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RECENT DEVELOPMENTS IN EXPERIMENTAL PLASMA SPECTROSCOPY (WITH PARTICULAR REFERENCE TO APPLICATIONS OF TUNABLE LASERS)

by

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ABSTRACT

Recent progress in experimental spectroscopic work on plasmas is surveyed, with particular reference to developments in experimental technique. Developments in emission spectroscopic work described stem mainly from availability of new plasma sources, particularly at very high densities. Considerable attention is paid to present and future applications of frequency tunable lasers to plasma physics problems. An elementary derivation of the basic principles of such work is used to illustrate the fundamental differences from laser spectroscopy of isolated atoms and neutral vapours, and to show why applications to plasmas (to date) remain fairly limited. A survey is given of some recent results obtained at Imperial College, and elsewhere.

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1. INTRODUCTION

This is the second of two papers concerned with recent developments in plasma spectroscopy. The present paper is concerned primarily with recent developments in experimental technique, including applications of tunable laser techniques to plasma spectroscopic problems, and with new experimental results stemming therefrom, whereas the first paper* (referred to throughout as I) discussed present theoretical limitations in attempting to treat the spectroscopy of dense and ultra-dense plasmas. Where recent experimental investigations are specifically of relevance to high-density effects, the results themselves are discussed in I, and the investigations are then referred to here only in so far as novel experimental techniques are involved. Other investigations, at lower densities, or of more general consequence are described in the present paper. No attempt is made at a general review of the copious literature of emission plasma spectroscopy, but rather the paper concentrates on a few developments which appear to the author to be of particular significance at the present time, before turning to the topic of laser spectroscopy of plasmas.

Developments in orthodox emission spectroscopy of plasmas tend to result either from new plasma sources becoming available, or from sophistication in diagnostic techniques. As well as reviewing experimental details of several new plasma devices (in so far as their application to fundamental spectroscopic problems is concerned), attention is therefore also paid to recent diagnostic developments making use of fixed frequency lasers.

A fairly comprehensive discussion is given of the problems of applying frequency tunable lasers to plasma spectroscopic work, illustrated primarily with results obtained by the author's own group, In this instance, there appears to be no very wide ranging review (in so far as plasmas are concerned) in the published literature. The advantages of applying tunable lasers to plasma spectroscopic problems are obvious enough, often paralleling those in work on neutral gases and vapours, but there are a number of very important points of difference in work on plasmas. In several cases there are technical limitations due to the fundamental nature of the plasma itself, which do not seem to have been fully appreciated in the past. A detailed treatment is therefore given from first principles so that the differences from work on neutral gases appear naturally. Attention is given to technical requirements in terms of laser design and performance, and to the limits on the density and temperature of plasmas which can presently be studied with these methods. Finally some recent results are surveyed and future prospects discussed.

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2. NEW PLASMA SOURCES AND NEW TECHNIQUES IN EMISSION SPECTROSCOPY

The most significant recent development in emission spectroscopy of plasmas - which in many situations remains the only feasible technique for reasons to be discussed below in the section on tunable laser spectroscopy - has been the use of new plasma sources, particularly those operating at high densities. Figure 1 shows a plot of sources in current experimental use (the significance of the lines representing characteristic plasma parameters is discussed in Paper I). A somewhat similar plot was given in my earlier review in 1972, and comparison shows two main developments in the period since. One is that Tokamak plasmas at moderate density and very high temperature have indeed come into use for fundamental atomic physics investigations, in addition to their primary role in fusion research. Particularly significant for fundamental spectroscopic work is the long (0.1 to 1 second) observation time now available at high temperature ($T_e \approx 1$ keV), and the consequent excellent signal - to - noise levels, together with the fact that very high ion stages can be generated in a plasma in which the time scale is long enough for equilibrium ionisation to have been attained, and for transient effects to become negligible. Examples of such work will be described below. The other main development has been a fairly dramatic increase by about 3 orders of magnitude in maximum electron density available in laboratory sources. Largely, this has been due to the development of laser-compression experiments in which densities exceeding 10^{23} cm^{-3} are being achieved (see e.g. Ya'akobi et al 1977, Key et al 1978). These latter experiments, of course, are very complex involving multi-million dollar laser installations with output powers in the range 10^{10} to 10^{12} watts. However in several cases the experimental installations are nevertheless accessible to fundamental physics investigations in addition to work aimed at producing thermonuclear reactions. For example, such a programme is maintained in a university department at the University of Rochester, New York, and in the United Kingdom the Science Research Council operates a Central Laser Facility for basic physics research at the Rutherford Laboratory for use by university scientists throughout the country. Nevertheless, the fact remains that laser compression is a complex and expensive means of reaching very high densities and temperaturs. A most interesting recent development, therefore, has been the realization that low-inductance vacuum sparks - which for some time have been known to give very high ionization stages indeed (Lee and Elton 1971) - produce densities not far short of those achieved in current laser-compression programmes. Datla and Griem (1978) at the University of Maryland and Negus and Peaock (1978) at the Culham Laboratory, using a variety of measurement techniques have deduced densities from simple low-inductance vacuum sparks approaching, or exceeding 10^{22} cm^{-3}. Moreover, these densities are approached in a plasma which may reach temperatures as high as 10 keV, with a consequent energy density comparable to that in laser-compression experiments. Earlier measurements on such sparks had not shown such high densities, probably through the lack of spatial and temporal resolution, and there are indications at present that the lifetime of the small (1 micron) hot, dense plasma produced may be very short indeed, and the densities at present deduced correspondingly still a lower bound (Peacock, private communication). The mechanism of production of the dense hot regions in these vacuum sparks remains a matter of discussion although with some agreement that it must be due to intense electron beams (see e.g. Cilliers et al 1975 and Negus and Peacock 1978). Negus and Peacock (1978) have pursued a suggestion of Shearer (1975) that the initial stage in the production of the ultra-dense region may be a collapse due purely to radiation cooling, followed by a semi-explosive deposition of energy once a critical density is reached. These plasmas remain very difficult to handle in practice, due to the small size of the emitting region, and to problems with irreproducibility both temporally and in spatial location. Nevertheless they do offer the prospect of studying phenomena at very high energy densities with relatively simple equipment, and for a very modest outlay, which, unlike laser-compression experiments, is within the budget of typical university departments.

Results of laser-compression experiments have been quite widely reported in the literature, and at international conferences. No exhaustive review of this fast-developing field will be attempted here. Suffice it to say that impressive extensions of experimental technique have been achieved, particularly at very short wavelengths with extensive use of curved crystal spectrometers. Spatially-resolved spectra have been obtained by a number of groups, clearly showing differences between the outer-shell of the imploded micro-balloon and the compressed core (see e.g. Key et al 1978), and with electronic streak cameras very high temporal resolutions have been obtained (on the pico-second time scale) allowing the different temporal evolution e.g. of resonance and inter-combination line intensities to be used for diagnostic purposes. A number of groups have also obtained results showing pressure-broadened profiles for resonance lines of very high ionization states such as Ne X and Si XIII. (See e.g. Ya'akobi et al 1977, and Key et al 1978). Interest and difficulties in interpreting these observations due to high density effects are discussed in Paper I.

Examples of work performed with the vacuum-spark sources mentioned above include line profile studies by Datla and Griem (1978) and spectral classifications for helium-like and lithium-like molybdenum by Beier and Kunze (1977). These latter authors also observed possible plasma-frequency satellites close to the intercombination line in helium-like molybdenum, but the interpretation of such features is still severely limited by lack of knowledge of the detailed space-resolved conditions in such very energetic plasmas.

Agains, in the available space only a few examples can be given of recent work utilizing Tokamak plasmas. These will be chosen from those involving studies of collisional processes, rather than wavelength spectroscopy (in which area there has also been important use made of the special properties of these long-lived, moderate density high temperature sources where e.g. many forbidden transitions can be observed in the laboratory for the first time).

One particularly elegant experiment on ionization and recombination rates in high ($z \simeq 30$) ionization stages of molybdenum was recently reported by Breton et al (1978) using the TFR machine in France. These authors made use of a unique, reproducible instability which can occur under particular conditions in Tokamak plasmas, the so-called 'saw-tooth' instability. This instability literally modulates the plasma density and temperature of the plasma in a saw-tooth fashion, at a frequency conveniently related to ionization rates, so that observation of the temporal behaviour of line-radiation allows deduction of appropriate rate coefficients.

Another, separate, use of non-stationary effects in Tokamak plasmas is that due to Peacock and Summers (1978). These authors studied the spatial behaviour of emission of the resonance and intercombination lines of He-like Oxygen in the DITE tokamak at Culham under quiescent conditions. Using the known spatial variation of density and temperature, they were able to interpret spatial changes in the transitions observed as being due to non-stationary ionization effects, emitting ions diffusing significantly during relevant excitation relaxation times. From these XUV spectroscopic observations Peacock and Summers were able to deduce the ion diffusion rate in the plasma, a rather fundamental parameter!

Finally, spectroscopic observations of Tokamaks already extend to very short wavelengths, in the crystal region. One example is the work of Bitter, Vangoeler et al at Princeton, who have observed the resonance and intercombination lines of helium-like iron, Fe XXV, and nearby dielectronic satellites in a plasma with an electron temperature in excess of 8×10^6 K, with the exceptionally good spectral resolution for this wavelength region of 0.2 milliAngstroms.

In the context of collision rate measurements, one should also note the recent work of Brooks, Datla and Griem (1978), at Maryland on dielectronic recombination rates. Use of theta-pinches for collision rate studies in the VUV is, of course, well established (see e.g. Kunze 1972), but Brooks et al's work is particularly notable in two respects. Firstly, they have observed dielectronic recombination rates, e.g. in Fe IX and Fe X, which agree quite well with the low-density limit of the very well known calculations by my namesake, Dr A Burgess, (1965), but not with the values expected at densities in the theta-pinch used. Secondly, Datla et al have overcome the old experimental problem of how to insert elements such as Fe into a plasma without using volatiles (such as carbonyls) containing other contaminants, and which often cause other problems e.g. with vacuum systems. Brooks et al have used laser-evaporation of a thin foil, followed by slow diffusion (over a period of mseconds) into the theta-pinch region with good success. At the present time, the shot-to-shot reproducibility of this method appears only to be limited by that of the available laser, and of details of foil preparation (R Datla, private communication). This relatively simple technique for inserting trace elements could find much application also in other plasma spectroscopic work, e.g. on line-shapes or tunable laser spectroscopy of plasmas.

