Plasmon-Mediated Radiative Energy Transfer across a Silver Nanowire Array via Resonant Transmission and Subwavelength Imaging

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Directional control over the excitation energy transfer between different nanosystems is of critical importance for the emerging field of nanophotonics and has various prospective applications ranging from biological detections to quantum information processing.1–14 For instance, the excitation energy transfer between semiconductor quantum dots (QDs) is employed to demonstrate quantum operations,4 and the stimulated interactions between active optical dipoles and surface plasmons are used to generate plasmonic lasing.5–8 Comparing with nonradiative energy transfer (such as Dexter and Förster processes),15,16 radiative energy transfer has sufficient distance range but poor efficiency and directionality. The surface plasmons of the exquisitely designing and optimizing metal nanostructures are powerful tools to enhance the efficiency of both radiative and nonradiative energy transfers.1,17 The Ag films have been used by Andrew et al. to first demonstrate plasmon-mediated radiative energy transfer from donor to acceptor dye molecules over distances longer than 100 nm,1 which proceeds in three processes, converting optical dipole of the donors to the surface plasmon on one interface of a Ag film, then cross coupling of two surface plasmons on the opposite interfaces of the film, and finally transferring excitation energy to the acceptors on the opposite side. On the basis of this principle, the corrugated nanostructures have been used by Feng et al. to enhance the cross coupling of the two surface plasmons on the opposite interfaces.17

A two-dimensional (2D) standing metal nanowire array could be a good candidate to assist radiative energy transfer due to its near-field coupling and imaging behaviors.20,21 The excitation energy of nanoemitters can be efficiently converted to the surface plasmons through strong coupling near the tips of the Ag nanowires,10 and the corresponding Purcell factor, P (the ratio of the spontaneous emission rate into the plasmon modes over emission into other channels), is predicted to be as high as 103.22,23 Metal nanorods support both longitudinal and transverse surface plasmon resonances (abbreviated by LSPRs and TSPRs, respectively) by the free electrons near the metal surface oscillating perpendicularly to and along the long axis of the nanorods. Resonant transmission through a Au nanorod array with a far-field excitation is reported by Lyvers et al.,24 which is found to be caused by the half-wave plasmon modes induced by TSPRs in the nanorod array. Furthermore, Ono et al. have reported that the metal nanorod array

**ABSTRACT**

Efficient plasmon-mediated excitation energy transfer between the CdSe/ZnS semiconductor quantum dots (QDs) across the silver nanowire array up to 560 nm in length is observed. The subwavelength imaging and spectral response of the silver nanowire arrays with near-field point-source excitations are revealed by theoretical simulations. Our studies demonstrate three advantages of the nanosystem: efficient exciton—plasmon conversion at the input side of the array through near-field strong coupling, directional waveguidance and resonant transmission via half-wave plasmon modes of the nanowire array, and subwavelength imaging at the output side of the array. These advantages allow a long-range radiative excitation energy transfer with a high efficiency and a good directionality.

**KEYWORDS:** surface plasmon · silver nanowire array · radiative energy transfer · semiconductor quantum dots · subwavelength imaging
has strong near-field focus effects and can generate subwavelength images of a z-polarized nanoemitter at the opposite side of the array when the emission wavelength of the nanoemitter is resonant with LSPRs of the metal nanorods. In this paper, we theoretically investigate half-wave plasmon oscillations, resonant transmission, and subwavelength imaging behaviors of the Ag nanowire arrays with near-field point-source excitations and experimentally demonstrate exciton energy transfer between CdSe/ZnS QDs and the corresponding dynamic processes across the nanowire array with length up to 560 nm.

