Spatio-temporal resolution studies on a highly compact ultrafast electron diffractometer

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Abstract

Time-resolved diffraction with femtosecond electron pulses has become a promising technique to directly provide insights into photo induced primary dynamics at the atomic level in molecules and solids. Ultrashort pulse duration as well as extensive spatial coherence are desired, however, space charge effects complicate the bunching of multiple electrons in a single pulse. We experimentally investigate the interplay between spatial and temporal aspects of resolution limits in ultrafast electron diffraction (UED) on our highly compact transmission electron diffractometer. To that end, the initial source size and charge density of electron bunches are systematically manipulated and the resulting bunch properties at the sample position are fully characterized in terms of lateral coherence, temporal width and diffracted intensity. We obtain a so far not reported measured overall temporal resolution of 130 fs (full width at half maximum) corresponding to 60 fs (root mean square) and transversal coherence lengths up to 20 nm. Instrumental impacts on the effective signal yield in diffraction and electron pulse brightness are discussed as well. The performance of our compact UED setup at selected electron pulse conditions is finally demonstrated in a time-resolved study of lattice heating in multilayer graphene after optical excitation.

1. Introduction

The initial steps in chemical reactions or condensed matter transformations are fundamentally characterized by subnanometer atomic motions on a femto- to picosecond timescale [1, 2]. Time-resolved electron or x-ray diffraction techniques have become a promising tool with sufficient temporal precision to directly deliver insights into ultrafast phenomena at the molecular level [1–4]. Based on the pump–probe approach, ultrashort laser pulses start the reaction dynamics and either x-ray or electron probes are used to provide a time-delayed series of structural diffraction patterns in a stroboscopic manner. Major limitations of these methods have been the photon flux for x-ray sources and the temporal resolution of electron pulses as a result of space charge [1, 2]. Next generation source concepts, such as free-electron lasers and electron diffractometers with RF compression, promise to observe structural dynamics in a single-shot with pulse durations as short as a few fs [4].

Time-resolved diffraction with electron probes intrinsically differs from approaches with x-rays. Electrons possess a considerably higher scattering cross section, which in turn restricts their use to ultrathin samples or surfaces [1–5]. X-ray sources can provide pulses of extraordinary transverse coherence [6], but the shorter wavelength of high-energy electrons can effortlessly resolve interatomic distances [1–5]. However, a main advantage of pulsed electron diffractometers is their relatively small size in comparison to large-scale facilities, such as synchrotrons and free-electron lasers. Ultrafast electron diffraction (UED) table-top apparatuses have been, for example, applied to molecules in the gas phase [7–10], laser-induced melting [11–13], dynamics in graphite [14–19], charge density waves [20, 21], motions in organic salt [22], superstructure dynamics [23] and many others.
In UED experiments, ultrashort electron pulses are generally created by laser-induced emission. Mainly three different source concepts can be distinguished [5]: Photoelectric emission from planar metal surfaces [7, 15, 24–32], emission from sharp metal tips [33–37] and photo-ionization of ultracold gases [38–42]. Emitted electrons are accelerated either by electrostatic fields to lower energies of 0.2–1.5 keV [33–36] as well as high energies of 30–95 keV [15, 16, 24–32, 37, 42, 43] or with RF photo-injectors up to 3–5 MeV [44–47]. Regardless of the final electron energy, the temporal resolution of a UED setup basically depends on the electrons’ initial energy spread, the applied acceleration field strength, the propagation distance to the sample and in particular the charge density of the pulse [1–5, 25]. Coulomb repulsion between the electrons broadens the ultrashort bunches during their propagation from the point of generation to the sample. For bunches containing several electrons, space charge effects therefore limit the minimal electron pulse duration to ~200 fs (full width at half maximum (FWHM)) at the sample position [3, 48]. Without additional compression techniques [30–32, 43, 49, 50], a trade-off must be made between the required temporal resolution and the number of electrons per pulse. However, transient diffraction data with an adequate signal-to-noise ratio can only be obtained by detecting $10^6–10^9$ electrons at each pump–probe delay within a maintainable measurement time [2–5, 43]. Depending on the source concept, different approaches are thus pursued; ranging from the single-electron regime in combination with high repetition rates [5, 35–37, 51], over the application of multi-electron pulses at a lower number of pump–probe cycles [15, 16, 24–29, 42] to single-shot experiments with compressed dense electron bunches [22, 30, 32, 43–47, 50]. In this connection the irreversible or reversible nature of the investigated dynamics additionally determines the method of choice [2, 3, 5].

Apart from the temporal response of an UED experiment, a sufficient spatial resolving capacity of the diffraction patterns is required for achieving atomic resolution, especially of solid systems with large unit cells or complex biomolecules. The evaluation of the spatial resolution is directly connected to a detailed discussion of the electron bunch coherence properties [1–5, 25, 52]. In diffraction the transverse degree of coherence characterizes the interference capability of scattering centres that are far apart. Accordingly, the transverse coherence length of an electron bunch must overlap the entire system under investigation to achieve unrestricted structural information. The longitudinal coherence is of minor impact for uncompressed electron pulses [3, 5, 25, 32]. The transverse coherence length is normally expressed by the electron bunch lateral momentum spread in which global and local momentum distributions must be distinguished [52]. An optimization of the electron emission process can thus improve the coherence by using for example ultracold gases [38–42] or well-tuned laser parameter [51]. Furthermore, a reduction of the electron source size increases the lateral coherence as well, but space charge effects set limits [33–37, 53]. Transverse coherence lengths of 1–5 nm have been reported so far for multi-electron pulses from planar sources with pulse $1/e^2$-radii of 170–500 µm at the sample [21, 25, 31, 51, 54]. The application of single-electron pulses from a minimized source has recently extended the lateral coherence to 20 nm at a $1/e^2$-radius of 154 µm [53]. Moreover, the ongoing developments of tip-based [35, 37] and ultracold electron sources [40–42] promise further improvements of the spatial resolution in future challenging UED experiments.

In this contribution, we present extensive spatio-temporal resolution studies on a highly compact ultrafast electron diffractometer by means of experiments and simulations. The focus is here directed towards the robust generation and successful application of uncompressed multi-electron pulses with excellent spatio-temporal properties in the lower space charge regime. In section 2, the experimental details of our UED setup are described and the unique characteristics of the electron source design are pointed out. In section 3, we investigate the interplay between the spatial, temporal and signal resolution and whose impact due to space charge effects and instrumental imperfections. For this purpose, the transverse coherence length is firstly determined in dependence of the electron source size by different methods. Secondly, electron pulses of various charge densities from different source sizes are temporally characterized. Thirdly, the diffraction peak intensity and instrumental brightness are discussed on the basis of previously observed coherence lengths and pulse durations. Finally, the performance of our setup with optimized instrumental parameters is exemplified on the basis of an UED experiment on multilayer graphene. We conclude the contribution with a brief summary.

2. Experimental details

In this section, we describe the experimental details of our UED setup and point out the unique characteristics of the electron pulse source concept, whose performance has been analysed in the spatio-temporal resolution studies presented here. Furthermore, a brief introduction is given to the preparation technique of free-standing single crystalline graphite and graphene samples, which were used for the spatial resolution measurements and time-resolved pump–probe experiments in this work.
2.1. Electron diffraction setup

Experiments in this work were performed with the UED setup illustrated in Figure 1(a). The apparatus is powered by a Ti:sapphire multipass amplifier delivering (27 ± 3) fs (FWHM) laser pulses at a central wavelength of 795 nm and ∼800 μJ maximum energy per pulse at a repetition rate of 1 kHz. Beam pointing and output power are independently stabilized by a closed-loop control system. The laser pulses are first separated into a pump and probe arm (S1). The reflected probe pulses are frequency-tripled (THG), a half wave plate (HWP) enables attenuation and dispersion is compensated by a prism compressor (PC). Free electron bunches are generated within the vacuum chamber (VC) by tightly focussing (L1) the 3ω-pulses into the electron pulse source (EPS). A high voltage connection (HVC) supplies the dc source to accelerate the electron pulses (EP) to 40 keV. Dispersed electrons are mapped onto a multiplying detector (MD) by a cooled magnetic lens (CML) and finally recorded with a CCD. For the pump beam the fundamental ω-pulses are attenuated with a neutral density filter (ND), focussed (L2) onto the sample and the relative arrival time is controlled by an optical delay stage (DS1). For a temporal electron pulse characterization via grating enhanced ponderomotive scattering (dotted beam path) the pump pulses are further split (S2) and separately focussed (L3, L4) from opposite sides on the electron bunches. In this geometry the position of the standing light wave can be adjusted by a second delay stage (DS2). (b) Enlarged view of the sample region. The frequency tripled probe pulses (3ω) back-illuminate a gold coated photocathode (C). The emitted electron pulses (EP) first gain momentum in the acceleration region (AR), passing a pinhole (P) in the anode (A) and further propagate through a field free drift region (DR). Therein a sample holder (SH) controls the position of the sample (S) and diffracted electrons are finally mapped on the detector by a cooled magnetic lens (CML) with an iron pole piece gap (G).

