Research articles

Magnetic and hydrodynamic torques: Dynamics of superparamagnetic bead doublets

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A B S T R A C T

Rotating chains of magnetic microparticles have many applications in lab-on-a-chip technologies. The simplest such chain is the fluid-borne doublet, where two beads are in close contact, but remain unattached, allowing each bead to freely rotate. These beads typically have two components contributing to their net magnetic moment: (i) a superparamagnetic moment and (ii) a field-independent permanent moment. In a rotating magnetic field, there are magnetic torques that separately rotate the doublet and its constituent beads as well as a hydrodynamic torque from the bead-doublet coupling. This study investigates, through experiments and simulations, the dynamics of field-driven doublets. New dynamics were found for the case where the dominant torque stems from the hydrodynamic coupling.

1. Introduction

Actuating magnetic micro- and nano-particles has become important to the development of several lab-on-a-chip technologies because of their translation and rotation in applied external fields, capability for multiplexing, bio-compatibility, and cellular-relevant length scale. They have been investigated in many systems such as ferrofluids [1–3], magnetic chains [4–10], magnetotactic bacteria [11,12] and clusters [13,14]. The ensuing applications are broad. They include fluid mixers [2,4,8,9,15–18], micro-swimmers [19,20], sorting [21–25], drug delivery [20,26,27], micro-surgery [20,28], cell detection [29,30], as well as stiffness-, bio- and rheological-sensing [10,31–40]. To optimize these applications, the interplay between the hydrodynamic and magnetic forces affecting their dynamics must be understood. One of the simplest constructs that displays this interplay is the detached doublet consisting of two adjacent, but unattached, fluid-borne magnetic beads rotating in a magnetic field.

Magnetic microbeads that are typically used in lab-on-a-chip devices are created by embedding magnetic nanoparticles within a polymer matrix [41]. Two components contribute to their net magnetic moment: (i) a superparamagnetic Néel dipole \( m_H \) that rapidly aligns with an external field \( H \) due to the fast stochastic reorientation of the nanoparticle magnetic domains and (ii) a Brownian field-independent permanent moment \( m_P \) arising from the slow relaxation of the magnetization of the larger nanoparticles within the bead [42,33]. Studies have investigated how magnetic torques arising from the interaction between \( m_P \) and \( H \) (dipole-field interaction) affects the rotation of individual beads [33] as well as chemically attached [43] and detached [44] doublets. Detached doublets present an interesting case where the dipole-field torque directly rotates the individual beads in the presence of a time varying magnetic field and consequently affects the doublet’s rotation through the hydrodynamic coupling of the beads’ and doublet’s rotation.

Detached magnetic particles have previously been studied without including the hydrodynamic bead-doublet coupling [36,10]. With only the magnetic torque, the doublets rotate at the same rate as the external field up to some critical frequency, above which the doublet on average begins to slow down. Coughlan et. al. [44] found that the hydrodynamic bead-doublet coupling had a small perturbation on the rotation rate of detached doublets. The perturbation was small in this case because of the relative weakness of \( m_P \) and the dipole-field torque on their beads.

The present study finds new dynamics in the rotation rate of the doublets when the hydrodynamic bead-doublet coupling is the strongest torque. This previously unreported response arises from separate critical frequencies caused by the moments \( m_H \) and \( m_P \). Simulations reveal that the hydrodynamic torque \( \tau_H^{dtd} \) on the doublet is linear with respect to the rotation rate of the doublet \( \omega_D \) and of the individual beads \( \omega_B \), \( \tau_H^{dtd} = y_H^{dtd} \omega_D + y_B^{dtd} \omega_B \). The coefficients \( y_H^{dtd} \) and \( y_B^{dtd} \) represent the strength of the drag torque on the doublet and the hydrodynamic bead-doublet coupling respectively and are determined from the simulations. Analytical models based on these results compare favorably with measurements of the average doublet rotation rate \( \omega_D \) for beads with different strengths of superparamagnetic \( m_H \) and ferromagnetic \( m_P \) moments. This enables the permanent moment \( m_P \) of the beads to be
non-invasively determined from the models by measuring $\sigma_0$ — an advantage over conventional techniques for measuring $m_0$ that require either non-symmetric particles [45] or spherical beads attached with small particles [31,46], fluorophores [47] or coatings [44,48]. Furthermore, these findings provide a framework to leverage both the magnetic and hydrodynamic torques on rotating doublets. For example, our analysis reveals that the hydrodynamic bead-doublet coupling can increase the maximum torque and rotation rate of a doublet which is beneficial for many lab-on-a-chip applications.

2. Materials and methods

A 3-axis electromagnet provides time-varying magnetic fields for rotating the particles. The platform (Fig. 1) includes four orthogonal electromagnets (Magnetec Corp OP-1212) and a custom-made solenoid that dynamically creates magnetic fields in the $xy$- and $z$-directions respectively. The fields, which are rotated in the $xy$-plane at constant frequencies ($\sim$0.1–10 Hz), torque the magnetic beads as they are viewed under an optical microscope (Leica DM2500 MH) with a 40x objective. The bead dynamics are imaged with a high speed camera (Phantom Miro M120) at 100 frames per second. The orientation of the doublet in the videos is tracked by extracting the doublet from the image through thresholding and then measuring the orientation of its longest axis.

Invitrogen’s Dynabeads M-270 Carboxylic Acid (2.8 $\mu$m diameter) [41] and Bang’s Lab COMPEL Fluorescent Magnetic Carboxylic Acid beads (2.8 $\mu$m diameter) are utilized because of their comparable geometries and surface chemistries while displaying definitive differences in magnetic moments. Larger COMPEL beads (7.9 $\mu$m diameter) were separately used to visualize individual bead orientation during rotation.

