

Addressing the Correlation of Stokes-Shifted Photons Emitted from Two Quantum Emitters

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In resonance fluorescence excitation experiments, light emitted from solid-state quantum emitters is typically filtered to eliminate the laser photons, ensuring that only red-shifted Stokes photons are detected. However, theoretical analyses of the fluorescence intensity correlation often model emitters as two-level systems, focusing on light emitted exclusively from the purely electronic transition (the zero-phonon line), or they rely on statistical approaches based on conditional probabilities that neglect the quantum coherence between the emitters and the coherence between the electric fields they generate. Here, we propose a model to characterize the correlation of either zero-phonon line photons or Stokes-shifted photons. This model successfully reproduces the experimental correlation of Stokes-shifted photons emitted from two interacting molecules and predicts that this correlation is affected by quantum coherence. Besides, we analyze the role of quantum coherence in the Stokes-shifted emission from two distant emitters, showing a sharp peak at zero time delay due to the Hanbury Brown–Twiss effect.

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Theoretical descriptions of light emission from quantum emitters typically focus on the case of simple two-level systems (TLSs) [1–12]. However, many realistic systems—such as solid-state emitters, trapped ions, or trapped atoms—exhibit vibrational degrees of freedom that couple to the electronic states. This coupling gives rise to two main emission channels: the zero-phonon line (ZPL), corresponding to a direct transition between the electronic excited and ground states, and phonon sidebands, which involve transitions to vibrationally excited ground states and appear red-shifted in the emission spectrum. Accurate modeling of these emission processes, particularly in systems involving two coherently interacting quantum emitters, is necessary for the advancement of quantum photonics. For example, these coupled systems can find applications in quantum information [13–17], quantum-state engineering [18], and the generation of entangled photons [19].

Moreover, in usual resonance fluorescence experiments, the laser is tuned to the ZPL transition, but only the red-

shifted (Stokes) photons are detected, minimizing contamination from scattered laser light [20–23]. Under these conditions, the standard TLS framework proves insufficient—particularly for experiments involving emission from interacting solid-state emitters [24–26]. To overcome this problem, the correlation of Stokes-shifted photons can be described using a conditional-probability approach, as introduced in Refs. [24,25]. This approach is based on the calculation of the conditional probabilities of a fluorescence photon being emitted by the system at time $t + \tau$ following an earlier emission at time t that projects the state of the system [27–29]. However, this approach does not account for the influence of the quantum coherence between the two emitters nor the quantum coherence of the electric field that they emit (see Supplemental Material [30]). Thus, this approach can fail to describe experiments where coherence becomes important, as indicated by our results here.

In this Letter, we present a model that incorporates quantum coherence effects to describe the photon statistics of both zero-phonon line and Stokes-shifted emissions from quantum emitters, as illustrated in Fig. 1(a). We apply this model to analyze the emission from two interacting organic molecules at cryogenic temperature, revealing that

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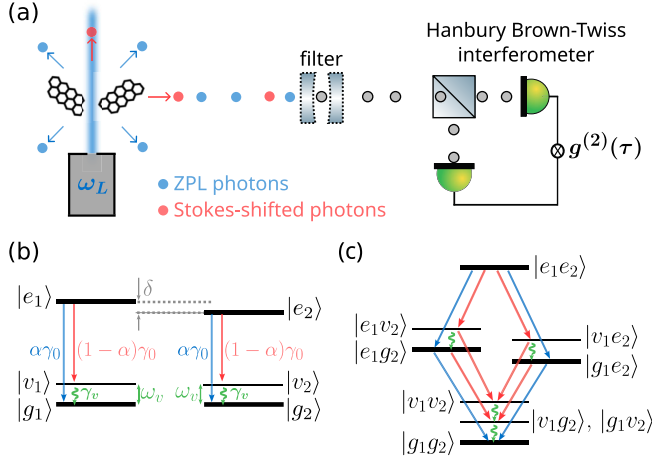


FIG. 1. (a) Schematic representation of light emitted from two interacting emitters, the filtering of this light, and the measurement of the intensity correlation $g^{(2)}(\tau)$. A laser beam at frequency ω_L (in blue) excites resonantly the pure electronic excited state of the quantum emitters, which then can emit a photon at the same frequency (blue circles) or a Stokes-shifted photon (red circles). A filter selects either the ZPL or Stokes-shifted light (gray circles represent these filtered photons). (b),(c) Energy levels and relaxation processes of two noninteracting emitters represented in the uncoupled local basis using the single-emitter representation in (b) and the two-emitter representation in (c). Blue arrows correspond to ZPL transitions and red arrows to Stokes-shifted transitions.

the statistics of ZPL and Stokes-shifted photons can differ significantly. We further explore the case of two distant, noninteracting emitters. In this regime, the model predicts a sharp feature at zero time delay due to the Hanbury Brown–Twiss effect, and we discuss the conditions necessary for its experimental observation.

