Laser Spinning of Nanotubes: A path to fast-rotating microdevices

Petr Král\textsuperscript{1,2} and H. R. Sadeghpour\textsuperscript{2}

\textsuperscript{1} Department of Chemical Physics, Weizmann Institute of Science, 76100 Rehovot, Israel
\textsuperscript{2} ITAMP, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts 02138

We show that circularly polarized light can spin nanotubes with GHz frequencies. In this method, angular moments of infrared photons are resonantly transferred to nanotube phonons and passed to the tube body by “umklapp” scattering. We investigate experimental realization of this ultrafast rotation in carbon nanotubes, levitating in an optical trap and undergoing mechanical vibrations, and discuss possible applications to rotating microdevices.

Carbon nanotubes\textsuperscript{4} have unique mechanical and electronic properties with many potential applications\textsuperscript{4}. They possess a huge Young modulus\textsuperscript{5} \(Y > 1\) TPa, which adjusts their autonomous mechanical oscillations to MHz frequencies\textsuperscript{3}. Moreover, their "molecular structures" remain naturally stable even at large deformations\textsuperscript{1}. Therefore, rotationally symmetric structures based on stiff nanotubes could form ideal piston-rods for nanoscale applications. In contrast to chemically driven bio-motors\textsuperscript{6}, spinning with Hz frequencies, such tubular structures could rotate very fast, if angular momentum is efficiently transferred to them and friction is reduced.

Small heteropolar molecules can be dissociated\textsuperscript{6}, if synchronously rotated with a dipolar laser trap, which accelerates its angular velocity. Larger molecules\textsuperscript{3} and micro-particles\textsuperscript{7} can be rotated by absorption of angular momentum from circularly polarized or "twisted" laser beams. Nanotubes are excellent candidates for this asynchronous driving, where the system rotational frequency is much smaller than the light frequency.

Here, we investigate ultrafast asynchronous rotation induced in nanotubes by excitation of their vibrational modes with circularly polarized light. The mode selection is restricted by radiational heating, since each photon absorbed by the tube transfers to it angular momentum \(\hbar\) and energy \(\hbar\omega\). The resulting heating can be limited in excitation of infrared (IR) \(A_{\alpha\gamma}\) or \(E_{\alpha\beta}\) phonon modes, active in graphite\textsuperscript{8} and nanotubes\textsuperscript{10}.

![FIG. 1. Scheme for nanotube (up) and tubular ring (down) spinning with angular velocity \(\omega_{\text{rot}}\) in a laser trap. Their rotation is induced by absorption of circularly polarized photons from a laser beam with intensity \(E^*\), propagating along the axis of rotational symmetry. Scattering of molecules with tubes damps the rotation.](image)

by friction with the surrounding molecules. In the lower configuration, a closed nanotube ring\textsuperscript{11} is analogously rotated by absorption of circularly polarized photons.

We can describe the excitation of nanotube phonons by circularly polarized light, and the subsequent relaxation, with the simplified Hamiltonian

\[
H = \sum_{\alpha} \hbar \omega_{\alpha} b_{\alpha}^\dagger b_{\alpha} + \sum_{\alpha, \pm} \hbar \omega_{\alpha, \pm} E_{\alpha, \pm}^\dagger(t) \left( b_{\alpha, \pm}^\dagger + b_{\alpha, \pm} \right) + \sum_{\alpha, \pm, \beta, \gamma} \left( c_{\alpha, \pm, \beta, \gamma} b_{\alpha, \pm}^\dagger b_{\beta} b_{\gamma} + H.c. \right) + H_d . \tag{1}
\]

The first two terms describe phonon modes \(\alpha = (\text{band}, k)\) and coupling of the chosen IR circularly polarized optical phonons, with operators\textsuperscript{12} \(b_{\alpha, \pm}^\dagger = 2^{-1/2} \left( b_{\alpha, \pm}^\dagger + i b_{\alpha, \mp}^\dagger \right)\) and \(b_{\alpha, \pm} = 2^{-1/2} \left( b_{\alpha, \pm} + i b_{\alpha, \mp} \right)\), to the light intensity \(E^+(t)\) of the same polarization. The third term denotes decay of these IR phonons, with wave vectors \(k \approx 0\), into phonon pairs with opposite wave vectors \(\pm k\), which most likely come from the same acoustical branch\textsuperscript{13}. These also can not carry angular quasi-momentum \(L\), which is passed to the tube by umklapp processes. The resulting tube rotation is predominantly damped by scattering with molecules, as described in \(H_d\).\textsuperscript{14}

In Fig. 2, we show two (doubly degenerate) IR modes in the elementary cell, with 40 atoms, of the (10,10)
wires, which propagate uni-directionally on the tube circumference in phase with the light polarization.

