Supplementary Material to the article "Near-field electromagnetic coupling in a nanosphere with a plasmonic core and an outer excitonic shell"

I. COEFFICIENTS OF THE SYSTEM OF EQUATIONS FOR CALCULATING PLASMON-EXCITON COUPLING CONSTANTS

Below we present the expressions for the coefficients $P_n^{(i)}$ and Q_n that appear in the system of the nonlinear equations (14). Solving this system yields the plasmon-exciton coupling constants g_+ and g_- as well as the frequencies $\breve{\omega}_{\rm pl} = \omega_{\rm pl} + \upsilon_{\rm pl}$, $\breve{\omega}_{\rm ex}^{+} = \omega_{\rm ex}^{+} + \upsilon_{\rm ex}^{+}$, and $\breve{\omega}_{\rm ex}^{-} = \omega_{\rm ex}^{-} + \upsilon_{\rm ex}^{-}$ shifted from their unperturbed values, $\omega_{\rm pl}$ and $\omega_{\rm ex}^{\pm}$, due to the electromagnetic interaction between the core and shell. The coefficients $P_n^{(i)}$ on the left-hand side of the system of equations (14) are given by

$$P_1^{(1)} = P_1^{(2)} = P_1^{(3)} = 1, \quad P_1^{(4)} = \cdots = P_1^{(11)} = 0, \tag{S1}$$

$$P_2^{(1)} = 2\gamma_{\rm ex}, \qquad P_2^{(2)} = P_2^{(3)} = \gamma_{\rm ex} + \gamma_{\rm pl}, \qquad P_2^{(4)} = \cdots = P_2^{(11)} = 0, \tag{S2}$$

$$P_3^{(1)} = \gamma_{\rm ex}^2, \quad P_3^{(2)} = P_3^{(3)} = \gamma_{\rm ex}\gamma_{\rm pl}, \quad P_3^{(4)} = P_3^{(5)} = P_3^{(6)} = 1, \quad P_3^{(7)} = P_3^{(8)} = -4, \quad P_3^{(9)} = P_3^{(10)} = P_3^{(11)} = 0, \tag{S3}$$

$$P_4^{(1)} = P_4^{(2)} = P_4^{(3)} = P_4^{(9)} = P_4^{(10)} = P_4^{(11)} = 0, \quad P_4^{(4)} = P_4^{(5)} = \gamma_{\rm ex}, \quad P_4^{(6)} = \gamma_{\rm pl}, \quad P_4^{(7)} = P_4^{(8)} = -4\gamma_{\rm ex}, \tag{S4}$$

$$P_5^{(1)} = \cdots = P_5^{(8)} = 0, \quad P_5^{(9)} = 1, \qquad P_5^{(10)} = P_5^{(11)} = -4. \tag{S5}$$

The coefficients Q_n on the right-hand side of Eqs. (14) can be expressed in terms of the basic parameters of the system: the permittivities of the metal core (4), the excitonic shell (6), and the surrounding medium as well as the volumes of the core $V_{\rm c}$, the shell $V_{\rm sh}$, and the total volume of the particle V. After a series of transformations, these coefficients take the following form

$$\begin{aligned} Q_1 = {} & \frac{3\varepsilon_{\rm sh}^{\infty} V_{\rm c} + (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty} (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}} \omega_{\rm p}^2 + \\ & \frac{3(f + 2\varepsilon_{\rm sh}^{\infty})(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + [f(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) + 2(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})(\varepsilon_{\rm m}^{\infty} + f + 2\varepsilon_{\rm sh}^{\infty})] V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty} (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}} \omega_0^2, \end{aligned} \tag{S6}$$

$$\begin{aligned} Q_2 = {} & \frac{3\varepsilon_{\rm sh}^{\infty} V_{\rm c} + (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty} (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}} \left(2\gamma_{\rm ex}\omega_{\rm p}^2\right) + \\ & \frac{3(f + 2\varepsilon_{\rm sh}^{\infty})(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + [f(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) + 2(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})(\varepsilon_{\rm m}^{\infty} + f + 2\varepsilon_{\rm sh}^{\infty})] V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty} (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}} (\gamma_{\rm pl} + \gamma_{\rm ex}) \omega_0^2, \end{aligned} \tag{S7}$$

