Abstract  Leaky plasmon modes (LPMs) in metal nanowires (NWs), which combine the physical characteristic of both “plasmonics” and “leaky radiation”, present distinguished performances in terms of guiding and radiating light. In contrast to traditional light-guiding in metal NWs with one single LPM, multiple LPMs are crucial for advanced uses such as augmenting data transmission channels, enhancing sensing performance, manipulating polarization and converting mode. Here, we demonstrate experimentally the control over multiple LPMs in pentagonal silver NWs. By combining far-field real-space imaging and leakage radiation microscopy, the three typical LPMs with fields mainly concentrating in corners surrounded by air are specifically identified. By manipulating excitation wavelengths and NW diameters, the number of the excited LPMs can be controlled. These findings reveal the physics of LPMs in silver NWs, thereby paving the way towards applying the high-order leaky modes in silver NWs for photonic integrated circuits, nanoscale confinement, plasmonic sensing, QD-nanowire coupling, etc.

Identification and control of multiple leaky plasmon modes in silver nanowires

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1. Introduction

Surface plasmon polaritons (SPPs), which are optically induced oscillations of free electrons at metal surfaces, hold the ability to confine light well beyond diffraction limit and to transport this energy over several micrometers [1–4]. Owing to these distinguished attributes, SPPs show significant potential for miniaturizing optical devices to the scale comparable with modern nanoelectronic devices and integrating optics and electronics on a single chip [4, 5]. Among various SPP-based nanostructures, chemically synthesized silver nanowires (NWs) [6] possess two distinctive advantages: (1) monocrystalline character results in low inner and surface scattering losses [7, 8]; (2) one-dimensional character contributes to ease of manipulation and assembly. The silver NWs have thus far been utilized as nano-waveguides [9–17], logic nano-gates [18, 19], nanorouters [20–24], nano-sensors [25–27], nano-antennas [26, 28–30], quantum-dot-nanowire couplers [31–33], etc., which has also been reviewed in reference [34].

For employing silver NWs for transporting light, they are typically deposited on a dielectric substrate and the leaky plasmon modes (LPMs) can be readily excited in this asymmetrical configuration (in contrast with NWs immersed in a dielectric surrounding) [26, 28, 35–39]. These LPMs are distinguished from bound plasmon modes by their lower effective refractive indices in comparison with those of substrates; therefore, their fields expand gradually and a portion of energy subsequently leaks into the substrates during propagation. As a result of combining the physical characteristic of both “plasmonics” and “leaky radiation”, the LPMs in silver NWs present several advantageous traits: (1) their propagation losses can be even lower than those of bound plasmon modes due to a decreasing fraction of energy confined within the silver [37, 39]; (2) enhanced light-matter interaction (such as coupling between polaritons and excitons in QDs, the interaction between polaritons and chemical molecules in sensors) can benefit from the leaky radiation of these LPMs [31]; (3) the leaky radiation itself provides an extra avenue for detection [26]. The LPMs have been mainly studied by implementing three strategies: (1) near-field imaging with SNOM [40–42], electron energy loss spectroscopy [17, 43] or photoemission electron microscopy [44], (2) far-field real-space imaging [26, 35, 45–51], and (3) leakage radiation microscopy (also called far-field Fourier-space imaging) [26, 28, 36, 39, 52, 53]. Based on the above approaches, a single LPM in a silver NW has been experimentally reported so far [26, 28, 36, 53]. References [26, 28] studied the control over radiation direction of single leaky plasmon mode on silver nanowires. Reference [36] theoretically predicted that multiple leaky modes can be supported
in a silver NW but only observed one leaky mode in experiment. Actually, multiple LPMs can be potentially exploited in various applications including augmenting data transmission channels based on mode-division multiplexing [54], enhancing sensing performance [25], manipulating polarization and converting mode [46, 55], etc. For data transmission in plasmonic integrated circuits, the use of Ag NW plasmonic waveguide for information transmission has been experimentally demonstrated in reference [56]. To further augment data transmission channels, mode-division multiplexing with multiple plasmon modes including both guided and leaky modes can be adopted since different modes are orthogonal. For exploiting leaky modes (especially high-order leaky modes) in sensing, enhanced performance (compared with exploiting guided modes in sensing shown in reference [57]) can be envisaged since their fields predominantly concentrate in the Ag/air interface. Besides, the aforementioned near-field imaging and the far-field real-space imaging are meant for the case that one plasmon mode is excited. For the case of two (or more) leaky modes that are simultaneously excited, only their total propagation behavior can be acquired while the propagation behavior for each specific mode cannot be resolved. Therefore, identification of each LPM in silver NWs is of vital importance.

