Letter

Nonradiant multiphoton states in quantum ring oligomers

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(Received 3 October 2023; accepted 7 June 2024; published 9 July 2024)

Arrays of coupled dipole emitters support collective single- and multiphoton states that can preserve quantum excitations. One of the crucial characteristics of these states is the lifetime, which is fundamentally limited due to spontaneous emission. Here, we present a mechanism of the external coupling of two states via the radiation continuum, which allows for an increase in the lifetime of both single and double excitations. As an illustrative example, we consider a ringlike ensemble of quantum emitters, demonstrating that upon slight optimization of the structure geometry, one can increase the lifetime of singly and doubly excited states with nonzero orbital momentum by several orders of magnitude. The proposed mechanism of multiphoton excitations lifetime control has a universal nature and might be applied for a wide class of open quantum systems and quantum ensembles besides the particular geometry considered in this Letter.

DOI: 10.1103/PhysRevA.110.L011501

Assembling quantum emitters in ordered systems allows for enhancing the light-matter interaction [1,2] which is crucial for quantum information [3,4], quantum sensing [5,6], and optomechanical [7–10] applications. Due to advanced trapping techniques, emitters can be assembled into the structured arrays in free space [11–14] or the vicinity of nanophotonic structures [15–18], which induces collective effects in single- and multiphoton regimes [19–30]. However, the reliable manipulation of quantum states requires their stability, which can be easily destroyed by the spontaneous emission inevitably present in open quantum systems. In this context, controlling the lifetime of quantum states remains one of the key problems in modern quantum optics.

Fortunately, this problem can be resolved by generating subradiant states characterized by suppressed spontaneous decay. Since the pioneer work of Dicke [31], the emergence of these states has been actively studied both theoretically [32–37] and experimentally [38–42]. Moreover, the spatial ordering of quantum dipole emitters can provide additional control over the lifetime of subradiant states for one-dimensional arrays in free space [26,43–45], near a waveguide [24,46–50], for two-dimensional arrays [51–53], or for single rings [43]. It has been shown that the radiative decay of large systems can be strongly suppressed, following either a polynomial $\propto N^{-\alpha}$ [44,48] or exponential $\propto e^{-\beta N}$ [43] asymptotic dependence on the number of quantum emitters in the array N. However, the suppression of radiative decay in smaller structures consisting of several to tens of emitters requires different approaches.

In this Letter, we demonstrate the feasibility of forming singly and doubly excited subradiant eigenstates in finite dipole ensembles based on the mechanism of external coupling, which was initially proposed by Friedrich and Wintgen [54] for open quantum systems. It has recently garnered significant attention in the field of nanophotonics for highly efficient nonlinear generation [55], lasing in single semiconductor nanostructures [56], achieving a strong nonlinear response [57], and engineering bound states in the continuum of extended periodic structures such as metasurfaces [58,59]. In this work, we demonstrate that this mechanism can also be extended to form doubly excited subradiant states. Our focus is on concentric rings of two-level dipole emitters, which have already attracted significant attention in quantum optics [60–66], owing to their high symmetry and relevance to various natural quantum systems such as organic molecules. The observation of long-lived doubly excited states with nonzero orbital momentum may find utility in quantum information protocols that involve beams with high angular momentum [34,67–69]. The proposed mechanism can be straightforwardly applied to ordered arrays with different symmetries and geometries to extend the lifetime of quantum excitations.

General formalism. Let us consider a double-ring ensemble of N two-level dipole emitters shown in Fig. 1. All emitters are located in the $z = 0$ plane. We also assume that all emitters

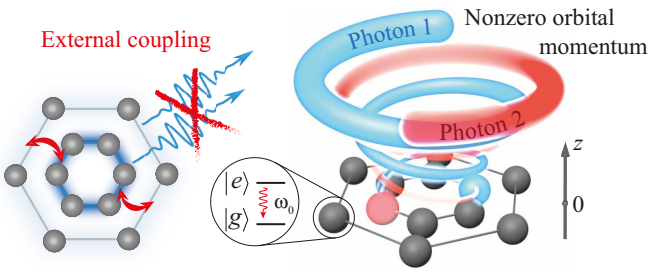

FIG. 1. An open system representing a double-ring oligomer of two-level dipole emitters. The doubly excited eigenstates can have nonzero orbital quasimomentum with radiative losses that can be strongly suppressed via the mechanism of external coupling.

