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**PRECISION MEASUREMENT OF THE $1s2s^1S_0 - 1s2p^1P_1$
INTERVAL IN HELIUMLIKE BERYLLIUM**

by

Timothy James Scholl

Department of Physics

**Submitted in partial fulfilment
of the requirements for the degree of
Doctor of Philosophy**

**Faculty of Graduate Studies
The University of Western Ontario
London, Ontario
November 1988**

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ABSTRACT

This work represents the first test of non-relativistic, relativistic, and quantum electrodynamic calculations for two-electron systems using a high precision measurement of the $2^1S - 2^1P$ interval of a heliumlike ion. The method of laser resonance spectroscopy on a fast ion beam of ions was used to measure the $1s2s^1S_0 - 1s2p^1P_1$ interval of heliumlike beryllium to be $16276.774(9) \text{ cm}^{-1}$. Absolute frequency calibration was provided by the iodine atlas of Gerstenkorn and Luc. This result is in excellent agreement with both theory and previous experiment with an improvement in experimental precision by a factor of 11.

The measurement of the width of the observed laser resonance yields a lower bound of $7.1(5) \text{ ps}$ on the lifetime of the 2^1P state. Careful analysis and modelling of the observed lineshape shows that the lineshape is completely understood and suggests a lifetime which is compatible with theory.

Another related experiment is proposed to measure the $1s2s^3S_1 - 1s2p^3P_1$ intervals in Be III using the same experimental technique. The analogous $2^1S - 2^1P$ and $2^3S - 2^3P$ intervals of heliumlike boron are also suggested as future experimental systems for tests of atomic structure theory.

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CHAPTER 1

Spectroscopy of One- and Two-Electron Atoms

The study of one- and two-electron atomic systems has for many years played an important role in our understanding of quantum mechanics. The relative simplicity of these atoms permits their energies to be precisely calculated, making them excellent systems for tests of atomic structure theory. Theoretical calculations of hydrogenic systems are extremely precise, equalling or surpassing the accuracy of experiment. Using Dirac's relativistic theory, calculations for a single electron in a fixed Coulomb potential are exact except for small corrections due to the motion and structure of atomic nuclei as well as quantum electrodynamic (QED) effects. For two-electron systems, no closed-form formulation analogous to the one-electron Dirac formalism exists; nevertheless, approximation methods of ever increasing precision are becoming available to challenge experiment.

1.1 One-Electron Systems

The first measurement of an atomic structure radiative (QED) effect was made by Lamb and Retherford in 1947.¹ The $2^2S_{\frac{1}{2}}$ and $2^2P_{\frac{1}{2}}$ levels of hydrogen (as well as hydrogenic ions) are predicted to be degenerate by the Dirac equation as a result of the inverse-square law for the Coulomb force between the electron and proton. A non-degeneracy was suspected as early as 1938 by Pasternack² who suggested spectroscopic observations of the components of the Balmer- α line could be explained by an upwards displacement of the $2^2S_{\frac{1}{2}}$ level relative to the $2^2P_{\frac{1}{2}}$ level. The first measurement of this interval (later named the Lamb shift) by Lamb, using direct rf resonance, clearly proved that these levels were not degenerate but separated by ~ 1058 MHz. This is small even in comparison to the 10969 Mhz

¹W.E. Lamb and R.C. Retherford, Phys. Rev. 72, 241 (1947).

²S. Pasternack, Phys. Rev. 54, 1113 (1938).

splitting of the P states for the $n = 2$ level of hydrogen. Many subsequent measurements of this interval have been made with the state-of-the-art measurement being 1057.845(9) MHz using the method of separated oscillatory fields on a fast atomic beam.³ The best theoretical prediction⁴ for the $2^2S_{\frac{1}{2}} - 2^2P_{\frac{1}{2}}$ interval is 1057.929(13) MHz which is not in good agreement with experiment. The hydrogenic Lamb shift scales approximately as $\frac{Z^4}{n^3}$, (where Z and n are respectively the atomic and principle quantum number), and precision measurements⁵⁻⁶ have been made by many different techniques in hydrogenic systems up to $Z = 18$.

Within the formal framework of QED, the Lamb shift is explained in terms of electron self-energy and vacuum polarization. The interaction energy between the electron and its own electromagnetic field is called the electron self-energy. The very presence of an electron (whether bound or free) implies an accompanying electromagnetic field which can react on the electron itself. Within QED, this process is described by an electron emitting a virtual photon and subsequently reabsorbing it. Since there is no way of turning off the interaction of the electron with its own electromagnetic field, the self-energy is an inseparable part of the observable rest energy of the electron. One can measure the difference of the bound-electron self-energy and the free-electron self-energy or the difference of the bound-electron self-energies for two distinct bound states. For electrons, this difference is essentially the Lamb shift (neglecting the much smaller vacuum polarization correction). H.A. Bethe, in 1947, estimated the electron

³S.R. Lundeen and F.M. Pipkin, Phys. Rev. Lett. 46, 232 (1981).

⁴P.J. Mohr, Phys. Rev. Lett. 34, 1050 (1975).

⁵G.W.F. Drake, Adv. At. Mol. Phys. 18, 399 (1982).

⁶H.W. Kugel and D.E. Murnick, Rep. Prog. Phys. 40, 297 (1977).

self-energy (E_{SE}) using Kramer's idea of mass renormalization as⁷

$$E_{SE} = \frac{8}{3\pi} \left[\frac{e^2}{4\pi\hbar c} \right]^3 \left[\frac{e^2}{8\pi a_0} \right] \frac{1}{n^3} \ln \left(\frac{E_\gamma^{(\max)}}{\langle E_I - E_{2^2S_{\frac{1}{2}}} \rangle_{ave}} \right).$$

The quantity $\langle E_I - E_{2^2S_{\frac{1}{2}}} \rangle_{ave}$ is the average excitation energy of the $2^2S_{\frac{1}{2}}$ state and $E_\gamma^{(\max)}$ is the maximum excitation (cut-off) energy which was chosen to be mc^2 to allow for the breakdown of his approach in the relativistic limit. The logarithm involving the average excitation energy is now known as the Bethe logarithm. Bethe's evaluation of this expression produced a shift of 1040 MHz for the $2^2S_{\frac{1}{2}}$ state.

Shortly after Lamb's original measurement, T.A. Welton in 1948 gave a qualitative explanation of the electron self-energy contribution to the Lamb shift in terms of a smeared electronic charge.⁸ He reasoned that since a quantized radiation field has a zero-point energy determined by the mean-square electric and magnetic fields, there are fluctuations in the vacuum radiation field that cause the electron to execute oscillatory motion smearing out its charge. For an electron bound in a non-uniform electric field such as that of the hydrogen nucleus, the potential experienced by the electron will differ from that appropriate to its mean position. In Welton's theory, only s -states suffer a shift; however in reality, states with $l \geq 1$ are also affected to a much smaller degree. This effect is a maximum for s -electrons through a modification of $|\Psi(0)|^2$ since the gradient of the potential is a maximum near the nucleus and $|\Psi(0)|^2 = 0$ for non- s -electrons.

A smaller but still significant contribution to the Lamb shift comes from vacuum polarization. A virtual electron-positron pair created in the Coulomb field of a nucleus behaves in such a way that the virtual electron is attracted to the

⁷H.A. Bethe, Phys. Rev. 72, 339 (1947).

⁸T.A. Welton, Phys. Rev. 74, 1157 (1948).

nucleus while the virtual positron is repelled. The result is that the nuclear charge observed at distances larger than the nuclear radius is less than the bare charge of the nucleus due to shielding of the nuclear charge by virtual electrons. In other words, the vacuum behaves like a polarizable medium due to the virtual electron-positron pairs. Since s -state electrons have a greater probability of penetrating the nucleus they are shifted to lower energy relative to other states due to this effect. In 1935, E.A. Uehling predicted this shift of the $2^2S_{\frac{1}{2}}$ level relative to the $2^2P_{\frac{1}{2}}$ level to be -27 MHz. The observed Lamb shift has the opposite sign and is about 40 times larger in magnitude since the majority of the effect is due to the electron self-energy.

1.2 Two-Electron Systems

The theory for calculating energy structure in two-electron atoms becomes more complicated because electron correlation effects must be included. The electron energy levels in heliumlike ions are also sensitive to radiative effects. The largest radiative contribution to heliumlike energy structure is from the one-electron Lamb shift although two-electron Lamb shift corrections are also important. It was recognized some years ago that the $1s2s - 1s2p$ transitions in heliumlike ions have attractive experimental properties:

- the transition frequency increases slowly with Z ($\sim Z$) so that the wavelengths of excitation for resonance experiments for $Z \leq 10$ are available from narrow-band coherent sources;
- the transition from the $1s2p^3P$ levels to the ground state is strongly suppressed, leading to a natural linewidth for the $1s2s^3S - 1s2p^3P$ transitions which is much smaller than that for hydrogenic Lamb shift transitions and scales approximately as Z ;
- the relative contribution to the total energy of relativistic and QED effects grows rapidly with Z (Z^3);

- as in hydrogenic ions, the lower level of the transition is metastable, allowing clean, low background measurements with mass-selected beams.

There is therefore a range of heliumlike ions where the wavelengths involved permit spectroscopic measurements of high precision on transitions with significant relativistic and QED contributions. In fact the ratio of the QED portion to the experimental linewidth is actually better than for the hydrogenic $n = 2$ Lamb shift. In the case of Li II, where laser spectroscopy has been used⁹⁻¹⁰, this ratio is ~ 350 , as compared with ~ 10 in hydrogen.

There have been experimental measurements of the $1s2s^3S_1 - 1s2p^3P_1$ and $1s2s^1S_0 - 1s2p^1P_1$ transition wavelengths, over a wide range of Z providing important checks on theoretical calculations. Excited two-electron ions have been produced by several different techniques including spark sources,¹¹ tokamaks,¹² beam-foil stripping,¹³ recoil ion sources,¹⁴ electron-beam ion sources (EBIS),¹⁵ and electron cyclotron ion sources (ECRIS)¹⁶ for spectroscopy.

For the low Z region, ions are produced either in a modest ion source¹⁷ or a

⁹R.A. Holt, S.D. Rosner, T.D. Gaily, and A.G. Adam, *Phys. Rev. A* **22**, 1563 (1980).

¹⁰E. Riis, H.G. Berry, O. Poulsen, S.A. Lee, and S.Y. Tang, *Phys. Rev. A* **33**, 3023 (1986).

¹¹B. Löfstrand, *Physica Scripta* **8**, 57 (1973).

¹²M.F. Stamp, I.A. Armour, N.J. Peacock, and J.D. Silver, *J. Phys. B* **14**, 3551 (1981).

¹³J. Désesquelles, *Nucl. Instrum. Methods B* **31**, 30 (1988).

¹⁴H.A. Klein, S. Bashkin, B.P. Duval, F. Moscatelli, J.D. Silver, H.F. Beyer, and F. Folkmann, *J. Phys. B* **15**, 4507 (1982).

¹⁵R.E. Marrs, M.A. Levine, D.A. Knapp, and J.R. Henderson, *Phys. Rev. Lett.* **60**, 1715 (1988).

¹⁶R.W. Dunford *et al.* in *Proceedings of the International Conference on ECR Ion Sources and Their Applications*, NSCL Report #MSUCP-47, East Lansing, MI, 1987, p. 573.

¹⁷R.A. Holt, S.D. Rosner, T.D. Gaily, and A.G. Adam, *Rev. Sci. Instrum.* **52**, 157 (1981).

spark source. Wavelengths are measured by using either a UV emission spectroscopy or to higher accuracy using direct resonance techniques.¹⁸ Ions with $Z \geq 10$ are usually produced by beam-foil stripping, tokamaks, or recoil ion sources and wavelengths measured with VUV and X-ray spectrometers. These techniques have produced a 700 ppm measurement of the $1s2s^3S_1 - 1s2p^3P_2$ transition in Kr xxxv¹⁹ which is equivalent to a $\sim 5\%$ measurement of the Lamb shift. This is the highest Z measurement of a $2^3S - 2^3P$ transition to date. At more modest Z , Berry *et al.* have measured two transitions in Cl xvi ($2^3S_1 - 2^3P_0$ and $2^3S_1 - 2^3P_2$) to 100 ppm and 20 ppm which, assuming exact relativistic calculations, are a test of the radiative corrections to $\sim 2.9\%$ and $\sim 0.6\%$ respectively.²⁰ In neutral helium, saturated absorption spectroscopy has been used²¹ to measure the $1s2p^3P_J - 1s3d^3D_J$ transitions to 8 parts in 10^9 . Using a two-photon measurement of the $2^3S_1 - 3^3D_J$ intervals,²² the $2^3S_1 - 2^3P_J$ intervals were determined to 0.05 ppm and the Lamb shift to $\sim 0.3\%$. Perhaps the most accurate measurement of both the absolute wavelength of and the Lamb shift contribution to a $2^3S_1 - 2^3P_J$ interval is a laser fluorescence measurement in $^7\text{Li II}$.²³ The absolute wavelengths were determined to 0.01 ppm and the QED corrections to $\sim 0.015\%$. These results are a factor of 50 times more precise than the best theoretical predictions. An even more precise experiment with the same atomic system has been performed but the results are not

¹⁸R. A. Holt, S. D. Rosner, T. D. Gaily, and A. G. Adam, Phys. Rev. A 22, 1563 (1980).

¹⁹S. Martin, J. P. Buchet, M. C. Buchet-Poulizac, A. Denis, M. Druetta, J. Désesquelles, J. P. Grandin, D. Hennecart, X. Husson, D. Lecler, and I. Lesteven, Phys. Rev. A 35, 2327 (1987).

²⁰H. G. Berry, A. DeSerio, and A. E. Livingston, Phys. Rev. A 22, 998 (1980).

²¹P. Juncar, H. G. Berry, R. Damaschini, and H. T. Duong, J. Phys. B: At. Mol. Phys. 16, 381 (1983).

²²E. Giacobino and F. Biraben, J. Phys. B: At. Mol. Phys. 15, 385 (1982).

²³E. Riis, H. G. Berry, O. Poulsen, S. A. Lee, and S. Y. Tang, Phys. Rev. A 33, 3023 (1986).

yet published.

A comparison of experimental measurements of the $1s2s^3S_1 - 1s2p^3P_J$ transitions with theoretical calculations for $Z \leq 26$ has been made by Goldman and Drake.²⁴ The theoretical error estimate is obtained from the uncalculated terms which represent the combined effects of electron correlation and higher-order relativistic corrections which are of order $(\alpha^4 Z^4)$ in the perturbative expansions in powers of (αZ) and Z^{-1} used to calculate two-electron energy structure. These terms scale approximately as $\pm 1.2(Z/10)^4 \text{ cm}^{-1}$. The theory and experiment agree within the specified uncertainties except for a few measurements. The precision of experiment and theory is comparable for $Z \geq 4$ but for $Z < 4$ the experiments are at least a factor of 10 more precise. Since QED calculations of the radiative corrections are carried out perturbatively, it is important to test the methods of calculation as precisely as possible for a wide range of Z . Experiments at higher Z are stringent tests of the perturbative expansions in powers of (αZ) for radiative corrections where they contribute a large fraction of the $1s2s - 1s2p$ interval. High precision resonance measurements at low Z , where electron correlation effects are more important, check perturbative expansions of $\frac{1}{r_{12}}$ (r_{12} is the electron separation) involving powers of $\frac{1}{Z}$.

²⁴S.P. Goldman and G.W.F. Drake, J. Phys. B: At. Mol. Phys. **17**, L197 (1984).

CHAPTER 2

Theoretical Background

Calculations for two-electron atoms and ions are more difficult than for one-electron systems because one must consistently include relativistic and QED effects as well as electron-electron correlations. Unfortunately, a relativistic two-electron Hamiltonian including QED effects to all orders cannot be uniquely defined. The rigorous starting point for many of these calculations is the fully covariant Bethe-Salpeter equation derived by Bethe and Salpeter from the Feynman form of QED²⁵ and from quantum field theory by Gell-Mann and Low.²⁶ No exact solution to this equation has been found but calculations based on expansions in powers of α , αZ , and Z^{-1} have been made.

