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Switching Between the NVT and NpT Ensembles Using the Reweighting and Reconstruction Scheme

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Abstract

Recently, we have developed several techniques in order to accelerate Monte Carlo (MC) molecular simulations. For that purpose, two strategies were followed. In the first, new algorithms were proposed as a set of early rejection schemes performing faster than the conventional algorithm while preserving the accuracy of the method. On the other hand, a reweighting and reconstruction scheme was introduced that is capable of retrieving primary quantities and second derivative properties at several thermodynamic conditions from a single MC Markov chain. The latter scheme, was first developed to extrapolate quantities in NVT ensemble for structureless Lennard-Jones particles. However, it is evident that for most real life applications the NpT ensemble is more convenient, as pressure and temperature are usually known. Therefore, in this paper we present an extension to the reweighting and reconstruction method to solve NpT problems utilizing the same Markov chains generated by the NVT ensemble simulations. Eventually, the new approach allows elegant switching between the two ensembles for several quantities at a wide range of neighboring thermodynamic conditions.

Keywords: Reweighting and reconstruction, Monte Carlo Markov chain, NVT and NpT ensembles

1 Introduction

Monte Carlo (MC) molecular simulations are based on the principles of statistical thermodynamics that provide a link between microscopic properties of matter and its bulk ones [1–3]. In other words, by MC molecular simulations we attempt to study matter in a detailed fashion by describing it at the molecular level utilizing the fundamental concepts of statistical thermodynamics. As a consequence of this detailed insight, molecular simulations are, in general, computationally much more expensive than classical models that are established based on the continuum assumptions. These detailed information, which are inquired from the microscopic

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level description, are collected in the so-called MC Markov chains. For instance, ensemble averages such as system's energy or pressure can be estimated by simply averaging relevant quantities from these chains. In addition, fluctuations around average could be used to estimate second derivative properties such as heat capacity and fluid compressibility [4].

To overcome the burden of the high computational cost, one could accelerate MC molecular simulations by extracting more information from these Markov chains. This might be possible, if the Markov chain generated by a single simulation at certain thermodynamic conditions could predict quantities at these prescribed conditions and beyond. Thus, merely a single simulation can replace multiple ones and save lots of computational time. This could be thought of as an alternative path to accelerate MC simulations instead of trying to modify the Metropolis algorithm itself [5–7].

For that purpose, researchers have developed several methods in order to retrieve more macroscopic information from the collected Markov chains. Typical examples of such techniques are histogram reweighting [8–10] and thermodynamic scaling [11–16]. In the first, collective distributions are properly re-weighted with some tunable factors in order to estimate ensemble averages at different conditions than the simulated ones. On the other hand, thermodynamic scaling refers to group of approaches developed for various ensembles based on the umbrella sampling concept [17, 18].

In this context, a novel, simple and self-consistent reweighting and reconstruction scheme has been introduced [19,20]. In contrast to the reported methods in literature, where unphysical non-Boltzmann distributions are utilized in addition to some tunable parameters, the proposed extrapolation schemes retain physically meaningful distributions and do not contain any parameters to be tuned. Nonetheless, the introduced method has been, so far, accommodated for systems of structureless identical Lennard-Jones (LJ) particles in canonical or NVT ensemble. However, in reality pressure and temperature are controlled, as they are usually tangible and measurable. In this article, we extend the reweighting and reconstruction scheme to encounter NpT systems (ensembles) using the same set of Markov chains created by NVT ensemble simulations.

In the following sections, the details of the simulation methods with the corresponding set of equations utilized to conduct this study are first described. Secondly, the generated results are reported and thoroughly discussed. Moreover, the algorithms and methods used in this work are validated by comparing the simulation results obtained versus relevant experimental data. Finally, the article closes with set of conclusions summarizing the objectives of this study.

2 Simulation Methods

The reweighting and reconstruction method has been originally designed for the canonical ensemble, which is often known as the NVT ensemble. In the NVT ensemble, the number of simulated particles (N), simulation cell volume (V) and temperature (T) are all kept fixed throughout the simulation. However, and as stated earlier, in real life experiments pressure (p) and temperature are usually kept constant. Motivated by this fact, scholars introduced the isothermal-isobaric ensemble or the NpT ensemble at which the pressure is imposed instead of the volume during the simulation. In this section, the original reweighting and reconstruction scheme developed for the NVT ensemble is at first described. Later, the tools to switch from the NVT ensemble to the NpT one are explained.

