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Published in:

Physical Review A (Atomic, Molecular and Optical Physics)

DOI:

[10.1103/PhysRevA.21.241](https://doi.org/10.1103/PhysRevA.21.241)

1980

[Link to publication](#)

Citation for published version (APA):

Fredriksson, K., Lundberg, H., & Svanberg, S. (1980). Fine- and hyperfine-structure investigation in the $5^{(2)}D-n^{(2)}F$ series of cesium. *Physical Review A (Atomic, Molecular and Optical Physics)*, 21(1), 241-247. <https://doi.org/10.1103/PhysRevA.21.241>

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Fine- and hyperfine-structure investigation in the 5^2D-n^2F series of cesium

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(Received 6 March 1979)

The authors have studied the fine-structure splittings in the F sequence of states in the cesium atom using high-resolution laser spectroscopy. The experiments were performed with a single-mode laser acting on the transitions from the lowest D state to the different F doublets. The Doppler broadening was reduced to about 20 MHz by means of the techniques of collimated atomic beams. For populating the D state, a further laser acting on the transition from the ground state to the second excited P state was used. About 10% of the P -state atoms decay to the 5^2D levels. All the studied F states were found to be inverted, with splittings as follows: $\Delta E(10^2F) = -873.2(2.0)$ MHz, $\Delta E(11^2F) = -667.2(2.0)$ MHz, $\Delta E(12^2F) = -522.0(3.0)$ MHz, $\Delta E(13^2F) = -416.0(3.0)$ MHz, $\Delta E(14^2F) = -336.8(1.0)$ MHz, $\Delta E(15^2F) = -276.3(1.0)$ MHz, $\Delta E(16^2F) = -229.3(1.0)$ MHz, and $\Delta E(17^2F) = -190.5(2.0)$ MHz. The results are compared with recent theoretical calculations. A further result of the present investigation is the hyperfine structure of the $5^2D_{3/2,5/2}$ states of ^{133}Cs . For the magnetic dipole interaction constant a and the electric quadrupole interaction constant b the following values were obtained: $a(5^2D_{3/2}) = +48.6(2)$ MHz, $b(5^2D_{3/2}) = 0.0(8)$ MHz, $a(5^2D_{5/2}) = -21.2(1)$ MHz, $b(5^2D_{5/2}) = 0.0(1.0)$ MHz. The inversion of the $5^2D_{5/2}$ state is especially noteworthy, but can be explained as due mainly to strong polarization effects.

I. INTRODUCTION

The fine-structure splittings in series of alkali-metal atomic states have attracted considerable interest during the last few years. Strong deviations from hydrogenlike behavior, including inversion of level ordering, were observed quite early for low-lying states of certain level sequences.¹ These perturbations have motivated a detailed experimental and theoretical study of the spin-orbit interaction in sequences of alkali-metal states. As the fine-structure splittings of high-lying states are quite small, especially for non- P states, high-resolution methods are needed. Laser techniques provide both an efficient population of highly excited states as well as a resolution frequently only limited by the radiative width of the excited state. Thus, Doppler-free two-photon spectroscopy,² quantum-beat spectroscopy,³ level crossing,⁴ rf resonance⁵ and microwave-resonance techniques⁶ have been used for studies in sequences of D states especially, but also P , F , and G states. A brief review of the general observations has been given in Refs. 5 and 7. A very interesting result of the experiments is that sequences, whose low-lying members have an inverted energy-level ordering, stay inverted even in the regime of high principal quantum numbers n . Thus, the perturbations causing the inversion scale with n approximately in the same way as does the "normal" interaction. Theoretical calculations for inverted sequences have recently been performed by several groups. Thus, Luc-Koenig,⁸ Holmgren *et al.*,⁹ as well as Sternheimer *et al.*¹⁰ have all been able to explain the inversion using

different approaches.

The lowest members of the F sequence in cesium have been known to be inverted for quite some time. An extrapolation to higher principal quantum numbers of the data given in Ref. 1 for $n=4$ to $n=10$, would indicate a possible crossover from inverted to normal level ordering somewhere above $n=10$. However, the more recent measurements by Eriksson and Wenåker¹¹ extending to $n=12$ make such a transition more doubtful. In order to clearly establish the behavior of this sequence we have performed accurate measurements in the 5^2D-n^2F spectral series. Results for the 12 and 13² F states were given in a previous Letter.¹² In the present paper we report the results of an investigation for all the states ranging from $n=10$ to 17. It is found that the sequence stays inverted and that the fine-structure splittings can be described by an expansion in odd powers of the effective principal quantum number n_{eff} .