We consider two almost identical quantum emitters (indexed by $j = 1, 2$), with a pure (0-phonon) electronic ground state $|g_j\rangle$ and a pure excited state $|e_j\rangle$. The transition dipole moment between these two states is denoted by μ_j and the transition frequency by ω_j . The detuning between the two transition frequencies is $\delta = \omega_1 - \omega_2$, with $\delta \ll \omega_1, \omega_2$. We consider an additional state $|v_j\rangle$ that corresponds to the 1-phonon state of a vibrational mode of frequency ω_v in the electronic ground state; see Figs. 1(b) and 1(c). In the rotating frame at the laser frequency ω_L , the unperturbed Hamiltonian of the system can be written as

$$\hat{H}_0 = \hbar \sum_{j=1}^2 \left[\frac{\Delta_j}{2} (|e_j\rangle\langle e_j| - |g_j\rangle\langle g_j|) + \frac{2\omega_v - \Delta_j}{2} |v_j\rangle\langle v_j| \right], \quad (1)$$

with $\Delta_j = \omega_j - \omega_L$. Further, the coherent dipole-dipole interaction between the two emitters is described by $\hat{H}_I = \hbar V (\hat{\sigma}_1^\dagger \hat{\sigma}_2 + \hat{\sigma}_1 \hat{\sigma}_2^\dagger)$, with V the coupling strength [12,30,31] and $\hat{\sigma}_j = |g_j\rangle\langle e_j|$ and $\hat{\sigma}_j^\dagger$ the ZPL lowering

and raising operators of emitter j , respectively. This interaction Hamiltonian considers that the emitters couple only through the purely electronic states. The coupling through the 1-phonon levels is not taken into account because it does not affect the dynamics of the emitters nor their light emission, as we consider short-lived vibrations. The total Hamiltonian is, thus, $\hat{H} = \hat{H}_0 + \hat{H}_I + \hat{H}_P$, where $\hat{H}_P = -(\hbar/2) \sum_{j=1}^2 (\Omega_j \hat{\sigma}_j^\dagger + \Omega_j^* \hat{\sigma}_j)$ is the pumping Hamiltonian. Here, $\Omega_j = \mu_j \cdot \mathcal{E}_j e^{ik_L \cdot r_j} / \hbar$ is the Rabi frequency, with k_L the laser wave vector and $\mathcal{E}_j$ the laser electric field at the position r_j of emitter j .

The state of the interacting emitters can be described by the density matrix $\hat{\rho}$, whose dynamics is governed by the Markovian master equation,

$$\begin{aligned} \frac{d}{dt} \hat{\rho} = & -\frac{i}{\hbar} [\hat{H}, \hat{\rho}] + \sum_{j=1}^2 \left(\frac{\alpha \gamma_0}{2} \mathcal{D}[\hat{\sigma}_j] + \frac{(1-\alpha)\gamma_0}{2} \mathcal{D}[|v_j\rangle\langle e_j|] \right. \\ & \left. + \sum_{k \neq j} \frac{\gamma_{jk}}{2} \mathcal{D}[\hat{\sigma}_j, \hat{\sigma}_k] + \frac{\gamma_v}{2} \mathcal{D}[|g_j\rangle\langle v_j|] \right) \hat{\rho}, \end{aligned} \quad (2)$$

where we have introduced the dissipators $\mathcal{D}[\hat{A}, \hat{B}] \hat{\rho} = 2\hat{A} \hat{\rho} \hat{B}^\dagger - \hat{B}^\dagger \hat{A} \hat{\rho} - \hat{\rho} \hat{B}^\dagger \hat{A}$ and $\mathcal{D}[\hat{A}] = \mathcal{D}[\hat{A}, \hat{A}]$ [36]. Here, α is the combined Debye-Waller/Franck-Condon factor, given by the fraction of photons emitted in the ZPL line [12,25,30,37], and γ_0 is the total decay rate from the excited state $|e_j\rangle$. Thus, we have fixed $\alpha \gamma_0$ as the decay rate in the ZPL and $(1-\alpha)\gamma_0$ as the decay rate into the 1-phonon state. γ_{jk} is the crossed-decay rate, including the effect of α [30], and γ_v is the vibrational decay rate, which we consider to be much larger than V .

