

Optimizing Fluorescence of Diamond Color Centers Encapsulated Into Core-shell Nano-resonators

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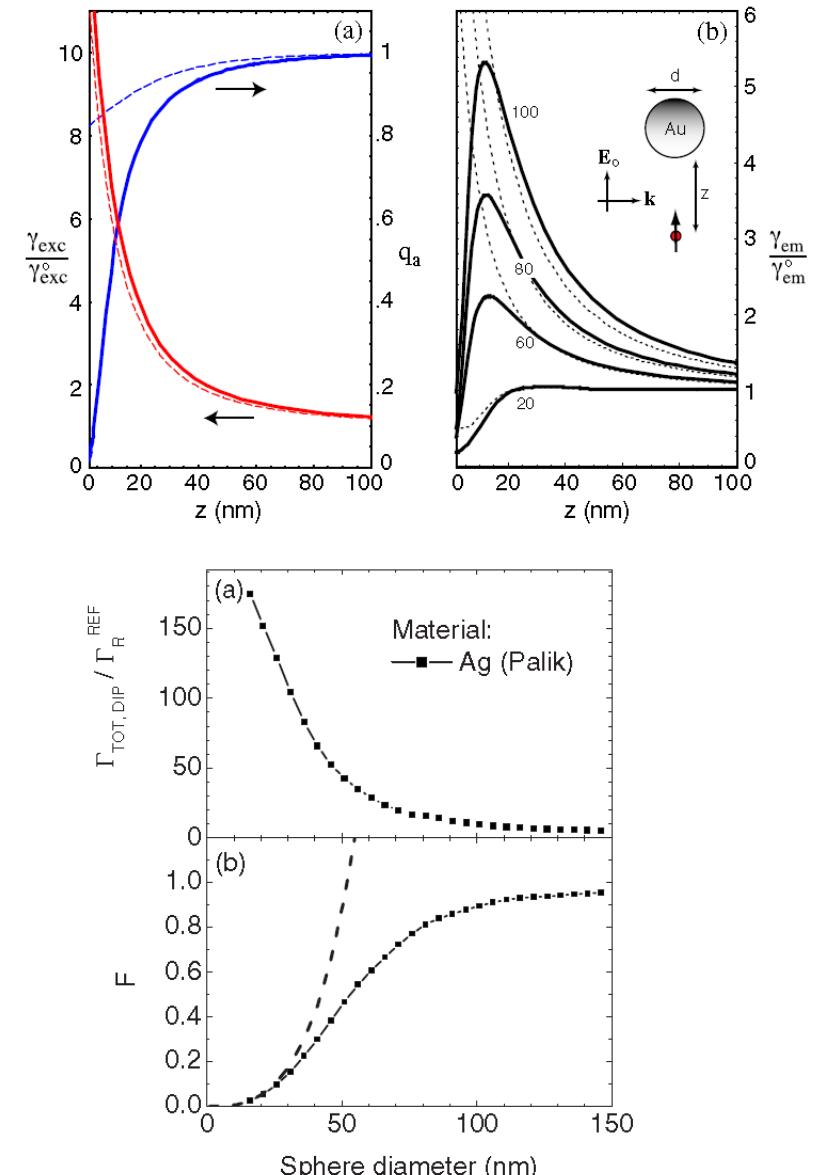


Dr. Mária Csete
COMSOL Conference
Boston, 2016

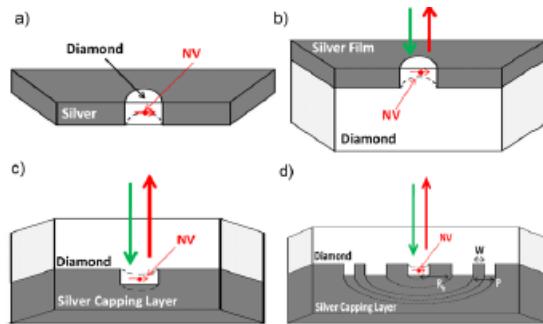
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Plasmonic enhancement of light emission

- Modification of fluorescence intensity, decay rate
 - E. M. Purcell et. al.: Phys Rev 69 (194) 37
- Trade-off between Purcell factor and QE
 - P. C. Das et. al.: Phys. Rev. B 65 (2002) 155416
 - P. Anger et. al.: Phys. Rev. Lett. 96 (2006)
- Intrinsic limits defined by losses
 - F. Wang et al.: Phys. Rev. Lett. 97 (2006) 206806
 - H. Mertens et al.: Phys. Rev. B 76 (2007) 115123



Enhancement of diamond color centers

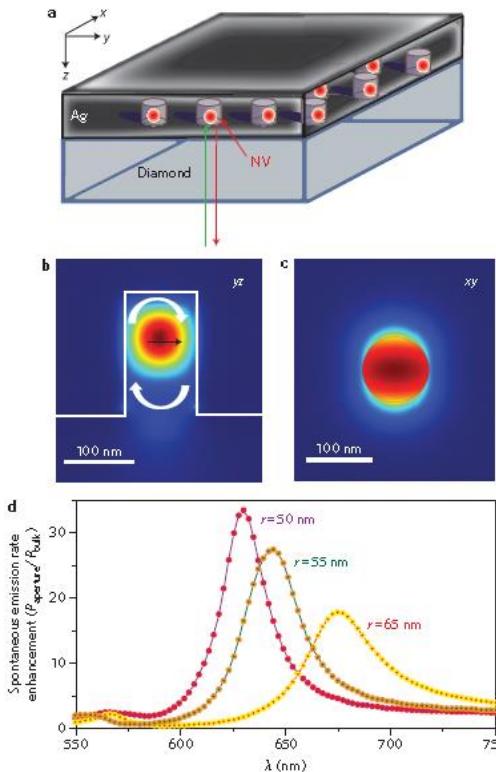


Nano-cavities and apertures to enhance emission

- I. Bulu, T. Babinec, B. Hausmann, J. T. Choy,
II. and M. Loncar, Optics Express 19 (6), 5268-5276 (2011)

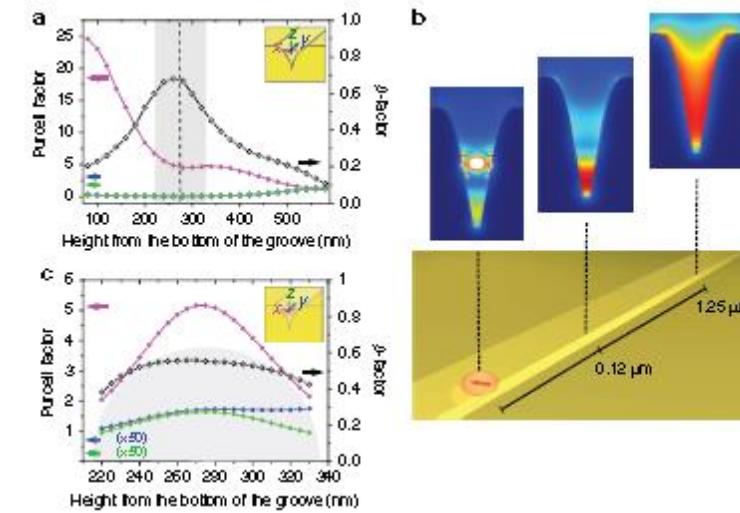
Array of nano-cavities to enhance emission and out-coupling

J. T. Choy, B. J. M. Hausmann, T. M. Babinec, Bulu, M. Khan, P. Maletinsky, A. Yacoby and M. Loncar, Nature Photonics 5, 738-743 (2011)



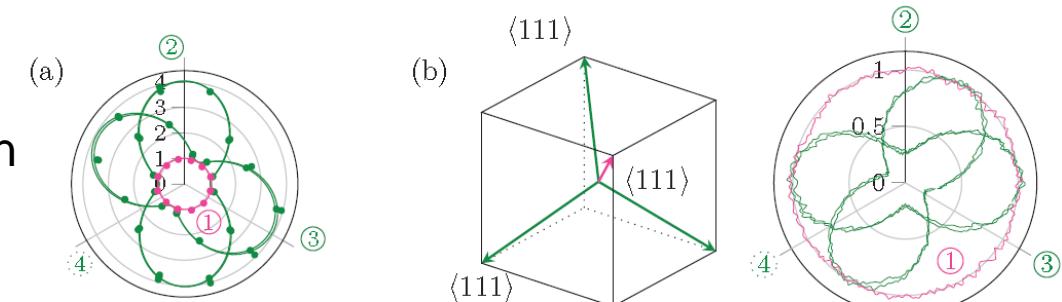
Perpendicularity of dipoles corresponding to excitation and emission

L. J. Rogers, K. D. Jahnke, M. W. Doherty, A. Dietrich, L. P. McGuinness, C. Müller, T. Teraji, H. Sumiya, J. Isoya, N. B. Manson, and F. Jelezko, Phys. Rev. B 89, 235101 (2014)



Positioning close to nano-channels

E. Bermúdez-Ureña, C. González-Ballesteros, M. Geiselmann, R. Marty, P. Radko, T. Holmgård, Y. Alaverdyan, E. Moreno, F. J. García-Vidal, S. I. Bozhevolyi and R. Quidant, Nature Communications 6, 7883



Principles to maximize radiative rate via spectral engineering

- **Excitation enhancement:** $\gamma^{excitation} / \gamma_0^{excitation} = |p \cdot E| / |p_0 \cdot E_0|$
- According to reciprocity radiative rate enhancement at excitation:
- **Emission enhancement**, total decay rate enhancement -> **Purcell factor**

$$F^{total} = \frac{\gamma_{uncoupledemitter}^{radiative} + \gamma_{coupledresonator}^{radiative} + \gamma_{emitterintrinsic}^{non-radiative} + \gamma_{coupledresonator}^{non-radiative} + \gamma_{quenching}^{non-radiative}}{\gamma_{uncoupledemitter,0}^{radiative} + \gamma_{emitterintrinsic,0}^{non-radiative}}$$

$$\left. \frac{\gamma^{radiative}}{\gamma_0^{radiative}} \right|_{emission} = \left. \frac{QE}{QE_0} \right|_{emission} \cdot \left. \frac{\gamma^{radiative}}{\gamma_0^{radiative}} \right|_{excitation} .$$

- Quantum efficiency

$$QE \Big|_{\gamma_{emitterintrinsic}^{non-radiative}=0, \gamma_{quenching}^{non-radiative}=0} = \frac{\gamma_{uncoupledemitter}^{radiative} + \gamma_{coupledresonator}^{radiative}}{\gamma_{uncoupledemitter}^{radiative} + \gamma_{coupledresonator}^{radiative} + \gamma_{coupledresonator}^{non-radiative}}$$

$$QE \Big|_{read-out} = \frac{P^{radiative}}{P^{radiative} + P^{non-radiative}}$$

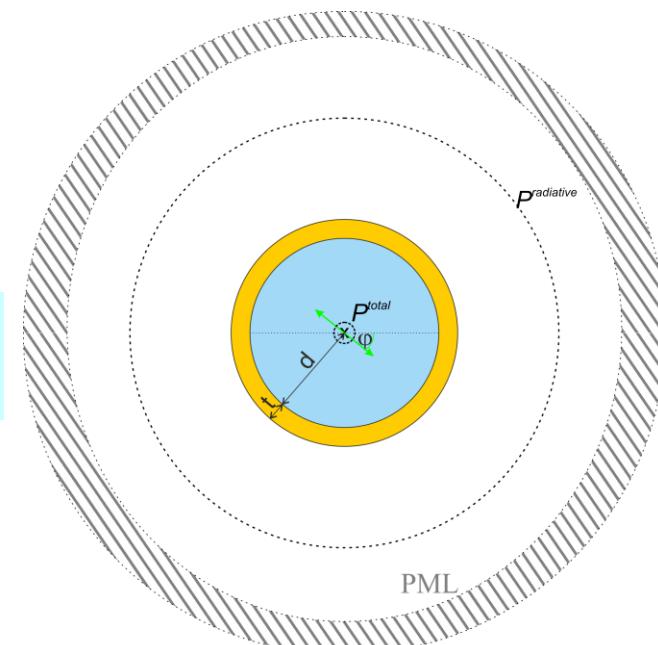
- Product of Purcell*QE -> radiative rate enhancement

$$F^{total} \cdot QE = \frac{\frac{\gamma_{uncoupledemitter}^{radiative} + \gamma_{coupledresonator}^{radiative}}{\gamma_{uncoupledemitter,0}^{radiative}}}{1 + \frac{1-QE_0}{QE_0}} = \frac{F^{radiative}}{1 + \frac{1-QE_0}{QE_0}}$$

