Highly nonlocal optical nonlinearities in atoms trapped near a waveguide: supplementary material

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1. EIT WITH NONLOCAL INTERACTIONS

Here we elaborate on the derivation of Eq. (1). Sec. A and B below review rather standard results, whereas the adiabatic expansion leading to nonlocal and nonlinear dispersion is discussed in Sec. C and D.

A. Effective 1d description

The system from Fig. 1 is modeled by the Hamiltonian

$$\begin{aligned} H_A &= \int d\mathbf{r} \rho(\mathbf{r}) [\hbar \omega_{eg} \hat{\sigma}_{ee}(\mathbf{r}) + \hbar(\omega_{eg} - \omega_{ed}) \hat{\sigma}_{dd}(\mathbf{r})], \\ H_F &= \sum_k \hbar(\omega_0 + ck) \hat{a}_k^\dagger \hat{a}_k, \\ H_{AF} &= -\hbar \int d\mathbf{r} \rho(\mathbf{r}) \sum_k [i g_k(\mathbf{r}) \hat{a}_k e^{ik_0 z} \hat{\sigma}_{eg}(\mathbf{r}) + \text{h.c.}], \\ H_{AC} &= -\hbar \int d\mathbf{r} \rho(\mathbf{r}) [i \Omega e^{-i\omega_c t} e^{ik_c z} \hat{\sigma}_{ed}(z) + \text{h.c.}], \\ H_{DD} &= \frac{1}{2} \int d\mathbf{r} \rho(\mathbf{r}) \int d\mathbf{r}' \rho(\mathbf{r}') U(\mathbf{r} - \mathbf{r}') \hat{\sigma}_{dd}(\mathbf{r}) \hat{\sigma}_{dd}(\mathbf{r}'), \end{aligned} \quad (\text{S1})$$

where $\hat{\sigma}_{ij}(\mathbf{r}) = |i\rangle\langle j|$ are the continuum (coarse-grained) atomic operators around $\mathbf{r}$ [1], and $\rho(\mathbf{r})$ is the atomic density. Here $\{k\}$ denote longitudinal modes guided by the waveguide with mode function $\mathbf{f}_k(x, y) e^{ikz} / \sqrt{L}$ and frequency ω_k , $\mathbf{f}_k$ being the profile of the fundamental transverse mode of the waveguide at wavenumber k , so that $g_k(\mathbf{r}) = \sqrt{\omega_k / (2\epsilon_0 \hbar)} \mathbf{d} \cdot \mathbf{f}_k e^{ikz} / \sqrt{L}$. Assuming that the relevant k wavenumbers form a relatively narrow band around $k_0 = \omega_0 / c$, we take a linear dispersion relation $\omega_k = \omega_0 + ck$ and ignore the dependence of the transverse profile on k , $\mathbf{f}_k(x, y) = \mathbf{e}_0 f_0(x, y)$.

The atoms are trapped along the waveguide between $z = 0$ and $z = L$ with a constant density (per unit length) along z , n_a , and a transverse profile $p(x, y)$ (per unit area) of width w around some point $r_a = (x_a, y_a)$. Considering the trapping width w to be much smaller than the spatial variation of $f_0(x, y)$ (e.g. $\sim 60\text{nm}$ and 250nm , respectively, in [2]), we can take $\rho(\mathbf{r}) = n_a p(x, y) \approx n_a \delta(x - x_a) \delta(y - y_a)$. Then, identifying the effective area $f_0(x_a, y_a) = 1/\sqrt{A}$ and the collective atomic operators $\hat{\sigma}_{ij}(z) = \int dx \int dy p(x, y) \hat{\sigma}_{ij}(\mathbf{r})$, and moving also to an interaction picture with respect to $H_A + \sum_k \hbar \omega_0 \hat{a}_k^\dagger \hat{a}_k$, we obtain the effectively 1d Hamiltonian, Eq. (2) in the main text.

B. Weak probe approximation

Using the Hamiltonian (2), we derive the following Heisenberg equations of motion,

$$\begin{aligned} (\partial_t + c\partial_z) \hat{\mathcal{E}}(z) &= n_a g^* \tilde{\sigma}_{ge}(z), \\ \partial_t \tilde{\sigma}_{ge}(z) &= -\Omega^{-i\delta_c t} \tilde{\sigma}_{gd}(z) + g \hat{\mathcal{E}}(z) [\hat{\sigma}_{ee}(z) - \hat{\sigma}_{gg}(z)] \\ &\quad - \gamma \tilde{\sigma}_{ge} + \hat{F}, \\ \partial_t \tilde{\sigma}_{gd}(z) &= \Omega^* e^{i\delta_c t} \tilde{\sigma}_{ge}(z) + g \hat{\mathcal{E}}(z) \hat{\sigma}_{ed}(z) e^{ik_c z} \\ &\quad - i n_a \int dz' U(z - z') \hat{\sigma}_{dd}(z') \tilde{\sigma}_{gd}(z), \end{aligned} \quad (\text{S2})$$

with $\tilde{\sigma}_{ge} = \hat{\sigma}_{ge} e^{-ik_0 z}$ and $\tilde{\sigma}_{gd} = \hat{\sigma}_{ge} e^{i(k_c - k_0)z}$. The second line of the equation for $\partial_t \tilde{\sigma}_{ge}$ describes the effect of the reservoir formed by all photonic modes not included in the Hamiltonian (non-guided modes), namely, spontaneous emission from the level $|e\rangle$ and its corresponding Langevin noise operator. We now assume that all atoms are initially in the state $|g\rangle$ and that the probe field $\hat{\mathcal{E}}$ is weak enough so that the photon density in

the medium is much smaller than the atom density hence the *individual* atoms respond linearly to the probe (i.e. the transition $|g\rangle \rightarrow |e\rangle$ is far from saturation): $\hat{\sigma}_{ij} = O(\hat{\mathcal{E}})$ for $i \neq j$, and $\hat{\sigma}_{gg} \sim 1$. Applying such linearization to Eqs. (S2) while retaining the nonlinearity due to the interaction U between *different* atoms, we can eliminate $\hat{\sigma}_{ge}$ and obtain the coupled equations for $\hat{\sigma}_{gd}$ and $\hat{\mathcal{E}}$, Eqs. (3) from the main text.

