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THE PERIODIC VARIATION OF LIGHT EMITTED FROM A RADIO FREQUENCY EXCITED SPECTROSCOPIC SOURCE

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THE PERIODIC VARIATION OF LIGHT EMITTED FROM A RADIO FREQUENCY EXCITED SPECTROSCOPIC SOURCE

by

S.M. Hamberger

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ABSTRACT

A very simple light source is described which is capable of emitting spectral lines whose intensities can be modulated up to 100 per cent at frequencies of the order of 10^7c/s. Experimental results are given which show the waveform of the light intensity emitted from a hydrogen source for a range of frequency and gas pressure. These results are explained by the fluctuations of the mean energy of electrons, the theory agreeing well with experiment for specific examples.

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CONTENTS

1. INTRODUCTION

1. The advantage of intensity modulated light sources for use in photometers, absorption spectrophotometers, etc., have been known for many years: in particular, a.c. coupled, tuned amplifiers can be used, with their inherent advantages over d.c. coupled amplifiers, to amplify detected signals. In these applications modulation frequencies have been low, typically 10-100 c/s, obtained either by a mechanical shutter or a discharge lamp operated by a.c. mains (e.g. Florida and Davey, 1953). Where high frequency modulation has been necessary, as in determinations of the velocity of light by modern versions of Fizeau's method (e.g. Bergstrand, 1950), or in some recent time resolved light absorption studies (Penner, 1961), the usual practice has been to modulate the beam from a steady source with a Kerr cell shutter by a high voltage r.f. supply from a valve oscillator. The principal disadvantages of this method are its inefficiency, (in practice the peak transmission does not exceed about 18 per cent of the source intensity (Zarem and Marshall, 1950)), and its inability to transmit certain wavelengths outside the visible region, and its elaboration.

2. The source to be described was developed for a particular experiment (Hamberger, 1961) in connection with transient hydrogen discharges. It was designed to emit hydrogen atomic line spectra for absorption measurements on a pulsed high current hydrogen discharge, to estimate the density of hydrogen atoms in various excited states. Since the discharge was transient, and also emitted intense radiation at these same wavelengths, it was necessary to modulate the intensity of the beam at some suitably high frequency in order to distinguish it in phase and frequency from the light emitted by the discharge. The modulation frequency depends on the particular conditions of the experiment in which the source is used. In our experiment a frequency of 3 Mc/s was considered suitable, and the principal requirements of the source were as follows:-

(a) it should emit strongly atomic hydrogen lines which had profiles determined by Doppler broadening only and with negligible self-absorption;

(b) the intensity of the lines should be modulated so that it had a maximum component at 3 Mc/s;

(c) the Doppler profiles could be easily controlled by varying the working temperature of the lamp over a wide range;

(d) it should be simple to make and easy to operate.

3. In this paper it is shown that these requirements may be fulfilled using an electrodeless high frequency discharge in a low pressure gas, in this case hydrogen or a mixture of hydrogen and helium. Although it is known that a high frequency discharge may emit a fluctuating light, (Frey, 1936), there appears hitherto to have been no systematic study of the conditions which govern the variation of light emitted from a discharge caused by a rapidly alternating electric field. In this work the calculated variation in mean electron energy is shown to account for the observed variation in emission.

2. DESCRIPTION OF THE SOURCE

4. The excitation of the atomic lines of hydrogen in a discharge has been discussed by many early workers, in particular by Wood, (1922). They have found that the atomic lines are best excited (in preference to the molecular spectrum) in the absence of metal or rough insulating surfaces, which are well known to act as catalysts aiding the recombination of atomic hydrogen. Hence it was decided to use an electrodeless discharge in a dielectric container.

5. The construction of the source is shown in Fig. 1. The vessel is filled with hydrogen, or a mixture of hydrogen and helium (see section 4) and an electrodeless discharge produced by an alternating electric potential applied between the two external silver coatings. The discharge is constricted into the central capillary tube of approximately 3 mm bore and 0.5 mm wall thickness, to enhance intensity and give an optically thin line source which is convenient for focussing on to a slit of a spectroscope. The thin wall enables the gas temperature to be controlled by cooling the outside of the capillary.

6. The first tubes, which were made of borosilicate glass, failed when local overheating by recombining atomic hydrogen softened the glass. The use of fused silica prevented this. A silica to glass graded seal is used for pumping.

7. To overcome the problems due to the removal of hydrogen to the walls during the discharge a titanium hydride replenisher is incorporated. This is of a type used in a large hydrogen thyratron* to control the operating pressure. A constriction between the discharge region and the replenisher chamber serves to prevent the discharge from reaching the replenisher and damaging it by sputtering.

8. It has been found that a better performance is obtained when a gas mixture of helium and hydrogen is used (see section 3). For this, spectroscopically pure helium is admitted at the required pressure just prior to sealing off.

9. The exciting power is obtained from a conventional 300 watt radio frequency transmitter, Type T1509. The matching transformer shown in the circuit of Fig. 2 enables a variable high voltage up to about 1000 V to be obtained conveniently near the source (minimising radiation from leads), and remote from the transmitter, to which it is coupled by a low impedance cable. The total power taken from the generator is normally less than 100 watts.

10. The temperature of the capillary tube is controlled by varying the convective cooling around it. By restricting it, wall temperatures of up to 750 ^{o}K are obtained; by cooling with cold nitrogen (from a Dewar of boiling liquid nitrogen) the wall temperature can be reduced to about 100 ^{o}K.

11. For processing, the device is sealed to a mercury pump and outgassed by baking at 500 ^{o}C. The replenisher is outgassed (using its heater winding), and the system filled with pure hydrogen through a palladium leak to a pressure of several torr with the replenisher hot. When the titanium pellets in the replenisher are allowed to cool, the pressure falls until all the hydrogen present is absorbed by the titanium. The heater current is then varied and the equilibrium pressure in the system found as a function of current (see Fig. 3). The replenisher is finally allowed to cool until the system is evacuated, and the lamp is sealed off.

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* G.E.C. Type CV 2418. The replenishers were supplied by M.O. Valve Co., Hammersmith.

