Electron dynamics in In$_x$Ga$_{1-x}$As shells around GaAs nanowires probed by terahertz spectroscopy

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Abstract

We present the electrical properties of GaAs/In$_x$Ga$_{1-x}$As core/shell nanowires (NWs) measured by ultrafast optical pump–terahertz probe spectroscopy. This contactless technique was used to measure the photoconductivity of NWs with shell compositions of $x = 0.20, 0.30$ and $0.44$. The results were fitted with the model of localized surface plasmon in a cylinder in order to obtain electron mobilities, concentrations and lifetimes in the In$_x$Ga$_{1-x}$As NW shells. The estimated lifetimes are about 80–100 ps and the electron mobility reaches 3700 cm$^2$ V$^{-1}$ s$^{-1}$ at room temperature. This makes GaAs/InGaAs NWs good candidates for the realization of high-electron-mobility transistors, which can also be monolithically integrated in Si-CMOS circuits.

Keywords: GaAs nanowires, terahertz spectroscopy, ultrafast dynamics, electron mobility, plasmon, carrier lifetime

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

1. Introduction

Thanks to their direct band gap, III–V semiconductors are excellent materials for optoelectronic and photovoltaic devices. The III–V semiconductor material of our interest is an alloy of GaAs and InAs called In$_x$Ga$_{1-x}$As. It has a composition-dependent band gap which can fit in the telecommunications wavelengths (1.31 and 1.55 μm) and a lower effective electron mass compared to GaAs [1]. GaAs/InGaAs heterostructures are particularly interesting because of their applications, such as high-electron-mobility transistors (HEMT), light emitting diodes, lasers, light detectors and solar cells [2]. However, due to the large lattice mismatch between In$_x$Ga$_{1-x}$As and the available substrates (typically GaAs or InP), not all choices of $x$ are feasible without forming dislocations [3]. For the same reason, it is impossible to integrate monolithically InGaAs-based heterostructures in Si-CMOS platform. A promising solution could be the use of GaAs/In$_x$Ga$_{1-x}$As core/shell nanowires (NWs) grown epitaxially on Si substrates. They represent a special case of nearly perfect epitaxial heterostructures due to the very small Si/GaAs interface area and due to the high surface-to-volume ratio, which loosens the lattice-mismatch restrictions.

Pump-probe terahertz spectroscopy is a perfect tool for probing electrical properties of semiconductor NWs (and other nanostructures as well) in a contactless way [4]. Scattering rates of charge carriers in semiconductors, as well as their plasmonic resonances for typical doping, are located in the terahertz (THz) band. So far, this technique has been successfully used to extract photoconductivity spectra of other types of NWs like GaAs, InAs and InP NWs [5], or
core/shell GaAs/AlGaAs NWs [6]. The spectra were analyzed using the localized surface plasmon (LSP) model and the estimations of mobility, concentration and lifetime of charge carriers in the NWs have been obtained. In our paper we apply this method to study the electrical properties of GaAs/InGaAs NWs. Since InGaAs has a smaller band gap than GaAs, the charge transport mainly occurs in the shell. This fact distinguishes GaAs/InGaAs NWs from other core/shell NWs previously studied by pump-probe terahertz spectroscopy, where only the core was photoexcited.

2. Methods

2.1. GaAs/InxGa1-xAs core/shell NWs

The GaAs/InxGa1-xAs NWs were grown on Si(111) substrates by molecular beam epitaxy [7]. First the GaAs cores of 25 nm diameter were grown axially in self-catalyzed vapor–liquid–solid mode. The core growth was performed at 615 °C, ending with 2 μm long NWs. Then the 80 nm thick InxGa1-xAs shells were grown conformally around the GaAs cores, at the lower temperature of 367 °C, in the vapor–solid mode. Figure 1 shows a scanning electron microscopy (SEM) image of the GaAs/InxGa0.8 As core/shell NWs on Si substrate. By the analysis of multiple SEM images we determine the relative standard deviations of the NW length and radius to be 2.4% and 2.9%, respectively. In this study we focused on three types of NWs with different compositions of In in the shell: x = 0.20, x = 0.30 and x = 0.44. Both the core and the shell were nominally undoped.

