Few-electron correlations after ultrafast photoemission from nanometric needle tips

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Free electrons are essential in such diverse applications as electron microscopes, accelerators and photoemission spectroscopy. However, the space charge effects of many electrons are often a problem and, when confined to extremely small space–time dimensions, even two electrons can interact strongly. Here we demonstrate that the resulting Coulomb repulsion can be highly advantageous, as it leads to strong electron–electron correlations. We show that femtosecond laser-emitted electrons from nanometric needle tips are highly anticorrelated in terms of energy because of dynamic Coulomb repulsion, with a visibility of 56%. We extract a mean energy splitting of 3.3 eV and a correlation decay time of 82 fs. The energy-filtered electrons display a sub-Poissonian number distribution with a second-order correlation function as small as $g^{(2)} = 0.34$, implying that shot-noise-reduced pulsed electron beams can be realized by simple energy filtering. We also reach the strong-field regime of laser-driven electron emission and gain insights into how the electron correlations of the different electron classes (direct or rescattered) are influenced by the strong laser fields.

Electron correlations are central to intensely investigated cooperative effects inside matter. The very nature and the relevant timescales of these effects bring together the fields of ultrafast physics and quantum (electron) optics. The direct detection of two or more correlated electrons is highly sought after. Ideally, the energy and momenta of the participating electrons would be measured directly, and photoemission spectroscopy with femtosecond time resolution is well suited to this. While the required photoemission set-ups with the proper detectors do in principle exist, it is imperative to first understand the electron correlations that arise dynamically after the electrons have left the sample (Fig. 1). For imaging ultrafast effects from small volumes in particular, these effects can quickly become dominant. We show here that for electrons that are tightly confined in space–time to the nanometre–femtosecond range, even two-particle effects are strong. Yet, the timescales are so fast that a surprisingly large average current can still be extracted before correlations due to Coulomb repulsion set in. Equally importantly, we show that these correlations in energy, in conjunction with an energy filter, can be used to attain electron beams with a sub-Poissonian counting statistics, which are highly relevant for imaging electron beam-sensitive specimens such as biological samples. Even experiments with heralded electrons are now conceivable, where the detection of one (energy-shifted) electron allows us to infer that another electron must have interacted with the sample (Extended Data Fig. 1). Sub-Poissonian electron sources can enhance the signal-to-noise ratio of many electron imaging devices, and heralded electron sources may allow novel quantum imaging modes. Our work shows strong energy correlations of electrons emitted from nanoscale solids, which is highly relevant for ultrafast electron beam applications including ultrafast electron microscopes, various kinds of time-resolved photoemission experiments and even nanophotonic particle accelerators. In close analogy with quantum optics, which deals with the particle properties of light and the quantum statistics of the photons, one may further argue that our work extends the field of quantum electron optics.

When the material under study is in the form of a nanometric needle tip, the electron source volume can be confined to extremely small length scales below ~10 nm. When the electrons are emitted with femtosecond laser pulses, an extremely high time resolution on the...
In total, 3.5% of all recorded events were two-electron events with both electrons recorded. We note that the given intensity already includes a field-enhancement factor of 3.7 obtained experimentally by an intensity sweep (see for example ref. 22).

Measuring the energy for each of the two electrons yielded the two-dimensional (2D) energy correlation map shown in Fig. 2a: the x axis shows the energy $E_1$ of one electron and the y axis the energy $E_2$ of the other, with arbitrarily chosen order. Two islands of high probability can be distinguished, one with a maximum at $E_1 = 41$ eV and simultaneously $E_2 = 39$ eV (and vice versa). At the respective axis, the energy spectra of each electron are shown (blue curves). On the x axis, we also plot a one-electron spectrum in green for comparison. While the ID spectra give no hint of correlations, the 2D map shows clear evidence of an anticorrelation: two-electron events with the same energy (that is, at the diagonal) are strongly suppressed and it is much more likely that electrons arrive with notably different energies. This anticorrelation is a signature of Coulomb repulsion between two emitted electrons within one laser pulse (in-depth discussion below). We note that we also observed spatial electron correlations, which are harder to analyse due to the specifics of our detector, and will need to be addressed in future work.

