Chaotic Internal Dynamics of Dissipative Optical Soliton Molecules

Youjian Song, Defeng Zou, Omri Gat,* Minglie Hu,* and Philippe Grelu*

When a laser cavity supports the propagation of several ultrashort pulses, these pulses can form compact bound states called soliton molecules. Soliton molecules are fascinating objects of nonlinear science, presenting striking analogies with their matter molecule counterparts. The relative timing and phase between the copropagating pulses are the most salient internal degrees of freedom of the soliton molecule. The soliton pair, composed of two identical pulses, constitutes the chief soliton molecule of fundamental interest. Its two major internal degrees of freedom allow self-oscillating soliton molecules, which have been recurrently observed. However, despite theoretical predictions, the low-dimensional chaotic dynamics of a soliton-pair molecule remain elusive. This article reports the observation of chaotic soliton-pair molecules within an ultrafast fiber laser by means of a direct measurement of the relative optical pulse separation with sub-femtosecond precision in real time. Moreover, it demonstrates an all-optical control of the chaotic dynamics followed by the soliton molecule by injection of a modulated optical signal that resynchronizes the internal periodic vibration of the soliton molecule. The fast error-free switching between ordered and chaotic soliton molecules enabled by pump current sweeping and external injection highlights the potential prospects of all-optical logic gates and chaotic communication using soliton molecules.

1. Introduction

Chaotic dynamics are characterized by aperiodicity, high sensitivity to initial conditions, and long-term unpredictability. These are intrinsic features and behaviors observable in most nonlinear systems. The manifestation of chaos in extended natural systems raises awe and fear, from the uncertain evolution of celestial bodies to the weather instability, respectively highlighted in the seminal works of Poincaré and Lorenz.[1,2] Therefore, over the past century, there has been a high impetus to investigate chaos within laboratory-confined systems over a wide range of scientific fields, from hydrodynamics and electromagnetics to quantum physics and optics.[3] Studying chaos in nonlinear optics is of particular interest since photonics exhibits handy systems such as the laser oscillator that can advance the fundamental understanding of chaotic dynamics and utilize chaos in technological innovations such as secure optical communications and Lidar sensing in automotive environments.[4,5] Alternatively, understanding the early warning signals signaling the onset of chaos helps circumvent its adverse manifestations in laser applications requiring high stability.[6–8]

The ultrafast laser constitutes an outstanding system for the investigation of chaos, being inherently wideband, highly nonlinear, and offering multiple parameter control schemes. Furthermore, the commonly accepted “dissipative soliton” paradigm explicates the great diversity of ultrashort laser pulse dynamics, which spans from stationary to chaotic pulse emission. Dissipative solitons are attracting states that result from the composite balance between dissipative and dispersive propagation effects and are potentially subjected to a great variety of bifurcations.[9] The standard mode-locked laser regime features a single laser pulse that retakes the same evolution along successive cavity roundtrips; this results from the existence of a stable focus attractor providing long term stability and robustness. Within the parameter space of the optical cavity, the laser can undergo abrupt bifurcations leading to chaotic pulse
Vivid illustrations of these chaotic dynamics include “soliton explosions,” which manifest as intermittent chaos in the pulse evolution over numerous cavity roundtrips, and “noise-like” pulse dynamics, characterized by a persisting noisy optical waveform and a complete loss of mutual coherence between successive roundtrips. Soliton explosions and noise-like pulsing belong to the category of “incoherent dissipative solitons,” which involves chaotic attractors to solve the apparent paradox of an average temporal localization for the pulse and its high instability. We emphasize that these chaotic solitons evolve around attractors of extended dynamical systems featuring a high dimensionality, with similar possibilities within Kerr microresonators, noting that the existence of low-dimensional chaotic soliton dynamics has been recently reported.

