Science Advances
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advances.sciencemag.org/cgi/content/full/7/2/eabd6705/DC1

Supplementary Materials for

Rotational Doppler cooling and heating

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Published 6 January 2021, *Sci. Adv.* **7**, eabd6705 (2021)
DOI: 10.1126/sciadv.abd6705

This PDF file includes:

Supplementary text

I. POLARIZABILITIES OF ROTATING PARTICLES

In this note, we formulate a classical model, which is capable of describing the optical polarizabilities of rotating particles of various geometries. By using this model, we successfully derive the optical polarizabilities of three distinctly different types of particles, namely, nanorods, nanocrosses, and nanodisks, which we assume to be rotating as shown in fig. S1. The results reassuringly satisfy general rules such as the optical theorem, as well as conservation of energy and angular momentum.

As shown in fig. S1, we further assume the particles to be thin along the z direction, around which they are taken to be rotating with angular velocity $\vec{\Omega}$. The rotation of the particle is manifested by the motion of the ionic potential in the material of which they are composed, which also imposes the rotation onto the conduction electrons. The latter are in turn assumed to completely mediate the interaction with external light. For the nanorod and nanocross in fig. S1 (B and C), the rotation is clearly observed from their continuously changing surface boundaries, while for axially symmetric particles, such as the nanodisk in fig. S1D, the rotation corresponds to a collective motion of electrons and ions with nonzero total angular momentum.

We determine the optical responses of these rotating particles from the internal dynamics of conduction electrons in the x-y plane under external light irradiation. In our model, we represent such oscillating electrons by a single point particle of effective charge Q and mass m subject to the classical equation of motion

$$m\ddot{\mathbf{r}} = -m\omega_0^2\mathbf{r} - m\gamma(\dot{\mathbf{r}} - \Omega r\hat{\varphi}) + m\tau\dddot{\mathbf{r}} + Q\mathbf{E}_\pm + \mathbf{F}^{\text{react}}, \tag{S1}$$

where $\mathbf{r}$ is the radial distance relative to the particle center in the lab frame, ω_0 is the intrinsic oscillator resonance frequency, γ is a phenomenological damping rate capturing the internal dissipation of electron oscillation energy, the dissipation force is proportional to the velocity with respect to the center of mass of the ionic background $\dot{\mathbf{r}} - \Omega r\hat{\varphi}$, $\mathbf{F}^{\text{react}}$ is the force imposed by the boundary defined by the particle geometry (i.e., the trapping ionic potential), and the Abraham-Lorentz force $m\tau\dddot{\mathbf{r}}$ with $\tau = 2Q^2/3mc^3$ introduces corrections due to radiation reaction.

As illustrated in fig. S1A, we represent the position vector in Cartesian coordinates as $\mathbf{r} = x\hat{\mathbf{x}} + y\hat{\mathbf{y}}$ in the lab frame and $\mathbf{r} = x'\hat{\mathbf{x}}' + y'\hat{\mathbf{y}}'$ in the frame rotating with the particle (primed variables refer to the rotating frame in what follows), with their respective unit vectors related through the transformation

$$\hat{x}' = \cos(\Omega t)\hat{x} + \sin(\Omega t)\hat{y}, \quad \hat{y}' = -\sin(\Omega t)\hat{x} + \cos(\Omega t)\hat{y}.$$

Using these relations, the transformation of the time derivative of any vector $\vec{v}$ from the lab frame to the rotating frame is given by $\dot{\vec{v}} = \dot{\vec{v}'} + \vec{\Omega} \times \vec{v}$. We can then apply this expression recurrently to find the relation between different orders of the time derivative of the position vector $\mathbf{r}$ in the lab and rotating frames as

$$\begin{aligned}
&\mathbf{r} = \mathbf{r}', \quad \dot{\mathbf{r}} = \dot{\mathbf{r}}' + \vec{\Omega} \times \mathbf{r}', \quad \ddot{\mathbf{r}} = \ddot{\mathbf{r}}' + 2\vec{\Omega} \times \dot{\mathbf{r}}' - \Omega^2\mathbf{r}', \\
&\dddot{\mathbf{r}} = \dddot{\mathbf{r}}' + 3\vec{\Omega} \times \ddot{\mathbf{r}}' - 3\Omega^2\dot{\mathbf{r}}' - \Omega^2(\vec{\Omega} \times \mathbf{r}'),
\end{aligned}$$

where we apply the fact that $\vec{\Omega}$ is perpendicular to $\mathbf{r}$. In this note, we use complex notation for time-harmonic quantities. We are interested in the optical response of the rotating particles under circularly polarized light. We write the electric field of right- (RCP, upper signs) and left-circularly polarized (LCP, lower signs) light in the lab frame as $\mathbf{E}_\pm = E_\pm e^{-i\omega t}\hat{\mathbf{u}}_\pm$ in terms of the corresponding unit vectors $\hat{\mathbf{u}}_\pm = (\hat{\mathbf{x}} \pm i\hat{\mathbf{y}})/\sqrt{2}$. In the rotating frame, the unit polarization vector $\hat{\mathbf{u}}_\pm$ is transformed into $\hat{\mathbf{u}}'_\pm = (\hat{\mathbf{x}}' \pm i\hat{\mathbf{y}}')/\sqrt{2}$ and the electric field thus becomes $\mathbf{E}_\pm = E_\pm e^{-i\omega_\mp t}\hat{\mathbf{u}}'_\pm$, which introduces a Doppler shift in

the light frequency according to $\hat{\mathbf{u}}_\pm = \mathrm{e}^{\pm i\Omega t}\hat{\mathbf{u}}'_\pm$. Together with the transformation of the time derivatives described above, we can explicitly rewrite Eq. (S1) in the rotating frame as

$$\ddot{x}' = -(\omega_0^2 - \Omega^2)x' - 2\Omega\dot{y}' - \gamma\dot{x}' + \tau(\dddot{x}' - 3\Omega\ddot{y}' - 3\Omega^2\dot{x}' + \Omega^3 y') + \frac{1}{m}(Q\mathbf{E}_\pm + \mathbf{F}^{\mathrm{react}})\cdot\hat{\mathbf{x}}', \tag{S2}$$

$$\ddot{y}' = -(\omega_0^2 - \Omega^2)y' + 2\Omega\dot{x}' - \gamma\dot{y}' + \tau(\dddot{y}' + 3\Omega\ddot{x}' - 3\Omega^2\dot{y}' - \Omega^3 x') + \frac{1}{m}(Q\mathbf{E}_\pm + \mathbf{F}^{\mathrm{react}})\cdot\hat{\mathbf{y}}', \tag{S3}$$

where the intrinsic resonance frequency $\sqrt{\omega_0^2 - \Omega^2}$ captures the effect of the centrifugal force, whereas the Coriolis force (second term on the right) directly couples the velocities in the two orthogonal directions. In what follows, we apply this model to calculate the optical response of rotating particles in different geometries.

