Artificial Cnoidal Wave Breathers in Optical Microresonators

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Breathers are localized structures that undergo a periodic oscillation in their duration and amplitude. Optical microresonators, benefiting from their high quality factor, provide an ideal test bench for studying the breathing phenomena. In the monochromatically pumped microresonator system, intrinsic breathing instabilities are widely observed in the form of temporal dissipative Kerr solitons which only exist in the effectively red detuned regime. Here, we proposed a novel bichromatic pumping scheme to create compulsive breathing microcombs via respectively distributing two pump lasers at the effectively blue and red detuned side of a single resonance. We experimentally discover the artificial cnoidal wave breathers and molecular crystal-like breathers in a chip-based silicon nitride microresonator, and theoretically describe their intriguing temporal dynamics based on the bichromatic pumping Lugiato-Lefever equation. In particular, the corresponding breathing microcombs exhibit diverse comb line spacing ranging from 2 to 17 times of the free spectral range of the resonator. Our discovery not only provides a simple and robust method to produce microcombs with reconfigurable comb line spacing, but also reveals a new type of breathing waves in driven dissipative nonlinear systems.

1. Introduction

Optical microresonators with high quality (Q) factor possess the ability to confine light in a small mode volume and obtain a photon lifetime up to several milliseconds, which greatly enhance the light-matter interactions,[1] thus emerging as an ideal platform for studying
nonlinear phenomenon. Over the last decade, microresonator-based Kerr frequency combs\(^{3,4}\) (“microcombs”) have set up a research upsurge in the field of Kerr nonlinear photonics and led to significant advances in a number of applications including high-capacity telecommunications\(^{5}\), spectroscopy\(^{6,7}\), ranging\(^{8-10}\), low-noise microwave/THz generation\(^{11-15}\), and optical neural network\(^{16-18}\). Since the mode-locked microcombs operated in the regime of temporal dissipative Kerr solitons (DKSs) was observed in crystalline microresonators\(^{19}\), the research focus rapidly move from modulation instability (MI) microcombs which exist in the effectively blue detuned regime\(^{20}\) to the soliton microcombs which exist in the effectively red detuned regime\(^{21}\). It was shown that different forms of DKSs, including traditional single/multiple soliton states and bound states such as soliton crystals (SCs)\(^{22,23}\) and perfect SCs (PSCs)\(^{24,25}\), exhibit a rich panel of dynamic instabilities\(^{26,27}\). Breathing state\(^{28-32}\) specifically a kind of localized patterns that undergo periodic oscillations in both duration and amplitude, is one of the most widely studied dynamic instabilities in microresonators, which can be well described by the Lugiato–Lefever equation (LLE)\(^{33,34}\). The intrinsic breathing instabilities under the monochromatic pumping scheme only exist in the detuning region between the modulation instability (MI) and stable DKS state. In the effectively blue detuned regime, the stable Turing rolls are directly connected with chaotic MI states\(^{27}\).

In addition to the common monochromatic pumping scheme, bichromatic pumping schemes have been employed as an important complement. This pumping scheme\(^{35}\) could be implemented either by launching two independent continuous wave (CW) lasers\(^{36}\) or modulating a single CW laser\(^{37}\). Bichromatic pumping scheme is previously adopted to pump two different resonances apart from each other with one or multiple free spectral ranges (FSRs) in order to generate MI microcombs with much lower pump power threshold\(^{35,38,39}\) or tunable comb line spacing (CLS)\(^{40}\). Auxiliary pump is commonly used to balance the unwanted thermal effect for reliable DKSs generation\(^{36,41}\). Recently, it was demonstrated that driving one resonance with two red-detuned monochromatic lasers could lead to the formation of
heteronuclear soliton molecules by accessing the multistability regime.\cite{42} When soliton microcombs are operated at breathing state, the introduction of a secondary pump could realize the stabilization and tuning of the breathing frequency.\cite{32}

In this work, we proposed a novel bichromatic pumping scheme where a single resonance is driven by two pump laser fields which are respectively located at the effectively blue and red detuned sides. The employment of two pumps provides additional degrees of freedom including the frequency difference and the power difference which are experimentally tunable. Even though the stronger pump is working at the effectively blue detuned side, an analogue of PSC breather\cite{24} is produced due to the existence of the red detuned pump, which features significantly larger bandwidth than monochromatically pumped Turing rolls. The microcomb states with CLSs ranging from 10 to 17 times of the FSR are observed in the experiment. Given that Turing rolls and PSCs are both periodic solutions of LLE, they are usually bracketed as cnoidal waves in nonlinear wave community,\cite{43–45} and here we refer the novel breathing states in our work as artificial cnoidal wave breathers (ACWBs). The breathing behavior of ACWBs is compulsive and the corresponding breathing frequency directly depends on the frequency difference of two pump lasers. In particular, breathing microcomb states with CLSs ranging from 2 to 9 times of the FSR referred to as molecular crystal-like breathers are also generated which are equipped with peculiar optical spectra. Verified by numerical simulations, these novel breathing states exhibit intriguing breathing behaviors which are significantly different from monochromatically pumped DKS breathers. High periodicity breathing and irregular breathing phenomena are also observed in the experiment. The demonstration of ACWBs and molecular crystal-like breathers may give some inspirations to the research of nonlinear wave dynamics, and the CLS reconfigurability may find its applications in diverse fields.

