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A pulse mass spectrometer for studying  
fast phenomena in gases at low pressures  
and in non-stationary plasmas  
TECHNIQUE AND APPARATUS

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Culham Laboratory,  
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1965

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A PULSE MASS SPECTROMETER FOR STUDYING FAST PHENOMENA IN  
GASES AT LOW PRESSURES AND IN NON-STATIONARY PLASMAS

Technique and Apparatus

by

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A B S T R A C T

A time-of-flight pulse mass spectrometer is described - known as a 'Chronotron' - for the analysis of neutral gas over a range of pressures from  $1.10^{-10}$  to 10 Torr, or of ions in a non-stationary plasma with an ion concentration of  $1.10^6$  to  $1.10^{16}$   $\text{cm}^{-3}$  without magnetic field, or in a strong magnetic field. Processes can be investigated of duration from  $1.10^{-7}$  sec. to continuous. Examples are given of applications of the method to the analysis of currents of molecules desorbed from solid surfaces, a study of the mechanism of sorption and of catalytic reactions on renewed surfaces using stable isotopes, the interaction of a hydrogen plasma and the walls of vacuum systems, the plasma in pulsed discharges and the passage of molecules through a highly ionized plasma.

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## 1. INTRODUCTION

In electronics, plasma physics and vacuum technology it is often necessary to study fast processes associated with time-dependent changes in the composition of gases, plasmas or of neutral or charged particles during their transition from the bulk to the surface of a material or in the opposite sense. Such processes include, for example, pulsed discharges in experimental thermonuclear devices, breakdowns in vacua and in gases at low pressures, the evolution and absorption of gases in vacuum systems in non-stationary conditions, and sorption- and catalytic effects in dynamic systems involving high and ultra-high vacua.

An effective method for studying fast processes is pulse mass spectrometry. Mass spectrometers designed for this purpose have to meet two specific requirements:

- (a) The analysis time over the whole mass spectrum under investigation must be short, in view of the short duration of the phenomena to be analysed.
- (b) The sensing unit of the spectrometer must not introduce changes in the process under investigation or in the composition of the analysed medium, even if this contains unstable molecules (such as radicals) or charged particles.

The first requirement, over a wide range of durations of the investigated effects, is met very adequately by the time-of-flight mass spectrometers having circuits ensuring the recording of the spectrum at the required time intervals. A theory of the ion motion in time-of-flight type mass analysers is being developed. The time-of-flight mass spectrometers described in <sup>(1)</sup> are not specifically designed for the study of fast phenomena.

To satisfy the second requirement, the sensing unit (chiefly the ion source) must be specially designed. It must be placed directly into the investigated medium to enable the analysed molecules or ions to reach the analysing region without experiencing previous collisions with the walls. Specially designed analysers have to be used for different ranges of gas pressure or plasma concentrations to be analysed. It is in this respect that spectrometers for the analysis of fast phenomena differ from mass spectrometers for analysing gases in which the optimum operating conditions are set up by using the inlet system.

For the investigation of vacuum problems arising in devices for studying plasmas and the physics of electrical discharges in vacua, a range of time-of-flight analysers has been developed designed for analysing neutral gases over a wide region of pressures, and for the analysis of ions in a non-stationary plasma. A recording circuit developed simultaneously permits the recording of the spectral over a wide region of durations. The complete system was given the name 'Chronotron', and is being manufactured industrially as model 'MSCH-3'<sup>(2)</sup>.

The results of several investigations carried out using the 'Chronotron' have been published in (3) and (4). In this paper the analyser and ancillary equipment are briefly described, and also a technique for studying fast processes, such as occur in various branches of electronics and vacuum technology.

## 2. THE 'CHRONOTRON' PULSE MASS SPECTROMETER

Three different types of neutral gas analyser for different ranges of pressure were designed and two types of ion analyser, one for operating in a volume without external magnetic poles, the other for operating in a strong magnetic field.

The sensing unit of the mass spectrometers for neutral gas analysis is formed by a pulsed ion source in which the analysed molecules are ionized by bombardment with an electron beam, and ion bunches are formed which are then separated according to their time of flight in the drift region. The sensing unit of the spectrometer for ion analysis consists of a system of diaphragms and an ion shutter where the plasma electrons and ions are separated and ion bunches are formed. The analyser is shown schematically in Fig.1.

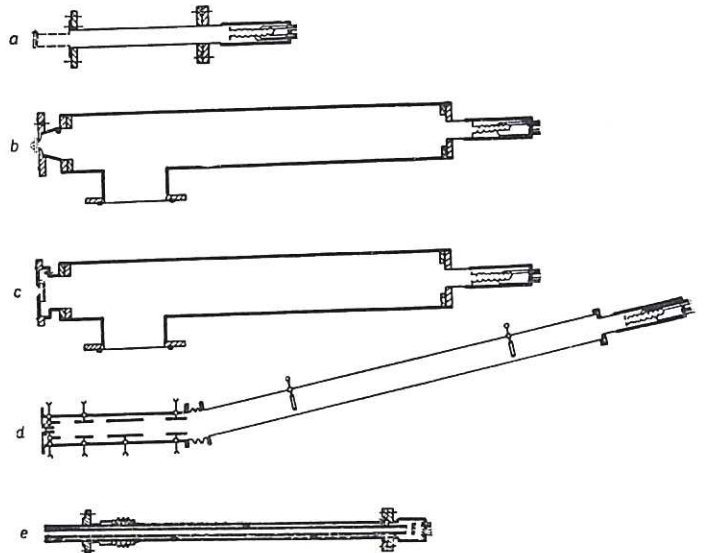


Fig.1

(CLM-TRANS 2)

The five analysers: A-neutral gas, at pressures from  $1.10^{-7}$  to  $1.10^{-5}$  Torr, B-neutral gas, at pressures from  $1.10^{-5}$  to  $1.10^{-2}$  Torr, C - neutral gas, at pressures  $1.10^{-2}$  to 10 Torr, D - ions in non-stationary plasma without magnetic field, E-for ions in a plasma located in a strong magnetic field

### A. Neutral Gas Analyser for Pressures from $1.10^{-10}$ to $1.10^{-5}$ Torr

The ion source is of the transverse type, the direction of the electrode beam forming a right angle with that of the ions. The source has a strip cathode giving currents up to 1A. The voltage pulses forming the electron beam are applied to electrode 1, those forming the ion bunches to the grid 2 (Figs.1(a) and 2). The source is located in the vacuum

