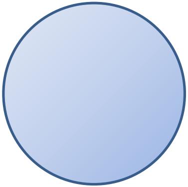
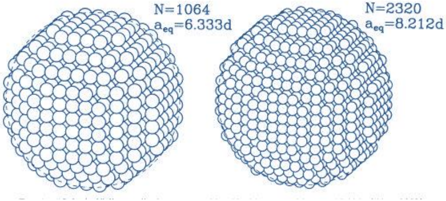
3. Interacting NPs

Discrete Dipole Approximation (DDA)

DDA





$$j = 1, \ldots, N$$

$$\mathbf{r}_j$$

$$\alpha_j^{\mathrm{CM}} = \frac{3d^3}{4\pi}\frac{\epsilon_j - 1}{\epsilon_j + 2}$$

$$\mathbf{E}_{\mathrm{inc},j} = \mathbf{E}_0 \exp(i\mathbf{k}\cdot\mathbf{r}_j - i\omega t)$$

$$\begin{cases} \mathbf{P}_j = \alpha_j \mathbf{E}_j \\ \mathbf{E}_j = \mathbf{E}_{\mathrm{inc},j} - \sum_{k\neq j} \mathbf{A}_{jk}\mathbf{P}_k \end{cases}$$

$$\boldsymbol{P}_j = \alpha_j\left(\boldsymbol{E}_{\mathrm{inc},j} - \sum_{k\neq j} \mathbf{A}_{jk}\boldsymbol{P}_k\right)$$

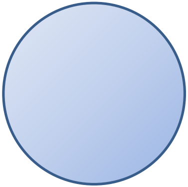
$$\begin{cases} \mathbf{A}_{jk} = \dfrac{\exp(ikr_{jk})}{r_{jk}}\left[k^2(\hat{r}_{jk}\hat{r}_{jk} - \mathbf{1}_3) + \dfrac{ikr_{jk} - 1}{r_{jk}^2}(3\hat{r}_{jk}\hat{r}_{jk} - \mathbf{1}_3)\right] & j \neq k \\ \mathbf{A}_{jj} \equiv \alpha_j^{-1} \quad \text{definition} & j = k \end{cases}$$

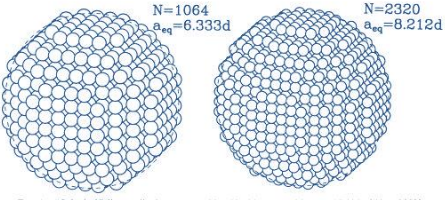
$$k \equiv \omega/c,\ r_{jk} \equiv |\mathbf{r}_j - \mathbf{r}_k|,\ \hat{r}_{jk} \equiv (\mathbf{r}_j - \mathbf{r}_k)/r_{jk},$$

$$\boxed{\sum_{k=1}^{N} \mathbf{A}_{jk}\mathbf{P}_k = \mathbf{E}_{\mathrm{inc},j}}$$

Draine, B. T.; Flatau, P. J. Discrete-Dipole Approximation For Scattering Calculations. *J. Opt. Soc. Am. A, JOSAA* **1994**, *11*, 1491–1499.

DDA





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$$\boldsymbol{P}_j = \alpha_j\left(\boldsymbol{E}_{\mathrm{inc},j} - \sum_{k \neq j} \mathbf{A}_{jk}\boldsymbol{P}_k\right)$$

solving

$$\sum_{k=1}^{N} \mathbf{A}_{jk}\mathbf{P}_k = \mathbf{E}_{\mathrm{inc},j}, \quad j = 1, \dots, N$$

$$\tilde{\mathbf{A}}\tilde{\mathbf{P}} = \tilde{\mathbf{E}}_{\mathrm{inc}}$$

$$C_{\mathrm{ext}} = \frac{4\pi k}{|\mathbf{E}_0|^2} \sum_{j=1}^{N} \mathrm{Im}(\mathbf{E}_{\mathrm{inc},j}{}^* \cdot \mathbf{P}_j),$$

$$C_{\mathrm{abs}} = \frac{4\pi k}{|\mathbf{E}_0|^2} \sum_{j=1}^{N} \left\{ \mathrm{Im}[\mathbf{P}_j \cdot (\alpha_j{}^{-1})^* \mathbf{P}_j{}^*] - \frac{2}{3} k^3 |\mathbf{P}_j|^2 \right\}$$

$$\mathbf{E}_{\mathrm{sca}} = \frac{k^2 \exp(ikr)}{r} \sum_{j=1}^{N} \exp(-ik\hat{r} \cdot \mathbf{r}_j)(\hat{r}\hat{r} - \mathbf{1}_3)\mathbf{P}_j$$

Draine, B. T.; Flatau, P. J. Discrete-Dipole Approximation For Scattering Calculations. *J. Opt. Soc. Am. A, JOSAA* **1994**, *11*, 1491–1499.

