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Kinetic Phenomena in Transport of Electrons and Positrons in Gases caused by the Properties of Scattering Cross Sections

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Abstract. Collisions of electrons, atoms, molecules, photons and ions are the basic processes in plasmas and ionized gases in general. This is especially valid for low temperature collisional plasmas. Kinetic phenomena in transport are very sensitive to the shape of the cross sections and may at the same time affect the macroscopic applications. We will show how transport theory or simulation codes, phenomenology, kinetic phenomena and transport data may be used to improve our knowledge of the cross sections, our understanding of the plasma models, application of the swarm physics in ionized gases and similar applications to model and improve gas filled traps of positrons. Swarm techniques could also be a starting point in applying atomic and molecular data in models of electron or positron therapy/diagnostics in radiation related medicine..

1. Introduction

In this paper we present a survey of some of the recent results of the physics of swarms of charged particles (we will confine our interest to electrons and positrons). Our first and necessary point is to illustrate some of the recent results obtained by the group(s) at the Institute of Physics in Belgrade (together with our collaborators). We also wish to illustrate

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how swarm physics connects on one side to atomic and molecular collisions (and thus to overall atomic and molecular physics) and on the other to non-equilibrium plasmas and their numerous applications. As the topic of swarms has not been addressed frequently at ICPEAC (although one of its satellites, Electron-molecule Collisions and Swarms, covers the topic very well) this presentation will necessarily be rather broad but not very detailed.

Collisions of electrons, atoms, molecules, photons and ions are the elementary processes occurring in plasmas. It may be argued that the level of individual collisions is the most fundamental level of phenomenology required to describe non-equilibrium collisional plasmas. That is so for two principal reasons: the first being that the duration of the collisions is many orders of magnitude shorter than the mean free time between the collisions. Thus we may bury all the quantum mechanics into the cross sections and basic properties of the energy levels and molecules. As a result, we may even use classical trajectories for charged particles and thus the Monte Carlo technique has had so much success. The second reason is related to the first and it is that the De Broglie's wavelength of particles is usually small compared to the mean volume per particle in the gas, at least until we reach very high densities (e.g. as in liquids). Thus electrons collide with only one target per collision.

A reductionist view of the science which dominated in the past declared that the more basic the phenomena were, the more fundamental they were. In that view of the world, the field theory and mathematics on their own may explain the psychological states of humans! A more realistic view which, luckily, prevails today is that there are layers of phenomenology, each with its own rules and foundations and each providing its accomplishments that are not trivially predicted at the more basic levels. In this way we may construct a path between atomic and molecular physics and the numerous modern applications of low temperature plasmas. As previously mentioned, there is no need to go deeper than the physics of collisional processes (including a range of collisions with surfaces). The next stage is the physics of swarms where collisions join the statistical physics and kinetic theory in addition to the surface processes. More detailed presentations of this realm of physics have been given in earlier texts [1,2], while more recent reviews have been given in references [3-5]. It is possible to say that little in the papers presenting the cross section data prepares us for the complex kinetic phenomena that evolve in the swarm physics, such as negative differential conductivity or negative absolute mobility [6,7].

The next layer of phenomenology is that of low temperature or more accurately non-equilibrium plasmas (NEP). It brings in space charge and other plasma effects, chemistry and many more different inputs. Swarm physics, represented by its kinetic phenomena, together with atomic collision data are the building blocks of the NEPs but little prepares us for the phenomena such as the spewing of the plasma bullet (ionization front) from the glass tube where an atmospheric pressure plasma jet (APPJ) is formed [8,9]. This device often produces a plasma bullet (ionization front) that actually moves faster, and is bigger and brighter, in the supposedly hostile world of atmospheric gases once it leaves the region of high field between the electrodes wrapped around the tube where more favourable gases for its formation dominate. But even at this level one cannot really envisage why and how such plasmas may induce, for example, preferential differentiation of human (periodontal ligament mesenchymal) stem cells into one out of four possible types of the cells [10].

