

Frequency Variation of the Mean Energy of r.f. Electron Swarms*

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Abstract

Starting from the general momentum and energy balance equations for charged particle swarms in a gas, as furnished by momentum-transfer theory, we obtain expressions for mean velocity and mean energy of an electron swarm in an r.f. electric field under spatially uniform conditions, in the frequency range $\omega\tau_e > 1$, where τ_e is the energy collisional relaxation time. If τ_e is a decreasing function of energy, it is shown that the cycle-averaged mean energy reaches a maximum at a certain frequency. Physical arguments are provided to support this result and the prediction is verified for a constant elastic cross section model by direct numerical solution of Boltzmann's equation.

1. Introduction

Periodic steady state plasmas driven by radio frequency and/or micro waves are widely used in microelectronic device fabrication and in gas laser excitation (see for example Makabe *et al.* 1994). In these new technological applications, the frequency covers the range 100 kHz to 2.45 GHz. Most plasmas are sustained between capacitively coupled parallel plates or in an inductively coupled cylinder, both in a collision dominated or in a collisionless regime. Radio-frequency electron transport in a collision dominated plasma, where binary scattering between an electron and neutral molecules is significant, is of key importance to understanding collective properties and behaviour of r.f. or microwave plasmas. In these circumstances, systematic theoretical investigation of an r.f. electron (swarm) transport is highly desirable.

The topic dealt with in this paper is the maximal property of the cycle-averaged mean energy of an electron swarm in an r.f. field, with respect to the frequency of this field. Fig. 1 below shows the typical frequency dependence, as obtained from numerical solution of Boltzmann's equation (Makabe 1991). Clearly the cycle-averaged energy increases with frequency at first, attains a maximum and then decreases monotonically with a further increase of frequency. Notice for H₂ that the velocity modulation also has maximum amplitude when the cycle-averaged energy has a maximum. However in CH₄, although the maximal property for the cycle average energy is evident, the distinct maximum in the velocity modulation occurs at higher frequencies for reasons to be discussed subsequently. This

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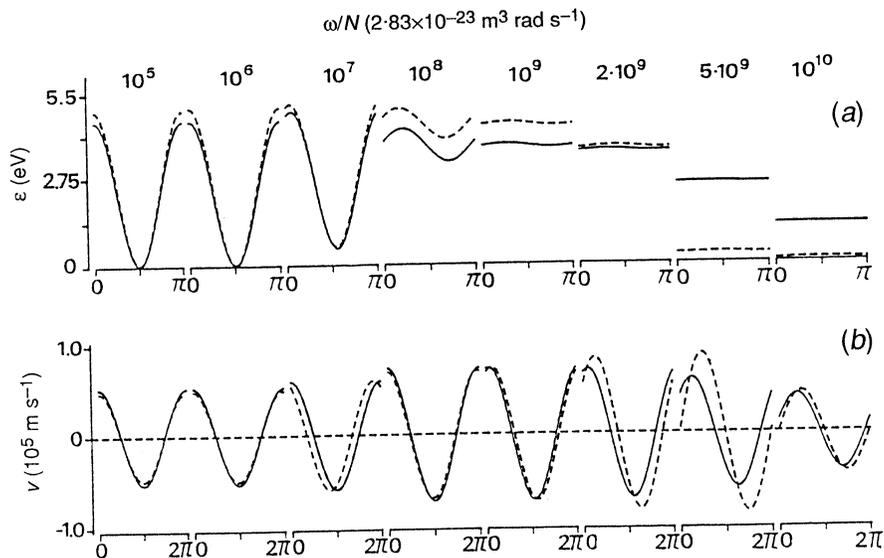


Fig. 1. Time modulation of the electron swarm parameters in H_2 (solid curve) and CH_4 (dashed curve) for different values of ω/N at $E/N = 50 \text{ Td}$: (a) ensemble averaged energy and (b) drift velocity.

maximal property in the cycle-averaged mean energy has also been observed by Yamanashi *et al.* (1991) and Loureiro (1993). We explain this phenomenon in Section 3 through an approximate, analytic solution of certain momentum and energy balance equations (Section 2). The results are augmented by a physical discussion in Section 4.