Other abrupt bifurcations may result from the laser multipulsing instability (MPI). Typically, when the pump power is augmented, the MPI leads to an increased number of intracavity pulses. Therefore, crossing the MPI with subsequent complex bifurcations leads to a virtually unlimited landscape of complex and chaotic ultrafast multi-pulse dynamics. The interaction between dissipative solitons is also known to lead to the spontaneous formation of compact bound states, also known as dissipative soliton molecules, since they display striking analogies with their matter molecule counterparts. Akin to the hydrogen molecule, the soliton-pair molecule is made of two identical constituents: it is the chief soliton molecule of fundamental interest, and the focus of our present investigations. Once formed, a stable optical soliton molecule can propagate indefinitely within the laser cavity. For a soliton molecule being stationary in its moving frame, the relative temporal separation and phase between the soliton constituents remain constant. Stationary soliton molecules are routinely generated in laser oscillators and can be represented as point attractors in an infinite-dimensional phase space. However, upon a change in the laser parameters, a Hopf-type bifurcation can take place and generate pulsating soliton molecules, represented as limit-cycle attractors, see Figure 1c.

These self-excited oscillations with time-varying properties can manifest in various ways, encompassing vibrating soliton molecules – where the soliton separation oscillates as well as phase-only oscillations. Recent laser experiments have implemented strategies of external periodic perturbation to probe and even induce transitions within soliton molecules, bringing in a close analogy to the approach of molecular spectroscopy. For instance, the resonant excitation of optical soliton molecules within a Kerr lens mode-locked Ti:sapphire laser probed the intramolecular interaction to reveal its anharmonic feature. The internal oscillatory dynamics of optical soliton molecules have been experimentally synchronized with a modulated signal injected into an ultrafast fiber laser. These experiments strengthen the
hypothesis that the individual solitons composing a multisoliton waveform are often governed by low-dimensional dynamical systems. Since optical solitons break phase symmetry as well as time-translation symmetry, the internal dynamics of a soliton-pair molecule has at least two degrees of freedom, which is enough for self-oscillations but not for chaos. As a matter of fact, there has not been any observation to date of low-dimensional chaos concerning the internal motions within soliton molecules, either in theory or in experiment. The seminal theoretical predictions of chaotic soliton-pair molecules, based on the complex Ginzburg–Landau equation, needed either the addition of an external drive\cite{35} or a soliton shape instability.\cite{25}

The motivation for discovering low-dimensional chaotic soliton molecules is twofold. It will strengthen the analogy with the molecules of matter, which are indeed subjected to nonlinear and chaotic vibrations.\cite{36} In addition, it will stimulate the development of effective low-dimensional interaction models applicable within a given range of system parameters inside otherwise highly multi-dimensional complex systems, improving the prospects of analysis and control of optical soliton molecules.

Hence, in this article, we demonstrate the excitation and all-optical control of a chaotic soliton-pair molecule within a passively mode-locked fiber laser. Such experiment requires an extremely well-resolved recording of the relative soliton separation in real-time, which is achieved with the balanced optical cross-correlation (BOC) technique.\cite{37} Indeed, BOC tracks the pulse separation variations within a vibrating soliton molecule with sub-femtosecond resolution, enabling the observation of its transitions to chaotic dynamics for the first time, to the best of our knowledge. The route through period doubling bifurcations to chaos is explicitly retrieved with a perfect reproducibility. The chaotic dynamics are qualitatively characterized by the direct BOC waveform, its radiofrequency (RF) spectrum, and phase portraits analysis. They are also quantitatively analyzed using the Lyapunov exponent and the correlation dimension analysis. Finally, we achieve an all-optical control of the chaotic dynamics followed by the soliton molecule, by means of a weak external signal injection, without altering the central laser parameters. The experimental observations are qualitatively well reproduced by numerical simulations.

