Blending organic electron donors and acceptors yields intermolecular charge-transfer states with additional optical transitions below their optical gaps. In organic photovoltaic devices, such states play a crucial role and limit the operating voltage. Due to its extremely weak nature, direct intermolecular charge-transfer absorption often remains undetected and unused for photocurrent generation. Here, we use an optical microcavity to increase the typically negligible external quantum efficiency in the spectral region of charge-transfer absorption by more than 40 times, yielding values over 20%. We demonstrate narrowband detection with spectral widths down to 36 nm and resonance wavelengths between 810 and 1,550 nm, far below the optical gap of both donor and acceptor. The broad spectral tunability via a simple variation of the cavity thickness makes this innovative, flexible and potentially visibly transparent device principle highly suitable for integrated low-cost spectroscopic near-infrared photodetection.
Mixtures of organic semiconductors with displaced frontier orbital energies are applied in organic light-emitting diodes (OLEDs), solar cells and photodetecting devices. In molecular blends that allow for charge transfer, it is energetically favourable for an excited electron to reside on the acceptor molecule (A) and a hole to reside on the electron donating molecule (D)\(^1\). An encounter of both charges at a donor–acceptor interface results in the formation of an intermolecular charge-transfer (CT) state which can redissociate into free carriers or recombine to the ground state\(^2\). Their decay can be optimized for efficient emission as recently demonstrated for ‘exciplex’-type OLEDs with a respectable external electroluminescence quantum yield of 15% (ref. 3).

Furthermore, CT states play an important role in organic photovoltaic and photodetecting devices, as they mediate between photo-generated excitons in neat absorbers and free charges\(^4\). As low-energy recombination centres, they limit the open-circuit voltage of organic solar cells\(^5,6\) as well as the minimum achievable low-energy recombination centres, they limit the open-circuit voltage of organic solar cells\(^5,6\) as well as the minimum achievable voltage of organic solar cells\(^5,6\) as well as the minimum achievable voltage of organic solar cells\(^5,6\) as well as the minimum achievable voltage of organic solar cells\(^5,6\) as well as the minimum achievable voltage of organic solar cells\(^5,6\) as well as the minimum achievable voltage of organic solar cells\(^5,6\) as well as the minimum achievable.

Example, for spectroscopic applications\(^18,19\), (D:A) blends offer a rather broadband absorption, while a tunable absorption above 1 m where materials rarely result in devices with reasonable responsivity at 36 nm. At short circuit, we achieve EQEs exceeding 20% with full-widths at half-maximum (FWHMs) down to 3 nm.

Results

The resonant cavity device architecture and a simplified energy level diagram are shown in Fig. 1a,b. We utilize microcavities formed by a fully reflecting and a partially transmissive silver (Ag) mirror\(^22–24\). The simultaneous use as electrodes allows a compact design incorporating both optical and electrical elements. The photo-active D:A blend is sandwiched between transparent transport layers which allow selective charge extraction towards the outer electrodes, analogous to state-of-the-art organic photovoltaic diodes\(^25–28\). Within the Fabry–Pérot interferometer formed by the two Ag mirrors, electromagnetic waves with a wavelength \(\lambda_{\text{res}}\) experience constructive interference when 

\[
\lambda_{\text{res}} = 2n\ell_{\text{cav}}/j,
\]

with \(j\) being a natural number. The effective cavity thickness \(\ell_{\text{cav}}\) is determined by the average index of refraction \(n\) and the effective geometrical cavity thickness \(\ell_{\text{cav}}\) that is, the distance between both Ag mirrors extended by the field penetration into both metal layers\(^29,30\).

