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Optoelectronics of Nanocomposites

Mahi R. Singh
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Western University

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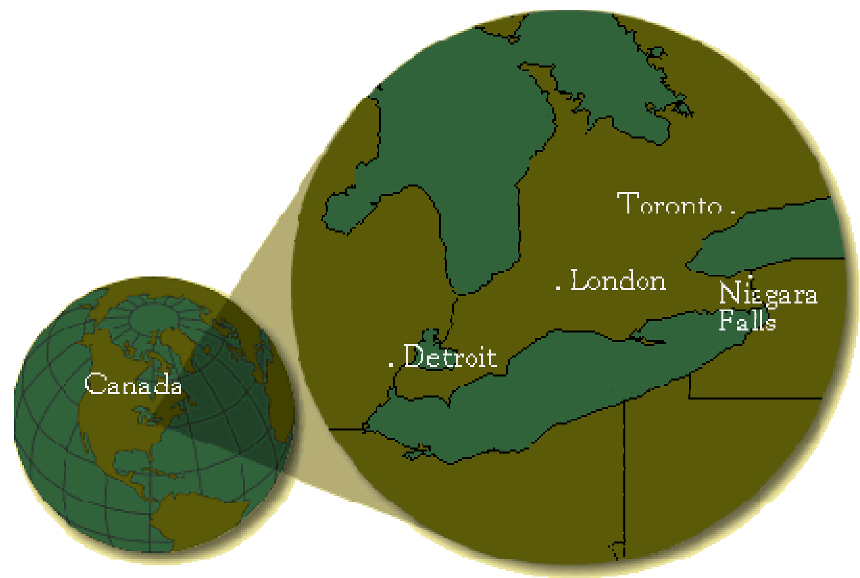
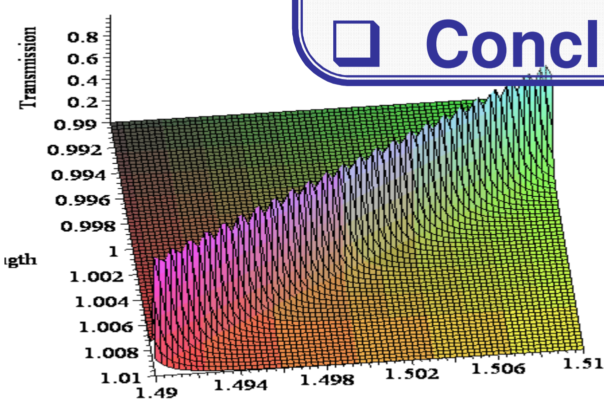


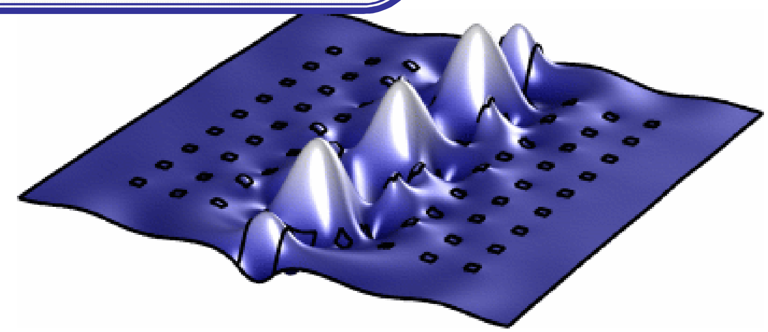
optics2014

Niagara Falls

Outline

- ❑ **Hybrid Metallic Nanomaterials**
- ❑ **Metallic nanomaterials: plasmonics**
  - Nobel metals : Gold, Silver
  - Graphene
  - Metamaterials
- ❑ **Photonic materials: Photonics**
  - Metamaterials
  - Photonic crystals
- ❑ **One- and two-photon switching**
- ❑ **sensors**
- ❑ **Photoluminescence enhancement**
- ❑ **Conclusion**





Collaborators

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C. Racknor

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M. Brzozowski

S. Balakrishan

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J. Flannery

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S. Mittler

Jai Sabrinathan

R. Lipson

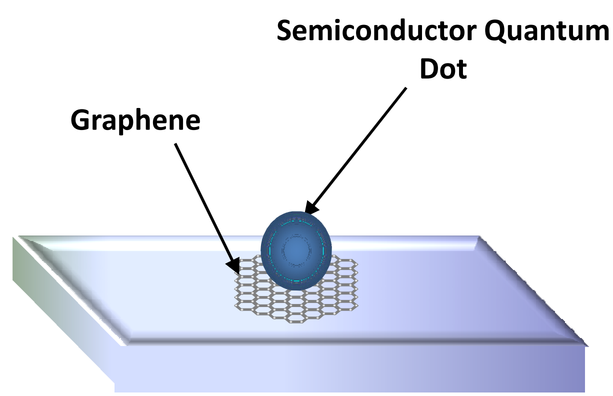
Hybrid Nanomaterials

Nanomaterials:

- Metamaterials
- Graphene
- Semiconductor nanoparticles (quantum dots, nanocrystals, nanowires, etc.)
- Metallic nanoparticles (spheres, nanorods, nanodisks, etc.)

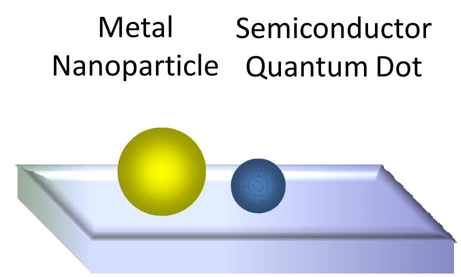
Substrate examples:

- ➢ Metamaterials
- ➢ Dielectric material
- ➢ Excitonic material
- ➢ Polaritonic material
- ➢ Polar materials
- ➢ Photonic crystal

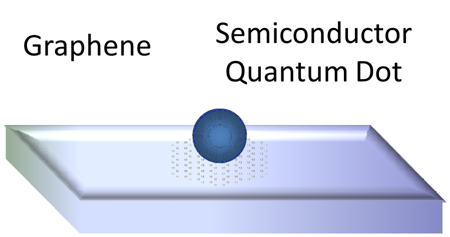


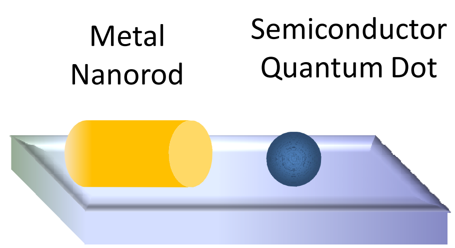
Hybrid Nanomaterials

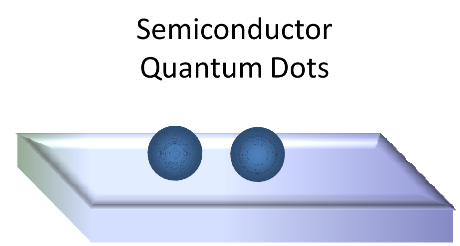
By using various combinations of nanostructures one can create enormous numbers of nanocomposite (hybrid) materials.

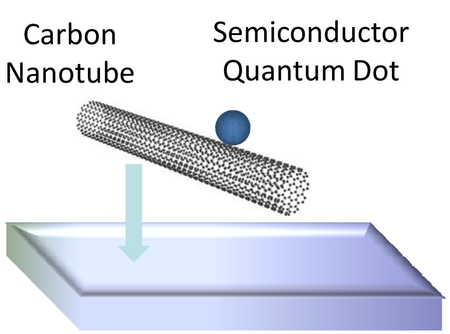


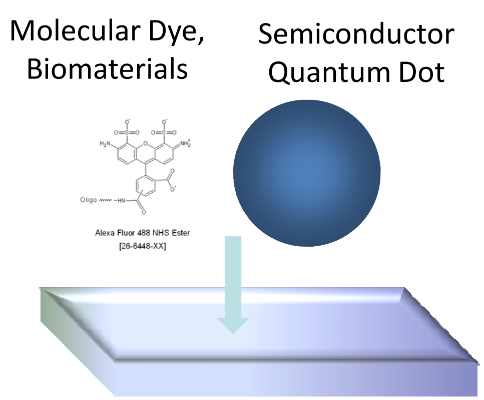
Substrates: dielectrics, photonic crystals, metamaterials

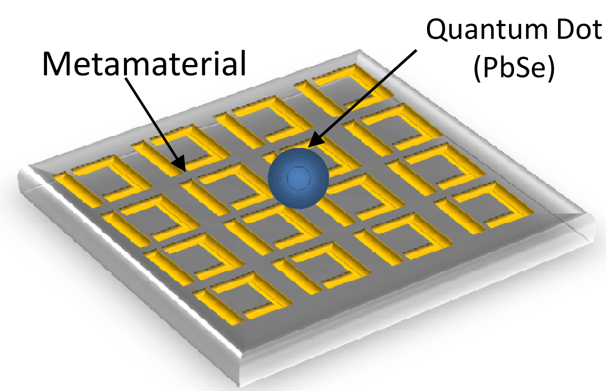












Metallic Nanomaterials: Plasmonics

J. Cox and Mahi Singh, Nanotechnology (2013)

- Metallic nanostructures: Gold, silver, copper
- Metals have free electrons which oscillate collectively to produce plasmons

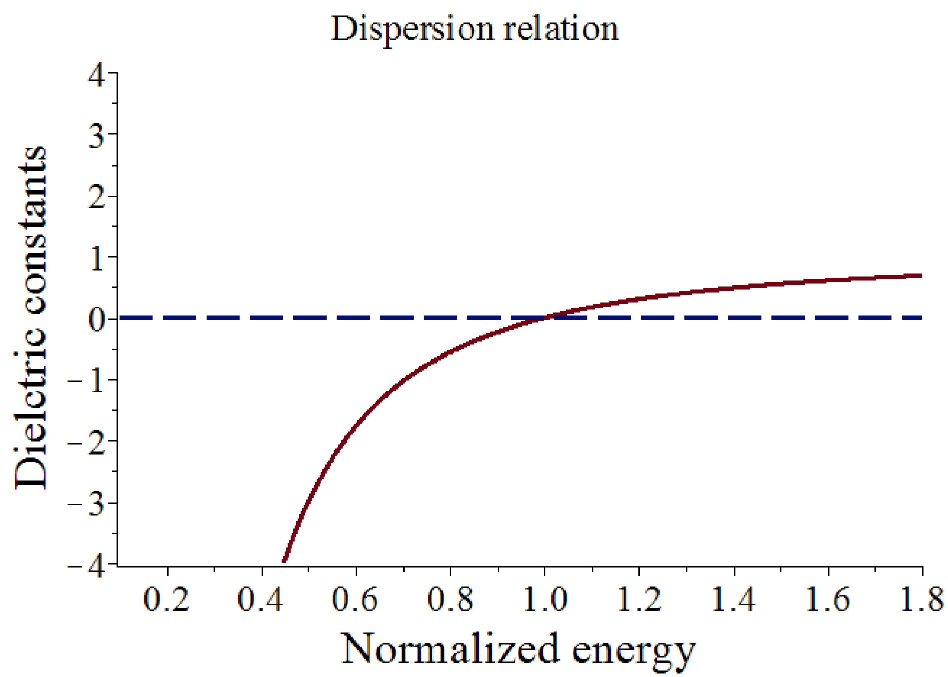
Dielectric constant

$$\in_m(\omega) \approx \left(1 - \frac{\omega_p^2}{\omega^2}\right)$$

$$\omega_p = plasmon$$

$$\omega_p = \sqrt{\frac{n(\varepsilon_F)e^2}{m}}$$

$$\varepsilon_F = \text{Fermi energy}$$



Dielectric constant is negative

Metallic Nanomaterial: Graphene

Graphene was invented by a Canadian physicist

P.R. Wallace (1915-2006)
McGill University, Montreal

Worked with Leopold Infeld, (Albert Einstein), NF Mott

1947: P.R. Wallace : Phys. Rev. 71, 622-634 (1947)

Graphite-moderated nuclear reactor project
(this project was part of a plan to develop nuclear weapons during the **II world war)**

Graphene is a gapless semiconductor

2010 Nobel Prize in Physics:
Andre Geim, Konstantin Novoselov

Mahi Singh and PR Wallace
Gapless Semiconductors

- M. SINGH and P.R. WALLACE , J. PHYSICS C 20, 2169, (1987).
- GUPTA, WALLACE and SINGH , J .PHYSICS C19, 6373 (1986).
- M. SINGH and P.R. WALLACE, SOLID STATE COMMUN. 53, 165 (1985).
- M. SINGH and P.R. WALLACE, J. PHYSICS C17, 5303 (1984).
- M. SINGH amd PR Wallace, J. PHYS. CHEM. SOLIDS 45,409 (1984).
- M. SINGH, P.R. Wallace and J. Leotin, J. PHYS. C17, 1385 (1983).
- M. SINGH and P.R. WALLACE , J. PHYS. C16, 3877 (1983).
- SINGH, LEOTIN and WALLACE, PHYS. STAT. SOLIDI B115, 105 (1983).
- M. SINGH and P.R. WALLACE , SOLID STATE COMM. 45, 9 (1983).
- M. SINGH, J. and P.R. WALLACE , PHYSICA B117, 441 (1983).
- SINGH, P.R. WALLACE, ASKENAZY, J. PHYSICS C15, 6731 (1982).
- SINGH, CISOWSKI, WALLACE, PORTAL, BROTO,
  Phys. Stat SOLIDI B114, 481 (1982).