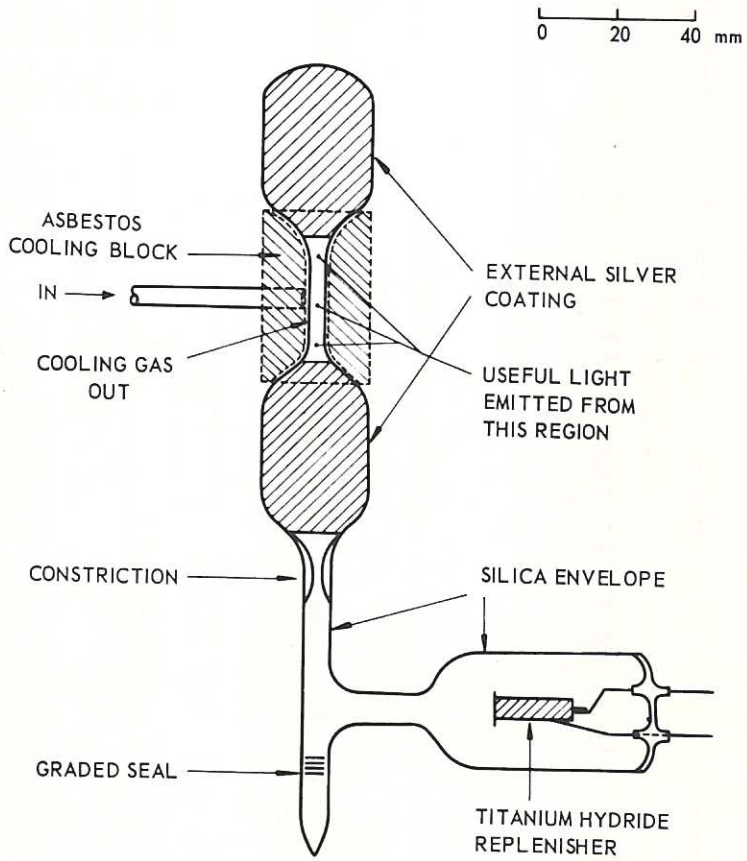


Fig. 1 Sketch of the Source

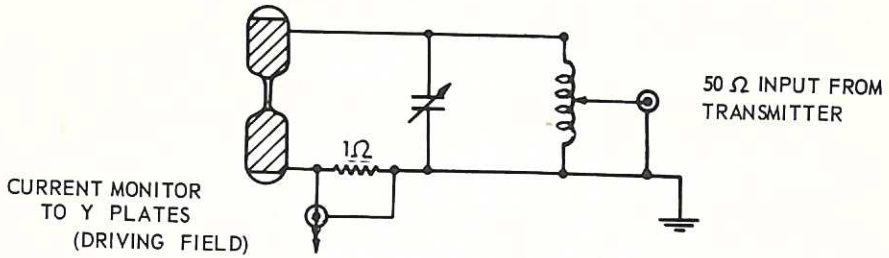


Fig. 2 Arrangement of Driving Circuit

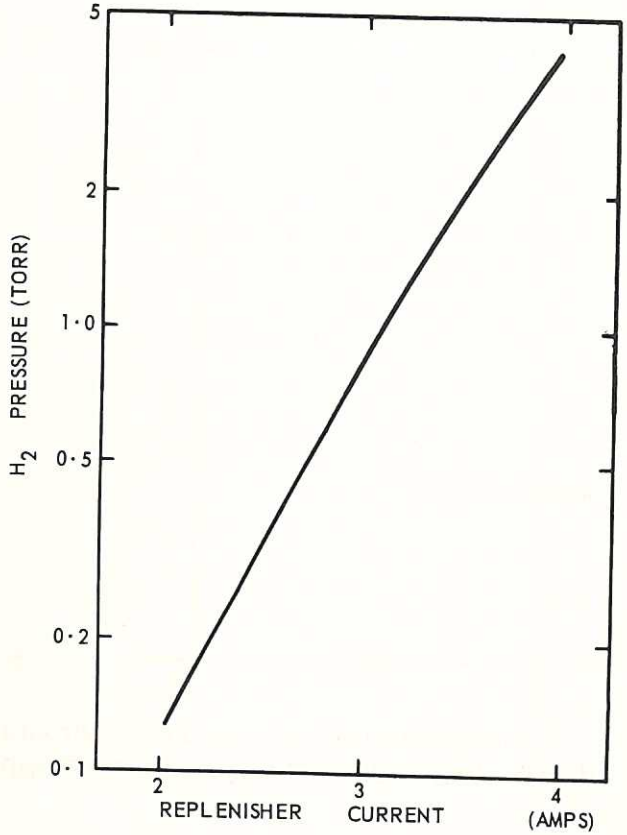


Fig. 3 Typical Replenisher Calibration

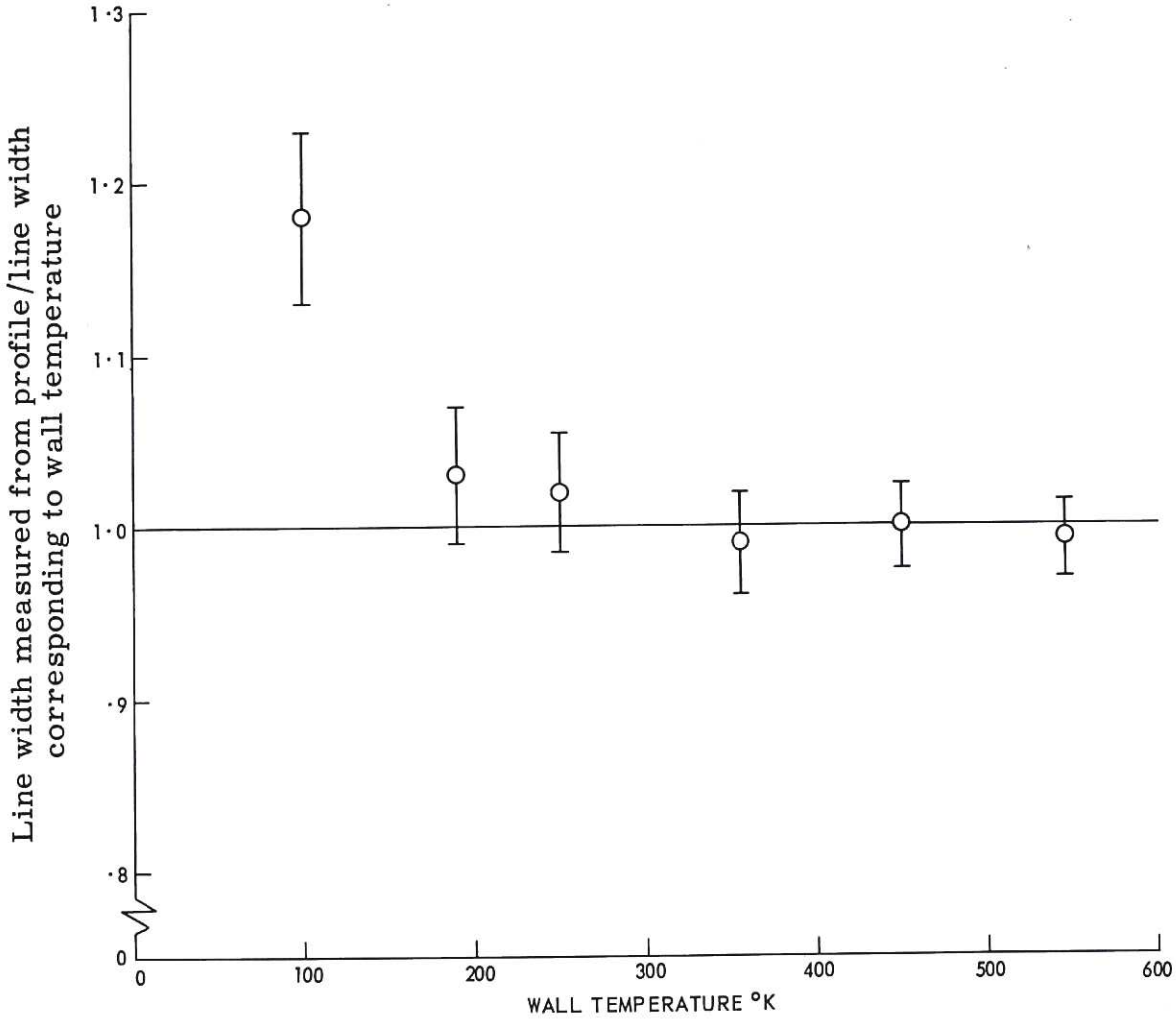
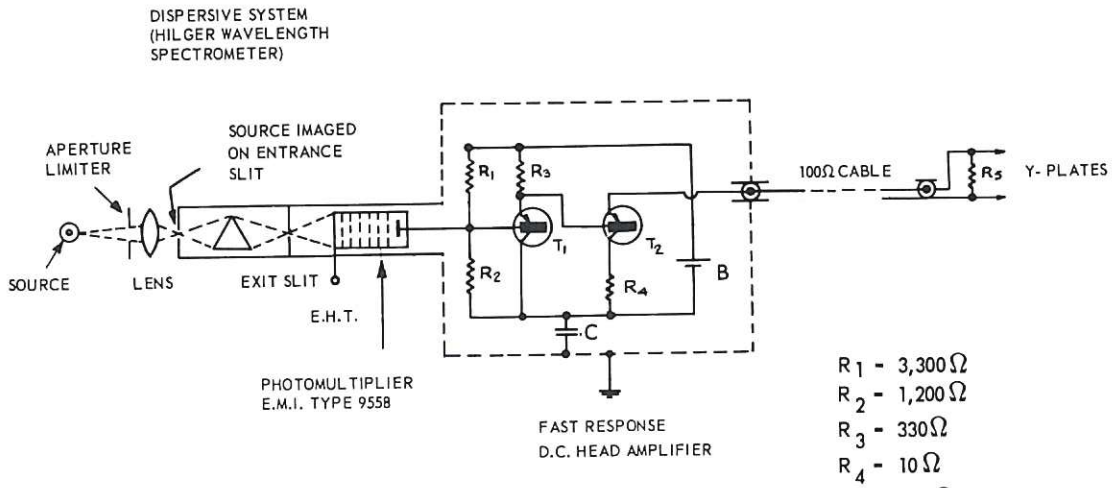


Fig. 4 Comparison of measured half-width of HeI Line 5016A with capillary outside wall temperature



R_1 - 3,300 Ω
R_2 - 1,200 Ω
R_3 - 330 Ω
R_4 - 10 Ω
R_5 - 100 Ω
C - 1000 pF
B - 1.35v CELL
T_1 - T_2 - 2N499

Fig. 5 Arrangement for observing fast modulation of spectral line intensity

3. PERFORMANCE

Spectral emission

12. The source, when filled with pure hydrogen, at first emits a bright Balmer spectrum, with the molecular lines only weakly excited. After running for some tens to hundreds of hours, the Balmer line intensity decreases considerably with a corresponding increase both in the intensity of the molecular spectrum and in the power dissipated in the tube. The resultant increased running temperature of the lamp also leads to the formation of a brown deposit on the inside of the capillary, which may be reduced silicon (Thonemann, 1953).

13. Clearly the recombination of atomic hydrogen at the walls has been enhanced. Reference to early work (Wood, 1922 and Series, 1957) on the subject indicates that a trace of water vapour is necessary, even on a glazed insulating surface, to prevent the catalytic action of the surface. It is possible that the continued operation of the lamp together with the gettering action of hot titanium removes the water vapour. Further, the deposits on the inside surface aid recombination.

14. In recent years several ways have been found to overcome this effect, including the deliberate introduction of other gases (e.g. water vapour) into the discharge. In particular, Series (1951, 1957) found that he could best excite the Balmer lines in a mixture of hydrogen and helium, the latter predominating; the main lines emitted were due to H and HeI, the H_2 spectrum being very weak. A similar mixture was therefore tried in the lamp; it was found that with a filling of 3 torr He and 0.1 to 0.2 torr H_2, the source emitted very strong Balmer lines, together with the HeI spectrum. No. HeII lines were detectable, and the molecular spectrum was very weak. In addition the running voltage and temperature of the source were greatly reduced.

