

# Control light propagation and polarization with plasmons for surface-enhanced Raman scattering

Hong Wei<sup>a</sup>, Zhipeng Li<sup>a</sup>, Yurui Fang<sup>a</sup>, Feng Hao<sup>b</sup>, Timur Shegai<sup>c</sup>, Tali Dadosh<sup>c</sup>, Yingzhou Huang<sup>a</sup>, Wenzhong Wang<sup>a,d</sup>, Zhenyu Zhang<sup>e,f</sup>, Gilad Haran<sup>c</sup>, Peter Nordlander<sup>b</sup>, Hongxing Xu<sup>a,g</sup>

<sup>a</sup>Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Box 603-146, Beijing 100190, China; <sup>b</sup>Department of Physics and Astronomy, Department of Electrical and Computer Engineering, Laboratory for Nanophotonics, Rice University, Houston, Texas 77005, USA; <sup>c</sup>Department of Chemical Physics, Weizmann Institute of Science, Rehovot 76100, Israel; <sup>d</sup>Central University for Nationalities, Beijing 100081, China; <sup>e</sup>Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA; <sup>f</sup>Department of Physics and Astronomy, University of Tennessee, Knoxville, TN 37996-1200, USA; <sup>g</sup>Division of Solid State Physics / The Nanometer Structure Consortium, Lund University, Box 118, S-22100, Lund, Sweden

## ABSTRACT

Surface plasmon resonances in metal nanostructures can lead to novel optical properties. The greatly enhanced electromagnetic field makes surface-enhanced Raman scattering (SERS) a highly sensitive spectroscopic technique. We employed Ag nanowires as plasmonic waveguide and achieved remote-excitation SERS at a few molecules level. The junctions between metal nanowires and nanoparticles offer hot spots for SERS, while the enhancement strongly depends on the laser polarization. We studied the polarization dependence in Au nanowire-nanoparticle systems of different geometry. The polarization of Raman-scattered light in SERS is a rarely studied topic. We found nanoantennas composed of a few nanoparticles can manipulate the polarization of emission light. A nanoparticle trimer is the simplest nanoantenna to realize the polarization control. By tuning the position and size of the third particle, emission polarization can be modified in a controllable way. In addition, the refractivity of the surrounding media also plays a crucial role for the emission polarization.

**Keywords:** surface plasmon, surface-enhanced Raman scattering, remote-excitation, polarization rotation

## 1. INTRODUCTION

Control of light on the nanometer scale is an important topic of nanophotonics. Based on the surface plasmon resonance of metal nanostructures, nanoplasmonics has become a booming subfield of nanophotonics. The manipulation of light intensity, propagation and polarization has been investigated in plasmonic nanostructures. As a spectroscopic phenomenon discovered over 30 years ago, surface-enhanced Raman scattering (SERS) has been an active topic of fundamental and applied research [1]. The dominating electromagnetic enhancement in SERS is caused by surface plasmon resonance, which is the typical example to manipulate light intensity with plasmons. In SERS systems, the Raman intensity enhancement can be over  $10^{10}$ , which leads to a high sensitivity and even single molecule can be detected by SERS [2, 3].

Recently, one dimensional metal nanostructures are employed to guide light below diffraction limit [4]. Due to the great potential applications in nanophotonic circuits, plasmonic waveguide is becoming a hot topic [5, 6]. Chemically synthesized Ag nanowires have good crystalline structures and are good candidates as plasmonic waveguides. Both far-

field and near-field properties are explored in Ag nanowire waveguide system [7-9]. Due to the momentum mismatch of photons and surface plasmons, light can not couple into the Ag nanowires from the midsection of the nanowires, and vice versa. Nanoparticles are employed to make up the momentum mismatch and help to couple photons into and out of the nanowire, in which system nanoparticles serve as nanoantennas [9, 10].

Metal nanoantennas can not only enhance the Raman intensity in SERS, but also can modify the light polarization. Polarization is another important property of light, while the manipulation of light polarization on the nanometer scale is a challenging subject in nanophotonics, which is less investigated compared with the intensity and propagation.

In this presentation, we summarize our recent studies on SERS, mainly include ref.[10-13]. For nanowire-nanoparticle system, surface plasmons can propagate in the nanowire and couple out as photons at the nanoparticle location on the nanowire. The coupling between the nanowire and nanoparticle can generate hot spots for SERS. By focusing the laser on the nanowire tip, propagating surface plasmons can be launched. If a molecule is located in the junction between the nanowire and nanoparticle, the Raman scattering can be excited remotely by means of propagating surface plasmons. The sensitivity of this unique excitation geometry is extremely high, and can allow single molecules detection. Interestingly, simultaneous multi-site remote SERS sensing can also be achieved. This new approach to SERS detection may have a variety of uses, providing improved detection of chemical information in less accessible geometries, for example, inside living cells, where the label-free detection method of SERS would yield highly valuable chemical information. For the wire-particle system, SERS are also studied by direct excitation. Large enhancement for Raman-scattered light in the wire-particle junction is found to depend strongly on the laser polarization. In many coupling nanostructures, the SERS enhancement is dependent of the incident laser polarization, since the coupling between nanostructures depends strongly on the direction of the incident electric field. On the other hand, the coupling between nanostructures can also induce the change of Raman polarization. We found that the polarization of Raman-scattered light can be strongly modulated by asymmetric Ag nanoparticle aggregates, of which a Ag nanoparticle trimer is the simplest nanostructure for such modulation. The multinanoparticle nanoantennas can serve as a polarization rotator and achieve the management of emission polarization on the nanometer scale by changing the geometry and surrounding media of the antennas.

