Applied Physics Letters

Strong coupling in hybrid plasmon-modulated nanostructured cavities

Zhen-Yu Zhang, Hai-Yu Wang, Jiang-Lin Du, Xu-Lin Zhang, Ya-Wei Hao, Qi-Dai Chen, and Hong-Bo Sun

Citation: Applied Physics Letters **105**, 191117 (2014); doi: 10.1063/1.4901445
View online: http://dx.doi.org/10.1063/1.4901445
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/105/19?ver=pdfcov
Published by the AIP Publishing

APPLIED PHYSICS LETTERS **105**, 191117 (2014)

Strong coupling in hybrid plasmon-modulated nanostructured cavities

Zhen-Yu Zhang, Hai-Yu Wang,[a)] Jiang-Lin Du, Xu-Lin Zhang, Ya-Wei Hao, Qi-Dai Chen, and Hong-Bo Sun[a)]
State Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, 2699 Qianjin Street, Changchun 130012, China

(Received 5 September 2014; accepted 30 October 2014; published online 13 November 2014)

The understanding of strong coupling between local restricted electromagnetic field of cavity and surface-plasmon (SP) on the metal surface lays the groundwork for many photonic applications. However, much of the fundamental photophysical properties underlying this performance such as how this strong coupling is induced by these two components, namely, the SP modes and cavity modes have remained unknown. Here, we present a study of a hybrid plasmon-modulated coupled system with Rabi splitting energy at 336 meV, and the coupled hybrid states are highly sensitive to the prosperity of SP mode which is induced by morphology of inlaid grating surfaces. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4901445]

The excitation of collective electron oscillations on metal surface, in short "surface-plasmon" (SP) constitutes the basis for the emerging field of surface plasmonics.[1] In all coupling fields, the different components cannot be regarded as isolated elements, therefore, the coupling between SP and the light coupled to the plasmons sensitively determines the functionality of the applications. Recently, researchers have paid close attention to investigate one question: How can we manipulate the coupling strength in a hybrid system? Two fundamentally different coupling mechanisms have to be distinguished so far: coupling via near-field and coupling via far-field. Plasmonic nanostructures can be coupled via their near-field when the hot spots (two adjacent) space is pronounced lower than the plasmon resonance wavelength, and this effect has garnered much interest.[1] Such as the apertureless near-field optical microscopy of single molecules,[2] the Au SP interference patterns using Scanning Near Field Optical Microscope (SNOM) optical microscope system,[3] and later, the plasmon-enhanced Raman Scattering by carbon nanotubes optically coupled with near-field cavities[4] have been investigated. All the near-field coupled investigations have in common that the fundamental SP resonance is hybridized due to interactions. In general, this coupling strength decreases when the hot spots distance increases, and when the inter-nanostructure distance exceeds the near-field regime, the hybrid localized plasmon modes will disappear and electrodynamic effects such as damping can no longer be neglected. Thus, transform the coupling from near- to far-field.

In this letter, we propose a far-field/microcavity/nanocavity cascading energy coupling scheme, and study the strong coupling between SP modes, excited by a two-dimensional (2-D) metallic grating array, and Fabry–Perot (F–P) cavity modes formed within a planar-like microcavity in the visible spectral range. Under TM-polarized excitation (electric field parallel to the incident plane), we observed a large Rabi splitting which is absent from the case under TE-polarized excitation (electric field perpendicular to the incident plane). At this strong coupling regime, we demonstrate the efficiency of energy exchange and mode conversion under TM-polarized excitation is sensitively dependent on the depths of gratings, namely, the intensity of SP modes.[5–7] Cavity is an appealing tool to enhance light-matter interactions within the framework of cavity quantum electrodynamics (cQED). Very interesting effects occur when the cavities are doped with emitters, such as nanostructures,[8,9] J-aggregates,[10,11] gratings,[12,13] nanoannulus,[14–18] have been recently demonstrated. As a useful function of cavities, the ability of coupling two radiating system via the optical far-field over a larger distance is very attractive, moreover, cavities have microscale dimensions with mode volumes at the order of or larger than the wavelength of light, and on the other hand, can be truly nanoscale with mode dimensions way below the wavelength of the light.[1]