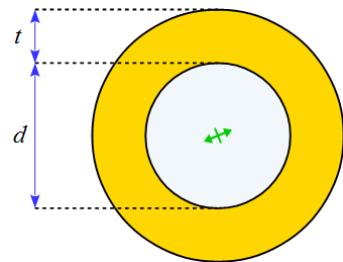
$$F^{radiative} \Big|_{read-out} = Purcell \cdot QE \Big|_{read-out} = \frac{P^{radiative}}{P_0^{radiative}}$$

- Corrected QE of the emitter

$$QE^{corrected} = \frac{P^{radiative}}{P^{radiative} + P^{non-radiative} + \frac{1-QE_0}{QE_0}}$$



Implementation into numerical methods & GLOBAL



Tuning dipole position and orientation

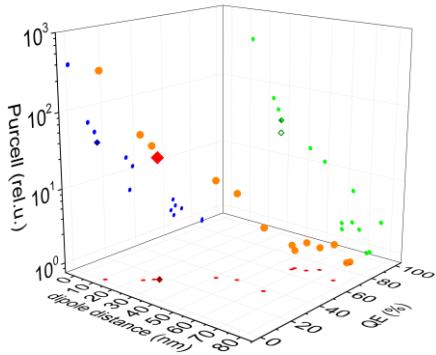
Conditional optimization

QE / Purcell factor optimization by stepping criterion regarding the Purcell factor / QE

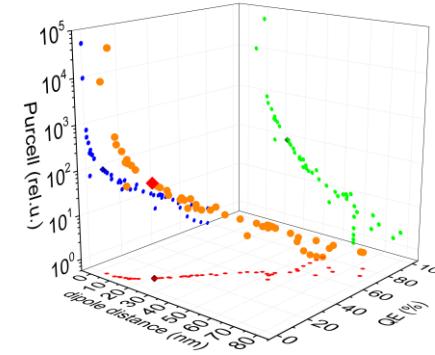
GLOBAL implemented via LiveLink for MATLAB: Sampling (Monte Carlo), Clustering (Single-link), Local searching (UNIRANDI, Random walk, BFGS)

T. Csendes et al.: The GLOBAL Optimization Method Revisited, Optimization Letters 2(2008) 445-454

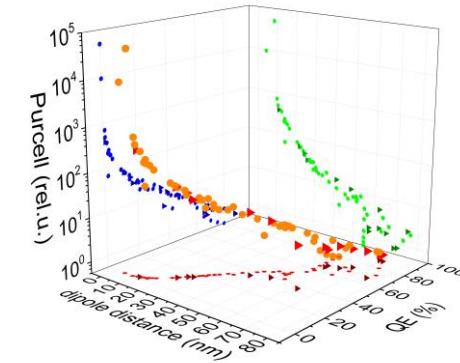
QE maximization with Purcell criterion



Purcell maximization with QE criterion



Integrated parameter cube

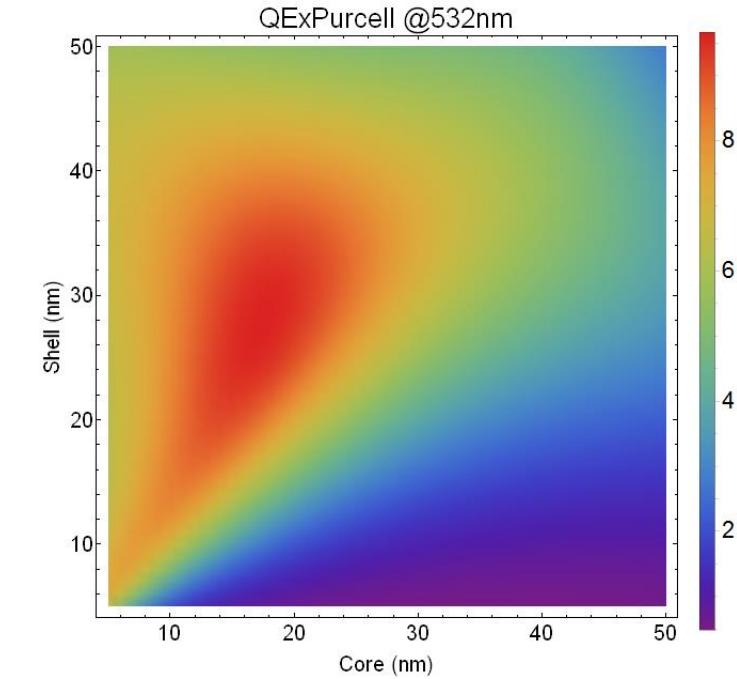
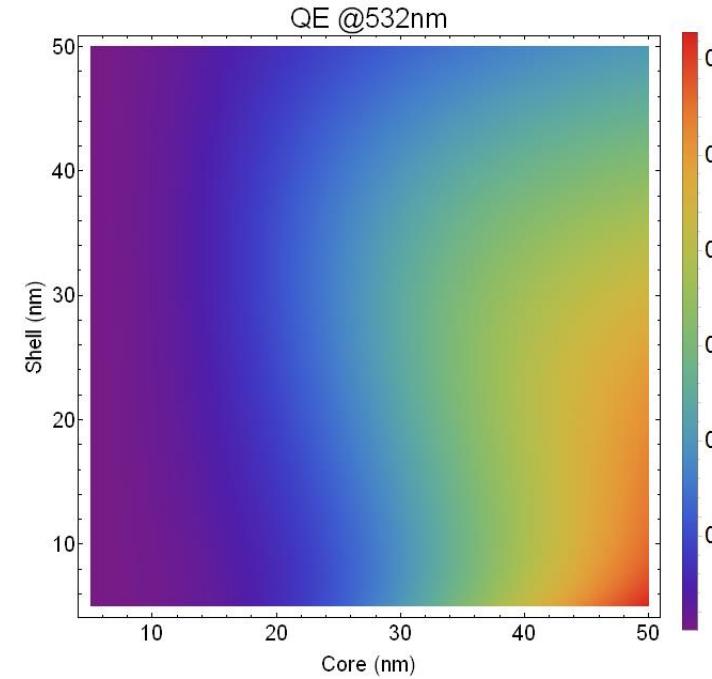
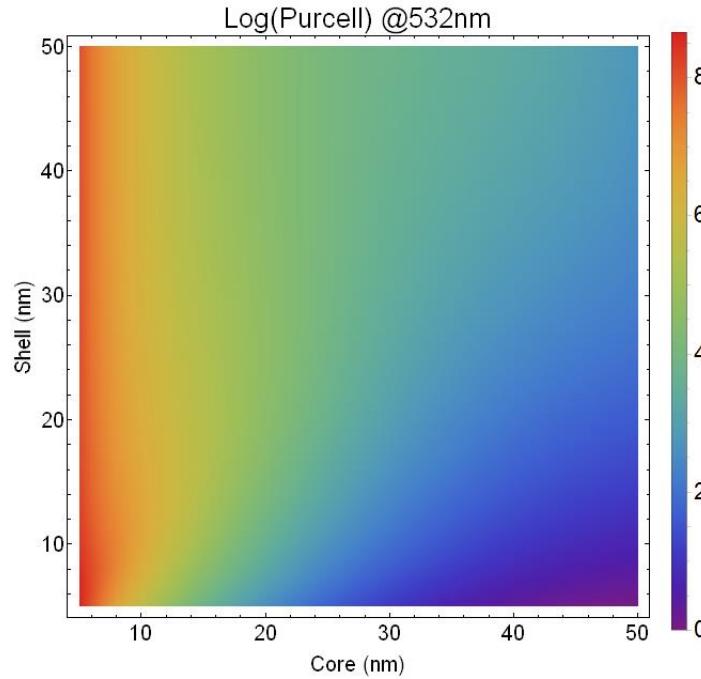


Extraction of wavelength dependent optical response
-radiated and absorbed power =>
-Purcell factor, QE
-radiative rate: Purcell*QE

Inspection of near-field at extrema
Enorm,
powerflow,
charge distribution

Different metals
Centralized dipole
De-centralized dipole
Different criteria
Different objective function

Parametric sweep before optimization



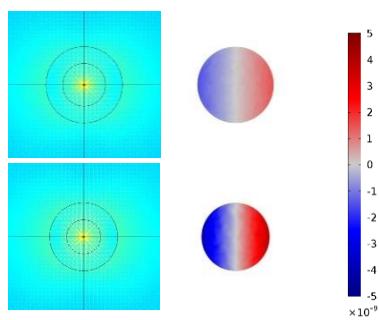
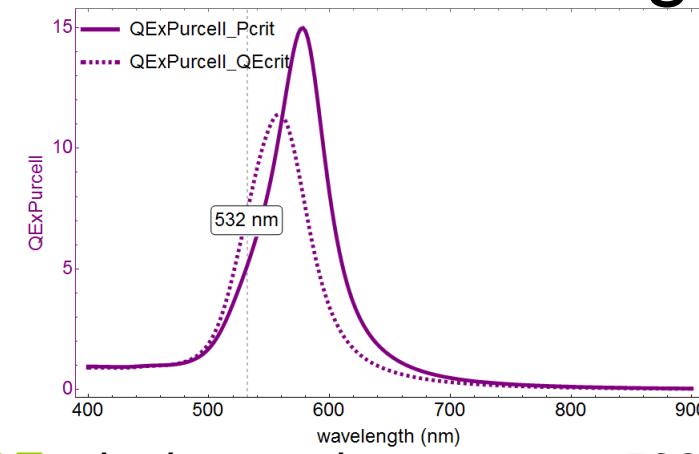
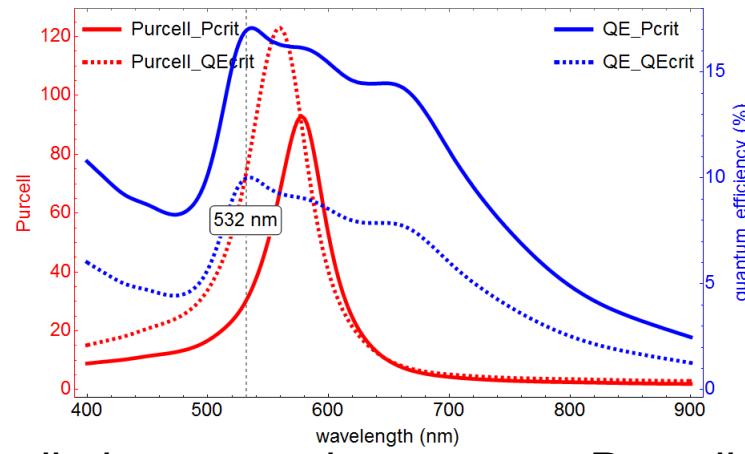
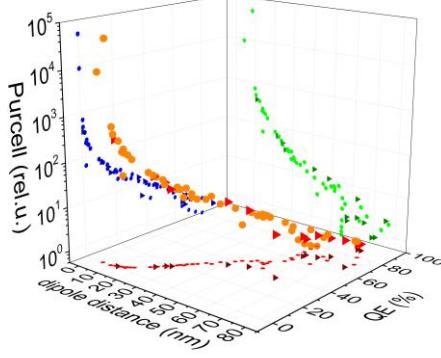
small cores are advantageous
to achieve large total decay rate
independently of shell thickness

