

Thermoelectric Signatures of Coherent Transport in Single-Molecule Heterojunctions

J. P. Bergfield*[†] and C. A. Stafford*[‡]

College of Optical Sciences, University of Arizona 1630 East University Boulevard,
Tucson, Arizona 85721, and Department of Physics, University of Arizona,
1118 East Fourth Street, Tucson, Arizona 85721

Received May 14, 2009; Revised Manuscript Received July 9, 2009

ABSTRACT

An exact expression for the heat current in an interacting nanostructure is derived and used to calculate the thermoelectric response of three representative single-molecule junctions formed from isoprene, 1,3-benzenedithiol, and [18]-annulene. Dramatic enhancements of the thermopower S and Lorenz number L are predicted when the junction is tuned across a node in the transmission function, with universal maximum values $S_{\max} = (\pi/3^{1/2})(k_B/e)$ and $L_{\max} = (7\pi^2/5)(k_B^2/e^2)$. The effect of a finite minimum transmission probability due, e.g., to incoherent processes or additional nonresonant channels, is also considered.

Electronic phase coherence underlies many of the unique and potentially revolutionary aspects of molecular device technology.¹ However, unequivocal experimental signatures of phase-coherent electron transport are difficult to obtain. Aharonov–Bohm interferometers have been used in quantum-dot^{2–4} and carbon nanotube⁵ studies both to determine the degree of coherence and to map the phase evolution of transmission by controlling the magnetic field and gate voltage, respectively. Phase information can also be extracted from asymmetric Fano line shapes arising from interference between resonant and nonresonant transport pathways;⁶ the degree of asymmetry is related to the degree of phase coherence, so an experiment needs to distinguish between a symmetric Breit–Wigner line shape and a slightly asymmetric one in order to demonstrate phase coherence.

A characteristic feature of coherent transport is the existence of nodes in the transmission spectrum.^{1,7} A transmission node can only arise from destructive quantum interference and as such constitutes a clear signature of coherent quantum transport in any system. Incoherent processes may also give rise to very low transmission probabilities, however, whose effect on the electrical conductance may be experimentally indistinguishable⁸ from that of a transmission node. In this Letter, we show that, in contrast to the electrical conductance, *thermoelectric effects are dramatically enhanced* in the vicinity of a transmission

node, thus providing a “smoking gun” for coherent transport that is testable in experiments on single-molecule junctions.

In order to investigate thermoelectric effects in single-molecule junctions, where Coulomb interactions play an essential role,⁹ we first derive an exact expression for the heat current in an interacting nanostructure. Using this result, together with the corresponding expression for the electrical current,¹⁰ we derive expressions for the linear thermoelectric response of the junction. Both the thermopower S and the Lorenz number L are calculated for three representative molecular junctions possessing transmission nodes within the HOMO–LUMO gap. The thermopower is predicted to reach a temperature-independent maximum value of $\pm\pi k_B/3^{1/2}e \approx \pm 156 \mu\text{V/K}$ near a transmission node, more than an order of magnitude larger than the values obtained in recent measurements^{11,12} of molecular junctions. Moreover, a 420% enhancement of the Lorenz number is also predicted at a transmission node. The dramatic enhancement of thermoelectric effects near a transmission node arises because the flow of entropy—an inherently incoherent quantity—is not blocked by destructive quantum interference.

We consider a junction consisting of a single molecule covalently bonded to M metallic electrodes (labeled $\alpha \in [1, \dots, M]$). The Hamiltonian of the system may be written

$$H_{\text{junction}} = H_{\text{mol}} + \sum_{\alpha=1}^M [H_{\text{lead}}^{(\alpha)} + H_{\text{T}}^{(\alpha)}] \quad (1)$$

where H_{mol} is the molecular Hamiltonian. Each electrode is modeled as a noninteracting Fermi gas

* Corresponding authors.

[†] College of Optical Sciences.

