

Supporting Information

Seed-mediated growth method for preparing the GNRs. 5 mL of 0.034 mol L⁻¹ HAuCl₄ aqueous solution was mixed with 20 mL of 2 mol L⁻¹ CTAB solution in a beaker. The mixture solution was stirred at 60 °C for about 15 min. Then 1 mL of 0.0005 mol L⁻¹ fresh NaBH₄ solution was injected into the seed solution. The color of the solution was immediately changed from yellow to dark brown. The seed solution was kept at 60 °C for 1 hour and then used for the synthesis of GNRs. The growth solution was prepared by mixing 0.03 mol Hexadecyltrimethylammonium bromide (CTAB), 0.0007 mol ortho-hydroxybenzoic, and 31.25 mL deionized water in a flask. After mixing, 0.5 mL of 0.004 mol L⁻¹ AgNO₃ solution, 0.156 mL of 0.00065 mol L⁻¹ L-ascorbic acid aqueous solution and 31.25 mL of 0.03375 mol L⁻¹ HAuCl₄ aqueous solution were added into the growth solution. The color of the solution turned orange at once and then became colorless. 0.226 mL of 12 mol L⁻¹ HCl solution was used to reduce the PH value below 7.0. Finally, 0.2 mL of seed solution was injected into the growth solution to initiate the growth of the GNRs. The final solution was kept at room temperature for 8 hours and GNRs solution was obtained.

Femto-second time-resolved transient absorption (TA) spectrometer. The TA spectrometer is consisted of three basic systems: Femtosecond laser, optical parametric amplifier (OPA) and an optical detection system. Femtosecond laser is generated by a mode-locked Ti:sapphire oscillator-seeded regenerative amplifier (peak wavelength is 800 nm; repetition rate is 1 kHz; pulse width is 100 fs). Then, the beam is divided into two components via a 9:1 beam splitter. The major component is sent to an optical parametric amplifier (OPA, TPR-TOPAS-U, America) to generate the 390 nm (0.8 μJ) pump pulse. The remaining component is sent into a CaF₂ crystal to generated the probe pulse (Spectrum range: 420-750

nm). The probe pulse is carefully automatic adjustment delay time with temporal resolution of 150 fs between pump pulse through a delay device controlled by the computer. After the pump pulse and the probe pulse with a slight angle were beamed through the sample, the intensities of the spectrally of probe pulses are monitored by a spectrometer (Ultrafast Systems). All the transient absorption measurements were carried out at room temperature.

Supporting Video

Video 1 records the spiral interferogram of the 543.6 nm (green) OVBs with OAM₊₁.

Video 2 records the spiral interferogram of the 543.6 nm (green) OVBs with OAM₋₂.