Instabilities with shear-thinning polymer solutions in the Couette-Taylor system

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Abstract. We have investigated the stability of viscoelastic Couette-Taylor flow induced by rotating only the inner cylinder in the case of a semi-dilute solutions of high molecular weight polyethylenoxide (Aldrich, 8·10^6 g/mol) in a 95% water / 5% isopropyl alcohol solvent mixture. The saturated instability mode appears in form of coupled counterpropagating spirals, which is a combination of propagating spirals and some harmonic modes. These modes can be described using an extended version of complex Ginzburg-Landau equations.

1. Introduction

The addition of small amounts of flexible polymer, with long linear chain, to a Newtonian liquid gives to the resulting solution a viscoelastic behavior. Among the various features of non-Newtonian behaviors observed with such solutions, turbulent drag reduction is one that has attracted much attention. Although this effect was discovered by Toms in 1949, there is no satisfactory theory available for its explanation. Research on the drag reduction problem requires the joint study of turbulence (which is not fully understood itself), and of viscoelastic flows, for which the lack of universal equations (such as the Navier Stokes equations for Newtonian flows) is a great drawback. Drag reduction, however, has important applications, such as lowering petroleum transportation cost in pipelines. These facts advocated the need for studies of viscoelastic flows in hydrodynamic model systems such as the Couette-Taylor system, for which various routes to turbulence have been identified for Newtonian flow [1]. This interest is reinforced by the increasing use of the Couette-Taylor system in biologically oriented apparatus with non-Newtonian flows, such as blood anticoagulant devices [2], or bioreactors, where cultures of biological cells benefit from low shear rate and mixing properties [3].

Experimental studies of the viscoelastic Couette-Taylor flow reported various results depending on the fluid rheological properties [4]. For solutions with low elasticity, inertia is the main driving force and, with the inner cylinder rotating, the circular Couette flow bifurcates to Taylor Vortex Flow, with moderate quantitative changes of the main characteristics such as the instability onset and wavenumber, as for instance in the work of Yi and Kim [5]. It should be highlighted that, by combining theoretical and experimental results,
Yi and Kim have been able to estimate the so-called second normal stress difference, a fluid rheological property which is difficult to measure with standard rheometric systems.

For Boger fluids, which are very elastic and very viscous fluids, Larson et al. [6] showed that the base circular Couette flow could bifurcate to a nonstationary “purely” elastic instability mode. Such a mode is triggered by the elastic effect only. It can be observed for vanishing Reynolds number, with only rotating the inner cylinder as well as with only rotating the outer cylinder. The characteristic time scale that governs elastic regimes is the polymer relaxation time. Further experimental investigations of elastic instability were done by Groisman and Steinberg [7], who confirmed key theoretical predictions and showed the progressive transition from inertial regime, governed by the viscous diffusion time, to elastic regime, governed by the elastic relaxation time. Experimental results with Boger fluids led, as an interesting side effect, to the identification of “thermoelastic” instabilities [8], for which viscous heating plays the main role in flow destabilization, with slower instability growth than for other regimes.

For solutions with intermediate elasticity, a purely elastic instability regime is not triggered but the critical instability mode may be qualitatively different from what would be expected in a Newtonian fluid. In such a case, the instability mode is usually referred to as “inertio-elastic”, as it results from the interplay between inertia and elastic effects. Of particular interest are flow patterns made of counterpropagating spirals observed in viscoelastic Couette-Taylor flow with only the inner cylinder rotating. For a Newtonian fluid, spirals are observed in the case of sufficiently counterrotating cylinders. For polymer solutions, counterpropagating spirals, named Rotating Standing Waves (RSW), were observed by Groisman and Steinberg [9][10], Baumert and Muller [11], Crumeyrolle et al. [12], with inner rotating cylinder only using different solutions. Groisman and Steinberg observed RSW as a second instability mode or as a critical mode, depending on the fluid elasticity. They reported for RSW a well-defined chess-board like flow pattern at onset. This can be depicted more generally as a pattern exhibiting a staggered-rows organization. Baumert and Muller also observed staggered-rows patterns. With aqueous solutions of polyethyleneoxide, Crumeyrolle et al. [12] have observed a flow pattern made of strongly coupled counterpropagating spirals. With a similar visualization technique to that of previous studies, strongly coupled counterpropagating spirals lead to a different, grid-like, flow pattern.

