The interplay between electron correlation and nuclear quantum effects makes our understanding of elemental hydrogen a formidable challenge. In this work, we compute the phase diagram of hydrogen and deuterium at low temperatures and high pressures (P > 300GPa) correctly accounting for accurate electronic correlations by quantum Monte Carlo, and for nuclear quantum effects and anharmonicity by the self-consistent harmonic approximation. We predict that phase III transits into the molecular metallic phase VI at 412(+/- 40) GPa, then to the atomic phase at pressures as large as 577(+/- 10) GPa. The transition pressures increase by 30 GPa and 63 GPa, respectively, if hydrogen is replaced with deuterium. Our results strongly support the experimental claim of a phase transition occurring at about 425 GPa into a metallic non-superconducting phase. However, for the atomic phase that is expected to host room temperature superconductivity, our data suggest much larger transition pressures than those predicted theoretically or attained so far experimentally.

In order to achieve these predictions, we had to carry out quantum Monte Carlo simulations for the electronic part that required a very accurate extrapolation to the thermodynamic limit, particularly sensitive in the metallic phases. We therefore performed calculations of very large supercell, containing up to 1024 atoms. To reach this size, we exploited the recently implemented GPU-compliant version of the TurboRVB quantum Monte Carlo code, which can harness the improved computational power with an almost-order-of-magnitude speed-up in large systems when GPU-accelerated cores are used.