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# Large Scale Zigzag Pattern Emerging from Circulating Active Shakers

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(Received 31 March 2023; accepted 21 June 2023; published 11 August 2023)

We report the emergence of large zigzag bands in a population of reversibly actuated magnetic rotors that behave as active shakers, namely squirmers that shake the fluid around them without moving. The shakers collectively organize into dynamic structures displaying self-similar growth and generate topological defects in the form of cusps that connect vortices of rolling particles with alternating chirality. By combining experimental analysis with particle-based simulation, we show that the special flow field created by the shakers is the only ingredient needed to reproduce the observed spatiotemporal pattern. We unveil a self-organization scenario in a collection of driven particles in a viscoelastic medium emerging from the reduced particle degrees of freedom, as here the frozen orientational motion of the shakers.

DOI: 10.1103/PhysRevLett.131.068301

Viscoelasticity, namely the tendency of a material to display both viscous and elastic response under external deformation, is commonly observed in a broad range of systems, from anelastic solids [1,2] to liquid crystals [3], micelles [4], concentrated colloidal suspensions [5,6] biopolymers [7,8], or living cells [9,10]. In viscoelastic fluids, the internal molecular rearrangement spans several timescales and length scales, provoking a series of intriguing phenomena including stress relaxation, hysteresis, memory, creep, or shear thickening [11]. While the bulk behavior of such fluids has been the matter of much research to date, emergent directions point toward investigating how such fluids mediate the organization of dispersed microscopic particles. Active [12–16], passive [17–19], or externally driven [20–25] particles in viscoelastic fluids are excellent model systems for many-body organization in elastic materials, while displaying promising applications in microrheology [26,27], tissue engineering [28–30], or microrobotics [31,32]. Indeed, fluid elasticity can directly affect the propulsion behavior of microswimmers [33–40], or even be used to obtain net motion via streaming flow when pairs of particles interact [36,41,42]. However, most of these works have been focused on single or few interacting particles, leaving the rich physics of ensembles a rich ground for exploration.

Here we demonstrate that a collection of active shakers made of driven magnetic microrotors can self-organize into large scale dynamic bands displaying a zigzag shape. First, we experimentally determine the flow field around a microrotor which originates from a microscopic version of the Weissenberg effect [43–45], and is astonishingly similar to that of a shaker force dipole [46]. Finite element simulations confirm that the elastic stresses around the periodically rotating particles drive a net dipolar flow field. We then characterize the growth process which starts at a microscopic level with pairs of rotors and grows beyond the millimeter scale. We found that the growth is scale invariant and linear with time. Using particle-based simulations based on a minimal model, we show that the shakerlike flow is at the origin of the instability, and it explains the observed constant angle of the bands. These results suggest that the formation of zigzag patterns is a general effect which could arise in a broad range of systems.

We disperse anisotropic microparticles in a solution of polyacrylamide (PAAM) at a concentration of 0.05% by volume in deionized water; see Supplemental Material (SM) for more details [47], which includes Refs. [48–51]. The PAAM is a linear, high-molecular weight polymer [$M_w = (5-6) \times 10^6$] and its addition to water made the solution viscoelastic; see SM [47] for further details. From previous works [52,53] we estimate the stress relaxation time of such diluted PAAM solution to be of the order $\tau \sim 3$ ms. Within our PAAM solution we then disperse homemade ferromagnetic hematite colloids, prepared using a sol-gel technique [47,54] and characterized by a peanut-like shape with two lobes with a long (short) axis equal to $\alpha = 2.6$ μm ($\beta = 1.2$ μm); see inset in Fig. 1(a). The particles display a permanent magnetic moment of amplitude $m \simeq 9 \times 10^{-16}$ Am$^2$ [55] and oriented perpendicular to their long axis. Once dispersed in the PAAM solution, the particles sediment due to density mismatch and float at an almost fixed elevation $h$ due to the balance between gravity and electrostatic repulsion with the close substrate.

