Research article

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A complete phase diagram for dark-bright coupled plasmonic systems: applicability of Fano's formula

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Abstract: Although coupled plasmonic systems have been extensively studied in the past decades, their theoretical understanding is still far from satisfactory. Here, based on experimental and numerical studies on a series of symmetry-broken nano-patch plasmonic resonators, we found that Fano's formula, widely used in modeling such systems previously, works well for one polarization but completely fails for another polarization. In contrast, a two-mode coupled-mode theory

(CMT) can interpret all experimental results well. This motivated us to employ the CMT to establish a complete phase diagram for such coupled plasmonic systems, which not only revealed the diversified effects and their governing physics in different phase regions, but more importantly, also justifies the applicabilities of two simplified models (including Fano's formula) derived previously. Our results present a unified picture for the distinct effects discovered in such systems, which can facilitate people's understanding of the governing physics and can design functional devices facing requests for diversified applications.

Keywords: Fano resonance; Fano's formula; coupled-mode theory; coupled plasmonic systems.

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1 Introduction

Plasmonic resonances in nano-metallic particles, caused by collective oscillations of free electrons inside the particles associated with electromagnetic (EM) waves, have attracted intensive attention recently. Compared to plasmonic structures possessing single-mode resonances, complex plasmonic systems involving two and more modes coupled together exhibit even more fascinating optical properties [1–16]. In particular, inter-mode couplings can generate “dressed” plasmonic modes in such systems, which can significantly modify the optical responses of the whole systems, leading to intriguing physical effects such as Fano resonances [1–6], Rabi oscillations [7–10], and plasmon-induced transparencies [11, 12]. Owing to their extraordinary properties such as local field confinement and freely engineered far-field spectrum line shapes, the coupled plasmonic systems were widely used in different application scenarios including bio-chemical sensing [17–20], plasmonic circuit [21–23], fluorescence enhancements [24–26], and more recently

Figure 1: Schematic of the coupled plasmonic system.

(A) The investigated samples that exhibit “Fano-like” resonances in the NIR regime. (B) SEM image of part of one fabricated sample.

meta-atom designs for realizing functional meta-devices [27–32].

Despite of numerous experimental progresses already achieved, theoretical understanding of these intriguing effects is still far from satisfactory. While numerical simulations can explain experimental findings well in most cases, they are basically repeating the experiments in computers and thus shed very little light on the inherent physics. Meanwhile, scientists have proposed several simplified models, such as Fano’s formula [33, 34] and the independent-oscillator model [35–38], to understand the rich physics discovered in different systems. Thus, it is highly desired to have a unified theoretical framework for such complex systems, which can not only help researchers choose the correct models to study their own systems, but more importantly, also guide people to design appropriate systems to meet their own application requirements.

In this paper, we combine experimental, numerical, and theoretical efforts to address these issues by establishing a complete phase diagram for plasmonic dark-bright-mode coupled systems. We start by studying a series of gold nano-square-resonators with broken symmetries experimentally, and show that while Fano’s formula can explain the spectra obtained under one incident polarization well, it completely fails for those obtained under another polarization (see Section 2). To understand such intriguing results, next we employ a two-mode coupled-mode-theory (CMT) [39–43] to re-examine the problems, and find that the CMT can explain all experimental findings well (see Section 3). Based on the two-mode CMT, we finally establish in Section 4 a complete phase diagram for such dark-bright coupled systems, which not only provides a unified picture to understand all the fascinating optical effects discovered, but more importantly, also

reveals the applicable regions of those simplified models derived previously. We conclude this paper in Section 5.

2 Experimental studies on a series of examples: issues arising

We start by experimentally investigating a series of samples that exhibit “Fano-like” resonances in the near-infrared (NIR) regime (Figure 1A).¹ As shown in Figure 1B, each fabricated sample contains a periodic array of 38 nm-thick gold patches with sizes 440 nm × 490 nm, arranged in a square lattice with a periodicity of 680 nm, deposited on a quartz (Qz) substrate. Each gold patch is drilled with a 260 nm × 260 nm air hole 260 nm with the center displaced from the patch center by a distance d , which is an important parameter to characterize the degree of symmetry-breaking in the sample. We have fabricated a series of samples with d varying from 0 to 80 nm with a step of 20 nm, with Figure 1B showing the scanning electron microscopy (SEM) image of one fabricated sample with $d = 60$ nm (see SEM images of all fabricated samples with different d in Section 1 of the Supplementary Material).

