Using this in situ EBL process, we can place quantum dot–waveguide (QD–WG) systems to emit single photons with high directionality. Low-temperature in situ electron-beam lithography enabled by cathodoluminescence mapping is used to select suitable QDs and to integrate them deterministically into linear WG structures at specific chiral points determined by numerical calculations. Excitonic and biexcitonic emission is observed from the fabricated QD–WG structure in a confocal micro-photoluminescence setup enabling the optical characterization in terms of directional emission of circularly polarized photons emitted by integrated QDs. The results show a high degree of anisotropy on the level of 54% for directional QD emission and antibunching in autocorrelation experiment confirming the fabricated QD–WG system, which is a prerequisite for using this effect in advanced applications in integrated quantum circuits.

Chiral light–matter interaction can lead to directional emission of two-level quantum emitters in waveguides (WGs). This interesting physics effect has raised considerable attention in recent years especially in terms of on-chip quantum systems. In this context, this work focuses on tailoring single semiconductor quantum dot–waveguide systems to emit single photons with high directionality. Low-temperature in situ electron-beam lithography enabled by cathodoluminescence mapping is used to select suitable QDs and to integrate them deterministically into linear WG structures at specific chiral points determined by numerical calculations. Excitonic and biexcitonic emission is observed from the fabricated QD–WG structure in a confocal micro-photoluminescence setup enabling the optical characterization in terms of directional emission of circularly polarized photons emitted by integrated QDs. The results show a high degree of anisotropy on the level of 54% for directional QD emission and antibunching in autocorrelation experiment confirming the fabricated QD–WG system, which is a prerequisite for using this effect in advanced applications in integrated quantum circuits.

Photonic waveguides (WGs) with integrated quantum dots (QDs) are promising building blocks for the realization of quantum-photonics circuits to implement, for instance, boson sampling and quantum gates for entanglement purification.

Interestingly, for propagating modes of light in WGs, where light is transversally confined, a unique property of spin-momentum locking can be found, which links the local polarization (transverse spin angular momentum) and the propagation direction. This interaction is the basis for chiral light-matter coupling effects with quantum emitters with many exciting opportunities to control the direction of single-photon emission and absorption in highly functional integrated quantum technology. It is worth mentioning that these opportunities are not only restricted to direct bandgap semiconductors realized in the GaAs/AlGaAs platform but are also compatible with quantum emitters in heterogeneous GaAs/Si$_3$N$_4$ structures to couple photons into complex low-loss Si-based chips. In this context, two-level systems providing well-defined polarized optical transitions, such as excitonic complexes in a QD, are most suitable for the integration into on-chip WG systems. By integrating such quantum emitters with WG systems one can achieve nonreciprocal, unidirectional photon emission and absorption to implement, for instance, optical circulators for single photons, single-photon controlled switches, controlled not (CNOT) gates, or complete spin networks.

