Analytic Theory of the Resonance Properties of Metallic Nanoslit Arrays

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Abstract—By applying formulation based on time reversibility, we provide the analytic theory of resonance properties of metallic nanoslit arrays. We model lossy resonant systems in which a resonance is induced by a single quasi-bound mode (QBM). It is consistent with the Fano resonance theory of quantum interference of auto-ionizing atoms and captures the essential characteristics of dissipative resonant systems. We show that time-reversibility requirements lead to analytical solutions for the resonant transmission and the associated nonreciprocal absorption in terms of a minimal number of independent basic parameters that include partial decay probabilities of resonant pathways and the non-resonant transmission amplitude. With a clear view of interfering electromagnetic field configurations and the associated absorbing processes, the theory reveals the essential physics of resonant optical transmission. In particular, the enhanced transmission peak is given by the product of partial decay probabilities and is independent of the non-resonant light wave amplitude. In a highly asymmetric coupling regime, the excitation of the QBM leads to anti-resonant extinction of the transmission, indicating a negative role of the QBM. The parity of the QBM determines occurrence of red or blue tails in the spectral profile. Absorbance measurements yield direct determination of the partial decay probabilities by which the main features of the resonant transmission are quantitatively explained. Thus, these basic parameters can be directly established experimentally. Full numerical calculations of the transmission spectra are in complete quantitative agreement with our analytical formulation for optical transmission mediated by both slit cavity modes and plasmonic modes.

Index Terms—Analytical models, nanostructured materials, periodic structures, resonance light scattering.

I. INTRODUCTION

NANOPIATTENDED surfaces with subwavelength periodicity are of immense interest for various potential applications. In particular, periodic metallic films support resonant energy localization and associated extraordinary optical transmission (EOT) [1], [2]. The resonance mechanisms behind EOT have been widely discussed in the literature and generally have been found to correspond to surface-plasmon polaritons (SPP) and Fabry-Pérot cavity modes [3]. We recently provided normalized parametric maps for an example idealized metal film with narrow slits, rendering clearly the regimes of dominance for these mechanisms [4].

In this paper, we treat the resonance interactions in detail, formulating their properties in terms of basic physical parameters. We model a nanostructured metal film as a dissipative quasi-bound resonator where the resonant (indirect) and nonresonant (direct) scattering pathways interfere. We apply the phenomenological coupled-mode theory due to Haus [5] to obtain the attendant transmission, reflection, and absorption spectra. For a resonance interaction that can derive from SPP or cavity mode, we find an analytic expression for the transmission spectrum in terms of the non-resonant transmission field amplitude and the radiation yields of the localized mode expressed in terms of relevant decay probabilities. We present numerical results obtained by rigorous coupled-wave analysis (RCWA) that show excellent agreement with the analytic theory.

The interference picture of EOT, i.e., Fano-type interpretation, has been accepted as a general explanation of the characteristic asymmetric spectral profile and red-shifted transmission peak [6], [7]. In the absence of an available theory that analytically decomposes the two interfering electromagnetic (EM) field configurations, numerical fitting typically applies this picture to experimental or simulated spectra [6], [8]. As the detailed information is implicitly held in the fitted phenomenological parameters, the interference picture associated with the formal quantum Hamiltonian mapping does not describe any further physics involved in the EM field interaction. Hence, to ameliorate this, we provide an analytic theory that clearly defines the two interfering EM field configurations in terms of basic parameters such as the non-resonant scattering amplitudes and partial radiation yields. We show that the basic parameters can be experimentally determined by absorbance measurements. The essential features of EOT, such as the enhanced maximum and anti-resonant extinction, are quantitatively described by a simple analytic formula.

II. FANO PROFILE AND TIME REVERSIBILITY

As discussed by Genet et al. [6], EOT can be explained as a resonant light scattering process where Fano interference takes place between two transmission pathways: the resonant pathway mediated by a discrete state, e.g., SPP [1], [9] or cavity mode [2], and the non-resonant pathway via a
continuum, analogous to Bethe’s contribution for hole arrays [10]. According to the Fano resonance interpretation of EOT, the transmitted intensity is given by an expression similar to that originally found for the quantum interference of an auto-ionizing atom as [6], [11]
\[
T(\delta) = |t_D|^2 \frac{(q + \delta)^2}{1 + \delta^2},
\]
where the reduced frequency is \( \delta = (\omega - \omega_0)/\gamma_t \) for a discrete state at \( \omega_0 + i \gamma_t \) and \( t_D \) is a transition probability amplitude via the continuum. The shape factor \( q \) describes resonant interference features such that the resonant enhancement is to that originally found for the quantum interference of an electron-scattering process using this quasi-reversibility argument [15]. In this paper, we prove that Eq. (2) correlates the phase difference and amplitude ratio of the EM fields pertaining to the two interfering pathways. The interference picture of EOT on this basis analytically decomposes the field configurations associated with the two interfering pathways in dissipative material systems, thereby yielding clear, improved Fano-type interpretation of this resonance effect.