2. Experimental Results on Chaotic Soliton Molecules

2.1. Monitoring the Extension of the Soliton Molecule in Real Time

Figure 1 displays the concept and implementation of the characterization of dissipative optical soliton molecules. These molecules are generated within a fiber ring laser cavity, which comprises an erbium-doped fiber for laser emission around the 1.5-micron telecom wavelength, and an effective and controllable ultrafast saturable absorber based on the nonlinear polarization evolution (NPE) that takes place in the single-mode optical fibers (see Experimental Section). Upon the adjustment of the pump power and the intracavity retarding waveplates, the laser can self-generate soliton-pair molecules in various dynamical states. The output soliton-pair molecules are beam-split into two branches and recombined with orthogonal polarization states using a Michelson interferometer, as sketched in Figure 1a. We then detect the real-time dynamics of the variations of the pulse separation within the soliton molecule by means of the BOC measurement technique. When the overlap between the two arms is scanned, the BOC outputs an S-shaped voltage signal (see Figure 1b). We set the zero crossing of the S-shaped curve as the working point, around which the voltage excursions of the BOC output are proportional to the variations of the pulse separation within the soliton molecule, whereas the laser intensity noise is canceled out by the balanced photodetection (more details about the BOC measurement technique are provided in Section A, Supporting Information).

The recording of the BOC voltage signal allows true real-time monitoring of the pulse separation at the sub-femtosecond level without relying on any heavy data post-processing. Supported by a spectral analysis, such monitoring facilitates a comprehensive exploration of complex and low-amplitude internal soliton molecule dynamics. In the case of stationary soliton molecules, the BOC output displays a flat curve with small random fluctuations, whereas for vibrating molecules, periodic trajectories are observed. We anticipate that a breakdown of the periodicity of the time-domain BOC signal can be, in case significant excursions of the BOC signal remain, a signature of chaotic dynamics within the soliton molecule (see Figure 1c).

2.2. Generating Chaotic Soliton Molecules

We have developed a controllable way to excite soliton molecules through a cascade of period-doubling bifurcations leading to chaos. Figure 2a is an extensive recording of the evolution of the RF spectrogram of the BOC output when the pump strength is gradually increased. As a starting point, we set a fundamental period-one (P1) oscillating soliton molecule. An example is shown in the lower part of Figure 2b, featuring quasi-sinusoidal oscillations of the pulse separation. At a pump driving current \( I = 509 \text{ mA} \) (optical pump power \( 305 \text{ mW} \)), the oscillation of the BOC output signal corresponds to an intramolecular vibration between the two solitons having an amplitude of \( \pm 80 \text{ fs} \). The oscillation frequency \( f_1 \) is measured from the corresponding RF spectrum displayed in Figure 2c, featuring a sharp peak located at 2.36 MHz accompanied by its second harmonic. The electronic noise floor is also displayed in Figure 2c (gray curve). Accordingly, the oscillation period corresponds to about 19 cavity roundtrips. We note that the oscillation period is not exactly an integer multiple of the roundtrip time, the latter phenomenon being called subharmonic entrainment and requiring a careful search of specific system parameters.\cite{15,38,39}

When the pump strength is increased, the P1 branch first maintains its stability with a slightly varying fundamental frequency, until a new set of spectral peaks appear at odd multiples of \( f_{1}/2 \) in the RF spectrogram (Figure 2a), signaling a period-doubling bifurcation to a new family of period-doubled (P2) states. This bifurcation manifests in the vibration observed in the time domain (Figure 2b) as a breaking of the symmetry of the oscillation pattern, which now repeats every two oscillations. We display the RF spectrum of the P2 state at \( I = 523 \text{ mA} \) in Figure 2c. The signal to noise ratio of the \( f_{1}/2 \) frequency
component is 42.6 dB, indicating a high stability. With a further increase of the pump driving current, the next bifurcation to the period-four (P4) family of steady states appears at $I \approx 525$ mA, see the vibration waveform in Figure 2b and its RF spectrum in Figure 2c. Finally, when the pump driving current increases above 526 mA, the sharp spectral peaks disappear and the RF spectrum becomes broadband, suggesting that the internal motion of soliton molecules has become chaotic. The two pulses, still bounded within the soliton molecule, experience irregular oscillations and their separation trajectory is aperiodic, in contrast with the periodic trajectories of the P1, P2, and P4 families. Here, a clear P1-P2-P4-...-chaos sequence is demonstrated, following the classical route of period-doubling bifurcation cascade to chaos. However, at a pump driving current exceeding 532 mA, the soliton molecule becomes once again periodic, first with limit cycle attractors belonging to a second P4 family, and then undergoes a reverse period-doubling bifurcation to reach a second P2 family. Finally, another transition to chaos starts for $I > 538$ mA. In Figure 2d, we plot the RF spectrum of the direct photodetection of the laser output: it yields the cavity repetition frequency $f_c$, where both P1 oscillations (gray curve), P2 oscillations (blue curve), and chaos (red curve) are considered for comparison. For the P1 state, the fundamental cavity repetition frequency $f_c$ is accompanied by sharp modulation sidebands, whose separation from $f_c$ indeed matches with the BOC-measured oscillation frequency of the soliton molecule.

