

Efficient narrow-band light emission from a single carbon nanotube p-n diode

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Electrically driven light emission from carbon nanotubes¹⁻⁸ could be used in nanoscale lasers⁹ and single-photon sources¹⁰, and has therefore been the focus of much research. However, high electric fields and currents have either been necessary for electroluminescence⁴⁻⁸, or have been an undesired side effect^{2,3}, leading to high power requirements and low efficiencies. Furthermore, electroluminescent linewidths have been broad enough to obscure the contributions of individual optical transitions. Here, we report electrically induced light emission from individual carbon nanotube p-n diodes. A new level of control over electrical carrier injection is achieved, reducing power dissipation by a factor of up to 1,000, and resulting in zero threshold current, negligible self-heating and high carrier-to-photon conversion efficiencies. Moreover, the electroluminescent spectra are significantly narrower (~ 35 meV) than in previous studies¹⁻⁸, allowing the identification of emission from free and localized excitons.

The basic building blocks of almost all of today's optoelectronic devices are p-n junction diodes, including photodetectors, light-emitting diodes (LEDs) and lasers. The demonstration of light emission from carbon nanotube p-n diodes is thus a fundamental step towards a possible technological use of nanotubes as nanometre-scale light sources. Here, we provide the first such demonstration. In our nanotube diodes (Fig. 1a), the p- and n-type regions are formed using the electrostatic doping technique introduced by Lee and colleagues¹¹. For details, refer to the Methods.

In Fig. 1b we present the I - V characteristics of a single-walled carbon nanotube diode under two different biasing conditions. The dashed green curve shows the drain-source current I_{DS} versus drain-source voltage V_{DS} when both gate biases are negative: $V_{GS1} = V_{GS2} = -8$ V. The tube then behaves as a p-type resistor and a symmetric, almost ohmic conduction behaviour is observed. The deviation from a completely linear I - V characteristic (dotted curve) at low V_{DS} is attributed to a voltage drop at the Schottky contacts between the metal electrodes and the nanotube. By applying gate biases of opposite polarity, a p-i-n diode is realized. The solid red line in Fig. 1b shows the I - V characteristic recorded with $V_{GS1} = -8$ V and $V_{GS2} = +8$ V. The device now clearly shows rectifying behaviour. The corresponding band structure is shown in Fig. 1c.

The left image in Fig. 2a depicts infrared emission from a device when the gate electrodes are biased at $V_{GS1} = -8$ V and $V_{GS2} = +8$ V and with a constant current of $I_{DS} = 240$ nA driven through the tube. In general, electrically excited light emission from semiconducting carbon nanotubes can be produced under ambipolar^{2,3} or unipolar^{4,5} operation. In the first case, both electrons and holes are injected simultaneously into the tube and their radiative recombination generates light. In the second case, a

single type of carrier, that is, either electrons or holes, accumulate kinetic energy in a high-field region within the device to generate excitons by means of impact excitation. The fact that no light is emitted when our devices are operated under unipolar conditions ($V_{GS1} = V_{GS2} = -8$ V; hole current; right image in Fig. 2a) shows that they are ambipolar light emitters. This is the behaviour we would generally expect an LED to show. The signal is still detectable at values of I_{DS} as low as ~ 10 nA. This is in contrast with all previous electroluminescence studies¹⁻⁸, where typically two orders of magnitude higher current levels are required to obtain light emission of comparable intensity. Moreover, the voltage drop across the intrinsic region is in the order of the bandgap (~ 1 V; see Fig. 1b), and therefore also 5-10 times smaller than in other devices¹⁻⁸, resulting, overall, in a power dissipation that is up to 1,000 times smaller. Under typical operation conditions, we estimate a power density of only ~ 0.1 W m⁻¹ in the tube, compared with the 10-100 W m⁻¹ in other devices. It is hence clear that the nanotube diodes are operated in an entirely different regime from all other electrically driven carbon nanotube light emitters to date. In fact, the power density is comparable to what is typically used in photoluminescence experiments¹², and thermal heating, which strongly influences the electroluminescence of metallic as well as semiconducting nanotubes⁷, does not have a role.

