Electron transport and propagation of negative streamers in liquid-phase xenon

Saša Dujko
Institute of Physics
University of Belgrade
Belgrade, Serbia
sasa.dujko@ipb.ac.rs

Ilija Simonović
Institute of Physics
University of Belgrade
Belgrade, Serbia
ilija.simonovic@ipb.ac.rs

Danko Bošnjaković
Institute of Physics
University of Belgrade
Belgrade, Serbia
dbosnjak@ipb.ac.rs

Zoran Lj. Petrović
Institute of Physics
University of Belgrade and Serbian Academy of Sciences and Arts
Belgrade, Serbia
zoran@ipb.ac.rs

Ronald White
College of Science and Engineering,
James Cook University
Townsville, QLD, Australia
ronald.white@jcu.edu.au

***Abstract*—The Monte Carlo method, initially developed for charged particle swarms in neutral dilute gases, is extended and generalized to investigate the transport processes of electrons in liquid-phase noble gases by accounting for the coherent and other liquid scattering effects. Electron transport coefficients, including the electron mobility, diffusion coefficients and ionization coefficient, are calculated as a function of the reduced electric field in liquid-phase xenon. Calculated transport coefficients are then used as an input in the classical fluid model to investigate the dynamics of negative streamers. Using the language of the contemporary kinetic theory of plasma discharges, in the present work among many important points, we investigate how various representations of inelastic energy losses in inelastic scattering events affect the electron transport and the macroscopic streamer properties.**

Keywords—Monte Carlo, liquid noble gases, transport coefficients, streamers, fluid models

I. INTRODUCTION

Understanding of the behavior of free electrons under the influence of electric field in liquids is of interest in both fundamental physics and in numerous technological applications. Those applications include the interdisciplinary field of plasma medicine [1], liquid dielectrics [2], plasma-water purification [3] and liquid particle detectors [4]. In particular, liquid-phase noble gases are used in the technology of the time-projection chambers, which are designed for detection of cosmic radiation and neutrinos [4], as well as in the search for dark matter particles [4]. Further optimization of such applications requires an accurate understanding of electron transport coefficients, streamer properties and the physical processes involved.

In our previous studies, we investigated the elastic scattering of electrons from liquid-phase argon [5] and liquid-phase xenon [6]. Electron transport coefficients were calculated in the sub-excitation energy region, e.g., for those values of the reduced electric fields, E/n_0, (where E is the electric field strength and n_0 is the neutral atom density) for which the mean energies are well below the first inelastic threshold. More recently, we have investigated the way in which electron transport coefficients are influenced by various representations of the inelastic energy losses in liquid-phase xenon with the special emphasis on the explicit effects of ionization (or interband transition having in mind that the electrons are quasi-free particles in liquid xenon) [7]. We have also discussed the fluid modeling methods with the aim of understanding electron transport and streamer propagation across the gas-liquid interfacial regions [8]. In this paper, as a part of our ongoing investigations of electron transport in liquid-phase noble gases in an electric field, we study the transition from an electron avalanche into a negative streamer ionization front and its propagation in liquid xenon. Calculations are performed using a fluid model in local field approximation. Using the electron scattering cross sections for both gas and liquid xenon, transport coefficients of electrons are calculated in Monte Carlo simulations to serve as input data for a fluid model used in this study.

We begin this study by briefly reviewing the basic elements of the fluid theory used to simulate negative streamers in liquid xenon in section 2. In section 3.A, we present the electron transport coefficients as a function of the reduced electric field. In the same section, we briefly discuss the cross sections for electron scattering in liquid xenon and the basic elements of the Monte Carlo method used for calculating electron transport coefficients. The development of negative streamers without formation of expanding gaseous filaments is discussed in section 3.B. In section 4 we present our conclusions and recommendations for future work.