But first, a word about the philosophy of our approach and a caution. While there has been a great deal of activity in recent times in developing numerical solutions of Boltzmann's equation for r.f. swarms (Winkler *et al.* 1987; Capitelli *et al.* 1988; Ferreira *et al.* 1991; Makabe and Goto 1988; Goto and Makabe 1990; Makabe 1991; White 1993; Loureiro 1993), very little progress seems to have been made in developing *analytic* solutions, in stark contrast to the case for d.c. fields. The situation in the mid-1960s was reviewed by McDaniel (1964), while Loureiro (1993) provided a more modern perspective, particularly with regard to the 'effective field' concept, of direct relevance to the present paper. We feel that there is an identifiable need at this time to try to fill the gap in the understanding of r.f. swarms through provision of simplified arguments and mathematics, and the vehicle we use for this purpose is momentum transfer theory, well known in the theory of d.c. transport (Mason and McDaniel 1988). The equations furnished by this theory, which form the basis for almost the entire discussion, are reviewed in Section 2. The cautionary note is that the reader should be careful to distinguish between the two averaging processes referred to here: The 'mean energy' for example refers to the instantaneous average of energy over the electron swarm, which can be further averaged in time, e.g. over one cycle of the oscillating field. The latter process is designated 'cycle-averaging' in what follows.

2. R.F. Swarm Model Equations

(2a) Momentum-transfer Theory, Balance Equations

In this paper we shall adapt the balance equation of momentum-transfer theory to charged-particle swarms in a.c. electric fields. This theory is well known for providing physical insight and good semi-quantitative results (typical accuracies $\sim 10\%$) for transport properties of ions, electrons and positrons in electrostatic fields (Robson 1984, 1986; Robson and Ness 1988; Ness and Robson 1988; Mason and McDaniel 1988). It provides a very useful contrast to the detailed, rigorous numerical analysis normally associated with the solution of Boltzmann's equation, but of course will never be a true alternative to the latter when high accuracy ($\lesssim 1\%$) is required. The method is applicable to both heavy and light charged particles and can therefore be used to analyse weakly ionised gases used in plasma processing and gas laser excitation. The space-time varying fields occurring in these plasmas can be incorporated directly into the equations of momentum-transfer theory without further ado, since the only approximations involved are in the collision terms; the field terms are exact and are of the same mathematical form, regardless of whether the field varies in space-time or not. This paper therefore constitutes the first step in a natural progression of extension of the d.c. momentum-transfer results to a.c. fields.

The theory, at its lowest level, is similar in spirit to the 'relaxation continuum' model of Makabe *et al.* (1992), but the origin and definition of the collisional relaxation times is more precise. In essence, momentum-transfer theory provides a scheme for systematic approximation of collision terms in moment equations generated by integrating Boltzmann's equation for each charged species with 1 , $m\mathbf{v}$, $\frac{1}{2}mv^2$, ... over all particle velocities \mathbf{v} . In the lowest approximation, momentum-transfer theory consists in employing collision terms of the same mathematical form as for the Maxwell (constant collision frequency) model, but with cross sections evaluated according to actual energy dependencies. The accuracy of the approximation can be assessed internally and consistently. Details can be found in Robson (1984, 1986) and Mason and McDaniel (1988).

In order to simplify the discussion, we consider only light swarm particles and assume spatially uniform conditions. The gas is assumed cold. In that case, it can be shown that equations (8) and (9) of Robson and Ness (1988) simplify to

$$\frac{d}{dt}\langle v \rangle + \tilde{\nu}_m(\langle \epsilon \rangle)\langle v \rangle = \frac{eE}{m}, \quad (1)$$

$$\tau_e(\langle \epsilon \rangle)\frac{d}{dt}\langle \epsilon \rangle + \langle \epsilon \rangle = \frac{1}{2}M\langle v \rangle^2 - \Omega(\langle \epsilon \rangle) \quad (2)$$

