Structured light provides an additional degree of freedom for modern optics and practical applications. The effective generation of orbital angular momentum (OAM) lasing, especially at a micro- and nanoscale, could address the growing demand for information capacity. By exploiting the emerging non-Hermitian photonics design at an exceptional point, we demonstrate a microring laser producing a single-mode OAM vortex lasing with the ability to precisely define the topological charge of the OAM mode. The polarization associated with OAM lasing can be further manipulated on demand, creating a radially polarized vortex emission. Our OAM microlaser could find applications in the next generation of integrated optoelectronic devices for optical communications in both quantum and classical regimes.

Light typically consists of a stream of linearly polarized photons, traveling in a straight line and carrying a linear momentum. However, it was recognized that beyond the linear momentum, circularly polarized light carries angular momentum ($\ell$). The angular momentum associated with the polarization degree of freedom, or spin angular momentum (SAM), can take only one of two values, $\pm \hbar$. In addition to the SAM, it was also demonstrated that a light beam can carry orbital angular momentum (OAM) ($m$). Such beams possess helical phase fronts so that the Poynting vector within the beam is twisted with respect to the principal axis. This fundamental discovery of an OAM opened a new branch of optical physics, facilitating studies ranging from rotary photon drag ($\ell$), angular uncertainty relationships ($\ell$), and rotational frequency shifts ($\ell$), to spin-orbital coupling ($\ell$). The OAM degree of freedom has enabled technological advances, for example, edge-enhanced microscopy ($\ell$). Moreover, in contrast to the SAM that can take only two values, the OAM is unbounded. OAM beams are thus being considered as potential candidates for encoding information in both quantum and classical systems. The combined use of spin and orbital angular momenta is expected to enable the implementation of entirely new high-speed secure optical communication and quantum teleportation systems in a multi-dimensional space ($\ell$), satisfying the exponentially growing demand worldwide for network capacity.

To date, most of the light sources only produce relatively simple light beams with spatially homogeneous polarization and planar wavefront. Generation of the complex OAM beams usually relies on either bulk devices, such as spiral phase plates, spatial light modulators, and computer-generated holograms ($\ell$), or recently developed planar optical components, including phase modulation-based metasurfaces ($\ell$–$\ell$), q-plates ($\ell$–$\ell$), and silicon resonators ($\ell$–$\ell$). Although the science of the OAM light beams on the micro- and nanoscale is still in its early days, it is likely to advance our knowledge of light interaction with conventional and artificial atoms (e.g., quantum dots) provided that the OAM beam is focused to subwavelength dimensions ($\ell$), facilitating on-chip functionalities for micromanipulation and microfluidics. Nevertheless, it remains a grand challenge to integrate the existing approaches for OAM microlasers on a chip. For an ultimate miniaturized optical communication platform, there is a necessity of independent micro- and nanoscale laser sources ($\ell$) emitting complex vector beams carrying the OAM information.

One approach to creating an OAM laser ($\ell$) is based on combining a conventional bulk laser with additional phase-front shaping components. Despite being straightforward, this approach relies on rather different device technologies and material platforms, and therefore it is not easily scalable and integratable. On the contrary, here we integrate the advantages of semiconductor microlasers with the pronounced changes in light propagation at the exceptional point to realize a fundamentally new, compact, active OAM source on a complementary metal-oxide-semiconductor (CMOS) compatible platform. We consider a microring cavity that supports whispering gallery modes (WGMs). These modes circulate inside the cavity and carry large OAM. However, because of the mirror symmetry of a ring cavity, clockwise and counterclockwise eigen-WGMs can be simultaneously excited, and their carried OAMs consequently cancel each other. This is evidenced by the quantized phase, taking values of either $0$ or $\pi$, azimuthally distributed in the ring, which results from the interference between two counterpropagating WGMs (fig. S1) ($\ell$). To observe the OAM of an individual WGM, it is essential to

