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2012

Zhang, H., Lu, H., Ho, H.-P., Zhou, Y., Yu, X., & Luan, F. (2012). Diffraction resonance with strong optical-field enhancement from gain-assisted hybrid plasmonic structure. *Applied Physics Letters*, 100(16), 161904-.

<https://hdl.handle.net/10356/95143>

<https://doi.org/10.1063/1.4704360>

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Citation: *Appl. Phys. Lett.* **100**, 161904 (2012); doi: 10.1063/1.4704360

View online: <http://dx.doi.org/10.1063/1.4704360>

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Diffraction resonance with strong optical-field enhancement from gain-assisted hybrid plasmonic structure

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(Received 14 November 2011; accepted 31 March 2012; published online 18 April 2012)

We propose and analyze the diffraction coupling of localized plasmon resonances (LPRs) through gain-assisted propagation surface plasmons (PSPs). The coupling process involves localization of incident light by LPR and LPR-PSP interaction. We demonstrate a significantly strong enhancement of electromagnetic power for LPRs in the event of diffraction resonance through incorporation of experimentally feasible optical gain to the PSP. Based on such phenomenon, we propose a hybrid plasmonic structure, which would potentially give rise to device realization of the nano-lasers. In addition, it is also a promising platform for applications such as surface enhanced Raman scattering, nonlinear optics, plasmonic trapping, etc. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4704360>]

The concentration of electromagnetic (EM) fields into subwavelength volumes is crucial to the performance of surface enhanced Raman spectroscopy (SERS),¹ nonlinear optics,² plasmonic trapping,³ photochemistry,⁴ and light-matter interactions.⁵ It is, therefore, imperative to address the question of achieving highest possible EM enhancement factor ($|E|^2$) from realistic designs. Surface plasmons (SPs) are often categorized into two classes: the propagating surface plasmons (PSPs) and the localized plasmon resonances (LPRs).⁶ It is generally accepted that a strong field enhancement arises from LPRs, and it is primarily responsible for the excitation of SERS.^{7,8} Indeed, giant EM enhancement has been demonstrated in a series of SERS strategies including gaps in metallic nanoparticle conglomerates,⁹ in the vicinities of sharp metallic tips,¹⁰ cascaded enhancements,¹¹ and diffraction resonances in silver nanoparticle arrays.¹² Another interesting and noteworthy approach towards high-field dependent single-molecule SERS (SMSERS) has already been demonstrated through a gain-assisted gold nanobox. In this design, the nanobox acts as a cavity for the surface plasmon amplification by stimulated emission of radiation (SPASER), so that a highly intensified EM field is created on the order of 10^8 .¹³ However, there are several drawbacks associated with aforementioned SERS designs including small hot-spot volume,^{9,10} fabrication difficulties,^{9,10,13} unstable chemical properties,¹² and unrealistic optical gain requirements.¹³ In this letter, we report a more feasible configuration for the resonant cavity of SPASER. Instead of a simple LPR, the hybrid SP resonances rely on diffraction coupling via PSPs to achieve a strong EM near field. Based on this hybrid plasmonic concept, we have designed a SPASER device by incorporating optical gain in the substrate^{14,15} in order to realize SPs' amplification. The device may constitute a platform for SERS detection, as well

as other high-field dependent applications at some convenient wavelengths such as 980 nm.

A schematic of the proposed hybrid plasmonic structure is shown in Fig. 1. The structure (from bottom to top) consists of an active PMMA substrate in which optical gain is made possible through doped with dye molecules, a 20 nm silver film and a 15–30 nm SiO₂ spacer. A two-dimensional square lattice of gold nanopillars with periodicity of $D = 865$ nm is deposited on top of the spacer layer. The diameter of the gold nanopillars is 150 nm and their thickness is 40 nm. We used finite-difference time domain (FDTD) simulation to calculate the near-field spectral intensity with the monitor placed at the edge of the nanopillar, as shown in Fig. 1. The monitor is oriented in a way such that the radial vector (from the disk center to the monitor) is along the polarization direction of the illumination. A linearly polarized plane wave (i.e., x-polarized light in Fig. 1) illuminates the device from the top at normal incidence. The dielectric permittivity values of gold and silver are taken from those reported by Johnson and Christy.¹⁶ The refractive index of the SiO₂ spacer is 1.46. The permittivity of the active region is complex valued and is expressed as $\epsilon = \epsilon' + \epsilon''i$. The real part is fixed at $\epsilon' = 2.25$, while the imaginary part is related to the amount of optical gain induced by external pumping. In this study, a Lorentzian index profile with a peak at

FIG. 1. Schematic of the proposed structure.