Nevertheless, we emphasize that the relative amplitude of these sidebands remains small, typically 35 dB or more below the main $f_c$ peak: this indicates a minor effect of the internal motion of the soliton molecule on the total energy of the soliton molecule. This observation is important to rule out any significant soliton shape instability contribution to the chaos observed in our experiments.[25] For the P2 state, a pair of sub-sidebands appears, with a frequency difference from $f_c$ being half of the initial P1 oscillating frequency. When reaching the chaotic states, the sharp modulation peaks disappear while the floor around $f_c$ increases by more than 10 dB compared with the P1 state, further demonstrating the aperiodicity of the chaotic state.

As a highlighted feature of the internal soliton molecular motions reported here, from periodic to chaotic, we emphasize again the fact that the optical soliton molecule always maintains its integrity, as a strongly bound soliton-pair system. This is linked to the fact that the relative timing variations, typically in the range of $\approx 100$ fs, remain small compared to the average

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**Figure 2.** Generation of chaotic soliton molecules in an ultrafast fiber laser. a) A full-record spectrogram shows the route of period doubling bifurcation to chaotic soliton molecules by increasing the pump strength. b) Vibration waveform of the pulses separation for P1, P2, P4 and chaotic soliton molecules. c) RF spectrum of the vibration waveforms displayed in (b). d) RF spectrum of the laser output for P1, P2, and chaotic soliton molecules. e) Average optical spectrum of a chaotic state recorded by an optical spectrum analyzer.
Figure 3. Statistical analysis of a chaotic internal soliton molecule motion. a) BOC output signal. b) A 3D phase space orbit reconstructed from the BOC output signal and the delayed BOC output signals with delay times $\tau$ and $2\tau$. The gray curves are projections of the phase portrait on three phase planes. c) Lyapunov exponent analysis of the BOC output signal. Inset: enlarged picture shows the exponent fitting result. d) Logarithmic plot of the correlation function $C_m(r)$ versus the radius $r$ for increasing embedded dimension $m$. e) Slope of the correlation function versus the radius $r$. A clear plateau is observed, with the black dashed curve giving an estimation of the correlation dimension.

We have also implemented a numerical simulation to qualitatively account for the observed pulse dynamics, based on the generalized nonlinear Schrödinger propagation equation. Its main results are presented in Section 3, with technical details included in the Supporting Information. We next analyze the statistics of the chaotic trajectories recorded in Figure 2b, an example of which is shown in Figure 3a. Figure 3b shows the chaotic orbit in a reconstructed 3D phase space, which is obtained from the BOC output signal of Figure 3a and the delayed BOC signals with delay times $\tau$ and $2\tau$. The delay time $\tau$ is chosen by the mutual information method, which is frequently used in the phase space reconstruction of 1D time series.[40] The geometric structures can be seen from the projections of the phase trajectory on the x–y, x–z, and y–z phase planes (gray curves). To quantitatively demonstrate that the reported interacting dynamics within the soliton molecule is chaotic, we extract the largest Lyapunov exponent $\lambda_L$ from the BOC signals. Lyapunov exponents quantify the divergence rate of nearby attractor trajectories in phase space and are widely used as a criterion for identifying chaos.[41] A necessary condition for chaos is the existence of at least one positive Lyapunov exponent $\lambda_1$. As shown in Figure 3c, the indicator of chaos is reflected by the average initial logarithmic divergence curve over all pairs of neighbors, where
an exponential divergence is reasonably linear with the Lyapunov exponent $\lambda_L \approx 6.5/\mu$s. We note that the divergence trajectory features saturation since the chaotic system is bounded in the phase space so that the average divergence cannot exceed the boundary of the strange attractor.