With these samples to hand, we then experimentally measured their optical transmission spectra under normally incident lights with two different polarizations. Open circles in the left panels in Figure 2A–E and Figure 3A–E depict the measured transmission spectra of

¹ We note that the fabricated structures have rounded corners at the edges. We have also performed simulations on rounded-edge structures (see Section 2 in the Supplementary Material), and found that the presence of rounded edges does not affect our main conclusions.

Figure 2: Experimental and numerical results of coupled plasmonic systems for x-polarization.

(A–E) Measured (open symbols in the left panels) and simulated (solid black curves in the right panels) transmission spectra of samples with different values of d , with (F) x-polarized incident light. (G, H) The simulated $\text{Re}(E_z)$ patterns of the symmetric nanostructure at frequencies corresponding to the dipole mode (labeled as the mode “I”) and the quadrupole mode (labeled as the mode “II”) for x-polarization. Dotted lines represent the resonant frequencies of the bright and dark modes with different values of d for x-polarization.

these samples with polarizations $\mathbf{E} \parallel \hat{x}$ and $\mathbf{E} \parallel \hat{y}$, respectively. The measured spectra exhibit intriguing evolutions as d varies from 0 nm (top) to 80 nm (bottom), indicating the important role played by the symmetry breaking. Specifically, the transmission spectra of the symmetry-protected sample (i.e. $d=0$ nm) contain two low-Q resonances at frequencies around 168 THz and 186 THz, for two different polarizations, respectively. As the degree of symmetry-breaking increases, two high-Q dips appear in the corresponding transmission spectra, which become stronger as d is further enlarged. Meanwhile, the transmission line-shapes of two different polarizations all exhibit interesting asymmetric properties, being typical features of Fano resonances [1–6, 44, 45].

We employed finite-difference time-domain (FDTD) simulations² to calculate the transmission spectra of these systems. The FDTD-simulated transmission spectra are shown in the right panels of Figure 2A–E and Figure 3A–E.

Apart from some deviations in the low-frequency regime (near 150 THz), the FDTD results are in reasonable agreement with experimental ones.³ In particular, all essential features of experimental spectra, such as evolutions of the dip positions and asymmetric line-shapes, have been reasonably reproduced by simulations. The FDTD simulations also revealed the nature of low-Q and high-Q modes, respectively. As shown in Figures 2G and 3G, the low-Q modes are essentially the dipolar resonances of the gold patch under excitations with two polarizations. As the patch length along the x direction is slightly longer than that along the y direction, it is not surprising to see that the frequency of the x-polarized mode is slightly lower than that of the y-polarized one. Because these dipole resonances can efficiently radiate to free space, they are usually called the “bright” modes. Meanwhile, FDTD simulations revealed that the high-Q modes for two different polarizations are two quadrupole resonances, as shown in Figures 2H and 3H, calculated in a typical sample with

² In our simulations, the permittivity of gold is described by the Drude model with plasma frequency 1.37×10^{16} rad/s and the collision frequency 2×10^{14} rad/s, and the refractive index of glass substrate is set as 1.5.

³ Discrepancies between experiments and simulations are possibly caused by adopting the simple Drude model for gold neglecting the contribution of inter-band transition, which also occur in other systems involving gold in the near infrared regime [18, 20].

Figure 3: Experimental and numerical results of coupled plasmonic systems for y -polarization.

(A–E) Measured (open symbols in the left panels) and simulated (solid black curves in the right panels) transmission spectra of samples with different values of d , with (F) y -polarized incident light. (G, H) The simulated $\text{Re}(E_z)$ patterns of the symmetric nanostructure at frequencies corresponding to the dipole mode (labeled as the mode “I”) and the quadrupole mode (labeled as the mode “II”) for y -polarization. Dotted lines represent the resonant frequencies of the bright and dark modes with different values of d for y -polarization.

$d=20$ nm⁴. The antiparallel distributions of the excited currents along two orientated arms are the typical features of quadrupole resonances, implying that such modes are “dark” in nature which cannot radiate to free space efficiently. In fact, in the ideal symmetry-protected case, such modes are completely dark when “seen” at the normal incidence (see Figures 2H and 3H).

However, although FDTD simulations can reproduce all essential features of experimental results well, very little physics can be gained from these calculations. In fact, the experimental spectra exhibit distinct behaviors for two polarizations as d increases, which cannot be easily understood by FDTD simulations. For example, it is not easy to understand how the symmetry-breaking parameter d dictates the evolutions of optical behaviors in two cases, which are obviously different.

