Zeitschrift: Bulletin de la Société Fribourgeoise des Sciences Naturelles = Bulletin der Naturforschenden Gesellschaft Freiburg

Herausgeber: Société Fribourgeoise des Sciences Naturelles

Band: 65 (1976)

Heft: 1: Mélanges en l'honneur du professeur Otto Huber à l'occasion de son 60e anniversaire = Festschrift zum 60. Geburtstag von Professor Otto Huber

Artikel: Three-body electron recombination in Plasma

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DOI: https://doi.org/10.5169/seals-308527

Three-Body Electron Recombination in Plasma

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At the beginning of this century Thomson (1924) posed a question (which arouse naturally in the course of his pioneering work, allowing him for the first time to measure the mass and charge of the electron), about the duration of life of the free electrons formed in the gas discharge. To explain the disappearance of the electrons and ions Thomson put forward the model of three- body recombination, which became the basis of all subsequent investigations in this field. The main idea of this model can be explained as follows. Let us consider a plasma consisting of the free electrons e, positive atomic ions A^+ and neutral particles. A free electron moving in the field of ion A^+ is characterized by the positive total energy E. It means that between the potential energy $-e^2/r$ of the electron in the field of A^+, its kinetic energy $mv^2/2$ and the total energy E the energy conservation relation $mv^2/2-e^2/r = E > O$ exists (r is the free electron distance from A^+). Suppose the free electron experiences at a distance $R = e^2/E$ from A^+ (where it has initially a kinetic energy $mv^2/2 = E + e^2/R = 2E$) a sudden collision with a "third particle" B (it can be a neutral particle or another free electron) and looses herewith a part of its kinetic energy in such a way, that the final kinetic energy $E' = mv'^2/2$ will be less than E. It means that after such collision the electron will have the *negative* total energy $mv'^2/2-e^2/R = E'-E < 0$, and thus it will

Bull. Soc. Frib. Sc. Nat. *65* (*1*), 76–87 (1976)

be captured by the Coulomb field of A^+. Equating E to the average electron thermal energy T (T is the temperature in the energetic units), we can say that an electron, experiencing the collision with a third particle inside a sphere around A^+ having the "Thomson radius" $r = e^2/T$, have a chance to be captured by A^+ and thus cease to exist as a free particle.

These qualitative considerations were used by THOMSON for the derivation of an approximate formula for the total rate of the free electron recombination in the recombination process $A^+ + e + B = A + B$ outlined above. Let us consider, following to THOMSON, the flux of the free electrons having the thermal velocity $v_T = (T/m)^{1/2}$ to an ion A^+. The capture cross- section of the electron in the Thomson process is of the order of $\pi r_T^2 w$, w being the probability that the electron will loose indeed the energy of the order T inside the Thomson sphere. The probability w is, evidently, of the order of $\frac{r_T}{L}\delta$, where L is the electron mean free path and δ is the probability of electron energy loss of the order of T in a binary collision with a third particle. Thus, the flux j of the electrons, experiencing the capture, to an ion A^+ is equal to $j = \pi r_T^2 \frac{r_T}{L} \delta v_T N_e$ (N_e is the density of the free electrons) and hence the total rate of the free electron recombination per a volume element is

$$dN_e/dt = -jN_+ = -\beta N_e^2 \quad (1)$$

(N_+ is the density of ions A^+ which coincides with N_e in a quasi-neutral plasma). The quantity β in (1) is the Thomson three- body recombination coefficient

$$\beta \simeq \pi r_T^3 v_T \delta / L \quad (2)$$

In a slightly ionized monoatomic plasma the role of the third particles carring away the free electron excessive energy play the neutral atoms A. In this case the probability $\delta \approx m/M$ (m is the electron mass and M is the mass of the atom A), while the electron mean free path is $L \approx 1/\sigma N_a$ (σ is the elastic scattering cross- section of electron by atom A and N_a is the density of atoms A). It means that the recombination coefficient β is given by the formula in this case

$$\beta \simeq e^6 N_a \delta\sigma / T^{5/2} m^{1/2} \quad cm^3/sec \quad (3)$$

On the other hand, in strongly ionized plasma, where the role of the third bodies play the free electrons, the mean free path $L \approx 1/r_T^2 N_e$ and $\delta \simeq 1$. So, for such plasmas the recombination coefficient is given by

$$\beta \simeq r_T^5 \, v_T \, N_e = e^{10} N_e / T^{9/2} m^{1/2} \; cm^3/sec \qquad (4)$$

With the development of plasma physics, especially during the last two decades, the investigation of the role of the three-body processes gained strongly in importance. Here is the list of the most important lines of research, in which the accounting for such processes is necessary.

