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STUDY OF OPTICAL-PIMPING TRANSIENTS IK RUBIDIUM AND CESIUM VAPORS

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STUDY OF OPTICAL-PUMPING TRANSIENTS IN RUBIDIUM AND CESIUM VAPORS

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Joseph Yellin, Richard Marrus, and W. A. Nierenberg

September 4, 1964

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> $\frac{1}{2} \leq \frac{1}{2}$  $\hat{\tau}$  $\sim 10^{11}$

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#### STUDY OF OPTICAL-PUMPING TRANSIENTS IN RUBIDIUM AND CESIUM VAPORS $*$

Joseph Yellin, Richard Marrus, and W. A, Nierenberg

Department of Physics and Lawrence Radiation Laboratory University of California, Berkeley, California /

September 4, 1964

#### ABSTRACT

The shapes of optical-pumping transients in rubidium vapor were studied in detail. Observed transients can be separated into a sum of exponential components. These components are found to be in agreement with a theoretical model based on complete mixing in the excited state. From a study of the times of the individual components as a function of light intensity, relaxation times can be deduced,

#### INTRODUCTION

Although a great deal of work has been done in the application of ~' optical-pumping techniques to the measurement of atomic collision cross sections,  $1-5$  much remains to be done in the understanding of the transient nature of the optical-pumping signal which itself carries information about atomic collision cross sections. The earliest theoretical investigation of the pumping transient structure as applied to an alkali with nuclear spin I =  $3/2$  is that of Franzen and Emslie.<sup>6</sup> We describe below some of the work we have done in extending the above-mentioned work, as well as its utilization in actual experimental situations. This work was undertaken to determine the feasibility of measuring atomic collision cross sections and relaxation times through the influence which these processes have on the structure of the pumping transients.

#### SKETCH OF PUMPING TRANSIENT STRUCTURE

If  $r_i$  is the absorption rate of the ith ground-state level of an alkali and  $\overrightarrow{p}_i$  is the population of that level, then the total absorption rate is  $\beta = \sum_{i} r_i^i p_i$ , the sum extending over all ground-state levels. If the 1

incident radiation is isotropic, we have  $r_i = r_i$  and  $\beta = constant$  as  $\sum_{i=1}^{n} N_i$ , where N is the total density. However, if the incident radiation is polarized so that a definite selection rule is operating, the absorption rates are generally not equal and, in particular, some may vanish  $[e, g, f]$  am=+1 and  $(F_{\text{max}})_{\text{ground state}} = (F_{\text{max}})_{\text{excited state}}$ , then  $r_{F-\text{max}} = 0$ . The more highly absorbing levels then deplete more rapidly than the levels with lower absorption rates, with the result that the atoms pile up in the less absorbing levels. The absorption by the vapor diminishes as a transient while a new equilibrium is being established through the "pumping" action of the radiation and the opposing thermalizing relaxation processes (wall collisions, collisions with foreign gases, spin exchange collisions, etc.).

To find the explicit form of the transient, we must solve for the populations  $p_i$ . These are governed by the phenomenological rate equations

$$
\dot{p}_i = \sum_{j=1}^n b_{ij} p_j + \frac{1}{nT} ,
$$

where T is the relaxation time and the transition rates  $b_{11}$  are given for a few cases in Tables I through IV. Tables V and VI give the relative absorption probabilities. The solution of this system is

$$
p_k = \sum_{i=1}^{R} a_{ki} \exp(-\lambda_i t) - \frac{1}{nT} \sum_{i=1}^{n} b_{ki}^{-1}
$$

where  $a_{ki}$  is the kth component of the ith eigenvector of the matrix b and  $\lambda_i$  is the eigenvalue corresponding to  $a_i$ ; b<sup>-1</sup> is the matrix inverse to b. <sup>1</sup> The absorption probability of the vapor is then a sum of exponentials,

