



Towards dielectric sensing via Babinet's principle-inspired plasmonic platforms

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Abstract

Localized surface plasmon (LSP) resonances in plasmonic micro-/nanostructures such as metallic spheres and cylinders, exhibit sensitivity to nearby external changes, such as variations in the refractive index of a dielectric close to the structures. This useful characteristic can be exploited in multiple fields, including medicine, and chemistry. Here, we make use of Babinet's principle of complementarity (a concept developed for low frequencies) to design plasmonic structures at optical frequencies. The structures consist of complementary metal-dielectric cylindrical dimers and apertures on a metal film. Their performance is evaluated both experimentally and numerically demonstrating their ability to be used for sensing applications.

1. Introduction

The confinement of electromagnetic (EM) waves at optical frequencies using localized surface plasmons (LSPs) in metallic micro-/nanostructures [1]–[3] and surface plasmon polaritons (SPPs) on metal-dielectric interfaces [4]–[7] has attracted the interest of the scientific community in the past years. LSP resonances [8]–[14], have been exploited in diverse applications such as solar cells [15], optical trapping [16] and photo-catalysis [17], among others.

The properties of LSP resonance within a plasmonic structure are influenced by parameters such as size, shape [18], [19] the metal composition and the surrounding dielectric [20]. This dependence means changing any one of these parameters will cause an observable shift in the spectral position of the LSP resonance including, for example, variations in the refractive index of a nearby dielectric film. Consequently, LSPs have excelled in many fields [1], [21], [22] where sensing local changes is required such as biosensing [23], [24] and surface-enhanced Raman spectroscopy [25], [26], among others.

The application of classical concepts, such as Babinet's principle in plasmonics has provided valuable insights for the design of plasmonic nanostructures [2], [18], [27]–[30]. Babinet's principle states that the diffraction patterns from

a set of infinitely thin, perfectly opaque particles and a complementary set of apertures in an infinitely thin, perfectly opaque sheet will be identical [31]. Therefore, in the context of LSPs, when comparing between the complementary plasmonic particle and aperture cases, it would be expected that the LSP resonances will be excited at similar spectral position. Moreover, their respective near-field electric and magnetic field distributions will also be complementary [27], [32]. However, when applying Babinet's principle to plasmonics, some slight discrepancies occur as metals, at optical frequencies, can no longer be considered as perfect electric conductors as is the case for lower frequencies [18].

In this communication, complementary plasmonic structures consisting of cylindrical metallic particle-dimers and aperture-dimers will be investigated when a thin dielectric film is positioned on top of them [33]. The spectral response of the proposed plasmonic structures is evaluated both numerically and experimentally. Moreover, the sensing performance of the two complementary plasmonic structures are evaluated and compared as the refractive index of the dielectric film is varied. An in-depth study of their performance will be discussed during the conference.

2. Results and discussion

The schematic representation of the particle-dimer and aperture-dimer on their xy -surface is shown in Fig. 1a,b, respectively. Here, the cylindrical dimers can be seen as gold (Au) features or apertures in an Au film. The dimers are placed on a silicon nitride (Si_3N_4) substrate with the whole structure immersed in air ($n_{\text{air}} = 1$). A cross-section of the complementary particle- and aperture-dimers are shown in Fig. 1c,d, respectively. The Au and Si_3N_4 layers are both 30 nm thick and they are modeled using experimental data from [34], [35], respectively. A dielectric film (which will act as the analyte to be sensed) is then positioned on top of the plasmonic structures with a variable refractive index, n_a . The dimers have a diameter D , and S is the separation distance between the two cylinders. For the study shown here, values of $D = 200$ nm and $S = 20$ nm were chosen due to accessibility for their experimental demonstration [33].

