Spectrally resolved x-ray beam induced current in a single InGaP nanowire

Lert Chayanun, Vilgailė Dagytė, Andrea Troian, Damien Salomon, Magnus Borgström and Jesper Wallentin

1 Synchrotron Radiation Research and NanoLund, Lund University, Lund, Sweden
2 Solid State Physics and NanoLund, Lund University, Lund, Sweden
3 European Synchrotron Radiation Facility, Grenoble, France

E-mail: lert.chayanun@sljus.lu.se

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Abstract
We demonstrate x-ray absorption fine structure spectroscopy (XAFS) detected by x-ray beam induced current (XBIC) in single n-i-n+ doped nanowire devices. Spatial scans with the 65 nm diameter beam show a peak of the XBIC signal in the middle segment of the nanowire. The XBIC and the x-ray fluorescence signals were detected simultaneously as a function of the excitation energy near the Ga K absorption edge at 10.37 keV. The spectra show similar oscillations around the edge, which shows that the XBIC is limited by the primary absorption. Our results reveal the feasibility of the XBIC detection mode for the XAFS investigation in nanostructured devices.

Supplementary material for this article is available online

Keywords: x-ray beam induced current (XBIC), x-ray absorption fine structure spectroscopy (XAFS), nanowire

(Some figures may appear in colour only in the online journal)

1. Introduction

X-ray absorption fine structure (XAFS) is an established method for investigating semiconductors, which can give information about the local atomic properties [1–3]. Recent developments in x-ray optics has made it possible to investigate single nanostructures, whose properties may differ substantially from bulk material [4, 5]. The signal from single nanostructures is inherently weak, which makes detection challenging. For instance, traditional transmission measurements suffer from poor contrast due to the weak absorption.

One alternative is to measure the electrical conductance, which gives a weak signal but also a low background for single nanostructures [6, 7]. The x-ray beam induced current (XBIC) is a more complex process than absorption and x-ray fluorescence (XRF), since the measured signal depends on local carrier transport properties in semiconductors. This makes it possible to use x-ray beams as a local probe, similar to electron beam induced current and scanning photocurrent microscopy [8, 9]. In particular, recent studies have demonstrated that x-ray beams can probe the interior of single nanowire (NW) devices [6, 7]. Moreover, NWs have shown a strong electrical response under hard x-rays [7]. The advantage of the x-ray beams over the electron and the laser probe beam is a longer penetration depth and a smaller diffraction limit, respectively [10–12]. Thus, nanofocusing of x-rays that reach the sub-10 nm regime [10, 11] could significantly enhance the spatial resolution of XBIC.

The process of generating charge carriers from x-rays starts with an absorption of a primary x-ray photon which excites an inner core electron that results in a core hole and a photoelectron. The absorption probability, $p_{abs}$, depends on the photon energy, the sample composition, as well as the geometry of the beam and the sample. Near an absorption edge, the absorption probability of the atoms will also depend...
on the local atomic environment. The relaxation of an electron from a higher state to the core hole releases an excessive energy in the form of a secondary photon or an electron through the processes called XRF and Auger electron emissions, respectively. Further electrons are excited by these secondary photons and electrons [6], at the same time as the electron–hole pairs thermalize to the band edges. The average number of carriers generated through the primary x-ray photon absorption is given by $\eta = E/\epsilon$, where $E$ is the photon energy, and $\epsilon$ is the ionization energy [13]. For the x-ray energy of 10.37 keV and the sample thickness of 180 nm used in this study, we have $\eta = 1867$ and $p_{abs} = 9.7 \times 10^{-3}$, for bulk In$_{0.56}$Ga$_{0.44}$P. This is in contrast to visible light for which only a single electron–hole pair is created per single photon event, $\eta = 1$.

The non-equilibrium charges will generate a current if there is an electric field, which can be internal, as in p–n junctions, or externally applied as in this work. The charge carriers can also recombine or get trapped in long-lived surface states before being detected, and the signal therefore depends on local carrier lifetimes and mobilities. Thus, XBIC can be used to investigate the local carrier transport properties. In addition, the signal from the studied sample might be affected by the x-ray interaction with the nearby components, such as the substrate, and the metal contacts. Note that the method here is distinct from the detection of Auger electrons, often called electron yield [1].

