


Modeling Bulk and Surface Characteristics of Cubic CeO_2 , Gd_2O_3 , and Gadolinium-Doped Ceria using a Partial Charge Framework — Supplementary Information

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Radial Distributions

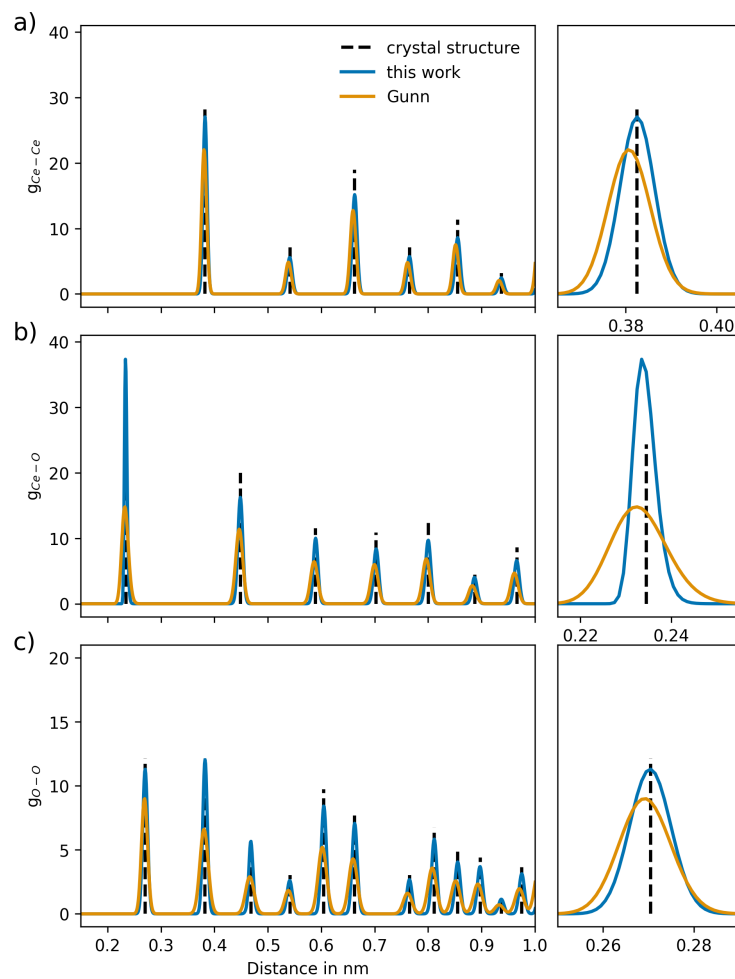


Figure S1: Comparison of the ion-ion pair distributions of CeO_2 at 298 K to the results found by using the potential of Gunn *et al.*

