

# Janus Swarm Metamaterials for Information Display, Memory, and Encryption

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Metamaterials are emerging as an unconventional platform to perform computing abstractions in physical systems by processing environmental stimuli into information. While computation functions have been demonstrated in mechanical systems, they rely on compliant mechanisms to achieve predefined states, which impose inherent design restrictions that limit their miniaturization, deployment, reconfigurability, and functionality. Here, a metamaterial system is described based on responsive magnetoactive Janus particle (MAJP) swarms with multiple programmable functions. MAJPs are designed with tunable structure and properties in mind, that is, encoded swarming behavior and fully reversible switching mechanisms, to enable programmable dynamic display, non-volatile and semi-volatile memory, Boolean logic, and information encryption functions in soft, wearable devices. MAJPs and their unique swarming behavior open new functions for the design of multifunctional and reconfigurable display devices, and constitute a promising building block to develop the next generation of soft physical computing devices, with growing applications in security, defense, anti-counterfeiting, camouflage, soft robotics, and human-robot interaction.

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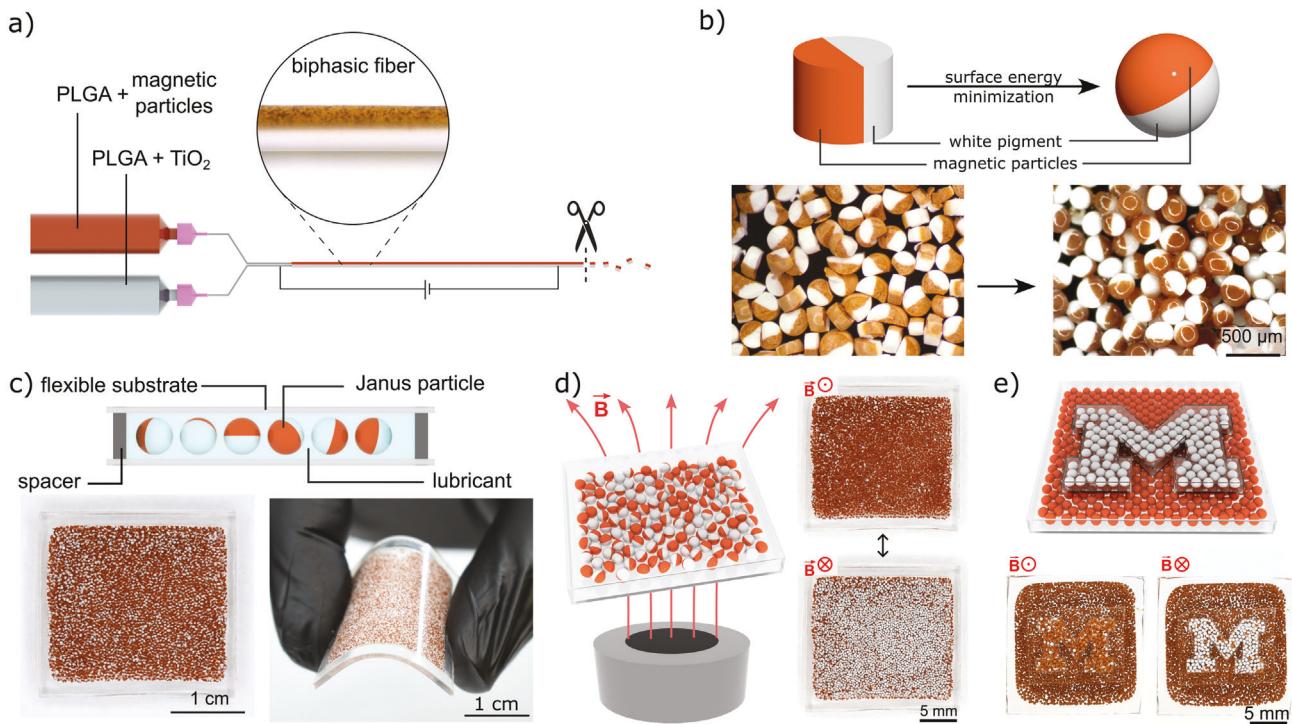
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## 1. Introduction

Metamaterials and metasurfaces, consisting of engineered structures with periodic arrangements of minimal building blocks, have been increasingly gaining interest to embody intelligence (processing and display of complex information) in responsive physical constructs.<sup>[1,2]</sup> Particularly, mechanical metamaterials have shown exciting potential in performing computing abstractions in physical systems,<sup>[3]</sup> such as mechanical memory for information storage,<sup>[4]</sup> physical mechanologic (from Boolean functions to integrated circuitry),<sup>[5-7]</sup> and encryption.<sup>[8,9]</sup> However, architected mechanical devices and responsive structures rely on compliant mechanisms (folding, bending, buckling, etc.) to achieve predefined states, which are difficult to miniaturize due to limitations in distributed actuation mechanisms at small scales, resulting in rather large structures

with limited portability.<sup>[2,10]</sup> Furthermore, metamaterials are often designed to fulfill a specific task with predefined target transitions from an initial state to a fixed final output configuration, with limited reconfigurability from a specific starting design. Instead of encoding information in the architected structure only, engineering these functions through multifunctional materials presents new opportunities for wide-spectrum programmability, miniaturization, and reconfigurability in metamaterials for performing advanced display and computing functions without compromising the device design space.<sup>[11-13]</sup>

In parallel to mechanical metamaterials, a broad diversity of responsive materials has been developed for non-emissive display functions, using physical changes in the material and incoming environmental light to display information.<sup>[14,15]</sup> Unlike traditional emissive displays, which use a front light or a backlight such as light-emitting diodes (LEDs) or liquid crystal displays (LCDs), non-emissive displays do not require a dedicated light source and use only reflected ambient light instead. As a result, they have near-zero power consumption, are highly energy-efficient, have high contrast, wide viewing angles, flexibility, and overall better readability and performance in a wide range of environmental conditions, which makes them very attractive for use in portable and wearable devices.<sup>[14,16]</sup> Common actuation methods in non-emissive display technologies include thermochromic (change color with temperature),<sup>[17]</sup> electrochromic (change color with applied current through electrochemical reactions),<sup>[15]</sup> electrophoretic/magnetophoretic



**Figure 1.** Fabrication of magnetoactive Janus particles (MAJPs) and display system. a) Electrohydrodynamic (EHD) co-jetting to fabricate bicompartimental PLGA fibers with magnetic particles and white pigment, followed by microsectioning to slice into biphasic cylinders. b) Shape transformation from Janus cylinders to particles through surface energy minimization. c) Flexible MAJP swarm display system. d) Magnetic actuation of MAJP swarms exhibiting two color states under opposed magnetic field. e) High contrast between the two swarm states.

