Threshold Limitations of the SPASER

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We present a semi-classical analytic model for spherical core-shell surface plasmon lasers (spasers). Within this model, we drop the widely used quasi-static approximation of the electromagnetic field in favor of fully electromagnetic Mie theory. This allows for precise incorporation of realistic gain relaxation rates that so far have been massively underestimated. Based on this, we obtain a quantitative understanding of the threshold characteristics that limit efficient spaser devices. Specifically, our model suggests how the threshold of spasers can be significantly reduced by introducing an emitter-free spacing layer between the gain medium and the metal core. Our model can be extended to complex plasmonic nanostructures using the recently introduced concept of quasi-normal modes.

Nanoscopic sources of coherent electromagnetic fields are essential elements for different fields in nanooptics, such as nanoplasmonics [1], metamaterials [2], and quantum plasmonics [3]. A surface plasmon laser (spaser) might be such a nanoscopic source [4, 5].

As compared to a laser, the obvious difference of a spaser is the use of plasmons instead of photons. These plasmons are inherently localized excitations and generally exhibit much smaller (mode) volumes than photonic cavity modes [6]. From an electromagnetic perspective, there is no reason to expect any further principle deviations from well-known (semi-classical) laser physics. For instance, Mie theory [7] completely describes the electromagnetic field for a (spherical) particle irrespective of the constituent material, i.e., whispering gallery modes of dielectric spheres and particle plasmons of metallic spheres are all included - where Gustav Mie did not make a difference, we will not do so either. However, as we will detail below, there are certain issues that have to be treated with care.

Recently, a number of spaser devices [8–14] have been characterized and extensive theoretical work has addressed fundamental and device-specific spaser properties [15–20]. However, several questions, even of a fundamental nature, remain to be answered. Perhaps the most important of these is related to the rather high spaser threshold. For instance, previous experiments placed very high demands on properties of the pump (e.g., high laser pulse intensities) [10], the synthesis of the spaser’s gain medium (e.g., dense incorporation of fluorophores) [8], and, quite generally, very high demands regarding the material quality [14]. Accordingly, these issues are reflected by the rather small number of publications that address spaser action in fully nanoscopic systems and/or systems working with organic gain media [8, 10].

Analytic theoretical descriptions have focussed so far on quasi-static [4, 11, 21-24] analyses that only consider a nanoparticle’s dipolar resonance. Rather numerical cold cavity analyses of actual devices have been used in order to show (i) that observed far-field pattern correspond to measured spectra etc. [11, 14], (ii) that the resonator under observation is unable to support ordinary purely optical modes [11, 14], and (iii) to estimate the minimum gain required to overcome losses within the resonator.

A complete description of spasing/lasing systems requires a fully quantum-mechanical treatment. In view of the dispersive and dissipative properties of metals and the open-system character of any spaser, this represents a formidable task.

In this work, we set ourselves the more modest, but also challenging goal of developing a fully electromagnetic semi-classical rate-equation approach. This allows for a quantitative investigation of the input-output characteristics, notably the quantitative determination of the spasing/lasing threshold.

The resonator in our model is based on the analytically derived eigenfrequency and quality factor of a dipolar mode of a metal sphere [21]. The interaction between resonator and gain medium is described by the radiative and non-radiative decay rates into all the different multipolar “modes” existing in the metal sphere (Fig. 1). All these quantities are obtained analytically from the full Maxwell equations via Mie theory. We are starting from the well-known system of a single emitter placed in front of a spherical particle [22] that is refined towards a system with an arbitrary number of homogeneously distributed and randomly oriented emitters. Finally, this system is merged into an effective single mode rate-equation model.

The radiative and non-radiative decay rates of emitters in the vicinity of spherical metal particles have been obtained some time ago using a multi-polar expansion of the electromagnetic field [22]. In particular, it has been found that for dipole emitters in close proximity (sub-wavelength distance) to the metal sphere, coupling of the emitter to higher-order multi-poles contributes significantly to the non-radiative decay rate. Staying within the language of Mie theory, these decay channels are off-resonant higher-order multi-polar “modes”. Within the language of solid-state physics, these non-radiative channels are excitations of various quasi-particles in the metal, e.g., electron-hole pairs, sometimes referred to as surface energy transfer (SET) (due to its similarity to Förster
The gain-medium model considers dipolar emitters ($\vec{d}$) in proximity to a lossy plasmonic resonator, specifically the decay rates into different channels like off-resonant higher order modes ($\gamma_{\text{offres}}$), farfield ($\gamma_{\text{rad}}$) and into the resonator-mode ($\gamma_M$). The emitters are randomly but homogeneously incorporated in a shell around the sphere of thickness $d_{\text{shell}}$ (between the radii $R$ and $R_2$) except for the emitter-free spacing layer of thickness $d_{\text{free}}$ ($R$ to $R_1$).

