Determination of lifetimes and hyperfine structures of the 8, 9 and 10 $^2D_{3/2}$ states of ^{133}Cs by quantum-beat spectroscopy

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Determination of lifetimes and hyperfine structures of the 8, 9 and 10 $^2D_{3/2}$ states of ^{133}Cs by quantum-beat spectroscopy

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Abstract. The states were studied by selective detection of the fluorescence generated in atoms of the vapour by stepwise excitation, using as sources a conventional caesium lamp for the first step from the ground state and a pulsed, tunable laser for the second. The techniques of delayed coincidence counting were applied to measure the time distribution of intensity of the fluorescent light. The observed modulation, representing in each case a superposition of the hyperfine intervals, showed a characteristic period which was interpreted as the reciprocal of the magnetic interaction constant a for the states in question. The relative simplicity of the modulation patterns is a consequence of the negligibly small quadrupole interaction in ^{133}Cs. With the polarizer oriented at $\cos^{-1} 3^{-1/2}$ to the analyser the modulation was suppressed, leaving a simple exponential decay from which the lifetime of the population, τ, could be determined.

The results are $a = 3.92 \pm 0.07$, 2.32 ± 0.04, 1.51 ± 0.02 MHz and $\tau = 148 \pm 3$, 208 ± 2, 310 ± 3 ns for the 8, 9 and 10 $^2D_{3/2}$ states, respectively. The values for a agree with those of Svanberg *et al*. Values of τ have not previously been reported.

1. Introduction

With the advent of tunable lasers, studies of highly excited states of atoms may be undertaken by the techniques of fluorescence spectroscopy. Small structures in these states may be explored by radio-frequency resonance or by methods based on coherent excitation of the states. Among these latter methods is that of 'quantum-beat' spectroscopy, that is to say, the study of modulation in exponentially decaying fluorescent light following the excitation of atoms by a pulse. The term 'quantum-beat' is used because the effect is a consequence of the oscillatory time-dependent probability of emission of light which is the same for all atoms excited at the same instant: it is not an effect arising from interference in the light from different atoms. It was studied originally for Zeeman structures by Dodd *et al* (1964), by Aleksandrov (1964) and by Hadeishi and Nierenberg (1965) and more recently, for hyperfine structures by Haroche *et al* (1973) and for fine structures by Haroche *et al* (1974). The same phenomenon is observed in beam foil spectroscopy (Andrä 1974).

The determination of structures in the alkalis is attractive because, being relatively simple atoms, they are good subjects for theoretical studies. A considerable number of measurements has been made of hyperfine structures in caesium. Early measurements of the ground states and excited P states have been followed recently by measurements on excited S and D states (see for example Gupta *et al* 1973, Svanberg *et al* 1973, Svanberg and Belin 1974, Tsekeris *et al* 1974). Our work provides confirmatory evidence for some values already reported, and additionally provides experimental values of some lifetimes

for which hitherto only theoretical estimates have been available. The states in question are 8, 9 and 10 $^2D_{3/2}$ of ^{133}Cs. Studies of some S states will be reported separately.

Fluorescence from these states was generated by the method of stepwise excitation. The first step, from the ground state 6 $^2S_{1/2}$ to the 6 ^2P states, was accomplished by continuous irradiation of the vapour by a conventional caesium lamp, and the second step, from one or other of the 6 ^2P states to the higher S or D states, by a pulsed, tunable laser.

The fluorescence was studied by measuring the distribution in time of photoelectrons following the laser pulse. Quantum-beat modulation was observed by placing a linear analyser either parallel or perpendicular to the polarizer in the laser beam: with the polarizer at $\cos^{-1}(\frac{1}{3})^{1/2}$ the coherent component of the fluorescent light was suppressed and the time dependence was a simple exponential decay, giving the lifetime of the excited state.