Assuming that the transition dipole moments of both emitters are identical ($\mu_1 = \mu_2$), the intensity correlation is given by [32,33,38,39]

$$g^{(2)}(\tau) = \frac{\langle \hat{E}^{(-)}(0) \hat{E}^{(-)}(\tau) \hat{E}^{(+)}(\tau) \hat{E}^{(+)}(0) \rangle_{ss}}{\langle \hat{E}^{(-)}(0) \hat{E}^{(+)}(0) \rangle_{ss}^2}, \quad (3)$$

where $\hat{E}^{(+)}(\tau)$ and $\hat{E}^{(-)}(\tau)$ are the positive-frequency and negative-frequency electric field operators in the Heisenberg picture, and $\langle \hat{A} \rangle_{ss} = \text{Tr}(\hat{A} \hat{\rho}_{ss})$ is the expected value of $\hat{A}$ at the steady state $\hat{\rho}_{ss}$. The experimental filtering is introduced through the proper definition of the electric field operators. The correlation of ZPL photons can be calculated using $\hat{E}_{\text{ZPL}}^{(+)}(t)/\xi_{\text{ZPL}} = \hat{\sigma}_1(t) + e^{i\phi_{\text{ZPL}}} \hat{\sigma}_2(t)$ in Eq. (3), whereas the correlation of Stokes-shifted photons can be obtained through $\hat{E}_{\text{St}}^{(+)}(t)/\xi_{\text{St}} = |v_1\rangle\langle e_1|(t) + e^{i\phi_{\text{St}}} |v_2\rangle\langle e_2|(t)$. Here, ξ_{ZPL} and ξ_{St} are proportionality constants that do not affect the intensity correlation and depend on the direction of the dipoles, the direction of detection $\hat{k}_d$, and the mean frequency of emission [i.e., $\omega_0 = (\omega_1 + \omega_2)/2$ in the case of ξ_{ZPL} and $\omega_0 - \omega_v$ in the

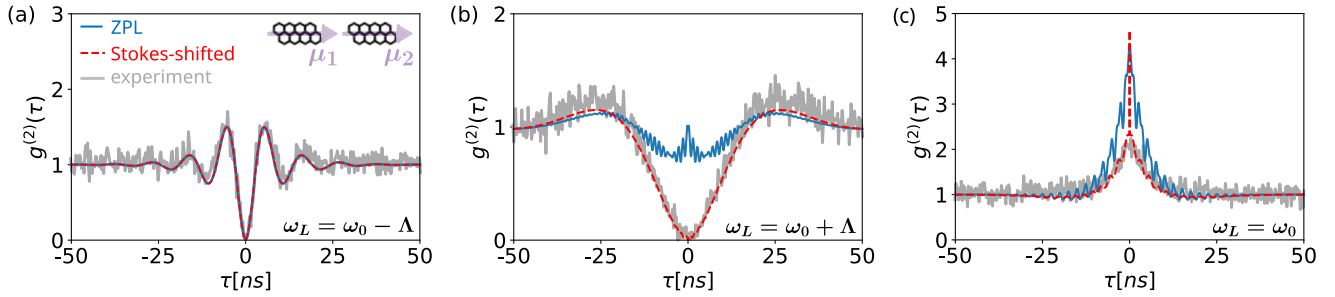


FIG. 2. Comparison of the correlation of ZPL photons and Stokes-shifted photons emitted from two strongly interacting dibenzanthanthrene (DBATT) molecules. The molecules are in a J-aggregate configuration, as depicted in the inset in (a), and have $1/\gamma_0 = 7.4$ ns. The laser is tuned resonantly to the (a) superradiant state $|\Lambda_- \rangle$ ($\omega_L = \omega_0 - \Lambda$), (b) subradiant state $|\Lambda_+ \rangle$ ($\omega_L = \omega_0 + \Lambda$), and (c) two-photon resonance ($\omega_L = \omega_0$). The simulated intensity correlation $g^{(2)}(\tau)$ of (solid blue line) ZPL and (dashed red line) Stokes-shifted light are plotted as a function of time delay τ . Solid gray line corresponds to the experimental results reported in Ref. [25]. All the parameters are specified in Supplemental Material [30].

