

# Towards a Precision Measurement of the Lamb Shift in Hydrogen-Like Nitrogen

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**Abstract.** Measurements of the  $2S_{1/2} - 2P_{1/2}$  and  $2S_{1/2} - 2P_{3/2}$  transitions in moderate  $Z$  hydrogen-like ions can test Quantum-Electrodynamic calculations relevant to the interpretation of high-precision spectroscopy of atomic hydrogen. There is now particular interest in testing calculations of the two-loop self-energy. Experimental conditions are favorable for a measurement of the  $2S_{1/2} - 2P_{3/2}$  transition in  $N^{6+}$  using a carbon dioxide laser. As a preliminary experiment, we have observed the  $2S_{1/2} - 2P_{3/2}$  transition in  $^{14}N^{6+}$  using a 2.5 MeV/amu foil-stripped ion beam and a continuous-wave  $CO_2$  laser operating on the hot band of  $^{12}C^{16}O_2$ . The measured value of the transition centroid,  $834.94(7) \text{ cm}^{-1}$ , agrees with, but is less precise than theory. However, the counting rate and signal-to-background ratio obtained indicate, that with careful control of systematics, a precision test of the theory is practical. Work towards constructing such a set-up is in progress.

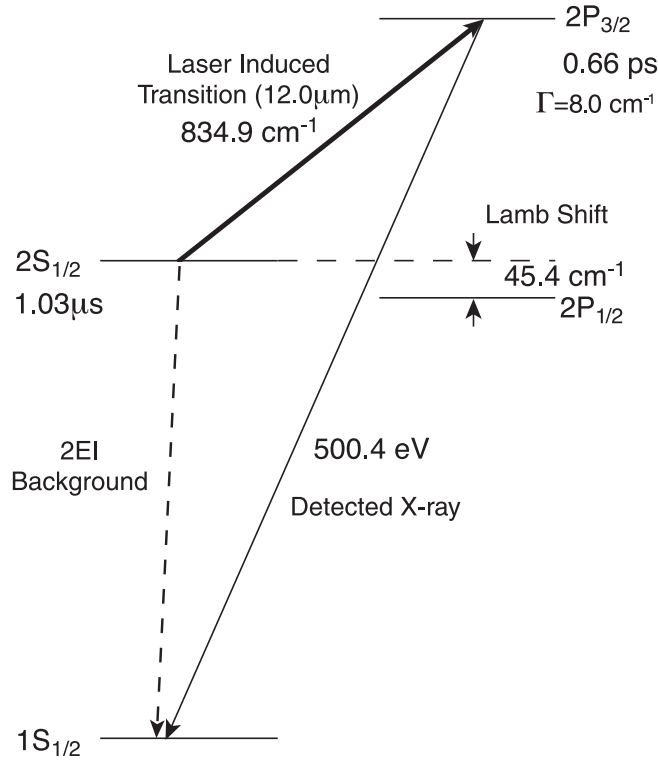
## 1 Introduction

An important problem in the theory of one-electron atoms is the calculation of binding corrections to the two-loop contribution to the self-energy, particularly for terms of order  $(\alpha/\pi)^2(Z\alpha)^2[(Z\alpha)^4/n^3]mc^2$  and higher [1,2,3]. Such terms limit the accuracy of QED corrections in atomic hydrogen to a few ppm, and limit the precision of the values for the Rydberg constant and the proton mean-square charge radius, that can be extracted from ultra-high precision spectroscopy of atomic hydrogen [4,5,6,7,8]. One way to study this problem is to make comparisons between theory and experiment in moderate  $Z$  ions. Because the terms of interest as a fraction of the measured intervals scale approximately as  $Z^2$ , a useful test can be provided by Lamb Shift measurements of lower precision than those in hydrogen.

For various ions the  $2S_{1/2} - 2P_{1/2}$  and  $2S_{1/2} - 2P_{3/2}$  transitions match the wavelengths of certain efficient lasers enabling spectroscopy with the fast-beam laser-resonance technique [9]. The most precise measurement of this type, achieved after considerable experimental development, was of the  $2S_{1/2} - 2P_{3/2}$  transition in  $P^{14+}$  [10], where an uncertainty equivalent to 0.15% of the  $2S_{1/2} - 2P_{1/2}$  interval was obtained. A high-power pulsed dye-laser producing intensities of over  $10 \text{ MW/cm}^{-2}$ , but with a duty cycle of about  $10^{-5}$ , was required to obtain a good signal-to-background ratio.

