Editors' Suggestion

Superlocalization Reveals Long-Range Synchronization of Vibrating Soliton Molecules

Said Hamdi, Aurélien Coillet, Benoit Cluzel, Philippe Grelu, and Pierre Colman*
Laboratoire Interdisciplinaire Carnot de Bourgogne UMR CNRS 6303, Université Bourgogne–Franche-Comté, 9 avenue Savary, 21000 Dijon, France

(Received 24 January 2022; accepted 2 May 2022; published 27 May 2022)

We implement a superlocalization method in the time domain that allows the observation of the external motion of soliton molecules in a fiber ring cavity laser with unprecedented accuracy. In particular, we demonstrate the synchronization of two oscillating soliton molecules separated by several nanoseconds, with intermolecular oscillations following the same pattern as the intramolecular motion of the individual molecules. These experimental findings indicate an interplay between the different interaction mechanisms that coexist inside the laser cavity, despite their very different characteristic ranges, timescales, strengths, and physical origins.

DOI: 10.1103/PhysRevLett.128.213902

Introduction.—Recently, the dynamics of optical soliton molecules (SMs) in ultrafast lasers has attracted increasing attention [1]. SMs are interacting solitons that form a bound state [2], which can exhibit periodic internal motions similar to the vibrations of molecules in chemistry Previous real-time experiments highlighted different oscillatory behaviors, such as vibrations, phase-only, or anharmonic oscillations [3–5]. The specific pattern depends on the laser parameters, namely, the existence of a limit-cycle attractor [6] combined with the laser noise [7] and other experimental perturbations [8–11]. Beyond their fundamental appeal, SMs could be involved in harmonic mode locking or in optical data manipulation [1,2]. Therefore, experiments now investigate SM dynamics for a larger number of interacting solitons. In a significant number of cases, solitons are not distributed in a regular train of pulses but form a supramolecular complex composed of several, often identical, molecules [10,12,13]. This raises the open question of the interplay between the mechanisms responsible for the dynamics governing each molecule and for the macro-organization of the complexes.

The dispersive Fourier-transform (DFT) technique [14] allows the single-shot real-time recording of the spectra over successive cavity round-trips at multi-MHz frame rates. After numerical processing, DFT spectra yield the dynamics of relative distance and phase between pulses that are constituents of the SM. As a major limitation, the observation window's T_{obs} is limited to typically 100 ps [8–10,12]. Such a practical limitation is bound on one side by the speed of the detection electronics and on the another side by the length of the dispersive line used for pulse stretching. Indeed, the need to avoid any overlap of the DFT traces for two subsequent pulses imposes a maximal value to the former. As a rule of thumb, an electronic bandwidth of ∼10 GHz and a repetition rate of ∼10 MHz for ∼200 fs pulses yields $T_{\mathrm{obs}} \sim 100$ ps. In addition, the DFT does not provide any information regarding the global motion of the molecule around the laser cavity. The direct observation, i.e., without pulse stretching, using a GHz-bandwidth oscilloscope provides a temporal resolution only down to a few tens of picoseconds. Such a resolution is not enough to observe the weak timing fluctuations that are expected to take place within the supramolecule. Indeed, the strongest mechanisms have the shortest range of interaction [1], while long-range interactions are much weaker [7,15,16]. Consequently, observing the latter would require acute measurements precision, about on par with the resolution provided by the DFT, but with much larger observation windows.

To do so, we have implemented on the timing channel a superlocalization procedure akin to what is done in fluorescence spectroscopy [17–19]. Thus, we got a timing resolution down to 0.14 ps, 100 times the native sampling resolution. Subtle changes of the molecules' cruising velocity can now be observed. We applied this new technique to the coevolution of two soliton-pair molecules [5], which are separated by ≃7.6 ns, one third the round-trip time (23.0 ns, FSR = 43.4 MHz). For a clear demonstration of the possibilities offered by combining short- and long-timescales measurement, two soliton-pair molecules indeed make the simplest multimolecular system.