## **2. EXCITING SERS BY PROPAGATING SURFACE PLASMONS**

The high sensitivity of SERS makes it have many applications and potentials in chemical and biological sensing and detections. In usual SERS measurements, the laser is focused directly on the position where Raman signal will be collected. Whereas, the separation of excitation and detection sites will provide flexibility in specific applications and new scheme design for SERS detection. By launching propagating surface plasmons in Ag nanowires, light can be guided tens of micrometers away from the excitation sites. Based on this, and considering the greatly enhanced electric field in the nanowire-nanoparticle junction, remote-excitation SERS can be achieved in Ag nanowire-nanoparticle system.

The Ag nanowires and nanoparticles are prepared by chemical method [14]. Figure 1a and 1b show the scanning electron microscopy (SEM) images of the typical Ag nanowire and nanoparticle mixtures as synthesized. The crystalline wires can serve as waveguides to guide light below diffraction limit. For a Ag nanowire with a particle adsorbed on as shown in Fig. 1c, when the 632.8 nm laser is focused on the wire tip, surface plasmons will be launched and propagate along the nanowire and finally couple out as photons at the discontinuities of the nanowire (nanoparticle location and another end of the nanowire in this system) as shown in Fig. 1d.

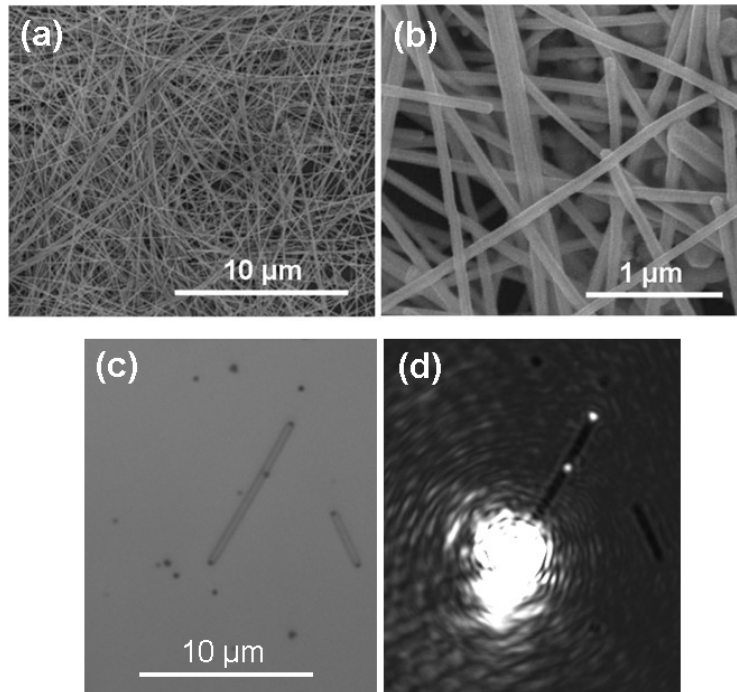


Figure 1: (a, b) SEM images of Ag nanowires with different magnifications (reproduced from ref. [10]). (c) White light reflection image of Ag nanowires on gold film. (d) Light guide image of the system in (c).

Figure 2 shows a Ag nanowire-nanoparticle system in which remote-excitation SERS was achieved. The probe molecule we used is malachite green isothiocyanate (MGITC). By focusing the laser light of 632.8 nm on the left wire tip, SERS signal of MGITC is detected at the particle position which is several micrometers away from the excitation spot. The experimental details can be found in ref.[10]. It should be mentioned that simultaneous multisite remote-excitation SERS was also achieved in our study. Both nanowire-nanoparticle junctions and nanowire-nanowire junctions can serve as hot spots for remote-excitation SERS. This multisite sensing capability is not only important for studying the propagation properties of surface plasmons in nanowires, but may be very useful in a variety of contexts, such as probing complex molecular processes in biological systems and constructing functional plasmonic networks.

