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Resolving single-electron and multi-electron distribution functions with event-based electron detectors

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Background

The combination of electron probe pulses with a synchronized optical sample excitation as applied in ultrafast transmission electron microscopy (UTEM) has enabled the investigation of ultrafast dynamics on the nanoscale [1]. Besides the traditional optical-pump/electron-probe approach, sufficiently fast event-based electron detectors, based for example on the TimePix3 chip architecture [2], can detect electron events with a temporal bin width of about 1.6 ns. Due to the stochastic electron trajectories in the sensor chip the spatial and temporal resolution is often limited, in particular at higher electron energies [3,4], and requires further data processing to overcome these limitations. In addition to the nanosecond temporal resolution of event-based detectors, such a detection scheme gives access to spatio-temporal electron-electron correlations and has been recently applied to the investigation of Coulomb-correlation within a photoelectron bunch generated by femtosecond photoemission from a Schottky emitter [5]. In general, these new kinds of detectors in principal provide a measurement of multi-electron distribution functions both in electron beams and electron pulses containing additional information with respect to averaged micrographs traditionally recorded.

Methods

To gauge the potential of event-based detectors for UTEM, the temporal response of a TimePix3 detector (Cheetah T3, Amsterdam Scientific Instruments) was characterized utilizing 200-fs electron pulses (400-kHz repetition rate, 200-keV electron energy, 1.3 electron/pulse) as generated in the Regensburg UTEM. As a photoelectron source, both a laser-driven Schottky and cold-field emitter were employed. In a first experiment, we experimentally collected a dataset of electron detection events for up to 4 x 10^6 electron pulses (Fig. 1 (a)). For each event, the TimePix3 collects relative detector position, time of arrival (ToA) and time over threshold (ToT). The events are clustered depending on their ToA and position, resulting in an average of 6.5 events per cluster (Fig. 1 (b) and (c)). The data is synchronized by assigning a timestamp to each photoemission laser pulse. A fully connected deep neural network model is trained using event data from 1.6 x 10^5 time-scrambled event cluster, so that it becomes possible to predict the ToA of the primary electron.

In a second experiment, electrons are emitted by a cold-field emitter gun (CFEG) and around 5 x 10^7 electrons are detected with the TimePix3. To find

Coulomb-correlated electron pairs, only clusters with a small mean ToA-difference (< 50 ns) from the dataset are selected. The electron pairs are statistically analysed and the influence of optical excitation, extraction fields and spatial distribution are examined.

Results

Using the non-clustered ToA events leads to a temporal spread of about 10 ns (Fig. 1 (d), grey curve). If a cluster algorithm is used and the mean ToA for each cluster is calculated, then the temporal spread can be reduced to 7 ns (Fig. 1 (d), red curve). The neural network reduces the predicted temporal distribution (for a part of the data not involved in the training) to 4.7 ns (FWHM) (Fig. 1 (d), green curve). In the case of the neural network the distribution is almost Gaussian while the ToA distribution in the previous cases shows a tail for larger ToA.

Applying closely spaced electron double-pulses with a temporal separation of 8 ns to TimePix3 detector (shorter than the intrinsic temporal resolution) shows a broadened flat top distribution (Fig. 1 (e), grey curve). With the implementation of the neural network, the two peaks can be distinctly separated (Fig. 1 (e), green curve), demonstrating the more general applicability of the approach.

Conclusion

In conclusion, we demonstrate, that the accurate prediction of the ToA by a neural network trained by femtosecond electron pulse data, resulting in an increased temporal resolution for the TimePix3 detector. This improved temporal resolution will also be helpful for the mapping of temporal correlations in the imaging of non-static nano-objects and non-trivial multi-particle correlations within electron pulses.

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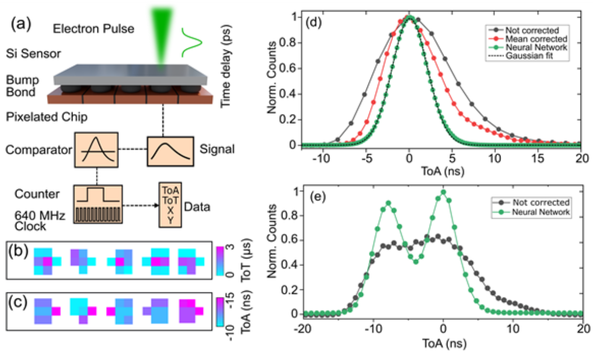
Graphic:



Keywords:

ultrafast-TEM, femtosecond-electron-pulses, laser-driven-cold-field-emitter, TimePix3, electron-electron-correlations

Reference:

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