[‡] Department of Physics.

$$H_{\text{lead}}^{(\alpha)} = \sum_{\substack{k \in \alpha \\ \sigma}} \varepsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma}, \quad N_\alpha = \sum_{\substack{k \in \alpha \\ \sigma}} c_{k\sigma}^\dagger c_{k\sigma} \quad (2)$$

where $c_{k\sigma}^\dagger$ creates an electron of energy $\varepsilon_{k\sigma}$ in lead α . Tunneling of electrons between the molecule and electrode α is described by the Hamiltonian

$$H_T^{(\alpha)} = \sum_{k \in \alpha} \sum_{n, \sigma} (V_{nk} d_{n\sigma}^\dagger c_{k\sigma} + \text{H.c.}) \quad (3)$$

where $d_{n\sigma}^\dagger$ creates an electron of spin σ on the n th atomic orbital of the molecule.

The starting point for our derivation of the heat current is the fundamental thermodynamic identity at constant volume, $TdS = dE - \mu dN$. Applying the identity to electrode α , one finds

$$I_\alpha^Q \equiv T_\alpha \frac{dS_\alpha}{dt} = \frac{d}{dt} \langle H_{\text{lead}}^{(\alpha)} \rangle - \mu_\alpha \frac{d}{dt} \langle N_\alpha \rangle \quad (4)$$

where I_α^Q is the heat current flowing from the molecule into electrode α and T_α and μ_α are the temperature and chemical potential, respectively, of electrode α . The time derivatives on the right hand side of eq 4 may be evaluated using standard quantum mechanics to obtain

$$\begin{aligned} I_\alpha^Q &= -\frac{i}{\hbar} \{ \langle [H_{\text{lead}}^{(\alpha)}, H_{\text{junction}}] \rangle - \mu_\alpha \langle [N_\alpha, H_{\text{junction}}] \rangle \} \\ &= \frac{i}{\hbar} \sum_{k \in \alpha} \sum_{n, \sigma} (\varepsilon_{k\sigma} - \mu_\alpha) [V_{nk} \langle d_{n\sigma}^\dagger c_{k\sigma} \rangle - V_{nk}^* \langle c_{k\sigma}^\dagger d_{n\sigma} \rangle] \end{aligned} \quad (5)$$

The second line of eq 5 is similar to the corresponding formula for the electrical current,¹⁰ except for the additional factor of $\varepsilon_{k\sigma} - \mu_\alpha$ inside the sum. The correlation functions in the second line of eq 5 may be formally expressed in terms of the junction's Green's functions using Dyson's equation,¹⁰ leading to the general result

$$I_\alpha^{(\nu)} = -\frac{i}{\hbar} \int_{-\infty}^{\infty} dE (E - \mu_\alpha)^\nu \text{Tr} \{ \Gamma_\alpha(E) (G^<(E) + f_\alpha(E) [G(E) - G^+(E)] \} \} \quad (6)$$

where $\nu = 1$ gives the heat current and $\nu = 0$ the number current. Here $f_\alpha(E) = \{1 + \exp[(E - \mu_\alpha)/k_B T_\alpha]\}^{-1}$ and $[\Gamma_\alpha(E)]_{n\sigma, m\sigma'} = 2\pi \delta_{\sigma\sigma'} \sum_{k \in \alpha} V_{nk} V_{mk}^* \delta(E - \varepsilon_{k\sigma})$ are the Fermi–Dirac distribution and tunneling-width matrix, respectively, for lead α . $G(E)$ and $G^<(E)$ are Fourier transforms of the retarded and Keldysh “lesser” Green's functions, $G_{n\sigma, m\sigma'}(t) = -i\theta(t) \langle \{d_{n\sigma}(t), d_{m\sigma'}^\dagger(0)\} \rangle$ and $G_{n\sigma, m\sigma'}^<(t) = i \langle d_{m\sigma'}^\dagger(0) d_{n\sigma}(t) \rangle$, respectively. Equation 6 is an exact formal result and provides a complete framework for calculating heat transport and thermoelectric effects in interacting nanostructures.

