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LETTER TO THE EDITOR

Measurements of the n^3D_1–n^3D_2 (n = 3–8) fine-structure separations in ^{4}He I by the beam–foil quantum-beat method

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Abstract. Beam–foil quantum-beat techniques have been used to measure the $^3D_1$–$^3D_2$ intervals of 1s*n*d (n = 3–8) in ^{4}He I. A two-spectrometer technique was used so as to obtain simultaneously a time calibration from the 1s3p ($^3P_1$–$^3P_2$) fine-structure separation. The following $^3D_1$–$^3D_2$ separations (in MHz) were found: 1323·6 ± 2·3 (n = 3), 553·0 ± 0·7 (n = 4), 284·1 ± 0·6 (n = 5), 165·3 ± 1·0 (n = 6), 101·6 ± 1·1 (n = 7) and 69 ± 3 (n = 8). These results are compared with previous measurements and theoretical values.

The beam–foil quantum-beat technique has been widely applied for atomic fine- and hyperfine-structure measurements (see e.g. Andrä 1974). However, the accuracy is often limited by beam spreading, velocity straggling and foil thickening which can introduce uncertainties in the time calibration of the in-flight radiative emission. We have developed a procedure which largely avoids such time-calibration errors. The foil-excited beam is viewed simultaneously by two optical spectrometers, one of which records a known quantum-beat pattern while the other records the quantum-beat pattern to be studied. In the He I spectrum the fine structure of the 1s3p ^3P term is known accurately and the $^3P_1$–$^3P_2$ separation at 658·55 ± 0·15 MHz (Lifsitz and Sands 1965) manifests itself in easily observable quantum beats in the beam–foil decay curves of the 3889 Å multiplet (1s2s ^3S–1s3p ^3P). We have used these for an accurate determination of the n^3D_1–n^3D_2 (n = 3–8) intervals. Comparison between experimental and theoretical values provides insight into the amount of singlet–triplet mixing and its n and l dependence.

The measurements were performed using 100–300 keV He$^+$ ions from the Stockholm 400 kV heavy-ion accelerator. The light from the beam was viewed by two optical monochromators and single-photon counting was performed with Peltier cooled photomultiplier tubes. The foil motion was achieved through a precision-ruling engine screw (linearity ±0·05%) which was driven by a stepping motor (resettable to ±0·01 mm). The size and number of steps were controlled by an on-line programming unit. The stepping was triggered by the accumulation of a fixed number of counts from a monitor phototube which viewed the beam through a fibre optics link at a fixed position downstream from the foil. The beam current and foil condition

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were also monitored by a Faraday cup at a constant distance from the foil, and a gate circuit interrupted the measurement if the current through the foil varied outside preset limits. At each position of the foil, the number of counts from each of the photomultipliers and the accumulator time were routed and stored in a 4000-channel analyser. After a prescribed number of foil sweeps the data were read on to paper tape. Computer analysis then corrected for background contributions, deconvoluted the multi-exponential decay and performed a Fourier transform and a least-squares fit. The result of a typical measurement is shown in figure 1.

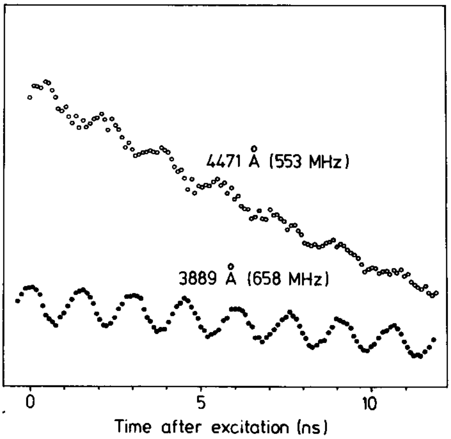

Figure 1. Quantum beats in the decay of the 4471 Å (1s2p ^{3}P–1s4d ^{3}D) and 3889 Å (1s2s ^{3}S–1s3p ^{3}P) lines in He I.

In order to obtain high precision in the determination of the 658·55 MHz calibration frequency, our step size was set at a distance corresponding to a few tenths of a nanosecond, and the intensity followed for 200 steps. Thus our sensitivity to the $^3D_1$–$^3D_2$ beats (which range from 70–1000 MHz) was high, but the sensitivity to the $^3D_2$–$^3D_3$ beats (which range from 5–70 MHz) was much lower. The $^3D_1$–$^3D_3$ beat frequency had too low an amplitude to be able to be extracted. Berry *et al* (1972) have computed the expected beam–foil beat intensity ratios for these intervals to be $A_{12}:A_{13}:A_{23} = 35:4:40$. Thus this measurement reports only the $^3D_1$–$^3D_2$ separations.