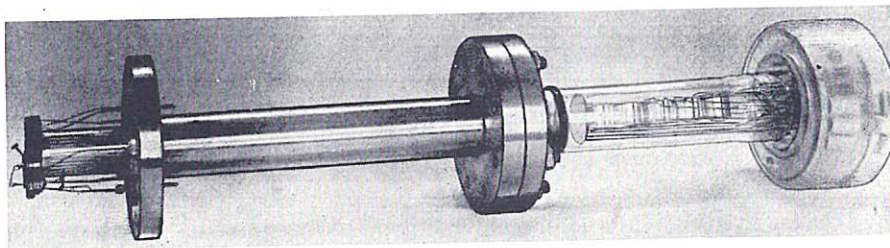


Fig.2 Analyser Type A (CLM-TRANS 2)

system in which the analysis takes place. The open source design permits free access of the molecular beams. The collimated molecular beams can be analysed by being fired through the source without impinging at the electrode. An open electron multiplier with box-type dynodes is used as an ion detector. The length of the drift space is 40 cm. The analyser is designed for operation in ultra-high vacua, and can be heated to 500°C. Depending on the object of the analysis two methods of ion bunching can be used. If, in addition to mass analysis, it is also necessary to measure the initial velocity of the neutral molecules (such as when studying cathodic sputtering) ionization is accomplished by a brief ( $1 \cdot 10^{-7}$  sec.) electron current pulse and the ions are accelerated out of the source by a constant electric field. The ions leaving the source retain with considerable accuracy the initial velocity distribution of the neutral molecules, which is determined at the detector from the shape of the bunches. For simple mass analysis a much higher accuracy and resolving power can be obtained by using pulses of the extracting voltage to shape the ion bunches, by bunching the ions in the ionization region, and by the 'space-time' focusing of the bunches<sup>(1)</sup>. With this method the duration of the electron pulse is larger than the time during which the ions are kept in the electron beam without experiencing the effect of the extracting pulse ( $\sim 2 \cdot 10^{-6}$  sec.); the ions are extracted by a pulse with a steep front and a duration exceeding the time during which the ions of larger mass are retained in the ionization region ( $2 - 5 \cdot 10^{-6}$  sec.).

The lower limit of the analyser working region is determined by the fact that at low pressures, when using pulsed ionization, the bunch only contains a small number of ions which leads to fluctuations in the peak heights on the mass spectrum. For analysis at pressures of the order of  $1 \cdot 10^{-10}$  Torr the electron pulse current must be a few hundredths of a milliamp. The number of ions  $N_i$  in the bunch is then given by

$$N_i = \frac{1}{e} J_e \Delta t n_0 \sigma_i d ;$$

where  $J_e$  is the electron beam current,  $\Delta t$  the time the ions are exposed to the electron beam,  $n_0$  the analysed gas concentration,  $\sigma_i$  the ionization cross section, and  $d$  the effective length of electron beam. When analysing hydrogen at a pressure of  $1 \cdot 10^{-10}$  Torr one has for an electron current  $J_e = 1 \text{ A}$ ,  $d = 2 \text{ cm}$ ,  $t = 2 \cdot 10^{-6} \text{ sec.}$  and  $N_i = 7 \cdot 10^3$ .

The upper limit of the pressure range is determined by scatter and the multiple ionization of the ions in the residual gases. The upper limit of the linear analyser characteristic is  $1 \cdot 10^{-5}$  Torr.

#### B. Neutral Gas Analyser for Pressures from $1 \cdot 10^{-5}$ to $1 \cdot 10^{-2}$ Torr

The ion source is of the longitudinal type, the directions of the electrons and ions being parallel. The source is located in the system in which the gas is analysed (Fig.1(b)).

The drift space and the collector have a separate pumping system and diffusion pump with a pumping speed of 500 litre/sec. The ion bunch has a small cross section ( $0.3 \times 5$  mm) and the bunches pass from the source into the drift space through a slit in the diaphragm separating the working region from the analyser pumping system. At the maximum operating pressure ( $1.10^{-2}$  Torr) the pressure does not exceed  $1.10^{-5}$  Torr; the pressure drop takes place over a length of about 1 cm. The ion detector is again formed by an electron multiplier. A pulsed ion source is used, with pulsed extraction and 'space-time' focussing. In a longitudinal type source, since the conditions for 'space-time' focussing are less favourable than in a transverse type source, the drift space is 1 metre long.

The upper limit of the operating region is determined by the non-linear analyser amplitude characteristic. The departure from the linear characterisation at high pressures is due to space charge effects on the ions in the source.

#### C. Neutral Gas Analyser for Pressures of $1.10^{-2}$ to 10 Torr

In this region of pressures the analyser cannot be placed directly into the system in which the analysis takes place, since no linear amplitude characteristic could then be obtained. The transverse type ion source is therefore placed behind an interchangeable diaphragm containing an aperture situated in the molecular gas beam ejected from the operating region (Fig.1(c)). The cathode of the source is placed close to the diaphragm (2 mm) and constitutes the source of a flat electron beam parallel to the plane of the diaphragm. The molecular beam of analysed gas intersects the electron beam through a solid angle of  $1.8\pi$ . The drift space is evacuated by a diffusion pump with pumping speed 500 litre/sec. The operating pressure region is split up into three smaller separate ranges for each of which a given size of diaphragm aperture is used. The aperture size is chosen such that at maximum pressure the mean free path of the analysed molecules is larger than the aperture diameter or the slit height. When this condition is fulfilled, the analysis of fast phenomena takes place without inertia (maximum lag  $10^{-5}$  sec.) and the composition of the analysed gas in the source does not change even if it contains unstable molecules.

The upper limit of the operating region (10 Torr) is given by the size of the smallest diaphragm aperture, which is 5 microns. The lower limit is imposed by the necessity of maintaining a certain ratio between the analysed gas concentration in the molecular beam and that in the drift space (where it should be at least two orders higher).

#### D. Analyser for Ions in a Non-Stationary Plasma

The sensing unit consists of a system of two apertured diaphragms of which the first is kept at the potential of the walls of the vacuum system containing the plasma, the second



at the negative potential which removes the ions from the plasma surface (Fig.1(d)). Both apertures must be smaller than the thickness of the plasma boundary layer (which in its turn depends on the Debye radius, the plasma concentration, and the potential of the second diaphragm). The ion shutter operates on the deflection principle and has the shape of a plate condenser between the plates of which passes the beam of accelerated ions from the plasma. To the shutter plates is applied a constant potential which deflects the beam and drives it from the collector. The ion bunches are formed by voltage pulses applied to the shutter plates; the pulses are of constant magnitude and opposite polarity. To suppress separation according to the masses, the shutter plate dimensions are chosen such as to make the time of flight of the ions through the shutter small compared to the duration of the negative pulse ( $2 \cdot 10^{-7}$  sec.). The analyser further includes an electrostatic corrector lens and two electrostatic focussing lenses by which the beam is aligned. The bunch collector is formed by an electron multiplier with box-type dynodes. To prevent the incidence of ultraviolet radiation at the collector, the drift tube of the analyser is suitably curved. In the region of the bend the ion path is curved by an electrostatic deflector system. Only electrostatic lenses are used in the analyser. The trajectories of ions of different mass are the same, so that the information about the mass composition is not influenced.