2. Bichromatic Pumping Scheme
In contrast to the common monochromatic pumping scheme\cite{19,20,46} and previous bichromatic pumping scheme working on different resonances,\cite{35,38,39} we drive a single resonance by two laser fields with distinct power, as shown in **Figure 1**. The laser fields with higher power referred to as primary pump is located at the effectively blue detuned side while the other with lower power referred to as secondary pump is located at the effectively red detuned side. The frequency difference is set to be several times larger than the half-width-half-maximum (HWHM) of the pumped resonance. Novel breathing states possessing periodic waveforms could be generated in the optical microresonator under this bichromatic pumping scheme.

**Figure 1.** An illustration of the proposed bichromatic pumping scheme. ACWB consisting of N optical pulses is breathing in the nonlinear optical microresonator. The red solid curve represents a breathing maximum while the blue dashed curve represents a breathing minimum. The corresponding averaged optical spectrum features a quasi-triangle envelope (in logarithmic scale).

Since only one resonance is pumped, the LLE model is sufficient to describe the dynamics of ACWBs by appending a secondary pump in the driving term. The bichromatic pumping LLE can be expressed as:

\[
\frac{\partial A(\phi,t)}{\partial t} = \left( -\frac{\kappa}{2} + i(2\pi \delta) \right) A + i \sum_{j=2} D_j \left( \frac{\partial}{i \partial \phi} \right)^j A - ig \left| A \right|^2 A + \sqrt{\kappa_{ex}} \left( s^{(b)} + s^{(r)} e^{2\pi i \delta t} \right) \tag{1}
\]

where \(A(\phi,t)\) represents the envelope of the intracavity field, \(\kappa\) is the cavity decay rate, \(2\pi \delta = \omega_b - \omega_p^{(b)} < 0\) is the frequency detuning of the effectively blue detuned primary pump from the pumped resonance, \(D_j\) is the \(j\)th order dispersion, \(\phi\) is the co-rotating angular coordinate which is related to the fast time coordinate \(\tau\) by \(\phi = 2\pi \tau \times \text{FSR}\).
\[ g = \frac{\hbar \alpha_0^2 c n_2}{n_0^2 V_{\text{eff}}} \] is the single photon-induced Kerr frequency shift with the refractive index \( n_0 \), the nonlinear refractive index \( n_2 \) and the effective mode volume \( V_{\text{eff}} \). \( \kappa_{\text{ex}} \) is the external coupling rate, \( \left| s^{(b,r)} \right|^2 = \frac{P^{(b,r)}_{\text{in}}}{\hbar \omega_0} \) is the driving photon flux where \( P^{(b,r)}_{\text{in}} \) respectively denote the power of the primary and secondary pump. \( 2\pi \Delta f = \omega_p^{(r)} - \omega_p^{(b)} \) is the frequency difference between two pump lasers, and \( \Delta f < \delta < 0 \) means that the secondary pump is effectively red detuned. For broader applicability of our study, we introduce the following normalized parameters for \( P^{(b,r)}_{\text{in}} \), \( \delta \) and \( \Delta f \), which are respectively defined as:

\[ f^{(b,r)} = \sqrt{\frac{8g \kappa_{\text{ex}} P^{(b,r)}_{\text{in}}}{\kappa^3 \hbar \omega_0}}, \quad \zeta_0 = \frac{4\pi \delta}{\kappa}, \quad \Delta \zeta_0 = \frac{4\pi \Delta f}{\kappa} \tag{2} \]

