Ultrafast non-linear hybrid mid-IR laser photoionization and micromarking of immersed diamond

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Direct interband and intragap photoexcitation by intense mid-infrared (MIR, 4.0 and 4.7 microns) femtosecond (fs) laser pulses was explored in ultrapure chemical-vapor deposited (CVD) diamond via acquisition of characteristic UV photoluminescence (PL) of free excitons and A-band PL of electrons anchored at deep donor-acceptor or dislocation-related traps, respectively. At lower laser intensities (¡ 10 TW/cm2) the excitonic PL yields exhibit highly non-linear dependences with (intensity x lambda2)-scaling and power slopes N=17 (4.0 microns) and 14 (4.7 microns), still insufficient to cross over the direct bandgap (6.5 eV) by 1.2 and 2.8 eV, respectively. Similarly high slope of 9 (4.7 microns for intragap (3.5 eV) photo-population of donor-acceptor traps is still insufficient for their direct excitation by 1 eV. At the intermediate (intensity x lambda2)-dependent values of the Keldysh parameter "gamma" about 1 such incomplete multi-photon excitation anticipates the hybrid total "multiphoton+tunneling" photoexcitation generally predicted by the Keldysh theory, but never unambiguously experimentally demonstrated. At higher laser intensities (¿ 10 TW/cm2) both the excitonic and A-band PL yields exhibit (sub)linear slopes, apparently, indicating formation of more strongly absorbing electron-hole plasma. These findings shed light on the hybrid "multiphoton+tunneling" character of Keldysh photoexcitation at intermediate values "gamma" and pave the way to defect/impurity band engineering of intragap non-linear optical properties in bulk dielectrics for their precise fs-laser nanomodification through IR-transparent low-melting solid-phase immersion.