Asymmetry in the interaction between an individual and its environment is generally considered essential for the directional properties of active matter, but can directional locomotions and their transitions be generated only from intrinsic chemical dynamics and its modulation? Here, we examine this question by simulating the locomotion of a bioinspired active gel in a homogeneous environment. We find that autonomous directional locomotion emerges in the absence of asymmetric interaction with the environment and that a transition between modes of gel locomotion can be induced by adjusting the spatially uniform intensity of illumination or certain kinetic and mechanical system parameters. The internal wave dynamics and its structural modulation act as the impetus for signal-driven active locomotion in a manner similar to the way in which an animal’s locomotion is generated via driving by nerve pulses. Our results may have implications for the development of soft robots and biomimetic materials.
undergoes periodic changes during the homogeneous BZR oscillations, causing the gel to undergo autonomous swelling and deswelling, reminiscent of the rhythmic muscle contractions in animals (17).

In a large enough gel sample, the propagation of chemical waves (spatiotemporal patterns of the concentration of oxidized catalyst) drives gel peristalsis, resulting in locomotion of the whole sample. This phenomenon is analogous to nerve pulses driving the locomotion of animals through deformable muscles (19). The gel lattice-spring model (gLSM) proposed by Balazs and coworkers (20–22) captures the chemomechanics dynamics of the BZ gel, i.e., the large-scale shape changes (23), locomotion, and collective behaviors of the gel sample (24).

The first effort toward understanding BZ gel locomotion reported (25) that the direction of motion results from a counter-effect of polymer-solvent interdiffusion during the chemical waves, which propels the gel against the direction of wave motion (retrograde wave locomotion). Subsequently, under a step-changed light intensity condition, another fundamental mode of locomotion, i.e., direct wave locomotion, and a retrograde-direct wave transition, were found, which was attributed to a “chemical anchoring effect” modulating the push-pull competition of the chemical waves (19). That study revealed that the internal dynamics (push-pull effects) of the system play a dominant role in the gel locomotion. A more complex form of locomotion, reciprocating migration of the gel, was subsequently found (12), in which an autonomous transition between retrograde and direct wave locomotion is rooted in the interaction between the system’s dynamic instability and a gentle spatial gradient of illumination. Like neuronal signals, which can be described, for example, by the Fitzhugh–Nagumo (26–28) or Hodgkin Huxley model (29), BZ waves, described by the Oregonator model, interacting with an active gel are nonlinear signal waves, which can serve as an analog of active (e.g., animal) locomotion.

It is, however, difficult to extract the essential origin of the gel locomotion from these earlier studies (11, 12, 19) due to the presence of differential illumination (i.e., the gel interacts with a heterogeneous environment), which introduces asymmetrical interaction into the system. In this work, we address the questions raised above by simulating a BZ gel that can move in a periodic space under a homogeneous environment; that is, no asymmetrical effect is introduced into the system from external conditions. We also examine the effects of modifying the parameters that govern the internal dynamics of the gel.

RESULTS

Locomotion modes and their transition in one spatial dimension

To investigate the gel locomotion in a homogeneous environment, we use pulse waves to propel a stimulus-responsive gel under uniform light intensity on a pseudo–one-dimensional (1D) ring. The 1D gel system can be seen as a collective of homogeneous synchronizing lattice oscillators. Once initialized, e.g., by a random fluctuation, the phase differences between these diffusion mechanics–coupled oscillators evolve to generate pulse waves that repeatedly propagate through the periodic boundary even in the absence of illumination, and no asymmetry is introduced into the gel sample via the boundary conditions or the environment (see fig. S1). The coupled oscillators lead to propagating pulse waves, resulting in directional locomotion of the entire gel. To speed up the simulations, we initiate pulse waves to propagate in the gel with the most likely wave number, i.e., one-pulse waves, according to fig. S2. A modified gLSM is used to study the gel locomotion, in which the kinetics of the photosensitive BZR, i.e., photoinduction and photoinhibition effects (30), the relationship between extreme values of the ν variable and the light intensity (19), and the illumination-induced bifurcation of local oscillations, are described by a two-variable photosensitive Oregonator model (11, 31). The chemical kinetics and the structure of the chemical waves can be modulated by varying the intensity of the homogeneous illumination (I), which allows us to control the gel locomotion propelled by the chemical waves. The details of the model, parameters, calculation procedures, and simulation settings are given in Materials and Methods.

We next consider the chemical dynamics and locomotion of a photosensitive BZ gel modulated by increasing levels of homogeneous illumination. Key transitions occur at the following light intensities $I_a = 0.0625$, $I_b = 0.0809$, $I_c = 0.0870$, and $I_d = 0.117$: (i) simple-to-complex pulse waves with interval-asymmetry structures (IASs), (ii) retrograde structured pulse waves to a stationary balance between the driving effects of primary and backfired waves, (iii) gel with no net motion to direct wave motion, and (iv) breaking up of the IASs. These phenomena are discussed in more detail below.