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980 nm, a 150 nm line width, and peak gain levels (ϵ'') ranging from 0 to -0.027 is used to fit the gain spectrum used in the active layer.¹⁴ We choose to terminate the simulation at 1.85 ps in order to allow the system energy decay to a negligible level and to ensure simulation convergence.

The full mechanism of diffraction coupling and hybrid SPs resonance can be briefly described as follows. Incident plane wave is first localized by LPRs in the nanopillars, whereby the nanopillars act as dipoles and excite the PSPs. In this case, the nanopillars couple their trapped EM energy to the silver film in the form of PSPs. The PSPs are then able to provide a path through which hybrid diffraction coupling between nanopillars is achieved. At certain wavelengths, the PSPs can form constructive interference, which is just the condition for diffraction resonance. These wavelengths are given by the extended Rayleigh cutoff wavelength $\lambda(i, j)_{m,air,PSP} = (D/m) \times n_{air,PSP} \times (i^2 + j^2)^{1/2}$, where D is the grating constant, m is an integer, $n_{air,PSP}$ is either the refractive index of air or the effective refractive index of PSP medium, and the integers i, j denote the grating diffraction order.¹⁷ This process gives rise to the resonance peaks indicated by the near-field intensity enhancement ($|E|^2$) spectra of the monitor as shown in Fig. 2. At this stage, four different thicknesses of the SiO₂ spacer are investigated. The inset of Fig. 2 shows the near-field spectrum of an isolated nanopillar when the spacer has a thickness of 20 nm. The maximum electric field intensity at the monitor is on the order of 10^5 . This amplitude is in line with results obtained from similar structures reported in literature.¹⁸ In Fig. 2, the peaks found in the 920–950 nm region (and also at 910 nm in the inset) are attributed to the dipolar LPRs of the nanopillars and their mirror images in silver film. The small peak at around 725 nm (and also at 643 nm in the inset) is induced by their quadrupole LPR modes. The general red shift in the spectra is due to collective plasmon resonance from interparticle coupling. The peak at around 865 nm comes from the (1, 0) order of normal diffraction coupling through air (i.e., $\lambda(1, 0)_{1,air} = 865$ nm). The peaks at 980 nm and 1328 nm (enlarged in the lower inset of Fig. 2) are from the (1, 1) and (1, 0) order of the diffraction resonance via PSPs coupling (i.e., $\lambda(1, 1)_{2,PSP} = 980$ nm and $\lambda(1, 0)_{1,PSP} = 1328$ nm, with $n_{PSP} \approx 1.6$ and $n_{PSP} \approx 1.54$, respectively, which will be further explained later). Their asymmetric shape suggests an associ-

FIG. 2. Near-field response spectra at the monitor for different SiO₂ spacer thicknesses. Upper inset: near-field response spectrum of an isolated nanopillar with spacer thickness = 20 nm. Lower inset: enlarged view of near-field spectra for the range of 1300–1350 nm.

ation with Fano resonance,¹⁹ which is induced by interference between two energy coupling paths, namely the path within a continuum of states (resonance of the isolated nanopillars and their images) and the one within a discrete state (the diffraction resonances via PSPs). As shown in Fig. 2, at 980 nm, the highest coupling efficiency takes place when the spacer is 20 nm thick. We also found out that the PSP induced diffraction resonances ($\lambda(1, 1)_{2,PSP} = 980$ nm) has comparable intensities with that of the diffraction resonance through air ($\lambda(1, 0)_{1,air}$). This happens because the EM energy of PSPs is more confined to a planar region than the case of free-space or air, despite that the silver film has an intrinsic ohmic loss. For SPs excited on the silver film, the penetration depth is usually more than 30 nm for a spectral range of 600–1500 nm.⁶ Hence, a 20 nm thick silver film is able to allow the optical gain applied to one side being used to compensate for the PSP losses on both sides.

