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Letter

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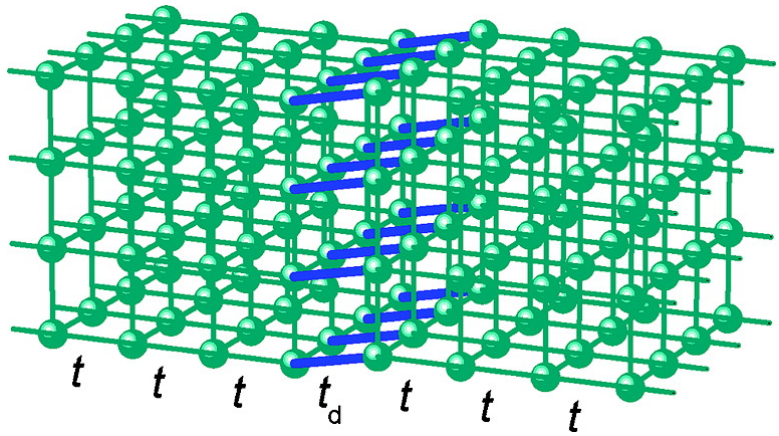
Nano Lett., **2008**, 8 (12), 4205-4209• DOI: 10.1021/nl801829w • Publication Date (Web): 29 October 2008



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NANO LETTERS

2008
Vol. 8, No. 12
4205-4209

Renormalization Plus Convolution Method for Atomic-Scale Modeling of Electrical and Thermal Transport in Nanowires

Chumin Wang* and Fernando Salazar†

Instituto de Investigaciones en Materiales, Universidad Nacional Autónoma de México, Apartado Postal 70-360, 04510, D.F., México

Vicenta Sánchez

Departamento de Física, Facultad de Ciencias, Universidad Nacional Autónoma de México, Apartado Postal 70-542, 04510, D.F., México

Received June 25, 2008; Revised Manuscript Received October 10, 2008

ABSTRACT

Based on the Kubo−Greenwood formula, the transport of electrons and phonons in nanowires is studied by means of a real-space renormalization plus convolution method. This method has the advantage of being efficient, without introducing additional approximations and capable to analyze nanowires of a wide range of lengths even with defects. The Born and tight-binding models are used to investigate the lattice thermal and electrical conductivities, respectively. The results show a quantized electrical dc conductance, which is attenuated when an oscillating electric field is applied. Effects of single and multiple planar defects, such as a quasi-periodic modulation, on the conductance of nanowires are also investigated. For the low temperature region, the lattice thermal conductance reveals a power-law temperature dependence, in agreement with experimental data.

The continuing work toward ever smaller devices has given rise to an intensive investigation of nanoscale electronics, which permits a lower power consumption, higher packing density, and possible quantum performance, when the size is smaller than its dephasing length. In such devices, nanowires constitute a basic building block. Also, the capability of nanowires to connect the molecules of living body suggests that they may have a broad biological utility.[1] For all these applications, an accurate modeling of the transport properties of nanowires is very helpful in the design of nanodevices.

A nanowire is an almost one-dimensional structure with unique electrical, optical, and transport properties, mainly due to its lateral confinement on the excitations, and consequently quantum behaviors are further evident. For instance, quantized electrical[2] and thermal[3] conductance of nanowires have been observed. However, these quantum behaviors are highly sensitive to the defects,[4] and their presence is in principle inevitable. Hence, it is of singular interest to quantify their effects. In particular, planar defects, made by a twin of several atomic planes, have been frequent in nanowires.[5] In this Letter, we report a study of the transport properties in cubically structured nanowires by using a previously developed real-space renormalization plus convolution method,[6] which allows an accurate evaluation of the Kubo−Greenwood formula having a high computa-

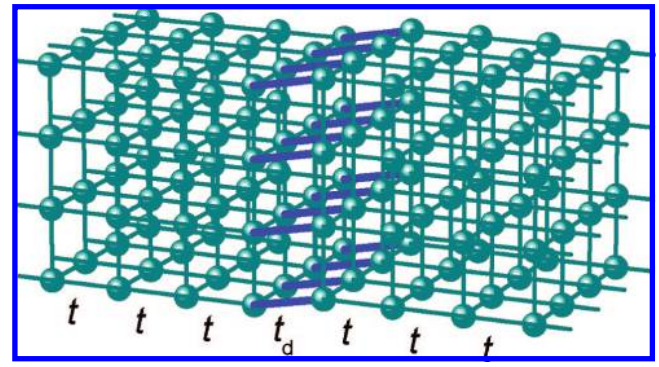

Figure 1. Schematic illustration of a nanowire segment around a planar twist defect depicted by the hopping integral t_d (thick blue lines). This cubically structured nanowire has a cross section of 4 × 4 atoms.