This is also related to the fact that for dispersive and dissipative material properties, the multi-polar "modes" are not anymore orthogonal. Physically speaking, these multi-polar "modes" exchange power between each other and, considering that higher-order multi-poles do not radiate but are prone to Ohmic losses, this leads to additional non-radiative decay processes (this is already captured by the framework of Ref. [22]). In fact, such cross-coupling effects are well-known in laser physics (excess noise or Petermann factor) [23] and, e.g., lead to modifications of the laser threshold and laser linewidth. Obviously, such effects cannot be captured when only considering dipolar modes in quasi-static [4, 10, 20] approaches.

Therefore, the representation of the gain medium by an ensemble of dipolar emitters, where each emitter is treated individually (since the decay rates are extremely sensitive on the precise position and orientation [22, 24]) is a key feature of our model, which clearly goes beyond the usual linear gain medium description. The decomposition of the rates into multi-polar orders (Mie coefficients) allows us further to analyze in detail how the emitted energy goes: to Ohmic losses/dissipation ($\gamma_{\text{inrad}}$), to far-field radiation ($\gamma_{\text{rad}}$), or to the actual resonator-mode ($\gamma_M$). Including an incoherent pump rate allows us to formulate corresponding rate equations for each emitter. Depending on the concrete realization of the spaser, these equations are subsequently averaged over the orientations or the emitters’ dipole moments and/or their positions (distances from the sphere, angular positions). The details of this rate-equation analysis are outlined in the supplemental material (SM).

Specifically, in the remainder we consider a spaser that operates on the (three energetically degenerate) dipolar resonances of a metal sphere (sphere radius $R$, eigenfrequency $\omega_{\text{ex}}$, quality factor $Q_{\text{ex}}$), i.e., a spaser, where the gain medium consists of randomly oriented emitters that have been uniformly distributed in a shell. The shell of thickness $d_{\text{shell}}$ (from $R$ to $R_2$, see Fig. 1) around the sphere may include an emitter-free part we call "spacing layer" of thickness $d_{\text{free}}$ (from $R$ to $R_1$) between metal and gain medium. We refer to this system as a lasing spaser, since the dipolar mode is "bright", i.e., this mode will effectively couple to the far-field and thus emit photons. As described in the SM, this system may be reduced through an averaging procedure to an effective single-mode rate-equation model that, in the stationary case, can readily be solved. After averaging, the decay rates can be rewritten in a simple (and intuitive) weighted sum of decay rates that stem from emitters that are oriented parallel ($\gamma_\parallel$) and perpendicular ($\gamma_\perp$) to the sphere’s surface: $\gamma(r) = \frac{1}{2} \gamma_\parallel(r) + \frac{1}{2} \gamma_\perp(r)$. Integration of $\gamma(r)$ from $R_1$ to $R_2$ yields overall decay rates that are used in the rate equations. The emission into a single dipole resonator-mode $\gamma_M$ can be calculated when we only consider the first order Mie coefficients and divide by the degeneracy of this mode, whereas we have to sum up 400 Mie coefficients (for short distances) to approach convergence when calculating the total decay rates. We want to point out here that the rate of stimulated emission into a single mode is nothing else than the rate of the corresponding spontaneous emission multiplied with the number of photons/plasmons present in that specific mode (this is already implemented in the rate-equations). Thus, by calculating $\gamma_M$ we can describe the whole spaser system. Throughout the paper gold is modelled through a Drude-Lorentz permittivity that was fitted to the data measured by Johnson & Christy [26] (fitting parameters see SM).

As can be inferred from Fig. 2(a), close to the sphere, the total decay rate is increased by orders of magnitude relative to the vacuum decay rate, an effect that is mainly due to enhanced coupling to non-radiative channels. This effect is well known (since it limits performance) in the field of plasmonic nanoantennas where often one is interested in boosting the radiative decay of single photon emitters. So far, this effect has largely been ignored in spaser theories, possibly based on the assumption that the resonant dipolar plasmon "mode" would dominate over the off-resonant higher-order multi-polar "modes". However, while this assumption is often justified in ordinary laser systems, it has no base when emitters are close to a metal sphere. Fig. 2(b) shows that even though the resonant dipole mode might be the dominant decay channel, it is by far not dominating the overall decay: $\gamma_M$ is orders of magnitudes smaller than $\gamma_{\text{tot}}$ and thus close-by-emitters can only provide a small ratio of their emitted energy to the actual resonator-mode.

