

## Tunable Hot-Electron Transfer Within a Single Core-Shell Nanowire

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We report the hot photoexcited electron transfer across the coaxial interface of a cylindrical core-shell nanowire. Modulation of the transfer rates, manifested as a large tunability of the voltage onset of negative differential resistance and of voltage-current phase, is achieved using three different modes. The coupling of electrostatic gating, incident photon energy, and the incident photon intensity to transfer rates is facilitated by the combined influences of geometric confinement and heterojunction shape on hot-electron transfer, and by electron-electron scattering rates that can be altered by varying the incident photon flux, with evidence of weak electron-phonon scattering. Dynamic manipulation of this transfer rate permits the introduction and control of a continuously adjustable phase delay of up to  $\sim 130^\circ$  within a single nanometer-scale device element.

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Understanding the effects of finite size and dimensionality on the interaction of light with semiconductor nanostructures, e.g., local enhancement of electromagnetic fields [1], optical absorption [2], exciton dynamics [3], multiexciton generation [4], and electron transfer across semiconductor heterostructures [5] is central to identifying and exploiting [6–10] novel modes of efficient conversion and transfer of energy. However, insight into the physics of hot-electron cooling and transfer across electrically interfaced heterojunctions, realized and studied extensively in real-space transfer (RST) devices, has been limited to planar interfaces in which local carrier densities are bounded principally by excited state lifetimes rather than finite geometric or mesoscopic symmetry considerations.

Here we report the tunable hot-electron transfer of photoexcited electrons across the coaxial interface of a core-shell nanowire. The rate of RST in the coaxial nanowire geometry can be tuned using three different mechanisms and is manifest as a tunability of the negative differential resistance, where the dependence of the transfer rate on an energy relaxation length scale is expected to possess a stronger power law dependence in the coaxial geometry than in a planar geometry. Tunability using an incident light wavelength indicates an ultrafast carrier dynamic, which is relevant to the study of hot carrier cooling. The controlled variation of incident photon flux on electron-electron interactions represents a promising means of investigating momentum relaxation and distribution in geometrically confined nanostructures. The introduction of a single nanoscale element possessing electrically and optically tunable complex impedance opens the possibility of assembling integrated circuits, including oscillators, amplifiers [11], phase shifters, frequency multipliers, phase-locked loops, and laser switches

[12], using fewer and far smaller elements, and with the versatility of a highly local optical interface [8,13–15].

Core-shell nanowires (CSNWs) each composed of a GaAs core and an  $\text{Al}_{0.33}\text{Ga}_{0.67}\text{As}$  shell were grown via metallorganic vapor phase epitaxy without the additional introduction of dopant. CSNWs were dispersed onto a 200-nm thick thermally grown  $\text{SiO}_2$  film on an electrically contacted Si(100) substrate for electrostatic gating, and electron beam lithography was used to define electrical contacts directly to the core near each end of the CSNWs. The NW cores possess a nearly intrinsic character at 300 K: they are slightly *p*-type on the basis of the recent demonstration of gate modulation of conductance within GaAs NWs without a shell [16], consistent with the unintentional doping (C) from precursor molecules [17], and thereby substantially suppressing the scattering by ionized dopants in the GaAs NW cores. We estimate the alloy composition  $x$  to be 0.33 in our  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  shells based on the results of the Voigt line shape fitting-based determination of the positions of the GaAs TO and LO phonon modes, and AlAs- and GaAs-like TO and LO modes for the  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  alloy in the Raman spectra collected from the core-shell NWs, thus permitting the extraction of values for  $x(\text{Al})$ . Values obtained for  $x$  were consistent (within a few percent) with those obtained from photoluminescence spectroscopy. The shells are expected to be weakly *n*-type due to unintentional doping (Si) from the Al-based precursor.