After calibrating our detection system against the infrared emission from a known black-body emitter and taking into account its collection efficiency (see Supplementary Information), we estimate an electroluminescence efficiency of $\sim 0.5 \times 10^{-4}$ to 1×10^{-4} photons per injected electron-hole pair. Given a radiative carrier lifetime of 10-100 ns in nanotubes¹³⁻¹⁵, we hence obtain a non-radiative lifetime τ_L of the order of a few picoseconds. This value is smaller than what is typically observed in photoluminescence measurements¹⁶ (~ 20 - 200 ps). It appears reasonable, however, because of the interaction with the environment and the higher carrier concentrations, which both cause an increase of the non-radiative decay rate¹⁷. We also acquired the electroluminescence spectra of our devices (for details see Methods). Figure 2b shows the spectrally dispersed emission of a nanotube diode at $I_{DS} = 200$ nA. It is composed of a single, narrow peak centred at ~ 0.635 eV, with a spectral width of ~ 50 meV (full-width at half-maximum, FWHM). Based on a correlation between the electroluminescence results with photoluminescence and Raman data¹⁸, we assign the electroluminescence peak to emission from the lowest-energy bright exciton state E_{11} in the nanotube (see Supplementary Information).

In Fig. 3a we present the results obtained from another device. As well as the dominant emission at ~ 0.755 eV (labelled X), a weaker luminescence band is observed at ~ 65 meV lower energy (LX). The possibility that X and LX originate from two separate tubes in a

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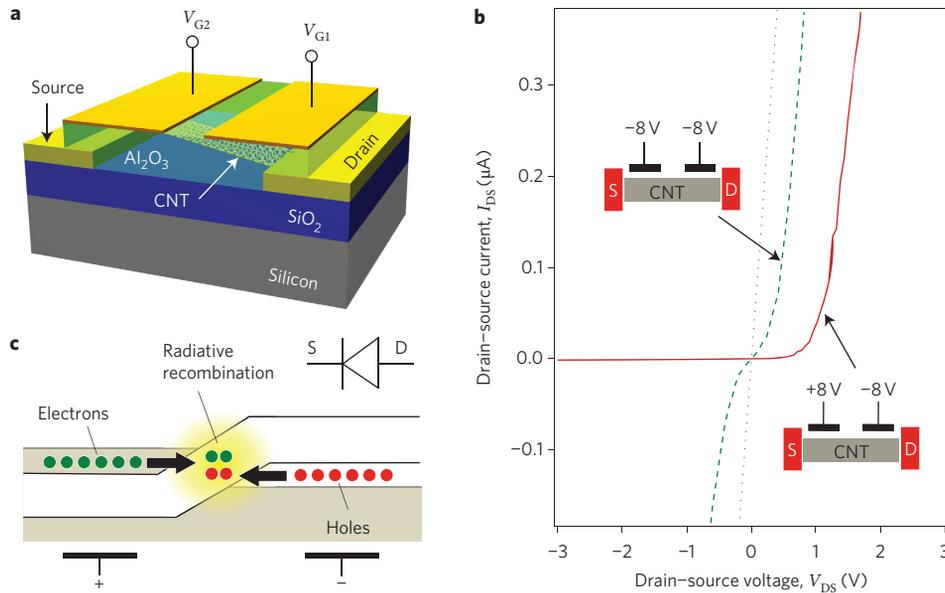


Figure 1 | Device structure and electronic characteristics. **a**, Schematic of the carbon nanotube LED. **b**, Electrical device characteristics for different biasing conditions. Solid red line: $V_{GS1} = -8$ V, $V_{GS2} = +8$ V. The nanotube is operated as a diode and shows rectifying behaviour. Dashed green line: $V_{GS1} = V_{GS2} = -8$ V. The nanotube behaves as a p-type resistor. The silicon bottom-gate was grounded during the measurements. The dotted grey line shows a completely linear I - V characteristic for comparison. **c**, Band structure of the nanotube diode when it is biased in the forward direction ($V_{DS} > 0$). Electrons and holes are injected into the intrinsic region and recombine partially radiatively and partially non-radiatively.

