Instantaneous in situ Raman probing of femtosecond laser-excited hot electrons, optical phonons and atomistic damage dynamics in diamond

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Ib-diamond plate with substitutional atomic nitrogen C-centers was exposed in its bulk by tightly focused 0.2-ps, 525-nm laser pulses. Stokes Raman scattering line related to 1332-cm^{-1} optical phonon was observed in the transmitted laser spectra. Its intensity demonstrates second-order dependence on laser pulse energy due to three-photon interband absorption, transforming at higher energies into a linear dependence for saturated absorption of dense electron-hole plasma. Raman scattering was related to a non-resonant spontaneous process, strongly enhanced by plasma emission of seeding optical phonons. Raman peak position wavenumber decreases versus pulse energy via photo-injection of plasma and then slowly increases due to the "electronic" stress in the near-critical plasma, until its final drop upon the onset of self-accelerated decay of optical phonons into acoustic ones (quasi-heating). Peak half-width monotonously increases versus pulse energy due to the self-accelerating optical-phonon decay during the 0.2-ps laser pulse. Structural conversion into NV- and other nitrogen-related centers becomes strongly enhanced in the quasi-heating regime. At the higher pulse energies all Raman lines disappeared along with the simultaneous photoluminescence intensity saturation for the novel color centers, demonstrating for the first time, the intriguing ultrafast "non-thermal" structural disordering in diamond.

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