We also analyze the correlation dimension $D_2$ by using the Grassberger–Procacia algorithm. The correlation dimension is an efficient diagnostic to distinguish between chaos and random motion. For a random motion, the measured $D_2$ grows linearly with the increasing embedding dimension $m$, whereas in the situation of chaos, $D_2$ saturates to an asymptotic value. Figure 3d displays the logarithmic plot of the correlation function $C_m(r)$ as a function of the radius $r$ for increasing $m$ and the corresponding correlation integral slope versus the radius $r$ is shown in Figure 3e. The curve manifests a clear saturation of the correlation integral slope, toward an estimated correlation dimension $D_2 \approx 2.28$. A finite value for the correlation dimension supports the hypothesis of chaotic internal soliton-pair dynamics and gives an estimate of the degree of complexity of the motion of the interacting solitons. A detailed description of the analyzing method involving the delay time $\tau$, the embedding dimension $m$, the Lyapunov exponent $\lambda_L$, and the correlation dimension $D_2$ is presented in Section E, Supporting Information.

We verify the reproducibility of chaotic soliton molecule generation, by using a pump current modulated at 100 Hz with a wide modulation amplitude such that the soliton molecules experience a complete circular sequence of bifurcations (P1 - chaos - P1), as observed in Figure 2a for a driving current ranging from 500 to 530 mA. The resulting real-time BOC output signals of the pulse separation are shown in Figure 4a, with Fourier spectral analysis in Figure 4b: the recording demonstrates a perfect reproduciability of the bifurcation sequences leading to or departing from chaos. During the first half modulation period, a switching occurs from stable P1 to P2 states. A transient P4 state is then observed before the onset of chaos. Subsequently, the system returns to the P1 state through the reverse sequence of bifurcations, initiated by the reduction of the intracavity energy. This dynamical switching between regular and chaotic oscillations is also illustrated in Videos S1 and S2, Supporting Information, where we videotaped this reversible process, showing the real-time evolution of the waveforms and the RF spectrograms after setting a longer modulation period suitable for a clear view in real time. Whereas the bifurcation sequence is repeatedly observed, the details of intra-molecular chaotic dynamics can vary substantially over consecutive experimental runs, with another example mirroring the analysis of Figure 3 shown in Figure S5, Supporting Information.

### 2.4. All Optical Control of Chaotic Soliton Molecules

Chaotic dynamics is prone to exhibit considerable reaction to additional perturbations. In chaos theory, it was shown that chaos can be suppressed to a limited cycle attractor by non-feedback methods such as the application of external periodic forces. The elimination of chaos is most effective when the drive frequency (frequency of the external force) is close to the eigenfrequency of self-excited oscillations that exist in the vicinity (in the parameter space) of the chaotic motion. In our experiment, the free-oscillation frequency of the vibrating soliton molecule varies within the range of 1–5 MHz, so we choose a drive frequency $f_d = 2$ MHz. We here demonstrate that the chaotic dynamics involving bound solitons in ultrafast fiber lasers can be suppressed by external cavity injection, without affecting the original laser parameters and the structure of the individual solitons. First, we experimentally verify that the soliton molecule remains a robust entity with respect to the injection signal, including within the chaotic region. Therefore, the soliton molecule presents an opportunity of external control. As shown in Figure 5a, we implement the external control of chaos via the injection of a modulated continuous wave (CW), which is an efficient method for pulse dynamics modulation. In the optical spectral domain, the CW is
selected at 1530 nm, far enough from the soliton molecule spectrum to avoid a deleterious influence on the mode locking regime within a suitable range of injection power. The CW is amplified by an Er-doped fiber amplifier (EDFA), whose output power is adjustable. An electro optic modulator with a bandwidth exceeding 1 GHz driven by a function generator imprints a sinusoidal intensity modulation on the injection signal. The latter is injected into the cavity via a 10% coupler in the counterpropagating lasing direction, thus directly influencing the gain saturation level of the fiber laser with a bandwidth up to several MHz.