In this paper, identification and control of multiple LPMs simultaneously excited in silver NWs are investigated by means of combining far-field real-space imaging and Fourier-space imaging. Firstly, the mode features of five typical plasmon modes including both bound modes and leaky modes in silver NWs are provided. Secondly, the experimental results for NWs with single, double and triple excited LPMs are presented sequentially. The control over the number of excited LPMs via manipulating excitation wavelengths and NW diameters is investigated. For one single LPM, the effective refractive index is wavelength-dependent whereas insensitive to NW diameters. For double LPMs, obvious beats with a fixed beat wavelength resulting from the interference between the two excited LPMs is obtained in the real-space image and their effective refractive indices are resolved in the Fourier-space image. For triple LPMs, a short excitation wavelength and a large NW diameter (532 nm and 454±5 nm, respectively) are required and the interference leads to complicated beats in the real-space image. Finally, conclusions are presented.

2. Mode analysis

For mode analysis, an infinitely long silver NW with a typical pentagonal cross section is situated on a SiO$_2$ substrate with a refractive index of 1.52. The diagonal length of the pentagon is defined as the diameter $D$ of the NW (also shown in Fig. 1), in accordance with the extracted diameter when the NW is under a scanning electron microscope (SEM). The permittivity data of silver from Johnson-Christy are adopted in simulations [58]. A finite element method (Comsol Multiphysics 4.4 with RF module) is used to investigate the plasmon modes in this configuration (see Supporting Information “Simulation Methods”). As an example, for a 454(±5)-nm-diameter silver NW at 532-nm wavelength, five typical plasmon modes with their mode distributions are illustrated in Fig. 1. These five modes can be divided into two categories:

(1) Bound plasmon modes ($B_1$ mode and $B_2$ mode). For these two modes with effective refractive indices (1.70 and 1.54 for $B_1$ mode and $B_2$ mode, respectively) larger than that of substrate, their fields are well confined on silver/glass interface and no leaky radiation can thus be detected using the far-field Fourier-space imaging. These two bound modes have been well investigated by far-field real-space imaging in reference [14, 15, 44, 45].

(2) Leaky plasmon modes ($L_1$ mode, $L_2$ mode and $L_3$ mode). For these three modes, their fields mainly concentrate in corners surrounded by air; accordingly, their refractive indices (1.09, 1.04 and 0.93 for $L_1$ mode, $L_2$ mode and $L_3$ mode, respectively) approach that (1.053) of infinite silver/air interface plasmon mode. Given that their effective refractive indices are considerably smaller than that of the substrate, a large portion of energy leaks into the substrate during propagation, thus enabling being detected using the far-field Fourier-space imaging.

**Figure 1** Electric intensity of five typical plasmon modes for a 454(±5)-nm-diameter pentagonal silver NW on a SiO$_2$ substrate ($n = 1.52$) at 532-nm wavelength. The two sequential data in the center of each graph denote the effective refractive index and the propagation length (unit: $\mu$m) of each mode, respectively. The length and direction of arrows indicate the intensity and direction of the electric field in the cross-section plane, respectively. For the mode index, the letter $B$ ($L$) represents a bound (leaky) mode; the subscript means its mode number.
Figure 2  The SEM image of a chemically synthesized silver NW whose diameter is 285 ± 5 nm. The inset denotes a zoom-in end face of the NW.

For $L_1$ mode, $L_2$ mode and $L_3$ mode, their respective effective refractive indices decrease sequentially. Their fields predominantly concentrate around the top corner, the two lower corners and the upper three corners, respectively. Note that the propagation lengths for both $L_1$ mode, $L_2$ mode (16 μm and 9.2 μm, respectively) are larger than those of the two bound plasmon modes, indicating the potential of exploiting leaky modes for enhanced light transmission. Among the three leaky modes, the $L_1$ mode can be easily detected while the $L_3$ mode can only be observed in thick NWs at short wavelengths in the far-field Fourier-space imaging, which will be experimentally demonstrated in the following sections.

