

## Complete characterization of the spasing (L-L) curve of a three-level quantum coherence enhanced spaser for design optimization

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We demonstrate that it is possible to derive an approximate analytical expression to characterize the spasing (L-L) curve of a coherently enhanced spaser with 3-level gain-medium chromophores. The utility of this solution stems from the fact that it enables optimization of the large parameter space associated with spaser designing, a functionality not offered by the methods currently available in the literature. This is vital for the advancement of spaser technology towards the level of device realization. Owing to the compact nature of the analytical expressions, our solution also facilitates the grouping and identification of key processes responsible for the spasing action, whilst providing significant physical insights. Furthermore, we show that our expression generates results within 0.1% error compared to numerically obtained results for pumping rates higher than the spasing threshold, thereby drastically reducing the computational cost associated with spaser designing. Published by AIP Publishing. <https://doi.org/10.1063/1.5025354>

The ability to confine and manipulate light on the nano-scale enables the development of ultra-fast, efficient and miniaturized optical circuits and systems, leading to many applications in nanotechnology.<sup>1–5</sup> Owing to the advancements in sophisticated nano-fabrication techniques, device sizes approaching tens of nanometers have been built and tested reliably.<sup>6</sup> However, miniaturization of optical devices below the sub-wavelength scale has been constrained by the fundamental limit of diffraction.<sup>7</sup> Therefore, confinement of light to unprecedentedly smaller volumes is essential for enabling very strong light-matter interactions in order to realize ultra-fast nano-scale optical devices. These challenges have been met successfully in the emerging field of quantum nano-plasmonics with the use of Surface Plasmons (SPs).<sup>2,8–12</sup> Generally, SPs are excited at metal-dielectric interfaces and as a result, they naturally experience intrinsic losses.<sup>13</sup> Therefore, energy must be transferred from an external source in order to replenish and sustain SP resonances.<sup>8</sup>

Coherent amplification of localized SPs via stimulated emission, theorized by Stockman and Bergman in 2003,<sup>14</sup> led to the discovery of spaser (Surface Plasmon Amplification by Stimulated Emission of Radiation) which is the nano-scale counterpart of laser. The effect of spasing was first demonstrated experimentally by Noginov *et al.* in 2009.<sup>15</sup> Since then, developing efficient, reliable and easily controllable spasers has become one of the major research areas in nano-plasmonics.<sup>16–19</sup>

A typical spaser consists of a plasmonic resonator (similar to the laser cavity) that supports SP modes and a gain medium that supplies energy to amplify them. Usually, the gain medium chromophores are modelled as two level

systems effectively, which have only one mode of external input in the form of an incoherent pump.<sup>20</sup> The intensity of the output optical field of a spaser can be represented by the number of localized SPs generated per spasing mode. These output characteristics of spasers are known as spasing curves<sup>20</sup> (a counterpart of the L-L curve for lasers<sup>7</sup>).

Due to the gain saturation caused by the plasmonic feedback on the gain medium chromophores, spasers generate fewer number of SPs while having higher thresholds. Dorfman *et al.*<sup>21</sup> have significantly improved the output efficiency of spasers by introducing another coherent optical field as an input to control the spaser output characteristics. They utilize a concept very much analogous to lasing without inversion (LWI) to enhance spaser performance. The chromophores in this scenario have been modelled as 3-level systems.

Owing to the number of coupled equations in this formulation, the equations naturally lend themselves to numerical solutions. However, an analytical solution closely approximating the exact numerical solution may not only provide valuable insights but may also contribute towards design optimization. Therefore, in this letter, we derive an analytical expression for the spasing curve of a spaser with 3-level gain medium chromophores. We have established the accuracy of the derived expression by comparing our results with detailed numerical simulations.

The basic configuration and the energy flow diagram of our system are illustrated in Fig. 1. A plasmonic nano-structure is surrounded by an optical gain medium which comprises homogeneously distributed generic 3-level quantum emitters. An emitter has a ground state ( $|1\rangle$ ), as well as two excited states ( $|2\rangle$  and  $|3\rangle$ ). The two inputs to the spaser are denoted by the incoherent pumping rate  $g$  (coupled to the transition  $|1\rangle \rightarrow |3\rangle$ ) and the Rabi Frequency of the coherent