III. ANALYTIC THEORY OF EOT BASED ON QUASI-REVERSIBILITY ARGUMENTS

We use a coupled-mode model schematically shown in Fig. 1(a). A resonant transfer of light involving a discrete state (blue arrows with coupling rates \( \kappa_1 \) and \( \kappa_2 \)) occurs on the coupling of an incident wave to a quasi-bound mode (QBM) resonant at \( \omega_0 + i \gamma_t \) and its re-emission to both sides of the film. Here, \( t_D \) and \( t_1 \) represent direct (non-resonant) transmission and reflection amplitudes of the continuum input (red arrows), respectively. This coupled-mode interaction is framed as a two-port resonator system shown in Fig. 1(b) where the QBM couples to two radiation ports and dissipates its energy to material losses via a non-radiative decay process (green arrows with decay rate \( \gamma_{nr} \)). As shown in APPENDIX A, in a slowly decaying regime where \( \gamma_{tot} << \omega_0 \), applying coupled-mode theory [5], [14], [16], the transmission coefficient is
\[
T(\delta) = t_D + \frac{2 \sqrt{|t_D|^2 |t_R|^2} e^{i\phi}}{1 + i\delta} \equiv t_D + t_R(\delta),
\]
where \( \phi = \arg(\kappa_1) + \arg(\kappa_2) \) is the resonant transfer phase at \( \delta = 0 \), \( \eta_n = |\kappa_n|^2/(2\gamma_{tot}) \) is the partial decay probability of the QBM to port \( n \), and \( \gamma_{tot} = |\kappa_1|^2/2 + |\kappa_2|^2/2 + \gamma_{nr} \) is the total decay rate. The decay probabilities \( \eta_n \) associated with the decay rates \( \gamma \) satisfy \( \eta_1 + \eta_2 + \eta_{nr} = 1 \) with \( \gamma_{nr} = \eta_{nr}\gamma_{tot} \) and \( \eta_n = \eta_{tot} \gamma_{tot} \). Fano-type interference between the non-resonant \( t_D \) and the resonant \( t_R(\delta) \) transfer amplitudes is explicit in Eq. (3). The transmitted intensity \( T = |T|^2 = |t_D|^2 + |t_R|^2 + 2Re(t_D \cdot t_R^*) \) becomes fully deterministic with minimal parameters of \( |t_D| \) and \( \eta_n \) once the phase difference between \( t_D \) and \( t_R \) is known.

As shown in APPENDIX B, there are two main consequences of Eq. (2). First, the amplitude ratio \( |t_D/t_R| \) and phase difference \( \Delta = \arg(t_D) - \arg(t_R) \) are related by
\[
\cos \Delta = -\eta_{rad} \left| t_D/\sqrt{t_R} \right|, \quad \sin \Delta = p \sqrt{1 - \cos^2 \Delta},
\]
where \( \eta_{rad} = \eta_1 + \eta_2 \) is the total radiation yield and \( p \) is the parity of the QMB assigned \( p = +1 \) to an even-like mode.
and $p = -1$ to an odd-like mode. Second, the nonreciprocal absorbance for light entering port $n$ is expressible as

$$A_n(\delta) = \frac{4\eta_1 \eta_{nr}}{1+\delta^2},$$

when $\eta_1 \neq \eta_2$, where the probability of absorption is $\eta_{nr} = 1-\eta_{rad} = 1-\eta_1 - \eta_2$.

Eqs. (3)–(5) reveal the essential physics of Fano interference in EOT and connect well with the paper by Genet et al. [6]. For lossless resonance systems, Eq. (3) reduces to Eq. (1) with the shape factor $q$ now expressed as

$$q = p \sqrt{4\eta_1 \eta_2 / |D_0|^2 - 1}. \quad (6)$$

The following properties emerge clearly: (i) the QBM’s parity $p$ determines whether a blue tail ($q > 0$) or a red tail ($q < 0$) is observed in the spectrum [6], [11]; (ii) the enhanced transmission maximum is $T_{max} = |D_0|^2 (q^2 + 1) = 4\eta_1 \eta_2$, independent of the non-resonant expectation $|D_0|^2$, e.g., standard aperture theory by Bethe [10]; (iii) a symmetric enhancement peak arises for $q = \pm \infty$, which is obtained when $D_0 = 0$, showing that nonzero $D_0$ causes the spectral asymmetry; (iv) a balance between the resonant and non-resonant contributions such that $4\eta_1 \eta_2 \approx |D_0|^2$ results in a symmetric extinction dip with $q = 0$, i.e., the QBM plays a negative role [17]. Figs. 1(c) and 1(d) illustrate the arguments (i)–(iv); these are computed by using Eqs. (3) and (4).

Representing Eq. (3) in the complex plane provides an intuitive understanding of the resonant interference. In Fig. 2(a), $\tau(\delta)$ given by Eq. (3) is a circle with radius $(\eta_1 \eta_2)^{1/2}$, its far-off-resonance end $\tau(\infty) (= \tau_\infty)$ lifted by $D_0$, and its axis connecting $\tau_\infty$ and $\tau(0) (= \tau_0)$ inclined counter-clockwise by $\Delta$ from the axis parallel to $D_0$. We refer to this circle as a resonance response circle (RRC). With the RRC representation of $\tau(\delta)$ in Fig. 2(a), the transmission maximum at $\delta = 1/q$ and minimum at $\delta = -q$ are geometrically determined by $\tau_{max}$ and $\tau_{min}$ at which the green line connecting the center of the RRC and the origin crosses the RRC.

In lossless cases, the amplitude-phase correlation in Eq. (4) is clearly visualized in the RRC as a geometric connection between its diameter and inclination. The phase difference between the non-resonant and resonant contributions $\Delta$ is determined so that the RRC always passes through the origin. Fig. 2(b) with corresponding spectra in Fig. 1(c) shows how amplitude-phase correlation affects the RRC and the $T$ spectrum. For a lossless system, anti-resonant null transmission is inevitable, i.e., $T_{min} = |T_{min}|^2 = 0$, and $T_{max}$ must be a square of RRC’s diameter, i.e., $T_{max} = |T_{max}|^2 = 4\eta_1 \eta_2$. We note that $\eta_1 = \eta_2 = 0.5$ is necessary for $T_{max} = 1$. It means that a lossless symmetric structure, e.g., a free-standing film in a resonant coupling regime, must exhibit a full transmission peak [2], [18], [19]; complete transmission is also obtainable even in an asymmetric system such as glass-supported films with conical holes or trapezoidal slits that balance the radiation yields of an SPP to each side of the film.

