

# Lawrence Berkeley National Laboratory

## Recent Work

### Title

OPTICAL PUMPING BY FORBIDDEN LINES

### Permalink

<https://escholarship.org/uc/item/66c5021q>

### Author

Yellin, Joseph.

### Publication Date

1972

Submitted to Journal of  
Physics B

RECEIVED  
LAWRENCE  
RADIATION LABORATORY

LBL-293  
Preprint 2

DOCUMENTS SECTION

OPTICAL PUMPING BY FORBIDDEN LINES

Joseph Yellin

January 1972

AEC Contract No. W-7405-eng-48

TWO-WEEK LOAN COPY

*This is a Library Circulating Copy  
which may be borrowed for two weeks.  
For a personal retention copy, call  
Tech. Info. Division, Ext. 5545*



LBL-293  
c. 2

34

## **DISCLAIMER**

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

OPTICAL PUMPING BY FORBIDDEN LINES<sup>†</sup>

Joseph Yellin

Lawrence Berkeley Laboratory  
University of California  
Berkeley, California 94720

January 1972

## ABSTRACT

An alkali optical pumping cycle in which the "pump" is accomplished by electric quadrupole transitions to the lowest  $^2D$  states is considered. It is shown that a large electronic spin polarization can be achieved in the s - d - p - s cycle. The magnitude of the cross-section for the absorption of a polarized photon in an s - d transition and the power required to achieve reasonable pumping times is estimated. The effect of collisional disorientation of the  $^2D$  states is explored and it is shown that disorientation of the  $^2D_{5/2}$  state causes the ground state electronic spin polarization to reverse in sign. Hyperfine structure is taken into account and the rate equations are solved for an alkali with nuclear spin  $I = 1/2$ .

## INTRODUCTION

1. Atomic states which are not connected to the ground state by electric dipole transitions may be prepared either directly by electronic or ionic impact excitation or indirectly by optical excitation of an intermediate state. The intermediate state may itself be metastable and require preparation by impact excitation. When electronic or ionic impact excitation is employed it may not be possible to make precision measurement on the atomic properties of the state because of the perturbing effects of the charged particles and their motions in external fields. Only when the state to be studied is sufficiently long lived so that the discharge or source of bombarding particle can be turned off or the atoms extracted from the excitation region before measurements commence is it possible to make precision measurements. However, this is not the usual case since highly excited states that decay to the ground state by forbidden transitions can decay by cascade involving electric dipole transitions. Furthermore, when direct impact excitation is used it is not possible to excite selectively some particular magnetic substate. Likewise, when the excitation is purely optical involving an electric dipole transition to some intermediate state which then decays to the metastable state, selectively is sacrificed and in addition serious noise problems may result from the cascade. The development of tunable lasers (e.g. dye lasers) should make it possible to excite directly metastable states by electric quadrupole transitions. In this way the states will be accessible to investigations by optical pumping and atomic beam techniques. We explore here the possibility and feasibility of optically pumping the ground state of an alkali atom in a beam or bulb experiment by direct

optical excitation of the lowest  $^2D$  states. The  $^2D$  states decay to the ground state through the lowest lying  $^2P$  states by electric dipole transitions. Spontaneous emission of quadrupole radiation is neglected. The pumping cycle is shown in figure 1 where rubidium is used as an example. Aside from general interest in excitation by forbidden optical transitions there is interest in the alkali  $^2D$  states. The inversion of these doublets is not understood and measurement of the hyperfine structure of these states may throw light on the problem. Recently Chang, Gupta and Happer (1971) proposed a method involving cascade transitions for measuring the  $^2D$  state hyperfine structure.

Forbidden lines arising from quadrupole transitions are observed in the laboratory and, on a larger scale, in emission nebulae. In the laboratory Segre (1930) has observed the  $4s - 3d$  transition in potassium in an absorption experiment. Prokofjew (1929) also observed the  $3^2D \rightarrow 4^2S$  transition and determined the strength to be  $A(3^2D \rightarrow 4^2S) = 220/\text{sec}$ . More recently Garstang (1966) calculated the rate as  $130/\text{sec}$  and Hartel and Ross (1968) measured the transition rate from electron impact studies getting  $500/\text{sec} \pm 20\%$ . As there is wide disagreement between these numbers we will estimate the transition rate for rubidium using hydrogenic wavefunctions. An order of magnitude calculation is adequate for our purposes. Similar transitions have been observed in other alkalis. The intensity of these transitions is very weak being characterized by a quadrupole oscillator strength that is several orders of magnitude smaller than that of the  $s - p$  dipole oscillator strength of the resonance lines. We calculate below the  $s - d$  transition rate induced by the absorption of circularly polarized light and then solve for the ground state electronic

spin polarization. In the simplified model used here hyperfine structure is neglected but this does not change the essential features of the problem. The inclusion of the nuclear spin is straight forward and will be discussed at the end.

