Unambiguous Evidence of Two Plasmon Decay During Ultrafast Laser Writing in Glass

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The interaction of femtosecond laser pulses with transparent media has been a focus of research due to its unique properties. It has been established that above a certain threshold, self-assembled nanogratings in silica glass can be induced [1]. Although the mechanism that triggers the nanostructure is still unclear, a theory has been introduced involving the mechanism of nanogratings formation based on two plasmon parametric decay [2]. A signature of two-plasmon decay is the generation of the 3/2 harmonic. Previously, in transparent media, only a weak 3ω/2 emission was observed at high energy thresholds [3]. Thus it remains unclear if two plasmon decay can be associated with self-assembled nanogratings formation. Here we present a thorough survey of 2^{nd}, 3^{rd} and 3/2 harmonic generations in fused silica for varying laser fluences within multiple regimes of optical laser writing and self-assembled nanostructures. We demonstrate that 3ω/2 can be observed at the energies close to the threshold of permanent material modification.

Regeneratively amplified, mode-locked Yb:KGW based femtosecond laser system (Pharos, Light Conversion Ltd.) operating at 1030 nm and delivering pulses of ~300 fs at varying repetition rates was used in the experiments. The laser beam was focused into the bulk of the silica via a ×10 (NA = 0.16) aspheric lens. A series of spectral measurements for varying pulse energy were taken translating the sample and at stationary positions and analysed with an Andor Shamrock SR-303i imaging spectrometer, where two gratings were used to vary the spectral range of the measurement (150 and 1200 lines/mm).

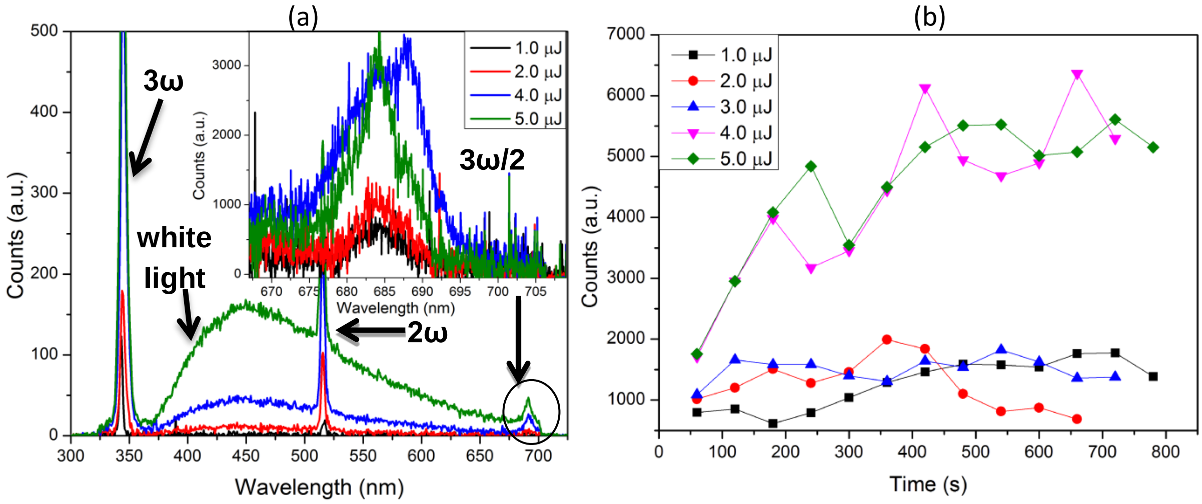

Fig. 1 a) Spectrum of harmonics for varying pulse energies (200 kHz repetition rate). White light emission and the double peak structure for 3ω/2 are observed as pulse energy is increased. b) Time dependence of 3ω/2 for varying pulse energies.

Analysing the light collected after the sample, we observed three distinctive peaks, corresponding to second, and third harmonics and 3ω/2. The spectrum measured for varying pulse energies revealed that 3ω/2 could be observed just above the permanent modification threshold, with a characteristic two-peak structure associated with forward and backward propagating plasmons and a bandwidth of ~20 nm (Fig. 1a). All three harmonics were increasing with pulse energy and evolved on the time scale of several minutes (Fig. 1b). The amplitudes seen for 3ω/2 are constant for low pulse energies where self-assembled nanogratings are formed indicating that two plasmon decay can play an important role in the formation of nanostructures. At higher pulse energies where strong damage occurs, the signal becomes erratic. Additionally, the observed emission of the three halves harmonic clarifies a long standing issue on electron concentration values in ultrafast laser material modification regimes. Since the two plasmon decay is possible for the electron concentration $n_e=n_{cr}/4$ [2,3], where n_{cr} is the critical plasma density, we could state electron concentration is at least 4×10^{20} cm^{-3}.

References

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