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Relaxation processes of electrons and positrons in gases in electric and magnetic fields crossed at arbitrary angles

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Abstract. A multi-term solution of the Boltzmann equation has been developed and used to investigate the temporal relaxation of charged-particle swarms under the influence of electric and magnetic fields crossed at arbitrary angles. A Monte Carlo simulation technique has been used to verify the Boltzmann equation analysis under hydrodynamic conditions and for an independent study of spatial relaxation of the electrons under non-hydrodynamic conditions. We present results for model and real gases highlighting the explicit influence of the magnetic field and non-conservative collisions on temporal and spatial relaxation characteristics, including the existence of transiently negative electron diffusivity. We present results for thermalization of the positrons in nitrogen highlighting the applicability of our theory for contemporary kinetic studies in the domain of positron physics.

1. Introduction

The theoretical investigation of charged-particle swarms moving in an unbounded gas in electric and magnetic fields is a topic of great interest both as a problem in basic physics and for its potential for application to modern technology. One of the major challenges in these investigations is an accurate representation of temporal and spatial relaxation of charged-particle swarms. Relaxation processes of a swarm of charged particles are related to various problems of gaseous electronics such as modeling of non-equilibrium plasma discharges, high-speed switching technique, swarm physics and physics of gas lasers [1]. The knowledge of temporal relaxation is essential for a better understanding of electron-molecule interaction as well as for a better understanding of transient transport phenomena in gases such as transient negative electron mobility [2] or transient negative electron diffusivity [3]. In the context of plasma modeling, the relaxation times of the electron transport properties are necessary input data for fluid plasma models such as the relaxation continuum model [4]. In addition, to fully appreciate the complex structure of the transport properties in radio-frequency (rf) electric and magnetic fields [5,6], a systematic investigation of the temporal relaxation of a swarm of charged particles in dc electric and magnetic fields is required.

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While the studies of temporal relaxation of the electrons are tremendously important from the point of view of fundamental physics, modern non-hydrodynamic studies of spatial relaxation of the electrons are motivated by the need to understand the electron kinetics in low-temperature plasmas, especially in the neighborhood of sources and boundaries. A number of theoretical methods to calculate the electron transport properties under these "non-hydrodynamic" conditions, especially those attempting to model or simulate the cathode fall of a dc [7] or sheath of rf discharges [8], revealed the non-local nature of electron transport. One of the principal aims of this paper is to study the non-local nature of electron transport when a magnetic field is present and non-conservative collisions are operative.

We begin this paper with a brief review of multi-term theory for solving the Boltzmann equation valid for both electrons and ions in time-dependent electric and magnetic fields crossed at arbitrary angles, incorporating the effects of non-conservative collisional processes. We focus on two situations: (i) temporal relaxation of the electrons and (ii) thermalization of the positrons. In addition to the Boltzmann equation analysis, we discuss our hydrodynamic and non-hydrodynamic Monte Carlo simulation codes. The former is used to test the Boltzmann equation analysis under the hydrodynamic conditions. The latter is used for studies of the effects of magnetic fields and non-conservative collisions on spatial relaxation of the electrons. After giving brief reviews of theoretical methods, we then give numerical examples for various cases of special interest, highlighting recent new results.

2. Theory

2.1. Multi-term theory for solving the Boltzmann equation

The behaviour of charged particles in gases under the influence of electric and magnetic fields is described by the phase-space distribution function $f(\mathbf{r},\mathbf{c},t)$ representing the solution of the Boltzmann equation

$$\frac{\partial f}{\partial t}+\mathbf{c}\cdot\frac{\partial f}{\partial \mathbf{r}}+\frac{e}{m}\left[\mathbf{E}+\mathbf{c}\times\mathbf{B}\right]\cdot\frac{\partial f}{\partial \mathbf{c}}=-J\left(f,f_0\right), \tag{1}$$

where **r** and **c** denote the position and velocity co-ordinates, e and m are the charge and mass of the swarm particle and t is time. The electric and magnetic fields are assumed to be spatially homogeneous with magnitudes **E** and **B** respectively. In what follows, we employ a co-ordinate system in which **E** defines the z-axis while **B** lies in the y-z plane, making an angle ψ with respect to the **E**. Swarm conditions are assumed to apply where the charged particle number density is much less than number density of neutral species and mutual interactions between swarm particles are negligible compared with swarm particle-neutral particle interactions. The right-hand side of Eq. (1) denotes the linear charged particle-neutral molecule collision operator, accounting for elastic, inelastic and non-conservative (e.g. ionizing and attaching) collisions. For elastic collisions we use the original Boltzmann collision operator [9], while for inelastic collisions we prefer the semiclassical generalization of Wang-Chang et al. [10]. The attachment and ionization collision operators employed are detailed in Ref. [11].