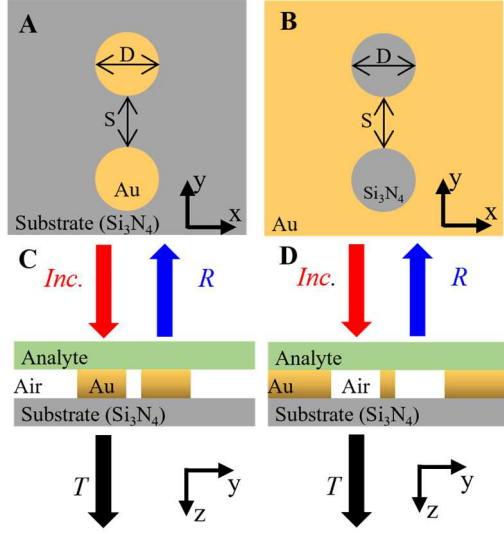


Figure 1. (a,b) Schematic of particle- and aperture-dimers. D and S represent the dimer diameter and separation, respectively. (c,d) Cross-section on the yz -plane with incident planewave (red). Reflection (blue) and transmission (black).

To evaluate the structures, numerical studies are carried out using the commercial software COMSOL Multiphysics®. An incident planewave, propagating in the z -direction, polarized along the x - (E_x) or y - (E_y) direction (as represented by the red arrow labelled $Inc.$ in Fig. 1b,d) is used to illuminate the plasmonic structures. By changing the polarization of the incident planewave, then complementary LSP resonant modes will be excited in each of the particle- and aperture-dimers. To determine the spectral position of each LSP resonance, the reflection (blue arrow in Fig. 1c,d) and transmission (black arrow in Fig. 1c,d) spectrum is calculated of each case. The spectral position of the LSP resonances of these plasmonic structures will observably change with variations of the n_a of the dielectric, enabling these devices to be used as a sensor.

For the analyte, a thickness of 50 nm is fixed such that the only variations in the spectral location of the LSP resonance will be due to changes of n_a . The sensitivity of each complementary plasmonic structure can then be determined by taking the ratio between the change in the LSP resonant spectral position and the change in the refractive index of the dielectric analyte, $S = \frac{\Delta\lambda}{\Delta n_a}$ [nm/RIU], where RIU stands for the refractive index unit [36], [37]. The calculated sensitivity for the complementary structures, as the n_a changes is shown in Fig. 2 with the results for the particle-dimers and aperture-dimers presented as red and blue curves, respectively. As the structures are illuminated with orthogonally polarized planewaves the E_x -illumination is represented by solid lines and the dashed lines represent the E_y -illumination.

For particle- and aperture-dimers illuminated by a E_x and E_y polarized planewave, respectively, the maximum values of the sensitivity (S_{max}) in Fig. 2 are $S_{max} = 207$ and 135 nm/RIU, respectively. The opposite polarization of the planewave, E_y and E_x polarized planewaves, respectively, obtain values of $S_{max} = 252$ and 222 nm/RIU, respectively. From Fig. 2, the plasmonic structures have the highest S_{max} when using E_y and E_x polarized planewaves on the particle- and aperture-dimers, respectively. This improvement in the performance of the complementary plasmonic structures is a result of complementary LSP modes with a field hotspot being excited. This field hotspot enhances the interaction with the dielectric analyte compared to the field distribution of LSP modes under orthogonal polarizations, resulting in higher sensitivities.

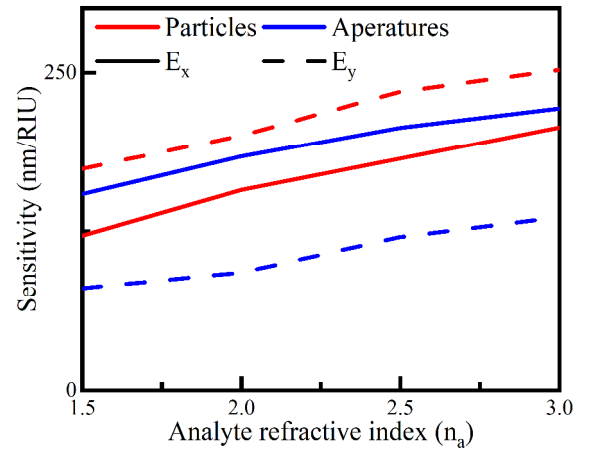


Figure 2. Sensitivity of the complementary particle-dimers (blue) and aperture-dimers (red) illuminated by an E_x (solid) and an E_y (dashed) polarized planewave as the refractive index of the dielectric analyte changes.

