FINE STRUCTURE OF HYDROGEN
USING QUANTUM BEAT SPECTROSCOPY

by

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Submitted in partial fulfillment of the requirements for the degree of Bachelor of Science at the
MASSACHUSETTS INSTITUTE OF TECHNOLOGY
May, 1975

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Chairman, Departmental Committee on Theses
ABSTRACT

A measurement of the fine structure in the 6D state of hydrogen is attempted using quantum beat spectroscopy. The experiment did not yield a value for the energy splitting. Various spectroscopic methods for measuring energy splittings are explained, the experiment is described, and possible reasons for its failure are discussed.
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ACKNOWLEDGEMENTS

This experiment was neither conceived nor performed in vacuo. There was a constant interaction with other people. Dan Kleppner originally suggested the experiment and aided my understanding of the theory of quantum beats. Assistance with the experimental apparatus, especially the laser and computer, was ably provided by Mike Littman, Ted Ducas, and Myron Zimmerman. Thanks are owed especially to Mike and Ted, but also include others too numerous to list. Funding and materials for the experiment were generously provided by the Research Laboratory of Electronics.

The gratitude I feel for my parents' support is far beyond that which words can express.
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I. MEASUREMENT OF SMALL ENERGY SPLITTINGS

I.1 INTRODUCTION

Quantum beat spectroscopy (QBS) is a modern technique used for measuring small energy splittings in atoms and molecules. Measurements have been made of Zeeman splittings,\textsuperscript{1-4} fine structure,\textsuperscript{5,6} and hyperfine structure.\textsuperscript{7} QBS involves the preparation of a coherent superposition of the states of interest. A coherent superposition of states can be prepared by a pulse of light if the duration of the pulse is short enough, and is at the frequency of the resonant radiation. The fluorescence observed then exhibits modulations at the frequencies corresponding to the energy splittings between the states. QBS was used in this experiment to measure the fine structure of the $6D$ state of hydrogen. QBS is not the only spectroscopic technique to measure small energy splittings in atoms and molecules. To aid in the understanding of QBS, a description of the different spectroscopic techniques that have been used to measure closely spaced energy levels is given. The limitations on the precision of the measurements of energy splittings are also discussed.

I.2 SPECTROSCOPY

A. Limitations on Measurements

Broadening of spectral lines limits the accuracy with which closely spaced structure can be measured. The elimi-
nation of broadening is important, because the measurement of closely spaced structure provides information about the atom or molecule as well as tests of theory. The shape of the spectral line is due to the type of broadening. Three types of broadening are considered here: (1) Doppler broadening, the largest, (2) the natural linewidth, which is the ultimate limit to elimination of broadening, and (3) pressure broadening. The parameter used to measure the amount of broadening is the full-width half-maximum (FWHM) frequency spread, which is written $\Delta \nu$. The frequency of a spectral line can be determined with a smaller error than the FWHM frequency spread using curve fitting techniques. Even so, the elimination of that broadening allows more accurate measurements of an energy splitting.

Doppler broadening is an example of inhomogeneous broadening, which means that the atoms do not interact with an E&M field identically. The inhomogeneously broadened line results from each atom in the ensemble emitting light centered at a different frequency, rather than the light from each atom being broadened with each photon centered at the same frequency. The measured frequency of a light emitter is a function of the relative velocity of the emitter and the observer

$$\nu = \nu_o \left(1 + \frac{v_x}{c}\right)$$

where:

$\nu$, $\nu_o$ are the frequencies measured by the observer and the emitter respectively.

$v_x$ is the component of velocity of the emitter in the direction of the observer.
Thus, if the distribution of velocities is \( g(v_x) \)

\[
I(v_x) = I_0 g(v_x).
\]

In a gas, the atoms have a Maxwell velocity distribution.\(^8\)

\[
I(\nu) \sim \exp(-Mv_x^2/2kT) = \exp\left(\frac{\left(\frac{Mc^2}{2kT}\right)\left(\nu-\nu_0\right)^2}{\nu_0^2}\right)
\]

This emission profile is a Gaussian with a FWHM frequency spread of

\[
\frac{\Delta \nu}{\nu_0} = \left(\frac{8 \ln2 kT}{Mc^2}\right)^{1/2}
\]

For hydrogen at 300°C, \( \Delta \nu/\nu_0 = 5 \times 10^{-6} \).

