Research Article

# Controlled Movement of Complex Double Emulsions via Interfacially Confined Magnetic Nanoparticles

Cassandra A. Zentner,[‡] Alberto Concellón,[‡] and Timothy M. Swager*

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**ABSTRACT:** Controlled, dynamic movement of materials through noncontacting forces provides interesting opportunities in systems design. Confinement of magnetic nanoparticles to the interfaces of double emulsions introduces exceptional control of double emulsion movement. We report the selective magnetic functionalization of emulsions by the *in situ* selective reactions of amine-functionalized magnetic nanoparticles and oil-soluble aldehydes at only one of the double emulsion's interfaces. We demonstrate morphology-dependent macroscopic ferromagnetic behavior of emulsions induced by the interfacial confinement of the magnetic nanoparticles. The attraction and repulsion of the emulsions to applied magnetic fields results in controlled orientation changes and rotational movement. Furthermore, incorporation of liquid crystals into the double emulsions adds additional templating capabilities for precision assembly of magnetic nanoparticles, both along the interface and at point defects. Applying a magnetic field to liquid crystal complex emulsions can produce movement as well as reorganization of the director field in the droplets. The combination of interfacial chemistry and precise assembly of magnetic particles creates new systems with potentially useful field-responsive properties.
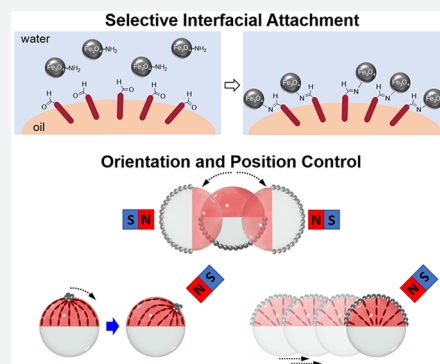


## INTRODUCTION

Particle stabilized emulsions, generally referred to as Pickering emulsions, have garnered attention for their high stability and biocompatibility when compared to traditional small molecule surfactant stabilized emulsions.[1,2] Particular attention has been paid to the fabrication of stimuli-responsive Pickering emulsions, which can be stabilized and destabilized with known triggers. Applied magnetic fields represent an attractive stimulus, as a result of ease of use and noninvasiveness considerations. Magnetic Pickering emulsions employing bare or functionalized $Fe_3O_4$ nanoparticles have shown utility in oil/water separation,[3,4] drug delivery/payload release,[5,6] and as templating materials.[7,8] Recent studies on the magnetic properties and specific control of magnetic Pickering emulsions have largely focused on the translational movement toward a magnet or the destabilization through dynamically switching magnetic fields.[3−6,9] Previous work by Liu et al. demonstrated the drastic change in magnetic properties when confining magnetic nanoparticles (MNPs) to a single emulsion's interface through interfacial electrostatic interactions.[10] Similarly, Ku et al. were able to control the magnetic properties of single Pickering emulsions through changes in interfacial MNP coverage.[11] Dispersed ferromagnetic nanoparticles, such as $Fe_3O_4$, exhibit superparamagnetic behavior in solution and lack the sufficiently large magnetic domains to prevent orientational relaxation of the aligned magnet moments after removal of external magnetic field. However, the interfacial confinement of MNPs can create extended magnetic domains and improved dipole−dipole interactions between MNPs to create a

ferromagnetic shell at the emulsion interface that by definition retains magnetization after removal of external magnetic field.[10−13] Studies of ferromagnetic layers in single emulsions open up exciting opportunities for the control of emulsions by magnetic fields.