The present study provide further results on the interactions of coupled counterpropagating spirals in the case of a semi-dilute solutions of high-molecular-weight polyethyleneoxide (Aldrich, 8·10^6 g/mol) in a solvent mixture made of 95% water / 5% isopropyl alcohol in volume. These results can be interpreted in the framework of coupled Complex Ginzburg-Landau Equations (CGLE) with resonant harmonic terms. Amplitude equations such as CGLE have proven to be useful, in particular close to instability onset, to model patterns exhibited by complex physical systems for which a mathematical description is difficult or not known precisely. The paper is organized as follows: the experimental setup is described in section 2, while results are reported in section 3. The section 4 introduces CGLE model and section 5 contains concluding remarks.

2. Experimental setup

The experimental setup consists of a Couette cell with coaxial horizontal cylinders. The inner cylinder is made of black Delrin with a radius \( a = 4.46 \) cm. The outer cylinder is made of Plexiglass with a radius \( b = 5.05 \) cm. The gap size between the cylinders is \( d = b - a = 0.59 \) cm and has a length \( L = 27.5 \) cm. The radius ratio is \( a/b = 0.883 \) and the aspect ratio is \( \Gamma = L/d = 46.6 \). The outer cylinder is fixed, while the inner cylinder is driven by a DC servomotor at the angular frequency \( \Omega \) (the experimental control parameter). Alternatively, we use the imposed shear rate \( \dot{\gamma} = \Omega a/d \) as the control parameter. We define \( e = (\Omega - \Omega_c)/\Omega_c = (\dot{\gamma} - \dot{\gamma}_c)/\dot{\gamma}_c \) as the criticality parameter, where \( \Omega_c \) is the critical rotation speed for which the base flow bifurcates and \( \dot{\gamma}_c = \Omega_c a/d \) the corresponding critical value of the imposed shear rate.
Polymer solutions were prepared by mixing an initial suspension of polyethyleneoxide (PEO, Aldrich, \(8 \times 10^6\) g/mol) in 40 ml of isopropyl alcohol with 760 ml of water. The concentration of PEO in the resulting solution is 500 weight ppm. The solution is maintained 5 days at rest at 4°C then approximately 14 h at room temperature. A final homogenization stage is performed with a magnetic shaker. Viscosity measurements with an AR2000 rheometer (TA Instruments) exhibited a shear-thinning behavior (figure 1). Such behavior is well described by the Carreau law

\[
\eta = \eta_0 \left[1 + (\lambda \dot{\gamma})^2\right]^{\eta/2},
\]

where \(\eta_0\) is the shear viscosity for vanishing shear-rate, \(\lambda\) a characteristic time and \(n\) the power law index for the shear-thinning behavior. Numerical values fitted from experimental data are given in table 1.

![Figure 1](image)

**Figure 1.** Shear viscosity of polymer solution for \(c = 500\) ppm of PEO in 95% water / 5% isopropyl alcohol.

**Table 1.** Carreau characteristics of the polymer solutions (PEO in 95% water / 5% isopropyl alcohol) obtained from the shear viscosity measurements.

| \(c\) (ppm) | \(\eta_0\) (mPa·s) | \(\lambda\) (s) | \(n\) |
|------------|----------------|---------------|------|
| 500        | 5.13           | 0.12          | 0.15 |
| 600        | 7.40           | 0.16          | 0.19 |
| 700        | 10.2           | 0.29          | 0.21 |

For flow visualization, 2% of Kalliroscope AQ 1000 (Kalliroscope Corp., MA, USA) has been added. Kalliroscope AQ 1000 is actually a dilute suspension of highly anisotropic reflective platelets in a mixture of propylene glycol and water. No significant change of the solution viscosity due to these platelets was observed. A linear 1024-pixels CCD camera (iDC 100, i2S, France) recorded the reflected light intensity \(I(x)\) from a line along the axial direction with 8-bits sampling. Recorded lines at regular intervals (0.2 s) are stacked together to provide space-time diagrams \(I(x,t)\) of flow patterns. Total acquisition time can last 27 minutes, over a length of 23 cm.

