Nonlinear propagation of fs laser pulses in liquids and evolution of supercontinuum generation

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Abstract: Nonlinear propagation of fs laser pulses in liquids and the dynamic processes of filamentation such as self-focusing, intensity clamping, and evolution of white light production have been analyzed by using one- and two-photon fluorescence. The energy losses of laser pulses caused by multiphoton absorption and conical emission have been measured respectively by z-scan technique. Numerical simulations of fs laser propagation in water have been made to explain the evolution of white light production as well as the small-scale filaments in liquids we have observed by a nonlinear fluorescence technique.

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1. Introduction

Filamentation of ultrashort high power laser pulses has been well observed for many years in gases, liquids and solids. And it still attracts considerable attention since many applications can be found such as white light production [1-3], pulse compression [4-7], remote sensing using lidar [8, 9], and optical switch [10]. Since the first observation of self-trapping of optical beams at the very dawn of nonlinear optics [11], quite a number of models have been developed to explain the nonlinear processes of laser pulses propagating in optical media [12-19]. It is now well recognized that self-phase modulation (SPM), dispersion, diffraction and plasma behaviour are the main causes of what we have observed such as supercontinuum generation, pulse splitting, X waves in the nonlinear propagation of such laser beam. The filamentation in bulk media is generally accompanied by conical emission (CE) which appears on a screen as a concentric rainbow with colors ranging outward from red to green. CE has been addressed as another form of paraxial X-wave and considered as a key for the interpretation of the out-of-axis energy emission in the splitting process of fs laser pulses in normally dispersive materials [15]. The filaments release a fraction of their energy through CE which interact with background and create additional filaments [20]. The formation of CE has been explained in different ways such as Cerenkov process, four-photon parametric process, plasma process and three-wave mixing picture [21-26]. However, a good understanding of the
formation and evolution as well as the role of CE in the propagation of fs laser pulses in transparent media is still lacking. Recently we have found that CE is inherently caused by the space-frequency coupling mainly from the interplay among SPM, dispersion and phase mismatching [27]. The strongly conical wave plays a major role of energy redistribution in space and induces energy loss in the on-axis part of the pulse [27,28].

The aim of this paper is to demonstrate nonlinear propagation of fs laser pulses in liquids and the dynamic processes of filamentation such as self-focusing, intensity clamping as well as evolution of white light production by using one- and two-photon fluorescence. The energy losses of laser pulses caused by multiphoton absorption (MPA) and CE have also been measured respectively by z-scan technique. Numerical simulations of fs laser propagation in water have been made to explain the evolution of white light production as well as the small-scale filaments in liquids we have observed by a nonlinear fluorescence technique. Conical emission plays an important role of energy redistribution and can prevent laser beam from self-guiding over a long distance.

2. Visualization of filamentation of fs laser pulses in liquids

A nonlinear fluorescence technique as described in Ref. [29,30] has been used to visualize the formation of filaments behind a slit (200 μm×5.0 mm). By this way we can observe the dynamic processes of filamentation such as self-focusing, intensity clamping, and white light production. The experimental setup is shown in Fig. 1. The Ti: sapphire laser parameters in front of the slit in standard operations are: 1 kHz, 800 nm, 2.0 mJ, pulse duration T=44 fs, and radius 𝑟1/𝑒=4.3 mm respectively. The slit is placed in front of a glass cell (the length of the cell is 20 mm). The dilute test solutions which have the same concentrations of ~10^16 cm^-3 are Coumarin 500, fluorescein 27 and DODCI in ethanol, and their absorption wavelengths are 395, 512, and 585 nm respectively. Here we call them blue, green and red dye respectively. The laser energy behind the slit was 44 μJ which corresponds to 1GW, or ~1.0×10^11 W/cm². The picture of multi-filamentation is taken from the side by a CCD camera. We use the slit arrangement because all the filaments are created in one column which can be projected onto the detector plane of the CCD array without any ambiguity.