The natural linewidth results from the Heisenberg uncertainty principle, which is \( \Delta E \Delta t \approx h \) or \( \Delta \nu \Delta t \approx 1/2\pi \). Since most of the atoms decay and emit a photon within one lifetime, the uncertainty in time for any one photon is of the order of the lifetime,

\[
\tau_i = \sum A_{ij}
\]

where \( A_{ij} \) are the Einstein A coefficients.

The natural linewidth has a Lorentzian lineshape, which is simply the Fourier transform of an exponential decay.

\[
I \sim \text{Re}\left[\int e^{-t/\tau} e^{i\omega t} dt\right] = \text{Re} \frac{1}{1/t + i\Delta \omega}
\]

\[
= \frac{\Gamma}{(1/\tau)^2 + (\Delta \omega)^2}
\]

where:

\[
\Gamma = \frac{1}{\tau} \text{ is one over the lifetime.}
\]

\[
\Delta \omega = 2\pi \Delta \nu = 2\pi (\nu - \nu_0).
\]

Thus, the FWHM frequency spread is \( \Delta \nu = \frac{1}{\pi \tau} \). In passing, it should be noted that one can obtain precision better than the natural linewidth by use of the Ramsey double resonance technique or by fitting the spectral lineshape to an ideal Lorentzian lineshape. The natural linewidth is a homogeneous
broadening, because each atom emits a spectral line with the natural linewidth.

Pressure broadening (which is homogeneous) arises from the perturbation of an atom during a collision. If the time between collisions is larger than the lifetime of the state of interest each atom radiates independently of all the others. Since the mean time between collisions increases when the pressure is decreased, pressure broadening becomes less important when the pressure is decreased. For a state with a lifetime of 100 nsec, pressure broadening is smaller than the natural linewidth below a pressure of 40 torr.

B. Elimination of Doppler Broadening

Several techniques are available for elimination of Doppler broadening. Beam spectroscopy minimizes the Doppler shift for the observer, because the component of beam velocity parallel to the direction of observation is zero; that is, the beam travels perpendicular to the line of sight. However, the number of atoms in a beam is usually smaller than the number of atoms in a cell; so, precision is gained at the cost of intensity. Radiofrequency (rf) spectroscopy can be used to measure small energy splittings by inducing electric dipole transitions between closely spaced energy levels to obtain a spectrum. The Doppler broadening in the rf range is about one part in $10^5$ ($10^{-7}$ cm$^{-1}$). This is usually a very small uncertainty in energy compared with the natural linewidth. A third way of eliminating Doppler broadening is to use laser spectroscopy - saturation spectroscopy, two photon spectroscopy, or quantum beat spectroscopy.
Saturation spectroscopy excites the atoms or molecules in a gas cell using monochromatic light from a laser (the bandwidth of the laser being much smaller than the Doppler broadening). A probe beam from the same laser traverses the saturated region in the opposite direction (see Fig. 1.1). When the laser excites atoms travelling with zero velocity along the axis of the laser beams, then the probe beam is not absorbed by the already excited atoms. If the probe beam intensity or the fluorescence from the initial state is monitored, a peak (or dip) in the intensity occurs at the center of the Doppler broadened line. The experiment described is just one example of saturation spectroscopy known as the Lamb dip. Other forms of saturation spectroscopy use a second laser as the probe beam, resulting in a peak (or dip) for some non-zero velocity ensemble.

Two photons propagating in opposite directions from the same source excite a transition at an energy equal to twice the energy of an individual photon, even though there is no resonant intermediate state. This is a second order process and is much less likely to occur than a stepwise excitation through a resonant intermediate state. All the atoms in a gas will find that the sum of the frequencies of the two light beams will be constant even though the frequency of each light beam will be different for atoms moving with speeds \( v_x \) parallel to the axis of the light beams (see Fig. 1.2).

\[
h\nu = h\nu_1 (1 - \frac{v_x}{c}) + h\nu_1 (1 + \frac{v_x}{c}) = 2h\nu_1
\]

where \( \nu_1 \) is the frequency of the laser.

