Laser spinning of nanotubes: A path to fast-rotating microdevices

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We show that circularly polarized light can spin nanotubes with GHz frequencies. In this method, angular momenta 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.

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Carbon nanotubes\textsuperscript{1} have unique mechanical and electronic properties with many potential applications.\textsuperscript{2} They possess a huge Young’s modulus \( Y \geq 1 \) TPa, which adjusts their autonomous mechanical oscillations to MHz frequencies.\textsuperscript{3} Moreover, their “molecular structures” remain naturally stable even at large deformations.\textsuperscript{4}

Therefore, rotationally symmetric structures based on stiff nanotubes could form ideal \textit{piston rods} for nanoscale applications. In contrast to chemically driven biomotors,\textsuperscript{5} spinning with Hertz 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 \textit{synchronously} rotated with a dipolar laser trap, which accelerates its angular velocity. Larger molecules\textsuperscript{7} and microparticles\textsuperscript{8} can be rotated by absorption of angular momentum from circularly polarized or “twisted” laser beams. Nanotubes are excellent candidates for this \textit{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 \textit{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) \( A_{2u} \) or \( E_{1u} \) phonon modes, active in graphite\textsuperscript{9} and nanotubes.\textsuperscript{10}

In Fig. 1 we show two schemes for spinning nanotubes. In the upper one, circularly polarized light beam propagates along the symmetry axis of the single-wall nanotube (SWNT) or multiwall nanotube (MWNT), levitating in an optical trap. The photon angular momentum is transferred to \textit{circularly polarized} phonons, counterpropagating on the tube circumference (see Fig. 2), and latter passed by scattering to the tube body. The angular momentum of light could be also directly passed to the nanotube in excitation of its dense rotational levels. The resulting tube rotation with angular frequency \( \omega_{\text{rot}} \) is mostly balanced 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

\[
H = \sum_{a} \hbar \omega_{a} b_{a}^{\dagger} b_{a} + \sum_{\alpha \zeta} \mu_{\alpha \zeta} E^{\pm}(t) (b_{\alpha \zeta}^{\dagger} + b_{\alpha \zeta}) + \sum_{\alpha \zeta, \beta \gamma} (c_{\alpha \zeta, \beta \gamma} b_{\alpha \zeta}^{\dagger} b_{\beta \gamma} + \text{H.c.}) + H_{d}.
\]

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

In Fig. 2, we show two (doubly degenerate) IR modes in the elementary cell, with 40 atoms, of the (10,10) nanotube. In the \( A_{2u} \) and \( E_{1u} \) modes, the atoms move out of plane and in plane, respectively, orthogonal to the tube axis,\textsuperscript{16} as shown by open circles. Combination of the two degenerate linearly polarized modes form a \textit{circularly polarized} phonon mode.
of either symmetry, which can absorb angular quasimomentum from circularly polarized photons. The atomic displacements break the tube symmetry and induce electric dipoles \((+-)\), 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. 1).

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

\[
\frac{dn_{A_{2u}}^+}{dt} = n_{A_{2u}}^+ - n_{A_{2u}}^+\textit{equil.},
\]

\(n_{A_{2u}}^+\) 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.\(^{10}\) 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\(^5\) \(f=0.004\), which we assume to approximately hold in C nanotubes. Its optical dipole moment is\(^{17}\) \(\mu_{A_{2u}} = e\sqrt{3\hbar/2m_{osc}}\omega_{A_{2u}} = 10^{-31}\) Cm, where \(m_{osc} = M_{\text{Carbon}}/2\) is the oscillator mass. Using the Fermi’s golden rule, and assuming that \(n_{A_{2u}}^+\textit{equil.} = 0\), we obtain the injection rate

\[
\dot{n}_{A_{2u}}^+ = \frac{2\pi}{\hbar} |\mu_{A_{2u}} E^+|^2 \rho(\omega_{A_{2u}}),
\]

where \(\rho(\omega_{A_{2u}})\) is the density of phonon modes at \(k=0\). An armchair \((10,10)\) nanotube of length \(l=1\) \(\mu\)m has \(n \approx 1.6 \times 10^5\) 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 \(\gamma_{A_{2u}}\approx 2\) to give the effective mode density \(\rho(\omega_{A_{2u}}) \approx 400/\gamma_{A_{2u}}\). For a field strength \(E^+ = 10\) kV/m, we then obtain from Eq. (3) that \(n_{A_{2u}}^+\approx 2.5 \times 10^3\) s\(^{-1}\). The IR phonons thus absorb the angular quasimomentum with the rate \(\dot{L}_{A_{2u}} = \hbar n_{A_{2u}}^+ \approx 2.5 \times 10^{-29}\) Nm.