In general, the retarded Green's function has the form⁹

$$G^{-1}(E) = G_{\text{mol}}^{-1}(E) - \Sigma_T - \Delta\Sigma_C \quad (7)$$

where G_{mol} may be evaluated by exact diagonalization of H_{mol} in the sequential-tunneling limit, and the self-energy terms $\Sigma_T + \Delta\Sigma_C$ describe the effects of finite tunneling width. Here the tunneling self-energy matrix

$$[\Sigma_T(E)]_{n\sigma, m\sigma'} = \delta_{\sigma\sigma'} \sum_{\alpha} \sum_{k \in \alpha} \frac{V_{nk} V_{mk}^*}{E - \varepsilon_{k\sigma} + i0^+} \quad (8)$$

can be calculated exactly, while the tunneling correction $\Delta\Sigma_C$ to the Coulomb self-energy can in general only be calculated approximately. However, in the middle of the HOMO–LUMO gap—the region of principal interest in this Letter—it can be shown⁹ that $\Delta\Sigma_C \approx 0$. Both $G(E)$ and $G^<(E)$ arise from time-ordered Green's functions on the Keldysh time-contour,¹³ so any prescription for calculating $G(E)$ also yields $G^<(E)$ without further approximations.

In many cases of interest in nanostructures, elastic processes dominate transport. This is the case if $\text{Im}\Delta\Sigma_C \approx 0$ and electron–phonon scattering is negligible. Then eq 6 may be simplified and cast in a form analogous to the multiterminal Büttiker formula^{14,15}

$$I_\alpha^{(\nu)} = \frac{1}{h} \sum_{\beta=1}^M \int_{-\infty}^{\infty} dE (E - \mu_\alpha)^\nu T_{\alpha\beta}(E) [f_\beta(E) - f_\alpha(E)] \quad (9)$$

where the transmission function is given by¹⁶

$$T_{\alpha\beta}(E) = \text{Tr} \{ \Gamma_\alpha(E) G(E) \Gamma_\beta(E) G^\dagger(E) \} \quad (10)$$

In linear response, the set of equations (9) (with $\nu = 0, 1$) may be further simplified and written in matrix form as

$$\begin{pmatrix} I_\alpha^{(0)} \\ I_\alpha^{(1)} \end{pmatrix} = \sum_{\beta} \begin{pmatrix} \mathcal{L}_{\alpha\beta}^{(0)} & \frac{1}{T} \mathcal{L}_{\alpha\beta}^{(1)} \\ \mathcal{L}_{\alpha\beta}^{(1)} & \frac{1}{T} \mathcal{L}_{\alpha\beta}^{(2)} \end{pmatrix} \begin{pmatrix} \mu_\beta - \mu_\alpha \\ T_\beta - T_\alpha \end{pmatrix} \quad (11)$$

where

$$\mathcal{L}_{\alpha\beta}^{(\nu)}(\mu, T) = \frac{1}{h} \int dE \left(-\frac{\partial f}{\partial E} \right) (E - \mu)^\nu T_{\alpha\beta}(E) \quad (12)$$

Here $f(E)$ is the equilibrium (zero bias) Fermi distribution with chemical potential μ and temperature T . We may then use the $\mathcal{L}$ functions to compactly encode a number of important transport properties¹⁷

$$G_{\alpha\beta}(\mu, T) = \frac{e^2}{h} \mathcal{L}_{\alpha\beta}^{(0)}(\mu, T) \quad (13)$$

$$S_{\alpha\beta}(\mu, T) = -\frac{1}{eT} \frac{\mathcal{L}_{\alpha\beta}^{(1)}(\mu, T)}{\mathcal{L}_{\alpha\beta}^{(0)}(\mu, T)} \quad (14)$$

$$\kappa_{\alpha\beta}(\mu, T) = \frac{1}{hT} \left(\mathcal{L}_{\alpha\beta}^{(2)}(\mu, T) - \frac{[\mathcal{L}_{\alpha\beta}^{(1)}(\mu, T)]^2}{\mathcal{L}_{\alpha\beta}^{(0)}(\mu, T)} \right) \quad (15)$$

where G is the electrical conductance, S is the thermopower (Seebeck coefficient), and κ is the electronic contribution to the thermal conductance. The linear-response transport coefficients of an interacting system thus have a structure identical to that of a noninteracting system, except that $T_{\alpha\beta}(E)$ must be calculated using the interacting Green's functions. It should be emphasized that eqs 13–15 can also be derived directly from eq 6, provided $\lim_{\Delta\mu \rightarrow 0} (\text{Im}\Delta\Sigma_C/\Delta\mu) = 0$ and $\lim_{\Delta T \rightarrow 0} (\text{Im}\Delta\Sigma_C/\Delta T) = 0$ (with similar conditions on the electron–phonon self-energy).