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have a ground state with $J = 0$ and an excited level with $J' = 1$ [37,70–74]. In such a structure, the lifetime of all three Zeeman sublevels ($J'_z = -1, 0, 1$) is the same $\tau_0 = 1/\gamma_0$ with the decay rate $\gamma_0 = k_0^3|\mathbf{d}|^2/(3\pi\hbar\epsilon_0)$, where $k_0 = \omega_0/c = 2\pi/\lambda_0$ is the wave number in vacuum, ω_0 is the transition frequency, and ϵ_0 is the vacuum permittivity. The transition dipole moment of emitters is fixed to be oriented along the z axis, $\mathbf{d} = |\mathbf{d}|\mathbf{e}_z$, which can be achieved by applying an external magnetic field isolating this transition from the two in-plane ones. The emitters are coupled via free-space electromagnetic modes, and their quantum states are governed by the effective non-Hermitian Hamiltonian (further, the Planck constant is set to be $\hbar = 1$) [62] $\widehat{H}_{\text{eff}} = -i\frac{\gamma_0}{2}\sum_{k=1}^{N}\hat{\sigma}_k^\dagger\hat{\sigma}_k + \sum_{k=1}^{N}\sum_{\substack{l=1,\\ l\neq k}}^{N} g(|\mathbf{r}_{kl}|,\omega_0)\hat{\sigma}_k^\dagger\hat{\sigma}_l$, where $\hat{\sigma}_k^\dagger$ ($\hat{\sigma}_k$) is the creation (annihilation) operator for excitation on emitter k, $|\mathbf{r}_{kl}| = |\mathbf{r}_k - \mathbf{r}_l|$ is the relative distance between emitters k and l, and the energy of noninteracting system $\omega_0\sum_{k=1}^{N}\hat{\sigma}_k^\dagger\hat{\sigma}_k$ has been subtracted. The coupling rate between two emitters is defined via the free-space electromagnetic Green's tensor [75] $g(|\mathbf{r}|,\omega_0) = (-3\gamma_0\pi/k_0)\mathbf{e}_z^T\cdot\mathbf{G}_0(\mathbf{r},\omega_0)\cdot\mathbf{e}_z$. Assumption of the Born-Markov approximation $g(|\mathbf{r}|,\omega) \approx g(|\mathbf{r}|,\omega_0)$ allows to avoid the dispersion of the coupling rate since $\gamma_0 \ll \omega_0$.

Ring oligomer. First, we consider an ensemble of N_d emitters arranged in a ring of radius R with the corresponding separation distance between neighbor emitters $a = 2R\sin(\pi/N_d)$. The eigenvalues of the system can be found by substituting the effective Hamiltonian into Schrödinger equation $\widehat{H}_{\text{eff}}|\psi\rangle = \varepsilon|\psi\rangle$ (Sec. S1 in Supplemental Material [76]). Each singly excited eigenstate of the ring can be associated with the orbital quasimomentum m due to the discrete rotational symmetry of the system, $|\psi\rangle = |\psi_{\text{ring}}^{(m)}\rangle$. As an illustrative example, we consider $N_d = 6$ dipoles, for which m can be equal to $0, \pm1, \pm2, 3$ (Sec. S2 [76]). These eigenstates have different parities under the symmetry operations from point group D_{6h} and transform according to the irreducible representations A_{2u}, E_{1g}, E_{2u}, and B_{1g}, respectively (Sec. S4 [76]). As a result, the eigenstates with $\pm m$ are degenerate, i.e., $\varepsilon^{(m)} = \varepsilon^{(-m)}$. In the basis of coupled emitters, the eigenstate with a quasimomentum m reads as $|\psi_{\text{ring}}^{(m)}\rangle = \sum_{k=1}^{N} c_k^{(m)}\hat{\sigma}_k^\dagger|g\rangle^{\otimes N_d}$, where $c_k^{(m)} = e^{im\varphi_k}/\sqrt{N_d}$ is the excitation probability amplitude for emitter k and $\varphi_k = 2\pi(k-1)/N_d$ (see Refs. [60,62] and also Sec. S2 in Supplemental Material [76]).

