

RDX Compression, $\alpha \rightarrow \gamma$ Phase Transition and Shock Hugoniot
Calculations from Density Functional Theory Based Molecular
Dynamics Simulations

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Supplementary Material

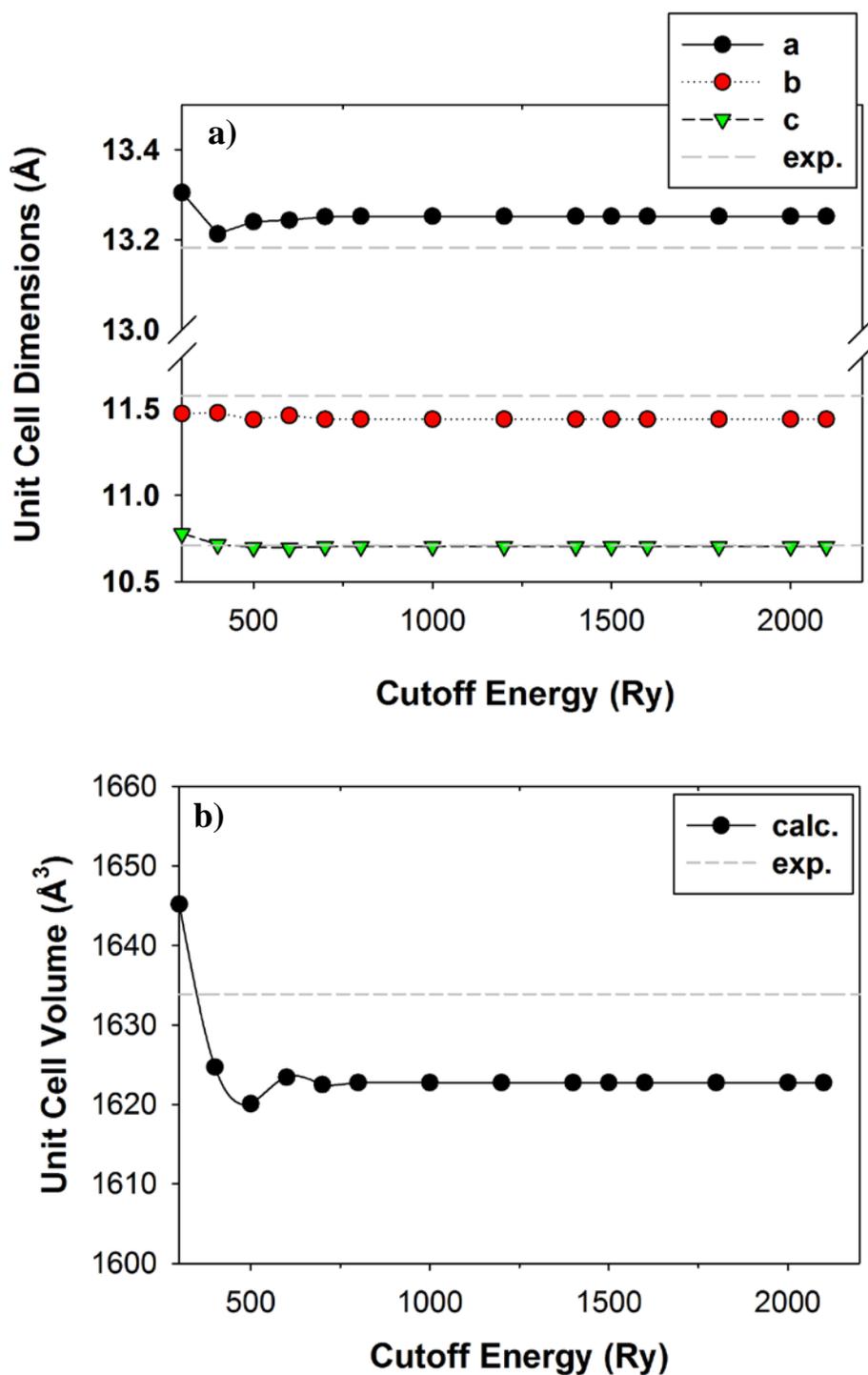


Figure S1. Variation of the predicted unit cell dimensions (a) and unit cell volume (b) for the α -RDX crystal with cutoff energy as obtained by using the CP2K program at the PBE-D2 level. The dotted lines indicate the corresponding experimental values from reference ¹.

