Is the classical two-term approximation of electron kinetic theory satisfactory for swarms and plasmas?

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Abstract

The "two-term" approximation (representation of the electron distribution by the first two terms of an expansion in spherical harmonics in velocity space) continues to occupy a central role in the low temperature plasma physics literature, in spite of the mass of evidence illustrating its inadequacy in the swarm (free diffusion) limit for many molecular gases. Part of the problem lies in the failure of many authors to specify quantitatively what they mean when they say that the two-term approximation is "acceptable". Thus for example, an error of 10% in transport coefficients may well be acceptable in many plasma applications, but for analysis of highly accurate swarm experiments to compare with ab-initio and beam derived cross-sections, 0.1% or less is required, making "multi-term" analysis mandatory. While reconciliation of the swarm and plasma literature along the lines of two different accuracy regimes may thus be possible, we dispute claims that the two-term approximation is generally satisfactory for inversion of swarm experiment data to obtain electron impact cross sections. The unsatisfactory nature of the accuracy assessment of other assumptions implicit in much of the modern plasma kinetic theory literature is also discussed.

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I. INTRODUCTION

The basic problem in the kinetic theory of dilute charged particles (e.g., ions, electrons, positrons, muons) in a neutral gas at equilibrium, governed by a Maxwellian velocity distribution function f_0, is to solve Boltzmann's equation [1]:

$$(\partial_t + \boldsymbol{c}\cdot\nabla + \boldsymbol{a}\cdot\partial_c)\ f = \left(\frac{\partial f}{\partial t}\right)_{\rm coll} \equiv -J(f,f_0) - J(f,f) - ... \tag{1}$$

for the charged particle velocity distribution function $f(\boldsymbol{r},\boldsymbol{c},t)$, subject to appropriate initial and boundary conditions. Here $\boldsymbol{a}$ is the acceleration on the particle, $\boldsymbol{c}$ and $\boldsymbol{r}$ are the particle's velocity and position respectively and t is time. This equation is fundamental for understanding weakly ionised gases of all descriptions, be they low temperature plasmas or charged particle "swarms", ie, the test particle or free diffusion limit of a plasma, where charged-particle – charged-particle interactions, as characterised by the second and subsequent terms on the right hand side of (1), are negligible. In this case, the charged-particle–neutral collision term $\left(\frac{\partial f}{\partial t}\right)_{\rm coll} \approx$ -$J(f,f_0)$ is the same in both cases, and the kinetic equation is linear. Note that $J(f,f_0)$ is usually taken to be the classical Boltzmann collision operator in the case of elastic collisions and the Wang-Chang, Uhlenbeck, de Boer semi-classical collision operator [2] (or a small mass ratio derivative [3]) for electron-molecule collisions. The difference is that for a plasma the field term $\boldsymbol{a} = \frac{q}{m}(\boldsymbol{E} + \boldsymbol{c}\times\boldsymbol{B})$ derives from space charge/currents, which must be found self-consistently from Maxwell's equations, while for a swarm the fields are externally specified. This rider aside, it is clear that there is a substantial degree of overlap between swarm and plasma kinetic theory, a fact that is not always acknowledged in the current low temperature plasma literature - see, for example, two recent reviews [4, 5]. In particular, statements made in these two papers claiming adequacy for the "two-term" approximation of the electron distribution function, i.e., terminating the expansion

$$f(\boldsymbol{c}) \approx \sum_{l=0}^{l_{\rm max}} \sum_{m=-l}^{m=l} f_m^{(l)}(c) Y_m^{[l]}(\hat{\boldsymbol{c}})\ , \tag{2}$$

with $l_{\rm max} = 1$, appear to be at odds with the results of "multi-term" kinetic theory of electrons swarms ($l_{\rm max}$ arbitrary) developed since the early 1970's [5–11]. (In equation

(2), and in other cases below where convenient, we have supressed the explicit space-time dependence.) Recent articles [12, 13] re-analyse the two-term approximation in the context of the swarm-plasma connection, and confirm some of these previous results without actually acknowledging them. All in all we are concerned by the lack of awareness apparent in much of the the modern low-temperature plasma literature concerning results long established in the swarm literature, especially regarding the two-term approximation, and feel that it is timely to give up to date, definitive statements concerning its accuracy and applicability.

