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Detection of microscope-excited surface plasmon polaritons with Rayleigh scattering from metal nanoparticles

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We propose a mapping tool of surface plasmon polaritons (SPPs) excited using an optical microscope. By combining dark-field and confocal microscopy, we can efficiently extract metal nanoparticle-induced Rayleigh scattering from background radiation, thereby leading to state-of-the-art SPP measurements. The method is verified to be sensitive to the dominant perpendicular field component of SPPs and be of high accuracy. We also use this method to reveal the conversion of spin angular momentum of light to the orbital angular momentum of SPPs under tight-focusing conditions. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4827264>]

Surface plasmon polaritons (SPPs) are light-induced charge density oscillations at a metal surface,¹ which, due to their unique features, are used widely in applications such as sensing,² imaging,³ and miniaturized photonic circuits.⁴ Mapping the near-field SPP distribution is crucial for understanding their fundamental properties and facilitating applications. Unfortunately, however, because of their evanescent nature, SPPs are extremely difficult to detect using conventional far-field microscopic systems. A common method is to use a near-field scanning optical microscope (NSOM)^{5–8} in which a bare tapered or metal-coated fiber is dipped into the SPP region to transfer some of its energy to the propagating waves.

However, this direct detection method is very sensitive to perturbations in the local environment in the vicinity of the probe, particularly for SPPs under microscope excitation scheme in which transmitted light from the microscope is very strong; hence, the measured SPP patterns usually deviate significantly from the theoretical patterns.^{9–11} Furthermore, both calculations and experiments have indicated high collection efficiency in the in-plane field,^{12,13} which is a subordinated component of SPPs. This jeopardizes the goal of SPP characterization because the dominant perpendicular field component is not imaged. Although other techniques have been proposed and demonstrated, including fluorescence detection¹⁴ and leakage radiation microscopy,¹⁵ they are restricted by the diffraction limit and are therefore not able to provide high spatial frequency information of SPPs.

In this letter, we propose an SPP detection method based on excitation from a microscope. Microscope excitation of SPPs, which employs a high numerical aperture (NA) objective lens with oil immersion to satisfy the plasmon wave-vector matching condition, shows great potential in applications such as ultrahigh-sensitivity surface plasmon resonance (SPR) sensing,¹⁶ super-resolved imaging,¹⁷ and surface-enhanced Raman spectroscopy.¹⁸ For most applications,

obtaining near-field SPPs is a prerequisite. This requires an efficient surface plasmon detection tool with high accuracy. As demonstrated in this letter, our method is able to detect the dominant perpendicular field of SPPs. This is a powerful surface plasmon detection tool and will be very useful for further exploration of SPPs.

The working principle of the proposed method is illustrated in Fig. 1. A collimated incident beam is first passed through an opaque circular mask to become an annular beam, which is subsequently tightly focused onto a metal/dielectric interface with a high NA oil immersion objective lens to excite SPPs on the metal surface. The size of the annular beam is therefore determined by the aperture size of the mask and objective lens. The incident angle is within the range of $\theta_{\text{mask}} < \theta < \theta_{\text{obj}}$, where θ_{mask} is the minimum incident angle restricted by the mask and θ_{obj} is the maximum incident angle allowed by the objective lens. The SPR angle θ_{spr} therefore should be between these limits to excite SPPs on the metal surface. The sample plane is located beneath the focal plane of the objective lens; hence, SPPs counter-propagate toward the center from the excitation ring. A metal nanoparticle (NP), immobilized on the metal surface and functioning as an SPP antenna, is employed to scatter the near-field surface plasmon waves. The induced Rayleigh scattering together with the transmitted annular beam is collected with another objective lens. As can be seen in Fig. 1, because of the vertical displacement between the nanoparticle and the focal point of the incident annular beam, Rayleigh scattering from the NP will be focused at a different point from that of the transmitted annular beam. As a result, by placing a micrometer-sized pinhole in the imaging plane of the collection objective lens—as is performed for confocal microscopy—we can exploit the scattering signals from the background radiation. This is because of the filter-like function of the pinhole in the imaging plane, which can pick up information from the object plane. By raster scanning the NP-film junction over the region of SPPs and recording the scattering intensity at each step, we can obtain a near-field SPP distribution.

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FIG. 1. Configuration of SPP detection method under excitation from a microscope.

