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Cesium Transition Probabilities for Optical Pumping*

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Optical transition probabilities are presented between the several magnetic sublevels of the Cs^{133} ground state via the first excited state, given separately for the two fine-structure components. Graphs of the populations of representative states are presented as a function of the time the atoms are illuminated, and a table is given of the populations of atoms absorbing integral numbers of photons from both D lines simultaneously, both polarized and unpolarized.

In order to compare the results of optical pumping[1,2] experiments with theory, it is necessary to have available detailed transition probabilities between the various magnetic sublevels of the ground state. Such probabilities have been published for sodium,[3,4] and the purpose of the present paper is to exhibit those for cesium-133 together with the state of an ensemble of Cs atoms subjected to any of several possible experiments. Although both circularly polarized and unpolarized light have been used for optical pumping,[3,5] it is only necessary to exhibit transition probabilities for the former, since unpolarized light may be regarded as a random mixture of the two senses of circular polarization. Considering only the case of circularly polarized incident light, then, one may briefly describe optical pumping by saying that a group of atoms is made to absorb photons from a directional beam of circularly polarized light and then radiates unpolarized light in all directions. Since the absorbed light carries angular momentum and the radiated light does not, the average angular momentum of the atoms changes; and the atoms, assumed to have zero angular momentum in the beginning, become polarized. The direction of the incident light defines the axis to which the angular momentum quantum numbers, F and m_F, of the atoms are referred; and other asymmetries in the experimental situation, such as magnetic fields, must either be absent or define the same axis. Since it is not worth the effort to entirely eliminate magnetic fields, the usual experiment includes an imposed magnetic field parallel to the desired axis; this has led to the misconception held by some that the magnetic field is necessary to define the axis.

TRANSITION PROBABILITIES

The quantum numbers of the Cs ground state are $6S_{\frac{1}{2}}$, and the first excited state is a doublet with the quantum numbers $6P_{\frac{1}{2}}$ and $6P_{\frac{3}{2}}$. The spin quantum number of the nucleus is $\frac{7}{2}$; and therefore the $J=\frac{1}{2}$ states have total angular momentum quantum numbers F of 3 and 4, while those of the $J=\frac{3}{2}$ state are 2, 3, 4, and 5. Because of the hyperfine interaction, m_I and m_J are not good quantum numbers; and F and m_F must be used. The matrix elements of the electron transition in the m_I, m_J scheme were calculated from standard formulas[6] and transformed into the F, m_F scheme using the matrices given on page 76 of reference 6. From these matrix

TABLE I. Transition probabilities, multiplied by sixteen for convenience in calculation, between ground-state sublevels in cesium excited by circularly polarized cesium $P_{\frac{1}{2}}$ resonance radiation ($\lambda=8943$ A). The index j of the initial state is given down the side and that of the final state, i, across the top.

	F, m_F	4, 4	4, 3	4, 2	4, 1	4, 0	4, −1	4, −2	4, −3	4, −4	3, 3	3, 2	3, 1	3, 0	3, −1	3, −2	3, −3
F, m_F	j \ i	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
4, 4	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
4, 3	2	0.0833	0.0208	0	0	0	0	0	0	0	0.1458	0	0	0	0	0	0
4, 2	3	0.0729	0.0911	0.0651	0	0	0	0	0	0	0.0755	0.1953	0	0	0	0	0
4, 1	4	0	0.1640	0.0938	0.1172	0	0	0	0	0	0.0234	0.1563	0.1953	0	0	0	0
4, 0	5	0	0	0.2344	0.1302	0.1771	0	0	0	0	0	0.0781	0.2031	0.1771	0	0	0
4, −1	6	0	0	0	0.2605	0.2083	0.2605	0	0	0	0	0	0.1563	0.2083	0.1563	0	0
4, −2	7	0	0	0	0	0.2344	0.3046	0.3985	0	0	0	0	0	0.2344	0.1953	0.1328	0
4, −3	8	0	0	0	0	0	0.1640	0.3646	0.6381	0	0	0	0	0	0.2735	0.2187	0.0911
4, −4	9	0	0	0	0	0	0	0.0729	0.3021	1.0417	0	0	0	0	0	0.2187	0.3646
3, 3	10	0.5832	0.1458	0	0	0	0	0	0	0	1.0208	0	0	0	0	0	0
3, 2	11	0.2187	0.2735	0.1953	0	0	0	0	0	0	0.2266	0.5858	0	0	0	0	0
3, 1	12	0	0.2735	0.1562	0.1953	0	0	0	0	0	0.0391	0.2605	0.3256	0	0	0	0
3, 0	13	0	0	0.2344	0.1302	0.1771	0	0	0	0	0	0.0781	0.2031	0.1771	0	0	0
3, −1	14	0	0	0	0.1563	0.1250	0.1563	0	0	0	0	0	0.0938	0.1250	0.0938	0	0
3, −2	15	0	0	0	0	0.0781	0.1016	0.1328	0	0	0	0	0	0.0781	0.0651	0.0443	0
3, −3	16	0	0	0	0	0	0.0234	0.0521	0.0911	0	0	0	0	0	0.0391	0.0312	0.0130

