Hydrodynamic approximation for ions in high-pressure rf glow discharges

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(Received 21 June 1994)

The use of the hydrodynamic approximation to describe the motion of ions in weakly ionized plasmas subjected to a high-frequency external electric field is critically examined. It is shown that if the frequency of this field is comparable with or larger than the ion collision frequency, then the response of the ions to the alternating electric field cannot be described by ion mobility. A possible alternative approach to the problem is presented.

PACS number(s): 52.25.Dg

rf glow discharges in plasmas have been widely used for many years in the microelectronics industry for materials processing, where low gas pressures of up to around 1 Torr are mostly used [1]. More recently, they have become of interest for use in exciting CO_2 and other gas lasers [2,3], in which case gas pressures of at least 30 Torr are generally used. A detailed understanding of the behavior of the particle densities and electric field in the sheath regions adjoining the electrodes is obviously important for materials processing, which takes place at the electrodes. However, such an understanding is also of importance for the plasmas used in gas lasers, even though the regions near the electrodes are not of major importance for the lasing process. At the pressures used in gas lasers, as opposed to those used for materials processing, the sheath regions do not screen the applied field from the positive column. In this case, the importance of the fields and consequent potential drops across the sheaths is that they determine the relationship between the applied voltage and the electric field in the positive column of the plasma, and it is the field there that determines the ionization and excitation rates of the molecules that lead to the operation of the laser. At the higher pressures used in this case, the mean free paths of the electrons and ions are much less than the width of the sheath region. For both pressure ranges, the problem of calculating self-consistently the electron and ion densities and velocities and the associated electric field is an extremely difficult one. Instead of attempting to solve the Boltzmann equation for the electron and ion distributions in phase space, use is often made of the fluid model or hydrodynamic approximation, in which electron and ion transport is described with the continuity and momentum transfer equations [4,5]. However, the equations for ions are expressed in terms of the ion mobility, and the use of this involves a very basic problem, namely the strong frequency dependence of the ionic mobility under the usual experimental conditions. While it is often stated that the ion motion cannot follow the high frequency rf field, the implications of this statement for the equations governing the system are usually ignored. In this paper, we examine this problem and describe one possible way of overcoming it by using a standard technique [6,7].

In order to treat a definite system, we consider the simplest possible one, namely that of a plasma between plane parallel electrodes, and also assume that the electron and ion mean free paths are sufficiently short for the particles to respond essentially to the local electric field. Let $F(x,t)$ denote the electric field in the plasma, N_e and N_i the electron and ion densities, respectively, μ_e and μ_i the electron and ion mobilities, respectively, α the ionization coefficient, and β the recombination coefficient. Then, if for the sake of simplicity, one ignores the contributions from diffusion to the electron and ion currents, one obtains the following set of three coupled nonlinear partial differential equations to describe the plasma in the hydrodynamic approximation:

$$\partial N_e/\partial t - \mu_e(\partial/\partial x)(N_e F) = \alpha\mu_e|N_e F| - \beta N_i N_e \equiv R(x,t) \ , \tag{1}$$

$$\partial N_i/\partial t + \mu_i(\partial/\partial x)(N_i F) = \alpha\mu_e|N_e F| - \beta N_i N_e \equiv R(x,t) \ , \tag{2}$$

$$\partial F/\partial x = (e/\varepsilon_0)(N_i - N_e) \ . \tag{3}$$

The above equations superficially look very reasonable, and have been used(with or without the addition of the diffusion components to the electron and ion currents) by various authors to calculate the temporal and spatial dependence of the electric field and of the electron and ion densities and currents [4,5]. However, if a rf potential of frequency ω is applied between the electrodes, the electric field F will have a major component of this frequency, and also harmonics at multiples of this frequency since the system of equations is nonlinear. According to the standard theory, the velocity $\mathbf{v}$ of a particle of mass m and charge q having an effective collision frequency ν_m in a field E satisfies the equation [8]

$$m d\mathbf{v}/dt + m\nu_m \mathbf{v} = q\mathbf{E} \ . \tag{4}$$

Hence the mobility $\mu(\omega)$ of such a particle in a field of frequency ω is just

$$\mu(\omega) = (|q|/m)/(\nu_m - i\omega) \ . \tag{5}$$

*Deceased.