These comments are not meant to suggest that other work using long-established plasma sources is by any means exhausted. To the contrary, the past five years have seen a great volume of work of much-refined accuracy using all of the sources shown in Fig. 1. An attempt to review this in any way would be unfair to many authors not included within the scope of the survey. Specific examples in the context of particular problems are given elsewhere in this paper, and also with reference to high density effects in Paper I. Perhaps the only general comment that need be made is that all of the sources shown in Fig. 1, other than those specifically mentioned above, have essentially become standard tools with properties which, if not always fully explained, are nevertheless controllable and amenable to detailed diagnostic work. Applications of laser-based diagnostic techniques, such as Thomson scattering (see below) have moved from being the preserve of a few fusion laboratories to a point where they have been employed in many fundamental physics investigations using plasmas, and other spectroscopic techniques have developed comparably, so that in most of the cases shown the density and temperature distributions can in principle be determined with high accuracy, spatially resolved, throughout the plasma. Similarly, control of plasma composition and impurity level has consistently improved - a notable example of this being the work of Grutzmacher and Wende (1977) on line profiles for hydrogen Lyman alpha where the impurity hydrogen concentration was successfully reduced to such a point that the hydrogen resonance transition was optically thin. The classic plasma-spectroscopic problem of inhomogeneity of the emitting region has also largely been overcome in the past few years, either by careful and systematic inversion of the data obtained from a plasma of space-varying but known characteristics (see e.g. Wiese 1972), or by the use of a plasma in which the density and temperature distributions have been shown to be homogeneous to the point at which corrections to the data of interest can be modelled and shown to be negligible (see e.g. Smith and Burgess 1978). The most difficult area of study probably remains that of resonance lines from neutral species, since in these cases boundary layers can exist outside the plasma itself in regions where accurate diagnostic techniques are not available. It is an amusing fact that the classical spectroscopist's jibe against plasma spectroscopy that "the conditions in the source are unknown" has been reversed by modern diagnostic techniques to the point where conditions in most plasma spectroscopic experiments are actually much better known than in the furnaces, etc., used in other spectroscopic work.

3. EXPERIMENTAL WORK USING FIXED-FREQUENCY LASERS

3.1 Diagnostic Applications

A number of reviews exist of applications of fixed-frequency lasers to plasma interferometry e.g. Jahoda (1971) and to scattering measurements e.g. by Evans and Katsenstein (1969). A very comprehensive discussion of applications of Thomson scattering measurements under virtually all plasma conditions has been published in the form of a book by Sheffield (1975), which in each case treats the underlying physics in detail. One very interesting advance has been the use of Thomson scattering techniques for investigation of magnetic field directions in plasmas, as opposed to the more conventional use for measurements of density and temperature distributions. These magnetic field direction observations depend upon the fact that the Thomson scattered spectrum is modulated at the electron cyclotron frequency only in that case where the scattering vector lies accurately perpendicular to the magnetic field, since in any other direction differential Doppler shifts smear out the modulated cyclotron frequency structure. Early attempts to observe such effects concentrated on the use of far infra red laser systems, since the requirements on the scattering vector direction are then less severe than for visible incident laser frequencies, but Sheffield (1972) pointed out the important fact that the restriction at visible frequencies could be turned to great advantage by using a detector system which preferentially discriminated in favour of the electron cyclotron frequency structures, hence allowing direct observation of the magnetic field direction. (In most magnetized plasmas the magnitude of the magnetic field is reasonably well known, and it is the direction which is least certain and most important for stability studies). The actual use of this very elegant and difficult techniques in a tokamak plasma for determination of magnetic field directions has been reported very recently by Forrest et al (1978).

3.2 Other Applications of Fixed-Frequency Lasers

One other application of fixed-frequency lasers (other than to plasma generation as per se, which is outside the scope of this paper) needs mention, because it is the subject of some considerable interest and spectroscopic study at the present time. This is the situation where a fixed laser frequency is used to couple energy selectively into plasma modes, via the resonant interaction occurring in the boundary of an inhomogeneous plasma at that spatial point where the density is such that either ω_{pe} or $2\omega_{pe}$ coincides with the incident laser frequency. (This occurs naturally in almost all laser-plasma generation experiments).

In laser compression experiments striking effects have been seen by several groups (see e.g. Key et al 1978), spectra showing intense emission from localised regions at $2\omega_{\text{incident}}$ and $\frac{3}{2}\omega_{\text{incident}}$ (corresponding to absorption of the laser beam at ω_{pe}, and subsequent direct emission of EM radiation by decay of two of the plasmons generated, or absorption at ω_{pe} and subsequent mixing with the incident laser light).

A difficult but very elegant experiment on related effects has been performed on a plasma focus device by Peacock et al (1977) at Culham Laboratory. These workers focussed a high intensity CO_2 beam (10.6 microns wavelength) into the relatively dense (10^{19} cm^{-3}), hot plasma, and then observed enhanced Thomson scattering of an incident Ruby laser beam from the same region of plasma with the fundamental aim of observing coupling effects due to ion acoustic modes.

4. APPLICATIONS OF FREQUENCY-TUNABLE LASERS TO PLASMA SPECTROSCOPIC PROBLEMS

4.1 General Remarks

The last 10 years have seen the appearance of a very wide range of frequency tunable laser systems, both pulsed and CW, which are well described in many reviews (see e.g. Laser Applications to Optics and Spectroscopy, ed.

Jacobs et al, Addison-Wesley 1975). Broadly speaking the whole of the visible spectral range is now covered, both in so far as pulsed high intensity lasers are concerned and with slightly more restrictive wavelength limits also by CW systems. Various other systems work in the near UV, frequency-doubling is possible, and in some specific instances frequency tripling in atomic vapours has extended wavelengths into the VUV although as yet only over rather isolated wavelength ranges. Some laser systems, the excimer and exciplex lasers, already directly give high (MW) power outputs in the UV and VUV and these have also been frequency-multiplied, e.g. to 570 Å (see Bradley et al, 1976). Down conversion schemes are also possible, so that (given a challenging enough scientific problem) a very wide range of wavelengths can be reached, at least in principle. In the visible and near UV tunable lasers have become standard laboratory items rather than research projects in their own right, and applications extend right through physics, chemistry and biology.

One rather startling contrast to all this activity is the still relatively very few applications reported of such lasers to plasma physics problems (see however Section 5 below) despite their immense impact elsewhere in atomic physics research. A cynic might comment that this is partially because once one has a tunable laser, there is a never-ending supply of interesting problems to investigate with a simple sodium vapour cell free from the worries inherent in using a complex plasma source. Also, the difficulties of a new technique are not always well adapted to the pressures of work and limitations of time on a complex fusion device, as we at Imperial College found to our cost in attempting in 1971 what I believe was the first ever application to such a plasma on the CLEO Tokamak at Culham (see e.g. Skinner 1974). (A rather similar investigation was, however, successfully carried out by Burakov et al 1977). Mainly, however, the reasons why relatively little work on plasmas has yet been reported are fundamental problems concerned with the physics of the plasma itself. The advantages of the tunable laser remain, but they have to be applied in different ways to those used in most atomic physics work - in some cases by developing special purpose plasma sources. These problems will now be reviewed.

BASIC AIMS

The general problems of interest in plasma spectroscopic research, varying from fundamental statistical mechanical problems, through pure atomic physics to applied diagnostic work on fusion devices have been surveyed in a previous review (Burgess 1972) and in various other papers in the literature (e.g. Cooper 1966; Kunze 1972). In paper I (Burgess 1978) specific physical problems at high plasma densities have also been discussed. The pertinent question at the present point is to consider which of the various properties of tunable lasers can be used to advantage in such work on plasmas. Roughly speaking, the advantages of lasers in general spectroscopic work can be listed as follows:-

a) High spectral resolution
b) Possibilities of spatial resolution (e.g. in scattering experiments).
c) High intensity, with the following consequences:-
  (i) High signal-to-noise ratios.
  (ii) Selective perturbation of specific level populations, etc.
  (iii) Excitation of specific collective modes of the system under study.
d) High time resolution - either of rapidly time-varying plasma sources themselves, or of fast rate processes in quasi-stationary plasmas.

(a), the possibility of high spectral resolution has been the basis of a very large number of applications in atomic physics (other than on plasmas), but is of relatively little consequence in work on plasmas where line-widths of interest are usually very broad (tenths of Å at least and quite conceivably tens of Angstroms). Even if Doppler-free techniques, such as saturable absorption spectroscopy, are applied, the pressure-broadened line-width is usually sizable. (As will be seen below, Doppler-free techniques in plasmas are rather difficult in practice. In addition, the time between velocity changing collisions is much shorter than in neutral gases, being only on the order of 10^{-9} secs for ions in a very modest plasma at 10^4 K and 10^{15} cm^{-3}).

Possibilities of spatial resolution (b), on the other hand are of very considerable consequence in plasma physics work, where sources are often inhomogeneous, and interpretation of emission spectroscopic work thereby limited. However, tunable laser work is still limited in this respect, both because of beam divergence problems with the simpler lasers, and for more fundamental reasons which will be discussed below limiting e.g. scattered light signals in a way that does not apply to say Thomson scattering.

All of the consequences of high intensity, (c), are of very considerable importance, and to some extent all three have already been exploited. (c)(ii) and (iii) are certainly the most fundamental and exciting, but it is easy to forget quite how important (c)(i) may be even if one uses the laser for purely conventional 'linear' spectroscopy and how limited signal-to-noise ratios are in emission plasma work. Examples are given later in the present paper, and in the paper elsewhere in the Symposium by myself, Kolbe and Playford.

Finally, whilst the high time resolution possibility (d), is of obvious importance, and has been much investigated in other applications of tunable lasers, it has not yet been much exploited on plasmas (although use of fixed frequency lasers for this purpose is fairly common-place).

LASER REQUIREMENTS FOR TYPICAL PLASMA CONDITIONS

We shall consider the requirements for a laser to be able to significantly alter atomic state populations first, and then treat those e.g. for application to, say, absorption measurements second, because the latter comes naturally into consideration of detection of changes produced in a pumping experiment as well as into orthodox absorption spectroscopic applications.

4.2 Pumping

One of the most important requirements in applying tunable lasers to plasmas is to be able to change significantly a selected atomic level population, whether in order to study particular atomic rate processes, to couple energy into some other process in the plasma (e.g. electron plasma waves), or simply to isolate some spatial region of the plasma by locally altering its emission properties.