We envisioned that further enhancements in controlled emulsion movement and orientation can be achieved by targeted placement and surface confinement of MNPs at double emulsion interfaces. Central to realizing new properties are the selective interfacial reactions needed to localize MNPs at only one of the specific interfaces of these more complex systems. In this work, we leverage previously demonstrated interfacial imine chemistry to selectively covalently attach amine-functionalized MNPs to one of the interfaces of double emulsions (Figure 1a).[14] In our design, water-dispersible amine-functionalized $Fe_3O_4$ magnetic nanoparticles (MNP-$NH_2$) react with surface active aldehydes, which can be present in hydrate form, to interfacially confine the MNPs (Figure 1b). Hydrate **1** and aldehyde **2** are selectively soluble in fluorocarbon (FC) and hydrocarbon (HC) oils, respectively, leading to specific, directed attachment of MNP-$NH_2$ at either
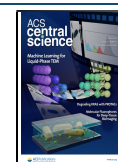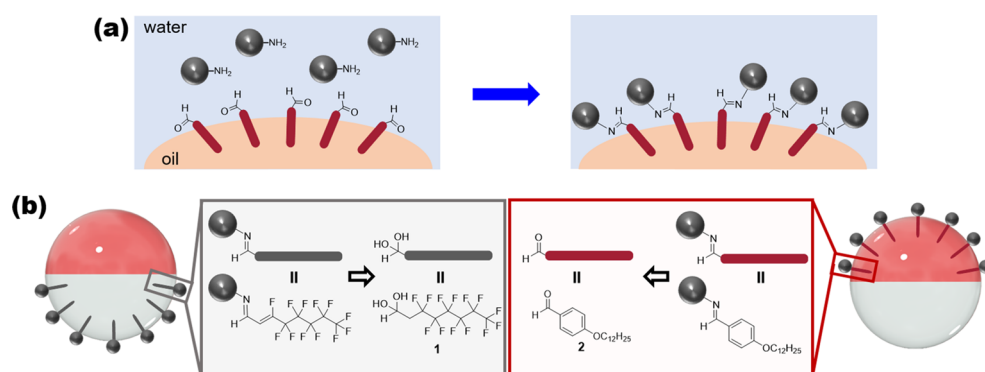
**Figure 1.** (a) Interfacial imine formation between amine-functionalized magnetic nanoparticles (MNP-NH$_2$) and oil-soluble aldehydes along the oil/water interface; (b) dynamic double emulsions decorated with magnetic nanoparticles and the retrosynthetic representation of the reaction of the MNP-NH$_2$ with the oil-soluble aldehyde or aldehyde hydrate.
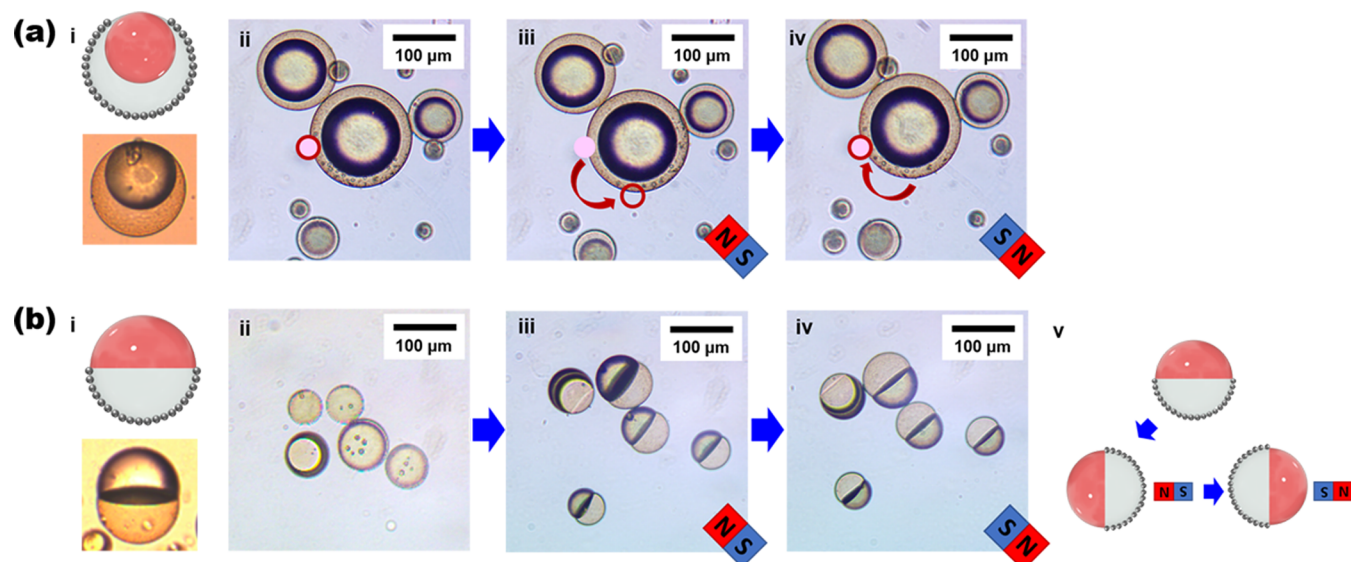


**Figure 2.** Magnetic response of MNP-NH$_2$ confined to the FC/W interface in HC/FC double emulsions with (a) HC/FC/W or (b) Janus morphology: (i) side-view schematic and image, (ii) top-view optical images before exposure to magnetic field, (iii) response to magnetic field, (iv) response to inversion of magnetic field, (v) schematic representation of Janus emulsion orientation change.

the FC or HC interface with water (W). This selective interfacial imine formation enables controlled manipulation of the orientation and directional movements of HC/FC double emulsions and induces ferromagnetic behavior. Double emulsions containing liquid crystals (LCs) provide for additional precision in MNP assembly. We have previously reported surfactant-driven control of the internal director fields and defects within LC double emulsions.[15] We now show that these features can be used to localize MNPs at particular points that can, in turn, strongly couple the LC organization to external magnetic fields.