The beat frequencies are a measure of the hyperfine intervals in zero magnetic field, $|\Delta F| = 1$ or 2, where the F values range from 5 to 2. Because the quadrupole contribution in ^{133}Cs is small the interval rule is obeyed to a very good approximation and the beat patterns, though often rather complicated (figures 3 and 4), represent the superposition of a relatively small number of oscillations rationally related to one another, the whole having a well-defined repetition period which is the reciprocal of the magnetic dipole interaction constant a. Hence a measurement of this period gives directly the value of a without the need for Fourier analysis.

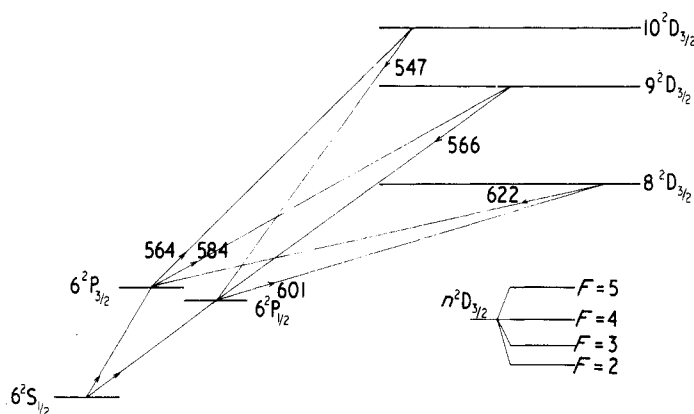The transitions studied are shown in figure 1.

Figure 1. Term diagram and transitions studied (wavelengths in nm). The intervals are not to scale. Hyperfine structure of a typical $D_{3/2}$ state is shown in the inset.

2. Description of the experiment

The disposition of apparatus is shown in figure 2. The fluorescence cell was cylindrical, 6·5 cm long, 3·5 cm diameter, with windows sealed parallel to one another as accurately as possible. By this construction parasitic light from the laser could be reduced to an acceptably low level. With caesium in the cell the temperature of the coldest part was thermostatically maintained at about 80 °C (vapour pressure 10^{-4} Torr). The earth's magnetic field inside the cell was compensated to within 10 mG.
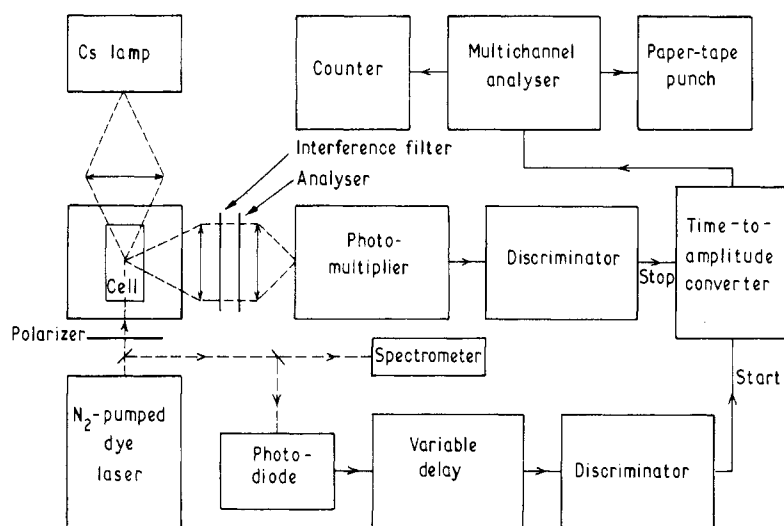
Figure 2. Arrangement of apparatus.

Incident radiation from the caesium lamp was unfiltered and unpolarized, and focused on the cell by a lens of large aperture. With the large amount of multiple scattering which must have occurred in the vapour, the first stage of excitation was substantially isotropic.

