Extremely nonlocal optical nonlinearities in atoms trapped near a waveguide

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Nonlinear optical phenomena are typically local. Here we predict the possibility of highly nonlocal optical nonlinearities for light propagating in atomic media trapped near a nano-waveguide, where long-range interactions between the atoms can be tailored. When the atoms are in an electromagnetically-induced transparency configuration, the atomic interactions are translated to long-range interactions between photons and thus to highly nonlocal optical nonlinearities. We derive and analyze the governing nonlinear propagation equation, finding a roton-like excitation spectrum for light and the emergence of long-range order in its output intensity. These predictions open the door to studies of unexplored wave dynamics and many-body physics with highly-nonlocal interactions of optical fields in one dimension.

Optical nonlinearities are commonly described by local nonlinear response of the material to the optical field, resulting in the dependence of the refractive index at point \( z \) on the field at the same point, \( E(z) \) [1]. Recently however there has been a growing interest in nonlocal nonlinear optics, namely, in mechanisms whereby the refractive index at \( z \) depends on the field intensity at different points \( z' \) in the material [2]. Mechanisms that give rise to such nonlocal nonlinearities include e.g. heat diffusion [3], molecular reorientation in liquid crystals [4] and atomic diffusion [5, 6]. This paper discusses a new regime of extremely nonlocal and controllable optical nonlinearities in atomic media, which is achieved by designing the interactions between atoms prepared in an electromagnetically-induced transparency (EIT) configuration.

EIT is associated with the lossless and slow propagation of light pulses in resonant atomic medium subject to coherent driving of an auxiliary atomic transition [7]. Since the early days of EIT, it has been explored as a means of enhancing optical nonlinearities [7–10]. A particular effective mechanism for giant optical nonlinearities is provided by dipolar interactions between atoms that form the medium: Since EIT can be described by the propagation of the so-called dark-state polariton [11], which is a superposition of the light field and an atomic spin wave, the inherently nonlocal dipolar interactions between atoms are translated to nonlocal nonlinearities in polariton propagation. In the case of dipolar interactions between Rydberg atoms in free space, most theoretical [12–15] and experimental [16–20] studies have focused on their remarkable strength, a useful feature in quantum information, whereas their nonlocal aspect has received less attention [21].

The present work rests on two previously explored mechanisms: that of EIT polaritons and that of modified long-range dipolar interactions in confining geometries, such as fibers, waveguides, photonic band structures [22] or transmission lines, that may entail possible giant vacuum-induced forces [23] and long-range entanglement generation [24]. Yet, we show that the combined effect of these mechanisms may allow for unfamiliar highly nonlocal optical nonlinearity. Namely, purely dispersive laser-induced dipolar interactions between atoms coupled to a nano-waveguide with a grating, which can extend over hundreds of optical wavelengths [25, 26], are shown to be translated via EIT into highly nonlocal optical nonlinearities.

To this end, we derive a nonlinear propagation equation for the (possibly quantum) EIT polariton field \( \hat{\Psi} \) along the fiber axis \( z \), that corresponds to a general interatomic potential \( U(z−z') \) and includes lowest-order nonadiabatic corrections,

\[
(\partial_t + v \partial_z) \hat{\Psi}(z) = -i(\delta_c + \delta_{NL}) [\alpha - C v^2 \partial^2_z] \hat{\Psi}(z), \quad \delta_{NL} = \alpha \int_L d\zeta U(z − \zeta') \hat{\Psi}^\dagger(\zeta') \hat{\Psi}(\zeta').
\]

Here the frequency shift and quadratic dispersion coefficients \( \alpha \) and \( C \), and group velocity \( v \) (specified below) are all real. The effect of the nonlocal interaction appears in the nonlinear detuning operator, \( \delta_{NL} \), which adds up to the detuning of the coupling field \( \delta_c \) (Fig. 1a), giving rise to a nonlocal and nonlinear frequency shift and quadratic dispersion.

This paper has two linked objectives: first, to elucidate the conditions under which such nonlocal and nonlinear propagation regime may occur; and second, to reveal, in this regime, a roton-like excitation spectrum of optical waves, which we predict to arise under EIT in atomic media trapped near a fiber-Bragg grating. This roton-like narrow-band spectrum is a signature of the tendency of light in this regime to exhibit spatial self-order, namely,
the state existence of a dipolar interaction $U$ and their longitudinal component $z$. Interaction between the atoms in the $|d\rangle$-level [see (b)] induces its energy shift $\delta_{NL}$ which is effectively added to the detuning $\delta_c$. (b) Laser-induced dipolar interactions: the laser with Rabi frequency $\Omega_L$ and detuning $\delta_L$ operates on the $|d\rangle \rightarrow |s\rangle$ transition of all atoms, $|s\rangle$ being an additional level, thus inducing a dipolar potential $U(z)$ between pairs of atoms ($z$ apart) populating the state $|d\rangle$. (c) Setup: atoms in the EIT configuration (black dots), illuminated by the fields $\mathcal{E}$ and $\Omega$ (thin blue arrow), are trapped at a distance $r_a$ from a nano-waveguide (gray cylinder) along $z$, from $z = 0$ to $z = L$. A far-detuned laser $\Omega_L$ (thick orange arrow), tilted by an angle $\theta_L$ from the $z$ axis, induces long-range interactions between the atoms, mediated by the waveguide modes.

crystal-like correlations, which can be revealed by measuring the photon intensity at the fiber’s output. The predicted self-order of light constitutes a new, hitherto unexplored, optical ”phase”, analogous to the spatial structure of cold atomic media subject to laser-induced dipole-dipole interactions [25] [27] [31].

The system

Consider a medium of identical atoms in an EIT configuration as in Fig. 1: The atoms are trapped at a distance $r_a$ from a nano-waveguide along its longitudinal $z$ axis [32] [33] (Fig. 1c). A strong (external) coupling field with Rabi frequency $\Omega$ drives the $|d\rangle \rightarrow |e\rangle$ atomic transition with detuning $\delta_c$ and wavenumber $k_c$ (Fig. 1a), whereas a weak (possibly quantized) field with carrier frequency $\omega_0$, wavenumber $k_0$ and envelope $\mathcal{E}$ is resonantly coupled to the $|g\rangle \rightarrow |e\rangle$ transition ($\omega_0 = \omega_{eg}$). Under tight transverse trapping (around $r_a$) with respect to the transition wavelength, the atomic positions can be characterized by their longitudinal component $z$ [32] [33]. We assume the existence of a dipolar interaction $U(z)$ between atoms in the state $|d\rangle$. Then, the energy of level $|d\rangle$ of an atom at $z$ is shifted by $\delta_{NL} \sim n_a \int dz' U(z - z') P_d(z')$, $n_a$ being the atomic density (per unit length) and $P_d(z')$ the occupation of state $|d\rangle$ in an atom at $z'$.