In Fig. 2 we show the development of a one-dimension filament array in ethanol behind a slit by using blue, green and red dye respectively. The fluorescence is excited by two-photon absorption of the fundamental laser beam and one-photon absorption of white light due to self-steeping. The pictures are taken by a single shot with the same condition except the dyes. For coumarin 500, the two-photon absorption of the fundamental laser beam is close to the resonance absorption and therefore the two-photon fluorescence signal emitted by Coumarin 500 is bigger compared to Flurescein 27 and DODCI. The picture size shown here is H×L=2.5 mm ×10.7 mm and L corresponds to the propagation distance from the left to the right. The three pictures (d), (e) and (f) which are taken with a higher spatial resolution (2.5 μm) correspond to the first filament at the top of Fig. 2(a), (b) and (c) respectively. Many small-scale filaments form during the propagation of a femtosecond laser beam in ethanol after the slit. The filaments develop from the irregularities of the beam profile at the beginning.
Fig. 2. Development of a one-dimension filament array in ethanol behind a slit. The fluorescence is excited by two-photon absorption of the fundamental laser beam and one-photon absorption of white light. The dyes in ethanol are (a) Coumarin 500, (b) Fluorescein 27, and (c) DODCI respectively. The pictures are taken by a single shot with the same condition except the dyes. The picture size shown here is H×L=2.5 mm×10.7 mm and L corresponds to the propagation distance from the left to the right. The three pictures (d), (e) and (f) below which are taken with a higher spatial resolution (2.5 μm) correspond to the first filament in (a), (b) and (c) respectively.

Here for example only one filament (the first one on the top in each case, by chance) is used to investigate the filamentation process in detail. In Fig. 3 we show the evolution of the fluorescence intensity from Coumarin 500 as well as the diameter of the first filament along the propagation. The fluorescence signal is integrated over the whole size of the first filament. For two-photon fluorescence the integrated signal $I_{\text{int}}(z)$ detected by the camera is proportional to $I_{\text{int}}(z) = \int I(r,z,t) rdrdt$, $I(r,z,t)$ is the intensity of the laser beam. If the laser intensity is assumed as a spatio-temporal Gaussian profile, the maximum laser intensity on the axis is proportional to $\sqrt{I_{\text{int}}(z)}/\Delta r/\Delta \tau$, $\Delta r$ is the radius of the laser beam, and $\Delta \tau$ is the time duration. The diameter of the laser beam decreases to its minimum and the fluorescence signal increases.
Fig. 3. Evolution of fluorescence intensity as well as the diameter of the first filament as a function of propagation distance. The fluorescence is excited in the blue dye in ethanol.

Fig. 4. Vertical line profile of fluorescence signal at the propagation distances $z=8.0$, 8.6 and 9.0 mm respectively. The fluorescence is excited by the blue dye Coumarin 500 in ethanol.

