Single-nanowire, low-bandgap hot carrier solar cells with tunable open-circuit voltage

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Abstract

Compared to traditional pn-junction photovoltaics, hot carrier solar cells offer potentially higher efficiency by extracting work from the kinetic energy of photogenerated ‘hot carriers’ before they cool to the lattice temperature. Hot carrier solar cells have been demonstrated in high-bandgap ferroelectric insulators and GaAs/AlGaAs heterostructures, but so far not in low-bandgap materials, where the potential efficiency gain is highest. Recently, a high open-circuit voltage was demonstrated in an illuminated wurtzite InAs nanowire with a low bandgap of 0.39 eV, and was interpreted in terms of a photothermoelectric effect. Here, we point out that this device is a hot carrier solar cell and discuss its performance in those terms. In the demonstrated devices, InP heterostructures are used as energy filters in order to thermoelectrically harvest the energy of hot electrons photogenerated in InAs absorber segments. The obtained photovoltage depends on the heterostructure design of the energy filter and is therefore tunable. By using a high-resistance, thermionic barrier, an open-circuit voltage is obtained that is in excess of the Shockley–Queisser limit. These results provide generalizable insight into how to realize high voltage hot carrier solar cells in low-bandgap materials, and therefore are a step towards the demonstration of higher efficiency hot carrier solar cells.

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(Some figures may appear in colour only in the online journal)
carriers through Si quantum dots in SiO₂ has been investigated [9, 10]. Ultra-fast, hot electron collection has been demonstrated in bandgap engineered GaAs/AlGaAs heterostructures [11, 12] and hot carrier transport across an InP/PbSe interface has been studied [13]. Hot carriers have been spectroscopically observed and predicted to result in solar cell efficiency enhancement in GaAsP/InGaAs quantum wells [14] and hot carrier photocurrent has been observed in a GaAs/InGaAs quantum well solar cell [15]. Ferroelectric insulators have been demonstrated to exhibit above bandgap photovoltages [16] and barium titanate, BaTiO₃, has been shown to exhibit power conversion efficiencies in excess of the Shockley–Queisser limit [17] due to hot carriers and the bulk photovoltaic effect [18, 19].

All of the above demonstrations of extraction of photogenerated, hot carriers have been performed in materials with a relatively large bandgap (i.e. \( E_G > 1 \) eV). However, the maximum power conversion efficiency achievable with a hot carrier solar cell depends upon the bandgap of the material, and the theoretically achievable efficiency in hot carrier solar cells is the highest in low-bandgap materials (i.e. \( E_G < 0.5 \) eV) [3, 4]. Recently, we reported single-nanowire, photothermoelectric devices that produced bipolar currents under illumination by different wavelengths of light [20]. Here, we point out that these devices are in fact, low-bandgap hot carrier solar cells as they were made of wurtzite (WZ) InAs, which has a room temperature bandgap of only 0.39 eV [21, 22]. In this work, we expand upon the discussion of these devices and show that they are hot carrier solar cells. We do this by comparing their measured current–voltage (\( I–V \)) curves to the Shockley–Queisser [23] detailed balance limit for an ideal pn-junction solar cell composed of the same absorbing material and showing that the open-circuit voltage of the highest resistance single-barrier device exceeds this limit. Then, we discuss the energy conversion process that allows achievement of this limit-breaking photovoltaic. Next, we demonstrate that photovoltage tunability through heterostructure engineering is a characteristic of the presented low-bandgap hot carrier solar cells by showing that when we increase energy filter transmissivity, we increase device conductivity and we decrease the achievable open-circuit voltage. Finally, we discuss topics for future work.

**Methods: device design and fabrication**

The devices in this study are based on single nanowires with either a single- or double-barrier heterostructure acting as an energy filter (figure 1). The basic principle for the generation of photocurrent and photovoltage in these hot carrier solar cells is illustrated in figures 1(b), (e) and relies on: (i) energy filters that separate photogenerated hot carriers (figures 1(c) and (d)), and (ii) absorption hot spots forming near the filters to give rise to photogenerated carriers in their vicinity (figures 1(b) and (e)). This localized increase in carrier concentration is possible because light absorption in a nanowire is not homogenous, but concentrated in hot spots corresponding to maxima of electromagnetic wave modes [20]. Electron–hole pairs are photogenerated predominantly in these hot spots and there establish a local non-equilibrium carrier temperature that can be much higher than the lattice temperature [20, 24, 25]. When an absorption hot spot is located within a hot-carrier diffusion length of a few hundred nanometers from an energy filter, hot electrons can diffuse across the filter before cooling. This charge movement results in a measurable photocurrent from which electrical power can be extracted (figure 1(b)) and the separation of photogenerated electrons and holes leads to the formation of an open-circuit voltage (figure 1(e)).

