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THE HARMONIC CONTRIBUTIONS TO THE ELECTRON VELOCITY DISTRIBUTION AND MACROSCOPIC QUANTITIES IN THE UNIFORM H_2 rf PLASMA[1)]

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The paper deals with the analysis of the harmonic contributions to the velocity distribution and relevant macroscopic quantities such as mean electron energy, particle current density and power input for an rf bulk plasma in H_2 over a wide range of the rf field frequency. The solution approach is based on the Fourier expansion technique applied to the non-stationary electron Boltzmann equation. A physical interpretation of the field frequency dependence of the harmonic contributions to the different quantities became possible by introducing lumped dissipation frequencies, both for energy and impulse.

I. INTRODUCTION

Recently comprehensive investigations of the periodic behaviour of the electron velocity distribution function and relevant macroscopic quantitites in the uniform bulk region of established rf discharges of inert and molecular gases [1—3] could be performed for not too high field frequency values. Especially it was found that at low rf field frequencies generally large modulation of the isotropic part of the velocity distribution function and of macroscopic quantities, as e.g. mean electron energy and mean collision frequencies, occur. When we increase the field frequency up to a critical frequency region, namely that covered by the lumped energy dissipation frequency ν_e/p_0 (which strongly depends on the kind of gas), a drastic reduction of this modulation is obtained. With further field frequency increase the just mentioned quantities tend to become time independent. However, when approaching ν_e/p_0, the anisotropic part of the velocity distribution and the relevant macroscopic quantities, i.e. the particle current density of the electrons and the power input from the rf field, remain still periodic functions with large modulation. It could be further found

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for an rf discharge in the inert gas Ne [2] that a reduction of the modulation of the latter quantities occurs only if the field frequency increases finally up to another critical frequency region, namely that covered by the lumped impulse dissipation frequency ν_i/p_0 in the gas considered.

The special aim of this paper is to investigate the different harmonic contributions to the isotropic and anisotropic distribution and to relevant macroscopic quantities in a wide field frequency range for an rf plasma in molecular hydrogen by using an appropriate solution technique for the non-stationary Boltzmann equation. These investigations are based upon the Fourier series expansions with respect to the time for both distribution parts in the Boltzmann equation and permit directly to determine the mentioned harmonic contributions up to very high field frequencies for a molecular gas. The field frequency dependence of the harmonic contributions to the different quantities obtained are microphysically interpreted by using the concept of energy and impulse dissipation frequency.

II. MAIN ASPECTS OF THE SOLUTION TECHNIQUE

The starting point is the non-stationary, spatially uniform Boltzmann equation for the electron velocity distribution function $F(\underline{v}, t)$

$$\frac{\partial F}{\partial t} - \frac{e_0}{m}\underline{E}\cdot\frac{\partial F}{\partial \underline{v}} = C^{el} + \sum_k C_k^{in} \tag{1}$$

in the rf field

$$\underline{E} = E\underline{e}_z, \quad E(t) = E_0 \cos(\omega t). \tag{2}$$

C^{el} and C_k^{in} are the collision integrals for elastic collisions and for several electron particle numbers conservative inelastic collision processes.

The Legendre polynomial expansion of $F(\underline{v}, t)$ in (1) gives in the Lorentz approximation finally two partial differential equations for the isotropic part $f(U, \bar{t})$ and for the first contribution $f_A(U, \bar{t})$ to the anisotropic part of the velocity distribution, where the electron energy U in eV and a normalized time scale $\bar{t}$ can be introduced according to $e_0 U = mv^2/2$ and $\bar{t} = p_0 t$ [2]. Furthermore, the same expansion yields the representation

$$\bar{U}(\bar{t}) = \int_0^\infty U^{3/2} f \,\mathrm{d}U,$$

$$\frac{j_e(\bar{t})}{n_e} = \frac{1}{3}\left(\frac{2e_0}{m}\right)^{1/2}\int_0^\infty U f_A \,\mathrm{d}U,$$

$$\frac{\bar{U}^F(\bar{t})}{p_0} = -\frac{E(\bar{t})}{p_0}\frac{j_e(\bar{t})}{n_e} \tag{3}$$

of the mean electron energy $\bar{U}$, of the electron particle current density j_e and of the mean power input $\bar{U}^F$ from the rf field. n_e denotes the electron density which is time independent due to the consideration of only conservative collision processes.