As in laser spectroscopy on neutral vapours, there are two stages to this. The first is simply to be able to alter the atomic level population of interest by an observable amount. The second is to be able to irradiate the transition of interest sufficiently strongly that is is saturated, that is the upper and lower state populations, n_2 and n_1, are 'equalised' or rather put into the ratio of their statistical weights, g_2/g_1 (as will obviously happen if the laser intensity is sufficient that absorption and stimulated emission in the resonant transition dominate all other processes since then $I_\nu B_{12} n_1 \simeq I_\nu B_{21} n_2$ with $g_1 B_{12} = g_2 B_{21}$).

Two complications arise in plasma spectroscopic work. The first is that very high laser intensities are required, because of the high competing collision rates. The second is that whereas in ordinary laser spectroscopy saturation almost always implies observability, in plasma work this is not true and a considerably higher intensity may be needed for any change in the upper level population to be observed than is required to saturate the transition.

To understand these points quantitatively we will work through the well-known theory for an isolated atom and then extend this to an atom in a plasma. Consider Fig.2.

4.2.1 Case 1 - Isolated Atom

Fig.2(a) shows the typical situation relevant in much work on neutral atomic vapours. The only mechanism opposing saturation is spontaneous radiative decay in the transition itself. The requirement on the laser power in order to significantly populate the upper level of this simple two level system is then obtained from the relevant rate equation for the upper level population:-

$$\int_0^{\Delta\Omega} \frac{n_1 I_\nu B_{12}}{4\pi}\, d\Omega = \int_0^{\Delta\Omega} \frac{n_2 I_\nu B_{21}}{4\pi}\, d\Omega + n_2 A_{21} \qquad (1)$$

where I_ν is assumed constant across the line profile ϕ_ν (so that the integration $\int I_\nu \phi_\nu d\nu$ can be carried out to yield the total rate), and $\Delta\Omega$ is the solid angle filled by the laser beam at the absorbing atom.
Hence the power required is given by:-

$$P = \int_0^{\Delta\Omega} I_\nu \Delta\nu \quad d\Omega$$

$$= \frac{4\pi A_{21}\ \Delta\nu}{B_{21}}$$

$$= \frac{8\pi\Delta\nu h\nu^3}{c^2} \qquad (2)$$

$\Delta\nu$ here is whichever is the larger of the homogeneous line-width of the transition, or the minimum output bandwidth of the laser. Notice that the intensity required is simply equivalent to an isotropic value of 2 photons per Hz per (wavelength)^2.

For typical visible transitions this power comes out to be somewhere in the region of 1 - 100 watts. Note the other features of the expression (2). P is independent of the transition probability, and whilst it depends on ν^3, this factor only varies by a small factor along a resonance series. Hence, as long as the laser power is ample to saturate the first member of a resonance series, one can probably carry out laser spectroscopy on any member of the series - witness all the recent work on laser spectroscopy of very high Rydberg states (see e.g. Ewart 1977). This contrasts very severely with the situation in a typical plasma, where the power required may well be Megawatts rather than watts, and increases very rapidly along a series, as we shall see below.

4.2.2 Case 2 - Two-level Atom with Collisions only between the 2 Levels

This case, Fig.2(b), is actually academic, since in most plasma situations when collisions are important (relative to spontaneous emission), upward transitions out of a given level dominate downward ones. (The exception to this rule is if the plasma is cooling and recombining fast, and far from equilibrium in so far as level populations are concerned). However, understanding of the 2 level atom with collisions is useful when we turn below to the case where collisions also cause upward excitation out of the upper level, so we will treat it briefly.

At first sight the expression for the power needed changes rather little, A_{21} in the derivation simply being replaced by the downward decay rate out of the upper level, Γ_{21} ($\Gamma_{21} > \Gamma_{12}$, so we may neglect Γ_{12} for these purposes. Hence we have

$$P = 8\pi\Delta\nu \frac{h\nu^3}{c^2} \frac{\Gamma_{21}}{A_{21}} \qquad (3)$$

Unfortunately, there are two major changes in this innocent-looking formula:-

1) Γ_{21} increases rapidly with increasing principal quantum number of the upper level (about as n^2), whereas A_{21} decreases rapidly.

2) $\Delta\nu$ is no longer the natural width, but is the pressure-broadened width. Unfortunately, the pressure-broadening collisions are almost never the same as those corresponding to Γ, but are much more frequent! Corresponding line-widths are therefore very large - e.g. 2Å for Hβ (4861 Å) at a density of only 10^{15} cm^{-3}. Moreover, these widths also increase rapidly along a series (again as about n^2).

So even within this simple model we have two important changes:-

1) Because of the high values of Γ and of $\Delta\nu$ (due to separate collisional mechanisms) the power needed to saturate a transition increases dramatically in comparison with the collision free case even for very modest plasma densities (see Fig.3).

2) Because of the n-scaling of Γ, $\Delta\nu$, and A_{21} the power needed to saturate increases as something like n^6 (or more) along a given series (see Fig.3).

Fig. 3 (solid lines) shows the power needed for stimulated emission to just equal collisional de-excitation for the Balmer series of hydrogen, neglecting collisional excitation upwards. (The reason why this is still a useful model, despite high upward collision rates, will become clear below). Collisional de-excitation rates in Fig.2 are calculated from Johnson (1972) and line widths are taken from Griem (1974). We see at once that in order to be able to do much laser spectroscopy on, say, the Hβ transition we need a tunable laser with something approaching One Megawatt output power, even if we restrict ourselves to relatively low plasma densities such as 10^{15} cm^{-3} (compare the plot of densities of typical plasma sources in Paper I). (Reasons why the requisite flux cannot simply be produced by focussing down are discussed in 4.3)

4.2.3 Case 3 - 2-level Atom populated also by Collisions Into and Out of the Upper Level (only)

It is useful to consider one more simple model before turning to the general case.

The rate equations for the long-term equilibrium of the situation shown in Figure 2(c) are:-

Upper Level

$$C + n_1 \int_0^{\Delta\Omega} \frac{I_\nu B_{12}}{4\pi}\, d\Omega = n_2 \int_0^{\Delta\Omega} \frac{I_\nu B_{21}}{4\pi}\, d\Omega + n_2(\Gamma_{21} + A_{21}) + n_2 D \qquad (4)$$

Lower Level

$$n_1 \int_0^{\Delta\Omega} \frac{I_\nu B_{12}}{4}\, d\Omega = n_2 \int_0^{\Delta\Omega} \frac{I_\nu B_{21}}{4}\, d\Omega + n_2(\Gamma_{21} + A_{21})$$

From the second equation, neglecting spontaneous emission, $\frac{n_2}{n_1} = \frac{g_2}{g_1}$ if the condition (2) is met, i.e. saturation depends only upon the collision rate between the two levels, and is not affected by D, (the remaining rate out of the upper level).

At first sight this sounds encouraging! The shock, however, comes if we substitute into the upper level rate equation (4). With $\frac{n_2}{n_1} = \frac{g_2}{g_1}$ the 2 terms dependent on the laser intensity are equal and cancel. The upper level rate equation then becomes:-

$$C = n_2 D + n_2(\Gamma_{21} + A_{21}) \qquad (5)$$

This is just the rate equation for the level in the absence of any laser radiation at all - so although we have equalized the level populations we have caused no long term change in the upper level population whatever! (This problem was already recognised by Measures in 1968, prior to the advent of practical tunable laser systems).

As already stated, it will be usual in a plasma for D, the upward rate collision out of the upper level of the pumped transition to exceed the downward rate, Γ. Consequently, we have the following situation:-

1) Significant enhancements of the upper level population of a pumped transition will be transient only, relaxing on a time-scale of 1/D - this is plotted for the first few levels of hydrogen in Figure 4. (The case where the lower level is also populated from the other levels is discussed below).

2) To achieve any instantaneous enhancement in the upper level population, the laser power must exceed:-

$$P = 8\pi\Delta\nu \frac{2h\nu^3}{c^2} \frac{D}{A_{21}} \tag{6}$$

Since D is usually larger than Γ_{21}, the power to observe significant (transient) enhancement of the upper level population exceeds that to saturate the transition.

This power is plotted for the Balmer series as a dotted line in Figure 3.

The two points that arise from this discussion therefore, are that, in a plasma, observation of changes in the upper level population, e.g. by looking at fluorescence scattering, may be a very hard way to do laser spectroscopy, and secondly that where a long-time enhancement of the upper level population does occur, it automatically provides information about the processes populating the lower level of the pumped transition (since without such processes the upper population relaxes to its initial value even during the laser pulse). In many cases it may be easier (i.e. require less laser power and lower time resolution) to study the lower level of a pumped transition (since usually this is appreciably changed in the long-time limit).

4.2.4 General Case of a Resonantly Pumped Atom in a Plasma

It will be clear that the general case of an atom with both upper and lower levels of the pumped transition coupled collisionally and radiatively to other levels cannot be solved analytically. The only alternative is to solve the entire coupled set of time-dependent collisional radiative rate equations, not only for the levels of the pumped transition itself, but also for the (time-varying) populations of all other levels in the atom. Such a treatment for hydrogen has been developed by Dr Valerie Myerscough of Queen Mary College, London, and has been used by her both in investigating possibilities for diagnostics of Tokamak devices (using Lyman-alpha pumping), (McIlrath, Myerscough and Koopman 1978), and interpreting a long series of experiments by my group on low temperature hydrogen plasmas pumped on H-alpha or H-beta. This code treats up to 20 levels of the hydrogen atom individually, includes all rate processes normally treated in standard time-dependent collisional radiative theories (e.g. Johnson and Hinnov 1972), and can calculate the time-dependent population changes induced by a laser pulse of arbitrary power and time-history.

In many cases, however, a much simpler model due to myself and C H Skinner (Burgess and Skinner, 1974) is quite useful which yields an analytic expression for the level populations of a saturated transition as a function of time by treating generally collision processes into and out of both states of the pumped transition, but ignoring all changes in the population of other levels. Comparisons with Myerscough's complete treatment show that in hydrogen this simple model is usually within about 20% of the predictions of a complete treatment (although not with the results of experiment, see below!).

The analytic expression for the upper level population as a function of time on the Burgess-Skinner model is then:-

$$n_2(t) = \left[\frac{n_1(o) + n_2(o)}{1 + g_1/g_2} - \frac{(C_1+C_2)g_2}{g_1D_1 + g_2D_2}\right] \exp\left(\frac{-g_1D_1 + g_2D_2}{g_2 + g_1}\right) + \frac{(C_1+C_2)g_2}{g_1D_1+g_2D_2} \qquad (7)$$

where C_1, D_1 and C_2, D_2 are the total rates into and out of the lower and upper levels of the pumped transition respectively (ignoring processes between the 2 levels themselves). Notice that since the laser remains on, the time-history depends on processes affecting the lower as well as the upper level, in contrast to techniques using short laser-pulses.