Increasing loss ($\gamma_{nr}$) without affecting $\kappa_n$ results in reduced RRC radius, $(\eta_1 \eta_2)^{1/2} = |\kappa_1 \kappa_2| / (|\kappa_1|^2 + |\kappa_2|^2 + 2\gamma_{nr})^{1/2}$, while $\Delta$ remains constant as shown in Fig. 2(c). Thus, the material loss suppresses $T_{max}$ while $T_{min}$ increases from zero because the RRC does not pass through the origin anymore. Note that Eq. (1) is not strictly valid with material losses. In particular, loss appreciably degrades $T_{max}$, but the destructive interference effect at $T_{min}$ is persistent for the low $|D_0|$ regime with $T_{min} \approx \eta_{nr}^2 |D_0|^4 / T_{max}$, which is far below the spectral background level $|D_0|^2$.

In dissipative films, even small loss coefficients may yield strong absorption when the system becomes resonant due to excitation of a QBM. Thus, absorption plays an important role in EOT [14], [20]. A main consequence of resonant absorption is nonreciprocity in the absorbance and reflectance for an asymmetric system [20], e.g., glass-supported films. From Eq. (5), the on-resonance (peak) absorbance is $A_n(0) = 4\eta_1 \eta_{nr}$, Thus, quantitative nonreciprocity is expressible as $|A_1(0)-A_2(0)| \approx 4\eta_{nr} |\eta_1-\eta_2|$. Nonreciprocity in $R_n$ must be identical to that in $A_n$ because reflectance $R_n = 1-T-A_n$ and $T$ is reciprocal in principle. The nonreciprocity measures the
difference in the QBM decay probability coefficients (radiation yields) $\eta_r$ and, therefore, directly relates to $T_{\text{max}}$, which takes small values under highly asymmetric $\eta_r$ as seen in Fig. 1(c).

Based on Eq. (5), the lossless limit of $T_{\text{max}}$ can be expressed in terms of $A_n$ as

$$
limit_{\eta_r \to 0} T_{\text{max}} = M = 4\gamma_1\gamma_2/\gamma_{\text{rad}}^2 = 4A_1A_2/(A_1 + A_2)^2. \quad (7)
$$

Thus $T_{\text{max}}$ for the lossless case is extracted from the lossy array. Equation (7) states that even for a lossless metal, $T_{\text{max}}$ is generally not unity.

IV. SURFACE-PLASMONIC EOT

We quantitatively check our theory with full numerical calculations [21]. The film is modeled as a Drude metal with collision and plasma frequency, $\Gamma = 4.52f_{\text{ur}} \times 10^{13}$ rad/s and $\omega_p = 7.54 \times 10^{15}$ rad/s, respectively, where $f_{\text{ur}}$ is an absorption factor. Frequency-dependent dielectric constant of the metal is given by $\varepsilon_{\text{metal}}(\omega) = 1 - \omega_p^2/(\omega^2 + i\Gamma\omega)^{-1}$. The absorption factor $f_{\text{ur}}$ adjusts the collision frequency of free electrons in the Drude metal such that $\gamma_{\text{nr}}$ of a mode supported by a metallic system is directly proportional to it [23]. Surface normal incidence is assumed.

For an optically thin film with slit cavity length short enough to thwart cavity mode (CM) resonance, an SPP at either interface of the film can operate as a dominant transmission-enhancing QBM [4]. For a thin metal film with thickness comparable to the skin depth, the metal film allows non-resonant transmission directly through it. Thus, Fano-type interference between the resonant and non-resonant pathways leads to a transmission spectrum highly asymmetric exhibiting an enhanced transmission peak and an anti-resonant extinction dip.

Fig. 3 shows numerical results for an optically thin nanoslit array with thickness $d = 40$ nm, period $\Lambda = 400$ nm, and slit width $w = 20$ nm. The resonance feature at $0.72 \mu m$ is attributed to the excitation of an SPP on the glass/metal interface. It is confirmed in the inset of Fig. 3(b) showing typical SPP character in the field near the glass/metal interface. The transmission spectra in Fig. 3(a) and a logarithmic replot in Fig. 3(b) clearly exhibit asymmetric profiles with $T_{\text{max}}$ increasing with increasing $f_{\text{ur}}$. In contrast to $T_{\text{max}}$, the anti-resonant dips ($T_{\text{min}}$) increase with $f_{\text{ur}}$. The effect of material absorption on $T_{\text{max}}$ and $T_{\text{min}}$ qualitatively agrees with the analytic theory description. Numerical absorption spectra are shown in Figs. 3(c) and (d).

To quantitatively verify the analytic theory, we extract from the computed absorbance spectra in Fig. 3 the basic resonance parameters ($f_{\text{tr}}, \eta_{\text{nr}},$ and $\eta_{\text{rad}}$) and compare the analytical and numerical spectral profiles. We first extract $f_{\text{tr}}$ by measuring the half-width at half-maximum (HWHM) of the absorbance peaks. HWHM ($\gamma_{\text{tot}}$) measured from the simulated absorbance spectra ($A_n$) in Figs. 3(c) and 3(d) is indicated in Fig. 4(a). It confirms that $\gamma_{\text{tot}}$ increases linearly with $f_{\text{nr}}$, implying $\gamma_{\text{nr}} \propto f_{\text{nr}}$. HWHM linearly extrapolated to $f_{\text{nr}} = 0$ is identified as $\gamma_{\text{rad}}$ and, then, we find $\eta_{\text{rad}} = \gamma_{\text{rad}}/\gamma_{\text{tot}}$. This method is valid for
most noble metals satisfying $|\text{Re}(e_{\text{metal}})| > |\text{Im}(e_{\text{metal}})|$ [23]. We estimate partial radiation yield $\eta_n$ by using

$$\eta_n = \eta_{\text{rad}} A_n / (A_1 + A_2).$$

(8)

This relation is found straightforwardly from Eq. (5). Except for $|t_0|^2$, all basic parameters for the analytic theory are obtained from the absorption spectra. We obtain the non-resonant transmittance $|t_0|^2$ by interpolating (4th-order polynomial) fit from the RCWA calculated transmission spectrum for far-off resonant wavelength ranges. The interpolated $|t_0|^2$ is indicated by the red dashed curve in Fig. 4(b) and shown as black curves in Figs. 3(a) and 3(b).