Figure 1. Schematic representation of the UED setup and detailed scale drawing of the electron pulse source. (a) Pulses from an amplifier system (1 kHz, 27 fs, 795 nm) are first separated into a pump and probe arm (S1). The reflected probe pulses are frequency-tripled (THG), a half wave plate (HWP) enables attenuation and dispersion is compensated by a prism compressor (PC). Free electron bunches are generated within the vacuum chamber (VC) by tightly focussing (L1) the 3ω-pulses into the electron pulse source (EPS). A high voltage connection (HVC) supplies the dc source to accelerate the electron pulses (EP) to 40 keV. Dispersed electrons are mapped onto a multiplying detector (MD) by a cooled magnetic lens (CML) and finally recorded with a CCD. For the pump beam the fundamental ω-pulses are attenuated with a neutral density filter (ND), focussed (L2) onto the sample and the relative arrival time is controlled by an optical delay stage (DS1). For a temporal electron pulse characterization via grating enhanced ponderomotive scattering (dotted beam path) the pump pulses are further split (S2) and separately focussed (L3, L4) from opposite sides on the electron bunches. In this geometry the position of the standing light wave can be adjusted by a second delay stage (DS2). (b) Enlarged view of the sample region. The frequency tripled probe pulses (3ω) back-illuminate a gold coated photocathode (C). The emitted electron pulses (EP) first gain momentum in the acceleration region (AR), passing a pinhole (P) in the anode (A) and further propagate through a field free drift region (DR). Therein a sample holder (SH) controls the position of the sample (S) and diffracted electrons are finally mapped on the detector by a cooled magnetic lens (CML) with an iron pole piece gap (G).
188 mm away from the electron source. A charge-coupled device (CCD) records the resulting pattern with one megapixel resolution. The number of electrons per pulse can be measured by an aluminium faraday cup and a pico-amperemeter.

Performing time-resolved pump–probe experiments the sample is excited by the fundamental of the amplifier system at 795 nm. Dispersion management after the amplifier for UV pulse generation and for propagation to the sample is adjusted such, that a pulse duration of (27 ± 3) fs (FWHM) is achieved in front of the sample. A long focal lens \( f = +500 \text{ mm} \) focuses the p-polarized laser pulses with 12° angle of incidence onto the sample. The beam is slightly over-focussed to avoid effects due to beam pointing and inhomogeneous excitation of ∼100 μm large samples (see figure 2). The spot size at the sample position was determined by a CCD beam profiler giving a 1/\( e^2 \)-diameter of ∼480 μm (see section 3.4 and figure 13(d) for details). The incident fluence is continuously variable via a reflecting neutral density filter and a maximum peak fluence of ∼60 mJ cm\(^{-2}\) can be used at the sample position in pump–probe experiments. A high precision optical delay stage controls the relative arrival time between the electron probe and optical pump pulse. By recording diffraction pattern snapshots at different time delays, laser induced structural dynamics in the sample can be observed directly.

The overall time resolution under normal incidence of electrons to the sample is mainly limited due to the electron pulse duration, which is further influenced by internal space charge effects and the initial energy spread. For a temporal pulse characterization, the electron bunches are directly cross-correlated with two counter-propagating laser pulses. This method uses the ponderomotive force of the pulsed laser field acting on the electrons in a standing-wave enhanced geometry [30, 31, 48]. As illustrated in figure 1(a) (dotted beam line), the fundamental pump pulses are splitted once again and are separately focussed \( f = +300 \text{ mm} \) with a 1/\( e^2 \)-radius of 33 μm and energy of ∼200 μJ per pulse at the electron bunches in a counter-propagating way. The position of the resulting stationary light wave is adjusted with the help of a second delay stage. By finally measuring the ponderomotively scattered electrons at different delay times on the detector, the local electron pulse charge density at a definite point in time and space can be reconstructed (see section 3.2 for details).

2.2. Single crystalline graphite and graphene sample preparation

Free-standing single crystalline graphite and graphene samples have been prepared in a three-step procedure (see [56] for details). First, macroscopic single crystal flakes of natural graphite are cleaved by multiple mechanical exfoliation with semiconductor grade adhesive tape [57]. The thinned graphite is afterwards deposited on top of an oxidized silicon wafer and its surface is carefully scanned in an optical microscope (see figure 2(a)). In the second step, residual flakes of adequate size and thickness are characterized by optical reflection [58] as well as atomic force microscopy, giving the possibility to directly count the number of graphene layers. In the final step, a selected flake is transferred on top of a Quantifoil 200 mesh TEM gold grid (90 μm hole size, see figure 2(b)). For an improved transfer process, the grid is partially covered by a perforated carbon film [56]. This amorphous support film is selectively removed from the sample grid hole before preparation by fs laser machining to reduce the electron diffraction background signal and avoid perturbing effects in pump–probe experiments. All experimental results shown in sections 3.1, 3.3 and 3.4 have been achieved using the 3 nm thick, nine layer graphene sample shown in figure 2.
3. Results and discussion

In experiments and simulations, we have investigated the transverse coherence length, electron pulse duration, elastic scattered peak intensity and instrumental peak brightness in dependence of the electron pulse charge density. For this purpose three spot sizes of the UV probe beam on the photocathode were chosen. The lateral profiles of the UV beam were measured with a scanning knife-edge method at three defined longitudinal beam positions: directly at the beam waist, 0.5 mm behind the waist and 3 mm in front. Provided that the beam profile had an overall Gaussian shape, the corresponding 1/e²-radii were: \((2.3 \pm 0.5) \mu m\), \((9.5 \pm 1.3) \mu m\) and \((28.0 \pm 1.2) \mu m\) respectively. A pointing jitter of the laser focus was not observed, but would be included in the measured results that were averaged over many shots. The electron emission is a single-photon process and thus permits to transfer the measured optical quantities linearly onto the electron source. Consequently, a simple adjustment of the longitudinal position of the UV pulse focussing lens (see figure 1(a)) allows a variation of the electron source area over more than two orders of magnitude. For all source sizes the emitted charge from the photocathode were measured to be linear in dependence of the UV laser power without saturation effects in the studied parameter range. Hence, the electron pulse emission is below the space charge limited regime and the virtual cathode effect has not been taking into account \([59, 60]\). For clarity, all source size dependent results presented in the following figures are colour-coded as follows: 2.3 \(\mu m\) source in red, 9.5 \(\mu m\) source in green and 28.0 \(\mu m\) source in blue.

3.1. Transverse coherence length

We have characterized the transverse coherence length of our UED setup for the mentioned three electron source sizes with particle tracking simulations and three experimental approaches. The N-body simulation was self-developed to get direct access to all tuneable parameters and their effects on the spatio-temporal resolution of the UED experiment (for simulation details see appendix B). Figure 3 displays the evolution of the electron bunch diameter from the generation at the cathode to the sample position for each of the three source sizes. The lateral expansion of the electron pulses during propagation, the global degree of coherence is enlarged similarly. In other words, the ratio of bunch radius to coherence length is conserved and the electron pulse transverse coherence can be approximated with knowledge of the initial coherence properties at the cathode \([53]\). From an analytical expression of the thermal emittance for metal cathodes \([61–63]\), we expect an initial transverse coherence length of \(\xi_i = (0.67 \pm 0.16) \text{ nm}\) for our electron pulse source, which is consistent with comparable
values reported for gold cathodes [51, 53, 64] (see appendix B.1 for details). Thereby, the transverse coherence length at the sample \( \xi_s \) can be valued from

\[ \xi_s \leq \frac{w_s}{w_i} \]

(1)

with the electron source \( 1/e^2 \)-radius \( w_i \) and electron pulse \( 1/e^2 \)-radius \( w_s \) at the sample [53]. Equality in equation (1) holds in the absence of space charge effects. Accordingly, we can get an upper limit of the electron pulses transverse coherence lengths at the sample position by measuring the ratio \( w_s/w_i \) for all three different electron source sizes. The source radii \( w_i \) are mentioned above and the corresponding radii at the sample \( w_s \) were determined from electron shadowgraphs as shown in the upper image row of figure 4. Therefore, the electron pulses were imaged onto the detector with the magnetic lens switched off. Using the shadow of the TEM sample grid (shown in figure 2) as a length gauge, the radii at the sample \( w_s \) were calculated by the second central moment of the radial intensity profiles on the detector, resulting in \( 1/e^2 \)-radii of \( w_{s,2.3} = (84.7 \pm 12.3) \mu m \), \( w_{s,9.5} = (64.4 \pm 10.9) \mu m \) and \( w_{s,28.0} = (57.4 \pm 10.4) \mu m \). From equation (1) we finally estimated transverse coherence lengths at the sample position of \( \xi_{s,2.3} < (24.7 \pm 14.8) \), \( \xi_{s,9.5} < (4.5 \pm 2.5) \) and \( \xi_{s,28.0} < (1.2 \pm 0.6) \) nm.