(1915-2006)

P.R. Wallace

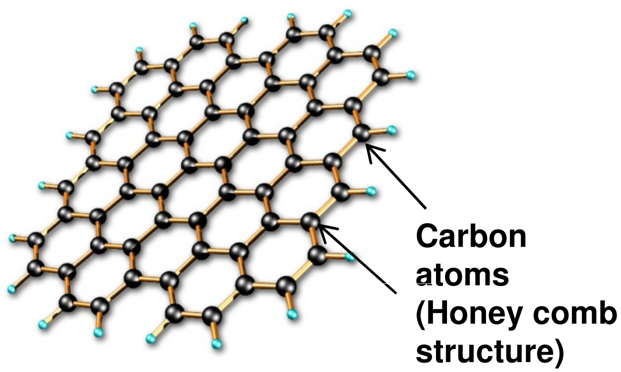
McGill University, Montreal

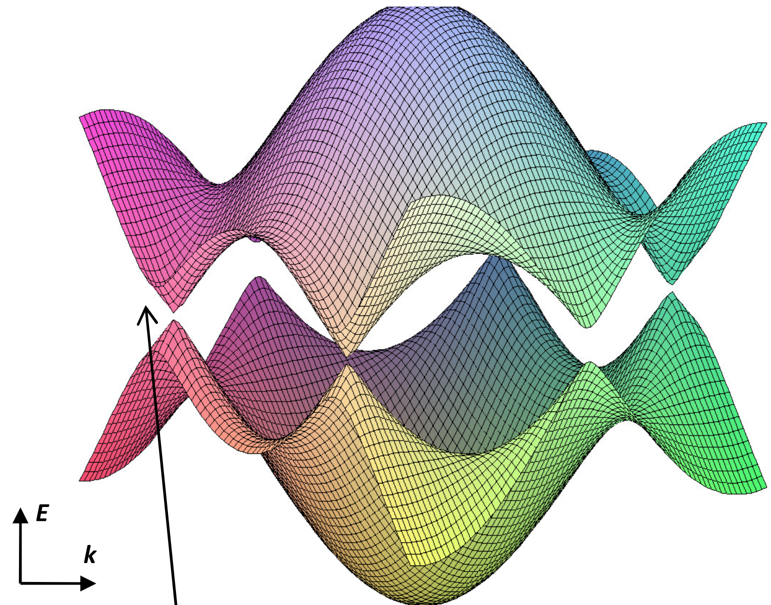
- I called him “SIR” and he did not like it.
- I have a graduate student who always calls me “SIR” and I like it and he is my favorite student.

Graphene : Plasmonics

P.R. Wallace, Phys. Rev. 71, 622-634 (1947)

- Graphene is a two dimensional sheet of carbon atoms which are arranged in the honey comb structure.



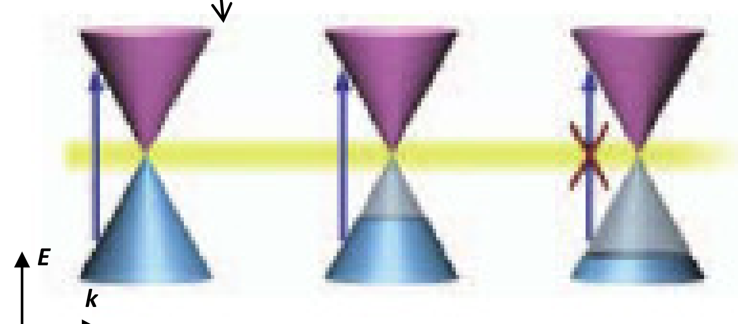


Mahi R. Singh, Electronic, Photonic, Polaritonic and Plasmonic Properties of aterials (Wiley Custom, Toronto, 2014)

Band structure:

- Gapless semiconductor and acts as a metal.
- k.p method
- Tight binding method

$$\varepsilon_k = \hbar v_F k$$



A.N. Grigorenko et al, Nature Photonics 6, 749 (2012)

Dielectric constant

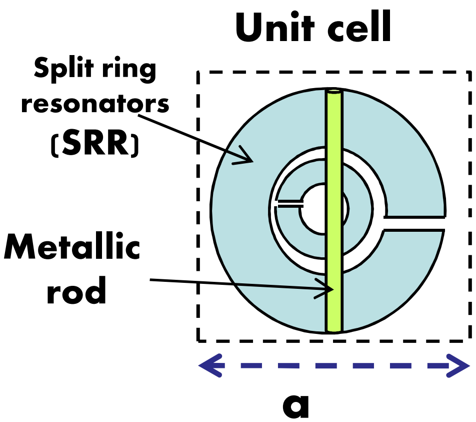
$$\in_m(\omega) \approx \left(1 - \frac{\omega_p^2}{\omega^2}\right) \qquad \omega_p = \sqrt{\frac{n(\varepsilon_F)e^2}{m}}$$

Dielectric constant is negative

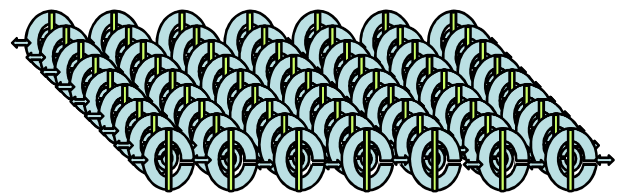
Metamaterials: Plasmonics

Mahi Singh. J. Cox, M. Brzozowski : J. Physics D: Applied Physics (2014)
Mahi Singh. C. Rackner, M. Brzozowski: Physical Review A (2014)

J.C. Bose, Proc. Royal Soc. 63, 146 (1988)
J.B. Pendry, Phys. Rev. Lett. 85, 3966 (2000)
Artificial materials made from negative dielectric constant an negative magnetic permeability



Unit cell =20-500nm, energy range= GHz-THz



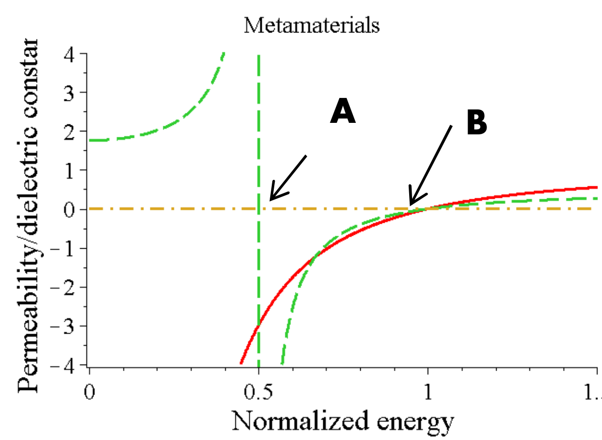
2-D-Metamaterial

3-D metamaterials

Dielectric constant negative magnetic permeability

$$\in_m(\omega) \approx \left(1 - \frac{\omega_p^2}{\omega^2}\right)$$

$$\mu_m(\omega) \approx \left(1 - \frac{F\omega^2}{\omega^2 - \omega_0^2}\right)$$



Note: ϵ and μ are negative between A and B

Dielectric constant is negative

Metamaterials: plasmonics

Mahi Singh. J. Cox, M. Brzozowski : J. Physics D: Applied Physics (2014)
Mahi Singh. C. Rackner, M. Brzozowski: Physical Review A (2014)

Natural Materials

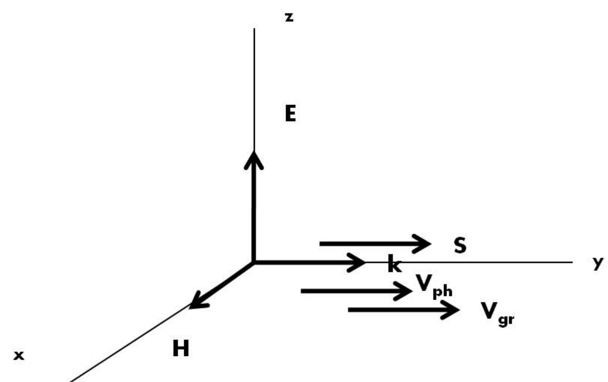
Maxwell equations:

$$\mathbf{k} \times \mathbf{E} = +|\mu|\,\omega \mathbf{H}$$

$$\mathbf{k} \times \mathbf{H} = -|\varepsilon|\,\omega \mathbf{E}$$

Poynting Vector (direction of the energy flow)

$$\mathbf{S} \propto \mathbf{E} \times \mathbf{H}$$



Right handed materials

Natural materials follow the Right Hand Rule (Right handed materials)

Metamaterials

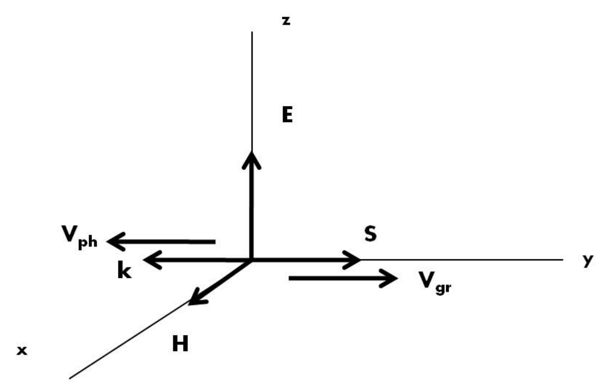
Maxwell equations:

$$\mathbf{k} \times \mathbf{E} = -|\mu|\,\omega \mathbf{H}$$

$$\mathbf{k} \times \mathbf{H} = +|\varepsilon|\,\omega \mathbf{E}$$

Poynting Vector (direction of the energy flow)

$$\mathbf{S} \propto \mathbf{E} \times \mathbf{H}$$



Left handed materials

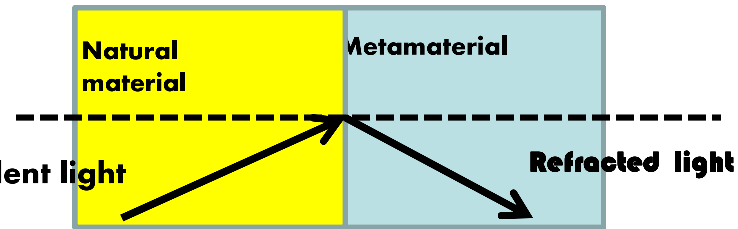
Metamaterials follow the Left Hand Rule (Left handed handed materials)

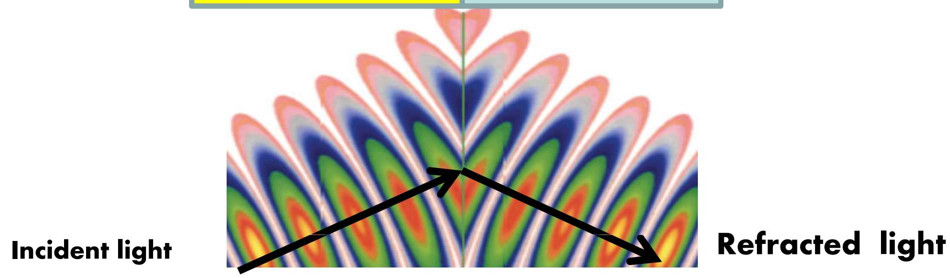
Metamaterials: plasmonics

Mahi Singh. J. Cox, M. Brzozowski : J. Physics D: Applied Physics (2014)
Mahi Singh. C. Rackner, M. Brzozowski: Physical Review A (2014)

Snell's law

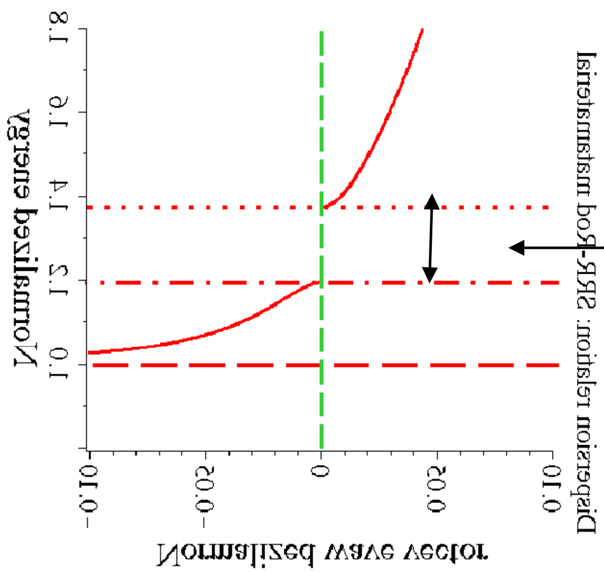
$$\frac{Sin(\theta_r)}{Sin(\theta_i)} = \frac{n_d}{n_m} = \frac{\sqrt{|\in_d||\mu_s|}}{s\sqrt{|\in_m||\mu_m|}}$$





Band structure

3-D metamaterials



Band gap

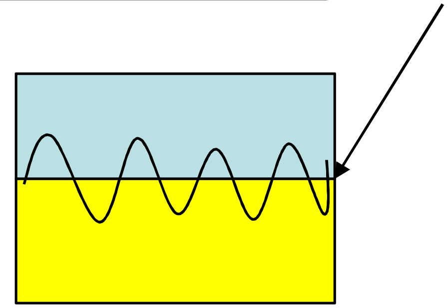
Cloaking: spaceships and individuals entirely invisible in light.