The radiative losses of a single ring do not exhibit any peculiarities [43,61,62] and the collective decay rate of subradiant eigenstates monotonically decreases with the ring size (Sec. S5 [76]). However, the situation can be drastically changed by adding a single emitter at the ring center, forming the *oligomer* ensemble, as shown in Fig. 2(a). The external coupling between the ring state with $m = 0$ and the central dipole via the radiation continuum leads to the formation of a long-living state. This can be understood by constructing the wave function for the oligomer's eigenstates from two contributions, $|\psi\rangle = c_a|g_{\text{ring}}\rangle\otimes|e_0\rangle + c_b|\psi_{\text{ring}}^{(0)}\rangle\otimes|g_0\rangle$, where $|g_0\rangle$ and $|e_0\rangle$ are the wave functions of the central dipole emitter in the ground and excited states, respectively, and $|g_{\text{ring}}\rangle \equiv |g\rangle^{\otimes N_d}$ is the ground state of the ring. The central emitter can couple only to the ring state with $m = 0$ due to symmetry considerations. The Hamiltonian in such a basis can be

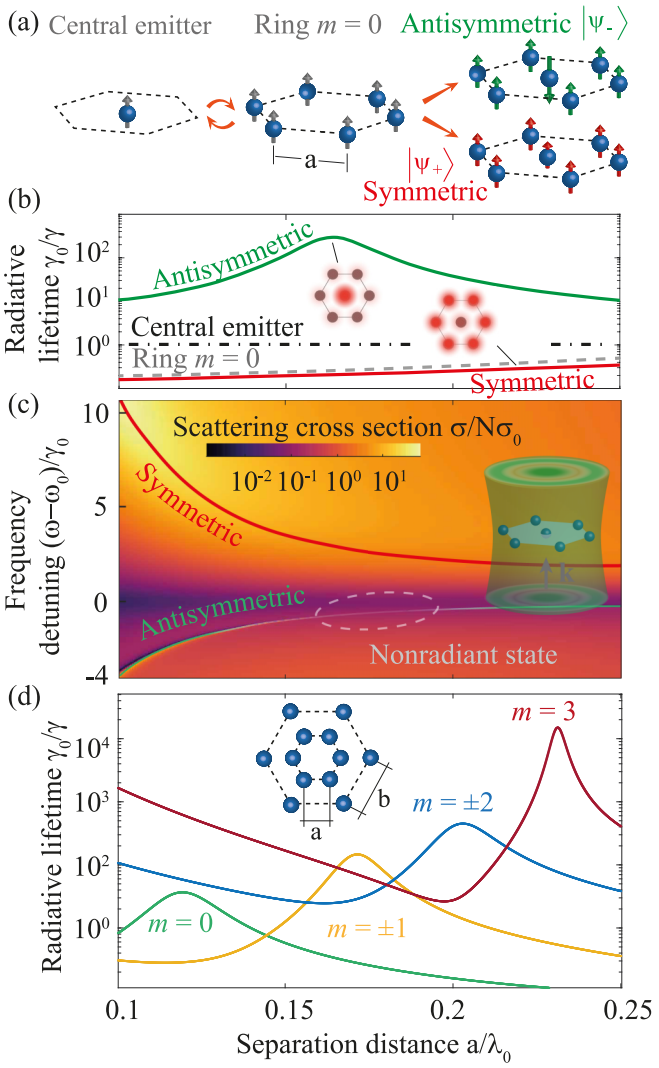

FIG. 2. (a) Schematic representation of the hybridization of ring states with a single emitter's state. (b) Lifetime enhancement for the states in (a). Inset: The excitation probability is shown in red for the oligomer eigenstates. (c) Scattering cross section (color coded) for the ring oligomer excited with the Bessel beam (inset). The red and green lines represent the eigenfrequencies of the oligomer eigenstates. (d) Lifetime enhancement for antisymmetric eigenstates with different m forming in two rings with $b/a = 2$ (inset).

represented as a sum of the unperturbed $\widehat{H}_0$ and interaction $\widehat{V}$ parts [63],

$$\widehat{H} = \widehat{H}_0 + \widehat{V} = \begin{pmatrix} \varepsilon_0 & 0 \\ 0 & \varepsilon_{\text{ring}}^{(m=0)} \end{pmatrix} + \begin{pmatrix} 0 & \varkappa \\ \varkappa & 0 \end{pmatrix}, \tag{1}$$

