Responses to referee reports for
"Multiscale response of ionic systems
to a spatially varying electric field".

First, I sincerely thank the referee for her/his comments. I have addressed all points made below.

1. I understand the concern raised by the referee.

   This work applies the extensively used non-equilibrium molecular dynamics simulation method. This method almost always uses "not experimental realizable" driving forces to probe the system response. Few examples: Simulations of Poiseuille flows where the fluid flow is driven by forces in the order of tera Newtons resulting in flow velocities (in a nanopore, that is) in the order 10 m/s. Temperature gradients of 100 Kelvin per nanometer. Water pulled through channels in cell membranes with an extreme large force that are nowhere present in cells.

   The interesting point here is that the system, i.e., the simulated model, respond to these synthetic forces in a *physical realistic* manner. In fact, this method is often used to calculate transport coefficients, e.g. the SLLOD method is applied in order to evaluate the shear viscosity very accurately. One can compare transport coefficient obtained with equilibrium simulations with the same coefficients extracted from non-equilibrium simulations and they agree in the limit of small forces as expected. Relevant for this work is the STF method (as referred to in the text). Here a large fictitious force field with amplitude of tera Newton and wavelength in order of Ångstrøm is applied to the system; the system response can be used to calculate the wavevector dependent viscosity. Even a (very synthetic) thermostat only removing specific modes for the temperature can be used! Hence, while the driving forces are "not experimentally realizable" the system response is *realistic*, thus, one is probing the correct dynamical behavior.

   There are theoretical foundations for these methods and I can recommend the text book by Evans and Morriss, Ref 7.

2. Very important point raised by the referee. The reason is a matter of computational speed; the shifted force method is much faster than any of the techniques based on the Ewald summation method.

   First, the maximum screening length is for the T_∞-system where in reduced units $\lambda_D = \sqrt{\epsilon T} \approx 1.6$, hence, $\lambda_D < 1.6$ for all systems. The cut-off is therefore around twice the screening length. The cut-off for the van der Waals interactions is quite large (the force is in the order of 10^{-6} at the cut-off distance), but is simply used here to reduce the number of simulation parameters.

   Indeed the shifted-force (SF) method may be questionable for non-inform systems. Especially, it will fail for confined systems - and so will the

standard Ewald methods. There are, however, strong indications that the SF method applies here: (i) the same results were found by letting $r_c = 6$ for selected situations, i.e., twice the cut-off distance. This simple check is always carried out. (ii) Comparing the non-equilibrium (i.e non-uniform) data with the predictions from the (uniform) linear response theory gives good agreement, Fig 4 b, also indicating that the SF method is applicable.

This concern is raised by both referee. To ensure that the SF method applies new simulations are carried out carefully comparing data using this method with data using the direct Ewald summation method. In the Ewald summation method the interaction energy is

$$V = \frac{1}{2} \sum_{\mathbf{n}} \sum_{ij} \frac{q_i q_j}{|\mathbf{r}_{ij} + L\mathbf{n}|}$$

$\mathbf{n}$ is an integer vector accounting for the replica systems. This direct method is not usually applied as the convergence is slow, however, for this particular simple and small system convergence is achieved quite quickly as seen in figure 4; using 124 replica systems suffices. This result is not

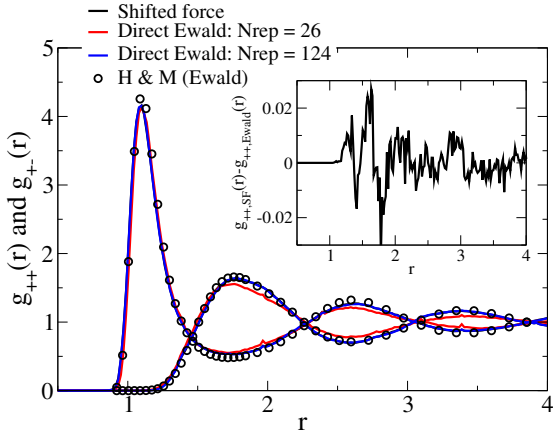

Figure 4: Radial distribution function for the molten salt system for the SF method and Ewald method using a different number of replica systems, N_{rep}. Also shown are the data points from Hansen and McDonald, Ref. 1 in the manuscript. The inset shows the difference between the SF and the Ewald method for N_{rep}=124 as this is not clear from the main figure.

surprising and is discussed in Refs. 16 and 17 in the manuscript; in Ref. 17 other properties are also compared.

Figure 5 shows an example for the charge density profile using the SF and Ewald methods in the non-equilibrium situation. Also, the corresponding spectra are shown. Clearly, the agreement is satisfactory.

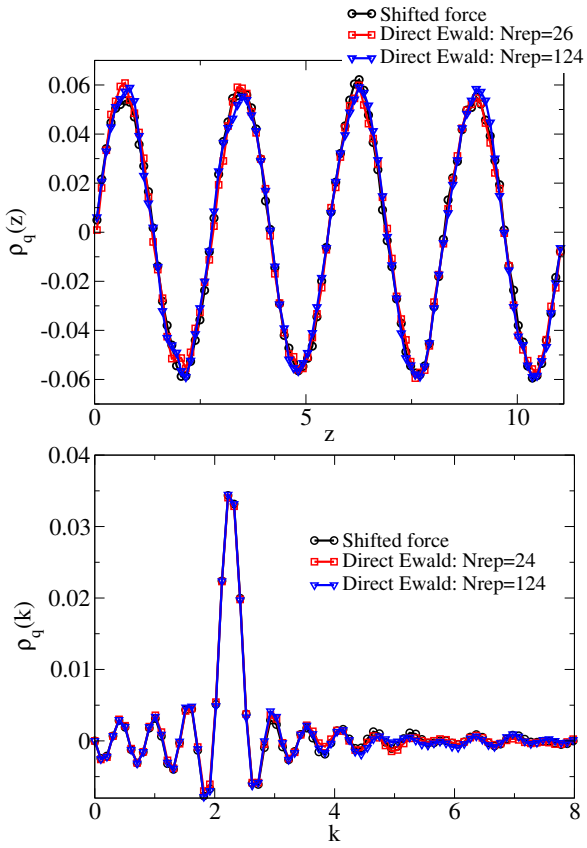

Figure 5: Upper figure: Charge profile results for the shifted force and Ewald methods. Lower figure: Corresponding spectra.

A paragraph justifying the application of the SF method is now included in the simulation details section.

This reply is copied in my response to the other referee.

3. I am not sure if I understand the question. As the referee correctly states the reduced charge (i.e. the charge in the simulations) is $q = q^*/\sqrt{\epsilon\sigma}$ where the q^* is the charge in units of $4\pi\epsilon_0$.

4. Indeed! Yurukawa is changed to Yukawa throughout the manuscript. Thank you for pointing this out.