In our previous work, we developed the necessary technology platform to systematically study directional emission from a single InGaAs QD embedded deterministically into ridge WG structures using charged excitonic states to confirm chiral coupling existing in multimode WGs. In that article, we provided both experimental results for highly off-center positioned QDs consistent with the numerical results for few quasi-transverse electric (TE) modes propagating along the WG. In the present work, using another sample based on the same wafer material, we prove the high repeatability of our technology on the one hand, and in contrast, we go beyond existing results by demonstrating the quantum nature of highly directional emission in our QD–WG structures. The applied in situ electron-beam lithography (EBL) fabrication process allows for a precharacterization of the sample to select spectrally and spatially isolated, optically bright QDs, to subsequently integrate them into linear WGs terminated e.g., with grating outcouplers. Using this in situ EBL process, we can place single QDs at well-defined off-center positions which, according to numerical simulations, exhibit maximum chiral light-matter interaction. The optical properties of deterministically processed...
QD–WG systems are studied via a spatially resolved (confocal) two-beam micro-photoluminescence (µPL) spectroscopy. In particular, we examine and compare the WG-coupled excitonic emission of deterministically integrated QDs via grating outcouplers from both ends of the linear WG. From the experimental data, we evaluate the polarized emission contrast related to directional emission induced by the chiral light-matter coupling. Moreover, the quantum nature of emission and directional coupling is studied by photon autocorrelation measurements from the outcoupler output of such a QD–WG system. Directional emission in the studied ridge WGs is expected for in-plane circularly polarized dipole emission, i.e., helicity of ±1, in highly off-center position of the integrated QDs. To determine the exact lateral displacement $\Delta x$, we perform numerical simulations based on Maxwell’s equations using a higher-order finite element method (FEM) implemented in the JCMsuite solver.[15] A 3D model of the dipole emission is schematically presented in Figure 1a, together with the WG cross-section in Figure 1b showing an 800 nm wide and 800 nm high WG structure, where dark (bright) regions correspond to GaAs (AlGaAs) layer forming distributed Bragg reflector (DBR) acting effectively as a cladding layer allowing for efficient wave guiding. After solving the problem for the preselected QD emission wavelength of 910 nm, we analyze the dipole coupling to the TE-like WG modes, which are calculated separately in a cross-sectional 2D model (see Figure 1b). The transverse magnetic (TM)-like modes in the WG are not considered here, as its out-of-plane polarized component of the electric field is dominating, and it does not contribute to the chiral coupling nor to the dipole coupling of the QD excitonic states. The chiral coupling with excitonic states is only feasible at a position where both in-plane components of the electric field of the modes, phase shifted of “π”, are present. The directional emission coupling factor $\beta \pm$ is evaluated by integrating the overlap between the dipole field distributions on both sides of the WG (see Figure 1a) with TE-modes by adding up all contributions. In the next step, we calculate the degree of anisotropy between left ($\ominus$) and right ($\oplus$) propagation direction in the WG, which is given by $(\beta_\ominus - \beta_\oplus)/(\beta_\ominus + \beta_\oplus)$ for a given right-hand or left-hand circular polarization of the dipole. Noteworthy, in the following experimental part, the calculated degree of anisotropy is compared to the degree of circular polarization $DCP = (I_\oplus - I_\ominus)/(I_\oplus + I_\ominus)$, where $I_\ominus$ is the intensity measured for right-hand (+) and left-hand (−) circular polarization for a single WG output, respectively. The latter is equivalent to the alternative approach using the selected polarization ($I_\oplus$ or $I_\ominus$) on both WG outputs, but this one can suffer from irregularities along the WG in experiment, which is not the case in the first case. In Figure 1c, the calculated degree of anisotropy is shown in dependence on the dipole position. This anisotropy shows characteristic oscillations due to higher order mode contributions below the displacement of $\Delta x = 300 \text{ nm}$, and the maximum value of $\approx 0.9$ occurs at the chiral point with a displacement of about $340 \text{ nm}$.[14] The QD–WG system is based on a semiconductor heterostructure grown by metalorganic chemical vapor deposition (MOCVD) on (100) GaAs substrate. The epitaxial growth starts with a 300 nm thick GaAs buffer layer and it is followed by 23 mirror pairs of AlGaAs/GaAs forming a DBR mirror whose 80 nm wide stopband is centered at 930 nm. In addition, the DBR mirror increases the intensity of the light emitted toward the top direction which is beneficial for the preselection of suitable QDs on the planar sample in the in situ EBL process. Moreover, in case of the WG with single D-shaped outcouplers, it enhances the photon extraction efficiency toward the collecting optics. The lower DBR is followed by the 230 nm thick GaAs one-layer cavity containing a single layer of InGaAs QDs of $\sim 10^5 \text{ cm}^{-2}$ areal density which is in the center of GaAs.