In two-electron systems, no QED effects are directly measurable as is the $2s_{\frac{1}{2}} - 2p_{\frac{1}{2}}$ splitting in hydrogenic systems. Measurements of the $n = 2$, $\Delta n = 0$ ($1s2s^1S_0 - 1s2p^1P_1$ and $1s2s^3S_1 - 1s2p^3P_1$) transitions in highly stripped ions have spurred interest in higher order QED corrections. Very simply, calculations of the non-relativistic two-electron energies (including electron correlations) and relativistic corrections are subtracted from the measured energies. The resulting QED remainder is then compared to theoretical QED corrections for those particular states of that ionic system. The $n = 2$, $\Delta n = 0$ intervals are extremely attractive experimentally since the Lamb shift contribution (ΔE_{LS}) to the interval increases dramatically as Z^4 while the non-relativistic energy (ΔE_{NR}) scales only as

²⁵E.E. Salpeter and H.A. Bethe, Phys. Rev. **84**, 1232 (1951).

²⁶M. Gell-Mann and F. Low, Phys. Rev. **84**, 350 (1951).

$\sim Z$. The ratio of the two is given approximately as²⁷

$$\frac{\Delta E_{LS}}{\Delta E_{NR}} \approx 1.4 \times 10^{-6} Z^3.$$

Another advantage is that for $Z \leq 10$, the wavelengths of these intervals can be produced by narrow-band coherent light sources so that very precise resonance measurements can be made.

Calculations of energy levels use different approximations in the low and high Z regions. For low Z systems highly accurate solutions to the non-relativistic Schrödinger equation are corrected by including relativistic effects as perturbations. This is accomplished by solving the Breit equation which can be considered as a relativistic extension of the Schrödinger equation. The Breit equation is similar in nature to the Dirac equation for one electron but, unlike the Dirac equation, it is not fully Lorentz invariant. A further low- Z approximation (Pauli approximation) to a simplified form of the Breit equation produces a Hamiltonian, $\mathcal{H}$, defined as

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_3 + \mathcal{H}_4 + \mathcal{H}_5 + \mathcal{H}_6$$

which describes all non-QED energy contributions. The zeroth order Hamiltonian, $\mathcal{H}_0$, is the non-relativistic Schrödinger Hamiltonian for two bound electrons written as

$$\mathcal{H}_0 = -\frac{\hbar^2}{2m} (\nabla_1^2 + \nabla_2^2) - \frac{Ze^2}{r_1} - \frac{Ze^2}{r_2} + \frac{e^2}{r_{12}}$$

where

r_1, r_2 are the coordinates of electrons 1 and 2 respectively and

r_{12} is defined as $|\mathbf{r}_1 - \mathbf{r}_2|$.

The sum of Hamiltonians $\mathcal{H}_1$ through $\mathcal{H}_6$ is known as the Breit-Pauli interaction. A physical description of the components of this interaction is given below.

$\mathcal{H}_1$ is the relativistic correction to the electron mass.

²⁷G.W.F. Drake in *Atomic Physics 8*, (Plenum, New York, 1983), p. 185.

$\mathcal{H}_2$ corresponds to the classical relativistic correction to the interaction between the electrons which is due to the retardation of the electromagnetic fields produced by the electrons.

$\mathcal{H}_3$ is the interaction between the spin magnetic moment and orbital magnetic moment of the electrons (spin-orbit coupling).

$\mathcal{H}_4$ is the Darwin term which has no classical analog; it arises from the Dirac Hamiltonian and can be explained in terms of *Zitterbewegung*.

$\mathcal{H}_5$ represents the interaction of the spin magnetic dipole moments of the two electrons.

$\mathcal{H}_6$ is the interaction with external fields (if any).

QED corrections to the energy of a given state are included using perturbation theory.

For higher Z , where these perturbations are no longer small, better results can be obtained by using the exact solutions to the one-electron Dirac Hamiltonian and then using perturbation theory to include electron-electron interactions and finally higher order radiative corrections (QED). This approach to these calculations leads to a Hamiltonian as follows:²⁸

$$\mathcal{H}_0 = \mathcal{H}_D(1) + \mathcal{H}_D(2) + V_{12} + B$$

where

$$\mathcal{H}_D(i) = c \boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta mc^2 - \frac{Ze^2}{r_i}$$

$$V_{12} = \frac{e^2}{r_{12}}$$

and

$\boldsymbol{\alpha}_i$ and β are the 4×4 Dirac matrices, B is the Breit operator representing the retarded Coulomb and magnetic interactions between two electrons.

Once again, QED corrections to the energy of a given state are included using

²⁸J.D. Bjorken and S.D. Drell, *Relativistic Quantum Mechanics*, (McGraw-Hill, New York, 1964), Chapter 4.

perturbation theory.

Another approach to these calculations is to perform a double perturbation expansion in powers of Z and Z^{-1} .²⁹⁻³⁰ This expansion can be written as

$$E = Z^2 \sum_n \sum_m E_n^{2m} Z^{-n} (Z\alpha)^{2m}$$

where the summation over n and m is from zero to infinity.

When the expansion is written as follows:

$$\begin{aligned} E = & \quad Z^2 E_0^0 + \quad \alpha^2 Z^4 E_0^2 + \quad \alpha^4 Z^6 E_0^4 + \dots \\ & + \quad Z^1 E_1^0 + \quad \alpha^2 Z^3 E_1^2 + \quad \alpha^4 Z^5 E_1^4 + \dots \\ & + \quad Z^0 E_2^0 + \quad \alpha^2 Z^2 E_2^2 + \dots \\ & + \quad Z^{-1} E_3^0 + \quad \alpha^2 Z^1 E_3^2 + \dots \\ & + \dots \end{aligned}$$

the first column sum is the exact non-relativistic energy (Coulomb interaction), and the second column sum gives all relativistic and Breit corrections of relative order α^2 (α is the fine structure constant). The Coulombic contribution has been calculated by a variational technique by Accad *et al.* as well as by summing the relativistic and Breit terms of the second column.³¹ The sum of the first row is the exact single-particle Dirac energies for two independent electrons and the sum of the second row represents the one-photon exchange term of the Breit interaction to all orders in $\alpha^2 Z^2$. The sum of the third row represents the two-photon exchange term of the Breit interaction to all orders in $\alpha^2 Z^2$.

A high precision variational calculation of the $1s^2 \ ^1S$ state of H^- and the $1s^2 \ ^1S$, $1s2s \ ^1S$, and $1s2s \ ^3S$ states of He has been made by Drake³² where the non-relativistic energy is calculated to a few parts in 10^{13} . This improved

²⁹A. Dalgarno and A.L. Stewart, Proc. Roy. Soc. (London) 75, 441 (1960).

³⁰H.T. Dcyle, Adv. At. Mol. Phys. 5, 377 (1969).

³¹Y. Accad, C.L. Pekeris, and B. Schiff, Phys. Rev. A 4, 516 (1971).

³²G.W.F. Drake, Nucl. Instrum. Methods B 31, 7 (1988).

variational calculation for two-electron systems uses a Hylleraas-type correlated basis set which is doubled by including powers of r_1 , r_2 , and r_{12} ($r_{12} = |r_1 - r_2|$ where r_1 and r_2 are the coordinates of electrons 1 and 2) twice with different exponential nonlinear parameters, along with the exact screened hydrogenic wave function. This basis set has been used to calculate the non-relativistic energy including second-order mass polarization effects with the remaining corrections (relativistic, relativistic recoil Lamb shift and finite nuclear size effects) added as perturbations. There is an almost constant discrepancy of $0.00048(6) \text{ cm}^{-1}$ between theory and experiment which has been attributed to the incomplete state of two-electron Lamb shift calculations. This technique has been extended to higher Z systems,³³ and produces the most accurate theoretical calculations for $2^1S - 2^1P$ and $2^3S - 2^3P$ intervals in heliumlike ions.

The best calculation of two-electron Lamb shifts for $2^3S - 2^3P$ (as well as $2^1S - 2^1P$) transitions in helium-like ions is by Goldman and Drake using a finite basis set.³⁴ They have calculated the leading terms in the $1/Z$ expansion of the two-electron Bethe logarithm for $1s^2 \ ^1S_1$, $1s2s \ ^1S_0$, $1s2s^3S_1$, $1s2p \ ^1P_1$, and $1s2p^3P_J$ states in order to determine the two-electron Lamb shift and hence, the $1s2s^3S_1 - 1s2p^3P_J$ transitions of heliumlike ions for $Z \leq 26$. Calculations of the $1s2s^3S_1 - 1s2p^3P_J$ energy levels are carried out by diagonalizing the following Hamiltonian (Goldman and Drake):

$$\mathcal{H} = (H_{NR} + B_P + E_{L,2} + H_M + H_{NS})_{LS} + R(H_D + V_{12} + B + E_{HO})_{jj} R^{-1} - \Delta$$

where

H_{NR} is the non-relativistic Hamiltonian ($\mathcal{H}_0$)

B_P is the Breit-Fauli interaction ($\mathcal{H}_1 - \mathcal{H}_0$)

$E_{L,2}$ is the two-electron Lamb shift correction

³³G.W.F. Drake, to be published.

³⁴S.P. Goldman and G.W.F. Drake, J. Phys. B **17**, L197 (1984).

H_M is the mass polarization correction³⁵

H_{NS} is the nuclear size correction³⁶

H_D is the sum of the one-electron Dirac Hamiltonians

$V_{12} = \frac{e^2}{r_{12}}$ is the electron-electron interaction

B is the Breit interaction including retardation³⁷

E_{HO} are higher order and one-electron QED corrections.³⁸

R is a unitary transformation from a jj -coupling basis set to the LS -coupling basis set. The E_{HO} term is corrected for the non-hydrogenic electron density at the nucleus. Terms which are counted twice such as the uncorrelated non-relativistic energy and the uncorrelated non-relativistic Breit-Pauli energy are subtracted in the quantity Δ .

To lowest order in an expansion in α , the two-electron Lamb shift correction is given by³⁹ (atomic units, with $1 \text{ au} = \alpha^2 mc^2$)

$$E_{L,2} = \frac{4}{3} Z \alpha^3 \left[\ln(Z\alpha)^{-2} + \ln[Z^2 R_\infty / \varepsilon(nLS)] + \frac{19}{30} \right] \langle \delta^3(r_1) + \delta^3(r_2) \rangle$$

where

R_∞ (Rydberg constant) = $\alpha/2 \text{ au}$ or 13.6 eV

$\langle \delta^3(r_i) \rangle$ is the matrix element of the wavefunction for the i th electron at the origin

$\varepsilon(nLS)$ is defined in terms of the two-electron Bethe logarithm.

The two-electron Bethe logarithm is defined by

$$\ln \varepsilon(nLS) = \frac{\sum_m |\langle \psi_0 | t | \psi_m \rangle|^2 \ln(E_m - E_0) / (E_m - E_0)}{\sum_m |\langle \psi_0 | t | \psi_m \rangle|^2 / (E_m - E_0)}$$

³⁵B.H. Bransden and C.J. Joachain, *Physics of Atoms and Molecules*, (Longman, London, 1983), Appendix 8.

³⁶P.J. Mohr, *At. Data Nucl. Data Tables* **29**, 453 (1983).

³⁷M.H. Mittleman, *Phys. Rev. A* **4**, 897 (1971).

³⁸P.J. Mohr, *Phys. Rev. A* **26**, 2338 (1982).

³⁹P.K. Kabir and E.E. Salpeter, *Phys. Rev.* **108**, 1256 (1957).

where

ψ_0 is the wavefunction for the nLS two-electron configuration with energy E_0

$$t = Z \sum_i \frac{r_i}{r_i^3} \text{ summed over all intermediate states.}$$

The summation over m extends over all bound states and leads to non-convergent results using standard variational basis sets for evaluation because of the large contribution from highly excited states. Goldman and Drake rewrite the two-electron Bethe logarithm as a ratio, expanding the numerator and denominator in powers of $1/Z$ as follows:

$$\ln \varepsilon(\text{nLS}) = A/B$$

$$A = Z^4[A_0 + A_1 Z^{-1} + 2(\ln Z)(B_0 + B_1 Z^{-1}) + \dots]$$

$$B = Z^4[B_0 + B_1 Z^{-1} + \dots].$$

The coefficients in the expansion of B can be obtained from the expression

$$B = 2\pi Z \langle \delta^2(r_1) + \delta^2(r_2) \rangle.$$

The value for A_0 can be determined from the condition $A_0/B_0 = \ln \varepsilon_0(\text{nLS})$ where $\ln \varepsilon_0(\text{nLS})$ is the lowest-order hydrogenic approximation for the two-electron Bethe logarithm defined by

$$\ln \varepsilon_0(\text{nLS}) = (1 + \delta_{L,0}/n^3)^{-1} (\ln \varepsilon_0(1s) + n^{-3} \ln \varepsilon_0(nL))$$

and $\ln \varepsilon_0(nL)$ is the hydrogenic Bethe logarithm for nuclear charge $Z = 1$. The quantity A_1 , defined in terms of a nearly divergent perturbation sum,⁴⁰⁻⁴¹ requires extensive calculation.⁴² Using the definitions for A and B , the two-electron Bethe logarithm is written

$$\begin{aligned} \ln(\varepsilon(\text{nLS})/R_\infty) &= A_0/B_0 + \ln 2 + 2\ln Z \\ &+ [(A_1 B_0 - A_0 B_1)/B_0^2] Z^{-1} + O(Z^{-2}) \\ &= \ln[\varepsilon_0(\text{nLS})(Z-\sigma)^2] + O(Z^{-2}) \end{aligned}$$

⁴⁰S.P. Goldman and G.W.F. Drake, J. Phys. B **16**, L183 (1983).

⁴¹A.M. Ermolaev and R.A. Swainson, J. Phys. B **16**, L35 (1983).

⁴²S.P. Goldman, Phys. Rev. A **30**, 1219 (1984).

where $\sigma = -(A_1 B_0 - A_0 B_1)/(2B_0^2)$ is a screening constant. The results for the two-electron Bethe logarithm for the ground state and first few excited states are

$$\ln(\epsilon(1^1S)/R_\infty) = \ln[19.7693(Z - 0.00615)^2]$$

$$\ln(\epsilon(2^1S)/R_\infty) = \ln[19.3943(Z - 0.02040)^2]$$

$$\ln(\epsilon(2^3S)/R_\infty) = \ln[19.3943(Z - 0.01388)^2]$$

$$\ln(\epsilon(2^1P)/R_\infty) = \ln[19.6952(Z - 0.00600)^2]$$

$$\ln(\epsilon(2^3P)/R_\infty) = \ln[19.6952(Z - 0.00475)^2]$$

It is interesting to note that the results for the two-electron Bethe logarithm do not differ very much from the hydrogenic case ($\sigma = 0$).

For the $1s2s^1S_0 - 1s2p^1P_1$ interval in heliumlike beryllium the breakdown of contributions to the total energy difference is⁴³

<u>Contribution</u>	<u>Energy (cm⁻¹)</u>
non-relativistic	16262.580
relativistic	9.211
mass polarization	7.850
QED	-2.868
nuclear structure	-0.006
<i>Total</i>	16276.766(30).

The Breit-Pauli interaction is included in the relativistic contribution. The QED contribution includes both two-electron Lamb shift corrections ($\sim 96.4\%$) and higher order and one-electron QED effects ($\sim 3.6\%$). The 0.030 cm^{-1} theoretical uncertainty represents uncalculated relativistic contributions of order ($\alpha^4 Z^4$).

⁴³G.W.F. Drake, Can. J. Phys. **66**, 586 (1988).

CHAPTER 3

Experimental Technique

3.1 Comparison of Techniques

Gas cell spectroscopy, performed by observing the transmission of light from a tunable dye laser through a sample gas or plasma, is a standard spectroscopic technique. Although the experimental procedure is rather uncomplicated and can produce a great deal of data, it has severe limitations that are a result of this simplicity. For example, the precision of the measurement is limited in most cases by Doppler broadening which can be rather large but may be reduced by saturated absorption spectroscopy at the expense of decreased signal. For systems which require high temperatures to provide a significant vapour pressure of gas or a strong discharge to produce ionic species, this further increases the Doppler broadening. Furthermore, the system may not be easily prepared in any highly excited state since these states are quenched by collisions.