Furthermore, the N-body simulations have been utilized to confirm the predicted transverse coherences for all three source sizes. Results of the simulated electron pulse intensity profiles are presented in the lower image row of figure 4 and show a good agreement with the experimental outcomes. It should be noted, that no free parameters were used in the simulations (see appendix B for details). The particle tracking algorithm makes the 6D phase-space distribution of an electron pulse accessible during propagation and allows to calculate the transverse normalized root mean square (RMS) emittance

\[ \epsilon_{n,x} \equiv \frac{1}{m_0 c} \sqrt{\left\langle x^2 \right\rangle \left\langle p_x^2 \right\rangle - \left\langle x \right\rangle \left\langle p_x \right\rangle^2} \]

(2)

with the particle coordinate \( x \), relativistic momentum \( p_x \), electron rest mass \( m_0 \) and speed of light \( c \). The brackets \( \langle \rangle \) stand for averaging over the entire particle ensemble of the bunch and the transverse emittance \( \epsilon_{n,y} \) in \( y \)-direction is given analogously [54, 65, 66]. Due to the cylindrical symmetry of our electron bunches we maintain \( \epsilon_{n,x} = \epsilon_{n,y} = \epsilon \) from the source to the sample. From simulations of all three source sizes we obtained mean transverse emittance values at the sample position of \( \epsilon_{s,2.3} = (0.8 \pm 0.1) \) nm, \( \epsilon_{s,9.5} = (2.6 \pm 0.4) \) nm and \( \epsilon_{s,28.0} = (7.5 \pm 0.3) \) nm for the three source radii of 2.3 \( \mu m \), 9.5 \( \mu m \) and 28.0 \( \mu m \), respectively. Moreover, the
transverse coherence length $\xi$, can be estimated from the transverse normalized RMS emittance $\varepsilon_s$ by

$$\xi \approx \frac{\hbar}{m_e c} \frac{w_s}{2\varepsilon_s},$$

where $\hbar$ is the reduced Planck constant [54, 65] (see appendix A for details). With equations (2) and (3), the particle tracking simulations for all three source sizes result in mean transverse coherence lengths at the sample position of $\xi_{2.3} \approx (19.2 \pm 2.6) \text{ nm}$, $\xi_{9.5} \approx (3.8 \pm 0.6) \text{ nm}$ and $\xi_{28.0} \approx (1.45 \pm 0.04) \text{ nm}$. Consequently, the simulated coherence properties are well consistent with the coherence values determined by the source expansion ratio giving an upper limit.

A direct experimental access to the transverse emittance and hence to the transverse coherence is provided by pepper-pot or multislit techniques [68]. In accordance with [67], we analysed TEM grid shadowgraphs as shown in figure 4 by fitting error functions to the intensity profile of a grid edge. Taking the detector point spread function (PSF) (see figure 8 and corresponding text) into account through deconvolution [67], we obtained mean transverse coherence lengths at the sample position of $\xi_{2.3} \approx (19.2 \pm 2.6) \text{ nm}$, $\xi_{9.5} \approx (3.8 \pm 0.6) \text{ nm}$ and $\xi_{28.0} \approx (1.45 \pm 0.04) \text{ nm}$. Consequently, the simulated coherence properties are well consistent with the coherence values determined by the source expansion ratio giving an upper limit.

However, the transverse coherence length of electron pulses not only limits the maximum resolvable distance in a sample, it further quantifies the resolution of the electron diffraction pattern in reciprocal space. More precisely, an increasing transverse coherence $\xi$ narrows the width of Bragg peaks in a pattern proportionally by

$$\xi = \frac{1}{\sigma_k},$$

where $\sigma_k$ is the standard deviation for Gaussian distributed Bragg peaks (see appendix A for details). Thus, we measured diffraction data for the three selected electron source sizes and extracted the particular transverse coherence from the respective peak width. Figure 5 presents the recorded diffraction signals, which were acquired from the multilayer graphene sample shown in figure 2 by integrating over $\sim 10^4$ incident electrons from $\sim 10^5$ laser shots in 11 s. Central undiffracted electrons were blocked by a beam stop and all patterns are shown before (right) and after background subtraction (left). In each case, the background-free elastic scattered signal was normalized to the sample electron flux and total elastically scattered intensity (see section 3.3 for details). Three source size dependent effects are readily identifiable: Bragg peak sharpness, peak intensity and peak-to-background intensity ratio increases with decreasing source size. 210 Bragg peaks, accentuated by black squares, were selected for further coherence analyses and are shown in detail in figure 6.
The Bragg peak width increases as expected and the peak intensity thus decreases (see also figure 6). Although the diffuse background is stronger for the smallest source due to the fact that more electrons transmitted the sample surrounding amorphous carbon film (see figures 2 and 4), the elastically scattered signal can be separated better from the background and the signal-to-noise ratio consequently increases with decreasing source size.

To further extract the transverse coherence for all three source sizes from the Bragg peak width, we fitted in each case a circular 2D Gaussian function to a series of peaks in a pattern. Figure 6 exemplifies the fitting procedure for the 210 Bragg peaks accentuated by black frames in the patterns of figure 5. All intensity profiles are well approximated by the circular 2D Gaussian distribution. Minor deviations only exist between 5% and 35% of the maximum peak intensity (see dashed lines in the peak profiles). This spreading of the measured peak tails arises from space charge effects and a nonlinear signal response, which is attributed to artefacts in the detector imaging system [69–71]. Data points affected from this broadening were therefore disregarded in the fitting procedure. Accordingly, we analysed the 24 most intense Bragg peaks in each pattern, resulting in an averaged peak standard deviation of \( \sigma_{k,I,2.3} = (0.727 \pm 0.009) \text{ nm}^{-1} \), \( \sigma_{k,I,9.5} = (0.924 \pm 0.008) \text{ nm}^{-1} \) and \( \sigma_{k,I,28.0} = (1.344 \pm 0.016) \text{ nm}^{-1} \). The corresponding instrumental transverse coherence lengths \( \xi_{2.3} = (1.38 \pm 0.01) \text{ nm} \), \( \xi_{9.5} = (1.08 \pm 0.01) \text{ nm} \) and \( \xi_{28.0} = (0.74 \pm 0.01) \text{ nm} \) were finally calculated from equation (4). It is obvious that these results differ from already ascertained coherence values. Most notably the expected scaling of the Bragg peak width with the source size was not reproduced. Consequently, the resolution of the electron diffraction patterns must be essentially affected by instrumental imperfections [71, 72]. We have taken in account two well-known basic causes: aberrations of the magnetic electron lens [71, 72] and signal spreading in the detector system [71, 72, 73, 74]. For each effect, a 2D PSF was determined in reciprocal space to further deconvolute the measured Bragg peak width [72]. Afterwards, the source-limited transverse coherence length \( \xi_s \) at the sample can be extracted from the mean instrumental Bragg peak width \( \sigma_{k,I} \) by

\[
\xi_s = \frac{1}{\sqrt{\sigma_{k,I}^2 - \sigma_{k,L}^2 - \sigma_{k,D}^2}},
\]

with the magnetic lens PSF width \( \sigma_{k,L} \) and detector PSF width \( \sigma_{k,D} \) (standard deviations).

The magnetic lens PSF represents a focused image of an electron point source onto the detector under aberrations and space charge effects. Especially within the space charge regime, an ideal electron point source is not experimentally realizable. Therefore, we determined the magnetic lens PSF by simulating the magnetic field

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**Figure 6.** Bragg peak fitting procedure for the coherence analysis of electron diffraction pattern from three different source sizes (1/e²-radii: 2.3 μm red scale, 9.5 μm green scale and 28.0 μm blue scale). The first column exemplarily shows the measured 210 Bragg peaks accentuated by black squares in the patterns of figure 5. The second column presents the corresponding fit results in the form of circular 2D Gaussian distributions. The third column compares line profiles of both peaks (along dashed lines in the images). Deviations from the Gaussian profile between 5% and 35% of the maximum peak intensity (between dashed lines in the peak profiles) are attributed to a nonlinear detector response and space charge effects. Affected data points (open circles) were thus excluded from the fitting procedure.
distribution of the solenoid with the freely available FEMM finite element program [75] and performed particle tracking simulations with implemented magnetic field. Figure 7(a) exemplifies projected particle trajectories of an electron pulse generated from the 2.3 μm source with (solid lines) and without (dashed lines) acting magnetic field in combination with the magnetic flux density distribution. The particle tracking algorithm additionally includes diffraction from 210 and 210 lattice planes in the multilayer graphene sample (see also figure 13(b)), the negative lens effect of the anode pinhole and space charge forces. The strength of lens aberrations and thus the width of the magnetic lens PSF basically depends on the abaxial electron pulse properties inside the solenoid field. Consequently, we separately determined a magnetic lens PSF for each of three studied source sizes due to their different electron pulse lateral diameters and divergence angles (as shown in figure 3). Additionally, the strong abaxial particle trajectories of diffracted electrons were taken into account by implementing the scattering process at the sample representatively in 210 direction. In order to finally obtain the source size dependent magnetic lens PSFs, the 210 Bragg peak intensity distribution on the detector was separately simulated based on a focussed electron point source with the abaxial properties of the particular finite source (see appendix B.2 for details). Figure 7(b) exemplarily presents the resulting magnetic lens PSF for the 28.0 μm electron source. Standard deviations $\sigma_{NL}$ for all three source sizes were determined by fitting each simulated lens PSF with a circular 2D Gaussian distribution. The results show a 10% increase of the PSF width from the largest to smallest source. (c) 2D Gaussian magnetic lens PSF with source-averaged standard deviation following from averaging over the fitted PSF widths of all three source sizes. (d) Comparison between source-averaged magnetic lens PSF (L-PSF) and mean fitted peak profiles (as shown in figure 6) illustrates that the Bragg peaks must be affected by lens aberrations.