However, in these experiments it can be shown that the signal for a given laser intensity scales as  $Z^{-6}$  while the background, from the two-photon decay of the

$2S_{1/2}$  level, scales as  $Z^6$ . Hence at lower  $Z$ , at the expense of somewhat reduced sensitivity to higher-order QED corrections, adequate signal-to-background can be achieved with much lower laser intensity, allowing continuous-wave lasers to be used. Apart from the great increase in overall signal, the necessary characterization of the laser beam at the interaction region, in time, frequency, power and spatial distribution, is considerably easier with continuous wave lasers. The  $2S_{1/2} - 2P_{3/2}$  transition in  $N^{6+}$ , see fig. 1, is near  $12.0 \mu\text{m}$ , and is accessible to the highly efficient  $\text{CO}_2$  laser. This system is therefore a good candidate for a precision Lamb Shift measurement.



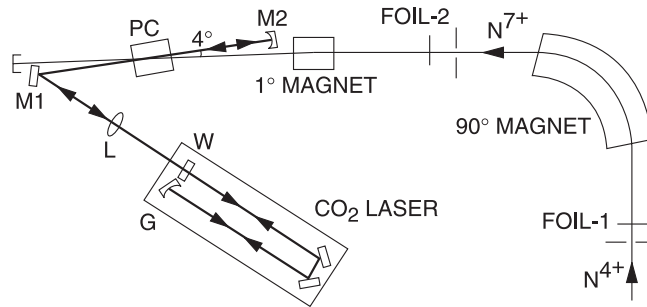
**Fig. 1.** Energy levels of  $N^{6+}$  relevant to the Lamb shift measurement

Ivanov and Karshenboim [11] have determined a theoretical value for the  $2S_{1/2} - 2P_{3/2}$  transition in  $^{14}\text{N}^{6+}$  of  $834.928(7) \text{ cm}^{-1}$ , where the error is dominated by the uncertainty in the two-loop binding corrections. Other QED corrections, and the nuclear size correction (based on the stated error of the nuclear rms charge radius measurements), contribute only about  $0.001 \text{ cm}^{-1}$ . Hence an experimental precision of  $\pm 0.002 \text{ cm}^{-1}$  or better would provide an interesting test of the two-loop corrections. This corresponds to subdivision of the  $8 \text{ cm}^{-1}$  natural resonance FWHM by a factor of 4000.

## 2 Survey Experiment

### 2.1 Experimental Arrangement

The experimental arrangement used for the survey experiment is shown in fig. 2. This was a modification of an existing set-up designed for co-linear spectroscopy of heliumlike ions [13,14]. A beam of 35 MeV  $^{14}\text{N}^{4+}$  ions was obtained from the Florida State University tandem accelerator. The accelerated ions were focused onto a  $10\text{ }\mu\text{g}/\text{cm}^2$  carbon foil, Foil-1 in fig. 2, placed near the entrance slits of a  $90^\circ$  momentum analysing magnet. Fully stripped ions were selected by the magnet and then passed through a second, nominally  $5\text{ }\mu\text{g}/\text{cm}^2$  foil, Foil-2. Here approximately 30% of the ions captured an electron to produce  $\text{N}^{6+}$ , of which the order of 10% are formed in the metastable  $2S_{1/2}$  state. Typical currents of  $\text{N}^{6+}$  were  $1 - 2$  particle-nA. The ions then traveled a distance of 4.1 m to the laser-ion interaction region. In order to make use of our existing interaction chamber it was necessary to deflect the ions by  $1^\circ$  using a magnet placed 1.2 m up-beam of the interaction region. The resulting laser-ion intersection angle, as subsequently measured by scanning a  $25\text{ }\mu\text{m}$  wire through the laser and ion beams, was  $4.07 \pm 0.15$  degrees. A small intersection angle was advantageous as regards increased laser-ion interaction time and hence signal. It also enabled the transition to be reached with laser wavelengths around  $11.1\text{ }\mu\text{m}$ , making use of the Doppler shift. A true co-linear geometry was not practical due to the problem of Stark quenching of the  $2S_{1/2}$  state in the motional electric field of the bending magnet.