A. Nanorod

Equation of motion. As shown in fig. S1B, we assume that the rotating nanorod to be always oriented along $\hat{\mathbf{x}}'$. This implies that the reaction force $\mathbf{F}^{\mathrm{react}}$ is always oriented along $\hat{\mathbf{y}}'$. Noticing that the frequency of incident circularly polarized light becomes $\omega_\mp$ in the rotating frame, we write the position vector of the effective charge as $\mathbf{r}' = \rho\mathrm{e}^{-i\omega_\mp t}\hat{\mathbf{x}}'$, whose time evolution is readily determined by solving Eq. (S2) as

$$\rho = QE_\pm/\sqrt{2}md_\mp,$$

where

$$d_\mp = \omega_0^2 - \Omega^2 - \omega_\mp(\omega_\mp + i\gamma) - i\tau\omega_\mp(\omega_\mp^2 + 3\Omega^2).$$

The position vector then becomes

$$\begin{aligned}\mathbf{r} &= \frac{Q}{\sqrt{2}md_\mp}E_\pm\mathrm{e}^{-i\omega_\mp t}\hat{\mathbf{x}}' \\ &= \frac{Q}{2md_\mp}E_\pm\mathrm{e}^{-i\omega t}(\hat{\mathbf{u}}_\pm + \hat{\mathbf{u}}_\mp\mathrm{e}^{\pm 2i\Omega t}).\end{aligned} \tag{S4}$$

Interestingly, in addition to the oscillatory component with the same frequency as the incident light, the coordinate vector $\mathbf{r}$ also contains components with shifted frequency $\omega \mp 2\Omega$ in the lab frame. With $\mathbf{r}$ explicitly given by Eq. (S4), we now calculate the time-averaged powers due to each of the force terms in Eq. (S1), which lead to different types of cross sections, as we discuss next.

Extinction. The extinction cross section of the particle σ_{ext} is determined by the time-averaged power of the work done by the light electric field, $P_{\mathrm{ext}} = \langle Q\mathbf{E}\cdot\dot{\mathbf{r}}\rangle$, which does not receive contributions from the components of $\mathbf{r}$ with shifted frequencies. With the light intensity written as $I = c|E_\pm|^2/8\pi$ (notice that we use the convention $\mathbf{E}^{\mathrm{ext}}(t) = \mathrm{Re}\{\mathbf{E}^{\mathrm{ext}}\mathrm{e}^{-i\omega t}\}$ for the time dependence of light of frequency ω), we find

$$\sigma_{\mathrm{ext}} = \frac{P_{\mathrm{ext}}}{I} = \frac{2\pi Q^2\omega}{mc}\mathrm{Im}\{\frac{1}{d_\mp}\}. \tag{S5}$$

Scattering. Scattered light is extracted from the electron oscillation through the work done by the Abraham-Lorentz force (the third term in Eq. (S1)). The scattering power is then given by $P_{\mathrm{sca}} =$

$-m\tau \langle \dddot{\mathbf{r}} \cdot \dot{\mathbf{r}} \rangle$, which readily leads to a total scattering cross section

$$\sigma_{\rm sca} = \frac{P_{\rm sca}}{I} = \frac{\pi Q^2}{mc} \frac{1}{|d_\mp|^2} \tau[\omega^4 + (\omega \mp 2\Omega)^4]. \tag{S6}$$

Alternatively, this expression for the scattering cross section can also be found by calculating the emission of the induced electric dipole in the rotating nanorod, $\mathbf{p} = Q\mathbf{r} = \mathbf{p}_\omega + \mathbf{p}_{\omega\mp 2\Omega}$, where $\mathbf{p}_\omega$ and $\mathbf{p}_{\omega\mp 2\Omega}$ are contributed by the two terms in Eq. (S4) associated with frequencies ω and $\omega \mp 2\Omega$, respectively. As the incident light frequency is ω, the emissions produced by $\mathbf{p}_\omega$ and $\mathbf{p}_{\omega\mp 2\Omega}$ correspond to elastic and inelastic scattering, respectively. The corresponding radiated powers are obtained using well-established expressions relating them to the emitting dipoles: $P_\omega = \omega^4 |\mathbf{p}_\omega|^2/3c^3$, which results in an elastic scattering cross section $\sigma_\omega = \pi Q^2 \tau \omega^4/mc|d_\mp|^2$, consistent with the first term in Eq. (S6); and similarly, $P_\omega = (\omega \mp 2\Omega)^4 |\mathbf{p}_{\omega\mp 2\Omega}|^2/3c^3$, which leads to the inelastic scattering cross section $\sigma_{\omega\mp 2\Omega} = \pi Q^2 \tau(\omega \mp \Omega)^4/mc|d_\mp|^2$, corresponding to the second term in Eq. (S6).

Absorption. Among the remaining terms in Eq. (S1), only the second term $-m\gamma(\dot{\mathbf{r}} - \Omega r\hat{\varphi})$ (i.e., dissipation) and the reaction force $\mathbf{F}^{\rm react}$ produce a nonzero contribution to the time-averaged power. For the nanorod, as we show below, the dissipation force $-m\gamma(\dot{\mathbf{r}} - \Omega r\hat{\varphi})$ does not contribute a time-averaged torque exerted on the particle, so it only acts by transferring energy from electron oscillations to thermal degrees of freedom. The reaction force $\mathbf{F}^{\rm react}$ directly changes the rotation of the ionic potential, and therefore, it alters the mechanical energy. Importantly, the reaction force $\mathbf{F}^{\rm react}$ disappears in the nanodisk, as it contains freely moving electrons, and therefore, the dissipation force also changes the mechanical energy of the particle.

The time-averaged power due to the dissipation force is $P_{\rm dis} = \langle m\gamma(\dot{\mathbf{r}} - \Omega r\hat{\varphi}) \cdot \dot{\mathbf{r}} \rangle$, where $\mathbf{r} = \rho e^{-i\omega_\mp t}\hat{\mathbf{x}}'$ and, for the rotating nanorod, we have $\hat{\varphi} = \hat{\mathbf{y}}'$. By expressing $\hat{\mathbf{y}}'$ in the lab frame as in Eq. (S4), we can finally obtain $P_{\rm dis}$ and the corresponding dissipation cross section

$$\sigma_{\rm dis} = \frac{2\pi Q^2}{mc} \frac{1}{|d_\mp|^2} \gamma\omega_\mp^2. \tag{S7}$$

