Feasibility Study of Mg Storage in Bilayer Silicene Anode Via Application of External Electric Field

Sumaiyatul Ahsan[1], M. F. N. Taufique[2], Hasan Al Jame[1], Saugata Sarker[1], Abrar Rauf[1], Sadiq Shahriyar Nishat[3], Md Tohidul Islam[4], Azmain Faek Islam[5], Md. Rafsun Jani[1], Md. Shafiqul Islam[1], Kazi Md. Shorowordi[1], Saquib Ahmed[6*]

[1] Department of Materials and Metallurgical Engineering (MME), Bangladesh University of Engineering and Technology (BUET), East Campus, Dhaka-1000, Bangladesh
[2] Pacific Northwest National Laboratory, Richland, WA 99354, USA
[3]Department of Materials Science and Engineering (MSE), Rensselaer Polytechnic Institute, 110 8th street, Troy, NY 12180, USA
[4]Department of Materials Design and Innovation, University at Buffalo, Buffalo, NY 14260, USA
[5]School of Mechanical and Materials Engineering, Washington State University Pullman, Washington 99163, United States
[6]Department of Mechanical Engineering Technology, SUNY – Buffalo State, 1300 Elmwood Avenue, Buffalo, NY 14222

*Corresponding author
Email: ahmedsm@buffalostate.edu, Phone: 1.716.878.6006 Fax: 1.716.878.3033

1. Themostatting

The flying ice cube [1] is a molecular dynamics simulation artifact in which the use of velocity rescaling thermostats sometimes causes the violation of the equipartition theorem, according to which energy is shared equally among all of its different parts in thermal equilibrium. In flying ice cube artifact kinetic energy is drained from some degree of freedoms and fed into others resulting into zero-frequency motions such as overall translation and rotation of system and a very damped internal motion (almost as if an ice cube or any other rigid body is flying through space). This is most notably seen in simulations of particles in vacuum. As can be noticed from our experimental setup, we have added vacuum in all three directions of our system, hence we must account for this unnatural phenomenon.

As is well known, the flying ice cube artifact arises from faulty rescaling of the velocities that doesn't account for canonical ensemble. In simple velocity rescaling, for thermal equilibration, velocities of all particles at the end of every timestep is rescaled by a factor λ to achieve a target instantaneous temperature , T_{target}:

$$\lambda = (\frac{K_{target}}{K})^{\frac{1}{2}}$$

$$K_{target} = \frac{1}{2} \times N_{DOF} k_B T_{target}$$

Here, a non-canonical distribution of kinetic energies is used as a means to rescale energy.

The canonical sampling through velocity rescaling (CSVR) thermostat developed by Bussi–Donadio–Parrinello [2] rescales the velocity of particles at the end of each timestep by a factor λ so that the kinetic energy exhibits the distribution of the canonical ensemble.

$$\lambda = (\frac{K_{target}}{K})^{\frac{1}{2}}$$

$$P(K_{target}) \propto K_{target}^{(N_{DOF}/2 - 1)} e^{-\beta K_{target}}$$

Here K_{target} is drawn from probability density function instead of microcanonical ensemble independent variable T_{target}. Hence it's experimentally proven to be least likely to show the artifact. [3]

1.1 Nose Hoover Thermostat:

1.1.1 MSD in x Direction:

1.1.1.1 Smooth

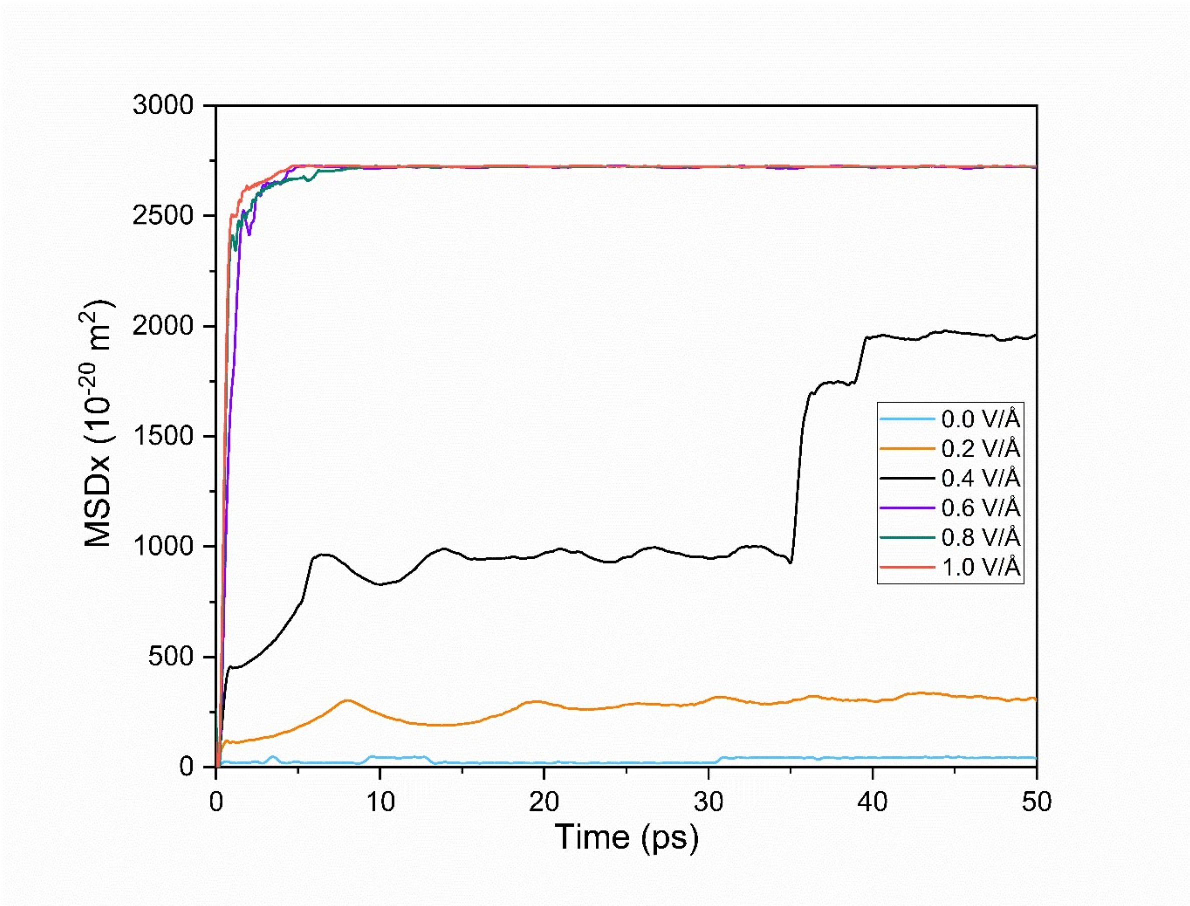

Figure S1 Smoothed curve: MSD behaviour in x direction vs time

1.1.1.2 Unsmooth

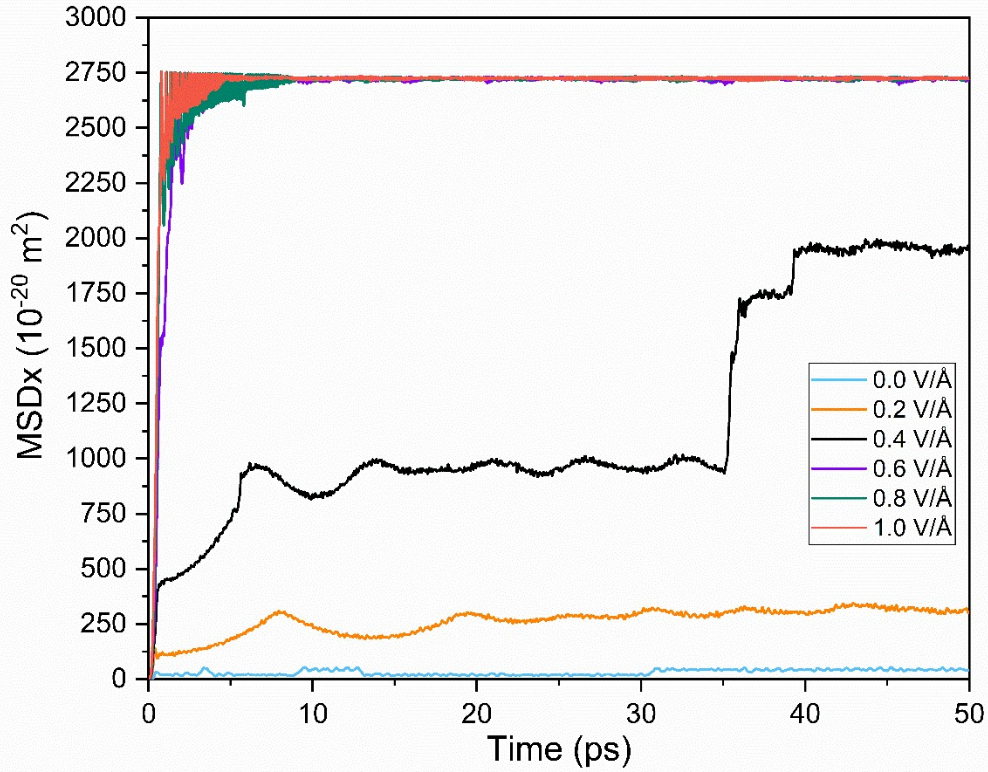

Figure S2 Unsmoothed curve: MSD behaviour in x direction vs time

1.1.2 MSD in y Direction:

1.1.2.1 Smooth

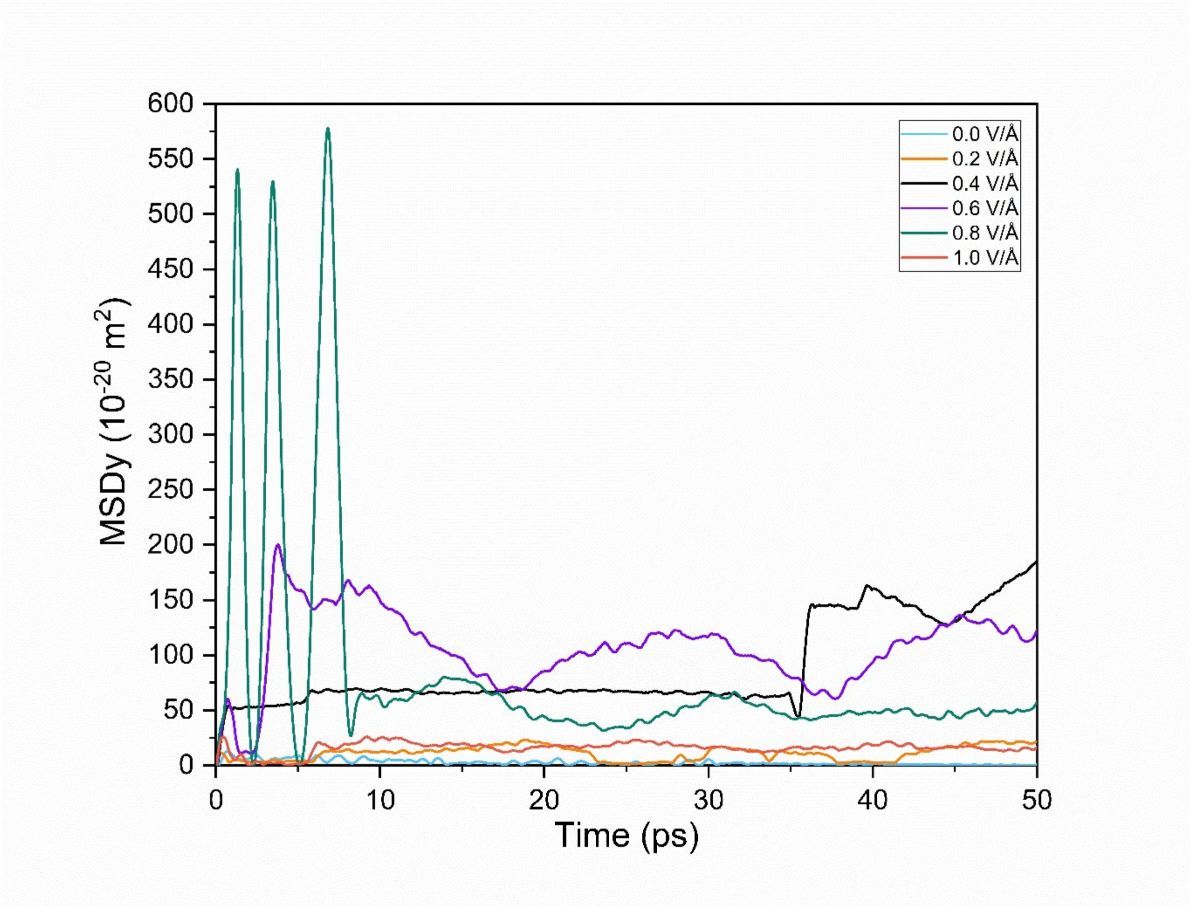

Figure S3 Smoothed curve: MSD behaviour in y direction vs time, demonstrating randomness.