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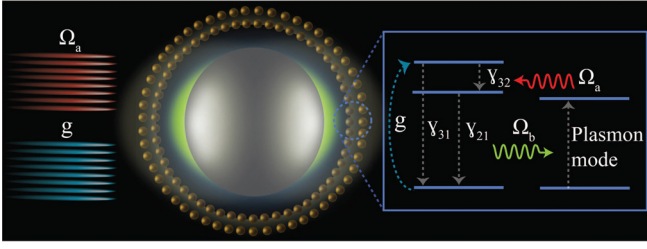


FIG. 1. Basic operation of a SPASER: Gain medium chromophores are excited using an incoherent pump ( $g$ ) as well as a coherent drive  $\Omega_a$ . The transition  $|2\rangle \rightarrow |1\rangle$  is coupled to one of the SP modes in the plasmonic nano-structure, which excites SPs via stimulated emission.

input field  $\Omega_a$  (coupled to the transition  $|2\rangle \rightarrow |3\rangle$ ). Since we are interested in the continuous wave (CW) operation of the spaser,  $g$  and  $\Omega_a$  will be provided continuously as long as the spaser works, in order to sustain the SPs. The spasing transition  $|2\rangle \rightarrow |1\rangle$  represented by the corresponding Rabi frequency  $\Omega_b$  is coupled with a surface plasmon mode in the nano-plasmonic resonator to transfer energy non-radiatively from the chromophores to the SPs, thereby sustaining the spasing process. The plasmonic field generated in the nano-structure provides an internal feedback to the gain medium during operation. The spontaneous decay rates in the 3-level emitters are denoted by  $\gamma_{21}$ ,  $\gamma_{32}$  and  $\gamma_{31}$ , as indicated in Fig. 1. The total number of 3-level quantum emitters (chromophores) in the gain medium is denoted by  $N_c$ .

To simplify our analysis, we approximate the 3-level emitters as dipoles.<sup>16</sup> Rotating Wave Approximation (RWA) has been deployed to neglect the rapidly oscillating terms in the Hamiltonian.<sup>7</sup> Then, the total Hamiltonian of the gain medium in the interaction picture can be written as

$$\mathcal{H}_{int} = \sum_p \{ -\hbar \Delta_b^{(p)} |1\rangle \langle 1| + \hbar \Delta_a^{(p)} |3\rangle \langle 3| - (\hbar \Omega_b^{(p)} |2\rangle \langle 1| + \hbar \Omega_a^{(p)} |3\rangle \langle 2| + c.c.) \}, \quad (1)$$

where  $c.c.$  denotes the complex conjugate and  $\hbar$  is the reduced Planck's constant. The Hamiltonian takes into account the summation of all the  $N_c$  chromophores and is denoted by  $\sum_p$ . We assume that the inter-chromophore interactions are comparatively weak enough to be neglected when writing the Hamiltonian.<sup>7,16,20</sup> In Eq. (1), detunings are defined as  $\Delta_a = \omega_{32} - \omega_a$  and  $\Delta_b = \omega_{21} - \omega_b$ ,<sup>22</sup> where  $\omega_{32}$  and  $\omega_{21}$  are the frequencies of the corresponding band gaps of the chromophores and  $\omega_a$  and  $\omega_b$  are defined as the frequencies of the coherent field and the plasmonic field, respectively.

Standard semiclassical theory has been adopted in the analysis, where the gain medium is treated quantum-mechanically and the SPs as well as photons are treated as classical quantities.<sup>20,21</sup> Therefore, we express both the plasmon annihilation operator,  $\hat{a}_n$ , and the photon annihilation operator,  $\hat{b}_m$ , as time varying complex numbers (C-numbers) with definitions  $a_n = a_{0n} e^{-i\omega_s t}$  and  $b_m = b_{0m} e^{-i\omega_{32} t}$ .<sup>16</sup>  $a_{0n}$  and  $b_{0m}$  are slowly varying amplitudes, whereas  $\omega_s$  is the frequency related to spasing. Based on the above information, the number of SPs per  $n^{\text{th}}$  spasing mode can be expressed as  $N_n = |a_{0n}|^2$ .<sup>7</sup>  $\Omega_b^{(p)}$  is expressed as  $-A_n d_{21}^{(p)} \nabla \psi_n a_{0n} / \hbar$ , where  $A_n = (\{4\pi \hbar \text{Re}[s(\omega_n)]\} / \{\epsilon_d \text{Re}[ds(\omega_n)/d\omega_n]\})^{1/2}$ . Here,  $s(\omega_n) = \epsilon_d / [\epsilon_d - \epsilon_m(\omega_n)]$ , where  $\epsilon_d$  is the bath permittivity