(moving and rotating charged pigment particles in suspension with electric or magnetic fields),<sup>[18–24]</sup> electrowetting (controlling the wetting of dyed solvents under an electric field),<sup>[25,26]</sup> photochromic (colloidal phase segregation upon illumination),<sup>[27]</sup> and mechanochromic mechanisms (based on structural coloration and deformation of photonic crystals),<sup>[28–30]</sup> which have been driven by the development of new responsive materials, methods of actuation, and engineered functional particles. However, despite recent advances, most non-emissive displays have limited functionality and exhibit simple synchronous behavior with global on/off transitions, are limited by the arrangement of electrodes with fixed boundaries, and overall lack flexibility, reconfigurability, and complexity in the displayed information.<sup>[14,15]</sup>

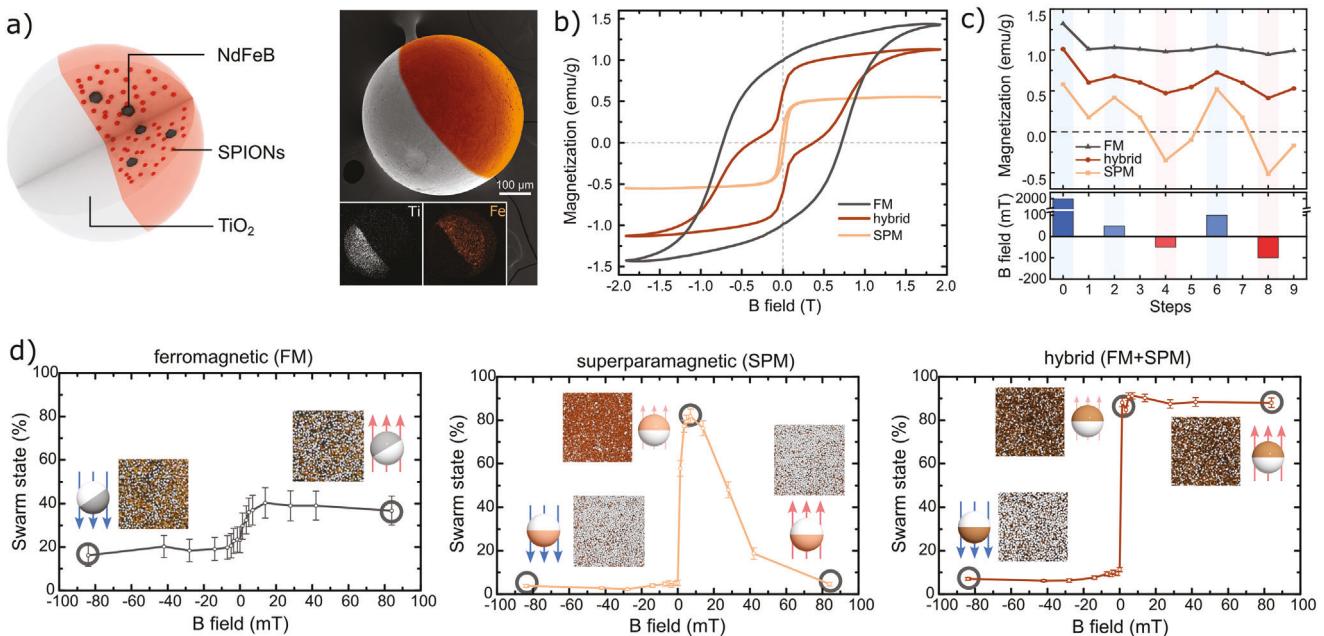
Here, we report a multifunctional swarm metamaterial system based on magnetoactive Janus particles (MAJPs) inspired by the dynamic skin coloration of cephalopods. The skin of cephalopods has chromatophores in its top layer, which are organs that contain sacs of pigment that rapidly expand and contract upon muscle actuation.<sup>[31]</sup> The collective actuation of chromatophores distributed throughout the skin enables adaptive coloration and pattern displays associated with many behavioral functions including camouflage, predation, and mating.<sup>[32]</sup> Inspired by the contraction and expansion of chromatophores and their emergent collective functions, we have designed MAJPs that can dynamically switch between predefined color states. MAJPs were designed and fabricated by electrohydrodynamic (EHD) co-jetting with engineered functional compartments for high contrast and addressable magnetic actuation.<sup>[21,33–35]</sup> The

programmable properties of MAJP swarms simultaneously enable fast response, remote actuation, and memory functions in electronics-free configurations, which massively facilitate integration in soft and flexible devices.<sup>[36,37]</sup> These modular configurations impart programmable magnetoactive behaviors and switching mechanisms in multifunctional active swarms, which enable advanced dynamic display, computation, non-volatile and semi-volatile memory, and encryption functions compatible with a soft, wearable devices.

## 2. Results

### 2.1. Design and Fabrication of Particle-Based Display System

The key active component in the proposed display system is the MAJPs, that is, particles with two functionally distinct compartments, each comprised of a precisely tailored composition with distinct magnetic and optical properties. These functional particles were fabricated by electrohydrodynamic (EHD) co-jetting, which produced polylactic glycolic acid (PLGA) bicompartimental fibers that were later transformed into bicompartimental MAJPs. Briefly, precursor solutions were prepared by dissolving and dispersing PLGA and particulate dopants in a mixture of solvents, which were then electrojetted using co-aligned needles under high voltage (Figure 1a; Figure S1, Supporting Information). During the EHD co-jetting process, charged liquid droplets are pulled from the needles by a high electric field, forming thin, long fibers that are rapidly solidified due to fast solvent



**Figure 2.** Properties and collective behavior of MAJPs. a) MAJPs are composed of magnetic particles (SPIONs and/or NdFeB) in one compartment, and white pigment ( $\text{TiO}_2$ ) in the other. SEM and EDS images show Ti and Fe in each compartment. b) Magnetic hysteresis loop of MAJPs with ferromagnetic NdFeB particles (FM), superparamagnetic iron oxide nanoparticles (SPIONs) (SPM), and hybrid (NdFeB and SPIONs). c) Magnetic remanence of MAJPs as a function of varying actuation fields, exhibiting different magnetization stabilities. d) States (brown color percentage) of different MAJP swarms as a function of actuation magnetic fields.