For these five modes, $B_1$ mode, $L_1$ mode and $L_3$ mode are bilaterally symmetric in terms of the charge density distributions with respect to the axis perpendicular to the NW/substrate interface (see Supporting Information “Charge Density Distribution Models”). Therefore, these three modes can be excited in silver NWs with flat terminations under parallel polarization (with respect to the NW direction). For $B_3$ mode and $L_2$ mode, their charge density distributions are bilaterally anti-symmetric and they can thereby be excited in silver NWs with flat terminations under perpendicular polarization (with respect to the NW direction) [33,46,59].

3. Experiment

The silver NWs used here are synthesized by a soft solution phase approach [6]. Silver NW diameters range from 150 nm to 450 nm with the lengths longer than 20 μm. The silver NWs are deposited on the SiO$_2$ substrate with a reflective index of 1.52. Figure 2 displays an SEM image of a typical NW with smooth surface and excellent uniformity. Before the SEM characterization, an ultrathin gold film is sputtered on the substrate to enhance the electrical conductivity. The termination of NW exhibits a spherical-like shape, which is explicitly demonstrated in the inset of Fig. 2. As has been pointed out in reference [14, 15, 34, 59], symmetric modes ($L_1$ mode, $B_1$ mode and $L_3$ mode here) and antisymmetric modes ($B_2$ mode and $L_2$ mode here) can be excited in silver NWs with flat terminations under parallel and perpendicular polarization (with respect to the NW direction), respectively; while other termination shapes (like the spherical-like shape here), both symmetric modes and antisymmetric modes can be excited regardless of the polarizations. The parallel polarization is found to show a relatively higher coupling efficiency than the perpendicular polarization; therefore, the parallel polarization is utilized in our experiments.

The leaky mode can be interpreted as representing guided modes beyond the cutoff. The leaky modes form a discrete set instead of a continuum, which is the case for the radiation modes. For the guided modes, their effective refractive index is above that of substrate and thereby only evanescent fields exist in the substrate, resulting in no detectable average power in the far field from the substrate side. For the leaky modes, their effective refractive indices are below that of substrate and a portion of energy radiates photons into the substrate during propagation. This leaking provides an avenue to access its propagation behaviors in the far field [60, 61].

In experiment, the angular distribution of the light intensity of the LPMs can be characterized by the angle $\theta$ defined in Fig. 3 and directly imaged in the Fourier plane of the microscope’s objective. As is shown in Figure 3, the propagation of LPM along the NW is denoted by $k_{\text{SPP}}$. Owing to the leaking characteristic, the LPM can be projected onto the Fourier plane for detection at the resonance angle $\theta_{\text{SPP}}$ given by $k_{\text{SPP}} = k_0 n_{\text{sub}} \sin \theta_{\text{SPP}}$.

The LPMs of NWs are characterized by a homemade confocal inverted microscope for both real-space imaging and Fourier-space imaging. The experimental set-ups for real-space and Fourier-space imaging are shown in Fig. 4. In both cases, the output of a laser (blue path) goes through
a polarizer and a beam splitter sequentially. The light is then focused on a NW termination to excite the LPM with a high numerical aperture immersion objective (Nikon, NA = 1.49). For the real-space imaging (green path in Fig. 4(a)), the leaky radiation is first collected by the same objective and then imaged on the intermediate image plane by an optical 4f system consisting of the objective and AL1. After that, the intermediate image is projected on a CCD through lens combination (AL2 and AL3). For Fourier-space imaging (orange path in Fig. 4(b)), the leaky radiation is first collected by the same objective as real-space imaging and imaged on the back focal plane of this objective. Then the image is projected on a CCD through lens combination (AL1 and AL2). In this case, AL3 is flipped out. In both real-space and Fourier space imaging, an adjustable stop (AS), which can be freely adjusted in size, position and even rotation in the intermediate image plane, is inserted to block the direct scattering light from the excited spot and promote signal noise ratio of the LPMs.

