Colloquium: Physically based fluid modeling of collisionally dominated low-temperature plasmas

R. E. Robson

Research School of Physical Sciences and Engineering, Australian National University, Canberra 2600, Australia

R. D. White

School of Mathematical and Physical Sciences, James Cook University, Townsville 4810, Australia

Z. Lj. Petrović

Department of Experimental Physics, Institute of Physics, 11080 Zemun, Serbia and Montenegro

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This colloquium examines the theoretical modeling of nonequilibrium low-temperature (tens of thousands of degrees) plasmas, which involves a juxtaposition of three distinct fields: atomic and molecular physics, for the input of scattering cross sections; statistical mechanics, for the kinetic modeling; and electromagnetic theory, for the simultaneous solution of Maxwell's equations. Cross sections come either from single-scattering beam experiments or, at very low energies (<0.5 eV), from multiple-scattering experiments on "swarms" in gases—the free diffusion or large Debye length limit of a plasma, where they are embedded in transport coefficient data. The same Boltzmann kinetic theory that has been developed to a high level of sophistication over the past 50 years, specifically for the purpose of unfolding these transport data, can be employed for low-temperature plasmas with appropriate modification to allow for self-consistent rather than externally prescribed fields. A full kinetic treatment of low-temperature plasmas is, however, a daunting task and remains at the developmental level. Fortunately, since the accuracy requirements for modeling plasmas are generally much less stringent than for swarms, such a sophisticated phase-space treatment is not always necessary or desirable, and a computationally more efficient but correspondingly less accurate macroscopic theoretical model in configuration space at the fluid level is often considered sufficient. There has been a proliferation of such fluid modeling in recent times and this approach is now routinely used in the design and development of a large variety of plasma technologies, ranging from plasma display panels to plasma etching reactors for microelectronic device fabrication. However, many of these models have been developed empirically with specific applications in mind, and rigor and sophistication vary accordingly. In this colloquium, starting from the governing Boltzmann kinetic equation, a unified, general formulation of fluid equations is given for both ions and electrons in gaseous media with transparent and internally consistent approximations, all benchmarked against established results. Thereby a fluid model is obtained that is appropriate for practical application but at the same time is based on a firmer physical foundation.

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I. INTRODUCTION

For many practical applications (including the treatment of organic materials, polymers, and integrated circuits), it is essential to have a nonequilibrium plasma with gas and ion temperature practically at room-temperature level (Lieberman and Lichtenberg, 1994, 2005). One may use the poor energy transfer in electron-molecule collisions to make sure that electrons gain high energy from the field and initiate plasma chemical processes, while the low degree of ionization means that their energy will not be transferred to ions and eventually to neutrals. Such plasmas, also known as *cold* or *low-temperature* plasmas, are typically far from equilibrium and can only be described by transport theory. In these low-temperature plasmas electron and ion-atom or molecule collision processes generally dominate the behavior of weakly ionized gases and act to suppress many (but not all) of the collective phenomena normally associated with high-temperature plasmas. For this reason low-temperature plasma physics has emerged as a distinct field, quite separate from fusion plasma physics, with an importance in its own right arising from its many high-technology industrial applications (Becker *et al.*, 2000).

The highly applied nature of the field has inevitably driven the modeling of these plasmas more towards empiricism, and unfortunately often away from its roots in fundamental physics. This schism further endangers progress in the field. The ever increasing demands of small-scale manufacturing processes can be met if and only if the basic physics is understood and basic physical principles are adhered to. These comments are particularly pertinent to the area of fluid modeling where typically empirical equations representing a balance of mass, momentum, and energy are constructed according to the specific application at hand and often driven by the demands of computational efficiency. Questions relating to the scientific rigor of this approach naturally arise, with benchmarking against established, accurate results, as well as tests of consistency between the various fluid models of major concern. As regards the latter, the radio-frequency discharge benchmark analysis of Surendra (1995) clearly demonstrates a high degree of variation between experiment and the various fluid models, and between the fluid models themselves. Given the disparate nature of the fluid models considered, this is hardly surprising. More importantly, the complexity of the benchmark system considered may well mask deficiencies in the basic physics of the models. As a general rule, we feel it would be better to start at the most fundamental level, getting the physics right for simple benchmarks and then proceeding to more complex situations. For plasmas, this means going right back to fluid descriptions in the swarm/test particle/free diffusion limit[1] and addressing questions of basic physics that remain unresolved even at this relatively simple level. A discussion of fluid modeling along these lines, emphasizing basic physics, is long overdue and is the task undertaken in the present colloquium.

Since collisions dominate in a weakly ionized gas, it is clear that there is an inseparable link with atomic and molecular physics, and that any modeling requires input, either directly or indirectly, of the cross sections (or interaction potentials) obtained through either one or more of the following means: (1) measurement results from single-scattering beam experiments (Brunger and Buckman, 2002), (2) results of theoretical *ab initio* calculations (Morrison and Trail, 1993; McEachran and Stauffer, 1997; Viehland, 2001), and/or (3) results from inversion of swarm experimental data (Crompton, 1994).

The first and most obvious point is that there must be correct implementation of the single-scattering collision data in the multiple-collision context of an extended gaseous medium, and it is here that some of the techniques of low-temperature plasma modeling employ a variety of treatments. This remark is not so much directed at modeling through Monte Carlo simulations or kinetic equations in which single-scattering cross sections enter into the calculations in a fairly clear way [see White *et al.* (2002) and remarks in Robson *et al.* (2003)], but rather at the macroscopic fluid level, where collisions are treated in a variety of ways which are not necessarily consistent with the data itself and/or the system under consideration. There are also other issues, associated with truncation of the set of fluid equations, that are indirectly associated with the correct representation of collisional phenomena. Thus at the fluid level of description one always has more unknowns than there are equations to solve, and therefore some closure hypothesis must be made. Surendra and Dalvie (1993) express dissatisfaction with the closure procedures reported in the plasma literature, and indeed it seems to us that this crucial step is too often dealt with in an uncritical, almost casual manner, especially as far as the heat flux *Ansatz* is concerned, in both the plasma and swarm literature. A critical discussion and new ideas, consistent with the correct

[1]For swarms the Debye length is much larger than the dimensions of the drift chamber, and the "free diffusion" or "test particle" situation prevails. On the other hand, a plasma is defined strictly as the state of matter where the Debye length is much smaller than any macroscopic length scale. In real gas discharges, these opposite extremes, plus a variety of intermediate conditions, may prevail at various positions in the discharge tube. The designation plasma tends to be used rather more loosely nowadays [see, e.g., the review of Winkler (2000)], possibly for this reason.

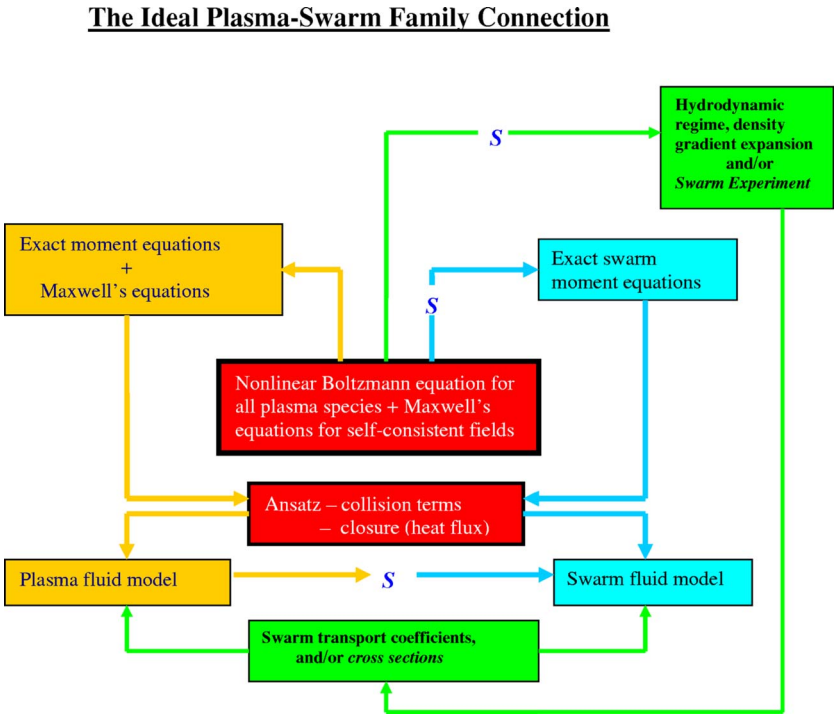

FIG. 1. (Color online) The steps involved in constructing fluid equations for both swarms and low-temperature plasmas, starting from the common Boltzmann equation for each of the ion and electron species, and proceeding through a common set of approximations for collision terms and closure. The symbol S denotes the swarm limit (low charge density, external fields, linear equations). Swarm fluid equations follow either directly as a limiting case of the plasma fluid equations or indirectly as moment equations of the swarm limit of Boltzmann's equation. In this picture, the plasma fluid equations are thus consistent with the numerous established results of swarm analysis (see Sec. V.D). In addition, it is legitimate in this scheme to employ measured or theoretically calculated swarm transport coefficients to replace approximate terms in either the swarm or plasma fluid equations, thereby considerably enhancing computational accuracy (see Sec. V.C).

portrayal of collision dynamics, is long overdue and is the main subject of this colloquium.

