# Machine-Learning Simulations Predict Phase Transformations of Fullerite in a Diamond-Anvil Cell

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**Abstract**

Fullerite is a molecular solid of $C_{60}$ cages held together by van der Waals forces. Under ambient conditions the molecules sit on a face-centered cubic lattice and rotate freely. While the collapse of the $C_{60}$ cages under high pressure and temperature is well established, the precise structural pathways under non-hydrostatic conditions remain incompletely characterized. Here we report the use of Molecular dynamics simulations with a Machine-Learning Interatomic Potential (MLIP) [1] to study phase changes in fullerite through a compression and decompression process. Both uniaxial and hydrostatic conditions are used to mimic diamond anvil cell (DAC) experiments using the stuffed cell and the gas-loaded cell respectively. All structures are compressed up to 100 GPa and decompressed to mimic sample recovery from a DAC.

Our simulation results quantitatively agree with experiment, reproducing the pressure threshold at which transformations are seen in the DAC [2] and provide an atomistic picture of the transformation mechanism of fullerite into higher-density carbon allotropes. Pressure-induced cross-linking of the fullerene molecules starts at 1.3 GPa for uniaxial conditions and 1.6 GPa under hydrostatic conditions. Collapse of the $C_{60}$ cages into tetrahedral amorphous carbon occurs around 30 GPa under uniaxial load and 45 GPa under hydrostatic load in good agreement with experiments. The density and $sp^3$ fraction predicted by the MLIP also quantitatively agree with experimental electron energy loss spectroscopy measurements. In conclusion, our results confirm that MLIPs can capture the full pressure-driven transformation pathway of fullerite, from molecular cross-linking to the collapse into ultrahard amorphous carbon, with quantitative agreement to experiment, opening the door to new preparation protocols for the formation of dense carbon phases.

[1] Qamar, M., Mrovec, M., Lysogorskiy, Y., Bochkarev, A., & Drautz, R. (2023). Atomic cluster expansion for quantum-accurate large-scale simulations of carbon. *Journal of chemical theory and computation*, *19*(15), 5151-5167.

[2] Heimes, H., Turner, E. P., Salek, A. G., Wierbik, J., Huang, X., Dunstan, W. R., ... & Bradby, J. E. (2025). Mechanisms of nanodiamond and amorphous diamond-like carbon formation following room temperature compression of C60. *Diamond and Related Materials*, 112776.