1.1.2.2 Unsmooth

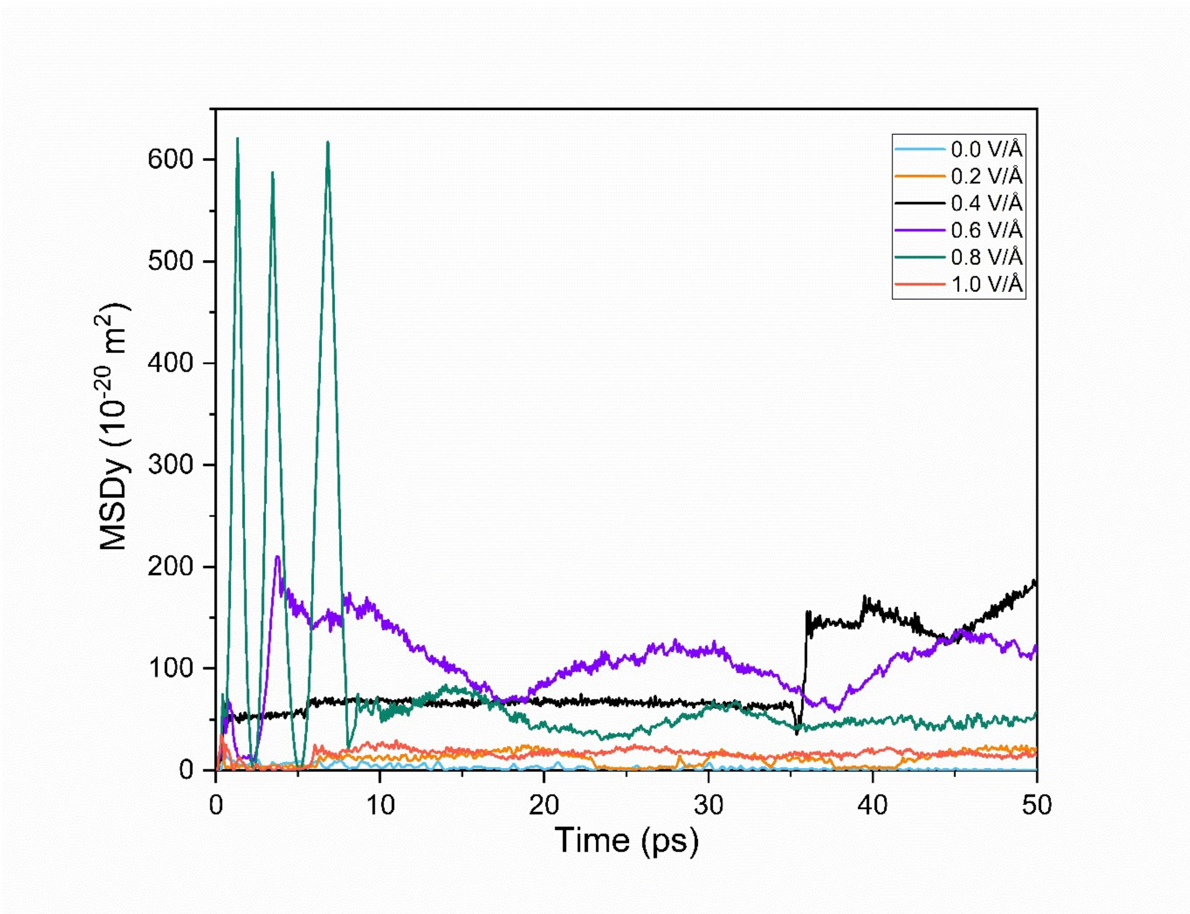

Figure S4 Unsmoothed curve: MSD behaviour in y direction vs time, demonstrating randomness.

1.1.3 MSD in z Direction:

1.1.3.1 Smooth

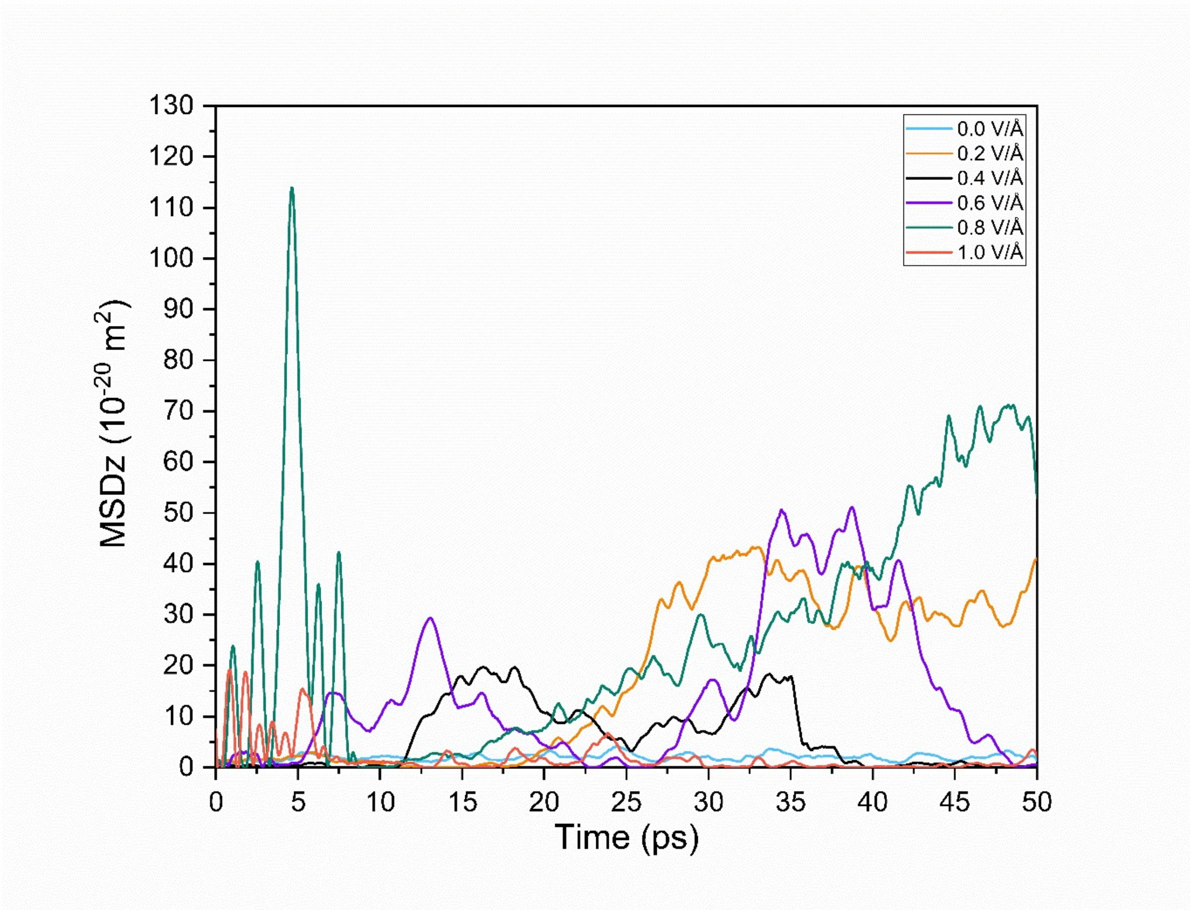

Figure S5 Smoothed curve: MSD behaviour in z direction vs time, demonstrating randomness.

1.1.3.2 Unsmooth

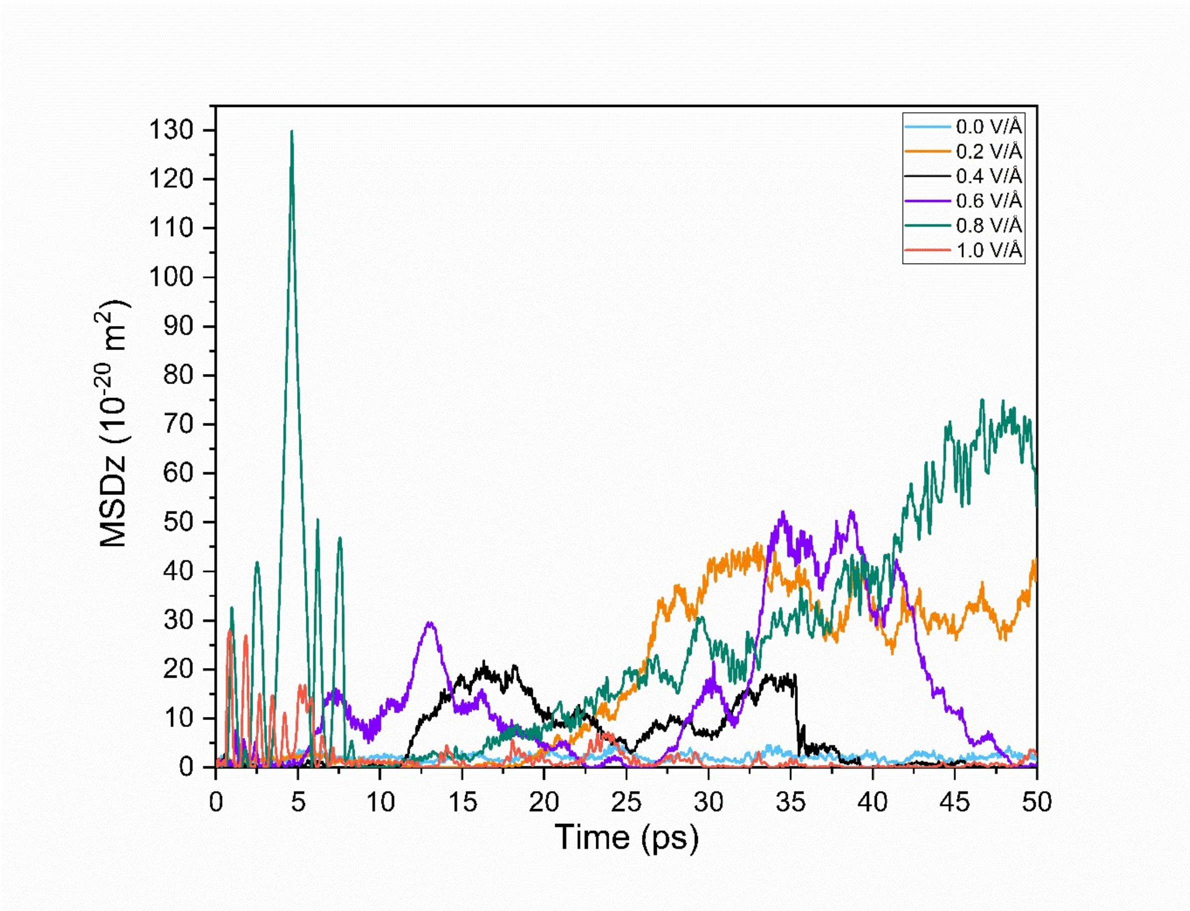

Figure S6 Unsmoothed curve: MSD behaviour in z direction vs time, demonstrating randomness.