The second major point is that it is axiomatic in physics that any more general theory must produce the results of a well-established, though more restrictive theory in the appropriate limit. The classical limit of quantum mechanics as $h \to 0$ and the nonrelativistic limit of mechanics at small velocities are perhaps the best known examples. Likewise, any fluid model of a low-temperature plasma must produce, in the limit of low charge densities, the well-established results of the test particle or swarm limit [see the review of White *et al.* (2002)]. One thing that strikes the reader surveying the literature of fluid models of low-temperature plasmas is the plethora of apparently different fluid equations currently in use. This begs a number of questions: What is their relationship to each other? Which set gives the most accurate results? Which has the firmer physical foundation? Which is the more flexible? Which can be applied to both ions and electrons? We can answer some, but not all of these questions, by drawing upon the experience for swarms since there the approximations and assumptions associated with collisional transfer terms, in particular, have generally been more closely examined and benchmarked (Robson, 1986; Mason and McDaniel, 1988). In the hydrodynamic regime (Kumar *et al.*, 1980; Mason and McDaniel, 1988) many useful and well-known empirical laws have been tested and established on the basis of low-order moment equations of the Boltzmann equation, e.g., the Wannier energy relation, the generalized Einstein relations, Blanc's law, Tonks's theorem, and so on. Importantly, corrections to these laws and internal consistency and accuracy tests have been developed over the years (Skullerud, 1984; Robson, 1986, 1994; Mason and McDaniel, 1988). Plasma fluid equations should be able to produce these established swarm results in the appropriate limit, but it is by no means obvious that they do. For all these reasons, it appears to us to be highly desirable at this point in time to establish a set of fluid equations applicable to both swarms and plasmas.

Third, we suggest that establishment of the validity of any approximation, together with the integrity of any associated computational procedure, requires, in addition to comparison with experiment, an estimate of its intrinsic accuracy by either (1) a comparison with an exact benchmark result, (2) a series of successive approximations, or (3) some internally consistent and logical method.

The question of accuracy requires some further discussion in its own right. To begin with, the motivation for using a fluid approach needs to be established: this is simply that it provides a relatively inexpensive computational means of analyzing low-temperature plasmas (and

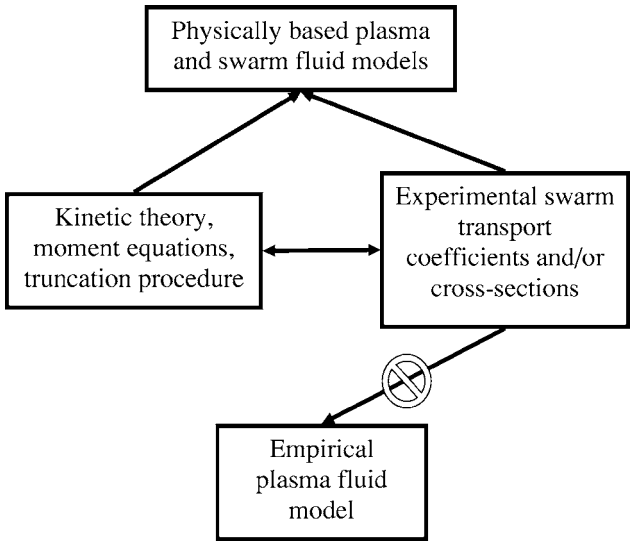

FIG. 2. The typical empirical fluid model of a low-temperature plasma suffers from the following defects: (i) any logical connection with exact moment equations derived from the Boltzmann equation is unclear; (ii) approximations for collision terms, and the closure hypothesis involving the heat flux, are not properly justified; and (iii) any commonality with swarm analysis and results is thus severed. Such an empirical model can therefore, in general, reproduce neither the swarm fluid equations nor the many well-known benchmark results and formulas established in the literature over many years (Sec. V.D). Furthermore, after having broken the nexus, the implementation of swarm transport coefficients in the empirical fluid equation model loses its physical foundation and can lead to unforeseen errors (see Sec. V.C).

charged-particle swarms) as compared with a full kinetic treatment, in which the Boltzmann equation is solved to high numerical precision (Winkler, 2000; White *et al.*, 2002). However, the penalty is a significant loss of accuracy, often no better than 10%, and sometimes much worse. For this reason results obtained from a fluid description of swarms (Robson, 1986; Robson, White, and Makabe 1997), for which errors in measured transport coefficients are typically 1% or less (Huxley and Crompton, 1974; Crompton, 1994), have hitherto been taken more as a qualitative to semiquantitative guide to physical understanding, rather than as a quantitative prescription. Plasma modelers, on the other hand, understandably take a more empirical and pragmatic standpoint, given that 10% is often quite an acceptable uncertainty even for quantitative purposes. There is thus an interesting contrast between accuracy requirements in the two fields and the manner in which the fluid description is viewed. Unfortunately, the literature has also become divided along these lines, even though results established in one area should carry over to the other, in appropriate limits. The present colloquium emphasizes this commonality and aims at strengthening the connection between the two fields.

Our view of the ideal way to proceed in fluid modeling is summarized schematically in Fig. 1, which emphasizes both physical foundations and internal consistency. The main limitations inherent in the more traditional empirical approaches are highlighted in Fig. 2.

Before embarking on the present overview specifically devoted to the modeling of charged fluids, we would like to emphasize that many of our observations concerning the approximations involved apply equally well to other areas of physics, whether the fluid is charged or neutral, and whether or not there are any driving fields. This remark applies particularly to the calculation of collision terms, which in this colloquium follow the prescription of momentum-transfer theory, a far more accurate, but far less well-known approximation than the popular mean-free-path theories of elementary kinetic theory [see p. 144 of Mason and McDaniel (1988)]. Just as numerous texts illustrate the application of the mean-free-path analysis to a whole spectrum of transport properties in a wide variety of physical problems, so can the discussion presented here be carried over from gaseous electronics to the more general arena. Hot atom chemistry and cold muon catalyzed fusion are just two pertinent examples (Robson, 1986, 1988), while recent research into ion clouds in ion traps deserves special mention (Viehland, 2005).

The outline of this colloquium is as follows. In Sec. II we review a simple, well-known example from plasma physics to illustrate the importance of the formulation of a plasma fluid model consistent with swarms, while in Sec. III general fluid equations are formulated for both ions and electrons, without any limiting assumptions, starting from the Boltzmann equation. Approximation of the collision terms is then discussed, along with the question of truncation, and in particular we focus upon the role of the heat flux vector. We then proceed from the general equations to the hydrodynamic limit, highlighting the transport coefficients, and then to the general, nonhydrodynamic problem. A benchmark solution is found for electrons in the free diffusion limit for plane-parallel geometry in Sec. IV, and the heat flux *Ansatz* is the subject of particular scrutiny. Finally, in Sec. V we address the use of swarm transport data in fluid models and make recommendations for the correct application in them.

II. THE IMPORTANCE OF PLASMA-SWARM CONSISTENCY

A. The textbook model of ambipolar diffusion

For both swarms and plasmas in the hydrodynamic, small gradient limit, the fluid equations derived below (see Sec. V.B) yield Fick's law of diffusion for each of the charged species:

$$\mathbf{\Gamma}^{(\pm)} = n^{(\pm)} K^{(\pm)} \mathbf{E} - D^{(\pm)} \nabla n^{(\pm)}. \tag{1}$$

For swarms of very low charge density, in which the Debye length (distance over which the field of a charged particle is effectively screened by surrounding charged particles) is larger than any relevant macroscopic dimension, the electric field $\mathbf{E}$ is externally prescribed, and the ion and electron density distributions $n^{(\pm)}$ evolve inde-

pendently of each other. For plasmas (high charge density, the Debye length is much much greater than the macroscopic dimensions), on the other hand, the ions and electrons interact with each other to produce a self-consistent space-charge field **E** and, as the usual textbook argument goes [see, e.g., Chen (1974)], the ions and electrons must move together, i.e.,

$$\mathbf{\Gamma}^{(+)} = \mathbf{\Gamma}^{(-)} \equiv \mathbf{\Gamma} \tag{2}$$

in order to maintain overall quasielectrical neutrality, $n^{(+)} \approx n^{(-)} \equiv n$. It is important to note that in both cases the same form of fluid equations (1) are used as the common starting point, and that the swarm mobility and diffusion coefficients for ions and electrons $K^{(\pm)}$ and $D^{(\pm)}$, respectively, appear in both cases. Of course, what one does with the fluid equations subsequently is a different matter. For swarms, one substitutes Eq. (1) into the respective equation of continuity for each species to generate two distinct diffusion equations, which may be solved independently of each other. For plasmas, one substitutes Eq. (1) into Eq. (2) to generate an expression for the space-charge field **E**, which in turn yields for the common flux

$$\mathbf{\Gamma} = -D_a \nabla n, \tag{3}$$

where

$$D_a = \frac{K^{(+)}D^{(-)} - K^{(-)}D^{(+)}}{K^{(+)} - K^{(-)}} \tag{4}$$

is the ambipolar diffusion coefficient. Equation (3) when substituted into the equation of continuity for either charge species then yields the ambipolar diffusion equation, which describes the common evolution of both ions and electrons.

B. Plasma-swarm consistency

The foregoing is a straightforward illustration, implicitly accepted throughout the plasma community, of how analysis of both swarms and plasmas proceeds from a common set of fluid equations (1), which in turn stem from the same approximations. Our suggestion is that the same philosophy should be adhered to in all situations, with a common general set of fluid equations for all circumstances, according to Fig. 1. One can make subsequent approximations for the plasma as needed, just as in the simple ambipolar diffusion problem above, but the common origin remains clear, as does the justification for using swarm transport data in the ultimate expression (4). It would be unthinkable to suggest that there should be two starting points instead of the one common point of departure (1), i.e., one set of fluid equations for swarms, and another, different set for plasmas, and yet that is precisely the sort of thing that is happening in current empirical plasma fluid modeling. The nature of the problem is illustrated schematically in Fig. 2: without a common foundation, inconsistencies ranging from the conceptual to the practical are inevitable. It would be incorrect, for example, to use swarm transport coefficients in a plasma fluid model which is itself inconsistent with basic swarm theory, and yet that is what is currently happening. [N.B. Given its consistency with Fig. 1, Eq. (4) does not suffer from this defect, and swarm data can be legitimately substituted.]

We shall return to this point again in the course of this colloquium.