We experimentally prepare a chaotic soliton molecule whose dynamics of inter-pulse separation is manifestly broadband, as attested by the RF spectrum of the BOC output signal (black curve in Figure 5b) at 550 mA pump strength. Then, we control this state by increasing the injected CW laser intensity, measured as the driving power of the external EDFA. At a driving power of 2.8 mW, the chaotic motion becomes intermittent, interspersed by P2-like oscillations (blue curve in Figure 5d) with corresponding spectral peaks at half-integer multiples of \( f_s \) located at about 2.5 MHz on top of a broad continuum (blue curve in Figure 5b). Robust P1 oscillations are obtained at the current drive of 9.2 mW, as attested by both the RF spectrum and the regular vibration waveform (red curves in Figure 5b,d). The complete and deterministic route of chaos control through period-doubling reversals via external cavity injection is shown in Figure 5c. Note that excessive injection power (>9.8 mW) will result in the disruption of the soliton molecule. The frequency components of the injection signal and beat notes are also evidenced by the RF spectrum in Figure 5b, confirming that the external injection is efficiently coupled to the chaotic motion. Overall, the route of external injection for chaos control evolves in a highly reproducible and reversible manner, so that the internal motion of soliton molecules reverts to chaos if we weaken the injection strength. For a clearer visual presentation of our results, we also videotaped this fully efficient switching of chaos control, see Section S3 and Video S3, Supporting Information.

3. Numerical Simulation of Chaotic Soliton Molecules

In this section, we conduct a numerical simulation to qualitatively verify the experimental observations. These simulations are based on a generalized nonlinear Schrödinger equation (GNLSE) including dissipative (gain/loss) terms, see Experimental Section for technical details. Figure 6 illustrates typical examples of numerically obtained soliton molecule dynamics that confirm the existence of a route from period-doubling bifurcations to chaotic soliton molecules. Figure 6a,b shows a P1 oscillation dynamics at an energy saturation \( E_{\text{sat}} = 0.63 \text{ nJ} \) (\( E_{\text{sat}} \) is proportional to the pump power). By Fourier transformation of the optical spectra displayed over consecutive cavity roundtrips in Figure 6a, an oscillation period of \( \approx 25 \) roundtrips for the relative soliton separation is retrieved and displayed in Figure 6b. The amplitude of the separation oscillation is \( \approx 0.05 \text{ ps} \). A vertical magnification of the soliton molecule energy is plotted in Figure 6a, showing the same periodicity. Starting from this P1 state, a transition to P2 occurs by increasing \( E_{\text{sat}} \) to 0.69 nJ. The period doubling characteristics are observed in the optical spectrum, the soliton molecule
energy, and the pulse separation, shown in Figure 6c,d. At $E_{\text{sat}} = 0.71$ nJ, the soliton separation dynamics enter a chaotic regime, see Figure 6e. We plot the 3D phase portrait of the chaotic state in Figure 6f. The complex geometrical structure of the strange attractor is obtained, in stark contrast with the limit cycles attractors of P1 and P2 states. The bifurcation diagrams based on the monitoring of the local maxima, for both the soliton molecule energy and the pulse separation, are shown in Figure 6g. The latter clearly shows the bifurcation route P1-P2-P4-...-chaos.

