



Plasmonics: Experiment, theory and applications

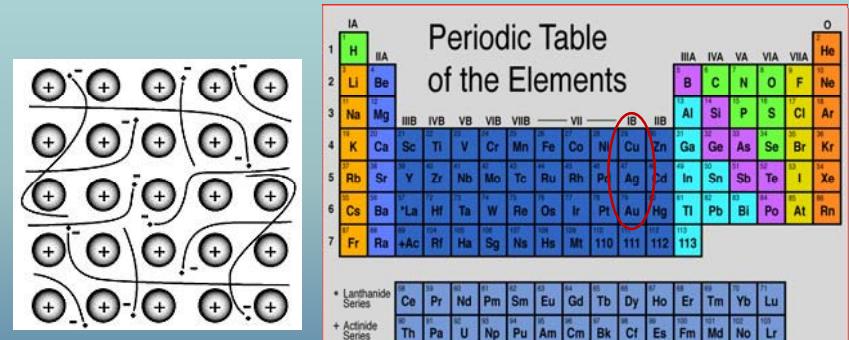
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Plasmonics: photonics with free charges

- A “sea” of free electrons in a background of rigid positive ions



- Best conductors:** noble metals Cu, Ag and Au
 - large electron charge density (about 1 free electron per atom)

$$N = \frac{\rho N_A}{A} \approx 10^{23} \text{ el/cm}^3$$



Drude theory of metals: DC

- Under the action of a constant external electric field
 - electrons get accelerated

$$\mathbf{F} = -e\mathbf{E}$$

- if the average time between collisions is τ , then

$$\langle \mathbf{p} \rangle = \mathbf{F} \tau \Rightarrow \langle \mathbf{v} \rangle = \frac{\langle \mathbf{p} \rangle}{m} = -\frac{e\mathbf{E}}{m} \tau$$

- this corresponds to a net current density

$$\mathbf{J} = -Ne \langle \mathbf{v} \rangle = \left(\frac{Ne^2 \tau}{m} \right) \mathbf{E}$$

- and thus to a DC conductivity

$$\mathbf{J} = \sigma_0 \mathbf{E} \Rightarrow \sigma_0 = \frac{Ne^2 \tau}{m}$$



Drude theory of metals: AC

- Under the action of an oscillating external electric field
 - within a time interval dt electrons get accelerated by the field, but also loose speed due to collisions

$$\frac{d \langle \mathbf{v} \rangle}{dt} = -\frac{\langle \mathbf{v} \rangle}{\tau} - \frac{e\mathbf{E}}{m}$$

- assume a harmonic time dependence for both \mathbf{E} and $\langle \mathbf{v} \rangle \sim e^{-i\omega t}$

$$-i\omega \langle \mathbf{v} \rangle = -\frac{\langle \mathbf{v} \rangle}{\tau} - \frac{e\mathbf{E}}{m}$$

$$\langle \mathbf{v} \rangle = -\frac{e\tau}{m(1-i\omega\tau)} \mathbf{E}$$

- resulting into the AC conductivity

$$\mathbf{J} = -Ne \langle \mathbf{v} \rangle = \sigma \mathbf{E} \Rightarrow \sigma(\omega) = \frac{\sigma_0}{1-i\omega\tau}, \quad \sigma_0 = \frac{Ne^2 \tau}{m}$$



Metal dielectric function

- Maxwell's equation for non-magnetic conductive media

$$\nabla \times \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{H}}{\partial t} \quad \nabla \times \mathbf{H} = \frac{4\pi}{c} \mathbf{J} + \frac{1}{c} \frac{\partial \mathbf{E}}{\partial t}$$

- Combine the two $\nabla \times (\nabla \times \mathbf{E}) = \nabla \times \left(-\frac{1}{c} \frac{\partial \mathbf{H}}{\partial t} \right)$

$$\nabla \times (\nabla \times \mathbf{E}) = -\frac{1}{c} \frac{\partial (\nabla \times \mathbf{H})}{\partial t}$$

$$\nabla \times (\nabla \times \mathbf{E}) = -\frac{1}{c} \frac{\partial}{\partial t} \left(\frac{4\pi}{c} \mathbf{J} + \frac{1}{c} \frac{\partial \mathbf{E}}{\partial t} \right)$$

$$\nabla \times (\nabla \times \mathbf{E}) = -\frac{4\pi}{c^2} \frac{\partial \mathbf{J}}{\partial t} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2}$$



Metal dielectric function

$$\nabla \times (\nabla \times \mathbf{E}) = -\frac{4\pi}{c^2} \frac{\partial \mathbf{J}}{\partial t} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2}$$

- Use the vector identity

$$\nabla \times (\nabla \times \mathbf{E}) = \nabla \cdot (\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E}$$

and due to Maxwell's first equation $\nabla \cdot \mathbf{E} = 0$

- so the wave equation $\nabla^2 \mathbf{E} = \frac{4\pi}{c^2} \frac{\partial \mathbf{J}}{\partial t} + \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2}$

- use the fact $\mathbf{E}, \mathbf{J} \sim e^{-i\omega t}$ and $\mathbf{J} = \sigma \mathbf{E}$

$$\frac{\partial \mathbf{J}}{\partial t} = -i\omega \mathbf{J} = -i\omega \sigma \mathbf{E} \quad \frac{\partial^2 \mathbf{E}}{\partial t^2} = -\omega^2 \mathbf{E}$$

$$\nabla^2 \mathbf{E} = -i\omega \frac{4\pi\sigma}{c^2} \mathbf{E} - \frac{\omega^2}{c^2} \mathbf{E} \quad \Rightarrow \nabla^2 \mathbf{E} + \frac{\omega^2}{c^2} \left(1 + i \frac{4\pi\sigma}{\omega} \right) \mathbf{E} = 0$$



Metal dielectric function

- Thus the metal dielectric function is

$$\Rightarrow \nabla^2 \mathbf{E} + \frac{\omega^2}{c^2} \underbrace{\left(1 + i \frac{4\pi\sigma}{\omega} \right)}_{\varepsilon(\omega)} \mathbf{E} = 0$$

$$\varepsilon(\omega) = 1 + i \frac{4\pi\sigma(\omega)}{\omega}$$

- so the wave equation $\nabla^2 \mathbf{E} + \frac{\omega^2 \varepsilon(\omega)}{c^2} \mathbf{E} = 0$

