Nematic Colloidal Micro-Robots as Physically Intelligent Systems

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Physically intelligent micro-robotic systems exploit information embedded in micro-robots, their colloidal cargo, and their milieu to interact, assemble, and form functional structures. Nonlinear anisotropic fluids such as nematic liquid crystals (NLCs) provide untapped opportunities to embed interactions via their topological defects, complex elastic responses, and ability to dramatically restructure in dynamic settings. Here a four-armed ferromagnetic micro-robot is designed and fabricated to embed and dynamically reconfigure information in the nematic director field, generating a suite of physical interactions for cargo manipulation. The micro-robot shape and surface chemistry are designed to generate a nemato-elastic energy landscape in the domain that defines multiple modes of emergent, bottom-up interactions with passive colloids. Micro-robot rotation expands the ability to sculpt interactions; the energy landscape around a rotating micro-robot is dynamically reconfigured by complex far-from-equilibrium dynamics of the micro-robot’s companion topological defect. These defect dynamics allow transient information to be programmed into the domain and exploited. Robust micro-robotic manipulation strategies are demonstrated that exploit these diverse modes of nemato-elastic interaction to achieve cargo docking, transport, release, and assembly of complex reconfigurable structures at multi-stable sites. Such structures are of great interest to future developments of LC-based advanced optical device and micro-manufacturing in anisotropic environments.

1. Introduction

Untethered mobile micro-robots are the focus of intensive research with diverse strategies for actuation, mobility, and interaction.[1–6] Given their scale, the development of these systems has exploited and inspired research in far-from-equilibrium colloidal systems. Micro-robot mobility is achieved by various mechanisms including self-propulsion and actuation under external fields, intersecting with the field of active colloids.[7–11] The physical dimensions of micro-robots make it challenging to integrate computational elements that imbue them with computational intelligence. Thus, micro-robots typically exploit physical intelligence[12] to perform essential tasks including colloidal-scale cargo capture, transport, and delivery. Physical intelligence refers to diverse interactions between the micro-robot, its cargo, or domain boundaries that can be harnessed to perform useful work, often drawing on concepts at the forefront of directed colloid manipulation and assembly. For example, micro-robot motion is exploited to generate...
hydrodynamic interactions that dictate cargo displacement,[13,14] and external electromagnetic fields are applied to generate and control colloid-micro-robot interaction.[6–17] Such interactions can be tailored by design of micro-robot and cargo shape, material properties, and those of the domain boundaries.[17] While many studies in this arena are motivated by potential biomedical applications,[6,13,14] there are important untapped opportunities for micro-robotics in technologically relevant environments to generate reconfigurable structures for functional metamaterials ranging from advanced optical devices[18–21] to energy harvesting materials.[22,23]

While micro-robots are typically studied in isotropic fluids, highly anisotropic domains provide important additional degrees of freedom for designing the interactions between a micro-robot and its cargo. For example, curvature fields at fluid interfaces have been designed to direct colloid motion by capillarity[24]; such interactions have recently been exploited for micro-robotic assembly, and cargo manipulation.[25–27] Nematic liquid crystals (NLC) are anisotropic fluids in which physical information can be embedded via the organization of nematogens and the presence of topological defects to generate emergent interaction among microscale objects in the domain.[28–37] The introduction of micro-robot dynamics dramatically expands the opportunity to sculpt such interactions. For example, micro-robot shape and anchoring conditions can mold nematogen orientation and dictate the formation of topological defects; the ability to reposition the micro-robot allows this information to be embedded at arbitrary sites in the domain. Furthermore, in far-from-equilibrium systems, the energy landscape around micro-structures can be dynamically reconfigured to generate dynamic defect structures[37–46] by an interplay of the elasticity and external fields in these highly nonlinear fluids. Such reconfigurable energy landscapes provide exciting opportunities for exploitation in untethered micro-robotic systems.

In this paper, we design and fabricate a four-armed ferromagnetic micro-robot that can be actuated using an external magnetic field (Figure 1a). The micro-robot’s shape and surface chemistry are designed to embed a nemato-elastic energy landscape that generates complex force fields (Figure 1b) on passive colloids. These emergent interactions drive the colloids along
2. Results and Discussion