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[1] W. E. Bell and A. L. Bloom, Phys. Rev. **107**, 1559 (1957).

[2] H. G. Dehmelt, Phys. Rev. **109**, 381 (1958).

[3] W. B. Hawkins, Phys. Rev. **98**, 478 (1955).

[4] W. Franzen and A. G. Emslie, Phys. Rev. **108**, 1453 (1957).

[5] W. B. Hawkins, Phys. Rev. **96**, 532 (1954).

[6] E. U. Condon and G. H. Shortley, *Theory of Atomic Spectra* (Cambridge University Press, New York, 1951), p. 63.

TABLE II. Transition probabilities, multiplied by sixteen for convenience in calculation, between ground-state sublevels in cesium excited by circularly polarized cesium $P_{\frac{3}{2}}$ resonance radiation ($\lambda = 8521$ A). The index j of the initial state is given down the side and that of the final state, i, across the top.

	F, m_F	4, 4	4, 3	4, 2	4, 1	4, 0	4, −1	4, −2	4, −3	4, −4	3, 3	3, 2	3, 1	3, 0	3, −1	3, −2	3, −3
F, m_F	j \ i	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
4, 4	1	1.5000	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
4, 3	2	0.3217	0.9803	0	0	0	0	0	0	0	0.0729	0	0	0	0	0	0
4, 2	3	0.0585	0.4127	0.6432	0	0	0	0	0	0	0.0377	0.0977	0	0	0	0	0
4, 1	4	0	0.1316	0.3752	0.4307	0	0	0	0	0	0.0117	0.0781	0.0976	0	0	0	0
4, 0	5	0	0	0.1880	0.2859	0.2968	0	0	0	0	0	0.0391	0.1015	0.0885	0	0	0
4, −1	6	0	0	0	0.2089	0.1967	0.2089	0	0	0	0	0	0.0781	0.1041	0.0781	0	0
4, −2	7	0	0	0	0	0.1880	0.1344	0.1463	0	0	0	0	0	0.1172	0.0976	0.0664	0
4, −3	8	0	0	0	0	0	0.1316	0.1006	0.1011	0	0	0	0	0	0.1367	0.1094	0.0455
4, −4	9	0	0	0	0	0	0	0.0585	0.0720	0.0779	0	0	0	0	0	0.1094	0.1823
3, 3	10	0.2917	0.0729	0	0	0	0	0	0	0	0.2604	0	0	0	0	0	0
3, 2	11	0.1094	0.1367	0.0977	0	0	0	0	0	0	0.2070	0.1992	0	0	0	0	0
3, 1	12	0	0.1367	0.0781	0.0976	0	0	0	0	0	0.1438	0.1940	0.2245	0	0	0	0
3, 0	13	0	0	0.1172	0.0651	0.0885	0	0	0	0	0	0.2877	0.1667	0.2747	0	0	0
3, −1	14	0	0	0	0.0781	0.0625	0.0781	0	0	0	0	0	0.3453	0.2155	0.3453	0	0
3, −2	15	0	0	0	0	0.0391	0.0508	0.0664	0	0	0	0	0	0.2877	0.3163	0.4896	0
3, −3	16	0	0	0	0	0	0.0117	0.0260	0.0455	0	0	0	0	0	0.1440	0.3294	0.8183

elements, the transition probabilities may be determined by use of the following equation[3]:

$$|b_{m0}(\infty)|^2 = \frac{4\pi^2\rho_\omega^2}{\Gamma\hbar^4} \sum_{n,n'} \frac{H_{mn}H_{n0}H_{mn'}^*H_{n'0}^*}{\gamma - i\omega_{nn'}}, \qquad (1)$$