The total number of the two degenerate linearly polarized modes forms a set of IR phonon modes, with the $A_{2u}$ and $E_{1u}$ symmetries. A circularly polarized light, $E^+$, excites phonon waves, which propagate uni-directionally on the tube circumference in phase with the light polarization.

nanotube. In the $A_{2u}$ and $E_{1u}$ modes, the atoms move out-of-plane and in-plane, respectively, orthogonal to the tube axis, as shown by open circles. Combination of the two degenerate linearly polarized modes forms a circularly polarized phonon mode, of either symmetry, which can absorb angular quasi-momentum from circularly polarized photons. The atomic displacements break the tube symmetry and induce electric dipoles ($\pm\cdot$), which follow in time the polarization of the circulating electric field $E^+$. The effect does not rely on coherent light and can be also realized in tubular rings (see Fig. 2).

As an example, we consider excitation of the $A_{2u}$ mode. The total number $n^+_{A_{2u}}$ of circularly polarized phonons, excited in the vicinity of $k = 0$, is given by the Boltzmann equation

$$\frac{\partial n^+_{A_{2u}}}{\partial t} = -\frac{n^+_{A_{2u}}}{\tau_{A_{2u}}} - \frac{n^+_{A_{2u};\text{equil}}}{\tau_{A_{2u}}}.$$  \hspace{1cm} (2)

$n^+_{A_{2u}}$ is their injection rate and $\tau_{A_{2u}} = 2\hbar/\gamma_{A_{2u}} \approx 2$ ps, is the relaxation time, where $\gamma_{A_{2u}} \approx 22$ cm$^{-1}$ is the width of the IR phonon lines in nanotubes. We neglect small populations $n^-_{A_{2u}}$ of phonons with the opposite polarization, resulting in scattering.

The absorption line of the $A_{2u}$ ($E_{1u}$) mode was observed near $\omega_{A_{2u}} = 870$ cm$^{-1}$ (1580 cm$^{-1}$) in both graphite and C nanotubes. In graphite, the $A_{2u}$ mode has an oscillator strength $f \approx 0.004$, which we assume to approximately hold in C nanotubes. Its optical dipole moment is $\mu_{A_{2u}} = e\sqrt{3}\hbar f/2m_{osc}\omega_{A_{2u}} \approx 10^{-31}$ Cm, where $m_{osc} = M_{\text{Carbon}}/2$ is the oscillator mass. Using the Fermi’s Golden rule, and assuming that $n^+_{A_{2u};\text{equil}} \approx 0$, we obtain the injection rate

$$n^+_{A_{2u}} \approx \frac{2\pi}{\hbar} |\mu_{A_{2u}}E^+|^2 \rho(\omega_{A_{2u}}),$$  \hspace{1cm} (3)

where $\rho(\omega_{A_{2u}})$ is the density of phonon modes at $k = 0$.

An armchair (10,10) nanotube of length $l = 1$ km has $n = 1.6 \times 10^5$ C atoms and $N = n/40 = 4000$ elementary cells ($A_{2u}$ modes with $k \neq 0$). About 10% of these modes (around $k = 0$) fall in the energy window $\gamma_{A_{2u}}$, thus giving the effective mode density $\rho(\omega_{A_{2u}}) \approx 400/\gamma_{A_{2u}}$.

For a field strength $E^+ = 10$ kV/m, we obtain from Eq. 3 that $n^+_{A_{2u}} \approx 2.5 \times 10^3$ s$^{-1}$. The IR phonons thus absorb the angular quasi-momentum with the rate $L_{A_{2u}} = \hbar n^+_{A_{2u}} \approx 2.5 \times 10^{-28}$ Nm.