- if we substitute for $\sigma(\omega) = \frac{Ne^2\tau}{m} \frac{1}{1-i\omega\tau}$ $\omega_p = \sqrt{\frac{4\pi Ne^2}{m}}$

$$\varepsilon(\omega) = 1 + i \frac{4\pi}{\omega} \frac{Ne^2\tau}{m} \frac{1}{1-i\omega\tau} = 1 - \frac{4\pi Ne^2}{m} \frac{1}{\omega^2 + i\omega/\tau}$$



Metal dielectric function

- Back to the wave equation

$$\nabla^2 \mathbf{E} + \frac{\omega^2 \varepsilon(\omega)}{c^2} \mathbf{E} = 0$$

$$\nabla^2 \mathbf{E} + k^2 \mathbf{E} = 0$$

- wave solution $\mathbf{E} \sim e^{i\mathbf{k} \cdot \mathbf{r}} e^{-i\omega t} = e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)}$ $\nabla^2 e^{i\mathbf{k} \cdot \mathbf{r}} = -|\mathbf{k}|^2 e^{i\mathbf{k} \cdot \mathbf{r}} = -k^2 e^{i\mathbf{k} \cdot \mathbf{r}}$

- wavevector and index of refraction

$$k = \sqrt{\frac{\varepsilon \omega^2}{c^2}} = \frac{n\omega}{c} \quad n(\omega) = \sqrt{\varepsilon(\omega)}$$

Διελεκτρική συνάρτηση: $\varepsilon(\omega) = \varepsilon_r + i\varepsilon_i$
Πολωτιμότητα του υλικού και απορρόφηση
Προκύπτει από την ηλεκτρονική διαμόρφωση

Διέκτης διάθλασης: $n = n_r + i n_i$
Διασκεδασμός και απορρόφηση
Καθορίζει τις ιδιότητες διάδοσης



Διελεκτρική συνάρτηση: $\epsilon(\omega) = \epsilon_r + i\epsilon_i$
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Δείκτης διάθλασης: $n = n_r + in_i$
Διασκεδασμός και απορρόφηση
Καθορίζει τις ιδιότητες διάδοσης

- **Wave propagation** $\mathbf{E} = \mathbf{E}_0 e^{i(\mathbf{k}\mathbf{r}-\omega t)} = \mathbf{E}_0 e^{i((\mathbf{k}_r+i\mathbf{k}_i)\mathbf{r}-\omega t)} = \mathbf{E}_0 e^{i(\mathbf{k}_r\mathbf{r}-\omega t)} e^{-k_i r}$
 - **along x direction** $\mathbf{E} = \mathbf{E}_0 e^{i(kx-\omega t)} = \mathbf{E}_0 e^{i((k_r+i\mathbf{k}_i)x-\omega t)} = \mathbf{E}_0 e^{i(k_r x-\omega t)} e^{-k_i x}$
 - Δύο όροι:
 - αρμονική κυματική διάδοση με k_r
 - Εκθετική απόσβεση με k_i
 - Η ροή ενέργειας δίνεται από το διάνυσμα Poynting $\mathbf{S} = \text{Re} \left\{ \frac{c}{8\pi} (\mathbf{E} \times \mathbf{H}^*) \right\} = \frac{cn}{8\pi} |\mathbf{E}|^2$
- $$|\mathbf{E}|^2 = |\mathbf{E}_0|^2 e^{-2k_i x} = |\mathbf{E}_0|^2 e^{-2n_i \alpha x / c} = |\mathbf{E}_0|^2 e^{-\alpha x}$$
- συντελεστής απόσβεσης $\alpha = \frac{2n_i \omega}{c} = 2n_i \frac{2\pi}{\lambda_0} = \frac{4\pi n_i}{\lambda_0}$



Metal dielectric function

- Drude dielectric function

$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega/\tau}$$

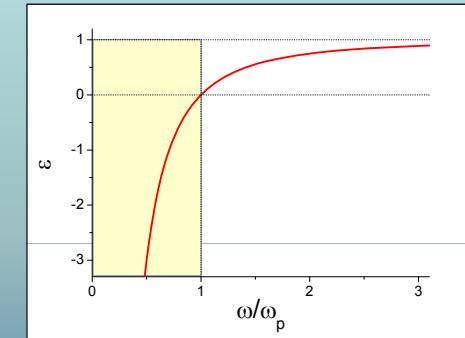
- For simplicity set $\tau = \infty$

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

- for $\omega < \omega_p$: reflection

- for $\omega > \omega_p$: transmission

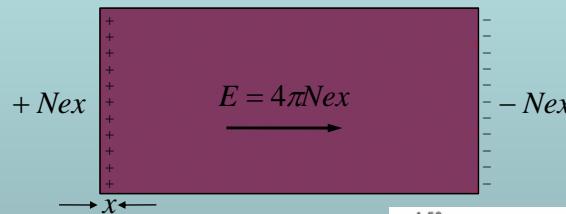
- for $\omega = \omega_p$: bulk plasmons



$\epsilon < 0$	$n = \text{imag} = in_i$	$R = \left \frac{in_i - 1}{in_i + 1} \right ^2 = 1$
$\epsilon > 0$	$n = \text{real} = n_l$	$R = \left \frac{n_l - 1}{n_l + 1} \right ^2 < 1$
$\epsilon = 0$	$n = 0$	longitudinal waves: $\epsilon \nabla \mathbf{E} = 0 \Rightarrow \nabla \mathbf{E} \neq 0$



Bulk plasmon

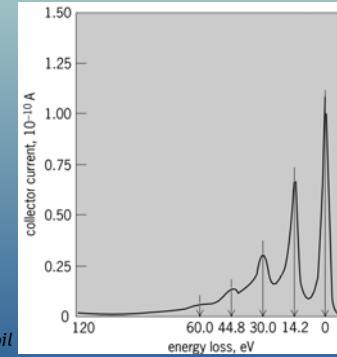


$$m\ddot{x} = -eE \Rightarrow \ddot{x} + \frac{4\pi Ne^2}{m}x = 0$$

self-sustained longitudinal charge oscillations with frequency ω_p

absorption peaks at $E = n\hbar\omega_p$

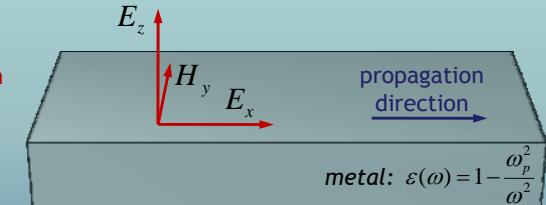
Number of detected electrons in a beam versus their energy loss during transit through a thin aluminum foil



