

Raman effect in self-focusing of few-cycle laser pulses in air

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Self-focusing of ultrashort pulses in air is investigated by means of numerical simulations. The role of the vibrational Raman effect and its dependence on pulse chirp is studied, with results shedding new light on the interpretation of the measurements of the critical self-focusing power. We also discuss computational modeling issues important specifically for few-cycle pulses. © 2013 Optical Society of America

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Self-focusing of few-cycle pulses in molecular gases can be affected by the impulsive excitation of vibrational Raman nonlinearity, which can shift the effective self-focusing threshold to higher powers. Numerical simulations in the field of laser filamentation usually do not take the impulsive vibrational response into account explicitly, perhaps due to long pulse durations, but also because its relative strength is not yet known. It is the aim of this work to explore signatures that can reveal the ratio between instantaneous Kerr and vibrational Raman nonlinearity. We also discuss practical issues that will be important for modeling and interpretation of future experiments with few-cycle pulses.

Several works motivated this study. Almost a decade ago, Bartels *et al.* [1] pointed out that the vibrational Raman effect can affect dynamics in high-power laser pulses with durations initially longer than the vibrational period of the given molecular gas medium. More recently, Odnher *et al.* demonstrated [2–4] that pulses undergoing filamentation can excite impulsive vibrational Raman response due to short-duration transient subpulses that occur in the filament core. Perhaps related to this is the observation made earlier by Chen *et al.* [5], who found that spectra in loosely focused filaments exhibit energy- and propagation-dependent red shift akin to soliton self-frequency shift in fibers. More refined experiments were recently performed by Uryupina *et al.* [6], who also proposed a simplified model based on the stimulated Raman effect. However, spatially and temporally resolved simulation that could capture these effects is still missing. These works dealt with relatively long pulses. Here we are interested in extremely short pulse durations, such as those studied by Laban *et al.* in [7]. They measured the critical power for self-focusing for a ~ 6 fs pulse at 800 nm, and found an unexpectedly high value of $P_c = 18$ GW. Because the pulse duration was sufficiently short to excite the vibrational Raman response in the impulsive regime, one could argue that it caused the increase in P_c . One of our aims is to show that there may be additional reasons behind an apparent increase in the self-focusing threshold.

Our simulation results were obtained with the unidirectional pulse propagation equation [8] simulator that

utilized the standard model for light–matter interaction [9]. Strong-field ionization rates for oxygen and nitrogen were parametrized as in [10], and the nonlinear index $n_2 = 7.8 \times 10^{-20}$ cm²/W was chosen according to recent measurements by Wahlstrand *et al.* [11]. Because of the short pulse duration, the rotational part of the nonlinear index can be neglected, and the only contributions to n_2 are instantaneous electronic and vibrational stimulated Raman effects. Since their relative strengths are not known, we perform comparative simulations for various values of fraction f that represents the electronic component. The remaining third-order nonlinearity is proportional to $(1-f)n_2$ and is implemented as the model described in [1] with the oscillator frequency corresponding to the nitrogen molecule vibration period of 14.3 fs [3]. Note that for the purposes of this work, the choice of any particular value of the nonlinear index is not important, and the parametrization in terms of f is merely a matter of convenience.

The initial beam had a waist of 2.2 mm, and the transform-limited FWHM pulse duration was 5 fs. In agreement with the findings of [7], the initial carrier-envelope phase did not affect observables utilized in this work. We explore different initial pulse chirps, and their effect on the self-focusing behavior. A convenient way to specify this in simulations is to set a location z_{TL} at which a transform-limited pulse forms in the absence of all nonlinear interactions, but with the gas chromatic dispersion fully taken into account. We specify z_{TL} with respect to the linear focus of a parabolic mirror with a focal distance of 75 cm.

An important modeling issue concerns focusing. It is common in the field of laser filamentation that focusing in a pulsed beam is implemented as an additional phase that is quadratic in radius. While this is certainly adequate for sufficiently long pulses, for the few-cycle durations relevant for this work, the reflection from a focusing parabolic mirror must be properly modeled. The effect of the mirror on the pulse is implemented as a quadratic-in-radius temporal shift of the pulse. Such an initial condition creates a focused waveform localized on a thin spherical shell with the center in the focal region. While in long pulses this reduces to the common phase-screen

model, it makes a difference in the temporal and radial profile of the pulse in the focal region for few-cycle pulses. In particular, it eliminates artifacts introduced by the simpler phase-screen implementation in which the peripheral portions of the beam take longer to arrive in the focus and thus prolong the on-axis pulse duration by an additional ≈ 10 fs.

