Velocity Map Imaging of Electrons Strong-Field Photoemitted from Si-Nanotip Arrays

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Abstract: We observe for the first time electron velocity/momentum distributions due to three-photon ionization and strong-field photoemission from Si-nanotip arrays using velocity map imaging.
OCIS codes: (100.3010) Image reconstruction techniques; (320.0320) Ultrafast optics.

Ultrafast and ultra-bright electron sources with spatially structured emission are an enabling technology for free-electron lasers, electron diffractive imaging, compact coherent X-ray sources [1] and attosecond science [2]. The interaction of sharp nanotips with intense few-cycle laser pulses are expected to potentially generate and provide precise control of confined coherent electron wave packets with attosecond duration. These wave packets can be used for the visualization of electron dynamics [3] and nanoplasmonic field dynamics [4]. In this work, we report on the characterization of the transverse electron momentum distribution upon photoemission from a surface when excited with 30-fs laser pulses at 800 nm, using a specially constructed velocity map imaging apparatus (referred to as "solid-state VMI" in the following). The transverse momentum distribution determines the emittance of an electron gun. For calibration purposes, we first performed measurements from flat Ti and Au surfaces and then studied the emission from a novel ultrafast optical-field emission cathode comprised of a large, dense and highly uniform array of nano-sharp high-aspect-ratio single-crystal silicon columns. Such field emitters offer an attractive alternative to UV photocathodes, while providing a direct means of structuring the emitted electron beam. Electromagnetic modeling and experimental characterization both estimate a field enhancement factor of ~10, leading to concentrated electron emission from a tightly localized volume around the tip apex. Particle tracing simulations reveal that the emitted electrons follow rapidly diverging trajectories due to laser acceleration after emission.

The velocity map imaging (VMI) technique has been developed to study electronic or molecular dynamics by detecting the emitted electron or ion distribution [5]. Depending on the extracting electric field created by a specialized electrode configuration, it focuses electrons with the same transverse velocity onto the same spot on a detector assembly consisting of a multi-channel plate (MCP), phosphor screen and CMOS camera, generating an image of the transverse velocity/momentum distribution.

Fig. 1 shows the schematic of the solid-state VMI, which consists of three electrodes (repeller, extractor, and ground electrode) [5] shielded with μ-metal tube with a flying distance of 49 cm in length followed by the detector assembly.

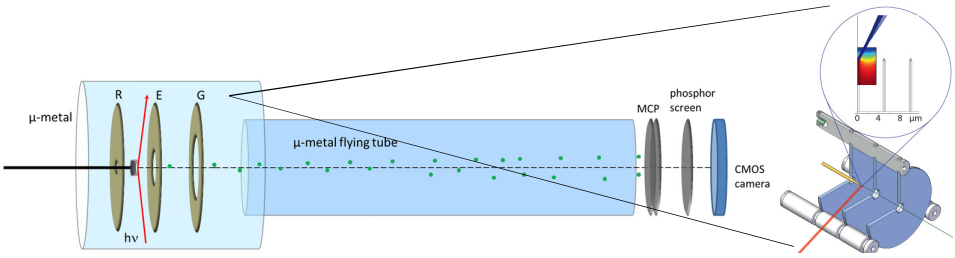

Fig. 1. Configuration of the solid-state VMI. The red solid lines represent the laser beam. R: repeller, E: extractor, G: ground electrode. The green dots represent the emitted electrons.

The Si-samples used in this experiment are uniform arrays of high-aspect-ratio doped silicon pillars, 38-μm high and 1-μm wide with a 5-μm pitch. Each pillar is topped by an ultra-sharp tip with an average radius of curvature of ~5 nm. The sample was treated with Piranha (H_2SO_4:H_2O_2 7:1) and BOE (NH_4F:HF 7:1) solutions to remove any organic residues and the thin native oxide right before mounting into the load lock chamber. Later,

the sample was transferred into the experimental high-vacuum chamber (pressure ~2×10^{-8} mbar) between the repeller and the extractor.

