Stark control of plexcitonic states in incoherent quantum systems

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Electro-optic control of quantum dots embedded in plasmonic nanocavities enables active tuning of photonic devices for emerging applications in quantum optics such as quantum information processing, entanglement, and ultrafast optical switching. Here, we demonstrate the coherent control of plexcitonic states in (i) off-resonant and (ii) resonant coupled quantum systems through the optical Stark effect. We analyze a hybrid plasmon-emitter system which exhibits tunable photoluminescence, Stark-induced transparency, and vacuum Rabi splitting due to a quadratic Stark shift in the degenerate states of a quantum emitter (QE). In addition, a resonantly coupled system shows path interference of hybrid plexcitonic states due to Stark-induced splitting in a two-level QE. Our study shows that the Stark tuning of plexcitons not only mitigates decoherence in the quantum system but it also stimulates the on/off switching of spontaneous photon emission in the visible regime. Such tunable systems can be used to operate photonic integrated circuits for applications in quantum computing and information processing.

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I. INTRODUCTION

Active control of quantum states in an incoherent system has become a new challenge for operating multifunctional *in situ* programmable photonic integrated circuits (PICs) [1–3]. Controlling these systems in real time and obtaining desired properties is crucial for the development of quantum technologies such as quantum information processing, quantum computing, and single-photon sources [4,5]. In this regard, quantum plasmonics provides the fastest route to achieve such control by manipulating the quantum properties of surface plasmons and excitons such as a quantum emitter (QE) through cavity quantum electrodynamics (CQED) [6,7]. Due to its extreme field confinement, the plasmon cavity entails profound coupling with QE which enables the coherent control of quantum devices at a single-photon level [7,8].

A coherent interaction of a plasmon cavity with an exciton generates mixed quantum states also known as plexcitonic dressed states which inherently possess all the information of a controlled quantum system [9]. Tuning these states as a function of coupling strength yields two distinct phenomena, i.e., Fano resonance (FR) and vacuum Rabi splitting (RS) for intermediate- and strong-coupling regimes, respectively [8,10,11]. Both coupling mechanisms enable coherent oscillations and allow the quantum superposition of different states which are essentially important for entanglement and quantum information [12,13]. So far FR and RS have been demonstrated in resonantly coupled plasmon-emitter systems while coherent control has been achieved by modulating the geometrical parameters of the structure [14], and this demands a challenging fabrication process.

Nevertheless, achieving coherent control in an off-resonant coupled quantum system is of potential importance for implementing active photonic functionalities such as ultrafast switching [15], signal processing, and lasing at the nanoscale [16,17]. One of the promising ways to manifest dynamical control over quantum systems is to use an external variable influence. At present, active tuning of polaritonic modes under CQED treatment has been explored either through resonant excitation [18], magnetic tuning [19], dielectric control [20], or by exploiting voltage-tunable two-dimensional (2D) materials [21] and quantum dots [22]. However, the coherent tuning of off-resonant quantum states in real time through the optical Stark effect (OSE) has yet to be discussed.

In contrast to previous studies discussing Fano resonance and tunability of polariton modes through different plasmonic structures [23–25], our work demonstrates an alternative approach of tuning plexcitonic modes of an incoherent quantum system via OSE. In this article, we study the Stark tuning of plexcitons in two scenarios: (i) In a hybrid plasmon-emitter system excited and coupled off resonantly, the probe (Stark) field shifts the eigenenergies of a three-level QE and coherently drives the off-resonant states close to resonance which leads to path interference. The coherent phase shift of plexcitons generates a transparency window which we refer to as Stark-induced transparency (SIT). Furthermore, we inspect how a small perturbation in the Stark field yields a large modulation in the vacuum Rabi splitting. (ii) In a resonantly coupled system, the Stark field lifts the degeneracy by splitting the excited state of a two-level QE which induces path interference of the hybrid plexcitonic modes in the system. Our concept of Stark tuning of hybrid modes explicates the coherent control of quantum devices as a proof-of-principle concept which can be used to mitigate the decoherence of a quantum system and also demonstrates active tuning of spontaneous photon emission in the visible regime.

