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Quasi-phase-matched high harmonic generation in gas-filled hollow-core photonic crystal fiber: supplementary material

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1. MODEL

A. On-axis HHG yield

The longitudinal on-axis build-up of the q^{th} harmonic in a PCF is the coherent sum over the emitted fields of single generation events along the z-axis of the waveguide. At the exit of the PCF ($z = L$), the harmonic signal can be written as:

$$I_q(L) \propto \left| \int_0^L S_q P \exp[-i(\int_0^z q k_f(z')dz' + \int_z^L k_q(z')dz')] \exp[\alpha(L-z)] dz \right|^2 \quad \text{(S1.1)}$$

where $S_q(z)$ is the nonlinear source term, $P(z)$ is the gas pressure, $k_f(z)$ and $k_q(z)$ are the propagation constants of fundamental and harmonic field and $\alpha(z)$ is the attenuation constant for the q^{th} harmonic order in argon. In case of a uniform gas-fill and no amplitude modulation of the driver pulse intensity, S_q, P, k_f, k_q and α are constants and (S1.1) simplifies to the known form [1]:

$$\begin{aligned} I_q(L) &\propto \left| \int_0^L S_q P \exp[-iqk_f z - i(k_q L - k_q z)] \exp[\alpha(L-z)] dz \right|^2 = \\ &\left| S_q P \exp(-ik_q L) \exp(\alpha L) \int_0^L \exp(i\Delta k z) \exp(-\alpha z) dz \right|^2 = \\ &\left| S_q P \exp(\alpha L) \int_0^L \exp(i\Delta k z) \exp(-\alpha z) dz \right|^2 \end{aligned} \quad \text{(S1.2)}$$

Due to mode beating in the NC-PCF, the electric field amplitude is modulated longitudinally. The nonlinear source term is assumed to be $S_q = |\chi_q E(z)^{\gamma}|^2$ where $\gamma \sim 5$ [2,3]. The effective nonlinearity χ_q corresponds to the harmonic order q and is set to 1 for a qualitative evaluation of the HHG build-up.

The attenuation constant in argon $\alpha = \alpha_{\mathrm{atm}} P(z)/P_{\mathrm{atm}}$ varies longitudinally with pressure $P(z)$ where α_{atm} is the attenuation constant in argon at atmospheric pressure P_{atm}.

Different gas pressures at the entrance and exit of the NC-PCF, P_0 and P_L, lead to a longitudinal pressure profile in the NC-PCF along the spatial coordinate z which can be approximated as [4,5]:

$$P(z) \approx \sqrt{P_0^{\,2} + \frac{z}{L}(P_L^2 - P_0^2)}\ . \tag{S1.3}$$

We assume that most of the high order harmonics are generated from parts of the driver pulses propagating in the fundamental waveguide mode. At low driving laser intensity the propagation constants for the driving laser in the fundamental LP_{01} mode and for the q^{th} harmonic in presence of ionized gas in the waveguide can be written as [3]:

$$\begin{aligned} k_f &\approx \frac{2\pi}{\lambda_0} + \frac{2\pi(1-\eta)P\delta_0}{\lambda_0} - P\eta N_{\mathrm{atm}} r_{\mathrm{e}} \lambda_0 - \frac{u_{11}^2 \lambda_0}{4\pi r^2} \\ k_q &\approx \frac{2\pi}{\lambda_q} + \frac{2\pi(1-\eta)P\delta_q}{\lambda_q} - P\eta N_{\mathrm{atm}} r_{\mathrm{e}} \lambda_q \end{aligned} \tag{S1.4}$$

where r is the radius of the NC-PCF, δ is the real part of the refractive index at the driving laser wavelength λ_0, and at the wavelength of the q^{th} harmonic λ_q [6], N_{atm} is the number density of argon atoms at atmospheric pressure, r_{e} is the classical electron radius, u_{11} is the first zero of the Bessel function $J_0(x)$ and η is the ionization fraction. In the present model, we apply a constant ionization fraction of $\eta = 0.01$. We neglect the modal contribution of the harmonic beam since its Rayleigh length is longer than L.

For the assumed longitudinal pressure distribution (S1.3) the inner integrals in equation (S1.1) can be evaluated exactly. The outer integral is then approximated numerically by using the global adaptive quadrature method.