and  $\epsilon_m(\omega_n)$  is the permittivity of the metal at plasmon resonance frequency  $\omega_n$ . The dipole moment element of the gain chromophores is given by  $d_{21}$  and the gradient of the potential function  $\nabla \psi_n \approx 1/\sqrt{V}$ , where  $V$  is the modal volume of the electric field generated by the plasmon mode, which is directly related to the size and the shape of the nano-particle.<sup>21,23</sup> Furthermore,  $\Omega_a$  is considered a constant since we assume that the driving field is strong enough to maintain the number of photons in mode  $m$  constant under operating conditions.<sup>21</sup>

The  $p^{\text{th}}$  gain medium chromophore is modelled as an open quantum system using the Liouville-von Neumann master equation,  $\dot{\rho}^{(p)} = -i/\hbar [\mathcal{H}_{int}, \rho^{(p)}] - \mathcal{L} \rho^{(p)}$ , where  $\dot{\rho}^{(p)}$  represents the time derivative of the density matrix.  $\mathcal{L}$  is the Lindblad super-operator that quantifies the interactions with the environment, such as spontaneous decay of the gain medium, incoherent pumping and dephasing (decoherence).<sup>24</sup> Lindblad terms have been extracted from Ref. 21, 25, and 26. The off-diagonal relaxation rates are  $\Gamma_{21} = 0.5(\gamma_{21} + g) + \gamma_{ph} + i\Delta_b$ ,  $\Gamma_{31} = 0.5(\gamma_{31} + \gamma_{32} + g) + \gamma_{ph} + i(\Delta_a + \Delta_b)$  and  $\Gamma_{32} = 0.5(\gamma_{31} + \gamma_{32} + \gamma_{21}) + \gamma_{ph} + i\Delta_a$ , where  $\gamma_{ph}$  is the dephasing (decoherence) rate of  $\rho_{ij}$ .<sup>21</sup> Hence, the emitter density matrix elements  $\rho_{ij}$  are given by the following complex valued coupled partial differential equations:

$$\dot{\rho}_{11} = \gamma_{21} \rho_{22} + \gamma_{31} \rho_{33} - g \rho_{11} + i(\Omega_b^* \rho_{21} - \Omega_b \rho_{21}^*), \quad (2a)$$

$$\dot{\rho}_{33} = -(\gamma_{31} + \gamma_{32}) \rho_{33} + g \rho_{11} - i(\Omega_a^* \rho_{32} - \Omega_a \rho_{32}^*), \quad (2b)$$

$$\dot{\rho}_{21} = -\Gamma_{21} \rho_{21} - i\Omega_b(\rho_{22} - \rho_{11}) + i\Omega_a^* \rho_{31}, \quad (2c)$$

$$\dot{\rho}_{31} = -\Gamma_{31} \rho_{31} - i\Omega_b \rho_{32} + i\Omega_a \rho_{21}, \quad (2d)$$

$$\dot{\rho}_{32} = -\Gamma_{32} \rho_{32} - i\Omega_a(\rho_{33} - \rho_{22}) - i\Omega_b^* \rho_{31}, \quad (2e)$$

$$\rho_{11} = 1 - \rho_{22} - \rho_{33}. \quad (2f)$$

We describe the stimulated emission of SPs as their excitation by the coherent polarization of the gain medium corresponding to the transition  $|2\rangle \rightarrow |1\rangle$ . Therefore, the time evolution of the plasmon annihilation operator should be of the same form as that of a 2-level gain medium,<sup>16</sup> which is expressed by invoking the Heisenberg equation of motion for  $a_{0n}$  as

$$\dot{a}_{0n} = -\Gamma_n a_{0n} + i \sum_p \rho_{21}^{(p)} \tilde{\Omega}_b^{(p)}. \quad (3)$$

The SP relaxation rate  $\Gamma_n$  is expressed as  $\gamma_n + i\Delta_n$ , where  $\gamma_n$  is the plasmon decay rate and  $\Delta_n$  is the detuning between  $\omega_n$  and the frequency of the  $|2\rangle \rightarrow |1\rangle$  transition ( $\omega_{21}$ ). The value  $\tilde{\Omega}_b^{(p)}$  is the single plasmon Rabi frequency denoted by  $\tilde{\Omega}_b^{(p)} = \Omega_b^{(p)} / a_{0n}$ . Next, we assume that all  $N_c$  chromophores interact with the SPs identically, and hence we omit the index  $p$  and set  $\sum_p \rightarrow N_c$  in Eq. (3).

