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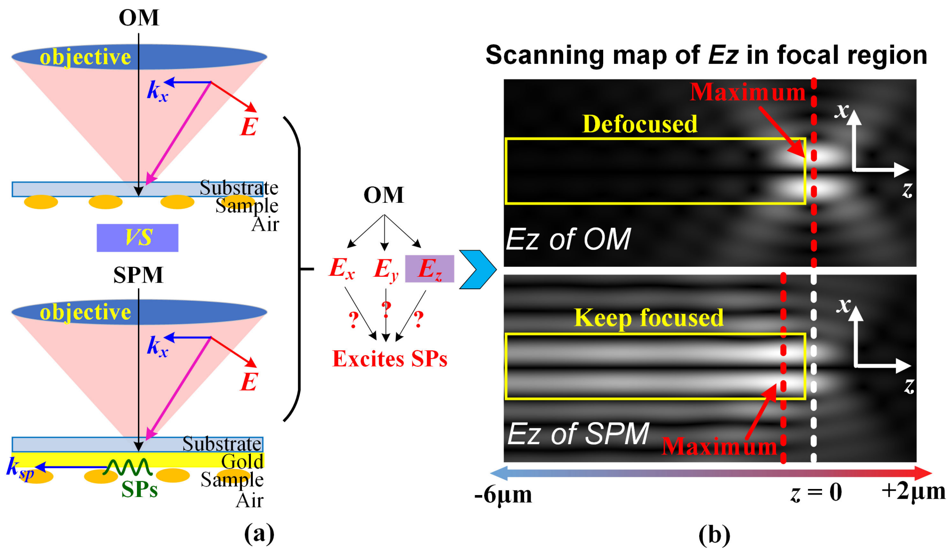

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Surface Plasmon Microscopy Versus Optical Microscopy: Ez Dominates in SPM

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Abstract: Objective-coupled surface plasmon microscopy (SPM) shows extremely high similarity with conventional optical microscopy (OM) in both configuration and theoretical model. And there is a common misunderstanding in SPM that all the three polarization components Ex, Ey, and Ez of the focused beam contribute to excitation of SPs, similar as the situation in OM that all the polarization components generate image contrast. Actually, this is not the case and some literatures have suggested 'only Ez excites SPs'. However, so far there has been no related theory to support this issue, and the common misunderstanding and corresponding errors are still applied in both academia and industry. The present work clarifies this significant issue for the first time. We theoretically prove that 'only Ez excites SPs' and further give two related phenomena as evidences.

Index Terms: Surface plasmon microscopy, surface plasmons, optical microscopy, Ez polarization, excitation.

1. Introduction

Objective-coupled surface plasmon microscopy (SPM) combines the high sensitivity of surface plasmons (SPs) and high lateral resolution of conventional optical microscopy (OM). It has been widely used in some application sceneries of biochemical sensing and life science which require high sensitivity and resolution [1], [2]. Fig. 1 shows the configurations of conventional OM and SPM respectively. One can observe the extreme similarity between the two configurations. Actually, due to the extreme similarities of configuration and model between SPM and OM, one tends to implant the features of OM into SPM directly and ignore the distinctive interaction between the focused beam and excited plasmonics in SPM. One common misunderstanding is that all the three polarization components *Ex*, *Ey*, and *Ez* of focused beam generated in OM contribute to excitation of SPs in SPM. However, is this the case?

In the excitation of SPs, there were two well-known conditions: *i)* the dispersion relation [3] and *ii)* only the TM mode (p-polarization) can excite SPs [4]–[6]. However, neither of the two conditions clarified which of *Ex*, *Ey*, and *Ez* excited SPs. Several groups claimed that only *Ez* excited SPs. For instance, Kano reached the conclusion based on the consideration that the experimentally acquired focal spot of SPM was consistent with that of *Ez* polarization component [7]. Argoul [8] and Zou [9] adopted '*Ez* excites SPs' directly in their studies. More specifically, Ref. [8] claimed that only the component of *Ez* dominated the resolution of SPM and Ref. [9] pointed out that only the component

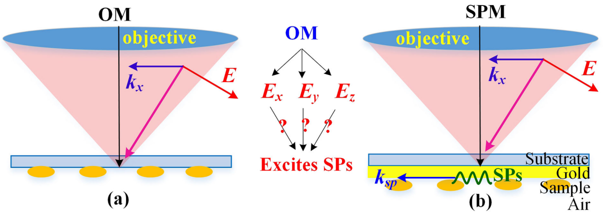


Fig. 1. (a) Configuration of conventional OM; (b) Configuration of SPM.