At low light intensity ($0.0 \leq I < I_a$), e.g., $I = 0.02$, as shown in Fig. 1A, simple pulse waves repeatedly propagate through the gel, propelling the gel to move opposite to the waves (inset view of the net displacement shown in Fig. 1B), generating retrograde wave gel locomotion. One can identify two wave regions in these simple waves: the wave front and the wave back are indicated in Fig. 1A. These regions respectively push and pull the local gel to move along the $+x$ and $-x$ directions alternately (“1-push” and “2-pull” in the inset of Fig. 1B), and the net displacement over 1 cycle is in the $-x$ direction, as shown in the inset of Fig. 1B. We emphasize that gel locomotion is driven by simple pulse waves even without illumination, i.e., $I = 0$, indicating that internal interaction between BZ waves and active matter is the origin of the stress asymmetry between push and pull.

When the intensity exceeds $I_a$, IASs begin to emerge in the simple pulse waves. As shown in Fig. 1, C, E, and G, the simple pulse wave is transformed into a complex pulse wave. An IAS is a structural unit of these complex pulse waves produced by an illumination-induced spatiotemporal distortion. A complete IAS is shown in the inset to Fig. 1G. Each complex pulse wave is formed by the arrangement of six IASs ($N_{IAS} = 6$ under illumination intensities $I_a < I < I_d$), and each IAS is made up of two local waves, that is, a primary wave and a backfired wave. Primary waves are local waves that propagate along the overall direction of the pulse waves (red arrow in Fig. 1G, inset), whereas backfired waves are local waves that propagate in the opposite direction (blue arrow in Fig. 1G, inset). Each IAS has four wave regions, the primary wave’s front and back and the backfired wave’s front and back, which are roughly located at numbers 1 to 4, respectively, in Fig. 1G. With increasing illumination ($I_a < I < I_b$), each IAS undergoes gradual distortion, as shown in Fig. 1 (C, E, and G). Note that the overall direction of pulse wave propagation does not change during this process. Under the driving force of these gradually distorting waves, the dynamics of the locus of the gel’s center, $R_c$, (adjusted for the moving baseline) changes gradually from quasiperiodic (Fig. 1D) to aperiodic oscillation (Fig. 1, F and H) with increasing illumination.

Meanwhile, the net displacement of the $R_c$ locus shows the gel locomotion transition from a retrograde wave ($I = 0.070$, Fig. 1D)
Fig. 1. Spatiotemporal plots of v and the motion of the gel center under homogeneous illumination. The two columns display spatiotemporal plots of v and position of the gel center versus time, respectively. The light intensity is (A and B) $I = 0.020$, (C and D) $I = 0.070$, (E and F) $I = 0.087$, and (G and H) $I = 0.110$. Color denotes the concentration of v. Labels 1 to 4 in (G) denote the approximate positions of the four wave regions in one interval structure, respectively, the primary wave's front and back and the backfired wave's front and back. Movie S1 shows the gel locomotion at $I = 0.0, 0.02, 0.070, 0.087$, and 0.116.
through a motionless state \( (I = I_a, \text{Fig. 1F}) \) and, ultimately, to a direct wave \( (I = 0.110, \text{Fig. 1H}) \). This transition is mediated by the IAS of the waves, and the zero net-displacement state of the gel is the critical state of the transition (light intensity \( I_a \) is the transition point). In this work, we have not found direct waves driven by simple pulse waves, which indicates that the presence of IAS may be necessary for the transition between retrograde and direct wave locomotion. When the intensity of illumination exceeds \( I_d \), the IAS breaks up into reverse-propagating pulse waves (see fig. S3), and the gel returns to retrograde wave locomotion comparable to that seen at low \( I \), but in the opposite direction. Note that the retrograde-direct transition can also be generated by adjusting other system parameters such as \( \varepsilon \) (Oregonator kinetic parameter), \( f \) (Oregonator stoichiometric parameter), \( \chi^* \) (chemistry-gel dynamic coupling parameter), \( K \) (wave number) and \( L_{gel} \) (size of the gel), as shown in figs. S4 to S7.

**Kinematic analysis of gel locomotion**

To clarify the origin of the homogeneous light-induced transition between locomotion modes, we quantify key characteristics of the pulse wave and the gel locomotion by introducing several parameters: the local curvature \( \kappa(x) \) of the chemical waves \( (\kappa) \), the mean velocity of the gel movement \( (V_g) \), and the mean net displacement of the gel center for each wave region \( (L_i) \). The first step needed to ensure that these characterizations are useful is the division of the pulse wave into regions. We accomplish this by using the total differential of \( v \) in the spatiotemporal plots, which captures the integrated dynamics of the pulse waves. The differential expression reads

\[
dv = v_x \, dx + v_t \, dt
\]

Using Eq. 1, we propose a criterion to divide the pulse wave regions, as shown in Table 1.

We choose this criterion based on the following reasoning. On the one hand, at the wave fronts of both simple (Fig. 1A) and complex (Fig. 1, C, E and G) pulse waves, the local value of \( v \) increases with time \( (v_t > 0) \), while at all wave backs, it decreases with time \( (v_t < 0) \). On the other hand, when the overall motion of the pulse waves lies along the +x direction (with \( I < 0.117 \)), the spatial gradients of \( v \) (i.e., \( v_x \)) are negative and positive, respectively, at the primary wave front and wave back, but for backfired waves, the signs of \( v_x \) are reversed due to the wave traveling in the −x direction, as shown in Fig. 1G. Therefore, the wave’s regions can be numerically identified by the criterion for \( v_t \) and \( v_x \) in Table 1. Using this criterion, the changes of the wave regions with increasing \( I \) may be distinguished, as shown in Fig. 2A. Simple pulse waves (e.g., the plot with \( I = 0.02 \)) contain only the primary wave’s front \( (1) \) and back \( (2) \). For complex pulse waves (plots with \( I = 0.07 \) to 0.116), the backfired wave front \( (3) \) and wave back \( (4) \) are adjacent to the primary wave front and wave back.

