Super-resolution optical imaging and memory enabled by scattering suppression of shell-coated gold nanorods

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**1.**

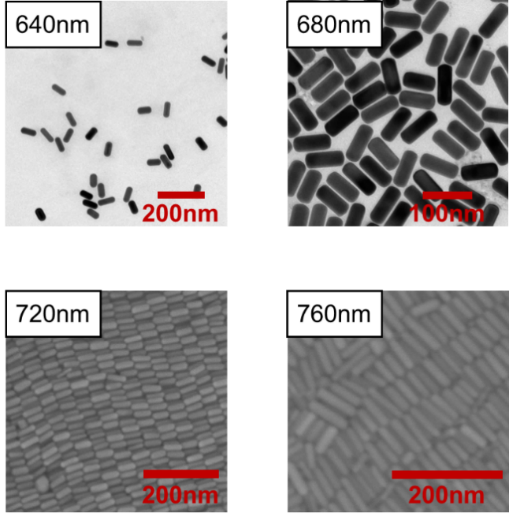


Fig. S1. TEM images of gold nanorods (GNRs) with plasmonic scattering peaks at 640 nm, 680 nm, 720 nm and 760 nm for the corresponded longitudinal diameter sizes: 57 nm, 63 nm, 72 nm and 84 nm.

**2.**



Fig. S2. Plasmonic scattering intensities of GNR (red) and GNS (blue) under varying power levels of a 640 nm excitation laser beam. The data were fitted using a polynomial fitting function.

In the experiment, we set the excitation laser (640 nm) power from 0.1 mW to 1 mW and measured the plasmonic scattering signals of gold nanosphere and GNR (the resonance peak at 540 nm and 760 nm respectively). The peak intensities of plasmonic scattering signals under different laser powers were plotted, as shown in Fig. S2. When the excitation laser power was below 0.3 mW (0.18 MW/cm²), the plasmonic scattering intensity exhibited a linear relationship with the excitation power [1]. However, when the excitation laser power reached 0.5 mW (0.3 MW/cm²), the plasmonic scattering intensity plateaued and saturated. This saturation behavior is attributed to the increased electron and lattice temperatures of the GNR under 640 nm laser excitation, which alter the dielectric constant and reduce the plasmonic scattering cross-section [2]. Notably, the plasmonic scattering intensity of a single GNR is significantly higher than that of a gold nanosphere, providing superior image quality in nanoparticle-based imaging applications.

**3.**

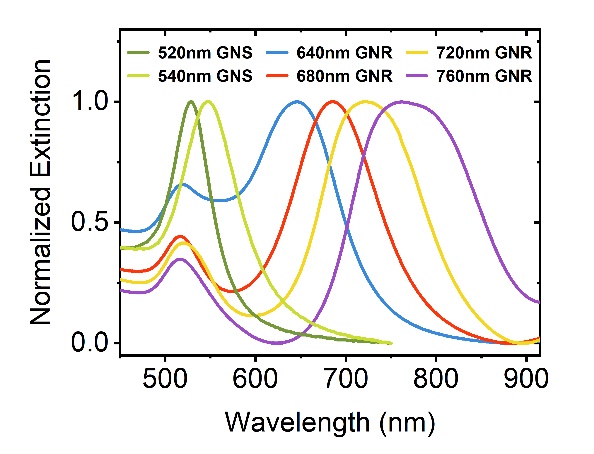


Fig. S3. Extinction spectra of gold nanoparticles with different sizes and shapes.

We measured the extinction spectra of gold nanoparticles with varying sizes and shapes in aqueous solutions. Gold nanospheres with diameters of 40 nm and 80 nm exhibited surface plasmon resonance (SPR) peaks predominantly in the 500-550 nm wavelength range. In contrast, gold nanorods demonstrated a significantly broader wavelength tunability range for their localized surface plasmon resonance (LSPR), spanning from the visible to near-infrared regions, far exceeding the spectral tunability of gold nanospheres.