1.1.4 Total MSD

1.1.4.1 Smooth

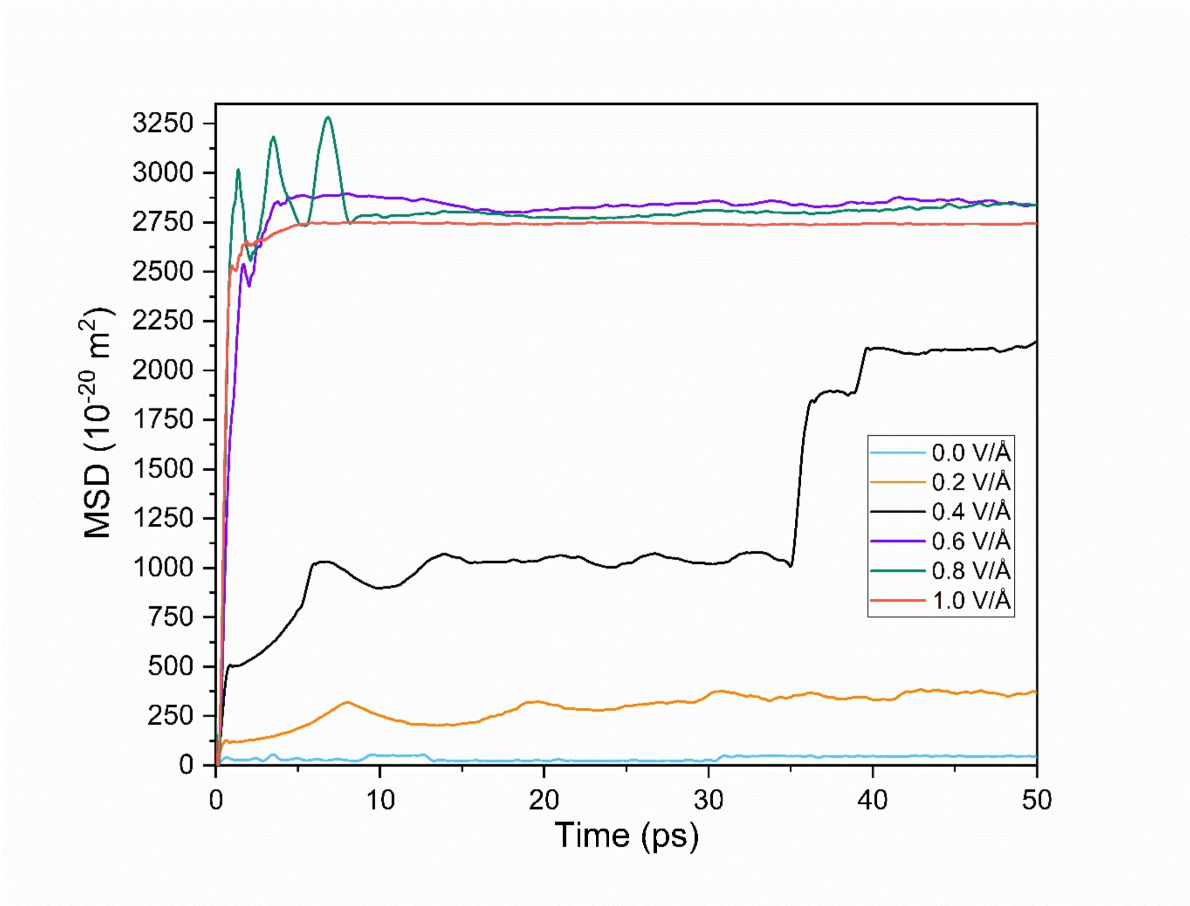

Figure S7 Smoothed curve: Total MSD vs time

1.1.4.2 Unsmooth

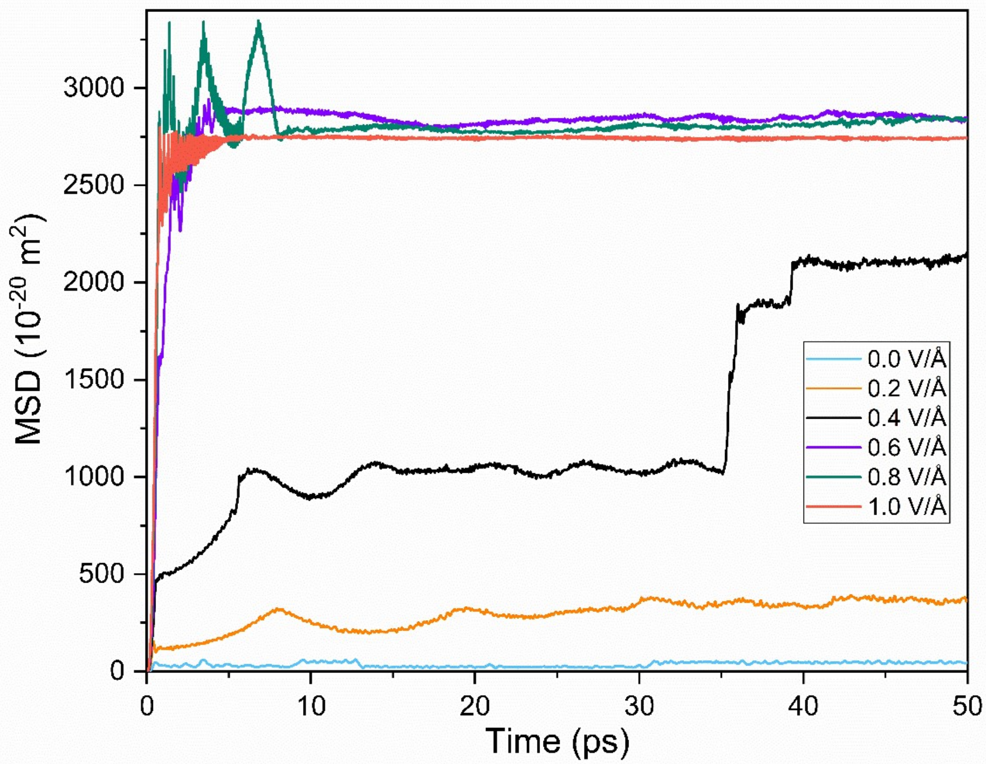

Figure S8 Unsmoothed curve: Total MSD vs time

1.1.5 Trajectory

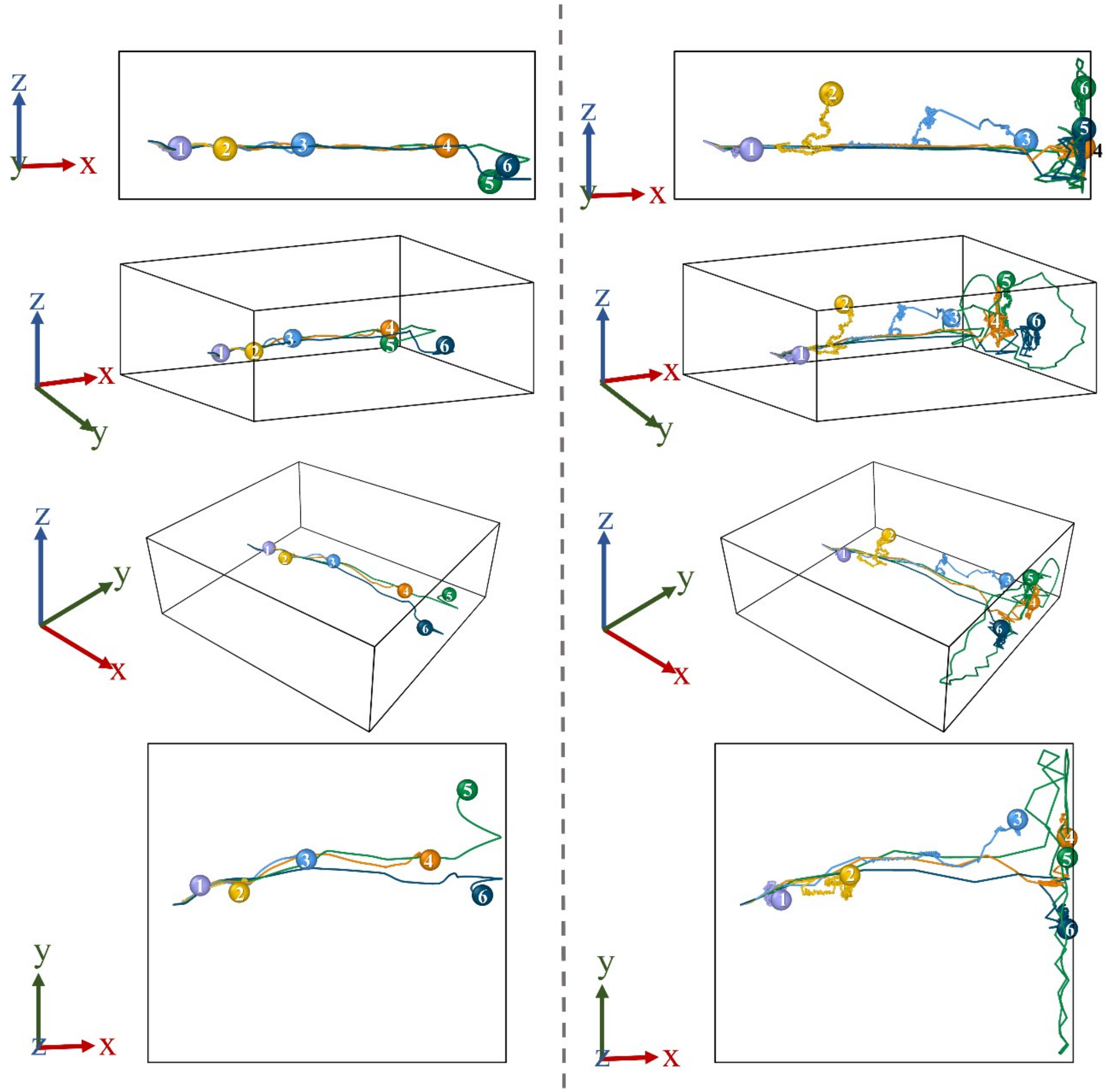

Figure S9 Left column represents trajectory for 1ps and right side represent 50ps. Data taken every 0.01ps & 0.1ps for left and right column respectively. From top to bottom: Front, Ortho, Perspective & Top view (1. lavender-0V/Å, 2. yellow-0.2V/Å, 3. blue-0.4V/Å, 4. orange -0.6V/Å, 5. green-0.8V/Å, 6. dark blue-1V/Å)