II. THEORETICAL METHOD

Simulations of negative streamers in liquid xenon are performed by using the classical fluid model. In this model the electron flux is obtained by assuming a steady-state of the momentum balance equation, and that the electron energy of the field-directed motion is much greater than the thermal contribution [9]. The generalized one-dimensional continuity equation for the electron number density is

$$\frac{\partial n_e(x,t)}{\partial t} = \frac{\partial}{\partial x}\left(D_L \frac{\partial n_e(x,t)}{\partial x} + \mu_e n_e(x,t) E \mathrm{sgn}(E)\right) + \left(\nu_i - \beta n_p(x,t) n_e(x,t)\right), \quad (1)$$

where $n_e(x,t)$ and $n_p(x,t)$ are the number densities of electrons and positive holes, respectively, which are functions of the coordinate x and time t. In this equation D_L and μ_e are the longitudinal diffusion coefficient and the electron mobility, respectively, $\boldsymbol{E}$ is the electric field,

This work was supported by the Grants No. OI171037 and III41011 from the MPNTRRS and also by the project 155 of the Serbian Academy of Sciences and Arts. RW acknowledges the support from the Australian Research Council.

oriented along the x-axis, while ν_i and β are the ionization rate and the recombination coefficient, respectively.

In addition to the electron continuity equation (1), the time evolution of the number density of positive holes is described by the number balance equation

$$\frac{\partial n_p(x,t)}{\partial t} = \left(\nu_i - \beta n_p(x,t)\right) n_e(x,t) \,, \tag{2}$$

where transport of positive holes has been neglected over the transient time scales considered in this study, owing to the significantly reduced mobility and diffusion of positive holes in liquid xenon [10].

The model is realized in a 1.5 dimensional (1.5D) setup. Thus, the total electric field in the system is evaluated as the sum of the uniform external electric field and the electric field due to space charge:

$$E(x,t) = E_0 + \frac{e}{2\epsilon_0\epsilon_r}\int_0^l \left(\operatorname{sgn}(x-x') - \frac{x-x'}{\sqrt{(x-x')^2+R_0^2}} \right) \left(n_p - n_e(x',t) \right) dx' \,, \tag{3}$$

where E_0 is the external electric field, e is the elementary charge, ϵ_0 and ϵ_r are the vacuum permittivity and the relative permittivity, respectively, and l is the length of the system. In this model, the space charge is contained inside cylinder with radius R_0 and the charge density varies along the axial direction only.

The recombination coefficient is given by the scaled Debye formula

$$\beta = \xi\beta_D = \xi\frac{4\pi e\mu_e}{\epsilon_0\epsilon_r} \,, \tag{4}$$

where β_D is the Debye recombination coefficient and ξ is the scaling factor which is taken to be 0.1 [11].

The above fluid equations are closed, assuming the local field approximation. According to this approximation the input terms, including μ_e, D_L, ν_i and β are assumed to be functions of the local instantaneous electric field. In the numerical implementation of our fluid model, the spatial discretization is performed by using the second order central finite difference, while the fourth order Runge–Kutta method is used for the integration in time.

III. Results and Discussion

A. Transport coefficients of electrons in liquid xenon

In case of electrons, the transport data needed for the solution of fluid equations (1) and (2) are μ_e, D_L, and ν_i. These electron transport data are calculated by using the Monte Carlo method. The Monte Carlo method, initially developed for charged particle swarms in neutral dilute gases [12], has been recently extended and generalized by including three effective scattering processes, which give a good representation of the coherent scattering of low energy electrons in non-polar liquids [7]. The validity of our Monte Carlo method has been tested by calculating the transport properties of electrons in the Percus Yevick model liquid. It was found that our results are in an excellent agreement with those calculated by Tattersall et al [13].

In order to account for excitations in liquid xenon, the set of inelastic atomic excitation cross sections of the Hayashi database was modified to form a set of excitation cross sections for intermediate excitons in liquid state. For example, the intermediate $n = 1\left[\Gamma\left(\frac{3}{2}\right)\right]$ and $n' = 1\left[\Gamma\left(\frac{1}{2}\right)\right]$ excitons have been observed at 8.2 eV and 9.45 eV, respectively [14]. The former has parentage in the excited atomic $6s'[3/2]_1$state, while the latter has parentage in the $6s'[1/2]_1$ state. As these intermediate excitons have a unique parentage, via the isolated atom's excited states, we thus approximate the cross sections for intermediate exciton excitations by cross sections of the corresponding atomic excitations. Likewise, the cross section for interband transitions is approximated by the cross section of the electron impact ionization, from the Hayashi's cross section set. However, the cross section is shifted by 2.1 eV, so that the threshold of the ioization is 9.22 eV in liquid xenon. This value corresponds to the $\Gamma\left(\frac{3}{2}\right)$ band gap in liquid xenon, which is the energy difference between the uppermost valence and the bottom of the conduction band. For simplicity, in the rest of this work the interband transition and the inelastic collisions will be referred to as ionization and excitations, respectively. For more details on the band structure and cross sections for electron scattering in liquid xenon, the reader is referred to [7].

