COULOMB-CORRELATED FEW-ELECTRON STATES IN A TRANSMISSION ELECTRON MICROSCOPE BEAM
Rudolf Haindl1,2, Armin Feist1,2, Till Domröse1,2, Marcel Möller1,2, John H. Gaida1,2, Sergey V. Yalunin1,2, and Claus Ropers1,2
1Max Planck Institute for Multidisciplinary Sciences, Am Fassberg 11, 37077 Göttingen, Germany
24th Physical Institute – Solids and Nanostructures, University of Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany

ABSTRACT
We report on the observation of Coulomb-correlated pair, triple, and quadruple free-electron states in a transmission electron microscope generated by femtosecond photoemission from a nanoscale field emitter. The electron number states exhibit energy and momentum correlations of about two electronvolts caused by strong acceleration-enhanced inter-particle energy exchange. State-sorted beam caustics reveal a longitudinal source increase and shift. We demonstrate electric-field control of transverse against energy correlations. In conjunction with apertures in the beam, this facilitates highly non-Poissonian electron beam statistics for microscopy and lithography. The high fidelity of few-electron state generation will open new experimental schemes for free-electron beams.

KEYWORDS
Electron emitters, Electron correlations, Coulomb interactions, Transmission electron microscopy

INTRODUCTION
Correlations between electrons are at the core of numerous phenomena in atomic, molecular, and solid-state systems. However, harnessing free-particle correlations remains challenging, with the recent implementation of nanoscale single-electron sources [1] and observation of antibunching in electron beams [2, 3, 4]. A powerful approach to induce strong electron-electron interactions is femtosecond-triggered photoemission from nanotips [5, 6] with high spatial and temporal confinement. Recently, femtosecond gating enabled strongly non-Poissonian beam statistics in free electron beams [4, 7, 8], with applications emerging in shot-noise-reduced electron microscopy and lithography.

In this work [8], we introduce event-based transmission electron microscopy for the detection of few-electron states from a laser-triggered nanoscale field emitter. We show that these states exhibit strong Coulomb correlations in few-electron states and discuss their generation mechanism. Furthermore, we discuss applications of beams of correlated electrons, including shot-noise reduction and electron heralding schemes.

PHOTOELECTRON GENERATION AND IMAGING

Experimental setup
The experimental work was carried out in two commercially available transmission electron microscopes (JEOL JEM 2100F and JEM F200) that have been modified to allow for photoexcitation of the electron source [8,9] (see Fig. 1a). After cooling the W(100)/ZrO emitter (radius of curvature r = 490 nm) to just below the continuous Schottky emission threshold at 1,150 K, the work function of the emitter is close to the photon energy $E_{ph} = 2.4$ eV of the photoexcitation laser. A laser pulse train with a 2 MHz repetition rate and 160 fs pulse length then generates ultrashort electron pulses by close-to-threshold laser-triggered Schottky emission. Subsequently, the photoelectrons are accelerated to a beam energy of 200 keV.

Event-based photoelectron detection
The photoelectrons are analyzed with a hybrid pixel electron detector based on the Timepix3 ASIC (EM CheeTah T3, Amsterdam Scientific Instruments). The camera is mounted behind the imaging energy filter of the microscope (see Fig. 1a) and images the spectral and spatial properties of the electrons. It records an event list of electrons with a pixel time resolution of 1.56 ns, better than the temporal spacing.
between subsequent laser pulses (500 ns). After single-electron localization [Ref. 10] from the event list, the detected electrons are matched to the generating photoemission laser pulse, and the number of electrons detected per pulse \( n = 1,2,3,4 \) is assigned (see Fig. 1b).

**Power scaling of few-electron states**

The power scaling of the few-electron states is shown in Fig. 1c. While in accordance to close-to-threshold laser-assisted Schottky emission, the single-electron \((n=1)\) rate scales linearly with laser power, the double-electron \((n=2)\) and triple-electron rate \((n=3)\) scale quadratically and cubically, respectively. This shows that few-electron states are generated by a \( n \)-photon, \( n \)-electron emission mechanism.

