Scale-invariant nonlinear optics in gases: supplementary material

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1. NUMERICAL METHODS

A. Filamentation

For the filamentation simulations, a uni-directional nonlinear envelope equation (the corresponding envelope equation of Eq. (1) in the main article but using a less restrictive paraxial approximation) was numerically integrated [1]. The equation includes the full pressure and frequency dependence of the wave number in the diffraction and the nonlinear term and is valid for pulses down to the single-cycle regime [2]. Note that the use of an envelope equation is not in contradiction to our discussion; the scaling can be formally shown both for envelope and field equations. The propagation equation follows from the nonlinear Helmholtz equation, adapting the method suggested by Feit and Fleck to introduce uni-directionality [3]. Argon is used as nonlinear medium and dispersion is treated by a Sellmeier-type expression for the linear refractive index [4]. Self-focusing originating from a Kerr-type nonlinearity is described by a nonlinear refractive index $n_2 = 0.98 \times 10^{-19} \text{ cm}^2\text{W}^{-1}$ for Ar at atmospheric pressure [5]. The interaction with the generated plasma, i.e. absorption and plasma defocussing, is treated in terms of a current density, whereas free electrons are generated via field ionization, numerically described within the generalized Keldysh-PPT theory (Perelomov-Popov-Terent'ev) [6, 7]. Avalanche ionization is included via a Drude-type model with a

phenomenological collision time of $\tau_C = 190 \text{ fs}$ at atmospheric pressure [8].

B. HHG

The numerical HHG simulations take both, propagation phenomena (phase matching effects) as well as the single atom dipole emission at the high harmonic frequencies into account and are performed in two steps. In the first step the generating laser field is propagated through the cylindrically symmetric gas medium, taking into account the time- and space-dependent neutral and plasma dispersion and optical Kerr effect, as described in Refs. [9, 10]. Then, in the second step the harmonic field is propagated using the generated high-order polarization as source term calculated using the strong-field approximation [11]. For the propagation of the harmonic field neutral dispersion and absorption are taken into account. The wave equations for both fundamental and harmonic fields are derived carrier resolved in paraxial approximation. In both equations a nonlinear source term proportional to the gas density is assumed. For the fundamental field absorption resulting from optical field ionization is taken into account (also scaling linearly with gas density), while neutral and plasma absorption are neglected. The numerical code has been verified by comparison with experimental results obtained with the intense harmonic beam line at Lund University.

C. Scaling error calculation

The normalized relative scaling errors for intensity $I_n = I_n(r, t)$ and fluence $F_n = F_n(r, z) = \int_{-\infty}^{\infty} I_n(r, t, z) dt$ displayed in Fig. 2 were calculated for the input parameter set $(\epsilon_{\text{in}}^n, p_n, f_n)$ and referenced to the corresponding parameter set with four times higher input pulse energy, all other parameters being adjusted according to the scaling relations, i.e. $(\epsilon_{\text{in}}^{n-1} = 4\epsilon_{\text{in}}^n, p_{n-1} = p_n/4, f_{n-1} = 2f_n)$. For the intensity and the fluence the normalized errors can be written as:

$$v_{\text{int}}^n = \frac{2 \int_{-\infty}^{\infty} dt \int_0^{\infty} dr r |I_n - I_{n-1}|}{\int_{-\infty}^{\infty} dt \int_0^{\infty} dr r I_n + \int_{-\infty}^{\infty} dt \int_0^{\infty} dr r I_{n-1}}, \quad (1)$$

$$v_{\text{flu}}^n = \frac{2 \int_{-\infty}^{\infty} dz \int_0^{\infty} dr r |F_n - F_{n-1}|}{\int_{-\infty}^{\infty} dz \int_0^{\infty} dr r F_n + \int_{-\infty}^{\infty} dz \int_0^{\infty} dr r F_{n-1}}. \quad (2)$$

The integrals were computed numerically on a finite grid in r and z using a normalized grid size such that $\Delta r/W_0 = \text{constant}$ and $\Delta z/z_R = \text{constant}$ with Δr and Δz denoting the grid unit size.