For gel locomotion driven by pulse waves, the mean velocity of gel movement \( (V_g) \) represents the push-pull effect of the waves with increasing light intensity. Integrating the instantaneous velocity of the gel center to retrograde wave locomotion comparable to that seen at low \( I \), but in the opposite direction. Note that the retrograde-direct transition can also be generated by adjusting other system parameters such as \( \varepsilon \) (Oregonator kinetic parameter), \( f \) (Oregonator stoichiometric parameter), \( \chi^* \) (chemistry-gel dynamic coupling parameter), \( K \) (wave number) and \( L_{gel} \) (size of the gel), as shown in figs. S4 to S7.

**Materials and Methods.** In Fig. 2C, when \( I < I_d \) (region I), \( \kappa(x) \) is always zero, which indicates that only simple pulse waves propagate in the gel. As \( I \) increases from \( I_a \) to \( I_d \) (region II), a spatially periodic organization of \( \kappa(x) \) begins to emerge and gradually strengthens, leading to six periodic structures (IAS) and distortion of the pulse waves. Meanwhile, comparing Fig. 2A with Fig. 2C, we see that when \( I > I_d \), wave regions 3 and 4 start to appear. The shape of the waves shortly after onset is seen at \( I = 0.07 \), near the transition point \( (L_d) \), in the spatiotemporal plots shown in Fig. 2 (A and B). The plots show a backfired wave front (wave region 3), which stems from the primary wave front (wave region 1). Its back (wave region 4) originates from the primary wave’s back (wave region 2). The complex dynamics in wave region 2 (primary wave back) play a key role in the transition of the locomotion mode and will naturally arise in our subsequent analysis.

The partial displacements \( L_i \) \( (i = 1, 2, 3, 4) \) serve as the blocks from which the gel locomotion is built, since they characterize the propulsive contribution from each wave region. The key lies in finding the spatiotemporal region that dominates the mode transition of the gel locomotion. Fig. 2D, the \( V_g-I \) curve, shows the net velocity of gel locomotion as a function of the illumination intensity in simple pulse wave–driven retrograde wave locomotion (region I, 1 May 2020

### Table 1. A criterion for identifying the subregions of chemical waves.

| Region of waves                  | \( v_t \) | \( v_x \) |
|----------------------------------|----------|----------|
| Primary wave's front (region 1) | > 0      | < 0      |
| Primary wave's back (region 2)  | < 0      | > 0      |
| Backfired wave's front (region 3)| > 0      | > 0      |
| Backfired wave's back (region 4)| < 0      | < 0      |
\( V_g < 0 \) and \( \kappa(x) = 0 \) as well as in complex pulse wave–propelled retrograde wave locomotion [region II, \( V_g < 0 \) and \( \kappa(x) \neq 0 \)] and complex pulse wave–driven direct wave locomotion [region II, \( V_g > 0 \) and \( \kappa(x) \neq 0 \)]. From Eqs. 3 and 4, and the above discussion, we have

\[ V_g T = L_{primary} + L_{backfire} \]  

where \( L_{primary} = L_1 + L_2 \) and \( L_{backfire} = L_3 + L_4 \) are the net displacements driven by the primary waves and the backfired waves, respectively. For a simple pulse wave, there is no \( L_{backfire} \). To examine the competition between the primary and backfired waves that determines the transition from negative to positive \( V_g \), we plot \( L_{primary} \) and \( L_{backfire} \) versus \( I \) in Fig. 2E. Here, the light intensity \( I_0 \) denotes the critical point where \( L_{primary} = L_{backfire} \). Near the retrograde-direct transition point \( I_0 \), \( L_{primary} \) increases roughly linearly with \( I \), while \( L_{backfire} \) (which is negative) decreases at a rate that diminishes with \( I \). A clearer view is shown in the inset of Fig. 2E, where \( L_{primary} \) increases linearly across \( I_0 \) and is always larger than \( I_1 \), indicating that \( L_{primary} \) dominates the transition from negative to positive \( V_g \) at \( I_0 \). The decomposition of \( L_{primary} \) and \( L_{backfire} \) into \( L_1 \) to \( L_4 \) with increasing illumination around \( I_0 \) is shown in Fig. 2F. One can see that \( L_1 \) is nearly constant, whereas \( L_2 \) decreases substantially, resulting in a negative-to-positive transition of \( L_{primary} \), indicating that \( L_2 \) dominates the change of \( L_{primary} \). On the other hand, for backfired waves, the two curves \( L_3 \) and \( L_4 \) are roughly symmetric about the zero line (horizontal dashed line in Fig. 2F), so that they almost cancel each other. In summary, wave region 2, which is the primary wave back, is the key region for the retrograde-direct transition of the gel locomotion.