Some of these limitations can be overcome by performing spectroscopy on a beam of ions. One efficient way of producing intense beams of ions is by beam foil stripping.⁴⁴ This technique allows the observation of many spectral lines of highly excited ions. The Doppler spread in the resulting ion beam is rather large, reducing resolution, and the broad lines may be blended with lines from spectra of other ionic species. Another problem which complicates data analysis is the large Doppler shift which must be eliminated from the observed wavelength. The Doppler shifts must be calibrated using previously measured lines which are present in the spectra and are unblended. A similar means of ion production is the recoil ion source where the ions produced by ion bombardment of the gas target are accelerated out of the

⁴⁴H.J. Andrä, *Physica Scripta* 9, 257 (1974).

source to form a beam of highly stripped ions which can be mass selected for spectroscopy.⁴⁵ The ensuing Doppler shifts are smaller and can be calculated from the accelerating potential or eliminated entirely by observing the spectrum directly in the recoil ion source.

Production of the ions in an ion source allows the preparation of mass and charge selected beams of ions in highly excited states and modest charge states. This of course virtually eliminates the possibility of obtaining spectra where the lines of interest are blended with lines from other systems. The Doppler spread, determined primarily by the velocity distribution of the ions in the region of the ion source where they are extracted, can be narrowed by decreasing the diameter of the source aperture and hence the volume of this region. A further narrowing can be accomplished by kinematic compression⁴⁶ of the Doppler velocity distribution arising from the acceleration process. The associated Doppler shift is usually not as large as in beam foil spectroscopy and can be eliminated by performing laser resonance spectroscopy with the laser and ion beams both counter-propagating and co-propagating. The two measured Doppler-shifted wavelengths can then be used to eliminate the Doppler shift without knowing the ion velocity.

Thus in general, the method of laser-induced fluorescence with fast-ion beams has the following experimental advantages:

- well-determined ion beam energy and Doppler shift;
- narrow ion beam Doppler profiles due to velocity compression;
- mass and charge state selection producing clean uncontaminated spectra;
- direct resonance measurement of energy intervals allowing unambiguous comparison of energy intervals with an external standard.

As a rule, direct resonance techniques produce the most accurate measurements of

⁴⁵H.G. Beyer, F. Folkmann, and K.H. Scharfner, Z. Phys. D **1**, 65 (1986).

⁴⁶S.L. Kaufman, Opt. Commun. **17**, 309 (1976)

atomic and molecular energy structure.

3.2 General Description of the Experiment

The technique of laser resonance spectroscopy uses the nearly monochromatic light of a laser to transfer the population in a two level system from the lower state which must be metastable to some upper state which is relatively short lived. The two level system which is of interest in this work is illustrated in Figure 1. Since the metastable $1s2s^1S_0$ level has angular momentum $J = 0$, it has no hyperfine structure. The hyperfine structure of the excited $1s2p^1P_1$ which is discussed in Appendix 1 is very small (less than 250 MHz) and cannot be resolved since the observed linewidth in this experiment, determined mainly by the ~ 8.2 ps lifetime of the upper state, is 22.5 GHz. The result of these two factors is that only one resonance can be observed as the laser drives electric dipole transitions between these levels. Once populated, the $1s2p^1P_1$ level can decay back to the metastable level or, with much greater probability, to the $1s^2\ ^1S_0$ ground state.

The experimental geometry, illustrated schematically in Figure 2, is chosen so that the laser and ion beams are collinear and overlapped in the region of the post-accelerator. Thus the laser light, as seen in the rest frame of the ion, is either red or blue Doppler-shifted depending whether the light is co- or counter-propagating. A laser resonance can be observed when the laser wavelength is tuned to resonance with the singlet transition for a particular ion velocity, transferring some population from the 2^1S_0 metastable level to the excited 2^1P_1 level. Any population in the 2^1P_1 state immediately decays to the 1^1S_0 ground state emitting a soft x-ray (~ 10 nm) which is recorded. Since the chosen geometry is collinear, it is necessary to restrict the interaction of the laser and the ion beam to the region directly in front of the microchannel plate used to detect the fluorescence. This is accomplished by increasing the ion velocity in the resonance region (post-accelerator) so that the laser resonance can occur only with this further

Figure 1. *Energy level diagram for heliumlike beryllium.*

The metastable $1s2s\ ^1S_0$ level is excited by 614.1 nm radiation from a tunable dye laser to the $1s2p\ ^1P_1$ level. This level decays with an 8.20 ps lifetime (theoretical) predominantly to the $1s^2\ ^1S_0$ ground state emitting a 10.0 nm photon which is detected by a microchannel plate array.

Figure 2. *Schematic layout of the experimental apparatus (not to scale).*

The ion beam travels from the source through the resonance region to the beam monitor in a vacuum of $< 10^{-7}$ Torr. The laser beam enters and exits the vacuum via Brewster windows (not shown) on both sides of the resonance region. The numbers 1 – 5 refer to detector signals input to the computer, while 0 refers to the computer-generated staircase ramp used to scan the dye laser.

Doppler shift.

The precision obtained by the laser resonance technique is determined by the width of the observed resonance which in most circumstances is a result of the angular divergence of the ion beam as well as the spread in ion energy. For this experiment the contribution to the experimental uncertainty from angular divergence is only 0.6 MHz/mrad while the velocity distribution contributes less than 60 MHz/V. These uncertainties are of little consequence when compared with the 19.4 GHz linewidth due to the lifetime of the 2^1P_0 state. The usual method of scanning across the laser resonance is to fix the laser frequency and change the ion velocity by scanning the voltage on the resonance region. Since this would necessitate a large voltage scan which is a significant fraction of the ion energy, systematic errors could occur as a result of ion focusing and deflection by the large electric fields from the accelerating potentials placed on the resonance region. To overcome this experimental problem, the voltage on the resonance region is fixed for a particular run while the laser frequency is scanned across the resonance. Since a laser does not scan linearly, it then becomes necessary to measure the wavelength of emitted light as the laser scans across the resonance. This is accomplished by picking off a small fraction of light from the primary laser beam and monitoring its transmission through an iodine absorption cell for absolute wavelength calibration as well as a Fabry-Perot étalon for interpolation between iodine calibration lines. Furthermore, to correct for any asymmetries in the observed lineshape which might arise from fluctuations in the total ion current or laser power during the acquisition of a spectrum, both the ion current and laser power are recorded during the scan. Thus, any distortion of the lineshape by drifts in these signals may be corrected during data analysis.

CHAPTER 4

Experimental Apparatus

4.1 Vacuum System

The ion beam apparatus, which was first assembled to perform a similar measurement in lithium,⁴⁷ is a vacuum system approximately 3.5 m long consisting of a high temperature ion source operating at 10 kV, a Wien velocity filter for mass selection, and an interaction chamber where the ion and laser beams are overlapped. The vacuum tanks, drift tubes and flanges are fabricated almost entirely from stainless steel. The vacuum is achieved by two 10 cm (4 in.) Varian VHS diffusion pumps and one 5 cm (2 in.) diffusion pump. All three of these pumps are water baffled to reduce oil back-streaming. One of the 10 cm diffusion pumps is mounted near the ion source to pump any gas which evolves from the ion source during operation. The second 10 cm pump, which is also fitted with a cold trap to further prevent back-streaming, evacuates the interaction chamber. The drift tube joining the velocity selector and the interaction region is evacuated by the 5 cm pump.

Since ultra-high vacuum is not necessary, the vacuum system is assembled with Viton O-rings. The ultimate pressure obtained by the pumps is $\sim 2 \times 10^{-8}$ Torr while the system is idle and after the ion source has been baked out to remove any gas and water vapour which is absorbed in the boron nitride insulators. While the source is operating, the system pressure as measured after the Wien filter and above the 5 cm pump rises to $\sim 8 \times 10^{-8}$ Torr as a result of outgassing of the boron nitride insulators in the ion source. The amount of evolved gas depends on the source temperature and the number of times it has been heated. A low operating pressure in the source is necessary since metastables are easily de-excited by

⁴⁷R.A. Holt, S.D. Rosner, T.D. Gaily, and A.G. Adam, Phys. Rev. A 22, 1563 (1980).

collision and multiply charged ions are more readily produced in low pressure discharges. The diffusion pumps are backed by a Sargent Welch rotary pump providing a backing pressure better than 50 mTorr. A foreline trap filled with activated alumina prevents contamination of the diffusion pump oil with rotary pump oil.

The rather long ion beam apparatus allows precise overlap of the ions and laser over 70 cm. The ions are focused by an Einzel lens immediately after they are extracted from the source. A set of deflection plates can be used to steer the beam vertically while the Wien filter provides mass selection as well as horizontal control. A shielded Faraday cup on a sliding piston can be lowered into the beam to measure ion beam current just after the Wien filter, for comparison with the current measured at the beam dump at the end of the apparatus. Ion currents of Be^+ in excess of $20 \mu\text{A}$ have been measured on this Faraday cup but due to space charge limits only 7 to $8 \mu\text{A}$ have been detected at the beam dump. The mass analyzed ion beam travels through a 1 m long drift tube where it can be further focused by a second Einzel lens before entering the interaction chamber where a set of steerage plates bend the beam horizontally through 5° to overlap the laser beam. A second set of steerage plates is used to make small final vertical corrections in the ion trajectory before the resonance region. Immediately after the resonance region, a second shielded Faraday cup can be moved into the beam axis to measure the ion current passing through this region allowing comparison with the ion current measured at the beam dump at the end of the interaction chamber as well the extracted ion current at the first Faraday cup. For 10 kV Be^+ ions the calculated space charge limit for the ion current measured at the beam dump is $11.3 \mu\text{A}$ while the limit for Be^{++} is $16.0 \mu\text{A}$. Ion currents of Be^+ in excess of $8 \mu\text{A}$ have been observed from the Danfysik 911A ion source which are probably space charge limited but the 1–5 nA beams of Be^{++} are easily transported by the ion optics the

entire length of the apparatus.⁴⁸ The entire interaction region is surrounded by three orthogonal pairs of square Helmholtz coils to cancel the earth's magnetic field to less than 1×10^{-6} T (10 mG) in the vicinity of the region where the laser and ion beam interact.

4.2 Ion Source

The major experimental hurdle is the production of the metastable ions, which requires removal of two electrons from the beryllium atom while leaving a third electron in an excited state ~ 120 eV above the ground state. Initially, it was expected that a commercial hollow cathode ion source (Danfysik 911A Heavy Ion Source) would produce sufficient currents of the desired ion. For efficient metastable production, an arc voltage of several times 120 V is necessary to ensure excitation of the metastable state with a single electron impact. The arc voltage of the Danfysik 911A source is only 70 to 90 V, which is far short of the voltage necessary for metastable production. Since engineering modifications to the source electrodes failed to raise the arc voltage to 120 V, the ion source was highly modified to resemble more closely a Penning discharge ion source. The Danfysik 911A ion source was based on a design by Sidenius for use in electromagnetic isotope separation.⁴⁹ The source uses a helically wound tungsten filament which is directly heated by a DC current. The electrons emitted are accelerated from the cathode through the gas or vapour to the anode by a positive potential, forming ions which are extracted through the hollow cathode by the electric field of the extraction electrode. The magnetic field (< 150 Gauss) produced by the cathode filament is cancelled by the windings of an auxiliary magnet coil coaxially mounted outside the vacuum. As is the case with most ion sources, the 911A operates best when the feed

⁴⁸R. Hutter in *Focusing of Charged Particles Vol II*, edited by A. Septier, (Academic Press, New York, 1967), Chapter 3.1.

⁴⁹G. Sidenius in *Proc. Int. Conf. on Electromagnetic Isotope Separators*, Marburg, 1970, p. 423.

material is a gas. However, the source is designed to operate with any compound which has a vapour pressure greater than 10^{-2} Torr at temperatures less than 1700°C . The temperature of the feed material and hence its vapour pressure is varied in one of three ovens which can be independently heated over a particular temperature range.

Although this ion source proved to be incapable of producing the necessary ions, its basic construction, involving stacked cylindrical electrodes and insulators held in a slotted sleeve for electrical connection as well as its ability to handle feed materials requiring high temperatures, made it an excellent starting point for the design of the Penning discharge ion source. The Penning discharge ion source (or PIG source as it is more commonly known since its design resembles that of a Penning ionization gauge) consists of two cathodes placed at either end of a cylindrical hollow anode. Electrons which are produced either by emission at one of the cathodes (heated cathode design) or in the discharge itself (cold cathode design) are attracted to the anode but are constrained to oscillate between the cathodes in helical orbits by the application of a magnetic field along the axis of the anode. Versions of the PIG source exist whose design employs ion extraction through an aperture in the anode (side extraction) or, as in this design, through the cathode (end extraction). Previous studies of the properties of these sources reveal that in general side extraction sources produce ion beams with higher charge states and heated cathode sources produce ion beams with smaller energy spreads.⁵⁰

Due to its Danfysik 911A parentage, the PIG ion source is constructed with end extraction and employs a heated cathode. This permits the ion source to produce beams of modest charge states with energy spreads of less than 10 eV. Ion beams with narrow velocity distributions are very desirable for laser spectroscopy in

⁵⁰G.D. Alton, Nucl. Instrum. Methods 189, 15 (1981).

order to minimize the Doppler contribution to the linewidth. A cross sectional view of the oscillating electron bombardment (PIG) ion source is shown in Figure 3.

A new molybdenum sleeve, 2.5 cm longer than the original, was fabricated to hold the stack of ion source electrodes and insulators containing the new anticathode and longer anode. The original ion source housing was also abandoned since a higher magnetic field was required to constrain the electrons to oscillating helical trajectories and to house this longer ion source. At the same time, a water jacket was added between the magnet spool and the vacuum chamber enclosing the ion source to remove the heat radiated from the ion source as well as the heat dissipated in the magnet. The new magnet is capable of producing a field of ~ 0.09 T (875 G) in the vicinity of the anode. This constrains the radius of electron orbits to ~ 1 mm which is smaller than the 3.8 mm inner radius of the 20 mm long tantalum anode.

The anode is positioned between the cathode and anticathode by boron nitride insulators, providing a gap on either end of 3.5 mm between the electrodes. The Danfysik cathode, a helix of tungsten captured between tantalum end pieces for electrical connection, is employed at the extraction end of the ion source allowing extraction of the ions through the hollow cathode. The anticathode, located between the anode and oven, is fabricated from tantalum with the same diameter opening as the cathode to pass the vapour from the oven charge into the anode cavity for ionization. Since beryllium has a melting temperature 1275°C , the high temperature oven of the Danfysik ion source is used to heat the beryllium metal to temperatures approaching its melting point. The boron nitride oven is machined to approximately the same dimensions as that of the Danfysik design with one notable change: the inside diameter of the oven was decreased from 4.1 mm to 3.2 mm to increase the wall strength since the oven was cracking too readily from either stresses in the material which are released by the high temperatures or from the differential expansion of boron nitride and the other metallic source parts. Boron

Figure 3. *Cross sectional view of the oscillating electron bombardment ion source.*

Electrons produced by the heated tungsten filament follow oscillating helical trajectories between the cathode and anticathode under the influence of the applied magnetic field (insulators are cross-hatched). Vapour produced by heating beryllium wire in the oven (not shown) is ionized by multiple electron impact and extracted through the cathode and mass-analyzed to form a beam of Be^{2+} ions.

nitride, which is sintered from powder, expands approximately 10 times more in the direction along which it is molded compared to the expansion perpendicular to the molding pressure. The 22 mm long oven cavity can hold enough beryllium to operate the ion source for 10 to 15 hours depending on the oven temperature, which is mainly controlled by the power dissipated in the oven winding.