To gain quantitative insights and understand the origin of the anticorrelation in detail, we modelled our system with a semi-classical simulation. The emission process was treated quantum-mechanically based on the emission of an electron in a laser field. This provided the starting parameters for a Monte Carlo simulation of the subsequent point-particle propagation (see Methods for details). With parameters matching the experiment, particularly for a laser pulse duration of 12 fs, a tip radius of $r_{tip} = 15$ nm, an applied static field of 0.3 V nm$^{-1}$ and a laser intensity of $7.4 \times 10^{12}$ W cm$^{-2}$, we obtained the black curve in Fig. 2b, which almost perfectly matches the experimental data (Methods). In the simulation, we assumed a true random character of the emission. Hence, the excellent agreement between the experiment and simulation shows that the Coulomb interaction between the two strongly space–time-confined electrons after the emission governs the spectra, as opposed to correlation effects in the emission process. This is corroborated by the scaling of the emission probabilities for one- and two- and three-electron events (Extended Data Fig. 3). From the simulation, we can further see that the detection efficiency did not play a major role, as the shape of the simulation (black curve) in Fig. 2b is similar for simulated detection efficiencies below unity.

The statistics of the detected electrons can be further quantified with the second-order correlation function $g^{(2)}(\Delta E, \Delta t) = \frac{I_{I+I} - I_{I+I}}{I_{I+I}}$, where $n$ is the number of detected electrons within -200 ns (that is, from within one laser pulse) and $\langle n \rangle$ is the mean value of $n$. It is clear from Fig. 2a that proper energy filtering can lead to sub- and super-Poissonian statistics: at the diagonal, the two-electron coincidences show a dip, which is not the case for two uncorrelated single electrons (see inset). Thus, the resulting ratio around the diagonal between two-electron and one-electron events will become strongly reduced, leading to a reduced $g^{(2)}$ (see Extended Data Fig. 4). A similar effect has recently been observed with a two-pixel detector.

Figure 3a shows $g^{(2)}$ as function of different energy filter settings (see the Methods for details). We clearly found sub-Poissonian (red)
**Fig. 2** Energy correlation in two-electron events. 

**a.** Energy spectrum of the two-electron events. The energy of one electron is plotted versus the energy of the other. A strong anticorrelation gap is observed along the diagonal, meaning that events with the same kinetic energy are strongly suppressed. The white outline marks a 50% contour line as a guide to the eye. The blue lines at the axes represent the energy spectra of the individual electrons. The green line shows the one-electron spectrum for comparison. The width of the one-electron spectrum is governed by the nonlinear emission mechanism, as well as possible Coulomb interactions with a non-detected electron. It is clear that the individual spectra of one- and two-electron events are very similar, and correlations only show up in the 2D representation between the two-electron events. The inset shows the corresponding map (same energy axes as in the main figure) for two one-electron events from two different laser pulses, with a clear maximum on the diagonal, as expected for uncorrelated events. 

**b.** The blue curve shows a histogram of the energy difference between the two electrons for the data in a. The strong dip at zero energy difference is caused by Coulomb interactions between the two electrons after emission. For comparison, from arbitrary one-electron events, we can generate uncorrelated two-electron events (inset in a). The green curve shows the energy difference of these events and exhibits no Coulomb-induced dip. A semi-classical simulation (black) quantitatively matches the experimental data. The inset shows the difference between the real two-electron events and the mixed one-electron events, both normalized to their respective number of events. Areas where the normalized fraction of two-electron events is higher (lower) than that of mixed one-electron events are marked blue (red).

**Fig. 3** Second-order correlation maps and electron number distributions. 

**a.** Map of $g^{(2)}$ for a complete dataset (similar to that in Fig. 2 with a slightly lower mean count rate of 7.5 kHz but for all events, not only the two-electron events). $g^{(2)}$ was calculated for different subsequently applied energy filters. Filtering was done around a central energy $E_{\text{central}}$ with a defined energy width $E_{\text{window}}$. The mean energy $E_0$ of the single electron distribution was subtracted on the y axis. Depending on the filter settings, areas of lower variance (red, sub-Poissonian distribution) and higher variance (blue, super-Poissonian distribution) can be found. 

**b.** Map of $g^{(2)}$ for a simulated dataset, showing good agreement with a. The colour scale in b also applies to a, c–e. 

**c.** Hit distribution for a Poisson-distributed source with a mean value of 0.5 electrons per pulse. 

**d.** Distribution for the same mean value after Coulomb interactions using a narrowband energy filter. The variance of the hit distribution is smaller than its mean value, indicating a sub-Poissonian distribution. 

**e.** Distribution with a broadband filter and higher central energy. Now a super-Poissonian distribution with large variance is found. To allow easier comparison, the Poisson distribution (grey bars) from c are also shown in d and e.
and super-Poissonian regions (blue). For comparison we show the same map generated for simulated data (Fig. 3b). Both maps show a quantitatively almost identical behaviour. We found the minimum $g^{(2)} = 0.34$ at the central energy of the electron distribution using a filter width of 0.5 eV.