In this stimulus-responsive gel with chemomechanical activity, the locomotion of the gel stems from the chemically induced asymmetry...
of the internal stress over the sample. We now analyze the internal dynamics of $V_p$ by examining the effect of homogeneous illumination on the kinematic terms, i.e., the mean values of the gel velocity $[(V_p^\text{mean})_i]$, the stress gradient $[(\sigma_x^\text{mean})_i]$, and the apparent friction coefficient $[(\xi')^\text{mean}]$ in each wave region, as shown in Fig. 3. The relationship between the local velocity of the gel and the stress is given by (21)

$$V_p = \frac{\sigma_x}{\xi'(\phi)}$$

where $\sigma_x = -\nabla(\pi_{\text{osm}} - \sigma_{\text{el}})$, $\xi'(\phi) = \Lambda_0^{-1}(1 - \phi)(\phi/\phi_0)^{3/2}$, $V_p$ is the grid velocity of the gel, $\sigma_x$ is the stress gradient, and $\xi'(\phi)$ is the apparent friction coefficient. The mean values of the kinematic parameters $[(V_p^\text{mean})_i]$, $[(\sigma_x^\text{mean})_i]$ and $[(\xi')^\text{mean}]$ represent the average over each wave region (Fig. 3, A, C, and D). $A_i$, the area that each wave region occupies in the plots (Fig. 3, A and B), is also shown in Fig. 3B.

Each displacement $L_i$ can be decomposed into $(V_p^\text{mean})_i \times A_i/S$, as shown in Fig. 3 (A and B). $(V_p^\text{mean})_i$ varies linearly with illumination near $I_c$ in all four wave regions. However, $(\sigma_x^\text{mean})_i$ has a markedly smaller slope. The areas of the wave regions also undergo a roughly linear change with $I$ around $I_c$; i.e., $A_3$ and $A_4$ increase, while $A_1$ and $A_2$ decrease. Our analysis of $(V_p^\text{mean})_i$ and $A_i$ suggests that the kinematics of $L_2$ are dominated by the velocity of the gel rather than the relative areas of the wave regions.

We next analyze the mechanics of the local gel velocity ($V_p$) under the driving of the chemical waves using Eq. 6. The mean value of the apparent friction viscosity ($\xi')^\text{mean}$ is investigated in Fig. 3D and fig. S8G. Under illumination $I$, (see fig. S8G), the distribution of $(\xi')^\text{mean}_i$ over each wave region is relatively uniform. Changes in the sign of $V_p$ arise from $\sigma_x$ rather than from $\xi'$, which is always positive. We can estimate $(V_p^\text{mean})_i$ in each region by averaging Eq. 6

$$(V_p^\text{mean})_i \approx \frac{[(\sigma_x^\text{mean})_i]}{[(\xi'(\phi)^\text{mean}]_{i}} (i = 1, 2, 3, 4)$$

As shown in Fig. 3C, each curve of $(\sigma_x^\text{mean})_i$ versus $I$ shows a trend that parallels the corresponding $(V_p^\text{mean})_i$ curve in Fig. 3A. Each $(\xi')^\text{mean}_i$ changes little with $I$, especially for the primary wave. As the inset to Fig. 3D shows that the ratio of $(\xi')^\text{mean}_i$ over $(\xi')^\text{mean}_2$ is almost independent of $I$, implying that the notable decrease of $L_2$ near $I_c$ (see Fig. 2F) is dominated by the mechanical term $(\sigma_x^\text{mean})_2$. Note that positive and negative $\sigma_x$ values favor direct and retrograde waves, respectively; thus, the changes in the pull effect $[(\sigma_x^\text{mean})_2]$ with illumination facilitate the transition from retrograde to direct waves.

The above analysis suggests that the insensitivity of $(\sigma_x^\text{mean})_2$ to changes in $I$ is the key to the locomotion transition. As shown in fig. S8 (E and F) (at $I = I_c$), $V_p$ and $\sigma_x$ display areas of both push and pull behavior within wave region 2. It seems clear that the $V_p$-induced decrease of $L_2$ (see Fig. 2F) and the $\sigma_x$-induced decrease of $V_p$ in wave region 2 (Fig. 3A) arise from the same mechanism. Both are generated from the internal competition between opposing domains in wave region 2. In Fig. 4A, at the critical light intensity ($I_c$), we can distinguish three sub-areas (2a, 2b, and 2c) in wave region 2 in which $\sigma_x$ is either negative (2a) or positive (2b and 2c). As the illumination level increases, the net force that generates the retrograde-direct wave transition arises from two effects. First, there are four subregions, that is, wave regions 1, 2a, 2b, and 2c, in the primary wave of complex pulses. Both push (wave region 1) and

![Fig. 3. Effect of light intensity on the kinematic parameters in each region of an IAS.](image)

Mean values of kinematic parameters in each wave region: (A) $(V_p^\text{mean})_i$ versus $I$. (B) $A_i$ versus $I$. (C) $(\sigma_x^\text{mean})_i$ versus $I$. (D) $(\xi')^\text{mean}_i$ versus $I$.
pull effects (wave region 2a) in the primary wave are strengthened with illumination (see fig. S8F). Second, with increasing illumination, two additional subregions (2b and 2c) contribute growing push effects to offset the pull effect from subregion 2a, leading to a nearly constant value of \((\sigma_x)_{\text{mean}}\) with increasing illumination. Briefly, the mixing of the push and pull effects within wave region 2 leads to weakening of the total pull effect and, ultimately, to transition from retrograde to direct wave gel locomotion.

