Influence of the atomic structure on the electric field enhancement in plasmonic nanostructures

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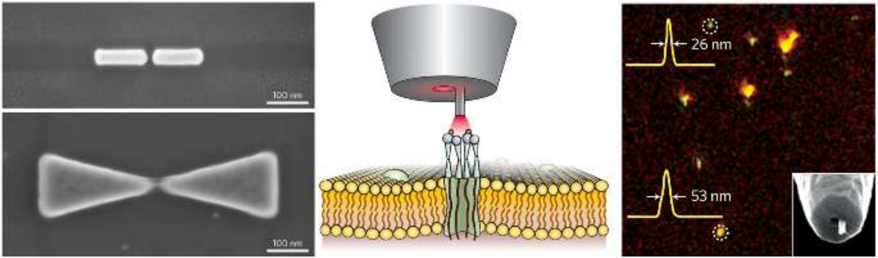
Why field enhancement in nanocavity?



Left panel: two antennas with gap sizes down to $\sim$ 10 nm, fabricated by focused ion beam milling. Center panel: schematic illustration of a biological imaging with optical antennas. Right panel: Fluorescently labelled antibodies imaged by the antenna probe.[1]

Understanding light-matter interaction, application to:

- Nanoplasmonic devices, such as nanosensor[2], nanoantennas[1]...
- Variety of technological applications as vibrational spectroscopies (SERS), solar cells ...

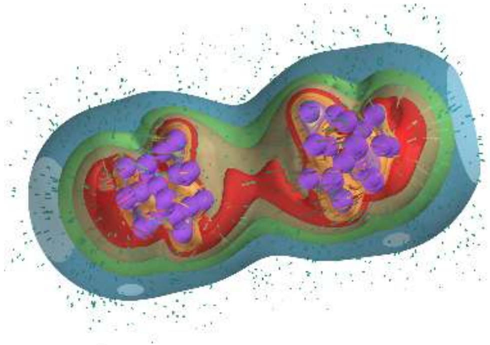
[1] L. Novotny, N. Hulst, Nature Phot. 5, 83 (2011).
[2] J. N. Anker et al. Nature Mater. 7, 442 (2008).

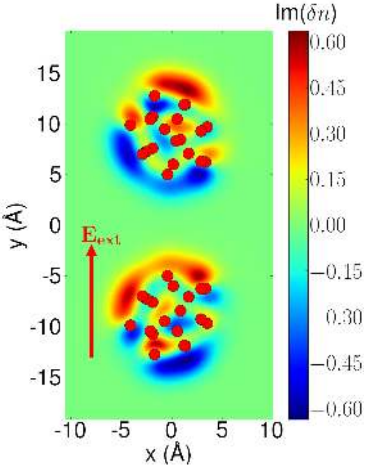
Field enhancement from quantum mechanics calculation

Density change and Induced electric field

$$[1 - \chi_0(\omega)K(\omega)]\,\delta n_\mu(\omega) = \chi_0(\omega)\mathbf{d}^\nu$$

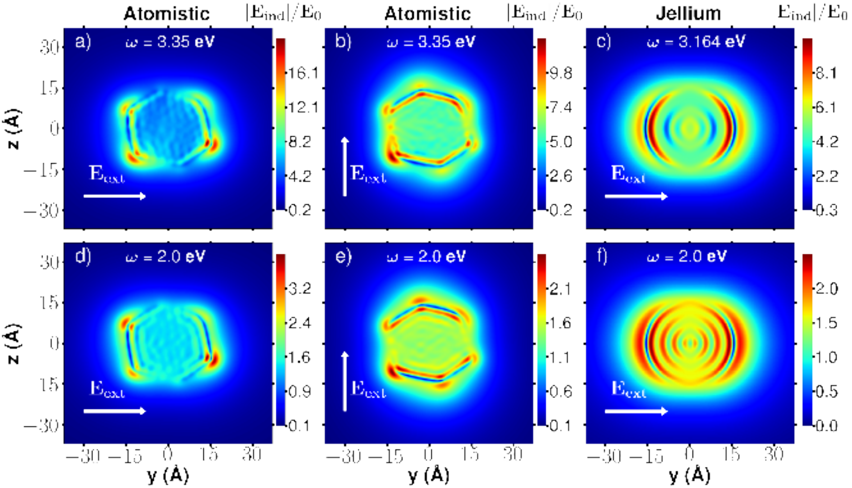
$$\mathbf{E_{ind}}(\mathbf{r},\omega) = -\nabla_r \int \frac{\delta n(\mathbf{r}',\omega)}{|\mathbf{r}-\mathbf{r}'|} d^3\mathbf{r}'$$