2.1 Primary Quantities

Primary quantities are obtained directly from the ensemble averages of the generated Markov chains in MC molecular simulations. In this work, we focus on two ensemble averages: the system's potential energy per particle ($\overline{E}$) and the system's pressure (p). To easily express and integrate these two terms in the reweighting and reconstruction scheme, two other generalized configurational quantities U_k and V_k that are functions of the distances separating the simulated particles are defined. For convenience and as all the simulated particles are assumed identical, these quantities can be normalized with respect to ε and σ. For instance, the normalized potential energy of the k^{th} configuration (U_k^*) is computed by (1); where, U_k is normalized by ε and distance r_{ij} is normalized by σ ($r_{ij}^* = r_{ij}/\sigma$). Furthermore, r_{ij}^* is divided by the normalized simulation box length ($L^* = L/\sigma$) such that $s_{ij}^* = r_{ij}^*/L^*$. Clearly, s_{ij}^* represents the normalized distance separating the i^{th} and j^{th} particles relative to L^*. Therefore, s_{ij}^* can only attain values between 0 and 1. Similar normalization scenario is applied to the other defined quantity in (2).

$$U_k^* = \frac{U_k}{\varepsilon} = 4\sum_{i<j}\left[\left(\frac{1}{L^* s_{ij}^*}\right)^{12} - \left(\frac{1}{L^* s_{ij}^*}\right)^{6}\right] \tag{1}$$

$$V_k^* = \frac{V_k}{\varepsilon} = 8\sum_{i<j}\left[2\left(\frac{1}{L^* s_{ij}^*}\right)^{12} - \left(\frac{1}{L^* s_{ij}^*}\right)^{6}\right] \tag{2}$$

Using the canonical averages of the defined configurational quantities above, the following equations can be derived to compute $\overline{E}^*$ and p^*, respectively

$$\overline{E}^* = \frac{E}{N\varepsilon} = \frac{1}{N}\left\langle U^* \right\rangle_{can} + E_{tail}^* \tag{3}$$

$$p^* = \frac{p\sigma^3}{\varepsilon} = \frac{\rho^*}{\beta^*} + \frac{\rho^*}{N}\left\langle V^* \right\rangle_{can} + p_{tail}^* \tag{4}$$

In these equations (1 - 4), ε and σ are model parameters related to the nature of the simulated particle, N is the total number of particles, β^* is the normalized Boltzmann's factor and equivalent to the reciprocal of the normalized temperature ($T^* = T/(\varepsilon/k_B)$), k_B is the Boltzmann's constant and ρ^* is the normalized number density ($\rho^* = \rho\sigma^3$). Whereas, $\langle X \rangle_{can}$ stands for the canonical average of property X over all the sampled configurations and subscript "*tail*" corresponds to the tail correction functions used to count for truncated interactions beyond the imposed cutoff radius ($r_c^* = 0.49L^* = s_c^* L^*$). These tail correction functions [4] can be computed using the following equations:

$$E_{tail}^* = \frac{8}{3}\pi\rho^*\left[\frac{1}{3}\left(\frac{1}{L^* s_c^*}\right)^{9} - \left(\frac{1}{L^* s_c^*}\right)^{3}\right] \tag{5}$$

$$p_{tail}^* = \frac{16}{3}\pi\rho^{*2}\left[\frac{2}{3}\left(\frac{1}{L^* s_c^*}\right)^{9} - \left(\frac{1}{L^* s_c^*}\right)^{3}\right] \tag{6}$$

2.2 Second Derivative Quantities

In addition to ensemble averages, the fluctuations of the sampled Markov chains can be used to estimate second derivative properties. In this paper, the isochoric heat capacity (C_v) and the isothermal compressibility (c_T) are considered. Similar to the previous part, one additional configurational quantity W_k is defined.