The upper limit of the plasma concentrations at which an analysis is possible, is determined by the dimensions of the smallest apertures which can be obtained in practice in the (thin wall) diaphragms. For apertures of  $5 \mu$  the maximum plasma concentration just permitting analysis equals  $10^{16} \text{ cm}^{-3}$ . The lower limit in the ultra-high vacuum region is determined, as in the case of neutral gas analysers, by the fluctuations in the peak amplitudes on the mass spectrum for small ion numbers in the bunches. At a plasma concentration of  $10^6 \text{ cm}^{-3}$  and pulse duration of  $10^{-7}$  sec. the total number of ions per bunch is approximately  $10^5$ .

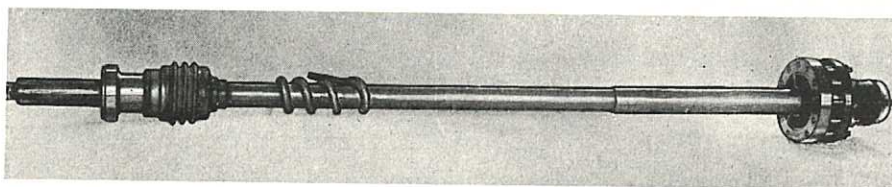


Fig.3 Analyser Type E (CLM-TRANS 2)

#### E. Plasma Ion Analyser for Operation in Strong Magnetic Fields

The analysers with ion trajectories corresponding to the magnetic lines of force can be used to analyse a plasma in a magnetic field of any given field strength. In a homogeneous field the ion trajectories are rectilinear. The sensing units are diaphragms similar to those used in the previous analyser (Figs.1(e) and 3). The ion-shutter, based

on the ion retardation principle, takes the shape of three plane grids; a positive d.c. potential, which slows down the ions to zero energy, is applied to the central grid. Rectangular negative pulses of duration  $1 - 2 \cdot 10^{-7}$  sec. are applied to the shutter.

An open secondary electron multiplier with 'transmitted emission' and with rectilinear electron trajectories following the magnetic field lines is used as detector. When the analyser is used in the homogeneous magnetic field, no shielding from ultraviolet radiation is possible. The range of plasma concentration analysed is the same as for the previous type.

#### Ancillary Equipment to the Chronotron Type MSCH-3

The ancillary equipment has to perform the following functions: to supply d.c. and pulsed voltages to the analyser; to form an image of the mass spectrum on the display screen; to provide a photographic and ciné recording of the spectrum. The equipment is common to all analyser models and permits recording of the mass spectrum over a wide range of durations of the investigated effect. By means of two generators with rectangular pulses with different circuits, pulse durations and amplitudes, the analysers can be used for any of the described applications. In the neutral gas analysers, ion bunching is achieved either by the 'space-time' focussing method, or with continuous or pulsed ionization, and the pulse ionization method with continuous ion extraction. In the plasma analysers, pulsed shuttering is used, in conjunction with pulse synchronisation by an external signal or by periodic pulses. The pulse repetition frequency which determines the periodicity of the separate spectral analyses, is 3 kc/s. For studying fast processes in a neutral gas at pressures of the order of  $10^{-10}$  Torr the high current electron beam (up to 5 A, 600 V) is controlled by means of an impulse generator.

The mass spectrum is recorded by three methods:

- (a) for continuous examination of the spectrum, the display screen is photographed with a ciné camera with a frequency of 32 frames per second and with time markers.
- (b) for simple recording of the spectrum with a time resolution up to  $2 \cdot 10^{-7}$  sec. and a duration of the analysis of  $(4 - 60) \cdot 10^{-6}$  sec., with a possibility of moving the instant of analysis from the start of the effect under investigation, to  $2 \cdot 10^{-4}$  sec. (recorded by taking photographs of the display screen).
- (c) for repeated recording of the spectrum on a single photograph at intervals of  $3 \cdot 10^{-4}$  sec., with a possibility of moving the time of analysis from the start of the effect to  $8 \cdot 10^{-3}$  sec. For the purpose of a qualitative evaluation of the effects being investigated, when their durations are not excessively short, visual observation of the spectrum on the display screen is a convenient method.

The evaluation of the spectrum and calibration according to the masses, are performed for all analyser models by conventional mass spectrometric methods. The most suitable is