large cores with thin shells
are advantageous
to improve the QE

an optimal core-shell
combination exist

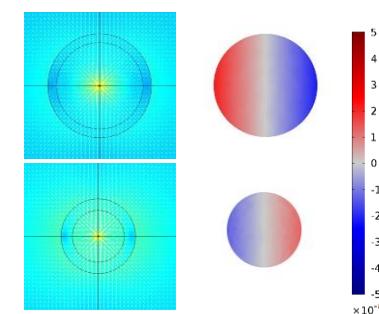
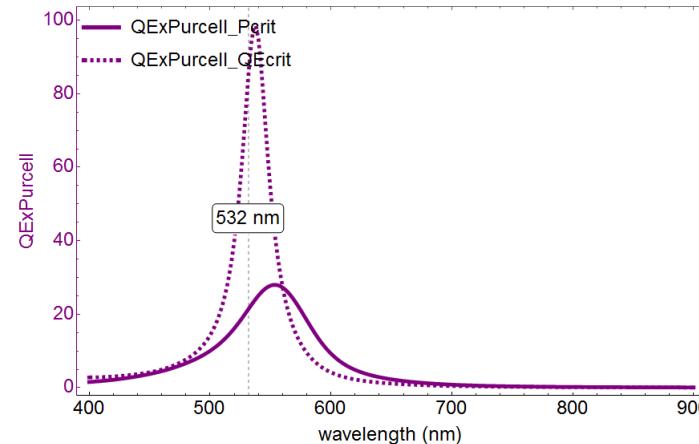
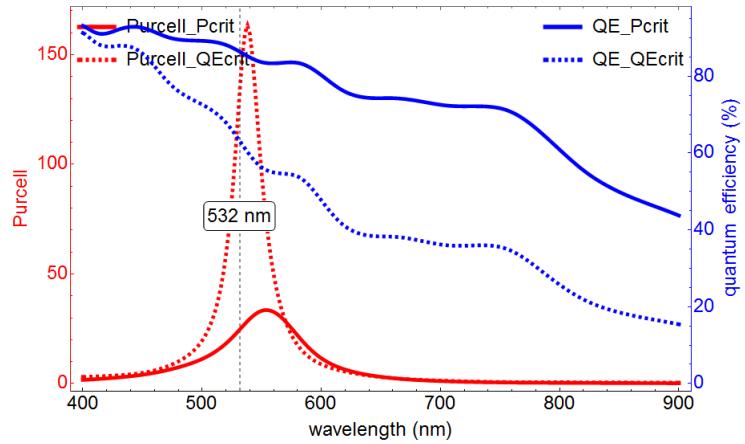
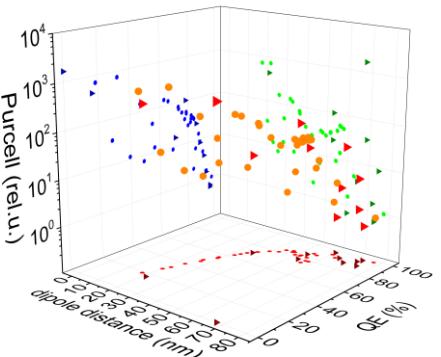
NV and SiV color center excitation enhancement: Au/Ag core-shell

Integrated parameter cube



~10/10 radiative rate enhancement, Purcell / **QE** criterion: peaks at $\lambda \gg / > 532$ nm
QE criterion: radiative rate larger, detuning smaller

Integrated parameter cube



$\sim 10 / 100$ radiative rate enhancement, Purcell / **QE** criterion: peak at $\lambda > / \sim 532$ nm
QE criterion: radiative rate larger, detuning smaller

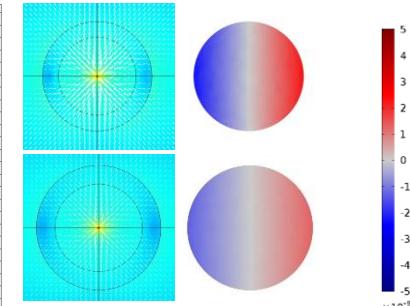
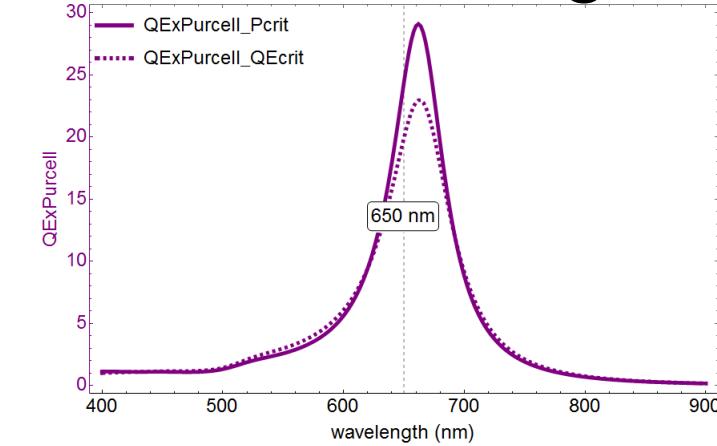
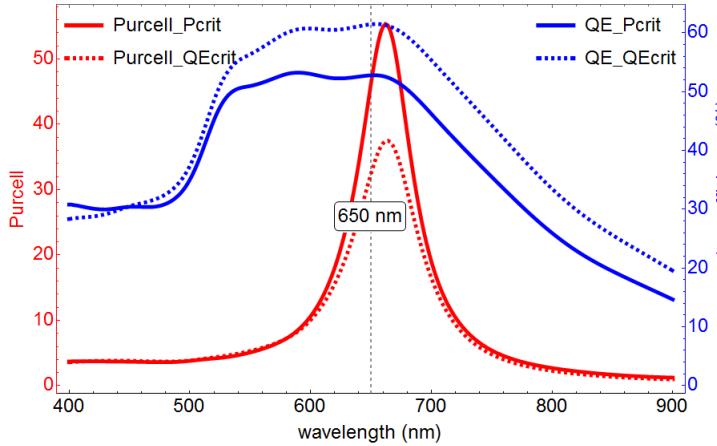
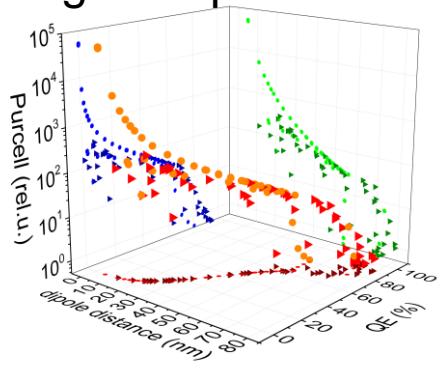
silver better:

larger QE and radiative rate, smaller detuning
larger core, larger distance

	criterion	P	Q(%)	QxP	$\Delta\lambda(\text{nm})$	2r(nm)	t(nm)	r/t	d(nm)	$\varphi(^{\circ})$	
Purcell	10	24.93	86.32	21.52	22	105.66	25.66	2.06	52.83	0.00	
	Ag/Au	0.82	5.08	4.16	0.48	2.01	1.25	1.61	2.01		
	QE	50	136.36	63.06	85.98	6	63.63	14.02	2.27	31.81	0.00
	Ag/Au	1.84	6.30	11.58	0.23	1.52	0.67	2.27	1.52		

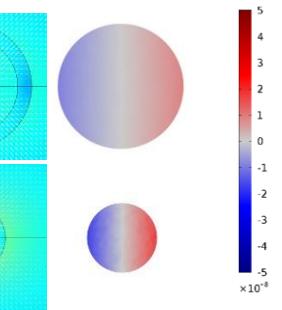
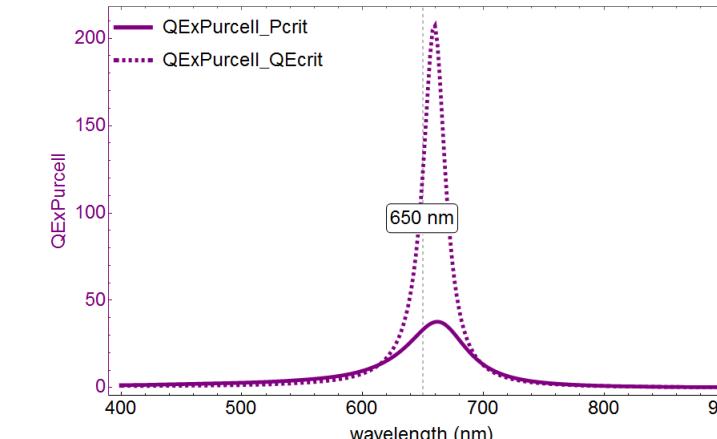
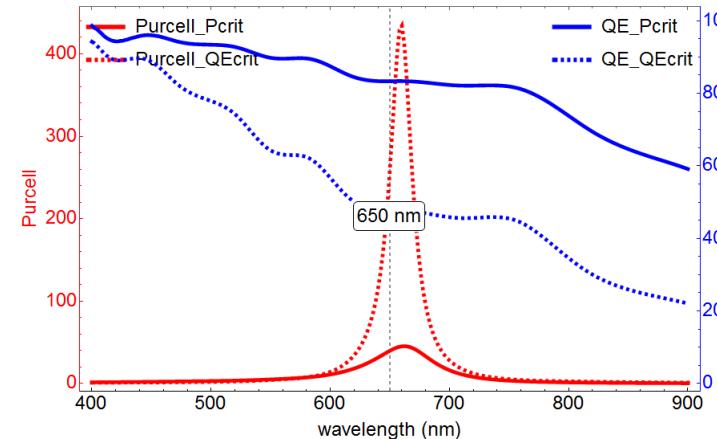
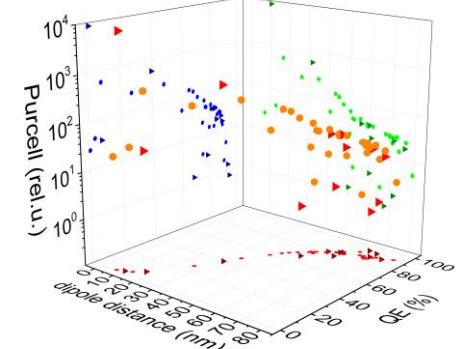
NV color center emission enhancement: Au/Ag core-shell

Integrated parameter cube



$\sim 10 / 10$ radiative rate enhancement, Purcell / QE criterion: peaks at $\lambda > \sim 650$ nm
Purcell larger radiative rate, same detuning

Integrated parameter cube



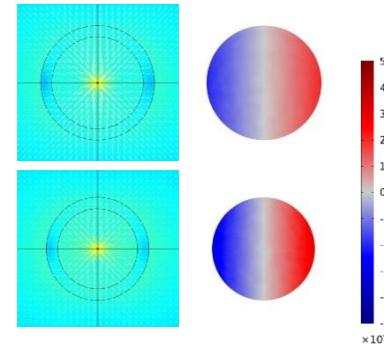
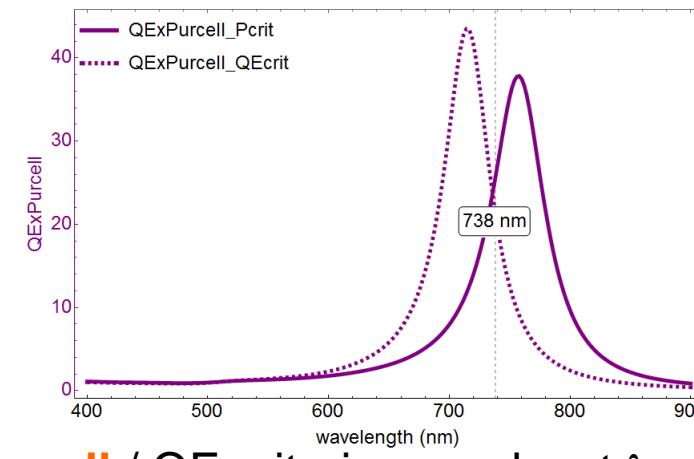
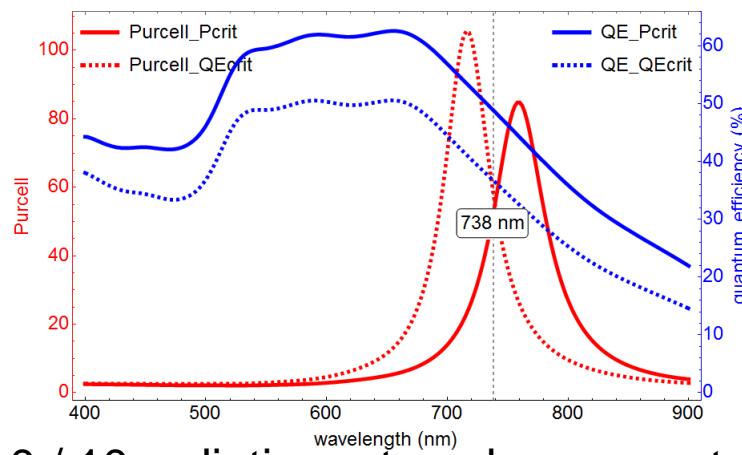
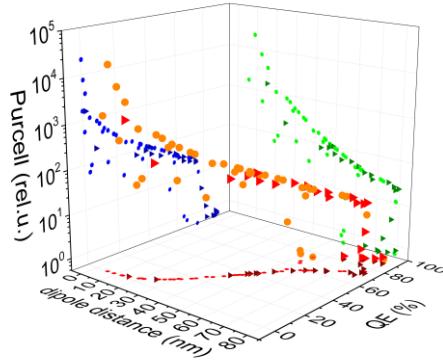
$\sim 10 / 100$ radiative rate enhancement, Purcell / QE criterion: peak at $\lambda > \sim 650$ nm
QE larger radiative rate, smaller detuning

silver better: slightly larger QE and radiative rate,
 same/smaller detuning
 larger/smaller core, larger/smaller distance

	criterion	P	Q(%)	QxP	$\Delta\lambda(\text{nm})$	2r(nm)	t(nm)	r/t	d(nm)	$\varphi(^{\circ})$
Purcell		25	39.73	83.43	33.14	12	119.21	16.96	3.52	59.60
	Ag/Au	0.86	1.58	1.36	1.00	1.25	0.88	1.43	1.25	0.00
QE		20	142.29	64.85	92.28	10	86.13	10.77	4.00	43.06
	Ag/Au	4.40	1.06	4.64	0.83	0.80	0.47	1.70	0.80	0.00

