Title: One-photon and two-photon switching in graphene and optical nanomaterials

Date: Mar 08, 2013 02:30 PM

URL: http://www.pirsa.org/13030094

Abstract: In this talk we will discuss both the one-photon and two-photon switching mechanisms in hybrid nanomaterials made from two or more semiconductor, metallic and optical nanostructures. The most prominent examples of these nanostructures are graphene, semiconductor quantum dots, metallic nanoparticles, and photonic and polaritonic crystals. Advances in nanoscience have allowed for the construction of these new classes of hybrid nanomaterials. Optical excitations in semiconductor nanostructures are electron-hole pairs (excitons) whereas excitations in metallic nanostructures are surface plasmons which are collective oscillations of electrons. Therefore, the combination of these systems can provide attractive opportunities to modify, design and control optical properties and to observe new phenomena which are based on exciton-plasmon interactions. It is expected that this research will provide a theoretical road map for the development of optical sensing and optical switching.

Photon switching in nanomaterials

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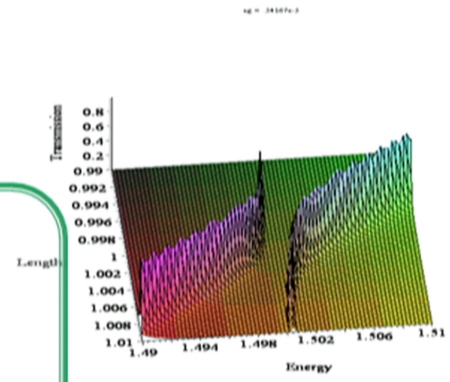
Department of Physics and Astronomy
Western University

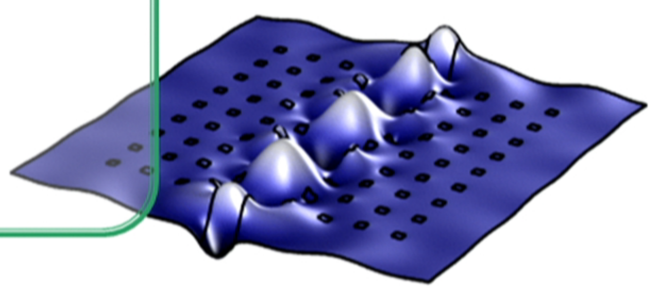
(The University of Western Ontario)
London, Canada

Perimeter Institute **(March 8, 2013)**

Outline

- ❑ Hybrid Nanomaterials
- ❑ Photonic materials
    - Polaritonic
    - Excitonic
    - Photonic crystals
    - Metamaterials
- ❑ One photon switching
- ❑ Two-photon switching
- ❑ Photoluminescence quenching
- ❑ Second harmonic generation
- ❑ Plasmonic heating
- ❑ Conclusion





Klaus von Klitzing

(Noble Prize 1986)

PMI, Stuttgart, Germany

Quantum Hall Effect

September 18 and 19, 2013

Western, London

Host: Mahi Singh

Collaborators

Graduate Students

J. Cox

A. Hatef

D. Schindel

C. Racknor

Ifte Haque

C. Tahani

Undergraduate Students

j. Richmond

J. Flannery

J. McKechnie

M. Bizozowski,

Scientists

Scott Barrie

Chandra Shekhar

S.P. Singh

International collaborations

UK

Y. Zhang

D. Birch

Y. Chen

Spain

M. Antón,

F. Carreño,

S. Melle,

O. Calderón,

E. Cabrera-Granado,

USA

S. Sadeghi

S. Pinnepalli

R. Agarwal

G. Gumbs

Argentina

C. Bildering

A. Bragas

Canadian collaborations

M. Zinke-Allmang

S. Mittler

Jai Sabrinathan

R. Lipson

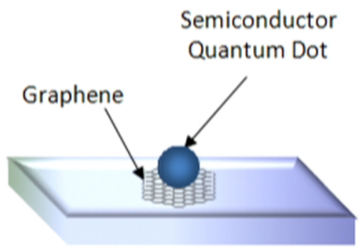
Hybrid Nanomaterials

Nanomaterials:

- **Semiconductor nanoparticles (quantum dots, nanocrystals, nanowires, etc.)**
- **Metallic nanoparticles (spheres, nanorods, nanodisks, etc.)**
- **Graphene and carbon nanotubes**
- **Molecular dyes, biological molecules (DNA, RNA, proteins)**
- **Metamaterials**

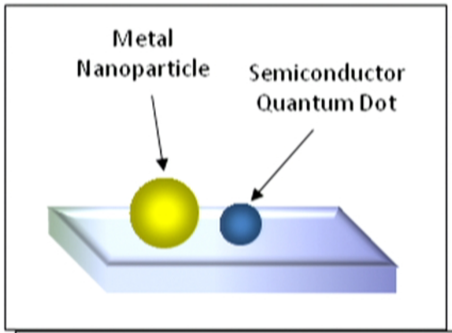
Substrate examples:

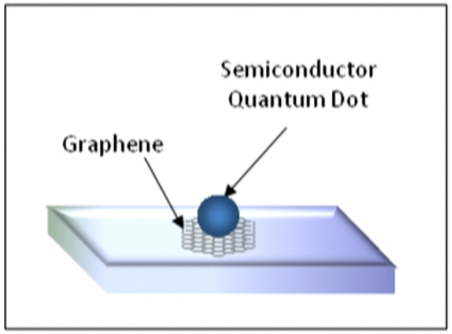
- ➢ **Metamaterials**
- ➢ **Dielectric material**
- ➢ **Excitonic material**
- ➢ **Polaritonic material**
- ➢ **Glass**
- ➢ **Photonic crystal**

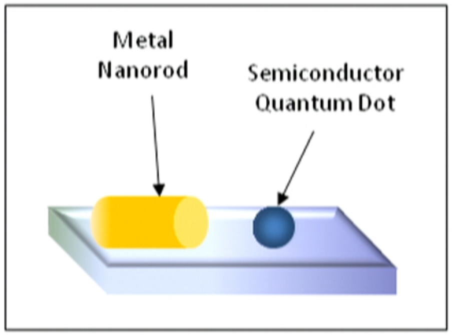


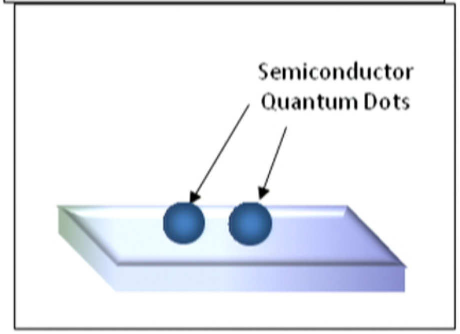
Hybrid Nanomaterials

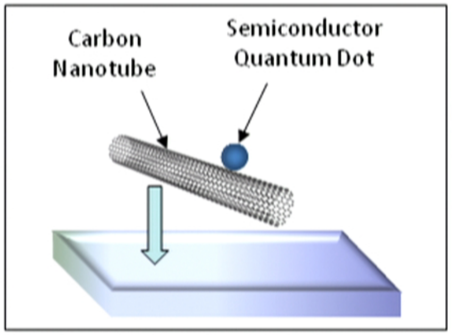
By using various combinations of nanostructures one can create enormous numbers of nanocomposite (hybrid) materials.

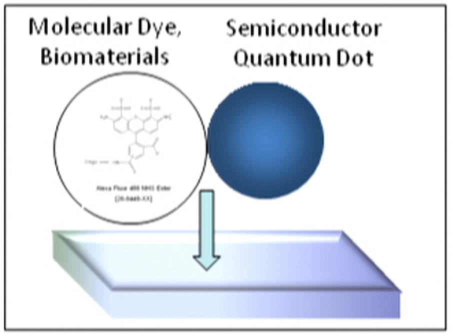


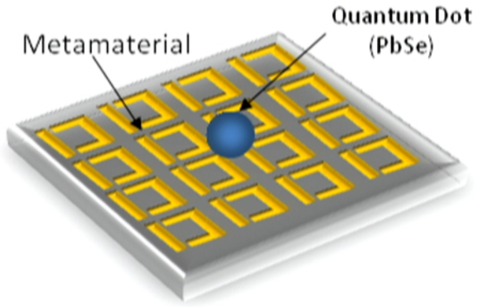






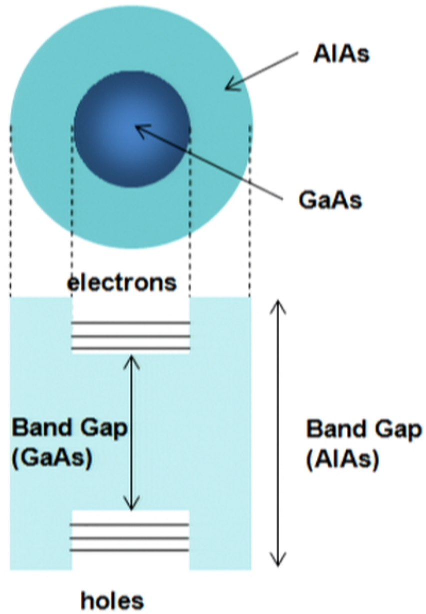


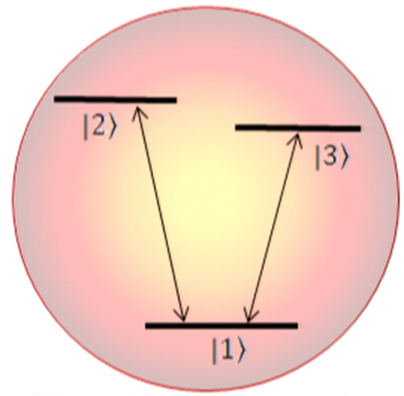




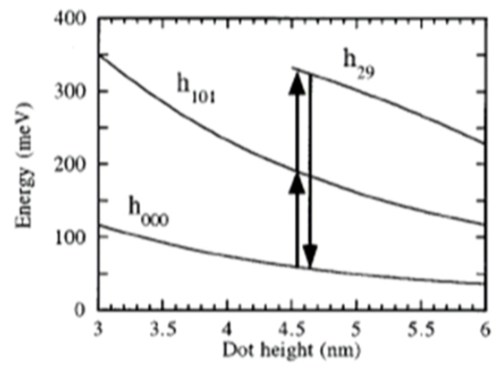
Quantum Dot: excitons

- Examples: CdSe/ZnS; GaAs/InAs; GaAs/AlAs; CdS
- Electron-hole pair are called excitons.
- Excitons energy depends on the size of quantum dots





