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Hyperfine-Structure Studies of Highly Excited D and F Levels in Alkali Atoms Using a cw Tunable Dye Laser*

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Highly excited S, D, and F levels in cesium and rubidium have been populated utilizing a two-step optical excitation method. In the first step an rf lamp is used to transfer atoms into the first P states, and the subsequent excitation is performed with the tunable radiation of a cw dye laser. We give results for the hyperfine structure of several D and F states in Cs^{133} and Rb^{87}, obtained in level-crossing and optical double resonance experiments.

Until recently, accurate hyperfine structure studies of excited states in alkali atoms have been confined to the sequences of ^2P levels, which are readily available for observation through direct excitation with resonance radiation. Detailed studies of P levels have revealed several effects which cannot be explained by a simple, one-electron picture of the alkali atoms. These include magnetic core polarization, electric-quadrupole shielding, and possible deviations from expected Landé g_J factors.[1] Clearly, a corresponding knowledge of the properties of non-P states is of great interest for the understanding of the various atomic interactions.

It is well known that the D-state fine-structure intervals are often anomalously small and even inverted, and recent experimental work has shown that similar, large anomalies occur in the D-state hyperfine structure. Data on D-state hyperfine structure are still quite limited, and F-state data are nonexistent. The scarcity of data can be traced to inadequacies of existing experimental methods. Archambault *et al.*[2] have made rough estimates of the hyperfine structure of the $5^2D_{5/2}$ state in Na^{23} and the $9^2D_{5/2}$ state in Cs^{133}, which were produced by electron bombardment. With the recently introduced cascade decoupling[3] and cascade rf spectroscopy methods[4] the Columbia group has investigated the hyperfine structure of several S and D levels in the alkali atoms.[5] In these experiments the states are produced by cascading from a P level, excited by a conventional rf lamp. However, for reasons of intensity, this method is limited to comparatively low-lying S and D levels as the absorption oscillator strengths in the S-P transition sequence decrease very rapidly with increasing P-state energy, and conventional lamps are not very efficient sources for the higher resonance lines. To get access to more highly excited non-P levels we have utilized a two-step excitation method,[6] taking advantage of recent advances in the technology of tunable lasers. In the first step the strong D_1 and D_2 lines from a powerful rf lamp are used to transfer atoms from the ground S state to the first excited P states. In the second step, the intense tunable radiation from a cw dye laser, operating with rhodamine 6G, is used to excite atoms from a P level to a highly excited S or D level. Absorption of light by atoms in the $5^2P_{3/2}$ level of

Rb has earlier been demonstrated by Bradley *et al.*,[7] utilizing high-power pulsed dye lasers. Further, a two-step excitation of the 3^2D term in lithium has been used in a fine-structure investigation by Smith and Eck.[8] In this case the light for the excitation was provided in a discharge in the studied alkali vapor. However, as in the electron excitation experiments of Archambault *et al.*,[2] it was hard to achieve a very well defined excitation situation. The excitation method described in this Letter is particularly useful since it can be used in conjunction with conventional optical double resonance (ODR) and level crossing (LC) methods to investigate the hyperfine structures and fine structures of a large number of excited states. As an example, we discuss some of our recent measurements for a number of excited D and F states in rubidium and cesium.

In performing experiments on the highly excited alkali states it is very valuable to have reliable estimates on absorption oscillator strengths, natural lifetimes, and branching ratios. We have made extensive computer calculations of these quantities utilizing the Coulomb approximation method of Bates and Damgaard.[9]

