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Unveiling real-time pulsation characteristics and programmable switching of 3D optical soliton molecules

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Abstract: Three-dimensional (3D) dissipative solitons, arising from the interplay of spatiotemporal nonlinear interactions, exhibit substantial similarities to other multi-dimensional physical phenomena. In particular, the investigation of 3D soliton molecules holds great potential for deciphering the underlying mechanisms of molecular complexity and addressing unresolved challenges in nonlinear systems. However, the pulsation of 3D soliton molecules, beyond stationary states, remains largely unexplored in the experiment. Here, we reveal the real-time pulsation characteristics and programmable switching of 3D soliton molecules, enabled by a single-shot multispeckle spectral-temporal measurement technique. Diverse dynamics of 3D molecules are captured, including the generation of period-doubling and beyond, desynchronization-synchronization transition, and controllable fast switching under programmable pump modulation. The mechanism of pulsation characteristics of the 3D soliton molecule is elucidated by numerical simulation. Further studies indicate the gain-induced nonlinear spatiotemporal coupling drives the 3D soliton molecule with frequency-locking in the spectral domain and noticeable mode dissipation in the spatial part, forming a unique dissipative soliton state. Our findings provide a new perspective for unveiling the complexity of 3D soliton molecules, and bring new insights into the underlying nonlinear physics of multi-dimensional lasers as well as a variety of applications.

Keywords: 3D soliton molecule; spatiotemporal coupling; synchronization; programmable switching; Hopf bifurcation.

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1 Introduction

Three-dimensional (3D) light fields that emerge and propagate across higher-dimensional spaces have attracted substantial attention for their potential in enabling high-capacity information transmission and uncovering multi-dimensional nonlinear phenomena prevalent in disciplines such as chemistry, physics, biology, and materials science^{1–5}. Extensive research has explored the dynamics of optical solitons within multimode fiber systems, including octave-spanning supercontinuum generation, dispersive wave produc-

tion, enhanced nonlinear interactions via mode coupling, spatial beam self-cleaning, and self-organized instabilities^{6–10}. Notably, the 3D dissipative optical soliton, a localized wave exhibiting particle-like characteristics, has been experimentally observed in spatiotemporal mode-locked (STML) fiber lasers. In these systems, the transverse and longitudinal modes of the generated 3D solitons are synchronously locked, forming coherent spatiotemporal structures^{11,12}. This concept of 3D dissipative solitons paves the way for advancements in generating high-energy pulses, manipulating complex spatiotemporal light fields, and

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dissecting the intricate behaviors inherent to higher-dimensional nonlinear systems.

In general, dissipative soliton generation requires delicate balance between dispersion and nonlinearity as well as gain and loss. In single-mode fiber (SMF) lasers, the dispersion corresponds to chromatic dispersion. While in multi-mode fiber (MMF) lasers, intermode dispersion constitutes a new dimension that needs to be balanced for 3D dissipative soliton formation. Steady propagation of 3D soliton in MMF lasers needs subtle balance between gain, nonlinear loss (spatial saturable absorber, nonlinear coupling), linear loss (spectral filter, spatial filter, linear coupling), nonlinearity and dispersion. They differ from 1D solitons in SMFs as the ultrashort pulses within MMFs are made up of several modal pulses that can have different spectra. 3D dissipative solitons in MMF lasers are also different from light bullets in free space nonlinear systems, as mode confinement of the MMFs gives limited and discrete spatial modes (i.e., diffraction is naturally counteracted). Depending on the value of intermode dispersion, different mechanisms (Kerr nonlinearity, spatiotemporal saturable absorber, spatial coupling/filtering) can contribute differently to balancing the intermode dispersion, thereby generating the 3D dissipative soliton with complex spatial speckle structure. Spatiotemporal saturable absorber (SA) effect can reset the walk-off by saturable absorbing weak modes and redistributing energy among modes.

Compared to the one-dimensional (1D) optical soliton counterpart^{13–18}, 3D soliton dynamics that evolve from stochastic initial seeds in multiple dimensions exhibit much more complicated behavior^{19–22}. Some interesting 3D soliton dynamics have been studied through different methodologies, e.g., buildup dynamics of single or multiple solitons, birth and internal phase vibration, and rogue waves in 3D solitons^{23–25}. Beyond stationary single 3D soliton, spatiotemporal nonlinear interactions between individual solitons can give rise to bound configurations of 3D solitons, termed ‘3D soliton molecule’. In contrast to their 1D analogs, such as the well-studied temporal soliton molecule in single-mode fiber lasers^{26–29}, these 3D soliton molecules, governed by spatiotemporal coupling (STC), embody multi-dimensional complexities that align more closely with the fundamental concept of molecules observed in biochemistry, quantum superfluid systems, and gaseous environments. Furthermore, perturbation to physical parameters such as polarization, gain, loss, dispersion, and nonlinearity can induce diverse dynamical behaviors in 3D soliton molecules, thereby further increasing the possibility of optical soliton analogy with matter molecules.

Moreover, intelligent optoelectronics currently is witnessing an unprecedented demand fueled by the explosive growth of global data traffic and the rapid development of next-generation information technologies, all-optical computing and quantum information processing, with its funda-

mental goal being to develop programmable, high-capacity and low-loss optical signal sources and regulation technologies. Particularly, 3D soliton molecules are ideal candidates, as their excellent particle-like and dynamic properties break the limitations of traditional 1D solitons in single-mode fiber lasers and show great potential to solve the capacity and programmability bottlenecks of conventional optical information processing systems. Though soliton molecules have been extensively studied in ultrafast fiber lasers in recent years with their applications expanding to optical communications, all-optical storage and precision spectroscopy, most existing research focuses on the temporal or spectral dynamics of 1D soliton molecules, leaving the spatiotemporal coupling mechanisms, real-time pulsation characteristics and programmable switching of 3D soliton molecules largely unexplored. Meanwhile, conventional regulation methods for soliton molecules rely on passive intracavity balance of loss, dispersion or nonlinear effects, making active programmable control and fast dynamic switching hard to achieve and thus restricting their practical application in intelligent optoelectronic systems requiring flexible signal manipulation. In addition, the growing demand for encrypted information transmission and high-capacity all-optical storage applications urgently calls for novel optical encoding schemes based on programmable soliton dynamics, all of which makes the exploration of the real-time pulsation characteristics, programmable switching mechanisms and nonlinear dynamics of 3D soliton molecules highly significant for filling the research gaps in intelligent optoelectronics and promoting the practical application of soliton-based technologies.

Despite these fascinating prospects, research into the dynamics of 3D soliton molecules remains in its early stages. Most investigations rely primarily on time-averaged measurements, focusing on parameters such as tunable wavelength, pulse number, and pulse intervals in 3D soliton molecules^{30–33}. Therefore, unable to uncover the complex 3D soliton molecule dynamics, typically unpredictable, time-varying, and non-repeatable. While several studies have revealed the buildup and internal phase vibration of 3D soliton molecules^{23,24}, their key real-time pulsation characteristics, including Hopf bifurcation, transitions from desynchronization to synchronization, and controllable fast switching, remain largely unexplored in the experiment. Pulsation soliton molecules, as localized spatiotemporal structures with periodic oscillatory behavior, represent fundamental configurations in numerous nonlinear physical systems and are closely associated with a wide range of critical nonlinear phenomena^{34–41}. Although recent studies have reported experimental investigations of 1D pulsation soliton molecules^{36,42–45}, disclosing the real-time pulsation characteristics of 3D soliton molecules remains an undressed challenge. Furthermore, deciphering the complex dynamics of 3D soliton molecules requires real-time

measurement techniques with high spatiotemporal resolution, extended recording duration, and multidimensional measurement capabilities. However, the emergence of multi-scale disorder, stemming from spatiotemporal dispersions and intense nonlinear interactions, poses significant obstacles to both theoretical and experimental research.

In this work, we unveil the real-time pulsation characteristics and programmable switching of 3D soliton molecules using a multispeckle spectra-temporal measurement technique. Diverse dynamics of 3D pulsation soliton molecules are resolved, encompassing period-doubling and beyond from stationary states, desynchronization-synchronization transition, and controllable fast switching via programmable pump modulation. Further studies indicate the gain-induced nonlinear spatiotemporal coupling drives the 3D soliton molecule with frequency-locking in the spectral domain and noticeable mode dissipation in the spatial part, forming a unique dissipative soliton state. The underlying mechanism of pulsation characteristics of the 3D soliton molecule is clarified by numerical simulation. Specifically, the cascaded Kerr nonlinearity (induced by gain and GRIN fibers) plays a vital role in the period doubling operation of 3D soliton molecule. Combined saturable absorber (SA) response and fiber-induced nonlinearity shape 3D soliton molecules, leading to distinct spatiotemporal output features across successive cavity roundtrips. Further, numerical simulation indicates that synchronization of periodic energy evolution across transverse modes is critical to realizing sub-harmonic entrainment of 3D soliton molecules. These results represent a key step in deciphering the underlying nonlinear physics and developing controlling strategy of 3D soliton molecules in multi-dimensional laser systems.

