

Study of optical phase transduction on localized surface plasmon resonance for ultrasensitive detection

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Abstract: Localized surface plasmon resonance (LSPR) has shown its remarkable applications in biosensing, bioimaging, and nanophotonics. Unlike surface plasmon polariton (SPP), the current studies regarding LSPR as biosensor were restricted in probing the extinction spectra, and thus limit the performance in biosensing and bioimaging. Here, we reveal that optical phase of LSPR provides an acute change at resonance beyond extinction spectra, which permits an ultra-high sensitivity in phase interrogation. We found that optical phases of LSPR show two orders of magnitude higher sensing resolution than extinction spectra among the same nanostructures. For the first time, we demonstrated the feasibility of probing optical phase transduction in LSPR for biosensing, and the sensitivity is superior to not only the extinction spectra among the same metallic nanostructures, but also the LSPR sensors among the current literatures. In summary, the exploitation of LSPR by phase interrogation essentially complements the sensitivity insufficiency of LSPR, and provides new access to understanding and using the rich physics of LSPR.

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1. Introduction

Localized surface plasmon resonance (LSPR), a collective oscillation of electrons in metallic nanostructures, is induced directly by the incoming electromagnetic waves without any coupler for momentum conservation [1,2]. Owing to various merits such as coupler-free excitation for sensing, sensitive response to the local refractive indices, significant enhancement of the local electric fields, and the comparable size with biomolecules, LSPR technique brings a significant impact to the fields of biochemical sensing and optoelectronics that is incompetent by the state-of-the-art technique of surface plasmon polariton (SPP), and further leads to remarkable applications in molecular interaction [3–8], medical therapeutics [9,10], intracellular imaging [9], and nanophotonic devices [11,12]. Unlike SPP as a well-developed biosensing platform, yet, the current researches regarding LSPR for biosensing are restricted on characterizing its extinction cross section as a function of excitation wavelengths (i.e., extinction spectra) only, and thus radically limit its sensitivity and further applications to compete with other sophisticated biochemical-sensors [13,14]. As a consequence, a variety of arrangements were demonstrated recently to promote the sensitivity on extinction spectra by longer resonant wavelengths, specific operating configuration, or well-designed nanostructures [15–21]. Rather than the extinction spectra, in this work we reveal that optical phase of LSPR provides an acute change at resonance, permitting an ultra-high sensitivity beyond extinction spectra according to our algorithm and common-path optical phase system. For the first time, we demonstrated the feasibility of probing optical phase transduction in LSPR for biosensing, and the sensitivity is superior to not only the extinction spectra among the same metallic nanostructures, but also the LSPR sensors among the current reported studies.

2. Materials and methods

2.1 Common-path phase interrogation system for LSPR measurement

A home-made common-path phase interrogation system was served for LSPR measurement under total internal reflection (TIR) and coupler-free configurations. In the TIR configuration (Fig. 1(a)), a 3 mW laser diode with 633 nm in wavelength was used as the light source, and a linear polarizer in front of the laser to improve the extinction ratio of polarization state up to $10^5:1$. A half-wave plate was used to obtain equal components of s- and p-polarized waves corresponding to the glass substrate for loading nanostructures. The SF-11 glass with refractive index of 1.78 is used for prism and the substrate. Rather than extract optical phases by using such common interferometry as heterodyne approaches [22], we designed a common-path system and an algorithm to extract optical phases in an uncomplicated and highly sensitive way. After LSPR was excited through evanescent waves, the reflectance passed through a quarter-wave plate and a rotating linear polarizer for extraction of optical phase information: When passing through the quarter-wave plate, the traveling s- and p-polarized waves were changed into left- and right-circular waves respectively by introducing