15. The effect of wall temperature is interesting: it is found that the Balmer line intensity increases as the temperature is decreased. This effect is shown in Fig. 9, which shows the intensity of H_α increasing considerably when the temperature is reduced from about 700 ^oK to 200 ^oK. This effect is not satisfactorily explained. It is known that reduction of the wall temperature generally encourages the recombination of atomic hydrogen and so should reduce the atomic line emission. A possible explanation is that helium adsorbed on the surface effectively poisons its catalytic action; at lower temperatures more helium is adsorbed, enhancing this effect.

16. The gas temperature, which is needed for calculations involving the hydrogen line profile (Hamberger, 1961), has been measured from the profiles of the helium singlet line 5016Å resolved by a Fabry-Perot etalon. The Balmer lines themselves have fine structure separations of the order of a Doppler width and so are not suitable for direct measurement. The assumption that the two types of molecules are at the same temperature is justified by a simple calculation which shows that in our lamp a hydrogen atom makes about 20 or 30 collisions with a helium atom (reducing its energy difference by 2/5 each time) for every collision with another hydrogen atom; and about 50 collisions in all before striking the wall. Further, it may be easily shown that the r.f. currents flow uniformly through the gas at the frequencies considered (spin depth $\gg$ diameter) and consequently the heating is uniform.

17. To check that the gas is, in fact, in good thermal contact with the wall, the Doppler widths were compared with those calculated on the assumption that the gas has the same temperature as the outside of the capillary wall, which

was measured by a thermocouple. This is shown in Fig. 4. The temperature drop in the wall was calculated to be less than 10 deg C - the radial heat flow through the capillary wall was measured calorimetrically and found to be about 5 watts. It is seen from Fig. 4 that the agreement is within the accuracy of measurement at temperatures above 200 ^{o}K, while at lower temperatures the gas was hotter than was indicated by the thermocouple. This may be due to the method adopted of force-cooling only the capillary section of the lamp, which results in large axial temperature gradients between the capillary walls and the hot bulbs at the ends.

18. No measurements have been made of the absolute intensity of the source, which clearly depends on the operation. From the discharge conditions it is estimated that $\sim 10^{15} - 10^{16}$ Hα photons per second are emitted per steradian for each centimeter of capillary, corresponding to a flux of about 10 mW cm^{-2} at the surface.

Modulation

19. The time variation in the emission of a specified line was observed using a small glass prism monochromator fitted with a photomultiplier behind the exit slit: this was connected to a head amplifier designed to give a uniform response from d.c. to at least 10 Mc/s (see Fig. 5). The high frequency response of the photomultiplier system (including the oscilloscope) was checked by observing the effect of a very short light pulse ($\sim 10^{-8}$ sec), from a fast scintillator: the rise time was not more than 2.10^{-8} seconds corresponding to an upper frequency cut-off of approximately 15 Mc/s.

20. Typical results for a lamp filled with pure hydrogen are shown in Figs. 6 - 8, in which the upper trace shows the observed intensity of the Hα line (increasing downwards) with a superimposed trace corresponding to zero intensity (obtained by closing a shutter). The lower trace is proportional to the current in the discharge tube (Fig. 2).

21. The pressure of hydrogen was varied by controlling the heater current in the replenisher, using the calibration obtained during processing. A typical calibration is shown in Fig. 3.

22. From the oscillograms in Figs. 6 - 8 it is clear that the emission is, in general, markedly non-sinusoidal i.e. it does not follow the curve of power input. The pulses of light become shorter, at a given frequency, as the pressure increases; and, at a given pressure, as the frequency decreases. These observations are in good agreement with the emission waveforms which are calculated in the next section.

23. When the source is intended for use with a receiver tuned to the fundamental frequency of light modulation (i.e. twice the driving frequency) conditions are chosen to maximise the component of the light waveform at this frequency. For example, a visual Fourier analysis of the Hα traces in Fig. 6 shows that a pressure of 2 torr gives a purer 3 Mc/s component than the others shown.

24. The modulation of Hα light at 3 Mc/s from a source filled with a He-H_2 mixture is shown in Fig. 9, for different temperatures. The peak intensity of the light increases by about ten times in cooling from 700 ^{o}K to 170 ^{o}K, but the modulation is not complete at low temperatures. A direct measurement of the 3 Mc/s component of the emitted light shows that the increase in useful Hα intensity is about twenty times.

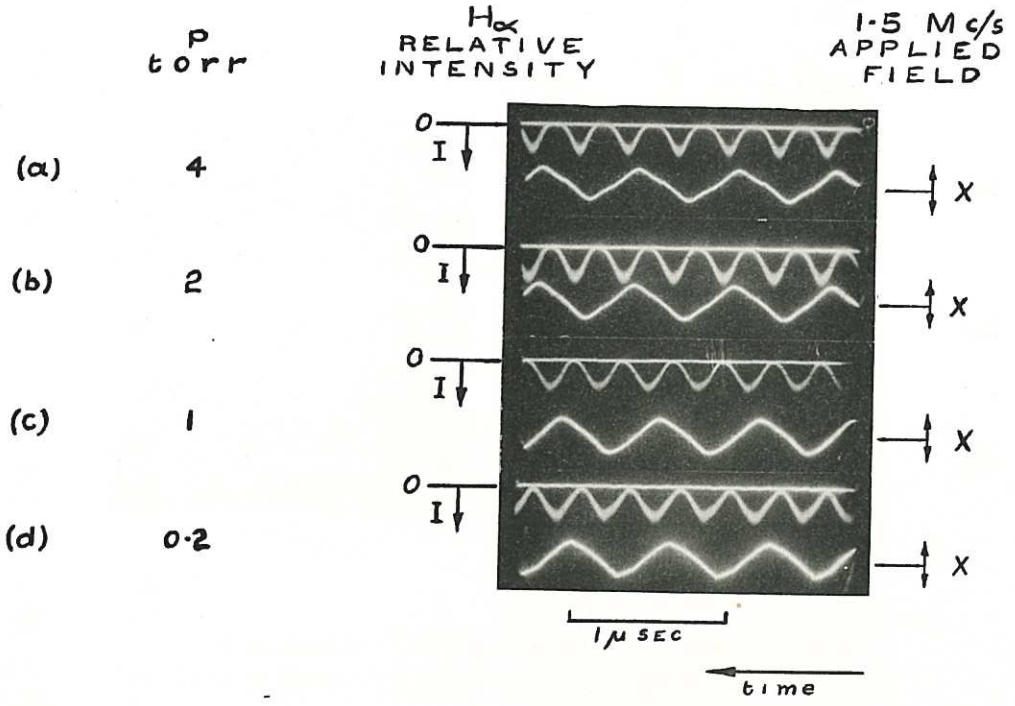


Fig. 6. Emission of H_α from a 1.5 Mc/s discharge in H_2 at various pressures (light modulated at 3Mc/s)

NOTE. In Figs. 6 - 8 the intensity scales are arbitary and should not be used to compare the emission intensity under different conditions.

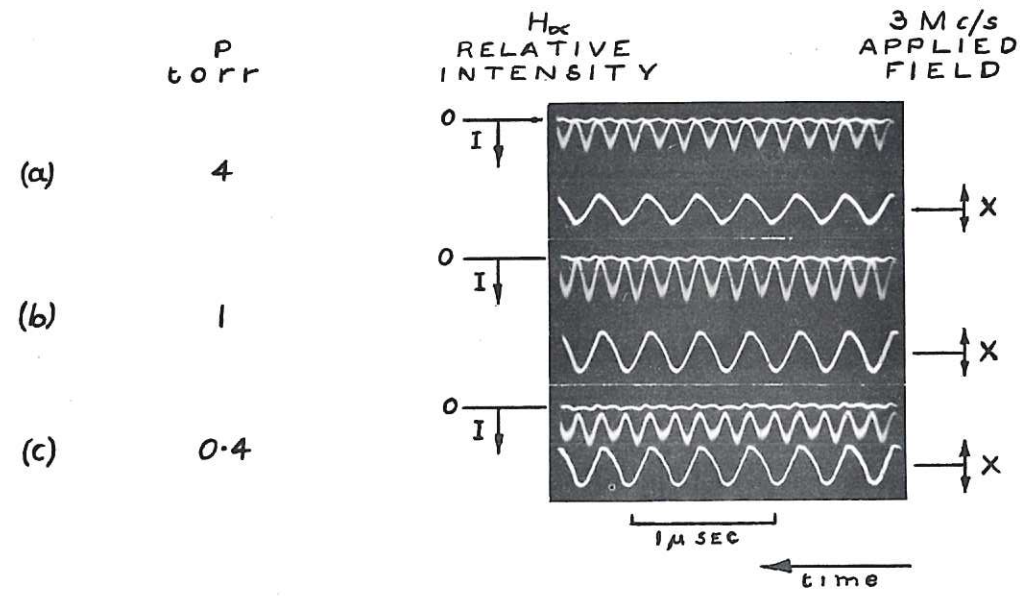


Fig. 7. Emission of H_α from a 3 Mc/s discharge in H_2 at various pressures (light modulated at 6 Mc/s)