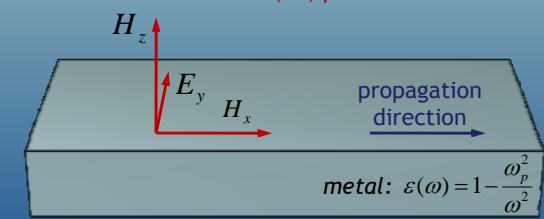
Surface plasmon polaritons

Two independent polarizations:

Transverse magnetic (TM) polarization



Transverse electric (TE) polarization





Surface plasmon polaritons

- Only the TM has plasmon solution
- SPP solution is obtained from combining:
 - boundary conditions
 - Maxwell's equation
 - wave equation

boundary conditions at $z=0$

- continuity of the parallel E
 $E_x = E'_x$
- continuity of the parallel H
 $H_y = H'_y$
- continuity of the perpendicular D
 $\epsilon_d E_z = \epsilon_m E'_z$

Maxwell's equation

$$\nabla \times \mathbf{H} = \frac{\epsilon}{c} \frac{\partial \mathbf{E}}{\partial t} \Rightarrow \begin{cases} \frac{\partial H_y}{\partial z} = -\frac{\epsilon}{c} \frac{\partial E_x}{\partial t} & ckH_y = -i\epsilon_d \omega E_x \\ \frac{\partial H_y}{\partial x} = \frac{\epsilon}{c} \frac{\partial E_z}{\partial t} & ckH'_y = i\epsilon_m \omega E'_x \end{cases}$$

$\Rightarrow \kappa = -\kappa' \epsilon_d / \epsilon_m$

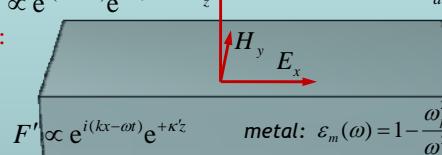
wave equation

$$\nabla^2 \mathbf{E} + \frac{\omega^2 \epsilon(\omega)}{c^2} \mathbf{E} = 0 \Rightarrow \begin{cases} -k^2 + \kappa^2 + \omega^2 \epsilon_d / c^2 = 0 \\ -k^2 + \kappa'^2 + \omega^2 \epsilon_m / c^2 = 0 \end{cases}$$

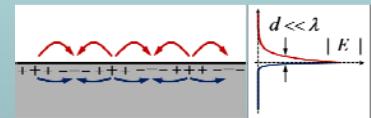
SPP dispersion

$$k = \frac{\omega}{c} \sqrt{\frac{\epsilon_d \epsilon_m}{\epsilon_d + \epsilon_m}}$$

$F \propto e^{i(kx-\omega t)} e^{-\kappa z}$



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



Surface plasmon polaritons

Surface plasmon polariton (SPP)

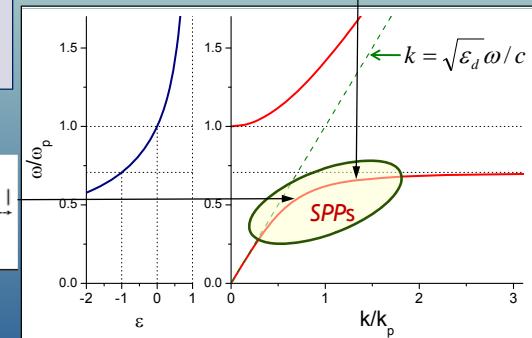
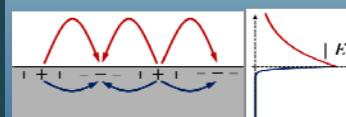
$$\left. \begin{array}{l} \kappa = -\kappa' \epsilon_d / \epsilon_m \\ k = \frac{\omega}{c} \sqrt{\frac{\epsilon_d \epsilon_m}{\epsilon_d + \epsilon_m}} \end{array} \right\}$$

SPP condition

$$\epsilon_m(\omega) < -\epsilon_d$$

Surface plasmon

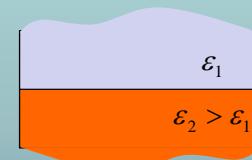
$$\epsilon_m(\omega) = -\epsilon_d \Rightarrow \omega_{SP} = \frac{\omega_p}{\sqrt{1 + \epsilon_d}}$$



Guided light

Free waves in 3D

$$\mathbf{E} = \hat{\mathbf{E}} E_0 e^{i(k_x x + k_y y + k_z z - \omega t)} \quad \left. \begin{array}{l} k_x^2 + k_y^2 + k_z^2 = \epsilon \frac{\omega^2}{c^2} \\ \nabla^2 \mathbf{E} + \epsilon \frac{\omega^2}{c^2} \mathbf{E} = 0 \end{array} \right\}$$



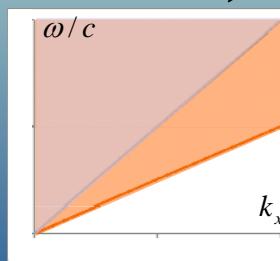
Assume an interface between two materials

- x direction is uniform, and so k_x is conserved
- in material 1

$$k_x^2 = \epsilon_1 \frac{\omega^2}{c^2} - k_y^2 - k_z^2 \Rightarrow k_x \leq \sqrt{\epsilon_1} \frac{\omega}{c}$$

- in material 2

$$k_x^2 = \epsilon_2 \frac{\omega^2}{c^2} - k_y^2 - k_z^2 \Rightarrow k_x \leq \sqrt{\epsilon_2} \frac{\omega}{c}$$



Surface plasmon polaritons

Surface plasmon polariton (SPP)

$$\left. \begin{array}{l} \kappa = -\kappa' \epsilon_d / \epsilon_m \\ k = \frac{\omega}{c} \sqrt{\frac{\epsilon_d \epsilon_m}{\epsilon_d + \epsilon_m}} \end{array} \right\}$$