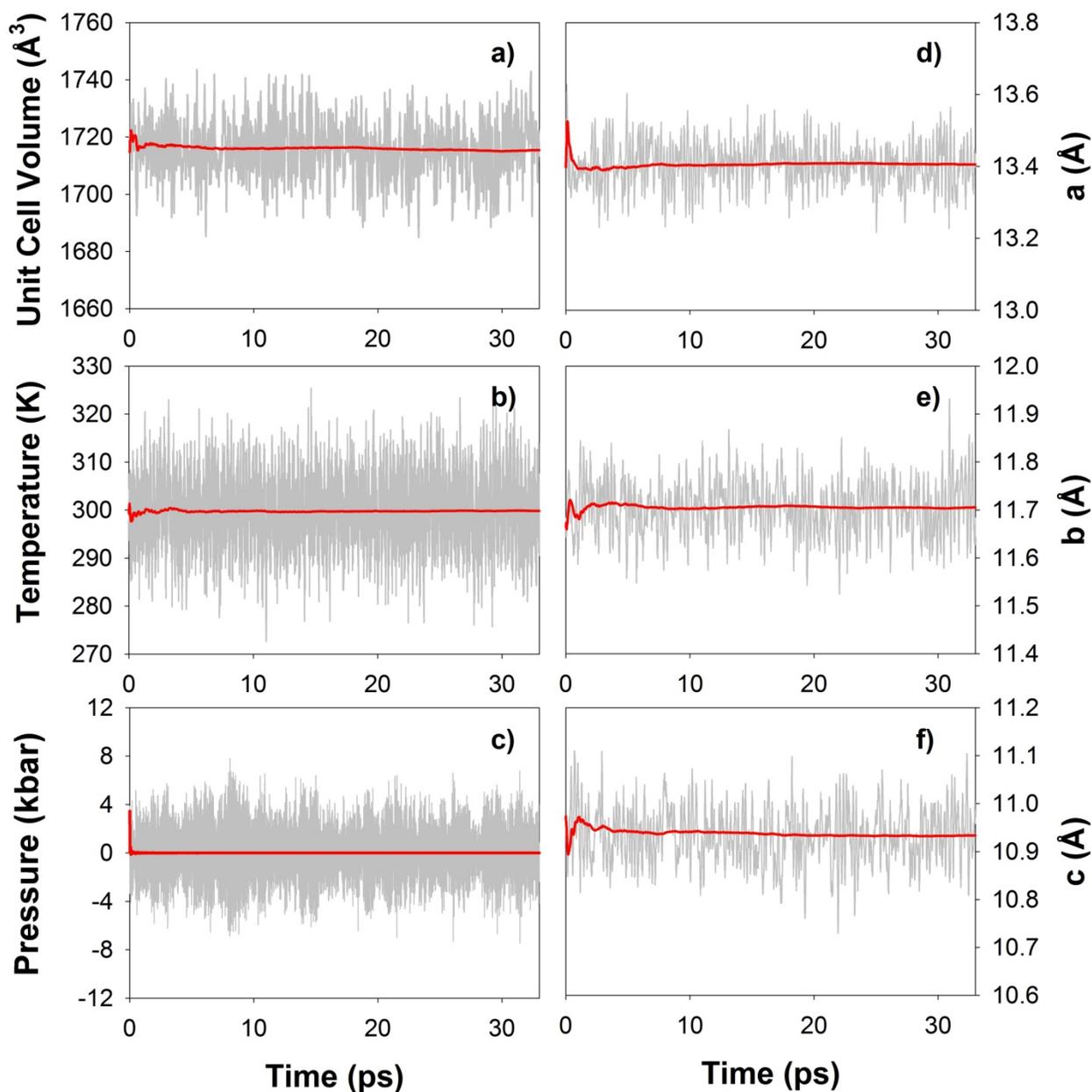
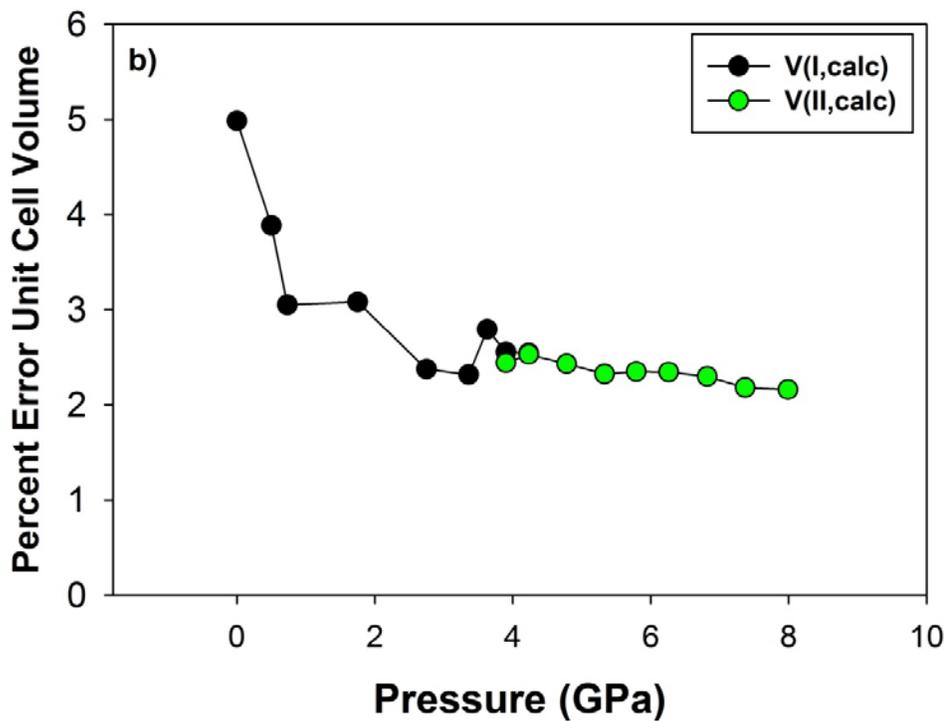