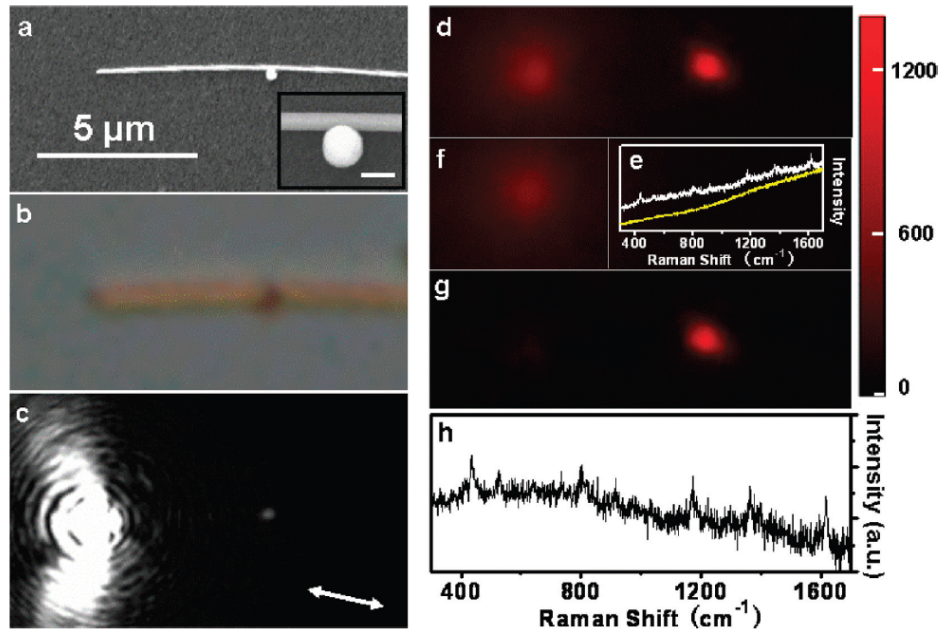


Figure 1: Remote-excitation SERS of MGITC molecules excited through propagating plasmons. (a, b) SEM and white light reflection images of a nanowire-nanoparticle system. (c) Light guide image of the same system by focusing the 632.8 nm laser light on the left nanowire end. (d) Raman image obtained at 436  $\text{cm}^{-1}$  Raman peak. (e) Raman spectra from the laser spot at the left end of nanowire and the remote site of the wire/particle junction, respectively. The remote-excitation SERS spectrum is obtained by separating the Raman collection spot from the laser spot. (f) The fluorescence background image of smooth ITO glass only. (g) The Raman image after background subtraction of panel f from panel d. (h) The remote-excitation SERS spectrum after fluorescence background correction with the spectra taken from panel e. (reproduced from ref. [10])

In the system we studied, the remote-excitation SERS has a very high sensitivity. It was achieved at single molecule level. To further validate our experiment results, we did electromagnetic calculations using the commercial COMSOL Multiphysics software package. The calculated remote-excitation SERS enhancement at the nanowire-nanoparticle junction is about  $10^7$  to  $10^8$  for a perfect solid nanosphere next to a cylindrical nanowire, which should have underestimated the actual enhancement since the imperfections in the experimental system can greatly enhance the local field. In addition, the MGITC molecules have an absorption peak centered around 633 nm. So the resonant Raman scattering will contribute a lot to the detected Raman signal. Therefore, the single molecule level sensitivity can be achieved for remote-excitation SERS in Ag nanowire-nanoparticle system. The calculated results also show that the remote-excitation SERS enhancement is only weakly dependent on the polarization of the laser light.

### 3. SERS INTENSITY DEPENDENCE ON INCIDENT LASER POLARIZATION

The largest electromagnetic field enhancement for SERS usually occurs in the junctions of nanoparticle aggregates. The strength of electromagnetic coupling between nanoparticles depends strongly on the polarization of excitation light, so the SERS intensity also shows strong polarization dependence [15]. For the Au nanowire-nanoparticle coupled system, we investigated how the Raman intensity of probe molecules (MGITC) in the nanowire-nanoparticle junction depends on the polarization of incident laser. In Fig 3a, a Au nanowire-nanoparticle aggregate is shown. From the Raman mapping image we can see that the nanowire-nanoparticle junctions are hot spots for SERS. Furthermore, the enhancement in these hot spots depends strongly on the laser polarization. The inset in Fig 3b shows a nanowire-

nanosphere system. The Raman spectra measurements show that the Raman intensity is strongest when the laser polarized across the nanowire and the nanoparticle. Finite element analysis simulations using COMSOL Multiphysics software show that the electric field in the nanowire-nanoparticle junction is much stronger for perpendicular polarization to the nanowire than for parallel polarization, as shown in Fig 3c.

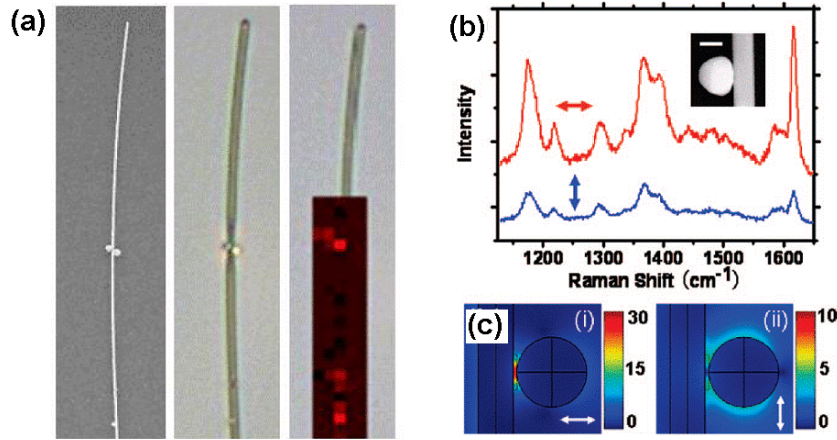


Figure 3: (a) SEM, optical microscopy and Raman image of a Au nanowire-nanoparticle aggregate. (b) SERS spectra of MGITC at two different polarizations for the nanowire-nanoparticle shown in the inset. (c) Calculated electric field for a gold sphere of radius 50 nm at 5 nm from a wire of radius 25 nm for perpendicular (i) and parallel (ii) polarization to the wire. (adapted from ref. [11])