In the established rf plasma the isotropic and anisotropic distribution can be given the Fourier series expansion

$$\begin{aligned} f(U, \bar{t}) &= \sum_{n=-\infty}^{\infty} F_n(U)\,\mathrm{e}^{in\bar{\omega}\bar{t}}, \quad F_{-n}(U) = F_n^*(U), \\ f_A(U, \bar{t}) &= \sum_{n=-\infty}^{\infty} F_n^A(U)\,\mathrm{e}^{in\bar{\omega}\bar{t}}, \quad F_{-n}^A(U) = F_n^{A*}(U), \quad \bar{\omega} = \omega/p_0. \end{aligned} \tag{4}$$

Substitution of (4) into the mentioned partial differential equation system for f and f_A yields

$$in\bar{\omega}F_n - \frac{1}{6}\left(\frac{2e_0}{m}\right)^{1/2}\frac{E_0}{p_0}\frac{1}{U^{1/2}}\frac{\mathrm{d}}{\mathrm{d}U}[U(F_{n+1}^A + F_{n-1}^A)] - \frac{1}{U^{1/2}}\frac{\mathrm{d}}{\mathrm{d}U}\times$$
$$\times\left(U^{3/2}2\frac{m}{M}\frac{\nu_d}{p_0}F_n\right) + \sum_k\frac{\nu_k^{in}}{p_0}F_n - \sum_k\left(\frac{U+U_k^{in}}{U}\right)^{1/2}\frac{\nu_k^{in}(U+U_k^{in})}{p_0}F_n(U+U_k^{in}) = 0,$$
$$n = 0, 1, 2, \ldots,$$

$$\left(\frac{2e_0}{m}\right)^{1/2}\frac{E_0}{2p_0}U^{1/2}\frac{\mathrm{d}}{\mathrm{d}U}(F_{n+2} + F_n) - \left(\mathrm{i}(n+1)\bar{\omega} + \frac{\nu_d}{p_0} + \sum_k\frac{\nu_k^{in}}{p_0}\right)F_{n+1}^A = 0,$$
$$n = -1, 0, 1, 2, \ldots, \tag{5}$$

where ν_d is the collision frequency for the impulse transfer in elastic collisions, ν_k^{in} the total collision frequency and U_k^{in} the corresponding energy loss for the kth inelastic collision process. For the isotropic distribution the natural normalization $\int_0^\infty U^{1/2} f(U, \bar{t})\,\mathrm{d}U = 1$ can be used. Its Fourier expansion leads to the conditions

$$\int_0^\infty U^{1/2}F_0(U)\,\mathrm{d}U = 1, \quad \int_0^\infty U^{1/2}F_n(U)\mathrm{d}U = 0, \quad n = 1, 2, 3, \ldots \tag{6}$$

for the Fourier coefficients of $f(U, \bar{t})$. A detailed observation of the coupling in (5), particulary the possibility of the elimination of the Fourier coefficients