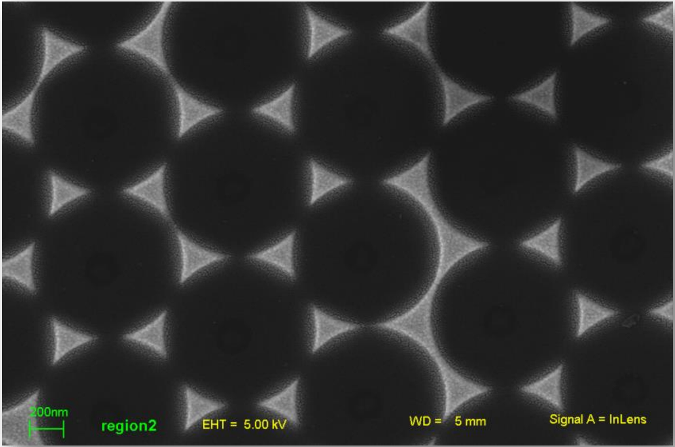
3. Interacting NPs

3.3 Array 2D - NSL

Array 2D - NSL

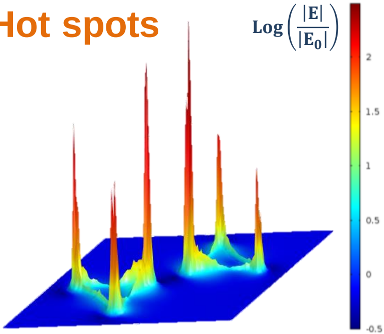
NPA

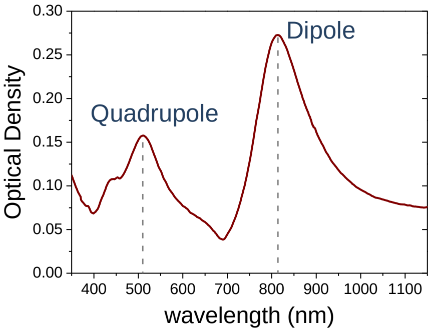
Nano-Prism arrays

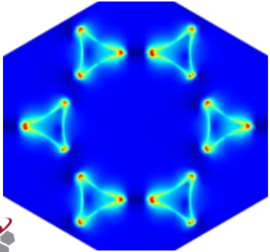


Interacting nanostructures

Hot spots

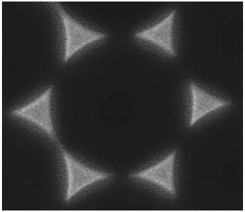






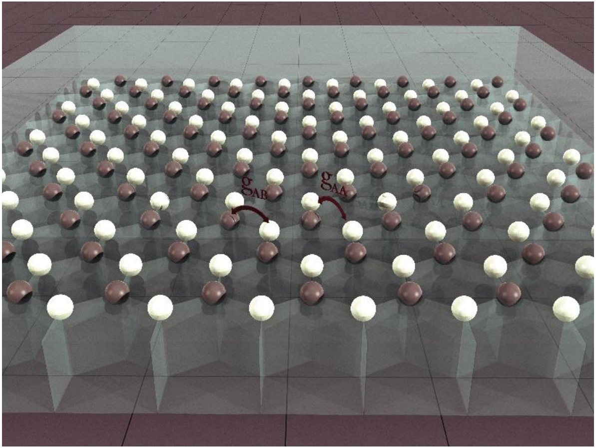
Interaction:

- SPR red-shift
- Local-field enhancement ~ 100 @ SPR
- Hot-spots



Array 2D - NSL

Diffractive dipolar coupling in non-Bravais plasmonic lattices



G. Pirruccio et al., Nanoscale Adv., 2020,2, 1261-1268

Array 2D - NSL

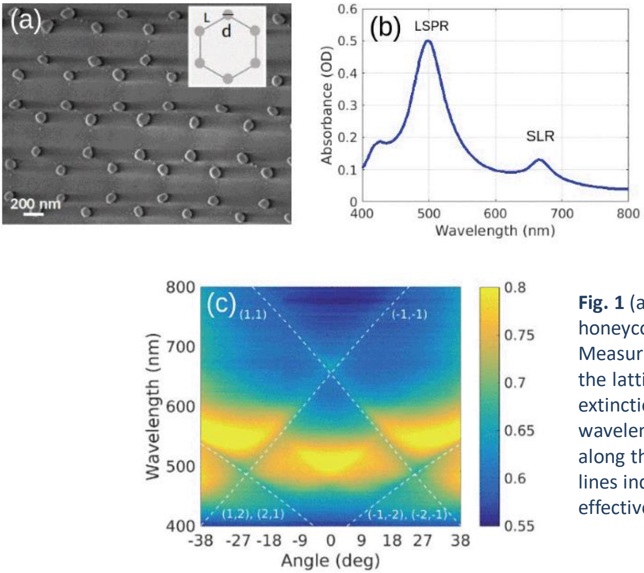
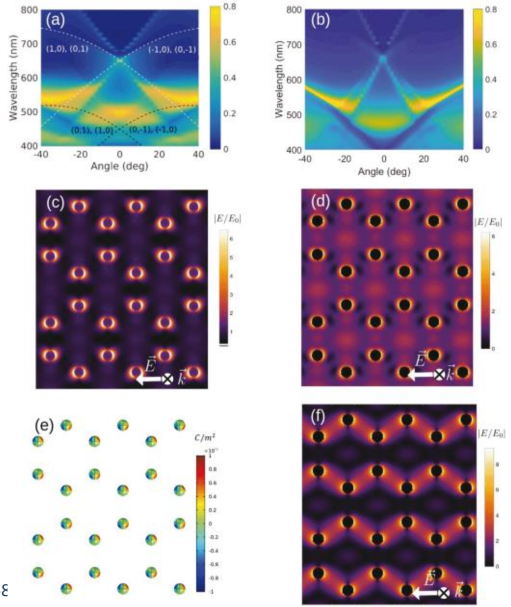


Fig. 1 (a) SEM image of the honeycomb plasmonic lattice. (b) Measured absorbance spectrum of the lattice. (c) Measured s-polarized extinction as a function of the wavelength and angle of incidence along the Γ − M trajectory. Dashed lines indicate RAs calculated with an effective refractive index of 1.47

G. Pirruccio et al., Nanoscale Adv., 2020,2, 1261-1268

Array 2D - NSL

Fig. 2 (a) Finite element method simulation and (b) spectral representation calculation of the s-polarized extinction map along the Γ – M trajectory. Dashed curves in (a) indicate RAs. (c) Simulated and (d) calculated spatial distribution of the normalized electric field amplitude for the LSPR, around λ = 500 nm. (e) Simulated surface charge density and (f) calculated spatial distribution of the normalized electric field amplitude for the SLR peak at λ = 660 nm. (c–f) are plotted in the plane crossing the nanospheres along their diameter, at normal incidence and for s-polarized incident light.



G. Pirruccio et al., Nanoscale Adv., 2020,2, 1261-1268

Nonlinear response of materials

The response of materials to an optical field $\boldsymbol{E}$ is described by the material polarization $\boldsymbol{P}$:

$$\mathbf{P} = \varepsilon_0 \left[\chi^{(1)}\mathbf{E} + \chi^{(2)}\mathbf{E}^{\mathbf{2}} + \chi^{(3)}\mathbf{E}^{\mathbf{3}} + \ldots + \chi^{(n)}\mathbf{E}^{\mathbf{n}} + \ldots \right]$$

Second-order effects:

- second harmonic generation (SHG)
- optical parametric amplification (OPA, OPO)
- ...

Third-order effects:

- third harmonic generation (THG)
- optical Kerr effect
- four-wave mixing (FWM)
- ...

The **local-field enhancement effects** can be exploited to design plasmonic nanosystems with amplified and tunable nonlinear optical response.

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- **optical Kerr effect**
- four-wave mixing (FWM)
- ...

The **local-field enhancement effects** can be exploited to design plasmonic nanosystems with amplified and tunable nonlinear optical response. !

$$\varepsilon = \varepsilon_0 \left[\chi^{(1)} + 3\chi^{(3)} \mathbf{E}^2 \right]$$

$$\begin{cases} n(I) = n_0 + n_2 I \\ \alpha(I) = \alpha_0 + \beta\, I \end{cases}$$