respectively, where $\langle v \rangle$ is the average velocity in the direction of the field E , $\langle \epsilon \rangle$ the mean energy, and

$$\tilde{\nu}_m = \nu_m + \nu_I + \frac{2}{3}\langle \epsilon \rangle \nu'_a \approx \nu_m, \quad (3)$$

$$\tau_e = \left(\frac{2m}{M} \nu_m \right)^{-1}, \quad (4)$$

$$\Omega = \tau_e \left[\sum_i \bar{v}_i \epsilon_i + \nu_I \epsilon_I + \left(\nu_I + \frac{2}{3} \langle \epsilon \rangle \nu'_a \right) \langle \epsilon \rangle \right]. \quad (5)$$

We denote by m and M the masses of the swarm particle and neutral molecule respectively, while ν_m , ν_a , ν_I and \bar{v}_i denote ensemble averaged collision frequencies for momentum transfer, attachment, ionisation and the i th inelastic process respectively, while ϵ_I and ϵ_i denote the ionisation energy of the molecule and threshold energy for the i th inelastic process respectively.

D.C. field. If E is a constant in equation (1), then a steady state is eventually attained, i.e.

$$\frac{d\langle v \rangle}{dt} = 0, \quad \frac{d\langle \epsilon \rangle}{dt} = 0,$$

where the average ('drift') velocity $v_d \equiv \langle v \rangle$ and mean energy $\epsilon_d \equiv \langle \epsilon \rangle$ are given by the solution of the equations

$$\nu_m(\epsilon_d) v_d = eE/m, \quad (6)$$

$$\epsilon_d = \frac{1}{2} M v_d^2 - \Omega(\epsilon_d). \quad (7)$$

Once the cross sections Q are specified as functions of energy ϵ , collision frequencies can be found [e.g. $\nu_m(\epsilon_d) = N \sqrt{2\epsilon_d/m} Q_m(\epsilon_d)$] and the equations can be easily solved. [Note that cross sections characterised by sharp thresholds must be treated more carefully (Robson 1984).]

The following useful identities follow from direct differentiation of (6) and (7):

$$M v_d \frac{dv_d}{d \ln E} = (1 + \Omega') \frac{d\epsilon_d}{d \ln E}, \quad (8)$$

$$-M v_d^2 \frac{d \ln \nu_m}{d \epsilon_d} = (1 + \Omega') \frac{K'}{1 + K'}, \quad (9)$$

where $K = v_d/E$ is the mobility coefficient, and

$$\Omega' \equiv \frac{d\Omega}{d\epsilon_d}, \quad K' \equiv \frac{d \ln K}{d \ln E}. \quad (10)$$

Negative differential conductivity (NDC) arises when $1 + \Omega' < 0$ (Robson 1984).

A.C. field. We now consider harmonically varying fields

$$E(t) = E_0 \cos \omega t. \quad (11)$$

For simplicity, we henceforth write v for $\langle v \rangle$ and ϵ for $\langle \epsilon \rangle$ and hence equations (1) and (2) become

$$\dot{v} + \nu_m(\epsilon)v = \frac{eE(t)}{m}, \tag{12}$$

$$\tau_e(\epsilon)\dot{\epsilon} + \epsilon = \frac{1}{2}Mv^2 - \Omega(\epsilon) \tag{13}$$

respectively. These are nonlinear in v and ϵ and must in general be solved numerically. Although this can be readily carried out, the avenue which we explore in this paper is to obtain physical insight through analytic (albeit approximate) solution of the equations.

Before going into these details, it should be noted that quite general symmetry arguments can be invoked (Robson 1995, present issue p. 347) to demonstrate that $v(t)$ and $\epsilon(t)$ can be represented by Fourier series involving, respectively, only odd and even harmonics of ω , i.e. showing only leading terms (see also Goto and Makabe 1990, Margenau 1948; Margenau and Hartman 1948):

$$v(t) = v_0 \cos(\omega t - \varphi_m) + \dots, \tag{14}$$

$$\epsilon(t) = \epsilon_0 + \epsilon_1 \cos(2\omega t - \varphi_e) + \dots, \tag{15}$$

where v_0 , ϵ_0 , ϵ_1 , φ_m and φ_e are appropriate amplitudes and phase shifts respectively. This sort of representation is very useful in discussing the analytic solution of (12) and (13).

