Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information for:

Enhanced thermoelectric figure of merit in thin GaAs nanowires

Xiaolong Zou,[a] Xiaobin Chen,[b] Huaqing Huang,[c] Yong Xu,*[d] and Wenhui Duan*[c]*

a. Department of Materials Science and nanoEngineering, Rice University, Houston, Texas 77005, USA
b. Department of Physics, Center for the Physics of Materials, McGill University, Montréal, Québec, H3A 2T8, Canada
c. Department of Physics and State Key Laboratory of Low-Dimensional Quantum Physics, Institute for Advanced Study, Collaborative Innovation Center of Quantum Matter, Tsinghua University, Beijing 100084, People's Republic of China
d. Department of Physics, McCullough Building, Stanford University, Stanford, California 94305-4045, USA

E-mail: xz20@rice.edu; yongxu@stanford.edu; dwh@phys.tsinghua.edu.cn

I. Landauer formula for thermoelectric properties

In the Landauer approach, the electronic current I and electronic thermal current I_Q can be expressed in terms of electron transmission function $\Xi(E)$. Supposing that the conductor is connected by two leads with chemical potential of $\mu_{+,-}$ and temperature of $T_{+,-}$,

$$I = \frac{2e}{h}\int dE\Xi(E)[f_+(E) - f_-(E)],$$

(1) $$I_Q = \frac{2}{h}\int dE\Xi(E)[(E-\mu_+)f_+(E) - (E-\mu_-)f_-(E)],$$

(2)

where $f_{+,-}(E) = 1/\exp[(E-\mu_{+,-})/k_BT_{+,-} + 1]$ is the Fermi-Dirac distribution function.

In the linear response region (i.e. voltage difference $\Delta V = (\mu_+ - \mu_-)/e = 0^+$ and temperature difference $\Delta T = T_+ - T_- = 0^+$), I and I_Q can be written as

$$I = \frac{2e^2}{h} I_0 \Delta V + \frac{2ek_B}{h} I_1 \Delta T \,, \tag{3}$$

$$I_Q = \frac{2ek_B T}{h} I_1 \Delta V + \frac{2k_B^2 T}{h} I_2 \Delta T \,, \tag{4}$$

where

$$I_n = \int dx \frac{x^n e^x}{(e^x + 1)^2} s(x) \,, \tag{5}$$

$x = (E - \mu)/k_B T$ $\mu = (\mu_+ + \mu_-)/2$, and $s(x) = \Xi(xk_B T + \mu)$. Then the electronic conductance σ, Seebeck coefficient S and electronic thermal conductance κ_e are:

$$\sigma = \frac{2e^2}{h} I_0 \,, \tag{6}$$

$$S = \frac{k_B}{e} \frac{I_1}{I_0} \,, \tag{7}$$

$$\kappa_e = \frac{2k_B^2 T}{h} (I_2 - \frac{I_1^2}{I_0}) \,, \tag{8}$$

On the other hand, thermal current contributed by phonons I_Q^p can be expressed in terms of phonon transmission function $\Xi_p(\omega)$:

$$I_Q^p = \frac{1}{2\pi} \int d\omega \Xi_p(\omega) \hbar\omega [f_+^B(\omega) - f_-^B(\omega)] \,, \tag{9}$$

where $f_{+,-}^B(\omega) = 1/\exp[(\hbar\omega)/k_B T_{+,-} - 1]$ is the Bose-Einstein distribution function. In the linear response region,

$$I_Q^p = \frac{1}{2\pi} \int d\omega \Xi_p(\omega) \hbar\omega \frac{\partial f^B(\omega)}{\partial T} \Delta T \,, \tag{10}$$

Then the phonon thermal conductance κ_p is

$$\kappa_p = \frac{k_B^2 T}{h}\int_0^\infty dx \frac{x^2 e^x}{(e^x-1)^2} s_p(x), \tag{11}$$

where $x = \hbar\omega / k_B T$ and $s_p(x) = \Xi_p(x k_B T / \hbar)$.