To reveal the physics behind the experimental and numerical results presented in Figures 2 and 3, we now employ Fano’s formula [1], originally developed for electron systems and widely used in photonics recently, to

interpret the obtained results. According to Fano’s theory, the transmission spectra $|t|^2$ of such bright-dark-coupled systems can be modeled by the formula [33–34]

$$|t|^2 = |t_0|^2 \frac{|\alpha + \varepsilon|^2}{|i + \varepsilon|^2}, \quad (1)$$

where t_0 is the complex transmission coefficient of the symmetry-protected sample (with $d=0$ nm), $\varepsilon = (f - F_q + i\Gamma'_q)/\Gamma_q$ with F_q denoting the resonant frequency of the “dressed dark mode”, $\Gamma'_q(\Gamma_q)$ standing for the absorption (radiation) loss of this dressed mode, and $\alpha = \cot\delta$ is the Fano parameter (with δ being the phase shift of the “continuum background”) which can be treated as a fitting parameter here to model the degree of asymmetry of the line-shape [35, 46]. It is clear that Fano’s formula [Eq. (1)] only needs four fitting parameters (Γ_q , Γ'_q , F_q , α), which collectively define the properties of the dressed dark modes loaded on the bright-mode background given by t_0 retrieved from the symmetry-protected spectra.

We now use Eq. (1) to fit the FDTD-simulated transmission spectra. Through varying the four parameters (Γ_q , Γ'_q , F_q , α), we obtained the Fano spectra that best fit the corresponding FDTD spectra, and then depicted the results as dotted

⁴ As this mode is completely “dark” in the symmetry-protected sample, we choose the sample with the weakest symmetry breaking to approximately illustrate the field pattern of this mode.

Figure 4: Analyses on the applicability of Fano's formula.

(A, E) FDTD-simulated (solid black curves) and Fano-formula-fitted (red dotted curves) spectra of samples with different values of d , shined by normally incident lights with two different polarizations. (B–C, F–G) Retrieved Fano-formula parameters as functions of d . (D, H) RMSE between the Fano-fitted and FDTD spectra as functions of d in two different polarizations.

lines in Figure 4A and E. The retrieved Fano parameters are depicted in Figure 4B, C and 4F, G. Compared with the FDTD spectra, the Fano spectra can reproduce all FDTD spectra with x -polarization well, but exhibit significant deviations for the y -polarization cases, especially for samples with $d > 40$ nm. Such deviations are more clearly seen in Figure 4D and H, where the root-mean square errors (RMSE)⁵ between the Fano and FDTD spectra are depicted as functions of

⁵ The root-mean square error is defined as $\sigma = \sqrt{\int_{f_1}^{f_2} |t_{\text{Theory}} - t_{\text{FDTD}}|^2 df} / \sqrt{\gamma_p}$, where t_{theory} and t_{FDTD} is the transmittance amplitude of corresponding theoretical formula and FDTD simulation, and the light's frequency from f_1 to f_2 are our considering waveband. γ_p is the radiation loss rate of the bright mode.

d for both polarizations. While RMSE remain at very small values for all the cases studied under x -polarization, they become significantly enlarged for the y -polarization cases with $d > 40$ nm. We emphasize that such deviations cannot be diminished by varying the fitting parameters, but rather are caused by the fact that the bright-mode dip in FDTD simulated y -polarized spectra exhibits a clear red shift as d increases. The latter can *never* be modeled by Fano's formula [Eq. (1)] assuming a “passive” background.

Apart from the issues arising in the fitting processes, another problem is that Fano's theory cannot tell us why the fitting parameters should behave like those depicted in Figure 4B, C and 4F, G, which are important to understand the underlying physics. For example, it is very difficult to

understand, based on Fano's formula only, why the dark-mode dip in the x -polarized spectra undergoes a strong red shift as d is increasing while the frequency shifts toward an *opposite* direction for the y -polarized spectra. All these difficulties motivate us to employ a more generic theory to re-examine the same problems, in order to explain the not only experimental results but also to help better understand the inherent physics.

3 Re-explain the experimental results by the CMT

In Section 2, we have seen that the failure of Fano's formula is essentially caused by its assumption that the “bright” mode only provides a “passive” background. Such an assumption is certainly correct for the electron case and is also valid for photonic systems with dark mode interacting “weakly” with the bright mode. However, in general the bright mode should also be treated as a “mode” rather than a passive background. Based on such understanding, we now establish a generic theoretical framework based on the two-mode CMT to re-examine the optical properties of such symmetry-broken systems, which naturally support two “modes” as already revealed by the FDTD simulations (see Figures 2 and 3).