2.1. Micro-Robot Design and Its Static Defect Configurations

We fabricate a ferromagnetic four-armed micro-robot using standard lithographic methods followed by PVD sputtering of a layer of Ni (≈20 and ≈200 nm for thin and thick coating, respectively) and subsequent treatment with dimethyloctadecyl [3-(trimethoxysilyl)propyl] (DMOAP). Untreated SU-8 is known to have degenerate planar anchoring\(^\text{[47,48]}\) that we confirm for untreated SU-8 features under cross polarizers in our laboratory. DMOAP-treated Nickel surfaces are known to have perpendicular, homeotropic anchoring that we also confirm under cross polarizers (data not shown). Thus, the resulting micro-robot has homeotropic anchoring on its Ni-coated top and side surfaces, and degenerate planar anchoring on its bottom face that is untreated SU-8. When placed in a uniform planar cell filled with the nematic liquid crystal 4-cyano-4″-pentylbiphenyl (5CB) (Figure 1a), the micro-robot molds the local director field, a headless vector \(n\), which describes the orientational symmetry of the nematogens. The micro-robot’s arms and wells have curvatures designed to generate gentle distortions in the domain to promote lock-and-key assembly of passive colloids (Figure 1a,b). Given the micro-robot’s complex anchoring and sharp edges, its defect configurations differ significantly from their well-known counterparts on smooth spherical particles with uniform anchoring.

Once placed in the planar cell, two different defect structures emerge depending on the gap thickness between the two plates. For highly confined systems where the ratio of cell thickness to micro-robot thickness \(h/H\) is \(≈1.2\), the system assumes a metastable defect configuration with a quadrupolar symmetry, with two defects at the tip of the two arms aligned perpendicularly to the far field director (Figure S1, Supporting Information). For less confined systems (\(h/H\) = 2), a stable configuration emerges with dipolar symmetry, with a single defect visible at the tip of one of the arms aligned parallel to the far field director as shown in Figure 2a,b and Figure S2 (Supporting Information). Numerical simulation (Figures 1a and 2c) reveals that this defect is a disclination loop that is anchored on two locations on the micro-robot’s degenerate planar face and extends along the micro-robot’s side toward its homeotropic face. The stability and shape of the dipolar structure in simulations depend on the details of the side surface anchoring (Figure S3, Supporting Information). Simulation comparing the companion defect structure of an all homeotropic micro-robot and micro-robot with a hybrid anchoring condition shows similar defect configurations (Figure S4, Supporting Information). In experiments, the quadrupolar structure irreversibly transforms to the dipolar structure under external perturbation. In addition to these defects, simulation reveals zones of diminished order along with the sharp edges of the micro-robot.

To study these micro-robots in interaction with passive colloids under weak confinement, DMOAP-treated silica colloids (\(2a = 25 \mu m\)) with homeotropic anchoring are suspended with the micro-robot in SCB; this suspension is introduced into the planar cell in the isotropic state, and subsequently quenched into the nematic state by cooling below the isotropic-nematic transition temperature (\(T_{\text{IN}} = 35 ^\circ C\)). We focus on the weakly confined case in which both the micro-robot and the colloid carry dipolar defects. Under the action of an external magnetic field (details in Experimental Section), the micro-robot can translate or rotate with complex defect dynamics that are harnessed to interact with passive colloids.

2.2. Directed Assembly of Colloids in Nematic-Elastic Force Field

The micro-robot embeds a complex energy landscape in the surrounding NLC that generates emergent interactions that drive colloids along with distinct paths. The path followed by a particular colloid depends on the colloid’s polarity, its initial position, and the pose of the micro-robot. We enumerate the interactions that occur for a micro-robot at a fixed position with a defect on its left arm in interaction with colloids with either...
Five types of attractive interactions were observed: i) dipole-chaining, in which the colloid chains with the dipolar loop adjacent to the robot with its companion defect pointing outward, oriented along with the far field director (Figure 3a), ii) an antiparallel configuration, in which the colloid assembles with its defect oriented facing the defect of the micro-robot assemble into an anti-parallel structure (Figure 3b), iii) a dipole-on-hill configuration, in which the colloid docks on the curved tip of the robot arm with its companion defect pointing toward the robot (Figure 3c), and iv) a dipole-in-well configuration, in which the colloid assembles with its defect oriented to the defect of the micro-robot assemble into an anti-parallel structure (Figure 3d).
configuration, in which the colloid docks in a well between two arms of the micro-robot with its companion defect pointing outward, oriented along the far field director (Figure 3d), and, finally, v) a hybrid configuration where the colloid partially docks well with its companion defect tilted toward the nearest arm (Figure 3e). The dipole-chaining and antiparallel dipole configurations are reminiscent of dipole-dipole interactions of uniform colloids with homeotropic anchoring, although the details of the defect configurations on the micro-robot differ. Note that the anti-parallel dipole configuration relies on the micro-robot and colloid being in close proximity. The other three cases are a recapitulation of lock-and-key interactions in which colloids interact with gentle distortion fields seeded by the curved boundaries, as shown in the insets to Figure 3c–e.