to its maximum after the first 8 mm of propagation. However the fluorescence intensity increases to another maximum at 8.6 mm. In Fig. 4 we show the linear fluence profile integrated across a vertical strip of 10 μm wide of the fluorescence signal at the propagation distances $z=8.0$, 8.6 and 9.0 mm respectively. The linear profile which corresponds to the beam profile of the filament is scanned vertically from Fig. 2(d). At the first maximum position $z=8.0$ mm, the linear profile is very narrow, the diameter is $\sim 12$ μm (full width at half its maximum, FWHM), and the laser intensity is clamped here. However at the second maximum position $z=8.6$ mm, the linear profile becomes broader and has a pedestal signal with a radius of $\sim 40$ μm (here the radius is meant for the radius of the pedestal signal and calculated from the maximum fluorescence intensity to the minimum fluorescence intensity since the profile is not Gaussian). The radius of the pedestal signal increases rapidly and becomes $\sim 70$ μm at $z=9.0$ mm. The speed the pedestal signal spreads outward can be defined by a diverging angle $\theta = \arctan(\Delta r / \Delta z)$. $\Delta r$ is the change of the radius of the pedestal signal and $\Delta z$ is the propagation distance. For blue dye this diverging angle is calculated from the experimental data to be $\sim 4.4^\circ$. The diverging angles for green and red dyes are calculated as $\sim 3^\circ$ and $\sim 2.3^\circ$ respectively in the same way by analyzing Fig. 2(e) and (f) respectively. This is one proof that the increase of fluorescence intensity at the second maximum position together with the pedestal signal shown in Fig. 2 is caused by one-photon fluorescence due to the absorption of the spreading blue light of the supercontinuum [29]. The blue light spreads outward at the largest angle which exhibits the sign of CE [21,22].
The evolution of fluorescence intensity emitted by blue, green and red dye respectively from the same filament is shown in Fig. 5(a). An interesting thing shown in Fig. 5(a) is the big difference of the position of the second peaks which correspond to one-photon fluorescence emitted by blue, green and red dyes respectively. It shows an “anomalous” phenomenon that blue light (from the blue dye) comes first, and then green light later and red light comes last. Shot to shot measurements is shown in Fig. 5(b) and they are highly reproducible in our experiments. Our explanation is that this “anomalous” phenomenon is related to CE. Since blue light diverges more rapidly compared to red light, the intensity of blue light decreases more rapidly. Blue light can not last and be amplified over a longer distance as red light can. So it ends earlier.

For comparison with our numerical simulation, in Fig. 6(a) we show a one-dimension filament array in water behind a slit. The test dye is Rhodamine B and its maximum absorption wavelength is 556 nm. (The reason why we use Rhodamine B in water as our test sample is because the known optical parameters of water are better studied than ethanol.) The evolution of fluorescence intensity along the first filament is shown in Fig. 6(b). The second peak shows the evolution of white light whose spectrum covers the absorption band of Rhodamine B.
3. Energy loss caused by CE and MPA

![Diagram showing experimental setup for z-scan measurement of energy loss caused by CE and MPA]

The visualization of filamentation of Ti:sapphire femtosecond laser pulses in liquids shows that only small-scale filaments can be observed in our case. The filaments can only persist for only around 2 mm and have diameters around 12 μm. In our case white light is produced in a very short distance after the laser intensity is clamped. White light which is produced from the fundamental laser beam diverges and spreads outward rapidly as shown in Fig. 4. This is the well known CE. However we have observed it at its very beginning of production by this fluorescence technique. The filaments dye out coincidently with the production of white light, which implies that white light is not in phase with the fundamental laser beam and its outward spreading by the way of CE will unavoidably take some energy from the fundamental laser beam inside the filament.
We have measured the energy loss caused by nonlinear effect such as MPA as well as CE respectively by using a z-scan technique. The experimental setup is shown in Fig. 7(a). A metal mesh (5×5 meshes, unit cell 497 µm × 497 µm, wire width 54µm) is placed in the laser beam before the laser passes through a sample cell in which water is filled. The cell is 12 mm long and the inside length is 10 mm. The position of the mesh can be scanned from the surface of the cell to 400mm away along the propagation axis. By changing the mesh distance multi-filamentation and white light production can be controlled [30, 31] because the laser intensity on the entrance surface of the sample cell will change due to the diffraction of the laser beam in free space by the mesh. Moreover, the steep intensity gradient of the near field diffraction pattern caused by the mesh is very efficient for the creation of filaments and white light without making the fundamental laser beam diverging as a focusing lens usually does. In order to measure the energy loss caused by MPA, an energy meter is placed close to the exit surface of the sample cell to measure the energy of the laser after it propagates through the cell. This arrangement can guarantee most of the laser beam including the white light be collected by the energy meter. Three measurements are made in this case. 1) Energy measurement of the laser beam without the sample cell as the position of the mesh is scanned. 2) Energy measurement with the sample cell included but the laser pulse is adjusted to be 1 ps to avoid nonlinear effect. 3) Energy measurement with the sample cell included but the laser pulse is adjusted to be 44fs to include strong nonlinear effect. The measurement result is shown in Fig. 7(b). The position 0 of the mesh distance on the x-axis corresponds to the surface of the sample cell. The energy measurement of the laser beam with a pulse duration of 1 ps shows the transmission efficiency of the sample cell is about 91%. Therefore the linear absorption of the laser by the sample cell is very small if the reflectivity of the two outer surfaces (around 8%, without including the two inner surfaces) is considered. When the laser pulse is adjusted to be 44 fs, by scanning the position of the mesh, strong white light and CE can be observed. The energy measurement shows the maximum energy loss is less than 3%. It is mainly caused by nonlinear absorption like MPA. Placing the energy meter 2 meters away from the exit surface of the cell along the propagation axis, the same z-scanned measurements of laser energy are shown in Fig. 7(c). After the nonlinear propagation and filamentation in the liquid, the laser beam including white light continues to propagate in free air. And at 2 meters away, most white light except the fundamental laser beam will not be collected by the energy meter due to CE. So by this way we can measure the conversion efficiency of CE due to the strong filamentation of laser pulses in water. The maximum conversion efficiency of CE is about 40% when the mesh distance is set as 50 mm. So the energy loss caused by CE is much higher compared to MPA. When the position of the mesh is scanned from the surface of the sample cell to 400 mm away along the propagation axis, the laser intensity distribution on the entrance surface of the sample cell will change due to the diffraction of the laser beam in free space by the mesh. The change of the laser intensity on the entrance surface of the sample cell will have influence on the filamentation of fs pulses in liquids as well as the conical emission and MPA.