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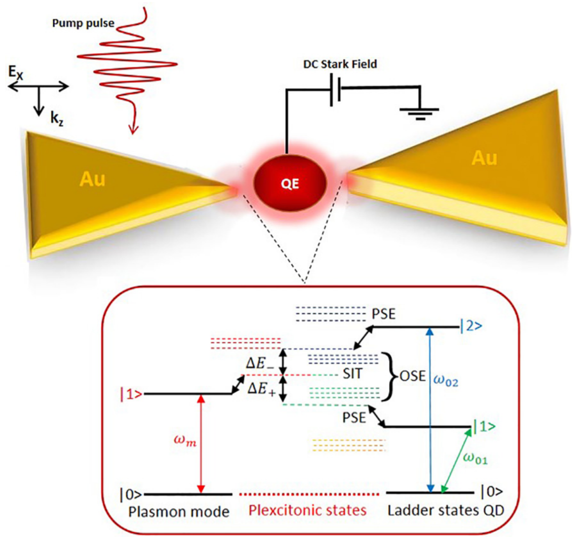

FIG. 1. Schematic diagram of a hybrid system of an Au bow-tie nanoantenna and a three-level QE. QE is attached to an external voltage bias which induces Stark shifts in the hybrid plexcitonic states.

II. THEORETICAL FRAMEWORK

We investigate the dynamics of a hybrid quantum system by formulating an analytical model that simply describes the interaction between a plasmon mode and QE in the context of a coupled harmonic oscillator [26]. The hybrid plasmon-emitter system consists of an Au bow-tie nanoantenna [27] and a QE, placed at a distance R between the nanodimer, as shown in Fig. 1. The bow-tie nanoantenna is irradiated with a p-polarized light of frequency (ω_o) and amplitude (E_o), which creates intense localized dipole modes (LSPs). We define ($\hbar\omega_m\hat{a}^\dagger\hat{a}$) as the unperturbed energy of the LSP mode with annihilation ($\hat{a}$) and creation ($\hat{a}^\dagger$) operators. The dipole interaction of the driving pulse with the plasmon mode is expressed as $\mathcal{M} = E_o\mu_m e^{-i\omega_o t}(\hat{a}^\dagger) + \text{H.c.}$ [20], where μ_m is the dipole matrix element defined as $\mu_m = -i\sqrt{12\pi\epsilon_o\eta r^3\hbar}$ with r as the edge size of the bow-tie triangle. Although our methodological approach differs for two different cases (off-resonant or resonant coupling), the defined total Hamiltonian is mathematically the same. Hence, QE is specified as a three-level system in the ladder configuration with basis states expressed as raising $\hat{\sigma}^\dagger = |e\rangle\langle g|$ and lowering $\hat{\sigma} = |g\rangle\langle e|$ operators for both cases. The transition energies of $|1\rangle$ and $|2\rangle$ excited states of QE are defined as $\hbar\omega_{01}$ and $\hbar\omega_{02}$, respectively, while the ground state energy is taken as zero. Since the pump field is off resonant to the plasmon mode and QE, we assume that there is no interaction between the pump field and the QE for both cases. Therefore, we neglect the dipole coupling of QE. The interaction between the plasmon mode and QE is quantified through a phenomenological constant (f) measured as the coupling strength between two oscillators. We choose the value of (f) normalized to the frequency of the excitation field according to the weak- ($f = 0.01\omega_o$) and intermediate- ($f = 0.05\omega_o$) coupling regimes as referenced in Ref. [10]. After applying rotating-wave and dipole approximations [19], we define the total Hamiltonian for a three-level QE interacting with the plasmon mode [28,29] as follows,

$$\hat{\mathcal{H}} = \hbar\Delta_m\hat{a}^\dagger\hat{a} + \hbar(\Delta_{01} - \Delta E)\hat{\sigma}_{11} + \hbar(\Delta_{02} + \Delta E)\hat{\sigma}_{22} + i\hbar f\sum_j(\hat{a}^\dagger\hat{\sigma}_{0j} - \hat{a}\hat{\sigma}_{0j}^\dagger) + \mathcal{M}, \tag{1}$$