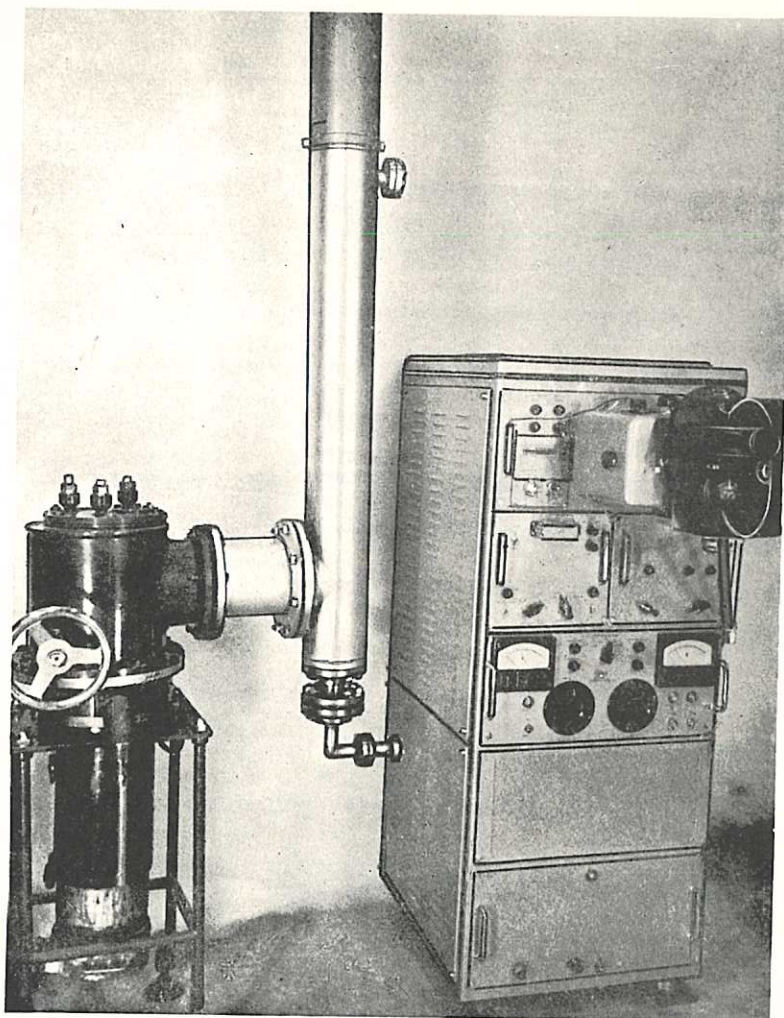


Fig. 4 (CLM-TRANS 2)  
Ancillary equipment of chronotron MSCH-3 and B-Type analyser

the internal standard method. Fig.4 shows the whole ancillary equipment, together with a model B analyser.

### 3. THE PULSE METHOD OF STUDYING FAST PROCESSES

It is not feasible within the limitations of this paper to review in detail the various applications and techniques for using the pulse method in conjunction with time-of-flight mass spectrometers. We have therefore only to refer to three applications:

- (a) Analysis of the composition of a neutral gas, or of charged particles participating in the effect, without influencing the effect itself.
- (b) Introduction into the investigated medium of stable isotopes of those elements contained in the system, and study of their behaviour (method of marked atoms).
- (c) Molecular probing of the medium under investigation that is, introduction into the investigated medium of a beam of particles not affecting the phenomenon, and study of the changes in the composition of the beam due to the effect under investigation. Some examples are also given of applications of the pulse technique to the study of effects varying considerably from one another both in the properties of the medium and in their speed. A detailed account of such phenomena is given in (3) and (4).

(a) Analysis of Currents of Molecular Particles Desorbed from Solid Surfaces

When studying the kinetics of the sorption of gases at low pressures the 'flash filament' method is often used. The pulse mass spectrometric method further increases the advantages of this technique and extends the range of information it supplies.

The measurement is carried out as follows<sup>(3)</sup>: a sample of the metal under investigation is placed into the large vacuum system containing the gases the sorption of which is being investigated. The ion source of the chronotron (type A) is placed in its close vicinity. Following the elapse of a certain period of time the sample is heated by passing a current pulse, and the atoms and molecules desorbed from the sample are analysed by the chronotron whilst passing once through the source. The vacuum pumps are continuously in operation during the measurement so that the pressure increase in the system during the desorption of the gases remains negligibly small. In contrast to the 'flash filament' method normally employed, where a gas filling a small vacuum system is being analysed, analysis of the desorbed gases in the molecular stream eliminates the errors associated with a selective sorption of the liberated gases at the walls. Recording of the time

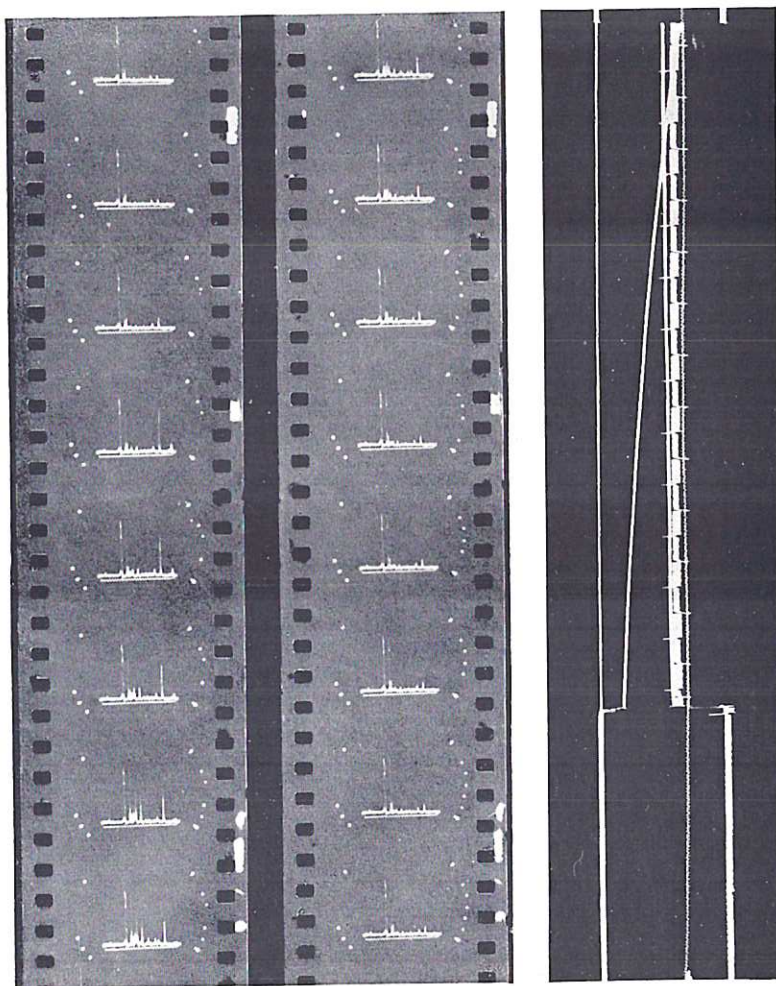


Fig. 5 (CLM-TRANS 2)  
Recorded spectra in the molecular current of desorbed  
gases, and temperatures on ciné strip

dependence of the desorption from the specimen during its heating, in conjunction with the recording of the temperature, permits a study of numerous details and aspects of the effect.

Fig.5 shows a number of frames from a filmstrip recording the spectrum of the desorbed gases with time markers and simultaneous temperature recording. The total duration of the desorption processes is one second. From the peak amplitudes on the spectrum corresponding to the various masses the concentration of the molecular current of the desorbed gases can be determined at any given moment, i.e. momentary value of the desorption rate. Fig.6

shows the results of an evaluation of such a record for the peak with mass number 28 (sorption of gases with composition indicated on the tungsten specimen). The curve of the desorption rate shows two maxima, the first corresponding to the desorption of carbon monoxide, the second to that of nitrogen. From the curve shapes various conclusions may be drawn about the structure of the sorbed layer and the effect of the surface structure on its characteristics.