2 Results

2.1 Experimental setup and principle

The experimental setup for generating 3D soliton molecules (SMs) in a multi-mode fiber laser is illustrated schematically in Fig. 1(a). The laser cavity includes a 3 m few-mode gain fiber (Nufern LMA-YDF-15/130-VIII) with a core diameter of 15 μm and support 6 modes, a 1.6 m few-mode passive fiber (Nufern LMA-GDF 15/130, support 6 modes) with same core diameter of the gain fiber, and a 2.5 m multi-mode graded-index (GRIN) fiber (Thorlabs GIF625, support 650 modes) with a core diameter of 62.5 μm , yielding a fundamental repetition rate of 25.8 MHz. To induce higher-order mode excitation, a large core offset $\sim 10 \mu\text{m}$ was introduced during the fusion splicing between the few-mode gain fiber and the passive multi-mode GRIN fiber, facilitating mode conversion from low-order to higher-order modes through mode coupling. Mode-locking in this 3D soliton laser was achieved via nonlinear polarization evolution (NPE) mechanism, which relies on intensity-dependent transmission characteristics to stabilize pulse formation. Figure 1(b) and 1(c) display the typical temporal pulse train and beam profile containing speckle grains (SGs), indicative of the stationary doublet 3D soliton molecule output.

The exemplary illustration of doublet 3D soliton molecule evolution under gain-induced spatiotemporal coupling is shown in Fig. 1(d), indicating the complex transient dynamics of 3D soliton molecules in both time and space domains. This can be further verified according to the mode-resolved pulses temporal intensity evolution (Fig. 1(e)), which signifies obvious intensity variation and energy transfer of multiple transverse modes across roundtrips. By precisely controlling

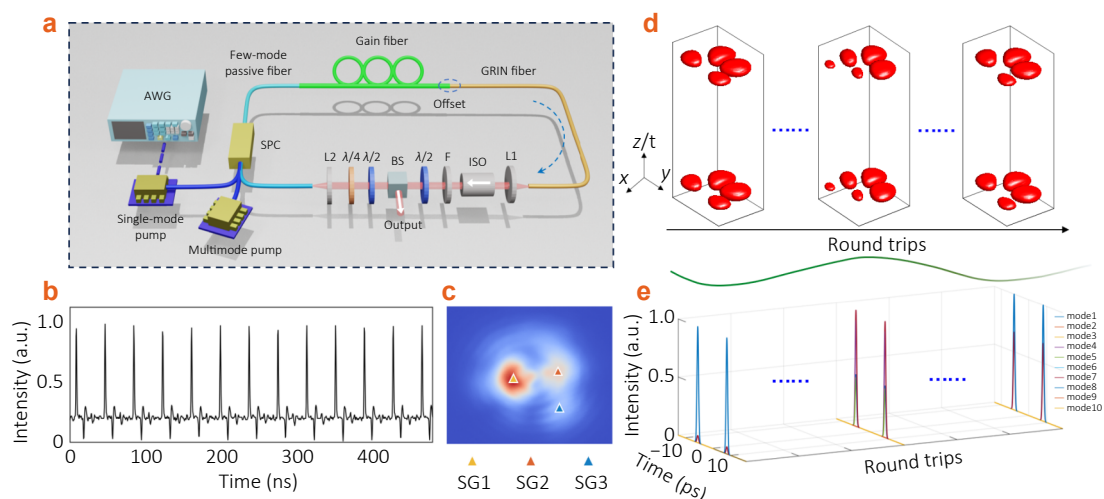


Fig. 1 | Schematic diagram of the spatiotemporal mode-locked fiber laser (STML) and typical characteristics of the 3D soliton molecule. (a) Schematic diagram of the multimode mode-locked fiber laser. BS: beam splitter. L: lens. F: filter. ISO: isolator. SPC: signal-pump combiner. $\lambda/2$: half-wave plate. $\lambda/4$: quarter-wave plate. AWG: arbitrary waveform generator. Typical pulse train in (b) and beam profile in (c) of doublet stationary 3D soliton molecule. (d) Exemplary illustration of doublet 3D soliton molecule evolution across multiple roundtrips under gain-induced spatiotemporal coupling. (e) Corresponding mode-resolved pulses temporal intensity evolution across multiple roundtrips.

pump power for gain tuning and exploring the parameter space through intracavity polarization, STML of 3D soliton molecules with diverse pulsation characteristics can be realized.

The output laser beam was directed through a beam splitter and then expanded using a telescopic magnification system, enabling clear resolution of individual SGs. This magnified beam was fed into a real-time measurement setup where spectral-temporal signals from multiple SGs were simultaneously captured by distinct single-mode probes. Off-line data processing and reconstruction techniques were employed to visualize the multispeckle spectral-temporal evolution of 3D soliton molecules across successive roundtrips (RTs). Additional details regarding the experimental system can be found in the Methods and Supplementary Information Fig. S1.

2.2 Period-doubling and beyond of 3D soliton molecule

Investigate the 3D soliton molecules possessing diverse characteristics, which can serve as a potential cross-disciplinary framework for exploring the complexities inherent in matter molecules. Notably, the 3D feature of optical solitons can be recognized through their intricate mode profile, wherein numerous speckle grains (SGs) arising from the coherent superposition of 3D optical solitons can be clearly distinguished. In this experiment, the stationary doublet 3D soliton molecule can be generated at a pump power of 1.4 W. Further increasing the pump power, the period-doubling and multiple-period bifurcation of 3D soliton molecule can be resolved in the experiment. To achieve precise pump power control, we add a multimode pump (27 W maximum output power) and single-mode pump laser (600 mW maximum output power) to the two arms of the signal-pump combiner (SPC). Therefore, the 0.1 mA adjustment precision of the pump current can be achieved through the single-mode laser diode. Figure 2(a) depicts the radiofrequency (RF) spectrum evolution of the 3D soliton molecule along the pump current increase (Measured by a radio spectrum analyzer connecting the whole laser output beam through a beam splitter). At low pump current (up to 726 mA), the STML laser generates a stationary doublet 3D soliton molecule, indicated by a single frequency component in the RF signal at the fundamental repetition rate f_r (see Fig. 2(a) and 2(d)). As the pump current increases, period-doubling behaviour emerges, characterized by the appearance of a sideband at half the repetition rate ($f_r/2$) with a broad spectral peak (see Fig. 2(e)). The intensity of the sideband gradually increases along the pump current and subsequently converges to a stable value, as shown in Fig. 2(b) (from 727 mA to 758 mA). Finally, the stable period-doubling of the 3D soliton molecule is formed at the pump current beyond 758 mA, as revealed by the appearance of the RF narrow peak ($f_r/2 = 12.9$ MHz) with high intensity

(see Fig. 2(f)). This transition from stationary to pulsating operation is associated with the ubiquitous dynamics termed ‘Hopf bifurcation’ and has now been further validated in the STML domain of 3D soliton molecules.

To quantify the mode content of 3D soliton molecules in different states along the pump current increase, we employed the stochastic parallel gradient descent (SPGD) algorithm to investigate the mode proportion of the STML output^{46,47}. This method retrieves the mode content only using the captured beam profile and is relatively simple. Here, the lowest 20 modes of the passive multi-mode GRIN fiber were considered for the SPGD-based mode decomposition. Although the passive GRIN fiber can support more than 100 modes, this simplification is still reasonable, as most energy of the 3D soliton is concentrated in the lower-order modes. The measured and retrieved beam profiles of 3D soliton molecule in the stable period-doubling state (pump current 765 mA) are presented in the inset of Fig. 2(g). Good agreement can be observed between the measured and retrieved beam profiles (correlation coefficient > 0.998). Encouraged by the high accuracy of our retrieve method, we further retrieve the mode content of 3D soliton molecule in stationary, modulated period doubling, and stable period doubling states, as shown in Fig. 2(g). The fundamental mode signifies a relatively small variation, while the higher-order modes (modes 2, 3, 8, and 10) display a stronger change, which also confirms the gain-induced spatiotemporal nonlinear interactions lead to the mode-dependent dissipation in the MMF laser. Further, we probe the optical spectra of 3D soliton molecule with stable period-doubling in three different SGs (see Fig. 2(c) at pump current 765 mA). The distinct optical spectra across three SGs show the multi-dimensional complexities of 3D soliton molecules.

