

Supporting Information

Scattering and Extinction Torques: How Plasmon Resonances Affect the Orientation Behavior of Nanorod in Linearly Polarized Light

Xiaohao Xu, Chang Cheng, Yao Zhang, Hongxiang Lei*, and Baojun Li*
State Key Laboratory of Optoelectronic Materials and Technologies, School of
Physics and Engineering, Sun Yat-Sen University, Guangzhou 510275, China

*E. mail: leihx@mail.sysu.edu.cn (H.L.); stslbj@outlook.com (B.L.)

1. Lorentz model for comprehending the reversal of phase of current density \mathbf{J} when the light wavelength (or frequency) passes through the plasmon resonance wavelength (or frequency).

A plasmon is a collective oscillation of the free electrons in a noble metal. The electron gas can be modeled as an ideal mechanical oscillator subject to an electric field \mathbf{E}_{loc} , which is known as the Lorentz model (see Figure S1). The equation of motion for such an oscillator is (see ref. 20 in the main text):

$$m\mathbf{x}'' + b\mathbf{x}' + K\mathbf{x} = -q\mathbf{E}_{\text{loc}}, \quad (1)$$

where \mathbf{x} is the oscillator displacement from equilibrium, K is the spring constant, b is the damping constant, and m and $-q$ are the mass and charge of the oscillator, respectively. Assuming the applied field varies harmonically in time with frequency ω as $e^{-i\omega t}$, with the substitution $\omega_0^2 = K/m$ and $\gamma = b/m$, the oscillatory part of the solution to this equation is given by $\mathbf{x} = x_A e^{i\Theta}(-q\mathbf{E}_{\text{loc}}/m)$, where the amplitude $x_A = 1/[(\omega_0^2 - \omega^2) + \gamma^2 \omega]^2$ and the phase $\Theta = \tan^{-1} \gamma\omega/(\omega_0^2 - \omega^2)$. It indicates that when the angular frequency ω of external field passes through a resonant angular frequency ω_0 , the phase Θ of the displacement of electron gas will shift π (i.e. $\mathbf{x} \rightarrow -\mathbf{x}$), resulting in a reversal of charge density phase $\rho \rightarrow -\rho$. On the other hand, the phase Θ' of velocity \mathbf{x}' of electron motion is $\Theta' = \tan^{-1} (\omega^2 - \omega_0^2)/\gamma\omega$, which varies slowly around ω_0 , so it can be treated as a constant when ω passes through resonant angular frequency ω_0 . As a result, the current density, which is given by $\mathbf{J} = \rho\mathbf{x}'$, become opposite (i.e., $\mathbf{J} \rightarrow -\mathbf{J}$).

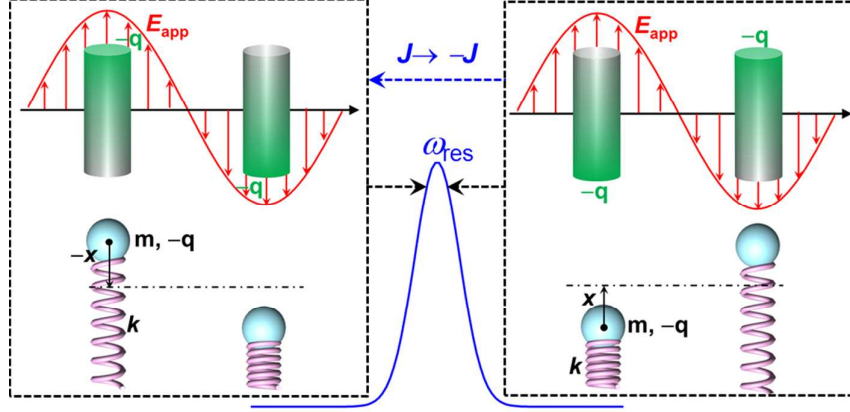


Figure S1. Illustration for Lorentz model. The electron gas is modeled as an ideal mechanical oscillator (mass m and charge $-q$) subject to an electric field \mathbf{E}_{loc} . The equation of oscillator motion: $m\mathbf{x}'' + b\mathbf{x}' + K\mathbf{x} = -q\mathbf{E}_{\text{loc}}$, where \mathbf{x} is the oscillator displacement from equilibrium, K is the spring constant, b is the damping constant.

2. Details of FDTD simulations.

Three-dimensional finite-difference time-domain (FDTD) simulations were performed using the commercial software package “FDTD Solutions” (Lumerical, Inc.). The simulation region is $1.05 \times 0.14 \times 0.14 \mu\text{m}^3$, and a mesh with cell size of 8 nm^3 was used (2572500 cells); the time step is 0.004 fs. The dielectric function of silver was modeled with parameters adjusted to match tabulated values from ref. 22 in the text. In this software package, option “tolerance” stands for the convergence criteria for the optimization to terminate, and it was set to 0 to ensure the optimization will run until the maximum number of generations has been reached.

