Observation of Enhanced Diffusivity in Magnetically Powered Reciprocal Swimmers

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We report on the development of a system of micron-sized reciprocal swimmers that can be powered with small homogeneous magnetic fields, and whose motion resembles that of a helical flagellum moving back and forth. We have measured the diffusivities of the swimmers to be higher compared to nonactuated objects of identical dimensions at long time scales, in accordance with the theoretical predictions made by Lauga [Phys. Rev. Lett. 106, 178101 (2011)]. Randomness in the reciprocity of the actuation strokes was found to have a strong influence on the enhancement of the diffusivity, which has been investigated with numerical calculations.

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Locomotion at nano- and microscales in fluidic environments is dominated by viscous forces, for which living microorganisms such as various species of bacteria, spermatozoa, etc., employ special swimming strategies [1,2] to achieve propulsion. The idea is to use nonreciprocal swimming strokes, which correspond to a set of cyclic deformations that are not time reversible in nature, allowing the microorganisms to have directed propulsion at short time scales. The effects of thermal fluctuations can play an important role in their dynamics [3,4], since the rotational diffusion of the swimmer may result in a loss of orientation, thereby giving rise to diffusive behavior at long time scales, where the effective diffusivity can be several orders of magnitude higher compared to an object of similar dimensions undergoing Brownian motion only. The situation is expected to be different for reciprocal swimmers which cannot achieve any net displacement over one period of actuation, but are still subject to translational and rotational Brownian displacements in the presence of thermal noise. Theoretically, this problem has been considered by Lauga [5], who showed that for scales larger than rotational diffusion, significant enhancement over Brownian diffusivities are also possible with reciprocal swimmers, with strong dependence on the time period of reciprocal actuation, $f^{-1}$ and the orientational diffusion time $\tau_\Omega$. Thus, reciprocal actuation can have significant influence over cellular motility; as shown by Lauga [5], over 2 orders of magnitude advantage over Brownian diffusivity for micron-sized marine bacterial species, *Shewanella putrefaciens* (CB-32 genome).

In order to confirm the predicted enhancement of diffusivity, it was first necessary to develop a system of externally powered reciprocal swimmers, whose sizes are small enough to have appreciable Brownian diffusion in time scales relevant to the experiment. With current advances in nanotechnology, it has been possible to obtain several nano- and microscale propulsion systems [6,7] that can be actuated externally, quite commonly with magnetic fields [8]. An important and relevant example is that of magnetized helical nanostructure [9,10], which can be propelled in fluids by a rotating magnetic field, along a direction defined by the sense of rotation of the field and the handedness of the helix. This is an important aspect in which the system of magnetic propellers differ from autonomous swimmers, such as self-propelled biological microorganisms or chemically powered catalytic nanorods [11,12], whose direction of motion, in the absence of any interactions, is completely random and only governed by thermal fluctuations. Interestingly, as we show in this Letter, the same system of helical nanostructures in the presence of a nonrotating, but oscillating magnetic field can demonstrate a back-and forth motion, whose directionality remains unrelated to the characteristics of the external drive, and therefore, qualifies as a reciprocal swimmer.

The working principle of the system of reciprocal swimmers can be understood in the following way. Consider a helix lying on $x$-$y$ plane pointing an arbitrary direction, whose magnetic moment is perpendicular to the long axis of the helix. In the presence of a magnetic field oscillating along the $z$ axis, for the moment to remain aligned with the field, the structure would turn either clockwise (CW) or counterclockwise (CCW), and therefore go forward or backward by an amount $\pm p_h/2$, where $p_h$ is the hydrodynamic pitch [13] of the helix. Most crucially, the body frame displacements would be unrelated to the orientational diffusion of the helix in the $x$-$y$ plane, as is the case for autonomous swimmers, whose direction of motion, in the absence of any interactions, is only governed by thermal fluctuations. To check this idea experimentally, we have fabricated magnetic helical nanostructures using a method described in detail before [14,15]. To actuate the helices, a square wave magnetic field of frequency $f_B$ is applied to a coil placed on a microscope, and a glass chamber containing the helices dispersed in deionized...
water was placed at the focal plane of the microscope objective. The helices (schematic shown in Fig. 1(a), SEM image in the Supplemental Material [16]) were made of silica with a thin metal (magnetic) coating on one side of the helix. During the experiments, the helices remained confined in a plane perpendicular to the optical axis, and we could not detect any out of plane motion. The coating was necessary to render the structures magnetic, and helped us to visualize the rotational dynamics of the helices. As shown in Fig. 1(b), an almost perfect sequence of clockwise and counterclockwise rotations was observed (also see movie S1 in the Supplemental Material [16]). This near-perfect reciprocal behavior occurred due to the asymmetry in the weight distribution of the helix brought about by the weight of a thin coating of the magnetic material, whose line of action did not coincide along the direction of the magnetic moment. The angle $\phi$ between these directions (defined in Fig. 1(c)) was estimated from microscope images to be $\leq 20^\circ$, but could not be measured with higher precision due to limitations of the imaging system.