**Fig. 2.** Set-up used for the survey experiment. G is a diffraction grating, W a window, L a lens, M1 and M2 are mirrors, PC is a proportional counter

The laser system [13] was based on a flowing gas, 7 m discharge length, industrial  $\text{CO}_2$  laser, originally rated at 350 W. The laser cavity was extended to produce an intracavity waist, with spotsize approximately 1.5 mm in diameter at the interaction region. A 75 lines/mm concave diffraction grating enabled selection of different laser lines. In order to produce a range of wavelengths to map out the broad resonance, the laser was successively operated on the P-23 to

P-41 rotational lines of the  $[01^11] - [11^10, 03^10]_I$  (hot) band of  $^{12}\text{C}^{16}\text{O}_2$ . Precise wavenumbers for these lines were obtained from ref. [15]. To facilitate background subtraction the laser discharge was switched at 500 Hz with a duty cycle of 50%. Typical time averaged intracavity powers of 150 W at discharge currents of 10-40 mA, depending on the laser line, were obtained. The intracavity power was determined by measuring the power transmitted through the nominally 99% reflecting mirror M2, and using the manufacturer's data for the transmission as a function of wavelength. Unfortunately, presumably due to thermal effects and hysteresis effects in the laser discharge, it was often difficult to achieve stable and reproducible lasing, and retuning between different laser lines often took several minutes. There was also difficulty in resolving the components of the closely spaced hot-band doublets.

X-rays from the ions were detected by a proportional counter mounted above the midpoint of the interaction region. The counter, operated with flowing pure  $\text{CH}_4$  at 300 mbar, was fitted with a  $0.6\text{ }\mu\text{m}$  thick aluminized polypropylene window. The effective detector aperture, 2.5 cm above the ion beam, subtended a solid angle of 0.4 sterad at the center of the interaction region, the solid angle falling to zero at points 3 cm up and down-beam. Hence the observation region was reasonably well matched to the trapezoidal (when viewed from above) laser-ion interaction region, of approximate total length 5 cm. Allowing for losses due to window absorption and the window supporting grids, the effective quantum efficiency for detecting the 500 keV Lyman- $\alpha$  X-rays was estimated to be 40%.

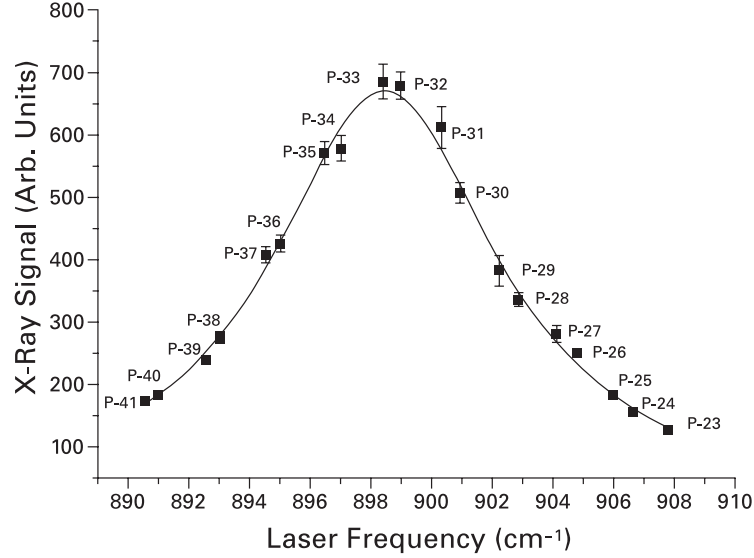
The measured laser induced signal was  $1.0 \times 10^5\text{ s}^{-1}$  per particle-nA. This is consistent with an estimated laser induced transition probability, based on time dependent perturbation theory, of 1.7%, together with the above estimate of detection probability, and a  $2S_{1/2}$  fraction of approximately 10%.

The two-photon decay of  $2S_{1/2}$  produces a continuous spectrum peaked near 250 eV. Sensitivity to the high energy part of this spectrum is suppressed in our detector by the K-absorption edge of carbon in the polypropylene window, while the lower part is discriminated electronically by a suitable threshold setting. The observed background rate was 4 kHz/particle-nA. Thus the signal-to-background ratio in the resonance peak was 25.