1.1.6 Interaction between Ion and Silicene

1.1.6.1 Smooth

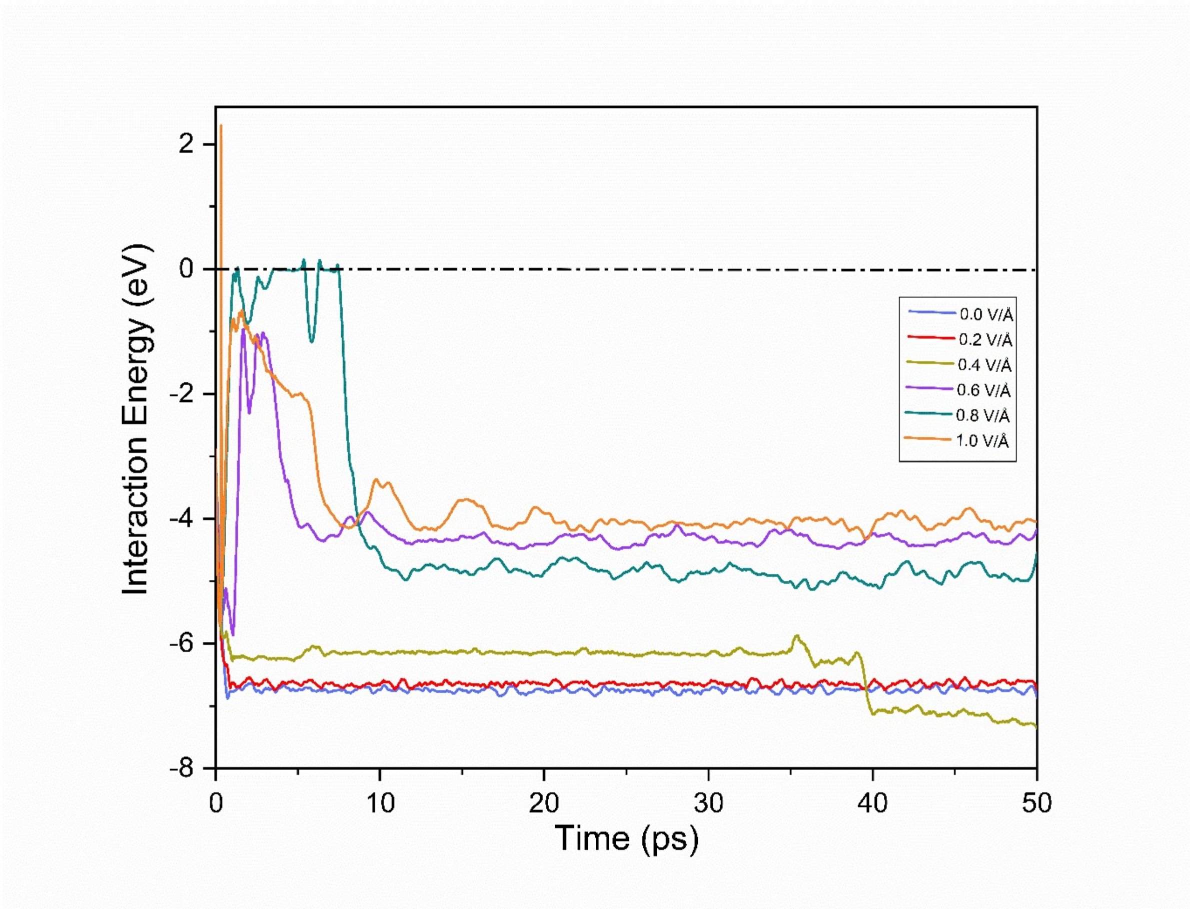

Figure S10 Smoothed curve: Interaction Energy vs time

1.1.6.2 Unsmooth

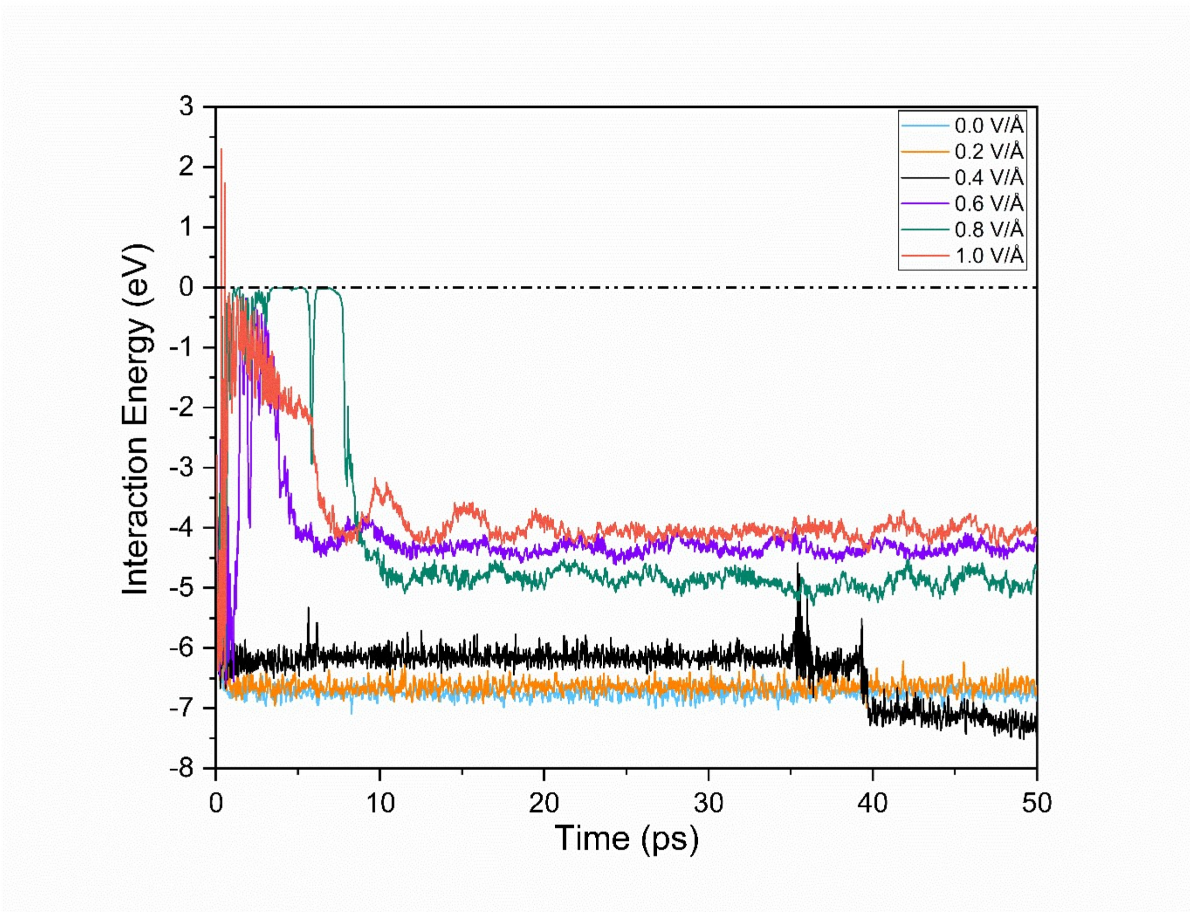

Figure S11 Unsmoothed curve: Interaction Energy vs time

1.2 Modified NVT:

1.2.1 MSD in x Direction:

1.2.1.1 Smooth

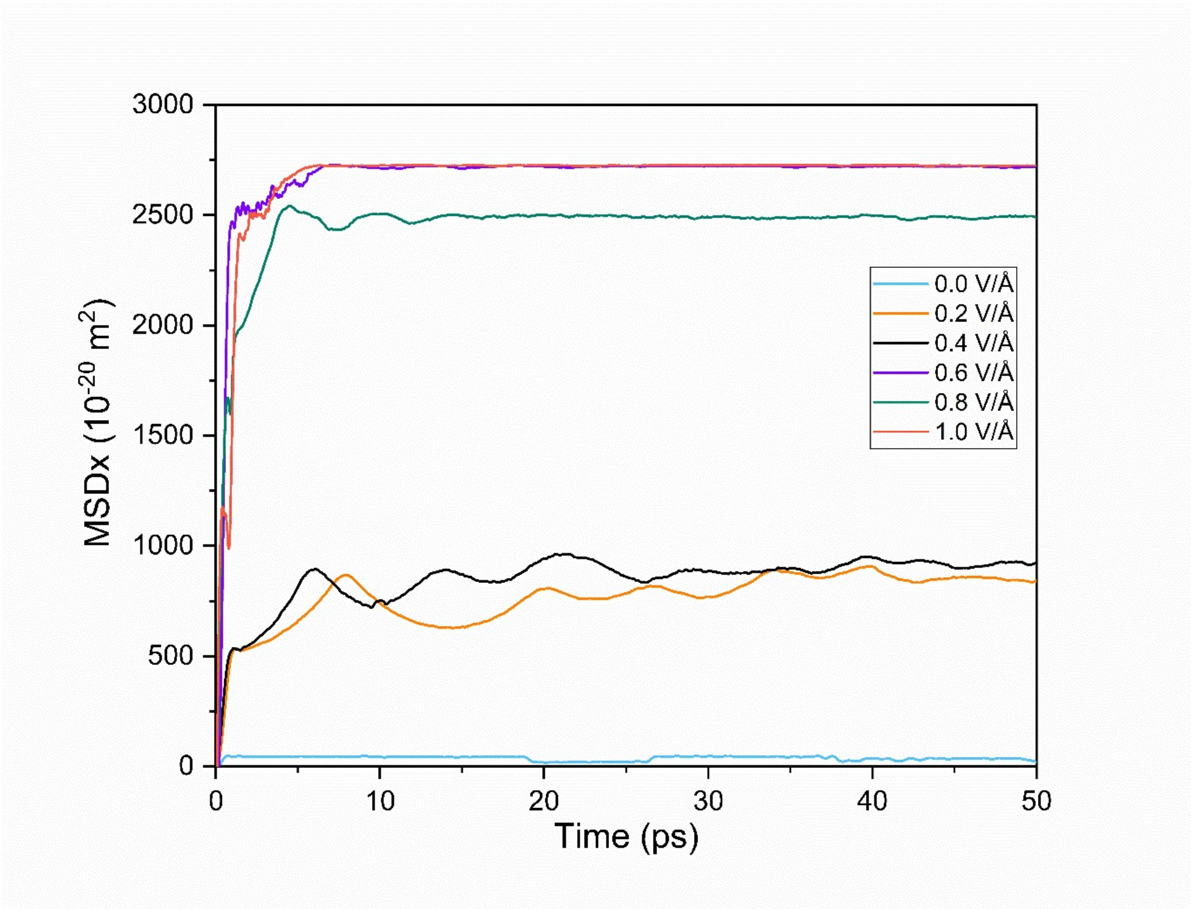

Figure S12 Smoothed curve: MSD behaviour in x direction vs time

1.2.1.2 Unsmooth

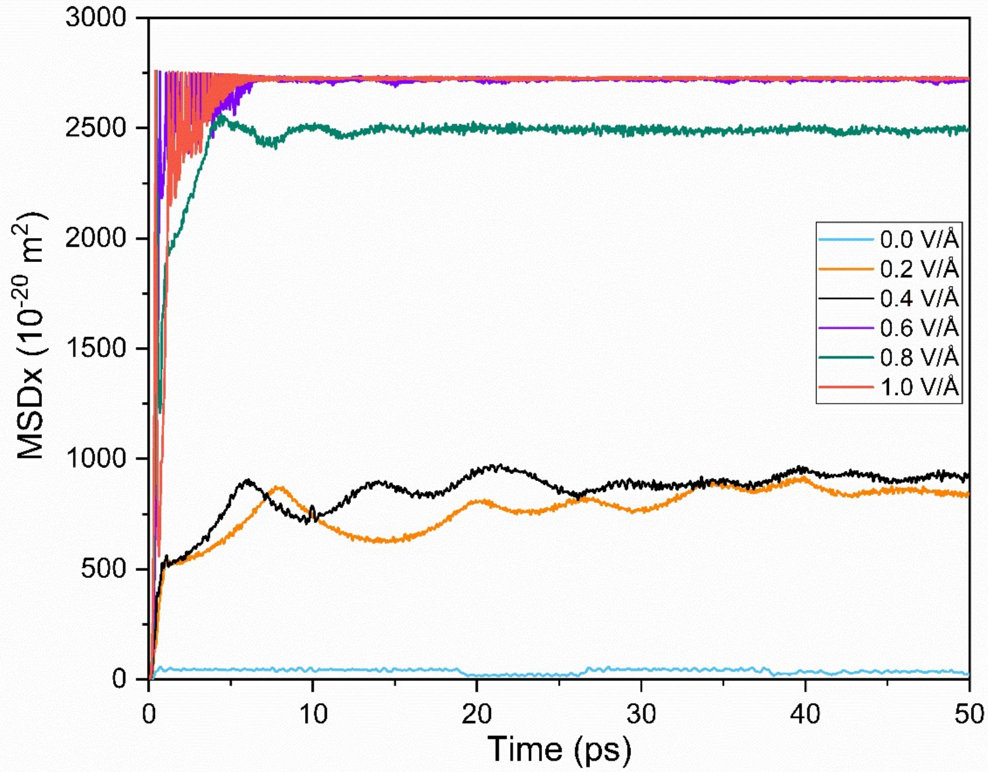

Figure S13 Unsmoothed curve: MSD behaviour in x direction vs time

1.2.2 MSD in y Direction:

1.2.2.1 Smooth