SPP condition

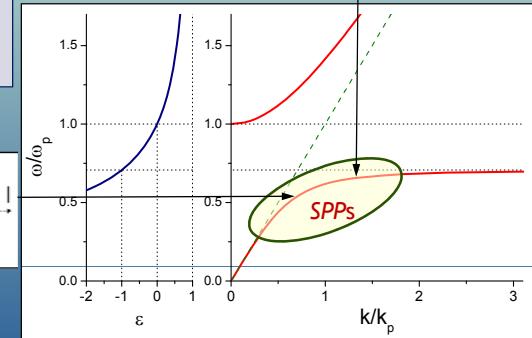
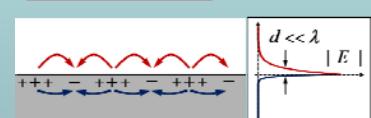
$$\epsilon_m(\omega) < -\epsilon_d$$

Surface plasmon

$$\epsilon_m(\omega) = -\epsilon_d \Rightarrow \omega_{SP} = \frac{\omega_p}{\sqrt{1 + \epsilon_d}}$$



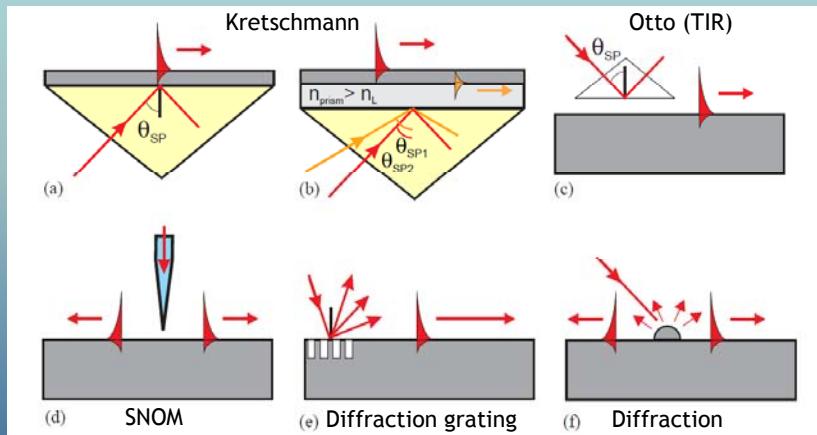
$$\epsilon_m(\omega) = 1 - \frac{\omega_p^2}{\omega^2}$$





Excitation of SPPs

- Surface plasmon polaritons exist below the light-cone
 - special care is needed to excite them



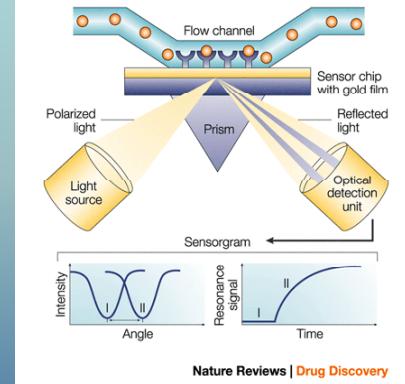
Zayats, Smolyaninov and Maradudin, Phys. Rep. 408, 131 (2005)



SPP applications

Plasmonics: Experiment, Theory and Applications

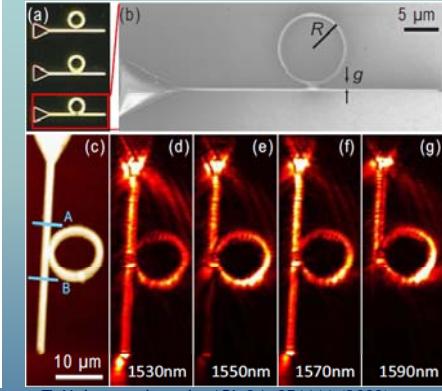
biological sensors



Nature Reviews | Drug Discovery

M.A. Cooper, NRDD 1, 515 (2002)

subwavelength waveguides

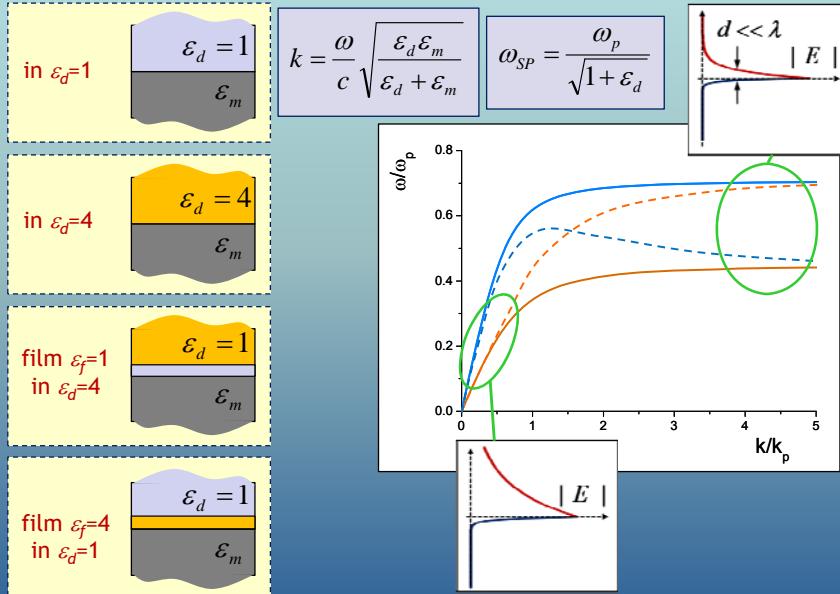


T. Holmgard et al., APL 94, 051111 (2009)



More complex SPPs

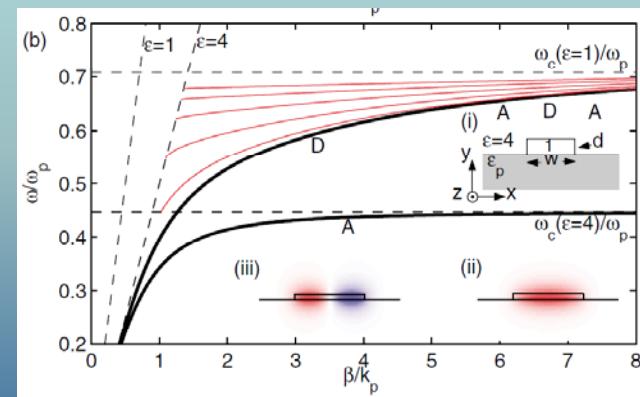
Plasmonics: Experiment, Theory and Applications



