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Investigation of the fine structure in the n^2D sequence of sodium using level-crossing spectroscopy

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Abstract. The fine-structure splittings in the n^2D sequence of sodium were measured for $n = 4-9$. An accuracy of about 0.1% was obtained in level-crossing measurements on an atomic beam. The D states were populated by step-wise excitation, using an RF lamp and a cw dye laser. A method for measuring the sign of the spin-orbit coupling constant is demonstrated. With the present measurements, the highly anomalous n^2D sequence in sodium has now been studied for $n = 3-16$.

1. Introduction

The alkali atoms, which have only one electron outside closed shells, are likely to behave as hydrogen-like systems; indeed, many properties like the general energy-level arrangement, the radiative lifetimes and Stark-interaction constants, can be quite well understood using hydrogen-like wavefunctions. However, some striking anomalies in the fine-structure (FS) splittings were observed early, e.g. the low-lying D states in sodium, the lowest D state in rubidium and the F states in rubidium and caesium are inverted (Moore 1971). This is totally inexplicable in a hydrogen-like picture. Early attempts to explain the inversion of alkali-like doublets were made by Phillips (1933) and by Araki (1939). A detailed theory for the spin-orbit interaction was developed by Blume and Watson (1962, 1963) and by Blume *et al* (1964). Using this theory, and applying many-body perturbation techniques, the anomalous fine structure could be studied in more detail at this laboratory (Holmgren *et al* 1976). Theoretical calculations on fine-structure intervals in alkali atoms have recently also been performed by Foley and Sternheimer (1976) and by Luc-Koenig (1976). Of special interest are the inverted D states of sodium, since light atoms are especially suitable for calculations. The inversion is found to be due to strong polarization effects in non-s-type subshells of the electron core. The FS splittings in the n^2D sequence of sodium are small, and because of this only the $n = 3-6$ members had until recently been measured by interferometry (Meissner and Luft 1937). With the introduction of the two-photon spectroscopy method, the 4^2D state was extensively studied (Pritchard *et al* 1974, Hänsch *et al* 1974, Bjorkholm and Liao 1974, Biraben *et al* 1974). With similar techniques Levenson and Salour (1974) extended the measurements to the 5^2D state. In a recent letter we demonstrated that a considerable increase in accuracy could be obtained for the 4^2D state by using the level-crossing (LC) technique (Fredriksson and Svanberg 1975). In the present work we have

extended the LC measurements to the 5, 6, 7, 8 and 9²D states. The D states were populated by step-wise excitations using an RF lamp and a CW dye laser. We have also tested a method for measuring the signs of the spin-orbit coupling constants using high-resolution laser spectroscopy.

Recently, the FS splittings for the 9–16²D levels of sodium were measured by quantum-beat spectroscopy (Fabre *et al* 1975). With the present measurements, the anomalous n^2 D sequence has been studied for $n = 3$ –16. The inversion has been experimentally established for $n = 3, 4, 5, 6$ and 10, 11, 12 (Fabre and Haroche 1975).

2. Theoretical considerations

In the present work we have determined the FS intervals by measuring the positions of level crossings induced by an external magnetic field. Thus it is necessary to relate the measured quantities to the splitting between the FS levels in zero magnetic field. In the presence of a spin-orbit interaction and an external magnetic field B , the energy-level diagram of an atomic state is given by

$$H = \frac{\delta W}{l + \frac{1}{2}} l \cdot s + \mu_B B (g_s s + g_l l). \quad (1)$$

Here δW is the spin-orbit splitting, μ_B is the Bohr magneton, and g_s and g_l are the Landé factors corresponding to the angular momentum s for spin and l for orbital motion.