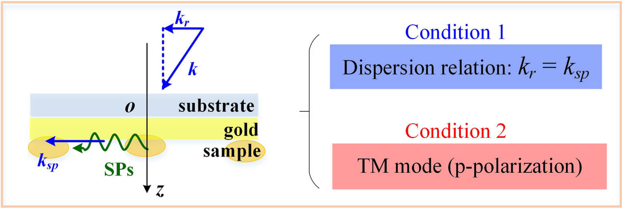


Fig. 2. Two fundamental conditions about SPs excitation: dispersion relation and TM mode matching.

of *Ez* should be taken into account in calculation of plasmonic fields. However, there has been no rigorous theory to prove this fact. And the question that which of *Ex*, *Ey,* and *Ez* excited SPs is still ambiguous and the common mistake is still widely applied. For example, the calculation in Ref. [4] considered all the three components to investigate the property of SPs even though *Ex* and *Ey* are independent with SPs excitation; and Ref. [1] directly took the resultant spot size of the three components as the theoretical resolution of SPM.

This work aims to theoretically prove that SPs are excited by the component of *Ez* polarization in objective-coupled SPM. We also demonstrate two phenomena associated with the self-focusing effect of SPs as further evidences. To the best of the authors' knowledge, this is the first time to clarify this issue and give an explicit proof on this issue.

2. Fundamentals of SPs Excitation

Fig. 2 shows the two fundamental conditions in excitation of SPs.

2.1 Dispersion Relation

The dispersion relation requires the matching of wave vectors of incident beam and SPs [3]. As shown in Fig. 2, the direction of the wave vector k_{sp} is consistent with the propagation direction of SPs, locating at the interface between the metal (gold) film and the dielectric (sample). And the value of k_{sp} is expressed as:

$$k_{sp} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} \tag{1}$$

where ω is the plasmon frequency, c is the speed of light in vacuum, ε_1 and ε_2 are the dielectric functions of the metal (gold) and the dielectric (sample) respectively. As for the wave vector of incident beam, only the component along the interface is capable to satisfy the dispersion relation. The wave vector component of incident beam along the interface k_r is expressed as:

$$k_r = k\sin\theta = 2\pi n \sin\theta / \lambda \tag{2}$$

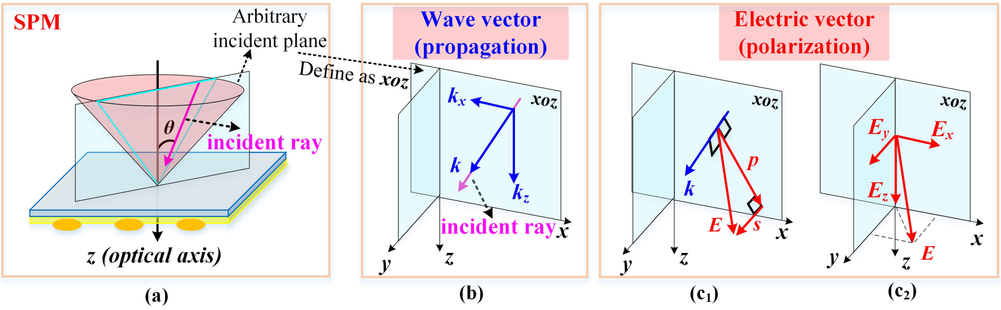


Fig. 3. Definition and decomposition of vectors in the beam cone. (a) Select an arbitrary incident ray in an arbitrary incident plane. The selected plane is defined as the *xoz*-plane. (b) Decompose the wave vector *k* into k_x and k_z. (c) Decomposition of the electric vector *E*. (c_1) Decompose the electric vector *E* into p-polarization and s-polarization. Note that *E* is orthogonal to *k* and is not in the *xoz*-plane. (c_2) Decompose the electric vector *E* into *Ex*, *Ey*, and *Ez*.

where *k* is the wave vector of incident beam, θ is the incident angle, *n* is the refractive index of the substrate, and λ is the wave length of incident beam. And the dispersion relation is given by:

$$k_{sp} = k_r \tag{3}$$

Note that both the definitions of k_{sp} in Eq. (1) and k_r in Eq. (2) are independent with the polarization components. And the independence makes the dispersion relation insufficient to directly clarify which of *Ex*, *Ey*, and *Ez* excites SPs.

