Ultrafast field-driven monochromatic photoemission from carbon nanotubes

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***Abstract*—Ultrafast electron pulses, combined with laser-pump and electron-probe technologies, allow for various forms of ultrafast microscopy and spectroscopy to elucidate otherwise challenging to observe physical and chemical transitions. However, the pursuit of simultaneous ultimate spatial and temporal resolution has been largely subdued by the low monochromaticity of the electron pulses and their poor phase synchronization to the optical excitation pulses. State-of-the-art photon-driven sources have good monochromaticity but poor phase synchronization. In contrast, field-driven photoemission has much higher light phase synchronization, due to the intrinsic sub-cycle emission dynamics, but poor monochromaticity. Such sources suffer from larger electron energy spreads (3 - 100 eV) attributed to the relatively low field enhancement of the conventional metal tips which necessitates long pump wavelengths (> 800 nm) in order to gain sufficient ponderomotive potential to access the field-driven regime[4]. In this work, field-driven photoemission from ~1 nm radius carbon nanotubes excited by a femtosecond laser at a short wavelength of 410 nm has been realized. The energy spread of field-driven electrons is effectively compressed to 0.25 eV outperforming all conventional ultrafast electron sources. Our new nanotube-based ultrafast electron source opens exciting prospects for attosecond imaging and emerging light-wave electronics.**

Keywords—ultrafast photoemission; carbon nanotube; strong field; low energy spread

I. INTRODUCTION (*HEADING 1*)

Highly coherent electron pulses, pumped by ultra-fast lasers, allow for the unprecedented study of various ultra-fast phenomena (*1*). In contrast to its quantum photon-driven counterpart, classical field-driven photoemission holds great promise. Significant system-level functional improvements manifest through the exploitation of material-driven alternative emission mechanisms; optical tunneling coupled to strong optical field excitation may ultimately result in electron generation with pulse duration down to half an optical cycle (*2*). However, the relatively large dimension (~ 10 nm radius) of conventional metallic, Spindt-like nanostructures necessitates long pump wavelengths to access the field-driven photoemission regime (*3*). This significantly increases beam energy spread and temporal width (~ few femtoseconds) due to the phase dependent emission feature. As a result, the typical energy spread is, as reported elsewhere, several eV or higher, which substantially limits their practical applications.

Carbon nanotubes (CNTs) have attracted much interest due to their exceptional electrical, chemical, mechanical and optical properties. In particular, CNTs strongly couple to static electric fields due to their unique geometry, and show intriguing structural stability under such otherwise aggressive environments. Collectively, these properties enable challenging electron emission systems to be realized at the nanoscale, for the first time, such as; highly coherent field electron emission with low energy spread, small virtual source size and high brightness, all of which cannot be simultaneously achieved using traditional metallic structures. Nevertheless, CNT-based electron emission studies have reported to date, almost exclusively, on the use of static field. Static electric field-driven CNTs based electron sources operate as continuous wave devices, or have relatively long pulse durations (~ns), comparable to the pulse width of an electronic drive signal. This can be overcome by using ultrafast optical field, which potentially offers great opportunities to break through the bottleneck mentioned above and promote the performance of ultrafast electron sources.

Here, we report on a single wall CNTs (SWCNTs)-based ultra-fast electron source, in which photoemission is strongly governed by an optical tunneling effect driven via ultra-fast optical fields. The present source demonstrates high coherence in a largely monochromatic beam (<0.25 eV).

II. EXPERIMENTAL DETAILS

The structure of the SWCNT-based photoemission is schematically illustrated in Fig. 1A. SWCNTs were grown by

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chemical vapor deposition and had a nominal tube radius distribution between~ 0.5 and ~ 1 nm (Fig. 1B, C). A schematic depiction of the experimental electron emission setup is shown in Fig. 1D. Photoelectron emission from SWCNT arrays was triggered with 100 fs laser pulses, with a central wavelength of 410 nm, at an 80 MHz repetition rate from a Ti: Sapphire ultrafast laser. A standard Si photodiode power sensor was used to measure the laser power. The laser was normally incident on the SWCNT tip via front illumination, which was focused to a 1.25 μm (FWHM) spot at the SWCNT cluster apex. This photocathode samples were mounted in a high-vacuum chamber (10^{-7} Torr). The anode was adjacent to the photocathode some 400 μm distant using a thick mica insulating spacer, thus isolating the anode from the cathode. The anode, together with the insulating separator, was placed directly on the surface of the photocathode with the SWCNT arrays centrally aligned. A Keithley 2400 source measurement unit was used to bias the anode with voltages of up to 50 V, with the anode current measured. Unless otherwise stated, the current measurements presented in the main text are those recorded at the anode. Every current data, collected by source meter, was acquired from an arithmetic average of 100 repeated measurements.