1. *Physics of shock- waves and high- temperature gasdynamics*

Electrons formed in a gas behind a strong shock- wave (where a high temperature can be produced) recombine via three- body recombination process $A^+ + e + B = A + B$, which determines hence, to some extent, their structure and other physical properties. Such shock-waves arise e. g. around the bodies (space vehicles, meteorites) falling in the Earth' atmosphere and thus the plasma kinetics (involving also study of the three- body recombination processes) becomes an essential part of space – physics.

2. *Combustion processes and MHD- research*

The electrons formed in flames as a result of combustion reactions can disappear via three- body recombination process $A^+ + e + B = A + B$ (B is an atom or a molecule) which, consequently, influences the flame properties. On the other hand, the study of the electron distribution in flames can serve as a tool for investigating the elementary, e. g. the three- body recombination processes. The plasma arising as a result of combustion processes and flowing with high velocities in hypersonic nozzles can serve for the MHD- method of electric current generation. The distribution of electrons in such nozzles is often governed by the electron three- body recombination processes.

3. *Fusion research*

The electrons in high- temperature plasmas used for fusion purposes, e. g. in a stable, high- pressure gas discharge being explored now by

Kapitza (1970), can disappear via three- body recombination process $A^+ + e + e = A + e$, which greatly influences the plasma properties in this perspective method of generating the fusion nuclear energy.

It is therefore natural, that the great efforts were devoted during the last decade to clarify the nature of the three- body recombination process both theoretically and experimentally. A short account of these research, in particular those performed in Fribourg, is given below.

In mathematically consequent way the Thomson model of three-body recombination was formulated by Pitayevskii (1962). To understand this approach, let as follow the fate of an electron captured initially by the Thomson process on a highly excited level of A* atom, which can be considered as hydrogen- like. This electron with the total negative energy $- \varepsilon$ ($\varepsilon = I/n^2$, $I = 13{,}6$eV, n is the principal quantum number) will experience then the further excitation and de-excitation collisions with the third particles, characterized by the high cross- sections. (If the third particle is an electron, these cross- sections exceed πr^2, $r = a n^2$ being the effective radius of the excited electron in A* atom, and a the Bohr's radius). As long as the differences between the neighboring energy levels $\Delta\varepsilon = 2I/n^3$ remain small compared with T, the excitation and de-excitation collisions will be equi-probable and thus, the captured electron will perform (like a Brownian particle) the random walk between the upper levels of A* locsing or acquiring a small fraction of its energy in a single encounter with a third particle. It is very essential to notice here, that the probabilities of radiative transitions of the captured, bound electron (as long as it remains on the upper levels of A* and plasma is not too thin) are negligible compared with probabilities of collision – induced transitions. (The total probability of radiative de-excitation is of the order of $10^{10}/n^5$ sec^{-1}; for $n = 10$ and the plasma with parameters $N_e = 10^{14}$ cm^{-3}, $T \approx 1$ eV, this probability is smaller by a factor 10^{-5} than the probability of transition ($\approx a^2 n^4 V_T N_e$) provoked by collision with a free electron). It means, that the electrons captured on the highly excited states of A* remain their for a long time and, besides, they have about an equal chance to "fall down" to the lower states of A* or be excited (in particular to the continuum of the free electrons). As a consequence, an approximate thermodynamical equilibrium between the upper states of A* atoms and the continuum of the free electrons arises, i. e. the populations $N_o(n)$ of A* atoms (or the density of such atoms per a volume element), corresponding to a principal quantum number n, are connected with the density of free electrons by the Saha formula

$$N_o = h^3 n^2 N_e^2 \exp(\varepsilon /T) / (2 \pi m T)^{3/2} g^+ , \qquad \varepsilon = I/n^2 \quad (5)$$

(Here g^+ is the statistical weight for the A^+ ion).