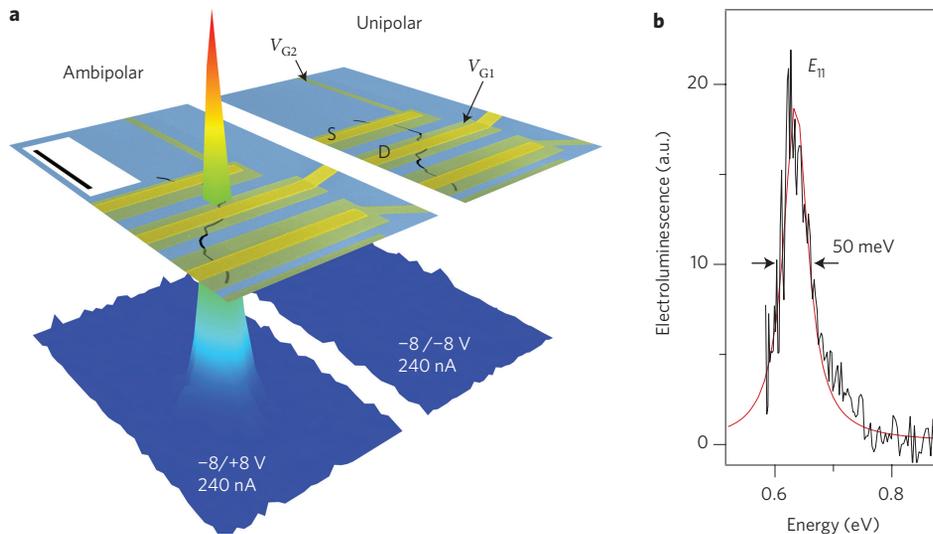


Figure 2 | Identification of the light emission mechanism. **a**, The upper plane is a scanning electron microscopy image of a carbon nanotube p-n diode. The nanotube is shown in black, the drain (D) and source (S) electrodes are yellow, and the overlapping gate electrodes are in green/yellow. Note that the rightmost electrode and gate belong to a different device and are not contacted. Scale bar, 10 μ m. The lower plane is a surface plot of the infrared emission. A microscopy image of the device (not shown) was taken under external illumination to verify that the emission is localized at the position of the tube. Infrared emission is observed at the position of the tube when the device is operated as an LED ($V_{GS1} = -8$ V, $V_{GS2} = +8$ V, $I_{DS} = 240$ nA; left image). In contrast, no emission is observed when a unipolar current of equal magnitude is driven through the nanotube ($V_{GS1} = -8$ V, $V_{GS2} = -8$ V, $I_{DS} = 240$ nA; right image). **b**, Electroluminescence spectrum of a nanotube diode at $I_{DS} = 200$ nA.

multiwalled carbon nanotube can be ruled out. The small energy spacing translates into a diameter difference that is much less than twice the graphite lattice-plane distance. To further confirm that the two emission peaks do not stem from a small bundle of nanotubes, we characterized the tube by resonance Raman spectroscopy and atomic force microscopy (AFM). In the Raman measurements, we tune the excitation laser energy between 2.0 and 2.5 eV, that is, across the E_{33} -range that corresponds to the diameter range of

our sample (see Supplementary Information). Only one single radial breathing mode (RBM) centred at $\Omega_{RBM} \approx 200$ cm^{-1} is observed, from which the nanotube diameter is determined to be¹⁹ $d_t = 248/\Omega_{RBM} \approx 1.24$ nm. From the AFM cross-section we extract a similar diameter. Those measurements, and the fact that double-peak spectra similar to the one in Fig. 3 have also been observed in other devices, support our claim that both emission features originate from a single-walled tube. Upon decreasing the gate