**4.**

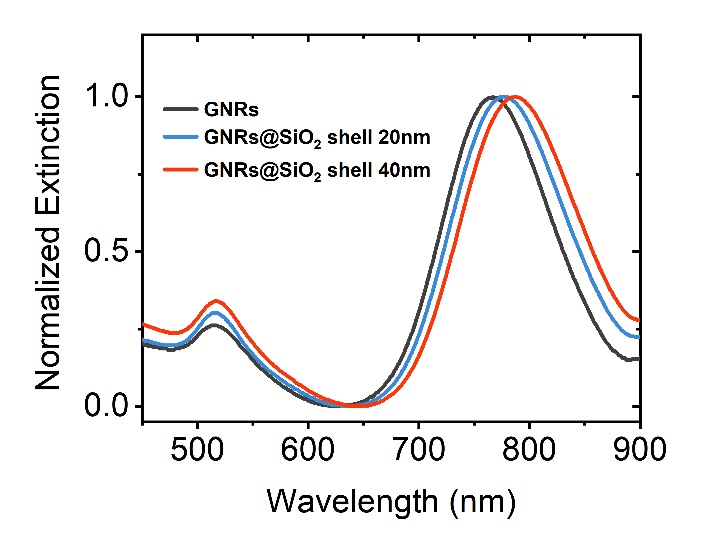


Fig. S4. The resonance peak of GNRs (length 85nm, width 24nm) used in the experiment is located at 765 nm. For the GNRs coated with silica shells, the resonance peak undergoes a red shift, appearing at 775 nm and 785 nm. Since the silica coating on the GNRs has a higher refractive index than the aqueous medium, the red shift of the resonance peak occurs.

**5.**

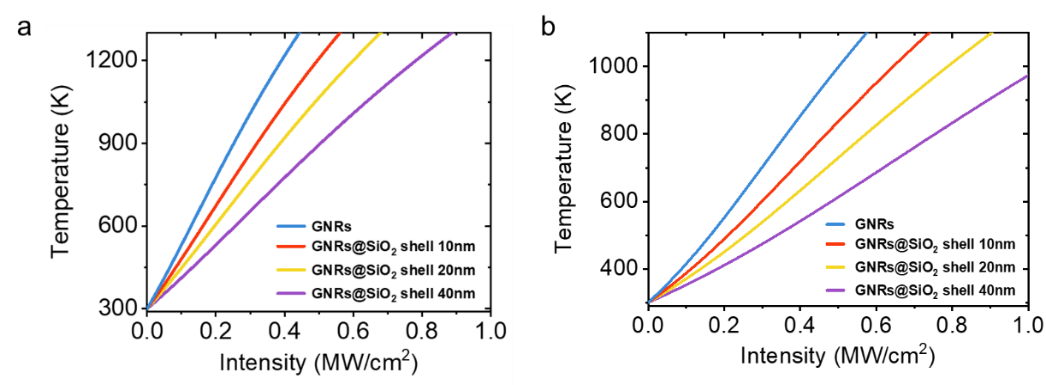


Fig. S5. Theoretical estimation of temperature evolution of GNRs (760 nm plasmonic scattering peak) with different silica shell thicknesses (0-40nm) in both air (a) and oil-immersed (b) surrounding environment.

The temperature from nonlinear heating of a GNR under the resonant CW plane wave illumination *TNP* can be expressed as [1]：

， is the total absorption power; is the absorption cross section and is the irradiance of the incoming light）。

: the effective thermal conductivity of substrate and environment (the values of depend on the surrounding environment and the different shell thicknesses of GNRS on the glass substrate, : 0.17, 0.2, 0.24, 0.32 and 0.3, 0.38, 0.46, 0.6 for estimating GNRs with 0 to 40 nm shell thickness values in air and oil surrounding environment [2])

: the radius of a sphere with the same volume as the GNRs (the thermal inhomogeneity is quite small ([3]) since the aspect ratio of GNRs used in our study is less than 4, therefore the GNRs (aspect ratio: 85nm : 25nm) can be estimated as a near-sphere scenario.)

: the dimensionless thermal-capacitance coefficient with an aspect ratio of D/d.

**6.**



Fig. S6. Simulation results of a GNR’s temperature evolution under the pulsed laser irradiation

When gold nanoparticles are irradiated by pulsed laser light, the absorption process can be divided into three distinct stages:

1. Electron absorption. When irradiated by pulsed laser light, a portion of the incident pulse energy is absorbed by the free electron gas within the nanoparticle. These electrons, being significantly lighter and more responsive than the lattice ions, rapidly thermalize into a Fermi-Dirac distribution on a timescale of . This process creates a nonequilibrium state within the nanoparticle: the electron temperature increases, while the lattice (phonon) temperature remains unchanged. The absorbed energy is given by:

Where is the absorption cross section, is the average irradiance， is the pulse repetition frequency. is the total absorption power.