3. Transforming the formulas of scattering, extinction and total torques.

Firstly, we express the incident electric field \mathbf{E}_{inc} and scattering electric field \mathbf{E}_{sca} as the superposition of three orthogonal components: $\mathbf{E}_{\text{inc}} = E_{ix}\mathbf{e}_x + E_{iy}\mathbf{e}_y + E_{iz}\mathbf{e}_z$, $\mathbf{E}_{\text{sca}} = E_{sx}\mathbf{e}_x + E_{sy}\mathbf{e}_y + E_{sz}\mathbf{e}_z$, where the unit vectors \mathbf{e}_x , \mathbf{e}_y , \mathbf{e}_z are in the directions of positive x , y , z axes, respectively. Because of the linear boundary conditions, the amplitude of field scattered by an arbitrary particle should be a linear function of the amplitude of the incident field (see ref. 20 in the main text). Accordingly, the relation between incident and scattering fields can be written in matrix form:

$$\begin{bmatrix} E_{sx} \\ E_{sy} \\ E_{sz} \end{bmatrix} = \langle S \rangle \begin{bmatrix} E_{ix} \\ E_{iy} \\ E_{iz} \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} & S_{13} \\ S_{21} & S_{22} & S_{23} \\ S_{31} & S_{32} & S_{33} \end{bmatrix} \begin{bmatrix} E_{ix} \\ E_{iy} \\ E_{iz} \end{bmatrix}, \quad (2)$$

and

$$\begin{bmatrix} H_{sx} \\ H_{sy} \\ H_{sz} \end{bmatrix} = \langle T \rangle \begin{bmatrix} H_{ix} \\ H_{iy} \\ H_{iz} \end{bmatrix} = \begin{bmatrix} T_{11} & T_{12} & T_{13} \\ T_{21} & T_{22} & T_{23} \\ T_{31} & T_{32} & T_{33} \end{bmatrix} \begin{bmatrix} H_{ix} \\ H_{iy} \\ H_{iz} \end{bmatrix}, \quad (3)$$

where $\langle S \rangle$ and $\langle T \rangle$ are two 3×3 amplitude matrixes that linearly transform the incident field to scattering field. For a given incident field and nanorod, the matrixes are generally functions of position and are independent of polarization direction θ relative to the $+x$ direction. For a linearly polarized incident electric field, which lies in the x - y plane, we have $E_{ix} = E_{inc} \cos \theta$, $E_{iy} = E_{inc} \sin \theta$, $E_{iz} = 0$, and $\mathbf{E}_{inc} = E_{inc} \cos \theta \mathbf{e}_x + E_{inc} \sin \theta \mathbf{e}_y$, where E_{inc} is the amplitude of incident electric field \mathbf{E}_{inc} . Substituting these components into formula 2, we obtain $\mathbf{E}_{sca} = (S_{11}E_{inc} \cos \theta + S_{12}E_{inc} \sin \theta) \mathbf{e}_x + (S_{21}E_{inc} \cos \theta + S_{22}E_{inc} \sin \theta) \mathbf{e}_y + (S_{31}E_{inc} \cos \theta + S_{32}E_{inc} \sin \theta) \mathbf{e}_z$. Similarly, the incident and scattering magnetic field can be written as $\mathbf{H}_{inc} = H_{inc} \sin \theta \mathbf{e}_x + H_{inc} \cos \theta \mathbf{e}_y$ and $\mathbf{H}_{sca} = (T_{11}H_{inc} \sin \theta + T_{12}H_{inc} \cos \theta) \mathbf{e}_x + (T_{21}H_{inc} \sin \theta + T_{22}H_{inc} \cos \theta) \mathbf{e}_y + (T_{31}H_{inc} \sin \theta + T_{32}H_{inc} \cos \theta) \mathbf{e}_z$, respectively, where H_{inc} the amplitude of incident magnetic field \mathbf{H}_{inc} . To obtain the relation between the optical torques and θ , we may extract the function that related to θ from formulas 1, 3, 4 in the main text but neglect the detailed forms of other terms that is independent on θ . Considering that the tensors $\langle M_{sca} \rangle$, $\langle M_{ext} \rangle$ and $\langle M \rangle$, that can include θ , are just the multiplication (e.g., $|\mathbf{E}_{inc}|^2$ or $E_{ix}E_{ix}^*$ or $E_{ix}E_{iy}^*$) of two electromagnetic components, the torques are expected to be the linear combination of $\sin^2 \theta$, $\cos^2 \theta$, and $\sin \theta \cos \theta$, i.e.,

