nature portfolio

https://doi.org/10.1038/s43586-021-00034-1

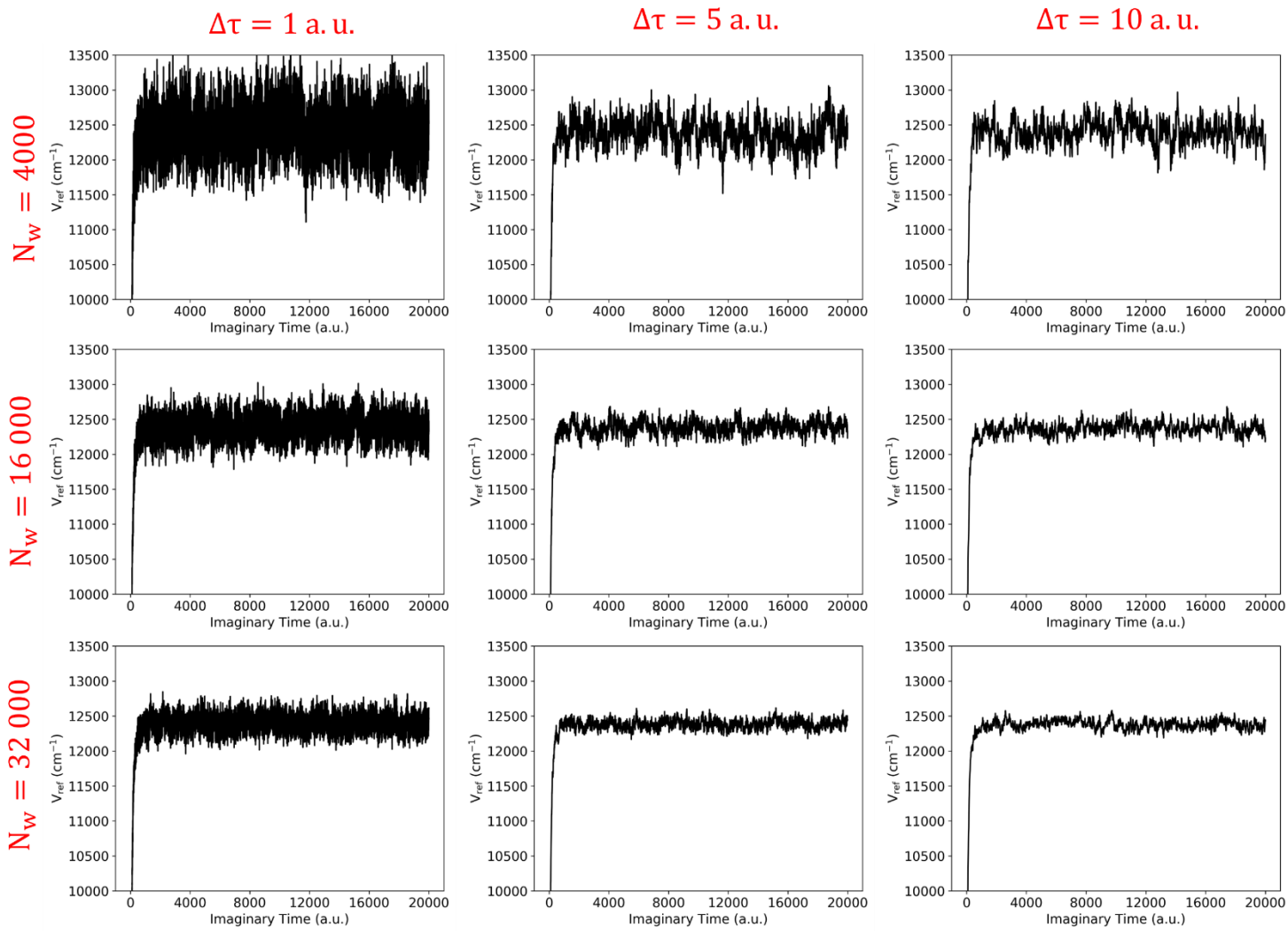
Supplementary information

Computational molecular spectroscopy

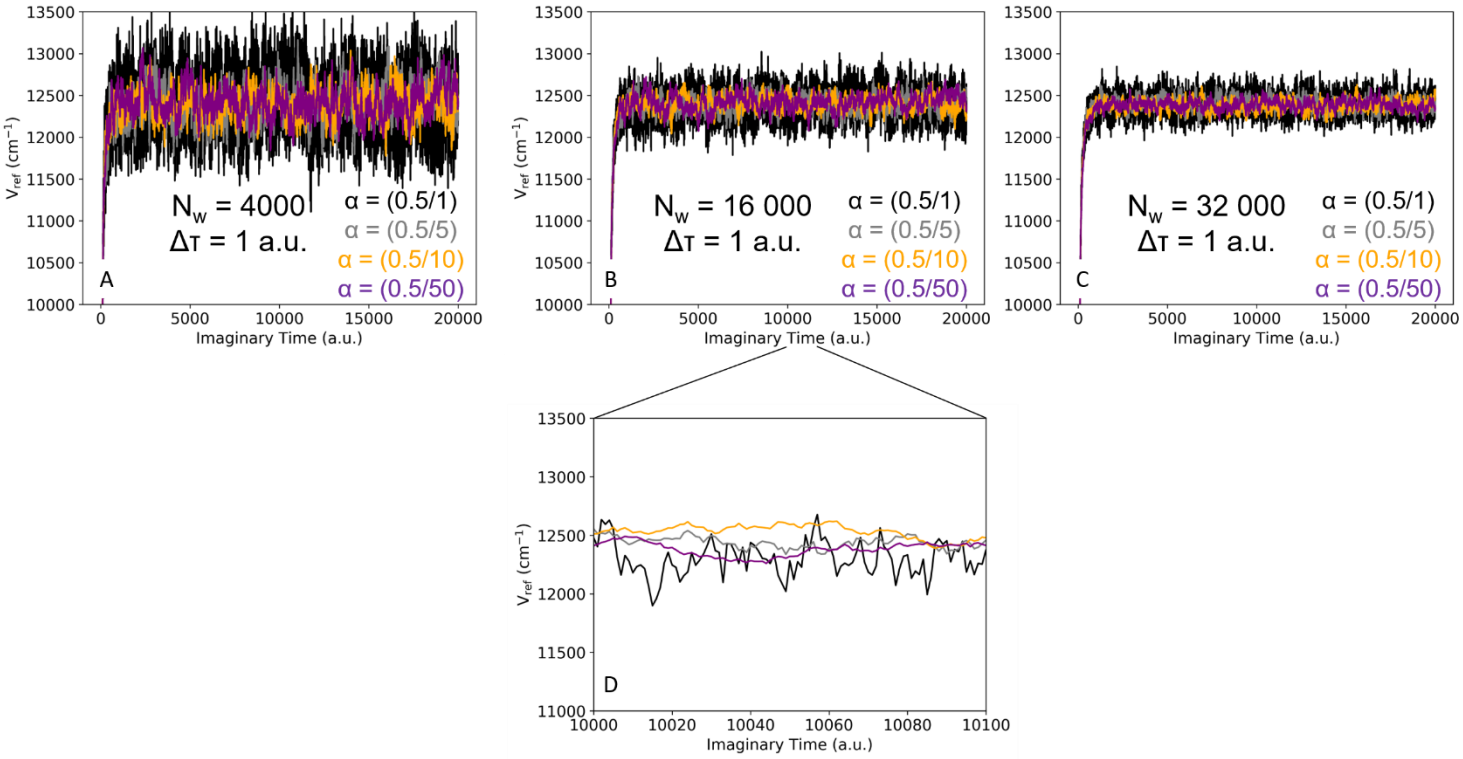
In the format provided by the authors and unedited

Supplementary Information for Computational Molecular Spectroscopy

Puzzarini *et al.*



Supplementary Figure 1: V_{ref} plotted a function of imaginary time for the ensemble sizes and time increments indicated.



Supplementary Figure 2: V_{ref} plotted a function of imaginary time for the ensemble sizes and time increments indicated. The value of α (reported in Hartrees) is varied to show how it affects the size of the fluctuations in V_{ref}. In panel D, we plot an expanded view of the region of the middle panel B.

Supplementary Table 1: Averaging Zero Point Energy over different Time Intervals, $N_w = 4000$[a]

Time Averaged (a.u.) [b]	$\Delta\tau$ = 1 a.u.	$\Delta\tau$ = 5 a.u.	$\Delta\tau$ = 10 a.u.
400 – 20 000	12 404 (14)	12 394 (11)	12 408 (12)
8000 – 20 000	12 409 (17)	12 382 (17)	12 408 (17)
12 000 – 20 000	12 412 (24)	12 385 (26)	12 409 (25)
