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Temporal Light Modulation of Photochemically Active, Oscillating Micromotors: Dark Pulses, Mode Switching, and Controlled Clustering

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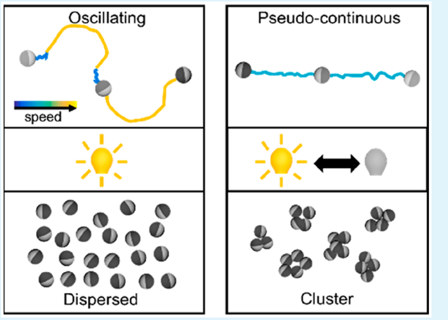
ABSTRACT: Photochemically powered micromotors are prototype microrobots, and spatiotemporal control is pivotal for a wide range of potential applications. Although their spatial navigation has been extensively studied, temporal control of photoactive micromotors remains much less explored. Using Ag-based oscillating micromotors as a model system, a strategy is presented for the controlled modulation of their individual and collective dynamics via periodically switching illumination on and off. In particular, such temporal light modulation drives individual oscillating micromotors into a total of six regimes of distinct dynamics, as the light-toggling frequencies vary from 0 to 10^3 Hz. On an ensemble level, toggling light at 5 Hz gives rise to controlled, reversible clustering of oscillating micromotors and self-assembly of tracer microspheres into colloidal crystals. A qualitative mechanism based on Ag-catalyzed decomposition of H_2O_2 is given to account for some, but not all, of the above observations. This study might potentially inspire more sophisticated temporal control of micromotors and the development of smart, biomimetic materials that respond to environmental stimuli that not only change in space but also in time.



KEYWORDS: *colloids, micromotors, oscillators, temporal illumination, clusters*

1. INTRODUCTION

The last two decades have witnessed a surge of research interests in micromotors (or "microrobot"), smart colloids that convert external energy stored in the environment into autonomous, microscopic motion,[1−5] due to their usefulness in fundamental studies of active matters,[6−8] as well as in potential applications.[9−13] Prominent examples of proposed applications for micromotors include microcleaners that sense and clean contaminants in polluted water,[14−16] and biomedical nanobots that sense, deliver, capture, and cure in human bodies.[17−20] These scenarios, and many others, often involve complex environments[21−27] and thus require a precise control over where the micromotors are located (space), as well as when they carry out specific functions (time), at both an individual and ensemble level.[28−30]

Strategies of spatiotemporal control of micromotors are therefore urgently needed. In this regard, the spatial navigation of a micromotor has been extensively studied, and reported strategies include external magnetic guidance,[31,32] biomimetic chemotaxis,[33−38] or physical microstructures that steer micromotors along specific pathways.[24−26,39−42] The effects of these navigation strategies can be monitored via advanced medical imaging techniques, even in vivo.[17,43] Temporal control of the dynamics of a micromotor, however, is much less studied. This becomes especially important for micromotors moving in complex environments, where a single mode of dynamics, i.e., the classic directional motion with a variable speed, might not suffice.

A convenient way to achieve spatiotemporal modulation is by electromagnetic waves, and micromotors propelled by light emerge as a good model system for this purpose.[44−46] These photoactive micromotors have in recent years gained popularity for being remotely controllable and tunable by varying light in its directionality,[47,48] intensity,[49] wavelength,[50,51] or polarization.[52] However, the effect of illuminating frequency on the dynamics of a micromotor, as well as their collective behaviors, has been largely unexplored, even though temporal modulation of light is critical in a variety of dynamic processes ranging from the ocular growth of young animals[53] to pulsed laser sources.[54,55] This lack of research might suffer from two limitations. First, halogen or mercury light sources are often used to illuminate photoactive micromotors, yet the nature of their operating mechanisms prohibits fast light switching. Second, the speeds of most micromotors scale (more or less) linearly with the applied light intensity and drop to zero almost

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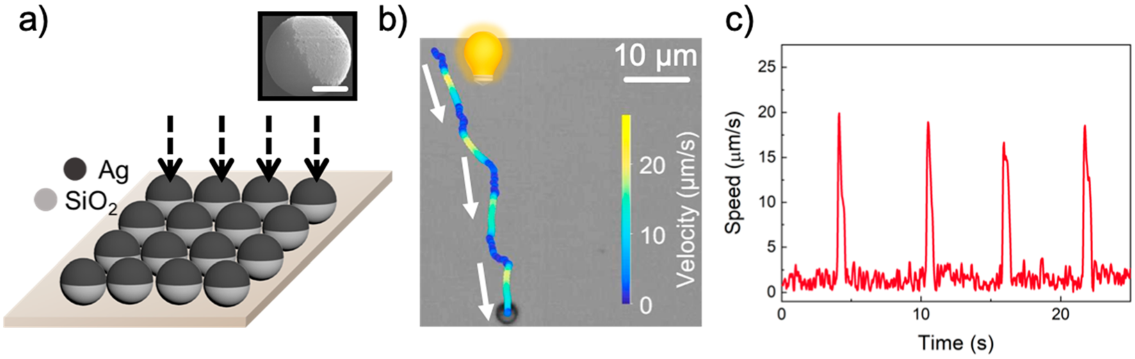

Figure 1. Fabrication and intrinsic oscillations of SiO_2−Ag Janus motors. (a) Janus motors are fabricated by depositing Ag on SiO_2 microspheres. Inset: scanning electron micrograph of a SiO_2−Ag Janus particle (scale bar 1 μm). (b) Trajectory (speeds color-coded) and (c) instantaneous speeds of one Janus motor pulsating forward away from its Ag cap for 30 s at 0.75 wt % H_2O_2, 400 μM KCl, and light intensity of 500 mW/cm^2.