We may now explain the physical reasoning that leads to Eq. (1). In Fig. 2 we plot the complex linear susceptibility $\chi$ of the EIT medium to the probe field as a function of its detuning $\Delta_p$, in the presence of a coupling field detuned by $\delta_c$. In our case is given by $\Delta_c = \delta_c + \delta_{NL}$ (Fig. 1a). When $\Delta_c = 0$ (Fig. 2a), the absorption coefficient $\Im \chi$ is symmetric with respect to $\Delta_p$ whereas the dispersion $\Re \chi$ is antisymmetric, so that no (real) quadratic dispersion exists for the probe envelope centered around $\Delta_p = 0$. By contrast, when $\Delta_c \neq 0$ is introduced (Fig. 2b), $\Re \chi$ is no longer antisymmetric and quadratic dispersion exists, which explains the term $\Delta_c C_L \partial^2 \chi$ in Eq. (1). However, this comes at the price of non-vanishing losses at $\Delta_p = 0$. For this reason we choose to work in the so-called Autler-Townes regime $\Omega \gg \gamma$, $\Delta_c$, where $\gamma$ is the width of the level $|e\rangle$. Then, for $\Delta_c$ smaller than the single-atom transparency window, $\Delta_c \ll \Omega^2/\gamma$, but still larger than $\gamma$, the absorption per atom can become negligible while dispersion is still significant, as illustrated in Fig. 2b. This explains the lossless propagation described by Eq. (1) with real parameters $\alpha, v, C$. As long as the absorption, associated with dissipation due to spontaneous emission at rate $\gamma$, is negligible, so are the noise effects of vacuum fluctuations; Eq. (1) then holds in operator form without additional Langevin quantum noise operators.

The formal derivation of Eq. (1) is sketched in the Appendix. Here we merely point out that the polariton field $\Psi$ is given by the superposition $\Psi(z) = \cos \theta \mathcal{E}(z) - \sqrt{n_a L} \sin \theta \sigma_{gd}(z)$, where $n_a$ is the number of atoms per unit length in the medium, $\sigma_{gd}(z)$ is the atomic $|g\rangle ↔ |d\rangle$ spin at $z$ and $\theta$ is the EIT mixing angle. The lossless propagation regime governed by Eq. (1) with $\alpha = \sin^2 \theta, v = \cos^2 \theta c, C = \sin^2 \theta (2 - 3 \sin^2 \theta)/\Omega^2$ is obtained by assuming all detunings to be smaller than the EIT transparency window $\delta_{tr} = \Omega^2/(\gamma \sqrt{OD})$, where $OD$ is the optical depth of the atomic medium.

![FIG. 1:](image1.png)

(a) EIT atomic configuration: the probe field $\mathcal{E}$ is resonantly coupled to the $|g\rangle \rightarrow |e\rangle$ transition whereas the coupling field $\Omega$ is coupled to the $|d\rangle \rightarrow |e\rangle$ transition with detuning $\delta_c$. Interaction between the atoms in the $|d\rangle$-level [see (b)] induces its energy shift $\delta_{NL}$ which is effectively added to the detuning $\delta_c$. (b) Laser-induced dipolar interactions: the laser with Rabi frequency $\Omega_L$ and detuning $\delta_L$ operates on the $|d\rangle \rightarrow |s\rangle$ transition of all atoms, $|s\rangle$ being an additional level, thus inducing a dipolar potential $U(z)$ between pairs of atoms ($z$ apart) populating the state $|d\rangle$. (c) Setup: atoms in the EIT configuration (black dots), illuminated by the fields $\mathcal{E}$ and $\Omega$ (thin blue arrow), are trapped at a distance $r_a$ from a nano-waveguide (gray cylinder) along $z$, from $z = 0$ to $z = L$. A far-detuned laser $\Omega_L$ (thick orange arrow), tilted by an angle $\theta_L$ from the $z$ axis, induces long-range interactions between the atoms, mediated by the waveguide modes.

![FIG. 2:](image2.png)

Linear susceptibility of the EIT atomic medium to the probe field as a function of its detuning $\Delta_p$. (a) For total detuning of the coupling field $\Delta_c = 0$, the absorption $\Im \chi$ (red dashed line) and dispersion $\Re \chi$ (blue solid line) are symmetric and antisymmetric, respectively, with respect to $\Delta_p$, so that no (real) quadratic dispersion exists for the probe envelope centered around $\Delta_p = 0$. (b) For $\Delta_c \neq 0$, $\Re \chi$ is not antisymmetric, so that quadratic dispersion exists, giving rise to the term $\Delta_c C \partial^2 \chi$ in Eq. (1), with $\Delta_c = \delta_c + \delta_{NL}$ (Fig. 1a).
Excitation spectrum of polariton waves

Let us assume a continuous wave (CW) polariton, and find the dispersion relation of wave excitations around this CW background, in analogy to the Bogoliubov spectrum of excitations in a Bose–Einstein condensate (BEC) \[39\]. The CW solution of Eq. (1) is \( \psi(t) = \psi_0 e^{-i(\delta_c + \nu_p U_0) t} \), with \( \psi_0 = |\psi_0|^2 \), \( \nu_p = \alpha^2 |\psi_0|^2 \) being the effective photon density per unit length and \( U_0 \) the \( k = 0 \) component of the spatial Fourier transform of the potential \( U_k = \int_{-\infty}^{\infty} dz U(z) e^{-ikz} \). Here we have neglected edge effects by assuming \( l < z < L - l \), \( l \) being the range of the potential \( U(z) \). The dispersion relation of small fluctuations \( \varphi(z,t) \) around the large average CW field \( \langle \Psi \rangle = \psi(t) \) are found as usual upon inserting \( \Psi = \psi(t) + \varphi(z,t) \) into Eq. (1) and linearizing it by keeping the fluctuations \( \varphi \) to linear order. Then, introducing the ansatz \[39\],

\[
\varphi(z,t) = e^{i(\delta_c + \nu_p U_0) t} \left[ u_k e^{ikz} e^{-i(\omega_k + kv) t} - v_k^* e^{-ikz} e^{i(\omega_k + kv) t} \right],
\]

into the linearized equation for \( \varphi \), \( u_k \) and \( v_k \) being c-number (Bogoliubov) coefficients, and using standard procedures \[39\], we find the modified Bogoliubov spectrum [see Supplementary Information (SI)]

\[
\omega_k = \sqrt{\omega_k^0 + 2\nu_p U_k}, \quad \omega_k^0 = (\nu_p U_0 / \alpha + \delta_c) C \nu^2 k^2.
\]

This means that a polariton wave distortion (about the CW solution) with a wavenumber \( k \) relative to the carrier wavenumber (inside the EIT medium), oscillates at a frequency (relative to the carrier frequency \( \omega_0 \))

\[
\omega(k) = \alpha \delta_c + \nu_p U_0 + kv + \omega_k.
\]

The dispersion relation \[39\] is composed of the detuning \( \delta_c \) due to the coupling field, the self-phase modulation of the CW component \( \nu_p U_0 \) (analogous to the chemical potential in a BEC \[39\]), the linear dispersion \( \omega_k \), and the modified Bogoliubov excitation spectrum \( \omega_k \). The spectrum \( \omega_k \) is determined by the interplay between the interaction Fourier transform \( U_k \) and the "kinetic-energy" quadratic dispersion \( \omega_k^0 \), which is affected by both the detuning \( \delta_c \), and by the \( k = 0 \) component of \( U_k \). This interplay is further discussed below for \( U(z) \) resulting from laser-induced interactions near a waveguide grating.