C. Adiabatic expansion

The equations of motion for the polaritons, Eqs. (6) in the main text, were obtained directly from the ones for the atoms and weak probe field, Eq. (3) without further approximations. In order to arrive at our central equation, Eq. (1), we then perform an adiabatic expansion in the spirit of Ref. [1]. The main idea is to consider an overall EIT configuration which is very close to both single-photon and two-photon resonances, namely, a slowly varying probe envelope $\hat{\mathcal{E}}(z)$ and small detunings for the coupling field. In turn, this implies small temporal derivatives of both polariton fields on the one hand, and small linear and nonlinear detunings on the other hand, respectively. Namely, our small parameter in the expansion, denoted T^{-1} , scales as

$$T^{-1} \gtrsim \partial_t \hat{\Psi}, \partial_t \hat{\Phi}, \delta_c, \hat{S}. \quad (\text{S3})$$

For example, in the adiabatic limit $T^{-1} \rightarrow 0$ we take the lowest order in Eqs. (5), obtaining

$$(\partial_t + c \cos^2 \theta \partial_z) \hat{\Psi} = -i \sin^2 \theta [\delta_c + \hat{S}(z)] \hat{\Psi}, \quad \hat{\Phi} = 0. \quad (\text{S4})$$

In order to find the lowest order corrections to this limit, we first go back to Eqs. (5) and insert the equation for $\hat{\Phi}$ into that of $\partial_t \hat{\Psi}$, obtaining an equation of the form

$$(\partial_t + c \cos^2 \theta \partial_z) \hat{\Psi} = D' + D'', \quad (\text{S5})$$

where D' are terms associated with dispersion effects and D'' with dissipative effects. In the following we focus on each of these terms separately.

Dissipative terms. We find,

$$\begin{aligned} D'' = & -\frac{\sin^2 \theta \cos^2 \theta}{|\Omega|^2} \gamma [c \partial_z - i(\delta_c + \hat{S})] [\partial_t + i(\delta_c + \hat{S})] \hat{\Psi} \\ & + \frac{\sin \theta \cos^3 \theta}{|\Omega|^2} \gamma [c \partial_z - i(\delta_c + \hat{S})] [\partial_t + i(\delta_c + \hat{S})] \hat{\Phi} \\ & - \frac{\sin \theta \cos^2 \theta}{\Omega} \sqrt{n_a} [c \partial_z - i(\delta_c + \hat{S})] \hat{F}. \end{aligned} \quad (\text{S6})$$

For the lowest order correction to the adiabatic limit, we insert $\hat{\Psi}$ and $\hat{\Phi}$ from the adiabatic solution, Eqs. (S4) into D'' . Then, the second line in Eq. (S6) vanishes, whereas the first line scales as $\sin^2 \theta (\gamma/|\Omega|^2) T^{-2} \hat{\Psi}$ [noting from Eqs. (S4) and (S3) that $c \cos^2 \theta \partial_z \sim T^{-1}$]. Then, this term of D'' is negligible with respect to the leading contribution of the left hand side of Eq. (S5) (scaling as $T^{-1} \hat{\Psi}$) if

$$T^{-1} \ll \frac{|\Omega|^2}{\gamma} \frac{1}{\sin^2 \theta}. \quad (\text{S7})$$

For typical EIT conditions with $v \ll c$, we have $\sin^2 \theta \approx 1$ and we get that the single- and two-photon detunings of our system, T^{-1} , should be within the bandwidth $|\Omega|^2/\gamma$, often referred to as the single-atom EIT transparency window. It should be noted however that the usefulness of this single-atom

transparency window in predicting the losses in our system relies on the expansion in T^{-1} , i.e. on very narrow detunings, and that in the Autler-Townes regime for example, considerable losses are expected for $T^{-1} \sim \Omega \ll |\Omega|^2/\gamma$. Nevertheless, the real condition for lossless propagation has to take into account the entire medium and not just a single atom. It can be found by noting that the first line in D'' provides a decay rate for $\hat{\Psi}$ from which we find that the losses in a medium of length L are negligible for

$$T^{-1} \ll \frac{|\Omega|^2}{\gamma \sqrt{OD}} \equiv \delta_{tr}, \quad (\text{S8})$$

where δ_{tr} is the transparency window of the entire medium, as usual [1], which for large OD can become much narrower than both $|\Omega|^2/\gamma$ and Ω .

Finally, the quantum fluctuations induced by the reservoir formed by the vacuum of all non-guided modes, captured by the $\hat{F}$ term, are related to the losses by the fluctuation-dissipation theorem. Therefore, when losses are negligible, so is the effect of these fluctuations, upon comparison to the reservoir-free quantum fluctuations of $\hat{\Psi}$. We have verified this, using the equation for $\hat{\Phi}$ in Eq. (5), by showing that the variance which $\hat{F}$ adds to $\hat{\Phi}$ is negligible with respect to the vacuum noise of $\hat{\Phi}$.

Dispersive terms. We find,

$$\begin{aligned} D' = & -\frac{\sin^2 \theta \cos^2 \theta}{|\Omega|^2} [c \partial_z - i(\delta_c + \hat{S})] \partial_t [\partial_t + i(\delta_c + \hat{S})] \hat{\Psi} \\ & + \frac{\sin \theta \cos^3 \theta}{|\Omega|^2} [c \partial_z - i(\delta_c + \hat{S})] \partial_t [\partial_t + i(\delta_c + \hat{S})] \hat{\Phi} \\ & - i \sin^2 \theta (\delta_c + \hat{S}) \hat{\Psi}. \end{aligned} \quad (\text{S9})$$

Inserting the adiabatic solutions, Eqs. (S4), into the above expression, we obtain D' up to order $T^{-3} \hat{\Psi}$. Then, for approximately lossless propagation as in Eq. (S7), we take $D'' \approx 0$, and Eq. (S5) takes the form

$$(\partial_t + c \cos^2 \theta \partial_z) \hat{\Psi} = (i\hat{A} + \hat{B} \partial_z + i\hat{C} \partial_z^2 + \hat{D} \partial_z^3) \hat{\Psi}, \quad (\text{S10})$$

with hermitian (real) coefficients $\hat{A}, \hat{B}, \hat{C}$ and $\hat{D}$ expanded up to order T^{-3}, T^{-2}, T^{-1} and T^0 , respectively. Taking the lowest non-vanishing order for each coefficient, we find $\hat{S} \approx \hat{\delta}_{NL}$ and end up with Eq. (1) from the main text, plus an extra $\hat{D}$ -coefficient term with $\hat{D} = -(\sin^2 \theta/|\Omega|^2) v^3$. Nevertheless, for the regime explored in this work, namely, that of a probe-pulse bandwidth smaller than the combined detuning $\delta_c + \hat{\delta}_{NL}$, such cubic dispersion term is negligible (this was also verified by repeating calculations while including this term).