In the Fourier-space imaging, the excited LPMs are discretely projected in Fourier-space planes to retrieve the effective refractive indices. The simulated propagation length is obtained by calculating the complex effective index $n_{\text{eff}}$ (including both real and imaginary parts) of each mode. The imaginary part of the complex effective index is directly related to the propagation length $L$ and working wavelength $\lambda$ by the formula [39]:

$$L = \frac{\lambda}{4\pi \text{Im}(n_{\text{eff}})}$$

The experimental propagation length is obtained by fitting the leaky intensity during propagation in the real-space image. The linear fitting and nonlinear fitting are adopted to obtain the experimental propagation length in both single and double LPMs, respectively. The details for the fitting are provided in the Supporting Information “Fitting for Single LPM” and “Fitting for Double LPMs”. According to the number of the detected LPMs in the silver NW, three sections are divided: (1) single LPM, (2) double LPMs and (3) triple LPMs.

### 3.1. Single LPM

As is pointed out above, the $L_{1}$ mode shows the highest refractive index among three LPMs, rendering it readily detectable even in thin NWs at long excitation wavelengths.

In a real-space image of a NW excited by a focused laser beam at one termination, the luminous lines located along the NW are indications of the radiation from the propagating LPM. Figure 5(a) clearly depicts this behavior for a typical 224(±5)-nm-diameter NW with a 45-μm (only 21 μm is shown in the figure) length at the 640-nm wavelength. Long NWs are utilized with the purpose of avoiding the plasmon resonance owing to the termination reflection. The relative intensity of the LPM is extracted along the luminous lines from the excited spot to the other end. Figure 5(c) presents the natural logarithm of the relative intensity (black and red dots from “Right line” and “Left line”, respectively) as a function of the distance away from the excited spot. These two dot lines almost coincide and both show linear dependence on the propagation distance with a negligible fluctuation due to the smooth surface and monocrystalline character of the silver NW. Therefore, only one LPM propagates in the NW and its propagation length can be calculated from the slope of two dot lines by a fitting method with the formula $I = I_0 e^{-x/r_0}$, where $I_0$ is the relative intensity at the starting point of the selected calculation range, $r_0$ is the $1/e$ damping length (i.e. propagation length) of the LPM, and $x$ is the distance away from the starting point. The fitting line demonstrates good agreement with the experimental data, confirming the validity of the direct measurement method used in this work. The extracted propagation length by fitting is 3.3 μm.
The wave vector of \( \mathbf{L}_1 \) mode is analyzed by Fourier-space imaging. Figure 5(b) displays the distribution of the relative wave vector \( (k_x/k_0, k_0) \) for \( \mathbf{L}_1 \) mode. The calibration of the Fourier space image is implemented based on both total internal reflection (TIR) at the Air/SiO\(_2\) interface and the numerical aperture angle of the objective. The inner circle \( (k_x/k_0 = 1.0) \) refers to the critical angle for total internal reflection (TIR) at the Air/SiO\(_2\) interface. The outer circle \( (k_x/k_0 = 1.49) \) refers to the numerical aperture angle of the objective, which is the maximum angle of the collected light. In the Fourier image, the LPM is recognized as a straight bright line along the perpendicular axis \(+k_x/k_0\), representing a constant effective refractive index in the \( x \) direction and a broad wave vector range in the \( y \) direction, which is a key characteristic of the leaky mode. From the relative intensity along the axis \(+k_x/k_0\) in the Fourier-space image (Fig. 5(d)), the position of the intensity peak presents the effective refractive index of the LPM in the \( x \) direction, and its value is extracted to be 1.04. In the Fourier-space image, the symmetric/antisymmetric behavior along the line is strongly related to the radiation direction of leaky plasmon mode [26]. The detected LRM includes a broad wave vector range in the \( y \) direction, which shows diverse polarization. By placing a polarizer before the Fourier imaging, both symmetric and antisymmetric LRMs can be obtained [26]. Nevertheless, the value of the effective refractive index for the leaky plasmon mode cannot be affected.