Three-level quantum dot



T. Brunhes et al., *Appl. Phys. Lett.* 75, 835 (1999).

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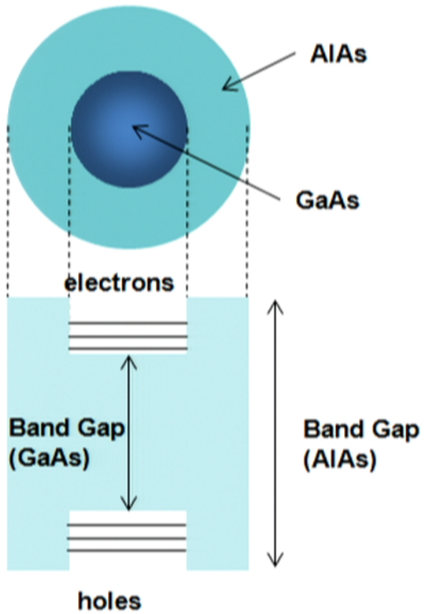
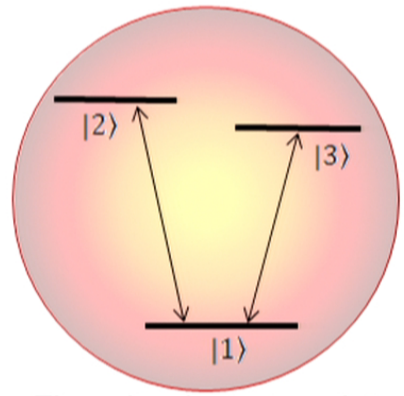
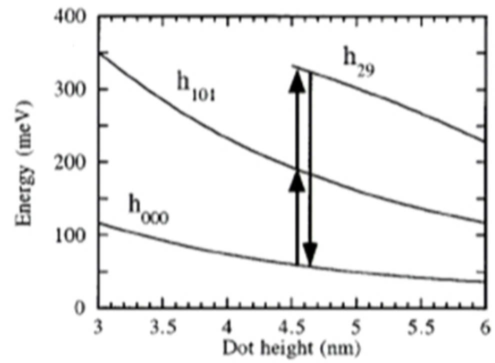




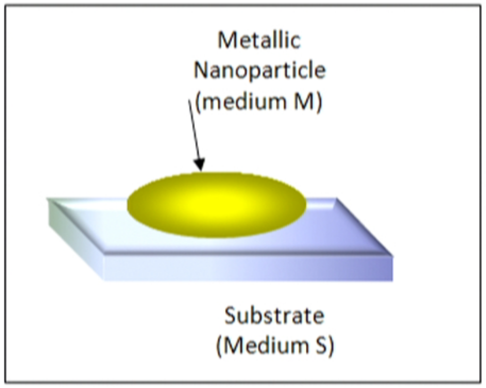
Three-level quantum dot



T. Brunhes et al., *Appl. Phys. Lett.* 75, 835 (1999).

Metallic Nanostructures (MNP)
Bound surface plasmon polaritons

Mahi Singh et al.,
Applied Physics Letters 99, 181106 (2011)



- Metallic nanoparticles have free electrons. at the surface.
- Electromagnetic field interacts with plasmons of the metallic nanoparticle
- The dielectric constant of metals can be NEGATIVE

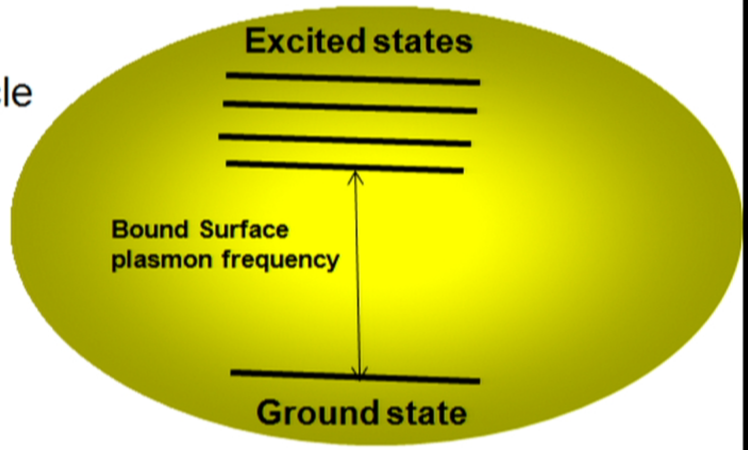
$$\epsilon_m(\omega) = 1 - \frac{\omega_p^2}{\omega^2}$$

- Electric field produced by the nanoparticle

$$E \approx -\left(\frac{\epsilon_m(\omega) - \epsilon_s}{\epsilon_s + \varsigma_{size}\left[3\epsilon_s + 3\epsilon_m(\omega)\right]}\right) E_{applied}$$

- The polarizability of the nanoparticle gives bound surface plasmons by putting it equals to zero

$$\alpha(\omega) \approx \left(\frac{\epsilon_m(\omega) - \epsilon_s}{\epsilon_s + \varsigma_{size}\left[3\epsilon_s + 3\epsilon_m(\omega)\right]}\right)$$



MNP:
Graphene and carbon nanotubes

Graphene was invented by a Canadian physicist

(1915-2006)
P.R. Wallace
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 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 and I like it and he is my favorite student.

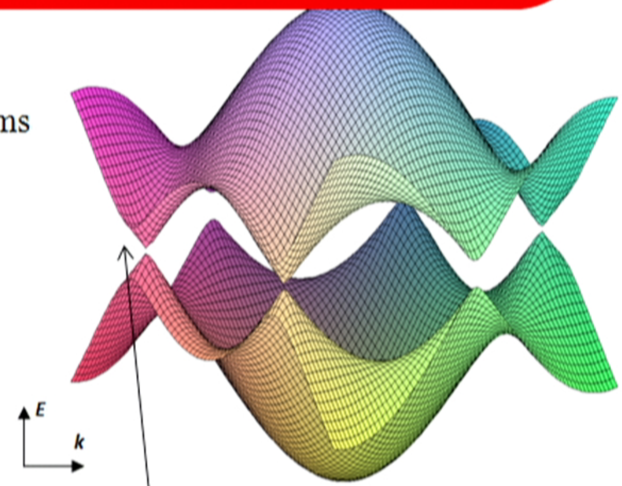
Graphene Band Structure

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

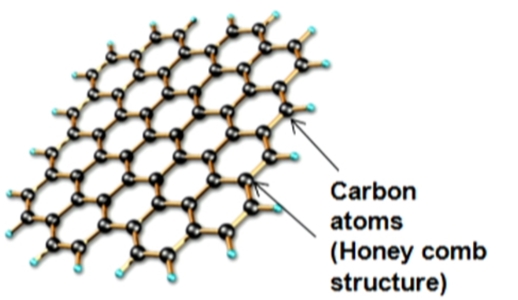
- Graphene is a two dimensional sheet of carbon atoms which are arranged in the honey comb structure.
- Gapless semiconductor and acts as a metal.

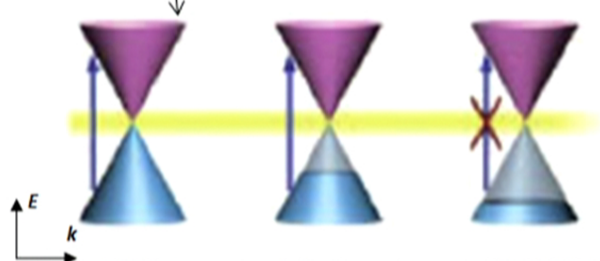
Band structure:

- k.p method
- Tight binding method



Mahi R. Singh, Introduction to Nanomaterials (Western Book Store, London, Canada 2012)



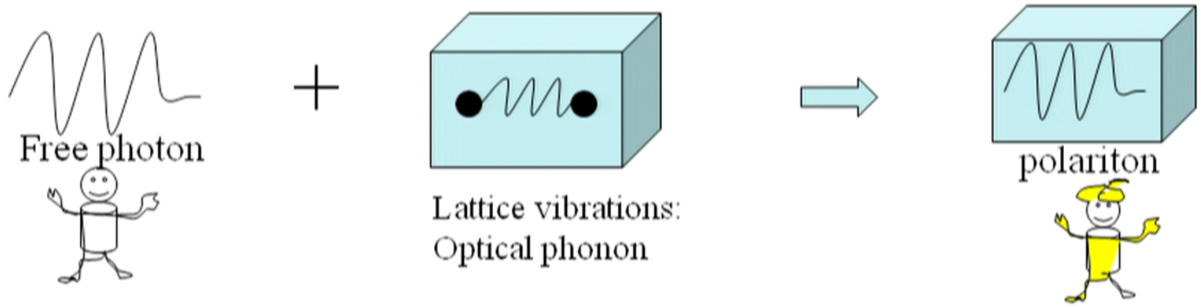


A.N. Grigorenko et al, Nature Photonics 6, 749 (2012)

