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Self-assembly of magnetic colloids under unsteady fields

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The use of magnetic fields offers an external, versatile way of controlling self-assembly of colloids. This review provides an exhaustive overview of unsteady fields that can vary in one, two, or three dimensions of space, as a powerful tool to direct the self-assembly of magnetic colloids into structures with tunable properties. Unlike steady fields, unsteady (nonstationary) fields can overcome the limitations of classical dipolar interactions, leading to a much wider range of structures, ranging from dense crystalline aggregates to 3D spanning networks, or dynamic clusters. The ability to precisely control the amplitude, frequency, and field direction allows for fine-tuning the interplay of interparticle forces, resulting in controllable assembly pathways. This review analyzes how different types of unsteady fields influence the morphology and dynamics of the self-assembled structures. Key parameters, such as the Mason number, are discussed to characterize the governing driving forces, and potential applications are highlighted.

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Introduction

Directed self-assembly makes use of external stimuli to drive the formation of complex ordered structures from individual components in both natural and engineered systems. Magnetic colloids have emerged as a versatile class of materials for designing self-assembled structures due to their responsiveness to externally applied magnetic fields. This responsiveness, combined with the ability to tune not only the colloid properties (e.g. size, shape, surface functionalization) but also the magnetic interactions (e.g. magnitude, attractive/repulsive nature), provides a powerful tool for potential applications in areas ranging from photonics and materials science to biomedicine and robotics [1]. Moreover, by modifying the magnetic interactions, it is possible to fine-tune the overall interactions between particles, including the balance between field-induced and intrinsic interactions (such as van der Waals, electrostatic, and hydrogen bonding forces). This enables the exploration of various phenomena that arise when competing driving forces are at play during self-assembly [2]. The directed self-assembly of magnetic colloids allows for unprecedented control over the morphology, topology, and functional properties of the resulting structures, which cannot be achieved through static self-assembly.

While the physicochemical properties of colloids (such as size, shape, and surface properties) enable access to a variety of new structures and dynamic behaviors, this review solely focuses on the influence of magnetic field protocols, particularly introducing a new degree of complexity by making the field time dependent (i.e. unsteady) in three dimensions. Traditionally, magnetic colloids have been assembled under uniaxial steady magnetic fields [3]. Under steady fields, particles become magnetized and interact. To a first approximation, it is assumed that each particle acquires a magnetic dipole with a magnitude $\vec{m} = V_p 3\beta\vec{H}$, where V_p is the particle volume, β is the contrast factor defined as $\beta = (\mu_p - \mu_c)/(\mu_p + 2\mu_c)$ (with μ_p and μ_c being the magnetic permeabilities of the particles and the carrier, respectively), and $\vec{H}$ is the external magnetic field.

In reality, however, due to the presence of neighboring particles, higher-order multipoles are also induced. Furthermore, the field acting on these particles is not solely the external field but rather the total field, which includes the perturbations created by neighboring particles. In addition, the linear relationship between $\vec{m}$ and $\vec{H}$ is valid only for superparamagnetic materials or ferromagnetic materials under very weak fields. Despite these complexities, such approximations are adopted for simplicity under the so-called mean magnetization

approximation (MMA), as they still capture the essential interaction characteristics. According to the MMA, the force between two particles is expressed as follows:

$$\overrightarrow{F} = -\frac{12\pi\mu_c a^6\beta^2 H^2}{r^4}\left[\left(3\cos^2\theta - 1\right)\hat{r} + \sin(2\theta)\hat{\theta}\right] \quad (1)$$

where a is the particle radius, r is the interparticle distance, and θ is the angle between the dipole direction and the interparticle vector. This anisotropic interaction is attractive along the direction of the field and repulsive in the perpendicular plane. Consequently, particles tend to aggregate into individual chains or dense columnar networks aligned with the field lines, depending on the particle concentration.

In contrast to steady magnetic fields, unsteady ones offer additional methods to control self-assembly and dynamics [4]. Unsteady magnetic fields can modulate interparticle interactions, expanding the range of achievable assemblies. For instance, at high field frequencies, the particle's slow response, either in terms of its motion within the viscous carrier or its dipole relaxation, results in particle interactions that differ significantly from the previously described dipolar interactions. In addition, magnetic torques arise due to the misalignment between the dipole and the external field ($\overrightarrow{\tau} = \overrightarrow{m} \times \overrightarrow{H}$) [5]. The ability to finely adjust external field parameters — such as amplitude, frequency, or orientation — allows for dynamic and reversible control of the assembled structures, paving the way for reconfigurable systems [6] and for structures that are closer to energy minima, and thus equilibrium [7]. Unsteady magnetic fields can also be used in controlling active systems, leading to nonequilibrium behaviors that often result in emergent collective dynamics, such as swarming, clustering, and phase separation [5]. The study of magnetic active colloids bridges the gap between equilibrium and nonequilibrium self-assembly, providing insights into both fundamental physics — serving as models for atomic or molecular systems — and potential technological applications [8].

In this review, we present a comprehensive overview of the self-assembly of magnetic colloids under unsteady magnetic fields. The review is structured into three sections, discussing assembly under magnetic fields that vary in one (uniaxial), two (biaxial), and three (triaxial) spatial dimensions, mostly on homogeneous, spherical particles not considering here neither patchy nor Janus particles in detail. As the dimensionality of the field increases, new degrees of freedom emerge, and the design space broadens. In each section, we highlight what in our opinion are the most relevant field configurations concerning equilibrium and nonequilibrium structures and their dynamics.

