Electron gun for diffraction experiments off controlled molecules

Nele L M Müller 1, Sebastian Trippel 1, Karol Długolecki 1 and Jochen Küpper 1,2,3,4

1 Center for Free-Electron Laser Science, DESY, Notkestrasse 85, 22607 Hamburg, Germany
2 The Hamburg Center for Ultrafast Imaging, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany
3 Department of Physics, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany
4 E-mail: jochen.kuepper@cfel.de

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Abstract
A dc electron gun, generating picosecond pulses with up to $8 \times 10^6$ electrons per pulse has been developed. Its applicability for future time-resolved diffraction experiments on state- and conformer-selected laser-aligned or oriented gaseous samples was characterized. The focusing electrodes were arranged in a velocity-map imaging spectrometer configuration. This allowed us to directly measure the spatial and velocity distributions of the electron pulses emitted from the cathode. The coherence length and pulse duration of the electron beam were characterized by these measurements combined with electron trajectory simulations. Electron diffraction data from a thin aluminum foil illustrated the coherence and resolution of the electron-gun setup.

Keywords: controlled molecules, electron diffraction, molecular physics, laser physics, alignment, orientation, imaging

Diffractive imaging is a promising approach for unravelling the microscopic details of chemical processes through the recording of so-called molecular movies in the gas-phase, which trace the structural dynamics of individual molecules and nanoparticles at the atomic level. Electron and x-ray diffraction are well established tools for investigating the structures of solid state samples [1], for example in transmission electron microscopy [2] or x-ray crystallography [3, 4]. Furthermore, electron diffraction has been found to have broad applications for gas-phase structure determination in chemistry [5]. Recent developments have mainly focused on realizing time-resolved experiments in order to study structural dynamics, where x-ray and electron diffraction serve as complementary approaches [6–11].

To be able to record structural changes during the ultrafast molecular processes of small complex molecules in the gas-phase, signals from many identical molecules have to be averaged. Gas-phase investigations pose the challenge that the sample might comprise different isomers and sizes [12]. In addition, the molecules in the gas-phase are typically randomly oriented. It is therefore important to provide samples, which are as clean and defined as possible, to allow for experimental averaging over multiple electron pulses. Clean molecular samples can be generated by their spatial separation according to shape [13–15] and size [16]. Controlling the spatial orientation of the molecules leads to an enhancement of the information that can be retrieved from a diffraction pattern, as proposed theoretically [17–20] and demonstrated experimentally for x-ray [21, 22], as well as for electron diffraction [23, 24]. Strong alignment or orientation is generally necessary [11, 20] for three-dimensional structure reconstruction [24] and can be provided in cold supersonic molecular beams by strong-field laser alignment and mixed-field orientation [25–29]. The low density of these controlled gas-phase samples requires sources of large cross-section particles or photons with a high level of brightness, while still ensuring atomic resolution. Electron sources can meet these requirements even in table-top setups.

The first sources that enabled the creation of electron pulses short enough to study ultrafast processes in molecules
or materials were dc electron guns. Here, electrons were created from metallic surfaces by short laser pulses and accelerated in dc electric fields [7, 8], yielding sub-pico-second electron pulses of moderate coherence and brilliance. Radio-frequency cavities allow for the temporal compression of electron pulses through phase-space rotation, shortening the pulse duration to below 100 fs with electron numbers of $10^6$ per pulse and electron spot sizes below 100 $\mu$m [30]. Compact dc guns can achieve comparable properties by increasing the acceleration fields and reducing the path length, during which the electron pulse can expand [31–34]. Ultra-fast single-electron sources [35] avoid the problem of space charges, but rely on very high repetition rates to achieve sufficient electron fluxes for diffractive imaging experiments. The use of ultra-cold atoms as electron sources increases the coherence [36, 37]. Other possible sources for time-resolved electron diffraction are low energy electron setups [38] or laser-induced electron diffraction [39, 40].

If the molecular samples are prepared in the necessary strongly-controlled fashion, their densities are typically on the order of some $10^8$ molecules $\text{cm}^{-3}$ [21, 41]. Assuming Rutherford scattering, for the prototypical molecule 2,5-diiodobenzenitrile an effective cross section on the order of $10^{-15}$ cm$^2$ can be derived for our experimental geometry, with a beam stop blocking a solid angle of $1.3 \times 10^{-3}$ sr. To align or orient the molecules, they are typically exposed to laser fields with intensities of 1 TW cm$^{-2}$ [25], which can be achieved by focusing the ps-duration mJ-pulse-energy laser beam to 100 $\mu$m [42]. For a 500 $\mu$m thick molecular beam this results in an interaction volume of about $5 \times 10^{-6}$ cm$^3$. The number of molecules in this volume and the given cross section lead to an elastic scattering signal on the detector $S_{\text{elastic}}$ of $5 \times 10^{-5}$ per electron. In order to achieve a diffraction pattern containing some $10^9$ scattered electrons on the detector within 24 hours, bright electron sources with $10^9$ electrons per second are needed. For experimental repetition rates on the order of 1 kHz [42], this corresponds to $10^6$ electrons per shot with an electron beam focus size of approximately 100 $\mu$m. The setup presented here produced the necessary electron numbers and allowed for a characterization of the electron beam to ensure, for instance, the required transverse coherence length.

