

1 Test for interference effect for low sample rate over the long simulation run.

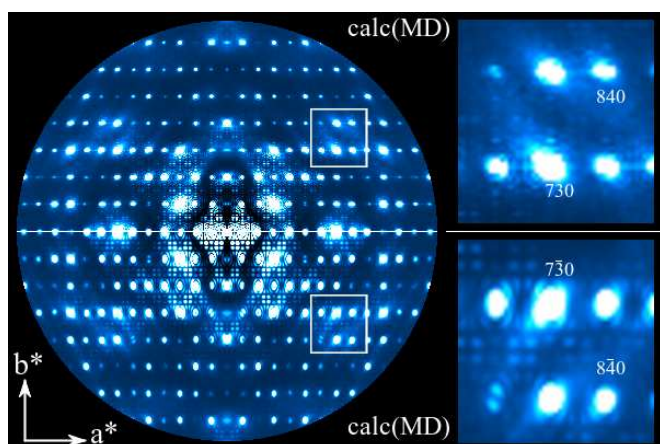


Figure 1

If the long simulation time allowed for long wavelength correlations (phonons) to develop then the correlation length could be bigger than half of the simulation box causing interference effects. This was checked by limiting the lot size to half of the simulation box size. The top hemi-sphere corresponds to trial #12 as described in **Table 1** and shown in **Figure 5(h)** in the main article. Here the lot size is set to $8 \times 15 \times 8$ and a R -value was calculated to be 21.13%. The bottom hemi-sphere corresponds to the same trial simulation only the lot size for the diffuse scattering calculation was set to $6 \times 8 \times 6$ and gave an R -value 20.73%. The results imply that the high intensity artifacts which are centered at Bragg positions could be coming from interference related mostly to the time sampling.

2 Result for increasing the lot size for MD trajectories with the larger simulation box.

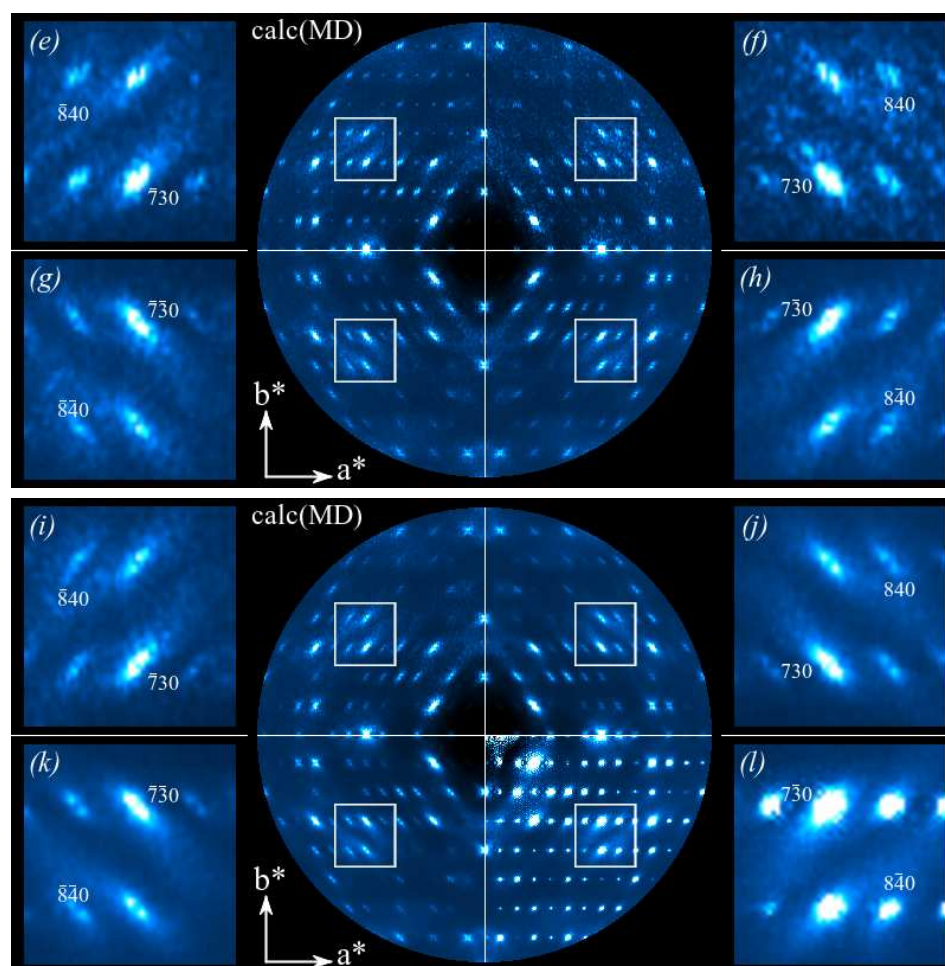


Figure 2

Output $hk0$ diffuse diffraction sections calculated from the MD simulation of Aspirin (form I) using methods detailed in **section 2** and shown in **Figure 5** of the main article. The results correspond to those trials where the simulation box size was set to $12 \times 17 \times 12$ unit cells. These images are calculated using a lot size that is set to match this simulation box size and represent the effect of increasing the lot size of the diffuse scattering calculation for each of those trials. Each trial output has been assigned to a quadrant such that the corresponding trial from **Figure 5** in the main article has the same lettering, thus (e)–(l) are calculated from the MD trajectories for trials #5 – 12 as described in **Table 1**. The respective R -values are 20.34, 20.58, 19.95, 20.26, 19.93, 19.87, 19.87 and 21.24 %. See main text for further details on the differences.