Accessing orbital angular momentum of quantum-ring excitons via directional semiconductor luminescence

J V Nieminen, O Vänskä, I Tittonen, S W Koch and M Kira

1 Department of Micro- and Nanosciences, Aalto University, PO Box 13500, FI-00076 Aalto, Finland
2 Department of Physics and Material Sciences Center, Philipps-Universität Marburg, Renthof 5, 35032 Marburg, Germany

E-mail: julius.nieminen@aalto.fi

Keywords: quantum ring, photoluminescence, orbital angular momentum of light

Abstract
A full quantum theory is developed for semiconductor quantum ring photoluminescence. The computations show that the orbital angular momentum (OAM) of individual excitons yields strong angle-dependent emission. The characteristic OAM signatures also survive significant amount of dephasing, suggesting efficient quantum-level coupling between quantum-ring excitons and the OAM of light.

1. Introduction
The realization of schemes that make orbital angular momentum (OAM) of light detectable has opened new possibilities to study light–matter interaction [1–4]. While circularly polarized light can excite a specific spin state in matter [5, 6], helical phase fronts of light beams carry and excite any angular momentum \( l \phi \), where \( l \) is integer valued [4]. These wavefronts are mathematically manifested in the \( \phi \) dependent mode functions with the azimuth angle \( \phi \) [4]. The OAM properties of light have been utilized in, for example, optical tweezers [7], imaging [8], and laser ablation [9]. Furthermore, the OAM aspects are particularly interesting for both classical [10, 11] and quantum information [12, 13] applications, because OAMs can encode high-dimensional states. To utilize the whole potential of OAM, e.g., in quantum information networks [14], it is important to find microscopic systems where the coupling of OAM between light and matter is efficient enough and experimentally detectable.

Systems involving atoms have been extensively studied to achieve the needed quantum-level OAM light–matter coupling. The manipulation of electronic transitions and center-of-mass movement of atoms with OAM of light have been theoretically studied. As a result, the center-of-mass effects are typically found to be much more pronounced than the electronic ones [15–18]. However, the experimental verification of light–matter coupling in a single-atom level has been found to be challenging [4, 19], due to the small size of individual atoms, whereas macroscopic states, such as Bose–Einstein condensates of atoms, are large enough to interact efficiently with OAM light modes. Consequently, OAM coupling between light and matter has been demonstrated with different types of collective excitations of various atomic ensembles [20–23] even at the single-photon level [24, 25].

In semiconductors, the Wannier excitons [26] are quasiparticles consisting of electron–hole pairs bound together by the Coulomb interaction. Due to their relatively large size, exciton states should interact with the OAM of light more efficiently than an atom. This type of coupling has already been demonstrated with semiconductor excitations in mesoscopic thin film structures [27, 28]. The OAM effects can be particularly interesting in quantum-confined structures with circular symmetry, especially when the movement of electrons is confined in all three dimensions. In such quantum-dot-like systems, the electronic states have the same \( e^{i\phi} \) dependence as the OAM light modes enhancing the electron–light OAM coupling. However, the electronic transitions in conventional quantum dots suffer partly from similar small-size restrictions as transitions in atoms, a limitation that may be circumvented by a quantum ring (QR) [29]. The doughnut-like topology of a QR makes it possible to introduce a fully quantized charge-carrier system with a large diameter and a strong OAM.
dependence [29, 30]. The resulting interactions can even occur with OAM modes of a plane-wave field that has a zero OAM, on average [31]. As a consequence, one should be able to efficiently excite excitons of varying OAM in QR systems.

In this work, we study quasi-stationary photoluminescence (PL) emitted from a QR structure by varying excitation conditions. We show that even a single QR produces a significant OAM coupling and that the angle-resolved PL is highly dependent on the OAM distribution of excitons. We also demonstrate that the angle-dependent PL can efficiently be applied to investigate OAM coupling between excitons and light. Furthermore, we find that the different OAM channels can be resolved even when the scattering and dephasing effects become appreciable. The applicability of the effect is studied in two QR systems with different physical radii.

2. Idea of angle-dependent PL from rotating excitons

Following a fully microscopic theoretical approach [26, 32], we derive the steady state of semiconductor luminescence equations [32, 33]. The full derivation and complete PL spectrum $I_{PL}(\omega, \theta)$, used in all our computations, is presented in the appendix. In this work, we consider only emission of s-polarized light to eliminate unnecessary complications resulting from scalar products between the dipole-matrix element and polarization vectors of light modes [34]. The physical consequences depicted by the full quantum theory can be illustrated via a simplified Elliott luminescence formula [26, 33] for the QR emission spectrum:

$$I_{PL}^{\omega \theta} = \sum_{M} \frac{J_{M}^{s}(nRq \sin \theta) \left( \Delta N_{M,1s} + N_{M,1s}^{s} \right)}{(E_{M,1s} - \omega \gamma)^2 + \gamma^2},$$