One important aspect of the present discussion concerns the adequacy of two-term theory for inversion of swarm data to obtain electron-atom and molecule cross sections [4, 14–16]. For this and other applications a clear specification of accuracy requirements, for example, as outlined in Section II, is required. In Section III the two-term and other approximations are discussed in more detail. We then give a brief discussion in Section IV concerning determination of electron impact cross sections from swarm and beam experiments and from ab initio quantum mechanics calculations.

II. SPECIFICATION OF ACCURACY

Since equation (1) must in general be solved numerically, and a number of approximations made, it is necessary to specify an *accuracy criterion* for one or more of the calculated quantities *at the outset*. For example, if it were desired to obtain the z-component of the average velocity (for convenience we suppress space-time dependence)

$$v_z \equiv \frac{\int d\boldsymbol{c}\; c_z\; f(\boldsymbol{c})}{\int d\boldsymbol{c}\; f(\boldsymbol{c})}\;, \tag{3}$$

to within a relative error ε, the normal practice would be to carry out successive approximate numerical solutions of (1) until the following condition were satisfied over the desired range of fields:

$$\left| v_z^{(N+1)} - v_z^{(N)} \right| \leq \varepsilon\;\; \left| v_z^{(N)} \right|\;, \tag{4}$$

where N is an integer denoting a parameter or group of parameters (see below for specific examples) associated with the numerical approximation used, and $v_z^{(N)}$ denotes the N^{th} approximation. Alternatively, more rigorously and with correspondingly greater computational

demands, one could apply an accuracy test to the distribution function $f(\boldsymbol{r}, \boldsymbol{c}, t)$ itself over all points $\boldsymbol{c}$ of interest in velocity space, or to its angular "moments" $f_m^{(l)}(c)$ (see equation (2)) over all relevant speeds, again for a specified range of field strengths. The choice is usually determined by the experimental data with which one wishes to compare the theoretical results.

It is our impression that claims made in recent review articles that

- "...the two term approximation ... does a remarkably good job of describing the transport of electrons in ... most gases" [4];
- "... restriction of the velocity distribution expansion to the lowest two terms already leads to good approximations under many plasma conditions" [17]

are representative of the opinion widely held by many in low temperature plasma kinetics (see e.g. recent reviews and articles [18–23]). Taken at face value, these claims and many others like them in the modern literature appear questionable. For one thing, it is meaningful to discuss approximations and compare results, be they of a theoretical or experimental nature, only after specifying the "error bars", as outlined above. In the context of the present discussion, a quantitative specification is required as to what "good" means. After all, an error of 10% in transport coefficients may well be acceptable in many plasma applications, and the above statements would be true if they contained that proviso. However, for analysis of highly accurate swarm experiments (required to compare with beam and/or ab-initio calculations of cross-sections), accuracy to within 0.1% or better is required, and a "multi-term" analysis or Monte-Carlo simulation is mandatory. If authors were prepared to qualify statements concerning the two-term approximation in this way, reconciliation of the swarm and plasma literature would be possible.

III. APPROXIMATIONS IN ELECTRON KINETIC THEORY

A The two-term approximation vs muliterm theory

The kinetic theory of dilute ions and electrons in gases underwent something of a revolution in the mid to late 1970's, with the lifting of two restrictions that had hitherto hindered progress:

1. Viehland and Mason [24, 25] developed a rigorous, systematic technique for solving Boltzmann's equation for ions in electric fields of arbitrary strength, for realistic ion-atom interaction potentials; and

2. Lin *et al.* [6] adapted this technique to electrons and light ions, and developed the first of what have become to be known as "multi-term" solutions of Boltzmann's equation. (Note that multi-term analysis for electrons had already commenced in a limited fashion some years previously [11], and the scaling of two-term errors with m/m_0 reported recently in [12, 13] had already been observed by then.) Subsequent multi-term techniques were developed in the early 80's (see the review article [26] and references therein).

Both areas have progressed significantly beyond that time, with many new techniques and applications (see the reviews [5, 10, 27]. In this paper we focus our attention primarily on electrons, but many of our remarks could apply equally well to ions as well, since equation (1) is the *same* for both species, and many of the numerical solution techniques are also identical [6, 10, 11, 28].