We first used the proposed method to map the near-field SPP distribution generated by a tightly focused, linearly polarized Gaussian beam at a wavelength of 532 nm. The NAs of the oil-immersion objective lens (Olympus, APO N, 60 $\times$) and circular mask were 1.49 and 0.87, respectively. The corresponding incident angle of the annular beam was $35^\circ < \theta < 79.8^\circ$. A thin Ag film with a thickness of ~ 50 nm was deposited onto a cleaned cover glass using electron beam deposition. The SPR angle was 44.1° , which was within the annular beam. Ag nanospheres with diameters of ~ 60 nm (nanoComposix) were immobilized on the Ag film to scatter the surface plasmon waves. The immobilization process is described as follows: A droplet of dilute Ag colloids (100 μ l, $\sim 10^9$ nanospheres/ml) was applied onto a Ag film and allowed to evaporate naturally; the sample was ready for use after water-rinsing and air-drying at ambient temperature. A collection fiber (OceanOptics) with a 50- μ m aperture diameter was placed in the imaging plane through a fiber coupler (Thorlabs, F810SMA-543). This fiber acts as a pinhole to select the desired object plane as well as coupling the NP-induced Rayleigh scattering to a photo-multiplier tube (Hamamatsu, H5784-20) for further amplification.

The SPP mapping result is presented in Fig. 2(a), which shows a pattern of two separate lobes centered across the axis of symmetry. This is because of the requirement for p -polarization to excite an SPP. For focused, linearly polarized light, the beam is only fully p -polarized in the direction

parallel to the incident polarization (white arrow), whereas in the other directions, the components of the p -polarized beam are diminished according to Malus's law. The inset in Fig. 2(a) is the calculated perpendicular field (E_z) component in the central area using the Richard-Wolf vectorial diffraction method.^{19,20} There is a good match between the experimental and calculated results. To further verify the E_z component sensitivity of our method, we performed a cross-section comparison between the scanning result and the calculated E_z and E_r components of the SPPs, as shown in Fig. 2(b). In contrast to the E_z component, for which the central spot is dark, there is a bright fringe in the center for the E_r component because of constructive interference, and its bright fringes are interlaced. The comparison clearly demonstrates that our approach is able to efficiently detect the dominant E_z component of SPPs. Moreover, the excellent agreement between the experimental and theoretical results also indicates that the metal NP on the Ag film have little influence on SPPs or at least do not change the near-field profile.

We further applied our method to measure the plasmon wavelength, which is an important parameter determining the dispersion relationship of SPPs. The standing wave in Fig. 2(a) can be used to obtain the plasmon wavelength. However, this is not a precise method because the periodicity of the fringes in the central area is larger than half the SPP wavelength. A better approach is to generate a regular standing wave and use it to scale the plasmon wavelength. Here, we use radially polarized light as the excitation source, but instead of full-beam excitation, only one pair of beam segments is employed simply by inserting a rectangular opaque mask. As a result, SPPs are excited from a pair of arcs and counter-propagate toward the center (Fig. 3(a)). As long as the expanding angle θ of the arcs is small enough, a standing wave with uniform periodicity will be generated.

Fig. 3(b) shows a calculated SPP standing wave between the excitation arcs when the expanding angle is 40° . The period of the standing wave is 257 nm. Note that this is not exactly the same as half the plasmon wavelength (253 nm), but we can use this standing wave to scale the plasmon wavelength. The corresponding mapping result and cross-section comparison are presented in Figs. 3(c) and 3(d), respectively. We can see that the measured pattern is in good agreement with the calculated one. The period is measured to be 261 nm. This leads to a plasmon wavelength equal to 514 nm ($261/257 \times 506$ nm), which is 1.6% larger than the theoretical value. The slight increase of λ_{SPP} is believed to

FIG. 2. SPP mapping results for a tightly focused, linearly polarized gaussian beam, verifying the E_z component sensitivity of our method. (a) Two dimensional mapping result (unit: V) along with the calculated E_z component (inset). The white arrow indicates the incident polarization. (b) Cross-section comparison between the scanning result (red) and the calculated E_z (green) and E_r (blue) SPP components.