where $\varepsilon_0 = -i\gamma_0/2$ and $\varepsilon_{\text{ring}}^{(m=0)} = -i\gamma_0/2 + \sum_{k=2}^{N_d} g(|\mathbf{r}_{1k}|,\omega_0)$ are the energies of the excited central emitter $|e_0\rangle$ and the excited ring eigenstate $|\psi_{\text{ring}}^{(0)}\rangle$, respectively, while the coupling rate is given by $\varkappa = \sqrt{N_d}g(R,\omega_0)$. Although $\varkappa$ is proportional to $\sqrt{N_d}$, the radius of the ring increases and g decreases with N_d, leading to the overall decrease of the $\varkappa$. Consequently, the external coupling in such an oligomer is strong enough to induce noticeable lifetime modification only for not very large N_d.

As depicted in Fig. 2(a), the interaction between subsystems leads to the appearance of symmetric $|\psi_+\rangle$ and antisymmetric $|\psi_-\rangle$ hybridized eigenstates with $m = 0$. Their decay rates are defined by the imaginary part of eigenenergies $\gamma_\pm = -2\,\mathrm{Im}(\varepsilon_\pm)$ where $\varepsilon_\pm$ are given in Sec. S6 [76]. The calculations reveal that the antisymmetric eigenstate possesses resonantly increased lifetime by a factor of $\tau_-/\tau_0 = \gamma_0/\gamma_- \approx 230$ for the particular separation between emitters $a/\lambda_0 \approx 0.16$ [see Fig. 2(b)]. At this point, the phase shift between the probability amplitudes for the central dipole and the ring reaches π exactly, while the excitation is primarily located at the central emitter [see the inset in Fig. 2(b) and Sec. S6 in Supplemental Material [76]]. Such a suppression of radiation, caused by external coupling between the ring and the central emitter states, resembles the destructive interference between the radial modes in dielectric cylindrical cavities [77,78].

The long-living oligomer state can be excited with tightly focused Bessel beams possessing a longitudinal component of the field [79]. In Fig. 2(c), one can see the scattering cross section (SCS) σ for the oligomer ensembles of different sizes illuminated by a Bessel beam with orbital angular momentum $\ell = 1$ and spin $s = -1$ adding up to the total angular momentum $m = 0$. The SCS of the oligomer is normalized by $N\sigma_0$ where $N = N_d + 1 = 7$ is the total number of emitters in the system, and σ_0 is the SCS for the central emitter (Sec. S7 [76]). One can observe a resonant enhancement of the SCS for the symmetric (superradiant) eigenstate $|\psi_+\rangle$ and a drastic narrowing of the spectral line corresponding to the antisymmetric (subradiant) eigenstate $|\psi_-\rangle$ when approaching the optimal size condition.