where $\Delta_m = (\omega_m - \omega_o)$ is the detuning of the plasmon mode frequency ω_m from the incident field frequency ω_o, and $\Delta_{0j} = (\omega_{0j} - \omega_o)$ the detuning of the QE transition frequency ω_{0j} from the driving source frequency ω_o. To induce a Stark shift in the excitonic levels, QE is attached to the external voltage bias, as shown in Fig. 1. Under the second-order perturbation treatment, the Stark shift (ΔE) produced in the excited states of QE is referred to as a quadratic Stark shift, which is calculated through the relation $\Delta E = -1/2\alpha E^2$, where E is the applied electric field and α is the polarizability of the atomic system, derived from $9/2(a_o)^3$ with a_o as the Bohr radius [30]. In our simple model, we ignore electron spin and other effects such as the relativistic correction and Lamb shift by considering these effects as small in comparison to the applied electric field. In contrast to the linear Stark effect, a quadratic Stark shift results from the induced dipole moment (μ_{qe}) of the energy states after the application of an external voltage bias. The change in μ_{qe} is proportional to the strength of the applied electric field. In this way, as the electric field strength increases, the induced dipole moment becomes more significant, which causes a pronounced shift in the plexcitonic states. In addition, when the interaction of the system with the environment reservoir is taken into account, our plasmon-emitter system acts as an open quantum system under the Markovian approximation [31].

The dynamics of the hybrid plasmon-emitter system is evaluated through the Heisenberg-Langevin approach [32] which provides a simple method to evaluate operators, and handle damping and the interaction with an external field. The equations are as follows:

$$\partial_t\hat{a} = \frac{i}{\hbar}[\hat{\mathcal{H}}, \hat{a}] - \frac{\gamma_m}{2}\hat{a}, \tag{2}$$

$$\partial_t\hat{\sigma}_{0j} = \frac{i}{\hbar}[\hat{\mathcal{H}}, \hat{\sigma}_{0j}] - \frac{\gamma_{0j}}{2}\hat{\sigma}_{0j}. \tag{3}$$

These equations combine the Heisenberg equation of motion with the damping terms resulting from the Markovian interaction with the reservoir which determines the decay rates of the plasmon mode ($\gamma_m = 72$ meV) and the excitonic levels of QE (γ_{0j}). The equations of motion derived for the plasmon mode amplitude $\langle\hat{a}\rangle$ and off-diagonal density matrix elements $\langle\hat{\sigma}_{0j}\rangle$ of QE using bosonic commutation relations are as follows:

$$\partial_t\langle\hat{a}\rangle = -[i(\Delta_m + \gamma_m/2)]\langle\hat{a}\rangle + f\langle\hat{\sigma}_{01}\rangle + f\langle\hat{\sigma}_{02}\rangle + \mathcal{M}, \tag{4}$$

$$\partial_t\langle\hat{\sigma}_{01}\rangle = -[i(\Delta_{01} + \Delta E) + \gamma_{01}/2]\langle\hat{\sigma}_{01}\rangle + f\langle\hat{a}\rangle, \tag{5}$$

$$\partial_t\langle\hat{\sigma}_{02}\rangle = -[i(\Delta_{02} - \Delta E) + \gamma_{02}/2]\langle\hat{\sigma}_{02}\rangle + f\langle\hat{a}\rangle. \tag{6}$$

We deliberately select smaller amplitudes for the probe field. This choice offers the additional advantage of minimizing other nonlinear effects, thus enhancing the precision of our approach in the weak-field limit. Therefore, while driving the analytical solutions, we employ a weak-field approximation by considering the intensity of the incident field is sufficiently

