

# Simulations of strongly coupled ions at a liquid-gas interface in time projection chambers

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**Abstract:** Molecular dynamics computer simulations of a Lennard-Jones fluid are used to describe the liquid-gas interface created in massive two-phases argon time projection chambers. The study intends to show the formation of the interface from a single phase state. The results allow us to set the initial conditions needed for future simulations of this type of systems. Additionally, once the interface is easily recreated, it shows a first approximation to the experimental scenario with the introduction of ions, as well as an explanation of the limitations encountered during the simulations.

## I. INTRODUCTION

Time projection chambers (TPC) have become a popular three dimensions tracking detector in many physics experiments since their invention in 1972 [1]. Tracking trajectories and interactions inside the chamber enables the identification of incoming particles. This is particularly useful considering the difficulty in detection weakly interacting particles. As a result, there has been an interest in the field of neutrino physics for this kind of detectors. Its utility has already been proved on different experiments. For instance, the Deep Underground Neutrino Experiment (DUNE) used in the detection of neutrinos [2] [3]. Furthermore, TPCs are being proposed for new experiments in other fields of physics, shown by the DarkSide-50 experiments [4] in hopes of detecting Weakly Interacting Massive Particles (WIMPs), considered to be the constituents of dark matter. This increased interest for these detectors instigate the study of the physics underneath.

There are several types of TPCs. Single phase detectors, composed of one phase medium which present a simplified scheme, and double phase detectors whose lower energy threshold allows the detection of smaller collisions. Liquid noble gases are used as mediums in these detectors due to their low background radiation, low energy threshold and low production cost [5]. As a result, massive liquid argon time projection chambers (LArTPCs) have gained interest as a proposed medium, as seen in the ICARUS experiment [6]. Consequently, we focus our study on them.

Double phase LArTPC are composed of two regions. A bulk section, where a large quantity of liquid argon is stored, and a thinner layer of gas, where the signal is extracted. The chamber is placed between a cathode, at the bottom of the liquid, and an anode, at the top of the gas, in order to collect the electrons. Usually, optical modules are placed at the electrodes to detect the scintillation lights produced inside. How-

ever, some LArTPC collect the electrons directly.

External energetic particles interact with the atoms inside the bulk. As a consequence, the collision produces the ionization and excitation of the atoms at rest, thus, liberating an electron-ion pair. Additionally, the relaxation process produces a photon, causing the scintillation light. The application of an external field separates the electron from the ion, preventing their possible recombination. Depending on the purity of the medium and its ions concentration, it is possible for the charges to recombine before reaching the detectors. In case the electron reaches the detector, the electric field increases its kinetic energy to ionize and excite the atoms in the gas, producing a Townsend discharge [7]. This avalanche increases the number of free electrons in order to secure the electrical conduction through the gas. Moreover, it makes possible the detection of a signal proportional to the amount of electrons crossing the interface from the liquid. On the other hand, it produces an increase on the number cations that are pushed by the electric field to the cathode.

Once the electrodes have collected the signals, a 3D position of the collision is obtained, using the 2D positions captured and the drift time between the signals detection [1].

The collected data enables the identification of different types of collisions, allowing us to focus on the detection of a specific interaction.

A recent study of the ions dynamics in massive LArTPCs [7] presents some concerns with these types of detectors, such as the lost of uniformity in the field due to the ions remaining longer times in the chamber than the electrons, and the injection of cations from the gas layer through an interface. In our study, we focus on the formation of this interface in double phase TPCs detectors. Due to their accuracy, the understanding of the behaviour between the particles and the charges at the interface is required. The distribution of the cations may add some insight on their interactions, especially as the avalanche increases their amount near the interface.

Liquid-gas interfaces are difficult to describe macroscopically. As a result, macroscopic studies tend to consider unknown parameters to indicate certain behaviours of the interface [7]. This paper aims to pro-

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vide some help in their determination.

In order to discuss the interface microscopic behaviour, molecular dynamics (MD) simulations are used. As it is common in MD, the problem presented is simplified in pursuance of a more systematic study. In other words, the simulation of the particles on a liquid-gas interface is an approximation to what is found in the experiment.