Generation of two-mode correlations

The parametric process described by the foregoing modified Bogoliubov theory also entails the dynamic generation of two-mode squeezing, i.e. pairs of entangled polaritons with wavenumbers \( \pm k \). The analysis is similar to that of propagation in fibers with local Kerr nonlinearity \[40 \[41\]. Upon inserting the expansion of small quantum fluctuations in the longitudinal wavenumber modes \( k, \delta(z) = \sum_k \hat{a}_k e^{ikz} / \sqrt{L} \), into the linearized equation for \( \varphi(z,t) \), we obtain coupled first-order differential equations (in time) for \( \hat{a}_k(t) \) and \( \hat{a}_k^{†}(t) \), whose solution is a dynamic Bogoliubov transformation (SI),

\[
\begin{align*}
\hat{a}_k(t) &= e^{-i(\nu_p U_0 + \delta_c + kv) t} \left[ \mu_k(t) \hat{a}_k(0) + e^{i2\phi_k} \nu_k(t) \hat{a}_k^{†}(0) \right], \\
\mu_k(t) &= \cos(\omega_k t) - i \frac{\nu_p U_k + \omega_k^0}{\omega_k} \sin(\omega_k t), \\
\nu_k(t) &= -i \frac{\nu_p U_k}{\omega_k} \sin(\omega_k t).
\end{align*}
\]

The number of entangled pairs generated after propagation time \( t \) wavenumbers \( \pm k \) can be quantified by the so-called squeezing spectrum, whose optimum is given by \( G_k = (|\mu_k| - |\nu_k|)^2 \) \[40\] \((G_k < 1 \) signifies entanglement), or by the normalized second-order (intensity) correlation function \( g^{(2)}(z, z', t) = \langle \hat{\Psi}^\dagger(z) \hat{\Psi}^\dagger(z') \hat{\Psi}(z') \hat{\Psi}(z) \rangle / \langle \hat{\Psi}(z) \hat{\Psi}(z') \rangle \rangle \langle \hat{\Psi}(z) \hat{\Psi}(z') \rangle \rangle \)

(all fields measured at the fiber’s output after propagation time \( t = L/v \)), where the averaging is performed with respect to the initial probability distribution, e.g. the quantum state, of the polariton (probe) field. For initial zero-mean fluctuations (around the CW solution) with average polariton occupation at mode \( k \), \( \langle \hat{a}_k(0) \hat{a}_k^{†}(0) \rangle = N_k \delta_{kk} \), and vanishing anomalous correlations \( \langle \hat{a}_k(0) \hat{a}_{-k}^{†}(0) \rangle = 0 \), we find (SI)

\[
g^{(2)}(z, z') \approx 1 + \frac{2\alpha^2}{\pi \nu_p} \int_0^\infty dk \left[ |\mu_k|^2 N_k + |\nu_k|^2 (N_k + 1) + (2N_k + 1)|\mu_k||\nu_k| \cos \phi_k \cos|k(z - z')| \right],
\]

where \( \phi_k = \text{arg}(\mu_k \nu_k) \) and we used \((1/L) \sum_k \rightarrow \int dk / (2\pi) \). As we shall see below, these correlations may reveal the ordering of a nonlocal system caused by pair-generation at preferred \( k \)-values.

Laser-induced interaction via waveguide grating

We now turn to the specific case of laser-induced interatomic dipolar interaction \[42\]. Consider another laser with Rabi frequency \( \Omega_L \) which is detuned by \( |\delta_L| \gg \Omega_L \) from the transition \( |d\rangle \rightarrow |s\rangle, |s\rangle \) being a fourth atomic level (Fig. 1b), and assume that this transition is distinct and separated from the transitions used for EIT (Fig. 1a) either spectrally or by polarization. We further take the laser to be oriented at an angle \( \theta_L \) with respect to the waveguide (Fig. 1c), and that the nano-waveguide incorporates a grating, i.e. periodic perturbation of the refractive index with period \( \Lambda = \pi / k_B \) (see e.g. Refs. \[35 \[43\]). Then, for \( \omega_L \) inside and close to the edge of the photonic bandgap associated with the grating (the
probes field’s carrier frequency $\omega_0$ being outside the gap), the laser-induced interaction potential between the atoms becomes \cite{25}

$$U(z) = -U_L \frac{1}{2} \cos(k_L^0 z) \cos(k_B z) e^{-|z|/l}, \quad (7)$$

where $l$ can extend over hundreds of wavelengths \cite{25}. Here $k_L^0 = k_L \cos \theta_L$ with $k_L$ the laser wavenumber, and $U_L$ depends on the laser parameters, atomic transition and effective area at $r_a$ (see Appendix). The resulting spatial Fourier transform $U_k$ then consists of four Lorentzian peaks of width $\sim 1/l$, centered around the spatial beating frequencies $\pm (k_L^0 - k_B)$ and $\pm (k_L^0 + k_B)$.

**Roton and Anti-Roton spectra**

Let us focus on the peak of $U_k$ around $k_R \equiv k_L^0 - k_B$ and its effect on the dispersion relation (spectra), Eq. \cite{3}. We first consider the case of anomalous dispersion, where the signs of $\omega_k^0$ and $U_k$ are opposite. In analogy to BEC, this describes the case of an attractive potential $U_k$ that competes with the “kinetic energy” $\omega_k^0$. Then, for $k$-values satisfying $|\omega_k^0| > 2n_p |U_k|$, $\omega_k$ is real and exhibits a dip around $k_R$, in contrast to the case of a local potential for which $U_k$ is independent of $k$ (standard Bogoliubov spectrum) and this feature is absent. This is seen in Fig. 3a, where both the analytical results of Eq. \cite{0} and numerical simulations of the nonlinear equation \cite{1} (Appendix), are plotted and shown to agree very well. The narrow-band “dip” of this $\omega_k$ spectrum is in analogy with the roton minimum in He II \cite{14}. It reflects the fact that wave distortions about the CW field with spatial frequencies around $k_B$ cost less energy and are hence favorable. This feature implies that the intensity of the polariton field in its ground state would tend to self-order with typical wavenumber $k_R$ \cite{14}.

Turning to the case of normal dispersion, where the signs of $\omega_k^0$ and $U_k$ are identical, $\omega_k$ exhibits an “anti-rotor” peak around $k_R$ (Fig. 3b). This means that distortions of spatial frequencies around $k_R$ are costly, so that the system prefers to avoid these spatial variations. This behavior again manifests the tendency of the system to order, since it indicates the spatial distortions that the system is unlikely to be found in, should it be in its ground state.