Far from the micro-robot, the hedgehog companion defects on the colloids align with the far-field director with either rightward-facing or leftward-facing companion defects aligned with the far field director; this orientation defines the colloids’ polarity, as shown in the insets to Figure 3f(i), g(f), respectively. Colloid-micro-robot interactions depend on this polarization and the colloid’s initial position with respect to the micro-robot. The trajectories of all colloids that assembled on the micro-robot are superimposed in the micro-robot frame for colloids with rightward-facing defects in Figure 3f(i) and leftward facing defects in Figure 3g(i). In these figures, trajectories are color-coded in terms of their final mode of assembly. For example, the green curves represent the colloids’ trajectories as they approach the micro-robot and dock in the dipole-in-well configuration, while the blue curves represent the hybrid configuration in which the colloids are first repelled away from the micro-robot and then are attracted to the final equilibrium positions, resulting in complex, curved trajectories. All five assembly configurations and their trajectories are predicted by numerical analysis (Figure 3f(ii), g(f)).

We consider the free energy of point dipoles aligned along the local director field, \( F_{\text{dipole}} = 4\pi K p \cdot (V \cdot n) \), where \( K \) is the elastic constant and \( p \) is the dipolar orientation. Predicted colloid trajectories corresponding to the negative gradient in free energy are calculated for colloids with right-facing defects (Figure 3f(i)) and left-facing defects (Figure 3g(i)) in the far-field. Only the director gradients in the horizontal plane are considered in the analysis to avoid any influence of the director field ansatz on the sidewall of the micro-robot. These distinct modes of assembly depend strongly on initial relative positions, polarizations of the colloid, and differ in range and strength, allowing for selective directed assembly.

To better understand these interactions, we track trajectories and compare the normalized separation distance to their equilibrium position \( d_i/a \) as a function of time \( t_i - t_i \), where \( t_i \) is the time when the colloid reaches its equilibrium position as shown in insets to Figure 3h–l. The dipole-chaining and hybrid configurations have longer-ranged interaction, with attraction observed for distances as large as 6a from the colloids’ equilibrium positions, while the dipole-on-hill and dipole-in-well configurations have attractions up to separation distances of roughly 2a. Since colloids move with negligible inertia, i.e., with Reynolds number \( Re = \frac{Du}{\mu} \sim 10^{-7} - 10^{-6} \), where \( \rho \) is the density of 5CB, \( u \) is the speed of the colloid, and \( \mu \) is the average viscosity of 5CB, inertia can be neglected and the average viscosity of 5CB, inertia can be neglected and the energy of interaction \( U \) between a colloid and the micro-robot can be inferred from the energy dissipated along the colloids’ trajectories by hydrodynamic drag. Estimates for \( U \) (shown in Figure 3h–l) are calculated for each colloid-micro-robot configuration by integrating the viscous drag force along the trajectories of the colloids

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than those observed for the colloid with homeotropic anchoring and a dipolar companion defect. The magnitude of the attractive forces is also weaker than those between the micro-robot and a homeotropic colloid. These weaker interactions limit the speed at which a micro-robot can translate and retain colloidal cargo. For example, random planar colloids that assemble via their surface defect near the micro-robot's defect (Figure S7a, Supporting Information) can be retained at speeds of ≈ 0.28 µm s⁻¹, but are released at micro-robot velocities of ≈ 0.90 and ≈ 2.28 µm s⁻¹. For these latter cases, the drag on the colloidal particle overcomes the force of interaction. For comparison, the stronger interactions between the homeotropic particle and micro-robot in a dipole-chaining configuration retains colloidal cargo at translational speeds as high as ≈ 2.90 µm s⁻¹.

2.3. Far-From-Equilibrium Defect Dynamics

Under a rotating magnetic field, a far-from-equilibrium defect emerges whose dynamics are influenced by the micro-robot's hybrid anchoring conditions, complex shape, and sharp edges. Under slow rotation, the dipolar loop, situated at rest on a micro-robot arm aligned with the far field director, extends as it is placed in an antagonistic orientation with respect to the anchoring imposed on the planar bounding surfaces of the cell. At higher rotation rates, backflow becomes significant in addition to this geometric frustration, as the director field and flow field become coupled to leading order. Upon rotation by \( \frac{\pi}{2} \) radians, the defect becomes unstable and hops to the arm that has become aligned with the far field as shown in Figure 4a and Movie S2 (Supporting Information). As the defect hops, the dipolar structure interacts with a portion of the disclination loop beneath the micro-robot. When rotation ceases, the defect reverts to the dipolar configuration on the arm aligned with the director over time scales characterized by the relaxation dynamics of the system, given by \( \tau = \frac{L\gamma_1}{K} \), where \( L \) is the characteristic length of the micro-robot, \( \gamma_1 \) is the rotational viscosity of NLC. Under continuous slow rotation, the defect "travels" via periodic extension, interaction, hopping, and contraction in the opposite sense of the micro-robot's rotation. The ratio of the viscous to elastic forces is defined by a dimensionless Ericksen number \( Er = \frac{\gamma_1 vL}{K} = \omega \tau \), where \( \omega \) is the angular velocity of the rotating micro-robot. For example shown in Figure 4a, the Ericksen number \( Er \) equals 1.55. Similar behavior has been observed for \( Er \) as low as 0.06. Numerical simulations suggest distinct modes of defect hopping. Simulations of the rotating micro-robot with a dipolar loop (Figure 4b and Movie S3, Supporting Information) show dynamic elongation of the loop.