The distribution (5) is an approximate one, since the electrons reaching, as a result of the diffusion process described above, some lower level, where the radiation probability becomes appreciable, will disappear in an irreversible way "falling down" to the ground state of A. The presence of such irreversible flux of recombining electrons, which modifies the distribution (5), can be, however, accounted for via the diffusion- type Fokker- Planck equation for the distribution function N(n), providing the mathematical equivalent to the qualitative picture of the electron random walk between the upper levels of A*. This equation, in which the role of the independent variable plays the principal quantum number n, can be written in the form (see SAYASOV, 1976)

$$\frac{\partial N}{\partial t} = - \frac{\partial j}{\partial n} \qquad j = b(n)\left(\frac{\partial N}{\partial n} - \frac{\partial \ln N_o}{\partial n} N\right) \quad (6)$$

where N_o is the Saha distribution (5) and b (n) is the diffusion coefficient

$$b(n) = \frac{1}{2} \int w(n, n')(n - n')^2 dn' \quad (7)$$

w (n, n') being the probability of collision-induced transition in A* between the levels with the quantum numbers n and n′. The quantity j has a meaning of the total flux of the recombining electrons, i. e. j/N_e^2 is just the recombination coefficient introduced by THOMSON. For an ideal plasma satisfying the condition $r/{}^3_T N_e \ll 1$ one can always use the equation (6) in the quasistationary form j = const, since for such plasmas the effective time of reaching the quasistationary regime j = const is much shorter than the effective plasma decay time. The equation j = const (with j defined by (6)) allows a solution $N = j N_o \int_{n_o}^{n} dn/N_o b(n)$. Using the boundary condition $N \to N_o$ for $n \to \infty$, we can rewrite this solution in the form

$$N = N_0 \int_{n_0}^{\infty} \frac{dn}{N_0 b} \Big/ \int_{n_0}^{\infty} \frac{dn}{N_0 b} \qquad n_0 \simeq 1 \tag{8}$$

while the recombination coefficient $\beta = j/N_e^2$ is defined simultaneously as

$$\beta = 1/ N_{en}^2 \int_0^{\infty} \frac{dn}{N_0 b} \qquad n_0 \simeq 1 \tag{9}$$

The formula (9) found first by PITAYEVSKII (1962) was used then in a number of papers for the calculation of the recombination coefficient corresponding to the different three- body reactions $A^+ + e + B = A + B$. PITAYEVSKII (1962) and VESELOVSKII (1967) considered the process in which the role of the third bodies play the atoms; the expression for β found in this way differs from the original formula of Thomson (3) only in a numerical factor. DALIDCHIK and SAYASOV (1966, 1967) investigated the recombination process, in which the role of the third body B a molecule experiencing the rotational excitation plays, and found the detailed form of the Thomson – type recombination coefficient (2) as a function of the basic molecular parameters.

The recombination coefficient β for the process $A^+ + e + e = A + e$ playing the fundamental role in the kinetics of strongly ionized plasmas was calculated in the framework of Fokker – Planck theory outlined above by GUREVICH and PITAYEVSKII (1964). However, they made a simplifying assumption about the transition probabilities $w(n,n')$ entering the diffusion coefficient (7) implying, that the velocities of the free electrons exceed greatly the velocities of the bound excited electron in A atom. (This assumption, evidently, is not valid for the binding energy $\varepsilon \gtrsim T$). This restriction was removed by SAYASOV (1976), who has found a more exact expression for the diffusion coefficient b (n) valid also for the levels with $\varepsilon \approx T$. The universal distribution for the populations N (n, l), pertaining to a state with definite values of the principal quantum number n and the angular momentum l of the excited electron in A^*, is represented by the formula (SAYASOV, 1976)