2. EXPERIMENTAL METHODS

The experiments were performed with a Titanium:Sapphire chirped pulse amplification laser chain, delivering up to 5 mJ pulses, with 20 fs duration (FWHM), centered around 800 nm, at a repetition rate of 1 kHz. The beam size was adapted to a diameter ($1/e^2$ of the intensity profile) of approx. 11 mm with a reflective telescope, followed by a 10 mm aperture. The filaments were generated inside an Ar-filled tube using spherical mirrors of different focal lengths ($f = 0.5, 0.75, 1, 1.5, 2, 2.5$ m). The tube was terminated with thin (0.5 mm) fused silica windows in order to minimize the impact of dispersion and undesired nonlinear interaction. The Ar pressure was set according to the scaling law ($p = 8, 3.5, 2, 0.98, 0.50, 0.32$ bar), while the pulse energy was fine-tuned with a $\lambda/2$ -plate followed by a transmissive polarizer in order to match the super-continuum spectra originating from the filament to best possible agreement for the different combinations of pulse energy, Ar pressure, and focal length. For the lowest input pulse energy, a second polarizer plate was installed in order to improve the polarization contrast.

Behind the filament tube, the bright center of the filament was selected with an aperture and sent to a pulse compressor, comprised of movable fused silica wedges and chirped mirrors in double-angle configuration (Ultrafast Innovations). The pulses were characterized using the dispersion scan technique (d-scan) [12]. The d-scan is based on recording the second harmonic spectrum as function of dispersion, in our case realized by moving one of the wedges. The spectral phase is retrieved with a numerical algorithm that iteratively reproduces the measured d-scan trace (second harmonic spectrum vs. wedge insertion) based on a measured fundamental spectrum, see Fig. S1. For the temporal pulse retrievals shown in Fig. S1 (a), a wedge insertion was chosen corresponding to a minimized pulse duration and best agreement with pulse profiles retrieved for other parameter combinations. Note that an offset third-order phase remains uncompensated in the employed compression scheme, as visible in the d-scan traces, showing a clear variation of spectral frequency distribution as a function of wedge insertion.

To ensure, that the focused pulses enters the nonlinear interaction regime with approximately identical phase properties, the

Fig. S1. D-scan pulse retrieval: (a), Measured second harmonic intensity as a function of wedge insertion (vertical scale with arbitrary offset). The displayed data were recorded with $f = 0.75$ m, $\epsilon_{\text{in}} = 0.26$ mJ and $p = 3.5$ bar, see also the corresponding spectrum and pulse retrieval in Figure 3 in the main article. (b), Corresponding numerically retrieved d-scan trace. The dashed line indicates the wedge insertion minimizing the pulse duration.

second order phase of the laser pulse was slightly adjusted with a Dazzler between the different filament configuration. The adjustment takes into account, that the tube length was changed as $L \propto \eta$ (and accordingly $f \propto \eta$) while the scaling model requires $L \propto \eta^2$ (and $f \propto \eta^2$). For different filament configurations, this implies a change of the linear dispersion upon propagation of the laser pulse though the tube. However, as the beam diameter before focusing D was kept constant (the scaling model predicts $D \propto \eta$), the focusing geometry f/D was correctly changed according to the scaling model.

3. DETAILS ON THE SCALING LIMITS

As mentioned in the main article, deviations from exact scale-invariance arise at high gas densities. Here, we discuss these limits and their relevance for nonlinear processes like filamentation and harmonic generation.

We start from the Forward Maxwell Equation (Eq. (3) in the main article):

$$\left[\frac{\partial}{\partial z} - \frac{i}{2k(\omega, \rho)} \Delta_{\perp} - iK(\omega, \rho) \right] \hat{\mathcal{E}}(r, z, \omega) = \frac{i\omega^2 \hat{\mathcal{P}}_{\text{NL}}(r, z, \omega, \rho)}{2k(\omega, \rho)c^2\epsilon_0}. \quad (3)$$

The interaction with the generated plasma is described via a current density, which can be included into the medium's nonlinear response through the transformation $\hat{\mathcal{P}}_{\text{NL}} \rightarrow \hat{\mathcal{P}}_{\text{NL}} + i\hat{\mathcal{J}}/\omega$.