![Figure 4](https://www.advancedsciencenews.com)  
**Figure 4.** Defect hopping around the 4-armed micro-robot. The defect exhibits a hopping instability. a) Experimental time-series images of defect hopping from one arm (labeled as 1) to another arm (labeled as 2) of the 4-armed micro-robot during a π/2 clockwise rotation. \( \theta \) indicates the angle between the director (red arrow) and the diagonal (dashed yellow arrow) of the micro-robot. Scale bars are 20 µm. b) Time-series images of numerical simulation of the dipolar disclination loop defect hopping between the two arms of the 4-armed micro-robot (labeled as 1 and 2) during clockwise rotation. The red curves represent for the companion topological defect.
and sliding of the defect's pinning point on the micro-robot’s edge. This sliding mechanism allows the loop to move from one arm to another. Simulations of a micro-robot with a dipolar hedgehog companion defect (Figure S8 and Movie S3, Supporting Information) show another mode of hopping. In this case, the defect hops between neighboring arms of the micro-robot through the bulk phase due to strong dynamic alignment of the director field. Structurally, both pathways for defect hopping occur in regions of large director field deformations, and the motion of the defect reduces the elastic free energy of the system. Both experiment and simulation capture hopping of the defect between adjacent arms on the rotating micro-robot, as the system attempts to preserve the overall dipolar orientation with respect to the far-field. Under continuous rotation at angular velocity \( \omega \) that challenge the natural relaxation dynamics, the defects form smeared-out dynamic structures with periodic rearrangements that depend on the Ericksen number \( E_r = \omega t \). Under high \( E_r \approx 18 \) (Movie S4, Supporting Information), for example, the defect becomes significantly elongated and remains “smeared out” while hopping along the structure every \( \frac{\pi}{2} \) radians determined by the geometric symmetry of the four-armed micro-robot, lagging the alignment of the arm tips without relaxing back to the equilibrium structure. The extent of defect elongation is positively related to \( E_r \); larger \( E_r \) leads to greater defect elongation around the micro-robot. These defect dynamics play a very important role when interacting with colloidal cargo.

2.4. Cargo Juggling and Release

The emergence of far-from-equilibrium defects during micro-robot rotation is evidence of nonlinear restructuring of the director field and provides an important means to manipulate colloidal cargo. Figure 5a and Movie S5 (Supporting Information) shows a colloid assembled in the upper left well in the hybrid configuration at \( t = 0 \) s. As the micro-robot rotates, the assembled colloid moves around the micro-robot influenced by steric hindrance and hydrodynamics and encounters the micro-robot’s dynamic defect at \( t = 25 \) s. The elongated disclination loop of the micro-robot and the hedgehog defect from the colloid then merge to form a shared defect that carries the colloid along with it at \( t = 29 \) s. Upon further rotation, the shared defect separates to restore the colloid’s companion hedgehog defect and the micro-robot’s elongated defect (\( t = 35 \) s). These dynamics place the colloid in a repulsive configuration on the hill of the arm adjacent to its initial docking site, with its defect pointing outward; the colloid is repelled from the micro-robot (\( t = 47 \) s). Colloids assembled in other configurations can also be released by rotation via similar defect dynamics.

In another example, two identical colloids docked on the micro-robot can be juggled, rearranged, and restructured by far-from-equilibrium defect dynamics. The micro-robot with assembled colloids, shown in Figure 5b and Movie S6 (Supporting Information) at \( t = 0 \) s, is initially at rest, with colloid 1 (shown in red) docked in the bottom left well in the hybrid configuration and colloid 2 (shown in yellow) docked in the upper right well in the dipole-in-well configuration. As the micro-robot rotates in the clockwise direction, its defect lags behind the arm, elongates, and merges with colloid 1’s companion defect to form a shared structure (\( t = 19 \) s). This merged, shared defect carries colloid 1, rearranging its orientation and position on the micro-robot (\( t = 46 \) s). Thereafter, the merged defect further elongates along the sharp edges of the micro-robot and encounters the colloid’s companion hedgehog defect (\( t = 66 \) s), forming a merged defect that is now shared with both colloids. This larger loop also rotates and re-positions colloid 2. Finally, as shown in Figure 5b at \( t = 77 \) s, the merged defect becomes unstable and contracts; the micro-robot recovers its original defect structure, and the colloids’ companion hedgehog defects are restored. However, the changes in the positions and orientations of the colloids alter their ensuing interactions with the micro-robot. Colloid 1 eventually stably docks in the dipole-in-well configuration on the upper right. Colloid 2, however, placed in an antagonistic orientation at the tip of the arm with