The solution of the secular equation for the matrix of this Hamiltonian gives rise to the Breit-Rabi formula for the energy $E(J, m_J)$ of a sublevel, characterized by the quantum numbers J and m_J (Ramsey 1963):

$$E(J, m_J) = -\frac{\delta W}{2(2l + 1)} + g_l \mu_B B m_J \pm \frac{\delta W}{2} \left(1 + \frac{4m_J}{2l + 1} X + X^2 \right)^{1/2} \quad (2)$$

where

$$X = (g_s - g_l) \frac{\mu_B B}{\delta W}.$$

The plus sign corresponds to the larger of the two J values and the minus sign to the smaller one. In figure 1 the corresponding energy-level diagram for an inverted D state is shown. Both energy and field are expressed in units depending on δW . In this way a diagram, valid irrespective of the value of δW , is obtained. In the figure, the $\Delta m_J = 2$ level crossings are indicated. The magnetic field values for these crossings can be calculated using equations (2). We get

$$\begin{aligned} B(\frac{5}{2}, \frac{5}{2}; \frac{3}{2}, \frac{3}{2}) &= \frac{4g_l + g_s}{5g_l(g_l + g_s)} \frac{\delta W}{\mu_B} \\ B(\frac{5}{2}, \frac{3}{2}; \frac{3}{2}, -\frac{1}{2}) &= \frac{[100g_l^4 + (g_s - g_l)^4 - 28g_l^2(g_s - g_l)^2]^{1/2} - (g_s - g_l)g_l}{5[4g_l^2 - (g_s - g_l)^2]g_l} \frac{\delta W}{\mu_B} \\ B(\frac{5}{2}, \frac{1}{2}; \frac{3}{2}, -\frac{3}{2}) &= \frac{[100g_l^4 + (g_s - g_l)^4 - 28g_l^2(g_s - g_l)^2]^{1/2} + (g_s - g_l)g_l}{5[4g_l^2 - (g_s - g_l)^2]g_l} \frac{\delta W}{\mu_B}. \end{aligned} \quad (3)$$

The crossings have been labelled by the quantum numbers J and m_J of the participating levels. From equations (3) it is evident that by an experimental determination of either of these crossing positions, the interval δW can be obtained.

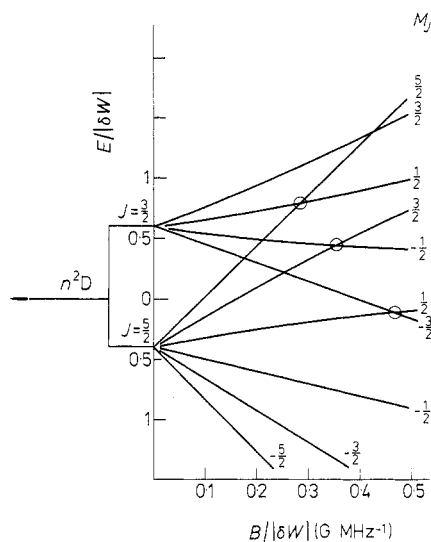


Figure 1. Energy-level diagram for an inverted D doublet. Three $\Delta m_J = 2$ level crossings are indicated. The diagram is valid for all values of $|\delta W|$.

In the level-crossing method (Breit 1933, Franken 1961), two crossing sublevels are excited coherently using σ light. If the σ component of the fluorescent light, released in the decay of these sublevels is detected, an interference effect in the scattered light intensity will be observed as a change in the angular distribution. The coherence effect, induced at the level crossing, cannot be transferred in a cascade decay. Thus the detection must follow in a spectral line originating in the studied state. For a suitable geometrical arrangement, the LC signal can be detected as a curve of Lorentzian shape. The full half-width ΔB of the Lorentzian is determined by the lifetime τ of the excited state and by the rate dv/dB at which the levels cross:

$$\Delta B = \left[\pi \tau \left(\frac{dv}{dB} \right) \right]^{-1}. \quad (4)$$

As a matter of fact, each of the discussed crossings for ^{23}Na (nuclear spin $I = \frac{3}{2}$) is made up of four crossings, since each of the participating levels is split up into four levels by the hyperfine interaction. However, the hyperfine interaction in the studied states is extremely weak and results only in a very small, symmetric broadening of the signals.

