Optical sensing works most efficiently around the singularity of resonances. In the pursuit of high quality-factor and sensitivity, non-plasmonic nanosensors are desired as metallic materials are intrinsically lossy. However, standalone resonant systems of dielectric nanoparticles (NPs) generally do not possess a pole or its identification has proven to be hard across the complex frequency domain. To solve this problem, an active external cavity is designed and the dielectric NP is put inside it to formulate a cavity-NP (C-NP) system. The dielectric NP has dimensions comparable with the effective wavelength in the particle material and the coupled resonance is shown to exhibit a pole when a singular optimum gain is applied, overcoming the no-pole-limitation of dielectric NPs. The underlying physics of the coupled system is studied with the pseudo-orthonormal eigenmode method (POEM), which can treat such non-Hermitian systems and quickly pinpoint the singularity from the real frequencies. The POEM study generates a set of guidelines that facilitate device design and experimental optimization. Through dynamic finite-difference time-domain simulations, the all-dielectric gain-assisted cavity-NP structure thus identified is shown to reach a pole at small optical gain. When used as a sensor, the system accommodates nanoscale sensing volume and giant sensitivity when operated around its pole.

1. Introduction

The optical nanosensor, a device that detects nanoscale changes in optical responses, generally observes the refractive index variation in the near field of a nanoparticle (NP) from far field. Both the resultant sensitivity and detection limit rely on the NP’s resonant response to the electromagnetic field. Sensitivity will be highest when operated around a resonant singularity, where quality factor and field localization are at their strongest.\(^1\)\(^{-}\)\(^3\) Identifying the resonant singularity is therefore key to pushing the sensitivity of a nanosensor. For instance, in the classical framework, surface plasmon amplification by stimulated emission of radiation (SPASER)\(^4\)\(^,\)\(^5\) can be characterized by the singularity of the dipolar resonance in a subwavelength metallic NP. Such resonance singularity is described by the mathematical pole (\(\epsilon_{\text{NP}} - \epsilon_{\text{h}}\))/\(\epsilon_{\text{NP}} + 2\epsilon_{\text{h}}\), which basically reveals the correlation between NP’s complex polarizability \(\alpha\) and the dielectric constants of the NP \(\epsilon_{\text{NP}}\) and host material \(\epsilon_{\text{h}}\).

Metallic NPs are most commonly preferred out of the NPs that utilize symmetric Lorentzian resonance for nanosensing. This is mostly because metal’s dielectric constant has a negative real part that results in a small denominator of the Clausius–Mossotti relation.

\[\alpha \propto \frac{(\epsilon_{\text{NP}} - \epsilon_{\text{h}})}{(\epsilon_{\text{NP}} + 2\epsilon_{\text{h}})}\]

This yields a divergent behavior of the polarizability, subsequently giving rise to strong plasmonic light-scattering and sub-diffraction mode volume.\(^7\) However, plasmonic NPs generally have low Q-factors due to high dissipative loss in the material. In order to increase the resonance Q-factor and sensitivity, it is shown in classical theory that a metallic NP is able to create a complete cancellation in the denominator when optical gain is introduced to the immediate surroundings of the NPs,\(^8\)\(^9\) leading to the creation of SPASER.\(^4\)\(^,\)\(^5\) Such device is predicted to be sufficient for single-molecule detection.\(^9\)\(^,\)\(^10\) Yet, the application of plasmonic lasers as nanosensors has several practical challenges. First, as the NPs are usually embedded in the gain medium for efficient amplification, the sensing targets are then insulated from the sensitive detection spot where the field is localized. Second, the optical pump power required by the necessary gain is generally applied directly onto the plasmonic structure and such high power could damage the subjects to be detected. Third, too strong a field localization is sometimes
counterproductive as it traps unwanted objects indiscriminately. We should note that nanosensing is about sensing nanoscale items rather than a demand for nanoscale mode volume at all costs. Although plasmonic NPs inherently support nanoscale mode volume, it is not the only way to realize nanosensing. Alternatively, it can be realized by optimizing the sensitivity and device configuration through other suitable structures as well.

With smaller losses and wider ranges of transparency windows, devices made of dielectric material can generally achieve much higher Q-factors. However, it is difficult to achieve pole singularity in Lorentzian resonance for a sensing particle made of dielectric materials due to intrinsic and practical limitations. According to the Clausius–Mossotti relation, the dipolar resonance of a dielectric NP anticipates no pole when the NP has subwavelength dimension, due to the fact that the dielectric constant of dielectric has a positive real part. If the particle size slightly increases to be comparable with the effective wavelength in the NP material, its optical response is no longer characterized solely by dipolar resonance but by multipolar modes as well. In this case, although a mathematical solution to pole might exist, unrealistic gain is usually required to compensate the low-Q nature that is omnipresent for most achievable modes in such NPs.