where $\hbar \omega$ is photon energy, $\theta$ is the emission direction indicated in figure 1, $q = \omega/c$ is the wavenumber, $c$ is the speed of light in vacuum, $R$ is the radius of the QR, $n$ is the refractive index, and $J_{M}$ is the Bessel function of the first kind. The exciton states have energy $E_{M,s}$ and the two quantum numbers $M$ and $s$ are related to the center-of-mass OAM $\hbar M$ and $\nu$ symmetry of the relative motion, respectively. The emission strength is proportional to the exciton population $\Delta N_{M,s}$ and to the plasma contribution $N_{M,1s}^{s}$. Equation (1) gives the PL intensity accurately when the emission is dominated by the $\nu = 1s$ contribution, the thickness of the ring is considerably smaller than the wavelength of photons and the oscillator strengths for different $1s$ states are equal; full computations include effects beyond these simplifications. We describe dephasing with a phenomenological constant $\gamma$. It defines the linewidth of PL peaks and its value is affected by various three-body scattering processes stemming from the Coulomb as well as from phonon interactions [26].

When 1s population dominates the many-body state, which is studied in here, excitonic PL originates almost exclusively from the exciton densities $\Delta N_{M,1s}$ [26] that justifies setting $N_{M,1s}^{s} = 0$ in (1); again, this assumption is applied only for illustrative purposes whereas the plasma contributions are fully included in our computations. By using $N_{M,1s}^{s} = 0$, the PL intensity becomes directly dependent on $J_{M}^{s}(nRq \sin \theta)$, which allows us to distinguish the different OAM states from the $\theta$ dependence of the PL. More specifically, excitons rotating with different absolute values of $|M|$ should have distinctly different angular dependence. For example, the behavior of $J_{M}(x)$ near $x = 0$ is proportional to $x^{M}$ such that only the $M = 0$ exciton can emit light to $\theta = 0$ angle (if the plasma part is omitted). However, for larger angles also excitons with $M \neq 0$ will contribute to the $\theta > 0$ emission. This idea is schematically presented in figure 1 for two particular excitons (blue spheres) with $M = 0$ and $M = 1$; OAM of $M = 1$ state is indicated by the blue arrow. The resulting PL contributions to different angles $\theta$ emitted from these excitons are indicated by the red arrows and curves that are in the $y$-$z$ plane.

---

Figure 1. Schematic presentation of the QR system and the angle-of-emission ($\theta$) dependence of PL emitted by excitons (blue spheres) with OAM quantum numbers $M = 0$ and $M = 1$. The luminescence contributions to different angles in the $y$-$z$ plane are shown by red arrows and curves. The rotation of $M = 1$ is indicated by the blue arrow.
3. Results

To resolve the OAM dependence of PL in more detail, we calculate PL spectra for angles $\theta = 0$ and a larger angle for two different GaAs-based QR model systems. We choose $\theta = \pi/2$ to represent the larger angle as the limiting value of an experiment. In both systems, the lateral thickness of QR, difference between the inner and outer radii, is 35 nm and the height is 3 nm. In figure 2(a), the results are shown for the smaller QR that has a radius of 50 nm. The corresponding results for the larger QR with a 150 nm radius are presented in figure 2(b). Other details of geometry and material parameters for these model structures are given in the appendix.

In both systems, we consider five different initial excitation conditions. The red solid line in figure 2 is the computed PL from type $\nu = M_0$ excitons. We also analyze emission for equal $|N| = M_1$ (black solid), $|N| = M_2$ (red broken), and $|N| = M_3$ (light blue dotted) occupations. In all of these four cases, we have chosen $\Delta N_{M,1_1} = 10^{-3}$ while all $\nu \neq 1s$ populations are set to zero. We also investigate the emission from a thermal exciton distribution and the related results are indicated by the shaded area. This distribution follows from $\Delta N_{M,\nu} = N_0 \exp (-E_{M,\nu}/k_BT)$ with $N_0$ chosen so that the occupation of type $(M = 0, \nu = 1s)$ exciton is equal for the thermal distribution and for the pure $M = 0$ excitation. The temperature is $T = 30 K$ and a relatively small dephasing value $\gamma = 0.06 \text{ meV}$ is used. PL is normalized with respect to the maximum emission obtained when $M = 0$ and $\theta = 0$.

Figures 2(a-i) and (b-i) show PL for $\theta = 0$ and confirm that only $M = 0$ excitons produce considerable PL to the direction parallel to the symmetry axis of QR. The angle $\theta = \pi/2$ produces a completely different result, as seen in figures 2(a-ii) and (b-ii). Now, PL emitted from the $M = 0$ excitation is significantly reduced while type $\neq M_0$ excitons dominate the PL spectra (excluding the type $|N| = 3$ exciton in the small QR that only yields a minor resonance near $\hbar \omega - E_{0,1_1} = 0.8 \text{ meV}$). This verifies the assumption of the relation between the angle-of-emission-dependent PL and the OAM distribution of excitons in systems with sufficiently low carrier densities.