Panels a–c of Figure 1 show results for isoprene, 1,3-benzenedithiol, and [18]-annulene junctions, respectively. Each junction has a transmission node in the center of the HOMO–LUMO gap (see Figure 1, upper panels). Both the thermopower S (middle panels) and the Lorenz number $L = \kappa/GT$ (lower panels) are strongly enhanced in the vicinity of the midgap nodes. Although transmission nodes are generic features of coherent transport, midgap nodes are experimentally advantageous for two reasons: (i) When the lead chemical potential is in the gap, the molecule is charge-neutral, and (ii) the mismatch between the metal leads' Fermi energies and the center of the HOMO–LUMO gap is typically small (a few electronvolts or less), so available gating techniques¹⁸ should be sufficient to tune across the node.

In Figure 1, isoprene and benzenedithiol were modeled using a semiempirical π -electron Hamiltonian that accurately

describes Coulomb interactions and π -conjugation.^{1,9,19,20} [18]-Annulene was modeled using Hückel molecular orbital theory ($\Sigma_C = 0$). The interaction and hopping matrix elements were taken from ref 20. The tunneling-width matrix was taken in the broad-band limit²¹ whereby $\Gamma_{no,mo'}^\alpha(E) = \Gamma_\alpha \delta_{\sigma\sigma'} \delta_{na} \delta_{ma}$, where a is the π -orbital connected to lead α . The junctions were taken to be symmetric, with $\Gamma_1 = \Gamma_2 = 0.5$ eV. It should be emphasized, however, that the principal findings of this Letter depend only on the existence of transmission nodes and not on the specific form of the molecular junction Hamiltonian.

The dramatic enhancement of thermoelectric effects predicted at a transmission node arises because entropy transport is less sensitive to quantum interference than is charge transport. The entropy current I_a^0/T_α represents the flow of disorder, which quantum mechanically is related to the purity of a system's state where, for example, a pure state has zero entropy current. As such, the entropy current inherently involves incoherence and therefore cannot be completely blocked by destructive quantum interference, whereas the electrical current *can* be completely coherent and therefore *may* be completely blocked by destructive quantum interference. Since thermopower is the entropy per unit charge carried by an electrical current and the Lorenz number is the ratio of the entropy conductance to the electrical conductance, both are strongly enhanced at a transmission node.

Since the integrands in the numerator and denominator of eq 14 both include a thermal-averaging factor of $-\partial f/\partial E$, the thermopower does not diverge when the chemical potential is tuned across a transmission node but instead approaches a finite value which is *independent* of temperature (see Figure 1, middle panels). This latter point should be emphasized, since the thermopower also increases in mag-