(2b) Approximate Solution of Balance Equations

We now write for the mean energy

$$\epsilon(t) = \epsilon_0 + \delta\epsilon(t), \tag{16}$$

where $\delta\epsilon(t)$ is the oscillatory part of the energy, assumed small compared with the cycle-averaged value ϵ_0 ,

$$\delta\epsilon \ll \epsilon_0. \tag{17}$$

As we shall see, this corresponds to a frequency regime such that

$$\omega\tau_e > 1. \tag{18}$$

In what follows, we thus linearise in the small quantity $\delta\epsilon$. Thus, for example, we expand

$$\nu_m(\epsilon) = \nu_m(\epsilon_0) + \delta\epsilon \nu'_m(\epsilon_0) + \dots, \tag{19}$$

$$\Omega(\epsilon) = \Omega(\epsilon_0) + \delta\epsilon \Omega'(\epsilon_0) + \dots \tag{20}$$

to first order in $\delta\epsilon$.

To this order of approximation, the solution of the momentum balance equation (12) in the periodic steady state (after all transients have decayed) is

$$v(t) = \frac{eE_0}{m} \int_0^\infty d\tau \cos \omega(t - \tau) \exp \left\{ - \int_0^\tau d\tau' \nu_m(\epsilon(t - \tau')) \right\} \\ \approx \frac{eE_0}{m} \int_0^\infty d\tau \cos \omega(t - \tau) e^{-\nu_m(\epsilon_0)\tau} \left\{ 1 - \nu'_m(\epsilon_0) \int_0^\tau d\tau' \delta\epsilon(t - \tau') \right\}. \quad (21)$$

If the energy balance equation (13) is integrated over a cycle and (20) is employed, we find that the cycle-averaged energy ϵ_0 satisfies

$$\epsilon_0 = \frac{1}{2} M v_{rms}^2 - \Omega(\epsilon_0), \quad (22)$$

where v_{rms} denotes the root-mean-square drift velocity. Then the linearised energy balance equation is

$$\tau_e(\epsilon_0) \delta\dot{\epsilon} + \delta\epsilon [1 + \Omega'(\epsilon_0)] = \frac{1}{2} M (v^2 - v_{rms}^2). \quad (23)$$

Henceforth, the ϵ_0 dependence of Ω , τ_e , etc. will not always be made explicit.

We seek solutions of (21) and (23) corresponding to the lowest-order time harmonics, as indicated in (14) and (15). Thus, if (14) is substituted into the r.h.s. of (23), we find

$$\delta\epsilon(t) = \epsilon_1 \cos(2\omega t - \varphi_e), \quad (24)$$

where

$$\epsilon_1 = \frac{\frac{1}{2} M v_{rms}^2}{1 + \Omega'} \cos \theta, \quad (25)$$

$$\varphi_e = \theta + 2\varphi_m, \quad (26)$$

$$\theta \equiv \tan^{-1} \left(\frac{2\omega\tau_e}{1 + \Omega'} \right). \quad (27)$$

If (24) is then substituted into the r.h.s. of (21) we obtain (14) once again with

$$v_0 = \frac{eE_0/m}{\sqrt{\omega^2 + \nu_m^2}} \left[1 - \frac{1}{4} p \cos \theta \cos(\varphi_e - \varphi_m) \cos \varphi_m \right], \quad (28)$$

$$\varphi_m = \varphi - \frac{1}{4} p \cos \theta \sin(\varphi_e - \varphi_m) \cos \varphi_m. \quad (29)$$

In these expressions

$$\varphi \equiv \tan^{-1}(\omega/\nu_m), \quad (30)$$

$$p \equiv \frac{M v_{rms}^2}{1 + \Omega'} \frac{d \ln \nu_m}{d \epsilon_0}. \quad (31)$$