**Dynamic origin of the transition from retrograde to direct wave locomotion**

Why do multiple areas of push and pull effects coexist within wave region 2? We seek here to answer this question by examining the dynamical details of the stress gradient. In the gLSM, \(\sigma_x\) is described by the following equation

\[
\sigma_x = B(\phi, \nu) \phi_x - \chi^* \nu_x
\]

(8)

where

\[
B(\phi, \nu) = 1 - 1/(1 - \nu) + 2 \chi(\nu) + 3 \chi(\nu)^2 - \nu_x + C(\nu_x/\nu^2 - 1)/(1 - \nu_x)
\]

The right side of Eq. 8 contains two composite terms, \(B(\phi, \nu)\phi_x\) and \(\phi \nu_x\) (\(\chi^*\) is set to 1.0), where \(\phi\) and \(\nu\) are dynamical variables of the system. The quantities, \(\nu, \phi, \nu_x, \phi_x, B\) are shown for pulse waves at several illumination intensities in fig. S8 (A to D and H, respectively). Sub-areas of wave region 2 are marked as 2a, 2b, and 2c in each of these plots. From fig. S8H, we see that \(B\) is always negative. The volume fraction, \(\phi\), is positive by definition. As shown in Fig. 4B and fig. S8C, \(\nu_x\) is always positive in wave region 2. So, according to Eq. 8, for \(\sigma_x\) to be positive, \(\phi_x\) must be negative, i.e., we require a pushing force. Further, Fig. 4B shows that negative \(\phi_x\) and small positive \(\nu_x\) occur in areas 2b and 2c of the pulse waves to give a push effect, offsetting the pull effect in area 2a of the pulse waves.

The dynamic term \(\phi_x\) in region 2 of the pulse wave is the key factor for the mode transition of the gel locomotion. As shown in Fig. 4B, \(\phi_x\) is antiphase to \(\nu_x\). The evolutions of \(\phi, \phi_x, \nu, \) and \(\nu_x\) obey reaction-diffusion-mechanics coupling dynamics according to Eq. 9.

As shown in Fig. 4C, the dynamics of local oscillations at the gel center undergo a bifurcation as the level of illumination is increased, changing from simple oscillations (e.g., the inset showing the \(u-v\) limit cycle with a stable periodic orbit at \(I = 0.020\)) to complex oscillations (e.g., the \(u-v\) trajectories at \(I = 0.070, 0.087,\) and 0.110), with the bifurcation occurring at \(I_0\). Because the pulse waves are generated from the diffusion of local oscillations through the gel, such a complex local oscillation appears to be necessary to support the backfired waves (see fig. S8A). In addition, as shown in the insets of Fig. 4C, when \(I > I_0\), with increasing illumination the orbit expands in the \(u-v\) plane, which leads to a greater variation of \(v\) over time and space. The changes in \(\phi\) are strongly correlated with those in \(v\), which accounts for the stimulus-responsive nature of the gel. Thus, over time, an illumination-enhanced oscillatory amplitude of \(v\) leads to an increasing amplitude of \(\phi, \phi_x,\) and \(\nu_x\) oscillations (see fig. S9, C and D). In essence, the parameter-induced instability of simple local oscillations constitutes the dynamic origin of the backfired waves, which leads to the transition from retrograde to direct wave locomotion of the gel in the absence of external asymmetries.

**DISCUSSION**

In summary, we used a modified gLSM model to study the locomotion of a mechanosensitive BZ oscillating gel in a periodic space. Two locomotion modes, that is, retrograde and direct wave locomotion, as well as the transition between them, can be found in such a symmetrical system by adjusting kinetic or mechanical parameters.

![Fig. 4. Mixed push and pull effects within primary wave back and dynamic bifurcation diagram of maximum of \(v\) at the gel center.](image-url)

(A) Sub-areas of \(\sigma_x\) plot at \(I = I_c\). Red and black dashed lines, respectively, denote \(\nu_x = 0\) and \(\nu_x = 0\). (B) Spatial profiles of \(v, \phi_x, \nu_x,\) and \(\nu_x\) within wave region 2 at \(I = I_c\) along the vertical red line in (A). (C) Dynamic bifurcation diagram of maximum of \(v\) at the gel center (\(V_{\text{max-center}}\)) versus \(I\). The insets are the trajectories of the \(u-v\) oscillations at different light intensities.
The numerical results indicate that the retrograde-direct transition of the gel locomotion occurs only if the gel is driven by complex pulse waves with IAS. Detailed analysis reveals that the complex pulse waves host four wave regions, including two push and two pull areas. Changes in the structure of these regions as the parameters are varied modulate the competition between the push and pull effects to generate the transition between modes. We have found that the dominant factor lies in the behavior of the primary wave back, in which homogeneous light-enhanced structural changes of the IAS inhibit the pull effect from this wave region to produce the retrograde-direct transition. Analysis of the chemical dynamics uncovers the origin of the locomotion, which is that the simple pulse waves lose stability to a more complex waveform, as modulated by the homogeneous illumination intensity and/or other parameters.