F_1^A, F_3^A, ... by using the second equation of (5), and the consideration of the normalization conditions (6) show that (5) can be separated into two independent equation systems, one for the F_0, F_1^A, F_2, F_3^A, ... and a further one for F_0^A, F_1, F_2^A, F_3, ... Particularly the zero normalization of all F_1, F_3, ... according to (6) leads to the conclusion that the second system has only the trivial solution and can thus be neglected. Therefore, the problem to determine the harmonic contributions of the isotropic and the anisotropic distribution is reduced to the first system for the functions F_0, F_1^A, F_2, F_3^A, ... A natural truncation of this hierarchy is obtained when considering the equations of (5) for even n's up to a certain even $2l$, i.e. the equations for $n = 0, 2, 4, \ldots, 2l$ and neglecting in the second equation of (5) the term F_{n+2} for $n = 2l$.
Thus the Fourier series expansion of the isotropic and anisotropic distribution for the non-trivial solution of the truncated system (5) can be written in the reduced form

$$f(U, \bar{t}) = \sum_{n=-l}^{l} F_{2n}(U)\,\mathrm{e}^{\mathrm{i}2n\bar{\omega}\bar{t}}, \quad F_{-2n}(U) = F_{2n}^*(U),$$

$$f_A(U, \bar{t}) = \sum_{n=-l}^{l+1} F_{2n-1}^A(U)\,\mathrm{e}^{\mathrm{i}(2n-1)\bar{\omega}\bar{t}}, \quad F_{-(2n-1)}^A(U) = F_{2n-1}^A(U). \qquad (7)$$

From this complex representation follows the real representation

$$f(U, \bar{t}) = F_0 + \sum_{n=1}^{l} 2|F_{2n}(U)| \cos(2n\bar{\omega}\bar{t} + \varphi_{2n}(U)),$$

$$f_A(U, \bar{t}) = \sum_{n=1}^{l+1} 2|F_{2n-1}^A(U)| \cos((2n-1)\bar{\omega}\bar{t} + \varphi_{2n-1}^A(U)), \qquad (8)$$

where $|F_{2n}|$, $|F_{2n-1}^A|$ and φ_{2n}, φ_{2n-1}^A denote the magnitude and the phase of the complex harmonics F_{2n} and F_{2n-1}^A.
The corresponding Fourier expansions of the macroscopic quantities given in (3) read then

$$\bar{U}(\bar{t}) = \sum_{n=-l}^{l} \bar{U}_{2n}\mathrm{e}^{\mathrm{i}2n\bar{\omega}\bar{t}}, \quad \bar{U}_{2n} = \int_0^\infty U^{3/2}F_{2n}\,\mathrm{d}U, \quad \bar{U}_{-2n} = \bar{U}_{2n}^*, \qquad (9)$$

$$\frac{j_e(\bar{t})}{n_e} = \sum_{n=-l}^{l+1} \left(\frac{j_e}{n_e}\right)_{2n-1} \mathrm{e}^{\mathrm{i}(2n-1)\bar{\omega}\bar{t}}, \quad \left(\frac{j_e}{n_e}\right)_{2n-1} = \frac{1}{3}\left(\frac{2e_0}{m}\right)^{1/2} \int_0^\infty UF_{2n-1}^A\,\mathrm{d}U,$$

$$\left(\frac{j_e}{n_e}\right)_{-(2n-1)} = \left(\frac{j_e}{n_e}\right)_{2n-1}^*, \qquad (10)$$

$$\frac{\bar{U}^F(\bar{t})}{p_0} = \sum_{n=-l-1}^{l+1} \left(\frac{U^F}{p_0}\right)_{2n} \mathrm{e}^{\mathrm{i}2n\bar{\omega}\bar{t}},$$

$$\left(\frac{\bar{U}^F}{p_0}\right)_{2n} = -\frac{E_0}{2p_0}\left[\left(\frac{j_e}{n_e}\right)_{2n-1} + \left(\frac{j_e}{n_e}\right)_{2n+1}\right], \quad 2n = 0, 2, \ldots, 2l,$$

$$\left(\frac{\bar{U}^F}{p_0}\right)_{2l+2} = -\frac{E_0}{2p_0}\left(\frac{j_e}{n_e}\right)_{2l+1}, \quad \left(\frac{\bar{U}^F}{p_0}\right)_{-2n} = \left(\frac{\bar{U}^F}{p_0}\right)_{2n}^*, \quad 2n = 0, 2, \ldots, 2l+2. \qquad (11)$$