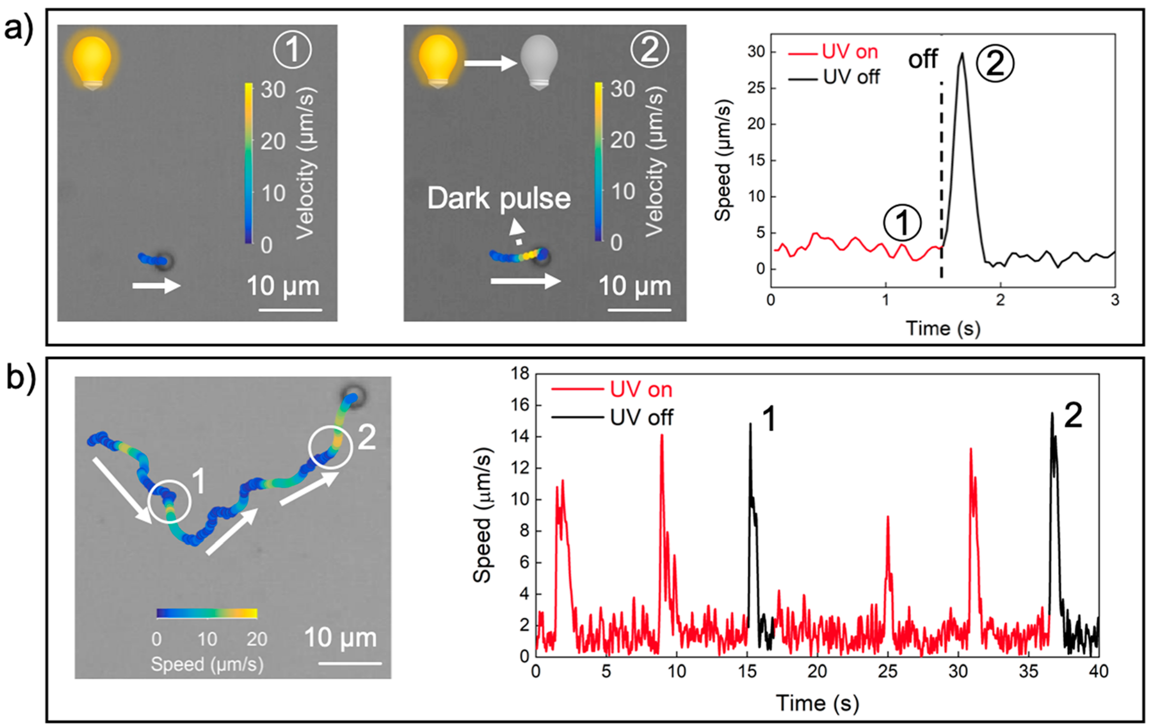

Figure 2. "Dark pulses" of a single oscillators. (a) Trajectories (left and center) and instantaneous speeds (right) of a Janus motor when turning off the UV light after 1.5 s of illumination with 500 mW/cm^2 at 0.75 wt % H_2O_2 and 400 μM KCl. (b) Trajectory (left) and instantaneous speeds (right) of a Janus motor during two cycles of switching the light on and off at 0.5 wt % H_2O_2, 400 μM KCl and 500 mW/cm^2 light intensity. Numerical labeling in micrographs match those in the data plots.

instantly when light is switched off. Toggling light therefore grants little benefit, as the micromotor switches between "go" and "stop".

A micromotor that shows a continuous variation of behaviors under different illuminating frequencies, on the other hand, might usher in a rich phase of dynamics and an expanded toolbox useful for a variety of applications. Photoactive micromotors with a response to light that is entirely different from typical micromotors are needed for this purpose. In this article, we show a prototype micromotor that points to this direction. Our strategy stems from recent reports by the Sen lab and us,[56−59] where silver (Ag)-containing microparticles show spontaneous and regular oscillations in speeds when immersed in H_2O_2 and KCl, and illuminated with *continuous lighting*. Moreover, pioneering experiments by Steinbock et al. and Petrov et al. suggest rich nonlinear effects of chemical oscillators when subject to temporal light modulation.[60,61] Inspired by these earlier studies, we here take oscillating Ag Janus micromotors as a model system and examine the changes in their dynamics, both individually and among a population, when light is applied *intermittently* (i.e., temporal modulation at a switching frequency of 10^{-1} to 10^3 Hz).

