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ABSTRACT.

The two-photon decay of the $2^2S_1$ state of atomic hydrogen has been directly observed using a fast coincidence counting technique. A hydrogen metastable beam apparatus has been constructed using a duoplasmatron proton source and charge-exchange in cesium vapour, and its characteristics measured. Tests are described which firmly establish the existence of the two-photon decay of atomic hydrogen, and the observed lifetime due to this process is consistent with the predicted value. The angular distribution of the emitted photons does not disagree with the predicted $(1 + \cos^2\theta)$ factor.
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I. Introduction.

I.1. Relevant history of the hydrogen atom.

The first real understanding of atomic structure began with Bohr's theory of the hydrogen atom in 1913\(^1\), which at the time satisfactorily explained the gross structure of the spectral lines. Sommerfeld's fine structure theory of 1916, was confirmed in 1926 by Paschen's observations of the fine structure of singly ionized helium and of the Balmer series of hydrogen\(^2\). After the problems of the intensities were resolved by the introduction of Dirac's theory of the electron and the electron spin, attention was concentrated on the fine structure of the Balmer alpha line. A slight shift in one of the fine structure components seemed to exist, but the resolution of the optical observations was not good enough to show a real discrepancy with the theory. In 1947, the historic experiment of Lamb and Retherford\(^3\) conclusively confirmed the existence of a discrepancy between the Dirac theory and the Balmer alpha fine structure. Using an atomic beam technique and microwave spectroscopy, they found that the \(2S_{\frac{1}{2}}\) and the \(2P_{\frac{1}{2}}\) levels of atomic hydrogen were not degenerate as predicted by the Dirac theory, but the \(2S_{\frac{1}{2}}\) level was shifted upwards in energy by about \(4.37 \times 10^{-6}\) eV, or 1.058 GHz. Bethe\(^4\) immediately interpreted this result in terms of the self-interaction of the electron in non-relativistic, second-order perturbation theory,
obtaining approximately the observed value for what is now known as the Lamb shift. Since then, the development of relativistic quantum electrodynamics by Schwinger, Tomonaga and Feynman has given an almost complete picture of the atomic hydrogen energy spectrum.

1.2. The metastable state.

Before the Lamb shift was discovered, it was realised that the $2S_\frac{1}{2}$ state of atomic hydrogen should be metastable, in the absence of perturbing effects. The only state below it in energy was the ground state, on the basis of the Dirac theory. Electric dipole transitions to the ground state are strictly forbidden by the parity selection rule, as are electric quadrupole transitions, and although magnetic dipole transitions are not strictly forbidden if the exact Dirac wavefunctions are used, the lifetime due to this mode of decay was calculated by Breit and Teller to be about two days.

Maria Goeppert-Mayer in 1931 developed the theory of two-photon decay, through which the parity selection rule, for transitions between states of the same parity, can be satisfied by the introduction of virtual transitions to intermediate states of different parity. In 1940, Breit and Teller examined the possible modes of decay of the $2S_\frac{1}{2}$ state of atomic hydrogen, and found that in the absence of perturbing effects, the lifetime was limited by two-photon decay to the ground state, giving a transition probability of about $7 \text{ s}^{-1}$. However, they
assumed the degeneracy of the $2S_\frac{1}{2}$ and the $2P_\frac{1}{2}$ states, and so even very small electric fields would be sufficient to mix them by the Stark effect. Since the $2P_\frac{1}{2}$ state was known to have a lifetime of about $1.6 \times 10^{-9}$ s, it appeared that in practice the $2S_\frac{1}{2}$ state would not be metastable in the laboratory.

The discovery of the Lamb shift showed that the lifetime of the $2S_\frac{1}{2}$ state was considerably longer in an electric field, (e.g., in a field of 2000 V,m$^{-1}$, the lifetime is of the order of a microsecond), and that metastable beams could be produced and studied in the laboratory.

Because the simultaneous emission of two photons in a single transition gives rise to a form of continuous radiation, the two-photon decay was invoked by Spitzer and Greenstein\textsuperscript{7} in 1951 to explain the spectra of certain gaseous planetary nebulae. They calculated the spectrum and transition rate of two-photon emission from the metastable state. Their value for the transition probability is in good agreement with the more accurate calculation of Shapiro and Breit\textsuperscript{8}, and the more recent calculation of Klarsfeld\textsuperscript{9} of $8.2283 \times 10^6$ s$^{-1}$. They also found that the two-photon decay gives rise to a 'bluish' continuum in the visible region of the spectrum, which appeared consistent with the astronomical observations.

Salpeter\textsuperscript{10} and Feinberg\textsuperscript{11} pointed out in 1958, that parity and time-reversal violating effects could affect the lifetime of the metastable state. Such an effect could be a permanent electron dipole moment, or as shown
by Zeldovich and Perelemov\textsuperscript{12}, the presence of neutral currents in the weak interaction, which would violate parity. Evidence for the existence of neutral currents has recently been reported\textsuperscript{13}.

The work reported in this thesis is the first observation of the unperturbed two-photon decay of the metastable state of atomic hydrogen.

1.3. The natural decay of metastable states.

This section describes some of the most relevant observations of the natural decays of metastable hydrogenlike and heliumlike atoms. Heliumlike atoms have two metastable states, the $2^1S_0$ and the $2^3S_1$ states. The $2^1S_0$ state decays by two-photon emission, and the $2^3S_1$ decays predominantly by single photon emission through the relativistic magnetic dipole process\textsuperscript{14}. Although the $2^3P_2$ state of neutral helium decays by an allowed electric dipole transition, it has been pointed out that for values of $Z$ greater than about 17, the heliumlike ions decay predominantly by a single photon magnetic quadrupole process\textsuperscript{15}.

The first attempt to observe the spontaneous decay of a metastable atom was made by Fite et al.\textsuperscript{16} in 1959. They tried to measure the lifetime of the $2S_g$ state of atomic hydrogen due to single photon decay, caused by parity non-conserving effects such as those discussed in the last section. They used a thermal beam of atomic hydrogen excited to the metastable state by
electron impact. By monitoring the Lyman-alpha signal produced by the metastable atoms in a field-free region, and with a strong electric field applied, they succeeded in setting a lower limit to the single photon decay lifetime of $2.4 \times 10^{-3}$ s. This is about 50 times shorter than the predicted two-photon lifetime.

The first direct observation of a two-photon decay was made by Lipeles et al., in 1965. They observed, in coincidence, the spontaneous two-photon decay of the $2S_1$ state of singly ionized helium. An electron bombardment source was used to ionize and excite the atoms which were then accelerated to an energy of about 15 eV. The ion beam was modulated by rf quenching at the Lamb shift frequency so that the signal originating from the metastables could be separated from the background. Since the two-photon spectrum of He$^+$ is predicted to extend from 30.4 nm to infinity, open-ended photomultiplier tubes were used inside the vacuum system. A delayed coincidence technique was employed. The beam modulation frequency was 208 Hz and the time spectrum with the quench field on and off were recorded in separate sections of the multi-channel analyser memory. Lipeles et al. obtained results consistent with the predicted $(1 + \cos^2 \theta)$ angular distribution, and in a later publication the same group reported a rough spectral distribution, using broad-band filters, not inconsistent with the theoretical predictions.

In 1972, a group from the same laboratory directly measured the natural lifetime of the same state of helium
by a decay-in-flight experiment. A similar ion source and quenching system was used, connected to an 8-metre length of pipe fitted with a movable Auger detector. After corrections were made for collisional quenching, they measured a lifetime of \(2.04 \pm 0.81, -0.34\) ms, which is consistent with the predicted two-photon lifetime of \(0.1215 Z^{-6}\) s, or 1.999 ms.

Schmieder and Marrus\(^1\) observed the two-photon decay of the \(2S_{1/2}\) state of hydrogenlike argon in 1970, and measured its lifetime by a beam-foil time-of-flight method. They accelerated argon ions (\(Z = 18\)) in the +14 charge state to an energy of 412 MeV in a heavy ion linear accelerator, and passed them through a thick metal foil from which they emerged distributed in a variety of charge states. The fully stripped ions were magnetically selected and passed through a thin carbon foil in which the probability of single electron capture was high. Thus a beam of hydrogenlike argon (Ar\(^{17+}\)) was produced. The two-photon spectrum of this ion extends from 3.34 keV downwards in energy, so solid-state detectors were used. A delayed coincidence technique was again used with the added advantage that the photon energies could be simultaneously monitored. The sum of the photon energies that formed the coincidences were found to form a peak at about 3.3 keV, indicating that the two-photon decay was indeed being observed. By varying the position of one of the detectors relative to the foil position, and only accepting photon energies in the region of 1.67 keV (two-photon peak), the lifetime was measured to be
(3.54 \pm 0.25) \times 10^{-9} \text{ s}, compared with the predicted value of 3.47 \times 10^{-9} \text{ s}, taking into account the relativistic magnetic dipole decay rate which increases as \( Z^{10} \), and has a lifetime of 1.1 \times 10^{-7} \text{ s} in this case. The mixing of the \( 2S_\frac{1}{2} \) state with the \( 2P_\frac{3}{2} \) state of \( \text{Ar}^{17+} \) which would result from the parity violating effects as mentioned above, was estimated to be less than 6 \times 10^{-4} of the \( 2P_\frac{3}{2} \) state.

In subsequent experiments Marrus and Schmieder\textsuperscript{14} also observed the forbidden decays of the heliumlike ion \( \text{Ar}^{16+} \), the \( 2^1S_0 \rightarrow 1^1S_0 \), the \( 2^3S_1 \rightarrow 1^1S_0 \), and the \( 2^3P_2 \rightarrow 1^1S_0 \) transitions. These results are given in table I.

Table I.

<table>
<thead>
<tr>
<th>State</th>
<th>( t_{\text{expt}} ) (10(^{-9}) s)</th>
<th>( t_{\text{theor}} ) (10(^{-9}) s)</th>
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<tbody>
<tr>
<td>( 2^1S_0 )</td>
<td>2.3 \pm 0.3</td>
<td>2.35</td>
</tr>
<tr>
<td>( 2^3S_1 )</td>
<td>172 \pm 30</td>
<td>210</td>
</tr>
<tr>
<td>( 2^3P_2 )</td>
<td>1.7 \pm 0.3</td>
<td>1.49</td>
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The \( 2^1S_0 \) and the \( 2^3P_2 \) lifetimes are in good agreement with theory, but the \( 2^3S_1 \) lifetime shows a small discrepancy. However, relativistic corrections to the theory, which have not been taken into account in the above theoretical lifetimes, may improve the agreement. Using the same techniques, the lifetime of hydrogenlike sulphur (\( S^{15+} \)) in the \( 2^2S_\frac{1}{2} \) state was found to be, \( 7.3 \pm 0.7 \times 10^{-9} \text{ s} \), in agreement with the predicted value of \( 7.23 \times 10^{-9} \text{ s} \).
Prior and Shugart\textsuperscript{23} in 1971 and Prior\textsuperscript{24} in 1972 measured the lifetimes of the $2^1S_0^o$ and the $2^2S_1^o$ states of singly ionized lithium and singly ionized helium, respectively, using an ion trapping technique. The ions were generated by electron bombardment and were contained by electric and magnetic fields, while an open-ended photomultiplier viewed the storage region. As the excited ions decayed in the trap, an exponential decay curve was formed in a multi-channel scaler coupled to the detector output. In the case of helium, the ion trap could be irradiated with microwaves at the Lamb shift frequency, to ensure that it was the $2^2S_1^o$ state under observation. The Li$^+$ $2^1S_0^o$ state was found to have a lifetime of $(5.03 \pm 0.26) \times 10^{-4}$ s, in good agreement with the theoretical value of $5.13 \times 10^{-4}$ s. The He$^+$ $2^2S_1^o$ lifetime was measured to be $(1.922 \pm 0.082) \times 10^{-3}$ s, also in good agreement with the predicted value of $1.899 \times 10^{-3}$ s. Prior\textsuperscript{24} also calculated, on the basis of the He$^+$ result, that less than $4.7 \times 10^{-5}$ of the $2^2P_1^o$ wavefunction was mixed with the $2^2S_1^o$, an order of magnitude improvement over Marrus and Schmieder's result.

Van Dyck, Johnson and Schugart\textsuperscript{25} measured the lifetime of the $2^1S_0^o$ state of neutral helium in 1971, by direct observation of the metastable atoms. A beam of helium atoms was excited to the $2^1S_0^o$ state by the impact of a short pulse of electrons, after which the atoms drifted through collimating slits, and through an 80% transparency copper grid, from which they caused electrons to be
emitted by the Auger effect. These electrons were collected by an electron multiplier. After passing through a further drift region of about 5 metres, the beam was collected by a copper plate, from which the Auger electrons were counted by another multiplier. The electron beam was pulsed continuously, and the time distribution of the arrival of the Auger electrons recorded. By finding the ratio of the number of counts registered by the second detector to that registered by the first, and plotting this ratio on a semi-log scale as a function of the velocity, a straight line was obtained whose slope was the required lifetime. The result obtained was $(1.97 \pm 1.0) \times 10^{-2}$ s, in agreement with the predicted value of $1.95 \times 10^{-2}$ s.

