Modification of photoluminescent nitrogen optical centers by femtosecond laser pulses in treated natural diamond

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Direct laser writing by focused ultrashort laser pulses enables local modification of color centers in diamonds with excellent three-dimensional positioning accuracy, providing unique capabilities for quantum technologies, magnetometry, laser generation and photoluminescence (PL) micromarking. To shed light on currently unknown underlying atomistic transformations, we exposed electron-irradiated and annealed natural IaAB-type diamond with an initially high content of various nitrogen-vacancy centers (NV, H3, N3 and H4) to ps, 525-nm laser pulses focused inside the sample. The modified areas were characterized by Fourier-transform infrared (FT-IR) and ultraviolet(UV)-visible transmission spectroscopy, as well as confocal Raman and PL microspectroscopy with excitation wavelengths of 405 nm and 532 nm. We report on a local decrease in originally strong PL yield of NV, H3 and H4 centers with a monotonic dependence on both pulse energy and exposure time, as well as a rise of H1b and H1c lines in the FT-IR spectra accompanied by a drop in the concentration of H1a center. The results indicate that laser-induced processes make possible both creation and destruction of NV, H3 and H4 centers influenced by their initial concentration and the parameters of laser modification, which complements known results and can be useful for adjusting of laser writing regimes for PL micromark inscription or local color modification.