### CROSS-SECTION FOR QUADRUPOLE TRANSITIONS

2. The probability per unit time of a transition for a one electron atom is given by (see e.g. Schiff 1968)

$$w_{mm'} = \frac{4\pi^2 e^2}{m^2 c \omega h} J(\omega) |\langle n'\ell'sj'm' | (\underline{e} \cdot \underline{p}) e^{i\mathbf{k} \cdot \mathbf{r}} | n\ell s j m \rangle|^2 \quad (1)$$

where  $h\omega$  is the energy associated with the transition,  $J(\omega)$  is the intensity of incident radiation in photons/cm<sup>2</sup>sec per unit frequency interval,  $\underline{e}$  is the polarization vector of the incident radiation,  $\underline{p}$  the momentum of the valence electron and  $\underline{k}$  is the propagation vector.  $e$  and  $m$  are the charge and mass of the electron respectively. When the electric dipole term vanishes the next term of importance in the matrix element is  $i(\underline{e} \cdot \underline{p})(\underline{k} \cdot \underline{r})$ . We assume that we have a circularly polarized wave with positive helicity moving along the axis of quantization ( $z$  axis). Then using spherical vectors we have  $\underline{e} = e_1$ ,  $\underline{k} = k e_0$  where  $e_1 = -\frac{1}{\sqrt{2}}(e_x + ie_y)$  and  $ke = \omega$ . Thus we have to evaluate the matrix element

$$A = \langle n'\ell'sj'm' | -ikp_1 r_0 | n\ell s j m \rangle$$

where  $p_1 = -\frac{1}{\sqrt{2}}(p_x + ip_y)$  and  $r_0 = z$ . From the commutator of the atomic Hamiltonian,  $H$ , with  $\underline{r}$  and of  $\underline{r}$  with  $\underline{p}$  we have  $p_1 r_0 = \frac{im}{2\hbar} [H, r_0 r_1] - 1/2(p_0 r_1 - p_1 r_0)$ . The first term on the right gives rise to the electric quadrupole transition E2 and the second term to the magnetic dipole transition M1. Since M1 vanishes exactly in LS coupling we have to evaluate

$$\text{only } A = \frac{mk}{2\hbar} \langle n'\ell'sj'm' | Hr_0r_1 - r_0r_1H | n\ell'sjm \rangle = \frac{mk\omega}{4\sqrt{\pi}} \langle n'\ell'sjm' | (x) r^2 C_1^2(\theta, \psi) | n\ell'sjm \rangle$$

where we have expressed  $r_0r_1$  in terms of a spherical harmonic of rank 2,

$$C_1^2(\theta, \psi) = \left(\frac{5}{4\pi}\right)^{1/2} Y_{21}(\theta, \psi). \text{ Applying standard techniques of Racah}$$

algebra we get (Edmonds 1957)

$$A^2 = \frac{m^2 k^2 \omega^2}{16\pi} (2\ell'+1)(2\ell+1)(2j'+1)(2j+1) \begin{pmatrix} \ell' & 2\ell \\ 0 & 0 \end{pmatrix}^2 \begin{pmatrix} j' & 2j \\ -m & m \end{pmatrix}^2 \left\{ \begin{matrix} \ell' & j' & s \\ j & \ell & 2 \end{matrix} \right\}^2 (x)$$

$$\left( \int_0^\infty R_{n'\ell'}(r)r^2 R_{n\ell}(r)r^2 dr \right)^2$$

where  $R_{n\ell}(r)$  is the radial wave function. For an s - d transition  $\ell' = 2$ ,

$\ell = 0$ ,  $j = 1/2$  and

$$A^2 = \frac{m^2 k^2 \omega^2}{80\pi} (2j'+1) \begin{pmatrix} j' & 2 & 1/2 \\ -m & 1 & m \end{pmatrix}^2 \left( \int_0^\infty R_{n'2}(r) r^2 R_{n0}(r) r^2 dr \right)^2 \quad (2)$$

with  $j' = 3/2$  or  $5/2$ . Combining (1) and (2) we have

$$w_{mm'} = \frac{e^2 a_0^3 k^3}{40\hbar} (2j'+1) \begin{pmatrix} j' & 2 & 1/2 \\ -m & 1 & m \end{pmatrix}^2 \left( \int_0^\infty R_{n'2}(r) r^2 R_{n0}(r) r^2 dr \right)^2 J(\omega)$$

where  $r$  is now expressed in atomic units (a.u.). We define the pumping

time  $\tau$  by

$$\tau^{-1} = \sum_{mm'} w_{mm'} = \sigma(\omega) I(\omega) \quad (3)$$

where

$$\sigma(\omega) = \frac{e^2 \omega^3 a_0^4}{200 c^3 \hbar \Delta\omega} (2j'+1) \left( \int_0^\infty R_{n'2}(r) r^2 R_{n0}(r) r^2 dr \right)^2$$

in the cross-section for the absorption of light.  $I(\omega) = J(\omega)\Delta\omega$  and  $\Delta\omega$

is the spectral width of the exciting light which is assumed to have a flat

distribution. In conventional optical pumping experiments,  $\tau$  may vary

from 1 msec to 1 sec or more. We can now estimate the photon flux that would



be required to achieve a pumping time of 100 msec. The radial integral is first evaluated using hydrogenic wavefunctions. This should be somewhat more accurate than for electric dipole transitions as the integral of  $r$  is dominated by contribution at large  $r$  where the alkali wavefunction is more hydrogenic. Thus for the  $5s - 4d$  transition we have,