To study the angular dependence of PL more thoroughly, the accurate $I_{\text{pl}}(\omega_0, \theta)$, where $\hbar \omega_0 = E_{0,1_1}$, is plotted in figures 3(a-i) and (b-i) as a function of angle $\theta$ for the smaller and the larger QR, respectively. Here, we use the same linestyles as in figure 2; the thermal $I_{\text{pl}}$ is indicated by the gray dashed lines. The excitation conditions are assumed to be the same as previously, with the exception that we have chosen $\gamma = 1 \text{ meV}$ and only $M \geq 0$ excitons are assumed instead of $M = \pm |M|$ excitations to simplify the analysis; in the thermal excitation all $\pm |M|$ states are present.
It is interesting to notice that the full PL in figures 3(a-i) and (b-i) for $M$ excitations has the same form as $I_{nRq}^M(nRq \sin \theta)$ with relative deviations typically less than a few percent. Thus, the $\theta$ dependence of PL can be explained via the properties of the Bessel functions and the more precise $I_{\omega \theta \gamma}^s$ in (1) is then an extremely accurate approximation of the full PL. The main reason for the deviations between $I_{\omega \theta \gamma}^s$ and the full calculation stems from the used infinitesimally thin QR assumption when (1) is formulated. In addition, for regions where PL intensity is small, the deviations between Bessel functions and $I_{\omega \theta \gamma}^0$ can be much larger, resulting from the omission of the plasma contribution.

By considering the properties of $I_{nRq}^M(nRq \sin \theta)$ or directly PL in figures 3(a-i) and (b-i), we see that at larger $\theta$ angles we observe more oscillations using the larger QR. This is directly related to the $nRq$ argument of the Bessel functions; a larger QR radius simply induces a stronger $\theta$ and OAM dependence in PL. Since $M$ is fixed and $q$ is nearly constant (defined roughly by the band gap), small enough QRs cannot emit light through excitons with large OAM numbers. Figure 3(a-i) shows that for the smaller QR, only type $M=0, 1, 2$ excitons produce appreciable PL. At the same time, figure 3(b-i) shows that the larger QR emits light to all $M$ states analysed. In fact even $M=5$ still produces a significant PL.

As PL measurements can detect intensity variations over several orders of magnitude [35], it is useful to calculate also relative differences of expected PL experiments by introducing

$$\Delta PL = \frac{I_{PL}^0(\theta, 0, \gamma) - I_{PL}^1(\theta, 0, \gamma)}{I_{PL}^0(\theta, 0, \gamma) + I_{PL}^1(\theta, 0, \gamma)}.$$  (2)

The computed $\Delta PL$ is shown in figures 3(a-ii) and (b-ii) using the same linestyles, excitations and $\gamma$ as in figures 3(a-i) and (b-i). We observe that $\Delta PL$ changes appreciably when $\theta$ exceeds roughly $\pi/6$ for the smaller and $\pi/20$ for the larger QR. In other words, OAM effects are detectable by comparing $\theta$ dependence of PL in a rather small angular range. As a general trend, the $\theta$ dependence becomes slower as $|M| > 0$ is increased. For the larger QR, $M=1$ excitation produces an additional peak (black line, figure 3(b-ii)) close to the value $\theta = \pi/3$; this stems from the zero-crossing of $I_{nRq}^1$ at this specific value, see figure 3(b-i). Interestingly, $\Delta PL$ depends strongly on $\theta$ only for pure excitonic emitters. For the thermal state, the shown $\Delta PL$ remains essentially constant. To resolve OAM for this case, one obviously needs to analyze the full spectrum and its $\theta$ dependence, as shown in figure 2.

In general, (1) shows that both plasma and exciton populations contribute to PL. However, the plasma source $N_{M,\rho}^p$ is typically orders of magnitude smaller than the $\Delta N_{M,\rho}$ population because it depends quadratically on density at the low density regime whereas $N_{M,\rho}^s$ dependency is linear. Therefore, the influence of
plasma on PL spectra is weak whenever the system contains exciton populations [35]. As a general trend, the plasma contributions produce a very weak $\theta$ dependence. Consequently, the OAM dependence of PL stems essentially from the excitons.

Experiments can resolve individual exciton resonances only if the broadening of resonances remains small enough. Therefore, it is important to check how well the OAM effects can be resolved as a function of the dephasing $\gamma$. For this purpose, figure 4 presents $\Delta \text{PL}(\pi/2, \gamma)$ as a function of $\gamma$ for the smaller QR with the same excitation conditions and linestyles as in figure 3. For $M = 0$ excitation, $\Delta \text{PL}$ is independent of $\gamma$, whereas $\Delta \text{PL}$ saturates to the values for other initial conditions indicated in figure 3(a-ii) as soon as $\gamma$ approaches the energy difference between the considered $M$ exciton and $\hbar\omega_0$. Consequently, even when the spectral separability of exciton resonances is lost the $\theta$ dependence of $\Delta \text{PL}$ remains and is not influenced by increasing values of $\gamma$.