2.2 TM Mode (P-Polarization) Excites SPs

The second condition refers to the mode matching that only the TM mode (p-polarization) excites SPs [4], [5], [10]. This condition was generally demonstrated by the sharp dip and sudden phase transition around the optimal excitation angle θ_{sp} on the reflection and transmission coefficients when using p-polarization [4]. And the crescent on the reflected back focal plane of SPM illuminated by linearly polarized beam (Fig. 5 in Ref. [5]) further supported this condition experimentally. However, this condition could not guarantee which component of the p-polarization (*Ex*, *Ey* and *Ez*) accounts for the excitation of SPs, either [4], [6].

In summary, the two excitation conditions of SPs are widely adopted in related works. However, neither of the two conditions can prove that '*Ez* excites SPs' directly. In the following analysis, we prove that *Ez* excites SPs by taking the two conditions into account simultaneously.

3. Excited SPs in Objective-Coupled SPM

This section is to theoretically prove that only the *Ez* component excites SPs in objective-coupled SPM. We especially emphasize the importance of the direction matching between the wave vectors and the electric vectors in the following derivations. For clear clarification, we illustrate the definition and decomposition of involved vectors in Section 3.1, and elaborate the derivation process in Section 3.2.

3.1 Definition and Decomposition of Vectors in the Focused Beam Cone

In this work, we utilize the common decomposition and definition of wave/electric vectors in Richards-Wolf vector diffraction theory [11], [12]. Note that we only concern the involved terms and the readers who are interested in detailed definitions and decomposition process are suggested to refer to Refs. [4], [6], [11], [12]. The schematic diagram is shown in Fig. 3. Fig. 3(a) illustrates the geometry of the focused beam cone and the sensor-chip. Fig. 3(b) illustrates the decomposed

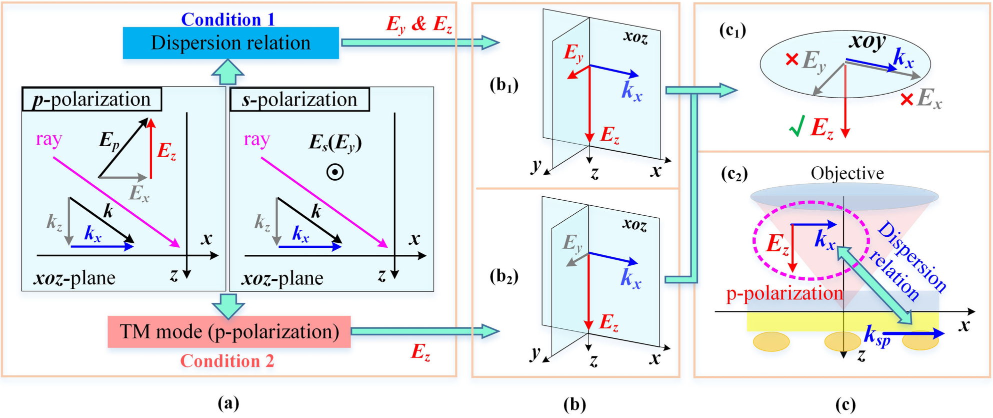


Fig. 4. Schematic diagram of proof procedure. (a) The decomposed vectors of the selected incident ray. (b) The two excitation conditions clarify that only *Ez* excites SPs. (b_1) Condition 1: k_x corresponds to *Ey* & *Ez* and excludes *Ex*. (b_2) Condition 2: *Ez* is p-polarization while *Ey* is s-polarization. (c_1) Only *Ez* excites SPs. (c_2) Agreement between the excitation conditions and the fact that *Ez* excites SPs.

components of the wave vector *k*. And Fig. 3(c) illustrates the decomposed components of the electric vector *E*.