RESULTS AND DISCUSSION

Theoretical Simulations of Subwavelength Imaging by Silver Nanowire Arrays. We begin the studies with computational simulations of the field distributions of a point-source coupling to the surface plasmon of a Ag nanowire array. The simulations are performed by using finite difference time domain (FDTD) method with commercial software FDTD Solutions 6.0.5. In the simulations, the point-source is situated at \(z = 0\) nm and the distance between the point-source and the input side (left end) of the nanowire array is \(z_m = 20\) nm (see Figures 1a and 2a), and the 2D hexagonal Ag nanowire array has a period \(a = 100\) nm, wire diameter \(d = 50\) nm. The refractive index of the medium is 1.61.

When a point-source is located at the central axis of a nanowire (as shown in Figure 1), only z-polarized source can be strongly coupled to the nanowire array via excitation of LSPRs. The propagating field along the nanowires and the output field at the opposite side reach the maxima at one or multiple half-wave resonances of the plasmons (the number of half-wave oscillations is expressed by \(m\)). Figure 1a shows the plasmon resonances with \(m = 7\) at \(\lambda = 590\) nm for the nanowire array with length \(L = 540\) nm (the simulations with \(m = 1, 2,\) and 3 are presented in Figure S1 in Supporting Information). The coupled electromagnetic field is mainly confined and propagates near the surface of the central Ag nanowire. It clearly indicates that the electromagnetic field of a point-source strongly couples to and resonantly transmits through the nanowire array via half-wave plasmon modes.

After resonant transmission through the nanowire array, the subwavelength images of the point-sources are generated at the output side (right end) of the array due to the near-field imaging effect. Figure 1b

![Figure 1. Seventh harmonic half-wave plasmon resonances of the Ag nanowire array (\(d = 50\) nm, \(a = 100\) nm, and \(L = 540\) nm) and the subwavelength imaging of a z-polarized point-source at the center of a nanowire. (a) Field distributions along the nanowires (\(x = 0\), YOZ plane). (b) Subwavelength image at the output plane (\(z_{out} = 24\) nm, XOY plane). The dashed circles represent the positions of the Ag nanowire array. (c) Cross-sectional field distributions at \(x = 0\) and \(z_{out} = 14, 18, 24, 30,\) and \(40\) nm.](image-url)
shows the subwavelength image at the plane with a distance $z_{\text{out}} = 24$ nm to the output side of the array via the seventh half-wave plasmon mode ($m = 7$). The $y$ cross-section field distributions at $x = 0$ and $z_{\text{out}} = 14, 18, 24, 30, \text{and } 40$ nm are presented in Figure 1c, which demonstrates the formation of the subwavelength image at $z_{\text{out}} = 24$ nm, and the full width at half-maximum (fwhm) of the image is as small as $1.4d$. Note that the fwhm of the subwavelength image increases from $\sim 70$ to $\sim 105$ nm when the distance $z_{\text{out}}$ increases from 24 to 100 nm (see Figure S3 in Supporting Information). Interestingly, a $y$-polarized source is more efficiently coupled to the nanowire array than a $z$-polarized source when it is located in the middle of two nanowires (see Figure S2 in Supporting Information), and two subwavelength images are generated around two nanowires in this case. Figure 2a shows the modulated half-wave oscillations ($m = 7$) of a $y$-polarized near-field source with $\lambda = 590$ nm, and the lateral field distribution around a single nanowire is asymmetric due to the transverse plasmons.

Comparing with a single nanowire, the Ag nanowire array has strong lateral near-field interaction between the nanowires as well as large effective lateral cross section, which significantly increases the plasmon coupling to the $y$-polarized sources (see Figure S4 in Supporting Information). Therefore, the nanowire array generates subwavelength images with much stronger intensity. The collaboration of the longitudinal and transverse plasmons of the nanowire array leads to efficient output of the subwavelength images.

