Title	Influence of nanoparticle dimensions on Rabi splitting strength
Author(s)	Lawless, Julia; Hrelescu, Calin; Elliott, Carolyn; Peters, Lisanne; McEvoy, Niall; Bradley, A. Louise
Publication date	2021-10-29
Original citation	Lawless, J., Hrelescu, C., Elliott, C., Peters, L., McEvoy, N. and Bradley, A. L. (2021) 'Influence of nanoparticle dimensions on Rabi splitting strength', 2021 Conference on Lasers and Electro-Optics (CLEO), San Jose, CA, USA, 9-14 May. Available at: https://ieeexplore.ieee.org/abstract/document/9571584 (Accessed: 28 March 2022)
Type of publication	Conference item
Rights	**© 2021, the Authors.**
Item downloaded from	http://hdl.handle.net/10468/12997

Influence of Nanoparticle Dimensions on Rabi Splitting Strength

Julia Lawless[1]*, Calin Hrelescu[1], Carolyn Elliott[1,3], Lisanne Peters[2], Niall McEvoy[2] and A. Louise Bradley[1,3]**

1. School of Physics and AMBER, Trinity College Dublin, College Green, Dublin 2, Ireland.
2. School of Chemistry and AMBER, Trinity College Dublin, College Green, Dublin 2, Ireland.
3. IPIC, Tyndall National Institute, Cork, Ireland.
*lawlesju@tcd.ie,**bradlel@tcd.ie

Abstract: Strong coupling was detected between single gold nano-bipyramids and monolayer MoS_2. It was demonstrated that the coupling strength increases with nanoparticle size, even without increasing the number of excitons coupled into the system.

Rabi splitting is a light-matter interaction in which the rate of exchange of energy between a quantum emitter and an optical resonator mode is larger than their intrinsic dissipation rates. When these conditions are met, the resonance splits into two new eigenstates with energies above and below the original excitations. This has a myriad of applications including a low threshold laser, optical switching and modification of the rate of chemical reactions [1].

In the past, observations of Rabi splitting have been confined to cryogenic temperatures. With the developments of nanoscience, however, it has become possible to achieve Rabi splitting at room temperature, and with plasmons of metal nanoantennae instead of a closed optical cavity.

In this work, strong coupling is demonstrated using single gold bipyramids drop-cast onto monolayer MoS_2 flakes on a SiO_2/Si substrate. Bipyramids of 60 to 115 nm in length were chemically synthesised using a wet chemistry approach, and the MoS_2 flakes were grown on the SiO_2/Si substrates using chemical vapour deposition. The polaritons were detected by dark field scattering, which was correlated with scanning electron microscopy images which showed the sizes of the bipyramids.

Bipyramids prove to be the ideal geometry to achieve the effect of Rabi splitting in this system, for several reasons. Firstly, the electric field enhancement is confined at the very sharp tips of the bipyramid. This effect is important because the coupling strength, $g \propto \frac{1}{\sqrt{V}}$, where V is the mode volume of the resonator [2]. The mode volume is further reduced due to the asymmetric nature of the bipyramid system, with only one of the bipyramid's sharp tips in contact with the surface (see figure 1a). This effect further improves the bipyramid's capacity for strong coupling because it tilts the plasmon resonance towards the MoS_2 layer. This is important because similar structures involving different types of nanoparticles can have a reduced coupling strength due to the misalignment of the plasmon resonance with the dipole moment [3].

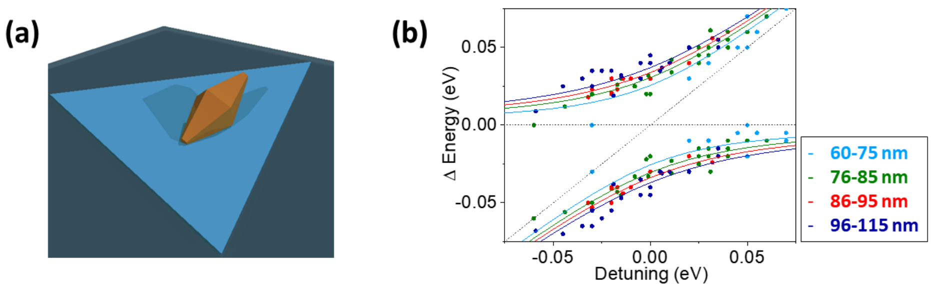

Fig. 1. (a) Schematic of a single bipyramid on a monolayer MoS_2 flake on an SiO_2/Si substrate. (b) Rabi splitting plot of the energy of the two polaritons plotted against the detuning of the uncoupled plasmon from the A exciton of MoS_2. A larger splitting is seen for longer bipyarmids.