To demonstrate the concept, we use a photo-active blend comprising buckminsterfullerenes (C\(_{60}\)) as electron acceptor and zinc phthalocyanine (ZnPc) as electron donor whose chemical structures are depicted in Fig. 2b. Their energy levels are displaced as illustrated in Fig. 1b. The ZnPc:C\(_{60}\) blend is known to display a substantially redshifted CT state as compared to the neat absorbers\(^31\). A volume ratio of 1:1 provides a maximum contact interface between ZnPc and C\(_{60}\). A sensitive EQE spectrum of a reference photovoltaic device with minimized cavity effects is depicted in Fig. 2a as crossed green line. The measurement confirms the presence of CT absorption in the NIR above 850 nm, that is, below the optical gap of neat C\(_{60}\) (700 nm) and ZnPc (815 nm) whose absorption spectra are shown as grey lines marked as I and II, respectively.

Cavity-enhanced CT absorption. To make use of the broadband weak CT absorption, we greatly enhance the optical field in the NIR utilizing a resonant microcavity. The thicknesses of the transport and Ag layers are optimized via optical transfer-matrix simulations\(^32\) for an absorber blend of 50 nm, as shown in Supplementary Fig. 1. To reach narrowband cavity resonances in the spectral region of CT absorption, we choose transport layer thicknesses of above 60 nm which exceeds those typically used in organic solar cells\(^25–27,33\). The EQE spectrum of a device fabricated with optimum transparent electrode (18 nm) and transport layer thicknesses (both about 80 nm, more details in Supplementary Table 1) is depicted in Fig. 2a on linear and logarithmic y-axis scales.
Angular dependence of resonance. Since the device class introduced here is based on optical interference, the cavity enhancement depends on the angle of light incidence. Figure 3 shows the absorption (3a) and EQE spectra (3b) of a device resonating at 950 nm, marked as A in Fig. 2a, as a function of the angle of incidence with respect to the substrate normal. The resonance wavelength decreases for non-normal excitation, with the peak position following a parabolic dispersion, leading to a reduction in resonance wavelength of more than 50 nm for angles above 45°. As illustrated in Fig. 3c, this behaviour is reproduced by transfer-matrix simulations. Close to normal incidence, there is however a rather broad angular range, spanning from −20° to +20°, where the resonance shift does not exceed 10 nm which is substantially smaller than the FWHM of 36 nm.

Identification of parasitic absorption. In the reported devices, the peak EQE is currently limited by parasitic absorption, which decreases the number of photon transits through the D:A blend. This observation is evidenced in Fig. 4a depicting the experimentally measured device absorption (filled area) and the corresponding EQE (hatched area) of three previous ZnPc:C60 devices whose EQEs peak at 910, 950 and 995 nm, respectively. While the peak EQE significantly drops (−59%) with increasing λres, the device absorption remains almost unchanged (−3%). Therefore, parasitic absorption greatly exceeds CT absorption. More insight into the origin of the parasitic absorption at the cavity resonance is obtained from optical simulations, depicted in Fig. 4b. We identify the transmissive top electrode, where almost every second photon absorption occurs, as main responsible. Moreover, the transparent and reflective electrode together yield every second photon absorption occurs, as main responsible. Angularly and spectrally resolved behaviour of a ZnPc:C60 sample resonating at 950 nm when the angle of incidence is 0° (device marked as A in Fig. 2a). (a) Experimental device absorption. (b) Experimental EQE. (c) Simulated device absorption. All graphs are normalized. For wavelengths below 800 nm, the neat absorber sequence extinguishes light outside the microcavity (see a,c) and, therefore, efficiently reduces the photo-response in the visible spectrum (compare b).