Here we select an arbitrary incident plane as the example in Fig. 3(a). For convenient expression, we define the selected plane as the *xoz*-plane. An arbitrary incident ray is located within the plane. The wave vector of the selected incident ray is shown in Fig. 3(b). Since the direction of the wave vector is consistent with the propagation direction of the incident ray [13], the wave vector is within the *xoz*-plane. According to the definition of *xoz*-plane, the wave vector *k* contains two orthogonal components k_x and k_z.

The electric vector of the selected incident ray is shown in Fig. 3(c). The direction of the electric vector denotes the polarization direction of the incident ray [13]. In this work, we emphasize the direction of 'polarization' and 'electric vector' and the two terms are used interchangeably. According to the characteristics of transverse wave, the electric vector is orthogonal to the wave vector [13]. Fig. 3(c_1) shows that the electric vector has two components p-polarization and s-polarization. The two polarization components are within and orthogonal to the *xoz*-plane respectively. And the resultant electric vector *E* is thus out of the *xoz*-plane. In Fig. 3(c_2), the electric vector is further decomposed into *Ex*, *Ey*, and *Ez* components.

3.2 Theory of Ez Exciting SPs

This section theoretically proves that SPs are excited by the *Ez* polarization only. The schematic diagram is shown in Fig. 4. Fig. 4(a) shows the decomposed vectors of the selected incident ray. In Fig. 4(b), we utilize the two conditions of SPs excitation mentioned in Section 2 to clarify that only *Ez* excites SPs. And Fig. 4(c) shows the agreement between the excitation conditions and the fact that *Ez* excites SPs. The detailed derivation is illustrated as follows.

3.2.1 Condition 1: Dispersion Relation: In the arbitrarily selected *xoz*-plane, the dispersion relation is expressed as $k_x = k_{sp}$ [14]. It means that the excitation of SPs is functioned by the k_x component, not the k_z component of the incident ray. Here we prove that only *Ey* and *Ez* components are capable to satisfy the dispersion relation while the *Ex* component has no contribution.

We first give an intuitive derivation by the geometry relation shown in Fig. 4(a). The main idea is to connect the wave vector k_x to polarization components *Ex*, *Ey*, and *Ez* according to the orthogonality between propagation and polarization. The result is shown in Fig. 4(c_1). The wave vector k_x is perpendicular to the electric vector of either *Ey* or *Ez* while parallel to the electric vector *Ex*. If k_x corresponds to *Ex*, there would be electric field along the propagation direction k_x, which is contradictory with the transverse characteristic of the incident beam. In sub-conclusion, only the *Ey* or *Ez* component is capable to satisfy the dispersion relation of SPs, while the *Ex* polarization component does not account for the excitation of SPs.

To further emphasize the sub-conclusion, we give the transverse characteristics in the form of Eq. (4):

$$k_x E_x + k_y E_y + k_z E_z = 0 \tag{4}$$

The dispersion relation indicates that the excitation of SPs is functioned by the k_x component. Here we prove that the wave vector k_x coexists with *Ey* and *Ez* components while is independent with the *Ex* component. The analysis is divided into two steps.

Step 1): k_x coexists with the *Ey* and *Ez* components. Since the incident ray is a transverse wave, Eq. (4) is always true. When *Ey* and *Ez* components are zero, the wave vector k_x must be zero whatever the value of E_x is. It means that k_x always coexists with *Ey* and *Ez* components.

Step 2): k_x is independent with the E_x component.

1) When the electric vector *Ex* in Eq. (4) is zero, the value of wave vector k_x can be arbitrary. It indicates that the wave vector k_x is independent with the *Ex* component.
2) When *Ex* in Eq. (4) is non-zero, upon rearrangement of k_x, the wave vector k_x is described as:

$$k_x = -(k_y E_y + k_z E_z)/E_x \tag{5}$$

Combine Eq. (5) with the dispersion relation:

$$k_{sp} = k_x = -(k_y E_y + k_z E_z)/E_x \tag{6}$$

One may think that *Ex* also contributes to excitation of SPs. However, this interpretation is inappropriate. Actually, Eq. (5) can be further simplified into a function of the resultant wave vector *k*. When considering the case of the selected incident ray above, Eq. (6) can finally be expressed as:

$$k_{sp} = k_x = -k \sin\theta \tag{7}$$

The minus denotes the direction of k_x. As a result, Eq. (6) is actually another expression of dispersion relation in Eq. (2). It does not mean that *Ex* is related with k_x and contributes to excitation of SPs. As shown in Fig. 4(c_1), k_x is independent with *Ex* due to the transverse characteristic of the incident beam.