The beads are suspended in a 0.06 mm deep fluid well made from double-sided tape sandwiched between a glass slide (Fisher) and a cover slip (Thermo-Scientific). The tape binds the device together and prevents leakage. The glass slide is coated with polyethylene glycol (PEG) or casein to reduce non-specific binding of beads to the substrate. The superparamagnetic beads are also diluted in 0.1% Triton solution (Sigma–Aldrich) to decrease the amount of non-specific binding.

The average doublet rotation rate $\omega_D$ is measured with a given doublet for the entire field frequency range. Once the field rotations are initiated and transient bead movement subsides, videos are recorded for 60–90 s to include multiple (>6) rotations of the doublet. Doublets were considered permanently attached if an applied $z$-field could not separate the beads. The attachment was caused by non-specific binding between the beads’ surfaces. The doublets do not bind to the surface and stay oriented in the $xy$-plane for the entire frequency range. Between measurements, the doublets were magnetically rolled to a new location [49] to inhibit non-specific binding to the surface, and a $z$-field is reapplied to confirm the beads remain attached or detached.

To assess the bead-doublet hydrodynamic coupling, steady-state Navier-Stokes equations were numerically solved. This simulation includes interactions between the beads as well as near wall effects from the substrate. Translational and rotational Brownian fluctuations were neglected in the simulations because they did not significantly affect the doublet rotation at these bead sizes. The two beads were assumed to have a 100 nm gap between their closest points and a 50 nm gap between the beads and the flat surface [50,51]. The simulation geometry enclosed the beads in a large cylinder with a diameter $\sim 40\times$ the bead diameter (Fig. 2a) in order to minimize the wall-induced anisotropic effects on the steady state solution, and it included a “refinement box” around the beads that was more finely discretized to increase the numerical accuracy of the simulation in that region. A no-slip velocity boundary (Dirichlet condition) and a vanishing pressure gradient (Neumann condition) were imposed on all surfaces including the walls of the cylinder.

A finite volume-based iterative steady-state solver [52] was written using the Semi-Implicit Method for Pressure-Linked Equations (SIMPLE) algorithm. The finite volume method is a discretization scheme that is commonly used to solve the incompressible Navier-Stokes equations:

$$\rho \left( \frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right) = -\nabla p + \mu \nabla^2 \mathbf{v},$$

where $\mu$ is the kinematic viscosity, $\rho$ the density, $p$ the pressure, and $\mathbf{v}$ the velocity of the fluid. Together with the incompressible continuity equation $\nabla \cdot \mathbf{v} = 0$.

Fig. 2. (a) Schematic of the fluid domain and the bead geometry simulated. Navier–Stokes equations were solved at higher resolution in the refinement box near the beads. (b) Fluid flow generated by two beads spinning at $\omega_B = 5 \text{ rad s}^{-1}$ ($\omega_D = 0$) is visualized by the blue trace of several tracer particles, initiated at random locations near the beads. Narrow white arrows mark the tracer particles’ direction of motion. Dashed arrows show the direction of rotation of the beads, while wide arrows indicate the hydrodynamic torque on the doublet due to its coupling to $\omega_B$. 
\[ \nabla \cdot \mathbf{v} = 0, \] the Navier-Stokes equations can be iteratively solved for a discretized fluid volume using the SIMPLE algorithm. Our model did not make any assumptions about the linearity of the low Reynolds number system. Instead, the full Navier-Stokes equations were solved. These solutions are used to determine the net force on a bead by numerically integrating the normal pressure and the shear stress contributions. The resulting net forces (Fig. 2b) on the two beads produce the hydrodynamic torque on the doublet and the beads about their centers of mass.

3. Theory

The magnetic moment for our beads is composed of a superparamagnetic Néel moment \( \mu_{H} \) and a permanent Brownian moment \( \mu_{B} \) oriented at \( \theta_{B} \) (Fig. 3a). The net bead magnetic moment \( \vec{m} = \vec{m}_{H} + \vec{m}_{B} \) is oriented at \( \theta_{H} \). Both beads are assumed to have the same magnetic moments \( \mu_{H} \) and \( \mu_{B} \). When two beads lie adjacent to one another, they form a doublet with a long axis oriented at \( \theta_{D} \). For detached doublets (Fig. 3b), the individual beads rotate at angular frequency \( \omega_{H} \) which may differ from the doublet frequency \( \omega_{D} \). For attached doublets the beads must rotate synchronously such that \( \omega_{H} = \omega_{D} \).

The next sections summarize the three torques encountered — the dipole-field interaction, the dipole-dipole interaction, and the bead-doublet hydrodynamic coupling — and the doublet’s dynamics when these torques combine to rotate detached and attached doublets.

3.1. Dipole-field and dipole-dipole torques

In an external field \( H \), \( \mu_{B} \) experiences the dipole-field torque \( \tau_{H}^{D} \) given by [45]:

\[
\tau_{H}^{D} = \vec{m} \times \mu_{B} \mathbf{H},
\]

(1)

where \( \mu_{B} \) is the permeability of free space. For detached doublets, \( \tau_{H}^{D} \) acts on the individual beads and not on the doublet, but for attached doublets, the torque acts on the doublet.