A set of ridge QD–WGs are defined using low-temperature in situ EBL realized by a customized scanning electron microscope (SEM), which is extended with a helium flow cryostat, a cathodoluminescence (CL) spectroscopy unit, and a homemade EBL pattern generator. The in situ EBL fabrication process flow is presented schematically in Figure 2a,b. The process begins with spin-coating the sample with a 100 nm thick layer of dual-tone EBL resist CSAR 62 (AR-P 6200). Then the sample is mounted to the cold finger of the SEM cryostat and is cooled down to 10 K.[16] At this temperature, we perform 2D CL scanning of the sample’s surface to select suitable QDs based on their CL intensity, emission wavelength (target wavelength about 910 nm), spatial position, as well as spectral pattern. This CL mapping process is performed using a homogeneous electron

![Diagram](image-url)

**Figure 1.** a) Calculated intensity profiles on both sides of the DBR-ridge-WG for circularly polarized dipole emission. High-emission anisotropy is achieved due to the rather large lateral displacement $\Delta x = 350 \text{ nm}$ of the dipole (marked with red circle). b) Cross-sectional view of the DBR-WG structure together with the illustration of the first five quasi-TE propagating modes that significantly contribute to the chiral coupling with the off-center dipole emitter. c) Mode coupling $\beta$-factor representing directional emission with (±) notation related to both directions along the WG together with the degree of anisotropy in dependence on the QD (dipole) off-center position.
dose of 10 mC cm\(^{-2}\) (50 ms exposure time, 0.5 nA beam-current) with a grid size of 500 nm. As a result, the resist behavior changes to soluble in the 10 \(\times\) 20 \(\mu\)m\(^2\) mapping area. In the subsequent low-temperature EBL step, a linear WG and grating outcouplers are patterned with high alignment accuracy of 30–40 nm\(^{-1}\) with respect to a preselected QDs in each mapping field. In this step, we apply gray-scale lithography with a homemade proximity correction procedure\(^{[14]}\) to avoid irregularities at the WG edges. For the EBL step, the electron dose is increased to 50 mC cm\(^{-2}\), so that the exposed areas become insoluble again.\(^{[18]}\) These areas are maintained in the subsequent development process (at 300 K in the cleanroom) and act as etch masks in the final plasma-enhanced reactive ion-etching step in which the deterministically patterned QD–WG structure is transferred into the semiconductor material. An SEM image of a QD–WG structure fabricated deterministically by in situ EBL is presented in Figure 2c.

First, we present spectroscopic data obtained during the in situ EBL process and from subsequent process evaluation. Figure 3a shows a CL map which shows pronounced local luminescence from a single QD at a wavelength of (908.0 \(\pm\) 0.5) nm. The associated CL spectrum from this QD (QD position: \(x = (9.13 \pm 0.03)\) \(\mu\)m, \(y = (5.93 \pm 0.04)\) \(\mu\)m within a 9.5 \(\times\) 9.5 \(\mu\)m\(^2\) mapping field) is shown in Figure 3b—lower panel. Next, the CL spectrum is compared to a µPL spectra taken at 8–10 K from the corresponding QD–WG structure after processing for high (>5 \(\mu\)W, red trace) and low (<0.5 \(\mu\)W, black trace) excitation powers of 787 nm diode laser, as shown in Figure 3b—upper panel. This comparison confirms that the deterministic sample processing does neither influence the wavelength nor the spectral pattern of the selected QD in a significant way. This is an important point, which will become crucial for future developments targeting at more complex QD–WG circuits with multiple spectrally matched QDs. In the presented µPL studies, we observe saturation of the emission line at 908.13 nm, suggesting recombination from neutral or charged excitonic state. The other lines are most probably originating from higher order excitonic states of the same QD, such as biexciton, or from other QDs.

Next, we explore chiral light-matter coupling and the related directional emission of a QD–WG structure, which is expected at the chiral point\(^{[2,4]}\) for highly off-center QD position, as indicated previously by numerical simulations. For this QD–WG with a width of (825 \(\pm\) 25) nm, the lateral displacement \(\Delta x\) is set to (350 \(\pm\) 25) nm during the in situ EBL process. We investigate the DCP of top emission from the outcouplers. For this specific position for the normalized Stokes parameter, one obtains \(V/I = \text{Im}(E_x^c E_x^n + E_y E_z^c)/(E_x^c + E_z^c) = \pm 1\), where \(E_x^c\) (\(E_x^n\)) is the time-dependent electric field in-plane vector component perpendicular (parallel) to the WG axis “z”. Accordingly, at the chiral point DCP = \(\pm 1\) is naturally expected.