A similar experiment on the same state of helium was performed by Pearl in 1970, but he obtained a value of $(3.8 \pm 0.8) \times 10^{-2}$ s, in serious disagreement with the value of Van Dyck et al., as well as with theory.

Moos and Woodworth reported in 1973 what must be one of the lowest transition rates ever measured, that for the spontaneous decay of the $2^3S_1 \rightarrow 1^3S_0$ state of neutral helium. This decay is by a relativistic magnetic dipole process, with a predicted lifetime of 7,874 s. They observed the 62.556 nm line of this transition in the afterglow of a flowing helium discharge, with a vacuum uv spectrometer. The metastable density in the discharge tube was determined by measuring the absorption of the 388.9 nm line ($2^3S_1$ to the $3^3P$ states) from a helium resonance lamp.
The $2^3S_1$ lifetime was thus measured to be 4,150 s, with an experimental uncertainty of a factor of three, which overlaps the predicted value.

Although not a spontaneous transition, the observation of stimulated two-photon emission from the metastable state of atomic hydrogen by Bräunlich and Lambropoulos in 1970, is of direct interest to the present work. These authors used a pulsed Nd-glass laser at a wavelength of 1,590 nm to stimulate the corresponding 137.3 nm photon, as well as a photon of wavelength equal to that of the incident photon. The metastable beam was produced by charge exchange of protons in cesium vapour and the emitted short wavelength photon was observed with a vacuum-uv spectrometer. Anti-Stokes Raman scattering of the laser photons was also observed, in which the incident photon was absorbed, and the emitted photon was at the incident photon frequency, plus the Lyman-alpha frequency. These results were in good agreement with the theory of Zernik on optical quenching of the metastable state.

I.4. The present experiment.

Although the theory of the two-photon decay is well established, giving good agreement with a number of experiments, it still remained for the decay to be observed and the lifetime to be measured for the simplest atom - hydrogen. The reason for this anomaly lies in the $z^6$ dependance of the two-photon decay
probability, which means that hydrogen has the lowest
decay rate of all the one-electron atoms.
A number of previous attempts to observe the decay had
failed\textsuperscript{30,31} without any convincing reason. As a result,
it was suspected that the predicted transition
probability of 8.228 \( s^{-1} \) might not hold for hydrogen\textsuperscript{30}.
Although this transition probability has been observed
to hold for all the hydrogenlike atoms investigated
to the limit of experimental accuracy, it must be
admitted that hydrogen is a special case, and so
observation of the two-photon decay of hydrogen and a
measurement of the corresponding lifetime are of
importance and should be investigated.

Previous attempts to observe the hydrogen two-photon
decay utilised a hydrogen oven-electron bombardment type
of metastable source, similar to the type used in the
original Lamb-Retherford experiment. It has been
pointed out by Donnally\textsuperscript{32} that charge exchange of protons
in cesium vapour should give a large cross-section for
capture into the metastable state. This has since been
verified in a number of experiments\textsuperscript{33,34,35}. It was
decided to utilise this type of source, with its
potentially higher metastable densities in a new attempt
to observe the two-photon decay of the \( 2^2S_\frac{1}{2} \) state of
atomic hydrogen.
II. Theory of the decay of the $2S^\frac{1}{2}$ state.

The energy diagram of the relevant states of atomic hydrogen is shown in Fig. 1, (neglecting hyperfine structure). The only states lower in energy than the $2S^\frac{1}{2}$ state are the $2P^\frac{1}{2}$ and the $1S^\frac{1}{2}$ states. The splitting between the $2S^\frac{1}{2}$ and the $2P^\frac{1}{2}$ states is known as the Lamb shift. It will be shown below that of all possible decay modes, the fastest is by the emission of two photons in the electric dipole approximation. Destruction of the metastable state is also discussed.

II.1. General perturbation theory.

The interaction of electrons and photons can be described by Schrödinger's equation,

$$i\hbar \frac{d}{dt}|\Psi\rangle = H|\Psi\rangle,$$

where $|\Psi\rangle$ is the state vector of the whole system, and $H$ is the Hamiltonian of the whole system, which can be written as,

$$H = H^1 + H^2 + H_1$$

$$H_0 = H^1 + H^2$$

where $H^1$ represents the energy of the electrons alone, and $H^2$ the energy of the photons alone, and $H_1$ their interaction.

If $H_1$ is small compared to $H_0$, we can treat it as a perturbation on $H_0$, and so solve the Schrödinger equation independantly for the electrons and the photons.
Fig. 1

Energy level diagram of the first two levels of atomic hydrogen, neglecting hyperfine structure.
where \( |\psi\rangle \) is the state vector of the electrons, and \( |n\rangle \) is the state vector of the photons. Then the state vectors of the Hamiltonian \( H_0 \) are products of the \( |\psi\rangle \) 's, and the \( |n\rangle \) 's. Since \( H_0 \) does not depend explicitly on the time, its eigenfunctions (state vectors) represent stationary states, and the interaction Hamiltonian \( H_1 \) causes transitions between these stationary states. Thus we can write,

\[
|\psi\rangle = |\psi\rangle |n\rangle
\]

The problem is more easily treated in the Interaction picture, where the state vectors and the Hamiltonian are transformed by,

\[
|\psi^I\rangle = e^{iH_0 t/\hbar} |\psi\rangle
\]

\[
H^I = e^{iH_0 t/\hbar} H e^{-iH_0 t/\hbar}
\]

Then the Schrödinger equation can be written as,

\[
\text{i} \hbar \frac{d}{dt} |\psi^I\rangle = H^I |\psi^I\rangle,
\]

where \( H^I \) is the interaction Hamiltonian in the Interaction picture. The interaction Hamiltonian between the electrons and the photons in the Schrödinger picture is,

\[
H_1 = -\frac{e}{2m} \hat{\mathbf{A}} \cdot \hat{\mathbf{p}} + \frac{e^2}{2m} |\mathbf{A}|^2
\]
\( \mathbf{A} \) corresponds to the classical vector potential and can be expressed in terms of emission and absorption operators, \( a^+ \) and \( a \), \(^{36}\)

\[
\mathbf{A} = \sum_{\sigma = 1}^{2} \sum_{n} \frac{n}{2\varepsilon_{\sigma} \omega} \mathbf{\hat{e}}_{n\sigma} (a_n^+ e^{-i\mathbf{k}_n \cdot \mathbf{r}} + a_n e^{i\mathbf{k}_n \cdot \mathbf{r}})
\]

where \( \mathbf{\hat{e}}_{n\sigma} \) is a unit vector in the direction of polarization, and \( \sigma \) determines the two possible polarization states. If \( P_f \) represents the probability for a transition from an initial state \( i \) to a final state \( f \), in the energy range \( dE_f \), then the transition probability per unit time is,

\[
w = \frac{1}{t} \int_P P_f \rho_f dE_f
\]

where \( \rho_f \) is the density of final states.

It can be shown \(^{36}\) that \( P_f \) expressed in terms of the scattering operator \( S \), is,

\[
P_f = \langle \psi_f | S | \psi_i \rangle^2
\]

The scattering operator is defined as,

\[
S = 1 + \frac{1}{i\hbar} \int_0^t H_i^I(\tau) d\tau + \left( \frac{1}{i\hbar} \right)^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 H_i^I(\tau_1) H_i^I(\tau_2)
\]

The first term in the expansion does not give rise to any transitions, since the initial and final states are orthogonal. The second term can give rise to transitions involving both one and two photons. The third and subsequent terms give transitions with
two and more photons involved.

II.2.1. Single photon emission.

In this case the transition probability per unit time is,

\[ W = \frac{1}{\hbar} \int_{t_0}^{t} \left| \langle \Phi_f | H | \Phi_i \rangle \right|^2 \rho f dE_f \]  

II.14.

Changing the Hamiltonian to the Schroedinger picture, and performing the integrations, assuming large \( t \), it can be shown that,

\[ W = \frac{2\pi}{\hbar} \left| \langle \Phi_f | H | \Phi_i \rangle \right|^2 \rho f \]  

II.15.

The density of final states for the whole system is,

\[ \rho f = \frac{2V}{(2\pi)^2} \frac{\omega^2}{\hbar c} 4\pi \]  

II.16.

Of the two terms in the interaction Hamiltonian, equation II.9, only the first term gives to single photon transitions. The second, being quadratic in the vector potential, couples the electron to the radiation field twice, and so gives only two-photon transitions, and we ignore it here.

Thus using II.16, II.9, and II.10,

\[ W = \frac{e^2 m \omega}{\epsilon_0 m c^3} \left| \langle \Phi_f | e^{-i\vec{K} \cdot \vec{r}} \vec{A} \cdot \vec{\nabla} | \Phi_i \rangle \right|^2 \]  

II.17.

The exponential term can be expanded in a Taylor series,

\[ e^{-i\vec{K} \cdot \vec{r}} = 1 - i\vec{K} \cdot \vec{r} + \frac{1}{2}(\vec{K} \cdot \vec{r})^2 - ... \]  

II.18.

When II.18 is substituted into the matrix element II.17 above, the result is analogous to a multipole expansion
in classical radiation theory, the first term being the electric dipole term, the second the electric quadrupole and magnetic dipole term, etc.

II.2.2. The electric dipole approximation.

When only the first term in the expansion of the exponential is included in the matrix element in equation II.17, the transition probability is given in what is known as the electric dipole approximation, as,

\[ \mathbf{w} = \frac{2\pi \hbar}{\varepsilon_0 m c} |\langle \Psi_f | \hat{\mathbf{e}} \cdot \mathbf{r} | \Psi_i \rangle|^2 \]  

II.19.

Converting the matrix element from the dipole velocity to the dipole length form,

\[ \mathbf{w} = \frac{e^2 \omega^3}{\varepsilon_0 \hbar c^2} |\langle \Psi_f | \mathbf{e} \cdot \mathbf{r} | \Psi_i \rangle|^2 \]  

II.20.

Applying II.20 to the case of the \(2S_\frac{1}{2}\) state of atomic hydrogen, which has only two states of lower energy, the \(2P_\frac{1}{2}\) state and the \(1S_\frac{1}{2}\) ground state, it is easily seen that the matrix element vanishes between the two \(S\) states by symmetry, an example of the parity selection rule. The value of the \(2S_\frac{1}{2}\) to \(2P_\frac{1}{2}\) matrix element is, \[\sqrt{\frac{5}{3}} a_0^2\] where \(a_0\) is the Bohr radius. Thus the transition probability for the \(2S_\frac{1}{2}\) to \(2P_\frac{1}{2}\) transition is,

\[ \mathbf{w}(2S\rightarrow2P) = \frac{e^2 \omega^3}{\hbar m c^2} \sqrt{\frac{5}{3}} a_0^2 = 1.8 \times 10^{-12} \text{ s}^{-1} \]

which is a negligible rate.

II.2.3. Higher multipole approximations.

In the electric quadrupole approximation, the \(2S \rightarrow 1S\) and the \(2S \rightarrow 2P\) spontaneous transitions are forbidden by
the "$\Delta l = 0, \pm 2$, except for $l = 0 \rightarrow 0$" selection rule\(^3\). In the magnetic dipole approximation, the $2S \rightarrow 2P$ transition is forbidden by the $\Delta l = 0$ selection rule, and the $2S \rightarrow 1S$ transition is forbidden, in the Pauli approximation, since the radial wave functions are orthogonal in this approximation. However if the exact Dirac wave functions are used, the matrix element of the spin operator is not exactly zero, and so these transitions are not strictly forbidden, but their transition probability is smaller than that for allowed electric dipole transitions by a factor of $10^{-13}$ for hydrogen. The lifetime due to this decay mode has been estimated for hydrogen to be about 2 days\(^6\).

In the higher multipole approximations the $2S \rightarrow 1S$ transition is forbidden by the "No $J = \frac{1}{2} \rightarrow \frac{1}{2}$" selection rule.

II.3.1. Two photon emission.

We now have to consider both the second and third terms in the expression for the scattering operator, equation II.13. For the second term, only the second part of the interaction Hamiltonian, equation II.9, contributes to two photon emission. However, since the wavelength of the radiation being considered, 121.6 nm, is large compared with the dimensions of the region over which the electron wave functions are appreciably different from zero, ($a_o$), the exponential terms can be taken out of the integral expressions for the matrix elements. The orthogonality of the electron
wave functions then causes all but the diagonal elements to vanish. Thus the $|\vec{A}|^2$ term does not contribute to two photon emission in this case.

