Dynamics and interactions of magnetically driven colloidal microrotors

Cite as: Appl. Phys. Lett. 120, 081601 (2022); https://doi.org/10.1063/5.0076574
Submitted: 26 October 2021 • Accepted: 05 February 2022 • Published Online: 22 February 2022

Raúl Josué Hernández Hernández, Thomas M. Fischer and Pietro Tierno

COLLECTIONS

This paper was selected as an Editor’s Pick

Timing is everything. Now it’s automatic.
A new synchronous source measure system for electrical measurements of materials and devices
Learn more
Dynamics and interactions of magnetically driven colloidal microrotors

We study the pair interactions between magnetically driven colloidal microrotors with an anisotropic shape. An external precessing magnetic field induces a torque to these particles spinning them at a fixed angular frequency. When pair of rotors approach each other, the anisotropic particles interact via dipolar forces and hydrodynamic interactions (HIs) excited by their rotational motion. For applied field spinning close to the magic angle, dipolar interactions vanish and the dynamic assembly of the pair is driven only by HIs. Further, we provide a theoretical description based on the balance between dipolar forces and HIs that allow understanding the role of anisotropy on the collective dynamics. Investigating microscopic colloidal rotors and understanding their collective dynamics are important tasks for both fundamental reasons, but also to engineer similar fluid stirrers that can be readily used for precise microscale operations or as microrheological probes.

Published under an exclusive license by AIP Publishing. https://doi.org/10.1063/5.0076574
of the pair due to HIs and the particles set at a constant separation distance. By using fast video microscopy and particle tracking routines, we quantify such interactions and put forward an analytical model that can describe the relative separation distance as a function of the shape anisotropy of both rotors.

As magnetic microrotors, we use hematite microellipsoids characterized by a long (short) axis equal to $b = 1.8 \pm 0.11 \mu m$ ($a = 1.3 \pm 0.12 \mu m$), see Fig. 1(a). More details on the size distribution of these particles can be found in the supplementary material and, on the synthesis process, in previous works. The particles are dispersed in ultra-pure water (milliQ, Millipore) and functionalized with the surfactant sodium dodecyl sulfate (SDS), 0.12 g of SDS in 80 ml of high-deionized water. This concentration of SDS corresponds to 5.2 mM, thus lower than the critical micelle concentration in water (8.2 mM). This avoids the formation of micelles, which would decrease the adsorption of the surfactant to the particle surface. Finally, the pH of the resulting solution is adjusted to 8.5–9.5 by adding tetramethylammonium hydroxide (Sigma-Aldrich). As shown in Fig. 1(a), the particles are characterized by a small, permanent dipole moment $m = 2.3 \times 10^{-16}$ A m$^2$ oriented along the direction perpendicular to the particle long axis $b$. The samples are prepared by loading ~10 ml of the particle suspension in a chamber composed of a previously cleaned coverslip and a microscope slide, which are separated by two parafilm stripes and sealed with grease.

The external magnetic fields are generated by a set of magnetic coils arranged on the stage of an optical microscope (Eclipse Ni, Nikon). The latter is equipped with an oil immersion objective (100× NA = 1.3 or 40× NA = 0.75, Nikon) with an additional 0.45× TV lens before the camera to increase the field of view. Four coils are arranged by pairs perpendicular to each other and connected to a power amplifier (AMP-1800, Akiyama) driven by an arbitrary waveform generator (TGA12104, Aim-TTi). An additional fifth coil is placed below the sample stage connected to an independent power supply (TTi El 302) to apply a static magnetic field along an axis ($z$), perpendicular to the sample plane ($x$, $y$). Prior to the experiments, we use a teslameter (FM 205, Projekt Elektronik GmbH) to calibrate the field amplitude and its homogeneity at the particle location. Videos of the particles are recorded with a CMOS camera (AC640-56-Basler) working at 100 fps, and the particle positions are extracted using tracking routines. More details on the experimental set-up are given in the supplementary material.

Once in water, the hematite ellipsoids sediment toward the bottom of the experimental chamber due to density mismatch. The presence of the surfactant on the particles surface prevents them to irreversibly stick to the glass substrate due to attractive Van der Waals interactions. Above the glass surface, the hematite ellipsoids are quasi two-dimensionally confined, showing thermal fluctuations. From the particle positions, we extract the mean square displacement (MSD) and find a diffusive motion with a diffusion coefficient, $D = 0.073 \mu m^2 s^{-1}$; more details are given in the supplementary material.