Thus, a detector of the excited state can determine an energy separation without Doppler broadening.
Figure 1.1 - The Lamb dip provides a means for finding the center of a Doppler line by punching a "hole" in the Doppler line with a characteristic width of the natural linewidth.
TWO PHOTON SPECTROSCOPY

Figure 1.2 - Though atoms with different velocity components in the x direction see different frequencies, their sum is always a constant. To find the energy, simply tune the laser until the upper state gets populated.
In saturation and two photon spectroscopy, small energy splittings must be measured by finding the difference in frequency between two spectral lines. QBS allows the direct measurement of a small energy splitting in an excited state. However, QBS measurements of ground state energy splittings cannot be done according to quantum electrodynamics.\textsuperscript{10}

1.3 QUANTUM BEAT SPECTROSCOPY

A. Level Crossing

Level crossing, a phenomenon related to quantum beats, can also be used to measure small energy splittings directly. The energy levels of an atom are dependent upon the magnetic field the atom experiences. Thus, at certain values of the magnetic field, two different energy levels may become degenerate; (the energy splitting becomes less than the natural linewidth). At that magnetic field, the intensity of the fluorescence of a particular polarization changes radically. This phenomenon was first noted by Hanle (1926) in the special case where the magnetic field is zero. Breit (1933)\textsuperscript{11} presented a theory which explained level crossing. The first level crossing experiment with a non-zero magnetic field was done by Franken (1959)\textsuperscript{12}.

Consider ground states $|m\rangle$, $|m'\rangle$ and excited states $|\mu\rangle$, $|\mu'\rangle$ where $|\mu\rangle$ and $|\mu'\rangle$ are excited from state $|m\rangle$ and decay to a state $|m'\rangle$. When $|\mu\rangle$ and $|\mu'\rangle$ are degenerate, a crossing occurs and the intensity of the fluorescence after excitation goes as:

$$I \sim \left| f_{m\mu} g_{\mu m} + f_{m\mu'} g_{\mu' m'} \right|^2$$

where:
\[ f_{m\mu} = \langle m \mid \hat{r} \cdot \hat{r} \mid \mu \rangle \] is the dipole matrix element for the excitation of state \( |\mu\rangle \) from \( |m\rangle \) with light of polarization \( \hat{r} \).

\[ e_{\mu'm'} = \langle \mu' \mid \hat{e} \cdot \hat{r} \mid m' \rangle \] is the dipole matrix element for the decay of \( |\mu\rangle \) to \( |m'\rangle \) with light of polarization \( \hat{e} \).

When the excited states do not cross, the intensity of the fluorescence is the sum of the intensities from the states \( |\mu\rangle \) and \( |\mu'\rangle \) individually.

\[ I \sim |f_{m\mu} e_{m\mu'}|^2 + |f_{m\mu'} e_{\mu'm'}|^2 \]

Thus, the resonance fluorescence with a particular polarization is different when the levels are crossed (see Fig. 1). Two states can be thought to share the same photon if that photon can excite both of the upper states; this occurs in level crossing.

The energy splitting in zero magnetic field can be calculated if the magnetic field at which level crossing occurs and the \( g_J \) factors for each state are known. The similarities between QBS and level crossing arise from the fact that in both phenomena the excited states can share the same photon. By using excitation with a short duration, thus insuring that the energy uncertainty of the photon is large, a single photon can excite the states \( |\mu\rangle \) and \( |\mu'\rangle \). This is the basis for QBS; it is a pulsed version of level crossing.

B. Theory

If two states that decay to the same ground state are excited coherently, the intensity of the fluorescence with a particular polarization observed at a right angle to the incoming radiation will not be a simple exponential. Instead, it is a modulated exponential; the modulation
Figure 1.3 - Level crossing occurs in a magnetic field only when the difference in $m_j$ is 0 or 2.
frequency being equal to the frequency of the energy splitting \( \nu_{\text{mod}} = \Delta E / h \). Assume that a multi-level system is excited by a pulse of polarization \( \vec{f} \) with a duration short enough that the excited states cannot be resolved. The instantaneous rate at which a photon of polarization \( \vec{g} \) is emitted by the excited atom is \(^{13}\):

\[
R(\vec{f}, \vec{g}, t) = \sum f_{\mu m} f_{\mu', m'} g_{\mu, m'} g_{\mu', m} \exp\left[ \left( 2\pi i \nu(\mu, \mu') - \Gamma \right) t \right]
\]

where:

- Constants due to geometrical factors are set equal to one.
- \( \nu(\mu, \mu') = \nu_{\text{mod}} \) is the frequency separation between the excited states \( |\mu\rangle \) and \( |\mu'\rangle \).