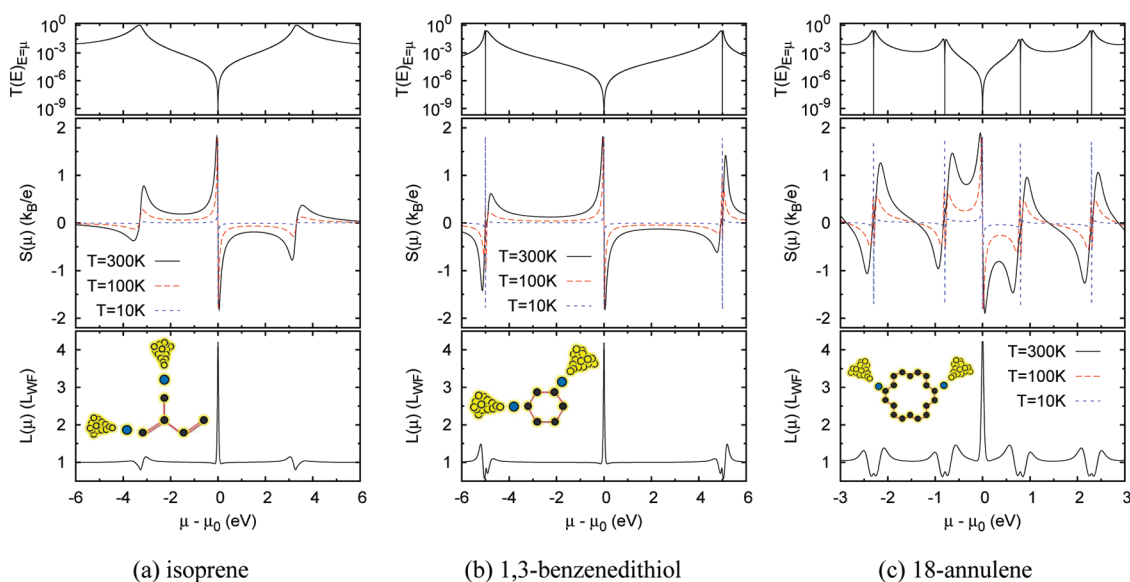


Figure 1. Transmission probability (upper panels, logarithmic scale), thermopower (middle panels), and Lorenz number (lower panels) as a function of lead chemical potential for three single-molecule junctions possessing transmission nodes at the center of the HOMO–LUMO gap, $\mu_0 \equiv (\epsilon_{LUMO} + \epsilon_{HOMO})/2$. Here $\Gamma_1 = \Gamma_2 = 0.5$ eV for all three junctions. The thermopower spectrum $S(\mu)$ is calculated at three different temperatures, demonstrating the invariance of the peak value $S_{\max} = \pi k_B/3^{1/2} e \approx 156 \mu V/K$ near the transmission node. The Lorenz number, given in units of $L_{WF} = (\pi^2/3) (k_B/e)^2$, peaks at a temperature-independent value of $21/5$ at the transmission node.

nitude and changes sign as the chemical potential crosses a transmission resonance.²² In the case of a resonance, however, the peak value is dependent upon both the temperature and lead-molecule coupling.⁹ From eq 14, it is evident that S is enhanced if $\mathcal{T}^{(1)}$ is large and $\mathcal{T}^{(0)}$ is finite (transmission resonance) or if $\mathcal{T}^{(0)}$ is small and $\mathcal{T}^{(1)}$ is finite (transmission node). Near a quadratic transmission node $T_{\alpha\beta}(E) \propto (E - \mu_0)^2$, it can be shown by direct evaluation of eq 14 that the thermopower reaches a universal maximum value²³ of $\pm S_{\max} = \pm \pi k_B / 3^{1/2} e \approx \pm 156 \mu\text{V/K}$, consistent with the numerical results shown in Figure 1. $S_{\max}$ is over an order of magnitude larger than the values obtained in recent measurements of single-molecule junctions.^{11,12}

The Lorenz number for each molecular junction is shown in the lower panels of Figure 1, normalized by the value predicted by the Wiedemann–Franz (WF) law

$$L_{\text{WF}} = \frac{\kappa_0 h}{T e^2} \equiv \frac{\pi^2}{3} \left(\frac{k_B}{e} \right)^2 \quad (16)$$

where $\kappa_0 = (\pi^2/3)(k_B^2 T/h)$ is the thermal conductance quantum.²⁴ For each molecule considered, the Lorenz number varies by tens of percent from the WF law as the chemical potential crosses an electronic resonance. This variation is comparable to the variation among different metals.²⁵ At a quadratic transmission node, the Lorenz number may be evaluated directly as

$$L_{\max} = \frac{1}{(eT)^2} \frac{\mathcal{T}_{12}^{(2)}(\mu, T)}{\mathcal{T}_{12}^{(0)}(\mu, T)} \Big|_{\mu=\mu_{\text{node}}} = \frac{7\pi^2}{5} \left(\frac{k_B}{e} \right)^2 \quad (17)$$

where we have assumed that the node is at least $k_B T$ away from any transmission peak. The peak Lorenz number is 420% larger than the WF value, several times greater than the variation near a transmission peak at low temperature. The peak Lorenz number is also universal, although the width of the peak depends on temperature.

Recent mean-field simulations suggest that tunneling through σ -orbitals may wash out any exact transmission nodes in small conjugated molecules.^{7,26} We can include the effects of any such additional nonresonant channels—or incoherent processes outside the scope of eq 10—phenomenologically by adding a small constant ε to the transmission probability: $T'_{12}(E) = T_{12}(E) + \varepsilon$. The dependence of the Lorenz number and thermopower on ε and temperature near the center of the HOMO–LUMO gap of an isoprene junction are shown in Figure 2 and Figure 3, respectively. Interestingly, we find that if a finite minimum transmission probability reduces the predicted enhancement of thermoelectric effects at low temperatures, the full effect is restored at sufficiently high temperature (see Figure 3). This behavior may be understood analytically by using $T'_{12}(E) = \gamma(E - \mu_0)^2 + \varepsilon$ in eq 12, which gives