By solely varying the light switching frequency while keeping all other conditions constant, an oscillating micromotor exhibits one of six distinct modes of dynamics: intrinsic oscillation, "dark pulses", Brownian motion, triggered oscillation, pseudocontinuous motion, and semi-intrinsic oscillation. Details of each observation are given in Results and Discussion. Moreover, we show that temporal modulation of a population of oscillating micromotors leads to controlled clustering, and close packed crystals of tracer colloids are

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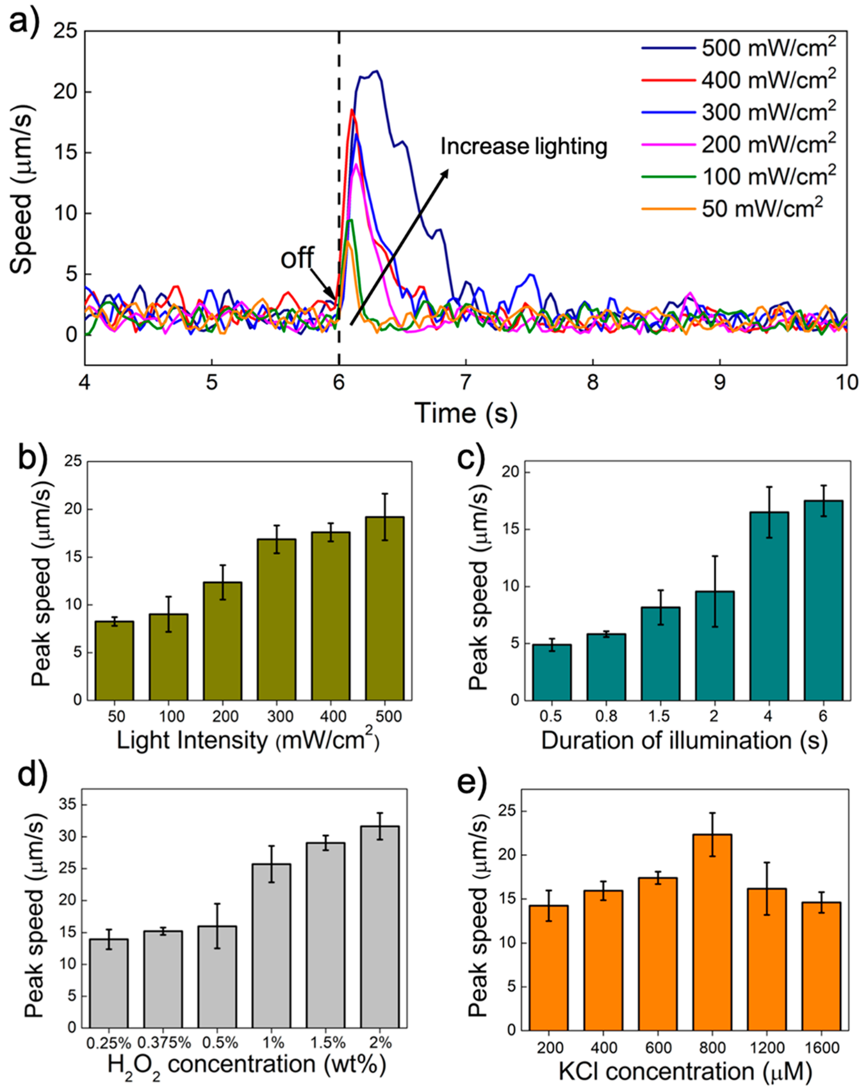

Figure 3. Modulating the dark pulses of a SiO_2−Ag Janus motor. (a) Instantaneous speeds of motors before and after shutting off light of different intensities. Experiments were conducted with 0.5 wt % H_2O_2 and 400 μM KCl. (b−e) Peak speeds of dark pulses by varying (b) light intensity (extracted from panel a), (c) duration of illumination, (d) concentrations of H_2O_2, and (e) concentrations of KCl. Experimental conditions are 0.5 wt % H_2O_2, 400 μM KCl, and 3 s of illumination at 500 mW/cm^2 unless a parameter is the variable.

formed. A qualitative mechanism is proposed to partially rationalize the above observations. We anticipate this study to inspire more sophisticated temporal control of micromotors, oscillating or not, that ultimately contributes to the development of smart, biomimetic materials that adapt to a changing environment, useful for sensing and actuation applications.