Finally, it should be noted that our derivation of Eq. (1) can be shown to be consistent with a similar adiabatic expansion in Ref. [1], upon relating the linear and nonlinear detunings $\delta_c + \hat{S}$ in Eqs. (5) to the time-dependence of the EIT mixing angle in [1] (Sec. V therein).

D. Significant dispersion in the lossless regime

The question is now under which conditions the above *dispersive* corrections to adiabaticity, giving rise to the ∂_z^2 term in Eq. (1), are significant while losses are nevertheless negligible. By comparing the first line of D' to that of D'' in Eqs. (S9) and (S6), respectively, we observe that dispersive effects are dominant over dissipative ones when $T^{-1} \gg \gamma$. On the other hand, for lossless propagation we have from (S7), $T^{-1} \ll |\Omega|^2/\gamma$, leading to the Autler-Townes condition $\Omega \gg \gamma$, as also explained

in Sec. 2A and Fig. 2 of the main text. More realistically, we consider lossless propagation with the condition (S8), leading to the requirement

$$\gamma \ll T^{-1} \ll \frac{|\Omega|^2}{\gamma \sqrt{OD}}, \quad (\text{S11})$$

and hence to a modified Autler-Townes-like condition $\gamma OD^{1/4} \ll \Omega$. In this work, we consider a case where deviations from adiabaticity are mainly due to the overall detuning of the coupling field, $\Delta_c = \delta_c + \delta_{NL}$ (in analogy to the intuitive explanation of Sec. 2A in the main text), so that the condition (S11) has to be satisfied for $T^{-1} \sim \Delta_c$.

2. BOGOLIUBOV THEORY FOR A NONLOCAL INTERACTION

A. Excitation spectrum

The polariton wave excitation spectrum (dispersion relation) around the CW solution, Eq. (8) in the main text, is found as follows. We first write the polariton field as a sum of an average CW $\langle \hat{\Psi}(z, t) \rangle = \psi(t)$ and a small fluctuation $\hat{\phi}(z, t)$,

$$\hat{\Psi}(z, t) = \psi(t) + \hat{\phi}(z, t), \quad (\text{S12})$$

where the average CW solution is that of self-phase modulation, $\psi(t) = \psi_0 e^{-i(\alpha\delta_c + n_p U_0)t}$ 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 $U(z)$,

$$U_k = \int_{-\infty}^{\infty} dz U(z) e^{-ikz}. \quad (\text{S13})$$

The integral for U_0 that arises in the above CW solution $\psi(t)$ 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 (S13). 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 $L \gg l$ (see also Sec. 2D below).

Inserting Eq. (S12) into the nonlinear equation (1) in the main text and keeping terms only to linear order in the perturbation $\hat{\phi}$, we obtain

$$\begin{aligned} (\partial_t + v\partial_z)\hat{\phi}(z) &= -i(\alpha\delta_c + n_p U_0)\hat{\phi}(z) \\ &+ i(n_p U_0 / \alpha + \delta_c) C v^2 \partial_z^2 \hat{\phi}(z) - i n_p \int_L dz' U(z - z') \hat{\phi}(z') \\ &- i n_p e^{i2[\phi - (\alpha\delta_c + n_p U_0)t]} \int_L dz' U(z - z') \hat{\phi}^\dagger(z'). \end{aligned} \quad (\text{S14})$$

This linearized equation is valid as long as the fluctuations $\hat{\phi}$ around the mean ψ are small, i.e. for

$$|\psi| \gg |\langle \hat{\phi} \rangle|, \sqrt{\langle \hat{\phi}^\dagger \hat{\phi} \rangle}, \sqrt{\langle \hat{\phi}^2 \rangle}. \quad (\text{S15})$$

In order to find the dispersion relation of the fluctuations it is enough to consider the case of a classical field $\hat{\phi} \rightarrow \phi$. We then insert the ansatz for $\phi(z, t)$ from Eq. (7) in the main text into the linearized equation (S14), obtaining coupled equations for u_k and v_k

$$\begin{pmatrix} \omega_k^0 + n_p U_k - \omega_k & -n_p U_k \\ -n_p U_k & \omega_k^0 + n_p U_k + \omega_k \end{pmatrix} \begin{pmatrix} u_k \\ v_k \end{pmatrix} = 0. \quad (\text{S16})$$

In order to arrive at the equations (S16), we used

$$\int_0^L dz' U(z - z') e^{ikz'} \approx \int_{-\infty}^{\infty} U(\xi) e^{ik\xi} e^{ikz} = U_k e^{ikz}, \quad (\text{S17})$$

which again amounts to neglecting edge effects, and where in the last equality we assumed that $U(z)$ is symmetric and real so that $U_{-k} = U_k$. Eq. (S16) 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 ω_k from Eq. (8).

B. Dynamic Bogoliubov theory

Here 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. (10) in the main text. We expand the quantum field $\hat{\Psi}(z, t)$ in spatial Fourier modes $\hat{a}_k(t)$, $\hat{\phi}(z) = \sum_k (1/\sqrt{L}) e^{ikz} \hat{a}_k$, and the transformed modes $\hat{c}_k(t)$ as,

$$\begin{aligned} \hat{\phi}(z, t) &= \sum_k \frac{1}{\sqrt{L}} e^{ik(z-vt)} e^{i[\phi - (\alpha\delta_c + n_p U_0)t]} \hat{c}_k(t), \\ \hat{a}_k(t) &= e^{i\phi} e^{-i(\alpha\delta_c + n_p U_0 + kv)t} \hat{c}_k(t), \end{aligned} \quad (\text{S18})$$

where the operators $\hat{a}_k(t)$ satisfy the equal-time commutation relations $[\hat{a}_k(t), \hat{a}_{k'}^\dagger(t)] = \delta_{kk'}$ and hence so do the operators $\hat{c}_k(t)$. Inserting Eq. (S18) into the linearized equation (S14) and neglecting edge effects as in Eq. (S17), 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 $\hat{c}_{-k}^\dagger$,

$$\partial_t \begin{pmatrix} \hat{c}_k \\ \hat{c}_{-k}^\dagger \end{pmatrix} = -i \begin{pmatrix} \omega_k^0 + n_p U_k & n_p U_k \\ -n_p U_k & -\omega_k^0 - n_p U_k \end{pmatrix} \begin{pmatrix} \hat{c}_k \\ \hat{c}_{-k}^\dagger \end{pmatrix}. \quad (\text{S19})$$

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. (10) in the main text.