(a) Spherical-harmonic expansion. The directional dependence of the phase-space distribution function in velocity space is represented in terms of a spherical harmonic expansion

$$f\left(\mathbf{r},\mathbf{c},t\right)=\sum_{l=0}^{l_{\max}}\sum_{m=-l}^{l}f_m^{(l)}\left(\mathbf{r},c,t\right)Y_m^{[l]}\left(\hat{\mathbf{c}}\right), \tag{2}$$

where $\hat{\mathbf{c}}$ represents the angles of **c**. The value of l_{max} is incremented until some predefined accuracy criterion is satisfied. This value indicates the deviation of the velocity distribution function from isotropy.

(b) Density gradient expansion. Assuming the hydrodynamic regime, the spatial dependence is represented by

$$f_m^{(l)}(\mathbf{r},c,t)=\sum_{s=0}^{2}\sum_{\lambda=0}^{s}\sum_{\mu=-\lambda}^{\lambda}f(lm\,|\,s\lambda\mu;c,t)G_\mu^{(s\lambda)}n(\mathbf{r},t)\,, \tag{3}$$

where $G_\mu^{(s\lambda)}$ is the irreducible gradient operator [11]. Truncation at s = 2 is necessary to determine transport coefficients up to and including diffusion when non-conservative collisions are operative.

(c) Sonine polynomial expansion. The speed dependence of the coefficients in Eq. (3) is represented by an expansion about a Maxwellian at an arbitrary temperature T_b, in terms of Sonine polynomials

$$f(lm\,|\,s\lambda\mu;c,t)=w(\alpha,t)\sum_{\nu=0}^{\infty}F(\nu lm\,|\,s\lambda\mu;\alpha,t)R_{\nu l}(\alpha c)\,, \tag{4}$$

where

$$R_{\nu l}(\alpha c)=N_{\nu l}\left[\frac{\alpha c}{\sqrt{2}}\right]^l S_{l+1/2}^{(\nu)}\left(\frac{\alpha^2c^2}{2}\right), \tag{5}$$

$$w(\alpha,c)=\left[\frac{\alpha^2}{2\pi}\right]^{3/2}\exp\left\{-\frac{\alpha^2c^2}{2}\right\}, \tag{6}$$

$$\alpha^2=\frac{m}{kT_b}, \tag{7}$$

$$N_{\nu l}^2=\frac{2\pi^{3/2}\nu!}{\Gamma(\nu+l+3/2)}\,, \tag{8}$$

and $S_{l+1/2}^{(\nu)}\left(\frac{\alpha^2c^2}{2}\right)$ are Sonine polynomials. Using the appropriate orthogonality relations of the spherical harmonics and modified Sonine polynomials the following system of coupled differential equations for the moments $F(\nu lm\,|\,s\lambda\mu;\alpha,t)$ is generated:

$$\sum_{\nu'=0}^{\infty}\sum_{l'=0}^{\infty}\sum_{m'=-l'}^{l'}\Big[\partial_t\delta_{\nu\nu'}\delta_{ll'}\delta_{mm'}+n_0J_{\nu\nu'}^{l}\delta_{ll'}\delta_{mm'}+\omega(000)\delta_{\nu\nu'}\delta_{ll'}\delta_{mm'}$$

$$+i\frac{q}{m}E(lm\,|\,l'm10)\alpha_t\left\langle\nu l\middle\|K^{[1]}\middle\|\nu'l'\right\rangle\delta_{mm'}$$

$$+\frac{qB}{m}\left\{\frac{\sin\psi}{\sqrt{2}}\left[\sqrt{(l-m)(l+m+1)}\delta_{m'm+1}-\sqrt{(l+m)(l-m+1)}\delta_{m'm-1}\right]-im\cos\psi\delta_{m'm}\right\}\delta_{\nu\nu'}\delta_{ll'}$$

$$-n_0J_{0\nu'}^{0}F(\nu lm\,|\,000)(1-\delta_{s0}\delta_{\lambda0}\delta_{\mu0})\delta_{l'0}\delta_{m'0}\Big]F(\nu'l'm'\,|\,s\lambda\mu)=X(\nu lm\,|\,s\lambda\mu)\,, \tag{9}$$