$$R_{50}(r) = \frac{2}{5\sqrt{5}} \left(1 - \frac{4}{5}r + \frac{4}{25}r^2 - \frac{4}{375}r^3 + \frac{2}{9375}r^4\right) e^{-r/5}$$

$$R_{42}(r) = \frac{1}{64\sqrt{5}} r^2 (1 - r/12) e^{-r/4}$$

$$\left(\int_0^\infty R_{50}(r) r^2 R_{42}(r) r^2 dr\right)^2 = 3.2 \times 10^4 \quad (\text{a.u.}) \text{ and}$$

$$\begin{aligned} \sigma(5s^2S_{1/2} - 4d^2D_{3/2}) &\approx \frac{7.7 \times 10^{-10}}{\Delta\omega} \text{ cm}^2, \quad \sigma(5s^2S_{1/2} - 4d^2D_{5/2}) \\ &\approx \frac{11.6 \times 10^{-10} \text{ cm}^2}{\Delta\omega} \end{aligned}$$

where we have used  $k(5s - 4d) = 19355 \text{ cm}^{-1}$ . From (3) we get  $I \sim 10^{17}$  photons/cm<sup>2</sup>-sec corresponding to an energy flux of  $S = h\nu I \approx 33 \text{ mW/cm}^2$  if we take for  $\Delta\omega$  the natural width of the transition  $\sim 10 \text{ mHz}$ . A pumping time of 1 msec will require a flux of  $\approx 3.3 \text{ W/cm}^2$ .

#### OPTICAL PUMPING TRANSITION PROBABILITIES & GROUND STATE ORIENTATION

3. The relative transition probabilities  ${}^2S_{1/2} m \rightarrow {}^2S_{1/2} \bar{m}$  are given by

$$B_{m\bar{m}} = \sum_{j''m''qk} \left| \langle {}^2S_{1/2m} | c_k^1 | {}^2P_{j''m''} \rangle \right|^2 \left| \langle {}^2P_{j''m''} | c_q^1 | {}^2D_{j',m'} \rangle \right|^2 \left| \langle {}^2D_{j',m'} | c_1^2 | {}^2S_{1/2\bar{m}} \rangle \right|^2$$

and if  $B_{m\bar{m}}$  is normalized so that  $\sum_{m\bar{m}} B = 1$  then

$$B_{\overline{m}\overline{m}} = 25(2j'+1) \begin{pmatrix} j' & 1/2 & 2 \\ m' & -m & 1 \end{pmatrix}^2 \sum_{j''q} (2j''+1) \begin{pmatrix} 1 & j'' & j' \\ k & \overline{m} & -m'' \end{pmatrix}^2 \begin{pmatrix} 1 & 1/2 & j'' \\ k & m & -m'' \end{pmatrix}^2 \left\{ \begin{matrix} 2 & j'' & 1/2 \\ j'' & 1 & 1 \end{matrix} \right\}^2 \quad (4)$$

The relative transition probabilities for pumping through the  ${}^2D_{3/2}$  or  ${}^2D_{5/2}$  state are given in Table 1 for the case of no reorientation in the  ${}^2D$  states. The transition rates are given by  $R_{-1/2 \ 1/2} = B_{-1/2 \ 1/2} \sigma(\omega) I(\omega)$  etc.

It is implicit in the above that the spectral width of the exciting light is broad compared to any splitting which may occur in the  ${}^2S_{1/2}$  and  ${}^2D$  states. Since we are here considering the ideal case of an alkali with zero nuclear spin the condition is easily satisfied in low magnetic field.

The rate equation for the density  $n_{\pm 1/2}$  ( ${}^2S_{1/2 \pm 1/2}$ ) of ground state atoms is

$$\frac{dn_{-1/2}}{dt} = -R_{-1/2 \ 1/2} n_{-1/2} + R_{1/2-1/2} n_{1/2}$$

or in terms of the polarization  $P_e = (n_{1/2} - n_{-1/2})/n$ ,  $\frac{dP_e}{dt} = -R_+ P_e - R_-$

where  $R_{\pm} = R_{1/2-1/2} \pm R_{-1/2 \ 1/2}$  and  $n = n_{1/2} + n_{-1/2}$ . Thus

$$P_e(t) = \frac{R_-}{R_+} (e^{-R_+ t} - 1)$$

The maximum polarization that can be achieved  $P_e(\infty)$  is given in Table 2(a). In an atomic beam experiment the atoms are illuminated only for a short time  $t \cong \ell/\overline{v}$  where  $\ell$  is the length of the illumination region and  $\overline{v}$  the average velocity of the atoms. Consequently, the polarization is  $P_e \cong -R_- \frac{\ell}{\overline{v}} = (B_{-1/2 \ 1/2} - B_{1/2-1/2}) \ell/\tau v$ . For typical geometries and beam velocity we can get  $P_e \sim 1\%$  with a  $\tau = 1$  msec. Such a polariza-