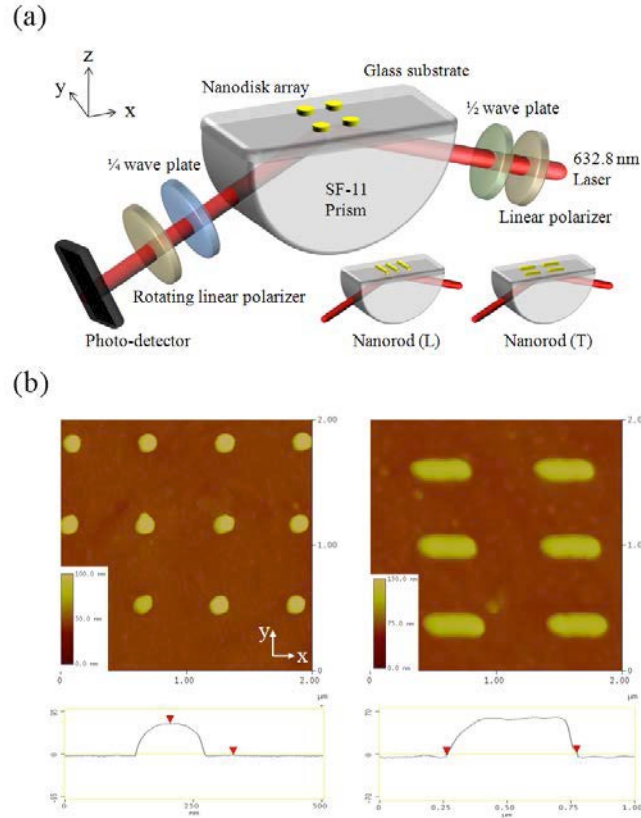


Fig. 1. LSPR probed by phase interrogation under TIR configuration. (a) Phase interrogation was first operated under TIR configuration. Three kinds of nanostructure arrays served LSPR sensing: nanodisk, nanorod (L), and nanorod (T). For nanorod (L), the electric field of s-polarized wave was parallel to the long axis. For nanorod (T), the electric field of s-polarized wave was parallel to the short axis. (b) AFM image of nanodisk and nanorod arrays prepared by electron beam lithography.

a $\pi/2$ phase retardation. The rotating linearly polarizer was then used to selectively record optical intensity in different rotation angles. The electric field of the s- and p-polarized waves after the quarter-wave plate and rotating linear polarizer can be described in Eq. (1) and Eq. (2) respectively,

$$\vec{E}_s = \frac{A_s}{\sqrt{2}} \begin{pmatrix} 1 & 0 \\ 0 & e^{i\pi/2} \end{pmatrix} \begin{pmatrix} 1 \\ 1 \end{pmatrix} = \frac{A_s}{\sqrt{2}} \begin{pmatrix} 1 \\ i \end{pmatrix}, \text{ where } A_s = |A_s| e^{i\phi_s} \text{ S wave after } 1/4 \text{ wave plate} \quad (1a)$$

$$\vec{E}_p = \frac{A_p}{\sqrt{2}} \begin{pmatrix} 1 & 0 \\ 0 & e^{i\pi/2} \end{pmatrix} \begin{pmatrix} 1 \\ -1 \end{pmatrix} = \frac{A_p}{\sqrt{2}} \begin{pmatrix} 1 \\ -i \end{pmatrix}, \text{ where } A_p = |A_p| e^{i\phi_p} \text{ P wave after } 1/4 \text{ wave plate} \quad (1b)$$

$$\vec{E}_s' = \begin{pmatrix} \cos^2 \theta & \sin \theta \cos \theta \\ \sin \theta \cos \theta & \sin^2 \theta \end{pmatrix} \vec{E}_s = \frac{A_s}{\sqrt{2}} e^{i\theta} \begin{pmatrix} \cos \theta \\ \sin \theta \end{pmatrix} \text{ S wave after linear polarizer} \quad (2a)$$

$$\vec{E}_p' = \begin{pmatrix} \cos^2 \theta & \sin \theta \cos \theta \\ \sin \theta \cos \theta & \sin^2 \theta \end{pmatrix} \vec{E}_p = \frac{A_p}{\sqrt{2}} e^{-i\theta} \begin{pmatrix} \cos \theta \\ \sin \theta \end{pmatrix} \text{ P wave after linear polarizer} \quad (2b)$$

where AS and AP are the amplitudes of the s- and p-polarized waves, ϕ_s and ϕ_p are the optical phases of the s- and p-polarized waves, and θ is the rotation angle of linear polarizer corresponding to the fast axis of quarter wave plate. The optical intensity of reflectance $I(\theta)$ encounters the photo-detector can be described as Eq. (3),

$$I(\theta) = \frac{1}{2} \left\{ |A_s|^2 + |A_p|^2 + 2|A_s||A_p| \left[\cos(\phi_{p-s}) \cos 2\theta + \sin(\phi_{p-s}) \sin 2\theta \right] \right\} \quad (3)$$

where ϕ_{p-s} is the difference between ϕ_s and ϕ_p . After we separately recorded the intensity in $\theta = 0, \pi/4, \pi/2$, and $3\pi/4$, the ϕ_{p-s} can be extracted through Eq. (4),

$$\tan(\phi_{p-s}) = \frac{I(\pi/4) - I(3\pi/4)}{I(0) - I(\pi/2)} = \frac{\sin(\phi_{p-s})}{\cos(\phi_{p-s})} \quad (4)$$