On the other hand, non-plasmonic NPs with asymmetric Fano resonances are known to support high Q-factors and potentially address the abovementioned issues.\(^{[11]}\) Although gain-assisted coupled resonant systems have been reported in many cases,\(^{[12–14]}\) the underlying physics in relation to lasing singularity as well as its accurate identification has yet to be investigated. Coupled oscillator model, as the most intuitive choice of theoretical framework for Fano resonance,\(^{[15]}\) does not consider eigenmodes and thus uncovers little of the physics. In general, the coupled oscillator model is unable to decouple the unknown frequency term analytically, resulting in an exhaustive sweep across the complex frequency plane in search for pole.\(^{[15]}\) The widely used temporal coupled-mode (TCM) theory for treating resonant optical system is generally restricted to Hermitian sub-systems without gain due to energy conservation, time-reversal symmetry, and reciprocity.\(^{[16]}\) Approximations have to be implemented in order for TCM to treat non-Hermitian systems.\(^{[17]}\)

In this work, we introduce a new theoretical framework that can treat non-Hermitian systems without any approximations. The pseudo-orthonormal eigenmode method (POEM) sets out from eigenmodes of a coupled resonance and reveals its physics through pole-identification across the complex frequency domain. Through the POEM study, we show that the no-pole-limitation for a dielectric NP of comparable size with the effective wavelength can be changed by creating gain-assisted Fano resonance in the cavity-nanoparticle (C-NP) system. Because POEM does not make any approximation and is able to study systems that are non-Hermitian, it retains more information about the system so that higher accuracy can be achieved for the solution of singularity as it is pushed from the complex plane onto the real frequency axis. Findings of the POEM study serve as design and optimization guidelines. When used together with finite-difference time-domain (FDTD) modeling, it can facilitate the establishment of an explicit non-plasmonic C-NP nanosensor of ultra-high Q-factor and sensitivity. In the meantime, such nanosensor design also possesses practical merits. For example, since gain material exists only in the external cavity on which optical pump is exerted, the pathway to the NP will be unobstructed, and the sensing target will safely reach the sensing spot. Unlike SPASER, which utilizes a metallic NP core and crams the gain medium into nanoscale volume, the C-NP architecture offers better gain stability by having significantly longer length for light-gain interaction inside the micrometer scale high-Q cavity. Moreover, in contrast to plasmonic structures, dielectric sensors are generally more compatible with complementary metal-oxide-semiconductor processes and facilitate mass production.

### 2. Theory: The Pseudo-Orthonormal Eigenmode Method

The C-NP system can be modeled by two coupled driven optical resonators\(^{[11,15,18]}\) described by the following matrix equation:

\[
\begin{pmatrix}
\Omega_1 & g \\
g & \Omega_2
\end{pmatrix} - \begin{pmatrix}
\omega & 0 \\
0 & \omega
\end{pmatrix} \begin{pmatrix}
P_1 \\
P_2
\end{pmatrix} = \begin{pmatrix}
f_1 \\
f_2
\end{pmatrix}
\]

(1)

where \(\omega\) is the oscillating frequency, \(P_i\) is the total dipole/multipole moment of the NP from the ports where the electromagnetic energy radiates, \(g \cdot P_2\) is cavity’s total dipole/multipole moment that is coupled to the NP, and \(g\) is the coupling factor between the NP and cavity. The complex resonant frequencies of the NP and cavity are denoted by \(\Omega_1 = \omega_1 - i\gamma_1\) and \(\Omega_2 = \omega_2 - i\gamma_2\), where \(\gamma_1\) and \(\gamma_2\) are the loss factor of the NP and cavity, respectively. Table 1 lists the meaning of all the essential symbols used in this study.

| Symbol | Meaning |
|--------|---------|
| \(P_1, P_2\) | Total dipole/multipole moment at ports of NP and cavity, respectively |
| \(\Omega, \omega\) | Frequency |
| \(\Omega, P\) | Momentum of polarized electron |
| \(F\) | A driving momentum exerted by the external field |
| \(g\) | Coupling factor between NP and cavity; \(g = |g| \exp(\phi_g)\); \(|g|\) - magnitude of coupling; \(\phi_g\) - direction of alignment between the two coupled dipole/multipole moments |
| \(\gamma_1, \gamma_2\) | Intrinsic loss factor of NP and cavity before coupling |
| \(\Delta P/\Delta \omega\) | Dipole/multipole moment change due to a sensing event |

By making the following assignments

\[
M = \begin{pmatrix}
\Omega_1 & g \\
g & \Omega_2
\end{pmatrix}, \ P = \begin{pmatrix}
P_1 \\
P_2
\end{pmatrix}, \ F = \begin{pmatrix}
f_1 \\
f_2
\end{pmatrix}
\]

(2)

we have Equation (1) rewritten as \([M - \omega I]P = F\), where \(I\) is identity matrix and \(MN_i = \alpha_i N_i\) \((n = 1, 2)\) using the nth eigenstate \((N_n)\) and eigenvalue \((\alpha_n)\) of matrix \(M\). Note that the completeness of these eigenstates, forming a complete set, is different from that of the conventional Hermitean matrices in quantum mechanics. Here \(M\) is a complex symmetric matrix, whose eigenstates are able to form a complete, pseudo-orthonormal
basis with the operation $N_i^T N_j = 0$ and $N_j^T N_i = 0$.\[19,20\] In this work, these operations are the pseudo-inner product defined as the simple Euclidean type inner product.\[19,20\]

Using the eigenstates of $M$ to get the expansion of $P$ we have,

$$P = \sum_{n=1,2} \beta_n N_n$$  \hspace{1cm} (3)

$$\sum_{n=1,2} \alpha_n \beta_n N_n = \omega \sum_{n=1,2} \beta_n N_n = F$$  \hspace{1cm} (4)

where $\beta_n$ is the expansion coefficient. Pre-multiplying Equation (4) by the transpose matrix $N_n^T$ to obtain $\alpha_n \beta_n N_n^T - a_n \omega \sum_{n=1,2} \beta_n N_n^T = N_n^T F$, we have the expansion coefficient given by Equation (5).

$$\beta_n = \frac{N_n^T F}{N_n^T \sum_{k=1,2} (\alpha_k - \omega)}$$  \hspace{1cm} (5)

In Equation (5), as $\omega$ tends to $\alpha_n$, the expansion coefficient reaches a pole singularity and tends to infinity. In this case, it implies that the $n$th eigenmode is dominant. The eigenvectors that correspond to the two eigenmodes are listed in Equation (6).