Further, in the absence of external asymmetric environmental interaction, or even in an isolated system without illumination (movie S1), autonomous active locomotion can be driven by interaction between simple internal nonlinear pulses and active responsive matter within the system. This interaction can be modulated by system parameters to change the locomotion direction and velocity, i.e., interaction with an external asymmetric environment is not a prerequisite for active locomotion. Here, by adjusting the system parameters, complex waves (local complex oscillations or chaos) result in reversal of the direction of locomotion. It is unlikely that such a transition can be realized without the loss of stability of simple pulse waves to more complex waves.

Coupling between nonlinear pulse signals, responsive active matter and a structured boundary [e.g., asymmetric system construction (7, 10) or appendages (32) like setae, mucus, limbs, or wings] should result in more diverse and sophisticated autonomous locomotion and collective behavior without the need for external stimulation.

Last, adaptability and “intelligent” motion of active matter in response to environmental stimuli have been reported, such as dynamic asymmetric light (9) and magnetic field–induced (13) versatile locomotion. Addition of internal nonlinear driving signals and active population communication would increase such systems’ computational ability to respond to environmental stimuli, producing functionally complex and potentially evolutionary motion that would enhance the level of intelligence in active matter. One can envision that designed DNA signals with different chemical reaction networks (33, 34) might be coupled with mechanical forces to construct a biocompatible active system for delivering and releasing drugs by using both internal and external control.

**MATERIALS AND METHODS**

Our BZ gel model is constructed by coupling the Yashin-Balazs model (gLSM) (20–22) with the two-variable form of the Amemiya model of the photosensitive BZR (11, 31). The governing partial differential equations (PDEs) for a 1D gel are

\[
\begin{align*}
\frac{du}{dt} &= -\nabla V_p \\
\frac{dv}{dt} &= -u \nabla V_p + \frac{V_p}{1 - \phi} (u \nabla V_p) + (1 - \phi) \nabla (u \nabla V_p) + F(u, v, \phi, I) \\
\frac{dv}{dt} &= -v \nabla V_p + G(u, v, \phi, I)
\end{align*}
\]

where \( V_p = \nabla \sigma / \zeta(\phi) \)

\[
\nabla \sigma = \nabla (\phi + \ln(1 - \phi) + \chi(\phi)^2 - \chi* \phi v + c_\phi v_0 \left[ (\phi/\phi_0)^{-1} \lambda_2^{-2} - \phi (2 \phi_0)^{-1} \right])
\]

\[
\zeta(\phi) = (\lambda_0 (1 - \phi))^{-1} (\phi/\phi_0)^{3/2}
\]

and \((11, 20–22, 31)\). The functions \( F(u, v, \phi, I) \) and \( G(u, v, \phi, I) \) describe the reaction kinetics of the BZR (modified Oregonator model). We replace the simple Oregonator in the Yashin-Balazs model with a two-variable photosensitive Oregonator model that incorporates both photoinduction and photoinhibition in the Ru(bipy)-catalyzed BZR. The functions \( F(u, v, \phi, I) \) and \( G(u, v, \phi, I) \) that describe the gel-coupled BZR are

\[
\begin{align*}
F(u, v, \phi, I) &= (1 - \phi)^2 u - \phi - (\phi + IP_{1}) (1 - \phi) \left[ u - q (1 - \phi)^2 |u + q (1 - \phi)^2|^{-1} + IP_{2} \right] \\
G(u, v, \phi, I) &= \varepsilon \left[ (1 - \phi)^2 u - (\phi - \delta) v + [0.5 P_{1} + P_{2}] \right]
\end{align*}
\]

Here, \( \varepsilon, f, \) and \( q \) are the Oregonator parameters. \( I \) denotes the dimensionless homogeneous light intensity. \( P_{1} \) and \( P_{2} \) are rate coefficients that characterize the two key photochemical reaction steps. Our simulation of the gel uses 200 grid points (denoted by \( S \)), and the gel length \( L_{gel} \) is set at 126.0 U. All times, space, variables, and parameters are dimensionless (7, 12, 21). The choice of model parameters ensures an oscillatory gel medium.

The PDEs were numerically integrated using the gLSM computational approach (20–22), using an unequal distance grid approximation for the 1D Laplacian operators (22) (first and second derivatives). The integration time step was \( \Delta t = 1.0 \times 10^{-4} \), and the space step was varied according to the local \( \lambda_{ij} \), where \( \lambda_{ij} \) characterizes the degree of swelling along the unrestricted direction of the gel along the long(x) axis of the tube. The other two (restricted) dimensions are characterized by \( \lambda_{rs} \), which is fixed. Initially, the BZ gel was assumed to be in the steady state. The initial volume fraction was \( \phi_{0} = \frac{V_{0}}{L_{gel}^{3}} \). The default values for our model simulation were \( f = 1.0, \varepsilon = 0.3, \chi* = 1.0, P_{1} = 0.124, P_{2} = 0.77, \lambda_{ij} = 1.26, \) and \( \lambda_{rs} = 1.06 \).