Longer detection wavelengths. While providing the proof of principle, ZnPc:C60 has a CT absorption band limited to wavelengths not exceeding 1,100 nm. To detect photons with lower energies, we exchange ZnPc by 2,2′,6,6′-tetraphenyl-4,4′-bipyramidrine (TPDP), chemical structure in Fig. 5b) as donor with an quenches the photo-response stemming from strong absorption above the optical gap of ZnPc, as shown in Fig. 1a and Supplementary Fig. 2.
corresponding EQE as hatched area. The experimental device absorption is shown as filled area and the TPDP:C60 detectors at short circuit. The reference sample (TPDP:C60 blend) with layers above 700 nm. For the cavity-enhanced devices, the resonance wavelength minimal interference effects has negligible absorption for wavelengths around 20%, this work underlines experimentally that CT states can be rather efficiently converted into photocurrent—which is contrary to their previous perception as trap states. Given the achieved dark currents, which are still limited by extrinsic device shunts, we estimate an upper limit for the specific detectivity at the resonance wavelength of the ZnPc:C60-based detectors of 10¹¹ Jones at short circuit (details are provided in the Supplementary Methods and Supplementary Fig. 5). We expect a further reduction in noise current and enhancement in detectivity by optimizing the absorber layer thickness or introducing undoped blocking layers.

A further increase in EQE and specific detectivity is expected when improving the ratio between CT absorption and the competing parasitic absorption. A first group of approaches aims to enhance the interfacial absorption, for example, by increasing the D:A blend thickness or by exploiting intercalating D:A blends with rather high CT absorption coefficients. A second strategy consists of reducing the competing amount of parasitic absorption. As discussed in Fig. 4, the EQE of the presented ZnPc:C60 devices can be improved via transport layers with suppressed NIR absorption. A much more drastic enhancement in EQE height and FWHM is expected when replacing the conducting Ag mirrors, being the dominant source for parasitic absorption: for this purpose, low loss mirrors such as distributed Bragg reflectors or other high-quality resonators, paired with NIR transparent electrodes with a high in-plane conductivity would offer a promising perspective.

A multitude of NIR applications in biomedicine, pharmacy and agriculture relies on spectroscopic analysis—such as disease detection, determining blood concentrations of glucose, oxyhemoglobin and water, analysing and interacting with brain functions, raw material and on-line quality monitoring, or determining nutrient compositions and optimal harvest dates. Following previously reported approaches, the realization of an organic NIR spectrometer would require several different donor or acceptor materials with varying optical gaps. The replacement of the neat material extinction by the interfacial CT absorption which extends over several hundreds of nanometers provides an elegant, robust and cheap alternative: here, all detection wavelengths within the spectrometer range can be addressed solely by a thickness variation for a single D:A blend, as outlined in Fig. 5a. Especially for the analysis of chemical compositions as common task for.

Discussion
The device concept outlined in this work has great potential as an original class of organic, narrowband NIR detectors, with an easily tunable detection wavelength. It is worth emphasizing that the wavelength selectivity, provided by these devices, is explicitly due to the weakly absorbing nature of CT states: in contrast, more strongly absorbing neat material transitions do not allow such a strong cavity enhancement and narrowband resonances as observed here, except within their narrow absorption tail region (for more details see the Supplementary Discussion). Moreover, the outlined detection principle undergoes a paradigm change: while conventionally excitons diffuse from either the absorbing donor or acceptor to a joint D:A interface to decay into CT states, those steps are skipped here in favour of a direct absorption of the latter. Demonstrating detectors with EQEs around 20%, this work underlines experimentally that CT states can be rather efficiently converted into photocurrent—which is contrary to their previous perception as trap states. Given the achieved dark currents, which are still limited by extrinsic device shunts, we estimate an upper limit for the specific detectivity at the resonance wavelength of the ZnPc:C60-based detectors of 10¹¹ Jones at short circuit (details are provided in the Supplementary Methods and Supplementary Fig. 5). We expect a further reduction in noise current and enhancement in detectivity by optimizing the absorber layer thickness or introducing undoped blocking layers.

On a variation of the excitation intensity, we observe no deviation from a linear photo-response over more than 5 orders of magnitude, as shown in Supplementary Fig. 6. We measure rise and fall times (10 to 90% or reverse) of 3 and 151 ns, respectively (see Supplementary Fig. 7 and Supplementary Table 1). The latter is partly delayed by discharge dynamics of a resistor–capacitor circuit of a 0.25 mm² large device which might be accelerated by reducing the device area. However, this response time is already sufficiently short for numerous applications related to NIR photodetection such as contact-free movement detection, non-invasive subsurface vision or night vision.

