Twirling DNA Rings - Swimming Nanomotors Ready for a Kickstart

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We propose a rotary DNA nanomachine that shows a continuous rotation with a frequency of $10^2$ to $10^4$ Hz. This motor consists of a DNA ring whose elastic features are tuned such that it can be externally driven via a periodic temperature change. As a result the ring propels itself through the fluid with a speed up to microns per second.

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The long lasting dream of scaling mechanical devices and machines down to the nanoscale (as popularized by R. Feynman and carried on by several visionary groups worldwide) continues to fire the imagination of researchers – now in the third generation. Among many experimental difficulties that appear in this context, choosing the proper material for the assembly of a nanodevice turns out to be crucial. Important material requirements are: stability, self-assembly ability, modularity, replicability, switchability, experimental tractability. Presently one of the most promising materials fulfilling those requirements is DNA. Assemblies based on DNA hybridisation chemistry as well as conformational DNA transitions were successfully exploited to generate periodically switchable nanodevices. However, despite their beauty and conceptual originality all of these devices suffer one major problem: the large kinetic barriers involved in the switching process boost their switching time per cycle to $\sim 10^3$ s, four orders of magnitude slower than their natural counterparts (biological molecular motors). A natural question arises then: Can one achieve subsecond switching times with a DNA nanodevice? Can such a device be operated in some manner to swim directionally and faster than $1\, \mu m/s$? In this letter we show theoretically the principal feasibility of such DNA nanomachines.

Let us in the following propose a surprisingly simple nanomotor: a DNA miniplasmid, cf. Fig. 1(a). We will show that despite its structural simplicity a miniplasmid can be turned into a nanomachine able to produce $fN$ forces and self-propelling at speeds of several microns per second through the fluid. In order to run the plasmid as a motor we use here the Euler-angle $\psi$ (cf. Fig. 1(a)) as the relevant degree of freedom. The main idea now is to induce a directed current $\langle \dot{\psi} \rangle$ – in a manner similar to the rotation of a closed rubber tube around its central circular axis – via non-equilibrium fluctuations and the ratchet effect, cf. Fig. 1(b). As a result the twirling ring generates a hydrodynamic flowfield (shown in Fig. 1(c)) that remarkably induces an efficient self-propulsion of the motor as detailed below.

The elastic distortion energy of a DNA ring with radius $R$ parametrized by the arc length parameter $s$ will in general be described by three Euler angles $\theta(s), \phi(s)$ and $\psi(s)$ via $E_{el} = \frac{1}{2} k_B T \int_0^{2\pi R} \sum_{i=1,2,3} l_i (\omega_i - \kappa_i)^2 ds$ with $\omega_1 = \phi' \sin \theta \sin \psi + \theta' \cos \psi, \omega_2 = \phi' \sin \theta \cos \psi - \theta' \sin \psi$ and $\omega_3 = \phi' \cos \theta + \psi'$. Here $l_1$ and $l_2$ are the two principal bending persistence lengths and $\kappa_1$ and $\kappa_2$ are intrinsic curvatures in two corresponding perpendicular directions. $l_3$ denotes the twist persistence length and – for simplicity – we choose $\kappa_3 = 0$. The parameters $\kappa_i$ and $l_i$ reflect the anisotropic bendability as well as intrinsic bendedness of the plasmid; here for sim-
before we compute the friction constant $\zeta$ that we will compute below. As a source of non-equilibrium we will choose here a time-dependent variation of temperature $T(t)$, cf. Ref. [13].

Before we compute the friction constant $\zeta$ we need to shed some light on the low Reynolds number hydrodynamics of the twirling DNA ring. The latter turns out to be peculiarly related to the inviscid (ideal) fluid vortices (rings of smoke) and as a matter of fact both of them propagate in analogous manner. To see this we first remark that for a reasonable ring radius $R = 10 \text{ nm}$ (a typical miniplasmid of about 200 bp) and the DNA helix radius $r_0 = 1 \text{ nm}$ the slender body approximation [14] is valid with the slenderness parameter $\varepsilon = r_0/R = 0.1$.