3. Numerical Simulation of Artificial Cnoidal Wave Breathers

Figure 2 shows the simulated dynamics of a typical ACWB state with CLS = 13 × FSR (see Supporting Animation 1 for the detailed evolution process), which is based on the realistic parameters of a silicon nitride (Si3N4) resonator used in the following experiment (see Section 4.1). The simulated intracavity power evolution and the temporal envelope evolution of the ACWB are respectively displayed in Figure 2b, c. Similar with the monochromatic pumping DKS breathers,\[29,30\] the ACWBs exhibit a periodic oscillation in their amplitude as well as a periodic compression (CP) and stretching (SP) in their duration. Intriguingly, the total intracavity power and the temporal envelope of ACWB do not share the same breathing period as shown in Figure 2a, which is different from the intrinsic breathing instabilities. After a power breathing period, the low-intensity pedestals and the high-intensity pulses exchange their positions in the co-rotating angular coordinate rather than returning to its initial state, which means that the intracavity waveform has twice the breathing period of the intracavity power (see Figure 2b, c). Figure 2d displays the corresponding optical spectrums sampled at seven moments over a power breathing period. During the process of power decline, the reduced
intensity difference between the pedestals and the pulses results in approximate frequency-doubled temporal waveform (A in Figure 2a, b) and specific instantaneous optical spectrum (A in Figure 2d) which shows complex envelope due to the interference between the ACWB pulses. The averaged spectrum features a typical triangle-shaped envelope of breathers but with some enhanced comb lines (see red arrows in Figure 3a) distinguished from the PSC breathers.[24]

We note that although it is hard to directly measure the detailed evolution of the intracavity waveforms as shown in Figure 2c in the experiment, these enhanced comb lines are important characteristics of ACWB’s averaged optical spectrum acquired by the optical spectrum analyzer.

Figure 2. Numerical simulations of a typical ACWB state with CLS=13×FSR under $f_1^2 = 13.5$, $f_2^2 = 3$, $\zeta_0 = -0.8$ and $\Delta \zeta_0 = -6.8$. a) Intracavity intensity patterns of ACWB sample at successive breathing minimums (SP1 and SP2), breathing maximums (CP1 and CP2) and an approximate frequency-doubled moment (A) depicted in Figure 2b. b) Evolution of the intracavity power over 10 power breathing periods. c) Evolution of the intracavity pulse waveform. d) Optical spectrums of ACWB sampled at seven moments over a power breathing period. The averaged spectrum over one period features a quasi-triangle envelope (in logarithmic scale) with some enhanced comb lines.

Traditional DKS breathers in microresonators exhibit periodic energy exchange between comb lines around the center and the wings, which is related to the Fermi-Pasta-Ulam recurrence.[28] Different from the DKS breathers, the ACWB microcombs show almost in-phase
oscillation as displayed in Figure 3b, indicating that the breathing phenomenon is directly caused by the periodically oscillating pump power under our bichromatic pumping scheme. Thus, this kind of breathing state could be referred to as the artificial breathing state distinguished from the intrinsic breathing state. Another important characteristic of ACWB microcombs is that the enhanced comb lines exhibit much smaller breathing depth which is defined as $(P_{\text{max}} - P_{\text{min}}) / (P_{\text{max}} + P_{\text{min}})$ with $P_{\text{max(min)}}$ being the maximum (minimum) power of each comb line, resulting from the specific evolution dynamics of the intracavity waveform. For example, as shown in Figure 3a, the comb lines with mode index $\mu = \pm 13$ possess almost the same averaged power as comb lines with mode index $\mu = \pm 26$, which makes it convenient to directly compare their breathing depth. The simulated breathing depth of mode $\mu = 26$ is around five times smaller than mode $\mu = 13$ (see Figure 3c).

Figure 3. a) Simulated averaged optical spectrum of the ACWB state with CLS=13×FSR. The red dashed lines indicate the triangle envelope of the spectrum, and the enhanced comb lines are marked by the red arrows. b) Simulated power evolution of the comb lines around the center ($P_{\text{center}}$) and wings ($P_{\text{wings}}$) as depicted in (a) ($P_{\text{center}}$ and $P_{\text{wings}}$ are scaled to the same range). c)
Simulated power evolution of the comb lines with mode index $\mu = 13$ and $\mu = 26$, and the numbers in brackets denote the corresponding breathing depth.

