Polaritonic frequency-comb generation and breather propagation in a negative-index metamaterial with a cold four-level atomic medium

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(Dated: February 13, 2019)

We develop a concept for a waveguide that exploits spatial control of nonlinear surface-polaritonic waves. Our scheme includes an optical cavity with four-level N-type atoms in a lossless dielectric placed above a negative-index metamaterial layer. We propose exciting a polaritonic Akhmediev breather at a certain position of the interface between the atomic medium and the metamaterial by modifying laser-field intensities and detunings. Furthermore, we propose generating position-dependent polaritonic frequency combs by engineering widths of the electromagnetically induced transparency window commensurate with the surface-polaritonic modulation instability. Therefore, this waveguide acts as a high-speed polaritonic modulator and position-dependent frequency-comb generator, which can be applied to compact photonic chips.

Nonlinear plasmonics (and polaritonics) [11] in waveguide geometries are of strong interest for schemes enabling strong cross-phase modulation [2], amplification and lasing [3], modulators [4] and detection [5]. Controlling and exciting nonlinear surface polaritons (SPs) is challenging as the strength of the nonlinear processes and their efficiency depend strongly on (metallic) nanostructure roughness [6, 7] which is experimentally challenging to minimize. We circumvent this problem by formulating an approach that spatially controls nonlinear SP waves, and we explore its application for modulation [4] and frequency-comb generators [8].

For spatial control of nonlinear surface-polaritonic waves, we suggest driving four-level N-type atoms (4NAs) [9] on the surface of a negative-index metamaterial (NIMM) [10] as depicted in Fig. 1. These components are contained in a stable cavity and serve as a nonlinear planar waveguide. The atoms are dopants in a transparent medium over a thickness of several dipole-transition wavelengths. These atoms are driven by three co-propagating fields a pump signal (s), a weak probe signal (p), and a standing wave coupling signal (c), all assumed injected from laser beams using the end-fire coupling technique. The cavity produces a resonant mode at the coupling frequency only, so this wave is illustrated as a standing wave.

\[ |4⟩ ↔ |1⟩, \ |3⟩ ↔ |1⟩ \text{ and } |3⟩ ↔ |2⟩ \text{ atomic transitions, respectively.} \]

The |4⟩ ↔ |1⟩, |3⟩ ↔ |1⟩ and |3⟩ ↔ |2⟩ atomic transitions, respectively. The 4NA medium in our waveguide is assumed as Pr^{3+} in Y_2SiO_5 with corresponding energy levels

\[ |1⟩ = |^3H_4, \pm 5/2⟩, \ |2⟩ = |^3H_4, \pm 3/2⟩, \]
\[ |3⟩ = |^1D_2, \pm 3/2⟩, \ |4⟩ = |^1D_2, \pm 5/2⟩. \] (1)

We assume inhomogeneous broadening of the atomic transitions to be in Lorentzian line shape [11]. The 4NA medium has atomic density \( N_a \), natural decay rates \( \Gamma_{nn} \) and dephasing rates \( \gamma_{nn} \) between levels \( |n⟩ \) and \( |m⟩ \) [17].

The signal (s), probe (p) and couple (c) laser fields interact with the 4NAs in the waveguide within an optical cavity of length \( \ell \). The signal detuning frequencies are
and the Rabi frequencies are $\Omega_{s,p,c}$ with

$$\Omega_c(x) = \Omega_c(0) \sin \frac{x}{\ell}$$

for constant Rabi-frequency coefficient $\Omega_c(0)$ and longitudinal coordinate, or position, $x$. The fields are evanescently confined to the NIMM-4NA interface with decay functions $\zeta_{c,p,s}(z)$. The decay functions are maximum at the interface, and we assume that $\zeta_c \equiv \zeta_p \approx \zeta_p$ [13].

We propose using a R6G ring dye laser as input sources parameters are also employed for the NIMM layer [10, 25]. Radiative decay is given by $\Gamma_N = 1/(U_0^2 |W|)$ if the imaginary parts of the GVD and SPM are much smaller than the real parts.