An AVCO model C5000 nitrogen laser was used to pump a dye laser. The dyes used in this work were rhodamine 6G for the 8D and 9D states and fluorescein disodium salt (FDS) for the 10D states. The dye laser produced flashes of about 5 ns duration, and the repetition rate normally used was 150 Hz. Rates up to 500 Hz were possible. Tuning was accomplished by rotation of a grating. A beam expander (10 ×) in front of the grating reduced the spectral line width to about 0·05 nm (5 GHz). The laser beam, about 2 mm in diameter, passed through a linear polarizer before entering the fluorescence cell. The desired wavelength was selected with the aid of an auxiliary spectrometer, and tuning to resonance monitored by measurement of the fluorescent intensity.

The fluorescent light in a wide angle cone perpendicular to the exciting beams was collected by an f/2·5 lens. Before reaching the photomultiplier it passed through an interference filter of bandwidth 6 nm to select a transition of wavelength different from that of the laser light, and also a linear analyser. Its intensity was measured as a function of time following the laser pulse by the techniques of delayed coincidence counting.

As shown in figure 2 the time-to-amplitude converter is started by a pulse from the laser flash and stopped by a pulse from the fluorescent light. The delay introduced into the former line ensured that no signal could be recorded until after the termination of the laser flash. The multichannel analyser (100 channels) sorted and stored pulses from the time-to-amplitude converter to form the time-decay spectrum. The contents of the memory were transferred to paper tape at the end of a run.

The power output from the laser was controlled so that, with a repetition rate of 150 Hz, the count rate in the analyser was between 20 and 50 Hz. A pile-up correction was applied to the recorded counts (Coates 1968). A background correction was also applied: cooling of the photomultiplier reduced the background by a factor of 100. Examples of decay curves, plotted logarithmically with the numbers of counts corrected as described, are shown in figures 3, 4 and 5.

The multichannel analyser was calibrated for time against the variable delay generator which itself was calibrated against a quartz-controlled oscillator.

3. Theory

The time dependence of the fluorescent light in these experiments is almost independent of the first stage of the excitation process and of the properties of the intermediate P states. The excitation of these states from the ground states is at a uniform rate and is almost entirely isotropic on account of the multiple scattering in the cell. There may be some small residual anisotropy arising from this excitation and also by stimulated emission from the D states, and this might have some small effect on the relative intensities of different components of the quantum beats, but we find excellent agreement between our experimental results and an analysis which supposes that the $|F, m_F\rangle$ components of the P states are statistically populated. These components form a set of ground states for the pulse excitation generated by the laser. We use the abbreviated notation $\{|\mu\rangle\}$ for these P states. The observed fluorescence is from a set of excited D states $\{|m\rangle\}$ to a set of lower P states $\{|\mu'\rangle\}$ which, in our experiments, are different from the $\{|\mu\rangle\}$.

The expression for the intensity at time t following a pulse from a set of equally populated states $\{|\mu\rangle\}$ is

$$I(t) = K \sum_{\substack{m,m' \\ \mu,\mu'}} \left(\frac{f_{m\mu}f_{\mu m'}g_{m'\mu'}g_{\mu'm}}{\Gamma - \mathrm{i}\omega_{mm'}} \right) \exp(\mathrm{i}\omega_{mm'} - \Gamma)t, \tag{1}$$

where f and g are electric dipole operators (Deech *et al* 1974 and earlier references given there). Γ is the decay constant of the states $\{|m\rangle\}$ and the $\omega_{mm'}$ are the Bohr frequencies†. Assumptions underlying equation (1) are: (i) that the duration of the pulse is short compared with Γ^{-1} and the $\omega_{mm'}^{-1}$; (ii) that the spectrum of the exciting light is broad compared with the hyperfine structure $\{|m\rangle\}$; and (iii) that the rate of excitation per atom is small compared with Γ. If (i) is violated the modulation is partially degraded but the periodicity remains unaltered. In our experiments the pulses were of duration about 5 ns and the periods that were actually measured ranged from 250 ns to 600 ns so that there was negligible degradation. Assumption (ii) was satisfied by a factor about 10^3. Assumption (iii)—which is equivalent to ignoring stimulated emission—might not have been satisfied in our experiments. The consequential transfer of population from the $\{|m\rangle\}$ to the $\{|\mu\rangle\}$ would not have been important. There can have been no transfer of coherence at hyperfine frequencies since the coherence time of the laser light was too short.