To find the power and cross section of the energy converted to mechanical motion, we first need to determine the reaction force $\mathbf{F}^{\rm react}$ acting on the electrons. Since $\mathbf{F}_{\rm react}$ is always along $\hat{\mathbf{y}}'$, by setting all terms related to y' to zero, we find from Eq. (S3)

$$\mathbf{F}^{\rm react} = \mp \frac{QE_\pm}{\sqrt{2}d_\mp} [i(\omega_0^2 - \omega^2) + \gamma\omega_\mp + \tau(\omega \mp 2\Omega)^3] e^{-i\omega_\mp} \hat{\mathbf{y}}'.$$

The power $P_{\rm react} = -\langle \mathbf{F}^{\rm react} \cdot \dot{\mathbf{r}} \rangle$ describes the part of energy that is extracted from electron oscillations and converted into mechanical motion. We find its corresponding cross section to be

$$\sigma_{\rm react} = \pm \frac{2\pi Q^2}{mc} \frac{1}{|d_\mp|^2} \Omega[\gamma\omega_\mp + \tau(\omega \mp 2\Omega)^3]. \tag{S8}$$

The absorption cross section should be the sum of dissipation and mechanical cross sections, that is,

$$\sigma_{\rm abs} = \sigma_{\rm dis} + \sigma_{\rm react} = \frac{2\pi Q^2}{mc} \frac{1}{|d_\mp|^2} [\gamma\omega_\mp\omega \pm \tau\Omega(\omega \mp 2\Omega)^3]. \tag{S9}$$

Effective polarizability. The rotating nanorod is not optically isotropic, so that in the lab frame it sustains a dipole of shifted frequency $\mathbf{p}_{\omega\mp 2\Omega}$, and consequently, the total dipole $\mathbf{p}$ is not simply proportional to the incident electric field vector $\mathbf{E}$. However, we still can define an effective polarizability of the rotating nanorod, which relates the electric field to the elastic electric dipole moment $\mathbf{p}_\omega = \alpha_\pm(\omega)\mathbf{E}_\pm$,

$$\alpha_\pm(\omega) = \frac{Q^2}{2m} \frac{1}{d_\mp}.$$

With this effective polarizability, the extinction cross section in Eqs. (S5) and the elastic scattering cross section shown by the first term in Eq. (S6) can be simply written in the well-known forms $\sigma_{\rm ext} = 4\pi k {\rm Im}\{\alpha_\pm(\omega)\}$ and $\sigma_\omega = 8\pi k^4 \left|\alpha_\pm(\omega)\right|/3$, respectively. In addition, the inelastic cross section (second term in Eq. (S6)) can similarly be written as $\sigma_{\omega\mp2\Omega} = 8\pi(\omega\mp2\Omega)^4\left|\alpha_\pm(\omega)\right|/3c^4$.

Energy and angular momentum conservation. Energy conservation is implied in Eq. (S1), since the different powers discussed above are calculated for each of the terms in Eq. (S1) yielding a nonzero time-averaged contribution. Energy conservation, simply stated as $P_{\rm ext} = P_{\rm sca} + P_{\rm abs}$, can be equivalently written in terms of the cross sections as $\sigma_{\rm ext} = \sigma_{\rm sca} + \sigma_{\rm abs}$, which is rigorously satisfied when inserting the expressions given in Eqs. (S5), (S6), and (S9).

Conservation of angular momentum in the system leads to an optical torque acting on the particle. Following similar procedures as for the calculation of different powers, we now rigorously evaluate the time-averaged torque contributed by each terms in Eq. (S1) according to $\langle \mathbf{r}\times\mathbf{F}\rangle$ for forces $\mathbf{F}$ given by $Q\mathbf{E}$, $-m\gamma(\dot{\mathbf{r}}-\Omega r\hat{\phi})$, $m\tau\dddot{\mathbf{r}}$, and $\mathbf{F}^{\rm react}$. Using the relation $\hat{\mathbf{u}}_\pm\times\hat{\mathbf{u}}_\mp - = \mp i\hat{\mathbf{z}}$, we find, after some straightforward algebra, all torque components to reduce to

$$\mathbf{M}_{\rm ext} = \pm\frac{Q^2}{4m}|E_\pm|^2{\rm Im}\{\frac{1}{d_\mp}\}\hat{\mathbf{z}}, \tag{S10a}$$

$$\mathbf{M}_{\rm dis} = 0, \tag{S10b}$$

$$\mathbf{M}_{\rm sca} = \mp\frac{Q^2}{8m}\frac{|E_\pm|^2}{|d_\mp|^2}[\tau\omega^3 - \tau(\omega\mp2\Omega)^3]\hat{\mathbf{z}}, \tag{S10c}$$

$$\mathbf{M}_{\rm react} = \mp\frac{Q^2}{4m}\frac{|E_\pm|^2}{|d_\mp|^2}[\gamma\omega_\mp + \tau(\omega\mp2\Omega)^3]\hat{\mathbf{z}}, \tag{S10d}$$

where $\mathbf{M}_{\rm ext}$ is the driving torque imparted by the incident light, whereas $\mathbf{M}_{\rm dis}$, $\mathbf{M}_{\rm sca}$, and $\mathbf{M}_{\rm react}$ are the frictional torques produced by dissipation, Abraham-Lorentz, and boundary-reaction forces, respectively. Because of Eq. (S1), the total torque acting on the electrons must be zero, so any change in angular momentum must be transferred to either the lattice or radiation. Additionally, the torque acting on the particle must nearly entirely end up in the ions because of their large mass compared with the electrons. Therefore, the reactions of both $\mathbf{M}_{\rm dis}$ and $\mathbf{M}_{\rm react}$ contribute to the torque acting on the ions $\mathbf{M} = -(\mathbf{M}_{\rm dis} + \mathbf{M}_{\rm react})$. Reassuringly, the power transferred to the ions as a result of this torque (i.e., $P_{\rm react} = \langle\mathbf{M}\rangle\cdot\vec{\Omega}$) is fully consistent with Eq. (S8).

We now provide an intuitive explanation of the total torque $\mathbf{M}$ acting on the particle. The total input number of photons can be directly written in terms of the extinction cross section as $N = \sigma_{\rm ext}I/\hbar\omega$, and considering that each circularly polarized photon carries angular momentum $\pm\hbar$, the total input angular momentum is $M_{\rm ext} = \pm\sigma_{\rm ext}I/\omega$, which is consistent with Eqs. (S10). Similarly, absorption directly results in transfer of angular momentum from photons to the particle, thus producing a torque $\pm\sigma_{\rm abs}I/\omega$. In contrast, elastic scattering maintains the angular momentum of the incident photons. As shown in Eq. (S4), the inelastic dipole $\mathbf{p}_{\omega\mp2\Omega}$ reverses the chirality of the incident light, so each scattered photon carries angular momentum of $\mp\hbar$, in contrast to the incident photons with angular momentum $\pm\hbar$. Taking into account the energy shift of inelastically emitted photons, we find the torque due to inelastic scattering to be $\pm2I\sigma_{\omega\mp2\Omega}\omega_\mp/\omega(\omega\mp2\Omega)$. Putting these results together, the total torque acting on the particle therefore becomes

$$\mathbf{M} = \pm I\left(\frac{\sigma_{\rm ext}}{\omega} + \frac{\sigma_{\omega\mp2\Omega}}{\omega}\frac{2\omega_\mp}{\omega\mp2\Omega}\right)\hat{\mathbf{z}}, \tag{S11}$$

which reproduced the result obtained based on Eqs. (S10).