The analytic theory curves in Figs. 3(a) and 3(b) are obtained by using partial radiation yields ($\eta_{\text{rad}}$ and $\eta_n$) purely estimated from the absorbance spectra and $|t_0|^2$ interpolated from the off-resonant transmittance. Thus we confirm excellent quantitative agreement between the analytic theory and RCWA calculations in describing $T_{\text{max}}$, $T_{\text{min}}$, spectral profile, and their dependence on the material absorption.

V. CAVITY-MODAL EOT

For an optically thick metal film, resonant CMs inside the nanoslits [2]-[4], [13], [17] correspond to the QBM in our model. Fig. 5 shows transmission and absorption spectra calculated by RCWA for a nanoslit array with thickness $d = 550$ nm, period $\Lambda = 400$ nm, and slit width $w = 20$ nm. Three resonant features at wavelengths 0.779, 0.978, and 1.395 $\mu$m are due to Fabry-Pérot (FP) resonances of the cavity modes inside the nanoslits as confirmed by the associated field distributions in Fig. 6. We label each mode as CM with $l$ indicating the number of nodes inside the cavity.

Qualitative agreement with the analytic theory is observed from the transmission and nonreciprocal absorption [20], [22] spectra in Fig. 5. For CM1 with $A_1 \approx A_2$, implying $\eta_1 \approx \eta_2$ according to our analytic theory interpretation, the peak transmittance $T_{\text{max}}$ is largest (0.934 for $f_{\text{ut}} = 0.1$) while $T_{\text{max}}$ for CM3 with $A_1 >> A_2$, implying $\eta_1 >> \eta_2$, is much smaller (0.107 for $f_{\text{ut}} = 0.1$). One might prefer high absorption of CM3 in Fig. 5(b) as the main reason for its low $T_{\text{max}}$. This interpretation, however, cannot explain the small transmission peak of CM3 in case of light incidence from glass substrate in Fig. 5(c) where CM3 exhibits much smaller absorption than CM1.

The large difference between $\eta_1$ and $\eta_2$ for CM3 is due to its spectral proximity to the SPP resonance wavelength at the glass/metal interface. Note excitation of surface field at the glass/metal interface in Fig. 6(c). In our case, the glass/metal interface becomes resonant due to the excitation of SPP at 0.721 $\mu$m (given by $\Delta f_{\text{glass}} e_{\text{metal}} / (\text{c}_{\text{glass}} + \text{c}_{\text{metal}})$), where $\varepsilon$ represents dielectric constant of the material indicated by its subscript. It is known in the literature [3] that the internal reflectance of a CM tends to 100% near the SPP resonance wavelength, suppressing coupling of the CM to the outside radiation. Thus, out-coupling of CM3 to glass substrate must be much smaller than that to air side, i.e., $\eta_1 >> \eta_2$.

We again apply the same analysis method as in Sec. IV to quantitatively check the validity of the analytic theory to a cavity modal EOT. Decay rates $\gamma_{\text{rad}}$ for CM1~CM3 are obtained by linearly extrapolating HWHM of the absorbance peaks in Figs. 5(b) and 5(c). The result is presented in Fig. 7(a).

For an optically thick metal film, $t_0$ is reasonably close to zero because there is no available transmission pathway other than the slit cavity. In this case, the analytic theory yields

$$T_{\text{max}} = 4\eta_1\eta_2 = M^2 \eta_{\text{rad}}^2$$

(9)

where $M$ can be given in terms of peak absorbances according to Eq. (7). We obtain $M = 0.9995$ for CM1, 0.912 for CM2, and 0.184 for CM3 from the peak absorbance in
Fig. 7. (a) Total decay rate $\gamma_{\text{tot}}$ (HWHM) versus absorption factor $f_{\text{nr}}$ for CM1 (red squares), CM2 (green circles), and CM3 (blue triangles). $\gamma_{\text{rad}}$ of each mode is inferred by linearly extrapolating $\gamma_{\text{tot}}$ to $f_{\text{nr}} = 0$. (b) Peak transmittance $T_{\text{max}}$ versus radiative decay probability $\eta_{\text{rad}}$, where symbols represent RCWA calculation results while solid curves represent Eq. (9) due to the analytic theory. Note that $M$ is the value of $T_{\text{max}}$ extrapolated to $\eta_{\text{rad}} = 1$ (corresponding to $\eta_{\text{nr}} = 0$).

Figs. 5(b) and 5(c). Fig. 5(b) shows excellent agreement in $T_{\text{max}}$ separately obtained by RCWA (symbols) and Eq. (9) (curves). Note that $M$ and $\eta_{\text{rad}}$ for $T_{\text{max}}$ in Eq. (9) are purely obtained from the absorption spectra.