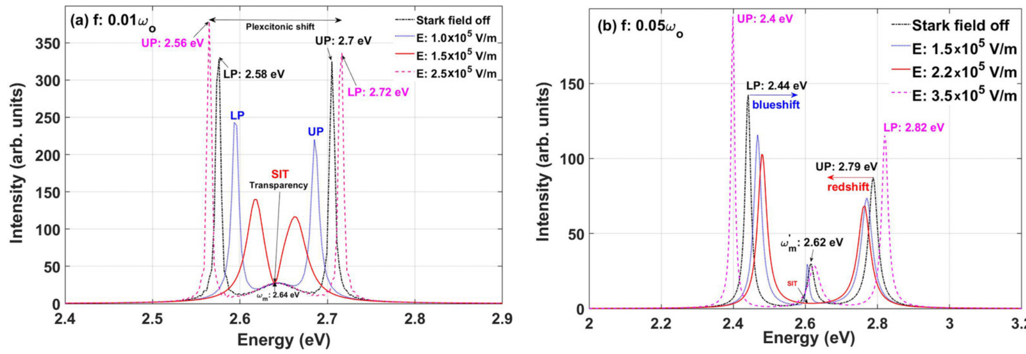

FIG. 2. Stark-induced shifts and transparency in the plexcitonic states (off-resonant coupling) for (a) weak- ($f = 0.01\omega_o$) and (b) intermediate-coupling strengths ($f = 0.05\omega_o$), respectively. A transparency window (SIT) appears at plasmon mode frequencies (a) $\omega_m = 2.64$ eV and (b) $\omega'_m = 2.62$ eV for an optimum value of the Stark field. The eigenfrequencies of the first and second excited states of QE are taken as $\omega_{01} = 2.58$ eV, $\omega_{02} = 2.7$ eV for the weak-coupling case and $\omega_{01} = 2.5$ eV, $\omega_{02} = 2.7$ eV for intermediate-coupling cases with decay rates $\gamma_{0j} = 5$ μeV, respectively.

weak to demonstrate our ability to perform Stark tuning even at low intensities. In this way, we evaluate the linear dynamics of the system and disregard higher-order terms, and hence we also ignore the noise operator terms. Moreover, in the weak-field limit, the excitonic population $\langle\hat{\sigma}^\dagger_{0j}\hat{\sigma}_{0j}\rangle \ll 1$ is minute, therefore, we have $\langle\hat{\sigma}_{00}\rangle = 1$ and $\langle\hat{\sigma}_{jj}\rangle = 0$ [19]. We perform the time evolution of Eqs. (4)–(6) numerically through the Runge-Kutta method using the MATLAB program and compute the output scattered intensity of the hybrid plasmon-emitter system given by the relation $I_{\text{sca}} = |\langle\hat{\sigma}_{0j}\rangle + \langle\hat{a}\rangle|^2$ [32,33].

A. Stark-induced shift in off-resonantly coupled plexcitonic states

We analyze the scattered intensity of a plasmon-emitter system in the weak-, intermediate-, and strong-coupling regimes [10], as shown in Fig. 2. The energy spectra show three plexcitonic states corresponding to the lower plexciton (LP), plasmon mode (PM), and upper plexciton (UP) in the hybrid system. The transition frequencies of QE, $\omega_{01} = 2.5$ eV and $\omega_{02} = 2.7$ eV, are kept off resonant to the PM frequency, $\omega_m = 2.64$ eV. The driving field is also taken off resonant from both QE and PM, with the excitation frequency $\omega_0 = 1.997$ eV. In the absence of the Stark field, a shift is observed in the excitonic levels due to the coupling of bare states with the plasmon mode, where such a type of nonresonant shift results in the bending of the bare energy levels. In the dispersive limit, when $\Delta = (\omega_{0j} - \omega_m) \gg f$ and $f \ll (\gamma_{0j} - \gamma_m)/2$, the shift in the plexcitonic states occurs due to the plasmonic Stark effect (PSE) [34]. In Fig. 2(a), we observe a PSE shift in the excitonic levels forming UP and LP states with energies 2.72 and 2.55 eV, respectively. When the Stark field is turned on, UP undergoes a redshift, while LP blueshifts towards the plasmon resonance frequency. Upon increasing the electric field strength, the incoherent plexcitonic states enter the resonant regime resulting in the path interference of UP and LP states with PM [see the red curve in Figs. 2(a) and 2(b)]. The coherent interaction of plexcitons due to the Stark field yields path interference [35–37] and a transparency window appears at the plasmon resonance frequency ($\omega_m = 2.64$ eV). We refer to this transparency as Stark-induced transparency (SIT).