B. Nanocross

Equation of motion. We consider a thin rotating nanocross formed by two perpendicular nanorods, which are fixed along directions $\hat{x}'$ and $\hat{y}'$, as shown in fig. S1C. Compared with the nanorod, the nanocross is optical isotropic, because it sustains two orthogonal degenerated dipolar modes. We denote the two dipoles along $\hat{\mathbf{x}}'$ and $\hat{\mathbf{y}}'$ as $\mathbf{p}_1$ and $\mathbf{p}_2$, respectively, and assume that the effective charge in each of the dipoles is Q, evolving according to the position vectors, $\mathbf{r}_1 = \rho_1 \mathrm{e}^{-i\omega_\mp t}\hat{x}'$ and $\mathbf{r}_2 = \rho_2 \mathrm{e}^{-i\omega_\mp t}\hat{y}'$. To find the optical response of the nanocross, we apply Eq. (S4) to both $\mathbf{p}_1$ and $\mathbf{p}_2$. The two charges are confined to the two perpendicular branches of the cross, so the direct velocity coupling described by the second term on the right of Eq. (S4) disappears. However, the two dipoles are not totally independent, since the reactive Abraham-Lorentz force produces coupling due to interference of the far-field radiation associated with each of them. For a circularly polarized incident field $\mathbf{E}_\pm = E_\pm \mathrm{e}^{-i\omega_\mp t}\hat{\mathbf{u}}'_\pm$, the equations of motion of the two charges reduce to

$$\rho_1(\omega_0^2 - \Omega^2 - \omega_\mp^2 - i\omega_\mp\gamma) - i\tau(\omega_\mp^3\rho_1 - 3i\Omega\omega_\mp^2\rho_2 + 3\Omega^2\omega_\mp\rho_1 + \Omega^3\rho_2) = \frac{Q}{\sqrt{2}m}E_\pm,$$
$$\rho_2(\omega_0^2 - \Omega^2 - \omega_\mp^2 - i\omega_\mp\gamma) - i\tau(\omega_\mp^3\rho_1 + 3i\Omega\omega_\mp^2\rho_1 + 3\Omega^2\omega_\mp\rho_2 - \Omega^3\rho_1) = \pm\frac{Q}{\sqrt{2}m}E_\pm,$$

as obtained from Eqs. (S2) and (S3) for $\mathbf{p}_1$ and $\mathbf{p}_2$, respectively.

The solution of these equations is readily found to be $\rho_1 = QE_\pm/\sqrt{2}mb_\mp$ and $\rho_2 = \pm i\rho_1$, where

$$b_\mp = \omega_0^2 - \Omega^2 - \omega_\mp(\omega_\mp + i\gamma) - i\tau\omega^3.$$

The electric dipole in the nanocross is a superposition of the two components along the orthogonal directions of the cross, that is,

$$\mathbf{p} = Q(\mathbf{r}_1 + \mathbf{r}_2) = \frac{Q^2}{\sqrt{2}mb_\mp}E_\pm \mathrm{e}^{-i\omega_\mp t}(\hat{\mathbf{x}}' \pm i\hat{\mathbf{y}}') = \frac{Q^2}{mb_\mp}E_\pm \mathrm{e}^{-i\omega t}\hat{\mathbf{u}}_\pm, \tag{S12}$$

where we only observe an electric dipole at the incident frequency ω in the lab frame. This is consistent with the intuition that rotating isotropic nanoparticles (e.g., molecules) do not produce rotational inelastic (e.g., Raman) scattering. Compared with the nanorod, the electric dipoles and associated radiation at frequencies $\omega \pm 2\Omega$ from the two branches of the nanocross cancel each other, and therefore, the Abraham-Lorentz force has no inelastic contribution, so that only the term $\tau\omega^3$ appears in $b_\mp$.

Cross sections. For optically isotropic particles, the circular polarizabilities can be strictly defined by $\mathbf{p}_\pm = \alpha_\pm(\omega)\mathbf{E}_\pm$, so from Eq. (S12) we find

$$\alpha_\pm(\omega) = \frac{Q^2}{mb_\mp}.$$

Following a similar procedure as for the nanorod, we can calculate the power associated with the scattering process of the rotating nanocross due to different terms in Eq. (S1), including the extinction power $P_{\mathrm{ext}} = Q\sum_i \langle \mathbf{E}\cdot\dot{\mathbf{r}}_i\rangle$, the scattering power $P_{\mathrm{sca}} = m\tau\sum_i \langle \ddot{\mathbf{r}}_i\cdot\dot{\mathbf{r}}_i\rangle$, and the power due to internal dissipation $P_{\mathrm{dis}} = -m\gamma\sum_i(\dot{\mathbf{r}}_i - \Omega r\hat{\varphi})\cdot\dot{\mathbf{r}}_i$. Obviously, this requires adding the contributions from the two branches. We finally find the following expressions for these cross sections:

$$\sigma_{\mathrm{ext}} = \frac{4\pi Q^2\omega}{mc}\mathrm{Im}\{\frac{1}{b_\mp}\}, \quad \sigma_{\mathrm{sca}} = \frac{4\pi Q^2}{mc}\frac{1}{|b_\mp|^2}\tau\omega^4, \quad \sigma_{\mathrm{dis}} = \frac{4\pi Q^2}{mc}\frac{1}{|b_\pm|^2}\gamma\omega_\mp^2.$$

To calculate the power associated with the mechanical force, we need to find the reaction force by considering the equations of motion of the charges evolving in each branch along perpendicular nanocross

directions. For example, for the branch aligned along $\hat{\mathbf{x}}'$, Eq. (S3) leads to

$$-i\Omega\omega_{\mp}\rho_1 - i\tau(\omega_{\mp}^3\rho_1 - 3i\Omega\omega_{\mp}^2\rho_2 + 3\Omega^2\omega_{\mp}\rho_1 + \Omega^3\rho_2) = \pm i\frac{Q}{\sqrt{2}m}E_{\pm} + F_1^{\text{react}},$$

where we also include the Abraham-Lorentz force due to radiation from the other oscillator of coordinate vector $\mathbf{r}_2$. Now, using the relations $\rho_1 = QE_{\pm}/\sqrt{2}mb_{\mp}$ and $\rho_2 = \pm i\rho_1$, we finally obtain the reaction force due to the first branch,

$$\mathbf{F}_1^{\text{react}} = \mp\frac{iQ}{\sqrt{2}mb_{\mp}}E_{\pm}(\omega_0^2 - \Omega^2 - \omega_{\mp}^2 \mp \Omega\omega_{\mp} - i\gamma\omega_{\mp})\hat{\mathbf{y}}'.$$

