

## LETTER TO THE EDITOR

# Quantum defect and fine-structure measurements of P, D, F and G Rydberg states of atomic caesium

L R Pendrill†, D Delande and J C Gay

Laboratoire de Spectroscopie Hertzienne de l'ENS, Université Pierre et Marie Curie, 4 place Jussieu, 75230 Paris Cedex 05, France

Received 26 July 1979

**Abstract.** Using a single-mode dye laser to excite and a thermoionic diode to detect highly excited caesium vapour atoms, we have extended the range of measured term values in caesium for the levels  $n^2P_{1/2,3/2}$  ( $20 \leq n \leq 60$ ),  $n^2D$  ( $54 \leq n \leq 58$ ),  $n^2F$  ( $23 \leq n \leq 109$ ) and  $n^2G$  ( $27 \leq n \leq 36$ ). The atomic transition wavenumbers were measured against the absorption spectrum of  $I_2$  vapour, affording an estimated uncertainty of  $\pm 0.01 \text{ cm}^{-1}$ .

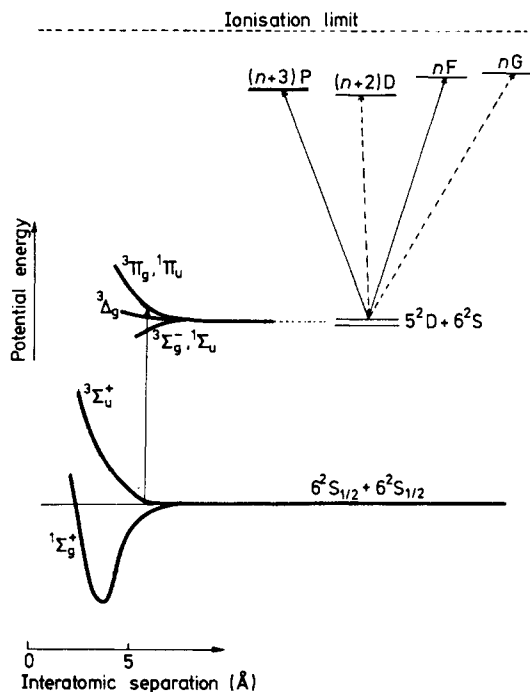
The ability to scan the wavelength of a highly monochromatic cw dye laser allows selective excitation of long sequences of closely spaced energy levels in atoms and molecules. Combining this with the high sensitivity of the thermoionic diode detector (Harvey and Stoicheff 1977), we have made measurements of an appreciable number of energy levels of Cs I, using as a wavenumber reference the well known absorption spectrum of molecular iodine (Gerstenkorn and Luc 1976, 1979).

The apparatus is largely identical to that described in earlier publications (Mirza and Duley 1978, Collins *et al* 1974). A cw monomode dye laser (Spectra Physics Model 580 dye cell), pumped by an argon ion laser, produced a beam of yellow light (power 50 mW, linewidth 10 MHz, Rhodamine 6 G) which was passed into a Pyrex cell containing pure caesium vapour at 200 °C (vapour pressure about 0.1 Torr). Highly excited atoms so produced were detected with a thermoionic diode, after the design of Harvey and Stoicheff (1977), i.e. the atoms being excited in an electrostatically shielded compartment.

Highly excited P, D, F and G levels of atomic Cs were excited by transitions from the  $5^2D$  atomic levels. The  $5^2D$  levels were populated by photodissociation of the  $Cs_2$  molecules (hybrid resonances, Collins *et al* 1974), the same wavelength light being used for both the molecular dissociation and the atomic excitation. The excitation scheme is depicted in figure 1.

For wavenumber calibration we monitored the absorption spectrum of molecular iodine simultaneously with the atomic spectra; passing part of the laser light through an iodine cell of length 50 cm and measuring the transmitted intensity with a photodiode. By referring to the iodine spectral atlas of the Aimé-Cotton Laboratory (Gerstenkorn and Luc 1976, 1979), we were able to measure the wavenumber (in vacuo) of each Cs spectral line to within an uncertainty of  $\pm 0.01 \text{ cm}^{-1}$ . This accuracy compares favourably with that of previous classical grating spectrometer and etalon measurements on

† Present address: JILA, University of Colorado, Boulder, Co 80309, USA.