In the conference, the validity of this work will be verified with experimentally fabricated devices. Furthermore, an in-depth analysis of the complementary electric and magnetic near-field distributions will be given alongside a more comprehensive discussion on the sensing performance. Further studies include the effect of changes of analyte configuration in the performance of the sensors such as varying the dielectric thickness, immersing/filling the particle-/aperture-dimers and using a rectangular particle of analyte [33].

3. Conclusion

In conclusion, we have shown in this communication, the design of complementary plasmonic particle- and aperture-dimers to support LSP resonant modes via Babinet's principle for complementarity. This was exploited to detect changes in the refractive index of a 50 nm thick sheet of dielectric analyte positioned on top of the complementary plasmonic structures by observing the change in the wavelength of the LSP resonance of each case. To evaluate the performance of these plasmonic-based sensing devices the sensitivity was calculated for each refractive index of the analyte, demonstrating sensitivities of 130 nm/RIU up

to 250 nm/RIU. Our results may be exploited in sensing devices within areas such as biology and chemistry.

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References

- [1] V. Pacheco-Peña, R. Alves, and M. Navarro-Cía, “Hidden Symmetries in Bowtie Nanocavities and Diabolo Nanoantennas,” *ACS Photonics*, vol. 6, no. 8, pp. 2014–2024, Aug. 2019, doi: 10.1021/acsp Photonics.9b00428.
- [2] T. Zentgraf *et al.*, “Babinet’s principle for optical frequency metamaterials and nanoantennas,” *Phys. Rev. B - Condens. Matter Mater. Phys.*, vol. 76, no. 3, pp. 4–7, 2007, doi: 10.1103/PhysRevB.76.033407.
- [3] V. Pacheco-Peña, M. Beruete, A. I. Fernández-Domínguez, Y. Luo, and M. Navarro-Cía, “Description of Bow-Tie Nanoantennas Excited by Localized Emitters Using Conformal Transformation,” *ACS Photonics*, vol. 3, no. 7, pp. 1223–1232, Jul. 2016, doi: 10.1021/acsp Photonics.6b00232.
- [4] S. A. Maier, *Plasmonics: Fundamentals and Applications*, 1st ed. 2007. New York, NY: Springer US, 2007. doi: 10.1007/0-387-37825-1.
- [5] J. A. Riley, N. Healy, and V. Pacheco-Peña, “Plasmonic meniscus lenses,” *Sci. Rep.*, vol. 12, no. 1, p. 894, 2022, doi: 10.1038/s41598-022-04954-0.
- [6] T. Zentgraf, Y. Liu, M. H. Mikkelsen, J. Valentine, and X. Zhang, “Plasmonic Luneburg and Eaton lenses,” *Nat. Nanotechnol.*, vol. 6, no. 3, pp. 151–155, Mar. 2011, doi: 10.1038/nnano.2010.282.
- [7] V. Pacheco-Peña, I. V. Minin, O. V. Minin, and M. Beruete, “Comprehensive analysis of photonic nanojets in 3D dielectric cuboids excited by surface plasmons,” *Ann. Phys.*, vol. 528, no. 9–10, pp. 684–692, Oct. 2016, doi: 10.1002/andp.201600098.
- [8] S. A. Maier, *Plasmonics: Fundamentals and Applications*. New York, NY: Springer US, 2007. doi: 10.1007/0-387-37825-1.
- [9] G. Barbillon, “Plasmonics and its Applications,” *Materials (Basel)*, vol. 12, no. 9, p. 1502, May 2019, doi: 10.3390/ma12091502.