evaporation. The resulting bicompartimental fibers had a constant diameter of  $300\text{ }\mu\text{m}$  and a clear boundary between the magnetic (brown) and the pigment (white) compartments (Figure S2, Supporting Information). The fibers were microsectioned into short cylinders (aspect ratio  $\approx 1$ ) using a conventional microtome and processed via ultrasonication in water. Under these conditions, the microcylinders underwent a shape-transformation process into spherical particles, a process that is driven by surface energy minimization (Figure 1b).<sup>[38]</sup> To fabricate particle-based display devices, a MAJP suspension was encapsulated between two poly(ethylene terephthalate) (PET) substrates and clear adhesive spacers, resulting in a flexible, transparent, and robust device casing (Figure 1c; Figure S3, Supporting Information). When exposed to a magnetic field, MAJPs exhibit a swarming behavior synchronously rotating with the external magnetic field direction (Figure 1d). The particle swarm exhibits emergent properties of a magnetomechanical metamaterial where magnetic stimuli are coupled to a mechanical response. Under an external magnetic field (magnetic stimulus), the particles experience a magnetic torque that depends on the actuation field (controllable), particle magnetization (tunable by particle design and fabrication), and the angle between them. As a result, the torque induces rotational motion (mechanical response) to align the particles with the field. By switching the magnetic field direction (normal to the display), two high-contrast color states can be observed, demonstrating fast and reversible two-state transitions in the swarm (Figure 1e; Movie S1, Supporting Information). The size of the MAJP particle was optimized for high-contrast switching between the two states (Figure S4, Supporting Information). While each individual particle has two possible color states (one compartment facing up or the other) in response to magnetic fields, the collec-

tive behavior of MAJPs exhibits visual emergent complexity in the form of swarming patterns with encoded information. In the following sections, we will engineer three MAJP types with distinct magnetic properties and switching behaviors under magnetic fields, and will design spatiotemporal structured fields to enable complex display, Boolean logic, memory, and encryption functions in programmable swarm metamaterials.

## 2.2. Design of Magnetoactive Janus Particles (MAJPs) and Swarming Behavior

Here, MAJPs are designed with two opposed compartments: a brown-colored compartment with predefined combinations of  $5\text{ }\mu\text{m}$  NdFeB ferromagnetic microparticles and  $50\text{ nm}$  superparamagnetic iron oxide nanoparticles (SPIONs), and a white-colored compartment with  $\text{TiO}_2$  pigment for contrast (Figure 2a). Scanning electron microscopy (SEM) and energy dispersion X-ray spectroscopy (EDS) confirmed a biphasic distribution of Ti and Fe, showing a clear boundary between compartments at the equator of the particle. NdFeB and SPION particles dispersed in the magnetic compartment are used to induce forces and torques on the MAJPs<sup>[37,39]</sup>; however, they each provide a distinct response to external magnetic fields (Figure 2b). Ferromagnetic MAJPs (FM, composed of NdFeB) have high saturation magnetization and high remanence (i.e., high magnetization is conserved when the field is removed). Superparamagnetic MAJPs (SPM, composed of SPIONs) have lower saturation magnetization and near-zero remanence (i.e., magnetization is not conserved when the field is removed). Hybrid MAJPs (composed of both NdFeB and SPIONs in the same compartment) have

combined properties, with high saturation magnetization but low-moderate remanence. These properties translate to distinct polarization behavior for each MAJP type (Figure 2c). After an initial pre-magnetization step, each MAJP type retained different levels of magnetization, and were then exposed to oscillating fields to characterize their switching behavior. While FM MAJPs preserved their magnetization, SPM MAJPs changed magnetization direction according to the applied field, oscillating around zero. Hybrid MAJPs exhibited a combined behavior, oscillating around its remanence but maintaining constant direction. Without an initial pre-magnetization step, all MAJP types oscillated around zero magnetization, and FM dopants contributed with weak magnetic moments compared to SPM (Figure S5, Supporting Information).

As a result of their distinct magnetic properties, each MAJP type exhibited characteristic collective switching behavior in particle swarms under varying magnetic fields (Figure 2d). FM MAJPs did not align synchronously and did not exhibit homogeneous collective color states in the swarm in either configuration (at negative and positive fields) due to their initial low magnetization. On the other hand, SPM MAJPs align synchronously under small fields, exhibiting homogeneous swarm color states with high contrast. However, their magnetization is reversed at larger fields ( $>20$  mT) due to their low remanence, thus switching their swarm state under the same field direction. Hybrid MAJPs (combining SPM and FM in the same compartment) can be aligned synchronously under small fields and then permanently magnetized to fix their polarization (Figure S6, Supporting Information). As a result, hybrid MAJPs exhibit homogeneous synchronous swarm alignment, high color contrast between swarm states, and high stability (swarm states are stable under the same field direction, regardless of magnitude). The composition of the hybrid magnetic compartment (NdFeB:SPION ratio) was optimized for stability and contrast (Figure S7, Supporting Information). Full-color hysteresis maps in a magnetic field sweep cycle of all three MAJP swarm systems (Figure S8, Supporting Information) confirmed the reversibility of the color-state transitions with their distinctive switching behaviors.

### 2.3. MAJP Swarms with Boolean Logic Functions

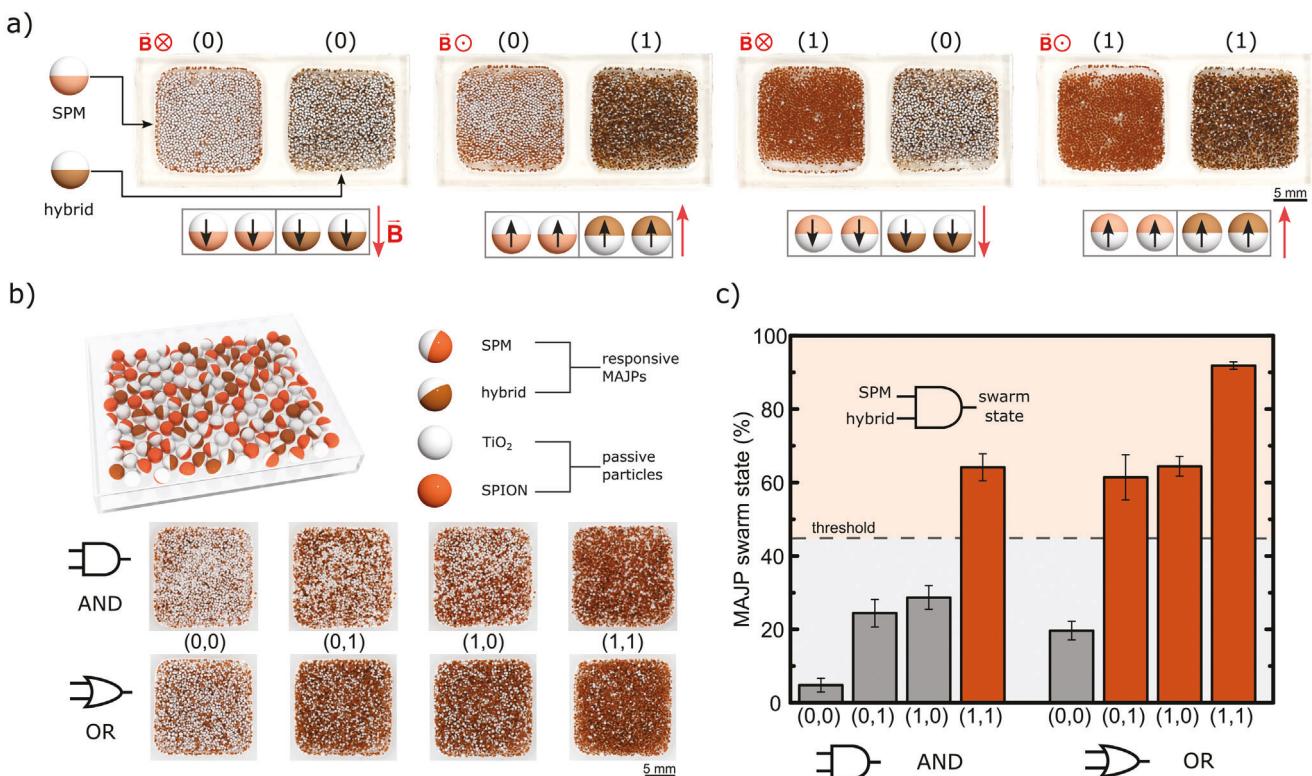
Leveraging their characteristic switching mechanisms, we have designed MAJP swarms with different responsive behaviors under the same input field. This approach enables programming multiple states in the swarms beyond the single binary states of individual particles. For example, we can define two states in a swarm as ON (1) and OFF (0) determined by the MAJPs orientation and collective displayed color (brown compartment up and white compartment up respectively). A swarm with a single type of MAJP can act as a binary 1-bit system that displays a collective color state as a function of magnetic field. By combining different types of MAJP swarms, we can program higher complexity responses in multi-bit display systems. To demonstrate this concept, we designed a 2-bit system combining SPM (left) and hybrid (right) swarms (Figure 3a) under the same magnetic field (both swarms subjected to the same global input field conditions at all times). Initially, both swarms are aligned together with the field (state 0,0), and can reversible alternate their state