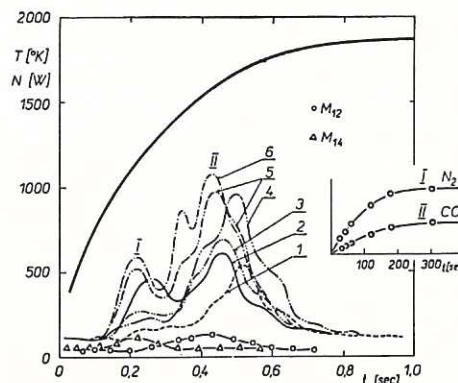


Fig. 6 (CLM-TRANS 2)  
Desorption rate of layers of mass 28 from a tungsten surface subjected to a rapid temperature rise

The quantity of different gas molecules which were present in the system and were sorbed at the metal surface during the period during which this surface was exposed to contact with these gases, is determined from the area of the curves shown in Fig.6 corresponding to each mass peak. From the relationship between the quantity of molecules sorbed and the duration of the exposure of the surface to the gases, the kinetic sorption characteristics can be plotted. In Fig.7 they are shown for the sorption of a gas mixture on tungsten. They indicate the selectivity of the sorption, and the mutual effect of the sorbed molecules on one another to the extent of the displacement of the one kind of sorbed molecules

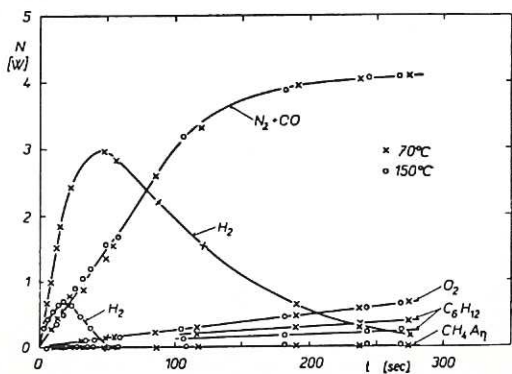


Fig. 7 (CLM-TRANS 2)  
Kinetic characteristics of the sorption on a tungsten surface of layers of a gaseous mixture

by others arriving at the surface in the course of the process. The use of pulse spectrometric techniques when studying sorption kinetics by the 'flash filament' method is a typical example of an application of the technique to various experiments relating to an analysis of molecular currents emitted from surfaces in the course of various surface effects. The pulse

method of analysis makes it possible to use modulated ion beams for separating the components of molecular beams according to their longer or shorter lifetimes.

(b) Study of the Mechanism of Sorption and Catalytic Reactions on Renewed Surfaces Using Stable Isotopes

As an example of the application of the pulse spectrometric method in conjunction with the use of stable isotopes, let us quote here the study of the sorption of hydrogen and the catalytic synthesis of methane on a renewed titanium surface at different temperatures.

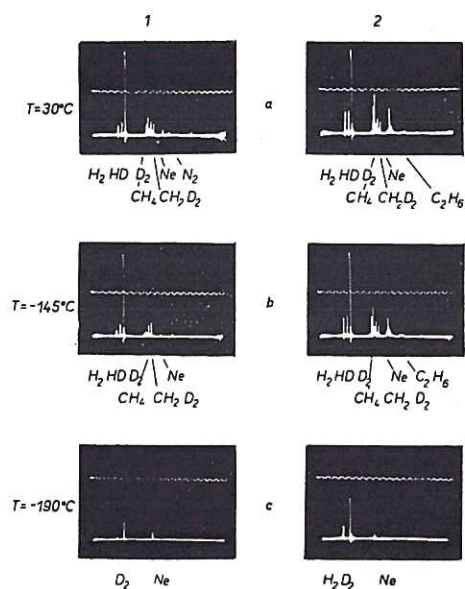


Fig. 8 (CLM-TRANS 2)  
Spectrum of gases above a titanium surface (1) without external effect and (2) bombarded with a beam of deuterium atoms for different surface temperatures

Fig.8 shows spectra of the composition of the gas above the titanium film once without any external action (Fig.8.1) and once during bombardment with an ion beam (Fig.8.2). Deuterium is admitted at a constant rate into the vacuum system at the walls of which the titanium film is deposited. The titanium layer is renewed by continuous evaporation of titanium. The spectra a, b, c, refer to various different temperatures of the film.

At room temperature the hydrogen molecules are adsorbed at the titanium surface and dissociate into atoms; an intense isotope exchange of hydrogen and deuterium takes place, associated with formation and liberation of DH. Simultaneously, methane is synthesized with the participation of deuterium from the labelled molecules. During the ion bombardment the principal component of the gases released are methane molecules bound weakly to the

surface. At sufficiently low temperature the sorption mechanism changes. The formation of DH and the synthesis of methane with deuterium are arrested; amongst the gases liberated during the ion bombardment, molecules containing carbon and radicals are practically absent. From the changes observed conclusions can be drawn about the transition from dissociative to non-dissociative chemisorption.

This example demonstrates the advantages of the pulse technique. At any given moment it provides information about all components of the gas mixture, and about all processes taking place simultaneously at the surface, and influencing one another.

(c) Interaction between the Hydrogen Plasma and the Walls of the Vacuum System

As an example of an application of the B type analyser we may use the study of interaction processes between a hydrogen plasma and the metallic walls of the vacuum chamber in toroidal thermonuclear devices<sup>(4)</sup>.

In toroidal devices of the quasi-stationary type the discharge vessels are subjected to stringent vacuum conditioning (heating, pumping-down to the ultra-high vacuum region). In these vessels effects were observed related to the physico-chemical interaction between the plasma and the walls. The gas pressure before applying the pulse is  $(1-2) \cdot 10^{-3}$  Torr, whilst following the pulse it drops by 30-60% due to the gas being bound to the walls. Impurities, containing especially carbon and oxygen, enter into the plasma. Fig.9 shows the spectra of the gas before and after the pulse obtained by means of a B-type analyser, and the variation of the partial pressures of the gas components and impurities, obtained from an evaluation of the spectra. In obtaining the spectrum the isotope method was used by alternating pulses in hydrogen and in deuterium. During the sorption of hydrogen at the walls, the gas previously chemisorbed during the previous pulses is liberated simultaneously with practically total exchange such that the gas present in the vessel following the pulse is no longer composed of the molecules admitted into the chamber previous to the given pulse but of molecules admitted into the chamber previous to earlier pulses (Fig.9,1a). The impurities present in the gas following the pulse consist of water vapour and carbon monoxide or also of methane when oil diffusion pumps are used. The water vapour enters into the volume as a result of chemical reduction reactions between the surface oxides and active hydrogen. This conclusion is based on a spectrum containing  $H_2O$ , HDO and  $D_2O$ . The remaining impurities present in the gas also originate in the course of surface catalytic reactions activated by the discharge.