III. FORMULATION OF A COMMON SET OF FLUID EQUATIONS

A. Kinetic equation

The starting point of any transport calculation is the kinetic equation, which can be written in generic form as

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\cdot\nabla + \frac{q}{m}[\mathbf{E} + \mathbf{v}\times\mathbf{B}]\cdot\frac{\partial}{\partial \mathbf{v}}\right) f = -J(f,F) - J(f,f), \tag{5}$$

where $f(\mathbf{r},\mathbf{v},t)$ is the charged-particle phase-space distribution function, $J(f,F)$ is the charged-particle–neutral molecule collision term, and F is the prescribed distribution function of the neutral gas (usually taken to be Maxwellian at temperature T_g). The definition of a swarm is that the degree of ionization should be negligible and that there should be no interactions with any products of collisions, excited, or charged particles; the Debye radius for a swarm experiment is infinite (or much larger than the swarm experiment). These conditions are satisfied very well in low current drift tube experiments. Usually, only one species of particle of mass m and charge q is present in the gas (or if more than one species is present, each may be treated separately), **E** and **B** are prescribed external fields, and the electron-electron or ion-ion interaction term $J(f,f) \approx 0$. On the other hand, for plasmas an equation like Eq. (5) exists for each charged species, **E** and **B** are determined self-consistently from Maxwell's equations (and external boundary potentials and currents); electron-excited state, excited state–excited state, electron-electron, ion-ion, and electron-ion interactions, as characterized by terms in $J(f,f)$, may be also important.

On the right-hand side of Eq. (5), the traditional collision operator of Boltzmann (1872), or its differential form in the case of electrons (Davydov, 1935), should be used for describing elastic collisions. To describe inelastic collisions, a fully classical collision operator can sometimes be justified for ions (Viehland, 1994), but for lighter particles, the Wang-Chang *et al.* (1964) semiclassical collision operator (in which neutral molecules in different excited states are effectively treated as different species) or its approximation in a finite difference form (Frost and Phelps, 1962) seems mandatory. The Waldmann-Snider quantum operator (Waldmann, 1958; Snider, 1960) may be required in some situations when the degeneracy of rotational states is important. Further interaction terms representing ionization, attachment, ion-molecule reactions, etc., could be added if desired (White *et al.*, 2002; Winkler *et al.*, 2002). Note that both

ion and electron components can be treated starting from the same form of Eq. (5), which can be solved using the same methods and techniques.

The task of kinetic theory is to solve Eq. (5) for f and hence to obtain quantities of physical interest such as velocity moments. In the fluid equation approach, one aims to obtain these moments directly, without knowing f. The next subsection addresses this question.

B. Fluid equations, truncation, and approximations to the collision terms

1. The general problem of truncation

Fluid equations may be derived either directly as moments of Eq. (5) or from first principles using physical arguments. For the present, we find it convenient to adopt the former approach. Basically, whether dealing with swarms or plasmas, fluid equations are generated by multiplying Eq. (5) by the set of functions $\phi_i(\mathbf{v})$ and integrating over all velocities to give equations for the averages $\langle\phi_i(\mathbf{v})\rangle$, where $\phi_1(\mathbf{v})=1$, $\phi_2(\mathbf{v})=mv$, $\phi_3(\mathbf{v})=\frac{1}{2}mv^2$, $\phi_4(\mathbf{v})=m\mathbf{v}\mathbf{v}$, and so on. Generally speaking, the equations are not closed—there are always more unknowns than equations to solve for. As Hazeltine and Waelbroeck (1998) put it, "The derivation of a closed set of fluid equations … generally requires serious approximation" to fix this problem. They go on to distinguish two possible types of closure mechanisms:

(a) *Truncation*, in which a high-order moment is assumed either to vanish or to be expressible somehow in terms of lower-order moments, using some empirical criterion. In more sophisticated kinetic-theory analysis [see, e.g., Mason and McDaniel (1988)], one increments the size of the moment equations systematically and closes the set when some convergence criterion is reached, thereby maintaining tight control. In contrast, at the fluid level such closure is achieved by a straight-out postulate or *Ansatz* and is uncontrolled.

(b) *Asymptotic closure*, which is based upon the smallness of some parameter. It is possible to control and refine such closure by successive approximations.

Both mechanisms are employed in what follows.

2. Approximation of the collisional transfer terms—general remarks

For simplicity, and to focus upon the essential points, we assume a swarm or plasma for which the degree of ionization is so low that charge-charge interactions, as represented by the term $J(f,f)$, are negligible, and focus entirely upon the charge-neutral term $J(f,F)$ in Eq. (5). This approximation can subsequently be relaxed and the theory refined accordingly without any great difficulty.[2] The collisional transfer terms generally contain an infinite number of moments, and must therefore be approximated by some closure hypothesis. We can express the collisional rate of change of property $\phi_i(\mathbf{v})$ and its approximation formally as

$$\int \phi_i(\mathbf{v})J(f,F)d^3\mathbf{v} = n\sum_{j=1}^{\infty} J_{ij}\langle\phi_j(\mathbf{v})\rangle \approx n\sum_{j=1}^{i} J_{ij}\langle\phi_j(\mathbf{v})\rangle, \quad (6)$$

where a velocity average is defined as

$$\langle\cdots\rangle = \frac{1}{n}\int \cdots f(\mathbf{v})d\mathbf{v}.$$

A closure hypothesis amounts to some particular way of specifying the collision rates J_{ij} (which are matrix elements of the collision operator with respect to the basis function set). Methods reported in the literature essentially follow the philosophy expressed in Sec. III.A, and fall into one of the following two categories described above.

(a) Truncation by empirical means (i.e., a guess!). For example, with $\phi_1(\mathbf{v})=m\mathbf{v}$, a favored approximation is to set $J_{1j}=\nu_c\delta_{j1}$, where ν_c is some collision frequency, so that the right-hand side of Eq. (6) is equal to $nm\nu_c\langle\mathbf{v}\rangle$. Alternatively, the velocity distribution function is often assumed to be a Maxwellian (another guess!), a method which also falls into the same category of uncontrolled approximation.

(b) Asymptotic, controlled closure via either (i) assumption of a specific form for the velocity distribution function, with systematically controlled refinements (Mason and McDaniel, 1988); or (ii) momentum-transfer theory with systematic approximations to the J_{ij}, benchmarked by exactly solvable models and internal error estimates. The small parameter in this case is ν_m', the energy derivative of the momentum-transfer collision frequency. Full details can be found in Robson (1986).

Whatever the method chosen, the collision terms (6) should be of exactly the same mathematical form for both swarms and plasmas: collisions are not influenced by either fields or the spatiotemporal behavior of the plasma. The latter effects are contained in the streaming terms, i.e., the left-hand side of the moment equations of Eq. (5), and it is here that the differences between

[2]Note that at the level of momentum and energy fluid equations, the mutual ion-ion or electron-electron interaction term $J(f,f)$ makes no contribution in any case since

$$\int d\mathbf{v}J(f,f)\phi_i(\mathbf{v}) \equiv 0$$

for $\phi_i=1, mv, \frac{1}{2}mv^2$, by virtue of conservation of particle number, momentum, and energy. On the other hand, electron-ion collisions do in fact contribute further terms to the right-hand side of the balance equations.

plasma and swarm analysis will be most obvious. We shall return to the specifics of evaluating the collision transfer rates in Sec. V.C.

In what follows, as a simplification to the notation, we dispense with the brackets $\langle\ \rangle$ representing averages when no ambiguity will arise, and simply write $\mathbf{v}$ for $\langle\mathbf{v}\rangle$, for example.

3. Fluid equations with momentum-transfer collision approximation

Momentum-transfer theory goes back to ideas expressed by Wannier (1953), in which the mathematical form of expressions derived for the constant collision frequency model are assumed to carry over to the general, energy-dependent collision frequency case (Robson, 1986; Mason and McDaniel, 1988). It is important to realize that since collisions *per se* are independent of the macroscopic characteristics of the system—fields, gradients, boundaries, etc.—the accuracy of the approximation is the same for both swarms and plasmas. Note also that the Wannier model is really just the lowest-order approximation in a scheme of successive approximations to the collision terms, with corresponding procedures for refinement and internal accuracy estimates being readily available (Robson, 1986; Robson and Ness, 1988). At the lowest level of approximation, this results in the following set of equations, equally valid for both swarms and plasmas, representing continuity, momentum, and energy balance, respectively, for particles of mass m and charge q,

$$\partial_t n + \boldsymbol{\nabla}\cdot n\mathbf{v} = n(\nu_I - \nu_a), \tag{7}$$

$$nm\frac{d\mathbf{v}}{dt} + \boldsymbol{\nabla}\cdot\mathsf{P} - nq(\mathbf{E}+\mathbf{v}\times\mathbf{B}) = -n\frac{mM}{m+M}\nu_m(\epsilon_{\mathrm{CM}})\mathbf{v} - nm\mathbf{v}(\nu_I + \xi\nu_a'), \tag{8}$$

$$n\frac{d}{dt}(\epsilon - \tfrac{1}{2}mv^2) + \boldsymbol{\nabla}\cdot\mathbf{J}_q + \mathsf{P}:\nabla\mathbf{v} = -n\nu_e(\epsilon_{\mathrm{CM}})[\epsilon - \tfrac{3}{2}k_BT_g - \tfrac{1}{2}(m+M)v^2 + \Omega(\epsilon_{\mathrm{CM}})] - n\epsilon(\nu_I + \xi\nu_a') - n\tfrac{1}{2}mv^2(2\nu_I + \xi\nu_a'), \tag{9}$$

where n, $\mathbf{v}$, and ϵ denote number density, average velocity, and average energy of the charged particles, ∇ is the normal gradient operator, while M and T_g refer to the molecular mass and temperature of the neutral gas, respectively. The pressure tensor P is defined as

$$\mathsf{P} = nm\langle\mathbf{VV}\rangle \equiv nk_B\mathsf{T} \tag{10}$$

and the heat flux vector as

$$\mathbf{J}_q \equiv \tfrac{1}{2}n\langle mV^2\mathbf{V}\rangle, \tag{11}$$

where $\mathbf{V}=\mathbf{v}-\langle\mathbf{v}\rangle$, while

$$\frac{d}{dt} = \partial_t + \mathbf{v}\cdot\boldsymbol{\nabla}$$

denotes the convective time derivative. The average collision frequencies for momentum and energy transfer,

$$\nu_m(\epsilon_{\mathrm{CM}}) = N\sqrt{\frac{2\epsilon}{\frac{mM}{m+M}}}\sigma_m(\epsilon_{\mathrm{CM}}), \tag{12}$$