In order to measure the roton and anti-rotor spectra, we first recall the meaning of the dispersion relation $\omega(k)$ from Eq. \cite{1}: without an interaction, the frequency associated with a wave envelope at wavenumber $k$ traveling inside the EIT medium is given by $\omega^{(0)}(k) = \omega_0 + vk + \delta_C U^2 k^2 = \omega(k) - n_p U_0 - (\omega_k - \delta_C U^2 k^2)$, so that $\omega_k$ (together with $n_p U_0$) expresses a frequency, or phase velocity, shift due to the nonlinearity. Namely, the wavenumber $k$ describes a spatial eigenmode of propagation, both with and without interaction, with an eigen-frequency $\omega(k)$ and $\omega^{(0)}(k)$, respectively. Now, suppose we let a weak quasi-CW pulse of length $L_p < L$ and frequency $\omega_p = \omega^{(0)}(k)$ enter the medium (on top of the strong CW), when the laser and hence the interaction $U_k$ are turned off. Since upon entering the medium the field does not change its frequency $\omega_p$, we deduce from the dispersion relation in the absence of interaction, $\omega^{(0)}(k)$ that the field inside the medium exhibits a perturbation $\varphi(z)$ at spatial frequency $k$ on top of the strong CW. Subsequently, when the entire pulse is in the medium, we immediately (non-adiabatically) turn on the laser $\Omega_L$, and hence the interaction $U_k$, so that the temporal frequency of the perturbation $\varphi(z)$ at wavenumber $k$ becomes $\omega(k) = \omega^{(0)}(k) + n_p U_0 + (\omega_k - \delta_C U^2 k^2)$. The frequency shift $n_p U_0 + (\omega_k - \delta_C U^2 k^2)$ can be thought of as an extra energy acquired by the mode $k$ due to the interaction energy. Therefore, the spectrum $\omega_k$ can be inferred from the frequency shift, measurable by homodyne detection of the pulse $\varphi(z)$ that exits the EIT medium (Fig. 3c). A similar procedure was proposed

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3.pdf}
\caption{Dispersion relations for EIT polaritons with fiber-grating mediated atomic interactions (values of physical parameters used here are given in the Appendix). (a) Roton-like excitation spectrum (dispersion relation) $\omega_p$ for the potential of Eq. \cite{1} in the anomalous dispersion case (opposite signs of $\omega_k^0$ and $U_k$). The analytical results from Eq. \cite{3} (blue solid line) agree well with those of direct numerical simulations of Eq. \cite{1} (gray dots, see Appendix). Compared with the spectrum of a local interaction ($U_k$ independent of $k$, dashed red line), the roton-like spectrum exhibits a dip in a narrow band of $k$-values around $k_R = k_L^0 - k_B$. (b) Anti-roton peak of the spectrum $\omega(k)$ in a narrow band around $k_R$ in the normal dispersion case (identical signs of $\omega_k^0$ and $U_k$). (c) Possible homodyne detection scheme: the input probe field consists of a CW field + perturbation at wavenumber $k$ and frequency $\omega^{(0)}(k)$. The field is split before entering the EIT medium ($z = 0$), so that a local oscillator of $\omega^{(0)}(k)$ is formed (lower arm) by e.g. filtering out the CW component. Then, mixing the output signal ($z = L$) with the local oscillator reveals their phase difference, from which $\omega_k$ can be inferred (see text).}
\end{figure}
for measuring the tachyon-like spectrum of polaritons in inverted media 46.

Dynamical instability: pair generation

In the anomalous dispersion case, consider now a sufficiently strong interaction such that for a narrow band of $k$-values around $k_R$, where $U_k$ is peaked, the condition $2n_p|U_k| > |\omega_0^k|$ is satisfied and $\omega_k$ becomes imaginary. Then, field perturbations around $k_R$ become exponentially unstable (Fig. 4a), resulting in parametric amplification and generation of entangled photon pairs in this narrow band of unstable $k$-values. The strength of the amplified perturbation and generated field is characterized by the magnitude of the coefficients $\mu_k(t)$ and $\nu_k(t)$ from Eq. (5), which grow exponentially with propagation time $t$ and are largest for the narrow peak around $k_R$. The resulting squeezing spectrum $G_k$ at the output $t = L/v$ (Fig. 4a) may be measured by homodyne detection 10, 45.

Dynamical instability: emergence of self-order

Perhaps the most remarkable implication of the extremely nonlocal potential of Eq. (7) is the dynamic formation of order in the system. Consider that at $t = 0$ there exist fluctuations around the CW, with a spatial spectrum $N_k = N_0 e^{-|k/q_0|^2}$, i.e., “δ-correlated” noise limited by the EIT transparency window of width $\delta_v = v q_0$. Then, fluctuations at $k$-values around the peak $k_R$ will be parametrically amplified as they propagate through the medium, as verified in Fig. 4b, where the spatial spectrum of the polariton field, $N_k(t) = \langle \hat{a}^\dagger_k(t)\hat{a}_k(t) \rangle$, at different propagation times $t$ ($t = L/v$ describing the output field) is calculated both analytically and via direct (classical) numerical simulations of Eq. (1). This suggests that the system becomes spatially ordered with a extremely nonlocal potential of Eq. (7) is the broadband instability of the local-interaction case (red dotted line). The instability is accompanied by generation of quantum entanglement, characterized by a narrow-band squeezing spectrum $G_k$ (blue solid line; $G_k < 1$ quantifies entanglement between ±$k$ photon modes), in contrast to the broadband spectrum for a local interaction (horizontal red solid line). (b) Dynamics of spatial spectrum $N_k(t)$ of the field intensity for different propagation times $t$ inside the medium ($t = L/v$ at the output). The emergence of a large peak around $k_R$ (and $-k_R$) out of the initial Gaussian polarization is clearly seen. Results of both the linearized theory (solid lines) and numerical simulations of Eq. (1) (dots) are shown: slight differences at later $t$-values are attributed to nonlinear corrections. (c) Self-ordering of the field intensity: considering the narrow peaks of $N_k(t)$ around ±$k_R$, fluctuations at these $k$-values become dominant, resulting in ordered intensity correlations $g^{(2)}$ which grow with propagation time $t$ ($t = L/v$ at the output). Excellent agreement between the theory, Eq. (6) (solid lines) and numerical simulations of Eq. (1) (dots) is observed. The correlations oscillate with a period $2\pi/k_R \approx 6.1$ mm and a range of a few $l \approx 2.7$ mm (where $L = 2.68$ cm and $v = 4340$ ms$^{-1}$), determined by the long-range interaction $U(z)$. This is in contrast to the local-interaction case, where the correlations vanish ($g^{(2)} = 1$) after a short distance $\pi/q_0 \approx 1.75$ mm, which is determined by the bandwidth $q_0$ of the initial fluctuations [see inset: local (red thin line) and nonlocal (blue thick line) cases for the output field at $t = L/v$].

m in our example, see Appendix), the light intensity clearly becomes ordered due to the long-range interaction $U(z)$, as can also be seen by the comparison to the local interaction case (inset of Fig. 4c).
Imperfections and scattering

So far we have considered a purely coherent evolution of the polariton field. Here we address three main sources of scattering and loss of field excitations (see also SI). First, the interaction we consider involves the illumination of the atoms by an off-resonant laser, which is accompanied by an incoherent process of scattering of laser photons $\Omega_L$ from the $|d\rangle \rightarrow |s\rangle$ transition to non-guided modes at rate $R_{fs}$ [12]. This process limits the coherence time of the $\sigma_{sd}$ spin wave and hence that of the polariton to be below $\sim R_{fs}^{-1}$, which in the example of Figs. 3 and 4 is nevertheless much longer than the experiment time $L/v$ (see Appendix).