* Corresponding author. E-mail: chumin@servidor.unam.mx.
† Current address: Departamento de Física-Química, Instituto de Física, Universidad Nacional Autónoma de México, Apartado Postal 20-364, 01000, D.F., México.

10.1021/nl801829w CCC: $40.75
Published on Web 10/29/2008

tional efficiency.[7] Furthermore, its real-space character permits the modeling of nanowires with a wide range of lengths, as well as the presence of single and multiple planar defects, such as quasi-periodically arranged ones. For the electron case, a tight-binding Hamiltonian is employed, while the Born model is used to describe the normal vibration modes. The results of these semiempirical models without fitting any parameters show a good qualitative agreement with experimental data.

Modeling Electronic Transport. Let us consider a simple s-band tight-binding Hamiltonian ($\hat{H}$) with null on-site energies given by[8]

$$\hat{H} = \sum_{\langle i,j \rangle} \{ t_{i,j} |i\rangle\langle j| + t_{j,i} |j\rangle\langle i| \} \quad (1)$$

where $t_{i,j} = t_{j,i}$ is the hopping integral between nearest-neighbor atoms i and j, denoted by $\langle i,j \rangle$. For the sake of simplicity, a uniform nearest-neighbor bond length ($a = 1$) is taken.

The electrical conductivity along the nanowires (σ_{xx}) can be calculated by means of the Kubo−Greenwood formula[9]

$$\sigma_{xx}(\mu, \varpi, T) = -\frac{2e^2\hbar}{\pi m^2 \Omega} \int_{-\infty}^{\infty} \mathrm{d}E \frac{f(E+\hbar\varpi) - f(E)}{\hbar\varpi} Tr[\hat{p}_x \tilde{G}(E+\hbar\varpi) \hat{p}_x \tilde{G}(E)] \quad (2)$$

where e and m are the electrical charge and the mass of electron, respectively, Ω is the system volume, and $f(E) = \{ 1 + \exp[(E-\mu)/k_B T] \}^{-1}$ is the Fermi−Dirac distribution with the Fermi energy μ, the Boltzmann constant k_B, and temperature T.

$$\hat{p}_x = \frac{im}{\hbar}[\hat{H}, x] = \frac{ima}{\hbar} \sum_j \{ t_{j,j+1} |j\rangle\langle j+1| - t_{j-1,j} |j\rangle\langle j-1| \}$$

is the projection of the momentum operator along the oscillating electrical field with frequency ϖ, and $\tilde{G}(E) \equiv G^+(E) - G^-(E)$ is the discontinuity of Green's functions, being $G^+(E)$ and $G^-(E)$ the retarded and advanced one-particle Green's functions, respectively.[8]

The trace in eq 2 at $\varpi = 0$, defined as $\mathrm{Trace}(E) \equiv \mathrm{Tr}[\hat{p}_x \tilde{G}(E) \hat{p}_x \tilde{G}(E)]$, for a one-dimensional (1D) periodic chain is given by[6]

$$\mathrm{Trace}^{1D}(E) = Tr[\hat{p}_x \tilde{G}^{1D}(E) \hat{p}_x \tilde{G}^{1D}(E)] = \frac{(N_{||}-1)^2 m^2 a^2}{2\hbar^2} \quad (3)$$

which is a constant for $|E| \leq 2|t|$ due to its ballistic nature. In eq 3, $N_{||}$ is the number of atoms along the applied electrical field. For a cubically structured nanowire with planar defects as shown in Figure 1, the Hamiltonian of eq 1 is separable, i.e., $\hat{H} = \hat{H}_{||} \otimes \hat{I}_{\perp} + \hat{I}_{||} \otimes \hat{H}_{\perp}$, being $\hat{H}_{||}$ ($\hat{I}_{||}$) and $\hat{H}_{\perp}$ ($\hat{I}_{\perp}$) the Hamiltonian (the identity of the corresponding Hilbert space) of the parallel and perpendicular subsystem with respect to the applied electric field, respectively.[10] Therefore, the convolution theorem can be expressed as[6]

$$\mathrm{Trace}(E) = \sum_v \mathrm{Trace}^{||}(E - E_v) \quad (4)$$

or

$$\sigma_{xx}(\mu, \varpi, T) = \frac{1}{\Omega_{\perp}} \sum_v \sigma_{xx}^{||}(\mu - E_v, \varpi, T) \quad (5)$$

where $\mathrm{Trace}^{||}$ and $\sigma^{||}$ are the trace and conductivity of the parallel subsystem, respectively, and $\Omega_{\perp}$ and E_v are the volume and eigenvalues of the perpendicular subsystem, i.e., $\hat{H}_{\perp}|v\rangle = E_v|v\rangle$. In general, the dc electrical conductivity is proportional to Trace.