The waves can arise spontaneously due to random fluctuations. Initially, the variables \( u, v, \) and \( \phi \) were assigned their stationary values \( (u_{ss}, v_{ss}, \phi_{ss}) \) (21, 22), where \( u_{ss} \) and \( v_{ss} \) are the solutions (22) of \( F(u_{ss}, v_{ss}, \phi_{ss}) = 0, G(u_{ss}, v_{ss}, \phi_{ss}) = 0 \) at \( \chi* = 0.0 \). We then added random noise (50% of the amplitude of \( u_{ss} \)) to all grid points, which induces synchronous oscillations or pulse waves. Figure S1 shows a schematic for a self-oscillating gel driven by pulse waves under periodic boundary conditions. In fig. S2, we examine the relative stability of different wave numbers in the absence of illumination by simulating 1000 independent runs with different random initial conditions at gel sizes \( L = 126.0, 315.0, \) and \( 630.0 \). At all gel lengths studied, the most probable outcome is a single wave propagating over the entire gel. To speed up the calculation, we initialize a pulse wave in the dark with \( K = 1 \) in the subsequent simulations. The illumination
effect on the locomotion of the system is studied by alternately allowing the wave to reach a stable behavior, recording its characteristics, and then increasing the illumination by 0.001. For all spatiotemporal plots, the data spacing in time and \( x \) are 0.5 and 1.0 U, respectively.

We define the curvature \( \kappa(x) \) at each grid point in the gel to characterize the local distortion of the chemical wave, which can be written as

\[
\kappa(x) = \frac{d^2 t}{d x^2} \left( \frac{1}{1 + \left( \frac{dt}{dx} \right)^2} \right)^{3/2}
\]

where \((x, t)\) gives the position of a wave peak. To simplify the calculation, we calculate the local \( \kappa(x) \) across each wave and then store the data in one row, which is assigned to the time \( t \) at which the wave peak crosses the gel center.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/6/18/eaa9125/DC1