We can understand the angular quasi-momentum umklapp processes by unrolling the nanotube, and loosely binding many such sheets into a superlattice of lattice constant $a_s = 2\pi r$, where $r$ is the tube diameter. Then, the IR phonons modes have the transversal wave vector $K_0 = 2\pi/a_s$, which falls in the middle of the second Brillouin mini-zone of size $Q = K_0$. In a two-phonon umklapp decay, the momentum conservation is $K_0 + K_1 + K_2 = Q$ (transversal wave vectors of the decayed acoustical phonons are $K_{1,2} \approx 0$), where the vector $Q$ interconnects centers of the first and second mini-zone. In the nanotube, we can vector multiply this identity by $h\tau$, and obtain the (umklapp) angular quasi-momentum conservation $L_0 + L_1 + L_2 = hQ \times r$ ($L_{1,2} = 0$), where $L_0 = hK_0 \times r \approx h$.

In Eq. 3 these processes are represented by the relaxation time $\tau_{A_{2u}}$, which could be derived from Eq. 3 following Klemens [3]. Its experimental value, $\tau_{A_{2u}} \approx 2$ ps, is in agreement with decay times of the suggested processes realized in other systems [4]. Since ab initio calculations of the phonon matrix elements $c_{\alpha\beta\gamma}$ are lacking, we use this value of $\tau_{A_{2u}}$ in our modeling. Eq. 2 then gives the steady-state angular quasi-momentum in the $A_{2u}$ phonon bath $L_{A_{2u}} = h\Delta n^+_{A_{2u}} = L_{A_{2u}}\tau_{A_{2u}} \approx 5.2 \times 10^{-41}$ J s.

The angular momentum is transferred to the tube body at the rate $\dot{L} \approx L_{A_{2u}}$. Nanotubes in liquids or under atmospheric conditions would rotate slowly, since collisions with the surrounding molecules quickly dissipate the acquired angular momentum. On the other hand, in low-vacuum environment, with realistic collisional rates $\kappa \approx 10^{-12} - 10^{-13}$ cm$^3$ s$^{-1}$, damping times of the order $\tau_{\text{damp}} \approx 10$ s are readily achievable. The tube thus keep a steady-state angular momentum $\dot{L} \approx L_{A_{2u}}\tau_{\text{damp}} \approx 2.5 \times 10^{-28}$ J s.

The nanotube rotation frequency $\omega_{\text{rot}}$ can be found upon calculating its principal moments of inertia $I_{\text{rot}}$ [21]

$$A = B = M \left( \frac{r^2 + r^2}{4} + \frac{l^2}{12} \right), \quad C = M \frac{r^2 + r^2}{2}.$$  \hspace{1cm} (4)

Here $M = \rho l$, $r_c$, $r_s$, and $l$ are the nanotube mass ($\rho$ is the linear density), exterior and interior radii, and length, respectively. For the (10,10) armchair nanotube with $r = (r_c + r_s)/2 \approx 0.68$ nm and $l = 1$ km, we obtain $M \approx 1.9 \times 10^{-20}$ kg and $A \approx 1.6 \times 10^{-33}$ kg m$^2 \approx 1.8 \times 10^5$ C.

Finally, we find the rotation speed $\omega_{\text{rot}} = L/C \approx 28$ GHz for this elementary nano-mechanical device. Centrifugal acceleration on its surface is enormous, $a = \cdots$
\[
\omega_{\text{con}} = \frac{C \omega_{\text{rot}}}{2A} \pm \sqrt{\left(\frac{C \omega_{\text{rot}}}{2A}\right)^2 + \frac{2S_0}{\rho}} \quad . (6)
\]

We can see that the modal frequencies depend on \(\omega_{\text{rot}}\) due to gyroscopic effects [20]. Since the ratio \(C/A \approx 1^{-2}\) is small, the effects are suppressed by the potential \(U\), so that \(\omega_{\text{con}} \approx \sqrt{2} \omega_{\text{cyl}}\). From Eq. (4) we find that they begin to play a role for tube lengths \(l < (r_c + r_i) \sqrt{3} \omega_{\text{rot}}/\omega_{\text{cyl}} \approx 130\) nm. If the trap is suddenly switched off, a micron-long nanotube rotating with frequency \(\omega_{\text{rot}} = 28\) GHz, and initially disturbed on its side, would precess with the frequency \(\omega_{\text{precess}} = C \omega_{\text{rot}}/A \approx 175\) kHz.