---

**Fig. 1. Design of OAM microlaser.** (A) Schematic of the OAM microlaser on an InP substrate. The diameter of the microring resonator is 9 $\mu$m, the width is 11 $\mu$m, and the height is 15 $\mu$m (500 nm of InGaAsP and 1 $\mu$m of InP). Thirteen-nanometer Ge single-layer and 5-nm Cr/11-nm Ge bilayer structures are periodically arranged in the azimuthal direction on top of the InGaAsP/InP microring, mimicking real index and gain/loss parts of an EP modulation at $n' = n + 0.01$ to support unidirectional power circulation. The designed azimuthal order is $N = 56$ at the resonant wavelength of 1472 nm. Equidistant sidewall scatters with a total number of $M = 57$ are introduced to couple the lasing emission upward, creating an OAM vortex emission with a helical wavefront. Its topological charge is defined by $l = N - M = -1$. (B) Simulated phase distribution of emitted light. A spiral phase map for an OAM charge-one vortex is clearly demonstrated.

*These authors contributed equally to this work.
†Corresponding author. Email: feng@buffalo.edu (L.F.); natasha@buffalo.edu (N.M.L.)

---
introduce a mechanism of robust selection of either clockwise or counterclockwise mode. In conventional bulk optics, unidirectional ring lasers have been demonstrated by implementing a non-reciprocal isolator in the light path. The optical isolator breaks the reciprocity between counterpropagating waves, facilitating the desired unidirectional flow. This approach, however, is not feasible at the micro- and nanoscale, as the realization of micrometer-sized isolators is extremely challenging.

To overcome this fundamental limitation, we realize the unidirectional power circulation by introducing complex refractive-index modulations to form an exceptional point (EP) (Fig. 1A). Driven by non-Hermiticity (i.e., gain and loss in optics) (21, 22), an EP occurs when multiple eigenstates coalesce into one (23-26). In our device, EP operation is essential to obtaining OAM laser emission (20). The microring laser resonator is designed with 500-nm-thick InGaAsP multiple quantum wells on an InP substrate. The complex refractive-index grating is achieved by placing on top of InGaAsP along the azimuthal direction (θ) periodically alternate single-layer Ge and bilayer Cr/Ge structures, corresponding to the refractive index (n′) and gain/loss (n″) in the cavity, respectively:

\[ \Delta n = \begin{cases} 
  in'' & \text{for } 2\pi p < \theta < 2\pi \left( p + \frac{1}{4} \right) N \\
  n' & \text{for } 2\pi \left( p + \frac{3}{8} \right) N < \theta < 2\pi \left( p + \frac{5}{8} \right) N 
\end{cases} \]  

(1)

where \( N \) denotes the azimuthal number of the targeted WGM and \( p \) takes integer values from the set \( \{0, N - 1\} \). An EP is obtained when the amplitudes of index and gain/loss gratings are set equal (i.e., \( n'' = n' \)). At EP, the Fourier transform of the complex refractive-index modulation is one-sided, yielding one-way distributed feedback (27-29) and robust unidirectional laser emission above threshold, as shown by a detailed semiconductor rate equation analysis (20). As a result, the counterclockwise WGM unidirectionally circulates in the cavity carrying large OAM through the azimuthally continuous phase evolution (figs. S2 and S3) (20).

The OAM associated with the unidirectional power flow is extracted upward into free space by introducing sideward modulations periodically arranged along the microring perimeter (16). The azimuthal phase dependence of the targeted unidirectional \( N \)th WGM is given by \( \varphi = N\theta \). The sideward modulations coherently scatter light, with the phase continuously varying in azimuthal direction, defined by the locations of the scatters (Fig. 1A, inset). For \( M \) equidistant scatters, the locations of the scatters are given by \( \theta_s = 2\pi s/M \), where \( s \in \{0, M - 1\} \), resulting in the extracted phase \( \varphi_s = 2\pi s(N - M)/M \) that carries OAM. Because the physically meaningful phase is measured modulo 2\( \pi \), we can subtract 2\( \pi s \) from each of the extracted phases and derive

\[ \varphi_s = 2\pi s(N - M)/M \]  

(2)

Equation 2 shows that the extracted phase increases linearly from 0 to \( 2\pi(N - M) \), thereby creating a vortex beam with topological charge \( l = N - M \). Figure 1B shows the modeling result of the vortex laser emission from our OAM microlaser, where \( N = 56 \) and \( M = 57 \). The phase of

**Fig. 2. Scanning electron microscope images of OAM microlaser.** The OAM microlaser was fabricated on the InGaAsP/InP platform. Alternating Cr/Ge bilayer and Ge single-layer structures were periodically implemented in the azimuthal direction on top of the microring, presenting, respectively, the gain/loss and index modulations required for unidirectional power circulation.