4. Experimental Generation of Artificial Cnoidal Wave Breathers

4.1. Typical Artificial Cnoidal Wave Breathers

Figure 4a illustrates our experimental setup. A Si$_3$N$_4$ microring resonator with the cross-section of $1550 \times 800$ nm$^2$ and the radius of 240 $\mu$m is utilized for ACWBs generation. Figure 4b shows the measured laser-scanned transmission spectrum of the pumped resonance which exhibits a loaded optical quality (Q) factor of 1.2 million. The dispersion characteristic of the microresonator shown in Figure 3c is measured through the Mach-Zehnder interferometer (MZI)-based optical sampling technique, and the measured dispersion matches well with finite-difference time-domain (FDTD) simulated result, indicating an anomalous dispersion of $D_2 / 2\pi \sim 1.35$ MHz. In order to achieve bichromatic pumping with flexibly controlled power and frequency difference, a dual-parallel Mach-Zehnder modulator operating in the carrier-suppressed single sideband (CS-SSB) mode is employed to generate two laser fields via simultaneously modulating the continuous wave (CW) laser by two microwave signals from broadband voltage-controlled oscillators (VCOs). The two laser fields are then amplified by an erbium-doped fiber amplifier (EDFA) and then coupled to the optical microresonator via a lensed fiber.
Figure 4. a) Schematic of the experimental setup. The inset shows the scanning electron microscopy image of a Si$_3$N$_4$ microresonator with the radius of 240 μm. CW Laser: continuous-wave laser; DP-MZI: dual-parallel Mach-Zehnder interferometer; VCO: voltage-controlled oscillator; AWG: arbitrary waveform generator; EDFA: erbium-doped fiber amplifier; FPC: fiber polarization controller; EC: electric coupler; OC: optical coupler; OSA: optical spectrum analyzer; OSC: oscilloscope; ESA: electronic spectrum analyzer. b) Measured transmission spectrum (blue circles) of the pumped resonance centered around 1564 nm. The Lorentz fitting curve (red solid line) indicates a loaded quality factor ($Q_{\text{load}}$) of $1.2 \times 10^6$. c) The measured (blue circles) and FDTD simulated (red solid line) integrated dispersion ($D_{\text{int}} = \omega_{\mu} - \omega_0 - \mu D_{\text{FSR}}$) of the fundamental quasi-TE mode as a function of relative mode number $\mu$ ($\mu = 0$ is at 1564 nm). Here, $\omega_0$ is the angular frequency of the pumped mode, $\omega_{\mu}$ is the angular frequency of the $\mu$th cavity mode relative to $\omega_0$, and $D_{\text{FSR}}$ is the FSR measured at $\omega_0$. The inset shows the simulated mode profile of the fundamental TE$_{00}$ mode.

We design a two-step tuning method for reliably accessing the ACWB states as shown in Figure 5a. Firstly, the primary and secondary pump are separated far from each other and both located at the effectively blue detuned side. By manually scanning the CW laser, the two laser fields are simultaneously tuned towards the resonance from the short wavelength, which is referred to as “forward tuning” (stage I in Figure 3a). The secondary pump then scans into the resonance, exciting the microcombs and red shifting the resonance due to the strong thermal
effect (stage II in Figure 3a). The resonance shifts back after the secondary pump crossing the effectively zero detuned point, and then thermally locked to the high power primary pump in the effectively blue detuned regime\textsuperscript{[47]} thanks to the large frequency difference between the two pumps, accompanied by the emergency of chaotic microcombs (stage III in Figure 3a). Afterwards, we tune the secondary pump backwards from the red detuned side of the resonance by changing the voltage applied to the VCO. ACWBs could be generated at appropriate detune value (stage IV in Figure 3a). Although the frequency detuning is considered unstable when the pump is working at the red detuned regime,\textsuperscript{[47]} the stronger primary pump at the blue detuned side stabilizes the resonance and makes it possible to freely tune the secondary pump at the red detuned side without greatly impacting the resonance. A numerical simulation of this tuning process could be seen in the Supporting information.

Figure 5. a) An illustration of the two-step tuning method for ACWBs generation. b) Three optical spectrums sampled during the two-step tuning process as depicted in (a). II: Primary combs generated by the secondary pump; III: Chaotic combs generated by the primary pump; IV: ACBW microcombs with $\text{CLS}=13 \times FSR$. 
Taking the generation of an ACWB state with CLS=13×FSR as an example, we launch a total pump power of 26.5 dBm onto the coupled waveguide of the Si3N4 microring resonator with a power difference between the primary and secondary pump around 6.5 dB. While the two laser fields with an initial frequency difference approximately 1.5 GHz are simultaneously scanned from blue to red, the secondary pump firstly couples into the resonance and excites the microcombs (II in Figure 4b). After the secondary pump crosses the resonance and reaches the red detuned regime, the resonance shifts backwards due to the thermal instability and is blocked by the primary pump. We further tune the primary pump into the resonance and relatively noisy microcombs are excited as shown in Figure 4b (subgraph III). Then the secondary pump is tuned backwards while fixing the primary pump until the frequency difference is close to about 500 MHz. At this time, ACWB microcomb is generated after the single-FSR components between the supermodes with CLS=13×FSR decaying rapidly (see IV in Figure 4b). The detailed optical spectrum is displayed in Figure 6a, which shows a quasi-triangle envelope with enhanced comb lines at mode index $\mu = \pm 26$. In order to further characterize the breathing behavior of the ACWB, the comb lines with mode index $\mu = -13$ and $\mu = -26$ are respectively filtered out and then detected by fast photodetectors (PDs). The power evolution traces of two comb lines recorded by the oscilloscope are shown in Figure 6c. The breathing depth of the comb mode $\mu = -26$ is much smaller than the comb mode $\mu = -13$, although they have almost the same averaged power. The enhanced comb lines and the difference of the breathing depth agrees well with the simulated result in Section 3, which corroborates the intriguing breathing dynamics of the intracavity waveform as shown in Figure 2c. Figure 6b shows the corresponding RF spectrum of the ACWB microcomb, where the low noise spectrum background indicates the high coherence of the ACWB state (see Supporting information for more coherence characterization). We note that the poor linewidth of the breathing frequency
results from the high phase noise of the broadband VCO controlled by the AWG, and a pure breathing frequency could be expected when using a stable modulation source.