Once the refractive index of the analyte is changed, any shift in ϕ_{p-s} , would emerge through the excitation of LSPR. The system resolution of our common-path phase interrogation is 2×10^{-5} rad, which is comparable with the literature [22–24]. One thing to be noticed, the measured ϕ_{p-s} is independent of the intensity of s- and p-polarized waves (i.e., $|A_s|$ and $|A_p|$) based on Eq. (1) to Eq. (3) as long as the signal is well distinguished from noise; in other words, the intensity ratio between s- and p-polarized waves is not responsible for the detecting sensitivity, but their relative phase shift subject to environment change is. Since ϕ_{p-s} represents the optical phase difference between s- (ϕ_s) and p-polarized (ϕ_p) waves, the shift of ϕ_s and ϕ_p after introducing the sucrose solutions will determine the shift of ϕ_{p-s} . If the ϕ_s and ϕ_p shift simultaneously and equally, the shift of ϕ_{p-s} will be compensated, and stay invariant. If only ϕ_s (ϕ_p) shifts with ϕ_p (ϕ_s) staying invariant, the shift of ϕ_{p-s} will be totally contributed from the shift of ϕ_s (ϕ_p). In the conventional SPP, ϕ_s always stays invariant because only p-polarized wave can excite SPP, therefore, it is safe to ignore the contribution from ϕ_s . However, LSPR is free from this restriction, and thus the phase shifts from both s- and p-polarized wave have to be considered simultaneously. The detail of the common-path phase system and the algorithm is illustrated elsewhere [25].

Three kinds of Au nanostructural configurations were prepared for experiments, denoted as nanodisk, nanorod (L) with its long axis parallel to the electric field of s-polarized wave, and nanorod (T) with its short axis parallel to the electric field of s-polarized wave (Fig. 1(a)). Nanodisk and nanorod arrays were fabricated by electron beam lithography. Electron beam evaporator was used to deposit Au with deposition rate 0.5 Å/s. The area of as-prepared nanostructure array is $0.9 \times 0.9 \text{ mm}^2$, which is much larger than the laser beam size. Atomic force microscopy was used to measure the topography of nanostructure. From Fig. 1(b), the diameter of nanodisk is ranged from 140 nm to 164 nm and the periodicity is ~650 nm. Thickness of nanodisk array was measured as 70 nm. The length of nanorod is 512 nm and the width is 215 nm, of which the aspect ratio is around 2.5. The periodicity of nanorod array is 1002 nm in x direction, and 629 nm in y direction. Thickness of nanorod array was measured as 61 nm. We fabricated two nanorod arrays with the long axis parallel to and perpendicular to the electric field of s-polarized wave under common-path TIR configuration.

2.2 Sensitivity of Au nanostructural arrays measured by extinction spectra

The resonant wavelengths of these Au nanostructural configurations were first examined by extinction spectra through normal incidence in order to have comparison with phase interrogation. Extinction spectra of nanostructure arrays were measured by Fourier transform infrared spectrometer (FTIR, Hyperion 1000, Bruker Optics). From the extinction spectra (Figs. 2(a)-(c)), we can see that the nanodisk owns only one resonant mode which occurs at

