Plasmonic nonlocal response effects on dipole decay dynamics in the weak and strong-coupling regimes: Supplemental Material

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Strong-coupling with silver structures

The dipole spectra of silver structures are presented in Section 3.2 of the article. Here in Fig. 1 we show the dipole spectra for the same structures but as a function of ω_0 with an oscillator strength fixed to $f = 7 \times f_{\text{th}}$ in each case. In this way the oscillator strength is set well above the threshold and the strong-coupling regime is achieved. Thus anti-crossing is visible in the dipole spectra.

Strong-coupling with gold structures

We present here the dipole spectra of the same geometries as in Section 3.2 of the article but with gold rather than silver. Like in the Ag case, the effect of nonlocality in Au systems is to increase the oscillator strength threshold needed to observe Rabi splitting. For gold however, except for the cone dimer, only the lower branch is visible even at high oscillator strengths well above the estimated thresholds. A small and faint second branch emerges as seen on the maps for the local model, but we attribute this to a lower-energy resonance of the decay rate rather than to the main resonance where significant Rabi splitting is expected. The lack of observable Rabi splitting might be due to the high absorption of gold at these high frequencies. The cone dimer, a geometry with a more important field enhancement at the sharp tips, exhibits clear Rabi splitting. The decay

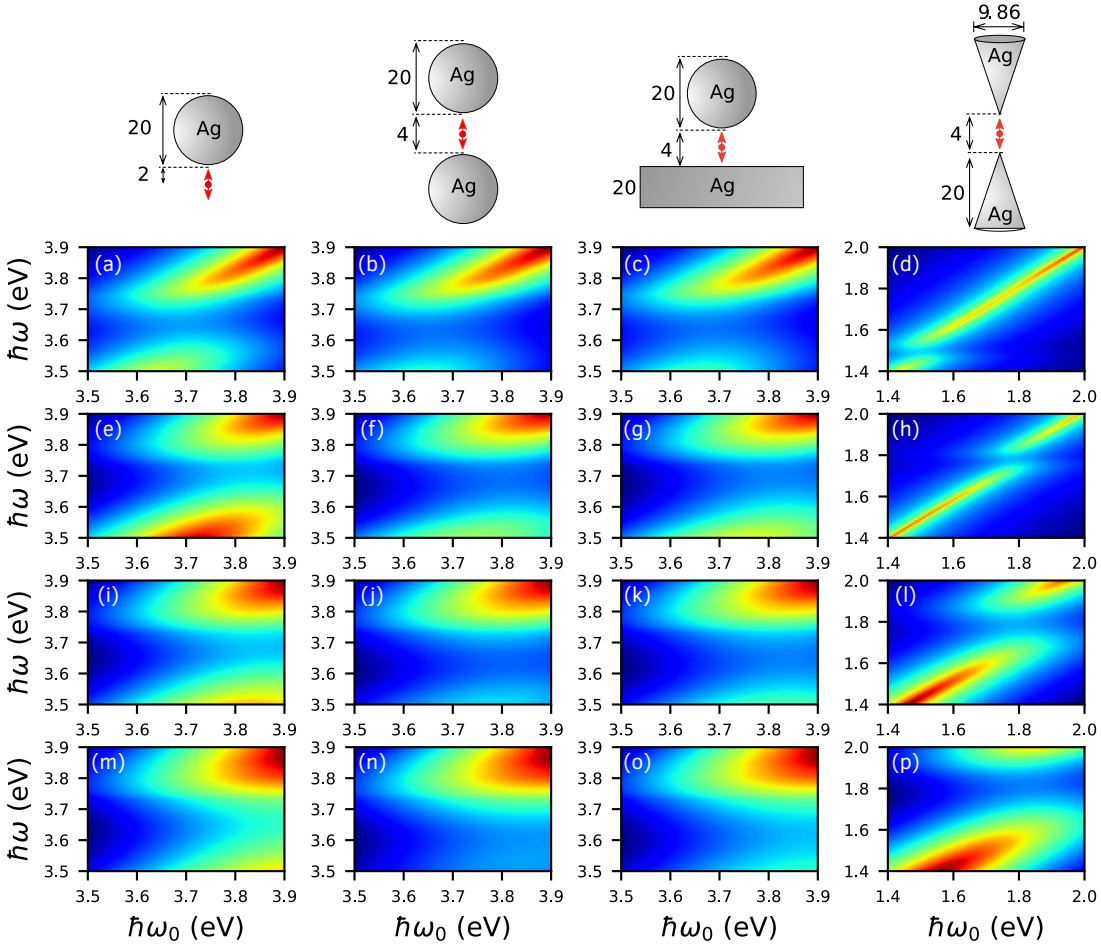

Figure 1: Dipole spectrum of an emitter in the vicinity of different Ag NP systems shown on the top (sizes are in nanometer), calculated using the local model (a-d), TF-HT model (e-h), the GNOR model with $A = 0.51$ (i-l) and the $A = 1$ (m-p), respectively. The spectra are shown as a function of the dipole transition frequency ω_0. The oscillator strength is fixed at a value above the threshold for Rabi splitting: $f = 7 \times f_{\rm th}$.