Electric Field: Metallic Nanomaterials

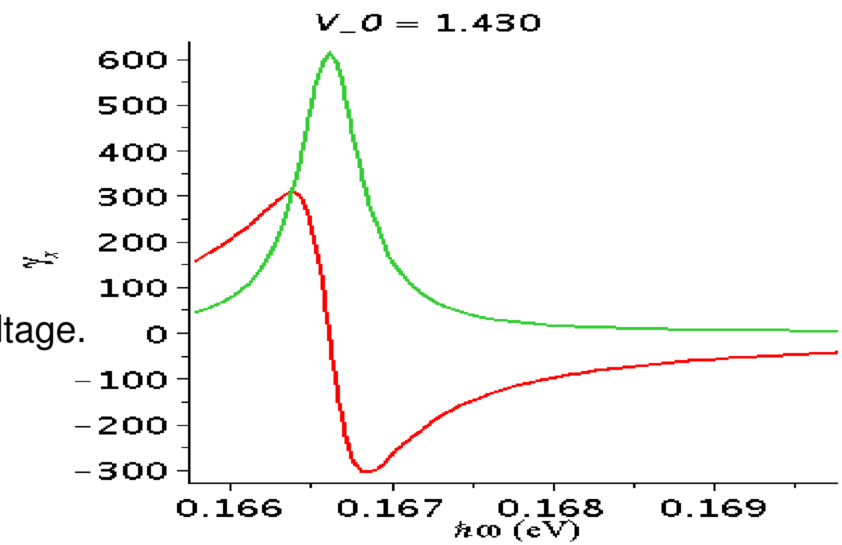
J. Cox and Mahi Singh, Nanotechnology (2013)

SPP Electric Field

$$E_{dipole} \approx \left(\frac{\in_m(\omega) - \in_s}{\in_s + \varsigma_{size}\left[3\in_s + 3\in_m(\omega)\right]} \right)$$



$$\in_m(\omega) \approx \left(1 - \frac{\omega_p^2}{\omega^2} \right) \qquad \omega_p = plasmon$$



Graphene: Gate voltage

- Plasmon frequency can be changed by applying the gate voltage.
- Gate voltage changes the Fermi energy.

$$\omega_p = \sqrt{\frac{n(\varepsilon_F)e^2}{m}} \qquad \varepsilon_F = \text{Fermi energy}$$

Parameters: Radius= 8 nm; v_F = c/300; μ = 10000 cm^{2}V^{-1}s^{-1}; ε_b = 10.89 (GaAs)

Photonic Crystals: Photonics

E. Yablonovitch, PRL 58, 2059 1987

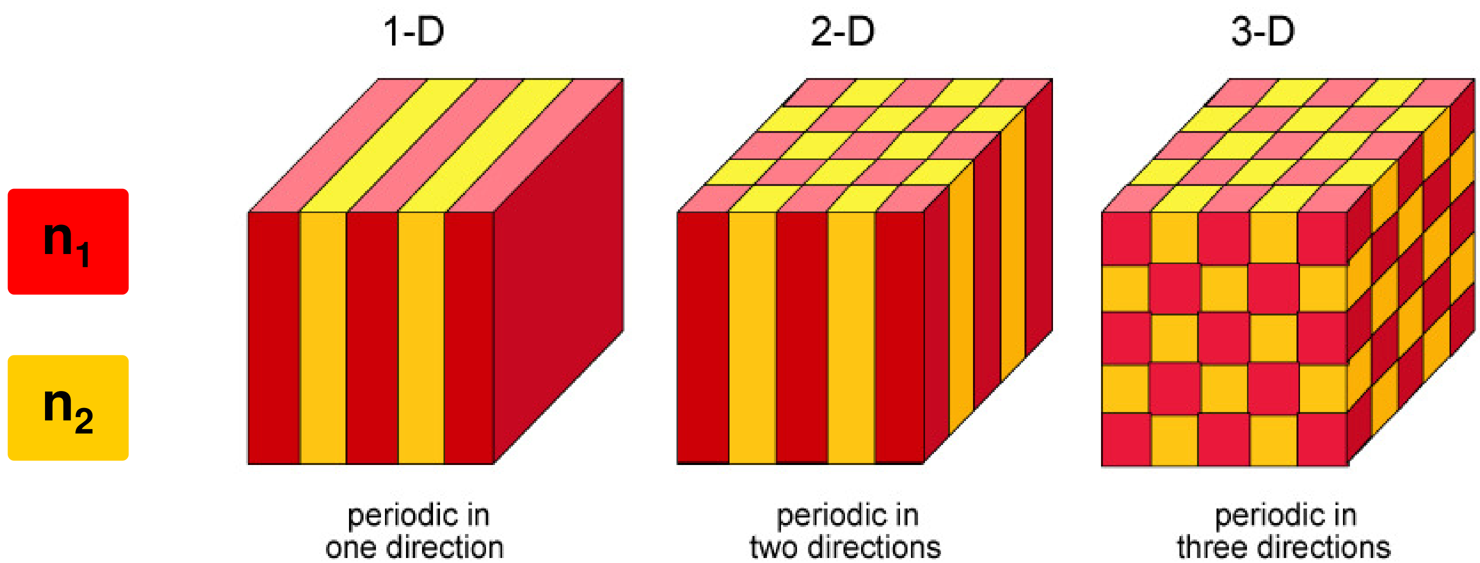
S. John, PRL 58, 2486 1987

- Periodic dielectric lattice
- Periodic dielectric constant function

Revenge of Electromagnetism

- Stop Bands: control of light propagation
- Complete and Partial gaps

Source: math.utwente.nl/~hammer/Metric/Illust/



Source: "Photonic Crystals", Joannopoulos, Meade and Winn.

Western Science

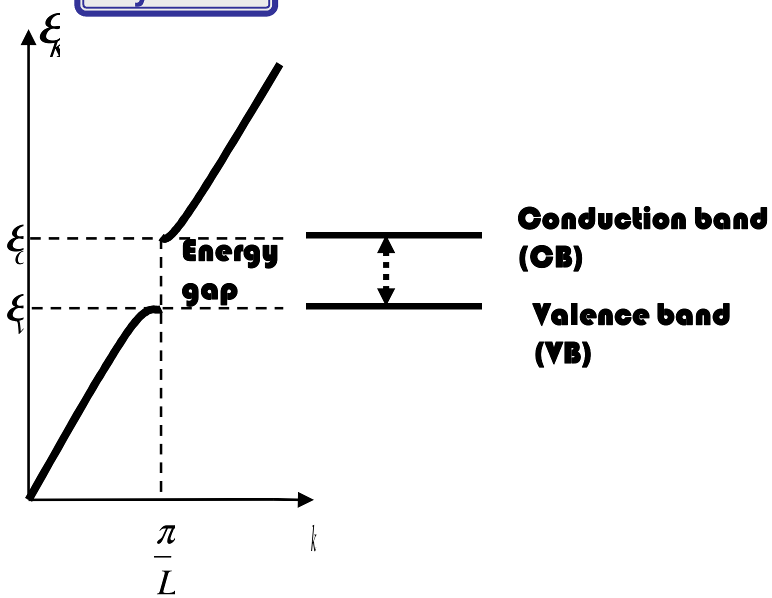
Photonic Materials: Photonics

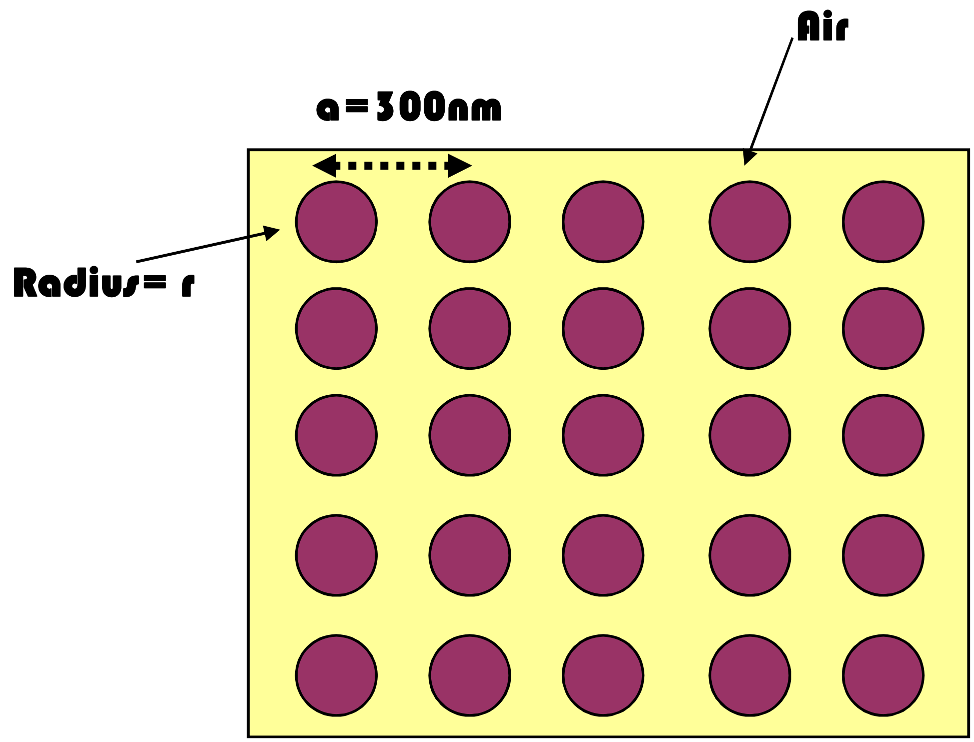
I. Haque and Mahi Singh. J. Phys. Condens. Matter 19, 156229 (2007)

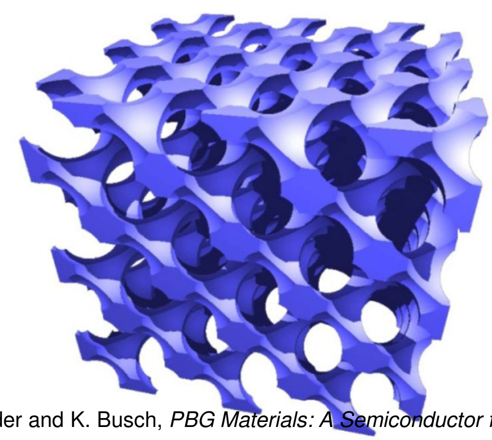
Dielectric spheres (Si) in air

Lattice constant a=300nm; Radius r/a=0.3; Spheres= Si (n=3.4); Background air (n=1)

Photonic crystal







Note: The location and value of the gap depend on the refractive index.

Source: S. John, O. Toader and K. Busch, *PBG Materials: A Semiconductor for Light*, 2002.