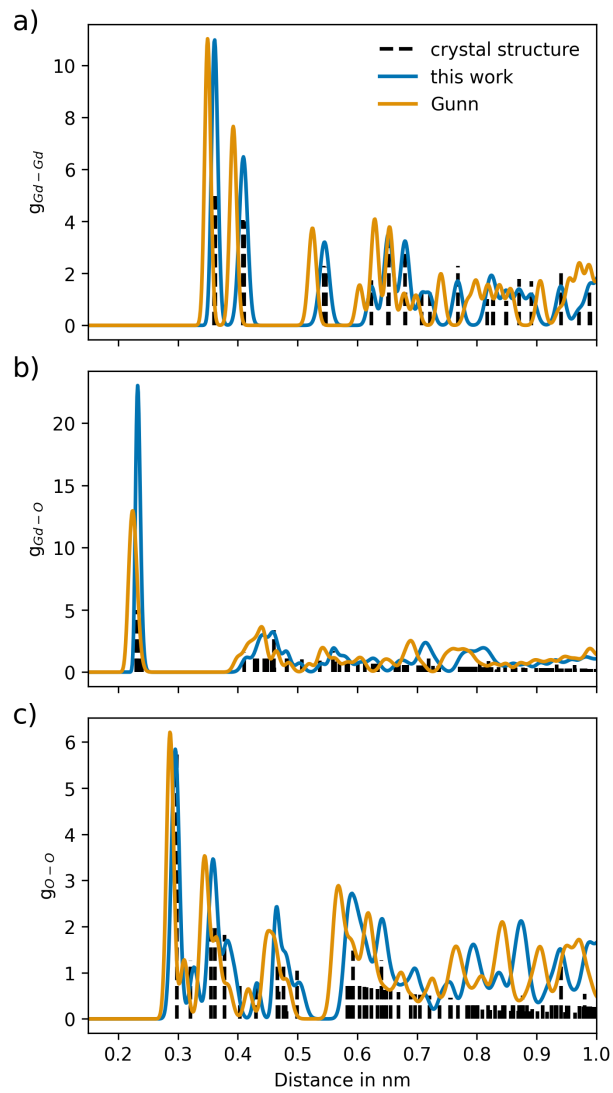


Figure S2: Comparison of the ion-ion pair distributions of Gd_2O_3 at 298 K to the results found by using the potential of Gunn *et al.*