For SERS, the Raman enhancement is proportional to  $E^4$ , which is the product of electromagnetic (EM) enhancement factor and Raman emission (RE) enhancement factor. EM enhancement is due to the local electromagnetic field enhancement associated with the incident light, while RE enhancement is caused by the antenna effect of the nanostructure. For the junction between a nanowire and a nanoparticle, the EM enhancement is proportional to the

square of the local electric field in the junction, i.e.  $G_{EM} = |E_{loc}(\omega_L, \theta) / E_0(\omega_L)|^2 = |E_{max}(\omega_L) / E_0(\omega_L)|^2 \cos^2(\theta)$ ,

where the  $E_0(\omega_L)$  is the incident electric field,  $E_{max}(\omega_L)$  is the maximum of the local electric field  $E_{loc}(\omega_L, \theta)$ ,  $\omega_L$  is the laser frequency and  $\theta$  is the polarization angle of the incident light with respect to a surface normal of the wire pointing to the nanoparticle. The RE enhancement is independent of the incident polarization since the induced electric

field is always in the direction across the junction, and is proportional to  $G_{RE} = |E_{max}(\omega_R) / E_0(\omega_R)|^2$ , where  $E_0(\omega_R)$  is the corresponding incident field at the Raman frequency  $\omega_R$ ,  $E_{max}(\omega_R)$  is the maximum of the local electric field  $E_{loc}(\omega_R, \theta)$  for the optimal polarization. Therefore, the total SERS enhancement factor is proportional

to  $G_{EM} G_{RE} = \left| \frac{E_{max}(\omega_L)}{E_0(\omega_L)} \right|^2 \left| \frac{E_{max}(\omega_R)}{E_0(\omega_R)} \right|^2 \cos^2(\theta) \approx \left| \frac{E_{max}}{E_0} \right|^4 \cos^2(\theta)$ . The calculated SERS enhancement results agree qualitatively with the experimentally measured Raman intensity.

Besides nanospheres, other shapes of nanoparticles in close proximity of a nanowire are also studied. In Fig 4, the Raman enhancement of three different systems is studied both experimentally and theoretically. It can be seen from Fig 4 that the calculations agree well with the experimental results, and SERS intensity has a  $\cos^2(\theta)$  dependence on the polarization of the excitation light. For perpendicular polarization, the calculated Raman intensity mainly comes from the nanowire-nanoparticle junction. For parallel polarization, the junction only contributes a small part to the calculated Raman intensity, while the outer surface of the nanoparticle contributes the dominant part. These results show that the polarization dependence of SERS in the nanowire-nanoparticle junctions is remarkably insensitive to the detailed

geometrical structures of the nanoparticles. Plasmon hybridization method was employed to analyze the plasmon coupling between the nanoparticle and the nanowire. The discrete plasmon states of the nanoparticle will couple to the continuous plasmon states of the nanowire when excited by incident light. And the coupling efficiency depends strongly on the incident polarization.