The dispersion length in a 5 fs pulse in air is as short as ~ 40 cm. This becomes an issue when the minimal pulse duration is to be achieved in the focal region. Intuition suggests that such a setting can be verified by maximizing the bandwidth of the self-phase modulation spectrum [7], because longer pulses should result in less broadening. However, this intuition-based argument is not correct. Figure 1 shows spectra obtained for various chirp settings. It is evident that the spectrum generated in the pulse that attains its minimal duration in the focus is in fact not the broadest. The data shown are for $f = 0.5$, and we note that this behavior is qualitatively the same for other values of f .

Figure 2 elucidates this finding in more quantitative terms. The upper panel shows the FWHM duration of the pulse (with all nonlinear interactions on) as measured in the focus. Graphs obtained for different pulse energies demonstrate that the optimal chirp setting resulting in the shortest pulse is the same as in the linear regime. This is not too surprising given the relatively tight focusing geometry. A more unexpected result is shown in the lower panel in Fig. 2. Here, the spectral width is quantified in terms of the quadratic deviation (in units of the central angular frequency) as if the spectrum was a probability distribution function. We have adopted this particular measure to deal with the varying spectral shapes (Fig. 1). The figure shows that the maximal spectral broadening *does not* occur for the minimal-duration pulses. The precise chirp setting that leads to the broadest spectrum may depend on how exactly the spectral width is quantified, but it is clear that the *broadest spectrum does not imply the shortest pulse*. Therefore, adjusting the pulse chirp to produce maximal spectral width does not ensure that the pulse duration will be shortest in the focus.

In [7], this criterion of maximal spectral broadening was used to presumably minimize the pulse duration

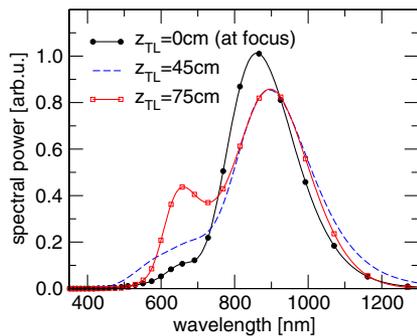


Fig. 1. Simulated spectra detected beyond the nonlinear focus of a few-cycle pulse. z_{TL} represent varying chirp settings—values mark locations at which a minimum-duration pulse would occur in the absence of nonlinear interactions. For $z_{TL} = 0$ cm and $z_{TL} = 75$ cm, the transform-limited pulse forms in the linear focus and at the focusing mirror, respectively.

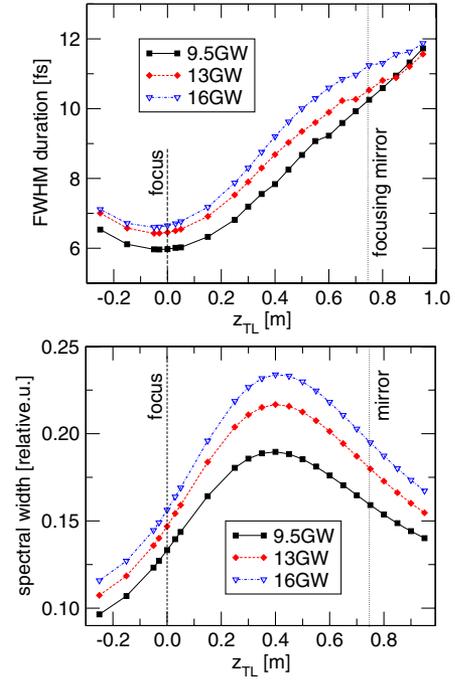


Fig. 2. Pulse duration in the nonlinear focus (top) and spectral width (bottom) as functions of the initial pulse chirp setting, expressed as a position at which a bandwidth-limited pulse would occur in the absence of nonlinear interactions. Contrary to intuition, maximal spectral broadening *does not* occur in the shortest-duration pulses.

at the focus location, and the peak power was estimated from the pulse energy and the pulse duration. One consequence of our counterintuitive finding is that such an approach can result in a wrong scale on the peak-power axis. Consequently, the apparent increase in P_c should not be interpreted as solely due to the influence of delayed nonlinearity.