Apart from the fine-structure splittings, the hyperfine-structure constants for the $5^2D_{5/2}$ and $5^2D_{3/2}$ states were determined with high precision. As for many other alkali-metal 2D states (see, e.g., Refs. 13 and 14) the $^2D_{5/2}$ level was found to be inverted while the $^2D_{3/2}$ level was normal. From a theoretical point of view the 5^2D state is quite interesting because it is the lowest D state in cesium, which is a heavy atom where relativistic effects can be expected to be important.

Our measurements were performed by observing fluorescence light from a collimated atomic beam, irradiated at right angles with a single-mode dye laser to obtain a narrow linewidth. The

laser was scanned over the substructure of the 5^2D-n^2F transition. In order to create the necessary population of the initial D state, we used the excitation scheme shown in Fig. 1. A multimode cw dye-laser beam, spatially overlapping the single-mode laser beam, was tuned to the $6^2S_{1/2}-7^2P_{3/2}$ transition at 4555 \AA , and in the decay of the $7^2P_{3/2}$ state, about 11% and 1% of the atoms branch into the $5^2D_{5/2}$ and $5^2D_{3/2}$ states, respectively. These states have conveniently long lifetimes around 1 \mu sec to serve as platforms for a subsequent $D-F$ excitation. Figure 1 also includes a diagram showing the substructure of a $5^2D_{5/2}-n^2F_{7/2,5/2}$ transition. In particular, the hyperfine splittings of the D state are given, expressed in the magnetic dipole interaction constant a and the electric quadrupole interaction constant b . The diagram is drawn for the case of inverted D -state hyperfine structure and inverted F -state fine structure. The hyperfine structure of the 2F states is extremely small and is neglected. The positions of the basic energy levels for high-lying members of the 2F sequence have been determined only recently by Mizra and Duley,¹⁵ who with multiphoton spectroscopy studied the levels up to $n = 65$. By time-resolved fluorescence spectroscopy g_J values and natural lifetimes for $^2F_{7/2}$ states

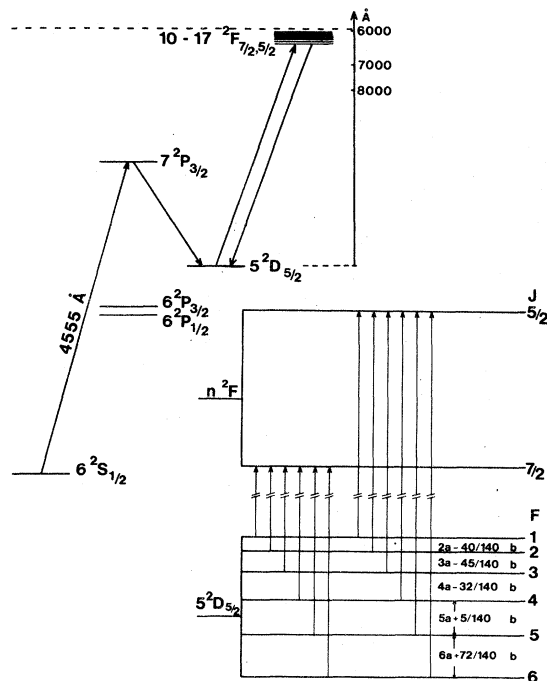


FIG. 1. Excitation scheme used in the studies of cesium 2F levels. A diagram showing the substructure of such a transition for the case of inverted F -state fine structure and inverted D -state hyperfine structure is included.

ranging from $n=11$ to 16 were recently measured in this laboratory¹⁶ using the excitation scheme given in Fig. 1. It is interesting to note that the properties investigated in Refs. 15 and 16 are very normal in contrast to those studied in the present work. In Sec. II of this paper we will present the experimental arrangement used in our studies of the anomalous fine and hyperfine structures. The measurements are described in Sec. III, whereas the results are discussed in the concluding Section IV.