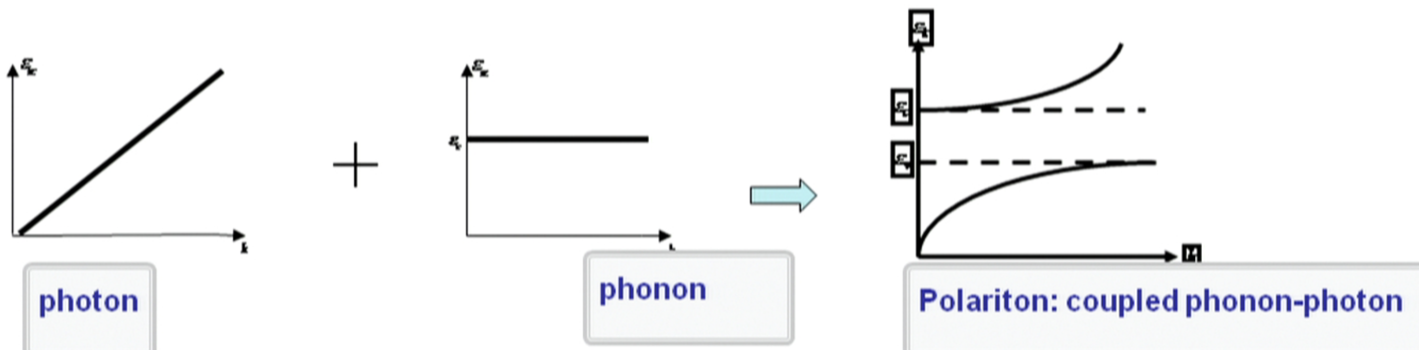
Polaritonic Materials

V. Rupasov and Mahi Singh: Phys Rev. Lett. (1997)

Example: Semiconductors (SiC, GaAs), oxides (MgO), ionic materials (LiF)



Dispersion Relation



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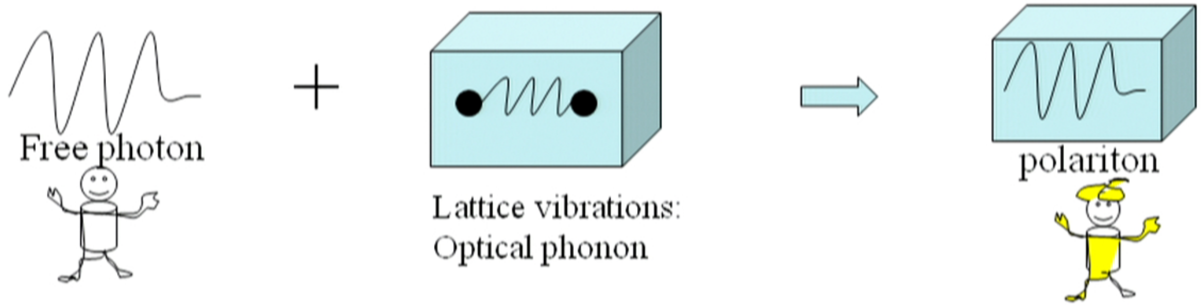

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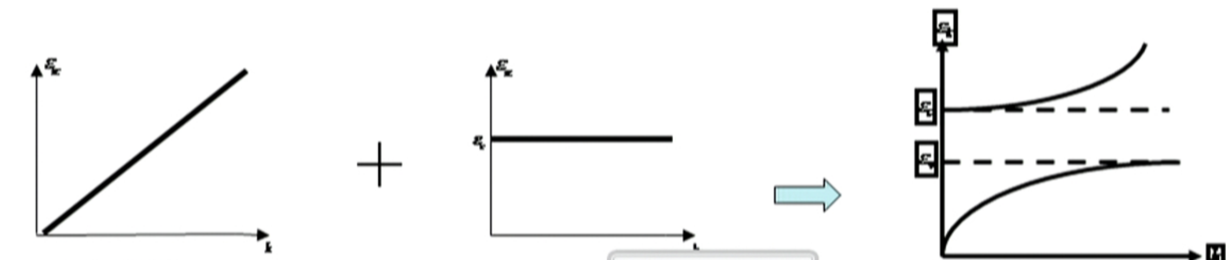


photon

phonon

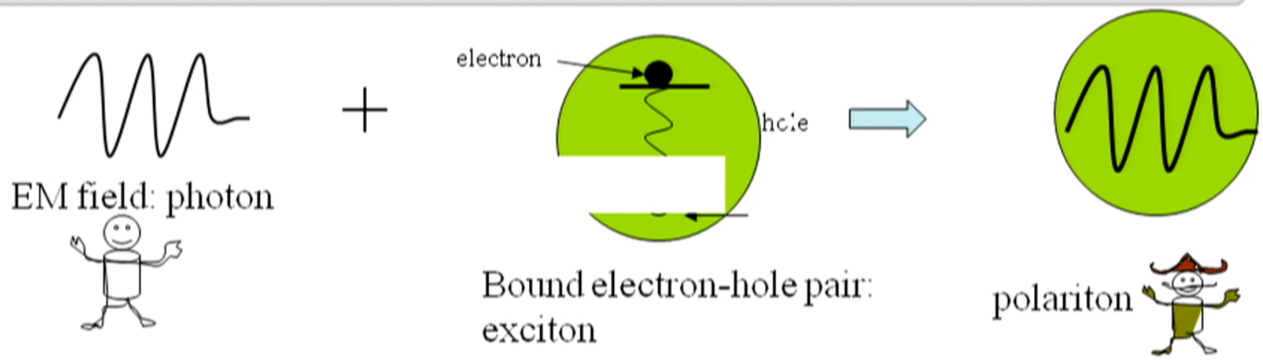
Polariton: coupled phonon-photon

Excitonic Materials

Cox, Singh and Racknor, **Nano Lett. 11, 5284 (2011)**

EXCITONS: Bound electron-hole pair

Example: Nanostructures : graphene, quantum dots, quantum wires, quantum wells



Excitonics
Example: GaN- Quantum Nanowire

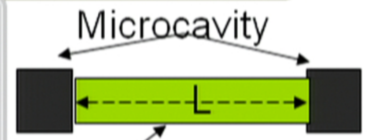
Mahi Singh: (Unpublished -2008)

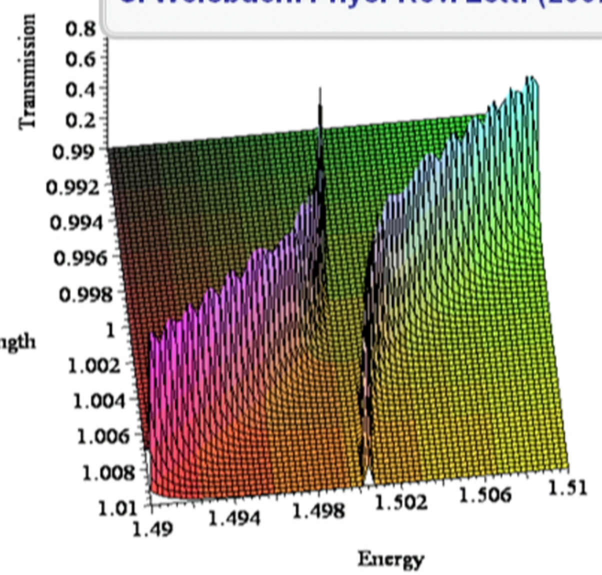
Transmission coefficient:
Excitonic Polariton

GaN

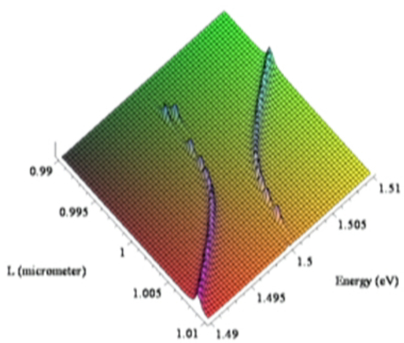
$\varepsilon_v = 1.5eV$
$\Delta = 10\mu eV$
$finesse = 30$