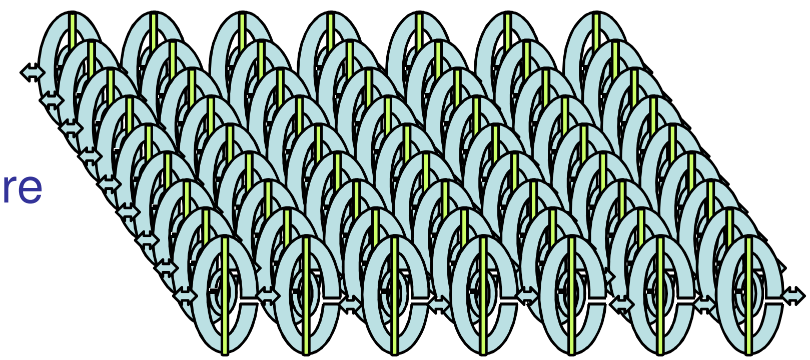
Metamaterials: Photonics

Mahi Singh. J. Cox, M. Brzozowski : J. Physics D: Applied Physics (2014)
Mahi Singh. C. Rackner, M. Brzozowski: Physical Review A (2014)

Band structure:

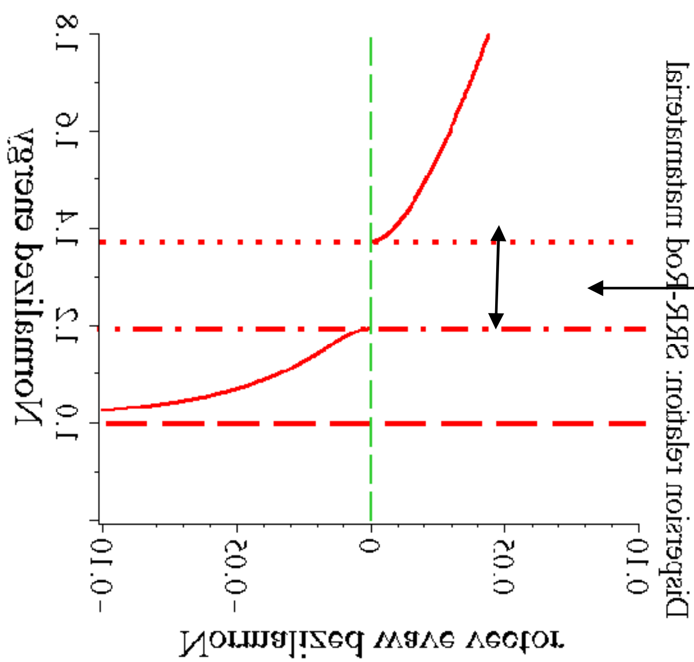
Metamaterials have Periodic structure

Solving Maxwell equations in
periodic refractive index



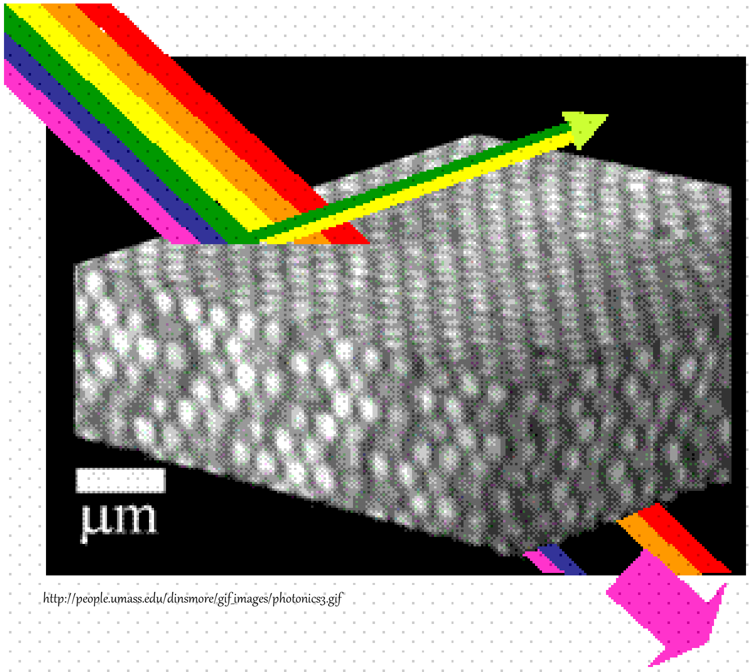
2-D-Metamaterial

3-D metamaterials



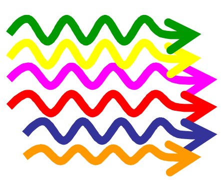
Band gap

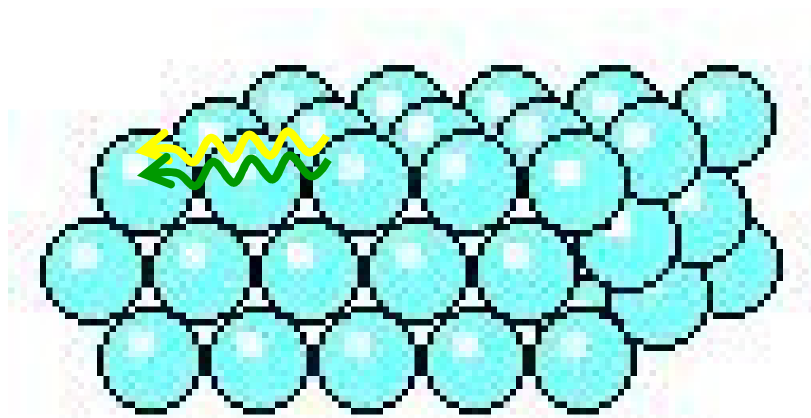
Metamaterials/photonic crystal



Reflected Colours
-Yellow
-Green

Transmitted Colours
-Purple
-Orange
-Red
-Blue

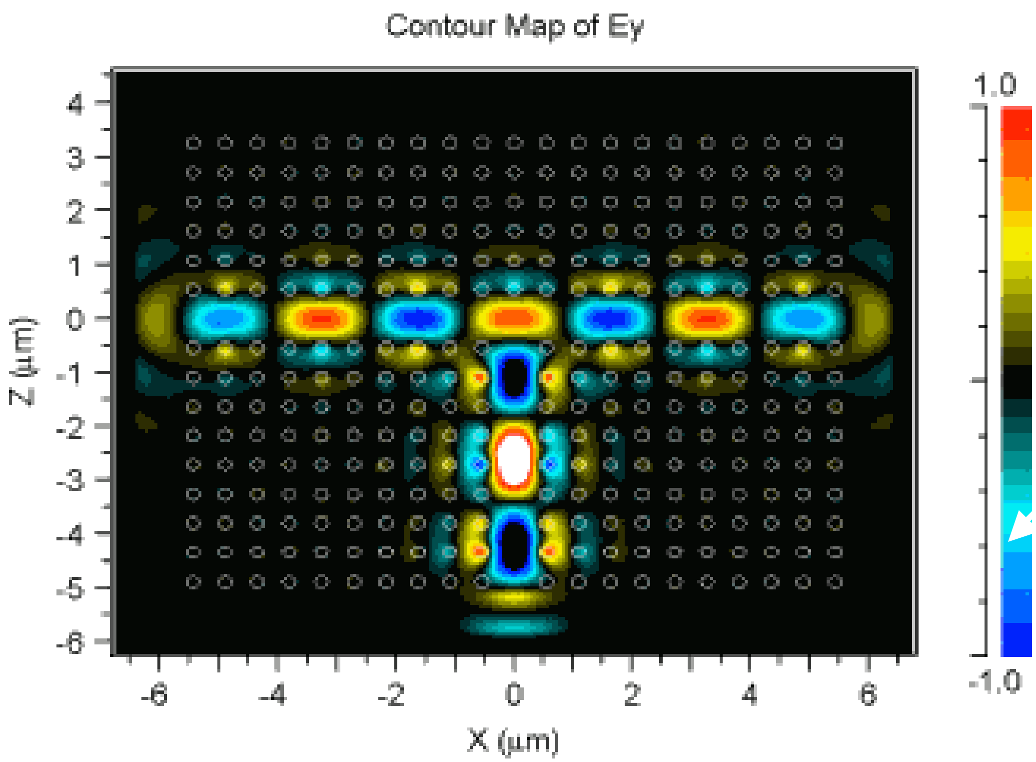


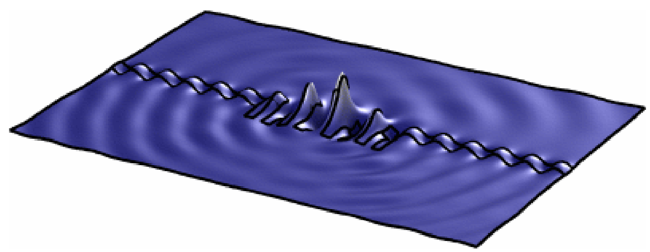


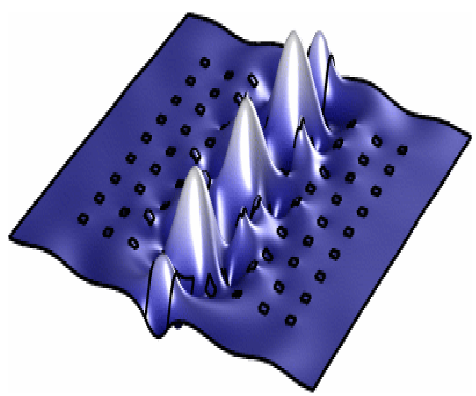
Photonic Crystals: Applications

- Photons are confined in a nano-size material

Photons are confined in 2-d







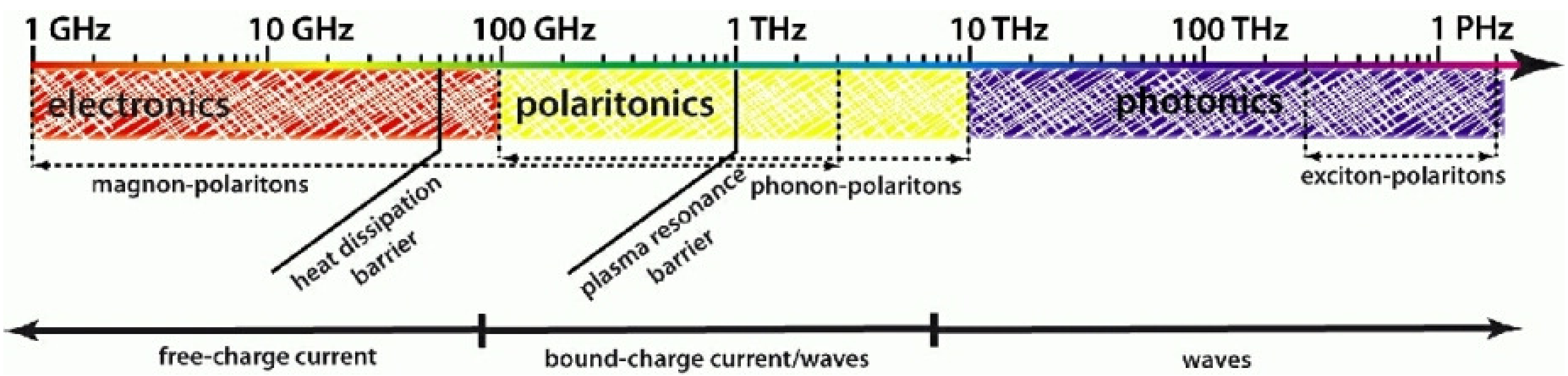
Source: math.utwente.nl/~hammer/Metric/Illust/
http://www.photeon.com/images/pc/animation.gif

90º Light Splitter

Applications: Polaritonic Materials

C. Racknor, Mahi Singh. Phys. Rev. B 82, 155130 (2010)
Cox, Singh and Racknor, **Nano Lett. 11, 5284 (2011)**

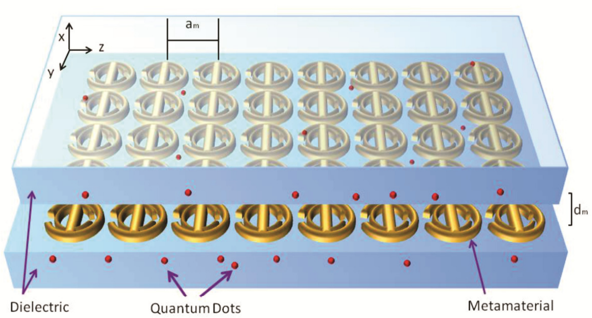
- Polaritonic materials have a band gap that lies in the terahertz frequency range.
- This opens a new realm of possibilities for opto-electronic devices because this range of frequencies is intermediate between the operational frequency ranges of photonics and electronics.