Under the usual operating conditions, the frequency ω is much greater than the plasma frequency ω_{pi} and the collision frequency ν_{mi} for the positive ions but much less than the corresponding frequencies ω_{pe} and ν_{me} for the electrons. For instance, at a typical frequency of 50 MHz, $\omega = 3\times10^{8}\ \mathrm{s}^{-1}$, and for a pressure of 30 Torr, with 10^{-7} of the molecules ionized, $\omega_{pi}\approx 6\times10^{7}\ \mathrm{s}^{-1}$ and $\omega_{pe}\approx 6\times10^{9}\ \mathrm{s}^{-1}$, while for a collision cross section of $10^{-16}\ \mathrm{cm}^{-2}$ one finds that for ions at room temperature and for electrons with energy 1 eV (typical for gas discharges), $\nu_{mi}\approx 4\times10^{6}\ \mathrm{s}^{-1}$ and $\nu_{me}\approx 7\times10^{9}\ \mathrm{s}^{-1}$. Thus for the electrons it is a reasonable approximation to use the static mobility $\mu_e(0)=e/m\nu_{me}$ in Eq. (1) even for the time-dependent field. For the ions, on the other hand, the static mobility which determines their response to the dc field in the sheath region is just

$$\mu_i(0)=e/m\nu_{mi} \ , \tag{6}$$

but the mobility that determines the response to the component of the field having the fundamental frequency ω of the applied potential is

$$\mu_i(\omega)\approx ie/m\omega \ , \tag{7}$$

since $\nu_{mi} \ll \omega$, while that appropriate to the response to the higher harmonics depends on the harmonic being considered. Since Eq. (2) is an equation in the time domain rather than in the frequency domain, it is certainly not valid to use in it a single constant ion mobility μ_i. Such a problem is well known in the theory of nonexponential dielectric response functions, for instance, where the system can be described either in terms of a frequency-dependent dielectric response or a time-dependent response function [9]. In our system, since the field in the plasma sheath contains both dc and high-frequency components, one cannot even introduce the equivalent of a time-dependent ion mobility $\mu_i(t)$ into Eq. (2) in place of the constant μ_i. Thus we conclude that not only does Eq. (2) not provide a reasonable approximation for the description of the ion motion, but also that it cannot be modified simply to provide such a description. This is the main point that we wish to make in this paper.

The simplest way to overcome this problem is to consider only the average motion of the ions, and to use the standard theory [6,7] for this motion in a field having a rapidly oscillating component. According to this theory, for an ion of charge e moving in a field

$$F(x,t)=E_0(x)+\sum_k E_k(x)\exp(-ik\omega t) \ , \tag{8}$$

the average motion is obtained by replacing the field $E(x,t)$ by the effective static field

$$E_{\mathrm{eff}}(x)=E_0(x)+(e/2m_i\omega^2)\sum_k (E_k dE_k/dx)/k^2 \ . \tag{9}$$

It follows that Eq. (2) should be replaced by

$$\mu_i(0)(\partial/\partial x)[N_i E_{\mathrm{eff}}(x)]=\langle R(x,t)\rangle \ . \tag{10}$$

The major problem with using Eq. (10) is that Eqs. (1) and (3) determine the development of the field $F(x,t)$ as a function of time, while Eq. (10) requires the Fourier components $E_k(x)$ of this field. Hence it will probably be necessary to use an iterative method to solve the set of Eqs. (1), (10) and (3). While this is much more difficult than the method currently used of just finding the numerical solution of the set of Eqs. (1), (2) and (3), it avoids the error of using an ionic mobility in Eq. (2).

The conclusion from this paper is that the hydrodynamic approximation with a constant ionic mobility cannot be used for ions in a weakly ionized plasma where the frequency of the driving field is comparable with or greater than the collision frequency of the ions with neutral atoms or molecules. On the other hand, even the numerical solution of the Boltzmann transport equations for the ions does not seem very practicable. Instead, the best approach within the hydrodynamic approximation is probably one which makes use of the effective time-averaged field experienced by the ions, as described above.

The authors gratefully acknowledge the receipt of financial support for this work from the Israel Ministry of Science and Arts and from the Research Authority of Bar-Ilan University.

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