We replace

$$t - \frac{x}{v_p} \mapsto \sigma := \frac{\tau}{\tau_p}, \quad x \mapsto s := \frac{x}{L_N},$$

ignoring the atomic absorption due to EIT window. We normalized GVD and SPM according to $g_l(x) = J(x)/J_{av}$. Dynamics of the normalized SP pulse envelope $u = [\Omega_p/U_0] \exp(-\alpha x)$ follows

$$\frac{1}{\sigma} \frac{\partial u}{\partial \sigma} - \frac{g_n(x) \partial^2 u}{2} - g_S(x)|u|^2 u = 0,$$

which is a dimensionless NLSE [23].

We propose employing the spatially modulated coupling laser for SP absorption-dispersion control during its propagation, which we illustrate by plotting SP absorption and dispersion in Figs. 2(a,b), respectively. Asymmetric absorption-dispersion profiles for the position-dependent SPs are evident, and we see the formation of multiple static EIT windows in the propagation direction by coupling laser modulation. We reduce atomic absorption by adjusting the spatially modulated control field and other laser field intensities for the wavelength corresponding to the $|3\rangle \leftrightarrow |1\rangle$ atomic transition (i.e., for $\omega \approx 0$). Therefore, points in the propagation direction correspond to $\omega = 0$ for the multiple EIT windows seen in Figs. 2(a,b). These EIT windows are suitable for propagating nonlinear polaritonic waves including Akhmediev breathers and frequency combs.

We choose realistic parameters to analyze performance of this polaritonic waveguide [24]. Radiative decay is quantified by $\Gamma_R = 9$ kHz and non-radiative decay by $\Gamma_{NR} = 6$ kHz. Atomic density is $N_a = 4.7 \times 10^{18}$ cm$^{-3}$. We propose using a R6G ring dye laser as input sources with $\lambda_l \approx 606$ nm, $\Omega_p = 28$ MHz, $\Omega_c(0) = 80$ MHz, $\Delta_e = 0.07$ MHz, $\Delta_n = 0.2$ MHz, and $\Delta_p = 0$. Realistic parameters are also employed for the NIMM layer [10, 25].

We suggest two sets of positions corresponding to $\omega \approx 0$ for stable propagation of nonlinear polaritonic waves.
including Akhmediev breathers and frequency combs. (i) At positions
\[ x_j^{(a)} = (-5.68 + 2j\pi)\ell, \quad j \in \{0, 1, \ldots \}, \quad (7) \]
onlinear SPs propagate with \( v_8 \approx 2.91 \times 10^{-2}c \) within \( \Delta x \approx \ell/2 \), and
\[ K_2 = (1.45 + 0.09i) \times 10^{-15} \text{ cm}^{-1}\text{s}^2, \quad (8a) \]
\[ W = (-1.47 + 0.11i) \times 10^{-15} \text{ cm}^{-1}\text{s}^2, \quad (8b) \]
respectively, are constant with \( g_0(x) \approx g_\text{in} \approx -1.01 \). Therefore, at these specific positions SPs propagate as polaritonic Akhmediev breathers. (ii) For
\[ x_j^{(l)} = (-2.61 + 2j\pi)\ell, \quad (9) \]
GVD and SPM are position-dependent within \( \Delta x \approx \ell/2 \) so
\[ K_{2av} = (5.87 + 0.25i) \times 10^{-18} \text{ cm}^{-1}\text{s}^2, \quad (10a) \]
\[ W_{2av} = (-1.01 + 0.04i) \times 10^{-15} \text{ cm}^{-1}\text{s}^2. \quad (10b) \]
At these points, SPs propagate with weak dispersion and strong nonlinearity as efficient polaritonic-frequency combs.

Exploiting the correspondence between energy levels of the Bogoliubov spectrum \( (E_{\pm1}) \) of the uniform Bose gas with kinetic energy \( \Delta k \) and energy transference between nonlinear polaritonic modes in EIT windows, we propose generating polaritonic side-bands with modulation frequency \( \Omega \) and growth rate \( b \) and thereby realize Akhmediev breather excitation. Our analysis shows energy transfer from the zeroth order \( (l = 0) \) polaritonic wave with propagation constant \( b' \), \( \Omega_p^0(x) = \exp(i b' x) \), to the first-order side-bands \( (l = \pm 1) \) by setting \( \Delta k = \Omega \) and \( E_{\pm1} = b \).

