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Hyperfine-structure measurements in the first excited *D* levels of potassium, rubidium, and cesium by cascade-fluorescence spectroscopy

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Hyperfine structures in the first excited D levels of ^{39}K, ^{85}Rb, ^{87}Rb, and ^{133}Cs have been measured by cascade-fluorescence spectroscopy. We find that the magnetic-dipole coupling constants A (in MHz) are $|A(^{39}\mathrm{K},3\,^2D_{3/2})| < 1.8$, $|A(^{39}\mathrm{K},3\,^2D_{5/2})| < 2.2$, $A(^{85}\mathrm{Rb},4\,^2D_{3/2}) = +7.3(5)$, $A(^{85}\mathrm{Rb},4\,^2D_{5/2}) = -5.2(3)$, $A(^{87}\mathrm{Rb},4\,^2D_{3/2}) = +25.1(9)$, $A(^{87}\mathrm{Rb},4\,^2D_{5/2}) = -16.9(6)$, and $A(^{133}\mathrm{Cs},5\,^2D_{5/2}) = -22.1(5)$.

I. INTRODUCTION

Recently, several measurements of the hyperfine structures (hfs) in the excited D levels of alkali-metal atoms have been reported.[1-7] Hyperfine structures in the second excited levels have been measured using cascade-fluorescence spectroscopy,[1] while stepwise excitation using a cw dye laser has been used for the third and the higher excited D levels.[2-7] It has been found that the hfs is inverted in all of the $D_{5/2}$ levels (where it has been possible to determine their sign), while the $D_{3/2}$ levels are normal. It is worth noting that the fine-structure intervals in the D levels of alkali atoms are anomalously narrow and are often inverted.[8] Since the discovery of the anomalous hfs,[9] considerable theoretical interest has been generated in the calculation of fine and hyperfine structures in the D levels.[10-15] Measurement of the hfs in the *first excited D* levels, therefore, are very important in providing a complete picture of the hyperfine interaction in alkali atoms. However, these levels are particularly difficult to reach by optical excitation. Stepwise laser excitation using first excited P^o level as the intermediate level requires a tunable cw laser in the infrared, which is not available. Although these levels can be reached by cascade excitation, they fluoresce in the infrared where photomultiplier tubes cannot be used. We have succeeded in investigating these levels by using a multiple cascade scheme, and in this paper we report measurement of the sign and magnitude of the magnetic-dipole coupling constants in the $4\,^2D_{3/2}$ and $4\,^2D_{5/2}$ levels of ^{85}Rb and ^{87}Rb and in the $5\,^2D_{5/2}$ level of ^{133}Cs. We give upper limits on the magnitude of the magnetic-dipole coupling constant in the $3\,^2D_{3/2}$ and $3\,^2D_{5/2}$ levels of ^{39}K. Since this work was completed, a new measurement of the hfs of $5\,^2D_{5/2}$ level of ^{133}Cs has been reported.[16]

The basic scheme of our method is illustrated in Fig. 1 for the $4\,^2D$ levels of rubidium. The schemes for potassium and cesium are similar. Atoms in the ground $5\,^2S_{1/2}$ level are excited to the $6\,^2P^o_{1/2,3/2}$ levels by second resonance lines (4202 and 4216 to Å) from a rubidium discharge lamp. Atoms in the $6P^o$ levels then decay to the $6\,^2S_{1/2}$ and $4\,^2D_{3/2,5/2}$ levels, in addition to the $5\,^2S_{1/2}$ ground level. The $4\,^2D_{3/2,5/2}$ level atoms, which are of interest here, fluoresce in a region (~15000 Å) where conventional photomultipliers cannot be used. Therefore we observe the 7800- and 7947-Å resonance lines which are emitted when the $4\,^2D$ level atoms decay via $5\,^2P^o$ levels to the ground state. Electronic polarization in the $6\,^2P^o$ levels is created by using circularly polarized 4202- and 4216-Å light. The polarization of the $6P^o$ levels is partially carried over to the $4D$ levels when the atoms decay from the $6P^o$ to the $4D$ levels.[17] The polarization of the $4D$ level is monitored by observing the circular polarization of the 7800- or the 7947-Å fluorescent light. Radio-frequency transitions induced between the magnetic sublevels of the $4D$ level are detected as a change in the polarization of the 7800- or the 7947-Å light.[18] Magnetic-dipole coupling con-

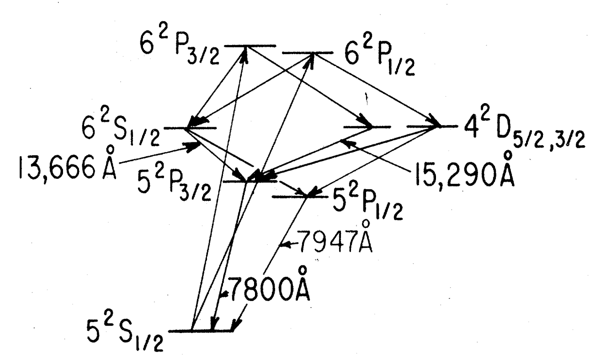

FIG. 1. Schematic illustration (not drawn to scale) of the two-step cascade scheme for rubidium. Second resonance lines ($5\,^2S_{1/2} \rightarrow 6\,^2P^o_{3/2,1/2}$) are used to excite the atoms, and 7800 and 7947-Å fluorescent lines are detected.

stants A for the D level are deduced from the observed transition frequency and the magnetic field. We have also performed cascade-decoupling[17] experiments to determine the sign of the magnetic-dipole coupling constants A.

The experiments reported in this paper are similar to those of Tai *et al.*[1] for the case of second excited D levels of Rb and Cs. However, the important difference between our experiments and those of Tai *et al.* is the presence of an additional cascading stage which introduces several problems. The major problem is that a number of intermediate levels are involved in the cascade. Most of the excited atoms actually cascade via the first excited S level, which contribute only to the noise in the signal of the ^2D levels. Moreover, the detected light, which connects the $5\,^2P^o$ levels to the ground state, can easily be trapped. Therefore, our experiment was carried out at low vapor density, at the expense of fluorescent intensity. In radio-frequency resonance experiments, there is often overlapping of the ^2D level resonances with resonances in the other levels. For example, if 7800-Å light is observed, resonances in the $6\,^2P^o_{1/2}$, $6\,^2P^o_{3/2}$, $6\,^2S_{1/2}$, $4\,^2D_{3/2}$, $4\,^2D_{5/2}$, and $5\,^2P^o_{3/2}$ levels can be observed. The problem of overlapping resonances is avoided, to some extent, by taking data at several different rf frequencies. An additional cascading stage also means that the atomic polarization is in general further degraded. The signal degradation is especially bad in the lowest $^2P^o$ level, which has a moderately large hfs for heavy alkali atoms so that, at the magnetic field at which we operate, the electronic and the nuclear spins are still partially coupled. This coupling affects the rf resonance signals from the D levels, making some of them too small to be observed.[18] The analysis of the cascade-decoupling signals becomes especially difficult because many branching ratios and intermediate level lifetimes are uncertain.