In the spectral domain, the output intensity $|E_{\text{out}}|^2$ reaches the maxima (i.e., resonant transmission) when the excitation wavelength of the point-source is resonant with the harmonic half-wave plasmon modes of the Ag nanowire array. The spectral responses of the Ag nanowire array ($L = 540$ nm) with near-field excitation are shown in Figure 3, where the output field intensities are read out at the plane $z_{\text{out}} = 24$ nm. Three resonant transmission peaks appear near 540, 590, and 650 nm in the visible region, which correspond to the sixth, seventh, and eighth harmonic plasmon resonances, respectively. Note that the spectral responses of the Ag nanowire array for the $y$-polarized and $z$-polarized point-sources are only slightly different, which means that the LSPRs of the nanowires play a key role in the half-wave plasmon modes of the array for either $y$-polarized or $z$-polarized excitations. We also note
that the short nanowire array with a single half-wave plasmon resonance ($m = 1$) for z-polarized point-sources corresponds to Ono’s superlens. The fwhm value of the subwavelength image of the z-polarized source is almost independent of the value of $m$, but the longer Ag nanowires ($L = 220$ nm) supporting the third plasmon mode ($m = 3$) can focus slightly more energy in the center peak of the near-field images (see Figure S1 in Supporting Information).

The theoretical simulations shown in Figures 1 – 3 reveal that a point-source (acting as a donor) can be efficiently coupled to the Ag nanowire array, and the subwavelength images can be generated at the opposite side. When an acceptor is located around the subwavelength images of the donor, efficient plasmon-mediated radiative energy transfer from the donor to the acceptor could be expected in this nanosystem.

**Nanostructures and Absorption Spectra of Silver Nanowire Arrays.** We turn to experimental demonstrations of long-range excitation energy transfer between QDs by using Ag nanowire arrays, which are electrochemically grown in anodic aluminum oxide (AAO) templates. Figure 4a,b shows the scanning electron microscopy (SEM) images of the AAO templates loaded with the Ag nanowires at the open-pore side and occupied side, respectively. In Figure 4b, the thin Al$_2$O$_3$ barrier layer on the AAO template is removed by sputtering technique to uncover the Ag nanowires. A cross-section SEM image of the AAO loaded with Ag nanowires is shown in Figure 4c. A transmission electron microscopy (TEM) image of the Ag nanowires prepared with electrodeposition charge $Q = 300$ mC is shown in Figure 4d. From the SEM and TEM images, we estimate that the Ag nanowire arrays have average wire diameter $d = 36 \pm 3$ nm, period $a = 97 \pm 5$ nm, and length $L = 560 \pm 40$ nm.

The absorption spectra of the Ag nanowire arrays with different lengths from 38 to 650 nm (the corresponding electrodeposition charge $Q$ from 20 to 360 mC) are shown in Figure 5. In Figure 5, a $p$-polarized source with an incident angle $60^\circ$ is used to excite LSPRs and TSPRs simultaneously. The absorption bands near 400 and 700 nm are caused by TSPRs and LSPRs of the Ag nanowire array, respectively. As the average Ag...
Radiative Energy Transfer between QDs across Silver Nanowire Arrays. The radiative energy transfer is monitored by photoluminescence (PL) emissions. As shown in Figure 6a, an s-polarized laser with a wavelength of 400 nm is slantwise (with Brewster angle $\theta_s \approx 50^\circ$) incident to the input sides of the samples, in which the Ag nanowires grow in the AAO template with the donor QDs_D absorbed on the surface of the Al$_2$O$_3$ barrier layer and the acceptor QDs_A deposited in the rest of the nanopores of the AAO template. The laser with a short wavelength of 400 nm is blocked by the Ag nanowire array, while the emissions from the donors with a longer wavelength of 560 nm can transmit through the array. Figure 6b shows the PL spectra of the donor-only, acceptor-only, and donor-acceptor samples (all the samples have the same wire length of $\sim 560$ nm). For the donor-only sample, the PL ($I_D$) spectrum shows a peak at $\sim 560$ nm, which red shifts about 15 nm compared with that of the dilute QDs_D solution in the absence of Ag nanowire array. This red shift of the PL peak is attributed to the nonradiative Förster energy transfer between the QDs_D in the assembled film and the transmission response of the Ag nanowire array. For the acceptor-only sample, the PL signals ($I_A$) are not observed in the whole detection region due to the complete block of the direct laser excitations by the Ag nanowire array. For the donor-acceptor sample, a new PL peak at $\sim 660$ nm from the acceptor QDs_A is observed, which is attributed to the radiative energy transfer from the donors to the acceptors. If the areas under the donor-only, acceptor-only, and donor-acceptor spectra from 450 to 780 nm are $I_{D}$, $I_{A}$, and $I_{D-A}$, respectively, then the energy transfer efficiency is $\eta_{\text{ET}} = (I_{D-A} - I_{D} - I_{A})/I_{D-A}$, where the correction factor $f$ can be calculated from the ratio of the donor emission intensities at 560 nm in the two spectra of donor-acceptor and donor-only samples. 1 From the experimental data shown in Figure 6b, the energy transfer efficiency $\eta_{\text{ET}}$ is calculated to be about 0.7, which means that 70% of the energy transmitted through the Ag array is coupled to the acceptors. Note that the local electric field around the acceptor QDs is enhanced about 2 orders of magnitude in the presence of the Ag nanowire array (see Figure S4 in Supporting Information for details), which indicates that the Ag nanowire array greatly enhances the energy transfer from the donors to the acceptors. The energy transfer efficiency can be further improved by fine-tuning the wire length and decreasing wire length variation.