$$S_{\max} = \pm \frac{\pi k_B}{\sqrt{3}e} [1 + (\tilde{T}/T)^2]^{-1/2} \quad (18)$$

and

$$\frac{L_{\max}}{L_{\text{WF}}} = \frac{21/5 + (\tilde{T}/T)^2}{1 + (\tilde{T}/T)^2} \quad (19)$$

where the crossover temperature is defined by $k_B \tilde{T} = (3\varepsilon/\gamma\pi^2)^{1/2}$. Here γ is a constant related to the specific resonance structure of the molecule and the lead-molecule couplings. If only the LUMO and HOMO levels contribute appreciably to the transport, then $\gamma \approx \Gamma_1 \Gamma_2 / 2\Delta^4$, where $\Delta = (\varepsilon_{\text{LUMO}} - \varepsilon_{\text{HOMO}})/2$. Larger molecules, such as [18]-annulene where σ -tunneling is negligible, are thus good candidates for experimental observation of the predicted enhancement of thermoelectric effects if σ -tunneling washes out transmission nodes in small conjugated molecules.

Although we have focused on predictions of the linear thermoelectric response of single-molecule junctions in this Letter, it should be emphasized that eq 6 is an exact result valid for arbitrarily large bias and temperature gradients applied to an interacting nanostructure. To analyze the performance of a nanoscale thermoelectric device, it is necessary to go beyond linear response theory. For example, eq 6 implies

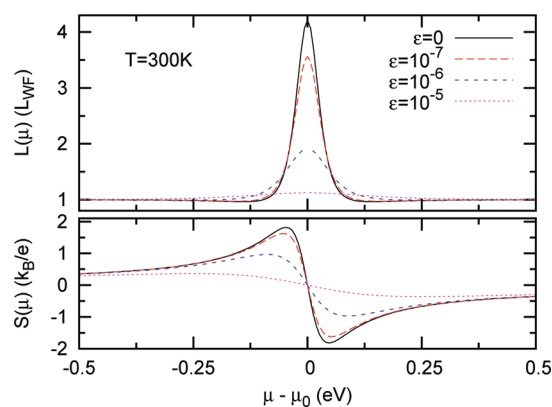


Figure 2. Lorenz number and Seebeck coefficient at $T = 300 \text{ K}$ near the middle of the HOMO–LUMO gap of an isoprene junction, for various values of the minimum transmission probability ε . Here $\Gamma_1 = \Gamma_2 = 0.5 \text{ eV}$.

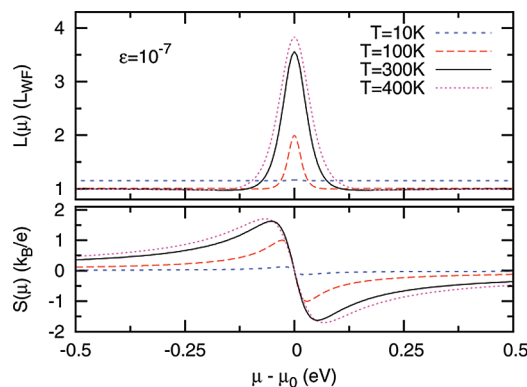


Figure 3. Lorenz number and Seebeck coefficient for an isoprene junction with $\varepsilon = 10^{-7}$ for various temperatures. Here $\Gamma_1 = \Gamma_2 = 0.5 \text{ eV}$.

$$\sum_{\alpha} I_{\alpha}^Q + \sum_{\alpha} \mu_{\alpha} I_{\alpha}^{(0)} = 0 \quad (20)$$

i.e., the total work done by the nanostructure against the external voltages equals the net heat flowing into the nanostructure. For a two-terminal heat engine with $T_2 > T_1$, the efficiency $\varepsilon = 1 - |I_{\alpha}^Q|/|I_{\alpha}^{(0)}|$, and it is necessary to go beyond linear response theory to evaluate this expression.

Acknowledgment. The authors thank P. Jacquod for useful discussions. This work was supported in part by AzRISE and a TRIF Photonics Graduate Fellowship.

Note Added after ASAP Publication: This paper was published ASAP on July 17, 2009. Equation 11 was updated. The revised paper was reposted on July 23, 2009.