We start to characterize the dynamics of the individual microrotors by using an in-plane rotating magnetic field with amplitude $B_0$ and angular frequency $\omega = B_0 (\cos(\omega t) \hat{x} - \sin(\omega t) \hat{y})$. Single particles spin with angular velocity $\omega_s$ due to the magnetic torque $\tau_m = |m| B \sin \phi$, which balances the viscous torque arising from the rotation in water, $\tau_v = -\zeta \beta$. Here, $\phi$ is the phase-angle between $B$ and $m$, see schematic in Fig. 1(a), $\beta = \omega t - \phi$, and $\zeta$ is the rotational friction of the ellipsoid in water. The torque balance equation in the overdamped limit leads to the dynamic equation, $\dot{\phi} = \omega - \omega_0 \sin \phi$, which predicts two dynamic regimes separated by a critical frequency $\omega_0$, as observed in Fig. 1(b). For $\omega < \omega_0$, the rotation is phase locked with the driving field, and $\phi$ is a constant, $\omega_0 = 2\omega_0$, being $z$ a pre-factor. In contrast, when $\omega > \omega_0$, the particle enters the asynchronous regime, where $\phi$ increases with $\omega_0$ and the average rotation of the rotor $\omega$ decreases as $\omega = 2\omega_0[1 - \sqrt{1 - (\omega/\omega_0)^2}]$. As shown in the inset of Fig. 1(b), the critical frequency scales linearly with the applied field, $\omega_0 = m B_0 / \zeta$. From the linear regression, we find that $\zeta = (1.61 \pm 0.13) \times 10^{-22}$ N s m, similar to previous values obtained for paramagnetic ellipsoids.

The complex dynamics of pair of microrotors result from the balance between magnetic dipolar forces and HIs excited by their rotary movement. We expect stronger HIs than spherical rotors due to the anisotropic shape, which will create a stronger vortex flow. To investigate such interactions, we choose to apply a precessing magnetic field, which allows tuning the dipolar forces from attractive to repulsive. Such a field reads as

$$ B = B_0 [\cos \vartheta + \sin \vartheta (\cos (\omega t) \hat{x} - \sin (\omega t) \hat{y})].$$

where $\vartheta$ is the precession angle and $B_0 = B \sin \vartheta$ is the in-plane component, Fig. 2(a). The time average dipolar forces between two equal point dipoles with magnetic moments $m$ on the same plane and at distance $r$ read as
Decreasing further... they are dragged by the flow field generated by the neighboring rotor. Small, and the particles in the pair circulate one around the other as...}

...effectively, we find that once formed at \( \vartheta = 55.26 \), the mean angular velocity of the pair decreases algebraically with the distance as \( \omega_p \sim \Delta r^{-3} \), Fig. 3. From the analysis of different trajectories, we can extract the magnetic torque applied to each particle as \( \tau_m = (1.2 \pm 0.3) \times 10^{-19} \text{N m T}^{-1} \), which is in good agreement with that calculated directly using the magnetic torque equation.

The interaction between the rotating hematite particles is a complex interplay between magnetic dipolar forces and the shape anisotropy, which induces HIs along the separation of the particle centers, \( \Delta r \). The mechanism of HIs between the anisotropic rotors can be explained by considering the periodic part of the dipolar interactions and the periodic flux in the region between the ellipsoids. Fluid is expelled from such region when the long axis rotates toward the line of separation, while it reenters the region when the long axis rotates toward a transversal orientation. Both situations repeat twice during a full field cycle, and the flux causes periodic (frequency \( 2 \omega \)) oscillations of the pressure. The mobility matrix that relates force and torque to speed and angular velocity for our system can be written as

\[
\begin{pmatrix}
\Delta \tau \\
\varphi_1 \\
\varphi_2 \\
\end{pmatrix}
= M
\begin{pmatrix}
F_{12} \\
\tau \\
\tau_1 \\
\tau_2 \\
\end{pmatrix},
\]

