Quasi-Chiral Interactions between Quantum Emitters at the Nanoscale

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We present a combined classical and quantum electrodynamics description of the coupling between two circularly-polarized quantum emitters held above a metal surface supporting surface plasmons. Depending on their position and their natural frequency, the emitter-emitter interactions evolve from being reciprocal to non-reciprocal, which makes the system a highly tunable platform for chiral coupling at the nanoscale. By relaxing the stringent material and geometrical constraints for chirality, we explore the interplay between coherent and dissipative mechanisms in the system. Thus, we reveal a quasi-chiral regime in which its quantum optical properties are governed by its subradiant state, giving rise to extremely sharp spectral features and strong photon correlations.

A quarter of a century ago, the theory of cascaded quantum systems was developed independently by Gardiner and Carmichael [1–3]. By construction, the theory describes distant source-target systems whereby non-reciprocal, unidirectional interactions arise naturally: the former is coupled to the latter while completely forbidding the opposite. Today, the emerging field of chiral quantum optics [4] seeks to realize, exploit and control physical systems exhibiting non-reciprocal light-matter interactions. Properly harnessed, the chiral coupling between quantum emitters (QEs) and photons at the quantum level promises a myriad of nontrivial applications in quantum communication, information and computing, including non-reciprocal single-photon devices [5], optical isolators [6], optical circulators [7] and even integrated quantum optical circuits [8].

In this Letter, we present a classical and quantum electrodynamics description of the most elemental physical platform yielding non-reciprocal interactions at the nanoscale: two circularly-polarized QEs placed on top of a flat metal surface supporting tightly confined surface plasmons (SPs). The high tunability of the system, which can be manipulated through the relative position of the QEs, their natural frequencies, and the distance from the metal surface, enables us to unveil a rich landscape of coherent and dissipative emitter-emitter couplings. This includes the much sought-after chiral (fully non-reciprocal) regime [4]. Through the comparison with the quantum cascaded formalism [9], we set a well-defined criterion for chiral coupling. Moreover, by relaxing the geometric and material parameters satisfying the unidirectional conditions, we investigate the evolution of the one- and two-photon far-field spectra of the system in the transition from the conventional (reciprocal) regime. This allows us to identify a less stringent, more easily accessible quasi-chiral configuration, in which the system develops extremely narrow spectral features and very strong photon correlations.

The starting point of our description for the two QEs and their electromagnetic (EM) coupling is the Hamiltonian

\[ H = \omega_0 (\sigma_i^1 \sigma_1^1 + \sigma_i^2 \sigma_2^2) + g_{12} \sigma_i^1 \sigma_2^1 \sigma_2^1 + g_{21} \sigma_i^2 \sigma_1^1 \sigma_1^1, \]

where \( \sigma_i^1 \) is the lowering (raising) operators of the QE-\( i \), and \( \omega_0 \) is its natural frequency. Hermiticity dictates that \( g_{12} = g_{21}^* \), which makes coherent interactions fully reciprocal. Next, we introduce damping in our model in the form of the inherent decay of both QEs and their dissipative coupling (collective decay). Both mechanisms are accounted for by the Master equation [10, 11]

\[ \frac{d}{dt} \rho = i [\rho, H] + \sum_{i,j=1,2} (\gamma_{ij}/2) \mathcal{L}_{ij} \rho + \sum_{i=1,2} (P_i/2) (\mathcal{L}_{ii} \rho)^d, \]

where \( \gamma_{ij} \) stand for self (\( i = j \)) and collective (\( i \neq j \)) decay rates, and \( \mathcal{L}_{ij} \rho = 2 \sigma_j \rho \sigma_i^d - \sigma_i^d \sigma_j^d \rho - \rho \sigma_i^d \sigma_j^d \) is the usual Lindblad superoperator \( (h = 1) \). Note that we have also included the incoherent driving rates \( P_i \), which feed population into both QEs (within the vanishing pump limit) [12]. Similarly to their coherent counterparts, the collective dissipative constants satisfy \( \gamma_{12} = \gamma_{21}^* \).