The proposed external coupling mechanism can be also applied to control the lifetime of eigenstates with $m \neq 0$. For instance, in the oligomer ensemble consisting of two concentric rings shown in Fig. 1, the symmetry of singly excited states in the isolated inner and outer rings, or $|\psi^{(m)}_{\text{in-ring}}\rangle$ and $|\psi^{(m)}_{\text{out-ring}}\rangle$, is the same. Therefore, the coupling between rings can lead to radiation suppression for arbitrary m. The mechanism of such an interaction can be also described within the framework of two coupled states with the wave function of the coupled system given as $|\psi^{(m)}_{\text{two-ring}}\rangle = c^{(m)}_a|\psi^{(m)}_{\text{in-ring}}\rangle \otimes |g_{\text{out-ring}}\rangle + c^{(m)}_b|g_{\text{in-ring}}\rangle \otimes |\psi^{(m)}_{\text{out-ring}}\rangle$. By scaling the oligomer size with the fixed ratio $b/a = 2$, one can reach the regime of resonantly enhanced lifetime for antisymmetric eigenstates with different m as shown in Fig. 2(d). Additional details on the superradiant symmetric states are provided in Sec. S6 [76].

Doubly excited states. The mechanism of external coupling can also be exploited to doubly excited states. Doubly excited quantum states form a manifold in the Hilbert space with a dimension of $N(N-1)/2$. Due to the symmetry of the wave function and the Pauli principle, they can be expanded over the wave function basis as $|\Psi\rangle = \sum_{k=1}^{N}\sum_{l=k+1}^{N} c_{kl}\hat{\sigma}_k^\dagger\hat{\sigma}_l^\dagger|g\rangle^{\otimes N}$. Importantly, one can characterize doubly excited eigenstates of ring oligomers with orbital quasimomentum m in a similar manner to the singly excited eigenstates. Moreover, a doubly excited eigenstate can be expanded over the products of singly excited eigenstates, $|\Psi^{(m)}\rangle = \sum_{m_1,m_2} v_{m_1,m_2}|\psi^{(m_1)}\rangle|\psi^{(m_2)}\rangle$, with $v_{m_1,m_2} \neq 0$ if $m_1 + m_2 = m \pmod{N_d}$. This condition for the quasimomentum immediately follows from representation theory. For example, a direct product of two wave functions of singly excited states entering E_{1g} ($m_1 = \pm 1$) and E_{2u} ($m_1 = \pm 2$) irreducible representations results in the wave function of the doubly excited state that enters one of the three irreducible representations $E_{1g} \otimes E_{2u} = B_{1u} + B_{2u} + E_{1u}$, with a total $m = 3$ (B_{1u}, B_{2u}), or $m = \pm 1$ (E_{1u}).