To investigate the strong coupling behaviour between the SP and F–P cavity modes, we fabricated a grating–cavity hybrid structure consisting of a two-dimensional silver grating array inlaid in microcavity by layer-by-layer fashion. Fig. 1 shows a schematic cross-sectional view of the hybrid plasmonic system. The samples were fabricated using physical vapor deposition for the mirrors, spin coating for spacer, and two beam interference for grating array. First, the

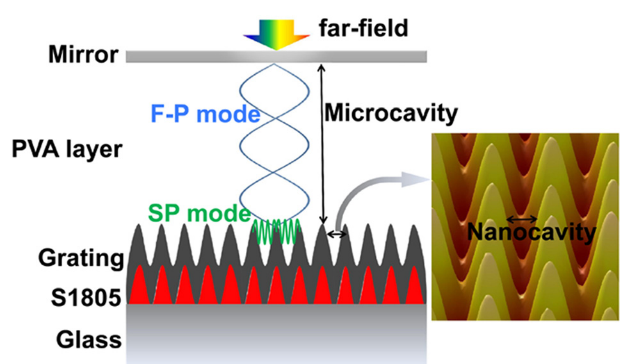

FIG. 1. Schematic cross-sectional view of the grating–cavity hybrid structure system.

[a)]Authors to whom correspondence should be addressed. Electronic addresses: haiyu_wang@jlu.edu.cn and hbsun@jlu.edu.cn.

corrugated gratings were fabricated on S1805 polymer resist coated on glass substrates by the two-beam interference method by employing 266 nm lasers. Then, a 50 nm thick sliver layer serving as the lower mirror is evaporated on corrugated gratings in high vacuum thermal evaporation. Subsequently, for the spacer we use the polyvinyl alcohol (PVA)-based spin-on in different mixing ratios with water, resulting in different space thicknesses. Finally, 30 nm of sliver is evaporated for upper mirror. The sliver gratings in the cavity acts as couplers to exciting SP along the interface, and the period of 300 nm and 325 nm are chosen to excite the first order SP mode (m = 1) that could coincide with the third F–P cavity mode in the visible spectral range. The strong coupling between the F–P mode and the SP mode is evident through a large mode splitting can be observed experimentally and a strong modal anti-crossing behavior in simulation. Angle-resolved refection detections under two different polarization states (TM and TE) were performed to mapping the dispersion relations of both the bare planar cavity and the hybrid grating–cavity systems by scanning the incident angle from 0° to 30°.

In analogy to a previous work,[12] in which the slow surface plasmon polaritons (SPPs) in plasmonic waveguiding bands formed by coupled plasmonic cavities is investigated, the parameter of nanostructure is critical due to it decisively determined the properties of plasmonics. Equally, the space of cavity has to be accurately controlled, due to the strong coupling can only be observed when the resonances of the photonic microcavity and the SP mode intersect. The atomic force microscopy (AFM) image of 300 nm period gratings are shown in the left panel of Fig. 2(a) and their profile are plotted in the right panel. Fig. 2(b) shows the experimental specular spectra for the doped microcavity containing corrugated gratings (black line) and the calculated specular refection spectra (red line) for the bare planar cavity under TM polarization. In Fig. 2(b), the coupling fashion is altered when the period changed, and the hybrid states are gradually distinguished when increasing of the grating depth from 20 nm to 80 nm for the two serial samples (300 nm and 325 nm). The intensity value of pronounced hybrid dips in reflectivity shows a trend of gradual increase with the increasing intensity value of SP modes which remain linearly proportional to the depths, and meanwhile, the intensity value of original co-resonant dips in reflectivity shows a trend of gradual decrease with the increasing coupled strength. The strong coupling of this SP mode with the F–P mode results in a large Rabi splitting of 336 meV in 300 nm period hybrid system, and 317 meV in 325 nm period hybrid system. The coupling leads to the lower polariton branch (LPB) and the upper polariton branch (UPB) with an energy separation so-called Rabi splitting, which is a direct measure of the splitting magnitude between the two components.