The Master equation above encompasses a remarkably rich phenomenology as a function of the coupling parameters \( \{g_{12}, \gamma_{12}\} \), which are in general complex quantities. Interestingly, it maps onto the cascaded Master equation if

\[ \left| \frac{g_{12}}{\gamma_{12}} \right| = \frac{1}{2}, \quad \arg \left( \frac{g_{12}}{\gamma_{12}} \right) = \frac{3\pi}{2}. \]  

(1)

The two equations above establish the magnitude and phase balance in the coherent and dissipative components of the QE interactions that give rise to chiral coupling. Specifically, Eqs. (1) yield complete isolation of QE-2 from the output of QE-1. Note that the damping rates must satisfy \( 0 \leq |\gamma_{12}| \leq \gamma_{11} \) (from now on, we assume \( \gamma_{22} = \gamma_{11} \)), and complete equivalence to the quantum cascaded formalism occurs only in the limit \( |\gamma_{12}| = \gamma_{11} = \gamma_{22} \) [9].

We parameterize our Master equation using EM calculations for the setup sketched in the left inset of Fig. 1 (a). We model the metal response through a Drude permittivity \( \varepsilon(\omega) \), with plasma frequency \( \omega_{pl} = 3.4 \text{ eV} \) (which corresponds to an asymptotic SP frequency \( \omega_{sp} \approx \omega_{pl} / \sqrt{2} = 2.40 \text{ eV} \)) and investigate the impact of metal absorption by varying the Drude damping \( \gamma_D \). The dipole moment of the two QEs is \( \mathbf{d} = |\mathbf{d}| (\hat{x} + i \hat{z}) / \sqrt{2} \), whose circularly-polarized character opens up novel degrees of freedom via the complex arguments of the coherent and dissipative...
tive coupling strengths. Within the formalism of macroscopic quantum electrodynamics [19], the coupling parameters have the form \( g_{ij} = \omega_0^2 d_i^\dagger \text{Re} \{ \mathbf{G} (\mathbf{r}_i, \mathbf{r}_j, \omega_0) \} d_j / \epsilon_0 c^2 \) and \( \gamma_{ij} = 2 \omega_0^2 d_i^\dagger \text{Im} \{ \mathbf{G} (\mathbf{r}_i, \mathbf{r}_j, \omega_0) \} d_j / \epsilon_0 c^2 \), where \( \mathbf{G}(\cdot) \) stands for the EM Dyadic Green’s function.

To gain insight into the emergence of chirality in our system, we compare numerical solutions of Maxwell’s equations against analytical predictions for the coupling strengths obtained by keeping only the plasmon-pole contribution to the Green’s function [12, 20]. From now on, we assume that both QEs are placed at the same height, \( z \), and introduce the variable \( x = x_2 - x_1 \) (position of QE-2 relative to QE-1). In the limit of vanishing metal losses and for \( k_{\text{sp}}(\omega_0)|x| \gg 1 \) (where \( k_{\text{sp}}(\omega_0) = (\omega_0/c)\epsilon(\omega_0)/\sqrt{\epsilon(\omega_0) + 1} \) is the SP wave-vector at the QE frequency), they read

\[
g_{12} = \eta(\omega_0) e^{-2\sqrt{k_{\text{sp}}(\omega_0)^2 - (\omega_0/c)^2} z} / (2\pi k_{\text{sp}}(\omega_0)|x|) \cdot e^{ik_{\text{sp}}(\omega_0)x} = g_{21}^*, \tag{2}
\]

and

\[
\gamma_{12} = 2i \text{sgn}(x) g_{12} = \gamma_{21}^*, \tag{3}
\]

with \( \eta(\omega) = (\omega/c)^3 d^2(\omega^2)/\epsilon_0 |\epsilon(\omega) + 1|^{3/2} |\epsilon(\omega) - 1| \). Importantly, Eqs. (2) and (3) naturally satisfy Eqs. (1), which proves that the QEs become chirally coupled when their interaction is fully mediated by confined SPs [4].