We realize active colloidal shakers by cyclically driving our particles back and forward along a fixed direction (here the $\hat{x}$ axis) using a time-dependent rotating field,

 

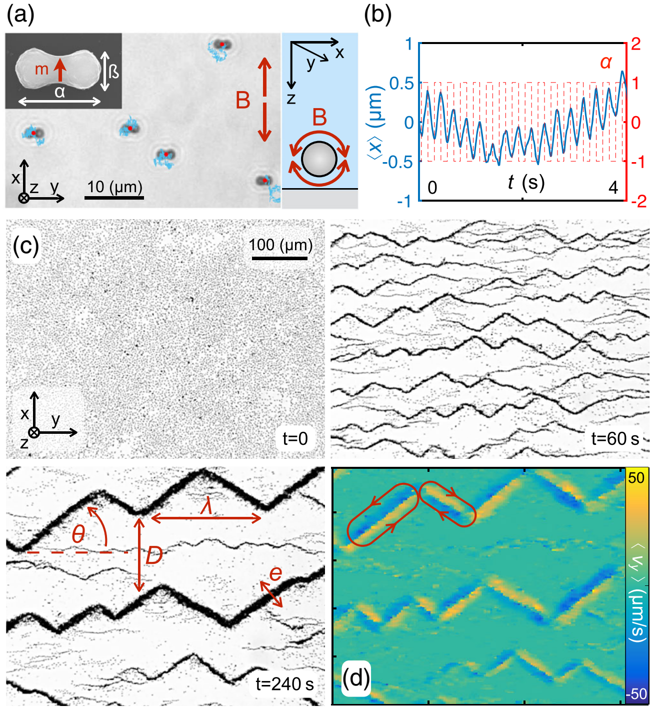


FIG. 1. (a) Trajectories of a dilute suspension of shakers after $N = 68$ cycles of the rotating field, Movie S1 in Ref. [47]. Top inset shows scanning electron microscope image of one hematite particle with the permanent magnetic moment $\boldsymbol{m}$. Small scheme on the right-hand side shows lateral view of one magnetic roller. (b) Mean displacement $\langle x \rangle$ versus time $t$ of shakers. Dashed line denotes the field direction of rotation. (c) Sequence of images illustrating the band formation from an initially disordered suspension ($t = 0$) and under a magnetic modulation with $f = 80$ Hz and $\delta t = 1/8$ s; see also Movie S3 in Ref. [47]. The image at $t = 240$ s shows two bands at a distance $D$ with wavelength $\lambda$, bond angle $\theta$, and thickness $e$. (d) Average horizontal velocity $\langle v_y \rangle$ of the shakers in bands. Red arrows denote the direction of the circulating particles, Movie S4 in Ref. [47].

$$\boldsymbol{B} = B[\sin(2\pi t \Delta f_-)\hat{\boldsymbol{x}} + \cos(2\pi t \Delta f_+)\hat{\boldsymbol{z}}], \qquad (1)$$

with $\Delta f_\pm = f \pm \delta f/2$ and $\delta f$ the frequency difference between the two field components along the $\hat{\boldsymbol{x}}$ and $\hat{\boldsymbol{z}}$ axis. The applied modulation periodically changes the direction of rotation every $\delta t = 1/(2\delta f)$, inducing a magnetic torque $\boldsymbol{\tau}_m = \boldsymbol{m} \times \boldsymbol{B}$ that sets the particles into rotational motion around their short axis with an angular speed $\Omega = 2\pi f$ for frequencies $f < 100$ Hz (synchronous regime). Moreover, since the applied field is circularly polarized along the $(\hat{\boldsymbol{x}}, \hat{\boldsymbol{z}})$ plane, it aligns the permanent moments of the particles, ensuring a fixed angular orientation. Here we fix $B = 5.5$ mT and $f = 80$ Hz.