Next, by writing the complex variables and coefficients in Eqs. (2a)–(2f) and Eq. (3) in the form of  $z = \text{Re}(z) + i\text{Im}(z)$  and by equating the real and imaginary parts of the equations separately, we derive a set of real valued, non-linear, coupled differential Eqs. (4a)–(4k). Note the labeling we have followed:  $\text{Re}(\rho_{cd}) = \rho_{cd}^R$ ,  $\text{Im}(\rho_{cd}) = \rho_{cd}^I$ ,  $\text{Re}(a_{0n}) = \mathcal{A}$  and  $\text{Im}(a_{0n}) = \mathcal{B}$ . Furthermore,  $\Delta_a$  and  $\Delta_b$  are taken as zero since

resonant coupling is assumed between relevant interactions. Hence, all  $\Gamma_{cd}$ s are assumed real.<sup>16,21</sup>

$$\dot{\rho}_{11} = \gamma_{21}\rho_{22} + \gamma_{31}\rho_{33} - g\rho_{11} + 2\tilde{\Omega}_b(B\rho_{21}^R - A\rho_{21}^I), \quad (4a)$$

$$\dot{\rho}_{33} = -(\gamma_{31} + \gamma_{32})\rho_{33} + g\rho_{11} + 2\Omega_a\rho_{32}^I, \quad (4b)$$

$$\dot{\rho}_{21}^R = -\Gamma_{21}\rho_{21}^R + \tilde{\Omega}_bB\rho_{22} - \tilde{\Omega}_bB\rho_{11} - \Omega_a\rho_{31}^I, \quad (4c)$$

$$\dot{\rho}_{21}^I = -\Gamma_{21}\rho_{21}^I - \tilde{\Omega}_bA\rho_{22} + \tilde{\Omega}_bA\rho_{11} - \Omega_a\rho_{31}^R, \quad (4d)$$

$$\dot{\rho}_{31}^R = -\Gamma_{31}\rho_{31}^R + \tilde{\Omega}_bA\rho_{32}^I + \tilde{\Omega}_bB\rho_{32}^R - \Omega_a\rho_{21}^I, \quad (4e)$$

$$\dot{\rho}_{31}^I = -\Gamma_{31}\rho_{31}^I - \tilde{\Omega}_bA\rho_{32}^R + \tilde{\Omega}_bB\rho_{32}^I + \Omega_a\rho_{21}^R, \quad (4f)$$

$$\dot{\rho}_{32}^R = -\Gamma_{32}\rho_{32}^R + \tilde{\Omega}_bA\rho_{31}^I - \tilde{\Omega}_bB\rho_{31}^R, \quad (4g)$$

$$\dot{\rho}_{32}^I = \Omega_a(\rho_{22} - \rho_{33}) - \Gamma_{32}\rho_{32}^I - \tilde{\Omega}_b(A\rho_{31}^R - B\rho_{31}^I), \quad (4h)$$

$$\rho_{11} = 1 - \rho_{22} - \rho_{33}, \quad (4i)$$

$$\dot{\mathcal{A}} = -\gamma_n\mathcal{A} + \Delta_n\mathcal{B} - N_c\tilde{\Omega}_b\rho_{21}^I, \quad (4j)$$

$$\dot{\mathcal{B}} = -\gamma_n\mathcal{B} - \Delta_n\mathcal{A} + N_c\tilde{\Omega}_b\rho_{21}^R. \quad (4k)$$

$N_n = |a_{0n}|^2$  and  $|a_{0n}|^2 = \mathcal{A}^2 + \mathcal{B}^2$ . Surface plasmons are modelled using C-numbers corresponding to bosonic number state representation, where  $N_n$  denotes the expected number of SPs generated in the spasing mode with frequency  $\omega_n$ .<sup>27</sup> The total energy output of the spaser is then given by  $\hbar\omega_n \times N_n$ . Hence,  $N_n$  gives a measure of the total output energy of the spaser. So, our aim is to simultaneously solve Eqs. (4a)–(4k) and derive an analytical expression for  $N_n$ . Such an expression would not only reduce the computational burden of calculating the output energy ( $\propto N_n$ ) of the spaser,<sup>7</sup> but would also provide numerous advantages for design optimization, whilst providing valuable physical insights into the operation of the device.