To understand the role of optical gain, as well as to demonstrate the mechanism of PSP induced diffractive resonance, we calculated the PSPs generated by a multilayer structure in the absence of the nanopillar array. A reflection pole method is formulated to enable computation of PSP's effective mode index.²⁰ When the spacer is 20 nm thick, from Fig. 3(a), we see that the real part of PSP's effective mode index (n_{PSP}) is about 1.6 at 980 nm, while ϵ'' varies from -0.01 to -0.03 , and it experiences a sudden drop because the mode cuts off at a longer wavelength. In Fig. 3(b), zero or negative values of the imaginary part denote zero-loss or amplified PSPs, respectively. When the absolute value of ϵ'' is larger than 0.01 (equivalent to a gain

FIG. 3. PSP's effective mode refractive index versus different optical gain levels (in terms of ϵ'') in the substrate with (a) for the real part and (b) for the imaginary part. (c) Near-field spectra for different gain levels. Inset shows an enlarged version of response spectrum when the optical gain approaches a level of $\epsilon'' = -0.025$.

coefficient $(2\pi/\lambda)\varepsilon''/\sqrt{\varepsilon'} = 427.42 \text{ cm}^{-1}$ at $\lambda = 980 \text{ nm}$), the optical gain can fully compensate the intrinsic PSPs loss in silver at around 980 nm. This critical gain level is in line with the value reported in literature.²¹ In reality, the overall structure requires a slightly higher gain level because of losses in the gold nanopillars and imperfect LPRs-PSPs coupling. The near-field spectra of the monitor for a 20 nm thick spacer and for different gain levels are shown in Fig. 3(c). Without optical gain, strong resonance is barely observable. As optical gain is introduced into the system, the near-field intensity at 980 nm increases dramatically. The inset in Fig. 3(c) reveals more details when the optical gain is $\varepsilon'' = -0.025$, a level that approaches but is still below the lasing threshold at 980 nm. This value corresponds to a gain coefficient of 1068.57 cm^{-1} and the near-field intensity has increased to $2.38 \times 10^7 \text{ V}^2/\text{m}^2$ in this case. If the gain level is further increased to reach the lasing threshold ($\varepsilon'' = -0.027$), amplification of light will balance the extinction of the system. To obtain the near-field spectra, the system is first illuminated by a multi-frequency pulse, followed by applying Fourier transform to its time-domain response. The lasing threshold is determined as such because the system will no longer decay but diverge in the time domain if the absolute value of ε'' is bigger than 0.027. Consequently, a SPASER-like phenomenon occurs with the emergence of a very sharp LPR peak. In Fig. 3(c), the plot produced with $\varepsilon'' = -0.027$ (i.e., the lasing threshold) is a manifestation of the SPASER-like process. In this study, we simply use negative imaginary part of permittivity to represent the substrate's optical gain, which is a common approach adopted by many other studies.^{13–15,22–26} Note that, such treatment of the optical gain accurately presents an effective gain level within the scope of small signal gain, and it contains simulated analysis of the entire dynamic saturation phenomenon in a real lasing system. Our treatment facilitates a comparison with the results of other work that have adopted similar treatments of the optical gain and reveals a lower threshold requirement in terms of gain level in our device.^{13,23} In view of the complexity of this type of system, many research groups are focusing on the analysis of more detailed and in-depth physical processes involved.^{24,27} It is shown that the fluorophore-doped PMMA can be represented by a dipolar gain medium having a permittivity with its imaginary part dependent on the pump light distribution and quenching effect from metal surfaces.²⁴ From an experimental viewpoint, a more detailed and comprehensive consideration of the pumping and quenching should be deliberated in order to achieve a specific effective gain for certain PSP modes.