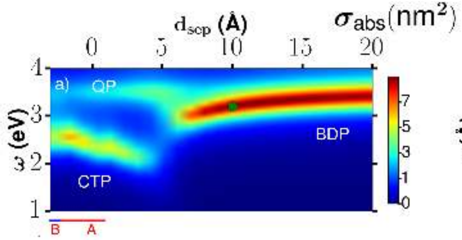


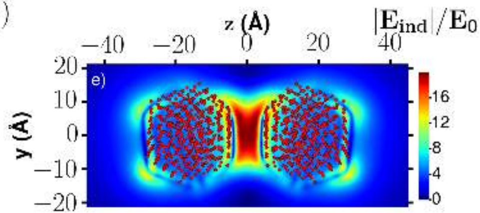
Density distribution of a Na_{20} dimer

TDDFT calculations with atomic-scale resolution: Atomic-scale lightning rod effect for Na_{380}.



Far field and near field for Na_{380}





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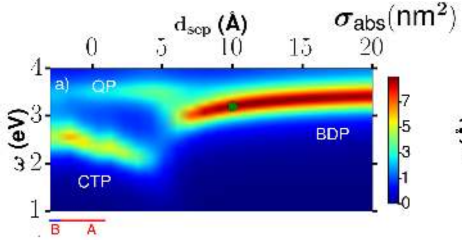

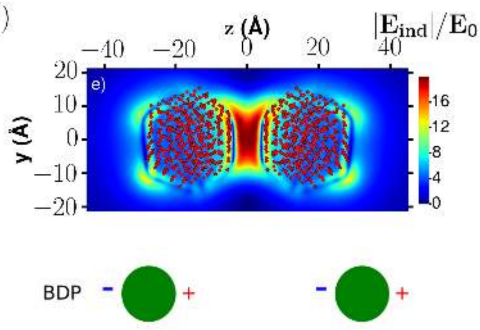

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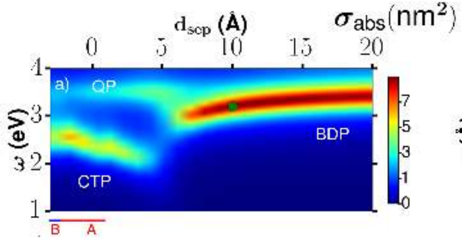
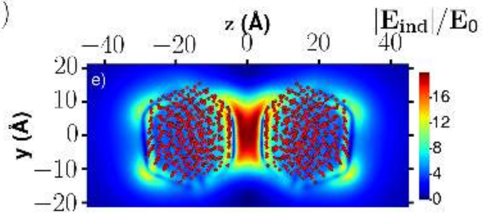
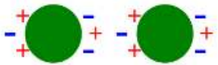