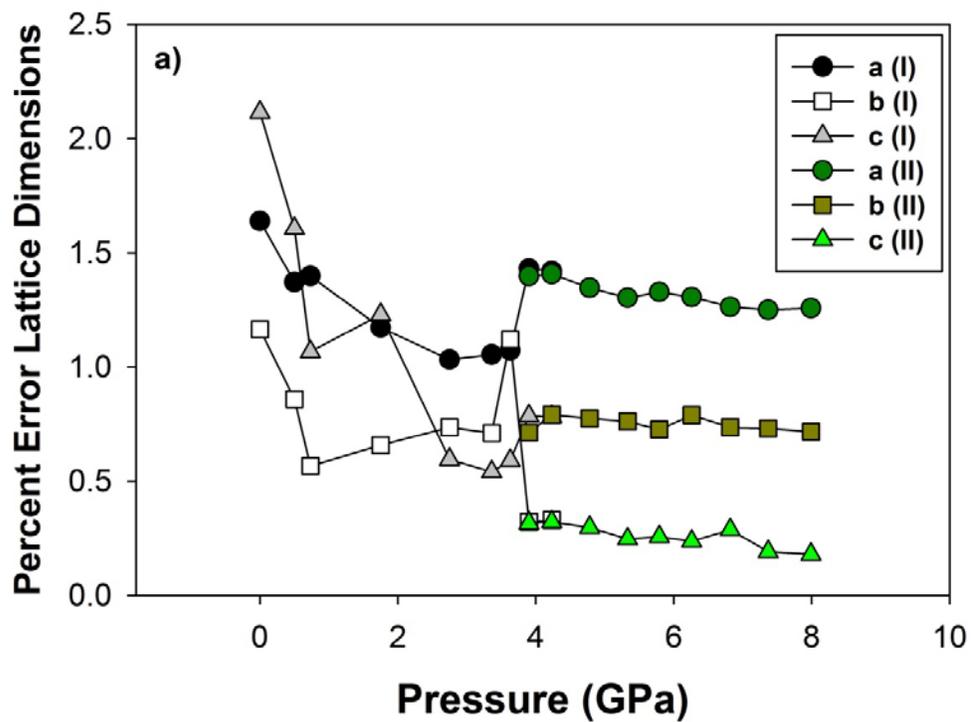