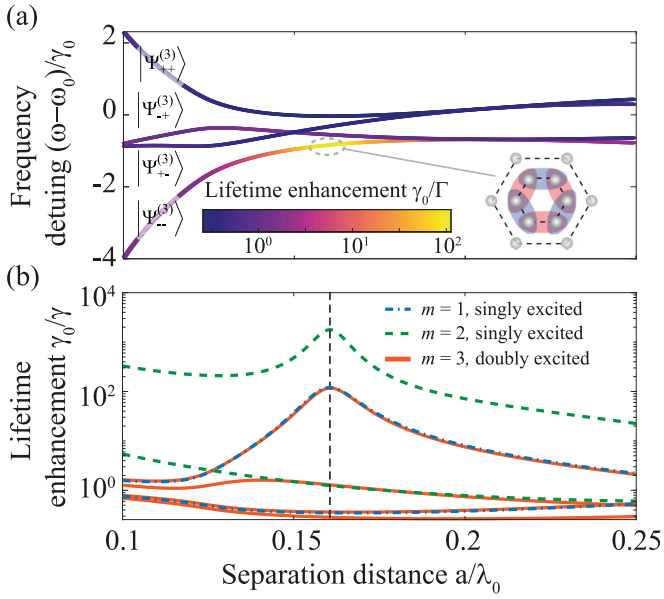

FIG. 3. (a) Eigenfrequency curves for eigenstates (2) where the color corresponds to the lifetime enhancement. The inset schematically shows the largest amplitudes c_{kl} for the nonradiant eigenstate. (b) The lifetime enhancement for singly excited eigenstates with $m = 1$ (blue dashed dotted) and $m = 2$ (green dashed) that form doubly excited eigenstates (2) with $m = 3$ (orange solid).

Two rings of $N_d = 6$ emitters support 61 doubly excited eigenstates including ten ones with the largest orbital quasimomentum $m = 3$ (Sec. S3 [76]). From now on, we will pay special attention to the eigenstates $|\Psi^{(3)}\rangle$ with $m = 3$, which also enter the B_{1u} irreducible representation. The indistinguishability of excitations and the Pauli principle imply that the double-ring oligomer supports only four possible doubly excited eigenstates of this type,

$$|\Psi^{(3)}_{s_1,s_2}\rangle = \frac{i}{2}\big(|\psi^{(+1)}_{s_1}\rangle|\psi^{(+2)}_{s_2}\rangle + |\psi^{(+2)}_{s_2}\rangle|\psi^{(+1)}_{s_1}\rangle - |\psi^{(-1)}_{s_1}\rangle|\psi^{(-2)}_{s_2}\rangle - |\psi^{(-2)}_{s_2}\rangle|\psi^{(-1)}_{s_1}\rangle\big), \quad (2)$$

where $s_1, s_2 = \pm$ correspond to symmetric (antisymmetric) singly excited eigenstates of the double-ring oligomer. We note that the other six doubly excited eigenstates with $m = 3$ have lower lifetimes and correspond to the B_{2u} symmetry since they contain a direct product of the singly excited eigenstates with $m_1 = 0$ (A_{2u} representation) and $m_2 = 3$ (B_{2u} representation).

Doubly excited eigenstates inherit their properties from at least two singly excited eigenstates, and therefore their lifetime can also be controlled with the external coupling mechanism. Indeed, the energy $\mathcal{E}^{(m)} = \omega^{(m)} - i\Gamma^{(m)}/2$ of a

doubly excited state $|\Psi^{(m)}\rangle$ can be written as (Sec. S8 [76])

$$\mathcal{E}^{(m)} = \sum_{m_1,m_2} \left|v_{m_1,m_2}\right|^2 [\varepsilon^{(m_1)} + \varepsilon^{(m_2)}], \tag{3}$$

whereas amplitudes v_{m_1,m_2} can be found based on the direct diagonalization of the effective Hamiltonian (Sec. S1 [76]). Hence, the energies of states (2) are $\mathcal{E}^{(3)}_{s_1,s_2} = \varepsilon^{(+1)}_{s_1} + \varepsilon^{(+2)}_{s_2}$ where $\varepsilon^{(m)}_{\pm}$ are given in Sec. S6 [76]. Consequently, the radiative decay of states (2) can also be decomposed into the sum $\Gamma^{(3)}_{s_1,s_2} = \gamma^{(+1)}_{s_1} + \gamma^{(+2)}_{s_2}$. Hence, the suppression of radiative losses for eigenstates (2) can be achieved for a particular oligomer geometry when both singly excited eigenstates with $m_1 = 1$ and $m_2 = 2$ have the lowest radiative losses. By varying the inner (a/λ_0) and outer ring $(b/a$ ratio) sizes independently, we find the optimal parameters to be $b/a = 2.2$ and $a/\lambda_0 \approx 0.16$ (Sec. S9 [76]). Indeed, for these parameters, the fully antisymmetric eigenstate $|\Psi^{(3)}_{--}\rangle$ has a lifetime that is two orders of magnitude larger than that of a single emitter [see Fig. 3(a)]. Moreover, this point is characterized by the maximal lifetime of both antisymmetric singly excited eigenstates with $m = 1$ and $m = 2$ [see Fig. 3(b)]. The radiative losses for the antisymmetric state with $m = 2$ are much smaller than those for $m = 1$, i.e., $\gamma^{(2)}_{-} \ll \gamma^{(1)}_{-}$, therefore, the overall radiative losses for the nonradiant doubly excited eigenstate $|\Psi^{(3)}_{--}\rangle$ are $\Gamma^{(3)}_{--} \approx \gamma^{(1)}_{-}$ [see Fig. 3(b)]. Additionally, we can emphasize that the form of the nonradiant eigenstate $|\Psi^{(3)}_{--}\rangle$, given by Eq. (2), implies that both excitations within this state are predominantly localized on the inner ring [see the inset in Fig. 3(a)], inheriting the properties of $|\psi^{(\pm1)}_{-}\rangle$ and $|\psi^{(\pm2)}_{-}\rangle$ nonradiant eigenstates.