2. RESULTS AND DISCUSSION

2.1. Single Oscillators: "Intrinsic Oscillations" upon Continuous Lighting. Before we introduce the various modes of motion upon temporal modulation of light, we first briefly review the oscillating dynamics of a Ag oscillating motor under continuous light, as has been first described by the Sen lab,[56] and more recently by us in more detail.[57]

In particular, we focus on Janus SiO_2−Ag microparticles, which were prepared by physically depositing 50 nm of Ag on one side of SiO_2 microspheres of 3 μm in diameter (Figure 1a). They were suspended in an aqueous solution that contained H_2O_2 (0.25−2 wt %) and KCl (200−1600 μM) and exposed to 365 nm UV light (generated by a LED lamp). More details of the experiments can be found in Experimental Section. It has been previously shown that these SiO_2−Ag Janus particles under such conditions would alternate between a long resting stage and an episode of short, fast motion with the SiO_2 hemisphere forward (see Figure 1b,c and video S1). This interesting, nonlinear dynamics is believed to arise from a spontaneous oscillation between a slow oxidation of Ag by H_2O_2 into AgCl (eq 1) and a photodecomposition of AgCl back into Ag (eq 2) that is autocatalytic due to the production of plasmonic Ag nanoparticles:[56,57]

$$2Ag + H_2O_2 + 2H^+ + 2Cl^- \rightarrow 2AgCl + 2H_2O \qquad (1)$$

$$4AgCl + 2H_2O \xrightarrow{\hbar\nu} 4Ag + O_2 + 4H^+ + 4Cl^- \qquad (2)$$

Equation 2 releases a flux of H^+ and Cl^- that diffuses away from the active particle in different rates. As a result, an electric field is spontaneously produced and propels the negatively charged active particle by a mechanism termed "self-diffusiophoresis".[57,62,63] Periodic release of ions thus give rise to periodic oscillation in particle speeds. Note that such

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oscillation under continuous lighting occurs spontaneously, and its dynamics is governed mainly by light intensities and chemical concentrations. To distinguish it from similar pulses triggered by switching light, discussed later, these spontaneous pulses are hereafter referred to as "intrinsic oscillations".

Besides periodically pulsating under illumination, the same SiO_2−Ag motors exhibit a single pulse of motion upon switching off the light (see SI video S2), termed "dark pulses". Figure 2a shows the trajectory of one SiO_2−Ag motor before and after light was turned off, and a dark pulse is characterized by a sharp rise and a quick decay of its instantaneous speeds. The peak speed of a dark pulse can even surpass that of intrinsic oscillations. During this dark pulse, the Janus motor moved away from the Ag cap, as it would during intrinsic oscillations. A few details on the dark pulses are worth mentioning. First, such a dark pulse was a singular but not random event. As shown in the Figure S1 and video S3, a few Janus particles pulsated forward at their own rhythms under continuous illumination but collectively and simultaneously dashed forward once light was turned off. These motors then stayed diffusive afterward in the darkness. Second, the dark pulse is insensitive to how fast light is switched off (i.e., turned off instantly or dimmed down slowly, characterized by a "ramp time") but is sensitive to the initial intensity from which light decreases. A dark pulse always occurred as the light was reduced below a certain threshold intensity (tentatively determined to be ∼40%). These data are given in the Supporting Information (Figures S2 and S3).

Does a dark pulse change the surface properties of a Janus particle and thus how it pulsates when illuminated again? To answer this question, we have manually switched off the light for a pulsating SiO_2−Ag Janus motor before switching it back on after a short dark period (Figure 2b, video S4). This on/off switching process was repeated a few times. The motor exhibited a sharp dark pulse every time the light was turned off but returned to intrinsic oscillation when the light was turned back on. The periods and intensity of its intrinsic oscillation remained more or less the same despite of dark episodes. These results suggest that dark pulses do not permanently modify the motor, and that intrinsic oscillation and dark pulses are likely independent processes.

2.2. Modulating Dark Pulses. Understanding and using dark pulses require a systematic investigation of the effect of experimental parameters on the pulsing dynamics. These parameters include concentrations of chemicals such as H_2O_2 and KCl, which are essential ingredients for a SiO_2−Ag Janus particle to oscillate. In addition, lighting conditions, such as illumination intensity and duration before light is switched off, are found to significantly affect dark pulses, too. These results, presented in Figure 3 and discussed in the following, suggest that dark pulses are not random and trivial process but rather an inherent feature whose intensity is tunable by judiciously choosing combinations of experimental parameters.

For example, from the instantaneous speeds of an oscillating motor in Figure 3a and b, we can clearly see that the amplitude of the dark pulse can be regulated easily by modulating light intensity before switching light off. In addition, results from Figure 3c,d show that the dark pulses are more intense if particles are illuminated for a longer period of time and at high concentrations of H_2O_2. With respect to the supply of KCl (Figure 3e), the amplitude of a dark pulse increased then decreased as the concentration of KCl increased. This trend agrees with previous studies where elevated ionic strength results in a decrease in speed of phoretic colloidal motors driven by a self-generated electric field,[64−67] such as those powered by self-electrophoresis or self-diffusiophoresis. This reasoning was further supported by adding KNO_3 to the solution, where the peak speeds of dark pulses monotonically decreased as KNO_3 concentration increased (Figure S4). Finally, preliminary results show that Janus particles made of SiO_2 microspheres of 2 μm diameter showed a stronger dark pulse than those of 3 μm diameter (Figure S9). The effect of sizes on the dynamics of oscillating micromotors will be investigated in more details in a separate study.

2.3. Mechanism of Dark Pulse. Before we move on to other modes of motor dynamics upon switching light, we attempt to propose a qualitative mechanism to explain dark pulses, which serve as the building block for many of the observations described below. The process of dark pulse is tentatively dissected into the following three stages.