The dipole-dipole torque \( \tau_{D}^{D} \) for two beads each with net magnetic moments \( \vec{m} = \vec{m}_{H} + \vec{m}_{B} \) separated by a distance \( 2r \) (twice the bead radius) is given by [43]:

\[
\tau_{D}^{D} = \frac{3 \mu_{0} m_{H}^{2}}{4 \pi (2r)^{3}} \sin(2(\theta_{H} - \theta_{B})),
\]

(2)

which causes a doublet, attached or detached, to rotate around its center.

![Fig. 3. (a) The doublet angle \( \theta_{D} \) is oriented along the direction connecting the centers of the beads. Induced magnetic moment \( \mu_{H} \) is in direction of the external field which is oriented at \( \theta_{B} \). The permanent magnetic moment \( \mu_{B} \) is fixed along some direction oriented at \( \theta_{B} \). The net magnetization \( m \) is oriented at \( \theta_{H} \) which is between \( \theta_{B} \) and \( \theta_{D} \). (b) For detached doublets, the beads rotate independently from the doublet. The doublet rotates at frequency \( \omega_{D} \) around the center of the two beads. Whereas the individual beads rotate around their center at frequency \( \omega_{H} \). For attached doublets, the beads and doublet rotate at the same frequency \( \omega_{H} = \omega_{D} \).](image)

3.2. Bead-doublet hydrodynamic torque

For detached doublets, while the dipole-field torque (Eq. (1)) acts on the beads and not directly on the doublet, the rotation of the individual beads creates an additional hydrodynamic torque on the doublet. This hydrodynamic bead-doublet coupling is the result of a rotating particle experiencing more drag on the side closer to the adjacent bead than its other sides, essentially causing the particle to “roll” around the adjacent bead and the doublet to rotate.

In low-Reynolds number dynamics, the forces and torques on a body are linearly related to its velocity and angular velocity respectively by drag coefficients that depend on the physical geometry [44,54]. As a result, a linear relationship between the hydrodynamic torques \( \tau_{H}^{Hyd} \) and \( \tau_{D}^{Hyd} \) and \( \omega_{H} \) and \( \omega_{D} \) is expected and confirmed by the simulations (Fig. 4). Thus, for a doublet

\[
\tau_{D}^{Hyd} = \gamma_{D}^{H} \omega_{D} - \gamma_{D}^{B} \omega_{B},
\]

where \( \gamma_{D}^{H} = 0.34 \text{ pN} \text{ s rad}^{-1} \) and \( \gamma_{D}^{B} = 6.60 \times 10^{-2} \text{ pN s rad}^{-1} \) are the doublet drag coefficients from the doublet (D) and bead (B) rotation respectively, \( \gamma_{D}^{H} \omega_{D} \) is the hydrodynamic torque resisting the rotation of the doublet, \( \gamma_{D}^{B} \omega_{B} \) represents an additional hydrodynamic torque on the doublet arising from the hydrodynamic bead-doublet coupling. Similarly for the individual rotating beads within the doublet (Fig. 4b),

\[
\tau_{B}^{Hyd} = \gamma_{B}^{H} \omega_{B} - \gamma_{B}^{D} \omega_{D},
\]

where \( \gamma_{B}^{H} = 9.37 \times 10^{-2} \text{ pN s rad}^{-1} \) and \( \gamma_{B}^{D} = 3.32 \times 10^{-2} \text{ pN s rad}^{-1} \) are the corresponding drag coefficients for the bead. Our simulations included interactions between the beads as well as near wall effects with the substrate which are reflected in these values for the drag coefficients. Changes in the simulated bead-substrate distance from 50 nm to 150 nm yield negligible (<3%) changes in all the drag coefficients except \( \gamma_{D}^{B} \) which would decrease by 16%.

The drag coefficients resulting from our simulations align with expectations based on analytical results for beads well separated from the surface. For two isolated and decoupled beads (radius \( r = 1.4 \mu m \)) in water (dynamic viscosity \( \eta = 8.9 \times 10^{-3} \text{ Pa s} \)) where each bead is translating around a fixed Eulerian point at the bead’s edge, \( \gamma_{D} \) may be analytically approximated by Stokes’ laws as \( \gamma_{D}^{H} = 12 \pi \eta r^{2} \approx 0.09 \text{ pN s rad}^{-1} \). Because of the additional drag on the beads from being near the surface and each other, the actual value of \( \gamma_{D}^{H} \) for non-isolated beads is expected to be larger but within the same order of magnitude which is confirmed by the results of the simulations which yielded \( \gamma_{D}^{H} = 0.34 \text{ pN s rad}^{-1} \). Similarly, our simulated value for \( \gamma_{B}^{H} (9.37 \times 10^{-2} \text{ pN s rad}^{-1}) \) is greater than but within an order of magnitude of the analytical value via Stokes’ Law for a rotating isolated bead \( \gamma_{B}^{H} = 8 \pi \eta r^{2} \approx 6.14 \times 10^{-2} \text{ pN s rad}^{-1} \) due to the near wall effects. While \( \gamma_{B}^{D} (6.60 \times 10^{-2} \text{ pN s rad}^{-1}) \) is difficult to analytically approximate, the effect of the flow field from the individual bead on the rotation of the doublet is expected to be smaller than the drag on the larger doublet \( (\gamma_{D}^{B} = 0.34 \text{ pN s rad}^{-1}) \) as it moves through the fluid.