Figure 5 shows an experimental result showing essentially this form, in this case observing the fluorescence produced by pumping on the He Iλ 5876 $2^3P - 3^3D$ transition (see Skinner 1974 for details). As already remarked, the existence of a long-time 'plateau' enhancement of the upper level population clear in the experimental trace - automatically means that rates into and out of the lower state of the pumped transition are significant, and provides data on them. As pointed out in Skinner's and my 1974 paper, this is a potential means of obtaining from visible frequency spectroscopy alone such data for first-excited states, which would otherwise require VUV spectroscopic techniques.

An illustration that the lower state of the transition really does affect the time behaviour of the fluorescence - and also a warning that the Burgess-Skinner solution must always be checked for validity - is shown in Figure 6, again taken from Skinner, 1974. This shows the results of pumping the singlet transition equivalent to the $2^3P - 3^3D$ line shown in Figure 5. In this case, pumping on the $2^1P - 3^1D$ 6678 Å transition, a quite different time-history is produced. Collision rates for the upper levels of these two transitions are closely similar. The difference, therefore, lies purely in the lower levels of the pumped lines. However, the analytic form (7) is not produced. The reason must be that the population of one of the nearby levels populating 2^1P - probably 2^1S - is not time-stationary during the laser pulse (i.e. as assumed by the Burgess-Skinner model).

Clearly, such laser pumping experiments can provide a wealth of previously inaccessible detail on specific collision rates. We will return to quantitative results obtained for hydrogen below (Section 5).

4.3 Probing and Detection

The difficulties of laser pumping produced by the high collision rates in a plasma have already been discussed. One obvious consequence is that in many situations available laser powers will be inadequate to cause saturation and only small relative changes in level populations will occur. This causes obvious difficulties in observing the consequences of such a laser pumping experiment. What is perhaps not so obvious is that there are severe difficulties in detection even if saturation is achieved. Again, there is a strong contrast with work on isolated atoms or neutral vapours. In these cases background emission is usually very low, line-centre optical depths can be high (e.g. in furnaces), and the background electron density is low or indeed zero. The experimenter therefore has a choice of detecting fluorescence emission, laser-induced optical depth changes, or causing photo-ionisation out of the excited state and detecting the electrons so generated (see e.g. Ewart and Purdie 1976). Alternatively, he can look for bulk changes induced in the gas as a whole, such as sound waves, providing very sensitive detection mechanisms as in opto-acoustic spectroscopy (see e.g. Colles, Angus and Marinero, 1976).

In a hot plasma opto-acoustic and photo-electron techniques are impossible for obvious reasons, and the choice is between fluorescence observations and optical depth probing, both of which have difficulties of their own.

FLUORESCENCE SCATTERING

Two things need to be considered in a fluorescence scattering observation, the absolute signal level and the ratio of signal-to-background. The sensitivity of an experiment is limited by four main considerations:-

a) The scattered intensity saturates at high enough laser power. Beyond a certain level, nothing can be gained by producing a more powerful pump laser (but as Figure 3 shows, the idea of saturation may be academic, because it can be hard enough to produce an adequate laser in the first place!). On the other hand, provided the fluorescence signal is still observable, saturation is a great experimental convenience, because it means that shot-to-shot fluctuations in the laser output do not matter much.

b) The enhancement produced in a fluorescent scattering experiment is transient (see Figures 4 and 5), usually being shorter than the input laser pulse.

c) Even the initial transient in the fluorescence is likely to represent a relatively small increase above background (at least until far UV lasers become generally available), simply because easily accessible transitions for laser pumping in typical plasmas have $h\nu < kT$. Consequently, the ratio of level populations prior to the laser pulse - and hence the enhancement generated - is small. The good enhancement shown for He I 5876 in Figure 5 is simply because this result was taken in a cool (1 eV) recombining plasma where the first excited states were very overpopulated (relative to equilibrium).

d) Finally, the initial absolute level populations are also likely to be small, since for most atomic or ionic species of interest in plasma spectroscopy, the only transitions accessible with presently available lasers are between excited states. However, this turns out to be not as serious a limitation as (c) - estimates show that e.g. in a Tokamak at say 1 keV temperature, the absolute scattered signal from a neutral hydrogen population of only 1 part in 10^7 would be detectable.

The crucial problem therefore is signal-to-background ratio. Low absolute level populations are not a particular problem, since they also imply low background emission. (b) and (c) above combined are the real difficulty because they imply limits on the spatial resolution that can be obtained. For instance, estimates (unpublished) show that for residual hydrogen in a Tokamak, pumping on Hα, a local enhancement of between 2 - 3 is still possible at temperatures of a few hundred electron volts. However, one wishes to spatially isolate say 1 cm^3 in a plasma perhaps 50 cm across. The signal-to-background ratio, therefore, is going to be very low. For this reason UV sources tuned to the Ly-α transition are being specifically developed, e.g. via frequency tripling at University of Maryland and Culham Laboratory or using the argon excimer laser (Professor D J Bradley's group at Imperial College).

Observations on lower temperature plasmas using fluorescence scattering are discussed in Section 5.

RAYLEIGH SCATTERING

One possible way round the background emission problem is via Rayleigh scattering observations, tuning a laser close to but not on resonance with a transition of interest (to a first approximation the scattered light being at the input laser frequency). This technique has been used by Wrobel, Steuer and Rohr (1976).

Four points need noticing :-

1) The scattered intensity is still limited by saturation since eventually stimulated Rayleigh scattering exceeds the spontaneous rate. Neglecting collisions the scattered intensity is limited to the identical maximum value that pertains for a resonantly pumped system.

2) The input laser intensity required to cause saturation is much higher than for resonant pumping, varying as $(\Delta\omega)^2$ where $\Delta\omega$ is the frequency separation from line-centre.

3) The lack of saturation can be an advantage, since the effective cross-section is independent of input intensity, allowing easy calibration (see Wrobel et al 1976).

4) Since electron collisional rates involve much larger energy transfers than likely laser de-tunings, the pumped states will be depopulated via collisions out of the virtual upper stage at a rate closely similar to that occurring for resonant excitation, so that similar transient effects may occur if saturation is approached. (Below saturation collisions are not very important, see Wrobel et al 1976).

Use of laser powers sufficient to saturate the Rayleigh scattering in a plasma is presently unlikely. Even ignoring saturation effects, in most cases near-resonant tuning turns out to be essential before Rayleigh scattering is likely to be observable, because of the dependence of the (low intensity) cross section on the frequency-detuning ($\propto\Delta\omega^{-2}$). Scattering off ground state atoms of interest is therefore unobservable in plasmas with visible frequency lasers. For ground state atomic helium, for example, the Rayleigh scattering cross-section at 6000 Å is only about 10^{-6} of the Thomson scattering cross-section (i.e. the effective Rayleigh cross-section is about 10^{-31} cm^2). Correspondingly, it would be necessary to tune a laser to within about 1 Å of the helium resonance line at 584 Å before the effective cross-section for Rayleigh scattering off the helium atoms became comparable to the Thomson cross-section for scattering from free electrons (which will usually have a comparable or higher number density in cases of interest). Even for visible transitions and except in partially ionized plasmas it may be very difficult indeed to produce scattering observable above the Thomson background unless tuned so close to resonance that background emission from the line wings becomes a severe problem. This may indeed nullify any advantage over actual resonant scattering, particularly if the transition of interest is pressure broadened and consequently has extended line-wings.

DETECTION VIA LASER-INDUCED OPTICAL DEPTH CHANGES

The other simple possibility for laser spectroscopic work on plasmas (i.e. other than detection of laser-induced scattering) is to observe changes in level populations in optical pumping experiments via the consequent laser-induced optical depth changes. Such observations, of course, are widely used in conventional laser spectroscopy, e.g. in saturable absorption spectroscopy. One attraction in work on plasmas is that, as shown, above it is easier to alter the population of the lower level of a pumped transition than the upper one. The difficulty in a plasma comes about because of the very low optical depths per unit path for transitions of interest - often 10^{-6} to 10^{-8} of that typical of laser spectroscopic studies on neutral gases. This large difference does not come about through any single effect, but through a number of small factors working in common.

A useful relationship for the optical depth per centimetre path at the centre of a Lorentzian (pressure-broadened) line is:-

$$\tau \text{ (per cm path)} = \frac{D\, n_i\, f_{12}}{118\, \Delta\nu}$$

where $\Delta\nu$ is the line half-half width (in Hz), n_1 the population of the lower state of the transition, f_{12} is the absorption oscillator strength and D the optical path in cm.

The three important differences in a plasma are:-

1) Lines of interest in the visible spectral region are almost invariably out of excited states, so that n_1 is usually very low (typically not exceeding say 10^{-3} of the electron density for anything other than the ground state).

2) The line-width, $\Delta\nu$, is large - e.g. equivalent to 1 Å for the 4861 Å Hβ transition in a plasma at only 10^{15} electrons cm^{-3} as opposed to typical Doppler widths in work on neutral gases of milli-Angstroms.

3) The f_{12} value is small for transitions between excited states.

As an illustration of these three effects working in common, consider the case of Hβ in a pure hydrogen plasma at $T_e = 10^4$ K, $n_e = 10^{15}$ cm^{-3}. In this case the predicted line-centre optical depth per centimetre path is only about 1.2×10^{-3}, so that several metres optical path are necessary in order to use laser absorption spectroscopy, either in a straightforward manner, or to detect the results of a laser-pumping experiment. Once again, there is a pressing need for VUV lasers operating at typical resonance line wavelengths, where much larger optical depths (and hence also optical depth changes) occur.

An alternative possibility proposed by the present author is that of detecting small laser-induced population changes via interferometric observations of the consequent changes in anomalous dispersion close to resonance. This has the advantage that the sensitivity can be varied to suit a wide range of transitions by varying the detuning from resonance, but the disadvantage that it is necessary to operate with a reference beam also in the plasma if absolute population densities are required since the free electron contribution to the refractive index is relatively very large. Long optical paths are still essential, the population change for one fringe shift to be produced being given by:-

$$\Delta n = \frac{m\ f_{12}}{2\pi\ e^2} (\omega^2 - \omega_o^2) \frac{\lambda}{D}$$

where D is the optical path, m the electron mass and ω_o the resonance frequency of the transition of interest.

This technique - known as Refractive Index Modulation Spectroscopy (or RIMS) is under study using the 10-metre plasma column at Imperial College.

4.4 Choice of Laser Design

At the present stage of development, there are three easily accessible and alternative types of dye laser systems suitable for work involving selective excitation of specific transitions in plasmas:-

1) Solid-state (ruby, Nd-Yag or freq-doubled ruby or Nd-Yag) pumped dye lasers.

