

Dynamic Heterogeneity In The Monoclinic Phase Of CCl_4

Supporting Information

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Method for Detecting Rotational Jumps

In order to detect the rotational jumps of the CCl_4 molecules we perform a detailed analysis of the trajectories based on an algorithm published by N. J. Carter and A. Cross; *Nature* **435** 308 (2005). Let us consider the coordinates of each one of the Cl atoms, relative to their bonded C atom, and denote them by $r_{n,\alpha,i}$. Here α represent any of the Cartesian coordinates x , y or z ; the index n denote molecule and $i = 1, 2, 3, 4$ refers to each one of the Cl atoms of molecule n .

$$f_{n,\alpha,i}(t) = \frac{\langle r_{n,\alpha,i} \rangle_- - \langle r_{n,\alpha,i} \rangle_+}{\sqrt{\frac{S_-}{\Delta t} + \frac{S_+}{\Delta t}}} \quad (1)$$

where the angular brackets represent time average, the subindices $-$ and $+$ indicate the lapse of length Δt before and after time t , respectively; and S_- and S_+ are the corresponding standard deviations. Whenever there is a rotational jump the mean values and standard deviation are affected. Consequently, the test function $f_{n,\alpha,i}(t)$ reflects those changes with a spike. The spike could be upwards or downwards depending on the direction of change of the average coordinate.

In order to have a single function per molecule, the twelve individual $f_i(t)$ for an individual molecule can be combined in the following way:

$$\mathcal{F}_n(t) = \sum_{i=1,2,3,4} \sum_{\alpha=x,y,z} |f_{n,\alpha,i}(t)| \quad (2)$$

By careful inspection of the simulation trajectories and the response of the test functions we selected a time lapse $\Delta t = 50$ ps. In Figure S1 we show an example of the variation of the relative coordinates $x_{n,\alpha,i}(t)$ corresponding to the four Cl atoms of molecule n , which undergoes a jump at $t = 2971.39$ ns. The figure also shows the corresponding four individual response functions of each Cl atom:

$$F_{n,i}(t) = \sum_{\alpha=x,y,z} |f_{n,\alpha,i}(t)| \quad (3)$$

along with the final curve of the test function $\mathcal{F}_n(t)$. A threshold value of $\mathcal{F}_n(t) = 50$ was used to determine the rotational jumps.

