Velocity-map imaging for emittance characterization of multiphoton-emitted electrons from a gold surface

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A velocity-map-imaging spectrometer is demonstrated to characterize the normalized transverse emittance of photoemitted electron bunches. The two-dimensional (2D) projected velocity distribution images of photoemitted electrons are recorded by the detection system and analyzed to obtain the normalized transverse emittance. With the presented distribution function of the electron photoemission angles a mathematical method is implemented to reconstruct the three-dimensional (3D) velocity distribution curve. As a first example, multiphoton emission from a planar Au surface is studied via irradiation at a glancing angle by intense 45 fs laser pulses at a central wavelength of 800 nm. The reconstructed energy distribution agrees very well with the Berglund-Spicer theory of photoemission. The normalized transverse emittance of the intrinsic electron bunch is characterized to be 0.52 and 0.05 π·mm·mrad in X- and Y-directions, respectively.

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I. INTRODUCTION

Time-resolved imaging of both transient molecular structure and condensed phase dynamics with picometer-femtosecond spatiotemporal resolution has recently become possible with the advent of x-ray free-electron lasers (XFELs) [1–7]. The high x-ray brilliance, coherence, and ultrashort pulse durations available from these sources are the key properties [8] that open up unprecedented opportunities for new science. Therefore, precise control of the x-ray pulse characteristics, including spectral coverage and temporal and spatial beam profiles are of utmost importance for advanced applications. These parameters are directly influenced by the properties of the electron bunch generating the x-ray pulses. Therefore, the accurate characterization of the electron beam quality is indispensable for assessing available approaches in order to enable improvements of the underlying electron beam technology. In addition, high quality electron bunches are instrumental in experiments where materials are studied using electron diffractive imaging [9–12].

The key measure in electron beam quality is electron beam emittance, i.e., the transverse phase-space distribution of the generated electron bunches. To quantify electron beam emittance as a function of photocathode composition and emission mechanisms, we demonstrate a velocity-map-imaging (VMI) spectrometer that allows us to directly access the transverse momentum distribution of photoemitted electrons, enabling the measurement of normalized transverse emittance from various cathodes. Usually, emission mechanisms are classified as thermionic emission, photoemission, or tunneling emission under extraordinarily high electric fields. More recently, nanostructured and plasmonic photocathodes used with multiphoton or strong-field optical emission have been used as improved electron sources [13–18]. Both, the experimental characterization and the theoretical description of the electron emittance from such cathodes is highly important, which motivates the direct VMI measurements developed here.

As a first proof-of-principle example, we report on quantitative measurements of multiphoton emission from a 400 nm thick Au thin film at room temperature, which was excited with 45-fs laser pulses centered at 800 nm. These measurements additionally allowed us to benchmark the performance of this new experimental setup. Quantum-yield-dependent measurements were performed by recording the events of electrons impinging on the detector when varying the average laser power and the polarization angle, respectively. These experimental results confirm that four-photon emission occurs from the planar Au surface. In our experiments the 2D transverse velocity/momentum distribution of photoemitted electrons was directly imaged onto the detector. An experimental 3D energy distribution was reconstructed from the measured 2D VMI data using a mathematical algorithm (vide infra) and compared to the theoretically derived 3D-space energy distribution from the Berglund-Spicer photoemission model [19–22]. The very good agreement of our experimental results with the theoretical model demonstrates the applicability of VMI for the characterization of the normalized transverse emittance of photoemitted emitters.

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II. EXPERIMENTAL SETUP

The velocity-mapping technique maps the velocity coordinates of particles onto a 2D detector without, to first order, the influence of the spatial coordinates. To achieve this, a configuration of electrostatic lenses, in the simplest case using three parallel electrodes, is employed to spatially tailor the electric fields [23–25]. The electric fields can be also used to image and magnify the spatial coordinates suppressing the effect of velocity coordinates, which is then referred to as spatial-map imaging (SMI) [23]. The spectrometer demonstrated here aims to characterize the electron emittance via characterizing the average spread of electron coordinates in position-and-momentum phase space.

