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# Electrodynamical enhancement of optical transitions in semiconductor and metal–semiconductor nanostructures

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**Abstract**

Semiconductor and metal–semiconductor nanostructures are shown to exhibit electrodynamical resonances analogous to the Fröhlich resonance for metal nanoparticles in a dielectric host. If the transition frequency of an optical transition in the nanostructure core coincides with one of the resonance frequencies of the nanostructure, the strength of the optical transition is dramatically enhanced by up to 4–6 orders of magnitude. The resonance frequencies are determined by dielectric permittivities of materials of host and nanostructure, and by sizes of the nanostructure. That enables to tune the resonance frequencies to desired values in an extremely wide spectral range—from ultraviolet to terahertz, engineering thus optical properties of high-efficiency nanostructured optical materials for numerous applications.
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## 1. Introduction

The rate of radiative spontaneous decay, $\Gamma_0$, of a dipole transition of frequency $\omega$ and dipole momentum $d$ in a point-like emitter, such an atom or a molecule, embedded into a dielectric host of permittivity $\varepsilon_{\text{host}}$ is described by well-known expression [1] (speed of light $c = 1$):

$$\Gamma_0 = \frac{4}{3}\omega^3 d^2 \sqrt{\varepsilon_{\text{host}}}. \tag{1.1}$$

However, in the case of colloidal semiconductor nanocrystals experiments show an essential depression of the decay compared to the value given in the above equation. To explain this depression, Wehrenberg, Wang, and Guyot-Sionnest [2] noted that Eq. (1.1), derived for point-like emitters, does not take into account the local field effect, or the screening of the radiation field inside a nanocrystal of finite size. In a spherical nanocrystal of permittivity $\varepsilon_{\text{NC}}$ embedded into a dielectric host of permittivity $\varepsilon_{\text{host}}$, the internal electric field (inside the nanocrystal) $\mathbf{E}_{\text{int}}$ is weaker in comparison with a uniform external field $\mathbf{E}_{\text{ext}}$ in the host [1]:

$$\mathbf{E}_{\text{int}} = \frac{3}{2 + \varepsilon_{\text{NC}}/\varepsilon_{\text{host}}}\mathbf{E}_{\text{ext}}. \tag{1.2}$$

Therefore, the expression (1.1) should be replaced by:

$$\Gamma = \frac{4}{3}\omega^3 d^2 |S|^2 \sqrt{\varepsilon_{\text{host}}} \equiv \Gamma_0 |S|^2, \tag{1.3}$$

where $\Gamma_0$ is the decay rate for a point-like emitter (1.1), and

$$S = \frac{3}{2 + \varepsilon_{\text{NC}}/\varepsilon_{\text{host}}} \tag{1.4}$$

is the screening factor. The estimates of radiative lifetime done with Eqs. (1.3) and (1.4) for PbSe and CdSe colloidal nanocrystals [2] show a good agrement with experimental data. Despite the radiation field is obviously strongly nonuniform, accurate computations [3] of the decay rate based on the Maxwell equations and conventional boundary condition for the radiation field on the nanocrystal–host interface just reproduce Eqs. (1.3) and (1.4). While for core–shell nanocrystals, the developed approach [3] results in the following expressions for the decay rate of an optical transition localized in the core:

$$\Gamma = \frac{4}{3}\omega^3 d^2 |W|^2 \sqrt{\varepsilon_{\text{host}}} = \Gamma_0 |W|^2, \tag{1.5}$$

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where the screening factor $W$ was found to be:

$$W = \frac{9}{\left(2+\frac{\varepsilon_{\text{core}}}{\varepsilon_{\text{shell}}}\right)\left(2+\frac{\varepsilon_{\text{shell}}}{\varepsilon_{\text{host}}}\right)+2\left(1-\frac{\varepsilon_{\text{core}}}{\varepsilon_{\text{shell}}}\right)\left(1-\frac{\varepsilon_{\text{shell}}}{\varepsilon_{\text{host}}}\right)\left(\frac{R}{R+\Delta}\right)^3}. \tag{1.6}$$

Here, $\varepsilon_{\text{core}}$, $\varepsilon_{\text{shell}}$ and $\varepsilon_{\text{host}}$ are permittivities of the core, shell and host, while $R$ and $\Delta$ are respectively the radius of the core and the thickness of the shell. In the case of uniform external field, the above expression was derived in Ref. [1] for the particular case $\varepsilon_{\text{core}} = \varepsilon_{\text{host}} = 1$.