A schematic of the experimental setup is shown in figure 1. The electrons were photo-emitted from a copper cathode via one-photon absorption after irradiation with short UV laser pulses. The pulses were generated by third-harmonic generation (THG) of near-infrared pulses 30 fs in duration from a Ti:sapphire laser (TSL) system with a repetition rate of 1 kHz. Based on the pulse duration of the near-infrared laser pulse at the THG setup, and the dispersion in subsequent optical elements, we estimated a pulse duration of 370 fs for the UV light. The central wavelength was 265 nm with a spectral width of 4 nm. The light enters and exits the chamber through anti-reflection coated windows. The pulse impinged on the cathode under an angle of 70 $^\circ$ normal to the surface of the cathode. The electrons were accelerated and focused by three electrodes in velocity-map imaging spectrometer (VMI) configuration. The applied potential at the cathode was $U_c = -15$ kV. The voltage on the focusing electrode was varied between $U_f = 0$ kV and $-15$ kV. The third electrode was kept at ground potential. The asymmetric electrode shape allowed the laser beam to pass and impinge on the cathode. The holes within the electrodes were large enough to avoid clipping the electron beam. On the one hand, this reduced the background signal in the diffraction experiments, as there was no electron scattering off the electrodes. On the other hand, it allowed for the steering of the electron beam’s position by changing the laser-spot position on the cathode. The voltage on the focusing electrode $U_f$ determined the position of the electron beam focus along the $z$ direction. The electrode configuration allowed us to characterize the electron pulse by applying the corresponding voltages for spatial- and velocity-mapping [43–45]. It is possible to create electric fields that allow for the mapping of either the spatial distribution of the electrons at the cathode or their respective velocity distribution as a 2D projection onto the detector [43, 44]. The spatial distribution of the electron beam was recorded by a position-sensitive detector consisting of a multi-channel plate (MCP) with a phosphor screen and a CMOS camera (Optronis CamRecord CL600x2). The detection system was read out with a 1 kHz repetition rate, which allowed for single electron counting in the case of a few electrons per pulse. At large electron numbers, the gain of the detector had to be reduced to avoid damaging the MCP. With lower gain, single electrons could not be resolved any more. In order to reduce the background from scattered light or other sources, the detector can be gated. A Faraday cup connected to an electrometer (Keithley 6514 electrometer) was used to measure the electron number per pulse. To further characterize the electron pulses, we performed diffraction experiments with a thin aluminum foil on a TEM grid, which was introduced into the electron beam path. In this case the direct electron beam was blocked by a copper or aluminum beam block.

![Figure 1](image-url)
The electron gun was designed for ultra-high vacuum. Here, the final pressure was $4 \times 10^{-9}$ mbar using a turbo-molecular pump with a pumping speed of 300 l s$^{-1}$, limited by outgassing from the cable of the Faraday cup and from the PEEK material of the electron gun insulators. This low pressure is essential when investigating thin gas-phase samples in order to reduce the scattering by background gas. We expect to achieve pressures of a few $10^{-10}$ mbar in the final setup when replacing all PEEK insulators by MACOR or alumina.

In figure 2 the electron number per pulse is shown for $U_f = -13.2$ kV as a function of the laser pulse intensity for two laser polarizations. This focusing voltage corresponded to the focus of the electron beam being close to the detector surface. The number of electrons increased linearly with the laser power, as expected for a one-photon effect of 265 nm light with a spectral width of 4 nm on copper, which has a work function of 4.7 eV. No saturation was observed. The number of generated electrons depended on the laser polarization: for p-polarization (red curve, field vector in plane of incidence) more electrons were generated than for s-polarization (blue curve, field vector parallel to cathode surface). This is in accord with the reflectivity of copper being higher for s-polarized light than for p-polarized light, which was confirmed by measuring the laser power for both polarizations after the cathode. It was possible to obtain up to $8 \times 10^6$ electrons per shot, which is sufficient for the planned diffraction experiments on dilute gas-phase samples delivered by the controlled-molecules apparatus.