SPP waveguides

Plasmonics: Experiment, Theory and Applications

- Surface-plasmon-assisted guiding of broadband slow and subwavelength light in air



Karalis, Lidorikis, Ibanescu, Joannopoulos and Soljacic, PRL 95, 063901 (2005)



Metallic nanoparticles

- A metallic nanoparticle inside a electromagnetic field
 - $a \ll \lambda \Rightarrow$ electrostatic limit

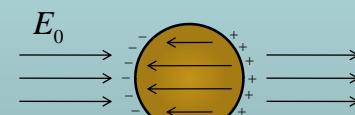


$$\Phi_{in} = -\left(\frac{3}{\epsilon_m + 2\epsilon_d}\right)E_0 r \cos \theta$$

uniform

$$\Phi_{out} = -E_0 r \cos \theta + \left(\frac{\epsilon_m - \epsilon_d}{\epsilon_m + 2\epsilon_d}\right)E_0 \frac{a^3}{r^2} \cos \theta$$

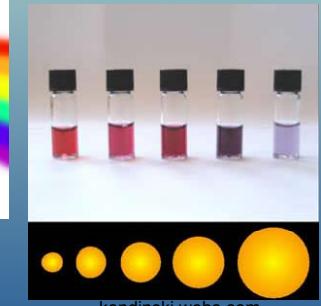
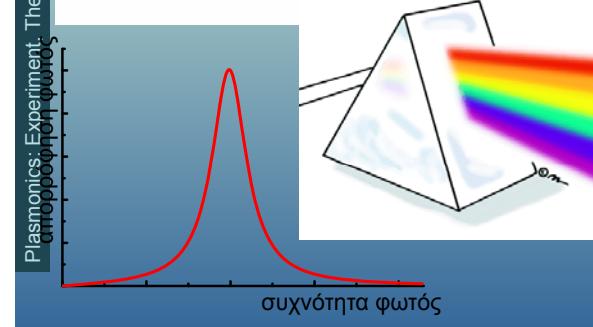
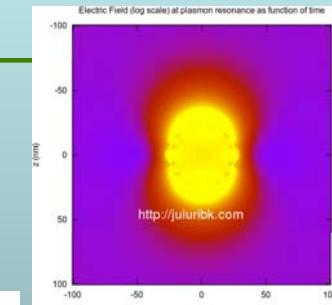
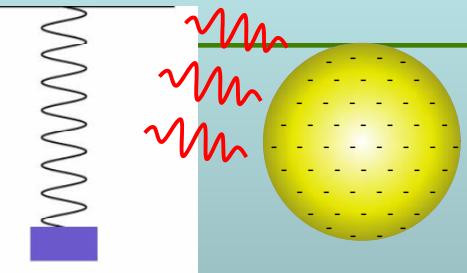
uniform dipole



Inside an electric field a nanoparticle gets polarized

$$p(t) \cong 4\pi a^3 \left(\frac{\epsilon_m - \epsilon_d}{\epsilon_m + 2\epsilon_d}\right) E(t)$$

- strong scattering
- strong local fields



Localized surface plasmon resonance

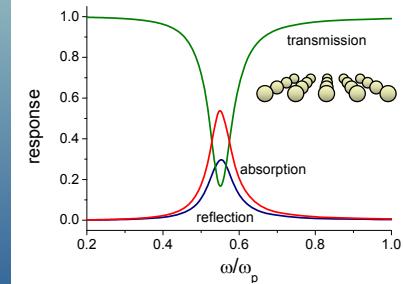
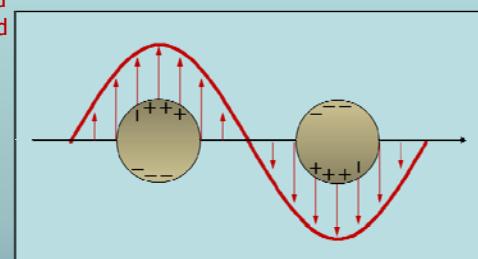
- Inside an electromagnetic field the nanoparticle gets polarized

$$p(t) \cong 4\pi a^3 \left(\frac{\epsilon_m - \epsilon_d}{\epsilon_m + 2\epsilon_d}\right) E(t)$$

- Surface Plasmon Resonance

$$\epsilon_m(\omega) = -2\epsilon_d \Rightarrow$$

$$\omega_{LSPR} = \frac{\omega_p}{\sqrt{1+2\epsilon_d}}$$



- At resonance we get extreme optical behavior

- local fields get maximized
- scattering gets maximized
- absorption gets maximized

- Assume the metal dielectric

$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega/\tau}$$

- We calculate the effective medium produced by the NPs

- Maxwell-Garnett formula

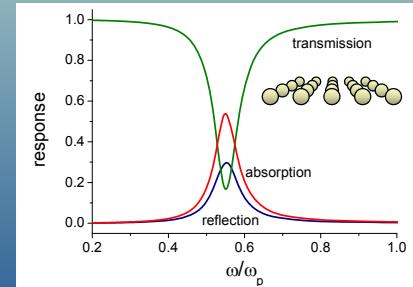
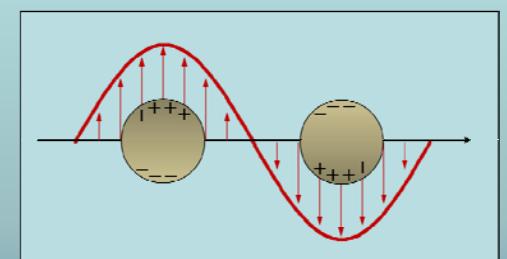
$$\frac{\tilde{\epsilon} - \epsilon_d}{\tilde{\epsilon} + 2\epsilon_d} = f \frac{\epsilon_m - \epsilon_d}{\epsilon_m + 2\epsilon_d}$$

for $\epsilon_d = 1$:

$$\tilde{\epsilon} = 1 + \frac{f\omega_p^2}{(1-f)\omega_{LSPR}^2 - \omega^2 - i\omega/\tau}$$

- This is a Lorentzian!