Of course, also the decay rates of Ref. [22] have a limited range of applicability. For instance, once the emitters approach the metal to within the sub-5 Å-regime additional processes such as charge transfer may occur, which are not included in the theory. Note, however, such processes would typically lead to even further increased non-radiative decay rates. Consequently, while our model would fail to be fully quantitative for such
FIG. 2. Orientation-averaged decay rates of a dipolar emitter as a function of distance to a gold sphere (R=7 nm and outer refractive index n_{out}=1.46). The emission frequency of the emitter is resonant with the sphere’s dipole modes (λ_{res}=534 nm). All rates are normalized to the corresponding vacuum decay rate. (a) The solid orange curve shows the total decay rate γ_{tot}, the dashed black the non-radiative decay rate γ_{nrad} and the red dotted curve the decay rate into a single (resonant) spaser mode γ_{M}. (b) Ratio of the decay rate into the spaser mode to the total decay rate γ_{M}/γ_{tot}.

small distances, it would still provide a conservative estimate for distances down to 1 Å.

Our model of a metallic sphere in a homogeneous outer medium of refractive index is rather close to realistic scenarios such as those in which dye-doped coatings of metal particles have been used and the spasers are immersed in ethanol [8], since this represents a situation of reasonable refractive index matching (n_{shell}=n_{out}). In any case, also the implementation of layered spherical structures, e.g., shells of different materials is not causing any conceptual problems in Mie theory [3].

At this point we are ready to compute the actual stationary input-output curves of the lasing spaser model, i.e., the number of plasmons in the resonator-mode as a function of pump. Specifically, we utilize realistic values as reported by Noginov et al. [3]: Fig. 3 depicts two stationary input-output curves of spasers with different emitter configurations (different d_{free} values) but otherwise identical parameters, i.e., R=7 nm, d_{shell}=15 nm (R_s=22 nm), n_{out}=1.46 and an emitter density of 0.06225 nm^{-3} (for the resonator we find ω_{res}=2(3.07 - 0.115)i eV and correspondingly Q_{res} = Re[ω_{res}] / (-2Im[ω_{res}]) = 10.06). One spaser is equipped with an emitter-free spacing layer of d_{free}=5 nm between emitters and sphere whereas the other spaser has almost no spacing layer (d_{free}=0.1 nm due to the above mentioned limited validity of the decay rates). For the calculations we use the measured lifetime of τ=4.3 ns in ethanol [3] with an refractive index of n_{eth}=1.33 to extract the vacuum decay rate of γ_{0}=1/(n_{eth}τ)=1.75 × 10^{8} s^{-1}.

The spaser with an emitter-free spacing layer between the emitters and the sphere’s surface exhibits an almost three orders of magnitude lower threshold (so does a spaser with same emitter number/slightly higher emitter density) even though its total number of emitters is less (N=2337) than for the spaser without spacing layer. This is surprising at first glance as one may think that additional gain medium - especially in the most intensive zone of the plasmon field - would be always beneficial for laser action. However, the situation is more complex: Emitters close to the metal not only decay extremely fast due to direct electron-hole excitation (Ohmic losses), these emitters are also the most efficient re-absorbers of plasmons due to the high field intensities close to the sphere. Accordingly, this leads to more electron-hole pairs instead of more plasmons. In other words: Emitters in close proximity to the sphere’s surface are efficient preventers of spaser action as they rapidly funnel emerging plasmon fields into Ohmic loss channels. This issue can however be simply neutralized by introducing a protective emitter-free spacing layer between metal and gain medium.

Further, this conclusion is in very good agreement with the published spaser devices that do not utilize localized but propagating plasmons and semiconductors as gain medium [11–14] and exhibit rather low thresholds, i.e.,
at room-temperature or in continuous wave mode, respectively. In these publications it is argued that low-index spacing layers are used to define a certain “hybrid” propagating mode that is guided mainly inside this spacing layer between high-index gain-materials and metal to minimize propagation losses [25]. As we now indirectly demonstrated, these designs are also very effectively avoiding the problematic issue with enhanced loss channels.