The summation is over all the states \( |m\rangle, |m'\rangle, |\mu\rangle, |\mu'\rangle \).

The excitation pulse can be provided by light, a thin metal foil, or electron impact\(^{15}\). Light excitation has become the simplest to perform since the advent of inexpensive, widely-tunable pulsed dye lasers.

The amplitude of the modulations in the exponential is dependent upon the polarization. In this experiment, the excitation pulse is linearly polarized; the direction of polarization determines the z axis. The excitation pulse travels in the y direction. Fluorescence travelling in the x direction is measured; first the light with z polarization, then the light with y polarization (see Fig. 1.4). It is an experimental observation that the amplitude of the modulation of the intensity of z polarized light is negative two times the amplitude of the modulation of the intensity of y polarized light. The calculation for the expected fluorescence in a QBS experiment confirms this observation (see Appendix).

The observed intensity is proportional to \( R \), the rate of
Figure 1.4 - Geometry of observation of fluorescence in a quantum beats experiment.
\[
R(\hat{z}, \hat{z}, t) = \sum z_{\mu m \mu m'} z_{\mu m' \mu m'} \exp \left[ (2\pi i \nu(\mu \mu') - \Gamma) t \right]
\]
\[
R(\hat{z}, \hat{y}, t) = \sum z_{\mu m \mu m'} y_{\mu m' \mu m'} \exp \left[ (2\pi i \nu(\mu \mu') - \Gamma) t \right]
\]

If \( z_{\mu m \mu m'} z_{\mu m' \mu m'} = (-2) y_{\mu m' \mu m'} \)

for each \( |m'\rangle, |\mu\rangle, |\mu'\rangle \) where \( \nu(\mu \mu') \neq 0 \), then it is clear that the fluorescence polarized in the \( z \) direction has twice the amplitude and the opposite sign of fluorescence polarized in the \( y \) direction. The matrix elements can be expressed as a 3j symbol time a reduced matrix element\textsuperscript{16}.

\[
\langle J M_J | z | J' M_{J'} \rangle = \left( \begin{array}{cc} J & J' \\ M_J & -M_J \end{array} \right) \langle J M_J || r || J' M_{J'} \rangle
\]
\[
\langle J M_J | y | J' M_{J'} \rangle = \frac{1}{\sqrt{2}} \left[ \left( \begin{array}{cc} J & J' \\ M_J & -M_J \end{array} \right) + \left( \begin{array}{cc} J & J' \\ M_J & -M_J \end{array} \right)^{-1} \right]
\]
\[
\langle J M_J || r || J' M_{J'} \rangle
\]

The reduced matrix elements cancel and the 3j symbols can be calculated. The theory confirms the experimental results.

It has been stated that the excitation pulse must be shorter than the inverse of the frequency separation. The natural question is how much shorter. Consider the real part of \( R(\hat{f}, \hat{g}, t) \):

\[
R(t) = (I_0 + I_{\text{mod}} \cos 2\pi \nu t) e^{-\Gamma t}
\]

where only a two level system is being considered.

Assume, for the sake of convenience, that the excitation pulse can be modeled as a square wave with a duration of \( t_{\text{exc}} \).

Mathematically, the fluorescence is found by convolving the fluorescence from an impulse with the shape of the excitation pulse.

\[
R(t_o) = \int_{-\infty}^{\infty} \rho(t-t_o) (I_0 + I_{\text{mod}} \cos 2\pi \nu t) e^{-\Gamma t} dt
\]

where \( \rho(t-t_o) \) is the shape of the pulse.

\[
\rho(t-t_o) = \begin{cases} 
1/t_{\text{exc}} & \text{for } t_o < t < t_o + t_{\text{exc}} \\
0 & \text{for all other } t.
\end{cases}
\]
Thus, \( R(t_0) = \frac{1}{t_{exc}} \int_{t_0}^{t_0 + t_{exc}} e^{-\Gamma t} (I_o + I_{mod} \cos 2\pi \nu t) \, dt \)