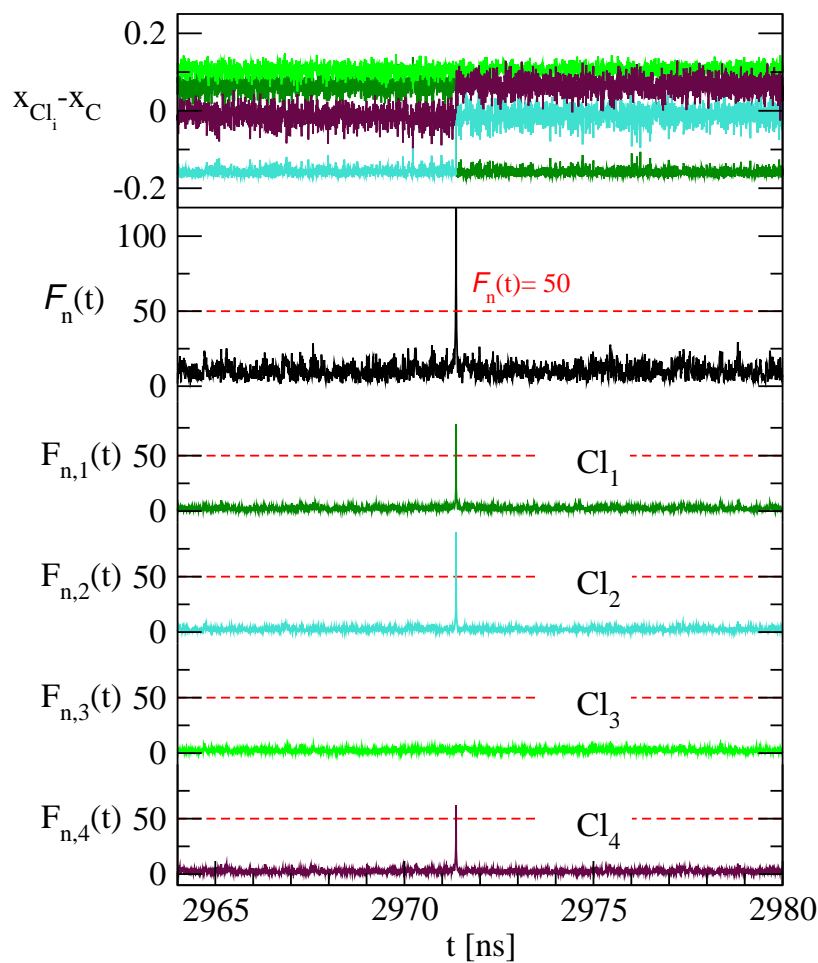


Figure S1: Top: Variation of the relative x coordinates of the four Chlorine atoms of molecule n in green (Cl_1), cyan (Cl_2), light green (Cl_3) and orange (Cl_4). The four individual response functions are also shown in the corresponding color, along with the final curve of the test function (in black). The threshold value of 50 is shown in red.

Radial Distribution Functions

In order to analyze the crystalline structure of the simulation supercell, we calculated the Carbon-Carbon radial distribution function for each possible combination of Carbon atoms within the different 4 groups of molecules in the system, at the all the simulation temperatures. We observed a good agreement of the function's peaks with the experimental values for the Carbon-Carbon distances on the monoclinic cell. On figure S2 we show the results at the lowest and highest simulation temperatures.

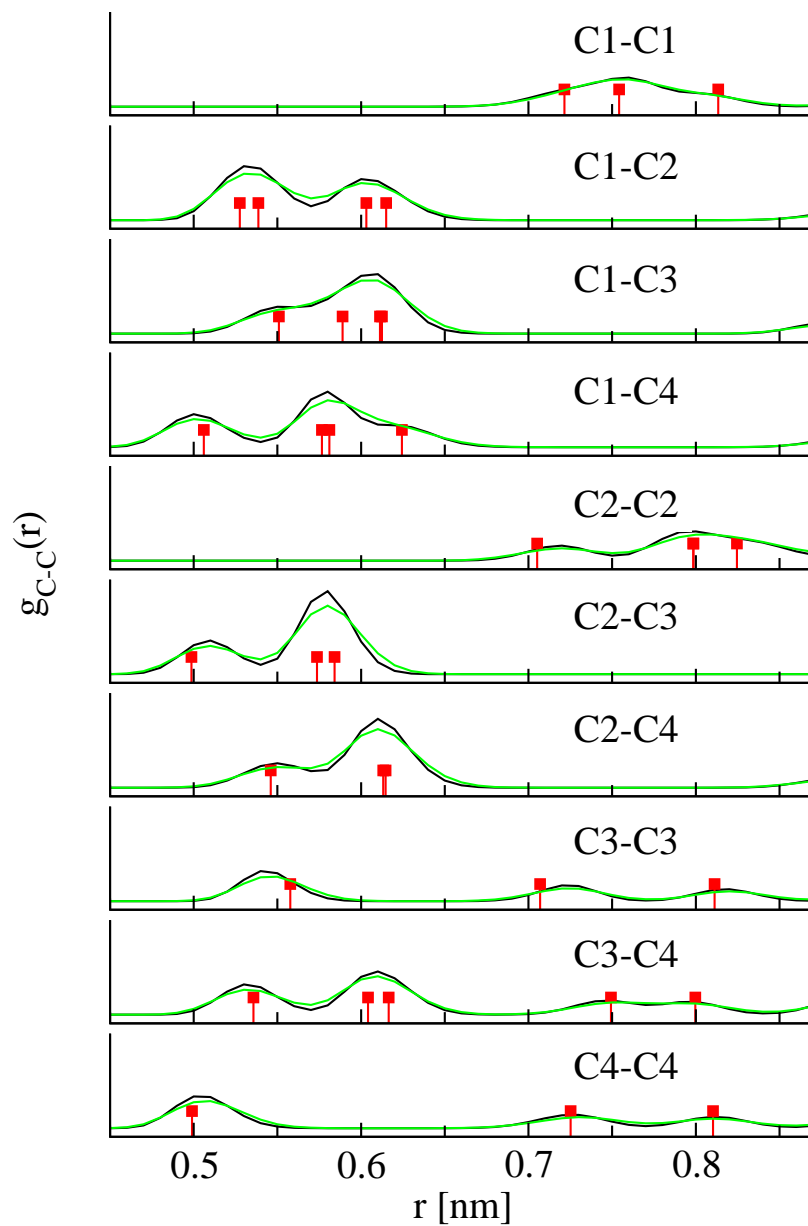


Figure S2: Carbon-Carbon radial distribution function for all possible combination of groups in the system at 160 K (black line) and 220 K (green line). On the graph the experimental values are also shown in red.