Internal motion: Twin molecules.—The fiber ring laser cavity is composed of 1-m erbium-doped fiber closed by 3 m of single-mode fiber (SMF) [see Fig. 1(a)]. Nonlinear polarization evolution in the fibers followed by discrimination through a polarizing beam splitter (PBS) result in a nonlinear transfer function that is responsible for the mode locking. Experimental data are recorded by means of a 35-GHz photodiode connected to a 40-GHz bandwidth 80-GS/s oscilloscope. The first channel records the DFT spectrum after a chromatic dispersion of −49 ps/nm. It yields the information about the internal motion for each

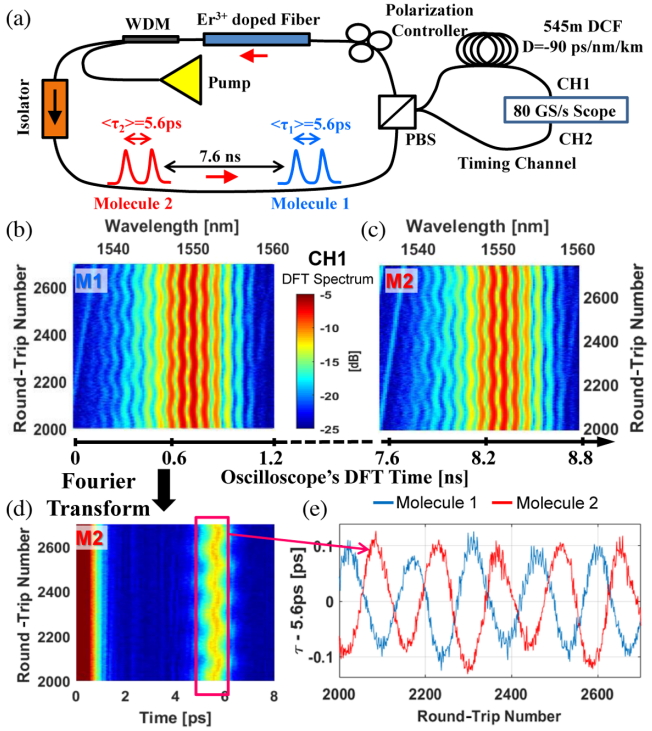

FIG. 1. (a) Experiment setup: PBS, polarizing beam splitter; HDF, 545-m highly dispersive fiber ($D = -90.15$ ps/nm/km); pump, 980-nm laser diode; WDM, pump-signal multiplexer. Single-shot spectra in logarithmic color scale for (b) the leading and (c) the trailing SM. Knowing the time of passage of each molecule, the oscilloscope time (lower axis) can be converted into wavelength (top axis). (d) Temporal autocorrelation traces after Fourier transform of (c). (e) Internal vibration motion for each molecule. Blue (red) line stands for the leading (trailing) molecule. They both exhibit an average soliton separation of 5.57 ps.