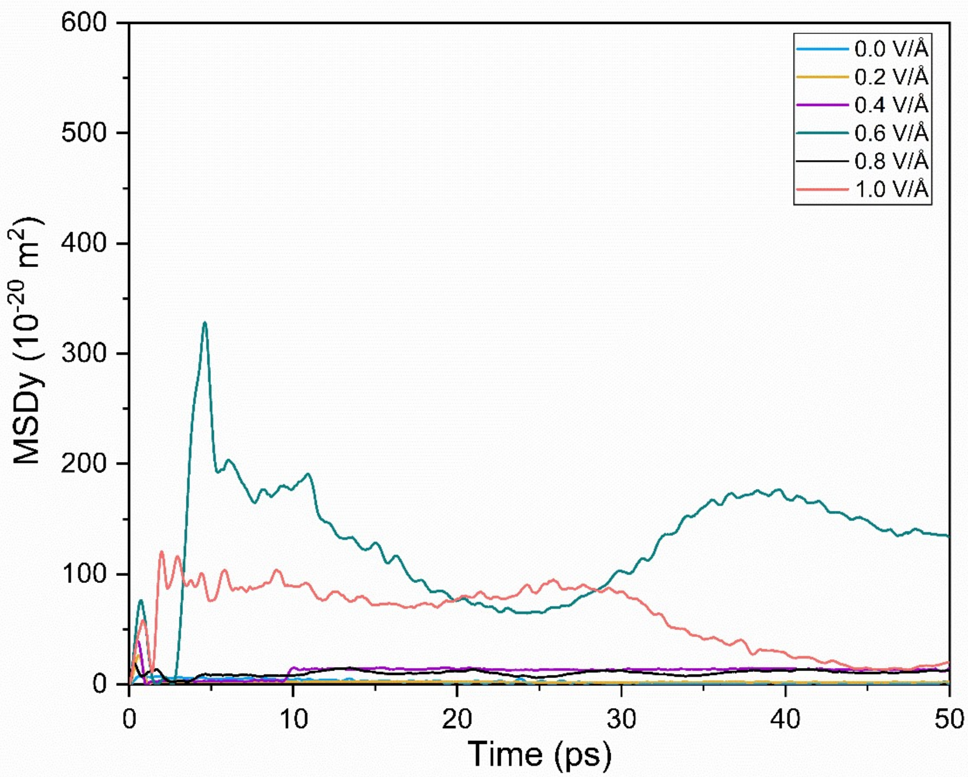

Figure S14 Smoothed curve: MSD behaviour in y direction vs time, demonstrating randomness.

1.2.2.2 Unsmooth

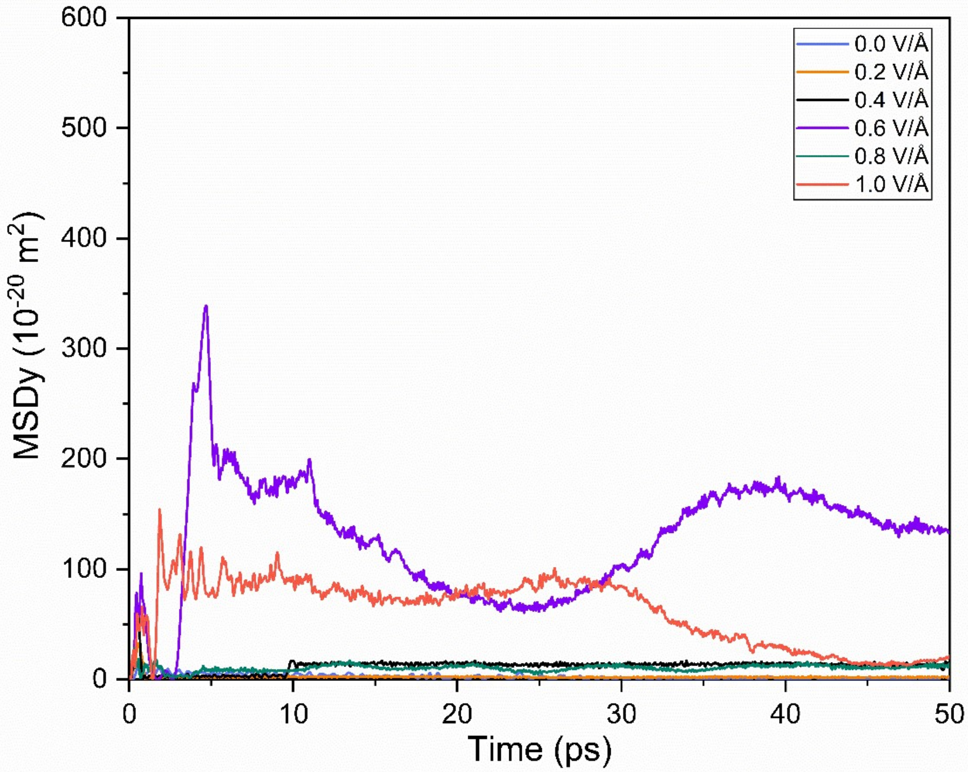

Figure S15 Unsmoothed curve: MSD behaviour in y direction vs time, demonstrating randomness.

1.2.3 MSD in z Direction:

1.2.3.1 Smooth

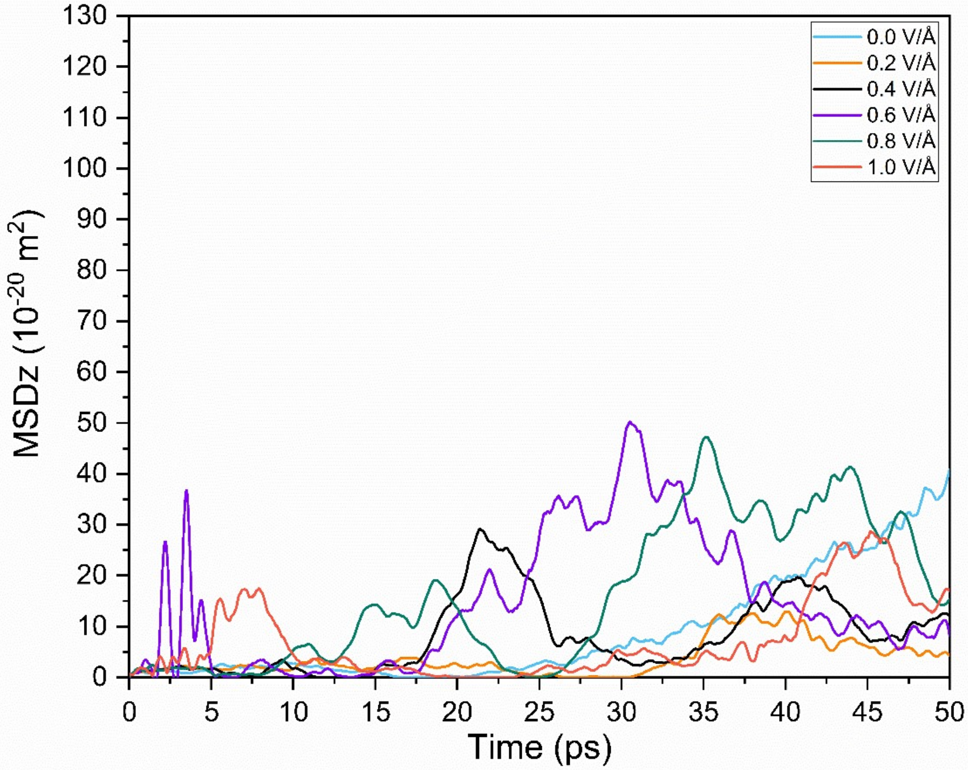

Figure S16 Smoothed curve: MSD behaviour in z direction vs time, demonstrating randomness.

1.2.3.2 Unsmooth

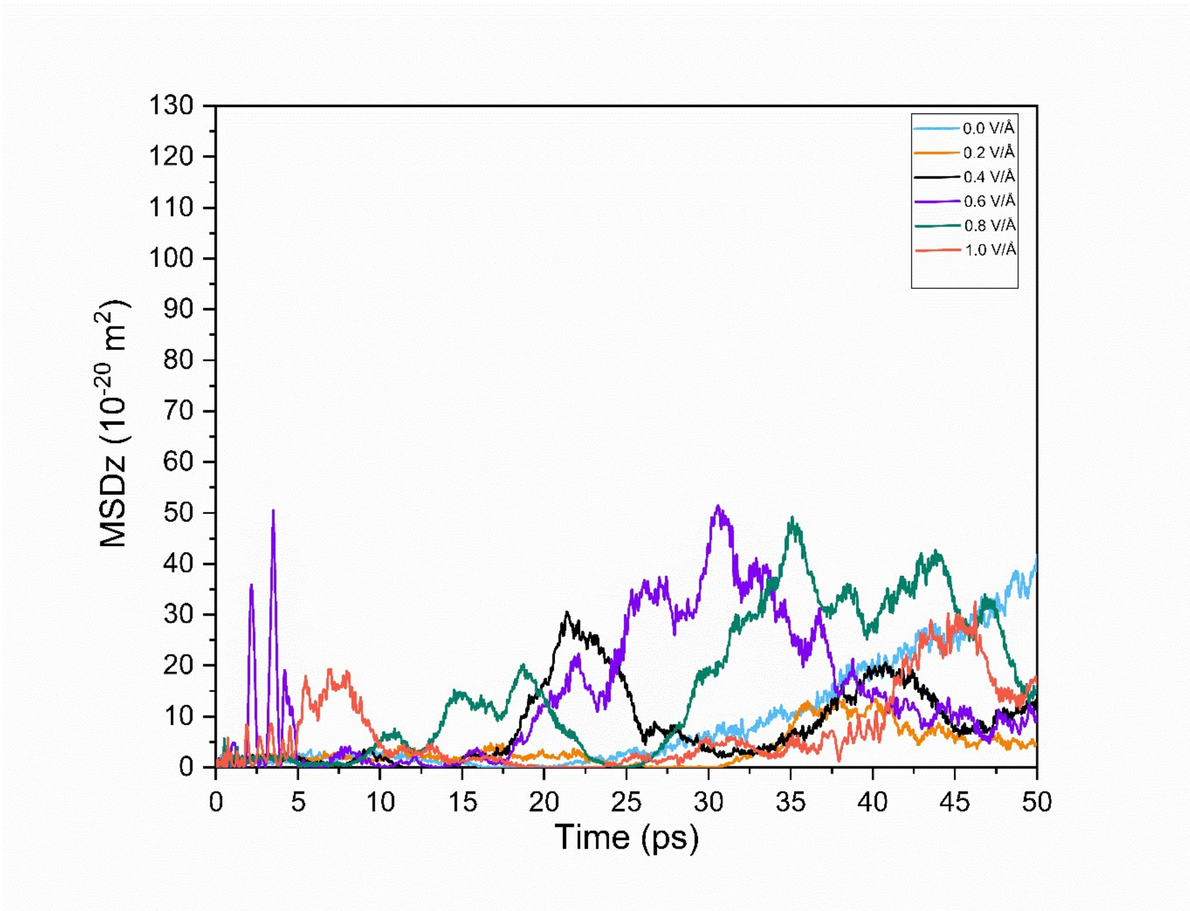

Figure S17 Unsmoothed curve: MSD behaviour in z direction vs time, demonstrating randomness.