2) N_2 pumped dye lasers.

3) Flash-lamp pumped dye lasers.

In many applications flash-lamp pumped systems have experimental advantages, as well as being simple and cheap. On pulsed plasma devices where the plasma lasts milliseconds at most, and the time between shots is minutes (if not hours!), it is impossible to exploit the high repetition rate possibilities of N_2 or Nd-Yag pumped systems. The short pulse lengths of these systems are rarely essential - given that in a plasma relaxation effects can equally be observed during a sustained laser pulse - and in many situations the longer pulses from flash-lamp pumped lasers confer signal-to-noise advantages. Finally, whilst flash-lamp pumped systems are harder to tune to really narrow line-widths (because of the low-Q cavity resulting from the poor optical quality of the lasing dye), this is not often significant in plasmas where line-widths of interest are usually large in any case.

There are two main choices of design for flash-lamp pumped dye lasers, namely co-axial flashlamp or confocal cavity. In the former the pumping light is generated in a narrow (0.3 mm) annular gap surrounding the dye cell, whereas in the latter a linear flash tube is used at one focus of an elliptical cavity. Both have advantages, and both are commercially available from many suppliers. The confocal laser probably wins out for mode control - and hence beam divergence and/or narrow band tuning (involving a series of intra-cavity Fabry-Perots) - whereas the co-axial system probably has the edge in actual intensity, and also in ease of home construction (the most critical parameter being accurate control of the flash-tube gap size in order to prevent filamenting). In many cases, the beam divergence and intensity available from either type of system can be upgraded by using a flash-lamp pumped laser to excite a separate small dye cell, thus avoiding optical problems due to inhomogeneities in the flash-lamp pumped system. Fielding (1974) has described a transverse pumped system of this type, and McIlrath, Koopman and Mahon at the University of Maryland have used a longitudinally pumped single-mode system (as the first stage of a frequency multiplication scheme generating Ly-α, T J McIlrath, private communication).

One problem at the output powers needed for work on plasmas is burn damage to the intra-cavity Fabry-Perot etalons. With suitable systems, this can be overcome by using two flash-lamp pumped lasers in tandem as an oscillator-amplifier system. Such a system, due to J Salter in my group at Imperial College, is shown in Figure 7. The (tuned) oscillator stage in this case puts out about 50 kW in a 300 ns pulse into a 0.3 Å bandwidth, whilst the amplifier stage provides a gain of about 10 to a total narrow-band output of 0.5 MW. The basic design of home-constructed co-axial flash-lamp laser we use - which is very similar to many other systems described already in the literature - is shown in Figure 8. Further details are given in Skinner (1974).

5. EXPERIMENTAL APPLICATIONS OF TUNABLE LASERS TO PLASMAS

In this section a brief description will be given of some recent applications of tunable lasers to plasma spectroscopy, with a considerable bias towards the work of the author's own group at Imperial College. One or two particularly interesting future applications which have been discussed theoretically but not yet exploited in practice will also be mentioned.

5.1 Applications to Orthodox, 'Linear', Spectroscopy

It was already mentioned in Section 4.1 that the simplest properties of lasers, i.e. high intensity and good beam quantity, in themselves offer important advantages in plasma work even if the laser is only used for orthodox (linear) absorption spectroscopy. In fact, until the advent of tunable lasers, only very rarely indeed was it possible to use any technique other than straight-forward emission spectroscopy, because of the high intrinsic plasma brightness. The limitations of emission spectroscopy are the lack of control over signal levels, and the fact that available optical paths are small (many laboratory plasma sources having dimensions of only a few centimetres at most), so that many interesting features remain too weak to be observed. These difficulties in emission work (which still constitute over 99% of all published plasma spectroscopy) and the consequent limitations on accuracy have often been the prime reason why important theoretical problems have remained unsolved.

Tunable lasers of even a very modest kind (e.g. CW dye lasers) are immediately bright enough to overcome the plasma background problem in nearly all sources (although of course restrictions on available wavelength range remain). The remaining difficulty is then the low optical depth per unit path described in Section 4.3. These can be overcome by multiple-passing (although , because of the refractive index of a plasma, beam shearing problems due to small density gradients should not be underestimated even in quite quiescent sources), and/or by the use of special purpose plasma sources. (However, it has also to be realised that the type of problem which can be tackled by a long path approach may be limited by time response problems. Figure 4 shows that if one wishes to study transient effects due to collisions for the Balmer series the maximum optical path over which a probe signal can be studied without effectively time integrating is often only a few centimetres, unless of course pump and probe signals can be made to travel at the same velocity throughout).

We have built a special purpose plasma source at Imperial College specifically for tunable laser spectroscopy, consisting of 10 z-pinch devices of alternating polarity in sequence, which gives us a plasma of column length 10 metres and diameter 16 centimetres. This machine is designed to be used partly in laser pumping and wave-mixing experiments, see below, but a major application is in absorption spectroscopy of very weak features. (Some of this work, in application to plasma satellite features in thermal plasmas, and to quasi-molecular satellite features in He perturbed by He^+ is discussed in the contributed paper in this symposium by myself, Kolbe and Playford). Presently, we have achieved total optical paths of 180 metres by multi-passing, the limit being set partly by optical considerations and partly by restrictions of our present (home-built) CW jet-stream dye laser. To date we have studied H and He line profiles, and also continuum absorption (using an Ar^+ laser as a background source), most of which work is still being written-up for publication (see Burgess, Kolbe and Playford 1978). An example of a pressure broadened profile of H-alpha, obtained with a 10 pass set-up (total optical path of 100 metres) is shown in Figure 9. With these techniques we have been able to study features out to a few x 10^{-6} of line-centre intensities. Put another way this equivalently means we could detect a level population of only 10^5 atoms/cm^{-3} or 1 part in 10^{10} of the plasma electron density. At the moment we are setting up an experiment to attempt a direct measurement of the magnetic dipole transitions out of the ground term of N II at 6548.1, 6583.6 Å ($A_{21} \simeq 0.001$, 0.003).

5.2 Fluorescence Scattering Observations of Collision Rates

The most widely reported application of tunable lasers to plasmas has been the study of collision processes via laser-induced fluorescence. Collins, Johnson and Shaw (1972) used such methods on a flowing afterglow in order to study collision processes for He (5^3P), and in a subsequent paper Collins and Johnson (1972) also studied He molecules. They observed a transient spike, similar to that shown in Figure 5, i.e. as observed and predicted by Burgess and Skinner (1974), but alternatively interpreted this in terms of co-operative radiation processes between separate atoms rather than simple collisional effects. Burrell and Kunze (1972), working with a short-pulse laser and powers below saturation carried out time-delay measurements, similar to those used in orthodox fluorescence studies of neutral vapours, to study collision processes between high states of He in a plasma. Recently, Himmel and Pinnekamp (1977) again used short pulse laser techniques (below saturation) to study collision rates for n=3, 4 and 5 in a very low density, cool, hydrogen plasma.

One of the problems of short pulse techniques is that information is only obtained on collision rates for the upper state of the pumped transition. However, by working with long, saturating pulses one can obtain information also on the lower pumped state. As pointed out by Skinner and myself (Burgess and Skinner 1974), this allows measurements via purely visible spectroscopy of rate processes for the upper states of VUV resonance lines.

We have applied this technique to rather higher density plasmas (10^{14} cm^{-3} < n_e < 5 x 10^{15} cm^{-3}) than those in the other papers cited, and have studied transitions in helium (Burgess and Skinner 1974; Skinner 1974) and hydrogen (Skinner 1974; Burgess, Kolbe and Ward 1978; Ward, Burgess, Skinner and Myerscough - to be submitted to J.Phys.B.). An example of fluorescence, resonantly pumping on H-alpha, is shown in Figure 10, see Skinner (1974, Ph.D thesis, University of London). As can be seen, this has the general form predicted by equation (7) above. However, there are very serious problems in interpreting such results in terms of the expected electron collision rates in hydrogen at these densities. Skinner in his thesis applied the simple model described in Burgess and Skinner (1974), and already concluded that effective collision rates in the plasma were lower than expected. Subsequently, to avoid the approximations inherent in a 2-level model, we have used Dr Valerie Myerscough's many level, time-dependent, collisional radiative code already mentioned in Section 4.2.4 (see also McIlrath, Myerscough and Koopman 1978) in the interpretation of a long series of laser-saturated fluorescence studies on H-alpha and H-beta. These show conclusively that under our plasma conditions ($10^{14} < n_e < 5 \times 10^{15}$ cm^{-3}, 0.4 eV < T_e < 1.0 eV), experiment and theory cannot be reconciled unless usually adopted collision rates are reduced by up to an order of magnitude. Using the established values there is good agreement for the initial transient peak, but the subsequent decay is much slower than predicted, and the final equilibrium plateau is far too high. (Notice, that from the arguments of Section 4.2.3. this immediately establishes something about the relative value of the rates into the upper and lower levels of the pumped transition). This is not the place to discuss the interpretation in detail - suffice it to say that one cannot explain the results either by invoking technical problems (optical depth, time-response, etc), nor by other laser-induced processes (photoionization, non-linear effects, Rayleigh scattering, etc).

5.3 'Double-Resonance' Techniques

It has already been mentioned that in laser-pumping in a plasma it is always easier to produce a long-term change for the lower level of the pumped transition than for the upper one. This suggests that one should probe in absorption on one transition (always remembering the optical depth limitations described in 4.3), whilst pumping on another line with a common lower state, thus bearing obvious relation to the classical techniques of optical double resonance.

We have made such observations on a 75 cm plasma column, pumping with one laser on H-alpha and probing with another on H-beta, and these are described in Burgess, Kolbe and Ward (1978). We have also made preliminary observations on He, pumping on $2^3P - 4^3D$, 4471 Å and probing on $2^3P - 3^3D$ 5876 Å using the full 10 metre plasma column (G Kolbe, unpublished). The set-up for the hydrogen observations is shown in Figure 11.

The results of such work are fully described in Burgess, Kolbe and Ward (1978). What needs mention here is that one advantage of the technique is that it becomes possible to prepare a plasma with only one level population significantly perturbed from its equilibrium value. This contrasts with the inevitable situation in fluorescence scattering, and is a great help in interpretation since multi-level theoretical models are not needed. The trick is to wait long enough after the saturating pulse is cut-off by the Pockel's cell. The lower pumped level then remains far from equilibrium, whilst the population lost from it is rapidly shared amongst many higher levels which hence return to very close to their equilibrium values.