C. Intensity correlations

Intensity correlations are characterized by the normalized second order coherence function defined by [3]

$$g^{(2)}(z, z') = \frac{G^{(2)}(z, z')}{G^{(1)}(z)G^{(1)}(z')} \quad (\text{S20})$$

with

$$\begin{aligned} G^{(2)}(z, z') &= \langle \hat{\Psi}^\dagger(z, t) \hat{\Psi}^\dagger(z', t) \hat{\Psi}(z', t) \hat{\Psi}(z, t) \rangle, \\ G^{(1)}(z) &= \langle \hat{\Psi}^\dagger(z, t) \hat{\Psi}(z, t) \rangle. \end{aligned} \quad (\text{S21})$$

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,

$$\begin{aligned} \hat{\Psi}(z, t) &= \frac{1}{\sqrt{L}} \sum_k e^{ikz} \hat{a}_k(t) \approx \frac{a_0(t)}{\sqrt{L}} + \frac{1}{\sqrt{L}} \sum_{k \neq 0} e^{ikz} \hat{a}_k(t), \\ a_0(t)/\sqrt{L} &= \sqrt{n_p / \alpha^2} e^{i\phi} e^{-i(\alpha\delta_c + n_p U_0)t}, \end{aligned} \quad (\text{S22})$$

where $\hat{a}_k(t)$ is given by Eq. (10) in the main text. Inserting Eq. (S22) into $G^{(1)}(z)$ from Eq. (S21), we find

$$G^{(1)}(z) = \frac{n_p}{\alpha^2} + \frac{1}{L} \sum_{k \neq 0} \left[|\mu_k|^2 N_k + |v_k|^2 (N_k + 1) \right], \quad (\text{S23})$$

where we have assumed the following statistics of the initial fluctuations $\hat{a}_{k \neq 0}$: $\langle \hat{a}_k(0) \rangle = 0$, $\langle \hat{a}_k^\dagger(0) \hat{a}_{k'}(0) \rangle = N_k \delta_{kk'}$ 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. (S22) into $G^{(2)}$ from Eq. (S21), keeping terms only to second order in the fluctuations $\hat{a}_{k \neq 0}$, in accordance with the assumptions (S15), and obtain

$$\begin{aligned} G^{(2)}(z, z') &= \frac{n_p^2}{\alpha^4} + 2 \frac{n_p}{\alpha^2} \\ &\times \frac{1}{L} \sum_{k \neq 0} \left\{ \left[|\mu_k|^2 N_k + |v_k|^2 (N_k + 1) \right] (1 + \cos[k(z - z')]) \right. \\ &\left. + |v_k| |\mu_k| (1 + 2N_k) \cos[k(z - z') - \phi_k] \right\}, \end{aligned} \quad (\text{S24})$$

where $\phi_k = \arg(\mu_k v_k)$. Inserting $G^{(1)}$ and $G^{(2)}$ from Eqs. (S23) and (S24) in Eq. (S20), we note that to lowest order in the fluctuations we can take $G^{(1)} \approx n_p / \alpha^2$ such that $g^{(2)} \propto G^{(2)}$. Then, 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. (11) from the main text.

D. Neglecting edge effects: real space and k -space descriptions

In sections 2A and 2B we neglect edge effects by using the approximation (S17) for a medium length L much larger than the potential range l . The reasoning, relying on a real-space description, goes as follows: Each "slice" of the field $\varphi(z)$ (or more generally, Ψ) enters the medium at $z = 0$ and leaves it at $z = L$. When it is well within the bulk of the medium, i.e. around points z that are at a distance larger than l from both edges, $l < z < L - l$, this slice interacts with all slices between $z \pm l$, which can practically be described as if it interacts with all slices between $(-\infty, \infty)$ due to the exponential decay of the potential with range l . This is the essence of the approximation (S17). When the slice is close to the edges at $z \in [0, l], [L - l, L]$, the interaction with other slices is not symmetric around z anymore and the overall effect on the slice at z is different than within the bulk of the medium. However, for $l \ll L$, bulk-like effects dominate over the boundary-like edge effects. Therefore, we expect that treating all z -points as if they are within the bulk medium should make a good approximation.

This point can also be seen in k -space. Introducing into the linearized equation (S14) the Fourier decompositions of $\hat{\varphi}$ and $U(z)$,

$$\begin{aligned} \hat{\varphi}(z) &= e^{i[\phi - (\alpha \delta_c + n_p U_0)t]} \int_{-\infty}^{\infty} \frac{dk}{2\pi} \hat{c}_k e^{ik(z - vt)} e^{\mp i \omega_k}, \\ U(z) &= \int_{-\infty}^{\infty} \frac{dk}{2\pi} U_k e^{ikz}, \end{aligned} \quad (\text{S25})$$

with the $\mp$ sign in ω_k is for $\pm k$, respectively, and using the notations $u_k = c_k$ and $v_k^* = -c_{-k}$, we obtain

$$\begin{aligned} 0 &= (\omega_k^0 - \omega_k) u_k + n_p U_k \int_{-\infty}^{\infty} \frac{dk'}{2\pi} e^{[\omega_k - \omega_{k'} + v(k - k')]t} \delta_L(k - k') u_{k'} \\ &- n_p U_k \int_{-\infty}^{\infty} \frac{dk'}{2\pi} e^{[\omega_k - \omega_{k'} + v(k - k')]t} \delta_L(k - k') v_{k'}, \end{aligned} \quad (\text{S26})$$

with $\delta_L(k) = L \sin(kL/2) / (kL/2)$. For an infinite medium $L \rightarrow \infty$, where edges are absent, we have $\delta_L(k) \rightarrow \delta(k)$ and the above equation indeed reproduces Eq. (S16) and hence the spectrum ω_k from the main text. However, in general, it describes the mixing of different k -modes due to the finite medium, such that each k -space point is coarse-grained with a kernel of spectral width $\sim 1/L$. Nevertheless, considering that the dependence of u_k, v_k and ω_k on k is mostly determined by U_k , they vary at a scale $1/l$ which is slower compared to $1/L$ variation, for $l \ll L$. Therefore, in the integrations over dk' in Eq. (S26) we can replace $\delta_L(k - k') \rightarrow \delta(k - k')$, reproducing Eq. (S16).

