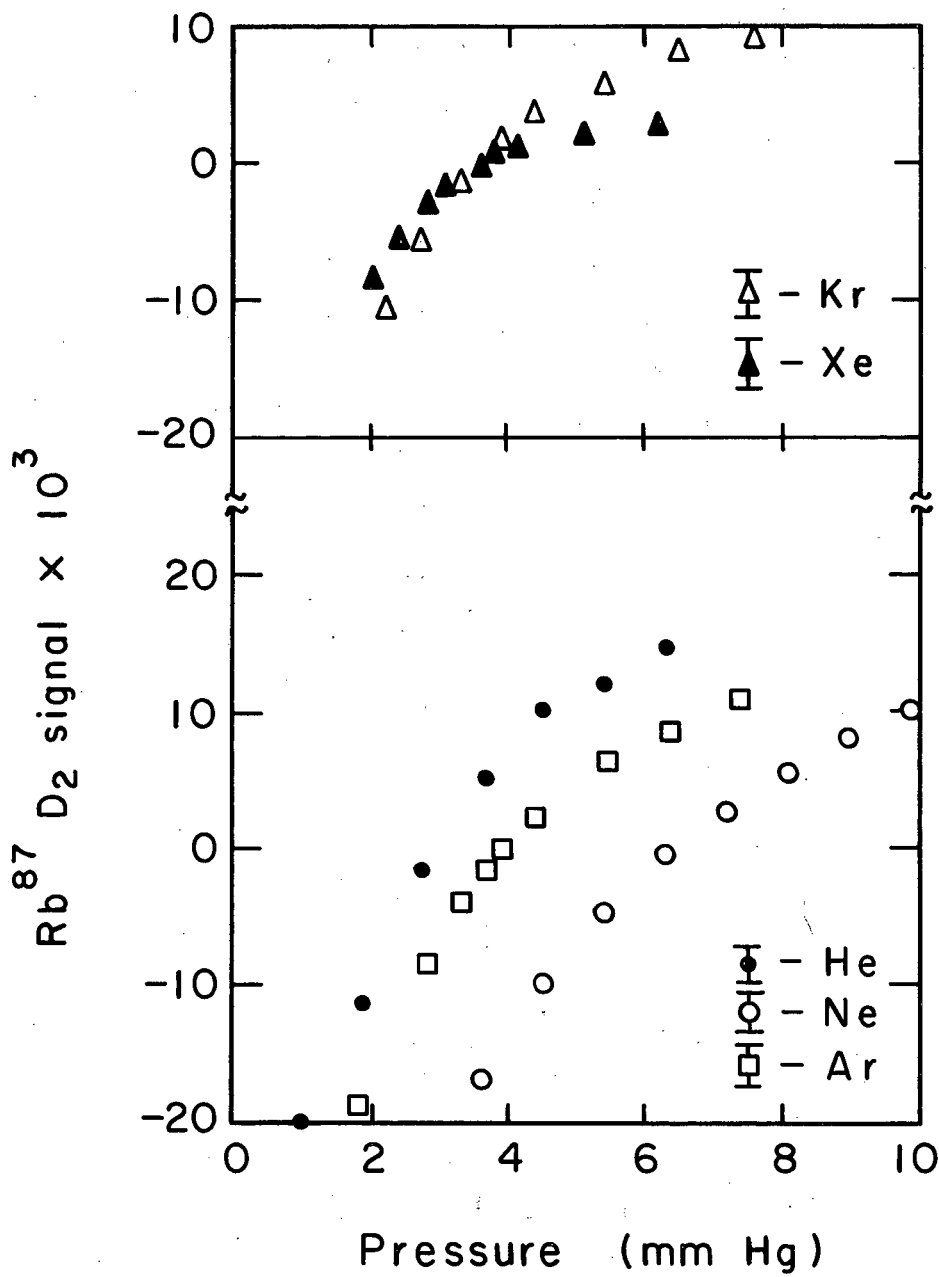
$$\frac{1}{T_1} = g D_0 \frac{p_0}{p} + \sigma_{gs} N_0 \bar{v} \frac{p}{p_0},$$

where g is a geometrical factor which for our cell is 1.34, D_0 is the diffusion coefficient at standard conditions, p_0 is atmospheric pressure, p is the buffer-gas pressure, σ_{gs} is the ground-state disorientation cross section, and N_0 is the density of buffer-gas atoms at atmospheric pressure and temperature appropriate to the experiment. Values of D_0 and σ_{gs} measured for Rb and Cs in a number of experiments¹⁰⁻¹⁵ give $T_1 \approx 10$ to 20 msec in the pressure range where nulls are observed. The pumping time was estimated to be $\tau_{\text{pumping}} \approx 1$ msec from the pumping transients. To play it safe,