1.2.4 Total MSD:

1.2.4.1 Smooth

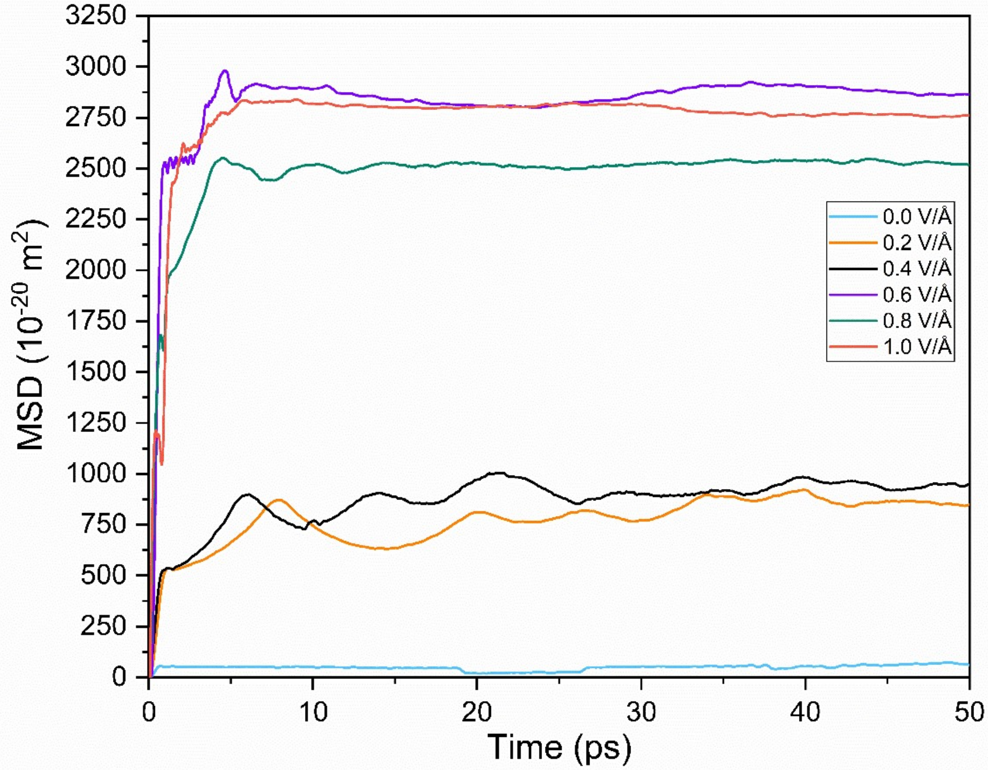

Figure S18 Smoothed curve: Total MSD vs time

1.2.4.2 Unsmooth

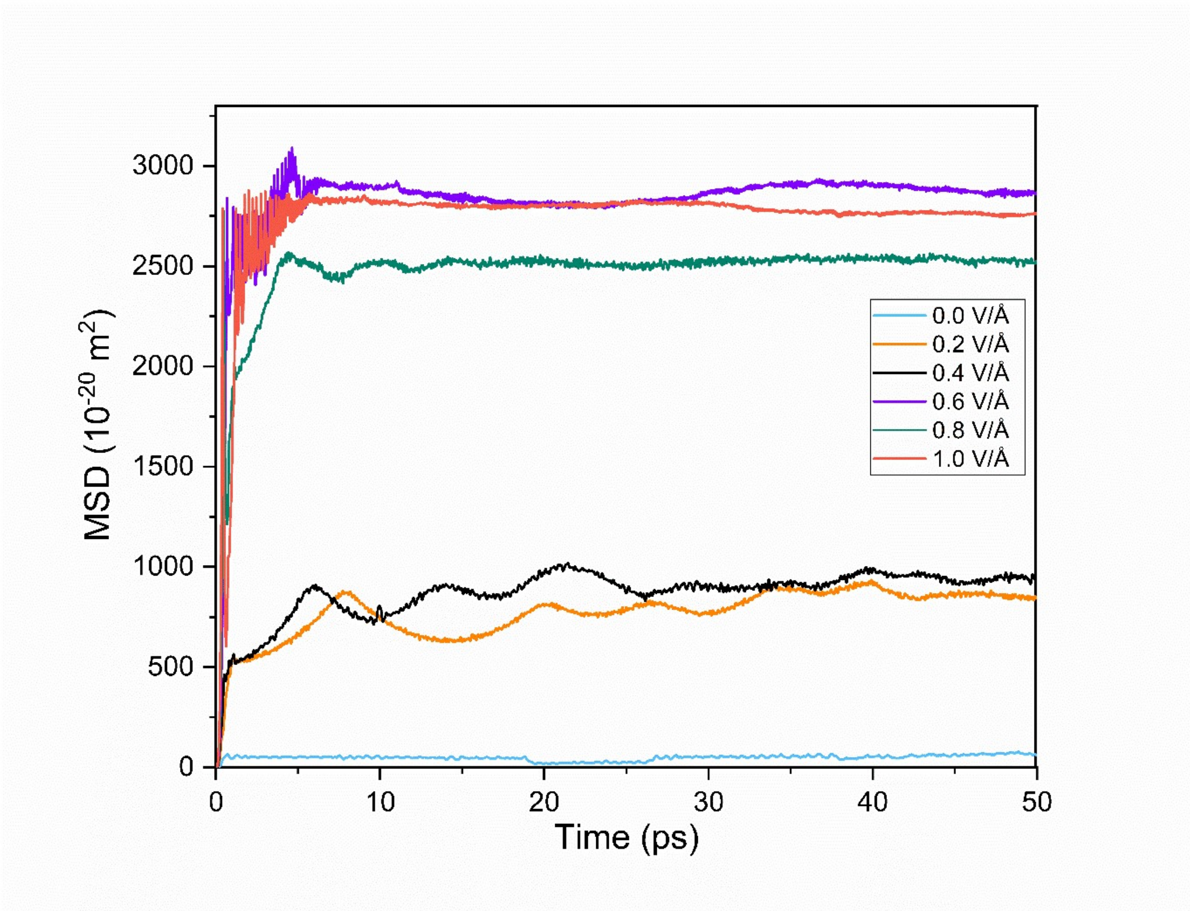

Figure S19 Unsmoothed curve: Total MSD vs time

1.2.5 Trajectory

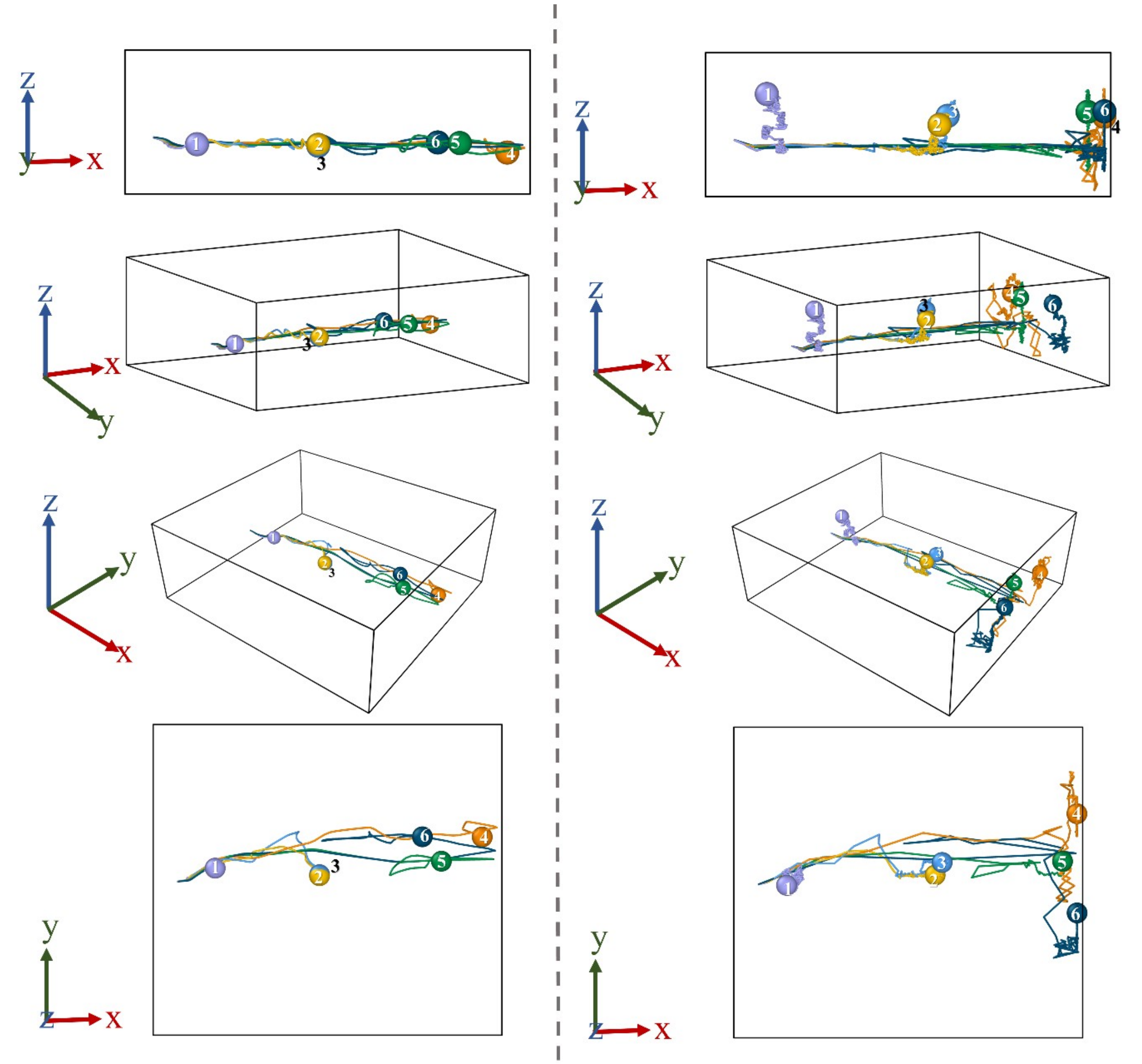

Figure S20 Left column represents trajectory for 1ps and right side represent 50ps. Data taken every 0.01ps & 0.1ps for left and right column respectively. From top to bottom: Front, Ortho, Perspective & Top view (1. lavender-0V/Å, 2. yellow-0.2V/Å, 3. blue-0.4V/Å, 4. orange -0.6V/Å, 5. green-0.8V/Å, 6. dark blue-1V/Å)