VI. DECOMPOSITION OF TOTAL FIELD INTO RESONANT AND NON-RESONANT CONFIGURATIONS

The EM field is readily decomposed into configurations associated with two interfering pathways in this theory. For port 1 (air-side) incidence, the total magnetic field can be written as $H_{\text{tot}} = h_{1}-\rho h_{1}^{*}+h_{2}^{*}+H_{\text{QBM}}$, where $H_{\text{QBM}}$ is the field associated with the QBM and $h_{2}$ is the normalized field for incoming light at port $n$; thus, $-h_{2}^{*}$ represents the normalized field for outgoing light at port $n$. We define two orthogonal projection operators $Q$ and $P$ that respectively project a field configuration onto the resonant and non-resonant subspace of a Hilbert space formed by the normalized field solutions of Maxwell’s equations. The coupled-mode pictures in Figs. 1(a) and 1(b) derive the resonant configuration $H_{\text{R}} = Q H_{\text{tot}} = a(k_{1}^{*} h_{1}+k_{2}^{*} h_{2}+\sigma k_{1}^{*} h_{1}^{*}-\sigma k_{2}^{*} h_{2}^{*})+H_{\text{QBM}}$, and non-resonant configuration $H_{\text{D}} = P H_{\text{tot}} = H_{\text{tot}}-H_{\text{R}}$, where $a = k_{1}/(2 \gamma_{\text{rad}})$ and $\sigma = (\eta_{\text{rad}}-\eta_{\text{nr}}+i \delta)/(1-i \delta)$ (see APPENDIX C). Note that $H_{\text{R}}$ completely describes $H_{\text{QBM}}$ in $H_{\text{tot}}$ and can be obtained by two coherent wave incidences with amplitudes $a k_{1}^{*}$ from port 1 and $a k_{2}^{*}$ from port 2. On the other hand, if we have incident waves associated with $H_{\text{D}}$, the QBM is not excited. Fig. 8 presents the RCWA simulation results for these two situations and confirms the validity of the field decomposition based on our theory.

VII. CONCLUSION

In summary, we developed a quasi-reversible Fano interference theory of the resonance properties of metallic nanoslit arrays that is consistent with the original Fano resonance theory on quantum interference of auto-ionizing atoms. Our formulation captures the essential characteristics of dissipative resonant systems such as the metallic nanoslit arrays treated in this paper. We show that the quasi-reversibility requirement derives the analytic description of the resonant transmission and associated nonreciprocal absorption in terms of a minimal number of independent basic parameters, i.e., partial decay probabilities ($\eta_{1}$ and $\eta_{2}$) and non-resonant transmission amplitude ($\eta_{0}$). With clear definitions of two interfering electromagnetic field configurations and the associated absorbing processes, the theory reveals the essential physics of resonant interference in the EOT phenomenon: (i) the enhanced transmission peak is given by the product of partial decay probabilities (4$\eta_{1}\eta_{2}$), independent of the non-resonant expectation $|\eta_{0}|^{2}$; (ii) in a highly asymmetric coupling regime where $4\eta_{1}\eta_{2} \approx |\eta_{0}|^{2} << 1$, excitation of the QBM leads to an anti-resonant extinction of the transmission indicating a negative role of the QBM; (iii) parity of the QBM determines occurrence of red or blue tails in the spectral profile; (iv) absorbance measurements yield direct determination of the partial decay probabilities by which main features of the resonant transmission are quantitatively explained. Thus, these basic parameters can be directly established experimentally.

Further extension of our theory to a multiple-QBM system may offer concomitant analytic descriptions pertinent to other resonant nanophotonic systems. In guided-mode resonance devices or nanoplasmonic templates, the coupling between localized modes provides a variety of useful spectra including filters [24] and polarizers [25] or a plasmon-induced transparency [26]. We believe that the quasi-reversible interference picture can be applied to such systems as well as others that exhibit general aspects of QBM resonances to further elucidate their essential physics.

Fig. 8. Decomposition of the total field into resonant ($H_{\text{R}}$) and non-resonant ($H_{\text{D}}$) configurations for CM2 in Fig. 5. The total field $H_{\text{tot}}$ is a superposition of the following: (left panel) $H_{\text{R}}$ that completely describes the field associated with the QBM in $H_{\text{tot}}$, (right panel) $H_{\text{D}}$ that does not excite the QBM (cavity mode in this case). Note arrows and their amplitudes for coherent combinations of incoming and outgoing waves associated with $H_{\text{D}}$ and $H_{\text{R}}$. 

![Graph](Image_1.png)


**APPENDIX**

**A. Transmission and Reflection Coefficients**

Using coupled-mode theory [5], we model an extraordinary optical transmission (EOT) system as a two-port resonant scattering problem assuming the non-resonant pathway to be lossless and include absorption to cause non-radiative decay of the QBM. This is schematically depicted in Fig. 1(b), where \( \psi_{n=}r(t) \) represents amplitudes of the coupled radiation modes at port \( n \) (+: incoming mode, −: outgoing mode). For a QBM with amplitude \( \Phi(t) \), the modal amplitudes are assumed to describe slowly varying envelopes of corresponding mode fields detailed as follows. The electric (\( E \)) and magnetic (\( H \)) fields associated with the QBM can be expressed as

\[
E_{\text{QBM}}(r, t) = \Phi(t)E_{\text{QBM}}(r)e^{-i\omega t} + (C.C) \quad (A1)
\]

and

\[
H_{\text{QBM}}(r, t) = \Phi(t)H_{\text{QBM}}(r)e^{-i\omega t} + (C.C). \quad (A2)
\]

The fields associated with the coupled radiation mode are then

\[
E_n(r, t) = [\psi_{n=}r(t)E_n(r) + \psi_{n=}r(t)E_n(r)]e^{-i\omega t} + (C.C) \quad (A3)
\]

and

\[
H_n(r, t) = [\psi_{n=}r(t)H_n(r) - \psi_{n=}r(t)H_n(r)]e^{-i\omega t} + (C.C). \quad (A4)
\]