II. THEORY

The basic theory of cascade-fluorescence experiments has been treated extensively in previous publications.[1,17,18] We recapitulate some of the important relations for completeness.

The hfs Hamiltonian for an atom with electronic and nuclear angular momenta J and I, respectively, placed in a magnetic field H is

$$\mathcal{H}=hA\vec{I}\cdot\vec{J}+hB\,\frac{3(\vec{I}\cdot\vec{J})^2+\frac{3}{2}(\vec{I}\cdot\vec{J})-I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}+\mu_B g_J\vec{J}\cdot\vec{H}+\mu_N g_I\vec{I}\cdot\vec{H}\,, \qquad (1)$$

where g_J and g_I are the electronic and nuclear g factors, and μ_B and μ_N are the Bohr magneton and nuclear magneton, respectively. A and B are the magnetic-dipole and the electric quadrupole coupling constants, respectively. The aim of our experiments is to determine the value of the magnetic-dipole coupling constant A. The electric quadrupole coupling constant B is usually too small to be determined in our experiments.

The general expression for fluorescent light intensity of polarization $\hat{u}$, detected in a small solid angle $\Delta\Omega$, in a two-step cascade experiment is given by[17]

$$\frac{\Delta I}{\Delta\Omega}=\frac{16}{9}\left(\frac{e}{mc}\right)^8\frac{\omega_{eb}\omega_{bs}\omega_{sf}}{c\hbar^5\omega_{eg}^2}\,\frac{u}{(2I+1)(2J_g+1)}\sum_{\mu\nu}\sum_{ij}\sum_{kl}\sum_{mn}\langle i|\vec{p}|k\rangle\cdot\langle l|\vec{p}|j\rangle \times\langle k|\vec{p}|m\rangle\cdot\langle n|\vec{p}|l\rangle\langle m|\hat{e}\cdot\vec{p}|g\mu\rangle\langle g\mu|\hat{e}^*\cdot\vec{p}|n\rangle\langle j|\hat{u}\cdot\vec{p}|f\nu\rangle \times\langle f\nu|\hat{u}^*\cdot\vec{p}|i\rangle(\Gamma_e+i\omega_{mn})^{-1}(\Gamma_b+i\omega_{kl})^{-1}(\Gamma_s+i\omega_{ij})^{-1}\,. \qquad (2)$$

Here $\hat{e}$ is the polarization and u the energy density of the exciting light, and the subscripts g, e, b, s, and f denote the ground, excited, first-branch, second-branch, and the final levels, respectively. In the case of Rb, the ground level is the $5\,^2S_{1/2}$ level, the excited levels are the $6\,^2P^o_{3/2}$ and $6\,^2P^o_{1/2}$ levels, the first-branch level is either the $4\,^2D_{3/2}$ level or the $4\,^2D_{5/2}$ level, the second-branch level is either the $5\,^2P^o_{1/2}$ level or the $5\,^2P^o_{3/2}$ level, and the final level is the $5\,^2S_{1/2}$ level. The magnetic sublevels of the ground level are denoted by μ, those of the excited level by m and n, those of the first branch by k and l, those of the second branch by i and j, and those of the final level by ν. The frequency differences are denoted by ω, and the decay rates are denoted by Γ. The observed fluorescence given by (2) must be weighted with appropriate branching ratios for different cascading routes which contribute to the observed fluorescence. We have evaluated (2), for the specific states in which we are interested using an IBM 360 computer.

A. Cascade decoupling

Equation (2) predicts that any longitudinal electronic polarization produced by the exciting light is partially retained in the cascade.[17] The polarization of the fluorescent light usually increases with the external magnetic field due to the decoupling of the electronic and nuclear angular momenta. The shape of the decoupling curve, predicted by (2), is sensitive to both the sign and the absolute value of the magnetic-dipole coupling constant.[1] We have used the decoupling technique to determine the sign of A in the $5\,^2D_{5/2}$ level of ^{133}Cs.

B. Cascade radio-frequency spectroscopy

Equation (2) gives the intensity of the fluorescent light of a given polarization in the absence of radio-frequency fields. In most of our experiments we induce magnetic-dipole transitions between the magnetic sublevels of the atoms. These transitions are induced by rf fields of a few hundred MHz for applied magnetic fields of a few hundred gauss. When an rf transition is induced, it decreases the electronic polarization in that level, and it is detected by observing a decrease in the polarization of the fluorescent light. The theory of cascade rf spectrcscopy is discussed in detail by Gupta *et al.*[18]

We deduce the magnetic-dipole coupling constant from the frequency of the rf and the magnitude of the magnetic field at which the transition is observed. Although we calculate the transition frequencies as a function of the applied magnetic field by diagonalizing (1) with the aid of an IBM 360 computer, we give below a simple expression to facilitate the discussion. In most of our experiments, the external magnetic field is so high that m_J and m_I are very nearly good quantum numbers. Under these conditions, the frequency of a transition between levels characterized by (m_J, m_I) and (m_J-1, m_I) is given by

$$\begin{aligned}\nu = {} & g_J \frac{\mu_B}{h} H + A m_I \\ & + \frac{hA^2}{2g_J\mu_B H}\left[(I^2+I-m_I^2)+m_I(2m_J-1)\right] \\ & + \frac{3B}{4}\,\frac{[I(I+1)-3m_I^2](1-2m_J)}{J(2J-1)I(2I-1)} \\ & + \text{higher order terms.}\end{aligned} \tag{3}$$