Figure 1b shows the Rabi splitting plot of the energy of the two polariton peaks plotted against the energy of the original, uncoupled plasmon for several hundred different bipyramids. The energy is plotted in terms of the detuning from the energy of the A exciton of MoS_2. The coupled oscillator model was used to fit the results [4]:

$$E_{up/lp} = \frac{E_{pl} + E_{ex}}{2} \pm \sqrt{g^2 + \frac{1}{4}\left[(E_{pl} - E_{ex}) - \frac{i}{2}(\gamma_{pl} - \gamma_{ex})\right]^2} \quad (1)$$

with the larger and smaller energy values corresponding to the upper and lower polaritons, respectively. The coupling constant is given by g, the energy of the plasmon and exciton are given by E_{pl} and E_{ex} and the spontaneous dissipation rates of the uncoupled plasmon and exciton are given by γ_{pl} and γ_{ex}.

The plot clearly shows that the coupling strength increases for larger bipyarmids. The Rabi splitting for bipyramids tuned to overlap in energy with the exciton, Ω, was shown to range from ≈ 55 meV for a 70 nm long bipyramid to ≈ 80 meV for a 100 nm long bipyramid.

There are two reasons for this increased coupling strength for larger bipyramids. Firstly, a larger bipyramid has a larger electric field strength at its tip (figure 2) because of the increased number of electrons increasing the strength of the plasmon. This increases the interaction with the MoS_2 layer. Secondly, in order to keep the plasmon energies tuned to the energy of the exciton, the aspect ratio (length to width) had to be reduced for the longer bipyramids. Therefore, the angle between the plasmon direction and the substrate was larger for the longer bipyramids, causing the plasmon to overlap more strongly with the MoS_2 layer.

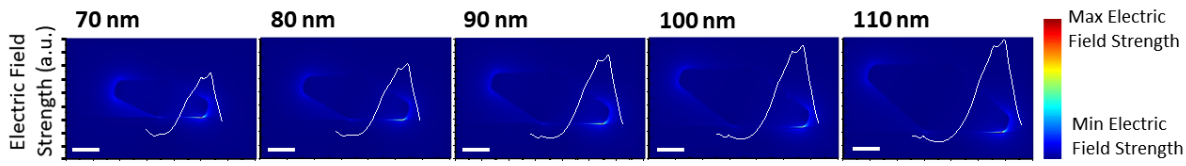

Fig. 2. Electric field maps of the center cross section for differently sized bipyramids. White overplots show the average electric field strength for the given slice along the bipyramid's length. White scale bars show 10 nm.

As discussed above, the bipyramid is an interesting shape of nanoparticle to achieve strong coupling, because the electric field enhancement is largely confined to the tips. Therefore, the mode volume or the resonator does not increase for larger sizes (as is demonstrated in in the full-width-half-maxima in figure 2), as is true for many other nanoparticles. This means that the number of excitons coupled into the system does not increase for larger bipyramids. This demonstrates an entirely new mechanism by which an increased coupling strength can be achieved when the size of a nanoparticle is increased [5].

In conclusion, it has been demonstrated how the size and aspect ratio of a bipyramid can determine the coupling strength within the strong coupling system. This effect of increasing the coupling strength with larger particles has never previously been seen with particles of different shapes without increasing the number of excitons coupled into the system. This emphasises the practicality and novelty of bipyramids in a strongly coupled system.

References

1. E. Cao, W. Lin, M. Sun, W. Liang, and Y. Song, "Exciton-plasmon coupling interactions: from principle to applications," *Nanophotonics*, 7(1), 145-167 (2018)
2. A. Demetriadou, J. M. Hamm, Y. Luo, J. B. Pendry, J. J. Baumberg, and O. Hess. "Spatiotemporal Dynamics and Control of Strong Coupling in Plasmonic Nanocavities," *ACS Photonics*, 4(10), 2410-2418 (2017)
3. M. Stührenberg, B. Munkhbat, D. G. Baranov, J. Cuadra, A. B. Yankovich, T. J. Antosiewicz, E. Olsson, and T. Shegai, "Strong Light-Matter Coupling between Plasmons in Individual Gold Bipyramids and Excitons in Mono- and Multilayer WSe_2," *Nano Letters*, 18(9), 5938-5945 (2018)
4. P. Törmä and W. L. Barnes, "Strong coupling between surface plasmon polaritons and emitters," *Rep. Prog. Phys.*, 78(1), 013901 (2015)
5. J. Lawless, C. Hrelescu, C. Elliott, L. Peters, N. McEvoy, and A. L. Bradley, "Influence of Gold Nano-Bipyramid Dimensions on Strong Coupling with Excitons of Monolayer MoS_2," *ACS Applied Materials & Interfaces*, 12(41), 46406-46415 (2020)