II. EXPERIMENTAL ARRANGEMENT

A schematic diagram of the experimental setup is shown in Fig. 2. Cesium atoms in an atomic beam, formed in a vacuum system, were excited in two steps by the light from two cw dye lasers. The atomic beam was collimated to a ratio of about 1:50. For the first excitation step a coumarin 47 multimode dye laser, pumped by the uv lines of an argon-ion laser, was used. The dye laser had a mode separation of about 300 MHz. In order to reduce the nonuniformity in the excitation caused by the random modedrift, the output mirror of the dye laser was piezoelectrically rapidly scanned. The single-mode dye laser (Coherent Radiation Model 599), used in the second step, was pumped by the light from a krypton-ion laser. With a pump power of 2.5 W, about 100-mW single-mode power could be obtained using the dye rhodamine 6G. Part of the light from the single-mode laser was split off to a monochromator and three Fabry-Pérot interferometers. The monochromator had a resolution of 30 GHz and was used for a rough wavelength setting. Two

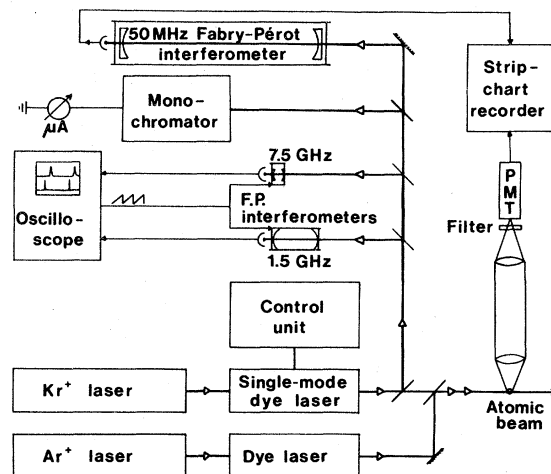


FIG. 2. Schematic diagram of the setup used in the laser-induced fluorescence measurements on a collimated atomic beam.

interferometers were of the spectrum-analyzer type with free spectral ranges of 1.5 and 7.5 GHz, respectively. They were connected to an oscilloscope for convenient monitoring of the single-mode laser operation. The third Fabry-Pérot interferometer, with a 50 MHz free spectral range, was used for accurate calibration of the frequency sweep. This interferometer, with spherical mirrors mounted 75 cm apart in a multipass arrangement, was Invar-stabilized and mounted in an evacuable chamber.

The two laser beams were made to overlap using a dichroic mirror. Since the fluorescent light had to be detected at the single-mode laser wavelength, precautions to reduce stray light were taken. The laser beams were directed into and out of the vacuum system through long tubes with apertures to screen light scattered in the entrance and exit beam windows. The interior of the system was blackened and the fluorescent light was detected by a photomultiplier tube with the interior of the vacuum pumping line as a background. The signal from the photomultiplier was fed to a two-channel strip-chart recorder, also monitoring the 50-MHz Fabry-Pérot fringes.

III. MEASUREMENTS

A. Calibration measurements

The Invar-stabilized Fabry-Pérot interferometer, used to monitor the laser scans, was calibrated in measurements on a sodium atomic beam. The same experimental setup as employed in the fine-structure measurements was utilized. The D_1 line in sodium, consisting of four components due to the hyperfine splittings of the states involved in the transition, was studied by scanning the single-mode dye laser and monitoring the decay back to the ground state. The simultaneously

recorded fringes from the Fabry-Pérot interferometer were thus evaluated in terms of the 1771.7-MHz splitting of the ground state and the 188.6-MHz splitting of the lowest P state.¹⁷ The free spectral range, i.e., the fringe separation, was found to be 49.97(5) MHz from 40 measured curves. It is important to consider interferometer drifts due to temperature changes, both during calibration runs and in subsequent measurements. Such drifts could easily be corrected for in measurements by comparing the positions of a given transition component in subsequent scans, as related to the Fabry-Pérot fringes.

B. Measurements of fine-structure intervals

The excitation scheme of the fine-structure measurements was shown in Fig. 1. The last step, $5^2D_{5/2} \rightarrow n^2F_{7/2,5/2}$, in the excitation process was first effectuated with the dye laser in multimode operation in order to set the monochromator to the proper wavelength, where fluorescence light from the F state was released. Then, using the monochromator for guidance, the laser, operating in a single mode, was adjusted to the wavelength region of interest. By electronically scanning the single-mode laser, the atoms were excited in the individual components of the transition, and the fluorescence light of the subsequent decay from the 2F state was detected. An experimental curve of the $5^2D_{5/2} \rightarrow 10^2F_{7/2,5/2}$ transition is shown in Fig. 3 together with the fringes from the Fabry-Pérot interferometer. Since the first step of the excitation was accomplished with a multimode dye laser, there were rather large fluctuations in the efficiency of the population of the second excited $^2P_{3/2}$ state as a result of the drift of the laser modes. This resulted in a large amplitude noise as well as defects in the