The probability of finding the atom in the final state $f$ is now,

$$ P_f = \left| \langle \Phi_f | S'| \Phi_i \rangle \right|^2 $$  \hspace{1cm} \text{II.21.} $$

where from equation II.13,

$$ S' = \frac{1}{(\hbar n)^2} \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \ H^I_1(\tau_1) H^I_2(\tau_2) $$  \hspace{1cm} \text{II.22.} $$

Proceeding as in the single photon case,

$$ P_f = 4 \left( \frac{e}{m} \right)^4 \sum_j \frac{\langle \Phi_f | \vec{p} \cdot \vec{A} | \Phi_j \rangle \langle \Phi_j | \vec{p} \cdot \vec{A} | \Phi_f \rangle}{E_1 - E_j} $$  \hspace{1cm} \text{II.23.} $$

where $\sum_j |\Phi_j\rangle \langle \Phi_j|$ is the unit operator, and the $|\Phi_j\rangle$'s form a complete set of basis states.

The initial state of the system is,

$$ |\Phi_i\rangle = |\Phi_i\rangle, \ \text{i.e., no photons}, $$

and the final state is,

$$ |\Phi_f\rangle = |\Psi_f\rangle |\omega_1\rangle |\omega_2\rangle, \ \text{i.e., two photons}. $$

where $\omega_1$ and $\omega_2$ are the angular frequencies of the photons.

The vector potential now is,

$$ \vec{A} = \sqrt{\frac{\hbar}{2eV}} \left[ \frac{\hat{a}_1^+ e^{-iK_1 \cdot \vec{r}}}{\sqrt{\omega_1}} + \frac{\hat{a}_2^+ e^{-iK_2 \cdot \vec{r}}}{\sqrt{\omega_2}} \right] $$  \hspace{1cm} \text{II.24.} $$

Substituting equation II.23 in equation II.23,
The two terms can be illustrated by Feynman diagrams, as shown in Fig. 2.

\[
P_f = \frac{\hbar^2 c^2}{m_e^2 e^2} \frac{1}{\omega_1 \omega_2} \left| \sum_j \frac{\langle \Psi_f | \hat{e}_2 \cdot \vec{p} | \psi_j \rangle \langle \psi_j | \hat{e}_1 \cdot \vec{p} | \Psi_1 \rangle}{E_j - E_j} \right|^2
\]

The W's are the electron energies, the solid lines represent the electron, the dashed lines the photons, and the vertices, the interactions. The decay can proceed by either diagram, so the probabilities are summed.

We have,

\[
E_1 = W_i, \quad E_1' = W_j + \hbar \omega_i,
\]
\[
E_j = W_j + \hbar \omega_i, \quad E_f = W_f + \hbar \omega_i + \hbar \omega_i.
\]

So,

\[
(E_1 - E_1') = W_i - W_j - \hbar \omega_i = \hbar (\omega_{ij} - \omega_i),
\]
\[
(E_1 - E_j') = W_i - W_j - \hbar \omega_i = \hbar (\omega_{ij} - \omega_i).
\]

Where \( \hbar \omega_{ij} \) is the energy difference between states \( W_i \) and \( W_j \).
Using these equations, and changing from the dipole velocity to the dipole length form of the matrix element, II.25 becomes,

\[ P_f = \frac{e^4}{c^2 \hbar^2} \omega_1 \omega_2 \left| \sum_j \frac{\langle \hat{\Psi}_f | \hat{\sigma}_1 \cdot \vec{r} | \hat{\Psi}_j \rangle \langle \hat{\Psi}_j | \hat{\sigma}_2 \cdot \vec{r} | \hat{\Psi}_f \rangle}{\omega_{1j} - \omega_1} \right|^2 + \frac{\langle \hat{\Psi}_f | \hat{\sigma}_2 \cdot \vec{r} | \hat{\Psi}_j \rangle \langle \hat{\Psi}_j | \hat{\sigma}_1 \cdot \vec{r} | \hat{\Psi}_f \rangle}{\omega_{1j} - \omega_2} \right|^2 \]

II.26.

The transition probability per unit time is again given by II.15, where now the squared matrix element is given by \( P_f \) from II.26. Since there are now two photons in the final state, the density of final states can be seen to be,

\[ \rho_f = \rho_1 \rho_2 \delta_\omega_1 \]

II.27.

Thus,

\[ w = \frac{2e^4}{\hbar^2 c \hbar c \hbar^2} \langle |M|^2 \rangle \omega_1^3 \omega_2^3 \, d\omega_1 \]

II.28.

\[ = \mathcal{A}(\omega_1) \, d\omega_1 \]

where \( M \) is the matrix element in II.26, and the average is taken over the directions of propagations of the photons, (through the density of final states function).

The total transition probability, \( \mathcal{A} \) is obtained by integrating II.28,

\[ \mathcal{A} = \frac{1}{\hbar} \int_0^{\omega_{1f}} \mathcal{A}(\omega_1) \, d\omega_1 \]

II.29.

where \( \omega_{1f} \) is the energy difference between the initial and final states. The factor of \( \frac{1}{\hbar} \) is introduced to
avoid counting each pair of photons twice. By conservation of energy, we must have,

$$\omega_1 + \omega_2 = \frac{E_i - E_f}{\hbar} = \omega_{if} \quad \text{II.30.}$$

The intermediate states $|\psi_i\rangle$ in equation II.28, are $P$ states, since the virtual transitions involved are electric dipole transitions. These virtual transitions, which need not conserve energy, can occur to all possible intermediate states, continuum as well as discrete, so the summation in the matrix element must also be considered as an integral over the continuum.

II.3.2. Angular distribution.

The dipole length operator $\mathbf{r}$ in equation II.26, is diagonal in nuclear and electronic spin, so the effects of fine and hyperfine structure can be neglected in applying equation II.28. Then the $S$ levels can be considered single and the $P$ levels triply degenerate, $(m = +1, 0, -1)$. Following Breit and Teller$^6$, we can consider each $P$ level in terms of three linearly independent $P$-state wave functions, having the transformation properties of the cartesian coordinates, $x, y, z$, and denoted by, $\alpha, \beta, \gamma$.

For each state $j$, we have,

$$\sum_{\alpha, \beta, \gamma} (\hat{e}_1 \cdot \mathbf{r})_j (\hat{e}_2 \cdot \mathbf{r})_j \psi \psi_j = (e_{1x} \mathbf{x}^{x}_j (e_{2x} \mathbf{x}^{x})_{\alpha i}
+ (e_{1y} \mathbf{y}^{y}_j (e_{2y} \mathbf{y}^{y})_{\beta i}
+ (e_{1z} \mathbf{z}^{z}_j (e_{2z} \mathbf{z}^{z})_{\gamma i}$$
using the symmetry and orthonormality of the wave functions, and where the matrix elements \( \langle \Psi_f | A | \Psi_j \rangle \) are denoted by \( (A)_{fj} \).

The sum over \( j \) is then,

\[
\sum_{\sigma, \pi, \nu} (\hat{\theta}_1 \cdot \hat{\theta}_2)_{fj} (\hat{\theta}_2 \cdot \hat{\theta}_1)_{ji} = z_f z_j (e_{1x} e_{2x} + e_{1y} e_{2y} + e_{1z} e_{2z})
\]

where \( n \) is the principal quantum number of the \( P \) states, and the matrix elements \( z_{fn} \) are between the state \( f \) and the state of type \( \nu \) with principal quantum number \( n \).

The above analysis shows that the transition probability has a dependence on the polarizations of the photons of the form \( (\hat{\theta}_1 \cdot \hat{\theta}_2)^2 \). When the photon detectors used are not polarization sensitive, as in the present experiment, the transition probability must be averaged over the directions of polarization.

We chose two coordinate systems, \( x, y, z, \) and \( x', y', z' \), with unit vectors along the axes, \( \hat{x}, \hat{y}, \hat{z}, \) and \( \hat{x'}, \hat{y'}, \hat{z'} \), respectively, so that the propagation vector \( \hat{k}_1 \) of photon 1 points along \( \hat{x} \), and propagation vector \( \hat{k}_2 \) of photon 2 points along \( \hat{x'} \), and \( \hat{x} = \hat{x'} \). Then the polarization vectors are,

\[
\hat{e}_1 = \cos \phi \hat{x} + \sin \phi \hat{y}
\]

\[
\hat{e}_2 = \cos \phi' \hat{x'} + \sin \phi' \hat{y'}
\]
where $\phi$ is the angle between $\hat{e}_1$ and the x axis, and $\phi'$ is the angle between $\hat{e}_2$ and the x' axis. Then,

$$\hat{e}_1 \cdot \hat{e}_2 = \cos \phi \cos \phi' + \sin \phi \sin \phi' \cos \theta,$$

II.33.

where $\theta$ is the angle between the propagation directions of the photons, i.e., $\cos \theta = \hat{f} \cdot \hat{f}' = \hat{e} \cdot \hat{e}'$.

Squaring II.33 and averaging over $\phi$ and $\phi'$ independently, we obtain,

$$\langle (\hat{e}_1 \cdot \hat{e}_2)^2 \rangle_{av} = \frac{1}{2}(1 + \cos^2 \theta),$$

II.34.

Therefore if the polarizations are averaged, the photon pair have maximum probability of being emitted with the angle between their directions of propagation being either 0° or 180°.

If the average is taken over the propagation directions as well as over the polarizations, then it is easily seen that,

$$\langle (\hat{e}_1 \cdot \hat{e}_2)^3 \rangle_{av} = \frac{1}{2},$$

II.35.

II.3.3. Spectral distribution.

From the energy conservation condition, equation II.30, it can be seen that the transition probability as given by equation II.28, has a single maximum at,

$$\omega_1 = \omega_2 = \frac{1}{2}\omega_{1f},$$

where $\omega_{1f}$ is the Lyman-alpha frequency, and is symmetrical about $\omega_{1f}$. Fig. 3 shows $A(\omega_1)$ plotted from values tabulated by Spitzer and Greenstein.\(^{17}\)

The transition probability per unit time as a
Fig. 3.

Predicted two-photon spectrum plotted on an energy scale.
function of wavelength is of much more interest in experimental work, and since we have,

\[ A(\omega_1) d\omega_1 = A' (\lambda_1) d\lambda_1 \]

\[ = A(2\pi c) d(2\pi c) = -\frac{2\pi c}{\lambda_1^2} A(2\pi c) d\lambda_1 \]

then,

\[ A'(\lambda_1) \propto \frac{1}{\lambda_1^2} A(\omega_1) . \]

Fig. 4 shows \( A'(\lambda_1) \) derived in this way from Fig. 3.

II.3.4. Natural lifetime.

Incorporating equations II.35, and II.32 into the expression for the transition probability, II.28, we get,

\[ A(\omega) d\omega = \frac{2e^4}{3\epsilon^2 \hbar^2 c^6 n^2} \left[ \sum_{n=0}^{\infty} \int \frac{d^2 f_{n^2}}{\omega_2 - \omega_1} \right] \left[ \frac{1}{\omega_2 \omega_1 - \omega_1} + \frac{1}{\omega_2 - \omega_1} \right]^2 d\omega_1 \]

\[ \times \omega_1^3 d\omega_1 \quad \text{II.36.} \]

Equation II.36 has been integrated by Breit and Teller\(^6\), Spitzer and Greenstein\(^17\), and more recently by Klarsfeld\(^9\), who obtained,

\[ A = 8.2283 \pm 0.0001 \text{ s}^{-1}. \]

In the above derivation of equation II.36, the existence of the Lamb shift was ignored. When the fact that the \( 2P_{\frac{3}{2}} \) level is below the \( 2S_{\frac{1}{2}} \) level is included, it can be seen that one of the denominators approaches zero, when one of the photon frequencies approaches the Lamb shift frequency, and consequently an infinitely large value is obtained for the transition probability.
Fig. 4.
Predicted two-photon spectrum plotted on a wavelength scale.
But when the radiation width of the $2P_{\frac{3}{2}}$ state is taken into account, it can be shown\textsuperscript{38} that this contribution is equal to the probability for the cascade, $2S \rightarrow 2P \rightarrow 1S$. But it has been shown above that the probability for the $2S \rightarrow 2P$ transition is negligibly small, and so the contribution from the cascade can be neglected. Furthermore, the existence of the $\omega_1^3 \omega_2^3$ term in the expression II.36, serves to reduce the transition probability for small differences of the initial and final energy levels, such as that due to the Lamb shift.

It must be pointed out that the above calculations and results are non-relativistic, but the corrections involved should be unimportant for low values of $Z$, especially for hydrogen.

II.4.1. Perturbations.

The three main processes by which the metastable state can be destroyed before undergoing its spontaneous decay are, interactions with electric fields, interactions with magnetic fields, and through collisions. These will be discussed separately below, inasmuch as they affect the present experiment.

II.4.2. Electric fields.