It is necessary to check Eq. (7) and Eq. (8), since there the denominator I_0 can be nearly zero when the Fermi level μ is located well within the band gap. For this purpose, we consider a simple case in which only one conduction band takes part in transport. The electron transmission function writes as $\Xi(E) = \Theta(E - E_{CBM})$, where E_{CBM} is the conduction band minimum and Θ is the step function. In Eq. (5), $s(x) = \Theta(x - x_0)$, where $x_0 = (E_{CBM} - \mu)/k_B T$. When μ is well below E_{CBM}, i.e., $x_0 \gg 1$, $L_0 \cong e^{-x_0}$, $L_1 \cong (1+x_0)e^{-x_0}$, and $L_2 \cong (x_0^2 + 2x_0 + 2)e^{-x_0}$. Then both $\sigma \cong \frac{2e^2}{h} e^{-x_0}$ and $\kappa_e \cong \frac{2k_B^2 T}{h} e^{-x_0}$ are nearly zero as expected, since no electronic state is close to the Fermi level. Physically, *S* is expected to be nearly zero as well. But here $S \cong -\frac{k_B}{e}(1+x_0)$ is artificially large, originated from the zero divide zero problem. Fortunately, this problem does not affect the calculation of power factor $P = \sigma S^2$, which approaches zero for large x_0. Based on Eqs. (6), (7), (8), and (11), the thermoelectric figure of merit *ZT* can be calculated.

II. Band structures for *wz*3 NWs

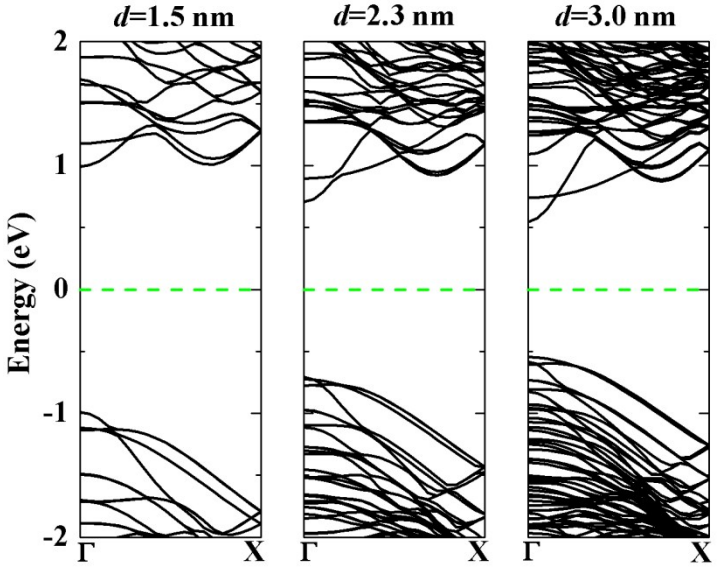

Figure S1. Band structures for *wz*3 NWs with different diameters. All the three GaAs NWs have direct band gap located at the Γ point. The band gap increases from 1.1 eV for 3.0-nm NW to 2.0 eV for 1.5-nm NW. The Fermi level (green dashed line) is set to the middle of the gap.