The presence of a solid surface breaks the spatial symmetry and produces a rolling transport due to the rotation-translation hydrodynamic coupling [56]. In the limit $\delta f = 0$, the field does not switch, and the hematite particles roll above the substrate by acquiring a frequency tunable propulsion speed $v_x \sim 2\pi\beta f$ [57], with $\beta$ the particle short axis. In contrast, for a finite frequency delay $\delta f = 4$ Hz, individual particles perform small oscillations of amplitude $\Delta x \sim 0.5$ μm and zero average velocity; see Fig. 1(b) herein and Movie S1 in Ref. [47].

The small amplitude time-reversible motion of the particle should produce no net displacement in a Newtonian fluid [58], as shown in Movie S2 in Ref. [47]. Instead, we find that, at high normalized particle density $\phi = 0.117 \pm 0.002$, the shakers organize in complex dynamic bands which grow linearly with time. As shown in the sequence of images in Fig. 1(c), a system of randomly distributed particles evolves into a structured array of bands with zigzaglike shape after a few seconds of magnetic driving. The bands grow first by acquiring nearest particles, located on their lateral sides, and then via a continuous merging; see Movie S3 in Ref. [47]. These dynamic bands acquire a zigzag shape with branches arranged at a constant angle of $\theta_l = \pm 31°$ delimited by cusps. A careful inspection of the particle velocity within a band, Fig. 1(d), reveals that the shakers move collectively, forming rotating vortices with fast circulating edge currents up to 50 μm $s^{-1}$; see Movie S3 in Ref. [47]. Each branch of a band is made of a large scale vortical flow of particles, and cusps within a band connect vortices of opposite chirality, similar to a two gears system. These bands were observed to form also for smaller peanuts ($\alpha = 1.8$ μm, $\beta = 1.3$ μm), or by varying $f \in [40, 100]$ Hz and for $\delta f > 0.75$ Hz.

To understand these unexpected, complex structures we analyzed the flow field generated by a single microrotor. We obtain the velocity field around a microrotor, averaged over many field periods, via particle tracking velocimetry; see SM for details [47]. Note that in a Newtonian fluid hydrodynamics interactions are time reversible so that no net particle displacement would emerge. Strikingly, the obtained flow field in Fig. 2(a) displays strong similarities with that generated by a shakerlike force dipole [46]. Shakers are a category of squirmers that generate a flow pattern similar to that of several microorganisms such as *Escherichia coli* bacteria [59] but without the polar component, which prevent them from self-propelling. These particles exert stresses on the surrounding fluid [60–62] that drive a flow field displaying an attracting part at the particle sides and a repulsive one at the tips, with a recirculation vortex between these two regions. An estimate of the Deborah number, $\mathrm{De} = 2\pi f \tau$, yields $\mathrm{De} \approx 1.5$, which suggests that this particular flow field results from the fluid elasticity. Thus, the particle rotation induces a normal stress difference along the $\hat{\boldsymbol{x}}$ axis which induces a flow toward the rotor and, by volume conservation, the fluid is expelled toward the tips; see Fig. 2(b). We confirm this hypothesis by computing the flow velocity $\mathbf{v}$ around a periodically rotating ellipsoid via three-dimensional numerical simulations. We consider a stress tensor $\mathbf{T} = -p\mathbf{I} + \eta_s(\nabla\mathbf{v} + \nabla\mathbf{v}^T) + \sigma$ as the sum of a Newtonian contribution (viscosity of water $\eta_s$)