References

- (1) Cardamone, D. M.; Stafford, C. A.; Mazumdar, S. *Nano Lett.* **2006**, *6*, 2422–2426.
- (2) Yacoby, A.; Heiblum, M.; Mahalu, D.; Shtrikman, H. *Phys. Rev. Lett.* **1995**, *74*, 4047–4050.
- (3) Schuster, R.; Buks, E.; Heiblum, M.; Mahalu, D.; Umansky, V.; Shtrikman, H. *Nature* **1997**, *385*, 417–420.
- (4) Avinun-Kalish, M.; Heiblum, M.; Zarchin, O.; Mahalu, D.; Umansky, V. *Nature* **2005**, *436*, 529–533.
- (5) Bachtold, A.; Strunk, C.; Salvetat, J.-P.; Bonard, J.-M.; Forró, L.; Nussbaumer, T.; Schönenberger, C. *Nature* **1999**, *397*, 673–675.
- (6) Clerk, A. A.; Waintal, X.; Brouwer, P. W. *Phys. Rev. Lett.* **2001**, *86*, 4636–4639.

- (7) Solomon, G. C.; Andrews, D. Q.; Goldsmith, R. H.; Hansen, T.; Wasielewski, M. R.; Van Duyne, R. P.; Ratner, M. A. *J. Am. Chem. Soc.* **2008**, *130*, 17301–17308.
- (8) Mayor, M.; Weber, H. B.; Reichert, J.; Elbing, M.; von Hänisch, C.; Beckmann, D.; Fischer, M. *Angew. Chem., Int. Ed.* **2003**, *42*, 5834–5838.
- (9) Bergfield, J. P.; Stafford, C. A. *Phys. Rev. B* **2009**, *79*, 245125.
- (10) Meir, Y.; Wingreen, N. S. *Phys. Rev. Lett.* **1992**, *68*, 2512–2515.
- (11) Reddy, P.; Jang, S.-Y.; Segalman, R. A.; Majumdar, A. *Science* **2007**, *315*, 1568–1571.
- (12) Baheti, K.; Malen, J.; Doak, P.; Reddy, P.; Jang, S.-Y.; Tilley, T.; Majumdar, A.; Segalman, R. *Nano Lett.* **2008**, *8*, 715–719.
- (13) Haug, H.; Jauho, A.-P. *Quantum Kinetics in Transport and Optics of Semiconductors*; Springer: Berlin, 1996; Vol. 123.
- (14) Büttiker, M. *Phys. Rev. Lett.* **1986**, *57*, 1761.
- (15) Sivan, U.; Imry, Y. *Phys. Rev. B* **1986**, *33*, 551–558.
- (16) Datta, S. *Electronic Transport in Mesoscopic Systems*; Cambridge University Press: Cambridge, U.K., 1995.
- (17) Finch, C. M.; García-Suárez, V. M.; Lambert, C. J. *Phys. Rev. B* **2009**, *79*, 033405.
- (18) Tao, N. J. *Nat. Nanotechnol.* **2006**, *1*, 173–181.
- (19) Chandross, M.; Mazumdar, S.; Liess, M.; Lane, P. A.; Vardeny, Z. V.; Hamaguchi, M.; Yoshino, K. *Phys. Rev. B* **1997**, *55*, 1486.
- (20) Castleton, C. W. M.; Barford, W. J. *Chem. Phys.* **2002**, *117*, 3570–3582.
- (21) Jauho, A.-P.; Wingreen, N. S.; Meir, Y. *Phys. Rev. B* **1994**, *50*, 5528.
- (22) Paulsson, M.; Datta, S. *Phys. Rev. B* **2003**, *67*, 241403.
- (23) Nakanishi, T.; Kato, T. *J. Phys. Soc. Jpn.* **2007**, *76*, 034715.
- (24) Rego, L. G. C.; Kirczenow, G. *Phys. Rev. B* **1999**, *59*, 13080–13086.
- (25) Ashcroft, N. W.; Mermin, N. D. *Solid State Physics*; Holt, Rinehart and Winston: New York, 1976.
- (26) Ke, S.-H.; Yang, W.; Baranger, H. U. *Nano Lett.* **2008**, *8*, 3257–3261.

NL901554S