In the spirit of the slender body theory one approximates the flow-field around the twirling ring by superimposing rotlets $\mathbf{u}_{\text{rot}}(\mathbf{x}; s) = \Gamma \delta(s) \times (\mathbf{x} - \mathbf{c}(s)) / |\mathbf{c}(s) - \mathbf{x}|^3$ placed along the ring centerline $\mathbf{c}(s)$ with arclength parameter $s$. The rotlet strength $\Gamma = 1/2\omega_c r_0^2$ is given in terms of the angular velocity $\omega_c$ of the ring about $\mathbf{c}(s)$. The full velocity profile is then given by $\mathbf{u}(\mathbf{x}) = \int_0^{2\pi R} \mathbf{u}_{\text{rot}}(\mathbf{x}; s) \, ds$; cf. also the stream lines around the rotating ring shown in Fig. 1(c). When integrating $\mathbf{u}(\mathbf{x})$ over the DNA ring (slender torus) surface in the limit of small $r_0/R$ one obtains a net translational velocity in the $z$-direction:

$$v_z(\omega_c) = \frac{R^3}{2R} \left( \ln \left( \frac{8R}{r_0} \right) - \frac{1}{2} \right) \omega_c$$  (3)

The fact that Eq. 3 coincides with the well known expression from ideal flow vortex theory [15] should not surprise if we recall that a rotlet $\mathbf{u}_{\text{rot}}(\mathbf{x}; s)$ is nothing else but the expression for the velocity field of an ideal point vortex. But despite this kinematic analogy between the twirling DNA and an ideal vortex ring, dynamically they are quite different. The propagation of an ideal vortex ring does not require any external forces/torques and is governed by conservation of kinetic energy and momentum. In sharp contrast to that the low Reynolds-number (Stokes) flow is governed by dissipation and the motion of twirling DNA ring requires the action of a torque $N_c = 8\pi^2 x_R^2 \eta R \omega_c (\eta = 10^{-3} \text{ Pa s}, the water viscosity)$ about the central axis $\mathbf{c}$. The latter expression can be verified by integrating the tangential stresses generated by $\mathbf{u}(\mathbf{x})$ over the ring surface. More generally by virtue of the linearity of the Stokes equations we can derive a resistance matrix ($M_{kl}$) relating the angular velocity $\omega_c$ (about the circular axis $\mathbf{c}$) and velocity $v_z$ (in the $z$-direction) with the corresponding external torque $N_c$ and force $F_z$:

$$\begin{pmatrix} F_z \\ N_c \end{pmatrix} = 4\pi^2 \eta \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix} \begin{pmatrix} v_z \\ \omega_c \end{pmatrix}$$  (4)

Combining the previous expressions obtained for $v_z(\omega_c)$ and $N_c(\omega_c)$ ($F_z = 0$) together with the result of Johnson and Wu [14] for the drag on a rigid slender torus we obtain entries in the leading order: $M_{11} = 2R \left( \ln 8/\varepsilon + 1/2 \right)^{-1}$, $M_{22} = 2R^2 \varepsilon$ and $M_{12} = M_{21} = r_0^3 \left( \ln 8/\varepsilon - 1/2 \right) \left( \ln 8/\varepsilon + 1/2 \right)^{-1}$. Note that the symmetry of the resistance matrix being a general feature of swimmers in the Stokes flow [17] provides a good check for the consistency of the involved calculations. Having