Fig. 4(a) shows the electric field intensity pattern when $\varepsilon'' = -0.025$ and $\lambda = 980 \text{ nm}$, for a unit element of the grating in a XY plane at the nanopillar-spacer interface. In this case, the EM enhancement factor is on the order of 10^7 . It is observed that the maximum field intensity focuses on the circumference of the nanopillar rather than some narrow gaps or small spots. Such distribution pattern increases the SERS detection volume, which in turn makes the SMSERS device more feasible. Figure 4(b) shows the directional pattern of the Poynting vector at the lower surface of the silver film, which clearly reveals the underlying mechanism of the PSP induced diffraction resonance. In Fig. 4(b), the diagonal in a quaternary

FIG. 4. (a) The local-field distribution pattern for a unit element of the grating when we use $\varepsilon'' = -0.025$ and resonant wavelength of 980 nm. It comes from an XY plane located at the nanopillar-spacer interface. (b) Directional pattern of the Poynting vector at the lower surface of the silver film. The dash box represents the two nearest nanopillar (highlighted by dash circle) in a diagonal direction. (c) Schematic of the charge distribution for highlighted region shown in (b).

unit of the grating contains two entire wave profiles ($m = 2$) that are associated with the (1, 1) order diffraction resonance generated through PSP coupling with (i.e., $\lambda(1, 1)_{2,PSP} = 980 \text{ nm}$ with $n_{PSP} \approx 1.6$). We also schematically show in Fig. 4(c) the charge distribution around a cross-section covering two nanopillars. The laser threshold in terms of gain coefficient in our design is $\sim 10^3 \text{ cm}^{-1}$, which is within the limits of semiconductor polymers and laser dyes (highly concentrated).⁶ As compared to the device scheme based on nanobox,¹³ our design offers several significant merits: simple fabrication, lower gain requirement, and larger “hot spots” volume. With these attributes, diffraction coupling of LPRs through gain-assisted PSPs offers an ideal platform for SERS. In SERS theory, it is well accepted that the metal nanoparticles act as efficient antenna that couple light energy (with frequency ω) to nearby molecules while at the same time capture and re-radiate the energy of the Raman-shifted light (with frequency ω'). Kerker *et al.* have demonstrated that this leads to a strong enhancement factor which is proportional to the local field $|E(\omega)|^2$ multiplied by $|E(\omega')|^2$.^{7,8} It is possible to further optimize this platform through better selection of nanoparticles to cater for the amplification of specific peaks or to carefully choose the grating constants in order to generate multi-peak resonance for double-resonance SERS,²⁸ as well as SMSERS, which usually requires an enhancement factor of 10^6 – 10^8 .¹

In conclusion, we have presented a theoretical analysis of diffraction coupling in a gain-assisted hybrid plasmonic structure. Based on this concept, we demonstrate a device that can provide an EM enhancement factor as high as 10^7 . Our hybrid structure is observed to exhibit merits such as multiple tunable resonant peaks and large high-field volume, which are attractive for ultra-sensitive SERS detection, nonlinear optics, plasmonic trapping, etc. Moreover, the Fano resonances in the hybrid structure are very useful for sensing applications based on customized plasmonic properties. Finally, our results reveal that gain-assisted hybrid SPs may provide a SPASER design framework in which the gain medium is separated from the nanoparticle.

The authors would like to acknowledge the financial support from RGC Central Allocation Grant (HKUST 2/07C) for a research studentship (HXZ), from Chinese University of Hong Kong Group Research Grant (3110048) and a research student (HFL). Support from the Singapore

Institute of Manufacturing Technology (SIMTech) for exchange student scholarship (HXZ and HFL) is gratefully acknowledged.