We can understand the angular quasimomentum Umklapp processes by unrolling the nanotube, and loosely binding many such sheets into a superlattice of lattice constant \(a_z = 2\pi r\), where \(r\) is the tube diameter. Then, the IR phonons modes have the transversal wave vector \(K_0 = 2\pi/l_{z}\), which falls in the middle of the second Brillouin minizon 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 \(K_{1,2} = 0\)), where the vector \(Q\) interconnects centers of the first and second minizon. In the nanotube, we can vector multiply this identity by \(hr\) and obtain the (Umklapp) angular quasimomentum conservation \(L_0 + L_1 + L_2 = hQ \times r\) \((L_{1,2} = 0)\), where \(L_0 = hK_0 \times r = \hbar\).

Here \(M = \rho l, r_1, r_1,\) 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 \(r = (r_1 + r_2)/2 \approx 0.68\) nm and \(l = 1\) \(\mu\)m, 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 \(\omega_{rot} = L/C \approx 28\) GHz for this elementary nanomechanical device. Centrifugal acceleration on its surface is enormous, \(a = r\omega_{rot}^2 \approx 0.5 \times 10^{12}\) m/s\(^2\) \(\approx 10^{11}\) g. This value surpasses by two orders of magnitude the acceleration obtained with sub-millimeter steel balls,\(^{21}\) and by five orders of magnitude acceleration in the fastest centrifuges.\(^{22}\) Since for \(a = 10^{11}\) g, the force on each C atom, \(F = 13\) \(\mu\)eV/\(\AA\) is still negligible with respect to chemical forces (1 eV/\(\AA\)), the tube rotation could be
further increased. On this path to “teragravity,” unique parameters of nanotubes can play a pivotal role.

We can now discuss in more details practical spinning experiments. Isolated SWNT or MWNT have been grown, for example, on an atomic force microscope tip, which can be later placed inside an optical trap. The nanotube can be severed from the tip using, for instance, a focused electron beam. Detached tubes could be also transported to the trap by recently developed nanotweezers. The optical trap can be formed by two linearly and mutually parallel polarized, counterpropagating laser beams.

Nonresonant scattering of trap-beam photons from the nanotube with polarizability $\alpha$ produces a force, oriented in the direction of increasing light intensity $I$, that results in the potential $U = -\alpha I/2c$ ($c$ is the speed of light). The longitudinal $\alpha_{z} \approx 500 \text{ Å}^2/\text{atom}$ and radial $\alpha_{r} \approx 25 \text{ Å}^2/\text{atom}$ static polarizabilities of semiconducting nanotubes are quite different, and this difference is even larger in metallic tubes. Therefore, the tube in the trap remains oriented along the beam polarization axis, where it experiences the trapping potential

$$U = -U_{0}e^{-\sqrt{2\alpha_{z} I/x^2}} - (U_{0} - S r^2)e^{-\sqrt{2\alpha_{r} I/x^2}}.$$  \hfill (5)

Here $x,y,z$ ($r = x^2 + y^2$) are the tube center-of-mass coordinates, and $S = S_{0}I = U_{0}/\alpha_{z}^2$ is the trap rigidity. To prevent thermal escape of the tube from the trap, we consider a trap depth $U_{0} = n \alpha_{z} I/2c \approx 10$ eV, and obtain $I \approx 1.2 \text{ GW/cm}^2$.

The trap laser frequency must be below the band gap $E_{g} \approx 1$ eV, and away from the frequencies of the tube internal modes, see below. Assuming that $\sigma_{r} \approx 1 \mu$m, we find $S_{0} = U_{0}/11\sigma_{r}^2 = 1.6 \text{ J/m}^3$.

Small amounts of defects and adsorbants on the tube walls do not prevent its spinning, but can shift its rotation frequency. In accordance with the De Laval principle of self-balancing, such a partially coated nanotube floating in the trap would rotate around an eccentric axis. Rapid rotation of the nanotube can be also limited by its mechanical vibrations in the trap, as discussed below. To avoid its large oscillations, the critical frequencies should be quickly passed during the acceleration.