Secondly, material imperfections and defects in the waveguide grating structure may give rise to scattering of photons off the guided mode. This effect is analyzed for dipolar interactions in Ref. [20], in terms of the so-called cooperativity of the waveguide structure, $P$, whose square root $\sqrt{P}$ gives the ratio between coherent (dipolar interactions mediated by waveguide modes, $U$) and incoherent processes (scattering to non-guided modes via the imperfections). For a realistic waveguide material of length $\lambda_L$, $P \sim 10^4$ [20]. Since the imperfection-mediated incoherent processes accumulate with the system size, and considering the dipolar interaction range as the effective system length $l \sim 3000\lambda_L$ (Appendix), we estimate $P \sim 10^3\lambda_L/l \sim 3.3$, giving rise to an imperfection-mediated scattering rate of $R_{im} \sim |U_L|/\sqrt{3.3}$ per atom. The effect of $R_{im}$ on the spectrum $\omega_k$ and the observables mentioned above can in principle be made arbitrary small, however, by noting that $\omega_k$ depends on $n_p U_L$ whereas $R_{im}$ depends solely on $U_L$. Then, decreasing $|U_L|$ while keeping $n_p U_L$ constant (by increasing the CW power $n_p$) reduces $R_{im}$ but keeps $\omega_k$ unchanged. This reflects the fact that $R_{im}$ is a single-polariton loss mechanism and hence independent of $n_p$, whereas the effects discussed above are cooperative.

Finally, we consider losses and decoherence of spinwave excitations of the EIT medium due to the nonvanishing detuning of the coupling field, $\Delta_c = \delta_c + \delta_{NL}$, where here $\delta_{NL} = n_p U_0/\alpha$ is the mean-field value of $\delta_{NL}$ [Eq. (1)] that appears in the linearized equation for $\psi$.

The transmission through the EIT medium of length $L$ is given by $e^{-\langle 1/2 \rangle_k \chi'' v L}$, leading to an imaginary frequency $-i(1/2) k_0 \chi'' v$ to be added to $\omega_k$, $\chi'' = \text{Im} \chi(\Delta_p = 0, \Delta_c)$, being the imaginary part of the EIT susceptibility [7] evaluated for simplicity at the center of the probe pulse ($\Delta_p = 0$) and in the presence of the coupling field detuning $\Delta_c = \delta_c + \delta_{NL}$. In the SI we show that the effect of the EIT losses on $\omega_k$ can be significantly decreased by reducing both $n_p$ and $\delta_c$.

In principle, all loss terms discussed above should be accompanied by corresponding quantum noise terms, which are not considered here, restricting this discussion to the classical-field case.

Conclusions

This study predicts a new and hitherto unexplored regime of nonlinear optics; namely, that of highly nonlocal interactions between photons in one dimension (1d). These nonlocal optical nonlinearities arise for light propagation inside driven atomic media in the vicinity of a waveguide. We have derived the nonlinear equation that governs light propagation in this regime, and have analyzed it around its CW solution, finding a narrow roton-like dispersion relation (Figs. 3a,b) and squeezing (entanglement) spectrum (Fig. 4a), and the emergence of order in the field-intensity (Figs. 4b,c), all of which reflect the tendency of the system to self-organize, which in turn results from the long-range interactions between photons.

We stress that the length-scale associated with order, $k_R = k_L \cos \theta_L - k_B$ is not imposed solely by the external grating with period $\Lambda = \pi/k_B$, but rather depends also on the wavenumber of the laser $k_L$ and its orientation $\theta_L$, reflecting the origin of order in the self-interaction of the light field. This is in contrast to order in e.g. an optical lattice, where the atoms are situated at lattice sites determined by the potential imposed by an external laser.

This work opens the way to experimental and theoretical investigations of new nonlinear wave phenomena, especially by venturing beyond the linearized regime and exploring the role of the nonlinear dispersion term $\delta_{NL} C v^2 \partial^2 z$. Specific directions of further research may include: (1) Nonlocal nonlinear optics in 1d, concerning the study of solitons, where the lensing effect created by the nonlinear refractive index change is now highly nonlocal, following $U(z)$. (2) Thermalization in 1d. This important issue has been considered both experimentally [18] and theoretically [49] for isolated BEC in 1d. Our proposed scheme may give access to important modifications in the possible route to thermalization affected by the nonlocal character of the designed interactions $U(z)$ and the nonlinear dispersion (“mass”). (3) Effects of non-additivity of systems with long-range interactions [50] may be studied here for quantum/classical optical fields. (4) Nonlocal interactions in other confining geometries, e.g. cavities, may give rise to other unfamiliar nonlocal optical nonlinearities. (5) The intriguing possibility of measuring field amplitudes and not only intensities, via homodyne detection, provides a possible advantage over the study of quantum fields in matter waves (atoms in a BEC), where measurements using a local oscillator are less straightforward.
Derivation of Eq. (1)

Equation (1) is derived as follows. The field envelope \( \mathcal{E}(z) = \sum_k \hat{a}_k e^{ik_0 z}/\sqrt{L} \), with commutation relations \([\hat{a}_k, \hat{a}_k^\dagger] = \delta_{kk'}\) and hence \( [\hat{\mathcal{E}}(z), \hat{\mathcal{E}}^\dagger(z')] = \delta(z-z')\), is assumed to be spectrally narrow and is guided by a transverse mode of the fiber with effective area \( A \) at position \( r_a \) and polarization vector \( \mathbf{e}_0 \). The Hamiltonian in the interaction picture is

\[
H_{AF} = -\hbar n_a \int dz \left[ ig\hat{\mathcal{E}}(z) e^{ik_0 z} \hat{\sigma}_{eg}(z) + h.c. \right],
\]

\[
H_{AC} = -\hbar n_a \int dz \left[ \Omega e^{-i\delta t} e^{ik_0 z} \hat{\sigma}_{ed}(z) + h.c. \right],
\]

\[
H_{DD} = \frac{1}{2} n_a^2 \hbar \int dz \int dz' U(z-z') \hat{\sigma}_{dd}(z) \hat{\sigma}_{dd}(z'),
\]  

(8)

and \( H_F = \sum_k \hbar c \hat{\alpha}_k^\dagger \hat{a}_k \), where \( g = \sqrt{\omega_0/(2\kappa_0 A)} d \cdot \mathbf{e}_0 \), \( d \) being the dipole matrix element of the \( |g\rangle \rightarrow |e\rangle \) transition, and \( \sigma_{ij}(z) = |i\rangle \langle j| \) for an atom at \( z \), with \( i,j \) representing the states \{\( g,d,e \)\}. We write the Heisenberg equations for the operators \( \hat{\mathcal{E}}(z) \), \( \hat{\sigma}_{ee}(z) \) and \( \hat{\sigma}_{gd}(z) \) and keep only linear terms in \( \hat{\mathcal{E}}(z) \), including nonlinearity solely through the potential \( U(z-z') \). We then transform them into equations of motion for the dark and bright polaritons, \( \hat{\Psi} \) and \( \hat{\Phi} \), respectively,

\[
\begin{pmatrix}
\hat{\Psi}(z) \\
\hat{\Phi}(z)
\end{pmatrix} =
\begin{pmatrix}
\cos \theta & -\sqrt{N} \sin \theta \\
\sin \theta & \sqrt{N} \cos \theta 
\end{pmatrix}
\begin{pmatrix}
\hat{\mathcal{E}}(z) \\
\hat{\sigma}_{gd}(z)/\sqrt{L}
\end{pmatrix},
\]

(9)

where \( \tan^2 \theta = n_a g^2/\Omega^2 \) and \( \sigma_{gd}(z) = \hat{\sigma}_{gd}(z) e^{i(k_0 - k_a)z} e^{-i\delta t} \). Next, we take the first nonadiabatic correction to EIT [11] by inserting the equation of motion for \( \hat{\Phi} \) into that of \( \hat{\Psi} \) and assume all detunings to be smaller than the EIT transparency window \( \delta_t = \Omega^2/(\gamma \sqrt{OD}) \), with \( OD = (n_a/A) L \sigma_a \) and \( \sigma_a \) the cross section of the \( |g\rangle \rightarrow |e\rangle \) transition, finally obtaining Eq. (1).