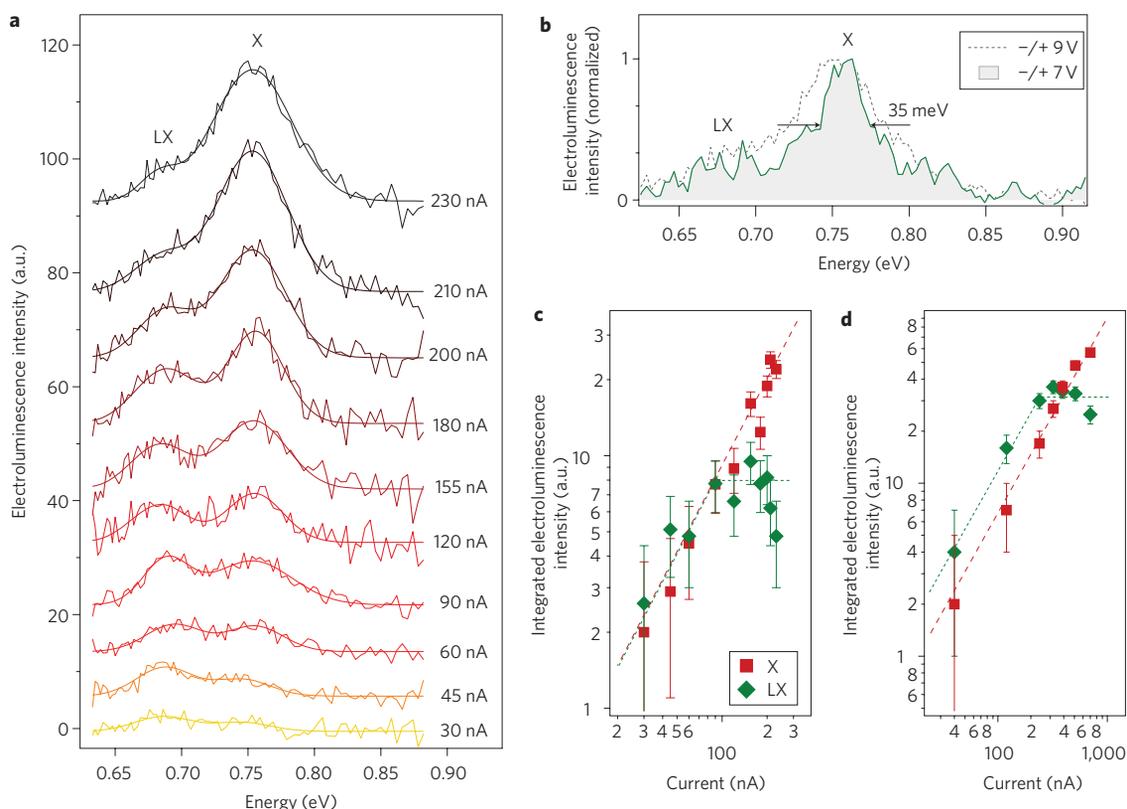


Figure 3 | Electroluminescence spectra. **a**, Electroluminescence spectrum of a nanotube diode recorded at different drain-source currents I_{DS} between 30 and 230 nA. Gate biases of $V_{GS1} = -9$ V and $V_{GS2} = +9$ V were applied. The data can be fitted well with two Gaussians and, at low currents, we extract widths of ~ 45 meV (FWHM) for the individual contributions. Besides the strong exciton emission (labelled X), a weaker satellite peak at lower energy is observed (LX). This is attributed to localized exciton emission. **b**, Comparison between electroluminescence spectra at two different gate biases (normalized). Solid green line, $V_{GS1} = -7$ V, $V_{GS2} = +7$ V; dashed grey line, $V_{GS1} = -9$ V, $V_{GS2} = +9$ V. The spectral width of the $-/+7$ V measurement is only ~ 35 meV (FWHM). **c**, Red symbols: the free exciton emission (X) shows an approximately linear increase with current. Green symbols: localized exciton emission (LX). The electroluminescence saturates as the current exceeds ~ 100 nA. The electroluminescence versus current dependence shows no threshold behaviour. The dashed lines are guides to the eye. **d**, Same as **c**, but for a different device.

bias voltages from $-/+9$ V to $-/+7$ V, the doping in the p- and n-regions decreases and so does the infrared emission intensity (for the same V_{DS}). However, as shown in Fig. 3b, we also observe a further reduction of the spectral width. We now extract a linewidth of only ~ 35 meV (FWHM). This value is ~ 2 to 8 times smaller than what has been reported in all previous electroluminescence measurements so far^{1–8}, and approaches that typically observed in room-temperature photoluminescence¹² (~ 25 meV).