## RESULTS AND DISCUSSION

Magnetic Fe$_3$O$_4$ nanoparticles (MNPs) were synthesized through the hydrolysis of FeCl$_3$ and FeSO$_4$, resulting in polydisperse nanoparticles. Amine functionalization was achieved by attachment of 2-aminoethyl phosphonic acid to the MNP surface, following a published procedure (full characterization in Supporting Information 3.3 and Figure S1).[16] The resulting nanoparticles are dispersible in water, with some observed aggregation of the nanoparticles, ranging from ~50−180 nm in size (Figure S1c,d). Magnetic measurements

of MNP and MNP-NH$_2$ solutions demonstrated minimal loss of magnetic saturation with functionalization (Figure S1e). Magnetic properties of nanoparticles strongly depend on their size and shape as well as their magnetic character.[12] Sufficiently small Fe$_3$O$_4$ nanoparticles (<30 nm) have both single domain structure and a flip in magnetization that can occur due to thermal effects (superparamagnetic effect),[17] and thus, our polydisperse 10.6 ± 2.6 nm sized MNP-NH$_2$ are superparamagnetic in solution; however, we also observed a small magnetic hysteresis suggesting the particles or aggregates partially assemble under applied field (Figure S1e). Studies on the MNP size effects on magnetic properties of emulsions were prohibitive as a result of aggregation of MNPs in water during and following functionalization, regardless of starting poly- or monodispersity. For this reason, bulk synthesized, polydisperse MNPs were used in our studies.

We initially studied the confinement of MNP-NH$_2$ to the interfaces of double emulsions comprising diethylbenzene and 2-trifluoromethyl-3-ethoxyperfluorohexane (HFE-7500) enriched with hydrate **1**. Polydisperse emulsions were fabricated by phase separation bulk emulsification with *in situ* imine formation,[14,18] with the expectation that MNP-NH$_2$ would

attach to the FC/W interface during emulsification, leading to HC-in-FC-in-water double emulsions (HC/FC/W). Emulsification attempts in the absence of cosurfactants resulted in selective MNP-NH$_2$ attachment to the FC/W interface; however, incomplete phase separation upon cooling was observed, potentially as a result of particles being internalized and thereby inhibiting clean phase separation (Figure S2). This effect is analogous to Pickering particles preventing coalescence in emulsions.[19] To counteract this phenomenon, cosurfactants were introduced in the aqueous phase during emulsification (Supporting Information 4.2). Low concentrations (0.01 wt %) of FC-surfactant Zonyl FS-300 (Zonyl) helped to promote full phase separation but did not inhibit *in situ* imine formation, allowing for successful MNP-NH$_2$ attachment (Figure 2a). The anticipated HC/FC/W emulsions were obtained with uniform coverage of MNP-NH$_2$ at the FC/W interface. The droplet morphology is not fully encapsulated, with some HC/W interfacial area present; however, no MNP-NH$_2$ particles were attached to the HC/W interface. Additionally, emulsions were stable to coalescence for greater than 3 months. In control experiments, immediate coalescence of the droplets or no nanoparticle attachment were observed in the absence of hydrate **1**, confirming imine formation is necessary for stabilization of the emulsion and selective interfacial functionalization (Figure S4). These control studies also demonstrate that minimal unreacted MNP-NH$_2$ is present at the interface, signifying that any observed magnetic response is as a result of interfacial confinement due to imine formation.

To elucidate if the interface confined MNP-NH$_2$ in HC/FC/W double emulsions have ferromagnetic behavior, we observed their responses to an external magnetic field. Upon exposure to a magnet, instant rotational motions in space were observed to maximize the magnetic alignment of the functionalized surface with the applied field (Figure 2a-ii to iii, Video S1). Upon inversion of the magnetic field (N−S pole flip), the double emulsions rotated back in the opposite direction (Figure 2a-iii to iv, Video S1). In addtion to the rotation observed, smaller droplets also partially tilt toward and away from the magnet with changes in magentic field (Video S1). Further, the emulsions rotated in space with movement of the magnet (N-pole) around the sample (Video S2). The rotational movements induced by the attraction to and repulsion away from the magnetic field are consistent with ferromagnetic behavior, and emulsions are exhibiting N−S pole orientation that remains even after the removal of magnetic field. If the droplets had a static ferromagnetic ordering, 180° rotation with the N−S pole flip is expected. However, the extent of rotation varied across the sample and 180° rotation was not always observed. The emulsion droplets are polydisperse; therefore, exact droplet size impact on response was not obtained, but in general, smaller droplets displayed a greater response/rotation to magnetic field. Our results suggest that interfacial confinement increased the magnetic domains size and promoted the cooperative alignment of magnetic moments leading to the observed rotational movements; however over the experimental time frame, relaxation of the MNP organization/magnetic dipoles can result in some randomization of magnetic moments and prevented full 180° rotation in all droplets. We reason a fully "locked-in" state of the MNPs at the interface of HC/FC/W emulsions was not achieved, resulting in some freedom of movement. We also note that other factors, including increased viscous resistance in larger droplets, could prevent full rotation.