The schematic of the spectrometer is shown in Fig. 1. The sample is mounted on the top of the sample holder, which can be retracted into a load-lock chamber. The load lock is designed for exchanging the sample without breaking the ultra-high vacuum (UHV) of the imaging system. When performing the electrostatic imaging experiments, the sample holder is transferred into the main chamber and brought in contact with the repeller plate to make sure they are at the same electric potential. The main chamber, maintained at 10⁻⁹ mbar, contains a stack of three cylindrically symmetric plates, labelled repeller (R), extractor (E) and ground (G) electrodes in Fig. 1. They are arranged in parallel, separated by 15 mm, and, with applied potentials, serve as the electrostatic lens. This is followed by a 0.5 m drift tube, which ends with a detector assembly consisting of a double microchannel plate (MCP, Chevron configuration), a phosphor screen (P46), and a CMOS camera (Optronics CL600×2) for recording images of the electron distributions. The full configuration is shielded against stray fields by a µ-metal tube. A 800-nm 45-µs Ti:Sapphire laser amplifier with a 3-kHz repetition rate was used to illuminate the sample at a glancing incidence angle of ~84°, with a laser focal intensity spot size of ~17×160 µm² root-mean-square (RMS) on the sample. In our experiments, single-shot electron-distribution images are read out at a repetition rate of 1 kHz, limited by the camera-acquisition frame rate. The average number of electrons emitted per pulse is on the order of one or less, thus, space charge effects, which were reported before [26], are excluded.

To calibrate and optimize the spectrometer field configuration for both SMI and VMI, a fixed potential of -6 kV was applied to both the repeller plate and the sample holder while the ground plate being grounded; see supplementary information for details. While scanning the extractor voltage from -5.8 kV to -4.3 kV, we observed the focusing behavior of the electron bunch depending on the extractor voltage [27]. This behavior is revealed by the RMS of the electron bunch size in X- and Y-directions on the detector shown in Fig. S1 (supplementary information). The SIMION [28] software is used to simulate the electric field configuration and to calculate the electron trajectories from a 2D Gaussian source with σₓ = 140 µm and σᵧ = 15 µm, yielding an RMS behavior curve that fits the experimental results. SMI is obtained at the minimum RMS size, i.e., at an extractor voltage of -5560 V, corresponding to a magnification factor of 7.5. From the measured SMI data, the RMS size is analyzed to be σₓ = 158 µm and σᵧ = 20 µm, which is in good agreement with the simulated electron bunch size and the laser focal spot size. The extractor voltage for VMI conditions is found at -4790 V according to the SIMION simulations and the calibration factor of velocity-per-pixel is 8014 m/s/pixel on the detector. The details of the simulations and experimental calibration are described in the supplementary material. In order to minimize field distortions, the sample front surface should be placed in the same plane as the repeller front surface. However, samples of different thickness lead to a position offset with reference to the repeller front, which strongly influences the field configuration. Therefore, the extractor voltage for operating in SMI and VMI mode are optimized by voltage adjustments of [50, -50] V and [400, -200] V, respectively, to correct for a position offset of [−0.5, 0.5] mm. In this case, re-adjusting the potential right after exchanging a sample is necessary, but quick (vide infra).

III. EXPERIMENTAL RESULTS

Fig. 2a shows the photoemitted electron yield as a function of incident laser energy on a logarithmic scale. The data, shown in red, were measured and averaged over four measurement sequences and the error bars show the corresponding standard deviations of the photoemitted electron counts due to laser fluctuations. The blue line reflects the results of a linear regression analysis that yielded a slope of cₑ ≈ 3.94, with a standard error of 0.04 and a coefficient of determination R² ≈ 0.999.