The CSNWs exhibit significant photocurrent sensitivity ( $\sim 0.1 \mu\text{A/W}$ ) at 300 K in their low-bias voltage response to either monochromatic laser or broadband illumination as evidenced by remarkably small dark currents ( $< 50 \text{ fA}$ ), amounting to more than 3 orders of magnitude in the linear variation in the photocurrent. A nearly linear photocurrent-voltage relationship for small DC bias  $V_{\text{bias}}$

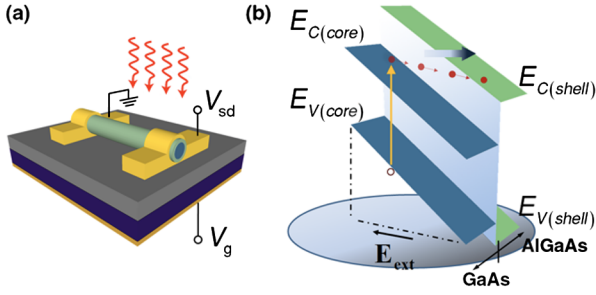


FIG. 1 (color online). Schematic illustrations of (a) NW device configuration under illumination and under source-drain bias  $V_{sd}$ , and (b) electronic band diagram depicting the real-space transfer of photoexcited electrons from the NW core to the shell conduction bands under large bias field  $E_{ext}$ .

at 300 K indicates that photo-generated carriers encounter a negligibly small barrier when collected. However, the application of a larger bias reveals negative differential resistance (NDR) in the photocurrent, where a threshold voltage  $V_{th}$  is defined by the onset of the NDR. For  $V_{bias} \approx V_{th}$  photoexcited electrons in the GaAs NW core can acquire sufficient energy from the large component of  $E$  parallel to the NW axis, exceeding the GaAs-Al<sub>0.33</sub>Ga<sub>0.67</sub>As conduction band offset ( $\Delta E_c \approx 0.255$  eV [18]), and undergo RST [19–23] into the shell [Fig. 1(b)]. For  $V > V_{th}$ , the curve bends down, and with a further increase in the applied electric field  $E$ , the slope of the curve becomes nearly zero and the photocurrent is saturated.

For sufficiently large  $E$ , the RST of photoexcited electrons from the higher electron mobility NW core to the lower-mobility and wider-gap shell, and the accompanying observation of NDR in photocurrent density  $J_{h\omega}$ , can be described by  $dJ_{h\omega}/dE = q\delta n[\mu_1 - f\Delta\mu - (df/dE)\Delta\mu E]$ , where  $\Delta\mu = \mu_1 - \mu_2$ ,  $\mu_1$  and  $\mu_2$  denote

GaAs core and Al<sub>0.33</sub>Ga<sub>0.67</sub>As shell electron mobilities (bulk values are  $\sim 8000$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> [11] and  $\leq 500$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> [11]), respectively,  $\delta n$  is the concentration of photo-induced electrons within the GaAs core, and  $f$  is an  $E$  field-dependent fraction of carriers photo-excited within the core that reach the shell. A peak in the current is expected when  $dJ_{h\omega}/dE = 0$ , i.e.,  $\mu_1/\Delta\mu = f + Edf/dE$ , from which estimates of  $f$  at  $V_{th}$  can be obtained by numerically solving the above ( $0.23 \leq f \leq 0.45$ ). Because of the complexity of solving the Boltzmann transport equation involving RST, Monte Carlo simulations are usually applied to simulate the velocity  $v - E$  relationship [19,24,25]. Assuming  $E$  is constant along the axis of the NW, we estimate that our measured  $V_{th}$  corresponds to  $E \approx 10$  kV/cm. This is comparable to the value of  $\sim 3$  kV/cm in bulk structures calculated by Monte Carlo simulations and obtained experimentally [21,24]; an increase is consistent with predictions of increases in the threshold voltage for decreased dimensionality from bulk to two dimensions [19].

On what basis can the observed NDR in photocurrent be attributed to RST? RST is directly related to the energy that a hot electron accrues from the incident photon energy. Incident radiation with a larger energy difference ( $E_{h\omega} - E_{g,GaAs}$ ) will generate a broader distribution of electron energies in excess of  $E_{g,GaAs}$  (and higher electron temperatures), requiring a lower electric field to undergo RST. In fact, we verified this dependence by observing a shift in  $V_{th}$  with incident photon energy, i.e.,  $dV_{th}/dE_{h\omega} < 0$  [Fig. 2(a)]. In addition to its potential application for a wavelength-selective nonlinear device element, significantly, this finding also indicates that the transit time for electrons to reach the RST threshold is comparable to, or shorter than, the hot-electron cooling time, indicating the potential for this device in high-frequency RST-based nanoscale optoelectronics.

