Nitrogen aggregates in yellow HPHT and CVD synthetic diamonds.

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The identification of natural and synthetic diamonds is an important gemmological task. Structural impurities in diamonds play a critical role in this task because they characterize the diamond growth and post-growth conditions, which differ between natural and synthetic diamonds. Optical centers in yellow diamonds partially overlap with those in colorless diamonds but also exhibit distinct differences. In recent years, there has been a growing number of synthetic yellow diamonds on the market, many of which are irradiated and annealed to alter their color. As a result, gemmological laboratories face challenges in the identification of yellow diamonds.

We studied synthetic yellow diamonds of both HPHT and CVD origin using a combination of spectral methods. Since 2023, the N3 center (415 nm) - previously considered an indicator of natural diamonds - has been observed in synthetic diamonds. In natural diamonds, the N3 center can be detected in both absorption and photoluminescence (PL) spectra, whereas in synthetic diamonds, its peak is weaker and observable only in PL spectra. Beginning in 2024, the N3 center has become common in synthetic yellow diamonds submitted by clients, with a more distinct peak in HPHT samples (Fig. 1a) and a weaker peak in CVD samples (Fig. 2). In natural diamonds, the N3 center arises from nitrogen aggregation during prolonged geological processes, while in synthetic diamonds, it can form during post-growth treatments (Vins *et al*., 2021).

In addition to the N3 center, a prominent PL series with zero-phonon lines (ZPLs) at 523.6 nm and 626.3 nm was detected in all HPHT yellow diamonds (Fig. 1a). These optical centers form in type Ib diamonds after irradiation and annealing (Dobrinets *et al*., 2013). The 580 nm center is attributed to Ni-related defects (Yelisseyev and Kanda, 2007).

FTIR spectroscopy revealed that the HPHT yellow diamonds are type Ia, with nitrogen detected in the 1100–1300 cm^{-1} range. One sample exhibited a platelet-related peak at 1364 cm^{-1}, a feature typically associated with natural post-growth annealing. Two synthetic yellow CVD diamonds (1.03 ct, cushion-cut) submitted by clients showed weak nitrogen-related peaks at 1250–1380 cm^{-1} and hydrogen-related peaks at 3107 cm^{-1} in FTIR spectra. Their PL spectra displayed weak N3 and H3 centers, along with a $[Si\text{-}V]^{-1}$ doublet at 736,6/736,9 nm (Fig. 2).

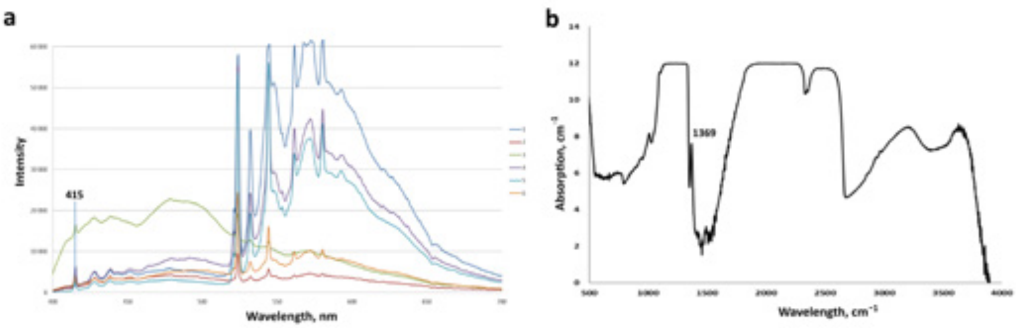


Fig. 1. a – PL spectra of 6 yellow synthetic HPHT diamonds, recorded at liquid nitrogen temperature. Excitation 365 nm. b – FTIR spectrum of yellow HPHT synthetic diamond with B2 (platelets) peak.