Beryllium, a known carcinogen, (as well as its compounds) is extremely toxic when handled in the form of a powder.⁵¹ Exposed skin can become inflamed and sores infected with beryllium are slow-healing. Acute intoxication due to inhalation of beryllium fumes or dust (300 mg/m^3) may result in inflammation of mucous membranes causing pain and difficulty in swallowing and breathing. Exposure to higher concentrations can cause immediate death. Chronic exposure attacks mainly the lungs causing pulmonary granulomatous disease (Berylliosis) up to 15 years after short exposures to low concentrations. The threshold limit value for exposure to the metal dust is 0.002 mg/m^3 for long term exposure. Because of the toxic effects of exposure of the lungs to beryllium dust, only beryllium in the form of wire was used as a feed material for the oven. This wire which is approximately 1.5 – 2 mm in diameter is safely handled wearing vinyl gloves and a face mask. The wire is easily cut into 18 mm lengths for loading into the oven without producing any dust.

Connections to the various ion source components are made via tantalum leads to stainless steel posts fastened to vacuum feedthroughs. Three simple DC 10 V – 30 A supplies power the magnet, filament, and oven. The unregulated power supplies, which were built to the same specifications ($< 0.5\%$ ripple) as those sold by Danfysik, are voltage controlled by autotransformers making them exceedingly robust. The arc potential is produced by a Hewlett Packard 6448B

⁵¹Johnson Matthey Aesar Group, *Material Safety Data Sheet for Beryllium*, 1986.

regulated power supply. A $900\ \Omega$ resistor was connected in series with the arc plasma to limit the arc current to 0.67 A, protecting the supply from ion source transients. Since the entire ion source is biased ~ 10 kV above ground these power supplies were floated at the source potential by a 20 kV isolation transformer. Ion source wiring connections are detailed in Figure 4.

After a great deal of investigation of source operation, it was found that the following source conditions gave the optimum balance of ion source stability, signal size, and lifetime of the beryllium feed material:

<u>Electrode</u>	<u>Voltage</u>	<u>Current</u>
Anode	660 V	35 mA
Filament	8.8 V	29 A
Oven	8.6 V	18 A
Magnet	8.6 V	24 A.

Source conditions such as these produce 3 nA ion beams of Be^{2+} and resonance signals of 500 counts/s. The source has been operated in two configurations:

- 1) cathode – anode – anticathode – oven (extraction through cathode)
- 2) anticathode – anode – cathode – oven (extraction through anticathode).

In general, larger beam currents of Be^{2+} were observed with the 2nd configuration with a corresponding increase in signal but this was at the expense of source stability as measured by ion current fluctuation.

4.3 Resonance Region and Fluorescence Detection

Fluorescence from the ion beam is detected in the resonance region where a post-accelerator (see Figure 5) Doppler shifts the ions into resonance with the $1s2s^1S_0 - 1s2p^1P_1$ transition causing fluorescence from the 2^1P state to the 1^1S ground state. This prevents the laser from being in resonance with the ions for the entire overlap region and restricts the fluorescence to the region inside the post-accelerator electrode where it is viewed through high transmission mesh by the detector. For a 10 kV ion beam of Be^{2+} the rate of change of frequency as a

Figure 4. *Wiring diagram of the oscillating electron bombardment ion source.*

The entire ion source and its power supplies are biased ~ 10 kV above ground by a high voltage power supply. The ion source power supplies are fed by the output of the 20 kV isolation transformer. The magnet (labelled a) is the external solenoid which produces the axial magnetic field causing helical electron trajectories. The $900\ \Omega$ ballast resistor for the anode power supply is not shown.

Figure 5. *Resonance region (to scale).*

A negative bias is placed on the centre electrode of the post-accelerator to Doppler shift the laser light, as seen in the rest frame of the ions, into resonance with the $2^1S - 2^1P$ interval. The 10 nm fluorescence to the ground state is viewed by the MCPA through a polypropylene window which shields the detector from charged particles.

function of voltage applied to the electrode is 53 MHz/V. Since the width of the resonance is ~ 20 GHz, the bias on the resonance electrode must be at least 380 V to avoid ion-laser beam interaction outside the region. A typical fixed resonance electrode bias would be in the range -500 to -1000 V.

The 10.0 nm laser-induced fluorescence from the 2^1P to the 1^1S state is detected by a microchannel plate array (MCPA). The MCPA functions as an array of channeltron detectors. That is, the MCPA is a group of hollow glass cylinders bound together like a honeycomb, each cylinder containing a resistive secondary-emission coating. A photon which enters the input end of one of the cylinders liberates an electron from the coating via the photoelectric effect. This photo-electron is accelerated by the axial electric field placed across the ends of the cylinder and strikes the wall further down the channel to produce secondary electrons. The cascade and multiplication process continues down the channel until the end is reached or gain saturation occurs. For this particular application two 25.4 mm aperture MCPA's are used with the first one feeding the second one to increase the sensitivity. A 2000 V potential was applied across both MCPA's to produce a total gain of 10^7 for each individual photo-electron liberated by the VUV photon.

Since the MCPA is also sensitive to charged particles, and it is situated very close to the ion beam to increase its solid angle of detection, it is necessary to shield the detector by placing a window in front of it to reduce the background counts from stray charged particles which would easily mask the resonance signal. It is somewhat difficult to produce a rugged window which is transparent in the 100 Å spectral region. 6 μm mylar has been used as a window material up to 44 Å and is fairly resistant to rupture; however, it is opaque to radiation beyond 44 Å. Other window materials such as cellulose nitrate, Formvar, and Zapon in thicknesses ranging from 1000 to 5000 Å, supported on mesh have been used as windows in

proportional gas counters for detection of soft x-rays. Even with a mesh support these windows are extremely fragile and have a life span of only one month in vacuum due to evaporation. Polypropylene, on the other hand, when stretched to a thickness less than $1\text{ }\mu\text{m}$ makes an excellent soft x-ray window. Measurements of $1\text{ }\mu\text{m}$ polypropylene windows show the following transmission properties:⁵²

<u>Wavelength ($\text{\AA}$)</u>	<u>Transmission (%)</u>
8.3	80
44	75
67	70
113	35.

Polypropylene can be stretched very uniformly and irreversibly from an initial thickness of $\sim 26\text{ }\mu\text{m}$ to less than $1\text{ }\mu\text{m}$ by placing the film over a cylindrical vacuum chamber approximately 30 cm in diameter and securing it between two flanges with O-rings. As the chamber is slowly evacuated with a rotary pump the polypropylene is stretched into a parabolic shape. The stretching process is continued until interference fringes can be seen in the film when illuminated with a bright white light source and air is admitted allowing the chamber to return to atmospheric pressure. Since the stretching process is irreversible, the polypropylene remains stretched to a thickness less than $1\text{ }\mu\text{m}$ over a circular region of about 12 cm at the bottom of the paraboloid and can be cut out with scissors and placed between pieces of cardboard until it is needed or attached to a mount by a thin layer of vacuum grease. These pi. hole-free windows last several years in a vacuum environment and are extremely tolerant to small pressure gradients during pumping cycles.

The stretched polypropylene film and aperture forms an unsupported 25.4 nm window on the end of an aluminum shielding sleeve which houses the MCPA. Using 35% as the transmission of the fluorescence through the window,

⁵²A.J. Caruso and H.H. Kim, Rev. Sci. Instrum. **39**, 1059 (1968).

7.5% as the photon detection efficiency of the MCPA, and 2% as the solid angle efficiency the overall efficiency of the detection system is 0.05%. The MCPA pulses are amplified a further factor of 100 by a model 156b Chronetics dual amplifier and then discriminated with a 50 mV threshold by a Chronetics 151A discriminator. The latter produces negative pulses which are inverted and then counted by the computer. Assuming only 0.55% of the metastable population is pumped to the excited state (see Appendix 2) and using the calculated detection efficiency, a typical resonance signal of 500 counts/s from a 3 nA beam of Be^{2+} translates into a 2% metastable content or a 60 pA beam of 2^1S metastables. Detection of this small signal is only possible because of the near-zero background (< 6 counts/s) which is a result of the polypropylene window and the insensitivity of the MCPA to detected photons from ion collisions with residual gas.

4.4 Lasers and Wavelength Measuring Equipment

The laser system used to excite the 2^1S metastable Be^{2+} ions consists of a Coherent CR-699-21 actively stabilized ring dye laser pumped by a Spectra-Physics model 171-19 argon ion laser. The S-P Ar^+ laser is capable of producing 18 W of light in the visible region of the spectrum and 2.5 W in the ultraviolet region. Since the Ar^+ laser is pumping a dye which absorbs more strongly the green region of the spectrum, the Ar^+ laser is run single-line by the installation of a prism to produce ~ 7 W of light at 514.5 nm. To produce a more stable output mode from the dye laser, the aperture control on the pump laser is adjusted to produce a TEM_{00} mode so that the laser beam cross section is round without a hole in the centre (TEM_{01}).

The Coherent CR-699-21 ring laser is actively stabilized by an external box of electronics. This results in a specified frequency jitter of less than 500 kHz and frequency drift of less than 50 MHz/hr. The frequency jitter is insignificant compared to the precision of this experiment and frequency drift is inconsequential

since the frequency is measured as the dye laser is scanned by observing its transmission through an iodine absorption cell. Ring lasers are travelling-wave lasers which do not produce a standing-wave pattern of light oscillation in the dye stream. At high pump powers this prevents a second frequency which has antinodes in places where the first frequency has nodes from reaching lasing threshold. As a result the ring laser operates more efficiently single mode since the laser can accept higher pump powers and the finesse of the frequency selecting devices in the cavity can be lowered and hence the operating losses. Using 7 W of 514.5 nm light to pump the KRS (Kiton Red S — also known as Sulforhodamine B) dye, the output power of the ring dye laser was optimized to 0.4 W at 614 nm (5.7% conversion efficiency).

Since the scan non-linearity of the ring laser is about 2% and the 30 GHz maximum scan length is only nominal, it is necessary to monitor the wavelength continuously as it scans, not just at the peak of the resonance. The laser can be scanned internally, manually, or externally. Externally generated scans are accomplished by providing a ± 5 V signal from the computer to the scan input jack which drives the scan generator according to the setting of the scan width control. The scan voltage from the computer is actually a ± 10 V signal which is divided in half by a voltage dividing circuit and smoothed by a 3.6 ms time constant which eliminates any rapid transients which might cause mode hops during the scan. A small portion of the dye laser output (~ 50 mW) is picked off by a thick beam splitter for frequency measurement. One of these beams is directed into a high-finesse Fabry-Perot étalon to monitor the mode structure. The second laser beam is split into three to be shared between a monochromator, the iodine absorption cell, and a second low-finesse Fabry-Perot étalon. The monochromator is used to make a coarse wavelength measurement (± 0.1 Å) to ensure the wavelength is near the resonance. The exact laser wavelength is measured as the

laser scans across iodine absorption lines which have been previously measured and tabulated in an atlas.⁵³ Knowing the approximate wavelength from the monochromator, it is possible to match the pattern and shape of the observed absorption lines with those shown in the atlas and hence establish an absolute frequency scale to 0.002 cm^{-1} (60 MHz). Since there is generally no iodine line at the exact centre of the resonance and since the laser scan is not absolutely linear, it is necessary to interpolate between absorption lines using frequency markers from a low-finesse Fabry-Perot étalon. The free spectral range (FSR) of this étalon (which is actually a pressure and temperature stabilized ring laser reference cavity for blue dyes) is $\sim 0.99\text{ GHz}$ and has a finesse of 2 at 614 nm. This provides frequency markers which can be located to better than 2 MHz at 1 GHz intervals throughout the laser scan.

A beam expander is used to collimate and expand the primary laser beam by a factor of four to improve the overlap with the larger ion beam. The laser light enters the vacuum system through a quartz Brewster window and passes through the resonance region before exiting through another Brewster window. The laser beam is retro-reflected by a mirror back through the vacuum apparatus so that it is nearly incident on the output aperture of the dye laser assuring that the overlap between laser and ion beams is identical for both parallel and anti-parallel spectra while the dye laser stability is not degraded by light which is coupled back into the laser cavity.

⁵³S. Gerstenkorn and P. Luc, *Atlas du spectre d'absorption de la molécule d'iode entre 14800–20000 cm⁻¹* (Editons du C.N.R.S., Paris, 1978).

4.5 Data Acquisition System

The acquisition of a single spectrum requires the simultaneous recording of five signals as the laser frequency is scanned. These five signals are

- 1) Laser-induced fluorescence (MCPA pulses)
- 2) Iodine absorption (analog voltage)
- 3) Laser power (analog voltage)
- 4) Étalon transmission (analog voltage)
- 5) Ion beam current (analog voltage).

Analog to digital (A/D) and digital to analog (D/A) conversions as well as pulse counting are handled by a RTI-815 multifunction board manufactured by Analog Devices which plugs into a PC/AT expansion slot. The RTI-815 board is configured to give 8 channels of 12 bit A/D conversion for input voltages in the ± 10 V range and 2 channels of 12 bit D/A conversion for output voltages in the range ± 10 V. There are 3 counter/timer channels available with maximum count rates of 100 kHz and gate times programmable from 2 μ s to 655.35 s with a time base accuracy of 0.01%. Thus for a 30 GHz scan, the minimum step in the staircase ramp generated by the computer to scan the laser is ~ 7.32 MHz; but in fact data was acquired only every 58.6 MHz which is more than ample for the ~ 20 GHz wide laser resonance and sufficient for determining the centres of étalon transmission peaks to 2 MHz. The data acquisition board can be programmed in a variety of languages including Fortran, which is used for this application.

The data acquisition program is written to display the five input signals simultaneously as separate graphs in real-time allowing the operator to halt data acquisition should the laser hop modes during the scan or the ion beam fluctuate excessively in energy or intensity. While the computer counts MCPA pulses (signal 1) for a set dwell time, it samples the analog input signals (signals 2-5) as many times as possible so that it records the average analog signal during that counting

period. Since the computer monitors the ion beam current, it can reject data which has been acquired while the ion beam current is fluctuating outside a given tolerance so that any distortions in the fluorescence lineshape due to fluctuations in the ion source behaviour can be minimized. Typical data acquisition parameters are

Dwell Time:	250 ms/channel
Channels:	512
Ion Beam Tolerance:	5.0%

yielding spectra with ~ 200 counts/channel at the peak of the resonance. Spectra were taken in groups of four, two with the laser and ion beams co-propagating ([P]arallel) and two counter-propagating ([A]nti-parallel) forming a PAAP or a APPA pattern. These patterns allow comparison of two similar measurements taken several minutes apart in order to check for drifts in the ion beam energy that would shift the position of the resonance peak. Approximately 15 minutes elapse during the acquisition of four spectra for typical data acquisition parameters.

CHAPTER 5

Data Analysis

5.1 General Considerations

In general, energies of atomic transitions are obtained from spectra by combining a model of the observed lineshape with a wavelength or frequency calibration. In most cases, the lineshape model is fit to the observed lines using a non-linear least-squares (NLLS) fitting program⁵⁴ to vary parameters in the model such as the background, linewidth, and peak centre. Very simply, the NLLS program varies the model parameters to minimize the weighted sum of the squares of the residuals (χ^2). Once the χ^2 is minimized, the values of the parameters for the linewidth and peak centre represent the best statistical estimate for the observed spectrum. Analysis of the data for this experiment requires the establishment of an absolute frequency scale for the measurement of laser wavelengths in the laboratory frame. It is then necessary to extract the Doppler-shifted wavelength as seen in the rest frame of the ions in order to determine the $2^1S_0 - 2^1P_1$ interval.

A typical spectrum composed of five signals is shown in Figures 6 and 7. The laser-induced-fluorescence signal is recorded as counts from an MCPA. A laboratory frequency scale is established using the signal from an iodine absorption cell and the fringes recorded from a Fabry-Perot étalon. The laser power and ion beam current are recorded as a measure of the operational stability of the laser and ion source. Even though the dye laser output power is fairly constant across the entire frequency scan and the ion beam current was monitored to eliminate large fluctuations in the current, the resonance signal was normalized to both laser power and ion current in order to remove the effect of the remaining variations.