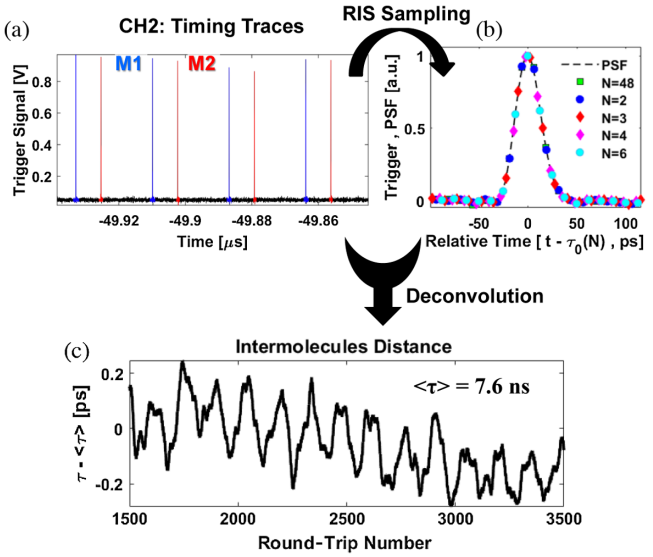

FIG. 2. (a) Timing trace: one peak per molecule and per RT. (b) Point spread function for $M1$: the random interleaved sampling (RIS) between the cavity RT time and the oscilloscope's sampling clock is shown for a few pulses. Dashed black: reconstructed PSF after averaging over 4221 RTs. (c) Evolution of the distance between the molecules ($M1$, $M2$) with sub-20-fs resolution. The average intermolecular separation is $\langle \tau_{\text{ext0}} \rangle = 7.58$ ns.

molecule. The second channel records the direct pulsed laser output to track down the global motion of the molecules around the cavity. It reveals the presence of two sets of pulses, and the DFT signal confirms that both sets are actually stable pairs of solitons separated by 5.57 ± 0.01 ps.

The internal vibration of each SM is obtained by Fourier transform of the DFT signal, as illustrated by Figs. 1(b)–1(d). In Fig. 1(e), the two molecules both follow a common periodic oscillation with an amplitude of 92 ± 6 fs and a periodicity of 143.5 round-trips (RTs). The correlation coefficient is 0.89 for a relative delay of 87.2 RTs. This strong (anti)correlation means that the two oscillations belong to the same family minus some drifts, as discussed later in this Letter. Since the existence of SMs is fixed by a common dissipative attractor [6], it is not surprising that the two pairs exhibit similar features. However, the two molecules are not isolated physical objects. Instead, they are likely to interact, as the existence of weak long-range (ns) interactions has been established [7,16,20]. Therefore, one could wonder whether the two molecules following a common vibration pattern would benefit from an additional synchronization mechanism. In theory, several distinct vibration patterns may coexist [21–23].

External motion: Point spread function deconvolution.— To investigate this matter further, we improved the native 12.5-ps (80-GS/s) sampling resolution by performing a point spread function (PSF) deconvolution, similar to what is done in fluorescence microscopy to achieve spatial superresolution [17–19]. In a nutshell, by superposing together the pulses acquired at each round-trip, the aliasing between the cavity-free spectral range and the oscilloscope's internal sampling clock performs a random interleaved sampling; hence, the shape of the PSF is retrieved with a tenfold improvement in temporal resolution [Fig. 2(b)]. Note that the PSF is defined using a self-consistent procedure so that a perfectly recurring and stable signal is not required [24]. Once the high resolution PSF is obtained, the time of passage of each pulse can be determined with a precision down to 140 fs. The residual uncertainty is due to a mixture between the oscilloscope's and laser intrinsic jitters (see Supplemental Material [25] for more details).

The advance or delay for each molecule compared to the average round-trip time is shown in Fig. 2(c). The most striking feature is that the intermolecular distance oscillates, following a pattern very similar to the internal motion: same periodicity and nearly the same amplitude (106 ± 18 fs versus 92 ± 6 fs). Similar to the internal

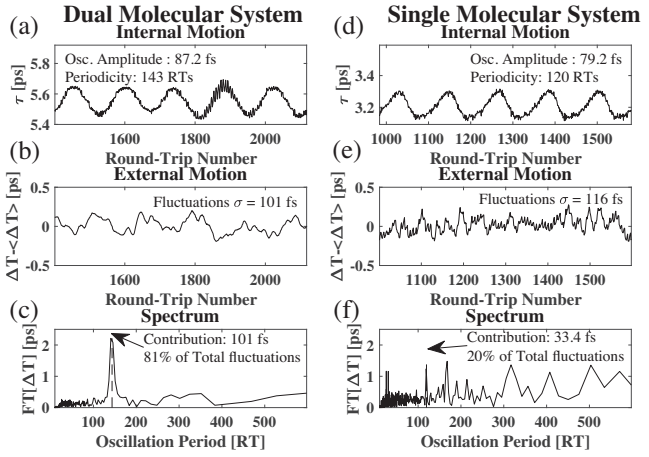

FIG. 3. (a) $M1$ Internal motion. (b) Intermolecular distance vs the round-trip number. (c) Fourier transform of (b): the dashed line indicates the period of the internal vibration. (d)–(f) Same as (a)–(c), but typical results for a single-molecule system.