tion is easily detectable by atomic beam techniques. When the alkali is optically pumped in a bulb the ground state spin relaxation time  $T_1$  must be taken into account as well as collisional reorientation of the excited state (Kastler, Cohen-Tannoudji, 1966). The latter effects the relative transition rates. The polarization is then given by  $P_e = \frac{R_-}{R_+ + T^{-1}}$  ( $e^{-(R_+ + T^{-1})t} - 1$ ). When a buffer gas is employed to increase the ground state relaxation time it is to be expected that alkali-buffer gas collision will disorient the  $^2D$  states more rapidly than the  $^2P$  states because of the higher angular momentum of the  $^2D$  states. Table 3 gives the relative transition probabilities in the case that the  $^2D$  states are completely disoriented. It is to be noted in Table 2(b) that when the pumping proceeds through the  $^2D_{5/2}$  state the ground state electronic spin polarization reverses in sign as the  $^2D_{5/2}$  state is disoriented. Thus for a certain value of buffer gas pressure the polarization vanishes. A determination of the null-signal buffer gas pressure can then yield information about the  $^2D_{5/2}$  state disorientation cross-section. A similar situation occurs in the normal alkali optical pumping cycle when the pumping proceeds through the  $^2P_{3/2}$  state. Here too the  $^2S_{1/2}$  state polarization reverses in sign as a result of collisional mixing of the  $^2P_{3/2}$  sub-levels (Fricke and Haas, 1966; J. Yellin, 1966).

#### HYPERFINE STRUCTURE

4. The above results are for an alkali with nuclear spin  $I = 0$  and are intended to show that the  $^2S_{1/2}$  ground state can be oriented by quadrupole transition in an  $s - d - p - s$  cycle. Any real alkali possesses an hyperfine splitting which in the ground  $^2S_{1/2}$  state may be much larger

than the spectral width of lasers. The  $^2D$  hyperfine splitting is negligible by comparison. Inclusion of the nuclear spin in the rate equation is straightforward. Instead of the transition coefficient  $B(^2S_{1/2} m_j \rightarrow \bar{m}_j)$  we now have

$$B_{Fm; \bar{F}\bar{m}} = \sum | \langle n^2 S_{1/2} \bar{F} \bar{m} | C_k^1(\theta, \psi) | n^2 P_j, I F m \rangle |^2 \quad (6)$$

$$\times | \langle n^2 P_j, I F m | C_p^1 | n^2 D_j, I F' m' \rangle |^2$$

$$\times | \langle n^2 D_j, I F' m' | C_1^2 | n^2 S_{1/2} I F m \rangle |^2$$

and there result  $4I + 1$  coupled rate equations. The solution of these equations has been discussed by Franzen and Emslie (1957) for the normal optical pumping cycle. If only one hyperfine level is pumped we can obtain an approximate value for  $P_e$  by noting that  $P_e \approx \frac{\Delta n(F)}{n}$  where  $\Delta n(F)$  is the difference in population of the  $F = I \pm 1/2$  states. This can be seen by reference to the Breit Rabi diagram for the  $^2S_{1/2}$  state. All magnetic substates belonging to the  $F = I - 1/2$  hyperfine state connect to  $m_j = -1/2$  whereas all but one magnetic substate belonging to  $F = I + 1/2$  connect to  $m_j = 1/2$  in the Paschen-Back limit. The approximation becomes better as  $I$  increases. To obtain  $\Delta n(F)$ , (6) is summed over all  $m, \bar{m}$  to get the total transition probability  $F = I \pm 1/2 \rightarrow F = I \mp 1/2$ . The two level system is then solved as for the case of no hyperfine structure with the restriction that now only one hyperfine level is excited. It is clear that in the absence of relaxation all of the atoms will be pumped into one hyperfine level leading to a large electronic spin polarization. We calculate below the population and polarization of the ground state of an alkali with nuclear spin  $I = 1/2$ , e.g.  $^{129}\text{Cs}$ . Although this case is not practical for conventional optical

pumping techniques as all alkali with  $I = 1/2$  are radioactive, it is nevertheless of interest in connection with atomic beam\* experiments (Marrus, Wang, Yellin, 1969).

We consider the case that the  $^2S_{1/2}$  hyperfine splitting is small compared with the bandwidth of the exciting radiation. The transition rates in units of  $\tau^{-1}$  are given in Table 4 for the case of no reorientation in the excited state  $^2D_{5/2}$  and in Table 5 for the case that the magnetic substates of  $^2D_{5/2}$  are completely mixed before emission. The relative absorption probabilities  $r_{Fm_F}$  of the four magnetic sublevels of the  $^2S_{1/2}$  state are given in Table 6. The rate equation for the occupation probabilities,  $P_{Fm_F}$ , of the ground state sublevels

$$\frac{dp_{F'm'_F}}{dt} = \sum_{Fm_F} B'_{Fm_F;F'm'_F} P_{Fm_F}$$

where  $B'_{Fm_F;F'm'_F} = B_{Fm_F;F'm'_F}$  for  $Fm_F \neq F'm'_F$  and  $B'_{Fm_F;Fm_F} =$   
 $= - \sum_{F'm'_F} B_{Fm_F;F'm'_F}, \quad Fm_F \neq F'm'_F$

are readily solved for the equilibrium population which are given in Table 7 along with the expectation value of  $F_z$ ,  $\langle F_z \rangle = \sum_{Fm_F} m_F P_{Fm_F}$ , and the absorption probability for the incident radiation,  $a = \sum_{Fm_F} r_{Fm_F} P_{Fm_F}$ .