$$\begin{aligned} Q_3 = {} & \frac{[3\varepsilon_{\rm sh}^{\infty} V_{\rm c} + (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}] \gamma_{\rm ex}^2 + [3(f + 2\varepsilon_{\rm sh}^{\infty}) V_{\rm c} + (f + 2\varepsilon_{\rm sh}^{\infty} + 4\varepsilon_{\rm h}) V_{\rm sh}] \omega_0^2}{3\varepsilon_{\rm sh}^{\infty} (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}} \omega_{\rm p}^2 + \\ & \frac{3(f + 2\varepsilon_{\rm sh}^{\infty})(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + [f(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) + 2(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})(\varepsilon_{\rm m}^{\infty} + f + 2\varepsilon_{\rm sh}^{\infty})] V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty} (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}} \gamma_{\rm pl}\gamma_{\rm ex}\omega_0^2 + \\ & \frac{3(f + \varepsilon_{\rm sh}^{\infty})(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V + 2(f + \varepsilon_{\rm sh}^{\infty} - \varepsilon_{\rm h})(f + \varepsilon_{\rm sh}^{\infty} - \varepsilon_{\rm m}^{\infty}) V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty} (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}} \omega_0^4, \end{aligned} \tag{S8}$$

$$\begin{aligned} Q_4 = {} & \frac{3(f + \varepsilon_{\rm sh}^{\infty})(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V + 2(f + \varepsilon_{\rm sh}^{\infty} - \varepsilon_{\rm h})(f + \varepsilon_{\rm sh}^{\infty} - \varepsilon_{\rm m}^{\infty}) V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty} (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}} \gamma_{\rm pl}\omega_0^4 + \\ & \frac{3(f + 2\varepsilon_{\rm sh}^{\infty}) V_{\rm c} + (f + 2\varepsilon_{\rm sh}^{\infty} + 4\varepsilon_{\rm h}) V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty} (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}} \gamma_{\rm ex}\omega_{\rm p}^2\omega_0^2, \end{aligned} \tag{S9}$$

$$Q_5 = \frac{3(f + \varepsilon_{\rm sh}^{\infty}) V_{\rm c} + (f + \varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty} (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}) V_{\rm sh}} \omega_{\rm p}^2\omega_0^4. \tag{S10}$$

II. DETAILS OF THE DERIVATION OF FORMULAS FOR COUPLING CONSTANTS AND SHIFTS OF PLASMONIC AND EXCITONIC RESONANCE FREQUENCIES

Here we outline a technique for solving the system of equations (14), which is used to calculate the plasmon-exciton coupling constants g_+ and g_- in core-shell hybrid nanoparticles and to determine the frequencies $\breve{\omega}_{\rm pl}$ and $\breve{\omega}_{\rm ex}^{\pm}$ shifted

from their unperturbed values due to the electromagnetic coupling between the core and shell. It should be noted that the expression for the shifted plasmon frequency can be derived directly from equations with the indices of $n = 1$ and 2 of the system (14). This leads to the relation

$$\breve{\omega}_{\rm pl}^2 = \frac{3\varepsilon_{\rm sh}^{\infty} V_{\rm c} + (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty}\,(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty})\,(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm sh}}\omega_{\rm p}^2. \tag{S11}$$

The corresponding shift relative to the unperturbed Fröhlich frequency (5) is equal to

$$\upsilon_{\rm pl} = \breve{\omega}_{\rm pl} - \omega_{\rm pl} = \omega_{\rm p}\left\{\left[\frac{3\varepsilon_{\rm sh}^{\infty} V_{\rm c} + (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty}\,(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty})\,(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm sh}}\right]^{1/2} - \frac{1}{\sqrt{\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}}}\right\}. \tag{S12}$$

In the limiting case of $\varepsilon_{\rm sh}^{\infty} \to \varepsilon_{\rm h}$ we obtain the Fröhlich frequency of the bare metal core, $\breve{\omega}_{\rm pl} \to \omega_{\rm pl}$. Note that the frequency $\breve{\omega}_{\rm pl}$ is equal to the plasmon resonance frequency of the nanoparticle consisting of the metal core and the passive shell with a permittivity $\varepsilon = \varepsilon_{\rm sh}^{\infty}$.