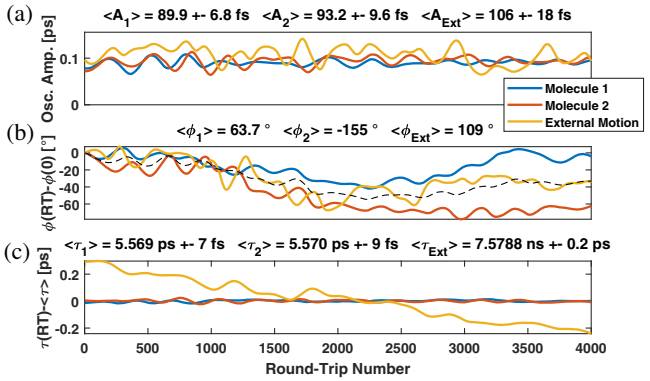

FIG. 4. (a) Instantaneous oscillation amplitude for molecules $M1$ (A_1), $M2$ (A_2), and for the intermolecular oscillation (A_{ext}). (b) Similar to (a) but regarding the relative phase of the oscillation. Black dashed: result of Eq. (1). (c) Evolution of the average soliton separation for $M1$ (blue) and $M2$ (red), and of the distance between the two molecules (yellow).

motions, this external oscillation has its own phase offset. The lower electronic bandwidth of the photodiode and the oscilloscope (40 GHz) acts as a low pass filter that filters out the subpicosecond features of the molecule. Therefore, the time of passage measured here corresponds to the time of passage of the molecule's center of mass, and it is not too sensitive to the internal motion of the molecule. As seen in Fig. 3(e), the vibration of a molecule is neither sensitive to its global motion nor coupled to it.

To understand better the specificity of this intermolecular oscillation, we compared in Fig. 3 the dynamics of the dual-molecule system with that of a single molecule recorded from the same laser setup. Both dynamics show nearly the same internal oscillatory motion [Figs. 3(a)–3(d)]. Using Parseval's theorem, most of the external motion (81%) of the two-molecule systems is explained by an oscillation tuned to the internal vibration state. On the contrary, for the single-molecule system the trajectory is mainly composed of noise fluctuations which are not related to the internal vibration. The same goes for the global drift of the two molecules (see Supplemental Material [25]). There does not seem to exist any coupling between the internal oscillations and fluctuations of the round-trip time. The remaining 20% contribution synchronized to the internal oscillation remains hypothetical: a small influence of the molecule dynamics on its own velocity, or a periodic exchange of energy between the two solitons that would cause an apparent motion of its center of mass [26,27]. Indeed, a vibrating SM is not a simple two-parameter oscillator [27]. However, the comparative analysis of the single-molecule systems rules out this phenomenon as being the cause of the external motion observed for the two-molecule systems.

Long-range interaction and synchronization.—Clues regarding any interaction between the different oscillators can be inferred from the oscillators' response to fluctuations. Indeed, since the oscillators are separated by several nanoseconds, they do not experience the same random noise. Hence, fully independent and noninteracting oscillators would slowly drift from each other following a random walk pattern.

For slow and weak fluctuations (adiabatic regime), the motion $\tau(t)$ of a harmonic oscillator around its equilibrium position τ_0 can be described as $\tau(t) = \tau_0(t) + A(t)\cos[\omega t + \phi(t)]$, where τ_0, A, and ϕ are the equilibrium position, the oscillation amplitude, and the phase offset, respectively. These parameters fluctuate under the action of noise and drift of the lasing conditions. Therefore, it is possible to get a finer description of the dynamics by monitoring the evolution of the latter three parameters, as shown in Fig. 4.