case of ξ_{St} [1,6,8]. Additionally, $\phi_{\text{ZPL}} = -(n\omega_0/c)\hat{\mathbf{k}}_d \cdot \mathbf{r}_{12}$ and $\phi_{\text{St}} = -[(\omega_0 - \omega_v)/c]\hat{\mathbf{k}}_d n \cdot \mathbf{r}_{12}$ [2,40], with n the refractive index of the host medium, c the speed of light in vacuum, and $\mathbf{r}_{12} = \mathbf{r}_2 - \mathbf{r}_1$.

We demonstrate next that the correlation of ZPL photons and of Stokes-shifted photons can be drastically different. With this purpose, we simulate the experimental configuration in Ref. [25], which measures the statistics of the Stokes-shifted photons emitted from two strongly interacting DBATT molecules (in a J-aggregate configuration [30]) in the normal direction to $\boldsymbol{\mu}_j$ and $\mathbf{r}_{12}$ (i.e., $\phi_{\text{ZPL}} = \phi_{\text{St}} = 0$). The mean frequency ω_0 of these molecules corresponds to a vacuum wavelength of 618 nm. For simulations, we consider the DBATT vibrational mode $\hbar\omega_v = 31.86$ meV [34], and $1/\gamma_v = 10$ ps based on experiments in Ref. [41]. At $\tau \gg 1/\gamma_v$, the simulations are not affected by the value of ω_v and γ_v nor by including a larger number of vibrational modes (see Supplemental Material [30]). Other molecular parameters (including V , γ_{12} , and δ) are extracted from independent measurements of the excitation spectra [30], so that no fitting parameters are considered.

We plot in Fig. 2(a) the intensity correlation $g^{(2)}(\tau)$ when the laser is tuned resonantly to the superradiant state, which in the J-aggregate configuration corresponds to the hybrid state $|\Lambda_- \rangle = -\sin\theta|g_1e_2\rangle + \cos\theta|e_1g_2\rangle$ [12], with transition frequency $\omega_0 - \Lambda$. Here, $\tan(2\theta) = -2V/\delta$ and $\Lambda = \sqrt{V^2 + (\delta/2)^2}$. We find that the intensity correlation of Stokes-shifted photons (dashed red line) is almost identical to the correlation of ZPL photons (solid blue line), both exhibiting antibunching and Rabi oscillations. This effective TLS behavior arises because the excitation is sufficiently weak and the laser is far detuned from both the two-photon transition and the transition to the antisymmetric state, thereby isolating the dynamics of the superradiant state [12]. The intensity correlation obtained experimentally in Ref. [25] (solid gray line) is well reproduced by the two simulations. On the other hand, Fig. 2(b) shows the intensity correlation when the laser is tuned resonantly to the

subradiant state $|\Lambda_+ \rangle = \cos\theta|g_1e_2\rangle + \sin\theta|e_1g_2\rangle$, with transition frequency $\omega_0 + \Lambda$. In this regime, the simulated correlations for Stokes-shifted and ZPL photons are significantly different. The Stokes-shifted photons again show antibunching, Rabi oscillations, and excellent agreement with the experiment. In contrast, the ZPL photon correlation also displays clear Rabi oscillations, but with $g^{(2)}(0) \approx 1$ and an additional faster oscillation at frequency 2Λ , indicating interference between the superradiant and subradiant pathways [12]. Such interference does not affect the Stokes-shifted photon correlation, since the emitters couple only through the ZPL (see Supplemental Material [30]).