- The free electrons oscillate as a driven oscillator
- NPs are like polarizable atoms



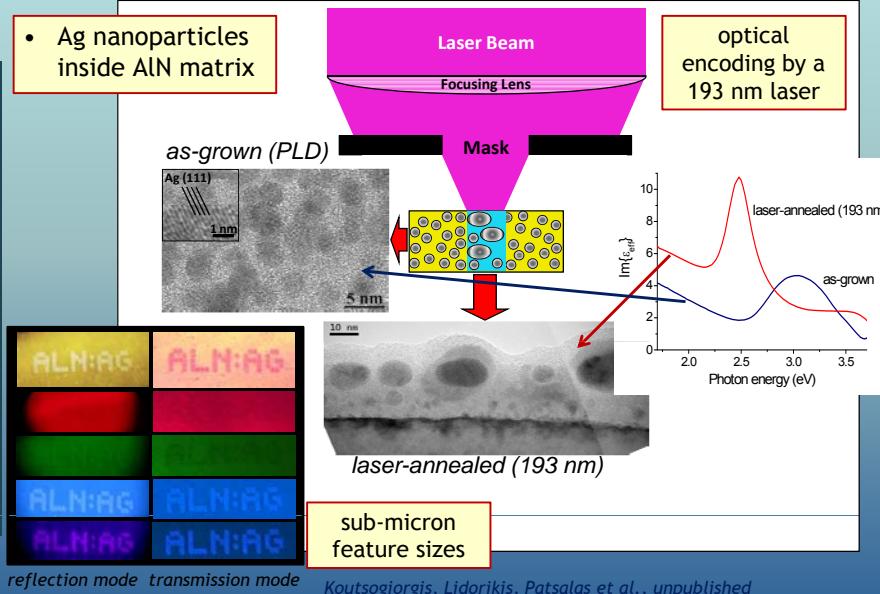


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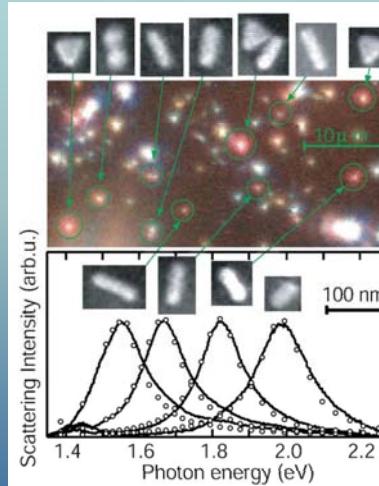


Optical encoding with LSPRs

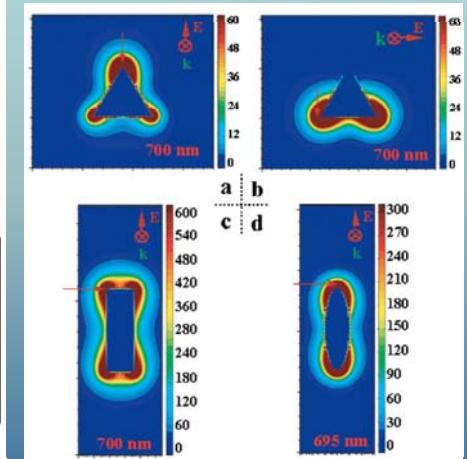
- Ag nanoparticles inside AlN matrix



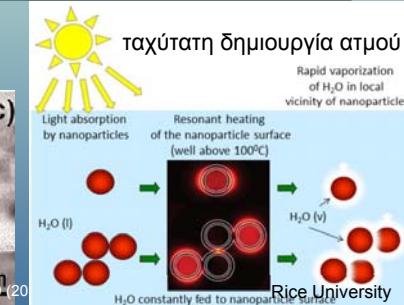
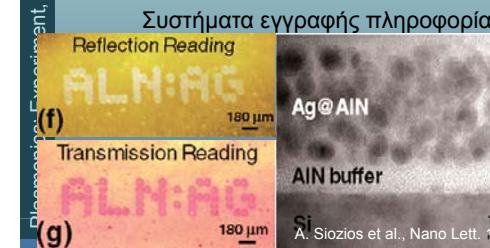
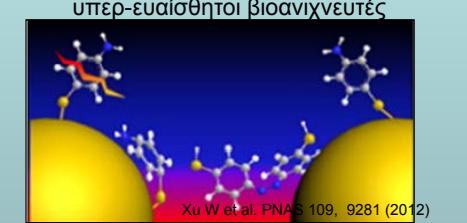
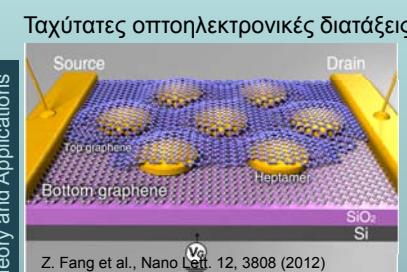
Au and Ag nanoprisms



Maier et al., JAP 98, 011101 (2005)