The PL intensity of the acceptor ($I_{A}$) in the donor-acceptor sample has the relationship

$$I_{D-A}^{(A)} \propto N^{(D)} \Gamma^{(D)} (\lambda_{\text{exc}}) I_{\text{exc}}^{(D)} \Gamma_{\text{rad}}^{(D)} \Gamma_{\text{nonrad}}^{(D)} T^{(D)} (\lambda_{\text{em}}) N^{(A)}$$

(1)

where $N^{(D)}$ and $N^{(A)}$ are the number of the donors and acceptors.
acceptors, respectively. \( \Gamma_{\text{rad}} \) and \( \Gamma_{\text{nonrad}} \) are the radiative and nonradiative decay rate of the donor QDs, respectively; \( |\mathbf{\mathbf{E}}^f(\lambda_{\text{exc}})|^2 \) is the field enhancement factor of the donors at the excitation wavelength, and \( \mathbf{T}^f(\lambda_{\text{exc}}) \) is the near-field transmittance of the array at the emission wavelength of the donors; \( |\mathbf{\mathbf{E}}^f(\lambda_{\text{exc}} = 400 \text{ nm})|^2 \) is calculated by using FDTD method, and the maximal enhancement factor reaches about 8 near the end of the nanowires. The geometry is the same used in Figure 1, but the excitation source is replaced by a 400 nm s-polarized plane wave with an incident angle of 50°.

The dependence of the acceptor PL \( I_{\text{PL}} \) on the coating layers of the donor films (\( n_\text{layer} \)) is shown in Figure 6c. The value of \( I_{\text{PL}} \) is almost zero for the three coating layers of the donor films due to the quenching effect of the Ag nanowire array. This quenching effect can be efficiently depressed by increasing the thickness of the spacing between the QDs and the Ag nanowires. \( \delta_0 \) dramatically increases when \( 4 < n_\text{layer} < 10 \), which is attributed to the effects of the large field enhancement factor \( |\mathbf{\mathbf{E}}^f(\lambda_{\text{exc}})|^2 \) and the strong near-field exciton–plasmon coupling at both excitation and emission wavelengths of the donor QDs.

