PETER

Plasmonic EPR Interaction Model

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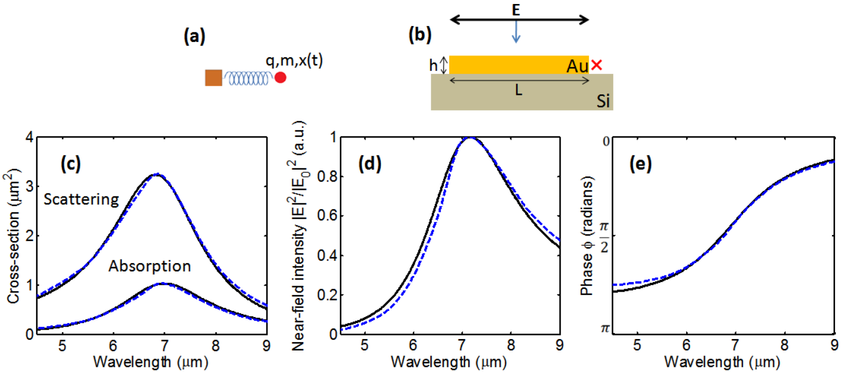
Single antenna spring model

- Tomáš Neuman et al.
  Importance of Plasmonic Scattering for an Optimal Enhancement of Vibrational Absorption in SEIRA with Linear Metallic Antennas
  The Journal of Physical Chemistry C, **119** (47), (2015)

- Mikhail A. Kats et al.
  Effect of radiation damping on the spectral response of plasmonic components
  Opt. Express **19**, 21748-21753 (2011)



$$\vec{p}\left(\omega_0^2 - \omega^2 - \mathrm{i}\omega\gamma_\mathrm{i}\right) = \alpha\left(\vec{E}_0 + \vec{E}_\mathrm{RR}\right) \quad (1)$$

$$\frac{\mathrm{d}W}{\mathrm{d}t} = \frac{\omega}{2}\mathrm{Im}\left\{\vec{p}^* \cdot \vec{E}_\mathrm{RR}\right\} = \frac{\omega\,|\vec{p}|^2}{12\pi\varepsilon_0}k^3 \qquad \Rightarrow \qquad \vec{E}_\mathrm{RR} = \mathrm{i}\frac{\omega^3 n^3}{6\pi\varepsilon_0 c^3}\vec{P} \quad (2)$$

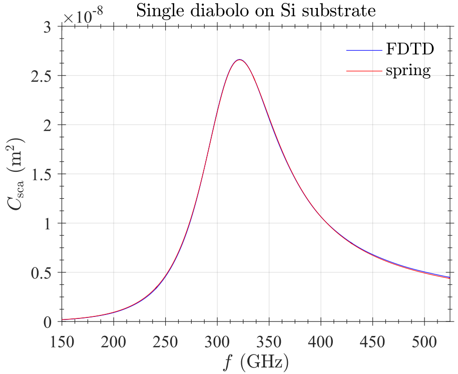
$$\vec{p} = \frac{6\pi\varepsilon_0 c^3}{n^3}\frac{\gamma_\mathrm{r}}{\omega_0^2 - \omega^2 - \mathrm{i}\omega\left(\gamma_\mathrm{i} + \gamma_\mathrm{r}\omega^2\right)}, \quad (3)$$

$$\gamma_\mathrm{r} = \frac{n^3}{6\pi\varepsilon_0 c^3}\alpha \qquad \Rightarrow \qquad \text{losses due to radiation} \quad (4)$$

Cross-sections

$$C_{\text{sca}} = \frac{\omega^4 |\vec{p}|^2}{12\pi\varepsilon_0 c^3} \,/\, I_0 = \frac{6\pi\varepsilon_0 c^2}{n^2} \frac{\gamma_{\text{r}}^2 \omega^4}{\left(\omega_0^2 - \omega^2\right)^2 + \omega^2 \left(\gamma_{\text{i}} + \gamma_{\text{r}}\omega^2\right)^2} \tag{5}$$

$$C_{\text{abs}} = -\frac{\omega}{2} \text{Im}\left\{\vec{p}^* \cdot \left(\vec{E}_0 + \vec{E}_{\text{RR}}\right)\right\} \,/\, I_0 = \frac{6\pi\varepsilon_0 c^2}{n^2} \frac{\gamma_{\text{r}}\gamma_{\text{i}}\omega^2}{\left(\omega_0^2 - \omega^2\right)^2 + \omega^2 \left(\gamma_{\text{i}} + \gamma_{\text{r}}\omega^2\right)^2} \tag{6}$$