$$\nu_e(\epsilon_{\mathrm{CM}}) = \frac{2mM}{(m+M)^2}\nu_m(\epsilon_{\mathrm{CM}}), \tag{13}$$

like the momentum-transfer cross section $\sigma_m(\epsilon_{\mathrm{CM}})$ from which they derive, are prescribed functions of the mean energy in the center of mass,

$$\epsilon_{\mathrm{CM}} = \frac{M\epsilon + m\frac{3}{2}k_BT_g}{m+M},$$

where k_B is Boltzmann's constant. The same goes for the average ionization and attachment collision frequencies $\nu_I(\epsilon_{\mathrm{CM}})$ and $\nu_a(\epsilon_{\mathrm{CM}})$, respectively. Note that the latter is really a total ionization frequency summed over all possible channels i,

$$\nu_I = \sum_i \nu_I^{(i)},$$

in which ions are produced in excited states characterized by energies $\epsilon_I^{(i)}$. The term Ω represents the average energy lost in one collisional energy relaxation time ν_e^{-1} through nonelastic processes and is given by

$$\Omega = \frac{M}{m+M}\sum_\alpha \epsilon_\alpha\frac{\vec{\nu}_\alpha - \overleftarrow{\nu}_\alpha}{\nu_e(\epsilon_{\mathrm{CM}})} - \sum_i \Delta\epsilon_I^{(i)}\frac{\nu_I^{(i)}}{\nu_e(\epsilon_{\mathrm{CM}})}. \tag{14}$$

The inelastic channels α are governed by threshold energies ϵ_α and collision frequencies for inelastic and superelastic processes $\vec{\nu}_\alpha$ and $\overleftarrow{\nu}_\alpha$, respectively. The latter are also prescribed functions of ϵ_{CM} but need to be specified more carefully in terms of the corresponding cross sections, $\sigma_I(\epsilon_{\mathrm{CM}})$.[3]

[3]It appears that the best way to take into account the collisions with excited molecules in plasmas is by treating them as a separate gas in the mixture with the buffer gas. This is so because such particles will have considerably different cross sections for both elastic and inelastic collisions and, in particular, the thresholds are different, which is essential for maintenance of some plasmas such as the H mode of inductively coupled plasmas. In that case the energy gain in superelastic collisions requires coupling to an additional equation for the kinetics of excited species. This is solved trivially in the case of swarms because the basic assumption in that case is that the gas is not perturbed by charged particles so only thermal population of excited species requires consideration. In plasmas, however, nonequilibrium kinetics of charged particles may lead to nonequilibrium population of excited species. With that in mind the application of momentum-transfer or other equations to plasmas needs no further consideration.

Note that to first order attachment enters the momentum and energy balance equations in terms of the derivative of the attachment collision frequency,

$$\nu_a' \equiv \frac{d\nu_a}{d\epsilon},$$

whereas to the same order for ionization only the ionization collision frequency ν_I appears. This reflects the different ways in which attachment and ionization affect average charged-particle properties, both qualitatively and quantitatively, as has been discussed elsewhere (Robson, 1986; Robson and Ness, 1988). Higher-order momentum-transfer-theory corrections could be added to the right-hand side if desired, without in anyway altering the generality of the arguments presented below.

For swarms, **E** and **B** are externally specified, spatially uniform fields, but for plasmas, they are generally space and time dependent, and must be obtained self-consistently with Maxwell's equations. For example, for capacitively coupled plasmas (Lieberman and Lichtenberg, 1994), this means solving the fluid equations in conjunction with Poisson's equation,

$$\boldsymbol{\nabla} \cdot \mathbf{E} = \sum_i n_i q_i / \epsilon_0, \tag{15}$$

where the sum on the right-hand side extends over all charged species i.

We emphasize that approximations have been made only on the right-hand sides of Eqs. (8) and (9), and that the left-hand side remains exact. There is, however, further approximation to be made before they are in a useful form.

C. The streaming terms

In order to proceed further, we must specify the pressure tensor P (or equivalently the temperature tensor T) appearing on the left-hand side (the *streaming* terms) of the balance equations (8) and (9). The former generally presents less of a problem, especially for the case of light, charged particles for which the pressure tensor becomes a scalar proportional to the mean energy [see Eq. (16)]. However, with one crucial exception (see Sec. IV.A), the latter always leads to a closure problem. If one formulates a balance equation for $\mathbf{J}_q$ by taking the appropriate velocity moment of Eq. (5), a term $\langle v^4 \rangle$ generally results, which in turn is given by a higher-order equation containing yet higher-order moments and so on. This chain of equations must be broken by some physically based *Ansatz* in order that a solution can be effected, which falls into the category of an uncontrolled truncation.

The significance of streaming terms, and consequently the severity of the closure problem for the modeling of any particular experimental arrangement, depends upon the way in which spatial variations can be treated. In many cases of interest it is simply not possible to describe the plasma or swarm even qualitatively at the hydrodynamic level, and the full set of nonhydrodynamic fluid equations must be employed. Moreover, $\mathbf{J}_q$ plays a central role under these circumstances (Surendra and Dalvie, 1993) and must be expressed in terms of the lower-order moments n, $\langle \mathbf{v} \rangle$, and $\langle \epsilon \rangle$ in order to close the equations, as has long been recognized (Golant *et al.*, 1980). Nevertheless, as Surendra and Dalvie (1993) point out, it is quite common in plasma modeling for $\mathbf{J}_q$ to be set either implicitly or explicitly to zero [e.g., by assuming a drifted Maxwellian velocity distribution function (Ingold, 1989)] or to be represented by a Fourier type of expression that relates it to $\nabla\varepsilon$.

On the whole, $\mathbf{J}_q$ is dealt with in a rather *ad hoc* fashion in the literature, with no standardization or benchmarking and little or no physical or mathematical justification of the *Ansatz*. A notable exception to this is the work of Suchy and Altman (2003), who present a general, quantitative argument as to why $\mathbf{J}_q$ might be expected to be small under near-equilibrium conditions. In general, however, it appears that heat flux plays a major role in determining plasma behavior (Winkler, 2000), and it seems that solutions to the fluid equations are generally quite sensitive to the choice of this *Ansatz*, both qualitatively and quantitatively, as shown below.

All in all, the present situation regarding $\mathbf{J}_q$ is very unsatisfactory, and we believe that a much more thorough and critical analysis is required.

IV. HEAT FLUX *Ansatz* AND BENCHMARKING

From this point on we consider for simplicity only particle-conserving collisions, but it is emphasized that ionization and attachment could be included as in Eqs. (8) and (9). Our view is that it is necessary to get the simpler problem correct first before moving on to the more complicated one.

A. Light particles, heat flux *Ansatz*

We now focus on light particles, such as electrons or muons, for which $m/m_0 \ll 1$, $\mu \approx m$, $\epsilon_{\mathrm{CM}} \approx \epsilon \gg \frac{1}{2}mv^2$ and the elements of the pressure tensor are given by

$$P_{ij} \approx \frac{2}{3} n \epsilon \delta_{ij}. \tag{16}$$

Equations (8) and (9) then become, in the absence of ionization or attachment,

$$nm\frac{d\mathbf{v}}{dt} + \frac{2}{3}\boldsymbol{\nabla}(n\epsilon) - nq(\mathbf{E} + \mathbf{v} \times \mathbf{B}) = -nm\nu_m(\epsilon)\mathbf{v} \tag{17}$$

and

$$n\frac{d\epsilon}{dt} + \boldsymbol{\nabla} \cdot \mathbf{J}_q + \frac{2}{3} n\epsilon \boldsymbol{\nabla} \cdot \mathbf{v} = -n\nu_e(\epsilon)\left[\epsilon - \frac{3}{2}k_B T_g - \frac{1}{2}m_0 v^2 + \Omega(\epsilon)\right], \tag{18}$$

where

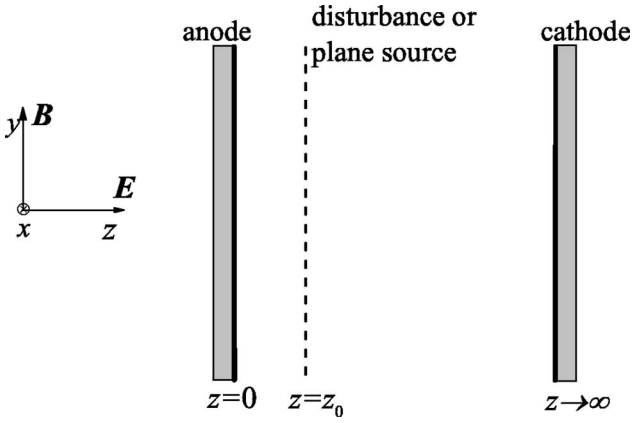

FIG. 3. An idealized model of the steady-state Townsend experiment, in which electrons emitted at a constant rate from an infinite plane source drift and diffuse in a uniform gas under the influence of uniform external fields.

$$\Omega = \sum_{\alpha} \epsilon_{\alpha}(\vec{\nu}_{\alpha} - \overleftarrow{\nu}_{\alpha})/\nu_e(\epsilon) \tag{19}$$

and $\nu_e(\epsilon) \approx (2m/m_0)\nu_m(\epsilon)$. Equations (7), (17), and (18) comprise three equations in four unknowns, n, $\mathbf{v}$, ϵ, and $\mathbf{J}_q$, and the latter must be prescribed through an additional *Ansatz*, as explained above.