**Figure 1.** Excitation scheme. Potential energy of two Cs atoms plotted against the interatomic separation. The transitions indicated by broken lines are 'forbidden' transitions.

caesium (Lorenzen and Niemax 1979, Sansonetti *et al* 1979 and private communication, Eriksson and Wenåker 1970, Kleiman 1962).

In deducing the term values and quantum defects of the excited states from the observed transition wavenumbers, we took the ionisation limit of Cs I to be  $31406.454 \text{ cm}^{-1}$  and the term values of the  $5^2D_{3/2}$  and  $5^2D_{5/2}$  levels to be  $14499.257 \text{ cm}^{-1}$  and  $14596.845 \text{ cm}^{-1}$ —these values were measured by Eriksson and Wenåker (1970).

Figures 2(a) and 2(b) give two examples of the spectra obtained; the first shows the higher members of the fundamental series  $5^2D_{3/2} \rightarrow n^2F$  with  $n$  as high as 99, and the second displays the observed allowed transitions to the  $n^2P$  levels as well as some of the 'forbidden' transitions observed to the  $n^2G$  levels. At other wavelengths, other transitions were observed between the  $5^2D_{5/2}$  level and the  $n^2F$  and  $n^2D$  levels. The occurrence of the forbidden transitions may indicate the presence of stray electric fields although these were expected to be small, particularly since the high Rydberg F levels, having giant electric polarisabilities, appear to be unperturbed. We intend to present a separate communication on these forbidden transitions.

The quantum defect  $\mu$  of each level was deduced from the measured term value  $T$  by the Rydberg formula:

$$T = -R/(n - \mu)^2 \quad (1)$$

where  $n$  is the principal quantum number and  $R = 109737.86 \text{ cm}^{-1}$ , the caesium Rydberg constant.