The upper trace in each case shows the variation in light intensity: the lower is proportional to the driving current

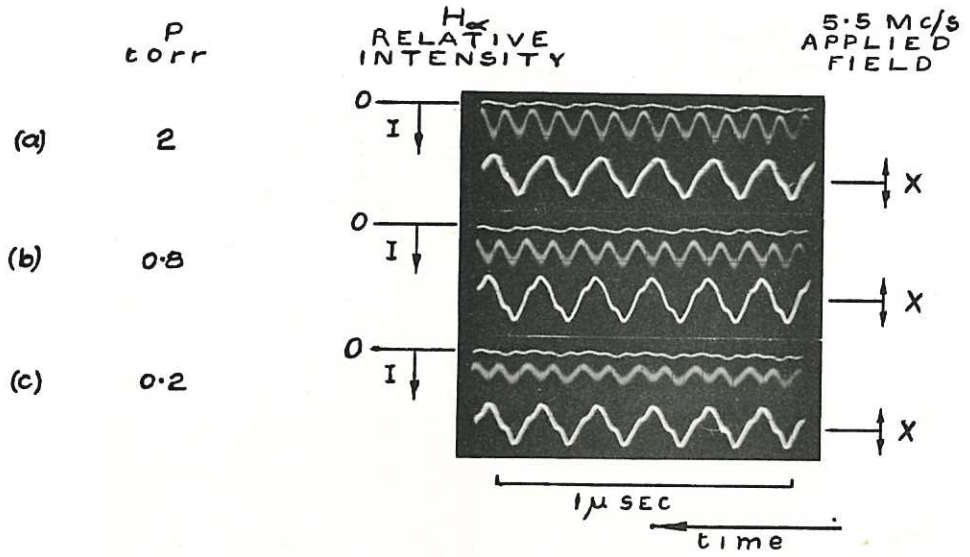


Fig. 8.. Emission of H_α from a 5.5 Mc/s discharge in H_2 at various pressures (light modulated at 11 Mc/s)

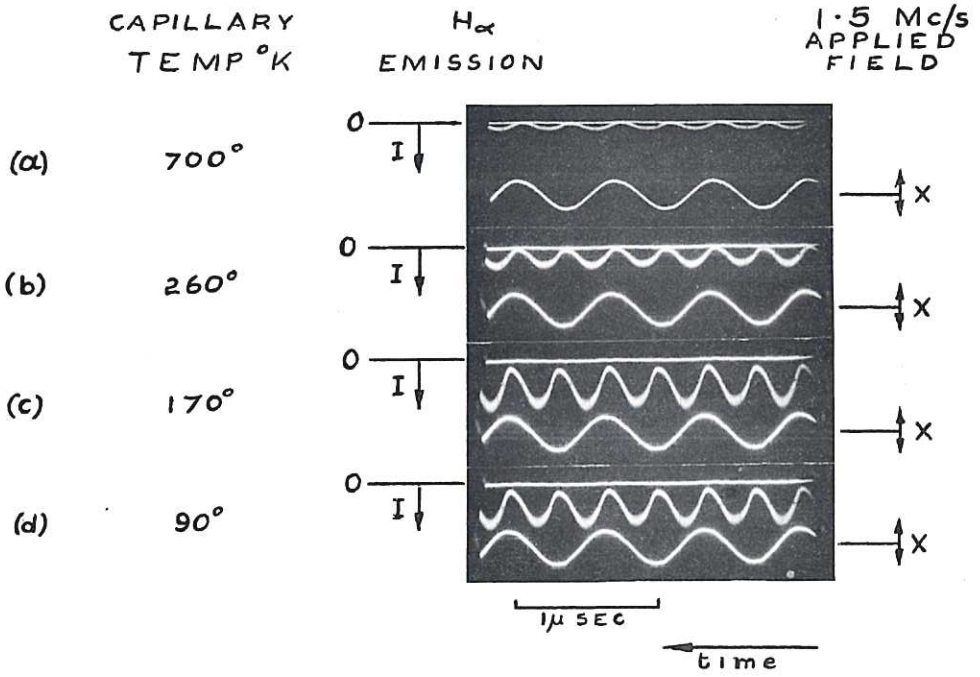


Fig. 9. Variation in H_α emission from a 1.5 Mc/s discharge in $H_e - H_2$ mixtures with wall temperature (intensity scale is same for all traces).

4. EXPLANATION OF THE MODULATION OF THE EMISSION

25. The rate of excitation of atoms by electron impact in a low pressure discharge is proportional to the electron density, the neutral atom density, and to an excitation function which increases with the mean electron energy. Table I shows some typical discharge conditions. At these pressures the deionization is governed by ambipolar diffusion (von Engel, 1955). The electron density therefore, decays with a time constant given by $\tau_{el} = \Lambda^2/Da$ where Λ is a characteristic diffusion length, and D_a the ambipolar diffusion coefficient. Even at the lowest pressures this time is of the order of 10^{-5} seconds, compared with the emission decay time $\sim 10^{-7}$ seconds seen from the oscillograms. It follows that variations in electron density cannot account for the modulation. Similarly, the density of neutral atoms cannot vary significantly, firstly since the rate of recombination to molecules is slow (the estimated life time of an atom before recombination in the capillary is between 10^{-1} and 10^{-3} seconds (Goodyear and von Engel, 1961); while the fraction lost by ionizing collisions with electrons is not important at the low degree of ionization attained ($N_e/N_o \triangleright 10^{-4}$).

Variation of mean electron energy in the alternating field

26. On the other hand, under certain conditions the mean electron energy may vary appreciably with the periodic field; this has been studied by Townsend (1932) and by Harries and von Engel (1954). Electrons will gain energy from the field and lose energy by collisions (both elastic and inelastic) with heavy particles, the rate depending on the frequency and particular kind of collision. The electron distribution will itself depend on the rate at which electrons exchange energy between themselves compared with that at which they impart energy to the heavy particles. If the former mechanism is sufficiently rapid the distribution will be Maxwellian (see para. 37 et seq.).

27. Suppose electrons make ν collisions per second with heavy particles and lose on average a fraction $\varkappa$ of their energy in each collision, then the decay of mean electron energy ε is

$$\frac{d\varepsilon}{dt} = - \varkappa\nu\,\varepsilon \qquad (1)$$

i.e. the decay is characterised by a time $\tau = {}^1/\varkappa\nu$ (which, in general, varies with ε). If τ is very small compared with a half-period of the field the mean energy will closely follow the variation of the field, as it would in d.c. discharge which is slowly varied, but without change in electron density because of the long diffusion decay constant.

28. A suitable equation can be written down (Francis, 1960) to describe the energy balance of a 'mean' electron in an electric field $X_o \sin \omega t$. Provided an electron makes many collisions in one half cycle of the field ($\omega \ll \nu$) the average energy $\delta\varepsilon$ gained in time δt in which electrons drift a distance δx in the direction of the field is

$$\delta\varepsilon = eX_o \sin \omega t . \delta x - \varkappa\nu\varepsilon . \delta t \qquad (2)$$

The drift velocity in the field may be expressed as

$$\frac{dx}{dt} = \frac{bX_o}{p} \sin \omega t \qquad (3)$$

where b is the electron mobility at a pressure of 1 torr and p is the pressure in torr. These results lead to

$$\frac{d\varepsilon}{dt} + \varkappa\nu\varepsilon = \frac{ebX_o^2}{p} \sin^2 \omega t \qquad (4)$$

which will apply to our discharge, since for hydrogen at p = 1, $\nu \approx 10^{10}$ sec^{-1}, compared with $\omega \approx 10^7$ sec^{-1}.

TABLE I

Typical Discharge Conditions in the Capillary Region

			Text Reference Para.	Notes
Pressure p (torr)	4	0.2		
Atom density N_o (cm^{-3})	2.10^{17}	10^{16}		1
Electron density N_e (cm^{-3})	5.10^{12}	10^{12}	4.2	2
Electron energy ε (eV)	0-2.6	3-9.2	4.1	6
Reduced electric field (peak) X_o/p ($Vcm^{-1}torr^{-1}$)	14	80		3
Current density J_o (peak) (Acm^{-2})	4	4		3
Time constant for decay of ε (sec)	10^{-8}	2.10^{-8}	4.1,	4
Time constant for decay of N_e (sec)	$\sim 10^{-4}$	$\sim 10^{-5}$	4	5
Quarter period of modulation (sec)	8.10^{-8}	8.10^{-8}		
Frequency of applied field $\frac{\omega}{2\pi}$ (sec^{-1})	1.5×10^6	1.5×10^6		
Depth of modulation ($^{max}/_{min}$) per cent	100	95	3.2,5	
Relevant diagrams:	Figs.6(a) & 14(a)	6(d) & 14(b)		

Notes 1. assumed $\sim {}^{2}/_{3}$ dissociated

2. estimated from J_o and drift velocity $^{X_o}/_{p}$.
3. Measured with valve voltmeter
4. Increase of energy loss rate $k\nu$, Figure 11, taken at mean value of ε
5. Based on ambipolar diffusion time constant = Λ^2/D_a
6. Calculated from energy equation.