A complete description of the solutions is included in Section S1, Supporting Information.

$$N_1 = \left(\frac{\alpha_1 - \Omega_2}{g}\right), \quad N_2 = \frac{g}{\alpha_2 - \Omega_2}$$  \hspace{1cm} (6)

3. Pinpointing the Pole by POEM

For a given set of known parameters ($\Omega_1, \Omega_2, g$), although it is theoretically feasible to sweep the complex frequency for a pole, it is often challenging as the iterative computations required by the sweep are blind and time-consuming, with no guarantee that the solution converges on a real $\omega$. Without an explicit expression for the root, some mathematical information is lost, making it difficult to understand the physical nature of the system. By analytically solving for the pole, POEM reveals the physics of the C-NP system while demonstrating the possibility of achieving a singularity and high-Q performance for the dielectric NP.

On the other hand, the expression $\Delta P/\Delta \omega$, which signifies the dipole/multipole moment change under a sensing event—an event that is generally interpreted as a local refractive index change upon a sensing event—serves as a characterization index of the sensitivity and is infinitely large at the pole (Equation (5)). In fact, $\Delta P/\Delta \omega$ is proportional to $-1/(\alpha_n - \omega)^2$ and the sensing performance is immediately linked to the system’s closeness to the singularity (Section S1, Supporting Information).

When $\omega_n = \omega_n^* \approx 2\pi f_n$, $g \approx |g| \sin 2\phi_0 \neq 0$, the values of $f_n$ and $g$ satisfy Equation (7) in order for the system to have a pole at a real $\omega$ (derivation in Section S1, Supporting Information).

$$(\gamma_1 + \gamma_2)^2 \gamma_1 \gamma_2 + (\gamma_1 + \gamma_2)^2 |g|^4 \cos 2\phi_0 = |g|^2 \sin^2 2\phi_0$$  \hspace{1cm} (7)

Without loss of generality, the intrinsic frequencies of both resonators are $\omega_1 = \omega_2 = 1$. As an example, $\gamma_1$ is set as 0.25, $|g|$ as 0.004, and $\phi_0$ as 0.9$\pi$. Plugging them into Equation (7), $\gamma_1$ is solved as $-5 \times 10^{-5}$. Leveraging the direct identification of eigenmodes, POEM is able to pinpoint and manifest the resonance in full details. Figure 1a,c,e shows the $|P|_1^2$ topologies around a pole across the $\omega - \gamma_1$, $\omega - |g|$, and $\omega - \phi_0$ planes, respectively, while Figure 1b,d,f reveals $|P|_1^2$ spectra corresponding to an evolving series of $\gamma_1$, $|g|$, and $\phi_0$, respectively. As illustrated by Figure 1a,c, the same pole can be found in the $\omega - \gamma_1$ and $\omega - |g|$ planes when $\gamma_1 = -5 \times 10^{-5}$ and $|g| = 0.004$. When a pole singularity is reached, $|P|_1^2$ reaches infinity, which is represented by a large number in the plots (peak values differ in graphs as different numerical resolutions are used). The lineshape evolution in Figure 1b ($|g| = 0.004$, $\phi_0 = 0.9\pi$) and Figure 1d ($\gamma_1 = -5 \times 10^{-5}$, $\phi_0 = 0.9\pi$) indicate that asymmetric lineshape—known as the Fano resonance—is prominent before the pole.\[21\]

The peak part of the asymmetric lineshape grows when $\gamma_1$ decreases (Figure 1b) or when $|g|$ increases (Figure 1d); as predicted by POEM, the Q-factor of the peak culminates at $\gamma_1 = -5 \times 10^{-5}$ and $|g| = 0.004$, respectively; it then diminishes as $\gamma_1$ grows more negative or as $|g|$ increases further. Note that negative $\gamma_1$ denotes gain in the cavity. From Figure 1a,b, it can be concluded that the pole singularity is the result of an optimum gain. From Figure 1c,d it can be concluded that the pole also needs an optimum coupling strength between the resonators—neither under-coupling ($|g| = 0.001$) nor over-coupling ($|g| = 0.005, 0.01$) yields a pole.

In general, with the remaining parameters defined, $\gamma_1$ or $|g|$ can be solved analytically from Equation (7). Specifically, in order for Equation (7) to have a root at all, \(\sqrt{|\gamma_1 \gamma_2|}\) should be no larger than $|g|$ (derivation in Section S1, Supporting Information). The condition sets up a primary criterion for the singularity identification while bearing crucial implications: for a certain coupling strength, the level of gain in the two-resonator system needs to be optimum, that is, a gain that is too small or big will preclude the singularity.

