Supercontinuum assisted trapped electron accumulation in titanium oxide gel by femtosecond laser pulses

A. I. Kuznetsov[1], A. Kanaev[2], and N. Bityurin[1*]

[1] *Institute of Applied Physics RAS, 603950, Nizhniy Novgorod, Russia,*
[2] *LIMHP C.N.R.S., Institut Galilée, Univerité Paris-Nord, 93430 Villetaneuse, France*
bit@ufp.appl.sci-nnov.ru

Abstract: Long-lived trapped electrons are generated in titanium oxide gels in a convergent part of a loosely focused beam from a millijoule, femtosecond Ti:Sapphire laser. The appearance of trapped electrons is accompanied by the generation of the white supercontinuum. Considering the relevant physical mechanisms of trapped electron accumulation, we show that two-photon interband transitions with participation of photons at the fundamental frequency and from the blue part of the supercontinuum explain the experimental findings.

OCIS codes: (320.7110) Ultrafast nonlinear optics; (140.3390) Laser materials processing

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1. Introduction

In media where the refractive index has a linear dependence on laser intensity, $n = n_0 + n_2 I$ (Kerr nonlinearity), the laser beam becomes unstable when its power exceeds a certain critical power $P > P_{cr}$ [1]. This instability results in beam filamentation. Multiple filamentation of almost collimated powerful IR femtosecond beams propagating in methanol and in water was investigated in Refs. [2-4]. The filamentation is accompanied by a strong spectral broadening of the incoming laser radiation. The symmetrical lineshape corresponding to the self-modulation of a laser pulse caused by stationary Kerr nonlinearity, evolves into an asymmetric spectrum of white light. Recently, the interrelations between optical breakdown and filamentation of femtosecond beams in gases, liquids, and solids have been thoroughly investigated [5-10]. In contrast to the breakdown-induced light, which is irradiated from the modified area isotropically, the white light generated through filamentation propagates in the direction of the incoming radiation, forming a conical emission. One of the causes of this supercontinuum (SC) generation is the self-modulation of the laser pulse upon the fast refractive-index change caused by emerging plasma [11-13]. It is argued in Refs. [14-16] that the filament is formed mainly by competition between beam self-focusing due to Kerr nonlinearity and defocusing caused by the plasma. In solids, the plasma density in the filament needed to overcome the Kerr-nonlinearity-induced self-focusing in the collimated beam should be much smaller than the plasma density needed for breakdown [17]. In recent publications [18, 19], the specific modification caused by such weak plasma within the filament has been suggested for application in laser writing of light-guiding structures within dielectrics. In contrast to material alteration caused by breakdown, this modification, resulting in changes of the refractive index, is not permanent and could be erased by annealing. These alterations are attributed to the formation of color centers and/or densification [20] but the

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detailed mechanism of the effect of the weak plasma that leads to such alterations is not yet fully understood.

In this respect, it is interesting to investigate the filamentation in media with a large number of electronic traps possessing memory of the electronic excitations. Such a medium could serve as a model for investigating phenomena caused by weak plasma.

In this communication, we consider the femtosecond laser-induced modification of a polymeric titanium oxide (Ti-O) based wet gel. This material possesses the solid component, which is a Ti-O polymeric network (4.5 mol %), and the liquid portion (95.5 mol %), which is mainly ethyl alcohol. Initially, the gel enclosed into a quartz cell is transparent in the visible spectral region. Ti^{4+} ions, constituting the gel chains, trap photo-excited electrons, forming Ti^{3+} centers. The number density of such traps can be about $10^{20}cm^{-3}$. It was demonstrated in [21-25] that these traps can be filled by UV bandgap (300 ÷ 370nm) irradiation. The creation of Ti^{3+} centers results in the appearance of a broad absorption band between 350 nm and 2500 nm with a maximum near 600nm (darkening).