The calculations assume an infinitely long spin relaxation time for the  $^2S_{1/2}$  state. When no reorientation occurs in the  $^2D_{5/2}$  state the  $^2S_{1/2}$  polarization is negative and the absorption of incident radiation increases by about 9%. Complete mixing of  $^2D_{5/2}$  sublevels causes the  $^2S_{1/2}$  polari-

---

\* In atomic beam experiments it is not necessary to use polarized light but an improved signal/noise is obtained with polarized light.

zation to reverse and the absorption to decrease by about 6% so that the net change in absorption resulting from mixing of the  $^2D_{5/2}$  state sublevels is 15%. The time evolution of the four magnetic substates of  $^2S_{1/2}$  is shown in figures 2 and 3 along with the absorption and polarization.

#### CONCLUSION

5. An orientation of the ground  $^2S_{1/2}$  state of an alkali can be expected in an s - d - p - s optical pumping cycle. The power levels required to achieve reasonable pumping times (10 - 100 msec) are well within the capabilities of conventional dye lasers. Mixing of the magnetic sublevels of the  $^2D$  states either by collisions or radio-frequency resonance can be observed by monitoring the ground state electronic spin polarization by the conventional methods of optical pumping or atomic beams. It is implicit in the above that coherence effects may be neglected. The excitation is assumed continuous or to have a pulse width long compared with the lifetime of the  $^2D$  states.

REFERENCES

- †Work was performed under the auspices of the U. S. Atomic Energy Commission.
- S. Chang, R. Gupta, and W. Hopper, Phys. Rev. Letters 27, 1036-38 (1971).
- A. R. Edmonds, Angular Momentum in Quantum Mechanics (Princeton University Press, Princeton, New Jersey, 1957).
- W. Franzen and A. G. Emslie, Phys. Rev. 108, 1453-58 (1957).
- J. Fricke and J. Haas, Z. Naturforschg 21A, 1319-20 (1966).
- R. H. Garstang, J. Chem. Phys. 44, 1308-9 (1966).
- I. V. Hertel and K. J. Ross, J. Phys. B1, 697-704 (1968).
- A. Kastler and C. Cohen-Tannoudji in Progress in Physics, Vol. V, Edited by E. Wolf (North Holland Publishing Co., Amsterdam, 1966).
- R. Marrus, E. Wand and J. Yellin, Phys. Rev. 177, 122-26 (1969).
- W. Prokofjew, Z. Phys. 57, 387-93 (1929).
- Leonard J. Schiff, Quantum Mechanics, Third Edition (McGraw-Hill Inc. 1968).
- E. Segre, Z. für Physik 66, 827 (1930); E. Segre and C. J. Bakker, Z. für Physik 72, 724 (1931).
- J. Yellin, University of California, Lawrence Radiation Laboratory Report UCRL-17175 (October 1966).

## TABLE CAPTIONS

Table 1: Transition probabilities for the optical pumping cycle

${}^2S_{1/2m_j} \rightarrow {}^2D \rightarrow {}^2P \rightarrow {}^2S_{1/2\bar{m}_j}$  assuming no hyperfine structure and no reorientation in the excited states.

Table 2: Equilibrium electronic polarization of the  ${}^2S_{1/2}$  state for

pumping through the  ${}^2D_{3/2}$  or  ${}^2D_{5/2}$  state. (a) No reorientation in the  ${}^2D$  states. (b) Complete disorientation of the  ${}^2D$  states.

Table 3: Transition probabilities for the optical pumping cycle

${}^2S_{1/2m_j} \rightarrow {}^2D \rightarrow {}^2P \rightarrow {}^2S_{1/2\bar{m}_j}$  assuming no hyperfine structure and complete disorientation of the  ${}^2D$  states.

Table 4: Transition probabilities  $B_{F_m, \bar{F}_m}$  for optical pumping of an

alkali with nuclear spin  $I = 1/2$  by left circularly polarized light propagating along the z-axis. The alkali is excited to the lowest  ${}^2D_{5/2}$  state by electric quadrupole transitions and decays to the ground state by two successive electric dipole emissions involving the lowest  ${}^2P_{3/2}$  state. No reorientation in the  ${}^2D_{5/2}$  and  ${}^2P_{3/2}$  states is assumed.

Table 5: Transition probabilities  $B_{F_m, \bar{F}_m}$  for optical pumping of an

alkali with nuclear spin  $I = 1/2$  by left circularly polarized light propagating along the z-axis. The alkali is excited to the lowest  ${}^2D_{5/2}$  state by electric quadrupole transitions and decays to the ground state by two successive electric dipole emissions involving the lowest  ${}^2P_{3/2}$  state. Complete disorientation of the  ${}^2D_{5/2}$  state is assumed.