- [10] L. Novotny and B. Hecht, “Surface plasmons,” in *Principles of Nano-Optics*, no. May, Cambridge: Cambridge University Press, 2012, pp. 369–413. doi: 10.1017/CBO9780511794193.014.
- [11] E. Ozbay, “Plasmonics: Merging Photonics and Electronics at Nanoscale Dimensions,” *Science (80-*, vol. 311, no. 5758, pp. 189–193, Jan. 2006, doi: 10.1126/science.1114849.
- [12] M. Zhang *et al.*, “Nanoimprinted Chiral Plasmonic Substrates with Three-Dimensional Nanostructures,” *Nano Lett.*, vol. 18, no. 11, pp. 7389–7394, Nov. 2018, doi: 10.1021/acs.nanolett.8b03785.
- [13] H. Aouani, M. Rahmani, M. Navarro-Cía, and S. A. Maier, “Third-harmonic-upconversion enhancement from a single semiconductor nanoparticle coupled to a plasmonic antenna,” *Nat. Nanotechnol.*, vol. 9, no. 4, pp. 290–294, 2014, doi: 10.1038/nnano.2014.27.
- [14] N. J. Greybush, V. Pacheco-Peña, N. Engheta, C. B. Murray, and C. R. Kagan, “Plasmonic Optical and Chiroptical Response of Self-Assembled Au Nanorod Equilateral Trimers,” *ACS Nano*, vol. 13, no. 2, pp. 1617–1624, 2019, doi: 10.1021/acsnano.8b07619.
- [15] P. Li, X. Jiang, S. Huang, Y. Liu, and N. Fu, “Plasmonic perovskite solar cells: An overview from metal particle structure to device design,” *Surfaces and Interfaces*, vol. 25, p. 101287, 2021, doi: 10.1016/j.surf.2021.101287.
- [16] B. J. Roxworthy *et al.*, “Application of plasmonic bowtie nanoantenna arrays for optical trapping, stacking, and sorting,” *Nano Lett.*, vol. 12, no. 2, pp. 796–801, 2012, doi: 10.1021/nl203811q.
- [17] M. Lemos de Souza, D. Pereira Dos Santos, and P. Corio, “Localized surface plasmon resonance enhanced photocatalysis: an experimental and theoretical mechanistic investigation,” *RSC Adv.*, vol. 8, no. 50, pp. 28753–28762, 2018, doi: 10.1039/c8ra03919d.
- [18] M. Horák *et al.*, “Limits of Babinet’s principle for solid and hollow plasmonic antennas,” *Sci. Rep.*, vol. 9, no. 1, pp. 1–11, 2019, doi: 10.1038/s41598-019-40500-1.
- [19] M. W. Chu, V. Myroshnychenko, C. H. Chen, J. P. Deng, C. Y. Niou, and F. J. G. De Abajo, “Probing bright and dark surface-plasmon modes in individual and coupled noble metal nanoparticles using an electron beam,” *Nano Lett.*, vol. 9, no. 1, pp. 399–404, 2009, doi: 10.1021/nl803270x.
- [20] K. R. Ryu and J. W. Ha, “Influence of shell thickness on the refractive index sensitivity of localized surface plasmon resonance inflection points in silver-coated gold nanorods,” *RSC Adv.*, vol. 10, no. 29, pp. 16827–16831, 2020, doi: 10.1039/d0ra02691c.
- [21] V. Pacheco-Peña and M. Navarro-Cía, “Understanding quantum emitters in plasmonic nanocavities with conformal transformation: Purcell enhancement and forces,” *Nanoscale*, vol. 10, no. 28, pp. 13607–13616, 2018, doi: 10.1039/c8nr01527a.
- [22] V. Pacheco-Peña, M. Beruete, A. I. Fernández-Domínguez, Y. Luo, and M. Navarro-Cía, “Description of Bow-Tie Nanoantennas Excited by Localized Emitters Using Conformal Transformation,” *ACS Photonics*, vol. 3, no. 7, pp.