The GaAs core and the InxGa1-xAs shell are significantly lattice-mismatched for higher values of x, which results in large strain. In [7] it has been demonstrated that the thin GaAs core is tensile-strained while the InxGa1-xAs shell is almost strain-free. The higher the indium content in the shell, the larger the tensile strain in the core. The band gap of unstrained bulk GaAs is 1.424 eV at 300 K and it decreases with increasing the tensile strain, i.e. the x in the shell. Nevertheless, our calculations (self-consistent Schrödinger–Poisson using the ‘nextnano’ software) demonstrate that the band gap of the strained GaAs core is always larger than that of the relaxed InxGa1-xAs shell. For example, the calculated band diagram at the Γ-point of the Brillouin zone for x = 0.20 is plotted in figure 2(a). Our results are similar to earlier simulations of GaAs/InAs core/shell NWs [8] which showed that, for thick shells, the conduction band edge of the InAs shell has a negative offset with respect to the GaAs core, while the valence band is rather flat. In agreement with the calculated band gaps, photoluminescence (PL) measurement on an ensemble of GaAs/In0.2Ga0.8As NWs shows a peak at 1.09 eV, which can be attributed to electron-hole recombination in the shell (figure 2(b)). After the growth, NWs of each type were transferred from the silicon substrate to separate z-cut quartz substrates in order to perform terahertz measurements. The area fill fraction \( f_{\text{fill}} \), or simply the fill factor, of the NWs was estimated for each sample from the images taken by optical microscope in dark field mode (figure 3). First, using the image processing software ‘ImageJ’, the fraction \( f_{\text{a}} \) of the image covered with NWs was extracted. But, due to the diffraction limit of the optical microscope, NWs on the image appear thicker than

![Figure 1. Side-view SEM image of as-grown GaAs/In0.2Ga0.8 As core/shell NWs on Si(111) substrate.](image1)

![Figure 2. (a) The simulated band diagram for GaAs/In0.23Ga0.77 As core/shell NWs showing the conduction band minimum (CBM) and the valence band maximum (VBM). The modeling includes the effect of strain. \( E_s \) and \( E_v \) are band gaps of the NW core and shell, respectively. (b) PL spectrum of GaAs/In0.3Ga0.8As NWs excited by a laser with 532 nm wavelength at the temperature of 300 K.](image2)
they actually are. Then the real fill factor of NWs was calculated using the formula \( f_{\text{fill}} = f_u \cdot d/d_w \), where \( d_w \) is the apparent NW thickness and the real one \( d \). Furthermore, since the NWs are randomly oriented (figure 3), only some of them are oriented predominantly in the direction of the THz probe polarization and, therefore, contribute to the plasmonic response (this will be discussed further in section 2.3). Therefore, the effective fill factor \( f_w \) was finally obtained from \( f_w = x_{\text{fill}} \cdot f_{\text{fill}} \), where \( x_{\text{fill}} \) stands for the fraction of the NWs which are effectively probed.

### 2.2. Optical pump—terahertz probe (OPTP) spectroscopy

We utilize OPTP spectroscopy to characterize the charge transport in the nominally undoped NWs. Near-optical pump pulses with wavelength of 800 nm (1.55 eV) and duration of approximately 60 fs are generated by a Ti:sapphire femtosecond laser amplifier and used to photoexcite electron-hole plasma in NWs. THz pulses, covering the frequency range between 0.1 and 2.7 THz, are generated by a large area photococonductive antenna [9] and detected, after being transmitted through the sample, by means of electro-optical sampling in a ZnTe crystal. Plasma oscillations of charge carriers in NWs are probed by the temporally delayed THz pulses, and their signature can be observed in the photoconductivity spectra.