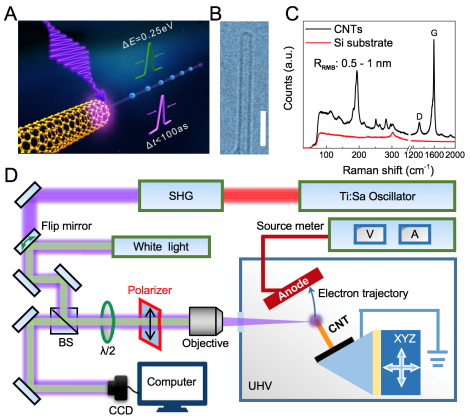

Fig. 1. Highly coherent SWCNT-based photoemission source.

III. Results and Discussion

Photoemission currents (I) were recorded as a function of incident laser power (P), as shown in Fig. 2A. We find a tip-exclusive signal at both 410 nm and 820 nm pump wavelengths. The results suggest two distinct operational advantages of our SWCNT device; a broad operation bandwidth and a highly nonlinear response. Note that the broadband operation of our SWCNT device highlights the strongly localized field enhancement capable of wide wavelength operation due to the extremely sharp tip geometry. The emission current at low pump power was proportional to the fourth power of laser power (Fig. 2A), suggesting above-threshold multiphoton photoemission, as the measured SWCNT work function (Φ) was ~ 4.4 eV. A deviation from the fourth power law was found when the pump power was > 3 mW. This presents strong evidence of classical field-driven photoemission. To further confirm this, the optical field-dependence of the emission current was analyzed by fitting with Fowler-Nordheim (FN) theory (Fig.2b). The calculated field enhancement factor was ~27. At an incident laser power of 3 mW (approximately 17.6 GV/m local optical field, F), Keldysh parameter $\gamma = \sim 1.85$. This further demonstrates that our photoemission operates in the strong optical field-driven regime, which is comparable to the Keldysh parameter of the previously reported field-driven photoemission sources pumped at longer wavelengths (*3*).

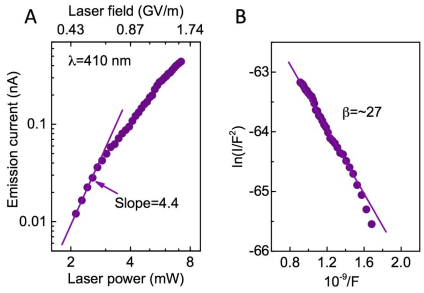

Fig. 2. Field-driven photoemission at 410 nm pump wavelength.

We also measured photoemission currents (I) as a function of anode bias voltage (V, Fig. 3A), in order to elucidate the underpinning dynamics of observed photoemission. The total energy spread of the generated photoemission can be determined directly from the differential spectrum of the transport characteristics. As depicted in Fig. 3B and Fig. 3C, the total energy spread for the present SWCNT-based electron source increases with pump power and wavelength. A very narrow energy spread of 0.25 eV was measured, approaching the spread suggested by our simulations (0.23 eV). We believe that the observed ultra-low energy spread allows for concurrently low beam divergence and narrow emitting energy level distribution.

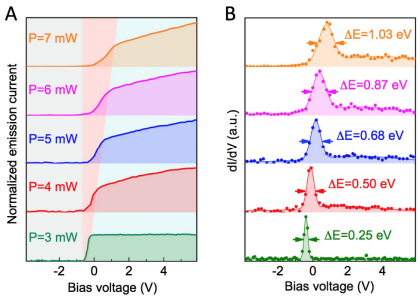

Fig. 3. Total energy spread. (A) Dependence of the normalized emission current on bias voltage (I-V) at various incident power. (B) dI/dV curves at different power, the width of the peaks (FWHM) indicates the total energy spreads.

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