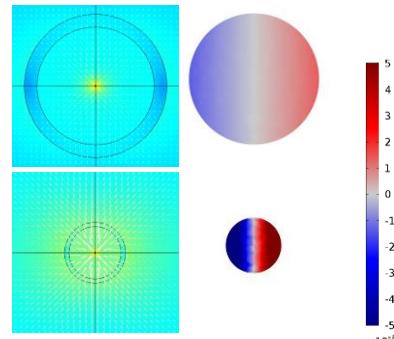
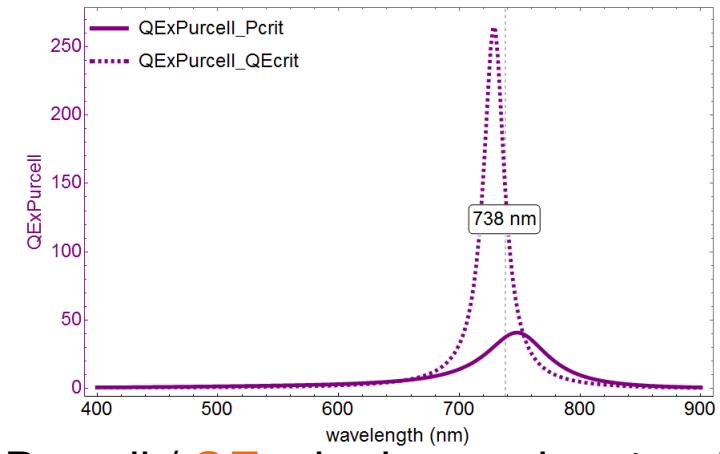
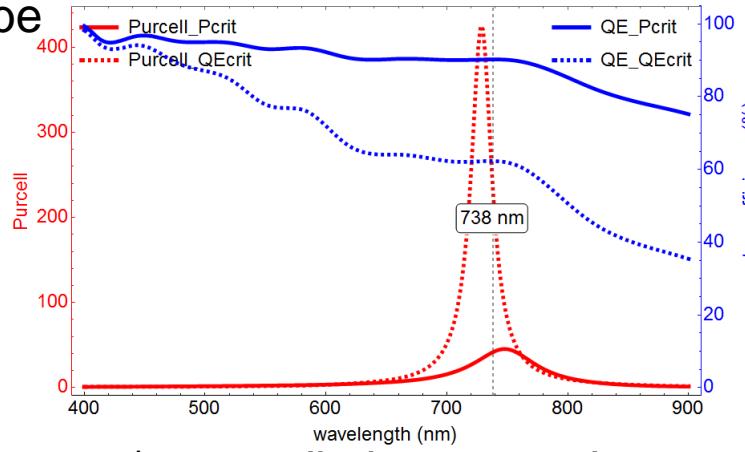
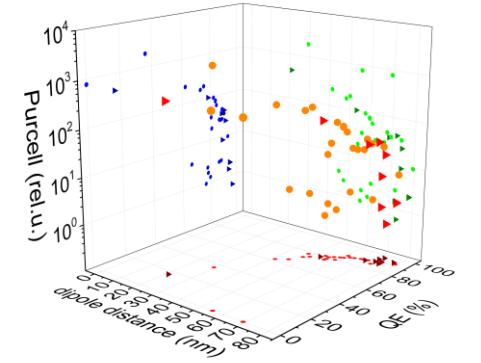
SiV color center emission enhancement: Au/Ag core-shell

Integrated parameter cube



$\sim 10 / 10$ radiative rate enhancement, **Purcell** / **QE** criterion: peaks at $\lambda > / < 738$ nm
Purcell larger radiative rate, smaller detuning

Integrated parameter cube

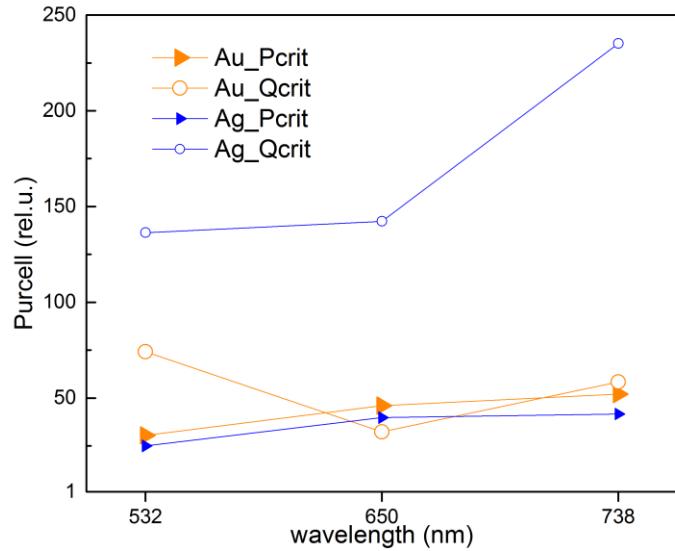


$\sim 10 / 100$ radiative rate enhancement, Purcell / **QE** criterion peak at $\lambda > / \sim 650$ nm
QE larger radiative rate and smaller detuning

silver better: enhanced QE and radiative rate,
smaller detuning
larger/smaller core, larger/smaller distance

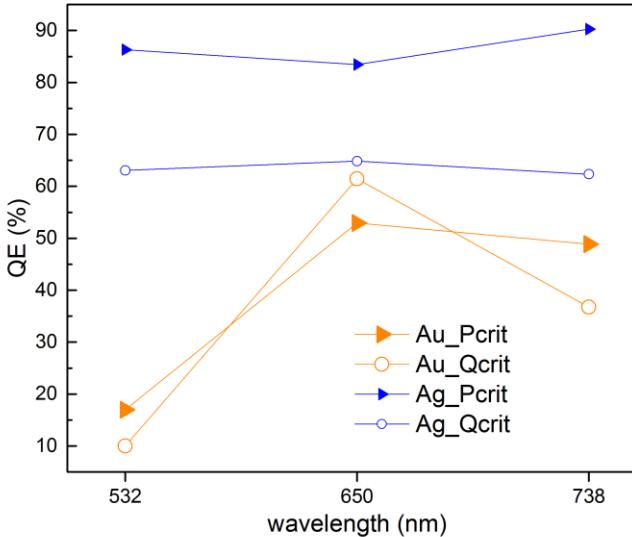
	criterion	P	Q(%)	Qcorr(%)	QxP	$\Delta\lambda(\text{nm})$	2r(nm)	t(nm)	r/t	d(nm)	$\varphi(^{\circ})$
Purcell	10	41.62	90.26	74.21	37.57	10	144.00	15.39	4.68	72.00	0.00
	Ag/Au	0.80	1.85	1.78	1.48	0.50	1.36	1.20	1.13	1.36	
	20	235.12	62.33	60.03	146.54	-2	82.76	7.55	5.48	41.38	0.00
	Ag/Au	4.03	1.70	1.89	6.84	0.09	0.90	0.58	1.56	0.90	

Centralized dipoles: single wavelength tendency



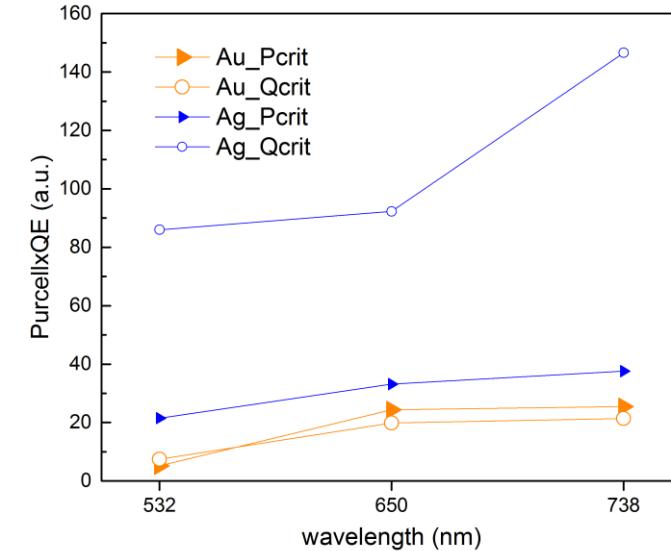
Total decay rate:
increase with wavelength
except in configuration_Au_Q_{crit}

Silver better
except configuration_Ag_P_{crit}



QE:
peaks at 650 nm
except in configuration_Ag_P_{crit}

Silver better

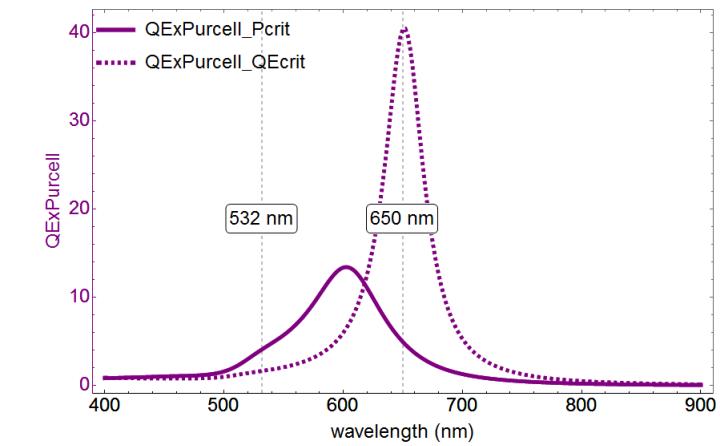
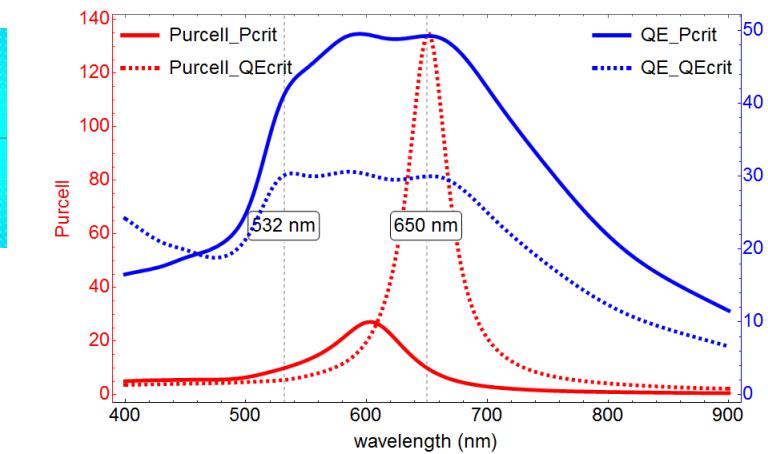
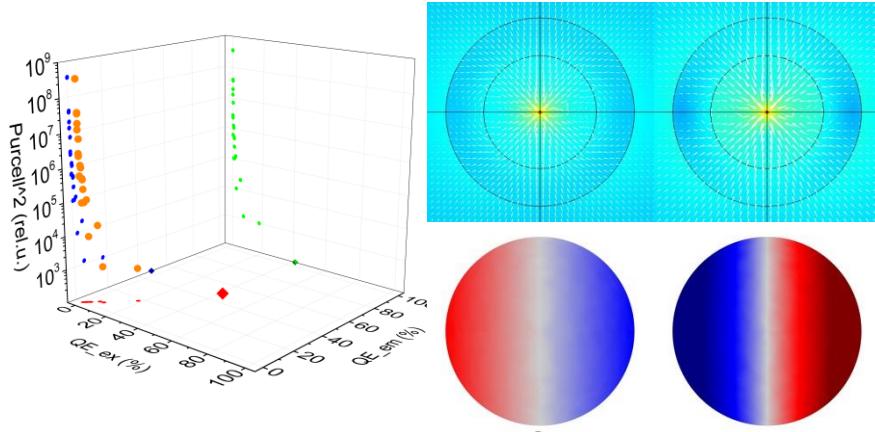