The sign of δW is not obtained in LC experiments of the kind discussed here. By observing the influence of an electric field on LC or quantum-beat signals, the sign can generally be inferred (Hogervorst and Svanberg 1975, Fabre and Haroche 1975). It is also possible to obtain the sign by observing relative intensities of components, registered with high-resolution optical spectroscopy methods. During the course of the present experiments, we tested a method where the fluorescent light from the resolved $^2D_{3/2}$ and $^2D_{5/2}$ states of the $n = 4$ level was studied. With defined excitation and detection conditions, the theoretical intensity ratios can easily be calculated (Condon and Shortley 1963). Since the components can be identified by their relative intensities, the correct order of the levels can be established.

A steady field was produced in a pair of Helmholtz coils and a sweep field in another pair. The fields were aligned with the north-south direction in the laboratory. The vertical component of the earth's magnetic field was compensated by means of a third pair of Helmholtz coils, not shown in the figure. For the field assignment, the currents for the main and sweep coils were taken through precision resistors and the voltages across these resistors were measured. The two fields were calibrated using optical pumping in the ground state of Cs^{133} .

Fluorescent light, released in the decay of the D state, was observed in the direction of the field, using the interior of a wide pumping line as a black background. The σ light, selected for wavelength with a 500 mm Bausch and Lomb grating monochromator, was detected with an EMI 9558 BQ photomultiplier tube. To reduce the dark current, Peltier cooling of the photomultiplier was used.

For lock-in detection of the LC signals, a rotating Polacoat PL 40 linear polarizer was used in the detection light beam. A reference signal for the lock-in amplifier was derived from a vane-switch oscillator. The lock-in signal was fed to a Laben averaging multichannel analyser. This analyser produced a reference voltage, which was used to control a repetitive linear field sweep, synchronous with the channel advancement. The signal accumulated in the multichannel analyser could be read out on a XY recorder. By reversing the direction of the main field for a fixed sweep-field direction and taking the average of the two measured signal positions, the influence from the horizontal component of the earth's magnetic field as well as from time constants in the lock-in amplifier and in the sweep were eliminated.

3.2. Level-crossing measurements

The excitation and fluorescence scheme and the corresponding lines are given in figure 3. The present LC measurements were performed for the D states with main quantum numbers ranging from 5 to 9. Corresponding experiments for the 4^2D level have been reported earlier (Fredriksson and Svanberg 1975). In order to obtain sufficient efficiency in the step-wise excitation, we operated with a fairly dense atomic beam so that multiple scattering was obtained in the first excitation step. This increased the fraction of atoms in the short-lived P state ($\tau = 16$ ns). An intense

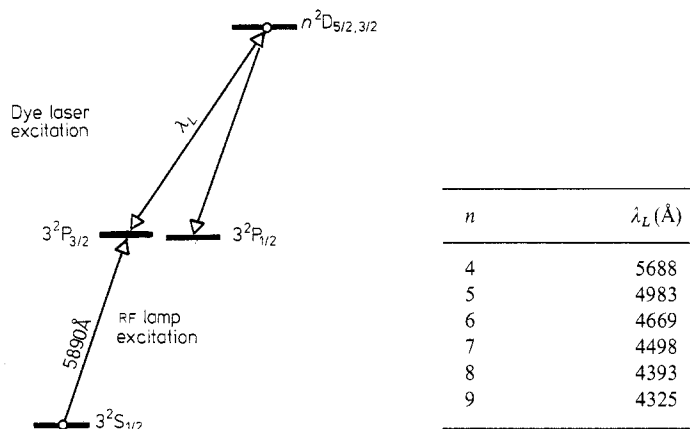


Figure 3. Excitation and fluorescence scheme and wavelengths for the laser-induced transitions used.

resonance light was observed from the scattering volume. It was necessary to use filters in front of the monochromator to reduce the stray yellow light in the monochromator housing.

With the geometrical arrangement used, it is possible to detect the LC signals as Lorentzian curves, provided that the proper phase is set for the lock-in amplifier.