3. LASER-INDUCED DIPOLE-DIPOLE INTERACTION

The laser-induced interaction potential between a pair of atoms, e.g. at positions z_1 and z_2 along the z -axis, is given by [4]

$$U(z_1, z_2) = -\frac{|\Omega_L|^2}{\delta_L} \hbar \Delta_{dd}(z_1, z_2) \cos[k_L^z(z_1 - z_2)], \quad (\text{S27})$$

with the same notations from Sec. 5 (and Fig. 1c) of the main text and where $\Delta_{dd}(z_1, z_2)$ is the so-called resonant dipole-dipole interaction evaluated at the laser frequency ω_L . $\Delta_{dd}(z_1, z_2)$ is responsible for dispersive interactions (excitation exchange) between the atoms mediated by virtual excitations of the photon modes that couple to the atoms. Therefore, the spatial dependence of $\Delta_{dd}(z_1, z_2)$, which in turn determines the potential $U(z_1, z_2)$, directly follows that of the photon modes. And in a confined geometry, these can alter dramatically and lead to modified and long range dipolar interactions Δ_{dd} and potentials U [4–8]. For a dipolar interaction at a frequency inside the bandgap of a waveguide grating, we obtain [6, 7]

$$\begin{aligned} \Delta_{dd}(z_1, z_2) &\approx \gamma \eta(z_1, z_2) \cos[k_B(z_1 - z_2)] e^{-|z_1 - z_2|/l}, \\ \eta &= \frac{1}{\sqrt{1 - \omega_L/\omega_u}} \frac{3\pi \hbar \sqrt{\hbar \Delta n}}{4\omega_L^2/c^2} E(z_1) E^*(z_2), \\ l &= \frac{1}{\sqrt{1 - \omega_L/\omega_u}} \frac{1}{k_B \sqrt{\hbar \Delta n}}. \end{aligned} \quad (\text{S28})$$

Here γ is the emission rate of the dipolar transition ($|d\rangle \rightarrow |s\rangle$ in our case, see Fig. 1c in the main text) and $E(z_m)$ is the transverse profile of the waveguide mode for atom m (units of inverse length), evaluated at the transverse position of the atoms r_a . Its dependence on z_m is that of the envelope of a Bloch wave and is hence periodic with the grating period Λ . Then, considering atoms trapped Λ -apart (which can be achieved using a laser with a wavelength $2\Lambda = 2\pi/k_B$ not too far from the atomic transitions), i.e. at positions $z_m = z_0 + m\Lambda$, we have $E(z_m) = E(z_0)$ identical for all atoms, so that $1/|E(z_0)|^2 \equiv A$ becomes the effective area of the mode and the energy potential depends only on the relative coordinate $z = |z_1 - z_2|$. In this respect, we note that the continuum description of EIT used here is a spatially coarse-grained picture [1] where all atomic and field operators are envelopes with a spatial resolution that must be limited by the transparency window, several orders of magnitude narrower than typical wavelengths of the probe field and atomic transitions (and hence Λ). Therefore, the interacting polariton field description of Eq. (1) in the main text does not resolve the individual atomic positions (Λ -apart), and we use the continuous potential from Eq. (12).

4. QUANTITATIVE ILLUSTRATION

Let us specify the system parameters used in the illustration in Figs. 3 and 4. Motivated by the experiment in Ref. [2], we

take the wavelength, dipolar matrix element and radiative decay rate of the D1 line of Cs atoms [9] as the typical parameters of all dipolar atomic transitions in Fig. 1, and consider atoms 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 an effective area of the fiber's HE_{11} transverse mode $A = 4.42 \mu\text{m}^2$ [6]. Taking an atom density of $n_a \sim 16 \times 10^6 \text{ m}^{-3}$, length $L = 2.68$ cm, and coupling laser with Rabi frequency $\Omega = 4 \times 10^8 \text{ s}^{-1}$ ($2 \times 10^8 \text{ s}^{-1}$ and $1.2 \times 10^8 \text{ s}^{-1}$ in Figs. 3b and 4, respectively) and detuning $\delta_c = -3.84 \times 10^7 \text{ s}^{-1}$ in Fig. 3a ($4.795 \times 10^6 \text{ s}^{-1}$ and $-3.64 \times 10^6 \text{ s}^{-1}$ in Figs. 3b and 4), we obtain $\alpha = 0.999839$ (0.99996 and 0.999986 in Figs. 3b and 4), $v = 48216 \text{ ms}^{-1}$ (12055 ms^{-1} in 4340 ms^{-1} in Figs. 3b and 4, respectively) and $\delta_{tr} = 8.66 \times 10^7 \text{ s}^{-1}$ ($2.16 \times 10^7 \text{ s}^{-1}$ and $7.79 \times 10^6 \text{ s}^{-1}$ in Figs. 3b and 4), so that Figs. 3a,b and 4a,b are effectively cut at $k = q_{tr} = \delta_{tr}/v = 1795 \text{ m}^{-1}$. For the Bragg grating imprinted on the fiber/waveguide [10, 11], 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 $I = 2 \times 10^4 \text{ Wm}^{-2}$ ($0.5 \times 10^4 \text{ Wm}^{-2}$ and 10^2 Wm^{-2} in Figs. 3b and 4), yielding $U_L = 2\eta R_{fs} = 1.14 \times 10^6 \text{ s}^{-1}$ ($2.85 \times 10^5 \text{ s}^{-1}$ and 5696 s^{-1} in Figs. 3b and 4), where $\eta \sim 12$ is the ratio between emission to the fiber grating modes and to free-space, and $R_{fs} = \gamma|\Omega_L|^2/(2\delta_L^2) = 47443 \text{ s}^{-1}$ (11860 s^{-1} and 237 s^{-1} in Figs. 3b and 4) is the resulting scattering rate to free space from the $|d\rangle \rightarrow |s\rangle$ transition, leading to a potential range of $l \approx 3027\lambda_L \approx 0.1L \approx 0.0027\text{m}$. The orientation angle is taken to be $\theta_L = 0.131$, so that $k_R = k_L \cos \theta_L - k_B = 1019 \text{ m}^{-1}$. We assume a CW background with a power 2×10^{-12} W in Fig. 3a (10^{-10} W and 4×10^{-10} W in Figs. 3b and 4), giving $n_p \approx 187 \text{ m}^{-1}$ (37347 m^{-1} and 414981 m^{-1} in Figs. 3b and 4). For the $N_k(t)$ and $g^{(2)}$ calculations in Figs. 4b,c, we consider an initial Gaussian spectrum of intensity fluctuations, $N_k = N_0 e^{-(k/q_{tr})^2}$, with $N_0 = 5$. Finally, wherever a comparison with the local-interaction case is made, the local potential is taken as $U(z) = (1/8)U_L \delta(z)$.