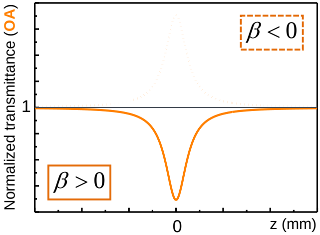
optical Kerr effect

The z-scan technique

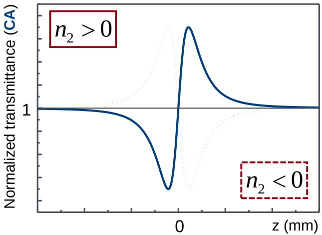
M. Sheik-Bahae *et al.*
IEEE J. Quantum El. **26** 760 (1990)

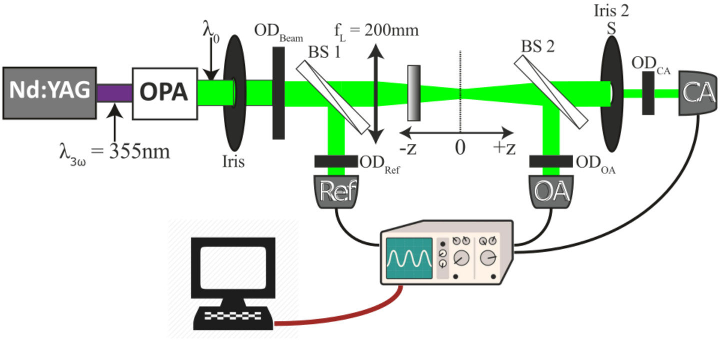
$$\begin{cases} n(I) = n_0 + n_2 I \\ \alpha(I) = \alpha_0 + \beta\, I \end{cases}$$

Open Aperture (OA)



Closed Aperture (CA)





The z-scan set-up

M. Sheik-Bahae *et al.*
IEEE J. Quantum El. **26** 760 (1990)

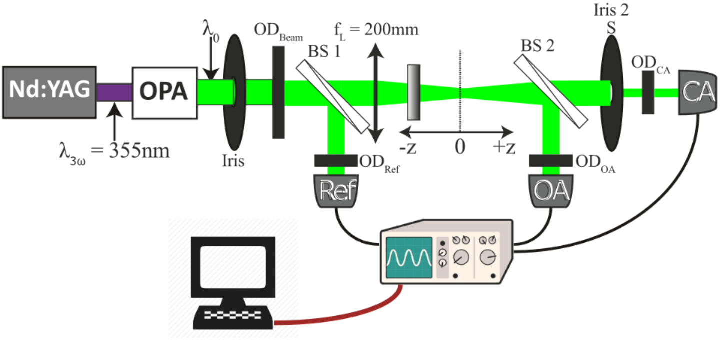
$$\begin{cases} n(I) = n_0 + n_2 I \\ \alpha(I) = \alpha_0 + \beta\, I \end{cases}$$

mode-locked Nd:YAG + OPA

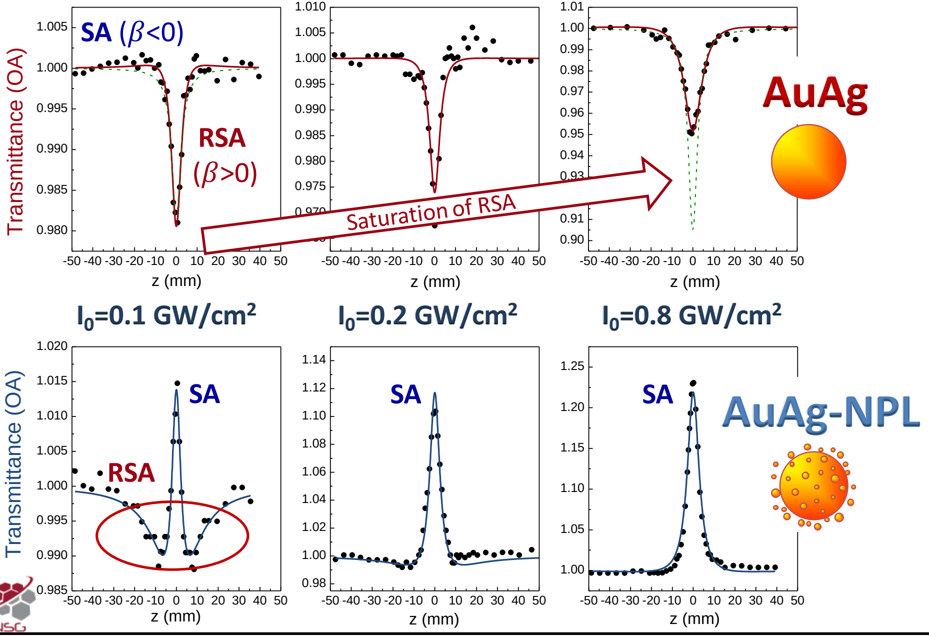
- E ≅ 30 mJ (@ 1064 nm)
- Δt ≅ 15 ps
- RR = 10 Hz
- λ = 420 – 2000 nm

pure electronic effects!

tunability!