Below, we show that the irradiation of gels by a loosely focused millijole IR femtosecond laser results in the creation of the Ti^{3+} centers. The amount of captured electrons can easily be determined by knowing their absorption cross-section, which was obtained in previous investigations of UV irradiation [25]. We show that IR femtosecond modification is threshold-like with respect to the laser intensity. The investigation of the kinetics of the accumulation of Ti^{3+} centers will provide useful information on the electronic processes involved.

2. Experiment

In our experiments, we used a Ti-sapphire femtosecond laser system (Tsunami+ Spitfire, *Spectra Physics*) which delivers femtosecond pulses with a pulse energy (E_p) of ~ 2 mJ and minimum pulse duration (t_p) of 45 fs at 800 nm (spectral FWHM ~ 25 nm) with a repetition rate tunable from 10 Hz to 1 kHz. Samples were irradiated in a fixed geometry (see Fig. 1). A 6-mm diameter laser beam (at e^{-1} of intensity) was directed by a spherical mirror (F = 50 cm) to a 0.5-mm quartz cell filled with gel or ethanol. The cell was located in the converging part of the beam 10 cm before the geometrical focus in the region free of filamentation in air. The beam diameter at the cell entrance was ~1.25 mm. t_p was tuned by a grating compressor (*Spectra Physics*). E_p was varied by use of neutral glass optical filters. Changes in t_p and E_p were controlled by a single-shot autocorrelator (SSA) (*Spectra Physics*) and a powermeter, model 407 (*Spectra Physics*). The absorption changes of the irradiated gels were measured in

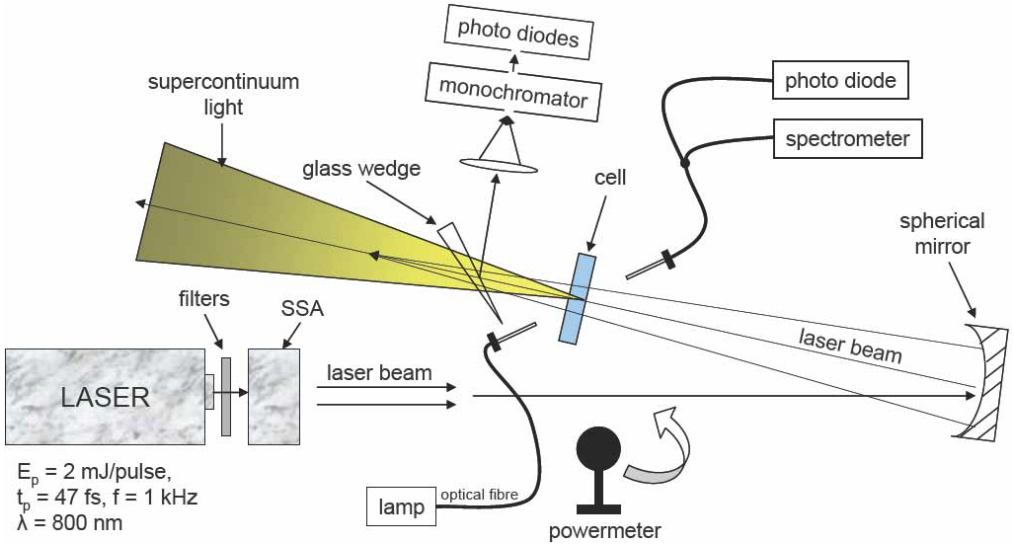

Fig. 1. Experimental setup.

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situ by an optical fiber probe using a spectroscopic lamp. A quartz fiber transmitted the signal either to a 2000 USB compact spectrometer (*Ocean Optics*) or to a photo diode coupled to a 60 MHz TDS1020 digitizing oscilloscope (*Tektronix*), which allowed the absorption kinetics analysis. The measurements of the absorption changes of the sample from pulse to pulse were performed at 800 nm. We monitored changes in absorption in the center of the irradiated region, 0.25 mm in diameter, where the laser beam intensity at the cell entrance is almost constant. Quantitative emission spectra measurements were carried out using a monochromator (*Solar-TII LTD*, Belarus) coupled to photo diodes calibrated for femtosecond radiation in the near IR-visible-near UV spectral region. Gel preparation is described elsewhere [22].