Space-time diagram processing is based on 2 dimensional Fourier analysis and complex demodulation. The aim of complex demodulation (Hilbert transform) is to extract from the real space-time signal the complex amplitude and phase of the modulations of a particular mode. This is more conveniently expressed with Fourier transforms. Let us assume that the 2d
spectrum \( \hat{I}(q,\omega) \) of \( I(x,t) \) exhibits a non-monochromatic peak. This peak is described as the result of the modulation of a monochromatic mode \( \exp(i(q_0 x - \omega_0 t)) \) by a low-frequency modulation \( M(x,t) \). The space-time signal \( I(x,t) \) is then \( I(x,t) = M(x,t) \cdot \exp(i(q_0 x - \omega_0 t)) \).

The Fourier convolution theorem states that \( \hat{I}(q,\omega) = \hat{M}(q,\omega) \ast \delta(q - q_0, \omega + \omega_0) \), the spectrum \( \hat{I}(q,\omega) \) is made of the spectrum \( \hat{M}(q,\omega) \) translated at \( (q_0, -\omega_0) \) in the 2d spectrum \( \hat{I}(q,\omega) \). From \( \hat{I}(q,\omega) \), Fourier filtering around a peak of interest followed by Fourier inverse transform allows to recover the complex quantity \( I(x,t) = A(x,t) \exp(i\varphi(x,t)) \), from which space-time diagrams for amplitude \( A(x,t) \) and phase \( \varphi(x,t) \) can be obtained. The derivatives of the phase with respect to space and time give the local frequency and wavenumber of the pattern: \( q = q_0 + \frac{\partial \varphi}{\partial x}, f = f_0 + \frac{1}{2\pi} \frac{\partial \varphi}{\partial t} \).

3. Results

We present results obtained near onset for \( c = 500 \text{ ppm} \). Similar results have been obtained for \( c = 600 \text{ ppm} \) and \( c = 700 \text{ ppm} \). The first instability from the base circular Couette flow occurs at \( \Omega = 0.372 \text{ Hz} \) via a Hopf bifurcation giving rise to a pattern of counterpropagating spirals which are separated in space (figure 1). For \( \varepsilon = 0.003 \), the space-time diagram (figure 2) exhibits a rich dynamics. In fact, one can distinguish zones of strongly coupled counterpropagating spirals near the left and right edges, zones of left traveling spirals and zones of right traveling spirals. It must be highlighted that the system can be slowly driven back below the onset and then slowly forward again above the onset without significant hysteretic behavior, thus the transition is a supercritical Hopf bifurcation.

The Fourier 2d power spectrum exhibits definite peaks (figure 3), corresponding to the left- and right-propagating spirals are located at \( (-q, \omega) \) and \( (q, \omega) \) respectively. Second harmonic modes \( (-2q, 2\omega) \) and \( (2q, 2\omega) \) propagate with the same phase velocity as the fundamental modes. The spectrum exhibits, aside from these peaks, two large peaks corresponding one to a stationary spatial harmonic mode \( (2q,0) \), and the other to a temporal homogeneous harmonic mode \( (0,2\omega) \). These unlocked harmonic modes result from a multiplicative coupling between right and left spirals [12]. Locked harmonics \( (\pm 2q, 2\omega) \) can be ignored in the following as their intensity in the power spectrum is relatively small.

![Figure 1](image1.jpg)  
**Figure 1.** Space-time diagram of the observed regime at the onset of instability.  