1.2.6 Interaction Energy:

1.2.6.1 Smooth

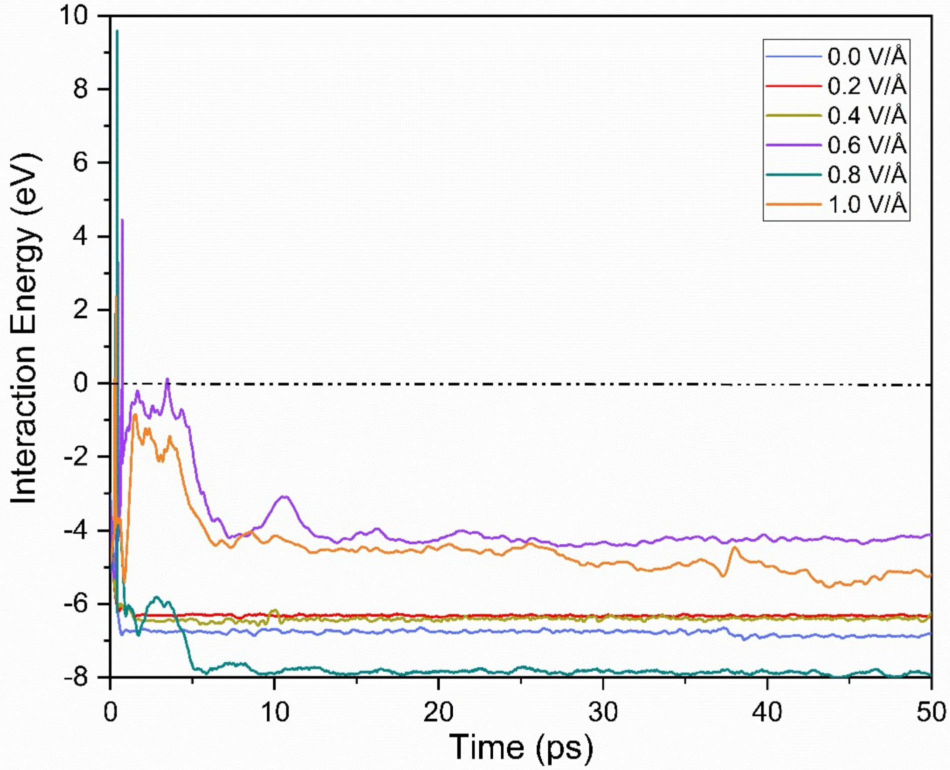

Figure S21 Smoothed curve: Interaction Energy vs time.

1.2.6.2 Unsmooth

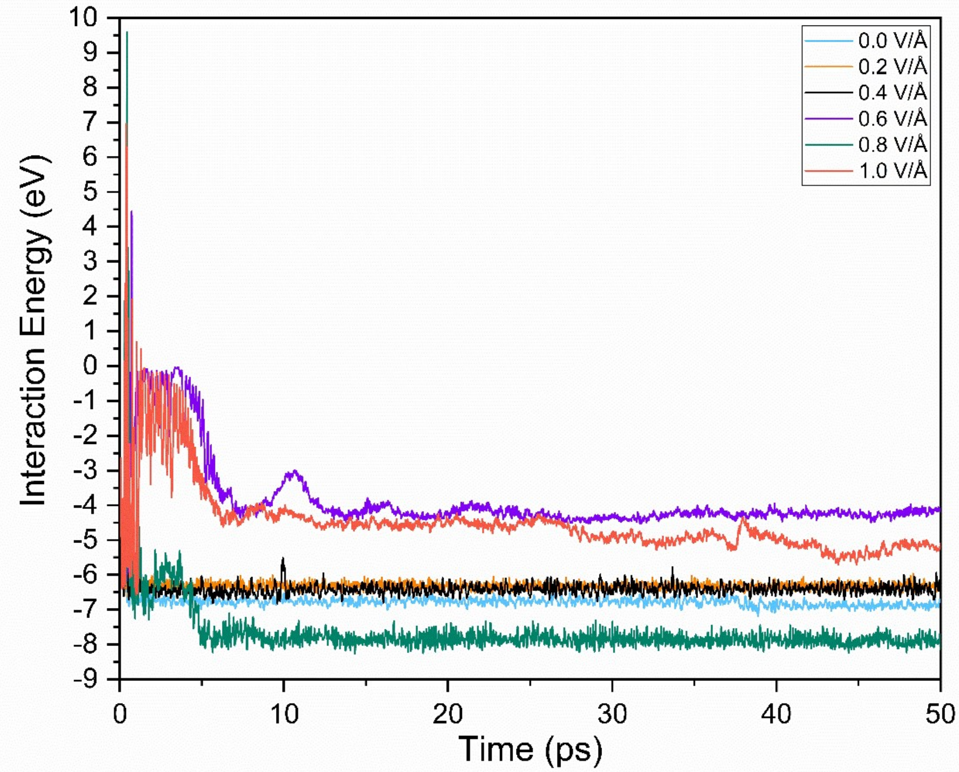

Figure S22 Smoothed curve: Interaction Energy vs time.

1.3 CSVR Thermostat:

1.3.1 MSD in x Direction:

1.3.1.1 Unsmooth

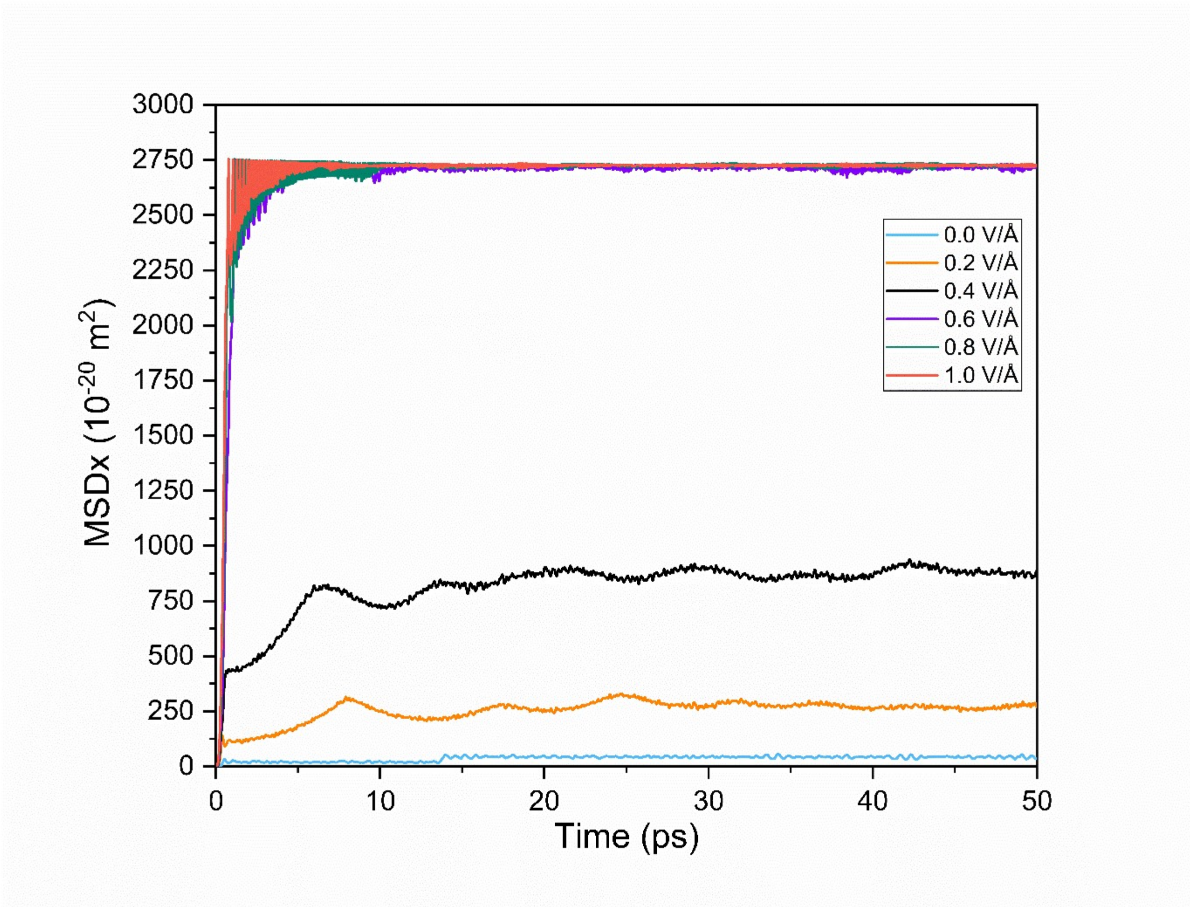

Figure S23 Unsmoothed curve: MSD behaviour in x direction vs time

1.3.2 MSD in y Direction:

1.3.2.1 Unsmooth

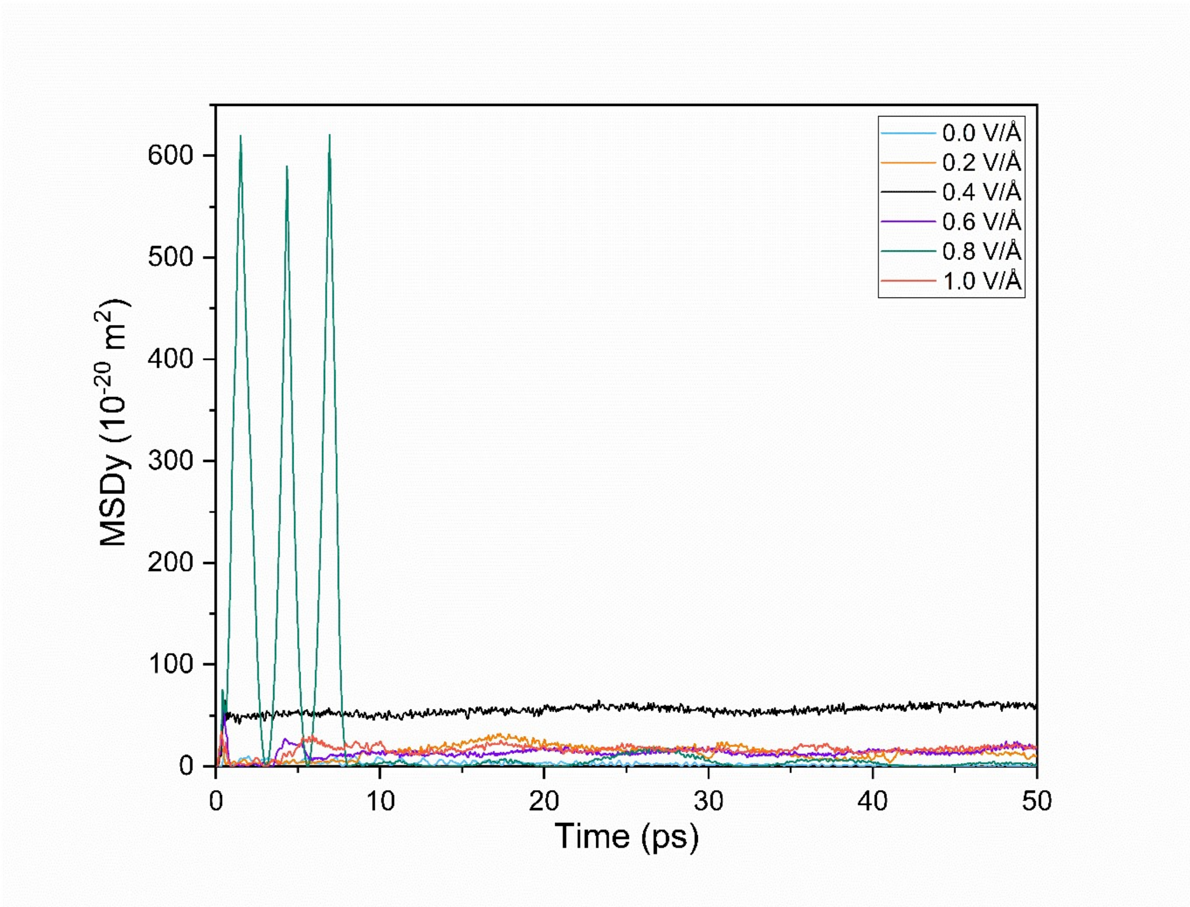

Figure S24 Unsmoothed curve: MSD behaviour in y direction vs time, demonstrating randomness.