⁵⁴W.H. Press, B.P. Flannery, S.A. Teukolsky, and W.T. Vetterling, *Numerical Recipes* (Cambridge, Cambridge, 1986), Chapter 14.

Figure 6. *Resonance signal and iodine absorption calibration lines.*

The upper graph shows the fluorescence detected by the MCPA as a function of laser frequency. A symmetric Lorentzian function, used to find the peak centre of the resonance, is plotted also. The lower graph records the transmission of laser light through an iodine cell as the frequency of the dye laser is scanned. This provides an absolute frequency calibration.

Figure 7. *Etalon transmission, laser power and ion current signals.*

The three graphs from top to bottom depict typical étalon transmission, laser power, and Be^{2+} ion current as a function of laser frequency for a 30 GHz laser scan. The ion current is monitored for fluctuations which exceed 5% which halts data acquisition until the tolerance is met again. The fluorescence signal is subsequently normalized to the laser power and ion current.

A detailed analysis has been developed describing the population transferred by laser excitation from the 2^1S level to the 2^1P level which fluoresces mainly to the 1^1S ground state (see Appendix 2). This model of a two level system damped by fluorescence to the ground state shows that, for the range of experimental parameters (laser power, interaction time etc.) encountered, the signal varies linearly with laser power. This justifies a linear normalization of the fluorescence signal to variations in the laser power. Furthermore, the analysis suggests the observed fluorescence signal is well-modelled by a single Lorentzian function neglecting distortions in the lineshape by Doppler broadening. The behaviour of the ion source, on the other hand, has not been extensively investigated. Calculations of metastable ion content from ion source parameters are not easily made so that there is no known functional dependence of metastable ion content with total ion current. Despite this lack of knowledge, a linear normalization of the fluorescence to the ion current for small variations is quite reasonable.

5.2 Frequency Calibration

The iodine atlas⁵⁵, used to establish the frequency scale for fluorescence, is a compilation of iodine absorption lines measured by Fourier transform spectroscopy. It contains more than 22,000 measured absorption lines and extends from 14,800 to 20,000 cm^{-1} . The original atlas data was obtained by observing the absorption of light from a tungsten iodine quartz lamp with a Fourier transform spectrometer after the light passed through an iodine cell. A hollow cathode uranium lamp was positioned between the absorption cell and the spectrometer to provide uranium emission lines for calibration.⁵⁶ The wavelengths of these lines were measured

⁵⁵S. Gerstenkorn and P. Luc, *Atlas du spectre d'absorption de la molécule d'iode entre 14800 - 20000 cm^{-1}* , (Centre National de la Recherche Scientifique, Paris, 1978).

⁵⁶S. Gerstenkorn, P. Luc, A. Perrin, and J. Chauville, *Astron. Astrophys.* **58**, 255 (1977).

independently by comparison with krypton spectra.⁵⁷⁻⁵⁸ The Doppler width at room temperature of the observed lines varies from 0.011 to 0.015 cm^{-1} . Using a resolving power of $\sim 10^6$, the absorption lines recorded by the Fourier transform spectrometer are claimed to be nearly symmetrical. The reported internal coherence of the iodine lines is 0.0007 cm^{-1} while the absolute accuracy using the external uranium standard is $\pm 0.006 \text{ cm}^{-1}$. The absolute error arises as a result of the accuracy of the uranium lines used for calibration (0.004 cm^{-1}) and possible small misalignments between the light source for absorption and the uranium lamp.

Improved progress in the identification of the iodine spectrum⁵⁹⁻⁶⁰ and the precision measurement of the hyperfine structure of numerous lines⁶¹⁻⁶² has led to a recalibration of the absorption spectrum using the "internal standard method".⁶³ The internal standard method uses a single hyperfine component measured by saturated absorption laser spectroscopy (typical experimental uncertainties range from 0.0003 cm^{-1} to 0.00002 cm^{-1}) and the known hyperfine structure to calculate the centre of gravity of all the unresolved hyperfine components for that line. This centre of gravity is then compared to the corresponding line centre measured by Fourier transform spectroscopy to check for systematic errors in the calibration of the Fourier transform spectrometer data. Therefore, one must be sure that the calculated centre of gravity of the hyperfine structure of a given line really corresponds to the observed peak position using Fourier transform spectroscopy.

⁵⁷V. Kaufman and G.J.J. Humphreys, J. Opt. Soc. Amer. 59, 1614 (1969).

⁵⁸S. Gerstenkorn and P. Luc, *Nouv. Rev. Optique* 7, 149 (1976).

⁵⁹S. Gerstenkorn and P. Luc, J. Mol. Spectrosc. 77, 310 (1979).

⁶⁰P. Luc, J. Mol. Spectrosc. 80, 41 (1980).

⁶¹J.E.M. Goldsmith, E.W. Weber, and T.W. Hänsch, Phys. Rev. Lett. 41, 1525 (1978).

⁶²L. Hlousek and W.M. Fairbank Jr., Opt. Lett. 8, 322 (1983).

⁶³S. Gerstenkorn and P. Luc, *Revue de Physique Appliquée* 14, 791 (1979).

To avoid the problem of possible blends in the absorption spectrum, the wavenumbers of the iodine lines are calculated from molecular constants derived from an exhaustive analysis of the entire iodine atlas. These calculated wavenumbers, not the measured line centres, are compared with the centres of gravity derived from saturated absorption laser spectroscopy. A comparison of the centres of gravity of 9 different lines with the calculated wavenumbers (from the molecular constants) shows a systematic shift. The wavenumbers determined by Fourier transform spectroscopy are too high by $0.0056 \pm 0.001 \text{ cm}^{-1}$. Furthermore, the accuracy of the corrected absolute wavenumbers in the atlas was determined to be $\pm 0.002 \text{ cm}^{-1}$ (two standard deviations) over the entire region of the atlas. The internal standard method has been applied a second time with a total of 36 different hyperfine components measured by saturated absorption spectroscopy covering a wider range of the atlas ($16,300 - 17,600 \text{ cm}^{-1}$).⁶⁴ This most recent recalibration reports an atlas correction which is proportional to the wavenumber as it should be in Fourier spectroscopy and is given by $\sigma_{\text{cor}} = \sigma_{\text{atlas}} (1 + \delta)$ where $\delta = -3.8 \times 10^{-7}$. For the iodine absorption lines which are used for calibration of both co-propagating and counter-propagating beryllium spectra, this correction becomes $\sigma_{\text{cor}} = \sigma_{\text{atlas}} - 0.0062 \text{ cm}^{-1}$. Obviously, any further change in the atlas correction must also be applied to the experimental determination of the $1s2s^1S_0 - 1s2p^1P_1$ interval in Be III. Although there has been three different atlas corrections for the iodine atlas, it is highly unlikely that the present atlas correction will change by a significant fraction of the experimental uncertainty.

To check the correspondence between the calculated centre of gravity and the peak position,⁶⁵ the authors of the iodine atlas investigated the experimental

⁶⁴S. Gerstenkorn and P. Luc, *Atlas du spectre d'absorption de la molécule d'iode entre 14800 - 20000 cm⁻¹ complément*, (Centre National de la Recherche Scientifique, Paris, 1987).

⁶⁵S. Gerstenkorn and P. Luc, *Revue de Physique Appliquée* **14**, 791 (1979).

lineshape. The observed lineshapes of the iodine absorption lines result from the convolution of three functions: the source profile (P_0) which can be calculated from hyperfine structure, the Fourier apparatus function ($F = \frac{\sin x}{x}$), and a rectangular function (T) due to the finite aperture of the Fourier interferometer. At room temperature, each hyperfine component for weak absorption is taken to have a theoretical Doppler width of ~ 350 MHz. The width of the two apparatus functions is 450 MHz. The calculated line profile (P_2) is then given by $P_2 = P_0 \bullet F \bullet T$ where $\bullet$ denotes convolution. The resulting profile is almost symmetrical and the discrepancy between the position of the maximum of the line profile and the centre of gravity is negligible. Following this recipe, the iodine absorption lines recorded in this experiment are convolved with F and T to produce a nearly symmetrical lineshape whose centre (maximum) is taken to be the wavenumber (neglecting the atlas correction) quoted in the atlas for that line.

5.3 Determination of Line Centres

Data analysis is accomplished using a program written in Gauss (a matrix oriented language) running on a PC/AT compatible. Six columns of 512 rows of data are read representing the laser scan voltage and the five data signals. The iodine absorption signal is immediately convolved with F and T to produce the line profile P_2 whose centre frequency can be found in the iodine atlas. To establish a linear but not absolute frequency scale, the Fabry-Perot fringe peaks are fit with an Airy function, $\mathcal{A}(\Delta\nu)$, which models the transmission of light through an étalon as a function of frequency.⁶⁶ The Airy function can be written as

$$\mathcal{A}(\Delta\nu) = A / [1 + F \sin^2(\pi\Delta\nu / \text{FSR})]$$

⁶⁶E. Hecht and A. Zajac, *Optics*, (Addison-Wesley, New York, 1974).

where

A is the peak amplitude (maximum transmission),

$$F = \frac{4R}{(1 - R)^2}, \text{ (R is the reflectivity of the étalon mirrors)}$$

$\Delta\nu = \nu - \nu_0$, is the frequency offset from maximum transmission, and

FSR is the free spectral range of the étalon (~ 1 GHz).

For a typical laser scan 28 or 29 étalon peaks are fit using 15 points centred around each étalon transmission peak. The NLLS fit to the peaks determines their centres to better than 2 MHz.

The étalon peak centres would be equally spaced if the laser frequency scan was truly linear. To compensate for any non-linearities in the laser scan, a linear but non-absolute frequency scale is established by arbitrarily assigning the frequencies of the peak centres to be 1 GHz (the approximate FSR of the étalon) times the peak number (1 – 29) and fitting this data as a function of scan voltage. A 6th degree polynomial is fit to this data showing a maximum non-linearity of less than 0.1%. To establish an absolute frequency scale, the observed iodine absorption lines, whose centres are known from the iodine atlas, are determined by a quadratic fit to 11 points centred around the peak of the absorption. This fit uses the nearly-symmetric convolved absorption data and the linear (but arbitrary) frequency axis. Depending on the spectrum, three to ten absorption lines are analyzed producing as many absolute calibration points for a linear fit of absolute wavenumber versus peak centre measured on the arbitrary frequency axis. This determines the frequency offset for the frequency axis as well as the overall scaling parameter. When this parameter is multiplied by the 1 GHz separation assigned to the interval between étalon peaks, the result is the FSR of the étalon. The maximum residual for the peak frequency of any iodine absorption line as measured

with the now absolute frequency scale and the value listed in the atlas for that line is less than 0.002 cm^{-1} (60 MHz) which fits well with the $\pm 0.002 \text{ cm}^{-1}$ accuracy of the lines measured in the iodine atlas. Many different iodine lines are used for calibration by varying the ion beam energy (Doppler shift). This prevents the final measurement of the $2^1\text{S} - 2^1\text{P}$ interval from being influenced by errors in the atlas for an occasional absorption line and is a good check for systematic trends with ion beam energy. In all, 54 different absorption lines are used for calibration, 20 lines for parallel overlap spectra and 34 for anti-parallel overlap.

The line centre of the resonance signal is determined by modelling the observed lineshape (normalized to the laser power and ion beam current) with a Lorentzian whose amplitude, full-width at half-maximum, and peak centre are fit parameters. Specifically, the Lorentzian fitting function is

$$\mathcal{L}(\Delta\nu) = B + A / [1 + (2\Delta\nu / \text{FWHM})^2]$$

where

B is the background which is determined experimentally,

A is the peak amplitude,

$\Delta\nu$ is the frequency offset from the centre of the resonance, and

FWHM is the full-width at half-maximum.

Using this model, the uncertainty in the peak centre of the fluorescence is 0.004 to 0.008 cm^{-1} (120 to 240 MHz) due to statistical uncertainty from the scatter in the fluorescence signal. The error in the peak centre due to calibration of the frequency axis is 0.002 cm^{-1} due solely to the uncertainties in absolute wavenumbers used for calibration of the iodine absorption lines. Finally, 0.0062 cm^{-1} is subtracted from the wavenumber of the peak centre to correct for the systematic error in the iodine absorption lines used for absolute frequency calibration.

The experimental background is extremely small (typically 5 counts/s)

arising mainly from collisions of the ion beam with background gas and the inherent noise in the MCPA. The background is determined by blocking the laser light outside the vacuum and counting for a fixed period of time. This measurement is repeated after the acquisition of four spectra or more often when the ion current is not too stable. The peak centre is very insensitive to the background. The FWHM, on the other hand, is extremely sensitive to the background and, in some cases, a negative background produced by NLLS modelling yields an erroneously large value for the FWHM. Thus, to produce an accurate measurement of the FWHM and hence place a lower bound on the lifetime of the 2^1P_1 level, the data are analyzed with a fixed background model using the measured background. Typical results for the FWHM from the fitting program are 20 to 25 GHz with statistical uncertainties of 0.3 to 0.6 GHz depending on the signal-to-noise present in the spectrum. The weighted average for 133 measurements of the FWHM is 22.5 ± 1.5 GHz.

A further experimental complication arises due to the geometry of the resonance region (see Appendix 3). The MCPA can view fluorescence from regions of the post-accelerator where there are large electric potential gradients. Therefore, the ion beam has an effective Doppler distribution due to the acceleration of an individual ion within the viewing region of the MCPA. This effective Doppler distribution is distinct from the usual Doppler distribution due to the spread of ion energies present in the ion beam when it is extracted from the ion source. In fact, the usual Doppler distribution is negligible compared to this effective Doppler distribution. This effective Doppler distribution broadens the observed resonance and produces an asymmetry which reverses for parallel and anti-parallel geometries and thus does not distort the calculation of the Doppler-free interval.

5.4 Calculation of Doppler - Free $2^1S_0 - 2^1P_1$ Intervals

The resulting Doppler shifted line centres from the fit spectra are analyzed in the groups of four as they were acquired ($P_1A_1A_2P_2$ or $A_1P_1P_2A_2$ where P_i represents a parallel overlap of laser and ion beams while A_i represents an anti-parallel overlap). If the velocity distribution of the ions remains constant during the acquisition of a group of four spectra, the peak centre for pairs of spectra with the same overlaps should be identical. That is, for a $P_1A_1A_2P_2$ group of spectra, the P_i spectra should have identical line centres and likewise the A_i spectra. Should there be small drifts in the ion source operating conditions producing a change in the ion beam energy, this would be most noticeable in the differences in the line centres for the first and fourth spectra in the group of four. Before calculating the Doppler-free wavenumber for the $2^1S_0 - 2^1P_1$ transition from these wavenumbers, measured in the laboratory frame, the A_i and P_i spectra are averaged separately within a group of four spectra to produce a single value for both the anti-parallel and parallel Doppler shifted wavenumber. The error assigned to each wavenumber is the larger of either an internal estimate of the error or an external estimate. For the parallel case, these errors calculated for the average wavenumber σ_P (defined as $\sigma_P = (\sigma_{P_1} + \sigma_{P_2}) / 2$) are

$$\delta\sigma_P^2 = (\delta\sigma_{P_1}^2 + \delta\sigma_{P_2}^2) / 4, \quad (\text{internal estimate})$$

$$\delta\sigma_P^2 = (\sigma_{P_1} - \sigma_{P_2})^2 / 2, \quad (\text{external estimate})$$

where

$\delta\sigma_{P_i}$ is the uncertainty in the wavenumber σ_{P_i} for the i^{th} measurement of the line centre for a parallel overlap.