Figure S2. Variations in time (represented with gray lines) of the (a) unit cell volume, (b) temperature, (c) pressure and (d-f) unit cell lattice parameters during a BOMD trajectory in the NPT ensemble for α -RDX at ambient pressure and $T=300$ K. In each case the red line represents the running average of the respective quantity.



Figures S3. Variation with pressure of the percent error for (a) the predicted lattice dimensions and (b) unit cell volume for selected BOMD results relative to the available experimental data.¹⁻⁴

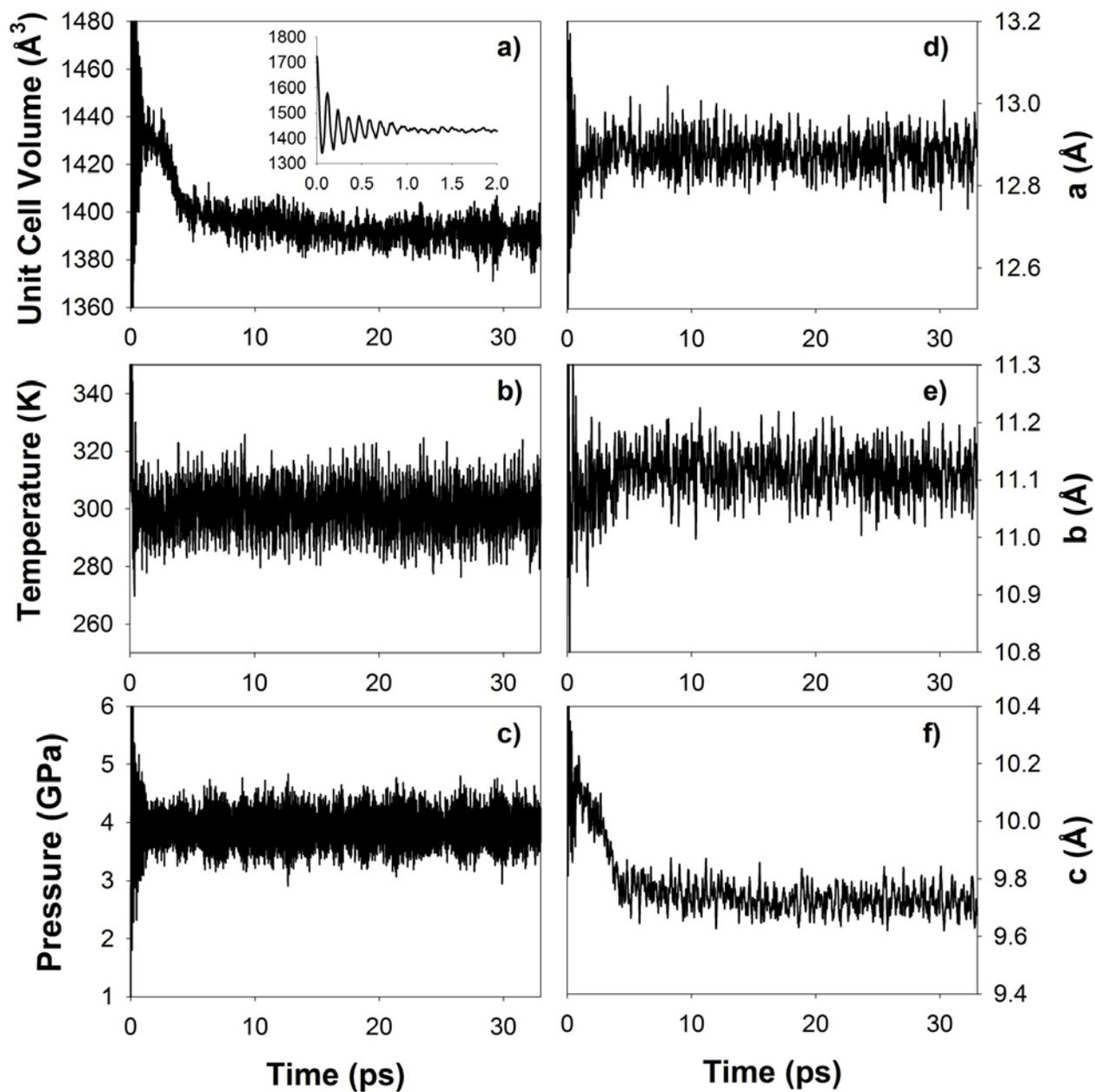


Figure S4. Variations in time of the unit cell volume, temperature, pressure and unit cell lattice parameters during a BOMD trajectory in the NPT ensemble for a trajectory at $P=3.9$ GPa and $T=300$ K initially started from the optimized configuration at ambient state of α -RDX. Detail of the unit cell volume variation in the first 2 ps is provided in the inset of panel a).