In general $2J(2I+1)$ different transitions corresponding to different values of m_I and m_J are possible. Under the conditions of our experiment (large H), the second-order term in A is much smaller than the first-order term. Also, the last term in (3) is quite small compared to the second term because $B \ll A$. Therefore, for a given value of m_I, transitions corresponding to different values of m_J are close together. In the experiments reported here, either the separation between transitions corresponding to different values of m_J are smaller than the natural widths of the transitions, or we apply a sufficiently intense rf magnetic field that we observe multiple-quantum transitions between transitions of the same m_I but different m_J. Thus we observe $(2I+1)$ well-resolved resonances, each resonance characterized by a value of m_I. If the polarization in the excited levels is not too complicated, the frequency at which each of these resonance occurs is independent of m_J because the average value of $(2m_J-1)$ for $m_J = J, J-1, \ldots -J+1$ is zero.[19] The resonance may be broadened, however, depending on the value of B. We have not been able to deduce the values of B in our experiments; however, we have been able to give upper limits on the value of B in some cases from the widths of these resonances. Signal-to-noise ratios in our experiments are such that we are unable to observe single-quantum resonances at low rf powers. For example, in the $5\,^2D_{5/2}$ level of ^{133}Cs there are $2J(2I+1)=40$ single-quantum resonances, while there are only $(2I+1)=8$ multiple-quantum transitions. Therefore we gain a large factor in the signal-to-noise ratio when we observe the multiple-quantum resonances.

We note that (3) assumes that the quadratic shift of the m_J sublevels due to the decoupling of L and S in an external magnetic field of a few hundred gauss is negligible. This shift is completely negligible for the $5\,^2D_{5/2}$ level of ^{133}Cs. However, the fine-structure interval in the $4\,^2D$ level of Rb is small enough (13.36 GHz) that, for a magnetic field of 250–400 G, different m_J sublevels shift on the order of 1 MHz. If the polarization of the $4\,^2D$ level is not too complicated (i.e., if different m_J resonances are of equal strength), this causes a broadening of the multiple-quantum resonance without any significant shift of the center frequency.

Finally, two effects can cause different resonances corresponding to different values of m_I to be of unequal size. One of these is due to the coupling of I and J. If the hfs in one of the intermediate levels is large enough so that I and J are coupled to each other at the magnetic field of interest, this coupling can modify the intensities of different m_I resonances by different amounts.[18] The second effect which can give rise to unequal strengths of different m_I resonances is due to "nonwhite" optical excitation. If the frequency distribution of the exciting light is such that atoms are mainly excited out of only one hfs component of the ground level, certain amount of nuclear polarization can be produced in such excitation. This nuclear polarization manifests itself in different sizes of different m_I resonances. For example, consider excitation of a $D_{3/2}$ level by spontaneous decay of a $P^o_{1/2}$ level which is optically excited from an $S_{1/2}$ ground level. If atoms are excited mainly out of the $F=(I+1/2)$ component of the ground-level hfs by σ_+ exciting light, the strength of m_I resonances in the $P^o_{1/2}$ and the $D_{3/2}$ levels increases with increasing magnetic field (for constant rf frequency) for a positive A, while it decreases with increasing magnetic field for negative A.[1] For σ_- exciting light, the effect is reversed. We have used this effect to determine the sign of A in the D levels of Rb. One does not even have to know the absolute sense of circular polarization (σ_+ or σ_-), because one can deduce the sign of A simply by comparing the relative magnitudes of the D-level resonances with those of the P^o-level resonances. Since the P^o-level hfs are known to be positive, the sign of the D-level hfs can easily be deduced. A detailed description of this effect is given by Tai *et al.*[1]

III. EXPERIMENT

The apparatus used for this experiment is shown schematically in Fig. 2. Second resonance lines (4202 and 4216 Å for Rb, and 4555 and 4593 Å for Cs) from a microwave-excited resonance lamp are selected by an interference filter. This light is circularly polarized by a Polaroid linear polarizer and a mica quarterwave plate, and irradiates a Pyrex cell containing alkali metal along the axis of the apparatus (z axis). The fluorescent light, selected by an interference filter [50 Å full width at half maximum (FWHM)], is detected by a photomultiplier (PM) tube in a solid angle of about 0.3 sr along the z axis. We observe D_1 light (7947 Å for Rb) for the case of $4\,^2D_{3/2}$ level and observe the D_2 light (7800 Å for Rb and 8521 Å for Cs) for the case of $D_{5/2}$ levels. The alkali-metal cell is placed inside an oven to get the desired vapor density. These experiments were performed at approximately 30 °C for Cs, 60 °C for Rb, and 90 °C for K. For radio-frequency-spectroscopy experiments, the cell is placed inside a $\lambda/4$ or $\lambda/2$ resonance cavity[18] which produces rf magnetic fields of the order of 1 G. Quasistatic magnetic fields are produced in the z direction by a pair of Helmholtz coils which were calibrated by optical pumping of Rb. To prevent the D_1 and D_2 resonance lines from the lamp from reaching the PM tube, we used a Schott colored-glass filter (BG18) in front of the lamp. The fluorescent light passes through a rotating quarterwave plate and a fixed linear polarizer. In this way the fluorescent light intensity was modulated at 13 Hz, the degree of modulation being proportional to the degree of circular polarization of light. As a result, synchronous detection using a lock-in amplifier was made possible. Further signal averaging was accomplished by a PDP-8/S minicomputer. In decoupling experiments, we observe the circular polarization of the fluorescent light as a function of the quasistatic magnetic field in the z direction. In radio-frequency resonance experiments, we apply an rf magnetic field of fixed frequency, sweep the quasistatic magnetic field in the z direction, and observe a sharp change in the polarization of the fluorescent light as we go through the rf resonance. The magnetic field was scanned at the rate of about 3 min/sweep. However, signal averaging of several hours was required to achieve the signal-to-noise ratio represented by Figs. 3–11.

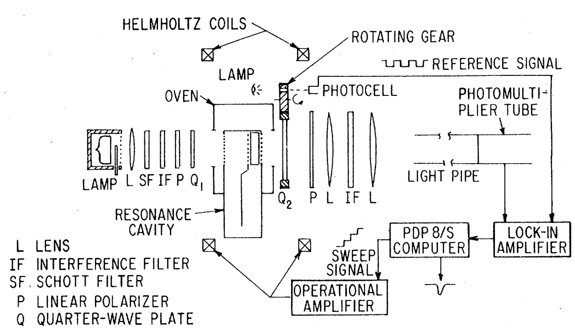

FIG. 2. Schematic diagram of the apparatus used in cascade radio-frequency experiments.