1.3.3 MSD in z Direction:

1.3.3.1 Unsmooth

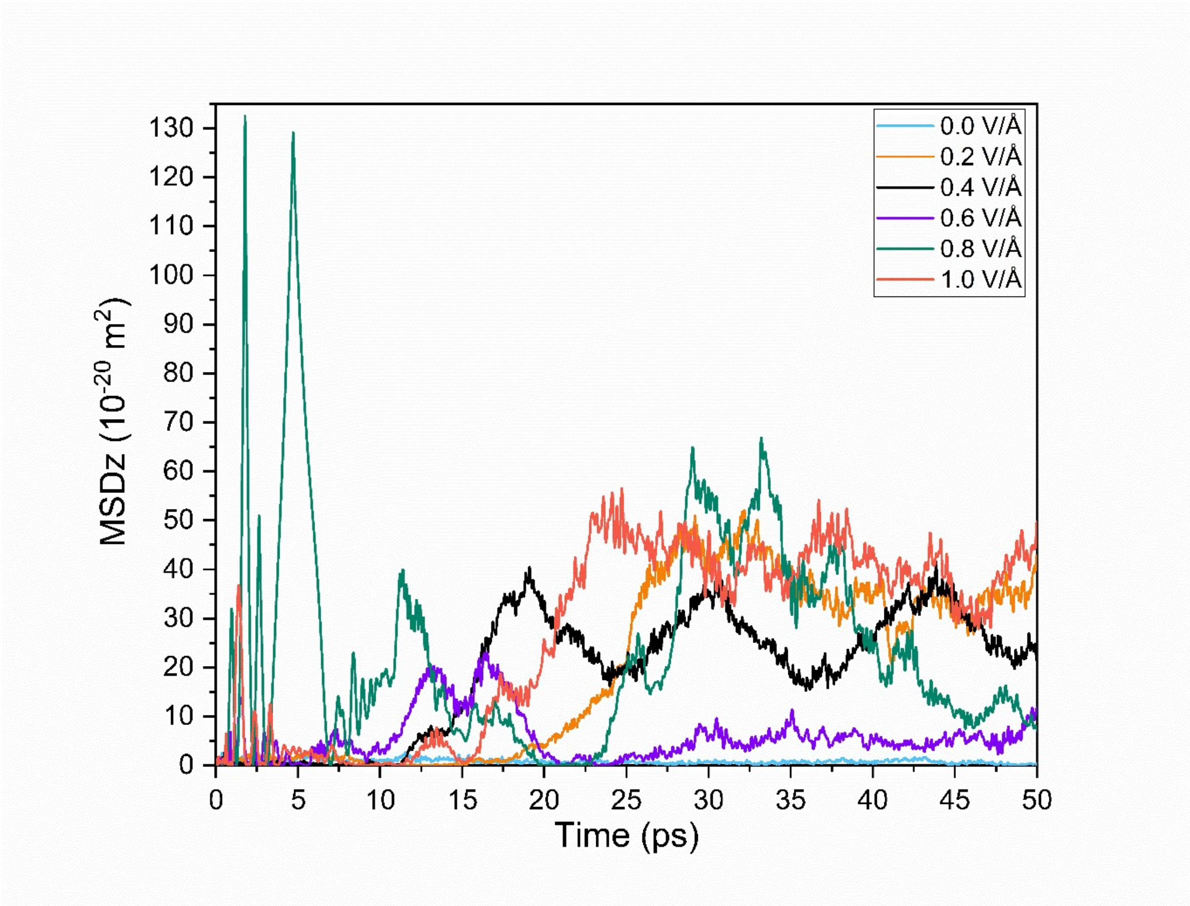

Figure S25 Smoothed curve: MSD behaviour in z direction vs time, demonstrating randomness.

1.3.4 Total MSD:

1.3.4.1 Unsmooth

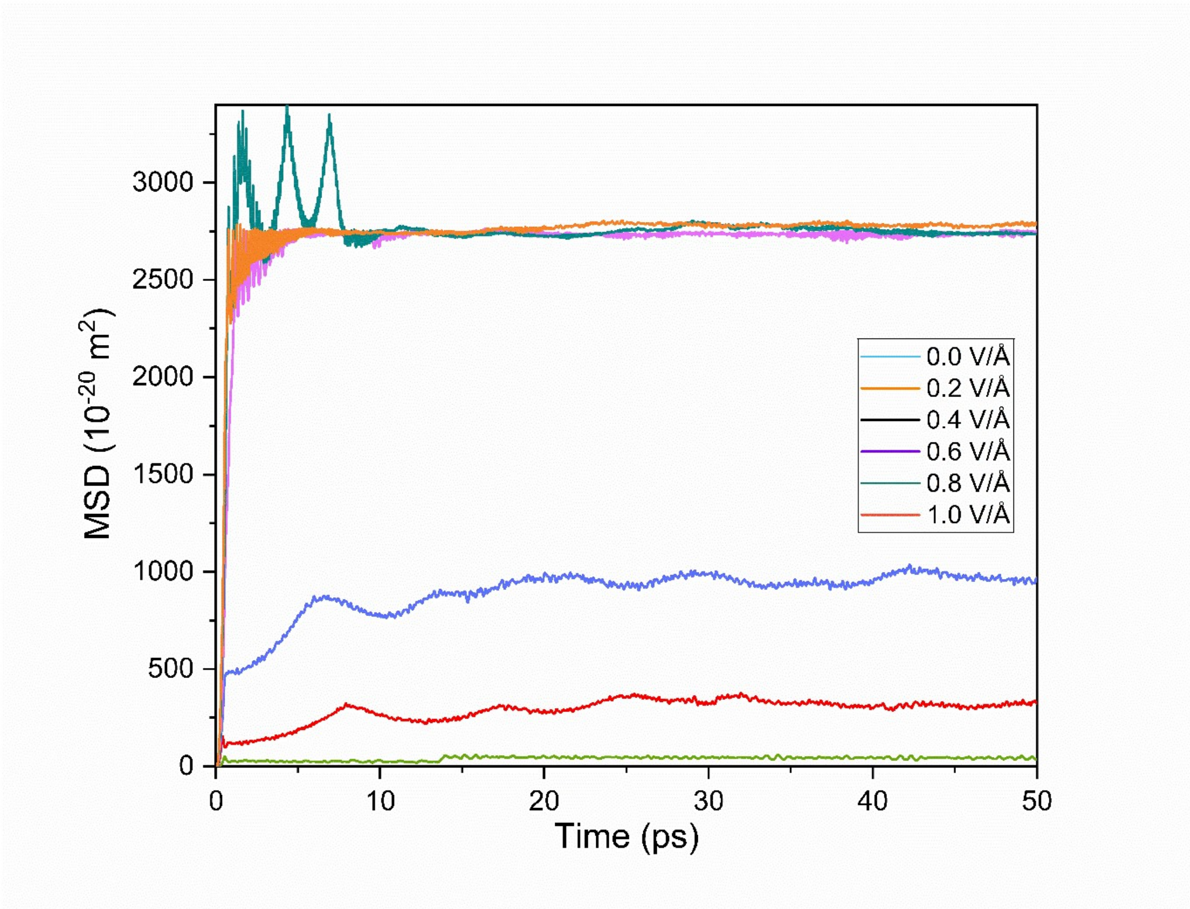

Figure S26 Unsmoothed curve: Total MSD vs time

1.3.5 Interaction Energy:

1.3.5.1 Unsmooth

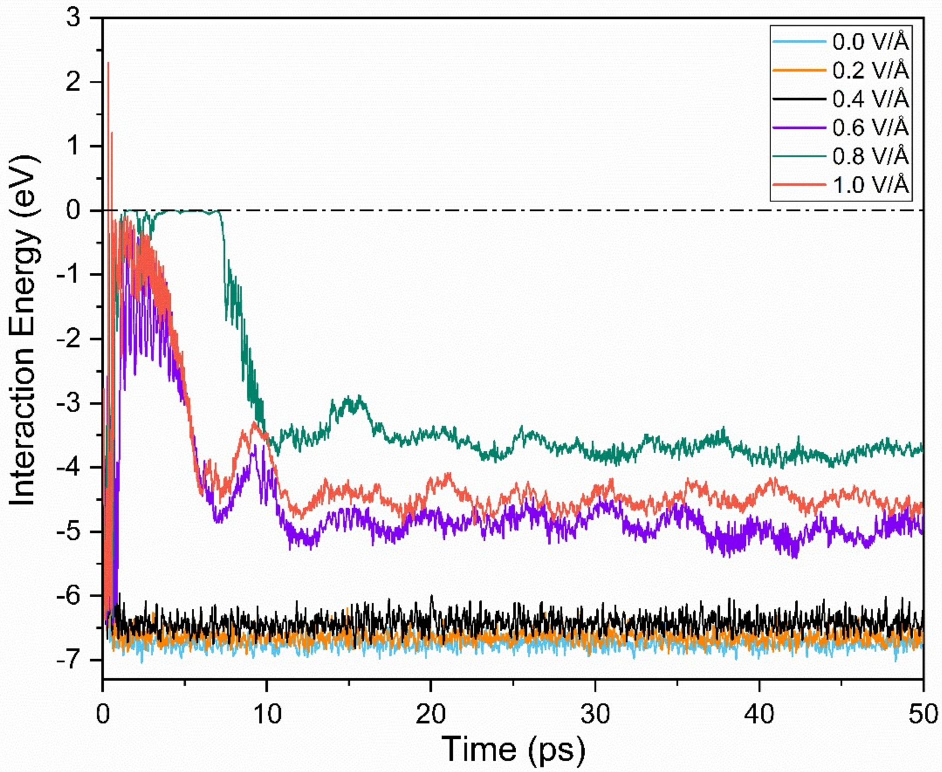

Figure S27 Unsmooth curve: Interaction Energy vs time.

2. References:

[1] S. C. Harvey, R. K. Tan, and T. E. C. Iii, "The flying ice cube: Velocity rescaling in molecular dynamics leads to violation of energy equipartition - Harvey - 1998 - Journal of Computational Chemistry - Wiley Online Library," vol. 19, no. 7, pp. 726–740, 1997, [Online]. Available: http://onlinelibrary.wiley.com/doi/10.1002/(SICI)1096-987X(199805)19:7%3C726::AID-JCC4%3E3.0.CO;2-S/pdf.

[2] G. Bussi, D. Donadio, and M. Parrinello, “Canonical sampling through velocity rescaling,” *J. Chem. Phys.*, vol. 126, no. 1, 2007, doi: 10.1063/1.2408420.

[3] E. Braun, S. M. Moosavi, and B. Smit, “Anomalous Effects of Velocity Rescaling Algorithms: The Flying Ice Cube Effect Revisited,” *J. Chem. Theory Comput.*, vol. 14, no. 10, pp. 5262–5272, 2018, doi: 10.1021/acs.jctc.8b00446.