Full interpretation is given in the paper referenced. Once again, as with the fluorescence work discussed in 5.1, we see large departures from predicted collision rates and this is shown for n=2 in figure 12.

5.4 Laser 'Burn-through' Experiments

Given a plasma column long enough to have measurable optical depth, a very simple method of measuring collision rates becomes possible. This is simply to measure the transmission of the plasma column as a function of input laser intensity and hence, by observing the eventual saturation, deduce the competing collision rate. (This, for once, is a rather simpler technique to interpret on a plasma than on a neutral gas because of the high collision rates and large homogeneous line-widths). Skinner (1974) first made such observations in helium, and we have since applied them to measurement of collision rates in hydrogen. An advantage of the technique is that (for the long-time behaviour during a laser pulse) the rate that matters is only that between the levels of the pumped transition themselves (see 4.2.3). Once again, in a hydrogen plasma we find this rate to be much lower than predicted (see Burgess, Kolbe and Ward 1978).

Such a procedure offers a very simple means of studying a wide range of rate processes - note that the upper level population does not necessarily have to change at all for the measurement to succeed (see 4.2.3 above). The major difficulty in terms of really quantitative application is to have adequate knowledge of the spatial intensity profile of the input laser beam - most long-pulse dye lasers run multimode. One needs a high-power (1 MW), single spatial mode output of long (> 100 ns) duration. The best way to achieve this seems to be with a suitable flash-lamp dye laser pumped dye laser - see e.g. Fielding (1974) - but using longitudinal pumping of the final oscillator stage. We are constructing such a laser specifically in order to continue our collision rate determinations via 'burn-through' measurements using the 10-metre plasma column.

5.5 Diagnostic Applications to Toroidal Magnetically-Confined Plasmas

The most straightforward application of tunable lasers to toroidal plasma experiments, particularly Tokamaks, is to the determination of neutral hydrogen densities, which in some machines are quite high (10^7 - 10^9 cm^{-3}) despite the elevated electron temperatures (0.5 - 1 keV). The neutral hydrogen concentration is of interest both in studying the re-cycling of the plasma from the walls (in most Tokamaks at present the containment time is less than the plasma duration) and also possibly in experiments using neutral beam heating (e.g. DITE at Culham).

As far as I am aware, the first attempt to use a dye laser to measure neutral hydrogen concentrations was by Skinner, Fielding and myself on the CLEO device at Culham in 1972-73 (see C Skinner, Ph.D thesis, University of London 1974). However, as these experiments were unsuccessful (for lack of machine time), I suppose the usual rules mean one should not mention them! Burakov et al, 1977, however, have succeeded in making H-alpha scattering observations on a Tokamak in order to measure neutral hydrogen distributions.

The major problem with H-alpha scattering is the low enhancement possible (because of the high temperature) and hence the limitations on possible spatial resolution. One way to overcome this would be to use a laser operating on Lyman-alpha. For this reason two groups (McIlrath at Maryland and Fielding at Culham) are presently building frequency up-conversion systems (the Maryland group has one operational), and Professor D J Bradley and Dr M H Hutchinson at Imperial College are building an argon-excimer laser for application to Tokamak diagnostics. A further advantage of operating on Lyman-alpha is that theoretical interpretation in terms of the underlying neutral population is more direct (although this is not necessarily as absolute as might seem at first sight) - see McIlrath, Myerscough and Koopman 1978.

There is another interesting prospect for resonance scattering diagnostics beyond measurement of neutral concentrations, and that is the possibility of spatially resolved measurements of magnetic field directions (in a Tokamak, the field magnitude is usually known to reasonable accuracy *a priori*). One way of doing this, using Thomson scattering, has already been mentioned in Section 3 (see also Forrest et al 1978). The alternative, using fluorescence scattering, which has not yet been achieved, (in contrast to the Thomson scattering technique), is a very simple application of one of the effects first described by Hanle. If an incident laser beam is polarised with its E-vector pointing in the direction of the observer, in the absence of any magnetic field no scattering would be observed parallel to the E-vector from a classical dipole excited by the E-field. In the presence of a magnetic field the scattered radiation would be polarised perpendicularly to the magnetic field (the dipole precessing around the latter). Observation of this polarisation direction (spectrally unresolved!) then would give a very simple measurement of the magnetic field direction. I first proposed this technique in application to Tokamak plasmas in 1970 (D D Burgess, EMR agreement CUL845 with Culham Laboratory), and it is further described in C H Skinner's subsequent Ph.D thesis (University of London 1974). A major problem is that atomic hydrogen is <u>not</u> a classical dipole - the spin-orbit interaction causes depolarisation of the fluorescence. We studied the consequences of the depolarisation effect theoretically for plasma applications at both low and high incident laser intensities. (An even more serious problem for hydrogen at Tokamak densities however is probably depolarisation due to proton collisions - V Myerscough, private communication). A better prospect would be to seed the plasma with helium and pump on a transition with a ^1S ground state (e.g. $2^1S - 3^1P$, 5016 Å), and this still seems a feasible way of determining magnetic field directions.

Finally, one should note that the temporal behaviour of the fluorescence during a long-lasting laser pulse is a unique function of both n_e and T_e, see equation (7). This could have useful diagnostic applications in some circumstances.

5.6 Experiments Involving Mixing of EM radiation with Longitudinal Plasma Waves

Two classes of experiments involving coupling with plasma waves can be distinguished, those involving fully-ionized plasmas, and those involving coupling via bound electrons in partially-ionised plasmas. Quite a lot of work has already been done on the first type, but the second may offer extra possibilities because the coupling is much stronger (wave-vector matching being eased by the centre-of-mass recoil of the atom), and hence thresholds for non-linear effects can be much lower.

With the advent of very high density plasmas, direct coupling of laser radiation into plasmas, via resonance at the electron plasma frequency, with the input laser frequency, has become quite widely studied (see e.g. Peacock et al 1977) and has already been discussed in Section 3. However, such coupling is also possible in lower density systems via the mixing of two laser beams in the plasma, separated by ω_p. This possibility was first pointed out by Kroll, Ron and Rostoker (1964). Experimentally the technique places very tight constraints on the <u>angular</u> relationship of the two input laser beams, because of the great difference between the dispersion relation for plasmons and that for photons, and the consequent difficulties of phase-matching. Nevertheless, such an experiment has been successfully carried out using two dye lasers tuned ω_p apart by Stansfield et al (1971). It should be noticed that optical generation of plasmons in such wave-mixing experiments in fully-ionised plasmas does not become self-stimulated (to the order described by Kroll et al 1964) because, essentially, of the very low plasma frequency compared with that of the two input laser beams.

An alternative is to use coupling of EM radiation and plasma waves via bound electrons. Such coupling, in the form of emission plasma satellite lines near allowed and forbidden transitions has been known for some time (see Paper I and Kunze and Griem, 1968). The possibility of stimulated plasmon Raman scattering via bound electrons, with a threhold at which the plasmon production becomes non-linear (which can be used to measure the effective damping rate for Langmuir waves), was first pointed out by Burgess, Richards and Mahon (1971). Related calculations have also recently been reported by Bretagne and Godart (1978) who claim that the threshold for self-stimulated plasmon production will be much higher than our original estimate. The difference, however, stems almost entirely from estimates of the initial population of the pumped transition - Bretagne and Godart interpreting our quoted value as being a theoretical over-estimate (e.g. the L.T.E. value being much lower) whereas actually our value was based on measurements in a recombining Helium plasma, where the first-excited states are overpopulated by several orders of magnitude.

Such non-linear effects should, therefore, still be observable by a suitable choice of plasma, with existing dye lasers (e.g. providing fluxes in the plasma of a few tens of Megawatts per cm^2). However, since the whole possibility of self-stimulated plasmon production stems from a large initial population difference of the initial and final atomic levels of the pumping scheme, such measurements will be limited to rather low temperature plasmas (until VUV tunable lasers become widely available). A major difficulty in such experiments is not so much that of pumping, but that of detection, for the reasons already discussed in Section 4.

5.7 Other Plasma Wave-Mixing Possibilities

Once one realizes that atoms in a plasma emit and absorb plasmons (quanta of electron waves) as much as they do photons (indeed rather better - see Alekseev and Nikitin 1966) a whole range of possible wave-mixing experiments seems feasible analogous to established 4-wave mixing processes with photons, such as CARS (Coherent Anti-Stokes Raman Spectroscopy - see e.g. Levenson, 1977). Considerable theoretical consideration is needed once one or more of the waves involved are longitudinal plasma oscillations - particularly over questions such as coherence (the plasma waves always containing a very high density of modes) and wave-vector matching. However, experimentally in a plasma one has e.g. the advantage of there being two opposing contributions to the overall refractive index of opposite sign, that due to the free electrons being always negative for transverse waves, which hence can be adjusted (relatively) by suitable choice of conditions. It would seem that coherent interactions of EM radiation with plasma modes via bound electrons pose a wide, and as yet relatively untreated, field for study. Conceivably, this could be of more than academic plasma physics significance as studies of possible x-ray lasers in ionized media develop, where stimulated cross-coupling to low frequency plasma modes could cause difficulties.