In the experiment we record simultaneously the THz waveforms of the transmitted THz field without photoexcitation \( E(t) \) and its change due to the photoexcitation \( \Delta E(t) \). The photoconductivity spectrum \( \Delta \sigma(\omega) \) of NWs can be extracted from the Fourier transformed spectra \( \tilde{E}(\omega) \) and \( \Delta E(\omega) \). It is directly proportional to the relative decrease of the transmitted field \( -\Delta E(\omega)/E(\omega) \) and inversely proportional to the effective fill factor \( f_w \). By repeating the outlined procedure for different pump-probe delays \( \tau \) we extract time-dependent photoconductivity spectra \( \Delta \sigma(\omega, \tau) \) and get insight in the relaxation dynamics of the photoexcited charge carriers in the NWs. More details about the data analysis can be found in [10].

### 2.3. Photoconductivity spectra of NWs

Conductivity of bulk metals or semiconductors containing free charge carriers can often be modeled with the classical Drude model. According to this model, the resonant frequency of free charge carriers, called plasma frequency, is given by

\[
\omega_p = \sqrt{\frac{ne^2}{\varepsilon_0 \varepsilon_m m^*}},
\]

where \( n \) is the concentration of the charge carriers, \( m^* \) their effective mass, \( e \) elementary charge, \( \varepsilon_0 \) vacuum permittivity and \( \varepsilon_m \) relative permittivity of the material. Since the bulk plasma oscillations are longitudinal, they cannot be directly excited by transverse electromagnetic waves. But, in the case of particles which are smaller than the wavelength of the external field, conductivity spectra have different, Lorentzian-like shape, and simple Drude model cannot be applied anymore [11]. Instead, a more appropriate model of LSP should be used.

In an external electric field the electrons and holes start to move in opposite directions but, unlike in the case of bulk material, they will be confined and have to stay at the boundaries of the nanoparticle. This results in a depolarization field and a restoring force giving rise to the LSP resonance at the frequency

\[
\omega_0 = \sqrt{\frac{gne^2}{\varepsilon_0 \varepsilon_m m^*}} = \sqrt{\gamma} \omega_p,
\]

where \( g \) is a geometrical factor which depends on the nanoparticle material, its geometry and the surrounding dielectric medium and can vary between 0 and 1. In contrast to a bulk plasmon, the LSP couples to transverse EM waves and, therefore, can be detected as an absorption peak in an OPTP experiment. The photoconductivity of the charge carriers is given by [11]:

\[
\Delta \sigma(\omega) = \frac{ne^2}{m^*} \frac{\gamma}{(\omega_p^2 - \omega^2 + i\gamma \omega)},
\]

where \( \gamma \) is a scattering rate of the charge carriers. The holes in photoexcited GaAs or InGaAs have much larger effective mass than the electrons and their contribution to the LSP can be neglected. Therefore, we consider only the photoconductivity of the electrons in the following analysis.

In order to find out the geometrical factor \( g \), the NWs were approximated as In\(_{0.45}\)Ga\(_{0.55}\)As cylinders. The assumption comes from the fact that, although both core and shell are photoexcited by the pump, plasmonic response of the electrons comes predominantly from the shell since its volume is much larger compared to the core. Furthermore, after the photoexcitation the carriers stay in the shell since, for every studied composition, the corresponding strained core still has a slightly larger band gap (section 2.1). This was also shown in the scanning near-field infrared microscopy study of nonlinear plasmonic response in highly-doped GaAs/In\(_{0.45}\)Ga\(_{0.55}\)As core/shell NWs, where the electrons from the shell were probed [12].