Table 6: Relative absorption probabilities of the magnetic sublevels of an alkali with nuclear spin  $I = 1/2$  when the excited state is  $^2D_{5/2}$  and the light is left circularly polarized with the direction of propagation along the z-axis.

Table 7: Equilibrium populations, polarization and absorption probability of the ground state of an alkali with nuclear spin  $I = 1/2$  optically pumped with left circularly polarized light propagating along the z-axis. The excited state is the lowest  $^2D_{5/2}$  and decays to the ground state through the  $^2P_{3/2}$  state. No relaxation in the  $^2S_{1/2}$  state is assumed. (a) No reorientation of the  $^2D_{5/2}$  state. (b) Complete disorientation of the  $^2D_{5/2}$  state.

Table 1

		${}^2D_{3/2}$		${}^2D_{5/2}$	
		$m_j$	$-1/2$	$1/2$	$-1/2$
$\bar{m}_j$	$-1/2$	19/180	32/90	8/15	3/15
	$1/2$	13/90	71/180	2/15	2/15

Table 2

$P_e^{(\infty)}$	${}^2D_{3/2}$	${}^2D_{5/2}$
(a)	- 19/45	- 1/5
(b)	- 1/2	- 1/3

Table 3

		${}^2D_{3/2}$		${}^2D_{5/3}$	
		$m_j$	$-1/2$	$1/2$	$-1/2$
$\bar{m}_j$	$-1/2$	1/8	3/8	1/3	1/6
	$1/2$	1/8	3/8	11/3	1/6

Table 4

		$m_F$			
		0	-1	0	1
$\bar{F}$	$\bar{m}_F$	0	-1	0	1
	0	0	0.15000	0.03333	0.01667
	-1	0.05000	0.198148	0.09444	0.02917
1	0	0.02917	0.101852	0.12176	0.05833
	1	0.02083	0.00000	0.01713	0.02917

Table 5

		$m_F$			
		0	-1	0	1
$\bar{F}$	$\bar{m}_F$	0	-1	0	1
	0	0	0.0625	0.0833	0.0625
	-1	0.0625	0.0833	0.0625	0.0417
1	0	0.0625	0.0833	0.0625	0.0417
	1	0.0625	0.0833	0.0625	0.0417

Table 6

		$m_F$			
		0	-1	0	1
$r_F$	$m_F$	0	-1	0	1
		0	1	4/3	1

Table 7

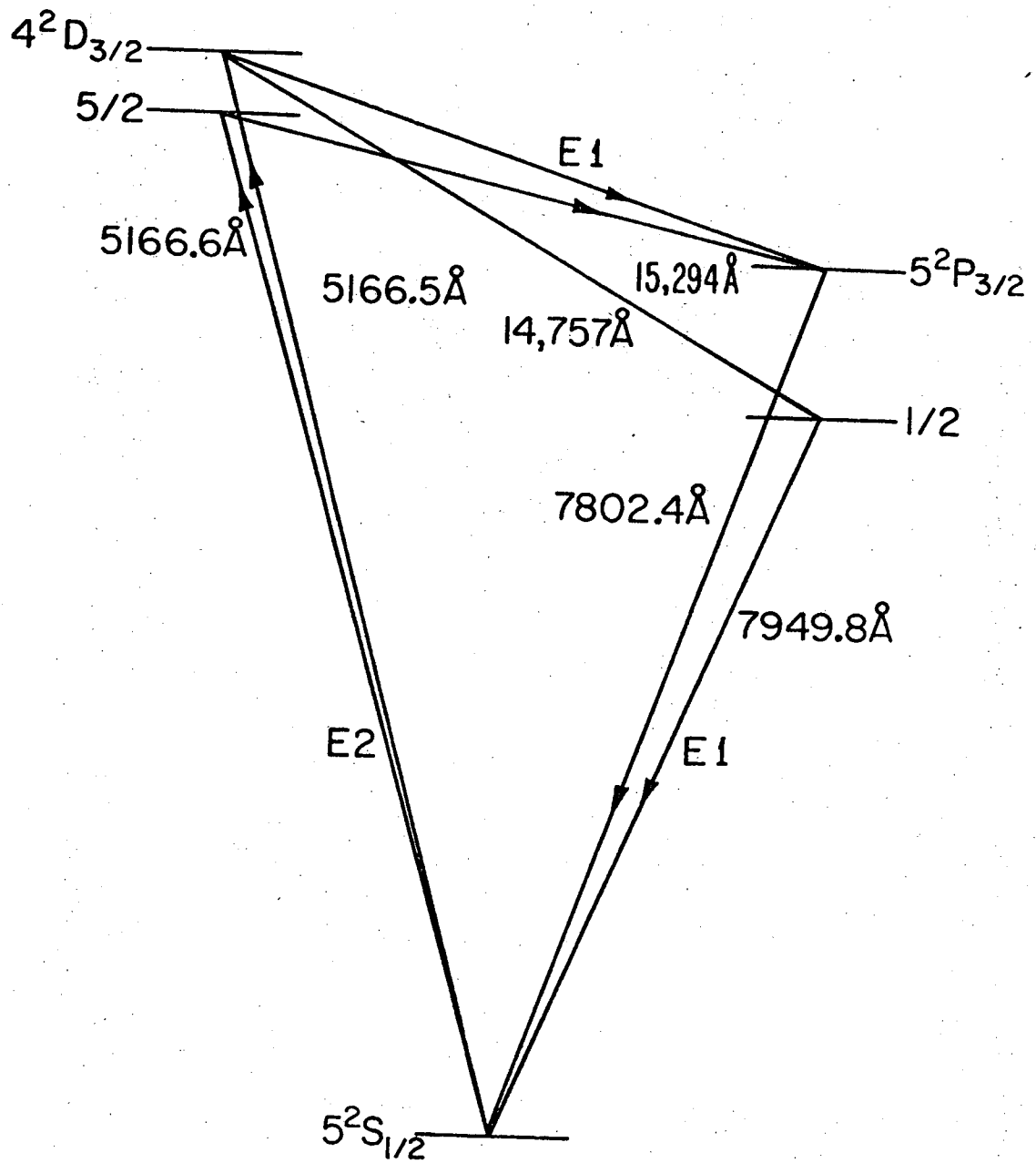
$F_{m_F}$	$P_{m_F}$	
	(a)	(b)
0 0	0.214648	0.235294
1-1	0.347744	0.176470
1 0	0.360211	0.235294
1 1	0.077397	0.352941
$F_z$	-0.270347	0.176470
a	1.090116	0.941176