In Fig. 7(d) we show the z-scan measurement of energy loss caused by CE for water, ethanol and CS₂ respectively. The reference is given as the energy measured when the pulse duration is adjusted to be 2 ps. It can be seen that energy loss caused by CE for CS₂ is as high as 86%. The energy loss caused by CE for water and ethanol is 38% and 41% respectively. Small difference of energy loss between water and ethanol is attributed to their small difference of nonlinear refractive index (2.7×10^-16 cm²/W and 5.0×10^-16 cm²/W respectively) and linear refractive index (1.33 and 1.36 respectively). However, for CS₂, its nonlinear refractive index is much higher (2.4×10^-15 cm²/W [32]) and the linear refractive index is 1.63 (a large linear refractive index means a large group velocity dispersion), a much stronger CE can be observed. That is why the energy loss caused by CE is much higher compared to water and ethanol because CE for CS₂ has a larger diverging angle. CE is quite dependent on different materials. Available theories of nonlinear propagation equation of waves normally...
include three main parameters which might have contribution to CE. They are $n_2$ (SPM), group velocity dispersion (GVD) and multi-photon ionization (MPI) rate (plasma). Most of the phenomena we have observed can be explained by these parameters. However these three parameters are not independent but related, which makes it difficult to explain which parameter is more important in some cases. Some early experiments show that supercontinuum generation is dependent on bandgap of materials. For some materials such as CS$_2$, supercontinuum cannot be generated when the bandgap is smaller than a threshold and thus the clamping intensity is low [2]. Here what we have observed from the measurement of energy loss caused by CE indicates that CE for CS$_2$ is much stronger compared to water which has a larger bandgap of 7.5 eV.

We have measured beam profiles of CE centred at red ($\lambda$=680nm) and green ($\lambda$=520nm) light for water and CS$_2$ respectively. The experimental setup is shown in Fig. 8(a). A metal mesh (5×5 meshes, unit cell 497 µm × 497 µm, wire width 54µm) is placed in the laser beam before the laser passes through a sample cell in which water or CS$_2$ is filled. The cell is 12 mm long and the inside length is 10 mm. The energy and the pulse duration of the laser pulse after passing through the mesh are 0.3 mJ and 44 fs respectively. The distance between the mesh and the cell is ~ 7 cm. CE which is produced from filamentation of fs laser pulses in water or CS$_2$ is stopped by a screen plate and imaged onto a CCD camera. The distance between the...
screen plate and the cell is ~15 cm. Red and green band-pass filters are placed respectively in front of the screen plate to select CE centred at red and green light respectively. The bandwidths of the filters are ~70 nm. The measured beam profiles of CE centred at red and green light for CS2 and water are shown respectively in Fig. 8(b) and (c) respectively. It is shown that the profiles of CE have ring structures and CE for CS2 is much stronger compared with water.