$$W_k^* = \frac{W_k}{\varepsilon} = 8\sum_{i<j}\left[10\left(\frac{1}{L^* s_{ij}^*}\right)^{12} - 3\left(\frac{1}{L^* s_{ij}^*}\right)^{6}\right] \tag{7}$$

Using the three canonical averages and the fluctuations of these defined quantities both properties under interest can be computed as follows [13]:

$${C_v}^* = \frac{C_v}{k_B N} = \frac{1}{N}{\beta^*}^2\left(\left\langle {U^*}^2\right\rangle_{can} - \left\langle U^*\right\rangle_{can}^2\right) \tag{8}$$

$${c_T}^{*-1} = \frac{{c_T}^{-1}\sigma^3}{\varepsilon} = \frac{\rho^*}{\beta^*} + \rho^*\left\langle W^*\right\rangle_{can} - 2\beta^*{\rho^*}^2\left(\left\langle {V^*}^2\right\rangle_{can} - \left\langle V^*\right\rangle_{can}^2\right) + c_{tail}^* \tag{9}$$

$$c_{tail}^* = \frac{16}{3}\pi{\rho^*}^2\left[\frac{10}{9}\left(\frac{1}{L^* s_c^*}\right)^{9} - 3\left(\frac{1}{L^* s_c^*}\right)^{3}\right] \tag{10}$$

2.3 Reweighting and Reconstruction Workflow

In the NVT ensemble, the general case for applying the reweighting and reconstruction scheme is to predict an ensemble average $\langle A^*\rangle_{can}$ at certain temperature and density that are different from the ones used to generate the MC Markov chain. In other words, the general case will be to extrapolate ensemble averages along paths of changing temperature and density at same time. This is possible by first reconstructing and then reweighting the original sampled Markov chain using the following relation

$$\left\langle A^*\left(\beta_n^*, L_n^*\right)\right\rangle_{can} = \frac{\sum_{k=1}^{M}\exp\left(-w_k\right)A_k^*\left(\beta_o^*, L_n^*\right)}{\sum_{k=1}^{M}\exp\left(-w_k\right)} \tag{11}$$

$$w_k = \beta_n^* U_k^*\left(\beta_o^*, L_n^*\right) - \beta_o^* U_k^*\left(\beta_o^*, L_o^*\right) + N\left[\beta_n^* E_{tail}^*\left(L_n^*\right) - \beta_o^* E_{tail}^*\left(L_o^*\right)\right] \tag{12}$$

In equations (11) and (12), reweighting factors ($\exp(-w_k)$) are related to the temperature and energy difference between the reference case and the targeted one. The quantity, A, could be replaced by any of the quantities U, U^2, V, V^2 or W with "o" and "n" subscripts representing the original thermodynamic conditions and the new ones at where the targeted point exists, respectively. Meanwhile, M is the number of sampled configurations that are forming the collected Markov chain out of the total created configurations.

2.4 Switching Scheme

Before describing the switching mechanism from the NVT to the NpT ensemble, the application of the classical reweighting and reconstruction scheme in predicting p^* is first explained. Starting from equation (4), the following relation shows how p^* at a new temperature (β_n^*)

and density (ρ_n^*) can be estimated using the information from a Markov chain generated at neighboring β_o^* and ρ_o^*.

$$p^*\left(\beta_n^*, L_n^*\right) = \frac{\rho_n^*}{\beta_n^*} + \frac{\rho_n^*}{N}\left\langle V^*\left(\beta_n^*, L_n^*\right)\right\rangle_{can} + p_{tail}^*\left(L_n^*\right) \tag{13}$$

In the equation above (13), the ideal part and the tail correction function are directly modified to adhere with the new prescribed conditions (β_n^* and ρ_n^*=$N/{L_n^*}^3$). However, the residual part $\langle V^*\left(\beta_n^*, L_n^*\right)\rangle_{can}$ is computed as explained in equation (11); such that, the reweighting factors are evaluated as exactly shown in equation (12).

$$\left\langle V^*\left(\beta_n^*, L_n^*\right)\right\rangle_{can} = \frac{\sum_{k=1}^{M}\exp\left(-w_k\right)V_k^*\left(\beta_o^*, L_n^*\right)}{\sum_{k=1}^{M}\exp\left(-w_k\right)} \tag{14}$$