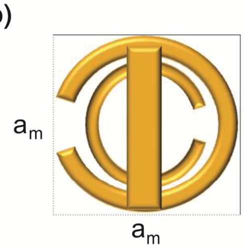
Graphene/Quantum Dot Hybrid deposited on a Photonic Crystal /Metamaterials

- We consider a nanocomposite consisting of a graphene nanoflake and a QD
- The graphene-QD nanocomposite is embedded in a photonic crystal
- When external laser fields are applied, plasmons in graphene interact with excitons in the QD
- Photonic crystal serves as an electromagnetic reservoir for the QD

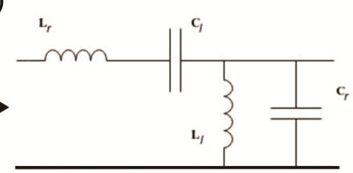
1.a)

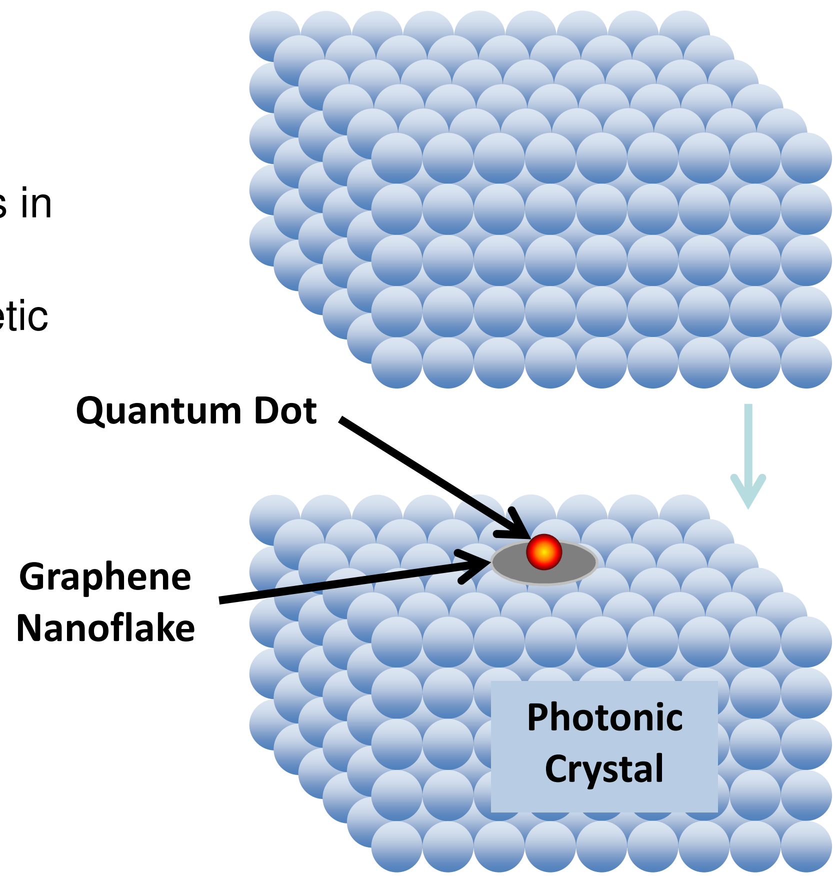


1.b)



1.c)





Cox , Singh et al, Physical Review B 86, 125452 (2112)
Singh ,Cox et al , Advance Materials (2013)

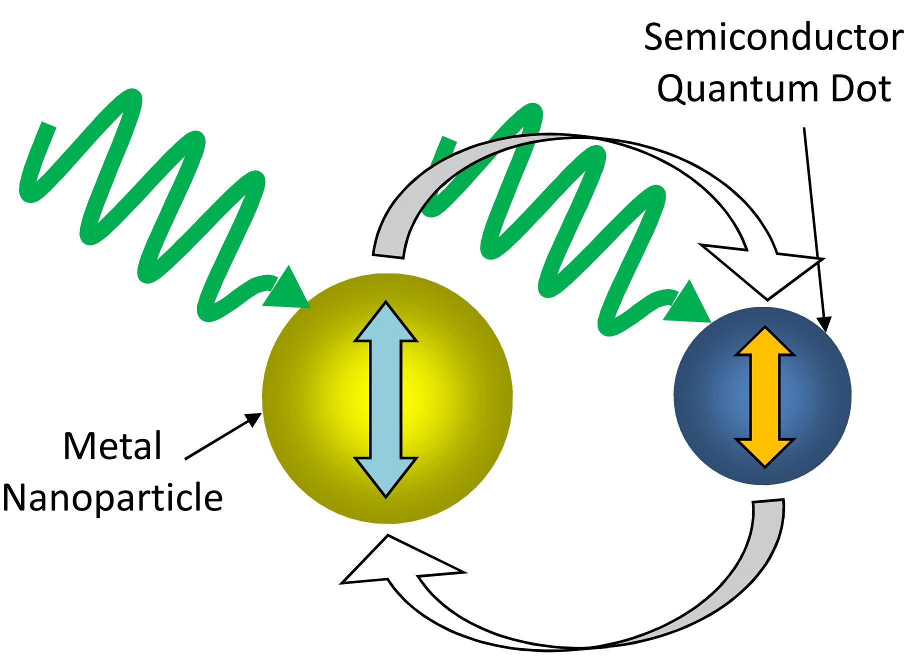
Dipole-Dipole Interaction

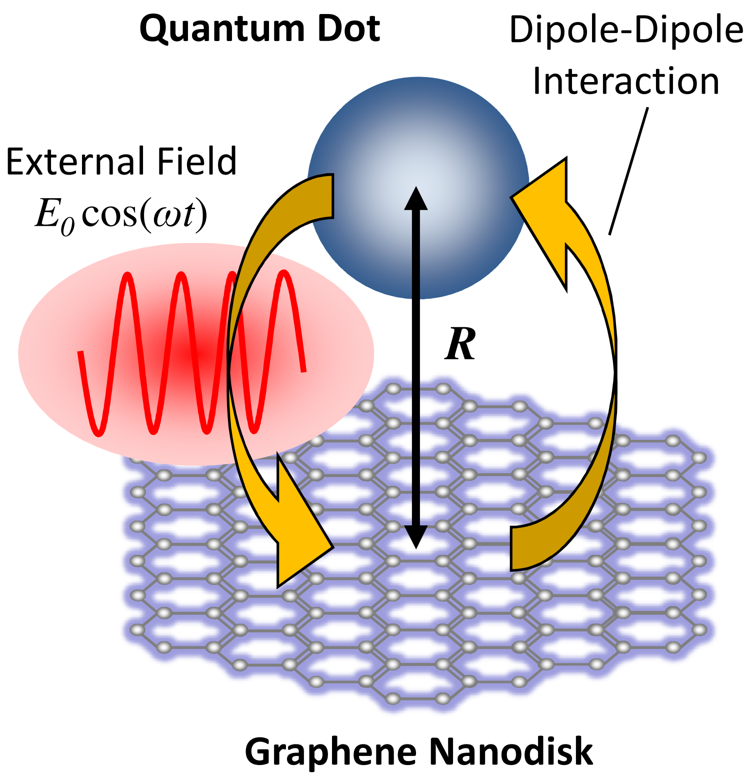
- In QD-MNP hybrids, excitons in the QD and localized surface plasmons in the MNP interact via the dipole-dipole interaction (DDI)

- This interaction is strong when the QD and MNP are in close proximity and their optical excitation frequencies are resonant

Cox and Singh, Physical B 86, 125452 (2012)
Hatef and Singh, Nanotechnology 23, 205203 (2012)

$$E_{dip} \approx \frac{P_{MNP}}{R^3}$$



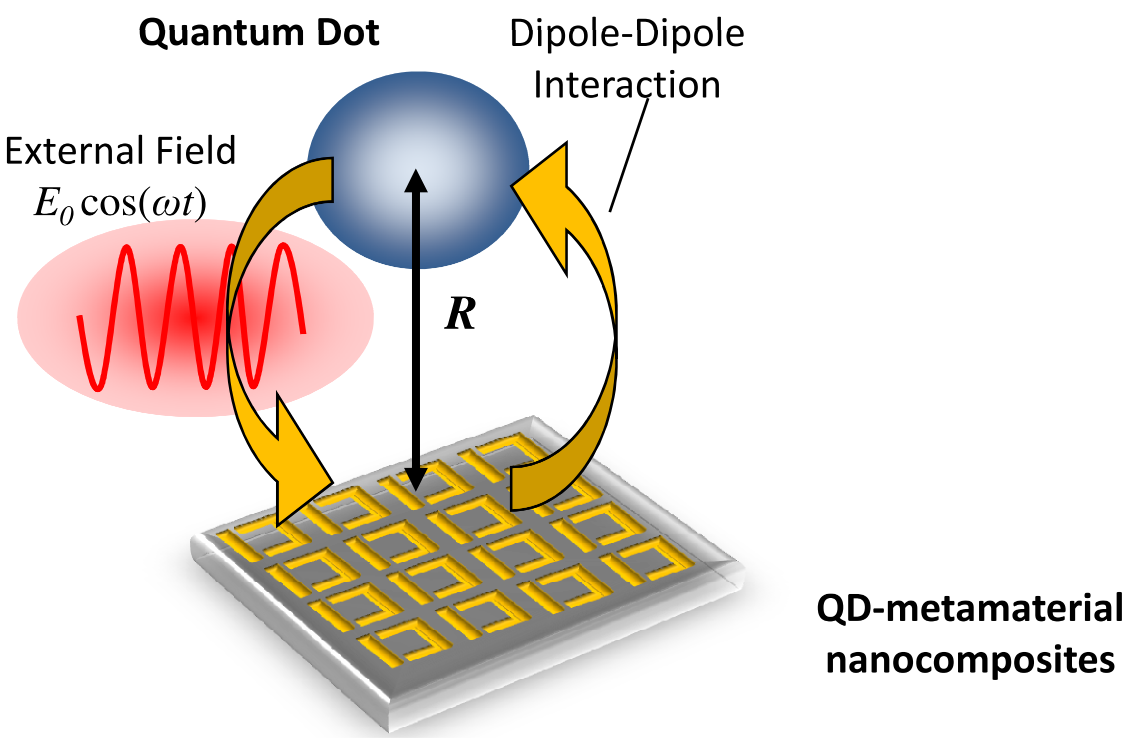


Excitons-SPPs Interaction

Mahi Singh. J. Cox, M. Brzozowski : J. Physics D: Applied Physics (2014)

➢In QD-MNP hybrids, excitons in the QD and localized surface plasmons in the MNP interact via the excitons-SPPs (dipole-dipole interaction (DDI)).

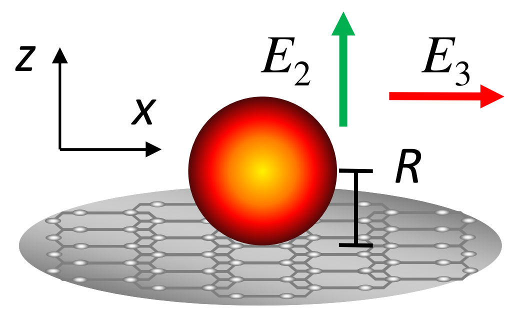
➢This interaction is strong when the QD and MNP are in close proximity and their optical excitation frequencies are resonant

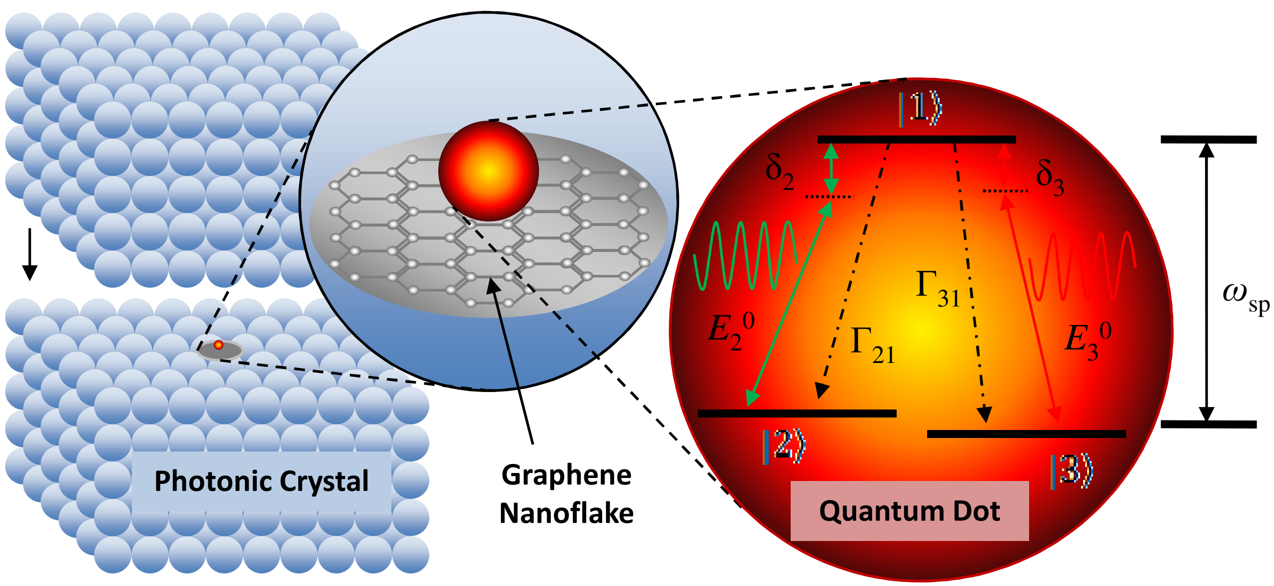


Graphene/Quantum Dot Hybrid

Cox, Singh et al., Physical Review B 86, 125452 (2012)