Elevated HOMO level which reduces the onset energy of CT absorption. A sensitive EQE measurement of a TPDP:C60 solar cell with minimal interference effects reveals a remarkably broad and featureless CT band from 725 nm down to 1,600 nm, as shown in Supplementary Fig. 4. Utilizing the approach outlined above, we tune the cavity by thickness adjustments of both transport layers (marked as triangle down) — and for selected devices also by a thickness variation of the TPDP:C60 blend (Δ). For more details on the layer thicknesses used, we refer the reader to Supplementary Table 2. Following this strategy, we achieve a remarkably broad resonance tunability from 810 to 1,550 nm as shown in Fig. 5a. We like to emphasize that the latter detection wavelength, addressing CT states I eV below the optical gap of both neat C60 and TPDP, settles among the highest wavelengths achieved with organic photodetectors so far. Up to now, we observe rather low internal quantum efficiencies for the material blend TPDP:C60, decreasing with blend thickness from 20 to 3% However, we expect future alternative D:A combinations to achieve higher internal yields for CT excitation in this wavelength range.

Discussion
The device concept outlined in this work has great potential as an original class of organic, narrowband NIR detectors, with an
NIR spectroscopy, an even further extension of the detection wavelengths into the infrared would be desirable. Hereby, the outlined strategy will provide photosensitivity also beyond 1,550 nm on a proceeding reduction of the CT absorption onset, by an appropriate choice of the frontier energy levels of both D and A. However, using organics, we expect a detection limit at about 2 μm. Here, fundamental interatomic vibration modes will cause strong parasitic absorption and, consequently, reduce the number of photon transits in the resonator.

In summary, we introduce an innovative class of organic narrowband NIR photodetectors based on mixtures of C60 and donor materials with a high HOMO level. An optical cavity device architecture enhances the photocurrent for wavelengths within the intermolecular CT absorption band. Using mixtures of ZnPc:C60, we obtain narrowband photodetection at wavelengths below the optical gap of ZnPc and C60 with EQEs of above 20% and spectral widths down to 36 nm. For photodetectors based on TPDPC+C60 blends with a lower CT absorption onset, we demonstrate a tunability of the resonance wavelength over a strikingly broad range from 810 to 1,550 nm by simple variations of the cavity thickness. We believe that, due to its mechanical flexibility, light weight, scalability, low fabrication cost and potential transparency at visible wavelengths, the introduced device class will become a valuable candidate for integrated spectroscopic NIR photodetection.

Methods
General fabrication procedure. Precoated glass is used either as a neat rigid substrate or with a prestructured layer of 90 nm indium tin oxide (ITO; Thin Film Devices, USA) deposited on top. Flexible devices are processed on 125-μm-thick films of planarized polyethylene naphthalate (pPEN; Teonex (R) PQ41M, DuPont Teijin Films, UK). Before device deposition, the flexible pPEN substrates are covered with 20 nm of aluminium oxide (AlOx), as a gas barrier by means of plasma-enhanced atomic layer deposition (Sentech SI ALD LL, Sentech Instruments, Germany), as earlier reported in ref. 52. The subsequent layers composed of organics, fullerene, oxides and/or metals are deposited via thermal evaporation under controlled vacuum with a base pressure of 10−8 mbar (K.J. Lesker, UK). Evaporation rates, layer thicknesses and, where applicable, mixing ratios are controlled via quartz crystal microbalances, with rates not exceeding 1 Å s−1. The geometrical intersection of the bottom Ag or ITO electrode and the top Ag or aluminium electrode defines a photo-active area of 6.4 or 0.25 mm2.