For a full characterization of the electron beam, the electron spot size at the detector was measured for various focusing voltages $U_f$, including those for spatial- and velocity-mapping. In figure 3(a) the root-mean-square (RMS) spot size of the electron beam in the $x$ and $y$ dimension, $\sigma_x$ and $\sigma_y$, are plotted as a function of $U_f$. Here, the laser intensity was reduced to less than 10 MW cm$^{-2}$ to create approximately five electrons per pulse from the cathode, and space charge effects were therefore negligible. The spot size decreased with increasing $U_f$ until it reached a focus, at the detector, for about $U_f = -13$ kV. Raising $U_f$ led to a further defocusing of the electron beam, i.e. the focus was placed before the detector. The exact voltage at which to place the focus onto the detector depends on the initial size of the electron cloud. Therefore, the foci in the $x$ and $y$ dimension had slightly different focusing behaviour.

In spatial imaging mode, $U_f = -13.3$ kV, the spatial distribution of emitted electrons was mapped onto the detector, which is depicted as an inset in figure 3(a). In this case, all electrons, which started from a certain coordinate on the cathode, hit a corresponding point on the detector, in first order independent of their momentum [44]. The magnification factor $m$ for spatial imaging was calibrated in the experiment by translating the laser-focus spot on the cathode using the focusing lens. From a known displacement of the electron beam on the cathode $\Delta x_C$ and the corresponding measured displacement of the electrons on the detector $\Delta x_D$ a magnification factor of $m = \Delta x_D / \Delta x_C = 3.9$ was determined. This agreed with the simulated value. For simulations of electric fields and trajectories, finite-element methods were
used (COMSOL Multiphysics). The inferred RMS sizes of the electron beam at the cathode were $\sigma_x = 85(3) \, \mu m$ and $\sigma_y = 31(1) \, \mu m$; the values in parenthesis depict one standard deviation. The difference in spread originated from the laser impinging on the cathode under an angle of incidence of 70°. The angle led to an effective broadening of the photoemission laser by a factor of approximately three in the $x$ direction, while the $y$ dimension was unchanged. Thus, the created electron beam was broader in the $x$ direction than in the $y$ direction on the cathode, which was confirmed in the spatial imaging measurements.

In velocity map imaging mode ($U_f = -11.35 \, kV$) the transverse velocity distribution was mapped onto the detector, which is shown as the second inset in figure 3(a). The velocity spread was similar in both dimensions. With the simulated magnification factor of $m_v = 0.9$ for velocity mapping and a simulated electron time of flight of 4.1 ns, an energy spread of $\sigma_E = 0.1 \, eV$ was obtained. This agrees well with the previously reported value $\sigma_E = 0.13 \, eV$ [46].

In order to characterize the electron beam, further simulations and measurements at various focusing voltages $U_f$ were performed. It was possible to retrieve the spatial and velocity distribution of the electrons in the $x$ and $y$ dimension from the experiment, but the corresponding values in the $z$ dimension had to be simulated. Electric fields were calculated using finite-element methods (COMSOL Multiphysics) and the electron trajectories in these fields were simulated using ASTRA [47]. The initial spatial distribution at the cathode was taken from the measurements described above. Together with a Fermi–Dirac distribution for the one-photon emission, this led to the emittance values of $\epsilon_x = 0.026 \, \pi \, mrad \, mm$ and $\epsilon_y = 0.010 \, \pi \, mrad \, mm$, and the energy spread in the $z$ direction of $\sigma_E = 0.2 \, eV$. Fitting the emittance to the transverse velocity distributions retrieved from VMI mode, while keeping $\sigma_E$ constant, resulted in $\epsilon_x = 0.029 \, \pi \, mrad \, mm$ and $\epsilon_y = 0.012 \, \pi \, mrad \, mm$, in good agreement with the values obtained from the Fermi–Dirac distribution. Using the fitted input parameters, the overall dependence of the electron beam spot size at the detector on the focusing voltage was simulated. The results are depicted by the magenta and cyan lines in figure 3(a) and are, again, in good agreement with the experimental results. This indicates that the simulated $\sigma_E$ was also sensible. Due to the good agreement between experiment and simulation it is possible to deduce the properties of the electron beam from the simulations, including size, coherence length and pulse duration, for its whole propagation. The coherence length $L_c = h \sigma_x/(m_0 c \epsilon)$, with the electron mass $m_0$ and the speed of light $c$, was determined using ASTRA [47]. At the sample position (11 cm downstream from the cathode) $L_c$ was deduced to be 3 nm in the $x$ dimension and 1.2 nm in the $y$ dimension. The pulse duration at this position was simulated to be 1.4 ps.