$$T_{sca}(\theta) = \alpha_1 \sin^2 \theta + \alpha_2 \cos^2 \theta + \alpha_3 \sin \theta \cos \theta = \zeta_{sca} \sin 2(\theta - \varphi_{sca}) + \xi_{sca}, \quad (4)$$

$$T_{ext}(\theta) = \alpha_4 \sin^2 \theta + \alpha_5 \cos^2 \theta + \alpha_6 \sin \theta \cos \theta = \zeta_{ext} \sin 2(\theta - \varphi_{ext}) + \xi_{ext}, \quad (5)$$

$$T_{tot}(\theta) = \alpha_7 \sin^2 \theta + \alpha_8 \cos^2 \theta + \alpha_9 \sin \theta \cos \theta = \zeta_{tot} \sin 2(\theta - \varphi_{tot}) + \xi_{tot}, \quad (6)$$

where coefficients $(\alpha_1, \alpha_2, \alpha_3, \alpha_4 \dots)$, $(\zeta_{sca}, \zeta_{ext}, \zeta_{tot})$, $(\varphi_{sca}, \varphi_{ext}, \varphi_{tot})$ and $(\xi_{sca}, \xi_{ext}, \xi_{tot})$ are independent on θ . For cylindrical silver nanorods, the torques exerted on them should be vanished when $\theta = 0$ and 90° due to symmetric, thus $(\varphi_{sca}, \varphi_{ext}, \varphi_{tot})$ and $(\xi_{sca}, \xi_{ext}, \xi_{tot})$ are determined to be zero. Accordingly, formulas 1, 3, 4 in the main text are simplified to be

$$T_{tot} = \zeta_{tot} \sin 2\theta, \quad (7)$$

$$T_{ext} = \zeta_{ext} \sin 2\theta, \quad (8)$$

$$T_{sca} = \zeta_{sca} \sin 2\theta, \quad (9)$$

respectively. Obviously, the coefficients ζ_{sca} , ζ_{ext} and ζ_{tot} stands for the scattering, extinction and total torques at $\theta = 45^\circ$. The formula 7–9 implies that if the torques at a certain θ ($\theta \neq 0$ and 90°) is known, then the torques at each θ can be derived. Furthermore, it tells us that, if the objects can be oriented (i.e., $\zeta_{tot} \neq 0$), there exist two possible balanced states, i.e., $\theta = 0$ or 90° . Orientational balance occurs when the

total torque T_{tot} vanishes with positive derivative with respect to θ (i.e., $T_{\text{tot}}(\theta) = 0$ and $dT_{\text{tot}}(\theta)/d\theta > 0$). Therefore, when $\zeta_{\text{tot}} > 0$, the nanorods are forced to align parallel to the polarization (i.e., $\theta = 0$), whereas when $\zeta_{\text{tot}} < 0$, they will align perpendicular to the polarization (i.e., $\theta = 90^\circ$). Likewise, the directions of T_{sca} and T_{ext} are determined by the signs of ζ_{sca} and ζ_{ext} , respectively. For $\zeta_{\text{sca}} \text{ (or } \zeta_{\text{ext}}) > 0$ and $\zeta_{\text{sca}} \text{ (or } \zeta_{\text{ext}}) < 0$, T_{sca} (or T_{ext}) tends to orient the nanorod parallel and perpendicular to the light polarization, respectively.

4. Spectra of scattering, extinction and total torques for a silver nanorod with length 200 nm and diameter 100 nm and its extinction spectrum.

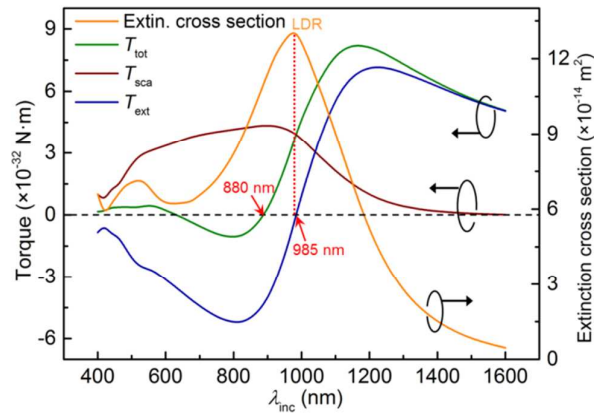


Figure S2. Spectra of scattering, extinction and total torques for a silver nanorod with length 200 nm and diameter 100 nm, together with extinction spectrum of the nanorod. The zero point of T_{ext} occurs at 985 nm, coinciding with the longitudinal dipole resonance (LDR). Because the scattering torque T_{sca} is always positive, the zero point (occurring at 880 nm) of the total torque T_{tot} is on the blue side of the LDR (occurring at 985 nm).