Evidently, the generation of the XBIC signal is more complex than the XRF process. Here, the XBIC and the XRF signals are compared with spectrally resolved excitation, to attain the XAFS spectrum from single nanowire devices. We find that the spectra are qualitatively similar, despite the underlying differences.

2. Methods

The sample was In$_{0.56}$Ga$_{0.44}$P single NWs with axial $n^+\cdot i\cdot n^+$ doping profiles which were grown via the vapor–liquid–solid method by the use of Au seed particles in a metal organic vapor phase epitaxy system (supplementary material is available online at stacks.iop.org/NANO/29/454001/mmedia). The NWs also had an InP nucleation segment at the base and a GaP segment right below the Au particle. The NW diameter was 175 nm. The nominal length of each section was monitored by LayTec EpiR DA UV optical reflectometry system [14] which gives the lengths of InP nucleation segment 230 nm, $n^+\cdot$InGaP 290 nm, $i\cdot$InGaP 1200 nm, $n^-\cdot$InGaP 400 nm, and GaP 120 nm. The NWs were transferred to a SiO$_2$ coated Si substrate with predefined bond pads and alignment markers. Electrical contacts were made to single NWs with electron beam lithography and metal evaporation of Ti and Au (10/230 nm). The NWs were excited with the nanofocused x-ray beam (∼65 nm diameter) at beamline ID-16B at the European synchrotron radiation facility, Grenoble, France (figure 1(a)) [15]. The XRF as well as the XBIC signals were collected over the NW at a bias of 0.05 V (figure 1(a)). For the spectrally resolved XRF and conductance measurement, the x-ray photon energy was scanned around the Ga K-edge energy (∼10.37 keV) at the position of the NW where we attained the highest photocurrent signal.

3. Results and discussions

3.1. Spatially resolved XBIC and XRF

The $I$–$V$ characteristics of the device in dark and under x-ray excitation at the flux $\Phi = 1.6 \times 10^7$ s$^{-1}$ is illustrated in figure 1(b). In both cases there is a linear relation between the current and the applied bias. Furthermore, the fluctuating signal manifests the noise of a few fA. The linear current–voltage relation makes it possible to calculate an electrical conductance which represents the XBIC signal in this report. The electrical conductance is about two orders of magnitude higher under x-ray excitation ($\sigma = 1.3 \times 10^{-12}$ $\Omega^{-1}$) compared with the dark conductance ($\sigma = 3.6 \times 10^{-14}$ $\Omega^{-1}$).

The superpositioned image of the conductance and XRF signals in figure 1(c) was gathered by a two-dimensional scan over the NW with a step size of 50 nm and a collection time of 0.2 s per point. The conductance is shown as green in this image (figure 1(c)). The XRF signal was collected as a spectrum at each position from which the intensities at the certain energies, corresponding to the emission from Au and Ga, were extracted (supplementary material). They are displayed in figure 1(c) as blue and red areas for the Au metal contacts and the NW, respectively.

Lineplots of the signals in figure 1(c) along the center of the NW are shown in figure 1(d). The strong XRF signal from
Ga atoms on the left (figures 1(c) and (d)) indicates the GaP segment near the top of the NW which is used as the reference position \((x = 0)\). Then, we could draw the dashed lines indicating the nominal segments of the NW in figures 1(c) and (d). Apparently, most of the XBIC signal (green area) is collected from the middle segment. We observed no significant conductance peak at the contact edge, which would indicate a Schottky like contact between the NW and the metal contacts as previously reported [16].

The conductance profile in figure 1(d) shows an exponential decay on both sides, which could be fitted by \(G(x) \propto \exp(-x/L)\), where \(L\) is a characteristic decay length [17]. The decay lengths are \(L_l = 203 \pm 19\) nm and \(L_r = 514 \pm 20\) nm for the left and right slopes, respectively. Comparing these lengths to the beam diameter (~65 nm), the decay is not limited by the size of the probe beam, so the characteristic decay length revealed here could be used to investigate the local carrier transport properties [16, 18, 19].