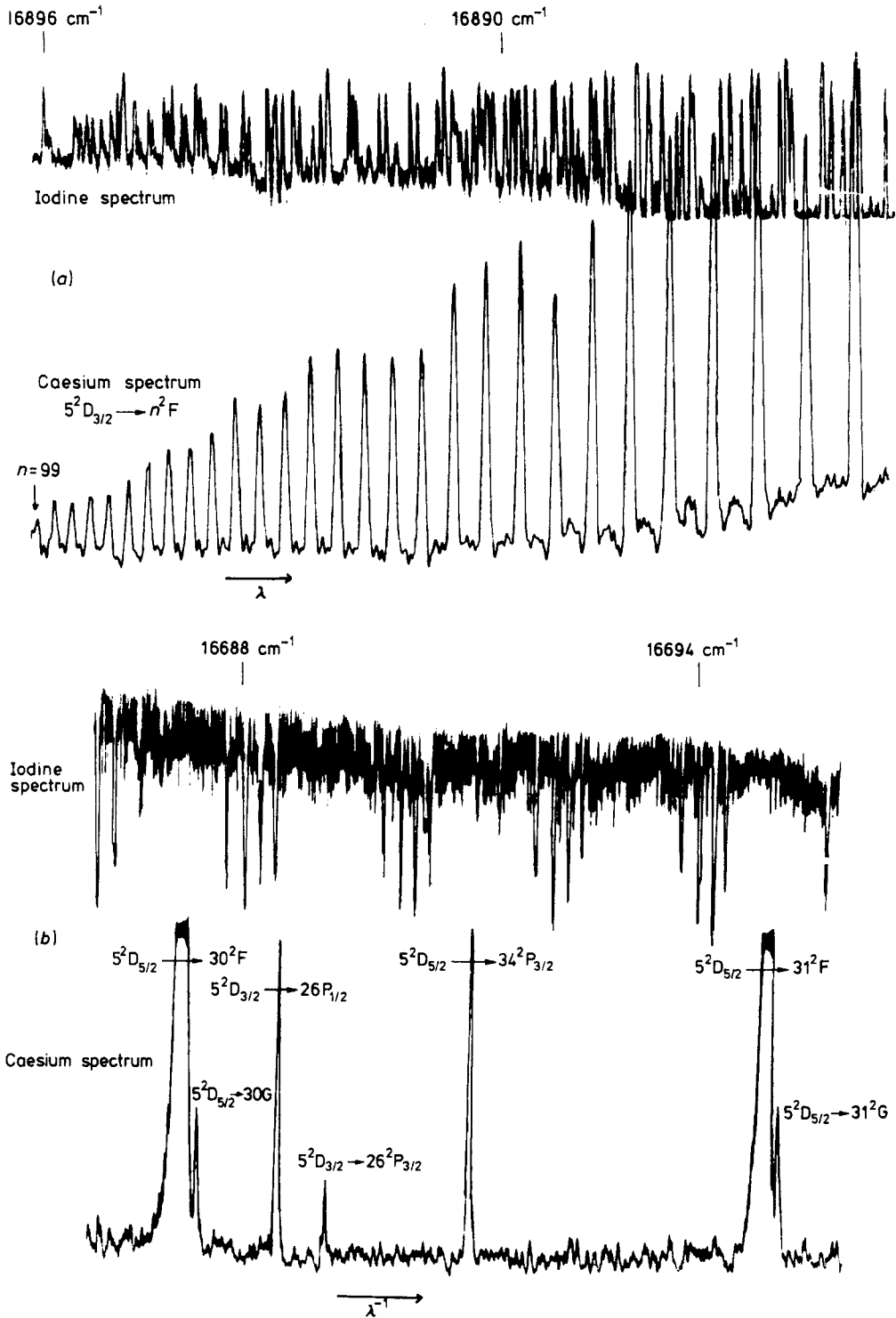


Figure 2. Caesium and iodine spectra. Diode current versus laser wavelength: (a) around  $16890 \text{ cm}^{-1}$ , (b) around  $16690 \text{ cm}^{-1}$ .

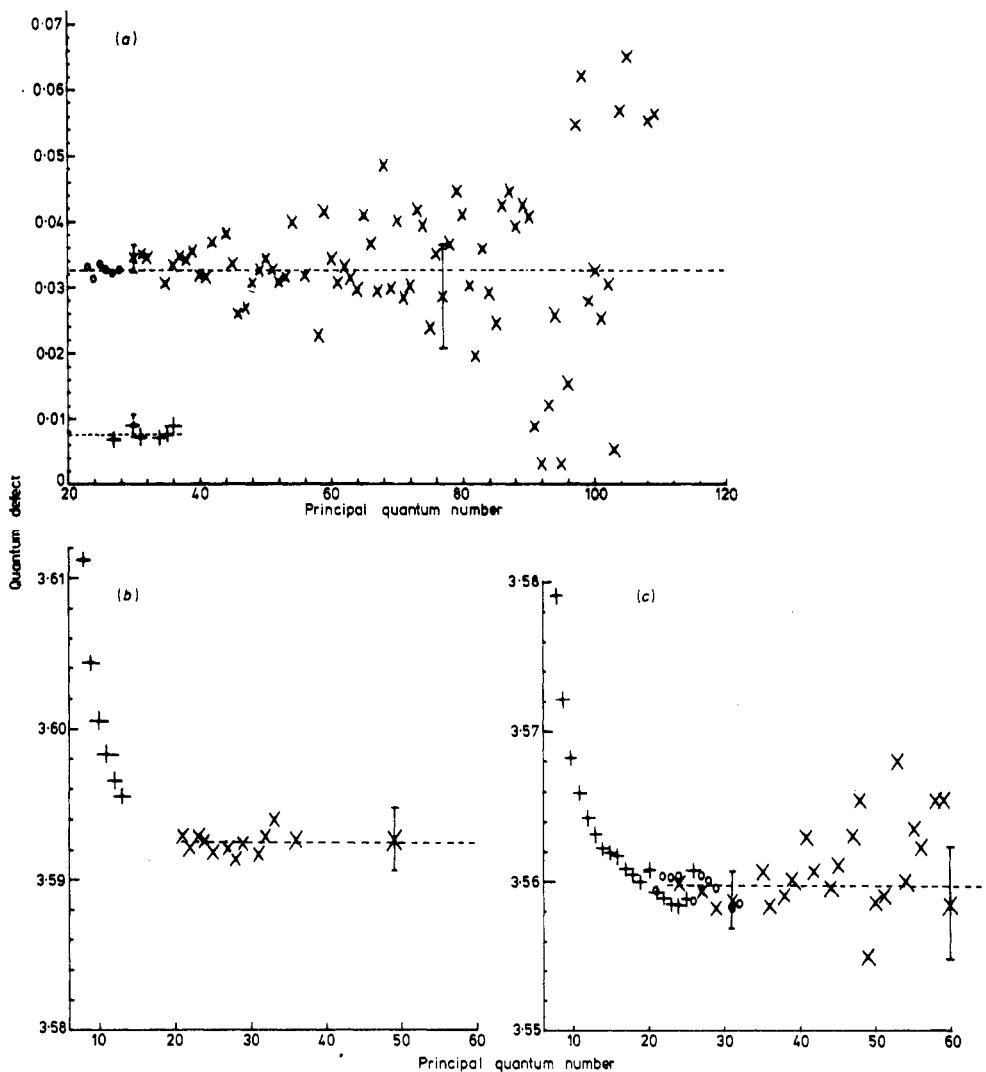
Our results for the quantum defects are summarised in figures 3(a), (b) and (c), in which  $\mu$  is plotted against  $n$  for the  $n^2F$ ,  $n^2G$  levels, the  $n^2P_{1/2}$  levels, and the  $n^2P_{3/2}$  levels, respectively.