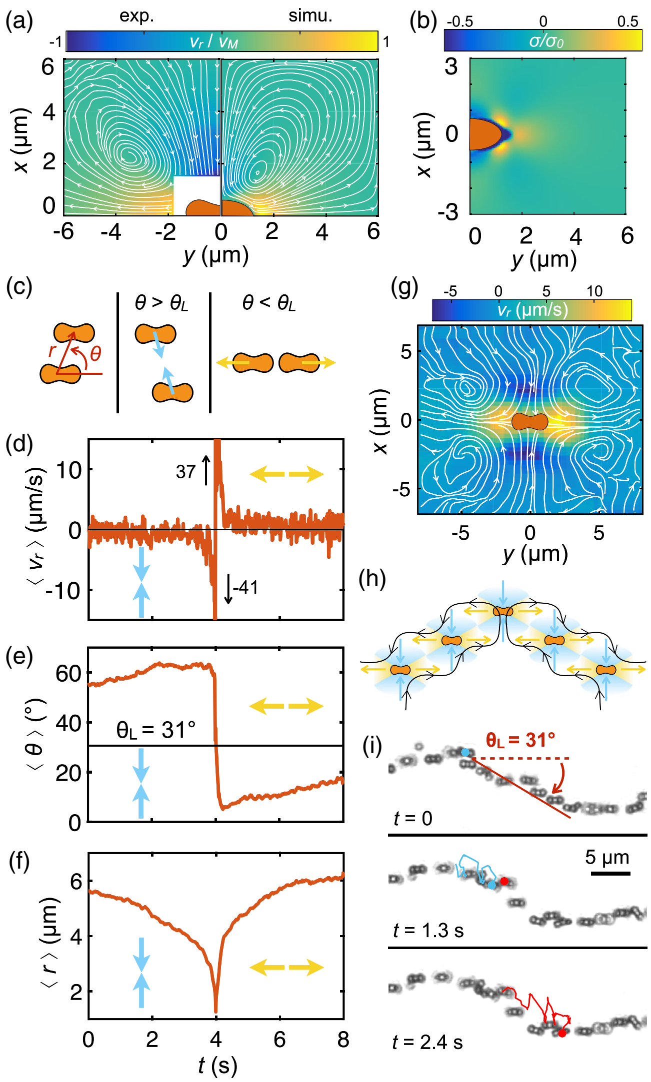


FIG. 2. (a) Normalized flow field created by a shaker $v_r/v_M$ in the $(x, y)$ plane. The color codes for the radial velocity $v_r$ are normalized by the maximal radial velocity $v_M$. It shows regions of attractions ($v_r < 0$) and repulsion ($v_r > 0$) separated by $\theta_l$. Left-hand (right-hand) panel refers to experimentally measured (simulated) flow field ($v_M = 2.2\ \mu\mathrm{m\,s^{-1}}$ for the experiments, $v_M = 22\ \mu\mathrm{m\,s^{-1}}$ for the simulation; see also the SM [47]). (b) Normalized first normal stress difference in the $(x, y)$ plane with $\sigma_0 = (\eta_s + \eta_p)2\pi f$; see text. (c) Schematics of two microrotors in three configurations: attraction (repulsion) arises when $\theta > \theta_l$ ($\theta < \theta_l$) with $\theta_l = 31°$. (d)–(f) Evolution with time of (d) the average radial velocity $\langle v_r \rangle$, (e) the relative angle $\langle \theta \rangle$, and (f) the radial distance $\langle r \rangle$ between two approaching microrotors. In all images blue [black] (yellow [gray]) arrows indicate attraction (repulsion) between the pair, Movie S5 in Ref. [47]. (g) Top view of the flow velocity generated from the interaction of two shakers. (h) Assembled structure from the flow produced by the shakers. (i) Sequence of snapshots showing a one-particle thin band with superimposed two particle's trajectories, Movie S6 in Ref. [47].

coming from the solvent and a viscoelastic one $\sigma$ introduced by the PAAM. Here $p$ is the pressure that enforces the incompressibility condition, $\nabla \cdot \mathbf{v} = 0$. We use the Oldroyd-B constitutive model, which predicts a constant viscosity but a nonzero normal stress difference [63], via an additional constitutive equation:

$$\tau\left(\frac{\partial}{\partial t}\sigma + \mathbf{v} \cdot \nabla\sigma - \nabla\mathbf{v} \cdot \sigma - \sigma \cdot \nabla\mathbf{v}^T\right) + \sigma = \eta_p(\nabla\mathbf{v} + \nabla\mathbf{v}^T), \tag{2}$$

where $\eta_p$ is the polymer viscosity. Such a model reduces to only two parameters, $\eta_p$ and $\tau \sim 3$ ms. The polymer viscosity can be extracted from the zero-shear viscosity of the solution, which from the experimental measurements is $\eta_0 \approx 2\eta_s$; thus, we estimate $\eta_p \approx \eta_s$. See SM [47] for further technical details.