We start by studying the dynamical evolutions of modes in such systems, neglecting losses due to both radiation and absorption, for the moment. According to the Hamiltonian formalism for photonic systems established in Refs. [47–50] the two modes described in Figure 2G, H and Figure 3G, H are two “eigenmodes” of the symmetry-protected system (with a Hamiltonian given by $\hat{\mathbf{H}}_0$): $\hat{\mathbf{H}}_0|\Psi_p\rangle = 2\pi f_p|\Psi_p\rangle$, $\hat{\mathbf{H}}_0|\Psi_q\rangle = 2\pi f_q|\Psi_q\rangle$. In symmetry-broken systems with a Hamiltonian given by $\hat{\mathbf{H}} = \hat{\mathbf{H}}_0 + \hat{\Delta}$ where $\hat{\Delta}$ accounts for the additional potential contributed by the air-hole dislocation, the total wave function $|\Psi\rangle$ of the systems can be expressed as a linear combination of the bases $|\Psi_p\rangle$ and $|\Psi_q\rangle$: $|\Psi\rangle = a_p|\Psi_p\rangle + a_q|\Psi_q\rangle$, where a_p and a_q are the amplitudes of the two modes. Putting this expression to the Hamiltonian equation $i\frac{\partial}{\partial t}|\Psi\rangle = \hat{\mathbf{H}}|\Psi\rangle$ and utilizing the orthogonal property between $|\Psi_p\rangle$ and $|\Psi_q\rangle$ dictated by the symmetry, one can straightforwardly derive the following equations governing the time evolutions of two mode amplitudes:

$$\frac{1}{2\pi} \frac{\partial}{\partial t} \begin{pmatrix} a_p \\ a_q \end{pmatrix} = -i \begin{pmatrix} f_p + \kappa_{pp} & \kappa_{pq} \\ \kappa_{qp} & f_q + \kappa_{qq} \end{pmatrix} \begin{pmatrix} a_p \\ a_q \end{pmatrix}, \quad (2)$$

where κ_{pp} and κ_{qq} denote the on-site corrections by the perturbation ($\hat{\Delta}$) and $\kappa_{pq} = \kappa_{qp} = \kappa$ describes the near-field coupling between two original modes, again contributed by the perturbation ($\hat{\Delta}$).

We now extend Eq. (2) (valid only for the closed systems) to the open systems studied here by adding back in the absorption/radiation losses of the modes, as well as the couplings of the modes to the excitation port:

$$\frac{1}{2\pi} \frac{\partial}{\partial t} \begin{pmatrix} a_p \\ a_q \end{pmatrix} = -i \left[\begin{pmatrix} f_p + \kappa_{pp} & \kappa_{pq} \\ \kappa_{qp} & f_q + \kappa_{qq} \end{pmatrix} - i \begin{pmatrix} \gamma'_p & 0 \\ 0 & \gamma'_q \end{pmatrix} \right] \begin{pmatrix} a_p \\ a_q \end{pmatrix} + \begin{pmatrix} -\gamma_p & X \\ X & -\gamma_q \end{pmatrix} \begin{pmatrix} a_p \\ a_q \end{pmatrix} + \begin{pmatrix} d_{1p} & d_{2p} \\ d_{1q} & d_{2q} \end{pmatrix} S^+. \quad (3)$$

Here, $\gamma_{l=p,q}$ and $\gamma'_{l=p,q}$ account for, respectively, the radiation and absorption losses of the two modes, X is the far field coupling, $S^+ = (s_1^+ s_2^+)^T$ denotes the strength of incident excitation, and $d_{j=l=p,q}$ describes the coupling between external light and the bright mode at the port labeled by $j=1, 2$. Here, we note that the near-field coupling has no influence on the dissipation of the system, as those coupling parameters ($\kappa, \kappa_{pp}, \kappa_{qq}$) are essentially real numbers. Such an approximation is valid as the real part of ϵ_{gold} (associated with the real parts of those coupling parameters) is much larger than the imaginary part of ϵ_{gold} (associated with the imaginary parts of those coupling parameters) in this frequency regime. According to time inversion symmetry and energy conservation, we get $d_{2p} = \eta d_{1p} = i\eta \sqrt{2\gamma_p} / (\eta^2 + 1)$ where η is parameter to describe the asymmetries of the modes' radiations toward two ports, caused by the presence of a substrate here [42]. It is worth noting that, as the dark mode is completely radiation-free, its coupling with the far field and with the bright mode is naturally 0, i.e. $\gamma_q = X = |d_{1q}|^2 = |d_{2q}|^2 = 0$ THz. To avoid using two parameters, we have assumed that $\gamma'_p \approx \gamma'_q = \gamma'$ for simplicity.⁶