Surface Properties

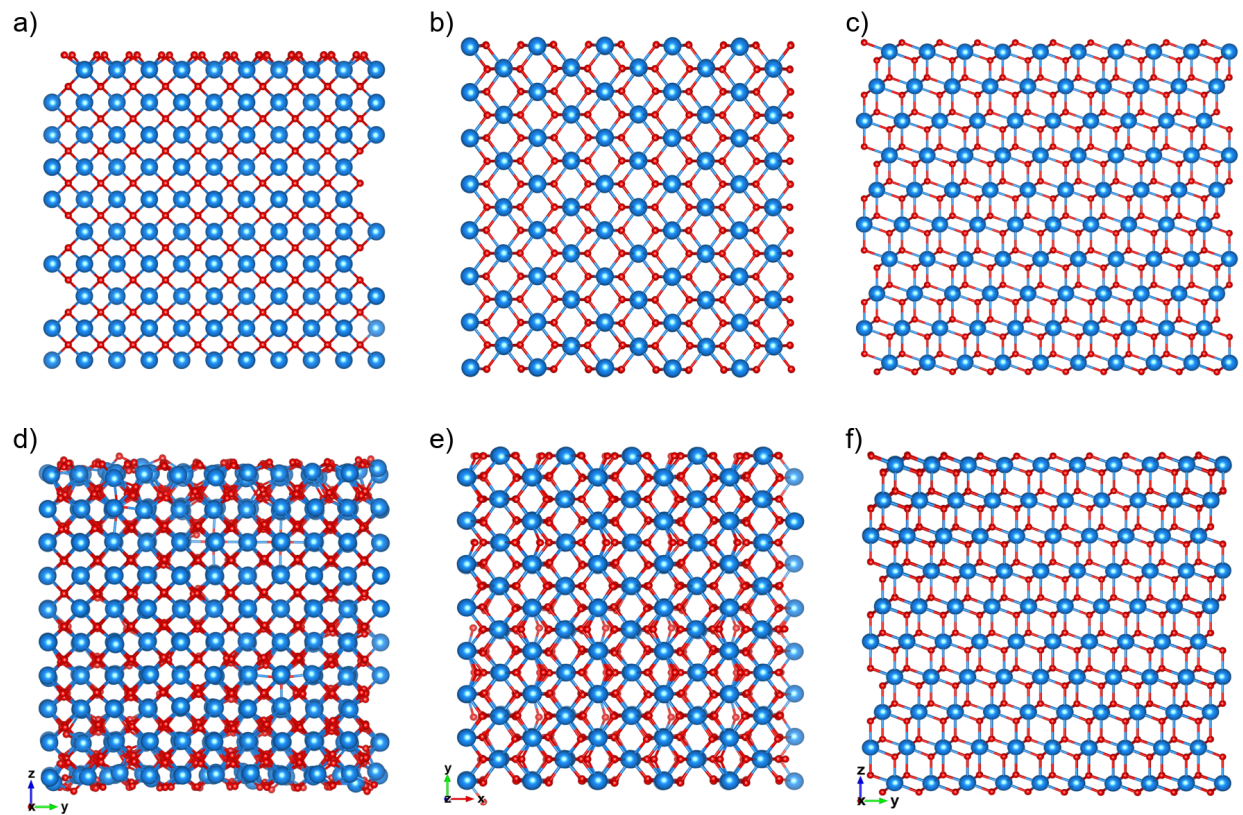


Figure S3: Comparison of the structure of a 2D-periodic CeO₂ (001), (101) and (111) model system determined *via* low temperature (10K) simulations using the partial charge model developed in this study (a, b and c) to results using the full-charge potentials by Gunn *et al.* (d, e and f) at 10 K under the same conditions. For a, c, d, and f the non-periodic direction is represented along the z-axis. For b and e the non-periodic direction is represented along the y-axis.