The coupling is evident revealed by an anti-crossing of the resonances when the SP mode resonance is detuned around the photonic microcavity resonances. The simulations were performed using scattered matrix method to account for this coupling in the hybrid nanostructures. The unperturbed resonant mode, pure SP mode, and the hybrid coupling modes excited by TM-polarized incident light are depicted in the dispersion diagrams of Figs. 3(a)–3(c). It is

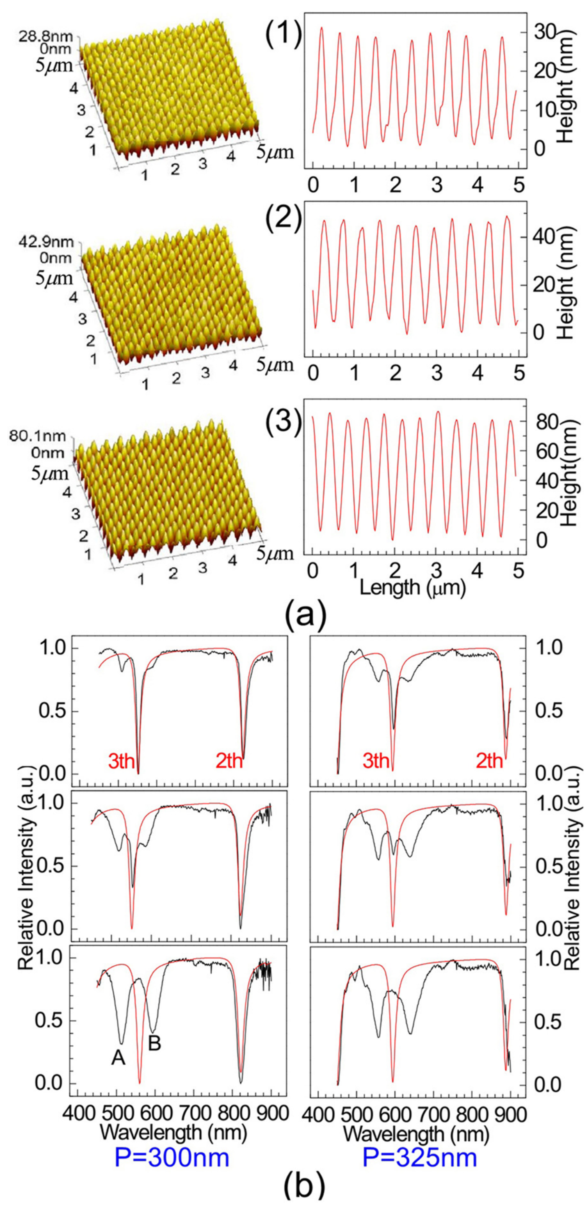

FIG. 2. Plots of strong coupling. (a) The AFM images of 300 nm period gratings, depths: (1) 22.8 nm, (2) 42.9 nm, and (3) 80.1 nm. (b) The calculated reflectance spectra of undoped cavity are plotted as red curves, and the experimental reflectance spectra of the doped structures are plotted as black curves, the period in left panel is 300 nm and the right panel is 325 nm.

clearly seen from Fig. 3(b) that the first SP mode linearly proportional to the periods is excited under TM-polarized incident light within the wavelength range covering the spectral window of the F–P cavity. Fig. 3(c) maps the calculated dispersion relations of the hybrid system under TM-polarized excitation geometry, and we can see from Fig. 3(c) that the strong coupling between the SP and F–P cavity modes occurs where the energy gap between the two branches reaches the minimum value showing a splitting energy. The dispersion relation of this coupled system indicate that, when the mode splitting occurs, the SP and F–P cavity modes should be coexisting in every hybrid state, and lead to the hybrid state are half-SP, half-cavity hybrid. We attribute the deviation of the hybrid energy position between simulation and experiments to the geometrical imperfect such as the outline of the corrugated gratings is not the strict lattice and the cavity is not the F–P cavity strictly.