$$
\beta(t) = \sum_{i=1}^{n} c_i \exp(-\lambda_i t) + \text{const.}
$$

						m <sup>1</sup>							
		$F^{\dagger} = 2$						$F' = 3$					
$\mathbf F$	${\bf m}$	$-2$	$-1$	$\mathbf{0}$	$+1$	$+2$	$-3$	$-2$	$-4$	$\pmb{0}$	$\mathbf{1}$	$\overline{c}$	3 <sup>7</sup>
$2^{\circ}$	$\mathord{\text{--}} 2$	$-\frac{76}{243} - \rho$	$\pmb{0}$	$\pmb{0}$	$\mathbf 0$	$\pmb{0}$	$\frac{25}{81}$	25 $2\overline{43}$	$\pmb{0}$	$\pmb{0}$	$\pmb{0}$	$\pmb{0}$	$\pmb{0}$
	$-1$	$\frac{10}{243}$	$-\frac{16}{27} - \rho$	$\pmb{0}$	$\pmb{0}$	$\pmb{0}$	$\frac{20}{81}$	$\frac{50}{243}$	$\frac{4}{27}$	$\pmb{0}$	$\mathbf 0$	$\mathbf 0$	$\pmb{0}$
	$\pmb{0}$	$\frac{4}{81}$	$rac{1}{9}$	$-\frac{22}{27} - \rho$	$\pmb{0}$	$\pmb{0}$	$\mathsf{O}\xspace$	$\frac{20}{81}$	$\frac{10}{27}$	$rac{5}{27}$	$\mathbf 0$	$\pmb{0}$	$\pmb{0}$
	$+1$	$\pmb{0}$	$rac{2}{27}$	$\frac{17}{81}$	$-\frac{212}{243} - \rho$	$\mathbf 0$	$\pmb{0}$	$\pmb{0}$	$\frac{4}{27}$	$rac{2}{9}$	$\frac{52}{243}$	$\pmb{0}$	0
	$+2$	$\pmb{0}$	$\pmb{0}$	$\frac{4}{81}$	$\frac{50}{243}$	$-\frac{20}{27} - \rho$	0 <sup>1</sup>	$\pmb{0}$	$\pmb{0}$	$\frac{4}{81}$	$rac{29}{243}$	$rac{5}{27}$	$\pmb{0}$
$\overline{\mathbf{3}}$	$-3$	$\boldsymbol{0}$	$\pmb{0}$	$\pmb{0}$	$\overline{0}$	$\mathbf 0$	$-\frac{84}{81}$ - $\rho$	$\pmb{0}$	$\pmb{0}$	$\pmb{0}$	$\pmb{0}$	$\pmb{0}$	$\pmb{0}$
	$-2$	$rac{25}{243}$	$\mathbf 0$	$\pmb{0}$	$\pmb{0}$	$\pmb{0}$	$\frac{29}{81}$	$-\frac{280}{243} - \rho$	$\mathbf 0$	$\pmb{0}$	$\mathbf 0$	$\,0\,$	$\mathbf 0$
	$-1$	17 $\overline{243}$	$\frac{4}{27}$	$\pmb{0}$	$\mathbf{O}$	$\boldsymbol{0}$	$\frac{10}{81}$	$\frac{85}{243}$	$-\frac{32}{27} - \rho$ .	$\pmb{0}$	$\mathbf 0$	$\mathbf 0$	0
	$\pmb{0}$	$\frac{4}{81}$	$rac{1}{9}$	$rac{5}{27}$	$\pmb{0}$	$\mathbf 0$	$\pmb{0}$	$\frac{20}{81}$	$rac{2}{9}$	$-\frac{67}{81} - \rho$	$\pmb{0}$	$\mathbf 0$	$\pmb{0}$
	$+1$	$\pmb{0}$	$\frac{4}{27}$	$\frac{10}{81}$	$\frac{52}{243}$	${\bf 0}$	$\pmb{0}$	$\pmb{0}$	$\frac{8}{27}$	$\frac{10}{81}$	$-\frac{116}{243} - \rho$	$\mathbf 0$	$\mathsf{O}\xspace$
	$+2$	$\pmb{0}$	$\pmb{0}$	$rac{20}{81}$	$\frac{50}{243}$	$\frac{5}{27}$	$\pmb{0}$	$\pmb{0}$	$\pmb{0}$	$\frac{20}{81}$	$rac{5}{243}$	$-\frac{8}{27} - \rho$	$\vec{0}$
	$+\,3$	$\pmb{0}$	$\pmb{0}$	$\mathbf 0$	$\frac{20}{81}$	$rac{5}{9}$	$\ddot{\phantom{0}}$ 0	$\mathbf 0$	$\pmb{0}$	$\rm \ddot{o}$	$\frac{10}{81}$	$rac{1}{9}$	$-\rho$