Without magnetic material

$$C_{\text{abs}}^{(0)} = \frac{\omega}{2}\varepsilon_0\varepsilon'' \int \mathrm{d}V \left|\vec{E}\right|^2 / I_0 \tag{7}$$

$$\Downarrow$$

With magnetic material

$$C_{\text{abs}} = C_{\text{abs}}^{(0)} + \frac{\omega}{2}\mu_0\mu''(\omega) \int \mathrm{d}V \left|\vec{H}_{\parallel}\right|^2 / I_0 \tag{8}$$

Assumption: weak magnetic material (1^{st} order perturbation) ⇒ field distribution remains the same, only the overall amplitude will be scaled

$$C_{\text{abs}}^{(\text{m})} = \frac{\left|\vec{P}\right|^2}{\left|\vec{P}^{(0)}\right|^2} k\mu''(\omega) \int \mathrm{d}V \frac{\left|\vec{H}_{\parallel}^{(0)}\right|^2}{\left|\vec{H}_0\right|^2} = \frac{\left|\vec{P}\right|^2}{\left|\vec{P}^{(0)}\right|^2} k\mu''(\omega) V \eta_{\text{avg}} \tag{9}$$

We further introduce damping parameter $\gamma_{\rm m}$ in analogy with $\gamma_{\rm i}$

$$\Downarrow$$

$$\vec{p} = \frac{6\pi\varepsilon_0 c^3}{n^3} \frac{\gamma_{\rm r}}{\omega_0^2 - \omega^2 - {\rm i}\omega\left(\gamma_{\rm i} + \gamma_{\rm m} + \gamma_{\rm r}\omega^2\right)}, \tag{10}$$

Assumption: the magnetic transition is tuned to the plasmonic resonance and the linewidth of the former is much smaller than the linewidth of the latter

$$C_{\rm abs}^{\rm (m)} = \frac{6\pi c^2}{n^2} \frac{\gamma_{\rm r}\gamma_{\rm m}}{\left(\gamma_{\rm i} + \gamma_{\rm m} + \gamma_{\rm r}\omega_0^2\right)^2} = \frac{\left(\gamma_{\rm i} + \gamma_{\rm r}\omega_0^2\right)^2}{\left(\gamma_{\rm i} + \gamma_{\rm m} + \gamma_{\rm r}\omega_0^2\right)^2} k\mu''(\omega) V \eta_{\rm avg} \tag{11}$$

$$\Downarrow$$

$$\gamma_{\rm m} = k\mu''(\omega) V \eta_{\rm avg} \, \frac{n^2}{6\pi c^2} \frac{\left(\gamma_{\rm i} + \gamma_{\rm r}\omega_0^2\right)^2}{\gamma_{\rm r}} \tag{12}$$

Extinction cross-section

$$C_{\text{ext}}^{(0)} = \frac{6\pi c^2}{n^2} \frac{\gamma_{\text{r}}}{\gamma_{\text{i}} + \gamma_{\text{r}}\omega_0^2} \tag{13}$$

$$\Delta C_{\text{ext}} = -\frac{6\pi c^2}{n^2} \frac{\gamma_{\text{r}}}{\left(\gamma_{\text{i}} + \gamma_{\text{r}}\omega_0^2\right)^2} = -k\mu''(\omega) V \eta_{\text{avg}} \tag{14}$$

$$\frac{\Delta C_{\text{ext}}}{C_{\text{ext}}^{(0)}} = -\frac{\mu''(\omega)\eta_{\text{avg}}}{6\pi} \left(k^3 V\right) \frac{\frac{\gamma_{\text{i}}}{\omega_0^2} + \gamma_{\text{r}}}{\gamma_{\text{r}}} \tag{15}$$

However, we use diabolo arrays ⇒ what is different?

