Electrically driven optical antennas

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Unlike radiowave antennas, so far optical nanoantennas cannot be fed by electrical generators. Instead, they are driven by light1 or indirectly via excited discrete states in active materials2,3 in their vicinity. Here we demonstrate the direct electrical driving of an in-plane optical antenna by the broadband quantum-shot noise of electrons tunnelling across its feed gap. The spectrum of the emitted photons is determined by the antenna geometry and can be tuned via the applied voltage. Moreover, the direction and polarization of the light emission are controlled by the antenna resonance, which also improves the external quantum efficiency by up to two orders of magnitude. The one-material planar design offers facile integration of electrical and optical circuits and thus represents a new paradigm for interfacing electrons and photons at the nanometre scale, for example for on-chip wireless communication and highly configurable electrically driven subwavelength photon sources.

Radio- and microwaves can be generated by currents that oscillate within antennas driven by high-frequency voltage sources that extend up into the 100 GHz regime. Sources for optical and infrared radiation are traditionally based on transitions between quantum states or bulky thermal sources because conventional electrical circuits are unable to generate oscillating currents with frequencies in the high terahertz regime4. As a result, the well-developed and powerful concepts of antenna theory are difficult to apply to optical radiation, the opposite of Feynman’s anticipation5. However, in 1976 it was already shown that visible light can be generated through electron tunnelling in large-area vertically stacked metal–insulator–metal (MIM) junctions6. Soon after, it was proposed theoretically that such light emission is caused by quantum-shot noise that results in broadband current fluctuations, a picture that was recently proved conclusively7,8. Theoretical considerations suggest that quantum yields of up to 10% may be achieved using this mechanism9,10. However, experimental observations of light emission using scanning tunnelling microscopy typically yield much lower efficiencies11–13.

Here we exploit quantum-shot noise to generate optical-frequency current oscillations within an in-plane antenna gap and thus create, for the first time, an electrically driven optical antenna. We show that coupling to a well-defined radiative antenna mode increases the efficiency of light emission by two orders of magnitude and provides full control over the properties of the emitted photons. As the optical antenna and tunnelling device are fully integrated, additional functionalities, such as gate electrodes, gap modifications and additional passive or active elements, may easily be incorporated. Furthermore, ultrafast amplitude and frequency modulation can be achieved.

To realize an electrically driven optical antenna the challenge is to implement a lateral tunnel junction in the feed gap of an electrically connected optical antenna on an insulating transparent substrate14. To this end, we combined the top-down focused-ion beam (FIB) structuring of single-crystalline gold flakes on glass substrates15 and the atomic force microscopy nanomanipulation of monodisperse spherical gold nanoparticles covered with a ligand shell. When pushed into a tailor-made antenna gap, the molecular spacer layer that covers the nanoparticles leads to a comparatively stable atomic-scale gap16 (see Fig. 1a) even at ambient conditions, in which all the experiments were performed.

To apply a d.c. voltage between the antenna arms we connected them at the minimum optical-field points with single-crystalline gold nanowires, which leaves the antenna resonance unchanged14.

Figure 1 | Electrically driven optical antenna. a, Electron micrograph of a lateral tunnel structure: an electrically connected single-crystalline gold nanoantenna loaded with a coated gold nanoparticle on a glass substrate. $V_{\text{a}}$, applied voltage; $e^-$, electron flow; $h\nu$, light emission. b, Tunnel gap with applied voltage. $E_f$, Fermi energy or electrochemical potential.

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These connectors extend for some micrometres before being attached to macroscopic contact pads (Supplementary Information). We fabricated a large number of devices and the respective scanning electron microscopy (SEM) images suggest that, typically, the particle is not exactly centred in the antenna gap, in particular after a voltage has been applied. Therefore, as tunnel resistances increase exponentially with gap size, for most devices the applied voltage drops over a single junction, as sketched in Fig. 1b. Owing to surface self-diffusion and the high d.c. fields, gold atoms rearrange around the gap, which over time leads to an increasing current (Supplementary Information). Given that the experiments were conducted at room temperature and ambient conditions, the light emission was sufficiently stable for intermittent measurements to be performed on the same structure for hours to days.

In Fig. 2a an exemplary current versus voltage (I–V) curve is plotted. With increasing voltage the current first increases linearly and later superlinearly. The corresponding Fowler–Nordheim representation (inset) is fully compatible with a dominating single tunnel junction that shows direct tunnelling for low and field emissions at high voltages as well as a single dip at the transition voltage $V_T$. A fit to the data using a single-barrier model is consistent with a 1.3 nm gap and a barrier height of 2.6 eV (Supplementary Information). Such reduced barrier heights are commonly observed at ambient conditions. Many reproducible I–V curves can be recorded for such a junction, although they exhibit current fluctuations typical for tunnel experiments at the nanoscale (Supplementary Information).