3. Results and discussion

Under the experimental conditions described above, IR femtosecond pulses induce darkening of the material, which is similar to the darkening by UV photons with the same spectrum of induced absorption. For this reason, it can be deduced that generation of trapped electrons occurs inside the gel under IR femtosecond irradiation. No significant change in the spectral lineshape is observed with the increase of the gel absorbance $A(\lambda) = \int_0^l \alpha(\lambda, x)dx$ during the experiment. Here α is the absorption coefficient and l is the cell width. Experiment shows a linear dependence of the absorbance on the pulse number (i) at each wavelength at the beginning of the darkening, thus allowing us to determine the initial rate of the absorption growth: $R(\lambda) = f \times (A(\lambda, i) - A(\lambda, 0))/i$, where f is the pulse repetition rate. Figure 2 shows R measured at 800 nm as a function of pulse energy at a fixed pulse duration (a) and pulse duration at a fixed pulse energy (b). It is seen that these dependences have both common and different features. They provide independent experimental information and taken together are important for determination of the relevant mechanism. As can be seen from the figure, the initial rate demonstrates a nonlinear dependence on E_p and/or t_p. This is understandable, when it is taken into account that the wavelength of irradiation belongs to the transparency range of the material. However, this darkening cannot be provided by the 2-photon absorption at the wavelength of the fundamental frequency (FF) because of the small photon energy $2\hbar\omega < E_{gap}$.

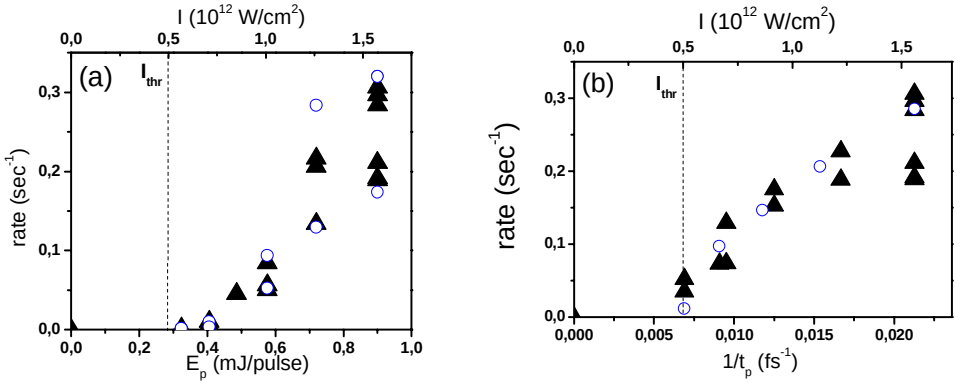

Fig. 2. Dependence of the initial rate of the gel absorption growth R at 800 nm in a 0.5 mm cell (triangles) on laser intensity at the entrance to the cell (a) when changing the laser pulse energy at fixed pulse duration t_p = 50 fs, (b) when changing the pulse duration at fixed pulse energy E_p = 0.9 mJ. Thickness of the quartz window is 1.5 mm. Other irradiation parameters are: d_{beam} = 1.25 mm, and f = 100 Hz. Circles indicate values of normalized products, $\varepsilon_{max} \times \varepsilon_{650}/t_p$, of spectral densities of the output radiation (see Fig. 3) at the wavelengths of the spectral maximum and at λ=650 nm.