IV. RESULTS

A. $4\,^2D_{5/2}$ level of ^{85}Rb and ^{87}Rb

Figure 3 shows six barely resolved rf resonances in the $4\,^2D_{5/2}$ level of ^{85}Rb. Figure 3 and all other figures in this article have arbitrary polarization scale. Figure 4 shows the same resonances in ^{85}Rb atoms when these atoms were excited by light from an ^{87}Rb lamp. The two curves in Fig. 4 correspond to σ_+ and σ_- excitation. Ground-level hfs in ^{85}Rb is only 3.3 GHz while it is 6.8 GHz in ^{87}Rb, therefore the ^{85}Rb atoms are mainly excited out of their ground-level $F=3$ hfs component. Therefore we can use the unequal sizes of different resonances corresponding to various values of m_I to determine the sign of A, as mentioned in Sec. II B. We note that for σ_+ excitation, the $6\,^2P^o_{3/2}$ resonances at low magnetic fields have a much larger magnitude than those at high field, while the opposite is true for the $4\,^2D_{5/2}$ resonances. With σ_- excitation, the trend is reversed. Since $6\,^2P^o_{3/2}$ level hfs is known to be normal, our data indicate that the hfs in the $4\,^2D_{5/2}$ level is inverted. We should mention that it is difficult to avoid accidental overlap of some

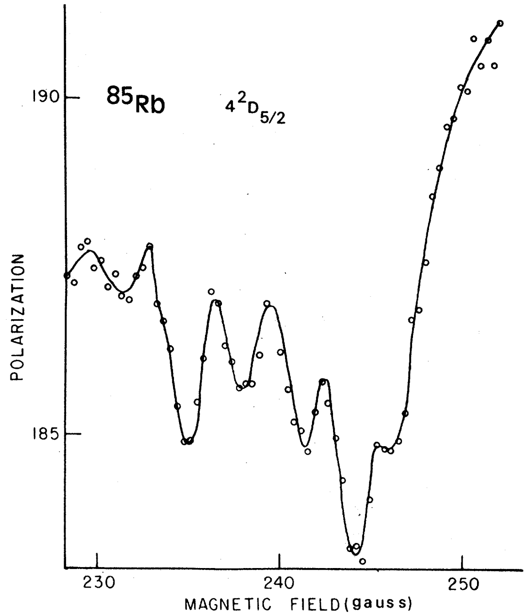

FIG. 3. Radio-frequency resonances in the $4\,^2D_{5/2}$ level of ^{85}Rb at a transition frequency of 403.6 MHz. There are six barely resolved peaks corresponding to $m_I = -\frac{5}{2}$ to $+\frac{5}{2}$. The solid line is drawn freehand for visual aid and does not represent any fitting curve.

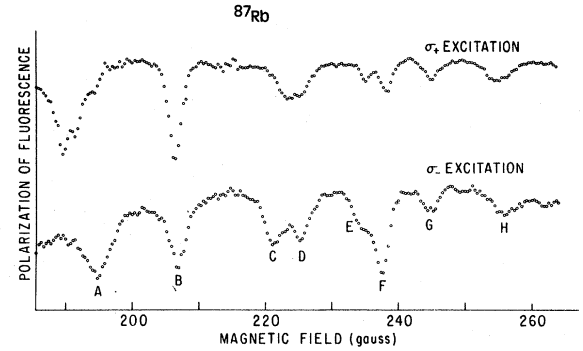

FIG. 5. Radio-frequency resonances in the $6\,^2P^o_{3/2}$ level (A, B, C, and F) and $4\,^2D_{5/2}$ level (D, E, G, and H) of ^{87}Rb atoms excited by light from an ^{85}Rb lamp. We identify D, E, G, and H resonances with m_I quantum numbers of $-\frac{3}{2}$, $-\frac{1}{2}$, $\frac{1}{2}$, and $\frac{3}{2}$, respectively. The transition frequency was 403 MHz.

of these resonances with those in some of the intermediate levels in these experiments. There are a number of transitions in the $5\,^2P^o_{3/2}$ level in the magnetic-field region covered by Figs. 3 and 4. We do not observe these resonances, and we believe that they are so weak and so broad that their presence does not significantly affect the positions of the $4\,^2D_{5/2}$ level resonances. This is because the radiative lifetime of the $5\,^2P_{3/2}$ level is very short [$\tau = 25.5$ nsec (Ref. 20)] and our rf fields are not intense enough to give a significant signal. Also, the hfs in the $5\,^2P^o_{3/2}$ level is large enough[21] ($A = 25$ MHz, $B = 26$ MHz) that different m_J resonances for a given value of m_I are spread

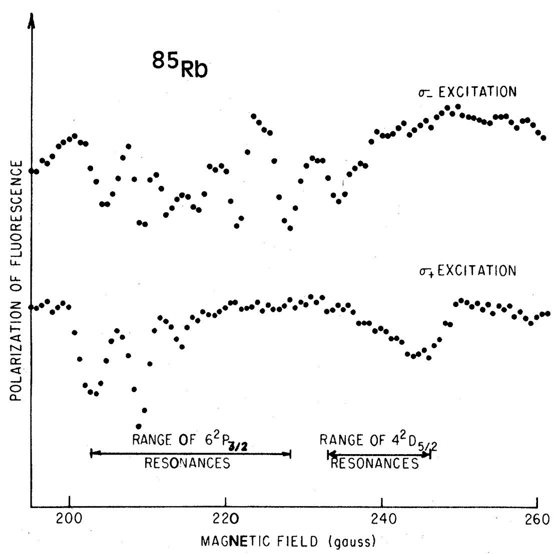

FIG. 4. Radio-frequency resonances in the $6\,^2P^o_{3/2}$ and the $4\,^2D_{5/2}$ levels of ^{85}Rb atoms excited by light from an ^{87}Rb lamp. The $4\,^2D_{5/2}$ resonances are not resolved here. The relative amplitudes of the $P^o_{3/2}$ and $D_{5/2}$ resonances change in opposite directions as excitation changes from σ_+ to σ_- (see the text).