For sufficiently high voltages, the tunnelling of electrons is accompanied by the emission of visible photons. The emitted photons are collected through a high numerical-aperture (NA) microscope objective and directed towards a sensitive detection system (see Methods). The number of photons increases linearly with the tunnelling current. A typical far-field emission spot is overlaid with an SEM micrograph of the antenna area and depicted in Fig. 2b. The recorded Airy pattern is colocalized with the antenna and exhibits a full-width at half-maximum (FWHM) of ~350 nm, close to the diffraction limit. Hence, the device acts as an electrically driven subwavelength photon source.

The spectrum of the emitted light is expected to depend on the applied voltage and the antenna resonance. Therefore we characterized the antenna resonance by white-light dark-field spectroscopy (see Methods). The scattering spectrum shows that the particle-loaded antenna has two resonances: one at lower energies around 770 nm and a second at higher energies around 590 nm. Owing to its stronger field confinement and enhancement, only the lower energy resonance significantly enhances light emission (Supplementary Information). The influence of the applied voltage on the light emission is presented in Fig. 2c, in which we plot electroluminescence spectra for applied voltages that range from 1.3 nm to 1.4 nm and a barrier height of 1.435, $l_{\text{offset}} = 50$ pA, $l_{\text{leakage}} = 0$; for details, see the Supplementary Information).

Figure 2 | Electro-optical characterization. a, I–V plot of a connected antenna with a tunnel gap. For applied voltages above 1.5 V the light detected has an intensity that grows linearly with the current. Inset, Fowler–Nordheim representation of the same I–V data and fit (solid line) using the Simmons model (with image charges, $s = 1.27$ nm, $\phi_{\text{eff}} = 2.56$ eV, $w = 2.14$ nm, $f_{\text{CTAB}} = 1.435$, $l_{\text{offset}} = 50$ pA, $l_{\text{leakage}} = 0$; for details, see the Supplementary Information). b, Electron micrograph of a structure superimposed with the subwavelength emission spot showing an Airy pattern with a FWHM of 350 nm. A weak background illumination was used to visualize the outline of the structure. c, Electroluminescence (EL) spectra for various applied voltages (open symbols). Solid red line is the scattering spectrum of an unbiased antenna; solid coloured lines represent the calculated electroluminescence spectra obtained according to the model described in the text. A global scaling factor was used to match the experimental data. All the presented spectra are corrected for the transfer function of the optical set-up. WL, white light; Exp, experimental.
responses are almost identical. Consequently, in good approxi-
mation, it is possible to model the electroluminescence spectrum
d by multiplying the spectral density of the current fluctuations with the experimentally determined far-field scattering spectrum of the antenna, which includes both the excitation spectrum and the emission characteristics. The solid lines in Fig. 2c are calculated according to this model for different applied voltages \( V \) and are in excellent agreement with the respective electroluminescence spectra. Most importantly, the model predicts the observed blue shift of the electroluminescence and that the energy of emitted photons cannot be larger than the applied voltage when considering only linear processes (quantum cutoff)\(^2\). That the onset of photon emission at high energies appears at the applied voltage confirms the effective single-junction tunnelling model.

We next demonstrate that the observed electroluminescence spectrum can be modified by changing the antenna geometry. For comparison, we also investigated a non-resonant reference structure that consists of two wires several micrometres long between which a tunnelling gap was prepared. Figure 3 depicts electron micrographs of the corresponding structures (left) and normalized scattering as well as the matching electroluminescence spectra recorded at a sufficiently high voltage (right). The electroluminescence spectra are corrected using the transfer functions of all the optical elements and the quantum efficiency of the detector. The external quantum efficiencies (QEs) and applied voltages are stated. The left column shows electron micrographs of the corresponding structures.

