Utilizing transverse momenta and finite transmission bandwidth to enhance the performance of low-dimensional thermoelectrics

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Low-dimensional systems with sharp features in the density of states (DOS) have been proposed as a means to improving the efficiency of thermoelectric devices. Quantum dot (QD) systems, which offer the sharpest DOS achievable, however, suffer from low power outputs while bulk (3-D) thermoelectrics, while displaying high power outputs, offer very low efficiencies. Here, we analyze the use of a resonant tunneling diode (RTD) structure that combines the best of both aspects, that is, DOS distortion with a finite bandwidth due to confinement that aids the efficiency and a large number of current carrying transverse modes that enhances the total power output. We show that this device can achieve a high power output ($\sim 0.3 \text{MW/m}^2$) at efficiencies of $\sim 40\%$ of the Carnot efficiency due to the contribution from these transverse momentum states at a finite bandwidth of $kT/2$. We then provide a detailed analysis of the physics of charge and heat transport with insights on parasitic currents that reduce the efficiency. Finally, a comparison between our RTD device and a QD with comparable bandwidth reveals that a similar performance requires ultra-dense areal QD densities of $\sim 10^{12} / \text{cm}^2$.

Introduction: The thermoelectric figure of merit, $zT$, has traditionally been used to evaluate the performance across various thermoelectrics and is defined as:

$$zT = \frac{S^2 \sigma}{\kappa_{el} + \kappa_{ph}} T,$$

where $S$ is the thermopower (Seebeck coefficient), $\sigma$ is the electrical conductivity and $\kappa_{el}$ and $\kappa_{ph}$ are the electronic and lattice (phonon) thermal conductivities respectively. Efforts to improve the thermoelectric performance have focused on reducing $\kappa_{ph}$ through nanostructuring several interfaces in the device $^{13}$, or improving the power factor $S^2 \sigma$ by modifying the electronic density-of-states (DOS) $^{4}$. Following the original proposal of Hicks and Dresselhaus on $zT$ enhancement in quantum well (QW) and wire heterostructures $^{4}$. Sofo and Mahan $^{6}$ proposed that the optimum “transport distribution”, which is related to the the density of states in the device $^{10}$, for thermoelectric performance was a delta function, achievable in an ideal quantum dot (QD). It was subsequently shown that with a delta-like transport distribution, thermoelectric operation proceeds reversibly at Carnot efficiency $\eta_C$ under open circuit (Seebeck) voltage conditions$^{11,12}$. Reversible operation however necessitates zero power output, while finite power output occurs at efficiencies lower than $\eta_C$ $^{12}$. The figure of merit $zT$, which is an indicator only of maximum efficiency, is hence not the best indicator of thermoelectric performance specifically if the operation at optimum power is to be considered $^{8,12,15}$.

This power-efficiency tradeoff, which is an inevitable result of the bandwidth due to level broadening in QD devices, was further elucidated in subsequent works $^{8,12}$. A plot comparing efficiency and power output in a single-level QD device as a function of the level broadening $^{8}$, adapted in Fig. 1(a), clearly demonstrates the disparity between the maximum efficiency $\eta_{max}$ and the efficiency at maximum power $\eta_{max} P$ for low broadening (low power) and the severe degradation in the efficiency for high broadening (high power). This study also demonstrated the possibility of very high ($\text{upto 2} \text{MW/m}^2$) power outputs for 3-D thermoelectrics, albeit at a low efficiency ($\sim 20\%$ of $\eta_C$) and reinforced the conclusion that $zT$ is not a good indicator of the maximum power point. Also, by evaluating the power factor $S^2 \sigma$ per mode for 1, 2 and 3 dimensional thermoelectric devices in both ballistic and diffusive regimes, it was shown in $^{10}$ that while low-dimensional devices did offer some improvements, greater importance should be attached to the shape and magnitude of the number of current carrying modes $M(E)$. They also pointed out the high packing fraction and low size requirements for quantum wells and wires in order to realize their potential benefits over bulk thermoelectrics. It seems evident from the above discussion that optimizing thermoelectric performance necessitates thinking beyond the $zT$ picture so as to combine engineering of current carrying modes in addition to confinement.