When $|g| = 0.004$, $\gamma_1 = -5 \times 10^{-5}$, $\phi_0$ varies from 0 to $2\pi$, and the $|P|_1^2$ topology repeats itself every $\pi$ cycle, only the range of $[0, \pi]$ is investigated (Figure 1f). Two poles are found near $\phi_0 = 0.1\pi$ and $0.9\pi$, respectively, with opposite dip/peak sequences in the $\omega$ domain. Symmetric resonances appear for $\phi_0 = 0$ (peak, not shown), $\pi/2$ (dip), and $\pi$ (peak), which serve as the three nodes that divide $[0, \pi]$ into two subsections within which the resonance lineshapes have opposite dip/peak sequences—dip-peak in $[0, \pi/2]$ and peak-dip in $[\pi/2, \pi]$. Similar to the observations in Figure 1b,d, both the peak power and resonance Q-factor increases before the pole and subsequently decreases in Figure 1f. Interestingly, we found that a singularity does not exist for the coupled system when $2\phi_0 = 0$ (Section S1, Supporting Information). In other words, $P_1$ and $P_2$ cannot be in parallel or orthogonal with each other for any singularity to occur; the asymmetric Fano lineshape is a necessary condition.

Additionally, $\phi_0$ directly influences the phase of $P_1/P_2$, a parameter used to determine the material nature of the NP, based on the different polarization nature of dielectric and metallic NPs in the presence of an external electromagnetic field. At the pole, $P_1/P_2$ is equal to $(\alpha_1 - \Omega_2)/g$ or $g/((\alpha_2 - \Omega_2)$. As illustrated in the right panel of Figure 1e, when the phase of $P_1/P_2$ is close to the
positive real axis, the NP is dielectric in nature; if the phase is close to the negative real axis, the NP is metallic in nature. Phenomenologically, the region where phase of \((\phi_{1} - \Omega_{2})/g\) is between \(-\pi/4\) and \(\pi/4\) represents systems that are mainly dielectric while the region confined within \(3\pi/4\) to \(5\pi/4\) denotes systems that are mostly metallic. All poles identified in Figure 1 fall in the dielectric region.

In the C-NP system, the dielectric NP’s ability to produce a pole by implementing assisting resonance from the external environment originates from the coupling of the two modes. The resultant resonance is not the symmetric Lorentzian resonance which is almost universal for standalone metallic NPs, but the asymmetric Fano resonance. Through the interference of scattered field from a low-Q and a high-Q resonance, the dielectric C-NP system fosters Fano resonance that is characteristic of asymmetric lineshapes.\(^{[21]}\) Of the two interacting resonances, the high-Q resonance usually experiences larger phase change, sometimes as large as \(\pi\) if gain is implemented. Consequently, it is able to pass the phase jump onto the low-Q resonance through coupling (as substantiated by the phase change of dipole/multipole moment in Equation (5) when a pole is realized), resulting in the singularity being identified at a practical gain.

4. Non-Plasmonic C-NP Nanosensor

4.1. Design Steps

The C-NP system analyzed by the POEM framework in previous section is materialized here with an explicit structure. Figure 2a illustrates the detailed configurations of the design. The C-NP structure is primarily composed of an open ring and a rectangular nanocube that is placed in the ring’s opening, where the two facets of the ring-opening both maintain a 50 nm gap with the nanocube. The nanocube’s spans in \(x\), \(y\), and \(z\) axis are 400, 350, and 200 nm, respectively, while the ring is built from rectangular waveguide with cross-sectional dimensions of \(350 \times 200\) nm. Both the ring and nanocube are made of SiN (for its wide transparency window that spans from blue to the near infrared\(^{[22]}\)), resting on a SiO\(_2\) substrate and immersed in water to mimic a bio-chemical sensing scenario. Fabrication-induced losses, such as sidewall roughness, are not considered here. Gaussian beam polarized in \(x\)-axis is focused and incident from above onto the nanocube, covering an area slightly larger than the nanocube and gaps combined. The incident light is scattered by the nanocube into the ring where it continues to circulate. Transmitted light...
is then monitored under the NP in the substrate. For sensing, the adsorption of bio-chemical molecules on the NP is modeled as a uniform dielectric layer with 30 nm thickness and slightly higher refractive index than water ($\Delta n$). The visible spectral range is focused as the target molecules generally have higher refractive index in short wavelength.[23,24] The sensing targets attach themselves to the five exposed surfaces of NP which have been surface-functionalized to selectively grab targeted molecules.[25] Consequently, optical transmission would experience a change that signifies the occurrence of a sensing event. For simplicity, we abbreviate the open ring as R, the nanocube as NP, the ring-nanocube structure as $R + NP$, the $R + NP$ structure with adsorption of the sensing target as $R + NP + A$, and the NP with adsorption as $NP + A$.