Solution of (1) for both ions and electrons usually begins with the decomposition of velocity dependence of $f(\boldsymbol{r}, \boldsymbol{c}, t)$ in (2) in directions $\widehat{\boldsymbol{c}}$ in velocity space with $l_{\max}$ an arbitrary positive integer. Early in the last century Lorentz [29] represented $f(\boldsymbol{c})$ for *light particles* in a bath of heavy scatterers by the first two terms of such an expansion, effectively by setting $l_{\max} = 1$, giving rise to the nomenclature "two-term" approximation. There are well based physical grounds [6, 30, 31] for assuming this near-isotropy in velocity space for *electrons* undergoing mainly *elastic* collisions, where kinetic energy exchange between the electron and

the neutral is relatively small, but the situation is not so clear-cut when *inelastic* collisions involving a much larger exchange of energy are important. For ions, (or to take another example in transport theory, neutrons [32], where (2) would be designated as the "$P_{l_{\max}}$" approximation) no such low order approximation has ever been considered appropriate, given that even in elastic collisions there is a large fractional energy exchange between the swarm and neutral particles, and the velocity distribution is generally significantly distorted from spherical symmetry (see also [33, 34]). The velocity distribution function of electrons in molecular gases can often assume a similar appearance [9, 35], for the same reason, *i.e.*, large energy exchange in collisions, and thus any distinction between ions and electrons based upon asymmetry of $f(\boldsymbol{c})$ is lost: for strong inelastic proceses, the electron distribution function starts to look like an ion distribution function, and it should come as no surprise that the electron velocity distribution function also requires a *multi-term* representation, *i.e.*, $l_{\max} > 1$ in (2).

These ideas are made concrete in Figs. 1-2. In Figure 1, we display the variation of the velocity distribution functions with ion mass for charged particles in a gas of hard-spheres. For smaller mass ratios (see Fig. 1A), the energy transferred per elastic collision is small and the velocity distribution function is near-isotropic. For higher mass ratios however (see Fig. 1B), the energy transferred per elastic collision increases and consequently so too does the anisotropy of the velocity distribution function. For molecular gases (e.g. CO_2 shown in Fig. 2) the energy transferred per inelastic collision is generally large (relative to the incident energy) at this field and the electron velocity distribution function has the associated anisotropic character displayed in the ion velocity distribution functions. Other recent papers dealing with visualisation of the velocity distribution function can also be consulted [34–36].

So when can these distortions in $f(\boldsymbol{c})$ be considered "small"? Although there are physical arguments and simplified estimates [6, 30, 31], ultimately the answer depends upon the imposed accuracy criterion! If the imposed accuracy criterion were applied to the drift velocity W (equal to the average velocity v_z in the stationary, uniform state) of electrons in Argon as in (4) with $N = l_{\max}$, then setting $\varepsilon = 0.1\%$ would generally lead to the

conclusion that for Argon at the field strength (and *monatomic* gases, where *only* elastic collisions are involved), the two-term approximation would probably suffice (see Table I and reference [37]). If we extend our accuracy requirements to include diffusion coefficients and keep $\varepsilon = 0.1\%$, then convergence can only be achieved by setting $N = l_{max} = 5$. On the other hand, even in a case like electrons in CH_4 (see Table I), which is very often used to illustrate the necessity of a multi-term representation, we would find that (4) would hold with $N = l_{\max} = 1$, *i.e.*, the two-term approximation for drift velocity would be satisfied over a wide range of fields, if we were to set $\varepsilon \approx 10\%$. Two-term theory would also be satisfactory for (e,CH_4) diffusion coefficients if we relaxed the accuracy requirement and set $\varepsilon \approx 20\%$. Further relaxation of the accuracy requirement would be required to make the same statement about $f_m^{(l)}(c)$ for all speeds c, and an even greater reduction would be needed for the velocity distribution function $f(\boldsymbol{c})$.

There are of course exceptions to the above general rules, and the two-term theory sometimes even gets the physics wrong [38]. For example, if we consider the reduced transverse diffusion coefficient $n_0 D_T$ for electrons in methane under the influence of an a.c. electric field of various reduced field frequencies ω/n_0 as shown in Fig. 3. At lower frequencies (viz. Fig. 3A), the instantaneous relative errors between the two-term and converged multi-term results can be as high as 50%. At a higher frequency (see Fig. 3B) we observe that the two-term approximation actually predicts oscillations that are π out of phase to those of the converged multi-term results. Instantaneously the relative error can be as high as 70%.