In the present calculations, we cover a range of reduced electric fields between 10^{-3} and 10^3 Td (1 Td = 10^{-21} Vm^2). The number density of xenon atoms is 1.4×10^{28} m^{-3}, while the temperature of the background liquid is 163 K. In our simulations, we usually follow 10^6 electrons except in the limit of the lowest values of E/n_0. Due to numerous elastic collisions in which only a fraction of the initial electron energy is transferred to a heavy xenon atom, the efficiency of energy transfer is very low in the limit of the lowest E/n_0. As a consequence, the relaxation of energy is very slow and requires a large computation time. In order to optimize the simulation speed, the simulations were usually begun with 10^4 electrons and after the relaxation to the steady state the electron swarm scaled up to 10^6 electrons. The details of this procedure are given elsewhere [7].

In Fig. 1 we show the dependence of the electron mobility on E/n_0. It should be noted that the density normalized mobility $n_0\mu$ and density normalized diffusion coefficients n_0D_L and n_0D_T shown in Fig. 2, are not independent of the neutral atom density [15]. These transport coefficients are given as a function of E/n_0, so that any linear dependence on density (as occurs in the dilute-gas limit) has been removed. Thus, we have a true comparison of the gas and liquid phases.

Calculations are performed assuming the following two scenarios: (*i*) no electronic excitations (case 1), and (*ii*) all electronic excitations from the gas-phase are included (case 2). Both the bulk and flux mobility components are shown. The bulk transport coefficients, are associated with the swarm's centre of mass transport and spread about its centre of mass. In Monte Carlo simulations, the bulk transport coefficients may be determined from the rate of changes of the appropriate averages of the positions of the electrons in the configuration space. The flux transport coefficients should be interpreted in terms of averages over the ensemble in velocity space. For example, the flux mobility is associated with the average velocity of the ensemble in the swarm. In liquid and gas xenon, these two sets of transport coefficients are equal in the absence of ionization.

For comparison, the theoretical [6,7] and experimental values [16] of mobility are displayed at the same figure, along with the mobility in gaseous xenon.

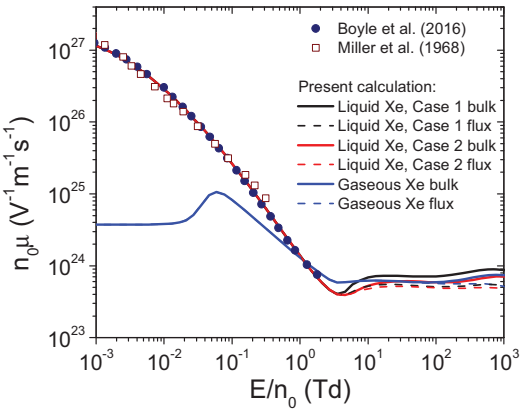

Fig 1. Variation of the electron mobility with E/n_0. Our Monte Carlo results, for liquid and gaseous xenon, are compared with the measurements (Miller et al. (1986)) and theoretical calculations (Boyle et al. (2016)). It should be noted that all three dashed lines for the flux properties emerge from the solid lines of the same colour above 10 Td.