by switching the field direction (state 1,1). However, the magnetization of SPM swarms can be rewritten at higher fields, while preserving the magnetization of hybrid swarms. This reversible transition enables access to the intermediate 0,1–1,0 states where SPM and hybrid swarms align synchronously but oppositely under the same homogeneous field (Figure S9; Movie S2, Supporting Information). Therefore, by controlling the global magnetic field direction and intensity, four interchangeable states can be accessed in this 2-bit, two-swarm MAJP system (which could be readily extended to higher number of bits for higher complexity). Hybrid particles can also be reprogrammed and their polarization reversed by applying an initial high-intensity pulse to give access to further programmable states and responses under the same field (Figure S10, Supporting Information).

In addition to programming swarms as individual bits, we can combine different types of MAJPs in a heterogeneous swarm configuration. This approach enables the design of MAJP complex swarms with programmable output states as a function of a single actuation stimulus, akin to other metamaterial structures developed for mechanical computing.<sup>[3]</sup> To demonstrate this concept, we have integrated responsive (SPM and hybrid) MAJPs and passive (solid TiO<sub>2</sub> and SPION) particles into complex swarms to provide a broad spectrum of programmable functions (Figure 3b). We designed swarm logic gates with AND and OR Boolean functions, with a programmable collective output state as a function of swarm composition and MAJP magnetoactive response (Figure 3c). These MAJP logic swarms exhibit the programmed behavior corresponding to their respective Boolean function truth tables. We discretized the output states of the complex logic swarms based on MAJP calibration (Figure S11, Supporting Information) and human color perception tests (Figure S12, Supporting Information), resulting in a consensus on/off switching threshold at 45%. This concept of functional responsive swarms can be easily miniaturized and integrated into multicell devices, and further expanded with dynamic and structured actuation fields for more advanced complex functions.

### 2.4. Display and Memory Functions of MAJP Swarms

MAJP swarms can display complex information by actuating selected particles only, exhibiting locally structured patterns rather than global on/off states. Actuation magnetic fields were spatially programmed by designing small-scale permanent magnet arrays arranged in specified orientations (Figure S13, Supporting Information), resulting in heterogeneously structured fields with programmed on and off regions. When applied to MAJP swarms, the particles align heterogeneously to the programmed structured field and display complex patterns in the swarm (Figure 4a). Since magnetic actuation does not require direct contact, the actuation arrays can be moved remotely, thus generating spatiotemporally heterogeneous fields and resulting in dynamic patterns with complex information (such as moving figures, rotating directions, sliding text, etc.) (Figure 4b; Figure S14 and Movie S3, Supporting Information). This selective switching actuation approach expands the display capabilities of MAJP swarms beyond 1-bit global behavior, both providing new design opportunities for emergent swarming patterns and enabling dynamic display functions. Furthermore, this information can be stored in the



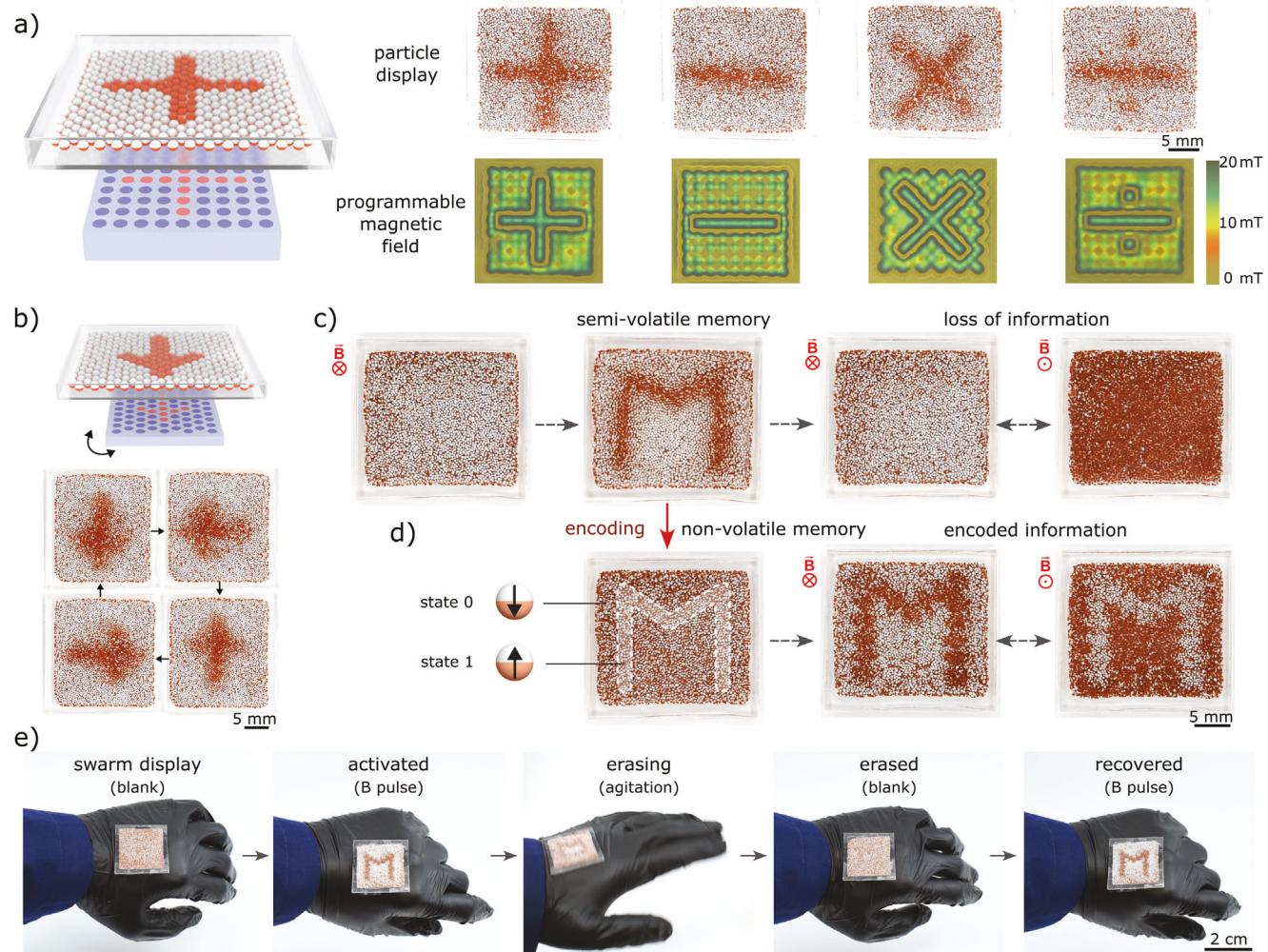
**Figure 3.** Selective actuation and switching behavior MAJP swarms with responsive logic functions. a) Selective switching behavior of MAJP swarms (SPM and hybrid) with 4 interchangeable 2-bit states by controlling the actuation magnetic field direction and intensity. b) MAJP swarms with AND and OR logic gate functions based on the individual input state of each particle type. c) Output state of AND and OR MAJP logic swarms.