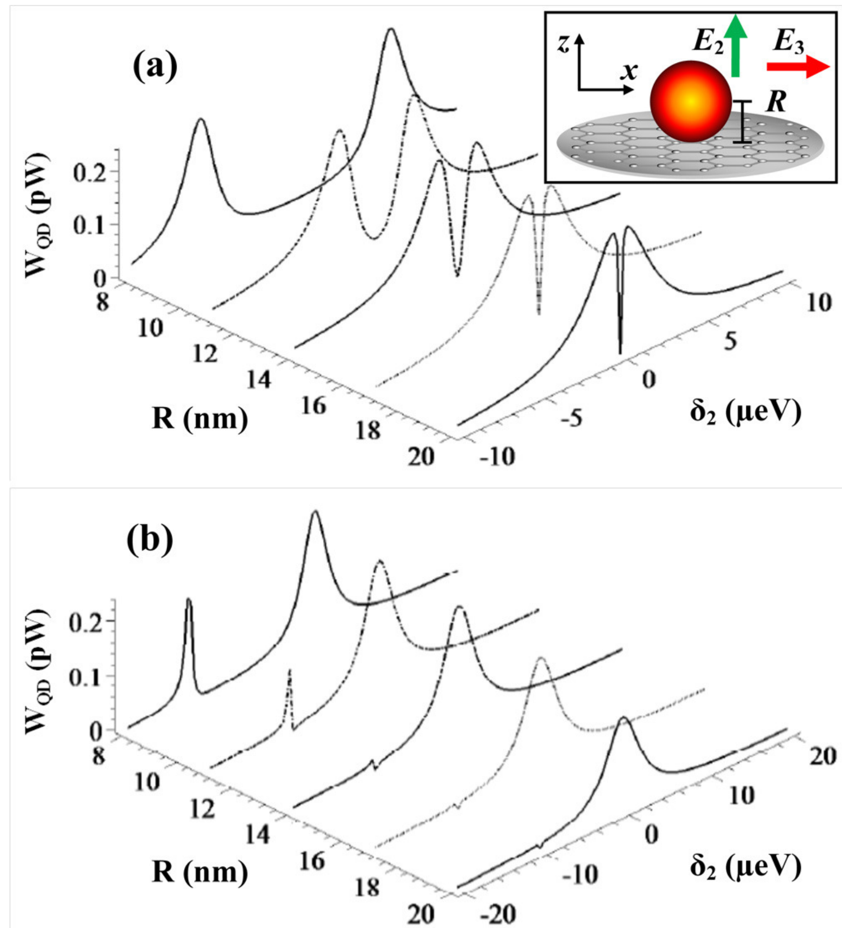
- It is considered that the resonance frequencies of the QD lie near ω_{sp} = 0.803 eV
- Initially we consider the case where the transition dipole moments (fields) μ_{12} (E_2) and μ_{13} (E_3) are aligned along the *z*- and *x*-directions, respectively
- Therefore only μ_{13} and E_3 couple to graphene





Power Absorption in QD

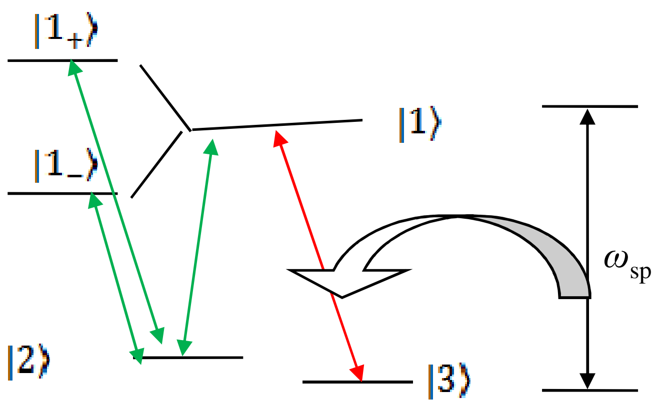
- Here the power absorbed by the QD is calculated while varying the graphene-QD separation *R*
- Narrow minima for larger values of *R* is due to electromagnetically induced transparency
- For small values of *R*, the spectrum splits into two peaks due to the DDI
- Power is absorbed by the QD at two frequencies
- (a) $\delta_3 = 0$; (b) $\delta_3 = 10$ μeV



Cox , Singh et al. Physical Review B 86, 125452 (2112)

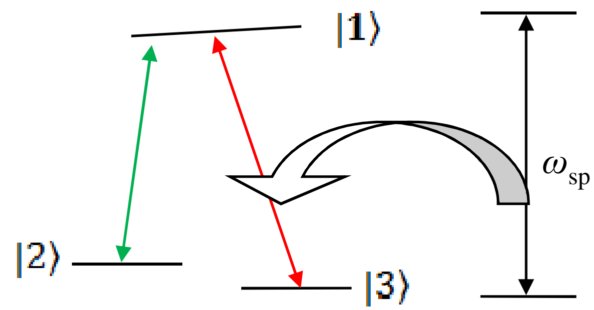
Switching Mechanism: One-photon spectroscopy

Cox, Singh et al., Physical Review B 86, 125452 (2012)

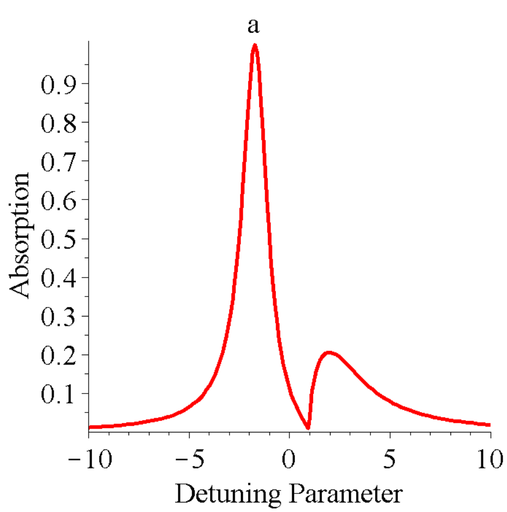


ABSORPTION:

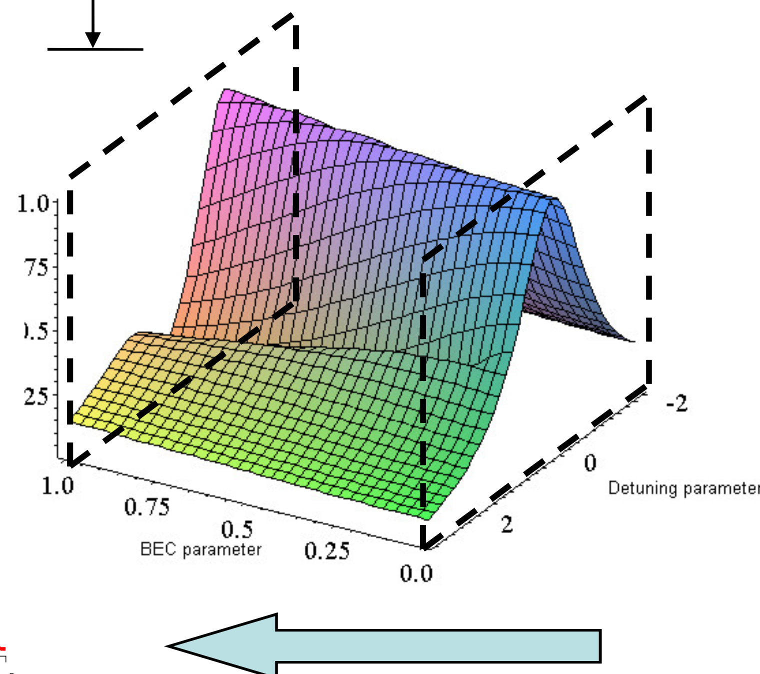
- transition from state from 2 to 1
- DDI: changes with R
- DDI: Gate voltage



Strong DDI coupling

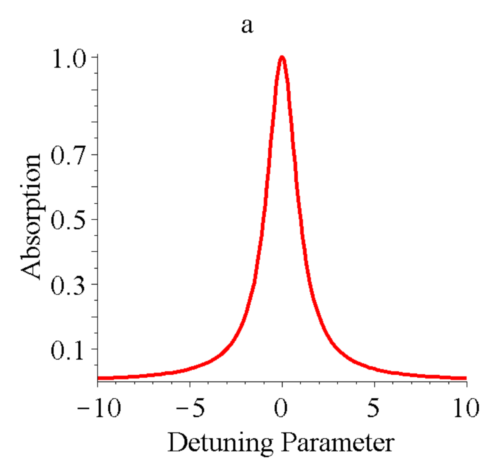


Photon-atom coupling TWO PEAKS.



coupling

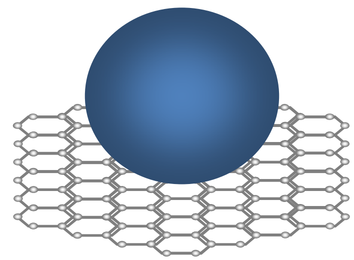
Weak DDI coupling



Photon-atom coupling = 0 ONE PEAK.

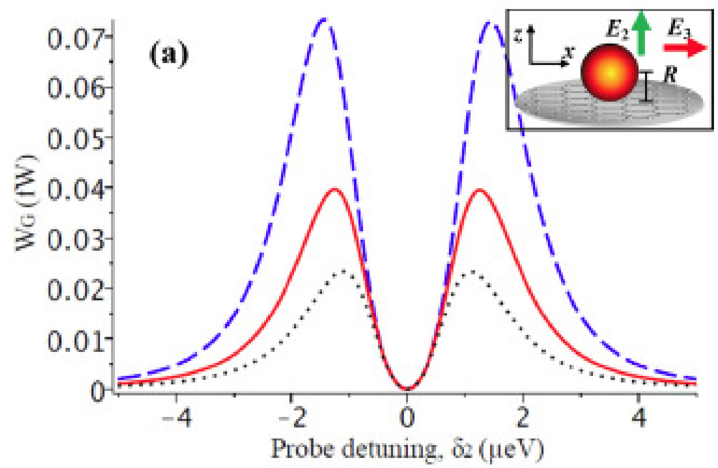
Sensing Mechanism: Substrate effect

- When a QD is in contact with biomolecules, molecular beacons, DNA or aptamers, its dielectric constant can be modified.
- This effect has also been verified experimentally by Dong et al., where upon integrating a molecular beacon to a CdTe-QD it was found that the fluorescence quenching due to graphene is modified.



H. Dong et al., Anal. Chem 82, 5511 (2010).

- ε_d = 10 (dotted curve)
- ε_d = 12 (solid curve)
- ε_d = 14 (dashed curve)
- Energy transfer to graphene when the QD dielectric constant is changed
  - Detection of biological molecules

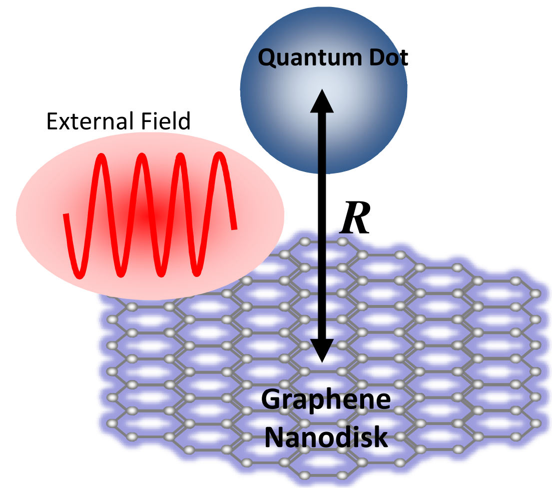


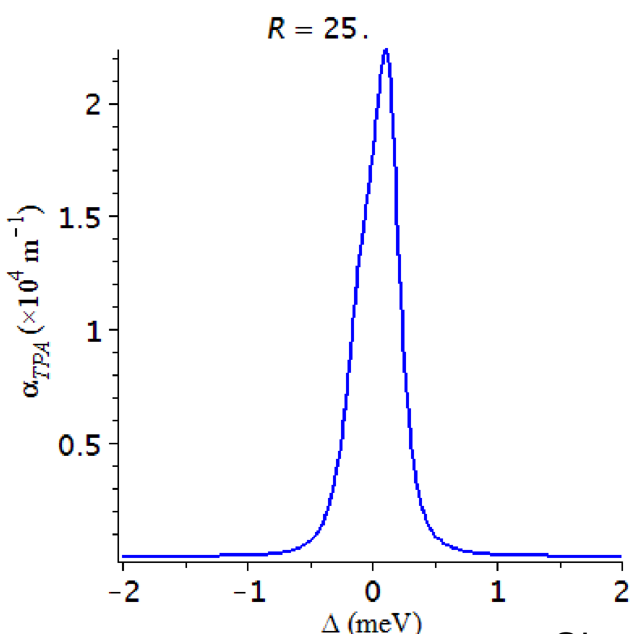
$$E_{dip} \approx \left(\frac{1}{[(2\in_b + \in_d)/3\in_b]} \frac{P_{QD}}{R^3} \right)$$