632.8 nm. On the other side, the nanorod showed two resonant modes as the electric field of incidence perpendicular to the short axis (i.e. transverse mode) and to the long axis (i.e. longitudinal mode). Resonant wavelength of the transverse mode is in 577.2 nm and that of the longitudinal mode is in 794.1 nm. Resonant modes of nanodisk and nanorod were simulated by boundary element method (BEM) [26,27], and the charge density distribution of nanostructures' eigen-modes were constructed (Fig. 2(d)). Au dielectric constant is polynomial fitted from the literature [28], and wavelength lower than the database was estimated by Drude model with plasma frequency 2.15×10^{15} Hz [29]. From the comparison between extinction spectra and simulation, the resonant modes in extinction spectra were confirmed as dipolar modes. In addition, for nanodisk, there exists the second dipolar mode along z direction besides the dipolar mode along the in-plane direction, which the resonant wavelength of this second mode was simulated as 205 nm. For nanorod, there also exists a third dipolar mode besides the transverse and longitudinal modes, which the resonant wavelength of the third mode was simulated as 184 nm. The simulation suggested that the resonant wavelengths along z direction in the nanodisk and the nanorod were shorter than 205 nm, which were not coupled by the probing wavelength used in our experiment. Accordingly, we could only consider the resonant modes along inplane direction in the following experiments.

Sensitivities of LSPR probed by extinction spectra were measured through the shift of extinction peak in wavelength. We introduced glucose solution with 2 M concentration, and the extinction peaks of nanostructures shifted to longer wavelength. Refractive index of glucose solution was estimated through the equation $n = 1.325 + 1.515 \times 10^{-4} \times C$, where n represents the refractive index of glucose solutions and C is the concentration in grams per liter [30]. The molecular weight of glucose is 180.16 g/Mol. From the refractive indices

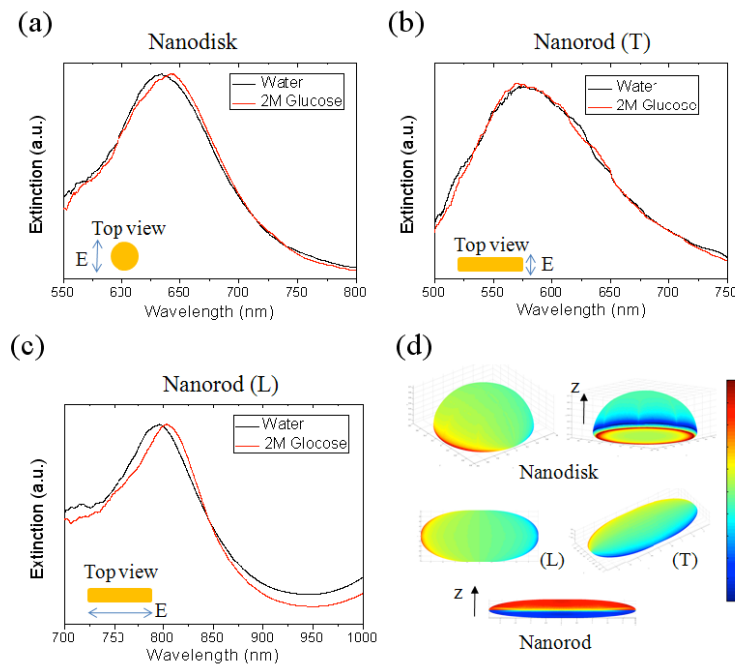


Fig. 2. Extinction spectra of nanostructures. (a) Nanodisk owns one dipolar resonant wavelength which is 632.8 nm. (b), (c) Nanorod owns two dipolar resonant wavelength which transverse mode occurs at 577.2 nm (nanorod (T)), and longitudinal mode occurs at 794.1 nm (nanorod (L)). 2 M glucose solution was introduced in order to measure the sensitivity, and both nanodisk and nanorod showed the red shift on resonant wavelengths. (d) Charge density distribution of nanostructures' eigen-modes.

difference and the corresponding wavelength shifts, sensitivity of the nanodisk is 172.34 nm/RIU, sensitivity of the nanorod in the transverse mode is 60.27 nm/RIU, and that in the longitudinal mode is 194.18 nm/RIU. The sensitivities of as-prepared Au nanostructural configurations are similar to the literature [20]. By considering the system resolution of FTIR in used as 0.03 nm, the sensing resolution of nanostructures in wavelength interrogation became 1.74×10^{-4} , 4.98×10^{-4} , and 1.54×10^{-4} RIU for nanodisk, nanorod in transverse mode, and longitudinal mode, respectively.