Uniaxial

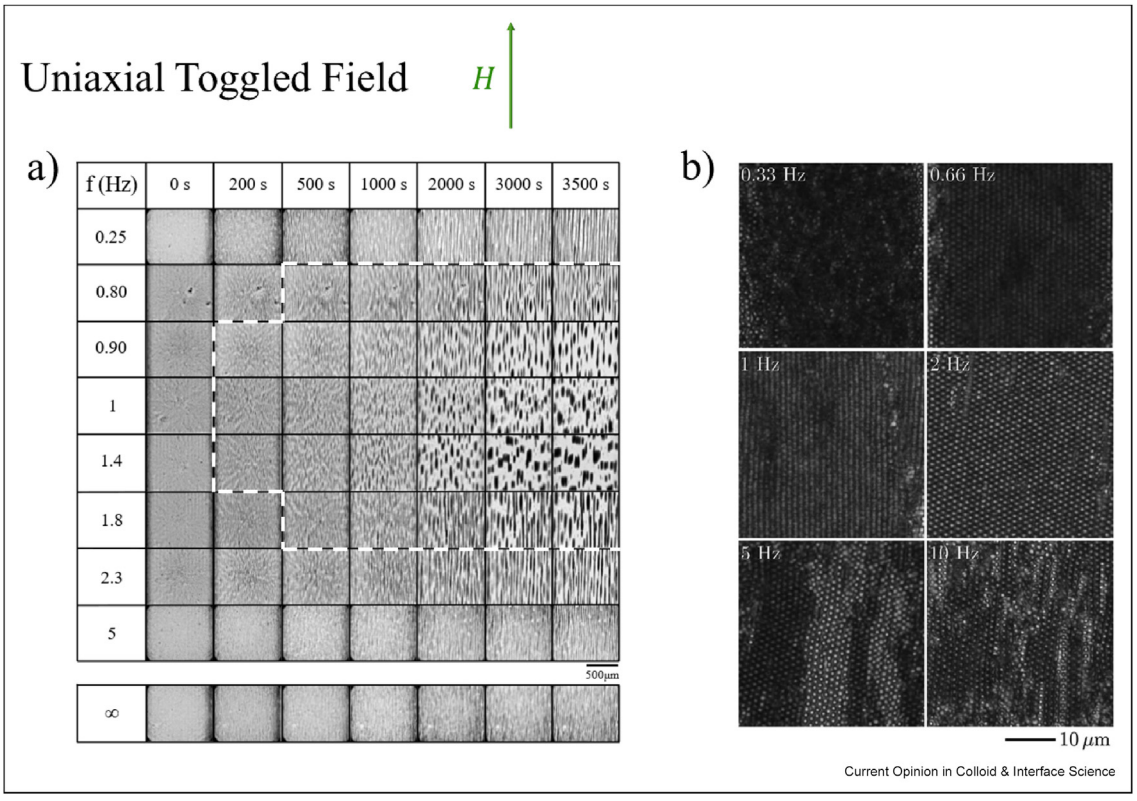
Toggled fields

The behavior of magnetic colloids under a uniaxial steady field has been widely examined due to their notable mechanical properties [3]. Steady magnetic fields typically induce the formation of columnar structures in the field direction that are especially suitable for momentum transfer applications. However, generated structures are not energetically optimal because they are often metastable and trapped in local energy minima. In contrast, the employment of unsteady magnetic fields can promote significantly different structuring dynamics, allowing particles to escape these kinetically trapped states and reorganize over time into more energetically favorable configurations forming defect-free crystals [9].

Since the pioneering works of Promislow and Gast [10], pulsed magnetic fields have been used to obtain highly crystalline, minimum energy structures. After applying a sequence of field pulses, the classical columnar structures collapse into dense ellipsoidal aggregates. These final aggregate shapes result from the interplay of demagnetization fields, magnetic surface energy, and interaggregate interactions. However, the dynamics and conditions required for the aggregate formation have not been well understood until the last decade. First, Swan et al. pointed out the frequency of the pulsed field (f) as a key parameter in the transformation of structures into dense ellipsoidal aggregates [11]. When the field is off, thermal motion allows for structural reorganization, leading to a gradual relaxation toward a final state. The pulse frequency introduces a characteristic time scale ($\sim f^{-1}$). Comparing this one with the diffusive time scale t_d, driven by Brownian motion, defines the critical frequency for forming these condensed structures. At very low frequencies, particle correlations and structural integrity are lost between cycles, causing the colloid to remain arrested. Conversely, at very high frequencies, particles do not have sufficient time for restructuring. Therefore, a narrow frequency range around $f \sim t_d^{-1}$ is optimal for observing depercolated ellipsoidal structures (Figure 1a).

Within this critical depercolation regime, the magnetic field strength significantly influences the dynamics of structure collapse. At low field strengths, there is a two-stage coarsening process; an initial growth limited by diffusive percolation, followed by a ballistic growth during structure collapse, with the transition time decreasing as field strength increases. At high field strengths, stronger lateral dipolar repulsion between aggregates leads to a single (i.e. one stage) rapid growth phase [12]. Recent studies show that adjusting the duty ratio ξ (i.e. the time the field is turned on during each toggling cycle) provides finer control over structural evolution. Under specific field conditions, a novel

Figure 1



Self-assembly under uniaxial toggled fields. **(a)** Structure evolution over time as a function of toggling frequency. Depercolated ellipsoidal aggregates regime, marked with dashed lines, occurs around the critical frequency $f \sim t_d^{-1}$ [12]. **(b)** High-resolution microscopy images of the self-assembled structures. The structuration under the critical frequency yields highly crystalline structures close to energy minimum [11]. Reproduced from Ref. [11] with permission from the Royal Society of Chemistry.

percolating microstructure can form perpendicular to the magnetic field, exhibiting continuous dynamics of rotation, rupture, and coalescence [13].

Microscopy and scattering techniques demonstrate that the internal structure of these ellipsoidal aggregates corresponds to a crystalline arrangement of particles, consistent with thermodynamic predictions [9] (Figure 1b). Controlling the internal crystalline order of the microstructures can significantly influence the macroscopic properties of magnetic colloids. Toggled fields can be thought as an analogous method to thermal annealing and therefore to reduce defects, enhancing certain physicochemical properties such as conductivity [14] and rheological performance [15]. In particular, Camacho et al. [15] demonstrated that by superimposing a saturating toggled magnetic field, it is possible to enhance the rheological response, promoting coarsening and crystallization. These annealed structures exhibit up to 30% higher yield stresses compared to those formed under conventional uniaxial steady fields in good agreement with numerical calculations on magnetic lattices. From a wider perspective, the ability to finely control the formation of colloidal crystals with diverse morphologies has also potential applications in the development of smart materials, photonic gels and crystals, or improved coatings [7].

Biaxial

The range of possible field protocols increases significantly by extending the field generation to a plane. For instance, any 2D Lissajous figure can be generated when superimposing harmonic signals in two perpendicular directions. Here, we will focus our attention on two particular configurations due to their simplicity and demonstrated utility in the literature: rotating and perturbating fields.