3.3. Detached doublets

For detached doublets, the dipole-dipole interaction torques the doublet into the direction of the field. At low Reynolds’ number, the net torque equals zero, and thus, the magnetic torque is given by

\[
\tau_{D}^{D} = \frac{3 \mu_{0} m_{H}^{2}}{4 \pi (2r)^{3}} \sin(2(\theta_{H} - \theta_{B})),
\]

(3a)

which yields the rotation rate

\[
\omega_{D} = (1-\varepsilon_{0})\omega_{H}\sin(2(\theta_{B}-\theta_{D}))+\varepsilon_{0}\omega_{B},
\]

(3b)

where \( \varepsilon_{0} = \frac{\mu_{0} m_{H}^{2}}{4 \pi (2r)^{3}} \) and the constant \( \varepsilon_{0} = \frac{\gamma_{D}^{H}}{\gamma_{D}^{B}} \approx 0.20 \) is a measure of the relative strength of the hydrodynamic bead-doublet coupling. At low frequencies up to the doublet’s critical frequency \( \omega_{D}^{c} \), the doublet...
will rotate synchronously with the field \( \omega_B = \omega_H \). At frequencies above \( \omega_B^\ast \), the doublet will rotate slower than \( \omega_H \) because drag overcomes the magnetic torque. Likewise, the beads will rotate synchronously with \( \omega_B \) up to the bead’s critical frequency \( \omega_B^\ast \) and at higher frequencies will also desynchronize with \( \omega_H \).

\( \ast \omega_B^\ast \) is dependent on the strength of the dipole-dipole torque and the hydrodynamic bead-doublet coupling and reaches its maximum value \( \gamma_B^D \) when there is maximal torque from the beads’ rotation (i.e. \( \omega_B = \omega_H \)) at \( \omega_B^\ast \) which occurs when \( \omega_B^\ast \geq \omega_B \). When the bead-doublet coupling is strong, the maximum average doublet rotation rate \( \Omega_D^{\text{MAX}} \) may be larger than \( \Omega_H \), even though the doublet is not rotating synchronously with \( H \).

Similarly, the magnetic torque on the individual beads is given by

\[
\tau_B^M = \mu_c m_p H \sin(\theta_H - \theta_B) = \gamma_B^B \omega_B^2 - \gamma_B^D \omega_B^2.
\]

and the bead rotation rate

\[
\omega_B = (1-c_B) \gamma_B^B (\theta_H - \theta_B) + c_B \omega_B.
\]

where \( c_B = \frac{\mu_c m_p H}{\gamma_B^D - \gamma_B^B} \) and \( \gamma_B^D / \gamma_B^B \approx 0.35 \). \( \omega_B^* \) is dependent on the dipole-
field torque and the hydrodynamic bead-doublet coupling from the rotating doublet and reaches its maximum frequency \( \omega_D \) when the hydrodynamic bead-doublet coupling is largest (\( \omega_D = \omega_H \)) at \( \omega_D^\ast \) which occurs when \( \omega_H^\ast \leq \omega_D^\ast \).

Because of the interdependence of the bead and doublet dynamics, \( \omega_H^\ast \) and \( \omega_D^\ast \) fall into two cases depending on their relative critical frequency values (\( \omega_H^\ast \) and \( \omega_D^\ast \)) or equivalently on whether the dimensionless parameter \( \frac{\omega_B}{\omega_H} = \frac{\omega_B}{m_H} \left( \frac{\omega_B^2 - \omega_H^2}{\omega_H^2} \right)^{1/2} \) is greater than or less than one. When \( m \approx m_H = \frac{\gamma V_H}{\omega_H} \),

### 3.3.1. Case I (\( \omega_H^\ast > \omega_D^\ast \))

As illustrated in Figs. 5a and b, Eqs. (3) and (4) lead to

\[
\sigma_H = \begin{cases} 
\omega_H & \text{if } \omega_H < \omega_H^\ast \\
\omega_H - (1 - c_D) \sqrt{\omega_H^2 - \omega_B^2} & \text{if } \omega_H^\ast < \omega_H < \omega_B^\ast \\
\omega_B & \text{if } \omega_B^\ast < \omega_H 
\end{cases}
\]

\[
\sigma_D = \begin{cases} 
\omega_D & \text{if } \omega_D < \omega_D^\ast \\
\omega_D - (1 - c_D) \sqrt{\omega_D^2 - \omega_B^2} & \text{if } \omega_D^\ast < \omega_D < \omega_B^\ast \\
\omega_B & \text{if } \omega_B^\ast < \omega_D 
\end{cases}
\]

In this case, \( \omega_H = \omega_B + k_H \sqrt{(\omega_B^2 - \omega_H^2)^2} \) where \( k_H = \omega_B (1 - c_D) \). The values for \( m_H \) and \( m_B \) for the beads can be determined from the two critical frequencies \( \omega_H^\ast \) and \( \omega_B^\ast \). The doublet and individual bead rotations fall into three distinct regions: (1) \( \omega_D < \omega_H^\ast \), (2) \( \omega_H^\ast < \omega_D < \omega_B^\ast \), and (3) \( \omega_B^\ast < \omega_D \).

When \( \omega_D < \omega_B^\ast \), both the individual beads and the doublet rotate at \( \omega_H^\ast \). The doublet does not align with \( H \) but trails it by a fixed angle that increases with \( \omega_D \) until the dipole-dipole torque reaches its maximum at \( \omega_H^\ast = \omega_B^\ast \).