If an electric field is applied to a hydrogen atom in the metastable state, its lifetime is reduced due to
Stark mixing of the 2P states with the metastable $2S_{1/2}$ state. It is to be expected that the $2P_{3/2}$ state gives a much smaller contribution to electric field quenching than the $2P_{1/2}$ state, since it is much further from the $S$ state. Neglecting the $2P_{3/2}$ state, Bethe and Salpeter obtain for the metastable lifetime in an electric field $E$,

$$t_s = t_p \left[ 1 + \frac{\alpha^2}{(1 - \sqrt{1 + \alpha^2})^2} \right]$$ \hspace{1cm} \text{II.37.}

where, \(\alpha = \frac{2.3 \text{e}_a \text{E}}{L}\),

with $t_p$ is the lifetime of the $2P_{1/2}$ state, $E$ is the electric field strength, and $L$ is the Lamb shift. For low field strengths, (i.e., $\alpha \ll 1$, or $E \ll 100 \text{V cm}^{-1}$),

$$t_s = t_p \left[ \frac{475}{E} \right]^2$$ \hspace{1cm} \text{II.38.}

with $E$ expressed in Volts per centimetre.

Using time-dependent perturbation theory, Lamb and Retherford developed a more general expression,

$$\frac{1}{t_s} = \frac{|\langle b | e^b \frac{\gamma}{\hbar^2} | a \rangle|^2}{\hbar^2 (\omega^2 + \gamma^2/4)} \cdot \chi$$ \hspace{1cm} \text{II.39.}

where $\hbar \omega$ is the energy difference between the initial and final states, $|a\rangle$ and $|b\rangle$ respectively, \(\chi = 1/t_p\) is the radiation width of the final state. When $|a\rangle$ represents the $2S_{1/2}$ state, and $|b\rangle$ the $2P_{1/2}$ state, and $\hbar \omega$ the Lamb shift, the expression II.39 reduces to the Bethe formula II.37. Since, as can be seen from Fig. 1, the energy difference between the $2S_{1/2}$ state and the $2P_{1/2}$ state is about 10 times the energy difference between the $2S_{1/2}$ state and the $2P_{3/2}$ state, it follows that the
contribution of the $2P^\frac{3}{2}$ term is about 1% of that of the $2P^\frac{1}{2}$ term, justifying its neglect in the Bethe formula II.37. Drake interpreted the quenching of a metastable atom on suddenly entering an electric field in terms of a two-photon induced transition, with one of the photons at zero frequency, to deduce the polarization of the emitted Lyman-alpha radiation. (The $2P^\frac{3}{2}$ state must be included here to obtain the correct polarization value.)

II.4.3. Magnetic fields.

Under the influence of an external magnetic field, the 2S and 2P levels split into a number of components corresponding to the possible values of the magnetic quantum number $m_j$, i.e., two for the $j = \frac{1}{2}$ state and four for the $j = \frac{3}{2}$ state. Because of the separation of the $2S^\frac{1}{2}$ and the $2P^\frac{3}{2}$ states, we can again neglect the latter. At about 575 gauss there is a crossing between the $2S^\frac{1}{2}$ ($m_j = -\frac{1}{2}$) and the $2P^\frac{1}{2}$ ($m_j = +\frac{1}{2}$) levels. Fields of this magnitude can easily be excluded from an apparatus, but lower fields can still quench the metastable state through the motional electric field $\vec{E}$, for atoms moving in a magnetic field $\vec{B}$, with velocity $\vec{v}$,

$$\vec{E} = \vec{v} \times \vec{B}$$

II.40.

producing Stark mixing.
II.4.4. Collisions.

The quenching of metastable hydrogen atoms through collisions can occur via three main processes,

1. electron loss,
   \[ H(2S) + X \rightarrow H^+ + X + e^- \]

2. electron capture,
   \[ H(2S) + X \rightarrow H^- + X^+ \]

3. de-excitation,
   \[ H(2S) + X \rightarrow H(1S) + X + L_\alpha \]

At thermal energies, the de-excitation process is the only possible reaction, due to the low energy available. Fite et al.\(^{16}\) found that water vapour had a cross-section for quenching metastables as large as \(1000 \text{ \(a_0^2\)}\), while molecular hydrogen and nitrogen gave about \(100 \text{ \(a_0^2\)}\). The high value for water vapour can be understood in terms of the long-range permanent dipolar field of the molecule, while the other molecules have only quadrupolar fields.

As the energy of the projectiles is increased, the de-excitation process becomes less important due to the shorter time spent in the target field.

Electron capture cross-sections are also found to decrease with increasing energy\(^{40}\), but more slowly than the de-excitation process. Electron loss cross-sections have been observed to increase with increasing energy\(^{40a}\) and are relatively unimportant at the moderate energies of interest here, (about 1 keV, or 0.2 a.u.).

The only target species we need consider are the main constituents of the background gas, which were observed
to be, (using a quadrupole mass spectrometer), water vapour, molecular hydrogen and molecular nitrogen. The total quenching cross-section for H$_2$O and N$_2$ have been measured by Dose et al.$^{41}$ in the energy range of 2 $\rightarrow$ 60 keV. Their results at 2 keV give about 25 $\sigma_0^2$ for both species, increasing with decreasing energy. Extrapolated to 1 keV, the probable upper limit is about 40 $\sigma_0^2$. Byron et al.$^{42}$ have measured the total quenching cross-section for molecular hydrogen and nitrogen in the energy range of interest, and found a value of about 8$\sigma_0^2$ for both, at an energy of 1 keV. Although the two results are not consistent, we can take about 40 $\sigma_0^2$ as an upper limit for quenching of the metastables by the background gas in the region of 1 keV.

The effects of the perturbations are two-fold; (1), they reduce the metastable density, and (2), if they occur within the observation region, can give rise to a radiation background.

Electric and magnetic fields only contribute to (1), since these processes give rise to Lyman-alpha photons, which the photomultipliers are not sensitive to. Since the time taken for the metastable atoms, at 1 keV energy, to traverse the apparatus is about 2 $10^{-6}$ s, perturbations must be kept low enough so that the reduced lifetime is, say, at least 10 times this value. This requirement gives, from equation II.38, that the average electric field $E$ along the beam length must be,

$$E \lesssim 4.5 \text{ V cm}^{-1}$$

and the magnetic field, perpendicular to the beam axis, must be,
Collisions with the background gas can give rise to processes (1) and (2). If we take the average background pressure in the metastable beam path as about $10^{-7}$ torr, or a thickness over 1 metre of, $\pi = 3.5 \times 10^{15}$ molecules m$^{-2}$, then the fractional loss of metastables over this length can be approximated as,

$$f \approx \pi \sigma$$

where $\sigma$ is the cross-section for metastable destruction, about $40 \text{ma}^2$ or $4 \times 10^{-19}$ m$^{-2}$, from above. This gives,

$$f \approx 1.4 \times 10^{-3}$$

a negligible loss.

For process (2), we can assume, as a worst case, that each collision gives rise to a detectable photon, through excitation of the background gas, then the radiation background produced by collisional quenching can be approximated as,

$$R \approx j(2S)\pi \sigma$$

where $j(2S)$ is the metastable flux, in atoms per second, and $\pi$ is now the thickness of the background gas in the observation region. For a pressure of $10^{-8}$ torr, and an observation region length of about $3 \times 10^{-2}$ m, $\pi = 10^{13}$ m$^{-2}$. We then have,

$$R \approx 2.5 \times 10^4 \text{ s}^{-1}$$

the significance of which will be discussed in section III.13.
III. Apparatus.

III.1. Introduction.

A schematic diagram of the main components of the apparatus is shown in Fig. 5. A duoplasmatron ion source, (1), with electrostatic lens system, (2), and analysing magnet, (3), are used to produce a proton beam. The protons are converted into hydrogen atoms in a cesium charge-exchange cell, (7). The detection region consists of an ultra-violet transmitting quartz tube, (11), which is observed by a pair of photomultiplier tubes, (10).

After leaving the detection region, the beam is monitored by a photomultiplier, (14), which detects the Lyman-alpha radiation emitted by the metastable atoms on entering the quench field, (13), and by a neutral detector/faraday cup, (15), which measures the current of the protons or of the neutrals in the beam, using a reversible potential on the guard ring.

The fast photomultiplier tubes are connected to a delayed coincidence circuit, which monitors the distribution of arrival time differences of pulses from the photomultiplier anodes. This method is described by Belcher. After leaving the charge-exchange cell, but before entering the detection region, the beam passes through two pre-quenching systems, (8), and (9). The pre-quench plates, (8), fulfil two functions, the removal of the charged fraction from the beam, and the periodic pre-quenching of the metastables, in order that
the background can be monitored. The pre-quench wires, (9), are used to fully quench the metastables to provide a zero check.

III.2. Production of the metastable beam.

The traditional source of metastable hydrogen atoms has until recently been of the Lamb-Retherford type, consisting of an oven to dissociate molecular hydrogen, and an electron beam to excite the atoms to the metastable state. With a source of this type, the beam intensity and collimation obtainable have been limited by the cross-section for the excitation, $0.16 \text{ mb}^2$, the electron beam intensity obtainable and the large gas loads that must be handled by the vacuum system.

Another method which has been used to form a metastable beam is by charge transfer of protons in a gas target. The maximum cross-sections for this process are small, however and occur at relatively high energies, for example, for molecular hydrogen, about $0.27 \text{ mb}^2$, at 30 keV, and for argon, about $0.28 \text{ mb}^2$ at the same energy.

In 1964, Donnally et al. proposed an improved source of low-energy metastable hydrogen atoms using the nearly resonant process of charge-exchange of protons in cesium vapour,

$$\text{H}^+ + \text{Cs} \rightarrow (\text{H}(2S), \text{H}(2P)) + \text{Cs}^+ - 0.49 \text{ eV}. \quad \text{III.1.}$$

Massey has pointed out that when the energy defect
Fig. 5.

Schematic diagram of the apparatus. (For key, see page 36.)
Key to Fig. 5, 'schematic diagram of the apparatus'.

(1) duoplasmatron,
(2) einzel lens, (lens chamber),
(3) analysing magnet,
(4) alignment bellows,
(5) retractable faraday cup, (chamber DP I),
(6) gate valves,
(7) charge-exchange cell,
(8) pre-quench plates, (chamber DP II),
(9) pre-quench wires, (chamber UHV I),
(10) coincidence photomultipliers,
(11) quartz tube with bellows,
(12) turntable,
(13) quench plates, (chamber UHV II),
(14) metastable monitor with oxygen filter,
(15) neutral detector/faraday cup,
(16) collimating apertures.
for the charge changing process is small, the cross-section is expected to be large and occur at low energies. This has since been verified in a number of experiments. Pradel et al. have found that the maximum cross-section for the above process is about $60 \pi a_0^2$, at an energy of about 600 eV, and at a cesium target thickness of about $1.2 \times 10^{14}$ atoms cm$^{-2}$.

Since the rate of two-photon decays of a metastable beam at a fixed detector location is proportional to the metastable density, a large cross-section at a low energy is required for the maximum rate, and so the near resonant charge-exchange process is used here.

III.3. Vacuum system.

The vacuum system consists of five main stainless steel vacuum chambers, individually pumped, Fig. 5. These are designated, in order from the ion source, as the lens chamber, the differential pumping chamber I, (DP I), the differential pumping chamber II, (DP II), the ultra high vacuum chamber I, (UHV I), and the ultra high vacuum chamber II, (UHV II). All of the flanges on the UHV chambers are sealed with aluminium gaskets. Most of the flanges on the rest of the system are sealed with Viton 'O' rings.

The lens chamber is pumped with a six-inch type EO6 oil diffusion pump, with a water-cooled chevron baffle to prevent backstreaming of the pump oil, and is isolated from the chamber by a quarter-swing butterfly valve.
The 600 l s\(^{-1}\) diffusion pump is backed by a type ES 330 334 l min\(^{-1}\), oil sealed rotary pump. All the diffusion pumps in the system are filled with polyphenyl ether pumping fluid, (Santovac 5), with a vapour pressure in the 10\(^{-10}\) torr region, and is highly resistant to cracking. Cleaver et al.\(^{49}\) found that under intense electron bombardment, this fluid gave rise to conducting films, and no evidence for the build up of insulating layers on the ion source or the lens system was found here.

Chamber DP I is evacuated by a two-inch EO-2 150 l s\(^{-1}\) oil diffusion pump and an ES50 50 l min\(^{-1}\) oil sealed backing pump, with thermoelectrically cooled chevron baffle and butterfly valve. Typical operating pressures in this chamber are 1 \(10^{-7}\) torr when the ion source is not running, and 1 \(10^{-6}\) torr with the source on.

The pumping stack attached to chamber DP II is similar to that on DP I, with the addition of a liquid nitrogen cold trap, as the pressure in this chamber was observed to be dominated by water vapour. Typical operating pressure in this chamber is 5 \(10^{-7}\) torr, the ion beam having little effect.