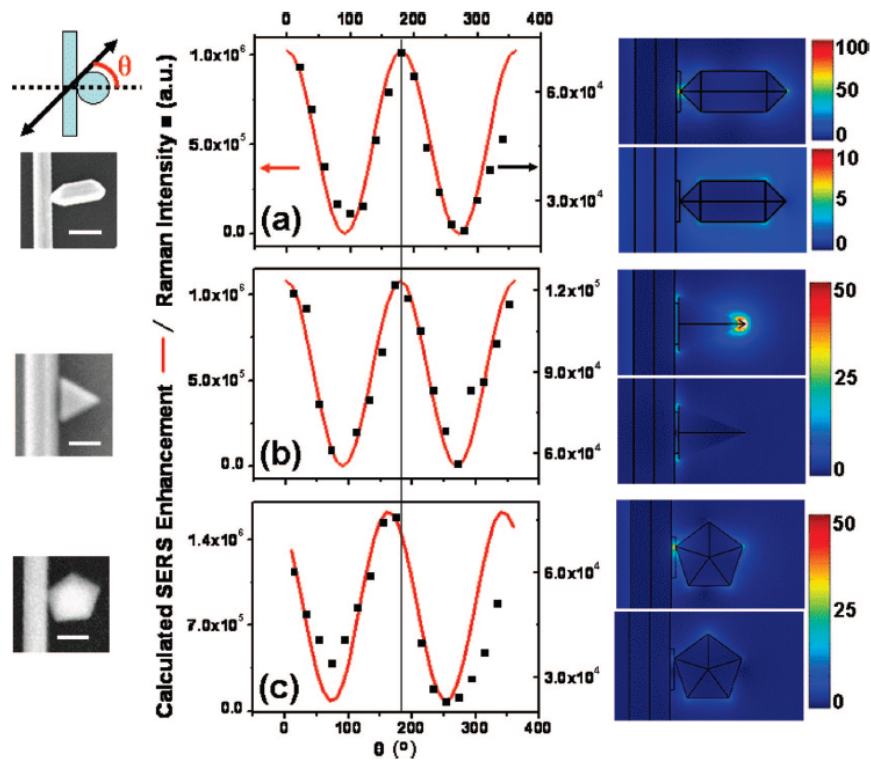


Figure 4: Measured (squares) and calculated (lines) SERS intensity as a function of polarization angle  $\theta$  defined in the inset for different shape nanoparticles adjacent to a nanowire. The SEM images of the wire-particle system investigated are shown on the left, and the electric field distributions from theoretical calculations are shown on the right for perpendicular (upper plot) and parallel polarization (lower plot). The scale bar in the SEM images is 1  $\mu\text{m}$  in (a) and 200 nm in (b) and (c). (reproduced from ref. [11])

#### 4. ASYMMETRIC NANOANTENNAS FOR POLARIZATION CONTROL OF EMISSION LIGHT

One challenging topic about the manipulation of light on the nanometer scale is the control of light polarization. Aggregates of metal nanoparticles can serve as nanoantennas to enhance the Raman emission intensity. In addition, these multiparticle nanoantennas can also manipulate the Raman polarization. Nanoparticle dimer, a simple nanoantenna, has been used in single molecule SERS study about ten years ago [2]. The strong plasmon coupling between nanoparticles results in greatly enhanced electric field in the junctions between the nanoparticles. However, much less is known about how these multiparticle nanoantennas affect the polarization of emission light.

We employed single molecule SERS to investigate the effect of nanoantenna on the Raman polarization. Experimentally, nanoantennas are composed of Ag colloidal nanoparticles prepared by the citrate reduction method of Lee and Meisel [16]. Raman measurements were performed on a home-built Raman spectrometer. The laser polarizations were varied by a halfwave plate, and the Raman-scattered light was split into two orthogonal components ( $I_{\parallel}$ ,  $I_{\perp}$ ). The depolarization

ratio  $\rho$  can be obtained by  $\rho = \frac{I_{\parallel} - I_{\perp}}{I_{\parallel} + I_{\perp}}$ . For the dimer shown in Fig 5a, the Raman intensity changed with the laser

polarization and maximized when the laser polarized parallel to the dimer axis, as shown in Fig 5b. The depolarization ratio of the Raman-scattered light is wavelength independent and reaches the maximum value of 1 when the incident light polarized along the dimer axis, as can be seen in Fig 5c, indicating the emission light is linearly polarized. Calculation results using generalized Mie theory (GMT) agree well with the experimental results.

For the trimer nanoantenna, the properties are quite different. For a nanoantenna composed of three nanoparticles, there are three possible locations for the molecule. Fig 5d shows an example. The Raman intensity in Fig 5e reaches maximal value at an angle of about  $75^\circ$ , which is not the direction of the axis of any two particles. GMT calculation results show that when the molecule is in the junction between particle 2 and 3, the experimental and the theoretical results are in very good agreement. The axial symmetry of the dimer composed of particle 2 and 3 is broken by particle 1, which results in the rotation of the Raman intensity and depolarization ratio patterns (Fig 5e and 5f). As can be seen from the patterns in Fig 5f, the depolarization ratio never reaches 1, which means the emission light is not linearly polarized anymore, but elliptically polarized. Furthermore, the depolarization ratio is wavelength dependent. The symmetry break in the nanoantennas leads to significant changes in the optical properties.

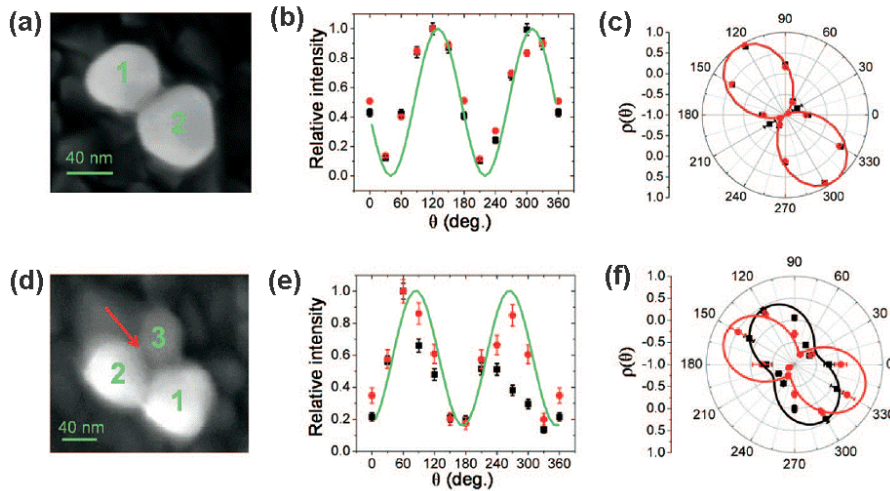


Figure 5: Polarization response of a nanoparticle dimer and a nanoparticle trimer. (a) A SEM image shows a dimer of nanoparticles, which is tilted about  $40^\circ$  from the vertical direction. (b) Normalized Raman intensity at 550 nm (black) and 583 nm (red) as a function of the angle of rotation of the incident polarization. The green line is the result of a GMT calculation. (c) Depolarization ratio measured at 555 nm (black) and 583 nm (red). Black and red lines are the results of GMT calculations performed at 555 and 583 nm, respectively. (d-f) Corresponding data for a nanoparticle trimer. More details can be found in ref. [12]. (adapted from ref. [12])