Realistic values for the excitation rate $\gamma_p$ of the gain medium can be easily calculated and correlated with actual laser flux densities $\Phi$ in experiments, e.g., via tabulated molecule absorption cross sections $\sigma_{abs}$. Doing so for the values given in Ref. 8 ($\Phi=10^{25}$ photons s$^{-1}$ cm$^{-2}$, $\sigma_{abs}=2.55 \times 10^{-16}$ cm$^{-2}$ and $N=2700$ emitters) we derive an overall excitation rate ($\gamma_p=\Phi\sigma_{abs}N$) of $6.9 \times 10^{-12}$ s$^{-1}$ (see orange vertical line in Fig. 3). Unfortunately the spacing layer in Ref. 8 is not defined with great accuracy but only called a “thin sodium silicate shell” which is used for stabilization during the chemical synthesis of gold nanospheres. Also, presumably there are variations from spaser to spaser in the measured particle ensembles, e.g., the emitters might be somehow inhomogeneously distributed over the shell, so that we expect to find the threshold for the spasers in Ref. 8 somewhere inbetween the two input-output curves in Fig. 3.

Nevertheless, our model explains qualitatively why lower thresholds have been found in Ref. 10 as compared to Ref. 8. First, the use of nanorods is lifting the three-fold degeneracy of the dipolar mode in a sphere, leading to slightly lower pump requirements (see the factor X in Eq. (21) in the SM). Much more important, however, is the rather thick ($\approx 10$ nm) porous silica layer used and the coverage with emitters. This procedure will most likely lead to a decaying emitter density towards the metal nanorod, effectively realizing a spacing layer.

The limitations of our approach are very similar to those that are known for rate-equations such as modeling of mode beating dynamics and spectral narrowing. Consequently, actual threshold values will presumably be even slightly above those that our model predicts. However, our model clearly demonstrates the paramount importance to accurately consider all decay channels, also those that originate from coupling to off-resonant higher-order multi-poles and the cross-coupling of non-orthogonal ”modes” (akin to the Peterman factor 27 of ordinary lasers) which naturally arises in the context of spasers. A strategy of simply bringing emitters extremely close to the metal surface in order to maximally exploit the plasmonic field enhancement 10 clearly underestimates the detrimental effects of the loss channel enhancement afforded by the non-resonant higher-order multi-polar ”modes”.

Furthermore, it has been suggested that plasmonic resonators with the smallest mode volumes/highest Purcell factors would perform the best 2 3 10. This approach may be less beneficial than expected as small mode volumes would require the gain medium to be positioned very close to the metal. Rather, we suggest that spaser design considerations should be based on the ratio of energy that an emitter channels into the resonator-mode to the overall emitted energy (see Fig. 2(b)).

At this point, it is worth noting that besides the non-orthogonality of ”modes” a quantum-optical treatment of the spaser would also require that these ”modes” are normalized to an appropriate volume. This latter issue has recently been addressed 29 31. For instance, by applying the numerical approach of Ref. 29 for ”modes” of arbitrary particles to the analytically solvable case of a sphere, we have confirmed that the dipolar contributions to the rates as described in Ref. 22 are exactly reproduced. This means that our analysis presented above can be extended to arbitrary particles and ”modes”.

In conclusion, our work provides a clear physical picture of threshold issues in spaser devices along with corresponding design guidelines. The unavoidable cross-coupling of ”modes” in plasmonic systems and the direct coupling of the gain medium to off-resonant decay channels heavily influence spaser thresholds. Consequently, their impact has to be minimized by carefully designed emitter-free spacing layers that balance the beneficial effect of gain enhancement through plasmonic fields with the detrimental effects of off-resonant decay channels. Our model can be suitably enhanced by the above-discussed numerical approaches 29 31 for determining ”modes” for complex plasmonic nanoparticles. So, it provides a rather easy tool to use for computing minimum pump power requirements for a given design and will thus be extremely helpful for future experimental works. In addition, our semi-classical model may provide the basis for full quantum-optical treatment of spaser action.

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The purpose of this supplemental material is to provide a detailed derivation of the semi-classical SPASER equation as used in the main manuscript. The strategy of the derivation is as follows: Starting from the results of radiative as well as non-radiative decay rates of an electric dipole emitter interacting with all the multi-polar resonances (Mie resonances) of a metal sphere [1], we formulate the associated rate equations (including incoherent pumping of the emitter). Specifically, we develop the theory by focussing on a SPASER whose gain medium operates on the dipolar plasmon-polariton resonances of the metal sphere.

At this point, it is important to note that a particle plasmon-polariton is a composite object with an electromagnetic “mode profile” both inside and outside the metal sphere.