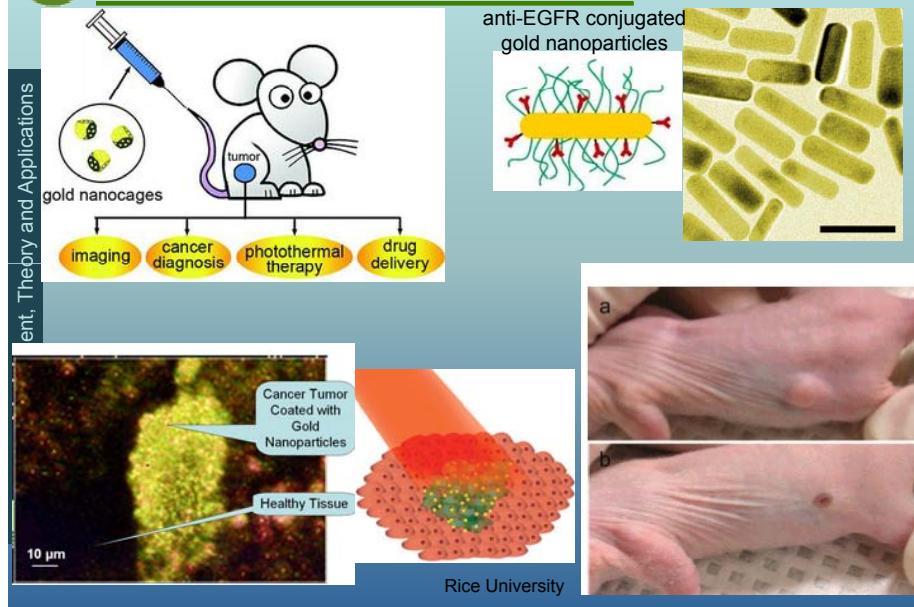
Εφαρμογές πλασμονικών νανοσωματιδίων





Εφαρμογές πλασμονικών νανοσωματιδίων στην ιατρική

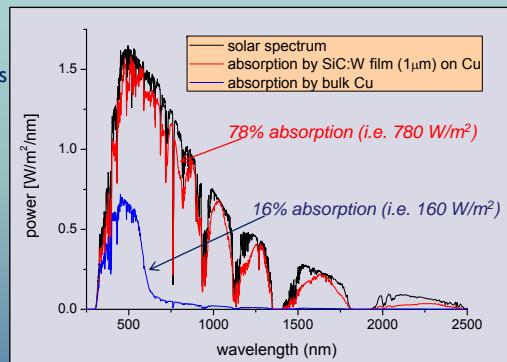
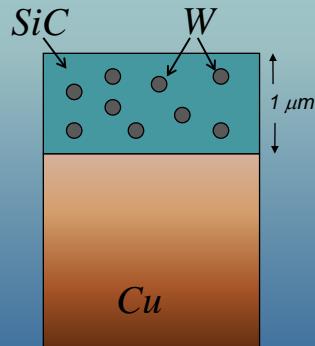
Plasmonics: Experiment, Theory and Applications



Solar harvesting

Plasmonics: Experiment, Theory and Applications

- A thin composite film of SiC with W nanoparticles
 - absorbs most of spectrum
 - inert and stable (protection)
 - withstands high temperatures ($>1000^{\circ}\text{C}$)



Bellas and Lidorikis, unpublished

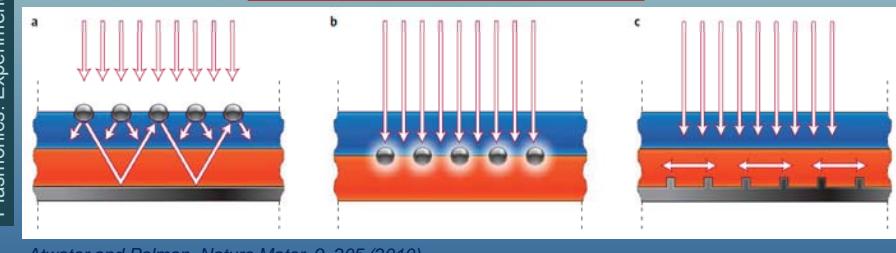


Enhanced solar absorption

Plasmonics: Experiment, Theory and Applications

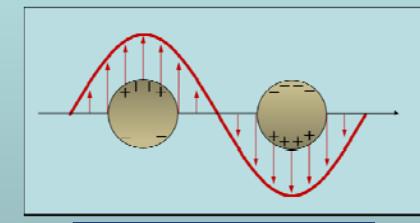
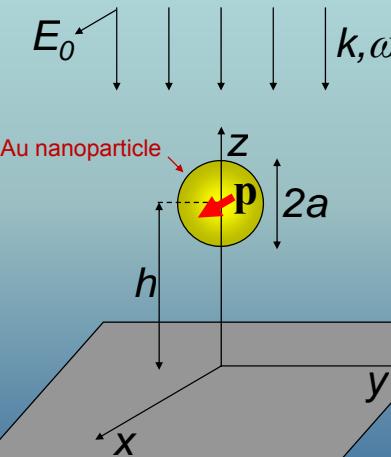
- Metallic nanoparticles and/or nanostructures can be used:
 - on the surface
 - inside the semiconductor
 - on the back contact
- Enhancement due to:
 - Scattering from the nanoparticles: increased optical path
 - LSPR near-fields: increased fields driving the absorption

Particularly important for organic and thin film solar cells



Theory of SPR near-field enhancement

Plasmonics: Experiment, Theory and Applications



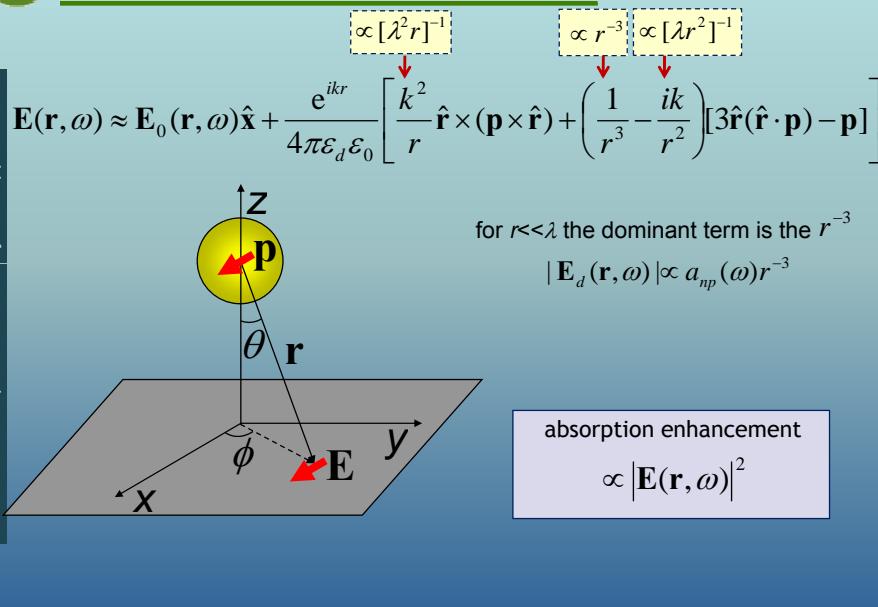
$$\mathbf{p}(\omega) = \epsilon_0 \epsilon_d \alpha_{np}(\omega) \mathbf{E}_0$$

for $a \ll \lambda$

$$\alpha_{np}(\omega) \equiv 4\pi a^3 Q \cong 4\pi a^3 \left(\frac{\epsilon_m - \epsilon_d}{\epsilon_m + 2\epsilon_d} \right)$$



