

## Quantum Chemical Simulation of Materials Phase Transition Under High Pressure and Temperature Conditions

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The combination of high pressure and high temperature enables the observation of phase-transition behavior in materials otherwise inaccessible under ambient conditions. Individually, increasing pressure and temperature often have opposite effects on materials properties, and the exact nature and details of the thermal-mechanical coupling necessary for chemical transformations to take place remain poorly understood and difficult to decipher experimentally. We developed an innovative quantum chemical simulation approach to address this problem, which we applied to investigate the graphitic-boron nitride (BN) transition to cubic-BN. The phase transition is found to be favorable at a pressure of 10 GPa and a temperature of 2060 K for cubic-BN, which is in good agreement with experimental reports. Our simulations unveil the BN phase transition reactive coordinate and reaction trajectories in unprecedented structural detail, yielding reaction profiles necessary to examine thermal-mechanical coupling effects. As pressure compresses the potential well of phonon modes, and enables the system to absorb a higher degree of thermal energy, it indirectly confers a specific phonon mode, with atomic displacements normal to the graphitic-BN layer, significant anharmonic behavior. The atomic vibration pattern of this specific phonon mode allows efficient transfer of thermal energy to chemical bonds and drives bond breaking and rearrangement. The underlying reaction mechanism of solid-state phase transition upon thermal-mechanical loading is discussed in terms of compression effects on phonon modes, and how thermal energy transfer to specific modes can initiate chemical reactions, in an attempt to bridge the important knowledge gap between high-pressure, high temperature chemistry and mode-selective chemical synthesis.