Photon emission and spatial correlations. Finally, the radiative decay of doubly excited states can be characterized by the second-order correlation function, which may be necessary for the future design of potential detection schemes. This function allows for describing spatial correlations between photons emitted by a state $|\Psi\rangle$ when detectors D_1 and D_2 are positioned at coordinates $\mathbf{r}^D_1$ and $\mathbf{r}^D_2$, respectively, and reads as [63,80]

$$g^{(2)}\left(\mathbf{r}^D_1, \mathbf{r}^D_2\right) = \frac{\sum_{\alpha,\beta}\langle\Psi|\hat{E}_{\beta,2}\hat{E}_{\alpha,1}\hat{E}^{\dagger}_{\alpha,1}\hat{E}^{\dagger}_{\beta,2}|\Psi\rangle}{\sum_{\alpha,\beta}\langle\Psi|\hat{E}_{\beta,1}\hat{E}^{\dagger}_{\beta,1}|\Psi\rangle\langle\Psi|\hat{E}_{\alpha,2}E^{\dagger}_{\alpha,2}|\Psi\rangle}. \tag{4}$$

Here, $\alpha, \beta = x, y, z$ denote the components in the Cartesian coordinate system, and $\hat{E}_{\alpha,1(2)} \equiv \hat{E}_{\alpha}(\mathbf{r}^D_{1(2)})$ is the electric field operator that creates a photon at the detector position $\mathbf{r}^D_{1(2)}$ with the polarization along the α axis. The corresponding electric field operator can be expressed via the free-space Green's tensor [43] $\hat{\mathbf{E}}^{\dagger}(\mathbf{r}) = k_0^2/\epsilon_0 \sum_{k=1}^{N} \mathbf{G}_0(\mathbf{r} - \mathbf{r}_k, \omega_0) \cdot \mathbf{d}\hat{\sigma}_k$. Singly excited states can be characterized by the far-field radiation pattern of a single photon $p(\theta, \varphi)$ (Sec. S7 [76]).

In Fig. 4(a), two possible configurations of a two-photon detection scheme are presented: One when a first detector is in the polar position $(\theta^D_1 = 0)$, while a second one scans over the sphere (configuration 1), and another when both detectors are placed in the same point $\mathbf{r}^D_1 = \mathbf{r}^D_2$ (configuration 2). The second-order correlation function (4) of the nonradiant eigenstate $|\Psi^{(3)}_{--}\rangle$ [see Eq. (2) and Fig. 3] is shown in Fig. 4(b). It exhibits typical hexagonal