To reveal the underlying physics further, we diagonalize the matrix containing the near-field coupling strength κ in Eq. (3), and arrive at the following equation describing the evolutions of the amplitudes of two collective modes $\tilde{a}_{\pm}$:

$$\frac{1}{2\pi} \frac{\partial}{\partial t} \begin{pmatrix} \tilde{a}_+ \\ \tilde{a}_- \end{pmatrix} = -i \left[\begin{pmatrix} \tilde{f}_+ & 0 \\ 0 & \tilde{f}_- \end{pmatrix} - i \begin{pmatrix} \tilde{\gamma}' & 0 \\ 0 & \tilde{\gamma}' \end{pmatrix} \right] \begin{pmatrix} \tilde{a}_+ \\ \tilde{a}_- \end{pmatrix} + \begin{pmatrix} -\tilde{\gamma}_+ & \tilde{X} \\ \tilde{X} & -\tilde{\gamma}_- \end{pmatrix} \begin{pmatrix} \tilde{a}_+ \\ \tilde{a}_- \end{pmatrix} + \begin{pmatrix} \tilde{d}_{1+} & \tilde{d}_{2+} \\ \tilde{d}_{1-} & \tilde{d}_{2-} \end{pmatrix} S^+. \quad (4)$$

⁶ Here, we note that the magnitude of eigen wave-functions of bright mode and dark mode are similar (see Figures 2G, H and 3G, H), so that the intrinsic loss of these two mode can be approximately same.

Here we introduced the following quantities $\bar{f} = (f_p + \kappa_{pp} + f_q + \kappa_{qq})/2$, $\Delta f = (f_q + \kappa_{qq} - f_p - \kappa_{pp})/2$ and $\Delta \tilde{f} = \sqrt{\Delta f^2 + \kappa^2}$, so that we can rewrite the original parameters as $\tilde{f}_\pm = \bar{f} \mp \Delta \tilde{f}$, $\tilde{\gamma}_\pm = \gamma_p (\Delta \tilde{f} \pm \Delta f) / 2\Delta \tilde{f}$, $\tilde{\gamma}' = \gamma'$, and $\tilde{d}_{j\pm} = \pm d_{jp} \kappa / \sqrt{\Delta \tilde{f} (\Delta \tilde{f} \mp \Delta f)}$ ($j=1, 2$).

We note that the far-field coupling $\tilde{X}$ between two collective modes is not a free parameter, but is determined by $\tilde{X} = -\sqrt{\tilde{\gamma}_+ \tilde{\gamma}_-} = -\frac{1}{2} \kappa \gamma_p / \sqrt{\Delta f^2 + \kappa^2}$, according to the energy conservation [51]. As the trace of a matrix remains invariant under orthogonal transformations, we find that $\tilde{\gamma}_+ + \tilde{\gamma}_- = \gamma_p$. Obviously, $\tilde{X}$ reaches its maximum as $\tilde{\gamma}_+ = \tilde{\gamma}_-$ which corresponds to the situation that the near-field κ is infinite, and is exactly 0 in the case of $\kappa=0$. In general, the far-field coupling $\tilde{X}$ is an increasing function of κ . This property may play an important role to help us understand the physics, as we explain in the following.

Through standard CMT analyses, we find that the transmission coefficients and reflection coefficients are given by

$$\begin{aligned} r &= r_{Qz} - \frac{2}{\eta^2 + 1} \frac{\tilde{W}_+ \tilde{\gamma}_- + \tilde{W}_- \tilde{\gamma}_+ - 2\tilde{X}^2}{\tilde{W}_+ \tilde{W}_- - \tilde{X}^2}, \\ t &= t_{Qz} - \frac{2\eta}{\eta^2 + 1} \frac{\tilde{W}_+ \tilde{\gamma}_- + \tilde{W}_- \tilde{\gamma}_+ - 2\tilde{X}^2}{\tilde{W}_+ \tilde{W}_- - \tilde{X}^2}, \end{aligned} \quad (5)$$

where $\tilde{W}_l = -i(f - \tilde{f}_l) + \tilde{\gamma}_l + \tilde{\gamma}'_l$ with $l=p, q$. Here, r_{Qz} and t_{Qz} are the transmission and reflection coefficients of the quartz substrate alone. Due to the presence of the quartz substrate, the asymmetry coefficient is non-zero, and is found to be $\eta = \sqrt{n}$ with n being the refractive index of the quartz substrate. Detailed derivations of Eq. (5) can be found in Section 3 of the Supplementary Material.