## 2.2 Procedure

The  $2S_{1/2} - 2P_{3/2}$  resonance was scanned at a fixed beam energy and interaction geometry by selecting different laser lines by manually adjusting the diffraction grating and adjusting the discharge current to maintain approximately constant laser power. For each laser line the difference in X-ray counts, laser(on) - laser(off), as well as the average laser power and beam current, was recorded for 30 seconds. The data from such a scan is shown in fig. 3. The counting statistics in our 30 s sample correspond to a measurement of the signal level to better than .1%. The much greater scatter observed in the data of fig. 3 is believed to be due to non-reproducible changes in the shape and position of the laser mode at the interaction region as the wavelength is changed, and also due to variations in the sensitivity of our proportional counter, and variations in the sensitivity and

baseline shifts in our measurements of laser power and beam current used for the normalization. Because this experiment was intended as a survey, no great attempt was made to study and reduce these problems.



**Fig. 3.** Resonance curve obtained by changing the laser line. P-J denotes a vibrational-rotational line of the hot-band of  $^{12}\text{CO}_2$

The laser-induced X-ray count rate data, normalized to laser power and beam current, were fitted with a Lorentzian using a least-squares technique to obtain the resonance centroid in the laboratory frame. The average resonance width, corrected to the rest frame of the ions, was  $8.5 \pm 0.4 \text{ cm}^{-1}$ , compared to the natural width of  $8.0 \text{ cm}^{-1}$ . This is consistent with some saturation in the transition probability and also in the detection sensitivity of the proportional counter at increased count rate. The wavenumber of the resonance centroid, in the rest frame of the moving ion, is obtained using the relativistic Doppler formula,

$$\omega' = \omega\gamma(1 - \beta \cos(\theta)) \quad (1)$$

where  $\omega'$ ,  $\omega$  are the centroids in the moving and laboratory frames, respectively,  $\beta = (\text{ion velocity})/c$ ,  $\gamma = (1 - \beta^2)^{-1/2}$ , and  $\theta$  is the intersection angle between the ion and laser beam in the laboratory frame. An initial value for the beam velocity was obtained from an NMR measurement of the magnetic field of the  $90^\circ$  magnet, using the standard laboratory calibration which was based on an accurately measured proton induced nuclear resonance. This was corrected for energy loss in the foil using the manufacturer's quoted thickness, and a tabulation of energy loss of fast ions in carbon [16]. An improved calibration and estimate of foil thickness was obtained by inducing the  $2^1S_0 - 2^3P_{1,F=1}$  transition in

$N^{5+}$  using the  $\text{CO}_2$  laser, also with X-ray detection. The wavenumber of this narrow resonance has been previously measured to be  $986.5799(7) \text{ cm}^{-1}$  [13]. The procedure was to transmit a  $N^{5+}$  beam through the analysing magnet, and scan resonances by varying the beam velocity, with and without the second foil in place. By using the P-28 line of the regular,  $10.4 \mu\text{m}$  band, and using the laser beam at  $186^\circ$  to the ion beam, these resonances occurred at approximately 17.5 MeV. The magnetic field in the analysing magnet, when transmitting  $N^{5+}$ , was then close to the field used when transmitting  $N^{7+}$  at the higher energy.

### 2.3 Results

Our result for the corrected centroid of the  $^{14}\text{N}^{6+} 2S_{1/2} - 2P_{3/2}$  transition is  $834.94(7) \text{ cm}^{-1}$ . The fitting error,  $0.05 \text{ cm}^{-1}$ , was obtained from the scatter of centroids of three separate data sets and is consistent with the error supplied by the fitting routine. To this were added, in quadrature, contributions due to the  $0.15^\circ$  error in the intersection angle ( $0.013 \text{ cm}^{-1}$ ), and errors due to the magnetic field calibration equivalent to 3 gauss ( $0.036 \text{ cm}^{-1}$ ), and a conservative 40% uncertainty in the 25 keV energy loss in the carbon foil ( $0.0086 \text{ cm}^{-1}$ ). The error in the magnet calibration was based on the simple difference between the “standard” calibration and the more reliable one obtained from the  $N^{5+}$  resonances. This error, and in fact all the errors associated with the Doppler shift can be reduced by an order-of-magnitude by careful attention to detail, using procedures we have already developed for measurements on heliumlike ions. There was no statistically significant shift in the centroid when the “adder” foil was relocated to be only .4 m upstream of the interaction region. This shows that the interaction between the laser and the possible small fraction of long lived Rydberg states, which survives to the interaction region, is not an obvious obstacle to a precision measurement. The hyperfine splitting of the  $2S_{1/2}$  level in  $^{14}\text{N}^{6+}$  is approximately  $0.22 \text{ cm}^{-1}$ , and produces a negligible broadening and asymmetry to the resonance in the present experiment.