Quantitative illustration

Let us specify the system parameters used in the illustration in Figs. 3 and 4. Motivated by the experiment in Ref. [22], we take the wavelength, dipolar matrix element and radiative decay rate of the D1 line of Cesium atoms [47] as the typical parameters of the dipolar atomic transitions in Fig. 1, where the atoms are trapped at a distance \( r_a \sim 500 \) nm from the center of a tapered fiber with radius \( a = 250 \) nm and refractive index \( n = 1.452 \), leading to \( A = 4.42 \mu m^2 \) [25]. Taking an atom density of \( n_a \sim 16 \times 10^6 \) m\(^{-1}\), length \( L = 2.68 \) cm, and coupling laser with Rabi frequency \( \Omega = 4 \times 10^8 \) s\(^{-1}\) \((2 \times 10^8 \) s\(^{-1}\) and \(1.2 \times 10^8 \) s\(^{-1}\) in Figs. 3b and 4, respectively) and detuning \( \delta_c = -3.84 \times 10^7 \) s\(^{-1}\) in Fig. 3a \((4.795 \times 10^6 \) s\(^{-1}\) and \(-3.64 \times 10^6 \) s\(^{-1}\) and in Figs. 3b and 4), we obtain \( \alpha = 0.999839 \) (0.99996 and 0.999986 in Figs. 3b and 4), \( v = 48216 \) ms\(^{-1}\) (12055 ms\(^{-1}\) in 4340 ms\(^{-1}\) in Figs. 3b and 4, respectively) and \( \delta_{tr} = 8.66 \times 10^7 \) s\(^{-1}\) \((2.16 \times 10^7 \) s\(^{-1}\) and 7.79 \times 10^6 \) s\(^{-1}\) in Figs. 3b and 4, so that Figs. 3a,b and 4a,b are effectively cut at \( k = q_{tr} = \delta_{tr}/v = 1795 \) m\(^{-1}\). For the Bragg grating imprinted on the fiber/waveguide [35, 43], we assume periodic perturbations \( \Delta n = 0.02 \) of the refractive index about \( n \) with period of length \( \Lambda = 396 \) nm. For the laser induced interaction we take a detuning \( \delta_L = -2\pi \times 0.65 \) GHz and intensity \( I = 2 \times 10^4 \) Wm\(^{-2}\) \((0.5 \times 10^4 \) Wm\(^{-2}\) and 10^2 Wm\(^{-2}\) in Figs. 3b and 4), yielding \( U_L = 2\eta R_{fs} = 1.14 \times 10^6 \) s\(^{-1}\) \((2.85 \times 10^5 \) s\(^{-1}\) and 5696 s\(^{-1}\) in Figs. 3b and 4), where \( \eta \approx 12 \) is the ratio between emission to the fiber grating modes and to free-space, and \( R_{fs} = \gamma (\Omega_L^2/2\delta_L^2) = 47443 \) s\(^{-1}\) \((11860 \) s\(^{-1}\) and 237 s\(^{-1}\) in Figs. 3b and 4) is the resulting scattering rate to free space form the \(|d\rangle \rightarrow |s\rangle \) transition, leading to a potential range of \( l \approx 3027 \lambda_L \approx 0.1L \approx 0.0027 m \). The orientation angle is taken to be \( \theta_L = 0.131 \), so that \( k_{RL} = k_L \cos \theta_L - k_B = 1019 \) m\(^{-1}\). We assume a CW background with a power \( 2 \times 10^{-12} \) W in Fig. 3a \((10^{-10} \) W and \( 4 \times 10^{-10} \) W in Figs. 3b and 4), giving \( n_p \approx 187 m^{-1} \) \((37347 \) m\(^{-1}\) and 414981 m\(^{-1}\) in Figs. 3b and 4). For the \( N_\kappa(t) \) and \( g(2) \) calculations in Figs. 4b,c, we consider an initial Gaussian spectrum of intensity fluctuations, \( N_\kappa = N_{0g}(\kappa)/\sigma_\kappa^2 \), with \( N_0 = 5 \). Finally, wherever a comparison with the local-interaction case is made, the local potential is taken as \( U(z) = (1/8)U_L b(z) \).

Numerical simulations of Eq. (1)

In order to obtain the \( \omega_k \) spectrum, we perform numerical simulations of the full nonlinear equation (1) using a three-term splitting method, with an initial field comprised of the CW solution \( \psi \) and weak perturbations \( \varphi \) at a spatial frequency \( k \). From the dynamics of the field, we can extract the temporal oscillation frequency of the field \( \omega(k) \) leading to the spectrum \( \omega_k \). The results of the simulation of Eq. (1) agree very well with those of a simpler split-step method simulations, where the nonlinear dispersion term \( \delta_{NL} C u^2 \partial_z^2 \) is approximated by its mean-field value, \( \delta_{NL} = n_p U_0/\alpha \). For the case of instability (Fig. 4b,c), we begin with the initial intensity fluctuations with a spectrum \( N_\kappa \) and run simulations with the mean-field nonlinear dispersion term \( \delta_{NL} = n_p U_0/\alpha \). Then, after propagation time \( t \), we measure \( N_\kappa(t) = |a_1^\dagger(t) a_1(t)| \) and \( g(2) = \langle I(z) I(z') \rangle / \langle I(z) \rangle \langle I(z') \rangle \), with \( I(z) = |\Psi(z)|^2 \) for a classical field \( \Psi \).

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SUPPLEMENTARY INFORMATION

Bogoliubov theory for a nonlocal interaction

Here we wish to present the derivation of the polariton wave excitation spectrum (dispersion relation) of perturbations around the CW solution, Eq. (3) in the main text. We first write the polariton field as a sum of an average CW solution and a small fluctuation \( \hat{\varphi}(z,t) \),

\[
\hat{\Psi}(z,t) = \psi(t) + \hat{\varphi}(z,t),
\]

(S1)

where the average CW solution is that of self-phase modulation, \( \psi(t) = \psi_0 e^{-i(\alpha \delta_e + n_p U_0)t} \) with \( \psi_0 = \sqrt{n_p/\alpha^2} e^{i\phi} \), \( U_0 = U_{k=0} \), \( U_k \) being the spatial Fourier transform of the potential \( U(z) \),

\[
U_k = \int_{-\infty}^{\infty} dz U(z) e^{-ikz}.
\]

(S2)

The integral for \( U_0 \) that arises in the CW solution runs over \( z' = 0 \) to \( z' = L \) around some point \( z, \int_0^L dz' U(z - z') \), and not from \( z' = -\infty \) to \( z' = \infty \) around \( z = 0 \) as in the definition \( U_k \). However, for a symmetric potential \( U(z) = U(-z) \) with range \( l \) and points \( z \) well within the medium, i.e. \( l < z < L - l \), the edges \( z = 0 \) and \( z = L \) have no effect, so that \( U_0 \) can be approximated as \( \int_0^L dz' U(z - z') \approx \int_{-\infty}^{\infty} dz' U(-z') \) for all such points \( z \). Thus, we may neglect the edge effects of a sufficiently long medium \( L \gg l \).