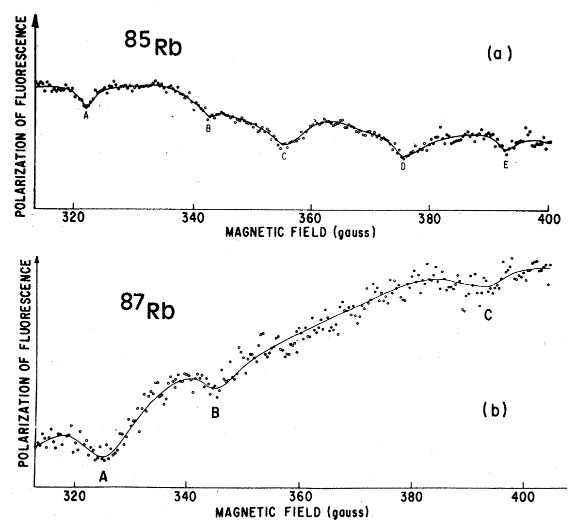

FIG. 6. Radio-frequency resonances in the $4\,^2D_{3/2}$ levels of (a) ^{85}Rb and (b) ^{87}Rb at 403.2 MHz. In (a), A, C, and E are $6\,^2P^o_{1/2}$ level resonances and B and D are the $4\,^2D_{3/2}$ level resonances. We identify B and D with m_I quantum numbers of $\frac{5}{2}$ and $-\frac{5}{2}$, respectively. In (b), A, B, and C correspond to $m_I = \frac{3}{2}, \frac{1}{2}$, and $-\frac{3}{2}$, respectively. The solid lines are drawn freehand for visual aid and do not represent any fitting curve.

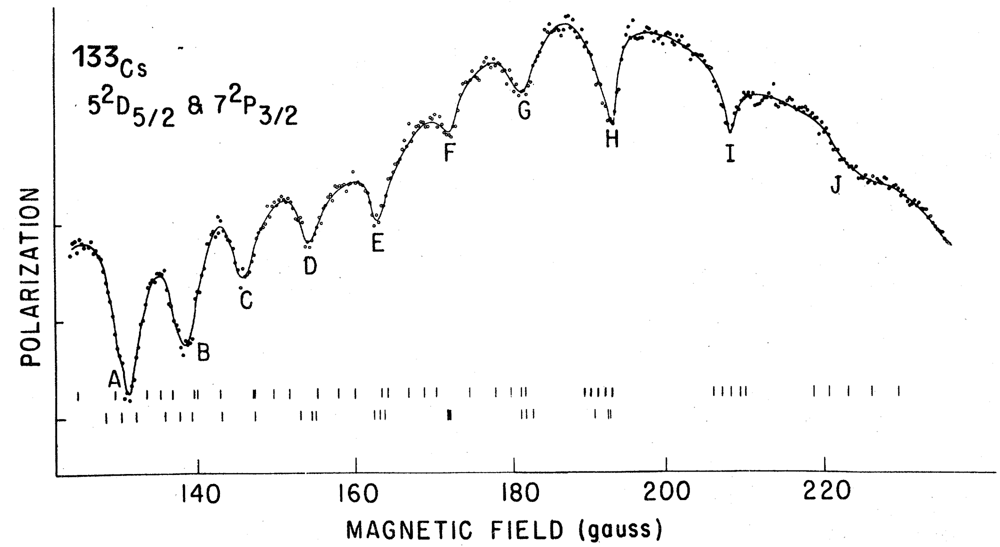

FIG. 7. Radio-frequency resonances in the $5\,^2D_{5/2}$ level (I, J) and in the $7\,^2P^o_{3/2}$ level (A–H) of ^{133}Cs at 302.8 MHz. Top row of markers gives the calculated positions of the $5\,^2D_{5/2}$ level resonances while the bottom row gives the expected positions of the $7\,^2P^o_{3/2}$ level resonances. Resonances I and J correspond to $m_I = \frac{5}{2}$ and $= \frac{7}{2}$, respectively. The solid line through the data points has been drawn to guide the eye.

out quite a bit giving rise to broad and weak signals.

Figure 5 shows the rf resonances in the $6\,^2P_{3/2}$ and the $4\,^2D_{5/2}$ levels of ^{87}Rb. The ^{87}Rb atoms were excited by light from an ^{85}Rb lamp. The resonances marked by letters A, B, C and F are in the $6\,^2P^o_{3/2}$ level, while those marked by D, E, G, and H are in the $4\,^2D_{5/2}$ level. There should be two other resonances in this field range, but they are too weak to be observed. There should be a group of $5\,^2P_{3/2}$ level resonances centered at 224 G which is difficult to observe because of the short radiative lifetime and moderately large hfs for this level, as discussed above for the case of ^{85}Rb. Another resonance at approximately 250 G, due to the $6\,^2P_{1/2}$ level, is too weak to be observed because of poor branching[22] into the observed channel. The difference in the relative sizes of the various $6\,^2P^o_{3/2}$ and $4\,^2D_{5/2}$ level resonances, unfortunately, are not as pronounced as for the case of ^{85}Rb. However, these data are consistent with

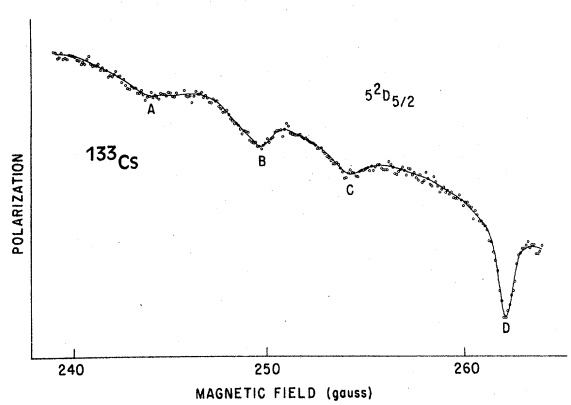

FIG. 8. Radio-frequency resonances in the $5\,^2D_{5/2}$ (B, D) and the $7\,^2P^o_{3/2}$ (A, C) levels of ^{133}Cs at a transition frequency of 415.4 MHz. Solid line through the data points has been drawn to guide the eye.

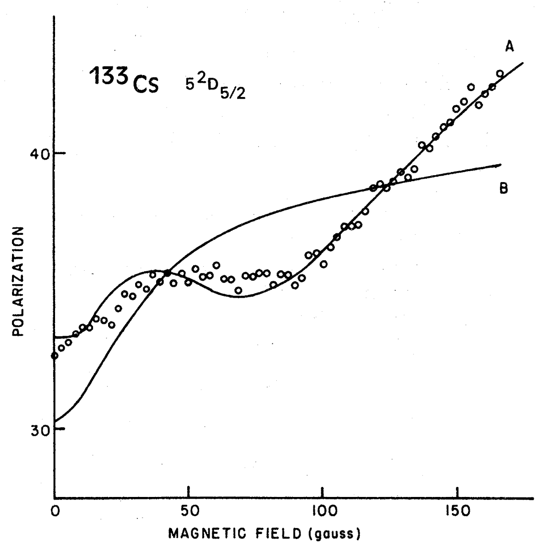

FIG. 9. ^{133}Cs $5\,^2D_{5/2}$ level decoupling data with σ_+ excitation. Curves A and B are the least-squares-fitted curves assuming an hfs constant of -22 and $+22$ MHz, respectively.