The initial continuous illumination sets the stage for what happens when lighting is removed. According to the mechanisms proposed in the literature,[56,57] upon being added in H_2O_2 and KCl, the silver coating on a Janus particle converts to AgCl. At the same time, the produced AgCl begins to decompose back into Ag under UV light. These two reactions, described by eqs 1 and 2, produce a heterogeneous nanostructure containing Ag nanoparticles decorated on AgCl. This Ag@AgCl nanostructure serves as a photocatalyst for the decomposition of H_2O_2.[68] Specifically, upon absorbing a photon, AgCl produces an electron−hole pair. The hole oxidizes water into O_2 (eq 3), while the electron migrates to AgCl reduces H_2O_2 into H_2O (eq 4), as in the following:

$$\text{on Ag:}\quad H_2O_2 + 2h^+ \rightarrow 2O_2 + 2H^+ \tag{3}$$

$$\text{on AgCl:}\quad H_2O_2 + 2H^+ + 2e^- \rightarrow 2H_2O \tag{4}$$

$$\text{overall:}\quad H_2O_2 \xrightarrow{\text{Ag@AgCl}} H_2O + \frac{1}{2}O_2 \tag{5}$$

Importantly, we speculate that the photoelectrochemical decomposition of H_2O_2 suppresses the chemical catalysis on the surface of Ag toward the decomposition of H_2O_2. Once the light is turned off, however, photocatalysis (eqs 3−5) stopped and the Ag-catalyzed H_2O_2 decomposition therefore became the only reaction. This is indirectly supported by the fact that a great number of bubbles that appeared as light was turned off (video S10). The concentration gradients of H_2O_2 and O_2 then lead to a diffusiophoretic force, with possible electrokinetic contributions, perhaps in the same way Pt-coated Janus particles are able to propel in H_2O_2 away from the Pt cap.[69] We also performed a simple experiment (video S5) showing that SiO_2−Ag Janus particles that were previously undergoing Brownian motion suddenly moved away from their Ag caps upon adding one drop of H_2O_2 (final concertation 0.5 wt %), which to some extent lends credibility to the mechanism above.

For the third stage, a dark pulse usually stopped within ∼1s, as shown clearly in Figure 2a and Figure 3a. The pulsed activity in the dark was not sustainable or repetitive, most likely because Ag on the particle surface, while catalyzing H_2O_2 decomposition, was also oxidized by H_2O_2 into AgCl (eq 1).[56,57] Once it was converted, catalysis stopped, and propulsion consequently ceased.

The above mechanism can qualitatively explain the effect of varying experimental parameters on the dynamics dark pulses

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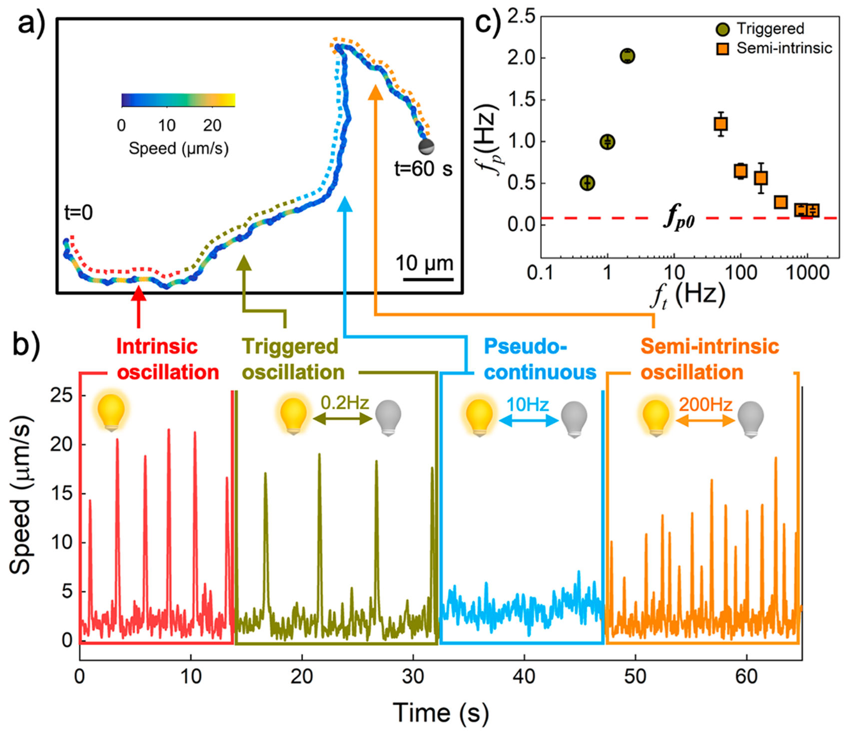