![Figure 6](image)

**Figure 6.** a) Optical spectrum of the ACWB microcomb with CLS=13×FSR. The red dashed lines indicate the triangle envelope of the spectrum, and the enhanced comb lines are marked by the red arrows. b) The corresponding RF spectrum (blue trace) of the ACWB in (a) and the PD noise floor (red trace). Resolution bandwidth (RBW) of the measured traces is 200 Hz. c) Recorded fast power evolution of the filtered comb mode $\mu = -13$ and mode $\mu = -26$.

Moreover, ACWB states with different CLS could be generated when changing the total pump power, frequency difference, power difference and the relative pump-resonance frequency detuning of the two laser fields. The ACWB microcombs with CLS ranging from 10 to 17 times of the FSR are observed in the same microresonator by bichromatically pumping the same resonance, as shown in **Figure 7**. The transition between these different ACWB states
are intermediated by chaotic microcombs. Those ACWB spectra are all characterized by the typical quasi-triangle envelopes with two enhanced comb lines (see Figure 7), which is consistent with the simulated characteristics. In some cases, two ACWB states could be switched between each other by simply change the relative pump-resonance frequency detuning without modifying other parameters of the two pump lasers (i.e. only need to tune the wavelength of the CW laser), such as the switching from the 12-FSR CLS state to the 13-FSR CLS state (see Figure 7). Different from the Turing rolls (or primary combs) whose CLSs are determined by the well-defined parametric gain\cite{20,27,44} or the perfect soliton crystals (PSCs) whose CLSs are determined by the spectral position of the avoided mode crossings (AMXs),\cite{22,24,25,25} we have not yet summed up the general rules for generating the ACWB microcomb with a specific CLS. From the perspective of LLE simulations, we find that ACWB states with several different CLSs could be generated under exactly the same pump parameters with different initial states. For example, ACWB states with CLSs ranging from 13 to 17 times of the FSR could be generated under the same parameters used in Figure 2 by only changing the initial state of the simulation.

Figure 7. Optical spectra of the ACWB microcombs with different CLSs ranging from 10 to 17 times of the FSR. The red dashed lines indicate the triangle envelope of the spectrum, and the enhanced comb lines are marked by the red arrows. The pumped mode is around 1564 nm for all microcombs. The parameters (total on-chip power, power difference, frequency difference) of the two pump lasers are set as (26.3 dBm, 5.6 dB, 530 MHz), (26.8 dBm, 5.6 dB, 535 MHz), (26.5 dBm, 6.5 dB, 520 MHz), (26.5 dBm, 6.5 dB, 520 MHz), (28.4 dBm, 6.4 dB,
We note that the generated ACWB states are very robust, which could exist for hours in our free-running system even with poor amplitude and frequency noise originating from the broadband VCOs driven by the AWG. This robustness could be attributed to the strong primary pump which is working at the effectively blue detuned regime and thermally locking the resonance. Moreover, the frequency difference of the two pump lasers need not to be at a fixed value for maintaining an ACWB state, and could be tuned for tens of megahertz (MHz) without losing the specific ACWB.

4.2. Molecular Crystal-like Breathers

Except for typical ACWB microcombs, novel breathing states with CLSs ranging from 2 to 9 times of the FSR are also generated when we continuously scan the two pump lasers in their power-detuning phase plane. Some of the experimentally measured optical spectra are displayed in Figure 8 (see Supporting information for more optical spectra under different pump parameters).