To study these Eqs. (4a)–(4k) in detail and observe the time evolution of  $\rho_{cd}$ ,  $\mathcal{A}$  and  $\mathcal{B}$ , we carry out some indicative numerical simulations. A silver nano-sphere of radius 40 nm is used as the plasmonic nano-structure. All parameter values are taken from Ref. 20, 21, 28, and 29, where  $\omega_n = 2.5$  eV,  $\gamma_n = 5.3 \times 10^{14}$  s<sup>-1</sup>,  $\Delta_n = 3.2 \times 10^{12}$  s<sup>-1</sup>,  $\gamma_{21} = 4 \times 10^{12}$  s<sup>-1</sup>,  $\gamma_{32} = 4 \times 10^{11}$  s<sup>-1</sup>,  $\gamma_{31} = 4 \times 10^{10}$  s<sup>-1</sup>,  $N_c = 6 \times 10^4$  and  $\epsilon_d = 2.25$ . The bulk permittivity values of silver are obtained from the Johnson and Christy model.<sup>28</sup> The time evolution of  $\mathcal{A}$  is given in Fig. 2 for an incoherent pumping rate ( $g$ ) of  $24 \times 10^{12}$  s<sup>-1</sup>. Due to the inherent slowly varying nature of the C-numbers ( $a_{0n}$  and  $\rho_{cd}$ s for  $c \neq d$ ),<sup>20</sup> multiple rounds of detailed numerical simulations show that all time derivatives

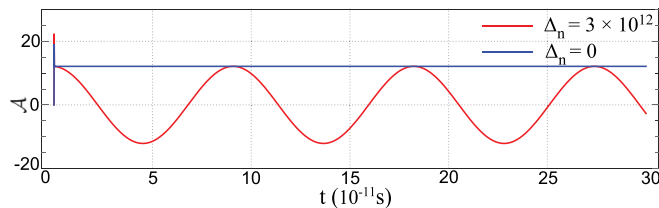


FIG. 2. Time evolution of  $\mathcal{A}$  when  $\Delta_n = 0$  and  $\Delta_n = 3 \times 10^{12}$  s<sup>-1</sup>. When the spasing transition ( $|2\rangle \rightarrow |1\rangle$ ) is resonantly coupled with the SP mode, the complex number representation of the plasmon annihilation operator ( $a_{0n}$ ) after the initial transients becomes constant over time. When there is significant detuning (i.e.,  $\Delta_n \neq 0$ ),  $a_{0n}$  varies sinusoidally over time.

in Eqs. (4a)–(4k) do not become zero simultaneously, even in the steady state for any  $g$  or  $\Omega_a$ . Since  $\Delta_n \ll \gamma_n, \omega_n$ , we now assume that the  $|2\rangle \rightarrow |1\rangle$  transition and the SP mode are resonantly coupled (i.e.,  $\Delta_n = 0$ ) and carry out the same set of numerical simulations. Note that such an assumption is physically justifiable as well, since the system designers always strive to achieve near or perfectly resonant coupling. The time evolution of  $\mathcal{A}$  when  $\Delta_n = 0$  is given in Fig. 2, where  $\mathcal{A}$  becomes constant in the steady state, unlike when  $\Delta_n \neq 0$ . This is the same for all other variables ( $\rho_{cd}$  and  $\mathcal{B}$ ) for all combinations of  $g$ ,  $\Omega_a$  and system parameters. Therefore, it is possible to set all time derivatives in Eqs. (4a)–(4k) to 0, which yields a set of algebraic equations that can be solved. By simultaneously solving the system of non-linear algebraic equations, we derive the full explicit analytical expression for  $N_n$  as follows:

$$N_n = [(-8\gamma_n\mathcal{F}_5((\Gamma_{21}\Gamma_{31}\gamma_n + \gamma_n\Omega_a^2)\mathcal{F}_{10} + N_c\tilde{\Omega}_b^2(\Gamma_{31}\Gamma_{32}\mathcal{F}_4 + \Omega_a^2\mathcal{F}_7)) + \mathcal{F}_8^2)^{\frac{1}{2}} - \mathcal{F}_8] \frac{1}{\mathcal{F}_9}, \quad (5)$$