It is helpful to discuss the physical meanings of all independent CMT parameters ($f_p, f_q, \gamma_p, \gamma', \kappa, \kappa_{pp}, \kappa_{qq}$) before starting to fit the FDTD spectra with the CMT ones. f_p, f_q, γ_p , and γ' describe the properties of two *original* modes supported in the symmetry-protected sample (with $d=0$ nm),⁷ which are independent of the symmetry-breaking parameter d . Meanwhile, $\kappa, \kappa_{pp}, \kappa_{qq}$ describe the “coupling” between two original modes, which depend “sensitively” on the symmetry-breaking parameter d , and must be

treated as fitting parameters as functions of d . Based on these understandings, we successfully retrieved all fitting parameters as functions of d , based on which the CMT spectra can best fit the corresponding FDTD spectra.

Figure 5A and D compare the CMT-fitted transmittance spectra with the FDTD ones for two different incident polarizations, showing excellent agreement between them. The eigenmode-related parameters in our CMT model are found to be $f_p=167.6(186.3)$, $f_q=294(217.5)$, $\gamma_p=59(56)$, $\gamma'=4(3.8)$, all in units of THz, for the case of x - (y -) polarization, respectively. Distinct from Figure 4, now the RMSE between the CMT spectra and the FDTD ones (see Figure 5C and F) are always very small independent of d and polarization. To fully understand how the CMT spectra evolve as d varies, we must know how the three coupling parameters ($\kappa, \kappa_{pp}, \kappa_{qq}$) depend on d , as in the CMT framework [e.g. Eqs. (2)–(5)] *only* these three coupling parameters are varying against d . As shown in Figure 5B and E, in both polarizations, the inter-mode coupling κ are obviously stronger than the on-site corrections κ_{qq}, κ_{pp} , and are an increasing function of d , which are very reasonable as the inter-mode coupling must be stronger in the case of larger symmetry-breaking. The CMT can be easily extended to more complicated systems possessing more resonant modes (see Section 4 of the Supplementary Material).

To understand why Fano's formula fails in certain cases, we now derive Fano's formula out from the more general framework (e.g. the two-mode CMT) under certain approximations. Such a derivation can reveal the inherent correction between Fano's formula and the CMT, and in particular, can help us understand when and why Fano's formula fails in certain cases. As Fano's formula deals with *weak-coupling* with a *passive* background [52], we first introduce a dimensionless parameter K

$$K = \left| \frac{\tilde{X}}{\tilde{f}_+ - \tilde{f}_-} \right| = \left| \frac{1}{4} \frac{\kappa \gamma_p}{\Delta f^2 + \kappa^2} \right| \quad (6)$$

to quantitatively characterize the strength of $\tilde{X}$. We call K as the *effective* far-field coupling. It is a total effect of frequency detuning Δf and the near-field inter-mode coupling κ . In the case of $K \leq 0.25$, we assume that $\tilde{X}$ is weak enough and can be safely ignored. Then, we can derive from Eq. (5), the following approximate expressions of reflection and transmission coefficients:

$$\begin{aligned} r &\approx r_{Qz} - \frac{2}{\eta^2 + 1} \left(\frac{\tilde{\gamma}_+}{\tilde{W}_+} + \frac{\tilde{\gamma}_-}{\tilde{W}_-} \right), \\ t &\approx t_{Qz} - \frac{2\eta}{\eta^2 + 1} \left(\frac{\tilde{\gamma}_+}{\tilde{W}_+} + \frac{\tilde{\gamma}_-}{\tilde{W}_-} \right). \end{aligned} \quad (7)$$

⁷ The resonant frequencies of those dark modes were obtained by the searching-eigenmode method in finite element method (FEM) simulations. Here, we excited the nano-patch arrays with evanescent waves at different frequencies, and seek for those frequencies at which the responses of the systems diverge. Such frequencies are defined as the eigen-frequencies of the dark modes, with associated field patterns being the related eigen-wavefunctions.

Figure 5: CMT modelling of the bright-dark coupled resonators.

(A, D) FDTD-simulated (solid black curves) and CMT-fitted (red dotted curves) transmission spectra for samples with different values of d , shined by normally incident light with two different polarizations. (B, E) Retrieved CMT parameters as functions of d for two different polarizations. (C, F) RMSE of CMT spectra (red circle symbols) with (C) x-polarized and (F) y-polarized incident waves.

Interestingly, we find that Eq. (7) actually represent the independent oscillator model (IOM) widely used in the community [35–38].