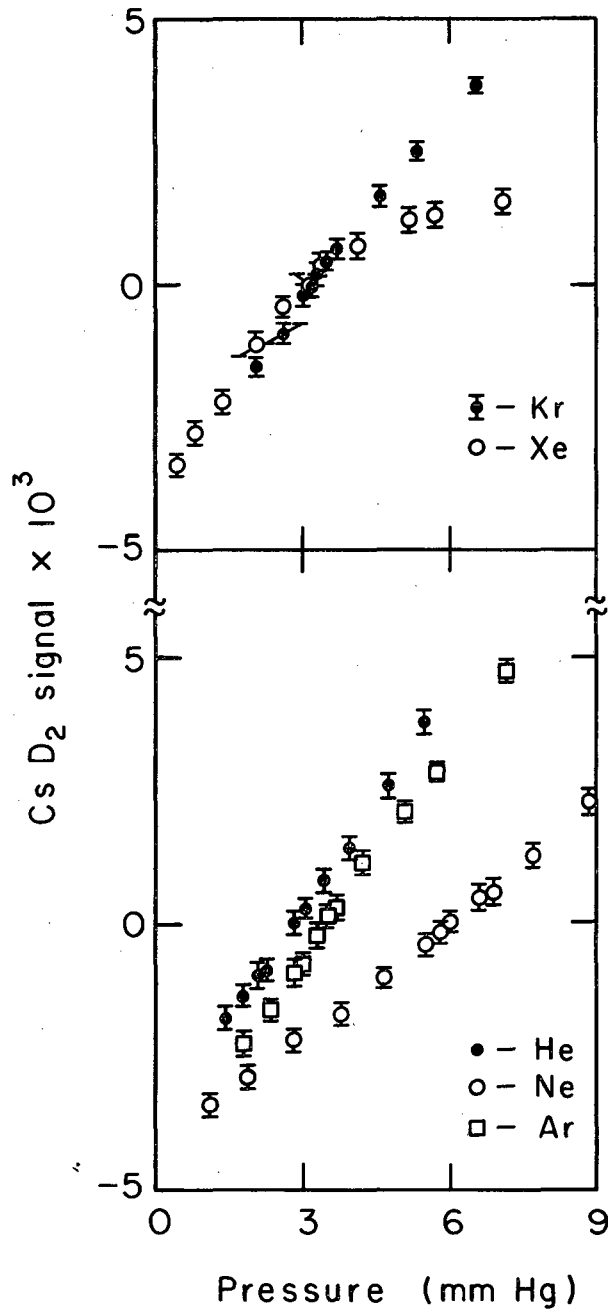
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Fig. 5. D₂ pumping signal vs buffer-gas pressure for Rb⁸⁵.



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Fig. 6. D₂ pumping signal vs buffer-gas pressure for Rb⁸⁷.



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Fig. 7. D₂ pumping signal vs buffer-gas pressure for Cs.

we decreased the light intensity by as much as 300% and repeated the measurements. The null points then occurred at a pressure $\approx 10\%$ lower than with maximum light intensity, but the ratios of pressures at which null points were observed was unchanged within the experimental error. This gives us confidence that the ground-state relaxation was unimportant. The results are summarized in Table I and compared to the intermultiplet mixing cross

Table I. Comparison of relative intramultiplet and intermultiplet mixing cross sections.