The Fowler-Dubridge model for the n-th order pho-
The electron yield mostly depends on the bulk absorption coefficient, expressed as term \((1 - R)^n\) in the Fowler-Dubridge model [30]. \(R\) is calculated by Fresnel equations with \(n_1 = 1\) and \(n_2 = 0.189 + i4.71\) [32] at an incidence angle of 84°. The plotted \((1 - R)^4\) curve fits very well with the data, which proves again the 4-th order multiphoton process.

A velocity-map image from a planar Au surface is shown in the inset of Fig. 3a. The image was integrated over 6×10^4 laser shots with an energy of ~50 nJ, corresponding to a peak intensity of 4×10^10 W/cm^2 on the cathode. Generally, in laser-induced multiphoton emission the emitted electron velocity vectors exhibit cylindrical symmetry along the direction normal to the sample surface. Therefore, the center of mass (COM) of the image is set as coordinate origin. The corresponding angle-integrated radial velocity distribution of the projected electrons is plotted in Fig. 3a as black line. To allow for comparison with the theoretical model, the 3D velocity/energy distribution is required. Introducing a novel mathematical method similar to the Onion Peeling algorithm [33], we are able to reconstruct the momentum/energy distribution when the angular distribution of emitted electrons is known. Fortunately, for multiphoton emission, the intensity of photoemitted electrons at various angles \(\theta\) can be derived from the Berglund- Spicer model [20] as

\[
I(\theta) \propto N^2 \cos \theta \cdot \frac{1}{1 + \alpha l(E)} \cdot \frac{1}{\sqrt{1 - N^2 \sin^2 \theta}}
\]

where \(\alpha\) is the optical absorption coefficient, \(l(E)\) is the electron-electron scattering length for an electron of kinetic energy \(E\), and \(N\) expresses the electron analogy of refraction at the vacuum-metal boundary [34]. For a small \(N\) (our case, \(N = 0.275\), i.e., an incident photon energy \(n\nu\) comparable to the work function \(W\), the equation can be simplified to \(I(\theta) \propto \cos \theta\) [35, 36]. Therefore, the 3D velocity distribution can be reconstructed as is described in detail in the supplementary information.

The reconstructed velocity distribution is plotted as blue line in Fig. 3a, and the smoothed energy distribution shown in Fig. 3b. The energy distribution of the emitted electrons shows an energy spread of ~1 eV, which corresponds to the energy difference between a four-1.55 eV-photon excitation and the Au work function of 5.31 eV.

IV. DISCUSSION

The Berglund- Spicer three-step model is employed as the analytic expression for the kinetic energy distribution of the photoemitted electrons. As the model is derived for single-photon emission, it is implied in our analysis that the electrons at an initial energy state \(E_0\) absorb sufficient number of photons instantaneously, rather than sequentially, to be pumped to a higher energy state \(E = E_0 + n\nu\). The kinetic energy distribution for single-photon emission [19] is adapted to multiphoton emission.
Fermi level

0 0.5 1 1.5 2
Velocity (m/s) ×10⁶

0 1.1-1.1
1.1
-1.1
0
×10⁶
×10⁶
0
0.2
0.4
0.6
0.8
1
1.2Norm. counts

2D ... emitted electrons. Since we estimate the absorbed peak intensity for the recorded image, Fig. 3a, to be ∼4×10⁹ W/cm²

1. To evaluate Equation 3, the probability of a photon

\[ E \] where

\[ \sqrt{\frac{\pi k}{\lambda}} \]

related to both independent of electron energy.

\[ \alpha = 4 \frac{\pi k}{\lambda} \] and taken as a constant

\[ \frac{\alpha}{\alpha + 1/l(E)} \]

\[ AE \]

\[ 1 + 4 \left( \frac{E - E_f}{nh\nu} - 1 + \ln \frac{nh\nu}{E - E_f} \right) \] (3)