The proposed mechanism based on weight asymmetry is depicted schematically in Fig. 1(c), where under the presence of a magnetic field, e.g., along $+\hat{z}$ direction, the torques due to the weight of the magnetic material and due to the coupling of the magnetic moment with the applied field balance each other, which cause the equilibrium direction of the moment to be slightly deviated (angle denoted as $+\alpha$) from $+\hat{z}$. Upon reversal of the direction of the magnetic field, i.e., to $-\hat{z}$ direction, the helix turns clockwise to a position where the moment is slightly deviated (angle denoted as $\beta$) from $-\hat{z}$, again determined by the counteracting torques. Similarly, when the field direction is reversed back to $+\hat{z}$ direction, the helix would turn in the counterclockwise direction, thus undergoing a full cycle of reciprocal motion.

To check if the near perfect reciprocal motion was indeed related to the weight of the magnetic film, we investigated the dynamics of the helix due to the torque induced by gravity alone. The helix was magnetized such that the film was above the SiO$_2$ part of the helix (left, Fig. 2) upon application of a magnetic field along $+\hat{z}$ direction. When the field was turned off, the helix turned almost a half rotation in the clockwise direction, such that the magnetic film was below the dielectric. On the other hand, when the magnetic field was along $-\hat{z}$ direction, the film was situated below the SiO$_2$ part (right, Fig. 2). As the field was turned off, the helix rotated slightly in the counterclockwise direction, which was too small to be distinguished from thermal fluctuations. The gravitational torque due to the magnetic film was therefore responsible for the heavy side to be preferentially located at the bottom.

In the sequence of turns shown in Fig. 1(b), certain events ($q = 2/15$) did not fit in a perfectly reciprocal sequence. This “randomness” could be explained by assuming a rotation in a direction not in accordance with the torque balance, possibly mediated by the rotational diffusion of the helix around its long axis. For the experiments discussed in this Letter carried out with helices of almost identical geometry, the degree of randomness ($q$) varied between 0% and 25%, and this variation was due to a distribution of angles $\phi$ among different helices, since smaller $\phi$ would imply a greater possibility of having a random turn mediated by thermal noise. Interestingly, the occurrence of the random events were almost always preceded and followed by similar events, as would be expected when the dynamics is primarily governed through a deterministic balance of torques. To elaborate on this, the random events (indicated within parentheses) typically were in a sequence of type I: CW-CCW-(CCW)-CW or CCW-CW-(CW)-CCW, but were very seldom in a sequence of type II: CW-CCW-(CCW)-(CW)-CW or CCW-CW-(CW)-(CCW)-CCW. A detailed statistics of the sequence of random turning events for all the experimental data presented in this Letter is provided in the Supplemental Material [16]. Also, the value of $q$ was almost zero with larger helices (SEM image and movie S2 in the Supplemental Material [16]), whose rotational

![FIG. 1 (color online).](image)

(a) Schematic and expected dynamics of a magnetized helix under an oscillating field. Upon reversal of the field direction, the helix can have either clockwise or counterclockwise rotation, such as to have the moment aligned to the applied field. (b) An example sequence of turning events of the helices. The occurrence of two random events has been marked with arrows. (c) Proposed mechanism behind the observed near-perfect reciprocal behavior, along with the variation of the magnetic field.
diffusion constant around the long axis was significantly smaller, and therefore, any nonzero $\phi$ could give rise to perfectly reciprocal motion with smaller effect from thermal fluctuations.

To confirm the proposed mechanism behind the origin of near perfect reciprocal motion, we modeled the dynamics of the helix, in the absence of thermal noise as

$$\gamma I \frac{d\alpha}{dt} = mB_z(t)\sin\alpha + WR\sin(\alpha + \phi).$$

Here, $\gamma I$ corresponds to the rotational drag of the helices about the long axis ($\sim 10^{-21}$ kg m$^2$/s), $m$ and $W$ refer to the magnetic moment ($\sim 10^{-16}$ Am$^2$) and weight ($\sim 10^{-14}$ N) of the magnetic film, respectively. We assumed $R (= 0.5 \mu m)$ to be the radius of the helix, which provides an approximate estimate of the gravitational torque ($\sim WR$). The magnetic field $B_z(t)$ was assumed to have a functional form similar to the experimentally applied field, which was close to a square wave. The detailed method of obtaining the numerical values for these quantities has been provided in the Supplemental Material (Secs. III and IV) [16]. The time evolution of angle $\alpha$ is given by $\alpha(t + dt) = \alpha(t) + \Delta t(d\alpha/dt)$, where $\Delta t = 10^{-6}$ s refers to the time step of the numerical calculation. As shown in Fig. 3(b) of the Supplemental Material [16], the dynamics of $\alpha$ in the absence of thermal noise showed a perfect reciprocal motion, where the calculated time of rotation for a half turn, denoted by $T_{rot}$, was found to be $\sim 13$ ms and was reasonably close to the experimentally measured value of $T_{rot}(\sim 10$ ms).