4. Discussion and Conclusion

Whereas oscillating optical soliton molecules have been the subject of several seminal investigations, this article presents the first experimental demonstration of the route for excitation and all optical control of a chaotic soliton molecule in optical resonators and lasers. We emphasize on the fact that the chaos investigated here refers to the internal dynamics of the soliton molecule and is assessed to be low-dimensional, leaving the soliton molecule energy practically unchanged. This can be understood physically by the chosen situation where the variations of the inter-pulse separation are of the order of one percent of that separation, therefore perturbing only slightly the overall shape of the soliton molecule. Therefore, to observe such internal chaos requires a characterization method endowed with an exquisite sub-femtosecond sensitivity and true real-time monitoring. The BOC method fully qualifies, doing so without relying on any heavy post-processing of data. This allows to record experimentally the vibration waveform with the necessary high precision to observe and analyze the experimental internal chaos. We therefore demonstrated that the chaotic dynamics of bound pulses within a soliton molecule can be accessed and manipulated in an unambiguous way. The BOC method appears particularly suited in the investigation of the complex nonlinear dynamics of soliton molecules in optical oscillators, as much as it is widely used for timing jitter characterization in ultrafast lasers and ring resonators.\[44,45\] We anticipate that it will be a useful asset in precise investigation of nonlinear dynamics effects such as stochastic resonance\[46\] and subharmonic entrainment\[39\] of soliton molecules.

Following the present investigation, the intramolecular dynamics of optical soliton molecules is gaining diversity as it further strengthens the analogy with matter-like molecules. Indeed, our study constitutes a strong confirmation of the hypothesis that a wide range of soliton molecule oscillations can be viewed as low-dimensional dynamical systems that feature limit cycle attractors as well as strange attractors. This hypothesis stays reasonably valid if the amplitude of the intramolecular motion remains sufficiently small compared to the average separation between solitons.
The fast-error-free switching between ordered and chaotic soliton molecules enabled by pump current sweeping and external injection highlights the potential prospects of all-optical logic gates and chaotic communication using soliton molecules. Beyond the fundamental case of soliton-pair molecules, there is a large variety of molecular complexes, macro-molecules, and soliton crystals that could be similarly investigated. It is reasonable to assume that the chaotic dynamics observed in our experiment could be extended to three-soliton interactions in lasers, and even more complicated multi-body dynamics such as interactions within soliton molecular complexes and supramolecular structures in parallel optical-soliton reactors. More generally, the findings of chaotic interactions among dissipative solitons will be of significant interest in the context of analogous non-linear systems, such as Bose–Einstein condensates and hydrodynamics.

5. Experimental Section

Laser Setup: The chaotic soliton molecules under investigation were generated from an Er-doped fiber laser, mode-locked by the NPE technique. NPE provided a virtual quasi-instantaneous saturable absorber effect, whose transfer function can be widely adjusted by altering the orientation of the intracavity wave plates, leading to multifarious mode-locked states. The cavity incorporated two types of fibers: a 0.5-m long EDF (Liekki Er 110-4/125) served as the gain medium and the others were standard single-mode fiber (SMF 28). By changing the length of the SMF, two mode-locking states with different fundamental repetition frequency and net chromatic dispersion were obtained. The total length of the laser cavity of 46.4 (μm) yielded a fundamental repetition frequency of 44.8 (50.6) MHz, corresponding to a roundtrip time of 22.3 (19.76) ns. The laser had a net anomalous dispersion ≈−0.07 (−0.06) ps² at 1.55-μm wavelength. In both states, the mode locking threshold was at a pump driving current of about 380 mA (optical pump power ≈230 mW) and the threshold of MPI was at about 455 mA (optical power ≈270 mW). Soliton molecular chaotic dynamics were observed in both mode-locking states, proving that the observation was not a coincidence resulting from specific laser parameters.

Balanced Optical Cross-Correlator: A single-crystal BOC scheme had been implemented to detect the internal soliton molecular motion. The BOC setup consisted of a 4-mm type-II phase-matched periodically poled KTiOPO₄ (PPKTP) crystal (facet 1) coating: HT @ 1550 nm and 775 nm, facet 2 coating: HT @ 775 nm and HR @ 1550 nm), a dichroic mirror (HT @ 1550 nm and HR @ 775 nm), a focusing lens (f = 30 mm) and a balanced photodetector (Thorlabs PDB420A). The output of the BOC was detected by an oscilloscope (Agilent in-signal) for monitoring of intramolecular pulse separation in real time and a RF spectrum analyzer (RIGOL DSAS115) for spectrum analysis of chaotic dynamics.