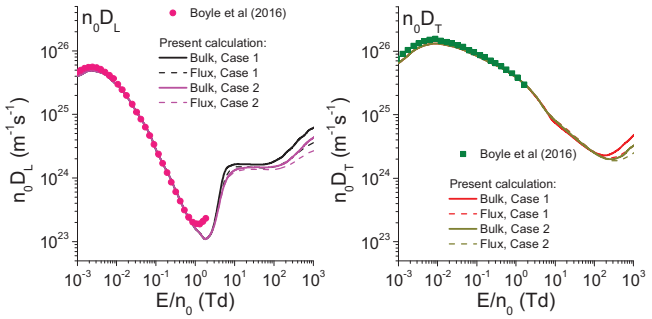

Fig. 2. Variation of the longitudinal (left panel) and transverse diffusion coefficients with E/n_0. Our Monte Carlo results are compared with the theoretical calculations (Boyle et al. (2016)).

The agreement between our Monte Carlo results and those obtained from a multi term solution of the Boltzmann equation is excellent.

For the lower values of E/n_0 we observe that the electron mobility in the liquid phase exceeds the mobility in the gas phase by more than two orders of magnitude. This is a clear sign of the reduction of the rate of momentum transfer of the lower energy electrons in liquid xenon. The lowering of the rate of momentum transfer follows from the modification of the scattering potential and the coherent scattering effects. Due to these liquid scattering effects, the electric field accelerates electrons more efficiently in liquid xenon than in gaseous xenon, which in turn leads to a significant enhancement of the electron mobility as compared to the gas xenon.

In Fig. 2 we show the variation of the longitudinal and the transverse diffusion coefficient as a function of E/n_0. The agreement between our Monte Carlo results and those evaluated from the solution of Boltzmann's equation for the lower values of E/n_0 is very good. For the higher values of E/n_0, we observe that the diffusion coefficients are reduced with an increase of the number of excitations used in the modeling. Due to the explicit effects of ionization, the bulk values of both n_0D_L and n_0D_T are greater than the corresponding flux values.

In Fig. 3 we show the variation of the ionization rate coefficient with E/n_0. We observe that the ionization rate is monotonically increasing function of E/n_0 for both the liquid- and gas-phase xenon. We also observe that the ionization rate is increased by reducing the number of

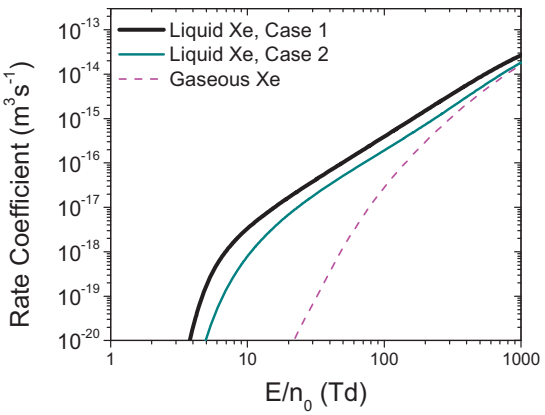

Fig. 3. Variation of the ionization rate coefficient with E/n_0. Calculations in gaseous xenon are compared with those in liquid xenon.

excitations. Likewise, the ionization coefficient in liquid xenon is significantly greater than the ionization coefficient in gaseous xenon. This can be expected due to the reduction of the threshold for ionization in the liquid phase. In addition, electrons can lose a significant amount of energy in a wide range of inelastic scattering processes at energies lower than the threshold energy for ionization in gaseous xenon. Likewise, there is a far lower number of inelastic scattering processes with thresholds which are lower than the threshold for ionization in the liquid phase compared to the gas phase.

B. Negative streamer fronts in liquid xenon

In Fig. 4 we show the formation and propagation of a negative streamer under the influence of the externally applied electric field of 77 Td. The initial Gaussian is positioned in the close vicinity of the cathode. The electric field is oriented to the right, so the negative fronts propagate to the left. The initial densities of electrons and positive holes are equal reflecting the macroscopic neutrality of a plasma. In addition, these densities are selected in such way that the space charge effects are negligible. The values of l and R_0 are set to 5×10^{-5} m and 1×10^{-5} m, respectively. The particular value of R_0 is chosen as an educated guess taking into account the width of the initial distribution and the spreading due to transverse diffusion. The length of the system l is determined by the requirement that the streamer velocity relaxes to a stationary value. The simulation in the gaseous xenon employs transport data for electrons for the gas phase scaled to the liquid density. We employ the bulk transport coefficients as an input in fluid simulations of negative streamer fronts in both the liquid and gas phases.