**COULOMB-CORRELATED FEW-ELECTRON STATES**

**Characteristic spectral features of few-electron states**

Event-averaged spectra are dominated by the \( n=1 \)-state spectrum, which exhibits a single peak at the beam energy \( E_0 = 200 \text{ keV} \) (see Figs. 1c, 3a). Strikingly, the electron pulses with \( n>1 \) exhibit a distinctive spectral shape with the number of peaks identical to the number of electrons in the pulse (see Fig. 3b-d), indicating a strong interaction between the electrons in the pulse. The \( n=2-4 \)-states are plotted with respect to the state-averaged energy \( \bar{E} \).

**Coulomb-correlated few-electron states**

Using the event-based measurement scheme, we can analyze the inter-electron interaction pulse-by-pulse. For \( n=2 \), a projection of the energies of electrons A and B onto a 2d-energy pair histogram (Fig 2a) shows the same feature as the \( n=2 \) spectrum: a strong dip at the central beam energy \( E_0 \). Therefore, two electrons almost never arrive at the detector with the same electron energy.

A particle-tracing simulation gives a quantitative explanation of the experimental results. When two electrons being injected into a static electric field with a time separation of \( 50 \text{ fs} \), the initial inter-particle Coulomb force is acceleration-enhanced. While the two electrons repel each other in the center of mass frame (difference velocity \( \Delta v = v_1 - v_2 \)), both electrons are accelerated by the external field in the laboratory frame (mean velocity \( \bar{v} \)). The final kinetic energy difference is proportional to \( \Delta v \bar{v} \). During acceleration, the inter-electron Coulomb force decrease is outweighed by an increase in the mean velocity and accumulates to a final energy difference of around 2 eV (see Fig. 3b).

Remarkably, two-dimensional energy correlations for number states \( n=3,4 \) also reveal a regular arrangement (see Fig. 3c,d), in agreement with simulations.

**State-sorted beam caustics**

Coulomb-correlations also affect the transverse beam properties. From the beam caustics shown in Fig. 4a, we identify a number state-dependent virtual source shift and increase. We attribute these two observations to transverse Coulomb deflection of electrons, which spread the \( n \)-dependent electron beam (see sketch in Fig. 4b).

---

**Figure 2:** Event-resolved spectra with a distinctive spectral shape for \( n=1-4 \) (panels a-d).

**Figure 3:** a) Energy histogram of \( n = 2 \)-states with electrons A and B. b) Particle trajectory simulation of the emitter geometry (sketch on top). The accumulated energy difference, electron velocity and Coulomb energy is plotted against travel distance of electrons. c,d) Sorted energy histogram for \( n=3,4 \) with electrons A,B,C,D.

**Figure 4:** a) State-resolved beam caustics recorded by varying the last condenser lens. b) Mutual Coulomb interactions of \( n=2 \)-states shift the virtual source in beam direction and increase the virtual source.
STATISTICAL CONTROL OF FEW-ELECTRON STATES

Electric field control of correlation gap and antibunching

As most energy difference is transferred within the emitter assembly (see inset in Fig. 5a), we analyzed how a change in extraction voltage influences the correlation. While Coulomb interactions affect longitudinal and transverse correlations, the electrostatic configuration during acceleration shapes the ratio between the two perpendicular components.

