X-Ray Pump-Probe Study of Diamond Formation with and without Color Centers

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Abstract

Renowned for its exceptional hardness and unique properties, diamond holds immense promise for diverse technological applications spanning mechanics, electronics, and photonics. Color centers, impurities formed by vacancies in the diamond lattice, introduce additional electronic states within the wide bandgap, making diamond suitable for quantum technology applications. In particular, with negatively charged silicon-vacancy (SiV) centers are emerging as promising single-photon emitters due to their narrow zero-phonon line transition at 738 nm. [1] An efficient method to synthesize diamond, with or without SiV centers, is the high-pressure high-temperature (HPHT) method. Identifying ideal precursors and understanding the kinetics of the phase transitions and the Si incorporation mechanism in the lattice are essential to fully exploit this method. [2] Diamondoids, consisting of hydrogen-terminated carbon cages superimposable on the diamond lattice, emerge as promising candidates for synthesizing high-quality diamond and exhibit lower pressure–temperature (P–T) thresholds compared to other hydrocarbons for diamond formation. [3] Moreover, adamantane, the smallest diamondoid with the chemical formula $C_{10}H_{16}$, can be functionalized with Si atoms (AdaSi) and used as a precursor for SiV center formation. [4] Despite these advances, the time-resolved structural dynamics of diamond and color center formation remain underexplored.

We recently performed time-resolved X-ray pump–probe experiments on compressed diamondoids utilizing 2.2 MHz X-ray pulse trains at the High Energy Density (HED) instrument of the European X-ray Free-Electron Laser Facility (EuXFEL).[5] We heated pure diamondoids, including adamantane, and an Si-functionalized AdaSi sample in a diamond anvil cell using trains of 200 ultrashort X-ray pulses, each 25 fs in duration with a 443 ns interpulse spacing, delivering high pulse energy and brightness. Submicrosecond temporal resolution X-ray diffraction (XRD) patterns revealed the formation dynamics and conditions of diamond across five samples at high pressures ranging from 13 to 20 GPa. The fastest diamond nucleation was observed in pure triamantane, where diamond was detected at around 30 µs at 19 GPa. We also determined energy and energy-delivery-rate thresholds for SiV formation in AdaSi samples, which show a strong pressure dependence, requiring higher energies at lower pressures. Post-recovery XRD and Raman spectroscopy revealed various carbon-based products, including graphite and amorphous carbon, while photoluminescence confirmed the presence of SiV centers. This study provides an ultrafast, *in situ* perspective on diamondoid-to-diamond conversion and SiV center incorporation, offering insights for developing efficient synthesis routes for color centers targeting a wide range of technological applications.

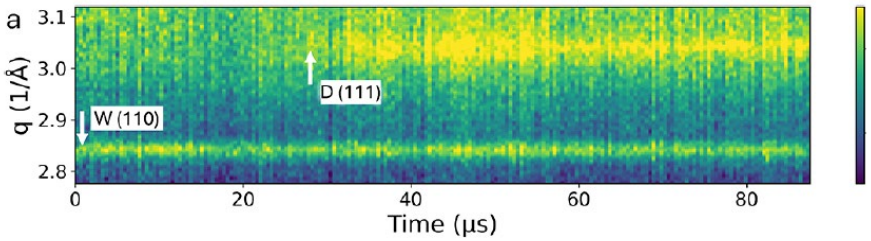

Figure 1. Batch view of 200 XRD patterns from the pulsed X-ray train with a 443 ns interpulse spacing, showing the emergence of the diamond (111) peak at approximately 27 *µ*s (run 605). The onset of the diamond (111) peak is labeled “D (111)” and the tungsten (110) peak is labeled “W (110)”, with arrows indicating both. [5]

[1] L. Rogers et al., *Nat Commun* **5**, 4739 (2014).

[2] H. Yang, Y. Ma, and Y. Dai, *Funct. Diam.*, **1** (1), 150–159 (2022)

[3] S. Park et al., *Sci. Adv.*, **6** (8), p. eaay9405(2020)

[4] Y.K. Tzeng et al., *Nat Commun* **15**, 7251 (2024)

[5] M. Han, A. Celeste et al., *JACS*, accepted (2026)