Figure 4. Mode switching of an oscillating motor via light toggling. (a) Trajectory and (b) instantaneous speed of an oscillating motor at continuous lighting (red), and toggling frequency of 0.2 Hz (green), 10 Hz (blue), and 200 Hz (orange), respectively. Light was toggled on-the-fly. Data were extracted from video S6. Experiments were conducted at 1 wt % H_2O_2, 400 μM KCl, and 400 mW/cm^2 light intensity. (c) Frequency of the oscillating motor (f_p) depends on the frequency of toggled light (f_t). The intrinsic frequency (f_{p0}) is labeled for reference. Note that data in panel c were acquired from a different set of experiments and do not match those from panel a or b.

shown in Figure 3. For example, strong intensity (Figure 3a,b) and long exposure (Figure 3c) of illumination is helpful for the generation of Ag on the particle surface in stage 1, which increases the rate of H_2O_2 decomposition during stage 2 and ultimately leads to a stronger dark pulse. Similarly, a higher concentration of H_2O_2 is also beneficial to this catalytic reaction (Figure 3d) and thus a stronger pulse. Higher concentrations of Cl^- lead to more AgCl during stage 3. This in turn produces more Ag particles after switching on light, resulting in a stronger dark pulse.

2.4. Triggered Pulses and Pseudocontinuous Motion upon Toggling Light. Because switching light off produces a dark pulse, periodically switching light on and off, referred hereafter as "toggling light", naturally leads to periodic pulses each time the light is turned off. This "triggered oscillation" is phenomenological similar to "intrinsic oscillations", in the sense that motors oscillate at a fixed frequency in both cases, defined as f_p (where p stands for particle). However, triggered motors are entrained to the light toggling frequency, defined as f_t (where t stands for trigger). Below, we describe the various dynamics—intrinsic oscillation, triggered oscillation, pseudo-continuous motion, and semi-intrinsic oscillation—of a Janus Ag micromotor upon periodic lighting, as the toggling frequencies was raised from 0 Hz (continuous lighting) to ~kHz (video S6 and Figure 4).

A Ag Janus particle was first exposed to continuous lighting and oscillated at f_p of ~0.4 Hz (Figure 4, the first regime colored in red). At very low toggling frequency, $10^{-1}<f_t < 6$ Hz, the frequency of a micromotor f_p is entrained to f_t, because a dark pulse is produced each time light is switched off (Figure 4b, the second regime colored in green). At an intermediate frequency, $6 < f_t < 30$ Hz, the oscillation of a micromotor becomes less noticeable (Figure 4b, the third regime colored in blue). Instead, the motor moves sluggishly with its SiO_2 hemisphere leading, because the removal of each short episode of light gives a quick but weak burst of action. These small bursts combine into a weakly intermittent motion that appears to be continuous, therefore the name "pseudocontinuous". It is worth noting that the average speeds of Janus motors in pseudocontinuous mode can be continuously regulated by varying light intensity and f_t, shown in Figures S5 and S6. Finally, at higher toggling frequencies, $30 < f_t < 10^3$ Hz (Figure 4b, the last regime colored in orange), episodes of lighting become so dense that a micromotor oscillates as if under continuous lighting, with a frequency f_p that is higher than its intrinsic frequency f_{p0} (under continuous lighting) but lower than the toggling frequency f_t. This regime is termed "semi-intrinsic oscillation", with an f_p that is continuously tunable by varying f_t, reaching a plateau at an f_t of kHz (Figure 4c). More examples of the above dynamics under various toggling frequencies can be found in Figure S7. In all these experiments, duty cycles of each lighting cycle were set to be 0.5. Moreover, we note that the threshold f_p that separates the pseudocontinuous regime and the semi-intrinsic oscillation regime is sensitive to concentrations of H_2O_2 and KCl, discussed in Figure S8.

2.5. Reversible Clustering of Oscillating Motors upon Temporal Light Modulation. Simultaneous modulation of an ensemble of micromotors could be useful in many proposed applications, as demonstrated in a number of recent studies with various strategies.[22,36,70,71] In this section, we show that temporal light modulation is a powerful technique that not only switches oscillating micromotor among various modes of individual dynamics (discussed above) but also transforms a