Figure 3(b) shows the spot sizes for $2 \times 10^5$ electrons per pulse, where space charges had a significant effect. For the detection of $2 \times 10^5$ electrons per pulse in figure 3(b), the detector voltage had to be reduced and single electron detection was not possible. This implies that figures 3(a) and (b) are only qualitatively comparable. A stronger asymmetry was observed in velocity map imaging mode than above. This could not be reproduced using cylindrical symmetry in electric fields and initial velocities, which was a good approximation in the simulations for small numbers of electrons. Using finite-element simulations it was possible to qualitatively determine the origin of the asymmetry in velocity map imaging mode, but a full simulation of all 3D trajectories for $2 \times 10^5$ electrons was not possible due to too high a computational cost. Simulations for small numbers of electrons showed that the trajectories of the electrons far from the central axis of the spectrometer were disturbed by the asymmetry of the electric field due to the opening in the electrodes, see inset in figure 1. This became more pronounced when space charges led to a significant broadening of the electron distribution. In the case of $2 \times 10^5$ electrons per pulse the radial distribution between the cathode and the focusing electrode was increased by an order of magnitude compared to the case when small numbers of electrons were present. This led to a larger magnification factor in the vertical direction in velocity map imaging mode, and therefore contributed to the asymmetry in the detector image. Secondly, the space charge effect itself led to an asymmetry in the velocity distribution if the electron spot was asymmetric. For an ellipsoid with homogeneous charge density, the velocity in the direction of the shorter axis is higher [48]. In our case, the
velocity distribution along the y direction was larger, as the size of the cloud is smaller in this dimension. Simulating similar electron densities in smaller, but asymmetric volumes showed an asymmetric velocity distribution as well. The velocity was higher in the direction of the smaller expansion, corresponding to the y direction in the experiment. Both effects resulted in the vertical broadening of the electron pulse in velocity-mapping mode.

Simulations in cylindrical symmetry (ASTRA) for one million electrons per pulse provided an approximate value for the pulse duration at the sample position of 60 ps. Although this was much longer than in the case of a few electrons/pulse, it is sufficiently short for the diffractive imaging of aligned and oriented molecules, which we can routinely create and control for hundreds of picoseconds [42, 49].

A thin polycrystalline aluminum sample was used to test the electron-optical properties of the generated electron pulses, for instance, its coherence length and spatial resolution. The inset in figure 4(a) shows a diffraction pattern for 10^3 electrons per pulse averaged over 10^6 pulses, i.e. about 15 min at 1 kHz. The electron beam was focused on the detector, which resulted in a nearly collimated beam at the position of the sample. The typical diffraction rings of a polycrystalline sample were observed [1]. The corresponding radial distribution as a function of momentum transfer q is plotted in figure 4(a). The peaks can be assigned to the allowed face-centred cubic crystal structure reflections for aluminum, and the corresponding Miller indices (hkl) are used to label the peaks [1].

The inset in figure 4(b) shows a diffraction pattern averaged over 10^3 pulses (~1 s) with 10^6 electrons per pulse. A 6 mm wide beam stop was used, but the MCP voltage still had to be reduced to avoid damaging the detector due to many electrons scattered over a small s. Reducing the MCP voltage reduces the gain of the detector system, i.e. the signal on the camera per impinging electron. With this lower gain single, or a few, electrons could not be detected any more. Therefore, the peak intensities in figures 4(a) and (b) cannot be compared quantitatively. Diffraction peaks are still visible in figure 4(b), except for the largest s where the electron number and gain were too small. This implies that the transverse coherence of the electron pulses was larger than 234 pm, while approximately 1 nm was expected from the simulations. The spatial resolution of the imaging experiment was better than 234 pm, the interatomic distance corresponding to the (111) reflection in the diffraction pattern of aluminum. The restriction in resolution due to the lower detector gain will not occur in the envisioned gas-phase experiments, as the sample density will be much smaller. Thus, single-electron detection will also be possible for large electron numbers per pulse.

Using the implemented spectrometer it was possible to experimentally obtain the emittance, i.e. the initial transverse spatial and velocity distributions of the electrons. The combination with simulations allowed for the deduction of further values, such as the coherence length and pulse duration of the propagated electron pulses. Compared to other sources with time resolutions on the order of 1 ps or below [7, 8], our setup did produce electron pulses with a duration of 1.5 ps for small numbers of electrons/pulse. More importantly, our table-top setup allows for a stable production of >10^6 electrons/pulse at a repetition rate of 1 kHz with an estimated pulse duration of 60 ps. Nevertheless, due to the negligible cross-sections, radiation damage can be neglected even on these long timescales. For the prototypical 2,5-diodobenzonitri-}

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