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**Title:** Efficient simulation of ultrafast quantum nonlinear optics with matrix product states

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Efficient simulation of ultrafast quantum nonlinear optics with matrix product states: supplemental document

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This document provides supplementary information to “Efficient simulation of ultrafast quantum nonlinear optics with matrix product states.”

1. DERIVATIONS OF SYSTEM HAMILTONIANS

In this section, we connect the dimensionless Hamiltonians in the main text, i.e., Eq. (1) for $\chi^{(3)}$ and Eq. (28) for $\chi^{(2)}$ nonlinear waveguides, to conventional lab-frame Hamiltonians in order to relate the normalized parameters to experimentally measurable ones.

A. $\chi^{(3)}$ nonlinear waveguide

The Hamiltonian for a $\chi^{(3)}$ nonlinear waveguide in the lab frame can be written as [1, 2]

$$
\hat{H}_{\text{lab}} = \hbar \int d\xi \left( \omega_0 \hat{\Phi}_\xi^+ \hat{\Phi}_\xi + \frac{i \omega'_0}{2} \left\{ \left( \partial_\xi \hat{\Phi}_\xi^+ \right) \hat{\Phi}_\xi - \hat{\Phi}_\xi^+ \left( \partial_\xi \hat{\Phi}_\xi \right) \right\} - \frac{\omega''_0}{2} \hat{\Phi}_\xi^+ \hat{\Phi}_\xi \hat{\Phi}_\xi^+ \hat{\Phi}_\xi - \frac{1}{2} \xi^{(3)} \hat{\Phi}_\xi^+ \hat{\Phi}_\xi \right),
$$

(S1)

where $\xi$ is the spatial coordinate, and $\hat{\Phi}_\xi$ is a local photon-polariton field annihilation operator with commutation relationships $[\hat{\Phi}_\xi, \hat{\Phi}_\xi^+] = \delta(\xi - \xi')$. We have approximated the energy dispersion of the waveguide $\omega(k)$ in the vicinity of the carrier wavevector (i.e., frequency) $k_0$ up to quadratic order as

$$
\omega(k) \approx \omega(k_0) + \omega'(k_0)(k - k_0) + \frac{1}{2} \omega''(k_0)(k - k_0)^2
$$

(S2)

and have written $\omega_0 = \omega(k_0)$, etc. The nonlinear coupling parameter $\xi(3)$ is related to the effective nonlinear coefficient $\gamma$ (with units of [power$^{-1}$ length$^{-1}$]) via $\xi(3) = \hbar \omega_0 \gamma v^2$ [1], where $v = \omega'_0$ is the group velocity at the carrier wavelength.

To further simplify the Hamiltonian, we move into a rotating frame given, which at lab-frame time $\tau$ is given by a unitary

$$
\hat{U} = \exp \left( -i \omega_0 \tau \int d\xi \hat{\Phi}_\xi^+ \hat{\Phi}_\xi \right).
$$

(S3)

As a result, we obtain a rotating-frame Hamiltonian $\hat{H}_{\text{rot}} = \hat{U}^\dagger \hat{H}_{\text{lab}} \hat{U} - i \hbar \frac{\partial}{\partial \tau} \hat{U}^\dagger \hat{U}$, which eliminates the trivial phase-rotation terms $\omega_0 \hat{\Phi}_\xi^+ \hat{\Phi}_\xi$ from the Hamiltonian Eq. (S1) so that

$$
\hat{H}_{\text{rot}} = \hbar \int d\xi \left( \frac{\iota \omega'}{2} \left\{ \left( \partial_\xi \hat{\Phi}_\xi^+ \right) \hat{\Phi}_\xi - \hat{\Phi}_\xi^+ \left( \partial_\xi \hat{\Phi}_\xi \right) \right\} - \frac{\omega''_0}{2} \hat{\Phi}_\xi^+ \hat{\Phi}_\xi \hat{\Phi}_\xi^+ \hat{\Phi}_\xi - \frac{1}{2} \xi^{(3)} \hat{\Phi}_\xi^+ \hat{\Phi}_\xi \right).
$$

(S4)

The dynamics of the field operators under Eq. (S4) are given by

$$
i \left( \partial_\tau + \iota v \partial_\xi \right) \hat{\Phi}_\xi = -\frac{\omega''_0}{2} \hat{\Phi}_\xi^+ \hat{\Phi}_\xi - \xi^{(3)} \hat{\Phi}_\xi^+ \hat{\Phi}_\xi^+ \hat{\Phi}_\xi^+ \hat{\Phi}_\xi.
$$