![Figure 2](image2.jpg)  
**Figure 2.** Space-time diagram of the observed regime just above onset (\( \varepsilon = 0.003 \))
Figure 3. Two-dimensional power spectrum of: a) the critical pattern (figure 1), b) the pattern just above onset (figure 2). Colour scale: power (arbitrary units). Powers for left modes ($q < 0$) and right modes ($q > 0$) have been added for convenience.

Therefore the pattern signal can be represented as follows

\[
I(x,t) = \text{Re} \left\{ A(x,t) \exp(i(qx - \omega t + m \theta)) + B(x,t) \exp(-i(qx + \omega t - m \theta)) + U_{\omega}(x,t) \exp(-2i(\omega t - m \theta)) + U_{q}(x,t) \exp(i(2q x)) \right\}
\]

(1)

where $A$, $B$, $U_{\omega}$, and $U_{q}$ are the complex amplitudes of the relevant modes. For each of these amplitudes, we have introduced a spatial profile as the time-averaged value, e.g.

\[
\langle A \rangle_{t}(x) = \frac{1}{T} \int_{0}^{T} A(x,t) dt
\]

where $T$ is the total acquisition time. Signal acquisition was performed at fixed value of $\theta$, so that we have ignored the angular dependence in our study.

Fourier filtering allows inspection of the space-time diagram of each mode (figure 4) and complex demodulation gives space-time diagrams of the corresponding amplitudes (figure 5). These diagrams show clearly that a right-propagating spiral largely fills the right part of the flow system and only a small boundary region of the left part. Symmetric behavior was observed for the left-propagating spiral, which fills a large part on the left of the system and only a small boundary region of the right part. In this case, propagating modes cover each other only in the two limited region near the boundaries, while a clear split occurred in the middle of the system for propagating modes. From figure 4, it appears that on a given part of the system (left or right), the two propagating modes are out of phase. Such phase shift is also observed for a single propagating mode (left or right) when the patterns for left and right part of the system are compared.
Figure 4. Individual space-time diagram for fundamental modes and unlocked harmonics obtained after the Fourier filtering ($\varepsilon = 0.003$.)
Figure 5. Space-time diagrams of mode amplitude moduli after complex demodulation ($\varepsilon = 0.003$.)

Figure 6. Time-averaged amplitude moduli of the modes ($\varepsilon = 0.003$.)
The spatial profiles of averaged amplitudes over the full acquisition time (27 min) are shown in figure 6a. The amplitude for propagating modes vanishes in the center of the system, where harmonic modes dominate (figure 6b).

The harmonic mode patterns (figures 4-5) exhibit slow modulations and amplitude holes (depression of amplitude in space and time). These amplitude holes occur in a regular fashion, leading to a well-localized depression in the time-averaged amplitudes (figure 6b). No perturbations were observed, however, on the propagating modes. From the space-time distribution of the phase fields, we have extracted the spatial distribution of frequency that is reported for fundamental modes in figure 7a. This frequency distribution shows that frequencies in the left and right parts of the experiment do not match each other. This leads to the low-frequency modulations of the harmonics and generate spatio-temporal dislocations due to collisions of two spirals with localized annulation of amplitude. The dislocations are well evidenced in the spatio-temporal phase diagram of unlocked harmonics (figure 7b).

**Figure 7.** ($\varepsilon = 0.003$) a) Variation of the fundamental frequency in space; b) Phase space-time diagram $\varphi = \text{Arg}(U_\omega(x,t)\exp(-2i\omega t))$.

**Figure 8.** Space-time diagram of the pattern for a) $\varepsilon = 0.008$; b) $\varepsilon = 0.048$. 
Figure 8 presents space-time diagrams of the flow pattern for increased values of the control parameter. Time-averaged amplitudes are reported for four values of $\varepsilon$ in figures 9. No split in the middle of the system is visible; the propagating modes are no longer separated. Amplitude holes are now observed in propagating modes, while only harmonics exhibited such feature at $\varepsilon = 0.003$. For $\varepsilon = 0.008$ the unlocked harmonics have approximately the same intensity; however, for $\varepsilon = 0.048$ intensity of spatial harmonic is lower than that of the temporal harmonic: when $\varepsilon$ increases, the time-averaged amplitude $\langle U_q \rangle_t$ decreases below $\langle U_\omega \rangle_t$ (figure 9).