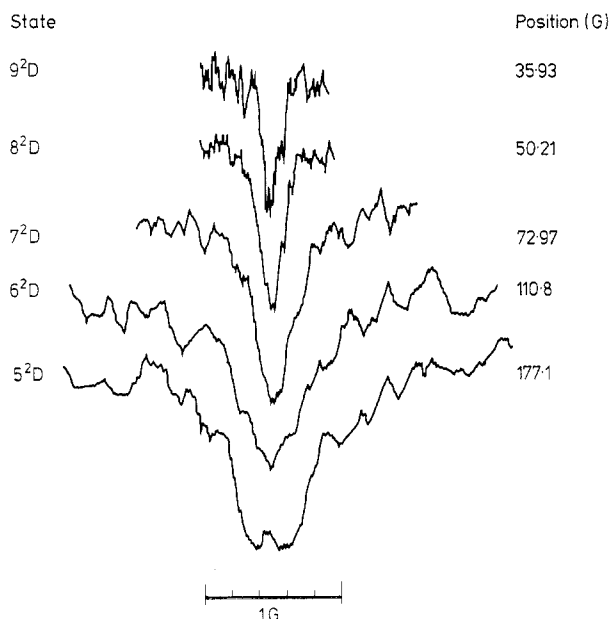


Figure 4. Experimental curves for the $n = 5$ to $n = 9$ states. Each curve has been sampled for 1–2 h.

The phase was adjusted before the runs by symmetrizing high-field LC curves. The strong zero-field level crossing (Hanle effect) was less suited for this adjustment, as there is a partial coherence transfer at zero field from the first excitation step. As the angle between the RF-lamp light beam and the laser beam is 135° , a dispersion-type curve will mix into the Lorentzian signal (Hogervorst and Svanberg 1975). For all the studied states the first LC signal was measured, eliminating shifts as discussed above. Further, for the 4^2D and 7^2D states the second crossing was also studied. In figure 4 experimental curves for the first crossing are shown. An integration time of 1–2 h was needed for each curve. The rapid increase in lifetime with higher n values (Kaiser 1975, Gallagher *et al* 1975), resulting in narrower LC signals (equation 4), can clearly be seen from the figure. In table 1, the measured magnetic-field values for the first crossing are given, obtained from several measurements. A field-calibration uncertainty of 0.03% is added to the statistical error. Further, an uncertainty for possible residual asymmetry has been included in the quoted field-value errors. The measured field positions for the second level-crossing signal in 4 and 7^2D (369.7(3) and 91.80(9) G, respectively) are entirely consistent with the corresponding positions for the first crossings. Using equations (3) with $g_l = 1.0000$ and $g_s = 2.0023$ (Schwinger-corrected), the values for the FS splittings, shown in the table, were obtained. At the field strengths used in these experiments higher-order corrections for other 2D states are completely negligible.

Table 1. Measured positions of the first level crossing, and the fine-structure splittings deduced from the level-crossing data.

n	$B(\frac{3}{2}, \frac{3}{2}; \frac{3}{2}, \frac{1}{2})$ (G)	$ \delta W $ (MHz)
4	293.68 (0.25)	1028.3 (0.6)
5	177.13 (0.17)	620.0 (0.6)
6	110.84 (0.11)	388.02 (0.35)
7	72.978 (0.070)	255.43 (0.25)
8	50.208 (0.050)	175.75 (0.17)
9	35.930 (0.035)	125.77 (0.12)

4. Laser-spectroscopy experiments

In order to obtain sign information for the sodium D states we tested a high-resolution laser spectroscopic method. The set-up was based on the arrangement used in the LC measurements. However, in this experiment we used a more collimated atomic beam (1 : 20) and performed a step-wise excitation of the sodium atoms using two lasers. Both laser beams were irradiated perpendicularly onto the atomic beam, in opposite directions to each other, and with overlapping paths. In the first step a cw rhodamine 6G dye laser was used to transfer atoms into the $3^2P_{3/2}$ level. The dye laser was operated in multi-mode. Using a piezo-electric mount for the output mirror of the dye laser, the mode structure was rapidly scanned across the absorption hyperfine lines, ensuring a 'white' excitation as a time average.