For imaging applications, the Fano factor $F = \frac{\langle n^2 \rangle - \langle n \rangle^2}{\langle n \rangle}$, the ratio of variance and mean number of electrons per laser shot, is of central importance. Immediately after emission our electron beam followed Poisson statistics where $F = 1$. Dynamically, this evolved, after proper filtering, into a sub-Poissonian distribution, leading to $F = 0.97 \pm 0.004$ in the experiment, which is close to the results of the simulations ($F = 0.88$) with a chosen mean value of 0.5 electrons per pulse before filtering (Methods). Here $F$ was only limited by the small average count rate necessary to avoid saturating our detector. With a mean number of two electrons per pulse, our simulations show that we can already achieve $F$ down to 0.68 ($g^{(2)} = 0.4$) and up to 1.48 ($g^{(2)} = 1.9$), as shown by the clear redistribution from Poisson to sub-Poisson and super-Poisson distributions (Fig. 3c–e; see Methods). Sub-Poissonian electron beams are crucial for shot-noise-reduced quantum imaging, which would be an extremely attractive feature for today’s electron imaging application because of new single-electron-counting detectors.

To obtain insights into the temporal behaviour of the correlations, we focused a beam consisting of two copies of the same laser pulse with an adjustable time delay onto the tip to trigger the emission (intensity for each pulse: $5.0 \times 10^{12}$ W cm$^{-2}$). In Fig. 4a–c, three electron energy correlation maps and their corresponding energy difference spectra are shown for three different time delays. For a time delay of $\tau = 200$ fs (Fig. 4a), no anticorrelation gap is visible because the mean temporal separation was sufficiently large to prevent a substantial Coulomb interaction. As the time difference of the laser pulses decreased ($\tau = 90$ fs, Fig. 4b), the anticorrelation gap emerged along the diagonal. For zero delay (Fig. 4c), we again observed the clear gap, this time in conjunction with a broadened energy difference spectrum, which is due to the increased instantaneous intensity and resulting strong-field effects (see discussion below).

In Fig. 4d we show electron energy difference plots in the range of $\tau = -300$ fs ... + 300 fs in the form of a 2D map. In this map we averaged vertically over blocks of $\Delta \tau = 7.6$ fs for better statistics, thus no interferometric effects are visible. The 2D map shows the anticorrelation gap as function of the delay between the two laser pulses: the gap smoothly opened up for time delays smaller than ~170 fs and vanished for larger time delays. From this plot we could extract three characteristic features shown in Fig. 4e,f: (1) the width of the energy gap, extracted by fitting a double-Gaussian function to the energy differences (three example spectra are shown in Fig. 4d), (2) the repulsion visibility and (3) $g^{(2)}$, all as function of pulse delay.

Owing to the Poissonian distribution of the emission statistics, half of the two-electron events were composed of one electron triggered by the first and one by the second laser pulse (case 1), and the other half were triggered by one laser pulse only (case 2, compare with Fig. 2).
Fig. 5 | Laser intensity dependence of the observed anticorrelation gap and strong-field effects. **a**, Energy difference spectra as a function of incident laser intensity. The intensity steps are \( I = (0.8, 0.9, 1.0, 1.2, 1.5, 1.6, 1.9, 2.3) \times 10^{13} \text{ W cm}^{-2} \) (coloured lines). As the intensity increases, the gap becomes less pronounced. The repulsion visibility of the anticorrelation gap drops with increasing intensity (inset). A shoulder, representing the famous rescattering plateau, is clearly visible in the semi-logarithmic plot. The 10 \( U_e \) cutoff for the highest intensity is at 13.3 eV, in good agreement with the expected value of 13.5 eV. c–f, Electron correlation spectra showing the energy of one electron versus that of the other for laser intensities of \( 0.8 \times 10^{13} \text{ W cm}^{-2} \) (c), \( 1.0 \times 10^{13} \text{ W cm}^{-2} \) (d), \( 1.6 \times 10^{13} \text{ W cm}^{-2} \) (e) and \( 2.3 \times 10^{13} \text{ W cm}^{-2} \) (f). The white 50% contour lines show that the energy separation becomes suppressed for higher intensities. g, Simulation results for the energy spectra as a function of start time difference \( \Delta t = t_1 - t_f \) without Coulomb interactions for the four event classes shown in h; the colour code in g also applies to g and h. Four possible event classes for two-electron events in the strong-field regime (see main text for details). i, Corresponding spectra with Coulomb repulsion between the electrons switched on. All classes clearly shift to larger \( \Delta t \). Class 2 now reaches final energies above 10 \( U_e \), a region that cannot be reached for classical point-particles without interaction (see ROI 1). Both g and i were calculated for a laser intensity of \( 1.8 \times 10^{13} \text{ W cm}^{-2} \).

independent of the total count rate. Therefore, only case-1 electrons showed a delay-dependent energy gap; case-2 electrons led to a constant offset (Fig. 4e). This offset equals roughly half of the maximum energy gap at \( t = 0 \) fs, resulting from the fact that the numbers of case-2 and case-1 electrons were similar (Methods).

