On the relative roles of higher-order nonlinearity and ionization in ultrafast light-matter interactions

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Far off-resonant ultrafast and nonlinear light-matter interactions are studied using a one-dimensional atomic model. Results from a pump-probe diagnostic reveal that any higher-order nonlinear refraction is masked by ionization-induced defocusing before it becomes significant. On the other hand, we show that signatures of a higher-order nonlinearity may still be manifest via low-order harmonics of the pump center frequency. Implications for filamentation of femtosecond pulses are pointed out. © 2012 Optical Society of America

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Although filamentation of femtosecond high-intensity pulses has attracted interest for many years, a complete understanding supported by a self-consistent theory is yet to be achieved. A topic of particular contention involves the higher-order Kerr effect (HOKE) in atomic and molecular gases [1,2]. Key attributes claimed for the HOKE include ultrafast nonlinear changes in refractive index that are higher than linear in intensity, that it changes sign for high enough intensity, and that it can occur before significant ionization appears. Observations of the HOKE spurred a revision of the accepted paradigm for femtosecond filamentation [3], suggesting that the role of plasma in filaments is not as important as previously thought. A number of papers followed, both supporting (e.g., [4–6]) and rejecting (e.g., [7–10]) this view.

Given that the HOKE could be a game changer in our understanding of filamentation, there has been relatively little theoretical elucidation of its microscopic origins. Teleki *et al.* [11] studied nonlinear refraction in gases using a one-dimensional (1D) atomic model and found nonlinear refraction akin to the HOKE in the quasi-static limit, but conjectured that plasma defocusing would effectively mask the HOKE. In contrast, Bree *et. al* [6] found that the HOKE can precede significant ionization based on the nonlinear Kramers–Kronig relations applied to multiphoton ionization of the atoms. Most recently, Volkova *et al.* [12] have performed three-dimensional (3D) quantum simulations of a silver atom and conclude that they find no reason to modify the existing paradigm of filamentation.

In this Letter, we study the relative roles of the HOKE and ionization in gases using the 1D atomic model of [11] comprising an electron in an attractive delta-function potential that approximates the electron-ion Coulomb potential. This model, which has a single bound state, the ground atomic state, and a continuum of freed electron states with positive energies, serves as an exactly soluble, albeit approximate, model for light-matter interactions involving ground-continuum transitions. There are two main reasons for the relevance of this study: First, it has been suggested that the HOKE arises precisely from ground-continuum atomic transitions, ground-bound state transitions yielding at most a saturable self-focusing nonlinearity. The 1D atomic model therefore provides a means to assess whether ground-continuum atomic transitions can produce the features of the HOKE alluded to in the first paragraph, and we find it cannot. On the other hand, we present evidence that signatures of a higher-order nonlinearity can be found in the low-order harmonics of the pump center frequency. Second, 3D atomic quantum simulations, e.g., [12,13], involve separating the nonlinear response from the much larger linear response in the full optical response, and this can be prone to numerical issues. It was recently shown in [14] that the 1D atomic model allows for an exact solution for the nonlinear optical response, making it immune to numerical issues, so that qualitative comparison between the more precise 3D atomic model and the reduced 1D atomic model can serve to bolster the conclusions of [11,12].

Our diagnostic approach involves a pump-probe scheme involving the electric field profile shown in Fig. 1. Specifically, we drive the 1D atomic system with a strong pump pulse with a region of constant intensity, and linear leading and trailing edges. At the same time, the system is exposed to a much weaker probe, the duration of which exceeds that of the pump. In this way, we may sample the

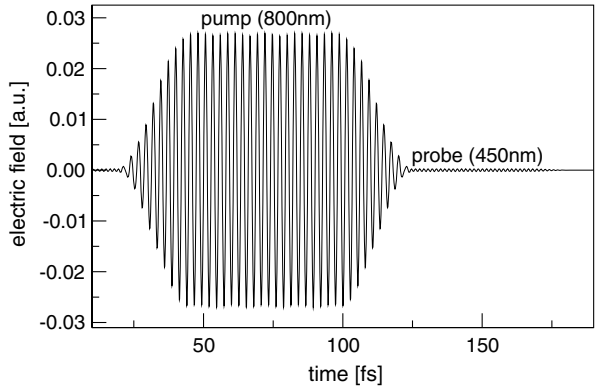

Fig. 1. Electric field of the pump-probe waveform. The probe ($\lambda = 450$ nm) appears as a weak background with the duration extending before and after the much stronger pump pulse ($\lambda = 800$ nm).

system response at times before, during, and after the pump pulse. Physically, our goal is to extract the *effective* susceptibility experienced by the probe due to the modification of the medium by the strong pump.