where $J_{\nu\nu'}^{l}$ and $\left\langle\nu l\middle\|K^{[1]}\middle\|\nu'l'\right\rangle$ are reduced matrix elements, $\omega(000)$ represents the net creation rate and $(lm\,|\,l'm10)$ is a Clebsch-Gordan coefficient. The explicit expressions for the RHS are given in Ref. [12] for the case of crossed dc electric and magnetic fields. These expressions are modified in this paper in order to consider the time-dependent fields crossed at arbitrary angles in the presence of non-conservative collisions. Discretising in time using an implicit finite difference scheme converts the system of coupled differential equations into a hierarchy of coupled matrix equations. To establish the transport coefficients of interest we are required to solve the following members of the hierarchy (s,λ,μ) = (0,0,0), (1,1,0), (1,1,1), (2,0,0), (2,2,0), (2,2,2). The explicit expressions for both bulk and

flux transport coefficients including the mean energy and gradient energy vector components are given in Ref. [6]. Transport coefficients of interest in this work include the mean energy (ε), drift velocity components (W_x, W_y, W_z) and diagonal diffusion tensor elements (D_{xx}, D_{yy}, D_{zz}).

2.2. Monte Carlo simulation technique

The other method for investigating the transient effects of charged-particle transport in dc electric and magnetic fields is a Monte Carlo simulation technique. In the context of hydrodynamic studies, it is assumed that the charged-particle swarm develops in an infinite space. At time $t = 0$, electrons are initially released from the origin according to Maxwellian velocity distribution with the mean starting energy of 1 eV under the influence of electric field only. After relaxation to the steady state in which both the swarm transport parameters and distribution function do not change in time, the magnetic field is applied and the relaxation process is followed accurately in time. An extremely large number of electrons (typically 10^7) have been followed in a neutral gas in order to obtain good statistics of the output data, particularly the diffusion coefficients. It is assumed that the electron density is sufficiently small so that Coulomb interaction between the electrons as well as shielding of the filed is negligible. All calculations are performed at zero gas temperature.

Spatial relaxation of the electrons has been studied by our steady-state Townsend Monte Carlo simulation code. The primary electrons are isotropically released one by one from the cathode surface into the half space with an initial energy ε_0. When an ionization collision occurs, a set of all dynamic properties (the moment of an ionization collision, the position of new electron, the starting energy and velocity) of secondary electron are established and placed at the stack. When primary electron reaches the anode surface or disappears in an attachment collision event, the first available electron from the stack is followed. These secondary electrons from the stack are released isotropically. In an attachment collision the electron is consumed and hence not followed further. If the stack is empty, the next primary electron is released and the whole procedure repeats. Thermal motion of the background neutral particles and electron-electron interactions are neglected. The electrodes are considered to be perfectly absorbing.

In both the hydrodynamic and non-hydrodynamic versions of Monte Carlo codes, we follow the spatiotemporal evolution of each electron through time steps governed by the minimum of two relevant time constants: mean collision time and cyclotron period for **E**×**B** field. These finite time steps are used to solve the integral equation for the collision probability in order to determine the time of the next collision. Once the moment of the next collision is established, the nature of the collision is determined by using the relative probabilities of the various collision types. All electron scattering is assumed to be isotropic regardless of the collision nature.

The way of sampling of transport properties depends upon the manner of swarm observation [13]. The definitions and corresponding formulae for the electron transport coefficients under the hydrodynamic conditions were given in our previous publications [14]. The spatially resolved electron transport properties have been obtained using the following formula [13]:

$$\langle \xi \rangle_j = \frac{\frac{1}{\Delta z}\int_{z_j-\Delta z/2}^{z_j+\Delta z/2} \xi f_{SST}(z,\mathbf{v})\,dz dv}{\frac{1}{\Delta z}\int_{z_j-\Delta z/2}^{z_j+\Delta z/2} f_{SST}(z,\mathbf{v})\,dz dv} \approx \frac{\sum_{k=1}^{N} \xi_k^j \Delta t_k^j}{\sum_{k=1}^{N} \Delta t_k^j}, \tag{10}$$

where $f_{SST}(z,\mathbf{v})$ is the steady state distribution function, ξ_k^j is the value of the quantity to be sampled when kth electron is contained in jth box, Δt_k^j is the residence time of the electron in that box and N is the total number of electrons which appear there. Electrons moving towards both the cathode and anode must be considered and sampled.