Figures 3(a-c) present the spectral-temporal dynamics of doublet 3D soliton molecule with stable period-doubling in three different SGs (pump current 765 mA). The spatiotemporal structure of the 3D soliton molecule generates dense spectral interference fringes exhibiting distinct spectral evolution features across three SGs, as depicted in Fig. 3(a-i), and 3(c-i). The DFT spectra of two successive RTs in Fig. 3(a-iii)-(c-iii) further verifies the period-doubling characteristics of 3D soliton molecule with obvious central amplitude variation in the spectrum. Field autocorrelation traces were calculated from the results of DFT spectra to reveal the corresponding temporal dynamics, indicating the spatiotemporal dual-soliton molecule interaction during evolution (Fig. 3(a-ii)-(c-ii)). The more significant period-doubling characteristic can be resolved in SG2 with apparent sidelobe fluctuation during evolution (Fig. 3(b-ii)) compared to the other two SGs, which is consistent with the spectra amplitude variation shown in Fig. 3(b-iii).

At a higher pump current, we also observe the doublet 3D soliton molecule with 4-period pulsation, which can be clearly identified in the RF spectrum of the global measured

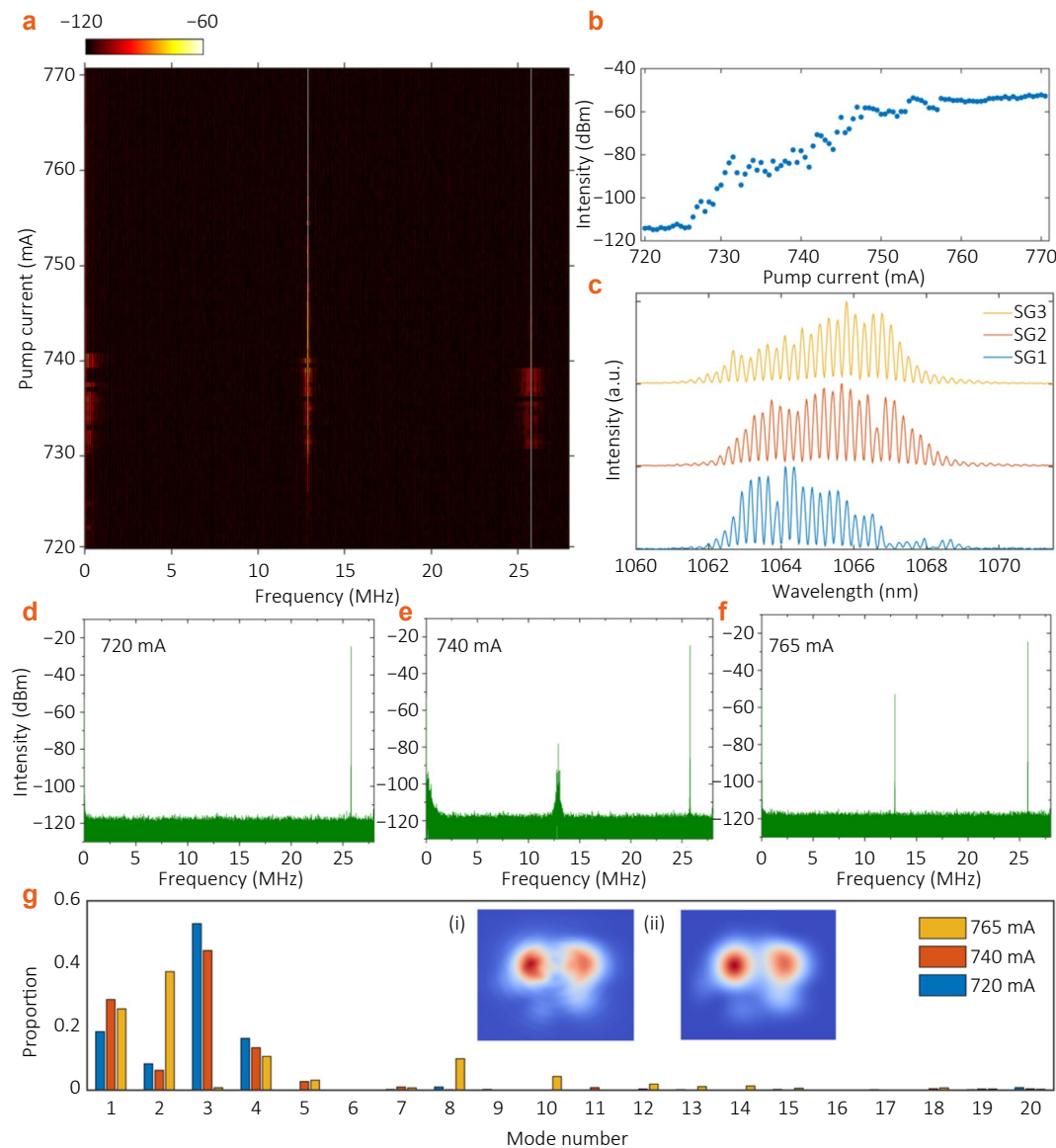


Fig. 2 | Map of the laser spectral intensity in the space of RF and pump current, showing the evolution of period-doubling dynamics of 3D soliton molecule. (a) Measured RF spectrum as a function of pump current. (b) Intensity of RF sideband along with pump current increase, showing 3D soliton molecule from stationary transition to period-doubling state. (c) Optical spectra of different SGs of 3D soliton molecule corresponding to the pump current of 765 mA. (d), (e), (f) single-shot RF spectra at different pump currents, corresponding stationary, modulated period-doubling, and stable period-doubling state of 3D soliton molecule. (g) Retrieved mode proportion under different pump currents for the 3D soliton molecule. (i, ii) Measured and retrieved beam profiles of stable period-doubling 3D soliton molecule at pump current 765 mA.

beam (Fig. 3(f)). The spectral-temporal dynamics of the 3D soliton molecule in two different SGs are shown in Fig. 3(d-i) and 3(e-i), corresponding to obviously different spectra evolution features. Periodic variation of the sidelobe with 4 RTs in the field autocorrelation traces is more conspicuous in SG1 than SG2 (Fig. 3(d-ii) and 3(e-ii)), demonstrating a stronger pulsation feature. This can be further verified in the RF spectrum for the two SGs, in which the sideband ($f_r/4$) of SG1 has a much higher intensity ~ 9 dB compared to SG2 (indicated by the arrows in Fig. 3(g-i) and 3(g-ii)). These findings further suggest the significance of

multi-dimensional characterization in comprehensively quantifying the pulsation trait of 3D soliton molecules and dissecting their underlying dynamics. The spectral-temporal evolution of the 3D soliton molecule with 4-period pulsation in SG3 and single-shot spectra within a pulsation period in three different SGs can be found in Fig. S5. By precisely tuning the pump power and intracavity polarization setting, the spectral-temporal dynamics resolved by speckles of 3D soliton molecules with 5- and 6-period pulsations can also be observed, which is presented in Supporting Information Figs. S6 and S7.