First, the internal oscillation is more stable that the external one Fig. 4(a). Noting the small temporal extension of each SM (< 7 ps here), the moderate amplitude excursions may result from a stronger stabilizing interaction. Indeed, dissipative solitons are subjected to phase-sensitive short-range interactions [28,29]. This strong binding attenuates the impact of noise. In addition, the impact of noise depends on the separation between the oscillator's constituents: It filters out lower frequency that then impacts the molecule globally (drift) but not its internal motion. The weaker interaction and greater sensitivity to low frequency noise explain the faster drift observed for the external oscillator, as displayed on Fig. 4(c). That said, considering its spatial extension, the external oscillator exhibits relative fluctuations that are proportionally much smaller.

Long-range (nanosecond) interactions are very difficult to model precisely in fiber lasers because they can combine physical effects such as gain depletion and recovery, electrostriction, and random walk on a noise floor [7,15,16,20]. In contrast with short-range interactions,

long-range effects are much weaker. In addition, they are sensitive to the optical intensity but not to the relative optical phase between them. For close-by solitons, the relative optical dephasing is an important feature that controls the attraction and repulsion, hence, the molecule's vibration. *A priori*, there does not exist any mechanism that would control the relative optical phase offset between the very distant molecules. However, a crucial point here is that the internal motion of the molecule is an oscillator that possesses its own phase, which is of different nature from the optical phase while still being closely related to it. Therefore, it is still possible that long-range interactions become sensitive to the phase of the SM oscillators. Some indications of the possible synchronization between the three oscillators can be found in Fig. 4(b) that features the evolution of the relative phase offset $\phi(RT) - \phi(0)$. As the two molecules evolve, their respective internal motions experience different phase fluctuations and drift away significantly from each other. When it comes to the external motion, we see that its phase evolution is similar to the one that would experience a simple springlike oscillator when excited at both ends by two oscillatory forces that would derive from the internal motion. Basically, the phase evolution of such a spring oscillator would then satisfy

$$\phi_{\text{spring}}(t) = \frac{\phi_1(t) + \phi_2(t)}{2} + \delta, \tag{1}$$

where $\phi_1(t)$ and $\phi_2(t)$ are the relative phases of the two driving forces, which we assume here to be the relative phases of the internal motion for each molecule. δ is a constant phase offset that accounts for a delayed response to the external driving taken here as $\delta \simeq 20°$ (equivalent to an offset of eight RTs), which yields the dashed line in Fig. 4(b). Therefore, the present data analysis indicates that the external oscillation is most likely driven by the combination of the internal motions of the SMs. Conversely, the internal oscillations being out of phase from each other [Fig. 1(e), and Fig. S3(c) in the Supplemental Material [25] for the second set of SMs] seems to indicate the existence of an underlying synchronization mechanism impacting the internal degree of freedom of the two molecules.

Discussion and conclusion.—The above analysis is limited by the number of recorded round-trips (4221 RTs), itself limited by the oscilloscope memory, so that we cannot give a definite statement regarding the strength of the synchronization between the internal and external motions of the SMs. Nevertheless, the progress of our argument in that direction is made possible by the super-localization techniques and refined data analysis which shed new light on the complex nonlinear dynamics of multiple SMs. Therefore, by presenting such tools and approach, our Letter should stimulate further experiments aiming to explore the dynamics of soliton molecular complexes with acute precision. We have also analyzed a second experimental dataset, namely consisting of another set of oscillating soliton-pair molecules, which is presented in the Supplemental Material [25]. Endowed with similar but not identical features, it confirms the existence of an external oscillation relative to the separation between the SMs. It also leaves the possibility for synchronization between the external oscillator and the internal ones.