We thereby obtain the wave with amplification factor \( b = -ib' \) according to
\[ \Omega_{\pm1}^p(x) = e^{ib}, \quad \tilde{x} := x \left[ K(\omega) + \frac{1}{2L_N} \right]. \quad (11) \]

In our scheme, a stable polaritonic Akhmediev breather propagates for \( \alpha \approx 0.25, \quad \Omega = 0.8 \) and \( b = 0.73 \) within \( \Delta x = \ell/2 \) within EIT windows such that \( \delta \omega_{\text{EIT}} \approx 30 \text{ MHz} \) as shown in Fig. 3(a).

Our waveguide serves as a fast-phase modulator according to stable polaritonic-breather propagation. To this aim, we rewrite the surface-polaritonic Akhmediev breather solution as \( \Omega_p^{\text{AB}} = |\Omega_p^{\text{AB}}| \exp \{ \text{i} \arg (\Omega_p^{\text{AB}}) \} \).

For our realistic parameters, \( \text{arg}(\Omega_p^{\text{AB}}) \approx \pi \) which is the phase shift between initial and recovered plane SP waves after breather formation. The time duration for the breather excitation-recurrence cycle in our nonlinear waveguide is \( \delta t = 12 \text{ ps} \). Therefore, our waveguide modulates polaritonic frequencies up to a few GHz and can be applied as a fast surface-polaritonic phase modulator.

We propose efficient polaritonic-frequency combs by rewriting
\[ g_t(x) = g_t^c + g_t^p(x), \quad t \in \{ D, N \}. \quad (13) \]
in terms of constant and position-dependent parts. The frequency combs can be excited at specific positions \( x_j^{(l)} \), where nonlinear SPs exhibit low GVD \( (|g_D^p(x)| \ll 1) \) and strong nonlinearity \( (|g_N^p(x)| \approx 1) \). Therefore, we neglect GVD and replace \( g_D[\partial^2 u/\partial \sigma^2] \rightarrow 0 \) \( (6) \) and assume \( g_N^c \approx -1 \). The resultant expression admits an initial SP wave with input power \( P_0 \) of the form
\[ u(x) = \sqrt{P_0} \exp \left[ -i P_0 \int dx' g_N(x') \right]. \quad (14) \]
We claim that stable propagation of nonlinear SPs in the weak-dispersion limit depends on the EIT-window widths and the normalized nonlinear coefficient $g_0^N(x)$.

We propose efficient surface-polaritonic frequency combs by SP propagation along the interface shown in Fig. 3(a) with $\delta \phi = 0.1 \pi$, $P_0 \approx 10 \ \mu W$. Then we numerically solve the NLSE together with initial condition (14) within $-3\ell < x_0^{(f)} < -2.5\ell$. We obtain a modulated EIT window, strong nonlinearity and consequently efficient polaritonic frequency combs. Specifically, for $x_0^{(f)}$ with $\Delta \omega_{\text{EIT}} = 25 \text{ MHz}$ and $Q_0 \approx 0.01$, frequency combs up to $\delta \omega_{\text{comb}} \approx 11.2 \text{ MHz}$ with stability $|\Omega_p(x)| \approx 0.87|\Omega_p(x = 0)|$ are excited. However, outside the EIT window, the generated polaritonic combs are highly unstable due to high atomic absorption.

This model allows us to develop a condition to generate efficient surface-polaritonic frequency combs via position-dependent GVD and SPM. To this aim, we consider $\Delta x = x_0 = \varepsilon$ as a small propagation length and add a perturbative term to Eq. (14) of the form

$$u(x, t) = \bigg\{ \sqrt{P_0} + \varepsilon p(x)e^{i\omega t / \tau_p} \bigg\} \times \exp \left[ -iP_0 \int_{-\ell/2}^{\ell/2} dx' g_N(x') \right].$$

We also expand the SP-wave perturbation frequency around the EIT-window centre ($\omega_s$) as a function of the relative polaritonic frequency comb mode number ($\nu$) in the presence of SPs dispersion

$$\omega = \omega_s + K_1(x)\nu + \frac{K_2(x)}{2} \nu^2 + \cdots$$

with $\{K_{i>2}\}$ related to higher-order dispersion. Efficient frequency combs are generated by suppressing higher-order dispersion [16], i.e., $|K_2(x)| \ll c|K_1(x)|^2$. Specifically, at $x_0^{(a)}$, $K_1 \approx 0.35$, $K_2 \approx 2.16 \times 10^{-10}$ and $|K_2/cK_1^2| \approx 10^{-18} \ll 1$, which yields efficient polaritonic-frequency combs as shown in Fig. 4(a).