## 2. Bare nanocrystals

### 2.1. Fröhlich resonance

If the nanocrystal permittivity is greater than the host permittivity, $\varepsilon_{\text{NC}} > \varepsilon_{\text{host}}$, the factor $S$ describes the depression of the decay due to screening of the radiation field inside the nanocrystal. Fröhlich was the first to note [4,5] that for a metal nanocrystal with the complex frequency-dependent dielectric function

$$\varepsilon_{\text{NC}}(\omega) = \operatorname{Re}\varepsilon_{\text{NC}}(\omega) + i\operatorname{Im}\varepsilon_{\text{NC}}(\omega) \tag{2.1}$$

the system metal nanocrystal plus dielectric host exhibits a resonance at the frequency $\omega = \Omega$ when the real part of the denominator in the expression (1.4) vanishes:

$$\operatorname{Re}\varepsilon_{\text{NC}}(\Omega) = -2\varepsilon_{\text{host}}. \tag{2.2}$$

At the resonance frequency, the screening factor is found to be:

$$S(\Omega) = \frac{3i}{2}\frac{\operatorname{Re}\varepsilon_{\text{NC}}(\Omega)}{\operatorname{Im}\varepsilon_{\text{NC}}(\Omega)}. \tag{2.3}$$

If $\operatorname{Re}\varepsilon_{\text{NC}}(\Omega) \gg \operatorname{Im}\varepsilon_{\text{NC}}(\Omega)$, the above expression results in a huge resonance enhancement of the radiation field inside metal nanocrystals.

In our case of semiconductor nanocrystals, the real part of the dielectric function (2.1) is negative in the vicinity of the transverse–longitudinal splitting, i.e. in the frequency range between the transverse, $\Omega_T$, and longitudinal, $\Omega_L$, frequencies of an optical phonon, $\Omega_T < \omega < \Omega_L$. In this frequency range, the dielectric function is well modeled by the expression:

$$\varepsilon_{\text{NC}}(\omega) = \varepsilon_\infty \frac{\omega^2 - \Omega_L^2 + i\omega\gamma}{\omega^2 - \Omega_T^2 + i\omega\gamma}, \tag{2.4}$$

where $\varepsilon_\infty$ is the high-frequency permittivity (at $\omega \gg \Omega_L$), and $\gamma$ is the relaxation parameter. Separating the real and imaginary parts of the dielectric function:

$$\begin{aligned}\operatorname{Re}\varepsilon_{\text{NC}}(\omega) &= \varepsilon_\infty \frac{(\omega^2-\Omega_L^2)(\omega^2-\Omega_T^2)+(\gamma\omega)^2}{(\omega^2-\Omega_T^2)^2+(\gamma\omega)^2} \\ &\simeq \varepsilon_\infty \frac{\omega^2-\Omega_L^2}{\omega^2-\Omega_T^2},\end{aligned} \tag{2.5}$$

$$\operatorname{Im}\varepsilon_{\text{NC}}(\omega) = \varepsilon_\infty \frac{\gamma\omega(\Omega_L^2-\Omega_T^2)}{(\omega^2-\Omega_T^2)^2+(\gamma\omega)^2} \simeq \varepsilon_\infty \frac{\gamma\omega(\Omega_L^2-\Omega_T^2)}{(\omega^2-\Omega_T^2)^2}, \tag{2.6}$$

we find from Eq. (2.2) the following equation for the Fröhlich resonance frequency $\Omega$:

$$\varepsilon_\infty \frac{\Omega^2 - \Omega_L^2}{\Omega^2 - \Omega_T^2} = -2\varepsilon_{\text{host}}. \tag{2.7}$$

In the second approximate equities in Eqs. (2.5) and (2.6), we took into account that, as a rule, the relaxation parameter $\gamma$ is much smaller than $\Omega_T$ and $\Omega_L$, while the frequency $\omega$ is sufficiently far from both $\Omega_T$ and $\Omega_L$.