Rotating fields

This particular field configuration consists of the superposition of two sinusoidal fields of equal frequency f and amplitudes in phase quadrature. Given two arbitrary orthogonal directions $\hat{i}$ and $\hat{j}$, a rotating magnetic field can be written as follows:

$$\vec{H}(t) = H[\sin(2\pi ft)\hat{i} + \cos(2\pi ft)\hat{j}] \quad (2)$$

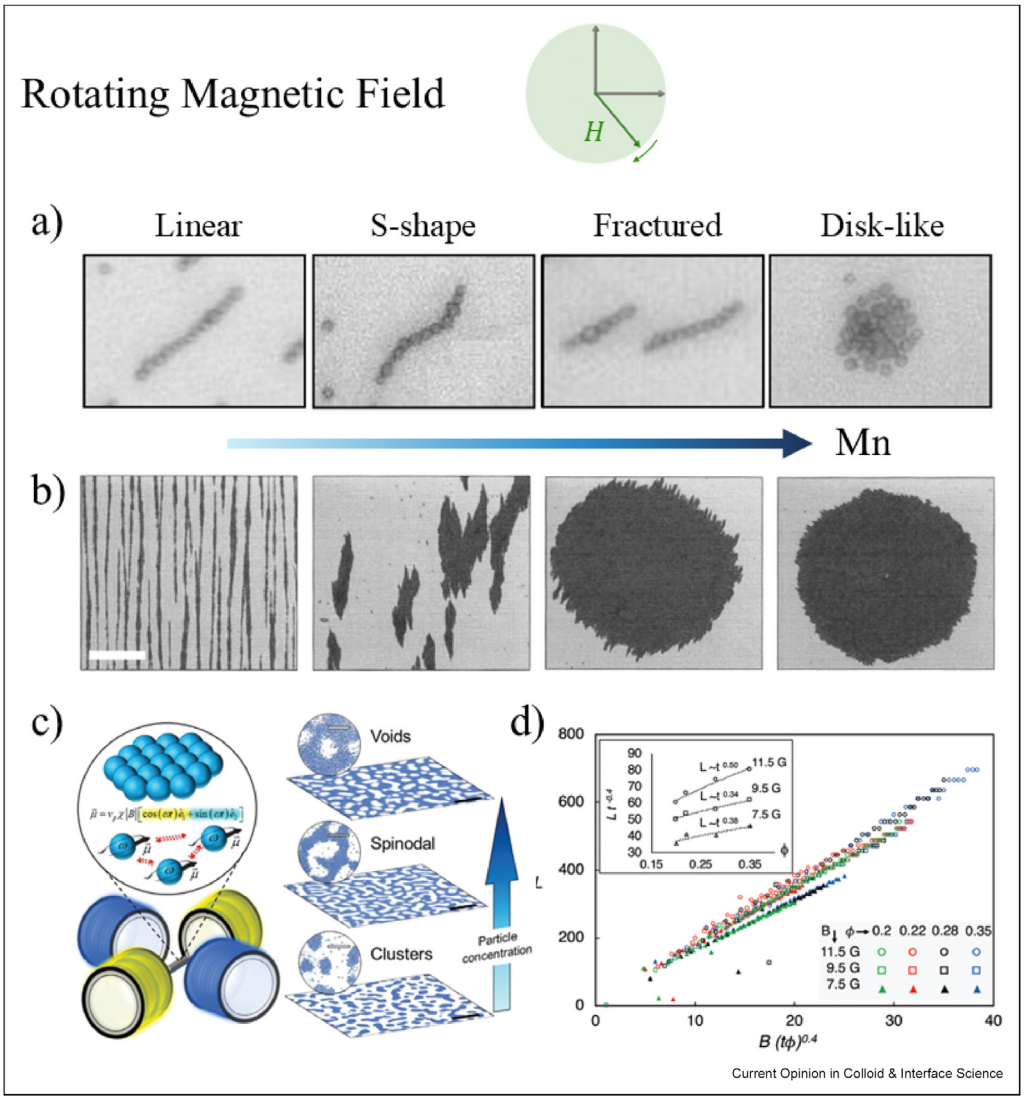
It has been used extensively in controlled self-assembly of colloidal crystals [16], micromanipulation [17], magnetic walkers [18], and photonic applications [19].

The behavior of magnetic colloids under rotating fields is dictated by the Mason number (Mn), a dimensionless number that measures the relative importance of hydrodynamic to magnetic forces. Mn was originally introduced for electro- and magneto-rheological fluids under shearing flows but was later extended to describe the dynamics of field-induced structures under unsteady fields. In the case of rotating fields, in the linear magnetization regime, it reads as follows [20,21]:

$$\mathrm{Mn} = \frac{32\pi\eta f}{\mu_c \beta^2 H^2} \tag{3}$$

Here η is the viscosity of the carrier liquid. Based on the Mason number, two distinct regimes can be identified when considering the structures induced by rotating magnetic fields. At low Mn (Mn≪1) particles form chain-like structures following the rotating field that can be

Figure 2



Self-assembly under rotating magnetic fields. **(a)** At low Mn and ϕ, particles assemble in chains that rotate with the field. As Mn is increased, the hydrodynamic forces tend to bend the chain to reduce drag eventually fracturing it. In the high Mn limit, viscous drag dominates, and time-averaged interactions yield the formation of disk-like clusters [36]. **(b)** At higher ϕ, chains are not capable to rotate independently, and both collision and interaggregate interactions favor the formation of disk-like aggregates at much lower Mn values [21]. Reprinted figure with permission from [21] Copyright 2025 by the American Physical Society. **(c)** In the time-averaged interaction regime, sheet-like structures are obtained with morphologies that are dependent on particle loading [26]. Reprinted figure with permission from Royal Society of Chemistry. **(d)** The characteristic length scale of these aggregates can be collapsed onto a master curve that follows a power law with time [26]. Reprinted figure with permission from Royal Society of Chemistry.

either straight, S-shaped, U-shaped, or even thick rods, depending on the frequency and the particle volume fraction. Conversely, at high Mn ($Mn \gg 1$), hydrodynamic interactions are capable to break the chains, leading to the formation of isotropic disk-like clusters (sheets), in dilute (concentrated) colloids, contained in the plane of rotation. The dynamics of self-assembly in both regimes have been extensively studied over the past few decades.