In these equations, \( E_{\text{QBM}}(r) \) and \( H_{\text{QBM}}(r) \) are frequency-domain solutions of Maxwell equations for the QBM while \( E_n(r) \) and \( H_n(r) \) denote the incoming radiation mode at port \( n \). Correspondingly, \( E_n^*(r) \) and \( -H_n^*(r) \) represent the outgoing radiation mode at port \( n \). “(C.C)” refers to the complex conjugate. The modal fields are assumed to be normalized such that

\[
2 \int_V d^3r \left( \frac{1}{2} \frac{d}{d\omega} |E_{\text{QBM}}|^2 + \frac{1}{2} \mu_0 |H_{\text{QBM}}|^2 \right) = 1, \quad (A5)
\]

\[
2 \int_{\partial V} d^2r \cdot \text{Re} \left( E_n \times H_n^* \right) = 1, \quad (A6)
\]

where \( V \) and \( \partial V \) represent a volume containing the QBM and boundary surface of \( V \), respectively. On this basis, we can take |\( \Phi(t) \)| and \( |\psi_{n=}r(t)|^2 - |\psi_{n=}r(t)|^2 \) as time-averaged energy content in the QBM and power incoming to the resonator through port \( n \). Defining a non-radiative decay rate by [27]

\[
\gamma_{nr} = \omega \int_V d^3r e^\nu |E_{\text{QBM}}|^2, \quad (A7)
\]

provides the power absorbed by the QBM as

\[
\int_V d^3r \omega e^{\nu} \langle |E_{\text{QBM}}|^2 \rangle = 2\gamma_{nr} U_{\text{QBM}}(t), \quad (A8)
\]

where \( \varepsilon = \varepsilon' + i\varepsilon'' \) is the dielectric constant of material inside \( V \) and \( \langle \cdot \rangle \), denotes the time average of its inner part.

For a QBM resonant at frequency \( \omega_0 \) in a slowly decaying regime such that \( \gamma_{\text{rad}} + \gamma_{nr} << \omega_0 \), \( \Phi(t) \), \( \psi_{n=}r(t) \), and \( \gamma_{nr} \)

in Eqs. (A1)–(A4), (A7) satisfy coupled-mode equations [5], [14], [16] given as

\[
\frac{d}{dt}\Phi(t) = \left[ i(\omega - \omega_0) - \gamma_{\text{rad}} - \gamma_{nr} \right] \Phi(t) + (\langle |\kappa| \rangle) |\psi_{+}(t)|, \quad (A9)
\]

\[
|\psi_{-}(t)| = C |\psi_{+}(t)| + \Phi(t) |\kappa|, \quad (A10)
\]

where the radiation decay rate is \( \gamma_{\text{rad}} = (|\kappa|)^2 / 2 + 1 / 2 \) and the non-resonant scattering matrix is

\[
C = \begin{bmatrix} r_1 & t_1 \\ t_2 & r_2 \end{bmatrix}. \quad (A11)
\]

Bra-ket notation represents a column vector \( |u\rangle = (u_1 u_2)^T \) with its complex-conjugate transpose \( |u\rangle = (u_1^* u_2^*) \). For stationary excitation in which \( d\Phi dt = 0 \), the coupled-mode Eqs. (A9) and (A10) are reduced to

\[
\Phi = \frac{1}{\gamma_{tot} - 1 - i\delta} \left( C + \frac{1}{\gamma_{tot} - 1 - i\delta} |\kappa|^2 \right) |\psi_{+}\rangle, \quad (A13)
\]

where total decay rate \( \gamma_{tot} = \gamma_{\text{rad}} + \gamma_{nr} \) and reduced frequency \( \delta = (\omega - \omega_0)/\gamma_{tot} \). By applying \( |\psi_{+}\rangle = (0 1)^T \) for port 1 incidence or \( |\psi_{+}\rangle = (1 0)^T \) for port 2 incidence to Eq. (A13), we obtain the transmission and reflection coefficients

\[
\tau(\delta) = \psi_{+} = t_D = \frac{2\eta_h \psi_{+}}{1 - i\delta}, \quad (A14)
\]

\[
\rho_\nu(\delta) = \psi_{-} = r_n + \frac{2\eta_n}{1 - i\delta} e^{i\kappa_n}, \quad (A15)
\]

respectively, where \( n \) denotes the incident port, \( \nu \) indicates the transmission port, e.g., \( \nu = 2 \) for \( n = 1 \), partial decay probability \( \eta_n = |\kappa|^2 / 2\gamma_{tot} \), resonant transmission phase \( \phi = \arg(\kappa_1) + \arg(\kappa_2) \), and resonant reflection phase at port \( n \varphi_n = 2\arg(\kappa_n) \).