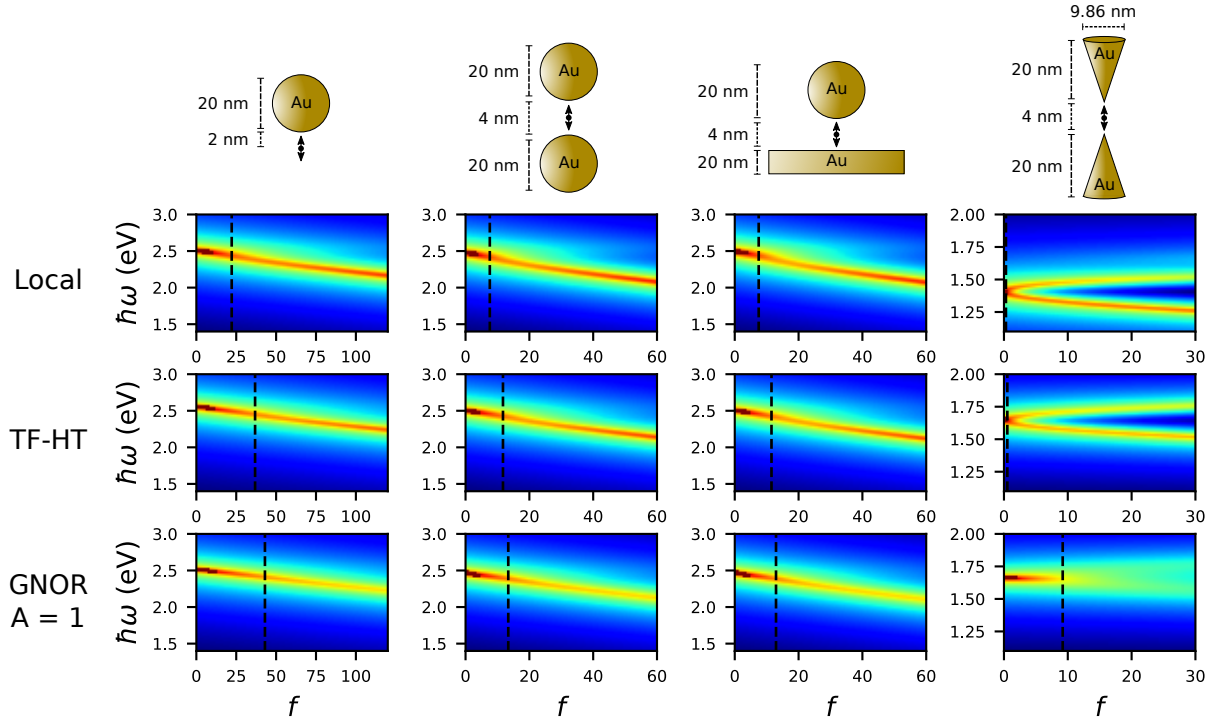

Figure 2: Dipole spectrum of a dipole in the vicinity of different gold NP systems. The dipole transition frequency is chosen to be at the peak of the total decay rate. The dashed vertical lines in the map plots indicate the oscillator strength threshold.

rate peak is at a lower frequency than for the other geometries, thus far from the spectral region of high absorption.

The dipole spectra are also shown as a function of ω_0 for a fixed oscillator strength $f = 7 \times f_{\text{th}}$ in Fig. 3. Although the oscillator strength is well above the threshold for Rabi splitting, anti-crossing is not as evident as for silver.

Dipole filed for a dipole oriented along $\hat{\mathbf{x}}$ in cylindrical coordinates

While in the article we focused on a dipole oriented along the z direction, it is still possible to apply the same 2.5D technique for an arbitrarily oriented dipole. In this case one should also consider an orthogonal direction to $\hat{\mathbf{z}}$, which without loss of generality can be assumed to be $\hat{\mathbf{x}}$. For a dipole oriented along $\hat{\mathbf{x}}$ we get:

$$
\begin{aligned}
E_\rho &= \frac{|\mathbf{p}_c|}{8\pi\varepsilon_0}\left\{\left(3\cos^2\theta - 1\right)\left(\frac{1}{r^3} - \frac{ik_0}{r^2}\right) + \frac{k_0^2}{r}\sin^2\theta\right\} e^{ik_0 r}\left(e^{i\phi} + e^{-i\phi}\right), \\
E_\phi &= \frac{|\mathbf{p}_c|}{8i\pi\varepsilon_0}\left\{\frac{1}{r^3} - \frac{ik_0}{r^2} - \frac{k_0^2}{r}\right\} e^{ik_0 r}\left(e^{i\phi} - e^{-i\phi}\right), \\
E_z &= \frac{|\mathbf{p}_c|}{8\pi\varepsilon_0}\left\{-\frac{k_0^2}{r} + \frac{3}{r^3} - \frac{3ik_0}{r^2}\right\}\sin\theta\cos\theta e^{ik_0 r}\left(e^{i\phi} + e^{-i\phi}\right),
\end{aligned}
\tag{1}
$$