where \( E_f \) is the Fermi energy of Au. \( C(E) = 0.5 \times (1 - \sqrt{W/E}) \) for \( E \geq W \) is a semiclassical threshold function. \( l(E) \) is the electron-electron scattering length, which is proportional to \( E^{-3/2} \). The absorption efficient \( \alpha \) is calculated from the extinction coefficient \( k = 4.71 \) as \( \alpha = 4\pi k/\lambda \) and taken as a constant \( \alpha = 7.7 \times 10^5 \text{ cm}^{-1} \) independent of electron energy. \( K \) is a correction factor related to both \( C(E) \) and \( \alpha l(E) \), which is between 0.5 to 1. To evaluate Equation 3, the probability of a photon carrying energy \( h\nu \) is calculated from the measured laser spectrum in the range from 760 to 850 nm. To overcome the barrier of 5.31 eV, an electron is assumed to always absorb four photons \((n \equiv 4)\). Absorption of various photon energies leads to slight difference of the quantum yield at a certain emitted kinetic energy as one can see from Fig. 3b. The main consequence of absorbing photons with various energies is the spectral/intensity broadening, which is illustrated by the color coding in Fig. 3b, but with an essentially unchanged spectral shape. We mention that (3) only includes the emitted electrons that experience none or one electron-electron scattering process during transport to the metal-vacuum surface. Electron-electron scattering is dominant over electron-phonon scattering and reshapes the energy distribution on a fast timescale, i.e., during an ultrashort laser pulse.

The density of states (DOS), i.e., the number of states available for electrons at a certain energy level, is shown in the inset of Fig. 3b. During the photoemission process, an energy state \( E_0 \) is first occupied by an electron, which is then excited to a higher energy state \( E \), which was empty. As fermions, electrons obey the Pauli exclusion principle. In thermal equilibrium, the possibility of electrons to occupy an available energy state is given by the Fermi-Dirac (FD) distribution \( f_{FD} \). However, excitation of a metal with ultrashort strong laser pulses initially creates a nonequilibrium distribution that then thermalizes via electron-electron scattering towards a Fermi-Dirac distribution. In gold, this thermalization occurs on a timescale of hundreds of femtoseconds [37, 38]. Subsequently, the electrons cool down by dissipating energy into the lattice via electron-phonon scattering occurring on a longer picosecond timescale. In the following discussion, where we employ the Berglund-Spicer model in our analysis, we assume that the electronic system can be described by a Fermi-Dirac distribution with quasi-equilibrium electron temperature \( T_e \). Hence, the appropriate densities of states and FD distributions are multiplied with the energy distribution as \( N(E) dE \ f_{FD}(E_0) \ DOS(E_0) \ (1 - f_{FD}(E)) \ DOS(E) \), resulting in the spectrum shown in Fig. 3b.

The best fit with our reconstructed experimental energy distribution is obtained for an electron temperature of 6000 K. This is comparable to previously observed electron temperatures of 7000 K in surface-enhanced multiphoton emission from copper [39]. The high energy tail of the spectrum indicates that very “hot” electrons are photoemitted by the femtosecond laser pulse, consistent with the high excess energy deposited into the electronic system. For the energy tail up to 4 eV, except for the high temperature, another process that might need to be taken into account is above-threshold photoemission (ATP), i.e., the absorption of one (or more) extra photon, occurring together with the four-photon process [40]. Moreover, for our experimental conditions, we can neglect tunnel ionization, which could result in high energy emitted electrons. Since we estimate the absorbed peak intensity for the recorded image, Fig. 3a, to be \( ∼4 \times 10^9 \text{ W/cm}^2 \),
taking into account Fresnel losses. This implies a Keldysh parameter \( \gamma = \sqrt{W/2U_p} \approx 17 \gg 1 \), which is well in the multiphoton emission regime; here, \( U_p \propto \lambda^2 I \) is the ponderomotive energy with laser wavelength \( \lambda \) and intensity \( I \).