To include the effect of thermal fluctuations, Eq. (1) was modified to include a random fluctuation,

$$\alpha(t + dt) = \alpha(t) + \Delta t \frac{d\alpha}{dt} + \xi(t),$$

where $\xi(t)$ is a random Brownian rotation at temperature $T$, given by $\langle \xi(t)\xi(t') \rangle = (2kT/\gamma I)\delta(t - t')$, where $k$ is Boltzmann constant. The time evolution of $\alpha$ for $\phi = 20^\circ$ including thermal noise is shown in Fig. 3(c) of the Supplemental Material [16], where certain rotations ($\sim 27\%$) could be observed which were in the opposite direction to a perfectly reciprocal sequence. To summarize the results of the numerical calculations, perfectly reciprocal motion could be predicted in the absence of thermal noise, but showed some randomness when thermal fluctuations were included in the calculations. The degree of randomness depended strongly on the angle $\phi$ and was found to be significantly smaller with larger helices, thereby confirming the proposed mechanism and the experimental observations.

To confirm the predictions of higher diffusivity in reciprocal swimmers compared to Brownian objects of the same dimensions, we measured the positions of the helical nanostructures, both with and without an oscillating magnetic field. Trajectories imaged for 50 s for these two cases have been plotted in Fig. 3(a), where larger trajectories were observed when the structures were actuated with magnetic fields. These images were taken at 53 frames per second.
randomness, we assumed a fraction \( q \) and one was generated by a computer program, given by \( r_n \) sequence, and then reversed the sign of the \( n^{th} \) velocity pulse, if \( r_n < q \). Once \( u(t) \) was obtained, we estimated the enhancement of diffusivity \( \Delta D \) numerically using Eq. (3) (following Ref. [5]),

\[
\Delta D = \frac{1}{2} \left\{ \lim_{t \rightarrow \infty} \int_{0}^{t} u(t')u(t')e^{-\left(t-t'/\tau\right)/\tau} dt' \right\}.
\] (3)

where the averaging was done over one time period and 100 different realizations of velocity sequences. For comparison, as shown in Eq. (4), an analytical formula for \( \Delta D \) was also obtained (see Supplemental Material [16]) assuming perfect \( (q = 0) \) reciprocal motion, where the body frame velocities were mathematically described as a series of impulse functions of alternate signs.

\[
\Delta D = p_{B}^2 f_B \tan \left( \frac{1}{4f_B \tau_r} \right)/8.
\] (4)

The numerical results for different values of \( q \), along with the analytical formula [Eq. (4)] are shown in Fig. 4, where the enhancement of diffusivity was seen to depend very strongly on the randomness in the actuation sequence. The finite rotation time of the helix was of lesser significance (especially at low frequencies), as could be seen from the simulation results at \( q = 0\% \) when compared with the estimates obtained from Eq. (4). Also shown in the same figure are the experimental results for different helices (but of same dimensions), all of which had different degrees of randomness \( (q) \) as measured from video analysis. The experimental measurement of \( q \) was obtained by estimating the number \( (Q) \) of random turns in an observation sequence of total \( N \) turns, thereby providing an estimate of measurement error for \( q \), given by \( \delta q = \sqrt{Q(1-Q)/N} \). The experimentally obtained \( \Delta D \) for a particular actuation frequency \( (f_B) \) and randomness \( (q \pm \delta q) \) was in agreement with the theoretical predictions, except for the experimental data point with \( q = (16 \pm 5)\% \) obtained at \( f_B = 1 \) Hz, where \( \Delta D \) was found to be slightly higher than theoretical predictions.

As far as we are aware, all experimental demonstrations [17, 18] of reciprocal actuation so far has been obtained with macroscale objects, which automatically precludes the investigation of phenomena [19, 20] that can arise from thermal fluctuations, such as the observation of enhanced diffusivity reported in this Letter. The structures and the method described here are the first experimental realization of a microscopic reciprocal swimmer, which can be in fluidic suspension for extended periods of time and generate hydrodynamic flows, thus providing a powerful system to test various theoretical predictions [21–25] about the behavior of a collection of reciprocal swimmers coupled via hydrodynamic interactions. The swimmers are chemically compatible with most fluidic media and it will be interesting to see how they behave in non-Newtonian fluids [18, 26, 27] or in close proximity to a deformable [17] surface, for which interesting theoretical predictions and macroscale experimental realizations exist.
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