The second-step excitation to the D state was performed using a pulsed dye laser (Molelectron DL-400 pumped by a Molelectron UV-400 nitrogen laser). The dye-laser linewidth was about 10 GHz. In order to be able to obtain a high-resolution spectrum with this laser, we used the technique described by Grundevik *et al* (1976). By means of a Tropel model 240 Fabry-Pérot interferometer, operating in the filter mode, and inserted in the dye-laser beam, six transmission maxima, 20 MHz broad and 1500 MHz apart were obtained. By scanning the Fabry-Pérot piezo-electrically, the $^2D_{5/2}$ and $^2D_{3/2}$ levels were excited separately. We observed the fluorescent light released in the decay back to the $3^2P_{3/2}$ and $3^2P_{1/2}$ states unresolved. Boxcar techniques were used for the detection.

In order to define a quantization axis, a magnetic field of about 5 G was applied. For the first step π light was used. However, due to multiple scattering, the excitation tends to be isotropic. In the second step, unpolarized light was employed. For the detection, only the σ components were measured.

In figure 5 an experimental curve for the 4^2D state is shown. The curve has two peaks with an intensity ratio of about 8:1. The peaks are slightly above 1000 MHz apart, and thus correspond to the two fs components. The hyperfine structure in the $3^2P_{3/2}$ state, ranging over about 110 MHz, is not resolved. The Fabry-Pérot interferometer is piezo-electrically scanned towards 'redder' wavelengths, and the small component is observed before the large one. A theoretical calculation of the relative intensities results in eight or ten times stronger intensity for the $^2D_{5/2}$ state, depending on whether π or unpolarized excitation is assumed for the first step. Thus it is evident that the stronger of the measured signal components is due to the $^2D_{5/2}$ state, which obviously has a lower energy than the $^2D_{3/2}$ state. Thus

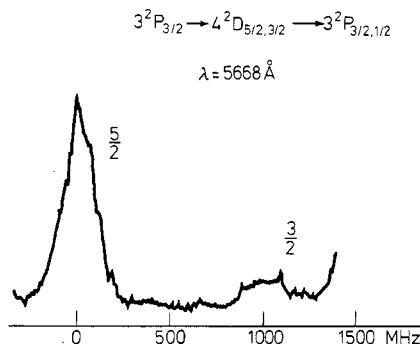


Figure 5. Fluorescence-light spectrum of the 4^2D state, obtained by narrow-band laser excitation of a collimated atomic beam. The free spectral range of the Fabry-Pérot filter was 1500 MHz.

the 4^2D state is inverted, which of course is known both from the early interferometric measurements and from the two-photon-absorption experiments. However, the present method can be used for measuring the signs of δW in all the states studied in this work. Unfortunately, we were lacking mirrors for our Fabry-Pérot filter for the appropriate wavelength region.

5. Discussion

In table 2 the FS splittings obtained in this work are given together with literature values for n^2D states in Na. For states, where LC as well as other measurements have been performed, the LC results obtained in this work are in general much more accurate. This is because the resolution in the LC measurements is only limited by the Heisenberg uncertainty relation and a very accurate calibration can be obtained for magnetic fields. Our result for the $4D$ state agrees well with the two-photon-spectroscopy values. Also for the $5D$ state, a good agreement with the result of Levenson and Salour (1974) is obtained. The 9^2D state has been measured by quantum-beat spectroscopy as well as with LC spectroscopy. The results agree within their limits of error. Thus the sequences of LC and quantum-beat measurements are nicely tied together.