Table I. Matrix of coefficients  $b_{ij}$  for an alkali with nuclear spin I = 5/2 with no reorientation in the P state;<br> $\rho = 1/\beta_0 T$ . Primes refer to initial state. For I = 3/2, see reference 6.

 $-2-$ 

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						$\overline{m}$ <sup>t</sup>							
			$F' = 3$										
$\mathbf F$	m	$-2$	$-1$	$\pmb{0}$	$+2$	$\pm$ 1	$-3$	$-2$	$-1$	$\mathbf 0$	$\mathbf{1}$	$\mathbf{z}$	3
$2^{\circ}$	$-2$	$-\frac{11}{36} - \rho$	$\frac{1}{18}$	$\frac{1}{12}$	$rac{1}{9}$	$\frac{5}{36}$	$\frac{1}{6}$	$rac{5}{36}$	$\frac{1}{9}$	$\frac{1}{12}$	$\frac{1}{18}$	$\frac{1}{36}$	$\pmb{0}$
	$-1$	$\frac{1}{36}$	$-\frac{11}{18} - \rho$	$\frac{1}{12}$	$rac{1}{9}$	$rac{5}{36}$	$\frac{1}{6}$	$rac{5}{36}$	$\frac{1}{9}$	$\frac{1}{12}$	$\frac{1}{18}$	$\frac{1}{36}$	$\pmb{0}$
	$\pmb{0}$	$\frac{1}{36}$	$\frac{1}{18}$	$-\frac{11}{12} - \rho$	$rac{1}{9}$	$\frac{5}{36}$	$\frac{1}{6}$	$\frac{5}{36}$	$rac{1}{9}$	$\frac{1}{12}$	$\frac{1}{18}$	$\frac{1}{36}$	$\pmb{0}$
	$\pm$ 1	$\frac{1}{36}$	$\frac{1}{18}$	$\frac{1}{12}$	$-\frac{11}{9} - \rho$	$\frac{5}{36}$	$\frac{1}{6}$	$\frac{5}{36}$	$\frac{1}{9}$	$\frac{1}{12}$	$\frac{1}{18}$	$\frac{1}{36}$	$\pmb{0}$
	$+2$	$\frac{1}{36}$	$\frac{1}{18}$	$\frac{1}{12}$	$rac{1}{9}$	$-\frac{55}{36} - \rho$	$\frac{1}{6}$	$\frac{5}{36}$	$\pmb{1}$ $\overline{9}$	$\frac{1}{12}$	$\frac{1}{18}$	$\frac{1}{36}$	$\pmb{0}$
$\mathbf{3}$	$-3$	$\frac{1}{36}$	$\frac{1}{18}$	$\frac{1}{12}$	$rac{1}{9}$	$\frac{5}{36}$	$-\frac{11}{6} - \rho$	$\frac{5}{36}$	$rac{1}{9}$	$\frac{1}{12}$	$\frac{1}{18}$	$\frac{1}{36}$	$\mathbf 0$
	$-2$	$\frac{1}{36}$	$\frac{1}{18}$	$\frac{1}{12}$	$rac{1}{9}$	$\frac{5}{36}$	$\frac{1}{6}$	$-\frac{55}{36} - \rho$	$rac{1}{9}$	$\frac{1}{12}$	$\frac{1}{18}$	$\frac{1}{36}$	$\pmb{0}$
	$-1$	$\frac{1}{36}$	$\frac{1}{18}$	$\frac{1}{12}$	$rac{1}{9}$	$\frac{5}{36}$	$\frac{1}{6}$	$\frac{5}{36}$	$-\frac{11}{9}$ $-\rho$	$\frac{1}{12}$	$\frac{1}{18}$	$\frac{1}{36}$	$\pmb{0}$
	$\pmb{0}$	$\frac{1}{36}$	$\frac{1}{18}$	$\frac{1}{12}$	$rac{1}{9}$	$\frac{5}{36}$	$\frac{1}{6}$	$\frac{5}{36}$	$rac{1}{9}$	$-\frac{11}{12} - \rho$	$\frac{1}{18}$	$\frac{1}{36}$	$\pmb{0}$
	$\pmb{1}$	$\frac{1}{36}$	$\frac{1}{18}$	$\frac{1}{12}$	$\frac{1}{9}$	$\frac{5}{36}$	$\frac{1}{6}$	$rac{5}{36}$	$rac{1}{9}$	$\frac{1}{12}$	$-\frac{11}{18} - \rho$	$\frac{1}{36}$	$\pmb{0}$
	$\mathbf{2}$	$\pmb{1}$ $\overline{36}$	$\frac{1}{18}$	$\frac{1}{12}$	$rac{1}{9}$	$\frac{5}{36}$	$\frac{1}{6}$	$\frac{5}{36}$	$\frac{1}{9}$	$\frac{1}{12}$	$\frac{1}{18}$	$-\frac{11}{36} - \rho$	$\mathbf 0$
	$\mathbf{3}$	$\frac{1}{36}$	$\frac{1}{18}$	$\frac{1}{12}$	$\frac{1}{9}$	$\frac{5}{36}$	$\frac{1}{6}$	$\frac{5}{36}$	$rac{1}{9}$	$\frac{1}{12}$	$\frac{1}{18}$	$\frac{1}{36}$	$-\rho$