So far the problem is forward, as the only targeted unknown is p^*. Nonetheless, another way to visualize this problem is by assuming that the new pressure ($p^*\left(\beta_n^*, L_n^*\right)$) and β_n^* are given and to look for their corresponding ρ_n^*. In standard MC simulations, such new formulation requires designing a new experimental setup which, as mentioned earlier, is called the NpT ensemble. In other words, the numerical experiment has to be rerun again with the same expensive computational cost. On the other hand, this work shows that such a switch between the NVT and NpT ensembles is fairly simple when using the reweighting and reconstruction scheme. Basically, the same equation in (13) is used but this time to search for L_n^* value that successfully retains the prescribed pressure $p^*\left(\beta_n^*, L_n^*\right)$. In the "Results and Discussion" section the proposed scheme will be further elaborated with more details.

3 Results and Discussion

The extensive analysis conducted to test and validate the proposed approach, is based on a set of 750 Markov chains generated by MC molecular simulations in NVT ensemble. This set spans the range of $0.05 \leq \beta^* \leq 0.75$ and $0.02 \leq \rho^* \leq 1$. The β^* range is chosen such that the system is always at a temperature higher than the critical temperature to avoid entering the two-phase envelope. On the other hand, ρ^* values are decided such that cases from sparse to highly dense systems are considered. In generating the 750 chains, a system of 256 uniformly distributed structureless LJ particles in a cubic cell with dimensions fulfilling the prescribed system's ρ^* is initially used. Starting from these initial configurations, a sequence of successive configurations is created by introducing elemental changes to the system, known as MC trials. For this particular case, particle displacement was the only possible MC trial. Therefore, at each MC step a particle is randomly chosen and displaced. For each run, a total of 61,440,000 MC trials was performed to generate the same number of configurations. The first 10 % of these configurations were discarded to account for equilibration. Out of the 90% left, only one configuration was sampled every 1024 steps. As a result, each of these 750 Markov chains is made of 54,000 sampled configurations. As a matter of fact, for the reweighting and reconstruction method only the sum of the short range ($1/s^{12}$) and long range ($1/s^6$) interactions need to be separately collected in the Markov chains.

This section of the paper is divided into three parts. In the first one, the results corresponding to solving the NpT problem, at which ρ^* is the main output, are discussed. In the second part, the other three quantities of interest are estimated by switching back to the regular reweighting and reconstruction scheme, as the densities corresponding to the new pressures are

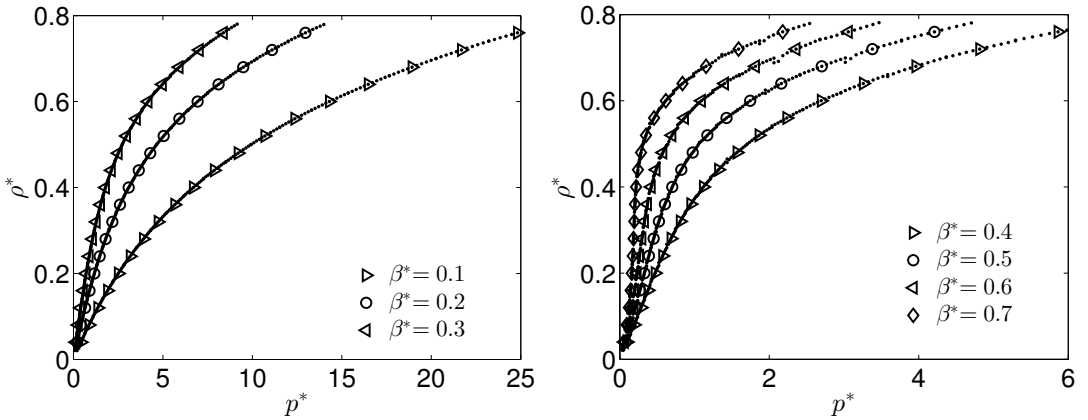

Figure 1: Extrapolated ρ^* $(\cdot)$ along seven different isotherms ($\beta^* = 0.1$, 0.2 ... and 0.7). Markov chains used as source points for extrapolation are plotted as symbols. Each source is used to extrapolate the adjacent five dots from each side. The plots are split into two for clarity.

now known. Finally, comparing the extrapolation results against methane gas experimental data validates all the algorithms and techniques used in this work.