The strongest $\gamma$ dependence is found with the thermal distribution, where the separability of OAM excitons is based on spectral resolution. Figure 4 shows that this separability for the smaller QR at 30 K is basically lost for dephasing greater than 0.5 meV. After this, a $\Delta \text{PL}$ measurement at a single spectral point detects a mixture of OAM effects that tend to average to zero. This result implies that OAM dependence is easier to observe when the system contains individual excitons.

In a realistic situation, a single exciton state is accompanied by a thermal background of excitons so that it is interesting to study how large singular exciton populations must be present in order to detect clear OAM signatures. To determine this precisely, we compute $\Delta \text{PL}(\theta, \gamma)$ when a singular density, $\Delta N_{M,1s}$, of $(M, 1s)$ excitons is added to the otherwise thermal distribution $\Delta N_{\text{th}}$. We assume that the total density $\Delta N_{\text{tot}} = \Delta N_{M,1s}^{\text{add}} + \sum_j \Delta N_{j,\theta}^{\text{th}}$ is constant and define the added singular fraction via

$$F_{\text{add}} \equiv \frac{\Delta N_{M,1s}^{\text{add}}}{\Delta N_{\text{tot}}}.$$  

Figure 5 presents $\Delta \text{PL}$ as a function of $F_{\text{add}}$ for the addition of $M = 0, ..., 3$ type excitons with the same linestyles as in figures 3 and 4; results for the smaller and the larger QR are shown in frames (a) and (b), respectively. We use 30 K exciton temperature for $\Delta N_{j,\theta}^{\text{th}}, \gamma = 1$ meV, and determine $\Delta \text{PL}(\theta, \gamma)$ at that $\theta$ angle which maximizes $\Delta \text{PL}$ for each $F_{\text{add}}$ separately.

Figure 5 shows that already small fractions of additional $M = 0$ and $M = 1$ excitons can be detected from the thermal background with both QRs. In the small QR, a much larger fraction of type $M = 2$ excitons is needed for clear separation while type $M = 3$ excitons are not detectable until almost 100% of excitation includes them. In the larger QR, these states can also be easily separated from the thermal distribution, which indicates that excitonic OAM effects are easier to detect in larger QRs using angle-resolved PL.

4. Summary and conclusions

As a summary, we have studied angle-of-emission resolved PL from a semiconductor QR structure. We have shown that the angle dependence of light emission from excitons is directly related to the absolute value of the center-of-mass OAM quantum number $M$ of excitons. Thus, this effect can be used to measure excited OAM distributions of excitons in QRs. The effect is strongest in the low-density regime of charge carriers when only
excitons with one $|M|$ value are excited. Both the plasma contribution to PL and the existence of other $M$ channels of excitons will reduce the usability of angle-resolved PL to detect explicit densities of $|M|$ excitons. We studied the separability of $M$ excitons from the thermal distribution and found that in small QRs the detection of additional excitons with $|M| > 1$ needs a considerable fraction of these particular states compared to the total density. In contrast, we found that PL from larger QRs can be used to observe excitons with higher $M$ numbers from the angle-of-emission independent background. In addition, the dephasing independence of our results indicates that our scheme of characterization of OAM states is highly robust and can be used to interpret measurements under different experimental conditions.

In the present work, we investigate QRs with isotropic electron and hole masses close the $\Gamma$ point. Many current QR experiments [36–38] are realized with GaAs-type systems where the mass is indeed highly isotropic [39, 40], validating our predictions for such systems. However, there are several materials such as Ge and ZnO whose effective masses can be highly anisotropic [41, 42] even in nanostructures [43–45]. In such systems, the mass anisotropy breaks the rotation symmetry such that the OAM is not a good quantum number for the electronic states. For example, circular OAM states become elliptic, which also modifies the selection rules for the angle-of-emission dependent transitions. For small modifications, one can use perturbation theory to access the small changes in the predicted angle-of-emission dependence. Since the level of mass anisotropy can be systematically controlled by the growth direction, strain, quantum-confinement geometry, etc., it clearly is interesting to study the interplay of this symmetry breaking and the directional OAM effects in further investigations.

Besides extensions to study mass anisotropy, one could also explore the intricate many-body scattering effects [32] behind the broadening of PL resonances and the lifetimes of excitations. It has already been verified that the luminescence times in QR systems are considerably increased when measurements are performed at high temperatures, which can be possibly related to the spreading of excitations over dark states [38, 46]. How well this spreading conserves OAM of exciton states, as well as many other aspects of light–matter coupling in QRs, could be resolved by measuring time- and angle-dependent PL simultaneously. These kinds of measurements together with our results can lead to new technologies in which microscopic systems capable of storing and further manipulating OAM information are needed.

Acknowledgments

The work at Aalto University was supported by the Euramet SIB04 project and the Academy of Finland (project 129043). The EMRP is jointly funded by EMRP participating countries within EURAMET and the European Union. The Marburg authors thank the Deutsche Forschungsgemeinschaft for partial support through SFB 1083 and grant KI 917/2-1. We also acknowledge the provision of the Micronova Nanofabrication Centre of Aalto University.