Between chambers DP I and UHV II, there is a pumping aperture, 20 mm long and 10 mm diameter, which also serves to collimate the beam. Chamber UHV I is pumped by a four-inch EO4 600 l s\(^{-1}\) oil filled diffusion pump, and an ES200 190 l min\(^{-1}\) oil sealed rotary backing pump, with thermoelectrically cooled chevron baffle and butterfly valve.
Chamber UHV II pumping stack consists of an E06/ES330 combination, as for the lens chamber, with thermoelectric baffle, liquid nitrogen cold trap, and butterfly valve. Both chambers UHV I and UHV II, being all metal gasket sealed, could be baked to 400°C by means of convection heaters totalling 7 kW distributed beneath them. A double-walled aluminium baking jacket, with the cavity filled with ceramic fibre, which could be fitted completely over these two chambers with a slot at one end to allow the connection to DP II to pass through. After 24 hours baking at temperatures between 350° and 400°C, and with all the cold traps filled, the pressure in UHV I eventually reached 4.5 \times 10^{-9} \text{ torr}, and in UHV II, 1 \times 10^{-9} \text{ torr}. The presence of an ion beam had no detectable effect on the pressure in UHV I, and caused the pressure in UHV II to rise to about 2 \times 10^{-9} \text{ torr}.

In all the chambers, the pressure is monitored by Bayard-Alpert type ionization gauges with X-ray limits of 2 \times 10^{-10} \text{ torr}, and the vacuum composition could be monitored with a quadrupole mass spectrometer in UHV II.

III.4. Ion source.

The ion source is a Von Ardenne type duoplasmatron, manufactured by Edwards High Vacuum, and designed to produce high brightness, low emittance proton beams at energies less than 5 keV, Fig. 6.

Electrons are produced by a tantalum filament heated by a 15 V, 60 A ac supply, and an arc is struck between
Fig. 6.

Ion source and power supplies.
the filament and anode. An intermediate electrode constructed of magnetic stainless steel constricts the arc, which is focused to a small area by a longitudinal magnetic field, thus producing a high degree of ionization in a small region at the anode. A small orifice (0.15 mm diameter) in the anode permits the plasma so produced to leak into the high vacuum region where the ion flux is extracted and shaped into a beam.

The arc is powered from a 300 V 5 A stabilised supply, with a 100 ohm variable series resistor to protect the supply from current surges. A 2000 ohm variable resistor is used for starting the arc by applying a high voltage between the intermediate electrode and the filament. In normal operation, this resistor is reduced to a low value, keeping about 30 V on the intermediate electrode for stable operation. (A higher voltage increases the velocity of the ions striking the filament, and as a result, its lifetime is greatly reduced.)

Hydrogen gas from a pressurised bottle is fed through a water vapour trap filled with silica gel, and a fine-control needle valve to the duoplasmatron. A temperature compensated Pirani gauge is used to monitor the hydrogen pressure in the source, which is typically about 1 torr.

With an arc current of about 1 A, and a filament current of 50 A, the source ran with reasonable stability for periods normally exceeding 100 hours, (limited by the filament lifetime).
III.5. Lens system and analysing magnet.

A double electrostatic lens system is used to extract, accelerate and focus the ion beam. The first lens is an accelerate-decelerate gap lens, Fig. 6, which controls the primary beam divergence. The second lens is a three-electrode decelerate-accelerate Einzel type, which controls the focus of the ion beam on target, without changing the beam energy. A voltage of -90 V is permanently applied to the outer elements of the Einzel lens, which then act as electron repellers forcing electrons back into the beam to maintain space-charge neutralization.

A standard C-type electromagnet is used to separate the different mass components of the beam. The pole pieces are shaped to allow stigmatic focusing, so that the circular cross-section of the beam, and thus the maximum current density, can be maintained at the target.

The duoplasmatron produces the ion species, $H_1^+$, $H_2^+$, and $H_3^+$ in the approximate percentages of the total beam of, 20%, 40%, and 40%, respectively. The proton beam is deflected through an angle of 30°, and a stainless steel bellows attached to chamber DP I assists in beam alignment. Fig. 7 indicates how the proton current output varies with beam energy, as measured by the faraday cup in chamber UHV II.
Proton output current versus beam energy. Error bars show error estimated from repeated measurements.
III.6.1 Charge-exchange cell.

The cesium charge-exchange cell is based on a design of Bacal et al.\textsuperscript{50}. This is a closed-cycle cell based on the open heat pipe principle, Fig. 8. The advantages of this cell lie in, reducing to a low value the flux of cesium atoms through the cell ports, through a reasonable choice of cell geometry, and continuously returning to the cell evaporator the cesium metal which condenses on the cell walls.

The central region of the cell, (the evaporator), is heated to a temperature at which the desired cesium vapour pressure is obtained, and a temperature close to, but a few degrees higher than, the melting point is imposed on the extremities of the cell. The internal surface of the cell is lined with several layers of fine stainless steel wire mesh (wick) tightly fitting against the walls. The liquid cesium that condenses on this is spread out by capillary action and does not get a chance to 'ball up' and block the beam. The liquid metal builds up a density gradient along the cell, and flows from the extremities of the cell back to the central evaporator. This continuous recycling of the cesium in the cell, consisting of successive evaporations and condensations also leads to a continuous purification of the metal. The cesium loss rate through the ports for this type of cell is reported to be\textsuperscript{50} $6.1 \times 10^{-4}$ g hour\textsuperscript{-1}, at an evaporator temperature of 110\textdegree C and a condenser temperature of 32\textdegree C. Loss rates approaching this value have been observed here.
Fig. 8.
Cesium charge-exchange cell.
The cell can be isolated from the vacuum system by means of gate valves at either end, and has three auxiliary ports, one supporting a Langmuir-Taylor hot-wire ionization detector, and another supporting a needle valve for flushing the cell with argon when charging with cesium.


Fig. 9 and Fig. 10 show how the neutral detector current and the metastable monitor count rate vary with the thickness of the cesium target respectively. Both curves show a peak. The neutral detector current is proportional to total number of neutrals produced in the interaction III.1, including the metastables. The reason for the decrease, in this case, is probably scattering out of the beam, and to a lesser extent, electron loss in multiple collisions, at high target thicknesses.

The peak in the metastable monitor curve is much more pronounced. The more rapid decrease in this case, is due to the fact that the primary destruction mechanism for the metastable state in cesium is that of de-excitation to the ground state, as shown by Pradel et al.36. They measured a cross-section for this mechanism of $50 \, \mu a_0^2$. At high target thicknesses, where multiple collisions occur, almost all the metastables produced are also de-excited. The shapes of these curves, Fig 9 and Fig. 10, agree quite well with the observations of Pradel et al.36, and
Fig. 9.

Total neutrals produced versus cesium target thickness. Error bars show error estimated from fluctuations due to beam instabilities.
Fig. 10.
Metastable flux versus cesium target thickness.
Error bars show one standard deviation.
Spiess et al. The position of the maximum in the metastable curve, Fig. 10 is about a factor of 2.5 higher than that observed by Pradel and Spiess, whose results are consistent with each other. The target thicknesses shown here have been calculated from the temperature of the cesium evaporator block, using a calibrated thermocouple at the position shown in Fig. 8, and the formula of Langmuir and Taylor,

\[
\log p = 11.0531 - 1.35 \log T - 4041 T^{-1}
\]

where \( p \) is the pressure in torr, and \( T \) is the absolute temperature. The thickness, \( \pi \), is then given by,

\[
\pi = 9.64 \times 10^{18} \frac{p l}{\tau}
\]

where \( l \) is the effective length of the target through which the beam passes in cm, and \( \pi \) is in atoms cm\(^{-2}\).

Although it was ensured that the system was in equilibrium, it is possible that the vapour pressure calculated was too high, due to impurities in the cesium, and so account for the discrepancy.

A different design of charge-exchange cell with low-conductance entrance and exit ports was tried at first, and this design was also suitable for use with gas targets. Fig. 11 shows how the number of metastables produced per second varies with different thicknesses of argon in the cell. Argon, at low energies, has a relatively large cross-section for metastable production, about 0.1 ma\(^2\), compared with that of molecular hydrogen, which has about 0.01 ma\(^2\). The target thickness here...
Fig. 11.
Metastable flux (relative) versus argon target thickness.
Error bars show statistical error plus error due to instabilities.
was monitored using an ion gauge in an adjacent vacuum chamber, and assuming a linear relationship between this and the argon pressure in the charge-exchange cell. However a different geometry was used for metastable detection from that used for Fig. 10, so a direct comparison is not possible. Nevertheless the difference in the shapes of the two curves, Fig. 10 and Fig. 11 is of interest. At high target thicknesses in argon, the metastable flux reaches a limiting value of about 25% of the peak height, while for cesium, this limiting value is very small. It is apparent that this ratio is very sensitive to the ratio of the cross-sections for formation of metastables from protons, and from ground state atoms. The excitation cross-section from the ground state can be expected to be of the same order of magnitude for both argon and cesium, while the charge transfer cross-section is nearly resonant in cesium and non-resonant in argon.

The metastable beam density as a function of beam energy is shown in Fig. 12, (closed circles). This is obtained by measuring the output rate of the metastable monitor, and dividing by the beam velocity. The error bars show the statistical uncertainty, plus an uncertainty due to beam instabilities, estimated from the scatter in repeated measurements. The shape of the curve depends on the shape of the cross-section curve, and the ion source proton output versus energy curve, Fig. 7. The metastable density displays a maximum at about 1.1 keV.

The neutral detector output current is also shown in Fig. 7, the error bars showing the estimated error due to
Fig. 12.
Metastable beam density (closed circles) and total neutral flux (open circles) versus beam energy, error bars show statistical error plus error due to instabilities.
the scatter in repeated measurements. The main contribution to the random background in the coincidence time spectrum, when the charged component has been removed, is from the neutral current through the quartz tube, which is measured by the neutral detector. Fig. 12 can be used to determine the energy for the maximum signal to noise ratio for detection of the two-photon decay, the position of which is not immediately obvious. This will be discussed in a later section of this chapter.

III.7. Quenching arrangement.

It was observed that of the three main components of the beam after leaving the charge-exchange cell, protons, metastables and ground state neutrals, the protons, as would be expected, gave rise to by far the largest singles background. Apart from the deterioration in the signal to noise ratio, the effect of the charged component colliding with the background gas almost drove the electronics into saturation, which effect gave rise to a series of oscillations in the time spectrum.

If the beam is passed through a pair of parallel plates with an electric field $E$ between them, and if the time spent by a particle in the field is $t$, then the deflection of the ions in the beam is proportional to $Et^2$. The number of metastables quenched, for relatively low values of $E$, can be seen from equation II.38 to be proportional to $E^2t$. It can be seen that if $t$ is made large enough and $E$ small enough, it is possible to remove all the ions from the beam without quenching.
an appreciable number of metastables. These conditions are satisfied by a long pair of plates, with a low electric field applied. In the present experiment, the length of the charge removal plates is limited by the size of the vacuum tanks. A pair of plates of length 60 mm, and a spacing of about 40 mm were employed, which also serve as pre-quench plates for the metastable component.

With a pure proton beam passing through the apparatus, it was found that 99.95% approximately of the beam was restricted from entering the quartz tube at an applied voltage of $\pm 45$ V. Fig. 13 shows how the metastable monitor count rate varies with the voltage applied to the pre-quench plates. It can be seen that at $\pm 45$ V, about 27% of the metastables are quenched. Although these figures could be greatly improved upon, they were found acceptable.

In Fig. 13, the points are the measured values, the error bars show one standard deviation, and the solid line is the calculated curve for the remaining fraction of metastables after passing through the pre-quench plates with a voltage $V$ applied. It is easily shown from equation II.38, that the remaining fraction is,

$$f = e^{-kV^2}$$  \hspace{1cm} \text{III.4.}$$

where $k$ is a constant which depends on the beam velocity and the plate geometry. The value of $k$ used here is chosen for the best fit to the measured points, and is,

$$k = 1.374 \times 10^{-4} \, \text{v}^{-2}$$
Fig. 13.

Lyman-alpha count rate versus pre-quench plate voltage.

Points are the measured values, the solid curve is fitted using equation III.4.

Error bars show one standard deviation.
The fact that the fit is so good can be taken as evidence that it is indeed the metastable state which is under observation.

The voltage on the pre-quench plates is switched alternately between \( \pm 45 \text{ V} \) and \( \pm 250 \text{ V} \), shown by the dashed lines in Fig. 13 to separate the radiation due to the metastables from that due to the other components of the beam, as well as that due to external influences.

The pre-quenching wires, shown at (9) in Fig. 5, consists of two rows of eight wires each, 8 mm between each wire and 20 mm between the rows. This system is based on a design of Tuan et al. and is designed so as not to deflect any residual charged component in the beam, while fully quenching the metastable component. A voltage of \( \pm 1000 \text{ V} \) is applied, which alternates in sign every two wires. Under all conditions tried with this system, large deflections of the charged component were produced, however they were still used in a null test on the two-photon signal, the charged component giving a very small effect.