In the paper, we examine the particles behaviour at the liquid-gas interface of argon atoms, which are described by a Lennard-Jones potential. Once the interface is formed and its creation process speeded, we introduce ions in the medium to achieve a first approximation to the experimental scenario. However, simulations of such massive systems show some limitations that we face in order to achieve our objective.

## II. SIMULATION PROCEDURE

To simulate the interface, we use MD computer simulations. This is a useful computational technique to obtain the dynamics of multicomponent systems by solving the time integral Newtons motion equations. Consequently, obtaining accurate trajectories of the particles in the system. Furthermore, MD simulations can offer some insights to experimental approaches and results [8], therefore, allowing us to offer a new perspective on macroscopic properties of experimental interest.

As the interfaces are highly dependent of the temperature, we use the Nosé-Hoover thermostat, which sets a heat bath to accurately describe a constant temperature system by removing or adding the necessary energy.

### A. Interaction potentials

One important requirement for a MD simulation is the definition of the interaction potentials between the system particles. In our case, we consider a Lennard-Jones pairs potential ( $V_{LJ}$ ) to describe the interaction of the argon atoms.

$$V_{LJ}(r) = 4\epsilon \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^6 \right] \quad (1)$$

The parameters values  $\epsilon$  and  $\sigma$  for the argon are found at Table.I. Despite the Lennard-Jones potential being not an exact representation of the interaction, its good accuracy and low computational cost makes it a good choice for the study of noble gas fluids.

Additionally the ions interact using the electrostatic potential (Eq.2)

$$V_e(r) = -\frac{1}{4\pi\epsilon_0\epsilon_r} \frac{q_1q_2}{r} \quad (2)$$

Being  $\epsilon_r$  the dielectric constant of the argon  $\epsilon_r = 1$

[9].

It is common in molecular simulations to set a cut off distance where the interactions stop being considered. Otherwise the amount of computation needed for all the particles would be too time consuming. In Lennard-Jones interaction pairs, it is considered that a cut off distance of  $2.5\sigma$  is enough [10], where  $\sigma$  is the effective size where  $V_{LJ} = 0$ . However, we increase it to get more accurate results by setting it to  $R_c = 15\text{\AA}$ .

### B. Simulation box

Another requirement is the setting of a simulation box. An increase in the simulation box size while maintaining the density of the system constant, will result in an increase of computational interactions. Therefore, a compromise must be set between accuracy and efficiency. However, by using periodic boundary conditions, bigger systems can be simulated. This is useful when recreating massive chambers. These allow us to consider a system far from the sides, that otherwise would affect the behaviour studied. Nevertheless, periodic conditions introduce more interfaces outside the box that may affect how the particles get in the system.

We expect the appearance of surface tension since we are in a two phases scenario, which will be formed while the system tries to reach its minimum energy by minimizing its interfacial area. Consequently, we design the simulation box to favour this area to be a slab instead of an sphere. The threshold for this conditions is shown in Eq.(3):

$$\frac{L}{H} \leq 3\sqrt{\frac{\pi}{2}} \quad (3)$$

Where L is the side of the box whereas H is the height. We set a simulation box of (x,y,z)=(40,40,200)  $\text{\AA}$ , where we expect each phase to fulfil the condition.

### C. Computational software

To calculate the equations of motion for all the particles in the medium, we use a MD calculator software called Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [11]. LAMMPS is an open source scientific program efficient at computing the time integration for the motion equations of many particles systems. It allows us to set a simulation box and fill it with particles, indicate the interaction potentials and initial conditions of the system.

To create our system we set a random initial distribution of the atoms. However, LAMMPS may overlap some particles, making the simulation fail and the results useless. In order to solve this problem, we minimize the energy of the particles for a few time steps before beginning the simulation.

The input parameters for all the simulations proposed in the paper are shown in Table I.

Parameter	Value	Units
Simulation box	(40,40,200)	Å
Particle mass $m$	39.984	$g/mol$
Temperature $T$	130	K
Well depth $\epsilon$	0.997	$KJ/mol$
Effective size $\sigma$	3.44	Å
Cut off distance $R_c$	15	Å
Time step $dt$	1	$fs$

TABLE I: General parameters used in all the simulations.