From these complex representations similar real representations as given in (8) for both distribution parts can be obtained. For the solution of the truncated system (5) it is convenient to replace the Fourier coefficients $\mathrm{Re}(F_1^A)$ and F_{2n-1}^A, $n \geq 2$ according to $G_{2n} = F_{2n-1}^A + F_{2n+1}^A$ by the functions G_{2n}. The resulting system of ordinary differential equations (with additional difference terms) for the functions F_{2n} and G_{2n} then contains $4l + 2$ real function components and is weakly singular at small energies with the singular point $U = 0$, strongly singular, however, for large energies with the singular point $U = \infty$. Thereby appropriate series expansions have been assumed for the collision cross sections involved in the different collision frequencies ν_d and ν_k^{in} near the singular points and immediately above the thresholds of the inelastic collision processes, respectively. Because of the different character of the singularities of the just mentioned system separate considerations of the structure of its general solution are necessary in the region of small and large energies, respectively. It was found that the general solution at small as well as that at large energies contains $2l + 1$ non-singular and $2l + 1$ singular fundamental solutions. The desired, i.e. physically relevant solution has to be sought within the non-singular part of the general solution (NSPG) both at small and large energies, with both NSPGS's involving a total of $4l + 2$ free parameters. The physically relevant solution can be uniquely determined (i) by the construction of the NSPGS at small as well as large energies, (ii) by a continuous connection of these at an appropriate connection point U_c and (iii) by additional normalization.
A special technique was developed to isolate numerically all contributions to both NSPGS's starting firstly from a sufficiently large energy down to the connection point U_c and secondly from the singular point $U = 0$ up to U_c to find thus both NSPGS's. This technique leads to the solution of the system in an approximation order with $4l + 2$ terms via a $(2l + 1)$-fold backward and a $(4l + 2)$-fold forward integration in order to construct both NSPGS's from the singular points to U_c.

III. RESULTS AND DISCUSSION

The calculations have been performed for an rf plasma in H_2 at a field amplitude $E_0/p_0 = 23$ V cm^{-1} Torr^{-1} applying the collision cross sections used in [2]. Figs. 1 to 4 show the harmonic contributions to the isotropic distribution

in dependence on the electron energy for the field frequency values $\omega/p_0 = \pi \cdot 10^7, \pi \cdot 10^8, \pi \cdot 10^9$ and $10^{10}\ s^{-1}\ Torr^{-1}$. These and all further results are obtained in an approximation with $l = 2$, i.e. with $4l + 2 = 10$ terms.
At $\omega/p_0 = \pi \cdot 10^7$ large contributions of the harmonics F_2 and (to a lesser extent) F_4 can be observed at higher electron energies (above ≈ 10 eV) and somewhat smaller contributions at lower energies (below ≈ 10 eV). For lower field fre-

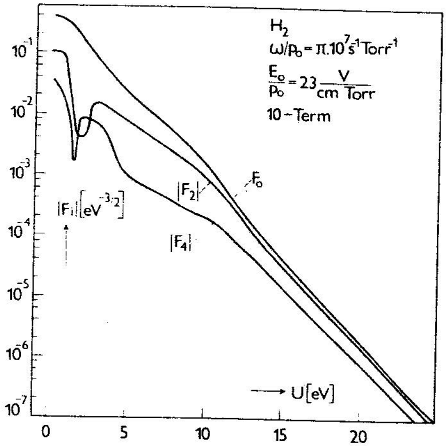

Fig. 1. Harmonic contributions to the isotropic distribution at $\omega/p_0 = \pi \cdot 10^7\ s^{-1}\ Torr^{-1}$ in dependence on the electron energy.