C. Weisbuch: Phys. Rev. Lett. (2007)



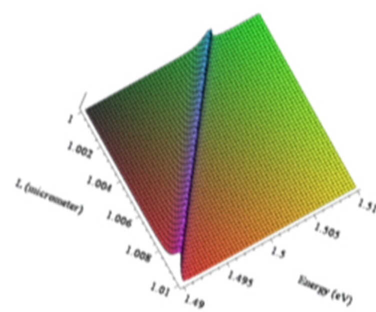


Exciton-polariton



Zero gap

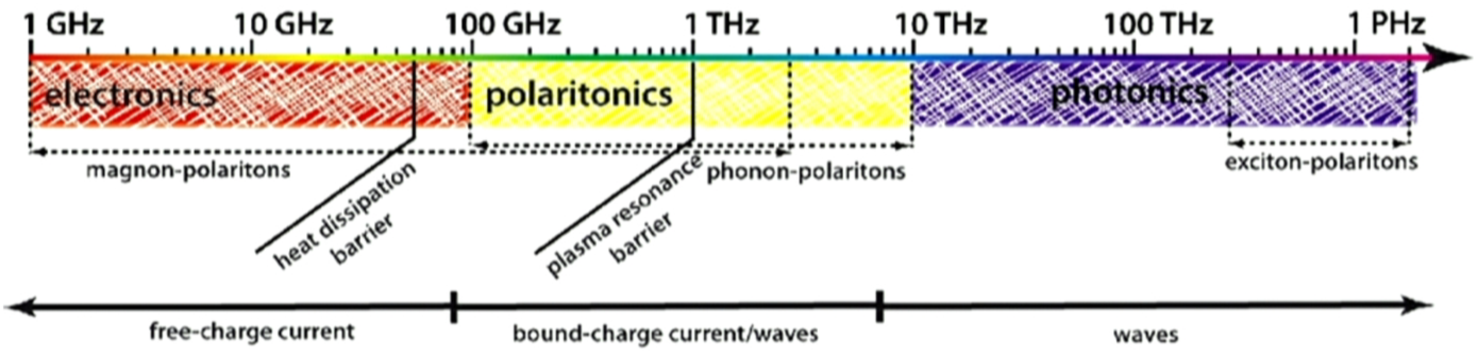
photon



Applications: Polaritonic Materials

C. Racknor and 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.**



Photonic Crystals

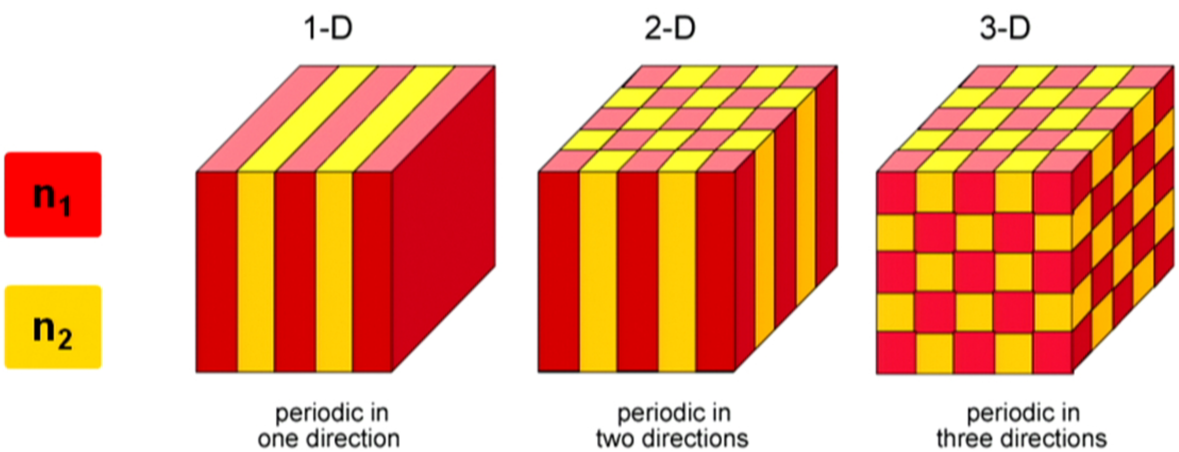
E. Yablonovitch, PRL 58, 2059 1987

S. John, PRL 58, 2486 1987

- Periodic dielectric lattice
- Periodic dielectric constant function

- 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

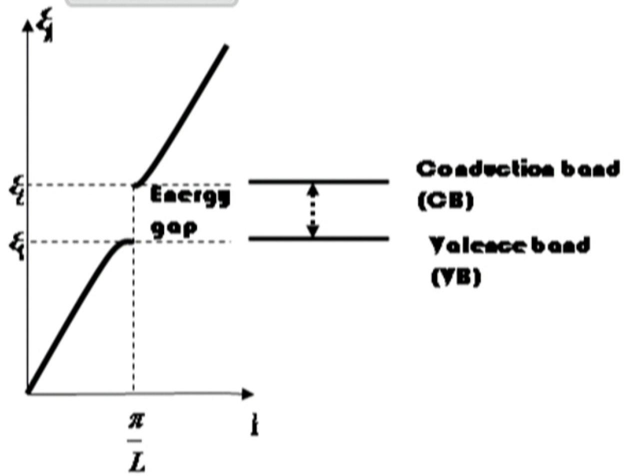
Photonic Materials: Dielectrics

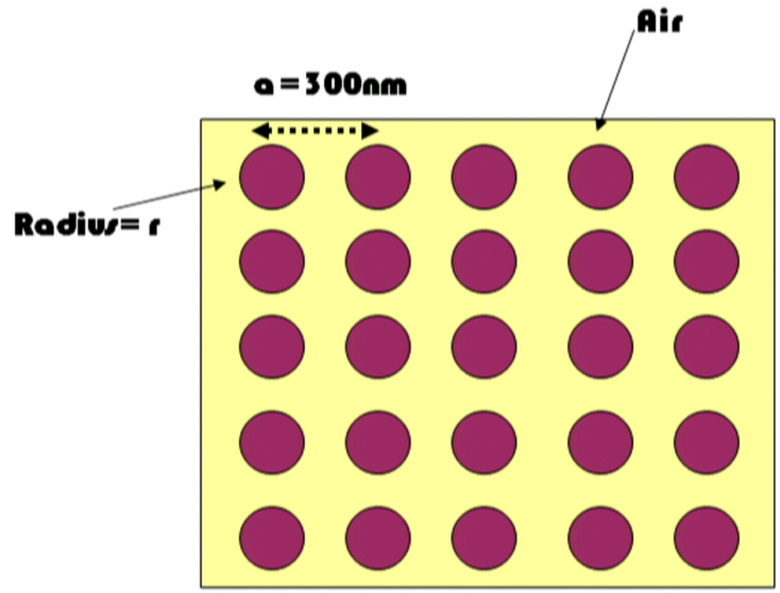
I. Haque and Mahi Singh. J. Phys. Condens. Matter 19, 156229 (2007)

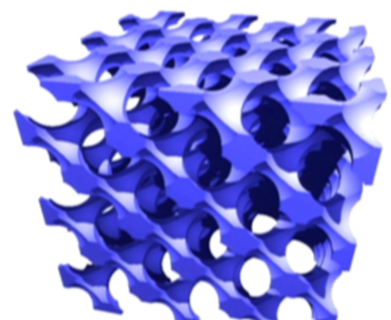
Dielectric spheres (Si) in air

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.

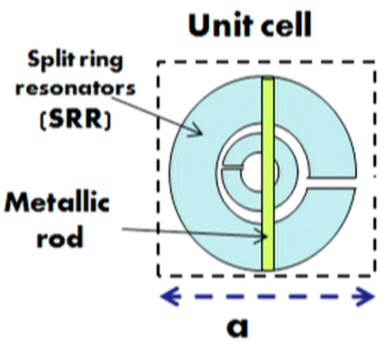
Source: S. John, O. Toader and K. Busch, *PBG Materials: A Semiconductor for Light*, 2002.

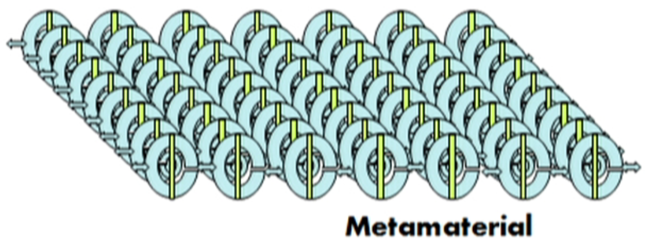
Metamaterials

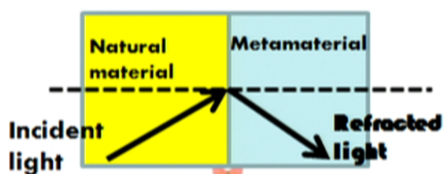
Singh. Cox, Rackner, Marek : Applied Physics Letters (2013)

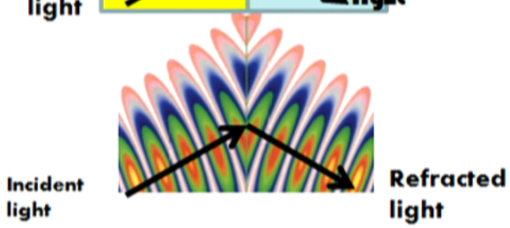
Artificial materials made from negative dielectric constant an negative magnetic permeability

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









Snell's law

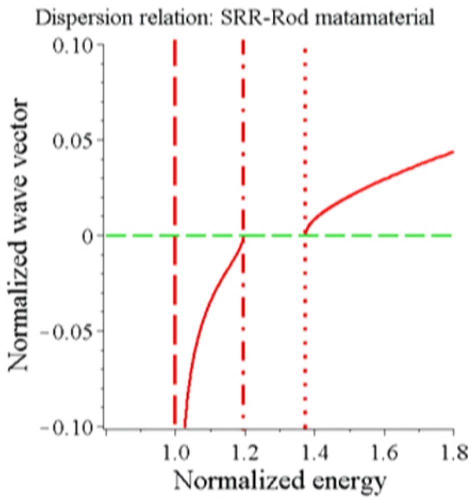
Metamaterials

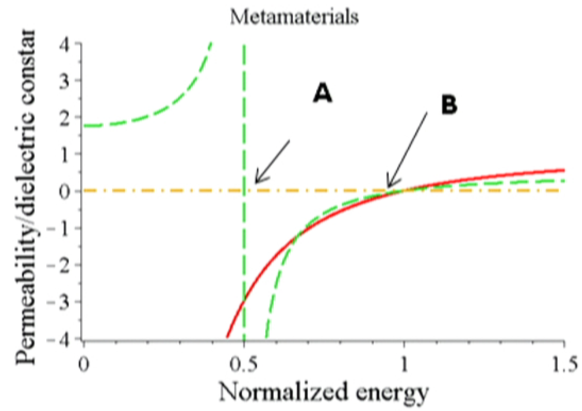
Singh. Cox, Rackner, Marek : Applied Physics Letters (2013)