### III. LIQUID-GAS INTERFACE CONSTRUCTION

To simulate the liquid-gas interface, we make sure we are in a possible coexistence area, hence, we choose an initial density of  $\rho = 0.55g/cm^3$  at  $T = 130K$  [12]. Considering the volume of our simulation box, we calculate the number of atoms needed.

The duration of the simulations is 7000000 (7M) time steps. As stated before, we add the atoms in a random distribution and proceed to minimize the energy before starting the simulation. Once the minimization has been done, we capture the initial distribution, Fig.(1a). After the simulation, the system presents an interface, Fig.(1b). However, as we are working on periodic boundary conditions, the gas phase occupies the middle of the box while the liquid phase is halved between two simulation boxes. Hence, we observe two interfaces inside the box.

The density values are analysed so a more time efficient set of initial conditions can be applied. We average the density of the vertical profile during the last 5M time steps, where the phases start to be defined, Fig.(2). This allows us to have a sample big enough to avoid the typical fluctuations of microscopic systems.

We see the height of the interface to be approximately 40 Å. Additionally, both phases occupy similar sizes inside the box. From Fig.(2), we obtain the values of the liquid phase ( $\rho_l = 0.88g/cm^3$ ) and the gaseous phase ( $\rho_g = 0.18g/cm^3$ ).

To check the values veracity, we create another simulation box by splitting the original into halves (40,40,100). We proceed to create the corresponding amount of particles according to their densities in each region and start the simulation once the energy has been minimized. However, in this case we simulate for 5000 time steps. We display the final result, Fig.(1c), to see that the interface is correctly created. This method creates the interface fast enough so that we can consider the introduction of ions to an already existing interface.

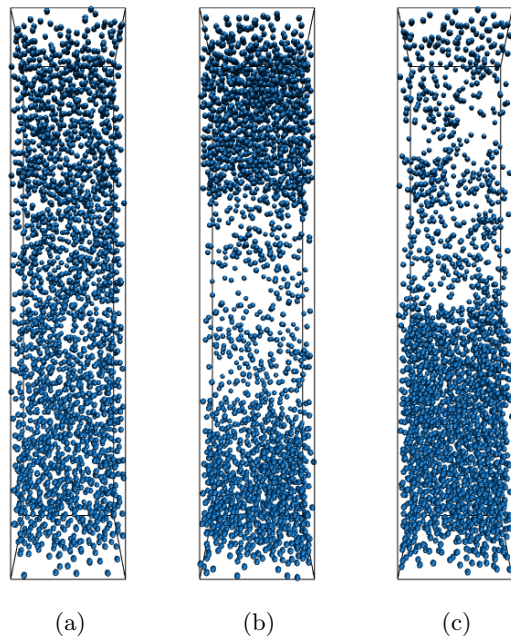


FIG. 1: Argon atoms distribution inside the simulation box. (a) shows the initial distribution after the minimization process. (b) shows the results in the final distribution and (c) shows the final distribution using the obtained densities.

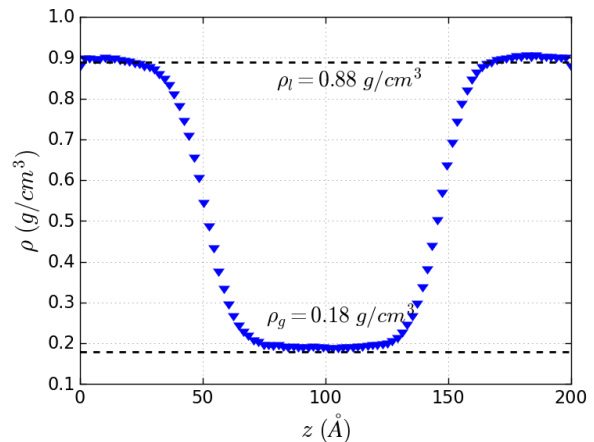


FIG. 2: Average densities for different horizontal slices of the chamber during the last 5M time steps.