The two peaks in Fig. 3 cannot be identified as the E_{11} exciton transition and the E_{11} continuum. The exciton binding energy that we estimate for a 1.24-nm-diameter tube embedded in $\text{SiO}_2/\text{Al}_2\text{O}_3$ ($\epsilon_{\text{eff}} \approx 5.7$) is²⁰ $E_b \approx 0.12$ eV, that is, almost twice as large as the observed splitting. More importantly, the continuum transition carries only a small fraction of the spectral weight²⁰ (see also Fig. 4). We can also exclude phonon-assisted emission, because of the different current dependencies of the two peaks. Low-energy satellite peaks have repeatedly been observed in photoluminescence measurements and have been attributed to localized exciton states^{10,16,21,22}. We thus assign the peak X to ‘free’ exciton emission and LX to emission from weakly localized excitons. It is not possible to determine from our optical measurements the physical mechanism of the exciton localization. It might be due to environmental fluctuations, leading to the formation of quantum-dot-like states, or brightening of intrinsic dark states at structural defect sites¹⁴. Note that in one of our devices, the low-energy emission feature LX was initially not present, but developed after stressing the tube by passing a high current through the device. This

observation supports the assignment of LX to emission from a defect site.

Figure 3c depicts the current dependence of the X and LX emission intensities as extracted from Fig. 3a. The free exciton emission X shows a linear increase with current. This is in contrast to previous studies of electroluminescence from nanotubes^{1–8}, which exhibit current thresholds of >1 μA for light emission. The nanotube diodes thus constitute threshold-less nanoscale light emitters. The localized exciton LX rises linearly at low currents but saturates as the current exceeds ~ 100 nA. Saturation of exciton emission in nanotubes is a characteristic signature of Auger-mediated exciton–exciton annihilation^{23–25}, which is known to be strongly enhanced in tightly confined one-dimensional systems²⁶. It sets in when more than one exciton is present in the tube, that is, when the electron–hole pair injection rate $I_{DS}/2q$ (q is the electron charge) exceeds the inverse carrier lifetime τ_L^{-1} . Therefore, $\tau_L = 2q/100$ nA ≈ 3 ps, which is in agreement with the τ_L estimated from the electroluminescence efficiency above. The sudden saturation further suggests that $\tau_A^{\text{LX}} \ll \tau_L$, with τ_A^{LX} being the LX–LX annihilation lifetime. From the absence of any noticeable X saturation, on the other hand, we expect the X–X annihilation lifetime τ_A^{X} to be much longer than τ_L . In fact, following ref. 26, we estimate ($E_b = 0.12$ eV, $E_g = 0.755$ eV + E_b) $\tau_A^{\text{X}} \approx 12$ ps. The fact that the emission from defect sites is of comparable strength to the emission from the rest of the tube further points towards a strong exciton nucleation (locally increased exciton density) at the low-energy defect sites. Figure 3d depicts the results from another device.