Appendix

The modeling of our system starts from the two-band envelope-function approximation [26] where the wave functions of electronic states are given by $\psi_{\lambda, m}(r) = w_\lambda(r)\xi_{\lambda,m}(r)$. Here, $w_\lambda(r)$ is the crystal periodic Bloch
function at the $\Gamma$ point, $\xi_{\lambda,m}(r)$ is the confinement function, and $\lambda$ is the band index with values $\lambda = c$ or $\lambda = v$ for the conduction and valence band, respectively. Furthermore, we assume that the lateral thickness and height of the studied QRs are so small that the relevant electronic states for our calculations remain in their ground state with respect to the radial- and $z$-directional excitations. Thus, the confinement index $m$ defines the OAM quantum number of single-particle states with respect to the axis of the circularly symmetric ring. The assumed symmetry of QRs is used to separate the azimuth–angle dependency of the confinement functions, giving $\xi_{\lambda,m}(r) = f_{\lambda,m}(\rho, z) e^{im\phi}$, where $\rho$ is the radial coordinate. The last unknown part of the electronic wave function, $f_{\lambda,m}(\rho, z)$, and the carrier energies, $E_{m}$, are solved numerically from the $\rho$–$z$–$\phi$ directional effective mass Schrödinger equation

$$\left\{ -\frac{\hbar^2}{2m_{\parallel}^2} \left[ \frac{1}{\rho} \frac{\partial}{\partial \rho} \left( \rho \frac{\partial}{\partial \rho} \right) - \frac{m^2}{\rho^2} \right] - \frac{\hbar^2}{2m_{\perp}^2} \frac{\partial^2}{\partial \zeta^2} + U_\lambda(r) \right\} \xi_{\lambda,m}(r) = E_{m} \xi_{\lambda,m}(r),$$  \hspace{1cm} (A.1)$$

where $m_{\parallel}^2$ and $m_{\perp}^2$ are the in-plane and $z$-directional effective masses and $U_\lambda$ is the confinement potential.

The explicit form of $U_\lambda(\rho, z)$ is taken to be zero inside the QRs with parabolic cross section and given by $\lambda$-band offset $U_\lambda$ between the QR and the buffer material. The model rings fabricated from GaAs are embedded inside AlGaAs. The values for the band offsets and effective masses are evaluated by following similar steps as in [47] and by assuming confinement-induced heavy–light–light-hole energy splitting at the $\Gamma$ point. The final values $U_c(\rho, z) = 330$ meV, $U_v(\rho, z) = -210$ meV, $m_{c,\perp} = m_{c,\parallel} = 0.0670 m_0$, $m_{v,\perp} = -0.1100 m_0$, and $m_{v,\parallel} = -0.3500 m_0$ are obtained by using material parameters in [40].

The many-body properties of our system are derived from the system Hamiltonian $H = H_0 + H_C + H_D$ [26]. In this, the single-carrier and photon contributions are

$$H_0 = \sum_{\lambda, m} E^c_\lambda a^\dagger_{\lambda,m} a_{\lambda,m} + \sum_{\bf q} \hbar \omega_\bf q \left( B^\dagger_\bf q B_\bf q + \frac{1}{2} \right),$$  \hspace{1cm} (A.2)$$

where $a^\dagger_{\lambda,m}$ and $a_{\lambda,m}$ are creation and annihilation operators, respectively, for a carrier in band $\lambda$ with OAM index $m$. The corresponding operators for photons with a mode index $\bf q$ are $B^\dagger_\bf q$ and $B_\bf q$. The carrier–carrier interactions are given by

$$H_C = \frac{1}{2} \sum_{\lambda, \lambda'} \sum_{M, m, \lambda} V_{m\lambda M}^{\lambda\lambda'} a^\dagger_{\lambda,m+1} a^\dagger_{\lambda',m} a_{\lambda',m+1} a_{\lambda,m},$$  \hspace{1cm} (A.3)$$

which includes the Coulomb-matrix element [26]

$$V_{m\lambda M}^{\lambda\lambda'} = \frac{e^2}{4\pi \varepsilon_0 \varepsilon} \times \int d^2r d^2r' \frac{\xi^\dagger_{\lambda,m+1}(r) \xi_{\lambda',m+1}(r') \xi_{\lambda',M+1}(r') \xi_{\lambda,m}(r)}{|r - r'|},$$  \hspace{1cm} (A.4)$$

where $\varepsilon$ is the dielectric constant of the structure and we have already applied the OAM selection rule for Coulomb interaction. The dipole Hamiltonian [26]

$$H_D = - \sum_{\bf q, M, m} \left[ F_{m\bf q}^M B^\dagger_\bf q a^\dagger_{\bf q,m+M} a_{\bf q,m} - \left( F_{m\bf q}^M \right) B_\bf q a^\dagger_{\bf q,m+M} a_{\bf q,m+M} \right],$$  \hspace{1cm} (A.5)$$

introduces the light–matter interaction to the system, where

$$F_{m\bf q}^M = i \varepsilon_\bf q d_{\bf q,\lambda} \int d^2r e^{i\bf qr} \xi_{\bf q,m+M}(r) \exp \left( i \bf q \cdot r \right) \xi_{\bf q,m}(r).$$  \hspace{1cm} (A.6)$$