Finally, one should welcome another change in the attitude that happened recently. It has been slightly over 100 years since the discovery of electron. Its discoverer J.J. Thompson toasted at Christmas receptions: "To the electron and may nobody find its application.". Needless to say, the previous century being labeled as the century of the electron means that some applications were eventually found. The attitude that applied is not fundamental has, however, changed. Luckily non-equilibrium plasmas offer one of the quickest and most abundant fronts of development of new applications and each application brings in requirements for new phenomena to be included. For example, attempting to apply NEPs to

medicine requires an understanding of a large part of the relevant medical knowledge. Following publication of a major review by David Graves, of the mechanisms coupling reactive species from the plasmas with biological triggers [11], there is now no room for plasma and atomic physicists to claim that medical processes need not be understood from their viewpoint, they simply have to learn them (Latin terms and all). Nevertheless one could claim that at the deepest relevant level leading to such applications, one may find atomic and molecular collisions however remote from the final outcome those may be [10].

2. Electron swarms in gases, cross section data sets and kinetic phenomena

Swarms may be simply defined as ensembles of particles (in this case electrons and positrons) moving in the background gas under the influence of external fields (if charged), limited by the walls of the vessel. These particles do not suffer effects of any significance due to interactions between themselves (Coulomb force, shielding of the external field) and also have negligible chances of colliding with the remnants of previous collisions. In other words, they move in the external fields affected mainly by the collisions with the pristine background gas.

Swarms bring transport theory and other aspects of statistical physics to the table, and often effects of surfaces may be needed albeit only in specific situations (e.g. a Steady State Townsend experiment). The transport may be well represented by a single particle distribution function, so the standard Boltzmann equation (BE) is appropriate. However, due to 7 degrees of freedom, a complex theoretical treatment is required for solving the BE. Due to the complexity of the cross sections (the dependence on the energy that can only be tabulated) and hence collision operator, the final result has to be obtained numerically. The resulting energy distribution function is however not something that can be measured, and the swarm physics focuses on averaged properties such as transport coefficients (drift velocity, diffusion tensor, ionization coefficients) or rates for specific processes (excitation or chemical).

Initially swarm physics was developed when techniques of electrochemistry were applied to study properties of charged particles in gases, especially when their elementary nature became obvious. However, they quickly proved to be a very good source of data for cross sections for the dominant processes especially after numerical solutions to the BE became available. The advantage of the technique was originally significant, as it provided good absolute calibration, and results for He were only matched by theory and beam techniques some ten to twenty years later [1]. Most importantly, if a full set of swarm facilities is used the resulting cross section set provides good number, momentum and energy balances for the charged particles in the gas and is thus directly applicable in the modeling of plasmas.

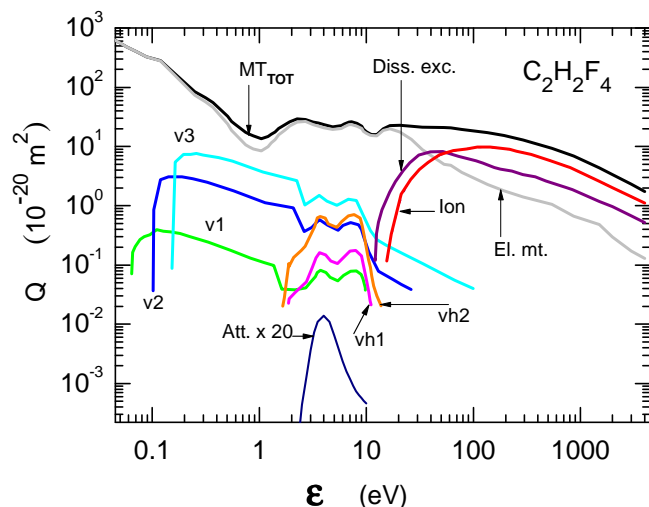


Figure 1. The cross section set for $C_2H_2F_4$ [12], MT- total momentum transfer, v-vibrational modes, Att-attachment, ion-ionization, El mt elastic momentum transfer, Diss. Exc. Dissociative excitation..