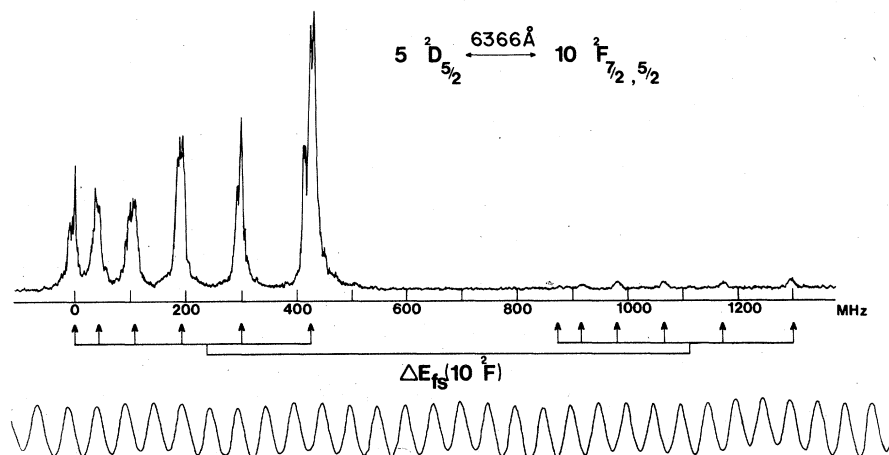


FIG. 3. Experimental curve recorded for the $5^2D_{5/2} \rightarrow 10^2F_{7/2,5/2}$ transition together with calibration fringes from the 50-MHz Fabry-Pérot interferometer.

relative intensities of the individual components of the studied transitions. Thus, the expected 20:1 ratio of the intensities of the two hyperfine-structure patterns is not fully verified in individual recordings. From the experimental curve it is obvious that the fine-structure of the 2F state is inverted, since the low-intensity lines appear at higher frequencies. Further, the hyperfine structure of the $5^2D_{5/2}$ state is shown to be inverted.

Since the fine-structure splittings in the lower 2F states are larger than the total hyperfine splitting of the $5^2D_{5/2}$ state, the experimental curves for these states will consist of a twice-repeated line pattern, as is shown in Fig. 3. For states with n larger than 12 this is not the case. The two hyperfine line patterns will then be overlapping. Such a case is given in Fig. 4, where the $5^2D_{5/2}$ - $15^2F_{7/2,5/2}$ transition is shown. Again, the inversion of the 2F state is evident, and the same result was obtained for all the other studied states. In Table I the fine-structure splittings of the $n=10$ to 17^2F states are given together with the wavelengths used for the excitation and detection.

C. Hyperfine structure of the $5^2D_{5/2,3/2}$ states

As has been pointed out earlier in this paper, the hyperfine structure of the $5^2D_{5/2}$ state is found to be inverted, i.e., the magnetic-dipole interaction constant a is negative. Since every single fine-structure measurement in this work also contains information of the hyperfine structure of this state, as is seen in Figs. 3 and 4, there is

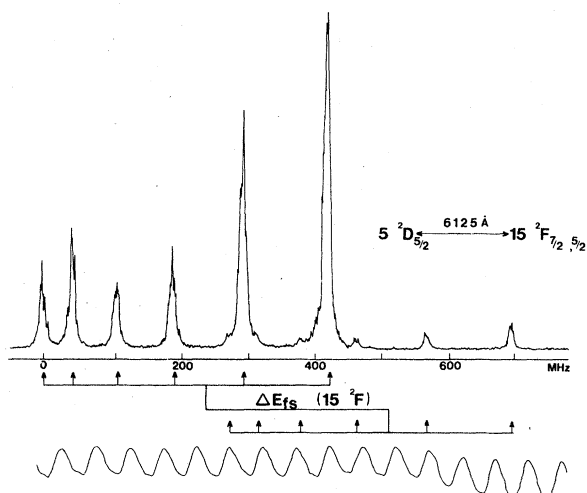


FIG. 4. Experimental curve recorded for the $5^2D_{5/2}$ - $15^2F_{7/2,5/2}$ transition. Because of the small value of the F -state fine-structure splitting, the two hyperfine-structure patterns partially overlap.