- ¹C. L. Haynes, A. D. McFarland, and R. P. Van Duyne, *Anal. Chem.* **77**, 338A–346A (2005).
- ²S. Kim, J. Jin, Y.-J. Kim, I.-Y. Park, Y. Kim, and S.-W. Kim, *Nature (London)* **453**, 757–760 (2008).
- ³M. Juan, M. Righini, and R. Quidant, *Nat. Photonics* **5**, 349–356 (2011).
- ⁴K. Watanabe, D. Menzel, N. Nilius, and H.-J. Freund, *Chem. Rev.* **106**, 4301 (2006).
- ⁵T. H. Taminiau, F. D. Stefani, F. B. Segerink, and N. F. van Hulst, *Nat. Photonics* **2**, 234 (2008).
- ⁶V. M. Shalaev and S. Kawata, *Nanophotonics with Surface Plasmons* (Elsevier, Amsterdam, The Netherlands, 2007).
- ⁷M. Kerker, D.-S. Wang, and H. Chew, *Appl. Opt.* **19**, 4159 (1980).
- ⁸T. J. Davis, D. E. Gómez, and K. C. Vernon, *Phys. Rev. B* **82**, 205434 (2010).
- ⁹P. J. Schuck, D. P. Fromm, A. Sundaramurthy, G. S. Kino, and W. E. Moerner, *Phys. Rev. Lett.* **94**, 017402 (2005).
- ¹⁰X. Wang, Z. Liu, M. D. Zhuang, H. M. Zhang, X. Wang, Z. X. Xie, D. Y. Wu, B. Ren, and Z. Q. Tian, *Appl. Phys. Lett.* **91**, 101105 (2007).
- ¹¹K. Li, M. I. Stockman, and D. J. Bergman, *Phys. Rev. Lett.* **91**, 227402 (2003).
- ¹²S. Zou and G. C. Schatz, *Chem. Rev. Lett.* **403**, 62–67 (2005).
- ¹³Z. Y. Li and Y. Xia, *Nano Lett.* **10**, 243 (2010).
- ¹⁴X. F. Li, S. F. Yu, and A. Kumar, *Appl. Phys. Lett.* **95**, 141114 (2009).
- ¹⁵A. Kobyakov, A. R. Zakharian, K. M. Gundu, and S. A. Darmany, *Appl. Phys. Lett.* **94**, 151111 (2009).
- ¹⁶P. B. Johnson and R. W. Christy, *Phys. Rev. B* **6**, 4370 (1972).
- ¹⁷V. G. Kravets, F. Schedin, and A. N. Grigorenko, *Phys. Rev. Lett.* **101**, 087403 (2008).
- ¹⁸H. C. Kim and X. Cheng, *Opt. Express* **17**, 17234 (2009).
- ¹⁹A. E. Miroshnichenko, S. Flach, and Y. S. Kivshar, *Rev. Mod. Phys.* **82**, 2257 (2010).
- ²⁰E. Anemogiannis, E. N. Glytsis, and T. K. Gaylord, *J. Lightwave Technol.* **17**, 929 (1999).
- ²¹M. A. Noginov, V. A. Podolskiy, G. Zhu, M. Mayy, M. Bahoura, J. A. Adegoke, B. A. Ritzo, and K. Reynolds, *Opt. Express* **16**, 1385 (2008).
- ²²A. A. Govyadinov, V. A. Podolskiy, and M. A. Noginov, *Appl. Phys. Lett.* **91**, 191103 (2007).
- ²³J. A. Gordon and R. W. Ziolkowski, *Opt. Express* **15**, 2622–2653 (2007).
- ²⁴I. De Leon and P. Berini, *Phys. Rev. B* **78**(16), 161401 (2008).
- ²⁵N. I. Zheludev, S. L. Prosvirnin, N. Papasimakis, and V. A. Fedotov, *Nat. Photonics* **2**(6), 351–354 (2008).
- ²⁶A. K. Sarychev and G. Tartakovsky, *Phys. Rev. B* **75**, 085436 (2007).
- ²⁷G. Colas des Francs, P. Bramant, J. Grandidier, A. Bouhelier, J.-C. Weeber, and A. Dereux, *Opt. Express* **18**, 16327 (2010).
- ²⁸Y. Chu, M. G. Banaee, and K. B. Crozier, *ACS Nano* **4**(5), 2804–2810 (2010).