3.1 Solving the NpT Problem

After conducting the necessary MC simulations in NVT ensemble, a data bank of MC Markov chains is available. In order to solve the NpT problem, p^* is first computed for each chain corresponding to specific β^* and ρ^* using equation (4). Therefore, for every chain in the generated data bank the three thermodynamic states $\beta^* = 1/T^*$, ρ^* and p^* are known. Assuming that we want to estimate ρ^* at a given p_x^* and β_x^*, the first step is to determine the suitable MC Markov chain from the generated set of chains as the source of extrapolation. Considering the case of extrapolation along isothermal paths, the chain with β^* equal to β_x^* and p^* closest to the targeted p_x^* is chosen as the source point of extrapolation.

In Figure 1, seven isotherms are reproduced. Along each isotherm only 19 Markov chains were used as source points for extrapolation. From each source point, five adjacent ρ^* values are predicted from each side at different p^* values such that the extrapolations from two consecutive source points meet at the middle. In fact, these predictions are made using equation (13) while solving for L_n^* as both $p^*\,(\beta_n^*, L_n^*)$ or p_x^* and β_n^* or β_x^* at extrapolation points are known. According to Figure 1, all the isotherms are well reproduced as the transitions from one extrapolation set to another are, in most occasions, smooth. However, in agreement with results reported in our previous papers [19, 20], the ranges of extrapolation are dependent on the thermodynamic states of the source points of extrapolation. It is observed that at low temperatures (high β^*) the ranges get narrower as the system approaches the critical temperature.

3.2 Switching back to the NVT Ensemble

As the ρ_x^* values associated with different pressures (p_x^*) of interest are all known, it is possible now to shift back to the regular forward reweighting and reconstruction scheme to solve for the other three properties considered using equations (3), (8) and (9). In this regard, three more figures are generated reproducing isotherms of $-\overline{E}^*$ (Figure 2), C_v^* (Figure 3) and c_T^* (Figure

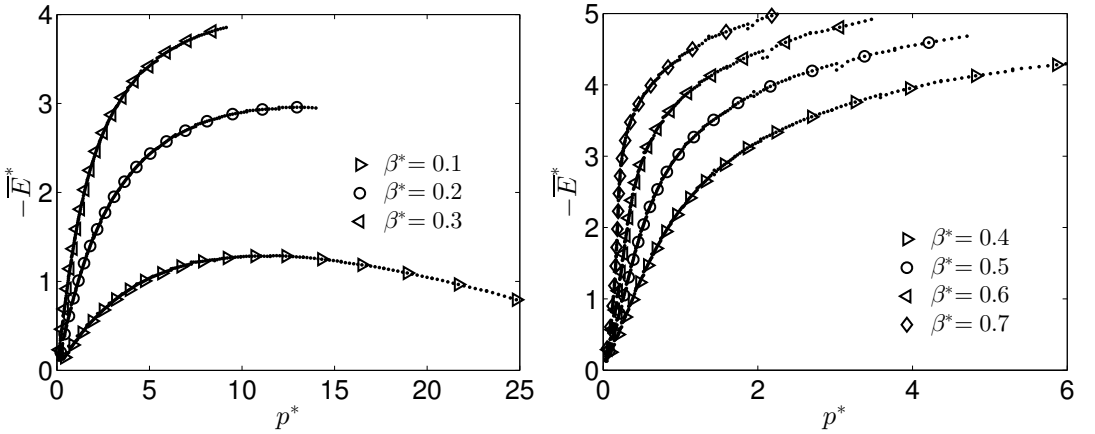

Figure 2: Same as Figure 1 but for extrapolating $-\overline{E}^*$.

