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LETTER TO THE EDITOR

Determination of hyperfine structure and $g_J$ factors in the sequences of $^2D$ states in alkali atoms using a tunable dye laser

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Abstract. We have extended recently reported hyperfine structure studies of $^2D$ states in alkali atoms to several new states. The $^2D$ states were populated in a two-step excitation scheme, involving a conventional RF lamp and a cw tunable dye laser. Using level crossing and optical double resonance techniques we have determined the hyperfine structure of the $8^2D_{3/2,5/2}$ states in $^{87}$Rb and the $11, 12, 13, 14^2D_{3/2,5/2}$ states in $^{133}$Cs. Further, $g_J$ factors for several of these states were determined.

The alkali atoms have a comparatively simple structure and are particularly suitable for spectroscopic investigations as well as for theoretical calculations. These atoms provide natural test cases for modern many-body perturbation calculations. The advent of such detailed computational schemes has renewed the interest for the experimental investigation of the fine and hyperfine structure of excited states. In the alkali atoms only the sequences of $^2P$ states are directly optically accessible in a single-step excitation process of the electric dipole type. Consequently, for a long time only these excited alkali states were studied with high resolution resonance methods. Excited non-$P$ states can be populated in the cascade decay of highly excited $^2P$ levels. Several lower $^2S$ and $^2D$ states have recently been investigated at Columbia Radiation Laboratory using cascade decoupling (Gupta et al 1972a) and cascade RF spectroscopy techniques (Gupta et al 1972b, Gupta et al 1973).

In order to get access to more highly excited non-$P$ states, the feasibility of a two-step excitation scheme, taking advantage of the recent developments in the technology of tunable dye lasers, has been demonstrated (Svanberg et al 1973a, b). In a first excitation step the strong $D_1$ or $D_2$ line from a powerful RF discharge lamp is used to populate one of the levels in the first excited $P$ doublet. From this level transitions are induced to an excited $S$ or $D$ level using a cw dye laser, tuned to the corresponding transition. The hyperfine structure (HFS) of several $^2D$ levels excited in this way, could be measured by the level crossing (LC) and optical double resonance (ODR) methods (Svanberg 1973, Svanberg et al 1974). We have now extended these measurements to several other $^2D$ states in Rb and Cs. These states have become accessible using new dyes, extending the wavelength region covered by rhodamine 6G, which was used in the previous investigations. In this way we have now obtained HFS information on the $8^2D_{3/2,5/2}$ states in $^{87}$Rb and on the $11, 12, 13, 14^2D_{3/2,5/2}$ states in $^{133}$Cs. Furthermore, the Landé $g_J$ factor has been determined for several of these states.

Our experimental arrangement is based on the set-up described by Svanberg and Rydberg (1969) and is similar to the arrangement used in the earlier laser investigations (Svanberg et al 1973b, 1974). An alkali vapour scattering cell is placed in a stabilized...
oven (70–100 °C) and is surrounded by Helmholtz coil systems, generating a well defined external magnetic field. The strong $D_1$ and $D_2$ lines from an alkali RF lamp are isolated by a Schott colour glass filter and the light is incident upon the scattering cell in the direction of the magnetic field. In this way the first excited $^2P$ levels are populated in a first excitation step. In the second step atoms are transferred to a highly excited $^2D$ level by means of an appropriately tuned dye laser, the intense light beam of which traverses the scattering cell at right angles to the external field. We used a Coherent Radiation Model 490 jet stream dye laser, pumped by a Coherent Radiation Model CR8 argon ion laser. In the present measurements the dye rhodamine 110, dissolved in ethylene glycol, was used. The multi-mode bandwidth of the dye laser was about 0.5 Å. Fluorescent light following the two-step excitation was detected in the direction of the magnetic field. A Bausch and Lomb 500 mm grating monochromator with the slit parallel to the laser beam was used and the wavelength selected light was detected with a photomultiplier tube. The lock-in technique was employed for the registration of LC and ODR signals. In the LC measurements a rotating polarizer was used in the detected beam, whereas the RF field, generated in a two-turn coil around the scattering cell, was modulated in the ODR experiments. The lock-in output signal was stored in a Laben Correlation multichannel analyser, which also controlled a repetetive magnetic field sweep.