If the frequency of an optical transition in the nanocrystal $\Omega_{\text{TR}}$ equals to the resonance frequency $\Omega$, $\Omega_{\text{TR}} = \Omega$, the radiative decay rate of the transition is enhanced by the gain factor $G(\Omega)$:

$$\Gamma(\Omega) = \Gamma_0 G(\Omega), \tag{2.8}$$

where

$$G(\Omega) = \left|S(\Omega)\right|^2 = \left|\frac{3i}{2}\frac{\operatorname{Re}\varepsilon_{\text{NC}}(\Omega)}{\operatorname{Im}\varepsilon_{\text{NC}}(\Omega)}\right|^2. \tag{2.9}$$

Frequencies of optical vibrations in different semiconductor materials range from approximately 10 microns to submillimeters. For example, in silicon carbide (SiC) $\Omega_T = 23.8$ THz and $\Omega_L = 29.1$ THz, while in lead selenide (PbSe), $\Omega_T = 1.96$ THz and $\Omega_L = 6.14$ THz [6]. That obviously enables to design high-efficiency optical media operating from mid-IR to terahertz-frequency ranges.

### 2.2. SiC nanocrystals

As an example, we estimate the resonance enhancement of an intraband optical transition in silicon carbide (SiC) nanocrystals. The dielectric function of SiC is well modeled by the expressions (2.4)–(2.6) with $\varepsilon_\infty = 6.52$, $\Omega_T = 793.9\ \text{cm}^{-1}$ (and the wavelength $\Lambda_T = 12.6\ \mu\text{m}$), $\Omega_L = 970.1\ \text{cm}^{-1}$ ($\Lambda_L = 10.3\ \mu\text{m}$), and $\gamma = 4.763\ \text{cm}^{-1}$ [7]. Note that the relaxation parameter $\gamma$ is much less than the optical phonon frequencies, $\gamma/\Omega_T \simeq 0.006$ and $\gamma/\Omega_L \simeq 0.005$.

The solution of the resonance condition (2.7) results in:

$$\Omega = \Omega_L\left[\frac{1+2(\varepsilon_{\text{host}}/\varepsilon_0)}{1+2(\varepsilon_{\text{host}}/\varepsilon_\infty)}\right]^{1/2} = \Omega_T\left[\frac{\varepsilon_0+2\varepsilon_{\text{host}}}{\varepsilon_\infty+2\varepsilon_{\text{host}}}\right]^{1/2}, \tag{2.10}$$

where

$$\varepsilon_0 = \varepsilon_\infty\left(\frac{\Omega_L}{\Omega_T}\right)^2 = 9.73$$

is the static permittivity of SiC.

Here and hereafter, in all our numerical estimates we accept the refractive index of a host matrix $n_{\text{host}} = 1.5$ (and hence, the host permittivity $\varepsilon_{\text{host}} = n_{\text{host}}^2 = 2.25$), because this value is typical for many solvents, glasses, and polymers. Then the resonance frequency is estimated to be:

$$\Omega \simeq 0.93\Omega_L \simeq 902\ \text{cm}^{-1} \tag{2.11}$$

and corresponding resonance wavelength $\Lambda \simeq 11$ μm. Using Eqs. (2.3), (2.5), (2.6), and (2.10) we easily find:

$$S(\Omega) = -i\frac{3\varepsilon_{\text{host}}(\varepsilon_0 - \varepsilon_\infty)}{(\varepsilon_\infty + 2\varepsilon_{\text{host}})(\varepsilon_0 + 2\varepsilon_{\text{host}})}\frac{\Omega}{\gamma} \tag{2.12}$$

that is estimated to be:

$$\left|S(\Omega)\right| \simeq 0.1\frac{\Omega}{\gamma} \simeq 19 \tag{2.13}$$

and hence, the gain factor $G(\Omega) = |S(\Omega)|^2 \simeq 3.6 \times 10^2$.

Note also, that nonradiative decay of the resonance transition should be depressed, because the transition frequency is less than the frequency of longitudinal optical phonon, $\Omega_{\text{TR}} < \Omega_L$.