The above settings are acquired by considering the results of FDTD simulations and design rules derived by POEM. Similar to most theoretical models such as TCM, POEM starts from collecting empirical values by fitting data of the numerical simulations and subsequently performing iterative parametric optimization following the conclusions of the POEM model. Unlike most of the other frameworks, POEM is able to pinpoint a singularity from the complex plane and provide a rather accurate estimate of the design so that the time and resources required by numerical simulations, especially FDTD calculations involving dynamic gain medium, is considerably cut down. The process to reach an estimate for a singularity-bearing C-NP structure usually includes three steps. First, in an effort to fulfill the precondition $\omega_1 = \omega_2$, NP and ring are so designed that their individual resonances are brought close to each other and at least $\omega_1 \approx \omega_2$ is satisfied. Next, in order to guarantee $|g| \geq \sqrt{|\gamma_1| |\gamma_2|}$, the coupling factor $|g|$ (proportional to R’s Purcell factor in FDTD simulation) is maximized in the starting design before optical gain is applied. This is done to avoid blind sweep of the complex plane for a possibly non-existent singularity. Finally, after determining the parameters $(\omega_1, \omega_2, \gamma_1, g)$ for the passive system, we use the solution to Equation (7) to quickly evaluate the optical gain level ($\gamma_2$) required by a pole.

4.2. Passive Structure

To make a fair comparison of the sensing capability of different designs, ratio between the change in collected optical power and the incident power ($\Delta T/T$) is monitored.[26,27] As the time-averaged Poynting vector is proportional to the square magnitude of electric dipole/multipole moment, $\Delta |P|^2$ is calculated in the POEM study to reflect the transmitted power change in the event of molecule adsorption. The $|P|^2$ values are normalized to the spectra peaks of respective bare NPs (i.e., the NP and R + NP).

Coupling between the NP and the external cavity is expected to be optimum when their resonances coincide ($\omega_1 = \omega_2 = 1$). $\gamma_1$ and $\gamma_2$ are 0.25 and 0.00015, respectively; the magnitude of coupling factor $|g|$ is 0.004 and $\phi_1$ is 0.9$\pi$. Upon adsorption of the sensing target, there is a frequency shift on the high-Q resonance due to the added material ($\Delta \omega_0 = 0.0005$). The full parameter settings of the different C-NP systems are summarized in Section S2, Supporting Information. As illustrated by the plots of Figure 2b, adsorption of the sensing target in C-NP architecture yields a sharp asymmetric profile while that of the NP alone results in relatively flat and close-to-zero $\Delta |P|^2$.

The Purcell factor of a structure (the open ring in our case) serves as the major indicator for coupling strength between the NP and the R, (i.e., $|g|$). It is calculated as the ratio of emitted power by a dipole/multipole source (the NP) when the structure is present and when it is absent from the environment.[28] Purcell factor of the open ring is calculated and plotted alongside the nanocube’s scattering cross section across the spectral axis (Figure 2c). It is observed that the Purcell factor exhibits a peak close to the wavelength of 650 nm. To investigate sensing performance under extreme conditions, a small refractive index of 0.05 between the A layer and the water background is set ($\Delta n = 0.05$). The $\Delta T$ spectra corresponding to the R + NP system in Figure 3a show that the $\Delta T$ sensitivity almost follows the spectral profile of the Purcell factor in Figure 2c, with the largest $\Delta T$ (32.8% of incident power) occurring near a wavelength of 655 nm. The resonance at this wavelength has the highest Q-factor in the

![Figure 2](image-url)
Figure 3. a) Transmitted power change ($\Delta T$) of the R + NP (left $y$-axis, solid line) and the NP (right $y$-axis, dashed line) structures by FDTD simulations, $\Delta T_{R+NP} = T_{R+NP} - T_{NP}$, $\Delta T_{NP} = T_{NP} - T_{NP}$. Selected peaks indicated by colors and symbols at 554 nm (green, square), 588 nm (yellow, triangle), 655 nm (red, circle), and 735 nm (crimson, diamond). Inset from left to right: electric field distribution in the $xy$ plane at the structure waist for NP at 655 nm, R + NP at 657 nm, R + NP at 655 nm, and R + NP at 655 nm showing entire R, respectively; first three insets have the same coordinates, all field intensities are normalized to a unified color scale. b) Spectral lineshapes of the R + NP (dashed line) and R + NP + A (solid line) structures zooming in on the selected wavelengths (a).