**B. Quasi Reversibility, Amplitude-Phase Correlation, and Nonreciprocal absorption**

The source-free frequency-domain Maxwell equations

\[
\nabla \cdot \mathbf{E} = 0, \nabla \cdot \mathbf{H} = 0, \nabla \times \mathbf{E} = i\omega \mu \mathbf{H}, \quad \text{and} \quad \nabla \times \mathbf{H} = -i\omega \varepsilon \mathbf{E}
\]

are invariant under a set of operations that \( \mathbf{E} \rightarrow \mathbf{E}^*, \mathbf{H} \rightarrow -\mathbf{H}^*, \varepsilon \rightarrow \varepsilon^*, \mu \rightarrow \mu^* \) [14], [28]. Note that the set of operations in this case corresponds to not only the time reversal of the field-scattering processes but also to the loss-gain interchange (sign changes in the imaginary parts of \( \varepsilon \) and \( \mu \)). Thus, phase-front propagation as well as power flow change direction, i.e., Re(\( \mathbf{E} \times \mathbf{H}^* \)) → Re(\( \mathbf{E}^* \times -\mathbf{H} \)) = –Re(\( \mathbf{E} \times \mathbf{H}^* \)). Therefore, for time-harmonic fields, if a certain incoming field configuration is scattered to the corresponding outgoing field configuration by an object with \( \varepsilon \) and \( \mu \), the time reversal of the outgoing field configuration should be scattered to the time reversal of the originally incoming field configuration by the object with its loss exchanged with gain in the same amount (e → \( \varepsilon^* \) and \( \mu \rightarrow \mu^* \)). Scattering process by an absorbing object is time reversible under the allowance of loss-gain interchange. However, the loss-gain
interchange at the macroscopic level is physically forbidden by
the second law of thermodynamics because it is inevitable that
the absorbed energy is thermally redistributed into other quanta
in an irreversible way. That is why we use the term “quasi-
reversibility” instead of “reversibility” when considering time
reversal of the scattering process by an absorbing object such
as a metallic structure.

For a general case where \( N \) pairs of incoming and outgoing
waves are coupled by a scattering process, quasi reversibility
can be discussed concisely by using the scattering matrix
\((S\text{-matrix)}\) formalism. Let \( S \) denote the \( S \)-matrix, which relates
the incoming and outgoing modes, \(|\psi_+\rangle\) and \(|\psi_-\rangle\), respectivly,
by the equation \(|\psi_-\rangle = S|\psi_+\rangle\). Quasi-reversibility
requires that time-reversed outgoing waves scatter to the
time-reversed incoming waves. The time-reversed situation
is obtained by taking \(|\psi_+\rangle \rightarrow |\psi_-\rangle^*\) for the incoming
wave configuration, \(|\psi_-\rangle \rightarrow |\psi_+\rangle^*\) for the outgoing wave
configuration, and \(S(e^*) \rightarrow S(e^*)\). This results in the \(S\)-matrix
equation \(|\psi_+\rangle^* = S(e^*)|\psi_-\rangle^*\). By requiring this equation to
be identical to the \(S\)-matrix equation for the original situation
\(|\psi_-\rangle = S(e)|\psi_+\rangle\), we obtain
\[
S^*(e^*) = S^{-1}(e) \quad \text{for quasi-reversibility.} \quad (A16)
\]

Note that reciprocity requires the \(S\)-matrix to be symmetric;
thus, the quasi-reversibility condition of Eq. (A16) is independ-
ent of the reciprocity. Also, we note that Eq. (A16) reduces to
the well-known condition of reversibility in lossless cases,
i.e., \(S^* = S^{-1}\), as \(\text{Im}(e)\) approaches zero [5]. Consequently,
Eqs. (A16) can be considered an extension of reversibility in
lossless systems to absorbing systems.

Using Eq. (A13), we find
\[
S(e) = C + \frac{|\kappa\rangle \langle \kappa^*|}{\gamma_n + \gamma_r - i(\omega - \omega_0)}, \quad (A17)
\]
and
\[
S(e^*) = C + \frac{|\kappa\rangle \langle \kappa^*|}{\gamma_n - \gamma_r - i(\omega - \omega_0)}. \quad (A18)
\]

Note in Eq. (A18) that \(\gamma_r \rightarrow -\gamma_r\) according to Eq. (A7)
where \(\gamma_r \propto \text{Im}(e)\). Here, we assume the non-resonant
scattering to be lossless, i.e., \(C^{-1} = C^\dagger\), and this assumption
restricts \(C\) in Eq. (A11) to take the form [5]
\[
C = \begin{bmatrix}
-\rho d \Delta_{22} & \pm i t e^{i (\xi_1 + \xi_2)} \\
\pm i t e^{i (\xi_1 + \xi_2)} & -\rho d \Delta_{22}
\end{bmatrix}. \quad (A19)
\]

For a homogenous metallic film with a thickness much larger
than the skin depth, the reflection and transmission
formula yields \(\xi_n \approx \tan^{-1}|\xi_n|/|\kappa_{\text{metal}}|^{1/2} \ll 1\)
where \(n = 1\) or 2 denotes the ports and take (+) sign for the
off-diagonal elements (non-resonant transmission coefficient)
under the definition of \(\psi_{n\pm}\) based on the EM field configu-
rations in Eqs. (A3) and (A4). Accordingly, it is reasonable
to use Eq. (A19) with \(\xi_n \ll 1\) and (+) sign for the off-diagonal
elements when describing the non-resonant response of a perforated
film possessing sub-wavelength holes or slits.

Applying Eqs. (A17)–(A19) to the quasi-reversibility
requirement of Eq. (A16) yields
\[
1 + t\sqrt{\eta_n}/\eta_n e^{i \Delta} = r e^{i \Delta_n}, \quad (A20)
\]
where \(\Delta = \xi_1 + \xi_2 + \pi/2 - \arg(\kappa_1) - \arg(\kappa_2), \quad \Delta_n = 2(\xi_n - \arg(\kappa_n)),\) and \(\nu\) is exclusive index as used in Eq. (A14).