If \( t_{exc} \ll 1/\Gamma \), then \( e^{-\Gamma t} \) is a constant over times between \( t_0 \) and \( t_0 + t_{exc} \). Performing the integration results in:

\[
R(t_0) = e^{-\Gamma t_0} \left[ I_o + I_{mod} \cos(2\pi \nu (t_0 + \frac{1}{\nu} t_{exc})) \frac{\sin(\pi \nu t_{exc})}{\nu t_{exc}} \right]
\]

To observe the modulation in quantum beats with half the amplitude of quantum beats induced by an instantaneous pulse requires \(^{10}\)

\[
\frac{\sin(\pi \nu t_{exc})}{\pi \nu t_{exc}} = 0.5
\]

which yields \( t_{exc} = 5 \) nsec for an energy splitting of 130 MHz.

II. MEASUREMENT OF FINE STRUCTURE IN HYDROGEN

II.1 ORIGIN OF FINE STRUCTURE

The simplicity of hydrogen allows accurate comparison with theoretical models. Therefore, the measurement of energy separations, especially small ones, have been of great interest. Fine structure, which causes energy splittings between states with different \( j \) values, is a result of (1) the relativistic motion of the electron and (2) the interaction of the electron's spin magnetic moment with its orbital angular momentum. Together, these effects result in an energy shift of:

\[
E = \frac{\alpha^2}{4 \pi^2 \hbar^2} \left( j - \frac{4n}{j+\frac{1}{2}} \right) \text{Ry for hydrogen (Z=1)}.
\]

where:

- \( \text{Ry} = 109,737 \, \text{cm}^{-1} \) is the Rydberg (ionization energy for H)
- \( \alpha = 0.007297 \) is the fine structure constant
- \( n \) is the principal quantum number.
Measurements of the fine structure in hydrogen have been performed using all the methods described in Chapter I. The most accurate measurements of the fine structure in n=3, 4, and 5 have been done by Pipkin (1971)\textsuperscript{13} using precision rf spectroscopy. The experimental values are in good agreement with theory\textsuperscript{19} (see Table 1). If the fine structure measurement in this experiment were made to an accuracy of .1%, then a deviation from the energy splitting calculated from the above formula would be found. The deviation is predicted by quantum electrodynamics and is known as the Lamb shift.

II.2 EXPERIMENTAL DESIGN

The experimental setup is shown in Fig. 2.1. Polarized light at 4102 A from a pulsed tunable dye laser shines through a hydrogen discharge cell containing atomic hydrogen in the 2p state. Fluorescence at the same wavelength (n=2→n=6 transition) is detected by a photomultiplier (PMT) after the light passes through a linear polarizer and a monochromator. The output from the PMT is the input to a sampling scope. A computer time averages the output from the sampling scope and also allows off-line manipulation of the data. Data taken with the linear polarizer axis perpendicular to the polarization of the laser light is subtracted from the data taken with the polarizer axis parallel to the laser polarization. This data manipulation enhances the percentage modulation of the exponential decay.

A. Pulsed Tunable Dye Laser

The dye laser is pumped by a nitrogen laser (Avco C950), which has a 10 nsec pulse duration at the wavelength 3371 A. The repetition rate of the nitrogen laser can be set between
HYDROGEN ENERGY LEVELS

2p state - Fine Structure \((j=1/2 \rightarrow j=3/2)\) is 11 GHz

Hyperfine structure in \(j=1/2\) is 60 MHz
Hyperfine structure in \(j=3/2\) is 24 MHz

<table>
<thead>
<tr>
<th>Level</th>
<th>Lifetime (nsec)</th>
<th>Fine Structure* (nsec)</th>
<th>Wavelength of 2p(\rightarrow)nd (Angstroms)</th>
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<tr>
<td>3D</td>
<td>20</td>
<td>0.3</td>
<td>6563</td>
</tr>
<tr>
<td>4D</td>
<td>40</td>
<td>2.1</td>
<td>4861</td>
</tr>
<tr>
<td>5D</td>
<td>75</td>
<td>4.5</td>
<td>4340</td>
</tr>
<tr>
<td>6D</td>
<td>120</td>
<td>7.5</td>
<td>4102</td>
</tr>
<tr>
<td>7D</td>
<td>190</td>
<td>12</td>
<td>3970</td>
</tr>
<tr>
<td>8D</td>
<td>280</td>
<td>18</td>
<td>3889</td>
</tr>
</tbody>
</table>