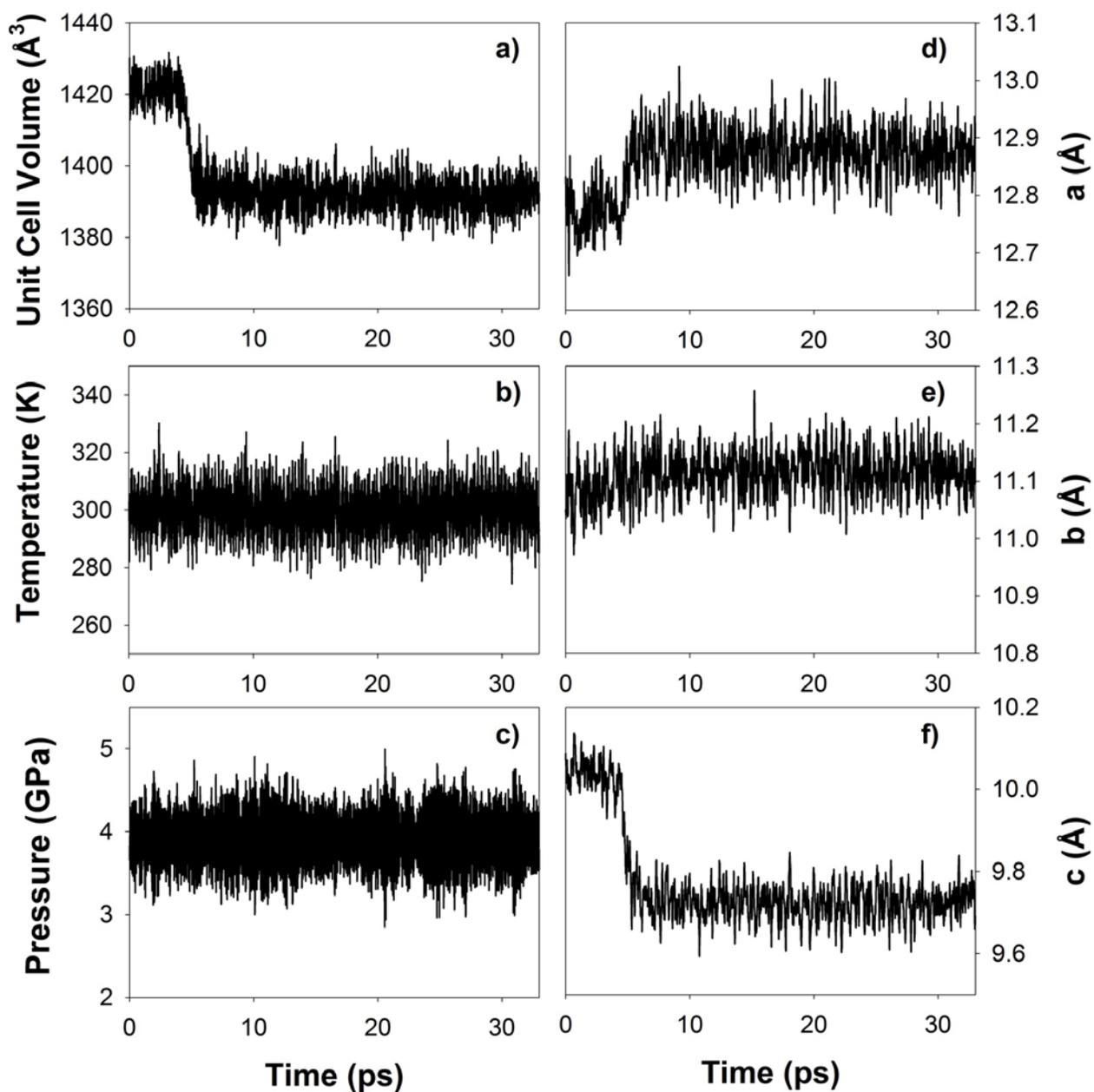


Figure S5. Variations in time of the unit cell volume, temperature, pressure and unit cell lattice parameters during a BOMD trajectory in the NPT ensemble for a trajectory at $P=3.9$ GPa and $T=300$ K initially started from and equilibrated configuration at $P=3.36$ GPa in the α -RDX phase.