When one gradually increases incident laser beam intensity by increasing E_p at a fixed t_p or decreasing t_p at a fixed E_p, one observes the onset of the generation of white light *within the*

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gel, and of conical emission, which is easily seen by the appearance of colored circles in the outgoing light. The intensity threshold of the white light generation inside the sample is close to the threshold of the sample darkening. This allows us to put forward a hypothesis that these two processes are interconnected. To verify this hypothesis, we performed quantitative spectral measurements of the emitted SC radiation at varied laser intensity. Corresponding threshold intensity in the gel is very close to threshold intensity of SC generation in pure ethanol. Therefore, further we use ethanol, the major component of the gel, as a model system to study SC spectra behavior in the gel to avoid the effects of sample darkening during the irradiation. An analysis of the E_p and t_p dependences of the SC spectra generated in ethanol, shown in Fig. 3 reveals that a detectable darkening of the sample correlates with the appearance of the blue asymmetric part of the outgoing spectrum.

The reason for such correlation can be the following. For ethanol the typical critical power is $P_{cr} = 6.5\times10^5$ W [26]. At the onset of darkening, the value of intensity in our experiments was $I = 5\times10^{11}$ W/cm^2 with $P = 6\times10^9$ W >> P_{cr} . An estimate of the retardation integral $B = 2\pi\lambda^{-1}\int_0^l n_2 I(z)dz$ [1], calculated along the beam propagation path through both the entrance window of the cell and the gel layer, yields the value $B > 4$ at the onset of darkening. This estimation, along with the observation that the induced absorption growth correlates with the onset of conical emission, allows us to suggest that the darkening occurs when the beam becomes instable. The filamentation is usually accompanied by ionization. The shape of the SC spectrum shown in Fig. 3, as was argued in numerous publications (see, e. g. [13]), corresponds to phase modulation of the beam when changes in the refractive index are provided by the ionization. Free electrons generated due to ionization can be trapped by Ti^{4+} ions and thus contribute to the gel darkening. Below we consider the ability of this mechanism to explain the observed gel absorption growth.

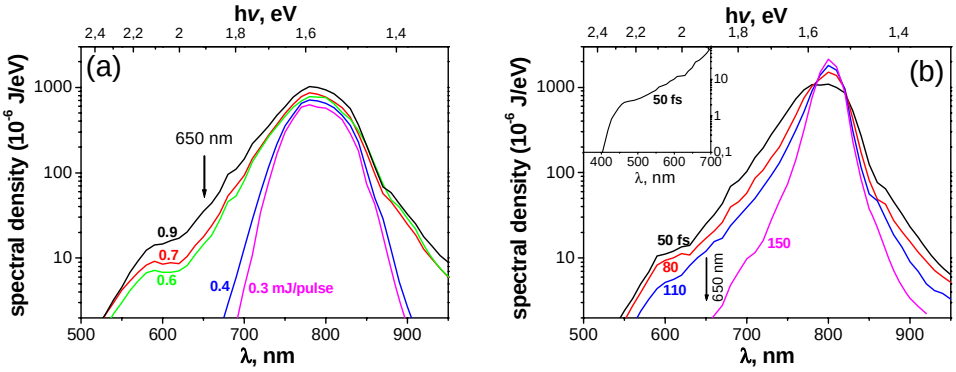

Fig. 3. Spectral density of supercontinuum radiation measured in J/eV generated in 0.5 mm quartz cell filled with pure ethanol for different values of the laser intensity at the cell entrance (a) when changing the laser pulse energy at fixed pulse duration t_p = 50 fs, (b) when changing the pulse duration at fixed pulse energy E_p = 0.9 mJ. Other irradiation parameters are: d_{beam} = 1.25 mm, and f = 100 Hz. Values of variable parameters are listed in the figures.

The amount of free electrons formed within filaments can be estimated in case of high value of retardation integral $B \cong 10$ (E_p = 0.9 mJ, t_p = 50 fs). We consider a simplified model where the laser light intensity, I_f, and plasma concentration within the filament, N_e, are determined by the competition between the focusing provided by the Kerr nonlinearity and defocusing caused by plasma ($n_2 I_f \approx \Delta n_e N_e$) and each part of the beam participates in the formation of a filament. Following the main ideas of the simplified model developed in Ref. [27], we suggest that plasma within the filament is formed by the multiphoton ionization that is provided within the filament by the intensity I_f for the pulse duration t_p. This model neglects the effect of group velocity dispersion (GVD) on plasma density considered in Ref. [28]. It is

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reasonable for short distances ~1mm, as is the case of our experiment. In Ref. [29] it was shown that variations of GVD significantly influence the extent of the SC spectrum, keeping both the light intensity and plasma density within filament practically unchanged.