**Fig. 3. Characterization of OAM lasing.** (A) Evolution of the light emission spectrum from PL to ASE, and to lasing at 1474 nm, as the peak power density of pump light was increased from 0.63, to 0.68, to 2.19 GW m\(^{-2}\), respectively. (B) Input-output laser curve, showing a lasing threshold of \( \sim 1 \text{GW m}^{-2}\). (C) Far-field intensity distribution of the laser emission exhibiting a doughnut-shaped profile, where the central dark core is due to the phase singularity at the center of the OAM vortex radiation. (D) Off-center self-interference of the OAM laser emission, showing two inverted forks (marked with arrows) located at two phase singularities. Originating from the superposition of central helical and outer quasiplanar phases intrinsically associated with OAM, the double-fork pattern confirms the OAM vortex nature of the laser radiation.
The OAM characteristics, such as the vortex stable single-mode lasing with a sideband suppression pattern would cause spatial gain inhomogeneity, leading to a decrease in the laser slope efficiency, multilateral mode operation, and unstable laser emission. In our OAM microlaser, unidirectional power flow oscillating in the cavity eliminates the unwanted spatial hole-burning effect that would be created by the interference pattern of two counterpropagating WGMs. The preferential gain saturation in the antinodes of the interference pattern would cause spatial gain inhomogeneity, leading to a decrease in the laser slope efficiency, multilateral mode operation, and unstable laser emission. In our OAM microlaser, unidirectional power flow forced at the EP modulation (fig. S3) (20) enables efficient and stable single-mode lasing with a sideband suppression ratio of ~40 dB (Fig. 3A). In the transition from broadband photoluminescence (PL), to amplified spontaneous emission (ASE), and finally to lasing (Fig. 3, A and B), the emission peak stabilized at the same resonant wavelength, demonstrating the avoidance of multimode oscillation typically existing in a microring cavity. The OAM characteristics, such as the vortex nature and the phase singularity, were characterized by analyzing the spatial intensity profile of lasing emission and its self-interference (fig. S4) (20). In the far field, we observed the intensity of lasing emission spatially distributed in a doughnut shape with a dark core in the center (Fig. 3C). The observed dark center is due to the topological phase singularity at the beam axis where the phase becomes discontinuous, as predicted in Fig. 1B. The presence of the OAM was then validated by the self-interference of two doughnut-shaped beams split from the same lasing emission. In each doughnut beam, because of its OAM, optical phase varies more markedly with a helical phase front close to the central singularity area, whereas the outer doughnut area is of a relatively uniform quasiplanar phase front. At the observation plane, we intentionally created a horizontal offset between two doughnut beams, so that the dark center of one beam overlapped with the bright doughnut area of the other, and vice versa. The resulting interference patterns between the helical and quasiplanar phase fronts revealed two inverted forks (Fig. 3D), as the quasiplanar and helical phases were reversed at the centers of two doughnuts. For both of them, the single fringe split into two at the fork dislocation, evidently confirming that the radiation from our OAM laser was an optical vortex of topological charge \( l = -1 \).

The polarization properties of the demonstrated OAM microlaser can be designed on demand. In particular, radially polarized beams, characterized by a nonuniform spatial distribution of their polarization vector, have enabled unique functionalities, such as high-spatial resolution microscopy by their sharp focusing (30). Although the conventional schemes require external optical components, such as geometric phase-based diffraction elements (9), radially polarized beams can be directly produced from our OAM microlaser. In a microring cavity, the resonant mode can be designed to be either quasi-transverse magnetic (TM) or quasi-transverse electric (TE). The radially polarized component of the quasi-TM mode is tightly confined at the microring perimeter and sensitive to sidewall modulations, facilitating the outcoupling of this mode from the laser (fig. S5) (20). Therefore, in our microring cavity, the dominant oscillating mode is designed to be a quasi-TM mode, and its scattering by the sidewall modulation results in the radially polarized OAM lasing. In experiments, the polarization state of the OAM lasing was validated. After transmission through a linear polarizer, the doughnut profile splits into two lobes aligned along the orientation of the polarizer (Fig. 4). The two lobes remained parallel to the polarization axis regardless of the rotation of the polarizer, manifesting pure radially polarized OAM lasing. Additionally, in contrast to linearly polarized OAM modes that are not compatible with optical fibers, fibers can support radially polarized OAM eigenmodes.