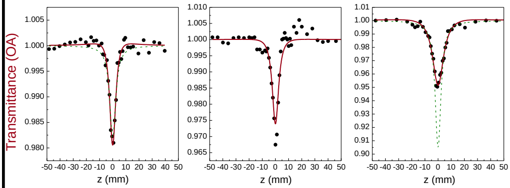


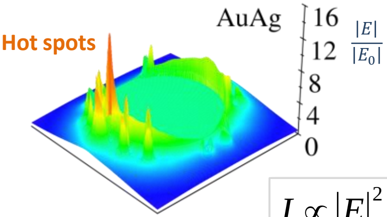
Open Aperture results



Open Aperture results

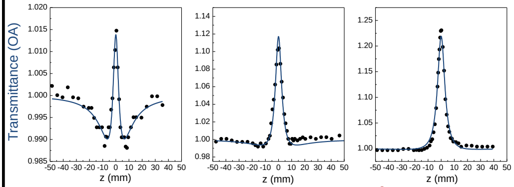
AuAg



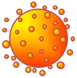


$$I \propto |E|^2$$

Effective intensity



AuAg-NPL



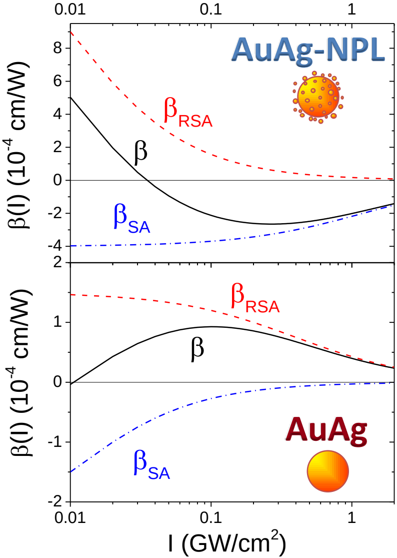
Nonlinear absorption

$$\alpha(I) = \alpha_0 + \beta\, I$$

Nonlinear absorption coefficient

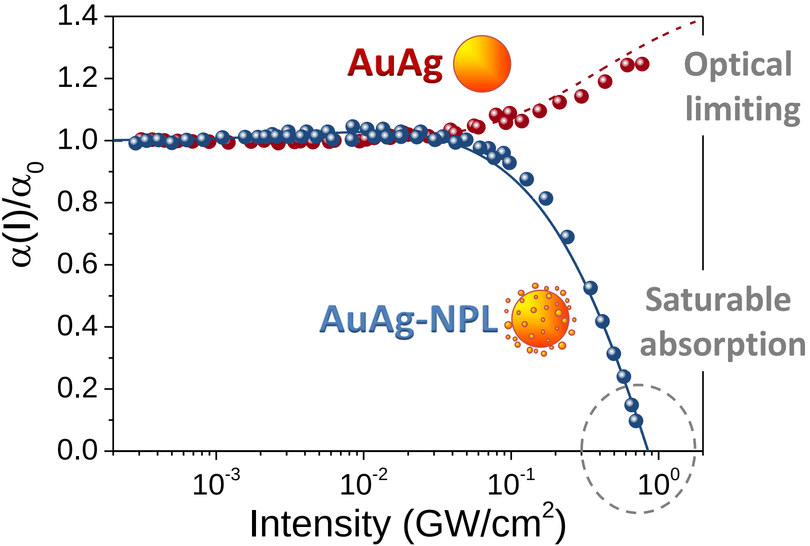
$$\beta(I) = \beta_{RSA}(I) + \beta_{SA}(I) = \frac{\beta_+}{1 + \frac{I}{I_s^+}} + \frac{\beta_-}{1 + \frac{I}{I_s^-}}$$

	AuAg	AuAg-NPL
β_+ (10^{-3} cm/W)	0.15 ± 0.03	1.9 ± 0.7
I_s^+ (10^8 W/cm^2)	4 ± 2	0.09 ± 0.04
β_- (10^{-4} cm/W)	-3 ± 1	-4 ± 1
I_s^- (10^9 W/cm^2)	0.010 ± 0.001	1.2 ± 0.1



T. Cesca *et al.* Optics Express. **20** 4537 (2012)

Nonlinear absorption



T. Cesca *et al.* Optics Express. **20** 4537 (2012)