FIGURE CAPTIONS

Fig. 1. The s-d-p-s optical pumping cycle.

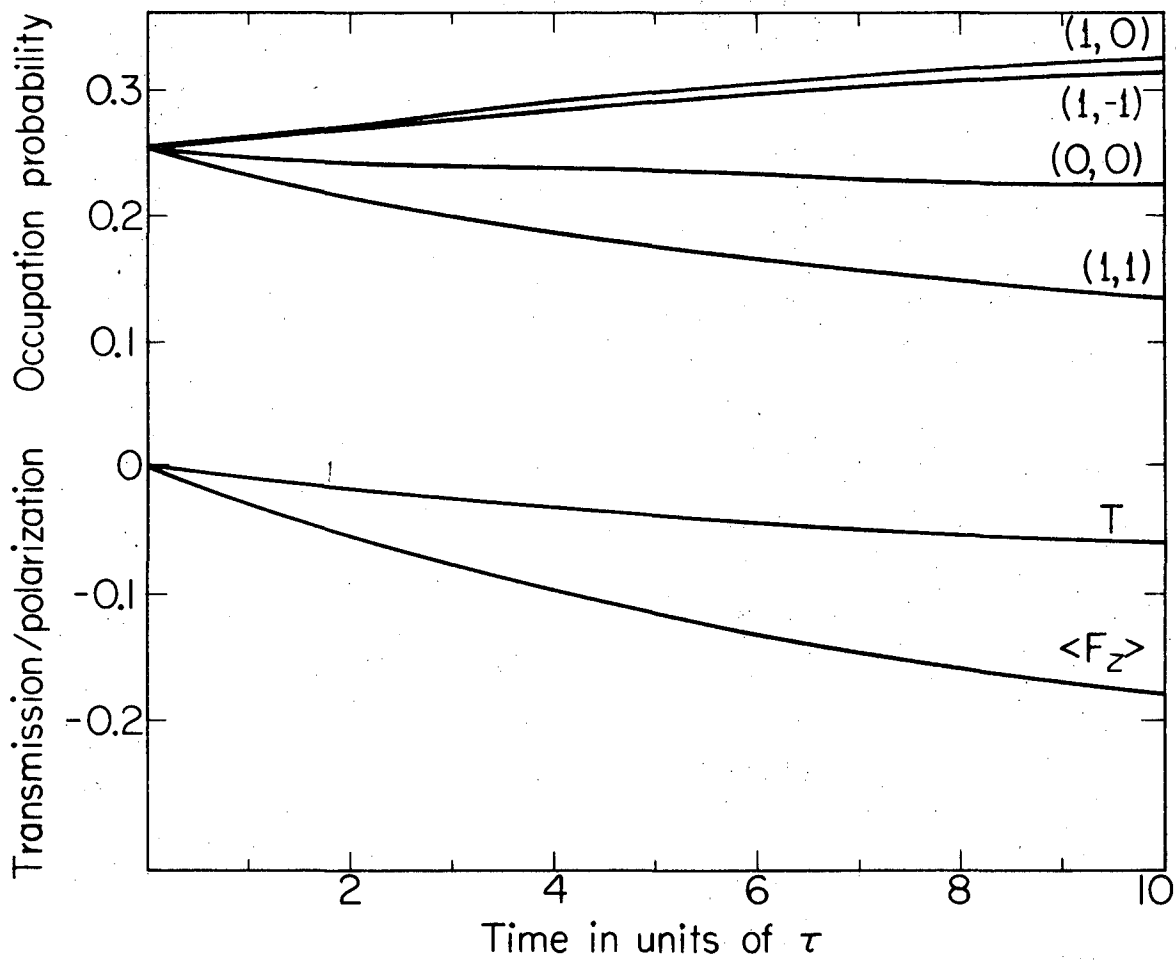
Fig. 2. Time dependence of the four magnetic sublevels of the  $^2S_{1/2}$  state of an alkali with nuclear spin  $I = 1/2$  for an  $s_{1/2} - d_{5/2} - p_{3/2} - s_{1/2}$  optical pumping cycle. Also shown are the polarization of the ground state and the transmission of the pumping light by the alkali vapor. No reorientation is assumed for the excited state.