We analysed the lateral signal spreading of the detector system according to [73, 74]. The detector PSF was estimated to be a 2D Gaussian distribution and its width was determined by fitting circular 2D Gaussians to single-electron detector events. For this purpose, the electron beam was mapped on the detector with removed sample grid and inactive magnetic lens. Images were recorded under same conditions as diffraction patterns in figure 5. Statistically occurring detector events around the central electron spot have to be taken into account to reconstruct the source-limited transverse coherence length from the diffraction pattern resolution.

Figure 7. Aberration analysis and simulation results of the magnetic lens PSF. (a) FEMM-simulated magnetic flux density distribution of the magnetic lens in false colours as well as selected particle trajectories of a focussed (solid lines) and unfocussed (dashed lines) electron pulse under the influence of the electrostatic lens effect inside the anode (A), diffraction by the sample (S) and space charge forces (see appendix B.2 for details). (b) Simulated magnetic lens PSF for the 28.0 μm source size plotted on same scale like Bragg peaks in figure 6, but with doubled image resolution. Individual PSFs for each of the three studied source sizes were fitted by circular 2D Gaussian distributions, resulting in a 10% increase of the PSF width from the largest to smallest source. (c) 2D Gaussian magnetic lens PSF with source-averaged standard deviation following from averaging over the fitted PSF widths of all three source sizes. (d) Comparison between source-averaged magnetic lens PSF (L-PSF) and mean fitted peak profiles (as shown in figure 6) illustrates that the Bragg peaks must be affected by lens aberrations.
the dashed line) displays averages of the fitted PSFs for 10 different peak intensities, which agree well with the corresponding measured single events. The variation in the signal gain is well known for micro-channel plate detectors and characterized by the width of the pulse height distribution [76]. A first comparison between the detector PSF in figure 8(b) and the magnetic lens PSF in figure 7(c) (both plotted on same scale) already demonstrates, that the major impact on the Bragg peak broadening is attributed to the signal spreading in the detector system. The line profiles in figure 8(c) provide a direct assessment of the detector PSF, magnetic lens PSF and mean diffraction peak widths. The detector PSF width represents 76% of the Bragg peak width in case of the smallest source size in contrast to 54% for the lens PSF. Deconvolution reveals that aberrations of the magnetic lens and the signal spreading in the detector causes a sevenfold broadening of the Bragg peaks for the 2.3 μm source. Finally, we calculated for all three source sizes the source-limited transverse coherence lengths $\xi_s$,

\[
\begin{align*}
\xi_{s,2.3} & = (10.0 \pm 14.4) \text{ nm}, \\
\xi_{s,9.5} & = (1.6 \pm 0.1) \text{ nm} \quad \text{and} \\
\xi_{s,28.0} & = (0.85 \pm 0.01) \text{ nm} \quad \text{from equation (5).}
\end{align*}
\]

Although instrumental imperfections have been taken into account, those outcomes are consistently smaller than values obtained by the other methods. We attribute the deviations to additional broadening effects like fluctuations in the magnetic lens current, a lack of sample flatness, crystal strains and inhomogeneities [71, 72]. Thus, the full information content of complex electron diffraction patterns could only be partially retrieved with the quantitative knowledge of detector and magnetic lens PSFs [72].

Table 1 summarizes the four different approaches and their results to the transverse coherence length for three altered electron pulse source sizes.

| Source size (μm) | Via source expansion (upper limit) | Via simulated emittance | Via TEM grid method (lower limit) | Via deconvoluted Bragg peak width |
|-----------------|------------------------------------|------------------------|----------------------------------|----------------------------------|
| 2.3 ± 0.5       | 24.7 ± 14.8                        | 19.2 ± 2.6             | 14.6 ± 6.2                      | 10.0 ± 14.4                      |
| 9.5 ± 1.3       | 4.5 ± 2.5                          | 3.8 ± 0.6              | 2.9 ± 0.2                       | 1.6 ± 0.1                        |
| 28.0 ± 1.2      | 1.2 ± 0.6                          | 1.45 ± 0.04            | 1.3 ± 0.1                       | 0.85 ± 0.01                      |

The Bragg peaks are predominantly broadened by signal spreading in the detector system and the magnetic lens PSF has less impact.
3.2. Temporal electron pulse characterization

Different approaches are used for measuring the temporal profile of ultrashort electron bunches in table-top experiments [29, 48, 77, 78]. In our UED setup we have characterized electron pulse durations by grating enhanced ponderomotive scattering according to [30, 31, 48, 79]. This cross-correlation method uses the ponderomotive interaction of the electron bunches with two counter-propagating laser pulses in a standing-wave enhanced geometry (see figure 9(b)). By measuring the ponderomotively scattered electron signal at different delay times on the detector, the FWHM of the electron pulse duration \( \tau_e \) can be reconstructed from FWHM \( \tau_s \) of the signal time trace (see figure 9(d)) by

\[
\tau_e = \sqrt{\tau_s^2 - \tau_l^2 - \tau_f^2}
\]

with the laser pulse duration \( \tau_l \) and the FWHM focal transit time \( \tau_f \) of the electron pulse through the joint laser focus. For a comprehensive description of the grating enhanced ponderomotive scattering technique see [48]. We have experimentally realized the characterization method as illustrated in figure 1(a) (dotted beam line) and detailed in figures 9(a) and (b). The fundamental pump pulses were split to \( \sim 200 \mu J \) per pulse and separately focussed (\( f = +300 \) mm) to a spot size of \( \Omega = (39 \pm 2) \mu m \) (FWHM) onto the electron bunches. The focal intersection was placed as close as possible to the sample position (1.5 mm in front) to achieve a meaningful estimation of the electron pulse duration in pump–probe experiments. A correct retrieval of the electron pulse duration was just acquirable by a background-free detection of scattering signal. For this purpose, electron pulses leaving the anode were centrally sliced by a 13 \( \mu m \) narrow copper slit before the residual centre part was scattered by the ponderomotive force of the standing-wave laser field (see figure 9(b)). Figure 9(c) shows logarithmically scaled detector images of the sliced electron pulses at various temporal laser–electron pulse overlap. Important geometrical facts are represented by dashed lines for clarity. (d) Gaussian fitted time traces from the 9.5 \( \mu m \) source at various charge densities with vertical offset for clarity. The electron pulse duration correlated trace width increases with the number of electrons as expected (dashed lines at FWHM are given as guides to the eye).

Figure 9. Temporal electron pulse characterization via grating enhanced ponderomotive scattering [48]. (a) Detailed scale drawing of the characterization geometry (overview shown in figure 1(a)). Electron pulses (EP) leaving the anode pinhole (P) are partially blocked by a copper slit (CS) in the sample holder (SH). The residual centrepiece is scattered by the ponderomotive force of counter-propagating laser pulses (\( \omega \)) 1.5 mm in front of the sample position. (b) Schematic illustration of characterization geometry. The copper slit transmits in x-direction only a 13 \( \mu m \) flat bunch slice to allow a background-free detection of electrons scattered inside the intensity grating, which has dimensions of 8 \( \mu m \) (from pulse duration) and 39 \( \mu m \) (from spot size) FWHM in x- and z-direction respectively. (c) Logarithmically scaled raw detector images of the sliced electron pulses in the x-y-plane, with (lower panel) and without (upper panel) laser-electron pulse overlap. Important geometrical facts are represented by dashed lines for clarity. (d) Gaussian fitted time traces from the 9.5 \( \mu m \) source at various charge densities with vertical offset for clarity. The electron pulse duration correlated trace width increases with the number of electrons as expected (dashed lines at FWHM are given as guides to the eye).
space charge forces. By fitting Gaussian functions to the time traces, we obtained in each case an electron pulse duration $\tau_e$ from the FWHM $\tau$ of the trace fit by means of equation (6) with the focal transit time $\tau_f = (346 \pm 21)$ fs (FWHM) and laser pulse duration $\tau_l = (27 \pm 3)$ fs. The transit time $\tau_f = \Omega/v_z$ is given by the focal spot size $\Omega$ and the mean electron longitudinal velocity $v_z = 112$ nm fs$^{-1}$. For instance, the evaluation of the lowermost and topmost signal time trace in figure 9(d) resulted in FWHM electron pulse durations of $(120 \pm 62)$ fs and $(410 \pm 18)$ fs for $49 \pm 79$ and $5040 \pm 102$ electrons per pulse, respectively. Convoluted with the pump pulse duration of $12$ fs (RMS) and the $22$ fs (RMS) effect of the non-collinear sample excitation, these values correspond to RMS temporal resolutions of $(57 \pm 28)$ fs and $(176 \pm 9)$ fs.