The rms drift velocity is given by

$$v_{rms} = v_0/\sqrt{2} = \frac{eE_{eff}}{m\nu_m(\epsilon_0)}, \quad (32)$$

where

$$E_{eff} \equiv E_{eff}^{(0)}[1 - \frac{1}{4}p \cos \theta \cos(\varphi_e - \varphi_m) \cos \varphi_m], \quad (33)$$

$$E_{eff}^{(0)} \equiv \frac{E_0/\sqrt{2}}{\sqrt{1 + \omega^2/\nu_m^2(\epsilon_0)}} = \frac{E_0}{\sqrt{2}} \cos \varphi. \quad (34)$$

Equations (22) and (32) are of identical mathematical form to the d.c. equations (7) and (6) respectively. Hence, just as the d.c. field E determines the d.c. transport properties v_d and ϵ_d , so are v_{rms} and ϵ_0 determined by E_{eff} . For this reason, we may think of E_{eff} as constituting an equivalent or 'effective' d.c. field, for cycle-averaged a.c. transport properties. Note that $E_{eff}^{(0)}$ in equation (34) resembles a more conventional definition of effective field, sometimes employed in the analysis of a.c. transport (Ferreira *et al.* 1991; Loureiro 1993), although elsewhere the definition of ν_m often appears somewhat arbitrary. The value of E_{eff} differs from $E_{eff}^{(0)}$ through the factor in square brackets in (33), and this in turn differs from unity only over a certain range of frequencies, as explained below.

By (22), (24) and (25) it is clear that the assumption (17) holds if

$$\cos \theta = \frac{1 + \Omega'}{\sqrt{(1 + \Omega')^2 + (2\omega\tau_e)^2}} \quad (35)$$

is small and a sufficient condition for this to be true is if (18) holds. Beyond this, however, there is no restriction on the applied frequency.

Many of the results for the d.c. case carry over to the a.c. case in terms of E_{eff} . Thus (8) and (9) become

$$Mv_{rms} \frac{dv_{rms}}{d(\ln E_{eff})} = (1 + \Omega') \frac{d\epsilon_0}{d(\ln E_{eff})}, \quad (36)$$

$$Mv_{rms}^2 \frac{d(\ln v_{rms})}{d\epsilon_0} = -(1 + \Omega') \frac{K'_{rms}}{1 + K'_{rms}}, \quad (37)$$

where $\Omega' = d\Omega(\epsilon_0)/d\epsilon_0$ and

$$K'_{rms} = \frac{d(\ln K_{rms})}{d(\ln E_{eff})}, \quad (38)$$

$$K_{rms} \equiv v_{rms}/E_{eff}. \quad (39)$$

Equation (31) for the important parameter p can thus be written as

$$p = -K'_{rms}/(1 + K'_{rms}). \quad (40)$$

Finally, note that just as for the d.c. case, NDC can arise, in the sense that $dv_{rms}/dE_{eff} < 0$, if $1 + \Omega' < 0$.

3. Frequency Variation of Transport Properties

The variation of ϵ_0 and v_{rms} with ω is controlled entirely by $E_{eff}(\omega)$ and, since ϵ_0 is a monotonically increasing function of E_{eff} , it follows that if the effective field has an extremal at a certain frequency, then so will the cycle-averaged energy. That is, it is sufficient to explore the frequency dependence of the effective field (33) in order to understand how cycle-averaged properties depend upon frequency.

On the r.h.s. of (33), $E_{eff}^{(0)}$ is a monotonically decreasing function of ω and controls the behaviour of E_{eff} at high frequencies. In contrast, the term in square brackets may actually increase with frequency, provided that

$$p > 0, \quad (41)$$

and approaches unity in the high frequency limit. This combination of factors means that E_{eff} , and hence ϵ_0 and v_{rms} , may reach a maximum at some frequency, provided that (41) holds at that frequency.