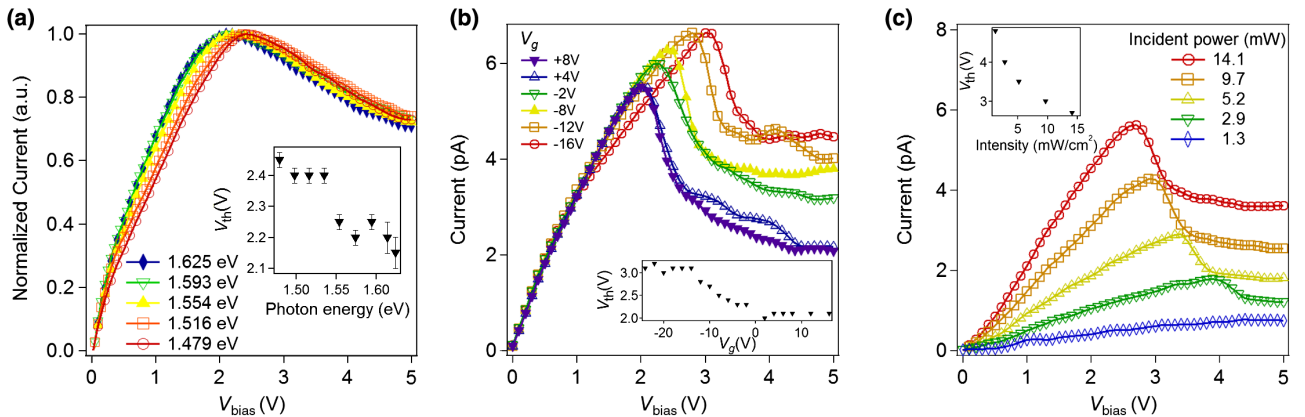


FIG. 2 (color online). Photocurrent-voltage traces, collected at 300 K, for different values of (a) laser excitation wavelength demonstrating tunability (trace is normalized by the peak current at  $V_{th}$  (Inset: decrease of  $V_{th}$  for increasing laser excitation energy)); (b) substrate gate bias  $V_g$ , as denoted in the legend (Inset: the increase in  $V_{th}$  for larger negative gate voltages, demonstrating the electrostatic gate tuning of  $V_{th}$ ); and (c) incident optical intensity, as denoted in the legend (Inset: dependence of  $V_{th}$  with incident optical intensity).

$V_{th}$  depends on  $\Delta E_c$  and thus we propose that the rate of RST and the field associated with its onset in a CSNW can be manipulated via the electrostatic gating of the entire length of the CSNW, given its radial proximity to the gate electrode. The measured photocurrent vs  $V_{bias}$  with substrate gating demonstrates that  $V_{th}$  can be tuned by up to  $\sim 50\%$ , with  $dV_{th}/dV_g < 0$ , confirming that it is electrons (and not holes) in the GaAs core that are undergoing RST [Fig. 2(b)]. The additional field contributed by  $V_g > 0$  provides a radial component of momentum to the electrons in the GaAs core (transverse to the heterojunction) to enter the AlGaAs shell. Nonuniformity in the radial field distribution from gate coupling is expected: a maximum field is reached where the NW is in physical contact with the gate oxide [26].

The asymmetric NDR response of our devices to gating can be understood by considering the additional contribution to the overall capacitance owing to the NW shell-core interface in the form of a bipolar ( $n-p$ ) junction-isolated channel. Electron transfer across the NW heterojunction under  $V_g > 0$  is analogous to a negatively biased  $p-n$  junction, in which saturation can be attained with small  $V_g$ . For  $V_g < 0$ , however, the bipolar channel is under positive bias and saturation is not observed for an appreciable range of  $V_g$ . A more detailed study of the extent of applicability of RST to observations of NDR on the photocurrent in CSNWs should consider momentum and energy relaxation mechanisms under  $E$ . However, on the basis of the strong electrostatic gating effect, of estimates of energy differences between the conduction band offset and transition energies associated with different conduction band minima in  $k$  space, and of the observed excitation energy dependence of RST, we assert that it is much more energetically favorable for electrons to undergo RST than intervalley transition(s) [18,27,28].