Figure 1(a) plots the absolute value of the effective coupling strength \( |g_{12} - i\gamma_{12}/2| \), which combines the dissipative and coherent components, normalized to \( |g_{12}| \) as a function of the distance between the two QEs (\( \omega_0 = 2.35 \) eV, \( \lambda_0 = 527 \) nm, and \( z = 5 \) nm). Numerical calculations for two different \( \gamma_D \) (green dashed and blue dotted lines) are compared against the lossless analytical result (red solid line). Remarkably, for \( |x| \gtrsim 0.1 \lambda_0 \approx 53 \) nm, the QE interactions are governed by SPs (analytical and numerical predictions are in perfect agreement) and the effective coupling becomes directional: its magnitude approaches \( 2|g_{12}| \) for \( \text{sgn}(x) > 0 \) and vanishes for \( \text{sgn}(x) < 0 \). We can also observe that the inclusion of metal absorption does not degrade the chiral character of the QE coupling, which is insensitive to \( \gamma_D \). The right inset shows that the only effect of the Drude losses is to reduce the efficiency of the dissipative coupling mechanism, weighted by \( |\gamma_{12}/\gamma_{11}| \). Note that recent reports have demonstrated that this ratio (also termed as the \( \beta \)-factor) can be optimized in complex plasmonic structures [21, 22], well beyond our proof-of-principle proposal.

Figure 1(b,c) analyze the deviation of SP-assisted coupling from Eqs. (1) as a function of the QE distance and natural frequency \( z = 5 \) nm). We can observe that both the magnitude ratio (b) and phase difference (c) undergo oscillations around the chiral condition (white regions), which become stronger as \( x/\lambda_0 \) and \( \omega_0 \) decrease. Both maps are obtained from analytical expressions [12], and demonstrate the possibility of varying the degree of chirality in QE coupling exclusively through SPs. Red solid lines in Figs. (d) and (e) plot cuts at \( \omega_0 = 2.35 \) eV in panels (b) and (c), respectively. Exact numerical calculations for increasing \( z \) are shown in dashed green (30 nm), dotted blue (60 nm), dot-dashed orange (150 nm), and thin pink (300 nm) lines. The deviations from Eqs. (1) are significantly larger than in the upper panels. This demonstrates that the tunability of the system increases further when the contribution of free-propagating modes to the Dyadic Green’s function becomes comparable to SPs.

Now we have proven the tuning of emitter-emitter interactions in our EM set-up, we investigate its influence in the quantum optical properties of the system. Figure 2(a) reveals the hallmark of chirality in the QE populations. It shows the steady-state population of QE-2, \( n_2 = |\sigma_2^\dagger \sigma_2| \), normalized to its population in isolation, \( n_0 = P_2/(P_2 + \gamma_{22}) \), versus \( g_{12}/|\gamma_{12}| \) and \( \text{arg}(g_{12}/|\gamma_{12}|) \). From now on, and unless stated otherwise, we take \( \gamma_{12} = |\gamma_{12}| = \gamma_{11}^* \). As anticipated, when Eqs. (1) are fulfilled (intersection of the horizontal and vertical long-dashed white lines) one indeed finds \( n_2 = n_0 \). This is a true manifestation of chiral coupling, since the output of the QE-2 drives QE-1, but the opposite is prevented. Note that
we find regions with $n_2/n_0$ both larger and smaller than unity away from the chiral conditions.