The right-hand panel of Fig. 2(a) shows that the computed velocity field in the plane $z = h$ and averaged over one period $\delta t$ displays the same features as that measured in the experiments and confirms that the mechanism driving the dipolar flow structure is the first normal stress difference; see Fig. 2(b). This result is consistent with the seminal works by Giesekus [64] and Fosdick and Kao [65], who showed that the first normal stress distribution around a rotating sphere generates streamlines that increase with even powers of De. It follows that the flow field introduced by the first normal stresses does not change sign upon changing the direction of the particle rotation leading to a nonzero average over one period.

We then analyzed the interactions between a pair of shakers using data from 33 separate experiments. Because of the field alignment, the magnetic particles display negligible orientational motion and the relative orientation can be described in terms of a single angle $\theta$, Fig. 2(c). The relative velocity field between the pair has a similar structure to the velocity field generated by a single one. Two particles attract each other when they are side by side and repel when positioned tip to tip. The transition between the two regions occurs at an angle $\theta_l = 31°$, Fig. 2(c). When two particles are initially arranged such that $\theta > \theta_l$, they approach first slowly and keeping their relative orientation constant. Near close contact, $r = 2\ \mu$m, we observe a rapid sliding process which causes a speed-up effect reaching relative velocities up to $v_r = 40\ \mu\mathrm{m\,s^{-1}}$, Figs. 2(d)–2(f). Such a process rearranges the rotors from side by side ($\theta \sim 90°$) to tip to tip ($\theta \sim 0°$), Movie S5 in Ref. [47]. Thus, close particles arrange themselves at an angle $\theta_l$ where attraction and repulsion are minimized and, by drawing the flow lines, one recovers the direction of rotation of the vortices, Fig. 2(h). During band growth, defects arise in the form of cusps which sink incoming particles expelling them from the opposite side. Such a

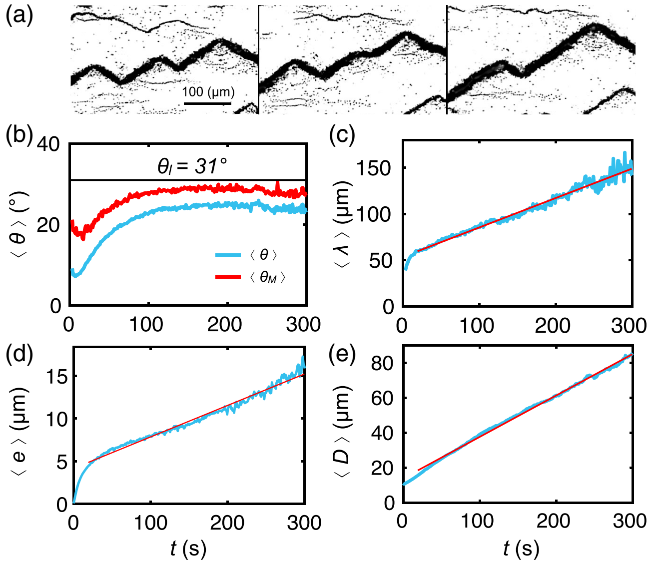


FIG. 3. (a) Sequence of snapshots showing the annihilation of a cusp (total duration $\Delta t = 30$ s), Movie S7 in Ref. [47]. Coarsening dynamics: time evolution of the mean angle $\langle\theta\rangle$ (b), wavelength $\langle\lambda\rangle$ (c), thickness $\langle e\rangle$ (d), and distance $\langle D\rangle$ (e). After a short transitory, $\lambda$, $e$, and $D$ all scale linearly with time (red line) while $\theta$ set to a stationary value of $\theta_l = 31°$. In (b) $\langle\theta\rangle$ corresponds to the full average over the experimental data, weighted by the size of the zigzag bands, while $\langle\theta_M\rangle$ is its maximal value over the bands.