Nanowires are ideal for hot carrier solar cells for several reasons. First, their optical properties are highly tunable [26]: the concentration and confinement of light inside the nanowire (i.e. photonic effects) can be combined with the electromagnetic generation of surface-confined, oscillating electron plasmas at metal-dielectric interfaces (i.e. plasmonic effects) to control the position of spatially well-defined photon-absorption hot spots within the nanowire. This enables the ideal, nearby positioning of energy filters for fast carrier separation and work extraction (figures 1(b), (e)). Second, because of radial strain relaxation, nanowires are more amenable to bandgap engineering than planar devices [27, 28]. This enables heterostructures of materials of desirable bandgaps and band offsets to be selected and fabricated with atomic precision and with low defect densities. Third, likely because of reduced electron–phonon interaction in nanowires, the temperature of photogenerated carriers can be much higher than that of the lattice [20, 25]. Finally, a single-nanowire device setup enables the use of a backgate (figure 1(f)) to tune the carrier concentration during experiments [29]. This enables us to experimentally access different conductivity regimes within a single device.

In the proof-of-principle demonstration of [20], we used WZ InAs as the absorber material because of its small bandgap, \( E_G = 0.39 \) eV [21, 22], corresponding to light with bandgap wavelength \( \lambda_G = 3180 \) nm, allowing absorption of a broad spectrum of light. Furthermore, InAs exhibits high electron–hole asymmetries of effective mass and mobility, enabling photogeneration of high-energy, fast-diffusing electrons and low-energy, slow-diffusing holes, thereby assisting in electron collection across the energy-filter and charge separation. As the barrier/energy-filter material, we used InP (\( E_G = 1.34 \) eV, \( \lambda_G = 925 \) nm) [30]. We further defined two types of InAs/InP heterostructures (figure 1(a)), namely (i) single, thermionic barriers because they are predicted to produce the highest thermoelectric power [31, 32] (figure 1(c)) and (ii), double-barriers—which have been previously used in hot carrier solar cell experiments [11, 12]—because of the energy filtering effect [33] of resonant tunneling structures (figure 1(d)).

InAs/InP nanowire heterostructures with atomically sharp interfaces were grown using chemical beam epitaxy (figure 1(a)). Nanowires were transferred to an SiO₂ substrate equipped with a backgate, and we electrically contacted individual nanowires by electron beam lithography (figure 1(f)). Contacts were fabricated with a 400 nm inner separation, ensuring that hot carriers would only need to travel a maximum of about 200 nm to be collected.
across the heterostructure before they cooled—a much shorter distance than an estimated hot-carrier diffusion length in InAs (see supporting information is available online at stacks.iop.org/NANO/28/434001/mmedia for more information). The InAs material was naturally n-type and no pn-junction was present within the nanowires. Both types of energy filters used were grown into the same nanowires (figure 1(a)), and contacts were placed around the structure of interest in different devices (figure 1(f)). For clarity, in the following sections of this paper, devices in which contacts were placed around a double-barrier quantum dot will be referred to as double-barrier devices and devices in which contacts were placed around a single, thermionic barrier will be referred to as single-barrier devices.

Devices were electrically characterized in vacuum in a variable-temperature ($T = 6–300$ K) probe station with optical fiber access. DC electrical measurements were made using the measurement circuit shown in figure 1(f) using a Yokogawa 7651 DC source, a Stanford Research Systems SR570 current preamplifier, a Hewlett Packard 34401A multimeter and a Keithley 2636B SourceMeter. For photovoltaic characterization we used light generated by a supercontinuum laser and selected by a monochromator resulting in a Gaussian spectrum with a center wavelength of 720 nm and a full-width at half-maximum of 140 nm. Integration of the spectrum’s spectral irradiance results in a computed irradiance of 17.6 kW m$^{-2}$ and integration of the spectrum’s spectral photon flux results in a computed total above-bandgap photon flux of $6.77 \times 10^{22}$ photons m$^{-2}$ (see supporting information for method details).

**Results and discussion**

Dark and illuminated current–voltage (I–V) curves of the single-barrier device show that it was fabricated properly and that it produces electrical power when illuminated (figure 2). The dark current–voltage curve of the single-barrier device is symmetric and exponentially increasing under both forward and reverse bias (figure 2(a)). This is the characteristic current–voltage shape for thermionic emission over a barrier and confirms that the device