The XBIC peak is related to the \(n\)-\(i\)-\(n\) doping profile of the nanowire. In the highly doped \(n\)-segments, the electric field is too weak to drive the charge carriers to the contacts before they recombine. Instead the electric potential falls almost entirely over the middle segment, which gives a strong electric field that efficiently moves the charge carriers.

### 3.2. X-ray photon flux variation XBIC

Next, the flux dependence of the NW response to the x-rays was investigated. The maximum flux used was \(\Phi = 1.6 \times 10^8\) s\(^{-1}\) which gives the conductance as red trace in figure 2(a). By reducing the flux by half to \(\Phi = 0.8 \times 10^8\) s\(^{-1}\), the conductance was decreased almost 3 times (figure 2(a)). Further decreasing flux generated a weak signal which could not be detected. A super linear relation was already observed in a similar experiment [7], which was attributed to the charge carrier trapping at the NW surface leading to photogating and photodoping effects [20]. In photogating, trapped charge carriers behave like a wrap-gate on the NW, changing the Fermi level. Then, trapped charges would induce an excessive amount of the opposite charge carrier in the center of the NW called photodoping.

An expected linear relation between the general photocconductance and the photon flux is \(G = q \eta p_{abs}(\mu l/\Phi)\Phi\) where \(q\) is the elementary charge, \(\mu\) is the carrier mobility, \(l\) is the length of the active region, and \(\Phi\) is the incident photon flux. With the long-lived traps, the photocconductance after switching the beam on becomes time dependent which can be written as:

\[
G(t) = q \eta p_{abs} p_{trap}(\mu l/\Phi)(1 - \exp(-t/\tau_d))\Phi, \tag{1}
\]

where \(t\) is time, \(p_{trap}\) is the trapping probability, and \(\tau_d\) is the detrapping lifetime. The conductance of this InGaP NW device is assumed to be dominated by electrons with mobility \(\mu = 200\) cm\(^2\) V\(^{-1}\) s\(^{-1}\) [21].

To further understand the trapping mechanism, we performed time-resolved measurements where the conductance was measured as a function of time after switching the x-ray beam on (figure 2(b)). At small \(t\), the rate of the photoconductance is constant, \(dG/dt = q \eta p_{abs} p_{trap}(\mu l/\Phi)\Phi\). giving the calculated trapping probability, \(p_{trap} \propto 1.45 \times 10^{-10}\), from the slope of the plot. Then, we fitted the time resolved photoconductance with equation (1) (dashed line in figure 2(b)) which yields the detrapping lifetime of \(\tau_d = 3.41\) s. In a similar experiment performed on a 100 nm diameter InP NW [7], the trapping probability was much higher, \(p_{trap} = 2.3 \times 10^{-6}\). We speculate that the lower trapping probability is due to the larger NW diameter here, 175 nm.

Furthermore, the device performance can be quantified by the photoconductive gain, \(g\), which is defined as the ratio between the conductance from the collected charge carriers and the absorbed charge carriers [7, 20]. By dividing equation (1) with the carrier absorption term, \(q \eta p_{abs} \Phi\), and setting \(t \to \infty\), the photoconductive gain can be written as \(g = p_{trap}(\mu l/\Phi)\Phi\), which yields \(g = 0.34\) for our device. The low gain is due to the low trapping probability.

### 3.3. Spectrally resolved XBIC and XRF

The ability to detect XBIC in a single NW opens up the possibility to use it as a new detection mode for XAFS investigations which are conventionally measured by the XRF or the absorption techniques. The weak signal from those techniques is a general problem for spectroscopic investigations of single nanostructures. Besides, the comparison of the

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\text{Figure 2. Flux and time dependence of XBIC (a) spatial photoconductance of the NW device at two different x-ray photon fluxes, } \Phi = 0.8 \times 10^8\text{ s}^{-1}\text{ (blue line) and } \Phi = 1.6 \times 10^8\text{ s}^{-1}\text{ (red line). (b) Time resolved photoconductance (blue squares) measured at } \Phi = 1.6 \times 10^8\text{ s}^{-1}. \text{ The solid red line is the linear fit for the rate of the photoconductance at short time and the dashed red line is the fit by the use of equation (1).}
\]
The XBIC at the atomic level. Energy could shade the light on the underlying mechanism of XAFS from the XBIC and the XRF signals at the Ga K-edge.