When \( \omega_B^\ast < \omega_D < \omega_H^\ast \), the individual beads continue to rotate at \( \omega_H^\ast \). Whereas, \( \sigma_H \) slows when \( \omega_H^\ast \) just exceeds \( \omega_H^\ast \). The doublet cannot rotate fast enough to keep up with \( H \) so it increasingly trails further behind it. When \( H \) is too far ahead of the doublet, the doublet will slow down and rotate backwards to realign with it. This process creates a periodic oscillatory motion (Supplementary Video 2). While the dipole-dipole torque \( \tau_B^\ast \) decreases with \( \omega_B^\ast \) on average, the hydrodynamic bead-doublet coupling continues to increase linearly because the beads are still rotating at \( \omega_H^\ast \) as evident in Fig. 5b. Once the hydrodynamic bead-doublet coupling dominates, \( \sigma_D \) increases linearly with \( \omega_D^\ast \) such that \( \sigma_D \approx \omega_D^\ast \omega_D^\ast \approx 0.2 \omega_H^\ast \). If \( \omega_H^\ast \) is sufficiently large, the hydrodynamic bead-doublet coupling can rotate the doublet faster than the dipole-dipole coupling can and rotate the doublet fully back to \( \omega_H^\ast \).

When \( \omega_H^\ast > \omega_B^\ast \), the beads can no longer rotate at \( \omega_H^\ast \), decreasing the hydrodynamic bead-doublet coupling and causing \( \sigma_D \) and \( \sigma_B \) to decrease (Fig. 5c) as both the beads and doublet start to exhibit periodic oscillatory motion.

### 3.3.2. Case II (\( \omega_H^\ast < \omega_D^\ast \))

As illustrated in Figs. 5c and d, from Eqs. (3) and (4)

\[
\sigma_H = \begin{cases} 
\omega_H & \text{if } \omega_H < \omega_H^\ast \\
\omega_H - (1 - c_D) \sqrt{\omega_H^2 - \omega_B^2} & \text{if } \omega_H^\ast < \omega_H < \omega_B^\ast \\
\omega_B & \text{if } \omega_B^\ast < \omega_H 
\end{cases}
\]

\[
\sigma_D = \begin{cases} 
\omega_D & \text{if } \omega_D < \omega_D^\ast \\
\omega_D - (1 - c_D) \sqrt{\omega_D^2 - \omega_B^2} & \text{if } \omega_D^\ast < \omega_D < \omega_B^\ast \\
\omega_B & \text{if } \omega_B^\ast < \omega_D 
\end{cases}
\]

where \( \omega_D = \omega_B^\ast + k_B \sqrt{(\omega_B^2 - \omega_D^2)^2} \) for \( k_B = \omega_B (1 - c_D) \).

When \( \omega_D < \omega_H^\ast \), the doublet rotates at \( \omega_H^\ast \). At \( \omega_H^\ast \), the individual beads are already rotating slower than \( \omega_H^\ast \) and contribute less hydrodynamic torque. As such, the critical frequency \( \omega_H^\ast \) is up to 14% below \( \omega_D^\ast \).

The lower limit of the critical frequency (\( \omega_B^\ast \approx 0.86 \omega_D^\ast \)) occurs if there is no dipole-field torque (i.e. \( \omega_B^\ast = 0 \)) and the upper limit (\( \omega_D^\ast = \omega_B^\ast \)) occurs if the beads rotate at \( \omega_B^\ast \) when \( \omega_D^\ast \) is reached (i.e. \( \omega_D^\ast = \omega_B^\ast \)).

When \( \omega_B^\ast > \omega_D^\ast \), both \( \sigma_H \) and \( \sigma_B \) decrease (Fig. 5d) as both the beads and doublet exhibit periodic oscillatory motion.

### 3.4. Attached doublet

Attached doublets are similar to the detached case except the beads are rigidly attached (\( \omega_B = \omega_D \)). Hence, they are affected by both the magnetic torques \( \tau_B^M \) and \( \tau_D^M \) and the hydrodynamic torques \( \tau_B^H \) and \( \tau_D^H \) (43) yielding the hydrodynamic drag coefficient \( \gamma_{AD} = \gamma_B^H - \gamma_D^H + 2(\gamma_B^M + \gamma_D^M) \).

The doublet rotation rate becomes

\[
\omega_D = \frac{1}{\gamma_{AD}} \left( (\gamma_B^M - \gamma_D^M) \omega_B \sin(2(\theta_H - \theta_B)) + 2(\gamma_B^M - \gamma_D^M) \omega_B \sin(\theta_H - \theta_B) \right)
\]

showing that \( \omega_B \) not only depends on \( \sigma_B \) and \( \sigma_D \), but also on the angle \( \phi \) (= \( \theta_B - \theta_D \)) between the permanent moment and the doublet axis.

### 3.5. Maximum rotation rate

Many lab-on-a-chip applications would benefit from an increased maximum torque or rotation rate. The maximum average doublet rotation rate \( \sigma_B^{MAX} \) in turn is dependent on the ratio of the critical frequencies and whether the beads are attached or detached (Fig. 6).

For a detached doublet, based on Eqs. (5) and (6),

\[
\sigma_B^{MAX} = \begin{cases} 
\omega_B^0 & \text{if } \omega_B < \omega_B^0 \\
\omega_B^0 - (1 - c_D) \sqrt{(\sigma_B^0)^2 - (\sigma_D^0)^2} & \text{if } \omega_B^0 < \omega_B < 4.63 \omega_B^0 \\
(\sigma_B^0)^2 - (\sigma_D^0)^2 & \text{if } \omega_B^0 \geq 4.63 \omega_B^0 
\end{cases}
\]

where 4.63 \( \omega_B^0 \) approximates \( 1 + (1 - c_D^2) \). For an attached doublet, the maximum rotation rate is also depen-
dent on the fixed angle \( \phi = \theta_0 - \theta_L \) (Eq. (7)). Both magnetic and hydrodynamic torques are maximized when \( \phi = \pi/4 \) which yields

\[
\Omega_{D}\text{MAX} = \frac{(y_D^2 - y_D^0 + 2y_D^0 y_D^0) \tau_B}{y_D^0 - y_D^0 + 2y_D^0 y_D^0 - 2y_D^0}.
\]

If the beads attach while in an external magnetic field, \( \phi = 0 \).