29. In this form it will be possible to use experimentally measured values of $\varkappa$, ν, b. Since the discharge (in pure hydrogen) occurs in a mixture of molecules and atomic gas of uncertain composition it is not simple to derive from known cross-sections meaningful values of the quantities $\varkappa,\nu,b$ which depend critically on the energy range assumed for the electrons, the degree of dissociation and the energy distribution. Inelastic collisions with hydrogen molecules become very important at quite low energies (0.1 eV)(Frost and Phelps, 1961), while at about 2 eV the main energy loss is due to excitation of atoms to the 2P state; the various processes are discussed in papers on r.f. ion sources (e.g. Thonemann, 1953; Goodyear and von Engel, 1961).

30. A simple two-stage approach suffices for our problem. We shall first assume that only atomic hydrogen is present, and that for most of the time the electron energy is below 2 eV, so that elastic collisions dominate and the damping coefficient $\varkappa\nu$ is at its least. This reduces equation (4) to a simple linear form which may be solved easily.

31. The result shows that the energy follows the field quite closely except at very low pressures (within the frequency range used in the experiment): it follows that, if other energy losses are considered the electron energy will be even more closely in step with the field, so that we are justified in using at all times values of $\varkappa,\nu,b$ and ε as functions of X/p measured in H_2 discharges under steady conditions. This leads to a less convenient (non-linear) equation from which more limited information can be extracted.

Elastic collisions with atoms only (hydrogen fully dissociated)

32. The fraction $\varkappa$ becomes simply $^{2m}/_{M} \approx \frac{1}{900}$ and the collision frequency ν is $N_o \overline{\sigma_o v}$ where σ_o is the elastic scattering cross-section, v the random electron velocity, and N_o the density of neutral atoms $\approx 7.10^{16} p$ cm^{-3}. Experimental values (Fite, 1958) of σ_o show that in the range 0.5 to 7 eV, $\sigma_o V \approx$ constant = $1.3 \times 10^{-7} \pm 12\%$ cm^3/sec. Thus $\varkappa\nu$ is a constant $\approx 10^7$ p sec^{-1} and the solution to equation (4) is

$$\varepsilon = \frac{ebX_o^2}{2p\varkappa\nu}\left\{1 - \cos\phi\cos(2\omega t - \phi)\right\}, \quad \tan\phi = \frac{2\omega}{\varkappa\nu} \qquad (5)$$

The first term on the r.h.s. represents the mean energy which the electron would gain in a d.c. field equal to the r.m.s. value of the a.c. field, and will be denoted by ε_o. When the frequency is low, i.e. $\omega \ll \varkappa\nu$, the last equation becomes

$$\varepsilon = \varepsilon_o (1 \cos 2\omega t) \qquad (6)$$

i.e. the energy varies sinusoidally between 0 and $2\varepsilon_o$ at twice the driving frequency. At the other extreme of high frequency, i.e. $\omega \gg \varkappa\nu$ we have

$$\varepsilon = \varepsilon_o \left(1 - \frac{\varkappa\nu}{2\omega} \sin 2\omega t\right) \qquad (7)$$

and the energy again varies sinusoidally at 2ω about the same average ε_o, but with a reduced amplitude, $\varepsilon_o \frac{\varkappa\nu}{2\omega}$, and a phase change of $\pi/2$. At p = 2 torr, $\varkappa\nu \approx 2.10^7$ sec^{-1} so that for a frequency of 1.5 Mc/s, $\frac{\varkappa\nu}{2\omega} \approx 1$. These results are shown diagramatically in Fig. 10.

General hydrogen discharge (not fully dissociated)

33. Having shown above that at pressures of a few torr and a frequency of a few Mc/s the damping due to elastic collisions alone ($\varkappa \sim 10^{-3}$) is sufficient to make the electron energy roughly follow the instantaneous field, we now insert values of $\varkappa,\nu,b$ measured in d.c. discharges (Townsend, 1947). Fig. 11 gives values of $\varkappa,\nu,\varepsilon$ and the product $\varkappa\nu$ for X/p in the range 0 to 50 V/cm/torr.

In the range $0 < X/p < 15$, $\kappa\nu$ varies linearly with mean energy to a very close approximation

$$\frac{\kappa\nu}{p} = 10^7\,\varepsilon, \text{ where } \varepsilon \text{ is in eV;}$$

but at larger X/p, $\kappa\nu$ rises much more steeply. Putting this in equation (4) we obtain

$$\frac{d\varepsilon}{dt} + 10^7\, p\varepsilon^2 = \frac{ebX_o^2}{p}\sin^2\omega t \qquad (8)$$

which is of the form

$$\frac{d\varepsilon}{dz} + A\varepsilon^2 = B\sin^2 z \qquad (9)$$

where

$$A = 10^7\, p/\omega$$

$$B = ebX_o^2/p\omega$$

The solutions of (9) may be expressed in terms of Mathieu functions (Harries and von Engel, 1954; McLachlan, 1947). Equation (9) has positive continuous solutions in which ε has a period of π and a r.m.s. value of $\sqrt{B/2A}$. Further, when $(AB) > 16$, ε varies between a small value and $\sqrt{B/A}$.

34. Suppose we take as an example the case of Fig. 6(a). i.e. $p = 4$ torr, $f = 1.5$ Mc/s ($\omega \approx 10^7$) for which we have measured the electric field in the capillary tube and found X_o to be 56 V/cm. Using measured values of mobility (Bradbury and Nielson, 1936, 1937), we find the quantities, $B = 28$ eV/cycle, $A = 4$ in our units. As $AB = 112$ and > 16, so that the condition for large variation in ε is satisfied. The maximum energy is then $\sqrt{B/A} = \sqrt{7} = 2.6$ eV.

35. The energy equation (9) has also been solved numerically for the above conditions: the solution is shown graphically in Fig. 12(a). Shown for comparison is the solution of the linear equation normalized to the same peak energy. For $X/p > 15$ the losses rise sharply with mean energy, and a better approximation is found to be

$$\kappa\nu/p = 3.23 \times 10^7\,(e^{\cdot 31\varepsilon} - 1) \qquad (10)$$

This is used to explain the modulation of Figure 6(d), where the experimental quantities are:-

$$p = 0.2 \text{ torr}$$

$$f = 1.5 \text{ Mc/s}$$

$$X_o/p = 80 \text{ V/cm/torr.}$$

The energy equation (4) takes the form

$$\frac{d\varepsilon}{dz} + P(e^{0.31\varepsilon} - 1) = B\sin^2 z \qquad (11)$$

where

$$\left.\begin{aligned} P &= 3.23 \times 10^7\, p/\omega = 0.67) \\ B &= eX_o \cdot \frac{bX_o}{p}\,\frac{1}{\omega} = 100 \end{aligned}\right\} \qquad (12)$$

36. The solution of (11) was obtained on the 'Mercury' computor and is shown in Fig.12(b). It will be seen that the mean electron energy ε oscillates between the values 9.2 eV and 3.0 eV, with a slight phase lag in the field (cf. Fig. 10, case $\kappa\nu \ll \omega$), the shape being non-sinusoidal and non-symmetrical about the maxima.

The energy distribution

37. We need now to compare the rate at which electrons lose energy to heavier particles with the rate at which they exchange it amongst themselves. If these are comparable the distribution will certainly be Maxwellian. However,

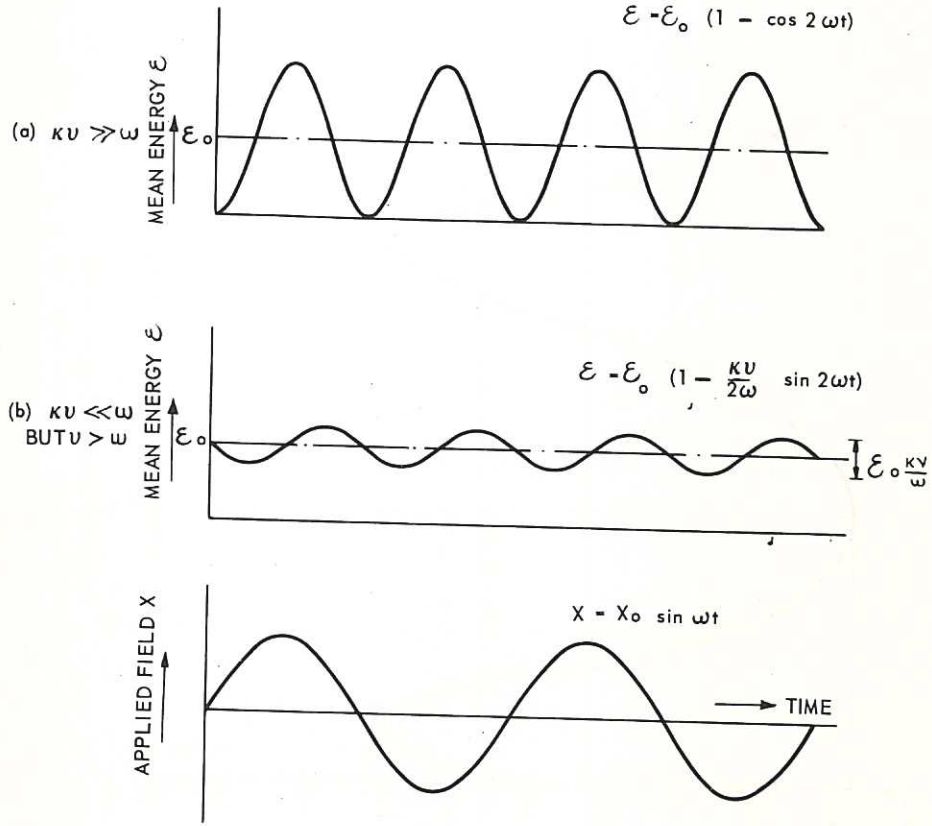


Fig. 10 Variation of mean electron energy $\mathcal{E}$ with periodic field X for
a. High damping
b. Low damping
When only elastic collisions between electrons and atoms are considered