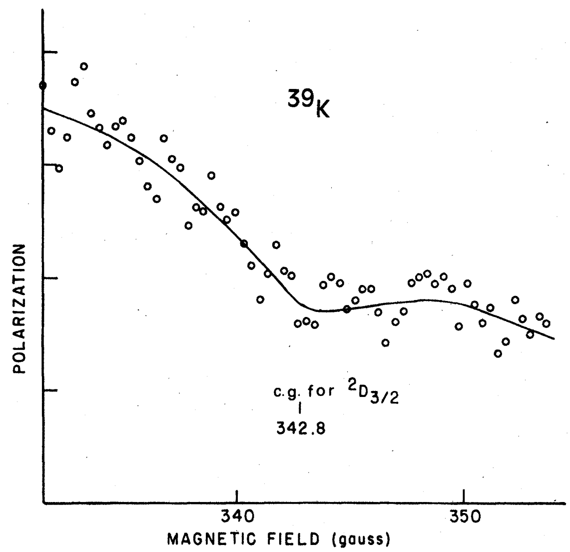

FIG. 10. Unresolved rf resonances in the $3\,^2D_{3/2}$ level of ^{39}K at a transition frequency of 383.9 MHz.

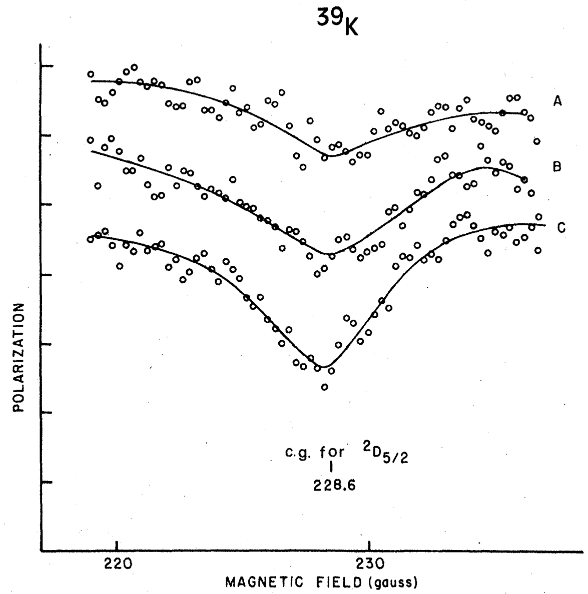

FIG. 11. Unresolved rf resonances in $3\,^2D_{5/2}$ level of ^{39}K at a transition frequency of 384.0 MHz. Curves A, B, and C correspond to rf power of 10, 20, and 34 W.

an inverted hfs in the $4\,^2D_{5/2}$ level, as deduced from the ^{85}Rb data. Each peak in Fig. 5 is a group of three (for P^o level) or five (for D level) unresolved resonances as pointed out in Sec. II B. Slight shifts in the positions of the peaks in Fig. 5 for two different polarizations are due to changes in the relative magnitudes of the overlapping resonances within each group. From these data we deduce the values of A for $4\,^2D_{5/2}$ levels of ^{85}Rb and ^{87}Rb, and these are listed in Table I.

B. $4\,^2D_{3/2}$ level of ^{85}Rb and ^{87}Rb

Radio-frequency resonance data for the $4\,^2D_{3/2}$ levels of ^{85}Rb and ^{87}Rb are shown in Figs. 6(a) and 6(b), respectively. In Fig. 6(a), resonances A, C, and E are due to the $6\,^2P^o_{1/2}$ level. Between points B and D, there should be a total of $2I+1=6$ resonances due to the $4\,^2D_{3/2}$ level. We believe some of these overlap with resonance C and some between points C and D are too small to be observed due to recoupling of the nuclear spin in the $5\,^2P^o_{1/2}$ level. Only two $4\,^2D_{3/2}$ resonances, B and D, can be distinctly observed. In Fig. 6(b) for ^{87}Rb, we can see three out of a total of $2I+1=4$ resonances. The second resonance on the high-field side of the center of gravity, somewhere between points B and C, is again too small to be observed. Such identification of the $4\,^2D_{3/2}$ resonances for ^{85}Rb and ^{87}Rb yields hfs constants which agree with the ratio of the nuclear magnetic-dipole moments of the two isotopes. We have ob-

TABLE I. Experimental and theoretical values of hfs in the first excited D levels of K, Rb, and Cs.

Atom	State	A_{exp} (MHz)	B_{exp} (MHz)	A^{a}_{Theo} (MHz)	A^{b}_{Theo} (MHz)
^{39}K	$3\,^2D_{3/2}$	< 1.8			
	$3\,^2D_{5/2}$	< 2.2			
^{85}Rb	$4\,^2D_{3/2}$	+ 7.3(5)	< 4		
	$4\,^2D_{5/2}$	− 5.2(3)			
^{87}Rb	$4\,^2D_{3/2}$	+25.1(9)	<10	+ 7.78	+28.6
	$4\,^2D_{5/2}$	−16.9(6)	<11	−14.98	−15.6
^{133}Cs	$5\,^2D_{5/2}$	−22.1(5)		−21.42	

[a] G. Belin, L. Holmgren, and S. Svanberg, Phys. Scr. 13, 351 (1976); 14, 39 (1976).
[b] I. Lindgren, J. Lindgren, and A.-M. Mårtensson, Z. Phys. A279, 113 (1976).

served these resonances with narrow-line excitation (i.e., by exciting ^{85}Rb atoms with an ^{87}Rb lamp and vice versa) and from the relative size of different resonances, we conclude that the sign of the hfs in the $4\,^2D_{3/2}$ level of Rb is normal. From a measurement of the positions of these resonances we have deduced the hfs constants listed in Table I.

