A Nanometer-sized Femtosecond Electron Source at 80 MHz Repetition Rate

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Abstract. We observe multiphoton electron emission from ultrasharp metallic tips illuminated with 7-fs-light pulses. Local field enhancement confines this emission to the tip apex, demonstrating the potential of this source for ultrafast electron imaging with nanometer-resolution.

Femtosecond electron and X-ray diffraction are currently among the most intriguing topics in ultrafast science, allowing for probing structural dynamics of molecular and solid state systems with previously unachievable temporal resolution. Despite substantial recent progress, these experimental techniques are still in an early stage, and large efforts are currently put into the development of sophisticated femtosecond electron [1] or X-ray [2] sources suitable for experiments with high temporal resolution. In electron diffraction, overcoming temporal smearing due to spatial propagation effects and to Coulomb repulsion of electron bunches produced at kHz repetition rates is particularly challenging. Ultimately, therefore, a point-like source of single electrons with temporal resolution in the regime of few femtoseconds would be highly desirable.

In this paper, we describe and demonstrate a novel approach towards realizing such a point-like ultrafast electron source. By illuminating ultrasharp gold tips with 7-fs pulses from an 80 MHz Ti:sapphire oscillator, we induce emission of an intense flux of up to 10^7 electrons per second. Due to the local field enhancement this emission is strongly localized at the apex of the metallic tip with a radius of curvature of only few tens of nanometers. We demonstrate the multiphoton character of the electron generation in the absence of a tip bias voltage.

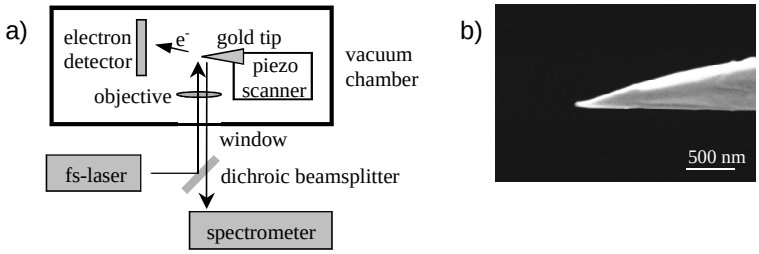

Fig. 1. (a) Schematic of the experiment. Electron emission and nonlinear light scattering are detected as a gold tip is scanned through a focused 7-fs laser in the plane perpendicular to the optical axis. (b) Electron microscope image of one of the tips.

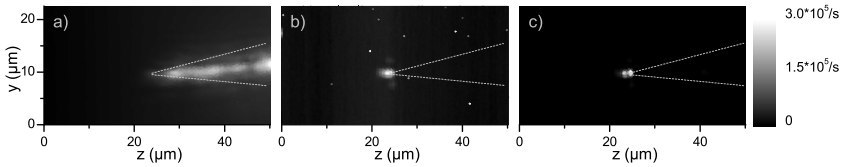

Fig. 1. (a) Spatial scan of the fundamental laser light backscattered from a sharp gold tip (indicated by the dotted lines). (b) Nonlinear light generation localized at the very end of the tip. (c) Simultaneously recorded electron emission (linear gray scale).

In the experiments, an electrochemically etched gold tip with a radius of curvature of about 20 nm (Fig. 1) is mounted on a piezo scanner inside a high-vacuum chamber. 7-fs-light-pulses from an 80MHz Ti:sapphire oscillator with a center wavelength of 800 nm are focused onto the tip with a Cassegrain mirror objective. The electrons emitted from the tip are detected with a micro-channel plate opposing the tip and are counted with an electronic discriminator. The linear and nonlinear scattered light is collected in a back reflection geometry, spectrally resolved and detected with a liquid-nitrogen-cooled CCD camera. Electron emission and spectrally resolved light scattering are recorded simultaneously while the tip is scanned through the laser focus.