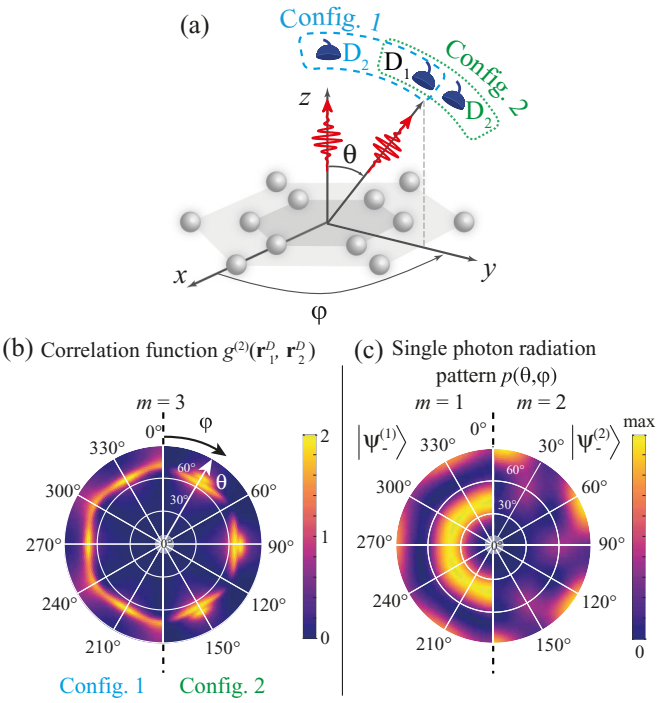

FIG. 4. (a) Configurations for detecting two-photon correlations within doubly excited states. (b) Far-field distribution of $g^{(2)}$ function (4) for the nonradiant eigenstate $|\Psi^{(3)}_{--}\rangle$. (c) Far-field radiation patterns for the singly excited antisymmetric eigenstates $|\psi^{(+1)}_{-}\rangle$ and $|\psi^{(+2)}_{-}\rangle$, which form the nonradiant eigenstate $|\Psi^{(3)}_{--}\rangle$. The results in (b) and (c) are shown considering the symmetry of distributions.

features with a maximal correlation function at a nodal line around $\theta \approx 60°$–$70°$ for both configurations. This behavior can also be explained by radiation patterns for the singly excited eigenstates $|\psi^{(+1)}_{-}\rangle$ and $|\psi^{(+2)}_{-}\rangle$ with $m = +1$ and $m = +2$, respectively, shown in Fig. 4(c). While maximal emission of the $m = 1$ eigenstate is observed around $\theta = 35°$, the emission of the $m = 2$ eigenstate is mainly concentrated around the $\theta = 90°$ plane, which results in a maximal correlation function at an intermediate polar angle as shown in Fig. 4(b).

Discussion and conclusion. As a remark, while the presented result mainly corresponds to linearly polarized σ_z transitions, the external coupling mechanism can be also applied for creating subradiant states for circularly polarized $\sigma_{\pm}$ transitions with the dipole moments lying in the plane of the oligomer structure; see Sec. S10 in Supplemental Material [76]. The recent progress in experimental techniques [41,81] also motivated by a number of fascinating theoretical concepts [43,66,82–84] proposed for quantum arrays with subwavelength spacing give hope that the small spacing between the atoms required for the observation of the discussed nonradiant states will be reached soon.

Finally, we have exploited the Friedrich-Wintgen mechanism to demonstrate the formation of subradiant singly and doubly excited eigenstates with a given orbital momentum in the ring oligomers. We have shown that the oligomers can be viewed as two subsystems of emitters supporting states that interact if they possess the same symmetry. The proposed mechanism relies on the destructive interference between the

subsystems resulting in the formation of antisymmetric states characterized by suppressed radiative losses for preoptimized oligomer geometry. The suggested approach is not limited to the systems considered in this Letter and can be applied to control radiative losses of multiphoton states in various open quantum systems.

Acknowledgments. The authors acknowledge K. Frizyuk, A. Bogdanov, I. Iorsh, and Y. Kivshar for fruitful discussions. The work was supported by the Russian Academic Leadership Program Priority 2030. The calculations of the radiative lifetime were supported by the Russian Science Foundation Grant No. 22-72-10047.

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