*Values are approximate; they do not represent the latest accepted values. Experimental and theoretical results agree.
Figure 2.1 - Setup for observation of quantum beats
1 and 100 pulaes per second. The peak power of the laser pulse is about 100 kilowatts. A lens focuses the laser light into the dye cell perpendicular to the axis of the dye laser. The dye laser operates at 4102 Å using dye 409 (New England Nuclear). The optics is mounted on magnetic bases (Oriel Corp.) for ease of handling. The dye laser consists of a dye cell, mirror, grating, and telescope (see Fig. 2.2).

The quartz dye cell (Molectron) has dimensions of 1x1x6 centimeters. A small magnetic stirrer in the base of the dye cell agitates the dye. The stirrer is revolved by a small motor below the dye cell. Therefore, no pump to flow the dye is necessary. After about two hours of lasing the dye molecules dissociate and the dye must be replaced. An optically flat, uncoated quartz window is the mirror at one end of the dye laser. The mirror has an anti-reflection coating on its rear surface to prevent lasing off of that surface. Only 4% of the light needs to be reflected because of the high gain of the dye.

The grating (Oriel Corp.) is responsible for tuning the frequency of the dye laser. The grating is placed at a relatively small angle to the laser axis to make the dispersion \( \left( \frac{1}{d \nu_{\text{tune}}} \right) \) large. When the grating is at an angle of approximately 30° to the axis of the laser, the reflected light back along the axis has the maximum intensity and a large dispersion. The telescope (Spectra-Physics) expands the spatially small laser beam so that it fill the grating. Using the whole surface area of the grating increases the number of waves which interfere, causing a proportional decrease in the FWHM angle.
Figure 2.2 - Standard Setup for a pulsed tunable dye laser. The grating provides a large dispersion at its reflection maximum. The path of the lasing light is mostly as follows: (1) after laser pulse, 4% of fluorescence from dye cell reflects from the mirror; (2) stimulated emission as the light passes back through the dye cell again; (3) telescope and grating select appropriate wavelength; (4) stimulated emission with passage again through dye cell provides most of the power; and (5) 96% transmission at front window provides one pass laser pulse.
distribution for a single wavelength\textsuperscript{20}. Thus, the laser has a much smaller FWHM wavelength spread, which results in a larger spectral density. The telescope lenses have coatings to prevent lasing between the lens and the mirror.

The dye laser has at 4102 Å a peak power output of one kilowatt for pulses 4 nsec long. The repetition rate of the dye laser is the same as the rate of the nitrogen pump laser. The FWHM wavelength spread of the dye laser is approximately .5 Å.

B. Hydrogen Discharge

The hydrogen discharge cell is a cylindrical pyrex cell with an inner diameter of 1 cm (outer diameter is 3/8 inch) and a length of 10 cm (see Fig. 2.3a). The cell has a "horn" to prevent light reflection into the monochromator and two glass to ¼" Kovar seals to allow a gas flow through the cell. Two optically flat windows provide the ends of the cylindrical cell.

The gas flow system is driven by a forepump (Duoseal 1405) capable of achieving an ultimate pressure of 4 microns. The hydrogen flow, which is adjustable, is typically large enough to maintain a pressure of 100 microns, a desirable pressure for a hydrogen discharge\textsuperscript{21}. The cell is vibrationally isolated from the rest of the vacuum system by two stainless steel, flexible hoses. A Pirani gauge on the pump side of the cell measures the pressure. A zeolite trap is in the foreline to prevent contaminants from entering the system. The apparatus is capable of handling two gas flows, though only hydrogen is normally used. The connection are made using
HYDROGEN DISCHARGE

(a) GAS FLOW SYSTEM

(b) DISCHARGE CIRCUIT - CLASS C AMPLIFIER

Figure 2.3 - Hydrogen Discharge System.
Capacitances in picofarads. Resistances in ohms.
Torr-Seal, an epoxy.

Originally, the hydrogen discharge cell was stainless steel tubes in the shape of a cross with windows at the ends of the tubes (see Fig. 2.4). An electrical feedthrough (from Ceramaseal) capable of handling radiofrequencies was located in the top of the cell. A double loop of wire in the discharge cell provided enough power to run a discharge. However, the power density in the discharge was small due to the large volume of the cell—about 1 liter. Connections in this cell were made using soft solder.