The analytical solutions for the dispersion relations of the LSP modes in cylinder were given in [13], and also quantized along the NW axis by taking into account the finite length \( L \) of the NWs [14]. Since OPTP spectroscopy probes NWs in the far-field regime and the NWs are much smaller than the...
THz wavelength ($L \ll \lambda$), only the lowest-order modes are expected to be excited. In this case the dispersion relation is:

$$\omega_0(m, qR) = \omega_p \sqrt{\varepsilon_m - \varepsilon_{en}/\alpha_m(qR)},$$

(3)

where $\varepsilon_m$ and $\varepsilon_{en}$ are static relative permittivities of the NW shell and surrounding medium, $R$ is the radius the NW, $q$ is the longitudinal component of the plasmon wavevectors and $m$ is the order of a transverse mode. $\alpha_m(x) = J_m(x)K_n(x)/K_m(x)J_n(x)$ (where $J_n(x)$ and $K_n(x)$ are modified Bessel functions).

The lowest-order longitudinal LSP mode corresponds to $m = 0$ and $q = \pi/L$, whereas the lowest transverse LSP mode corresponds to $m = 1$ and $q = 0$. Figure 4(a) shows the LSP dispersions calculated using equation (3) for the case of 20% In. We use $\varepsilon_{en} = 1$ and the static relative permittivity $\varepsilon_m = 13.23$ for In$_{0.2}$Ga$_{0.8}$As [1] since the frequency range of the THz probe lies below TO phonons for all considered values of $x$ [7]. Since $qR$ is proportional to the NW radius-to-length ratio $R/L$, one can see from figure 4(a) that the thinner and/or longer NWs will have lower longitudinal LSP frequencies (blue curve). On the other hand, the perpendicular LSP resonance (green curve) is almost constant and, in the case of InGaAs NWs, close to the bulk plasma frequency $\omega_p$.

By taking the square of equation (3) one gets the geometrical factor $g = \omega_0^2/\omega_p^2$ as a function of $R/L$. The dependence $g(R/L)$ for the mentioned longitudinal mode ($m = 0$) in the cylindrical In$_{0.2}$Ga$_{0.8}$As NW is depicted by the blue curve in the figure 4(b). From there, by taking into the account the aspect ratio of our NWs, for the lowest-order longitudinal and transverse modes the geometrical factors were found to be $g(m = 0, q = \frac{\pi}{L}) = 0.228$ and $g(m = 1, q = 0) = 0.93$, respectively.

In the study by Joyce et al [10] cylindrical NW geometry was approximated with a prolate ellipsoid. The argument for the approximation came from the comparison of the numerically calculated dielectric polarizabilities for the two geometries [15]. For the sake of comparison we calculated the dependence $g(R/L)$ for the ellipsoidal In$_{0.2}$Ga$_{0.8}$As NW using the analytical expression from [10] and plotted it in the figure 4(b) (red line) along with the previously mentioned one (equation (3)) for the cylinder (blue line). It can be seen that this approximation holds true for radius-to-length ratios much smaller than one. However, for the higher values of $R/L$ the appropriate model should be used. In fact, the study [15] made comparison of the dielectric polarizabilities for the two geometries only for positive values of the NW dielectric permittivity (or, using the notation from the paper, positive values of the permittivity contrast $\varepsilon_i/\varepsilon_a$), which, in the case of frequency dependent polarizations, corresponds to the frequencies far above the LSP resonance. Therefore, in order to strictly compare LSP frequencies of the cylindrical and elliptical NWs, one should compare their polarizabilities for a sufficiently large range of negative permittivities as well (i.e. also negative permittivity contrasts).

As already discussed, only the lowest-order longitudinal $g(m = 0)$ and transverse $g(m = 1)$ modes are expected to be excited by the THz probe. But, although the NWs in all of our samples are randomly oriented, only one resonance was observed in all experiments. According to the results of previous studies on highly oriented NWs [16, 17] we assign the observed plasmon resonances to the longitudinal LSP mode. The suppression of the transverse resonance in the THz photoconductivity can be explained either by electron scattering from the NW surfaces [18] or Landau damping in the electron-hole plasma [19].