Recently, a thermoelectric generator $^{16}$ based on a hot central cavity with a single energy level coupled to two cold contacts through resonant tunneling diode (RTD) heterostructures was proposed. These devices, which can be implemented as AlGaAs/GaAs/AlGaAs QWs, can potentially combine the benefits of a sharp DOS and high power output due to transport through perpendicular momentum modes as depicted in Fig. 1(b). Given the proof-of-concept that was developed in the aforesaid work, it is important to dwell into the details of the transmission spectra such as level broadening and the effect of excited levels that accompany a realistic RTD structure.

In this paper, we present a quantitative study of the performance of an RTD-based thermoelectric (Fig. 1(c)) with a realistic transmission line width $kT/2$ of the ground state energy level. We show that the power-
efficiency tradeoff is not as severe in this device as in QD devices, and it is possible to obtain high power (upto 0.3MW/m²) at an efficiency of 40% of $\eta_C$ through it. We also present a detailed analysis of the physics of charge and heat transport in the RTD devices and quantify the effect of high energy resonances and parasitic reverse currents on power output and efficiency. Finally, we compare RTD and QD devices and conclude that a very high ($10^{12}$/cm²) QD density is needed to match the performance of RTDs. Moreover, the two devices show similar maximum efficiencies, which we explain in terms of the relative widths of the energy levels in the transport window.

Simulation Methodology: A schematic of the heterostructure we have used in our simulations is shown in Fig. (c). Note that the simulated device extends to infinity in the x- and y-directions as indicated by the dotted lines in Fig. (c). Quantization in the z-direction is achieved by sandwiching a layer of GaAs (white) between two AlGaAs barriers (dark blue). The reason for choosing the AlGaAs/GaAs system is two-fold. First, this material system is extremely well studied and characterized, and devices such as lasers and photodetectors employing this system have been researched for many decades [17]. Second, the lattice constant shows very little variation over all compositions of AlGaAs, and hence modifications to the device bandstructure due to strain are minimal [18]. We can thus model these devices fairly accurately using a simple tight-binding Hamiltonian within the one-band effective mass model [19]. The conduction band diagram of this heterostructure along the light blue plane is shown in Fig. 2. The figure also portrays the basic thermoelectric operation at zero bias. High energy electrons in the tail of the Fermi distribution in the hot contact ($T_H = 330K$) can move through the channel into the cold contact ($T_C = 300K$), but a reverse flow of electrons is prevented due to the absence of states in the channel at energies below the Fermi energy. This energy filtering will improve as the width of the conducting state reduces, which explains the increase in efficiency with reducing level width (Fig. 1(a)). The temperatures at the hot and cold contacts $T_H$ and $T_C$, are taken to be the same as in [8] to enable a direct comparison of device performance.

In order to investigate the device properties at various positions of the ground state energy level relative to the equilibrium Fermi level ($E_{F,eq} = E - \mu_e$), we vary three parameters: the width ($w_b$) and the height ($E_b$) of the AlGaAs barriers, and the width ($w_w$) of the GaAs well such that the level broadening is fixed at $kT/2$, where $T = (T_H + T_C)/2$. Previous studies of nanoscale thermoelectrics have primarily focused on very sharp quantized levels to maximize efficiency [8, 16, 20]. There are several problems with this approach. Firstly, a mean-field analysis such as that employed here and previously is only accurate in the limit of large coupling to device contacts [21], which inevitably leads to a large level broadening. Secondly, experimentally grown III-V QD spectra typically show a linewidth $> 10$ meV at room temperature [22, 23]. $kT/2$ is thus a realistic lower bound on the currently achievable level broadening. Further, a previous study [24] considered the optimal bandstructure for thermoelectric performance and found that when the lattice conductivity is taken into account, a broadened dispersion produces a higher $zT$ than an ideal Sofo-Mahan delta function [24]. It is thus

![Fig. 1](image_url)
The integration along the transverse co-ordinate is performed assuming periodic boundary conditions along the device [19, 25]. In order to apply a bias ($V_{\text{bias}}$) across the device, we change the Fermi level of the hot contact. The self-consistent calculation accounts for the resultant shift in the device transmission function. The calculated transmission is then used in the Landauer equations for charge and heat current densities [16]:

$$J = \frac{e m^*}{2\pi^2 \hbar^2} \int dE \int dE_z T(E_z)[f_H(E_z + E_L) - f_C(E_z + E_L)]$$