We now turn to the heat flux term which appears in the energy balance equation. For the special case of elastic collisions that are only described by a momentum-transfer collision frequency $\nu_m(\epsilon) \sim \epsilon$, the following exact expression can be derived by going to the next highest moment of the Boltzmann equation:

$$\mathbf{J}_q \approx -\frac{2}{3m}\nabla\left[\frac{n\xi(\epsilon)}{\nu_m(\epsilon)}\right] + \frac{1}{3}(5-2p)\frac{nq}{m\nu_m(\epsilon)} \times(\mathbf{E}+\mathbf{v}\times\mathbf{B}) - \frac{5}{3}n\epsilon\mathbf{v}, \tag{20}$$

where $\xi(\epsilon) = \epsilon^2$ and $p = 1$. We now postulate that Eq. (20) applies in the more general case, involving both elastic and inelastic collisions described by collision frequencies with arbitrary energy dependences, where

$$p = \frac{d\ln\nu_m}{d\ln\epsilon} \tag{21}$$

and $\xi(\epsilon)$ is proportional to the average of the square of the energy. As with momentum-transfer theory, we have at least anchored this *Ansatz* to a collision model for which we know the expression to be exact. However, unlike momentum-transfer theory, it is not obvious how to refine Eq. (20) by successive approximations. These considerations aside, the best way to establish the validity of Eq. (20) for practical purposes is to perform a benchmark calculation.

B. The benchmark model

As an application of the theoretical framework presented in the previous section, consider the simple infinite plane-parallel geometry configuration shown in Fig. 3; electrons diffuse and drift away from the plane source into an infinite gas at the same rate as they are emitted from the source. There exists a steady state, in which properties are independent of time and vary with position only in the z direction. The density of these electrons is assumed to be so low that they can be treated in the swarm (free diffusion) limit. This is an idealization of the steady-state Townsend experiment, though without any accompanying ionization. Note that this is an inherently nonhydrodynamic situation; that is, spatial variations in physical quantities cannot be accounted for by a functional dependence upon density and its gradients. Unphysical results occur, for example, if the diffusion equation is applied to this problem.[4]

In what follows, we seek analytic solutions in the asymptotic region far downstream from the source. Here gradients are small, and deviations from the spatially uniform state may be treated as small perturbations, facilitating linearization of the equations. A priority of the analysis is the benchmarking of the fluid equations via direct comparison of the solution obtained by applying various heat flux *Ansatz*, i.e., Eq. (20) and others already in the literature, with an exact solution of Boltzmann's equation for a simple collision-model case. Note that although we employ models for the purpose of carrying out the present calculations, our fluid equations are quite general and are suitable for solving problems in real gases. In all cases we take $B=0$ for simplicity, although it is emphasized that the solution for a nonzero magnetic field is a very straightforward extension of the analysis presented. The electric field is taken to be homogeneous for the purposes of analytic solution, but this restriction can also be lifted when necessary.

1. Analytic solution for the asymptotic region

We now set $q=-e$, $\mathbf{E}=(0,0,-E)$, $\mathbf{v}=(0,0,v)$, $\mathbf{J}_q=(0,0,J_q)$, $\nabla=(0,0,\partial/\partial z)$, and all derivatives with respect to time equal to zero in Eqs. (7), (17), (18), and (20) to get the fluid equations corresponding to Fig. 3:

$$\frac{\partial\Gamma}{\partial z} = 0, \tag{22}$$

$$\frac{2}{3}\frac{\partial(n\varepsilon)}{\partial z} = neE - nm\nu_m(\varepsilon)v, \tag{23}$$

[4]The diffusion equation

$$\frac{\partial n}{\partial t} + v_d\frac{\partial n}{\partial z} - D\frac{\partial^2 n}{\partial z^2} = 0$$

is valid strictly speaking only if both hydrodynamic and weak-gradient conditions prevail. Even in the asymptotic region at large distances downstream in Fig. 3, where gradients are weak, hydrodynamic conditions do not prevail, and therefore the diffusion equation is invalid.

$$-\frac{1}{\nu_e}\left[v\frac{\partial\varepsilon}{\partial z}+\frac{2\varepsilon}{3}\frac{\partial v}{\partial z}+\frac{1}{n}\frac{\partial J_q}{\partial z}\right]=\varepsilon-\frac{3}{2}kT_g-\frac{1}{2}Mv^2+\Omega(\varepsilon), \tag{24}$$

and

$$J_q=-\frac{2}{3m}\frac{\partial}{\partial z}\left[\frac{n\xi(\varepsilon)}{\nu_m(\varepsilon)}\right]+\frac{(5-2p)}{3}\frac{na\varepsilon}{\nu_m(\varepsilon)}-\frac{5}{3}\Gamma\varepsilon. \tag{25}$$

Equation (22) shows that the particle flux does not vary with position, i.e.,

$$\Gamma=nv=\text{const}. \tag{26}$$

In general, the equations shown above constitute a set of coupled nonlinear equations that must be solved numerically. Up to this point, the electric field may be considered to be space dependent, but in what follows it is assumed to be constant. By focusing on the asymptotic region, far downstream from the source, which for simplicity we take at the origin, i.e., $z=z_0=0$, approximate analytic solutions are possible. Thus we write

$$n(z)=n_0+n_1e^{Kz} \tag{27}$$

and similarly for all other quantities, where the subscript 0 denotes the spatially uniform regime attained as $z\to\infty$, the subscript 1 denotes a small perturbation, and K a wave number. The latter, which may in general be complex, controls the rate of decay to the uniform state and is the quantity of primary interest. Equation (27) and others of similar form for v, ε, and J_q are substituted into Eqs. (23)–(25), and the resulting equations are linearized in small quantities. Upon equating the coefficients of e^{Kz}, we obtain the following hierarchy of equations.

(i) *Spatially homogeneous equations*: To zero order in e^{Kz} we find equations corresponding to the limit $z\to\infty$:

$$eE=m\nu_m(\varepsilon_0)v_0,$$

$$\varepsilon_0=\frac{3}{2}kT_g+\frac{1}{2}Mv_0^2-\Omega(\varepsilon_0), \tag{28}$$

$$J_{q0}=-\frac{2}{3}p\Gamma\varepsilon_0.$$

(ii) *First-order spatially inhomogeneous equations*: To first order, after linearizing and equating coefficients of e^{Kz}, there follows

$$\frac{n_1}{n_0}+\frac{v_1}{v_0}=0,$$

$$(\kappa+p)\frac{\varepsilon_1}{\varepsilon_0}+(1-\kappa)\frac{v_1}{v_0}=0, \tag{29}$$

$$\frac{3}{2}\kappa\left[\frac{\varepsilon_1}{\varepsilon_0}+\frac{2}{3}\frac{v_1}{v_0}+\frac{J_{q1}}{\Gamma\varepsilon_0}\right]+\gamma\frac{\varepsilon_1}{\varepsilon_0}-2\frac{v_1}{v_0}=0,$$

where κ is a dimensionless wave number defined by

$$K=\frac{3}{2}\frac{eE}{\varepsilon_0}\kappa \tag{30}$$

and

$$\gamma=\frac{(1+\Omega')\varepsilon_0}{\varepsilon_0+\Omega-\frac{3}{2}kT_g}. \tag{31}$$

The linearized version of the heat flux *Ansatz* (25) is

$$\frac{J_{q1}}{\Gamma\varepsilon_0}=Q(\kappa)\frac{\varepsilon_1}{\varepsilon_0}, \tag{32}$$

where the dimensionless spectral heat flux is defined by

$$Q(\kappa)=\frac{A+B\kappa+C\kappa^2}{\kappa-1} \tag{33}$$

while

$$A=\frac{2p}{3},$$

$$B=\alpha\bar{p}-\frac{5}{3}\left(1+p-\frac{2p^2}{5}\right), \tag{34}$$

$$C=-\alpha(\bar{p}-p-1),$$

and

$$\bar{p}=\frac{\varepsilon_0\xi'(\varepsilon_0)}{\xi(\varepsilon_0)},$$

$$\alpha=\frac{\xi(\varepsilon_0)}{\varepsilon_0^2}. \tag{35}$$

Equations (29) and (32) together constitute four homogeneous equations in the four unknowns, $n_1,v_1,\varepsilon_1,q_1$, and therefore have a nontrivial solution if and only if the determinant of the coefficients vanishes. This leads to the secular equation

$$\frac{5}{2}\kappa^2+\left(\gamma+p-\frac{7}{2}\right)\kappa-(\gamma+2p)+\frac{3}{2}\kappa(\kappa-1)Q(\kappa)=0. \tag{36}$$

We seek solutions of Eq. (36) for which

$$\text{Re}(\kappa)<0, \tag{37}$$

which correspond to physical quantities decaying to a spatially uniform equilibrium state.

Before proceeding further, a comment of a general nature needs to be made. Both the qualitative and quantitative nature of the solutions of Eq. (36) depend sensitively upon the heat flux, i.e., upon $Q(\kappa)$. It is no good simply ignoring it, for if we take $Q(\kappa)=0$, Eq. (36) admits only real solutions, regardless of the strength of the inelastic processes (as measured by γ) or the value of p. Thus none of the oscillatory patterns of decay known to characterize this problem [e.g., in the Franck-Hertz experiment (Robson *et al.*, 2000; Sigeneger *et al.*, 2003)] can be reproduced with such a drastic assumption. Like-

wise, neglecting heat flux in even the simple case of elastic collisions governed by a constant collision frequency ($p=0$, $\gamma=1$) leads to solutions of Eq. (36) that are in error by more than 50%.

Substitution of Eq. (33) into Eq. (36) leads to a cubic equation for κ:

$$-\frac{3}{2}\alpha(\bar{p}-p-1)\kappa^3+\left(\frac{3}{2}\alpha\bar{p}-\frac{5}{2}p+p^2\right)\kappa^2 + \left(\gamma+2p-\frac{7}{2}\right)\kappa-(\gamma+2p)=0. \tag{38}$$

The solution can be effected after specification of the momentum transfer and inelastic collision cross sections, which in turn determine p and γ. However, there are two unspecified parameters, α and $\bar{p}$, relating to the heat flux, that need to be fixed before the problem is solved completely.