The time-dependent susceptibility is defined operationally through the following procedure: For a given temporal profile of the electric field $E(t) = E_{\mathrm{pump}}(t) + E_{\mathrm{pr}}(t)$, we calculate the nonlinear current J induced in the 1D atomic system using the exact solutions [14], then a second calculation is done with the pump alone. The difference of the two is therefore the response of the pump-affected system to the probe:

$$J_{\mathrm{pr}}(t) \equiv J(E_{\mathrm{pump}}, E_{\mathrm{pr}}) - J(E_{\mathrm{pump}}, 0) \approx \frac{\delta J}{\delta E_{\mathrm{pr}}} E_{\mathrm{pr}}. \quad (1)$$

For high intensities, $J(t)$ exhibits rich spectra including high-harmonics (see Fig. 2), but here we concentrate on the response around the frequency of the probe itself.

From the current probe response $J_{\mathrm{pr}}(t)$, we extract the component $J_{\mathrm{filt}}(t)$ corresponding to frequencies in the vicinity of the probe center frequency by applying a filter in the spectral domain with a bandwidth of one quarter of the center frequency. Having isolated $J_{\mathrm{filt}}(t)$ from its background, the polarization response $P_{\mathrm{filt}}(t)$ is obtained by integration in time. This then serves to define the effective nonlinear optical susceptibility by dividing the complex-valued analytic signals of the nonlinear polarization and probe (i.e., quantities for which $P_{\mathrm{filt}}(t) = \mathrm{Re}\{\mathcal{P}_{\mathrm{filt}}(t)\}$, $E_{\mathrm{pr}}(t) = \mathrm{Re}\{\mathcal{E}_{\mathrm{pr}}(t)\}$):

$$\Delta\chi(t) \equiv \mathcal{P}_{\mathrm{filt}}(t)/\mathcal{E}_{pr}(t). \quad (2)$$

The real part of this susceptibility reflects the time evolution of the nonlinear refractive index experienced by the probe. Note that this is a way to characterize a single atom response, and that propagation effects (i.e., phase matching) also affect the actually generated radiation.

If we denote by I_{sat} the intensity at which HOKE starts to manifest itself, then for peak pulse intensities $I < I_{\mathrm{sat}}$, we expect that the effective susceptibility will be proportional to the local pulse intensity. This case is illustrated qualitatively by the dashed line in Fig. 3 for the HOKE behavior shown on the left and for the pump-probe pulse in Fig. 1. In the first scenario in which HOKE can indeed manifest itself before ionization occurs, then for $I > I_{\mathrm{sat}}$, the qualitative variation of the effective susceptibility with time should take the form shown as the thin solid line in Fig. 1: The characteristic dip in the time variation of the time-dependent susceptibility appears since for $I > I_{\mathrm{sat}}$, the HOKE should depress the nonlinear susceptibility for the highest intensities in the pulse. For even higher intensities $I > I_c$, I_c being the intensity at which the HOKE reverses sign, the time variation of the susceptibility takes on the generic form shown as the thick solid line in Fig. 1.

In contrast, for the second scenario in which a combination of the standard Kerr effect plus defocusing due to ionized electrons provides a qualitative model, we would expect to see that the nonlinear susceptibility would increase monotonically in time until ionization occurs, after which the nonlinear susceptibility would decrease monotonically until the pump pulse terminates. After the pump pulse, a negative susceptibility persists as long as the freed electrons remain. For low intensities such that ionization does not occur, the time variation should be the same as the dashed line in Fig. 3.

Next we turn to which of the above two scenarios best reflects the results of our exact simulations based on the 1D atomic model for the nonlinear current and polarization. Figure 4 shows that we observe the second or standard scenario. While at lower intensities, the susceptibility follows the intensity envelope of the pump as expected for the Kerr effect, the almost linear decrease with time for higher intensities is clearly due to increasing number of "freed electrons," which produces defocusing or negative nonlinear refraction. Importantly, this behavior is qualitatively independent of the ionization energy of the system. That is, for higher E_g, the system exhibits weaker nonlinearity, and the ionization onset occurs at higher intensities. Our pump-probe diagnostic based on the 1D atomic model does not exhibit significant manifestations of the HOKE in the frequency

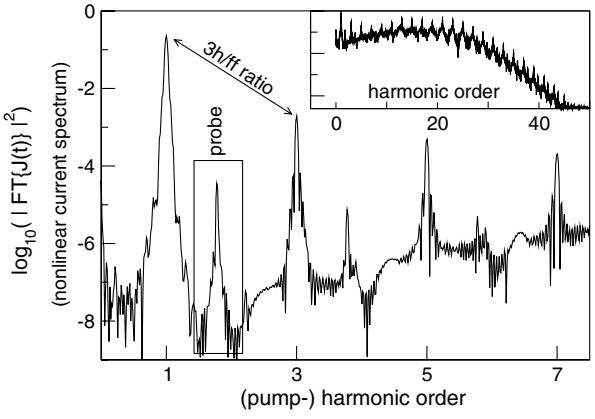

Fig. 2. Log-scale spectrum of the nonlinear current, $\log_{10}(|\mathrm{FT}\{J(t)\}|^2)$, with the inset showing its high-harmonic extent. The effective susceptibility is extracted from the vicinity of the fundamental probe frequency marked by a rectangle. Another interesting quantity is the ratio between the third- and fundamental-frequency powers indicated by the arrow.