Here, $d_{\bf q,\lambda}$ is the dipole-matrix element [26] between bands, $\varepsilon_\bf q = \sqrt{\hbar \omega_\bf q / (2\varepsilon_0)}$ is the vacuum field amplitude, and $n$ is the refractive index. In (A.3)–(A.6), we have already made the separation between center–of–mass OAM ($M$) and relative movement of carriers ($m$). Furthermore, we only consider s-polarized plane-wave modes, resulting in that the mode index $\bf q$ corresponds directly to the wave vector of the mode.

To formulate the QR Elliott luminescence formula for the plane-wave modes we follow the steps taken in [33] by applying the cluster–expansion method [26, 32]. In more detail, we define

$$f^c_m \equiv \left\{ a^\dagger_{c,m} a_{c,m} \right\}, \hspace{1cm} f^h_m \equiv 1 - \left\{ a^\dagger_{v,m} a_{v,m} \right\},$$  \hspace{1cm} (A.7)$$

$$\xi^M_{N,m',m} \equiv \Delta \left\{ a^\dagger_{v,m} a_{c,m'} a_{c,m+M} a_{v,m-M} \right\},$$  \hspace{1cm} (A.8)$$
\[ \Pi_{m}^{\text{QM}} \equiv \Delta \left( B_{q}^{*} a_{r,m,a_{e,m+M}} \right). \]  

(A.9)

where \( f_{m}^{e} (f_{m}^{h}) \) is the electron (hole) occupation, \( c_{X M',m'}^{M'} \) is the exciton correlation, and \( \Pi_{m}^{\text{QM}} \) is the photon-assisted polarization. In the next step, we solve the dynamics of the photon-assisted polarization and the two-photon correlation \( \Delta \langle B_{q}^{*} B_{q} \rangle \) that give the semiconductor luminescence equations [32]

\[
i\hbar \frac{\partial}{\partial t} \Pi_{m}^{\text{QM}} = \sum_{M,m} \left[ F_{m}^{\text{QM}} \left( \Pi_{m}^{\text{QM}} \right)^{*} - \left( F_{m}^{\text{QM}} \right)^{*} \Pi_{m}^{\text{QM}} \right],
\]

(A.10)

\[
i\hbar \frac{\partial}{\partial t} \Pi_{m}^{\text{QM}} = -\left( \hbar \omega_{q} + \gamma \right) \Pi_{m}^{\text{QM}} + \sum_{m'} A_{mmm'}^{M} \Pi_{m'}^{\text{QM}} - F_{m}^{\text{QM}} f_{m+M}^{h} - F_{m}^{\text{QM}} f_{m+M}^{e},
\]

(A.11)

for a QR system [33]. Here, for simplicity, we have only given the equation for \( \Delta \langle B_{q}^{*} B_{q} \rangle \) in (A.10) and already omitted the contribution of stimulated emission [26] in (A.11). Furthermore, we have assumed that long enough time has passed after the optical excitation, so that all optical coherences have already vanished [26] and that the system is in a homogeneous state [26] where all excitations are distributed evenly across the QR circumference. In the preceding equations, \( \gamma \) includes phenomenologically the triplet-correlation-induced dephasing [32] and

\[
A_{mmm'}^{M} = \delta_{m,m'} \bar{E}_{m}^{M} - \left( 1 - f_{m}^{e} - f_{m+M}^{h} \right) V_{mmm'-m}^{v,\nu,\nu',\nu''}.
\]

(A.12)

where the renormalized electron–hole-pair energy is given by

\[
\bar{E}_{m}^{M} = E_{g} + E_{m+M}^{e} - E_{m}^{h} - \sum_{m'} \left( V_{m,m+M-m}^{v,\nu,\nu'-m} + V_{m,m+M-M}^{v,\nu,\nu'-m} \right) f_{m'}^{e} - \sum_{m'} \left( V_{m,m'-m}^{v,\nu,\nu'-m} + V_{m+M,m'+M}^{v,\nu,\nu'-m} \right) f_{m'}^{h}.
\]

(A.13)

with the band gap \( E_{g} \) of the QR material.