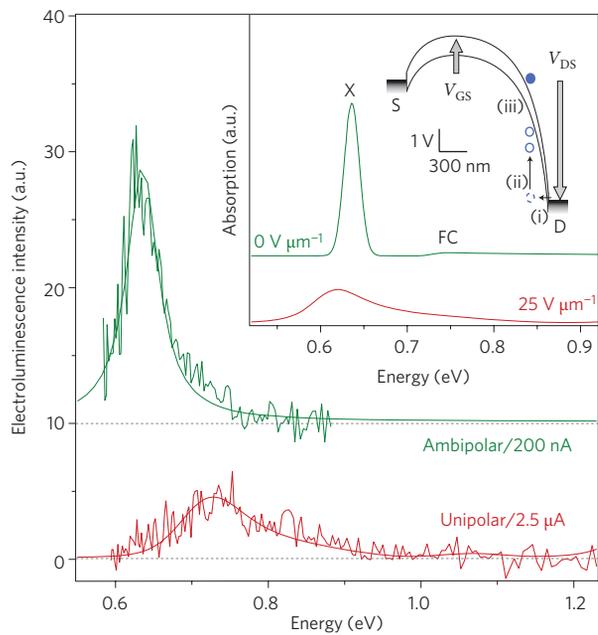


Figure 4 | Comparison between ambipolar and unipolar light emission. Green line: electroluminescence of a nanotube diode. Red line: electroluminescence emission from a FET device. The upper curve is offset for clarity. Both devices are made from the same, long tube and undergo the same processing steps, the only difference being the absence of top-gate electrodes in the FET device. Inset: calculated band structure²⁷ of the FET. In the FET emission measurements, biases of $V_{DS} = +5$ V and $V_{GS} = -20$ V were applied to the drain and (silicon) back-gate electrodes, respectively. An electric field of >25 V μm^{-1} occurs near the drain electrode. Holes are injected from the drain (i), accumulate kinetic energy in the high electric field (ii), and eventually generate electron-hole pairs by impact excitation (iii). Green line: calculated absorption spectrum of a 1.4-nm-diameter tube ($\epsilon_{\text{eff}} = 6.0$) at zero field. X is the exciton transition, FC denotes the band-to-band (free carrier) transitions. Red line: calculated absorption spectrum for a field of 25 V μm^{-1} . For direct comparison, we also overlay the calculated absorption with the measurement in the main panel. Inhomogeneous broadening was taken into account by convoluting the calculated spectra with Gaussians of 25 meV (FWHM, inset) and 50 meV (FWHM, main panel) widths. The simulation does not reproduce the experimentally observed blueshift of the FET emission with respect to X. This shift most likely arises from different dielectric environments in the two devices.

A similar behaviour is observed, but the onset of saturation now occurs at higher current (~ 250 nA), suggesting a higher concentration of defects in this tube. This is also consistent with a ~ 2 times stronger LX emission as compared to X emission at low currents. It might also be interesting to investigate the photon statistics of the LX emission, because it is believed that exciton localization plays an important role in the generation of quantum light from nanotubes¹⁰.

In Fig. 4 we compare the ambipolar emission from a diode with the unipolar emission from a back-gated field-effect transistor (FET) made out of the same, long tube. The FET emission amplitude is ~ 4 times smaller than the emission from the diode, although I_{DS} is 12.5 times higher. It is also spectrally broader (~ 180 meV) and exhibits a slightly asymmetric lineshape. The FET device was operated in the reverse bias regime, with $V_{GS} < 0 < V_{DS}$ and $|V_{GS}| > |V_{DS}|$. In this regime, holes are the majority carriers and generate electron-hole pairs by impact excitation. Most electron-hole pairs are generated at the peak field F_{max} near the drain electrode (see inset) and we estimate a lower limit of $F_{\text{max}} > V_{DS}/t_{\text{ox}} \approx 25$ V μm^{-1} , where we use the gate oxide thickness t_{ox} as the screening length²⁷. When

estimating the contribution of different broadening mechanisms to the emission linewidth we find that, under those biasing conditions, the dominant contribution is due to mixing of exciton and continuum states in the high electric field. The inset of Fig. 4 shows a simulation²⁸ of the field dependence of the optical absorption of a 1.4-nm-diameter tube. At zero field (0 V μm^{-1} ; green line), as is approximately the case in our diodes, there is no absorption in the energy range between the E_{11} exciton and the onset of the weak band-to-band absorption. The absorption, as well as the emission, are hence dominated by the E_{11} excitonic transition. In the FET (25 V μm^{-1} ; red line), however, owing to the high electric field, the exciton wavefunction mixes with the band-to-band continuum, which leads to spectral weight transfer from the excitonic peak to the continuum. The band-to-band absorption moves into the forbidden region and merges with the E_{11} exciton peak, resulting in a strongly broadened, asymmetric lineshape. At 25 V μm^{-1} , the simulated absorption extends over an energetic range of more than 150 meV. Owing to the very high carrier temperatures in those devices⁷, we expect the emission spectrum to be of comparable width to the absorption. Additional broadening mechanisms, such as Auger recombination^{23–25} and phonon broadening²⁹, will increase the width even further.