Recent swarm derived cross section sets cover many gases so we shall give only one example, for the 1,1,1,2-tetrafluoroethane ($C_2H_2F_4$) molecule [12]. Transport coefficients measured by a Pulsed Townsend technique were converted to cross sections, based on an initial set that was available in the literature due to S. Biagi. Results are shown in Figure 1.

A disadvantage of the swarm technique is that it is indirect i.e. it involves guessing of the cross section set and then comparing the calculated transport coefficients to the experimental data until agreement is

reached. In addition its resolution is poor, especially at higher energies, and the results potentially suffer from non-uniqueness.

Reliable results are usually obtained from drift velocities and characteristic energies (diffusion coefficient divided by mobility) for energies up to 1 – 2 eV, while typical electron energies in relevant plasmas are higher. If the ionization coefficient is used in the analysis one may extend the energy range of the set. Assuming that the measured ionization cross sections are very accurate we can fit the ionization rate by adjusting the middle range electronic excitation cross sections or dissociation to the ground state (which are often incomplete).

The accuracy of the resulting cross sections depends very much on the accuracy of the transport theory (or the corresponding Monte Carlo simulation (MCS)). Numerous tests need to be made to check the codes against specially designed benchmarks, for various aspects of the transport or properties of the processes [13]. On the other hand one needs to reopen, in a systematic fashion, the issue of anisotropic scattering. At low energies, due to the randomizing effect of frequent collisions, isotropic scattering is a good approximation provided that the momentum transfer cross section was obtained with that approximation. It has been shown, however, that for mean energies in excess of 20 eV or even for smaller energies when inelastic processes are very strong, one needs to include differential cross sections i.e. a full anisotropic model.

A plethora of atomic and molecular processes acting at the same time, that use up the energy gained from the field, leads to the formation of the shape of the electron energy distribution function (EEDF), and furthermore, but less obviously to the dependencies of the averaged properties, i.e. the transport and rate coefficients. Those processes finally lead to the functionality of low temperature plasmas and their many applications. From the viewpoint of fundamental physics the most interesting aspect of the swarm physics are the so-called kinetic phenomena [3,5]. Those represent an often counter intuitive behaviour of the collective properties, that cannot be predicted from the individual trajectories or from the shape of the cross sections (at least not without some experience). Those may be loosely classified according to the primary source of their existence (although the cross section magnitudes, shapes and properties are generally relevant) :

- **Dependence on the rates of momentum transfer and inelastic processes:** anisotropic diffusion; diffusion heating/cooling; enhanced mobility; negative differential conductivity (NDC); spatial separation of fast and slow particles-i.e the energy gradient, ...
- **Non conservative transport:** attachment heating/cooling; negative absolute mobility; difference between flux and bulk transport coefficients; positron NDC for bulk drift; skewed Gaussians, ...
- **Magnetic field induced:** magnetic field cooling; ExB drift; ExB anisotropy of diffusion,...
- **NDC for positrons in liquids**
- **Time dependent fields:** anomalous diffusion; limited relaxation; phase delays at high frequencies; time resolved NDC; transient negative diffusivity, heating of electrons due to cyclotron-resonance effects,...
- **Non-hydrodynamic:** Frank Hertz oscillations and Holst Oosterhuis structures; runaway ions; runaway electrons; thermalization/equilibration (non-local transport); increasing mean energies close to the boundaries; back-diffusion.