4. Numerical simulation

A numerical simulation of nonlinear propagation of an ultrashort laser pulse in water based on the model described in Ref. [17, 27] was made to explain the evolution of white light production as well as the small-scale filaments in liquids we have described above. The equation of propagation of axially symmetric electric field is formulated in the frequency domain in order to tackle the dispersion more precisely and can be expressed as [27]

$$\frac{\partial}{\partial t} \hat{E}(r, z, \omega) = \left[ \frac{i}{2k(\omega)} \nabla_z^2 + ik(\omega) \right] \hat{E}(r, z, \omega) + \frac{i\omega^2 \hat{P}_{nl}(r, z, \omega)}{2k(\omega)c^2\varepsilon_0} - \frac{\omega \hat{J}_f(r, z, \omega)}{2k(\omega)c^2\varepsilon_0}$$

(1)

This is the scalar unidirectional pulse propagation equation solved in the z direction. The four terms on the right side corresponds to diffraction, dispersion, SPM and plasma behaviour respectively. Here $\omega$ is the optical frequency, the wave number $nk = \omega/\varepsilon_0$.

$\hat{P}_{nl}(r, z, \omega)$ is the nonlinear polarization in the frequency domain of $P_{nl}(r, z, \omega)$ given by

$$P_{nl}(r, z, t) = 2\varepsilon_0 \eta_n n_2 I(r, z, t) E(r, z, t)$$

(2)

$n_b$ and $n_2$ are linear refractive index and nonlinear coefficient respectively. Higher order terms are neglected in Eqs. (2). $\hat{J}_f(r, z, \omega)$ can be described as:

$$\hat{J}_f(r, z, \omega) = \frac{e^2}{m(v-i\omega)} \eta_n (r, z, \omega) \otimes E(r, z, \omega) + \int \beta^{(n)} \int \hat{J}_{e}^{(n)}(r, z, \omega) \otimes E(r, z, \omega) / k_0$$

(3)

Where $\otimes$ is the convolution operator, $v$ is collision frequency, $I_p$ is the energy of bandgap, and $k_0 = 0.5ce_0$. The evolution of free electron density is described as

$$n_e(r, z, t) = \int I_{me}(r, z, \omega) d\omega$$

(4)

And the electron generation rate is expressed as

$$I_{me}(r, z, t) = \beta^{(e)} I_e^{(e)}(r, z, t) + \eta_m n_e(r, z, t) - \eta_c n_c(r, z, t)$$

(5)

Where $\eta_{me}$ and $\eta_{mc}$ represent cascade ionization and electron-ion recombinations rate respectively as described in Ref. [25, 33]. $\eta_{me} = (I(r, z, t) e^2 \tau_e / n_e m_c c_0 (1 + \alpha_t^2 \tau_e^2))$, where the collision time $\tau_e = 1$ fs for water and collision frequency $v = 1/\tau_e$. The value of $\eta_{me}$ is $2 \times 10^{-15}$ m$^3$s$^{-1}$. $\beta^{(e)}$ corresponds to coefficient of multi-photon transition from the valence to the conduction bands [27,33].