For a detailed quantification of the temporal pulse broadening with decreasing electron source size, we have investigated the electron pulse duration for the three studied sources as a function of the number of particles per pulse. Results of the ponderomotive scattering experiment in combination with simulated electron pulse durations are presented in figure 10. Calculated (transparent point clouds) and measured (circles) data are in remarkable good agreement. The simulated pulse durations were computed by the particle tracking algorithm used in section 3.1. The good agreement with the measured results was only achievable by a rigorous implementation of all experimental parameter including the copper slit as well. Mainly two effects can lead to deviations between the measured pulse duration of the sliced electron pulses $1.5$ mm in front of the sample position and the actual pulse duration of the unsliced electron pulses used for UED experiments. First, space charge effects can broaden the temporal pulse profile under further propagation to the sample and the measured electron pulse duration would be underestimated. However, the temporal characterization was only performed at the central part of the nearly Gaussian electron pulse profile which is more affected by space charge broadening than the electron pulse edges. This fact causes an overestimation of the pulse duration. In consideration of both opposite aspects, our simulations could finally reveal that the slicing effect is more significant and the unsliced pulse duration at the sample can be up to 6% shorter in comparison to the measured pulse duration. It should be noted, that no free parameters were used in all simulations of electron pulse durations (see appendix B for details).

The results of the temporal pulse characterization in figure 10 show that for all source sizes a minimal electron pulse duration of $\sim 120$ fs can be reached with less than 50 electrons per pulse close to the single-electron regime. This value is supported by theoretical considerations from [25, 51], providing a minimal pulse duration of $\tau_{e,\text{min}} = (120 \pm 38)$ fs for our instrumental parameters.

![Figure 10. Measured and simulated electron pulse durations at 1.5 mm in front of the sample position as a function of the number of electrons per pulse for each of the three studied source sizes (1/e²-radii: 2.3 μm red, 9.5 μm green and 28.0 μm blue). Simulated (transparent point clouds) and measured (circles) data show a remarkable good agreement at all source sizes. The theoretical minimal pulse duration of $\tau_{e,\text{min}} = (120 \pm 38)$ fs is indicated by the solid line (error ranges by dashed lines).](image)
Above ~50 electrons per pulse in the presence of space charge effects, the strength of the temporal pulse broadening strongly depends on the source size. Electron pulses with (2814 ± 14) carriers (more than sufficient to study reversible dynamics) still remain ultrashort with a FWHM pulse duration of (175 ± 44) fs in case of the largest 28.0 μm source, whereas they are strongly broadened to (705 ± 17) fs for the smallest 2.3 μm source with (2656 ± 338) electrons per pulse. The fourfold growth in pulse duration is put into perspective by taking into account that the initial charge density increases by more than two orders of magnitude. Furthermore, the drastically enhanced transverse coherence of electron pulses from the smallest source implicates increasing diffraction peak intensities (see figure 5 and section 3.3 for details) and allows a reduction of the number of electrons per pulse. For instance, diffraction patterns with excellent signal-to-noise ratio as presented in figure 13(b) are achievable within a minute exposure time using either (839 ± 124) electrons per pulse from the 2.3 μm source or (1500 ± 85) electrons from 9.5 μm source. Thus, FWHM pulse durations of (337 ± 17) fs or (230 ± 32) fs in combination with transverse coherence lengths up to 20 or 4 nm, respectively become applicable for high-precision UED experiments. Our comprehensive spatio-temporal resolution studies consequently reveal that the compactness of our electron source design provides at balanced conditions a temporal resolution of less than 200 fs along with high-definition electron diffraction.

3.3. Coherence-dependent diffraction peak intensity and instrumental brightness

As already indicated, the transverse coherence length of electron pulses not only defines the Bragg peak width in diffraction patterns (see section 3.1 for details), it consequentially affects the Bragg peak intensity at a steady electron flux through the sample. More precisely, the peak intensity of a Bragg spot increases quadratically with its underlying transverse coherence [52, 71]. To investigate this correlation, we analysed the diffraction peak intensity in dependence of the transverse coherence length for each of the three studied source sizes.

For this purpose, raw pattern images had to be normalized after background subtraction to obtain the quantitative diffraction data shown in figure 5. As each individual diffracted electron in a bunch only interferes with itself [5, 51, 71] and its scattering probability is mainly determined by sample intrinsic parameters [25, 71], we first normalized each background-free pattern \(I_{bf}(k_x, k_y)\) by its overall elastic scattered intensity

\[
I_n(k_x, k_y) = \frac{I_{bf}(k_x, k_y)}{\iint Id(k_x, k_y) \, dk_x \, dk_y},
\]

to eliminate disturbing influences due to laser and electron source fluctuations during the recording process. Afterwards, each standardized diffraction pattern \(I_n(k_x, k_y)\) was normalized by its particular incident electron flux at the sample as follows: We determined the source-size dependent flux by analysing the sample shadowgraphs presented in the first image row of figure 4. For each image, pixel values below 50% of the maximum transmitted intensity were set to zero and the remaining intensity was integrated. The 50% threshold facilitates for all three images a strict distinction between directly transmitted and scattered electrons (not visible in figure 4) by means of sharp boundary conditions (indicated by dashed lines in figure 4). The outcomes were normalized relative to the largest 28.0 μm source, resulting in a relative electron flux of 61% and 82% for the 2.3 μm and 9.5 μm source, respectively. The standardized diffraction pattern \(I_n(k_x, k_y)\) were finally divided by the relative electron flux values to obtain a consistent intensity scale for the diffraction data of all three studied source sizes presented in figure 5.

To reveal a potential quadratic dependence of the Bragg spot peak intensity on the transverse coherence length, we fitted the 110 and similar five permutation symmetric [110] Bragg spots (equal distances to the centre) in all three quantitative diffraction patterns by 2D Gaussian distributions (see section 3.1 and figure 6 for fit details). Averaging over the six fits from each of the three patterns provided the Gaussian peak profiles presented in figure 11(a). The direct comparison clearly shows a rise of the peak intensity with increasing transverse coherence. The 2.3 μm and 9.5 μm source achieves a 3.9 or 2.2 times higher peak intensity relative to the 28.0 μm source. However, a quadratic intensity dependence should result in a relative peak intensity enhancement by a factor of 138.4 or 3.5 assuming the deconvoluted transverse coherence lengths of 10.0 nm and 1.6 nm, respectively (see table 1). In order to check, we deconvoluted the 2D Gaussian distribution corresponding to the peak profiles in figure 11(a) with the magnetic lens PSF and the detector PSF (compare section 3.1). The 2D deconvolution was performed under conservation of the total peak volume. Figure 11(b) illustrates resultant profiles of the source-limited Bragg peaks on a single logarithmic scale. Their intensity and width drastically change for higher coherent electron pulses in absence of instrumental imperfections. The deconvoluted peak intensities of the 2.3 μm and 9.5 μm source are 144.6 and 3.7 times higher with respect to the 28.0 μm source and in good agreement with the expected ones (see above).

Furthermore, we analysed the peak intensity ratio in case of a prospective minimization of magnetic lens aberrations and signal broadening effects in the detector system. For this purpose, the source-limited Bragg peaks were reconvinulated by a magnetic lens PSF as well as a detector PSF, which both have half the widths of the
brightness depeak intensities due to instrumental imperfections, the magnetic lens and detector PSFs are included in the brightness, which is the most often used longitudinal energy spread is of minor importance [66]. To account for the above-mentioned decrease of Bragg conditions.

The optimized performance of time-resolved diffraction experiments, we parameters can be reliably retrieved by theoretical considerations and numerical simulations. With respect to an source relative to the 28.0 μm.

so far our comprehensive resolution studies have shown that all spatial and temporal electron pulse parameters can be reliably retrieved by theoretical considerations and numerical simulations. With respect to an optimized performance of time-resolved diffraction experiments, we finally introduce the transverse beam brightness, which is the most often used figure of merit for the electron pulse quality in applications where the longitudinal energy spread is of minor importance [66]. To account for the above-mentioned decrease of Bragg peak intensities due to instrumental imperfections, the magnetic lens and detector PSFs are included in the brightness definition. Accordingly, we have calculated the instrumental transverse normalized peak brightness $B_1$ in our cylindrically symmetric setup as follows:

$$B_1 \approx \left( \frac{m_0 c}{\pi \hbar} \right)^2 \sqrt{\frac{8 \ln 2}{2\pi}} \frac{N e \xi_1^2}{\tau_e w_2^2},$$

where $\xi_1$ is the instrumental transverse coherence length affected by instrumental imperfections, $w_2$ is the lateral electron pulse 1/e2-radius at the sample, $\tau_e$ is the FWHM electron pulse duration, $N$ is the number of electrons per pulse at the sample, $\hbar$ is the reduced Planck constant, $e$ is the elementary charge, $m_0$ is the electron rest mass and $c$ is the speed of light (see appendix B.3 for derivation details). The instrumental transverse brightness $B_1$ represents a figure of merit for the acquirable diffraction signal yield in space at a given resolution in time. We therefore simulated the instrumental brightness as a function of the electron source size and number of emitted electrons per pulse to obtain an ideal electron pulse setting for UED experiments. First, electron pulse durations $\tau_e$ and source-limited transverse coherence lengths $\xi_1$ were simulated by means of our particle tracking algorithm. Corresponding outcomes are summarized in figure 12. The pulse duration changes as expected and already discussed in section 3.2. The results of the lateral coherent properties reveal that a source-limited transverse coherence length $\xi_1$ above 10 nm is achievable in our setup with an electron source radius of less than 5 μm. Furthermore, we convoluted coherence values $\xi_1$ with the magnetic lens and detector PSFs from section 3.1. Separate calculations of individual magnetic lens PSFs for each of the simulated electron pulse settings were not included. Finally, the instrumental brightnesses $B_1$ were determined by equation (9). The right-hand graph in figure 12 shows the simulated result. It clearly demonstrates for our UED setup that smaller sources with radii below 5 μm indeed reach highest transverse coherences; the instrumental brightness however is less in comparison to larger sources. Reasons for this are enlarged pulse radii, longer pulse durations and less electrons per pulse at the sample that are all caused by intensified space charge forces. The higher transverse coherence of a smaller source would only shift the brightness distribution downwards with a significant improvement of both the magnetic lens and detector resolution as mentioned above.