The matrix \( A_{mmm'}^{M} \) in (A.12) defines the Wannier equation for excitons [26]:

\[
E_{m}^{M} \phi_{M}^{R} (m) - \left( 1 - f_{m}^{e} - f_{m+M}^{h} \right) \sum_{m'} V_{mm'm'-m}^{v,\nu,\nu',\nu''} \phi_{M}^{R} (m') = E_{M,v}^{e} \phi_{M}^{R} (m),
\]

(A.14)

where \( \phi_{M}^{R} (m) \) and \( E_{M,v}^{e} \) are the exciton wave function and energy for the exciton with center-of-mass OAM \( \hbar M \) and with a symmetry of relative movement \( \nu \). The index \( R \) in the exciton wave function denotes the right-handed solution of the Wannier equation. Whenever carrier populations are present, (A.14) has also left-handed solutions \( \phi_{M}^{L} (m) \) [26]. Equation (A.14) corresponds to the Wannier equation in quantum wires [26], which can be also seen from our numerically solved ground states (\( \nu = 1s \)) that are highly similar to the 1s solutions of quantum-wire excitons.

The intensity of PL, \( I_{\text{PL}} \), is defined by the photon flux [34]:

\[
I_{\text{PL}} (q) = \frac{\hbar}{2} \Delta \langle B_{q}^{*} B_{q} \rangle.
\]

To solve \( I_{\text{PL}} \), we consider a quasi-stationary state where the spontaneous–emission source [26] in the second line of (A.11) can be assumed to be constant in the studied time window. After evaluating the contribution of correlated electron–hole plasma [26] to the emission, we obtain the Elliot luminescence formula

\[
I_{\text{PL}} (\omega, \theta) = 2 \gamma \sum_{M,v} \left| F_{M,v}^{M} (\omega, \theta) \right|^{2} \left( \Delta N_{M,v} + N_{M,v}^{\text{ps}} \right) \left( E_{M,v} - \hbar \omega \right)^{2} + \gamma^{2},
\]

(A.15)

for the plane-wave mode with frequency \( \omega \) that propagates at an angle \( \theta \) with respect to the symmetry axis of QR. Here, the exciton distribution [32] is defined by \( \Delta N_{M,v} \equiv \sum_{m} \phi_{M}^{L} (m) \phi_{M}^{L} (m') c_{X M',m'+M}^{M'} \) and the plasma contribution by \( N_{M,v}^{\text{ps}} \equiv \sum_{m} \phi_{M}^{L} (m) f_{m+M}^{e} f_{m+M}^{h} \). The absolute square of effective oscillatory strength \( F_{M,v}^{M} (\omega, \theta) \) is given by
\[
\left| P_{M,s}^{\omega \theta} (\omega, \theta) \right|^2 = E_q^2 |d_{s1}|^2 \sum_m \phi_{M,s}^R (m) \int dp dz \rho_{M,s} (npq \sin \theta) \exp(inzq \cos \theta) \\
\times f_{c,m+M} (\rho, z) f_{c,m+M}^{*} (\rho, z),
\]
(A.16)

where we have used the Jacobi–Anger expansion before integrating over the azimuth angle.

The approximative form of the full \( I_{\text{PL}}^{1s} (\omega, \theta) \) given in (1) is obtained when we use the long wavelength limit approximation in (A.16) with respect to the lateral thickness and height of QR. This results in

\[
\left| P_{M,s}^{\omega \theta} (\omega, \theta) \right|^2 = E_q^2 |d_{s1}|^2 \sum_m \phi_{M,s}^R (m) \left( R^2 \right),
\]

where \( R \) is the radius of QR. Furthermore, our numerical calculations show that the oscillatory strengths \( \sum_m \phi_{M,s}^R (m) \) for 1s excitons are independent of \( M \) with high accuracy. And finally, when we consider only emission from the 1s states in arbitrary units, the constant-like group of terms \( 2\pi E_q^2 |d_{s1}|^2 \sum_m \phi_{M,s}^R (m) \left( R^2 \right) / h \) can be omitted to obtain the final form of approximative \( I_{\text{PL}}^{1s} (\omega, \theta) \).