Generally  $\mu$  is a function of  $n$  which may be described by the Rydberg-Ritz formula:

$$\mu = a + bT + cT^2 + dT^3 + \dots \quad (2)$$

(e.g. Eriksson and Wenåker 1970).

The terms in equation (2) which depend on  $T$  are important only for the low-lying energy levels, as exemplified in the results of the measurements of Lorenzen and



**Figure 3.** Quantum defect  $\mu$  plotted against principal quantum number  $n$ . (a)  $\circ$ ,  $5^2D_{3/2} \rightarrow n^2F$ ;  $\times$ ,  $5^2D_{5/2} \rightarrow n^2F$ ;  $+$ ,  $5^2D_{5/2} \rightarrow n^2G$  (this work). (b)  $\times$ ,  $5^2D_{3/2} \rightarrow n^2P_{1/2}$  (this work);  $+$ ,  $6^2S_{1/2} \rightarrow n^2P_{1/2}$  (Lorenzen and Niemax 1979). (c)  $\circ$ ,  $5^2D_{3/2} \rightarrow n^2P_{3/2}$ ;  $\times$ ,  $5^2D_{5/2} \rightarrow n^2P_{3/2}$  (this work);  $+$ ,  $6^2S_{1/2} \rightarrow n^2P_{3/2}$  (Lorenzen and Niemax 1979).

Niemax (1979) shown for comparison with our data in figures 3(b) and (c) for the levels  $n^2P_{1/2,3/2}$ . For all our measured quantum defects, we have found  $\mu$  to be constant ( $=a$ ) with  $n$ , as is expected for these higher energy levels: the resulting mean quantum defects are represented in figure 3 by the horizontal broken lines and their values are tabulated in table 1 together with the results of other experiments.

Table 1. Caesium quantum defects.

State		This work	Others
$n^2P_{1/2}$	$21 \leq n \leq 49$	3.5926(5)	3.5918(5) Lorenzen and Niemax (1979)
$n^2P_{3/2}$	$19 \leq n \leq 60$	3.5597(9)	3.5588(7) Lorenzen and Niemax (1979)
$n^2D$	$54 \leq n \leq 58$	2.476(3)	2.470(50) Kleiman (1962) (weighted mean $D_{3/2}, D_{5/2}$ )
$n^2F$	$23 \leq n \leq 109$	0.033(3)	0.033 Collins <i>et al</i> (1974)
$n^2G$	$27 \leq n \leq 36$	0.0077(8)	0.00686(3) Sansonetti <i>et al</i> (1979)

Our results for the quantum defects appear to be systematically slightly larger than those of other experimenters: the differences (which are within the quoted error bars) correspond to shifts towards the red of all the spectral lines of the order of  $0.005 \text{ cm}^{-1}$ . This value is not inconsistent with the expected shift (Lorenzen and Niemax 1979) due to  $\text{Cs}^* + \text{Cs}$  collisions at the vapour temperature ( $200^\circ\text{C}$ ) used in our experiments. There is qualitative agreement with theoretical predictions using a Fermi potential and a single scattering length approximation (Omont 1977).