C. $5\,^2D_{5/2}$ level of ^{133}Cs

The rf resonance data for the $5\,^2D_{5/2}$ level of ^{133}Cs are shown in Figs. 7 and 8. Data shown in Fig. 7 were taken at an rf frequency of 302.8 MHz. Resonances A to H are due to the $7\,^2P^o_{3/2}$ level and agree very well with their expected positions indicated by the bottom row of markers. We see only two $5\,^2D_{5/2}$ resonances, marked I and J. The top row of markers indicates the expected positions of various rf transitions in the $5\,^2D_{5/2}$ level calculated by assuming that $|A|=22.1$ MHz and $B=0$ for this level. At fields below 200 G, the $5^2D_{5/2}$ resonances badly overlap with the $7\,^2P^o_{3/2}$ resonances. Moreover, it is difficult to drive multiple-quantum resonances in this region because the magnetic field is not high enough to group these resonances according to their m_I quantum numbers. Therefore, only two resonances, marked I and J, are observed. In order to confirm our identification of these resonances, we have observed these resonances at 415.4 MHz as shown in Fig. 8. Here A and C are the $7\,^2P^o_{3/2}$ level resonances while B and D are $5\,^2D_{5/2}$ level resonances. Overlapping $7\,^2P^o_{3/2}$ and $5\,^2D_{5/2}$ resonances at G in Fig. 7 separate to become resonances A and B in Fig. 8 at higher magnetic field. Resonances C and D in Fig. 8 correspond to overlapping resonances at H in Fig. 7. Our identification of $5\,^2D_{5/2}$ resonances leads to the positions of all possible $5\,^2D_{5/2}$ resonances as shown by the top row of markers in Fig. 7. Because the agreement is very good with the positions and widths of the few observed $5\,^2D_{5/2}$ resonances, we believe the identification to be correct.

The sign of the magnetic-dipole coupling constant A for the $5\,^2D_{5/2}$ level is determined by the decoupling technique. Open circles in Fig. 9 show the decoupling data. Circularly polarized (σ_+) exciting light is used and circular polarization of the 8521-Å fluorescent light is observed as a function of a longitudinal (along the z axis) magnetic field. The solid curves represent a least-squares fit with two adjustable parameters, an overall scale factor, and the relative contribution of two major cascade routes. An overall scale factor is required because our polarizers are not perfect. We treat the relative contribution of the two major cascade routes as an adjustable parameter because the branching ratios are uncertain. The signal was calculated from Eq. (2) using the IBM 360 computer. The two dominant cascade routes are

$$6\,^2S_{1/2}\rightarrow 7\,^2P^o_{3/2}\rightarrow 7\,^2S_{1/2}\rightarrow 6\,^2P^o_{3/2}\rightarrow 6\,^2S_{1/2}$$

and

$$6\,^2S_{1/2}\rightarrow 7\,^2P^o_{3/2}\rightarrow 5\,^2D_{5/2}\rightarrow 6\,^2P^o_{3/2}\rightarrow 6\,^2S_{1/2}.$$

There are three other cascade routes with minor contributions to the detected signal:

$$6\,^2S_{1/2}\rightarrow 7\,^2P^o_{1/2}\rightarrow 7\,^2S_{1/2}\rightarrow 6\,^2P^o_{3/2}\rightarrow 6\,^2S_{1/2},$$

$$6\,^2S_{1/2}\rightarrow 7\,^2P^o_{1/2}\rightarrow 5\,^2D_{3/2}\rightarrow 6\,^2P^o_{3/2}\rightarrow 6\,^2S_{1/2},$$

and

$$6\,^2S_{1/2}\rightarrow 7\,^2P^o_{3/2}\rightarrow 5\,^2D_{3/2}\rightarrow 6\,^2P^o_{3/2}\rightarrow 6\,^2S_{1/2}.$$

Alternate ways of fitting with inclusion of these minor cascade routes yield essentially the same curves. From radio-frequency resonance experiments we deduce $|A(5\,^2D_{5/2})|=22$ MHz. In Fig. 9, the curves marked A and B represent best fits with $A=-22$ MHz and $A=+22$ MHz, respectively. These data are consistent with a negative dipole coupling constant for this level.

D. $3\,^2$D levels of ^{39}K

Radio-frequency resonance data for the $3\,^2D_{3/2}$ and the $3\,^2D_{5/2}$ levels of ^{39}K are shown in Figs. 10 and 11, respectively. The hfs in these levels are too small to be resolved; therefore, we obtain single resonances at the center of gravity ($H=h\nu/g_J\mu_B$) of the $D_{3/2}$ and $D_{5/2}$ levels. In Fig. 11, curves A, B, and C represent increasing rf power. From the widths of these resonances, extrapolated to zero rf power, we obtain upper limits on the hfs in these levels. In this analysis we have taken the radiative lifetime of these levels to be 40 nsec.[22]

V. DISCUSSION

The results of this work are compiled in Table I. The error bars represent two standard deviations of the mean plus allowance for possible systematic errors. As it was pointed out in Sec. II, we do not observe resolved resonances corresponding to different values of m_J. Therefore, we have been unable to determine the value of the electric quadrupole coupling constant B. We have, however, obtained upper limits on B from the widths of these resonances in some cases and these are shown in Table I.

We find that the hfs of both the $4\,^2D_{5/2}$ level of Rb and the $5\,^2D_{5/2}$ level of Cs are inverted, while that of the $4\,^2D_{3/2}$ level of Rb is normal. We have

TABLE II. A compilation of experimental values of magnetic-dipole coupling constant A in the D levels of alkali atoms. Only the absolute value of A has been determined where its sign is not indicated.

	$D_{3/2}$ State A (MHz)				$D_{5/2}$ State A (MHz)			
n	^{39}K	^{85}Rb	^{87}Rb	^{133}Cs	^{39}K	^{85}Rb	^{87}Rb	^{133}Cs
3	< 1.8[a]				<2.2[a]			
4		+7.3(5)[a]	+25.1(9)[a]			−5.2(3)[a]	−16.9(6)[a]	
5	0.44(10)[c]	+4.18(20)[b]	+14.43(23)[b]	+48.6(2)[f]	0.24(7)[c]	−2.12(20)[b]	− 7.44(10)[b]	−22.1(5)[a,g]
6	0.2(2)[c]	+2.28(6)[d]	+ 7.84(5)[d]	+16.30(15)[b]	0.1(1)[c]	−0.95(20)[d]	− 3.4(5)[d]	− 3.6(1.0)[b]
7		+1.34(1)[d]	+ 4.53(3)[d]	7.4(2)[e]		−0.55(10)[d]	− 2.0(3)[d]	− 1.7(2)[e]
8		+0.84(1)[d]	+ 2.84(2)[d]	+ 3.98(8)[e]		−0.35(7)[d]	− 1.20(15)[d]	− 0.85(20)[e]
9			1.90(1)[d]	+ 2.37(3)[e]			(−)0.80(15)[d]	− 0.45(10)[e]
10				+ 1.52(3)[e]				− 0.35(10)[e]
11				1.055(15)[e]				0.24(6)[e]
12				0.758(12)[e]				0.19(5)[e]
13				0.556(8)[e]				0.14(4)[e]
14				0.425(7)[e]				
15				0.325(8)[e]				
16				0.255(12)[e]				
17				0.190(12)[e]				
18				0.160(10)[e]				