We next explored the behavior of Janus emulsions with interfacially confined MNP-NH$_2$. To achieve Janus emulsions with magnetic functionality at the FC/W interface, HC-surfactant Tween-20 was added after the previously described *in situ* emulsification of magnetic HC/FC/W double emulsions. Significantly high concentrations of Tween-20 (0.5−1 wt %) were necessary to induce this morphology change. This is attributed to the jammed state of the confined MNP-NH$_2$ particles, which prevents quick morphology changes characteristic of surfactant stabilized double emulsions.[11] The resistance to reconfiguration can be attributed to the Pickering emulsion and may also be enhanced by magnetic ordering of the MNPs at the FC/W interface. The jammed state further resulted in "snowman" morphologies, wherein the area of FC/W interface is enhanced in the Janus state, which is a consequence of strong interactions of the MNPs with both the fluorocarbon and water (Figure 2b-i).

The MNP-NH$_2$ Janus emulsions instantly responded to a magnetic field, overcoming the preferred gravity aligned orientation by flipping onto their side (Figure 2b-ii to iii). Upon inversion of the magnetic field, the emulsions flipped 180°, exhibiting repulsion away from the magnetic field (Figure 2b-iii to iv, Video S3). Again, the emulsions are demonstrating N−S pole orientation owing to the ferromagnetic MNP layer, resulting in attraction toward and repulsion away from the magnetic field. The observed 180° inversions signal to an increase in dipole−dipole interactions and cooperatively as a result of a more locked-in state of the MNPs at the interface, and a reduction in competing relaxation allowing for retention of dipole alignment over the measurement time frame. In addition to an increase in jamming, the transistion from HC/FC/W morphology to a Janus state increases the asymmetry of MNP-NH$_2$ surface coverage on the emulsions. The intrinsic polar nature of asymmetric, Janus structures enhances ferromagnetic properties in materials.[20,21] Consequently, structural control of double emulsion morphology provides control over magnetic behavior. Ferromagnetic behavior is further demonstrated by the horizontal rotation of the Janus droplets as a magnet (N-pole) is moved around the samples (Video S4).

To confirm the interfacial confinement is necessary for observed responses to magnetic fields, we synthesized FC-dispersible magnetic nanoparticles (MNP-R$_F$, Figure S5). Janus emulsions with MNP-R$_F$ dispersed within the FC phase (1 mg/mL) exhibited translational movement toward the magnet without changes in droplet orientation (Figure S6). Inversion of magnetic field did not change the behavior of the emulsion movement or orientation. In total, all of the observations confirm that the interfacial confinement of MNPs imparts ferromagnetic behavior to our double and Janus emulsions.

To further study the confined state of the nanoparticles at the interface, we investigated the magnetization curve (magnetic field versus magnetic moment) of MNP-NH$_2$ dispersed in water in comparison to when they are attached to the interfaces of double emulsions (Figure 3a). Measurements of the magnetic properties of the emulsions were performed in a continuous phase without unreacted MNP-NH$_2$, ensuring the magnetic results were from confined MNP-NH$_2$ only. Confinement at the interface of HC/FC/W emulsions provides an increase in the magnetic saturation from 58.06 to 72.60 emu/g (Figure 3b). An additional increase in magnetic saturation to 77.25 emu/g was observed with
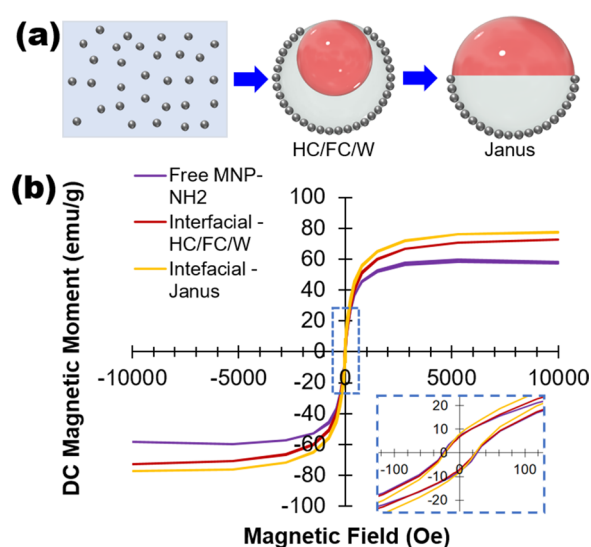
**Figure 3.** Magnetization curve of free MNP-NH$_2$ as well as confined MNP-NH$_2$ in HC/FC/W and Janus double emulsions. (a) Schematic of materials studied; (b) magnetization curve, inset − hysteresis.