2. Fixing the parameters of the heat flux *Ansatz*

a. Case of $\nu_m(\varepsilon)\sim\varepsilon$

In this case (assuming $T_g=0$ K) $p=1$, and the heat flux *Ansatz* (25) is exact with $\xi(\varepsilon)=\varepsilon^2$, that is, $\bar{p}=2,\alpha=1$. The coefficient of the cubic term in Eq. (38) vanishes identically in this case, leaving a quadratic:

$$\frac{3}{2}\kappa^2+\left(\gamma-\frac{3}{2}\right)\kappa-(\gamma+2)=0. \tag{39}$$

Note that this equation has only real solutions and that there will therefore be no oscillatory behavior regardless of the strength of the inelastic processes, as measured by γ. For this model also, the spectral heat flux (33) is independent of κ, and has the constant value

$$Q(\kappa)=-\frac{2}{3}, \tag{40}$$

regardless of the value of γ. In the special case where only elastic collisions occur, $\gamma=1$, Eq. (39) has one negative solution, namely,

$$\kappa=-1.26. \tag{41}$$

This model is the ideal benchmark as it is the only one for which an exact heat flux *Ansatz* can be written down, namely, Eq. (20). None of the other expressions found in the literature are, however, consistent with it. This point is further discussed below.

b. Case of $\nu_m=$ const

Contrary to expectations, the constant elastic collision frequency model $p=0,\gamma=1$, is not a trivial model, and provides another useful benchmark. In this case, we take $\xi(\varepsilon)=\alpha\varepsilon^2$ in the heat flux *Ansatz*, and hence $\bar{p}=2$. There is thus only one free parameter α and Eq. (38) becomes

TABLE I. Parameters for benchmark elastic collision models. For cases where exact analytic results are known, explicit fractions are given.

p	$-\kappa$	α	Q
0	$\frac{10}{49}$	3.56	1.05
0.5	0.7	0.83	−0.35
1	1.26	1.0	$-\frac{2}{3}$

$$-\frac{3}{2}\alpha\kappa^3+3\alpha\kappa^2-\frac{5}{2}\kappa-1=0. \tag{42}$$

An exact analytic solution of the Boltzmann equation is possible for this model, and a spectrum of wave numbers κ emerges (Robson *et al.*, 2000). The member with the lowest magnitude determines the asymptotic behavior at large z, and has the value

$$\kappa=-\frac{10}{49}. \tag{43}$$

Substitution of this value of κ into Eq. (42) yields

$$\alpha=3.56, \tag{44}$$

which with Eq. (33) yields the value of the heat flux spectral density

$$Q(\kappa)=1.05, \tag{45}$$

a value also obtained from the exact solution of Boltzmann's equation.

c. Case of $\nu_m(\varepsilon)\sim\varepsilon^{1/2}$

The Boltzmann equation has been solved numerically for the constant elastic cross-section model (Li, 1999), for which $p=0.5$, $\gamma=1$, and the wave number of smallest magnitude is found to have the approximate value

$$\kappa\approx-0.7. \tag{46}$$

As for the fluid description, for all elastic collision models we take $\xi(\varepsilon)=\alpha\varepsilon^2$ in the heat flux *Ansatz* and $\bar{p}=2$. Equation (38) becomes in this case

$$-\frac{3}{4}\alpha\kappa^3+(3\alpha-1)\kappa^2-\frac{3}{2}\kappa-2=0, \tag{47}$$

which with Eq. (46) yields

$$\alpha\approx0.83. \tag{48}$$

The value of the spectral heat flux can then be found from Eq. (33) to be

$$Q(\kappa)=-0.35. \tag{49}$$

3. Summary and recommendation for the heat flux *Ansatz*

The above results are summarized in Table I. On the basis of these findings for the asymptotic regime, we might go a step further and suggest a more explicit (and therefore more useful) form of the heat flux *Ansatz* by substituting

$$\xi(\varepsilon) = \alpha\varepsilon^2 \tag{50}$$

into Eq. (20) to obtain

$$\mathbf{J}_q \approx -\frac{2\alpha}{3m}\boldsymbol{\nabla}\left[\frac{n\epsilon^2}{\nu_m(\epsilon)}\right] + \frac{1}{3}(5-2p)\frac{nq\epsilon}{m\nu_m(\epsilon)}\mathbf{E} - \frac{5}{3}n\epsilon\mathbf{v}. \tag{51}$$

This might be expected to hold for those situations where elastic collisions dominate since on the basis of dimensional considerations, Eq. (50) provides the only possible way of forming a function with the dimensions of energy squared. However, when inelastic collisions are important, one has to consider the threshold energy ε_I and the energy function $\Omega(\varepsilon)$ as well, and there are then other ways of forming $\xi(\varepsilon)$. The question of how to do this is beyond the scope of this paper.

C. Other treatments of heat flux

The heat flux is represented in the literature in many ways [see, e.g., Meyyappan and Kreskovsky (1990); Park and Economou (1990); Surendra *et al.* (1990); Passchier and Goedheer (1993); Young and Wu (1993); Lymberopoulos and Economou (1995); Yoon *et al.* (1995); Bogaerts *et al.* (1999)], but the physical basis is not always clear, as discussed below, and benchmarking against established results is rare. For example, Bulowski *et al.* (1996) postulate in their equation (14) that (in our notation)

$$\mathbf{J}_q = -\frac{10}{9}\frac{n\varepsilon}{m\nu_m}\boldsymbol{\nabla}\varepsilon, \tag{52}$$

which in the linear regime leads to a spectral heat flux

$$Q_B(\kappa) = -\frac{5\kappa}{3}. \tag{53}$$

For the constant collision frequency model, substitution of the exact value, $\kappa=-10/49$, into the right-hand side gives $Q_B \approx \frac{1}{3}$, compared with the exact value of 1.05. For the other cases shown in Table I, Q_B has the wrong sign. The usefulness of Eq. (52) is therefore not clear, given that it fails these simple tests. Otherwise, the fluid equations of Bulowski *et al.* (1996) seem to be tailored to the constant collision frequency model, and there is no comparison possible with the more general formalism presented here.

A simple Fourier type of *Ansatz*, similar to that of Bulowski *et al.* (1996), has also been adopted in Gogolides and Sawin (1992) and Boeuf and Pitchford (1995), but again the justification is unclear. Golant *et al.* (1980) have a slightly more sophisticated *Ansatz* [see their equations (7.45) and (7.48)], but this too fails the benchmark comparison test for $p=1$. More recent modeling [see, e.g., the review of Morgan (2000)] also suffers from this problem.

Surendra and Dalvie (1993) are perhaps the only authors who have previously expressed concerns similar to ours. They comment that "common expressions for heat flux, such as Fourier heat conduction, are not particularly accurate" and (while referring to the Fourier *Ansatz* or the simple assumption, $\mathbf{J}_q=\mathbf{0}$) "one of the drawbacks for fluid models is that assumptions of sometimes questionable validity are required." They find that even for simple models "heat conduction can contain significant errors." We strongly support these observations. While it is one thing to see what is wrong, it is quite another to fix the problem. We have gone one step further and have provided what we believe is a more physically sound *Ansatz* (51), which should work satisfactorily (with judicious choice of the parameter α), at least when elastic processes dominate.

V. ON THE USE OF SWARM TRANSPORT DATA IN FLUID MODELS

A. General remarks

In plasma modeling, transport coefficients are viewed as parameters which may be obtained either from experimental swarm data, from solutions of the Boltzmann equation, or from Monte Carlo simulations. We note that swarm data appearing in the literature are in the form of tables of transport coefficients versus E/N (N is the neutral number density; Huxley and Crompton, 1974) and are used by some plasma modelers either directly, as in "local-field" theories, e.g., Morrow and Blackburn (2002), or indirectly as a function of some calculated effective field, as in the relaxation continuum theory model of Makabe and co-workers [see, e.g., Makable *et al.* (1992)]. In this section it is pointed out that

- transport coefficients are strictly defined only in connection with the hydrodynamic regime, and that their use in other circumstances may be problematic;
- care must be taken with the definition and interpretation of transport data appearing in the literature to avoid any possibility of mismatch;
- what really appears in fluid models that derive from Boltzmann's equation are not transport coefficients *per se*, but rather mean-energy-dependent collisional rates, as in Eqs. (12) and (13), and it is these quantities that should be evaluated from the swarm transport data;
- to this end, it is necessary to recast the data in the literature into the form of a table of transport coefficients versus mean energy, the sort of "look-up" table advocated by Robson, White, and Makabe (1997).

These issues are addressed in Secs. V.B and V.C below.

B. Definition of transport coefficients: The hydrodynamic regime

1. Flux-gradient relations, transport coefficient definitions

For most swarm experiments (Huxley and Crompton, 1974) and in many plasma discharges away from boundaries, fields are uniform and hydrodynamic conditions prevail. Here the space dependence of macroscopic properties is carried solely by the number density $n(\mathbf{r},t)$ [Kumar *et al.*, 1980]. If in addition, gradients are weak, most terms in the left-hand side of the general fluid equations (8) and (9) can then be regarded as small and solutions constructed accordingly. In effect, the entire space dependence is effectively projected out of the problem onto the diffusion equation—see Eq. (56). This is formalized in solutions of Boltzmann's equation by the familiar density-gradient expansion of the velocity distribution function (Kumar *et al.*, 1980), which to first order[5] in ∇n at the fluid level consists in writing

$$\begin{aligned} n\mathbf{v} &= n\mathbf{v}_0 + \mathsf{v}_1 \cdot \nabla n + \cdots, \\ n\varepsilon &= n\varepsilon_0 + \varepsilon_1 \cdot \nabla n + \cdots, \\ n\mathsf{T} &= n\mathsf{T}_0 + \mathsf{T}_1 \cdot \nabla n, \\ \mathbf{J}_q &= n\mathbf{J}_{q0} + \mathsf{J}_{q1} \cdot \nabla n, \end{aligned} \tag{54}$$

where the subscript 0 refers to the spatially uniform state. The first of these is just Fick's law of diffusion,

$$n\mathbf{v} = n\mathbf{W}^{(*)} - \mathsf{D}^{(*)} \cdot \nabla n, \tag{55}$$

in which

$$\begin{aligned} \mathbf{W}^{(*)} &= \mathbf{v}_0, \\ \mathsf{D}^{(*)} &= -\mathsf{v}_1 \end{aligned}$$

are the (flux) drift velocity and (flux) diffusion tensor, respectively. We return to discuss these definitions shortly.