It should be stressed that in the GMT calculations, only the geometry of the nanoantennas are used as input. To further investigate how the geometry affects the emission polarization, more GMT calculations are performed. Figure 6 shows the polarization rotation as a function of the position and the size of the third particle in a trimer nanoantenna. When the third particle is far away from the dimer, its coupling with the dimer is negligible and the polarization pattern stays the same with the dimer. When the third particle is moved close to the dimer, the plasmon coupling will rotate the emission polarization significantly, as shown in Fig 6a. Figure 6b shows three depolarization ratio patterns. It's clear that both the polarization direction and ellipticity are changed when the third particle is close to the dimer. Besides the position, the size of the third particle is also critical to the polarization modulation. In Fig 6c, the radii of the dimer are 24 nm and 35 nm, respectively. The polarization rotation angle is plotted as a function of the third particle radius (from 10nm to 100 nm) while the interparticle distance is kept at 1 nm. The polarization does not rotate when the third particle is small (radius < 20 nm). As the third particle size increases, both 555 nm and 583 nm Raman-scattered light show a nonmonotonic

polarization rotation. It should be noted that the polarization modulation is only wavelength-dependent when the third particle has a size in a certain range.

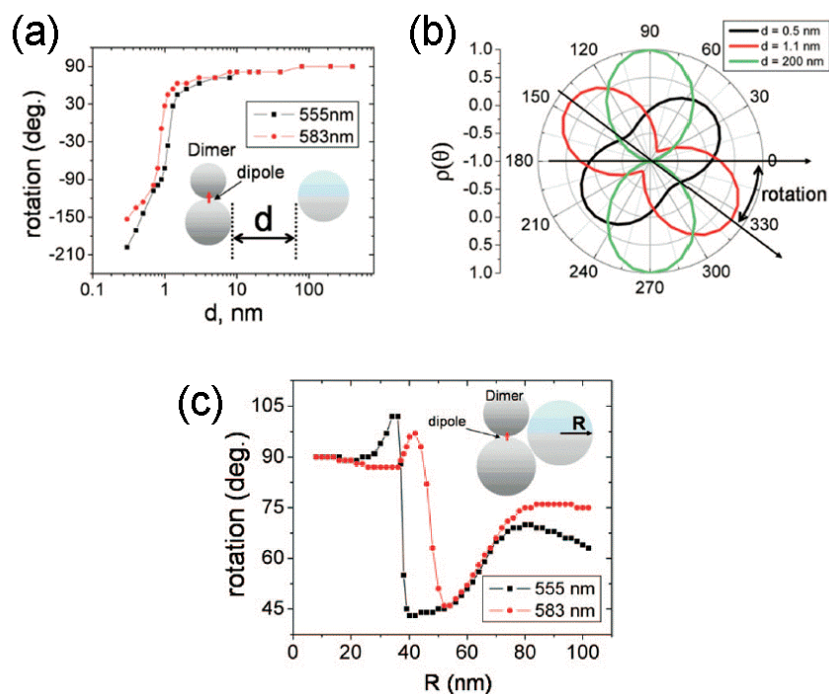


Figure 6: Polarization rotation as a function of the position (a, b) and the size (c) of the third particle in a trimer. (adapted from ref. [12])

The polarization control is studied in single molecule SERS systems, but it is not limited to SERS only. The emission properties of other emitters, such as dye molecules and quantum dots, can also be modulated. We further study the emitter-nanoantenna interactions in a general way using GMT. The emitter is treated as a dipole located in the junction between the nanoparticles with an orientation angle  $\alpha$ , as shown in Fig 7a. When the dipole is aligned with the dimer axis, great emission enhancement over  $10^5$  can be achieved, as shown in Fig 7b, when the nanoparticle radius is 40 nm and separation is 1 nm. When the dipole is rotated away from the dimer axis, the enhancement decreases following a  $\cos^2 \alpha$  relation. Then a third nanoparticle is added to form a trimer nanoantenna, and the emission enhancement factor can be larger than  $10^4$  over a broad wavelength range, as shown in Fig 7c. In both the dimer and trimer antennas, only the emission by the parallel component of the dipole can be enhanced significantly.