Inserting Eq. (S1) into the nonlinear equation (1) in the main text and keeping terms only to linear order in the perturbation \( \hat{\varphi} \), we obtain

\[
(\partial_t + v \partial_z) \hat{\varphi}(z) = -i(\alpha \delta_e + n_p U_0)\hat{\varphi}(z) + i(n_p U_0/\alpha + \delta_e)Cv^2\partial_z^2 \hat{\varphi}(z) - in_p \int_0^L dz' U(z - z') \hat{\varphi}(z') - in_p e^{2|\phi-(\alpha \delta_e + n_p U_0)t|} \int_0^L dz' U(z - z') \hat{\varphi}(z').
\]

(S3)

This linearized equation is valid as long as the fluctuations \( \hat{\varphi} \) around the mean \( \psi \) are small, i.e. for

\[
|\psi| \gg |\langle \hat{\varphi} \rangle|, \sqrt{|\langle \hat{\varphi}^\dagger \rangle|}, \sqrt{|\langle \hat{\varphi}^2 \rangle|}.
\]

(S4)

In order to find the dispersion relation of the fluctuations it is enough to consider the case of a classical field \( \hat{\varphi} \rightarrow \varphi \). We then insert the ansatz

\[
\varphi(z,t) = e^{i(\phi-(\alpha \delta_e + n_p U_0)t)} \left[ u_k e^{ikz} e^{-i(\omega_k + kv)t} - v_k^* e^{-ikz} e^{i(\omega_k + kv)t} \right],
\]

(S5)
\[ \omega_k^0 + n_p U_k - \omega_k = 0, \quad -n_p U_k = \omega_k^0 + n_p U_k + \omega_k \] (S6)

In order to arrive at the equations (S6), we used quantum field \( \hat{\Phi} \) to arrive at the dynamical Bogoliubov transformation with the coefficients of Eq. (5) in the main text. We expand the extra equation whose solution is the spectrum \( \omega_k \), which again amounts to neglecting edge effects, and where in the last equality we assumed that \( u \) is a c-number coefficient, into the linearized equation (S3), obtaining coupled equations for \( k = \pm 1 \) being c-number coefficients, into the linearized equation (S3), obtaining coupled equations for \( k = \pm 1 \):

\[
\int_0^L dz U(x - x') e^{ikx'} \approx \int_{-\infty}^{\infty} U(x) e^{ikx} = U_0 e^{ikx},
\]

which again amounts to neglecting edge effects, and where in the last equality we assumed that \( U(x) \) is symmetric and real. Eq. (S6) has a nontrivial solution only if the determinant of the matrix on the left-hand side is zero, yielding an extra equation whose solution is the spectrum \( \omega_k \) from Eq. (3).

**Dynamic Bogoliubov theory for a nonlocal interaction**

In this part we address the quantum description of the dynamics of the fluctuations \( \hat{\Phi} \) around the CW solution and arrive at the dynamical Bogoliubov transformation with the coefficients of Eq. (5) in the main text. We expand the quantum field \( \hat{\phi}(x, t) \) in spatial Fourier modes \( \hat{a}_k(t) \), \( \hat{\phi}(x) = \sum_k (1/\sqrt{L}) e^{ikx} \hat{a}_k \), and the transformed modes \( \hat{c}_k(t) \) as,

\[
\hat{\phi}(x, t) = \sum_k \frac{1}{\sqrt{L}} e^{ikx} e^{i\left[(\omega_k + n_p U_k) - i(\alpha \delta_k U_0 + k v)\right]t} \hat{c}_k(t), \quad \hat{a}_k(t) = e^{i\phi_k} e^{-i(\alpha \delta_k + n_p U_0 + kv)t} \hat{c}_k(t),
\]

where the operators \( \hat{a}_k(t) \) satisfy the equal-time commutation relations \( [\hat{a}_k(t), \hat{a}^\dagger_{k'}(t)] = \delta_{kk'} \) and hence so do the operators \( \hat{c}_k(t) \). Inserting Eq. (S8) into the linearized equation (S3) and neglecting edge effects as in Eq. (S7), we obtain an equation for \( \partial_t \hat{c}_k \). Then, upon taking its Hermitian conjugate, we end up with coupled equations of motion for \( \hat{c}_k \) and \( \hat{c}^\dagger_{-k} \):

\[
\partial_t \left( \begin{array}{c} \hat{c}_k \\ \hat{c}^\dagger_{-k} \end{array} \right) = -i \left( \begin{array}{cc} \omega_k^0 + n_p U_k & n_p U_k \\ -n_p U_k & -\omega_k^0 - n_p U_k \end{array} \right) \left( \begin{array}{c} \hat{c}_k \\ \hat{c}^\dagger_{-k} \end{array} \right).
\]

By diagonalizing the matrix, we find the solution for \( \hat{c}_k(t) \) from which we obtain the dynamics of \( \hat{a}_k(t) \) as the Bogoliubov transformation from Eq. (5) in the main text.

**Intensity correlations**

Intensity correlations are characterized by the normalized second order coherence function defined by

\[
g^{(2)}(z, z') = \frac{G^{(2)}(z, z')}{G^{(1)}(z)G^{(1)}(z')}
\]

with

\[
G^{(2)}(z, z') = \langle \hat{\Psi}(z, t)\hat{\Psi}(z', t)\hat{\Psi}(z', t)\hat{\Psi}(z, t) \rangle, \quad G^{(1)}(z) = \langle \hat{\Psi}(z, t)\hat{\Psi}(z, t) \rangle.
\]

Here we assume that all points of the field \( \hat{\Psi}(z) \) have experienced the same duration of interaction \( t \) upon arrival at the detector. The detector measures at different times different points of the field \( \hat{\Psi}(z) \), so that the correlations between detector-signals at different times are in fact correlations of the field in space as in \( g^{(2)}(z, z') \) which quantifies the autocorrelation of the field between \( z \) and \( z' \).

Upon expanding the field in its longitudinal Fourier modes, we recall that the \( k = 0 \) mode can be approximated by the strong average CW solution,

\[
\hat{\Psi}(z, t) = \frac{1}{\sqrt{L}} \sum_k e^{ikz} \hat{a}_k(t) \approx \frac{a_0(t)}{\sqrt{L}} + \frac{1}{\sqrt{L}} \sum_{k \neq 0} e^{ikz} \hat{a}_k(t), \quad \frac{a_0(t)}{\sqrt{L}} = \sqrt{\frac{n_p}{\alpha^2}} e^{i\phi} e^{-i(\alpha \delta_k + n_p U_0)t},
\]

\[ u_k \text{ and } v_k \text{ being c-number coefficients, into the linearized equation (S3), obtaining coupled equations for } u_k \text{ and } v_k \]

\[ (\omega_k^0 + n_p U_k - \omega_k, -n_p U_k) \begin{pmatrix} u_k \\ v_k \end{pmatrix} = 0. \]
where \( \hat{a}_k(t) \) is given by Eq. (5) of the main text. Inserting Eq. (S12) into \( G^{(1)}(z) \) from Eq. (S11), we find

\[
G^{(1)}(z) = \frac{n_p}{\alpha^2} + \frac{1}{L} \sum_{k \neq 0} \left[ |\mu_k|^2 N_k + |\nu_k|^2(N_k + 1) \right],
\]