2. Fluid equations at the hydrodynamic level

If we substitute Eq. (54) into Eqs. (7)–(9) and linearize in ∇n we find

$$\partial_t n + \mathbf{v}_0 \cdot \nabla n + \mathbf{v}_1 : \nabla\nabla n = 0, \tag{56}$$

$$\begin{aligned} -k\mathsf{T}_0 \cdot \frac{1}{n}\nabla n + q\left[\mathbf{E} + \left(\mathbf{v}_0 + \mathsf{v}_1 \cdot \frac{1}{n}\nabla n\right) \times \mathbf{B}\right] \\ = \mu\nu_m(\varepsilon_{\mathrm{CM},0})\mathbf{v}_0 + \mu[\nu_m(\varepsilon_{\mathrm{CM},0})\mathsf{v}_1 \\ + \nu_m'(\varepsilon_{\mathrm{CM},0})\mathbf{v}_0\varepsilon_1] \cdot \frac{1}{n}\nabla n, \end{aligned} \tag{57}$$

$$\begin{aligned} -\frac{\mathbf{J}_{q0}}{\nu_e(\varepsilon_{\mathrm{CM},0})} \cdot \frac{1}{n}\nabla n = \varepsilon_0 - \frac{3}{2}kT_g - \frac{1}{2}(m+M)v_0^2 \\ + \Omega(\varepsilon_0) + \{\varepsilon_1[1+\Omega'(\varepsilon_0)] \\ - (m+M)\mathbf{v}_0 \cdot \mathsf{v}_1\} \cdot \frac{1}{n}\nabla n, \end{aligned} \tag{58}$$

where : denotes a contraction over two tensorial indices, and

$$\Omega(\varepsilon_0) = \frac{M}{m+M}\sum_I \frac{\epsilon_I(\vec{\nu}_I - \overleftarrow{\nu}_I)}{\nu_e(\varepsilon_{\mathrm{CM},0})},$$

$$\varepsilon_{\mathrm{CM},0} = \frac{M\varepsilon_0 + m\frac{3}{2}k_B T_g}{m+M}.$$

Upon equating coefficients of ∇n, we get the following hierarchy of equations:

(a) *Spatially homogeneous equations*

$$q[\mathbf{E} + \mathbf{v}_0 \times \mathbf{B}] = \mu\nu_m(\varepsilon_{\mathrm{CM},0})\mathbf{v}_0, \tag{59}$$

$$\varepsilon_0 = \frac{3}{2}kT_g + \frac{1}{2}(m+M)v_0^2 - \Omega(\varepsilon_0). \tag{60}$$

(b) *First-order inhomogeneous equations*

$$-k\mathsf{T}_0 + q\mathsf{v}_1 \times \mathbf{B} = \mu[\nu_m(\varepsilon_{\mathrm{CM},0})\mathsf{v}_1 + \nu_m'(\varepsilon_{\mathrm{CM},0})\mathbf{v}_0\varepsilon_1], \tag{61}$$

$$-\frac{\mathbf{J}_{q0}}{\nu_e(\varepsilon_{\mathrm{CM},0})} = \varepsilon_1[1+\Omega'(\varepsilon_0)] - (m+M)\mathbf{v}_0 \cdot \mathsf{v}_1. \tag{62}$$

These equations have appeared many times in the literature [see, e.g., Robson (1986, 1994); Petrović and Vrhovac (1998); Li *et al.* (2002)], in which discussions of the balance equation for the spatially homogeneous temperature tensor T_0 can also be found. It is a matter of first solving the nonlinear equations (59) and (60), then the linear equations (61) and (62), and the problem is completely solved.

3. Practical application of hydrodynamic results

Equation (60) gives Wannier's famous energy-drift velocity relation in the absence of inelastic collisions ($\Omega = 0$), while Eqs. (61) and (62) together yield the generalized Einstein relations linking the diffusion tensor with the differential mobility (Robson, 1986). Tonk's theorem (Tonks, 1937) and the equivalent electric-field concept (Heylen, 1980; in which a configuration of **E** and **B** fields is replaced by a simpler one, with an effective **E** field only) result from Eqs. (59) and (60) (Robson, 1994). Blanc's law (an expression for mobility in a mixture in terms of a mole fraction weighted sum of inverse mobilities in the respective pure component gases) and corrections thereto result from a simple extension of the above theory to gas mixtures (effectively replacing the collision terms for a single gas by a weighted sum of collision terms corresponding to each component of the mixture;

[5]It is sufficient to go to first order only provided that collisions conserve particle number. In the presence of ion-molecule reactions, attachment, ionization, etc., second-order terms must be retained (Kumar, 1980; Robson and Ness, 1986).

TABLE II. Columns required to calculate collision transfer rates from the transport coefficients.

E/N	$\mu^{(*)}=W^{(*)}/E$	$D_{\perp}^{(*)}$	$\varepsilon/e\approx\frac{3}{2}D_{\perp}^{(*)}/\mu^{(*)}$
⋮	⋮	⋮	⋮

Milloy and Robson, 1973). These widely used relations are often considered to be semiempirical laws, but are in fact firmly based upon fluid equations in the hydrodynamic limit with the collision terms of momentum-transfer theory. The accuracy of the laws, and thus the accuracy of the fluid equations themselves, has been repeatedly tested over many years and found to be typically 10%, more or less.

The hydrodynamic fluid equations are also useful for providing a physical understanding of various phenomena that are otherwise described numerically through solution of Boltzmann's equation. Examples include the frequency dependence of the mean energy of electrons in rf fields (Robson *et al.*, 1995) and anomalous anisotropic diffusion (White *et al.*, 1995; Maeda *et al.*, 1997; Robson, White, and Makabe, 1997).

The scope of the fluid equations for application in the hydrodynamic regime is clearly enormous. We have not been able to find any fluid models in the low-temperature plasma physics literature that are capable of reproducing these established results in the hydrodynamic limit. This colloquium aims to redress such inconsistencies by reconciling the swarm and plasma literature.

4. Heat flux in the hydrodynamic regime

Note that only the spatially uniform heat flux $\mathbf{J}_{q0}$ appears at the level of the hydrodynamic regime, and that this term is generally considered to be small and therefore safe to neglect. The exception to this general rule is when negative differential conductivity prevails (Robson, 1984), in which case neglecting $\mathbf{J}_{q0}$ would lead to negative, unphysical diffusion coefficients. Since negative differential conductivity occurs in many practical applications that are typically the target of plasma models, for example, electrons in Ar/CF_4 mixtures (Kurihara *et al.*, 2000), it is important to treat heat flux correctly. Methods for obtaining $\mathbf{J}_{q0}$ for use in such practical circumstances were detailed by Robson, Hildebrandt, and Schmidt (1997).

C. Application of swarm data to fluid models

1. Calculation of collision rates

We now consider how the collisional transfer rates (the ν's) in the general, nonhydrodynamic equations (8) and (9) can be evaluated from hydrodynamic swarm data, using the fact that these depend upon space and time through the local, instantaneous mean energy, rather than the instantaneous local field. Our procedure is consistent with the work of others [see, e.g., Boeuf and Pitchford (1995), and references therein], but we show how the accuracy can be improved significantly with relative ease (Robson, White, and Makabe, 1997). In Sec. III.B.2 we made some general remarks on how the evaluation might proceed and now give details in the context of the hydrodynamic theory above.

To evaluate collisional transfer rates from experimental swarm transport data, one must first systematically reduce the fluid equations down to the swarm limit in the hydrodynamic regime as demonstrated in Sec. V.B.2 [see Eqs. (59)–(62)]. From this set of equations, one can then generate relationships between collisional transfer rates and measured swarm transport coefficients, and thereby the mean energy, in a self-consistent manner.

Let us consider a dc swarm experiment (or set of experiments) which measures electron mobility and transverse diffusion coefficients as functions of E/N. In the absence of ionization and/or attachment, the latter can be interpreted as flux transport coefficients, and we can immediately construct a table with headings of the form shown in Table II. Such a table is to be interpreted as specifying the function $\mu^{(*)}(\varepsilon)$ of mean energy, which has been approximated by the generalized Einstein relation in the last entry of the table, though a more accurate representation could be specified (Mason and McDaniel, 1988). The key point, however, is that instead of trying to evaluate collision frequencies as a function of mean energy from a knowledge of cross sections, one simply replaces these collisional rates in Eqs. (8) and (9) using Eqs. (59) and (60):

- momentum-transfer collision frequency ν_m:

$$\nu_m(\varepsilon)=\frac{e}{m\mu^{(*)}(\varepsilon)}; \tag{63}$$

- elastic energy-transfer collision frequency ν_e:

$$\nu_e(\varepsilon)=2\frac{m}{M}\nu_m(\varepsilon);$$

- inelastic energy-transfer collision parameter Ω:

$$\Omega(\varepsilon)=\frac{3}{2}kT_g+\frac{1}{2}(m+M)[\mu^{(*)}E]^2-\varepsilon.$$

The functional dependence upon ε thus follows directly from swarm experimental data. Alternatively, in the absence of swarm data, one can construct the table above from theoretically determined transport coefficients, i.e., either from hydrodynamic solutions of Boltzmann's equation or from Monte Carlo simulations. In any case, a dramatic increase in accuracy results, as was shown for a time-dependent problem (Robson, White, and Makabe, 1997). The error associated with the collision transfer terms in Eqs. (8) and (9) is compensated for by re-

writing the collision frequencies according to the above renormalization process.

If nonconservative collisional processes are significant, then the procedure remains essentially the same, but with the following modifications:

- The balance equations in the hydrodynamic regime are accordingly modified (Robson, 1986) and expansion to second order in ∇n is required (Kumar *et al.*, 1980) to obtain experimentally measured bulk transport coefficients.
- One must account for the fact that swarm experiments actually measure only bulk transport coefficients (Robson, 1991), which must be corrected to give the flux quantities required in Table II. Note that Boltzmann equation calculations and Monte Carlo simulations give both flux and bulk transport coefficients.