By fitting a Gaussian function to the energy gap width, we inferred the correlation decay time (that is, the temporal range of the electron–electron interaction) as \( \sigma_t = 81.7 \) fs (Fig. 4e); \( \sigma_{t,rep} \) as function of the delay shows a similar temporal behaviour with \( \sigma_{t,rep} = 68.1 \) fs (Fig. 4f), coinciding with the value for the repulsion visibility \( \sigma_{vis} = 69.1 \) fs. This timescale represents the emission time delay below which the electrons show a clear anticorrelation gap. Vice versa, and intriguingly, this timescale also yields a theoretical limit to the maximum emission rate of single-electron pulses of \( f_{rep,\text{max}} = 1/\sigma_t = 12.7 \) THz for the chosen quadrupole settings. This is the highest possible rate under which a train of single electron pulses with equidistant temporal spacing can be emitted from a nanometric tip in the observed solid angle without strong mutual interactions. The maximum current corresponding to \( f_{rep,\text{max}} \) equals \( I_{\text{max}} = e \times f_{rep,\text{max}} = 1.96 \mu\text{A} \), which is surprisingly large for most electron beam applications. It represents a current limit for this type of nanometric electron emitter below which an undistorted electron beam can be achieved. Such a beam corresponds to a deterministic single-electron source where in each emission event just one electron is emitted, with a constant temporal spacing. For a Poissonian-distributed emission process, such as standard laser-triggered electron emission or d.c.-field emission, this limit cannot be reached. However, pulsed laser-triggered deterministic single-electron emitters are conceivable30–32, hence a deterministic
high-current single-electron source seems within reach. Furthermore, it might be possible that by varying the experimental parameters (for example cooling or the emission regime), the initial wave packet could be changed, which could lead to different timescales in the two-pulse measurement. This technique could therefore be used as a tool to read out the initial conditions of the wave packet during emission.

To investigate how strong-field effects affect the anticorrelation gap, we varied the incident intensity (of again single laser pulses) from $8.0 \times 10^{12}$ W cm$^{-2}$ to $2.3 \times 10^{13}$ W cm$^{-2}$, resulting in the energy difference spectra shown in Fig. 5a. We observed that with increasing laser intensity, the gap depth at $\Delta E = 0$ eV reduced, leading to a reduced repulsion visibility (inset in Fig. 5a). While the gap width stays almost constant, the individual width of the two peaks increased notably and a plateau arose (Fig. 5a,b). This plateau is the famous tell-tale feature of field-driven dynamics: for certain emission times within the laser cycle the electron is driven back to the tip and scatters elastically off it. The maximum kinetic energy a rescattered electron can gain is ten times the ponderomotive energy (the $10U$ cutoff, well known in the strong-field community). This leads to an expected cutoff energy of 13.5 eV for the highest intensity of $2.3 \times 10^{13} \text{W cm}^{-2}$ in our measurement, which is visible in Fig. 5b.

Corresponding correlation maps for four different laser intensities are shown in Fig. 5c–f. At the lowest intensity Fig. 5c, the white 50% contour line divides the spectrum into two parts. With increasing intensities both parts continuously grow together until the contour line, as well as the highest count rate regions, show almost no gap (Fig. 5f). Thus, the strong driving of the electrons in the laser field starts to mask the mutual Coulomb interaction, leading to a reduction of the anticorrelation gap.

To understand the correlated electron dynamics in the strong laser field, we carried out a 3D simulation including both strong-field effects (three-step model and multi-electron Coulomb repulsion effects (Methods). Figure 5g shows the simulation results of the final energy difference of two electrons as a function of their emission time difference with Coulomb interactions switched off. When we classified the strong-field emitted electrons into direct and rescattered electrons, we obtained four combinations (Fig. 5h): (1) the first electron leaves the tip directly and the second undergoes rescattering; (2) the first electron undergoes rescattering whereas the second is emitted directly; (3) both electrons are emitted directly; (4) both undergo rescattering. These four classes are colour-coded in Fig. 5g, where the colours match those in Fig. 5h.