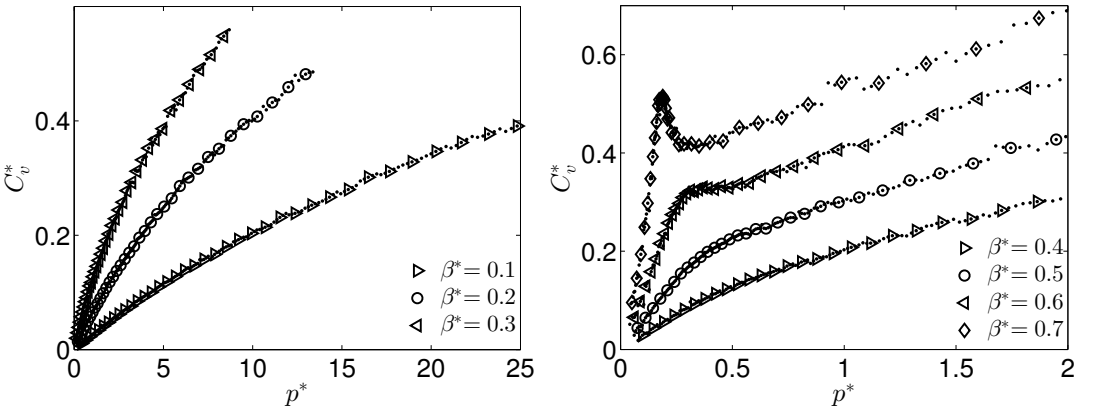

Figure 3: Same as Figure 1 but for extrapolating C_v^*.

4). Again here, the plots were split into two sets for clarity. Similar to ρ^*, extrapolation ranges diminish as source points approach critical temperature. In addition, it is evident that for second derivative quantities narrower ranges of extrapolation are expected when compared to primary quantities. To sum up, the MC Markov chains generated from NVT ensemble simulations are used to solve for density at a given pressure and temperature. To solve the inverse problem, the MATLAB built-in non-linear solver "fsolve" was used. Whenever densities are determined, the rest of the parameters are evaluated using the forward scheme.

3.3 Extrapolation Along Paths of Changing β and p

To take the method a step further, in fact, the extrapolation process does not have to solely be along isotherms. It is possible to extrapolate from a certain source point to other neighboring points at both different temperature and pressure conditions. As an example (Figure 5), source points ($\circ$) along $\beta^* = 0.3$ were used to extrapolate points ($\cdot$) at different pressures along two different isotherms $\beta^* = 0.25$ ($\triangleright$) and 0.35 ($\diamond$). The symbols ($\triangleright$ and $\diamond$) are only plotted to show

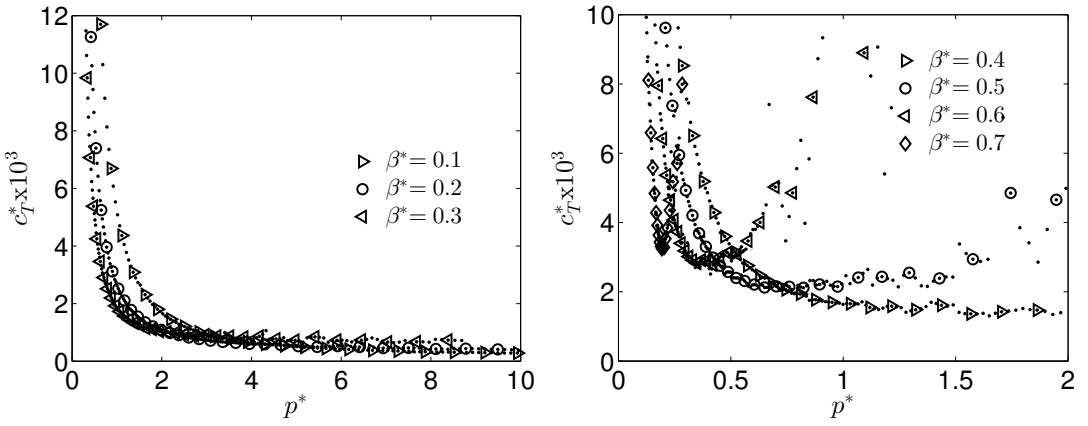

Figure 4: Same as Figure 1 but for extrapolating c_T^*.