A significant increase in the quadratic Stark shift is observed with an increase in the electric field strength which results in a significant redshift and blueshift of the UP and LP states from the bare energy levels, respectively. In the intermediate-coupling regime ($f = 0.05\omega_o$), the new hybrid states are UP = 2.79 eV, LP = 2.44 eV, and PM at $\omega'_m = 2.62$ eV. When the Stark field is turned on the UP level redshifts to a lower-energy level with a decrease in the energy of 26 meV and LP blueshifts to a higher level with an increase in the energy up to 39 meV. As the levels transit towards the resonant energy state of the polaritons, the coherent interaction yields interference along with transparency at 2.60 eV. The redshifts and blueshifts in the UP and LP states change the overall dynamics of the hybrid molecular system. The crossing of plexciton states to either side leads to vacuum Rabi splitting due to a quadratic shift in the LP mode with an energy difference of 226 meV from the polariton mode resonance and the shift in UP causes splitting of 194 meV. A small increase in the electric field transforms incoherent states to a resonantly oscillating coherent system. Similar effects are observed in the strongly coupled plexcitonic states, in which the Stark field induces a maximum Rabi splitting of $\Omega \leqslant 350$ meV. Here, Ω is the difference between transition frequencies of shifted levels with the corresponding polariton mode frequency (ω'_m).

To validate these shifts, we plot the photoluminescence (PL) spectra as a function of detuning (Δ_m). The photoluminescence for a cavity-emitter coupled system is defined as [38],

$$\text{PL} = \frac{\gamma}{\pi}\left|\frac{-if}{[\gamma_+ + i(\Delta/2 - i\Delta_m)^2 + (\Omega_R/2)^2]}\right|^2, \qquad (7)$$

where Ω_R is the complex form of the generalized Rabi frequency defined as $\Omega_R = \sqrt{4f^2 - (4\gamma_- + i\Delta_{0j})^2}$ with the energy dissipation rates $\gamma_+ = |\frac{\gamma_{oj}+\gamma_m}{4}|$ and $\gamma_- = |\frac{\gamma_{oj}-\gamma_m}{4}|$. The detuning between shifted energies of plexciton states and the plasmon mode is defined as $\Delta = (\omega'_{0j} - \omega'_m)$. We calculate the PL intensity for different values of the Stark field and evaluate the quadratic shift in UP and LP for three different coupling regimes as shown in Fig. 3. In the dissipative regime, the plexciton with nearly resonant states close to the zero detuning region [point of SIT in Fig. 3(a)] yields maximum

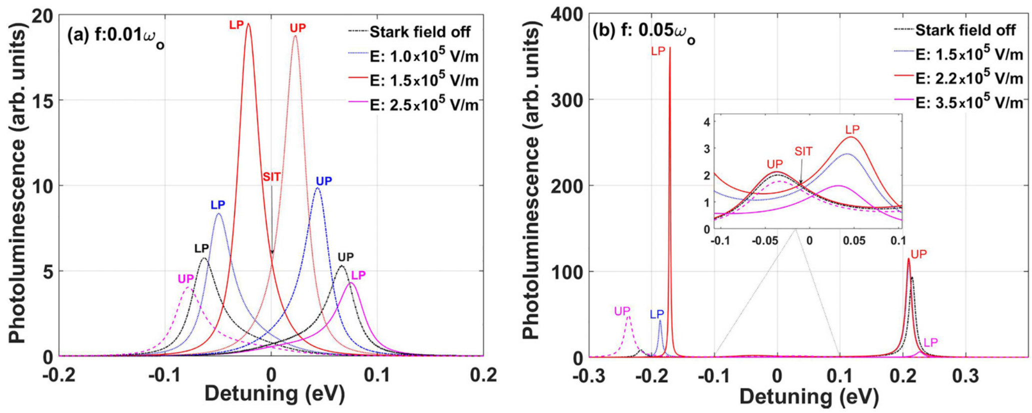

FIG. 3. Photoluminescence spectra of Stark-induced plexcitonic shifts as a function of detuning (Δ_m) for (a) weak- ($f = 0.01\omega_o$) and (b) intermediate- ($f = 0.05\omega_o$) coupling regimes. The spectral parameters are the same as mentioned in Fig. 2.