3. Results and discussion

3.1 Near-field excitation of LSPR in phase interrogation

The optical phases of LSPR were first probed by evanescent waves under total internal reflection (TIR) configuration because LSPR responds a greater cross section of scattering and extinction to the p-polarized evanescent waves [31]. The ultrasensitive LSPR probed by phase interrogation was demonstrated by measuring the analyte of sucrose solutions, and were operated under four different incident angles greater than the critical angle for the purpose of examining the sensitivity, and clarifying the sensing mechanism of LSPR by phase interrogation. From Fig. 3(a), nanorod (L) under a 50° incidence exhibited the highest sensitivity (10.95 rad/RIU) among the three nanostructural configurations, and the sensing resolution reached 1.83×10^{-6} RIU by taking the consideration of our system resolution. This sensing resolution is 10.93 times higher than the most sensitive LSPR sensors in current literature [17–20], where the highest sensitivity does not exceed 2×10^{-5} RIU by taking the consideration of the general system resolution to be 0.02 nm [19,20]. We further compared the sensing resolution between phase interrogation and extinction spectra on the same nanostructural configurations, and the results showed that the sensing resolution of phase interrogation was 35.29 to 84.15 times higher than extinction spectra on these three nanostructural configurations (Figs. 2(a)-(c)).

As linearly fitting the sensing curves of the nanodisk and the nanorods (T & L) under four detecting angles (i.e., 50°, 55°, 60°, 65°) in Fig. 3(a), the values of the adjusted R square range from 0.838 to 0.876 for the nanodisk, from 0.906 to 0.962 for the nanorod (T), and from

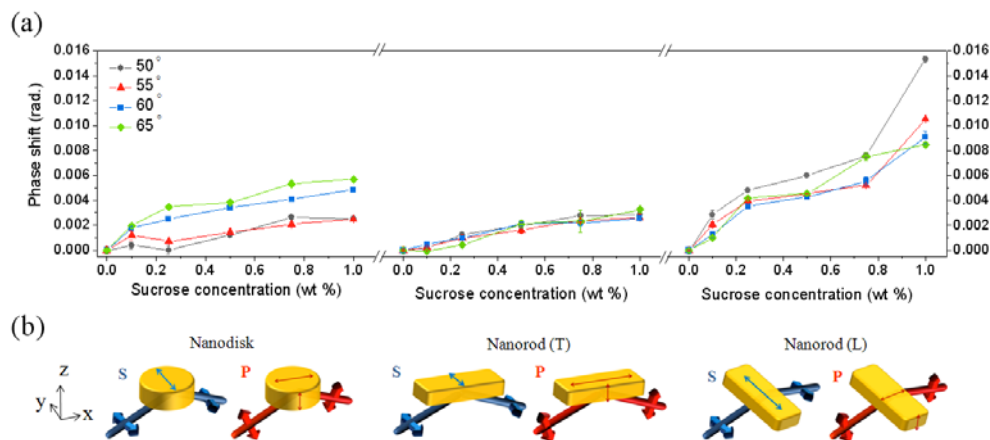


Fig. 3. Sensitivity measurements of LSPR by phase interrogation under TIR configuration. (a) Sucrose solutions with different concentrations were introduced gradually, and led to the phase shift of nanostructures. The highest sensitivity in the nanodisk was 4.06 rad/RIU under a 65° incidence. The highest sensitivity in nanorod (L) was 10.95 rad/RIU and in nanorod (T) was 2.34 rad/RIU under a 50° incidence. Five times of measurements were conducted in each nanostructural configurations in order to confirm the experiments. (b) Incidence can be decomposed into s- and p-polarized evanescent waves. Sensitivity differences among the three nanostructural configurations were due to the correlation of the resonance modes between s- and p-polarized evanescent waves.

0.853 to 0.939 for the nanorod (L), respectively. All the values of the adjusted R square in these sensing curves indicate that the phase shift of our LSPR linearly depends on the sucrose concentration gradient. Besides, the value of the adjusted R square for SPP is 0.898 as shown in Fig. 4(b), which is comparable with the LSPR sensing curve and further suggests that the fluctuation in the sensing curve stems from the instable factor of the detecting environment, including the noise of electric circuits and the fluctuation of flow rates. In addition to the detecting environment, the imperfection of the fabricated nanoparticles may also contribute to a flattened or fluctuated phase transition due to the electron scattering and electron-phonon scattering. Therefore, once the detection frees from the instable factor of detection environment and the imperfection of nanoparticle fabrication, the sensing curve should perform accurately with the less fluctuation. Here we have revealed a drastic promotion in sensing resolution merely by changing the probing method from extinction spectra to phase interrogation.