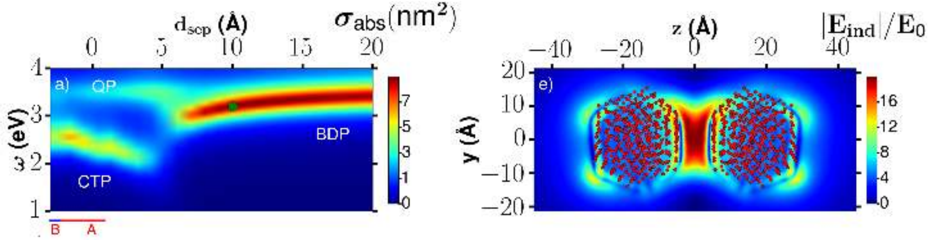


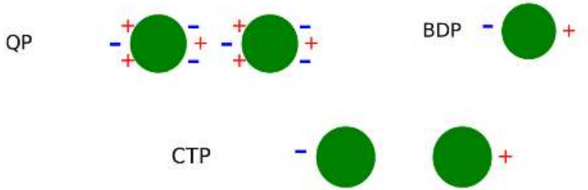
QP



BDP

Far field and near field for Na_{380}





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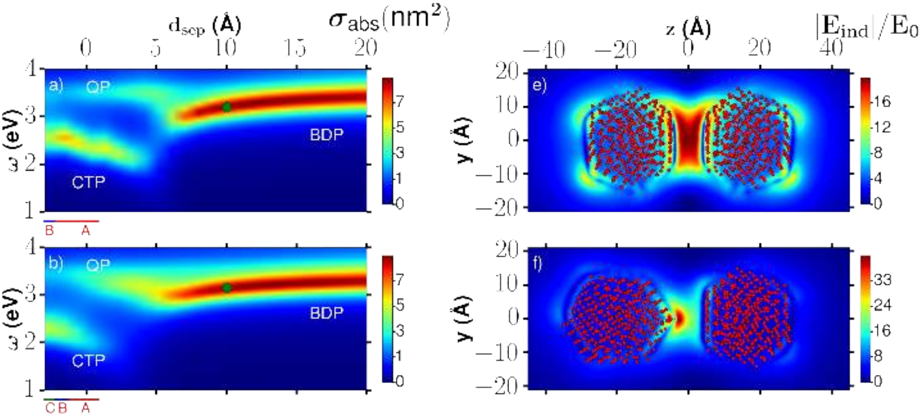

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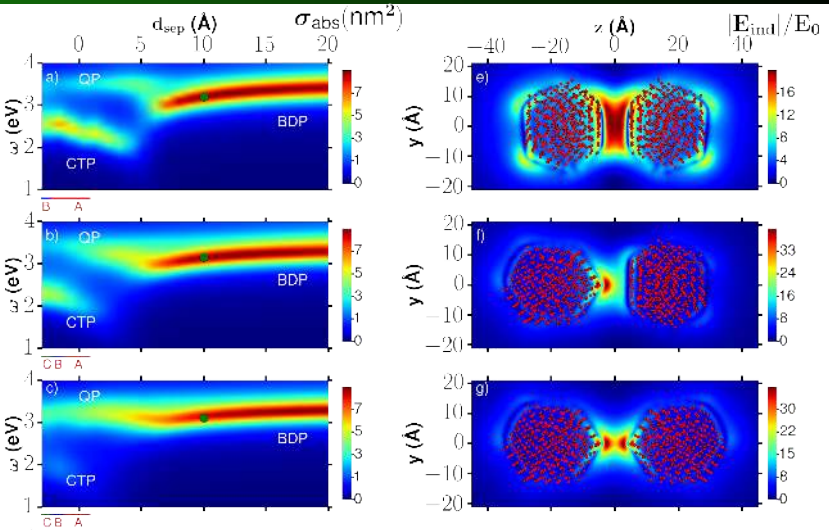
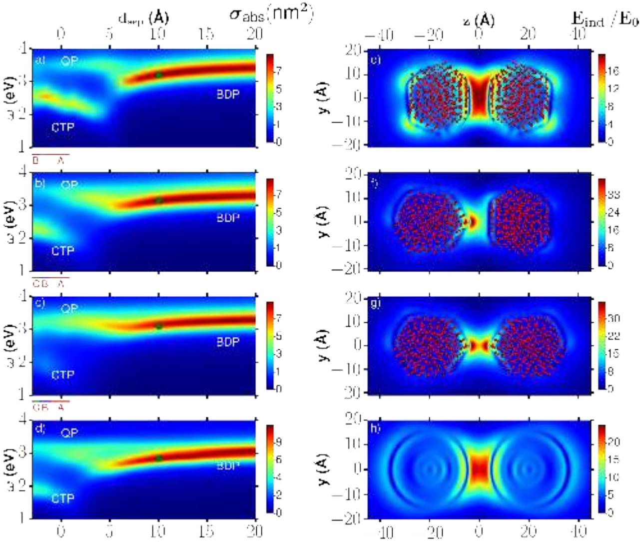
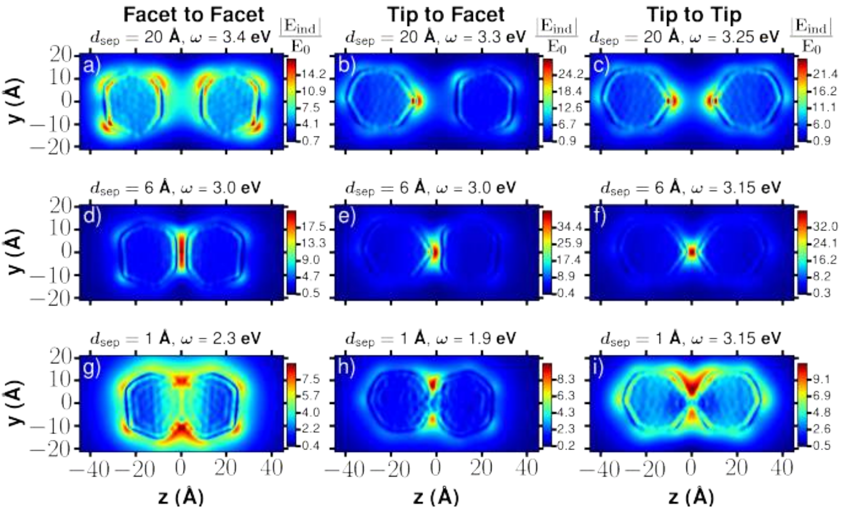