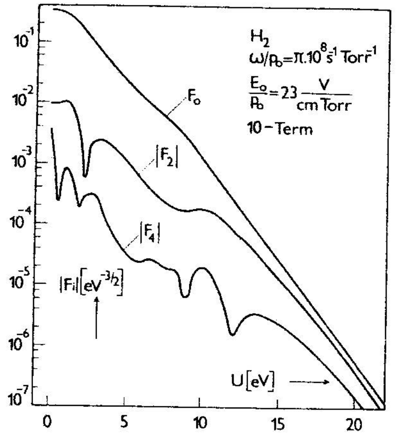

Fig. 2. Harmonic contributions to the isotropic distribution at $\omega/p_0 = \pi \cdot 10^8$.

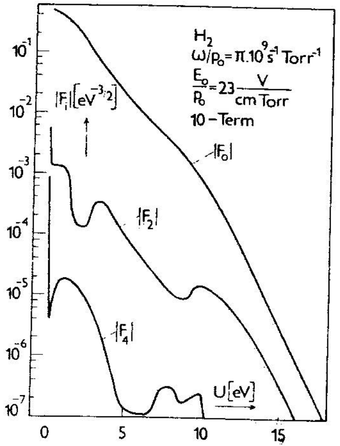

Fig. 3. Harmonic contributions to the isotropic distribution at $\omega/p_0 = \pi \cdot 10^9$.

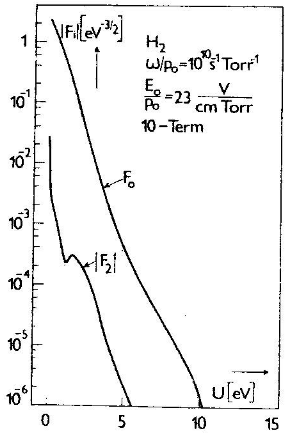

Fig. 4. Harmonic contributions to the isotropic distribution at $\omega/p_0 = 10^{10}$.

quencies these contributions become still larger. From Fig. 2 it can be seen that at $\omega/p_0 = \pi \cdot 10^8$ a drastic reduction of both harmonics, particularly at lower energies, occurs, whilst a larger contribution of F_2 remains at higher energies. With a further increase of the field frequency this reduction continues as seen from Figs. 3 and 4 and at $\omega/p_0 = \pi \cdot 10^9$ also F_2 is already remarkably smaller than F_0, even at the highest relevant energy. At such a field frequency the isotropic distribution is practically represented by its dc part F_0. Moreover, the comparison of Figs. 1 and 2 indicates that up to this field frequencies the dc part F_0 of the isotropic distribution shows only a slight change. Only when passing from $\omega/p_0 = \pi \cdot 10^8$ to 10^{10} a drastic depopulation in the dc part F_0 of the isotropic distribution at not too small energies takes place and the harmonic contributions F_2 and F_4 decrease further on.
Thus the question arises how such a drastic evolution of the harmonic contributions to the isotropic distribution can be understood. As already mentioned this becomes possible when using the lumped dissipation frequencies for energy and impulse which can be represented by the individual collision frequencies according to the expressions

$$\frac{\nu_e}{p_0} = 2\frac{m}{M}\frac{\nu_d}{p_0} + \sum_k \frac{\nu_k^{in}}{p_0}, \quad \frac{\nu_i}{p_0} = \frac{\nu_d}{p_0} + \sum_k \frac{\nu_k^{in}}{p_0}. \tag{12}$$

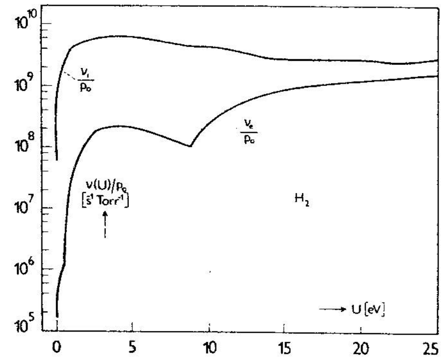

Fig. 5. Lumped energy and impulse dissipation frequency for H_2 in dependence on the electron energy.