Our values are presented and compared with other measurements in table 1. Our results for $n = 3$–7 are in agreement with the earlier beam–foil work of Berry *et al* (1972) but have much smaller uncertainties due to our improved velocity definition. For $n = 3$ our results are in agreement with, but of lower precision than, the level-crossing measurements of Kaul (1968) and Tam (1975). For $n = 4$ our result is slightly smaller than that of Tam, and our precision is still a little lower. It is also worth noting that Tam actually measured the $^3D_1$–$^3D_3$ and $^3D_2$–$^3D_3$ separations for $n = 3, 4$, the difference of which gives the $^3D_1$–$^3D_2$ intervals with slightly larger errors. For $n = 5$ and 6 it is clear that our beam–foil accuracies are comparable to, or better than, available level-crossing measurements. The beam–foil method can also be extended, without too much effort, to higher levels, which are difficult to reach by step-wise laser excitation.

Table 1. Fine-structure intervals $^3D_1$–$^3D_2$ in He I, measured from the 1s2p ^3P–1snd ^3D (n = 3–8) transitions.

Upper term	Wavelength (Å)	Interval (MHz) This work	 Earlier beam–foil[a]	 Other experiments[b]
3d ^3D	5875	1323·6 ± 2·3	1349 ± 25	1324·7 ± 0·4[c] 1327·2 ± 1·1[d] 1358 ± 30[e]
4d ^3D	4471	553·0 ± 0·7	536 ± 30	555·1 ± 0·3[c] 561 ± 30[e]
5d ^3D	4026	284·1 ± 0·6	290 ± 20	282 ± 2[f]
6d ^3D	3819	165·3 ± 1·0	150 ± 20	166 ± 3[f]
7d ^3D	3705	101·6 ± 1·1	92 ± 15	
8d ^3D	3636	69 ± 3		

[a] Berry *et al* (1972).
[b] This list is not complete, some older measurements being omitted.
[c] Tam (1975), level crossing.
[d] Kaul (1968), level crossing.
[e] Brochard *et al* (1957), Fabry–Perot.
[f] Dily and Descoubes (1971), level crossing.

For purposes of comparison with theoretical estimates we have computed the deviation from an n^{-3} scaling law for all values. A simple theoretical model which ignores, for example, singlet–triplet mixing, has been given by Bethe and Salpeter (1957). Using recent values for the fundamental constants (Cohen and Taylor 1973) we can write the frequency ν_B given by Bethe and Salpeter for the n^3D_1–n^3D_2 separation as

$$\nu_B = (35\,033 \text{ MHz})n^{-3} \qquad (1)$$

which accounts for the dominant part of the n dependence. Thus the percentage deviation $100 \times (\nu - \nu_B)/\nu_B$, where ν is the measured frequency for a given $^3D_1$–$^3D_2$

Table 2. Comparison of the percentage deviation $100 \times (\nu - \nu_B)/\nu_B$ between this experiment and theory.

n	Experiment a	Theory b	 c	 d	 e
3	2·0 ± 0·2	3·7	2·4	2·1	−1·5
4	1·0 ± 0·1	4·6	2·0	1·7	−1·6
5	1·4 ± 0·2	4·8			
6	1·9 ± 0·6	5·4			
7	−0·5 ± 1·1	5·7			
8	1 ± 4	5·2			

[a] This work (uncertainties represent standard deviations).
[b] Parish and Mires (1971).
[c] Van den Eynde *et al* (1972). Fine structure derived from their work by Tam (1975).
[d] Bessis *et al* (1964) and Ambry *et al* (1968). Fine structure derived from their work by Tam (1975).
[e] Araki (1937).

separation, provides a very sensitive comparison between theory and experiment, as is presented in table 2. Our results are clearly in disagreement with those of Parish and Mires (1971), and to a less extent with all other theoretical results available. It is interesting to note that Parish and Mires report singlet–triplet mixing parameters which are up to a factor of two less than those reported by Van den Eynde *et al* (1972). Tam (1975) has pointed out that the n^3D_2–n^3D_3 fine-structure separations seem to depart more severely from the n^{-3} scaling law. Measurements of increased precision for these frequencies are now in progress.

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References

Ambry C, Bessis N and Moser C M 1968 *Phys. Rev.* **170** 131–5
Andrä H J 1974 *Phys. Scr.* **9** 257–80
Araki G 1937 *Proc. Phys. Math. Soc. Japan* **19** 128–55
Berry H G, Subtil J L and Carré M 1972 *J. Physique* **33** 947–54
Bessis N, Lefebvre-Brion H and Moser C M 1964 *Phys. Rev.* **135** A957–9
Bethe H A and Salpeter E E 1957 *Handb. Phys.* **35** 1–454 (Berlin: Springer-Verlag)
Brochard J, Chabbal R, Chantrel H and Jacquinot P 1957 *J. Phys. Radium* **18** 596–602
Cohen E R and Taylor B N 1973 *J. Phys. Chem. Ref. Data* **2** 663–734
Dily D and Descoubes J-P 1971 *C. R. Acad. Sci., Paris* B **272** 1182–4
Kaul R D 1968 *J. Opt. Soc. Am.* **58** 429
Lifsitz J and Sands R H 1965 *Bull. Am. Phys. Soc.* **10** 1214
Parish R M and Mires R W 1971 *Phys. Rev.* A **4** 2145–60
Tam A C 1975 *Phys. Rev.* A **12** 539–50
Van den Eynde R K, Wiebes G and Niemeyer Th 1972 *Physica* **59** 401–18