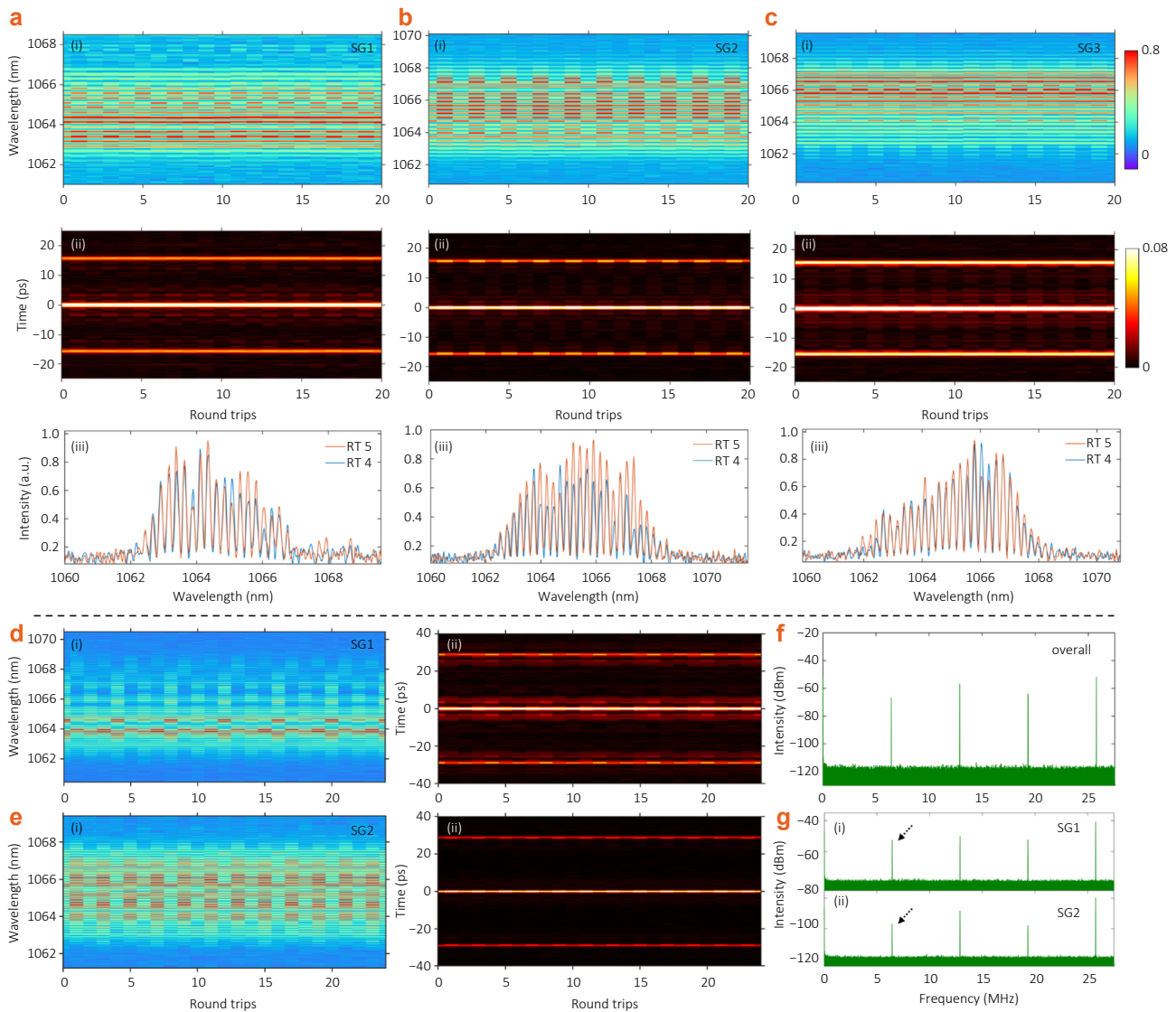


Fig. 3 | Spectral-temporal dynamics resolved by speckles of 3D soliton molecule with period doubling and 4-period pulsation. (a-i), (b-i), (c-i) Spectral evolution of the 3D soliton molecule with period-doubling in three different SGs. (a-ii), (b-ii), (c-ii) Field autocorrelation trace evolution corresponding to (a-i), (b-i), (c-i), respectively. (a-iii), (b-iii), (c-iii) Single-shot spectra of two successive roundtrips in three different SGs. (d-i), (e-i) Spectral evolution of the 3D soliton molecule with 4-period pulsation in two different SGs. (d-ii), (e-ii) Field autocorrelation trace evolution corresponding to (d-i), (e-i), respectively. (f) Single-shot RF spectra of global 3D soliton molecule with 4-period pulsation. (g-i), (g-ii) Single-shot RF spectra of SG1 and SG2, respectively.

To further understand the dynamics of 3D soliton molecules with period-doubling characteristic, a numerical simulation based on the generalized multimode nonlinear Schrödinger equations (GMMNLSE) is performed^{11,12}. During the simulation, only six transverse modes are considered in the GRIN fiber to reduce the huge computing time, and three transverse modes are considered for the gain fiber and the matched passive fiber (see simulation details in the Methods section). Through setting appropriate parameters for the saturable absorber (SA) and the saturation energy of the gain fiber, a characteristic evolution of 3D soliton molecules with period-doubling is depicted in Fig. 4. The simulated mode-resolved spectrum and temporal intensity

of 3D soliton molecule are shown in Fig. 4(a) and 4(b). Due to the different spatial distributions and gain competitions in the gain fiber, the locked multiple transverse modes have differences in pulse durations, spectral shapes, and intensities. Notably, the multiple transverse modes with period doubling characteristic can keep good synchronization, as indicated by the corresponding energy evolution (Fig. 4(c)). Here, the pulse energy is mainly concentrated on the four transverse modes, while the periodic energy evolution can still be observed in the LP11a and LP02 modes. The overall spectral evolution of the 3D soliton molecule, along with the single-shot spectra of two consecutive roundtrips, effectively mirrors the spectral period-doubling characteristic.

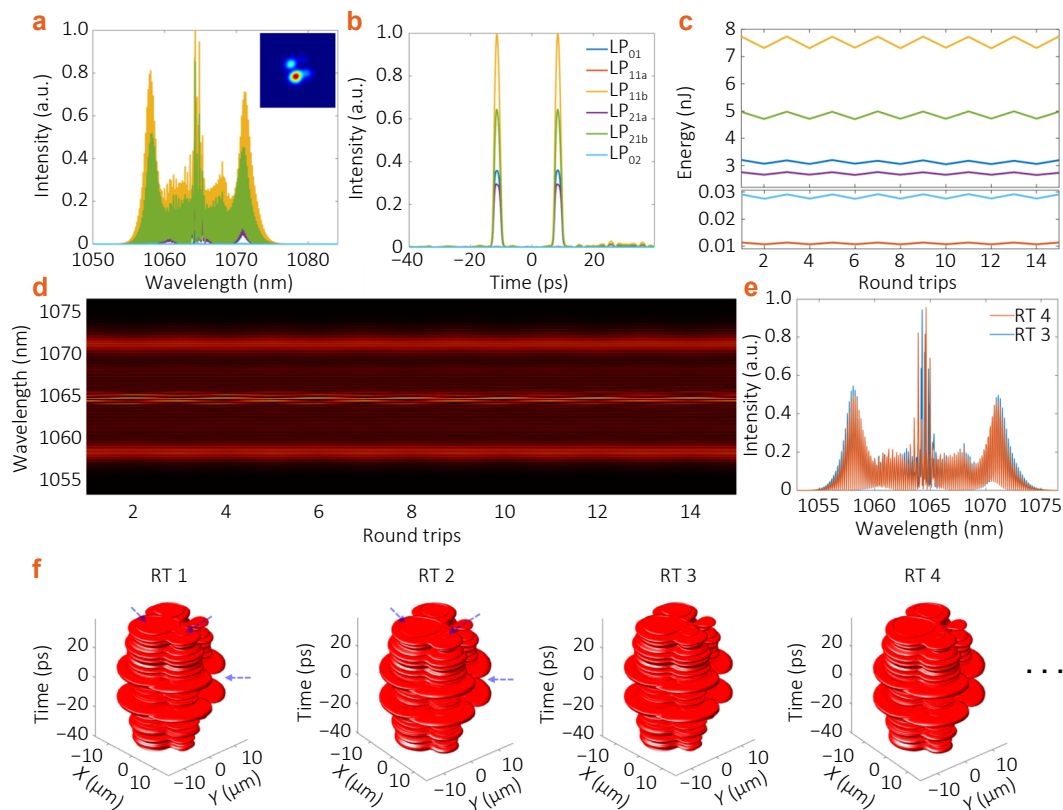


Fig. 4 | Numerical simulation of 3D soliton molecule with period doubling. (a) Simulated single-shot mode-resolved spectrum (insert corresponds to the beam profile) and (b) mode-resolved temporal intensity of 3D soliton molecule. (c) energy evolution of different transverse modes. (d) Total spectra evolution of 3D soliton molecule. (e) Single-shot spectra of two successive roundtrips from (d). (f) Spatiotemporal intensity of four successive output pulses of the 3D soliton molecule with period-doubling.

Figure 4(f) exhibits the spatiotemporal intensity of four successive output 3D soliton molecules. The intensity variation can be resolved (indicated by the blue arrows), further demonstrating the spatiotemporal period-doubling bifurcation of the 3D soliton molecule. Additionally, the simulated intracavity optical field evolution over two consecutive roundtrips of 3D soliton molecule is shown in Supporting Information Fig. S2.