$$\frac{N(n\,\ell)}{g} = \left(\frac{N}{g}\right)_0 \phi(\varepsilon/T), \quad g = 2(2\ell + 1)$$

$$(N/g)_o = h^3 N_e^2 / (2\pi mT)^{3/2} g^+ \quad (10)$$

$$\phi(x) = e^x \int_x^\infty x^{3/2} e^{-x} F^{-1}(x) dx \; / \int_0^\infty x^{3/2} e^{-x} F^{-1}(x) dx$$

$$F(x) = (8/\pi) \int_0^\infty t^{1/2} e^{-xt} dt / (1+t)^3 \quad , F(0)=1$$

(The calculations of GUREVICH and PITAYEVSKII correspond to simplifying assumption F (x) = F (o) = 1). For the recombination coefficient β an expression was derived by SAYASOV (1976)

$$\beta = 0{,}42 \; 10^{-8} \; T^{-9/2} \; N_e \quad cm^3/sec \quad (11)$$

which agrees well with the results of the much more complicated numerical calculations performed by Monte Carlo method.

A very convenient tool for the experimental investigation of the kinetical regularities governed by three- body recombination provide the afterglow plasmas. The afterglow device worked out earlier in Fribourg by SCHNEIDER and HUGENTOBLER (1972) allowing to perform the exact mesurement of the electron densities (with laser interferometry) as well as the temperature (with microwave diagnostics) in the recombining plasmas, made it possible to mesure the recombination coefficients β for some gases in a broad intervale of temperatures (HOLLENSTEIN, SAYASOV and SCHNEIDER, 1975). The idea of this method can be described as follows. The kinetic equation (1) for a strongly ionized plasma where β is proportional to N_e has the form

$$\frac{dN_e}{dt} = - \alpha(T) N_e^3 \quad (12)$$

where the coefficient α depends upon the temperature only. It means, that α can be expressed in a simple way through the electron density: $\alpha = - N_e^{-2}\, dlnN_e/dt$. As the plasma decay observed by HOLLENSTEIN, SAYASOV and SCHNEIDER (1975) obeys with a good accuracy the exponential law ($N_e \sim \exp(-t/\tau)$), see Fig. 1, one can say, that $\alpha = 1/\tau\, N^2$. The mesurements of the temperature decay T (t) performed simultaneously with the measurements of densities N_e (t) lead to the formulation of the power-type relation $N_e \sim T^p$ for the decaying afterglow- plasmas, implying the power-type relation $\alpha \sim T^{-2p}$ also for the