Table II. Matrix of coefficients  $b_{ij}$  for an alkali with spin I = 5/2. Complete mixing of the P state is assumed.

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 $\frac{1}{2}$  ,  $\frac{1}{2}$  ,  $\frac{1}{2}$ 

Table III. Matrix of coefficients  $b_{ij}$  for an alkali with nuclear spin I = 7/2 and no reorientation in the P state assumed.

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Table IV. Matrix of coefficients  $b_{ij}$  for an alkali with nuclear spin I = 7/2. Complete mixing of the P state is assumed.



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	$Rb$ <sup>85</sup>		$Rb^{87}$
$\lambda_i$	$\mathbf{c}_{\mathbf{i}}$	Λ,	$c_i$
1.9431	0.0207	1.9025	0.0407
1.5451	0.0414	1.2901	0.0853
1.1841	0.0605	0.7299	0.1508
0.8259	0.0868	0.1175	0.6589
0.4649	0.1376	0.0000	0.0643
0.0669	0.6019		
0.0000	0.0511	$\bullet$	

Table V. Decay rates and amplitudes for  $\rm\,Rb}^{87}$  and  $\rm\,Rb}^{85}$  with the  $\rm\,P$  state assumed disoriented and p=O, 01.

Table VI. Relative absorption rates  $r_{\rm m}$  of an alkali with nuclear spin I-5/2 for the transition  $S_{1/2}^{1} \rightarrow P_{1/2}$  with  $\Delta m=+1$ .



The quantities  $b_{ij}$  depend on the degree of reorientation which goes on in the excited state,<sup>1</sup> and can be readily calculated for the two extreme cases of no reorientation in the excited state and complete mixing in the excited state, The calculations shown are for the transition  $nS_1/2 \rightarrow nP_1/2$  (pumping with  $D_{\lambda}$  light), and assume that the emission line is sufficiently broad compared with the absorption line that the intensity distribution may be taken as flat. This assumption is justified in practice. Decay rates,  $\lambda_i$ , and approximate amplitudes for  $Rb^{85}$  and  $Rb^{87}$  are given in Table VII, It should be noted that the decay rates are in units of  $\beta_0$ , the average absorption probability per atom per unit time, The amplitudes are given for a particular value of the relaxation time, which is in units of  $\beta \tilde{0}^1$ .