The metastable monitor views a system of four plates, similar to a design of Spiess et al. The quenching field is applied to the two inner plates, which are 65 mm long and are spaced 40 mm apart. The fringe fields are reduced by a set of image plates placed 3 mm outside the inner plates. Spiess et al. have shown that the fringe field in front of the plates is very small when the outer plates are 4 mm longer than the inner ones, and so it will not appreciably influence the metastable beam in the
detection region. Fig 14 is a plot of the Lyman-alpha photomultiplier output against the voltage applied to the quench plates, with a metastable beam passing through. The normal operating voltage is ±500 V.


The neutral detector consists of a Faraday cup, with guard ring and a 90% transparency tungsten wire mesh over the cup entrance. With the guard ring and mesh biased negatively, it operates as an electron-suppressed Faraday cup, to measure the charged component of the beam, and with the charged component of the beam deflected by the quench field, and a positive potential on the guard ring and mesh, it measures the neutral component of the beam. The secondary electrons emitted by the neutrals on striking the cup surface, are accelerated out of the cup and give rise to a positive current flowing from the detector, which can be used to measure the neutral current, provided the secondary electron emission coefficient is known. Pradel et al.\textsuperscript{35} found the same value for the coefficient for both metastables and ground state neutrals, which is, 1.7 at a beam energy of 1 keV.

Fig. 15 shows how the detector output current varies with the potential on the guard ring, (actually the guard ring was grounded and a negative potential applied to the cup). It can be seen to approach an equilibrium value only after about 40 V is applied. This implies that the secondary electron energy spread extends to this value. The normal operating guard ring voltage is 45 V.
Fig. 14.

Lyman-alpha count rate versus quench plate voltage.

Error bars show one standard deviation.
Fig. 15.
Neutral detector output current versus guard ring voltage, for a constant beam. Error bars are estimated from scatter in repeated measurements.

A rough estimate of the neutral beam composition will be made at the cesium target thickness which gives the maximum number of metastables. It must be emphasised that this can only be a very crude estimate for some of the quantities required are not known very accurately. The main purpose of this exercise is to demonstrate the consistency of the measured parameters.

The flux of metastable atoms observed by the metastable monitor can be expressed as,

\[ F_m = \frac{n}{\Omega \eta T \alpha P} \ m \ s^{-1} \]

where \( n \) is the count rate of the metastable monitor, \( \Omega \) is the solid angle subtended by the monitor at the beam on entering the quench field, \( \gamma \) is the quantum efficiency of the photomultiplier at the Lyman-alpha wavelength, \( T \) is the transmission of the lithium flouride window, \( \alpha \) is the absorption of the oxygen at Lyman-alpha, and \( P \) is the correction for the polarization.

The metastable monitor count rate, from Fig. 10, at the maximum, is about \( 6.6 \times 10^3 \ m \ s^{-1} \). The solid angle, \( \Omega \), is estimated to be \( 6.5 \times 10^{-5} \) of a sphere. The photomultiplier quantum efficiency is taken from the manufacturers data to be \( 10\% \) at 121.6 nm. The lithium flouride window transmission, \( T \), is also taken from the manufacturers data to be \( 50\% \) at the same wavelength. The absorption coefficient of dry molecular oxygen at atmospheric pressure at the Lyman-alpha wavelength is
given by Carriere and de Heer\textsuperscript{57} as 0.7 cm\textsuperscript{-1}. Since the length a photon has to travel through the oxygen is about 4 cm, its transmission is about 6%.

It is well known that the radiation emitted by metastable atoms in an electric field is anisotropic\textsuperscript{58}. The measurements of Wijngaarden et al.\textsuperscript{58}, extrapolated to a field strength of 250 V cm\textsuperscript{-1}, the value of the quench field used here, give the anisotropy, $R$ as about 0.168. The anisotropy is defined as,

$$R = \frac{I_n - I_\perp}{I_n + I_\perp} \quad \text{III.6.}$$

where $I_n$ and $I_\perp$ are the intensities measured in the directions parallel to, and perpendicular to, the field lines respectively. The intensity measured here is $I_\perp$, and so we have,

$$\frac{I_\perp}{I_n + I_\perp} = \frac{1 - R}{2} = 0.416$$

i.e., the value measured here is too small by a factor of 0.416.

Combining these results, we get the metastable flux as,

$$F_m = 8.1 \times 10^{10} \text{ H(2S) s}^{-1}.$$  

The flux of neutral atoms is given by,

$$F_g = \frac{i_{nd}}{e \gamma} \quad \text{III.7.}$$

where $i_{nd}$ is the current from the neutral detector, $e$ is the electronic charge, and $\gamma$ is the secondary electron emission coefficient. Pradel et al.\textsuperscript{35} have measured $\gamma$ for 'dirty' stainless steel as about 1.7 for a neutral
beam energy of 1 keV. At the cesium thickness at which
the metastable flux is at a maximum, (about \(3 \times 10^{14}\) atoms \(\text{cm}^{-2}\))
the neutral detector current is about \(3.1 \times 10^{-7}\) A, from
Fig. 9. These values give the total neutral flux as,

\[ F_g = 1.1 \times 10^{12} \text{ atoms s}^{-1}. \]

which gives,

\[ \frac{F_m}{F_g} = 0.074 \text{ or } 7.4\%. \]

The maximum percentage of metastables in the neutral
beam measured by Pradel et al.\(^{35}\) was about 30\%, with a
similar experimental arrangement. Bearing in mind the
large uncertainties in most of the parameters used above,
the agreement is not bad.

**III.10.1. Detection arrangement.**

Between chambers UHV I and II is mounted a vacuum-tight
tube of synthetic quartz, (Spectrosil). Around this
tube is fixed a light-tight and reasonably air-tight
aluminium box. Fixed to one side of the box is a
turntable assembly, consisting of a rotor inside a stator
through which the quartz tube passes. One detector
(photomultiplier tube) can be mounted on the rotor, and
another on the stator, so that the angular distribution
of the coincidences can be investigated. The whole
turntable and its associated drive assembly is designed
to fit inside chamber UHV II.

Synthetic quartz has a short-wavelength cut-off of
about 160 nm, which allows about 56\% of the two-photon
spectrum to be observed. Since molecular oxygen begins
absorbing light at about 180 nm, it is desirable to remove it from the detection region. This can be achieved by flushing the detector box with argon.

III.10.2. Photomultiplier tubes.

The main selection criteria for selecting the coincidence photomultiplier tubes were the spectral response, and the timing accuracy. The spectral response was chosen to peak at 240 nm, - half the Lyman-alpha frequency, for both tubes.

The coincidence photomultiplier tubes used in producing the present results were an E.M.I. type 9816QB and a Phillips type XP1023. The 9816QB has a spectral response extending from 160 nm to 750 nm, (S20), a peak quantum efficiency of 28%, an anode pulse risetime of 2.3 ns, and a transit time difference of 0.9 ns for electrons leaving the centre and a point 18 mm from the centre, of the photocathode. The photocathode diameter is 50 mm.

The XP1023 has an S13 type spectral response, extending from 160 nm to 620 nm, a peak quantum efficiency of 25%, an anode pulse risetime of 1.8 ns, a transit time difference of 0.2 ns, for the same conditions as the 9816QB, and a photocathode diameter of 42 mm.

Both coincidence photomultiplier tubes were housed in 3 mm thick brass tubes to reduce spurious coincidences.

An EMR type 542G-08-18 photomultiplier tube was used as a Lyman-alpha monitor. This tube is solar blind with a lithium flouride window, and a spectral response
extending from 105 nm to 220 nm, peaking at the Lyman-alpha wavelength of 121.6 nm. A copper gasket sealed lithium fluoride window was mounted on chamber UHV II, and a flowing dried molecular oxygen filter was used between it and the photomultiplier as a Lyman-alpha filter. It is well known that molecular oxygen has a transmission window at this wavelength\textsuperscript{57}.

III.11.1. Electronics.

The electronic system is designed to have the best practical timing resolution, so matched 50 ohm cables are used throughout, and great care was taken to match the photomultiplier anodes to the cable. Although dc-coupled amplifiers and discriminators are used, it was felt desirable to operate the photocathodes at ground potential to avoid stray electric fields in the detection region. In consequence, the photomultiplier anodes had to be ac-coupled, however no evidence for pulse pile-up on the blocking capacitors was observed.

Initial operation of the detection system gave rise to a number of spurious coincidence peaks, but most of these were tracked down to either cross-talk between the coincidence channels, or electrical pick-up. After these effects had been eliminated, however, a small spurious coincidence peak still remained, which, after a number of tests were made, was ascribed to cosmic rays. Similar effects have been observed by Novick\textsuperscript{59}, and King et al.\textsuperscript{60}.
The solution adopted to eliminate this effect was to modulate the metastable component of the beam, using a dc electric quenching field, and synchronously switching the detection system. Although this completely eliminated the spurious effect, it also doubled the measurement period.

A schematic diagram of the electronic system is shown in Fig. 16. The anode pulses from the coincidence photomultipliers are amplified by a factor of 10 in fast amplifiers, type NE4634, which have a risetime of about 2 ns. The output pulses are fed to constant fraction of pulse height timing discriminators, ORTEC type 463. One of the discriminator outputs is taken directly to the start input of an ORTEC type 447 time-to-pulse-height converter (TAC), while the other is taken through a variable delay unit, to the stop input. The TAC produces an output pulse whose amplitude is proportional to the arrival time difference of the start and stop pulses. The TAC linearity is optimum in the centre of its timing range, so the variable delay is adjusted to position the true coincidence peak in this region. The TAC output is gated off by a switch unit when the voltage on the proton removal (pre-quench) plates is being changed, to avoid the possibility of spurious coincidences being generated.

The TAC output is taken directly to the input of a multi-channel pulse height analyser (MCPHA), an Intertechnique DIDAC 800. The subgroup of the MCPHA memory that is being addressed by the TAC output is also determined by
Fig. 16.

Schematic diagram of electronic system.
the switch unit, so that the time spectra for the metastable beam on and quenched are stored separately. The timing discriminator outputs are also coupled to fast scalers, which monitor the integrated singles counts.

The metastable monitor, which counts the Lyman-alpha photons emitted by the metastable beam on entering the quench field, is coupled to a slow shaping amplifier, type NE4603, and integral discriminator type NE4623. The discriminator output is fed to a ratemeter, and two scalers, which are gated alternately on and off by the switch unit. One of these scalers monitors the metastable beam, and the other the background. On subtracting the scaler readings, a measure of the integrated metastable current is obtained. A small error is introduced by the second scaler counting the photons emitted by the quenched metastable atoms, but this effect is small and can be estimated and allowed for. The ratemeter is taken to a two-pen chart recorder, the other channel of which monitors one of the singles rates via another ratemeter. The neutral detector output is monitored by an electrometer and another chart recorder.

III.11.2. Switch unit.

The timing of the switch unit is controlled by a 100 kHz, 0.1% accuracy crystal controlled clock. The switch unit fulfills six separate functions as follows, (1), it switches the pre-quench/proton_A field high and low, (2), it routes the metastable monitor output to one of two scalers,
depending on whether the pre-quench field is high or low, (3), it routes the TAC output to the appropriate subgroup of the multi-channel analyser memory, (4), it continuously monitors the timing of the high/low periods, (5), it gates the TAC off during a pre-quench field switching operation, for a precise amount of time, (0.655s), and (6), it switches the read/store function of the MCPHA.

The clock unit divider is normally set to produce one output pulse per minute, and the arrival of one of these pulses at the switch unit initiates a change in level of the pre-quench field, provided the start button has been pressed. If the stop button is pressed during a run, the unit only changes to a stop condition when a switching cycle is complete, thus preserving equal on-off times irrespective of when the run was started or stopped.

Fig. 17 shows a schematic of the switch unit. The start and stop buttons are followed by an 'anti-bounce' circuit, (R-S latch), whose output goes to logic '1' when the start button is pressed, and to a logic '0', when the stop button is pressed. This output is fed to the 'data' input of a D-type latch, whose output follows the data input when the clock (CK) input is high, and stores the data when the clock input goes low. The clock input is derived from the leading edge of the gating waveform, which in turn is derived from a J-K flip-flop, so that the latch outputs (Q and \( \overline{Q} \)) change state, when demanded by the start and stop buttons, at the beginning or end of a complete switching cycle. The
Fig. 17.
Schematic diagram of switch unit.
rest of the circuitry is then controlled by the latch outputs.

The two scalers which count the Lyman-alpha photons are gated on and off by pulses derived from the leading edges of the gating waveform and its complement, via 'One-shot' multivibrators, (O.S.). When the stop button is pressed, the scaler 'gate open' pulses are inhibited and at the end of a switching cycle, both scalers are gated off by a pulse derived from the latch output Q, fed through an OR gate together with the normal gating pulses.

The latch output also gates the switching relay, which is driven when the gate is open by the JK flip-flop, through a driving circuit. The relay switches the pre-quench plate voltage with a 0.2 us time constant to avoid 'spikes', the memory subgroups of the MCPHA, and also a 10 Hz frequency derived from the 100 kHz clock, to the timing scalers. These serve to check whether the relay contacts stick and give a spurious signal.