The power due to energy conversion into mechanical rotation is then given by $P_{\text{react},1} = -\langle \mathbf{F}_1^{\text{react}} \cdot \dot{\mathbf{r}}_1 \rangle$. Similarly, we can find the power due to mechanical forces acting on the branch aligned along $\hat{\mathbf{y}}'$, which is simply equal to the effect of the torque on the first branch (i.e., $P_{\text{react},2} = P_{\text{react},1}$) because of the symmetry of the particle. Similar to the nanorod, we show below that the dissipation force does not contribute to the torque acting on the ions, so the total mechanical power can be equivalently obtained as $P_{\text{react}} = \left\langle \mathbf{M}^{\text{react}} \cdot \vec{\Omega} \right\rangle = P_{\text{react},1} + P_{\text{react},2}$, where $\langle \mathbf{M} \rangle = 2\langle \mathbf{F}_1^{\text{react}} \times \mathbf{r}_1 \rangle$ is the torque acting on the particle and the factor 2 accounts for the contribution from the branch along $\hat{\mathbf{y}}'$. Using this expression for P_{react}, we finally obtain the corresponding cross section

$$\sigma_{\text{react}} = \frac{4\pi Q^2}{mc}\frac{1}{|b_{\mp}|^2}\gamma\Omega\omega_{\mp}.$$

Finally, the absorption cross section is given by $\sigma_{\text{abs}} = \sigma_{\text{dis}} + \sigma_{\text{react}}$.

Energy and angular momentum conservation. We can easily prove that the cross sections found above satisfy the well-known results $\sigma_{\text{ext}} = 4\pi k\text{Im}\{\alpha_{\pm}(\omega)\}$ and $\sigma_{\omega} = 8\pi k^4 |\alpha_{\pm}(\omega)|/3$, and furthermore, energy conservation stems from the fact that they satisfy the relation $\sigma_{\text{ext}} = \sigma_{\text{abs}} + \sigma_{\text{sca}}$.

Similar to the nanorod, the torque components acting on the rotating nanocross and associated with the force terms in Eq. (S1) can be equally obtained from $\sum_i^{1,2}\langle \mathbf{r}_i \times \mathbf{F} \rangle$, where $\mathbf{F}$ is chosen as $-\gamma m\dot{\mathbf{r}}$, $\tau m\dddot{\mathbf{r}}$, or $\mathbf{F}_i^{\text{react}}$. We find

$$\mathbf{M}_{\text{ext}} = \pm\frac{Q^2}{2m}|E_{\pm}|^2\text{Im}\{\frac{1}{b_{\mp}}\}\hat{\mathbf{z}}, \quad \mathbf{M}_{\text{sca}} = \mp\frac{Q^2}{2m}\frac{|E_{\pm}|^2}{|b_{\mp}|^2}\tau\omega^3\hat{\mathbf{z}}, \quad \mathbf{M}_{\text{dis}} = 0, \quad \mathbf{M}_{\text{react}} = \mp\frac{Q^2}{2m}\frac{|E_{\pm}|^2}{|b_{\mp}|^2}\gamma\omega_{\mp}\hat{\mathbf{z}}.$$

Compared with the nanorod, the expression for $\mathbf{M}_{\text{ext}}$ only differs in the definition of $b_{\pm}$ and a factor of 2 due to the fact that two charges are present in the nanocross. We note that the dissipation torque is still zero, and the contribution of inelastic scattering is absent in $\mathbf{M}_{\text{sca}}$ and $\mathbf{M}_{\text{react}}$. It is then easy to verify that these expressions satisfy $\mathbf{M}_{\text{ext}} = \mathbf{M}_{\text{sca}} + \mathbf{M}_{\text{react}}$, thus guaranteeing conservation of angular momentum. The total torque acting on the particle is $\mathbf{M} = -(\mathbf{M}_{\text{dis}} + \mathbf{M}_{\text{react}})$, which can be shown to satisfy the general relation in Eq. (S11) by setting $\sigma_{\omega\mp 2\Omega} = 0$.

C. Nanodisk

Equation of motion. For the nanodisk shown in fig. S1D, internal electrons can move freely and do not experience forces stemming from a potential boundary (i.e., $\mathbf{F}_{\text{react}} = 0$). We thus represent the electrons as a free two-dimensional oscillator with a total charge of $2Q$. For circularly polarized incident light, we write the incident electric field as $\mathbf{E}_{\pm} = E_{\pm}\text{e}^{-i\omega_{\mp}t}\hat{\mathbf{u}}'_{\pm}$, and due to the rotational symmetry of the particle, the position vector of the effective charge should be $\mathbf{r} = \rho\text{e}^{-i\omega_{\mp}t}\hat{\mathbf{u}}'_{\pm}$. Indeed, by substituting $\mathbf{E}_{\pm}$ and $\mathbf{r}$ into Eqs. (S2) and (S3), we find that these equations reduce to the relation

$$\rho = QE_{\pm}/mh_{\mp},$$

where

$$h_{\mp} = \omega_0^2 - \Omega^2 - \omega^2 - i\gamma\omega_{\mp} - i\tau\omega^3.$$

Cross sections. The circular polarizabilities (for RCP and LCP illumination) of the rotating nanodisk determined from the above equations reduce to

$$\alpha_{\pm}(\omega) = Q^2/mh_{\mp}.$$

Calculations similar to the cross sections of the rotating nanorod and nanocross can be carried out for the rotating nanodisk. By evaluating the extinction power $P_{\rm ext}$, the scattering power $P_{\rm sca}$, and the power of internal dissipation $P_{\rm dis}$, we readily find

$$\sigma_{\rm ext} = \frac{4\pi Q^2}{mc}\omega{\rm Im}\{\frac{1}{h_{\mp}}\}, \quad \sigma_{\rm sca} = \frac{4\pi Q^2}{mc}\frac{1}{|h_{\mp}|^2}\tau\omega^4, \quad \sigma_{\rm dis} = \frac{4\pi Q^2}{mc}\frac{1}{|h_{\pm}|^2}\gamma\omega\omega_{\mp}.$$

Energy and angular momentum conservation The above cross sections also satisfy $\sigma_{\rm ext} = 4\pi k{\rm Im}\{\alpha_{\pm}(\omega)\}$ and $\sigma_{\omega} = 8\pi k^4 \left|\alpha_{\pm}(\omega)\right|/3$, and again, energy conservation is readily corroborated from the expression $\sigma_{\rm ext} = \sigma_{\rm abs} + \sigma_{\rm sca}$ that they satisfy.