Pump Current Modulation: The pump laser diode (LD) was driven by a commercial LD controller (Thorlabs CLD1015), which has an external RF input port for pump current modulation. A 100 Hz sinusoidal modulation generated by a function generator (Rigol DG5102) was used to modulate the laser. The modulation amplitude can be adjusted by varying RF voltage from the function generator output. The modulation bandwidth can be as high as 10 MHz, while, in Er-fiber lasers, it was limited to few kilohertz due to the low pass filtering effect caused by the dumped gain relaxation oscillations. Ref. [31] used this scheme for intramolecular pulse separation manipulation. Here, this technique allowed to reproduce the bifurcation sequences leading to or departing from chaos.

All-Optical Control of Chaotic Soliton Molecules: The external optical injection was based on an all-fiber configuration. The 1530 nm CW laser was generated by a commercial low noise tunable laser (Santec TSL-550). The output was power amplified by a home-made all polarization maintaining EDFA. A fiber-coupled high speed electro optic modulator (Conquer KG-AM-15-2.5G) was used to sinusoidally modulate the intensity of the CW and the MHz modulation signal was from a function generator (Rigol DCS102).

Numerical Simulations: They were based on GNLS, which included dissipative (gain/loss) terms. The model also included a lumped saturable absorber (SA) to promote mode locking. Similar piece-wise propagation models had been widely used to reproduce complex pulsed laser dynamics, such as soliton pulses, soliton molecule vibrations, and incoherent dissipative solitons. The propagation equation in the optical fibers reads:

\[
\frac{\partial A}{\partial z} + \frac{i}{2} \left( \beta_2 + \frac{\gamma}{2} \right) \frac{\partial^2 A}{\partial \tau^2} - \frac{\gamma}{2} |A|^2 A + \frac{\beta_3}{6} \frac{\partial^3 A}{\partial \tau^3} + i \frac{\gamma}{2} |A|^2 A = E_{\text{in}} \frac{P}{|A|^2} + i |E_{\text{int}}|^2 A - i |A|^2 A + i E_{\text{ext}} A + i E_{\text{int}} A + i E_{\text{in}} A
\]

where A is the field envelope, \( \frac{\partial A}{\partial z} \) is the propagation coordinate, \( \frac{\partial^2 A}{\partial \tau^2} \) is the space-time co-moving with the pulses. The parameters of fiber dispersion and effective nonlinearity corresponded to the commercial fibers Corning SMF-28 (passive fiber) and Liekki Er 110-4 (active fiber) used in the experiment. For the active fiber, \( g = g_0(1 + |A|^2 \frac{dr}{dr_{sat}} + (\omega - \omega_0) \Delta \omega_0 \) models the EDF gain, where \( g_0 \) is the small-signal gain, \( E_{\text{sat}} \) is the saturation energy, \( \omega \) is the optical angular frequency, \( \omega_0 \) is the central angular frequency. The saturable gain \( g \) has an FWHM bandwidth \( \Delta \omega_0 \) of 50 nm. The SA was modeled by a power-dependent transmittance function: \( T = 1 - a_0 \omega_0 q_0 (1 + P/(P_{\text{sat}})^{-1}) \), where \( a_0 \omega_0 \) is the non-saturable loss, \( P_{\text{sat}} \) is instantaneous power, \( E_{\text{sat}} \) is saturation power. The SA was modeled with 8% non-saturable loss, 30% modulation depth, and 50 W saturation power. Guided by the experiment, the energy saturation coefficient \( E_{\text{sat}} \) of the modeled gain fiber was adjusted to seek irregular dynamics of interacting solitons. Operating well above the laser threshold, \( E_{\text{sat}} \) becomes proportional to the pumping power.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

bifurcations, chaos, dissipative solitons, fiber lasers, soliton molecules, ultrafast characterization, ultrafast lasers

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