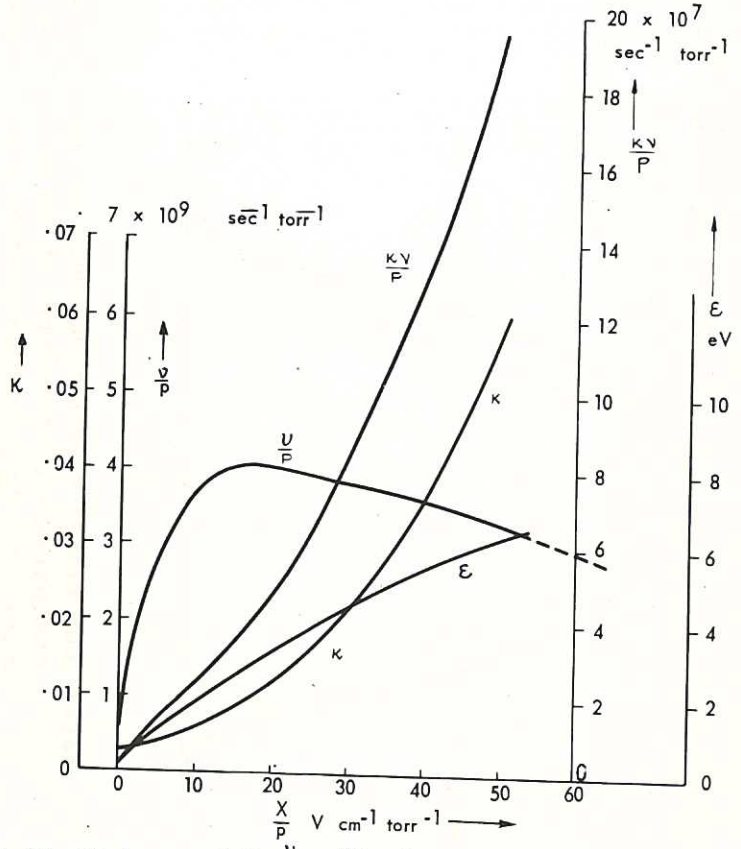


Fig. 11 Values of K $\frac{\upsilon}{p}$, $\frac{\kappa\upsilon}{p}$, $\mathcal{E}$ derived from Experimental data for H_2 (Townsend 1947)

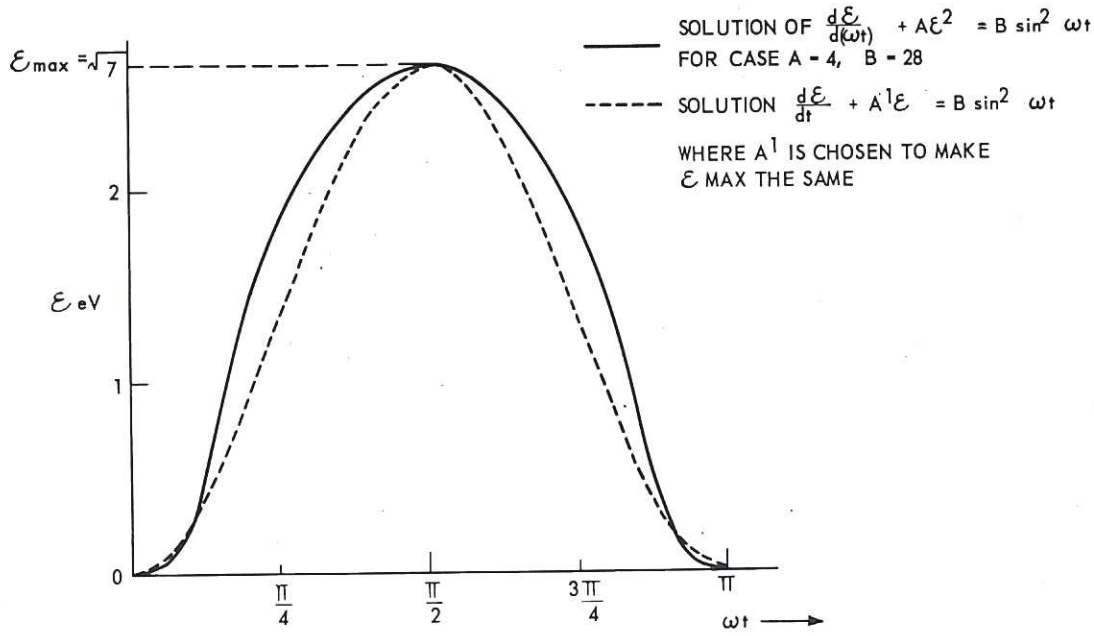


Fig. 12(a) Numerical solution of nonlinear energy Equation 9 compared with the form of the solution of the corresponding linear equation i.e. Equation 6. (Low X/p)

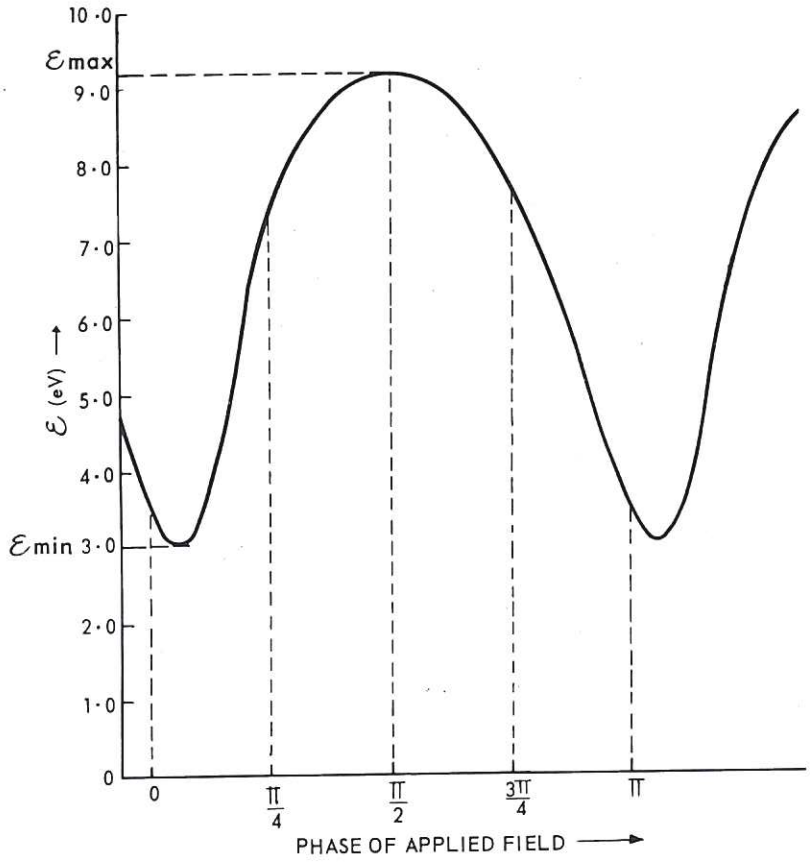


Fig. 12(b) Variation of mean electron energy at high X/p. Numerical solution of Equation 11.

collisions between electrons and heavy particles can often of themselves lead to a Maxwellian distribution, when the heavy particles have a wide range of excitation levels (von Engel, 1956) - this is so in a mixture of H and H_2. For example, the assumption of a Maxwellian distribution of electron velocities successfully accounts, at $X/p > 20$, for the ionization in hydrogen due to electron swarms of trivial densities (currents $< 10^{-8}$ amperes), in which there is virtually no interaction between the electrons (von Engel, 1956; Emeleus et al, 1936).

38. Furthermore, careful probe measurements in the positive columns of glow discharges in helium and argon (Klarfield, 1937; 1938) show that a Maxwellian electron energy distribution exists even when the electron-electron energy exchange time is one to four orders of magnitude longer than the exchange time with atoms.