FIG. 2: The rotational current $\langle \dot{\psi} \rangle$ and the induced translational velocity $v_z$ as a function of the dimensionless frequency $\tilde{f}$ of the temperature (potential) oscillations. The DNA ring has the following parameters: $R = 10 \text{ nm}$, $r_0 = 1 \text{ nm}$ (typical DNA minicircle), $l_1 = 45 \text{ nm}$, $l_2 = 50 \text{ nm}$, $\kappa_1 = \kappa_2 = (200 \text{ nm})^{-1}$ leading to the ratchet potential displayed in the inset. Displayed are the asymptotic expressions, Eqs. 4 (dashed line) and 5 (dashed-dotted line) together with the numerical solution of Eq. 2 (thin line) for a temperature ratchet with $A_T = 0.03$. The thick solid line corresponds to an oscillating potential ratchet with $A_E = 0.3$. See text for details.
the mobility relation one can consider different types of motion, e.g.: (i) The ring is twirling freely \((N_c = 0)\) and moving under the force \(F_z\) (or at fixed \(v_z\)). (ii) The ring is prevented from rotation \((\omega_c = 0)\) and moved by the force \(F_z\) (or at fixed \(v_z\)). (iii) The ring is held in position \((v_z = 0)\) by imposing a force \(F_z\) counterbalancing the action of torque \(N_c\). (iv) The DNA ring is free to move (no external force applied, \(F_z = 0\)) under an externally imposed torque \(N_c\) (or at given \(\omega_c\)). Comparing, for instance, the velocities \(v_z^i\) and \(v_z^i\) from cases (i) and (ii) one sees that \(v_z^i/v_z^i = \frac{M_k}{\det M_k} \approx (\epsilon/2)^2 \ln(8/\epsilon)\); i.e., a ring with an isotropic DNA sequence (able of twirling) settles faster than a ring with very high barriers to twirling \((\omega_c = 0)\), though in practice the difference is negligible, for instance \(\sim 1\%\) for \(\epsilon = 0.1\). By comparing cases (iii) and (iv) we conclude that a ring twirling at fixed \(\omega_c\) and forced not to translate requires a slightly larger torque \(N_c^i(\omega_c)\) than the unconstrained freely translating ring with \(N_c^i(\omega_c)\). However the relative difference is again small and of order \(O(\epsilon^2 \ln(8/\epsilon))\). Therefore by dropping this marginal correction to the leading order we obtain in both cases (iii) and (iv) the angular friction constant \(\zeta = N_c/\omega_c \approx 8\pi^2 \eta r_0^2 R\), the quantity that appeared above in Eq. 2. Note that the latter is the same (in our \(\epsilon \ll 1\) leading order expansion) as for a straight cylinder with radius \(r_0\) and length \(2\pi R\). Finally another interesting feature that can be read off Eq. 4 is the efficiency of the twirling ring propulsion. The latter is independent of the mechanism of twirling and can be defined as the ratio of the power \(P_0 = 2\pi^2 \eta M_1 \nu_z^2\) dissipated by a (for simplicity) rigid ring directly moved by a force as compared to the power \(P_{twirl} = \frac{1}{2} N_c \omega_c\) dissipated by twirling propulsion at the same translational speed. For a ring with \(R = 10\ nm\) we have \(P_0/P_{twirl} \approx 0.8\%\) which is comparable to the efficiency of bacterial propulsion by a rotating flagellum.

Having determined the friction constant \(\zeta\) we return to the ratchet dynamics given by Eq. 2 with the twirling potential Eq. 4. To obtain the directed twirling frequency \(\omega_c\) of motion, e.g.: \((\epsilon/2)^2 \ln(8/\epsilon)\) and the abbreviations \([\ldots]\) and \(\langle \ldots \rangle\) as defined in 13 but with the integrations with respect to \(\psi\). Furthermore the density distributions \(P_0\) and \(P_1\) from the upper expansion are given by \(P_1 = E_{el} / T\) (Boltzmann distribution in the adiabatic limit) and \(P_1 = \frac{1}{\pi} E\langle FC_1\rangle\) with \(c_1 = \frac{\partial T}{\partial \psi} - \frac{1}{2} F \frac{\partial}{\partial \psi} F^{-1}\).

Equations 5 and 6 together with Eq. 1 and \(\zeta = 8\pi^2 \eta r_0^2 R\) allows us to get the twirling speed \(\omega_c = \langle \psi \rangle\) and by virtue of Eq. 3 the induced translational velocity \(v_z(\omega_c)\) for arbitrary DNA elastic parameters \(l_i = 1.2\) and \(\kappa_i = 1.2\).