We can also calculate different contributions to the torque acting on the electrons. We find

$$\mathbf{M}_{\rm ext} = \pm\frac{Q^2}{2m}|E_{\pm}|^2{\rm Im}\{\frac{1}{h_{\mp}}\}\hat{\mathbf{z}}, \quad \mathbf{M}_{\rm sca} = \mp\frac{Q^2}{2m}\frac{|E_{\pm}|^2}{|h_{\mp}|^2}\tau\omega^3\hat{\mathbf{z}}, \quad \mathbf{M}_{\rm dis} = \mp\frac{Q^2}{2m}\frac{|E_{\pm}|^2}{|h_{\mp}|^2}\gamma\omega_{\mp}\hat{\mathbf{z}},$$

from where we also find the total angular momentum to be conserved (i.e., $\mathbf{M}_{\rm ext} = \mathbf{M}_{\rm sca} + \mathbf{M}_{\rm dis}$). The total torque acting on the rotating nanodisk is $\mathbf{M} = -\mathbf{M}_{\rm dis}$, which also satisfies the general relation given by Eq. (S11).

The dissipation force leads to a nonzero time-averaged torque acting on the particle. In contrast to the nanorod and nanocross, the dissipation power $P_{\rm dis}$ includes also the contribution of energy conversion into mechanical energy, as given by $P_{\rm mech} = \left\langle \mathbf{M} \times \vec{\Omega} \right\rangle$. The power of conversion to thermal energy is then

$$P_{\rm therm} = P_{\rm dis} - P_{\rm mech} = \frac{Q^2}{2m}\frac{|E_{\pm}|^2}{|h_{\mp}|^2}\gamma\omega_{\mp}^2.$$

II. ROTATIONAL DOPPLER COOLING LIMIT

According to Eqs. (S11) and (3), for a small rotation frequency Ω and an isotropic particle with $\gamma \ll \tau^{-1}$, the torque exerted by linearly polarized light reduces to

$$M \approx \frac{8\pi\Omega}{c}\partial_{\Omega}{\rm Im}\{\alpha_{+}(\omega)\}|_{\Omega=0} = -\beta\Omega,$$

so the dynamics of the particle angular momentum is governed by

$$J\dot{\Omega} = -\beta\Omega + M_{\rm fl}(t),$$

where $M_{\rm fl}$ is the fluctuating torque, and the cooling torque $\beta\Omega$ introduces a frictional torque by analogy to the frictional force in optical molasses. From the fluctuation-dissipation theorem, we can find the Einstein relation that determines the steady-state temperature

$$k_{\rm B}T_{\rm lim} = \frac{D}{\beta},$$

where $D = \gamma \left\langle \Delta L(t)^2 \right\rangle$ is the rotational diffusion coefficient. Considering that the dissipation of each incident angular momentum introduces $\hbar^2$ into the deviation of the particle angular momentum $\left\langle \Delta L(t)^2 \right\rangle$, we find the diffusion coefficient

$$D = \frac{2I}{\hbar}\sigma_{\text{ext}}|_{\Omega=0} \times \hbar^2 = \frac{8\pi\hbar}{c}\text{Im}\{\alpha_+(\omega)\}|_{\Omega=0}.$$

The above Einstein relation for the particle rotation can be further recast as

$$k_{\text{B}}T_{\text{lim}} = \hbar \left.\frac{\text{Im}\{\alpha\}}{\partial_\Omega \text{Im}\{\alpha\}}\right|_{\Omega=0}. \tag{S13}$$

We then use this relation to estimate the lowest temperature that is achievable by rotational Doppler cooling. For nanocrosses and nanodisks with the polarizability of Eq. (2), the two terms in Eq. (S13) can be explicitly found to be

$$\begin{aligned}
\text{Im}\{\alpha\}|_{\Omega=0} &= \frac{\gamma\omega}{(\omega_0^2-\omega^2)^2+\gamma^2\omega^2}, \\
\partial_\Omega \text{Im}\{\alpha\}|_{\Omega=0} &= \frac{\gamma(\omega_0^2-\omega^2)^2-\gamma^3\omega^2+4\gamma\omega^2(\omega_0^2-\omega^2)m}{[(\omega_0^2-\omega^2)^2+\gamma^2\omega^2]^2},
\end{aligned}$$

where $m = 1$ for crosses and $m = 0$ for disks. Inserting these two expressions into Eq. (S13), we find

$$k_{\text{B}}T_{\text{lim}} = \hbar\frac{[(\omega_0^2-\omega^2)^2+\gamma^2\omega^2]\omega}{(\omega_0^2-\omega^2)^2-\gamma^3\omega^2+4\omega^2(\omega_0^2-\omega^2)m} \xrightarrow{\gamma\ll\omega_0} \frac{[\delta^2+(\gamma/2)^2]\omega_0}{\delta^2-(\gamma/2)^2+2\delta\omega_0 m},$$

where $\delta = \omega - \omega_0$ is the frequency detuning, and the rightmost approximation corresponds to the low-damping limit, $\gamma \ll \omega_0$. The lowest temperature is determined by the minimum of the right-hand side of the above equation. In the low-damping limit $\gamma \ll \omega_0$, we find a Doppler cooling limit for the nanocross given by $k_{\text{B}}T_{\text{lim}} = \hbar\gamma/2$, which is achieved at $\delta = \gamma/2$; this is similar to the translational Doppler limit temperature. In contrast, in the limit $\gamma \ll \omega_0$, the Doppler cooling limit in the nanodisk is found to be $k_{\text{B}}T_{\text{lim}} = \hbar\omega_0$ for large frequency detuning $\delta \to \infty$. We note that for nanocrosses the Doppler cooling limit determined by the more rigorous expression in the above equation decreases with γ, so the result $k_{\text{B}}T_{\text{lim}} = \hbar\omega_0$ that is found for $\gamma \ll \omega_0$ is indeed the lowest achievable temperature. However, for nanodisks, the more rigorous temperature limit T_{lim} deceases as γ increases, so lower temperatures can be found in particles with stronger dissipation.

Figure S1

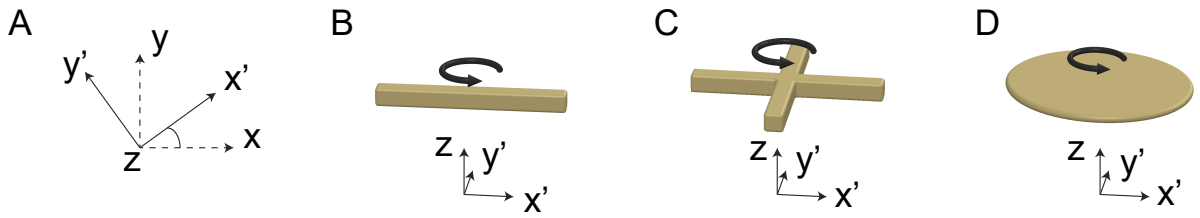

FIG. S1: **Choice of coordinate systems for discussions.** (**A**) Coordinate of the rotating frame (solid vectors) and the laboratory frame (dashed vectors). (**B** to **D**) Illustration of the orientation of three types of rotating particles in the rotating frame: nanorod (B), nanocross (C), and nanodisk (D).