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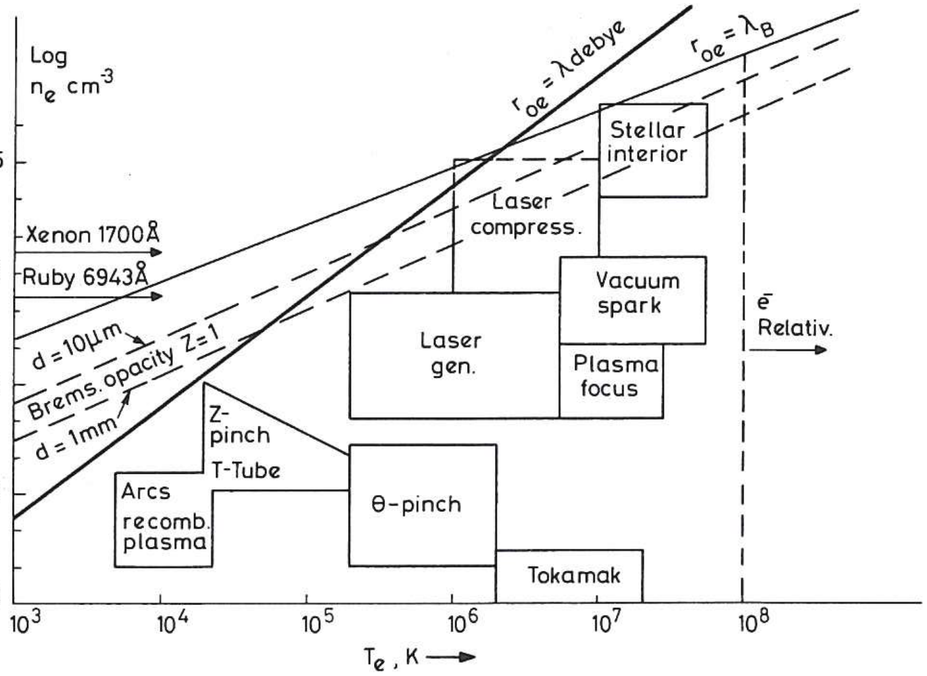
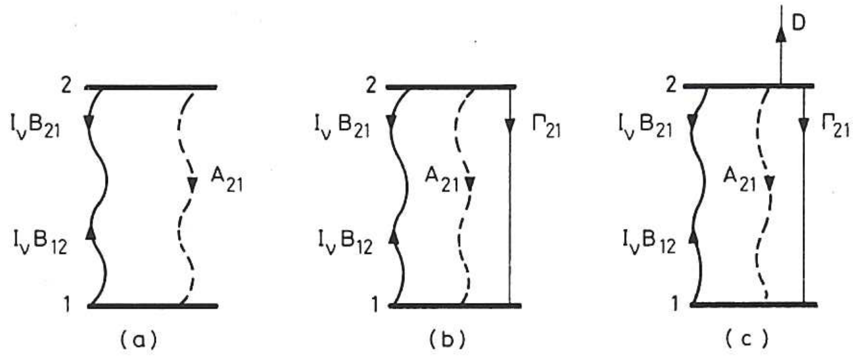


Fig.1 Laboratory plasma sources.



a) Power to saturate.

$$P = 8\pi \Delta\nu \frac{h\nu^3}{c^2}$$

(b) Power to saturate.

$$P = 8\pi \Delta\nu \frac{h\nu^3}{c^2} \frac{(\Gamma_{21} + A_{21})}{A_{21}}$$

(c) Power to saturate.

$$P = 8\pi \Delta\nu \frac{h\nu^3}{c^2} \frac{(\Gamma_{21} + A_{21})}{A_{21}}$$

(c) Power to transiently enchance upper level.

$$P' = 8\pi \Delta\nu \frac{h\nu^3}{c^2} \frac{D}{A_{21}}$$

Fig.2 Laser-pumped atom.

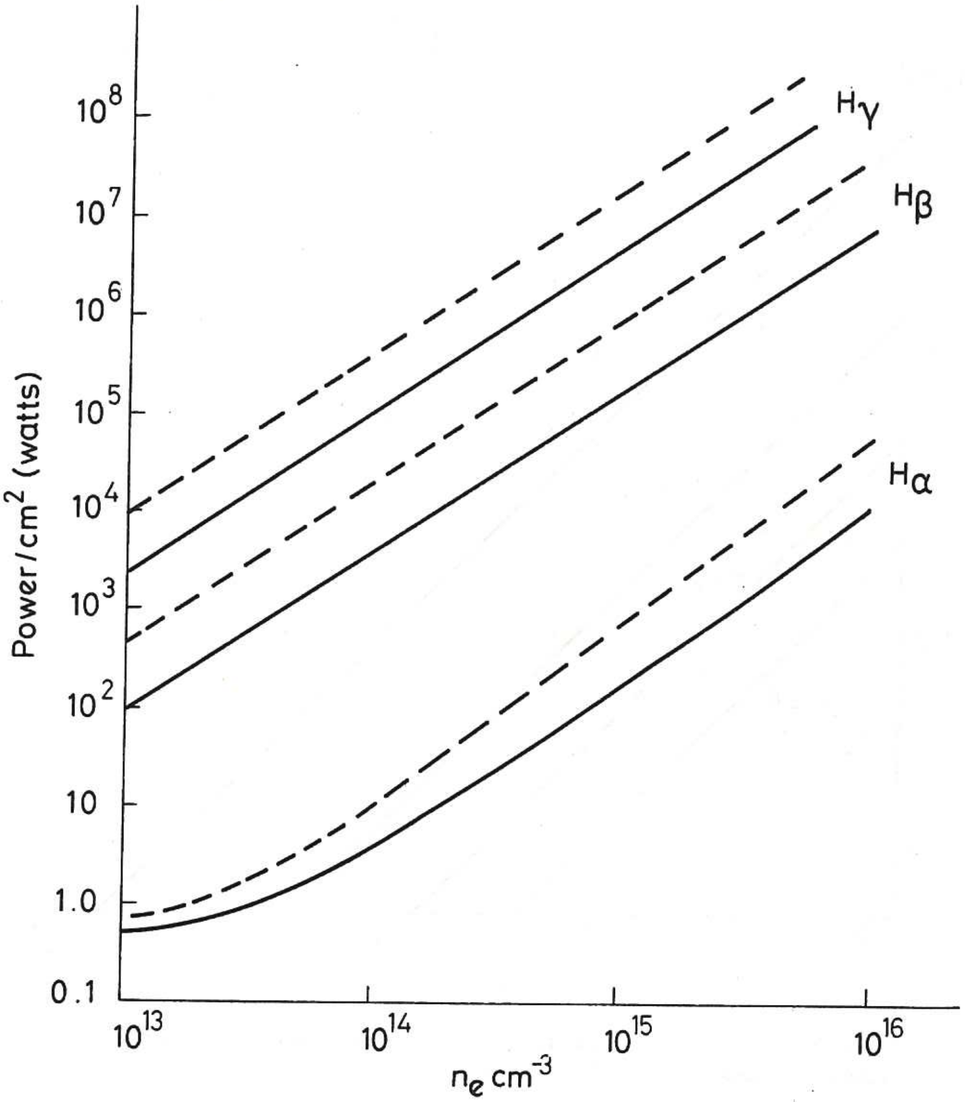


Fig.3 Laser power to saturate Balmer series. ———— = flux (into line half-width) to saturate. - - - - - - - = flux to cause significant enhancement of upper level population. (Based on collision rates of Johnson 1972, line widths from Griem 1974.)

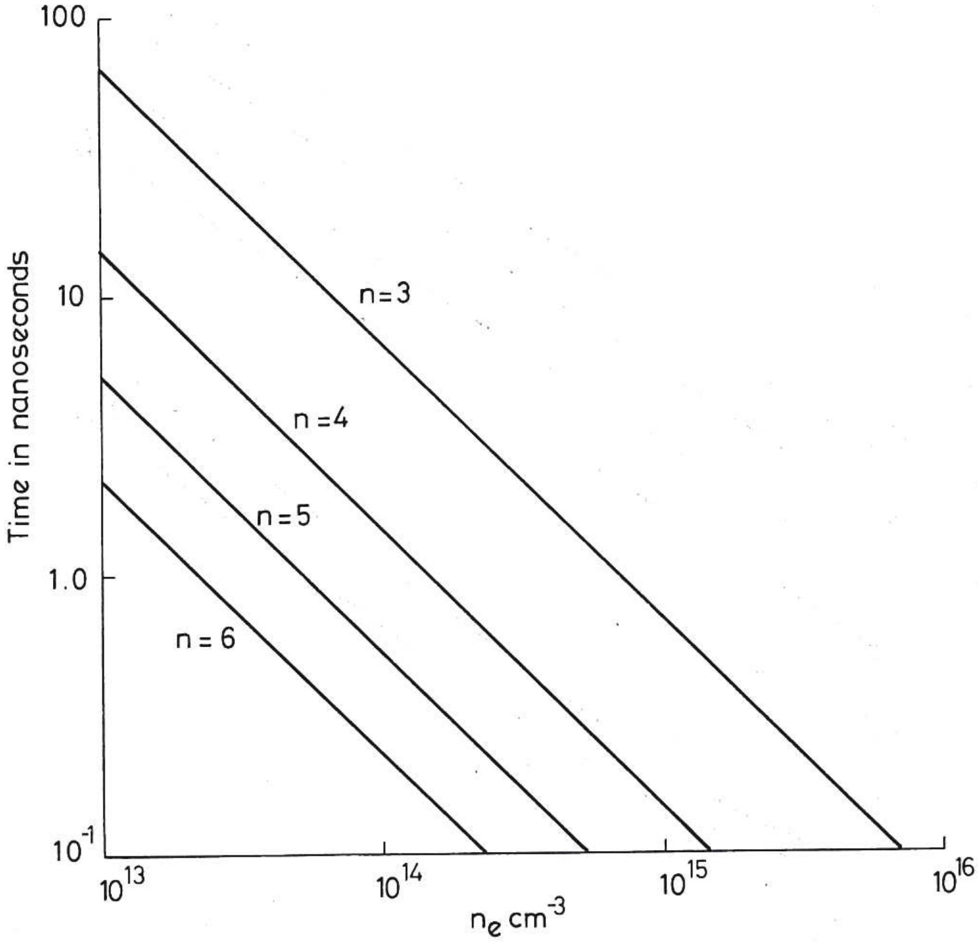
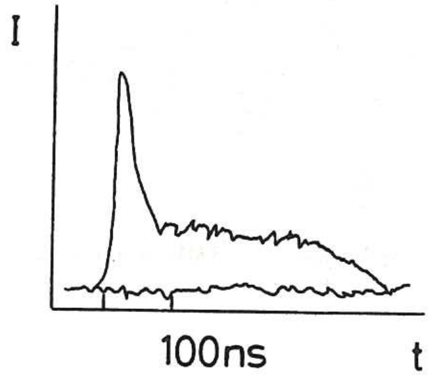
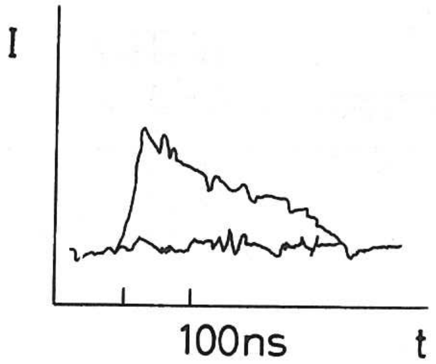


Fig.4 Decay times for transient fluorescence enhancement, Balmer series. (Based on collision rates of Johnson, 1972).