In the absence of gas filaments and trapping of electrons in the density fluctuations, the general features of the streamer profiles in the liquid xenon are the same as those of the streamers in gases [7]. We observe that the streamer front caries an overshoot of electrons, generating a thin space charge layer that screens the electric field in the streamer interior behind the front. In this screened interior region, the density of charge is not constant. The electron number density and the positive hole density are further reduced due to the recombination of electrons and positive holes. A similar decrease in the electron number density in the streamer interior and behind the front, is observed for streamers in electronegative gases, where electron attachment consumes the lower energy electrons. We observe that the streamer formation as well as streamer

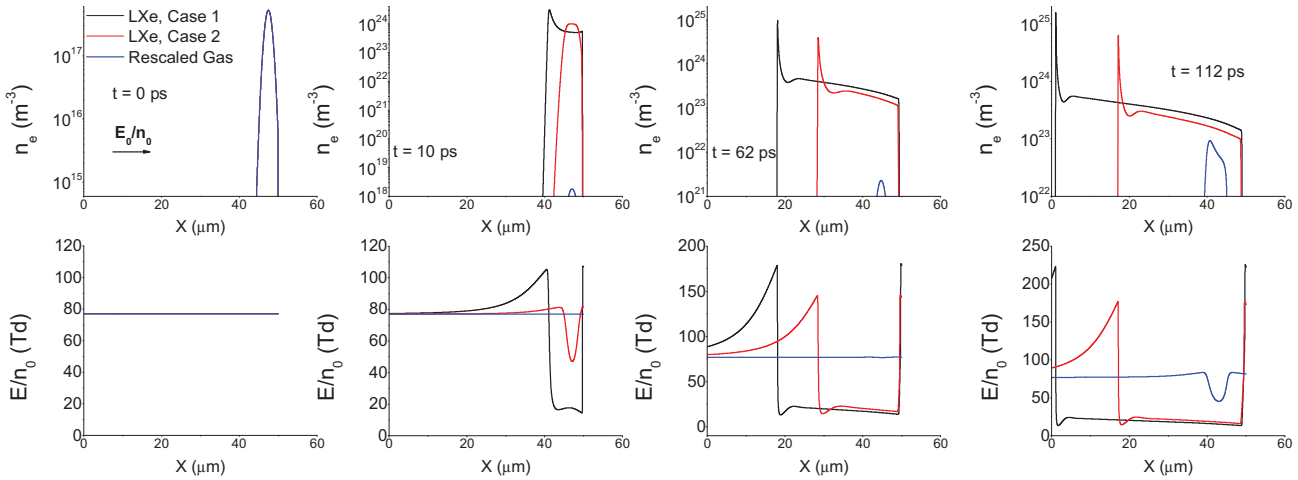

Fig. 4. The formation and propagation of a negative streamer in liquid xenon for E/n_0 = 77 Td. Here n_e refers to the electron number density while E/n_0 refers to the reduced resultant electric field.

propagation are greatly influenced by the number of excitations in the model. For example, the streamer velocity and the electron number density in the streamer interior are increased by reducing the number of excitationsIt can also be observed that the transition from an electron avalanche into a streamer is much slower in the case of the rescaled gas than in the liquid phase.

IV. Conclusion

Using a Monte Carlo simulation technique and 1.5 dimensional classical fluid model, we have investigated the influence of inelastic energy losses on both the transport properties of electrons and dynamics of negative streamers in liquid xenon. The cross sections for inelastic scattering and interband transitions of electrons in liquid xenon are approximated by using the cross sections for electron scattering on an isolated xenon atom. The *ab initio* cross section for elastic scattering in liquid xenon is taken and adopted in order to include the effects of coherent scattering and atomic potential screening which are critical for low-energy electron scattering. Calculations in the liquid phase are augmented by those in the gas phase. It is found that, above approximately 1 Td there is a significant difference between the values of transport properties determined by employing different representations of the inelastic energy losses. The electron mobility and diffusion coefficients, as well as the ionization rate coefficient are reduced with increasing number of excitations in the model. Likewise, it is found that the streamer properties, including the streamer velocity, the ionization degree in the streamer interior and the distribution of electric field strongly depend on the number of excitations which are included in the model.