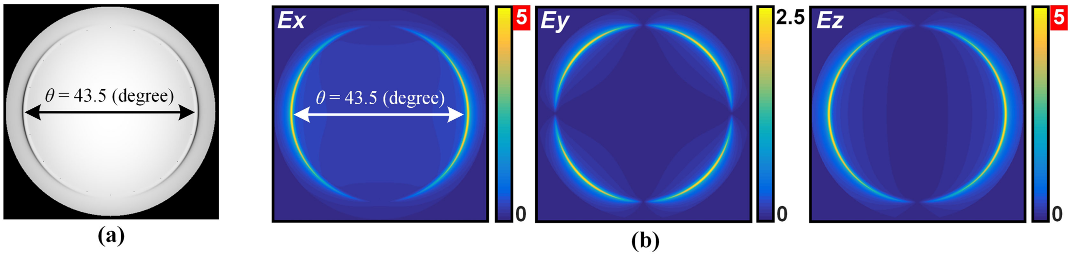
In conclusion, by combining the dispersion relation with the transverse characteristic, we prove that only *Ey* and *Ez* components are capable to satisfy the dispersion relation while the *Ex* component has no contribution.

3.2.2 Condition 2: TM Mode (P-Polarization) Excites SPs: Here we further clarify that only *Ez* excites SPs. As shown in Fig. 4(b_2), the *Ez* component is in the *xoz*-plane and corresponds to p-polarization, while the *Ey* component is perpendicular to the *xoz*-plane and refers to s-polarization. According to the condition 2, only *Ez* (p-polarization) can excite SPs, while the polarization component of *Ey* shows no contribution.

In conclusion, the condition 1 and 2 actually indicates that the polarization components locating in the *xoy*-plane (*Ex* and *Ey*) do not contribute to excitation of SPs. Thus, only Ez polarization which is perpendicular to the *xoy*-plane excites SPs as shown in Fig. 4(c_1). And the fact that *Ez* excites SPs is in good agreement with the excitation conditions. In Fig. 4(c_2), the matching of wave vectors k_x and k_{sp} corresponds to the dispersion relation (in blue). And the *Ez* polarization component corresponds to the p-polarization (in red).

TABLE 1
Parameters of Calculation

Light source	*NA* of objective	RI of immersion oil	RI of Au film	Excitation angle of SPs
633nm Uniform linear polarization	1.25	1.52	0.17+3.52i	43.5 deg



(a) (b)

Fig. 5. Back focal plane (BFP) and polarization components. (a) The BFP corresponding to the excitation angle of 43.5 degrees. (b) Distributions of *Ex*, *Ey*, and *Ez* on the BFP. The distribution of *Ex* and *Ez* are similar. The amplitude of *Ex* and *Ez* are comparable and the ratio is nearly 1:1.

4. Two Direct Phenomena of '*Ez* Dominates the Characteristic of SPM'

In this section, we give two direct and characteristic phenomena to further clarify that only the *Ez* component excites SPs: the self-focusing effect and lateral resolution of SPM. Before the discussion of the two phenomena, we quantify the ratio among the components of *Ex*, *Ey* and *Ez*. The distributions of *Ex*, *Ey*, and *Ez* on the back focal plane of the objective are calculated by [4]:

$$\begin{aligned} E_x(\theta,\varphi) &= \cos^{1/2}\theta \cdot E(\theta,\varphi)\left(\cos^2\varphi\cos\theta \cdot t_p(\theta) + \sin^2\varphi \cdot t_s(\theta)\right) \\ E_y(\theta,\varphi) &= \cos^{1/2}\theta \cdot E(\theta,\varphi)\cos\varphi\sin\varphi\left(\cos\theta \cdot t_p(\theta) - t_s(\theta)\right) \\ E_z(\theta,\varphi) &= \cos^{1/2}\theta \cdot E(\theta,\varphi)\cos\varphi\sin\theta \cdot t_p(\theta) \end{aligned} \tag{8}$$

where θ is the incident angle, φ is the azimuthal angle, and $E(\theta, \varphi)$ is the field distribution of the light source. The maximum of θ is determined by the numerical aperture (*NA*) of the objective and the refractive index of the immersion oil. For a uniform light source, $E(\theta, \varphi)$ is set to unity. The term $\cos^{1/2}\theta$ is incorporated to account for the effect of high numerical aperture. The term $t_i(\theta)$ is the multiple transmission coefficient of p- or s-polarization.