Our result is compared with the theoretical results of Johnson and Soff [12] and Karshenboim and Ivanov [11] in table 1.

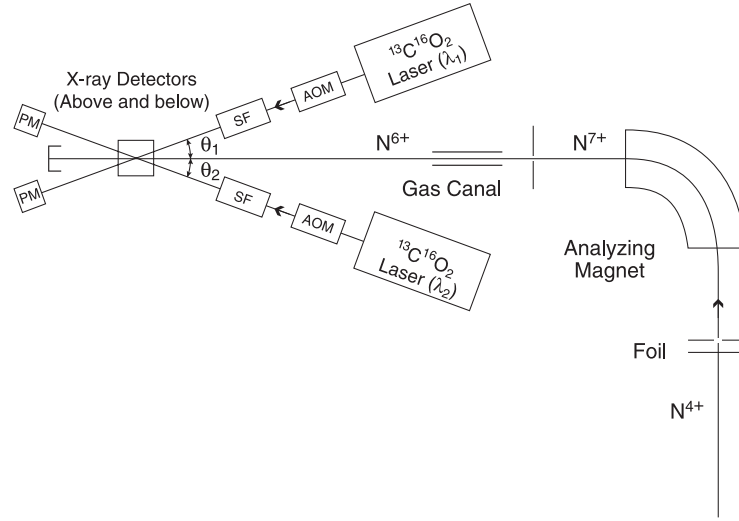
**Table 1.** Comparison of result of survey experiment with theory for the  $2S_{1/2} - 2P_{3/2}$  transition on  $\text{N}^{6+}$ . Units are  $\text{cm}^{-1}$

Theory	Johnson and Soff (1985)	834.913
	Ivanov and Karshenboim (2000)	834.928(7)
Experiment	This Work	834.94(7)

### 3 Ongoing Work

#### 3.1 Set-up using Two $^{13}\text{C}^{16}\text{O}_2$ Lasers and a $5^\circ$ Interaction Geometry

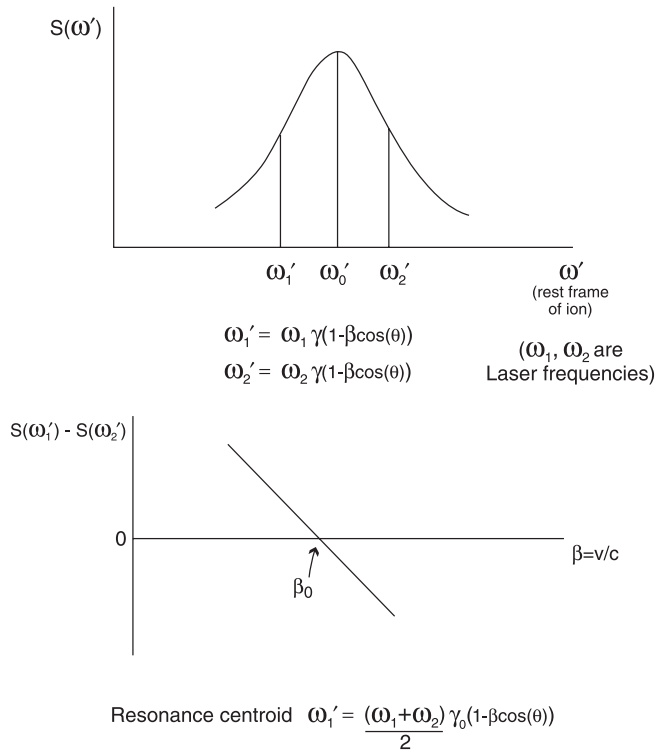
By far the most serious problems with the previous experiment were the difficulties of precisely measuring and leveling the intracavity laser power, and of keeping the interaction geometry constant, as the laser line was changed. To address these problems we are building a new setup where the large industrial  $\text{CO}_2$  laser operating on the hot-band is replaced by two scientific  $\text{CO}_2$  lasers operating on the regular band of  $^{13}\text{C}^{16}\text{O}_2$ , see fig. 4.