2.3 Desynchronization and synchronization of 3D soliton molecule

Remarkably, with a different intracavity polarization setting, desynchronization to synchronization of doublet 3D soliton molecule can be resolved by solely increasing the pump current, as evidenced by the measured RF spectrum (Fig. 5(a)). The 3D soliton molecule synchronizes when its pulsation frequency is a sub-harmonic of the laser cavity fundamental frequency. In contrast, during the desynchronized phase, the pulsation frequency of solitons no longer maintains an integer-multiple relationship with the fundamental repetition rate^{35,36}. Specifically, at low pump current (up to 798 mA), the 3D soliton molecule corresponds to an un-frequency locking state, the f_r not corresponding to an

integer time of pulsation frequency (f_p), which is further verified by the single-shot RF spectrum shown in Fig. 5(d). We term this STML laser regime ‘non-subharmonic pulsation’ (N-SUB) presents much broader linewidth of the RF sideband. Continuous increases in pump current induce the RF sideband intensity enhancement and apparent frequency drift, as shown in Fig. 5(b) and 5(c) (pump current 792-801 mA). Here, the specific measurement parameter of RF spectrum is frequency span range of 40 MHz, sampling data-points of 2000, and resolution bandwidth of 100 Hz. The large RF span range may cause the frequency fixed at several discrete points (Fig. 5(c)). The increased sideband intensity corresponds to a stronger pulsation ratio of the 3D soliton molecule, as further evidenced by the single-shot RF spectrum at pump current 798 mA (Fig. 5(e)). Besides the subharmonic peak appears ($f_p = f_r / 3$), there are also many spectral lines locate symmetrically on both sides of the subharmonic peak, thus, termed a ‘modulated subharmonic’ (M-SUB) structure. When the pump current is above 801 mA, the weakly modulated spectral lines disappear, and only the subharmonic peaks (SUB) exist, demonstrating the 3D soliton molecule achieves pulsation frequency-locking, corresponding to^{48,49} the winding number 1/3 ($f_p = 8.6$ MHz). The frequency-locking character of 3D soliton molecule can

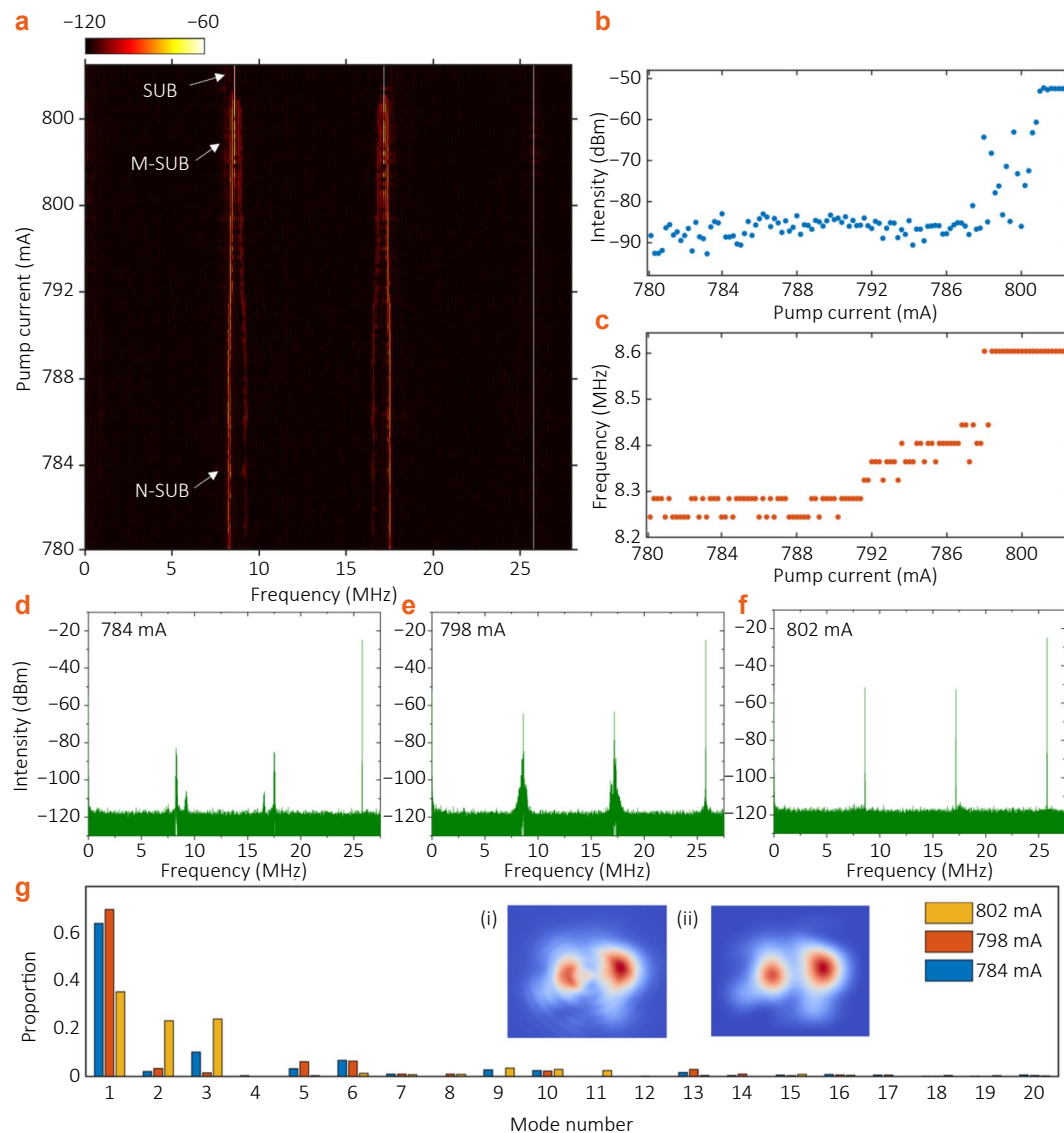


Fig. 5 | Map of the laser spectral intensity in the space of RF and pump current, showing desynchronization transition to synchronization of 3D soliton molecule with 3-period pulsation. (a) Measured RF spectrum as a function of pump current. Non-subharmonic (N-SUB), modulated subharmonic (M-SUB), Subharmonic (SUB). (b, c) Intensity and frequency along with pump current increase, showing 3D soliton molecule from N-SUB transition to SUB, the synchronization region of SUB state relating to the winding number $1/3$. (d, e, f) single-shot RF spectra at different pump currents, corresponding N-SUB, M-SUB, and SUB state of 3D soliton molecule. (g) Retrieved mode proportion under different pump currents for the 3D soliton molecule. (i, ii) Measured and retrieved beam profiles of SUB 3D soliton molecule at pump current 802 mA.

be further resolved in the single-shot RF spectrum (pump current 802 mA) in Fig. 5(f) with a pronounced intensity increase and narrow linewidth of the sideband. Subsequently, the intensity and frequency of the RF sideband will maintain constant when further improving the pump current (Fig. 5(b) and 5(c)).

Simultaneously, we execute the mode analysis to quantify the mode content of the 3D soliton molecule in different states. The measured and retrieved beam profile of 3D soliton molecule with frequency locking (pump current 802 mA) is shown in the inset in Fig. 5(g). Good consistency between them proves the precision (correlation > 0.997) of

our retrieve method is satisfactory. The retrieved mode contents of 3D soliton molecules in N-SUB, M-SUB, and SUB are present in Fig. 5(g). The mode proportion variation mainly concentrated on the first three modes (modes 1, 2, and 3) suggests the gain-induced spatiotemporal coupling and nonlinear interaction can cause the 3D soliton molecule with frequency-locking in the spectral domain and notable mode dissipation in the corresponding spatial part. The spectral-temporal dynamics of 3D soliton molecule with N-SUB and SUB states in three different SGs can be found in Supporting Information Fig. S4.