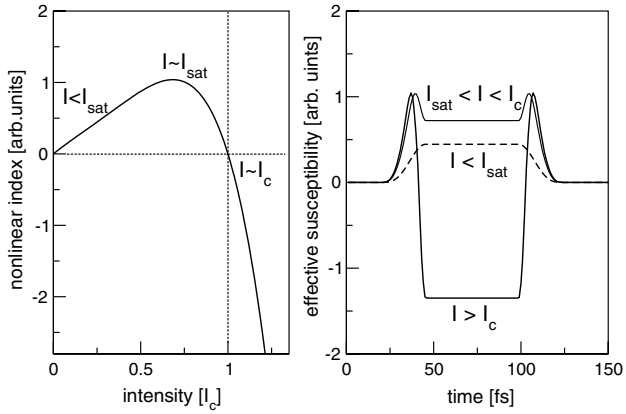

Fig. 3. Qualitative schematics of the effective time-dependent susceptibility (right) in a system with an instantaneous HOKE-like nonlinearity (left). Bat-ear features appear at both the leading and the trailing edge of the pump pulse for a sufficiently high intensity.

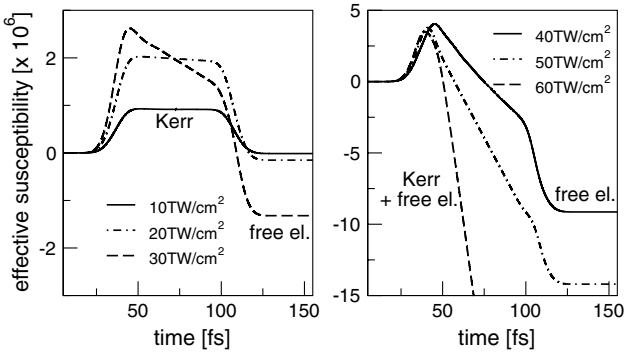

Fig. 4. The effective time-dependent susceptibility experienced by the probe at lower (left) and higher (right) pump intensities. Data shown for the ground-state energy of $E_g = 10$ eV. The same qualitative behavior is observed for other E_g values. Labels (Kerr, free electrons) indicate main contributing effects.

band of the probe field, in agreement with the conclusions arrived at based on the 3D atomic simulations of [12].

From the above observations, it seems that the effective susceptibility could be well approximated by the model currently used in filamentation modeling. At the same time, it has been shown that the nonlinear behavior of this model in the quasi-static regime deviates from the Kerr effect [11], raising the question of whether any signatures of higher-order nonlinearity can survive.

The answer is in the affirmative, and can be readily deduced from the example in Fig. 2, which shows that the peaks corresponding to the fundamental and third harmonic frequency have different heights. This is incompatible with the standard instantaneous Kerr effect plus the Drude plasma-like response from free electrons, and thus signals the influence of higher-order nonlinearities. Indeed, the Drude plasma does not contribute to the third harmonic at all, and the ratio between the spectral power of the fundamental and the third harmonic due to Kerr effect must be one. To see this, consider a local field $E_0 \cos(\omega t)$, for which the instantaneous Kerr polarization would be $P = \epsilon_0 \bar{n}_2 N_a E_0^3 \cos(\omega t)^3$, and the corresponding nonlinear current density is obtained as $J = \partial_t P = 3/4\epsilon_0 \omega \bar{n}_2 N_a E_0^3 [\cos(\omega t) + \cos(3\omega t)]$, showing that the ratio between the strength of the third and the fundamental harmonics of the *nonlinear current* is 1 (while it is 1/9 in the nonlinear polarization). Note that the effective susceptibility only describes the *source* that appears in the Maxwell's equations, and not directly the generated radiation. The quantitative assessments of this effect on the harmonic generation requires a full propagation analysis, and will be discussed elsewhere.

In conclusion, utilizing exact solutions of a simple 1D atomic model to describe the nonlinear response induced by a strong optical pulse, we have shown that the complex nonlinear behavior found in the quasi-static regime becomes masked at the fundamental frequency by the onset of ionization. The continuum states overwhelmingly dominate the defocusing part of the response at high intensity. Deviations from the usual nonlinear behavior, i.e., one described in terms of an instantaneous Kerr effect and plasma defocusing, are found in the lower-harmonic behavior. This will affect the conversion into third and fifth harmonic radiation. Qualitatively similar behavior is expected in femtosecond filaments for the part of the nonlinear response that couples bound and continuum electronic states. Because the strength of these effects increases with decreasing ionization potential, we expect that the excited states present in the filament plasmas could also provide significant contributions to the overall nonlinearity of the medium.

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