jamming of the MNP-NH$_2$ caused by the morphology change to the Janus state. These results confirm an increase in ferromagnetic character resulting from improved magnetic dipole alignment capability with confinement and densified packing of MNP-NH$_2$ at the emulsion interface and increased asymmetry. Our results are consistent with previous studies that have shown an increase in magnetization with confinement of magnetic nanoparticles in assemblies.[22,23]

Generally, ferromagnetic materials exhibit hysteresis in the magnetization curve as a result of the retention of magnetization. Previous studies have shown both increases and decreases of the magnetic coercivity as a result of the confinement of magnetic nanoparticles.[22−24] However, our emulsions do not exhibit measurable enhancements in coercivity (Figure 3b-inset). Liu et al. observed a large increase

in coercivity with full interfacial MNP coverage in their single emulsion system.[10] It is likely that we do not achieve a fully jammed interface needed to observe a similar increased coercivity. The polydisersity of the individual MNP-NH$_2$ particles and the aggregate sizes formed in water (Figure S1c) could lead to random assembly of the particles at the interface and less than ideal packing capabilities. Therefore, unlike in the assembly of monodisperse Fe$_3$O$_4$, a close-pack monolayer is not readily achieved. We reason this imperfect coverage could weaken the interaction between MNPs along the interface, resulting in lower coercivity. However, in our system, the weak ferromagnetic coupling manifests in an increase in magnetic saturation and significant macroscopic ferromagnetic control of the emulsions. Simple changes in symmetry are sufficient to induce dramatic increases in macroscopic ferromagnetic responses to magnetic field. Consequently, the emulsions are capable of amplifying small changes in coupling, resulting in a highly sensitive system.

Following successful generation of ferromagnetic behavior in HC/FC double emulsions, we applied the same strategy to double emulsions containing liquid crystals (LCs) to generate programmable MNP packing at interfaces, leading to a distinct magnetic response. LCs have long been used to organize (often align) solutes, and point defects in the director fields can be used to consolidate particles.[25−27] The latter highlights how topological defects present within a LC emulsion can precisely position polymers, biomolecules, or micrometer-sized particles.[28−30] As a result, LC templating provides a powerful approach for precisely controlling the interfacial organization/positioning of MNPs. Previous magnetic LC emulsions consisted of single emulsions, in which MNPs are dispersed within nematic phases, and the results were similar to what is observed with particles dispersed in one of the phases of an isotropic liquid. Specifically, this functionalization enables translational movements toward the magnet.[31] Alternatively, MNPs dispersed in cholesteric single emulsions showed a programmable assembly in the internal disclinations of the cholesteric phase.[32] Herein, we make use of Janus emulsions
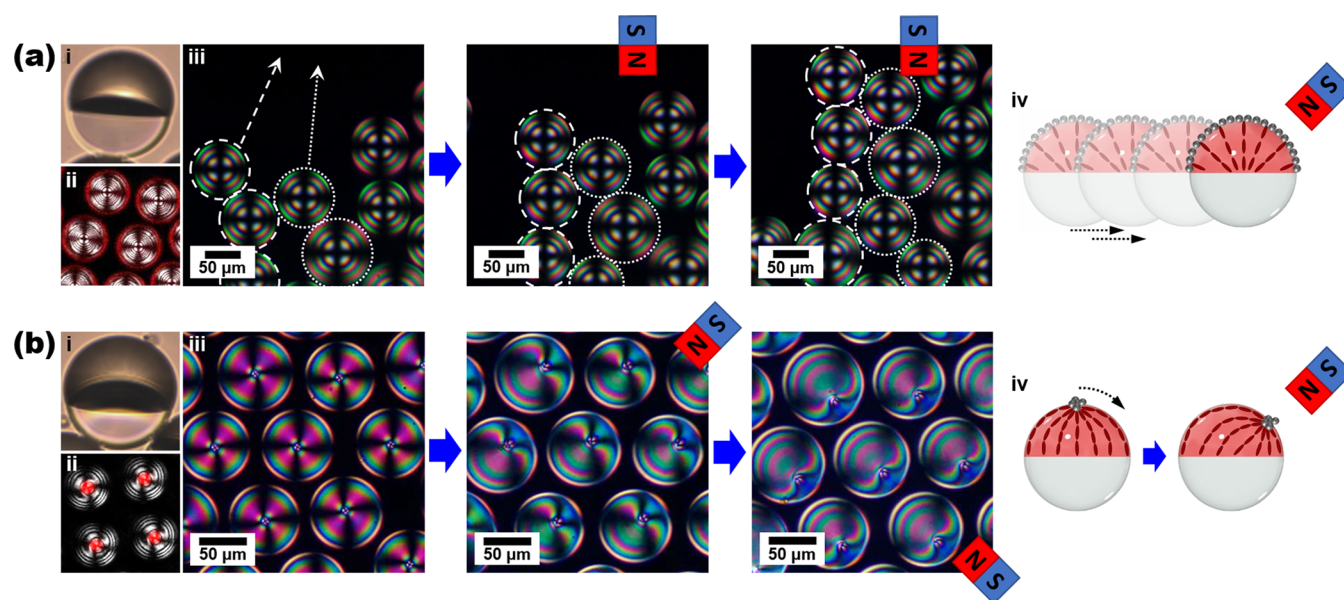