To validate our experimental findings and gain a better

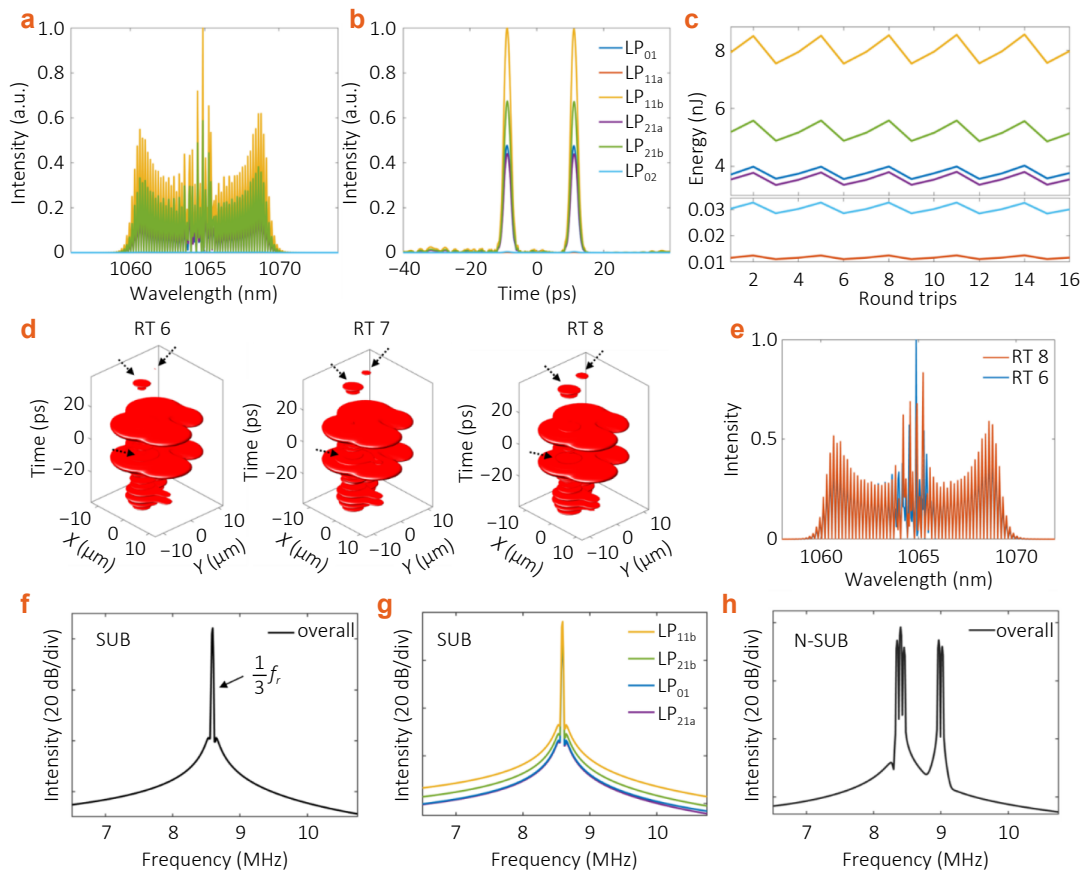


Fig. 6 | Numerical simulation of 3D soliton molecule with synchronization and desynchronization. (a) Simulated single-shot mode-resolved spectrum and (b) mode-resolved temporal intensity of 3D soliton molecule. (c) energy evolution of different transverse modes in SUB state. (d) Spatiotemporal intensity of three successive output 3D soliton molecule within a pulsation period. (e) Single-shot spectra within a pulsation period. (f) RF spectra of SUB 3D soliton molecule and (g) RF spectra of different transverse modes achieved from the Fourier transform from (c). (h) RF spectra of N-SUB 3D soliton molecule.

insight into the 3D soliton molecule complex evolution from N-SUB to SUB state, the numerical simulation is executed and shown in Fig. 6. The simulated mode-resolved spectrum and temporal intensity of 3D soliton molecule with 3-period pulsation are shown in Fig. 6(a) and 6(b). The spatiotemporal intensity evolution and single-shot spectra within one pulsation period (Fig. 6(d) and 6(e)) reflect the soliton molecule pulsation characteristics well. Notably, as evidenced by the corresponding energy evolution in Fig. 6(c), multiple transverse modes sharing the same pulsation period maintain excellent synchronization. Here, the pulse energy is predominantly centered on the four transverse modes, and the LP₀₁ and LP_{21a} mode has a more closer energy distribution. Further, the RF spectra of 3D soliton molecule with SUB and N-SUB states can be obtained by Fourier transformation of the simulated energy evolution (Figs. 6(c) and S3) shown in Fig. 6(f) and 6(h), which agree well with the experimental result (Fig. 5(f) and 5(d)). The only one RF side peak in Fig. 6(f) with high SNR and pulsation frequency of 8.6 MHz signifies the synchronization feature of 3D soliton molecule, relating to the winding number $1/3$. The simulated energy evolution of the N-SUB 3D soliton molecule can be found in Supporting Infor-

mation Fig. S3. As shown in Fig. 6(g), the RF spectra of different transverse modes further demonstrate synchronization of energy periodical evolution of different transverse modes plays a significant role in achieving the sub-harmonic entrainment of 3D soliton molecules. The lag synchronization or weak consistency of the energy evolution of different transverse modes will both deteriorate the periodicity and break the sub-harmonic state.

2.4 Controllable switching of 3D soliton molecule under programmable pump modulation

As observed in the above section, a linear increment in the pump power triggers the state transition of the 3D soliton molecule. Consequently, programmable electronic modulation of the pump current can be employed to achieve controllable deterministic switching of 3D soliton molecules. Figure 7 presents the consecutive switching of the 3D soliton molecule between stationary and pulsation states under programmable pump modulation. The periodic electronic modulation signals with a modulation frequency of 5 kHz are shown in the inset of Fig. 7(a) and 7(d) (red line). Obvious different spectral interference fringes can be resolved