However, the instrumental brightness graph in figure 12 demonstrates that the highest signal yield in combination with a short pulse duration as well as enhanced transverse coherence is achieved in case of the 9.5 μm source size and 1500 emitted electrons per pulse (indicated by a green filled white circle in the graph).
3.4. Performance-optimized time-resolved experiment on multilayer graphene

The performance of our highly compact ultrafast electron diffractometer is lastly exemplified in a time-resolved diffraction experiment with optimized electron pulse parameters. We have observed the in-plane lattice heating in multilayer graphene after fs laser excitation and analysed the results by means of the Debye–Waller theory according to [16].

The multilayer graphene sample shown in figure 2 was excited optically at the fundamental wavelength of the amplifier system with a mean incident fluence of \((6.2 \pm 0.4) \text{ mJ cm}^{-2}\) at a pulse duration of \((27 \pm 3) \text{ fs (FWHM)}\) (see section 2.1 for experimental details). Lattice dynamics were probed by electron pulses from the 9.5 \(\mu\text{m}\) source with \(1500 \pm 50\) electrons per pulse at the sample and measured pulse durations of \((230 \pm 32) \text{ fs (FWHM)}\).

High-definition diffraction patterns were recorded at 271 time delay values with variable temporal increments from \(-30\) ps before to 200 ps after laser excitation. Each delay step was approached a hundred times to average out short-term laser and instrumental fluctuations. For each approach, the diffraction patterns of 660 laser shots were added up. Consequently, a total exposure time of 66 s or \(\sim 10^8\) electrons were accumulated for each delay value, resulting in an overall measurement time of 12.5 h for time-resolved data at highest quality. Figure 13(b) exemplifies a resultant diffraction pattern at negative time delay.

For evaluation of the transient diffraction data, a total number of 101 Bragg peaks up to a lattice vector of \(|\mathbf{G}| = 18.4 \text{ Å}^{-1}\) were first indexed as shown in figure 13(b). Some diffraction spots were not considered due to perturbing stray light from the pump laser. Afterwards, each selected Bragg peak was individually fitted within a rectangular image section (indicated by squares in figure 13(b)) by a 2D Gaussian distribution (see section 3.1 and figure 6 for fit details). The fitting procedure enables the time-resolved analysis of the peak intensity and moreover a sub-pixel determination of the peak width and position. To give an impression of the instrumental performance, we will only focus on intensity changes in the following.

The evolution of peak intensities after laser excitation is exemplarily illustrated in figure 13(a) by six selected Bragg peaks in [210] direction of the direct lattice. The dynamics clearly show a fast and a slow decay component and were thus fitted by bi-exponential decay functions (indicated by solid lines) according to [16]. A complete evaluation of the transient diffraction data resulted in mean time constants of \((444 \pm 16) \text{ fs}\) and \((11.8 \pm 0.2) \text{ ps}\) for the fast and slow decay, respectively. The small error limits point out the high data quality and large number of analysable Bragg peaks. The underlying physical processes in graphite and graphene have been widely investigated and are still under debate [14–19, 80–85]. However, a detailed discussion of phonon decay mechanisms goes beyond the scope of this paper and will be presented elsewhere.

The inset in figure 13(a) exemplifies the excellent setup performance on basis of the residual from fitted 110 Bragg peak intensity dynamics (not shown in the main graph). Assuming that the peak intensity decay is perfectly described by the bi-exponential fit function, the residual represents an upper limit of the diffraction signal noise. A statistical analysis of the residual (indicated by the histogram on the right) resulted in an overall signal noise below \(\pm 0.23\%\) (standard deviation). Taking a mean intensity drop of 6.54\% for the 110 peak into

![Figure 12](image-url). Fundamental electron pulse parameters simulated as a function of electron source size and number of emitted carriers per pulse. White circles in each graph indicate for the three studied source sizes \((1/e^2\) - radii: 2.3 \(\mu\text{m}\) red filled, 9.5 \(\mu\text{m}\) green filled and 28.0 \(\mu\text{m}\) blue filled) the electron pulse conditions discussed in section 3.1. The left graph presents the charge density dependent FWHM electron pulse duration at the sample. Labelled white dashed lines highlight selected contours of isometric temporal resolution. The central graph shows the source-limited transverse coherence length along with labelled contour lines of selected constant electron pulse radii at the sample. It reveals that a transverse coherence above 10 nm is achievable with a source size of less than 5 \(\mu\text{m}\) radius. The right graph demonstrates the instrumental transverse normalized peak brightness given by equation (9).
account, a signal-to-noise ratio above 28 can thus be reached in time-resolved diffraction experiments. It should be additionally noted that the 3 nm thin graphene sample only provides a weak diffraction signal and the estimated signal-to-noise ratio therefore demonstrates a lower limit of the instrumental performance.

Finally, we evaluated the asymptotic intensity decrease of all indexed Bragg peaks in figure 13(b) to give an impression of the analysable amount of diffraction data and its quality. A recent UED study on a 1–3 nm thin graphite film [16] concluded that the thermal equilibrium is reached around 150 ps after laser excitation, which is in compliance with our results at comparable pump fluences. In addition, the final lattice temperature rise could be well reconstructed from both the intensity reduction of Bragg peaks combined with the Debye–Waller theory as well as from calculations including the absorbed laser fluence and temperature-dependent heat capacity of graphite. In the following, we determined the final lattice temperature of our sample first due to the Debye–Waller theory and secondly from the absorbed pump fluence according to [16].

The intensity of a Bragg peak \( I(G, T) \) is reduced for a given reciprocal lattice vector \( G \) and temperature \( T \) by the Debye–Waller factor, usually defined as \( \exp[-2W(G, T)] \). The equilibrium lattice temperature \( T_{eq} = T_0 + \Delta T \)
can thus be calculated from the asymptotic Bragg peak intensity \( I_{\text{asy}}(G, T_{\text{eq}}) \) by
\[
\ln[I_{\text{asy}}(G, T_{\text{eq}})/I_0(G, T_0)] = -2[W(G, T_{\text{eq}}) - W(G, T_0)]
\]
with the initial Bragg peak intensity \( I_0(G, T_0) \) at room temperature \( T_0 = (296 \pm 3) \text{K} \).
Assuming that the measured Bragg peaks in figure 13(b) with Miller indices \( h\bar{k}\bar{l} \) mostly represent in-plane lattice dynamics and the corresponding in-plane Debye temperature \( \Theta_d \) is considerably higher than the final temperature rise \( \Delta T \), \( W(G, T) \) is given by
\[
W(G, T) = \frac{1}{4} |G|^2 \langle u_{\text{m}}^2 \rangle = \frac{1}{4} |G|^2 \left[ \frac{6\hbar^2}{m_c k_B \Theta_d} \left( \frac{T}{\Theta_d} \right)^2 + \int_0^{\Theta_d/T} \frac{\varepsilon}{e^\varepsilon - 1} d\varepsilon \right],
\]
where \( \langle u_{\text{m}}^2 \rangle = \langle u^2 \rangle + \langle u^2 \rangle \) is the temperature-dependent in-plane atomic mean square displacement, \( m_c \) is the mass of a carbon atom, \( k_B \) is the Boltzmann constant and \( \hbar \) is the reduced Planck constant [16, 86, 87].
Therefore, we plotted the relative intensity decrease -\( \ln[I_{\text{asy}}(G, T_{\text{eq}})/I_0(G, T_0)] \) for all indexed Bragg peaks against the square of the absolute value of the reciprocal lattice vector \( |G|^2 \) and fitted the data set by a linear regression with weighting due to the errors of \( I_{\text{asy}}(G, T_{\text{eq}}) \). Figure 13(c) illustrates the corresponding results. The slope of the linear regression was further used as a constraint to solve equation (10) by additionally taking into account a temperature dependence of the in-plane Debye temperature with \( \Theta_d(T_0) = 1330 \text{K} \) [87] and \( \Theta_d(T_{\text{eq}}) = 1627 \text{K} \) [87, 88]. The theoretical temperature behaviour of the Debye temperature was adapted from [88] and scaled onto the experimental value at room temperature [87]. Thus, we finally obtained a laser-induced heating of the multilayer graphene sample to an equilibrium temperature of \( T_{\text{eq}} = (1234 \pm 19) \text{K} \). This precise temperature determination within the applied theoretical approximations could be achieved mainly by the large number of recorded Bragg peaks in combination with relatively low asymptotic intensity errors.