We next study under what condition the bright mode can be considered as a *passive* background, meaning that its contribution to the transmission remains nearly unchanged after the dark mode is added (i.e. $t_0 = t_{Qz} - \eta_t \tilde{\gamma}_+ / \tilde{W}_+ \approx t_{Qz} - \eta_t \gamma_p / W_p$). Obviously, we require that the changes in the center frequency and bandwidth of the bright mode are both negligible, yielding the following criterions:

$$\begin{aligned} \Delta\Gamma_b &= \tilde{\gamma}_+ - \gamma_p = \gamma_p (\Delta f - \sqrt{\Delta f^2 + \kappa^2}) / 2\sqrt{\Delta f^2 + \kappa^2} \ll \gamma_p, \\ \Delta F_b &= \tilde{f}_+ - f_p = \tilde{f} - \tilde{\Delta f} - f_p \approx \Delta f - \sqrt{\Delta f^2 + \kappa^2} \ll \gamma_p. \end{aligned} \quad (8)$$

Assuming that Eq. (8) can be satisfied, we expand Eq. (5) to the Taylor series and then drop the high-order terms (setting $\kappa_{pp} = 0$ as the on-side terms are not important), and finally arrive at the Fano-like formula – Eq. (1), in which the involved Fano parameters are expressed in terms of the CMT parameters as:

$$\begin{cases} t_0 = t_{0z} - 2\eta\tilde{\gamma}_+ / ((1+\eta^2)\tilde{W}_+) \\ \varepsilon = (f - \tilde{f}_- + i\tilde{\gamma}_-) / \tilde{\gamma}_- \\ \alpha = i(1 - 2\eta / ((1+\eta^2)t_0)) \end{cases} \quad (9)$$

Detailed derivations of Eq. (9) can be found in Section 5 of the Supplementary Material. Equation (9) clearly reveals the physical meanings of all Fano parameters in terms of the CMT parameters (see CMT-calculated Fano parameters in Section 6 of the Supplementary Material). Specifically, the background transmission t_0 is nothing but the transmission contributed by the bright mode, while ε describes the normalized frequency detuning of the dark mode, and α is a parameter dictated by the background properties including the asymmetry caused by the substrate.

Equations (6) and (8) reveal that K , $\Delta\Gamma_b$ and ΔF_b are three key parameters to judge whether the 2-mode CMT formula can be rewritten as the Fano's formula. Figure 6A–D depict how these parameters vary against d for two incident polarizations. Clearly, for the x -polarization, these parameters are always small enough to ensure that criterions [Eqs. (6) and (8)] can be well satisfied, which explains why Fano's formula works well (see Figure 6A and C). In contrast, for the y -polarization, the absolute values of these parameters increase significantly as $d > 20$ nm making criterions [Eqs. (6) and (8)] no longer well satisfied, leading to the failure of Fano's formula.

We can then establish a clear physical picture to understand the intriguing effects discovered in this work

with the help of Eq. (6). Although the direct inter-mode coupling κ takes *similar* values for two different polarizations, the frequency detuning Δf are very different in two cases, which makes the *effective* far-field coupling K behaving very differently. Specifically, while Δf is much larger than κ in the x -polarization case, the same is not true for the y -polarization case. As a result, the *effective* far-field coupling K in the y -polarization case is much stronger than that in the x -polarization case, which finally results in the failure of Fano's formula in the y -polarization cases.

4 Generic phase diagram derived from CMT

Now that the 2-mode CMT has been proven as a more general theoretical tool to study such a dark-bright coupled photonic system, we will employ it to derive a phase diagram for such systems, in order to illustrate the rich physics caused by the interplays between different parameters. In particular, such a phase diagram can reveal in which regions those simplified models derived previously are justified, which is important to help researchers to correctly use these models in further studies.

Previous analyses have shown that K and ΔF_b [see Eqs. (6) and (8)] are two crucial parameters to dictate the applicability of Fano's formula. Figure 7A contains a color map representing how $\Delta F_b/\gamma_p$ vary against $\Delta f/\gamma_p$ and κ/γ_p ,

Figure 6: Analyses on two CMT parameters related to the validity of Fano's formula.

The effective far-field inter-mode interaction K (A, B), changes of the radiation loss $\Delta\Gamma_b$ and frequency ΔF_b of the bright mode (C, D), calculated with the retrieved CMT as shown in Figure 5, for samples with different values of d and under excitations of normally incident lights with different polarizations. The shaded area denotes the region where Eq. (1) is no longer held.

Figure 7: A generic phase diagram for the dark-bright coupled plasmonic systems.