- 1223–1232, 2016, doi: 10.1021/acsphotonics.6b00232.
- [23] D. M. Kim, J. S. Park, S.-W. Jung, J. Yeom, and S. M. Yoo, “Biosensing Applications Using Nanostructure-Based Localized Surface Plasmon Resonance Sensors,” *Sensors*, vol. 21, no. 9, p. 3191, May 2021, doi: 10.3390/s21093191.
- [24] G. Das *et al.*, “Plasmonic nanostructures for the ultrasensitive detection of biomolecules,” *Riv. del Nuovo Cim.*, vol. 39, no. 11, pp. 547–586, 2016, doi: 10.1393/ncr/i2016-10129-y.
- [25] P. L. Stiles, J. A. Dieringer, N. C. Shah, and R. P. Van Duyne, “Surface-enhanced Raman spectroscopy,” *Annu. Rev. Anal. Chem.*, vol. 1, no. 1, pp. 601–626, 2008, doi: 10.1146/annurev.anchem.1.031207.112814.
- [26] S. Schlücker, “Surface-enhanced Raman spectroscopy: Concepts and chemical applications,” *Angew. Chemie - Int. Ed.*, vol. 53, no. 19, pp. 4756–4795, 2014, doi: 10.1002/anie.201205748.
- [27] J. D. Ortiz, J. P. del Risco, J. D. Baena, and R. Marqués, “Extension of Babinet’s principle for plasmonic metasurfaces,” *Appl. Phys. Lett.*, vol. 119, no. 16, p. 161103, Oct. 2021, doi: 10.1063/5.0065724.
- [28] M. Navarro-Cia, M. Beruete, and M. Soroll, “Electromagnetic Response of Extraordinary Transmission Plates Inspired on Babinet’s Principle,” in *Behaviour of Electromagnetic Waves in Different Media and Structures*, no. June, InTech, 2011. doi: 10.5772/18036.
- [29] F. Falcone *et al.*, “Babinet Principle Applied to the Design of Metasurfaces and Metamaterials,” *Phys. Rev. Lett.*, vol. 93, no. 19, p. 197401, Nov. 2004, doi: 10.1103/PhysRevLett.93.197401.
- [30] V. Krápek *et al.*, “Independent engineering of individual plasmon modes in plasmonic dimers with conductive and capacitive coupling,” *Nanophotonics*, vol. 9, no. 3, pp. 623–632, 2020, doi: 10.1515/nanoph-2019-0326.
- [31] M. Born and E. Wolf, “Elements of the theory of diffraction,” in *Principles of Optics*, Cambridge University Press, 2019, pp. 412–516. doi: 10.1017/9781108769914.011.
- [32] M. Hrtoň, A. Konečná, M. Horák, T. Šikola, and V. Krápek, “Plasmonic Antennas with Electric, Magnetic, and Electromagnetic Hot Spots Based on Babinet’s Principle,” *Phys. Rev. Appl.*, vol. 13, no. 5, p. 054045, May 2020, doi: 10.1103/PhysRevApplied.13.054045.
- [33] J. A. Riley, M. Horák, V. Krápek, N. Healy, and V. Pacheco-Peña, “Plasmonic sensing using Babinet’s principle,” *Nanophotonics*, pp. 53–56, Sep. 2023, doi: 10.1515/nanoph-2023-0317.
- [34] P. B. Johnson and R. W. Christy, “Optical Constant of the Nobel Metals,” *Phys. L Re View B*, vol. 6, no. 12, pp. 4370–4379, 1972.
- [35] H. R. Philipp, “Optical Properties of Silicon Nitride,” *J. Electrochem. Soc.*, vol. 120, no. 2, p. 295, 1973, doi: 10.1149/1.2403440.
- [36] M. Beruete, N. Engheta, and V. Pacheco-Peña, “Experimental demonstration of deeply subwavelength dielectric sensing with epsilon-near-zero (ENZ) waveguides,” *Appl. Phys. Lett.*, vol. 120, no. 8, 2022, doi: 10.1063/5.0079665.
- [37] M. Alagdar, B. Yousif, N. F. Areed, and M. Elzalabani, “Improved the quality factor and sensitivity of a surface plasmon resonance sensor with transition metal dichalcogenide 2D nanomaterials,” *J. Nanoparticle Res.*, vol. 22, no. 7, 2020, doi: 10.1007/s11051-020-04872-0.