Figure 5. Cargo juggling and release. a) Time-stamped images of cargo release of an assembled colloid between two arms of the micro-robot (labeled as 1 and 2) during clockwise rotation via dynamic defect interaction. The yellow arrows indicate the location of the (elongated) disclination loop of the micro-robot. b) Time-stamped images of cargo juggling of two colloids (labeled as 1 and 2) during clockwise rotation of the micro-robot via dynamic defect interaction. Scale bars are 20 \( \mu \)m.
its hedgehog defect pointing outward, moves away from the micro-robot. Thus, colloid 1 is retained and colloid 2 is repelled for $t = 77 \text{ s} - t = 135 \text{ s}$.

### 2.5. Micro-Robotic Directed Assembly of Colloidal Structures

Having demonstrated the ability to assemble, transport, and release passive cargo using our micro-robot, we further exploit the micro-robot for assembly of colloids and build structures by releasing these colloids near attractive sites on wavy walls.$^{[31,34,35]}$ For example, a colloid in a dipole chaining configuration ($\text{Figure 6a; Movie S7, Supporting Information}$) was carried as cargo by a micro-robot. This cargo was released near an attractive well on a wavy micro-structure via rotational defect dynamics like those described above, including defect elongation, merger, separation, and recovery. Once detached from the micro-robot, the colloid migrates into the attractive well ($\text{Figure 6a(iii)}$) and the micro-robot is driven away to retrieve a different colloidal building block in the domain. This process, which combines top-down direction by the micro-robot motion and bottom-up assembly via the emergent interactions between the colloid, the micro-robot and the wall, can be repeated and multi-element systems can be built. Depending on the design of the attractive sites, various colloidal structures can be constructed by the sequential addition of colloidal building blocks to prescribed sites. For example, using a bounding wavy wall as a construction site, as shown in $\text{Figure 6a}$, multiple structures were constructed including a 1D colloidal lattice ($\text{Figure 6b}$), a chain of seven colloids ($\text{Figure 6c}$) and a more complicated anisotropic structure ($\text{Figure 6d}$). Our approach is material independent as the nemato-elastic energy field is only dictated by the surface anchoring of the micro-robot and passive cargo that is defined by the initial surface treatment process and can be easily controlled. Thus, such micro-robotic assembly approach can be applied to functional building blocks made of differing materials for reconfigurable devices.

### 2.6. Trajectory Planning of Micro-Robot and Fully Autonomous Cargo Manipulation

The generation of strong magnetic field gradients on the micro- and smaller scales remains challenging and would hamper efforts to scale down this system to manipulate colloids of smaller radius. To address this issue, we exploit a defect-propelled swimming modality of nematic colloids using a purely rotating external field to actuate the micro-robot toward fully autonomous cargo manipulation.$^{[37]}$ Upon rotation, the companion defect of the micro-robot undergoes periodic rearrangement in which the defect depins from the micro-robot’s sharp edge and sweeps across the surface of the micro-robot; this occurs even as the defect hops between the micro-robots’ arms as shown in $\text{Figure 7a}$ and Movie S8 (Supporting Information). This defect sweeping motion acts as a swim stroke that drives micro-robot swimming; via this effect, micro-robot rotation generates unidirectional translation. For example, the micro-robot with its top arm initially positioned in contact with the dashed line in $\text{Figure 7a}$ traveled a distance of $30.3 \mu m$ in one period of rotation ($T = 160 \text{ s}$). Translational speed and direction are controlled by the rate and sense of rotation, as described in a detailed study of a rotating magnetic disk with similar hybrid anchoring in a separate study from our group.$^{[37]}$ We use this modality to actuate and control the micro-robot trajectories as shown in $\text{Figure 7b,c}$. Mirror symmetric changes in micro-robot trajectory with respect to the axis perpendicular to the far field director were achieved by reversing the micro-robot’s sense of rotation as shown in $\text{Figure 7b}$ and Movie S9 (Supporting Information). The direction of the external field of $T = 12 \text{ s}$ was reversed twice at the locations indicated by
the red dashed lines that led to an N-shaped trajectory of the micro-robot. More complex changes of direction are also possible. The direction of micro-robot translation depends on the rate of rotation, affording an additional degree of control. As shown in Figure 7c and Movie S9 (Supporting Information), by changing the period of rotation from 4 s to 8 s and finally to 16 s at the locations indicated by the red dashed lines, the micro-robot moves along a curved trajectory. This ability to steer the micro-robot relies on the rotation-rate dependent defect elongation that enhances broken symmetry in the system. With the ability to steer and make sharp turns while translating purely by tuning the rotation rate and direction, the micro-robot can be exploited for fully autonomous micro-robotic cargo manipulation.