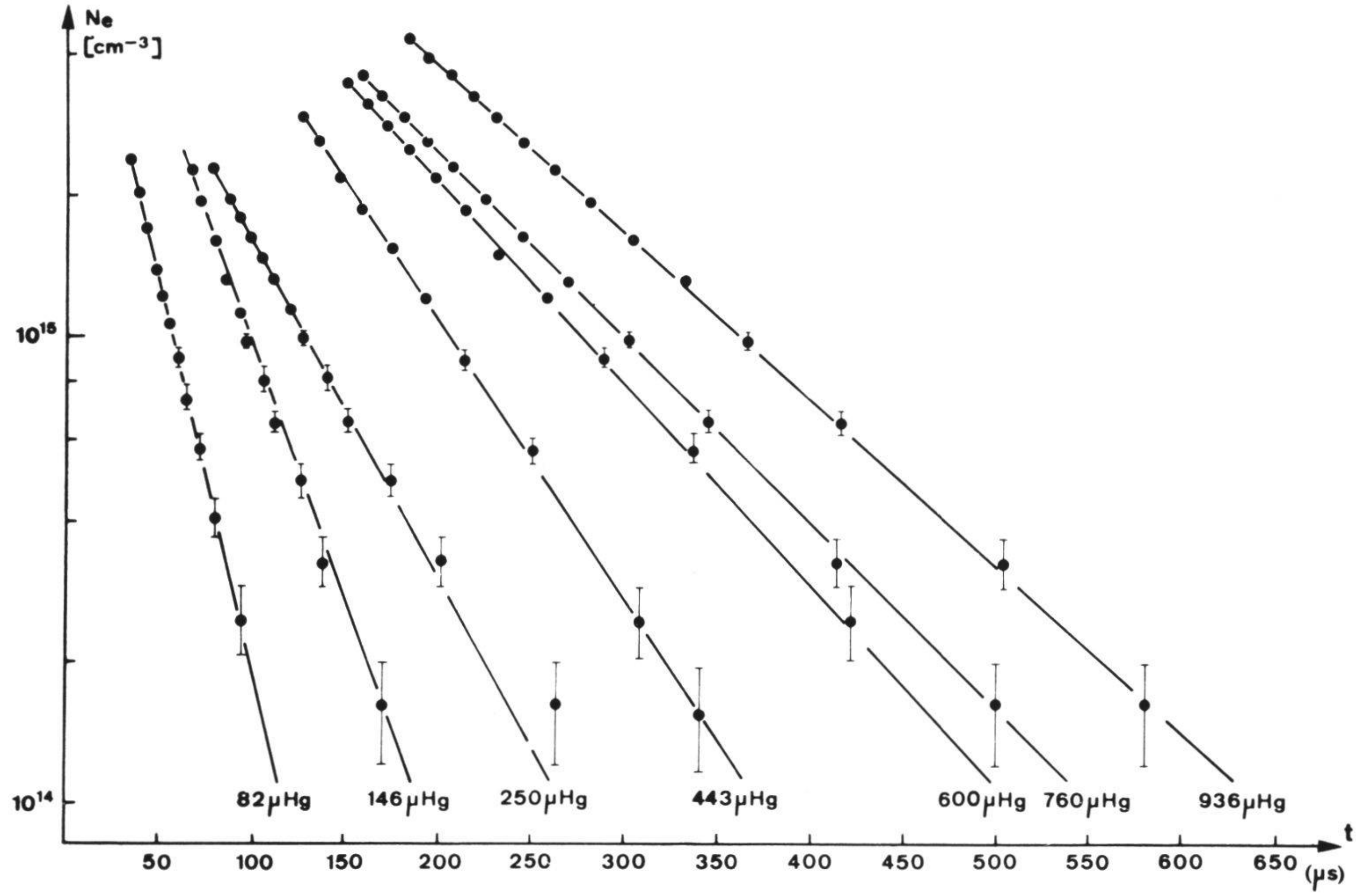

Fig. 1: The electron density Ne as the function of time (in μs) with pressure (in μHg) as parameter.

recombination coefficient. The temperature dependence of the recombination coefficient, found in this way by HOLLENSTEIN, SAYASOV and SCHNEIDER (1975) in a broader interval of temperatures of He- plasmas than those studied in the earlier works, is reproduced in Fig. 2. It is seen from Fig. 2, that experiment described above confirms the simple Thomson- type expression (11), as well as the results of the more complicated calculations performed by DRAWIN and EMARD (1971).

The universal Fokker- Planck distribution (10) was checked by comparison (Fig. 3) with the results of the extensive measurements of the population distribution in the recombining He- plasmas described by JOHNSON and HINNOV (1969) and by COLLINS and HURT (1968). As Fig. 3 shows, the relatively simple universal distribution (10) proves to be in a good agreement with the experimental results (for not too low excited levels of He). The knowlege of the temperature dependence of the recombination coefficient α is of prime importance for