(2)

and

$$J^Q_H = \frac{m^*}{2\pi^2 \hbar^2} \int dE \int dE_z T(E_z) [f_H(E_z + E_L) - f_C(E_z + E_L)]$$

(3)

The integration along the transverse co-ordinate is performed assuming periodic boundary conditions along these directions. The equations simplify to:

$$J = \frac{e m^*}{2\pi^2 \hbar^2} \int dE \int dE_z T(E_z)[F_H(E_z + E_L) - F_C(E_z + E_L)]$$

(4)

and

$$J^Q_H = J^Q_1 + J^Q_2,$$

where $J^Q_1$ and $J^Q_2$ are given by:

$$J^Q_1 = \frac{m^*}{2\pi^2 \hbar^2} \int dE_z T(E_z) [F_H(E_z + E_L) - F_C(E_z + E_L)]$$

(5)

$$J^Q_2 = \frac{m^*}{2\pi^2 \hbar^2} \int dE \int dE_z T(E_z) [G_H(E_z + E_L) - G_C(E_z + E_L)]$$

(6)

Here $F_i = \int_0^{\infty} dt \left( 1 + e^{t-x} \right)^{-1}$ and $G_i = \int_0^{\infty} dt t \left( 1 + e^{t-x} \right)^{-1}$, with $i = C/H$. It should be noted that our expression for heat current is different from [8] but agrees with [16]. However, we have found that this discrepancy makes only a minor difference numerically. The formalism described above enables us to study the energy distribution of the charge and heat currents, and hence characterize the parasitic components of current that bring down the efficiency. Since we have considered the full transmission spectrum and not just a single Lorentz-broadened peak for the device, we can also study the effect of high-energy, secondary resonances which will be present in real devices.

Simulation results and discussions: We present the efficiency in Fig. 3a and the power Fig. 3b from our device at various values of $E_{\text{pos}}$. We note that a power of 0.6 MW/m² at an efficiency of ~ 32% is obtained for $E_{\text{pos}} = kT$, and at $E_{\text{pos}} = 3.5kT$ the maximum efficiency touches 60% of $\eta_C$. Optimal performance, however, is obtained at $E_{\text{pos}} = 1.5kT$, with a power of 0.3 MW/m² at 40% of $\eta_C$. Although the powers we have obtained are approximately an order of magnitude below the thermionic power generators analysed in [8], they...
FIG. 4. (a) $zT$ and maximum power as a function of ground $E_{\text{pos}}$. It can be seen that the maximum $zT$ ($12$, at $E_{\text{pos}} = 3.5kT$) and maximum power ($0.6\text{MW/m}^2$, at $E_{\text{pos}} = kT$) are attained at very different points, thus highlighting the power-efficiency tradeoff. However, it is possible to obtain high power at high $zT$, as is evident at $E_{\text{pos}} = 1.8kT$ ($zT = 9$, power = $0.3\text{MW/m}^2$). (b) Efficiency derived from $zT$, calculated maximum efficiency and efficiency at maximum power ($\eta_{\text{maxP}}$) as functions of $E_{\text{pos}}$. $zT$ accurately predicts the maximum efficiency but overestimates $\eta_{\text{maxP}}$ in the region where power output is high.

function at nearly twice the efficiency which is a direct consequence of quantization in the transverse direction and the resulting narrowing of transmission spectrum.

We have also calculated $zT$ for the RTD device, which is plotted along with the maximum power as a function of $E_{\text{pos}}$ in Fig. 4(a). We immediately see that $zT$ and maximum power occur at very different $E_{\text{pos}}$, which supports the conclusions of several earlier studies that $zT$ is not a good indicator of the power performance of a thermoelectric. Corresponding to the maximum efficiency point at $E_{\text{pos}} = 3.5kT$ we get a $zT$ of 12. At the previously mentioned optimal $E_{\text{pos}}$ (1.5$kT$), $zT$ is 9. It is thus possible to obtain both a higher $zT$ and a higher power, and thus these devices present, what we believe, a better power-efficiency tradeoff than both QD and bulk thermionic devices by combining the advantages of both.