**Dynamics of Plasmon-Assisted Radiative Energy Transfer between QDs.** The dynamics of exciton–plasmon–exciton–photon conversion and excitation energy transfer are monitored by using a time-correlated single-photon counting (TCSPC) system. The PL decay rate of the donor QDs \( f_D \) is significantly fastened by the Ag nanowire array, as shown in Figure 7a. The PL decay trace of the QDs on the barrier layer of pure AAO template is single exponential, which means that the cooperative effects caused by the assembly of QDs can be neglected. The corresponding PL lifetime is measured to be about 10.7 ns. On the contrary, the PL of 10 coating layers of donor QDs transmitted through the Ag nanowire array decays in a two-component exponential form

\[
I(t) = A_f e^{-t/\tau_f} + A_s e^{-t/\tau_s}
\]

where \( A_f \) and \( A_s \) are the weight factors of the fast and slow decay processes, respectively; \( \tau_f \) and \( \tau_s \) are the corresponding lifetimes. By fitting the recorded data, we obtain \( \tau_f = 2.2 \text{ ns}, \tau_s = 10.7 \text{ ns} \), \( A_f/(A_f + A_s) = 0.64 \), and \( A_s/(A_f + A_s) = 0.36 \). The enhancement factor of the PL decay rate of the donors reaches as high as 4.9, which confirms a strong near-field coupling between the donors and the Ag nanowire array. The long lifetime contribution is from the donor QDs with longer distance to the nanowires, which are coupled to the plasmons weakly. Figure 7b shows the PL decay rate of the acceptors, which is slightly enhanced by the Ag nanowire array due to the large separation between the acceptors and nanowires because of short deposition time \( t_d \) (red curve in Figure 7b). As the deposition time is prolonged, the separation decreases, and the emission field of the acceptors is strongly coupled to the nanowires, which leads to a faster PL decay rate of the acceptor QDs (black curve in Figure 7b).

**CONCLUSIONS**

In summary, the Ag nanowire array enhances the excitation energy transfer between QDs by sequential processes: first, enhancing the excitation of excitons in the donors, then converting excitons to surface plasmons through near-field coupling, followed by resonant transmissions of surface plasmons via half-wave modes, consequently generating subwavelength images at the opposite side of the array through near-field imaging effect, and finally converting the excitation energy from the images of the donors to the acceptors. The FDTD simulations reveal a more than 10² enhancement of the electromagnetic field by the Ag nanowire array over a distance longer than 500 nm. The measured radiative energy transfer efficiency between the QDs reaches about 70% at the output side of the array. The nanowire array demonstrates the conversion of exciton–plasmon–exciton–photon with a higher conversion efficiency compared with single nanowire, and...
it assists radiative energy transfer with a longer transmitting distance compared with the Ag film. In addition, the array of longer Ag nanowires also supports multiple half-wave oscillations at multlwavelength and can be used for multicolor subwavelength imaging similarly to the stacked array of the short Ag nanorods (the exquisitely stacked arrangement could be better for color imaging). The metallic nanowire array with half-wave plasmon resonances gives rise to several prospective applications in nanophotonics.

**METHODS**

**Preparation of Samples.** The AAO templates were prepared by a two-step anodization process. First, the aluminum sheets (purity 99.99%) were degreased in acetone and electropolished under a constant voltage condition of 16 V for 4 min in a mixture of HClO4 and C2H5OH at 0 °C to smooth the surface morphology. In the first and second anodization processes, treated aluminum sheets were exposed to a 0.3 M oxalic acid solution under constant voltage of 40 V in an electrochemical cell at a temperature of about 4 °C. The alumina layer produced by the first anodization process was removed by wet chemical etching in a mixture of phosphoric acid (0.15 M) and chromic acid (0.60 M) at 60 °C for 1 h. The barrier layer of AAO templates was thinned by chemical pore widening and stepwise potential reduced anodization steps. First, the thickness of the barrier layer was decreased by chemically widening in oxalic acid solution at 30 °C for 3 h. Then the AAO templates were anodized with a stepwise reduced potential from 40 V down to 1 V in oxalic acid solution, and the barrier layer was further thinned.