**Fig. 4.** Schematic of set-up using two  $^{13}\text{CO}_2$  lasers

The power outputs of the lasers will be actively stabilized and matched using a system of thermopile power meters (PM) and acousto-optic modulators (AOM's). Spatial filters (SF) will be used to ensure a well defined laser mode at the interaction region. A system of beam scanners will be used to accurately characterize the laser and ion beams at the interaction region and to measure the intersection angles  $\theta_{1,2}$ .

Although various interchanges of laser wavelengths, power meters, etc. will be made to control systematics, the basic technique for the measurement is indicated in fig. 5. The lasers are set on laser lines approximately equidistant from, but on either side of the resonance centroid, and balanced in power. The lasers are chopped in anti-phase and the difference signal,  $S(\omega_1) - S(\omega_2)$  is recorded. The beam velocity is varied till the “zero-crossing” (where the signals are equal) is found. The resonance centroid (in the ion's rest frame) is then obtained from the relativistic Doppler formula and the mean of the two laser frequencies.



**Fig. 5.** Schematic of “zero-crossing” technique for determining the resonance centroid

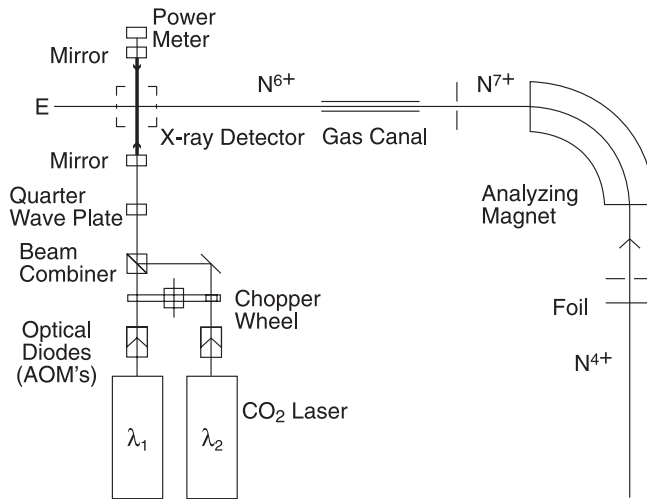
The beam velocity will be obtained by making use of a third laser, operating on the normal band of  $^{12}\text{C}^{16}\text{O}_2$ , to induce the  $N^{5+} 1s2s \ ^1S_0 - 1s2p \ ^3P_{1,F}$  resonances, which have been measured to better than 1 ppm. This hence gives the absolute value of  $\beta$  for the  $1s2s \ ^1S_0 \ N^{5+}$  ions to a comparable precision. The difference in energy loss between ions leaving the gas canal in the  $N^{6+} 2S_{1/2}$  state and in the  $N^{5+} 2^1S_0$  state has to be allowed for. But the effect on the  $2S_{1/2} - 2P_{3/2}$  measurement is only 1.2 ppm per keV energy shift. Hence the beam velocity can be determined with sufficient precision for a measurement at the level of a few ppm. Because of certain cancellations, the measurement is more sensitive to the angle between the laser beams, than to the angles of intersection of either beam with the ion beam. To achieve the required accuracy it will be sufficient to measure this angle to 0.2 mrad.

### 3.2 Set-up using a Transverse Geometry and a $^{14}\text{C}^{16}\text{O}_2$ Laser

Using a laser operating on  $^{14}\text{C}^{16}\text{O}_2$ , the regular band of which spans  $12.0 \mu\text{m}$ , it is possible to perform the experiment with a transverse geometry with a fast ion beam, or with a slower ion beam, for example from an ECR ion source. A possible setup is outlined in fig. 6. Because of the shorter interaction time, a build-up