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**Figure 1.** Single nanowire hot carrier solar cell. (a) Scanning transmission electron high angle annular dark field micrograph of an InAs/InP heterostructure nanowire with single- and double-barrier InP segments that each can act as an energy filter. In any given device, only one of the segments is used as an energy filter; the other one is not contacted. InAs and InP segments are false colored (InAs—pink, InP—yellow) as a guide to the eye. (b) Band diagram of a single-barrier device under short-circuit current conditions. The red area indicates the location of a hot spot of photon absorption and carrier generation. Steps 1–3 indicate the process of current generation: (1) photogeneration of an electron–hole pair; (2) diffusion of a hot electron across the barrier, followed by thermalization; (3) the electron leaves to the left and drives a current through the circuit, filling the photogenerated hole from the right. The electron quasi-Fermi levels, $E_{Fn}$ at the contacts are indicated by red lines. (c), (d) Band diagrams under short-circuit conditions and geometry of the heterostructures used in this work. (e) Band diagram of a single-barrier device under open-circuit voltage conditions at a bias of 0.37 V. (f) 3D illustration of a single barrier hot carrier solar cell with electrical measurement circuit. Spacing between the inner edges of the contacts is 400 nm.
The single-barrier device does not contain a pn-junction. The figures of merit of the illuminated single-barrier device (figure 2(b)) are as follows: short-circuit current, $I_{SC} = -13.3 \pm 0.2$ pA, open-circuit voltage, $V_{OC} = 368$ mV $\pm 5$ mV, and fill-factor, FF = 27.5 $\pm$ 0.4%.

To place these results into context, we computed the dark and illuminated current–voltage curves of a pn-junction diode using the Shockley–Queisser detailed balance method (see supporting information for details). We assumed an ideal pn-junction solar cell made of WZ InAs that has the same projected area and surface area as our nanowire device and that is illuminated by our experimental spectrum. In this way, we calculated the following figures of merit: $I_{SC} = -165.6$ pA, $V_{OC} = 251$ mV, and FF = 68.7%.

The high measured $V_{OC} = 368$ mV for the illuminated single-barrier device compared to that of an ideal pn-junction ($V_{OC} = 251$ mV) provides strong evidence that hot-carrier energy conversion is essential to the voltage generation in the presented device, and enhances the achievable voltage compared to a pn-junction made of the same contacted absorber material. Our interpretation is that in the presented device kinetic energy of hot photogenerated electrons is converted into voltage based upon a thermoelectric effect [7, 8, 20], extracting electrical power from the differential in carrier temperature across the thermionic barrier. Because of this mechanism, hot carrier solar cells are not bound by the Shockley–Queisser detailed balance limit, which assumes isothermal energy conversion.

While the single-barrier device exceeds the $V_{OC}$ of an ideal pn-junction solar cell made of WZ InAs, the measured $I_{SC} = -13.3 \pm 0.2$ pA and FF = 27.5 $\pm$ 0.4% are much lower than those for the corresponding ideal pn-junction ($I_{SC} = -165.6$ pA, FF = 68.7%). This results in a lower power conversion efficiency of the single-barrier device compared to an ideal pn-junction solar cell made of WZ InAs.
There are three possible reasons for this smaller $I_{SC}$: first, the nanowire does not absorb all of the light that it is incident upon its projected area as its diameter is too small to support guided modes at the illumination wavelength. Second, not all of the absorbed light is absorbed in the hot spot next to the energy filter. Third, some photogenerated electrons and holes are likely to recombine within the single-barrier device before they are separated across the energy filter. This could happen if (1) a hot electron cools before crossing the energy filter, (2) the cooled electron recombines with its hole before being recycled up to energies high enough to cross the energy filter or (3) the hot electron diffuses in the direction opposite to the energy filter and recombines without encountering the energy filter. The smaller FF in the single-barrier device compared to an ideal pn-junction solar cell made of WZ InAs is because the current–voltage curve of the single-barrier device is linear in the power producing region. This is a characteristic of thermoelectric devices [34] and hot carrier solar cells based on the bulk photovoltaic effect [16–19].

While hot carrier solar cells are based on a thermoelectric effect, they offer opportunities for high-efficiency energy conversion that are different than those offered by traditional thermoelectric devices. This is because the presence of hot carriers can lead to very large temperature differentials over very small distances and between different distributions of particles (e.g., electrons and phonons). In comparison to traditional thermoelectric devices—in which performance is limited by parasitic heat flow in the lattice [35]—the heat transfer to the lattice in a nanoscale hot carrier solar cell can be suppressed if hot carriers are extracted from the device before they cool to the lattice temperature, a process that can be enhanced if electron–phonon energy exchange is inhibited by phononic engineering. As discussed in [20], we estimate the differential in the electron (carrier) temperature in the presented devices to be 170 K across the single-barrier, a value that is consistent with measurements of the non-equilibrium carrier temperature sustained in photogenerated carrier populations generated in small diameter nanowires under steady-state illumination [25]. Such a large temperature gradient would not be sustainable in traditional thermoelectrics, where carriers and phonons generally are in local thermal equilibrium, and it significantly enhances the achievable thermoelectric energy conversion efficiency. Importantly, power optimization and efficiency limits of thermionic thermoelectric devices have been studied [31, 32, 36, 37] and it has been shown that maximum power can be achieved at $T_C = 300$ K using a $k_B T_H$ filter with a barrier height of $1.1 k_B T_H$ [31]. Given the estimated $T_H$ of 470 K, this corresponds to a barrier of 45 meV. In this optimal configuration, a thermoelectric efficiency limit at maximum power of $\sim38\%$ of the Carnot efficiency is predicted [31], corresponding to $\sim14\%$ efficiency for the given $T_C$ and $T_H$—a result which is in agreement with the quantum bounds on thermoelectric power and efficiency [37].