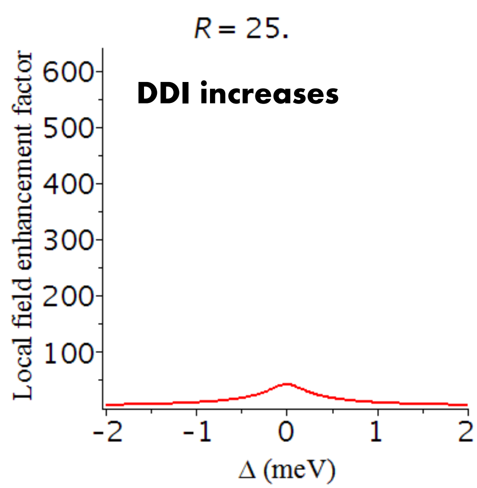
Cox and Singh, Physical Review B 86, 125452 (2012);
Singh, Racknor and Schindel **App. Phys. Lett. 101, 051115 (2012)**

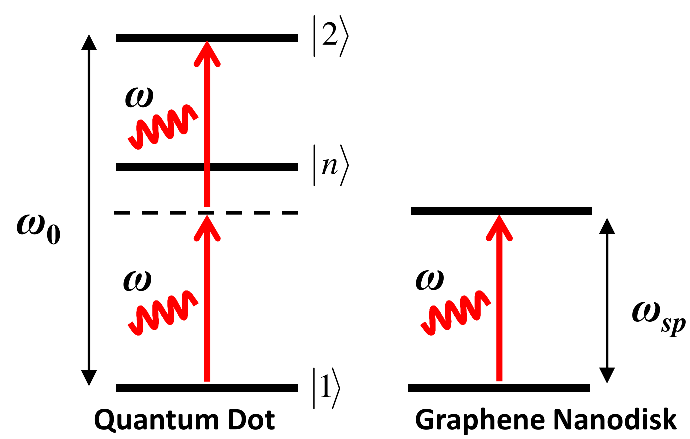
Two-Photon Process: QD-Graphene
DDI splitting

- Here the two-photon absorption coefficient is calculated as a function of the two-photon detuning parameter.
- The center-to-center distance between the quantum dot and graphene nanodisk, *R*, is varied





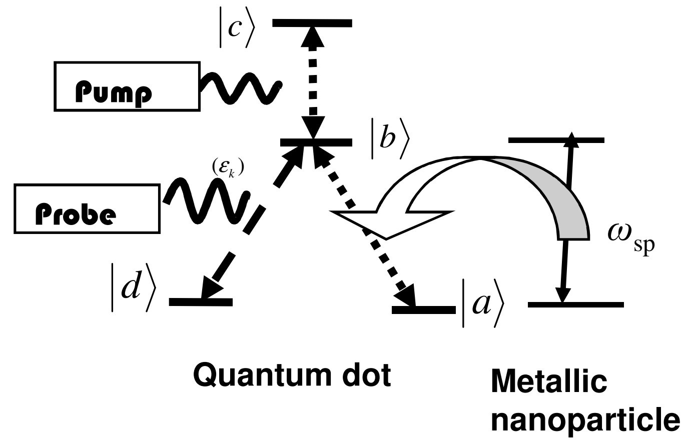




Singh, Cox et al., J. Phys.: Condens. Matter 25 (2013) 385302

One-Photon Process
DDI + Pump Field Effects

Singh: Optics Letters 34, 22 (2009)
Singh: Journal of Applied Physics 106, 063106 (2009.
Singh: Phys. Rev. B80 195303 (2009)
Singh: Phys. Rev. A 79, 013826 (2009).



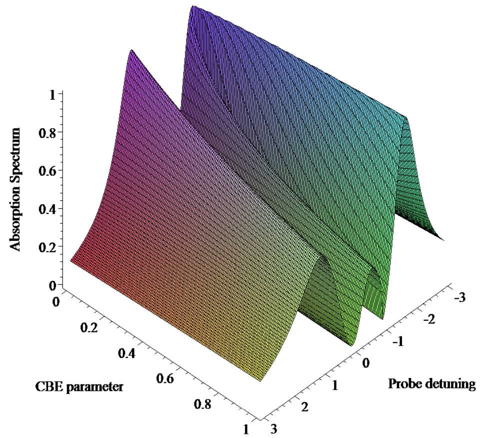
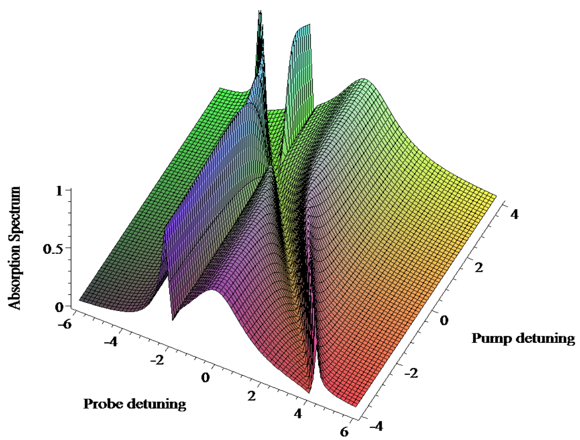
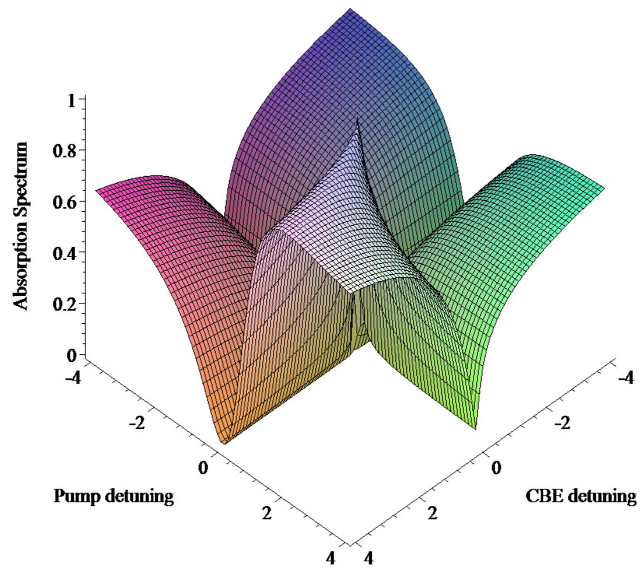


Fig.5





Sydney opera house ?

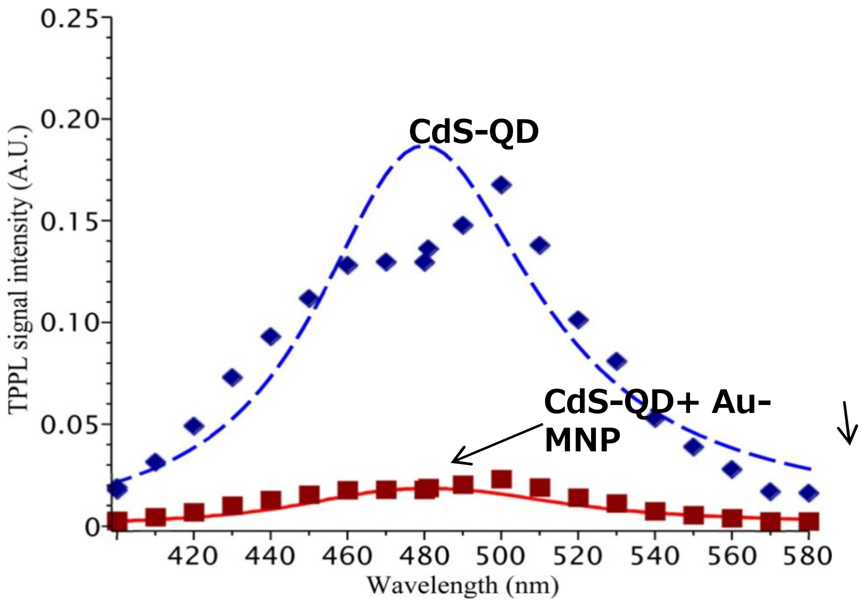
PHYSICAL REVIEW WEBSITE

Main Results: The system switches from one transparent state to two transparent states

Two-photon photoluminescence Quenching

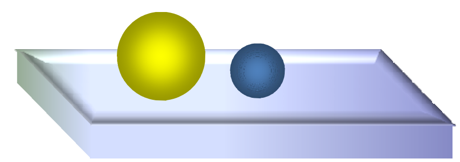
Cox, Singh, Bildering, Bragas, Advanced Materials 1, 460 (2013)

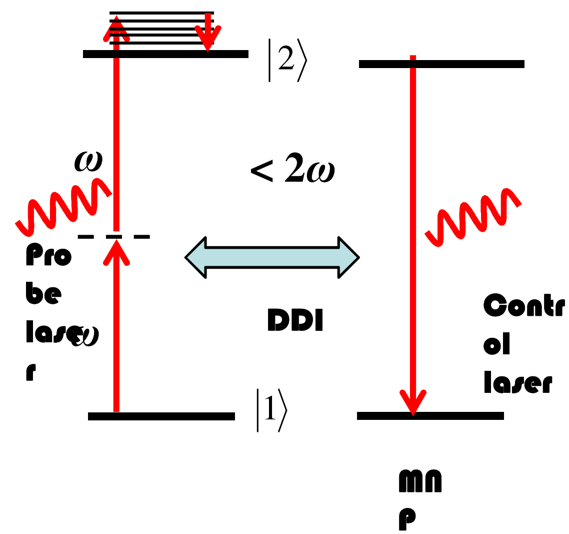
- Two-photon photoluminescence (TPPL) from CdS QDs alone (dashed curve) and from QD-Au MNP hybrid system (solid curve)
- TPPL from the QDs is quenched in the presence of the MNPs since the energy is transported from QD to MNP.



$$I_{2ph}^{QD} \approx W_{qd}^{0} \text{(energy emitted without MNP)}$$

$$I_{2ph}^{QD} \approx W_{qd}^{0} - W_{qd}^{mnp} \text{(energy emitted with MNP)}$$





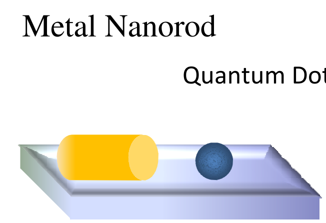
PL in QD

Second Harmonic Generation

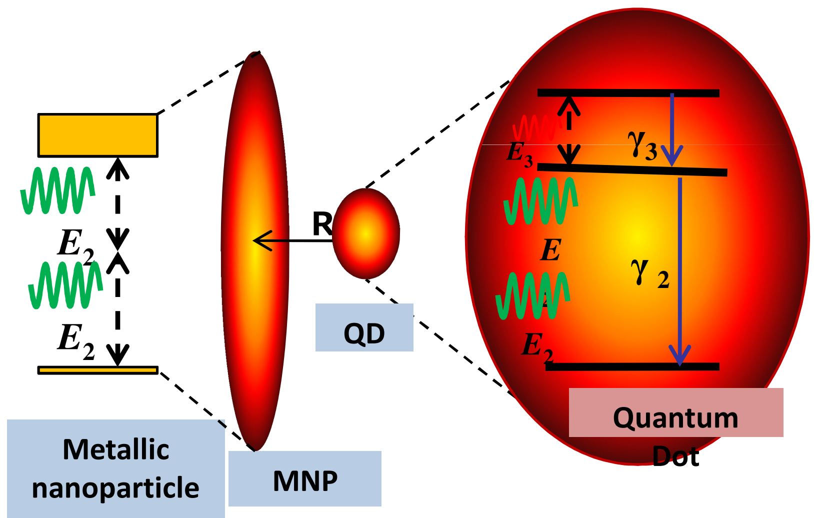
Singh., Nanotechnology 24 (2013) 125701

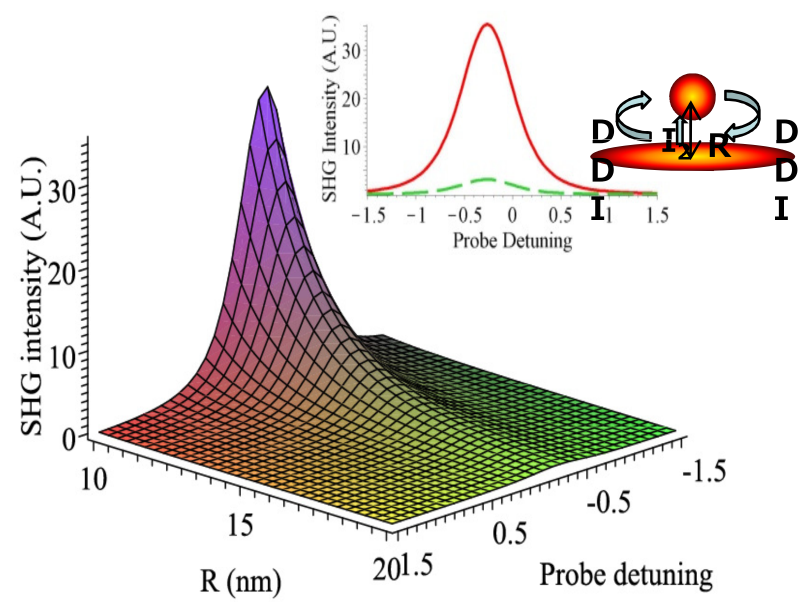
Enhancement effect:

We consider a QD-metallic nanorod hybrid deposited on s substrate



$$I_{SHG}^{QD} \approx \left|\Omega_{probe} + \Omega_{ddi} + \Omega_{SHG}^{mnp}\right|^2$$



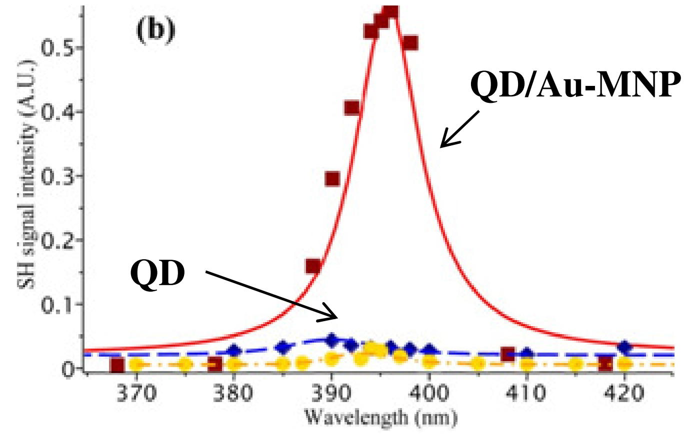


Enhancement of SHG is predicted due DDI

Switching on and off of the SHG is found

Second Harmonic Generation
Enhancement and switching

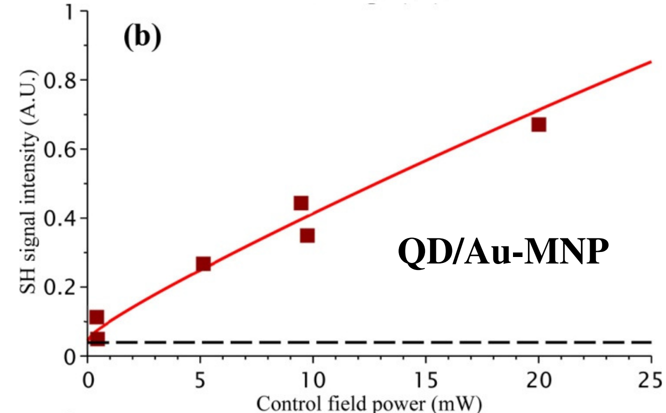
Cox, Singh, Bildering, Bragas, Advanced Materials 1, 460 (2013)



➢ Enhancement of SHG :

observed in 40 nm Au MNP-CdS-QD hybrid system . R = 41.5 nm.

$$I_{SHG}^{QD} \approx \left|\Omega_{probe} + \Omega_{ddi} + \Omega_{SHG}^{mnp}\right|^2$$



➢ Switching: **Due to control field intensity and frequency**

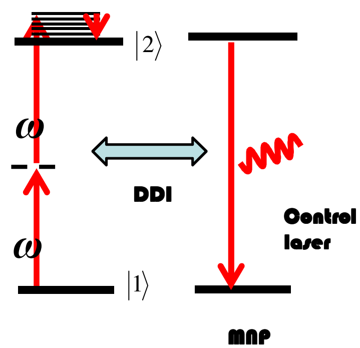
Switching DDI;: Dotted curve to solid curve

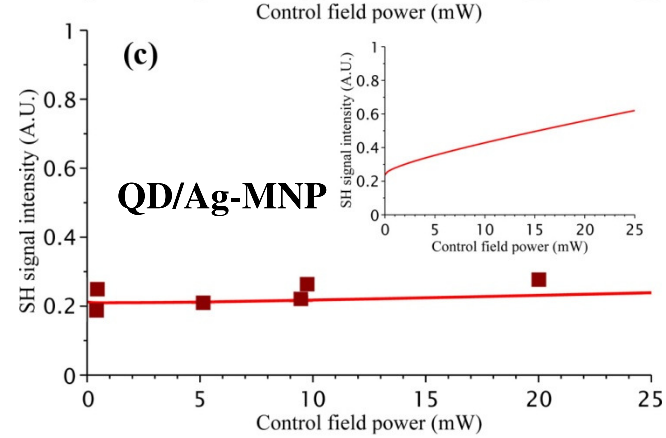
➢Dotted curve: The control field frequency is not resonance with the SPP of MNP: No DDI

$$I_{SHG}^{QD} \approx \left|\Omega_{probe} + \Omega_{SHG}^{mnp}\right|^2$$

➢Solid curve: With control field : DDI

$$I_{SHG}^{QD} \approx \left|\Omega_{probe} + \Omega_{ddi} + \Omega_{SHG}^{mnp}\right|^2$$





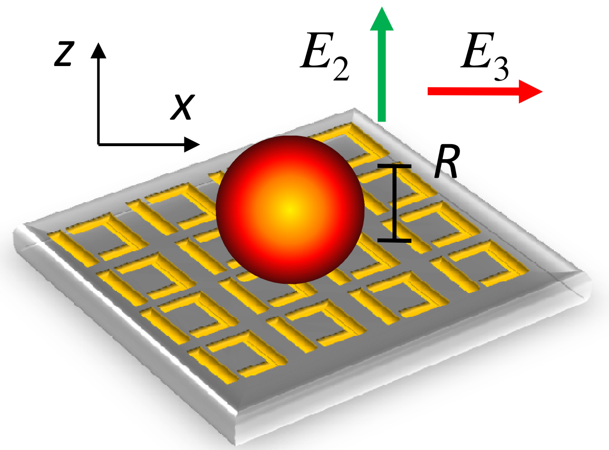
NO SHG in Ag-MNP- QD

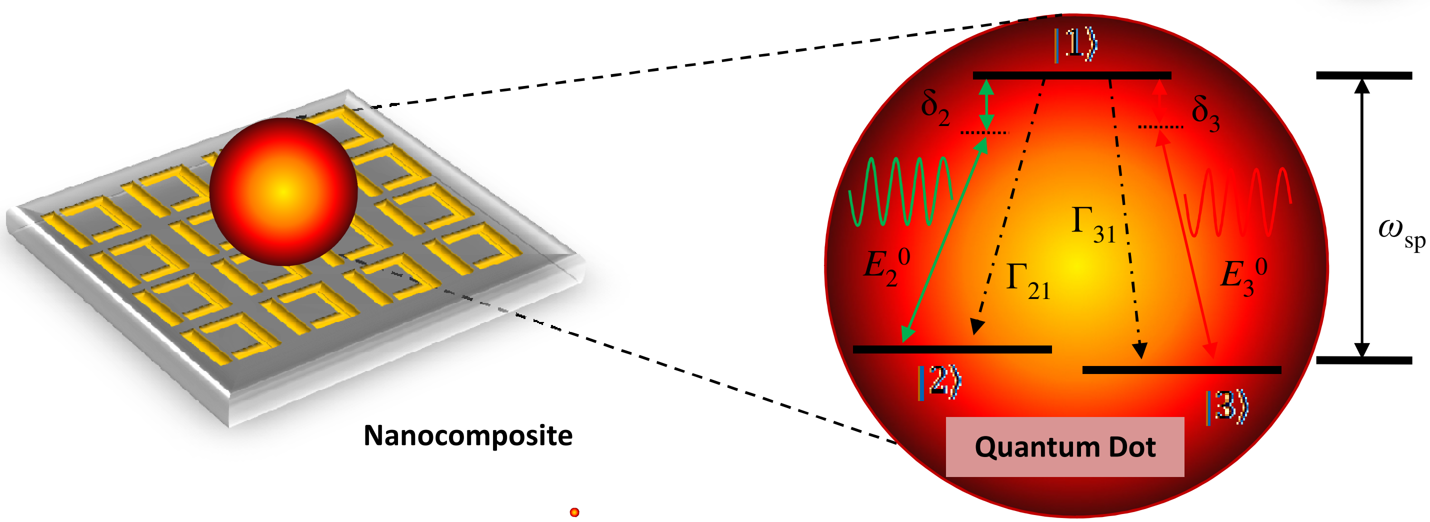
Control field frequency is not resonance with AU surface plasmon frequency.

Three level Quantum Dot

Mahi Singh. J. Cox, M. Brzozowski : J. Physics D: Applied Physics (2014)
)

- It is considered that the resonance frequencies of the QD lie near ω_{sp}
- Initially we consider the case where the transition dipole moments (fields) μ_{12} (E_2) and μ_{13} (E_3) are aligned along the *z*- and *x*-directions, respectively
- Therefore only μ_{13} and E_3 couple to SSP of metamterials



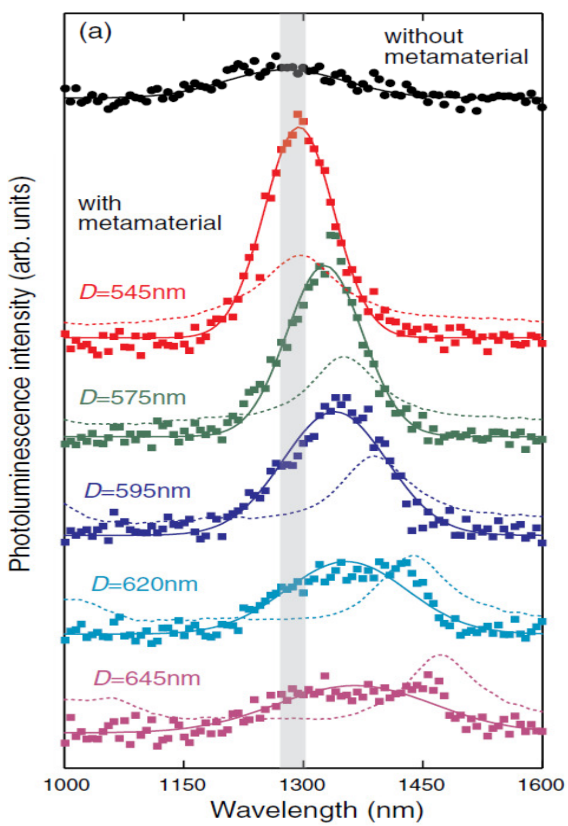


Enhancement of photoluminescence in metamaterials hybrid

Mahi Singh. J. Cox, M. Brzozowski : J. Physics D: Applied Physics (2014)
Mahi Singh. C. Rackner, M. Brzozowski: Physical Review A (2014)

- Peak height is enhanced in the presence of the metamaterial (DDI)
- Peak position changes with the unit cell size (shift in the SPP energy)

Tanaka et al. , PRL 105, 227403 (2010)

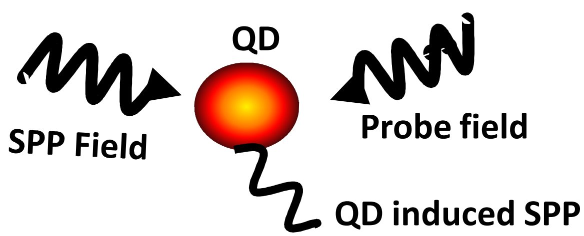


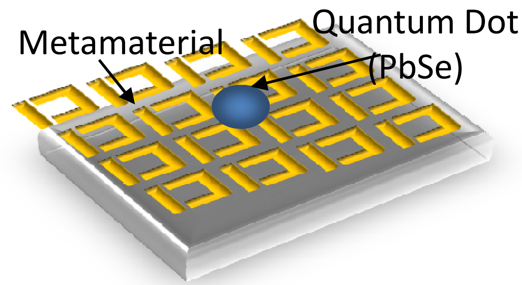
$$PL = QW_{abs}$$

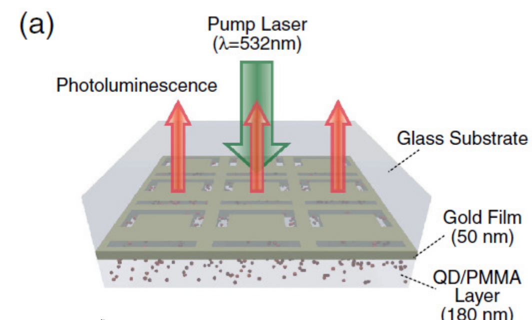
$$W_{abs} \rightarrow \left|\Omega_p + \Omega_{sp} + \Omega_{qd}\right|^2$$

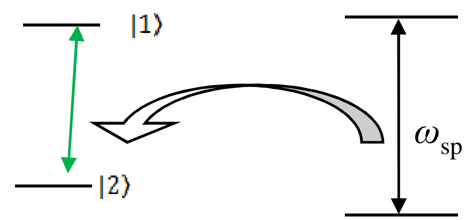
Q= quantum yield
W_abs = energy absorption rate

Ω_p = Probe field
Ω_{sp} = SPP field
Ω_{qd} = QD induced SPP









Conclusions

Hybrid Nanomaterials

- ❑Switching mechanism
- ❑Sensing mechanism
- ❑PL enahncement

Ane ke liye DHANYABAD

(Thanks for coming)

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