A great simplification in the transient structure can be effected. It is found that  $\beta$  can be approximated very well by just two exponentials if the excited state is mixed, whereas a single exponential suffices if there is no mixing. Experimentally it is very difficult to resolve all the exponentials, and one would expect to observe this approximate form. This is a very useful result, for it means the appearance of a second decay in the pumping transient testifies to mixing, and furthermore the lifetime and amplitude of the second decay are related to the degree of mixing. The above result also means that, provided there is no mixing, the transient may be taken as exponential and the relaxation time then deduced from  $1/\tau = cI + T^{-1}$ , where  $\tau$  is the lifetime, I the light intensity, and c a proportionality constant, The relaxation time T can then be determined from the intercept of the  $(1/\tau)$  axis, <sup>5</sup> If the excited state is mixed, which may be the case if a foreign gas is present in the absorption cell, one must separate out the two decays before the above technique can be used,

#### EXPERIMENT

Figure 1 shows the experimental apparatus in block form, The transient begins synchronously with the sweep of the multichannel scaler, and is regenerated many times, Each transient is added in phase to the previous one, while noise superimposed on the transients averages to zero because of the random phases of the noise Fourier components, Nonrandom noise, such as 60-cycle pickup, may get phase-locked, but can be minimized by choosing a proper sweep rate, Typically a signal-to-noise ratio of approx 1000:1 or better is obtained with the transient sampled at 1-ms ec intervals, The purpose of the rf is to equalize the populations upon completion of the transient in preparation for a new transient, which comes about when the rf is gated off.

#### **RESULTS**

Pumping transients have been observed in  $Rb^{87}$ ,  $Rb^{85}$ , and  $Cs^{133}$ under various conditions and show the general features predicted, These are shown in Figs. 2 through 5. The  $C_s$  pumping transient shown in Fig. 5 is dominated by the relaxation time as the pumping speed of the lamp is  $\approx$  5 msec;  $\tau$ , then, is essentially the relaxation time and is about three times the mean wall collision time, i.e. it takes three wall collisions to destroy the polarization for the Pyrex cell used,

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 $-3$   $-2$   $-1$  0  $+1$   $+2$   $+3$   $-4$   $-3$   $-2$   $-1$  0  $+1$   $+2$   $+3$   $+4$ 

3 Table VII. Relative absorption rates  $r_m$  for the transition  $S_{1/2}P_{1/2}$ 





Resonance lamp

MUB-4220

Fig. 1. Experimental apparatus.

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# Fig. 2. Optical-pumping transient in  $Rb^{87}$  with Ne buffer gas at 17-cm Hg at 24°C.



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### Fig. 4. Optical-pumping transient in Rb $^{87}$  at 52°C in a wall-coated cell.



 $\overline{C}$ 

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The relaxation time in Fig. 6 is determined from the longest decay, since it can be determined with considerably greater accuracy.

#### DISCUSSION

Only the simplest type of pumping transient has been considered. The effect of spin-exchange collisions has been ignored, and this neglect is probably justified for the experiments performed, as the alkali-alkali spin-exchange time is of no consequence at the operating temperature of  $22^{\circ}$ C, judging by the reported cross sections for spin exchange.<sup>5</sup> Obvious quantities to measure are ground-state relaxation times,. which can be related to the ground-state disorientation cross sections from diffusion theory, and cross sections for disorienting the excited state. Ground.-state disorientation cross sections for alkalis diffusing in noble gases has been measured recently by other techniques, but much remains to be done for the excited-state cross sections.<sup>14</sup> It has been assumed in the derivation of the pumping equations that there is no appreciable absorption of the resonance radiation. In practice, about  $2\%$ of the incident light is absorbed.

#### CONCLUSION

Observed pumping transients show the general features predicted, and it appears likely that cross sections for disorienting the P state of an alkali can be measured by the technique proposed. Relaxation times and therefore ground-state disorientation cross sections can be determined from the transients so long as one is careful to separate out the transient components. Work is currently in progress to determine the excited-state disorientation cross sections as well as ground-state disorientation cross sections.



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