This model allows for an assessment of the density of plasma electrons created per single laser pulse averaged over the laser pulse propagation volume inside the cell, $\overline{N}_e = I_{beam}(\sigma_e L_{cell})^{-1}(N_0 \beta_K \Delta n_e t_p / n_2)^{-1/(K-1)}$. Here β_K is the coefficient of K-photon ionization per a single ethanol molecule of density N_0. σ_e and Δn_e are the effective absorption cross-section and the absolute value of refractivity of plasma electrons estimated by Drude formulas, respectively. The estimated $\overline{N}_e$ can be compared to the averaged density of trapped electrons, $\overline{N}_{Ti^{3+}}$, calculated from the experimentally measured value of the increment of the optical density per pulse at the wavelength of probing light. For t_p=50fs and I_{beam}=1.5×10^{12}W/cm^2 the measured increment of absorbance at 800 nm per pulse is ΔD = 3×10^{-3}. Knowing the absorption cross-section of Ti^{3+} $\sigma_{Ti^{3+}}(800nm) = 2.5\times10^{-18} cm^2$ [25], we calculate that $\overline{N}_{Ti^{3+}} \cong 2.5\times10^{16} cm^{-3}$. For realistic sets of the parameters involved, the estimated density $\overline{N}_e$ appears to be ~ 10^{17} cm^{-3}, which is larger than the experimentally measured $\overline{N}_{Ti^{3+}}$, indicating the possibility for such electrons being captured by Ti^{4+} centers to contribute to the material darkening.

It should be noted that, at estimated intensities $I_f \approx 3\times10^{13}$ W/cm^2 the 3-photon absorption within the solid portion of the gel with subsequent direct trapping of corresponding excited electrons by Ti^{4+} ions could also contribute to the generation of Ti^{3+} centers. The real contribution of both of these mechanisms, however, could hardly be determined because of the lack of knowledge about the trapping efficiency of the electrons generated in such ways.

The distinguishing feature of the above mechanisms is that they suggest the generation of Ti^{3+} centers exclusively within the filaments. However, such mechanisms could hardly explain the shape of a modified region in a relatively thick 4 mm cell shown in Fig. 4(a), even if we take into account the space distribution of filaments in a Gaussian beam, as demonstrated in Ref. [30]. The modified region is diverging from the right to the left, while the initial beam propagates from the right to the left and is slightly focused (i.e., converging). The shape of this region correlates with the diverging shape of the SC beam, which starts generating just near the input surface of the sample. Such a shape of the modified region permits one to put forward the hypothesis that the SC radiation itself is involved in the modification process.

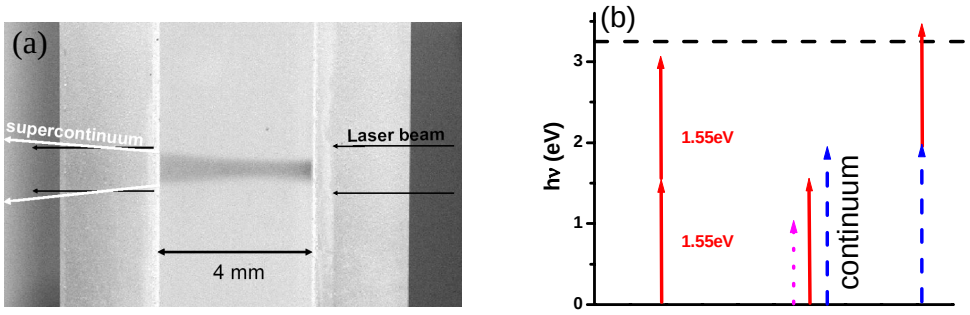

Fig. 4. (a). Photograph of the modified gel domain in a 4 mm cell. The laser beam propagates from the right to the left. (b) Scheme of 2-photon modification of the gel with participation of photons from the broadened spectrum.