Note: ϵ and μ are negative between A and B

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

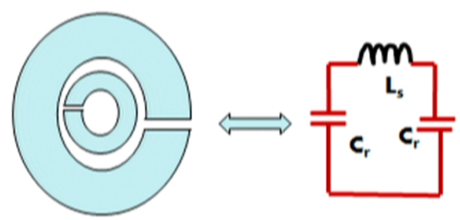
$$\mu_m(\omega) \approx \left(1 - \frac{F\omega^2}{\omega^2 - \omega_0^2}\right)$$





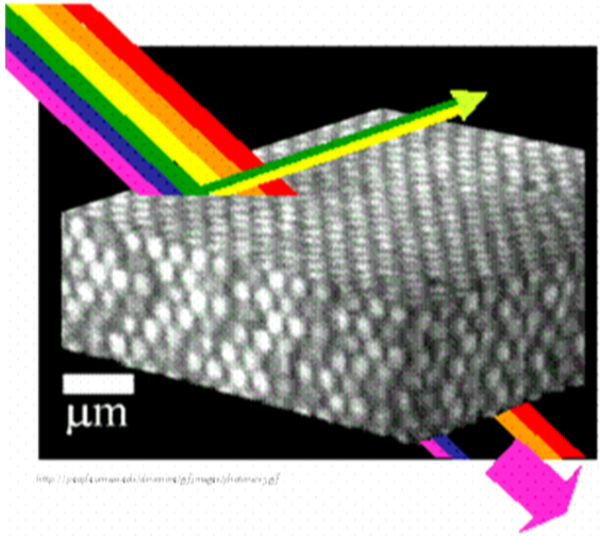
Band gap is found

Band structure: Transmission line theory



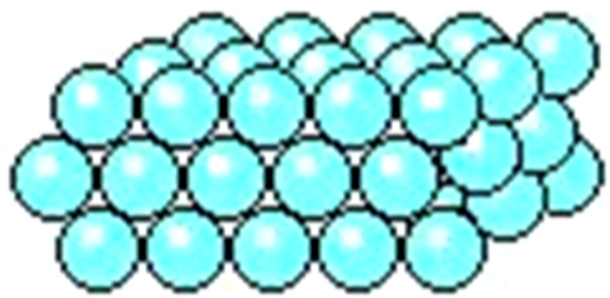
SRR–RLC circuit

Photonic Materials



Reflected Colours
-Yellow
-Green

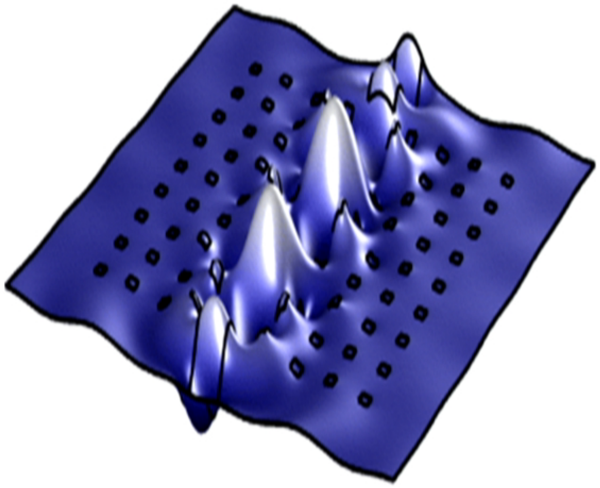
Transmitted Colours
-Purple
-Orange
-Red
-Blue

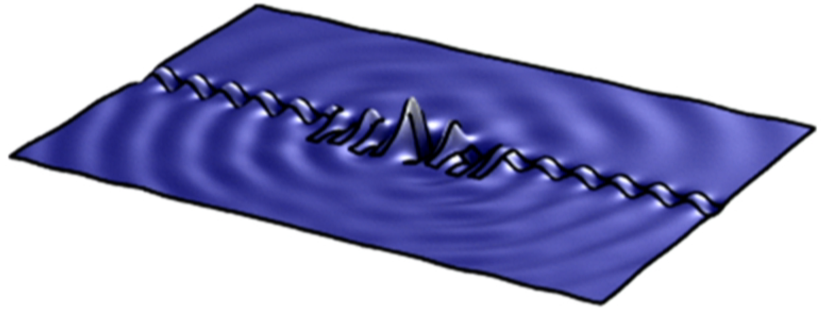


Applications;
Photonic Band gap Materials

- **Photons are confined in a nanosize material**

Photons are confined in 2-d





90º Light Splitter

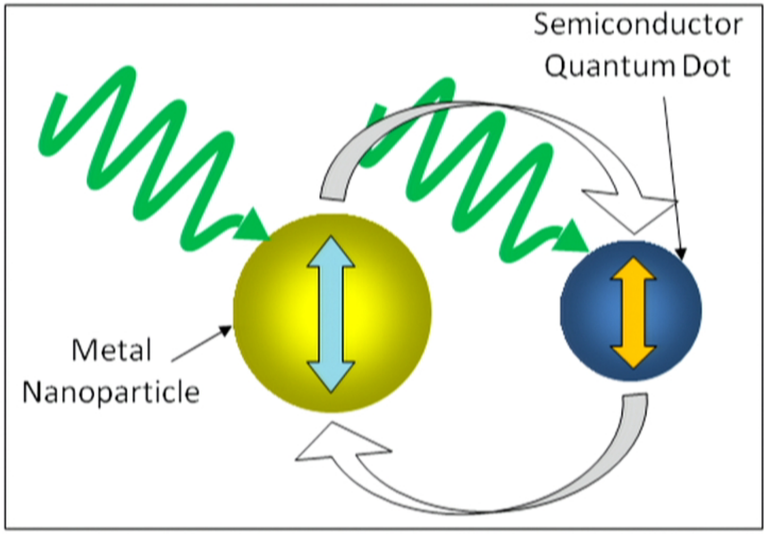
Dipole-Dipole Interaction

➢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)

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



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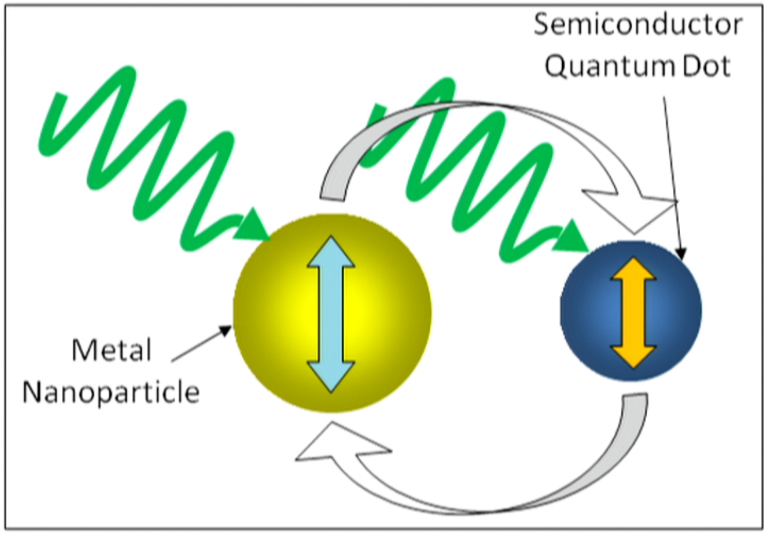
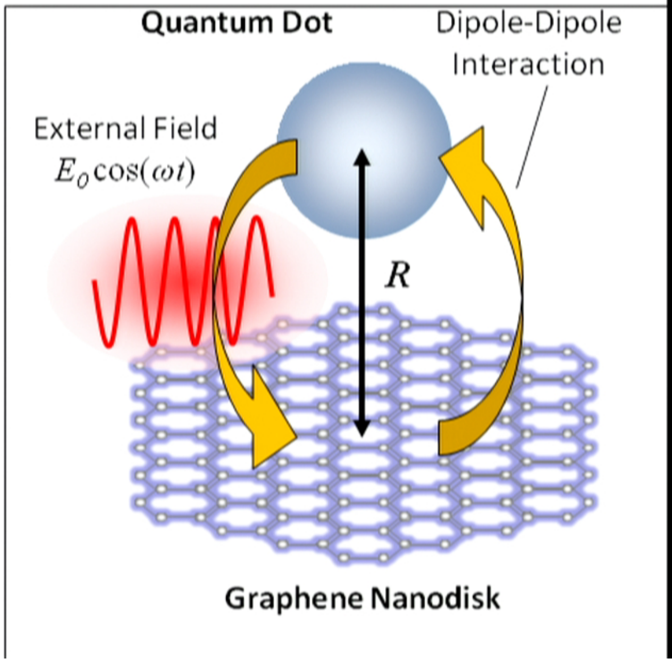
Hatef and Singh, Nanotechnology 23, 205203 (2012)