## 3. Core–shell nanocrystals

For optical transitions localized in the core of core–shell nanocrystals, the screening factor is given by Eq. (1.6). If in this expression, the real part of the complex dielectric function of the shell

$$\varepsilon_{\text{shell}}(\omega) = \operatorname{Re}\varepsilon_{\text{shell}}(\omega) + i\operatorname{Im}\varepsilon_{\text{shell}}(\omega) \tag{3.1}$$

is negative, $\operatorname{Re}\varepsilon_{\text{shell}}(\omega) < 0$, the real part of the denominator in the expression (1.6) vanishes at two resonance frequencies.

To avoid simple but more tedious computations, we restrict our further analysis to the case of a relatively thick shell of the thickness $\Delta \gtrsim R$. Is such core–shell nanocrystals, the geometry factor $\sigma = [R/(R+\Delta)]^3$ is much less than 1. Therefore, computing zeroes of the real part of the denominator in Eq. (1.6), we can omit the term proportional to $\sigma$. Then, the resonance frequencies are determined by the equations:

$$\operatorname{Re}\varepsilon_{\text{shell}}(\Omega_1) \simeq -2\varepsilon_{\text{host}}, \tag{3.2}$$

$$\operatorname{Re}\varepsilon_{\text{shell}}(\Omega_2) \simeq -\frac{1}{2}\varepsilon_{\text{core}}, \tag{3.3}$$

where we also omitted a contribution of small term $[\operatorname{Im}\varepsilon_{\text{shell}}]^2$. Note that at $\varepsilon_{\text{core}} = 4\varepsilon_{\text{host}}$ we deal with the degenerate case when $\Omega_1 = \Omega_2$.

At the resonance frequencies, the magnitudes of the function $W(\omega)$ are found to be:

$$\begin{aligned} W(\Omega_1;\sigma) &\\ \simeq{}& \frac{i}{\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_1)} \\ &\times \frac{18\varepsilon_{\text{host}}^2}{(\varepsilon_{\text{core}} - 4\varepsilon_{\text{host}}) + 2(\varepsilon_{\text{core}} + 5\varepsilon_{\text{host}})\sigma + 2i\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_1)}, \end{aligned} \tag{3.4}$$

$$\begin{aligned} W(\Omega_2;\sigma) &\\ \simeq{}& \frac{i}{2\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_2)} \\ &\times \frac{9\varepsilon_{\text{core}}\varepsilon_{\text{host}}}{-(\varepsilon_{\text{core}} - 4\varepsilon_{\text{host}}) + 2(2\varepsilon_{\text{core}} + \varepsilon_{\text{host}})\sigma + 2i\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_2)}, \end{aligned} \tag{3.5}$$

where we keep the terms proportional to $\sigma$ and $[\operatorname{Im}\varepsilon_{\text{shell}}]^2$, because they become essential, if $\varepsilon_{\text{core}}$ is sufficiently close to $4\varepsilon_{\text{host}}$.

In core–shell nanocrystals we still have one free parameter—the geometry factor $\sigma$. An appropriate choice of the geometry factor enables to increase one of the magnitudes: $W(\Omega_1;\sigma)$ or $W(\Omega_2;\sigma)$. If $\varepsilon_{\text{core}} < 4\varepsilon_{\text{host}}$, then at $\sigma = \sigma_1$, where

$$\sigma_1 = \frac{4\varepsilon_{\text{host}} - \varepsilon_{\text{core}}}{2(\varepsilon_{\text{core}} + 5\varepsilon_{\text{host}})}, \tag{3.6}$$

we find:

$$W(\Omega_1;\sigma_1) = \left[\frac{3}{2}\frac{\operatorname{Re}\varepsilon_{\text{shell}}(\Omega_1)}{\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_1)}\right]^2. \tag{3.7}$$

That results in the gain factor:

$$G(\Omega_1;\sigma_1) = \left[W(\Omega_1)\right]^2 = \left[\frac{3}{2}\frac{\operatorname{Re}\varepsilon_{\text{shell}}(\Omega_1)}{\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_1)}\right]^4. \tag{3.8}$$

The expression in the square brackets in the above formula is nothing but the screening factor at the Fröhlich resonance frequency, and hence, the gain factor in core–shell nanocrystals is equal to the square of the Fröhlich gain factor, $G_{\text{core–shell}} = [G_{\text{Fröhlich}}]^2$.