We vary the coupling-laser intensity for experimental control of EIT-window widths, leading to efficient polaritonic frequency combs shown clearly for $g_0 = 0$ and $\delta \phi = 0.1 \pi$ with initial condition (15) solving the NLSE numerically around $x_0^{(f)}$. The number of frequency combs increases by modulating coupling-laser intensity and by engineering the EIT-window widths shown in Fig. 4(c). Comparing our frequency combs to polaritonic Akhmediev breathers reveals that nonlinear waves generated at our propose position are more efficient than frequency combs excited by Akhmediev breathers, as seen in Fig. 4(b).

We describe the excitation of nonlinear surface-polaritonic waves including polaritonic Akhmediev breather and frequency combs by employing the concept of pass-band polaritonic modulation instability. We assume the initial SPs with dispersion length $L_D \approx L_N$ according to

$$u(x, t) = u_0 e^{i(k + K_0 + 1/2L_D)x + i\omega t / \tau_p},$$

with

$$k = g_0 |u_0|^2 e^{2\text{Im}[K_0(x)]x - \omega^2 / 2 - K_0(x) - 1 / 2L_N.}$$

Moreover, we assume $\Omega'$ as a modulation frequency, $\delta = (\omega_s - \omega_0) / \omega_s$ as the normalized perturbed frequencies, and $\kappa$ as a modulation parameter in the propagation direction. We have perturbed the SP waves in terms of $p(x), q(x) \ll 1$ as

$$u(x, t) = u_0 \left[ 1 + p(x)e^{-i\Omega' (\kappa \bar{x} - \tau)} + q^*(x)e^{i\Omega' (\kappa \bar{x} - \tau)} \right].$$

We also expand the SPs linear dispersion and the nonlinear coefficient as a power series of the normalized perturbation frequency

$$K(\delta) = K_0 + K_1 \delta + \frac{K_2}{2} \delta^2 + O(\delta^3), \ g_N \approx g_0 + g_{N1} \delta, \ (20)$$

and linearize the NLSE using Eq. (17) in the weak perturbation limit [28]. The perturbed-wave dispersion relation is

$$\left[ \kappa + \bar{K}_0 + (K_1 - 1)\delta + \frac{K_2}{2} \delta^2 \right]^2 + (g_0 + g_{N1} \delta)|u_0|^2 - \frac{\Omega'^2}{4} = 0,$$

with $\bar{K}_0 = K_0 + 1 / 2L_N$. The gain map for the perturbed-polaritonic waves, shown in Fig. 4(d), demonstrates that nonlinear-polaritonic waves are excited in EIT windows with $|\delta| < \delta_{\text{EIT}}$ and $0.5 < \Omega' < 1$ corresponding to pass-band polaritonic modulation instability.

In summary, we introduce a waveguide that exploits spatial control to excite nonlinear-polaritonic waves including Akhmediev breathers and frequency combs as specific cases. We propose a stable cavity comprising 4NAs in a lossless dielectric above the NIMM layer, on which SPs propagate. The 4NA medium is driven by three co-propagating signals, a pump signal ($s$), a weak probe signal ($p$), and a standing wave coupling signal ($c$), all assumed injected from laser beams using the end-fire coupling technique. We propose stable excitation of polaritonic Akhmediev breathers and energy transfer to other polaritonic side-bands at certain position of NIMM-4NA interface by modifying laser-field intensities and detunings through GVD-SPM modulation. Moreover, we demonstrate efficient polaritonic frequency-comb generation at a specified position of the waveguide by engineering EIT-window widths and decreasing GVD commensurate with the pass-band regime for polaritonic modulation instability. Our proposed waveguide has been analyzed for experimentally feasible conditions and should act as a high-speed polaritonic phase modulator and efficient frequency-comb generator.
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