Buffer gas	$\sigma_{3/2}$ relative to $\sigma_{3/2}(\text{Ar})$ at 22°C			$\sigma(P_{3/2} \rightarrow P_{1/2})$ relative to $\sigma_{3/2}(\text{Ar})^a$	
	Rb ⁸⁵ ($\pm 15\%$)	Rb ⁸⁷ ($\pm 15\%$)	Cs ¹³³ ($\pm 15\%$)	Rb at 67°C ($\pm 15\%$)	Cs at 38°C ($\pm 15\%$)
He	0.44	0.47	0.38	65.5	0.75
Ne	0.45	0.46	0.42	1.44	0.60
Ar	1.0	1.0	1.0	1.0	1.0
Kr	1.26	1.32	1.22	0.94	3.54
Xe	1.46	1.46	1.45	1.31	5.28

a. Data from Refs. 16 and 17.

sections measured by Krause et al.^{16, 17}

IV. ESTIMATE OF THE ABSOLUTE CROSS SECTIONS

We neglect the nuclear spin to get an estimate of $\sigma_{3/2}$. The level diagram for pumping with D_2 right circularly polarized light with the appropriate transition rates is shown in Fig. 8. The rate equations are readily solved and give for the equilibrium polarization

$$P = \frac{(b_{-+} - b_{+-})\beta_0}{\frac{1}{T_1} + (b_{-+} + b_{+-})\beta_0},$$

where b_{-+} is the transition rate for $|^2S_{1/2}, m_J = -1/2\rangle \rightarrow |^2S_{1/2}, m_J = +1/2\rangle$ and similarly for b_{+-} , and where $\beta_0 = \int I_\nu \sigma_\nu d\nu$, the average light-absorption rate. If we invoke the selection rules $\Delta m_J = 0, \pm 2$ for mixing proposed

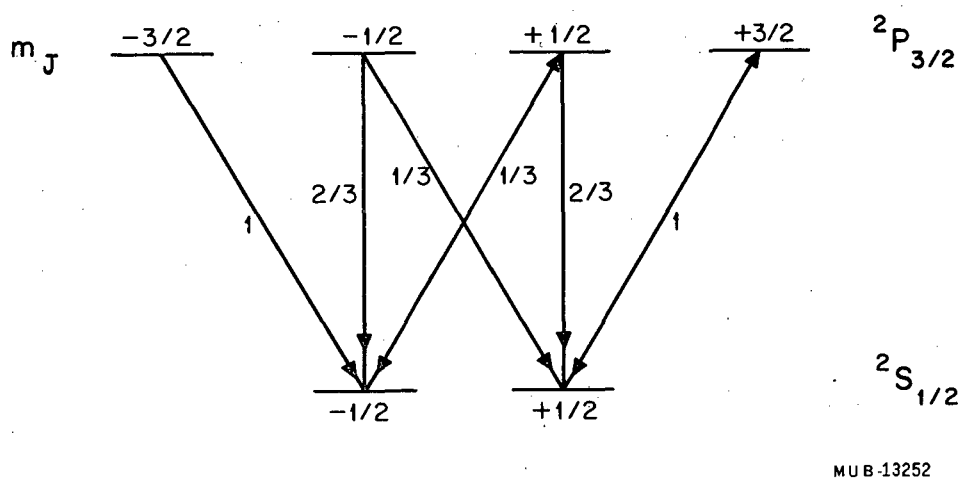


Fig. 8. Level diagram for pumping with right circularly polarized D_2 light, with nuclear spin $I = 0$ assumed.

by Bender¹⁸ on the basis of the Van der Wall interaction, or by Jordan¹⁹ on the basis of the quadrupole induced dipole interaction, it follows that the probability for scattering from $|^2P_{3/2}, m_J=+3/2\rangle$ to $|^2P_{3/2}, m_J=-1/2\rangle$ must be $1/3$ for the polarization to vanish. Thus $\sigma_{3/2} n \bar{v} \tau_{3/2} \approx 1/3$, where $\tau_{3/2}$ is the radiative lifetime of the $P_{3/2}$ state which is $\approx 3 \times 10^{-8}$ sec for Rb and Cs, therefore $\sigma_{3/2} \approx 10^{-15}$ cm² to 10^{-14} cm². In contrast, the intermultiplet mixing cross sections $\sigma(^2P_{3/2} \rightarrow ^2P_{1/2})$ are of the order 10^{-18} cm² for Rb and 10^{-20} cm² for Cs. Intramultiplet mixing cross sections for the $^2P_{1/2}$ state of Rb⁸⁷ recently deduced from a transient experiment are of the order 10^{-17} to 10^{-16} cm² (Ref. 6).

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