By this approach, three typical NWs with diameters of 185 ± 5 nm, 224 ± 5 nm and 303 ± 5 nm, are measured at wavelengths varying from 532 nm to 1000 nm. Such a wide range of wavelengths is achieved by exploiting two single-frequency lasers (532 nm and 640 nm) together with an ultra-narrow linewidth tunable laser (Spectra-Physics, Matisse TX-light, 750 nm-1000 nm). The dependence of the effective refractive indices on NW diameters and excitation wavelengths is illustrated in Fig. 6(a). For the three NW diameters, their effective refractive indices first drop from about 1.12 to about 1.04 with wavelengths increasing from 532 nm to 750 nm, and then fluctuate with wavelengths further increasing to 1000 nm. The effective refractive indices for the 303(±5)-nm-diameter NW are slightly higher than those of the other NWs. Figure 6(b) shows the simulated results. It can be seen that the effective refractive indices decrease rapidly at short wavelengths and slightly at long wavelengths and the effective refractive index for the 303(±5)-nm-diameter NW is generally the highest among those of the three NWs. It can be concluded that the effective refractive index of \( \mathbf{L}_1 \) mode shows little dependence on the NW diameter while it is highly wavelength-dependent, especially at short wavelengths. Therefore, the effective refractive index of \( \mathbf{L}_1 \) mode can be effectively controlled by adjusting the excitation wavelength.

The dependence of propagation lengths of NWs on NW diameters and excitation wavelengths is provided in Fig. 6(c). The propagation lengths for the 303(±5)-nm-diameter NW at 532 nm and 640 nm are not provided because more than one LPM is excited and the propagation length for \( \mathbf{L}_1 \) mode cannot be distinguished solely. For the 303(±5)-, 224(±5)- and 185(±5)-nm-diameter NWs, their propagation lengths are around 6 \( \mu \)m, 5 \( \mu \)m and 4 \( \mu \)m over the experimentally available excitation wavelength range, respectively; therefore, the propagation length of \( \mathbf{L}_1 \) mode can be extended by exploiting a thick NW. In
simulations shown in Fig. 6(d), the propagation lengths varies around 4  μm, 3  μm and 2  μm for the 303(±5)-, 224(±5)- and 185(±5)-nm-diameter NWs, respectively. The  L_1 mode in a thick NW also exhibits an enhanced propagation length. This can be interpreted as follows: as the NW diameter increases, the SPPs between the two interfaces (Ag/Air and Ag/SiO_2) gradually decouple due to the limited penetration depths into metal, resulting in reduced portion of energy within the silver and correspondingly the ohmic loss. The simulated propagation lengths are shorter compared with the experimental results. This discrepancy may be attributed to the reduced losses in chemically synthesized silver NWs in experiments relative to those used in simulations. Besides, the simulated propagation length of the LPM is affected by the substrate thickness.

### 3.2. Double LPMs

For a thick NW and a short wavelength, a high-order LPM (L_2 mode) besides L_1 mode can also be excited. Here, two LPMs (L_1 mode and L_2 mode) are observed in NWs with diameters from 249 ± 5 nm to 407 ± 5 nm at short wavelengths (532 nm and 640 nm) in experiment.

For a 309(±5)-nm-diameter NW, its real-space image at 532 nm is provided in Fig. 7(a). Two bright lines demonstrating typical interference patterns on the two sides can be unambiguously observed. Figure 7(c) presents the extracted intensities along both “Right line” and “Left line” in Fig. 7(a). As the distance away from the excited spot increases, linear superposition of the L_1 mode and L_2 mode with a wave-vector difference leads to beating pattern in the intensity. The obtained beat wavelength, which is a measure of the wave-vector difference between the two modes, is λ_b = 4.6 ± 0.2  μm (the distance between two adjacent peaks or valleys). By fitting the intensity along the NW (see Supporting Information Fig. S3 and Table S2), the propagation lengths for L_1 mode and L_2 mode are 4.84  μm and 2.75  μm, respectively. The propagation length of the beats is related to those for the two SPP modes and the fitted value is 3.51  μm. Moreover, the wave vectors of L_1 mode and L_2 mode are displayed in the Fourier-space image (Fig. 7(b)), where the two LPMs are recognized as two parallel bright lines between two circles. Figure 7(d) also exhibits relative intensity along the axis +k_x/k_0 in the Fourier-space image. The obtained effective refractive indices of L_1 mode and L_2 mode are 1.13 and 1.01, respectively. The corresponding beat wavelength between these two modes can be approximately calculated as L_c = 2π Re ((k_x,L_1 − k_x,L_2)^−1) = 4.4  μm [39], which is in accordance with the measured beat wavelength (4.6 ± 0.2  μm) from the real-space image.