where  $\mathcal{F}_1 = \Gamma_{32}(2\gamma_{31} + 2\gamma_{32} + g) + 6\Omega_a^2$ ,  $\mathcal{F}_2 = \gamma_{21}(\gamma_{31} + \gamma_{32} + g) + \gamma_{32}g$ ,  $\mathcal{F}_3 = \gamma_{21} - 3\gamma_{31} - \gamma_{32} - 3g$ ,  $\mathcal{F}_4 = \gamma_{21}\gamma_{31} + \gamma_{21}\gamma_{32} - \gamma_{32}g$ ,  $\mathcal{F}_5 = 2\gamma_{31} + 2\gamma_{32} + g$ ,  $\mathcal{F}_6 = \gamma_{21} + \gamma_{31} + 2g$ ,  $\mathcal{F}_7 = 2\gamma_{31}\Gamma_{31} - 2\Gamma_{31}g + 2\Gamma_{31}\gamma_{21} - \gamma_{21}g$ ,  $\mathcal{F}_8 = 2\gamma_n\Gamma_{31}\mathcal{F}_1 + \gamma_n\Gamma_{21}\mathcal{F}_2 + 2\gamma_n\Omega_a^2\mathcal{F}_3 + N_c\tilde{\Omega}_b^2\mathcal{F}_4$ ,  $\mathcal{F}_9 = 4\gamma_n\mathcal{F}_5\tilde{\Omega}_b^2$  and  $\mathcal{F}_{10} = \Gamma_{32}\mathcal{F}_2 + 2\Omega_a^2\mathcal{F}_6$ . Equation (5) can be used to analyze the number of SPs analytically, for  $g > g_{th}$ , where  $g_{th}$  is the spasing threshold. It gives a fully analytical characterization of the spasing curve since it contains both controlling parameters  $g$  and  $\Omega_a$  as well as all system specific parameters. We now compare the numerical simulations with the results generated by our analytical expression for the spaser proposed in Ref. 21. The error percentage ( $\mathcal{E}$ ) is defined as  $(N_n^s - N_n)/N_n^s \times 100\%$ , where  $N_n^s$  is the value obtained in numerical simulations. Figures 3(a) and 3(b) depict the spasing curve of  $N_n$  as  $g$  varies. Different curves have been obtained for various values of  $\Omega_a$ . It is evident that  $\mathcal{E}$  for the whole spasing regime ( $g > g_{th}$ ) remains less than 0.1% in all practical cases.<sup>16–18,20</sup> Studying the effect of dephasing (decoherence) on spasing is also of utmost importance for complete characterization.<sup>24</sup> Therefore, another comparison is made in Figs. 3(c) and 3(d) to investigate the effect of decoherence ( $\gamma_{ph}$ ) on spasing. While it becomes evident that the robustness of the spaser output against decoherence has been enhanced as  $\Omega_a$  is increased, it can also be seen that Eq. (5) can be used with an acceptable accuracy ( $\mathcal{E} < 0.1\%$ ) to study the effect of dissipative processes such as decoherence ( $\gamma_{ph}$ ) in a spaser.

We have ideally assumed a pure state initial condition for the density matrix elements when obtaining numerical results.<sup>21,22</sup> There is a slight disagreement between the numerical and analytical results for very small  $g$  values above the threshold for high  $\Omega_a$ . This could be due to the inability of the system to produce the expected response to high  $\Omega_a$  values at very low pumping rates, as the number of electrons in high energy levels is minute in the initial spasing build up. If a more practical mixed initial state is assumed instead,<sup>2,8</sup> even this slight disagreement will no longer be