For nanocrystals with the CdSe core of permittivity $\varepsilon_{\text{core}} = 6.2$ in a host matrix of permittivity $\varepsilon_{\text{host}} = 2.25$, the required geometry factor $\sigma_1 = 0.08 \ll 1$, and we find $\Delta \simeq 1.3R$.

If $\varepsilon_{\text{core}} > 4\varepsilon_{\text{host}}$, then at $\sigma = \sigma_2$, where

$$\sigma_2 = \frac{\varepsilon_{\text{core}} - 4\varepsilon_{\text{host}}}{2(2\varepsilon_{\text{core}} + \varepsilon_{\text{host}})}, \tag{3.9}$$

we derive analogous expressions for the magnitude $W(\Omega_2;\sigma_2)$:

$$W(\Omega_2;\sigma_2) = -\frac{3}{2}\frac{\operatorname{Re}\varepsilon_{\text{shell}}(\Omega_2)}{\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_2)}\frac{3\varepsilon_{\text{host}}}{\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_2)} \tag{3.10}$$

and the gain factor:

$$G(\Omega_2;\sigma_2) = \left[\frac{3}{2}\frac{\operatorname{Re}\varepsilon_{\text{shell}}(\Omega_2)}{\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_2)}\right]^2\left[\frac{3\varepsilon_{\text{host}}}{\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_2)}\right]^2. \tag{3.11}$$

For nanocrystals with PbSe core of permittivity $\varepsilon_{\text{core}} = 22.9$ in a host matrix of permittivity $\varepsilon_{\text{host}} = 2.25$, the required geometry factor $\sigma_2 = 0.14$, and we find $\Delta \simeq R$.

Thus, core–shell nanocrystals exhibit a huge enhancement of an optical transition localized in the core provided that the geometry factor is chosen to maximize the gain factor either at the frequency $\Omega_1$ or at the frequency $\Omega_2$. An appropriate choice of materials, including host material, and sizes of core–shell nanocrystals enables to develop extremely efficient optical media operating in an extremely wide spectral range—from ultraviolet to terahertz.

### *3.1. Semiconductor shell*

To show a high efficiency of core–shell nanostructures we estimate parameters the gain factor in core–shell nanocrystals with the shell made of SiC, focusing on the resonance frequency $\Omega_1$ that is controlled by host permittivity only. As in the case of bare SiC nanocrystals, for core–nanocrystals in a host of permittivity $\varepsilon_{\text{host}} = 2.25$, the resonance frequency $\Omega_1 \simeq 902\ \text{cm}^{-1}$ ($\Lambda_1 = 11$ μm). The core of the structure can be made of any semiconductor material[1] enabling to provide an intraband optical transition with the transition frequency $\Omega_{\mathrm{TR}} = \Omega_1$. However, the permittivities of core and host control the required geometry factor $\sigma_1$. For CdSe core of radius $R$ the shell thickness $\Delta = 1.3R$.

In the case of bare nanocrystals the Fröhlich gain factor is given by Eq. (2.9), and was estimated to be $G_{\text{Fröhlich}} \simeq 3.6 \times 10^2$. Therefore, for core–shell nanocrystals with the SiC shell, we immediately find

$$G_{\text{core–shell}}(\Omega_1;\sigma_1) = [G_{\text{Fröhlich}}]^2 = 1.3 \times 10^5. \tag{3.12}$$

Thus, semiconductor core–shell nanocrystals exhibit a huge enhancement of optical transitions lying in the spectral range from 10 microns to submillimeters. To discuss an electrodynamical enhancement of optical transition in the ultraviolet, visible and near-IR ranges, we need obviously to consider nanostructures with metal shells. The first example of such a nanostructure with the cobalt (Co) core and CdSe shell have been recently fabricated [8].