Figure S2

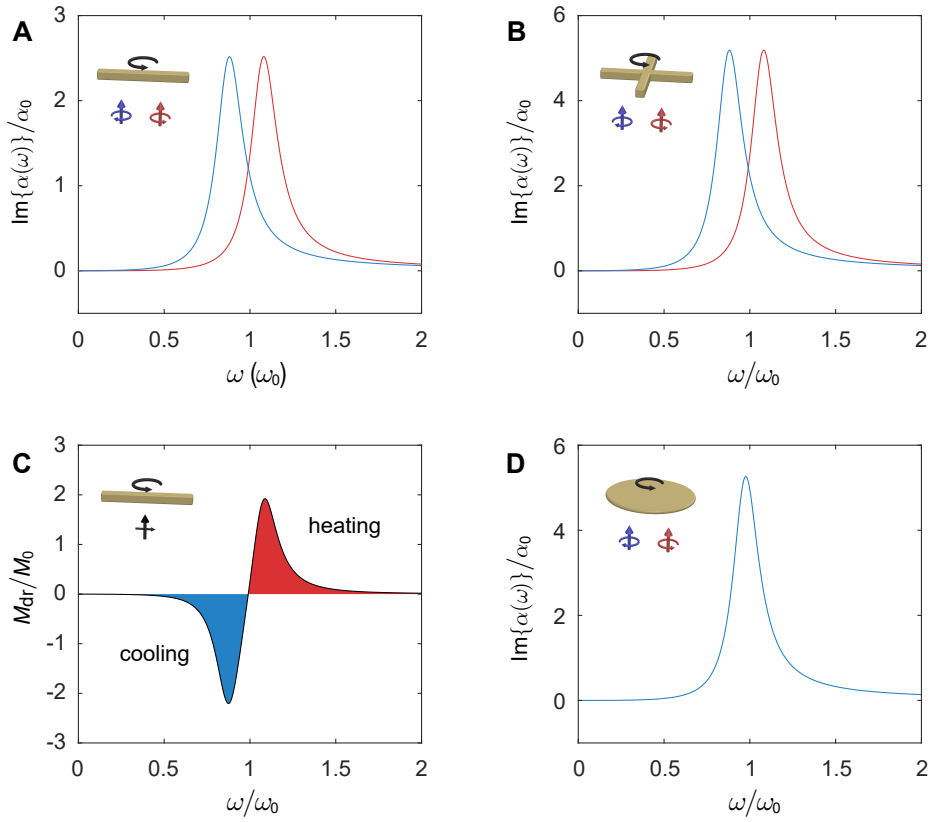

FIG. S2: **Optical response of dissipationless rotating nanoparticles.** (**A** to **C**) We consider a nanorod (A), a nanocross (B) and a nanodisk (C) with RCP (red) and LCP (blue) incident light. (**D**) Time-averaged torque acting on the rotating nanorod in (A) under linearly polarized illumination. In all cases, particles are rotating with angular velocity $\Omega = 0.1\omega_0$, and the radiative damping rate is $\tau^{-1} = 0.2\omega_0$. All frequencies are normalized to the particle resonance frequency ω_0, the polarizability is normalized to $\alpha_0 = Q^2/m\omega_0^2$, and the torque is normalized to $M_0 = \alpha_0|E_\pm|^2/2$. The nanorod (A) and nanocross (B) exhibit a CD similar to the dissipative scenarios in Fig. 2, while the CD is absent in the nanodisk (C). A light wave cannot produce an optical torque on dissipationless isotropic particles, such as the nanocross (B) and the nanodisk (C). However, an optical torque can arise on anisotropic nanoparticles (such as the nanorod in (A)) due to inelastic scattering, and consequently, linearly polarized illumination can lead to RDC and RDH in such particles.

Figure S3

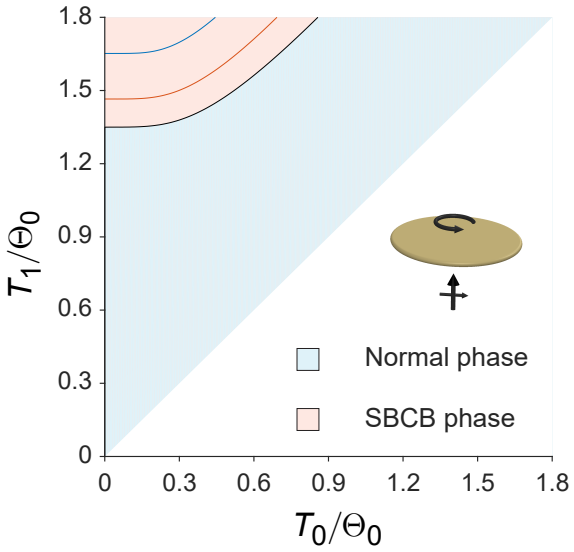

FIG. S3: **Stability of a nanodisk at rest under linearly polarized illumination with frequency $\omega = \omega_0$.** The blue, red and black curves correspond to the phase boundaries for $\gamma = 0.25\omega_0$, $0.35\omega_0$ and $0.35\omega_0$, respectively. The vacuum and particle temperatures T_0 and T_1 are normalized to $\Theta_0 = \hbar\omega_0/k_B$.

Figure S3 shows the stability of a nanodisk at rest under linearly polarized illumination with a light frequency $\omega = \omega_0$ falling into the RDH regime according to Fig. 2F. Compared with nanocrosses and nanorods, a higher particle temperature is required to break the rotational stability of the nanodisk. Because the circular dichroism of the rotating nanodisk derives from internal dissipation according to A_{disk} in Eq. (2), a disk with larger dissipation (i.e., larger γ) shows stronger circular dichroism, which further leads to a decrease in the threshold particle temperature, as illustrated by the phase boundaries shown for different γ's. This is in contrast to nanorods and nanocrosses, in which a small γ results in stronger circular dichroism and helps to reduce the threshold particle temperature.