In long nanotubes, one needs to consider also flexural vibrations [18, 21]. The critical flexural frequencies \(\omega_f\) can be evaluated from the equations for lateral deflections \(x(z), y(z)\) at different points \(z\) along the trap axis, if the rigid body approximation is abandoned. The equation for the \(x\) deflection is

\[
Y \frac{\partial^4 x}{\partial z^4} = -\rho \frac{\partial^2 x}{\partial t^2} - S_0 x + \frac{\sigma^2}{C \omega_{\text{rot}}} \frac{\partial^2 y}{\partial z^2}, \quad \text{with} \quad Y = \frac{\pi}{12} r_c^4 \rho, \quad S_0 = \frac{\pi}{4} r_c^3 \rho. \quad \text{Eq. (7)}
\]

Here \(Y\) is the Young modulus, \(I = \pi (r_c^4 - r_i^4)/4\) is the second moment of nanotube cross-section and the factors \(\sigma^2 = 2a = 2\rho(r_c^3 + r_i^3)/2\) are the densities of the moments of inertia [19], which correspond to the bulk expressions in Eq. (4) in the limit \(l \to 0\). The equation for the \(y\) deflection results from Eq. (7) by exchanging \(x \leftrightarrow y\) and a negative sign in the last term.

The flexural frequencies correspond to the solutions \(x = x_0 \cos(\omega_f t), y = y_0 \sin(\omega_f t)\) in Eq. (4). This substitution gives an ordinary differential equation, identical for both the \(x\) and \(y\) deflections. For simplicity, we apply the clamped-end approximation, with the boundary conditions \(x_0(z) = \pm \frac{1}{2} \), \(d^2 x_0(z) = \pm \frac{1}{2}\) \(d^2 z = 0\). The solutions are \(x_0(z) = A_0 \cos(\xi z)\) or \(x_0(z) = A_0 \sin(\xi z)\), where \(\xi = \sqrt{\left(\alpha / \sqrt{a^2 - 4 \beta Y I / 2 Y I}\right) / \alpha} \approx \alpha \omega_f^2 - c \omega_{\text{rot}} \omega_f, \beta = S_0 - \rho \omega_f^2.\) Therefore, \(\xi = n \pi/1, \text{ with } n = 1, 2, 3, \ldots\) indexing the eigenmodes, which leads to the critical flexural frequencies \(\omega_{fn} = \omega_f\) 

\[
\omega_{fn} = \sqrt{\frac{S_0/\rho + (n \pi/4)^4 Y I / \rho}{1 - (n \pi/4)^2 / 2}}. \quad \text{(8)}
\]

We use the values \(Y \approx 5.5\) TPa and \(h = r_c - r_i = 0.066\) nm, found in molecular dynamics simulations [28].

In Fig. 3 we show the dependence of the lowest critical frequencies \(\omega_{fn}\) on the tube length \(l\), calculated from Eq. (8) using the numerical values for \(Y, h, \rho, S_0\) and \(r\). For long tubes, the frequencies \(\omega_{fn}\) coincide with \(\omega_{cyl}\), while for shorter tubes \((l < 1.3\) \(\mu\)m), the bending term surpasses the trap term, and \(\omega_{fn} = (n \pi)^4 \sqrt{Y I / \rho}\). In the continuum description, gyroscopic effects become only important for high eigenmodes \(n \approx 1/l\). In the inset of Fig. 3 we also show the dependence of \(\omega_{fn}\) on \(n\) for
tubes of different lengths. The huge Young modulus $Y$ makes the density of critical frequencies $\omega_{fn}$ relatively low, especially for short nanotubes. This allows for a rapid traversal to the “supercritical state”, which is realized above the flexural or other vibrational frequencies.

Rotating nanotubes could form parts of nano-motors, centrifuges or stabilizers. Centrifugal studies could be performed inside microtubes with large diameters $d \approx 10 \mu m$ [29] or in assemblies made from nanotube rings, forming strong but flexible skeletal coats. One could also think about possible applications of rotating tubes in liquids. Slowly rotating coiled nanotubes [30] could, for example, propel microscopic systems, which would chemically power the rotation of these tubes that attached to their surfaces in bearings [31], as in bio-motors. We believe that unique properties of nanotubes made from carbon and other materials could foster applications with rotating micro-elements.

We would like to thank R. Saito and G. Dresselhaus for data on IR phonon modes and several useful discussions. PK would like to acknowledge support from EU COMO. This work was also supported by the US National Science Foundation through a grant to the Institute for Theoretical Atomic and Molecular Physics at the Harvard-Smithsonian Center for Astrophysics.