3. Results and discussion

3.1. Temporal relaxation of the electrons

In this section we consider the response of the electron transport coefficients to the application of a magnetic field under hydrodynamic conditions. Similar studies have been published previously for neon using the two-term approximation [15]. We extend these studies by (i) overcoming the inherent inaccuracies of the two-term approximation and (ii) addressing the temporal relaxation of spatial inhomogenities through a study of the diffusion tensor. In addition, this work is an extension of the work in [16] to consider the explicit influence of non-conservative collisions and the angle between the fields on the temporal relaxation profiles. Two specific situations have been considered: (i) the conservative Reid ramp model gas system [17]; and (ii) the ionization model gas of Lucas and Saelee [18]. The former has become the standard test for Monte Carlo simulations and Boltzmann equation solutions, particularly in light of known failure of the two-term approximation for this model. The latter is significant for demonstrating just how important it is to consider the ionization as a true non-particle conserving collision, and not simply just another inelastic process, as is sometimes done [21]. The initial conditions represent the steady state magnetic field free case where the electron swarm is acted on solely by a dc electric field (E/n_0 = 12 Td; B/n_0 = 0 Hx; 1 Td = 10^{-21} Vm^2; 1Hx = 10^{-27} Tm^3). At time t = 0, a magnetic field is switched on and the relaxation properties are monitored as a function of time (normalized time n_0t).

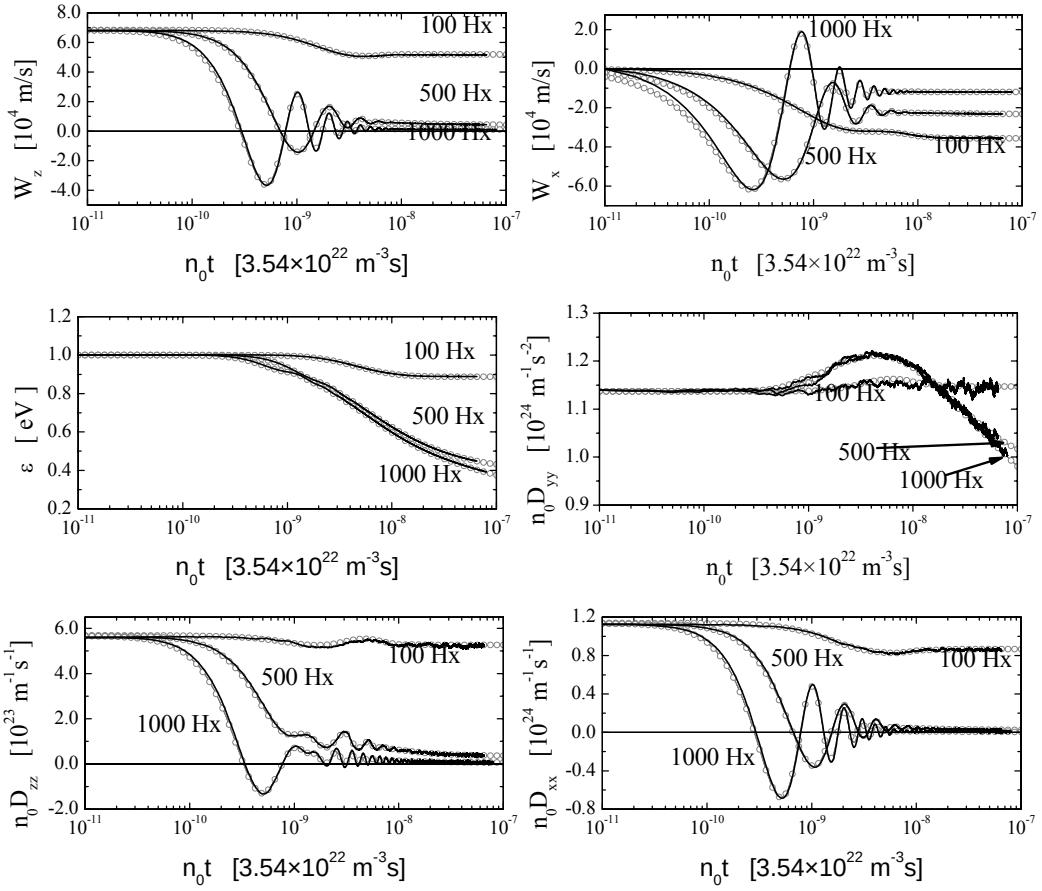

Figure 1. Temporal relaxation of the mean energy, drift velocity components and diagonal elements of the diffusion tensor for various applied magnetic fields in a crossed field configuration for electrons for Reid ramp model (solid lines-Monte Carlo; open symbols-Multi-term Boltzmann equation).