We observed the fine structure of the  $n^2P$  levels to principal quantum numbers considerably higher than has been possible via the principal series transitions. In our measurements, the signals from the transitions  $5^2D_{3/2} \rightarrow n^2P_{1/2}$  were larger than those from the  $n^2P_{3/2}$  levels—as expected from a simple statistical weight argument (Kuhn 1964) and as may be seen from figure 2(b)—whereas for the principal series transitions

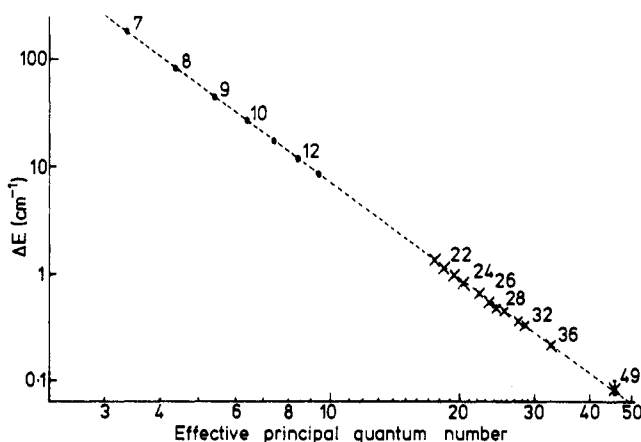


Figure 4. Log-log plot of the  $n^2P_{1/2,3/2}$  fine-structure interval  $\Delta E$  versus effective principal quantum number  $n^*$ .  $\times$ , this work;  $\circ$ , Lorenzen and Niemax (1979). The number beside each symbol indicates the principal quantum number.

$6^2S_{1/2} \rightarrow n^2P_{1/2}$ , the oscillator strength becomes very small compared with that of the  $n^2P_{3/2}$  levels for  $n$  above 20 (Raimond *et al* 1978).

Figure 4 shows a log-log plot of the  $n^2P_{1/2,3/2}$  fine-structure interval  $\Delta E$  versus the effective principal quantum number  $n^*$  ( $=n - \mu$ ). Our results (crosses) are compared with those of Lorenzen and Niemax (1979) (circles). The straight line, which is the result of a least-squares fit to our data, indicates a variation of  $\Delta E$  with  $n^*$  having a functional form:

$$\Delta E = K(n^*)^\alpha$$

where  $\alpha$ , given by the slope of the line, is found to equal  $-2.983 (\pm 0.030)$ . This is in close agreement with the value of  $-3$  expected from an hydrogenic model of the caesium atom.

We would like to thank Dr Andrew and Dr Niemax for giving us their two sets of results prior to publication and for valuable discussions. We are also grateful to Dr J Vigué for the loan of an iodine absorption cell.

## References

- Collins C B, Johnson B W, Mirza M Y, Popescu D and Popescu I 1974 *Phys. Rev. A* **10** 813-21  
 Eriksson K B and Wenåker I 1970 *Phys. Scr.* **1** 21-4  
 Gerstenkorn S and Luc P 1976 *Atlas du Spectre d'Absorption de la Molécule d'Iode* (Paris: Editions CNRS)  
 — 1979 *Revue Phys. Appl.* **14** 791-4  
 Harvey K C and Stoicheff B P 1977 *Phys. Rev. Lett.* **38** 537-40  
 Kleiman H 1962 *J. Opt. Soc. Am.* **52** 441-7  
 Kuhn H G 1964 *Atomic Spectra* (London, Harlow: Longmans)  
 Lorenzen C J and Niemax K 1979 *J. Quant. Spectrosc. Radiat. Transfer* in press  
 Mirza M Y and Duley W W 1978 *J. Phys. B: Atom. Molec. Phys.* **11** 1917-20  
 Omont A 1977 *J. Physique* **38** 1343-59  
 Raimond J M, Gross M, Fabre C, Haroche S and Stroke H H 1978 *J. Phys. B: Atom. Molec. Phys.* **11** L765-77  
 Sansonetti C, Andrew K L and Verges J 1979 *EGAS Conference Paris-Orsay*