1. Electron-phonon thermalization. Subsequently, the electron gas undergoes relaxation (cooling) through internal electron-phonon interactions, leading to lattice ion thermalization on a timescale of . This timescale is independent of nanoparticle size and remains constant for larger nanoparticles and moderate pulse energies, with approximately 1.7 ps. At this stage, the nanoparticle reaches internal equilibrium ， but remains out of equilibrium with the surrounding medium, which retains its initial ambient temperature.
2. External thermal diffusion. Energy diffusion into the surrounding medium typically occurs on a longer timescale , leading to nanoparticle cooling and medium heating. This timescale depends on nanoparticle size, ranging from 100 picoseconds to several nanoseconds. Given that electron-phonon thermalization is significantly faster than external thermal diffusion, the nanoparticle temperature reaches an initial maximum value, which can be directly estimated through energy conservation:

Following irradiation by a single laser pulse, the nanoparticle can be treated as a point heat source, and the subsequent thermal diffusion process can be described by the following equation:

The analytical solution after N pulses at any given time t is expressed as:

Since electron-phonon thermalization occurs significantly faster than external thermal diffusion, the initial temperature is considered uniform within the nanoparticle. Furthermore, due to the thermal conductivity , we can also assume that the nanoparticle temperature remains uniform throughout the evolution process [3]. The thermal diffusion equation for pulsed laser heating is given by:

Where ρ is the density and c is the specific heat capacity. The numerical solution can be obtained by using RK4 algorithm [3].

We now extend the model to gold nanorods. For gold nanorods with an aspect ratio less than 4, the uniform temperature approximation remains valid. Therefore, the gold nanorods used in experiments are treated as point heat sources. Under pulsed laser irradiation, the nanoparticle temperature depends solely on the nanoparticle volume.

Based on the experimental data, we calculated the temperature evolution of gold nanorods under pulsed laser irradiation, as shown in Fig S5. The average power of the pulsed laser is set to 2×105MW/cm2. For gold nanorods with a 20nm silica shell, the response time of the thermal lattice is about 20 ns scale.

For gold nanorods with different silica shell thicknesses, we adopt a temperature-dependent model. We calculated the thermal field distribution of gold nanorods in different local environments, as shown in Fig. 2(g).

**7.**

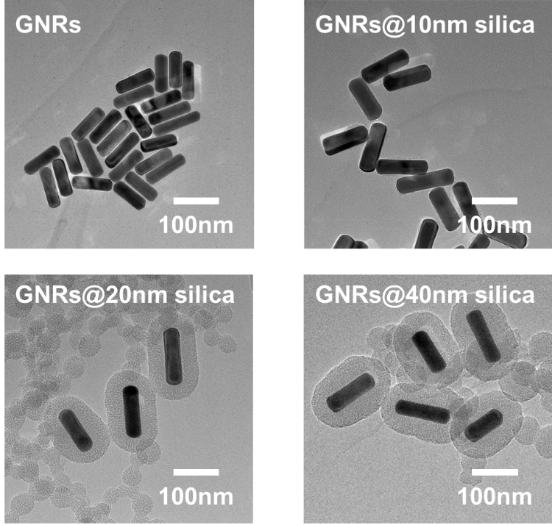


Fig. S7. Transmission electron microscopy (TEM) images of bare gold nanorods and gold nanorods coated with 10 nm, 20 nm, and 40 nm silica shells. The silica shells effectively isolate the particles.

**8.**

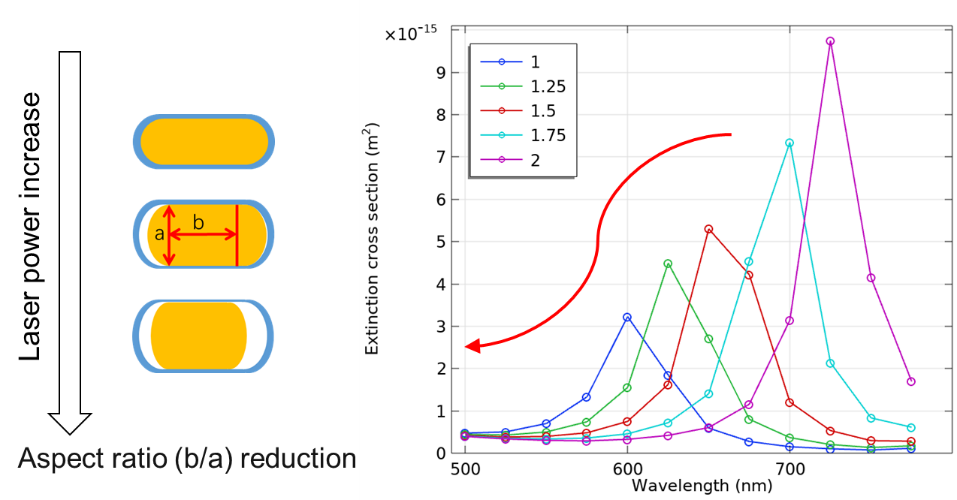
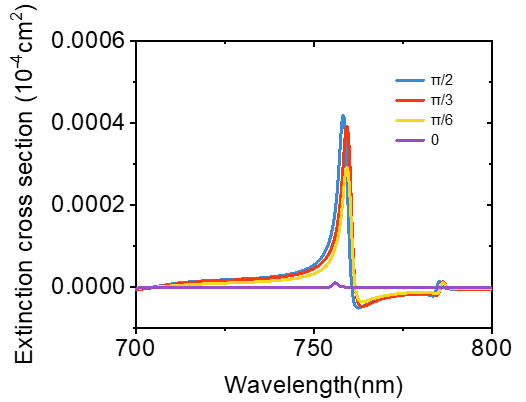