PL. On the other hand, in Fig. 3(b), the UP and LP peaks split with large detuning and maximum transitions occur at the point of crossing near zero detuning [see the inset of Fig. 3(b)] and as the levels move away towards large detuning, the PL from both UP and LP states decreases profoundly (purple dashed and solid curve). We also obtained similar results for the strong-coupling regime. The splitting of UP/LP states enhances further with an increase in field strength and maximum PL occurs at the induced transparency (the results are not shown here). The tuning of plexcitonic levels not only produces a spectral shift but it also switches the photoluminescence intensity to on/off by modulating the coherent dynamics of the hybrid system through an external probe (see Fig. 3). Moreover, our proposed system demonstrates the tuning of PL in the optical frequency range. In this way, Stark tuning of an incoherent quantum system transforms it into an actively tunable coherent photonic device that carries a strong potential for nanoscale lasing and on-demand PIC technologies.

B. Stark-induced splitting in resonantly coupled plexcitonic states

In this section, we illustrate the Stark splitting in a two-level QE through an external probe field which induces hybrid plexcitonic states and Rabi splitting in a resonantly coupled plasmon-emitter system. For this, we use the same Hamiltonian as defined in Eq. (1) by replacing Δ_{01} and Δ_{02} with a detuning parameter (Δ_q) and derive the equations of motion for the plasmon mode and off-diagonal density matrix elements of a two-level QE by using Eqs. (2) and (3) as follows,

$$\partial_t\langle\hat{a}\rangle = -[i(\Delta_m + \gamma_m/2)]\langle\hat{a}\rangle + f\langle\hat{\sigma}_{01}\rangle + f\langle\hat{\sigma}_{02}\rangle + \mathcal{M}, \quad (8)$$

$$\partial_t\langle\hat{\sigma}_{01}\rangle = -[i(\Delta_q + \Delta E_-) + \gamma_{01}/2]\langle\hat{\sigma}_{01}\rangle + f\langle\hat{a}\rangle, \quad (9)$$

$$\partial_t\langle\hat{\sigma}_{02}\rangle = -[i(\Delta_q - \Delta E_+) + \gamma_{02}/2]\langle\hat{\sigma}_{02}\rangle + f\langle\hat{a}\rangle, \quad (10)$$

where the probe field splits the excited state of QE with the transition frequency (ω_{ge}) and the shift in the levels is evaluated as ΔE (see Fig. 4). $\Delta_q = (\omega_{ge} - \omega_o)$ is the detuning of the two-level QE transition frequency (ω_{ge}) from the incident light frequency (ω_o). For the off-resonant excitation, the frequency of the pump field is taken as $\omega_o = 1.997$ eV. Nevertheless, the plasmon mode and QE couple resonantly with frequencies $\omega_m = 2.64$ eV and $\omega_{eg} = 2.68$ eV, respectively.