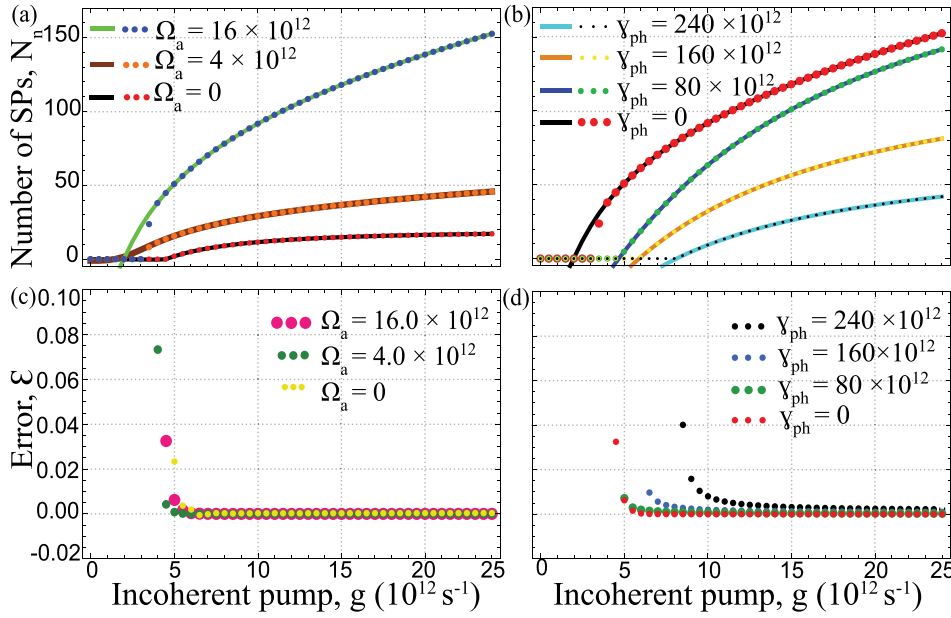


FIG. 3. (a)–(d) compare the spasing curves obtained both numerically and analytically. Dotted lines in (a) and (c) represent the numerical results and the solid lines represent our analytical results. (a) The spasing curves obtained both numerically and analytically for different values of  $\Omega_a$ , where  $\gamma_{ph} = 0$  for all cases. The corresponding error ( $\mathcal{E}$ ) values are shown in (c). (b) The spasing curves obtained both numerically and analytically for different values of  $\gamma_{ph}$ , where  $\Omega_a = 16 \times 10^{12} \text{ s}^{-1}$  for all cases. The corresponding error ( $\mathcal{E}$ ) values are shown in (d). Percentage error ( $\mathcal{E}$ ) is less than 0.1% for all cases in the spasing regime ( $g > g_{th}$ ).

present due to the initial presence of electrons in the upper energy levels to compensate for the low pumping rates. Furthermore, for practical pumping rates so far reported in the literature ( $g > 5 \times 10^{12} \text{ s}^{-1}$ ),<sup>16–18,20</sup> the values obtained using both types of initial conditions converge to our steady state analytical solution.

Figures 3 and 4 also suggest that apart from accurately characterizing the spaser output with  $\mathcal{E} < 0.1\%$  for  $g > g_{th}$  when  $\Delta_n \ll \gamma_n, \omega_n$ , it is also possible to use Eq. (5) to qualitatively understand the threshold conditions ( $g_{th}$ ) by setting  $N_n = 0$ . When  $\Delta_n$  is comparable or greater than  $\omega_n$  or  $\gamma_n$ , the predicted analytical values will not be within the accepted error bounds, i.e.,  $\mathcal{E} > 0.1\%$ . But such off-resonant conditions are not desired practically from a system designer's perspective since such conditions do not facilitate efficient energy transfer and there is some likelihood for system instability as well.<sup>7</sup>

Equation (5) can be tailored for different nanoparticles and gain media by varying  $\tilde{\Omega}_b$ , decay and dephasing rates and  $N_c$  accordingly. It is also possible to derive simpler expressions when the parameters are constrained to specific regimes. For example, if a certain spaser configuration satisfies the condition  $g > \gamma_{21} \gg \gamma_{32} \gg \gamma_{31}$ , its spasing curve can be derived in a more simplified form as follows:

$$N_n \approx \frac{\gamma_{32} N_c}{4\gamma_n} \left(1 - \frac{\gamma_{21}}{g}\right) - \frac{\gamma_{21}}{4\tilde{\Omega}_b^2} \left(g + \frac{2\Omega_a^2}{g}\right) + \frac{1}{4} \left[ \left(1 - \frac{\gamma_{21}}{g}\right)^2 \left(\frac{\gamma_{32} N_c}{\gamma_n}\right)^2 + \frac{8g N_c \Omega_a^2}{\gamma_n} \right]^{\frac{1}{2}}. \quad (6)$$

To simply model the main physical dependencies of the system, we considered the Taylor series expansion of Eq. (5) for the whole operating regime for fixed values of  $\Omega_a$ . We could approximate that  $N_n$  shows a logarithmic behavior as  $g$  varies, expressed as  $k_1 \ln(g - k_2 g_{th})$ , where  $k_1$  and  $k_2$  depend on system parameters and  $\Omega_a$ . Similar analyses can be done to simply approximate the relationships between other parameters.