Experimental Stage

To investigate nitrogen aggregation mechanisms, we conducted post-growth experiments on three CVD diamond plates (0.15 ct, 0.12 ct, and 0.65 ct) and one HPHT diamond plate (6×6×1 mm, 0.86 ct). The samples were electron-irradiated (3 MeV) and subsequently annealed at 1750–1800°C.

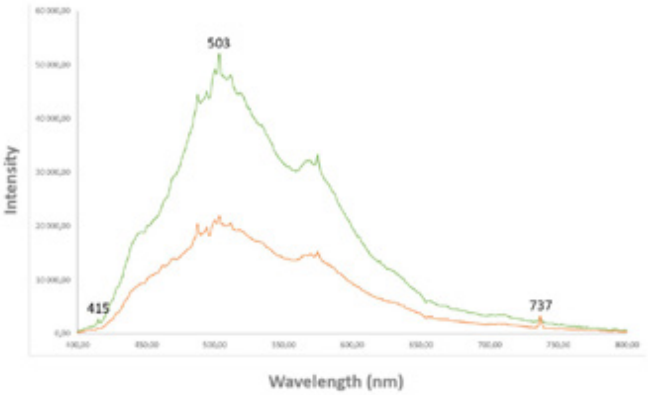
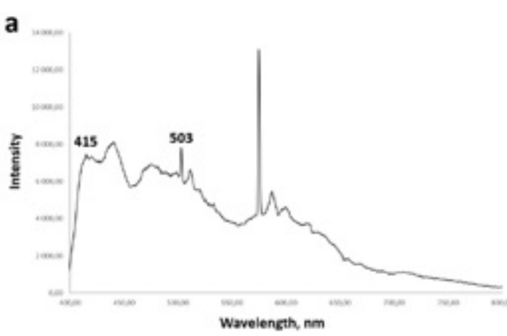


Fig. 2. PL spectra of two yellow CVD diamonds recorded at liquid nitrogen temperature with excitation 365 nm. There are peaks at 415 nm, 503 nm, 575 nm, and 737 nm.

Spectral characterization was performed before and after each stage. After irradiation, vacancies or GR1 centers (741 nm) appeared in the spectra. Following annealing, the N3 center emerged in CVD plates in PL spectra recorded at liquid nitrogen temperature. In annealed CVD plates, apart from 415 nm, additional 503 nm, 575 nm, and 737 nm peaks appear in Pl spectra recorded at liquid nitrogen temperature (Fig. 3a). Same peaks are recorded at polished CVD diamonds (see Fig. 2). In the annealed HPHT plate we did not observe the N3 center in PL spectra (Fig. 3 b) but see the H3 center (503 nm) that appeared after annealing. In the FTIR spectrum of the annealed HPHT plate there are A and C centers recorded as well as weak B2 (platelets) peaks. Probably further annealing is required for this sample in order to induce N3 center.

Our experiments confirm that the N3 center forms in synthetic diamonds after irradiation and annealing. The proposed mechanism involves:

1. As grown stage: Single nitrogen atoms are present (more abundant in HPHT, less in CVD diamonds).
2. Irradiation: Generates vacancies in the diamond lattice.
3. Annealing: Facilitates the formation of H3 aggregates, a fraction of which evolve into N3 aggregates.



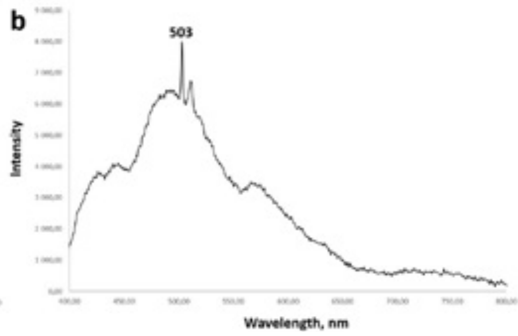


Fig. 3. PL spectra of synthetic CVD (a) and HPHT diamond (b) plates after irradiation and annealing.

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