3. Results and discussion

3.1. Photoconductivity measurements

During the measurements, the THz beam path and the sample were enclosed in a box which was purged with nitrogen. This was done in order to reduce the water vapor content of the air, since it is a strong THz absorber. All measurements were performed at room temperature. For the NWs with $x = 0.20$, pump fluences of 12.7 and 52.5 μJ/cm$^2$ were used, whereas for $x = 0.30$ and $x = 0.44$ the fluences were 6.3 and 12.7 μJ/cm$^2$. From each OPTP measurement, the photoconductivity spectra $\Delta\sigma(\omega, \tau)$ were extracted in the range of pump-probe delay times up to $\tau = 350$ ps.
Figure 5. (a) Color map shows the measured normalized frequency dependent real part of photoconductivity of a In$_{0.2}$Ga$_{0.8}$As shell NW sample at different delay times. Color scale from blue to yellow corresponds to the normalized values of \( \text{Re}\Delta\sigma \), from 0 to 1. (b)–(d) Real (black) and imaginary (red) parts of the photoconductivity spectra at various PP delay times. The dots and circles are extracted data from the measurements while the lines are LSP fits. Pump fluence was 52.5 \( \mu \text{J cm}^{-2} \).

Figure 5(a) shows an example of the temporal evolution of the plasmonic response in the NWs with \( x = 0.20 \). In order to make the plasmon dynamics more clear, we have normalized \( \Delta\sigma(\omega, \tau) \) for every delay time \( \tau \). Since this measurement was recorded at the high pump fluence of 52.5 \( \mu \text{J cm}^{-2} \) the LSP frequency for short delay times is above the covered frequency range and the plasmon peak becomes visible only about 50 ps after the photoexcitation. As the delay time increases, the LSP shifts to lower frequencies as a result of the electron-hole recombination.

Figures 5(b)–(d) show the plots of real (black squares) and imaginary (red dots) parts of the photoconductivity for three chosen delay times. The drop of the photoconductivity intensity with time again shows the recombination dynamics of charge carriers, because the intensity is proportional to the concentration of charge carriers, and the intensity is proportional to the carrier concentration (equation (2)). LSP frequency can be easily spotted, since it is approximately at the maximum of \( \text{Re}\Delta\sigma(\omega) \) and at the zero crossing of \( \text{Im}\Delta\sigma(\omega) \).

3.2. Estimation of electron concentration and mobility

The extracted photoconductivities were fitted with equations (1) and (2) (lines in figures 5(b)–(d)), using LSP frequency \( \omega_0 \) and scattering rate \( \gamma \) as fitting parameters. The fitting was successfully performed for all delay times, so the fitting parameters were obtained as a function of \( \tau \): \( \omega_0(\tau) \) and \( \gamma(\tau) \). For that purpose, for each shell composition the appropriate bulk dielectric constants \( \varepsilon_m \), and the geometrical factors \( g \), calculated using the expression (3), were used. Namely, for \( x = 0.20, 0.30 \) and 0.44, \( \varepsilon_m = 13.23, 13.42 \) and 13.70 [1]; and the geometrical factors \( g(m = 0) = 0.228, 0.230 \) and 0.234 were used, respectively.

Figure 6 shows the results of the fitting. Green, blue and red colors correspond to the pump fluences of 6.3 \( \mu \text{J cm}^{-2} \), 12.7 \( \mu \text{J cm}^{-2} \) and 52.5 \( \mu \text{J cm}^{-2} \), respectively. We estimated electron mobilities \( \mu(\tau) \) for each sample and fluence by inserting both the extracted \( \gamma(\tau) \) and the effective electron mass \( m^* \) of a corresponding bulk material into the expression \( \mu = e/m^*\gamma \). The values for \( m^* \) were also taken from [1]: \( m^* = 0.054, 0.050 \) and 0.045\( m_e \) for 20%, 30% and 44% In samples, respectively. Similarly, time dependent concentrations \( n(\tau) \) were estimated from the expression (1), using the LSP frequencies \( \omega_0(\tau) \) from the fit and the mentioned values for \( m^* \) and \( g(m = 0) \). These results are displayed in figure 7.