On another point of misunderstanding, it is sometimes claimed that the two-term approximation becomes increasingly difficult to satisfy at higher fields (see e.g. [39, 40]). There are, however, examples which clearly show that this is not the case, *e.g.*, Fig. 6 (CH_4) and Fig. 13 (CO_2) of reference [9] indicate that the two term results are worst at field strengths which facilitate the onset of significant inelastic collisions, and then become quite good at higher fields, as the relative importance of inelastic collisions declines. It is the latter which is the dominating criterion for the breakdown of the two-term approximation and *not* just the field strength per se.

B Other factors determining accuracy

1 Treatment of Ionisation

The treatment of ionisation in the kinetic theory of low-temperature plasmas and swarms is of vital importance and there often exist common approximations and assumptions in its treatment. The first approximation in widespread use in electron kinetics is the treatment of electron impact ionisation as no different from any other inelastic process, ignoring the true physics of the non-particle conserving collision process (see e.g. [17, 41]. It is rare to find any mention of the error associated with this assumption. In some cases the results are not even qualitatively correct under such an approximation [38]. In Figure 4 we display the relative error in the rate coefficient for the process of electron impact ionization in molecular oxygen, comparing the treatment as a true ionization process with that as a purely inelastic scattering process for electrons in oxygen. Over the range of fields considered here (appropriate to plasma operating conditions), the relative error increases with E/N and is as high as 120% - rendering the assumption totally invalid at such field strengths. An equivalent study has also been performed in Ar [42] and the errors are qualitatively similar. The specially-constructed Lucas-Saelee model shows that glossing over this assumption could be problematic and the reader is referred to the calculations of [9, 43] for further discussion. Otherwise it appears that error estimates can only be made on a case by case basis, and, like the other assumptions mentioned above, we feel strongly that it is incumbent on those making this particular approximation to provide such accuracy estimates.

Another issue of importance in the treatment of ionization is the partitioning of post-collision energy between the scattered and ejected electrons. In the absence of any further information, workers are generally forced to make an assumption on the partitioning in their kinetic theory. For field strengths operative in plasma discharges however, the transport properties can be very sensitive to the partitioning scheme. Such sensitivity is displayed in the ionisation rates in Figure 4 and extends to other transport properties. Two schemes (i) post-collision energy is shared equally (50-50) and (ii) post-collision energy is given all to one electron while the other has zero kinetic energy (0-100), are compared with the

rate determined by assuming all fractions of the distribution of post-collision energy are equiprobable. The 50-50 scheme tends to underestimate the equiprobable partitioning rates while the 0-100 scheme tends to overestimate them. Differences of up to 15% exist between the schemes over the range of fields considered. Equivalent calculations have been performed in Ar [42] and the sensitivity demonstrated. Thus, in the extraction of ionization cross-sections from swarm data one must keep in mind that transport properties are sensitive to both the magnitude of the ionization cross-section and the partitioning scheme for the post-collision energy.

2 Mass ratio expansion

Another approximation in widespread use in the electron kinetic theory is the representation of the actual Boltzmann integral collision term $J(f, f_0)$ by an approximation based upon the smallness of the electron-neutral molecule mass ratio m/m_0. We emphasise that as far as both elastic and inelastic collisions are concerned, and before any such approximation is made, $J(f, f_0)$ is of the *same mathematical form* to start with, regardless of whether we are talking about ions, electrons, positrons or muons, and that in *all* cases, it can be represented by the *same* mass ratio expansion, which we write formally as:

$$J(f, f_0) \approx \sum_{p=0}^{p_{\max}} (\frac{m}{m_0})^p \; J^{(p)}(f, f_0) \; . \tag{5}$$

Here $p_{\max}$ is a nonnegative integer, which, like $l_{\max}$, must strictly speaking be chosen by incrementation using an accuracy criterion like (4), with $N = p_{\max}$. For electrons $m/m_0 \sim 10^{-4} - 10^{-5}$ and we expect the series (5) to converge very rapidly. Indeed we have found that the normal approximation of setting $p_{\max} = 1$ and 0 for the elastic and inelastic collision terms respectively leads to an error in all electron transport coefficients of $\varepsilon \sim 10^{-5}$ over all fields of interest [34, 44]. The accuracy of the mass ratio approximation for the *distribution function* is expected to be somewhat less, but we have not carried out such an investigation. The great advantage of truncation at first order in mass ratio is that $J^{(0)}$ and $J^{(1)}$ can be represented in differential-finite difference form [3, 30, 45–47] and special

numerical techniques can be brought to bear to effect very accurate solutions of the approximate Boltzmann equation [5, 17]. However, it does not make sense to endeavour to obtain such numerical solutions to an accuracy greater than that of the approximate equation itself. At the time of writing, relative errors to within $\sim 10^{-5}$ for the numerical solution of the $p_{\max} = 1$ approximated Boltzmann equation were proclaimed (see e.g. [48]), which lies at the bounds of the limits of validity of the approximated collision term itself. If such high precision is required, the only way to obtain a true estimate of the *overall* accuracy of the calculation is to go to the $p_{\max} = 2$ representation of $J(f, f_0)$, but then advantages of simplification in mathematical form are lost, and it is not clear whether the special numerical techniques developed for the $p_{\max} = 1$ representation could continue to be applied.