3.2 Sensing mechanism of LSPR in phase interrogation

In fact, it is possible to undertake such ultrasensitive LSPR probed by phase interrogation by harnessing three enabling factors. First, the optical phase transition occurs only across the resonant wavelength, meaning that probing source right at the resonant wavelength will generate the highest sensitivity. Next, it is essential to excite LSPR by the p-polarized evanescent wave because it provides a much greater cross section of extinction and scattering than s-polarized evanescent wave or plane wave [31], leading to a greater phase shift and certainly higher sensitivity. Further theoretical verifications are conducted and will be published in the future. Finally, the sensitivity in our phase interrogation rests on the shift of ϕ_{p-s} ; thus, the case with one polarized wave invariant in the optical phase and the other polarized wave completely on resonance yields the highest sensitivity.

For example, the nanorod (L) array resonated strongly with p-polarized evanescent wave along the short axis of nanorod (Fig. 3(b)), whereas the resonance rarely occurred with s-polarized evanescent wave along the long axis of nanorod because the probing wavelength was far from the longitudinal mode. Nanorod (L) array satisfied the three aforementioned factors concurrently, and led the greatest phase shift to the best correlated sensitivity of LSPR among three nanostructural configurations. In contrast, the nanorod (T) array resonated mainly with s-polarized evanescent wave along the short axis of nanorod and no resonance with p-polarized evanescent wave (Fig. 3(b)); consequently, the sensitivity decreased significantly and was independent of the incident angle. For the nanodisk array, it resonated with s- and p-polarized evanescent waves simultaneously due to the only resonant mode at xy plane (Fig. 3(b)). The simultaneous excitation of s- and p-polarized evanescent waves compensated for the shift of ϕ_{p-s} in different amounts along the incident angle, therefore, the sensitivity was lower than the nanorod (L) array but higher than the nanorod (T) array.

3.3 Optical phase vs. incident angle plots of LSPR, SPP, and TIR

To further confirm that the phase shift in nanostructures derive from LSPR rather than SPP or TIR, we compared the phase curves vs. the incident angles and the sensitivity measurements between a continuous Au film with 50 nm thickness, a blank substrate without a nanostructure array, and the nanorod (L) array. As shown in Fig. 4(a), the Au film exhibited a regular SPR phase curve, in which a drastic optical phase transition occurred at 54.5°. The optical phase transition angle coincided with the resonant dip when we measured by angular interrogation. The phase curve obtained from the blank substrate was slightly varied along with incidences, since the optical phases of the s- and p-polarized waves were not identical under total reflection. The phase curve in nanorod (L) exhibited similar behavior with the blank substrate rather than the Au film substrate, and this similarity reflected the discrete arrangement of nanostructures on a glass substrate. The optical phases carried by reflectance are the ensemble of LSPR excitation and TIR in nanorod (L), and the deviation between nanorod (L) and the blank substrate is modulated by the coupling of LSPR. This result suggests that though TIR

was applied in experiments, SPR was not excited in nanostructures. From the sensitivity measurements (Fig. 4 (b)), the Au film exhibited the highest sensitivity due to SPR excitation, and this ultra-high sensitivity of SPR in phase interrogation has been fully studied in the literature [22–24]. The blank substrate exhibited a negligible phase shift as the increasing sucrose concentrations were introduced. The slight phase shift was due to the change of TIR circumstances. Sensitivity in nanorod (L) was 8.64 times higher than in the blank substrate, and this amplified sensitivity indicates that the shift of ϕ_{p-s} stemmed mainly from LSPR excitation, not TIR.