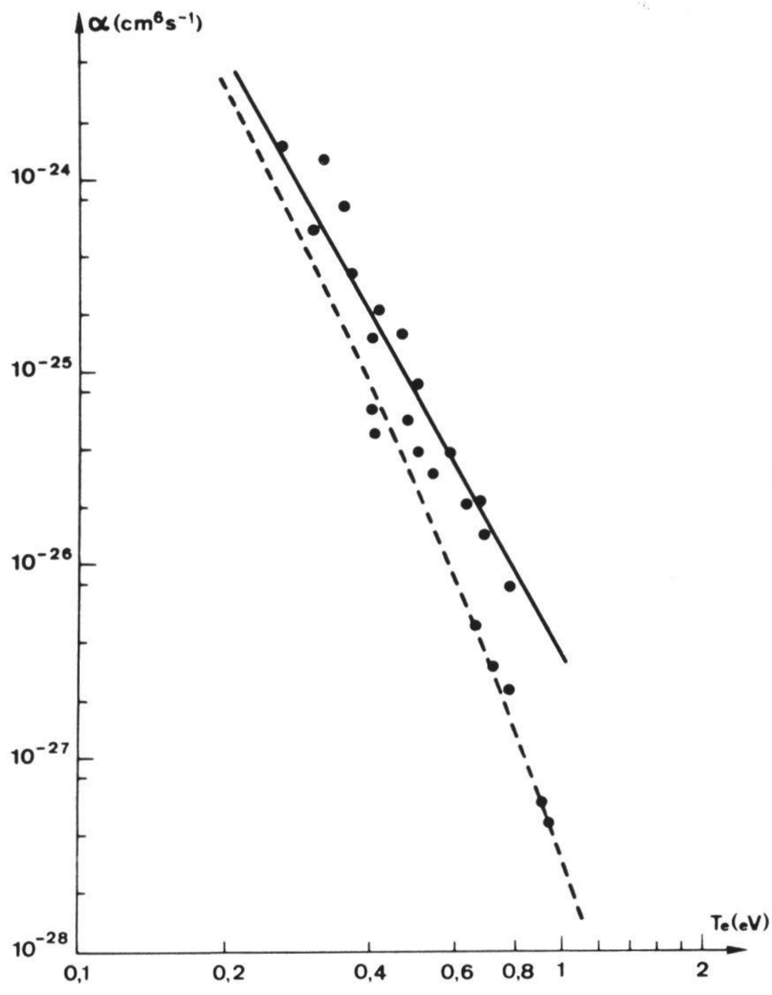

Fig. 2

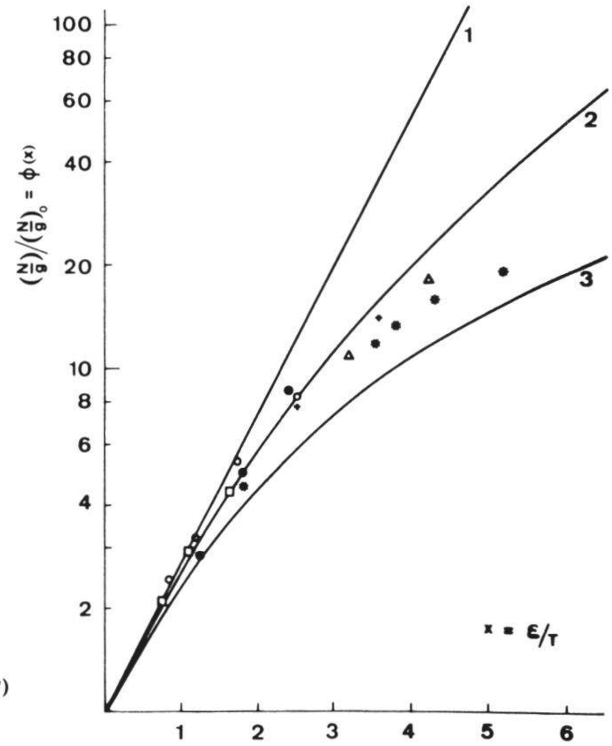

Fig. 3

Fig. 2: The recombination coefficient in a He- plasma as the function of tempareture (in eV). ● the experimental points from HOLLENSTEIN, SAYASOV, and SCHNEIDER (1975) : full line- Thomson – type formule (11); dashed line corresponds to the numerical calculations of DRAWIN and EMARD (1975).

Fig. 3: 1– Saha distribution corresponding to $\Phi = \exp(\varepsilon/T)$ in (10); 2– Fokker- distribution (10); 3– Fokker- Planck distribution corresponding to the simplified form of the diffusion coefficient b(n) used by GUREVICH and PITAEVSKII (1964). ●, ○, □, △, + experimental points corresponding to ^1S and ^3S – states of He- atoms and different conditions (JOHNSON and HINNOV (1969); * experimental points corresponding to the ^3D – states of He -atoms (COLLINS and HURT, 1968).

understanding of the kinetics of strongly ionized plasma decay. One of the questions which arise naturally in this context is–why the decay of the plasma afterglows produced in the different ways follows the exponentiall law (as in Fig. 1) notwithstanding the highly non- linear character of the equation (12) describing this decay? The following interpretation of this interesting phenomenon observed in a number of experiments, but never analysed before, was proposed by HOLLENSTEIN, SAYASOV and SCHNEIDER (1975).