![Figure 3](image1.png)

**Figure 3 | Tunability and efficiency.** a–d. Electroluminescence (open circles) and scattering spectra (solid lines) for different antenna geometries (a–c) and a non-resonant wire several micrometres long (d). All the spectra were corrected using the transfer functions of all the optical elements as well as for the quantum efficiency of the detector. The external quantum efficiencies (QEs) and applied voltages are stated. The left column shows electron micrographs of the corresponding structures.

from 1.7 to 2.0 V. Each spectrum features a peak and a high-energy cutoff that both blue shift with increasing voltage as the peak also gains intensity. A similar blue shift of the photon spectrum for increasing applied voltage has been observed before in MIM structures\(^2\). No further shift is observed for voltages sufficiently larger than the antenna resonance energy and the electroluminescence spectrum closely resembles the antenna scattering spectrum under these operating conditions (Supplementary Information). In general, the spectrum of emitted photons can be understood by considering the spectral density \( C(\omega) \) of the temporal fluctuations of the tunnelling current\(^2\), which can be expressed as \( C(\omega) = (eV - \hbar \omega) \) with spectral components at frequency \( \omega \) that reach well beyond the optical frequency range depending on the applied voltage \( V \). To model the interaction of the fluctuating tunnel current with the antenna resonance we assume that it can be represented by a sum of single-frequency dipoles located in the tunnel junction with amplitudes according to \( C(\omega) \). Photon creation is considered a two-step process. In a first step, the power extracted from these dipoles is increased by the resonantly enhanced local density of states in the antenna gap (near-field antenna response). In a second step, the antenna radiates into the far field and the emission spectrum is determined by the resonantly enhanced radiative decay rate of the antenna mode (far-field antenna response). Our single-crystalline antennas exhibit relatively narrow resonances and it has been shown that under such circumstances near- and far-field responses are almost identical\(^2\). Consequently, in good approximation, it is possible to model the electroluminescence spectrum with amplitudes according to

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\text{Electron micrographs of the corresponding structures (left) and normalized scattering as well as the matching electroluminescence spectra recorded at a sufficiently high voltage (right). The electroluminescence spectra are corrected using the transfer functions of all the optical elements and the quantum efficiency of the detector. The external quantum efficiencies (QEs) and applied voltages are stated. The left column shows electron micrographs of the corresponding structures.}
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For a quantitative comparison between resonant and non-resonant systems, we determined the external quantum efficiency of the electron-to-photon conversion (Supplementary Information) by dividing the number of emitted photons in the spectral range between 500 and 1,000 nm by the number of electrons that pass the tunnel junction (also see Fig. 3). The quantum efficiencies for the resonant antennas range from $10^{-3}$ to $10^{-4}$ depending on the detailed antenna shape and on the nanoparticle position. However, the obtained efficiency for the non-resonant wire is lower than $10^{-6}$. Consequently, the best antenna exhibits a two orders of magnitude larger efficiency compared to the non-resonant wire system and a one order of magnitude larger efficiency than typical values reported for scanning tunnelling microscope light emission at ambient conditions. Our results, therefore, clearly show the positive effect of the resonantly enhanced local density of states and the improved radiative decay rate provided by the well-defined in-plane antenna resonance. However, the relative contributions of both effects are difficult to quantify (Supplementary Information).

The antenna also governs the angular radiation pattern of the emitted photons. To obtain the radiation pattern we imaged the Fourier plane of the microscope objective (Fig. 4). The radiation pattern shows two lobes close to the critical angle similar to the pattern of a dipole in proximity to a glass surface (Supplementary Information). This is expected for the radiating mode as the charge oscillations on the antenna arms resemble that of two in-phase subdiffraction dipoles. Furthermore, the polarization of the emitted light measured after the microscope objective exhibits a $10:1$ ratio along the antenna axis (Supplementary Information). The obtained value is limited by the depolarization introduced by the high-NA collection optics.

The antenna wires not only support a resonant mode at optical frequencies, but are also used to apply a voltage across the tunnel junction that separates the antenna wires. In contrast to semiconductor or organic light-emitting devices, our device consists of a single material and its properties are not limited by discrete electronic states, but rather depend on the antenna architecture. As the fluctuations in the tunnelling current constitute a broadband source, the electroluminescence can be tuned to the blue as well as to the infrared spectral regions, for example, by using shorter and longer antennas, respectively. This implies that differently tuned devices can be integrated on-chip in close proximity on top of a wide variety of substrates. The planar geometry results in a voltage-independent gap, well-defined resonances and easy external access to the tunnel gap, as well as possibilities for extending the antenna, and electrode design.