The ordered array Ag nanowires were deposited in the nanopores of AAO templates by pulsed direct current (DC) electrodeposition in an electrolyte containing AgNO3 (0.03 M) and H2SO4 (0.03 M) with a Pt counter electrode. The deposition pulse was a 6 ms negative potential of 6 V, followed by a 6 ms positive potential of 6 V and a zero potential delay of 400 ms. The length of Ag nanowires was controlled by the deposition integral coulomb charge. The deposition was carried out by a galvanostat/potentiostat (PAR VersaSTAT3-200). The underlying Al substrate was dissolved by using CuCl2 solution.

The donor QDs_D were spin-coated and electrostatically adsorbed onto the surface of the Al2O3 barrier layer of the AAO template layer-by-layer. The freshly cleaned Al2O3 barrier layer was functionalized with poly(ethyleneimine) (PEI) to produce a positively charged surface, then a 150 μL droplet of 0.4 μM negatively charged QDs_D solution was dropped on the substrate and rotated for 30 s at 1000 rpm. This procedure was repeated until the needed layers of the QDs_D were achieved. The donor QDs_D and the acceptor QDs_A are commercial carboxyl CdSe/ZnS core–shell QDs, which were purchased from Invitrogen Corporation. The emission peaks of the QDs are adjusted by their sizes. The donor and acceptor QDs have emission intensities of phosphoric acid (0.15 M) and chromic acid (0.60 M) at 60 °C for 1 h. The barrier layer of AAO templates was thinned by chemical pore widening and stepwise potential reduced anodization steps.

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**Characterization of Samples.** The filled AAO templates were etched by a precise Ar ion polishing system (Gatan PIPS Model 691) and examined by SEM to determine the degree of pore filling. The SEM was performed by using a FEI SEM Sirion 200 operated at an accelerating voltage of 25.0 kV. The TEM was performed by using a JEOL 2010HT operated at 100 kV. The TEM samples were prepared by dissolving the AAO template containing Ag nanowires in NaOH solution. The absorption spectra were recorded by a UV–vis–NIR spectrophotometer (Varian Cary 5000) by using a p-polarized source with an incident angle of 60°.

**Optical Experiments.** The PL from the samples was collected by the reflection measurement. An s-polarized laser for the measurements of PL was generated by a mode lock Ti:sapphire laser (Cary 500) by using a pump–polarized source with an incident angle of 60°. The excitation wavelength was tuned to about 400 nm, which was near-resonant to the TSPRs of the Ag nanowire array. Therefore, the incident local field at the input side of the nanowire array was enhanced strongly, but the transmission of the excitation laser through the nanowire array was efficiently blocked. The scattering noise was filtered by a band-pass filter (BPF), following by a 70 mm focal length lens, which was used to excite the sample at a Brewster angle θb ≈ 50°. The luminescence from the sample was collected by the focusing lens and filtered by a couple of filters that consisted of a long wave pass filter (LWPF) and a short wave pass filter (SWPF) before entering the spectrometer detector (Avantes AvaSpec-2048TEC). The time-resolved PL decay traces were recorded by using a time-correlated single-photon counting system (PicoQuan GmbH). The PL measurement precision in our experiments for the samples with multilayered donor QDs reaches about 3% error range due to the high sensitivity of the detector and the strong PL signal from the assembled QDs.

**Computational Simulations.** The computational simulations were performed by using the FDTD method with perfectly matched layer (PML) boundary conditions. The cell size was 2 × 2 × 4 nm3. The nanowires were hexagonally arrayed with rod diameter = 50 nm and spacing = 100 nm. The refractive index of the matrix was 1.61, and the complex dielectric constants of the Ag were taken from Palik’s handbook.

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**Supporting Information Available:** Rod length dependences of the plasmon resonances and subwavelength imaging, excitation—polarization dependences of the plasmon resonances and imaging, output position dependences of the subwavelength image, plasmon coupling efficiency of a dipole to the silver nanowires. This material is available free of charge via the Internet at http://pubs.acs.org.

**REFERENCES AND NOTES**