Further, we show in Fig. 2(c) the intensity correlation when the laser is tuned to the two-photon resonance ($\omega_L = \omega_0$) corresponding to half the frequency between $|g_1g_2\rangle$ and $|e_1e_2\rangle$. In this case, both the correlations of ZPL photons and of Stokes-shifted photons are bunched, as this laser frequency enables the resonant excitation of the doubly excited state $|e_1e_2\rangle$, strongly increasing the probability of emitting photons in cascade [12,28]. Additionally, the two correlations exhibit oscillations of frequency Λ , corresponding to the detuning between the laser frequency and the transition frequency of the superradiant state. However, the ZPL correlation exhibits more pronounced oscillations and does not capture well the experimental measurements, whereas the Stokes-shifted correlation reproduces them very well. These results reveal that the correlation of Stokes-shifted photons and of ZPL photons emitted from two strongly interacting quantum emitters can be very different, and they emphasize the importance of an accurate description of each experimental configuration.

Next, we investigate the role of quantum coherence in the correlation of Stokes-shifted photons emitted from two quantum emitters. The off-diagonal elements of the electric field intensity operator expressed in the uncoupled basis can be associated with the first-order coherence of the electric field emitted [30,35], and the coherence between the emitters is encoded in the off-diagonal elements of the

density matrix [42]. For this analysis, we first substitute $\hat{E}_{\text{St}}^{(+)}(t)/\xi_{\text{St}} = |v_1\rangle\langle e_1|(t) + |v_2\rangle\langle e_2|(t)$ (where $\phi_{\text{St}} = 0$) into the general expression of the intensity correlation in Eq. (3). After some algebra (see Supplemental Material [30]), we obtain that the Stokes-shifted correlation is the sum of three contributions:

$$g_{\text{St}}^{(2)}(\tau) = \frac{G_d^{(2)}(\tau)}{\langle \hat{I}_{\text{St}}(0) \rangle_{ss}^2} + \frac{G_{\text{coh},I}^{(2)}(\tau)}{\langle \hat{I}_{\text{St}}(0) \rangle_{ss}^2} + \frac{G_{\text{coh},p}^{(2)}(\tau)}{\langle \hat{I}_{\text{St}}(0) \rangle_{ss}^2}. \quad (4)$$

We first consider the denominator, where we have introduced the operator $\hat{I}_{\text{St}}(\tau) = \hat{E}_{\text{St}}^{(-)}(\tau)\hat{E}_{\text{St}}^{(+)}(\tau)$ (which is proportional to the fluorescence intensity) with the steady-state expected value

$$\frac{\langle \hat{I}_{\text{St}}(0) \rangle_{ss}}{|\xi_{\text{St}}|^2} = 2p_{ee} + p_{eg} + p_{ge} + p_{ev} + p_{ve} + 2\text{Re}p_{ev,ve}. \quad (5)$$

Here, $p_{ab} = \langle a_1 b_2 | \hat{\rho}_{ss} | a_1 b_2 \rangle$ is the population of the state $|a_1 b_2\rangle$, and $p_{ev,ve} = \langle e_1 v_2 | \hat{\rho}_{ss} | v_1 e_2 \rangle$ is an off-diagonal element of the density matrix, which vanishes as long as the emitters couple with each other only through the pure electronic states. The use of the uncoupled local basis $|a_1 b_2\rangle$ (with $a_j, b_j \in \{e_j, g_j, v_j\}$) is convenient because the emission of Stokes-shifted photons projects the system into a localized state of this basis [29].

Further, we have decomposed the correlation in Eq. (4) into three different contributions. The first contribution involves only diagonal elements of $\hat{\rho}_{ss}$ and $\hat{I}_{\text{St}}(\tau)$, and its numerator is given by

$$\begin{aligned} \frac{G_d^{(2)}(\tau)}{|\xi_{\text{St}}|^4} = & p_{ee}[\langle v_1 e_2 | \hat{I}_{\text{St}}(\tau) | v_1 e_2 \rangle + \langle e_1 v_2 | \hat{I}_{\text{St}}(\tau) | e_1 v_2 \rangle] \\ & + p_{eg}\langle v_1 g_2 | \hat{I}_{\text{St}}(\tau) | v_1 g_2 \rangle + p_{ge}\langle g_1 v_2 | \hat{I}_{\text{St}}(\tau) | g_1 v_2 \rangle \\ & + p_{ev}\langle v_1 v_2 | \hat{I}_{\text{St}}(\tau) | v_1 v_2 \rangle + p_{ve}\langle v_1 v_2 | \hat{I}_{\text{St}}(\tau) | v_1 v_2 \rangle. \end{aligned} \quad (6)$$