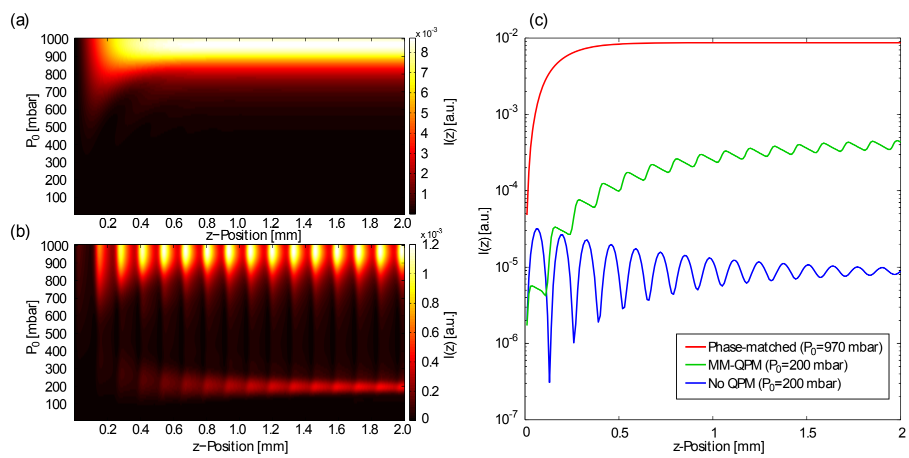

Fig. S1. Modeled on-axis build-up of H21 as a function gas pressure for a uniform gas-fill $P_0 = P_L$. (a) Intensity build-up of H21 at different pressures without MM-QPM. Phase matching of H21 is possible at P_0 = 970 mbar. (b) Build-up of H21 at different pressures in case of beating between the LP_{01} and LP_{13} mode. The enhanced HHG efficiency around P_0 = 200 mbar is the result of MM-QPM. (c) Non-phase-matched build-up of H21 at P_0 = 200 mbar without mode beating (blue curve). At the same pressure and with MM-QPM, the build-up efficiency is strongly enhanced (green curve). The red curve shows the build-up of H21 in case of phase-matched HHG.

In order to illustrate the HHG build-up in a NC-PCF with a uniform gas-fill at different pressures $P_0 = P_L$, we assume that a single driver pulse excites a single or a pair of modes at the entrance of the NC-PCF. The intensity build-up $I_{21}(z)$ of H21 at the entrance of the NC-PCF is illustrated in Fig. S1. The build-up model indicates that phase-matched HHG is possible at $P_0 = P_L = 970$ mbar in case the driver pulse propagates in a single waveguide mode (Fig. S1a). In case of mode beating between the LP_{01} and LP_{13} mode a strong increase in the build-up efficiency as a result of MM-QPM is observed for $P_0 = 200$ mbar (Fig. S1b). The phase-matched build-up at $P_0 = 970$ mbar is less efficient in this case since the driver pulse intensity is modulated longitudinally. Three different line-outs of Fig. S1a/b at different pressures are shown in Fig. S1c.

B. Model and experiment

We measured the harmonic spectrum H19-H29 in argon at an input pressure of 0.8 bar with IDC-QPM for the beating of the LP_{01} mode of the drive pulse with the LP_{02} mode of the control pulse and for the beating of the LP_{01} and LP_{21} mode. The pulse energy of control and drive pulse was 5 μJ. A comparison between relative harmonic brightness predicted by our model and HHG spectra measured for these two IDC-QPM settings is shown in Fig. S2.

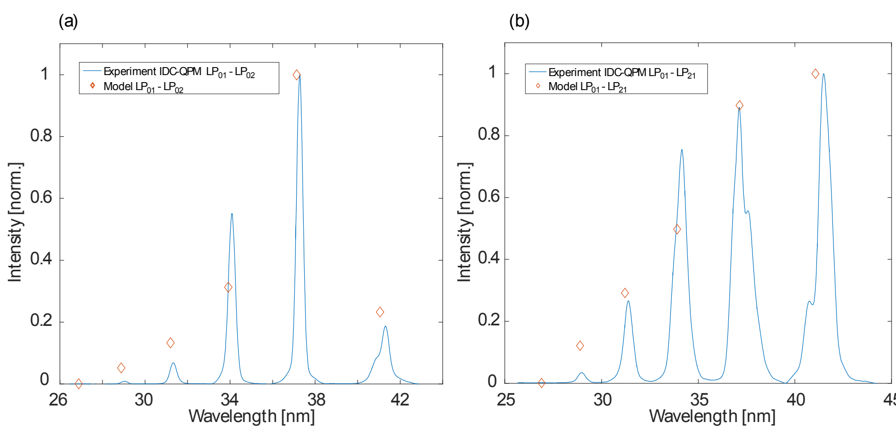

Fig. S2. Experimental HHG spectra and model predictions. (a) Harmonic spectrum with IDC-QPM of LP_{01} and LP_{02} mode (blue line) and relative harmonic brightness predicted by our model (orange diamonds). (b) Harmonic spectrum with IDC-QPM of LP_{01} and LP_{21} mode (blue line) and relative harmonic brightness predicted by our model (orange diamonds).

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