Since all these functions are performed by the same relay, they must be synchronous to a high degree. The switching period of one minute on and one minute off, is a compromise between the long periods required to reduce inaccuracies in the switching process, and the short periods required to average out drifts and instabilities.
III.12. Light pulser.

To locate the zero position in the time spectrum, all the relative delays associated with the two coincidence channels must be known, or measured. A fast pulse generator is of limited use, since no account is taken of the different transit times in the photomultipliers. A pulsed light source, if fast enough, measures all the relative delays, as well as the time resolution. An ordinary visible light emitting diode (LED) is used here, driven at a high current by a fast pulse. The pulse generator used is unusual, and is discussed below.

If the collector-base junction of a transistor is reverse biased with a sufficiently high voltage, the minority carriers are accelerated across the depletion region, causing ionization to occur by liberating bound electrons, and setting up a micro-plasma in the base region. This effect is called avalanche breakdown, and gives rise to a negative resistance characteristic, which can be utilised to produce fast risetime pulses.

The circuit used here is shown in Fig. 18.

![Fig. 18.](image-url)
A positive pulse, derived from a differentiated multivibrator output, of amplitude about 2 V is applied directly to the base. The transistor turns on rapidly, and avalanche breakdown is initiated. The collector voltage drops very rapidly, (the large value of the collector resistor prevents the transistor from destroying itself), and when the output capacitor has discharged itself through the load, the transistor recovers, ready for the next driving pulse. The transistor is run at a rate of about 1 kHz to avoid overheating.

With an ordinary type V405A silicon planar transistor, and careful construction, an output of amplitude 28 V, full-width at half maximum (FWHM) of 1 ns, and a risetime, which was limited by the sampling oscilloscope used, to 0.35 ns.

The LED used was a Fairchild type FLVIO, with a quoted light output risetime of 10 ns. By driving the LED at a much higher forward current than the manufacturers recommended maximum for a very short period, risetimes much shorter than the quoted figure can be achieved. If the LED driving pulse is too short in duration, no light output is observed. With the pulse generator modified to produce an output of 8 V, FWHM of 2 ns, and a risetime of 1 ns, the coincidence curve shown in Fig. 19 was observed. The full-width at half maximum of this curve is measured as about 3.2 ns.

Fig. 20 shows a coincidence curve produced by cosmic rays, over a period of about 18 hours. The FWHM here is measured as 2.25 ±.02 ns. If we assume that the cosmic
A positive pulse, derived from a differentiated multivibrator output, of amplitude about 2 V is applied directly to the base. The transistor turns on rapidly, and avalanche breakdown is initiated. The collector voltage drops very rapidly, (the large value of the collector resistor prevents the transistor from destroying itself), and when the output capacitor has discharged itself through the load, the transistor recovers, ready for the next driving pulse. The transistor is run at a rate of about 1 kHz to avoid overheating.

With an ordinary type V405A silicon planar transistor, and careful construction, an output of amplitude 28 V, full-width at half maximum (FWHM) of 1 ns, and a risetime, which was limited by the sampling oscilloscope used, to 0.35 ns.

The LED used was a Fairchild type FLV10, with a quoted light output risetime of 10 ns. By driving the LED at a much higher forward current than the manufacturers recommended maximum for a very short period, risetimes much shorter than the quoted figure can be achieved. If the LED driving pulse is too short in duration, no light output is observed. With the pulse generator modified to produce an output of 8 V, FWHM of 2 ns, and a risetime of 1 ns, the coincidence curve shown in Fig. 19 was observed. The full-width at half maximum of this curve is measured as about 3.2 ns.

Fig. 20 shows a coincidence curve produced by cosmic rays, over a period of about 18 hours. The FWHM here is measured as 2.25 ±.02 ns. If we assume that the cosmic
ray coincidences give a delta function stimulation, then the FWHM of Fig. 20 is the system resolution time. The LED light pulse duration can then be estimated as,

\[ t_{\text{led}} = \sqrt{(3.2)^2 - (2.25)^2} \text{ ns} \]

\[ = 2.3 \text{ ns approximately.} \]
Fig. 19.

Coincidence peak produced by LED light pulser.

Full width at half maximum = 3.2 ns.
Fig. 20.
Coincidence peak produced by cosmic rays.

Full width at half maximum = 2.25 ± .02 ns.
III.13. Signal to noise ratio.

If the number of two-photon decays in view of the coincidence detectors per second is \( N_0 \), and the detector efficiencies are \( e_1 \) and \( e_2 \) respectively, then the rate of true coincidences detected per second is,

\[
N_t = e_1 e_2 N_0
\]  

III.8.

If the resolving time of the coincidence circuit is \( t \), then it is easily shown that the randoms rate due to uncorrelated photons is,

\[
N_r = e_1 e_2 N_0^2 t
\]  

III.9.

(If a simple coincidence circuit is used, a factor of 2 appears in equation III.9, due to its symmetrical inputs.)

The true to random ratio is then,

\[
\frac{N_t}{N_r} = \frac{1}{N_0 t}
\]  

III.10.

so if either \( N_0 \) or \( t \) is made small enough, the true to random ratio can be made as large as desired. However, if the beam emits other uncorrelated photons with rate, \( aN_0 \) per second, the detectors have noise rates of \( n_1 \) and \( n_2 \) per second respectively, and the number of counts due to cosmic rays is \( N_{cr} \) per second, it can be shown that the true to randoms ratio is now,

\[
\frac{N_t}{N_r} = \left[ N_0 t \left( 1 + \frac{n_1 + N_{cr}}{e_1 N_0} + a \right) \left( 1 + \frac{n_2 + N_{cr}}{e_2 N_0} + a \right) \right]^{-1}
\]  

III.11.
Equation III.11 has a maximum value at,

\[ N_0 = (1 + a)^{-1} \left[ \frac{(n_1 + N_{cr})(n_2 + N_{cr})}{e_1 e_2} \right]^{\frac{1}{2}} \]  

III.12.

Now the error in the number of true coincidences, in the presence of the random background, is, from Poisson statistics,

\[ \Delta (N_t T) = \sqrt{N_t T + N_r T}, \]

where \( T \) is the time taken to accumulate the time spectrum. The fractional error in the number of true counts is then,

\[ f = \frac{\sqrt{(N_t T + N_r T)}}{N_t T} \]  

III.13.

Substituting equation III.11 into equation III.13,

\[ f = (e_1 e_2 T)^{-\frac{1}{2}} \left[ \frac{1}{N_0} + t \left( 1 + \frac{n_1 + N_{cr}}{e_1 N_0} + a \right) \left( 1 + \frac{n_2 + N_{cr}}{e_2 N_0} + a \right) \right]^{\frac{1}{2}} \]

- III.14.

Now if the beam intensity is increased without limit, \( (N_0 \to \infty) \), then we have,

\[ f = (1 + a) \left[ \frac{t}{e_1 e_2 T} \right]^{\frac{1}{2}} \]  

III.15.

which is independent on \( N_0 \).

Equation III.14 shows that the best statistical accuracy is achieved at the highest source strength, and from equation III.11, at the lowest true to random ratio.

In practice the onset of saturation limits the maximum count rates that can be tolerated. It can be seen, also, from Fig. 12, that \( N_0 \), which is proportional to the metastable density, and \( aN_0 \), which is proportional to the neutral current, do not have a linear relationship as was
assumed above by introducing the factor a, certainly above about 1.1 keV. Nevertheless, it can be seen that the region of maximum metastable density, around 1 keV, from Fig. 12, will give the best statistical accuracy, and therefore the best signal to noise ratio.

The expected background rate can be estimated from the number of photons emitted per second through collisional quenching of the metastables, $R$ estimated in section II.4.3, page 32a. Since the metastable fraction of the total neutral beam has been estimated in section III.9, page 61, as about 0.1, and if we take as an upper limit, the same cross-section for emission of a photon by a ground state neutral in collision with a background gas molecule, as for a metastable atom, then the number of photons emitted per second by the total neutral beam is,

$$ R' \approx 2.5 \times 10^5 \text{ s}^{-1} $$

The random coincidence rate produced by this background can be written, using equation III.9, as,

$$ N_r = e_1 e_2 (R')^2 t $$

where $e_1$ and $e_2$ are the products of the quantum efficiencies and solid angles subtended at the beam by the two coincidence detectors respectively. We have, $e_1 e_2 \approx 2.8 \times 10^4$. $t$ is the time delay per channel of the coincidence circuit, which is $2.5 \times 10^{-10}$ s. These values give,

$$ N_r \approx 4.5 \times 10^{-3} \text{ s}^{-1} $$

as the random background per coincidence channel.
IV. Results and discussion.


With a stable, dense metastable beam, produced by 1 keV protons at the optimum cesium thickness, the data collection and measurement methods are as described below.

The oxygen flow to the metastable monitor Lyman-alpha filter is increased until the count rate reaches an equilibrium value. Voltages of ± 250 V, and ± 45 V are applied to the switch unit and the start button is pressed. The high and low voltages are switched alternately to the pre-quench/proton removal plates, which switch the metastable component of the beam off and on respectively, through Stark quenching.

The output of the time to pulse height converter is switched synchronously to subgroups A and B of the multi-channel analyser memory. The subgroups contain 200 channels each, and each channel represents a time delay of $2.5 \times 10^{-10}$ s.

The monitored parameters are, apart from the coincidence time spectra; the integrated singles counts, one of the singles count rates, the metastable monitor output with the metastable beam on and off, the metastable monitor rate, the neutral detector current, and the integrated beam on and beam off times. The multi-channel analyser also provides, as well as the time spectra directly, the difference time spectrum, the integrated number of counts
in each of the three spectra, as well as the integral under each peak. The difference time spectrum should contain the two-photon peak. All these data are punched on paper tape for later analysis, either by hand or by computer, at the end of a run.

IV. 2. Background radiation.

The singles rate, and therefore the randoms background, was found to be critically dependent on slight changes in the focusing conditions, and in the direction of the beam, which tended to drift randomly with time. As a consequence, the signal to noise ratio could vary by up a factor of two between different runs, which lasted typically 20 hours.

The pressure dependence of the background was found to be negligible, i.e., at about $3 \times 10^{-8}$ torr in the detection region, a six-fold increase in pressure produced only a 1% increase in the singles rate. It can be concluded, then that the major contribution to the randoms background is due to beam-surface collisions, either in the quartz tube, or in chamber UHV II. As a result, the normal operating pressure in the detection region was allowed to remain in the $10^{-8}$ torr range, the liquid nitrogen cold traps being unfilled.

An optical spectrometer, observing (through a sectrosil window) the proton beam striking a stainless-steel surface, saw only the Balmer series of lines. It can be concluded that the Balmer-beta line is the main component of the background radiation, since the photomultipliers
sensitivities are very low at the wavelength of the Balmer-alpha line, which is 486.1 nm. The 9816QB photomultiplier tube always gave the highest singles count rate, when the beam was running, even though the quantum efficiencies of both photomultiplier tubes were similar, for since its spectral sensitivity extends to about 750 nm, it is sensitive to the Balmer-alpha line, although its sensitivity is much reduced at this wavelength.

Table IV.1 compares the number of random coincidences per memory channel observed, and those expected from the integrated singles counts. The expected randoms count $R$ per channel is calculated from equation III.9,

$$R = N_1 T = \frac{N_1 N_2 t}{T}$$

where $N_1$ and $N_2$ are the integrated singles counts from detectors 1 and 2 respectively, ($N_1 = e_1 N_0 T$ ; $N_2 = e_2 N_0 T$). The channel width, $t$, is fixed at $2.5 \times 10^{-10}$ s. The more accurate formula for $N_r$, used in III.11 is not used as some of the parameters are not accurately known. A small correction has to be applied owing to the different 'on' times of the singles circuit and the coincidence circuit. The coincidence circuit is gated off for 0.655 s every minute, as mentioned in section III.11.2, and about a 1.5% correction is required. The singles scalers also count the coincident signals, of course, but these are negligible compared with the randoms counts.