2D homogeneous isotropic layer: $\vec{P}(\vec{r}) = \alpha \vec{E}(\vec{r}), \qquad \alpha = \varepsilon_0 \varepsilon(\omega) \qquad (16)$

$$\vec{E}(\vec{r}) = \vec{E}_1(\vec{r}) + \int \mathrm{d}\vec{r}' \; \overleftrightarrow{G}(\vec{r} - \vec{r}')\vec{P}(\vec{r}') \tag{17}$$

$$\vec{P}(\vec{r}) = \alpha \left[\vec{E}_1(\vec{r}) + \int \mathrm{d}\vec{r}' \; \overleftrightarrow{G}(\vec{r} - \vec{r}')\vec{P}(\vec{r}') \right] \tag{18}$$

$$\Downarrow$$

Fourier space

$$\vec{P}(\vec{r}) = \int \mathrm{d}\vec{r} \; \vec{P}(\vec{q})\, e^{-\mathrm{i}\vec{q}\cdot\vec{r}}, \qquad \vec{P}(\vec{q}) = \frac{1}{(2\pi)^2} \int \mathrm{d}\vec{r} \; \vec{P}(\vec{r})\, e^{\mathrm{i}\vec{q}\cdot\vec{r}}, \tag{19}$$

$$\vec{P}(\vec{q}) = \alpha \left[\vec{E}_1(\vec{q}) + (2\pi)^2 \, \overleftrightarrow{G}(\vec{q})\vec{P}(\vec{q}) \right] \tag{20}$$

$$\text{2D structured layer:} \qquad \vec{P}(\vec{r}) = \alpha(\vec{r})\vec{E}(\vec{r}), \qquad \alpha(\vec{r}) = f(\vec{r})\varepsilon_0\varepsilon(\omega) \tag{21}$$

$$\vec{P}(\vec{r}) = \alpha(\vec{r}) \left[\vec{E}_1(\vec{r}) + \int \mathrm{d}\vec{r}\,' \; \overleftrightarrow{G}(\vec{r} - \vec{r}\,')\vec{P}(\vec{r}\,')\right] \tag{22}$$

$\Downarrow$

Fourier space

$$\vec{P}(\vec{r}) = \int \mathrm{d}\vec{r} \; \vec{P}(\vec{q})\, e^{-\mathrm{i}\vec{q}\cdot\vec{r}}, \qquad \vec{P}(\vec{q}) = \frac{1}{(2\pi)^2} \int \mathrm{d}\vec{r} \; \vec{P}(\vec{r})\, e^{\mathrm{i}\vec{q}\cdot\vec{r}}, \tag{23}$$

$$\vec{P}(\vec{q}) = \int \mathrm{d}\vec{q}\,' \; \alpha(\vec{q} - \vec{q}\,') \left[\vec{E}_1(\vec{q}\,') + (2\pi)^2\, \overleftrightarrow{G}(\vec{q}\,')\vec{P}(\vec{q}\,')\right] \tag{24}$$

$$\alpha(\vec{r}) = \sum_{mn} \alpha'(\vec{r} - \vec{R}_{mn}), \qquad \vec{\Lambda}_{mn} = \left(\frac{2m\pi}{L_x}, \frac{2n\pi}{L_y}\right) \tag{25}$$

$$\alpha(\vec{q}) = \sum_{mn} \alpha'(\vec{q}) e^{-\mathrm{i}\vec{q}\cdot\vec{R}_{mn}} = \alpha'(\vec{q}) \frac{(2\pi)^2}{L_x L_y} \sum_{mn} \delta\left(\vec{q} - \vec{\Lambda}_{mn}\right) \tag{26}$$