Around $\Delta E = 0$ and without Coulomb interactions, we mainly found events with two rescattered or two direct electrons. We repeated the simulation using the same starting conditions for each event but included Coulomb interactions (Fig. 5i). The four events classes are slightly fuzzier, but the more prominent change is that they are shifted with respect to the case without Coulomb interactions. We then defined two regions of interest (ROI) to analyse the main experimental features, namely the broadened spectrum (ROI 1) and the reduced gap depth (ROI 2). From Fig. 5i we can see that if the first electron is a rescattered one (high final energy) followed by a direct one (smaller energy), these events result in high energy differences (ROI 1): the trailing slow direct electron pushes the fast rescattered electron to even higher energies, surpassing the 10 $U_0$ line, which is the (classical) maximum energy for a single electron. In contrast, if a slow direct electron is followed by a fast rescattered electron, the first electron slows the second; both electrons end up with similar energies and thus around $\Delta E = 0$ eV (see ROI 2). This qualitatively explains our experimental findings in the inset of Fig. 5a, depending on which electron class comes first: we observed a decrease in the contrast when increasing the intensity as the influence of the laser field on the final energy of the electrons increases with increasing intensity.

With this microscopic explanation we can quantitatively understand how the spectra evolve as a combination of (single-electron) strong-field emission physics and (two- or multi-electron) Coulomb interactions. Besides the decrease in repulsion visibility (ROI 2), we see that electrons show up at higher energies than expected from single-particle models, as shown by ROI 1. We inferred that to observe the interaction between two electrons in the clearest way, it is beneficial to work with the lowest laser intensity possible. For our experiments, the intensity required for a high-contrast gap was typically below $1 \times 10^{13} \text{W cm}^{-2}$. This marks an important difference between our tip-based experiments and electron–electron interactions in non-sequential double ionization of atoms, where the typical intensity required to observe two-electron events is $>1 \times 10^{14} \text{W cm}^{-2}$ (refs. 16, 38). We suggest that the higher intensity required for atom-based correlation experiments is one of the main reasons why the observed anticorrelation gap is so much more pronounced in our case of needle-tip emission; with atoms, the anticorrelation gap shown in Fig. 2b is virtually invisible.

Owing to the semi-classical nature of our simulations, quantum statistical effects, such as the Pauli exclusion principle, were not included. However, the probability of electrons being measured at zero energy difference can be further suppressed by the Pauli exclusion principle. Thus, the Pauli principle could add to the energy gap measured at the detector in our case as well. To estimate its strength, we numerically solved the 1D time-dependent Schrödinger equation for two electrons with Coulomb interactions (Methods). The two insets in Fig. 6 show the square modulus of the two-electron wavefunction in energy space right after emission ($t = 0 \text{ fs}$) and after $100 \text{ fs}$ of propagation. Because of spatial and energetic overlap, we observed interference effects at $t = 0 \text{ fs}$. After $100 \text{ fs}$, the two-electron wavefunction showed a strong suppression along the diagonal. This energy separation demonstrates that quantum statistic effects cannot play a role after this timescale, as both spatial and energetic overlap would be required.
When we varied the initial separation of the two wave packets in the quantum simulation, we could extract the Coulomb-induced energy gap size after 100 fs of propagation (red curve in Fig. 6). For comparison, we carried out a semi-classical 1D simulation using the same initial starting conditions (blue curve). As both curves show almost perfect overlap, we conclude that the electrons in our case mainly behave like classical point-particles following the centre of mass of their wave packets, justifying our semi-classical modelling approach. It is only possible to clearly observe Pauli suppression when the kinetic energy is much higher (hundreds of kiloelectronvolts) and the initial temporal spread is larger, as vacuum dispersion and Coulomb interactions are reduced so that the wavefunction overlap during detection is much larger. This is due to fact that the TOF difference for these two electrons is nearly identical because of the very high kinetic energy and small energy spread of these electrons.

In conclusion, we observed a strong anticorrelation behaviour in two-electron emission from metal needle tips that was clearly identified as Coulomb interactions arising dynamically after emission. This repulsion between both electrons led to a prominent energy splitting with a visibility of up to $V = 56\%$ and a gap width of $-3.3\,\text{eV}$, which is much larger than the spectral width of electron beams in standard transmission electron microscopes $\leq 1.5\%$, for example. We observed a second-order correlation of $g^{(2)} = 0.34$; a sub-Poissonian pulsed electron beam is thus straightforwardly attainable by energy filtering, which is relevant for imaging applications. We show that the anticorrelation is most prominent when the emission times of the two electrons were separated by less than ~80 fs, which we identified as the correlation decay time. This timescale gave an upper current limit for electron correlations in direct two-electron emission experiments from solids, such as superconductors, the always-present electron correlations in direct two-electron emission experiments from metal nanotips.