Radiative rate enhancement:
increases with wavelength
according to wavelength dependency
of material limits

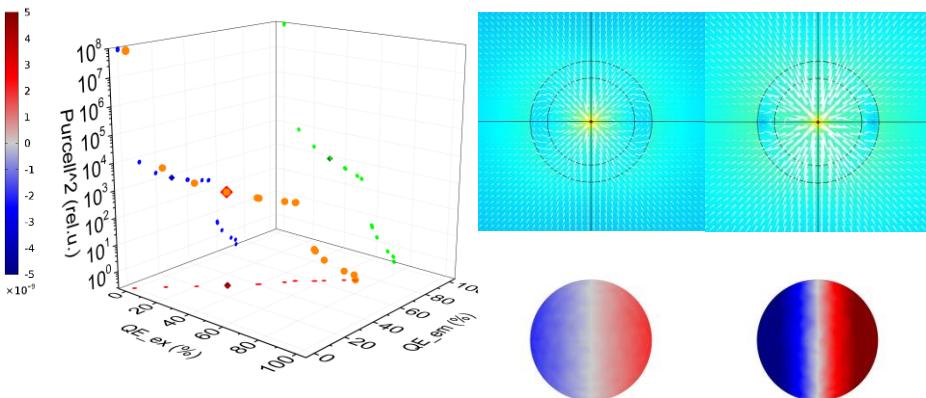
Silver better

NV center simultaneous enhancement via Au

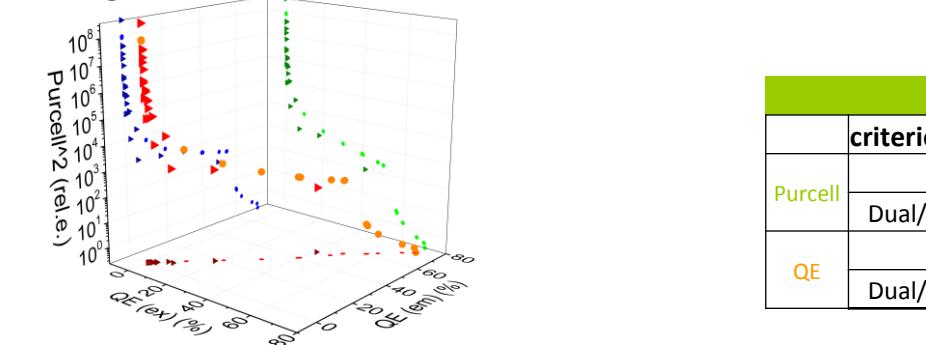
QE maximization with Purcell criterion



Purcell maximization with QE criterion



Integrated parameter cube



Purcell / QE criterion excitation / emission

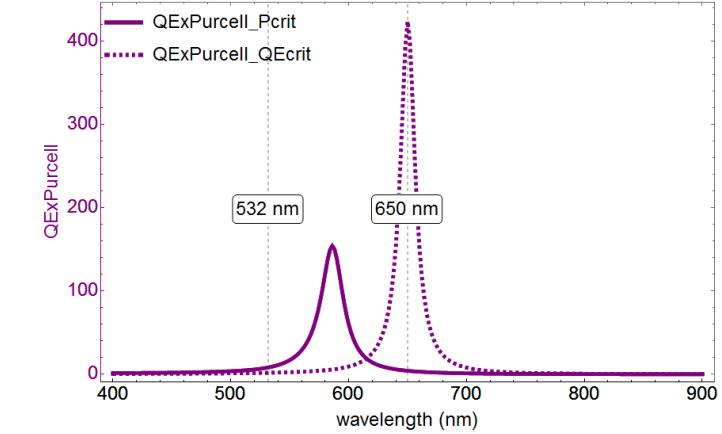
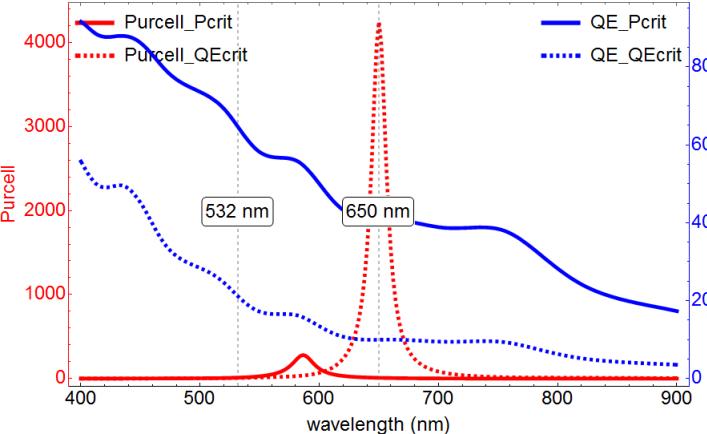
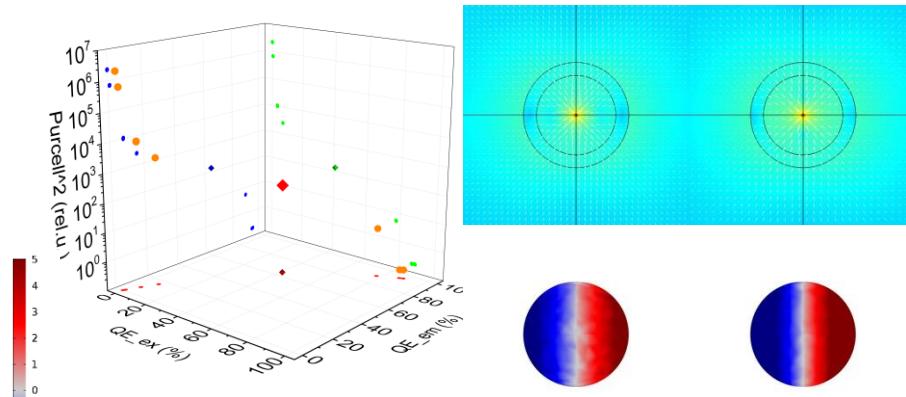
~1 – 10/1-10 excitation - emission enhancement,
single peaks at 532 nm < λ < 650 nm / 650 nm

Dual: both improves QE at excitation
with QE_{crit} better in emission enhancement, smaller detuning

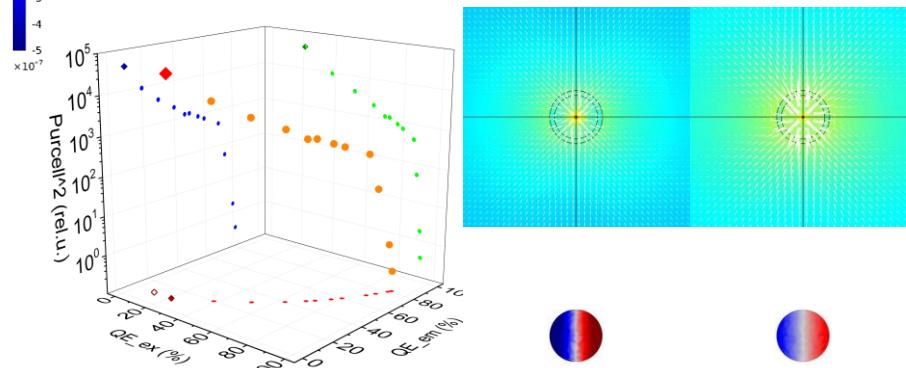
	criterion	excitation				emission				$\Delta\lambda(\text{nm})$	QxP^2	$2r(\text{nm})$	$t(\text{nm})$	r/t	$d(\text{nm})$	$\varphi(\text{°})$	
		P	Q(%)	QxP	$\Delta\lambda(\text{nm})$	criterion	P	Q(%)	QxP								
Purcell		10	10.03	41.29	4.14	70	10	10.00	49.28	4.93	-48	20.42	92.58	30.87	1.50	46.29	0.00
	Dual/Solo	0.33	2.43	0.80	1.52		0.22	0.93	0.20	-4.00							
QE		30	5.57	30.16	1.68	120	30	134.50	30.00	40.35	2	67.75	71.53	14.03	2.55	35.77	0.00
	Dual/Solo	0.07	3.02	0.23	4.62		4.16	0.49	2.03	0.17							

NV center simultaneous enhancement via Ag

QE maximization with Purcell criterion



Purcell maximization with QE criterion

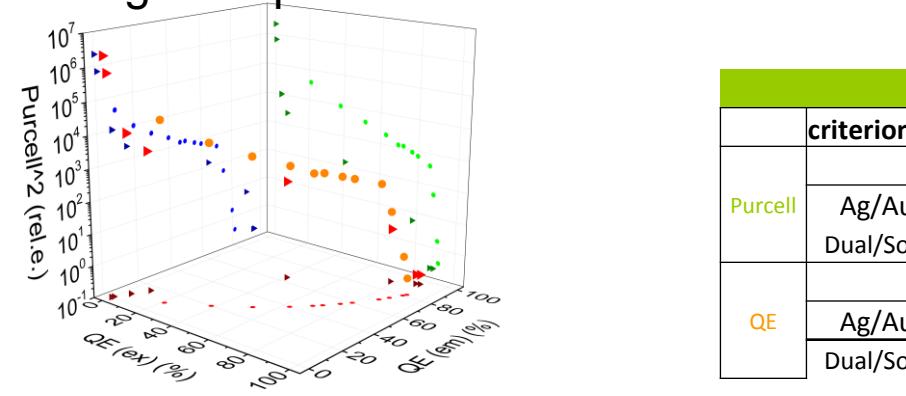


Purcell / QE criterion excitation / emission

~1 - 10/1-100 excitation - emission enhancement
single peaks at $532 \text{ nm} < \lambda < 650 \text{ nm} / 650 \text{ nm}$

silver is better in excitation and emission enhancement
smaller core with thinner shell, smaller dipole distance