The agreement between the measured and calculated values is satisfactory, which indicates the correct operation of the circuits.
<table>
<thead>
<tr>
<th>Run</th>
<th>$T$ (sec.)</th>
<th>$N_1$</th>
<th>$N_2$</th>
<th>$R_{\text{meas.}}$</th>
<th>$R_{\text{calc.}}$</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>$8.24 \times 10^4$</td>
<td>$5.98 \times 10^9$</td>
<td>$1.99 \times 10^9$</td>
<td>$2.95 \times 10^4$</td>
<td>$3.61 \times 10^4$</td>
<td>0.83</td>
</tr>
<tr>
<td>52</td>
<td>$7.42 \times 10^4$</td>
<td>$2.03 \times 10^9$</td>
<td>$1.06 \times 10^9$</td>
<td>$7.87 \times 10^3$</td>
<td>$7.25 \times 10^3$</td>
<td>1.09</td>
</tr>
<tr>
<td>53</td>
<td>$7.64 \times 10^4$</td>
<td>$1.60 \times 10^9$</td>
<td>$4.76 \times 10^8$</td>
<td>$3.24 \times 10^3$</td>
<td>$2.49 \times 10^3$</td>
<td>1.31</td>
</tr>
<tr>
<td>54</td>
<td>$8.28 \times 10^4$</td>
<td>$2.61 \times 10^9$</td>
<td>$8.83 \times 10^8$</td>
<td>$7.00 \times 10^3$</td>
<td>$6.96 \times 10^3$</td>
<td>1.01</td>
</tr>
<tr>
<td>55</td>
<td>$6.85 \times 10^4$</td>
<td>$1.95 \times 10^9$</td>
<td>$6.92 \times 10^8$</td>
<td>$5.72 \times 10^3$</td>
<td>$4.92 \times 10^3$</td>
<td>1.17</td>
</tr>
<tr>
<td>56</td>
<td>$7.15 \times 10^4$</td>
<td>$5.30 \times 10^9$</td>
<td>$1.84 \times 10^9$</td>
<td>$3.05 \times 10^4$</td>
<td>$3.41 \times 10^4$</td>
<td>0.90</td>
</tr>
<tr>
<td>60</td>
<td>$6.19 \times 10^4$</td>
<td>$2.91 \times 10^9$</td>
<td>$1.08 \times 10^9$</td>
<td>$1.27 \times 10^4$</td>
<td>$1.27 \times 10^4$</td>
<td>1.01</td>
</tr>
<tr>
<td>61</td>
<td>$8.84 \times 10^4$</td>
<td>$5.45 \times 10^9$</td>
<td>$2.85 \times 10^9$</td>
<td>$3.24 \times 10^4$</td>
<td>$3.24 \times 10^4$</td>
<td>0.89</td>
</tr>
</tbody>
</table>

Table IV.1.
Comparison of measured and calculated random coincidences per channel. $T$ = Total time, $N_1$ = integrated singles count from photomultiplier 9816QB, $N_2$ = from XP1023, $R_{\text{meas.}}$ = measured number of random counts per channel, $R_{\text{calc.}}$ = number of random counts calculated, (see text) Ratio = $R_{\text{calc.}} / R_{\text{meas.}}$ (corrected).
IV.3. The coincidence peak.

The spurious coincidences, presumably due to cosmic rays, arrive at a constant rate, depending only on the geometry and sensitivity of the detection system, and independently of all other conditions in the apparatus. This rate is about $0.25 \text{s}^{-1}$, and gives rise to relatively large peaks in the coincidence spectra for the metastable beam on and for the metastable beam quenched. On subtracting these spectra the residual coincidence peak is observed, approximately half the size of the spurious peak, under the optimum conditions expected for observation of the two-photon decay.

The number of counts under the peak is observed to be proportional to the metastable density, for a constant detection geometry. These coincidences are ascribed to the two-photon decay of metastable atomic hydrogen, and a number of tests will be described below which confirm this belief.

Difference time spectra obtained by the method described in section 1 are shown in Fig. 21 and Fig. 22. The total times taken to obtain these spectra were about 21 hours each. The number of counts under the peaks is about 4,000 in each case. Each point shown in the figures is the sum of two adjacent multi-channel analyser memory channels. The time delay corresponding to each point is then 0.5 ns. Both curves display a full width at half maximum of about 4.5 points as shown, or 9 memory channels, which gives the FWHM of each curve as about 2.25 ns.
Fig. 21.
Coincidence curve with a metastable beam passing through detection region, angle between detectors 90°. Each point represents delay of 0.5 ns. Total time about 21 hours.
Fig. 22.
Same as fig. 21, but with angle between detectors at 180°. Total time about 21 hours.
This is the same value as is observed for the resolution using the cosmic ray coincidences, in section III.12.

IV.4. Null tests.

Since the two-photon coincidence peak is obtained as a difference of two larger peaks, which increase at a constant rate, then for any difference in the collection times of, or the detection system characteristics for, the initial coincidence spectra, a spurious result will be obtained.

The scalers which monitor the on-off periods of the switching relay never showed a difference greater than 0.3%. This difference was probably mostly due to electrical pick-up.

With no beam running, but with the pre-quench voltage being switched normally, coincidence spectra were taken in a period of 14.2 hours. The coincidence peaks due to the cosmic rays appeared with the normal rate, giving peak integrals of about 9,000 counts in each spectrum. The difference spectrum gave a 'peak' integral of, 46 ± 190 counts, where the error is the statistical error. If the beam had been running, the expected peak integral was about 6,000 counts.

A similar test was carried out with the LED light pulser, for a shorter time, but with a much higher coincidence rate. The result was again zero within the error.

A null test, identical to the first one described above, but with a hot-wire ionization gauge switched on in one of the vacuum chambers to simulate the background due to the beam, was run for a period of about 37 hours.
In this case the singles rates were about 20% of their normal values with the beam running. Again a null result was obtained, with a difference peak integral of $84 \pm 300$.

Fig. 23 shows the coincidence spectrum obtained when a metastable beam was running normally, but with a voltage of $\pm 1,000$ V applied to the pre-quench wires, (position (9) in Fig. 5.). The voltage to the pre-quench plates was being switched as usual. The expected number of counts in the peak was about 8,200, or a peak height of about 2,500. This was estimated from the metastable monitor count rate immediately before and after the run, and the neutral detector output, which gave an indication of the beam stability throughout the run. The actual number of counts measured at the peak location was $240 \pm 900$. A number of such runs were made, and each one was null within the error.

The count distribution in Fig. 23 is not symmetrical about zero, which implies that some other component of the beam is affected by the switching system. This must be the residual charged component. It was pointed out in section III.7, page 53, that a small fraction of the protons enter the quartz tube when the pre-quench plate voltage is low. The pre-quench wires do not deflect the proton beam efficiently, having been designed to perform the opposite function. This test then shows that the residual charged component of the beam does not produce coincidences.
**Fig. 23.**

Coincidence spectrum with metastables fully quenched.

Total time about 22 hours.
IV.5. Observed characteristics of the two-photon decay.

IV.5.1. Angular distribution.

Fig. 24 shows the measured angular distribution of the true coincidences. The points are the integrated number of counts under the peak, normalised to the integrated metastable monitor output. The points at 90° and at 135° are further normalised to the value at 180°. The point at 135° took 42 hours to accumulate, while the other two points took about 21 hours each. The error bars show one standard deviation. The solid curve is a semi-analytical folding of the photocathode solid angles with a \((1 + \cos^2 \theta)\) function, for a point source. This folding operation has the effect of flattening the distribution curve. The agreement can be seen to be satisfactory.

If either the rate of true coincidences, or the rate of cosmic ray coincidences, is plotted against the angle between the photomultiplier tubes, there is no apparent correlation between the plotted points and the predicted angular distribution. Table IV.2 shows the normalised true coincidences, the true coincidence rate, and the cosmic ray coincidence rate, for comparison.

Table IV.2.

<table>
<thead>
<tr>
<th>Angle</th>
<th>Normalised true coincidences</th>
<th>True coinc. rate / sec.</th>
<th>Cosmic ray rate / sec.</th>
</tr>
</thead>
<tbody>
<tr>
<td>90°</td>
<td>((7.85 \pm .75) \times 10^{-5})</td>
<td>.116 ± .012</td>
<td>.442 ± .01</td>
</tr>
<tr>
<td>135°</td>
<td>((1.18 \pm .10) \times 10^{-4})</td>
<td>.114 ± .014</td>
<td>.238 ± .01</td>
</tr>
<tr>
<td>180°</td>
<td>((1.42 \pm .10) \times 10^{-4})</td>
<td>.112 ± .008</td>
<td>.289 ± .01</td>
</tr>
</tbody>
</table>
Normalised coincidence peaks at $180^\circ$, $135^\circ$, and $90^\circ$ compared with predicted angular distribution.
IV.5.2. Spectral distribution.

The photomultiplier tubes respond to about 56% of the two-photon spectrum, and since the spectral sensitivities of both tubes are similar, either tube can respond to either photon.

A glass filter, which has a low wavelength cut-off of about 500 nm was placed over the sensitive area of one of the photomultiplier tubes, causing the other tube only to respond to the short-wavelength photon. This reduces the spectral response of the whole coincidence circuit by about a factor of five. After a run lasting about 44 hours, no significant number of coincidences was recorded. Although a small peak was expected to be observed after this time, a detailed folding of the filter response with the two-photon spectrum and the photomultiplier response was not done, and so there is a very large uncertainty in the expected value. It can be concluded, therefore, that this result does not disagree with predictions of the two-photon spectral distribution.

IV.5.3. Lifetime.

A very rough estimate of the lifetime due to the two-photon decay can be attempted from the count rates observed. The number of coincidences arising from the two-photon decay can be expressed as,
\[
N(2\gamma) = \frac{j(2S)}{v} \eta G \gamma A
\]

where \( j(2S) \) is the flux of metastables through the quartz tube; \( v \) is the beam velocity, about \( 4.4 \times 10^7 \) cm s\(^{-1} \); \( G \) is the geometrical factor for coincidence detection, including the \((1 + \cos^2 \theta)\) term, which has been calculated to be, \( 1.5 \times 10^{-2} \); \( \eta \) is the length of the observation, about 3 cm; \( \gamma \) is the product of the peak quantum efficiencies of the coincidence photomultipliers, about \( 7 \times 10^{-2} \); \( F \) is the fraction of the two-photon spectrum which can be observed, about 0.56; \( A \) is the transition probability for the two-photon decay.

The metastable flux can be found from the metastable monitor output, which can be expressed as,

\[
N(L_m) = j(2S) \eta
\]

where \( \eta \) is the overall efficiency of the metastable monitor, which is, from section III.9, \( 8.1 \times 10^{-3} \).

Eliminating \( j(2S) \) from IV.2 and IV.3, we get the transition probability as,

\[
A = \frac{N(2\gamma)}{N(L_m)} \frac{\gamma F v}{\eta} \quad \text{IV.4.}
\]

The quantity \( N(2\gamma)/N(L_m) \) can be taken from table IV.4.2, for the \( 180^\circ \) position as about \( 1.4 \times 10^{-4} \). (The geometrical factor \( G \) above is calculated for the \( 180^\circ \) position.) Substituting all these values in IV.4 we get for the transition probability,

\[
A = 0.6 \text{ s}^{-1} \quad \text{or} \quad t_{2S} = 1.7 \text{ s}
\]
The predicted lifetime is\(^9\), \(0.12153 \pm 0.00002\) s. Considering the very large uncertainties in the above estimate, which are difficult to estimate themselves, but are expected to be at least a factor of 10, the measured value of 1.7 s cannot be considered to be in disagreement with the theoretical prediction.

IV.6. Suggestions for future work.

The apparatus is capable of running continuously for 100 hours or more with the minimum of attention. It should thus be possible to reduce the size of the error bars on the points for the angular distribution curve, and observe at a much greater number of angles.

A solar-blind photomultiplier tube is available, with spectral sensitivity extending from 120 nm to 300 nm, but with a small sensitive area and low quantum efficiency, and gain. However, it is not sensitive to the Balmer series and thus substantially reduces the random background. Because of its smaller size, the expected angular distribution curve should be less flattened, and so a much more accurate measurement of this quantity should be obtained.

The spectral distribution could be determined using broadband filters and very long measuring times.

The metastable monitor could be calibrated absolutely and the coincidence photomultipliers calibrated as well, and the geometry of the detecting systems determined more accurately, to give the first direct measurement of the metastable hydrogen lifetime, probably with an error of about 50\%. 
IV.7. Conclusion.

On the basis of the tests and measurements described above, it can be concluded that the coincidences observed are associated with the $\frac{3}{2}$ state of atomic hydrogen. The observation of the angular dependence of the coincidences make unlikely the possibility that they arise from cascades, excited in some way by the metastable state. Cascades would also be expected to produce an observable 'tail' in the shape of the coincidence curve. It can be finally be concluded that the two-photon decay of metastable atomic hydrogen has been observed, and its measured characteristics are in agreement with predictions.
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ERRATA.

Page 10, line 5, should begin:
"of singly-stimulated two-photon emission ... ".

Page 12, equation II.1., and subsequent equations, replace "h" by "\hbar".

Page 15, equation II.10., replace "\hat{A}" by "\hat{A}0".

Page 15, equation II.12., and page 19, equation II.23., insert modulus brackets around the respective matrix elements before squaring.

Page 19, last line should read: "Substituting equation II.24. in equation II.23, ".

Page 26, equation II.36., the multiplying factor should be: \( \omega_3^2 \omega_4^3 \omega_1 \).

Page 39, line 8, delete "which".

Page 69, line 4, read "gated" for "gated".

Page 70, line 4, read "limited" for "limited".

Page 93, reference 13 should read:
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