In Fig. 4(b) we plot the efficiency calculated from $zT$, the calculated maximum efficiency and the calculated efficiency at maximum power ($\eta_{\text{maxP}}$) as functions of $E_{\text{pos}}$. It is apparent that while $zT$ predicts the maximum efficiency quite accurately over the entire range of $E_{\text{pos}}$, in the region of significant power output, however, it overestimates $\eta_{\text{maxP}}$. This points once again to the unsuitability of $zT$ as the sole design parameter for low-dimensional thermoelectrics.

To better understand the factors responsible for limiting the efficiency, we analyze the physics of transport through the thermoelectric device in Fig. 5. $J_{\text{norm}}$ in Fig. 5(b) denotes the charge (heat) current density that has been normalized to the total charge (heat) current. Note that although Fig. 5 is plotted for $E_{\text{pos}} = kT$ and $V_{\text{bias}} = 5 \text{mV}$, the discussion that follows is very general.

Electrons in the device can be classified into three “transport windows” on the basis of their energies shown in the shaded regions in Fig. 5(a) and (b)): low energy, moving from the cold to hot contact; intermediate energy, moving from hot to cold contact and high energy, also moving from hot to cold contact. Fig. 5(c) is a zoomed in version of the low energy window. Charge current density here is negative, but much smaller in magnitude than the intermediate window, but the heat current is positive. We similarly zoom into the high energy window in Fig. 5(d). Due to their higher energy
these electrons make a higher fractional contribution to heat current than the charge current \[8\]. Both these regions together contribute in reducing the device efficiency.

As an interesting aside, we note that the presence of secondary resonances in the high energy window of the transmission spectrum (Fig. 5(a)) enhances the contribution of this window to the charge and heat currents (Fig. 5(c)). Such secondary resonances will be present in a realistic low-dimensional thermoelectric device, but their contribution to degrading the efficiency has not been considered previously \[8, 10, 20\]. Here, for example, if the secondary resonance is ignored, we find that the efficiency increases from 40% to 46% of \(\eta_C\), a difference of nearly 15%. A quantitatively accurate model of low-dimensional thermoelectric device performance must therefore take the entire transmission spectrum and not just a simple Lorentz-broadened levels into account.

**Comparison of QD and RTD devices:** Unlike QDs, RTD devices do not approach Carnot efficiency even close to the open circuit voltage, because of transport through transverse momentum states (Fig. 1(c)). This is easily seen from (5) and (6). The second term in (6) is not present in QDs. Even if we assume an ideally sharp delta-like energy level, which leads to the Carnot efficiency at the Seebeck voltage \(V_s\), we see that the second term will lower the efficiency for RTDs. It is also worthy to note that the RTD device features a higher Seebeck voltage \(V_s\) due to the contribution from transverse current carrying modes.

The advantage of RTD thermoelectrics however lies in the large power output. From \[8\] we see that at resonance widths of \(\sim kT/2\), a QD can give a thermoelectric power output of \(\sim 30pW\) per dot as noted from Fig. 1(a). To match the power density of an RTD (0.3MW/m\(^2\)), a QD density of \(\sim 10^{12}/cm^2\) is required \[10\]. Self-assembled III-V QDs typically display a density of \(\sim 10^{11}/cm^2\). Further, we see in Fig. 6(a) that the efficiency at maximum power is nearly the same for both the RTD and QD. While Fig. 6(a) is for \(E_{pos} = kT\), this observation is ubiquitous. While this may seem surprising at first, since a QD is expected to be a much more efficient energy filter than an RTD, we note that both charge and heat transport occur primarily in a window of width \(\sim kT\) around the equilibrium Fermi level, where the difference in occupation of the two contacts is significant \[10\]. Since the level width is also of order \(\sim kT/2\), both the RTD and QD are expected to behave similarly in terms of energy filtering. To confirm this we reduced the width of the dot level to \(kT/20\), upon which its maximum efficiency increased to 60% of \(\eta_C\) while the RTD efficiency remained almost unaffected. We thus conclude that not only can the RTD surpass QD thermoelectrics in terms of power output, but can also match its efficiency for realistic ground state energy level broadening.