He I $2\,^3P - 3\,^3D$
5876 Å fluorescence

Fig.5



He I $2\,^1P - 3\,^1D$
6678Å fluorescence

Fig.6

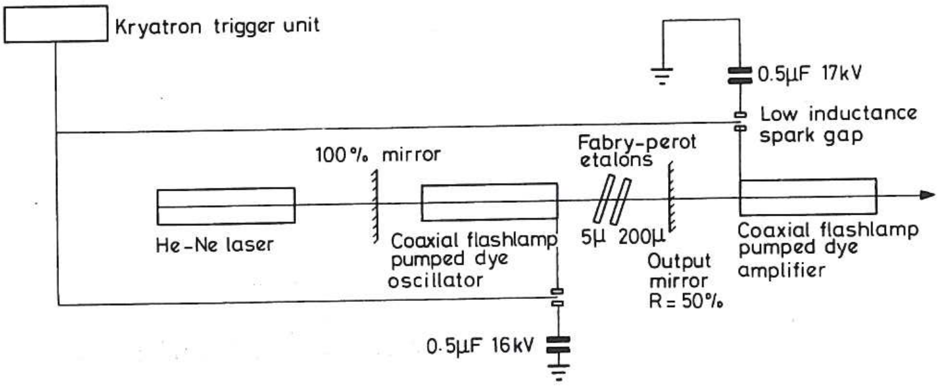


Fig.7 Oscillator-amplifier flash-lamp pumped dye laser (>0·5 MW into 0·3 Å).

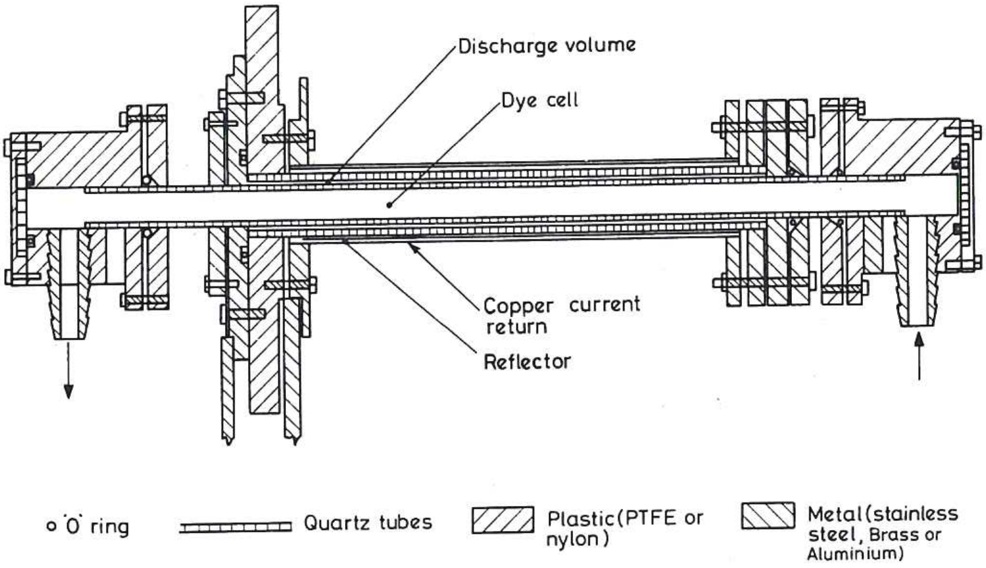


Fig.8 Coaxial laser.

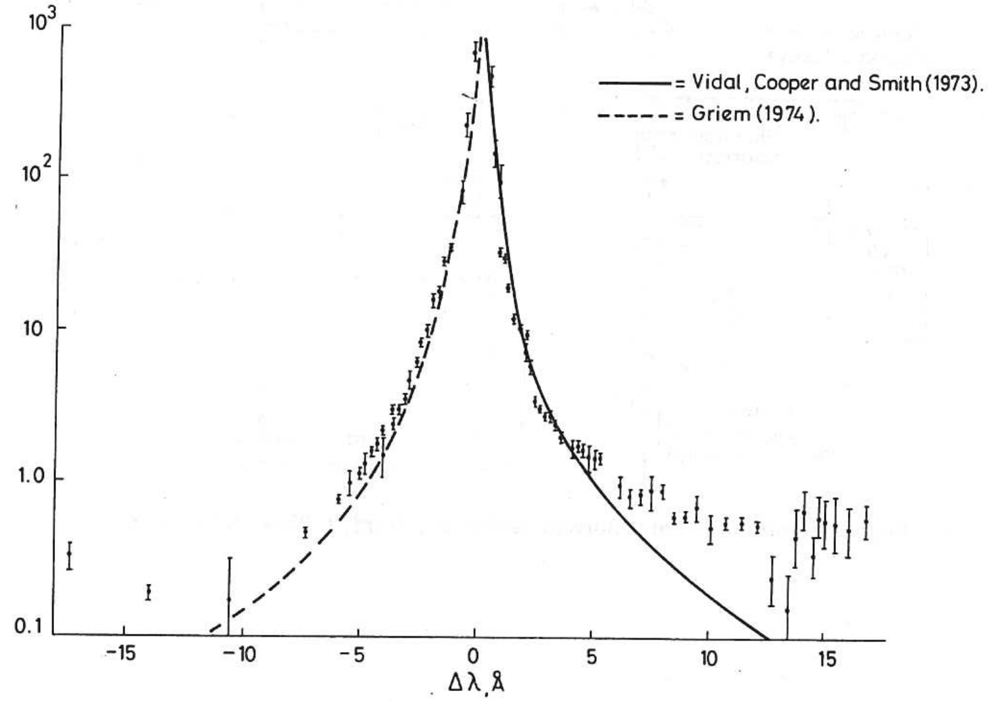
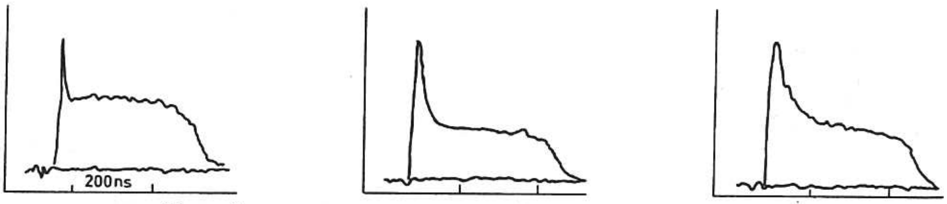


Fig.9 H-alpha absorption profile / 100 metres path, $n_e = 10^{15}$ cm^{-3}, $T_e = 10^4$ K.



$n_e = 0.65 \times 10^{15}$ cm^{-3}
$T_e = 0.7$ eV

$n_e = 0.5 \times 10^{15}$ cm^{-3}
$T_e = 0.5$ eV

$n_e = 0.4 \times 10^{15}$ cm^{-3}
$T_e = 0.4$ eV

Fig.10 Hα, laser-saturated fluorescence (see Skinner C.11., 1974).

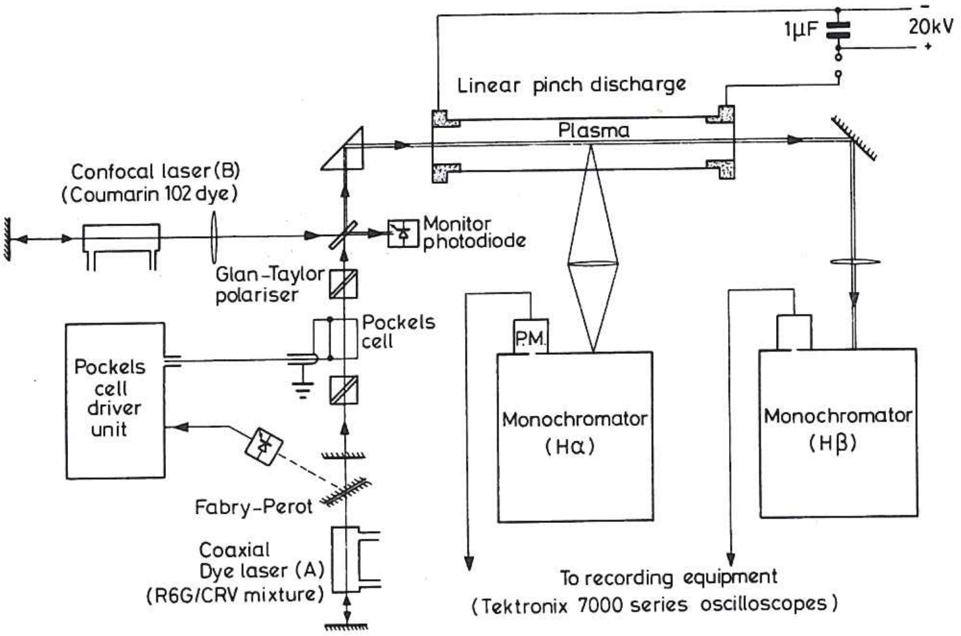
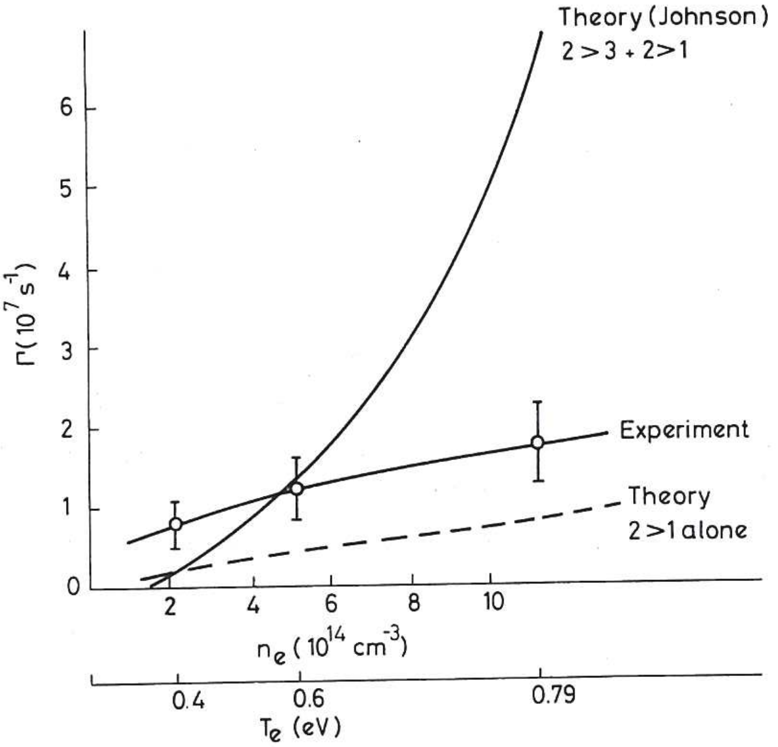


Fig.11 (Reproduced from Burgess, Kolbe, and Ward, J. Phys., B11, 1978).



(Reproduced from Burgess, Kolbe and Ward, J.Phys B11, 1978)

Fig.12 Decay out of n = 2.