Fig. S8. Simulations reveal the evolution of plasmonic resonance characteristics in GNRs with 10 nm shell under increasing laser power density. The rising nanosecond laser pulse fluence induces aspect ratio reduction in GNRs, driving both blue shift and attenuation of scattering cross-section. Thin silica coatings offer merely partial protection, where analogous hollow interfacial regions emerge between the GNR core and silica shell as the aspect ratio decreases.

**9.**



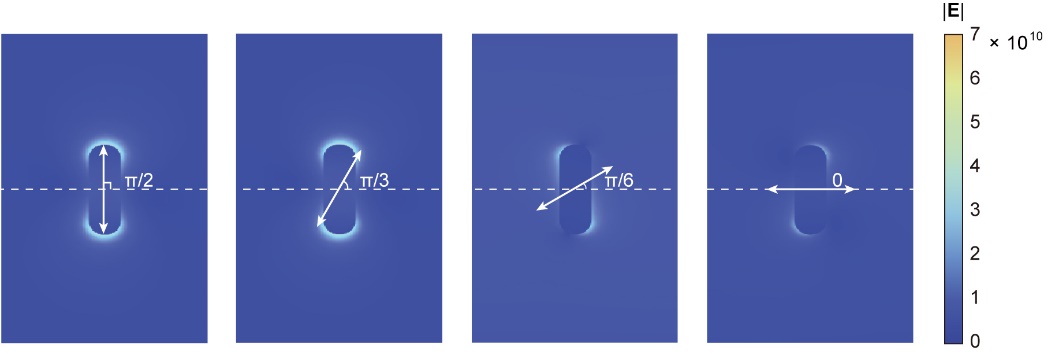


Fig. S9. (a)Simulated extinction cross section of a GNR as the polarization angle of the incident laser was varied. (b)Two-dimensional electric field enhancement contours of the GNR in different polarization angles. The incident laser polarization direction is marked by arrows.

The optical properties of anisotropic GNRs exhibit strong polarization dependence. Through simulations, we observed that adjusting the polarization angle of incident laser induces systematic variations in the electric field intensity, governed by symmetry rules (e.g., central or axial symmetry). In the simulations, a GNR with dimensions of 85 nm (length) × 25 nm (width) were illuminated by light at a wavelength of 775 nm. The polarization angle (θ) of the incident laser was systematically tuned to π/2, π/3, π/6, and 0. Fig. S8(a) displays the extinction cross-section curves of the GNR. As the polarization angle decreases, the extinction cross-section gradually diminishes. The maximum extinction cross-section occurs at θ = π/2, corresponding to the strongest plasmonic enhancement effect.

Fig. S8(b) illustrates the two-dimensional electric field intensity distributions of the GNR under the incident laser illumination with different polarization angles. The incident polarization direction is indicated by arrows, with polarization angles labeled. Notably, the symmetry of the electric field distribution (e.g., central or axial symmetry) varies as the polarization angle changes. Both the extinction cross-section and the localized field enhancement at the nanorod’s ends are inherently dependent on the polarization angle of the incident laser beam. By tuning the incident polarization angle, the field distribution can transit between distinct symmetric configurations (e.g., central symmetry to axial symmetry). These results underscore the significant potential of leveraging anisotropy in practical applications, particularly for polarization-engineered nanophotonic devices.

**References**

1. S. Chu, H. Wu, Y. Huang, *et al*., “Saturation and Reverse Saturation of Scattering in a Single Plasmonic Nanoparticle,” [ACS Photonics](https://doi.org/10.1021/ph4000218) **1** (1), 32-37 (2014).
2. Y. Chen, P. Lee, P. Shen, *et al*., “Study of Nonlinear Plasmonic Scattering in Metallic Nanoparticles,” [ACS Photonics](https://doi.org/10.1021/acsphotonics.6b00025) **3**(8), 1432-1439 (2016).
3. G. Baffou, and H. Rigneault, “Femtosecond-pulsed optical heating of gold nanoparticles,” [Phys. Rev. B](https://doi.org/10.1103/PhysRevB.84.035415) **84,** 035415 (2011).