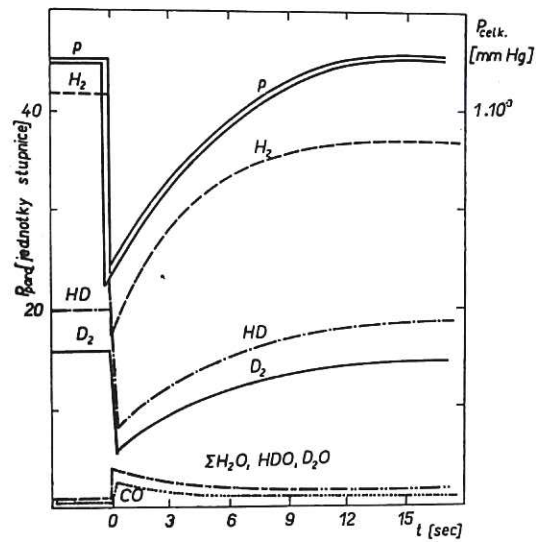
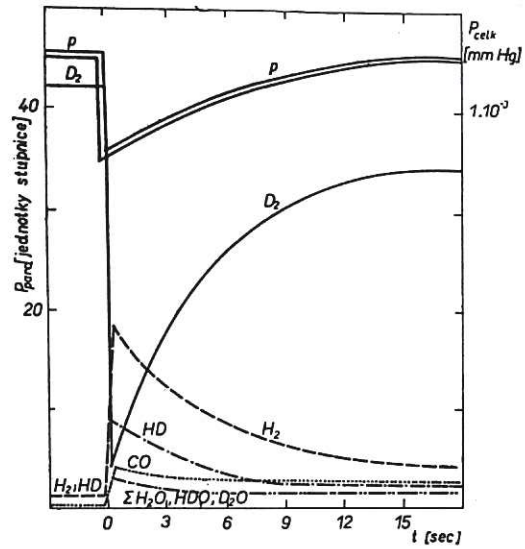
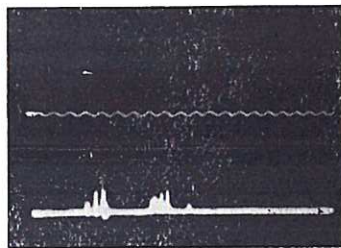
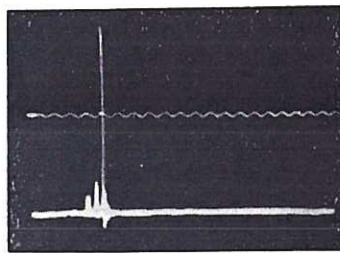
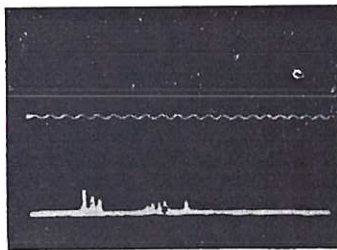
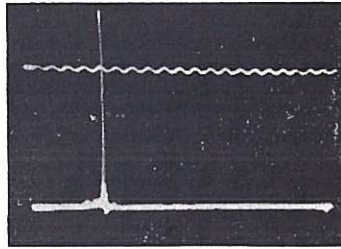


Fig. 9 (CLM-TRANS 2)  
 (1) Partial pressures of components and impurities in fillergas  
 (2) Spectra forming gas composition before and closely following pulse. A. Discharge in deuterium following discharge in hydrogen. B. Discharge in a deuterium and hydrogen mixture



(d) Composition and Pattern of Propagation of the Plasma During a Discharge between Metallic Electrodes in a Vacuum

In spark discharges struck between metal electrodes in a vacuum a non-stationary rapidly moving plasma is formed with a total containment time of the order of  $10^{-6}$  to  $10^{-5}$  secs. A spectrometric investigation of the composition of the plasma ions as a function of time permits the drawing of conclusions about the mechanism of the process. A D-type analyser is used.

Fig.10 shows spectra of plasma ions observed at different times following the initiation of the discharge ( $3 \times 10^{-7}$  to  $3.6 \times 10^{-6}$  sec.) with a time resolution of  $3.10^{-7}$  sec, in

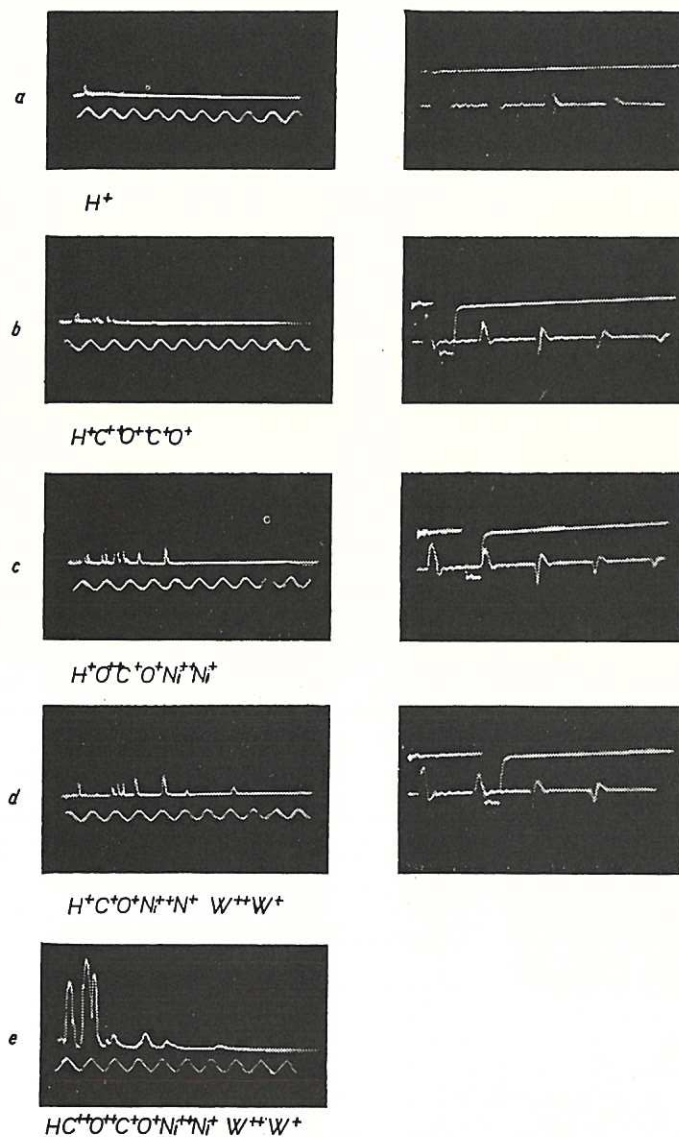


Fig. 10 (CLM-TRANS 2)  
Ion spectra of the plasma of a spark discharge entering the analyser at different times, with time markers. (a) delay time  $T - 0.3 \mu s$ ; (b)  $T - 1.7 \mu s$ ; (c)  $T - 2.7 \mu s$ ; (d)  $T - 3.6 \mu s$ ; (e) complete spectrum.