Further, we would like to emphasize that there are $2n + 1$ degenerate modes per multipole of order $n$. In particular, the dipolar resonances associated with the “principal quantum number” $n = 1$ consist of three dipolar multi-poles that are distinguished by their “magnetic quantum number” $m$. Consequently, for a dipole emitter of fixed (but arbitrary) orientation relative to a given coordinate system, we generally obtain a multi-mode-like description with energetically degenerate multi-polar resonances. Here, we hasten to stress, that we do not quantize the electromagnetic field. Rather our referring to “quantum numbers” is only meant as a loose analogy to help build intuition with regards to the multi-polar structure of the full (classical) electromagnetic treatment (Mie theory) of a metal sphere as opposed to a treatment within a quasi-static approximation which features a single dipolar resonance.

The above observations suggest the following implementation of our strategy:

(i) The decay rate of an emitter into a plasmon “mode” comprises both, a non-radiative and a radiative contribution. The concrete calculation of those contributions is given in section 1.

(ii) The threefold degenerate dipolar plasmon “modes” are regarded as SPASER “modes”. The decay into these “modes” may be described akin to the active medium’s emission rates into the lasing modes of an ordinary multi-mode laser.

(iii) The decay rates into higher-order multi-polar modes are interpreted as loss rates of the emitters. They thus play the same role as the so called non-radiative decay rates in the description of lasers.
(iv) The imaginary part of the SPASER “mode” frequencies (i.e., the eigen-frequencies of the metal sphere’s scattering matrix that correspond to the dipolar resonances) may be identified with the loss rates of the dipolar SPASER “modes”.

In particular, the above discussion regarding multi-mode-like behavior becomes important when, in a second step, we consider many emitters uniformly distributed on a shell around the metal sphere. This requires an averaging of the above-described single-emitter rate equations over all solid angles and, depending on the concrete physical realization, potentially an averaging over the dipoles’ orientations. Together with the standard semi-classical mean-field factorization between atomic and electromagnetic degrees of freedom [3], this will lead to an effective single-mode-like description of the dipolar Lasing-SPASER that involves corresponding effective (i.e., averaged) rates.

In a final step, we solve these effective rate equations in the stationary case for the plasmon-polariton occupation in the dipolar resonances of the metal sphere. As these plasmon-polaritons will radiate into free space in a known manner, we have thus solved the problem of a semi-classical Lasing-SPASER for a single metal sphere within a fully electromagnetic analysis (i.e., without a quasi-static approximation).

1 Review of Decay Rates near a Sphere

The electromagnetic problem of a single electric-dipole emitter (position \( \mathbf{r}' \), dipole moment \( \mathbf{p} \)) interacting with a conducting sphere (radius \( R \), complex dielectric constant \( \epsilon_{\text{sph}}(\omega) = \epsilon(\omega) + i\sigma(\omega)/\omega \), conductivity \( \sigma(\omega) \)) proceeds by considering the total field as the sums of the electromagnetic field of an emitter in a homogeneous medium, the induced field inside the sphere and the scattered fields outside the sphere. These fields may be expanded into the set of vector spherical harmonics centered around the sphere’s center and the unknown expansion coefficients for the scattered and induced fields are found by matching the boundary conditions of the total field on the sphere’s surface.

1.1 Relative Non-radiative Decay Rate

From these fields, the non-radiative energy loss rate \( W_A \) is obtained by integrating the Ohmic losses over the sphere volume \( \Omega \). Normalizing to the dipole’s emitted power in free space \( W_{R0} \) we obtain the emitters relative non-radiative decay rate

\[
\frac{\gamma_{\text{nrad}}}{\gamma_0} = \frac{W_A}{W_{R0}} = \frac{3 \pi^2 \varepsilon_0^2 \sigma}{2 \mu_0^4 \rho^2} \left( \frac{\rho_0}{\rho} \right) \sum_{\alpha \kappa} C_{\alpha \kappa} \left\{ |f_{\alpha \kappa}|^2 J_n^{(M1)}(k_\kappa, 0, R) + |g_{\alpha \kappa}|^2 J_n^{(N1)}(k_\kappa, 0, R) \right\}
\]

(2)

For the dielectric function we use a Drude-Lorentz model

\[
\epsilon_{\text{sph}}(\omega) = \varepsilon_r - \frac{\omega_p^2}{\omega^2 + i\gamma_1 \omega} - \frac{\omega^2}{\omega^2 - (\omega^2 - \Omega^2) + i\gamma_2 \omega}
\]