Here we take the excitation of SPs in air as the example to illustrate the ratio among the components of *Ex*, *Ey* and Ez. The related parameters such as wavelength, *NA*, refractive indices (RI), and excitation angle of SPs are listed in Table 1. The plasmonic sample is bare 46nm Au film exposed to air. Fig. 5(a) shows the corresponding BFP with excitation angle of 43.5 degrees. Fig. 5(b) shows that the ratio between *Ex* and *Ez* is nearly 1:1, which means that *Ez* is comparable with *Ex* and *Ey* in this case. Section 4.1 and 4.2 are both based on this condition.

4.1 Ez Accounts for Self-Focusing Effect in SPM

Self-focusing effect is one of the most distinctive characteristics of SPs. This effect refers to that SPs keep focused when the sample of SPM is moved closer to the objective and the strongest amplitude of focused SPs occurs at a plane with a certain defocusing distance instead of the focal plane of conventional OM [4], [15]. Previous publications have taken this effect into consideration in the practical implementation of SPM [16], [17]. However, the phenomenon that only *Ez* accounts

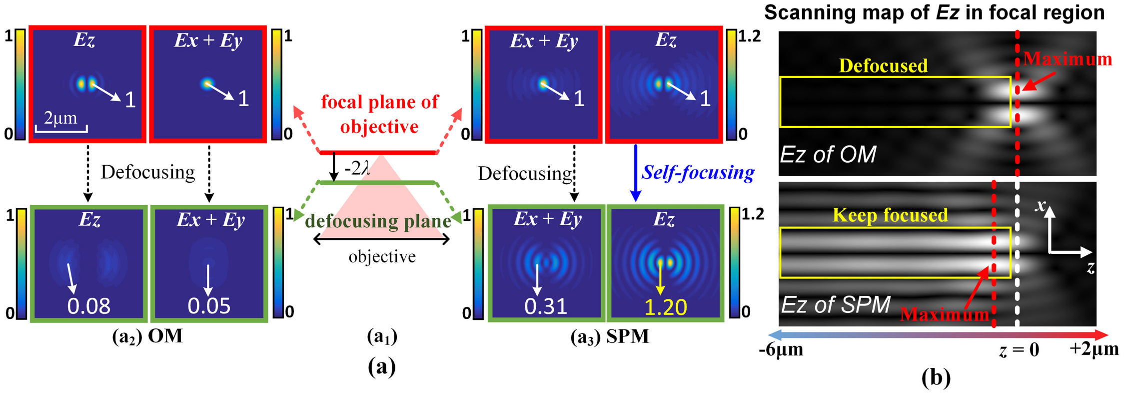


Fig. 6. *Ez* accounts for the self-focusing effect of SPs. (a) Intensity variations of *Ex*+*Ey* and Ez with defocusing. (a_1) The focal plane and -2λ defocusing plane. (a_2) The conventional OM shows divergences of both *Ex*+*Ey* and *Ez*. (a_3) The SPM shows only the divergence of *Ex*+*Ey* but gives a tighter *Ez*. (b) Focal field distribution of *Ez* on *xoz* plane in OM and SPM.

for the self-focusing effect has not been clarified. To elaborate this issue, we calculate the focal field distribution of *Ex, Ey*, and *Ez* with defocusing (the sample is moved away from the focal plane of the objective) in conventional OM and SPM according to the following expression:

$$e_i(x, y, z) = \int_{\theta_1}^{\theta_2}\int_{0}^{2\pi} E_i(\theta, \varphi)\cdot \exp[jk(x\cos\theta\cos\varphi + y\cos\theta\sin\varphi + z\sin\theta)]\sin\theta\, d\theta\, d\varphi \tag{9}$$

where e_i and E_i denote the amplitudes of the polarization components on the focal plane and BFP respectively; *x*, *y*, *z* are coordinates in the focal region; *j* is the imaginary unit; *k* is the wave vector in the immersion medium of the objective lens. The calculation results are shown in Fig. 6.