Whether doublets move faster while attached or detached depends on whether drag or magnetic torque increases more with attachment. When doublets become attached, the magnetic torque increases from the addition of the dipole-field torque \( \tau_D^H \) on the doublet, but the drag also increases from the addition of the hydrodynamic torque \( \tau_D^{\text{Hd}} \) from the beads. The faster state is generally dependent on the ratio of \( \omega_B^2 \) to \( \omega_D^2 \) (Fig. 6), except when \( \omega_B^2 \approx \omega_D^2 \), then it is highly dependent on \( \phi \). Otherwise, when \( \omega_B^2 > \omega_D^2 \), attached doublets have a faster maximum rotation rate than their detached counterparts; for \( \omega_B^2 < \omega_D^2 \), the doublets can rotate faster when detached.

4. Experimental results and discussion

The following sections will compare the measured \( \omega_D \) for detached Dynabead and COMPEL doublets with the models predicted above. It is preceded with a discussion justifying that the two types of beads investigated have a permanent moment \( m_P \), and is concluded with a comparison of \( \Omega_{D}\text{MAX} \) for attached and detached doublets.

4.1. Identification of permanent moment \( m_P \)

If the magnetic moment of a bead does not immediately align with \( H \), it may be the result of a permanent moment [13,16,29–31,35] or the presence of a magnetization [33,46]. An easy axis is caused by magnetic anisotropy resulting from either non-uniformly distributed magnetic nanoparticles or nanoparticles that exhibit shape anisotropy, and is characterized by a magnetization that is linearly dependent on \( H \) and can flip orientation along its axis [33,46].

A permanent moment results from large nanoparticles with a Néel relaxation time that is long compared with \( 2\pi/\omega_H \) and has a fixed magnitude. The orientation of the permanent moment is fixed [33,44,45] at low fields but can be re-magnetized [47] at high fields where \( H \) can overcome the coercive energy and flip the magnetization’s direction. Single bead studies [33,47] have demonstrated that 2.8 \( \mu m \) Dynabeads have a permanent moment and not an easy axis.

In order to identify the type of magnetization for our beads, attached doublets were studied, because both the dipole-field and dipole-dipole torque would directly affect the doublet rotation. When \( \omega_H \) is above the critical frequency, \( \omega_D \) will start to slow down and exhibit oscillatory motion. The doublet will oscillate at a frequency that is dependent on the type of magnetization that underlies its rotation [33,46,47]. For a permanent moment \( m_P \) fixed along a specific direction, the period of the oscillation is the time required for the field to move completely around the doublet yielding the oscillation frequency \( \Omega_1 = \omega_H - \omega_D \). Whereas for the dipole-dipole interaction \( \tau_D^D \) (Eq. (2)), an easy axis, or a permanent moment at high \( H \) that can flip directions, the oscillation period is the time required for the field to go halfway around the doublet yielding the oscillation frequency \( \Omega_2 = 2(\omega_H - \omega_D) \).

A doublet’s oscillation frequency can be directly determined from the power spectrum of \( \theta_L(t) \) (Fig. 7). Experimentally, the power spectrum of the COMPEL beads display much stronger frequency peaks at \( \Omega_1 \) than \( \Omega_2 \) (Fig. 7a) whereas the Dynabeads have pronounced peaks at both \( \Omega_1 \) and \( \Omega_2 \) (Fig. 7b). The presence of \( \Omega_1 \) in both COMPEL and Dynabeads confirms a fixed direction, permanent moment in both cases. Whereas, the relative weakness in \( \Omega_2 \) for COMPEL beads compared with Dynabeads, suggests that the dipole-dipole interaction and consequently \( m \) is weaker for COMPEL beads than Dynabeads at \( H = 20 \) Oe.

4.2. Detached COMPEL beads

As shown in Fig. 8a, the typical frequency response of a detached COMPEL doublet agrees with Eq. (5) allowing COMPEL beads to be used to explore bead dynamics when \( \omega_B^2 > \omega_D^2 \).

The time dependence of \( \theta_L \) is also measured (Fig. 8b). As expected, the doublet rotates at \( \omega_B \) when \( \omega_B < \omega_D \), and undergoes periodic oscillatory motion when \( \omega_B < \omega_B < \omega_D \). These oscillations were found to decrease in magnitude at higher field frequencies when the bead-doublet coupling provides greater torque than the dipole-dipole interaction. In the case where \( \omega_H > \omega_D \), the doublet was often found to rotate consistently at a frequency below \( \omega_H \) but not oscillate. The bead-doublet coupling does not appear to cause any periodic oscillations in \( \theta_L \), even though the individual beads are expected to oscillate.

When \( \omega_H > \omega_D \), we expect the decrease in \( \omega_D \) is due to \( \tau_B \) slowing. However, a direct measure of \( \tau_B \) could not be achieved because the
2.8 μm beads are spherically symmetric with any defects too small to be optically resolved. On the other hand, 7.9 μm COMPEL beads exhibit a similar \( \omega_D \) as the 2.8 μm beads and their orientation can be observed due to the presence of minor surface defects (Supplementary Videos 1 and 2). Between 1.4 and 1.8 Hz, \( \omega_D/2\pi \) increases past \( \omega_D/2\pi \), i.e., the individual 7.9 μm beads slow down and rotate asynchronously, which coincides with \( \omega_D/2\pi \) starting to decrease from 0.30 Hz to 0.19 Hz. This response for the 7.9 μm beads agrees with our model that the decrease in \( \omega_D \) is due to \( \omega_D \) slowing when \( \omega_D > \omega_D^* \).