The expression for $I(t)$ in terms of multipole moments $T_q^{(k)}$ is especially convenient for a discussion of the spatial properties of the fluorescent light. The (k, q) term in the expansion contains the factor $E_q^{(k)}U_{-q}^{(k)}$, the product of the multipole components of the light transmitted by polarizer and analyser, respectively (Carrington and Corney 1971). The time dependence of the (k, q) term appears as a sum similar to that in equation (1) with

$$\omega_{mm'} \equiv \omega(F, m_F) - \omega(F', m_{F'}), \qquad (m_{F'} = m_F - q),$$

which is independent of the m_F since external fields are reduced to zero. The coherence that is observed is between states with m_F values differing by q.

† In Deech *et al* (1974), $\omega_{mm'}$ should be defined as $-(E_m - E_{m'})/\hbar$.

3.1. Linear polarization

Choose the z axis of the spherical basis to be the direction of the electric vector of the incident light. Then the only non-vanishing components $E_q^{(k)}$ are those having $q = 0$. $E_0^{(0)}$ is a measure of the population of excited atoms, $E_0^{(1)}$ is zero, and $E_0^{(2)}$ is a measure of alignment in the excited states. Coherence is established between hyperfine states with the same value of m_F.

If the analyser tensor is first written with reference to an axis z' parallel to the electric vector of the transmitted light, again the only non-vanishing components are $U_0^{(0)\prime}$ and $U_0^{(2)\prime}$. Under a rotation by the angle θ which brings the z and z' axes into coincidence, the $q = 0$ components are $U_0^{(0)\prime}$ and $\frac{1}{2}U_0^{(2)\prime}$ $(3\cos^2\theta - 1)$ respectively. Hence the intensity of fluorescent light has a spherically symmetrical term with time dependence $\exp(-\Gamma^{(0)}t)$, while the modulated terms $\exp(i\omega_{FF'} - \Gamma^{(2)})t$ depend on the relative orientation of polarizer and analyser as $(3\cos^2\theta - 1)$. (A similar discussion in relation to modulation at the Larmor frequency has been given by Gunn and Sandle 1971.) A distinction has been drawn here between the decay constant for population, $\Gamma^{(0)}$, and for alignment, $\Gamma^{(2)}$, though it was not expected that these would have different values in our experiments since multiple scattering of fluorescent light was not taking place (the lower state was not the ground state).

4. Results

The results were displayed as logarithmic plots of fluorescent intensity (number of counts, corrected for pile-up and background) against time (channel number, calibrated as described above). In figures 3, 4 and 5 the results are shown for fluorescence from $9\,^2D_{3/2}$ with the angle θ between polarizer and analyser having the values 0, 90°, 54° respectively, corresponding to the values 1, $-\frac{1}{2}$ and 0 of the function $\frac{1}{2}(3\cos^2\theta - 1)$.

Complicated modulation is clearly seen under the envelope of an exponential decay. The stronger features of figure 3(a) are readily identified in figure 4(a) with opposite sign and with substantially reduced amplitude. In figure 5 the modulation has been very largely suppressed. The dependence of the modulation on θ agrees satisfactorily with the theory when it is recalled that the fluorescent light was collected over a relatively large solid angle. Closer study of figures 3(a) and 4(a) shows that the pattern is repetitive with period rather less than 500 ns. This is evidence that the intervals in the hyperfine structure are rational multiples of a constant frequency and therefore obey the interval rule. Measurements of hyperfine structure in other levels of caesium (Violino 1969, Schmieder *et al* 1970) have shown that the ratio b/a of the electric quadrupole to the magnetic dipole interaction constant is of the order 1 %, which supports our conclusion. It is shown in table 1 below that the fractional deviations from the interval rule are substantially smaller than the ratio b/a.