$$\Downarrow$$

$$\begin{aligned}\vec{P}(\vec{\Lambda}_{kl}) = \frac{(2\pi)^2}{L_x L_y} \sum_{mn} \alpha'(\vec{\Lambda}_{mn}) \Big[& \vec{E}_1 \delta\left(\vec{\Lambda}_{kl} - \vec{\Lambda}_{mn}\right) + \\ & + (2\pi)^2 \vec{\vec{G}}\left(\vec{\Lambda}_{kl} - \vec{\Lambda}_{mn}\right) \vec{P}\left(\vec{\Lambda}_{kl} - \vec{\Lambda}_{mn}\right) \Big]\end{aligned} \tag{27}$$

$$\vec{E}_{\mathrm{FF}}(\vec{r}) = (2\pi)^2 \int\limits_{|\vec{q}|<k_1,k_2} \mathrm{d}\vec{q}\;\; e^{-\mathrm{i}\vec{q}\cdot\vec{r}}\;\; \overleftrightarrow{G}_{\mathrm{FF}}(\vec{q})\,\vec{P}(\vec{q}) \tag{28}$$

Assumption: dense array $|\vec{\Lambda}_{mn}| > n\omega/c$ for $m \vee n \neq 0$

$$\Downarrow$$

Suppression of every $\vec{P}(\vec{q})$ except at $\vec{q} = 0$

$$\vec{E}_{\mathrm{FF}}(\vec{r}) = \frac{\mathrm{i}k_1}{2\varepsilon_0\varepsilon_1} t_{\mathrm{s}}\;\vec{P}(\vec{q} = 0)\, e^{\pm\mathrm{i}k_{1,2}z} \tag{29}$$

Antenna array spring model

$$\vec{P}(\vec{q}=0)\,\left(\omega_0^2-\omega^2-\mathrm{i}\omega\gamma_\mathrm{i}\right)=\alpha\left(t_\mathrm{s}\vec{E}_1+\frac{\mathrm{i}k_1}{2\varepsilon_0\varepsilon_1}t_\mathrm{s}\;\vec{P}(\vec{q}=0)\right) \tag{30}$$

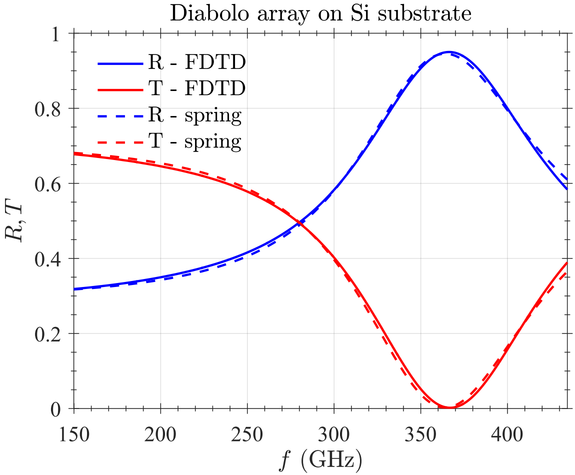
$$\vec{P}(\vec{q}=0)=\frac{2\varepsilon_0 c n_1}{\mu_1}\frac{\gamma_\mathrm{r}}{\omega_0^2-\omega^2-\mathrm{i}\omega\left(\gamma_\mathrm{i}+\gamma_\mathrm{r}\right)}, \tag{31}$$

$$T=(1-r_\mathrm{s})t_\mathrm{s}\left[1-\frac{\omega^2\left(\gamma_\mathrm{r}^2+2\gamma_\mathrm{r}\gamma_\mathrm{i}\right)}{\left(\omega_0^2-\omega^2\right)^2+\omega^2\left(\gamma_\mathrm{i}+\gamma_\mathrm{r}\right)^2}\right] \tag{32}$$

$$R=r_\mathrm{s}^2+t_\mathrm{s}\frac{\omega^2\left[(1-r_\mathrm{s})\gamma_\mathrm{r}^2-2r_\mathrm{s}\gamma_\mathrm{r}\gamma_\mathrm{i}\right]}{\left(\omega_0^2-\omega^2\right)^2+\omega^2\left(\gamma_\mathrm{i}+\gamma_\mathrm{r}\right)^2} \tag{33}$$

$$A=2t_\mathrm{s}^2\frac{\omega^2\gamma_\mathrm{r}\gamma_\mathrm{i}}{\left(\omega_0^2-\omega^2\right)^2+\omega^2\left(\gamma_\mathrm{i}+\gamma_\mathrm{r}\right)^2} \tag{34}$$

$$\gamma_{\mathrm{r}} \approx 0.31\omega_0, \qquad \gamma_{\mathrm{i}} \approx 0.02\omega_0 \tag{35}$$