The model (Eq. (5)) also quantitatively agrees with experimental results on the relative strength of the hydrodynamic bead-doublet coupling to the hydrodynamic drag as measured by \( \alpha_B = \gamma_B^2/\gamma_D^2 \) (Eqs. (3b) and (5)). In the region \( \omega_B < \omega_H < \omega_D^* \), a near-linear response \( \omega_B \approx \omega_B^* \) is observed at high frequencies (Fig. 8) when the hydrodynamic bead-doublet coupling is the dominant doublet torque. Based on the response of ten doublets, it is experimentally found that \( \omega_B = 0.20 \pm 0.04 \) which is in good agreement with the simulation-determined value of \( \omega_B = 0.20 \) (as found in Section 3.3). Because \( \gamma_B^2 \) is sensitive to the height above the substrate (decreasing 16% when the gap changes from 50 nm to 150 nm), the agreement in \( \gamma_B^2 \) validates our choice of a 50 nm bead-substrate gap in the simulations.

The model described by Eq. (5) includes assumptions that influence the fit to the data around the critical frequencies. For instance both beads are taken to have the same critical frequency \( \omega_D^* \) that leads to a sharp peak in the model at \( \omega_D^* \). When \( \omega_D^* \) for the beads is different, the response of \( \omega_D \) at \( \omega_D^* \) becomes smoother as seen in the measured data in Fig. 8. In addition, the current model ignores effects from thermally driven Brownian fluctuations [47] which causes the fits to over-estimate \( \omega_D \) around the critical frequencies. For example, even if \( \omega_D < \omega_D^* \), the addition of Brownian torque can cause the doublet to oscillate and, on average, slow down if \( \omega_B \) is sufficiently close to the critical frequency. Consequently, not including Brownian motion increases the predicted peak in \( \omega_D \) at the critical frequencies. Based on measurements of seven COMPEL doublets, the values of \( \omega_D \) at the critical frequencies based on the fits to Eq. (5), which do not include Brownian fluctuations, are 31 ± 7% and 26 ± 7% larger than the highest measured values of \( \omega_D \) at \( \omega_D^* \) and \( \omega_B^* \), respectively. Numerical simulations reported by Reenen et al. [47] for the simpler case of individually rotating 2.8 μm Dynabeads found a similar result where the exclusion of Brownian motion caused an increase of 16 ± 5% in the critical frequency.

From the critical frequencies measured using the doublet’s frequency response, both \( m_B \) and \( m_P \) can be determined for individual beads. Compared with single bead techniques that require coatings or fluorophores to identify the bead’s orientation, the present approach is a non-invasive means for determining \( m_B \). Based on a sampling of 13 doublets of 2.8 μm COMPEL beads at \( H = 20 \) Oe, the critical frequencies are \( \omega_B/2\pi = 0.40 \pm 0.02 \) Hz and \( \omega_B/2\pi = 3.60 \pm 0.16 \) Hz. As such, the maximum critical frequencies are \( \omega_D/2\pi = 0.40 \pm 0.02 \) Hz and \( \omega_D/2\pi = 5.17 \pm 0.17 \) Hz where \( \omega_D = \pi \omega_B/H \) and \( \omega_B = \omega_D/2\pi \). The resulting magnetic moments are \( m_B = (1.05 \pm 0.04) \times 10^{-15} \) Am² and \( m_P = (7.23 \pm 0.18) \times 10^{-15} \) Am². The magnetic susceptibility \( \chi = m/m_B \approx 0.37 \) if \( m_B \approx m_B^{MAX} \), which is justified because \( m_B \) is (5.9–6.8) \( m_B^{MAX} \) depending on the angle between \( m_B \) and \( m_P \).

4.3. Detached Dynabeads

A typical frequency response (shown in Fig. 9a) of \( \omega_D \) for detached Dynabeads at \( H = 20 \) Oe agrees with a fit based on Eq. (6), thereby allowing Dynabeads to be used to explore bead dynamics when \( \omega_B < \omega_D^* \).

As depicted in Fig. 9b, the doublet rotates at \( \omega_B \) when \( \omega_B < \omega_D^* \), and undergoes periodic oscillatory motion when \( \omega_B > \omega_D^* \). Unlike with the COMPEL beads, these oscillations continue for all the measured high frequencies, since the dipole–dipole torque, which causes the oscillations, remains the dominant torque.

Detached Dynabeads partially coated with a non-magnetic film were previously studied by Coughlan et al. [44] who found that the hydrodynamic bead-doublet coupling affected \( \omega_D \), but did not calculate the bead magnetic moment from \( \omega_D \). Based on six doublets at \( H = 20 \) Oe, the measured maximum critical frequencies are \( \omega_B/2\pi = 3.15 \pm 0.23 \) Hz and \( \omega_B/2\pi = 2.21 \pm 1.79 \) Hz. The resulting magnetic moments are \( m_B = (4.49 \pm 3.67) \times 10^{-16} \) Am² and \( m_P = (2.03 \pm 0.07) \times 10^{-15} \) Am². The lower accuracy for \( m_B \) is likely a result of the permanent moment being relatively weak for Dynabeads and not having a significant effect on doublet rotation. The magnetic susceptibility can be approximated in this case as \( \chi = m_B/m_B^{MAX} \approx 1.11 \) because \( m_B \) is roughly 45x greater than \( m_P \). As expected, the Dynabeads display a smaller value of \( m_B \) and a larger \( m_B^{MAX} \) than COMPEL beads. In comparison, Coughlan et al. [44] found comparable values for the magnetic moments of the same type of Dynabeads with \( \chi = 1.33 \) at \( H = 10 \) Oe and \( m_B = 3.0 \times 10^{-16} \) Am² through a bulk particle property measurement system and single bead analysis respectively. The magnetization among the beads vary considerably [47], which is likely the cause for the variations in the measured magnetic moments.