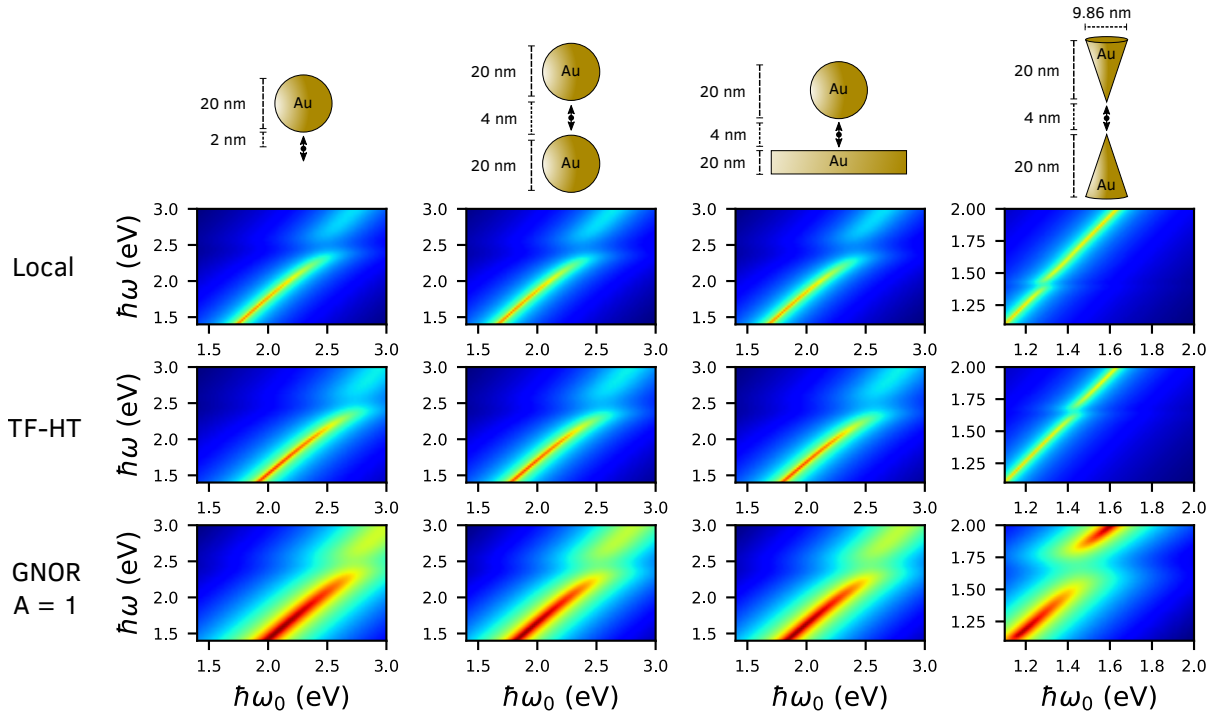

Figure 3: Dipole spectrum of a dipole in the vicinity of different gold NP systems as a function of the dipole transition frequency. The oscillator strength is fixed at $f = 7 \times f_{\mathrm{th}}$.

where analogously to Eq. (12), $r = \sqrt{\rho^2 + z^2}$ and $\theta = \arctan(\rho/z)$.

Classical decay rates near a metal sphere

For some geometries, such as a dipole near a metal sphere, an exact analytic solution for the decay rates exists within classical electrodynamics. For a dipole radially oriented with respect to the sphere, the rates normalized to their values in free space are given by [1]:

$$\frac{\gamma_{\mathrm{sp}}}{\gamma_0} = 1 + \frac{3}{2}\mathrm{Re}\sum_{l=1}^{\infty}(2l+1)\,l\,(l+1)\,b_l\left[\frac{h_l^{(1)}(kr)}{kr}\right]^2, \tag{2}$$

$$\frac{\gamma_{\mathrm{r}}}{\gamma_0} = \frac{3}{2}\mathrm{Re}\sum_{l=1}^{\infty}(2l+1)\,l\,(l+1)\left|\frac{j_l(kr) + b_l h_l^{(1)}(kr)}{kr}\right|^2, \tag{3}$$

where r is the distance from the dipole to the center of the sphere, b_l is the second Mie scattering coefficient, $h_l^{(1)}$ is the spherical Hankel function and j_l is the spherical Bessel function. Numerical implementations of these formulas are readily available [2], but they are limited by the maximum angular mode number l that they can reach. This becomes an issue when the dipole is close enough to the surface of the sphere where a high number of modes are necessary for the sums to converge. The limitation lies in the evaluation of the special functions that can yield numerical values not storable in 64-bit numerical data types. We found that implementing the computation using the NumPy and SciPy libraries allows to reach higher mode numbers with respect to other available implementations. This exact solution has been used to validate the FEM computations for the local model.

References

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