On the one hand, at low Mn, field-induced structures can follow the field although with a certain phase lag. Melle et al. made significant efforts to understand the dynamics of rotating chains at low frequencies [20,22,23]. At low particle volume fractions (below 0.02 in Ref. [21]), single-particle-width chains form, whose morphology and steady-state dynamics can be explained in terms of Mn. By simply balancing hydrodynamic and magnetic torques acting on the chains, it can be shown that their average length scales as $\sim Mn^{-1/2}$, which agrees well with experimental observations [23]. Such a torque balance also explains the appearance of a phase lag between the field and chain directions, as well as the development of an S-shape. Both the maximum phase lag and tension occur at the central bead of the chain, causing the chain to break at this point, thereby reducing its length for the chain to remain stable. The self-assembly process in the initial transitory regime (i.e. before reaching steady state) has also been studied under the frame of the Dynamic Scaling Theory either speeding up or down the growth dynamics depending on whether chains interact or not, respectively [24] (Figure 2a). Rotational dynamics can be enhanced using chains made of chemically linked particles whose rigidity can be tuned by the particle linker. These flexible chains serve as toy models of elastic rods that can rotate synchronously with the external field at low Mn, rotate asynchronously with the field when the frictional torque exceeds magnetic torque (i.e. large Mn), and even bend into complex shapes to minimize frictional torque [25]. Finally, at high-volume fractions, interchain interactions result in thick clusters with different rotational dynamics. In this case, the transition from anisotropic (chain-like) rotating structures to isotropic (disk-like) structures occurs at Mn much lower than 1 due to collisions between rotating aggregates. The resulting structures are large isotropic disks that rotate slowly, with short rod-like clusters rotating and circulating along the surface [21] (Figure 2b).

On the other hand, at high Mn, the behavior changes drastically. Hydrodynamic drag prevents the structures from following the field, and magnetic interactions become effectively isotropic in the field plane, as the dipole−dipole interaction averages over one field cycle. This averaged interaction is attractive in the field plane and repulsive in the perpendicular direction, leading to the aggregation of particles into disk-like clusters or sheets (Figure 2a). Once the critical Mn for averaged interactions is exceeded, the governing parameters of the resulting phase are the magnetic field strength H and the particle concentration ϕ. At low ϕ, colloids nucleate into isolated clusters that coarsen over time. As ϕ increases, percolating structures emerge, appearing as spinodal decomposition at moderate particle loadings and as a particle-rich phase with voids that coarsen over time at higher ϕ [16,26] (Figure 2c). Magnetic field strength also plays a crucial role in the final structure; increasing H transitions the structures from a disordered microscopic arrangement, due to significant Brownian motion, to a highly crystalline state. The final size of the structures is determined by the strength of the magnetic interactions. The dynamics of structure growth under large Mn have been shown to follow a power law, scaling with $H^2 t^{0.4} \phi^{0.4}$ [26] (Figure 2d). Hydrodynamic interactions influence not only cluster formation but also individual particles, accelerating cluster formation and inducing both single-particle and collective cluster rotation, as observed experimentally [27,28].

While the low and high Mn regimes are well understood, recent attention has also focused on the transition region [19,29]. As mentioned before, when the rotation frequency is gradually increased starting from low values, rotating chains break up into smaller chains that can resist the viscous drag. In the boundary region with intermediate frequencies, it has been shown that before the formation of disk-like clusters, rotating chains exist. These chains can interact between them to form disk-like clusters, leading to a mixture of chains and disks that can eventually reach a steady state.

Rotating fields are particularly useful in micromanipulation applications, including bead transport and assembly [30−32], magnetic rotors [33], and microswimmer propulsion [34,35].

Perturbating fields

Also referred to in the literature as *oscillating* or *fluctuating magnetic fields*, this configuration involves the superposition of a uniaxial steady magnetic field and an alternating sinusoidal field along an orthogonal axis. We will use the term "perturbating" because in the literature, the alternating field is usually of low intensity. However, in the discussion below, we will not limit ourselves to that particular case. In this field configuration, the resulting field vector traces a triangular area:

$$\vec{H}(t) = H_{st}\,\hat{i} + H_{alt}\sin(2\pi f t)\,\hat{j} \tag{4}$$

Here, $\hat{i}$ and $\hat{j}$ represent two arbitrary orthogonal directions, and H_{st} and H_{alt} represent the amplitudes of the steady and alternating fields, respectively. Perturbating fields differ slightly from rotating ones by introducing an additional degree of freedom: the maximum angle swept by the field

vector $\theta_p = \arctan(H_{alt}/H_{st})$. A Mason number can again be defined to balance hydrodynamic drag and interparticle magnetic interactions [37,38]:

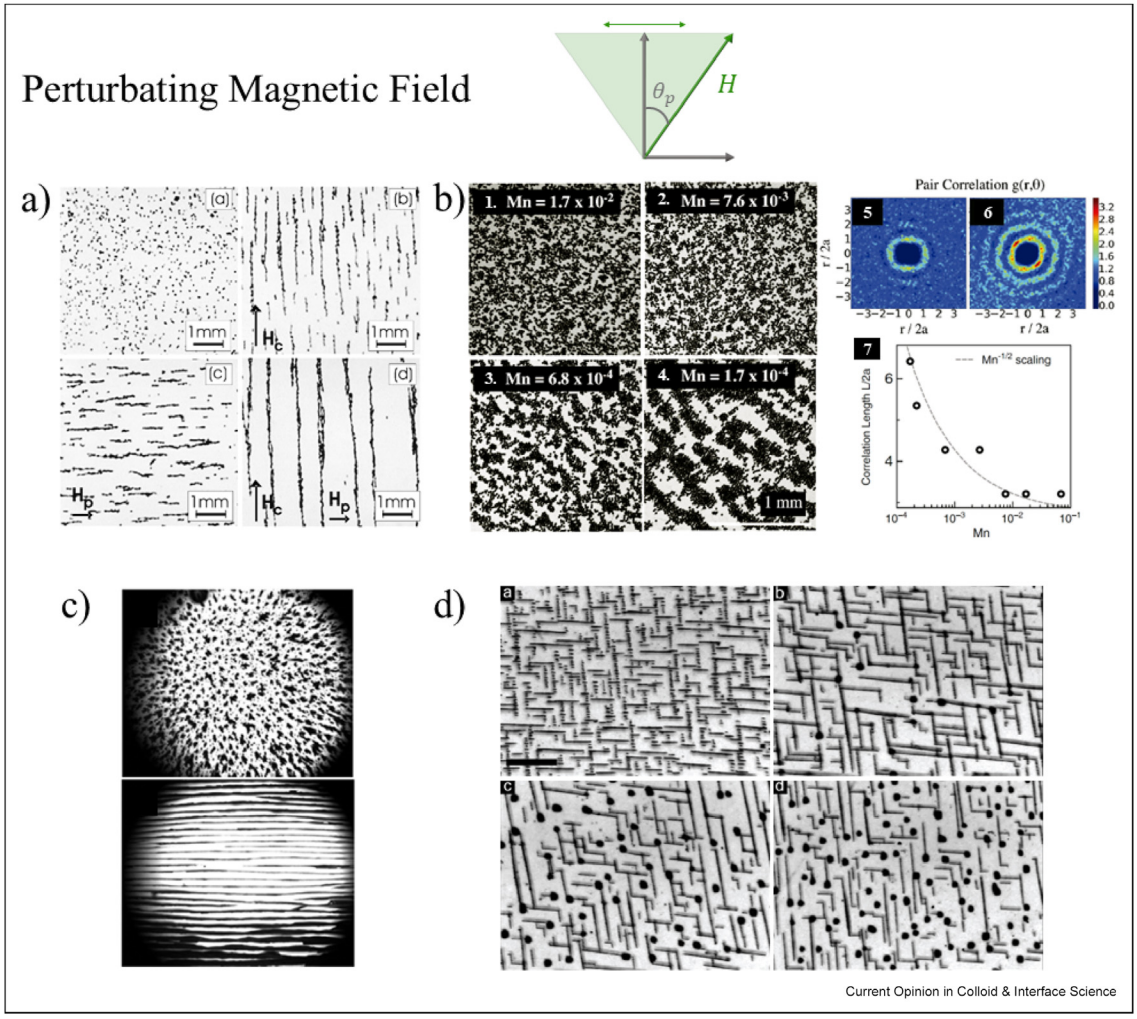
$$\mathrm{Mn} = \frac{32\pi\eta f\theta_p}{\mu_c\beta^2 H^2} \quad (5)$$