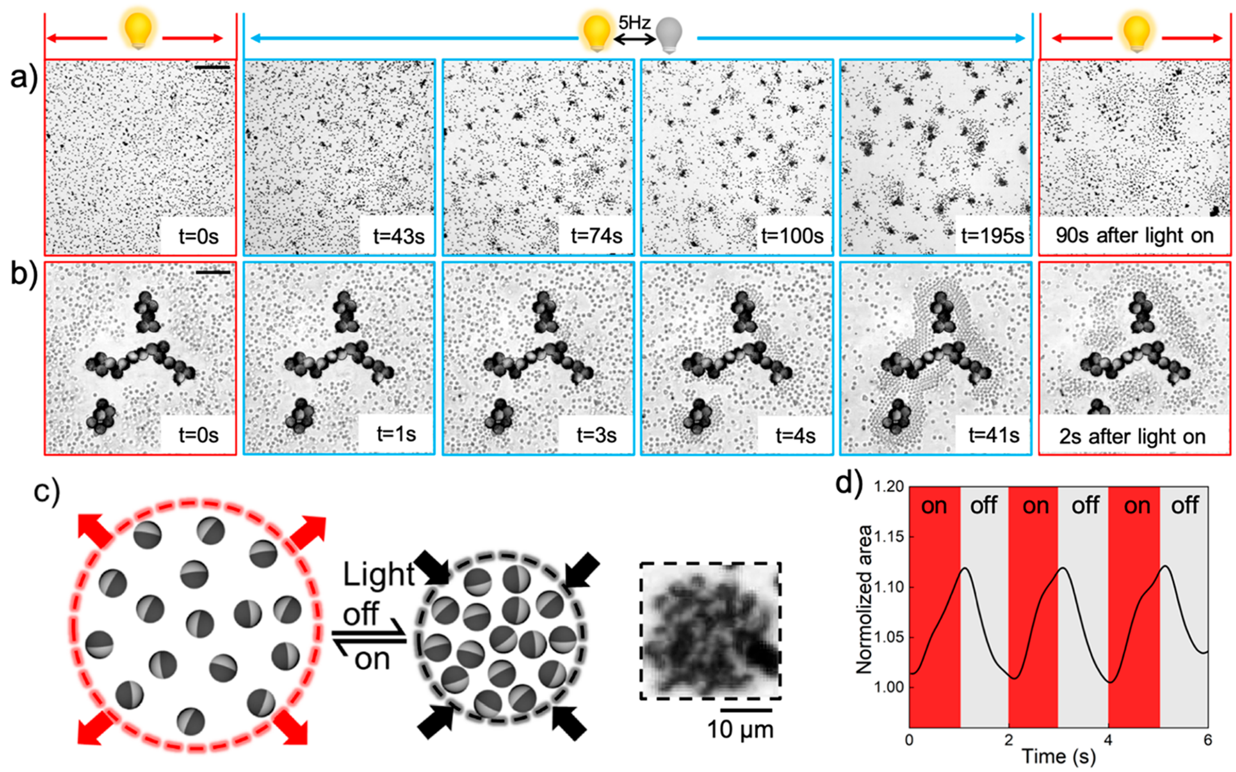

Figure 5. Reversible clustering of oscillating motors upon temporal light modulation. Optical micrographs of (a) reversible transition between dispersion and aggregation pattern (scale bar 100 μm) and (b) reversible self-organization of 1 μm SiO_2 tracer particles (scale bar 10 μm) by switching between continuous and intermittent (5 Hz) illumination. (c) Scheme of the contraction and expansion of a cluster by toggling light on/off. Inset: Janus particles orient randomly within a cluster. (d) Normalized area profile of one cluster over time during toggling light on/off at 0.5 Hz. Experimental conditions are 0.5 wt % H_2O_2, 400 μM KCl, and 500 mW/cm^2 light intensity.

group of oscillating micromotors from a uniform dispersion into clusters.

Our experiments, documented in video S7, began with continuous lighting, under which Ag Janus particles were somewhat uniformly dispersed in the solution without any significant aggregation (Figure 5a, $t = 0$). Waves of action propagated through the entire population, a common phenomenon[56] for oscillating motors that will be the focus of a different study. As the light was toggled on and off periodically at 5 Hz, however, particles gradually aggregated into many small clusters, which further served as nuclei to attract other nearby particles, eventually forming larger clusters within a few minutes (Figure 5a). As the lighting was switched back to be continuous, these clusters slowly expanded, and the population reversed back to be uniformly dispersed (Figure 5a, the last panel).

To better understand this aggregation process, a lower toggling frequency f_t of 0.5 Hz was applied, upon which a cluster of micromotors expanded and contracted periodically following f_t, as the light was switched on and off, respectively (video S8, Figure 5c). Its contraction and expansion was quantified by tracking the cluster area that oscillated over time (Figure 5d). Moreover, we note that, as shown in inset of Figure 5c, a cluster consisted of particles oriented in all directions, suggesting that they were not phoretically migrating toward the cluster center but rather were swept in by convection (e.g., substrate osmosis).

In addition to inducing clustering for oscillating micromotors, toggling light can drive the reversible self-organization of passive particles (video S9, Figure 5b). Although an earlier study by us has shown that,[57] under *continuous* illumination, tracer particles were periodically repelled and attracted from a Ag Janus oscillating motors, regular crystals were never observed, unlike the case with oscillating Ag_3PO_4 microparticles.[59] However, upon intermittent illumination at 5 Hz, negatively charged SiO_2 microspheres rapidly migrated and assembled around clusters of Ag Janus particles that were stuck on the substrate, forming crystalline clusters within seconds. A closer look reveals that SiO_2 microspheres were attracted more strongly than being repelled as light was turned off and on, respectively, thus achieving a net migration over time. The self-assembly process can be arbitrarily turned on and off by switching between periodic lighting and continuously lighting, respectively.