Figure 5a shows the extraction field dependence of the \( n=2 \)-energy difference function. For lower extraction fields, the correlation gap moves to lower difference energies, i.e., less correlation energy is accumulated during acceleration (see Fig. 5b). Rather, the Coulomb interaction is transferred preferably in transverse direction. As the electron beam passes beam limiting apertures before entering the column, electron trajectories with higher transverse correlation are more likely to be cut from the beam (see insets in Fig. 5b). This results in a reduced second-order correlation function \( g_2(0) \) of electrons in the same pulse (see Fig. 5b). Hence, the electric field acts as a control parameter the suppression of higher order number states.

\[ \text{Figure 5: a) Electron pair correlation functions for various extraction fields. Inset: electron optics schematic of emitter assembly. b) A decrease in extraction field reduces the correlation gap (red) and the second order correlation function at zero delay \( g_2(0) \). Inset: Coulomb deflection in conjunction with apertures causes a shift from longitudinal correlation to transverse correlation for small extraction fields. As electrons with higher transverse momentum are more likely to be cut from the beam, the transmission of \( n=2 \)-electrons at small extraction fields is reduced.} \]

Schemes for the creation of sub- and super-Poissonian electron beam statistics

Tailoring the Coulomb correlation of few-electron states offers multiple opportunities to favor electron number classes selectively. For example, an aperture placed in a beam crossover reduces the number of transmitted higher number states, thereby reducing the shot-noise in electron beams. Alternatively, spectral filtering can produce sub-Poissonian statistics by placing an energy slit in a spectrally dispersed plane, where mostly \( n=1 \) states are transmitted. Conversely, replacing the energy slit with a beam stop will mostly stop \( n=1 \) states, facilitating super-Poissonian beam statistics.

CONCLUSIONS

We introduced electron number states in transmission electron microscopy that enable new experimental schemes, such as correlated probing with multiple electrons. Furthermore, a manifold of applications arises from manipulating the beam statistics. While already a shot-noise reduced \( n=1 \)-state enhanced beam has great potential for imaging and lithography, a \( n=2 \) electron beam can serve as a source of heralded electrons, which allows for shot-noise free electron illumination. Here, one of the two electrons is measured on a detector and thereby heralds the second electron, which interacts with a sample. Thereby, the exact number of electrons that interacted with the sample can be counted. Finally, also the investigation of the Coulomb-correlation mechanism warrants further investigation. The well energy-separated few-body states could be quantum mechanically entangled, allowing them to be used as free-electron qubits.

ACKNOWLEDGEMENTS

We gratefully thank P. Kruit for fruitful discussions on stochastic Coulomb interactions. We are indebted to the members of the Göttingen UTEM team for constant support and useful discussions.

REFERENCES

[1] E. Bocquillon, et al., Science 339, 1054–1057 (2013). DOI: 10.1126/science.1232572X.
[2] H. Kiesel, A. Renz, and F. Hasselbach, Nature 418, 392–394 (2002). DOI: https://doi.org/10.1038/nature00911
[3] M. Kuwahara, et al., Phys. Rev. Lett. 126, 125501 (2021). DOI: https://doi.org/10.1103/PhysRevLett.126.125501
[4] S. Keramati, W. Brunner, T.J. Gay, H. Batelaan, Phys. Rev. Lett. 127, 180602 (2021). DOI: https://doi.org/10.1103/PhysRevLett.127.180602
[5] P. Hommelhoff, Y. Sortais, A. Aghajani-Talesh, and M. A. Kasevich, Phys. Rev. Lett. 96, 077401 (2006). DOI: https://doi.org/10.1103/PhysRevLett.96.077401.
[6] C. Ropers, et al., Phys. Rev. Lett. 98, 043907 (2007). DOI: https://doi.org/10.1103/PhysRevLett.98.043907
[7] S. Meier, J. Heimerl and P. Hommelhoff, Nat. Phys., in press (2023). DOI: https://doi.org/10.1038/s41567-023-02059-7
[8] R. Haindl, et al., Nat. Phys., in press (2023). DOI: https://doi.org/10.1038/s41567-023-02067-7
[9] A. Feist, et al., Ultramicroscopy 176, 63–73 (2017). DOI: https://doi.org/10.1016/j.ultramic.2016.12.005
[10] P. van Schayck, et al., Ultramicroscopy 219, 113091 (2020). DOI: https://doi.org/10.1016/j.ultramic.2020.113091

CONTACT

*R. Haindl, rudolf.haindl@mpipat.mpg.de