FIG. 3. Dependence of the critical flexural frequencies $\omega_{fn}$ on the nanotube length $l$. The two thin horizontal dashed lines correspond to $\omega_{cyl}$ and $\omega_{con}$. In the inset, we show $\omega_{fn}$ as a function of the bending modal number $n$ for nanotubes of different length.

[1] S. Iijima, Nature 354, 56 (1991).
[2] M. S. Dresselhaus, G. Dresselhaus, and P. C. Eklund, Science of Fullerenes and Carbon Nanotubes (Academic Press Inc., San Diego, 1996).

[3] M. M. Treacy, T.W. Ebbesen and J. M. Gibson, Nature 381, 678 (1996); R. Gao et al., Phys. Rev. Lett. 85, 622 (2000).
[4] O. Lourie, D. M. Cox and H. D. Wagner, Phys. Rev. Lett. 81, 1638 (1998).
[5] D. Walz and S. R. Caplan, Biophys. J. 78, 626 (2000). F. A. Samatey et al., Nature 410, 331 (2001).
[6] J. Karczmarek, J. Wright, P. Corkum and M. Ivanov, Phys. Rev. Lett. 82, 3420 (1999); D. M. Villeneuve et al., Phys. Rev. Lett. 85, 542 (2000).
[7] E. Santamato, B. Daino, M. Romagnoli, M. Settembre and Y. R. Shen, Phys. Rev. Lett. 57, 2423 (1986).
[8] E. Higurashi et al., Appl. Phys. Lett. 64, 2209 (1994); L. Paterson et al., Science 292, 912 (2001).
[9] R. J. Nemanich, G. Lucovsky and S. A. Solin, Sol. St. Commun. 23, 117 (1977).
[10] J. Kastner et al., Chem. Phys. Lett. 221, 53 (1994); U. Kuhlmann, H. Jantoljak, N. Pfander, P. Bernier, C. Journet and C. Thomsen, Chem. Phys. Lett. 294, 237 (1998).
[11] J. Lin et al., Nature 385, 780 (1997).
[12] A. A. Kiselev, Opt. Spectrosc. 53, 469 (1982).
[13] P. G. Klemens, in Solid State Physics, ed. by F. Seitz and D. Turnbull, (Academic Press Inc., NY, 1958), Vol. 7; P. G. Klemens, Phys. Rev. 148, 845 (1966).
[14] S. Usher and G. P. Srivastava, Phys. Rev. B 50, 14179 (1994).
[15] P. Kráล and M. Shapiro, Phys. Rev. Lett. 86, 131 (2001).
[16] R. Saito (private communication).
[17] R. W. Boyd, Nonlinear Optics, (Academic Press, 1992).
[18] L. D. Landau and E. M. Lifshitz, Elasticity Theory (Pergamon, Oxford, 1986).
[19] R. N. Arnold and L. Maudner, Gyrodynamics and its Engineering Applications, (Academic, NY/London 1961).
[20] S. Whitley, Rev. Mod. Phys. 56, 41 (1984); ibid 56, 67 (1984).
[21] J. W. Beams, Sci. Am. 204, 134 (1961).
[22] T. Mashimo, S. Okazaki and S. Shihabazaki, Rev. Sci. Instrum. 67, 3170 (1996).
[23] Ch. L. Cheung, J. H. Hafner, T. W. Odom, K. Kim and C. M. Lieber, Appl. Phys. Lett. 76, 3136 (2000).
[24] S. Trasobares et al., Eur. Phys. J. B 22, 117 (2001).
[25] P. Kim and C. M. Lieber, Science 286, 2148 (1999).
[26] A. Ashkin, Phys. Rev. Lett. 24, 156 (1970); M. J. Renn, R. Pastor and H. J. Lewandowski, Phys. Rev. Lett. 82, 1574 (1999).
[27] L. X. Benedict, S. G. Louie, and M. L. Cohen, Phys. Rev. B 52, 8541 (1995).
[28] B. L. Yakobson, C. J. Brabec and J. Bernholc, Phys. Rev. Lett. 76, 2511 (1996).
[29] M. Remškar et al., Appl. Phys. Lett. 69, 351 (1996).
[30] X. B. Zhang et al., Europhys. Lett. 27, 141 (1994); A. Volodin et al., Phys. Rev. Lett. 84, 3342 (2000).
[31] A. N. Kolmogorov and V. H. Crespi, Phys. Rev. Lett. 85, 4727 (2000); J. Cumings and A. Zettl, Science 289, 602 (2000).