At this point, it is essential to discuss more details of the SMs dynamics. Occurrences of multiple SM systems have been previously reported in the literature [12,30]. Studies focus either on the binding mechanisms controlling the equilibrium position between the solitons [9,10,30] or on the internal molecules' dynamics and their possible interaction [12]. A second distinction concerns the type of interactions that are involved. Indeed, they can be sensitive or not to the optical phase; hence, they have different attractive landscapes. Therefore, depending on the interactions, completely different dynamics are expected. In a nutshell, multimolecules systems are characterized by at least two main features: the macroscopic organization of the molecules and their internal dynamic. It is indeed important to distinguish between, on one side, the adiabatic adaptation of the equilibrium positions to an external change of the laser parameters [9], and on the other side, the intrinsic dynamics that occurs at constant laser parameters. Only the latter can be considered a true oscillator. Thus far, the intrinsic limit of the DFT technique, as discussed in the Introduction, limited the studies to situations where the supramolecular cohesion is governed by the same interactions as for the molecules binding: short-range interactions. Moreover, these are strong coherent interactions, which lead to a perfect locking of the molecules [12]. In this Letter, the long-range organization of the molecules, which are separated by one third of the cavity round-trip, is well ascribed by acoustic and opto-mechanical interactions [31,32]. These interactions are of different nature from a direct soliton interaction as they are not sensitive to the optical phase. Nevertheless, we have shown that they may be sensitive to the oscillator's phase and thus create a link between the internal motions. Such an exchange of information could, in particular, result in the two molecules having a preferable specific phase relationship, as it is observed here with anticorrelation between the internal vibrations states. Note that if the acoustic interaction is most likely responsible for the relative position between the two molecules, it may not necessarily be the mechanism involved in their relative synchronization. For example, the exchange of dispersive waves, albeit very weak, could also play a synchronization role since it is sensitive to the relative phase between solitons. Indeed, the synchronization effect is very weak, in particular, with regard to the laser noise, but it can accumulate over numerous round-trips [32] and hence lead to noticeable effects: The two molecules have the same oscillation

pattern. The direct observation of the said synchronization mechanism requires, however, an extremely precise timing with subpicosecond resolution.

To conclude, we have shown that SMs influence each other over the long range. Despite being separated by several nanoseconds, they cannot be considered independent objects. Interestingly, long-range interactions do not only determine the relative position of the molecules, but they can also serve as media to exchange dynamical information by coupling the internal degrees of freedom together. Despite the fact that such a possible synchronization will remain weak and cannot give rise to a strict locking, it is an open question whether different molecules could share the same vibration properties and can thus drive each other resonantly. Obviously, the mechanisms involved here would also be present in harmonically mode-locked fiber lasers [10,33].

The point spread function deconvolution that we have implemented is critical in order to unveil such subtle interactions, as this requires a very precise timing resolution below 300 fs. This technique provides new possibilities to study the dynamics of multipulse systems in fiber ring cavity lasers [10]. As a pure numerical postmeasurement processing, this deconvolution technique is easy to implement. Note that the final timing resolution after deconvolution depends on the quality of the experimental implementation. As a practical example, in this Letter, we have managed a 2 order of magnitude improvement of the resolution.

Concerning the specific topic of SM, an instantaneous phase and amplitude analysis—as shown in Fig. 4—is essential to describe accurately the vibration patterns. In particular, it provides information about the fluctuations, which are directly related to the laser noise. Therefore, fluctuations of the vibrations reveal the intrinsic properties of the limit-cycle attractor in response to noise. Thus far, the description of SMs has been mostly qualitative, with scarce information regarding the stability of the molecules' motion. We hope the present work will serve as incentive for more thorough and quantitative analysis of SM's vibration patterns.

This work was supported by the French "Investissements d'Avenir" program project ISITE-BFC (Contract No. ANR-15-IDEX-0003) and by the Agence Nationale de la Recherche project "CoMuSim" (Contract No. ANR-17-CE24-0010-01). The authors declare no conflicts of interest.

*Corresponding author.
pierre.colman@u-bourgogne.fr

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