In a co-propagating frame given by a spatial coordinate $\mu = \xi - \eta \tau$, Eq. (S5) is further simplified to give a quantum nonlinear Schrödinger equation (NLSE) [2]

$$
i \partial_\tau \hat{\Phi}_\mu = -\frac{\omega''_0}{2} \hat{\Phi}_\mu^+ \hat{\Phi}_\mu - \xi^{(3)} \hat{\Phi}_\mu^+ \hat{\Phi}_\mu^+ \hat{\Phi}_\mu^+ \hat{\Phi}_\mu.
$$

(S6)
Here, c-number substitutions of $\hat{\Phi}_\mu \rightarrow \Phi_\mu$ to Eq. (S6) leads to the classical NLSE [3].

Finally, to convert Eq. (S6) into a dimensionless form, we introduce normalized time and space coordinates $t$ and $z$ according to

$$
\tau = \frac{\omega_0' t}{(\xi (3))^2}, \quad \mu = \frac{\omega_0' z}{(\xi (3))^2},
$$

which produce operator dynamics

$$
\imath \partial_t \hat{\phi}_z = -\frac{1}{2} \partial^2_z \hat{\phi}_\mu + \hat{\phi}_\mu^{\dagger} \hat{\psi}_z, \quad \imath \partial_t \hat{\psi}_z = -\frac{\beta}{2} \partial^2_z \hat{\psi}_z + \frac{1}{2} \hat{\phi}_\mu^2 (S14)
$$

which is the same as Eq. (1) of the main text.

Using Eq. (S7), one can relate dimensionless coordinates used in the main text to experimental parameters. For instance, a normalized time of $t = 1$ corresponds to a lab-frame propagation distance of

$$
L_{\chi (3)} = \frac{|k_0'|}{\hbar \gamma \omega_0} = \frac{\lambda_0^3 |k_0'|}{\hbar \gamma c},
$$

which sets a characteristic single-photon nonlinear length. Here, $\lambda_0 = c / 2 \pi \omega_0$ is the carrier vacuum wavelength, and $k_0'$ is the group velocity dispersion with units of $[\text{time}^2 \cdot \text{length}^{-1}]$ and is related to $\omega_0'$ via $\omega_0' = -\nu^2 k_0'$. The absolute value in Eq. (S10) is taken to account for the potential negativity of $k_0'$. Similarly, a normalized width of $z = 1$ corresponds to a pulse duration in the lab frame of

$$
T_{\chi (3)} = \frac{|k_0'|}{\hbar \gamma \omega_0} = \frac{\lambda_0^3 |k_0'|}{\hbar \gamma c}.
$$

**B. $\chi^{(2)}$ nonlinear waveguide**

Similar procedures can be applied to $\chi^{(2)}$ nonlinear waveguides [1, 2]. Assuming that phase and group velocities are matched between the fundamental harmonic (FH) and the second harmonic (SH), the operator dynamics in the co-propagating lab frame become

$$
\imath \partial_t \Phi_\mu = -\frac{\omega_0''}{2} \partial^2_\mu \Phi_\mu + \zeta^{(2)} \Phi_\mu^{\dagger} \Psi_\mu, \quad \imath \partial_t \Psi_\mu = -\frac{\omega_0''}{2} \partial^2_\mu \Psi_\mu + \frac{\zeta^{(2)}}{2} \Phi_\mu^{\dagger} \Phi_\mu,
$$

corresponding to Eq. (S6) for the $\chi^{(3)}$ case. Here, $\Phi_\mu$ and $\Psi_\mu$ are FH and SH field annihilation operators, respectively, and they fulfill canonical commutation relationships $[\Phi_\mu, \Phi_\mu^{\dagger}] = [\Psi_\mu, \Psi_\mu^{\dagger}] = \delta(\mu - \mu')$. Second-order energy dispersion of FH and SH are given by $\omega_0''$ and $\omega_0''$, respectively. The nonlinear coupling parameter $\zeta^{(2)}$ is connected to the CW slope conversion efficiency $\eta$ of second-harmonic generation at $\omega_0$, which has the units of [power$^{-1} \cdot$ length$^{-2}$], via $\zeta^{(2)} = \sqrt{2 \hbar \omega_0 \eta \eta^3}$.