**Figure 4.** Magnetic response of MNP-NH$_2$-functionalized LC double emulsions with (a) radial and (b) monopolar organizations: (i) side-view microscopy images, (ii) confocal microscopy images, (iii) polarized-light optical microscopy images, (iv) schematic representations.
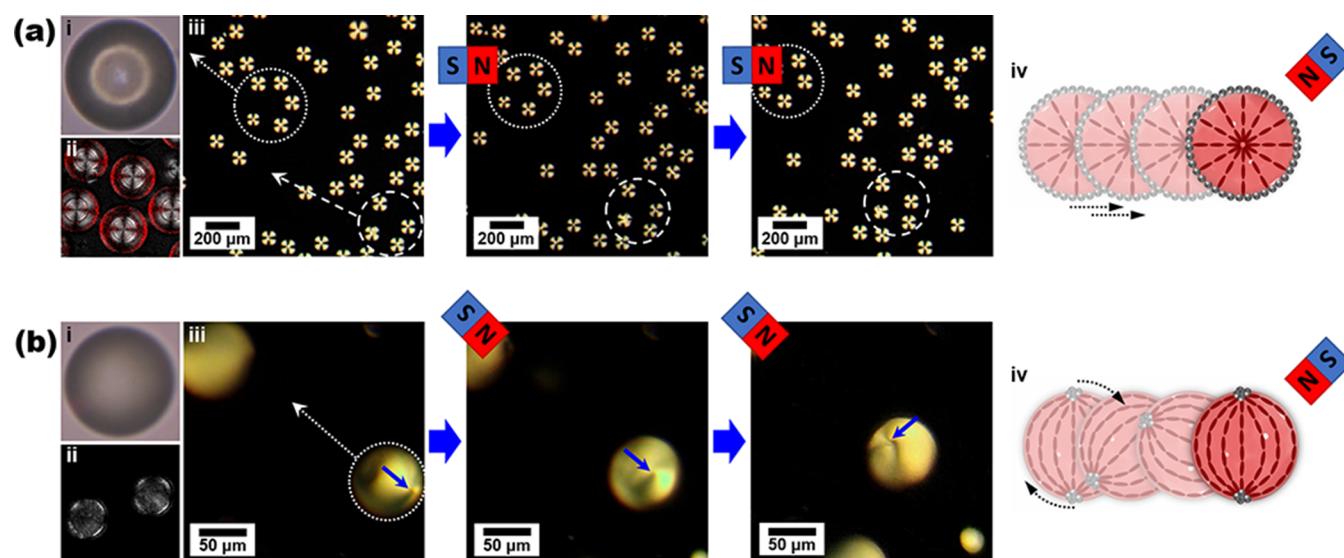
**Figure 5.** Magnetic response of MNP-NH$_2$-functionalized LC single droplets with (a) radial and (b) bipolar organizations: (i) side-view microscopy images, (ii) confocal microscopy images, (iii) polarized-light optical microscopy images wherein the point singularity containing the MNP-NH$_2$ is highlighted by a blue arrow, (iv) schematic representations.

comprising two immiscible phases, nematic LC (4-cyano-4′-pentylbiphenyl, 5CB) and FC oil (ethyl perfluorobutyl ether, HFE-7200), to organize MNPs. These Janus droplets were prepared and stabilized as reported previously by an evaporation-induced phase separation method that includes internal LC/FC surfactants (Figure S10).[15] Aldehyde **2**, which is exclusively soluble in 5CB (Figure S7), was added to the system to immobilize MNP-NH$_2$ at the LC/W interface via interfacial imine formation.