The relativistic Doppler formula relating the resonance frequency ν_0 (as seen in the rest frame of the ions) to the measured laser frequency ν_L (in the laboratory)

is

$$\nu_0 = \gamma(1 \pm \beta \cos \theta) \nu_L$$

where

$\beta = v/c$ is the fraction of the ion velocity compared to the speed of light,

$$\gamma = (1 - \beta^2)^{-\frac{1}{2}}.$$

For collinear geometry ($\cos \theta \cong 1$) and a parallel overlap,

$$\nu_0 = \gamma(1 - \beta) \nu_P,$$

and likewise for an anti-parallel overlap

$$\nu_0 = \gamma(1 + \beta) \nu_A,$$

where ν_P and ν_A are the resonant laser frequencies measured in the laboratory frame. Since both ν_P and ν_A are measured one can calculate both ν_0 and β as

$$\nu_0 = (\nu_P \nu_A)^{\frac{1}{2}}$$

$$\beta = (\nu_P - \nu_A) / (\nu_P + \nu_A).$$

Thus for measured line centres in wavenumbers the above equations can be rewritten as

$$\sigma_0 = (\sigma_P \sigma_A)^{\frac{1}{2}}$$

$$\beta = (\sigma_P - \sigma_A) / (\sigma_P + \sigma_A)$$

and

$$\delta \sigma_0 = \frac{1}{2} \sigma_0 (\delta \sigma_A^2 / \sigma_A^2 + \delta \sigma_P^2 / \sigma_P^2)^{\frac{1}{2}}$$

$$\delta \beta = 2(\sigma_P^2 \delta \sigma_A^2 + \sigma_A^2 \delta \sigma_P^2)^{\frac{1}{2}} / (\sigma_P + \sigma_A)^2.$$

Thirty groups of four spectra taken on different days with different beam energies were analyzed. The resulting Doppler-free $2^1S_0 - 2^1P_1$ intervals are

tabulated below as a function of beam energy/charge:

<u>Interval</u> (cm^{-1})	<u>Uncertainty</u> (cm^{-1})	<u>Beam Energy/charge</u> (V)
16276.778	0.009	9359
16276.773	0.008	9366
16276.776	0.003	9419
16276.782	0.006	9640
16276.763	0.002	9945
16276.768	0.005	9949
16276.757	0.005	9950
16276.767	0.003	9950
16276.791	0.012	10068
16276.784	0.016	10270
16276.765	0.006	10307
16276.780	0.009	10308
16276.774	0.013	10314
16276.766	0.005	10315
16276.773	0.007	10367
16276.786	0.012	10423
16276.787	0.006	10433
16276.785	0.003	10581
16276.782	0.005	10582
16276.778	0.003	10584
16276.787	0.005	10587
16276.784	0.006	10587
16276.774	0.004	11259
16276.769	0.008	11336
16276.773	0.010	11412
16276.783	0.010	11427
16276.786	0.005	11439
16276.762	0.007	11445
16276.778	0.005	11457
16276.756	0.010	11467.

The average value of the $2^1S_0 - 2^1P_1$ interval is $16276.776(9) \text{ cm}^{-1}$ for these 30 observations. Using the squares of the uncertainties for the intervals for weighting, the weighted average for the $2^1S_0 - 2^1P_1$ interval is $16276.774(9) \text{ cm}^{-1}$. A histogram of the 30 individual measurements of the $2^1S_0 - 2^1P_1$ interval is shown in Figure 8. The dominant contributions to the uncertainty are the shot noise in the photon counting and the drifts in the ion beam energy. The intervals are also plotted against ion beam energy in Figure 8 to illustrate the lack of any systematic trends as a function of ion energy. A linear fit of the transition wavenumber to the ion beam energy/charge determines a slope which, within error, is zero. In fact, a fit to

Figure 8. *Distribution of transition wavenumbers and ion energy dependence.*

The histogram illustrates the distribution of Doppler-free transition wavenumbers for all 30 measurements of the $1s2s^1S_0 - 1s2p^1P_1$ interval. The lower graph of transition wavenumber *vs* ion beam energy/charge illustrates the lack of systematic trends due to ion energy.

a straight line with zero slope (i.e. a determination of the weighted mean) gives a smaller chi-square per degree of freedom.

Some clustering of values of the $2^1S_0 - 2^1P_1$ interval at a given energy is evident from Figure 8. This clustering becomes more apparent with the application of a Runs test⁶⁷⁻⁶⁸ to the data to check for non-random fluctuations about the mean of the data. One statistically significant run is comprised of seven consecutively high measurements (that is, seven measurements which lie above the average of all the data). The expected number of runs of length seven for a set of 30 measurements is smaller than one by four standard deviations. Nevertheless, even for this conspicuous run, the mean of the seven measurements is greater than the weighted mean of all the data by only 0.009 cm^{-1} which is one standard deviation for the weighted average of all 30 measurements and the quoted experimental error. This clustering is probably a result of ion source conditions which vary from day to day and can not be controlled absolutely by the source parameters; however, this effect has been minimized by repeating the measurements with different source conditions and source configurations as well as by taking data over many different days.

⁶⁷W. Feller, *An Introduction to Probability Theory and its Application*, (Wiley, New York, 1968), p. 42.

⁶⁸W. Mendenhall, *Introduction to Probability and Statistics*, (Duxbury, North Scituate, 1979), p. 498.

CHAPTER 6

Conclusions

6.1 Experimental Results

The $1s2s^1S_0 - 1s2p^1P_1$ interval in Be III has been measured using laser resonance spectroscopy. This is the first high precision measurement of the $2^1S - 2^1P$ interval in a two-electron atom. Experimental results and theoretical calculations for this interval are compared in the following table:

Experiment	Edlén ⁶⁹ Löfstrand ⁷⁰ This work	16279.0 cm ⁻¹ 16276.8(1) cm ⁻¹ 16276.774(9) cm ⁻¹
Theory	Drake ⁷¹	16276.766(30) cm ⁻¹ .

The results are in excellent agreement with an earlier measurement by Löfstrand with an improvement in precision by a factor of 11. The first measurement of this interval in Be III by Edlén is probably not in agreement; however, no experimental precision is quoted. The best theoretical calculation of the $2^1S - 2^1P$ separation by Drake is in good agreement with this measurement but is less precise by a factor of three. The future inclusion of relativistic contributions of order ($\alpha^4 Z^4$) will increase the theoretical accuracy providing a more stringent comparison. Assuming exact calculations of the non-QED contributions to the interval, this experiment represents a 0.3% measurement of the Lamb shift. Since the iodine atlas has been used as a frequency standard, any further atlas correction must also be applied to this interval. Although there have been several recalibrations of the iodine atlas, it is highly unlikely that the present atlas correction (see Chapter 5) will change by a

⁶⁹B. Edlén, Ark. Fys. 28, 441 (1951).

⁷⁰B. Löfstrand, Physica Scripta 8, 57 (1973).

⁷¹G.W.F. Drake, Can. J. Phys. 66, 586 (1988).

significant fraction of the experimental uncertainty.

The width of the observed resonance laser resonance for the $2^1S - 2^1P$ interval (dominated by the lifetime of the 2^1P level which decays rapidly to the ground state) is increased by inclusion of fluorescence from ions in the fringing fields of the post-accelerator. This precludes an accurate measurement of the lifetime of the 2^1P state, setting only a lower limit of 7.1(5) ps. Even without this broadening, the accuracy of the measurement would be $\sim 7\%$ which is not a very precise test of the relativistic calculation of the 8.20 ps lifetime of the 2^1P state by Drake.⁷² Subsequent modelling of the observed lineshape due to the broadening by fluorescence from fringing regions shows that the widths of aggregate spectra suggest an unbroadened linewidth of 19.4 GHz corresponding to the theoretical lifetime (see Appendix 3). This model is very sensitive to the angular detection efficiency of the microchannel plate array which is very difficult to measure and has not been measured for this experimental wavelength.

6.2 Possible Experimental Improvements

The most obvious experimental improvement is the elimination of fringing fields in the region of the post-accelerator viewed by the microchannel plate array. One possible method is to place apertures in the electrodes to reduce the penetration of fringing potentials into the accelerating electrode where the fluorescence is viewed. Another method is the use of a series of parallel conducting plates with apertures which are aligned to permit the ion and laser beams to pass through. The potential placed on each plate could then be controlled separately which would better define the post-accelerating potential in the vicinity of the region viewed by the microchannel plate array between the plates.

⁷²G.W.F. Drake, Phys. Rev. A 19, 1387 (1979).

The present experimental accuracy is determined by the scatter in measurements. Further ion source development could perhaps produce an ion source with greater stability producing an ion beam with less energy drift and a higher metastable content. This would allow longer periods of data acquisition over which more data could be collected reducing statistical uncertainties in the peak centres and lineshape distortions caused by changes in the Doppler distribution of the ion beam. An even simpler method of increasing the amount of observed signal is to increase the solid angle of detection by employing several microchannel plate arrays. This is an excellent means of decreasing the data acquisition time for a spectrum since the observed background is negligible.

This experiment could be repeated with perpendicular geometry instead of the present collinear geometry. That is, the ions and laser beams would intersect at right angles which would eliminate the Doppler shift of the laser frequency seen by the ions. Although the elimination of the Doppler shift is a great experimental advantage, the system must be very carefully aligned because the lineshape is much more sensitive to any asymmetries in the overlap of the two beams and any deviation from the perpendicular.

6.3 Future Experiments

The most obvious subsequent experiment is a measurement of the $1s2s^3S_1 - 1s2p^3P_J$ intervals which can yield a much greater experimental precision because of the narrower linewidths which results from the longer lifetime of the $1s2p^3P_J$ states (the ultimate limit is 5.4 MHz due to the 29 ns lifetime of the $1s2p^3P_J$ levels). Another advantage will be increased fluorescence signals arising from the decreased damping of the excited state which permits more efficient transfer of population from the metastable state to the radiating state (see Appendix 2). This advantage will be partially offset by the ~ 372 nm laser excitation wavelength. This requires either intra-cavity frequency doubling in the

ring dye laser or a new Ar^+ ion laser with greater output in the deep UV region which is capable of pumping a dye with a tuning curve that covers this wavelength region. Either means of production of the necessary tunable radiation will provide substantially less power than is available at the present experimental wavelength. Since the fluorescence from the 2^3P state is back to the metastable 2^3S level, any scattered laser light will provide a large background signal. This can be eliminated by a carefully designed post-accelerator to eliminate both scattered light and fringing fields in the viewing region. One final experimental hurdle is the measurement of absolute wavelengths. If the laser wavelength is produced using frequency doubling, the primary wavelength can be measured using saturated absorption of iodine. For direct production using a UV dye, another absolute standard or a transfer standard will have to be found.

The next two-electron atom which would be possible to study is B IV. The ions could be produced from BF_3 in a commercial Penning ion discharge source or perhaps in the source developed for this work. The laser excitation wavelength for the singlet system is ~ 450 nm but the width of the resonance is at least 59 GHz which is three times worse than for this experiment. The $2^3\text{S} - 2^3\text{P}$ interval for two-electron boron is ~ 282 nm which would definitely require frequency doubling.

Laser resonance measurements of $2^1\text{S} - 2^1\text{P}$ and $2^3\text{S} - 2^3\text{P}$ intervals provide the best tests of QED calculations. The increasing availability of intense, narrowband UV light sources and novel ion sources should allow high precision measurements of this type to be extended to the region of $Z \sim 10$ to take advantage of the $\sim Z^4$ dependence of the Lamb shift. For heliumlike neon ($Z = 10$), the $1s2s^3\text{S}_1 - 1s2p^3\text{P}_1$ interval is ~ 127 nm. The width of the laser resonance due to the lifetime of the 2^3P state is ~ 0.852 GHz. Assuming one can measure the centre of the resonance to the same relative precision as in this experiment, this would test calculations of the Lamb shift to 3 parts in 10^6 (assuming exact non-relativistic

energies). With the continual refinement of theoretical calculations for two-electron atoms, these types of high precision experiments will be necessary to provide significant tests of QED in the future.

APPENDIX 1

Hyperfine Structure of the $1s2s^1S_0 - 1s2p^1P_1$ Transition

Since the total electronic angular momentum J of the metastable $1s2s^1S_0$ level is zero, it therefore has no hyperfine structure. The $1s2p^1P_1$ excited state on the other hand has total electronic angular momentum $J = 1$ which when coupled to the nuclear spin $I = 3/2$ produces three hyperfine components of total angular momentum $F = 5/2, 3/2$, and $1/2$. These hyperfine components are not resolved in the observed spectra because of the short lifetime of the 2^1P_1 level and the relatively small hyperfine structure but in principle they could alter the line profile and shift the centroid (defined as $\sum_{F'} (2F'+1) \langle \mathcal{H}_{fs} \rangle_{F'}$). The magnetic dipole hyperfine interaction can be written as⁷³

$$\mathcal{H}_{fs} = A(J) \mathbf{I} \cdot \mathbf{J}.$$

The centroid of three hyperfine components for an interaction of the form $\mathbf{I} \cdot \mathbf{J}$ with relative intensities proportional to $2F+1$ (where $\mathbf{I}$ and $\mathbf{J}$ couple to form $\mathbf{F}$) is exactly where it would be in the absence of the $\mathbf{I} \cdot \mathbf{J}$ hyperfine interaction. However, if the relative intensities of the fluorescence are not in the $2F+1$ ratio, or if there are other hyperfine interactions present, then a systematic shift would be present in the measured $1s2s^1S_0 - 1s2p^1P_1$ interval. Both of these situations are considered and are shown to be negligible to the present experimental precision.

Calculation of the $1s2p^1P_1$ Hyperfine Structure

The relativistic spin-orbit interaction written as

$$\mathcal{H}_{so} = \sum_i \xi(r_i) \mathbf{l}_i \cdot \mathbf{s}_i$$

⁷³G.K. Woodgate, *Elementary Atomic Structure*, (Clarendon, Oxford, 1980), Chapter 9.

mixes states with the same electronic angular momentum J .⁷⁴ In this case, the most important contribution to the $1s2p^1P_1$ state is the $1s2p^3P_1$ state. As a result the actual or observed $1s2p^1P_1$ state ($|1s2p^1\Psi\rangle$) should be written as

$$|1s2p^1\Psi\rangle = \cos\theta|1s2p^1P_1\rangle - \sin\theta|1s2p^3P_1\rangle$$

where the values of $\cos\theta$ and $\sin\theta$, the mixing coefficients, have been calculated to be 0.9999 and 1.7046×10^{-3} respectively.⁷⁵ The hyperfine energies are then given by

$$\Delta E_{\text{hfs}} = \langle 1s2p^1\Psi | \mathcal{H}_{\text{hfs}} | 1s2p^1\Psi \rangle$$

or

$$\begin{aligned} \Delta E_{\text{hfs}} = & \cos^2\theta \langle 1s2p^1P_1 | \mathcal{H}_{\text{hfs}} | 1s2p^1P_1 \rangle \\ & - 2\sin\theta\cos\theta \langle 1s2p^3P_1 | \mathcal{H}_{\text{hfs}} | 1s2p^1P_1 \rangle \\ & + \sin^2\theta \langle 1s2p^3P_1 | \mathcal{H}_{\text{hfs}} | 1s2p^3P_1 \rangle \end{aligned}$$

where the magnetic dipole hyperfine operator $\mathcal{H}_{\text{hfs}}$ is written as⁷⁶

$$\mathcal{H}_{\text{hfs}} = \left(\frac{\mu_0}{4\pi} \right) \left(2\mu_B \frac{\mu_I}{I} \right) \mathbf{I} \cdot \sum_i \left\{ \frac{\mathbf{l}_i}{r_i^3} - \frac{1}{r_i^3} \left(\mathbf{s}_i - 3 \frac{(\mathbf{s}_i \cdot \mathbf{r}_i) \mathbf{r}_i}{r_i^2} \right) + \frac{8\pi}{3} \mathbf{s}_i \delta^3(\mathbf{r}_i) \right\}.$$

Here $\mathbf{l}_i$, $\mathbf{s}_i$, and $\mathbf{r}_i$ are, respectively, the orbital angular momentum, spin, and position of the i^{th} electron, μ_B and μ_I are the Bohr magneton and nuclear magnetic moment and the summation is over both electrons. The first term in the summed brackets is the orbital-dipole term describing the interaction of the magnetic dipole moment of the nucleus with the magnetic field at the nucleus produced by the orbital motion of the electrons. The second term is the dipole-dipole term describing the interaction of the magnetic dipole moment of the nucleus with the magnetic dipole moment of the electron. The third term is the Fermi contact term which has no classical analogue. The first two terms are applicable only for electrons with $l \neq 0$ while the third term is non-vanishing only for electrons with

⁷⁴G.K. Woodgate, *op. cit.*, Chapter 7.