5. NUMERICAL SIMULATIONS OF EQ. (1)

In order to obtain the ω_k spectrum, we perform numerical simulations of the full nonlinear equation (1), where the field operator $\hat{\Psi}$ is replaced by its expectation value Ψ (classical field). We use a three-term splitting method, capable of dealing with the nonlocal and nonlinear dispersion coefficient that appears in Eq. (1), unlike the more common (two-term) split-step method. We study the dynamics of the field, initially comprised of the CW solution ψ and weak perturbations φ at a spatial frequency k , and extract the field's temporal oscillation frequency $\omega(k)$ leading to the spectrum ω_k . For weak enough perturbations where the Bogoliubov theory applies, the results of the simulation of Eq. (1) agree very well with those of the simpler split-step method simulations, where the nonlinear dispersion term $\delta_{NL} C v^2 \partial_z^2$ is approximated by its mean-field value, $\delta_{NL} = n_p U_0 / \alpha$, so that the equation becomes of semilinear type. For the case of instability (Fig. 4b,c), we begin with the initial intensity fluctuations with a spectrum N_k and run split-step simulations with the mean-field nonlinear dispersion term $\delta_{NL} = n_p U_0 / \alpha$. Then, after propagation time t , we measure $N_k(t) = \langle a_k^*(t) a_k(t) \rangle$ 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 Ψ .

The finite size of the medium L is treated by taking periodic

boundary conditions for the field $\Psi(z)$ and the potential $U(z)$, so that the k -space (spatial frequencies) is sampled at integer multiples of $2\pi/L$ as can be seen e.g. in Fig. 3 of the main text. This $2\pi/L$ limit on the k -space resolution simulates the coarse-graining in k -space with the kernel $\delta_L(k)$ of width $\sim 2\pi/L$ of the finite medium, as in Eq. (S26). This relation can be explicitly shown by considering the Fourier series of the periodic potential $U(z)$ and field fluctuation $\varphi(z)$, which are given by Eq. (S25) with the substitution $\int dk / (2\pi) \rightarrow \sum_k$, where k now assumes the discrete values $k = 2\pi q/L$ (with integer q), and where the Fourier coefficients c_k, U_k are replaced by

$$\tilde{c}_k = \frac{1}{L} \int_L dz \varphi(z) e^{-ikz}, \quad \tilde{U}_k = \frac{1}{L} \int_L dz U(z) e^{-ikz}. \quad (\text{S29})$$

Then, from Eq. (S14), and using the relation between Fourier series and Fourier transform coefficients, $U_k = L \tilde{U}_k$, we obtain

$$0 = (\omega_k^0 + n_p U_k - \omega_k) u_k - n_p U_k v_k, \quad (\text{S30})$$

with $k = 2\pi q/L$ and $u_k = c_k, v_k = -c_{-k}^*$. Eq. (S30) is nothing but the sampling of Eq. (S16) at the discrete k -values of the L -periodic medium. The relation to the L -finite medium result, Eq. (S26) is that coarse-grained values around each $k = 2\pi q/L$ are replaced in Eq. (S30) by their actual values. When the width of the kernel of the coarse-graining, $\sim 1/L$ in our case, is narrower than the typical change of the coarse-grained functions, $\sim 1/l$ in our case, then Eq. (S30) becomes a very good approximation of Eq. (S26). Therefore, for $l \ll L$ we expect the numerical simulations to approximate well the finite medium while also allowing to consider its limited spatial resolution by the discrete sampling of k -space.

6. SCATTERING AND IMPERFECTIONS

In the following we address the main loss and dephasing mechanisms which affect the polariton evolution. For each of them we estimate the rate at which they decohere the polariton field; then, by comparing the analytical expression of this decoherence rate with that of the spectra ω_k (or $\gamma_k = \text{Im}\omega_k$ in the case of instability), we study how the limitations it imposes on the observability of the spectra can be relaxed.

A. Scattering of off-resonant laser photons

Beginning with the illumination of the $|d\rangle \rightarrow |s\rangle$ atomic transition by the off-resonant laser Ω_L , it leads to an interaction potential $U(z)$, but is also accompanied by scattering of photons to non-guided modes at a rate $R_{fs} = \gamma|\Omega_L|^2/(2\delta_L^2)$ [4], which decoheres the spin wave $\bar{\sigma}_{gd}$, and hence the polariton, at rate R_{fs} . This rate has to be compared to the frequency scale $U_L = 2\eta R_{fs}$ of the coherent interaction-related effects such as ω_L , where η is a geometrical factor related to the mode-density of the guided modes. For the case of waveguide-grating modes, η can be larger than 1 ($\eta \sim 12$ in our numerical example, see Sec. 4 here) so coherent effects prevail [6]. In fact, a deeper reason for the dominance of the coherent effects discussed here (the spectra ω_k and γ_k) results from the fact that these are all *cooperative* effects, proportional to the density of excitations (EIT polaritons) n_p in the system, whereas the scattering R_{fs} is essentially a *single-atom* effect since photons are scattered to non-guided, non-confined modes (which mediate much weaker cooperative effects) [6]. Therefore, ω_k scales as $n_p U_L \propto n_p \eta R_{fs}$ [Eq. (8)] so that even for small η , it can be made much larger than R_{fs} by e.g. increasing n_p (increasing the laser power of the background CW probe field). Indeed, for the specific numerical example taken

Fig. S1. (a) Instability case. Spectrum of amplification $\gamma_k = \text{Im}\omega_k$ for the example considered in the main text (as in Fig. 4a therein): nonlocal potential (blue solid curve) and local potential (red dashed curve). (b) Overcoming cooperative decoherence $R_{im}^{(2)}$: anti-roton case. Same parameters as in the main text (Fig. 3b therein) apart from Ω which is increased by a factor 12. (c) Same as (b), however this time for the case of instability γ_k and where Ω is increased by a factor 8.5.

here (Sec. 4 here), we obtain $R_{fs} = 47443 \text{ s}^{-1}, 11860 \text{ s}^{-1}, 237 \text{ s}^{-1}$ for the roton, anti-roton and instability cases, respectively, yielding decoherence rates R_{fs} much smaller than the typical frequency resolution of the corresponding spectra ω_k of the roton and anti-roton (Figs. 3a and 3b), and γ_k of the instability (Fig. S1a here).