For particles heavier than electrons, more terms in (5) are certainly required to achieve the desired overall accuracy. For muons ($m_\mu \approx 200\ m_e$) for example, it has been found that transport coefficients accurate to 0.1% or so over the range of fields of interest can be achieved only by taking $p_{\max} = 3$ [49]. As expected in this case, the two-term approximation is inadequate for this accuracy criterion, and $l_{\max} = 2$ or more in (2) is required. For ions of mass equal to the neutrals, *i.e.*, $m/m_0 = 1$, it is found that $p_{\max}$ as high as 6-7 and $l_{\max}$ as high as 4-6 can be required for an accuracy of 1% or better in the transport coefficients over field strengths of interest [34].

IV. ELECTRON-MOLECULE IMPACT CROSS SECTIONS FROM SWARM DATA

A Swarm and beam experiments, ab initio quantum mechanical calculations

The greatest significance of swarm experiments in relation to fundamental and applied topics in electron scattering lies in their ability to furnish accurate cross sections at low energies. While advances in crossed-beam techniques have enabled measurement of relative angular distributions to an accuracy of a few percent even at energies as low as 0.5 eV [50], the necessity of determining from these data integral cross sections, by extrapolation to small and large angles followed by normalization, introduces significant additional error.

As a consequence, at energies below about 1eV, analysis of transport data from swarm experiments remains the most accurate way of determining integral cross sections for electron scattering. Of special interest is electron-molecule scattering, where analysis of transport coefficients can determine rotationally and vibrationally inelastic cross sections at energies very near threshold, typically from fractions of a millivolt (for rotational excitation) to a few tenths of an eV (for vibrational excitation). Swarm and beam derived cross sections have been complemented by many substantial *ab initio* quantum mechanical calculations for simple molecules [51]. The modern approach to determination of cross sections over a wide range of energies is a combination of all three techniques.

In principle, swarm-derived cross sections are most accurate at energies where at most one inelastic process is energetically accessible. When several inelastic channels are open, there may be problems with a lack of uniqueness, which can grow more serious with increasing energy above threshold. It is here that *ab initio* theoretical calculations can play a crucial role, e.g., by fixing rotational cross sections and thus allowing the determination via transport analysis of a unique vibrational cross section. A joint study of ro-vibrational excitation in molecular hydrogen along these lines is currently underway aimed at resolving the long-standing controversy surrounding the e, H_2 vibrational cross section [15, 52, 53]. Our experience with this problem in particular highlights the importance of examining with the greatest of care statements concerning the accuracy of transport analyses and the cross sections they yield.

B Unfolding the cross sections from transport data

Low energy electron impact cross sections can be "unfolded" from measurements of transport properties obtained from swarm experiments, in conjunction with solutions of (1). The early methods of deriving cross sections from swarm measurements of the drift velocity and transverse diffusion coefficient in an electric field has been extensively reviewed by Huxley and Crompton [14] and became accepted as competitive and complementary to other established techniques, such as crossed beam or total attenuation experiments. Originally the

two term theory was used to unfold the transport data, but in the early 1980's Haddad [7] tested this approximation using the then recently developed muliterm code [6], and found it wanting in a number of cases. More recently in the Heidelberg experiments [54, 55] using electric and crossed electric and magnetic fields, the four quantities W, Lorentz deflection angle α, and both longitudinal and transverse diffusion coefficients D_L and D_T respectively were measured, with accuracies of around 0.2% obtained for each of the former, and around 2% for each of the latter over the range of fields considered. It seems reasonable to demand that theoretical calculations should have an even greater precision, say better than 0.1%. It is precisely in this accuracy range that the two-term approximation is found to be wanting for many molecular gases, as evidenced by the large number of studies conducted over the past two decades (see the reviews [5, 10, 26]). At best, one is never sure that the two-term approximation holds to a specified accuracy until a multi-term analysis (or Monte Carlo simulation) has been run as a check. Schmidt and coworkers [54, 55] subsequently developed an fully automated procedure for extracting cross sections from their experimental transport data. With the multi-term analysis, accuracies typically around $1-3\%$ were achieved for the elastic momentum transfer cross section in the range $0.01 - 1.0eV$ for the gases considered, with uncertainties of $5 - 10\%$ for inelastic cross sections.