where $|b_{m0}(\infty)|^2$ is the transition probability from state 0 to state m, ρ_ω is the density of radiation oscillators at the resonance frequency, $1/\Gamma$ is the lifetime of the initial state, $1/\gamma$ that of the intermediate state, $\omega_{nn'}$ represents the energy difference between two intermediate states n and n', and H_{mn}, H_{n0}, $H_{mn'}^*$, and $H_{n'0}^*$ are the matrix elements between the initial and intermediate states and between the intermediate and final states. The result is averaged over the frequency distribution of the emitted and absorbed photons for a continuous spectrum incident. In the case of cesium, the interference terms resulting when $n \neq n'$ are unimportant, for the hyperfine splittings are large compared to the natural linewidth of 27 Mc/sec,[7] in both the ground state[8] and the excited states.[9] The smallest hyperfine separation is 152 Mc/sec, between the $F=2$ and the $F=3$ levels of the $6P_{\frac{3}{2}}$ state.

The resulting transition probabilities are tabulated in Table I for transitions occurring via the intermediate levels of the $P_{\frac{1}{2}}$ state ($\lambda = 8943$ A), and in Table II for transitions via the levels of the $P_{\frac{3}{2}}$ state ($\lambda = 8521$ A).

OCCUPATION PROBABILITIES

Experiments may be performed using either of the two fine structure lines for excitation[4,10] or by using an unfiltered light source, which provides both lines. Knowing the transition probabilities, one may calculate the state of an ensemble of atoms as a function of time for a given light source intensity by using the following equations[4]:

$$\dot{p}_i = \beta_0\left(\textstyle\sum_j B_{ij}p_j + \rho\right), \qquad (2)$$

where p_i is the probability of finding an atom in the state numbered i, B_{ij} is the probability of a transition from the state j to the state i, β_0 is the average light absorption probability per atom per unit time, and

$$\rho = 1/(16T\beta_0),$$

where T is the ground-state relaxation time. Except in certain special cases, these equations must be solved by numerical or analog computation; the solutions in Figs. 1 to 4 were obtained by the use of a differential analyzer.

Figure 1 shows the occupation probabilities of the several levels of the ground state as a function of time

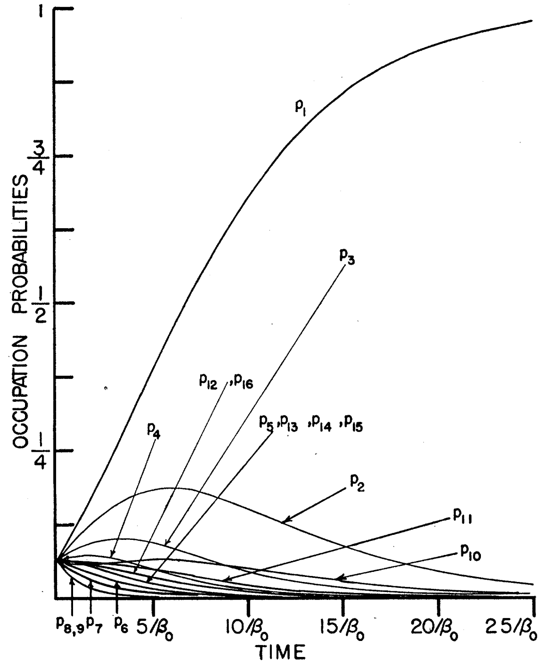

FIG. 1. Occupation probabilities p_i of the ground-state sublevels of cesium atoms illuminated by circularly polarized cesium $P_{\frac{1}{2}}$ resonance radiation ($\lambda = 8943$ A), as a function of time measured in units related to the incident light intensity β_0. The subscripts i have the same meaning as in Table I.

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[9] P. Buck, I. I. Rabi, and B. Senitzky, Phys. Rev. **104**, 553 (1956).

[10] H. G. Dehmelt, Phys. Rev. **105**, 1487 (1957).