Theory of SPR near-field enhancement



Absorption enhancement

- at a point

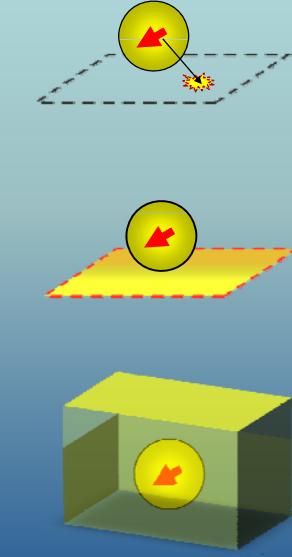
$$F(\mathbf{r}, \omega) \cong 1 + |\mathcal{Q}|^2 \left(\frac{a}{r} \right)^6 (3x^2 + 1) + \\ + 2|\mathcal{Q}| \left(\frac{a}{r} \right)^3 (3x^2 - 1) \cos \Delta$$

- if we integrate on a plane

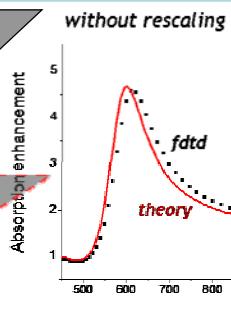
$$F(z, \omega) \cong 1 - \sigma(1 - \delta^2) + \\ + \frac{1}{4}\sigma|\mathcal{Q}|^2 \left(\frac{a}{z} \right)^4 (5\delta^4 - 2\delta^6) + \\ + 2\sigma|\mathcal{Q}| \left(\frac{a}{|z|} \right) \delta(1 - \delta^2 - k^2 z^2) \cos \Delta'$$

- if we integrate within a volume

$$F(\omega) \cong 1 - f + 2f|\mathcal{Q}|^2 + 2f \operatorname{Re}\{\mathcal{Q}\}$$



Planar absorption enhancement



- Multiple scattering \rightarrow Claussius-Mossotti

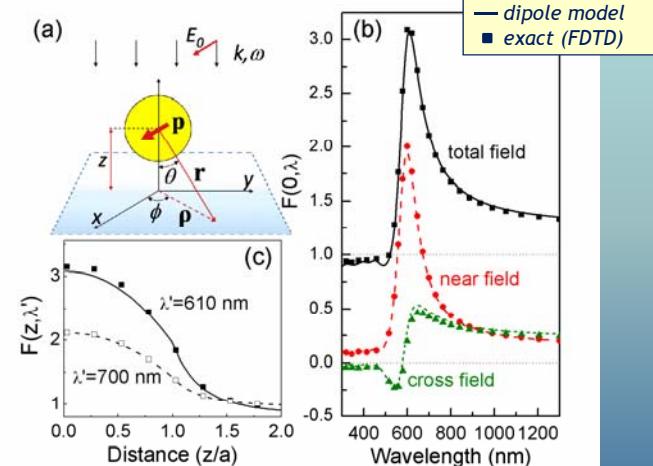
$$\alpha_{np} \rightarrow \frac{a_{np}}{1 - \frac{N}{3}\alpha_{np}} \Rightarrow \mathcal{Q} \rightarrow \frac{|\mathcal{Q}|}{1 - f \cdot |\mathcal{Q}|}$$

- Absorption in the nanoparticle \rightarrow reduced field strength

$$|E_0|^2 \rightarrow |E_0|^2 (1 - C_{abs}/L^2) = \gamma |E_0|^2$$

$$C_{abs} = k \operatorname{Im}\{\alpha_{np}\} = 4\pi a^3 k \operatorname{Im}\{Q\}$$

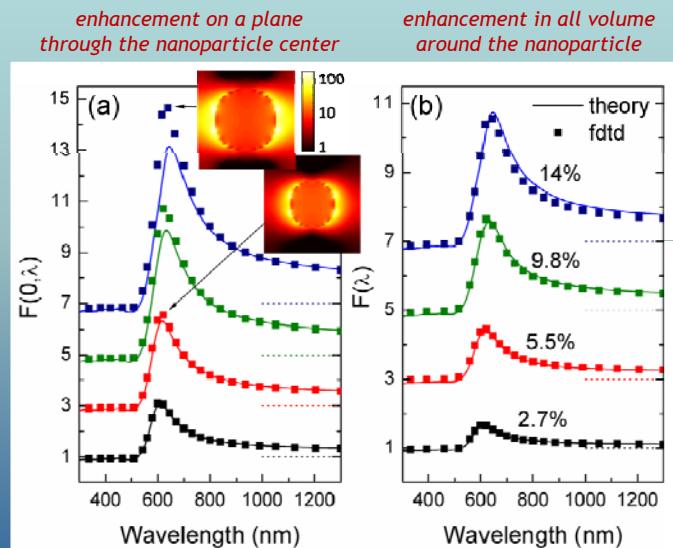
Planar absorption enhancement



Simple dipole model has excellent agreement with exact numerical simulations (FDTD)



Planar and volume absorption enhancement



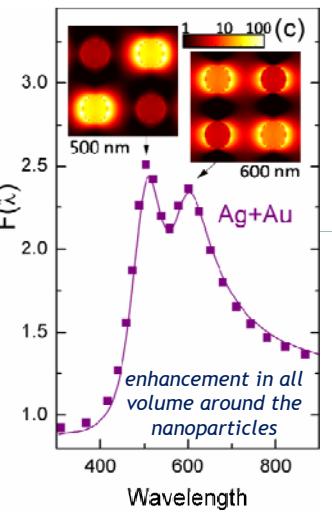
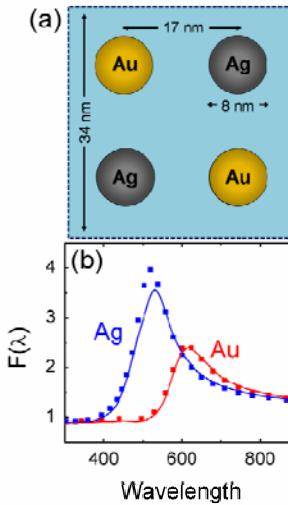
Volume enhancement of composites

if more than one type of nanoparticles is present, the rescalings become:

$$Q_i \rightarrow \frac{Q_i}{1 - \sum f_i Q_i}$$

$$\gamma = 1 - \frac{\sum f_i C_i}{L^2 f}$$

$$\sum f_i = f$$



combining different nanoparticles we can tailor the absorption spectrum of the semiconductor

Lagos, Sigalas and Lidorikis, submitted



Conclusions

- Exciting time for plasmonics
 - sensors
 - optical waveguides and components
 - encoding
 - biomedical
 - solar harvesting
 - enhanced light-matter interactions
 - and many more to come...

*Thank you for
your
attention!*