We have demonstrated a microring OAM laser producing an optical vortex beam with an on-demand topological charge and vector polarization states. This is enabled through combined index and gain/loss modulations at an EP, which breaks the mirror symmetry in the lasing generation dynamics and facilitates the unidirectional power oscillation. Finally, OAM vector laser beams might offer novel degrees of freedom for the next generation of optical communications in both classical and quantum regimes.

**REFERENCES AND NOTES**

20. Materials and methods are available as supplementary materials on Science Online.
ELECTROCHEMISTRY

Nanostructured transition metal dichalcogenide electrocatalysts for CO2 reduction in ionic liquid

Mohammad Asadi,1 Kibum Kim,1,2,3 Cong Liu,3s Aditya Venkata Addepalli,1 Pedram Abbasi,1 Poya Yasaee,1 Patrick Phillips,4 Amirhossein Behranginia,1 José M. Cerrato,2 Richard Haasch,6 Peter Zapot,7 Bijendra Kumar,7 Robert F. Klie,4 Jeremiah Abiade,1 Larry A. Curtiss,3† Aditya Venkata Addepalli,1 Amin Salehi-Khojin1†

Conversion of carbon dioxide (CO2) into fuels is an attractive solution to many energy and environmental challenges. However, the chemical inertness of CO2 renders many electrochemical and photochemical conversion processes inefficient. We report a transition metal dichalcogenide nanoarchitecture for catalytic electrochemical CO2 conversion to carbon monoxide (CO) in an ionic liquid. We found that tungsten diselenide nanoflakes show a current density of 18.95 milliamperes per square centimeter, CO faradaic efficiency of 24%, and CO formation turnover frequency of 0.28 s−1 at a low overpotential of 54 mV. We also applied this catalyst in a light-harvesting artificial leaf platform that concurrently oxidized water in the absence of any external potential.

CO2 reduction activities of similarly sized (~100 nm) TMDC NFs including MoS2, WSe2, MoSe2, and WSe2 were tested using a rotating disc electrode. All TMDCs were grown using a chemical vapor transport technique (33). Figure 1A shows cyclic voltammetry (CV) results of WSe2 NFs, and bulk MoS2 as well as Ag nanoparticles (Ag NPs) and bulk Ag as a representative noble-metal catalyst. All experiments were performed inside a two-compartment, three-electrode electrochemical cell (fig. S6) using an electrolyte of 50 volume percent (vol %) EMIM-BF4 and 50 vol % deionized water; this composition gives the maximum CO2 reduction activity (15). The polarization curves of all studied catalysts were obtained by sweeping potential between +0.8 and −0.764 V versus RHE (reversible hydrogen electrode; all potentials reported here are based on RHE) with a scan rate of 50 mV s−1 (Fig. 1A and fig. S8). We also performed chronoamperometry at different applied potentials for WSe2 NFs. The results indicate that the obtained current densities for all applied potentials are 10 to 20% less than the CV results with 50 mV s−1 scan rate (fig. S9). The difference is attributed to the charging current (capacitive behavior) in the CV measurements.

The CO2 reduction began at −0.164 V (overpotential of 54 mV) for WSe2 NFs, as confirmed by faradaic efficiency (FE) measurements (Fig. 1B). At this potential (overpotential of 54 mV), a current density of 18.95 mA/cm2 (normalized on the basis of geometrical surface area) was obtained for WSe2 NFs; by comparison, current densities were 0.19 mA/cm2 for bulk Ag, 1.57 mA/cm2 for Ag NPs, and 3.4 mA/cm2 for bulk MoS2. The CO formation FE for WSe2 NFs (Fig. 1B) and bulk MoS2 (12) were 24% and 3%, respectively. However, the Ag NPs and bulk Ag did not reduce CO2 at this overpotential. At −0.764 V potential, the recorded current density for WSe2 NFs was 330 mA cm−2, versus 3.3 mA cm−2 for bulk Ag, 11 mA cm−2 for Ag NPs, and 65 mA cm−2 for bulk MoS2. The CO formation turnover frequency (TOF) (Fig. 1C) (33), a measure of per-site activity of catalysts to produce CO, was 0.28 s−1 for WSe2 NFs versus 0.016 s−1 for bulk MoS2. However, this value was zero for Ag NPs, as they could not produce CO at this overpotential (54 mV). Figure 1C also shows that the CO formation TOF of WSe2 was approximately three orders of magnitude higher than that of Ag NPs in the overpotential range of 150 to 650 mV.