⁷⁵G.W.F. Drake, *Phys. Rev. A* **19**, 1387 (1979).

⁷⁶G.K. Woodgate, *op. cit.*, Chapter 9.

$l = 0$.

Since the triplet mixing coefficient $\sin\theta$ is very small, the hyperfine energies can be written as the sum of the first two terms only, that is

$$\Delta E_{\text{hfs}} = \cos^2\theta \langle 1s2p^1P_1 | \mathcal{H}_{\text{hfs}} | 1s2p^1P_1 \rangle \\ - 2\sin\theta\cos\theta \langle 1s2p^3P_1 | \mathcal{H}_{\text{hfs}} | 1s2p^1P_1 \rangle.$$

For the first term, the matrix element of the magnetic dipole hyperfine interaction operator is non-zero for the orbital-dipole term only. For the second term, one needs to consider only the Fermi contact hyperfine interaction, which is the largest of the three magnetic dipole hyperfine interactions for this case. Thus the hyperfine energies can be written as

$$\Delta E_{\text{hfs}} = \left(\frac{\mu_0}{4\pi} \right) \left(2\mu_B \frac{\mu_I}{I} \right) \\ \times \left\{ \cos^2\theta \langle 1s2p^1P_1 | I \cdot \sum_i \frac{\mathbf{l}_i}{r_i^3} | 1s2p^1P_1 \rangle \right. \\ \left. - 2\sin\theta\cos\theta \langle 1s2p^3P_1 | \frac{8\pi}{3} I \cdot \sum_i \mathbf{s}_i \delta^3(\mathbf{r}_i) | 1s2p^1P_1 \rangle \right\}.$$

The first matrix element is evaluated using a screened hydrogenic approximation yielding the result

$$\langle 1s2p^1P_1 | I \cdot \sum_i \frac{\mathbf{l}_i}{r_i^3} | 1s2p^1P_1 \rangle = \\ \frac{1}{2} \left[F(F+1) - I(I+1) - J(J+1) \right] \langle 1s2p^1P_1 | \frac{1}{r_{2p}^3} | 1s2p^1P_1 \rangle$$

where

$$\langle 1s2p^1P_1 | \frac{1}{r_{2p}^3} | 1s2p^1P_1 \rangle = \frac{1}{l(l+\frac{1}{2})(l+1)} \frac{1}{n^3} \left[\frac{Z}{a_0} \right]^3$$

is evaluated with $Z=3$, $n=2$, and $l=1$.⁷⁷

The second matrix element in the expression for ΔE_{hfs} can be evaluated using the

⁷⁷G.K. Woodgate, *op. cit.*, Chapter 2.

Wigner–Eckart theorem and tensor product identities.⁷⁸

$$\begin{aligned}
 \langle 1s2p \text{ LSJFM}_F | s_1 \delta^3(r_1) + s_2 \delta^3(r_2) | 1s2p \text{ L'S'I'F'M}'_F \rangle = \\
 \delta_{FF'} \delta_{M_F M'_F} \delta_{II'} \delta_{LL'} (-1)^{F+I+J'+2S'+J+L+1} \\
 \times \sqrt{\frac{I(I+1)(2I+1) 3(2J+1)(2J'+1) 3(2S+1)(2S'+1)}{6(2L+1)}} \\
 \times \begin{Bmatrix} J & 1 & J' \\ I' & F & I \end{Bmatrix} \begin{Bmatrix} J' & S' & L \\ S & J & 1 \end{Bmatrix} \begin{Bmatrix} s_2 & 1 & s_2' \\ S' & s_1 & S \end{Bmatrix} \\
 \times \langle 1s2p \text{ L} || \delta^3(r_1) || 1s2p \text{ L}' \rangle.
 \end{aligned}$$

The reduced matrix element involving the Dirac delta function can be written in terms of the usual matrix element using the Wigner–Eckart theorem and then evaluated using hydrogenic wavefunctions so that

$$\langle 1s2p \text{ L} || \delta^3(r_1) || 1s2p \text{ L}' \rangle = \sqrt{3} \frac{Z^3}{\pi a_0^3 n^3}$$

evaluated with $Z=4$ and $n=1$.

The resulting hyperfine structure is tabulated below

F	ΔE_{od} (MHz)	ΔE_{fc} (MHz)	ΔE_{hfs} (MHz)
5/2	-126.4	23.1	-103.3
3/2	84.3	-15.4	68.9
1/2	210.7	-38.5	172.2

where

$\Delta E_{od} = \cos^2 \theta \langle 1s2p^1P_1 | \mathcal{H}_d | 1s2p^1P_1 \rangle$ is the hyperfine energy due to the orbital–dipole interaction in the $1s2p^1P_1$ state,

$\Delta E_{fc} = -2\sin\theta\cos\theta \langle 1s2p^3P_1 | \mathcal{H}_c | 1s2p^1P_1 \rangle$ is the hyperfine energy arising from the Fermi contact interaction and the singlet–triplet mixing,

and

$\Delta E_{hfs} = \Delta E_{od} + \Delta E_{fc}$ is the total hyperfine energy.

⁷⁸A. Messiah, *Quantum Mechanics Volume II*, (Wiley, New York, 1962), p. 1076.

Calculation of the Relative Intensities of Fluorescence

The relative intensities of the observed fluorescence from the $1s2p^1P_1$ state can be calculated from the formula

$$|\langle aLSIJM_F | d_{\kappa}^{(1)} | a'L'S'IJ'M_F' \rangle|^2 = \\ (2F+1)(2F'+1) \begin{pmatrix} F & 1 & F' \\ -M_F & \Delta M_F & M_F' \end{pmatrix}^2 \begin{Bmatrix} J & 1 & J' \\ F' & 1 & F \end{Bmatrix}^2 \\ \times |\langle 1s2pLSIJ || d^{(1)} || 1s2sL'S'IJ' \rangle|^2$$

where $d_{\kappa}^{(1)}$ is the κ th component of the spherical tensor representation of the electric dipole operator and $\kappa = M_F - M_F'$. For laser excitation from the $1s2s^1S_0$ level to the $1s2p^1P_1$ level and fluorescence from the $1s2p^1P_1$ level to the $1s^2^1S_0$ ground state, the quantum numbers a and a' are defined as

Process	a	a'
excitation	$1s2p$	$1s2s$
fluorescence	$1s^2$	$1s2p$

The axis of quantization is chosen to be the direction of polarization of the laser light. Thus for electric dipole transitions driven by the laser from the $1s2s^1S_0$ state to the $1s2p^1P_1$ state the selection rule is $\Delta M_F = 0$. For fluorescence from the $1s2p^1P_1$ level to the ground state $1s^2^1S_0$ level, $\Delta M_F = 0, \pm 1$. The fluorescence is viewed by an MCPA with a solid angle of 0.256 steradian at an average angle 90° from the axis of quantization. For fluorescence where $\Delta M_F = 0$, the intensity varies as $\sin^2\theta$ while for fluorescence where $\Delta M_F = \pm 1$, the intensity varies as $\frac{1}{2}(1+\cos^2\theta)$ where θ is the viewing angle from the axis of quantization. Integrating these functions over the solid angle subtended by the MCPA yields the following angular dependence for a point source:

ΔM_F	Angular Factor
0	0.2372
± 1	0.1232

Note that the ratio of the angular factors is very nearly 2:1, which is the value for

$$\theta = 90^\circ.$$

With this information in hand and assuming uniform population of all $1s2s^1S_0$ $F = 3/2$ magnetic sublevels, one can calculate the relative population of the $1s2p^1P_1$ level produced by laser excitation as

F	M_F	Relative Population
5/2	$\pm 3/2$	0.2
5/2	$\pm 1/2$	0.3
3/2	$\pm 3/2$	0.3
3/2	$\pm 1/2$	0.0333
1/2	$\pm 1/2$	0.1667

and the following relative intensities of fluorescence from the $F = 5/2$, $3/2$, and $1/2$ states of the $1s2p^1P_1$ level as

F	Relative Intensity
5/2	0.5070
3/2	0.3437
1/2	0.1493.

Using the hyperfine energies calculated above and the relative intensities of fluorescence for the hyperfine components, the centroid of the calculated fluorescence is shifted a mere -3.0 MHz from where it would be in the absence of hyperfine structure. A simple model fitting a single Lorentzian to the sum of three Lorentzian curves separated by the hyperfine energies with amplitudes given by the expected fluorescence intensities yields a frequency shift of -3.0 MHz as expected and a line broadening of 3.0 MHz. The systematic shift in the line centre is of little consequence when compared with the experimental uncertainty and the line broadening is also negligible compared with the broadening from signals observed from fringing fields in the resonance region.

APPENDIX 2

Modelling of Laser-Induced Fluorescence

Consider two atomic states $|1\rangle$, $|2\rangle$ coupled by the electric field of a laser. The oscillating electric field drives transitions from the $|1\rangle$ state to the $|2\rangle$ state which is damped by fluorescence to an independent third state $|3\rangle$. The electric dipole interaction of the electric field $\mathbf{E}$, of the light with the atom is defined to be

$$\begin{aligned}\mathcal{V} &= \sum_i e \mathbf{r}_i \cdot \mathbf{E} \\ &= \sum_i e \mathbf{r}_i \cdot \hat{\mathbf{e}}_z E_0 \cos \omega t\end{aligned}$$

assuming a sinusoidally varying electric field. The matrix element of the electric dipole interaction between states $|1\rangle$ and $|2\rangle$ in the interaction picture can then be written as

$$\langle 2 | \mathcal{V} | 1 \rangle_I = e^{i(E_2 - E_1)t/\hbar} \langle 2 | \mathcal{V} | 1 \rangle$$

where

$$\langle 2 | \mathcal{V} | 1 \rangle = E_0 \cos \omega t \langle 2 | \sum_i e \mathbf{r}_i \cdot \hat{\mathbf{e}}_z | 1 \rangle = E_0 \cos \omega t d_{21}$$

and E_1 is the energy of the i th state. Therefore, the matrix element for the operator $\mathcal{V}$ can be rewritten as

$$\langle 2 | \mathcal{V} | 1 \rangle_I = \hbar b e^{i\Delta\omega t}$$

with the following definitions:

$$\begin{aligned}\Delta\omega &= \frac{(E_2 - E_1)}{\hbar} - \omega \\ &= \omega_{21} - \omega \\ 2b &= \frac{E_0 d_{21}}{\hbar}.\end{aligned}$$

The $e^{-i\omega t}$ term has been omitted since it is far off resonance (rotating wave approximation). The quantity $2b$ is the usual Rabi frequency.⁷⁹

If spontaneous emission is ignored from state $|2\rangle$ to state $|1\rangle$ but is included

⁷⁹N.F. Ramsey, *Molecular Beams*, (Clarendon, Oxford, 1956), Chapter V.

from $|2\rangle$ to state $|3\rangle$, then the equations of motion for the Schrödinger equation in the interaction picture are given by

$$i\dot{a}_1 = b e^{i\Delta\omega t} a_2$$

and

$$i\dot{a}_2 = b e^{-i\Delta\omega t} a_1 - i \frac{\Gamma}{2} a_2.$$

Here, $|a_i(t)|^2$ is defined as the probability of state $|i\rangle$ being populated at time t and Γ is the transition rate (A coefficient) for spontaneous emission from state $|2\rangle$ to state $|3\rangle$. With the transformation

$$a_1 = e^{i\Delta\omega t/2} c_1$$

and

$$a_2 = e^{-i\Delta\omega t/2} c_2$$

the explicit time dependence in the coupled differential equations for a_1 and a_2 can be eliminated, and the resulting equations can be solved exactly. These equations are

$$i\dot{c}_1 = \frac{\Delta\omega}{2} c_1 + b c_2$$

and

$$i\dot{c}_2 = -\frac{\Delta\omega}{2} c_2 + b c_1 - i \frac{\Gamma}{2} c_2.$$

With these boundary conditions,

$$c_1(0) = 1 \quad \text{and}$$

$$c_2(0) = 0,$$

the solutions to the differential equations are

$$c_1(t) = \frac{1}{2} e^{-\Gamma t/4} \left\{ \left[1 - \frac{1}{a} \left[\Delta\omega + i \frac{\Gamma}{2} \right] \right] e^{iat/2} + \left[1 + \frac{1}{a} \left[\Delta\omega + i \frac{\Gamma}{2} \right] \right] e^{-iat/2} \right\}$$

and

$$c_2(t) = -\frac{b}{a} e^{-\Gamma t/4} \left\{ e^{iat/2} - e^{-iat/2} \right\}$$

where

$$a^2 = (\Delta\omega + i\Gamma/2)^2 + (2b)^2.$$

When $(2b) \ll (\Gamma/2)$ (strong damping), a can be expanded as follows:

$$a \approx (\Delta\omega + i\Gamma/2)^2 \left[1 + \frac{1}{2} \frac{(2b)^2}{(\Delta\omega + \Gamma/2)^2} \right]$$

so that

$$\frac{1}{a}(\Delta\omega + i\Gamma/2) \approx 1 - \frac{1}{2} \frac{(2b)^2}{(\Delta\omega + \Gamma/2)^2}.$$

This can be substituted into the expression for $c_1(t)$, yielding the following expression for $|c_1(t)|^2$:

$$|c_1(t)|^2 \sim e^{-\left[\frac{\Gamma/2}{(\Delta\omega)^2 + (\Gamma/2)^2} \right] 2b^2 t}.$$

If the semiclassical approximation is made that the ground state depletion is small,

$|c_1(t)|^2$ can be expanded to get

$$|c_1(t)|^2 \sim 1 - \left[\frac{\Gamma/2}{(\Delta\omega)^2 + (\Gamma/2)^2} \right] 2b^2 t$$

or

$$|c_1(t)|^2 \sim 1 - \left[\frac{1}{1 + (2\Delta\omega/\Gamma)^2} \right] 2bt \frac{b}{(\Gamma/2)}.$$

This formula predicts a Lorentzian fluorescence signal as a function of laser frequency offset whose full-width at half-height is Γ and an amplitude proportional to $2bt \frac{b}{(\Gamma/2)}$. Therefore in the limit of extreme damping ($\Gamma \rightarrow \infty$), the fluorescence disappears. This is counter-intuitive since one would expect that as the damping from the $|2\rangle$ state to the $|3\rangle$ state is increased, there would be a corresponding increase in fluorescence signal.

A naïve picture assumes the average rate of population transfer from the $|1\rangle$ state to the $|2\rangle$ state depends only on $2bt$ (that is, laser intensity and interaction time) and not on γ (the damping of state $|2\rangle$). If one considers the analogy of the motion of a damped classical oscillator undergoing forced oscillations⁸⁰ with the above atomic system, the apparent paradox can be explained. The equation of

⁸⁰F.S. Crawford Jr., *Waves Berkeley Physics Course - Volume 3*, (McGraw-Hill, New York, 1968), Chapter 3.

motion for the damped oscillator is given by

$$M\ddot{x}(t) + M\Gamma\dot{x}(t) + M\omega_0^2x(t) = F_0\cos\omega t$$

where

x is the displacement of the oscillator

M is the mass of the oscillator

Γ is the damping constant

ω_0 is the resonant frequency of the undamped oscillator

F_0 is the amplitude of the driving force

ω is the frequency of the driving force.