After evaporation, all glass samples are covered with special encapsulation glasses which leave a sealed hollow volume filled with nitrogen above the device. A ultraviolet-cured epoxy glue (XNR 5592; Nagase ChemteX, Japan) is used to seal the sample at the rim of the encapsulation glass. All samples on flexible substrates are sealed with another flexible barrier film against oxygen and moisture. For this, a proprietary barrier glue (Tesa SE, Germany), containing a latent getter. The lamination is carried out at room temperature in inert atmosphere. AlOx (full area) onto the device. This is done using a 25-μm-thick, ultraviolet-cured, proprietary barrier glue (Tesa SE, Germany), containing a latent getter. The lamination is carried out at room temperature in inert atmosphere. AlOx, films of both flexible barriers are placed directly adjacent to the device to minimize edge diffusion.

ZnPc:C60 series. The layer sequences of all ZnPc:C60 samples are documented in Supplementary Table 1. For further details on the device structure, we refer to Supplementary Note 3. Mass-related mixing ratios are given in weight percent (wt%), whereas unitless ratios are volume specific. The utilized materials in order of deposition are: molybdenum trioxide (MoO3; Sigma-Aldrich, USA); gold (Au; General Gold und Silberscheidanstalt, Germany); as earlier reported in ref. 52. The subsequent layers composed of organics, fullerene, oxides and/or metals are deposited via thermal evaporation under controlled vacuum with a base pressure of 10−8 mbar (K.J. Lesker, UK). Evaporation rates, layer thicknesses and, where applicable, mixing ratios are controlled via quartz crystal microbalances, with rates not exceeding 1 Å s−1. The geometrical intersection of the bottom Ag or ITO electrode and the top Ag or aluminium electrode defines a photo-active area of 6.4 or 0.25 mm2.

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**Acknowledgements**

We thank the ‘Bundesministerium für Bildung und Forschung’ for funding within the scope of the projects InnoProfile 2.1 (03IP602A) and InnoProfile 2.2 (03IP602X), as well as the ‘Deutsche Forschungsgemeinschaft’ within the scope of SPI1839 (LE474/53-1). Moreover, we thank Annette Petrich for purification of the organic materials as well as Daniel Schütte, Andreas Wendel, Tobias Günther and Caroline Walde for sample preparation. We also thank Dr Mauro Furno (Novaled GmbH) and Sim4tec for providing the software for optical transfer matrix calculations. K.L. acknowledges his support as fellow of the Canadian Institute for Advanced Research (CIFAR).

**Author contributions**

B.S. and K.V. developed the concept and conceived the experiments with A.M., J.B., D.S. and C.K. Moreover, A.M. designed the mirrors and optically optimized the microcavities including simulations of suitable layer thicknesses. O.Z. synthesized the donor material TTDP. F.N. provided flexible substrates and encapsulation. B.S. and J.B. measured and optimized the device EQE. B.S., A.M. and M.B. measured or simulated the angle-dependent device characteristics. B.S. determined the experimental and simulated absorption under normal incidence. S.U. and D.S. measured the transient photocurrent. B.S. carried out measurements of the dynamic range, current–voltage characteristics, noise and specific detectivity. B.S. wrote the manuscript with feedback from K.V. Beyond, we thank Annette Petrich for purification of the organic materials as well as Daniel Schütte, Andreas Wendel, Tobias Günther and Caroline Walde for sample preparation. We also thank Dr Mauro Furno (Novaled GmbH) and Sim4tec for providing the software for optical transfer matrix calculations. K.L. acknowledges his support as fellow of the Canadian Institute for Advanced Research (CIFAR).

**Additional information**

**Supplementary Information** accompanies this paper at http://www.nature.com/naturecommunications

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**How to cite this article:** Siegmund, B. et al. Organic narrowband near-infrared photodetectors based on intermolecular charge-transfer absorption. *Nat. Commun.* **8**, 15421 doi: 10.1038/ncomms15421 (2017).

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