Next we address the role of the vibrational Raman effect in simulations inspired by the experiment of [7]. For several values of f , we record the position $z_{\max}(\log(P_i))$ of the peak in the plasma density generated around the nonlinear focus as a function of the initial peak power. The characteristic shape of $z_{\max}(\log(P_i))$ shows a crossover from an almost constant function at low powers to a linearly decreasing (in log scale) function for higher powers [12]. It has been shown [13] that the crossover power P_x can be significantly smaller than the nominal P_c as calculated from the nonlinear index n_2 . Nevertheless, it can reflect an increase in the effective self-focusing threshold in short pulses. In particular, as the pulse duration shortens below the molecular vibrational period, P_x is expected to shift to higher values.

This behavior is depicted in Fig. 3. It shows results for two settings of the initial chirp, and three values of $f = 1.0, 0.5, 0.0$. The bottom-row data were obtained for the initial chirp adjusted such that the minimal pulse duration is obtained in the focus. Comparison of different columns reveals dependence on the contribution of the vibrational Raman effect. The crossover power P_x (defined as the intersection of linear fits shown in Fig. 3) increases from 5.4 GW at $f = 1.0$ to 7.7 GW for $f = 0.0$. This trend is in line with expectations based

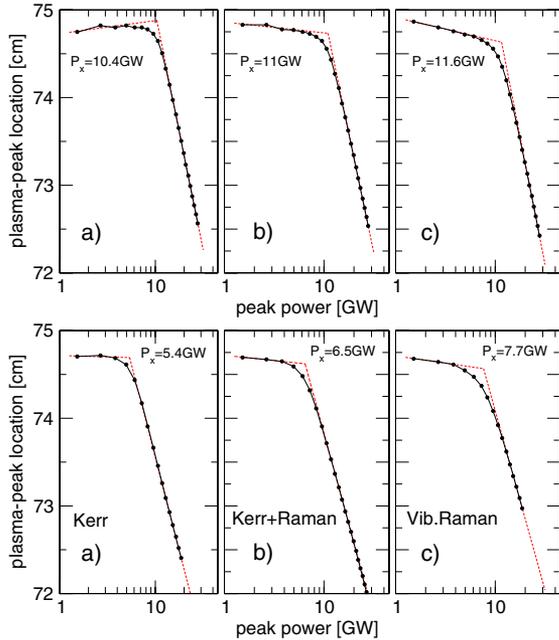


Fig. 3. Location of the maximal plasma density as a function of the initial peak power. Instantaneous to total (i.e., including vibrational) nonlinearity ratio $f = 1.0, 0.5$ and 0.0 in the left, middle, and right columns, respectively. Top row: $z_{TL} = 75$ cm. Bottom row: $z_{TL} = 0$ cm.

on qualitative modeling of self-focusing in Kerr–Raman media [14]. Varying f mainly affects the high-power portion of the crossover curve, which is the part that can be obtained more accurately in experiment.

The data in the top row illustrate what happens when the pulse duration is not minimized at the focus. Here, the transform-limited pulse forms at the parabolic mirror, and resulting pulse durations are therefore longer in the nonlinear focus. This is why P_x shifts to higher values. Longer pulses are also less efficient in impulsive excitation of the vibrational Raman response, and this manifests in a weaker dependence of P_x on f .

Comparing plots between rows, we see that the pulse chirp setting shows the strongest influence for pure Kerr nonlinearity, and its influence weakens with the increasing strength of the vibrational nonlinearity. The overall shape of the focus-location curve also changes with f . The crossover becomes more gradual and the low-energy portion more sloped as the contribution of the vibrational Raman effect increases.

In conclusion, we have presented comparative numerical simulations of self-focusing dynamics in few-cycle pulses. To readers interested in simulations, we have pointed out that focused few-cycle pulses require more

realistic initial conditions than those normally used in longer pulses. Contrary to intuition, maximal spectral broadening does not imply minimal pulse duration in the nonlinear focus. We therefore caution against the method adopted in [7], which relied on spectral broadening alone as an indicator of a shortest-duration pulse. This should be important for interpretation and design of future experiments employing ultrashort pulses. Our investigations indicate that pulses with durations of a few femtoseconds are sensitive to the ratio between the instantaneous and vibrational components of the nonlinear index. We speculate that the shift of the nonlinear focus location versus the pulse energy, measured for pulses with different chirps, can be used to estimate the contribution of the vibrational Raman effect. For example, Fig. 3 implies that $P_x(z_{TL})$ as a function z_{TL} depends on the value of f . An estimate of the latter will be important for simulation of extremely short pulses, and also for modeling of self-compression dynamics in longer-duration pulses.

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