FIG. 3. Measurement of SPP wavelength. (a) Schematic illustration of excitation configuration of a regular SPP standing wave for the plasmon wavelength measurement. A pair of SPP excitation arcs is induced by tightly focused, radially polarized light with only two beam segments. (b) Calculated SPP standing wave when the expanding angle of the arcs is 40° . (c) Corresponding measured SPP pattern, which agrees well with the calculated pattern. (d) Cross-section comparison. The measured period is 1.6% larger than that from the calculation. Scale bar in (c) is $1\ \mu\text{m}$.

be due to the modification of Ag permittivity from the bulk ($-10.1786 + 0.8238i$; this value was also used in the calculation) to the thin film.

Finally, we applied the proposed method to reveal the conversion of spin angular momentum (SAM) of light to the orbital angular momentum (OAM) of SPPs. The angular momentum, which includes SAM and OAM, is an important property of light that is used widely in applications such as particle manipulation, microscopic imaging, information communication, and quantum computation.^{21–23} The photonic SAM involves the polarization state of the photon, whereas the OAM relates to its phase (or wavefront). Recently, it was demonstrated that the SAM/OAM of light is able to convert to the OAM of surface plasmons through plasmonic vortex lenses,²⁴ and the OAM of SPPs was directly visualized through a particle trapping experiment.²⁵ Here, we demonstrate that the SAM to SPP-OAM conversion can also be realized through a tight-focusing configuration, and, by virtue of the proposed SPP detection method, the conservation of angular momentum number during this conversion is confirmed experimentally.

Fig. 4(a) gives the SPP pattern calculated using the Richard-Wolf vectorial diffraction method when the incident light is circularly polarized ($s=1$). The pattern shows a series of concentric rings and is verified to be the same as that of a plasmonic vortex (PV) with topological charge

FIG. 4. Revealing the conversion of spin angular momentum of light to the orbital angular momentum of SPPs under tight-focusing condition. (a) Calculated near-field SPP pattern under a tightly focused circularly polarized beam (spin angular momentum, $s=1$), showing a plasmonic vortex with topological charge “1” (orbital angular momentum, $l=1$). The inset gives the phase distribution in the central area. (b) Experimental mapping result (unit: V), in line well with the calculated pattern. The area of each contour map is $4\ \mu\text{m} \times 4\ \mu\text{m}$.

$l=1$. Generally, a PV with topological charge l is defined as²⁴

$$J_l(k_{\text{SPP}}r) \exp(jl\varphi), \quad (1)$$

where k_{SPP} is the SPP wavenumber, r and φ are the radial and azimuthal coordinates, and $J_l()$ is the Bessel function of first kind with order l . Kim *et al.*²⁴ reported that the PV carries an OAM proportional to the topological charge l . As a result, by tightly focusing circularly polarized light onto a metal/dielectric interface, the SAM of light is converted to the OAM of SPPs with conservation of the angular momentum number. From Eq. (1), the topological charge of a PV can be determined either by the primary ring size from the intensity distribution (left term) or by the spiral phase within the primary ring from the phase distribution (right term). The inset in Fig. 4(a) further confirms the topological charge of the obtained PV.

In the experiment, a circularly polarized beam with $s=1$ was tightly focused onto the silver/air interface to excite SPPs. The mapping result is shown in Fig. 4(b) and has a high degree of accordance with the theoretical result. The measured primary ring size is 295.6 nm . This is very close to the value predicted from the Bessel function (295 nm for $l=1$), which therefore verifies the topological charge of the obtained PV and the conservation of angular momentum number.

To summarize, we have developed a state-of-the-art SPP detection method, which, by combining the concept of dark-field microscopy (annular beam) and confocal microscopy (pinhole in the image plane of the objective lens), allows the Rayleigh scattering of SPPs induced by metal nanoparticles to be efficiently extracted from the background radiation. The perpendicular field sensitivity of our method is confirmed by comparing the mapping results with the calculated perpendicular and in-plane field components. This represents a significant advance because SPPs are strongly dominant in the perpendicular field, whereas conventional NSOM detection generally yields only in-plane field information. We further applied our method to perform systematic measurements of the SPP wavelength and verified the accuracy of these measurements. Finally, this method was applied to reveal the conversion of the SAM of light to the

OAM of SPPs. Owing to the high precision of the technique, we were able to confirm experimentally the conservation of angular momentum number during the conversion. We believe that our method can be employed as a powerful surface plasmon detection tool, with significant contributions to further investigation of SPPs in both fundamental science and the development of important applications.

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