With the assumption that deviations from the interval rule may be neglected, a measurement of the recurrence period for any recognizable feature in figures 3(a) or 4(a) gives directly the value of $1/a$. It is not necessary to allow for displacement of peaks or troughs by overlap from neighbouring features since the pattern—under our assumption—is exactly repeated. It is to be noted, however, that the statistical uncertainty of the points is larger for the smaller values of $\lg I$ so the best features to measure are the strongest peaks. Measurements from the experimental curves give $1/a$ to an accuracy approaching 1 in 200. The hyperfine intervals themselves for the $D_{3/2}$ states are $5a$,
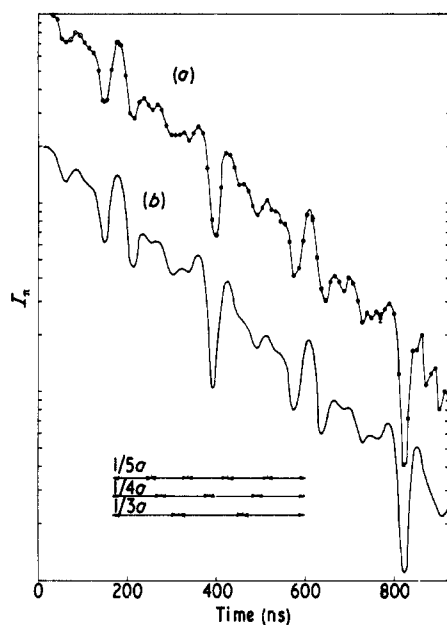
Figure 3. (*a*) Experimental decay spectrum for $9\ ^2D_{3/2} : I_\pi$ (logarithmic scale) against time (arbitrary origin). The full curve has been drawn to join the experimental points with no attempt to smooth statistical fluctuations. Error bars where shown represent one standard deviation. (*b*) Theoretical curve calculated from equation (1) using experimental values of *a* and Γ. Periods corresponding to the intervals 5*a*, 4*a* and 3*a* are indicated.
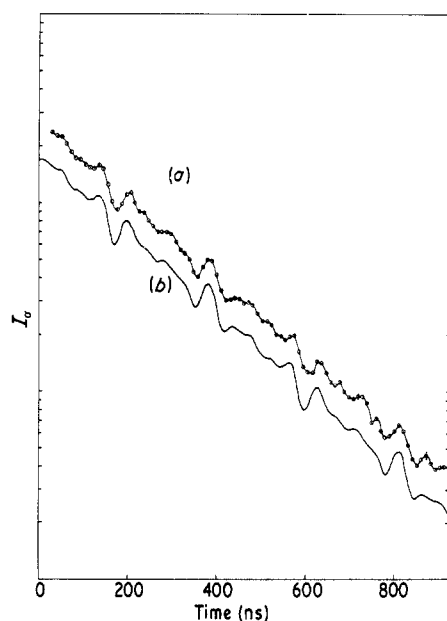

Figure 4. (*a*) Similar to figure 3(*a*), with I_σ instead of I_π. (*b*) The corresponding theoretical curve.
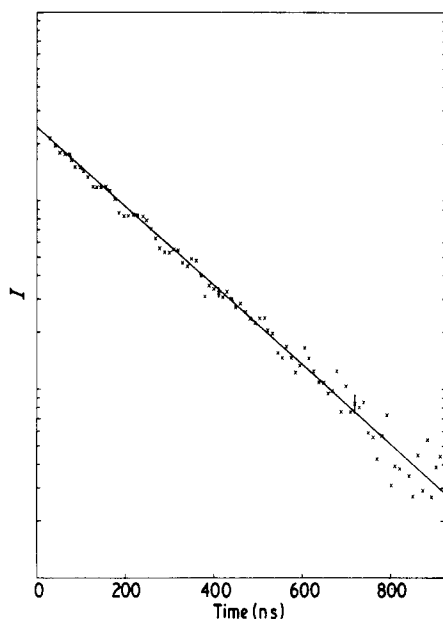
Figure 5. Experimental decay spectrum for $9\,^{2}D_{3/2}$ with $\theta = 54°$. The straight line is the least squares fit to the points. Some residual modulation is to be seen, arising from departure from the ideal geometrical conditions.