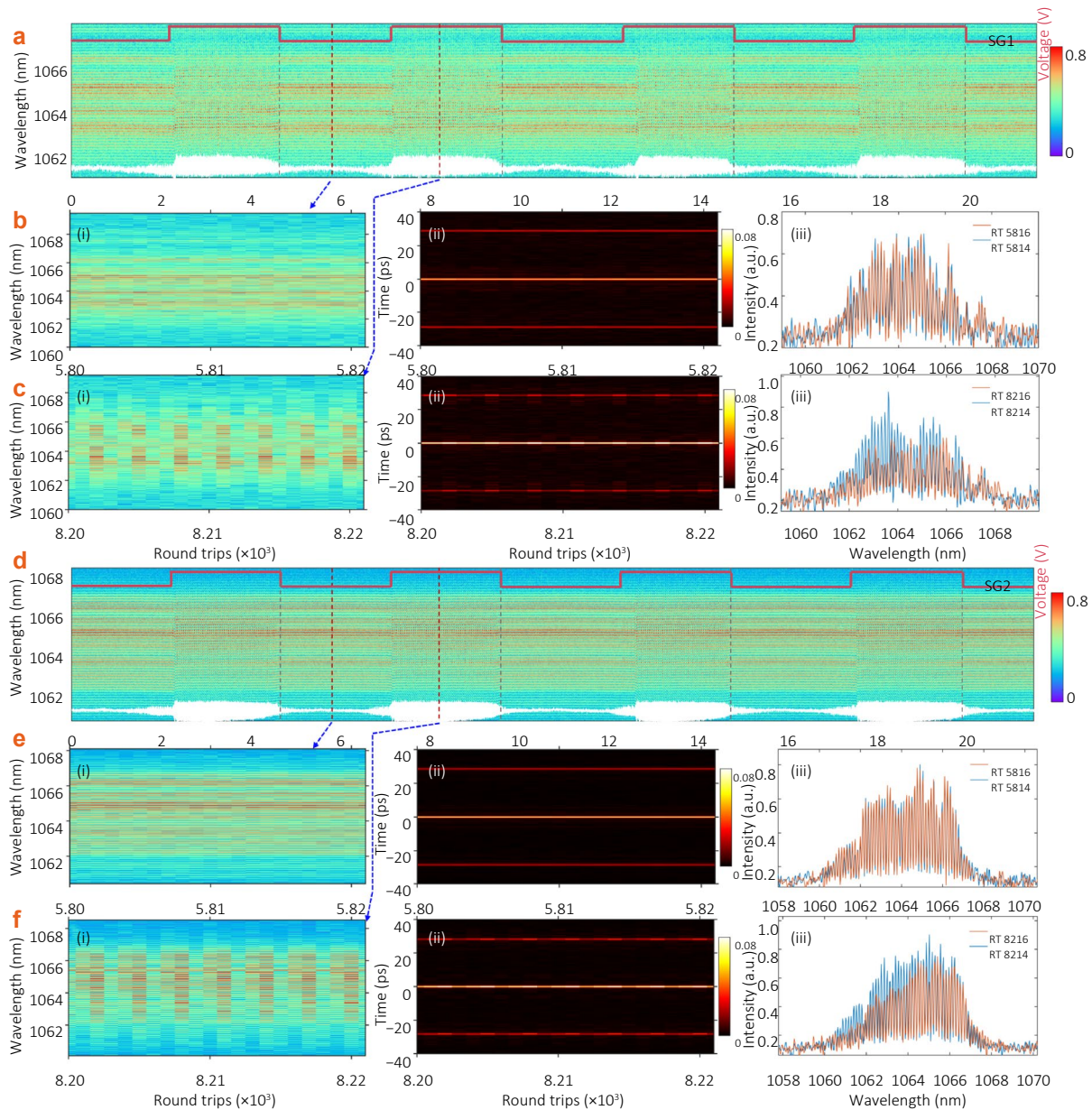


Fig. 7 | Controllable switching of 3D soliton molecule under programmable pump modulation. (a, b-i, c-i) Spectral evolution of the 3D soliton molecule in SG1. The white curve denotes the energy evolution. (b-i), (c-i) corresponding to the red dashed lines in (a). (b-ii, c-ii) Field autocorrelation trace evolution corresponding to (b-i), (c-i). (b-iii, c-iii) Single-shot spectra at the RT numbers of maximal and minimal energies within a pulsation period in SG1. (d, e-i, f-i) Spectral evolution of the 3D soliton molecule in SG2. (e-i, f-i) corresponding to the red dashed lines in (d). (e-ii, f-ii) Field autocorrelation trace evolution corresponding to (e-i), (f-i). (e-iii, f-iii) Single-shot spectra at the RT numbers of maximal and minimal energies within a pulsation period in SG2.

between two states during continuous switching, and further verified by the zoom-in plot of the DFT spectra evolution (Fig. 7(b-i), 7(c-i), 7(e-i), 7(f-i)). Combine the energy evolution of the 3D soliton molecule during continuous switching (white curve in Fig. 7(a) and 7(d)), the soliton molecule with low energy corresponding to a stationary state, and further verified by the field autocorrelation trace evolution and single-shot spectrum (Fig. 7(b-ii), 7(b-iii), 7(e-ii), 7(e-iii)). In sharp contrast, the 3D soliton molecule at higher energy signifies a strong pulsation characteristic with a

pulsation period of 3 RT, as indicated by the DFT spectra and field autocorrelation traces (Fig. 7(c-i), 7(c-ii), 7(f-i), 7(f-ii)). The apparent amplitude variation of optical spectra during one pulsation period further demonstrates the strong pulsation ratio of the 3D soliton molecule (Fig. 7(c-iii) and 7(f-iii)). Moreover, the pronounced difference in the spectra interference fringes in SG1 and SG2 suggests an underlying spatiotemporal interaction within the 3D soliton molecule. The corresponding whole field autocorrelation trace, relative phase, and RF spectrum evolution of the 3D soliton

molecule during continuous switching can be found in Supporting Information Figs. S8, S9 and S12. Further, the spatial structure evolution of the 3D soliton molecule during continuous switching is shown in Supporting Information Fig. S10, exhibits obvious mode content variation between the stationary and pulsation states. These results demonstrate that the gain-induced spatiotemporal coupling can drive the 3D soliton molecule to evolve in multiple dimensions, thereby generating diverse spatio-spectral-temporal landscapes and leading to the complex 3D soliton dynamics. To verify the reproducibility and stability of 3D soliton molecule during consecutive switching, we also calculate the correlation coefficient and can be found in Supporting Information Fig. S11.

Further, the controllable fast switching of 3D soliton molecule with versatile spectral characteristics is beneficial for potential applications in optical computing, information

encoding, and soliton communication. As proof of concept, we conducted the digital coding using two different energy values of 3D soliton molecule (high energy for '1' and low energy for '0'), corresponding to pulsation and stationary states, respectively. Here, the stationary and 3-period pulsation 3D soliton molecule generation corresponding to the pump power 1.450 W and 1.505 W, respectively. The pulsation characteristic of 3D soliton molecule under programmable pump modulation can be further verified by the RF spectra evolution, as shown in Fig. S13. Benefiting from the diverse spectrum landscapes across distinct SGs induced by nonlinear spatiotemporal coupling, dual-layer multi-letter encoding of 3D soliton molecule can be achieved owing to the opposite energy evolution in SG1 and SG2. As a result, the optical binary codes of QOM (10001|01111|01101, an abbreviation from Quality of Mode) and NPR (01110|10000|10010, an abbreviation from Nonlin-

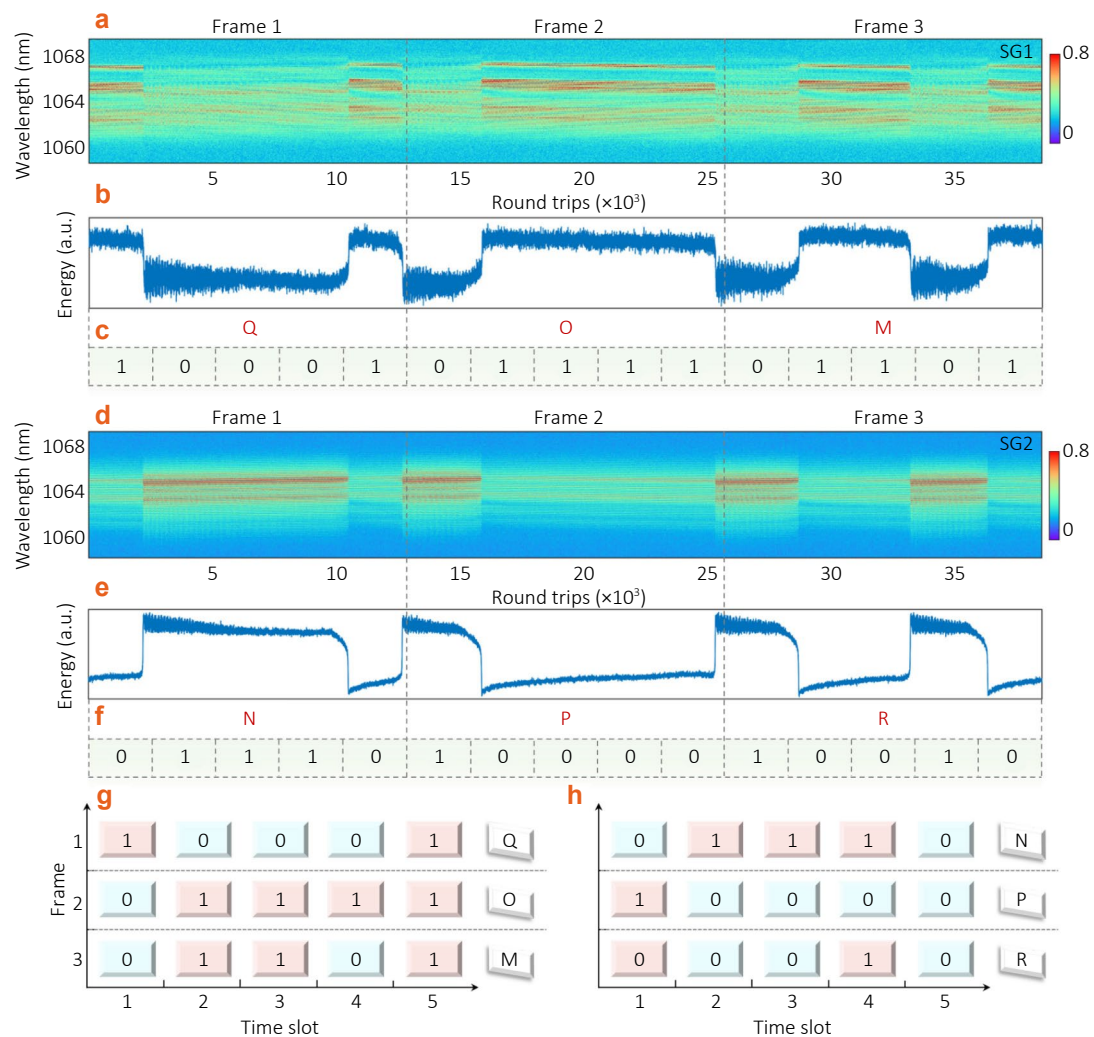


Fig. 8 | Exemplary dual-layer multi-letter encoding of 3D soliton molecule under programmable pump modulation. Successive recording of the encoded DFT streams in (a) SG1 and (d) SG2. (b, e) Corresponding energy evolution in SG1 and SG2. (c, f) ASCII binary codes of each letter which contain 5 binary bits. (g, h) Visualization of each letter. Each frame is composed of five time slots and represents a single letter.

ear Polarization Rotation) can be obtained by programming the pump current, as shown in Fig. 8. Each letter can be encoded by 5 bits with a total temporal duration of 500 μ s, corresponding to several kHz encoding streams. For visualization, the corresponding multi-letter streams are shown in Fig. 8(c) and 8(f). This dual-layer multi-letter test further verifies the availability of 3D soliton molecule-based digital encoding and the potential for applications in encrypted information transmission and high capacity all-optical storage.