Figure S4

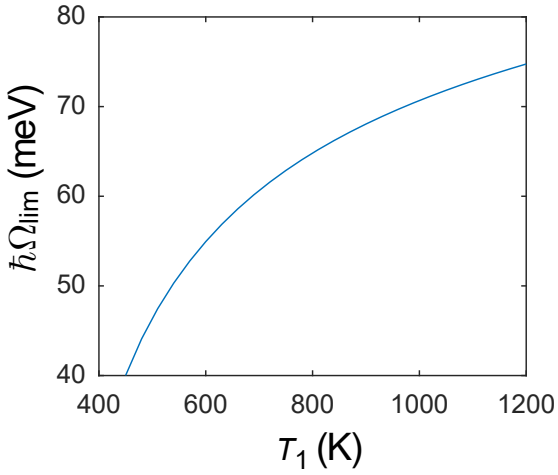

FIG. S4: **Rotation frequency of a nanocross in equilibrium under laser pumping.** The relative magnitudes of parameters ω_0, γ, and τ are the same as in Fig. 2 and 3; namely, $\gamma = 0.2\omega$ and $\tau^{-1} = 0.02\omega_0$, where $\omega = 1.1\omega_0$ is the laser frequency. We assume the vacuum at room temperature $T_0 = 300\,\mathrm{K}$ and $T_0 = 0.1\Theta_0$, so the particle resonance is located at $\hbar\omega_0 = 0.026\,\mathrm{eV}$, corresponding to a photon wavelength of $4.8\,\mu\mathrm{m}$. According to the phase diagram in Fig. 3A, for the chosen laser frequency $\omega = 1.1\omega_0$, when the particle temperature T_1 is above $0.13\Theta_0 \approx 390\,\mathrm{K}$, it starts rotating and eventually reaches an equilibrium rotation frequency Ω (black dots in Fig. 3C). The equilibrium frequency Ω_{eq} is determined by the laser intensity $I(T_1)$ or the equilibrium temperature T_1 at $\Omega = 0$. In the absence of other frictional mechanisms such as gas collisions, the rotation of the particle can be accelerated up to the THz regime.

Figure S5

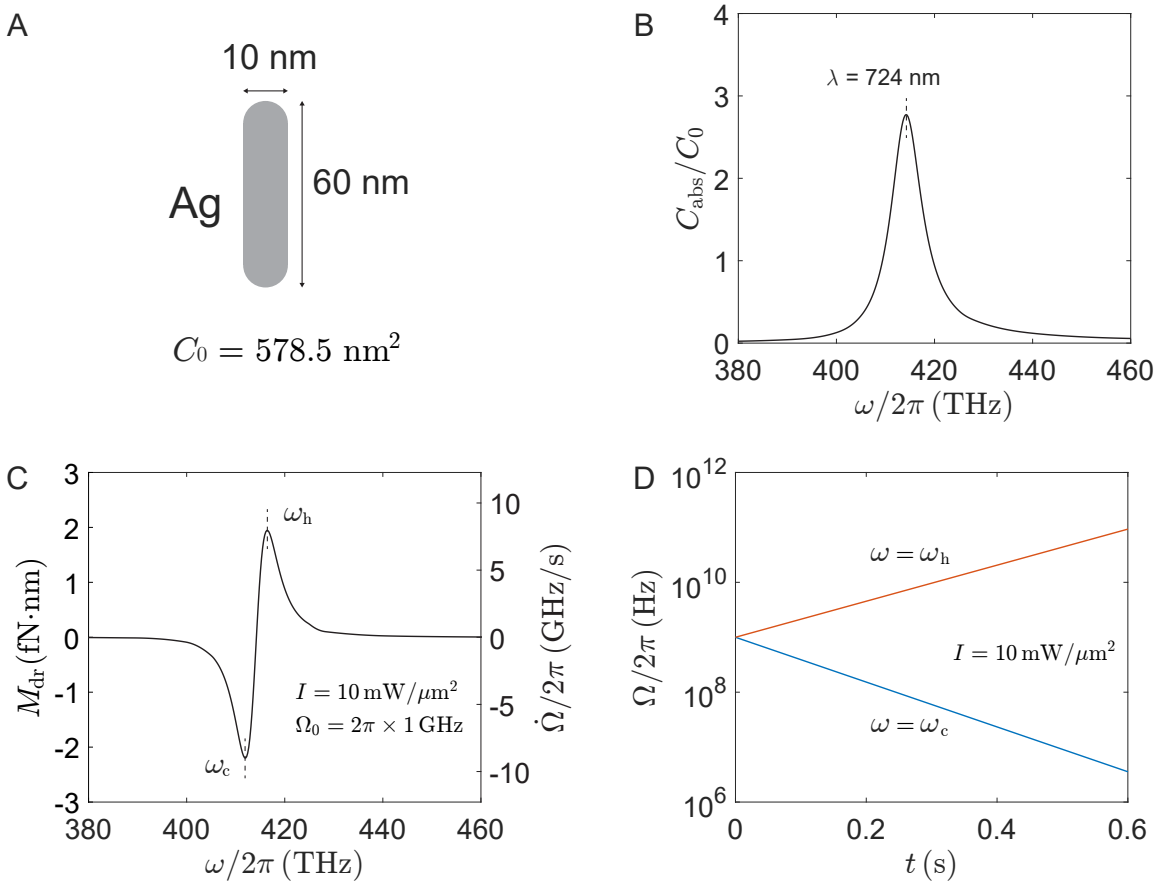

FIG. S5: **Cooling and heating rotation of a silver nanorod.** (**A**)Illustration of the studied silver nanorod, with a geometric cross section $C_0 = 578.5\,\mathrm{nm}^2$. (**B**) Absorption cross section of the silver nanorod at rest for different light frequencies, which shows a pronounced plasmon resonance peak. A linear polarization parallel with the long axis of the nanorod is considered here. (**C**) Optical torque exerted by a linearly polarized light on the nanorod rotating with a frequency $\Omega_0 = 2\pi \times 1\,\mathrm{THz}$. The resulting acceleration of the particle rotation $\dot{\Omega} = M_{\mathrm{dr}}/J_{\mathrm{rod}}$ is also scaled, where the moment of inertia of the nanorod is $J_{\mathrm{rod}} = 3.89 \times 10^{-39}\,\mathrm{kg}\cdot\mathrm{m}^2$, calculated based on the density of silver, $10.49\,\mathrm{g/cm}^3$. (**D**) Time evolution of the particle rotation for a laser with two frequencies, $\omega = \omega_{\mathrm{c}}$ and $\omega = \omega_{\mathrm{h}}$, working in the RDC and RDH regimes, respectively, where ω_{c} and ω_{h} are indicated in (C), and the initial rotation frequency is assumed to be $\Omega = \Omega_0$. In both (C) and (D), a moderate light intensity $I = 10\,\mathrm{mW}/\mu\mathrm{m}^2$ is assumed. For such light intensity, the rotation of the nanorod accelerates or decelerates by several orders of magnitude within a subsecond timescale. The minimum and maximum rotation frequencies achievable in the cooling and heating effects are bounded by the RDC limit and the steady condition shown in fig. S4, respectively.