### IV. IONS INJECTION AT THE INTERFACE

Once the initial conditions to form the interface have been obtained, we proceed with the study of the charged particles behaviour. It has been considered that the difference between the potential energies of the two phases favours the injection of ions into the liquid [7]. We begin the simulations setting a neutral charged system, i.e. the total charge inside the box is zero. This is a common procedure in MD simulations where there are periodic boundaries and the computa-

tions of the electric field only converges with net charge being zero. Nevertheless, LAMMPS has methods to deal with finite net charges different than zero but it is recommended to avoid it.

### A. Charged pairs

In massive LArTPCs, the creation of charge particles happens by the ionization of a neutral atom. Thus, after the collision, an electron-ion pair appears inside the chamber. We ignore the size difference between the electron and the ion to consider a simpler simulation where the ions are the charged particles being anions and cations. We set the same initial conditions obtained in the last section, Fig.(1c), and add 10 pairs of anions( $q_a = -1 eV$ ) and cations( $q_c = 1 eV$ ) to the system. Because we are adding charged particles already separated, we consider that the recoil energy has scattered the pair through the box. Therefore, a random distribution can be used to place the ions.

We add an electric field to the system. This field will apply a force to anions and cations equally, following the expression  $F = qE$ . We use the value of the uniform field considered for this type of detectors in the previous study of  $E = 1 kV/cm$  [7].

After the simulation, we observe that the ions have joined due to the attraction force. The field applied or thermal effects have not been able to keep them from recombine. Due to the  $V_{LJ}$ , the ions minimum distance between them is around  $3 \text{ \AA}$  ( $\sigma = 3.44 \text{ \AA}$ ). Therefore, we calculate the maximum value of the attraction field that a ion creates so we can compare its force with the one produced by the external field applied.

$$E_a(d = 3\text{\AA}) = \frac{1}{4\pi\epsilon_0\epsilon_r} \frac{|q|}{r^2} \approx 1.599 \text{ V/\AA} \quad (4)$$

Thus,  $E_a \gg E$ .

The experimental value of the applied field is negligible compared to the attraction field between the pairs. We check this by analysing the velocity evolutions on this system for different applied fields.

For  $E < E_a$ , the pair recombine and stops being affected by the field, as their total charge is zero. Otherwise, for  $E > E_a$ , the recombination is avoided and we see some oscillation, probably caused by the difference in mobility for each phase. Nevertheless, due to the magnitude of the field, these velocities increase the system energy significantly, hence, we are not longer at  $T = 130K$  and the densities of the coexistence differ from the ones used. This causes the formation of bubbles inside the box and the destruction of the interface itself. As a result, this is not a suitable simulation for our study.

We can justify these results by studying the Bjerrum length of the system. The Bjerrum length indicates the separation between two charged particles from which the electric interaction is bigger than the thermal effects. Once the electrostatic interaction is comparable

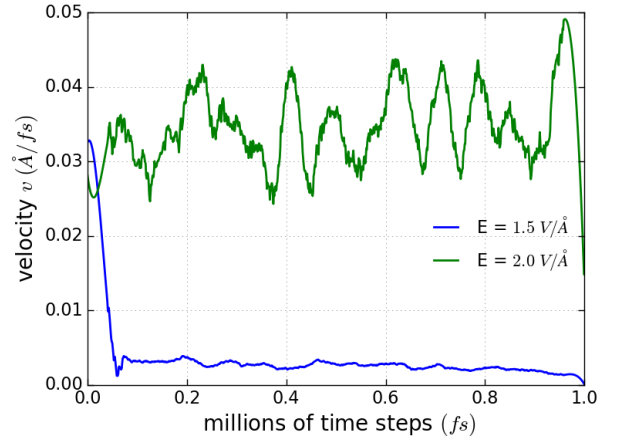


FIG. 3: Velocity evolution for an anion inside the chamber. A Savitzky–Golay filter is applied to smooth the fluctuations of the values at the representation.

to  $\kappa_B T$ , the system is driven by the attraction.