The laser parameters for the simulation are chosen to be as close as possible to those used in our lab. The central wavelength is 800 nm, and the spectral width is about 27 nm (FWHM) corresponding to a pulse duration of about 38 fs although the pulse duration in our experiments is measured as 44 fs. The diameter (FWHM) of the laser beam is chosen as 110 μm (corresponding to roughly the diameter of an initial hot spot before self-focusing in our...
Experiment) and the input power is 14.5 $P_\text{cr}$. The critical power for a cw beam collapse is $P_\text{cr}=2.74$ MW for a nonlinear coefficient $n_2=2.7\times10^{-16}$ cm$^2$ W$^{-1}$. The frequency dependent refractive index of water is chosen as expressed in a formula given in Ref. [34]. In our simulation, the linear loss caused by water is not included in the frequency dependent refractive index since the length of the propagation in water is only 1 cm and the linear absorption is very small and neglected. Similarly the influence of absorption of the dyes on the propagation of laser pulses in liquids is neglected since the concentrations of the dyes are very low. The incident laser beam profile for numerical simulation is chosen as a spatio-temporal Gaussian profile without chirp. In Fig. 9 we show the simulated evolution of the on-axis maximum laser intensity, the size of the laser beam and two-photon plus one-photon (556 nm for Rhodamine B) fluorescence as a function of propagation distance in order to compare the simulation results with the experiments. The two-photon plus one-photon fluorescence is calculated by $I_{\text{tot}}(z) \propto \int \int \left( I_2(r,z,t) + \beta I_1(r,z,t) \right) r \, dr \, dt \cdot I_\text{s}(r,z,t)$ and $I_1(r,z,t)$ are the intensity of the laser beam and that of the white light component at 556 nm (spectral width = 30 nm) respectively. $\beta$ is a coefficient which corresponds to the relative intensity of two-photon and one-photon fluorescence. The evolution of two-photon plus one-photon fluorescence intensity shown in Fig. 9 is close to what we have observed in Fig. 6(b). Compared with the evolution of the on-axis laser intensity, the first peak at 10.6 mm in Fig. 9 corresponds to the position where the laser intensity is clamped. The diameter of the laser beam reaches its minimum value of 10 $\mu$m. After that, the on-axis laser intensity decreases gradually, however the intensity of the white light at 556 nm continues to increase until the laser beam propagates to 12.7 mm.

The simulated evolution of the maximum on-axis laser intensity together with the evolutions of the intensities of white light centred at different wavelengths (at 636, 520, and 420 nm, and we call them red, green and blue light respectively) is shown in Fig. 10. The intensities of the red, green and blue light are calculated by a Fourier transformation of a segment of spectrum with a band-width of 30 nm and their central wavelengths are 636, 520 and 420 nm respectively. The evolutions of red light, green light and blue light related to the maximum on-axis laser intensity show a qualitative similarity with that shown in Fig. 5(a) although the simulation is made for water instead of ethanol used in Fig. 5. The intensity of red light can increase over a longer distance compared to green and blue light and therefore the onset of the peak intensity of red light occurs later. This can be explained as a consequence of CE because the diverging angle for blue and green light is larger. Blue light can not last and be amplified over a longer distance as red light can. So it ends earlier. White light is produced in a very short distance (~ 1 mm) after the laser intensity is clamped. It takes

![Fig. 9](image-url)
the form of CE and plays a very important role of energy redistribution in space and induces energy loss in the on-axis part of the laser pulse. CE provides a dissipative mechanism which can arrest catastrophic self-focusing and prevent laser beam from self-guiding over a long distance [27].

In this simulation, the peak electron density is calculated as $1.3 \times 10^{18} \text{ cm}^{-3}$. And the energy loss caused by MPA is ~ 5% which is comparable with the measurement of energy loss caused by MPA for water in Sec. 3. Our numerical simulations indicate that when the laser power for our laser parameters is lower than 8 times the critical power for strong self focusing, MPI as well as MPA plays a neglectable role [27]. In this case, CE which is mainly caused by SPM and the dispersion plays an important role of energy loss in the on-axis part of laser pulse.

5. Conclusion

In summary, we have demonstrated nonlinear propagation of fs laser pulses in liquids and the dynamic processes of filamentation such as self-focusing, intensity clamping as well as evolution of white light production by using one- and two-photon fluorescence. The energy losses of laser pulses caused by MPA and CE have also been measured respectively by z-scan technique. Numerical simulations of fs laser propagation in water have been made to explain the evolution of white light production as well as the small-scale filaments in liquids we have observed by a nonlinear fluorescence technique.