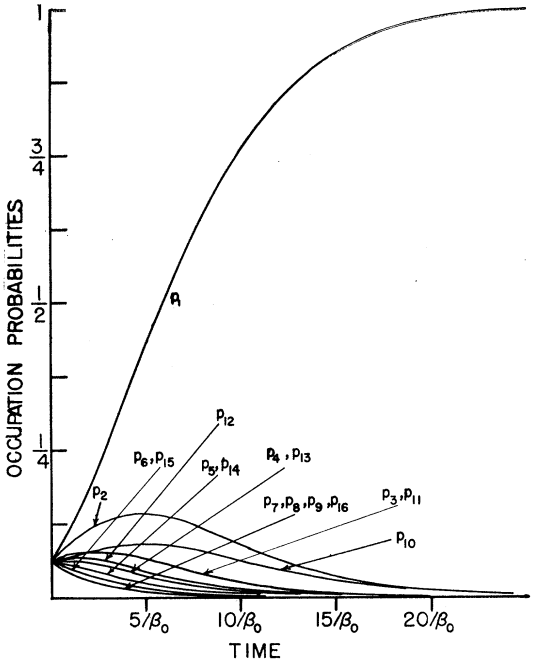

FIG. 2. Occupation probabilities p_i of the ground-state sublevels of cesium atoms illuminated by circularly polarized cesium $P_{\frac{3}{2}}$ resonance radiation ($\lambda = 8521$ A), as a function of time measured in units related to the incident light intensity β_0. The subscripts i have the same meaning as in Table I.

for pure $P_{\frac{1}{2}}$ (8943 A) radiation incident, and Fig. 2 shows them for pure $P_{\frac{3}{2}}$ radiation. Figure 3 shows the difference between the occupation probabilities of the two $m_F = 0$ levels for $P_{\frac{3}{2}}$ radiation incident, the $F = 3$ level having the greater population; these two levels have the same population at all times when $P_{\frac{1}{2}}$ radiation is incident.

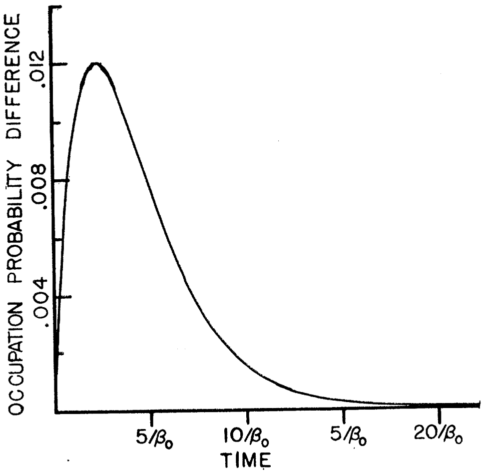

FIG. 3. Difference between the occupation probabilities of the two cesium ground-state sublevels having $m_F = 0$, illuminated by circularly polarized cesium $P_{\frac{3}{2}}$ resonance radiation ($\lambda = 8521$ A). The level with $F = 3$ has larger probability than the one with $F = 4$. The units of the time axis are inversely proportional to the incident light intensity, β_0.

Figure 4 shows the light absorption probability per atom, which increases for $P_{\frac{3}{2}}$ light and decreases to zero for $P_{\frac{1}{2}}$ light. This results from the fact that the 4, 4 level has a higher than average absorption probability for transitions to the $P_{\frac{3}{2}}$ level and zero probability to the other.

One of the special cases for which the occupation probabilities may be found exactly is the case of mixed illumination with equal intensities of both fine structure components; this case is simple because the absorption probabilities of all the levels of the ground state are the same, and the transition probability table obtained by combining Tables I and II may be treated as a matrix operator acting upon an occupation vector whose components are the occupation probabilities of the various levels of the ground state. Because of the double statistical weight of the $P_{\frac{3}{2}}$ level compared to the other, $\frac{1}{3}$ times Table I must be added to $\frac{2}{3}$ times Table II, a procedure which also gives the proper normalization of the result. The occupation vectors given in Table III are obtained by applying the operator n times to an initial occupation vector whose components are all equal to $\frac{1}{16}$, and gives the occupation probabilities of an atom which has scattered exactly n photons. The quantities for a real ensemble of atoms exposed to a known amount of light may be found by computing the probabilities of scattering each integral number of photons and summing the occupation vectors with these probabilities as weights.