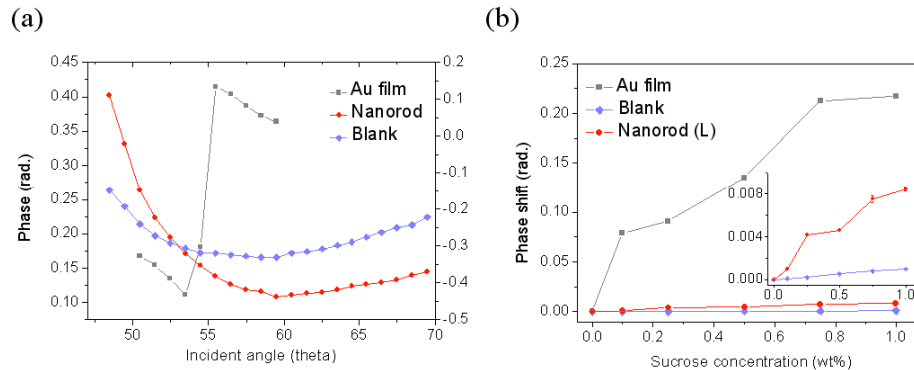


Fig. 4. Phase diagrams and sensitivity measurements of Au film (50 nm), blank substrate without a nanostructure array, and nanorod (L) array. (a) The phase diagram of Au film corresponded to a regular SPR phase diagram with the resonant angle of 54.5° (corresponding to the right axis). The optical phases changed along with the angle in the blank substrate were due to the optical phase difference between s- and p-polarized waves traveling under TIR (corresponding to the left axis). The phase curve in nanorod (L) is similar to the blank substrate (corresponding to the left axis), indicating that the optical phase change in nanorod (L) was induced by the ensemble of LSPR excitation and TIR, rather than SPR excitation. (b) The Au film exhibited a sensitivity of 155.32 rad/RIU, which is the regular sensitivity of SPR in phase interrogation. The sensitivity of the blank substrate was 0.7 rad/RIU under a 65° incidence, and the sensitivity of nanorod (L) was 6.05 rad/RIU under a 65° incidence.

3.4 Far-field excitation of LSPR in phase interrogation

Though we have successfully demonstrated the sensitivity improvement by phase interrogation under TIR configuration, it is more essential to validate this feasibility of phase interrogation under coupler-free configuration, which allows LSPR diverse biological applications *in vitro* or *in vivo* [4,32], and even biologically label-free imaging. Here we impinged the probing laser under a normal incidence from the glass substrate side, and adopted nanorod array for measurements because its anisotropic geometry helps avoid the phase shift compensation between the s- and p-polarized waves during detection (Fig. 5(a)). As shown in Fig. 5(b), the sensing resolution reached 1.94×10^{-5} RIU, which is 7.94 times higher than extinction spectra on the same nanostructure. Notice that the sensing resolution of coupler-free configuration is one order less than that operated by TIR, due to two major reasons. One is that coupler-free configuration is probed by a plane wave instead of an evanescent wave, which corresponds to a smaller cross section of scattering and extinction [31]; the other is that the transmittance in such coupler-free configuration suffers from substantial scattering attributable to the nanostructure array and dielectric layers. Nevertheless, phase interrogation under the coupler-free configuration still performed better sensing ability, almost one-order of magnitude greater than that obtained by measuring the conventional extinction spectra.

It is worth to mention that this sensitive phase transduction is mainly from the LSPR of the isolated nanostructure, but not from the periodic structure. Based on current

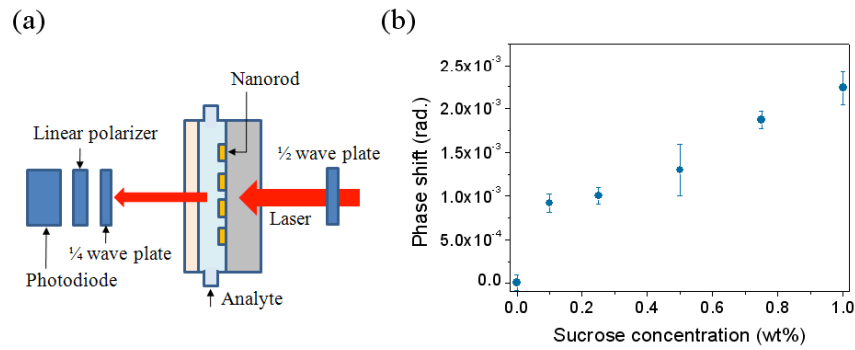


Fig. 5. LSPR probed by phase interrogation under coupler-free configuration. (a) Coupler-free configuration is operated by normal incidence from the glass side, and the transmittance is analyzed. Nanorod array was adopted for the measurement, and the included angle between the long axis of the nanorod and the electric field of incidence was 45° . (b) The sensitivity measurement shows that the sensitivity of LSPR under coupler-free configuration was 1.03 rad/RIU . The lower sensitivity than LSPR under the TIR configuration was obtained owing to the different excitation waves and probing methods: TIR configuration was excited by an evanescent wave and the reflectance was under analysis; coupler-free configuration was excited by a plane wave and the transmittance was under analysis.