Let us finally comment on the efficiency of the nanotube LEDs. Measurements of the photoluminescence efficiency of single-walled carbon nanotubes yielded values up to^{25,30} $\sim 1 \times 10^{-2}$, whereas in our devices we obtain at most $\sim 1 \times 10^{-4}$ photons per injected electron-hole pair. This difference of about two orders of magnitude can be understood by taking the following two factors into account. First, only a fraction of electrically induced electron-hole pairs have the right spin to populate radiative singlet exciton states; $[1 + 3\exp(\Delta/kT)]$ times as many populate non-radiative triplet states (k is the Boltzmann constant, T the temperature and Δ the singlet-triplet splitting). Using literature values^{13,14} for Δ , we estimate that this effect reduces the efficiency by about an order of magnitude. Second, the short non-radiative lifetime leads to an efficiency reduction by another order of magnitude. Possible routes for improving the efficiency would therefore be brightening of the triplet states—for example, by adding magnetic nanoparticles²¹—and/or suspending the nanotube to increase the non-radiative lifetime.

Methods

The p- and n-type regions in our nanotube diodes are formed using electrostatic doping. As illustrated in Fig. 1a, two separate gate electrodes that couple to two different regions of a single-walled carbon nanotube are used. One gate is biased with a negative voltage, drawing holes into the nanotube channel, and the other gate is biased with a positive voltage, resulting in an accumulation of electrons in the channel. In this way, a p-n junction can be formed and the devices behave very much like conventional semiconductor diodes¹¹.

In a first step of device fabrication, single-walled carbon nanotubes were grown by ethanol chemical vapour deposition (3–4 nm iron oxide nanoparticles) on a highly p-doped silicon substrate with $t_{\text{ox}} = 200$ nm thick thermal silicon oxide. The nanotubes were up to tens of micrometres long, their diameters ranging from 1 to 2 nm, as determined by AFM. The tube density was kept low (~ 1 tube per $1,000$ μm^2) to prevent the formation of nanotube bundles. Standard electron-beam lithography, electron-beam evaporation and lift-off were then used to fabricate the 50-nm-thick titanium contact electrodes. Titanium was chosen because it allows line up of the Fermi level close to the middle of the nanotube bandgap and hence efficient injection of both p- and n-type carriers. The devices were then annealed in vacuum and their FET characteristics were recorded using the silicon substrate as a back-gate. The back-gated devices exhibited clearly ambipolar transfer characteristics with high on/off ratios ($> 1 \times 10^3$). A 33-nm-thick Al_2O_3 gate dielectric was then deposited on top of the sample by atomic layer deposition. As well as acting as a gate oxide, this protected the nanotube from being influenced by chemical dopants and the devices were found to be stable over a period of months. The dielectric constant of Al_2O_3 was $\epsilon_{\text{Al}_2\text{O}_3} \approx 7.5$, as determined by C - V measurements. In a second lithography step, titanium split gates were fabricated. The width of the intrinsic (ungated) region between the split gates was 1 μm . The gate field regions that produce electrostatic doping between the edges of the contact electrodes and the edges of the gates were constructed to be 1–2 μm wide. Finally, contact windows were etched in the gate oxide at the position of the drain/source

pads using phosphoric acid. A schematic of the device is shown in Fig. 1a. The sample was mounted in an evacuated probe station and all measurements were performed at room temperature.

Electroluminescence from the sample was collected with a $\times 20$ microscope objective and imaged onto a liquid-nitrogen-cooled HgCdTe infrared camera (256 \times 256 pixel; IR Laboratories). A (cooled) 2,215-nm short-pass filter was mounted in front of the camera to reduce the background black-body radiation and increase the sensitivity of the measurement. Emission spectra were acquired by placing a GRISM (combination of a grating and prism) in the beam path between the objective and the infrared camera. The emission lines of a xenon spectral calibration lamp were recorded to calibrate the system. The raw data were corrected for the spectral transmission of all optical components in the beam path and the spectral sensitivity of the detector.

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Author contributions

T.M. and M.K. were responsible for the experimental work. All authors discussed the results and commented on the manuscript.

Additional information

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