In the case of perturbating fields, viscous drag and magnetic interparticle forces change with time during a magnetic field cycle. Hence, Equation [5] can be considered as an upper bound estimate. Moreover, in contrast to rotating fields, the magnetic field magnitude does not remain constant throughout the cycle. To account for this, the averaged field value during a period can be used: $H^2 = H_{st}^2 + H_{alt}^2/2$.

However, many authors restrict their analysis to the small perturbation angle approximation where $H \approx H_{st}$.

Similar to previous field configurations, self-assembled structures obtained under perturbating fields can be understood in terms of dimensionless numbers; in this case, the Mason number Mn and the perturbation angle θ_p. Most frequently, Mn is tuned by changing f and/or H. At $\mathrm{Mn} \ll 1$, particle structures are expected to move following the field, while $\mathrm{Mn} \gg 1$ places the system in a regime of time-averaged interactions. In addition to this transition, for a given Mn, it is anticipated the existence of a critical θ_p that must be exceeded in order to break the chains and allow particles to cluster into sheets [39].

Figure 3



Self-assembly under perturbating magnetic fields. **(a)** The superposition of a small-amplitude oscillatory perturbation orthogonal to the steady field component increases significantly the final size of the structures because repulsive regions of dipolar interparticle interaction are hindered by the perturbation [37]. Reprinted figure with permission from [37]. Copyright 2025 by the American Physical Society. **(b)** At high ϕ, low Mn perturbations induce a certain degree of crystallinity, as revealed by the pair correlation function. In this regime, the characteristic length of the aggregates decays with $\mathrm{Mn}^{-1/2}$ [39]. Reprinted figure with permission from [39]. Copyright 2025 by the American Physical Society. **(c)** In the high Mn, time-averaged interactions govern the self-assembly and chain-like structures are formed at low θ_p (upper snapshot) and sheets at high θ_p (lower snapshot) [38]. Reprinted with permission from IOP Publishing. **(d)** In the intermediate Mn regime, both chains and sheets (disks) can coexist [44]. Reprinted with permission from ACS. Copyright 2025 American Chemical Society.

Structures closely follow the field in the low Mn regime. Donado et al. pioneered the use of low f perturbating fields as a mean to promote interchain aggregation [37]. At low f and θ_p, chains oscillate following the field favoring the lateral aggregation, either by direct collisions between aggregates or through additional dipolar interactions [40]. This introduces new mechanisms for lateral chain aggregation that do not occur in the absence of the perturbation. An optimal frequency was reported for the maximum chain length under perturbating fields if compared to uniaxial steady fields [37] (Figure 3a). Moreover, an enhanced rheological response was found due to the thicker and stronger formed structures [38]. Similar to rotating fields, oscillating chains under high-angle perturbating fields θ_p can deform into S-shapes as a result of the balance between hydrodynamic and magnetic torques. Further increasing θ_p chains experience fracture events when their length exceeds a critical value that also scales as $\sim \mathrm{Mn}^{-1/2}$ [41]. At high particle concentrations, thick aggregates form instead of individual particle-width chains. The perturbating field enhances clustering, and a certain degree of crystallinity is induced as the structures become more densely packed [39]. Consistently, the length scale of these dense clusters also follows $\sim \mathrm{Mn}^{-1/2}$ (Figure 3b).

Time-averaged interactions come into play in the high Mn regime. The additional degree of freedom introduced by the perturbation angle allows for a wider range of achievable structures if compared to the rotating field case. In this regime, self-assembled structures tend to be static rather than dynamic. Very low perturbation angles ($\theta_p \sim 0^o$) result in static columnar structures similar to those observed under uniaxial steady fields. The same occurs as the perturbation angle approaches its maximum value ($\theta_p \sim 90^o$). Here, the averaged interaction becomes equivalent to that of a uniaxial steady field oriented in the perturbation direction, with a field amplitude equal to its root mean square value. Interestingly, in between these two extreme values ($0^\circ < \theta_p < 90^\circ$) sheets formation has been recently reported (Figure 3c), with a strongly loading-dependent morphology, similar to the case of rotating fields [42]. Such a similarity is expected because the averaged dipolar interaction for a perturbating field with $\theta_p = 54.7^o$ is identical to that of a rotating field. The growth dynamics of these layered structures also follow a power law, consistent with Dynamic Scaling Theory. In particular, initially formed chains "zipper" together to form sheet fragments, which further merge due to the attractive averaged interaction in the field plane [43].

The transition behavior from low to high Mn regimes has received limited exploration. Nagaoka et al. demonstrated that disk-like and chain-like aggregates coexist at intermediate frequencies and $\theta_p = 45^o$. Moreover, by tuning θ_p, they were able to assemble a crosshatched pattern [44] (Figure 3d).