The fundamental reasons for these effects lie in the interplay between the times or spatial scales required for relaxation of number, momentum and energy, and in the interplay between the source of energy and momentum (i.e. the external field) and the processes that dissipate those properties. One example of kinetic phenomena is particularly important for the world of Atomic and Molecular physics. Absolute negative mobility has been predicted by several authors. The phenomenological explanation requires a group of electrons to be released with energy of 2 eV in a mixture of argon with 0.5% of F₂ (or any other gas with a large thermal attachment). The majority of the electrons would be accelerated by the field and would have an increasing chance to collide. If scattering is isotropic then 50% of the electrons will scatter backwards and join the smaller group of electrons that move against the field. Although those lose energy, the decreasing cross section will reduce their chances of

redirection until they thermalize in the region of the Ramsauer Townsend (RT) minimum. There, the electric field would again accelerate the majority of the electrons in the expected direction. Thus, for a while, electrons would on average move against the field and current – mobility would be negative. If one adds small amount of F_2 the thermal attachment will eat up the thermalized electrons not allowing them to accelerate and the current would be negative perpetually [14,15]. Of course it has been shown that this does not mean that we have a source of free energy although entropy is in principle reduced. However we pay the price by producing a lot of negative ions which contribute to an even greater growth of entropy [16]. The importance of this example is that it provides a situation where atomic processes may be used to tailor the distribution function, and in essence act as Maxwell's demon (in this case the thermal attachment). It is also not a man made device. Requirement to maintain the second law of thermodynamics requires us to separate at least two kinds of transport coefficients. For drift velocities we may have an average over all electrons in all of the space (the flux drift velocity), while we may also follow the center of the mass of electrons and determine its velocity (the bulk drift velocity). The distinction between these two is due to the changing number of particles (non-conservative processes; attachment, positronium (Ps) formation for positrons or ionization for electrons) and the difference may be associated with the validity of the second law of thermodynamics [16].

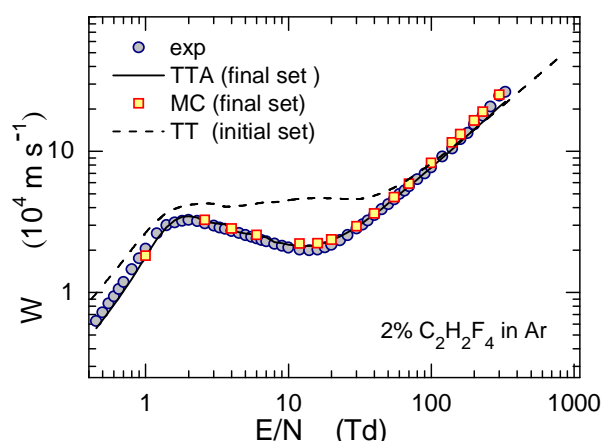


Figure 2. Fit of the experimental drift velocities (open circles) in $C_2H_2F_4$ with cross sections from figure 1 [12] (solid circles and line) and with an older set that was previously in use (see [12]). NDC is well developed between 2 Td and 20 Td.

We shall also show one example of the related phenomenon of negative differential conductivity (NDC), where drift velocity is reduced as the field increases and the mean energy increases due to the reduced control of the energy by inelastic process and increased randomization of directions in momentum transfer collisions. This example also shows how the structure in the drift velocity may be used to improve the uniqueness of the cross sections, as the calculations with another, similar, set does not show the experimentally observed NDC [12].

Kinetic phenomena, being shaped by the cross sections, provide an opportunity to strengthen the ability to normalize the cross section sets and also to modify and even define some of the applications or plasma properties. Thus those effects should be

recognized and their implications understood when one wants to model collisional NEP.

3. Direct application of swarm data and models in the physics of ionized gases

In some cases when space charge is not excessive, swarms may be used as a direct representation of the ionized gas (often under those conditions, however, all conditions are not met to call such systems a plasma). The first example is the physics of Townsend discharges. The fact that swarm models are exact for such circumstances (in the limit of vanishingly small currents), makes them perfect to determine atomic and molecular processes in gas phase [17] and on surfaces and to study gas breakdown as well sometimes even revealing new phenomena in experimental observations [18]. Further direct application of swarm data and theory is in attempts to optimize gaseous dielectrics. In principle, two directions of research are dominant. The first is replacing SF_6 by more ecologically acceptable gases and the second is to produce mixtures of such gases that would allow their operation without the need for expensive high pressure vessels.