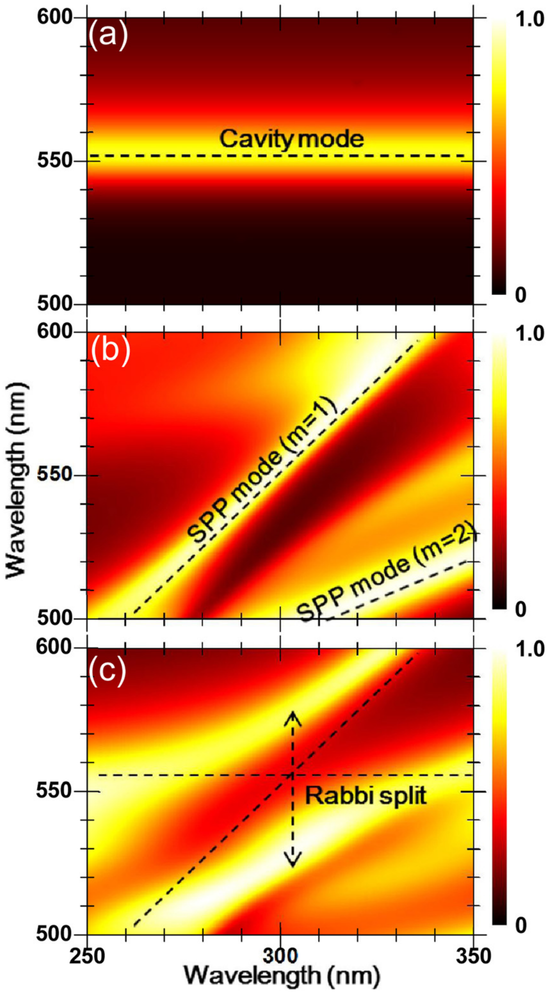

FIG. 3. Simulations: (a) Contour plots of the calculated reflectance of third unperturbed F–P mode, (b) contour plots of the calculated reflectance of sliver grating as a function of wavelength and period under TM-polarized light excitation, (c) contour plots of the calculated reflectance of the doped system consisting of a silver grating array as a function of wavelength and grating period under TM-polarized light excitation.

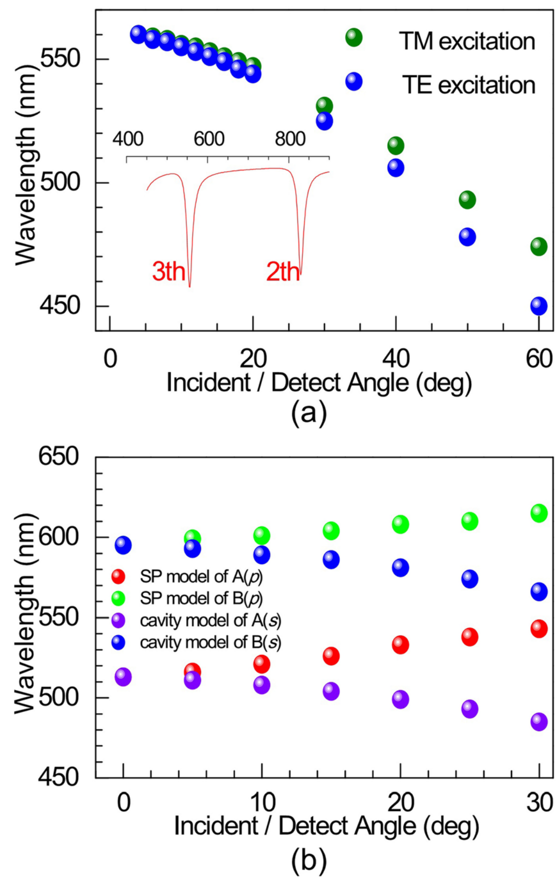

FIG. 4. (a) Angle-resolved phase plots of undoped microcavity when varying the incident angles from 0° to 60° under TM- and TE-polarized light excitation. (b) Angle-resolved phase mappings of doped hybrid system when varying the incident angles from 0° to 30° under *p*- and *s*-polarized light detection for SP mode and F–P mode studied, respectively.

The hybrid modes are grating- and cavity-induced simultaneously, and aims to elucidate the characters of the hybrid states, the measurements of phase in the TM- and TE-polarized refection beams from the hybrid grating–microcavity system are performed. Under TM-polarized incident light, both the SP and cavity modes can be excited simultaneously in this hybrid system within the same spectral window, and the sensitivity of the coupled photonic-plasmonic structure dependent on the angle arises from the strong phase dependence of the pure SP and unperturbed cavity mode.[7,11] Experimentally, the polarization before detector is *p* (TM)-polarized or *s* (TE)-polarized. Under *s*-polarized case, no SP mode exists and only the F–P mode is detected, while under *p*-polarized, reverse. The dots in Fig. 4(a) shows the phase which is a function of angle and wavelength, the unperturbed F–P mode shifts into higher energy when varying the incident angle from 0° to 60°, and the difference between TM- and TE-polarized case arises from the influence of SPPs.[11] Fig. 4(b) provides a deeper insight concerning the nature of the split modes, SP mode shifts into longer wavelength and photonic microcavity mode shifts into shorter wavelength when varying the incident angle from 0° to 30°. The SP mode of the hybrid energy of 300 nm period hybrid system (left panel in Fig. 2(b)) moves from 513 nm to 542 nm (A) and from 595 nm to 617 nm (B) under *p*-polarized detection; while the cavity mode moves from 513 nm to 484 nm (A) and from 595 nm to 576 nm (B) under *s*-polarized detection.