conjunction with time markers. The spectra refer to a discharge between a nickel and a tungsten electrode in a vacuum system using an oil diffusion pump (pressure  $1 \cdot 10^{-6}$  Torr) and a discharge pulse duration of  $5 \cdot 10^{-7}$  sec. The spectrum of the plasma ion includes singly and doubly ionized ions of the electrode materials, i.e. nickel and tungsten, and ions of the surface impurities, such as oxygen, carbon and hydrogen. The presence of material ions of both electrode materials, i.e. of the cathode and anode, can be attributed to the particular parameters of the source circuit (the discharge capacitance and voltage) at which the spectrum was measured. For different values of the precisely determined parameters only ions from the cathode or from the anode enter the spark discharge. During a period of  $1 \cdot 10^{-6}$  sec. following the discharge only hydrogen ions enter the entrance diaphragm of the spectrometer. During a further period of equal duration doubly and singly ionized carbon and oxygen ions reach the diaphragm, and during further similar periods also ions of the heavier metals. In the rapidly moving plasma of a spark discharge the ions are separated according to their mass. This means that the rapid motion of the plasma is not caused by the effect of the electrodynamic forces on the entire plasma. By means of the pulse method not only can qualitative information be obtained about fast processes but also information of a quantitative nature, such as the velocity of the separate plasma components.

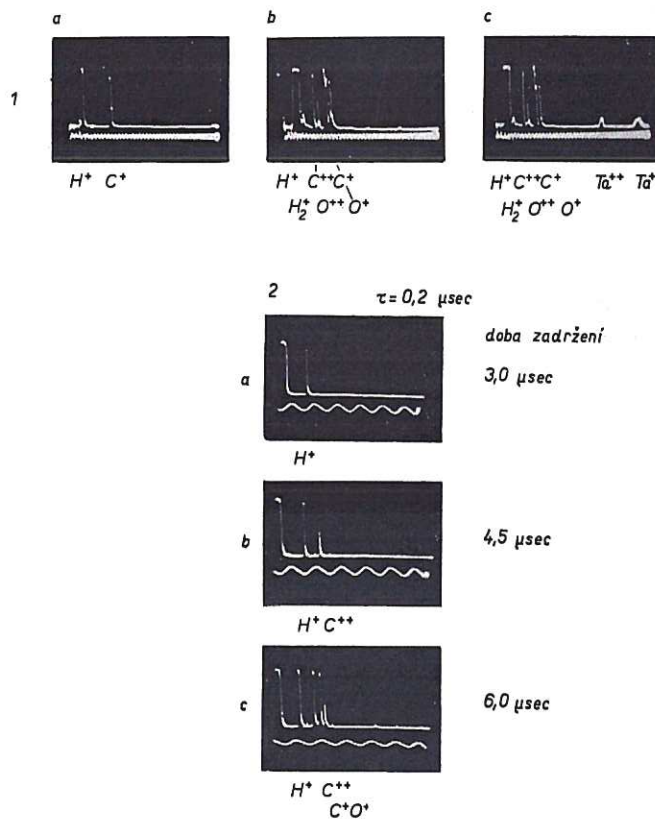


Fig. 11 (CLM-TRANS 2)  
 (1) Complete spectrum of the impurity ions of a hydrogen plasma from a microdischarge  
 (2) Spectrum showing composition of the impurities at different times following initiation of micro discharge

Fig.11 shows the complete ion spectrum and spectra obtained at different intervals, of micro-discharges taking place at a copper surface due to the action of a primary hydrogen plasma in a magnetic field. These spectra were obtained by the E-type analyser. The duration of the cathode spots during separate micro-discharges is approximately  $1 \cdot 10^{-5}$  sec. The complete spectra (Fig.11,1) indicate that the micro-discharge plasma contains chiefly carbon and oxygen ions, and to a lesser extent, and by no means in all discharges, also metal ions. The results indicate that the formation of the micro-discharges is attributable to charging-up of microscopic regions of dielectric at the surface by ions of the original plasma. On this basis it is possible to choose suitable constructional materials to limit contamination of the plasma. Spectra taken with a time resolution of  $2 \cdot 10^{-7}$  sec. (Fig.11,2) indicate a similar separation of the ions according to their mass during the propagation of a plasma from the cathode spots of micro-discharges parallel to the magnetic field in a low pressure gas as during propagation of a plasma in a vacuum without a magnetic field<sup>(1)</sup>.

(e) Passage of a Beam of Neutral Atoms containing Several Components, through a Highly Ionized Plasma

As a last example of applications of the pulse spectrometer method we refer to the molecular probing of the investigated medium where the different processes are not deduced from changes in the composition of the particles of the medium but from changes in the composition of molecular beams purposely introduced into the investigated medium and which do not by themselves influence phenomena taking place in the medium.

When 'irradiating' a plasma which is nearly 100% ionized with beams of atoms having thermal velocities, attenuation of the atomic beams occurs due to ionizations by the plasma electrons. The remaining processes are secondary. The relative attenuation of the beam is given by

$$\frac{N_0 - N}{N_0} = 1 - \exp\left(-n_e d \frac{\hat{\sigma}_i \delta_e}{\delta_0}\right) \approx n_e d \frac{\hat{\sigma}_i \delta_e}{\delta_0};$$

where  $N_0$  and  $N$  are the initial and final density of the beam,  $n_e$  the plasma concentration,  $d$ , the path length of the beam atoms in the plasma,  $\hat{\sigma}_i$ ,  $\delta_e$  are the ionization cross-sections of the atoms in the plasma (depending on the kind of atom and the electron temperature of the plasma),  $S_0$  the mean velocity of the atoms in the beam. In the case of a Maxwellian velocity distribution of the plasma electrons the relationship can be calculated between the ionization rate  $\hat{\sigma}_i \delta_e$  and the electron temperature, based on the relationship between the ionization cross-section and the electron energy, which is known for numerous gases. The time dependence of the plasma concentration and its electron

temperature (over a given range of these parameters) can then be measured by the pulse spectrometric method. The method is based on the bombardment of the plasma by a beam containing atoms of different kinds and with a different dependence between  $\hat{\sigma}_i \delta_e$  and the electron temperature, and the spectrometric determination of the attenuation of the beam for each component separately. By means of calibration curves plotted previously it is then possible, from the experimentally determined attenuation ratio

$$\left( \left( \frac{\hat{\sigma}_i \delta_e}{\delta_0} \right)_I : \left( \frac{\hat{\sigma}_i \delta_e}{\delta_0} \right)_{II} \right)$$

to obtain at each instant the electron temperature of the plasma and, from the absolute value of the attenuation  $n_e d (\hat{\sigma}_i \delta_e / \delta_0)$ , the plasma concentration.