The cylindrical whirl mode reflects the rigid-body vibrations of the tube orthogonal to the trap axis. The forward (backward) cylindrical frequencies are $\omega_{cyl} = \pm \sqrt{S_{0}/\rho} \approx \pm 9.2 \text{ MHz}$. In the conical whirl mode, the tube ends move in opposite directions with respect to the tube-trap axis. For a tube distorted through the angle $\theta$, the torsional moment is $M_{\theta} \approx \rho L \theta^2$, resulting in the Euler’s equation, $A\omega_{cyl}^2 = C\omega_{rot}^2 + S_{0}/L$, Using $A = \rho L^2/12$, valid for $L \gg r$, we obtain

$$\omega_{con} = \frac{C\omega_{rot}}{2A} \pm \sqrt{\left(\frac{C\omega_{rot}}{2A}\right)^2 + \frac{2S_{0}}{\rho}}.$$  \hfill (6)

We can see that the modal frequencies depend on $\omega_{rot}$ due to gyroscopic effects. Since the ratio $C/A \approx L^{-2}$ is small, the effects are suppressed by the potential $U$, so that $\omega_{con} \approx \sqrt{2\omega_{cyl}}$. From Eq. (6), we find that they begin to play a role for tube lengths $l < (r_{e} + r_{i}) \sqrt{3\omega_{rot}/\omega_{cyl}} \approx 130 \text{ nm}$. If the trap is suddenly switched off, a micron-long nanotube rotating with frequency $\omega_{rot} = 28 \text{ GHz}$, and initially disturbed on its side, would precess with the frequency $\omega_{pre} = C\omega_{rot}/L \approx 175 \text{ kHz}$.

In long nanotubes, one needs to consider also flexural vibrations. 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

$$YI \frac{\partial^2 x}{\partial z^2} = -\rho \frac{\partial^2 x}{\partial t^2} - S_{0}x + a \frac{\partial^2 x}{\partial z^2} + c \omega_{rot} \frac{\partial}{\partial t} \frac{\partial^2 y}{\partial z^2},$$  \hfill (7)

Here $Y$ is the Young’s modulus, $I = \pi(r_{e}^4 - r_{i}^4)/4$ is the second moment of nanotube cross section, and the factors $c = 2a = \rho(r_{e}^2 + r_{i}^2)/2$ are the densities of the moments of inertia, which correspond to the bulk expressions in Eq. (4) in the limit $l \rightarrow 0$. 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. (7). 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 $x_{0}(z = \pm l/2) = d\delta x_{0}(z = \pm l/2)/dz = 0$. The solutions are $x_{0}(z) = A_{0}\cos(\xi z)$ or $x_{0}(z) = A_{0}\sin(\xi z)$, where $\xi^2 = (\alpha^2 + \sqrt{\alpha^2 - 4BYI}/2YI)$, $\alpha = a_{rot} - c\omega_{rot}f_{w}$, and $B = S_{0} - \rho\alpha_{f}^2$. Therefore, $\xi = n\pi/l$, with $n = 1,2,3,\ldots$ indexing the eigenmodes, which leads to the critical flexural frequencies $(\omega_{fn} = \omega_{rot})$

$$\omega_{fn} = \sqrt{S_{0}/\rho + (n\pi/l)^2YI/\rho} / \left(1 - (n\pi/l)^2/2\right).$$  \hfill (8)

We use the values $Y \approx 5.5 \text{ TPa}$ and $h = r_{e} - r_{i} \approx 0.066 \text{ nm}$, found in molecular-dynamics simulations.

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 $\omega_{fn} = (n \pi/l)^2 \sqrt{Y/\rho}$. In the continuum description, gyroscopic effects become only important for high eigenmodes $n \approx l/r$. In the inset of Fig. 3, we also show the dependence of $\omega_{fn}$ on $n$ for tubes of different lengths. The huge Young’s 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 nanomotors, centrifuges or stabilizers. Centrifugal studies could be performed inside microtubes with large diameters $d \approx 10$ $\mu$m (Ref. 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 could, for example, propel microscopic systems, which would chemically power the rotation of these tubes that attached to their surfaces in bearings, as in biomotors. We believe that unique properties of nanotubes made from carbon and other materials could foster applications with rotating microelements.

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