Plasmonic keys for ultra-secure information encryption

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Arbitrary 3D polarization orientation is used to excite surface plasmon resonance in gold nanorods, creating highly secure encryption 'keys.'

As the need for information generation and storage increases, so does demand for information security systems. One way to achieve a high level of security is to use optical techniques to encrypt information in the polarization state of a writing beam, requiring a polarization 'key' for the visualization of the original information.

We can use gold nanorods to create these 'plasmonic' keys for encryption. By exciting the surface plasmon resonance in the nanorods, we increase scattering and local field enhancement. These effects can have a variety of applications, such as boosting the efficiency of light-trapping in photovoltaics and increasing photoluminescence quantum yields in metallic nanoparticles.

The excitation of surface plasmon resonance in gold nanorods is dependent on the polarization orientation of the incidence beam, so only nanorods that are aligned parallel to the polarization orientation of the beam can be excited efficiently. Until recently, we could only generate strongly enhanced longitudinal polarization by focusing a radially polarized light through a high numerical aperture (NA) objective. Here we describe how we achieved arbitrary 3D polarization orientation in the focal region using diffraction of a configured single beam (see Figure 1).

We configured the input polarization of the single beam by superimposing a radially polarized beam and an azimuthally polarized beam with weighting factors $\gamma$ and $\delta$, respectively, at the back aperture of a high NA objective: see Figure 1(a). We further modulated the amplitude of the superposed beam using an apodizer with function $P(\alpha, \varepsilon)$, where $\alpha$ is the azimuthal angle of the transmission aperture and $\varepsilon$ the normalized radius of the obstruction within the apodizer. By configuring the polarization and the amplitude of single beams, we could freely tune the out-of-plane orientation ($\theta$) and the in-plane orientation ($\beta$) of the focal polarization (see Figure 1).

Figure 1. Arbitrary 3D focal polarization control. (a) Schematic diagram of superposition of a weighted radially polarized beam ($\gamma$) and a weighted azimuthally polarized beam ($\delta$). (b) Configuring the weighting factor $\tan^{-1}\left(\frac{\varepsilon}{\Delta}\right)$ and the normalized radius of the concentric obstruction of the apodizer $P(\alpha = 0, \varepsilon)$ enables an orientation-unlimited flexibility of tuning the ratio of the longitudinal to transverse polarization intensity components $\frac{I_z}{I_p}$ from 0 to over 40, corresponding to a tunability of $\theta$ from $\frac{\pi}{2}$ to approximately 0.

This 3D polarization control allowed us to tailor the excitation of the randomly aligned gold nanorods. Figure 2(a)–(c) shows the calculated two-photon (2p) fluorescence rate images of gold nanorods with in-plane, out-of-plane, and arbitrary 3D alignments interacting with the configured 3D focal polarization. The experimental results in Figure 2(d) and (e) are consistent with the calculation. Once the illumination power exceeds the threshold, it can selectively melt correspondingly aligned gold nanorods.

Figure 2(d) and (e) shows the 2p images of gold nanorods with in-plane (blue) and out-of-plane (red) alignments before and after exposure to the laser illumination, configured with $\theta = \frac{\pi}{4}$ and $\theta \sim 0$, respectively. The results show that the orientation flexibility enables 3D selective melting of gold nanorods with corresponding alignments. We can control the melting rate by the power of the laser illumination, as well as the configured focal polarization orientation. Figure 3 shows nearly identical

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melting rates of gold nanorods with in-plane (blue squares) and out-of-plane (red circles) alignments excited by the illumination configured with $\theta = \frac{\pi}{4}$.

We can use the arbitrary 3D selective melting of gold nanorods as a means to encrypt the ‘key’ polarization information, providing the encryption with a far greater level of security than that of in-plane polarization control methods. In Figure 3, we demonstrate the process using five configured polarization orientations (indicated by the directions of red arrows). We can retrieve the five encrypted patterns only by 2p fluorescence imaging through raster scanning with the ‘key’ polarization orientation, which is the same as that used for information encryption. Otherwise, the information is read out as noise.

We could use arbitrary 3D polarization orientation, as described here, to provide greater information storage capacity without increasing the size of the recording medium. In our future work, we may employ this technique to develop polarization microscopy by tailoring the interaction between light and matter in an orientation-unlimited 3D manner. Possibilities for use include single molecule detection, multi-dimensional optical storage data, 3D displays and spintronics.

**Figure 2.** Selective excitation and melting of gold nanorods. The fluorescence rate imaging of gold nanorods with (a) in-plane, (b) out-of-plane and (c) arbitrary 3D alignment excited by the illumination configured at $\gamma = 1$, $\delta = 0$ and $P(\alpha = 0, \epsilon = 0.7)$. The two-photon fluorescence images of nanorods with in-plane (blue) and out-of-plane (red) alignments before and after high-power illumination under the excitation configured with (d) $\theta = \frac{\pi}{2}$, $\beta = 0$, (e) $\theta = \frac{\pi}{4}$, $\beta = \frac{\pi}{4}$, (f) The two-photon fluorescence contrast reduction of gold nanorods with in-plane (blue squares) and out-of-plane (red circles), respectively, under the excitation configured at $\gamma = 1$, $\delta = 0$ and $P(\alpha = 0, \epsilon = 0)$.

**Figure 3.** Demonstration of ultra-secure polarization information encryption. The red arrows in (a-e) indicate the five configured polarization orientations used for the information encryption at (a) $\theta = \frac{\pi}{2}$, $\beta = 0$, (b) $\theta = \frac{\pi}{4}$, $\beta = \frac{\pi}{4}$, (c) $\theta = \frac{\pi}{4}$, $\beta = \frac{2}{4}$, (d) $\theta \sim 0$, $\beta = 0$, and (e) $\theta = \frac{\pi}{14}$, $\beta = 0 \sim 2\pi$. The scale bar is 10 $\mu$m.

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References