![Figure 8](image)

**Figure 8.** Optical spectra of the molecular crystal-like breathers with different CLS ranging from 2 to 9 times of the FSR. The parameters (total on-chip power, power difference, frequency difference) of the two pump lasers are set as (28 dBm, 5.6 dB, 484 MHz), (27 dBm, 6.2 dB, 493 MHz), (28 dBm, 6.5 dB, 531 MHz), (26 dBm, 6.3 dB, 500 MHz), (29 dBm, 7 dB, 630 MHz), (25 dBm, 5.7 dB, 430 MHz), (26.5 dBm, 6 dB, 478 MHz), and (27 dBm, 6.5 dB, 530 MHz), respectively.
These novel breathing microcombs feature patterned spectral envelopes which indicate that complex periodical waveforms are generated in the microresonator. Figure 9b shows the simulated averaged spectrum of the breathing state with CLS=6×FSR, which shows remarkable agreement with the experimentally measured result (see Figure 9a). The corresponding intracavity waveform evolution process is displayed in Figure 9c (see Supporting Animation 2 for detail evolution process), which shows significantly different dynamics with typical ACWB states. As shown in Figure 9d, there are several pulses oscillating as a group within one period of the intracavity waveform (see dashed region in Figure 9d). The patterned envelope of the optical spectrum is exactly resulted from the interference between these multiple pulses. Here, we name these novel breathing states as “molecular crystal-like breathers” inspired by the terminologies “soliton molecules”\cite{42} and “soliton crystals”.\cite{22} It should be noted that not all of the experimentally measured optical spectra displayed in Figure 8 could be well reproduced in the general LLE simulations (Equation 1). We surmise that the wavelength-dependent quality factor, avoided mode crossings and high-order dispersion will affect the specific intracavity waveforms and the corresponding optical spectra.
Figure 9. a) Experimentally measured optical spectrum of a molecular crystal-like breather state with CLS=6×FSR. The total on-chip power, power difference and frequency difference of the two pump lasers are set as 29 dBm, 7 dB and 630 MHz, respectively. b) Simulated averaged optical spectrum of a molecular crystal-like breather state with CLS=6×FSR. c) Evolution of simulated intracavity pulse waveform. d) Simulated intracavity pulse waveform sampled at two breathing maxima depicted in (c) by the white dashed lines. The shaded regions in (d) indicate one period of the corresponding waveforms. For all the simulations, the parameters are set as $f_1^2 = 20$, $f_2^2 = 4$, $\zeta_0 = -2$ and $\Delta \zeta_0 = -10$, respectively.

4.3. High Periodicity Breathing and Irregular Breathing

Similar with the intrinsic breathing instability of monochromatically pumped DKS breathers,[26,30] we also observed high periodicity breathing and irregular breathing phenomena in the ACWB and molecular crystal-like breather states. Some experimental results are displayed in Figure 10. The period-2 breathing, period-4 breathing and irregular breathing phenomena are discovered in the microcomb states with 9-FSR, 16-FSR and 4-FSR CLSs respectively, which are characterized by the recorded frequency-domain RF spectra and time-domain power evolution of the corresponding microcombs (see Figure 10d-f). Experimentally, the high periodicity breathing and irregular breathing phenomena tend to appear when the primary pump is tuned further into the chaotic regime (i.e. increasing the power or decreasing the frequency detuning of the primary pump). We find that the period-2 breathing phenomenon is frequently observed in the molecular crystal-like breathers but relatively rarely found in typical ACWB states, which could be attributed to the special evolution dynamics of the intracavity waveform as shown in Figure 9d. In most of the ACWB states, tuning the pump parameters sometimes directly led to chaotic states without the appearance of the high periodicity breathing or irregular breathing. Numerically, these states can be well reproduced in the bichromatic pumping LLE simulations (see Supporting information for details).
Figure 10. High periodicity and irregular breathing phenomena. (a-c) Optical spectra of the microcombs with CLS equal to 9×FSR, 16×FSR and 4×FSR, respectively. (d-f) The RF spectra of the microcomb states in (a-c). The insets show the corresponding time-domain power evolution recorded by a fast photodetector. (d), (e) and (f) shows the period-2, period-4 and irregular breathing phenomena, respectively. The RBW of the measured traces is 100 kHz. The frequency difference is marked by the red arrow.

5. Conclusion

In conclusion, we have proposed a novel bichromatic pumping scheme and discovered the artificial cnoidal wave breathers and the molecular crystal-like breathers in a Si3N4 microresonator. The unique pumping scheme bridges the gap between the effectively blue and red detuned regime of a single resonance and excites a compulsive breathing behavior of microcombs. In particular, the ACWB states with CLSs ranging from 10 to 17 times of the FSR and the molecular crystal-like breathers with CLS ranging from 2 to 9 times of the FSR are produced by pumping exactly the same resonance of a single resonator, which indicates the remarkable flexibility of this pumping scheme for generating microcombs with reconfigurable
Moreover, the high periodicity breathing and irregular breathing phenomena were also observed in our experiment. Numerical simulation results agree well with the experiment results and reveal the intriguing breathing dynamics of the intracavity waveform which are significantly different from the intrinsic breathing instabilities of monochromatically pumped DKS breathers. On the one hand, the mechanism of these novel breathing phenomena is universal for any kinds of Kerr nonlinear resonators pumped bichromatically, which may broaden the research scope of nonlinear wave dynamics both theoretically and experimentally. On the other hand, the discovery of the ACWBs and molecular crystal-like breathers will help to develop reconfigurable microcombs which could meet different demands of applications such as optical communication, spectroscopy and photonic microwave/THz generation.