Although the literature of magnetic colloids under perturbating fields is scarce, these fields have been successfully applied in microswimmers and microfluidic devices [45], as well as providing an immediate solution to significantly enhance the rheological response of magnetorheological fluids [38].

Triaxial

This section encompasses unsteady magnetic fields that vary in all three spatial dimensions, creating new possibilities for field protocols that lead to novel particle assemblies and dynamics. We first focus on precession fields, the most widely studied triaxial unsteady field configuration. Then, we discuss other field configurations that result in exotic assemblies and dynamic behaviors.

Precession fields

Precession magnetic fields are created by superimposing a rotating field and a uniaxial steady field oriented along the axis orthogonal to the rotation plane. Given three arbitrary orthogonal directions $\hat{i}$, $\hat{j}$ and $\hat{k}$, a precession magnetic field can be written as follows:

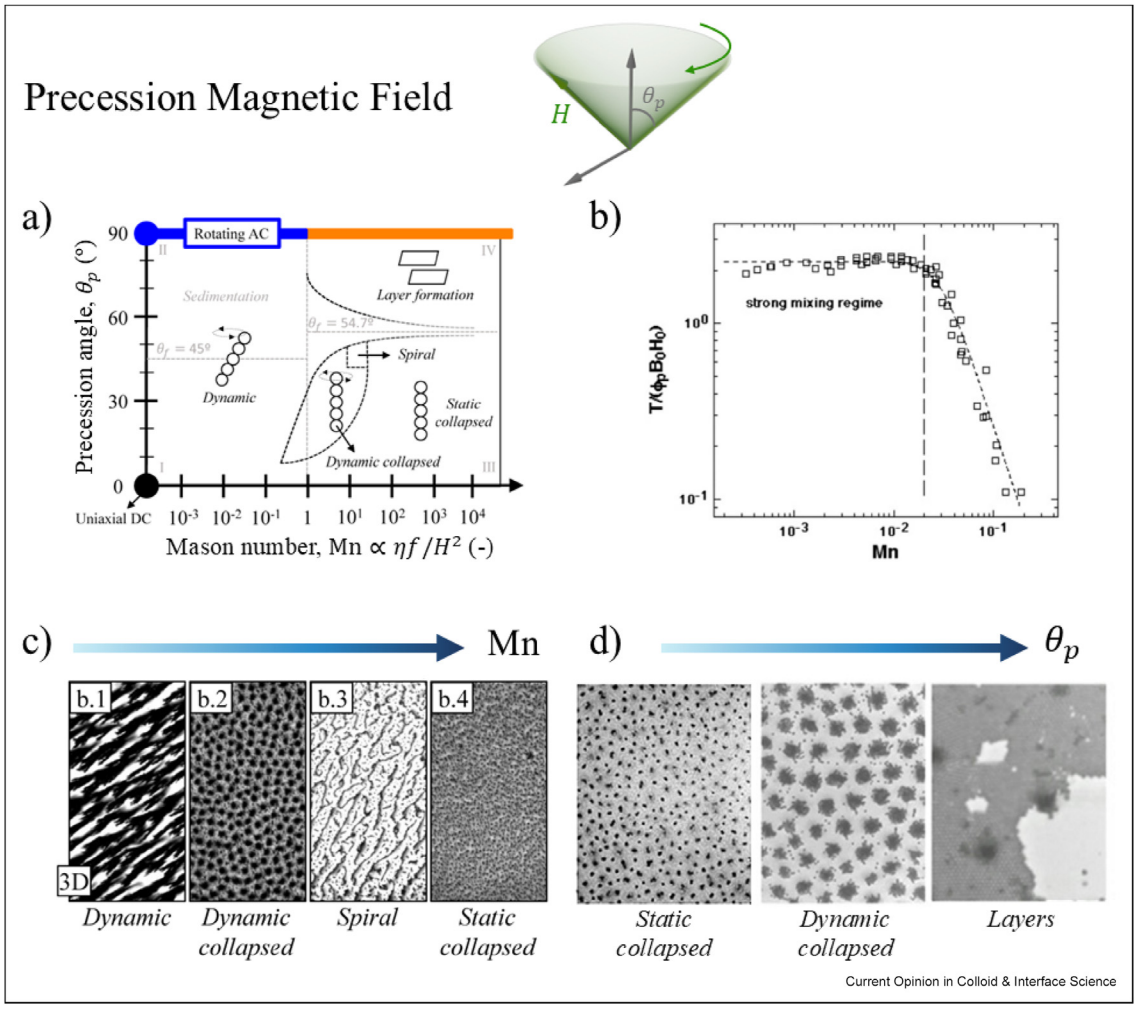
$$\vec{H}(t) = H\left\{\sin\theta_p[\sin(2\pi ft)\hat{i} + \cos(2\pi ft)\hat{j}] + \cos\theta_p\hat{k}\right\} \tag{6}$$

Here, $0^\circ < \theta_p < 90^\circ$ is the precession angle and represents the angle of the cone swept by the field vector. This configuration can be thought as a 3D extension of a perturbating field. It also bridges the gap between uniaxial steady ($\theta_p = 0^\circ$) and rotating fields ($\theta_p = 90^\circ$). Again, the distinction between dynamic and static structures can be made using the Mason number [38]:

$$\mathrm{Mn} = \frac{32\pi\eta f \sin\theta_p}{\mu_c\beta^2 H^2} \tag{7}$$

Interestingly, in contrast to perturbating fields, the angular velocity is now steady, and the field amplitude is constant along a cycle. Figure 4a shows the phase diagram for precession fields in terms of the two dimensionless numbers Mn and θ_p. In the low Mn regime, structures do have time to follow the field vector. Generated chain-like structures precess with the field and induce strong fluid vortices. For this reason, precession fields at low f are often referred to as *vortex fields* in the literature. Martin et al. [46,47] extensively studied this regime, focusing on its mixing properties at the microscale. They demonstrated that the mixing torque arises due to the phase lag between the field and the chains orientation. By