Since both, the measured quantum yield and the momentum distribution, are in quantitative agreement with the Fowler-Dubridge and Berglund-Spicer models, as one would expect from multiphoton emission from a planar Au cathode, the VMI spectrometer has successfully been implemented as a tool to characterize the photoemitted electrons from cathodes, especially to directly measure the transverse momentum distribution. Assuming there is no correlation between the location of emission and the transverse momentum, the RMS normalized emittance \( \epsilon_n \) is defined as [34]

\[
\epsilon_n = \frac{\sqrt{\langle \zeta^2 \rangle \langle \kappa^2 \rangle}}{mc}, \text{ with } \zeta \in \{X,Y\}
\]

(4)

where \( \langle \zeta^2 \rangle \) is the spatial spread and \( \langle \kappa^2 \rangle \) is the momentum spread of the electron bunch. From the velocity map image shown in the inset of Fig. 3a, the RMS normalized emittance of the planar Au photocathode irradiated by 45-fs 800-nm laser pulses with a focal spot size of \( \sigma_X = 161 \mu m \) and \( \sigma_Y = 17 \mu m \) is characterized to be \( \epsilon_{nx} = 0.52 \pi \cdot mm \cdot mrad \) and \( \epsilon_{ny} = 0.05 \pi \cdot mm \cdot mrad \) in the \( X \) and \( Y \)-directions, respectively. To decrease the intrinsic normalized emittance, in principle one needs to decrease either the emission area or the momentum spread. The former can be intuitively decreased by an extremely tight focal spot size or sharp tip surface, which geometrically limits the emission area. For reducing of the momentum/energy spread, choosing a material with appropriate work function and irradiated by a laser beam with matched photon energy, for example the photoemission of Cu under 266-nm laser irradiation, is expected to help. Further reduction is expected when entering the strong-field emission regime, where the electrons are considered to adiabatically tunnel through the surface barrier with zero initial momentum and are then driven by the instantaneous optical field [13, 41]. Under these conditions electrons are expected to be emitted with a relatively small divergence angle and significantly lower transverse momentum spread.

**V. CONCLUSIONS**

We demonstrated an electron spectrometer with VMI and SMI capabilities, which intuitively allows for the measurement of the normalized transverse emittance of photocathodes, i.e., through the direct observation of the transverse position and momentum distributions. We verified and benchmarked the capabilities of the instrument in a proof-of-concept experiment, in which we characterized the photoemitted electrons from a 400 nm thin Au film. For ultrashort femtosecond laser pulses with a peak intensity lower than \( 10^{12} \) \( W/cm^2 \) at 800 nm central wavelength, which would correspond to \( \gamma = 1 \), multiphoton emission is shown to be the dominant contribution to the entire electron current.

We intend to utilize this new setup for the emittance characterization of electron bunches strong-field emitted from nanotips under optical field irradiation. Such devices should show superior emittance [14, 17]. Moreover, the small radius of the sharp tips realize a field enhancement, which dramatically lowers the laser power required for entering the strong-field regime and thus avoids damaging of the cathodes. Our ongoing work aims at the characterization of electron emission from nanostructured array emitters, which are predicted to provide high-current low-emittance coherent electron bunches in the strong-field emission regime.

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Supplementary information: Velocity-map imaging for emittance characterization of multiphoton-emitted electrons from a gold surface

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I. SPECTROMETER CHARACTERIZATION

The electron spectrometer has been characterized experimentally, accompanied by simulations, in order to determine the focusing conditions for the SMI and VMI modes; see Fig. 1 in the main manuscript for the experimental setup. Fig. S1 shows the measured root-mean-square (RMS) in the X- and Y-directions of the spatial electron distribution on the detector as a function of the extractor voltage, together with the results from SIMION [1] electric field and particle trajectory simulations. A similar behavior as in Ref. 2 is observed.

The strongest focusing of the electron bunch onto the detector is achieved at an extractor potential of -5560 V, which is thus identified as the SMI voltage. The RMS at this voltage shows the magnified laser-surface-interaction area. The slightly different focusing behavior of the electron bunch in the X and Y-directions is attributed to the asymmetric initial electron bunch size, due to the glancing incidence irradiation, and the finite kinetic energy of the electrons.