Applying the scaling relations derived in the main article, deviations from exact scale-invariance of Eq. (3) arise mainly from a not exactly linear scaling of the mediums' linear and nonlinear responses with gas density. The physical quantity in Eq. (3) that carries explicit gas density dependence is the linear refractive index n [entering via $k(\omega, \rho) = n(\omega, \rho)\omega/c$]. Furthermore, the nonlinear response typically contributes additional density dependencies (see below), for example via the nonlinear refractive index n_2 and the collision frequency ν_c . The pressure dependence for n and n_2 can be evaluated via the atomic polarizability

α and the hyperpolarizability $\langle \gamma \rangle$ [13]:

$$(n + n_2 I)^2 - 1 \approx \rho \alpha + \frac{3 \rho \langle \gamma \rangle I}{8 \epsilon \epsilon_0^2 n_0}. \quad (4)$$

The pressure dependent terms in Eq. (3) that contribute to linear propagation can be written as:

$$n(\omega, \rho) \approx 1 + \frac{\rho \alpha}{2} - \frac{\rho^2 \alpha^2}{8}, \quad (5)$$

$$k(\omega, \rho) \approx \frac{\omega}{c} \left(1 + \frac{\rho \alpha}{2} - \frac{\rho^2 \alpha^2}{8} \right), \quad (6)$$

$$K(\omega, \rho) = k(\omega, \rho) - k(\omega, 0) \approx \frac{\omega}{c} \left(\frac{\rho \alpha}{2} - \frac{\rho^2 \alpha^2}{8} \right), \quad (7)$$

$$k^{-1}(\omega, \rho) \approx \frac{c}{\omega} \left(1 - \frac{\rho \alpha}{2} - \frac{\rho^2 \alpha^2}{8} \right). \quad (8)$$

Accordingly, for n_2 we obtain:

$$n_2 \approx \frac{3 \rho \langle \gamma \rangle}{16 \epsilon \epsilon_0^2 n_0} - \frac{1}{8} \left(\frac{3 \rho \langle \gamma \rangle}{16 \epsilon \epsilon_0^2 n_0} \right)^2 I \quad (9)$$

with I denoting the intensity.

$K(\omega, \rho)$ is proportional to gas density in leading order, as required for the scaling formalism. Higher (than linear) order contributions to the gas density dependence are generally very small, i.e. for Ar at a pressure of 10 bar the second-order correction of $k(\omega, \rho)$ is of relative magnitude 4×10^{-6} , while the first order amounts to 3×10^{-3} . Consequently, the quantity $k^{-1}(\omega, \rho)$, entering equation 3 via the diffraction and the nonlinear terms, can be considered gas density independent in good approximation.

The scalability of the nonlinear propagation terms [the right hand side of Eq. (3)] has to be evaluated individually for each term contributing to the nonlinear propagation phenomenon considered. For filamentation, the relevant contributions are: Kerr-effect, plasma defocusing, and nonlinear absorption resulting from optical field ionization and avalanche ionization, as listed below:

$$\begin{aligned} \frac{\hat{P}_{\text{NL}} + \frac{i \hat{J}}{\omega}}{2 \epsilon_0} = & \underbrace{n_0(\rho) n_2(\rho) (1 - \rho_e / \rho) I \mathcal{E}}_{\text{Kerr effect}} \\ & - \underbrace{\frac{1}{2 \rho_c} \frac{\omega^2}{v_c^2(\rho) + \omega^2} \widehat{\rho_e \mathcal{E}}}_{\text{Plasma defocusing}} \\ & + i \underbrace{\frac{1}{2 \omega \rho_c} \frac{v_c(\rho) \omega^2}{v_c^2(\rho) + \omega^2} \widehat{\rho_e \mathcal{E}}}_{\text{Absorption due to avalanche ionization}} \\ & + \underbrace{\frac{i}{\omega} \frac{n_0(\rho) c U_i}{2} \left[\frac{(\rho - \rho_e) W_{\text{OFI}}}{I} \mathcal{E} \right]}_{\text{Absorption due to field ionization}}. \quad (10) \end{aligned}$$

Here $\rho_e(r, z)$ is the plasma density, and $\rho_c = \epsilon_0 m \omega^2 / e^2$ the critical plasma density with m and e denoting electron mass and charge, respectively. W_{OFI} is the optical field ionization rate and U_i is the ionization potential of the gas medium. Exact

scalability according to our formalism can be achieved if all terms entering Eq. (10) are proportional to gas density. This implies, that the ionization ratio must be conserved upon scaling, i.e. $\rho_e(r, z) / \rho = \rho_e(r / \eta, z / \eta^2) / \rho = \text{const.}$. Note, that ρ_e and thus ρ_e / ρ can still vary rapidly in space and time. We briefly discuss below how the different terms scale with gas density.