Fig. 3. Time dependence of the four magnetic sublevels of the  $^2S_{1/2}$  state of an alkali with nuclear spin  $I = 1/2$  for an  $s_{1/2} - d_{5/2} - p_{3/2} - s_{1/2}$  optical pumping cycle. Also shown are the polarization of the ground state and the transmission of the pumping light by the alkali vapor. Complete disorientation of the excited state is assured.



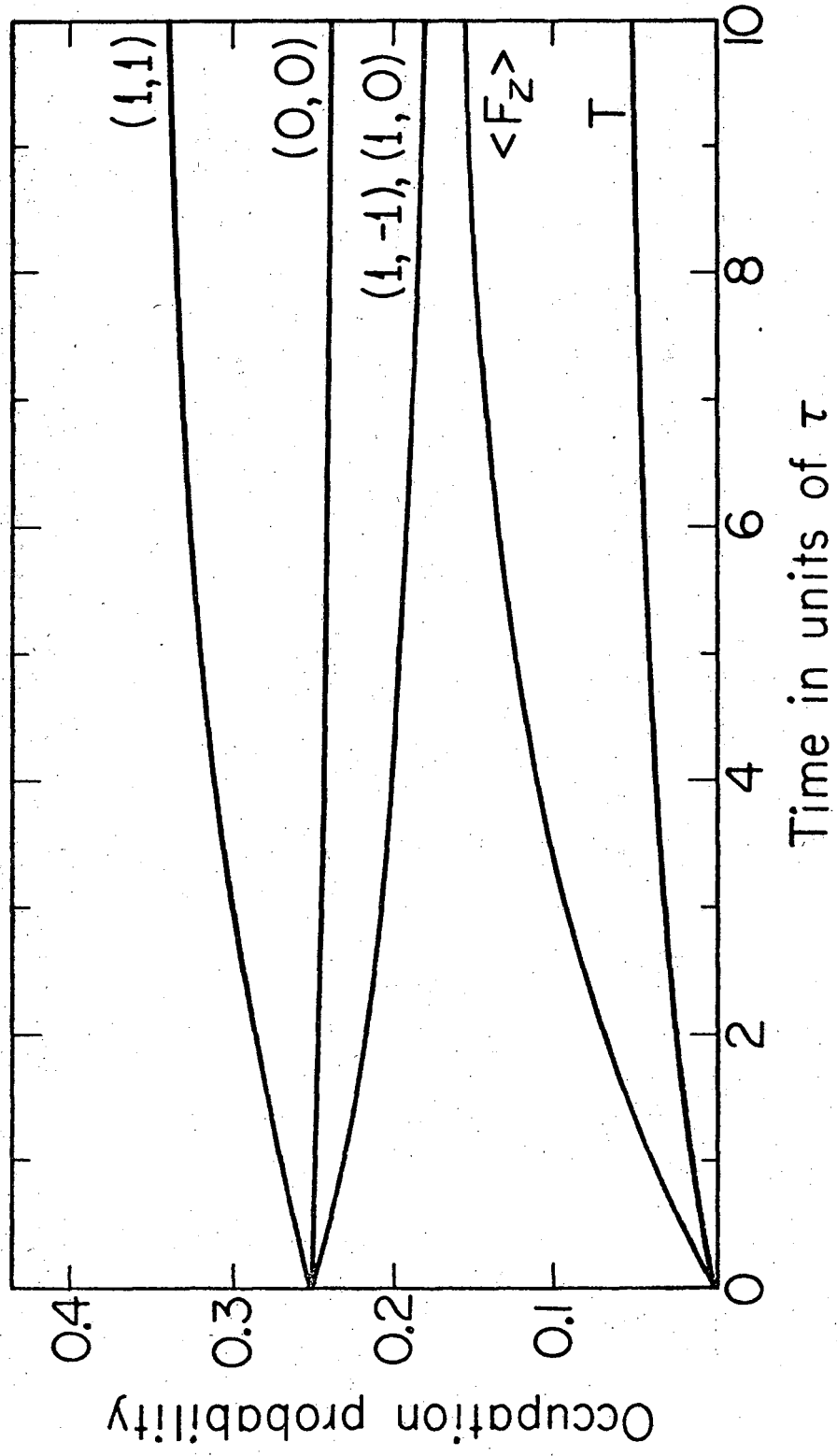
XBL721-2085

Fig. 1



XBL 721-2087

Fig. 2



XBL721-2086

Fig. 3



LEGAL NOTICE

*This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.*

TECHNICAL INFORMATION DIVISION  
LAWRENCE BERKELEY LABORATORY  
UNIVERSITY OF CALIFORNIA  
BERKELEY, CALIFORNIA 94720