How do hot carrier solar cells compare to pn-junction solar cells in terms of strategies to boost their open-circuit voltage? In pn-junction solar cells, increasing the open-circuit voltage requires the elimination of sources of non-radiative recombination in order to decrease bias-induced dark current and increase the ‘turn on’ voltage of the diode that comprises the solar cell. While reducing non-radiative recombination to increase short-circuit current is also important in hot carrier solar cells, of similar importance is engineering the energy-filtering, charge-separating heterostructure. To achieve a high open circuit voltage in a low-bandgap hot carrier solar cell, we find that it is necessary to have an energy filter that is highly resistive to low energy electrons and holes, while simultaneously highly transmissive to high energy electrons. An energy filter with these characteristics enables achievement of a large open-circuit voltage because (1) it prevents backflow leakage of cooled photogenerated electrons after they have transited the energy filter (2) it decreases the bias-induced dark current of the device and (3) it inhibits the movement of low-energy photogenerated holes, ensuring that ambipolar movement of photogenerated electron–hole pairs is avoided. In short, in a hot carrier solar cell, the photovoltage can be tuned by engineering the conductivity of the energy-filtering, charge-separating heterostructure.

Indeed, in our experiments, we observe an increase in the device conductivity and a decrease in the achievable open-circuit voltage when we use a double-barrier quantum dot (figure 1(d)) instead of a single, thermionic barrier (figure 1(c)) as the heterostructure energy filter. The increased conductivity of the double-barrier device in comparison to the single-barrier device can be attributed to the many current-carrying, resonant energy levels that exist below the barrier height in the quantum dot between the double-barriers. These energy levels result in a room temperature, zero gate voltage conductance that is approximately four orders of magnitude greater than that of a single-barrier device (figure 3(a)). Because of this high conductivity, to observe power-producing photocurrents and photovoltages under illumination, it is necessary to cool the double-barrier devices to $T = 6$ K and to apply a back-gate voltage of $V_G = -20$ V to suppress dark conductivity. Even then, the high transmissivity of the double-barrier heterostructure results in high illuminated conductivity and therefore, a low maximum open-circuit voltage of only $\sim17$ mV (figure 3(b)).

**Conclusion and outlook**

We foresee several routes to increasing the short-circuit current and the fill-factor of the presented low bandgap nanowire hot carrier solar cells. To increase the short-circuit current, we anticipate that the following strategies may be useful: (i) increasing nanowire diameter to increase absorption, (ii) optically designing contacts to increase absorption and to concentrate absorption on one side of and nearby an energy filter, (iii) passivating the nanowire surface to increase electron mobility and lifetime (iv) optimizing the placement, height and width of the energy filter, (v) tailoring the nanowire diameter, crystal phase and heterostructures to minimize electron relaxation rates due to phonon scattering, and (vi) adding a hole contact to collect holes and reflect electrons. Additionally, we anticipate that these optimization techniques may be applied in the modeling-guided design of vertical nanowire arrays [38] in which photons are absorbed more strongly closer to the tips of
the wires. Modeling suggests that it is possible to design nanowire diameter and array pitch such that a broadband absorption hot spot is present within the top 500 nm of the wires, where an energy filter could be placed within their hot-carrier diffusion length. Furthermore, additional concentration of longer wavelength light into this volume may be possible by use of plasmonic elements \[39, 40\]. Finally, to increase the fill factor, we anticipate adding band bending into the device by doping or by local gating to induce nonlinearity in the illuminated current–voltage curve.

Material choice will also play an important role in optimizing the devices described in this work. It is likely that by moving to absorbers with smaller bandgaps, higher carrier temperatures and efficiencies can be achieved as a larger fraction of photon energy will be converted into carrier kinetic energy. Moving to a wider bandgap barrier would likely enable larger open-circuit voltages by decreasing the thermionic dark current. However, maximum power has been predicted to be achieved with the estimated temperature difference at a barrier height of 45 meV \[31\], suggesting that a move to a narrower bandgap barrier material would be advantageous. In the end, to better understand the practical and the theoretical efficiency limits for these devices, and to determine the precise parameters of an ideal bandstructure, comprehensive optoelectronic and thermal device modeling will be required including self-consistent hydrodynamic simulations.

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