Another direction of research where swarm models and data are used abundantly (albeit that field has almost severed its connections with the swarm community) is that of the gas filled particle

detectors, including the nowadays most popular Resistive Plate Chambers (RPC)[19]. Using the Monte Carlo code developed to study swarms and obtain cross sections, and the newly established cross section set, we were able to calculate the time response of such devices [20] that agrees well with experiments. These results may now be used to optimize gas mixtures, operating conditions, chemistry and control the degree of ionization to speed up the counting rate. Other types of gas filled detectors may be modeled in a similar fashion.

The most important aspect in application of swarm physics, is in so called low temperature plasmas (we prefer to call them non equilibrium plasmas-NEP). We could spend much space on this issue, but it is only covered here as a brief introduction with more being found in reference [21]. The kinetic theory and the transport data all enter fluid models and together with the solution for the field distribution are the foundation of the theory. The hybrid models use the same data together with the cross section sets **that have to be complete** and thus be tested by the swarm technique, as do the kinetic codes. As one example we can describe capacitively coupled RF plasmas, which have sheaths close to the electrodes and with high fields that increase on one side and decrease on the other. During the reduction of the field electrons diffuse into that region and get accelerated into the plasma when the field starts increasing again. The diffusion flux of electrons is defined by the longitudinal diffusion coefficient, the one that shows anomalous behavior due to inability of the electron energy to respond the changes in the fields. This inability follows from the finite relaxation time of the electron energy which is strongly affected by the shape of the elastic cross section. On the other hand, for inductively coupled RF plasmas the ExB drift opens new channels to feed energy into the plasma [3]. Most models however assume constant (in space and time) transport coefficients, and neglect additional components of drift velocity and diffusion when magnetic fields are present. Nevertheless it has been difficult to impress upon the plasma modeling community that their models, when applied to simple low space charge limit benchmark situations, **should be able to replicate the swarm benchmarks**. Completing this exercise, however, would open many issues on the available cross sections and would forge a stronger link between atomic and molecular collision physics and the plasma modeling community. At the same time it would make binary collision experts aware of the data needs for the numerous plasma applications,

Another issue is that of the pertinent theory. As mentioned above, most frequently spatial and temporal uniformity are assumed in modeling. This is seriously wrong in cases of sharp gradients, in the profiles of plasma properties when hydrodynamic expansion of the theory is not an option (and is still being used in almost all cases). One such example is that of the streamers. Streamers are the basis for most high pressure discharges and recently a theory has been developed that includes proper treatment of transport across strong gradients in various streamer properties. Although the space charge made the final profile very robust, the improved theory produced results that had a significant change in the speed of propagation [22]. Streamers are an essential component of a number of atmospheric plasmas including lightning, sprite discharges in the upper atmosphere and atmospheric pressure plasma jets, which are being championed for novel medical procedures while having some intriguing physics on their own [8]. Other atmospheric discharges like aurora are often modeled [23] by using measured distribution functions from the atmosphere, in a procedure that resembles swarm models. It seems possible that a similar analysis should be made with distribution functions calculated having in mind all the available data and conditions at high altitudes.

4. Positrons in gases: swarms and (swarms in) traps

The absence of swarm experiments for positrons, with two exceptions [24], made us adopt a strategy that we do not advise for electrons. That is to collect the available cross sections, which are now generally available for several of the most important gases [25-27], and calculate the transport coefficients hoping to identify new kinetic phenomena that would justify building new swarm experiments. It was found that for gases with a strong positronium formation cross section, skewing of the positron swarm occurs due to preferential loss at the front of the group leading to a major reduction (NDC) for the bulk drift velocity. One such example is water vapour [28], which is critically

important for applications of positrons in medicine. Assuming that a set of cross sections is sufficiently complete, we may proceed to model tracks of positrons in water vapour allowing also for assessment of nanodosimetry [29].