We demonstrate fully autonomous micro-robotic cargo manipulation using our four-armed micro-robot under a programmable rotating magnetic field as shown in Figure 8 and Movie S10 (Supporting Information). The complete process was divided into four stages: i) locomotion and approach, ii) directed assembly, iii) transport, and iv) release. First, the micro-robot was driven toward a colloid, following an almost linear path of ≈ 137 μm with an average speed of 1.71 μm s⁻¹ under a clockwise rotating external field of T = 4 s (Figure 8(i)). Upon cessation of rotation, the micro-robot recovers its static dipolar defect and the colloid is attracted and migrates a distance ≈ 5.6α to dock in the dipole chaining configuration (Figure 8(ii)). Note that the time required for this docking process depends on the initial separation distance between micro-robot and colloid that determines the strength of the elastic interaction; this time can be greatly reduced if the micro-robot is placed closer to the colloid. Here, we parked the micro-robot relatively far from the colloid in order to demonstrate the range of this interaction. Once assembled, the pair was rotated counterclockwise under the same period of T = 4 s. During this process, the micro-robot followed a linear path while the colloid follows a helical trajectory and travels an effective distance ≈ 16.4α; the retention of the colloid is influenced by a complex interplay of defect-defect interaction and hydrodynamics (Figure 8(iii)). Finally, upon reducing the period of rotation to T = 20 s, the extent of defect elongation
is reduced, weakening the attractive interactions between the colloid and micro-robot’s defects. The colloid is released from the micro-robot (Figure 8(iv)), completing this fully autonomous cargo manipulation process.

2.7. Discussion of Open Issues

The long-ranged interactions between the particle and the microrobot rely on the ability of colloids suspended in NLCs to generate elastic distortions and companion defects. Theory indicates that this ability depends on the surface anchoring strength of the nematogens $W$ and the NLC’s elastic constant $K$. For DMOAP-treated silica surfaces in the 5CB, $W \approx 0.1 \text{ mJ m}^{-2}$. For 5CB, $K \approx 10^{-11} \text{ N}$ in the single constant approximation. Colloids are predicted to be unable to generate distortions and companion defects when their diameter $2R$ is approximately equal to the extrapolation length $\lambda = K W^{-1}$. For 5CB, the extrapolation length is $\approx 100 \text{ nm}$. Experiments in the literature indicate that DMOAP-treated silica nanoparticles in 5CB form defects and assemble via long ranged interactions with significant binding energies for diameters as small as $35 \text{ nm}$. These results may have implications in our study; they suggest that an appropriately scales micro-robot may interact with nanoscale colloids. Since free energy functional for NLC that captures the relevant energetics scales linearly with the size of the system, smaller systems will have proportionally smaller energies and ranges of interaction. However, there could be surprising outcomes associated with such changes in scale, as it is well-documented that microscale colloids and nanoscale colloids can have different metastable and stable states that may change the prevailing interactions.

The field of micro-robotics has spurred advancements in far-from-equilibrium soft matter colloidal physics. In this research, the highly nonlinear dynamic response of NLC revealed by the micro-robot’s motion has generated open fundamental questions that are worthy of detailed study. For example, the micro-robot has hybrid anchoring and rough sharp edges whose impact on dynamic defect pinning/depinning and defect elongation thresholds remain to be elucidated. The elongated defect undergoes multiple complex rearrangements including the swim stroke and defect hopping instabilities whose dependence on micro-robot properties and rotational dynamics warrant further study. In far-from-equilibrium micro-robot/cargo interactions, transient defect-defect interactions including defect sharing, merger, and separation play central roles in cargo fate. The settings in which defects remain distinct, merge, or re-separate in these highly nonlinear regimes are not known, and the physics that regulates these transitions remains unexplored. Pair interactions of micro-robots are also likely to be highly complex. Related studies on the simpler system of rotating disks with hybrid anchoring reveal complex pair interactions that depend strongly on rotation rate, defect-defect interactions, and degree of confinement. Greater fundamental understanding of such far-from-equilibrium behaviors would further develop NLC micro-robot physically intelligent interactions and would advance such system’s potential for tether-free micro-robotic cargo manipulation in technologically relevant settings. Furthermore, our results may spur research in active nematic systems such as microtubules whose nematogens consume chemical energy and dynamically reconfigure. The emergent behaviors harnessed in our system originate from the nematic fluid’s anisotropy; related effects should also emerge in active nematic systems, with the additional potential of harnessing the activity of these systems to drive micro-robotic motion and interaction.

