Visualization of Photocurrents in Nanoobjects by Ultrafast Low-Energy Electron Point-Projection Imaging

M. Müller, A. Paarmann, and R. Ernstorfer
Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany
m.mueller@fhi-berlin.mpg.de

Abstract: Ultrafast dynamics of photocurrents in semiconductor nanowires are investigated with femtosecond time and nanometer spatial resolution. We demonstrate the capability of time-resolved low-energy electron point-projection imaging as a novel tool for mapping transient fields at nanostructures.
OCIS codes: (320.7130); (180.0180);

1. Introduction

Semiconductor nanowires (NWs) and, in particular, heterostructured NWs are promising candidates for future nanoscale electronic and optoelectronic devices, as well as ideal model systems for exploring fundamental semiconductor physics on nanometer length scales. Within the last years there has been vast progress in controlling the doping level in both radial and axial direction during NW growth [1]. In order to understand the physics of carrier transport in these structures, it is of major importance to study the dynamics of charge carriers upon photoexcitation. So far, typical studies such as time-resolved photoluminescence and photoemission electron microscopy provide either spatially- or time-averaged information, respectively. In this regard, it is most appealing to combine nanometer spatial with femtosecond temporal resolution to directly measure the spatio-temporal evolution of photoexcited charge distributions in such nanoobjects.

Here, we present the first femtosecond time- and nanometer-spatially resolved investigation of ultrafast photocurrents in heterostructured InP NWs, employing our newly developed technique of femtosecond low-energy electron point-projection imaging (fsPPI). We show that low-energy electrons are ideally suited to detect the transient fields generated by ultrafast photocurrents on nanometer dimensions. The high sensitivity of this novel approach to weak fields allows for studying the detailed spatially resolved dynamics in a wide range of low-dimensional systems, in particular plasmonic and semiconductor nanostructures.

2. Methods

A schematic of our experimental apparatus is shown in Fig. 1a. We use tungsten nanotips as point source for femtosecond low-energy electron probe pulses generated by an ultrashort laser pulse [2]. The tip is placed ~20 µm in front of the sample and a point-projection image is recorded on the imaging screen with a magnification of up to 10^4. Figure 1b shows projection images of gold NWs spanning across 2 µm holes in a thin carbon substrate in continuous field emission and laser-triggered photoemission mode, respectively. In the pulsed mode, the electron emission is confined to a time window <10 fs as evidenced by the autocorrelation revealing a 3^{rd} order process, see Fig. 1c. Time resolution of the imaging is achieved by varying the delay between the electron probe pulse and an optical pump pulse exciting the sample, see Fig. 1a.

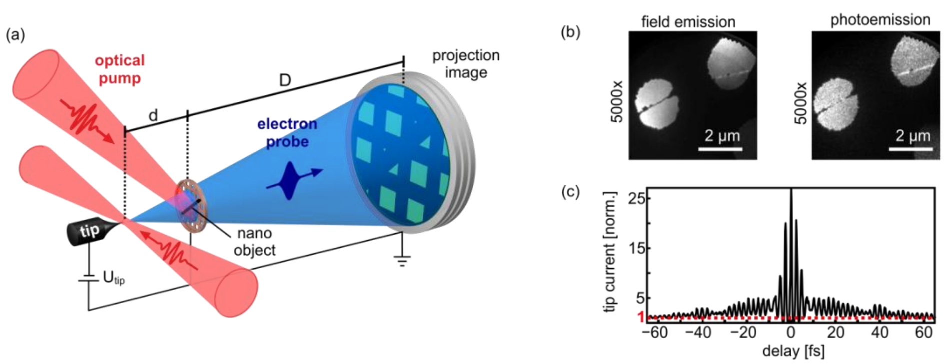

Fig 1: (a) Scheme for femtosecond point projection imaging. (b) Projection images of a 50 nm gold NW lying across 2 µm holes in a free-standing carbon thin film, for field emission and photoemission operation mode, respectively. (c) Interferometric autocorrelation of the nanotip current in the laser-triggered mode.

3. Results and Discussion

The fsPPI data of axially doped p-i-n InP NWs are presented in Fig. 2. The projection image in Fig. 2a shows the NW before the optical pump pulse arrives. The projected wire diameter appears bright and much larger than its real space diameter (30 nm) due to static lensing effects induced by local electric fields in the vicinity of the nanoobject, which depend critically on extrinsic parameters such as the influence of the tip field and intrinsic parameters like work function differences between the NW and the substrate. At temporal pump-probe overlap we observe a clear pump-induced change of the imaged wire profile, as shown in Fig. 2b. The intensity redistribution is most pronounced perpendicular to the wire, transiently changing the projected wire diameter. We also observe clear differences of these changes axially along the wire. The dynamics of these effects are analyzed in Fig. 3c, where we plot the wire diameter as a function of time delay. At both axial positions of the NW the signal shows a rapid initial drop with a ~200 fs time constant, followed by the relaxation of the effect on a ~1 ps time scale. While the time scales in both regions of the NW are similar, the magnitude is much larger in the top part, red curves in Fig. 2c.