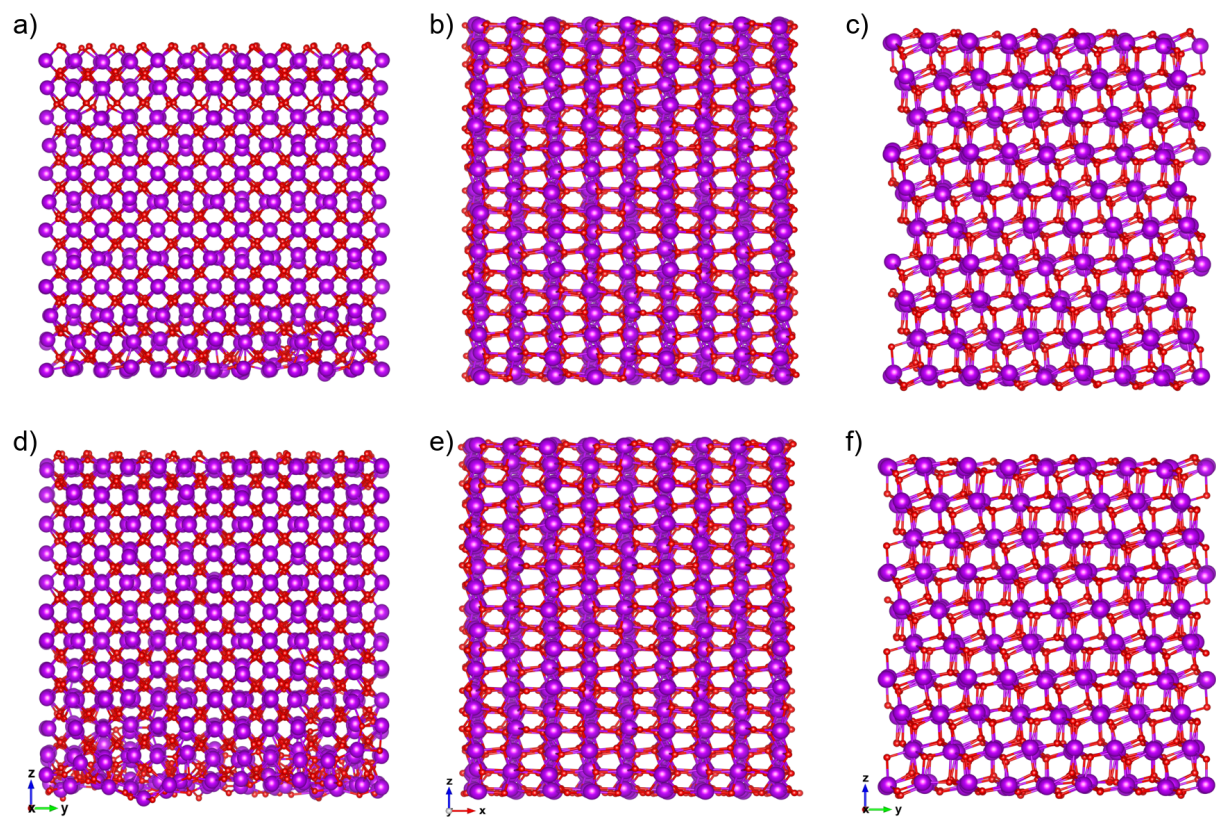


Figure S4: Comparison of the structure of a 2D-periodic $\text{Gd}_2\text{O}_3(001)$ and $\text{Gd}_2\text{O}_3(101)$ model system using the partial charge model developed in this study (a, b and c) to results using the full-charge potentials by Gunn *et al.* (d, e and f) at 10 K under the same conditions. For a, c, d, and f the non-periodic direction is represented along the z-axis. For b and e the non-periodic direction is represented along the y-axis.

Comparison of the Vibrational Behaviors

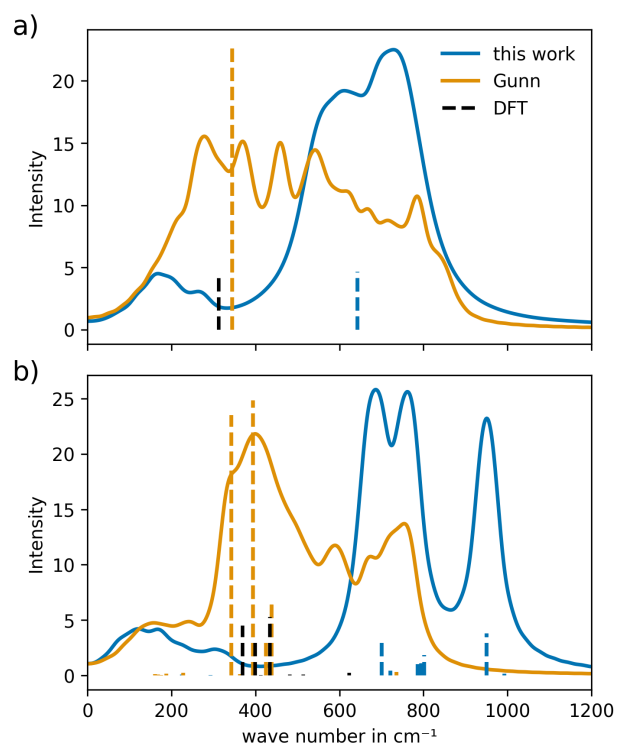


Figure S5: Comparison of the power spectra at 298 K to the results found by using a harmonic vibrational analysis for (a) CeO₂ and (b) Gd₂O₃, respectively.