In Fig.1 we show the temporal relaxation profiles of the electron transport coefficients for the Reid ramp model for various B/n_0 in a crossed field configuration. The results obtained by multi-term theory for solving the Bolztmann equation are compared with those obtained by Monte Carlo simulation technique. The excellent agreement between these methods validates the basis of transport theory as well as numerical integrity of both numerical codes. While the mean energy and B-component of the diffusion tensor show monotonic relaxation profile, the relaxation profiles of the drift and diffusion in the **E** and **E×B** direction exhibit a transition from monotonic decay to damped periodic decay as magnetic field increases. For these damped periodic profiles, the oscillations are on the time-scale of the gyro-orbits τ and the envelope decays on a time-scale for momentum relaxation τ_m together with a further relaxation on the time-scale of the energy relaxation time τ_e. Perhaps the most striking phenomena is the existence of transiently negative excursions of the diffusion tensor elements in both the **E** and **E×B** directions. The existence of transiently negative diagonal diffusion elements in swarms was observed in CO_2 [16] as well as in rf fields [19] and is explained in greater detail in [3].

In Fig. 2 we show the temporal relaxation profiles of D_{yy} as a function of the magnetic field strengths and angle between the fields. The results obtained by the multi-term theory (MT) are compared with those obtained by the two-term approximation (TTA) for solving the Boltzmann equation. In the early and intermediate stages of the relaxation process, the significant deviations between the temporal profiles can be observed. This is a clear indication that the initial distribution function and its initial evolution deviates substantially from isotropy in velocity space. However, as magnetic field and the angle between the fields increase, the deviations between TTA and MT results are significantly diminished. This suggests that the magnetic field acts to destroy the anisotropy of the velocity distribution function, consequently inducing enhanced convergence in the *l*-index. A similar effect was observed in methane [20].

Another striking property is the high sensitivity of the relaxation profiles to the angle between the fields. In contrast to other diagonal elements of the diffusion tensor, the oscillatory nature of D_{yy} is reduced as the angle between the fields is increased. Finally, in the limit of an orthogonal field configuration these profiles are monotonic (see Fig. 1). For an orthogonal field configuration the Lorentz force does not act in this direction and hence there are no imprinted oscillations on the diffusion coefficient in this direction. On the other hand, for small angles between the fields, the electrons are under the action of Lorentz force producing the oscillatory relaxation profiles. For a magnetic field of 1000 Hx and angle of 30^o between the fields, the Lorentz force is shown to induce the negative transient diffusivity.

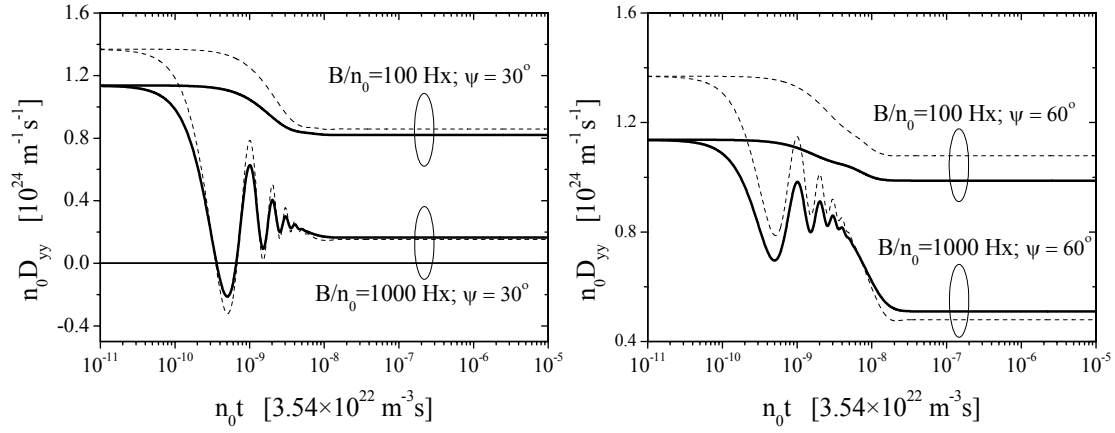

Figure 2. Temporal relaxation of D_{yy} for magnetic fields strengths of 100 and 1000 Hx and angles of 30 and 60 degrees between the fields for Reid ramp model (solid lines-MT; dashed lines-TTA)