The present work will be extended in a near future by investigating the propagation of positive and negative streamers in a point-to-plane geometry. We will also consider the influence of density fluctuations and gas filaments, as well as trapping of electrons in these structures, on both the electron transport and the streamer dynamics.

References

[1] M. G. Kong, G. Kroesen, G. Morfill, T. Nosenko, T. Shimizu, J. van Dijk and J. L. Zimmermann, "Plasma medicine: an introductory review," New J. Phys. vol. 11, 115012, 2009.

[2] V. Y. Ushakov, V. F. Klimkin and S. M. Korobeynikov, "Impulse Breakdown of Liquids," (Berlin: Springer), 2007.

[3] B. Jiang, J. Zheng, S. Qiu, M. Wu, Q. Zhang, Z. Yan and Q. Xue, "Review on electrical discharge plasma technology for wastewater remediation," Chem. Eng. J. vol. 236, 348, 2014.

[4] E. Aprile and T. Doke, "Liquid xenon detectors for particle physics and astrophysics, " Rev. Mod. Phys. vol. 82, 2053, 2010.

[5] G. J. Boyle, R. P. McEachran, D. G. Cocks and R. D. White, "Electron scattering and transport in liquid argon, " J. Chem. Phys. vol. 142, 154507, 2015.

[6] G. J. Boyle, R. P. McEachran, D. G. Cocks, M. J. Brunger, S. J. Buckman, S. Dujko and R. D. White, "*Ab initio* electron scattering cross-sections and transport in liquid xenon, " J. Phys. D: Appl. Phys. vol. 49, 355201, 2016.

[7] I. Simonović, N. A. Garland , D. Bošnjaković , Z. Lj. Petrović, R. D. White and S. Dujko, "Electron transport and negative streamers in liquid xenon, " Plasma Sources Sci. Technol. vol. 18, 015006, 2019.

[8] N. A. Garland, I. Simonović, G. J. Boyle, D. G. Cocks, S. Dujko and R. D. White, "Electron swarm and streamer transport across the gas–liquid interface: a comparative fluid model study, " Plasma Sources Sci. Technol. vol. 27, 105004, 2018.

[9] S. Dujko, A. H. Markosyan, R. D. White and U. Ebert, "High-order fluid model for streamer discharges: I. Derivation of model and transport data," J. Phys. D: Appl. Phys. vol. 46, 475202, 2013

[10] O. Hilt and W. F. Schmidt, "Positive hole mobility in liquid xenon, " Chem. Phys. vol. 183, 147, 1994.

[11] N. Y. Babaeva and G. V. Naidis, "Modeling of positive streamers in liquid argon, " Tech. Phys. Lett. vol. 25, 91, 1999.

[12] S. Dujko, Z. M. Raspopović and Z. Lj. Petrović, "Monte Carlo studies of electron transport in crossed electric and magnetic fields in CF_4, " J. Phys. D: Appl. Phys. vol. 38, 2952, 2005.

[13] W. J. Tattersall, D. G. Cocks, G. J. Boyle, S. J. Buckman and R. D. White, "Monte Carlo study of coherent scattering effects of low-energy charged particle transport in Percus-Yevick liquids, " Phys. Rev. E vol. 91, 043304, 2015.

[14] P. Laporte, J. L. Subtil, U. Asaf, I. T. Steinberger and S. Wind, "Intermediate and Wannier Excitons in Fluid Xenon, " Phys. Rev. Lett. vol. 45, 2138, 1980.

[15] Y. Sakai, "Quasifree electron transport under electric field in nonpolar simple-structured condensed matters," J. Phys. D: Appl. Phys. vol. 40, R441, 2007.

[16] L. S. Miller, S. Howe and W. E. Spear, "Charge Transport in Solid and Liquid Ar, Kr, and Xe, " Phys. Rev. vol. 166, 871, 1968.