magnetization states of MAJPs as a semi-volatile and non-volatile memory system. We have adapted conventional computer and mechanical memory terminology to describe our MAJPs memory functions<sup>[3]</sup>: (i) volatile memory requires power input (or in this case, an actuation field) to maintain the stored information (displayed pattern), but the information is lost when the field is removed. On the other hand, (ii) non-volatile memory does not lose information when the field is removed. In between of the two, (iii) semi-volatile memory has non-volatility but for a limited duration (the memory is degraded over time or disrupted by external agents). In semi-volatile memory mode (Figure 4c), a low-intensity structured field is applied which rotates and aligns the particles in a specific pattern. The particle orientation and displayed pattern remain after the field is removed, however, the information can be erased when a new field is applied. In non-volatile memory mode (Figure 4d), a high-intensity structured field is applied which rewrites the magnetization of particles in a specific pattern, encoding the information permanently. The displayed pattern is preserved after the field is removed and after new fields are applied. This non-volatile memory mechanism is further demonstrated in MAJP swarms integrated in a wearable soft display device mounted on a glove (Figure 4e; Movie S4, Supporting Information), where an “M” is encoded and displayed with an homogeneous field activation pulse. The pattern is not erased when the field is removed, but it can be erased with mechanical agitation (causing the particles to reorient randomly). However, since it has been encoded in the structured magnetiza-

tion of the swarm, the pattern is recovered and displayed again under another homogeneous field pulse without loss of information. Therefore, MAJP swarms and their integrated soft display devices offer highly-stable non-volatile memory without energy input and under mechanical perturbations or deformation, which enables their operation and deployment in wearable technology for unstable working environments.

## 2.5. Reconfigurable Encoding and Encryption of Complex Information

Taking advantage of the memory properties of MAJP swarms, we have designed an encryption mechanism to display information only under specific and programmable conditions. This mechanism is based on the XOR operation of superimposed non-volatile and semi-volatile memory functions on a SPM-MAJP swarm (Figure 5a). First, a pattern is encoded by applying a structured magnetic field (encoding key  $\vec{M}$ ), locally rewriting the magnetization of the swarm according to the predefined key (non-volatile writing). Then, a second structured field (reading key  $\vec{B}$ ) is applied, selectively actuating only those regions of the swarm where its encoded magnetization is misaligned (semi-volatile reading). As a result, we observe a complex displayed pattern due to the XOR operation of encoding and reading key fields  $\vec{M} \oplus \vec{B}$  in the swarm, generating a unique pattern that is not possible with either key field alone. This concept was adapted to the



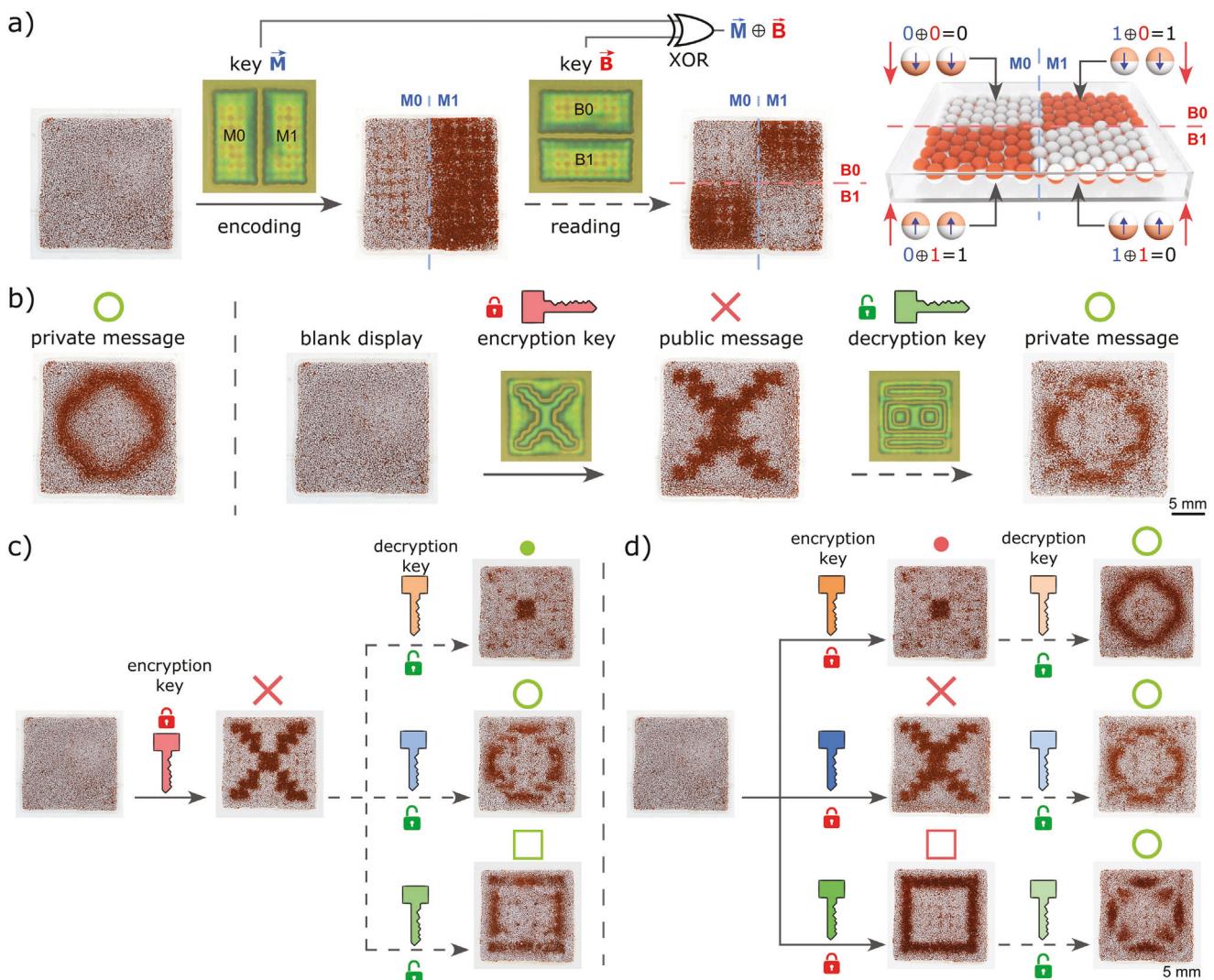
**Figure 4.** Swarming display and memory functions. a) MAJP swarm display with static patterns under preprogrammed structured magnetic fields. b) Dynamic MAJP swarm display patterns under structured rotating fields. c) Semi-volatile and d) non-volatile memory of structured display patterns, showing loss and conservation of encoded information respectively. e) Integration of a soft MAJP swarm display device on a glove, showing the recovery of encoded non-volatile information after mechanical agitation.