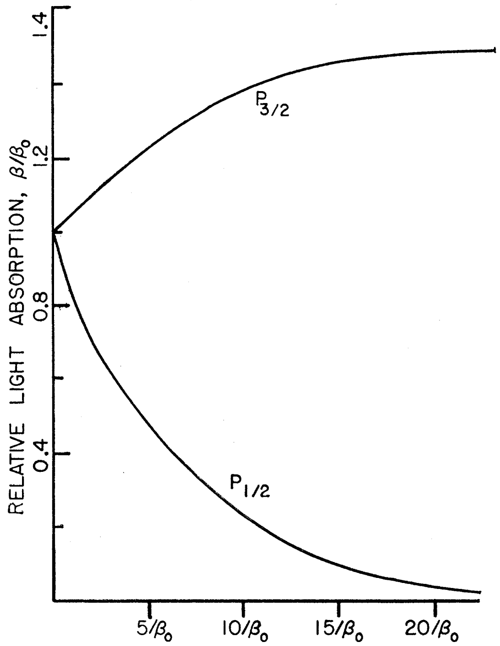

FIG. 4. Absorption probability of circularly polarized cesium $P_{\frac{1}{2}}$ or $P_{\frac{3}{2}}$ resonance radiation by cesium atoms relative to the initial absorption probability, as a function of the length of time the atoms have been illuminated measured in units inversely proportional to the incident light intensity β_0.

The effect of unpolarized light upon an atom may be calculated by applying an operator deduced from the operator for circularly polarized light by changing the sign of each m_F and adding half the result to half the original operator. Table IV gives the result for equal intensities of the two fine structure components, and the effect is to favor the occupation of states with high absolute values of m_F, as well as to change the occupation probabilities of the two $m_F=0$ states with respect to each other.

Since the time required to place a large fraction of atoms in one state with available light intensities is longer than the transit time of gas atoms across a reasonably sized vacuum chamber, it is common practice to use an inert buffer gas to increase the time that optically pumped atoms are illuminated.[10,11] Although inert, the buffer gas is capable of causing transitions between the various sublevels; and while this contributes to the relaxation time in the ground state, by far the fastest relaxation occurs in the excited state.[11] If the buffer gas pressure is increased to the point where complete relaxation occurs in the excited state during the time that an atom is excited but relaxation in the ground state is still small, the optical pumping is considerably hindered unless one sublevel of the ground state has zero absorption probability, which is the case for the $F=4$, $m_F=4$ level when only $P_{\frac{1}{2}}$ light is used. In this case the result of pumping will still be to place a large fraction of the atoms in that state.

TABLE III. Occupation probabilities of the ground-state sublevels of cesium atoms which have scattered exactly n photons of circularly polarized mixed $P_{\frac{1}{2}}$ and $P_{\frac{3}{2}}$ cesium resonance radiation with equal intensities in the two lines.

F, m_F \ $n=$	1	2	3
4, 4	0.11500	0.17784	0.25221
4, 3	0.09814	0.13018	0.15630
4, 2	0.08286	0.09427	0.09691
4, 1	0.06822	0.06758	0.06023
4, 0	0.05715	0.04797	0.03747
4, −1	0.04670	0.03365	0.02320
4, −2	0.03784	0.02318	0.01409
4, −3	0.03059	0.01552	0.00459
4, −4	0.02495	0.00996	0.00397
3, 3	0.06246	0.06815	0.07604
3, 2	0.06553	0.07132	0.07282
3, 1	0.06677	0.06735	0.06050
3, 0	0.06615	0.05993	0.04764
3, −1	0.06373	0.05168	0.03716
3, −2	0.05946	0.04401	0.02935
3, −3	0.05335	0.03726	0.02385

TABLE IV. Occupation probabilities of the ground-state sublevels of cesium atoms which have scattered exactly n photons of unpolarized mixed $P_{\frac{1}{2}}$ and $P_{\frac{3}{2}}$ cesium resonance radiation with equal intensities in the two lines.

F, m_F \ $n=$	1	2	3	∞
4, 4	0.06998	0.07448	0.07744	0.08551
4, 3	0.06436	0.06504	0.06530	0.06558
4, 2	0.06036	0.05922	0.05855	0.05668
4, 1	0.05796	0.05609	0.05505	0.05219
4, 0	0.05716	0.05510	0.05396	0.05089
3, 3	0.05791	0.05659	0.05647	0.05867
3, 2	0.06250	0.06274	0.06276	0.06243
3, 1	0.06525	0.06531	0.06480	0.06233
3, 0	0.06616	0.06597	0.06530	0.06237

Although the kind of pumping light used can strongly affect the results, one also sees that two or more pumping schemes may give very similar results for some types of experiments; for example, the use of polarized or unpolarized light makes little difference to the population difference between the two $m_F=0$ states, compared to their population difference in the absence of optical pumping.

ACKNOWLEDGMENT

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[11] P. Bender, thesis, Princeton University (unpublished).