Table 1. Deviations from the interval rule as fractions of $b/a = x$.

F values	5–4	4–3	3–2
Intervals	$5a(1 + x/7)$	$4a(1 - x/14)$	$3a(1 - 5x/21)$

$4a$ and $3a$. The corresponding periods can easily be recognized in recurring features of figures $3(a)$ and $4(a)$.

The decay constants $\Gamma^{(0)}$ and the corresponding lifetimes were obtained by finding least squares fits to sets of points such as those shown in figure 5.

For comparison with the experimental curves, theoretical curves were calculated from equation (1) using experimental values of a and Γ. Such curves are shown in figures $3(b)$ and $4(b)$. The high degree of correlation between the theoretical and experimental curves is evidence that the theory of resonance fluorescence which is commonly used for single-step excitation using thermal sources is valid also in this more complicated situation. The correlation also justifies our neglect of the electric quadrupole interaction.

In table 2 below we compare our results with those of other observers and with theoretical calculations. Our values of a are based on a number of independent measurements of the period of the strongest recurring features in curves such as figures $3(a)$ and $4(a)$. The quoted errors are statistical and correspond to one standard deviation. The uncertainty in the time calibration is smaller than the statistical error by an order of magnitude.

Table 2. Values of a and $\tau = 1/\Gamma^{(0)}$.

| Level | a(MHz) | | | $\dfrac{a \text{ (this work)}}{a \text{ (theor)}}$ | τ(ns) | | $\dfrac{\tau \text{ (this work)}}{\tau \text{ (theor)}}$ |
	This work	Other work	Theor		This work	Theor	
$8\ ^2D_{3/2}$	$3\cdot92\pm0\cdot07$	$3\cdot98\pm0\cdot12^a$	$5\cdot12$	$0\cdot76$	148 ± 3	160	$0\cdot92$
$9\ ^2D_{3/2}$	$2\cdot32\pm0\cdot04$	$2\cdot37\pm0\cdot07^a$	$3\cdot11$	$0\cdot75$	208 ± 2	240	$0\cdot88$
$10\ ^2D_{3/2}$	$1\cdot51\pm0\cdot02$	$1\cdot52\pm0\cdot03^b$	$2\cdot03$	$0\cdot75$	310 ± 3	366	$0\cdot86$

[a] Svanberg *et al* (1973).
[b] Svanberg *et al* (1975).

5. Conclusion

Our measurements for a are in good agreement with those of Svanberg *et al* (1973) obtained at Columbia by the 'level-crossing' technique. Dr Svanberg, in private correspondence, has informed us that independent measurements made at Göteborg are also in agreement and that the sign of a has been determined by Stark effect measurements as being positive. The experimental values are not in agreement with values calculated from the Goudsmit–Fermi–Segré theory (Kopfermann 1958), but the experimental and theoretical values are in a constant ratio, to within the experimental error.

We believe these are the first reported measurements of the lifetimes of these states, though it was clear from the level-crossing experiments that the lifetimes could not be grossly different from those calculated by the method of Bates and Damgaard (1949) which are given in the last column but one of table 2. The experimental values are lower than the calculated values, and the fractional difference becomes progressively greater in the sequence 8D, 9D, 10D, contrary to what one might expect. In this connection we recall the well known anomaly in the intensity ratio of the members of the doublets of the principal series (Fermi 1930, Fulop and Stroke 1973).

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