39. Consider a typical discharge in the range of parameters we have used for these sources, e.g. p = 4 torr, X_o/p = 14 V/cm/torr. The peak current density in the capillary has been measured and found to be 4 A/cm^2: the drift velocity at this value of X/p is 5.10^6 cm/sec, hence the electron density, averaged over the cross section of the tube, is 5.10^{12} cm^{-3}. At this density and a mean energy of 1.7 eV (corresponding to the peak energy of 2.6 eV deduced in the previous sections) the time for electrons to reach a Maxwellian distribution by self collisions (Spitzer, 1956) is $\tau = 10^{-8}$ secs.

40. The time required for an electron to share its energy with the heavy particles can be read from Fig. 11; at the mean value $\overline{X/p}$ = 9 V/cm/torr, this is $1.2\ 10^{-8}$ secs. These two values for the energy exchange times are sufficient to justify the assumption of a Maxwellian distribution at all times in the discharge. At lower pressures (e.g. p = 0.5 torr, X/p = 50, $\varepsilon \approx 6$ eV) the electron-electron exchange time is 10 times longer than that due to collisions with neutral atoms. In view of the experimental evidence quoted above, it again appears justifiable to assume a Maxwellian distribution, which we do in the following sections.

Variation of emitted light

41. It is theoretically possible to calculate the number of hydrogen atoms in any quantum state (e.g. unexcited, excited, ionized) provided the mean electron energy changes slowly, and an unchanging form of energy distribution is assumed. Such calculations have been made (McWhirter, 1959) by considering the various populating and depopulating processes, and assigning estimated probabilities for each of them. The resultant linear differential equations can then be solved by numerical methods.

42. In our discharge the electron energy is varying rapidly with time: since the probabilities of excitation and ionization also vary strongly with mean energy, and hence with time, an attempt at the type of analysis referred to leads to a system of difficult non-linear differential equation.

43. Fortunately, in our discharges the degree of ionization is low, the electron density remains constant with time, and the dominant state is the unexcited neutral atom. Here only a few processes play any significant part in determining the populations of the lower excited states. For example the excited state of hydrogen with principal quantum number n = 3 is chiefly formed by electrons which hit atoms in the ground state, and depopulated by spontaneous decay with the emission of radiation:

$$\frac{dN_3}{dt} = N_1 N_e \overline{\sigma v} - N_3 \Sigma A_3 \qquad (13)$$

Here N_e is the electron density, N_q in general is the density of atoms in the n = q state, and A_q the probability per second that such an atom will jump spontaneously to some lower state. σ is the cross section for excitation by electron collisions, and σv, the average over the distribution of electron velocities v, gives the excitation rate.

44. If the rate of production and loss of any given state are extremely rapid compared with the rate of variation of the applied field, then an equilibrium exists between them at all instants of time. This condition is satisfied when the mean lifetime of an excited state, $1/\Sigma A_3$, and the mean time interval between an electron making two exciting collisions $\frac{1}{N_1 \overline{\sigma v}}$ are each much less than the period of the field. Excited states are produced in a pulse every half period of the field, so that in equation (13) the term $\overline{\sigma v}$ could be replaced by a Fourier series of the form $\Sigma B_n \sin 2n\omega t$. The solution shows that provided $\Sigma A_3 \gg 2\omega$, the two terms on the right hand side are much greater than dN_3/dt. The instantaneous value of N_3 is then

$$N_3 = \frac{N_1 N_e \overline{\sigma v}}{\Sigma A_3} \qquad (14)$$

The emission intensity of a given line arising from a downwards transition from n = 3 states, e.g. H_α, is

$$I \propto N_3 A_{32} = \frac{A_{32}}{\Sigma A_3} N_1 N_e \overline{\sigma v} \qquad (15)$$

Here we are interested only in the variation with time due to the factor $\overline{\sigma v}$, since the other quantities are assumed to be constants on our short time scale.

45. The variation of $\overline{\sigma v}$ with mean energy, ε, may be calculated assuming a Maxwellian energy distribution of the electrons, and using a simple form of the dependence of σ on velocity (Woolley and Stibbs, 1953). The result of such a calculation is shown in Fig.13 for the appropriate transition.

46. If we now combine this with the time variations which were predicted for ε (in para. 32 et seq.) we can explain the experimental observations shown in Figs. 6 to 8 for a pure hydrogen discharge. (No attempt has been made to analyse a mixture of H_e and H_2 but one would expect the results to be qualitatively similar to those discussed in para. 32)

Application to specific cases

47. Consider once again our two examples in which p = 4 torr, f = 1.5 Mc/s, and p = 0.2 torr, f = 1.5 Mc/s, for which we have derived the variation of mean energy shown in Figs. 12(a) and (b). Using these results together with the data of Fig. 13 we may predict the variation in H_α emission to be expected. These are shown in Figs. 14(a) and (b). For comparison the experimentally observed waveforms (taken from the oscillograms of Fig. 6(a) and (d) are shown superimposed, both being normalized to the maximum intensity and to a periodicity in the driving field of π. At the higher pressure agreement between theory and experiment is well within the accuracy of measurement on the rising current edge. The slightly worse agreement on the falling current edge is probably due to the asymmetry of the driving field (see lower trace of Fig. 6(a). In the theory the field is assumed to be sinusoidal.) In the lower pressure example shown in Fig. 14(b) (p = 0.2 torr) the theoretical curve shows several features which are confirmed by the observations, e.g. the modulation is now not complete, the minimum emission being about 5 per cent of the maximum, and there is a slight phase shift (~ 0.1 radian) of the intensity waveform relative to the current wave form.

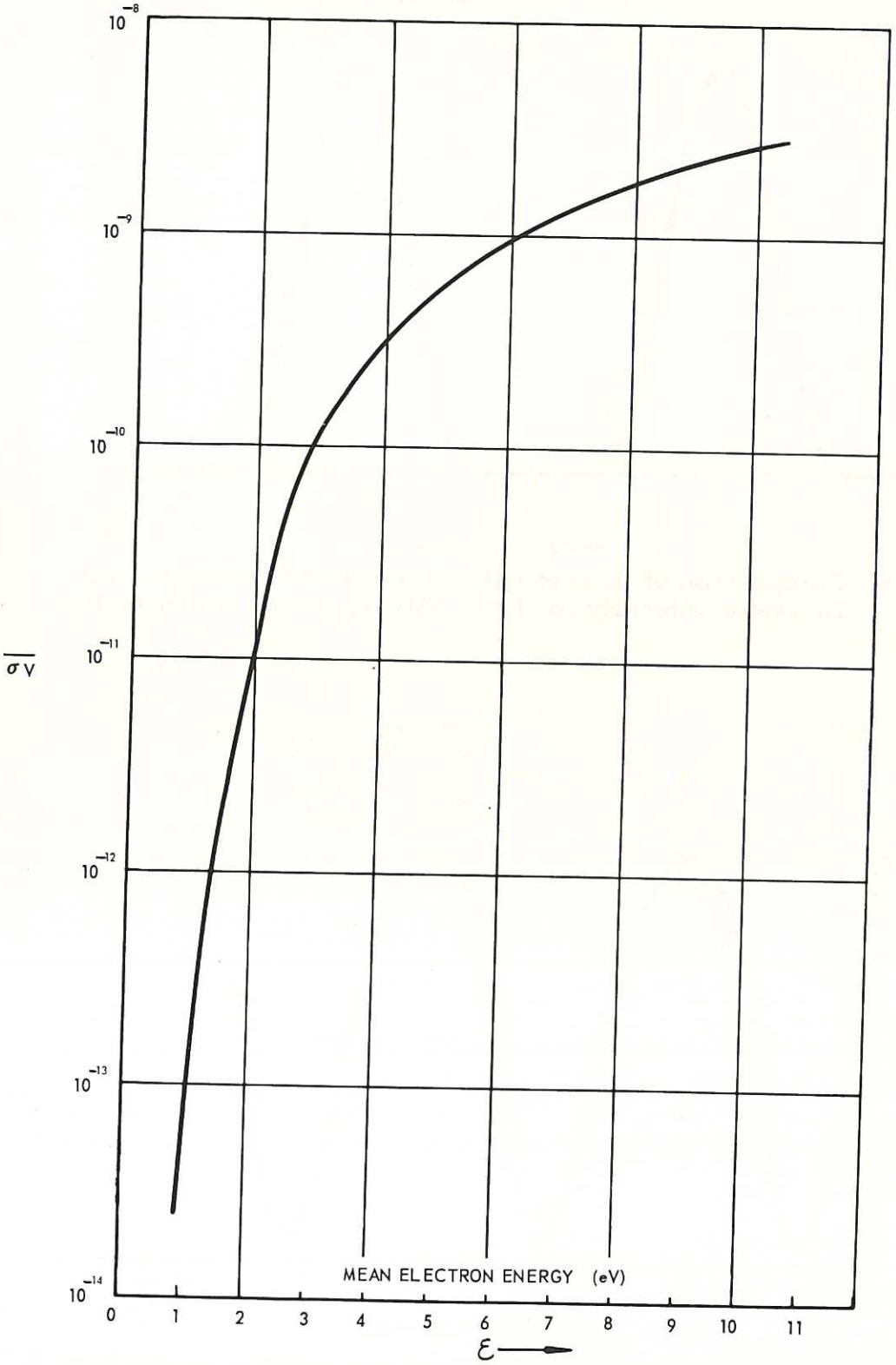


Fig. 13 Average excitation rate per electron per cm^3 for transition 1s→3p in atomic hydrogen