One should be aware that some of the critical devices in positron physics contain gas to reduce the energy of positrons, below the threshold for Ps formation, and then to further cool them so that the outgoing beams might have a very narrow energy spread. the Penning-Malmberg-Surko trap is usually separated to three stages, with pressures ranging from 10^{-3} Torr to 10^{-5} Torr, with pure N_2 at the front and mixture of N_2 and CF_4 in the last stage [30,31]. We have been able to apply the code originally developed for electron swarms (and tested against all known benchmarks) to model the Surko trap [32]. In figure 3 one can see the development of the distribution function from a single beam, through to multiple beams (due to inelastic collisions with electronic excitations), and to gradual development of the low energy distribution which becomes dominant and eventually decays to the Maxwell Boltzmann distribution at room temperature [33]. This is fully analogous to the equilibration of electron swarms with initial beam, followed by Frank Hertz like effects during the first collisions and subsequent development of a broad energy distribution demonstrating also that interpretation of the experiment using swarm phenomenology is appropriate (including of course a good set of cross sections). Having this tool it became possible to determine other aspects of trap operation: losses, optimum choice of potential drops and geometry. It led to some new proposals such as the idea of S. Marjanović for the inversion of the gases, whereby CF_4 would be used at the trap front and with the mixture still at the last stage, with lower potential drops that would help avoid Ps formation and allow efficiencies of up to 90%.

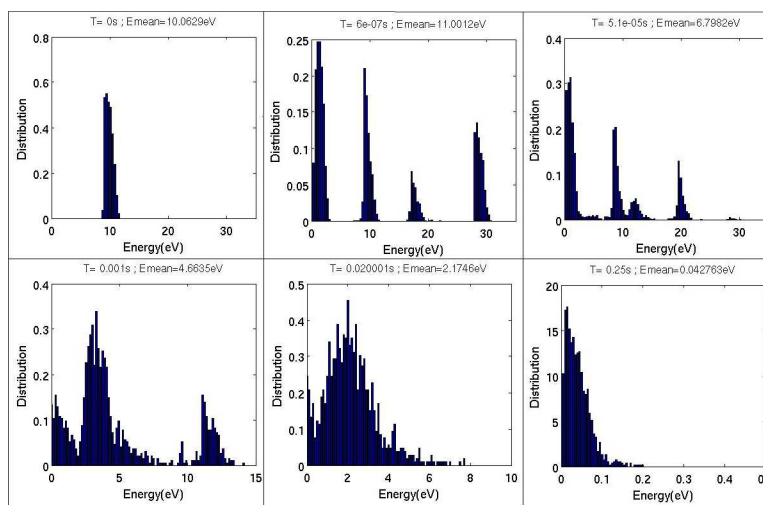


Figure 3. Temporal development of the energy distribution function in a positron trap [33].

A large number of elastic collisions, which happen during thermalization, leads to an expansion of the positron swarm in the trap. For many applications, however, increased density is required and thus additional narrowing in the final stage may be required. For this purpose a rotating wall stage has been developed that may operate in two regimes: single particle [34,35] and plasma regimes. A theory of the former has been provided in reference [36] where viscosity was added to a simple transport equation allowing the experiments to be fitted. In our

approach a swarm based Monte Carlo codes has been used with realistic sets for the cross sections [37]. The role of each of the processes has been elucidated, and it is possible to characterise all the salient features of the rotating wall trap. As the system develops with an entire ensemble, it appears that the term single particle rotating wall should be replaced by the swarm regime of the rotating wall.

5. Conclusion

The realm of the physics of ionized gases controlled by collisions without a significant effect of the Coulomb interaction between charged constituents, is known as swarm physics. It is in this area that the kinetic phenomena are observed most directly. The tools of swarm physics allow us to cross the path from the elementary microscopic collisional processes all the way to the macroscopic properties of swarms, plasmas and other forms of charged particle ensembles and their applications. It appears that for gas filled systems the phenomenology, tools and data of swarm physics provide the best way

to understand and even optimize the devices and their applications, while crossing the gap between microscopic cross sections and the large scale practical devices.

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