(1)

where \( \varepsilon_r = 5.967 \), \( \omega_p = 8.729 \), \( \gamma_1 = 0.065 \), \( \delta = 1.09 \), \( \Omega = 2.684 \) and \( \gamma_2 = 0.433 \).
where, following Ruppin’s notation all over the paper \[\text{I} \], the following abbreviations have been introduced:

\[
C_{mn} = \frac{(2n + 1)(n - m)!}{n(n + 1)(n + m)!}, \quad \epsilon_m = \begin{cases} 1 & \text{if } m = 0 \\ 2 & \text{if } m = 1 \end{cases}
\]

\[
I_n^{(M1)}(k, r_1, r_2) = \int_{r_1}^{r_2} j_n(kr) r^2 dr
\]

\[
I_n^{(N1)}(k, r_1, r_2) = \frac{1}{2n + 1} \int_{r_1}^{r_2} [(n + 1)|j_{n-1}(kr)|^2 + n|j_n(kr)|^2] r^2 dr
\]

\[
f_{amn}(k_o, r', \mathbf{p}) = \alpha_n p_{amn} = \frac{ik^3}{\varepsilon_o \pi} \alpha_n \left(M^3_{amn}(k_o r') \cdot \mathbf{p}\right)
\]

\[
g_{amn}(k_o, r', \mathbf{p}) = \beta_n q_{amn} = \frac{ik^3}{\varepsilon_o \pi} \beta_n \left(N^3_{amn}(k_o r') \cdot \mathbf{p}\right)
\]

Further, the summation in Eq. (2) runs over \(a \in \{e, o\}\) (even and odd modes with respect to \(m\)), \(n = 1, 2, \ldots, \) and \(m = 0, \ldots, n\), where the combination \((o0n)\) is excluded. In fact, the combination of \(a \in \{e, o\}\) and \(m = 0, \ldots, n\) together with the exclusion of \((o0n)\) is equivalent to a “magnetic quantum number” \(m = -n, \ldots, n\).

### 1.2 Relative Radiative Decay Rate

The radiative energy loss rate \(W_A \text{[I]} \) can be obtained by integrating the flux of the Poynting vector over a spherical surface that contains both, the dipole and the sphere. Normalizing to \(W_{00} \) we obtain the emitters relative radiative decay rate

\[
\frac{\gamma_{rad}}{\gamma_0} = \frac{W_R}{W_{00}} = \frac{3 \pi^2 \varepsilon_0^2}{2 k_0^2 \iota_0^2} \sum_{amn} C_{amn} \left\{ |s_{amn} + u_{amn}|^2 + |t_{amn} + v_{amn}|^2 \right\},
\]

where the following abbreviations have been introduced:

\[
s_{amn}(k_o, r', \mathbf{p}) = \frac{ik^3}{\varepsilon_o \pi} \left(M^1_{amn}(k_o r') \cdot \mathbf{p}\right)
\]

\[
u_{amn}(k_o, r', \mathbf{p}) = \frac{ik^3}{\varepsilon_o \pi} \left(N^1_{amn}(k_o r') \cdot \mathbf{p}\right)
\]

\[
t_{amn}(k_o, r', \mathbf{p}) = \frac{ik^3}{\varepsilon_o \pi} \left(M^3_{amn}(k_o r') \cdot \mathbf{p}\right)
\]

\[
v_{amn}(k_o, r', \mathbf{p}) = \frac{ik^3}{\varepsilon_o \pi} \left(N^3_{amn}(k_o r') \cdot \mathbf{p}\right)
\]

### 1.3 Decay Rates into Plasmon “Modes”

The absolute values \(\gamma_{nrad}\) and \(\gamma_{rad}\) of the non-radiative and radiative decay rates are obtained from (2) and (3) through multiplication by the experimentally determined free space decay \(\gamma_0\) of the considered emitter. Then, the decay rate of the emitter into a multipolar plasmon “mode” labeled \((amn)\) is associated with the respective multipolar contribution \(\gamma_{amn}\) to the total decay rate \(\gamma = \gamma_{rad} + \gamma_{nrad} = \sum_{amn} (\gamma_{amn,rad} + \gamma_{amn,nrad}) = \sum_{amn} \gamma_{amn}\).
1.4 Summary
The contributions to the radiative and non-radiative decay rates from each multi-polar resonance \( \{amn\} \) depends on the emitter’s position \( r' \) and the orientation of its dipole moment \( p \). Roughly speaking, for dipole-sphere separations above one wavelength of the emitted radiation, the sphere’s dipolar resonances dominate and – in contrast to the case of a dipole in front of a plane – there is practically no distance dependence. In contrast, for separations below one wavelength higher-order multi-polar resonance become increasingly important and eventually dominate for small separations \([1]\).