4.4. Maximum rotation frequency

\( \omega_D^{MAX} \) for attached and detached COMPEL and Dynabeads are summarized in Fig. 10. \( \omega_D^{MAX} \) is not directly dependent on the ratio of
the critical frequencies. In our case, the maximum frequency for COMPEL and Dynabead doublets are comparable, which is possible because they are each characterized by different magnetic moments: \( m \) is 181% larger for Dynabeads and \( m_P \) is 134% larger for COMPEL beads. However, the effect of attachment on \( \sigma_{\text{Hyd}}^{\text{MAX}} \) is dependent on the relative strength of \( \omega_B^D \) to \( \omega_B^P \) as predicted in Fig. 6.

When \( \omega_B^D > \omega_B^P \), attached doublets generally rotate faster than detached doublets, which is also observed with COMPEL beads. The attached doublets rotate over 4x faster (2.73 ± 0.15 Hz) than detached doublets (0.62 ± 0.04 Hz). The hydrodynamic bead-doublet coupling allows detached doublets to rotate faster than when only under a magnetic dipole-dipole torque \( \tau_D \), but it is a smaller effect on \( \sigma_{\text{Hyd}}^{\text{MAX}} \) than if the beads are attached. The hydrodynamic bead-doublet coupling allows \( \sigma_{\text{Hyd}}^{\text{MAX}} \) to be 50% greater than \( \sigma_D \) (0.62 ± 0.04 Hz versus 0.40 ± 0.02 Hz) but occurs when \( \omega_B^P \) is 9x greater (3.6 ± 0.16 Hz versus 0.40 ± 0.02 Hz).

When \( \omega_B^D < \omega_B^P \), detached doublets generally rotate faster than their attached counterparts as seen (Fig. 10) with Dynabeads where detached doublets rotated 20% faster (2.66 ± 0.08 Hz) than attached doublets (2.21 ± 0.11 Hz).

5. Conclusion

This study has addressed the role of hydrodynamic and magnetic torques on the rotational dynamics of a pair of magnetic beads. Simulations confirm a linear dependence of the hydrodynamic torque on \( \omega_B \) and \( \omega_B^D \), i.e. \( \tau_B^{\text{Hyd}} = \gamma_B^P \omega_B \omega_B^D - \gamma_B^P \omega_B \omega_B^D \) and \( \tau_B^{\text{Hyd}} = \gamma_P \omega_B \omega_B^D - \gamma_P \omega_B \omega_B^D \) which was experimentally confirmed by the doublet response (Figs. 8 and 9). Models for the cases where \( \omega_B^D > \omega_B^P \) and \( \omega_B^D < \omega_B^P \) agreed with \( \sigma_D \) measured for COMPEL and Dynabeads respectively which allowed the beads to be used as examples of the two regimes. Moreover, our simulation results were validated when the value for \( \omega_B^D \) reflects the drag-reduced bead-doublet coupling, matched the experimentally determined \( \omega_B^P = 0.20 \pm 0.04 \) from COMPEL beads.

For detached doublets, \( \chi \) and \( m_B \) were determined from the frequency-dependent response of \( \sigma_B \). This represents a non-invasive method of determining the permanent bead moment \( m_B \) of individual beads which is central to the dynamics of single and chain rotations. In contrast to the more precise value of \( m_B = (1.05 \pm 0.04) \times 10^{-15} \text{Am}^2 \) for COMPEL beads, our value of \( m_B = (4.49 \pm 3.67) \times 10^{-16} \text{Am}^2 \) for Dynabeads had more uncertainty due to the relative weakness of \( m_B \) and its modest effect on \( \sigma_B \).

It is also noted that while doublets have been assumed to rotate faster when detached [44], this is true only when \( \omega_B^D < \omega_B^P \) (Figs. 6 and 10). When \( \omega_B^D > \omega_B^P \), \( \sigma_{\text{Hyd}}^{\text{MAX}} \) is greater for attached doublets.

In general, micromagnetic actuation is hindered by the effects of viscosity which places a limit on the maximum rotation rate. Attachment is one way to overcome that barrier by exchanging torque between a body and its constituent parts. For the doublets, when the Dynabeads were detached or the COMPEL beads attached, \( \sigma_{\text{Hyd}}^{\text{MAX}} \) was able to increase by decreasing the rotation rate of the individual beads. Likewise, this effect could be extended to more complex many-body systems.

The findings presented in this study have broad applications. For instance, simple, non-invasive bead calibration can be performed by measuring the frequency-dependent \( \sigma_B \) to determine the relative strengths of permanent and induced moments. For drug delivery protocols, the calibrated, bio-compatible beads may be administered along with drugs. Because of the permanent moment, individual beads would be able to roll independently through small pores to reach the target site before chaining into larger structures for on-site micro-mixing [26]. The mixing rate would be increased for beads with a large permanent moment and could potentially lead to increased drug effectiveness. The results thus provide a framework to engineer emergent behaviors of magnetic microbeads in complex and crowded environments.

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