Both these frequencies are shown in Fig. 5 for H_2 in dependence on the electron energy. As mentioned in the Introduction large modulations of the isotropic distribution are possible for $\omega/p_0 \ll \nu_e/p_0$ since then at every instant during the alteration of the rf field a remarkable energy dissipation in collisions can still occur. In agreement with these statements large contributions of the harmonics F_2 and F_4 can be observed at lower field frequencies. If ω/p_0 becomes comparable to ν_e/p_0, a larger instantaneous energy dissipation in collisions is no longer possible, which leads to a distinct reduction of the isotropic distribution modulation. This is the case at $\omega/p_0 \approx \pi \cdot 10^8$ in the region of lower electron energies as to be seen from the values of v_e/p_0 in Fig. 5. Thus at these energies the harmonic contributions remarkably decrease as obvious from Fig. 2. The same situation occurs at higher energies at a field frequency of $\approx \pi . 10^9$ so that now in Fig. 3 the harmonic contributions become small also at higher energies. Only for field frequencies, which are relevant to those considered in Figs. 3 and 4, ω/p_0 becomes comparable or even larger than the impulse dissipation frequency v_i/p_0. Under these conditions also a larger instantaneous impulse dissipation in collisions is no longer possible, so that a reduction of the electron current density and of the resultant power input from the rf field occurs as it is clearly seen from results presented later. The reduced power input now causes the mentioned drastic depopulation of the dc part of the isotropic distribution with increasing field frequency in this frequency range.

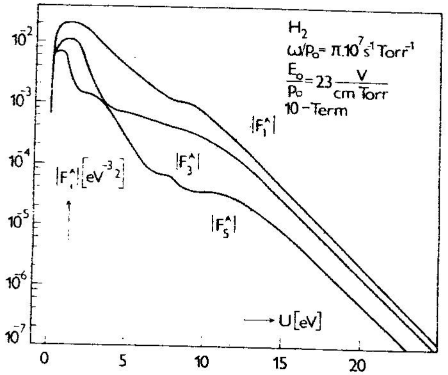

Fig. 6. Harmonic contributions to the anisotropic distribution at $\omega/p_0 = \pi . 10^7$.

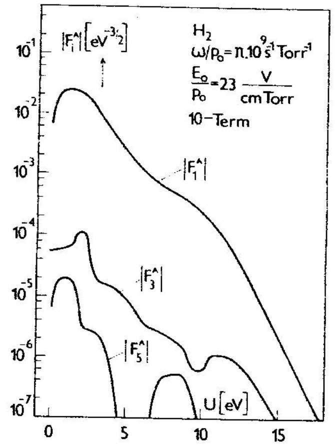

Fig. 7. Harmonic contributions to the anisotropic distribution at $\omega/p_0 = \pi . 10^9$.

To illustrate the harmonic contributions to the anisotropic distribution $f_A(U, \bar{t})$, Figs. 6 and 7 show the results for $\omega/p_0 = \pi . 10^7$ and $\pi . 10^9$. Of course according to (7), there is no dc part of f_A. At lower frequencies the higher harmonics F_3^A and F_5^A yield besides the lowest harmonic F_1^A a remarkable contribution to the anisotropic distribution. When ω/p_0 becomes comparable to v_e/p_0 the higher harmonics F_3^A and F_5^A strongly decrease, similar to the harmonics F_2 and F_4 of the isotropic distribution. This happens, as obvious, when passing from Fig. 6 to 7. Only if ω/p_0 becomes comparable with the impulse dissipation frequency v_i/p_0, also a strong reduction of the lowest harmonic F_1^A occurs. The onset of this last effect is already indicated by Fig. 7.