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Conflict of Interest
The authors declare no conflict of interest.

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Abstract

This supplementary material includes the bichromatic pumping LLE simulation details, other molecular crystal-like breathers generated in the Si$_3$N$_4$ microresonator, the tunability of the breathing frequency, the coherence characterization of the ACWB microcombs and the illustration of the animations.
I. Bichromatic pumping LLE simulations

Pump parameters setting strategy

In order to intuitively illustrate the general methods for setting the parameters of the primary and secondary pumps in the bichromatic pumping LLE simulations, we employ the stability chart (i.e. power-detuning phase plane)\(^1\) of the traditional monochromatic pumping LLE as a reference which is shown in Figure S1. According to a large number of simulation examples, we find that, in order to produce the ACWBs or the molecular crystal-like breathers, the primary pump should be located in the Turing rolls region or the edge of the chaotic MI region which is roughly indicated by the blue shaded region in Figure S1, while the secondary pump should be located below the DKS region which is roughly indicated by the red shaded region in Figure S1. The high periodicity and irregular breathing phenomena tend to happen when the primary pump get closer to the resonance. If the primary pump goes deep into the chaotic region or the secondary pump goes into the DKS region, the generated microcombs will evolve to obviously chaotic states.

Since the primary pump is located in the blue detuned regime where modulation instability exists, biochromatic pumping LLE simulations could be initiated with random noise. But it should be noted that the eventual breathing state is sensitive to the initial state, and initiating with periodical pulses will be helpful for finding the ACWB and the molecular crystal-like breather state with a specific CLS.
Figure S1. Probable parameters of the primary and secondary pump lasers illustrated in the stability chart of the monochromatic pumping LLE. The probable parameter regions for the primary pump and secondary pump are respectively shaded with blue and red. Note: the regions of DKS breather, transient chaos and spatiotemporal chaos are not displayed (see the reference[2] for detailed description).

Simulated two-step tuning process

Figure S2 shows the numerical simulated two-step tuning process for generating a 14-FSR CLS ACWB microcomb. Thermal effect is ignored in the simulation considering that the resonance is thermally locked by the blue detuned primary pump. Firstly, the pump parameters are set as $f_1^2 = 13.4$, $f_2^2 = 4.3$, $\xi_0 = -12$ and $\Delta \xi_0 = -10.5$. $\xi_0$ is tuned from -12 to -0.1 (“forward tuning”) in 375000 roundtrips initiated with one photon per mode noise (512 modes are considered). The secondary pump scans through the resonance from blue to red while the primary pump reaches near the resonance at the effectively blue detuned regime which excites chaotic microcombs. Then $\Delta \xi_0$ is tuned from -10.5 to -8.1 (“backward tuning”) in 250000 roundtrips. During the backward tuning process, the ACWB state with CLS=14×FSR is condensed from the chaotic pulses within an appropriate range of frequency difference $\Delta \xi_0$ (see blue shaded region in Figure S2a).
Figure S2. Simulated two-step tuning process. (a) Envelope of the intracavity power evolution trace. The Roman numbers are corresponded with four stages shown in Figure 5a in the main text. The black dashed line separates the forward and backward tuning process. (b) Evolution of the intracavity pulse waveform. The inset shows local details. (c) Evolution of the intracavity optical spectrum.

**High periodicity and irregular breathing**

Here we verify that the high periodicity breathing and irregular breathing phenomena could also be reproduced in the bichromatic pumping LLE simulations, as shown in Figure S3.
**Figure S3.** Simulated high periodicity breathing and irregular breathing phenomena. (a) Period-2 breathing state for a 9-FSR CLS state under $f_1^2 = 20$, $f_2^2 = 3.5$, $\zeta_0 = -2$ and $\Delta \zeta_0 = -10$ (see Supporting Animation 4). (b) Period-4 breathing state for a 16-FSR CLS state under $f_1^2 = 14$, $f_2^2 = 2.8$, $\zeta_0 = -0.1$ and $\Delta \zeta_0 = -6.1$ (see Supporting Animation 5). (c) Irregular breathing state for a 4-FSR CLS state under $f_1^2 = 21$, $f_2^2 = 4$, $\zeta_0 = -2.1$ and $\Delta \zeta_0 = -9.7$ (see Supporting Animation 6). In (a-c), top left: averaged optical spectrum; bottom left: RF spectrum of the corresponding microcomb over 200000 roundtrips (the equivalent resolution bandwidth is 500 kHz); top right: evolution of the total intracavity power; bottom right: evolution of the intracavity waveform.