In Fig. 3 we demonstrate the influence of non-conservative collisions on temporal relaxation of W_y and W_z for Lucas Saelee ionization model. We show the bulk and flux values of the drift velocity components as a function of various magnetic fields and angles between the fields. The parameter F controls the ionization degree. In the early stage of the relaxation process there is no distinction between the bulk and flux values of W_y indicating there is no spatial variation of the average electron energy in this direction. On the other hand, there is a clear distinction between the bulk and flux values for W_z. This suggests a strong variation of the average energy along the swarm in this direction. However, as the relaxation process proceeds in time, the synergism of magnetic field and ionization collisions on the temporal relaxation profiles becomes clearly evident. While the distinction between the bulk and flux values for W_y is more pronounced, the distinction for W_z is firstly diminished at the intermediate stage of a relaxation process and then upon reaching the steady state there are no differences between the bulk and flux values. These results suggest that due to complex interplay of the action of the magnetic field and the energy and momentum dissipation of the electrons in collisions, a complicated redistribution of high energetic electrons occurs.

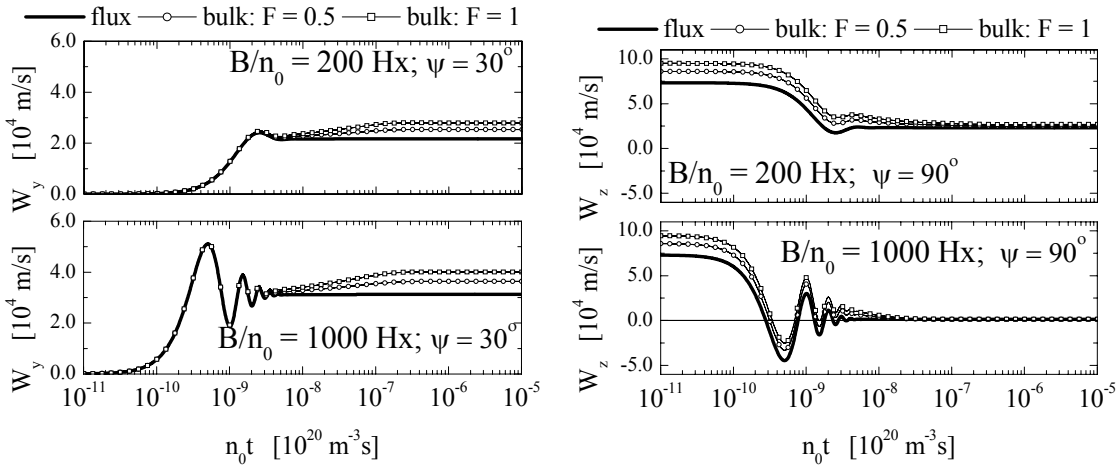

Figure 3. Temporal relaxation of the bulk and flux drift velocity components W_y and W_z for magnetic fields strengths of 200 and 1000 Hx and angles of 30 and 60 degrees between the fields for the ionization model of Lucas and Saelee.

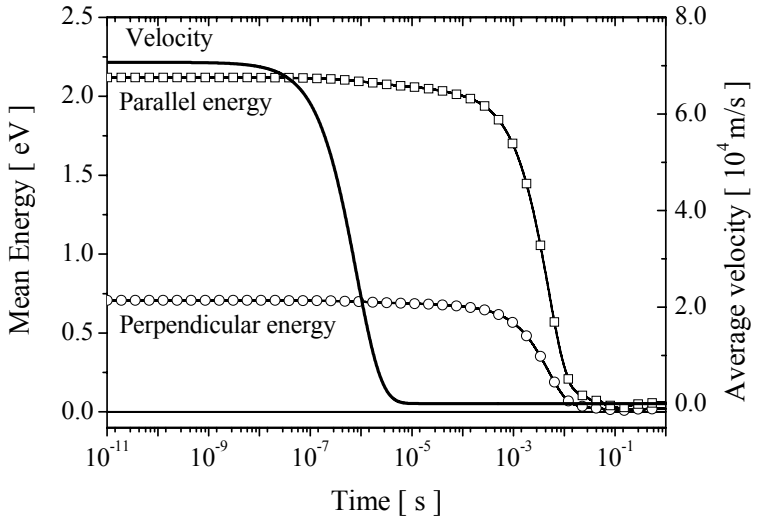

Figure 4. The relaxation times for thermalization of positrons in pure nitrogen.