The steady state solution $x_s(t)$ can be written as

$$x_s(t) = A_{ab} \sin\omega t + A_{el} \cos\omega t$$

where A_{ab} and A_{el} are the absorptive and elastic amplitudes respectively defined by

$$A_{ab} = \frac{F_0}{M} \frac{\Gamma\omega}{[(\omega_0^2 - \omega^2)^2 + \Gamma^2\omega^2]}$$

$$A_{el} = \frac{F_0}{M} \frac{(\omega_0^2 - \omega^2)}{[(\omega_0^2 - \omega^2)^2 + \Gamma^2\omega^2]}$$

The time-averaged power loss to the damping is given by

$$P_d = \frac{1}{2}M\Gamma\omega^2[A_{ab}^2 + A_{el}^2]$$

which can be shown to be equal to the time-averaged steady-state input power

$$P_i = \frac{1}{2}F_0\omega A_{ab}.$$

Thus for the limit of infinite damping ($\Gamma \rightarrow \infty$), no power can be absorbed by the oscillator and no power is dissipated via damping. That is, if the damping is so large that the mass fails to oscillate when the external force is applied, no power is absorbed and no power is dissipated by damping. This also holds true for the heavily damped fluorescence of the atomic system. It is untrue that the rate of population transfer remains constant as the damping is increased. The absorbed power from the electric field of the laser decreases with increased damping and hence the dissipated power (fluorescence) also decreases.

It is necessary to calculate the quantity $2bt$ to determine the percentage of transferred metastable population from state $|1\rangle$ to state $|2\rangle$. The value of b is determined by the laser power and the Einstein coefficient A_{21} for spontaneous emission from the $1s2p^1P_1$ level to the $1s2s^1S_0$ level. The square of the electric dipole matrix element between these two states is written in terms of A_{21} as follows:

$$d_{21}^2 = 3\pi\epsilon_0 \frac{\hbar^4 c^3}{\omega_0^3} \frac{A_{21}}{\sqrt{3}}$$

where

$$A_{21} = 0.0677 \times 10^8 \text{ s}^{-1} \text{ (theoretical value}^{81}\text{)}$$

$$\omega_0 = 3.07 \times 10^{15} \text{ rad/s (the angular frequency of the } 2^1S - 2^1P \text{ transition)}$$

so that

$$d_{21} = 4.90 \times 10^{-30} \text{ C-m.}$$

The electric field amplitude (E_0) of the laser can be determined from the average power per unit area, S_{av} , which is given by

$$S_{av} = \frac{1}{2}\epsilon_0 c E_0^2.$$

The typical laser power in the resonance region is 300 mW in a beam whose cross sectional area is $\sim 3.17 \times 10^{-5} \text{ m}^2$. This corresponds to an electric field of $2.67 \times 10^3 \text{ V/m}$ (assuming a uniform energy density). The b coefficient is given in terms of the following quantities:

$$b = \frac{E_0 d_{21}}{2\hbar} = 6.21 \times 10^7 \text{ rad/s.}$$

The effective interaction time t for the laser and ion beams is given by

$$t = \frac{d}{\beta c} = 4.37 \times 10^{-8} \text{ s}$$

where d (3 cm) is the length over which the velocity is nearly uniform in the post-accelerator electrode and β (2.29×10^{-3}) is the fraction of the speed of light at which the ions are travelling. The fraction of metastable ions which fluoresce is

⁸¹A.W. Weiss in *Atomic Transition Probabilities Hydrogen through Neon*, (U.S. Department of Commerce and National Bureau of Standards, Washington, 1966), p. 25.

given by the amplitude (A) of the Lorentzian

$$A = 2bt \frac{b}{(\Gamma/2)}.$$

Using the calculated values of b and t as well as the theoretical prediction of Γ , the fraction of metastable ions which fluoresce is 0.55%. This result also predicts that the observed fluorescence signal varies linearly with laser power motivating the normalization of fluorescence signal to the recorded laser power.

APPENDIX 3

Lineshape Modelling

Aggregate spectra, which are formed by overlapping many individual spectra, show slight asymmetries for both parallel (*P*) and anti-parallel (*A*) geometries. These asymmetries, which are reversed for the two geometries, are not apparent in individual spectra due to the reduced signal-to-noise ratio. Individual spectra for a particular geometry are combined using the parameters of the fit of a symmetric Lorentzian (see Chapter 5) to form a histogram of the aggregate spectra by overlapping peak centres at a bin arbitrarily assigned the value 0 GHz. In addition to matching peak centres, the recorded background counts are subtracted from each individual spectrum before the spectrum is sorted into histogram bins. The final histogram is then normalized using the fit peak amplitudes as a weighting factor and only the central ~ 30 GHz region, where the majority of spectra overlap, is used for further analysis. The resulting histograms are shown in Figure 9 and 10 along with a fitted symmetric Lorentzian curve to highlight the asymmetries. The resulting histograms have a shape which can be characterized as a Lorentzian with unequal left and right half-widths. The results of a non-linear least-squares fit of a Lorentzian with unequal left and right half-widths is summarized in the following table:

<u>Geometry</u>	<u>Left Half-Width</u> (GHz)	<u>Right Half-Width</u> (GHz)
<i>P</i>	12.42(9)	10.58(7)
<i>A</i>	10.48(7)	12.37(7)

which clearly shows the reversal of the asymmetry between *P* and *A* spectra.

This asymmetry can be explained by considering the viewing geometry of the resonance region as well as the potentials inside the post-accelerator. The resonance region geometry has been illustrated earlier (See Chapter 4, Figure 5).

Figure 9. *Histogram of aggregate spectra for parallel geometry.*

A histogram of the aggregate spectra for parallel geometry is produced by overlapping the peak centres of individual spectra. For parallel geometry, the histogram is clearly broader to lower frequency. A systematic trend is evident in the graph of the residuals for a symmetric Lorentzian function (the smooth curve in the upper graph) as a result of the asymmetry in the histogram.

Figure 10. *Histogram of aggregate spectra for anti-parallel geometry.*

A histogram of the aggregate spectra for anti-parallel geometry is produced by overlapping the peak centres of individual spectra. For anti-parallel geometry, the histogram is clearly broader to higher frequency. A systematic trend is evident in the graph of the residuals for a symmetric Lorentzian function (the smooth curve in the upper graph) as a result of the asymmetry in the histogram.

The potential distribution near the ends of two concentric cylinders is calculated by Spangenberg.⁸² Typically, ~ -1000 VDC is placed on the central electrode so that the potential on the axis in the post-accelerator electrode reaches the full -1000 V about 2.25 cm (the diameter of the post-accelerator electrode) from either grounded electrode. Similarly this potential penetrates into the end of the grounded electrode approximately the same amount. The region of potential gradient is viewed by part or all of the MCPA.

The effect of viewing the region of fringing fields in the post-accelerator is that the ions in the fringing field interact with the laser light with a range of Doppler shifts and the resulting fluorescence is detected by the MCPA. Using geometry where the laser and ion beams are anti-parallel for an example, the frequency of the laser light, as seen in the rest frame of the ions, is blue-shifted into resonance with the $2^1S - 2^1P$ transition. As the laser is scanned to higher frequency, the ions in the fringing field, where the Doppler shift is less than at the centre of the post-accelerator, become resonant with the laser light at a frequency greater than those ions which fluoresce at the centre of the post-accelerator region. The majority of the fluorescence is recorded from those ions which fluoresce at the centre of the post-accelerator. Since the magnitude of the Doppler shift viewed by the ions in the fringing fields is always less than at the centre of the post-accelerator, the fluorescence emitted in the fringing fields is recorded for frequencies in the scan larger than the peak of the resonance. This produces an asymmetric Lorentzian-like signal which is broader to higher frequency.

For parallel geometry, the ion and laser beams are propagating in the same direction so that the laser frequency is red-shifted in the reference frame of the ions. Since the magnitude of the Doppler shift for ions in the fringing fields is always less

⁸²K.R. Spangenberg, *Vacuum Tubes*, (McGraw Hill, New York, 1948), Chapter 13.

than near the centre of the post-accelerator, the ions which fluoresce in the fringing fields are resonant for frequencies less than those ions which fluoresce at the centre. Therefore, the signal for parallel overlap geometry is broadened to lower frequency. Since this asymmetry reverses from parallel to anti-parallel overlap geometry any small distortion in the determination of the line centres of individual spectra cancel when the geometric mean is calculated.

The degree of cancellation can be calculated by considering the geometric mean of the following observed peak centres:

$$\sigma_A' = \sigma_A + \delta\sigma$$

$$\sigma_P' = \sigma_P - \delta\sigma$$

where

σ_i' is the observed peak centre for either *P* or *A* geometry

σ_i is the true peak centre in the absence of asymmetric broadening

$\delta\sigma$ is the amount of peak centre shift.

The Doppler-free wavenumber of the $2^1S - 2^1P$ transition (σ_0) is given by

$$\begin{aligned}\sigma_0 &= \sqrt{\sigma_P' \sigma_A'} \\ &\cong \sqrt{\sigma_P \sigma_A} \left\{ 1 + \left[\frac{\sigma_P - \sigma_A}{\sigma_P \sigma_A} \right] \frac{\delta\sigma}{2} \right\}.\end{aligned}$$

Therefore, the shift in the Doppler-free wavenumber $\Delta\sigma_0$ is

$$\Delta\sigma_0 = \sqrt{\sigma_P \sigma_A} \left[\frac{\sigma_P - \sigma_A}{\sigma_P \sigma_A} \right] \frac{\delta\sigma}{2}$$

so that the calculated shift using typical values for σ_P (16313.18 cm^{-1}), σ_A (16240.45 cm^{-1}), and $\delta\sigma$ (0.133 cm^{-1} , estimated from a model described below) is

$$\Delta\sigma_0 = 0.0003 \text{ cm}^{-1}.$$

This systematic shift is negligible compared to the present experimental precision.

The lineshape modelling program establishes a cylindrical coordinate system with the *z*-axis along the axis of the post-accelerator and the origin placed equidistant from either end of the post-accelerator, directly in front of the MCPA.

At least part of the MCPA is capable of viewing fluorescence emitted along the z -axis for values of z , $-3.6 \text{ cm} \leq z \leq 3.6 \text{ cm}$. The z -axis is divided into small steps (typically .25 mm or less) and at each step a double integration (in θ and ϕ) is performed over the portion of the MCPA surface which is not vignetted by the electrodes of the post-accelerator. The MCPA detection efficiency is angle dependent⁸³ which further complicates the calculation. The detection efficiencies for MCPA's are not widely measured and few measurements include angular information. The best measurement of the angular dependence for a wavelength near 100 Å is a measurement by Parkes *et al.*⁸⁴ at 68 Å. These results are tabulated from the graph and interpolated using a spline and are included as a factor in the double integration. Each step along the z -axis is converted from a distance to the origin into a frequency offset from the Doppler-shifted frequency as viewed at the origin. Again, the potential distribution in the post-accelerator is tabulated from figures and interpolated using a spline. Using the voltage for a given value of z along the z -axis, the frequency offset is calculated using the relativistic Doppler formula

$$\nu(V) = \gamma(1 \pm \beta(V) \cos\theta) \nu_L$$

where

$$\beta(V) = v/c = \left\{ \frac{2qV}{mc^2} \right\}^{\frac{1}{2}} \text{ is the ion velocity compared with the speed of light}$$

$$\gamma = (1 - \beta^2)^{-\frac{1}{2}} \cong 1$$

ν_L is the frequency of the laser light measured in the laboratory.

The frequency offset $\Delta\nu$, is then

$$\Delta\nu = \pm \Delta\beta \nu_L$$

(assuming $\cos\theta = 1$) where $\Delta\beta$ is given by

$$\Delta\beta = \beta(z) - \beta(z=0)$$

⁸³O.H.W. Siegmund and Roger F. Malina, *Detection of EUV and soft X-rays with microchannel plates: A Review*, Space Sciences Laboratory, University of California, Berkeley (unpublished).

⁸⁴W. Parkes, R. Gott, and K.A. Pounds, IEEE Trans. Nucl. Sci. **17**, 360 (1970).

and the positive/negative signs are for *P/A* geometry respectively.

The resulting curve of the double integration over the MCPA face (including angular detection efficiencies) versus frequency offset is the effective Doppler distribution as a function of frequency offset and is illustrated in Figure 11. The final modelled signal is produced by convolving the effective Doppler distribution with the sum of three Lorentzian functions whose relative amplitudes and peak centres are determined by the hyperfine structure of the 2^1P state (see Appendix 1). The full-width at half-height of the Lorentzian functions is 19.42 GHz which corresponds to a theoretical value of $1.220 \times 10^{11} \text{ s}^{-1}$ for the *A* coefficient of the 2^1P level.⁸⁵ The peak centre of the lineshape model (the convolution of the hyperfine structure with the effective Doppler distribution) is shifted by $\sim 4 \text{ GHz}$ (0.133 cm^{-1}) from the peak centre of the hyperfine structure alone. The results of the convolution are plotted with the aggregate spectra in Figures 12 and 13. A NLLS fit was used to adjust the peak amplitude and peak centre of the model relative to the aggregate data. No other adjustable parameters were used to enhance the agreement between the data and model. A symmetric Lorentzian whose peak amplitude, peak centre, and width are fit to the aggregate spectra are also shown in Figures 9 and 10. The associated plots of the residuals between model and data shows systematic deviations in the fit of the symmetric Lorentzian fits due to the asymmetries present in the data. Although the effect is very small, it can be seen in the sinusoidal variation of the sign of the residuals. This effect is not seen in the residuals of the convolved model which clearly represents a better fit to the aggregate spectra. For individual spectra, a simple symmetric Lorentzian produces an acceptable fit because of the larger signal-to-noise ratio. The results of a fit of a symmetric Lorentzian function to a typical signal is shown in Figure 14. The

⁸⁵G.W.F. Drake, Phys. Rev. A 19, 1387 (1979).

Figure 11. *Effective Doppler distribution as a function of frequency offset.*

The effective Doppler distribution is calculated as a double integration over the MCPA face including angular detection efficiencies. The frequency scale is calculated from the relativistic Doppler formula using the potential distribution inside the electrodes of the post-accelerator.

Figure 12. *Modelled lineshape for parallel geometry.*

The modelled lineshape (smooth curve in upper graph) is plotted with the histogram of the aggregate P spectra. Only a horizontal shift and an overall scaling factor is used to fit the model to the data. The resulting residuals are clearly more uniform for the modelled lineshape than for the symmetric Lorentzian function (see Figure 9).

Figure 13. *Modelled lineshape for anti-parallel geometry.*

The modelled lineshape (smooth curve in upper graph) is plotted with the histogram of the aggregate *A* spectra. Only a horizontal shift and an overall scaling factor is used to fit the model to the data. The resulting residuals are clearly more uniform for the modelled lineshape than for the symmetric Lorentzian function (see Figure 10).

Figure 14. *Typical parallel resonance signal with symmetric Lorentzian function.*

A symmetric Lorentzian function is fit to a typical resonance signal. No asymmetry in the resonance signal can be seen in the graph of residuals because of the larger statistical uncertainties associated with individual spectra. The chi-square per degree of freedom for the NLLS fit is 1.164.

chi-square per degree of freedom is 1.164 using Poisson statistics to calculate the weights for the reduced chi-square. The same spectrum is also shown with a two-parameter fit (peak amplitude and centre) of the convolved model in Figure 15. Using the same weights, the chi-square per degree of freedom is 1.168. Either fitting function (the symmetric Lorentzian or convolved model) provides an adequate fit to find the peak centre of the UV signal because of the rather large scatter in a single UV spectrum.

The excellent agreement between the modelled lineshape and the aggregate spectra suggests a value for the A coefficient of the 2^1P level in agreement with that predicted by theory. This model depends greatly on the sensitivity of the MCPA as a function of angle which is difficult to measure and has not been measured at the wavelength observed in this experiment. Even though this model has been highly successful in reproducing observed spectra, one can not state with any confidence an exact value for the lifetime but one can remark that the width of the observed resonance can serve to set a lower limit of 7.1(5) ps on the lifetime of the 2^1P state. If a more accurate measurement is desired, an improved post-accelerator region would be necessary to eliminate fringing fields in the viewing region of the MCPA.

Figure 15. *Typical parallel resonance signal with modelled lineshape.*

The modelled lineshape is fit to the same resonance signal shown in Figure 14 using only a horizontal offset and overall scaling parameter. The large statistical uncertainties associated with individual spectra mask the expected asymmetry in the data. The "goodness of fit" for the NLLS fit as gauged by the chi-square per degree of freedom is the same as that for a symmetric Lorentzian function.

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