We interpret the observations being a result of one-photon above-bandgap absorption from the 800 nm pump pulse and consecutive radial separation of the photo-generated charge carriers due to band bending at the semiconductor surface. The radial photocurrent results in a transient reduction of the surface band bending. This is observable through imaging of the transient electric fields outside the NW modifying the electron lensing effect of the NW. The axial differences are likely due to the p-i-n structure of the NW. The linear fluence dependence of the transient diameter changes supports the picture of one-photon absorption from the pump pulse. We also observe a slight axial redistribution of intensity indicating axial photocurrents induced by the doping contrast. The analysis and interpretation of these dynamics are currently in progress. Noteworthy, the initial rise time of the effect is ~200 fs. Simulations studies [3] predict electron pulses with durations <100 fs for fsPPI, suggesting the initial dynamics are not limited by the instrument resolution but instead due to the dynamics of the photo-excited charge carriers.

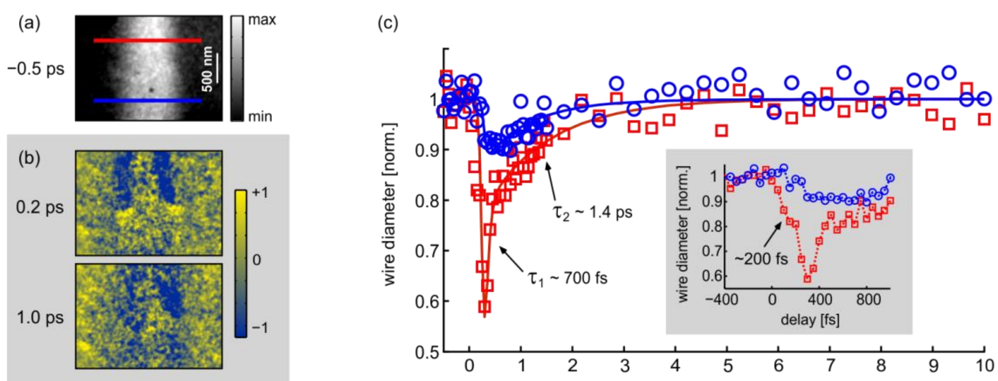

Fig 2: (a) Point-projection image of an InP NW at negative delay times between pump and probe. (b) Transient pump-induced image differences at two selected delay points. (c) Dynamics of the wire diameter in the projection image in two regions of the nanowire indicated with the colored lines in (a). The inset shows the ~200 fs initial dynamics of the diameter reduction.

4. Summary

Femtosecond low-energy electron point-projection investigations of axially doped InP nanowires are presented. We observe transient modifications of the projection images with a rise time of ~200 fs and decay times of ~1ps, which we interpret being the result of axial separation of photo-carriers due to surface band bending, inducing transient electric fields around the wire. Our data represents the to our knowledge the first real-space observations of femtosecond photocurrents in a nanoobject with nm spatial resolution making it very attractive for investigations of ultrafast dynamics in nanoscopic systems such as plasmonic nanostructures.

5. References

[1] M. Hjort, J. Wallentin, R. Timm, A.A. Zakharov, U. Håkanson, J.N.Andersen, E. Lundgren, L. Samuelson, M.T. Borgström, and A. Mikkelsen, "Surface Chemistry, Structure and Electronic Properties from Microns to the Atomic Scale of Axially Doped Semiconductor Nanowires", ACS Nano **6**, 9679 (2012)

[2] C. Ropers, D.R. Solli, C.P. Schulz, C. Lienau, and T. Elsaesser, „Localized Multiphoton Emission of Femtosecond Electron Pulses from Metal Nanotips", Phys. Rev. Lett. **98**, 043907 (2007)

[3] A. Paarmann, M. Gulde, M. Müller, S. Schäfer, S. Schweda, M. Maiti, C. Xu, T. Hohage, F. Schenk, C. Ropers, and R. Ernstorfer, „Coherent Femtosecond Low-Energy Single-Electron Pulses for Time-Resolved Diffraction and Imaging: A Numerical Study" ,J. Appl. Phys. **112**, 113109 (2012).