B. Scattering by imperfections in the waveguide structure

The effect of imperfections on the guided modes k is typically described by adding an imaginary part $-ik$ to their mode frequencies. This can be consistently shown by e.g. considering pure modes which are dipole-coupled to scatterers modelled as lossy and localized harmonic oscillators far-detuned from the modes k [12]. Recalling that the interaction $U(z)$ between the atoms is mediated by the photon modes k , and taking their width κ into account, U now becomes complex,

$$\begin{aligned} U(z) &= U'(z) - iU''(z), \\ U'(z) &= -\cos(k_L^2 z) \frac{\Omega_L^2}{\delta_L^2} \int_0^\infty d\omega \frac{G(\omega, z)(\omega - \omega_L)}{(\kappa/2)^2 + (\omega - \omega_L)^2}, \\ U''(z) &= -\cos(k_L^2 z) \frac{\Omega_L^2}{\delta_L^2} \int_0^\infty d\omega \frac{G(\omega, z)\kappa/2}{(\kappa/2)^2 + (\omega - \omega_L)^2}, \end{aligned} \quad (\text{S31})$$

where $G(\omega, z)$ is the spectrum of the photon-modes reservoir coupled to a pair of atoms z -apart [4, 5, 7, 12]. The existence of the imaginary part $-iU''$ reflects the fact that now the modes k not only mediate a dispersive interaction between the atoms (U'), but also mediate scattering of laser photons Ω_L to non-guided modes via the atoms and the imperfections in the waveguide.

For the modes k of a waveguide with a bandgap, as in our example, we have [6, 7]

$$G(\omega, z) \approx \gamma \frac{3\bar{n}\sqrt{\bar{n}\Delta n} \cos[k_B z(1 + \sqrt{\bar{n}\Delta n}\sqrt{\omega/\omega_u - 1})]}{4A\omega_L^2/c^2 \sqrt{\omega/\omega_u - 1}}, \quad (\text{S32})$$

with the effective area A from Sec. 3 here. We now turn to the estimation of three distinct decoherence rates associated with this imperfection-induced loss mechanism.

Single-atom scattering.— The resulting decoherence rate on a single atom $R_{im}^{(1)}$, is obtained by taking $z \rightarrow 0$ in $U''(z)$. Performing the integration in (S31) using (S32) we find $U(0) = -U_L/\sqrt{1 - i\kappa/(2\delta_u)}$ with $\delta_u = \omega_u - \omega_L$ (ω_u being the frequency of the upper bandedge) and $U_L = \eta|\Omega_L|^2/\delta_L^2\hbar\gamma$ [Eqs. (S27) and (S28)]. For our numerical example we first need

to estimate κ : current state-of-the-art nano-waveguide-based cavities can have an imperfection-induced Q -factor of up to $\sim 4 \times 10^5$ for a cavity resonance at $\sim 800 \text{ nm}$, so that $\kappa \sim 5 \times 10^9 \text{ s}^{-1}$. Going back to our numerical example from the main text, we have $\delta_u \sim 0.2 \times 10^9 \text{ s}^{-1}$, yielding $U(0) \approx -U_L(0.21 + i0.19)$, so the rate of this laser-induced scattering $R_{im}^{(1)} = U''(0)$ is comparable to the coherent potential $U'(0)$. We note however that $R_{im}^{(1)}$ describes a *single-polariton* decoherence rate and hence can be made much smaller than the cooperative effects represented by ω_k , by increasing n_p (as explained for the R_{fs} case above).

Cooperative scattering.— Atoms located at a distance z apart can also scatter photons in a cooperative manner, leading to a mutual decoherence rate $U''(z)$. The resulting *cooperative decoherence* per atom is then obtained by summing over interfering mutual scattering from all $|d\rangle$ -populated atoms in the medium, $R_{im}^{(2)} = n_p \int_L U''(z)$, where n_p is the density of polariton excitations in the medium (which is roughly the density of $|d\rangle$ -populated atoms). Performing the integrations on z and ω [see Eqs. (S31) and (S32)] we obtain $R_{im}^{(2)} \approx n_p|U_0|\kappa\Delta_u/(\kappa^2/4 + \Delta_u^2)$, which valid for $\Delta_u \gg \delta_u$ and where $\Delta_u = \omega_u(k_R/k_B)^2/(\bar{n}\Delta n)$ ($k_R = k_L^z - k_B$ being the roton wavenumber). We now need to compare $R_{im}^{(2)}$ to $\omega_k = \sqrt{\omega_k^0(\omega_k^0 + 2n_p U_k)}$. It is easy to show that $\omega_k^0 \propto C\bar{v}^2 \propto \Omega^2$, so that near the peak of ω_k , where $2n_p U_k$ is dominant over ω_k^0 , we have $\omega_k \propto \Omega$. On the other hand, $R_{im}^{(2)}$ essentially does not depends on Ω . Therefore, by increasing Ω and keeping n_p fixed, ω_k can in principle always be made dominant over $R_{im}^{(2)}$.

Considering our numerical example we find $\Delta_u = 1.45 \times 10^9 \text{ s}^{-1}$ satisfying the condition $\Delta_u \gg \delta_u \sim 0.2 \times 10^9 \text{ s}^{-1}$, so that with $\kappa \sim 5 \times 10^9 \text{ s}^{-1}$ we have $R_{im}^{(2)} \approx 0.87n_p U_0$. Then, for the anti-roton case, we find $R_{im}^{(2)} \sim 15 \times 10^5 \text{ s}^{-1}$, much larger than the anti-roton peak of a few 10^5 s^{-1} (Fig. 3b). This can be improved by increasing Ω by e.g. a factor of $f = 12$, yielding the pronounced anti-roton spectrum of Fig. S1b. Similarly, for the case of instability, we find $R_{im}^{(2)} \sim 3.3 \times 10^5 \text{ s}^{-1}$, larger than the amplification rate γ_k which is peaked at a value of about $1.5 \times 10^5 \text{ s}^{-1}$. Again, this can be remedied upon increasing Ω by a factor $f = 8.5$ for example, resulting in the amplification profile of Fig. S1c. For the roton case, we have $R_{im}^{(2)} \sim 0.3 \times 10^5 \text{ s}^{-1}$, which is already smaller than the required resolution of the roton "dip" of about 10^5 s^{-1} .