In the sequence of $^2D$ states in $^{133}$Cs, the rhodamines 6G and 110 cover the members with principal quantum number $n$, ranging from 8 to 14. Out of these the states

![Figure 1](image-url)  
**Figure 1.** Level crossing signals for the 11, 12, 13, and 14 $^2D_{9/2}$ states in $^{133}$Cs. The sampling time for each of the curves is 0.5–1h.
with \( n = 8, 9, 10 \) were investigated in the previous works, leaving the levels with \( n = 11, 12, 13, 14 \) to be studied here. For \(^{87}\text{Rb}\) the \( 8 \ ^2\text{D} \) doublet is investigated in this work extending the previous experiments on the \( 6 \) and \( 7 \ ^2\text{D} \) states. All the \( ^2\text{D}_{\frac{3}{2}} \) states were studied with the LC method. In \(^{133}\text{Cs} \) \((I = \frac{3}{2})\) as well as in \(^{87}\text{Rb} \) \((I = \frac{5}{2})\) there are three detectable level crossings besides the zero field crossing (Hanle effect). However, in \(^{133}\text{Cs} \) the second and third crossings are strongly overlapping, resulting in an unresolved signal structure. Experimental curves are shown in figures 1 and 2. Here the excitation and detection lines are also indicated. For an accurate evaluation of the HFS, the regions around the individual crossing signals were covered with short magnetic field sweeps. Care was taken to eliminate influences of asymmetries, time

![Figure 2. Optical double resonance curve for the \( 8 \ ^2\text{D}_{\frac{3}{2}} \) state, and level crossing curve for the \( 8 \ ^2\text{D}_{\frac{3}{2}} \) state of \(^{87}\text{Rb} \).](image)

| Atom | State | \(|a|\) MHz | \(|b|\) MHz | \(g_J\) |
|------|-------|-------------|-------------|--------|
| \(^{87}\text{Rb}\) | \( ^8\text{D}_{\frac{3}{2}} \) | 2.85(3) | 0.16(5) | 1.997(15) |
| \(^{87}\text{Rb}\) | \( ^8\text{D}_{\frac{3}{2}} \) | 1.2(2) | \(b/a > 0\) | |
| \(^{133}\text{Cs} \) | \( ^{13}\text{D}_{\frac{3}{2}} \) | 1.055(15) | | |
| \(^{133}\text{Cs} \) | \( ^{11}\text{D}_{\frac{3}{2}} \) | 0.24(6) | | 1.994(10) |
| \(^{133}\text{Cs} \) | \( ^{12}\text{D}_{\frac{3}{2}} \) | 0.758(12) | | 0.8001(10) |
| \(^{133}\text{Cs} \) | \( ^{12}\text{D}_{\frac{3}{2}} \) | 0.19(5) | | 1.996(10) |
| \(^{133}\text{Cs} \) | \( ^{13}\text{D}_{\frac{3}{2}} \) | 0.556(8) | | |
| \(^{133}\text{Cs} \) | \( ^{13}\text{D}_{\frac{3}{2}} \) | 0.14(4) | | 1.998(10) |
| \(^{133}\text{Cs} \) | \( ^{14}\text{D}_{\frac{3}{2}} \) | 0.425(15) | | |

Table 1. Absolute values of hyperfine interaction constants, and \(g_J\) values for \(^2\text{D}\) states in \(^{87}\text{Rb}\) and \(^{133}\text{Cs}\)

constants, and the earth's field. Thus the magnetic dipole interaction constant $a$, and for $^{87}\text{Rb}$ also the electric quadrupole interaction constant $b$, could be determined. The results are shown in table 1.

The $^{2}\text{D}_{5/2}$ states were all investigated with ODR. In all states the HFS turned out to be very small, and at a resonance frequency of about 110 MHz the Paschen–Back region for the HFS is well reached. Thus, $2I+1$ resonances of equal width and intensity are expected symmetrically spread around a centre of gravity, corresponding to the particular $g_J$ value of the state. In all cases only an unresolved structure was obtained. The halfwidth of this structure was determined, extrapolated to zero RF amplitude and low vapour pressure. From the measured halfwidth and theoretically estimated widths for the individual signal contributions, only very rough values for the $a$ factor could be obtained. From the position of the centre of gravity the $g_J$ value was determined. The magnetic field calibration, especially critical in these last measurements, was performed using optical pumping. An experimental ODR curve is included in figure 2. ODR experiments were also performed in the $^{12}\text{D}_{3/2}$ state for checking the $g_J$ value. The results of the ODR measurements are recorded in table 1. It should be noted, that these experiments only give the absolute values of the coupling constants. The signs, which are not necessarily positive due to the influence of core polarization (Gupta et al 1972b), will be determined in subsequent experiments. The $g_J$ values of the studied $^{2}\text{D}_{5/2}$ and $^{2}\text{D}_{3/2}$ states are found to agree well with the values predicted by the Landé formula: 1·2005 and 0·7995, respectively.

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