spectral range considered while the resonance linewidth is around 100 pm. Such concurrence of sensitivity and Purcell factor suggests that the optimum coupling factor [g] corresponds to the case where coupling is strongest, not contradicting with the findings of Figure 1c,d where an optimum [g] is sought. Since the coupling between the R and NP is generally classified as weak coupling, [g] should be maximized. Similar to the results of POEM in Figure 2b, the power change in transmission spectrum for the NP alone is small and flat as compared to that of the R + NP (Figure 3a). Due to a tradeoff between the spectral resolution and spectral coverage in FDTD calculations, Figure 3a is the result of a coarse spectral resolution (500 pm) and broad spectral range (250 nm). Consequently, not all peak features are accurately resolved. Figure 3b recalculates the structure with 50 pm spectral resolution for resonances where the dip wavelength occurs at around 554, 588, 655, and 735 nm, respectively. The results confirm that the sensor is most sensitive at the resonance of 655 nm and is progressively less sensitive as the wavelength moves away from it ($\Delta T_{554} = 5.0\%$, $\Delta T_{588} = 8.5\%$, $\Delta T_{735} = 5.6\%$). The external ring cavity is essential to bring about a high-Q resonance for the creation of even higher Q factor. The first three insets of Figure 3a zoom in on the NP to show, respectively, the electric field distributions in $xy$ plane for the standalone NP at 655 nm, the R + NP at an off-resonance wavelength of 657 nm, and R + NP at the resonance wavelength of 655 nm. Comparison among these field distributions reveals that the R + NP structure displays the strongest field localization on the NP when resonating with an external cavity. A fourth inset on the right illustrates the full view of the on-resonance R + NP, confirming that the R and the NP is complementary to each other in forging a closed pathway along which optical emission from the NP preferably scatters and circulates. As sensing takes place in the near field of NP, the sensitivity depends on the NP resonance as well. To demonstrate that the large $\Delta T$ or $\Delta|P_1|^2$ is attributed to the R + NP (C-NP) integration rather than the corners of the ring opening, sensing performance of the standalone R is investigated. For fair comparison of the R and R + NP, adsorption of sensing target in the standalone R is taken as the addition of a hollow adsorption enclosure (abbreviated as R + A). As a result of small $\Delta n$ and the meager amount of adsorbed molecules, the A layer gives rise to only 50 pm red shift in the resonance dip (Figure 4b by POEM and Figure 4d by FDTD). Without the NP, the resonance linewidth is about 600 pm for both the R and the R + A (Figure 4d); when the NP is in place, the resonance experiences roughly the same amount of spectral shift but resonance linewidth is reduced to 100 pm (Figure 4b by POEM and Figure 4e by FDTD), resulting in much larger $\Delta T$ (Figure 4f) and $\Delta|P_1|^2$ (Figure 4c).

4.3. Gain-Assisted Structure

As depicted in the inset of Figure 5, introducing the right amount of gain into the ring carries the system onto a singularity that is characterized by amplification in light scattering and resonance Q-factor. One important finding of the POEM study is that the system yields infinite sensitivity $\Delta P/\Delta\omega$ at its pole, leading to the apparent necessity to identify an exact pole-supporting design for maximum sensitivity. In this section, FDTD modeling
Figure 4. Results by POEM study on the effect of NP: a) $\mid P_1 \mid^2$ spectra for the R (black solid line) and R + A (red solid line), both curves normalized to the maximum of the R spectrum; b) $\mid P_1 \mid^2$ spectra for the R+NP (dotted line) and R + NP + A (dashed line), both curves normalized to the maximum of the R + NP spectrum; c) $\Delta \mid P_1 \mid^2$ spectra for the R (solid line) and R + NP (dashed line). $\Delta \mid P_1 \mid^2 = \mid P_1 \mid^2_{R} - \mid P_1 \mid^2_{R+NP}$. FDTD simulation results of the R and R + NP structure: d) power spectra of the R (square) and R + A structure (circle); e) power spectra of the R + NP (dotted line, square) and R + NP + A (dashed line, circle); f) $\Delta T$ for the R (solid line, square) and R + NP (dashed line, circle). $\Delta T_R = T_{R+A} - T_R$, $\Delta T_{R+NP} = T_{R+NP+A} - T_{R+NP}$.

Figure 5. Transmission spectra of the gain-assisted R + NP + A structure when $N_p = 2 \times 10^9$, $2.4 \times 10^9$, and $2.8 \times 10^9$ s$^{-1}$. Transmission of $N_p = 2.8 \times 10^9$ s$^{-1}$ is halved for sake of display clarity. Spectral data points are indicated by red spheres (resolution = 10 pm). Inset: schematic showing the introduction of optical gain into the ring to realize a pole singularity for the R + NP system.
with dynamic four-level atomic system is used to confirm the findings of POEM study and Additionally disclose a practical sensing device operating around its pole. Though FDTD method incorporating semi-classical model of the four-level atomic system is, to our knowledge, the best approach to describe and predict the optical properties of the active system, it is still exceedingly time-consuming. The analytical solution uncovered by POEM transforms the pole-searching endeavor from exhaustive sweep of a plane to a rather accurate pinpointing exercise; the guidelines thus interpreted equally scales down the challenge faced by design processes using FDTD modeling (see Section S4, Supporting Information, for details on FDTD modeling).

The gain medium is applied exclusively to the open ring, which is optically pumped at 488 nm and emits at 550 nm. As the optical pumping is applied solely to the ring and the NP is well isolated from it, the gain observed on the NP even when it is made of the same gain material is negligible. Hence, in practice, there is no need for selective doping into different devices that would potentially increase the fabrication complexity. Radius of the open ring is reduced to 2 μm so its peak Purcell factor is brought close to 550 nm. All the other structural settings of the passive system (Figure 2a) are kept—including the dimension of the NP, as there is little difference in the required individual resonance (ω1 ≈ ω2 still holds). Optical gain in the R is gradually increased by varying the pumping rate Np while ion concentration N0 is maintained at 1 × 10^23 m^−3. Details of the gain material can be found in Sections S3 and S4, Supporting Information.