In Figure 1, a segment of the nanowire with a cross section of 4×4 atoms is schematically illustrated, where a planar defect is represented by the hopping integral t_d (thick blue lines). For the case of multiple defects, t_d occurs at many planes along the nanowire, and in this case study, they are arranged following the Fibonacci sequence[11] [$\mathscr{F}(n)$] of generation n, i.e.,

$$\mathscr{F}(1) = \bullet \overset{t_d}{-} \bullet, \quad \mathscr{F}(2) = \bullet \overset{t_d}{-} \bullet \overset{t}{-} \bullet$$

and $\mathscr{F}(n) = \mathscr{F}(n-1) \oplus \mathscr{F}(n-2)$, where $\bullet$ symbolizes the atomic positions. For example

$$\mathscr{F}(4) = \bullet \overset{t_d}{-} \bullet \overset{t}{-} \bullet \overset{t_d}{-} \bullet \overset{t_d}{-} \bullet \overset{t}{-} \bullet$$

and then, a nanowire with a cross section of 4×4 atoms has 16 interconnected Fibonacci chains and it is additionally attached at each extreme of this nanowire 16 interconnected semi-infinite periodic leads with null on-site energy and hopping integral t.

Lattice Thermal Conductivity. For the case of phonons, let us consider an interaction potential (V_{ij}) between nearest-neighbor atoms i and j within the Born model[12]

$$V_{ij} = \frac{1}{2}(\alpha - \beta)\{ [\mathbf{u}(i) - \mathbf{u}(j)] \cdot \hat{\mathbf{n}}_{ij} \}^2 + \frac{1}{2}\beta [\mathbf{u}(i) - \mathbf{u}(j)]^2 \quad (6)$$

where $\mathbf{u}(i)$ is the displacement of atom i with respect to its equilibrium position and α and β are central and noncentral restoring force constants, respectively. The unitary vector $\hat{\mathbf{n}}_{ij}$ indicates the bond direction between i and j atoms. The Dyson equation for Green's function of phonons ($\mathbf{G}$) is[13]

$$(M\omega^2 \mathbf{I} - \mathbf{\Phi}) \mathbf{G}(\omega) = \mathbf{I} \quad (7)$$

where M is the atomic mass, $\mathbf{I}$ stands the matrix identity, and $\mathbf{\Phi}$ is the dynamic matrix, whose elements are

$$\Phi_{\mu\mu'}(i,j) = \frac{\partial^2 V_{ij}}{\partial u_{\mu}(i)\, \partial u_{\mu'}(j)} \quad (8)$$

being $\mu, \mu' = x, y,$ or z. It is worth mentioning that the Born model is widely used in the description of many semiconductor materials, despite its apparent simplicity and reduced fitting parameters.[14] Furthermore, for cubically structured nanowires $\mathbf{\Phi}$ is separable, as occurred in the electron case.[15]

The lattice thermal conductivity (κ) can be calculated by means of the Kubo−Greenwood formula given by[13,16]

$$\kappa_{xx}(T) = \frac{-4\hbar^2}{\Omega k_B T^2} \sum_l \frac{1}{2\pi} \int_0^{\infty} \mathrm{d}\omega \frac{\omega^2 \exp(\hbar\omega/k_B T)}{[\exp(\hbar\omega/k_B T) - 1]^2} Tr[\mathbf{A}_x \tilde{G}(\omega) \mathbf{A}_x \tilde{G}(\omega)]_l \quad (9)$$

where the sum of l is over the longitudinal and transversal modes and $\tilde{\mathbf{G}}(\omega)$ is the discontinuity of the phononic Green's function defined as in the electron case.[8] The elements of