understanding, localized surface plasmons coupling with a periodic structure can be realized from three mechanisms: One is the coupling through localized surface plasmonic field, by which the effective coupling is limited to the separation between isolated nanostructures in the value of around 100 nm in visible region [33,34]; second is the coupling through the aid of surface plasmon polaritons, by which the periodic pattern has to be built on a metallic continuum, usually made of Au or Ag thin film [35,36]; and third is the diffractive coupling, by which the diffraction order of the periodic pattern should meet the resonant wavelength of the single nanoparticle [37–40]. In our system, there is no metallic continuum and the separation between nano-structures is much larger than 100 nm . Therefore, the only possible route to have the coupling among plasmons is through diffractive coupling. Since the diffractive coupling between periodic pattern and single particle resonance may lead to the narrow resonance peak and enhanced electromagnetic field, it may potentially contribute to the sensitivity when phase interrogation is conducted. Since we measured the sensitivity of nanodisk and nanorod arrays under 0° , 50° , 55° , 60° , and 65° , the only possible condition to generate diffractive coupling was calculated as the nanodisk array under 60° or 65° incidences (63.396° , -3rd diffraction order, analyte mode in x-direction) based on the literature [40]. However, the nanodisk array in our work can be excited by s- and p-polarized waves simultaneously due to its isotropic structure, and thus both s- and p-polarization will lead to diffractive coupling if it is successfully generated [41–43]. The synchronous influence between s- and p-polarized waves will eventually compensate the phase shift of ϕ_{p-s} , and results in no effect in sensitivity. One thing to be noticed, the highest sensitivity, nanorod (L), is free from the diffractive coupling effect under the examined angles. As a consequence, the periodic pattern in our system should have no contribution on the sensitivity of detection.

4. Conclusion

In conclusion, we carried out the first-time demonstration of probing LSPR by phase interrogation for biosensing, and the results showed the superior sensitivity beyond not only the conventional extinction spectra among the same nanostructures, but also the LSPR sensors among the current reported studies. We examined the sensing mechanism by near-field and far-field excitations on three common nanostructural configurations in order to make a complete understanding of optical phase in LSPR as a biosensor. As excited by evanescent p-

polarized wave, sensing resolution reached 1.83×10^{-6} RIU, which is 84.15 times higher than values for probing by extinction spectra on the same nanostructures, and is 10.93 times higher than the most sensitive LSPR sensors in the current literature [17–20]. As excited by plane wave, sensing resolution reached 1.94×10^{-5} RIU, which is 7.94 times higher than the values for probing by extinction spectra. Sensitivity can be further maximized by satisfying three design rules: (1) probing wavelengths meet resonant wavelengths; (2) p-polarized evanescent wave provides much greater coupling cross sections than s-polarized evanescent wave and plane wave, and eventually contributed to the sensitivity enhancement; and (3) phase shift compensation between s- and p-polarized waves should be avoided. More important, by using our common-path optical phase system, it is available to probe LSPR under arbitrary angles by a single optical path, and this common-path design further improved the sensing resolution by filtering out unnecessary noises during the measurement; moreover, this design satisfies those bio-detecting circumstances that regularly probed by extinction spectra. In summary, LSPR owns two exceptional advantages that are incompetent by the state-of-the-art technique of SPP— coupler-free excitation for bio-sensing and even bio-imaging, and the comparable size with biomolecules. These two exceptional advantages allow an LSPR sensor to be applied as single particle detection for trace measurement [44], molecular ruler in studying the conformation change of biomolecules [45], ultra-sensitive nanosensors [46], intracellular imaging [9], and possibly better resolution of bio-imaging under the normal incidence. Here we enhanced its sensitivity by introducing the existence of phase transduction from LSPR. A common-path phase interrogation under near-field and far-field (i.e., coupler-free) excitation is utilized to demonstrated a comparable sensitivity with the conventional SPP. We believe that LSPR probed by phase interrogation will not just benefit the applications in biochemical sensing, but also create new access to understanding and using the rich physics of LSPR on metallic nanostructures.

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