Far field and near field compared to Jellium for Na_{380}

- Facet to facet
- Tip to facet
- Tip to tip
- Jellium



The near field dependence of the Na_{380} with the clusters separation



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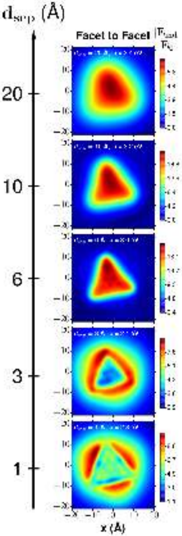

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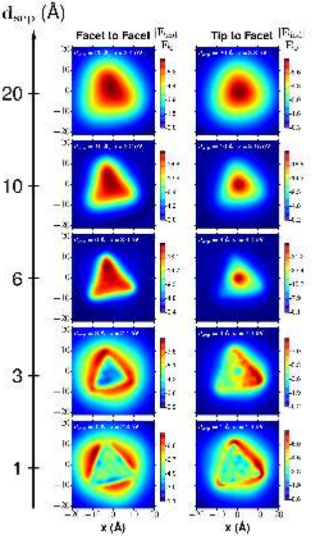

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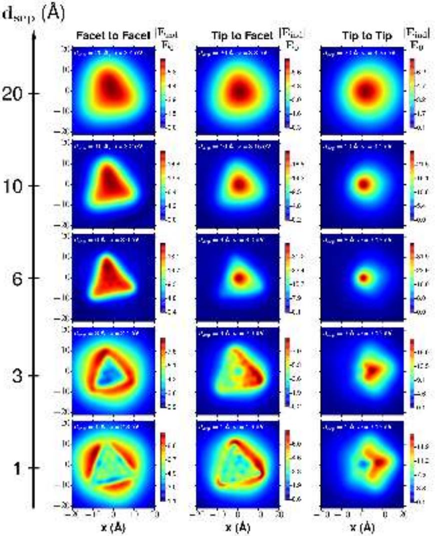