proposed encryption mechanism, where the structured encoding field has an encryption key function and the reading field has a decryption key function (Figure 5b; Figure S15, Supporting Information). For example, to communicate a target message of “○”, a MAJP swarm is encoded with an arbitrary encryption key field, displaying a public message of “×” when read with a homogeneous field without the proper key. Only when the unique complementary decryption key is applied the private message of “○” can be read. This swarm encryption system is versatile and scalable, and can be adapted to arbitrary key patterns to display different information under different conditions. For example, for a specific encrypted swarm displaying a public message of “×”, three different decryption keys were programmed to display private messages of “●”, “○”, “□” respectively (Figure 5c; Figure S16 and Movie S5, Supporting Information), thus displaying different messages from the same original encrypted message using. Oppositely, multiple encryption/decryption key pairings can be designed for a single message so that only the right combination

displays the target private message (Figure 5d; Figure S17 and Movie S6, Supporting Information). To illustrate this, three encryption keys displaying a public message of “●”, “×”, “□” were designed, with their respective complementary decryption paired keys to display a target private message of “○”.

### 3. Conclusion

The MAJP swarm systems described here in various device configurations have diverse programmable functions that arise from the rational design of a family of responsive bicompartimental Janus particles. By carefully designing the MAJP swarm configuration and its magnetoactive properties, one can tailor the responsiveness of MAJPs to magnetic fields based on varying magnetization, remanence, and stability, resulting in distinct and programmable switching behaviors. The design of unusual colloidal materials and their programmable behaviors opens a broad design space for complex display functions beyond global on/off



**Figure 5.** Encoding and encryption of information in MAJP swarm displays. a) Complex patterns are generated with a XOR operation of non-volatile magnetic encoding field and a semi-volatile reading field. b) Encryption is achieved by (i) encoding the particles with an encryption key field (non-volatile writing) and (ii) applying a complementary decryption key field (semi-volatile reading). c) Encryption using a single encryption key (single public message) and three different decryption keys reading different private messages. d) Encryption using multiple encryption keys (different public messages) and three different paired decryption keys reading the same private message.

switching, which constitutes a major breakthrough in the fields of reflective displays, responsive metamaterials, and soft robotic matter. More specifically, the design of MAJPs with tunable responsiveness under magnetic fields enabled non-volatile and semi-volatile memory mechanisms for the display of complex and dynamic graphic information in functional swarms (Table S1, Supporting Information). While the resolution of the graphics is intrinsically limited by the particle size and the structural resolution of the applied fields, these are easily scalable (both up and down) in size to fit particular message- and application-specific requirements. Due to their modular design (bottom-up hierarchical design of individual particles, their swarm switching properties, and swarm composition and structure to program the global response), MAJP display systems can also be scaled up to large swarms with wide working areas or miniaturized into smaller cells in multi-pixel metamaterial arrays, both pro-

viding a versatile platform for displaying more complex information. The integration of non-volatile and semi-volatile memory in heterogeneous particle swarms has enabled the design of logic gates and encryption functions for applications in mechanical computing and security (Table S2, Supporting Information).<sup>[3,40]</sup> Unlike other graphic encryption mechanisms that typically rely on single-key (null-to-image transitions),<sup>[8]</sup> the presented MAJP swarm system relies on a two-key XOR operator encryption mechanism, thus increasing the security and the specificity of the device (Table S3, Supporting Information). While most encryption systems relying on mechanical metamaterials are designed for a single specific message that cannot be modified once fabricated,<sup>[41]</sup> the MAJP swarm display system offers excellent versatility and reconfigurability as the keys can be immediately exchanged on demand for new messages. Furthermore, MAJP swarms are easy to deploy and integrate in flexible

devices as demonstrated, including wearables and other portable technology, for soft, flexible, durable, light-weight, electronics-free, and resilient (preserve function and encoded information under mechanical agitation and stress) performance. We note that MAJPs are not limited to configurations with two equally sized hemispheres, and complex particles with more than two compartments and/or different compartment sizes could provide access to a broad range of swarming behaviors and states that can be tailored toward particular functions. Because of their programmable switching properties and unique functions in computation, memory, and encryption functions, MAJP swarms are promising colloidal building blocks to develop the next generation of soft display devices, with growing applications in personal security, defense, anti-counterfeiting, camouflage, soft robotics, and human-robot interaction.