How fast can we operate the twirling ring machine? We shall assume some realistic parameter values for the DNA ring: \(R = 10\ nm\), \(r_0 = 1\ nm\) (typical DNA mini-circle) leading to \(\zeta = 2 \cdot 10^{-7} k_B T s\). Furthermore we set \(l_1 = 45\ nm\), \(l_2 = 50\ nm\), \(\kappa_1 = \kappa_2 = (200\ nm)^{-1}\) which corresponds to a rather modest anisotropy and intrinsic curvature. For the temperature variation amplitude we choose \(\Delta T = \pm 10\ K\), i.e., \(\Delta T \approx 1/30\) (at room temperature \(T_0 = 300\ K\)). Figure 2 provides a log-log plot of the rotational current and the corresponding drift speed of the ring as a function of the dimensionless frequency \(f\) of the temperature variation. The thin solid curve gives the numerical result obtained from Eq. 2; the two straight lines correspond to the analytical results for the two asymptotic cases, Eqs. 4 and 5. As can be seen from this plot the two limits show a \(f^{-2}\) and \(f^2\) dependence, respectively, in accordance with Eqs. 4 and 5. The maximal rotational current is achieved in the crossover region, namely \(\omega_c \approx 200\ rad/s\) for \(f \approx 10^{-1}\). Following Eq. 4 this implies a translational velocity of \(v_z = 50\ mm/s\).

Such fast temperature oscillations are technically feasible and might be most conveniently generated by adiabatic pressure variations e.g. by ultrasound. Another promising method is to use the inductive heating of metal nanocrystals that are covalently attached to the DNA ring. In fact, this method has been successfully used to control the hybridization behavior of DNA 20. This might also point towards an alternative way of driving the ratchet, namely via a periodic...
variation of the elastic properties of the ring. Operating the system close to the DNA duplex melting temperature is likely to induce strong oscillations in the overall ring stiffness. The thick solid line in Fig. 2 shows the rotational current obtained when the elastic energy is varied as $\dot{E}_{el} = \frac{dE_{el}}{dt} = \int_{-\infty}^{\infty} \frac{d}{dt} \left[ \frac{1}{2} \kappa (\psi - \psi_0)^2 \right] dt$ where we chose the relative amplitude $A_E = 0.3$. As can be seen from Fig. 2 the maximal current of this oscillating potential ratchet occurs roughly at the same frequency as that of the thermal ratchet but the value of $\omega_c$ is much higher, namely on the order of $2 \times 10^4$ rad/s which implies a quite notable translational velocity of $v_z = 5 \mu m/s$. As a comparison a typical bacterium moves at $30 \mu m/s$. Our ring ratchet (with oscillating potential) resembles in many respects “real” biological nanomotors. Besides its nanoscopic size (radius $10 \text{ nm}$), swimming efficiency ($0.8\%$) and speed ($4 \mu m/s$) it can generate forces and torques close to that of biomolecular motors. Although the net translational force resulting from Eq. 4 $F_z = 4 \pi M_{12} \omega_c \approx 0.6 \text{ fN}$ is comparably small (due to cancelling of most of the stresses), the local torque $N_c = 8 \pi \eta R \omega_c \approx 0.004 k_B T$ and the force acting at the DNA surface $F_{1loc} = N_c / r_0 = \zeta \omega_c / r_0 \approx 16 \text{ fN}$ are significant if we consider the simplicity of the mechanism behind.

From an experimental point of view one should be aware of the fact that a ring (twirling or non-twirling) looses its initial orientation almost instantaneously due to rotational diffusion. The typical relaxation time scale of this process is on the order $R^3 / (k_B T)$ (up to logarithmic corrections) which for a ring with $R = 10 \text{ nm}$ leads to $10^{-7} \text{ s}$. That means that a single twirling ring in solution will not perform any noticeable translational motion can be neglected. The correct time dependence term in Eq. (2.58) of Ref. [11] should read $\Delta \psi - \left( \Delta (\psi) \right) ^2$ instead of $\Delta \psi$ with $\Delta (\psi) = 1 - \frac{T (2m \hbar)}{4}$. Here we introduce the abbreviations $\int_0^\infty \left( \int \right) dx$ for definite and indefinite integration.

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