2 Lasing-SPASER Rate Equations

We are now in a position to formulate the equations for the dipolar SPASER “modes”. Specifically, we consider a number of identical two-level uncoupled emitters at different positions \( r' \) and with different dipole orientations \( p \) that interact with a single metal sphere. Consequently, we introduce a corresponding composite emitter label \( \mu = (r', p) \).

The emission rate of emitter \( \mu \) into the dipolar SPASER “mode” \( \lambda = (e01, e11, o11) \) (resonance frequencies \( \omega_\lambda \)) is denoted by \( \gamma_\lambda \mu \). In addition to the dipolar “modes” the emitters total loss rates, \( \gamma_t \mu \), are also composed of the higher-order multi-polar contributions to the emitters decay. These SPASER “modes” exhibit losses, i.e. they radiate into the far-field and are subject to dissipation within the sphere. The corresponding total loss rates of the SPASER “modes” are encoded in the \( Q_\lambda \)-values of the associated dipolar resonances and are determined by the imaginary parts of the poles of the sphere’s scattering matrix in the complex frequency plane.

The above program may now be executed along the lines of standard multi-mode laser theory \([2]\) and we obtain

\[
\frac{dn_\lambda}{dt} = \left( n_\lambda + \frac{1}{2} \right) \sum_\mu D_\mu \gamma_\lambda \mu + \frac{1}{2} \sum_\mu N_\mu \gamma_\lambda \mu - \frac{\omega_\lambda}{Q_\lambda} n_\lambda, (\lambda = e01, e11, o11) \quad (13)
\]

\[
\frac{dD_\mu}{dt} = N_\mu (\gamma_{\mu 0} - \gamma_{\mu 1}) - D_\mu \left( \gamma_{\mu 0} + \gamma_{\mu 1} + 2 \sum_\lambda \gamma_{\lambda \mu} n_\lambda \right). \quad (14)
\]

Here, \( N_\mu = N_{1,\mu} + N_{2,\mu} \) and \( D_\mu = N_{1,\mu} - N_{2,\mu} \) denote, respectively, the total number and the inversion of the two-level emitters with label \( \mu \) (\( N_{1,\mu} \) and \( N_{2,\mu} \) are the number of emitters in ground and excited states, respectively). In addition, we have introduced a pump rate \( \gamma_{\mu 0} \) for emitter \( \mu \). Finally, \( n_\lambda \) denotes the number of plasmons in SPASER “mode” \( \lambda \).

Eqs. (13) and (14) represent a system of \( N + 3 \) equations, where \( N \) is the total number of emitters. For a (spatial and/or orientational) distribution of the emitters, we have to average the Eqs. (13) and (14) accordingly. This is facilitated by summing Eq. (14) over the emitters,

\[
\sum_\mu \frac{dD_\mu}{dt} = \sum_\mu N_\mu (\gamma_{\mu 0} - \gamma_{\mu 1}) - \sum_\mu D_\mu \left( \gamma_{\mu 0} + \gamma_{\mu 1} + 2 \sum_\lambda \gamma_{\lambda \mu} n_\lambda \right). \quad (15)
\]

In order to make further progress, it is helpful to introduce the emitter density \( \rho_N(\mu) \) and the inversion density \( \rho_D(\mu) \) per “orientational angle” and to convert
the \( \mu \)-sums in Eq. (15) to appropriate integrals.

\[
\int d\mu \frac{dD_\mu}{dt} = \frac{1}{V_\mu} \int d\mu \rho_N(\mu) (\gamma_p - \gamma_t) - \int d\mu \rho_D(\mu) \left( \gamma_p + \gamma_t + 2 \sum_\lambda \gamma_{\lambda \mu} n_\lambda \right).
\]

(16)

In order to understand the underlying physics, we consider the simplest nontrivial case where the emitters are homogeneously distributed within a thin shell (inner radius \( R_1 \), outer radius \( R_2 \)) and, further, exhibit a uniform distribution of dipole orientations. Consequently, we set

\[
\rho_N(\mu) = \frac{N}{4\pi V_{sh}} \quad \text{and} \quad \rho_D(\mu) = \frac{D}{4\pi V_{sh}},
\]

(17)

where \( V_{sh} \) is the volume of the shell and \( N \) and \( D \) are the total number and total inversion within the thin shell. It is important to note (see section 1) that even for a homogeneous emitter density, the inversion density in general exhibits a strong dependence on the radial position and the dipole orientation of the emitters (by symmetry, the inversion density does not exhibit any angular dependence for a homogeneous emitter density). Therefore the second equation in (17) represents a mean-field-like approximation [3].