Our experimental arrangement is based on the apparatus described in Refs. 4 and 5. A powerful rf lamp was constructed for producing the light for the first excitation step. The beam of D_1 and D_2 light entered the resonance cell in the direction of the external magnetic field, which is also the detection direction. The laser beam, which has a diameter of about 3 mm, passes through the resonance cell at right angles to the field direction. We use a Spectra Physics model 370 tunable dye laser, which is pumped by the 5145-Å line of a Spectra Physics model 165 argon ion laser. In multimode operation the dye laser gives up to 300 mW output for an input power of 2 W. With rhodamine 6G as a dye, the usable wavelength region extends from about 5550 to about 6300 Å. A bandwidth of about 0.3 Å is obtained. Fluorescent light released in the decay of the highly excited state is detected by an RCA C31000F photomultiplier. The rf field required in the ODR measurements is produced in a cavity, tuned to about 147 MHz. Lock-in detection is used in the experiments and the signals are stored in a PDP-8/S computer, which also controls the linear magnetic field sweep.

In the two-step excitation process it is possible to take advantage of radiation trapping of the D_1 and D_2 resonance lines. In the multiple scattering process, an incoming photon can excite several ground-state atoms into a ^2P state so that more P-state atoms will be available for laser excitation. In this way we have achieved adequate populations for ODR and LC spectroscopy of the $9, 10, 11^2S_{1/2}$ and the $8, 9, 10^2D$ levels in Cs and the $8, 9^2S$ and $6, 7^2D$ levels in Rb. The interaction region between the atoms and the two light beams is about 0.5 cm^3 and contains at the usual operation temperature (100°C for Cs, 120°C for Rb) about 10^{13} atoms. The number of detected photons per second in a solid angle of about 0.15 sr is 10^6–10^8, depending on which particular state is studied and what detection line is used.

We have used the LC method to measure the hyperfine structure of the 8 and $9^2D_{3/2}$ states in Cs^{133}. Because of the multiple scattering of the D lines the different sublevels of the $6^2P_{3/2}$ state have approximately equal populations. With the tunable laser, σ transitions to the $^2D_{3/2}$ state are induced and the decay down to the $6^2P_{1/2}$ level is observed. To avoid direct leakage of the detection line from the rf lamp, a Schott RG10 color glass filter is used in the exciting beam to block all lines except the infrared ones. An interference filter with a half-width of 50 Å is used to isolate the detection line from the stray light from the strong laser beam. A rotating linear polarizer in the detected beam is used for lock-in detection of the LC signals. In Fig. 1 examples of the measured LC curves are shown. In

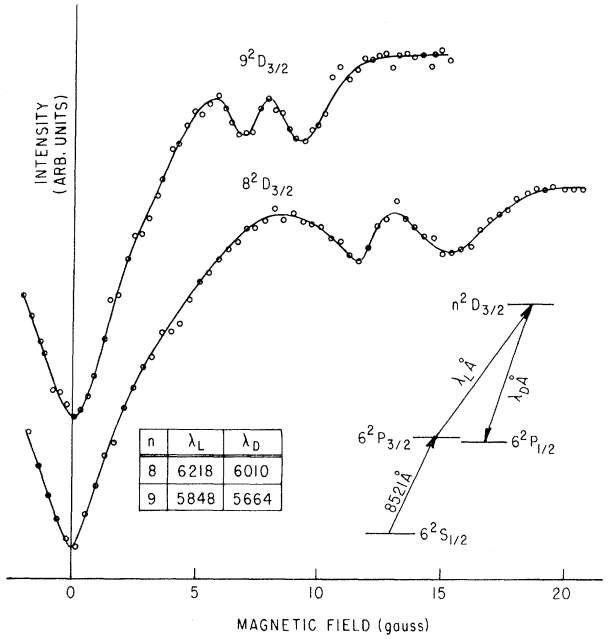

FIG. 1. Level-crossing signals for the 8 and $9^2D_{3/2}$ states in Cs^{133}. The sampling time for each of the curves is about 1 h.