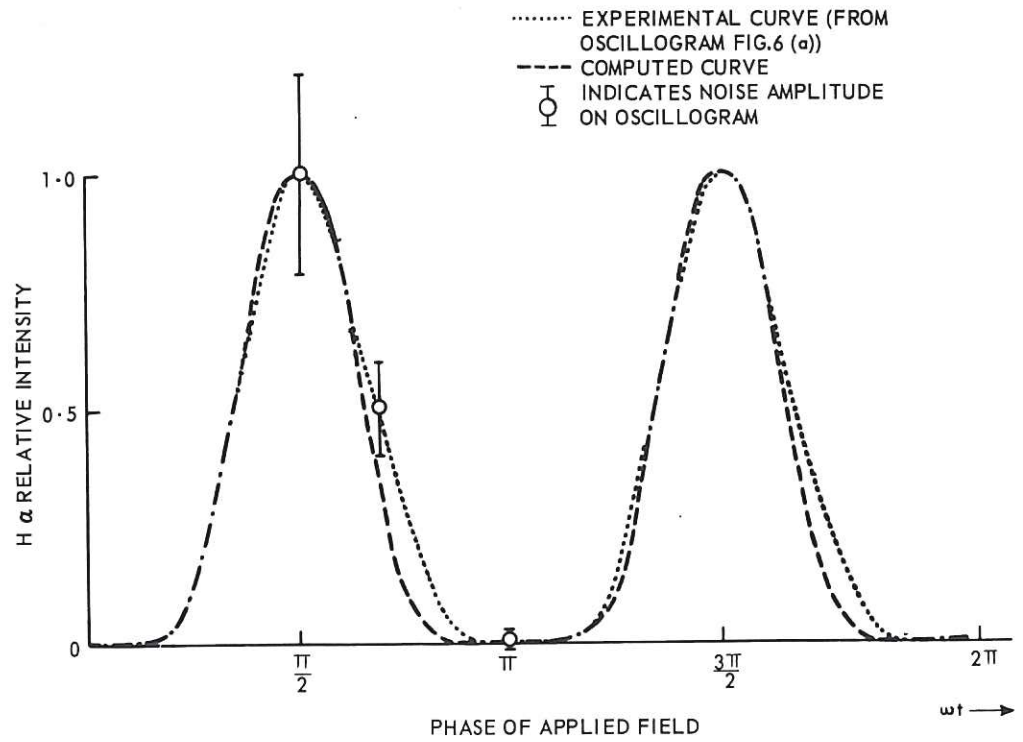


Fig. 14(a) Comparison of theoretical and measured variation in H Emission intensity for f = 1.5Mc/s, p = 4 torr (pure H_2)

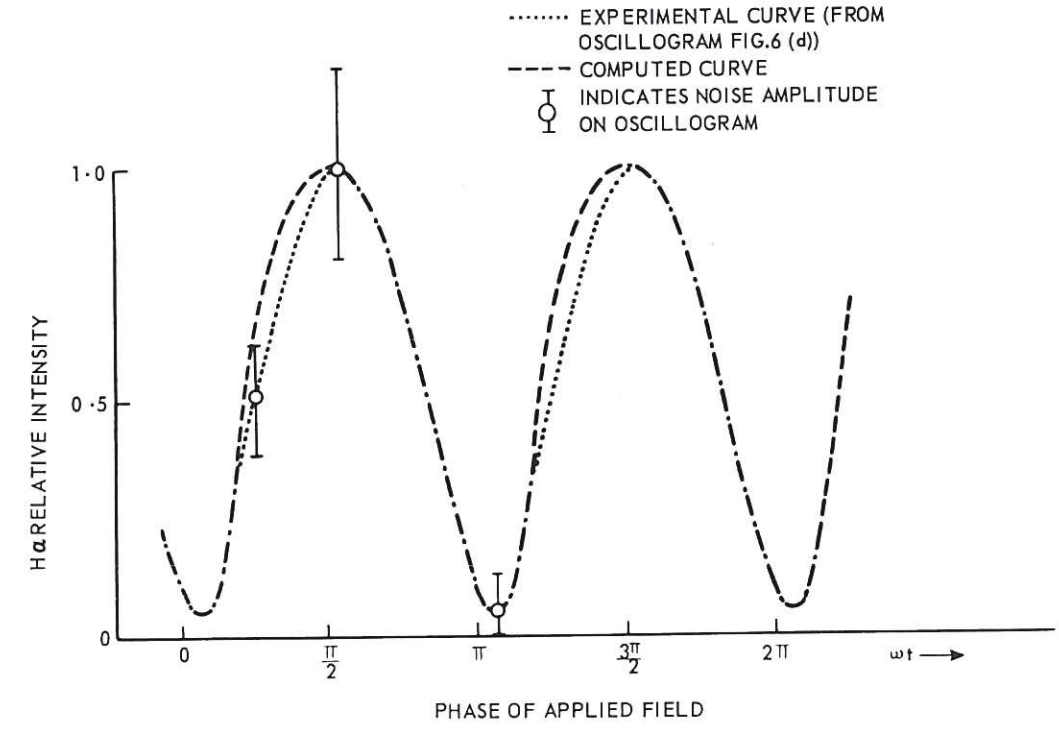


Fig. 14(b) Comparison of theoretical and measured variation in H Emission intensity for f = 1.5Mc/s, p = 0.2 torr (pure H_2)

5. DISCUSSION

48. The principles involved in the operation of the source at high frequencies are not limited to hydrogen lines nor presumably to those in the visible region of the spectrum. For instance, Lyman α would be similarly modulated, and could be extracted from the source through a window of suitable material, such as lithium fluoride. Equally, any atoms which may be present in the discharge as an impurity will be periodically excited by electron collisions and will therefore emit modulated light at the appropriate wavelengths. For example, modulated mercury lines have been seen in samples containing a trace of mercury vapour.

49. It should be clear from the previous section that the variation in ε needed for appreciable modulation of the excitation depends on the line required, and that ε need not be reduced to zero. For example, Fig. 13 shows that a fluctuation in mean energy between 2 and 3 eV would be sufficient to provide about 90 per cent modulation of $H\alpha$ (i.e. ratio of maximum to minimum intensity = 10:1).

50. It has been difficult to decide on the upper frequency limit of operation. We have observed hydrogen Balmer lines modulated at 22 Mc/s, but measurement at these frequencies was difficult (partly due to the lack of suitable detectors). Certain factors might be expected to become important at higher frequencies: for example the finite lifetime of the radiating atom would cause the light output to be smoothed when the reciprocal mean lifetime $\Sigma A_q \geq 2\omega$. An attempt has been made to observe this effect in excited states of hydrogen, for which the reciprocal mean life of the 5th level $\Sigma A_5 \approx 1.2 \times 10^7$ sec^{-1} (Allen, 1955). We should therefore expect appreciably less modulation of H_γ, which originates by decay of atoms with n = 5, at frequencies > 1 Mc/s. In fact we observed no noticeable decrease in the depth of modulation of H_γ as the applied frequency was increased to nearly 10 Mc/s. This apparent discrepancy may be due to the shortening of the lifetime of the excited atoms in the discharge conditions; the tabulated figure is, of course, obtained theoretically from the quantum mechanics of the unperturbed atom (Bethe and Salpeter, 1957), and would not apply in the presence of strong local fields. Alternatively it can be qualitatively explained by the depopulation of the n = 5 state by super-elastic collisions, whose rate also depends on the instantaneous electron mean energy.

51. It should also be noted that any contribution to light emission by radiative recombination would not be modulated in the same way as that due to the primary collision, since it depends on the product of charge densities, and not on the energies of electrons in the tail of the distribution. This effect would become important if current densities were increased considerably in an attempt to produce a very high intensity source, but is unlikely to be encountered when a sharp spectral line is required.

52. Although the device was developed for time resolved absorption spectroscopy there are other possible applications, for example, as a very fast stroboscopic lamp which might enable crystal vibrations to be studied visually. Another possible use is in optical (especially infra-red) communication (Beese, 1961). The radio frequency carrier could readily be frequency modulated to carry considerable information on one channel.. The inclusion of some caesium vapour would cause modulated Cs resonance lines in the near infra-red to be emitted strongly (λ = 8944, 8521 Å).

6. CONCLUSIONS

53. A simply constructed and operated source of intensity modulated spectral lines has been described, whose mode of operation is sufficiently well explained by a simple theory of binary exciting collisions whose rate depends on a fluctuating electron temperature. We have been able to deduce the necessary variations in electron energy to account for the detailed waveform of one spectral line by using experimentally measured collision parameters from d.c. discharges with similar values of X/p.

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