The process of finding the gain level that pushes the R + NP system onto a pole, designated as the critical gain, is illustrated in Figure 5. Critical gain is sought for the R + NP + A structure while Δn is reduced to 0.01. For Np <2.4 × 10^8 s^-1, the transmitted resonance exhibits asymmetric lineshape where the peak culminates at around 553.3 nm and slowly increases with Np. At Np = 2.0 × 10^9 s^-1, the peak power is 15.4 and the resonance’s full width at half maximum (FWHM) is about 20 pm. In this case, the corresponding gain coefficient is calculated as 16.1 cm^-1 and the system is said to be under-amplified because there is not enough gain for pole to occur. When Np increases to 2.4 × 10^9 s^-1 (gain coefficient ≈ 16.9 cm^-1), the emission peak experiences a power outburst to 1052 while the FWHM decreases to around 10 pm. Increasing Np further while keeping the gain below saturation results in resonance with a higher peak but smaller Q-factor; when Np = 2.8 × 10^9 s^-1 (gain coefficient ≈ 17.7 cm^-1), the peak grows to 2334 and FWHM to 50 pm. In this case, the system is considered over-amplified as the gain in this case is larger than the critical gain. Information on gain coefficient calculation can be found in Section S4, Supporting Information.

As summarized by the schematic illustration of Figure 6a, the gain-assisted R + NP system essentially differs from standalone resonant system by reaching a singularity at much lower gain than the lasing threshold. The symmetric Lorentzian resonance of a standalone system (|g| = 0) with loss (γ2) needs a hugely negative γ2 to fully compensate for the losses. Therefore, the Q-factor of a conventional laser based on symmetric resonance is primarily limited by gain saturation, and more often than not, even the highest Q-factor does not coincide with a pole. The R + NP system, on the other hand, involves asymmetric Fano resonances, where the coupling factor is comparable with γ1 and γ2, and the critical gain for pole requires a very small optical gain that can be readily achieved in laboratory conditions. Most importantly, the fundamental difference between pole and lasing threshold is that the pole is a singular point with infinite Q-factor corresponding uniquely to critical gain, where the Q-factor starts to diminish with smaller or bigger values.

It should be pointed out that the singularity created by critical gain in the R + NP + A system is not due to lasing of the open ring alone. In order to confirm this supposition, the same simulation is performed at critical gain but with two changes in the settings. First, given the structural orientation of inset in Figure 6b, we place two time-field monitors inside the ring at twelve o’clock (Monitor 12) and three o’clock (Monitor 3), respectively. Next, we reduce the simulation domain by shifting one side of the perfectly matched layer so that it cuts through the lower half of the ring (straight line, inset Figure 6b). Light incident on the NP creates two counter-propagating pathways in the ring. By excluding half of the ring from the simulation domain, counter-clockwise propagating light stops and only the clockwise propagating light reaches Monitor 3. The enveloped field amplitudes in Figure 6b confirm the fact that the current gain (critical gain) is not able to compensate the loss in the R.

When evaluating sensing performances of a high-Q system, such as the R + NP operated around a pole, comprehensive
assessment is no longer reflected by $\Delta T$ alone. Here, a new criterion is set as the ratio of non-overlapped area between the $T_{RNP}$ and $T_{RNP+A}$ spectra over the total integrated area (550–553 nm). The illustration in the inset of Figure 7c defines the calculation scheme of the non-overlapped area ratio (NOAR). The non-overlapped area is the area sum of all patches confined between the $T_{NP}$ and $T_{NP+A}$ spectra (patch A and B). Figure 7b illustrates an example of such non-overlapped area with pink shades. The total integrated area is the area confined between the transmitted curve and the straight line at $T = 0$ (patch A, B, C). Figure 7d shows an example of the total integrated area with green shades. The resultant NOAR is then calculated from $(A + B) / (A + B + C)$. The non-overlapped area accounts for the degree of apartness between the two resonances and NOAR serves as a compendious figure of merit (FOM) that simultaneously reflects the absolute power change, the relative spectral shift, and the resonance Q-factor. Specifically, sensors utilizing high-Q resonance lineshapes are especially immune to the effect of noise. In the four cases shown in Figure 7, the NOARs are calculated as 0.11%, 12.4%, 81.9%, and 50.0%, respectively.

In Figure 7a, both spectra dips possess linewidths around 350 pm while spectral shift and maximum $\Delta T$ upon adsorption of sensing target are 20 pm and 1.1%, respectively. A small NOAR is expected as both $\Delta T$ and resonance Q-factor are small. When the theoretical gain is incorporated and $N_p = 2.0 \times 10^9$ s$^{-1}$, the amplified peak of the $T_{RNP+A}$ spectrum has a FWHM of 20 pm (Figure 7b). Peak shift upon adsorption of sensing target is 30 pm. The gradual evolvement of peak’s Q-factor from Figure 7a (dashed line, circle) to Figure 7b (solid line, filled circle) aligns with the behavior depicted in Figure 1b when the external cavity turns from no gain ($\gamma_2 = 1.5 \times 10^{-5}$) to a small gain ($\gamma_2 = 0$).

At $N_p = 2.4 \times 10^9$ s$^{-1}$, resonance supported by the $R + NP + A$ structure ($T_{RNP+A}$) reaches a pole with FWHM = 10 pm (Figure 7c). Given the same gain, the $R + NP$ structure is under-amplified and the peak height is two orders of magnitude smaller than that of the $T_{RNP+A}$. When $N_p = 2.8 \times 10^9$ s$^{-1}$, both $R + NP$ and $R + NP + A$ structures are over-amplified. The FWHM increases to 45 pm for $R + NP$ structure and 50 pm for $R + NP + A$ structure. The spectral shift in peak resonance upon adsorption of sensing target is around 30 and 20 pm in Figures 7c and 7d, respectively. By comparing the NOAR among different gain conditions of Figure 7, it is found that when gain is set close to the system’s pole, an optimized balance between $\Delta T$, spectral shift, and resonance linewidth can be achieved. On the other hand, the lineshape evolution from pole (Figure 7c solid line) to over-amplification (Figure 7d solid line) is similar with the pattern shown in Figure 1b where $\gamma_2$ turns from just realizing the pole ($-5 \times 10^{-5}$) to being over-amplified ($-2 \times 10^{-4}$). Alternative sensing scenario based on pole singularity of the $R + NP$ structure is presented in Section S5, Supporting Information.