References
[1] Allen L, Beijersbergen M W, Spreeuw R J C and Woerdman J P 1992 Phys. Rev. A 45 8185–9
[2] Molina-Terriza G, Torres J P and Torner L 2007 Nat. Phys. 3 305–10
[3] Franke-Arnold S, Allen L and Padgett M 2008 Laser Photonics Rev. 2 299–313
[4] Yao A M and Padgett M J 2011 Adv. Opt. Photon. 3 161–204
[5] Gerardot B D, Brunner D, Dalgarno P A, Öhberg P, Seidl S, Kroner M, Karrai K, Stoltz N G, Petroff P M and Warburton R J 2008 Nature 451 411–4
[6] Hanson R and Awschalom D D 2008 Nature 453 1043–9
[7] Grier D G 2003 Nature 424 810–6
[8] Förhapter S, Jesacher A, Bernet S and Ritsch-Marte M 2005 Opt. Express 13 689–94
[9] Omatsu T, Chujo K, Miyamoto K, Okida M, Nakamura K, Aoki N and Morita R 2010 Opt. Express 18 17967–73
[10] Wang J et al 2012 Nat. Photonics 6 488–96
[11] Bozinovic N, Yue Y, Ren Y, Tur M, Kristensen P, Huang H, Willner A E and Ramachandran S 2013 Proc. Natl Acad. Sci. USA 110 12614–7
[12] Großblacher S, Jenniswein T, Vaziri A, Weihs G and Zeilinger A 2006 New J. Phys. 8 R75
[13] Fickler R, Lapkiewicz R, Plick W N, Krenn M, Schaeff C, Ramelow S and Zeilinger A 2012 Science 338 638–43
[14] Kimble H J 2008 Nature 453 1023–30
[15] Babiker M, Bennett R C, Andrews D L and Dávila Romero L C 2002 Phys. Rev. Lett. 89 143601
[16] Pícón A, Benseny A, Vázquez de Aldana J R,Mompart J,Plaja L,Calvo G F and Roso L 2010 New J. Phys. 12 020305
[17] Mondal P K, Deb B and Majumder S 2014 Phys. Rev. A 89 063418
[18] Scholz-Margraf H M, Fritzsche S, Serbo V G, Afanasiev A and Suhayzhynyy A 2014 Phys. Rev. A 90 013425
[19] Thamvanthi S, Kapale T K and Dowling P J 2008 Phys. Rev. A 77 033825
[20] Tabosoa J W R and Petrov D V 1999 Phys. Rev. Lett. 83 4967–70
[21] Inoue R, Kanai N, Yonehara T, Miyaomoto Y, Koashi M and Kozuma M 2006 Phys. Rev. A 74 053809
[22] Andersen M F, Ryu C, Clade P, Natarajan V, Vaziri A, Helmerson K and Phillips W D 2006 Phys. Rev. Lett. 97 170406
[23] Chen Q F, Shi B S, Zhang Y S and Guo G C 2008 Phys. Rev. A 78 053810
[24] Ding D S, Zhou S Y and Guo G G 2013 Nat. Commun. 4 3527
[25] Nicolas A, Veissier L, Giner L, Giacobino E, Mayne D and Lauriat J 2014 Nat. Photonics 8 234–8
[26] Kira M and Koch S W 2012 Semiconductor Quantum Optics (Cambridge: Cambridge University Press)
[27] Chen G, Shi B S, Zhang Y S and Guo G C 2008 Phys. Rev. B 78 053810
[28] Ding D S, Zhou S Y and Guo G G 2013 Nat. Commun. 4 3527
[29] Nicolas A, Veissier L, Giner L, Giacobino E, Mayne D and Lauriat J 2014 Nat. Photonics 8 234–8
[30] Kira M and Koch S W 2012 Semiconductor Quantum Optics (Cambridge: Cambridge University Press)
[31] Chen Q F, Shi B S, Zhang Y S and Guo G C 2008 Phys. Rev. A 78 053810
[32] Wang J et al 2012 Nat. Photonics 6 488–96
[33] Bozinovic N, Yue Y, Ren Y, Tur M, Kristensen P, Huang H, Willner A E and Ramachandran S 2013 Proc. Natl Acad. Sci. USA 110 12614–7
[34] Großblacher S, Jenniswein T, Vaziri A, Weihs G and Zeilinger A 2006 New J. Phys. 8 R75
[35] Fickler R, Lapkiewicz R, Plick W N, Krenn M, Schaeff C, Ramelow S and Zeilinger A 2012 Science 338 638–43
[36] Kimble H J 2008 Nature 453 1023–30
[37] Babiker M, Bennett R C, Andrews D L and Dávila Romero L C 2002 Phys. Rev. Lett. 89 143601
[38] Pícón A, Benseny A, Vázquez de Aldana J R, Mompart J, Plaja L, Calvo G F and Roso L 2010 New J. Phys. 12 020305
[39] Mondal P K, Deb B and Majumder S 2014 Phys. Rev. A 89 063418
[40] Scholz-Margraf H M, Fritzsche S, Serbo V G, Afanasiev A and Suhayzhynyy A 2014 Phys. Rev. A 90 013425
[41] Thamvanthi S, Kapale T K and Dowling P J 2008 Phys. Rev. A 77 033825
[42] Tabosoa J W R and Petrov D V 1999 Phys. Rev. Lett. 83 4967–70
[43] Inoue R, Kanai N, Yonehara T, Miyaomoto Y, Koashi M and Kozuma M 2006 Phys. Rev. A 74 053809
[44] Andersen M F, Ryu C, Clade P, Natarajan V, Vaziri A, Helmerson K and Phillips W D 2006 Phys. Rev. Lett. 97 170406
[45] Chen Q F, Shi B S, Zhang Y S and Guo G C 2008 Phys. Rev. A 78 053810
[46] Ding D S, Zhou S Y and Guo G G 2013 Nat. Commun. 4 3527
[47] Nicolas A, Veissier L, Giner L, Giacobino E, Mayne D and Lauriat J 2014 Nat. Photonics 8 234–8
[48] Kira M and Koch S W 2012 Semiconductor Quantum Optics (Cambridge: Cambridge University Press)
[49] Chen G, Shi B S, Zhang Y S and Guo G C 2008 Phys. Rev. A 78 053810