![Figure 9](image)

**Figure 9.** Time averaged amplitude moduli of the modes.

We have observed that the slow dynamics of propagating modes associated to amplitude holes exhibits anticorrelation between left and right modes, as illustrated on figure 10. We have calculated the correlation function between left and right mode amplitude moduli as follows

$$ C_X(\tau) = \frac{\sum_{t=0}^{T-\tau} \left( \langle A \rangle_X(t) \cdot \langle B \rangle_X(t+\tau) \right)}{\left( \sum_{t=0}^{T-\tau} \langle A \rangle_X^2(t) \cdot \sum_{t=\tau}^T \langle B \rangle_X^2(t) \right)^{1/2}} $$

where $T$ is the full acquisition time and $\langle \cdot \rangle_X$ is the spatial average of the amplitude modulus on a space interval $X$ from which space-time mean (on the same interval $X$) was subtracted, that is $\langle \cdot \rangle_X = \langle \cdot \rangle_X - \langle \cdot \rangle_{X \times T}$. Anticorrelation was evidenced, with $C(0)$ around -0.7 (figure 11). We have observed for each correlation curve (figure 11a and 11b) that anticorrelation at $\tau$
0 is followed by a well-defined maximum of $C(\tau)$. The correlation times associated to these positive maxima are roughly $\tau = 75\, \text{s}$ on the left side of the system and $\tau = 100\, \text{s}$ on the right side of the system. Other larger correlation times are also visible. Similar behavior is observed from $\varepsilon = 0.005$ up to $\varepsilon = 0.048$. For higher values of the control parameter, disorder leads to different correlation functions, with smaller time scales.

![Figure 10](image1.png)

**Figure 10.** Left mode amplitude modulus (red line) and right mode amplitude modulus (green line) averaged on: a) $x \in [0, 8.1] \, \text{cm}$, b) $x \in [14.9, 23] \, \text{cm}$.

![Figure 11](image2.png)

**Figure 11.** Correlation function between propagating mode moduli for: a) $x \in [0, 8.1] \, \text{cm}$, b) $x \in [14.9, 23] \, \text{cm}$.

For each complex amplitudes $A, B, U_\omega$ and $U_q$ we have introduced the space-time averaged value, e.g. $\langle A \rangle = \frac{1}{LT} \int_0^L \int_0^T |A(x,t)| dt dx$, where $L$ is the acquisition length. These quantities provide a synthetic view of the evolution of the mode above onset and can be used as order parameters for spatial inhomogeneous patterns.

We report in figure 12 the space-time averaged amplitudes scaled by the sum of averaged amplitudes $\langle I \rangle = \langle A \rangle + \langle B \rangle + \langle U_\omega \rangle + \langle U_q \rangle$, as a function of $\varepsilon$. A relative decrease of harmonics for large values of $\varepsilon$ is evidenced. The intensity of harmonics decreases below the level of propagating spiral when the criticality parameter is increased.
The investigation of the temporal power spectra for increasing values of the criticality parameter $\varepsilon$ (figure 13) shows that the fundamental frequency decreases with $\varepsilon$, as usually observed for spiral flow in Newtonian solutions [13].

For $\varepsilon = 0.156$ (figure 14), strongly disordered flow regime is reached, with a continuous frequency spectrum. We have observed, by varying the control parameter backward and forward, that when such strongly disordered flow has been reached and maintained a significant time, it results in loss of strong coupling properties, with modified onset and fundamental frequency of counterpropagating spirals, or even in the suppression of counterpropagating spirals in favor of Taylor Vortex Flow. This is ascribed to the probable mechanical degradation of the polymer macromolecules that can be expected for high strain rate.
Figure 14. a) Space-time diagram of observed flow for $\epsilon = 0.156$; b) time power spectra of the diagram; c) spatial power spectra of the diagram.