In conclusion, we have experimentally and theoretically studied the energy exchange between SP and F–P cavity modes via a Rabi splitting in a far-field/microcavity/nanocavity cascading energy coupling scheme. A giant Rabi-splitting energy of 336 meV was observed, and we also revealed the manipulation of strong coupling by modifying the SP ingredient employing the corrugated grating structures. We believe this cascading coupling design provides an efficient way for far-field coupling of efficient energy exchange between SP mode and F-P cavities mode, providing further concepts for plasmonic devices working.

We would like to acknowledge the financial support from the 973 project (Grant Nos. 2014CB921302 and 2011CB013003), Natural Science Foundation of China (NSFC Grant Nos. 21273096 and 61378053), and Doctoral Fund Ministry of Education of China (Grant No. 20130061110048).

[1]R. Ameling and H. Giessen, Laser Photonics Rev. **7**, 141 (2012).
[2]V. V. Protasenko and A. C. Gallagher, Nano Lett. **4**, 1329 (2004).

[3]P. Dvořák, T. Neuman, L. Břínek, T. Šamořil, R. Kalousek, P. Dub, P. Varga, and T. Šikola, Nano Lett. **13**, 2558 (2013).
[4]S. Heeg, A. Oikonomou, R. Fernandez-Garcia, C. Lehmann, S. A. Maier, A. Vijayaraghavan, and S. Reich, Nano Lett. **14**, 1762 (2014).
[5]A. M. Stefan, *Plasmonics: Fundamentals and Applications* (Springer, Berlin/Heidelberg, 2006).
[6]Y. Jiang, H.-Y. Wang, H. Wang, B.-R. Gao, Y.-W. Hao, Y. Jin, Q.-D. Chen, and H.-B. Sun, J. Phys. Chem. C **115**, 12636 (2011).
[7]Y.-W. Hao, H.-Y. Wang, Z.-Y. Zhang, X.-L. Zhang, Q.-D. Chen, and H.-B. Sun, J. Phys. Chem. C **117**, 26734 (2013).
[8]R. Ameling and H. Giessen, Nano Lett. **10**, 4394 (2010).
[9]S.-M. Chen, G.-X. Li, D.-Y. Lei, and K. W. Cheah, Nanoscale **5**, 9129 (2013).
[10]Y.-W. Hao, H.-Y. Wang, Y. Jiang, Q.-D. Chen, K. Ueno, W.-Q. Wang, H. Misawa, and H.-B. Sun, Angew. Chem. Int. Ed. **50**, 7824 (2011).
[11]S. Hayashi, Y. Ishigaki, and M. Fujii, Phys. Rev. B **86**, 045408 (2012).
[12]S. Balci, A. Kocabas, C. Kocabas, and A. Aydinli, Appl. Phys. Lett. **97**, 131103 (2010).
[13]Y.-D. Huang, H. Qin, B.-S. Zhang, J.-B. Wu, G.-C. Zhou, and B.-B. Jin, Appl. Phys. Lett. **102**, 253106 (2013).
[14]H.-Q. Le, G. W. Turner, J. R. Ochoa, M. J. Manfra, C. C. Cook, and Y.-H. Zhang, Appl. Phys. Lett. **69**, 2804 (1996).
[15]X.-Q. Wu, Y. Xiao, C. Meng, X.-N. Zhang, S.-L. Yu, Y.-P. Wang, C.-X. Yang, X. Guo, C.-Z. Ning, and L.-M. Tong, Nano Lett. **13**, 5654 (2013).
[16]H.-L. Zhang, C. Peng, A. Seetharaman, G.-P. Luo, and H.-Q. Le, Appl. Phys. Lett. **86**, 111112 (2005).
[17]B. Zhang, Z.-R. Wang, S. Brodbeck, C. Schneider, M. Kamp, S. Höfling, and H. Deng, Light Sci. Appl. **3**, e135 (2014).
[18]K. Ding and C.-Z. Ning, Light Sci. Appl. **1**, e20 (2012).