Janus droplets with different internal LC director organizations were used to study the different MNP-NH$_2$ arrangements. We first explored a radial configuration, in which 5CB molecules exhibit perpendicular (homeotropic) anchoring at the aqueous interface with no single point defect (singularity) formation at the LC/FC interface (Figure 4a). Polarized-light optical microscopy (POM) of these droplets showed the expected Maltese cross textures. The presence of aldehyde **2** did not affect the LC organization within the Janus droplets. In contrast to MNP-NH$_2$ coverage to the HC/FC droplets, which can be observed by side-view optical microscopy, the attachment of MNP-NH$_2$ to the LC/W interface could not be detected optically. The necessity for the Tween-20 cosurfactant and the lower propensity of aldehyde **2** to assemble at the interface relative to hydrate **1** both likely contribute to the lower MNP-NH$_2$ coverage.[14] The spatial organization of MNP-NH$_2$ at the LC/W interface is imaged by fluorescent labeling by reacting the MNP-NH$_2$ with *n*-succinimidyl ester rhodamine. Successful functionalization was confirmed by using a confocal microscope equipped with crossed polarizers. Merged fluorescent and polarized-light micrographs reveal bright fluorescence over the entire LC/W interface (the red fluorescent rings), confirming a MNP-NH$_2$ shell at the LC/W interface (Figure 4a-ii). In the control experiments under the same conditions without the MNP-NH$_2$ attachment step, no fluorescent ring was observed by confocal microscopy (Figure S8a), confirming the attachment of immobilized MNPs.

In the absence of an applied magnetic field, the MNP-NH$_2$-functionalized LC/FC Janus droplets randomly disperse in

water. Applied magnetic fields magnetize the droplets, and they undergo translational motion toward the magnet (Figure 4a-iii, Video S5). During movement, interemulsion attraction between the magnetic dipoles results in the formation of short-range linear chains. As individual droplets and droplet chains get closer in space to the magnet, the size of the chains gradually increased. However, we observed no noticeable change in the POM texture of the Janus radial droplets in response to magnetic fields, indicating that the field does not change the organization of the LC director field. When the magnetic field was removed, droplets stopped moving, chains lost their integrity, and droplets redisperse in water over the course of 4−5 h. LC/FC Janus emulsions did not exhibit the same ferromagnetic response (i.e., rotation) as was observed in HC/FC Janus emulsions. Lower overall coverage, and therefore a less jammed state, resulted in diminished response. Magnetization curves of LC emulsions could not be reliably measured, because the lower overall coverage of MNP-NH$_2$ resulted in a weaker magnetic response, and the mass of attached MNP-NH$_2$ was difficult to accurately obtain.

To guide our understanding of the influence of the LC organization on the interfacial assembly of MNP-NH$_2$, we investigated Janus droplets with a monopolar configuration. In this case, the singularity resides at the LC/W interface, and minimization of the elastic energy places it at the pole of the droplet. These droplets were obtained by using our previously reported method based on mesogenic surfactants that promote an alternative parallel (planar) LC anchoring at the LC/W interface (Figure S10).[15] Janus monopolar droplets concentrate the MNP-NH$_2$ assembly at the topological defect at the droplet's pole. Aldehyde **2**, which is not liquid crystalline, concentrates around the defect core to minimize the free energy and subsequently reacts with MNP-NH$_2$ at this location. POM images confirm that these Janus droplets exhibit a director profile that radiates from a point defect located at the pole, thereby confirming that the monopolar configuration was not altered after imine linking of the MNP-NH$_2$ (Figure 4b). Combined fluorescent and polarized-light micrographs reveal that the fluorescent signal (the red spots)

from rhodamine tagged interface-bound MNP-NH$_2$ coincide with the location of the topological defect of the monopolar LC Janus droplets (Figure 4b-ii). Moreover, heating these droplets into an isotropic phase ($T_{N-I}$ = 35 °C) produces a bright fluorescence over the entire LC/W interface (red fluorescence rings) (Figure S9a). All these observations confirm that MNP-NH$_2$ selectively localize at the LC topological defects. In the control experiments under the same conditions without MNP-NH$_2$ attachment step, no fluorescence was observed by confocal microscopy (Figure S8b).