3 Conclusions

In summary, we unveil the real-time pulsation characteristics and programmable switching of 3D soliton molecules using the multispeckle spectral-temporal measurement technique. Multiple dynamics of 3D soliton molecules are resolved, including the generation of period-doubling and beyond originating from the stationary operation. The distinguishable features of 3D soliton molecule in different SGs can be clearly observed, as indicated by the different speckle-resolved spectra and autocorrelation trace evolution. When observing the 3D soliton molecule from desynchronization transition to synchronization, noticeable mode dissipation can be resolved in the spatial part and forms a unique dissipative soliton state, which will not be observed in the 1D temporal soliton molecule. Similar spatial structure evolution can be viewed during the 3D soliton molecule from stationary transition to period-doubling operation, and continuous controllable switching under gain modulation. Numerical simulation further shows that the synchronization of the energy's periodic evolution in different transverse modes is crucial for achieving the sub-harmonic entrainment of 3D soliton molecules. Either lag synchronization or weak consistency in the energy evolution of different transverse modes will degrade the periodicity and disrupt the sub-harmonic state.

Moreover, controllable fast switching of 3D soliton molecule involving pulsation and stationary operation is achieved under programmable pump modulation, which is applicable to a series of soliton pulses used for binary 0-1 modulation, enabling data to be hidden within the multi-speckle of the 3D soliton. This can result in a dual-layer signal that carries both regular and encrypted data within the same soliton pulse sequence, which may stimulate advanced applications of coherent optical communications, all-optical storage, and optical computing. Benefit from the diverse spectrum landscapes across distinct SGs induced by nonlinear spatiotemporal coupling, dual-layer multi-letter encoding of 3D soliton molecule firstly be achieved owing to the opposite energy evolution in different SGs. Currently, limited by the electrical modulation speed of laser-diode driver (maximum modulation frequency of 100 kHz), thus, the encoding rates can be optimized by using faster electronic modulation devices. When further scaling the laser

cavity length with a higher repetition rate and sampling more SGs in the spatial structure of 3D soliton molecule, we anticipate the higher encoding rates can be achieved.

Further, the period-doubling mechanism of the 3D soliton molecules is theoretically uncovered, demonstrating that the cascaded Kerr nonlinearity induced by gain and GRIN fibers plays a vital role in the operation. The combined action of the SA response and fiber-induced nonlinearity sculpts 3D soliton molecules, resulting in notably distinct spatiotemporal output features across successive cavity roundtrips. Moreover, the gain-induced nonlinear spatiotemporal coupling drives the 3D soliton molecule to frequency-locking in the spectral domain and noticeable mode dissipation in the spatial part. Such an STML experimental platform, enabling generation of 3D soliton molecules and real-time multidimensional characterization, further deepens the analogy between optical solitons and matter molecules. Coupled with the digital applications growing demand for innovative soliton dynamics-based optical encoding schemes, exploring the nonlinear dynamics and programmable switching characteristics of 3D soliton molecules is of great significance for filling research gaps in intelligent optoelectronics and advancing the practical application of soliton-based technologies. We anticipate that our work shed new light on the complex nonlinear processes of the multi-dimensional laser system, and represent a key step in dissecting the underlying mechanisms and developing controlling strategy of 3D soliton molecules as well as the related applications.

4 Methods

Experimental setup: The experimental setup designed for investigating 3D soliton molecules integrates a multimode mode-locked fiber laser with a real-time single-shot multi-speckle spectral-temporal measurement system, enabling multidimensional signal visualization (see Supplementary Information Fig. S1 for detailed schematics). The STML fiber laser adopted a ring-cavity design, which integrates several key fiber components: a 3 m-long ytterbium-doped gain fiber (Nufern LMA-YDF-15/130-VIII) featuring a 15 μ m core diameter and capable of supporting 6 modes; a 1.6 m few-mode passive fiber (Nufern LMA-GDF 15/130) with a core diameter matching that of the aforementioned gain fiber; and a 2.5 m section of graded-index (GRIN) multimode fiber (Thorlabs GIF625) that has a 62.5 μ m core diameter and can support up to 650 modes. The gain fiber and GRIN fiber were fusion-spliced with a large core offset of approximately 10 μ m, a configuration intended to promote the excitation of higher-order modes. For the STML system to operate effectively, several critical components are required, including a bandpass spectral filter, polarization waveplates, and a polarization-dependent isolator. Laser output was extracted via a beam splitter (splitting ratio 50 : 50), expanded using a 5 $\times$ magnification telescope, and

then fed into the real-time measurement system. Within this system, the detected signal splits into three channels, each capturing individual speckle patterns of the multimode beam through single-mode fiber probes. The collected signals undergo optical time-division multiplexing before being divided into two paths. One branch preserved signal temporal information without dispersion, measured directly by a 20-GHz bandwidth high-speed photodiode paired with an oscilloscope (Keysight DSOV204A, 80 GS/s sampling rate). The second branch passes through a long dispersion fiber (0.6 ns/nm dispersion) for real-time spectral analysis via time-stretch dispersive Fourier transformation (TS-DFT) using another photodiode. This configuration achieves temporal and spectral resolutions of 50 ps and 0.1 nm⁵⁰, respectively.

Numerical simulation: To understand the complex dynamics of 3D soliton molecule, the numerical simulation is executed, including the gain competition between different modes in the few-mode gain fiber, the spatial-dependent gain saturation, the instantaneous spatial-dependent SA, the spectral and spatial filtering effect. The generalized multimode nonlinear Schrödinger equation (GMMNLSE) is used for the simulation of pulse propagation in fiber parts:

$$\partial_z A_p(t; z) = i\delta\beta_0^{(p)} A_p - \delta\beta_1^{(p)} \partial_t A_p + \sum_{m=2}^3 i^{m+1} \frac{\beta_m^{(p)}}{m!} \partial_t^m A_p + i \frac{n_2 \omega_0}{c} \sum_{l,m,n} S_{plmn}^K A_l A_m A_n^*, \quad (1)$$

where $A_p(t; z)$ denotes the temporal envelope of the electric field for the p -th transverse mode, $\beta_m^{(p)}$ represents the m -th-order dispersion coefficient corresponding to the p -th mode, S_{plmn}^K is the nonlinear coupling coefficients for the Kerr effects, n_2 is the nonlinear refractive index, $\delta\beta_0^{(p)}$ represents the propagation constant and $\delta\beta_1^{(p)}$ represents the group velocity of the p -th transverse mode.

In the few-mode gain fiber, to model the gain competition between different transverse modes and the spatial dependence of the gain saturation, the expression is shown as:

$$g(x, y, \omega; z) = \frac{g_0(\omega)}{1 + \int |A(x, y, t; z)|^2 dt / F_{sat}}, \quad (2)$$

$$A(x, y, t; z) = \sum_p \frac{F_p(x, y) A_p}{\sqrt{\iint F_p(x, y) dx dy}},$$

where $F_p(x, y)$ represents the mode profile of the p -th transverse mode, F_{sat} stands for the saturation time integrated intensity, g_0 denotes the small-signal gain. The gain spectrum was modeled by adopting a Gaussian profile with a full width at half maximum (FWHM) bandwidth of 40 nm.

For the saturable absorber effect that applied to the 3D optical field, the corresponding expression is shown as:

$$A_{out}(x, y, t; z) = A_{in}(x, y, t; z) \sqrt{1 - \frac{q}{1 + (|A(x, y, t; z)|^2 / I_{sat})}}, \quad (3)$$

where $A(x, y, t; z)$ is the summation of all modes, I_{sat} is the saturation intensity, q is the modulation depth. The simulation parameters of the STML fiber laser follow the typical experimental values.

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Author contributions

Y.Z., K.Y. conceived and performed most experiments and simulations. K.K.T., H.Z., and K.K.Y.W. supervised and guided the project. Y.Z., K.Y., Y.D., Y.L., K.K.T., H.Z., and K.K.Y.W. discussed the results and contributed to the writing of the paper.

Competing interests

The authors declare no competing financial interests.

Supplementary information

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