A carrier-envelope phase-stable 800-nm 30-fs Ti:sapphire laser amplifier with a 3-kHz repetition rate was used to illuminate the sample at an incident angle of ~85° and p-polarization with respect to the nanotip direction (see Fig. 1). The laser energy was ~6 μJ with a beam size of ~300 μm in diameter, which corresponds to an intensity of 2.2×10^{11} W/cm^2. The electrons were amplified in the MCP and hit the phosphor screen. The velocity/momentum distribution was recorded on the CMOS camera. During the experiments, the negative voltages supplied to repeller, sample holder and extractor were -6000 V, -5142 V, and -4371 V, respectively, which yields the electric field required for the VMI mode.

Two flat metal samples with different work functions were tested for reference, showing the results in Fig. 2 (b) and (c), where r is the radius of the high-intensity region of the VMI data. The x and y axes refer to the velocities V_x and V_y. Higher transverse kinetic energies cause wider electron distributions in the images. We assume that most electrons generated by the interaction between laser and metal samples are generated by multi-photon ionization. For Ti (work function ϕ=4.33 eV), three photons with 1.55 eV energy are necessary to free an electron, i.e., to overcome the work function. The remaining kinetic energy is therefore 0.32 eV. For Au with the larger work function ϕ=5.1 eV, four photons are required to free the electron, and therefore the remaining kinetic energy is 1.1 eV. The kinetic energy ratio of Au and Ti is 3.43, which is comparable to the ratio of ~4 in the radii of the distributions observed in the different VMI data. The slight difference is due to the nonlinear relationship between the pixels (velocity related) and kinetic energy $dE=mvdv$.

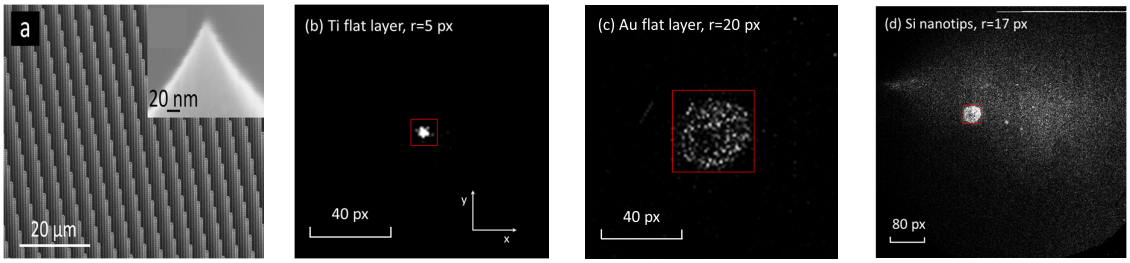

Fig. 2. (a) SEM of the high-aspect-ratio Si-nanotip array; VMI data of (b) flat Ti sample; (c) flat Au sample; (d) Si-nanotip array. The black and white scale represents the relative intensity of electrons detected. The red box indicates the center part of the VMI data.

Fig. 2(d) finally shows the VMI data of the Si-nanotip array. The bright spot with radius r=17 pixels corresponds to a kinetic energy of ~0.9 eV resulting from three-photon ionization [6], which is consistent with the work function of the Si-nanotips of 3.6 eV. The ~1 eV decrease of the initial Si work function can be due to the high doping level of ~10^{16} cm^{-2} carrier concentration, semiconductor affinity and also nonuniformity of the strong electric field near the tips. Fading away from the center, a significant background of electrons is observed with kinetic energy up to ~15 eV. This is consistent with earlier kinetic energy measurements using a time-of-flight (TOF) spectrometer [6], which showed a significantly higher energy contribution in the strong-field emission regime due to rescattering of electrons on the emission surface.

In conclusion, we have constructed a solid-state VMI that enables us to visualize the transverse momentum distribution of electrons emitted from a surface. Test measurements on Au and Ti surfaces gave consistent results relating excess electron energies upon multi-photon emission with the observed phase-space distribution. In the strong-field photoemission regime from Si nanotips we observe a strong emission peak corresponding to an excess energy of 0.9 eV and a large pedestal with kinetic energies up to ~15 eV. By optimizing the solid-state VMI operation conditions and laser pulse parameters, we can further optimize the electron emission process under strong-field conditions to ultimately arrive at optimized electron guns for electron diffractive imaging and coherent X-ray sources.

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