2. Additional comments on the use of hydrodynamic swarm data in plasma modeling

As for all transport coefficients across the entire scientific spectrum, mobility and diffusion coefficients are generally defined as constants in a flux-gradient relationship (cf. the thermal conductivity in Fourier's law of heat conduction or the viscosity coefficient in Newton's law of viscosity) and are universal properties of the charged-particle–gas combination for a given E/N. In swarm physics, quantities thus defined are called *flux* transport coefficients to distinguish them from the bulk transport coefficients actually measured in swarm experiments. Furthermore, it is only in the small gradient, hydrodynamic regime with uniform fields that Fick's law applies; transport coefficients can strictly speaking be defined only under such circumstances. Swarm experiments are carefully arranged to meet these requirements. Plasmas, on the other hand, may not be in hydrodynamic states, and care must be exercised in applying results from swarm theory and experiments.

Some error must result if Fick's law is used outside its regime of strict validity as, for example, in local-field modeling of discharges with nonuniform fields in which spatial nonhydrodynamic effects exist. Furthermore, if ionization and/or attachment are also significant, the error is compounded by the use of uncorrected swarm-derived transport coefficient data. It is important to be aware of the inherent limitations of using hydrodynamic formulas out of context (Morrow and Blackburn, 2002). Note that there have been some attempts to generalize definitions to nonhydrodynamic conditions (Vrhovac and Petrović, 1999).

Finally, we remark upon the anisotropic character of diffusion, which has long been known [see, e.g., Wannier's classic paper (Wannier, 1953) for charged particles in general, and the experimental work of Wagner *et al.* (1967) for electrons in particular]. The theory of anisotropic diffusion in an electric field is in fact now textbook material (Huxley and Crompton, 1974; Mason and McDaniel, 1988). It is therefore of some concern that it is a common practice for plasma modelers to assume that diffusion is isotropic, i.e., $\mathsf{D}=D\mathbf{1}$ (Sommerer and Kushner, 1992; Boeuf and Pitchford, 1995). For electric fields only, this amounts to ignoring the significant difference [sometimes a factor of 2 or 3 (Ness and Robson, 1986)] between electron diffusion coefficients parallel $D_{\parallel}$ and transverse $D_{\perp}$ to the electric field. The oversight is further compounded by using the simple Einstein relation to determine D [see, e.g., Sommerer and Kushner (1992); Boeuf and Pitchford (1995)], rather than the by now well-established generalized form of the Einstein relations involving differential mobility (Mason and McDaniel, 1988).

In the presence of a magnetic field, anisotropy is even more marked. Magnetic and electric anisotropies are in fact coupled and there have been recent investigations that have focused on the behavior of the diffusion tensor elements in swarms in the presence of electric and magnetic fields (Ness, 1993; White *et al.*, 1999). For diagonal elements of the diffusion tensor, there can be a large disparity in their respective magnitudes. Extended generalized Einstein relations have been developed to enable quick and accurate estimates of their magnitude (Li *et al.*, 2001). For off-diagonal elements of the diffusion tensor it has been well documented that these elements can have magnitudes of equivalent order to those of diagonal elements (Ness, 1994; White *et al.*, 1999). Hence any assumptions involving a diagonal diffusion tensor in fluid models would be subject to doubt. Up to now there have been few attempts to incorporate the off-diagonal elements of the diffusion tensor into fluid models (Uhm *et al.*, 1995).

D. Hybrid models

To complete this section, we point to the existence of promising hybrid models [see, e.g., Ventzek *et al.* (1993); Fiala *et al.* (1994); Donko (1998); Bogaerts *et al.* (1999)], in which the bulk of electrons and all (or most) ions and neutrals are described by fluid equations, while nonequilibrium, "fast" electrons are treated through Monte Carlo simulation, or possibly some kinetic technique. High-frequency and/or magnetic-field kinetic effects, for example, may be more satisfactorily analyzed in this way. Hybrid codes, together with proper treatment of effects of magnetic fields, have led to an explanation of new effects in power dissipation in inductively coupled plasmas (Tadokoro *et al.*, 1998; Vasenkov and Kushner, 2003). In early hybrid codes, Monte Carlo simulation was used to calculate the ionization (source) term, and this is still often used in dc discharge modeling. Present day hybrid codes, however, are more sophisticated (Ventzek *et al.*, 1993; Rauf and Kushner, 1998).

The fluid segment of these codes still suffers from the same limitations as for the pure fluid models discussed above, and there are additional problems arising from the need to connect the Monte Carlo simulation and fluid parts. However, the advantages of the more accurate treatment of electrons is considered justification for this additional complexity. Monte Carlo simulation also

TABLE III. Benchmark tests and models for computation of plasma and swarm transport properties.

Plasma property or model	References
Reid's (inelastic) ramp model	Reid (1979); Ness and Robson (1986)
Ionization models	Lucas and Saelee (1975); Ness and Robson (1986)
Attachment models	Ness and Robson (1986)
Anomalous anistropic diffusion	White *et al.* (1995); Maeda *et al.* (1997); Robson, White, and Makabe (1997)
Time-resolved negative differential conductivity	Jelenak *et al.* (1995); Petrović *et al.* (1997); Robson, White, and Makabe (1997); White *et al.* (1998); Bzenić, Petrović, *et al.* (1999)
Frequency dependence of rate coefficients	Goto and Makabe (1990); Robson *et al.* (1995)
Negative absolute mobility	Dyatko *et al.* (1997); Dyatko *et al.* (2000); Dyatko and Napartovich (2001); Dujko *et al.* (2003)
Spatial relaxation	Robson *et al.* (2000); Li *et al.* (2001); Winkler *et al.* (2002)
Time-resolved $\mathbf{E}\times\mathbf{B}$ transport	Tadokoro *et al.* (1998); Raspopović *et al.* (2000); Petrović *et al.* (2002); White *et al.* (2002)
Temporal relaxation	Maeda and Makabe (1994); Loffhagen *et al.* (1998); Bzenić, Raspopović, *et al.* (1999); Winkler *et al.* (2002)

provides a continuous supply of data necessary for the fluid model lookup table in Table II, as discussed above, obviating the need to rely upon the literature, and in that respect the scheme is more efficient.

On the negative side, the accuracy of the hybrid codes remains to be determined through standard benchmark tests. We are aware of only one such test reported in the literature, which is the calculation of transport coefficients in dc fields (Verboncoeur *et al.*, 1996). In that case the kinetic parts of several codes were tested against a two-term Boltzmann code for the case of neon, which is not a particularly good benchmark. Even then some of the predictions proved to be well outside the expected limits of accuracy. It would seem desirable for a greater range of benchmark tests to be carried out, for example, for electrons in both **E** and **B** fields for the situations shown in Table III.

Any failure to reproduce these results may imply faulty representation of the physics within the codes. At the same time these tests should be made in such a way as to provide direct guidance as to which of these phenomena are critical and which approximations are appropriate for the accuracy required in the plasma modeling. If done properly, hybrid models have the potential to provide a significant improvement over pure fluid equations in the sense that they provide the possibility of adding some critical physics due to electron kinetics and kinetics of fast heavy particles. These improvements over pure fluid codes justify the additional tests to establish limits of their accuracy by comparisons to swarm benchmarks. Further benchmarks such as Townsend discharges (Arslanbekov and Kolobov, 2003) and abnormal glow (Marić *et al.*, 2003) may be required to establish the accuracy of other parts of the model.

VI. DISCUSSION AND CONCLUDING REMARKS

In this paper we have identified what we believe to be the key issues associated with fluid modeling of low-temperature plasma discharges and charged-particle swarms, in both hydrodynamic and nonhydrodynamic circumstances, at the level of momentum and energy balance. To simplify matters, we have not considered nonconservative collision effects, such as ion-molecule reactions, ionization, or attachment, but these could be included by the addition of appropriate terms in basic fluid equations. We have concentrated on principles and simple arguments in an attempt to promote a modicum of unity among the plethora of fluid models in the literature, and have arrived at the following conclusions:

(a) All assumptions and approximations made in proceeding from Boltzmann's equation to the fluid equations should be systematically and clearly spelled out. Furthermore, we believe that whatever the form of the fluid equations eventually derived for modeling low-temperature plasma discharges, the established results of swarm theory must be reproducible as limiting cases. One such systematic approximation of collision transfer terms, momentum-transfer theory (Robson, 1986), was demonstrated to lead to straightforward fluid equations. We believe that there is much unexplored scope for using this in the full plasma problem, and indeed fluid equations have already been set out (Robson, White, and Makabe, 1997). We readily acknowledge, however, that there may well be other, better approximate formulations possible.

(b) Assumptions and approximations concerning closure of fluid equations in general, and the heat flux term in particular, are pivotal for nonhydrodynamic situations and should also be clearly spelled out. We have made an *Ansatz* for heat flux (20) that is exact for a particular interaction model and that should provide better results than existing *ad hoc* assumptions. We have noted several closure schemes in the literature, but in general they are not consistent with this benchmark model and often are not physically sound.

(c) On the use of swarm transport data in fluid models:

(i) Expressions used to relate collisional transfer rates to swarm transport coefficients must be consistent with the conditions under which the swarm experiments were conducted.

(ii) The origin of the swarm data must be known and, if appropriate, suitably modified before implementation in the fluid models.

(iii) There is a vast literature on swarms, providing empirical relations that may be useful for a fluid description of plasmas.

Benchmarking of numerical calculations, whether fluid, kinetic, Monte Carlo, or hybrid in origin, is essential for establishing the integrity of the respective approaches. The swarm literature contains many models also suitable for benchmarking plasmas, and we have made a strong recommendation that this be done in particular for hybrid models.

Finally, we stress that plasma modelers have to solve a large number of serious problems including lack of cross-section data for many species that are of critical importance, poor knowledge of boundary conditions and complex surface processes, complex geometries, a large number of relevant processes having time constants that are different by orders of magnitude, numerical diffusion due to a small number of grid points, generation of a significant number of radicals that change the composition of the gas, heating of the gas, etc. Thus one may be tempted to disregard the problems discussed in this paper on the basis that empiricism gives results of sufficient accuracy under certain circumstances. However, one never knows whether the empirical model will fail under different circumstances. Thus we firmly believe that it is essential to pursue a better foundation for the basic equations and procedures that are implemented in models of low-temperature plasmas.

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