### 3.2. Metal shell

Let us consider first the case of shell made of metal, e.g. an alkali metal, whose dielectric function is well described by the Drüde formula:

$$\varepsilon_{\text{shell}}(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i/\tau)}, \tag{3.13}$$

where $\omega_p$ and $\tau$ are respectively the plasma frequency and relaxation time. The real and imaginary parts of the dielectric function are then given by:

$$\operatorname{Re}\varepsilon_{\text{shell}}(\omega) = 1 - \frac{\omega_p^2\tau^2}{\omega^2\tau^2 + 1} \simeq 1 - \frac{\omega_p^2}{\omega^2}, \tag{3.14}$$

$$\operatorname{Im}\varepsilon_{\text{shell}}(\omega) = \frac{\omega_p^2\tau}{\omega(\omega^2\tau^2 + 1)} \simeq \frac{1}{\omega_p\tau}\left(\frac{\omega_p}{\omega}\right)^3. \tag{3.15}$$

Here, in the second approximate equities, we took into account that in alkali metals $\omega_p\tau \gg 1$ ranging from 79 for lithium (Li) to 285 for sodium (Na) [9], and the frequency range of our interest is not very far from the plasma frequency, i.e. $\omega\tau$ is also much greater than 1.

Then, Eqs. (3.2) and (3.3) result in the following expressions for the resonance frequencies:

$$\Omega_1 = \frac{\omega_p}{(1 + 2\varepsilon_{\text{host}})^{1/2}}, \tag{3.16}$$

$$\Omega_2 = \frac{\omega_p}{(1 + \varepsilon_{\text{core}}/2)^{1/2}}. \tag{3.17}$$

Since the plasma frequency in alkali metals ranges from 2.8 eV for cesium (Cs) to 6.2 eV for lithium (Li), the resonance frequency $\Omega_1$ for nanostructures with alkali shell in a host of permittivity $\varepsilon_{\text{host}} = 2.25$, $\Omega_1 = \omega_p/\sqrt{5}$, ranges in the interval 1.2–2.6 eV, covering thus the wavelength range from approximately 470 nm to 1 micron. Since the resonance frequency $\Omega_1$ does not depend on the core permittivity core, one can employ any semiconductor, in which the frequency of one of the optical transitions can be tuned by the core size to $\Omega_1$. While estimates of the resonance frequency $\Omega_2$ require to specify semiconductor material of the core.

Nevertheless, as well in the case of semiconductor shell, the choice of the geometry factor depends on the core permittivity. For the CdSe core of radius $R$, the required thickness of the shell was already estimated to be $\Delta = 1.3R$. Then, the magnitude of the function $W(\omega)$ at $\omega = \Omega_1$ is found to be:

$$W(\Omega_1;\sigma_1) = \left[\frac{3\varepsilon_{\text{host}}}{(1 + 2\varepsilon_{\text{host}})^{3/2}}(\omega_p\tau)\right]^2. \tag{3.18}$$

At $\varepsilon_{\text{host}} = 2.25$, we then get $W(\Omega_1;\sigma_1) \simeq [0.5\omega_p\tau]^2$. At a typical for alkali metals value $\omega_p\tau \sim 100$, we find $W(\Omega_1;\sigma_1) \sim 2.5 \times 10^3$, that results in the giant gain factor $G(\Omega_1;\sigma_1) = [W(\Omega_1;\sigma_1)]^2 \sim 6 \times 10^6$.

In many metals, the dielectric function is not described by the Drüde formula in the visible and near-IR spectral ranges. Therefore, we need to use experimental data to engineer core–shell nanostructures with enhanced optical transitions. Let us consider, for example, a nanostructure with the silver (Ag) shell. The frequency $\Omega_1$ is found from the condition (3.2). As it follows from the experimental data for silver [10], the real part of the dielectric function of the silver shell, $\operatorname{Re}\varepsilon_{\text{shell}}(\omega)$, reaches the magnitude $-2\varepsilon_{\text{host}} = -4.5$ at the frequency $\omega = \Omega_1 \simeq 3$ eV. While the imaginary part of the silver dielectric function is found to be $\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_1) \simeq 0.2$ [10]. At an appropriate choice of the core and shell sizes, we get for the gain factor:

$$G(\Omega_1;\sigma_1) = \left[\frac{3}{2}\frac{\operatorname{Re}\varepsilon_{\text{shell}}(\Omega_1)}{\operatorname{Im}\varepsilon_{\text{shell}}(\Omega_1)}\right]^4 \simeq \left[\frac{3}{2}\frac{4.5}{0.2}\right]^4 \simeq 1.2 \times 10^6. \tag{3.19}$$