The effective refractive indices of the two LPMs (L_1 mode and L_2 mode) versus NW diameters for the two excitation wavelengths (532 nm and 640 nm) are presented in Fig. 8. The effective refractive indices of L_1 mode are in accordance with those in thin NWs shown in Fig. 6(a, b), further demonstrating their little dependence on the NW diameter and strong dependence on the wavelength. For L_2 mode, the measured effective refractive index at 532 nm is larger than that at 640 nm for a fixed NW diameter. The measured effective refractive index of L_2 mode shows higher dependence on the NW diameter at 532-nm wavelength than 640-nm wavelength, agreeing with the simulated results shown in Fig. 8(b).
Figure 7 Visualization of double LPMs in a silver NW ($D = 309 \pm 5$ nm) at the 532-nm wavelength. (a) and (b) are a real-space image and a Fourier-space image, respectively. The yellow dash lines in (a) present a part of the silver NW (30 $\mu$m of total 52 $\mu$m length). (c) is logarithm of the intensity of the LPMs. The black and red dotted lines are extracted from the “Right line” and “Left line” in (a), respectively. (d) The relative intensity of leaky radiation versus the relative wave vector extracted from the $+k_x/k_0$ axis in (b). The inset in (d) shows the simulated effective indices (1.09, 1.00) and propagation lengths (6.3 $\mu$m, 3.2 $\mu$m) of the leaky modes $L_1$ and $L_2$ in the 309($\pm$5)-nm-diameter silver NW at the 532-nm wavelength, respectively.

Figure 8 Effective refractive indices ($n_{\text{eff}}$) of the two excited LPMs ($L_1$ mode and $L_2$ mode) versus NW diameters for two excitation wavelengths (532 nm and 640 nm). (a) and (b) are experimental and simulated results, respectively. Error bars are calculated from repeated experiments.

3.3. Triple LPMs

For a thicker NW at a short wavelength, besides $L_1$ mode and $L_2$ mode, a higher-order LPM ($L_3$ mode) can also be excited. Here, in a 454 $\pm$ 5 nm NW at the 532-nm wavelength, three LPMs are observed in experiment. Figure 9(a) presents the real-space image, where two bright lines signifying interference patterns on the two sides are evidently observed. In the Fourier-space plane shown in Fig. 9(b), three straight lines denoting the three excited LPMs can be distinguished. The effective refractive indices of the three excited LPMs ($L_3$ mode, $L_2$ mode and $L_1$ mode) are 1.00, 1.06 and 1.09, respectively, obtained from Fig. 9(d). Figure 9(c) shows the linear-superposition-induced interference between the three excited LPMs. Different from the results shown in Fig. 7(c), a constant beat wavelength cannot be extracted in experiment since the interference among three modes is much more complicated. (See Supporting Information “Fitting for Triple LPMs”)

4. Conclusion and Outlook

The LPMs in NWs, which present combined physical characteristics of both “plasmonics” and “leaky radiation”, show several advantageous traits in terms of low-loss propagation, enhanced light-matter interaction and accessible far-field radiation. The excitation of multiple LPMs in NWs are of vital importance for exploiting the high-order modes for augmenting data transmission channels based on mode-division multiplexing, enhancing sensing performance, manipulating polarization and converting mode, etc. The existing near-field imaging with SNOM or photoemission electron microscopy and far-field real-space imaging fail in the cases when more than one plasmon mode is excited. Here, by combining far-field real-space imaging and Fourier-space imaging, the three typical LPMs in pentagonal silver NWs are specifically identified. By manipulating the excitation wavelengths and the NW diameters, the number of the excited LPMs is controllable. The fields of the
three LPMs in silver NWs mainly concentrate in corners surrounded by air and their refractive indices are measured to be close to that of silver/air interface mode. These demonstrations reveal the physics of LPMs in silver NWs and thus create new opportunities for potentially utilizing low-loss metal NWs supporting LPMs as a basic building block for photonic integrated circuits, nanoscale confinement, plasmonic sensing, QD-nanowire coupling, etc.

Supporting Information

Additional supporting information may be found in the online version of this article at the publisher’s website.

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