**REFERENCES AND NOTES**

1. C. Bechinger, R. D. Leonardo, H. Lüöwen, Active particles in complex and crowded environments. *Rev. Mod. Phys.* **88**, 045006 (2016).
2. S. Ramaswamy, The mechanics and statistics of active matter. *Annu. Rev. Condens. Matter. Phys.* **1**, 323–345 (2010).
3. M. C. Marchetti, J. F. Joanny, S. Ramaswamy, T. B. Liverpool, J. Prost, M. Rao, S. R. Aditi, Hydrodynamics of soft active matter. *Rev. Mod. Phys.* **85**, 1143–1189 (2013).
4. T. Vicsek, A. Zafeiris, Collective motion. *Phys. Rep.* **517**, 71–140 (2012).
5. R. M. Alexander, *Principles of Animal Locomotion* (Princeton Univ. Press, Princeton, 2002).
6. A. J. Išpeert, Biorobotics: Using robots to emulate and investigate agile locomotion. *Science* **346**, 5508–5518 (2017).
7. P. Illien, R. Golestanian, A. Sen, Fuellmer motion: Phoretic motility and collective behaviour of active colloids. *Chem. Soc. Rev.* **46**, 5508–5518 (2017).
8. J. Anderson, Colloid transport by interfacial forces. *Annu. Rev. Fluid Mech.* **21**, 61–99 (1989).
9. S. Palagi, A. G. Mark, S. Y. Reigh, K. Melde, T. Qiu, H. Zeng, S. Y. Reigh, K. Melde, T. Qiu, H. Zeng, C. Parmeggiani, D. Martella, A. Sanchez-Castillo, N. Kapernaum, F. Giesselmann, D. S. Wiersma, E. Lauga, P. Fischer, Structured light enables biomimetic swimming and versatile locomotion of photosensitive soft microrobots. *Nat. Mater.* **15**, 647–653 (2016).
10. A. H. Gelebart, M. D. Jan, M. Varga, A. Konya, G. Vantome, E. W. Weijer, R. L. B. Selinger, D. J. Broer, Making waves in a photoactive polymer film. *Nature* **546**, 632–636 (2017).
11. X. Lu, L. Ren, Q. Gao, Y. Zhao, S. Wang, J. Yang, I. R. Epstein, Photophobic and phototrophic movement of a self-oscillating gel. *Chem. Commun.* **49**, 7690–7692 (2013).
12. L. Ren, M. Wang, C. Pan, Q. Gao, Y. Liu, I. R. Epstein, Autonomous periodic migration of an active material. *Proc. Natl. Acad. Sci. U.S.A.* **114**, 8704–8709 (2017).
13. W. Hu, G. Z. Lum, M. Mastrangeli, M. Sitti, Small-scale soft-bodied robot with multimodal locomotion. *Nature* **554**, 81–85 (2018).
14. V. Petrolli, M. L. Goff, M. Tadrous, K. Martens, C. Allier, O. Mandula, L. Hervé, S. Henkes, R. Skrepnek, T. Boudou, G. Cappello, M. Ballard, Confinement-induced transition between wavelike collective-cell migration modes. *Phys. Rev. Lett.* **122**, 168101 (2019).
15. T. Gao, Z. Li, Self-driven droplet powered by active nematics. *Phys. Rev. Lett.* **119**, 108002 (2017).
16. P. Fischer, A machine from machines. *Nat. Phys.* **14**, 1072–1073 (2018).
17. R. Yoshida, T. Takahashi, T. Yamaguchi, H. Ichijo, Self-oscillating gel. *J. Am. Chem. Soc.* **118**, 5134–5135 (1996).
18. Y. S. Kim, R. Tamate, A. M. Akimoto, Recent developments in self-oscillating polymeric systems as smart materials: From polymers to bulk hydrogels. *Mater. Horiz.* **4**, 38–54 (2017).
19. L. Ren, W. She, Q. Gao, C. Pan, C. Ji, I. R. Epstein, Retrograde and direct wave locomotion in a photosensitive self-oscillating gel. *Angew. Chem. Int. Ed.* **55**, 14301–14305 (2016).
20. V. V. Yashin, A. C. Balazs, Modeling polymer gels exhibiting self-oscillations due to the Belousov-Zhabotinsky reaction. *Macromolecules* **39**, 2024–2026 (2006).
21. V. V. Yashin, A. C. Balazs, Theoretical and computational modeling of self-oscillating polymer gels. *J. Chem. Phys.* **126**, 124707 (2007).
22. O. Kuksenok, V. V. Yashin, A. C. Balazs, Three-dimensional model for chemoresponsive polymer gels undergoing the Belousov-Zhabotinsky reaction. *Phys. Rev. E* **78**, 041406 (2008).
23. V. V. Yashin, A. C. Balazs, Pattern formation and shape changes in self-oscillating polymer gels. *Science* **314**, 798–801 (2006).
24. O. Kuksenok, P. Dayal, A. Bhattacharya, A. C. Balazs, Chemo-responsive, self-oscillating gels that undergo biomimetic communication. *Chem. Soc. Rev.* **42**, 7257–7277 (2013).
25. P. Dayal, O. Kuksenok, A. C. Balazs, Using light to guide the self-sustained motion of active gels. *Langmuir* **25**, 4298–4301 (2009).
26. R. FitzHugh, Impulses and physiological states in models of nerve membrane. *Biophys. J.* **1**, 445–466 (1961).
27. J. Nagumo, S. Arimoto, S. Yoshizawa, An active pulse transmission line simulating nerve axon. *Proc. IRE* **50**, 2061–2070 (1962).
28. R. FitzHugh, Motion picture of nerve impulse propagation using computer animation. *J. Appl. Physiol.* **25**, 628–630 (1968).
29. A. L. Hodgkin, A. F. Huxley, A quantitative description of membrane current and its application to conduction and excitation in nerve. *J. Physiol.* **117**, 500–544 (1952).
30. L. Ren, B. Fan, Q. Gao, Y. Zhao, H. Luo, Y. Xia, X. Lu, I. R. Epstein, Experimental, numerical, and mechanistic analysis of the nonmonotonic relationship between oscillatory frequency and photointensity for the photosensitive Belousov-Zhabotinsky oscillator. *Chaos* **25**, 064607 (2015).
31. T. Amemiya, T. Ohmori, M. Nakaia, T. Yamaguchi, Two-parameter stochastic resonance in a model of the photosensitive Belousov-Zhabotinsky reaction in a flow system. *J. Phys. Chem. A* **102**, 4537–4542 (1998).
32. M. Iwamoto, D. Ueyama, R. Kobayashi, The advantage of mucus for adhesive locomotion in gastropods. *J. Theor. Biol.* **353**, 133–141 (2014).
33. S. S. Wang, A. D. Ellington, Pattern generation with nucleic acid chemical reaction networks. *Chem. Rev.* **119**, 6370–6383 (2019).
34. I. R. Epstein, Making waves with DNA. *Physics* **8**, 12 (2015).

**Acknowledgments:** We are grateful to the Advanced Analysis and Computation Center of CUIMT for the award of CPU hours to accomplish the computations in this paper. **Funding:** This work was supported by the National Natural Science Foundation of China (grant nos. 21572882 and 21972165), the Natural Science Foundation of Jiangsu Province (grant no. BK20171186), and the NSF (grant no. CHE-1856484). **Author contributions:** All authors contributed to discussion, the execution of the study, and theoretical analysis. L.R., Q.G., and I.R.E. conceived the research and wrote the paper. L.R. and L.Y. developed the numerical codes. L.R., L.Y., Q.G., R.T., and J.W. performed the calculation and prepared the figures and movies. Q.G. and I.R.E. supervised the research. **Competing interests:** The authors declare that they have no competing interests. **Data and materials availability:** All data needed to evaluate the conclusions in this paper are present in the paper and/or the Supplementary Materials. Additional data related to this paper may be requested from authors.

Submitted 21 October 2019
Accepted 10 February 2020
Published 1 May 2020
10.1126/sciadv.aaz9125

**Citation:** L. Ren, L. Yuan, Q. Gao, R. Teng, J. Wang, I. R. Epstein, Chemomechanical origin of directed locomotion driven by internal chemical signals. *Sci. Adv.* **6**, eaaz9125 (2020).