Our work introduces correlation measurements into ultrafast electron emission from solids. We expect our work to herald quantum-enhanced electron imaging modes, opening up the field of quantum electron optics. Furthermore, when examining strong electron correlations in direct two-electron emission experiments from solids, such as superconductors, the always-present Coulomb interactions of the free electrons could easily mask quantum-cooperative effects in the electron correlation spectra. From our measured timescale we estimated a lower limit of the required quantum-cooperative effects in the electron correlation spectra. From our measured timescale we estimated a lower limit of the required quantum-cooperative effects in the electron correlation spectra. From our measured timescale we estimated a lower limit of the required quantum-cooperative effects in the electron correlation spectra.

In the final phase of manuscript writing we became aware of similar work by R. Haindl et al. $^4$, where multi-electron correlations are studied in a transmission electron microscope.

**Online content**

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-023-02059-7.

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Methods

Experimental details

Tip handling and vacuum system. Our experiments were carried out in an ultra-high-vacuum chamber with a pressure <10^{-9} mbar. The vacuum chamber was mounted on a damped optical table, as well as our laser system, to avoid vibrations. The [310] tungsten tips were inserted into the chamber via a load-lock system. The tips were characterized in situ by field ion microscopy and cleaned in the same step by field evaporation.

Laser system and optics. We used 12 fs pulses from a commercial optical parametric chirped-pulse amplifier from Laser Quantum. It operated at a repetition rate of 200 kHz with a central wavelength of 800 nm. We use chirped mirrors and fused silica wedges for dispersion compensation. The pulses were fed into the vacuum chamber, where they were focused with an off-axis parabolic mirror with a focal length of 15 mm.

For the two-pulse measurements, we used a dispersion-balanced Mach–Zehnder interferometer to generate two copies of the laser pulse with an intensity ratio of 1:1.

Heralding of electrons and single-electron sources

One potential scheme to generate heralded electrons is shown in Extended Data Fig. 1. Here we exploited the energy gap as number filter, one can spatially separate one-electron and two-electron events. The detection of one electron at the position of two-electron events (lower trajectory in Extended Data Fig. 1) allowed us then to predict the presence of a second electron with a different energy. Such an electron can then be used for shot-noise-reduced imaging of a sample. If two-electron events showed a pronounced repulsion larger than the energy width of the single electrons, they were separated in absolute energy space. Using an energy dispersive filter, one can spatially separate one-electron and two-electron events.

The detection of one electron at the position of two-electron events (lower trajectory in Extended Data Fig. 1) allowed us then to predict the presence of a second electron with a different energy. Such an electron can then be used for shot-noise-reduced imaging of a sample. This scheme is especially attractive when there is a high likelihood that the electron is being absorbed. If the likelihood for absorption is low, the presence of a second electron with a different energy. Such an electron can then be used for shot-noise-reduced imaging of a sample.

All these schemes originate from quantum optics with photon states and are typically experimentally realized using spontaneous parametric down-conversion. Twin beams can, for example, be generated that are frequency or momentum entangled. For a narrow pump pulse the measurement of one photon directly yields the exact frequency/momentum of the second photon. Such a direct conclusion is not yet possible using our scheme, but can be enhanced for a smaller energy spread of the single electron emission. For example, narrow single-electron emission with only 16 meV can be achieved with cooled Niobium tips, as recently shown in ref. 6. Using ultrashort laser pulses and such narrowband emission, our scheme could even allow spin entanglement as the number of available states is reduced for the case because the part of the beam considered was within the coherence length, which has recently been reported to be up to a ratio of 0.36 of the beam diameter. We determined the zoom factor by placing a transmission electron microscope grid close to the tip apex, generating a shadow image with and without quadrupoles.

Finally, we note that the front multi-channel plate was a funnel-type multi-channel plate with a high quantum efficiency of 86% (ref. 47). Therefore, we neglected quantum efficiency in our evaluation, but will address possible effects in future work on sub-Poissonian statistics.

Two-pulse measurement

We obtained the energy difference by fitting the sum of two Gaussian functions to the data in each line:

\[ f(x) = b_1 \times \left[ \exp\left(-\frac{(x+b_3)^2}{b_4^2}\right) + \exp\left(-\frac{(x-b_3)^2}{b_4^2}\right) \right] \]

with fitting parameters b₁, b₂, b₃, b₄. The parameter b₃ equals half of the energy width \( E_{\text{g}} = 2 \times b_3 \) in our definition. The values of \( E_{\text{g}} \) obtained as a function of \( r \) are shown in Fig. 4e. As stated, we could infer the maximum emission rate for single electron pulses of \( f_{\text{rep, max}} = 12.2 \text{THz} \) from \( E_{\text{g}}(r) \). Although no laser system with such a high repetition rate is currently available, one possibility would be to trigger the emission of electrons by a continuous terahertz field with a frequency of 12.2 THz.