If the claim that two term approximation is typically used for swarm analysis were true (and it is *not* in our experience), then we suggest the accuracy of cross sections derived on that basis is therefore open to serious doubt and the such cross-sections are possibly unsuitable for comparison with other theoretical and/or experimental techniques.

V. CONCLUDING REMARKS

This note suggests that when viewed in the cold hard light of numerical precision, the continued uncritical use of the two-term approximation as the standard-bearer of electron kinetics is unwarranted. In our opinion, the distinction between electron and ion kinetics becomes blurred for many molecular gases, and multi-term analysis should be regarded as the norm, not the exception, just as it is for ions in general. Windows of opportunity

certainly exist for continued use of the two-term theory in many cases, e.g., for plasma modelling requiring only accuracy of $\gtrsim 10\%$. However, in general when high precision of around 0.1% or better is required, a multi-term code (or Monte Carlo simulation) is mandatory for *all* molecular gases. For intermediate accuracies, say $\sim 1\%$, it is difficult to give a definitive statement, but we recommend that a multi-term analysis (or Monte-Carlo analysis) be applied, if only for checking the two-term results.

We have discussed two other approximations implicit in much of the literature on electron kinetics: the approximation of the collision operator to first order in mass ratio, and the approximate treatment of ionisation purely as an inelastic process. The former assumption leads to errors in *electron* transport coefficients typically around 10^{-5} and would only come into question when solutions of the Boltzmann equation to a similar accuracy or better were required. The latter cannot be glossed over and must be analysed on a case by case basis. We feel strongly that it is incumbent on authors making such an approximation to investigate and report just what error is involved.

Our overall recommendation is that in general the usual practice in physics of specifying "error bars" should be followed in the low-temperature plasma literature, and that in particular, the accuracy of calculations based upon the two-term approximation and other well known assumptions implicit or explicit in this literature should be clearly spelled out.

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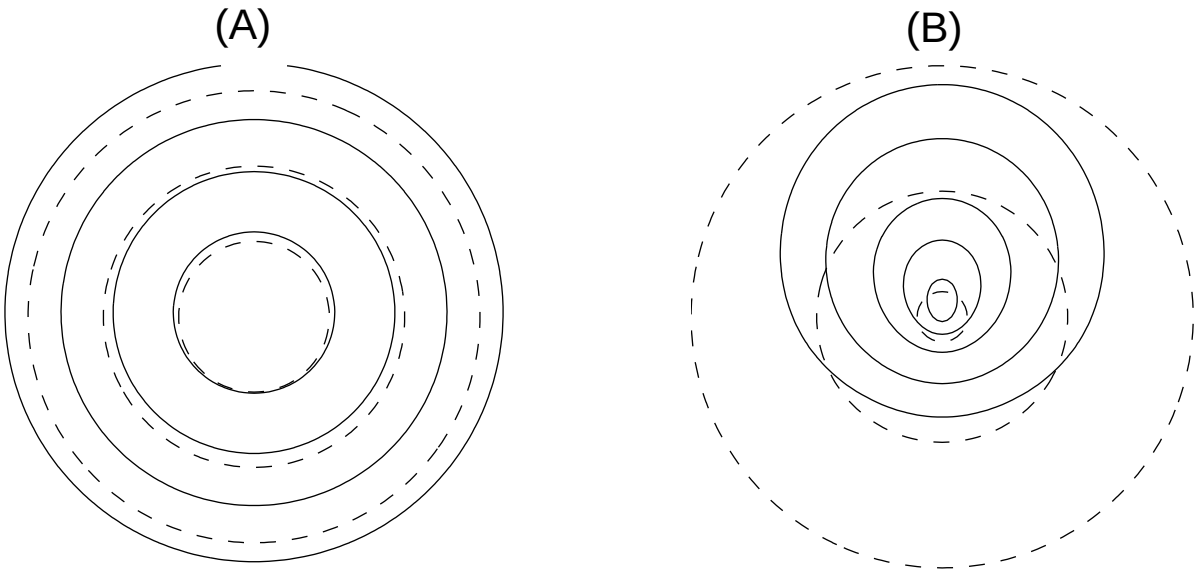