With this, we proceed to rewrite Eq. (15) and Eq. (13) to

\[
\frac{dD}{dt} = N (\overline{\gamma}_p - \overline{\gamma}_t) - D (\overline{\gamma}_p + \overline{\gamma}_t + 2 X_1 \gamma_M n_1),
\]

(18)

\[
\frac{dn_1}{dt} = \left( n_1 + \frac{1}{2} \right) D \overline{\gamma}_M + \frac{1}{2} N \overline{\gamma}_M - \frac{\omega_1}{Q_1} n_1.
\]

(19)

Here, \( \overline{\gamma}_\lambda \) are the averaged decay rates

\[
\overline{\gamma}_\lambda = \frac{1}{4\pi V_{sh}} \int d\mu \gamma_{\lambda \mu}
\]

\[
= \frac{1}{4\pi V_{sh}} \int _{R_1} ^{R_2} dr' r'^2 \int _0 ^{2\pi} d\phi' \int _0 ^{\pi} d\theta' \sin \theta' \int _0 ^{2\pi} d\phi \int _0 ^{\pi} d\theta \sin \theta \gamma_{\lambda}(r', p).
\]

(20)

In Eqs. (18), (19), and (20), the quantities \( \omega_\lambda, Q_\lambda, \) and \( \gamma_\lambda \) depend only on the “principal quantum number” \( n \) and are degenerate with the multiplicity of the corresponding “magnetic quantum number” \( m \) (the even and odd vector spherical harmonics with index \( m = 0, \ldots, n \) and (00n) excluded, see section 1). Since we are focussing on the dipolar SPASER “modes” with \( n = 1 \), we obtain the effective semiclassical dipolar SPASER equations

\[
\frac{dD}{dt} = N (\overline{\gamma}_p - \overline{\gamma}_t) - D (\overline{\gamma}_p + \overline{\gamma}_t + 2 X_1 \gamma_M n_1)
\]

(21)

\[
\frac{dn_1}{dt} = \left( n_1 + \frac{1}{2} \right) D \gamma_M + \frac{1}{2} N \gamma_M - \frac{\omega_1}{Q_1} n_1.
\]

(22)

Where we defined \( \gamma_M = \overline{\gamma}_1 \) and \( X_1 = 3 \) denotes the (geometrical) degeneracy of the dipolar resonances with respect to the “magnetic quantum number” \( \overline{\gamma}_p \)
and $\gamma_t$ have been defined analogously to $\gamma_\lambda$ in Eq. (20).

For the stationary solution to the effective semiclassical dipolar SPASER equations, we finally obtain

$$n_1 = \frac{\gamma M N Q (\gamma M X_1 + \gamma_t + \gamma_1) \pm \sqrt{8 \gamma^2 M N Q X_1 + \gamma M N Q (\gamma M X_1 + \gamma_t + \gamma_1) - \omega_1 (\gamma p + \gamma_t)^2}}{4 \gamma M N Q X_1}$$

(23)

The dipolar SPASER equations, Eqs. (21) and (22), together with the stationary solution, Eq. (23), represent the central object of study of our main manuscript. However, at this point we would like to emphasize that our derivation is very general and can be extended, for instance, to study mode competition between the dipolar and quadrupolar SPASER “modes”. The corresponding effective rate equations are

$$\frac{dD}{dt} = N (\gamma p - \gamma_t) - D (\gamma p + \gamma_t + 2 (X_1 \gamma M_1 n_1 + X_2 \gamma M_2 n_2)),$$

$$\frac{dn_1}{dt} = \left( n_1 + \frac{1}{2} \right) D \gamma M_1 + \frac{1}{2} N \gamma M_1 - \frac{\omega_1}{2 Q_1} n_1,$$

$$\frac{dn_2}{dt} = \left( n_2 + \frac{1}{2} \right) D \gamma M_2 + \frac{1}{2} N \gamma M_2 - \frac{\omega_2}{2 Q_2} n_2.$$

(24)

Here $X_1$ and $X_2$ denote, respectively, the multiplicities of the corresponding dipolar and quadrupolar resonances. Similarly, the resonance frequencies, $\omega_1$ and $\omega_2$, the quality factors $Q_1$ and $Q_2$, and the plasmon-polariton occupation numbers, $n_1$ and $n_2$ refer to dipolar and quadrupolar SPASER “modes”.

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