In Figure 4, we present polarization-resolved µPL spectra for QD emission at 908.1 nm of the selected QD–WG structure. This QD transition is most probably a charged exciton state due to relatively narrow linewidth of \(\approx\)70 \(\mu\)eV, as compared with the width of 140 \(\mu\)eV for excitonic states with a fine structure splitting of 70 \(\mu\)eV.\(^{[4]}\) As a reference measurement, we first excite and detect polarization-resolved µPL emission directly from the QD, which is located as schematically presented in the inset of Figure 4. As expected, in this case, we observe rather small DCP of \((-0.06 \pm 0.01)\). Next, by a raster-scanning, we move the collection spot along the WG to the position of the left and right outcouplers, as shown in the inset of Figure 4. In this

---

**Figure 2.** a,b) Schematic view of in situ EBL processing. In the first step (a), a 2D CL map is collected over the writing field which allows one to select a bright single QD and to integrate it directly afterward into (b) the target WG structure with high alignment accuracy. c) SEM image of the fabricated QD-800 nm wide and 80 \(\mu\)m long WG with D-shaped outcouplers on both ends.

**Figure 3.** a) 2D CL map taken within a (908.0 \(\pm\) 0.5) nm wavelength range to determine the position of a suitable QD. b) Comparison of CL and µPL spectral patterns from the selected QD before and after WG integration, respectively. The high-excitation power spectra in red color is red-shifted by 0.5 nm for clarity.
case, we collect only outcoupled emission from the same QD excitonic states, which first couple to quasi-TE modes and propagate a distance of about 40 μm along the WG. In this case, we observe a pronounced DCP of (0.54 ± 0.01) for the right side and (−0.22 ± 0.01) for the left side of the WG. The opposite sign of DCP clearly confirms that in the deterministically fabricated QD–WG system with highly off-center QD position, the chiral coupling, i.e., highly directional emission of circularly polarized photons through the WG, can be achieved. Nevertheless, in our experiment, the DCP is limited to 0.54 and the absolute value at both sides is not equal. To our understanding, this could be related to: 1) not ideal spatial matching of the QD position and the related transition dipole moment with the chiral position; 2) irregularities along the WG influencing mode structure; 3) outcoupler design is not fully polarization maintaining in principle (see Supporting Information); and 4) possible backreflection from the opposite side of the WG. The last point has, however, rather minor effect in case of 80 μm long WG structure, as we expect the attenuation of directional emission of about 0.36 dB μm⁻¹ and back-reflection of about 20%, which results in ~10⁻⁴ less intensity on the opposite side of the WG. Concerning our D-shape outcoupler design, it is expected that the initial in-plane polarization of the dipole in the WG, that couples mainly to TE-like guided modes, can change due to multimodal field interference which is a function of the emitter–outcoupler distance and also depends on the outcoupler’s geometry. We verified that for the circularly polarized dipole at highly off-center position characterized by ≈90% of directionality, which is close to chiral coupling for QD charged excitonic state, the outcoupled light has circularly polarized component of 48%, linearly polarized component of 15%, tilted (±45°) linearly polarized component of 18%, and unpolarized light component of 19%. This evaluation was made with a limited numerical aperture of 0.4 in the far-field of the top emission. In case of additional filtering applied in the experimental setup adjusted for a certain circular polarization state, which is realized using a quarter-wave plate and a linear polarizer, the relevant circular polarization component is then highly sustained of ≈74% (other components are reduced by a factor of 2), and therefore, noticeably reducing the experimental error of directionality contrast evaluation (further details can be found in Supporting Information).