TABLE I. Fine-structure splittings for the 10 to 17^2F states determined in the present work and wavelengths of the lines used for excitation and detection.

n^2F state	Fine-structure splitting (MHz)	Excitation and detection line (nm)
10	-873.2(2.0)	636.6
11	-667.2(2.0)	628.9
12	-522.0(3.0)	623.1
13	-416.0(3.0)	618.8
14	-336.8(1.0)	615.3
15	-276.3(1.0)	612.5
16	-229.3(1.0)	610.3
17	-190.5(2.0)	608.5

a very large amount of data for the determination of the hyperfine-structure constants. From 90 registration curves the magnetic-dipole constant a and the electric-quadrupole constant b are evaluated:

$$a(5^2D_{5/2}) = -21.2(1) \text{ MHz},$$

$$b(5^2D_{5/2}) = 0.0(1.0) \text{ MHz}.$$

Similar investigations of the $5^2D_{3/2}$ -state hyperfine structure were performed. The transition $5^2D_{3/2}$ - $13^2F_{5/2}$ was then studied. In Fig. 5 an experimental curve displaying the hyperfine components of the transition is shown. In this case only the ${}^2F_{5/2}$ state is reached because of the selection rule, and the structure is not repeated as it was in the earlier cases. Also included in the figure is a diagram showing the studied sublevel structure. For the magnetic-dipole and electric-quadrupole interaction constants we found

$$a(5^2D_{3/2}) = +48.6(2) \text{ MHz},$$

$$b(5^2D_{3/2}) = 0.0(8) \text{ MHz}.$$

The zero result for the quadrupole-coupling con-

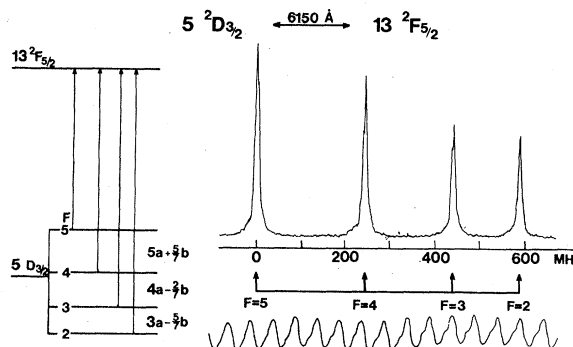


FIG. 5. Recording of the $5^2D_{3/2}$ - $13^2F_{5/2}$ transition showing the hyperfine structure of the $5^2D_{3/2}$ state, which is schematically given in the sublevel diagram.

stants for both states reflects the small value of the nuclear electric quadrupole moment for ^{133}Cs , 3 mb.

In the error limits given for the $5^2D_{5/2,3/2}$ hyperfine-structure constants and the 2F fine-structure splittings (Table I), statistical errors and possible errors in the free spectral range of the Fabry-Pérot interferometer as well as reasonable remaining systematic errors are included.

IV. DISCUSSION

The measurements reported in this work yield accurate fine-structure values for several highly excited 2F states in cesium and clearly show that this sequence stays inverted in the regime of high principal quantum numbers. In Table II experimental and theoretical values for this sequence are given. The fine-structure values are plotted in Fig. 6 versus the effective principal quantum number n_{eff} . (For the F states of Cs the effective principal quantum numbers are very close to integers.) The theoretical results for the 4, 5, and 6^2F states given by Sternheimer *et al.*¹⁰ are too low to be included in the figure. An attempt to smoothly join our data points with those for the lower states given by Eriksson and Wenåker¹¹ is met with difficulties, which appear particularly clear if a log-log plot is used. Evidently the splittings given in Ref. 11 for the $n=7, 8,$ and 9 states are too small. In order to describe the fine-structure splittings, an expansion in odd powers of n_{eff} of the type

$$\Delta E = A/n_{\text{eff}}^3 + B/n_{\text{eff}}^5 + C/n_{\text{eff}}^7$$

can be used. For the D sequences of sodium³ and rubidium² such expansions, using only two terms

($C=0$), have been very successful. However, for the F sequence of cesium a considerably better fit was obtained by also including an n_{eff}^7 term. Using the rather well-determined values for the $n=4, 5,$ and 6 states and the precision values for the 10 to 17 states, we obtain

$$A = -979.6 \text{ GHz}, B = 12\,220 \text{ GHz}, C = -33\,760 \text{ GHz}.$$

The corresponding curve is drawn as a full line in Fig. 6. By using the found expansion, it is possible to predict the most likely values for the fine-structure of the $n=7, 8,$ and 9 states: $-2190,$ $-1570,$ and -1150 MHz, respectively.