3. Conclusion

We have introduced the concept of driven micro-robots in NLC as physically intelligent systems imbued with the capability to sense, attract and assemble colloidal building blocks via material agnostic nemato-elastic interactions and to dynamically restructure their environment. This untethered micro-robotic platform in NLC can generate complex colloidal reconfigurable structures via a combination of top-down and bottom-up assemblies. The motion of micro-robots in NLC is strongly coupled to the highly anisotropic nematic organization, and vice versa, providing opportunity to dramatically reconfigure the elastic energy landscape and to write transient director fields into the domain for potential micro-robotic applications. Here, we have described micro-robots with shapes and surface chemistry designed to embed elastic energy landscapes and generate distinct emergent interactions with colloidal cargo. Furthermore, the micro-robot’s rotational motion can...
deform its companion topological defect to generate rich non-equilibrium defect dynamics. We have exploited such dynamics as virtual functional structures that generate modalities of motion and interaction to enable reconfigurable assembly of passive building blocks with remarkable degrees of freedom. Finally, we have demonstrated a fully autonomous cycle of cargo manipulation using a swimming mobility enabled by the dynamic defect, which propels micro-robot translation. This ability to generate dynamic force fields, dynamically restructure the topological defects, and exploit them as functional structures for colloidal assembly greatly expands the opportunities for assembly of reconfigurable functional systems.

We envision applications ranging from functional metasurfaces and devices to manage electromagnetic, including thermal, fields. Our approach, which exploits the NLC's anisotropic response to generate micro-robot-colloidal cargo interactions differs from existing approaches for reconfigurable devices that exploit nematic liquid crystal's optical birefringence. Should our approaches gain traction, the opportunity for impact is vast, as society has made tremendous investment in the grooming of liquid crystalline responses, for example, in the multi-billion thin-film transistor liquid crystal display (TFT-LCD) industry.

4. Experimental Section

Fabrication of Micro-Robots and Assembly of Planar NLC Cell: Micro-robots with critical dimensions shown in Figure 1a were fabricated out of SU-8 photosist (Kayaku Advanced Materials, Inc.) following lithographic processes on a supporting wafer. Thereafter, a layer of nickel was sputtered onto the surface using a Lesker PVD75 DC/RF Sputterer to make the colloids ferromagnetic. Subsequently, treatment with 3 wt.% solution of N-dimethyl-n-octadecyl-3-aminopropyl-trimethoxysilil chloride (DMOAP, Sigma–Aldrich) imposed homeotropic anchoring condition on the micro-robot's Ni-coated surfaces. Untreated SU-8 was known to have degenerate planar anchoring. Upon release from the wafer and dispersion in SCB, the resulting micro-robot had hybrid anchoring. The top and side Ni-coated surfaces were homeotropic while the bottom bare SU-8 surface, which was protected during DMOAP treatment, had degenerate planar anchoring. Glass slides were spin-coated with polyimide (PI-2555, HD Microsystems) and rubbed with a velvet cloth along the desired direction to impose uniform planar anchoring. Two glass slides with uniform planar anchoring were assembled in an antiparallel fashion and glued together using a UV sensitive epoxy with two layers of 15 µm plastic spacers in between. The resulting thickness of the cell was ≈ 50 µm. Silica spherical colloids of 2a = 25 µm (Sphatech Inc.) were also treated with DMOAP, washed, and dried before adding into 4-cyano-4'-pentylbiphenyl (SCB, Kingston Chemicals). Finally, a mixture suspension of SCB with micro-robots and passive colloids was introduced into the cell from the side by capillarity in the isotropic state of SCB before quenching down to the nematic state. Depending on the thickness of the nickel layer, the coated micro-robot could either appear transparent (nickel layer = 20 nm) or black (nickel layer = 200 nm). While the transparent micro-robot allowed to visualize the sweeping motion of the disclination line, micro-robots with thicker coating possess stronger magnetic moments, enabling faster rates of rotation and translation under external magnetic fields.

Application of External Magnetic Fields: Controlled rotations of the micro-robots were achieved by placing the assembled NLC cell in a rotating magnetic field generated by a custom-built magnetic control system. The system consists of two orthogonal pairs of electromagnetic coils (APW Company) mounted on an aluminum supporting structure arranged around the workspace. Visual feedback was provided by a CCD camera (Point Grey Grasshopper3 Monochrome) mounted on a Zeiss inverted microscope (ZEISS Axio Vert.A1). Each coil pair was powered independently using a programmable power supply (XG 850 W, Sorensen) whose outputs were controlled by a Python algorithm written in-house. Sinusoidal time-dependent voltages were applied on each pair and the waveforms were separated by a π/2 phase lag to achieve a rotating field. The field gradient was applied by using rectangular NdFeB magnets (K&J Magnetics, Inc.) held to the end of a tweezer. The magnet was placed ≈0.5 cm from the cell. The amplitudes of the magnetic field applied are measured using a magnetometer and were in the order of a few mTs, far below the magnetic Fredericksz transition threshold to reorient the NLC molecules, but sufficiently strong to overcome the drag and move the micro-robot in arbitrary directions.