We note that in this measurement (Fig. 4e) we observed a larger energy gap at \( r = 0 \text{ fs} \) than the gap shown in Fig. 2a. We suspect that this is because the quadrupole settings were different in the two-pulse case, resulting in a different magnification. Similarly, the exact shape and state of the needle tip can introduce small changes in the shape of the repulsion gap.

As we have noted in the caption of Fig. 4, the spectral shape also changes as function of pulse delay. The more both pulses overlap, the higher the instantaneous intensity will get. We can also see in the...
intensity sweep in Fig. 5a that the increased intensity led to higher energy electrons. At some point, a plateau could even arise due to rescattering of the electrons in the strong laser field.

### 3D point-particle simulation

For the simulation, we used a custom-made point-particle trajectory simulation in MATLAB R2019a. This semi-classical simulation used a quantum-mechanical emission probability function, calculated following ref. 27 for the initial emission time. The initial position was randomly Gaussian distributed at the tip surface and the initial tangential momentum also followed a random distribution, which was chosen according to experimental parameters. The number of emitted electrons was given by Poissonian statistics. After emission, we numerically integrated the equations of motion, including the static electric field resulting from the bias voltage at the tip, the oscillating laser field and Coulomb interactions between the electrons. The equation of motion of one electron with index \( i \) is given by:

\[
t_i = \frac{e}{m_e} E_i(r_1, \ldots, r_n)
\]

with the elementary charge \( e \). The total electrical field is given as

\[
E_i = -\sum_{j \neq i} \frac{e}{4\pi\varepsilon_0} \frac{r_j - r_i}{|r_j - r_i|^3} + E_{\text{static}}(r_i) + E_{\text{laser}}(r_i).
\]

Here \( \varepsilon_0 \) is the vacuum permittivity and \( r_j \) are the coordinates of two different particles \( i \) and \( j \). For \( E_{\text{static}} \), we used the static field of a spherical capacitor, as we modelled the tip as a sphere (that is emitting in the vertical axis the determined characteristic decay length48. The horizontal axis is the mean number of totally emitted electrons, and the vertical axis the determined average after filtering. Realizing even higher count rates would offer a route to achieving small \( F \), which is important for imaging.

While one- and two-electron events can be precisely reconstructed, events with more than two electrons are very difficult to disentangle. To avoid the influence of falsely reconstructed electron events, for the sub-/super-Poisson measurements, we focused on count rates where the unfiltered distribution had a small ratio of events with more than two electrons (1.6 × 10⁻²). The shown measurement had an average of 3.3 × 10⁻² electrons per pulse, resulting in \( g^{(2)} = 1.04 \) and \( F = 1.00 \) without filtering, representing an almost perfect Poisson distribution. Using simulations we demonstrated that smaller \( F \) can be achieved by increasing the average count rate, as shown in Extended Data Fig. 5c. The horizontal axis is the mean number of totally emitted electrons, and the vertical axis the determined \( F \) after filtering at the detector plane. The individual histograms (orange) represent the electron number distribution for each simulated data point. The smaller \( F \) gets, the more the difference from a Poisson distribution becomes apparent. In comparison, we show Poissonian distributions in grey for the corresponding mean particle number after filtering. For an average of five emitted electrons per pulse at the tip, we already observed a distribution with \( F = 0.5 \) around one electron per pulse on average after filtering. Realizing even higher count rates would offer a route to achieving small \( F \), which is important for imaging.

For the experimentally determined \( F \), we estimated the error bar by splitting our dataset into four independent subsets. For each subset we calculated \( F \) and then the standard deviation of the four obtained results, resulting in \( \Delta F = \pm 0.004 \).