The theoretical results given by Luc-Koenig⁸ well describe the inverted F sequence, with an overestimate of the splittings of slightly more than 20%. The values calculated for the lowest sequence members by Sternheimer *et al.*¹⁰ are, while correctly reproducing the inversion, almost a factor of 2 too large in magnitude. It is interesting to note that, while a first-order relativistic central-field approach is taken in Ref. 8, nonrelativistic wave functions are used in Ref. 10, and the inversion then is explained in terms of configuration interaction. An interesting discussion on the interrelation between these two approaches can be found in Ref. 8.

The dipole-coupling constants a , describing the magnetic hyperfine interaction of the $5^2D_{5/2}$ and $5^2D_{3/2}$ states, were found to be negative and positive, respectively. The value of $a(5^2D_{5/2})$ had previously been determined by Lam¹⁸ using rf spectroscopy. He obtained the value $-22.1(5)$ MHz, which is slightly larger and less accurate than the value obtained in the present work. The inversion of alkali-metal atom $^2D_{5/2}$

TABLE II. Experimental and theoretical values for 2F -state fine-structure splittings.

2F state n	Experimental values (MHz)		Theoretical values (MHz)	
	This work	Eriksson and Wenåker ^a	Luc-Koenig ^b	Sternheimer <i>et al.</i> ^c
4		-5435	-6711	-10 730
5		-4395	-5480	-10 268
6		-3109	-3864	-6 260
7		-2075	-2707	
8		-1391	-1934	
9		-1019	-1417	
10	-873.2(2.0)	-714	-1064	
11	-667.2(2.0)	-737	-817	
12	-522.0(3.0) ^d	-405	-639	
13	-416.0(3.0) ^d			
14	-336.8(1.0)			
15	-276.3(1.0)			
16	-229.3(1.0)			
17	-190.5(2.0)			

^a Reference 11.

^b Reference 8.

^c Reference 10.

^d Values given already in Ref. 12.

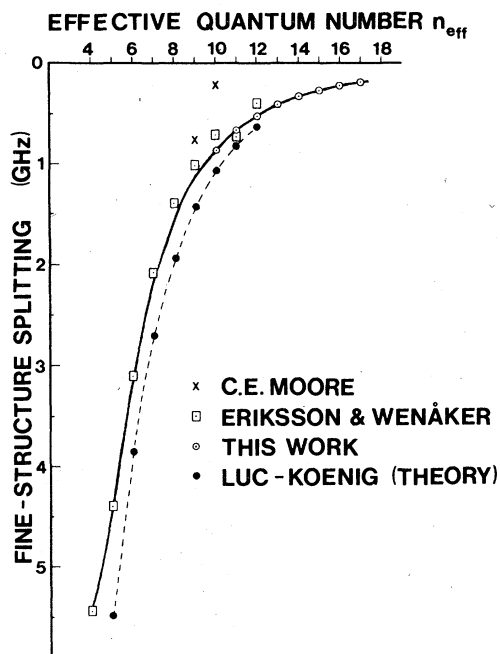


FIG. 6. Experimental and theoretical values for the fine-structure splittings in the F sequence of cesium plotted vs the principal quantum number. The full line represents the formula obtained for describing the F -state fine-structure splittings.

states has been successfully explained by Lindgren and co-workers.¹⁹ By using many-body perturba-

tion theory a very good agreement with experiment has been obtained, especially for the $4^2D_{5/2,3/2}$ state of rubidium.²⁰ While a detailed inclusion of correlation effects is necessary for obtaining accurate theoretical values, the $^2D_{5/2}$ -state inversions are basically due to polarization effects, induced in the core owing to the presence of the valence electron. Lindgren and co-workers have shown that such effects can be included to infinite order²¹ and in the context of a previous hyperfine-structure work on cesium²² their computer program could be used to generate theoretical values for the dipole constant for the $5^2D_{5/2}$ and $5^2D_{3/2}$ states also. The theoretical values of -21.4 and 23.3 MHz, respectively, correctly reproduce the signs of the coupling constants. However, the very good agreement in the case of the $5^2D_{5/2}$ state must be regarded as fortuitous. Apart from correlation effects, relativistic effects should also be important for a heavy atom like cesium. Presently, a relativistic treatment of polarization effects is being developed at this department,²³ and the 5^2D states will be good test cases for the potential of this refined approach.

ACKNOWLEDGMENTS

The authors gratefully acknowledge valuable discussions with Professor I. Lindgren. This work was supported by the Swedish Natural Science Research Council.

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