In the introduction, we discussed the hydrogen-like model for the alkali atom. The FS splitting for a D state of hydrogen is given by

$$\delta W_H = \frac{hcR\alpha^2}{6n^3} \quad (5)$$

where R and α are the Rydberg and fine-structure constants, respectively. For a hydrogen-like system we can then write

$$\delta W = \frac{hcR\alpha^2 Z_i^2}{6n_{\text{eff}}^3} \quad (6)$$

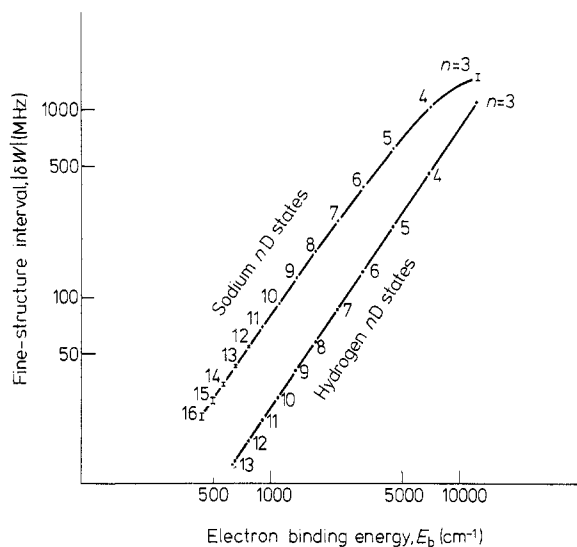
with

$$n_{\text{eff}} = \left(\frac{hcR}{E_b} \right)^{1/2} \quad (7)$$

where Z_i is the effective nuclear charge and E_b the electronic binding energy.

Table 2. Experimental and theoretical values for the fs splittings in n^2D states of sodium.

n	$ \delta W _{\text{exp}}$ (MHz)		$\delta W_{\text{theor}}^h$ (MHz)
	This work	Other work	
3		1482 (50) ^a	-1130
4	1028.3 (0.6)	1027 (16) ^b 1035 (10) ^c 1025 (6) ^d 1028.5 (3.0) ^e	-806
5	620.0 (0.6)	618 (12) ^f	-486
6	388.02 (0.35)	372 (50) ^g	-300
7	255.43 (0.25)		
8	175.75 (0.17)		
9	125.77 (0.12)	124.5 (1.5) ^h	
10		91.5 (1.0) ^h	
11		70.0 (0.7) ^h	
12		54.0 (0.5) ^h	
13		42.8 (0.6) ^h	
14		34.3 (0.8) ^h	
15		28.0 (1.0) ^h	
16		23.0 (1.0) ^h	

^a Meissner and Luft (1937).^b Bjorkholm and Liao (1974).^c Hänsch *et al* (1974).^d Pritchard *et al* (1974).^e Biraben *et al* (1974).^f Levenson and Salour (1974).^g Fabre *et al* (1975).^h Holmgren *et al* (1976).**Figure 6.** A plot of $\lg |\delta W|$ against $\lg E_b$ for the D sequence of hydrogen and sodium.

According to equations (5)–(7), a straight line with a slope of $\frac{3}{2}$ should be obtained if $\lg|\delta W|$ is plotted against $\lg E_b$. In figure 6 the curves for the hydrogen and sodium n^2D sequences are shown. For sodium a curved line is obtained. As the states with $n = 3, 4, 5, 6, 10, 11$ and 12 are known to be inverted, it is evident from the curve that the other ones are also inverted. The bend of the curve can be described by adding a term proportional to n_{eff}^{-5} in equation (6), as shown by Fabre and Haroche (1975). Clearly, the leading term, proportional to n_{eff}^{-3} , has a negative coefficient.

Recently, detailed many-body-perturbation calculations have been performed at this laboratory to shed light upon the anomalous D sequence (Holmgren *et al* 1976). The results for the $n = 3$ –6 levels are included in table 2. The inversion is reproduced and the numerical agreement is rather good. The inversion is found to be due to strong polarization effects in non-s subshells belonging to the core of electrons. Thus the contribution from the ‘closed shells’ is more important for the fine structure than the primary interaction of the d electron. So far, there is no experimental or theoretical evidence to show that the D sequence becomes ‘normal’ for high values of n .

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