Under a high  $E$  field, electron-electron scattering contributes to randomizing energy gained from the  $E$  field along the channel, resulting in an increased momentum relaxation rate and a shorter time scale for the carrier distribution to reach a steady state [19,29,30]. This is particularly beneficial for RST since the randomization process enables a larger fraction of transporting electrons in the channel to accrue a sufficient radial component of momentum to transfer across the interface.

Based on the principle that the electron-electron scattering rate is a function of carrier concentration (in our case  $\delta n$ ) we investigated the effect of this scattering rate on the onset of RST by varying the optical excitation intensity  $I_{h\omega}$ . Significantly, we observe that  $V_{th}$  can also be tuned by  $\sim 50\%$ , with  $dV_{th}/dI_{h\omega} < 0$  over the range  $0 \leq I_{h\omega} \leq 14.1 \text{ mW/cm}^2$  [Fig. 2(c)]. Below a threshold excitation intensity, however, the NDR feature is diminished, likely due to insufficient electron-electron scattering.

We rule out the possibility that the observed NDR is due to an unintended and unconfirmed presence of a tunnel

barrier in our devices: our experimental results and *a priori* knowledge that the GaAs NW cores are very weakly  $p$ -type [16], taken together with our estimate of the peak instantaneous net excess carrier concentration [31] under our experimental conditions ( $\delta n = 7.0 \times 10^{17} \text{ cm}^{-3}$  [32]), and the absence of an appreciable thermal contribution, indicate that a tunneling process cannot explain the observed NDR feature in the photocurrent and its tunability with electrostatic gating and incident photon flux.

We investigated the temperature dependence of the photocurrent in our CSNWs from 300 to 4.2 K. The strong NDR feature can also be seen at 4.2 K under higher power monochromatic (0.33 mW) laser illumination, where a nA-scale photocurrent and a more than 2:1 peak-to-valley photocurrent ratio are observed [Fig. 3(a)]. While a decrease in the low-bias photoconductance  $G_{h\omega}$  under a lamp illumination for decreasing temperature  $T$  from 300 to 160 K is seen, there is a remarkable absence of systematic variation in  $G_{h\omega}$  over the range from  $\sim 160$  to 4.2 K [Fig. 3(b)]. These results indicate that, for  $T \leq 160$  K, photoexcited carriers are not appreciably scattered by phonons in these devices.

We compare the probability of hot electrons to undergo RST in the cylindrical core-shell NW geometry to that for a planar structure, i.e., a thin layer of GaAs sandwiched between AlGaAs above and below it. Electron-electron scattering randomizes the momentum distribution while not changing the electron drift velocity. Assuming that

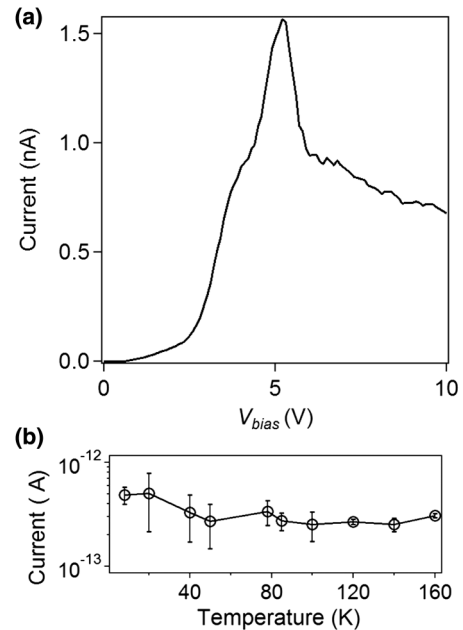


FIG. 3. (a) Measured nA-scale photocurrent response at 4.2 K and under 0.33 mW Ti:S laser irradiation. (b) Current at a fixed  $V_{bias}$  plotted as a function of  $T$ . The absence of a thermally activated contribution to the photocurrent response over this range of  $T$  as seen in (b) indicates that the scattering of photoexcited carriers by phonons is not significant.