Gas chromatography and differential electrochemical mass spectroscopy analyses indicated that CO and H2 were the only gas-phase products (5, 11, 12, 14–16) in the potential range of 0 to −0.764 V (13). The measured FE for WSe2 NFs/IL (Fig. 1B) showed that this system is highly selective for CO formation at high potentials (−0.2 to −0.764 V). However, at smaller potentials (−0.164 to −0.2 V), it produces a mixture of CO and H2 (synthesis gas). Figure S13 shows the selectivity (FE) results of all TMDCs tested in this study (33).

The catalytic performance of TMDC NFs was compared with that of other reported catalysts (Fig. 1D) by multiplying current density (activity) by CO formation FE (selectivity). At 100 mV overpotential, the performance of WSe2 NFs exceeded that of bulk MoS2 and Ag NPs tested under identical conditions in an ionic liquid by a factor of nearly 60. The performance of WSe2 NFs also exceeds those of Au NPs (17) and Cu NPs (18) by three orders of magnitude. Additionally, at this overpotential, the performance of WSe2 exceeded that of WS2 and MoSe2 NFs by factors of 3 and 2, respectively (Fig. 1D). We also performed chronoamperometric experiments to examine the electrochemical stability of WSe2 NFs in 50:50 vol % IL/deionized water. At the applied potential of −0.364 V (0.254 V overpotential), a small decay (10%) was observed after 27 hours of continuous operation of the three-electrode two-compartment cell (fig. S14) (13).

The photochemical performance of WSe2/IL was also studied using a custom-built wireless setup. This artificial leaf mimics the photosynthesis process in the absence of any external applied potential. The cell (Fig. 2A) (13) is composed of three major segments: (i) two amorphous silicon triple-junction photovoltaic (PV-a-si-3jn) cells in series to harvest light, (ii) the WSe2/IL cocatalyst and National Science Foundation Award (DMR-1506884) that facilitated the fabrication of the device and optimization of the EP modulation.

SUPPLEMENTARY MATERIALS
www.sciencemag.org/content/353/6298/464/suppl/DC1

†These authors equally contributed to this work. ‡Corresponding author. Email: salehih@uiuc.edu (A.S.-K.); curtissla@illinois.edu (L.A.C.)

1Department of Mechanical and Industrial Engineering, University of Illinois, Chicago, IL 60607, USA. 2Department of Mechanical Engineering, Chungbuk National University, Cheongju 361-763, South Korea. 3Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, USA. 4Department of Physics, University of Illinois at Chicago, Chicago, IL 60607, USA. 5Department of Civil Engineering, University of New Mexico, Albuquerque, NM 87131, USA. 6Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA. 7Conn Center for Renewable Energy Research, University of Louisville, Louisville, KY 40292, USA.

References (31–34)

10 April 2016; accepted 30 June 2016
10.1126/science.aaf8533

Growth of semiconductors and thin film deposition. (a) A digital photograph of the artificial leaf. The artificial leaf is composed of two amorphous silicon triple-junction photovoltaic (PV-a-Si-3jn) cells in series to harvest light (i), the WSe2/IL cocatalyst and National Science Foundation Award (DMR-1506884) that facilitated the fabrication of the device and optimization of the EP modulation.
Microlasers with a twist
Structured light, in the form of helical wavefronts, provides an additional degree of freedom to encode information for optical communications. Creating light beams with the desired amount of optical angular momentum, or twist, has usually been achieved with bulk optic devices. Miao et al. demonstrate a possible route for an integrated optics approach in which a twisted-light source with a controlled amount of optical angular momentum is generated internally to the designed device structure. These microlasers could find application in telecommunication and information technologies to increase the rate of information transmission.

Science, this issue p. 464