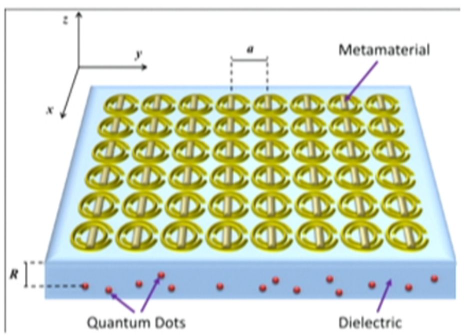
$$E_{dip} \approx \frac{P_{MNP}}{R^3}$$

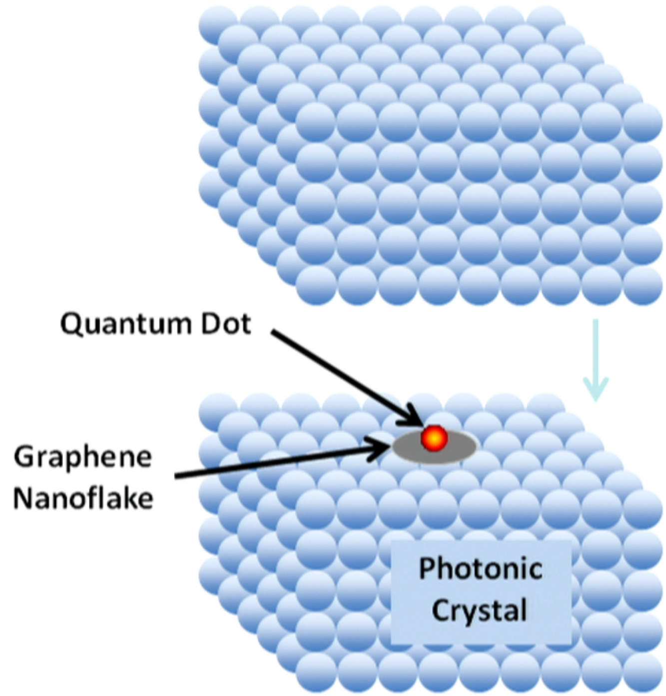




Graphene/Quantum Dot Hybrid deposited on a Photonic Crystal /Metamaterials

- 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





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

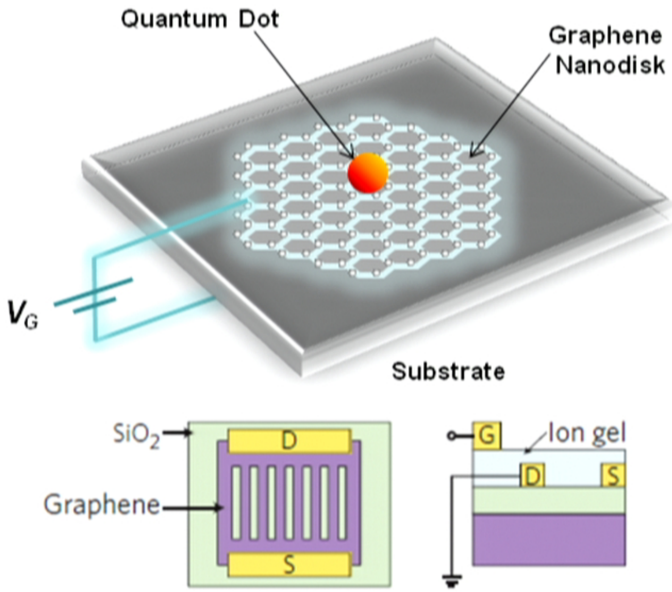
Graphene: Bound Surface Plasmons

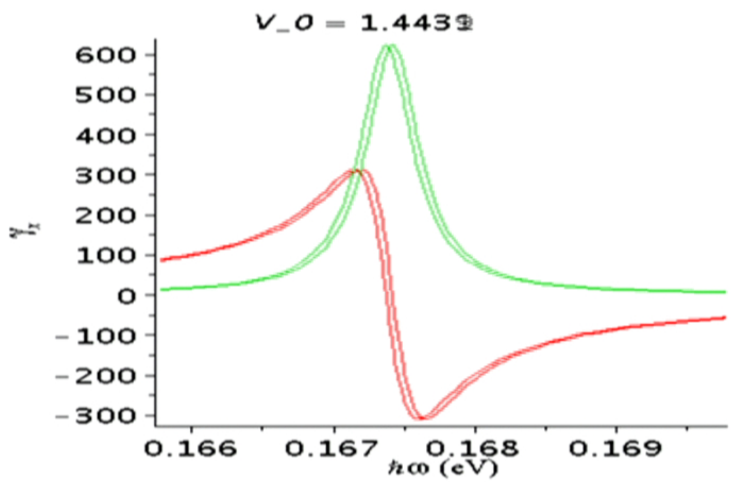
J. Cox and Mahi Singh, Nanotechnology (2013)

Gate voltage
Plasmon frequency can be changed by applying the gate voltage

$$\alpha \approx -\left(\frac{\epsilon_m(\omega) - \epsilon_s}{\epsilon_s + \varsigma_{size}\left[3\epsilon_s + 3\epsilon_m(\omega)\right]}\right)$$

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





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

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J. Cox and Mahi Singh, Nanotechnology (2013)

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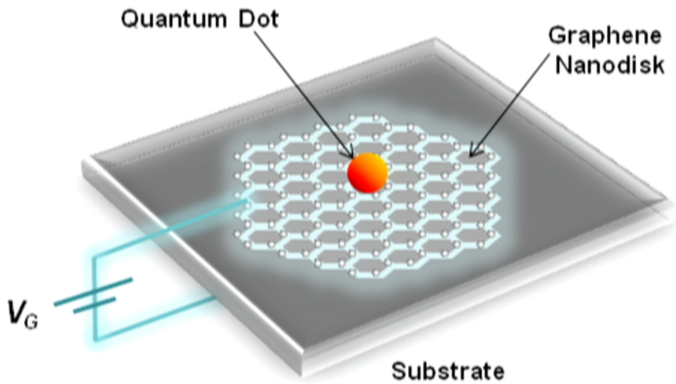
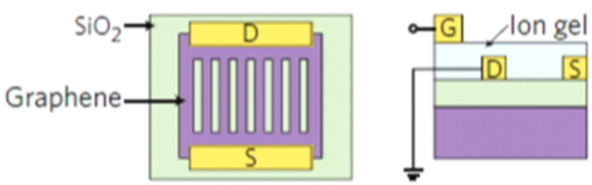
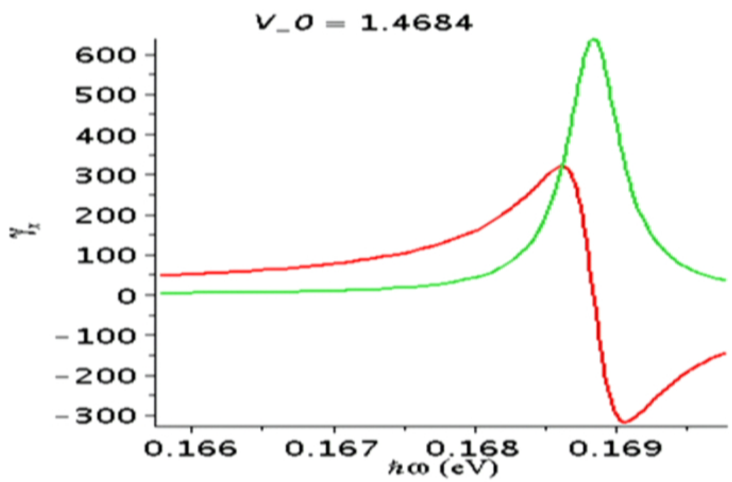






Parameters: Radius= 8 nm; v_F = c/300; μ = 10000 $cm^2V^{-1}s^{-1}$; ε_b = 10.89 (GaAs)

Theoretical Formalism

- Hamiltonian

$$H_{Total} = \begin{bmatrix} H_{QD} + H_{MNP} + H_{laser} \\ + H_{QD\_MNP} + H_{QD\_laser} + H_{MNP\_laser} \\ + H_{QD\_substrate} + H_{MNP\_lsubstrate} \end{bmatrix}$$

- Maxwell equation
- Schrödinger equation
- Density matrix method + Greens function method

$$\frac{d\rho_{22}}{dt} = 2\Gamma_{21}\rho_{11} - iR_2 e^{i\theta_2}\rho_{21} - i\Lambda_2\rho_{12}\rho_{21} + iR_2 e^{-i\theta_2}\rho_{12} + i\Lambda_2^*\rho_{21}\rho_{12}$$

$$\frac{d\rho_{33}}{dt} = 2\Gamma_{31}\rho_{11} - iR_3 e^{i\theta_3}\rho_{31} - i\Lambda_3\rho_{13}\rho_{31} + iR_3 e^{-i\theta_3}\rho_{13} + i\Lambda_3^*\rho_{31}\rho_{13}$$

$$\frac{d\rho_{12}}{dt} = -d_{12}\rho_{12} + iR_3 e^{i\theta_3}\rho_{32} + i\Lambda_3\rho_{13}\rho_{32} - iR_2 e^{-i\theta_2}(\rho_{11} - \rho_{22})$$

$$\frac{d\rho_{13}}{dt} = -d_{13}\rho_{13} + iR_2 e^{i\theta_2}\rho_{23} + i\Lambda_2\rho_{12}\rho_{23} - iR_3 e^{i\theta_3}(\rho_{11} - \rho_{33})$$