How do we understand the contraction and expansion of clusters (as well as tracer particles), in connection to the individual dynamics of oscillating micromotors, during light toggling? It is easy to see that clusters and tracer particles are undergoing triggered oscillation, at a toggling frequency of 5 Hz, in the same way as that seen with individual motors driven at the same frequencies (i.e., Figure 4b). Therefore, one naturally expects that the dark pulses for individual motors and the contraction of a cluster, both occurring as the light was switched off, share the same origin, i.e., the catalytic decomposition of H_2O_2 in the darkness. However, this effect might be complicated by additional contributions from substrate-driven osmosis, which becomes significantly stronger for clusters of active particles fixed to the substrate. In addition, we note potential contributions from transient photocurrents associated with photochemical reactions on Ag/AgCl surfaces,

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an effect that is under investigation. A detailed mechanism is currently lacking for the collective behaviors of oscillating micromotors under temporal light modulation.

3. CONCLUSION

To summarize, we have systematically studied a rich variety of dynamics for individual and a population of oscillating, Ag Janus micromotors, upon temporal light modulation. More specifically, we observe a surprising pulse of movement by these oscillating micromotors upon switching light off, termed "dark pulses", with a peak intensity tunable by experimental conditions. Based on this effect, periodically switching on and off illumination at low frequencies entrains a micromotor to an externally triggered frequency f_t, so it exhibits triggered oscillation ($f_t \sim 10^0$ Hz) or pseudocontinuous motion ($f_t \sim 10^1$ Hz). Higher toggling frequencies at 30−10^3 Hz give rise to a semi-intrinsic oscillation phase, where the oscillation frequency of a micromotor continuously varies with the illumination frequencies. At a population level, the same periodic illumination collects well-dispersed oscillating micromotors into many tight clusters that can be redispersed by switching the light on. Tracer microspheres under the same toggled lighting form close packed crystals around oscillating micromotors.

These observations are interesting for a number of reasons. First, although the oscillating micromotors described here only serve as a preliminary model, it inspires other stimuli-responsive materials with built-in temporal components. Second, the various modes of dynamics for an oscillating micromotor at different toggling frequencies (Figure 4) suggest a mismatch between the kinetics of the chemical reactions involved in the system, thus presenting an opportunity to probe the underlying mechanism for this oscillatory system (eqs 1 and 2). Finally, the controlled, reversible clustering presented at the end of this article is a useful technique to generate separated clusters of oscillating micromotors, which help us understand waves and interparticle communications, and help in the design of ensemble of micromachines that communicate and coordinate. This topic will be explored in a later study.

4. EXPERIMENTAL SECTION

4.1. Fabrication of Janus SiO_2−Ag Motor. We used the drop-casting method to prepare a monolayer of SiO_2 (3 μm in diameter) on a glass slide. Specifically, a certain amount of SiO_2 was suspended in ethanol and was dispersed by ultrasound. The suspension was then drop-casted on glass slides. The monolayer of SiO_2 was coated with a 40 nm silver layer via electron beam evaporation (e-beam evaporator HHV TF500). The as-prepared SiO_2−Ag Janus microparticles were released from the glass slides by ultrasonication and resuspended in deionized water. The typical morphology of the prepared SiO_2−Ag Janus particles is shown in Figure 1a inset. Although SiO_2 microspheres were chosen for this study, microspheres of other inert materials such as PMMA or PS give qualitatively similar results.

4.2. Motor Motion Experiment. The suspension of SiO_2−Ag particles was transferred into a rectangular capillary tube (VitroCom no. 3520-050, thickness 200 μm) or homemade chamber and observed from underneath with an inverted optical microscope (Olympus IX71), and UV light was applied from the top. The intermittent UV light was carried out with a LED UV light source (Thorlab M365LP1-C1, peak wavelength at 365 nm, maximum switching frequency 1.5 kHz). The light source was connected to a function generator (Agilent33210A), so that light signals can be temporally modulated. A ring-shaped lamp of white LED is placed around the UV lamp to provide ambient lighting for imaging, and we confirm that this white light is not sufficient, either in terms of intensity or in wavelength, to activate our micromotors or to induce their dark pulses. The motion of the Janus SiO_2−Ag motor was recorded by a CMOS camera (GS3-U3-51S5C-C, Point Gry) typically at 30 frames per second (fps). These videos were then analyzed by MATLAB codes courtesy of Hepeng Zhang from Shanghai Jiaotong University. Particle coordinates were extracted and were used to obtain trajectories and instantaneous speeds of motors.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsami.9b22342.

Intrinsic oscillations of a photochemically active motor upon continuous lighting (MP4)

"Dark pulses" of an oscillating motor (MP4)

"Dark pulses" of a few Janus motors (MP4)

Switching between intrinsic oscillation and "dark pulse" of an oscillating motor (MP4)

Propulsion of a few Janus motors when fueling H_2O_2 (MP4)

Mode switching of oscillating motor via light toggling (MP4)

Reversible transition between dispersion and aggregation pattern of oscillating motors upon temporal light modulation (MP4)

Reversible transition between expansion and contraction of a cluster of micromotors (MP4)

Reversible self-organization of passive particles on the micromotors upon temporal light modulation (MP4)

Many bubbles emerged once light is turned off among a suspension of SiO_2−Ag motors (MP4)

Description of Figures S1−S8 and videos S1−S10 in the text (PDF)

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Notes

The authors declare no competing financial interest.

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