**II. Other molecular crystal-like breathers**

**Figure S4** shows the optical spectra of other molecular crystal-like breathers generated in the experiment. The diverse spectrum envelopes of these microcombs indicate that a large variety of molecular crystal-like breathing pulses could be generated in the microresonators under the bichromatic pumping scheme. In other words, the novel bichromatic pumping method is very powerful and flexible for generating intriguing periodical optical pulses in the microresonators.

**Figure S4.** Experimentally measured optical spectra of other molecular crystal-like breathers. The parameters (total on-chip power, power difference, frequency difference) of the two pump lasers are set as (27 dBm, 6 dB, 450 MHz), (29.3 dBm, 7 dB, 596 MHz), (26.6 dBm, 5.7 dB, 473 MHz), (27 dBm, 6 dB, 503 MHz), (28.4 dBm, 6.5 dB, 541 MHz), and (25.6 dBm, 6 dB, 453 MHz), respectively.

**III. Tunability of the breathing frequency**

We also observed that the generated breathing microcombs can maintain in a certain range of frequency difference in the experiment. Taking a period-2 breathing state with CLS=2×FSR (see **Figure S5**) as an example, we tune the frequency of the secondary pump while keeping
the primary pump fixed. The period-2 breathing state maintains while the frequency difference is tuned from 556 MHz to 602 MHz.

Figure S5. (a) Optical spectrum of a period-2 breathing molecular crystal-like breathing state with CLS=2×FSR. (b) Evolution of the RF spectra during the tuning process. (c) RF spectra sampled at three different stages as depicted in (b). The resolution bandwidth of the measured traces is 100 kHz.

IV. Coherence characterization

In order to characterize the coherence of the artificial cnoidal wave breathers (ACWB), we employed an external CW laser (CoBrite DX4 DFB) to heterodyne with a comb line of the ACWB microcomb and then measured the beat note by electronic spectrum analyzer (R&S FSWP). We took an ACWB state with CLS=14×FSR as an example as shown in Figure S6. The heterodyne signal marked by the red arrow possesses a high signal-to-noise ratio of above 34 dB, clearly indicating high coherence of the ACWB state. The spurs around heterodyne signal exactly originate from the breathing behavior of the comb line.

The spectral purity of the ACWBs repetition frequency is another important parameter to characterize the coherence. However, direct measurement of the repetition frequency beat note is unavailable in our lab due to the large FSR (~100 GHz) of our Si$_3$N$_4$ microresonator.
Figure S6. (a) Optical spectrum of an ACWB state with CLS=14×FSR combined with a CoBrite DX laser. The inset shows the local details of the spectrum. (b) Top: RF spectrum of the ACWB. Bottom: RF spectrum of the heterodyne signal. The black arrows indicate the frequency difference (around 640 MHz) of the primary and secondary pump. The red arrow indicates the heterodyne frequency (around 7.5 GHz).

V. Illustration of the animations

The animations show the dynamics of some typical ACWB and molecular crystal-like breathers. In each animation, top: evolution of the total intracavity power; middle: evolution of the intracavity waveform; bottom: evolution of the optical spectrum.

**Animation 1**, an ACWB state with CLS=13×FSR state under $f_1^2 = 13.5$, $f_2^2 = 3$, $\zeta_0 = -0.8$ and $\Delta\zeta_0 = -6.8$.

**Animation 2**, a molecular crystal-like breather state with CLS=6×FSR under $f_1^2 = 20$, $f_2^2 = 4$, $\zeta_0 = -2$ and $\Delta\zeta_0 = -10$.

**Animation 3**, a molecular crystal-like breather state with CLS=3×FSR under $f_1^2 = 8$, $f_2^2 = 2$, $\zeta_0 = -1$ and $\Delta\zeta_0 = -6$.

**Animation 4**, period-2 breathing state for a 9-FSR CLS state under $f_1^2 = 20$, $f_2^2 = 3.5$, $\zeta_0 = -2$ and $\Delta\zeta_0 = -10$.

**Animation 5**, period-4 breathing state for a 16-FSR CLS state under $f_1^2 = 14$, $f_2^2 = 2.8$, $\zeta_0 = -0.1$ and $\Delta\zeta_0 = -6.1$.

**Animation 6**, irregular breathing state for a 4-FSR CLS state under $f_1^2 = 21$, $f_2^2 = 4$, $\zeta_0 = -2.1$ and $\Delta\zeta_0 = -9.7$.
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