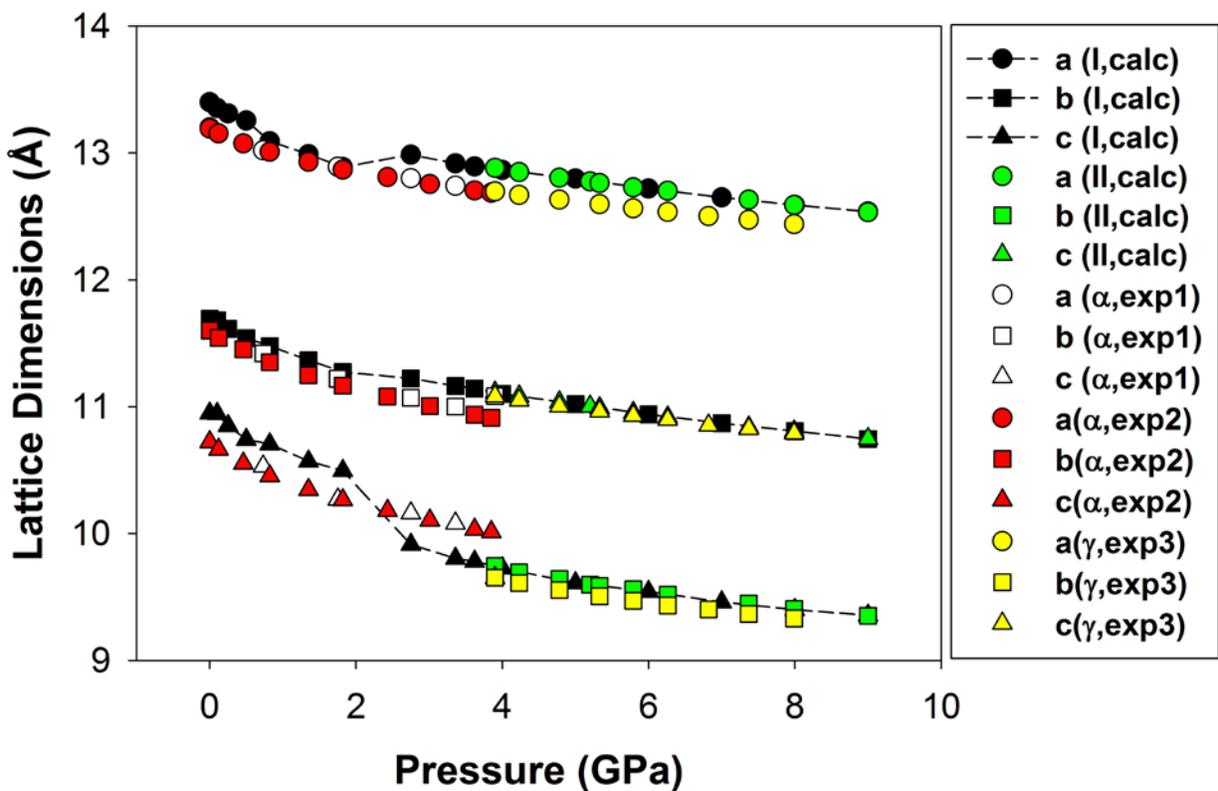
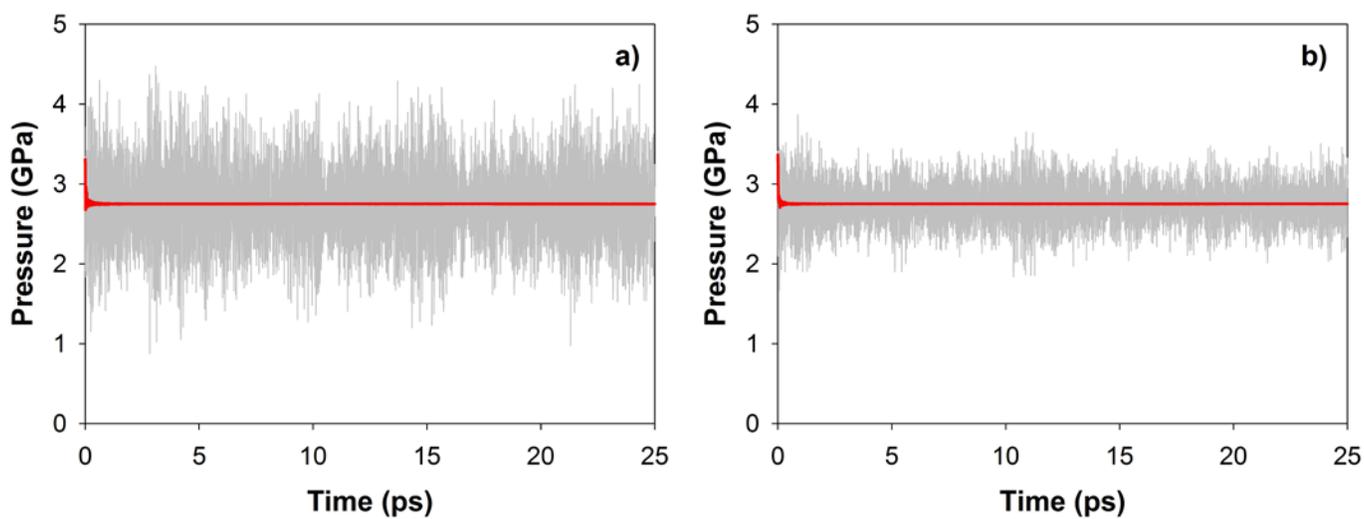
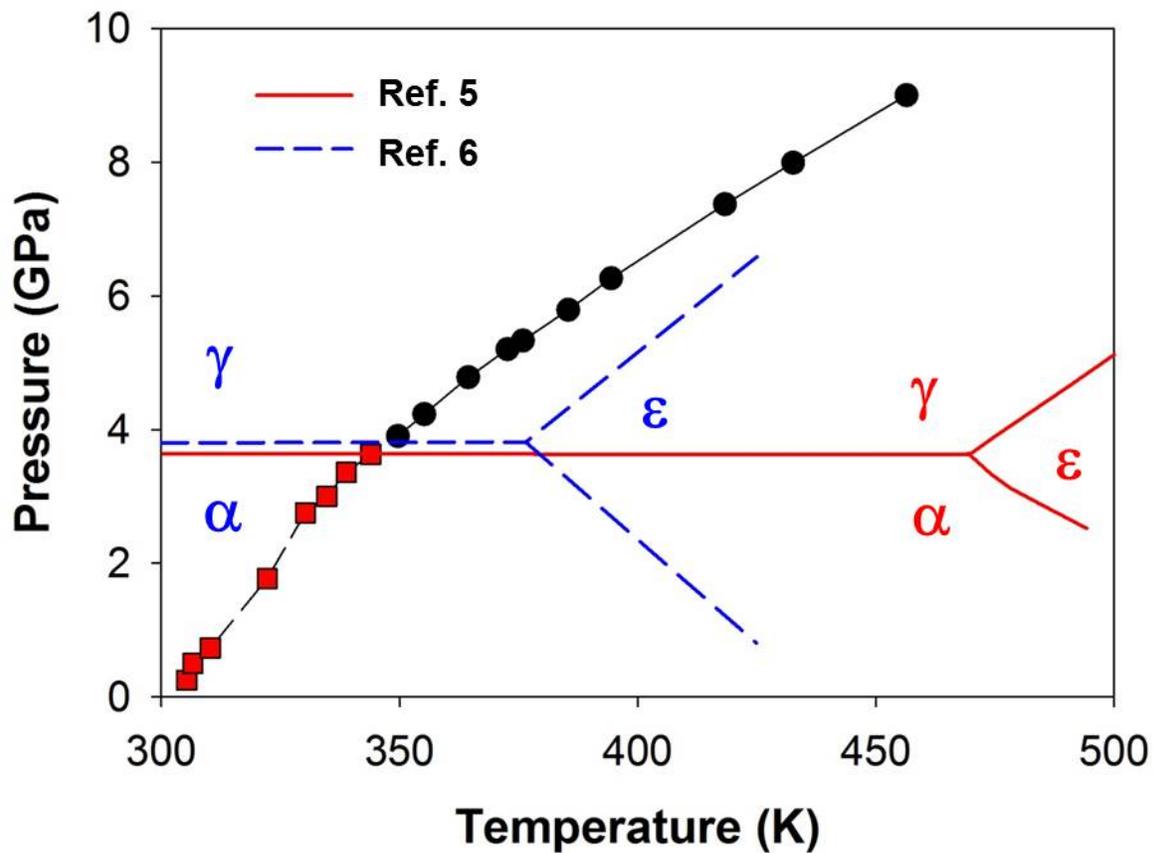


Figure S6. Variation with hydrostatic pressure of the predicted lattice dimensions of α -RDX and γ -RDX from BOMD calculations. A 1x1x2 supercell has been used in these simulations. The acronyms exp1, exp2 and exp3 refer to the corresponding experimental data from Refs. 2, 3 and 4. For the calculated data, the sets denoted with (I) and (II) indices correspond to crystal structures whose initial configurations are in the α - and γ -RDX phases, respectively.



Figures S7. Instantaneous values of the internal pressure in the RDX crystal (gray lines) during BOMD trajectories at $T=300$ K and $P=2.75$ GPa for the case of a simulation performed using (a) a $(1 \times 1 \times 2)$ supercell and (b) a $(2 \times 2 \times 2)$ supercell. The corresponding running averages are indicated by red lines.



Figures S8. The P-T Hugoniot calculated data overlaid on phase diagrams determined by Baer *et al.*⁵ and by Dreger and Gupta.⁶ Following the approach described in Ref.7 the data for the β phase in the original phase diagram from Baer *et al.*⁵ was reassigned to the ϵ phase.

References

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