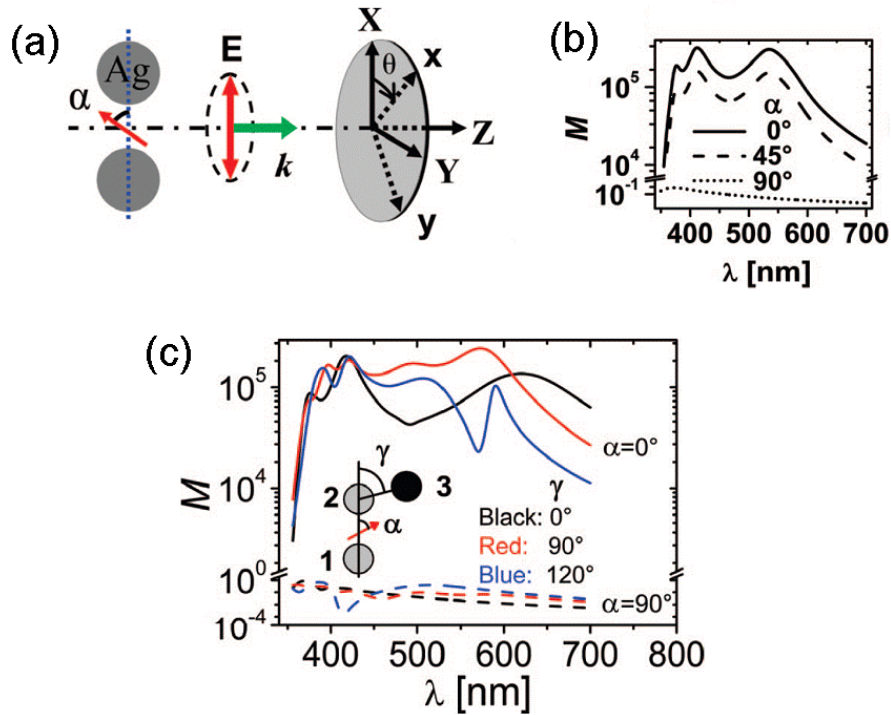


Figure 7: (a) Schematics of dipolar emission from a Ag dimer antenna. A dipole (red arrow) is located in the center between two Ag nanoparticles (radius is 40 nm, separation is 1 nm), with an orientation angle  $\alpha$ . The polarization of the emitted light is projected onto the x and y axes, and two orthogonal components of the emission are calculated for different rotation angles  $\theta$  of the xy axis with respect to the XY system to compute the depolarization ratio defined by  $\rho(\theta) = \frac{I_x(\theta) - I_y(\theta)}{I_x(\theta) + I_y(\theta)}$ . (b) Enhancement factor M of the dipole emission from dimer antenna as a function of wavelength at different orientation angles. (c) Enhancement factor M of the dipole emission from trimer antenna as a function of wavelength for different orientation angles of the dipole and positions of the third particle. (adapted from ref. [13])

For a dimer antenna, the emission polarization is along the dimer axis. When a third particle is introduced to the dimer axis, since the axial symmetry holds, the emission polarization of the linear trimer stays the same as the dimer, as shown in Fig 8a. When the third particle is moved clockwise away from the dimer axis, the emission polarization also rotates clockwise. After reaching the maximum rotation angle of about  $40^\circ$  (Fig 8b), the polarization will rotate in the reverse direction and moves back to  $0^\circ$  (Fig 8c). In the equilateral triangle geometry shown in Fig 8c, new symmetry is created and the third particle couples symmetrically with the other two particles, so the emission polarization is the same as the dimer antenna. In Fig 8d, the polarization angle and maximum depolarization ratio of the antenna emission are shown as a function of angle  $\gamma$ . As  $\gamma$  increases from  $0^\circ$  to  $120^\circ$ , the polarization angle increases first and then decreases to go back to  $0^\circ$ . At the maximal polarization angle, the depolarization ratio maximum is 0.7, which indicates that the emitted light is a bit elliptically polarized. As the size of the third particle increases, larger polarization rotation can be achieved. If a fourth particle is introduced, the emission polarization can also rotate further. It should be noted that the emission polarization is only determined by the antenna configuration, independent of the dipole orientation. For a trimer antenna shown in Fig 7c, the dimer composed of nanoparticle 1 and 2 determines the emission enhancement, while the nanoparticle 3 modifies the emission polarization.