Illuminating such a sharp metal tip with fs light pulses results in nonlinear light generation on the blue side of the laser spectrum [3,4]. Local field enhancement at the tip apex leads to a localization of the nonlinear light generation at the very end of the tip. This spatial localization is directly seen in Fig. 2, comparing images of the linearly scattered laser light (a) and the nonlinear emission (b) in the wavelength range between 400 and 600 nm. Here, the tip is scanned through the focus in the plane perpendicular to the optical axis. The nonlinear emission is composed of second harmonic radiation peaked around 400 nm and a broad background continuum between 450 and 700 nm. Its spot size, about 0.8 µm x 1.5 µm, is only given by the shape of the laser focus. Fig. 2(c) shows the simultaneously recorded electron signal. Strikingly, we find an intense emission of electrons generated at exactly the position as the nonlinear light generation. In the electron image, the spot size is reduced further to about 0.6 µm x 1.1 µm. Clearly, the same local field enhancement is responsible for both electron and nonlinear light emission.

To get first insight into the dynamics of the electron emission, a phase-stable pair of laser pulses was sent onto the tip, and the electron emission was recorded as a function of the pulse delay. The resulting interferometric autocorrelation (IAC) trace (Fig. 4a), has a FWHM of about 10 fs. For estimating a pulse duration from this measurement, knowledge about the underlying optical nonlinearity is needed. Recent results claim a one-photon assisted tunneling mechanism at high tip bias voltages close to 1 kV [1]. In contrast, the intensity-dependent measurement shown in Fig. 4b reveals a clear fourth-order dependence of the electron emission on the incident laser power in our experiments. This suggests emission from a short-lived non-thermal carrier distribution at energies above the 5.1 eV work function of gold. This transient distribution is induced by multiphoton absorption and ultrafast energy redistribution during the presence of

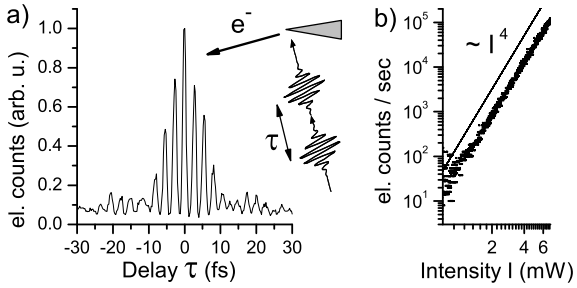

Fig. 3. (a) Electron signal as a function of relative delay between a pair of phase-locked laser pulses (interferometric autocorrelation). (b) Fourth-order intensity dependence of the electron emission from the tip

the pulse. Similar experiments for various bias voltages clearly support this interpretation. The IAC duration is about a factor of 1.5 longer than expected from an instantaneous fourth-order nonlinearity and the 6.7 ± 0.2 fs duration of our laser pulses.. Both temporal dispersion of the focused pulses and a resonantly enhanced nonlinearity may account for this slight broadening of the IAC trace. We find no IAC components beyond 30 fs delays that could evidence a storing of energy in the electronic system beyond that time. Therefore, it is not expected that the electron pulse duration close to the tip differs significantly from the optical pulse duration. It will be interesting to investigate how propagation towards a sample affects this time structure.

Comparing the emission rates from the tip apex with those from the tip shaft allows one to estimate the actual local electric field enhancement with respect to the incoming laser field: $\alpha = |E_{loc} / E_{inc}|$. The field enhancement factors from nonlinear light generation and electron emission are about α = 15 and 10, respectively. These values are in rather good agreement with theoretically predicted values ($\alpha \approx 12$) for gold tips with 10 nm radius [7]. This field enhancement and the high peak intensity of our 7-fs-pulses results in the high electron flux of about 10^7 electrons per second for zero tip bias voltage and at average laser power of only 15 mW.

In summary, we have demonstrated a novel nanometer-sized femtosecond electron source based on local field enhancement at ultrasharp metal tips. We believe that this new source carries potential for use in ultrafast electron microscopy and diffraction, as it avoids space charge effects previously encountered in experiments at lower repetition rates. It is expected that the electron yield can be increased substantially by tailoring the field enhancement at the tip by the use of, e.g., ion beam milling techniques.

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