Comparison of the M–O Potential

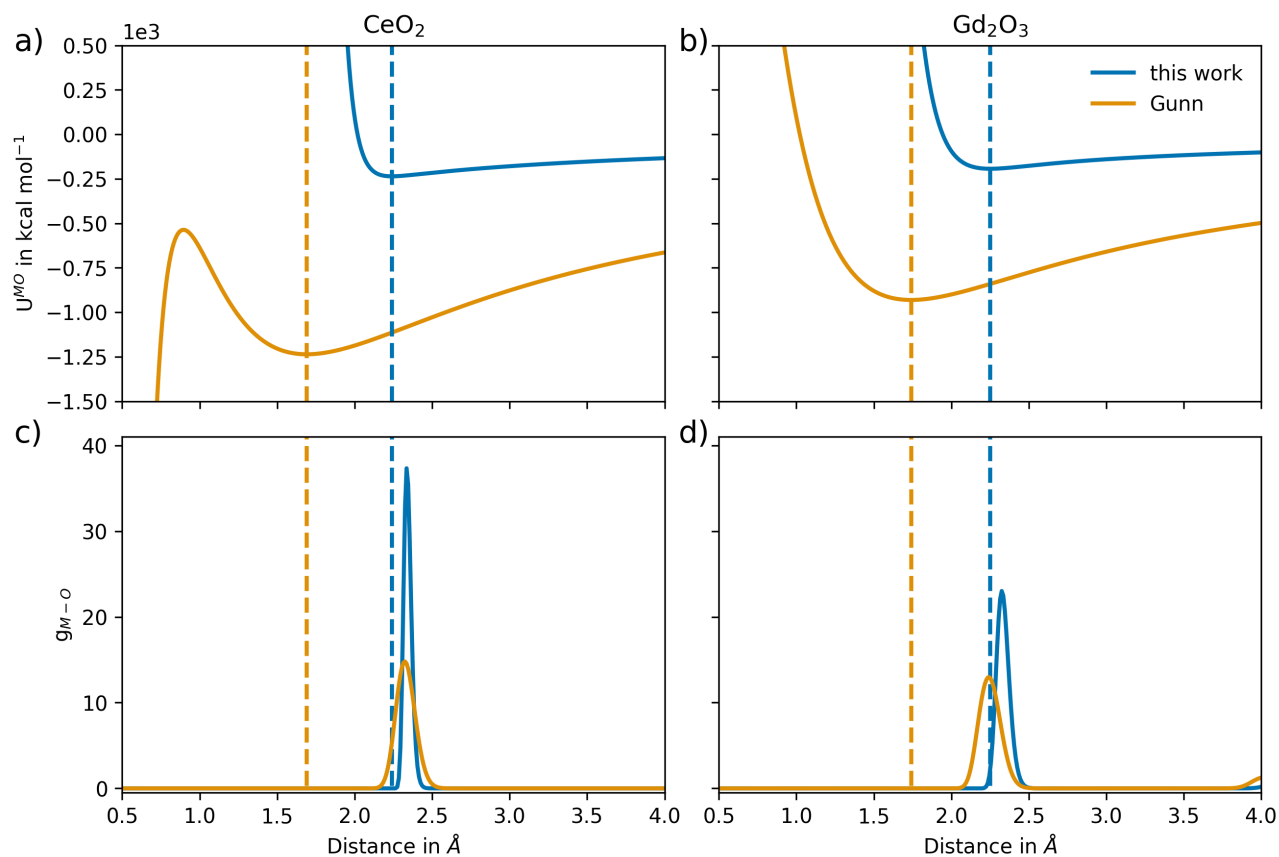


Figure S6: The potentials developed for a) Ce–O and b) Gd–O, and the M–O pair distributions of c) CeO₂ and d) Gd₂O₃ at 298 K. The potential of the novel potential is compared to the potential published by Gunn *et al.* The vertical dashed lines show the equilibrium distances of the respective M–O pair potential.

Comparison of the Computational Cost

Table S1 represents a comparison of the computational cost between the partial charge model developed in this study and respective ab-initio calculations. When comparing the molecular dynamics simulation, the partial charge model was applied to a 5x5x5 supercell of CeO₂ (1500 atoms), while the DFT calculation was used in conjunction with a 2x2x2 supercell cell (96 atoms). Molecular dynamics simulations are necessary for various property calculations, such as the radial distribution function (RDF), oxygen diffusion, and vibrational power spectra. The time comparison of vibrational analysis includes both geometry optimization and vibrational analysis. The vibrational analysis systems consist of a 4x4x4 supercell of CeO₂ containing 768 atoms for the partial charge model and a single unit cell (space group 225) containing 2 atoms for the DFT calculation. The table provides an estimate of the computational cost, as the actual computational cost depends on the specific hardware and settings used for the calculation. Performing property calculations with both methods would result in a similar ratio of computational cost. When comparing the computational cost of

Table S1: Comparison of the computational cost between the partial charge model developed in this study and ab-initio calculation using DFT.

Method	Partial Charge Model	DFT
Molecular Dynamics (0.1 ps)	3 10 ⁻⁵ h	24 h
Vibrational Analysis	0.05 h	54 h

the partial charge model to the DFT calculation, it is clear that the partial charge model is significantly faster. Furthermore, generating a vibrational power spectrum with DFT is much more time-consuming compared to a frequency calculation. This is because, in the former case, the calculation of the trajectory (20 ps) would take far longer than 54 hours. Therefore, a vibrational analysis is more appropriate for comparing the vibrational behavior of the partial charge model with the DFT calculation.