In conclusion, we have analyzed the thermoelectric performance of a finitely broadened RTD-based device and shown that it can attain high powers (0.3MW/m\(^2\)) at high efficiencies (\(\sim 40%\) of \(\eta_C\)) because of the combined benefits of a large number of transverse momentum modes to carry current and longitudinal energy quantization to enhance filtering. By considering the energy spectrum of the charge and heat currents we estimated the effects of higher energy resonances on the device performance. We also showed that the RTD might be preferable to the QD-based thermoelectric for realistic level broadening. We note however that this study is confined to consideration of the electronic component of thermal conductivity, and a complete understanding of low-dimensional thermoelectrics requires the inclusion of phonon heat transport too. The determination of the best low-dimensional thermoelectric considering both electron and phonon transport, as well as quantification of the power and efficiency performance of this thermoelectric is a fruitful avenue of future research.

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[1] G. J. Snider and E. S. Toberer, Nature Mater. 7, 105 (2008).
[2] T. C. Harman, P. J. Taylor, M. P. Walsh, and B. E. LaForge, Science 297, 2229 (2002).
[3] B. Poudel, Q. Hao, Y. Ma, Y. Lan, A. Minnich, B. Yu, X. Yan, D. Wang, A. Muto, D. Vashaee, et al., Science 320, 634 (2008).
[4] L. D. Hicks and M. S. Dresselhaus, Phys. Rev. B 47, 12727 (1993).
[5] L. D. Hicks and M. S. Dresselhaus, Phys. Rev. B 47, 16631 (1993).
[6] G. D. Mahan and J. O. Sofo, Proc. Natl. Acad. Sci. U.S.A. 93, 7436 (1996).
[7] J. P. Heremans, V. Jovovic, E. S. Toberer, A. Saramat, K. Kurosaki, A. Charoenphakdee, S. Yamanaka, and G. J. Snyder, Science 321, 554 (2008).
[8] N. Nakpathomkun, H. Q. Xu, and H. Linke, Phys. Rev. B 82, 235428 (2010).
[9] M. S. Dresselhaus, G. Chen, M. Y. Tang, R. G. Yang, H. Lee, D. Z. Wang, Z. Ren, J. P. Fleurial, and P. Gogna, Advanced Materials 19, 1043 (2007).
[10] R. Kim, S. Datta, and M. Lundstrom, J. Appl. Phys. 105, 034506 (2009).
[11] T. E. Humphrey and H. Linke, Phys. Rev. Lett. 94, 096601 (2005).
[12] B. Muralidharan and M. Grifoni, Phys. Rev. B 85, 155423 (2008).
[13] M. Esposito, K. Lindenberg, and C. Van den Broeck, Phys. Rev. Lett. 102, 130602 (2009).
[14] M. Esposito, K. Lindenberg, and C. Van den Broeck, EPL 85, 60010 (2009).
[15] M. Esposito, R. Kawai, K. Lindenberg, and C. Van den Broeck, Phys. Rev. Lett. 105, 150603 (2010).
[16] B. Sothmann, R. Sanchez, A. Jordan, and M. Buttiker, New J. Phys. 15, 095021 (2013).
[17] P. Bhattacharya, Semiconductor Optoelectronic Devices (Pearson Education, Inc., 1997).
[18] B. G. Streetman and S. K. Banerjee, Solid State Electronic Devices (Prentice-Hall, Inc., 2006).
[19] S. Datta, Quantum Transport: Atom to Transistor (Cambridge University Press, 2005).
[20] A. Jordan, B. Sothmann, R. Sanchez, and M. Buttiker, Phys. Rev. B 87, 075312 (2013).
[21] B. Muralidharan, A. W. Ghosh, and S. Datta, Phys. Rev. B 73, 155410 (2006).
[22] M. Srujan, K. Ghosh, S. Sengupta, and S. Chakrabarti, J. Appl. Phys. 107, 123107 (2010).
[23] S. Shah, K. Ghosh, S. Jejurikar, A. Mishra, and S. Chakrabarti, Mater. Res. Bull. 48, 2933 (2013).
[24] C. Jeong, R. Kim, and M. Lundstrom, J. Appl. Phys. 111, 113707 (2012).
[25] R. Lake, G. Klimeck, R. C. Bowen, and D. Jovanovic, J. Appl. Phys. 81, 7845 (1996).