The circuit for supplying the radiofrequency power is similar to circuits previously used in hydrogen maser work. The discharge circuit (see Fig. 2.3b) maintains a hydrogen discharge with 3 to 100 watts of power provided by a Lambda power supply. The circuit is not cooled by forced air, though heat is dissipated to the surroundings by conduction and convection. The nature of the hydrogen discharge is not a strong function of the power into the circuit. The power is coupled to the discharge by a solenoid with 20 turns wrapped around the discharge cell. The rf circuit operates at a frequency of 150 MHz. The capacitance in the tank circuit is provided by stray capacitance.

C. Detection of Fluorescence

The resonance fluorescence is filtered by a linear polarizer and a monochromator (Heath) set at 4102 Å, before being detected by a photomultiplier (RCA 7265). The PMT has a rise time of 2 nsec and a relatively low dark current. It was operated with an anode to cathode voltage of 2400 volts.
Figure 2.4 - Original Discharge Cell and Experimental Setup.
Under these conditions, the efficiency of photon detection is 10%. The output of the PMT is the input to a time averaging sampling scope - a Tektronix 7904 oscilloscope and a PDP-11/20 computer with lab peripherals.

The Tektronix oscilloscope has a sampling scope module with a rise time of 300 picoseconds. It is triggered by an electrical pulse from the nitrogen laser. The scope outputs the horizontal sweep, the vertical trace, and the trigger to the computer. The computer, using a sample-and-hold, digitizes the vertical trace of the scope when the horizontal sweep reaches a certain voltage. After the next trigger (laser pulse), the computer increments the voltage for the horizontal sweep at which it samples and holds the vertical trace. In this way, the computer completes a single sweep of data in two seconds. The sweep of the data is then repeated and averaged over time. The plot of the data is displayed on a scope (Tektronix) or can be plotted on a digital plotter (Hewlett-Packard 7210A).

II.3 EXPERIMENTAL RESULTS

A. Characteristics of Discharge

A good hydrogen discharge has a red color due to the Balmer \( \alpha \) line at 6563 A. A blue discharge indicates a large amount of molecular hydrogen or an absence of hydrogen, in which the residual nitrogen gives the blue color. A red discharge can also be obtained from water vapor. The discharge contains many additional lines due to the water vapor and oxygen, but the atomic hydrogen spectral lines are as strong in the water vapor discharge as in the hydrogen discharge.
Although much research has been done on hydrogen discharges, it is still a black art in which desirable conditions must be achieved by trial and error.

An estimate of the density of hydrogen atoms in the 2p state is required to know whether quantum beats are observable. A lower estimate of the density can be made by measuring the intensity of the atomic hydrogen spectral lines in the visible, of which the 6563 Å line is by far the strongest. From the intensity of the Balmer α line, it is calculated that quantum beats can be observed, though only marginally. An extremely difficult method for estimating the 2p density would be to know the processes taking place in the discharge, then to calculate the density from the collision cross sections\textsuperscript{24}.

B. Observation of Quantum Beats

Before the observation of quantum beats in hydrogen was attempted, we observed beats in cesium. The experiment was similar to that done by Haroche, Paisner, and Schawlow\textsuperscript{7} except that our laser bandwidth was wider than the hyperfine separation in the cesium ground state. Thus, the quantum beats in our experiment were a superposition of modulations at five different frequencies, instead of two sets of data each with modulations at three different frequencies. The measured frequencies were the same as those obtained by Haroche et. al., though much less accurate. The other major difference was that the cesium was present in an atomic beam rather than in a gas cell.

4102 A radiation is emitted constantly by the hydrogen discharge and provides a background above which additional
fluorescence should be observed after the laser pulse. This additional fluorescence was not observed in this experiment; therefore, no attempt could be made to see quantum beats. Many attempts were made to increase the signal to noise ratio. The discharge conditions - the flow rate, the power into the discharge, and the mixture of hydrogen to water vapor - were varied. Also, the interaction region, where the laser passes through the discharge, was imaged onto the PMT using a lens and a slit. Problems with radiofrequency noise pick-up in the electronics were solved by shielding the rf discharge circuit.

