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## Supporting Information For

### **Resonantly driven nonlinear dynamics of soliton molecules in ultrafast fiber lasers**

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In the following, we provide the technical details concerning:

- (A) The ultrafast fiber laser model used to numerically confirm the resonant excitation of soliton molecules observed in experiments.
- (B) The Duffing model and the fitting of the Backbone curve of soliton molecules.

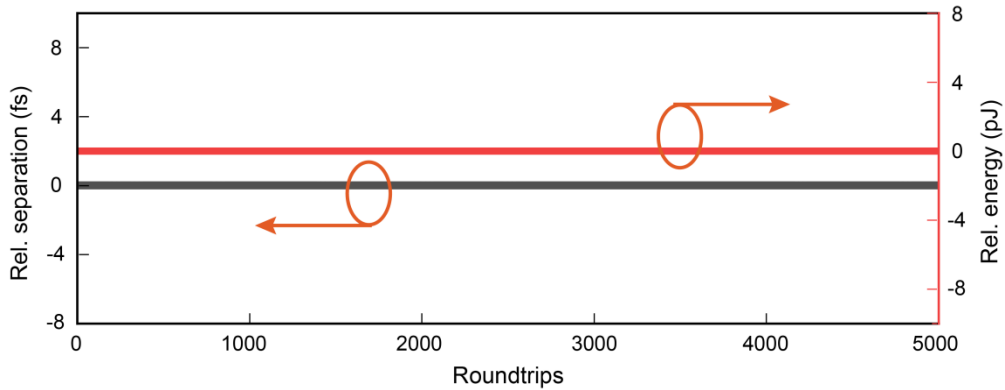
### (A) The ultrafast fiber laser model used to numerically confirm the resonant excitation of soliton molecules observed in experiments

We employ the generalized nonlinear Schrödinger equation (NLSE) to numerically reproduce the soliton molecule dynamics observed within the ultrafast fiber laser, aligning with the experimental findings:

$$\frac{\partial A}{\partial z} + \frac{i}{2} \left( \beta_2 + ig \frac{1}{\Omega_g^2} \right) \frac{\partial^2 A}{\partial \tau^2} = \frac{g}{2} A + \frac{\beta_3}{6} \frac{\partial^3 A}{\partial \tau^3} + i\gamma |A|^2 A \quad (\text{S1})$$

Where term  $A$  represents the field envelope,  $z$  is the propagation coordinate, and  $\tau$  corresponds to the fast time in the reference frame moving with the pulse. The parameters related to fiber dispersion and nonlinearity are selected from the Corning SMF-28 and Liekki Er 110-4. The saturated gain of the erbium-doped fiber with a gain bandwidth of 50 nm is defined as  $g = g_0/(1 + \int |A|^2 d\tau / E_{\text{sat}})$ , where  $E_{\text{sat}}$  is the saturation energy. For small-signal gain coefficient  $g_0$ , we apply a straightforward sinusoidal modulation  $g_0 = g_{0s}(1 + M_f \sin(2\pi f R_{\text{ts}}))$ , where  $R_{\text{ts}}$  is the laser roundtrips and  $g_{0s}$  refers to the small-signal gain coefficient corresponding to the initial stationary soliton molecules.  $M_f$  is the modulation depth, and  $f$  is the driving frequency.

A power-dependent transmittance function is adopted to model the ideal fast saturable absorber (SA) effect  $T = 1 - \alpha_{\text{ns}} - q_0(1 + P/P_{\text{sat}})^{-1}$ , where  $q_0 = 35\%$  is the modulation depth,  $\alpha_{\text{ns}} = 8\%$  is the non-saturable loss,  $P = |A|^2$  is instantaneous power, and  $P_{\text{sat}} = 50\text{W}$  is saturation power. In the simulation, the soliton molecule state with exceptionally stable mode-locking, characterized by extremely weak variations of intrapulse separation and energy fluctuations (see Fig. S1), is used as the initial condition. By keeping all other parameters constant and varying only the modulation depth  $M_f$ , we investigate the nonlinear response of the intrapulse separation of soliton molecules under different driving strengths.



**Figure S1.** Stability monitoring of the stationary soliton molecule (without modulation). The black curve shows the variation in intrapulse separation, while the red curve represents the variation in pulse energy, both demonstrating excellent stability.

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## (B) The Duffing model and fitting the Backbone curve of Soliton Molecules

The Duffing equation is widely used to describe phenomena such as resonance frequency shifts, bifurcations, and hysteresis in nonlinear systems, making it fundamental for understanding the dynamics of systems ranging from mechanical resonators to optical systems:

$$d^2x/dt^2 + 2\zeta\omega_0 dx/dt + \omega_0^2 x + \frac{k_3}{m_{\text{eff}}} x^3 = \frac{F}{m_{\text{eff}}} \quad (\text{S1})$$

where  $x(t)$  represents the displacement of the oscillator from its equilibrium position.  $m_{\text{eff}}$  is the effective mass,  $\omega_0$  is the eigenfrequency,  $\zeta$  is the linear damping coefficient. The external driving term is  $F=F_0\cos(\omega t)$ . The nonlinear term  $k_3x^3$  represents the cubic nonlinear stiffness (Duffing nonlinearity). Using a slowly varying amplitude  $r(t)$  and phase representation  $\varphi(t)$  for the displacement  $x(t)$ :

$$\begin{aligned} x(t) &= r(t) \cos[\omega t + \varphi(t)], \\ \dot{x}(t) &= -\omega r(t) \sin[\omega t + \varphi(t)] \end{aligned} \quad (\text{S2})$$

By substituting Equation S2 into the equation of motion and applying the method of averaging to eliminate the fast-oscillating terms. Essentially, assuming that  $r(t)$  and  $\varphi(t)$  vary much more slowly than the oscillation period  $2\pi/\omega$ , allowing us to average over one oscillation cycle:

$$\begin{aligned} \dot{r} &= \frac{F_0}{2m_{\text{eff}}\omega_0} \sin(\varphi) - \frac{\zeta_1}{2m_{\text{eff}}} r, \\ \dot{\varphi} &= -\frac{F_0}{2m_{\text{eff}}\omega_0 r} \cos(\varphi) + \frac{3k_3}{8m_{\text{eff}}\omega_0} r^2 \end{aligned} \quad (\text{S3})$$

To find the steady-state solution, we set  $\dot{r}=0$ ,  $\dot{\varphi}=0$  and eliminate  $\varphi$ , and the equation of the amplitude  $r$  could be obtained:

$$(\omega_0^2 - \omega^2)r + \frac{3k_3}{4m_{\text{eff}}} r^3 = \frac{F_0}{m_{\text{eff}}} \quad (\text{S4})$$

Considering that the cubic nonlinearity coefficient  $k_3$  is small, we can approximate the frequency shift due to the amplitude  $r$ . This gives:

$$\omega = \omega_0 + \frac{3k_3}{8m_{\text{eff}}\omega_0} r^2 \quad (\text{S5})$$

Equation S5 shows that the resonance frequency  $\omega$  depends on the oscillation amplitude  $r$ , and is applied to the nonlinear fitting of the Backbone curve for soliton molecules. In this case,  $k_3$ ,  $m_{\text{eff}}$ , and  $\omega_0$  are constants, while  $r$  corresponds to the oscillation amplitude of the intrapulse separation of soliton molecules.