The most important process responsible for the loss of the electron energy in the afterglow- discharge tubes is the electron thermal conductivity to the walls of the tube, which produces the energy flow to the walls of the order of $q^e \approx \chi dT/Rdr \approx \chi T/R^2$ (per a volume element). Here $\chi \approx k\ r_T^{-2}\ v_T \sim T^{5/2}$ is coefficient of the thermal conductivity, k is the Boltzmann constant, R is the radius of the discharge tube. This flow is compensated, however, to some extent by the heating due to the transfer of the Coulomb energy, which a recombining electron acquires in the field of a A^+-ion to the other electrons serving as the third bodies, thus increasing their kinetical energy. The energy production (per a volume element) due to this mechanism is $q_h \approx -E^*\, dN_e/dt = E^*\, \alpha\, N_e^3$, where $E^* \approx T$ is the energy released per one recombining electron. Equating approximately q_e and q_h, and assuming $\alpha \sim T^{-\delta}$, we get a relation $T \sim N_e^s$, $s = 3/(\delta + \frac{5}{2})$. Inserting this relation in (12), we reduce (12) to the equation $dN_e/dt = -N_e/\tau$ where parameter $\tau \sim N_e^b$, $b = 2(\delta-5)/(2\delta+5)$ is almost constant for $\delta \approx 5$, implying the exponential decay law for the temperature dependence of the recombination coefficient $\alpha \sim T^{-\delta}$ with $\delta \approx 5$, what is just the case for the three- body electron recombination of the type $A^+ + e + e = A + e$ (see (11)). The formula for the effective afterglow-decay time τ found in this way by HOLLENSTEIN, SAYASOV and SCHNEIDER (1975) proved to be in a good agreement with their experimental results as well as with the results described in a number of other papers. Thus, one can conclude, that the phenomenon of the exponential decay in the afterglow- plasmas is due to interplay of two strongly temperature- dependent processes: three- body electron recombination and the electron thermal conductivity, which leads to a relation between N_e and T reducing the basic kinetic equation (12) to the linear form $dN_e/dt = -N_e/\tau$, $\tau \approx$ const. In conclusion, it must be indicated here, that the simple theoretical approach based on the Fokker-

Planck equation (6) has its limitations. It can not, for instance, explain the very interesting phenomenon, observed e. g. by JOHNSON and HINNOV (1969) – the appearence of the inverted populations with N (n, l) ⟨N (n', l) for n'⟨n at the lower levels, which makes it possible, in principle, to use such recombining plasmas for creating the laser action. The more refined theoretical methods like those used by Drawin are necessary here. However, the experimental approach worked out by SCHNEIDER and HUGENTOBLER (1972) may prove to be useful for investigation of these fascinating phenomena as well.

Zusammenfassung

Hier wird eine kurze Übersicht über den heutigen Stand der plasmakinetischen Forschung im Zusammenhang mit der Elektron-Dreierstoßrekombination gegeben. Das Phänomen des exponentiellen Elektronendichtezerfalls in staık ionisierten Plasmen, beobachtet mit der Entladungsanlage des Physikalischen Instituts der Univ. Freiburg, wird besprochen. Eine elementare Deutung dieses Phänomens wird vorgeschlagen.

Résumé

On présente ici une brève vue d'ensemble de l'état actuel de la recherche en cinétique des plasmas, en relation avec la recombinaison électronique à trois corps. On discute la phénomène de la décroissance exponentielle de la densité électronique dans les plasma fortement ionisés, observés à l'aide des appareils à décharges de l'Institut de Physique de l'Université de Fribourg, et une interprétation élémentaire de ce phénomène est proposée.

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