We focus now in the parameter range given by the vertical white dashed line in Fig. 2(a), and investigate the properties of the coupled QEs when only the magnitude condition for chirality is met. As shown in Fig. 1, this regime is significantly more accessible that the chiral configuration, as the geometric (QEs height and separation) and material (QE natural frequency) constraints on the system are greatly relaxed. We explore the system through the normalized power spectrum \[ S(\omega) = \lim_{t \to \infty} \text{Re}\{\sum_{\omega_0}^{\infty} \xi^i(t) \xi(t + \tau) e^{i\omega\tau}/(\pi\eta_\gamma)\} \], which accounts for coherent superposition of the photon emission by both QEs, with $\xi = (\sigma_1 + \sigma_2)/\sqrt{2}$ and $\eta_\gamma = (\xi^i \xi)$. Figure 2(b) plots $S(\omega)$ versus $\arg(g_{12}/\gamma_{12})$ for $|g_{12}| = |\gamma_{12}|/2$. It reveals that, for certain phase differences, an extremely narrow peak emerges that completely dominates the spectral properties of the system. We term the configuration yielding this sharp spectral feature as \textit{quasi-chiral}, which smoothly evolves into the drastically lower and broader single-peaked spectrum characteristic of the \textit{reciprocal} \argarglit{arg}(g_{12}/\gamma_{12}) = 0, \pi, 2\pi] and \textit{chiral} \argarglit{arg}(g_{12}/\gamma_{12}) = \pi/2, 3\pi/2] regimes. Figures 2(c) and (d) analyze the sensitivity of the quasi-chiral peak to $|g_{12}|/|\gamma_{12}|$ and $|\gamma_{12}|/|\gamma_{11}|$, respectively. The grey solid spectrum in both panels is a cut along the white dot-dashed line in panel (b) $|argarglit{arg}(g_{12}/\gamma_{12})| = 5\pi/4$. Upon increasing the EM coupling strength, and thus away from the chiral condition, we find a remarkable robustness in the spectrum, which still presents a (slightly blue-shifted) prominent peak for $|g_{12}| = |\gamma_{12}|$ [panel (c)]. On the contrary, by decreasing the dissipative coupling $S(\omega)$ flattens, which indicates the crucial role that the $\beta$-factor plays in the emergence of the quasi-chiral phenomenology [panel (d)].

We now analyze in more detail the emission spectrum of the two coupled QEs. For that purpose, we study the three different EM coupling configurations marked by horizontal lines in Fig. 2(b): reciprocal \argarglit{arg}(g_{12}/\gamma_{12}) = \pi], quasi-chiral \[5\pi/4]\], and chiral \[5\pi/4]\]. The corresponding emission spectra are shown as the solid grey lines in Fig. 3 (a), (b) and (c), respectively (note the different vertical scales). The panels also display the decomposition $S(\omega) = \sum_{i=1}^{4} S_i(\omega)$ \[10, 12\], where the contributions $S_i(\omega)$ (non-solid color lines) arise from the four transitions that can take place in the system. They are illustrated by the diamond level scheme in the inset of Fig. 3(b), with vertices at frequencies \{0, $\omega_{rad}$, $\omega_{sub}$, $2\omega_0$\}. Note that

![Figure 2](image1.png)

![Figure 3](image2.png)
\( \omega_{\text{rad}} \) and \( \omega_{\text{sub}} \) result from the diagonalization of the Liouvillian \( \mathcal{L} \) and, therefore, vary with the parameters. We can observe that the reciprocally coupled spectrum [panel (a)] is governed by a single peak centered at \( \omega_{\text{rad}} \approx \omega_0 - |g_{12}| = \omega_0 - |\gamma_{12}|/2 \) which originates from the transition from the radiative to the ground state (green dashed line). Note that \( \arg(g_{12}/\gamma_{12}) = \pi \) in this case, which yields \( \omega_{\text{rad}} < \omega_0 \) [12].