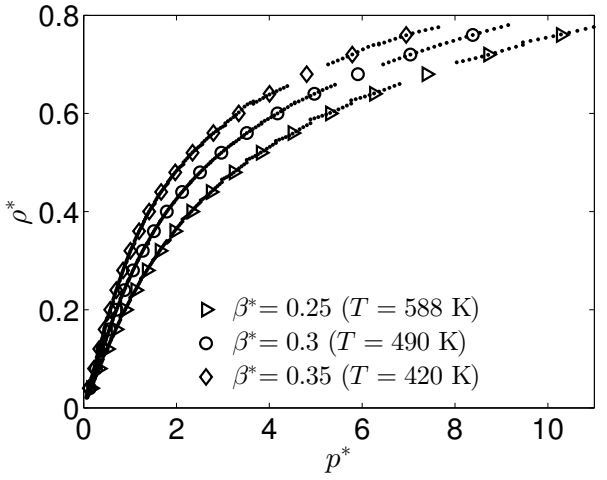

Figure 5: ρ^* ($\cdot$) values are extrapolated using Markov chains along $\beta^* = 0.3$ ($\circ$) as source points. Each source point is used to extrapolate along the same isotherm and the other two plotted ones. To better visualize the extrapolation area along different temperature and pressure conditions, the points extrapolated by one of the source points are removed.

how well the extrapolation was. In addition, the extrapolated points by one of the source points were removed to help visualize the extrapolation area covered by each source point.

Moreover and in order to get a clear idea about the extrapolation limits of the proposed scheme, the normalised quantities (β^*) are transformed into temperatures (T) with absolute units. By referring to Figure 5, the source points along $T = 490$ K isotherm ($\circ$) were successful in reproducing the other two isotherms with more than 70 K temperature difference from both sides. For more details about extrapolation limits, the two earlier papers are recommended [19, 20].

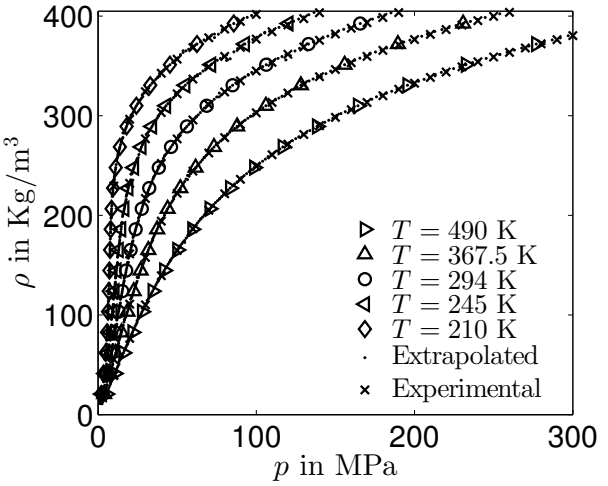

Figure 6: Comparison between ρ experimental values ($\times$) against simulated (Symbols) and extrapolated ($\cdot$) ones. Only five of the previous isotherms are reported as for the other two, for the best of our knowledge, no experimental data exists.

3.4 Validation

Finally the algorithms and extrapolation schemes used in this paper are all validated by comparing the data from direct simulations and extrapolated ones with respect to methane experimental data [21]. In specific, density values (Figure 6) are used in comparison. Moreover, and for better comparison, quantities in absolute units are reported instead of the normalized ones. Noting that, for converting quantities into real units $\varepsilon/k_B = 147$ K and $\sigma = 3.722$ Å were used [20].

4 Conclusion

In conclusion, motivated by the importance of the NpT ensemble in describing many of real life processes we have introduced a self-consistent scheme that allows switching between the NVT and NpT ensembles using the reweighting and reconstruction method. The proposed scheme extends the capabilities of the tremendously fast reweighting and reconstruction method by handling different types of systems depending on the imposed parameters. Results have shown that the range of extrapolation limits behaves similarly regardless of the switch between the two ensembles. In agreement with previous studies, primary quantities (ρ and $\overline{E}$) can be extrapolated for wider ranges when compared to second derivative ones (C_v and c_T). Meanwhile, the work is in progress to extend the reweighting and reconstruction method for other ensembles in order to widen the investigation scope. In addition, benefiting from this scheme by coupling it with flow simulators is under consideration.

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