Integrated parameter cube

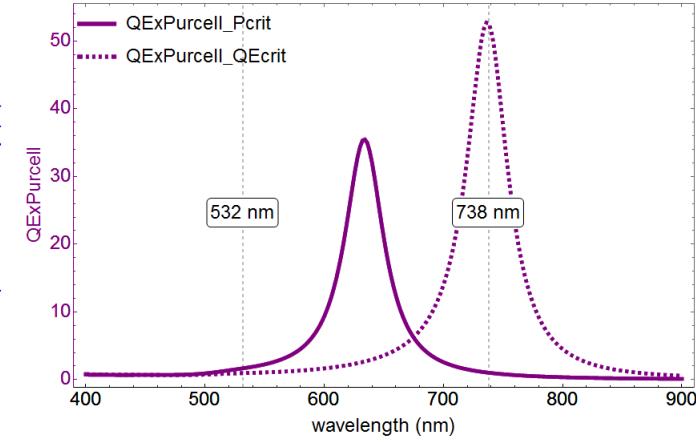
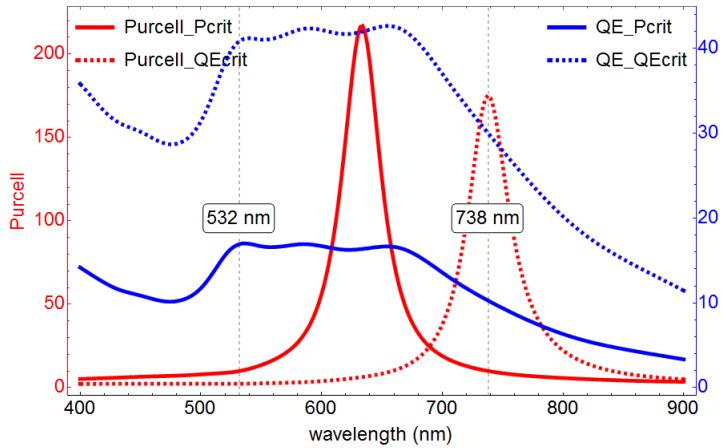
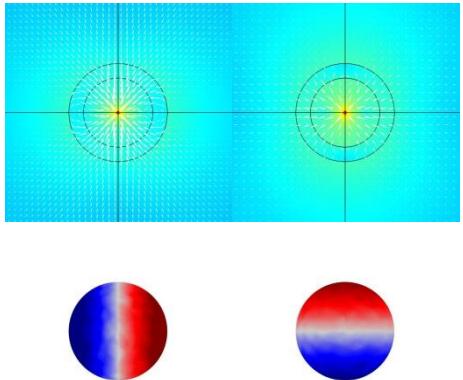
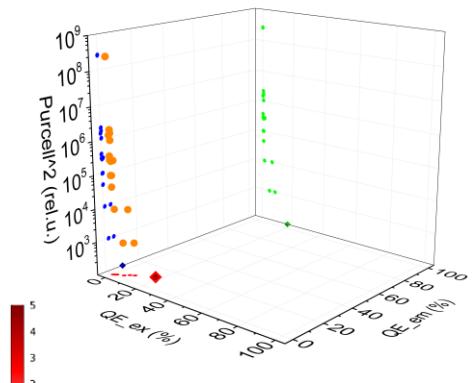


Dual: at excitation all quantities are smaller
with QE_{crit} better in emission enhancement, smaller detuning

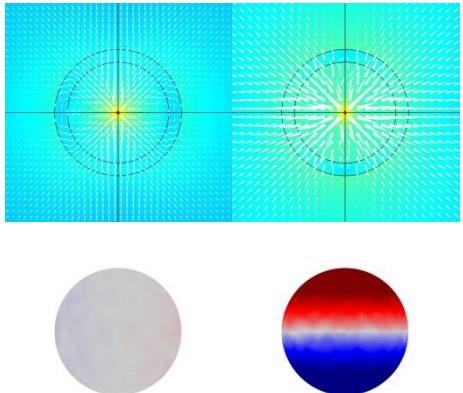
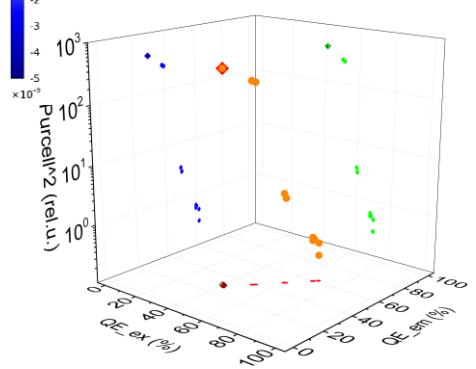
	criterion	excitation					emission					2r(nm)	t(nm)	r/t	d(nm)	$\varphi(^{\circ})$	
		P	Q(%)	QxP	$\Delta\lambda(\text{nm})$	criterion	P	Q(%)	QxP	$\Delta\lambda(\text{nm})$	QxP 2						
Purcell	10	10.56	40.80	4.31	54		10	13.02	64.49	8.40	-64	36.17	65.04	10.63	3.06	32.52	
	Ag/Au	1.05	0.99	1.04	0.77		1.30	1.31	1.70	1.33	1.77						
	Dual/Solo	0.42	0.47	0.20	2.45		0.33	0.77	0.25	-5.33							
QE	10	9.09	21.09	1.92	118		10	4.25E+3	10.00	424.54	0	814.08	35.51	3.92	4.53	17.75	0.00
	Ag/Au	1.63	0.70	1.14	0.98		31.56	0.33	10.52	0.00	12.02						
	Dual/Solo	0.07	0.33	0.02	19.67		29.84	0.15	4.60	0.00							

SiV center simultaneous enhancement via Au

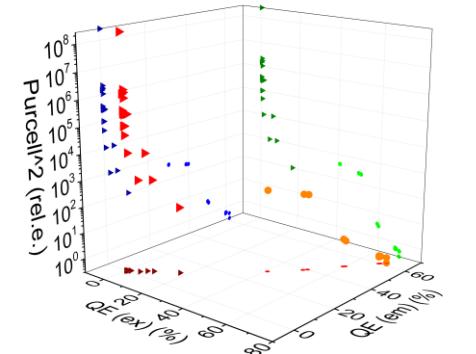
QE maximization with Purcell criterion



Purcell maximization with QE criterion



Integrated parameter cube



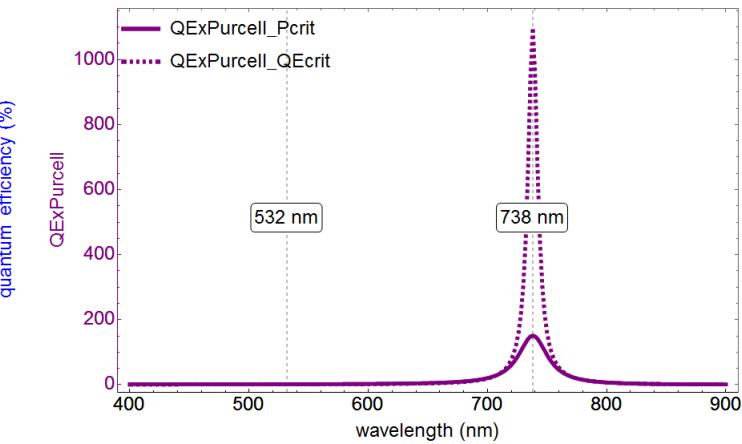
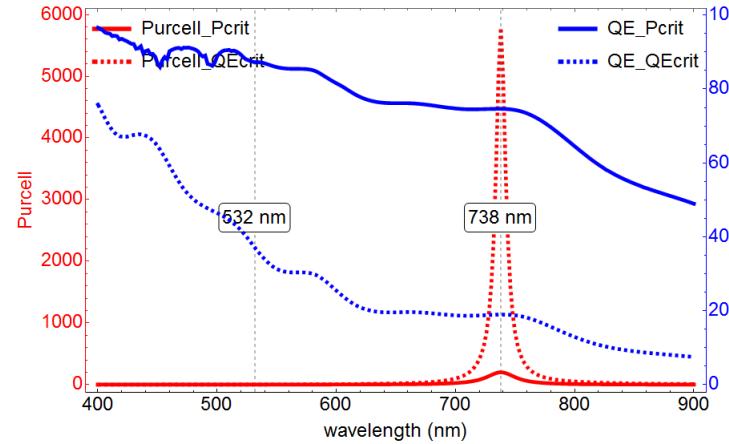
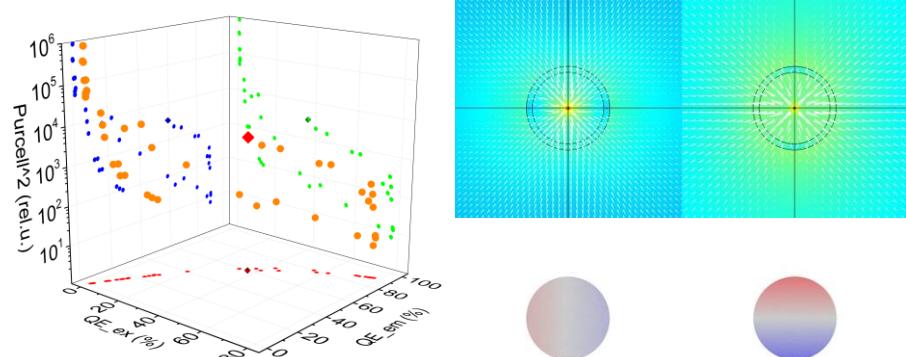
Purcell / QE criterion excitation / emission
~1-1/1-100 excitation-emission enhancement
single peaks at 532 nm < λ < 738 nm / 738 nm

Dual: both QE improves at excitation
with QE_{crit} better in emission enhancement, smaller detuning

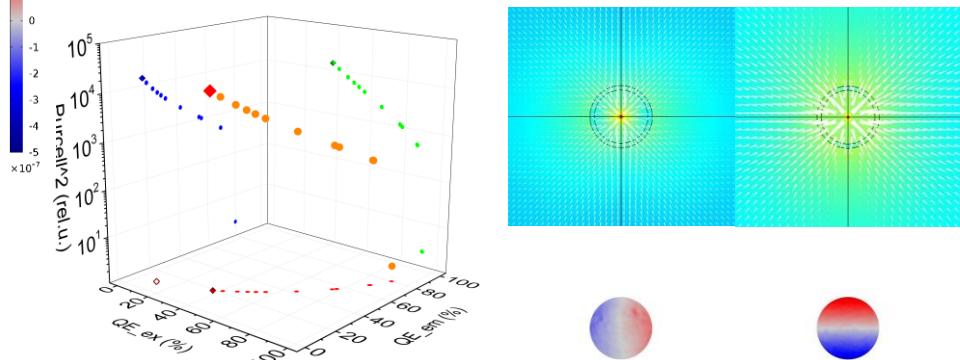
	excitation					emission												
	criterion	P	Q(%)	QxP	Δλ(nm)	criterion	P	Q(%)	Qcorr (%)	QxP	Δλ(nm)	QxP ²	2r(nm)	t(nm)	r/t	d(nm)	φ(°)	
Purcell		10	10.00	16.93	1.69	102	10	10.01	10.27	5.41	1.03	-104	1.74	56.80	11.96	2.38	28.40	0.00
	Dual/Solo	0.33	1.00	0.33	2.22			0.19	0.21	0.13	0.04	-5.20						
QE		40	2.38	40.87	0.97	204	30	175.09	30.01	28.54	52.55	-2	51.11	83.26	10.28	4.05	41.63	0.00
	Dual/Solo	0.03	4.09	0.13	7.85			3.00	0.82	0.90	2.45	0.09						

SiV center simultaneous enhancement via Ag

QE maximization with Purcell criterion



Purcell maximization with QE criterion

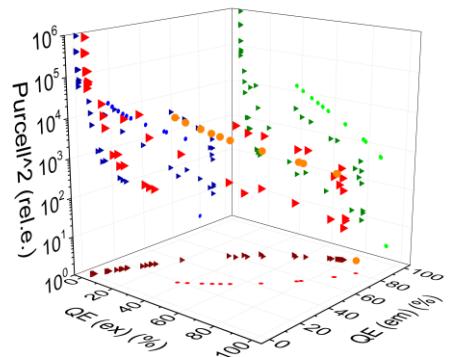


Purcell / QE criterion excitation / emission no detuning
 ~1-100/1-1000 excitation-emission enhancement
 single peaks at 738 nm