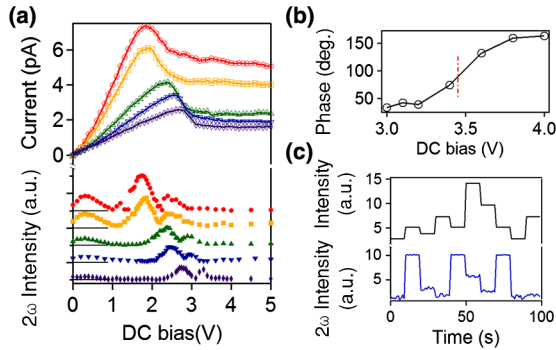


FIG. 4 (color online). (a) Top: photocurrent traces for selected values of incident power: 14.1 mW (red), 9.7 mW (yellow), 5.2 mW (green), 3.9 mW (blue), and 2.85 mW (magenta); bottom: measured  $2\omega$  photocurrent signals associated with a small (250 mV) ac voltage modulation (50 Hz) bias applied to the NW about  $V_{th}$ . (b) Demonstration of control of the photocurrent phase with DC  $V_{bias}$  for fixed incident power. The vertical dash-dotted red line denotes the value of  $V_{th}$  obtained from a DC  $I$ - $V$  trace under identical intensity. (c) Top: arbitrary time series of incident light power selected to shift the value of  $V_{th}$  in relation to a fixed  $V_{bias} = 2.4$  V; bottom: corresponding  $2\omega$  signal, demonstrating the optical amplitude modulation of the nonlinear response.

the NW diameter ( $2R$ ) and film thickness ( $t$ ) in the planar structure are each small in relation to the absorption depth (i.e., the photoexcited electrons are uniformly distributed within the GaAs NW core or thin film layer) and defining an energy relaxation length  $L$  within which (all) hot electrons lose energy when entering the AlGaAs shell (or film), the probability of hot electrons created within the GaAs NW core undergoing RST (and the relative degree of tunability) is, to leading order in  $L$ , stronger ( $\sim L^2$ ) than that for the planar geometry ( $\sim L$ ). (See [37] for Supplemental Materials)

We describe how the CSNW can be operated as an electrically or optically controllable nonlinear nanoscale device element. For example, in the introduction of an ac modulation to  $V_{bias}$  at frequency  $\omega$  about selected values of  $V_{th}$  defined by different values of incident optical intensity, we demonstrate full-wave rectification as seen by modulation frequency doubling, and tunability of its maximum along the bias voltage axis [Fig. 4(a)]. For fixed incident power, the phase difference (between that of the ac voltage applied to the sample leads and the generated ac photocurrent)  $\Delta\phi$  can be tuned continuously by bias voltage with  $d\phi/dV \approx 130^\circ/V$  [Fig. 4(b)], demonstrating the potential for these CSNW devices as programmable phase delay elements and for use in frequency multiplication. Further, the  $2\omega$  current response to a sequence of arbitrary-amplitude light pulses incident upon the CSNW device under ac bias voltage [Fig. 4(c)] demonstrates optical amplitude control of nonlinearity in photocurrent.

The introduction of a single nanoscale element possessing electrically and optically tunable complex impedance

opens the possibility of assembling integrated circuits, including oscillators, amplifiers [11], phase shifters, frequency multipliers, phase-locked loops, and laser switches [12] using fewer and far smaller elements, and with the versatility of a highly local optical interface [8,13–15]. While challenges owing to the effects of parasitic capacitance from the substrate must be addressed, we propose that, based on our observation of tunable frequency doubling at 1 kHz and on the inherent relaxation time scales for RST of  $\sim 10^{-12}$  s [19], practical GHz-range single nanowire devices exhibiting tunable phase should be within reach, particularly given recently reported evidence of intrinsic picosecond-scale response time characteristics of these CSNWs[33]. Finally, the dynamic control of hot-electron transfer rates in nanoscale heterojunctions as reported here is relevant for novel photovoltaic devices: a significant reduction in the time scale of a hot-electron transfer from semiconductor nanocrystals was demonstrated recently[34] renewing intense interest in efforts to approach the theoretical limit of quantum efficiency for a hot-electron transfer that far exceeds [35] the Shockley-Queisser limit [36].

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