3.2. Thermalization of the positrons

One of the key issues for optimum design and control of the positron traps is a knowledge of the relaxation times for thermalization of the positrons. As an initial study we investigated the relaxation time of positrons in pure nitrogen using a set of cross sections compiled in our laboratory. The subject of our study is a swarm of positrons initially with a drifted Maxwellian distribution (1.5×10^4 K with a drift of 7.0×10^4 m/s) entering the chamber. In Fig. 4 we demonstrate the thermalization of the mean energy (parallel refers to the initial drift direction) and the drift velocity. We can see that the thermalization time for the drift velocity is of the order of microseconds as compared with the thermalization times for energy being of the order of 10s of milliseconds.

3.3. Spatial relaxation of the electrons

In this section we demonstrate the ability of a magnetic field to control the spatial relaxation of the electrons. We extend the previous work on the idealised steady-state Townsend experiment [21] to include the explicit influence of the angle between the fields on spatial relaxation of the electrons when non-conservative collisions are operative.

In the case of an electric field only, previous work on the electron spatial relaxation revealed the complex nature of the relaxation process and associated basic mechanisms. The nature of the spatial relaxation profiles is dependent on the interplay between the power dissipated in elastic collisional processes, power dissipated in threshold collisional processes and the power deposited into the swarm by the field [21]. For certain gases, there exists a "window" of electric field strengths where the relaxation profiles are damped oscillatory in nature, and outside this electric field window the profiles are monotonic.

Before considering the effects of a magnetic field on spatial relaxation of the electrons we present one particularly interesting set of results associated with the spatial relaxation of the electrons in gas mixtures. In Fig. 5 we show the spatial relaxation of the mean energy in pure argon and mixtures of argon and CF_4 at E/n_0 of 15 Td. The cross sections for argon are displayed and detailed in [22] while the cross sections for CF_4 are given in [23]. We observe that the mean energy in pure argon exhibits a damped oscillatory relaxation along a decaying profile. However, by introducing a small amount of molecular admixture (e.g. CF_4) the oscillations are firstly suppressed and then entirely quenched in the limit of higher concentration of CF_4. By introducing a molecular admixture other collision processes, preferentially lower threshold vibrational excitations are introduced. These new collision processes lead to more efficient damping than elastic collisions, by virtue of larger and different energy loss mechanisms.

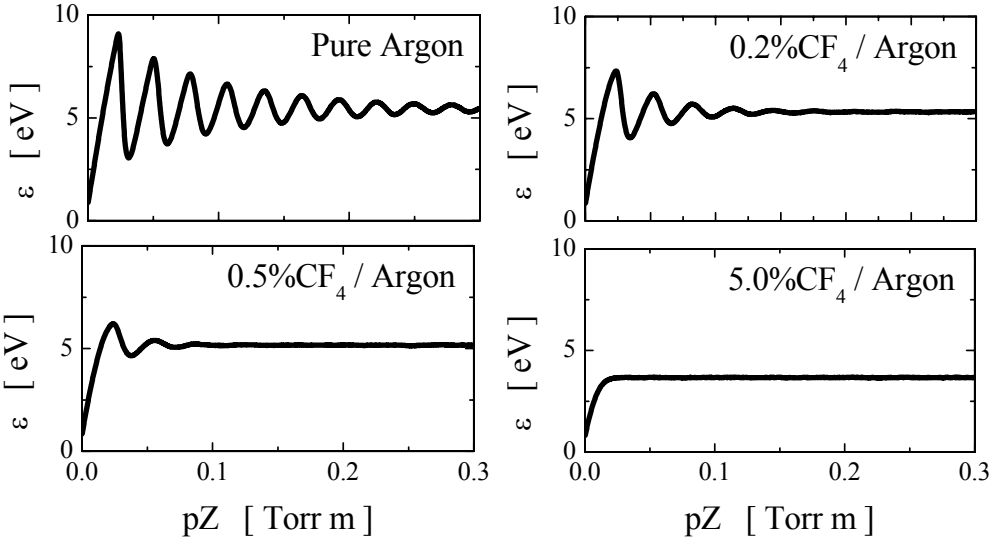

Figure 5. Spatial relaxation of the mean energy for pure argon and gas mixtures of argon and CF_4.

In Fig. 6 we show the effect of angle between electric and magnetic fields on spatial relaxation of the electrons for the Lucas-Saelee model. We observe that ε, v_z, v_x and v_y exhibit a damped oscillatory relaxation along a decaying profile for all ψ considered, although modulation is decreased with increasing ψ. We also note that v_x increases with the angle between the fields. This average velocity component has the worst statistic in our simulations. As ψ increases, the relaxation proceeds quicker while the period of oscillation becomes larger. As can be seen, the well known symmetry properties hold: $v_x = v_y = 0$ for $\varphi = 0^o$ and $v_y = 0$ for $\varphi = 90^o$. Another striking phenomenon is that v_y is comparable to v_x and v_z but yet it is common in the literature for plasma modellers to fail to include this quantity in their models. An extension of standard plasma models to include the **(E**×**B)**×**B** effects on electron transport may led to a better understanding of the power transfer to magnetically assisted/enhanced plasma reactors [24].