Furthermore, the values of γ_m, γ_{0j}, and $\mathcal{M}$ are the same as defined in Fig. 2. After performing the time evolution of Eqs. (8)–(10) numerically, we compute the scattered intensity ($I_{\rm sca}$) of a hybrid system as a function of excitation wavelength. Figure 4(a) shows the schematic of an energy-level diagram of plexcitonic states, a plasmon mode, and a two-level QE. We calculate the energy spectra of plexcitonic states with Stark splitting and shift for different values of the electric field. The black dotted curve in Figs. 4(b) and 4(c) indicates two peaks which correspond to hybrid modes in the absence of a probe field. The peak spectral positions of hybrid modes appear at $\omega'_m = 2.61$ eV and $\omega'_{ge} = 2.72$ eV for a weakly coupled system ($f = 0.02\omega_o$) and 2.52 and 2.8 eV for intermediate coupling ($f = 0.05\omega_o$), respectively. When the Stark field is turned on, the excitonic level of QE splits into two, which then couple to the existing polaritonic mode. The coupling of discrete states with the continuum mode results in the path interference of three coherent states giving rise to multiple hybrid modes. The new plexcitonic modes show three peaks with energies 2.52, 2.67, and 2.73 eV for PM, LP, and UP states, respectively [see red in Figs. 4(b) and 4(c)]. As the field strength increases the plexcitonic states indicate the signature of Mollow triplets [39], though in our case, Mollow triplets result from the interaction of intense LSPs with Stark-split excitonic levels. With the increase in the probe field, the LP redshifts and UP blueshifts with large detuning with PM. For $f = 0.05\omega_o$ coupling, the hybrid mode splits with an energy shift of 282 meV. In the presence of a Stark field, the splitting generates two new plexcitonic states with energies 2.7 eV (UP) and 2.8 eV (LP). As the field strength increases to 3.1×10^5 V/m, the plexciton dynamics drastically change with a maximum energy shift of 491 meV, [Fig. 4(c)].

We validate the plexciton tuning through photoluminescence spectra and evaluate the optical response of the system for different values of the Stark field. Figures 5(a) and 5(b) show the contour plots of PL for UP and LP states as a function of detuning (Δ_m). For the values of $E = 1.2 \times 10^5$ V/m and $E = 1.8 \times 10^5$ V/m, LP and UP peaks show mixed plexcitonic states, with a minimum energy difference between plexcitons of 57 meV [Fig. 5(a)] and 133 meV [Fig. 5(b)],

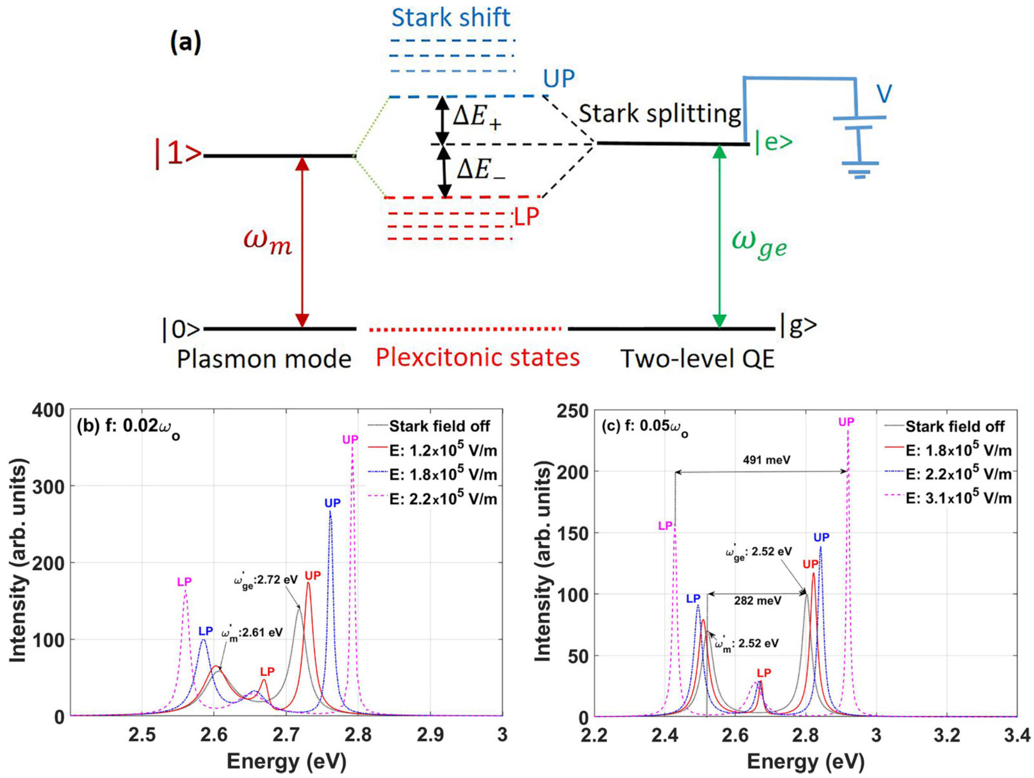

FIG. 4. (a) Schematic diagram of PM coupled to two-level QE ($\omega_m \approx \omega_{01}$), and the tuning of plexcitonic states through Stark-induced splitting and shifts. Stark-tuned hybrid plexcitonic states in the resonantly coupled plasmon-emitter system for (b) weak and (c) intermediate coupling.