silver is better in emission with P_{crit} and QE_{crit}

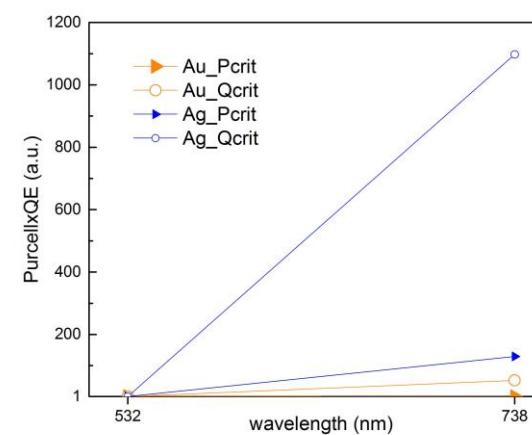
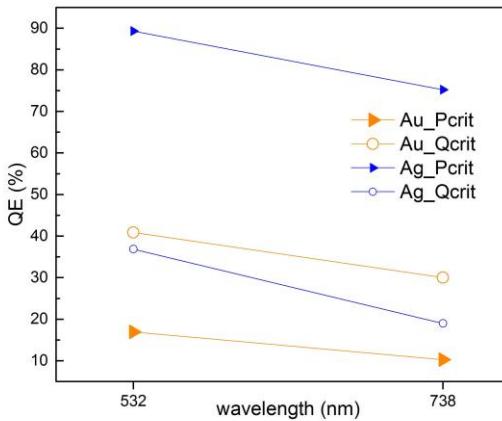
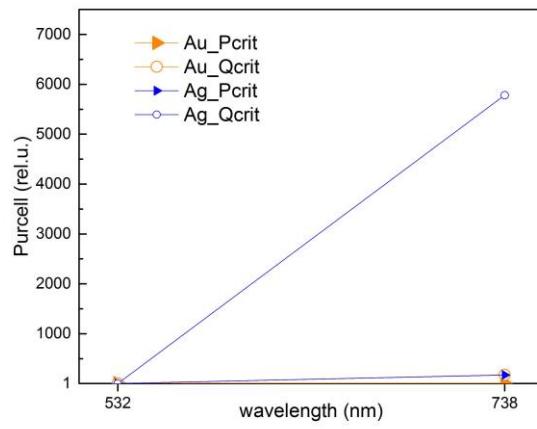
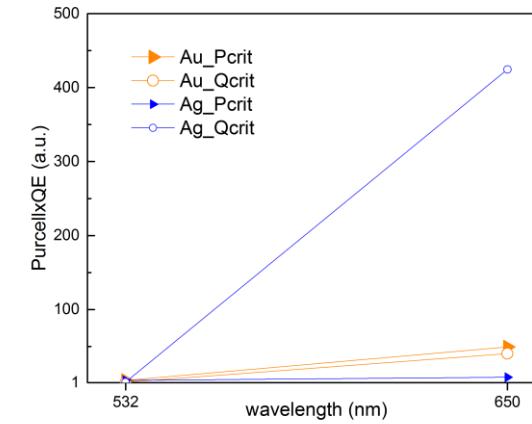
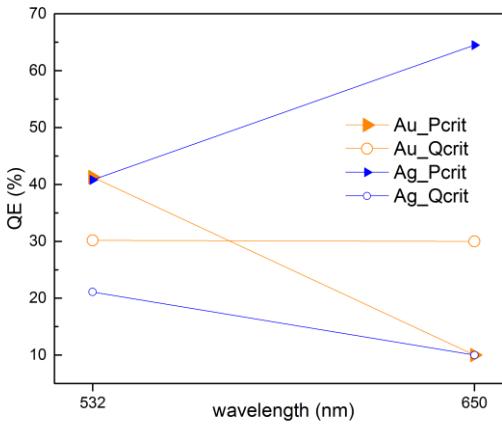
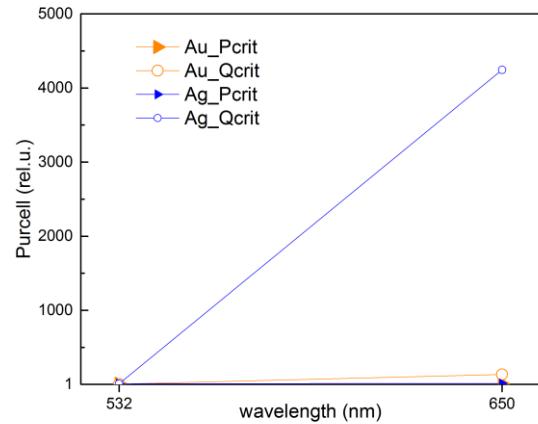
Dual: better in emission enhancement with P_{crit} and QE_{crit}

Integrated parameter cube



excitation								emission									
criterion	P	Q(%)	QxP	$\Delta\lambda(\text{nm})$	criterion	P	Q(%)	Qcorr (%)	QxP	$\Delta\lambda(\text{nm})$	QxP^2	2r(nm)	t(nm)	r/t	d(nm)	$\varphi(^{\circ})$	
Purcell	1	1.64	89.31	1.46	206	1	172.41	75.22	69.51	129.68	0	189.94	99.31	8.98	5.53	49.66	0.00
	Ag/Au	0.16	5.27	0.86	2.02		17.22	7.32	12.85	126.10	0.00	109.03					
	Dual/Solo	0.07	1.03	0.07	9.36		4.14	0.83	0.94	3.45	0.00						
QE	10	2.44	36.85	0.90	206	10	5.78E+3	18.98	18.95	1.10E+3	0	985.59	43.99	3.56	6.18	21.99	0.00
	Ag/Au	1.02	0.90	0.92	1.01		33.02	0.63	0.66	20.89	0.00	19.28					
	Dual/Solo	0.02	0.58	0.01	34.33		24.59	0.30	0.32	7.49	0.00						

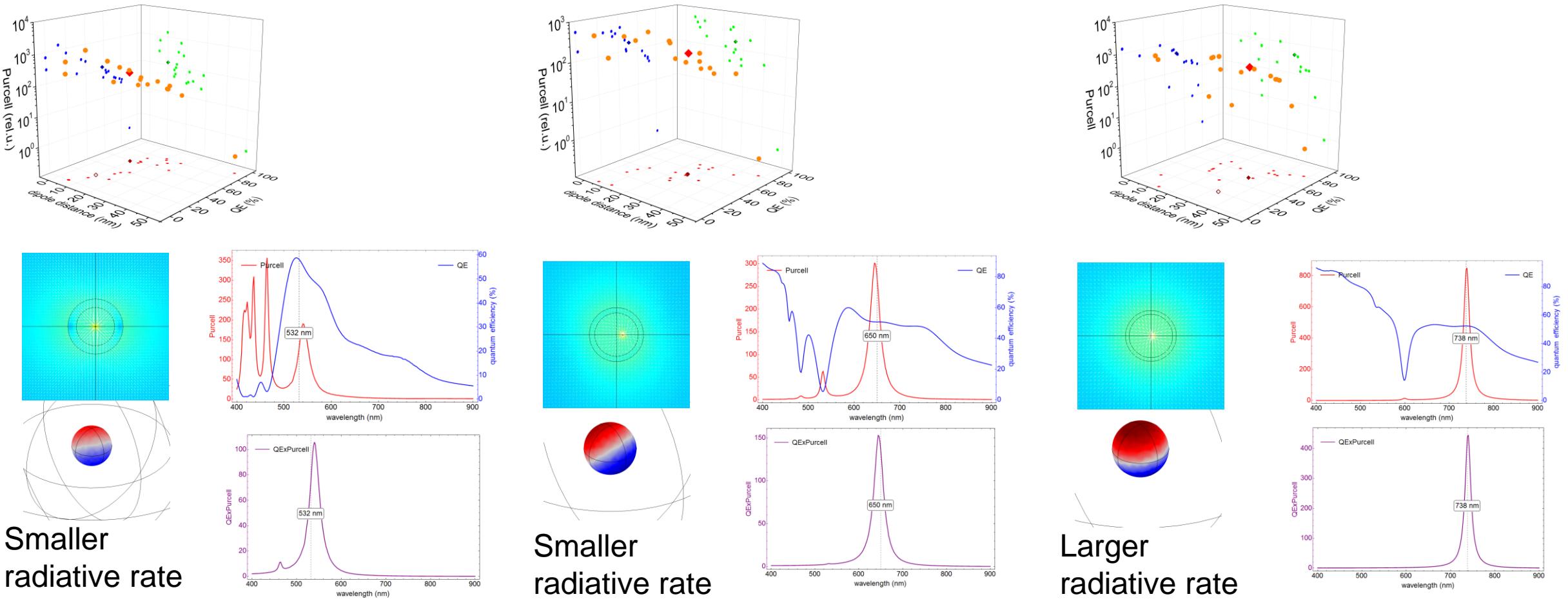
Centralized dipole: dual wavelength tendency



Larger number of artificial constraints =>
Discrepancies with respect to the expectations based on wavelength and material dependent limits
Radiative rate in accordance / contradiction with material limits in case of QE_{crit} / P_{crit}

NV and SiV color center excitation/emission enhancement, Ag

Single wavelengths, Purcell factor maximization with QE criterion; **de-centralized dipole**



Smaller
radiative rate

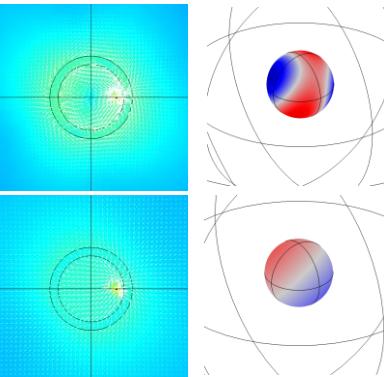
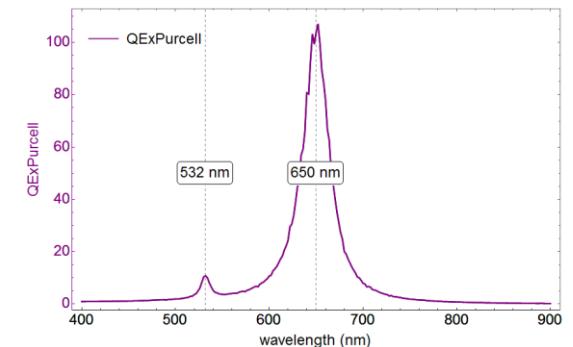
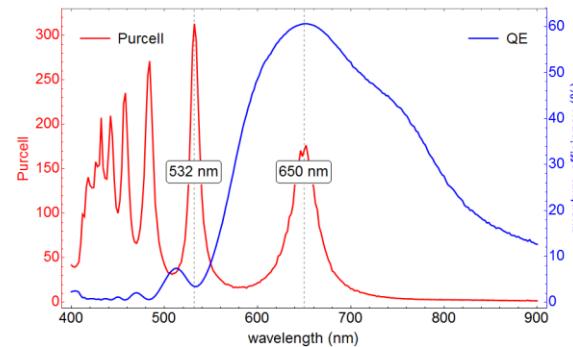
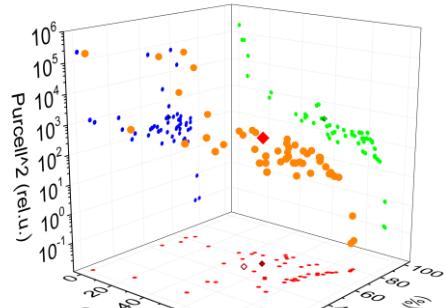
Smaller
radiative rate

Larger
radiative rate

	criterion (%)	P	Q(%)	Qcorr (%)	QxP	$\Delta\lambda(\text{nm})$	2r(nm)	t(nm)	r/t	d(nm)	$\theta (\circ)$	$\varphi (\circ)$
532		20	124.51	58.24		72.52	8	61.84	13.17	2.35	9.97	34.42
	shifted/central		0.91	0.92		0.84	1.33	0.97	0.94	1.03	0.31	
650		50	158.88	50.77		80.66	-6	72.27	8.99	4.02	25.98	39.82
	shifted/central		1.12	0.78		0.87	-0.60	0.84	0.83	1.01	0.60	
738		20	426.05	52.48	51.39	223.58	4	72.32	6.16	5.87	32.64	10.16
	shifted/central		1.81	0.84	0.86	1.53	-2.00	0.44	0.82	1.07	0.79	