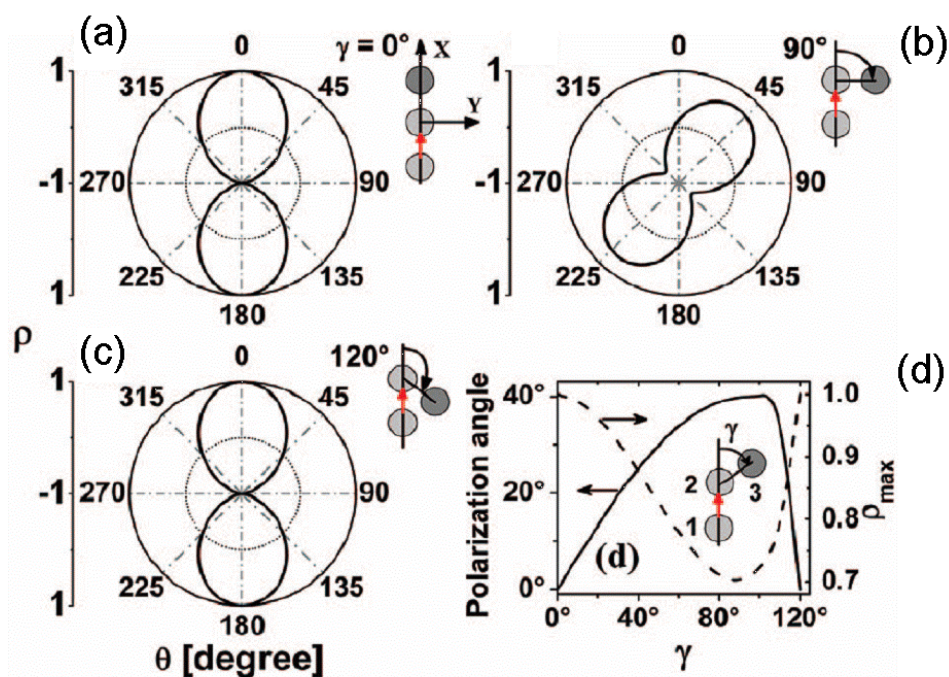


Figure 8: Depolarization ratio of the dipole emission from a Ag trimer nanoantenna with (a) linear, (b) right-angle, and (c) equilateral triangle configurations. (d) Polarization angle and the maximum depolarization ratio of the antenna emission as a function of angle  $\gamma$ . The radii of all three particles are the same (40 nm). The separations between particles 1-2 and 2-3 are kept at 1 nm. The wavelength of the dipole emission is 555 nm. (reproduced from ref. [13])

Besides the geometry of the nanoantennas, the refractivity of the surrounding medium ( $n_s$ ) can also influence the emission polarization significantly. Figure 9a shows the polarization angle as a function of  $n_s$ . When  $n_s$  is changed, the plasmon resonances will also be changed. So the polarization angle can be quite different when  $n_s$  is different. The emission enhancement factor also depends on the refractivity of the media, as shown in Fig 9b. The sensitive dependence of emission polarization on  $n_s$  can be very useful to control the emission polarization of dipoles by changing the refractivity of the medium. This property can also be used to design refractivity sensors based on the polarization rotation.

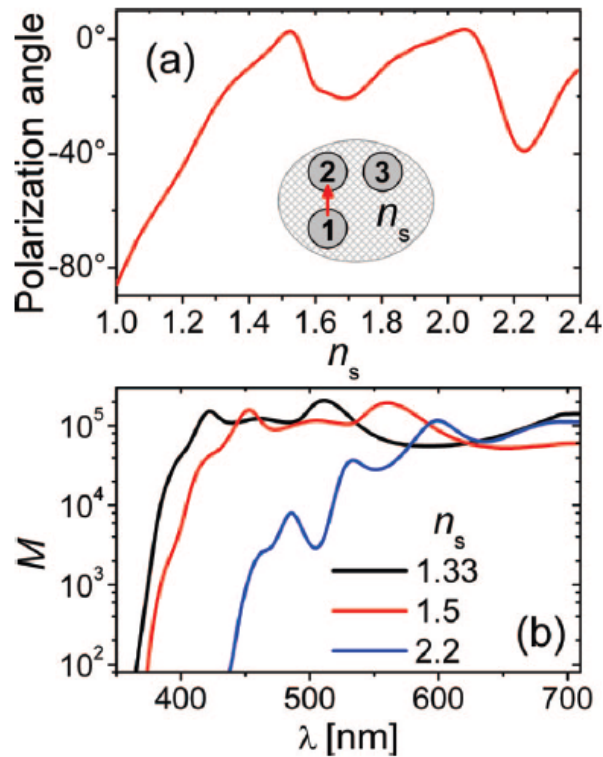


Figure 9: (a) Polarization angle of the emission from a right-angle trimer ( $R_1 = R_2 = 40$  nm,  $R_3 = 80$  nm) as a function of the refractivity of the surrounding medium  $n_s$ . The dipole, emitting at 555 nm, is denoted by a red arrow. The separation between the nanoparticle 1-2 and 2-3 is kept at 1 nm. (b) Emission enhancement from the same dipole-trimer system as a function of the emission wavelength of the dipole, for three different values of  $n_s$ . (reproduced from ref. [13])

## SUMMARY

By combining plasmonic waveguide and SERS, we achieved remote-excitation SERS in Ag nanowire-nanoparticle systems at single molecule level. This novel excitation scheme makes new applications possible. For the Raman enhancement, we studied the dependence of SERS intensity on the laser polarization in Au nanowire-nanoparticle systems. The polarization rotation of emission light from multiparticle nanoantennas was studied experimentally and theoretically. A polarization rotator at the nanometer scale can be realized by changing the geometry of the antenna and the refractivity of the surrounding media.

## ACKNOWLEDGEMENTS

We gratefully acknowledge financial support from the following sources: NSFC Grant No. 10625418, 10874233, MOST Grant No. 2006DFB02020, 2007CB936800, 2009CB930700, “Bairen Project” of CAS, the U.S. Army Research Office under Contract No. W911NF-04-1-0203, NSF under Grant EEC-0304097, the Robert A. Welch foundation under Grant C-1222, the historic generosity of the Harold Perlman Family and by the Israel Science Foundation, and the Swedish Research Council (VR). This work is also supported by Division of Materials Sciences and Engineering, Office of Basic Energy Sciences, Department of Energy Grant DEFG0205ER46209, and National Science Foundation Grant DMR-0606485.

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