Fig. 6(a_1) shows the focal plane and a negative defocusing plane of 2λ. Fig. 6(a_2) shows the distributions of *Ex*+*Ey* and *Ez* on the focal plane (red square) and defocusing plane (blue square) in OM. And Fig. 6(a_3) shows the case of SPM. One can find that when the sample is defocused, the conventional OM shows divergences of both *Ex*+*Ey* and *Ez,* while the SPM shows only the divergence of *Ex*+Ey but gives a tighter *Ez*. For clear clarification, we also show the distribution of *Ez* of OM and SPM on *xoz* plane (Fig. 6(b)). It clearly demonstrates that: 1) The *Ez* of OM is diverged when the sample moves away from the focal plane ($z = 0$); 2) The *Ez* of SPM keeps focused even at a large defocusing distance; 3) The maximum of *Ez* of SPM occurs at a certain defocusing plane. The above discussions show that only *Ez* accounts for the self-focusing effect of SPs.

4.2 Ez Dominates the Lateral Resolution of SPM

The lateral resolution depends on the minimal spot size in the focal region. Since the focusing process of SPM is affected significantly by SPs [4], [6], [15], [18], the lateral resolution of SPM depends a lot on the *Ez* polarization which excites SPs in SPM. Ref. [8] approximates the focal spot of SPM and evaluates the lateral resolution by the field distribution of *Ez* in SPM directly. This operation is consistent with the fact that SPs are excited by *Ez* and helps to understand the real theoretical resolution of SPM. For clear illustration, Fig. 7 gives a comparison of resolution of SPM and OM under linear and radial polarization illuminations. Both the focal spots of OM and SPM are calculated by the resultant of *Ex*, *Ey*, and *Ez*. The minimal focal spot of SPM is obtained on the defocusing plane defined in Fig. 6(a_1) while that of OM is obtained on the focal plane of the objective. One can observe that the focal spot of SPM shows similar profile and size with the field distribution of *Ez* in either linear or radial polarization. The results are consistent with the

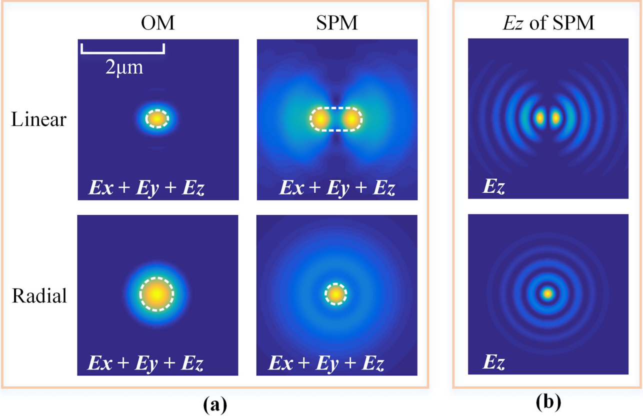


Fig. 7. SPM v.s. OM in lateral resolution. *Ez* dominates the resolution of SPM. (a) focal spots of OM and SPM under linear and radial polarizations. (b) *Ez* of SPM under linear and radial polarizations.

experiment in Ref. [19] that the resolution of SPM is worse than OM when using a linearly polarized illumination and the analysis in Ref. [8] that the lateral resolution of SPM exceeds the diffraction limit of OM when using radial polarization illumination. This effect shows the great significance of the *Ez* polarization component. In practical implementation of SPM, we suggest using illuminations whose *Ez* component generates a tightly focused spot.

5. Conclusion

This work theoretically proved that the polarization component *Ez* was the only excitation source of SPs. The key idea was to connect the excitation conditions of SPs to the polarization components of incident beam. To further verify the issue, we provided two direct and practical phenomena: i) *Ez* accounted for the self-focusing of SPM and ii) dominated the resolution of SPM. This work clarifies the misunderstanding about excitation of SPs and provides a fundamental for both academic research and practical application of SPM.

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