Figure 4



Self-assembly under precession magnetic fields. **(a)** Phase diagram of the generated structures as a function of Mn and θ_p [51]. Reprinted with permission from [51]. Copyright 2023, The Society of Rheology. **(b)** Torque density versus Mn in a suspension of magnetic spheres. At low Mn, dynamic chains following the field are able to generate a strong vorticity that can be useful in mixing applications [47]. Reprinted figure with permission from [47]. Copyright 2025 by the American Physical Society. **(c)** Effect of Mn in the low θ_p regime. As Mn is increased, structures start feeling time-averaged interactions and collapse in chains along the steady axis [51]. Reprinted with permission from [51]. Copyright 2023, The Society of Rheology. **(d)** Effect of θ_p in the high Mn regime. As θ_p is increased, layers are formed in the rotatory plane [57]. Reprinted (adapted) with permission from [57]. Copyright 2016 American Chemical Society.

balancing hydrodynamic and magnetic torques, they explained the morphology of the chains in their steady state, showing that the chains exhibit not only an azimuthal lag but also a polar angle smaller than that of the field relative to the steady axis. The chain length is limited by viscous torque, and longer chains can only grow by reducing the polar angle (collapsing toward the steady axis) or increasing the phase lag. Chains eventually reach a dynamic equilibrium, continuously breaking and reforming upon reaching a critical lag. Theoretical predictions were supported by experimental measurements of the mixing torque that appears to follow a universal behavior as a unique function of Mn. A strong mixing regime was observed at low Mn, with mixing power diminishing rapidly at higher Mn values (Figure 4b). Experiments involving anisotropic particles demonstrate an even stronger vorticity generation, as shown with ellipsoids or platelets [48,49]. In addition, by carefully depositing particles on a liquid–air interface, this mixing protocol has shown to be suitable to mix molecules, accelerating surface–chemical reactions, as well as in metamaterial applications [50].

In the high Mn regime, time-averaged interactions come into play. Two types of self-assembled structures can be distinguished depending on θ_p (Figure 4a). At low precession angles ($\theta_p \sim 0^o$), the averaged interaction is similar to that of a uniaxial steady field inhibiting structural transformations [51] (Figure 4c). When θ_p increases the rotational component becomes dominant,

and the averaged interaction resembles that of a rotating field. For $\theta_p > 54.7^o$, the repulsive component of the dipolar interaction induced by the steady field becomes negligible when compared to the attractive component induced by the rotating field, causing particles to collapse into isotropic structures, as observed in rotating fields. These structures can manifest as either separate disk-like clusters or connected layered/sheet-like structures, depending on the particle concentration.

Several studies have explored the assembly of dilute magnetic colloids in disk-like clusters under high-f high-θ_p precession fields [52,53]. Clusters formed under high-f fields rotate more slowly than the external field because of imbalanced magnetic forces at the edges of the cluster. Treating the cluster as a viscoelastic material, the rotation dynamics depends on cluster size, field strength and frequency, particle properties, and the viscoelastic characteristics of the clusters themselves. Using this rotational behavior, *in situ*—assembled microwheels can be precisely controlled in terms of rolling velocity and direction by carefully tuning f and θ_p [54]. Recent work has shed further light on the rotation mechanism at fluid interfaces [55]. In a different approach, Dobnikar et al. [56,57] eliminated the net rotation of clusters at high frequencies by reversing the field rotation direction every cycle. When the particle volume fraction is large enough, the superposition of high-f high-θ_p precession fields results in the formation of connected layered structures or sheets [51] where rotation dynamics are suppressed due to the even distribution of torques [57] (Figure 4d). Net forces only appear at the edges or near defect zones within the structures. Gravity can also play a significant role, as large layers tend to sediment rapidly if they form in a plane orthogonal to the direction of gravity.

The transition (i.e. intermediate Mn) regime has been explored in several works [51,57]. In this regime, colloidal assembly is inherently coupled to the system's dynamics, and averaged interactions fail to capture the emergent behavior, adding another level of complexity. These studies report novel dynamic structures, such as motile chains, spirals, and chaotic rotating ensembles, observed in both experiments and simulations. The utility of these dynamic structures has also been highlighted in recent work [42,58], showing that precession fields can promote lateral chain coalescence in high-concentration magnetorheological fluids, enhancing their rheological response, similar to the effects previously described in perturbating fields.

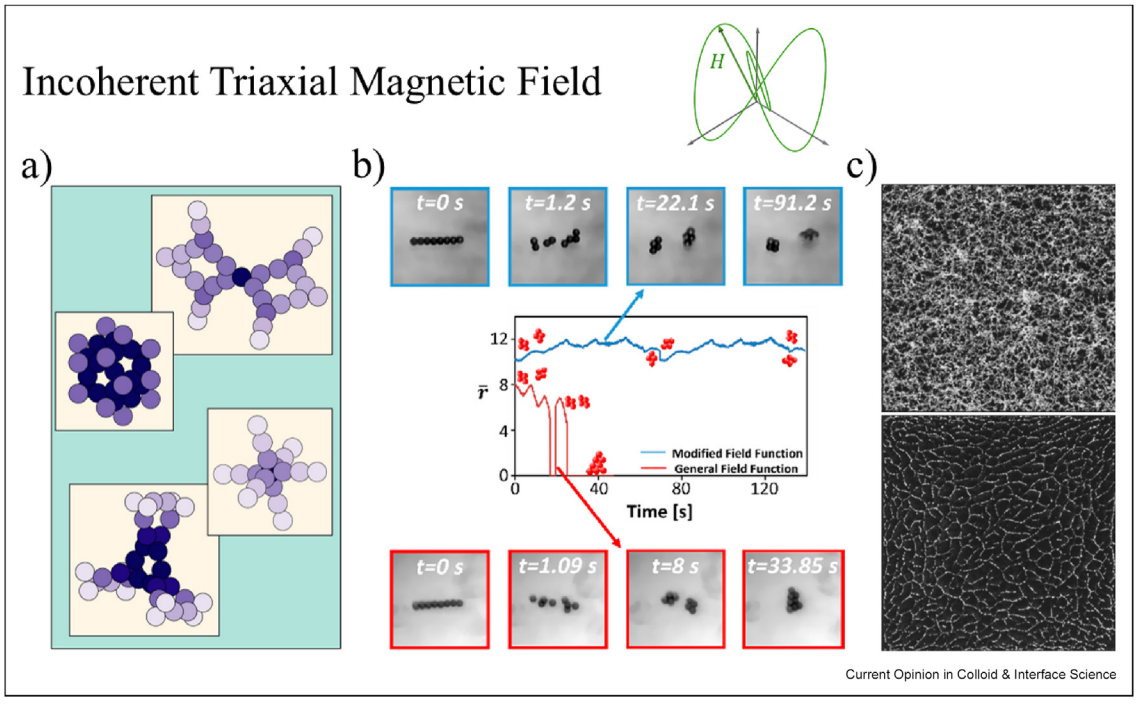
Most of the studies discussed so far have focused on spherical, uniform particles. However, more complex colloidal constituents can lead to new assemblies and dynamics. For example, Janus particles have been shown to form tubular structures [59], pearl chains, or close-packed aggregates [60]. Chemically linked chains exhibit S- and U-shapes in rotating fields but undergo shape transformations under precession fields. This transformation has been observed both theoretically [61] and experimentally [45], with chains forming complex helicoidal geometries. These helices can propel themselves under the same fields, suggesting potential applications in cargo transport through complex 3D environments. Nanoparticles under precession fields combined with light irradiation can form tornado-like microswarms capable of vertical motion and controlling chemical reaction rates by trapping reactants inside [62]. Peanut-shaped particles can act as microrobots, forming ribbons precisely directed via tumbling displacements under precession fields [63]. Finally, colloidal currents can be induced by inclined precession fields through spatial symmetry breaking [64].