The chiral system [panel (c)] presents a single, broader maximum which emerges due to the spectral overlapping of the emission of the two single-excitation states (\( \omega_{\text{rad}} = \omega_{\text{sub}} = \omega_0 \)). The two QEs are only weakly coupled and \( S(\omega) \) resembles the single QE spectrum. Finally, we find that, surprisingly, the sharp peak in the quasi-chiral emission [panel (b)] originates from the decay of the subradiant state, with \( \omega_{\text{sub}} \approx \omega_0 + 0.4|\gamma_{12}| \). Note that a second, much lower and broader maximum occurs at \( \omega_{\text{rad}} \approx \omega_0 - 0.4|\gamma_{12}| \).

To examine the causes of the evolution of the emission spectrum with \( \arg(g_{12}/\gamma_{12}) \), we plot in the right column of Fig. 3 the resonant frequencies \( \omega_i \) (d), the decay rates \( \Gamma_i \) (e), and the weights \( W_i \) (f) for the four \( S_i(\omega) \) contributions [10, 12]. We can observe that by varying the phase difference between coherent and dissipative couplings from the reciprocal to the chiral configuration, the frequency and decay rate of the radiative and subradiant states merge into the single QE values: \( \omega_0 \) and \( \gamma_{12} = \gamma_{11} \). In the evolution, the weight of the subradiant contribution increases faster than its linewidth, which gives rise to the formation, narrowing and blue-shifting of the quasi-chiral spectral maximum shown in Fig. 2(b).

In order to access the emission dynamics of the coupled QEs at a deeper level, we investigate the two-photon correlations in frequency, via the theory of two-photon spectra [24]. Using the formalism of Ref. [25], we compute the two-photon spectrum \( g_{12}^{(2)}(\omega_1, \omega_{11}) \) of our system (for photons detected at the frequencies \( \omega_1 \) and \( \omega_{11} \)). We assume zero time delay and access the correlations via detectors with spectral width \( \Omega = |\gamma_{12}|/5 \). We plot in Fig. 4 (a, b, c) spectra for the same \( \arg(g_{12}/\gamma_{12}) \) as in Fig. 3 (a, b, c). In the three panels, a dashed thick line marks the subradiant peak position and the dot-dashed thin one marks the radiative frequency. The reciprocal system (a) broadly demonstrates bunching [red regions, \( g_{12}^{(2)} > 1 \)], with little fine structure. However, upon evolving through the quasi-chiral configuration (b) towards the chiral limit (c), a remarkable butterfly structure [25] emerges due to the appearance of bands of antibunching [blue regions, \( g_{12}^{(2)} < 1 \)] related to the subradiant state. In panel (d), we map the autocorrelation function \( g_{11}^{(2)}(\omega_1 = \omega_{11} = \omega) \) versus the phase difference between coherent and dissipative coupling strengths. The close resemblance to the one-photon spectrum of Fig. 2(b) is immediately apparent. Importantly, by simple inspection we can conclude that the minimum of \( g_{11}^{(2)} \) overlaps almost exactly with the subradiant peak position. Most notably, the global minimum in the correlation function does not occur at the chiral, non-reciprocal conditions but when the two QEs are quasi-chirally coupled, further highlighting the singular optical properties the emerge in the quasi-chiral regime.

To conclude, we have investigated the emergence of chirality in the interactions between two quantum emitters. Exploring the interplay between coherent and dissipative mechanisms, we find naturally the non-reciprocal coupling configuration, which enables us to identify the physical conditions for its occurrence. Through analytical and numerical electromagnetic calculations, we have shown that tunable chiral interactions can be realized at a nanoscale platform consisting of two circularly-polarized emitters held above a metal surface. Finally, we have unveiled the rich quantum optical properties of the quasi-chiral regime, in which the conditions for non-reciprocity are only partially met. We have shown that the subradiant state of the system governs its one- and two-photon spectra, which gives rise to sharp spectral features and strong photon correlations. Our findings set the theoretical grounds and provide guidance towards the development and optimization of quantum optical functionalities associated with the fine tuning between coherent and dissipative light-matter interactions, beyond the highly stringent chiral regime.

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