Details on Numerical Modeling: Numerical simulations of static and dynamic nematic structures were performed using a Q-tensor order parameter formulation of nematodynamics. The scalar degree of order S and the director n are the largest eigenvalue and the corresponding eigenvector of the Q-tensor, respectively. Equilibrium configurations correspond to minima of the Landau-de Gennes free energy with volume density of

\[
f_{\text{vol}} = \frac{1}{2} Q_{ij} Q_{ij} + \frac{B}{2} Q_{ij} Q_{ij} Q_{kl} Q_{kl} + \frac{C}{4} (Q_{ij} Q_{ij})^2 + \frac{L}{2} (Q_{ij} Q_{ij}) (Q_{ij} Q_{ij})
\]

where A, B, and C are phase parameters that dictate the degree of order in equilibrium homogeneous director field \( S_{eq} \) and \( L \) is the elastic constant. Additionally, Fournier-Galatola planar-degenerate surface potential descends the anchoring of nematic molecules on the bottom surface of the active micro-robot

\[
f_{\text{surf}} = W (Q_{ij} - Q_{ij}^p)^2
\]

where \( Q_{ij} = Q_{ij} + \frac{S_{eq}}{L} \delta_{ij}, Q_{ij}^p = (\delta_{ij} - \nu \cdot \nu_i Q_{ij} \delta_{ij} - \nu \cdot \nu_i), \) and \( \nu \) is the surface normal. Off the micro-robot's side wall, top surface and cell's top and bottom boundaries, the director field is fixed.

Equilibrium structures are found by using a gradient descent for the Q-tensor

\[
\vec{Q} = \vec{H} \nabla \vec{H}
\]

where \( \vec{H} \) is the molecular field \( \vec{H} = \frac{1}{2} \left( \frac{\partial F}{\partial Q_{ij}} + \frac{\partial F}{\partial Q_{ij}} \right) + \frac{1}{2} \frac{\partial F}{\partial Q_{ij}} \delta_{ij} \) and \( \Gamma \) is the rotational viscosity parameter. On the planar degerenate surface, Q-tensor follows the dynamics of

\[
Q_{ij}^\text{surf} = \Gamma \left[ \frac{1}{2} (H_{ij}^\text{surf} + H_{ij}^\text{surf}) - \frac{3}{2} \frac{\partial F}{\partial Q_{ij}^\text{surf}} + \frac{1}{2} \frac{\partial F}{\partial Q_{ij}^\text{surf}} \right]
\]

where \( \Gamma \) is the surface rotational viscosity parameter, and \( H_{ij}^\text{surf} = -\frac{\partial F}{\partial Q_{ij}^\text{surf}} + \frac{1}{2} \frac{\partial F}{\partial Q_{ij}^\text{surf}} \) is the molecular field.

Simulations for the rotating micro-robot were solved in the rotating frame of the micro-robot, in which case the time derivative of the Q-tensor includes an additional term of \( Q_{ij} Q_{ij} - \Omega_{ij} Q_{ij} \), where \( \Omega_{ij} \) is the vorticity tensor of the rotating colloid corresponding to \( \vec{r} = 6 \).

Equation 3 was solved using a finite difference method on a 800 × 800 × 240 mesh. The dimensions of the micro-robot were \( H = 75 \Delta x, \quad r_1 = 45 \Delta x, \) and \( r_2 = 37.5 \Delta x \) in accordance to Figure 1a. Neumann boundary conditions are used in the lateral directions of the numerical simulation box. Mesh resolution is set to \( \Delta x = 1.5 \Delta x, \quad L(1 + BS_{eq}) = 9 \Delta x, \) where \( BS_{eq} \) is the nematic correlation length that sets the size of the defect cores. The following values of the model parameters are used: \( B/A = 12.3, \quad C/A = -10.1, \quad W = 0.5 L/\Delta x, \quad \Gamma_{\text{surf}} = \Gamma/\Delta x, \) and a timestep of 0.1(\Delta t)/{(\Gamma L)}

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.
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Conflict of Interest
The authors declare no conflict of interest.

Authors Contribution
T.Y., Y.L., F.S., and K.J.S. designed research. T.Y. and E.B.S. developed control cell; T.Y. and Q.X.Z. performed experiments and analyzed experimental data. M.R. and Ž.K. designed and conducted numerical simulations. T.Y. and Ž.K. contributed to figure preparation. All authors contributed to the writing of the manuscript and participated in discussions of the research.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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active soft materials, directed assembly, emergent interactions, nonlinear dynamics, topology

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