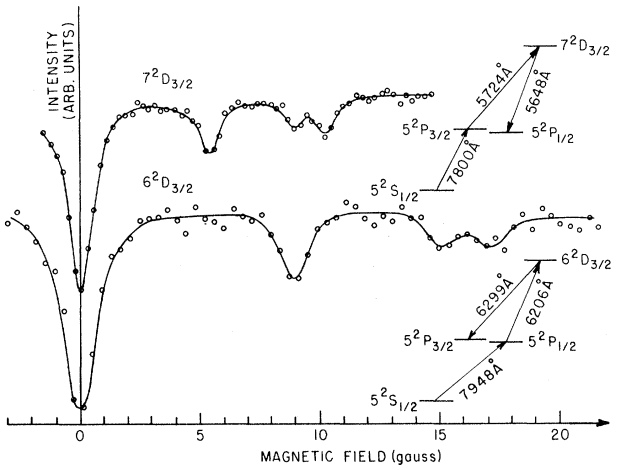

FIG. 2. Level crossing signals for the 6 and $7^2D_{3/2}$ states in Rb^{87}. Total sampling time for each curve, 1.5 h.

both $^2D_{3/2}$ states one resolved and two nonresolved crossing signals are obtained. From the positions of the crossings we obtain for the dipole interaction constant a

$$|a(8^2D_{3/2}\ Cs^{133})| = 3.98(12)\ \text{MHz}$$

and

$$|a(9^2D_{3/2}\ Cs^{133})| = 2.35(7)\ \text{MHz}.$$

The quadrupole interaction constant b is in both cases small, as the quadrupole moment of Cs^{133} is only -3 mb. In order to determine the signs of the coupling constants additional measurements have to be performed.[4]

Corresponding LC measurements were also made in the 6 and $7^2D_{3/2}$ states of Rb^{87}. In Fig. 2 experimental curves are shown. From the positions of the resolved crossings we obtain

$$|a(6^2D_{3/2}\ Rb^{87})| = 7.72(20)\ \text{MHz},$$

$$|b(6^2D_{3/2}\ Rb^{87})| = 0.6(4)\ \text{MHz}, \quad b/a > 0,$$

and

$$|a(7^2D_{3/2}\ Rb^{87})| = 4.55(15)\ \text{MHz},$$

$$|b(7^2D_{3/2}\ Rb^{87})| = 0.36(18)\ \text{MHz}, \quad b/a > 0.$$

We have also performed ODR measurements in the Paschen-Back region of the 8 and $9^2D_{3/2}$ states in Cs^{133}. The same geometry was used and the rf was 100% square-wave modulated for lock-in detection. Eight signals of equal intensity are expected around the center of gravity, corresponding to $g_J = 0.8$. The structure obtained is in both states unresolved and agrees closely with the theoretical structure calculated from the LC results.

As the transition between a $^2D_{5/2}$ state and a $^2P_{1/2}$ state is forbidden and it is not possible to use the laser transition for the detection due to the inevitable stray light, a different technique was used for measurements in the highly excited $^2D_{5/2}$ states in Cs. These states decay with a branching ratio of about 15% into the second excited $^2P_{3/2}$ level, and the wavelength (4555 Å for cesium) for the transition from this state to the ground state can be well isolated from both exciting lines. Schott BG3 and BG18 color glass filters were useful in further suppressing the background light. The LC method is not very useful in this case as the coherence at the crossing points is strongly degraded in the passage through the second state. Thus we used the ODR technique. The 8, 9, and $10^2D_{5/2}$ levels in Cs^{133} all turn out to have a very small hyperfine structure. In the Paschen-Back region an unresolved signal structure, consisting of eight closely spaced signals, is obtained. The width of a single component is calculated from the theoretically estimated natural lifetimes. In extrapolations to zero rf power we find the signal structure half-widths 4.5(5), 2.7(5), and 2.3(5) G. From these results only rough values for the dipole coupling constants can be obtained:

$$|a(8^2D_{5/2}\ Cs^{133})| = 0.9(4)\ \text{MHz},$$

$$|a(9^2D_{5/2}\ Cs^{133})| = 0.5(2)\ \text{MHz},$$

and

$$|a(10^2D_{5/2}\ Cs^{133})| = 0.4(2)\ \text{MHz}.$$

In Cs the ^2F levels from 4^2F to 7^2F can be populated by cascading from the ^2D levels excited in these experiments. The $8^2D_{5/2}$ and $8^2D_{3/2}$ levels have a 2% branching ratio into the $5^2F_{7/2}$ and the $5^2F_{5/2}$ levels, respectively. The decay of the F states to the $5^2D_{5/2}$ and $5^2D_{3/2}$ levels can be observed at 8079 and 8016 Å. We have observed unresolved ODR signals in these F states as shown in Fig. 3. The 5^2F doublet is inverted with a fine-structure splitting at only about 4410 MHz. Thus even for zero hyperfine structure, the ODR signals at an rf frequency of 147 MHz would be spread over 0.6 and 0.9 G in the $5^2F_{5/2}$ levels, respectively. The estimated individual signal full half-width is 2.7 and 2.8 G, whereas the measured half-width is about 4.0 G in the $5^2F_{7/2}$ as well as in the $5^2F_{5/2}$ states. From these values, rough upper limits for the hyperfine structure

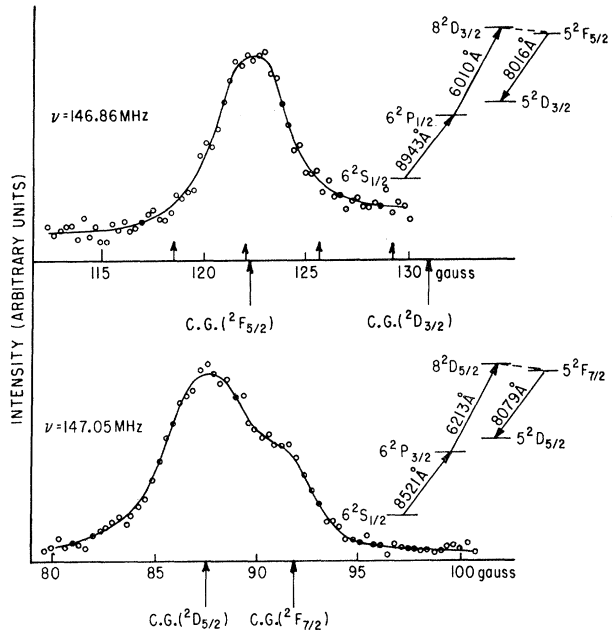

FIG. 3. Optical double resonance curves with signals from the $5^2F_{7/2}$ and $5^2F_{5/2}$ levels in Cs^{133}. The center of gravity (C.G.) for the F states and the position of the D resonance also occurring in these experiments are indicated. Sampling time for each of the curves is about 1 h.

constants are estimated:

$$|a(5^2F_{7/2}\ Cs^{133})| < 1.0 \text{ MHz}$$

and

$$|a(5^2F_{5/2}\ Cs^{133})| < 0.7 \text{ MHz}.$$

Measurements of the hyperfine structure of other highly excited alkali states and experiments for determining the signs of the coupling constants in the states studied in this work are in progress.

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Determination of the He^4-He^4 Repulsive Potential up to 0.14 eV by Inversion of High-Resolution Total–Cross-Section Measurements

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High-resolution measurements of the He^4-He^4 total scattering cross section are presented for reduced collision energies between 0.2 and 200 meV. Clear evidence for the Ramsauer-Townsend effect is observed and thirteen backward glory extrema are resolved. From these extrema we derive the energy dependence of the s phase shift. Applying the semiclassical inversion method proposed by Miller we then compute the repulsive potential up to 0.14 eV.

Knowledge of the interaction between two He^4 atoms in the ground state has been greatly increased in recent years. Relative differential scattering cross-section measurements at fixed energies[1-3] give sufficient information on the interaction potential only at energies which are not