4. Unlocked harmonics generation and Ginzburg-Landau model

The appearance of strong coupling between counterpropagating spirals that generate unlocked spatial and temporal harmonics may be described by CGLE model into which we have incorporated resonant terms for the harmonics dynamic. Following CGLE equations scaling from reference [14], we have introduced the following set of equations to describe the pattern (1):

\[
\begin{align*}
\frac{\partial A}{\partial t} + s \frac{\partial A}{\partial x} & = A + (1 + ic_1) \frac{\partial^2 A}{\partial x^2} - (1 + ic_2) A - \delta(1 + ic_3) B + U_\omega B^* \\
\frac{\partial B}{\partial t} - s \frac{\partial B}{\partial x} & = B + (1 + ic_1) \frac{\partial^2 B}{\partial x^2} - (1 + ic_2) A^2 B - \delta(1 + ic_3) A^2 + U_q^* A \\
\frac{\partial U_\omega}{\partial t} & = -\sigma(1 + ic_4) U_\omega + l(1 + ic_5) AB \\
\frac{\partial U_q}{\partial t} & = -\sigma U_q + l(1 + ic_5) AB^*
\end{align*}
\]  

(3)  
(4)  
(5)  
(6)

where $s$ is the wave group velocity, $\delta$ the coupling constant, which is positive for stable spirals, and $c_1$ the linear and nonlinear dispersion coefficients. The last terms in equations (3-4) are the simplest combinations that can be introduced from equation (1) in order to describe coupling between fundamental and resonant harmonic modes ($2q,0$) and $(0,2\omega)$. The equations (5-6) are the lowest-order equations allowed to describe the resonant harmonics as slave modes that are damped ($\sigma > 0$) in the absence of fundamental modes and sustained ($l > 0$) by coupling interaction between the fundamental, propagating modes. The damping constant is real for the mode $U_q$, while the oscillatory mode $U_\omega$ may have a frequency shift, hence the constant $c_4$ in equation (5). However, frequency spectra and demodulation of experimental
patterns suggest that the frequency of $U_{\omega}$ remains close to twice the propagating mode frequency. Hence, as a first approximation and for the purpose of this paper, we have assumed that $c_4=c_5=0$.

It appears that, in this context, some basic coupling effects in the equations can be evidenced by looking at steady solutions $U_{\omega} = AB \cdot 1/\sigma$ and $U_q = AB' \cdot 1/\sigma$ of equations (5-6). The steady solutions moduli are equal, as suggested by experimental observations (figure 6, see also figure 17 in reference [16]). Rewriting equations (3) and (4) with steady harmonics leads to

$$\frac{\partial A}{\partial x} = A + (1 + ic_1) \frac{\partial^2 A}{\partial x^2} - (1 + ic_2) \frac{A}{\partial \xi} \left( \delta(1 + ic_3) - \frac{I}{\sigma} \right) \|B\|^2 A$$

$$- \frac{\partial B}{\partial x} = B + (1 + ic_1) \frac{\partial^2 B}{\partial x^2} - (1 + ic_2) \frac{B}{\partial \xi} \left( \delta(1 + ic_3) - \frac{I}{\sigma} \right) \|A\|^2 B$$

From these equations we thus expect lowered coupling effect between left and right spirals. However, the last terms in equations (7-8) actually control rejection of one wave by the other; thus by lowering $\delta$, harmonics allow left and right spirals to coexist with larger amplitude at the same location in space. This in turn will drive harmonics through $AB$ and $AB'$ terms. Hence harmonics may be sustained permanently.

Such a scheme is consistent with the observed behavior at $\varepsilon = 0.003$ around $x = 5$ cm and $x = 20$ cm, where harmonics are roughly steady while counterpropagating spirals exhibit significant spatial overlap (figure 6). From amplitude profiles reported in figure 6, the ratio $1/\sigma$ is estimated to be 0.12. The scaled CGLE coefficients for propagating modes are estimated from value observed for spiral patterns in the same Couette-Taylor geometry [15] (table 2); $\delta$ will be approximated as 0.2.

| Table 2. Numerical values of scaled CGLE equations coefficients estimated from spiral patterns. |
| --- |
| $s = 1.1$ | $L = 6$ | $c_i = 0.06$ | $\delta \ll 1$ |

Equations (3-6) have been solved using the FEMLAB® (Lagrange quadratic finite elements, UMFPACK and DASPK solvers) with absorbing boundary conditions for all modes.