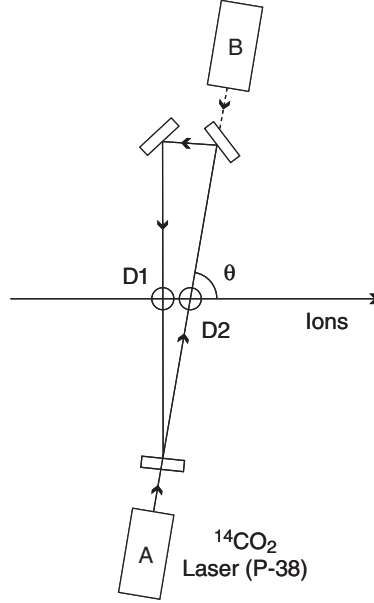


cavity will be required to obtain sufficient power ( $\sim 200$  W) with a fast beam. To avoid sensitivity to the first-order Doppler shift it is essential that the co- and counter-propagating beams be accurately co-linear. The chief technical difficulty, however, is that of monitoring and leveling the intracavity power as the laser wavelength is changed. Monitoring the power by monitoring the transmitted power through one of the mirrors suffers from the wavelength dependence, and possible power dependence (due to thermal effects) of the transmission of the high-reflection coatings.



**Fig. 6.** Schematic of set-up using two  $^{14}\text{CO}_2$  lasers

A possible solution to monitoring the laser power in the build-up cavity is indicated in fig. 7. This scheme is developed from the “auto-collimation” technique of Mueller et al. [17]. Our scheme (in its simplest variant) is based on a triangular ring build-up cavity, in which the direction of propagation can be precisely reversed, e.g. by alternately feeding it with two lasers, A or B. The two lasers operate on the same laser line, e.g. P-38 at  $833.684\text{ cm}^{-1}$ . By correctly selecting the intersection angle and beam velocity  $\beta$ , it is possible to arrange that the laser intersecting the ion beam under detector D2, will be Doppler shifted by equal amounts above and below the resonance centroid. Hence equal signals in D2 will be obtained, as the direction of propagation is alternated, provided the circulating power at the interaction region is kept constant. (We neglect the small relativistic change to the laser power in the moving frame here. This can be calculated and allowed for.) The resonance centroid, in the ion rest frame, is then given by  $\omega' = \gamma\omega_l$ , where  $\omega_l$  is the laser frequency and  $\gamma = (1 - \beta^2)^{-1/2}$ . In practice the ion beam velocity would be varied to search for the point of equal signals. For P-38, the required ion beam energy would be close to 19.2 MeV and, for the Doppler shifted frequencies to correspond to the half-height points on the



**Fig. 7.** Simplified schematic of set-up to facilitate power leveling in a build-up cavity

resonance,  $\theta = 85^\circ$ . Beams, propagating in either direction, intersect the ion beam under detector D1 at  $90^\circ$ , and are Doppler shifted (via the second-order Doppler shift) to the resonance peak. Hence equal signals should be obtained for the beams in either direction, with a relatively weak dependence on beam energy, enabling the exact balancing of the laser power.

#### 4 Conclusion

We have carried out the first measurement of the  $2S_{1/2} - 2P_{3/2}$  interval in  $N^{6+}$ . Our result is in good agreement with the theory, but the precision of  $0.07 \text{ cm}^{-1}$ , or .17% of the Lamb Shift interval, is not sufficient to provide a useful test. However the count rates and signal-to-background ratio achieved, 100 kHz/particle-nA and 25 respectively, are consistent with obtaining a statistical precision of  $\pm 0.001 \text{ cm}^{-1}$ . The beam current and detector solid angle can be increased in future experiments if necessary.

The Doppler shift, even with a near co-linear geometry, is *not* a serious limitation to the precision, at least at the level of  $\pm 0.002 \text{ cm}^{-1}$  for the  $2S_{1/2} - 2P_{3/2}$  interval. We are pursuing a set-up using a  $5^\circ$  intersection angle, with two  $^{13}\text{C}^{16}\text{O}_2$  lasers. The beam velocity calibration will make use of previously accurately measured transitions in heliumlike ions.

To reduce sensitivity to the Doppler shift further the experiment can be carried out using a transverse geometry on a fast beam, or on a slow beam, or

even with an ion trap, using a  $^{14}\text{C}^{16}\text{O}_2$  laser. Obtaining sufficient laser power to carry out the experiment with a fast beam may require the use of a laser build-up cavity and a technique for accurately monitoring the intracavity power.

With adequate laser equipment, and precise characterization of the interaction geometry, detector response, and measurement of laser power, sufficient precision to test the theory of the two-loop binding corrections should be obtainable.

## 5 Acknowledgments

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