In connection with \(\tau(\delta)\) and \(\rho_n(\delta)\) in Eq. (A14) and (A15),
respectively, \(t = |\eta_t|\) is non-resonant transmission magnitude,
\(r_n = |\eta_n|\) is non-resonant reflection magnitude, \(\Delta = \arg(\eta_t) - \phi\) denotes phase difference between the non-resonant and
resonant transmission at \(\delta = 0\), and \(\Delta_n = \arg(\eta_n) - \phi\) is phase
difference between the non-resonant and resonant reflection at
\(\delta = 0\). By solving Eq. (A20) for \(n = 1\) and 2, we finally obtain

\[
\cos \Delta_n = \frac{-\eta_n e^{i \phi}}{2\sqrt{\eta_n}} \quad \text{and} \quad \sin \Delta_n = p\sqrt{1 - \cos^2 \Delta}, \quad (A21)
\]

\[
\cos \Delta_n = \frac{1}{r} \left(1 + \frac{\eta_n}{\eta_n} \cos \Delta\right) \quad \text{and} \quad \sin \Delta_n = \frac{1}{r} \sqrt{\eta_n} \sin \Delta, \quad (A22)
\]

respectively, where the parity factor \(p = +1\) for \(\arg(\kappa_1) - \arg(\kappa_2) < \pi/2\) or \(-1\) for \(\arg(\kappa_1) - \arg(\kappa_2) > \pi/2\).

The nonreciprocal absorbance is directly obtained by using
Eq. (A12) in the absorbing power expression \(A_n = 2\gamma_n |\Phi|^2\)
for unit-amplitude light incidence at port \(n\) as
\[
A_n(\delta) = \frac{4\eta_n \eta_n}{1 + \delta^2}, \quad (A23)
\]

where the QBM absorbance probability is \(\eta_n = 1 - \eta_1 - \eta_2\).
The absorbance can be derived by energy conservation that
\(A_n + |\tau|^2 + |\rho_n|^2 = 1\). By introducing the intensity-phase
correlations of Eqs. (A21) and (A22) to Eqs. (A14) and (A15),
we arrive at Eq. (A23), showing that the intensity-phase
correlations satisfy the energy conservation requirement.

### C. Field Decomposition into the Resonant and Non-Resonant Configurations

Here, we describe how the total EM field \(\{E_{tot}, H_{tot}\}\) can be
decomposed into the resonant and non-resonant configurations.
We define two orthogonal complementary operators \(Q\) and
\(P\) that respectively project a field solution onto the resonant
and non-resonant subspaces in a Hilbert space formed by
all normalized field solutions of Maxwell’s equations for
a given system. The resonant configuration given by \(\{E_R, H_R\} = \{Q E_{tot}, Q H_{tot}\}\) fully describes excitation of the field
associated with the QBM in \(\{E_{tot}, H_{tot}\}\) while the
non-resonant configuration given by \(\{E_D, H_D\} = \{P E_{tot}, P H_{tot}\}\)
does not couple to the QBM at all. According to the definition
of modal amplitudes in Eqs. (A1)–(A4), the total field is given by
a superposition of fields associated with the port’s radiation
modes and the QBM as
\[
\begin{align*}
E_{tot} &= \psi_1 e_1 + \psi_2 e_2 + \psi_1 e_1^* + \psi_2 e_2^* + \Phi_{\text{QBM}} e^{-i\phi} \quad \text{in} \\
&\quad \text{QBM} + (C.C) \\
H_{tot} &= \psi_1 h_1 + \psi_2 h_2 - \psi_1 h_1^* - \psi_2 h_2^* + \Phi_{\text{QBM}} e^{-i\phi} \quad \text{in} \\
&\quad \text{QBM} + (C.C)
\end{align*}
\]
\(\text{(A24)}\)
\(\text{(A25)}\)

In the coupled-mode Eq. (A9) and its stationary form of
Eq. (A12), the part of incoming modes that excites the QBM
Φ is obviously the projection of $|\psi_+\rangle$ onto $|\kappa\rangle$. Thus, a normalized matrix $|\kappa\rangle^2 / \langle \kappa | \kappa \rangle = (2\gamma_{rad})^{-1} |\kappa\rangle^2 |\kappa\rangle$ projects the incoming mode onto the resonant configuration, i.e.,

$$Q |\psi_+\rangle = \frac{1}{2\gamma_{rad}} \left( |\kappa\rangle^2 / \langle \kappa | \kappa \rangle \right) |\psi_+\rangle = a |\kappa\rangle,$$

(A26)

where $a = (\kappa_1 |\psi_+\rangle + \kappa_2 |\psi_{2+}\rangle) / (2\gamma_{rad})$. $Q |\psi_+\rangle$ fully describes Φ as $|\kappa\rangle^2 |\kappa\rangle = \left( |\kappa\rangle^2 + r |\sigma\rangle^2 |\gamma\rangle^2 \right) |\psi_+\rangle$. Accordingly, projection of the outgoing radiation mode onto the resonant configuration is given by the scattered outgoing mode from $Q |\psi_+\rangle$, i.e.,

$$Q |\psi_−\rangle = SQ |\psi_−\rangle = a |\kappa\rangle,$$

(A27)

where $σ(δ) = (η_{rad} − η_{rad} + iδ)/(1 − iδ)$ and $S$ is the scattering matrix defined in Eq. (A17). On this basis, we finally have the resonant configuration as follows,

$$E_R = [a(k_1e_1^+ + k_2e_2) + \kappa_1σe_1^+ + \kappa_2σe_2^+] + \Phi e_{QBM} e^{-iat},$$

(A28)

$$H_R = [a(k_1h_1^+ + k_2h_2) - k_1σh_1^+ - k_2σh_2^+] + \Phi h_{QBM} e^{-iat},$$

(A29)

The projection onto the non-resonant configuration $P = 1 − Q$ is a complementary projector of $Q$. Thereby, the non-resonant configuration is obtained simply by the relation

$$\{E_D, H_D\} = \{PE_{tot}, PH_{tot}\} = \{E_{tot}, H_{tot}\} − \{E_R, H_R\}.$$
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