respectively. As the field strength increases, the spectral energy and the PL intensity of UP and LP are modulated in a profound manner. The UP plexciton blueshifts with a maximum energy of 148 meV and LP redshifts to 102 meV [Fig. 5(a)]. In contrast to this, for strongly coupled LP and UP modes, the spectral energy shifts are more pronounced with detuning of 264 and 341 meV, respectively [see Fig. 5(b)].

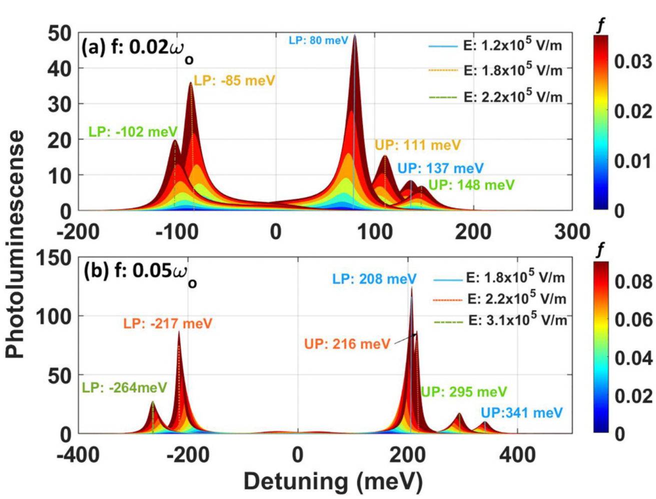

FIG. 5. Photoluminescence spectra of Stark-tuned plexcitonic states as a function of detuning (Δ_m) for (a) $f = 0.02\omega_o$ and (b) $f = 0.05\omega_o$. UP plexciton blueshifts and LP redshifts as the Stark field strength increases.

Here, some small peaks also appear close to zero detuning along with intense UP/LP peaks which show the signature of Rabi splitting [10] in the plexciton. Nevertheless, as the splitting between UP and LP states increases, the PL intensity decreases gradually, indicating the switching of spontaneous photon emission from high (on) to low (off). In this way, the Stark splitting of the exciton not only modulates the energy of hybrid states but also actively tunes the photoluminescence intensity (on/off) as a function of the probe field.

III. CONCLUSIONS

In summary, we theoretically investigate the coherent control of plexcitonic states in incoherent quantum systems through the optical Stark effect (OSE). We demonstrate the Stark tuning of hybrid plexciton modes in a coupled plasmon-emitter system. A small perturbation in the applied field modulates the energy eigenstates of the quantum emitter which modifies the hybrid modes in a coherent manner. At first, we evaluate the Stark-induced spectral shifts of plexciton states in an off-resonant coupled system. The pronounced resonant shifts in the excitonic levels result in the coherent interference of dressed states leading to tunable photoluminescence. We also report the Stark-induced transparency in hybrid states in both weak- and intermediate-coupling regimes. Furthermore, we investigated the dynamics of hybrid plexciton modes, in a resonantly coupled plasmon-emitter system, due to splitting of the energy eigenstate of QE. The Stark-induced splitting shows the signature of Mollow triplets in the plexcitonic modes with a maximum energy splitting up to 491 meV between the upper and lower plexcitons. We also explore the impact of Stark tuning of plexcitons on photoluminescence spectra, which shows the active control of photon transitions and on/off switching of spontaneous emission in the visible regime. Our proposed coherent optical control of quantum states through the Stark effect clearly indicates its potential in quantum information processing and quantum computing applications. In addition to this, such tunable signatures can be used for engineering dynamical nanophotonic systems with potential applications such as lasing, ultrafast switching, optical modulation, single-photon emission, and sensing applications.

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