Possible applications lie in the field of electronic and photonic hybrid circuitry. Owing to the instantaneous nature of the inelastic tunnelling process (unlike that of semiconductor diodes), the signal bandwidth of the device is not limited by the spontaneous recombination rate but by the device capacitance and the ohmic resistance of its leads. As the resistance is just around 100 Ω and the antennas are small, the capacitance is low and bandwidths up to the terahertz regime should be feasible. Large bandwidth optical transistor operation may also be realized by means of photon-assisted tunnelling or by integrating molecular switches into the tunnel junction. For quantum-communication applications, it would be exciting to imprint the fermionic nature of electrons onto photons to realize a source of single photons. For practical implementations it will be important to improve the stability further as well as the quantum efficiency of the devices. The tunnelling junction could be further stabilized by the deposition of an insulator or by exploiting a covalent bond of the molecular layer around the gold particle to the antenna arms. The quantum efficiency may be improved by enhancing both the inelastic tunnelling efficiency and the antenna efficiency (Supplementary Information). The efficiency of inelastic tunnelling is predicted to reach as high as 10% in the direct tunnelling regime. Our light-emission experiments were performed for voltages with a similar magnitude as the barrier height. In this regime the relative contribution of elastic tunnelling to the vacuum is increased. The tunnel barrier depends on the detailed shape of the gap as well as on the gap material. We expect that for optimized tunnel junctions a device operation in the direct tunnelling regime is possible. This would result in a larger quantum efficiency of the light-emission process. Cross-conjugated molecules in the gap could further increase the quantum efficiency by enhancing the inelastic tunnelling rate to 50% by quantum interference. Other types of molecules can introduce additional conduction channels, which increases the amplitude of the current noise. Last, but not least, antenna geometries that feature even higher field enhancements may be employed.

**Methods**

Methods and any associated references are available in the online version of the paper.

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Author contributions
J.K., R.K., J.P. and B.H. conceived the experiment. J.K., R.K. and J.P. designed the antennas. J.K. and M.E. designed and fabricated the electrode structure. R.K. grew the gold electrodes and transferred them. J.K. and R.K. milled the structures and performed the particle pushing. J.K. and M.E. designed and fabricated the electrode structure. R.K. grew the gold electrodes and transferred them. J.K. and R.K. milled the structures and performed the particle pushing. M.K. supervised the FIB fabrication. R.K. programmed experiment-control and data-acquisition software. J.K. and R.K. constructed the experiment, performed the measurements and analysed the data. J.K. performed the finite-difference time-domain simulations. J.K., R.K., J.P. and B.H. co-wrote the manuscript with input from all authors.

Additional information
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Competing financial interests
The authors declare no competing financial interests.
Methods

Fabrication. Single-crystalline gold flakes were grown on a borosilicate coverslip in a wet-chemical process via the reduction of chlorauric acid, HAuCl₄, in ethylene glycol. Flakes were then transferred to an electrode structure. The electrode structure was defined by optical lithography and consisted of a 20 nm chromium adhesion layer and a 100 nm gold layer. Nanoantennas were fabricated from the single-crystalline gold flakes by FIB milling (Helios Nanolab, FEI Company) using an acceleration voltage of 30 kV and a beam current of 1.5 pA. For each structure, the I–V characteristics were recorded to verify a resistance >1 TΩ (Supplementary Information). Then a diluted solution of gold nanoparticles (A11C-30-CTAB-1, Nanopartz) was drop-cast onto the antennas. To remove excess molecular surfactant, the sample was rinsed in 60 °C warm ultrapure water and exposed to oxygen plasma. Particles were manipulated by an atomic force microscope (Veeco Dimensions 3100) using a silicon cantilever (OMCL-AC240TSE, Olympus).

Optical characterization. A halogen lamp coupled to a multimode fibre served as an excitation source for the white-light dark-field scattering measurement. The light was focused on the back focal plane of an oil-immersion microscope objective (Plan-Apochromat, ×63, NA = 1.40, Zeiss) to illuminate the sample with a parallel light beam. In the detection path, direct reflection was blocked by a circular beam block, such that only the scattered light was collected. The scattered light was dispersed by a spectrometer (Shamrock 303i, 80 lines mm⁻¹ blazing at 870 nm) and detected by an electron-multiplied charge-coupled device (EMCCD) (iXon A-DU897-DC-BVF, Andor).

Electrical characterization. The electrodes were contacted by copper–beryllium probe needles (Semprex Corp.), which were mounted onto a probe head (DPP220, Cascade Microtech). I–V characteristics were recorded by a source meter unit (Keithley 2601A, Keithley Instruments Inc.) using a guard and noise shield.

Electro-optical measurements. Voltages were applied by the source meter unit (Keithley 2601A, Keithley Instruments Inc.) and electroluminescence was collected by an oil-immersion microscope objective (Plan-Apochromat, ×63, NA = 1.40, Zeiss) and detected through a spectrometer (Shamrock 303i, 80 lines mm⁻¹ blazing at 870 nm or a mirror) by an EMCCD (iXon A-DU897-DC-BVF, Andor). Typical integration times of the camera were 100 ms. The source meter unit and EMCCD camera were synchronized by a LabVIEW program to allow for a correlated data analysis.

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