Assumptions:

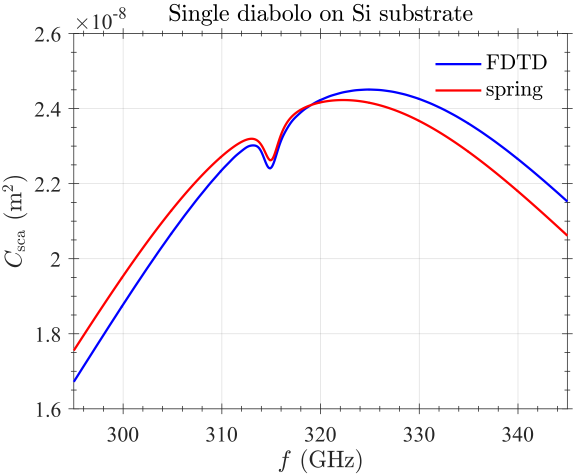
- homogeneous thin magnetic layer with relative permeability $\mu(\omega)$ that can be described by a Lorentz model with linewidth much smaller than the linewidth of the plasmonic resonance
- apart from amplitude scaling, the magnetic interaction does not affect the field distribution of the diabolo array

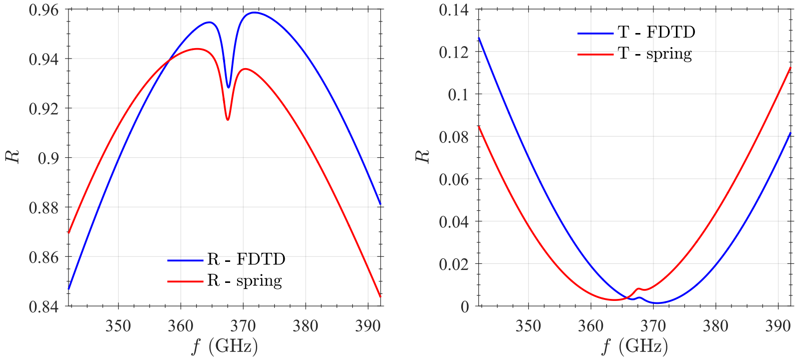
$\Downarrow$

$$\frac{\Delta T}{T^{(0)}} = \frac{k_1 d \mu''(\omega)}{\mu_1} \left[\frac{\gamma_\mathrm{r} + \gamma_\mathrm{i}}{\gamma_\mathrm{i}} \frac{\eta_\mathrm{avg}}{t_\mathrm{s}} - \frac{2\gamma_\mathrm{r} + (1 - r_\mathrm{s})\,\gamma_\mathrm{i}}{\gamma_\mathrm{i}} \right] \tag{36}$$

$$\frac{\Delta R}{R^{(0)}} = -\frac{k_1 d \mu''(\omega)}{\mu_1} \left[\eta_\mathrm{avg} \frac{\gamma_\mathrm{r} + \gamma_\mathrm{i}}{\gamma_\mathrm{r} - r_\mathrm{s}\gamma_\mathrm{i}} + (1 - r_\mathrm{s}) \left(1 + \frac{\gamma_\mathrm{r} + \gamma_\mathrm{i}}{\gamma_\mathrm{r} - r_\mathrm{s}\gamma_\mathrm{i}} \right) \right] \tag{37}$$

Single diabolo - FDTD vs. spring model




0.96
0.94
0.92
0.9
0.88
0.86
0.84
R
350
360
370
380
390
f (GHz)
R - FDTD
R - spring
0.14
0.12
0.1
0.08
0.06
0.04
0.02
0
R
350
360
370
380
390
f (GHz)
T - FDTD
T - spring