To have an idea of how the $R + NP$ structure works as a nanosensor under different amplification conditions, NOAR is
calculated for a range of pumping rates. As shown in Figure 8, the $N_p$ value spans from $0.1 \times 10^9$ to $50 \times 10^9$ s$^{-1}$, which corresponds to a gain coefficient of $12.3-112.1$ cm$^{-1}$. The sensitivity is close to zero when gain is small. It then starts to climb steeply at around $2.3 \times 10^9$ s$^{-1}$, before jumping to the pole when $N_p = 2.4 \times 10^9$ s$^{-1}$. However, NOAR does not peak at the pole. It continues to go up before reaching the maximum at $N_p = 2.48 \times 10^9$ s$^{-1}$ (inset of Figure 8). The sensitivity does not decrease instantly after the pole because the R + NP structure is still under-amplified when resonance intensity of the R + NP + A structure keeps on rising with the over-amplification. As gain increases further and both structures are over-amplified, the spectral overlap between the two resonances grows and NOAR decreases. The downturn of the NOAR after its peak is a quick drop at first, and subsequently much slower decline at around $N_p = 3 \times 10^9$ s$^{-1}$. As a result, the gentle slope of the over-amplified resonances unwraps a range of moderate NOARs (yellow region), that could potentially expand the dynamic range of the sensor. With gain saturation happening at around $N_p = 32 \times 10^9$ s$^{-1}$, the sensitivity falls and eventually tends to zero. From the inset of Figure 8, we observe that the giant NOAR bestowed by the pole is confined to the narrow band of over-amplification immediately after the pole (yellow area). This means that the extreme performance of the nanosensor is allowed not only for a single point, but is possible for a range of gain values. It also suggests that the R + NP system operating around a singularity possesses tolerance to gain fluctuation, where small perturbation in the gain conditions would not deviate the sensing performance so much that it totally loses effect.

In fact, sensitivity and stability are two opposing attributes that can never be resolved. A highly sensitive system is bound to be less resistant to external disturbances. In the practical implementation of a nanosensor operated around the pole, tradeoff must be found between the sensitivity and design tolerance. Tolerance assessment is essential as uncertainties in fabrication and experiments might result in deviations that totally invalidate the sensing performance. \cite{16, 17} In Section S6, Supporting Information, effect of different geometric variations are studied and the results show that the current R + NP structure has good tolerance for device perturbations, with pole successfully identified for most of the cases and an average NOAR of above 50% returned. In particular, those variation scenarios in which the ring cavity’s effective path length is altered (such as changing the waveguide width, ring radius, and refractive index) sometimes lead to failure of finding a pole and consequently rather small sensitivity. The reason for such behavior is because the perturbed cavity no longer supports a resonance wavelength equal to the new root of the perturbed system. In this sense, the advantage of high density of states as a result of long effective cavity length is evident: by having longer effective path length, higher density of resonance occurrence is achieved, which leads to increased probability of matching with the new root. To demonstrate such possibility, a pole-possessing R + NP structure with larger ring radius is found following the same design rules of POEM. As a result, pole singularity is identified at a critical gain level of 0.41 cm$^{-1}$ and an NOAR of 77.81%. Detailed explanations and results for the R + NP structure with larger ring radius can be found in Section S6.6, Supporting Information.

5. Conclusion

POEM, as a useful tool to perform fast pole-identification in the complex plane, is first formulated. Using POEM, we confirm the possibility of a non-plasmonic sensing system approaching its sensing singularity at real frequencies when given suitable structural settings. The pole is based on an asymmetric Fano resonance supported by the C-NP architecture. Following the guidelines derived in POEM, we reveal an explicit sensor structure, with practical material design. Giant sensitivity is demonstrated when the sensor leverages the singularity envisaged by the POEM study. Additionally, we define new FOM for gain-assisted nanosensing. Such FOM accounts for the resonance Q-factor, spectral shift, and power change, and equally holds in gainless scenarios. Of significance, the pole of the identified nanosensor requires a small optical gain, which is shown to be of a lower
level than that is needed to compensate the system’s total loss. Such small optical gain can be readily achieved, making experimental realization easier. The C-NP architecture, on the other hand, separates pump from the sensor, effectively insulating the sensing targets from the damaging pump pulse. Besides the active C-NP system investigated in this work, POEM can be used to study any other type of active or passive coupled systems as well. As the formalism suggests, POEM is not restricted to treat a system with only two elements, but can generally treat coupled resonance with any number of elements. Consequently, the design and method proposed by this work provide potential solutions to a wide range of applications including Fano laser and bound states in the continuum based on non-periodic structures.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

**Keywords**

lasing singularity, nanosensors, non-Hermitian systems, ultra-sensitivity

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