(S13)

where we have assumed the following statistics of the initial fluctuations \( \hat{a}_k(0) \): \( \langle \hat{a}_k(0) \rangle = 0, \langle \hat{a}_k(0)\hat{a}_k(0) \rangle = N_k \delta_{kk'} \) with \( N_k = N_{-k} \), and \( \langle \hat{a}_k(0)\hat{a}_{-k}(0) \rangle = 0 \). Moving to the second-order coherence, we insert Eq. (S12) into \( G^{(2)} \) from Eq. (S11), keeping terms only to second order in the fluctuations \( \hat{a}_k \), in accordance with the assumptions (S4), and obtain

\[
G^{(2)}(z, z') = \frac{n_p^2}{\alpha^2} + \frac{2n_p}{\alpha^2} \frac{1}{L} \sum_{k \neq 0} \left\{ \left[ |\mu_k|^2 N_k + |\nu_k|^2(N_k + 1) \right] \left( 1 + \cos[k(z - z')] \right) + |\nu_k||\mu_k|(1 + 2N_k) \cos[k(z - z') - \phi_k] \right\},
\]

(S14)

where \( \phi_k = \arg(\mu_k\nu_k) \). Inserting \( G^{(1)} \) and \( G^{(2)} \) from Eqs. (S13) and (S14) in Eq. (S15), we note that to lowest order in the fluctuations we can take \( G^{(1)} \approx n_p/\alpha^2 \) in the denominator, arriving at

\[
g^{(2)}(z, z') = 1 + \frac{2\alpha^2}{n_p} \frac{1}{L} \sum_{k \neq 0} \left\{ \left[ |\mu_k|^2 N_k + |\nu_k|^2(N_k + 1) \right] \left( 1 + \cos[k(z - z')] \right) + |\nu_k||\mu_k|(1 + 2N_k) \cos[k(z - z') - \phi_k] \right\}.
\]

(S15)

Finally, upon taking the continuum limit \((1/L) \sum_k \rightarrow (1/2\pi) \int_{-\infty}^{\infty} dk\), we split the integral into its positive and negative \( k \)-values and obtain Eq. (6) from the main text.

**Imperfections and scattering**

The scattering and loss mechanisms discussed in the main text represent single-atom (single-polariton) effects which may add a loss term of the type \( -\gamma \text{loss} \widetilde{\Psi}(z) \) on the right-hand side of the propagation equation, Eq. (1), and hence an imaginary part to the spectrum \( \omega_k \). In Fig. 1a, we plot \( \omega_k \) for the roton and anti-roton cases (blue solid line), compared with \( -\gamma \text{loss} \) for the three types of losses mentioned in the main text: 1. Scattering of off-resonant laser photons to non-guided modes, \( \gamma \text{loss} = R_{fs} \) (gray dashed line); 2. Imperfection-mediated scattering, \( \gamma \text{loss} = R_{im} \sim |U_L|/\sqrt{3.3} \) (orange dashed line); 3. EIT losses due to detuning of the coupling field, \( \gamma \text{loss} = R_{EIT} = -i(1/2)\kappa_0 \text{Im}\chi(\Delta_p, \Delta_c) \) (black dashed line). Considering the susceptibility of an EIT medium [2], we find

\[
R_{EIT} = v OD \frac{2(\Delta_p - \Delta_c)^2\gamma^2}{L \left[ 4\gamma^2(\Delta_p - \Delta_c)^2 + (\Omega^2 - 4\Delta_p(\Delta_p - \Delta_c))^2 \right]},
\]

(S16)

\( \gamma \) being the width of the atomic level \( |c\rangle \) and \( OD \) the optical depth of the medium (see main text). In all figures here, \( R_{EIT} \) is evaluated at the center of the pulse \( (\Delta_p = 0) \) and for \( \Delta_c = \delta_c + \delta_{NL} \), \( \delta_{NL} = n_p U_0/\alpha \) being the mean-field value of the nonlinear detuning.

In the roton case in Fig. S1a, using the same physical parameters from the main text (Fig. 3a and Appendix therein) we observe that the off-resonant scattering \( R_{fs} \) (gray dashed line) is negligible whereas \( R_{im} \) and \( R_{EIT} \) can become

![Graph showing the comparison between the lossless (real) spectrum \( \omega_k \) (blue solid line for nonlocal interaction and red dashed line for local interaction), and loss contribution (imaginary). Three loss mechanisms are discussed: off-resonant scattering (gray dashed line), imperfection-mediated scattering (orange dashed line), and EIT losses (black dashed line).](image-url)
FIG. S2: Effect of losses on the instability spectrum $\gamma_k$ (a) Lossless case (blue solid line for nonlocal interaction, red dashed line for local interaction) compared with that where imperfection-mediated scattering are included (black dashed line). (b) Same as (a) for EIT losses (black dashed line). (c) Same as (b), where $n_p$ and $\delta_c$ are reduced by a factor $f = 10$.

c omparable to the $\omega_k$ spectrum and may hence significantly alter the observable effects discussed in the main text. However, as explained in the main text, the effect of $R_{im}$ can in principle be made arbitrary small by decreasing $|U_L|$ and keeping $U_L n_p$ constant, leaving us with $R_{EIT}$ (black dashed line) as a dominant loss mechanism. Reexamining the expression for $\omega_k$, Eq. (3) in the main text, we observe that a reduction of both $\delta_c$ and $n_p$ by a factor $f$, results in a reduction of $\omega_k$ by the same factor. This linear scaling is in contrast with the nonlinear dependence of $R_{EIT}$ with $\Delta_c$ as per Eq. (S16). This scaling difference is an example of a possible method for obtaining coherent effects (represented by $\omega_k$) which are dominant over incoherent EIT losses, as shown in Fig. S1b, where $f = 10$ is taken.

Proceeding to the anti-roton case in Fig. S1c, we plot $-\gamma_{loss}$ for the three types of incoherent processes using the physical parameters from the main text (Fig. 3b and Appendix therein). It is seen that both the EIT losses (black dashed line) and those of off-resonant scattering $R_{fs}$ (gray dashed line) are negligible, leaving us with the imperfection-induced scattering, which in turn can be drastically reduced as explained above.

Finally, considering the case of an instability, where in the lossless case we have $\Im \omega_k = \gamma_k > 0$, losses give negative contribution to $\gamma_k$, hence degrading the parametric amplification associated with the instability. Here $R_{fs}$ still gives a negligible contribution to $\gamma_k$, as well as the effect of imperfections: in Fig. S2a we plot the lossless $\gamma_k$ (blue solid line) compared with $\gamma_k - R_{im}$ (black dashed line) obtaining very similar curves. Considering the EIT losses, in Fig. S2b we plot $\gamma_k$ (blue solid line) compared with $\gamma_k - R_{EIT}$ and observe a significant reduction of the amplification rate. Nevertheless, reducing both $\delta_c$ and $n_p$ by e.g. a factor $f = 10$ as in the roton case, may, in principle result in EIT losses which are negligible with respect to $\gamma_k$, as presented in Fig. S2c.

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