FIG. 1: Contour plot of the velocity distribution function for ions of masses 4×10^{-4} amu (A) and 4 amu (B) in a model gas of hard spheres of mass 4 amu at $E/n_0 = 1$Td (Note: the electric field force is vertically upward). The cross-section is independent of energy and is fixed at 6Å^2 while the neutral gas temperature is fixed at 295K. The values of the solid contours from largest to smallest radii in (A) are 0.3,0.6,0.9.1.2,1.5 $(\text{eV})^{-3/2}$; (B) are 25,50,100,150,200 $(\text{eV})^{-3/2}$. The energy scale is denoted by the dashed circular contours. The values of the energy scale contours of increasing radii are respectively for (A) are 0.3,0.6,0.9 eV while for (B) are 0.01, 0.05, 0.1 eV.

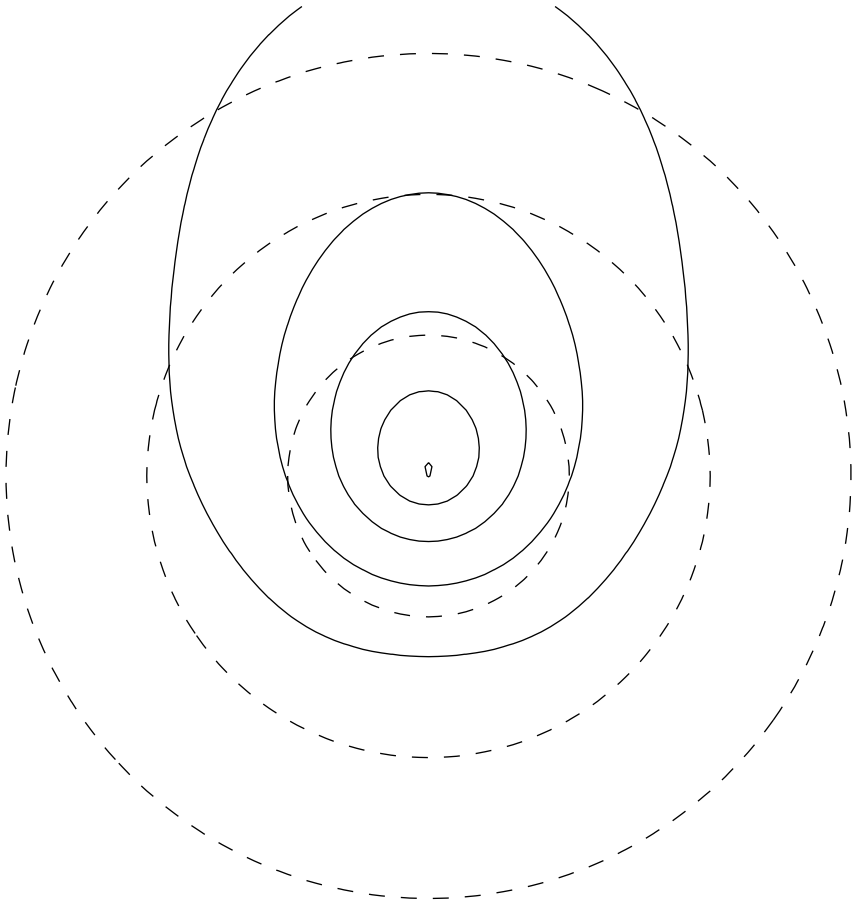

FIG. 2: Contour plots of the velocity distribution functions for electrons in carbon-dioxide at 293K subject to a reduced electric field of $E/n_0 = 5$Td (Note: the electric field force is vertically upward). The value of the solid line contour heights from largest to smallest radii are, respectively 0.5,1,2,4,8 $\text{eV}^{-3/2}$. The energy scale is indicated by the dashed concentric circular plots of increasing radii referring to 0.3,0.6 and 0.9 eV respectively.