Incoherent fields

This section is devoted to more general triaxial fields consisting in the superposition of three orthogonal sinusoidal fields with different amplitudes, frequencies, and phase lags [65]. In this way, any 3D Lissajous figure can be generated. However, a precise control over the phase lags is not easy to achieve, especially at high frequency, and requires the use of dedicated RLC circuits with capacitor banks [66] run in real time by control software tools [67].

Despite the vast design space offered by triaxial unsteady fields, only a few researchers have addressed these protocols, and thus, the available references are very scarce. Martin et al. have been the most prominent contributors to the theoretical and experimental framework to explain interparticle interactions in incoherent triaxial magnetic fields [68]. In terms of self-assembly, the time-averaged regime of incoherent fields presents opportunities for novel and exotic interactions. The simplest triaxial unsteady incoherent field is the so-called balanced case. In this particular configuration, all three oscillating field components have the same root mean square value. Under this configuration, the averaged dipolar interaction induced by the external field cancels out, meaning that any particle interaction must arise from the magnetization induced by neighboring particles. Therefore, these interactions need to be calculated self-consistently as they involve many-body effects. Interestingly, for the case of two isolated particles, the averaged interaction is central and short ranged, losing some of the basic features of the dipolar interaction [56,69]. Numerical simulations for a small number of particles have shown that noncompact structures are expected, with randomly oriented hexagonal sheets being the most energetically favorable arrangement [69] (Figure 5a). Further experimental works [65] have tested the stability conditions for forming singlets, doublets, or quadramers by adjusting the relative frequencies of the field components (Figure 5b).

Figure 5



Self-assembly under incoherent triaxial magnetic fields. **(a)** Low-energy structures under balanced triaxial fields. Many-body interactions lead the formation of noncompact aggregates [70]. Reprinted figure with permission from John Wiley and Sons. **(b)** Formation of stable and unstable quadramers. To obtain stable quadramers [65], superimposes periodic signals with a dissimilar frequency in one of the axes. Reprinted figure with permission from Elsevier. **(c)** In a triaxial field, foams are assembled (upper snapshot). Heterodyning the three components produces foams with optimal properties (lower snapshot) [73]. Reprinted figure with permission from [73]. Copyright 2004 by the American Physical Society.

For colloids with many particles, it is difficult to experimentally achieve the noncompact structures predicted by simulations due to sedimentation. To address this without resorting to complex stabilization methods, a slightly different frequency is applied to each axis. This technique, known as *heterodyning*, creates a secondary slow variation in the magnetic field compared to its base frequency. Magnetic colloids respond strongly to these beats, which can be viewed as an oscillation between coherent and incoherent interactions. Using heterodyned fields, isotropic foam-like networks can be self-assembled, offering optimal physical properties, such as enhanced magnetic permeability and mechanical resistance (see Figure 5c) [70].

By breaking the symmetry of the balanced triaxial field — either by varying the relative amplitudes along each axis or heterodyning only two components — anisotropy can be introduced into the system. This enables the tuning of physical properties to enhance specific characteristics in a desired direction [70].

More recent studies have explored the potential of incoherent fields as a powerful tool for generating vorticity, extending the mixing torque concept reported in precession fields. In this particular application, low frequencies are used so that structures can follow the field vector. Using complex symmetry considerations [71] or chaining models [72], the vorticity induced by any triaxial configuration can be predicted based on the relative frequencies and phases of the components, showing good agreement with experimental results. Consequently, triaxial magnetic fields provide precise control over the vorticity vector by fine-tuning field parameters.

Conclusions

Unsteady magnetic fields offer an extensive toolkit for manipulating colloids, driving the formation of unique structures and dynamics across various configurations. Starting with uniaxial toggled fields, researchers have demonstrated the ability to overcome kinetic barriers to achieve minimum energy assemblies with high crystalline order. Rotating fields further expand this by introducing a steady rotation that encourages particles to form compact, disk-like clusters. These fields allow for the development of highly ordered structures with specific rotational behaviors that can be fine-tuned by adjusting field parameters such as strength and frequency. Perturbating fields introduce an additional degree of freedom by superimposing a steady field with an oscillating one in an orthogonal direction. This creates a sweeping motion of the magnetic field, allowing

for the assembly of elongated chains and more robust structures. Perturbating fields are capable to enhance particle aggregation (at small angles) and induce deformations and even fractures in the particle chains (at large angles).

Precession fields, a combination of steady and rotating fields, add an additional dimension of complexity by introducing a precession angle in the 3D space. This creates a range of dynamic behaviors, from vortex-like mixing at low Mason numbers (Mn) to isotropic structures at higher Mn. The precession field's ability to enhance mixing on the microscale or drive novel particle assemblies makes it a valuable tool for applications in microfluidics, material design, and surface−chemical reactions [50,74,75]. Moving to more complex schemes, triaxial incoherent fields — where the field oscillates across all three spatial dimensions — further enrich the landscape of magnetic control. By carefully adjusting the relative amplitudes, frequencies, and phase lags of the magnetic components, the formation of noncompact, foam-like structures and anisotropic networks is reported. These assemblies are highly relevant for the development of functional materials and other advanced systems.

In summary, unsteady magnetic fields — whether uniaxial, biaxial, or triaxial — enable an unprecedented level of control over particle self-assembly and dynamics. As researchers explore more complex field configurations and particle types, these techniques will continue to advance applications in fields such as materials science, microfluidics, and nanotechnology.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

No data was used for the research described in the article.

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