The system of equations for determining the shifted frequencies $\breve{\omega}_{\rm ex}^{+}$ and $\breve{\omega}_{\rm ex}^{-}$ is derived from equations with the indices of $n = 1$, 2, and 3 of the system (14). This system can be written as

$$\left(\breve{\omega}_{\rm ex}^{+}\right)^2 + \left(\breve{\omega}_{\rm ex}^{-}\right)^2 = \frac{3\,(f + 2\varepsilon_{\rm sh}^{\infty})\,(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm c} + [f\,(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty}) + 2\,(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\,(\varepsilon_{\rm m}^{\infty} + f + 2\varepsilon_{\rm sh}^{\infty})]\, V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty}\,(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty})\,(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm sh}}\omega_0^2, \tag{S13}$$

$$\left(\breve{\omega}_{\rm ex}^{+}\breve{\omega}_{\rm ex}^{-}\right)^2 = \frac{3\,(f + \varepsilon_{\rm sh}^{\infty})\,(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h})\, V + 2\,(f + \varepsilon_{\rm sh}^{\infty} - \varepsilon_{\rm h})\,(f + \varepsilon_{\rm sh}^{\infty} - \varepsilon_{\rm m}^{\infty})\, V_{\rm sh}}{3\varepsilon_{\rm sh}^{\infty}\,(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty})\,(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm sh}}\omega_0^4. \tag{S14}$$

Further from solving equations (S13) and (S14), we obtain the expression for the shifted frequencies of the excitonic modes of the shell

$$\left(\frac{\breve{\omega}_{\rm ex}^{\pm}}{\omega_0}\right)^2 = 1 + \frac{f\left[X \mp \sqrt{X^2 + 8\frac{V_{\rm c}}{V}} - 4\frac{V_{\rm c}}{V}\right]}{(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\left[X \mp \sqrt{X^2 + 8\frac{V_{\rm c}}{V}}\right] - 4\frac{V_{\rm c}}{V}\,(\varepsilon_{\rm sh}^{\infty} - \varepsilon_{\rm h})}, \quad X = 1 - 2\frac{V_{\rm sh}}{V}\frac{\varepsilon_{\rm m}^{\infty} - \varepsilon_{\rm h}}{\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h}}. \tag{S15}$$

The respective shifts of the frequencies of the excitonic shell modes relative to their unperturbed values (8) are given by

$$\upsilon_{\rm ex}^{\pm} = \breve{\omega}_{\rm ex}^{\pm} - \omega_{\rm ex}^{\pm} = \omega_0\left\{1 + \frac{f\left[X \mp \sqrt{X^2 + 8\frac{V_{\rm c}}{V}} - 4\frac{V_{\rm c}}{V}\right]}{(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\left[X \mp \sqrt{X^2 + 8\frac{V_{\rm c}}{V}}\right] - 4\frac{V_{\rm c}}{V}\,(\varepsilon_{\rm sh}^{\infty} - \varepsilon_{\rm h})}\right\}^{1/2} - \omega_0\left\{1 + \frac{f\left[1 \mp \sqrt{1 + 8\frac{V_{\rm c}}{V}} - 4\frac{V_{\rm c}}{V}\right]}{(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\left[1 \mp \sqrt{1 + 8\frac{V_{\rm c}}{V}}\right] - 4\frac{V_{\rm c}}{V}\,(\varepsilon_{\rm sh}^{\infty} - \varepsilon_{\rm h})}\right\}^{1/2}. \tag{S16}$$

Here ω_0 and f are the central frequency and the effective oscillator strength of the excitonic transition, respectively, while $\varepsilon_{\rm m}^{\infty}$ and $\varepsilon_{\rm sh}^{\infty}$ are the background components of the permittivities of the metal core and the excitonic shell. Note that the frequencies $\breve{\omega}_{\rm ex}^{\pm}$ correspond to the resonance frequencies of the excitonic shell of the particle with a core with a constant permittivity $\varepsilon = \varepsilon_{\rm m}^{\infty}$. The shifts $\upsilon_{\rm ex}^{\pm}$ vanish when $\varepsilon_{\rm m}^{\infty} = \varepsilon_{\rm h}$.