Fig.12 shows a calibration curve for a beam of helium and argon atoms. It follows from the curve that simultaneous measurements can be made of the plasma concentration and electron temperature during irradiation of the plasma with a similar beam up to  $T_e$  values of approximately 30 eV. At higher temperatures the attenuation ratio of the helium and argon component of the beam is comparatively insensitive to temperature; for even higher plasma temperatures only the concentration can be determined with this method.

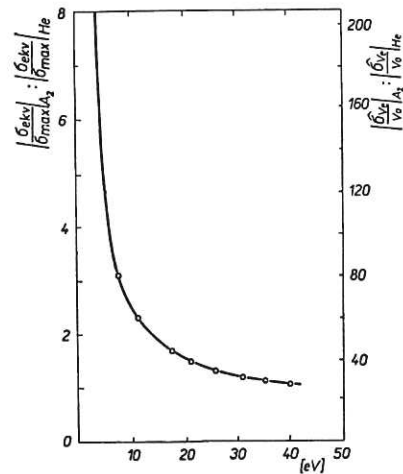


Fig.12 (CLM-TRANS 2)  
Calibration curve corresponding to a beam of helium and argon atoms, as used with the molecular probe method

The composition of the probe must be selected depending on the concentrations to be measured. For measuring high concentrations, atoms of light inert gases will be used; for measuring low concentrations heavy atoms are used, especially of the alkali metals. The molecular beams are detected with an A-type analyser. In view of its great sensitivity beams of low intensity can be used by which the plasma remains virtually unaffected. The variation in the intensities of the beam components is recorded by the auxiliary equipment of the chronotron using the recording method in accordance with the duration of the investigated effect in the plasma.

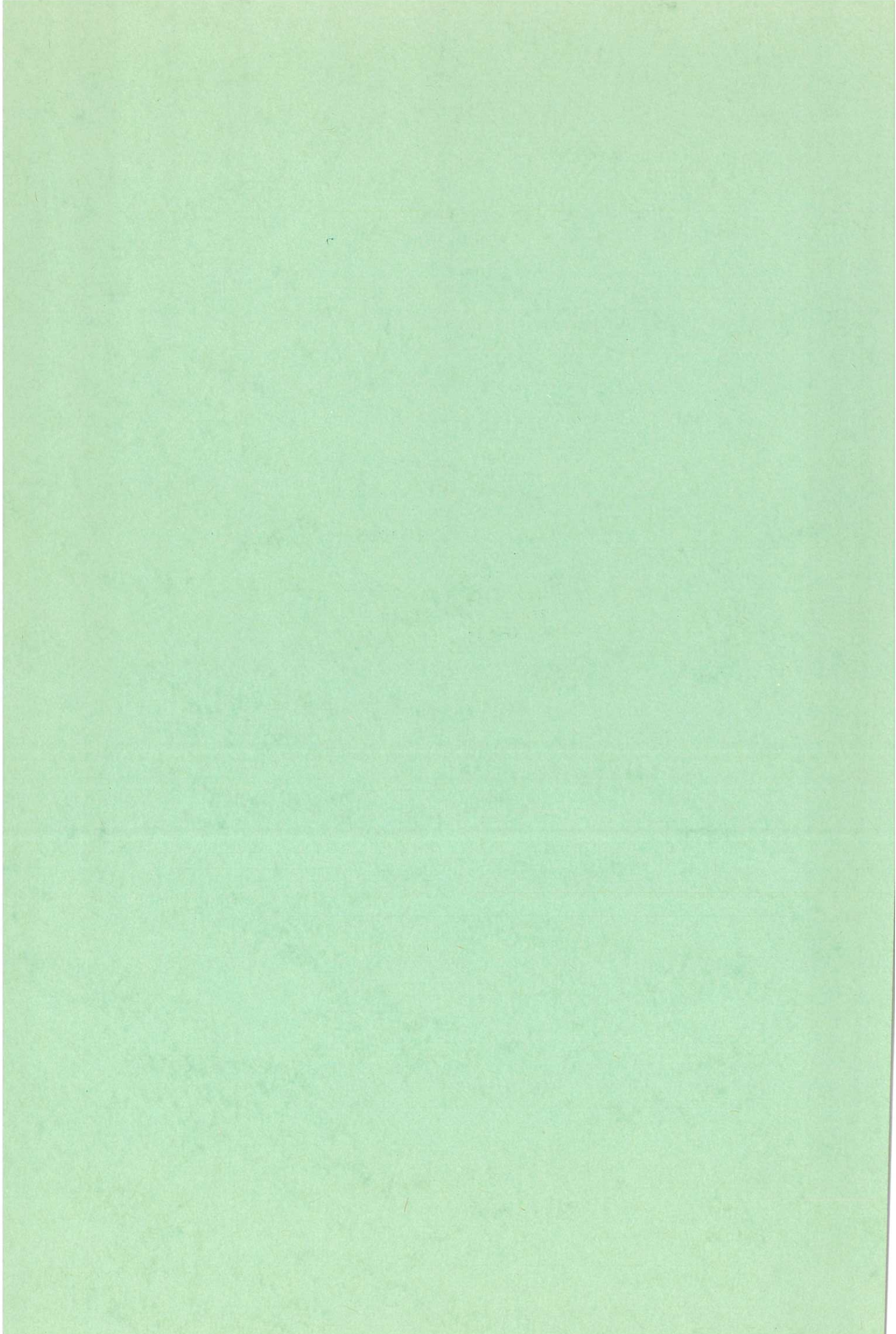
#### 4. CONCLUSIONS

The pulse spectrometer technique constitutes a valuable aid for the studying of fast processes in various fields of research - such as electronics, plasma physics, chemical kinetics and vacuum technology. With the equipment described effects can be analysed taking place in a neutral gas or a plasma over a wide range of pressures, plasma concentrations and durations of the investigated effects.

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