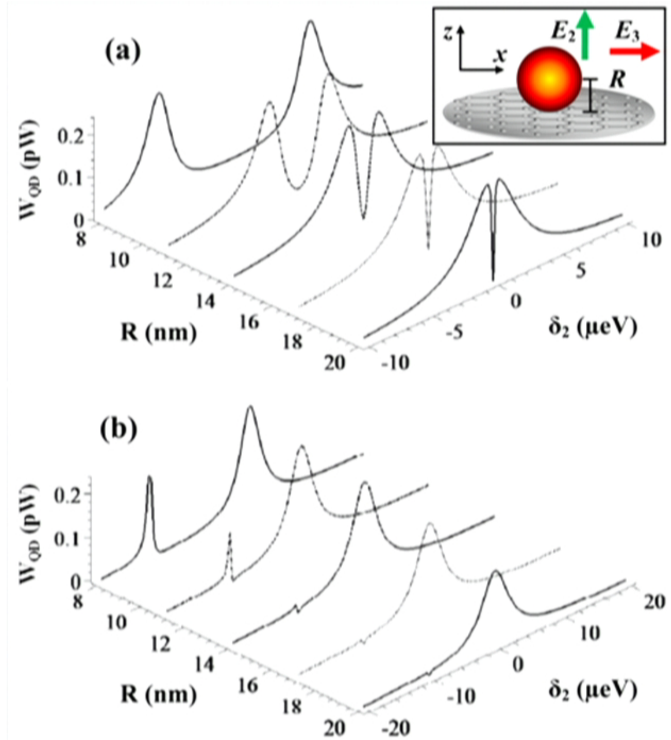
$$\frac{d\rho_{23}}{dt} = -i(\delta_2 - \delta_3)\rho_{23} + iR_2 e^{-i\theta_2}\rho_{13} + i\Lambda_2^*\rho_{21}\rho_{31} - iR_3 e^{i\theta_3}\rho_{21} - i\Lambda_3\rho_{13}\rho_{21}$$

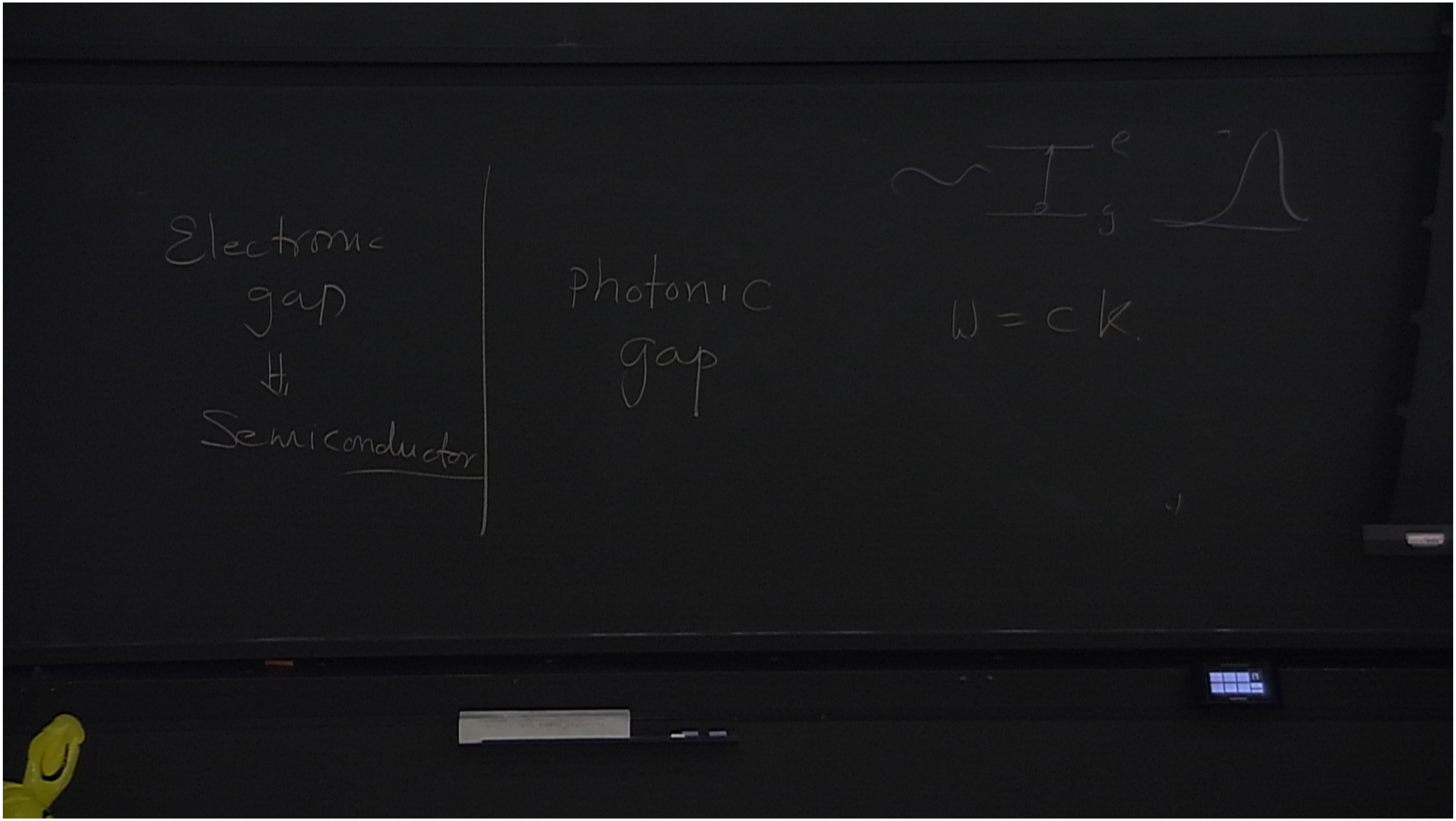
$$W_{QD} = \sum_{i=2,3} \hbar\omega_i\rho_{ii}\Gamma_{i1}$$

$$W_G = \sum_{i=2,3} \frac{g_{x,z}^2 V \mu_{1i}^2 \omega_i \,\mathrm{Im}(\gamma_{x,z})\, |\tilde{\rho}_{1i}|^2}{2\pi\epsilon_b\epsilon_{dr}^2 |\epsilon_{bg}|^2 R^6}$$

Power Absorption in QD

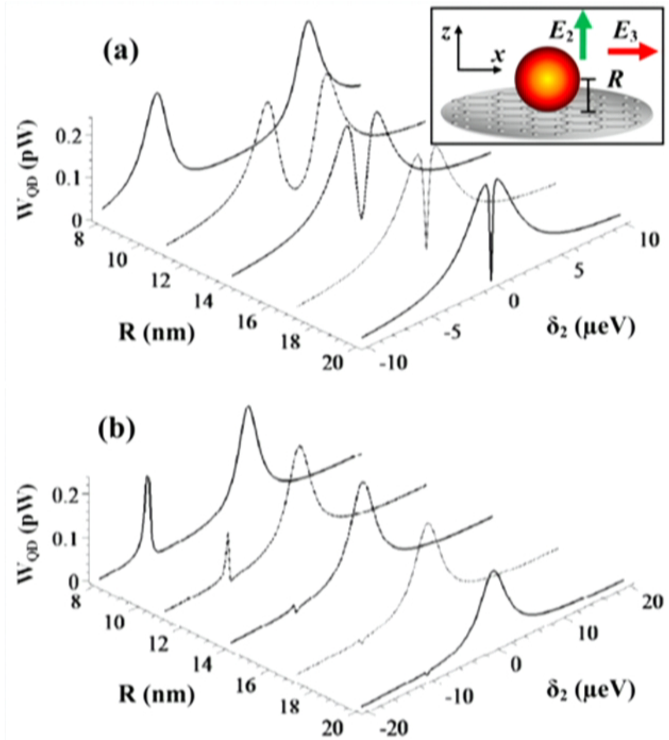
- Here the power absorbed by the QD is calculated while varying the graphene-QD separation R
- Narrow minima for larger values of R is due to electromagnetically induced transparency
- For small values of R, the spectrum splits into two peaks due to the DDI
- Power is absorbed by the QD at two frequencies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




Electronic
gap
Semiconductor
Photonic
gap
e
g
$\omega = ck$

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



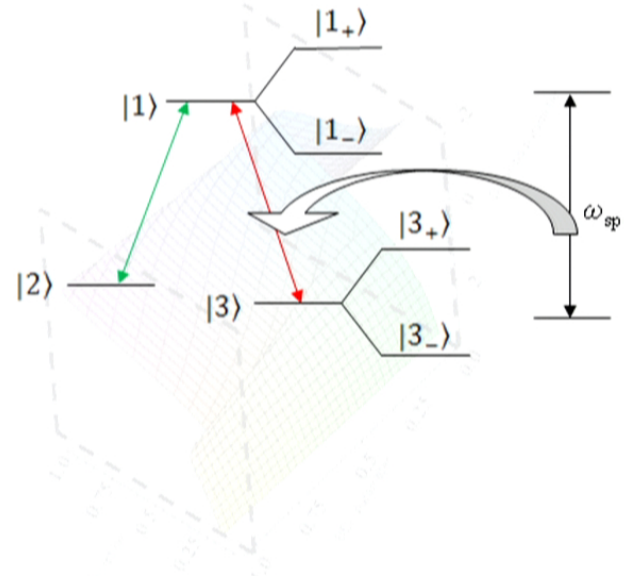
Switching Mechanism: One-photon spectroscopy