[a] Results of this work.
[b] C. Tai, W. Happer, and R. Gupta, Phys. Rev. A 12, 736 (1975).
[c] G. Belin, L. Holmgren, I. Lindgren, and S. Svanberg, Phys. Scr. 12, 287 (1975).
[d] G. Belin, L. Holmgren, and S. Svanberg, Phys. Scr. 13, 351 (1976).
[e] G. Belin, L. Holmgren, and S. Svanberg, Phys. Scr. 14, 39 (1976).
[f] K. Fredriksson, H. Lundberg, and S. Svanberg, Phys. Rev. A 21, 241 (1980).
[g] Recent measurement of Fredriksson et al. (Ref. f above) gives A ($5\,^2D_{5/2}$, ^{133}Cs) = −21.2(1) MHz.

been unable to determine the hfs of the $5\,^2D_{3/2}$ level of ^{133}Cs due to poor signal-to-noise ratio. The inverted hfs in the $D_{5/2}$ levels are consistent with hfs in the higher $D_{5/2}$ levels, which are also inverted. In Table II we list all the hfs that have been measured in the D levels of K, Rb, and Cs. In Table I we also give some theoretical values of the magnetic-dipole coupling constant A that have recently been calculated. Column 5 gives the values calculated by Belin *et al.*[6,7] A Hartree-Fock calculation has been used and polarization effects have been included to all orders. Correlation effects, however, have been neglected. The agreement with experiment is very good for the $D_{5/2}$ level. Column 6 gives the results of many-body calculations by Lindgren *et al.*[12] using approximate Brueckner or natural orbitals. These calculations include polarization effects to all orders and all lowest-order correlation effects. The agreement with experiment is excellent.

Recently Liao *et al.*[23] have determined the complete hfs Hamiltonian for the $4\,^2D$ level of Rb by cascade anticrossing spectroscopy. They find that the contact term $(\vec{S}\cdot\vec{I})$ has a very large contribution to the hfs of 4D level and that $\langle r^{-3}\rangle$ for the orbital and the dipole parts are completely different, in fact, they do not even have the same sign. Das and collaborators[11] and Lindgren and collaborators[13] have calculated these quantities and find good agreement with experiment. It will be very interesting if similar measurements could be carried out in the $5\,^2D$ level of Cs. Such an attempt has not yet been made, primarily, because of very large magnetic fields required to decouple L and S in this level.

Since these experiments were completed, a new measurement of the hfs of the $5\,^2D$ levels of ^{133}Cs has been performed by Svanberg *et al.*[16] by laser spectroscopy. They find $A(5\,^2D_{5/2}) = -21.2(1)$ MHz, in approximate agreement with our value, and $A(5\,^2D_{3/2}) = +48.6(2)$ MHz.

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[1]C. Tai, W. Happer, and R. Gupta, Phys. Rev. A 12, 736 (1975).

[2]S. Svanberg and P. Tsekeris, Phys. Rev. A 11, 1125 (1975); S. Svanberg, P. Tsekeris, and W. Happer, Phys. Rev. Lett. 30, 817 (1973).

[3]S. Svanberg and G. Belin, J. Phys. B 7, L82 (1974).

[4]W. Hogervorst and S. Svanberg, Phys. Scr. 12, 67 (1975).

[5]G. Belin, L. Holmgren, I. Lindgren, and S. Svanberg, Phys. Scr. 12, 287 (1975).

[6]G. Belin, L. Holmgren, and S. Svanberg, Phys. Scr. 13, 351 (1976).

[7]G. Belin, L. Holmgren, and S. Svanberg, Phys. Scr. 14, 39 (1976).

[8]C. E. Moore, *Atomic Energy Levels*, Natl. Bur. Stand. (U.S.) Circ. No. 467 (U. S. GPO, Washington D. C., 1958).

[9]R. Gupta, S. Chang, C. Tai, and W. Happer, Phys. Rev. Lett. 29, 695 (1972).

[10]H. M. Foley and R. M. Sternheimer, Phys. Lett. 55A, 276 (1976).

[11]T. Lee, J. E. Rogers, T. P. Das, and R. M. Sternheimer, Phys. Rev. A 14, 51 (1976); R. M. Sternheimer, J. E. Rogers, T. Lee, and T. P. Das, *ibid.* 14, 1595 (1976).

[12]L. Holmgren, I. Lindgren, J. Morrison, and A.-M. Mårtensson, Z. Phys. A276, 179 (1976); I. Lindgren, J. Lindgren, and A.-M. Mårtennsson, *ibid.* A279, 113 (1976); Phys. Rev. A 15, 2123 (1977).

[13]T. N. Chang, J. Phys. B 11, L583 (1978).

[14]E. Luc-Koenig, Phys. Rev. A 13, 2114 (1976).

[15]R. M. Sternheimer, J. E. Rodgers, and T. P. Das, Phys. Rev. A 17, 505 (1978).

[16]K. Fredriksson, H. Lundberg, and S. Svanberg, Phys. Rev. A 21, 241 (1980).

[17]R. Gupta, S. Chang, and W. Happer, Phys. Rev. A 6, 529 (1972).

[18]R. Gupta, W. Happer, L. K. Lam, and S. Svanberg, Phys. Rev. A 8, 2792 (1973).

[19]L. K. Lam, Ph.D. thesis, Columbia University, 1975 (unpublished).

[20]R. W. Schmieder, A. Lurio, W. Happer, and A. Khadjavi, Phys. Rev. A 2, 1216 (1970).

[21]H. A. Schüssler, Z. Phys. 182, 289 (1965).

[22]O. S. Heavens, J. Opt. Soc. Am. 51, 1058 (1961).

[23]K. H. Liao, L. K. Lam, R. Gupta, and W. Happer, Phys. Rev. Lett. 32, 1340 (1974).