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Ultrafast dynamics of photoexcited free carriers in CVD diamonds

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Abstract. We report on the experimental studies of the free-carrier dynamics and nonlinear optical properties in both bulk polycrystalline and single-crystal chemical vapor deposition (CVD) diamonds. The dynamics of nonlinear refraction and nonlinear absorption is measured by use of a time-resolved femtosecond pump-probe technique under UV excitation. Nonlinear refraction dynamics indicates a positive Kerr effect. Slow and fast components in nonlinear absorption transmittance are separated and used to determine the free-carrier lifetimes.

1. Introduction

High-purity diamond crystals have received much attention [1,2] owing to their potential applications in semiconductor physics and optoelectronics. Due to their transparency over a broad wavelength range from the UV to IR and to their excellent thermal conductivity, diamonds have become one of the major window materials. However, in the study of inertial confinement fusion (ICF) the high-intensity drive laser and laser plasma-induced X-ray can modify the optical properties of the window materials composed of diamonds, which leads to the changes in the strength and phase of the probe beam in diagnostic apparatuses and therefore to the increased uncertainty in the experimental results. Therefore, it is of crucial importance to investigate the ultrafast dynamics of the free carriers and nonlinear optical properties in bulk diamond.

In this work, we investigate the free-carrier dynamics and nonlinear optical properties in both bulk polycrystalline and single-crystal chemical vapor deposition (CVD) diamonds by use of a time-resolved femtosecond pump-probe technique employing a phase object. The dynamics of nonlinear absorption and refraction is determined by the differential transmittance in open- and closed-aperture configuration excited by the UV laser, respectively.

2. Samples and experimental setup

Both bulk polycrystalline and single-crystal diamonds of high purity were studied in this paper. Diamonds were prepared by Element Six, UK employing chemical vapor deposition method. We used a modified pump-probe technique with a phase object (PO pump-probe technique) [3] to measure the dynamics of transmittance changes. A mode-locked Yb:KGW-based femtosecond laser (515 nm, 190 fs, 20 Hz) was divided into a pump and a probe beams. An optical parametric amplification (OPA) system was employed to change the pump beam to the UV light. A PO, which consisted of a glass plate with a transparent dielectric disc deposited on it, was introduced into the probe beam. The pump and probe beams were focused on the same point of the sample. After passing through the sample, the probe beam was divided into two beams. A finite aperture was placed in front of one detector and its radius in the far field was adjusted to be equal to that of diffracted beam of the PO. Energy change of the probe beam was recorded as a function of delay time between the pump and probe beam.

3. Experimental results

Figures 1(a)-1(c) show the normalized transmittances in open-aperture configuration as a function of delay time for polycrystalline diamond. Transmittance changes indicate that nonlinear absorption includes the non-degenerate two-photon absorption (ND-TPA) and free-carrier absorption [4]. The ND-TPA dominates near the zero delay and the free-carrier absorption decays slowly with a fast (80 ps) and a slow (3-8 ns) components. A sharp peak in the vicinity of zero time delay for a closed-aperture configuration in Fig. 1(d) can be ascribed to a positive nonlinear refraction due to the Kerr effect. The FWHM of the trace is 0.3 ps, which is associated with the pulse width. Similar results are obtained for other excitation densities. The nonlinear refractive index of the diamond is extracted from the fitting to be $(1.4 \pm 0.2) \times 10^{-15}$ cm^2/W. Moreover, under high energy density the negative nonlinear refraction due to the free carriers will be observed at large time delay.

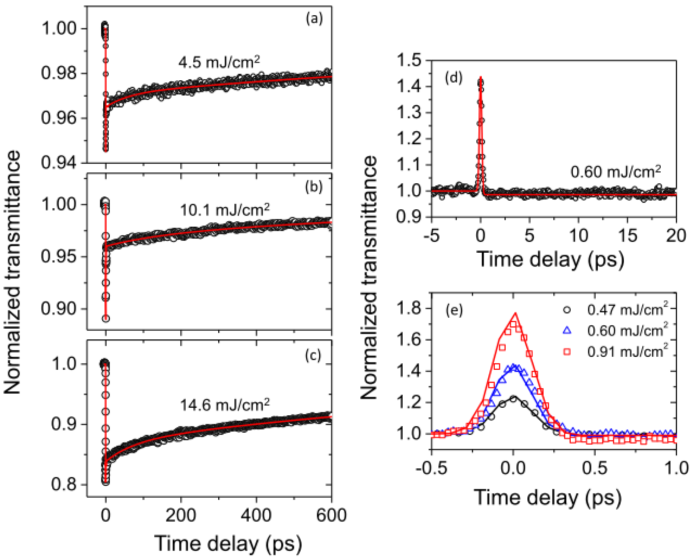

Fig. 1. Dynamics of (a)-(c) nonlinear absorption and (d)-(e) nonlinear refraction for the polycrystalline diamond under 351 nm excitation.

In addition, the transmittance changes in open-aperture configuration ranging from 480 to 680 nm are also investigated for both polycrystalline and single-crystal diamonds. The detailed results will be discussed somewhere else.

4. Conclusion

The optical nonlinearities and ultrafast free-carrier dynamics in both bulk polycrystalline and single-crystal CVD diamonds have been investigated by the PO pump-probe technique. We have found that both ND-TPA process and Kerr nonlinearity are positive. The free-carrier absorption (refraction) induced by the ND-TPA process is positive (negative). The decay of free-carrier absorption is relatively slow. This work will enable us to gain a better understanding of ultrafast dynamics of carriers and the nonlinearities in diamonds and therefore is beneficial to their applications in optics.

References

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