The characteristic changes of the harmonic contributions to the isotropic and anisotropic distribution just discussed are reflected in the corresponding field frequency dependence of the harmonic cintributions to the mean electron energy (determined by the isotropic distribution), electron particle current density and the power input from the rf field (both determined by the anisotropic distribution) as given by (9) to (11). Fig. 8 shows the harmonic contributions to the mean electron energy according to (9) over a wide field frequency range, i.e. from ω/p_0 much smaller than the energy and impulse dissipation frequency up to ω/p_0 remarkably larger than both dissipation frequencies. In agreement with the frequency dependence of the harmonic contributions to the isotropic distribution, $|\bar{U}_2|$ is comparable with the dc part $\bar{U}_0$ at low ω/p_0. When ω/p_0 approaches and exceeds v_e/p_0 (cf. Fig. 5), a drastic decrease of the harmonics $\bar{U}_2$ and $\bar{U}_4$ can be seen. However, up to field frequencies of $\omega/p_0 \approx 10^9$ the dc part $\bar{U}_0$ remains nearly unchanged. But if ω/p_0 approaches and exceeds v_i/p_0 (cf. fig. 5), also the dc part of $\bar{U}$ remarkably decreases. This last fact is a reflection of the above mentioned strong depopulation of the dc part F_0 of the isotropic distribution at these field frequencies.
Fig. 9 shows the harmonic contributions to j_e/n_e (i.e. to the drift speed) according to (10) in the same representation as in Fig. 8. Despite the larger contributions of the higher harmonics F_3^A and F_5^A to the anisotropic distribution at lower

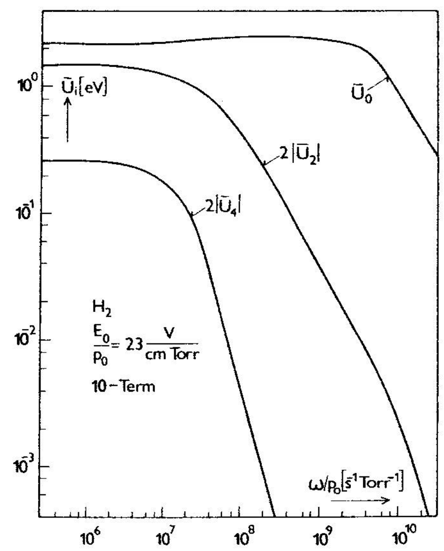

Fig. 8. Harmonic contributions to the mean electron energy in a wide field frequency range.

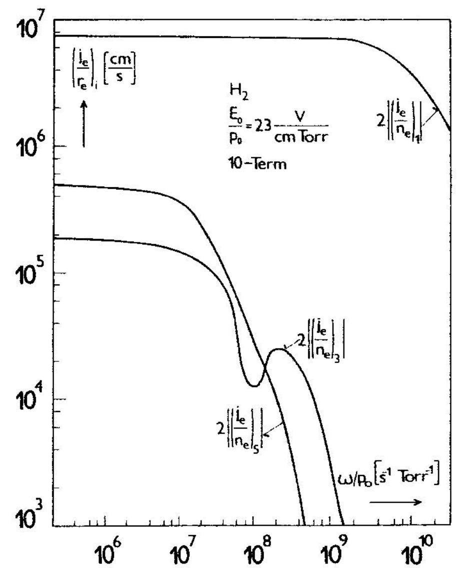

Fig. 9. Harmonic contributions to the electron particle number density.

field frequencies (cf. Fig. 6), the higher harmonic contributions $(j_e/n_e)_3$ and $(j_e/n_e)_5$ are already relatively small in comparison with the lowest harmonic $(j_e/n_e)_1$ in the range of lower ω/p_0 values. When ω/p_0 approaches and exceeds the energy dissipation frequency ν_e/p_0, the higher harmonics drastically decrease, however, the lowest harmonic remains nearly unchanged. Only if ω/p_0 becomes comparable with and larger than the impulse dissipation frequency ν_i/p_0, a remarkable reduction also of the lowest harmonic $(j_e/n_e)_1$ can be observed. This is in correspondence to the strong reduction of the lowest harmonic F_1^A with increasing field frequency in this frequency range (cf. Fig. 7).