As already mentioned above, we additionally calculated the final lattice temperature \( T_{\text{eq}} \) by means of the absorbed pump fluence and graphite heat capacity according to [16]. The incident fluence was first determined by measuring the pump laser profile at an equivalent sample position outside of the vacuum chamber with a CCD beam profiler. Figure 13(d) illustrates the obtained fluence distribution, which is adequately uniform over the sample area with a maximum fluence deviation of less than 20%. Averaging over the entire sample area resulted in a mean incident pump fluence of \( F = (6.2 \pm 0.4) \text{ mJ cm}^{-2} \). Fresnel’s equations were further used to calculate an absorption of \( \alpha = (0.135 \pm 0.010) \) for the 3 nm multilayer graphene sample at 795 nm by taking into account its specific optical conductance [89] and the angle of incident onto the sample. Thus, an equilibrium lattice temperature of \( T_{\text{eq}} = (1135 \pm 103) \text{K} \) could be finally determined from the incident pump fluence \( F \) by solving
\[
F = \frac{d}{A} \int_{T_0}^{T_{\text{eq}}} C_p(T) dT,
\]
with the temperature-dependent specific heat capacity per volume \( C_p(T) \) of graphite taken from [90], the sample absorption \( A \) and corresponding thickness \( d \).

Both results for the equilibrium lattice temperature, \( T_{\text{eq}} = (1234 \pm 19) \text{K} \) from the Debye–Waller analysis of Bragg peak intensities and \( T_{\text{eq}} = (1135 \pm 103) \text{K} \) from the absorbed pump laser fluence, agree within their error limits. In this connection, the quality of the time-resolved diffraction data is reflected in the considerable lower error of the resultant lattice temperature.

The presented time-resolved diffraction data on multilayer graphene could thus demonstrate that the studied excellent spatio-temporal resolution of our compact ultrafast electron diffractometer is unrestrictedly applicable for challenging future UED experiments.

4. Summary and conclusions

In this paper, we extensively investigated the interplay between spatial and temporal aspects of resolution limits in UED experiments on the basis of a newly designed, highly compact ultrafast electron diffractometer. The focus was directed to the robust generation of uncompressed multi-electron pulses with excellent spatio-temporal properties in the lower space charge regime as well as a successful application of these pulses to time-resolved experiments. In order to accomplish this, we have shown that the propagation distance of electron bunches from the source to the sample can be reduced threefold in comparison to so far most compact source designs by installing the magnetic lens behind the sample. Additionally, a static acceleration field strength above 10 MV m\(^{-1}\) could be achieved with a pressed in, glue-free implementation of the photocathode substrate. These improvements diminish the longitudinal and lateral electron pulse broadening and allowed a reduction of the electron source size to extend the transverse coherence while maintaining an ultrashort pulse duration. Accordingly, we investigated the dependence of the electron source size and charge density on the transverse coherence length, electron pulse duration, diffraction signal intensity and instrumental brightness for reaching an optimized instrumental performance.
Three different electron source sizes were studied with a variation of the source area over more than two orders of magnitude. The transverse coherence length of the electron pulses was determined by three experimental approaches and a self-developed particle tracking program. Moreover, we directly extracted the transverse coherence length from the fitted widths of measured diffraction peaks. The smallest studied source sizes achieved the best coherence values up to 20 nm. By taking magnetic lens aberrations and a detector signal spreading into account, we revealed an up to sevenfold enlargement of Bragg peaks due to instrumental imperfections. However, a deconvolution via instrumental PSFs finally confirms the expected dependence of the Bragg peak intensity on the coherence. Our highly compact UED setup can therefore provide multi-electron pulses at the sample with a maximum transverse coherence length of about 20 nm while maintaining a bunch $1/e^2$ radius of less than 85 μm.

Due to the fact that the coherence-improving source reduction inevitably increases space charge effects, we investigated the temporal pulse broadening as a function of the number of electrons per pulse for the three studied source sizes. Close to the single-electron regime, experimental as well as simulated outcomes have shown a minimal FWHM pulse duration of 120 fs for all source sizes resulting in overall temporal resolution of 60 fs (RMS). For 3000 electrons per bunch an outstanding RMS temporal resolution of 78 fs is reached using the largest investigated source radius at the cost of a low transverse coherence. Increasing the coherence to the highest value moderately reduces the RMS temporal resolution to 147 fs for ~1000 electrons per bunch.

On the basis of the spatio-temporal resolution studies, we further analysed influences on the diffraction signal intensity and electron pulse brightness. The expected quadratic dependence of the Bragg spot peak intensity with the transverse coherence length was only confirmed in absence of instrumental imperfections. Therefore, we extended the standard brightness definition to find optimal electron pulse settings for highest signal yields in space and time at a certain temporal resolution.

Finally, the full performance of our UED setup was demonstrated in a time-resolved experiment with optimized electron pulse parameters. We observed the in-plane lattice heating in multilayer graphene after signal yields in space and time at a certain temporal resolution. The high quality of the diffraction pattern thereby allowed the evaluation of 101 Bragg peaks at each pump–probe delay up to a reciprocal lattice vector of 18.4 Å$^{-1}$. The statistical analysis of a selected Bragg peak intensity decay additionally demonstrated the high signal-to-noise ratio in the transient data. Thus, we conclude that the excellent spatio-temporal capability of the presented electron diffractometer is certainly applicable for many prospective UED experiments and we expect a high impact of this design on the field of ultrafast structural dynamics.

As an outlook this setup can be combined with ultrafast laser control techniques (see for example [91] and references therein) to study light–matter interaction with shaped laser pulses in order to guide material response towards user-designed directions.

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Appendix A. Transverse coherence definition

The transverse coherence length of an electron beam is a measure of the maximum resolvable distance in a sample over which the wave function describing the electrons is considered to be in phase. For an electron pulse with Gaussian phase space distribution, the transverse coherence length $\xi_x$ along the lateral space coordinate $x$ is often related to the standard deviations of the uncorrelated half-angle divergence $\sigma_{\theta_x}$ or local uncorrelated transverse momentum spread $\sigma_{\nu_x}$ by

$$\xi_x \equiv \frac{\lambda}{2\pi \sigma_{\theta_x}} = \frac{\hbar}{\sigma_{\nu_x}}, \quad (A.1)$$

where $\hbar$ is the reduced Planck constant and $\sigma_{\theta_x} = \sigma_{\nu_x}/p_z$ in the paraxial approximation with the mean longitudinal momentum $p_z$ of the electrons. The transverse coherence length $\xi_x$ along the lateral space coordinate $x$ is given analogously. This definition is widely accepted in the field of UED [3, 5, 25, 40, 41, 54, 65].

From equation (A.1) directly follows an expression of the transverse coherence length $\xi_x$ in terms of the local uncorrelated transverse wave vector spread $\sigma_{k_x}$ by
In state of the art UED experiments the transverse coherence length at the sample is typically orders of magnitude smaller than the lateral electron pulse diameter [4, 5] and the global correlated half-angle divergence is about a few mrad (≤ 4 mrad in our setup). Under this conditions the transverse coherence length $\xi_x$ is only marginally reduced by the correlated angle divergence and can be well approximated analogously to equation (A.1) by means of the transverse emittance $\varepsilon_{n,x}$ as follows: In regions of a setup where the electron pulses are focused or collimated, the correlated angle divergence vanishes and the normalized RMS emittance $\varepsilon_{n,x}$ of equation (2) can be reduced to

$$\varepsilon_{n,x} = \frac{\sigma_x \sigma_{p,x}}{m_0 c} = \frac{w_x \sigma_{p,x}}{2m_0 c},$$

with the standard deviation $\sigma_x$ of the spatial electron pulse profile along the lateral coordinate $x$, the equivalent pulse $1/e^2$-radius $w_x = 2 \sigma_x$, the uncorrelated transverse momentum spread $\sigma_{p,x}$, the electron rest mass $m_0$ and the speed of light $c$ [5, 41, 65]. The emittance $\varepsilon_{n,y}$ for the lateral coordinate $y$ is given analogously. Hence, the combination of equations (A.1) and (A.3) results in the approximation of the transverse coherence length $\xi_x$ through the normalized emittance $\varepsilon_{n,x}$ [5, 41, 65],

$$\xi_x \approx \frac{\hbar}{m_0 c} \frac{w_x}{2 \varepsilon_{n,x}}.$$

This approximation has been used in our particle tracking algorithm instead of equation (A.2) due to a higher numerical robustness and insignificant deviations in the studied parameter regime.

### Appendix B. Particle tracking simulation

A particle tracking program was self-developed to get direct access to all tuneable electron pulse parameters and their effects on the spatio-temporal resolution of our UED setup. 3D charged particle trajectories were calculated under the combined influence of instrumental electromagnetic fields and space charge forces by grouping electrons inside a pulse to so-called macroparticles [93]. A single pulse has been typically simulated by means of 1000 macroparticles regardless of the number of electrons per pulse to allow significant statistical analyses and to give a feasible estimation of the space charge effects for pulses with less than 5000 electrons. Accordingly, each macroparticle has to carry a defined fraction of charge (at a constant mass-to-charge ratio) so that the entire electron pulse charge is fully represented. The macroparticles relativistic equations of motion have been solved relativistically by a 3D

$$\xi_x = \frac{\hbar}{\sigma_{p,x}} = \frac{1}{\sigma_{k,x}},$$

where $\sigma_{p,x} = \hbar \sigma_{k,x}$. Here, the absolute value of the electron wave vector is defined by $|k| = 2\pi/\lambda$, with the de Broglie wavelength $\lambda$. In case of an idealized diffraction experiment, without instrumental limitations and sample imperfections, the standard deviation of the Gaussian Bragg peaks in reciprocal space would be equal to the uncorrelated transverse wave vector spread $\sigma_{k,x}$ in equation (A.2) [25, 74, 92].