NV and SiIV color center excitation & emission enhancement, Ag

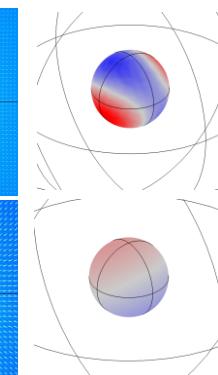
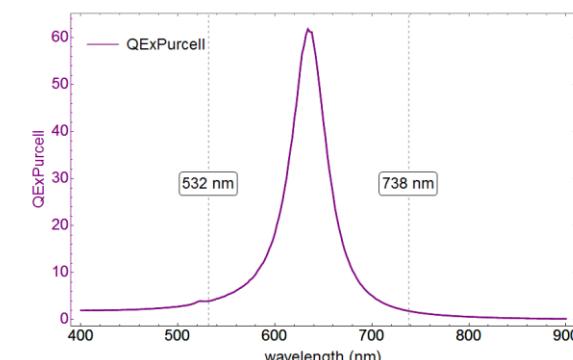
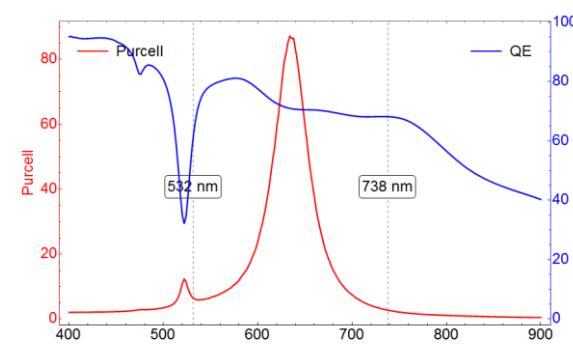
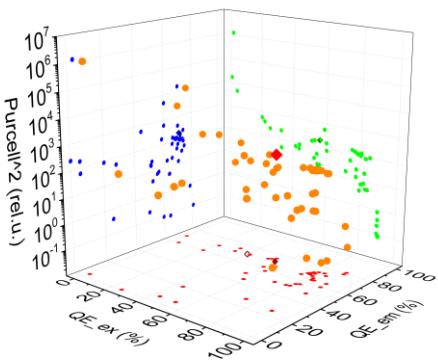
Dual wavelengths, Purcell maximization with QE criterion; **de-centralized dipole**



NV

Capable of resulting in enhancement at both wavelengths, better/weaker excitation/emission via de-centralized dipole

criterion (%)	excitation				emission				QxP^2	2r(nm)	t(nm)	r/t	d(nm)	θ (°)	φ (°)		
	P	Q(%)	QxP	$\Delta\lambda$ (nm)	criterion	P	Q(%)	QxP	$\Delta\lambda$ (nm)								
0	283.71	3.51	9.94	0	0	40	203.56	60.61	123.38	2	1226.92	83.10	10.57	3.93	9.52	47.48	123.04
shifted/central	31.20	0.17	5.19	0.00			0.05	6.06	0.29		1.51						



SiIV

Capable of resulting in enhancement at both wavelengths, better/weaker excitation/emission via de-centralized dipole

criterion (%)	excitation				emission				QxP^2	2r(nm)	t(nm)	r/t	d(nm)	θ (°)	φ (°)		
	P	Q(%)	QxP	$\Delta\lambda$ (nm)	criterion	P	Q(%)	Qcorr (%)	QxP	$\Delta\lambda$ (nm)							
40	3.18	0.64	2.04	-8	60	1.47	68.18	9.59	1.00	-104	2.05	94.09	13.51	3.48	40.47	33.31	15.75
shifted/central	1.31	0.02	2.28	-0.04		2.55E-4	3.59	0.51	9.15E-04	-	2.08E-3						

NV and SiV color center excitation & emission enhancement, Ag

Dual wavelength, Purcell² maximization, **de-centralized dipole**, corrected QE criterion

WRT non-corrected

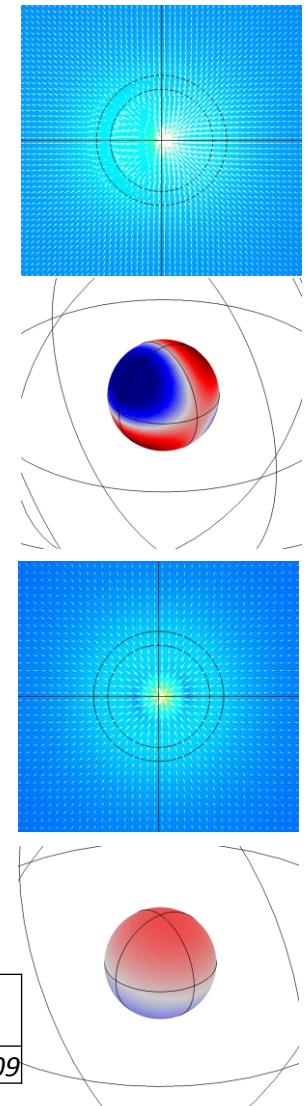
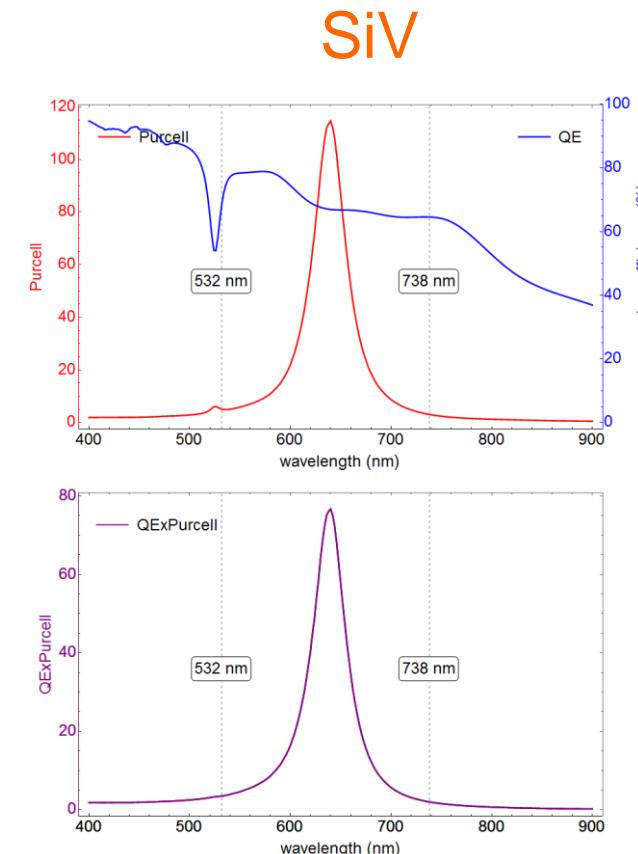
Excitation: better QE, smaller Purcell*QE, larger detuning

Emission: better Purcell, Purcell*QE, smaller detuning

WRT to central

Excitation: better QE, larger Purcell*QE, smaller detuning

Emission: better QE



criterion (%)	excitation				emission													
	P	Q(%)	QxP	$\Delta\lambda(\text{nm})$	QEcorr criterion (%)	P	Q(%)	Qcorr (%)	QxP	$\Delta\lambda(\text{nm})$	QxP^2	2r(nm)	t(nm)	r/t	d(nm)	$\theta (\circ)$	$\varphi (\circ)$	
70	2.38	71.42	1.70	108	correction applied	10	1.72	64.82	10.42	1.12	-98	1.90	89.27	12.11	3.69	41.44	12.73	190.09
	0.75	111.22	0.83	-13.50			1.17	0.95	1.09	1.11	0.94	0.92						
	0.97	1.94	1.89	0.52			2.98E-04	3.41	0.55	1.02E-03	-	0.00						

NV and SiV color center excitation & emission enhancement, Ag

Dual wavelength, QExPurcell² maximization, de-centralized dipole, corrected QE criterion

WRT non-corrected

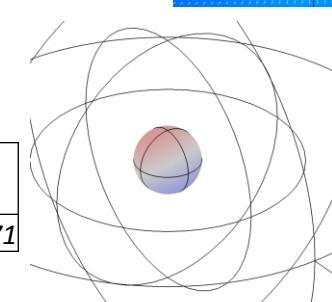
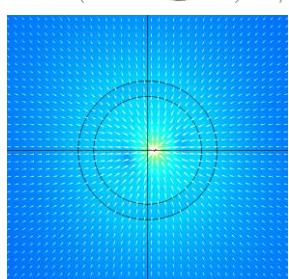
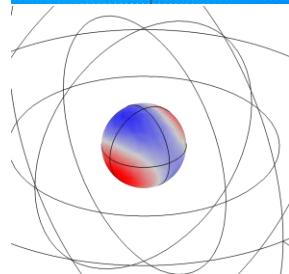
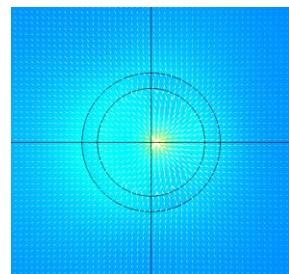
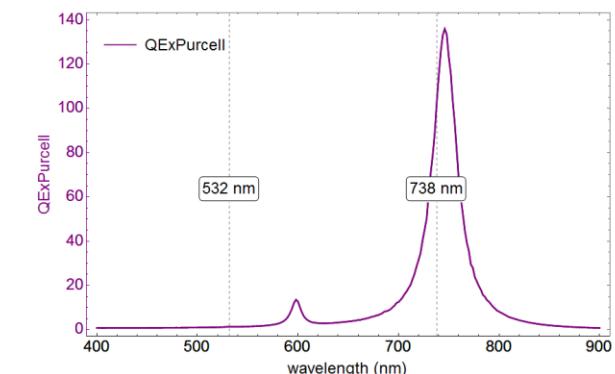
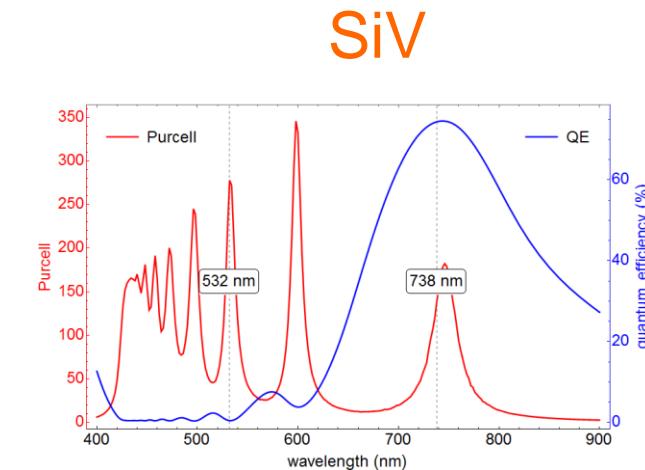
Excitation: better QE, smaller Purcell*QE, larger detuning

Emission: better Purcell, QE, Purcell*QE, smaller detuning

WRT to central

Excitation: better Purcell, larger Purcell*QE, smaller detuning

Emission: better QE



excitation					emission													
criterion (%)	P	Q(%)	QxP	Δλ(nm)	QEcorr criterion (%)	P	Q(%)	Qcorr (%)	QxP	Δλ(nm)	QxP^2	2r(nm)	t(nm)	r/t	d(nm)	θ (°)	φ (°)	
0	167.31	0.70	1.17	66	10	230.64	74.30	71.51	171.37	8	200.35	99.66	8.99	5.54	10.67	70.38	148.71	
corr/non-corr	52.56	1.09	0.57	-8.25		156.59	1.09	7.46	170.64	-0.08	97.61							
corr/central	68.65	0.02	1.30	0.32		0.04	3.91	3.77	0.16		0.20							

Conclusions on core-shell type nano-resonators

- Single wavelength optimization
 - Purcell*QE radiative rate enhancement increases with the wavelength
 - Radiative rate enhancement is smaller than 10^3
 - Optimization results in better QE and Purcell*QE enhancement in Ag-based core-shells
- Dual wavelength optimization
 - Purcell*QE enhancement is larger at the emission wavelength except configuration_Au_SiV_Purcell
 - In optimized configurations_532-650/738 the radiative rate enhancement smaller/larger than 10^3
 - Material dependence of radiative rate at the excitation and emission
 - slightly and more considerably larger in configuration_Ag_Purcell&QE for NV
 - slightly smaller and significantly larger in configuration_Ag_Purcell&QE for SiV
 - Dual with respect to single wavelength optimization
 - larger QE at the excitation in Au based core-shells in case of 532-650 and 532-738
 - larger Purcell*QE at the emission, smaller detuning both in Au and Ag-based configuration_Au/Ag_QE
- De-centralized with respect to centralized
 - Larger Purcell * QE at 738 nm single wavelength, and at the excitation in both cases of NV and SiV
- De-centralized with corrected QE criterion optimized for P^2 and Purcell*QE 2
 - smaller/larger excitation rate than in case of de-centralized /centralized, optimized with QE
 - larger/smaller emission rate than in case of de-centralized /centralized, optimized with QE.

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Prof. Tibor Csendes



Gábor Szabó

A. Szenes, B. Bánhelyi, L. Zs. Szabó, G. Szabó, T. Csendes, M. Csete: "*Enhancing diamond color center fluorescence via optimized plasmonic nanorod configuration*", Plasmonics, DOI: 10.1007/s11468-016-0384-1