We now focus on the frequency range

$$\nu_e < \omega \ll \nu_m \quad (42)$$

and observe that in this case (33) can be approximated by

$$E_{eff}(\omega) \approx \frac{E_0}{\sqrt{2}} \left(1 - \frac{1}{2}\omega^2\tau_m^2\right) \left(1 - \frac{A}{\frac{1}{2}\omega^2\tau_m^2}\right), \quad (43)$$

where

$$A \equiv \frac{1}{8}p(1 + \Omega')^2 \left(\frac{m}{M}\right)^2. \quad (44)$$

It is then readily shown that $\partial E_{eff}/\partial\omega = 0$, $\partial\epsilon_0/\partial\omega = 0$ when

$$\frac{1}{2}\omega^2\tau_m^2 = A^{\frac{1}{2}},$$

i.e.

$$\omega\tau_m = (p/2)^{\frac{1}{4}} (|1 + \Omega'|)^{\frac{1}{2}} \left(\frac{m}{M}\right)^{\frac{1}{2}} \quad (45a)$$

or equivalently

$$\omega\tau_e = \frac{1}{2}(p/2)^{\frac{1}{4}} (|1 + \Omega'|)^{\frac{1}{2}} \left(\frac{m}{M}\right)^{\frac{1}{2}}. \quad (45b)$$

Obviously (41) must hold in order for these equations to make sense physically. Given that we also expect $p \lesssim 1$ for most realistic situations, it is clear that equations (45) are consistent with the original assumption (42). Notice that the maximum value of effective field is

$$E_{eff} = E_0/\sqrt{2}(1 - A^{\frac{1}{2}})^2 \approx E_0/\sqrt{2}, \quad (46)$$

since A is small. Then maximum values of ϵ_0 , v_{rms} are found by substituting (46) in the r.h.s. of (31) and solving (22) and (31) simultaneously. Beyond this maximum, E_{eff} and ϵ_0 decrease monotonically with increasing frequency ω . This remark is also true for v_{rms} , unless a region of (cycle-averaged) NDC is encountered as the frequency is further increased, leading to an additional maximum in v_{rms} . We now examine the key requirement (41) in more detail. Obviously in the absence of ionisation, attachment and inelastic processes ($\Omega = 0$) and when $\nu_m = \nu_m(\epsilon_0)$ increases with ϵ_0 , equation (31) indicates that $p > 0$, and hence E_{eff} and ϵ_0 attain maxima. In this case (36) shows that v_{rms} is also a monotonically increasing function of E_{eff} , and therefore it, too, reaches a maximum at the same value of ω . The same remarks apply even in the presence of attachment, ionisation and inelastic processes ($\Omega \neq 0$), provided that $1 + \Omega' > 0$ at that frequency. On the other hand if at this frequency $1 + \Omega' < 0$ (a region of NDC in a cycle averaged sense), equation (41) cannot be satisfied unless ν_m is a decreasing function of energy, but this combination is improbable for realistic cases.

In summary, if ν_m is an increasing function of energy then, generally speaking, the cycle-averaged energy ϵ_0 and rms drift velocity v_{rms} simultaneously attain a maximum at a frequency satisfying (45). For combinations of elastic and cross sections such that $1 + \Omega' < 0$ in the relevant frequency range, however, no such extremal property is possible. An interesting exception to this general rule occurs when $1 + \Omega' < 0$ *outside* the frequency range for which ϵ_0 attains its maximum, for then v_{rms} may have an additional extremals as ω is varied. This is the case for CH_4 as shown in Fig. 1, as discussed below.

To make these results more concrete, we take a model where the electrons make only elastic collisions with the neutrals, the collision frequency being governed by a simple power law

$$\nu_m(\epsilon) \sim \epsilon^{(l+1)/2}, \quad (47)$$

where l is an arbitrary constant. In that case (22) and (31) together give

$$p = \frac{Mv_{rms}^2}{\epsilon_0} \frac{d(\ln \nu_m)}{d(\ln \epsilon_0)} = l + 1. \quad (48)$$

For the constant cross section model $l = 0$, $p = 1$ and the frequency for which energy reaches a maximum follows from (45a):

$$\omega\tau_m = 2^{-1/4} \left(\frac{m}{M} \right)^{\frac{1}{2}}. \quad (49)$$

We have also calculated the cycle-averaged mean energy over a range of frequencies for a constant cross section model by solving a multiterm Boltzmann r.f. code (White 1993) and the results are displayed in Fig. 2. The peak is quite broad, but the position of the maximum nevertheless appears to be very close to the predicted value (49).