In earlier paper [31] on IR femtosecond laser-induced color center generation in silicate glasses it was argued that the main reason of this phenomenon is single-photon absorption of

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photons from the UV tail of SC. The result of our investigation of the UV part of the SC is shown in the inset of Fig. 3(b). It can easily be estimated that the number of photons within the range of a single-photon modification (λ<370nm) is too small to provide any detectable darkening of the sample. According to [16] the UV border of the SC spectrum in the gel should have an even longer wavelength than in ethanol. On the contrary, Fig. 2 demonstrates an evident correlation between the initial rate of darkening as a function of the incident laser intensity and the product $\varepsilon_{max}\times\varepsilon_{650}/t_p$. Here, ε_{max} is the spectral density of the output beam at the maximum, which is close to the FF, and ε_{650} is the output spectral density at a wavelength of 650 nm. This product is proportional to the probability of 2-photon transition at corresponding wavelengths. The summed energy of two photons, one at the FF and one at 650 nm, is equal to the energy of a photon with a wavelength of ~355 nm, which belongs to the range of single-photon absorption of the gel leading to the creation of Ti^{3+} centers.

This correlation allows us to suggest that the main process responsible for the modification of the gel is *two-photon absorption when one photon is close to the fundamental frequency and the other comes from the supercontinuum broadening* (see Fig. 4(b)). Knowing the concentration of the created Ti^{3+} centers, we can assess the 2-photon absorption cross section $\delta(\lambda_1, \lambda_2)$ in the ranges of λ_1 = 790 nm ± 50 nm, and λ_2 = 650 nm ± 50 nm supposing that the above 2-photon absorption results in the creation of all detected Ti^{3+} centers with efficiency 0.25 [24]. The summed energy of these two photons $\hbar\omega = \hbar\omega_1 + \hbar\omega_2$ lies within 3.48 ± 0.24eV. Considering a rectangular pulse, and supposing that the SC is uniformly distributed within the whole area of the laser beam, we estimate the value of the 2-photon absorption cross-section per Ti^{4+} ion as $\delta = 5\times10^{-50}$ cm^4s. The value of δ, which should be considered as an estimation from above, is far from the upper limit for 2-photon absorption cross-sections measured in different media [32], thus proving the reliability of the considered mechanism.

If the SC is generated due to phase modulation caused by ionization, then the blue part of the spectrum is generated at the leading edge of the pulse. Because the dispersion is normal, the blue light propagates slower than the red light. This facilitates the interactions of photons within the filament. However, it is important that the discussed mechanism can also work in the region free of filaments.

Additional experiment was performed with two cells when SC light was generated in the first cell filled with ethanol and the second cell with the gel was located right after the first one. We observed the darkening of the gel in such geometry and the size of the modified region was close to the size of the SC beam in the gel cell. This provides a complementary proof of the proposed mechanism.

4. Conclusion

In conclusion, titanium oxide polymeric wet gels, being media with a large number of traps for electrons, provide an opportunity to elucidate the interaction of femtosecond pulses with materials. The trapped electrons are stored here as Ti^{3+} centers, which could easily be identified through the induced wide band optical absorption. We have studied the effect of loosely focused powerful IR femtosecond radiation on this medium. Our estimations show that excited electrons produced due to high- (three and more) order multiphoton absorption by a self-focused laser beam within the filaments and trapped as Ti^{3+} centers could contribute to the material modification. However, the mechanism which relates the electron excitation to the two–photon absorption when one photon is of the fundamental frequency and the other comes from the blue part of supercontinuum radiation can alone explain the main features of the observed titanium oxide gel alteration.

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