The different terms in this expression are related to the six different decay paths leading to the emission of Stokes-shifted photons [red arrows in Fig. 1(c)]. The contribution $g_{\text{St},d}^{(2)}(\tau) = G_d^{(2)}(\tau)/\langle \hat{I}_{\text{St}}(0) \rangle_{ss}^2$ to the intensity correlation in Eq. (4) is independent of quantum coherence, and it can be approximated by the correlation obtained with the conditional-probability approach [30]. Moreover,

$$\frac{G_{\text{coh},I}^{(2)}(\tau)}{|\xi_{\text{St}}|^4} = p_{ee}[\langle v_1 e_2 | \hat{I}_{\text{St}}(\tau) | e_1 v_2 \rangle + \langle e_1 v_2 | \hat{I}_{\text{St}}(\tau) | v_1 e_2 \rangle] \quad (7)$$

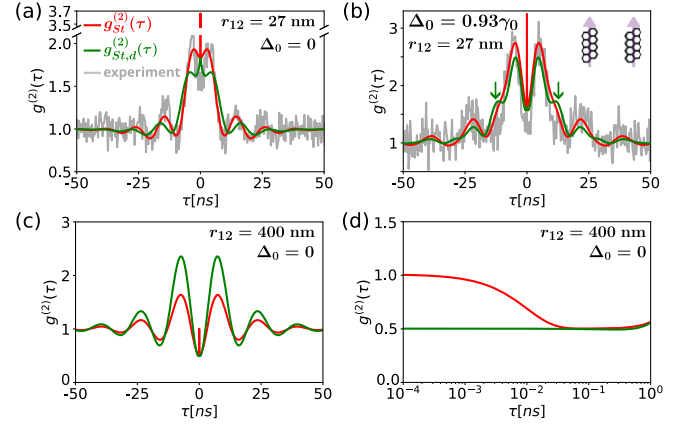


FIG. 3. Impact of quantum coherence in the correlation of Stokes-shifted photons emitted from two DBATT molecules. The molecules are in an H-aggregate configuration, as depicted in the inset in (b), and have $1/\gamma_0 = 7.4$ ns. We consider $r_{12} = 27$ nm in (a),(b) and $r_{12} = 400$ nm in (c),(d). The laser is tuned to the two-photon resonance $\Delta_0 = 0$ in all panels (with $\Delta_0 = \omega_0 - \omega_L$), except in (b), where $\Delta_0 = 0.93\gamma_0$. Red lines correspond to the simulation using the full model including quantum coherence in the emission, whereas green lines correspond to the simulation neglecting this coherence. Gray lines in (a),(b) correspond to experimental measurements. The rest of the parameters are specified in Supplemental Material [30].

is proportional to the population p_{ee} and to the off-diagonal elements $\langle v_1 e_2 | \hat{I}_{\text{St}}(\tau) | e_1 v_2 \rangle$ and $\langle e_1 v_2 | \hat{I}_{\text{St}}(\tau) | v_1 e_2 \rangle$ of the intensity operator $\hat{I}_{\text{St}}(\tau)$. $G_{\text{coh},I}^{(2)}(\tau)$ can be interpreted as the quantum interference between the emission paths of the two molecules (i.e., the Hanbury Brown–Twiss effect [43–45]). Last, $G_{\text{coh},p}^{(2)}(\tau)$ includes all terms involving off-diagonal elements of the density matrix [30] and, thus, the coherence between the emitters.

We first analyze the role of quantum coherence in the case of two DBATT molecules separated by a short distance ($r_{12} = 27$ nm). To this aim, we compare new experimental measurements of the correlation of Stokes-shifted photons emitted from a molecular H-aggregate configuration (the experimental setup in Supplemental Material [30]) with the simulated intensity correlations obtained with the complete model [Eq. (4)], as well as with the correlation $g_{\text{St},d}^{(2)}(\tau) = G_d^{(2)}(\tau)/\langle \hat{I}_{\text{St}}(0) \rangle_{ss}^2$, obtained by neglecting the role of quantum coherence in the emission. In particular, we plot in Figs. 3(a) and 3(b) the correlation of Stokes-shifted photons for this molecular pair when the laser is tuned to $\omega_L = \omega_0$ (resonantly to the two-photon resonance) and to $\omega_L = \omega_0 - 0.93\gamma_0$ (slightly detuned from the two-photon resonance), respectively. The rest of the parameters are specified in Supplemental Material [30]. The simulated $g_{\text{St}}^{(2)}(\tau)$ (red lines) shows a good agreement with the experimental measurements (gray lines) and appreciable differences from $g_{\text{St},d}^{(2)}(\tau)$ (green line). Specifically, if coherences