For future applications in quantum-photonic circuits, it is crucial to verify the quantum nature of directional emission. For this purpose, we measured the photon autocorrelation function $g^{(2)}(\tau)$ for the QD–WG structure under study. The experimental setup includes a fiber-coupled Hanbury–Brown and Twiss (HBT) configuration with two single-photon counting modules based on silicon avalanche photodiodes. In Figure 5a, we present...
the normalized $g^{(2)}(\tau)$ data taken for the emission directly from
the QD position under continuous-wave nonresonant excitation
787 nm. We fit the raw data histogram with a model function $g^{(2)}(\tau) = 1 - (1 - g^{(2)}(0))\exp(-\frac{\tau}{\tau})$ convoluted with the instru-
ment response function (IRF) describing the timing resolution of
the detection system given by $\exp(-|t|/\tau_{\text{IRF}})$ and $\tau_{\text{IRF}} = 350$ ps.
In this way, we obtain $g^{(2)}(0)_{\text{fit}} = 0.25$ and $\tau = (2.2 \pm 0.2)\text{ns}$ for
the convoluted function. By deconvolution with the IRF of the HBT
setup, the corrected $g^{(2)}(0)_{\text{dev}} = 0.20 \pm 0.06$, which is well
below the limit of the single photon emission. Next, we measured
the autocorrelation for the directional emission of the same QD
exciton line detected at the right outcoupler. In this case, we obtain
$g^{(2)}(0)_{\text{fit}} = 0.36$ and $\tau = (2.0 \pm 0.2)\text{ns}$ for the convoluted fit to the
raw data, as presented in Figure 5b, and deconvolution yields
$g^{(2)}(0)_{\text{dev}} = 0.25 \pm 0.06$. Noticeable antibunching given by
$g^{(2)}(0)_{\text{fit}} < 0.5$ confirms that our deterministic chirally coupled
QD–WG structure features directional coupling and emission at
the single-photon level, which is needed for further development
of on-chip quantum optics and nanophotonics. In future, further
reduction of the $g^{(2)}(0)$ value may be possible using quasi-
resonant p-shell excitation or strict resonant s-shell excitation
of the QD exciton, which reduces uncorrelated background emission. Interestingly, in case of strict resonant excitation, a high
suppression of the laser scattering should be feasible because
of on-chip spatial filtering of the outcoupled emission collected
with the confocal detection configuration at the grating outcoupler
at the end of the linear WG.

In summary, we presented the deterministic fabrication and
optical study of a QD–WG structure serving as a directional
single-photon source based on chiral light-matter interaction
between the off-center QD (displacement $\Delta = (50 \pm 25) \text{nm}$) and
propagating modes (chiral point expected for $\Delta x \cong 340$ nm)
of the multi-mode DBR ridge WG. Our device was fabricated via
low-temperature in situ EBL allowing us to integrate the QD
in the target off-center position of the chiral point within
30–40 nm alignment accuracy. Chiral coupling depending on
the spin configuration of excitonic state and helicity of emitted
photons is verified by polarization resolved µPL measurements
via high degree of polarization anisotropy of outcoupled emission
observed from one end of the linear WG. Performing photon autocorrelation measurements, we confirmed that quantum
nature of directional emission in terms of $g^{(2)}(0) = 0.31 \pm
0.06 < 0.5$ is maintained in our chirally coupled QD–WG sys-
tem. Our results on the controlled integration of single quantum
emitters shows a high potential to pave the way for upscaling
photonic WGs to more complex on-chip integrated quantum circuits with novel quantum functionality.

Supporting Information
Supporting Information is available from the Wiley Online Library or
from the author.

Acknowledgements
This research was supported by the Polish Ministry of Science and Higher
Education within Mobiliñośc Plus–V edycja. The research leading to these
results has received also funding from the German Research Foundation
through CRC 787 “Semiconductor Nanophotonics: Materials, Models,
Devices” and from the European Research Council under the European
Union’s Seventh Framework ERC Grant Agreement No. 615613.

Conflict of Interest
The authors declare no conflict of interest.