When increasing the extractor voltage, the electron bunch diverges. Based on our simulations, the extractor voltage for VMI is approximately -4790 V. For a full calibration of the spectrometer, the simulations were used to study the field configuration and the electron trajectories in those fields for the given electrode configurations and the particles initial distributions. In Fig. S1 the simulated RMS of the electron bunch, with electrostatic imaging, at the detector position is plotted as function of extractor voltage. The simulations were carried out given an initial spatial 2D Gaussian distribution of 2000 electrons for each simulated point. The center of mass (COM) of this distribution was given by (X,Y) = (0,0) and a Z-coordinate matching the sample surface with standard deviations of σX = 140 μm and σY = 15 μm. The initial momentum distribution was given by a uniform half sphere with an uniform kinetic energy distribution of electrons in the range of [0.1, 0.6] eV.

The COM of the electron distribution as a function of the initial starting position of the electrons, i.e., the laser focus position on the sample, was used to experimentally calibrate the voltage for velocity-map imaging. Fig. S2 shows the COM as function of the laser position for various voltages together with straight-line fits. A decrease of

FIG. S2. Position dependence of the center of mass of the electrostatic imaging on the detector on the initial source position for various extractor potentials from -5800 V to -4500 V.

FIG. S1. Experimental (hollow) and simulated (solid) root-mean-square deviations of electron spatial distributions on the 2D detector versus focusing extractor voltage in both X and Y-directions. The insets show SMI and VMI detector images for the indicated positions.

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the slope with decreasing extractor voltage is observed. Fig. S3 depicts the slope of each measurement in Fig. S2 as function of the extractor voltage together with a quadratic fit and corresponding simulation results. The error bar for the experimental points is given by the first-order coefficient error of each fitting curve with 95% confidence bounds. VMI mode is obtained at the zero crossing of this curve, i.e., at -4790 V, as for this voltage the extractor voltage (red circles) with a quadratic fit (red line).

VMI -6000 -4790 0 -6000
SMI -6000 -5560 0 -6000

Table I: Voltages (in V) applied for operation in SMI and VMI mode

|          | Repeller | Extractor | Ground | Sample |
|----------|----------|-----------|--------|--------|
| SMI      | -6000    | -5560     | 0      | -6000  |
| VMI      | -6000    | -4790     | 0      | -6000  |

an incident angle of 84°. The SIMION simulation results, also shown in Fig. S4, are in good agreement with the data.

The focusing conditions for the SMI and VMI mode depend strongly on the position of the sample inside the velocity-map imaging spectrometer. Fig. S5 shows the simulated extractor voltages necessary for SMI and VMI mode for various sample displacements with respect to the front surface of the repeller plate. These simulations show that either the sample position has to be known, or at least be reproduced, to a very high precision or calibration measurements have to be performed when a new sample is inserted into the spectrometer. Fortunately, with the protocol described in our manuscript this calibration can be done quickly. In addition the dependence

The focusing conditions for the SMI and VMI mode depend strongly on the position of the sample inside the velocity-map imaging spectrometer. Fig. S5 shows the simulated extractor voltages necessary for SMI and VMI mode for various sample displacements with respect to the front surface of the repeller plate. These simulations show that either the sample position has to be known, or at least be reproduced, to a very high precision or calibration measurements have to be performed when a new sample is inserted into the spectrometer. Fortunately, with the protocol described in our manuscript this calibration can be done quickly. In addition the dependence

**FIG. S3.** Slope of the experimental laser position dependent COM of the spatial distribution at the detector as function of the extractor voltage (red circles) with a quadratic fit (red line). Black points and the black line indicate the corresponding simulated results.

**FIG. S4.** The COM of electrostatic imaging on the detector as a function of the initial source position for SMI mode, i.e., an extractor voltage of -5560 V. The slope in Y-direction is the spatial magnification factor. The ratio between X- and Y-directions confirms the incidence angle of the laser beam of 84°.