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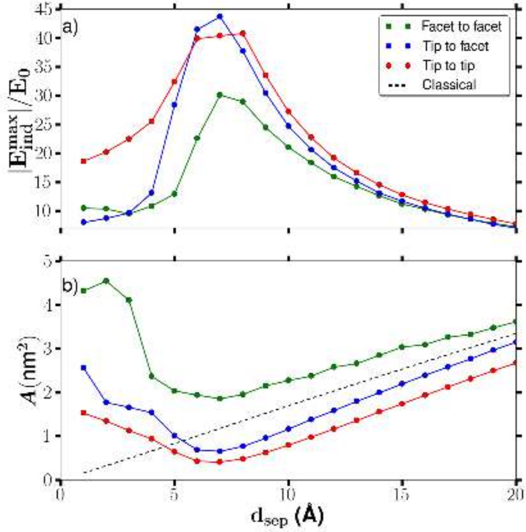

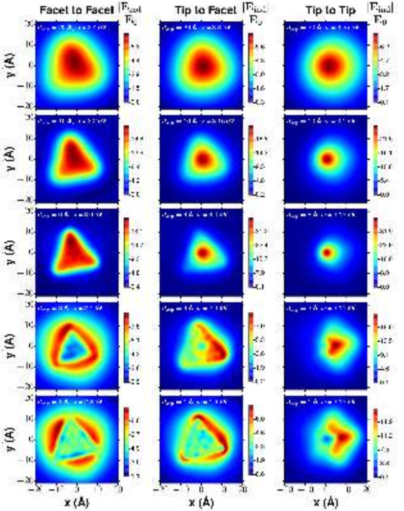

Conclusion

Ab initio TDDFT calculations of realistic models for a plasmonic nanogap:

- Our model provides quantum mechanic atomic-scale resolution of the electric field enhancement in contrast to classic[1], jellium[2]and quantum corrected[3]models.
- Thanks to this resolution we demonstrate a large dependence of the electric field enhancement on the geometrical details of the nanogap.
- With this model we wish now to look toward time dependence and EELS calculations . In order to get closer to the reality, the relaxation of the dimers is also a crucial forward step.

[1] Taylor, R. W. et al. ACS Nano 5, 3878-3887 (2011).
[2] Quijada, M. et al. Phys. Rev. A 75, 042902 (2007).
[3] Esteban R. et al. Nature comm. 3, 825 (2012)


Thank you for your attention

questions frame: TDDFT

Time-dependent Kohn-Sham equations

$$\left[-\frac{1}{2}\nabla^2 + V_{\text{eff}(\mathbf{r},t)}\right]\varphi_i(\mathbf{r},t) = i\frac{\partial}{\partial t}\varphi_i(\mathbf{r},t),$$

with the effective time-dependent potential,

$$V_{\text{eff}}(\mathbf{r},t) = V_{\text{ext}}(\mathbf{r},t) + \int \frac{n(\mathbf{r}',t)}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}' + V_{\text{xc}}(\mathbf{r},t)$$

Fast Fourier Transform

$$\mathbf{E}_{\text{ind}}(\mathbf{r},\omega) = -\mathrm{FT}^{-1}\left(\mathrm{FT}\left[\frac{\mathbf{r}}{|\mathbf{r}|^3}\right]\mathrm{FT}\left[\delta n(\mathbf{r},\omega)\right]\right)$$

questions frame: Cross section and polarizability in linear response TDDFT

Cross section σ

$$\sigma(\omega) = -\frac{4\pi\omega}{3c}\mathrm{Im}\left[P_{xx}(\omega) + P_{yy}(\omega) + P_{zz}(\omega)\right]$$

$$\text{with} \quad P_{ij}(\omega) = \int \mathbf{r}_i \chi(\mathbf{r}, \mathbf{r}', \omega)\mathbf{r}'_j d\mathbf{r} d\mathbf{r}'$$

Confinement A

$$A = \int_S \frac{|E_{\mathrm{enh}}(x, y_0, z)|^2}{|E_{\mathrm{enh}}^{\max}|^2} dxdz$$

questions frame: Interpenetration of the clusters