**Figure 3.** Spectrally resolved photoconductance and XRF (a) schematic of the interference process between the emitted photoelectron wave (blue circles) and backscattering wave from the neighboring atoms (red circles) within the lattice. (b) Plot of the spectrally resolved conductance (black) and XRF (blue). (c) Close up plot of (b) about the Ga K-edge peak. The interesting peaks of the spectrum are labeled as A, B and C.

XAFS from the XBIC and the XRF signals at the Ga K-edge energy could shed light on the underlying mechanism of the XBIC at the atomic level. Each result of the spectrally resolved measurements (supplementary material) shows fluctuations and non-reproducible spikes owing to the sensitivity of the equipment. Those results are averaged and shown in figure 3, with the conductance (black trace) and XRF (blue trace) spectra. Overall, the oscillation of the conductance follows the XRF signal, although there are some differences. In both spectra, we observed a rapidly increasing XRF signal at the Ga K-edge energy (figure 3(b)). From this result, we can trace the photoconductance signal back to the interaction between the x-ray and the Ga atoms. However, the pre-edge signal in the conductance measurement is not at the minimum level as detected in the post-edge region. The reason is that charge carriers can be generated also by x-ray absorption in In and P atoms. In contrast, the Ga K XRF signal can only result from excitation in the Ga K shell.

The signals show post-edge oscillations that are typically known as the extended XAFS, which is part of the XAFS spectrum. The oscillations result from the interference between the emitted photoelectrons from the target atoms and their backscattering waves from neighboring atoms (figure 3(a)). The change in energy of the electrons, which is coupled to their wavelength, leads to a variation in constructive and destructive interference. Consequently, the interference affects the absorption probability of the target atoms leading to the oscillations in the detected conductance and XRF.

The magnified spectra near the edge (figure 3(c)) reveal two consecutive peaks at the edge energy, labeled as peak A and B, in both the XRF and conductance plots. The peaks A and B are at 10.374 keV and 10.378 keV, respectively. The first distinct peak, C, in the post-edge region is at 10.397 keV. Although the XAFS results could reveal many atomic features of the sample, a quantitative interpretation of the XAFS results is beyond the scope of this report. We only qualitatively compare our result with the other relevant studies.

For peaks A and B, the XAFS from different Ga composition materials; e.g. GaAs and Ga2O3 [3, 22], shown a similar feature at the Ga K-edge energy. Following these studies, we interpret peak A and B as the transitions of the 1s electron to 4p and to the continuum, respectively. In the post-edge region where we observed peak C, similar results were found from the measurement of InGaN [23] and GaN [24]. The energy difference between Peak B and C could be related to the distance from Ga atoms to their neighboring atoms, which is related to the crystal structure. In our case, where the energy gap between peak B and C is about 17 eV, the results suggest that the sample has zinc blende structure [24]. The same result was also achieved from a transmission electron microscopy (TEM) study of NWs grown under similar conditions [25].

Similar measurements performed on the heterostructure p–n junction NW device exhibited a slightly inconsistency between these two signals, since the secondary electrons were affected by the heterostructure NW and the existence of the depletion region at the junction [6]. Due to the lack of the built-in electric field within the n⁺–i–n⁺ doped NW, we did not observe such an effect on the measured XBIC.

**4. Conclusion**

In conclusion, our results from the spectrally resolved XRF and XBIC demonstrate the feasibility to use electrical detection for XAFS measurements on the nanostructure devices. The two detection modes exhibit similar spectra. We observed a super linearly increasing XBIC signal as function of the x-ray photon flux, which is caused by photogating and photodoping effect due to the surface trapping. This technique could be used to study the local atomic environment and carrier transport properties of many categories of nanostructured electronic devices [26, 27].
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ORCID iDs

Lert Chayanun @ https://orcid.org/0000-0002-6195-1657
Vilgailė Dagiytė @ https://orcid.org/0000-0002-1740-8737
Andrea Troian @ https://orcid.org/0000-0003-1558-9228
Damien Salomon @ https://orcid.org/0000-0001-8334-7832
Magnus Borgström @ https://orcid.org/0000-0001-8061-0746
Jesper Wallentin @ https://orcid.org/0000-0001-5909-0483

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