are neglected in the emission, we observe that (i) the amplitude of the oscillations is notably modified, and (ii) a bump emerges at $\tau \approx \pm 10$ ns in Fig. 3(b) (see the green arrows), which is not measured in experiments. These two differences are due to $G_{coh,p}^{(2)}(\tau)$. Furthermore, $g_{St}^{(2)}(\tau)$ exhibits an extremely narrow peak at $\tau = 0$, with width comparable to the vibrational lifetime, which is due to $G_{coh,l}^{(2)}(\tau)$ and is not resolved experimentally [30]. In Supplemental Material [30], we provide additional measurements from different molecular pairs, which also provide good agreement with the simulations obtained with the complete model.

Finally, we analyze the correlation of Stokes-shifted photons emitted from two distant emitters and find that quantum coherence again plays an important role. We plot in Figs. 3(c) and 3(d) the simulated $g_{St}^{(2)}(\tau)$ (red lines) and $g_{St,d}^{(2)}(\tau)$ (green lines) for two detuned emitters ($\delta = 5\gamma_0$) separated by $r_{12} = 400$ nm (which yields a negligible dipole-dipole coupling) and $\omega_L = \omega_0$. The rest of the parameters are specified in Supplemental Material [30]. Figure 3(c) shows that both simulations exhibit oscillations at the generalized Rabi frequency [33], which at this laser frequency is equal to $\sqrt{(\delta/2)^2 + |\Omega|^2}$. The amplitude of these oscillations is significantly affected by quantum coherence, specifically, by $G_{coh,p}^{(2)}(\tau)$. Further, the complete model yields $g_{St}^{(2)}(\tau = 0) = 1$, with a fast decay to 0.5 in the timescale of the vibrational lifetime [tens of picoseconds, see Fig. 3(d)], which is a consequence of the Hanbury Brown–Twiss effect between the Stokes-shifted light emitted from the two molecules [30]. This fast decay is due to the loss of the initial coherence encoded in $G_{coh,l}^{(2)}(\tau)$ and is attributed to the influence of the internal vibrations of the emitters acting as a dephasing channel. This behavior of the Stokes-shifted correlation from two distant emitters is analogous to that of the ZPL correlation from two uncorrelated, equally pumped emitters [46], as it occurs in the limit of strong driving or large dephasing [50], for which available experiments have reported, as far as we know, $g^{(2)}(\tau = 0) \approx 0.5$ [51–54], in contrast with theoretical analysis that predicts a value of 1 [47–50,55,56]. Our analysis thus clarifies how this discrepancy is due to an insufficient time resolution of the detectors in the experiments.

In summary, we have presented a model to address the correlation of Stokes-shifted photons emitted from quantum emitters as well as that of the ZPL photons. We have shown an excellent agreement between the simulations obtained using this model and experimental measurements of the correlation of the Stokes-shifted photons emitted from two interacting DBATT molecules (using experimental data from Ref. [25] as well as new experimental measurements). Additionally, we have revealed that the intensity correlation of ZPL photons can exhibit significant differences with respect to the intensity correlation of

Stokes-shifted photons, depending on the molecular and laser parameters. Furthermore, we have provided evidence that quantum coherence can impact the emission of Stokes-shifted photons when the emitters are interacting and also when they do not interact. Finally, we have found that detectors with time resolutions smaller than the lifetime of the vibrations are needed to measure $g^{(2)}(\tau = 0) = 1$ in experiments on Stokes-shifted emission from two resonantly driven distant emitters.

Therefore, this Letter provides a foundation for extending the study of photon correlations emitted by systems with interacting quantum emitters, particularly in platforms that involve both electronic and vibrational states. Examples include atoms and ions in optical, magnetic, or electric traps as well as solid-state systems like quantum dots coupled to phonons and defect centers in diamonds, carbon nanotubes, and two-dimensional materials. This framework can also be generalized to ensembles with more than two coupled emitters [30], enabling the exploration of collective quantum phenomena in larger emitter networks.

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Data availability—The data that support the findings of this article are openly available [57].

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