Apart from space charge effects, the initial electron pulse phase space distribution directly after photoemission constitutes all important spatio-temporal pulse properties at the sample. Thus, an accurate imitation of the initial phase space distribution is essential to achieve a successful reproduction of experimental pulse parameters by means of the particle tracking simulation. The starting point is the random assignment of initial 3D positions and momenta to the macroparticles from the predefined phase space distribution. Thereby, the transverse spatial electron distribution has been assumed to have a Gaussian shape whose $1/e^2$-radius is given by the UV laser spot size on the photocathode (see section 3). The longitudinal spatial distribution and therefore the...
temporal shape of the electron pulse has been represented by a Gaussian profile as well. The emission of each macroparticle within the UV laser pulse duration (30 fs (FWHM)) has been simulated with a temporal resolution of 120 as. In this process, electromagnetic fields and space charge effects are already taken into account. The initial velocity distribution has been calculated via the three step model of photoemission in accordance with [61, 64]. The shape of the velocity distribution is thereby defined due to the following intrinsic instrumental parameters: UV pulse central photon energy $\hbar \omega = (4.64 \pm 0.03)$ eV, gold cathode workfunction $\phi_0 = (4.26 \pm 0.2)$ eV from [51, 64] and electric acceleration field strength $E_{acc} = U/d = (11.4 \pm 0.7)$ MV m$^{-1}$ with $U = 40$ kV $(10^{-4}$ relative stability) and $d = (3.5 \pm 0.2)$ mm. Furthermore, these instrumental parameters allow an analytical approximation of the intrinsic normalized emittance $\varepsilon_{n,i}$ from the following expression [61–63]

$$\varepsilon_{n,i} = \frac{w_i}{2} \sqrt{\frac{\hbar \omega - \phi_{eff}}{3m_0e^2} }, \quad (B.1)$$

with the electron source 1/$e^2$-radius $w_i$, the laser photon energy $\hbar \omega$, the effective work function $\phi_{eff}$, the electron rest mass $m_0$ and the speed of light $c$. Here, the effective work function $\phi_{eff} = \phi_0 - e^2E_{acc}/4\pi\varepsilon_0$ is the cathode work function $\phi_0$ lowered by the Schottky effect [61, 62], where $e$ is the elementary charge, $E_{acc}$ is the electric acceleration field strength and $\varepsilon_0$ is the vacuum permittivity. Thus, the combination of equation (B.1) with equation (A.4) results in an initial transverse coherence length of $\xi_1 = (0.67 \pm 0.16)$ nm for our setup, which provides the basis for further transverse coherence analyses in section 3.1.

B.2. Simulating the PSF of the magnetic lens
In section 3.1, magnetic lens PSFs are used to quantify the source-limited transverse coherence lengths in dependence of the electron pulse source size. The determination procedure of the magnetic lens PSFs for all three studied source sizes is described in the following.

For each of the finite source sizes, the experimental conditions were first reconstructed by tracing the overall propagation of macroparticle bunches (always representing 1500 electrons per pulse) from their point of generation at the cathode to the detector plane, including the diffraction process in 210 direction of the graphene sample (see figure 7). In these simulations, the minimal spot size of the resulting 210 Bragg peak on the detector could be well retrieved by optimizing a scale factor for the global magnetic field strength. It should be noted that for all source sizes the optimal scale factors differed less than 10% from the parameter-free finite element results of the FEMM program (see also figure 7). The actual determination of the PSFs was based on the reconstruction of the experimental fields.

Influenced by lens aberrations and space charge effects, a magnetic lens PSF simply represents the focussed intensity distribution of an electron point source on the detector. However, particularly within the space charge regime, an ideal point source is not realizable. Therefore, we determined the magnetic lens PSFs by mimicking point sources in the following way: Electron pulses from a finite source area were propagated until 1 mm behind the anode at $z = 4.5$ mm, where the electric acceleration field and magnetic lens fields are both negligible (see figures 3 and 7). At this point the projection of the lateral phase space distribution (shown in figure B.1 for the lateral coordinate $x$) clearly demonstrates the influence of the source size on the uncorrelated transverse velocity spread $\sigma_{v,x}$ (standard deviation of the velocity distribution along a given position $x$). This velocity spread $\sigma_{v,x}$ is directly related to the uncorrelated transverse momentum spread $\sigma_{p,x} = m_0 \gamma \sigma_{v,x}$ via the electron rest mass $m_0$ and the Lorentz factor $\gamma$. The momentum spread $\sigma_{p,x}$ further defines the transverse coherence length of the electron pulse (see appendix A for details). To finally achieve a macroparticle ensemble with point source properties, we altered the phase space distribution at $z = 4.5$ mm as follows: The projected distributions were linearly fitted independently in $x$- and $y$-direction and then the transverse velocity components of each macroparticle were set to the values of the linear fit functions at its position (see figure B.1 for details). This modification creates a laterally fully coherent electron pulse with a perfect linear position-momentum correlation like bunches from an electron point source. In addition, the method preserves the specific transverse diameter and divergence angle of the electron pulses from each of the three studied source sizes. The further propagation of the fully coherent electron pulses to the detector plane at $z = 188$ mm was simulated similarly to the finite source case above under the influence of diffraction and the predetermined optimal magnetic field strength. The final intensity distributions of the magnetic lens PSFs were achieved by integrating over 1000 single simulations cycles, while each macroparticle ensemble represented 1500 electrons per pulse.
B.3. Instrumental brightness definition

According to [66, 97], the transverse normalized peak brightness $B_n$ is defined by

$$B_n \equiv \frac{I_0}{4\pi^2 \varepsilon_{n,x} \varepsilon_{n,y}},$$  \hspace{1cm} (B.2)

with the peak current $I_0$ and the transverse normalized emittance $\varepsilon_n$ in $x$- and $y$-direction given by equation (2). Due to the cylindrical symmetry of our apparatus we have $\varepsilon_{n,x} = \varepsilon_{n,y} = \varepsilon_n$. The factor $4\pi^2$ originates from the assumption of a transverse 4D Gaussian shaped phase space distribution. Assuming a Gaussian profile for the longitudinal pulse distribution as well, the peak current $I_0$ is given by

$$I_0 \equiv \frac{Ne}{\sqrt{2\pi} \sigma_t},$$  \hspace{1cm} (B.3)

where $N$ is the number of electrons per pulse, $\sigma_t$ is the electron pulse duration (standard deviation) and $e$ is the elementary charge. From equation (A.4) follows an approximation of the normalized emittance $\varepsilon_n$ by the use of the source-limited transverse coherence length $\xi_t$ and $1/e^2$-radius of electron pulse $w_e$ at the sample. Taking into account that the source-limited coherence $\xi_t$ is normally reduced by instrumental imperfections to the instrumental coherence $\xi_I$ (see sections 3.1 and 3.3 for details), the instrumental transverse normalized peak brightness $B_I$ is finally given from the combination of the above equations (B.2), (B.3) and (A.4)

$$B_I \approx \left(\frac{m_0 c}{\pi \hbar}\right)^2 \frac{8 \ln 2}{2\pi} \frac{Ne \xi_I^2}{\tau_e w_e^2},$$  \hspace{1cm} (B.5)

where the standard deviation $\sigma_t$ of the electron pulse duration is additionally replaced by the FWHM duration $\tau_e = \sqrt{8 \ln 2} \sigma_t$.

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Figure B.1. Simulated projections of electron pulse $x$-$\gamma v_x$ phase space distributions at $z = 4.5$ mm for all three studied source sizes ($1/e^2$-radii: 2.3 $\mu$m red, 9.5 $\mu$m green and 28.0 $\mu$m blue transparent point cloud). The distributions clearly demonstrate the influence of the source size on the uncorrelated transverse velocity spread $\sigma_{v,x}$, which is expressed in different widths of the velocity distributions along a given particle position ($\sigma_{v,x}$ is exemplified by the inset in the right graph). An ideal electron point source would therefore lead to a perfect position-momentum correlation without any local velocity spread ($\sigma_{v,x} = 0$ for all $x$). Since the determination of the magnetic lens PSFs bases only on mimicked point sources (see text for details), we changed the velocity components of the macroparticle ensembles in our PSF simulations for each cycle at $z = 4.5$ mm as follows: the projected distributions were linearly fitted in $x$- and $y$-direction independently (fit results are shown as magenta dashed lines). Afterwards, the macroparticles velocity components were projected isopositional onto the linear fit results (exemplified by white arrows). The resulting particle distributions (shown as black points) thus obtained point source characteristics, while their specific transverse diameters and divergence angles (linear slopes of the distributions) were preserved.
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