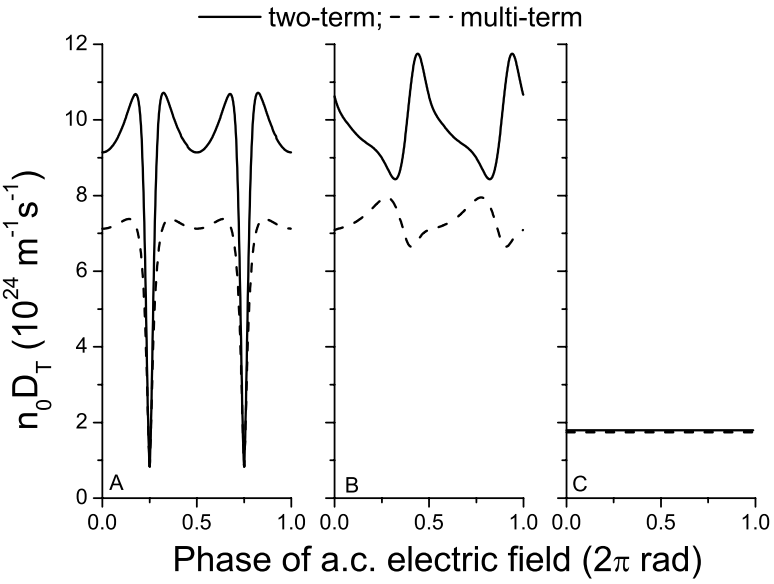

FIG. 3: Comparison of the two-term and multi-term profiles of the transverse diffusion coefficient for CH_4 under the influence of an a.c. electric field at various applied reduced angular frequencies ω/n_0 (rad m^3 s^{-1}): (A) 1x10^{-17} (B) 1x10^{-15} (C) 1x10^{-14}. ($E/n_0 = 5\cos\omega t$ Td where n_0 is the neutral number density and t is time).

TABLE I: Convergence in the l-index of the drift velocity and diffusion coefficients for electrons in methane (CH_4: E/n_0=5Td; Ar: E/n_0=0.5Td. n_0 is the neutral number density.)

Gas	Transport	l_{max}					
	coefficient	1	2	3	4	5	6
CH_4	W ($\times10^4$ms^{-1})	10.6	9.89	10.0	10.0	10.0	10.0
	n_0D_T ($\times10^{24}$m^{-1}s^{-1})	4.94	4.53	4.62	4.59	4.61	4.60
	n_0D_L ($\times10^{24}$m^{-1}s^{-1})	1.87	2.18	2.02	2.05	2.05	2.05
Ar	W ($\times10^3$ms^{-1})	2.515	2.515	2.515	2.515	2.515	2.515
	n_0D_T ($\times10^{25}$m^{-1}s^{-1})	1.737	1.685	1.693	1.690	1.691	1.691
	n_0D_L ($\times10^{24}$m^{-1}s^{-1})	2.042	2.043	2.043	2.043	2.043	2.043

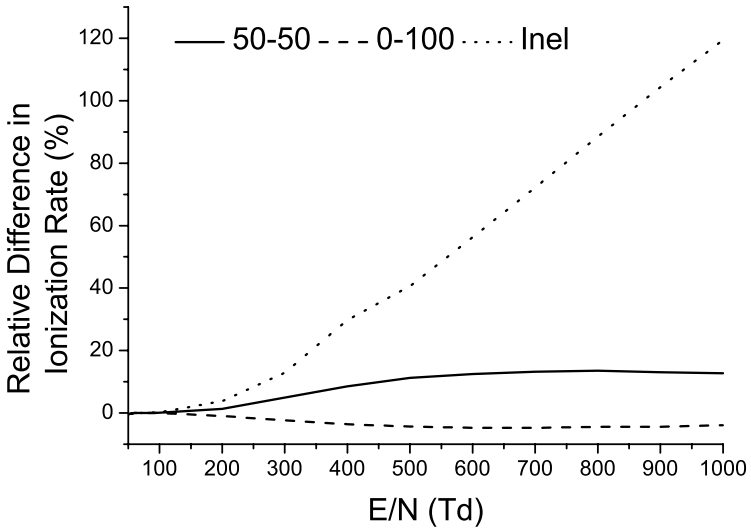

FIG. 4: Comparison of ionization rates for electrons in molecular oxygen at 300K calculated using the various approximations for the ionization process . The results are displayed as relative differences to those rates calculated assuming all fractions of the post-collision energy are distributed uniformly between scattered and ejected electrons. The various schemes are (i) ionization treated as an inelastic collision process (INEL) (ii) post-collision energy is equally shared between the scattered and ejected electrons (50-50) and (iii) post-collision energy is all given to one electron the other remaining stationary (0-100).