Polariton propagation losses.— Eq. (5) from the main text defines the polariton Ψ as a superposition of an atom (spin wave) and a propagating photon. The origin of both $R_{im}^{(1)}$ and $R_{im}^{(2)}$ discussed above are in its atomic component, whereas its photonic component, essentially comprised of the modes k , also decays at a rate κ due to photon propagation in the presence of imperfections. This means that the polariton field suffers from an additional decoherence given by κ times the fraction of its photon component $\cos^2 \theta$ (likewise, $R_{im}^{(1,2)}$ should be multiplied by $\sin^2 \theta$, however as typical of EIT, we have $\sin^2 \theta \approx 1$). Nevertheless, unlike all other decoherence rates discussed here (R_{fs} , R_{EIT} and $R_{im}^{(1,2)}$), this loss mechanism is *independent* of the existence of the interaction $U(z)$ (it does not

Fig. S2. EIT absorption-rate spectrum for a probe field envelope (wavenumber k around the carrier k_0) inside an EIT medium with a detuned coupling field Δ_c : Eq. (S33) with $\Delta_p = \delta_p + kv$ and $\Delta_c = \delta_c + n_p U_0/\alpha$ (physical parameters of Sec. 4 here). (a) Anti-roton case: $\delta_p = 0$ (blue curve) and $\delta_p = \Delta_c$. (b) Same as (a), for instability case. (c) Same as (a), for roton case.

depend on the laser Ω_L), and hence can be first measured in the absence of U and then corrected for in measurements where U is switched on.

C. EIT propagation losses

The analysis based on Eq. (1) of the main text is only exact for CW polaritons and vanishing coupling-field detuning $\Delta_c = 0$, however it makes a good approximation for a polariton bandwidth and Δ_c well within the transparency window $\delta_{tr} = cq_{tr}$, which is the regime considered in this work [as explained in Sec. 3A and the adiabatic expansion leading to Eq. (1)]. Here we consider the full expression for EIT losses and show that the above transparency-window considerations indeed lead to a low-loss regime. Moreover, we show that EIT-loss-related limitations on the observability of the cooperative spectra may possibly decrease by working around two-photon resonance.

The absorption per unit length of a probe field (wavenumber k_0) via medium with imaginary susceptibility χ'' is $(1/2)k_0\chi''$, yielding an absorption rate $(1/2)kv\chi''$, where v is the group velocity of the field. Using the probe susceptibility of an EIT medium [13, 14], we then obtain the EIT absorption rate,

$$R_{EIT} = \frac{1}{4} \frac{OD}{L} v \frac{(\gamma^2/2)(\Delta_p - \Delta_c)^2}{[\Delta_p(\Delta_p - \Delta_c) - |\Omega|^2]^2 + (\gamma^2/4)(\Delta_p - \Delta_c)^2}, \quad (\text{S33})$$

where Δ_c is the detuning of the coupling field Ω , Δ_p is the detuning of the probe, γ is the width of the atomic level $|e\rangle$ and OD is the optical depth of the medium (see main text). In our case, the probe field is a pulse hence the detuning for each k wavenumber around the resonant carrier k_0 is $\Delta_p = vk$. We can then plot the $R_{EIT}(k)$ loss-rate spectrum given the overall control detuning $\Delta_c = \delta_c + \delta_{NL}$, with the nonlinear contribution $\delta_{NL} = n_p U_0/\alpha$ evaluated by its mean-field value. Referring back to our numerical example, Fig. S2 presents the spectrum R_{EIT} within the transparency window $k \in [-q_{tr}, q_{tr}]$ for the three cases of anti-roton, instability and roton [recalling that $q_{tr} = \delta_{tr}/v = |\Omega|^2/(\gamma\sqrt{OD}v)$].

Focusing on the blue curves first, we note that for all three cases (a,b,c) R_{EIT} is not symmetric around $k = 0$ due to non-vanishing Δ_c , just as explained and illustrated by Fig. 2b of the main text. Comparing the EIT-loss rate spectrum R_{EIT} to the

coherent and cooperative dispersion spectra ω_k , we indeed see that for all three cases the coherent effects prevail as expected in our Autler-Townes regime (see Sec. 3A in the main text and Sec. 1D here): comparing Figs. 3(a), 3(b) (main text) and S1(a) (here) to Figs. 2S(c,a,b), respectively, we see that the coherent effects are larger by about an order of magnitude for all three cases. This demonstrates that our transparency-window-based considerations, leading to the requirement $\Delta_c, \Delta_p \ll \delta_{tr}$ (with δ_{tr} evaluated for the usual symmetric EIT window as in Fig. 2a, main text), are in general sufficient to ensure the dominance of coherent effects over EIT losses, in our regime.

In order to be able to observe the roton-like features of the spectra ω_k (and γ_k), we have to make sure that R_{EIT} is also smaller than the resolution required to observe these features around the roton feature at $k_R = 1019\text{m}^{-1}$ (rather than just being smaller than ω_k). For the anti-roton the required resolution of $\sim 10^5\text{s}^{-1}$ (Fig. 3b) indeed exceeds $R_{EIT} \sim 0.2 \times 10^5\text{s}^{-1}$ (Fig. S2a around $k_R \approx 1000\text{m}^{-1}$). For the instability case, the required resolution of $\sim 0.2 \times 10^5\text{s}^{-1}$ is of the same order of R_{EIT} (Fig. S2b, blue curve), however this can be improved by considering a detuned carrier for the probe, such that two-photon resonance around the carrier is established, i.e. for $\Delta_p = \Delta_c + kv$ (Fig. S2b, red curve). For the roton case the situation is more challenging, as the roton "dip" is more sensitive and requires a resolution of $\sim 10^5\text{s}^{-1}$, which is of the same order of magnitude as R_{EIT} (Fig. S2c).

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