**FIG. S5.** Top: SMI and bottom: VMI extractor potential for different position offset from sample front to the repeller front surfaces. FD stands for flying distance in the figure legend.
of the extractor voltage on the flight distance has been investigated (red points and lines). Our simulations show that this uncertainty is uncritical compared to the exact sample position in the spectrometer.

II. RECONSTRUCTION ALGORITHM

Our reconstruction algorithm for the conversion of the 2D projected velocity distribution to the 3D distribution is based on the assumption that the angular distribution of the photoemitted electrons is known. For our simulations, a cosine function \( I(\theta) \propto \cos \theta \) [3, 4], derived from the Berglund-Spicer model [5] as discussed in the main text, is applied in the algorithm. In addition, it is assumed that for multiphoton emission the angular distribution is independent of the modulus of the three dimensional velocity vector. The 3D velocity distribution is then obtained from the 2D projected distribution by a matrix method similar to Onion Peeling [6]. For multiphoton emission from a planar Au surface, the electrons are assumed to be photoemitted within a half sphere of \( \varphi \in [0, 2\pi], \theta \in [0, \pi/2] \). The photoemitted electron distribution has cylindrical symmetry with respect to the surface normal of the sample.

Fig. S6a shows a scatter plot for a single 3D velocity \( v_i \) distribution given by \( f(v, \theta) = \delta(v - v_i) \cos \theta \). Fig. S6b shows the projection of this distribution onto the 2D detector surface. It can be derived that the projected velocity distribution for this special case is

\[
P_i(v_x, v_y) = \int f(v, \theta) \, dv = \begin{cases} C & \text{for } v_{x,y} < v_i, \\ 0 & \text{otherwise} \end{cases}
\]

where \( C \) is a constant. As shown in Fig. S6b, the projected velocity distribution of \( f(v, \theta) \) is constant inside the circular phase-space area of radius \( v_i \). Furthermore, Fig. S6c shows the radial distribution obtained from the projected velocity distribution given by

\[
\rho_i(v_{2D}) = \int P_i(v_x, v_y) \, d\theta_{2D} = \begin{cases} 2\pi C \cdot v_{2D} & \text{for } v_{2D} < v_i, \\ 0 & \text{otherwise} \end{cases}
\]

where \( v_{2D} = \sqrt{v_x^2 + v_y^2} \). In the reconstruction, each radial distribution \( \rho_i(v_{2D}) \) is built up by a triangle as sketched in Fig. S6d. \( v_i \) is taken equally spaced and form the intervals confined by the neighboring gray dashed lines. The 2D projected distribution is related to the 3D distribution \( f_i \) by a transfer matrix \( M \).

\[
\rho_i = M f_i,
\]

with \( M \) given by:

\[
M = \begin{pmatrix}
1 & 1/4 & 1/9 & 1/16 & \cdots \\
0 & 3/4 & 3/9 & 3/16 & \cdots \\
0 & 0 & 5/9 & 5/16 & \cdots \\
0 & 0 & 0 & 7/16 & \cdots \\
\vdots & \vdots & \vdots & \vdots & \ddots
\end{pmatrix}
\]

The 3D distribution can finally be obtained by inversion of the measured 2D-projected distribution

\[
f_i = M^{-1} \rho_i.
\]

![Fig. S6](attachment:image.png)

**FIG. S6.** (a-c) Representation of a simulated electron bunch with a single 3D velocity \( v_i \) and an angular distribution of a cosine function: (a) in 3D, forming a spherical surface; (b) in 2D, yielding a uniform distribution in the detector plane; (c) in 1D, showing a linearly increasing radial velocity \( v_{2D} \) with distance from distribution COM. (d) A conceptual diagram of the reconstruction algorithm: The area of each red triangle at the bottom indicates the number of photoemitted electrons having the same 3D velocity. The corresponding distribution curve is plotted as blue curve. The black curve is the 2D projection distribution curve, summing up the number of photoemitted electrons within each interval of the same transverse velocity. The gray dashed lines indicate the transverse-velocity intervals used in this projection.

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