where \( F_{12} = \frac{\mu_0 \mu_r^2}{4 \pi} \frac{P_2(\cos \vartheta)}{2 \pi} \left( \frac{\cos \varphi - \cos \varphi_1}{\cos \varphi - \cos \varphi_2} \right) \) denotes the dipolar interaction force between the rotors, \( \tau, \tau_1, \) and \( \tau_2 \) are the total and individual magnetic torques, and \( \varphi, \varphi_1, \) and \( \varphi_2 \) are the corresponding phase-lag angles, see inset in Fig. 4. Further, \( M \) is the mobility tensor that depends on the conformations \( (\Delta r, \varphi_1 - \varphi, \varphi_2 - \varphi, \vartheta_1, \vartheta_2) \) of the rotors. We approximate the leading dependencies of the symmetric mobility matrix on the conformation by \( M_{11} = 1/6 \pi \eta a, M_{12} = 0, M_{13} = a \epsilon^2 \sin^2 \vartheta \sin(2[\varphi_1 - \varphi])/6 \pi \eta a^3, M_{20} = 1/8 \pi \eta a^2, \) and \( M_{31} = 1/3 \pi \eta a^2 \), where \( a \) is the mean radius of the rotor, \( \epsilon \) is the eccentricity of the rotor shape, and we assume that the rotor orientation to be locked to the magnetic field \( (\vartheta_1 = \vartheta, \varphi_1 = \omega t) \). The two rotors are circling around their centers of mass with frequency \( \omega_p \) and all other...
non-diagonal elements of the mobility matrix are assumed to vanish. The mobility coefficient, \(M_\perp\), arises due to the fact that liquid needs to flow out or needs to enter the region between the two rotors when the particle orientation changes from one major axis of the hematite particle aligned with the separation to the other. Hence, we find the following differential equation:

\[
\dot{\omega} = \frac{P_3 (\cos \theta) + \cos (2\theta)}{2} + \frac{\alpha^2}{\mu} \eta \omega, \tag{4}
\]

where we use adimensional units with \(\tilde{r} = [\omega - \omega_0] r, \tilde{\omega} = \Delta \tilde{r}/l\) with \(l = \sqrt{72\pi^2 \eta (\omega - \omega_0)/a \mu m^2 P_3 (\cos \theta)}\) and \(\alpha = 2a^2 \epsilon \sin \theta_1/\sqrt{3}l\).

A numerical solution of Eq. (4) yields an oscillatory behavior of the separation on short time scales. On longer time scales, the equation predicts a separation for small precession angle and an attraction for larger precession angles. Figure 4 shows a phase diagram of the long time behavior as a function of the anisotropy parameter and the precession angle. The line separating attractive from repulsive interaction starts at the magic angle \(\theta_m = 54.7^\circ\) and zero anisotropy. Then, it monotonously increases with the parameter \(\alpha\), thus the shape anisotropy of the rotors. This implies that increasing the particle anisotropy raises the strength of HIs producing a shift of the magic angle toward higher values.

In conclusion, we have investigated the non-equilibrium dynamics of pair of interacting magnetic microrotors driven by an external precessing magnetic field. We show that by tuning the precession angle, one can control the assembly of the pair and reduce the effect of magnetic dipolar interactions in favor of hydrodynamic ones. The controlled stirring of anisotropic, elongated particles in viscous fluids may find several applications apart from that of fluid mixers. For example, magnetic nanorods have been used to measure the rheological properties of monolayers at the water/air interface. When compared to the hematite particles, both type of particles should generate similar vortical flow when torque by a rotating magnetic field. However, the smaller lateral size of the nanorods will make more difficult their visualization under optical microscopy. In comparison with macroscopic rheometers, the small size of these rheological probes increases the sensitivity of the measurement while reducing the amount of material to be investigated. In this context, most of the studies so far have investigated the dynamics of single rotors in viscoelastic fluids. Thus, a potential future direction of this work could be to test how the dynamics of pair of rotors are altered by a complex non-Newtonian medium.

See the supplementary material for additional details on the experimental system, the measurements of the long and short axes of the hematite ellipsoids, and the diffusive properties of these particles.

We thank Helena Massana-Cid for experimental advice and Gaspard Junot for providing SEM images of the particles. This work received funding from the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation programme (Grant Agreement No. 811234). P.T. acknowledges support from MCIU (No. PID2019-108842GB-C21) and the Program ICREA “Acadèmia.”

AUTHOR DECLARATIONS

Conflict of Interest

The authors declare no conflict of interest.

DATA AVAILABILITY

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

REFERENCES