## 4. Experimental Section

**Magnoactive JANUS Particle (MAJP) Fabrication:** MAJPs were fabricated by first producing Janus microfibers from polylactic glycolic acid (PLGA) (ester termination, MW 50–75 KDa, Sigma Aldrich) solution through an electrohydrodynamic (EHD) co-jetting process. To prepare the solution, magnetic particles (<50 nm SPIONs and 5  $\mu\text{m}$  NdFeB microparticles) and 200 nm  $\text{TiO}_2$  particles were dispersed in a mixture of chloroform and N,N-dimethylformamide (DMF) (95:5 v v<sup>-1</sup>) using a Qsonica q700 ultrasonicator. PLGA was added to the dispersion at a 37.5% w/v concentration, maintaining a PLGA:particle ratio at 5.7% w/w. The PLGA/particle precursors were loaded in two separate 1 mL syringes with 25 G co-aligned needles. The precursor dispersions were then pulled from the needles by an 8 kV driving voltage (Gamma High Voltage Research) applied between the needle tip and the ground (15 cm distance) assisted by a syringe pump at a constant flow rate of 0.05 mL h<sup>-1</sup>. Co-jetted Janus microfibers were dried and stored in a vacuum desiccator for 2 days for solvent evaporation. The dried fibers were then embedded in optimal cutting temperature (OCT) cutting gel and microsectioned using a microtome (Epredia NX50) at 350  $\mu\text{m}$ . The sectioned microcylinders were suspended in OCT water solution (OCT served as surfactant and viscosity modifier), and were shape-transformed into spherical particles by surface energy minimization using ultrasonication (Qsonica q700 with 1/8" microtip probe for 15 min). The resulting MAJPs were washed with DI water to remove the OCT surfactant, and stored in DI water for later use.

**Scanning Electron Microscopy (SEM):** Janus particles were coated with a gold film (10–20 nm) in an SPI-Module Sputter Coater and imaged on the Thermo Fisher Nova 200 Nanolab SEM. The energy dispersive spectroscopy (EDS) function was used to image titanium and iron elements.

**Magnetic Characterization:** The magnetic hysteresis loops of MAJPs were measured at room temperature in a Lake Shore 7400 vibrating sample magnetometer. The particles were dried at room temperature in a vacuum desiccator and mounted on a sample holder. The measurements were made using a scanning field within the range of  $\pm 1.9$  T, and were background subtracted and normalized by the sample weight. The VSM was later utilized to test the magnetic stability of different MAJP types. After exposure to a high magnetic field (2 T), the magnetizations of the particles were measured at zero fields, and comparable fields within the actuation field range (50–100 mT).

**MAJP Swarm Cell Fabrication:** PET sheets with 0.15 mm thickness were laser-cut into the desired shapes (e.g., 3  $\times$  3 cm rectangles), and adhesive gaskets from mounting tapes were cut accordingly using a puncture tool. After attaching one side of the mounting tape gasket to one PET substrate, the MAJP suspension was transferred to the well and then sealed with another PET film on top (Figure S3, Supporting Information). Trapped air pockets were removed with two 18G 3" hypodermic needles (with one needle injecting water and the other suctioning air). Due to the elastomeric and self-adhesive properties of the mounting tape, the needle holes were sealed without leaking.

**Magnetic Actuation Using Homogeneous Fields:** MAJP swarms and devices were exposed to homogeneous magnetic fields (global direction) generated by either an electromagnet (Bunting BDE-4032-12, with a DC power supply Tekpower TP3005P) spanning  $\pm 84$  mT or permanent Nd-FeB magnets for larger fields. Actuation fields <20 mT were used to rotate the MAJPs by generating magnetic torques on the particles, and >50 mT were used to rewrite the polarity of SPM MAJPs. Alternating combinations of low (<20 mT) and high (>50 mT) magnetic fields were used to actuate SPM, FM, and hybrid MAJPs selectively in our swarm devices. A Nikon camera (D3500) was used to record the swarm states and transitions, and the images were later analyzed using Image (version: 1.53e) with the color threshold function to quantify brown versus orange pixels.

**Magnetic Actuation Using Structured Fields:** Structured magnetic fields (heterogeneous fields with varying intensity and direction over the actuation plane) were generated with arrays of small NdFeB permanent magnets ( $\phi$ : 2 mm, height: 1 mm, FINDMAG). Magnet array holders with 1 mm thickness with simple cubic or hexagonal close-packed 2 mm diameter holes and 1 mm spacings were 3D-printed (SL1S, Prusa research), and the disk magnets were then assembled in the holders with predefined directions and patterns, resulting in structured magnetic fields. The patterns of the structured fields were visualized using a magnetic flux display film (Learay Magnets).

**Design of Semi-Volatile and Non-Volatile Memory in MAJP Swarms:** MAJP swarms exhibited static semi-volatile patterns when a structured low-intensity field was applied (magnet array placed at >1 mm from swarm cell). Dynamic semi-volatile patterns were similarly exhibited under a mobile magnet array under the swarm cell. These patterns disappear after removal of the field and upon applying a new field. Non-volatile memory was achieved by selectively rewriting SPM MAJP swarms under high-intensity fields (>50 mT, achieved by positioning the magnet arrays at <1 mm under the swarm cell). The pattern was then displayed upon exposure to a new homogeneous field, remaining stable after the field is off and after mechanical agitation. These concepts were demonstrated on single swarm devices and on a wearable device mounted on a nitrile glove.

**Design of Boolean Functions in MAJP Swarms:** AND and OR logic gates were fabricated by combining responsive (SPM and hybrid) MAJPs and passive (solid color) particles into a single swarm. The two types of MAJPs were actuated independently, thus providing the two inputs of the gates, while the passive particles were added to tune the color baseline and the output response. To determine the transition threshold between the ON and OFF states, we performed a randomized color perception test with variations in MAJP swarm compositions with  $n = 46$  participants (HUM00236320, exempt from IRB review), resulting in a threshold of 45%. The MAJP swarm composition were then tuned accordingly: the AND swarm gate was composed of 40% SPM MAJP, 40% hybrid MAJP, and 20%  $\text{TiO}_2$  passive particles, and the OR swarm gate was composed of 40% SPM MAJP, 40% hybrid MAJP, and 20% SPION passive particles.

**Design of Encryption Mechanisms in MAJP Swarms:** The encryption process requires an encryption key and a decryption key, both generated with structured magnetic fields from arrays of permanent magnets with pre-programmed polarities. The encryption key is first applied at high-intensity, thus rewriting the magnetization of the swarm and encoding the desired pattern as semi-volatile memory. Under a homogeneous reading field (public message), the encryption pattern is displayed. The complementary decryption key field was applied at low intensity, performing an XOR function with the encryption key and displaying the private message only under the combination of the two keys.

## Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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## Conflict of Interest

The authors have submitted an invention disclosure related to this research.

## Author Contributions

Z.Z., J.L., and A.P.F. conceived, designed, and supervised the research. Z.Z. and J.R. synthesized the particles via EHD co-jetting. Z.Z. characterized the materials and fabricated the display cells. Z.Z. and A.P.F performed actuation, display, memory, and encryption experiments and analyzed the results. All authors participated in manuscript revisions, discussions, and interpretation of the data.

## Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

## Keywords

encryption, magnetoactive, mechanical computing, metamaterials, non-emissive display, particle swarm, reconfigurable structures, soft device

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- [1] K. Bertoldi, V. Vitelli, J. Christensen, M. van Hecke, *Nat. Rev. Mater.* **2017**, *2*, 17066.
- [2] P. Jiao, J. Mueller, J. R. Raney, X. Zheng, A. H. Alavi, *Nat. Commun.* **2023**, *14*, 6004.
- [3] H. Yasuda, P. R. Buskohl, A. Gillman, T. D. Murphey, S. Stepney, R. A. Vaia, Raney, *Nature* **2021**, *598*, 39.
- [4] T. Chen, M. Pauly, P. M. A. Reis, *Nature* **2021**, *589*, 386.
- [5] C. El Helou, P. R. Buskohl, C. E. Tabor, R. L. Harne, *Nat. Commun.* **2021**, *12*, 1633.
- [6] C. El Helou, B. Grossmann, C. E. Tabor, P. R. Buskohl, R. L. Harne, *Nature* **2022**, *608*, 699.
- [7] A. Pal, M. Sitti, *Proc. Natl. Acad. Sci. U.S.A.* **2023**, *120*, 2212489120.
- [8] M. Zhang, A. Pal, Z. Zheng, G. Gardi, E. Yildiz, M. Sitti, *Nat. Mater.* **2023**, *22*, 1243.

- [9] M. Zhang, H. Shahsavan, Y. Guo, A. Pena-Francesch, Y. Zhang, M. Sitti, *Adv. Mater.* **2021**, *33*, 2008605.
- [10] A. Rafsanjani, K. Bertoldi, A. R. Studart, *Sci. Rob.* **2019**, *4*.
- [11] K. Bhattacharya, R. D. James, *Science* **2005**, *307*, 53.
- [12] J. M. McCracken, B. R. Donovan, T. J. White, *Adv. Mater.* **2020**, *32*, 1906564.
- [13] X. Xia, C. M. Spadaccini, J. R. Greer, *Nat. Rev. Mater.* **2022**, *7*, 683.
- [14] D. Y. Kim, M.-J. Kim, G. Sung, J.-Y. Sun, *Nano Converg* **2019**, *6*, 21.
- [15] C. Gu, A.-B. Jia, Y.-M. Zhang, S. X. A. Zhang, *Chem. Rev.* **2022**, *122*, 14679.
- [16] Y. Kim, C. W. Moon, I. S. Kim, J. K. Hyun, *Chem. Commun.* **2022**, *58*, 12014.
- [17] C. Yu, Y. Li, X. Zhang, X. Huang, V. Malyarchuk, S. Wang, Y. Shi, L. Gao, Y. Su, Y. Zhang, H. Xu, R. T. Hanlon, Y. Huang, J. A. Rogers, *Proc. Natl. Acad. Sci. U.S.A.* **2014**, *111*, 12998.
- [18] R. Hattori, S. Yamada, Y. Masuda, N. Nihei, *J. Soc. Inf. Display* **2004**, *12*, 75.
- [19] Y. Komazaki, H. Hirama, T. Torii, *J. Appl. Phys.* **2015**, *117*, 154506.
- [20] T. Nisisako, T. Torii, T. Takahashi, Y. Takizawa, *Adv. Mater.* **2006**, *18*, 1152.
- [21] S. Hwang, T. D. Nguyen, S. Bhaskar, J. Yoon, M. Klaiber, K. J. Lee, S. C. Glotzer, J. Lahann, *Adv. Funct. Mater.* **2020**, *30*, 1907865.
- [22] S. Y. Lee, J. Choi, J. Jeong, J. H. Shin, S. Kim, *Adv. Mater.* **2017**, *29*, 1605450.
- [23] S.-H. Kim, S.-J. Jeon, W. C. Jeong, H. S. Park, S.-M. Yang, *Adv. Mater.* **2008**, *20*, 4129.
- [24] S. K. Nam, J. B. Kim, S. H. Han, S.-H. Kim, *ACS Nano* **2020**, *14*, 15714.
- [25] Y. Lu, B. Tang, G. Yang, Y. Guo, L. Liu, A. Henzen, *Micromachines* **2021**, *12*, 206.
- [26] J. Heikenfeld, K. Zhou, E. Kreit, B. Raj, S. Yang, B. Sun, A. Milarcik, L. Clapp, R. Schwartz, *Nat. Photonics* **2009**, *3*, 292.
- [27] J. Zheng, J. Chen, Y. Jin, Y. Wen, Y. Mu, C. Wu, Y. Wang, P. Tong, Z. Li, X. Hou, J. Tang, *Nature* **2023**, *617*, 499.
- [28] S.-U. Kim, Y.-J. Lee, J. Liu, D. S. Kim, H. Wang, S. Yang, *Nat. Mater.* **2022**, *21*, 41.
- [29] W. Wang, Y. Zhou, L. Yang, X. Yang, Y. Yao, Y. Meng, B. Tang, *Adv. Funct. Mater.* **2022**, *32*, 2204744.
- [30] M. Liu, J. Fu, S. Yang, Y. Wang, L. Jin, S. H. Nah, Y. Gao, Y. Ning, C. B. Murray, S. Yang, *Adv. Mater.* **2023**, *35*, 2207985.
- [31] R. Hanlon, *Curr. Biol.* **2007**, *17*, R400.
- [32] L. M. Mäthger, E. J. Denton, N. J. Marshall, R. T. Hanlon, *J. R. Soc. Interface* **2009**, *6*, S149.
- [33] K.-H. Roh, D. C. Martin, J. B. Lahann, *Nat. Mater.* **2005**, *4*, 759.
- [34] S. Bhaskar, K. M. Pollock, M. Yoshida, J. Lahann, *Small* **2010**, *6*, 404.
- [35] J. Yoon, K. J. Lee, J. Lahann, *J. Mater. Chem.* **2011**, *21*, 8502.
- [36] M. Li, A. Pal, A. Aghakhani, A. Pena-Francesch, M. Sitti, *Nat. Rev. Mater.* **2022**, *7*, 235.
- [37] Y. Kim, X. Zhao, *Chem. Rev.* **2022**, *122*, 5317.
- [38] K. J. Lee, J. Yoon, S. Rahmani, S. Hwang, S. Bhaskar, S. Mitragotri, J. Lahann, *Proc. Natl. Acad. Sci. U.S.A.* **2012**, *109*, 16057.
- [39] U. Bozuyuk, Y. Alapan, A. Aghakhani, M. Yunusa, M. Sitti, *Proc. Natl. Acad. Sci. U.S.A.* **2021**, *118*, 2022090118.
- [40] Y. Gao, S. F. Al-Sarawi, D. Abbott, *Nat. Electron.* **2020**, *3*, 81.
- [41] J. H. Kim, S. Jeon, J. H. In, S. Nam, H. M. Jin, K. H. Han, G. G. Yang, H. J. Choi, K. M. Kim, J. Shin, S.-W. Son, S. J. Kwon, B. H. Kim, S. O. Kim, *Nat. Electron.* **2022**, *5*, 433.