If we calculate the Bjerrum length for our study, we obtain

$$\lambda_B = \frac{1}{4\pi\epsilon} \frac{q^2}{\kappa_B T} \approx 3 \cdot 10^{-7} m = 3 \cdot 10^3 \text{ \AA} \quad (5)$$

In a  $L = 40\text{\AA}$  box, there will always be more attraction between the particles than thermal effects. We can not avoid the recombination of the ions, since the size of our simulation box, is too small to be compared to the experimental chambers. As stated before, the size of the simulation box is tied to the computational cost needed. Therefore, we must reassess the proposed methodology for the simulation.

### B. Positive charges

As stated in the previous study [7], the electrons drift velocity is bigger than the ions one ( $\nu_e \gg \nu_i$ ). Therefore the electrons remaining time inside the TPC is negligible in comparison with the ions times. This creates a difference in charged density inside the detector ( $\rho_i \gg \rho_e$ ). Even though this is a problem for the uniformity of the field, it allows us to assume that the electrons are absorbed fast enough to not consider them. Hence, we assume the case with only cations a suitable one for our simulation, as they are the main remaining charge at the interface.

In order to achieve an initial approximation to the experiment, we do not introduce an external field. As there is a total charge different than zero, while having an infinity periodic boundary, the system would experience a net force that would accelerate the particles. As a result, our initial consideration for the creation of the interface would not work as the temperature would rise and change the coexistence densities. Additionally, to achieve a big statistic sample, we perform with 34 simulations with different random initial positions for

the cations over a total sum of 129M time steps.

Once the simulation is completed, we see the cations being in a stable position. This leads to the conclusion that the repulsion potential between ions is the main interaction for the charges. As the previous case, it seems to be the expected behaviour when the Bjerrum length is bigger than the sides of the box.

In order to confront this problem, we adapt the simulations by supposing an hypothetical case where the charge value of the cations is low enough to make the Bjerrum length smaller than the box size. A length of  $\lambda_B = 6\text{\AA}$  is a good value to fulfil  $\sigma < \lambda_b < L$ . Thus, we set the charges to be  $q' = 0.045 eV$ . Additionally, as low charges apply less force to the system, we add again the experimental electric field,  $E = 1 \text{ KV/cm}$ . The final distribution of densities is shown in Fig.(4).

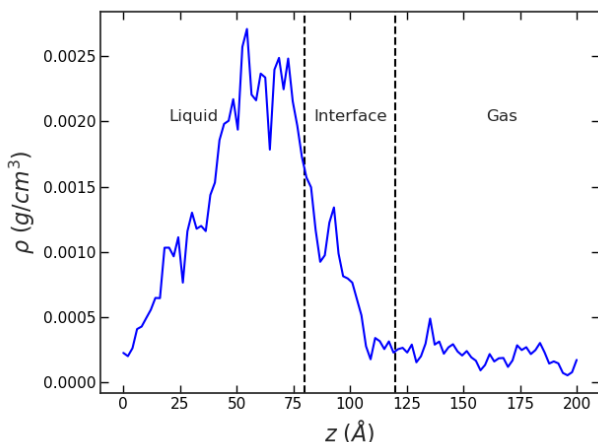


FIG. 4: Ions density considering smaller charges. The average value for each slab during 4M time steps is displayed.

We observe that when considering this scheme of lengths ( $\sigma < \lambda_B < L$ ), and therefore, smaller electric interactions, the cations tend to cross the interface to the liquid, thus, confirming that the positive charges are flavoured to pass from the gas to the liquid.

## V. CONCLUSIONS

We have successfully achieved the formation of an interface between liquid and argon gas through the use of molecular dynamics. However, the limitations in computational power have forced us to work in a simulation box too small to recreate the experimental conditions. We have obtained the distribution of cations at the interface of a Lennard-Jones fluid characterised as an argon medium. Nevertheless, the more descriptive distribution has been obtained in an hypothetical case where the value of the charges is smaller than the real one. Therefore, the results must be taken as a first approximation to the experimental description, even though they are a starting scenario for future studies such as the cations electric field dependency or the efficiency to pass positive ions from gas to liquid.

Once we have obtained some insight on the behaviour of argon atoms at the interface, future works could achieve an approximation to the macroscopic parameters related to the interface for different time-projection chambers.

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