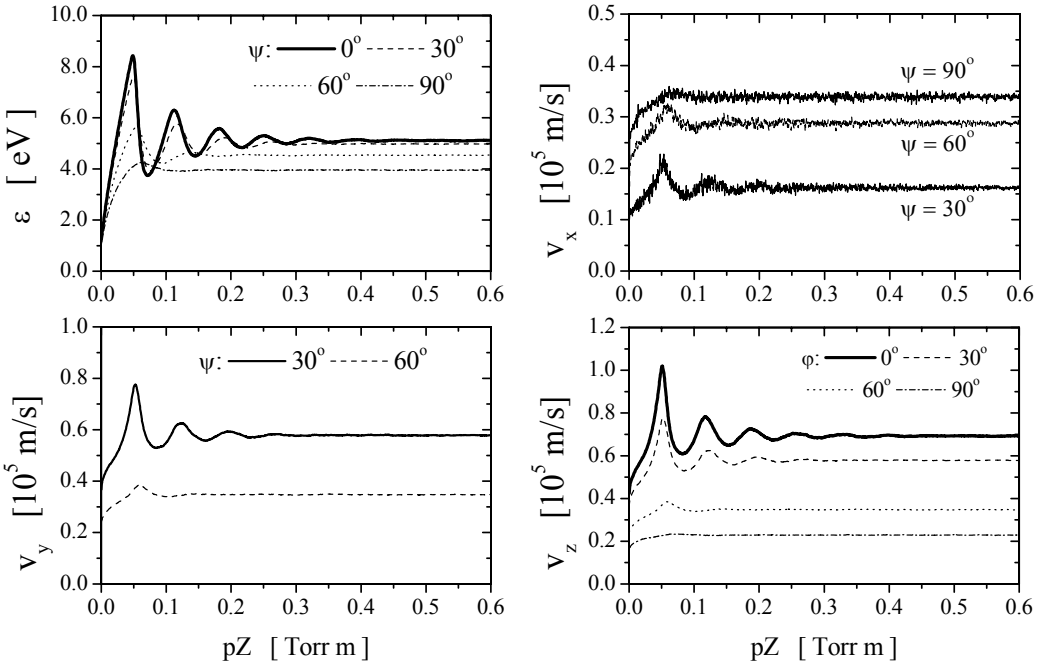

Figure 6. Spatial relaxation of the mean energy and average velocity components for Lucas-Saelee model at E/n_0 of 10 Td and B/n_0 of 200 Hx for various angles between the electric and magnetic fields as indicated at the graphs. The initial electron energy is 1.5 eV.

4. Concluding remarks

In this article we have presented a systematic study of the influence of magnetic fields and non-conservative collisional processes on the temporal and spatial relaxation properties of electron and positron swarms in gases. For the study of temporal relaxation under hydrodynamic conditions we have implemented (i) a time-dependent multi-term solution of Boltzmann's equation, and (ii) a time-resolved Monte-Carlo simulation. The results of the two independent techniques were compared for a benchmark gas and found to be in excellent agreement. In this study we focussed on (i) the explicit contribution of non-conservative processes to the bulk transport coefficients, and (ii) the influence of the angle between the electric and magnetic fields. The study yielded some interesting phenomena including the existence of transient negative diagonal elements of the diffusion tensor. In addition, we demonstrated that the traditional two-term approximation was invalid under certain conditions. As an interesting application of our theory, we also applied to study the relaxation time of positrons in molecular nitrogen. This is of particular relevance to understanding positron traps. For the study of

spatial relaxation under non-hydrodynamic conditions we implemented a spatially-resolved Monte-Carlo simulation. We demonstrated that we were able to suppress the traditional Franck_Hertz oscillations observed in pure Ar by the introduction of a small fraction of CF_4 into the system. In addition, for the first time, we have presented results that investigate the effect of varying the angle between the electric magnetic fields on the spatial evolution of the swarm. It was found that the nature of the profiles could be controlled by varying the angle: Frank-Hertz oscillations could be suppressed while oscillation periods could be modified.

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