The mobility \( \mu(\tau) \) tends to stabilize for longer delays \( \tau \) for all samples and pump fluences, thus, it was fitted with a constant. The fits are shown in figures 7(b), (d) and (f): orange, light blue and pink lines are for the fluences of 6.3 \( \mu \text{J cm}^{-2} \), 12.7 \( \mu \text{J cm}^{-2} \) and 52.5 \( \mu \text{J cm}^{-2} \), respectively. The results are presented in figure 8(a). The measured mobilities lie between 54% and 83% of the mobilities in the corresponding n-doped bulk InGaAs with similar electron concentration of 2.3 \( \times 10^{17} \text{ cm}^{-3} \) (blue line) [20]. Taking into account that the InGaAs shell surfaces are not passivated, the measured mobility values are rather high. In fact, the only types of NWs with higher electron mobility at room temperature reported up to now are InAs NWs having 6000 cm$^2$ V$^{-1}$ s$^{-1}$ [5] and their alloys with Sb, for example InAs$_{0.65}$Sb$_{0.35}$, reaching 8300 cm$^2$ V$^{-1}$ s$^{-1}$ [21]. The low impact of the non-passivated shell surface on the carrier scattering in our NWs may be explained by the smaller surface-to-volume ratio of the InGaAs shells compared to typical NWs, where the charge transport occurs only in the core. Our
results do not show a monotonic dependence of mobility on x. We speculate that this is because of the different structural quality and/or the different surface conditions (e.g. degree of oxidation) for InGaAs x shells with different x.

In order to determine electron lifetimes from \( n(\tau) \), we performed mono-exponential decay fitting for the first 100 ps. This particular time interval was selected for two reasons:

(i) at longer delays the relaxation rate changes for some samples and more complex modeling taking into account heterogeneity of the NWs and/or different recombination mechanisms [6] have to be applied; (ii) the error margins increase for longer \( \tau \) making the fitting less reliable. The fitting curves in figures 7(a), (c) and (e) have the same colors like the ones from the mobility fits. The measurement for \( x = 0.20 \) and 52.5 \( \mu \) cm\(^{-2} \) fluence was excluded from the analysis because the electron concentration was out of the measurement range during the first 50 ps.

The electron lifetimes for all samples as a function of pump fluence are plotted in figure 8(b). It can be seen that for a given NW type, the electron lifetime increases with the pump fluence. This dependence is characteristic for all NWs and it occurs due to the saturation of the surface traps at higher photoexcited carrier densities [10, 22]. That is, as the pump fluence increases, the number of available surface states drops and carriers can live longer.

The initial concentration of electrons versus pump fluence is shown in figure 8(c). One can easily see that for a fixed fluence, the initial concentration increases for the NWs with higher indium content. This can be attributed to stronger absorption of the pump pulse in shells with higher x that have lower band gap.

4. Conclusion

We performed OPTP spectroscopy on GaAs/In\(_{x}\)Ga\(_{1-x}\)As core/shell NWs. The extracted photoconductivities were successfully fitted with LSP model using only the plasmon frequency \( \omega_0 \) and the scattering rate \( \gamma \) as fitting parameters. The scaling between the plasmon frequency and its strength can be precisely determined using the analytical equation for the geometrical factor of a cylindrical NW with given dimensions. We estimate electron mobilities, concentrations and lifetimes in our In\(_{x}\)Ga\(_{1-x}\)As NW shells with various
compositions. The mobility values are found to be in the range of \(3000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}\) for unpassivated shells with \(x\) from 0.20 to 0.44. We anticipate that improved mobility values can be obtained in passivated structures, allowing for their employment in the future as nanoscale HEMTs on Si.

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