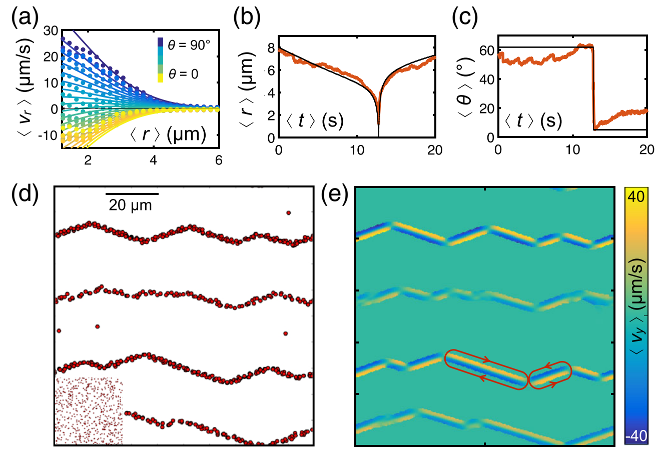


FIG. 4. (a) Average radial velocity $\langle v_r\rangle$ between two particles as a function of their relative distance $r$ and for different angle $\theta$ (color bar). Scattered data are experiments, while continuous lines are nonlinear regression using Eq. (9) in Ref. [47]. (b),(c) Average radial distance (b) and relative angle (c) as function of time for pairs of interacting particles. In both graphs, experiments (simulation) are represented by the red [gray] (black) curve. (d) Image showing the zigzag state obtained after a time $t = 50$ s, with $N = 601$ particles, Movie S8 in Ref. [47]. The inset shows the initial random configuration of the particles. (e) Corresponding color-coded plot showing the average horizontal velocity $\langle v_y\rangle$ with arrows denoting the direction of the circulating particles.

hypothesis is confirmed by observing the formation of one particle thin band, Fig. 2(i) herein and Movie S6 in Ref. [47], where the constituent particles detaching from the branch are dragged along the vortical edge current toward the nearest cusp.

The ensemble of shakers exhibits a self-similar behavior with scale invariance in time as they evolve to large scale structures, Fig. 1(c). During coarsening, the different band parameters such as wavelength [$\lambda$, Fig. 3(c)] thickness [$e$, Fig. 3(d)], and distance [$D$, Fig. 3(e)] grow linearly in time, while the bond angle rapidly saturates to $\theta_l$, Fig. 3(b) [66]. As shown in Fig. 3(a), a band is composed of a sequence of topological defects in the form of cusps that connect different branches in a zigzag manner. During coarsening, small branches disappear in favor of large ones inducing the annihilation of cusps, Movie S7 in Ref. [47]. This dynamically slow process ultimately would lead to a single straight band of particles, or to separate bands at very large distance between them. However, due to the slow velocity of coarsening and the large system size (observation window 0.66 mm, whole system size 1 cm), this state is difficult to reach experimentally. During coarsening, separate bands merge by reducing their distance until touching each other. Such a process is triggered in part by the cusp annihilation that increases the wavelength $\lambda$ and so the spatial extension of a band along the lateral direction ($\hat{\boldsymbol{x}}$). The fusion of two bands increases the interband distance $D$ and gives rise to a new structure with larger thickness $e$. This new band will in turn annihilate its cusps, starting a new cycle. The cusp's annihilation process ($[A] + [A] \to 0$) can be written as $(d[A]/dt) = -k[A]^2$, with $[A]$ the linear cusp concentration in a band and $k$ the annihilation rate constant. The solution of this equation is $[A](t) = a_0/(1 + a_0 kt)$, with $a_0 = [A]$ $(t = 0)$. By definition, $\lambda = 2/[A]$, one thus recovers the linear growth in time of the wavelength by writing $\lambda(t) = 2(kt + 1/a_0)$.