Keywords
chiral coupling, deterministic nanofabrication, quantum dots, single-
photon sources, waveguides

Received: March 6, 2020
Revised: May 27, 2020
Published online: June 22, 2020

[1] S. Mahmoodian, P. Lodahl, A. S. Sørensen, Phys. Rev. Lett. 2016, 117,
240501.
[2] I. Söllner, S. Mahmoodian, S. L. Hansen, L. Midolo, A. Javadi, G.
Kiršanské, T. Pregnolato, H. El-Ella, E. H. Lee, J. D. Song, S. Stobbe,
P. Lodahl, Nat. Nanotechnol. 2015, 10, 775.
[3] I. J. Luxmoore, N. A. Wasley, A. J. Ramsay, A. C. T. Thijsse, R. Oulton,
M. Hugues, S. Kasture, V. G. Achanta, A. M. Fox, M. S. Skolnick, Phys.
Rev. Lett. 2013, 110, 037402.
[4] R. J. Coles, D. M. Price, J. E. Dixon, B. Royall, E. Clarke, P. Kok,
M. S. Skolnick, A. M. Fox, M. N. Makinon, Nat. Commun. 2016,
7, 11183.
[5] P. Schnauber, J. Schall, S. Bounour, T. Hönne, S. I. Park, G. H. Ryu,
T. Heindel, S. Burger, J. D. Song, S. Rodt, S. Reitzenstein, Nano Lett.
2018, 18, 2336.
[6] M. Davaano, J. Liu, L. Sapienza, C. Z. Zhang, J. V. De Miranda
Cardoso, V. Verma, R. P. Mirin, S. W. Nam, L. Liu, K. Srinivasan,
Nat. Commun. 2017, 8, 589.
[7] M. Schwartz, E. Schmidt, U. Rengstl, F. Horung, S. Hepp, S. L. Portalupi,
K. Llin, M. Jetter, M. Siegel, P. Michler, Nano Lett. 2018,
18, 6892.
[8] S. Hepp, S. Bauer, F. Horung, M. Schwartz, S. L. Portalupi, M. Jetter,
P. Michler, Opt. Express 2018, 26, 30614.
[9] K. Y. Blokh, F. Nori, Phys. Rep. 2015, 592, 1.
[10] P. Lodahl, S. Mahmoodian, S. Stobbe, A. Rauschenbeutel,
P. Schneeweiss, J. Volz, H. Pichler, P. Zoller, Nature 2017,
541, 473.
[11] J. L. O’Brien, A. Furusawa, J. Vucković, Nat. Photonics 2009, 3,
687.
[12] A. Politi, M. J. Cryan, J. G. Rarity, S. Yu, J. L. O’Brien, Science
2008, 320, 646.
[13] P. Schnauber, A. Singh, J. Schall, S. I. Park, J. D. Song, S. Rodt,
K. Srinivasan, S. Reitzenstein, M. Davaano, Nano Lett. 2019,
19, 7164.
[14] P. Mrowiński, P. Schnauber, P. Gutsche, A. Kaganskiy, J. Schall,
S. Burger, S. Rodt, S. Reitzenstein, ACS Photonics 2019, 6, 2231.
[15] J. Pomplun, S. Burger, L. Zschirichedi, F. Schmidt, Phys. Status Solidi B
2007, 244, 3419.
[16] A. Kaganskiy, T. Heuser, R. Schmidt, S. Rodt, S. Reitzenstein, J. Vac.
Sci. Technol. B 2016, 34, 061603.
[17] M. Gschrey, F. Gericke, A. Schüßler, R. Schmidt, J.-H. Schulze,
T. Heindel, S. Rodt, A. Strittmatter, S. Reitzenstein, Appl. Phys.
Lett. 2013, 102, 251113.
[18] M. Gschrey, A. Thoma, P. Schnauber, M. Seifried, R. Schmidt,
B. Wohlfeil, L. Krüger, J.-H. Schulze, T. Heindel, S. Burger, F. Schmidt,
A. Strittmatter, S. Rodt, S. Reitzenstein, Nat. Commun. 2015, 6,
7662.
[19] Ł. Dusankowski, S. H. Kwon, C. Schneider, S. Höfling, Phys. Rev. Lett.
2019, 122, 173602.