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

ABSTRACTION:

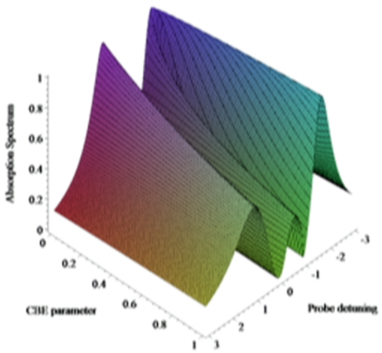
ABSORPTION:

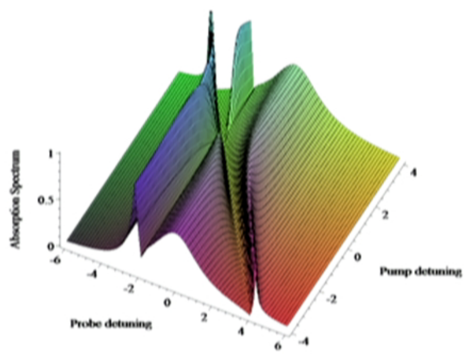
- transition from state from 2 to 1
- DDI: changes with R
- DDI: Gate voltage

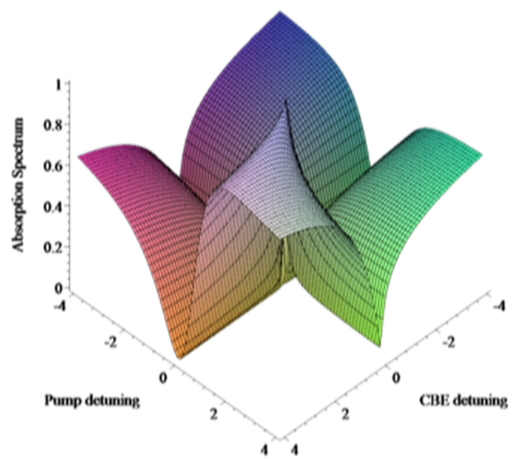


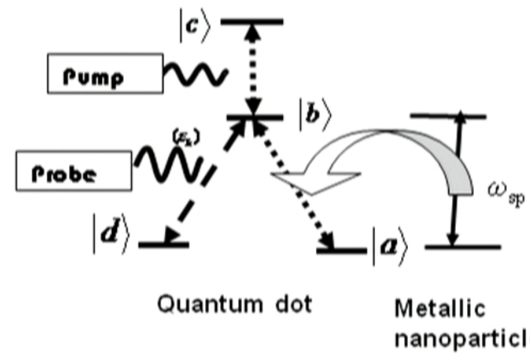
One-Photon Process
DDI + Pump Field Effects

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).









PHYSICAL REVIEW WEBSITE

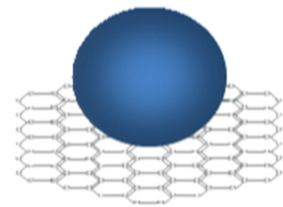
Main Results: The system switches from one transparent state to two transparent states

Sensing Mechanism: Graphene-QD hybrid

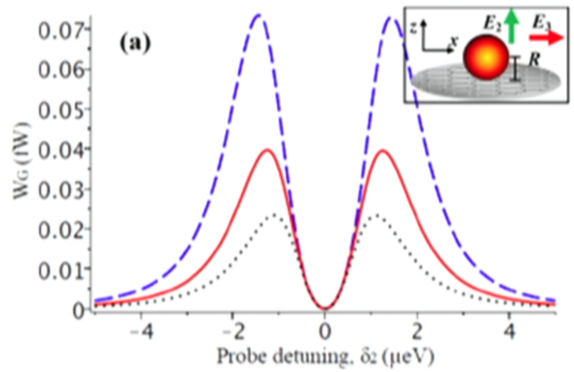
➢When a QD is in contact with biomolecules, molecular beacons, DNA or aptamers, its dielectric constant can be modified.

➢ 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).



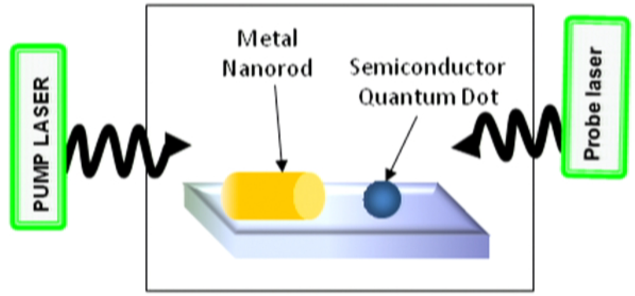
- ε_{qd} = 10 (dotted curve)
- ε_{qd} = 12 (solid curve)
- ε_{qd} = 14 (dashed curve)
- Energy transfer to graphene when the QD dielectric constant is changed
  - Detection of biological molecules



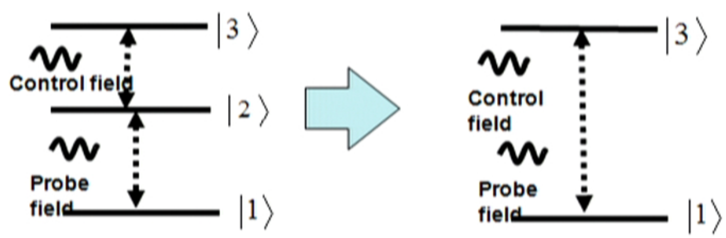
Cox and Singh, Physical Review B 86, 125452 (2012);
Singh, Racknor and Schindel App. Phys. Lett. 101, 051115 (2012)

Two-Photon Spectroscopy: *QD+MNP*

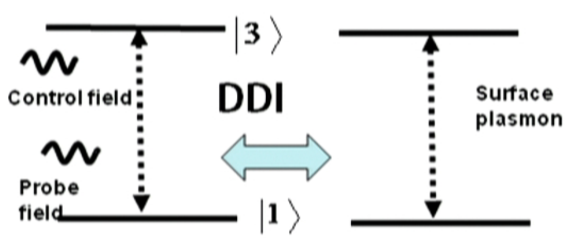
Mahi Singh, Nanotechnology (2012)
J.Cox, Singh, Bildering, Bragas, Advanced Materials (2013)
Racknor[1], Zhang, Birch[2], Chen, Appl. Phys. Lett. (2013)



Aim: Two-photon transition 3-level QD

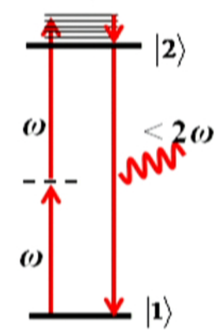


Quantum dot+ metallic nanorod



Two-photon transition in 2-level QD

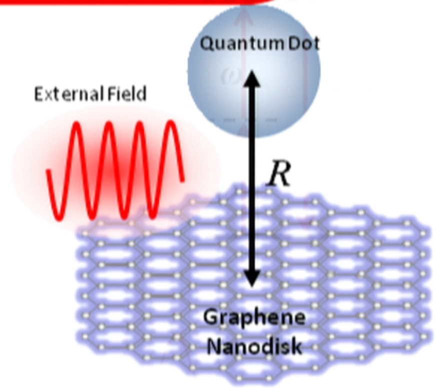
Quantum dot (2-level)

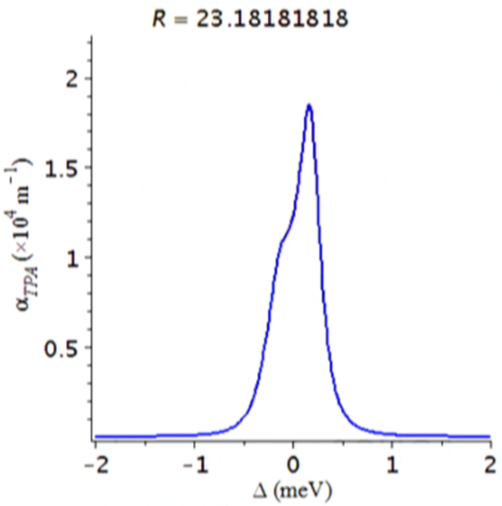


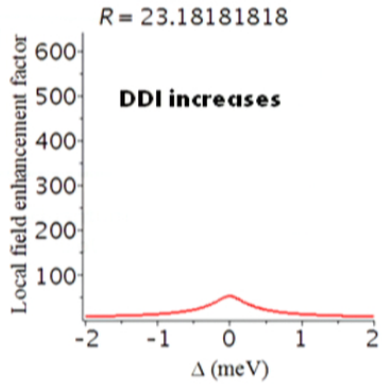
Two-photon absorption

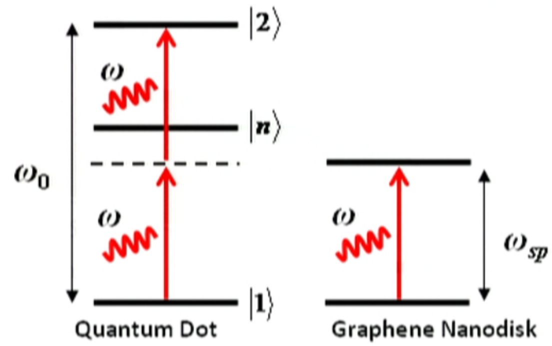
Two-Photon Process: QD-Graphene
DDI splitting

- 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









J. Cox, M. Singh, (2013))

Conclusions

Hybrid Nanomaterials

- Switching mechanism
- Sensing mechanism
- Plasmonic heating

Ane ke liye DHANYABAD