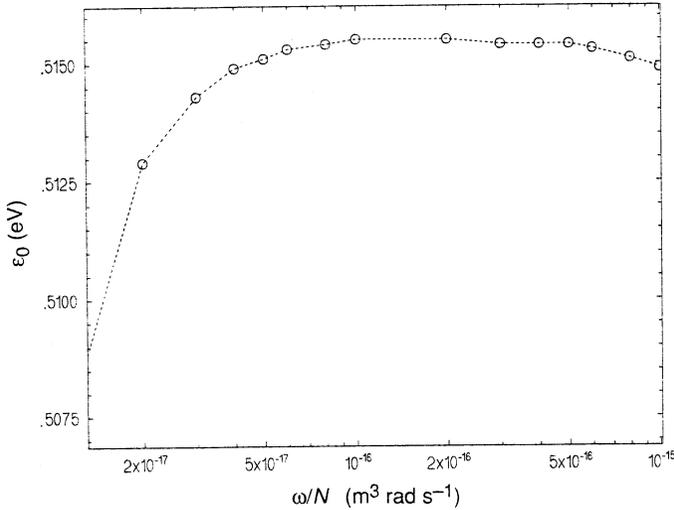


Fig. 2. Frequency variation of the cycle-averaged energy for the constant cross section model: $\sigma_m = 5 \text{ \AA}^2$, $E/N = 1 \text{ Td}$ and $T_0 = 0 \text{ K}$.

4. Discussion

The previous section gives the derivation of an expression (equation 45) for the frequency at which the cycle-averaged energy reaches a maximum, based upon the approximate momentum-transfer theory. It was shown that such a maximum is possible only if $\tau_e(\epsilon)$ decreases with energy. We now present a physical argument to supplement this mathematical discussion, following the thesis of Maeda (1993). For convenience, we refer to H_2 in Fig. 1. At low frequencies $\omega\tau_e < 1$, mean energy is a function of the instantaneous field and oscillates between a minimum of zero (where the field is zero) and a maximum value when the field is also a maximum. As frequency is increased, the minimum value of mean energy increases above zero, since $\omega\tau_e(\epsilon)$ is not small at low energies, and electrons cannot relax their energy. However, at higher energies, $\omega\tau_e(\epsilon)$ may in fact be small [$\tau_e(\epsilon)$ decreases with ϵ], allowing the electron energy to relax. The maximum value of ϵ attained in a cycle therefore remains virtually unchanged, but the minimum value increases, as frequency is increased. Therefore the cycle-averaged energy increases with frequency at first. As ω is further increased, however, $\omega\tau_e > 1$ over the entire cycle and electron energy cannot be relaxed. The modulation is therefore weaker and the cycle-averaged energy attains a maximum in this range of frequencies. Finally, at high frequencies when $\omega\tau_m > 1$, electrons are trapped by the oscillating field and energy is not fed in. As a result the mean energy falls with increasing frequency. The prediction on physical grounds is thus that the cycle-averaged mean energy has a maximum somewhere in the range specified by

(42), provided that the relaxation time decreases with increasing energy. This is consistent with the restriction (41) in the corresponding mathematical treatment.

For CH_4 in Fig. 1, this also follows the general pattern referred to above, but in contrast to H_2 , v_{rms} actually appears largest at frequencies well above the frequency corresponding to the maximum ϵ_0 . In fact, ϵ_0 falls dramatically while v_{rms} is still rising. This is the 'signature' of NDC (Robson 1984), and what is observed can be explained as in Section 3 in terms of an *additional* maximum in v_{rms} at high frequencies, following on from the (shallow) maximum in v_{rms} which occurs simultaneously with maximum ϵ_0 . This property of the drift velocity in CH_4 was observed and discussed by Goto and Makabe (1990) and displayed in their Fig. 8.

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