The fact that quantum beats were not observed does not imply that the experiment is impossible; but it is more difficult than originally anticipated. Quantum beats have so far been observed only in a few ideal systems where the upper state can be populated cleanly; that is, the excited state is populated only by the incident excitation pulse (i.e., not by a discharge). It was hoped that this experiment would show that observation of quantum beats can be done where there is background fluorescence at the same frequency.
APPENDIX - CALCULATION OF QUANTUM BEATS IN HYDROGEN

Consider that all the atoms of interest are located with equal probability in any of the states of $2p_{3/2}$. To calculate the percentage modulation of the exponential decay, the expression for $R(\hat{\mathbf{r}}, \hat{\mathbf{g}}, t)$ given in Chapter I must be evaluated. The matrix elements are given in Table 2\textsuperscript{25}. The matrix elements with a negative $M_J'$ are not given, but are identical to the matrix elements with positive $M_J'$; therefore, their omission does not affect the answer for the percentage of the signal which is modulated. Several transitions which decrease the percentage modulation have not been considered in this treatment. These transitions include the $2p \rightarrow 6s$, $2p_{1/2} \rightarrow 6d_{3/2}$, and $2s \rightarrow 6p$ transitions. Also, due to the fact that the laser pulse is about four nanoseconds long, the expected modulation is only about 5%. After subtracting data with perpendicular polarization from data with parallel polarization, the percentage modulation is about 15%.
HYDROGEN MATRIX ELEMENTS

\[ \langle J' M_J' | z | J M_J \rangle \] for the \(^2D \rightarrow ^2P_{3/2}\) transition \((J=\frac{3}{2})\)

\[ \begin{array}{cccc}
  M_{J'} & M_J & J'=3/2 & J'=5/2 \\
  3/2 & 3/2 & -\sqrt{3/5} & 2\sqrt{3/5} \\
  1/2 & 1/2 & -\sqrt{1/15} & 3\sqrt{1/5} \\
\end{array} \]

\[ \langle J' M_J' | y | J M_J \rangle \] for the \(^2D \rightarrow ^2P_{3/2}\) transition \((J=3/2)\)

\[ \begin{array}{cccc}
  M_{J'} & M_J & J'=3/2 & J'=5/2 \\
  5/2 & 3/2 & 0 & -\sqrt{3} \\
  3/2 & 1/2 & -\sqrt{1/3} & -3\sqrt{1/3} \\
  1/2 & -1/2 & -2\sqrt{1/5} & -3\sqrt{1/10} \\
  1/2 & 3/2 & -\sqrt{1/3} & \sqrt{1/10} \\
\end{array} \]

\[ R(z,z,t) = (-\sqrt{3/5})^4 + (-\sqrt{1/15})^4 + (2\sqrt{3/5})^4 + (3\sqrt{2/5})^4 \\
+ (-\sqrt{3/5})^2(2\sqrt{3/5})^2 + (-\sqrt{1/15})^2(3\sqrt{2/5})^2 \cos \omega t \]
\[ = 76 + 6\frac{18}{25} \cos t \]

\[ R(z,y,t) = (-\sqrt{3/5})^2(-\sqrt{1/15})^2 + (2\sqrt{3/5})^2(-3\sqrt{1/10})^2 + (-\sqrt{1/15})^2(-1/15)^2 \\
+ (3\sqrt{2/5})^2(-3\sqrt{1/10})^2 + (-\sqrt{1/15})^2(-1/15)^2 + (3\sqrt{2/5})^2(\sqrt{1/10})^2 \\
+ (-3\sqrt{3}/5)(2\sqrt{3/5})(-\sqrt{1/5})(-\sqrt{1/10}) + (-3\sqrt{1/10})(3\sqrt{2/5}) \\
-2\sqrt{1/15}(-\sqrt{1/15}) + (\sqrt{3/10})(3\sqrt{2/5})(-\sqrt{1/5})(-\sqrt{1/15}) \cos \omega t \\
= 35 - 3\frac{9}{25} \cos \omega t \]

\[ R(z,z,t) - R(z,y,t) = 41 + 10\frac{2}{25} \cos \omega t \] (25% modulation)

Table 2
FOOTNOTES


11. G. Breit, Revs. Mod. Phys. 5, 91 (1933)