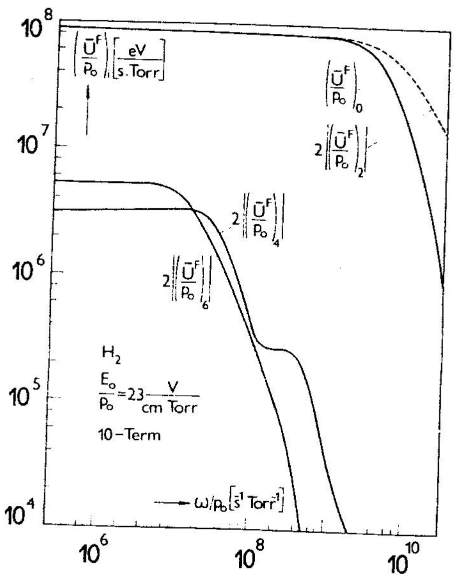

Fig. 10. Harmonic contributions to the power input from the rf field.

Finally Fig. 10 reports the harmonic contributions to the power input from the rf field according to (11) for the same field frequency range. Now the dc part $(\bar{U}^F/p_0)_0$ and, in addition, the lowest harmonic contribution $(\bar{U}^F/p_0)_2$ are nearly equal up to large field frequencies, whilst the higher harmonics $(\bar{U}^F/p_0)_4$ and $(\bar{U}^F/p_0)_6$ are already relatively small at low field frequencies and drastically decrease when ω/p_0 approaches and exceeds the energy dissipation frequency. Both quantities, the dc part and the lowest harmonic of the power input involve, according to (11), the lowest harmonic $(j_e/n_e)_1$. Therefore these quantities remain large and nearly equal up to high ω/p_0. However, the higher harmonics are small for all field frequencies since they are determined by the higher harmonic contributions to the electron current density (cf. (11) and Fig. 9). When ω/p_0 becomes comparable with and larger than the impulse dissipation frequency, the dc part and the lowest harmonic contribution to the power input from the rf field rapidly decreases. However, the second harmonic $(\bar{U}^F/p_0)_2$ remains larger than the dc part at these field frequencies. This fact means that at these ω/p_0 values the power input from the rf field to the electrons becomes even negative during certain parts of the rf period, i.e. the power flows partly back from the electron component to the rf field, which is a reflection of the increasing inertia of the electrons in the very rapidly varying rf field at large ω/p_0.

Concluding we would like to emphazise that the complex change with the increasing field frequency of the harmonic contributions to both distribution parts as well as to the relevant macroscopic quantities can be well understood on the basis of the two lumped dissipation frequencies for energy and impulse, i.e. by using quantities which can already be obtained from the atomic data of the relevant electron collision processes involved.

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ГАРМОНИЧЕСКИЕ ВКЛАДЫ В РАСПРЕДЕЛЕНИЕ СКОРОСТИ ЭЛЕКТРОНОВ И МАКРОСКОПИЧЕСКИЕ ВЕЛИЧИНЫ В ОДНОРОДНОЙ H_2 rf ПЛАЗМЕ

Работа посвящена анализу гармонических вкладов в распределение скорости и соответствующих макроскопических величин, таких, как энергия электронов, плотность потока частиц и мощность на входе, для rf объемной плазмы в H_2 в широком диапазоне частот поля. Анализ основан на технике Фурье-разложения, примененной для нестационарного уравнения Больцмана для электронов. Физическое объяснение зависимости полевой частоты гармонических вкладов от разных величин может быть дано, если ввести приведенные диссипативные частоты как для энергии, так и для импульсов.