Under the action of a magnetic field, the MNP-NH$_2$ labeled topological defect moves from the north pole to the side of the droplet adjacent to the magnet (Figure 4b, Video S6). Control experiments without crossed polarizers were carried out to ensure that we were not simply tilting the Janus droplets on their sides, which would generate the same POM texture (Figure S9b). These control experiments confirmed that under a magnetic field, the MNP-NH$_2$ labeled topological defect moves toward the magnet without an orientation change or physical translation of the Janus droplet. Once the magnetic field was removed, the topological defect relaxes to the initial state and location. After heating monopolar droplets into an isotropic phase and delocalization of MNPs, the Janus emulsions presented the same magnetic response (translational motion) as for Janus emulsions with radial organization (Figure S9a).

We also employed our methods to create MNP-NH$_2$-functionalized single-phase LC emulsions. By using mesogenic surfactants that control the LC anchoring at interfaces, we prepared LC droplets where mesogens are oriented perpendicular or parallel to the LC/W interface, leading to radial or bipolar configurations, respectively (Figure 5). When MNP-NH$_2$ were attached via imine formation to LC single emulsions with a radial configuration, a magnetic shell was created on the surface of the droplets, allowing translational motions by an external magnetic field in a controlled manner (Figure 5a, Video S7). In single emulsions with a bipolar configuration, two topological defects are located at the poles, and precise assembly of MNP-NH$_2$ at the defects was achieved. Bipolar LC MNP emulsions effectively roll toward the magnet, thereby displaying a controllable, but relatively unique, translational motion in response to an applied external magnetic field (Figure 5b, Video S8).

## CONCLUSION

In conclusion, we demonstrate new manipulations of HC/FC and LC/FC double and Janus emulsions through selective attachment of magnetic nanoparticles. Interfacial imine formation between selectively soluble aldehydes or aldehyde hydrates and water-dispersible MNP-NH$_2$ allowed for the strong attachment of MNP-NH$_2$ to the interfaces. Significant MNP-NH$_2$ coverage along the FC/W interface in HC/FC double emulsions resulted in macroscopic ferromagnetic behavior. Both encapsulated core–shell and Janus emulsions exhibited orientational attraction toward and repulsion away from an applied magnetic field owing to the induced magnetic N–S pole direction. Such responses resulted in the rotational movement and changes in orientation in space. The increase in ferromagnetic response was further revealed in magnetization curves, wherein an increase in magnetism with the confinement of MNP-NH$_2$ was observed. However, no increase in coercitvity was observed, indicating that small changes in the weak ferromagnetic coupling could induce large changes in macroscopic behavior. A structural-symmetry-dependent ferromagnetic response was also demonstrated. In addition, the results described above demonstrate that LC internal ordering is an efficient template to control the organization of MNP-NH$_2$ at the LC/W interface. We note that the LC-templated functionalization with MNP-NH$_2$ has features that make it distinct from the functionalization of emulsions comprising isotropic fluids. Although less ferromagnetic behavior was observed, the LC ordering and the selective concentration of MNPs at topological defects results in new ways to manipulate droplets through rolling and change the internal LC director field without changing the orientation or position of the droplet. The ease of attaching MNPs to droplets by interfacial imine forming reactions presents opportunities to create other active emulsion systems.

## ASSOCIATED CONTENT

### Ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acscentsci.0c00686.

> Supplementary Figures; Materials and Characterization Techniques; Synthesis and Characterization; Experimental Procedures (PDF)
>
> Video S1 − HC/FCdouble emulsions (HC/FC/W morphology) rotational response to repeated inversion of magnetic field (MP4)
>
> Video S2 − HC/FCdouble emulsions (HC/FC/W morphology) rotational response to horizontal movement of N-pole magnet around the sample (MP4)
>
> Video S3 − HC/FC double emulsions (Janus morphology) change in orientation in response to repeated inversion of magnetic field (MP4)
>
> Video S4 − HC/FC double emulsions (Janus morphology) rotational response to horizontal movement of N-pole magnet around the sample (MP4)
>
> Video S5 − LC/FC double emulsions (radial organization) movement towards the magnet (MP4)
>
> Video S6 − LC/FC double emulsions (monopolar organization) topological defect movement towards the magnet (MP4)
>
> Video S7 − LC single emulsions (radial organization) movement towards the magnet (MP4)
>
> Video S8 − LC single emulsions (bipolar organization) rolling towards the magnet (MP4)

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## Author Contributions

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## Notes

The authors declare no competing financial interest.

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