To rationalize our results, we set up a minimal simulation scheme that reproduces the self-organization scenario neglecting magnetic and steric interactions. The former are not considered given the relative low value of $\boldsymbol{m}$. For two hematite rotors $(i, j)$ aligned tip to tip [side by side] at the closest distance of $x_{ij} = \beta$ [$y_{ij} = \alpha$], the time averaged potential is relatively weak, given by $\langle U_d\rangle = -\mu_0 m^2/(8\pi x_{ij}^3) = -5.8 k_B T$ [$\langle U_d\rangle = \mu_0 m^2/(4\pi x_{ij}^3) = 1.1 k_B T$], with $\mu_0 = 4\pi \times 10^{-7}$ Hm$^{-1}$. Thus, we are left to consider the generated velocity field [see Fig. 4(a)] that we obtain directly from the experimental data. We use an empirical function which simultaneously fits all data [Eq. (9) in SM [47]] and provides the relative velocity $\boldsymbol{v}(r_{ij}; \theta_{ij})$ between particles at relative position $r_{ij}$ and orientation $\theta_{ij}$. Thus, we numerically integrate the equation of motion $(d\boldsymbol{r}_i/dt) = \sum_{i \neq j} \boldsymbol{v}(r_{ij}; \theta_{ij})$; more details on the implementation can be found in the SM [47]. Taking into account the different approximation used, the result is rather striking since it allows us to reproduce the band formation process and predict the correct circulation flow across the bands, as shown in Figs. 4(d) and 4(e). Thus, the zigzag pattern arises due to the shakerlike shape of the flow field generated by the particle rotation, Movie S8 in Ref. [47].

In conclusion, we show that zigzag bands emerge because of the shakerlike flow field generated by the magnetic rotors. In our system, this flow field results from the elastic stress created by the particle rotation within the PAAM solution. However, similar patterns have also been reported in Newtonian fluids for particles submitted to an ac field [67]. The physical origin of this instability was explained by macroscopic gradients in the electrolyte concentration due to the field-induced concentration gradients near the particles' surfaces [68,69]. Further, such instability was attributed to mutual polarization of particles, causing them to rotate [70,71], and recently to the presence of electrokinetic flows [72]. Our Letter shows that the zigzag instability is even more general, suggesting that it is not the forcing (magnetic or electric) nor the medium (Newtonian or viscoelastic) that matters but rather the type of hydrodynamic flow those systems create around the particles. The frozen orientation of the shakers plays an important role in stabilizing the zigzags. Indeed, in active nematics it was shown that freezing one orientation via a constant magnetic field induces a zigzag stripe phase [73]. In contrast, systems of force dipoles free to rotate and/or able to self-propel may not exhibit such stable structures as they generate turbulent dynamics. Thus, we find that the zigzag instability is a general phenomenon that would depend only on the symmetry of the flow field developed by the particles. A potential technological application of our Letter could be to use the vortices as a conveyor belt to transport particles or to mix fluids at small scales. It could also be used to localize magnetic inclusions in microfluidic devices. The oscillating field could be used, for example, to control the flow by inducing (removing) clogging when the oscillating field is switched on (off).

We thank Thomas M. Fischer and Jaume Casademunt for stimulating discussions, and Jordi Ortín for help with the rheological measurements. This work has received funding from the European Research Council (ERC Consolidator Grant Contract No. 811234). P. T. acknowledge support from the program "ICREA Acadèmia" and the Agència de Gestió d'Ajuts Universitaris i de Recerca (2021 SGR 00450). M. D. C. acknowledges funding by the Spanish Ministry of Science and Innovation (MCINN) and Next-GenerationEU program under the Ramon y Cajal (RYC2021-030948-I) fellowship, the retos de investigación Grant No. PID2020–113033 GB-I00 and the government of Aragon (LMP63_21).

*Note added.*—Recently we became aware of a theoretical work posted in arXiv [74], where a similar zig-zag structure was observed for active particles producing stresslets and forming canted conveyor belts.

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