(A) Normalized frequency shift of the bright mode $\Delta F_b/\gamma_p$ (color map) versus $\Delta f/\gamma_p$ and κ/γ_p . The whole space is divided into 4 parts by the white line ($\Delta F_b/\gamma_p = 1$), the black line ($\Delta F_b/\gamma_p = 0.1$) and the red line ($K = 0.25$). Green squares and red stars denote the positions of our samples for the cases of x- and y-polarizations. (B–E) Transmittance and reflectance spectra of four typical cases represented by the orange/green circles located at four different phase regions, calculated by the CMT, Fano's formula, and the IOM, respectively.

with a red line corresponding to the boundary defined by $K = 0.25$. Clearly, such a boundary line separates the whole phase space into two sub-regions – the white area (corresponding to $K > 0.25$) where the far-field inter-mode coupling $\tilde{X}$ plays very important role and the rest (with $K < 0.25$) where $\tilde{X}$ can be safely dropped. Outside the white area, the effective far-field inter-mode coupling K is so weak that the two “dressed” modes are essentially decoupled. Therefore, the IOM is well justified in this region. In contrast, inside the white area which is termed as the “critical” region, the effective far-field coupling between two modes are so strong that the IOM is no longer a good model and one has to use the original two-mode CMT to fully characterize the optical properties of the system.

In the region where the IOM works, we can further divide the phase space into three sub-regions separated by two additional phase boundaries, defined by $\Delta F_b/\gamma_p = 0.1$ (black line) and $\Delta F_b/\gamma_p = 1$ (white line), respectively. Based on the analyses given in last section, we understand that in the sub-region below the black line, the bright-mode, even after interacting with the dark mode, still remains nearly unchanged in resonant frequency, indicating that Fano's formula must work well. Meanwhile, in the sub-region above the white line, the bright mode must be strongly hybridized with the dark one. In particular, as the “dressed” bright mode exhibits a frequency shift larger than γ_p , we call this region as a “Rabi” region

[7–10]. In fact, even the “dressed” bright/dark modes can still be well treated as two independent oscillators in this region, their essential properties are completely different from those of the two original modes. Finally, the sub-region bounded by these two lines is called an “intermediate” region where the two “dressed” modes can be modeled separately but with non-negligible and moderate hybridizations.

We now choose a few representative examples to vividly illustrate the essential features of four different sub-regions defined in Figure 7A. Figure 7B–E plot, respectively, the CMT-calculated transmission/reflection spectra of four typical systems, corresponding to four orange circles located at different regions in Figure 7A. Obviously, for three samples labeled by “B”, “C” and “D”, the CMT-calculated spectra (solid lines) can always be well modeled by the IOM (open circles). On the contrary, the CMT-calculated spectra for sample “E” can never be well described by the IOM which can even exhibit unphysical reflection amplitudes exceeding 100%. We used Fano's formula further to fit these CMT calculated spectra, and depicted the best-fitting Fano lines in the same figures with dashed lines. Clearly, the Fano lines can only well describe the CMT spectra of sample “B”, among all four samples studied. These quantitative comparisons have reinforced our notions of four different phase spaces discussed in last paragraph.

Finally, we note that such a generic phase diagram can help us understand the intriguingly different behaviors of our own experimental results for two different polarizations. The green squares and red stars represent the positions of those samples with different d in our phase diagram, for the cases of x - and y -polarizations, respectively. Obviously, for the case of x -polarization, all samples are well located inside the Fano-applicable region as d increases, which explains why Fano's formula can well describe the optical properties of such a series of samples. In contrast, for the case of y -polarization, increasing d can drive our sample to transit from the Fano-applicable region to the critical region, resulting in the failure of Fano's formula as shown in Figure 7A. Moreover, we emphasize that both κ and Δf (not just κ) play important roles in determining the position of a system in the phase diagram. Only after considering these two parameters simultaneously, one can choose the correct formula to explain the related experimental/numerical findings, and even design devices exhibiting the desired spectrum line shapes based on our phase diagram.

5 Conclusions

In summary, based on experimental and numerical results on a series of symmetry-broken nano-plasmonic resonators, we found that Fano's formula works well for one incident polarization but fails completely for another polarization. This motivated us to re-examine the same problems employing the two-mode CMT, which explains all numerical/experimental results well. We then establish a generic phase diagram for such dark-bright-coupled systems based on the CMT, in which the simplified models previously developed (including Fano's formula and the independent-oscillator-model) are found to be valid only in specific phase regions. Our findings not only provide deeper physical understandings on such complex systems, but more importantly, identify the validity regions for those simplified models, which can help researchers choose correct models in their future studies.

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