III. Normalized *ZT* and *P* for *wz*3 NWs

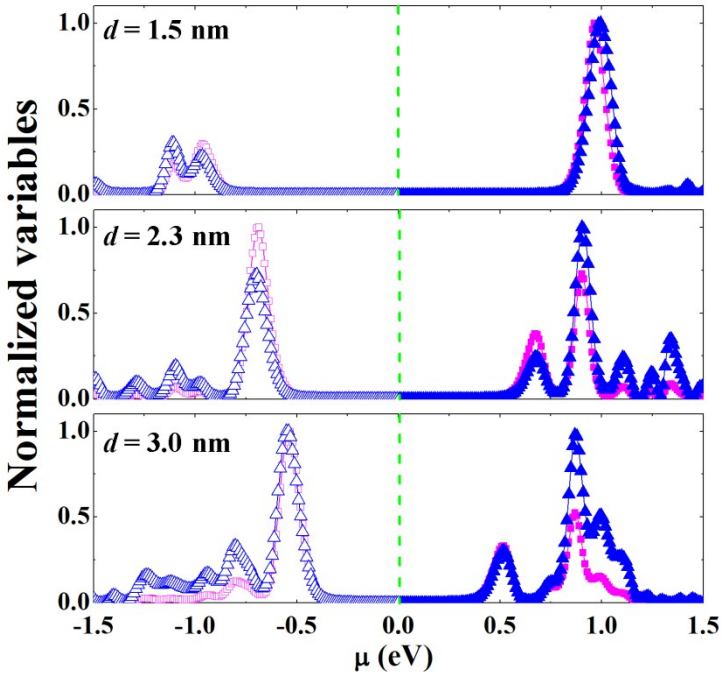

Figure S2. Normalized *ZT* (blue triangles) and *P* (pink squares) as functions of electronic chemical potential for *wz*3 NWs. Green dashed lines indicate the Fermi level. *ZT*/*P* values for *p*- and *n*-type NWs are shown in empty and filled symbols, respectively.

IV. VBM states for larger *wz*3 NWs

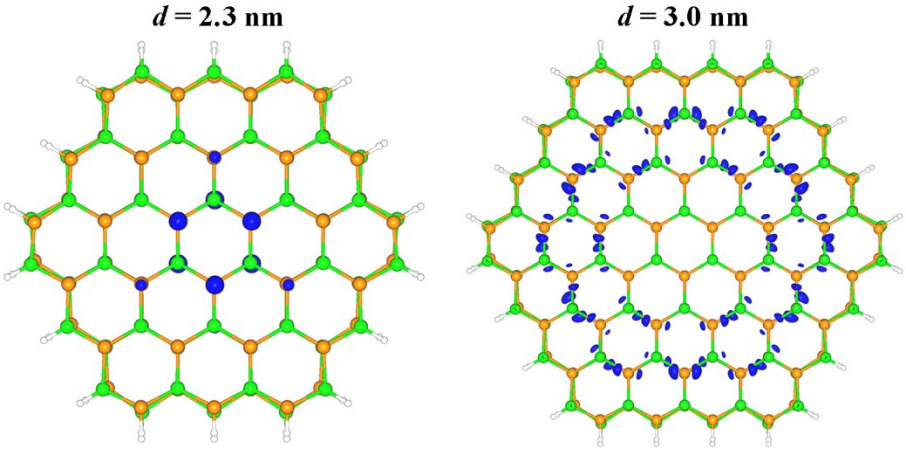

Figure S3. Partial charge density plots (isosurfaces in blue) of VBM states for bigger *wz*3 NWs. The green, orange, and white spheres represent the Ga, As and H atoms, respectively.

V. VBM and CBM states for 1.6-nm *zb*2 NW

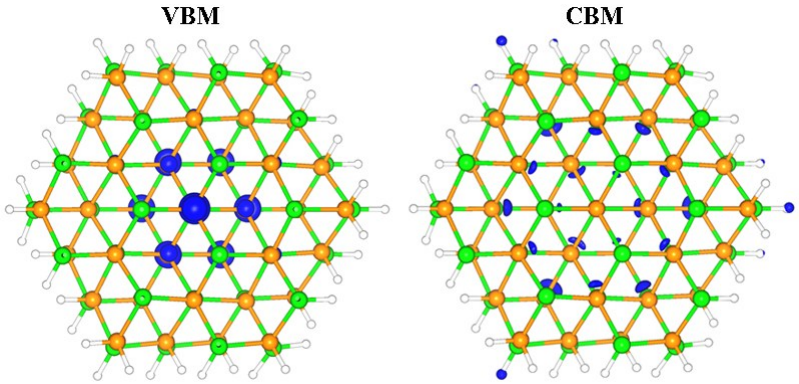

Figure S4. Partial charge density plots (isosurfaces in blue) of VBM and CBM states for 1.6-nm *zb*2 NW.