We report results obtained with zero group velocity ($s = 0$). We have chosen the initial conditions for propagating modes in form of $A(x) = F_0 \sin(\pi x/L)$ and $B(x) = F_0 \sin(-\pi x/L)$, in order to match the observed phase shifts in the experimental pattern. Initial values for harmonics were set to zero. With $c_i = c_j = c_k = 0$, stationary solutions with sustained harmonics are obtained. This is checked by solving the system (3-6) for $10^4$ dimensionless time units. Amplitude moduli distributions for fundamental modes are reported in figure 15a (solid line, $F_0 = 0.1$). Amplitude moduli distributions of harmonics are reported in figure 15b. Harmonics amplitudes $U_{\omega}$ and $U_q$ are equal. Non-zero amplitudes at $x = 0$ (figure 15b, dark line) can be obtained by adding a diffusion term $\zeta \partial^2 U / \partial x^2$ in the right-hand side of equations (5-6). For investigated values of $\zeta (0 < \zeta < 2)$, we have found that $\zeta$ has a negligible impact on the amplitude distributions of the fundamental modes (figure 15a). From the computed amplitudes, simulated space-time patterns are obtained (figure 16).
Figure 15. Distributions of the amplitude moduli for: a) stationary solutions for fundamental modes ($|A|=|B|$), dark line: $\zeta = 2$, grey line: $\zeta = 0$; b) stationary solutions for harmonics ($|U_ω|=|U_q|$), dark line: $\zeta = 2$, grey line: $\zeta = 0$. For all cases: $s = 0$, $c_1 = c_2 = c_3 = 0$, $\sigma = 1$, $l = 0.12\sigma$, $\delta = 0.2$.

Figure 16. Simulated individual space-time diagram for fundamental modes and unlocked harmonics ($\zeta = 2$).

Observed unsteady behavior of harmonics in the middle of the full system is associated with separated counterpropagating spirals, as seen on figures 5-6, where spatial overlap around $x = 12$ cm is negligible. Previous experimental results [12][16] suggested that roughly steady harmonics can also coexist with separated counterpropagating spirals for low values of the
control parameter. In this work steady harmonics in the separation zone were simulated with harmonics diffusion, in a case where large fundamental modes overlap was present on both sides of the separation zone.

5. Conclusion
For Newtonian flows, counterpropagating spirals were observed when both cylinders are counter-rotating [1]. With viscoelastic flows, counterpropagating spirals can be observed with only the inner cylinder in rotation. Present experimental results demonstrate that, with aqueous POE solutions, the interaction of counterpropagating spirals leads to generation of spatial and temporal harmonics. Amplitude of these harmonics is proportional to second power of spiral wave amplitude. Such harmonics were not observed in Newtonian liquids [17]. The increase of quadratic nonlinearity of the liquid is thus related to the viscoelastic properties. Appearance of spatial and temporal harmonics in our experiments may be related to the shear-thinning properties of the viscoelastic aqueous POE solutions, since this was not observed in any other viscoelastic solutions. Previous experimental studies [9] [10] [11] [18] were done with solutions for which shear-thinning was negligible, and numerical work [19] investigated constitutive equations without shear-thinning behavior. Ginzburg-Landau equations together with equations for coupled resonant harmonics have provided a phenomenological framework that describes part of our experimental results. Further investigations are required to elucidate the formation of unsteady harmonics pattern in regions exhibiting separated fundamental propagating modes. Experimental results with shear-thinning viscoelastic liquids could serve as further tests for constitutive equations of viscoelastic flow such as Giesekus equation [20] through direct numerical simulations or comparison of theoretically expected amplitude equations.

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