The first term in Eq. (10), describing the Kerr effect, scales proportional to gas density in leading order. Note that the correction $(1 - \rho_e / \rho)$, which takes into account that the Kerr nonlinearity mainly originates from the neutral medium (the contribution from the ionic species is usually much weaker), is independent from the density, if the ionization ratio is conserved.

The second term in Eq. (10), describing plasma defocusing is proportional to ρ_e and thus to ρ (if $\rho_e / \rho = \text{const.}$). Deviations from perfect scaling can occur because of the density dependence of the collision frequency ν_c . Within the framework of the Drude model, ν_c is a phenomenological quantity describing the frequency of collisions for a free electron in the plasma with heavier particles (atoms and ions). The time scale for avalanche ionization, i.e. the time for a free electron in the plasma to gain sufficient kinetic energy from the laser field in order to impact ionize another atom, is on the order of $1 / \nu_c$. For ν_c we assume a simple linear dependence on gas density [14]. For atmospheric pressure in argon $1 / \nu_c \approx 190$ fs; thus for wavelengths in the UV to near-infrared spectral range, usually used for nonlinear optics applications, the laser angular frequency is much larger than the collision frequency ($\omega \gg \nu_c$) and the density dependence of ν_c will not influence plasma defocusing significantly. For much higher pressure however, such that $\omega \approx \nu_c$, the gas density dependence of ν_c will lead to deviations from exact linear density dependence. Furthermore, in the extreme case that the plasma density ρ_e overcomes the critical density ρ_c the plasma will become opaque. In this regime, where back reflection from the plasma can occur, unidirectional propagation equations are not valid. For wavelength in the above stated range however this will not happen for pressures below several ten atmospheres ($p \gtrsim 70$ bar at room temperature for $\lambda = 800$ nm).

The third term in Eq. (10), describing nonlinear absorption of power due to avalanche ionization, is the only contribution, which does not scale linearly with gas density. This imposes a rather strict limitation for the validity of the scaling to conditions, where avalanche ionization is not important for the nonlinear interaction with the gas medium, i.e. conditions where either ionization generally is weak (at low intensity) or avalanche ionization is weak compared to optical field ionization, as is the case for short pulses or low gas density ($\tau \ll 1 / \nu_c$, where τ is the pulse duration). Both conditions are fulfilled for the parameter regime usually applied for filamentation and HHG, where often short laser pulses (typically below 100 fs) are employed and where the ionization ratio is usually rather low (typically $\rho_e / \rho < 0.1$).

The last term in Eq. (10), describing optical field ionization, is linearly proportional to gas density in good approximation.

To include processes like harmonic generation and wave-mixing an appropriate source polarization has to be added to the right-hand side of Eq. (10). Similar to the Kerr contribution, such terms scale approximately proportional to gas density in full agreement with our scaling formalism.

The validity of $\rho_e / \rho = \text{const.}$ can be evaluated via a rate equation, coupled to Eq. (10) and describing plasma generation

due to field and avalanche ionization:

$$\frac{\partial \rho_e}{\partial t} = (\rho - \rho_e)W_{\text{OFI}}(|\mathcal{E}|) + W_{\text{AVI}}(|\mathcal{E}|)\rho_e. \quad (11)$$

Optical field ionization is usually treated by the generalized Keldysh-PPT theory [6, 7] and the rate for avalanche ionization $W_{\text{AVI}} \equiv (\omega^2 I / \rho_c c n_0) [\nu_c / (\omega^2 + \nu_c^2)]$ follows from the Drude model [15]. While the first term on the right hand side of equation 11 scales linearly with gas density, thus confirming $\rho_e / \rho = \text{const.}$, the second term describing avalanche ionization does not show this linear dependence. Consequently, the ratio between optical field ionization and avalanche ionization changes with gas density. In extreme conditions, i.e. at high intensities if long pulses and/or high gas densities are used ($\tau \approx 1/\nu_c$), the ionization ratio will not be conserved upon scaling ($\rho_e / \rho \neq \text{const.}$), resulting in deviations from exact scale invariance.

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