We introduce normalized time and space coordinates $t$ and $z$ via

$$
\tau = \left(\frac{\omega_0''}{(\zeta (2))^4}\right)^{1/3} t, \quad \mu = \left(\frac{\omega_0''}{(\zeta (2))^2}\right)^{2/3} z,
$$

which leads us to normalized operator dynamics

$$
\imath \partial_t \hat{\phi}_z = -\frac{1}{2} \partial^2_\mu \hat{\phi}_\mu + \hat{\phi}_\mu^{\dagger} \hat{\psi}_z, \quad \imath \partial_t \hat{\psi}_z = -\frac{\beta}{2} \partial^2_z \hat{\psi}_z + \frac{1}{2} \hat{\phi}_\mu^2
$$

(S14)
with \( \beta = \omega''_b / \omega''_a \), \( \hat{\phi}_z = (\omega''_a / \zeta(2))^{1/3} \hat{\Phi}_\mu \), and \( \hat{\psi}_z = (\omega''_a / \zeta(2))^{1/3} \hat{\Psi}_\mu \). The coupled-wave equations Eq. (S14) are generated by a normalized Hamiltonian

\[
\hat{H} = \int dz \left( -\frac{1}{2} \hat{\phi}_z^2 \hat{\phi}_z^* - \frac{\beta}{2} \hat{\psi}_z^2 \hat{\psi}_z^* + \frac{1}{2} \left( \hat{\phi}_z^2 \hat{\phi}_z^* + \hat{\psi}_z^2 \hat{\psi}_z^* \right) \right),
\]

which is Eq. (28) of the main text.

Based on Eq. (S13), the single-photon nonlinear length \( L_{\chi^{(2)}} \) and the characteristic pulse duration \( T_{\chi^{(2)}} \) (i.e., for \( t = z = 1 \)) for a \( \chi^{(2)} \) nonlinear waveguide are related to experimental parameters via

\[
L_{\chi^{(2)}} = \left( \frac{\lambda_a^2 |k_0'|}{4 \hbar^2 g^2 c^2} \right)^\frac{1}{2}, \quad T_{\chi^{(2)}} = \left( \frac{\lambda_a |k_0'|}{2 \hbar \eta c} \right)^\frac{1}{4},
\]

where \( \lambda_a \) is the FH carrier vacuum wavelength.

C. Experimental prospects

In considering experimental prospects for \( \chi^{(3)} \) nonlinear waveguides, \( \gamma = 660 \text{ W}^{-1}\text{m}^{-1} \) at \( \lambda = 1.59 \mu\text{m} \) [4] leads to \( L_{\chi^{(3)}} = 1.5 \times 10^5 \text{ m} \), assuming \( |k_0'| = 1 \text{ fs}^{-2} \text{ mm}^{-1} \). It is important to note that \( L_{\chi^{(3)}} \) is a continuous-wave single-photon nonlinear length; generally, due to the nonlinear nature of the interactions, the effective nonlinear rate can be further enhanced with higher peak intensity. For the case of a Kerr soliton with fixed waveguide parameters, for instance, the effective self-phase modulation rate scales proportionally to the total photon number \( \bar{n} \), thanks to the larger peak intensity of the soliton pulse [5]. Signatures of this pulsed enhancement can already be seen in Fig. 3(d) of the main text.

Non-Gaussian quantum features are expected to be pronounced when the effective nonlinear rate exceeds the rate of linear loss, which can be interpreted as a strongly interacting regime. By fully leveraging the pulse-enhanced nonlinearities, as shown in Ref. [6], it seems plausible that non-Gaussian Kerr effects could be observed if the ratio of nonlinearity to loss in nanophotonic Kerr media could be improved by another order of magnitude from the present state of the art in \( \chi^{(3)} \) nonlinear waveguides.

Regarding \( \chi^{(2)} \) nonlinear waveguides, thin-film periodically poled lithium niobate (PPLN) waveguides operated in the short wavelength regime \( \lambda_a = 780 \text{ nm} \) have been theoretically predicted to achieve \( \eta = 140,000 \% \text{W}^{-1}\text{cm}^{-1} \) [7]. These numbers lead to \( L_{\chi^{(2)}} = 2.7 \text{ cm} \) for \( |k_0'| = 1 \text{ fs}^{-2} \text{ mm}^{-1} \), making fabrication of nonlinear straight waveguides with single-photon-level nonlinearity plausible. Such potential for high \( \chi^{(2)} \) nonlinearity could also provide a means to realize large \( \chi^{(3)} \) nonlinearity via a cascaded \( \chi^{(2)} \) process [8]. Finally, characteristic (i.e., 3 dB) attenuation lengths as long as 1 m have been realized in thin-film PPLN waveguides [9]. Taking these numbers for nonlinearity and loss together (and considering further the possibility for pulsed enhancement of nonlinearity as discussed above), we expect observation of significantly non-Gaussian quantum dynamics, such as those described in this work, to be possible in the near future.

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