## 4. Nanostructures with two shells

To prevent a direct electrical core–shell contact, it could be necessary to fabricate an intermediate shell between the core and metal (or even semiconductor) shell. Moreover, a sufficiently thick intermediate shell enables to essentially depress a radiationless energy transfer from the core to a metal shell with excitation of surface plasmons that undergo Landau damping [11].

An intermediate shell can be made of either semiconductor or even dielectric material like silica ($SiO_2$) [12]. If the thickness of the intermediate shell is much less than the core radius, it does not contribute essentially to electrodynamical properties of the structure, which are still described by the screening factor $W$ (1.6), and all of the above-derived results are thus valid.

A sufficiently thick intermediate shell essentially modifies Eq. (1.6). Now, the expression for the screening factor [3] contains, as it must be expected, three ratios of permittivities: $\varepsilon_{\text{core}}/\varepsilon_{\text{int.shell}}$, $\varepsilon_{\text{int.shell}}/\varepsilon_{\text{shell}}$, and $\varepsilon_{\text{shell}}/\varepsilon_{\text{host}}$. If the real part of the dielectric function of the shell is negative, the resonance fre-

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[1] We assume here that both core and shell semiconductor materials are chosen to provide also the localization of the considered optical transition in the core of the nanostructure.

quencies are found from the conditions:

$$\operatorname{Re}\varepsilon_{\text{shell}}(\Omega_1) \simeq -2\varepsilon_{\text{host}}, \tag{4.1}$$

$$\operatorname{Re}\varepsilon_{\text{shell}}(\Omega_2) \simeq -\frac{1}{2}\varepsilon_{\text{int.shell}}. \tag{4.2}$$

They are completely analogous to the conditions (3.2) and (3.3) for core–shell nanocrystals, but both of the resonance frequencies are independent of the core permittivity, because the resonance frequency $\Omega_2$ is determined by the permittivity of the intermediate shell. That provides us an additional flexibility in engineering nanostructured optical media with desired properties.

Although we discussed here only optical transitions localized in the core of core–shell nanostructures, the developed approach enables to consider optical transitions localized in one of the shells of the nanostructure or even extended over core and shell, as it occurs in ZnSe/CdSe nanostructures [13,14].

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## References

[1] L.D. Landau, E.M. Lifshitz, Electrodynamics of Continuous Media, Elsevier, Oxford, 1984.
[2] B.L. Wehrenberg, C. Wang, P. Guyot-Sionnest, J. Phys. Chem. B 106 (2002) 10634.
[3] V.I. Rupasov, Phys. Lett. A 363 (2007) 140.
[4] H. Fröhlich, Theory of Dielectrics, Oxford Univ. Press, London, 1949.
[5] C.F. Bohren, D.R. Huffman, Absorption and Scattering of Light by Small Particles, Wiley, New York, 1983.
[6] O. Madelung, Semiconductors—Basic Data, Springer, Berlin, 1996.
[7] W.G. Spitzer, D.A. Kleinman, Phys. Rev. 121 (1961) 1324.
[8] H. Kim, M. Achermann, L.P. Balet, J.A. Hollingsworth, V.I. Klimov, J. Am. Chem. Soc. 127 (2005) 544.
[9] N. Ashcroft, D. Mermin, Solid State Physics, Brooks Cole, NY, 1976.
[10] P.B. Johnson, R.W. Christy, Phys. Rev. B 6 (1972) 4370.
[11] I.A. Larkin, M.I. Stockman, M. Achermann, V.I. Klimov, Phys. Rev. B 69 (2004) 121403(R).
[12] V. Klimov, M. Petruska, private communication.
[13] S.A. Ivanov, et al., J. Phys. Chem. B 108 (2004) 10625.
[14] L.P. Balet, et al., Nano Lett. 4 (2004) 1485.