Finally, from the equations with the indices of $n = 4$ and 5 of the system (14) we have two relations

$$g_{+}^2\breve{\omega}_{\rm ex}^{+}\breve{\omega}_{\rm pl} + g_{-}^2\breve{\omega}_{\rm ex}^{-}\breve{\omega}_{\rm pl} = \frac{3\left[(\varepsilon_{\rm sh}^{\infty})^2 + 2\,(\varepsilon_{\rm h})^2\right] V_{\rm c} + (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})^2\, V_{\rm sh}}{2\left[3\varepsilon_{\rm sh}^{\infty}\,(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty})\,(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm sh}\right]^2} f V_{\rm sh}\omega_0^2\omega_{\rm p}^2, \tag{S17}$$

$$g_{+}^2\breve{\omega}_{\rm ex}^{+}\breve{\omega}_{\rm pl}(\breve{\omega}_{\rm ex}^{-})^2 + g_{-}^2\breve{\omega}_{\rm ex}^{-}\breve{\omega}_{\rm pl}(\breve{\omega}_{\rm ex}^{+})^2 = \frac{3\left(\varepsilon_{\rm sh}^{\infty 2} + f\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h}^2\right) V_{\rm c} + (\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\,(\varepsilon_{\rm sh}^{\infty} + f + 2\varepsilon_{\rm h})\, V_{\rm sh}}{2\left[3\varepsilon_{\rm sh}^{\infty}\,(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty})\,(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm sh}\right]^2} f V_{\rm sh}\omega_0^4\omega_{\rm p}^2 \tag{S18}$$

for determining the sought plasmon-exciton coupling constants. Expressions (S17) and (S18) yield

$$g_{\pm}^2 = \frac{f V_{\rm sh} V\,(\mathcal{A} \pm \mathcal{B})\,(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})^2\left(\omega_0^2/\breve{\omega}_{\rm ex}^{\pm}\right)\left(\omega_{\rm p}^2/\breve{\omega}_{\rm pl}\right)}{4\left[3\varepsilon_{\rm sh}^{\infty}\,(\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm c} + (\varepsilon_{\rm m}^{\infty} + 2\varepsilon_{\rm sh}^{\infty})\,(\varepsilon_{\rm sh}^{\infty} + 2\varepsilon_{\rm h})\, V_{\rm sh}\right]^2}, \tag{S19}$$

$$\mathcal{A} = 1 + 2\frac{V_\mathrm{c}}{V}\left(\frac{\varepsilon_\mathrm{sh}^\infty - \varepsilon_\mathrm{h}}{\varepsilon_\mathrm{sh}^\infty + 2\varepsilon_\mathrm{h}}\right)^2, \qquad \mathcal{B} = \frac{X\left[1 - 2\frac{V_\mathrm{c}}{V}\left(\frac{\varepsilon_\mathrm{sh}^\infty - \varepsilon_\mathrm{h}}{\varepsilon_\mathrm{sh}^\infty + 2\varepsilon_\mathrm{h}}\right)^2\right] - 8\frac{V_\mathrm{c}}{V}\frac{\varepsilon_\mathrm{sh}^\infty - \varepsilon_\mathrm{h}}{\varepsilon_\mathrm{sh}^\infty + 2\varepsilon_\mathrm{h}}}{\sqrt{X^2 + 8\frac{V_\mathrm{c}}{V}}}. \tag{S20}$$

The coupling constants are primarily determined by resonant parts of the polarizabilities of the metal core and the excitonic shell. However, they also depend on non-resonant parts of the polarizabilities, which are determined by the values of $\varepsilon_\mathrm{m}^\infty$ and $\varepsilon_\mathrm{sh}^\infty$. The final expressions for the quantities $\breve{\omega}_\mathrm{pl}$, $\breve{\omega}_\mathrm{ex}^\pm$, and $g_\pm$ are also provided in the article [see Eqs. (15)–(18)].