Equation (5) can be used to analytically study the relationship between any selected pair of parameters by setting others as constants, similar to the way it is done for  $g$  vs.  $N_n$ . To demonstrate the versatility of our solution, we have studied how  $N_n$  behaves as the number of chromophores ( $N_c$ ) is varied in Fig. 5(a). Then, we set  $N_n = 0$  in Eq. (5) and express the threshold number of chromophores  $N_{c,th}$  as a function of  $\Omega_a$ , depicted in Fig. 5(b). Such analyses for any set of parameters can be performed using our analytical solution. This enables the system designers to develop highly intuitive and methodical optimization schemes for physically meaningful parameter sets, which are much simpler than full-blown numerical approaches. In addition, by setting

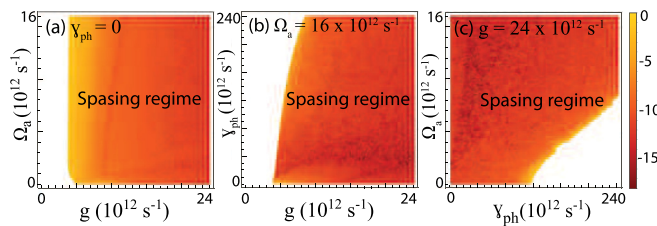


FIG. 4. (a)–(c) depict the two-dimensional plots for the logarithmic value of the absolute value of the error ( $\mathcal{E}$ ) percentage. (a)  $\log_{10}(|\mathcal{E}|)$  for a range of values of  $g$  and  $\Omega_a$  when  $\gamma_{ph} = 0$ . (b)  $\log_{10}(|\mathcal{E}|)$  for a range of values of  $g$  and  $\gamma_{ph}$  when  $\Omega_a = 16 \times 10^{12} \text{ s}^{-1}$ . (c)  $\log_{10}(|\mathcal{E}|)$  for a range of values of  $\Omega_a$  and  $\gamma_{ph}$  when  $g = 24 \times 10^{12} \text{ s}^{-1}$ . Error ( $\mathcal{E}$ ) is less than 0.1% for all cases in the spasing regime. Non-spasing regimes are shown in white.

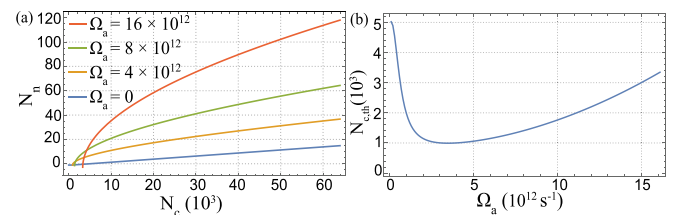


FIG. 5. (a) Depicts the variation of  $N_n$  as  $N_c$  varies for different values of  $\Omega_a$ . (b) The variation of minimum  $N_c$  (threshold) required to start spasing ( $N_{c,th}$ ) as  $\Omega_a$  varies in the configuration given in (a). Note that  $g = 15 \times 10^{12} \text{ s}^{-1}$  and  $\gamma_{ph} = 0$  for both (a) and (b).

$\Omega_a = 0$  and  $\gamma_{32} > g$  in Eq. (5) for a much faster  $|3\rangle \rightarrow |2\rangle$  population transfer,<sup>21</sup> it is possible to study the spasing output of a 2-level spaser<sup>20</sup> analytically.

In conclusion, we have derived an explicit analytical expression and simpler approximations to describe the spasing curves of a coherently enhanced spaser, whilst providing valuable physical insights into the operation of the spaser. The method we have followed in deriving the solution itself will aid similar nanoplasmonic systems to be solved analytically to very high accuracy. The derived expression provides the basis to calculate the optimal system parameters and inputs required in order to achieve desired spasing outputs, which is a capability that surpasses numerical solutions. Therefore, the proposed scheme enables the utilization of spasers in complex nanoplasmonic systems by enabling design optimization. In addition, it grants a faster way to determine the output energy generated by the localized SPs, eliminating the need for computationally expensive numerical simulations. The error of the spasing curve generated by our expression is under 0.1% for the whole operating regime above the spasing threshold, suggesting an almost perfect characterization.

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