### Two-particle time-dependent Schrödinger equation simulation

Contrary to the standard example of quantum mechanics textbooks, we were interested in the interaction of not just one but two particles. Following the approach of ref. 51, we numerically solved the two-particle time-dependent Schrödinger equation:

\[
i \frac{\partial}{\partial t} \psi(x_1, x_2, t) = H \psi(x_1, x_2, t).
\]

where the Hamiltonian in atomic units is given by:

\[
H = -\frac{1}{2} \frac{\partial^2}{\partial x_1^2} - \frac{1}{2} \frac{\partial^2}{\partial x_2^2} + V(x_1, x_2),
\]

In our case we used a soft Coulomb potential:

\[
V(x_1, x_2) = 1/(|x_1 - x_2| + a_0)
\]

to avoid singularities at \( x_1 = x_2 \). We set \( a_0 = 1 \), which is sufficiently small that the Coulomb potential resembles the classical one over most of the region of our grid. In this calculation we did not consider static and laser fields. The two wave packets were chosen to have similar properties to the experiment: a 10 eV start energy and 2 eV energy spread. For the numerical propagation a trade-off between computational time, numerical stability and propagation time was required. That is

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also the reason why we chose 10 eV instead of 40 eV, the latter leading to the computation time and storage capacity being eight times greater. Because of the dispersion of the wave packets over time, we needed a sufficiently large grid (~700 – 1,000 Å) while the spacing (<1 Å) needed to be so small that a large enough k-space was covered, at least three times the size of the start momentum. A smaller k-space leads to reflections of the wave packets and incorrect results. To avoid the wave packets leaving the grid, we used a self-adapting co-moving grid, which allowed us to reach propagation times beyond 100 fs. The electron emission from tips was not spin selective; we therefore ended up with three antisymmetric spatial wave functions and one symmetric spatial wave function. The energy gap shown in Fig. 6 is then the incoherent sum of all parts. For quantum statistics effects an overlap both in real and k-space is necessary. Coulomb repulsion, however, separates the wave function (especially in k-space), which is why we did not observe any difference between antisymmetric and symmetric parts after 100 fs of propagation.

Data availability
Source data are provided with this paper. All other data that support the plots within this Article and other findings of this study are available from the corresponding author upon reasonable request.

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Author contributions
S.M. and J.H. performed the experiment, analysed the data and generated the plots. S.M. performed the semi-classical simulations and J.H. performed the quantum-mechanical simulations. All authors wrote the manuscript.

Competing interests
The authors declare no competing interests.

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Extended Data Fig. 1 | Measurement scheme for heralded of electrons.
Electron events triggered from a metal needle tip are energetically separated by an Omega filter. When the energy width of single electron events is smaller than the mean Coulomb energy splitting, the electrons can be separated after the omega filter. Single and two-electron events are then separated in absolute energy. The measurement of one electron in the energy region prohibited for single electron events, only possible for two-electron events, leads directly to the knowledge of the presence of a second electron. By post-selection a deterministic electron source is achieved, which can be used for quantum imaging.
Extended Data Fig. 2 | Sketch of experimental setup. Femtosecond laser pulses focused by an off-axis parabolic mirror (OAP) trigger electrons (blue) from a metal needle tip. The highly coherent electron beam is magnified by two quadrupoles by a factor of up to 10 (only one quadrupole shown here). The two quadrupoles can be moved by a 3-axis manipulation stage. The multi-hit capability of the delay-line detector allows us to measure the position x and y, and the time of flight for each electron individually.
Extended Data Fig. 3 | Power scaling of n-electron events. Shown is the laser power vs. the counts per laser shot on a double-logarithmic scale. Because of the multi-photon photoemission process, the electron emission probability $P$ follows a power law $P \propto I^{n_{\text{ph}}}$, visible by the linear scaling in the double-logarithmic representation. Here, $n_{\text{ph}}$ denotes the number of absorbed photons. The slopes are $m = 2.8 \pm 0.4$ (one electron, red), $m = 6.5 \pm 0.5$ (two electrons, blue) and $8.9 \pm 0.7$ (three electrons, green). The slope for the total emission (sum of all events) is $m = 3.3 \pm 0.4$. The linear fits were weighted by the total count rate for each laser power. The inset shows the corresponding slopes for the one, two and three electron slopes with a linear fit (black line).
Extended Data Fig. 4 | Second order correlation function $g^{(2)}(E_1, E_2) = \frac{\langle N_{E_1}N_{E_2} \rangle}{\langle N_{E_1} \rangle \langle N_{E_2} \rangle}$ calculated for the energy map shown in Fig. 2(a). A filter excluding data points with less than 5 counts per bin removes data points dominated by noise (gray mask).
**Extended Data Fig. 5** | Electron energy filtering to shape the emission statistics. (a) Explanation of the energy filter used in Fig. 3: a central energy is chosen (orange line) and then an interval of $\pm E_{\text{window}}/2$ is used as filter area. (b) Experimental data shown in Fig. 3(a), with highlighted variables. (c) Fano factor for simulations from an average of 0.1 electron per pulse to 5 electrons per pulse being emitted. The insets show the resulting energy-filtered hit-distributions after propagation including Coulomb interactions.