16 000 – 20 000	12 412 (25)	12 383 (23)	12 408 (27)

[a]The numbers provided in parenthesis are the uncertainties in the reported zero-point energies. These uncertainties are calculated based on five independent DMC simulations.

[b]Time averaged refers to the amount of imaginary time (tau) that is used to average V_{ref}.

Supplementary Table 2: Averaging Zero Point Energy over different Time Intervals, $N_w = 16\,000$

Time Averaged (a.u.) [b]	$\Delta\tau$ = 1 a.u.	$\Delta\tau$ = 5 a.u.	$\Delta\tau$ = 10 a.u.
400 – 20 000	12 396 (8)	12 388 (5)	12 381 (5)
8000 – 20 000	12 396 (7)	12 385 (4)	12 385 (4)
12 000 – 20 000	12 393 (8)	12 387 (9)	12 388 (8)
16 000 – 20 000	12 396 (10)	12 386 (11)	12 382 (8)

Supplementary Table 3: Averaging Zero Point Energy over different Time Intervals, $N_w = 32\,000$

Time Averaged (a.u.) [b]	$\Delta\tau$ = 1 a.u.	$\Delta\tau$ = 5 a.u.	$\Delta\tau$ = 10 a.u.
400 – 20 000	12 396 (3)	12 396 (5)	12 383 (6)
8000 – 20 000	12 398 (4)	12 392 (5)	12 380 (8)
12 000 – 20 000	12 399 (8)	12 390 (6)	12 379 (8)
16 000 – 20 000	12 403 (10)	12 393 (7)	12 382 (8)