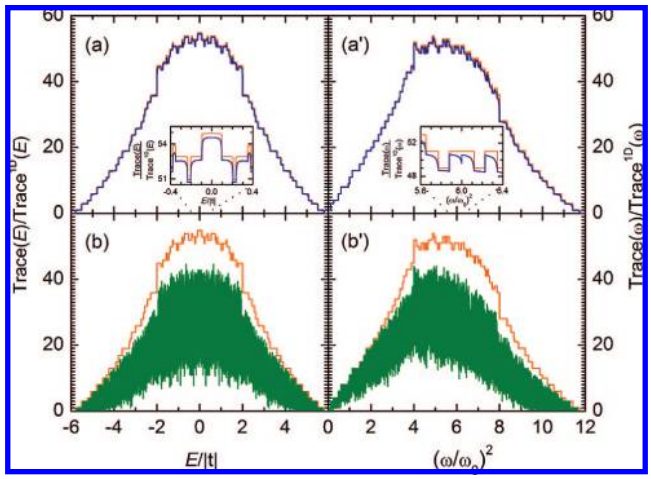

Figure 2. (a, b) Trace(E) and (a′, b′) Trace(ω) for nanowires with a single planar defect (blue lines) and quasi-periodically arranged defects (green lines), in comparison with the periodic case (orange lines), being $t_d = 0.95t$, $\alpha_d = 0.95\alpha$, and $\eta = 10^{-11}|t|$ or $10^{-11}\omega_0^2$. These nanowires contain $433494438 \times 9 \times 9$ atoms.

matrix $\mathbf{A}_x$ are $[\mathbf{A}_x]_{\mu\mu'}(i,j) = {}^1/_2(\mathbf{R}_i - \mathbf{R}_j)_x\Phi_{\mu\mu'}(i,j)$, where $\mathbf{R}_i$ is the equilibrium position of atom i.

It is easy to find that for a periodic chain the trace of a single mode in eq 9, Trace(ω) $\equiv Tr[\mathbf{A}_x\tilde{\mathbf{G}}(\omega)\mathbf{A}_x\tilde{\mathbf{G}}(\omega)]$, is

$$\text{Trace}^{1D}(\omega) = Tr[\mathbf{A}_x\tilde{G}^{1D}(\omega)\mathbf{A}_x\tilde{G}^{1D}(\omega)] = -\frac{(N_{||}-1)^2a^2}{8} \quad (10)$$

similar to the electron case of eq 3. Despite that the renormalization plus convolution method can be applied to both longitudinal and transversal modes, numerical results of the lattice thermal conductivity are only shown in this Letter for the longitudinal mode, where the studied nanowires are connected to two semi-infinite periodic leads with the same cross section. Single and multiple defects are introduced by α_d maintaining constant M, as occurred in the electron case. The nanowires with and without defects studied in this Letter have a length of 433494438 atoms corresponding to the Fibonacci sequence $n = 42$ built by adding those of $n = 41$ and $n = 40$, where the single planar bond defect is introduced at this interface.

Results. Calculations of both electrical and lattice thermal conductance of nanowires are carried out by using the renormalization plus convolution method,[6] i.e., the single-channel conductivity is calculated by means of a real-space renormalization procedure for each given E_v and this conductivity is summed through the convolution technique given by eq 5, which is particularly useful when the cross section has a reduced number of atoms, as is the case of a nanowire.

In panels a and b of Figure 2, Trace(E) is illustrated for nanowires without defects (orange line) and with single (blue line) and multiple quasi-periodically ordered defects (green line). Similarly, in parts a′ and b′ of Figure 2 Trace(ω) for the phonon case with $\beta = \alpha$ is plotted, being $\omega_0 = (\alpha/M)^{1/2}$. In the insets of parts a and a′ of Figure 2, magnifications of the spectra are shown. Nanowires used in these figures have $433494438 \times 9 \times 9$ atoms and their defects are characterized by $t_d = 0.95t$ or $\alpha_d = 0.95\alpha$. The imaginary part (η) of E or ω^2 in Green's function is $\eta = 10^{-11}\,|t|$ or $10^{-11}\omega_0^2$. Observe

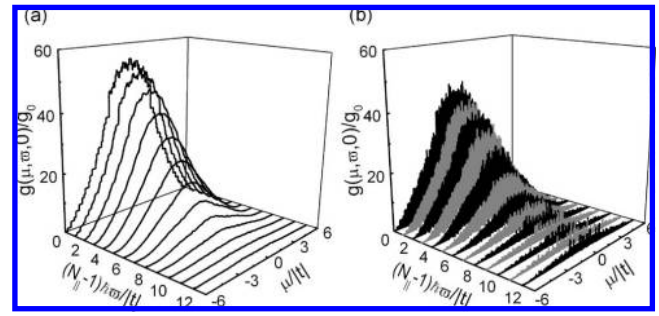

Figure 3. The electrical conductance at $T = 0$ [$g(\mu,\varpi,0)$] in units of $g_0 = 2e^2/h$ versus the Fermi energy (μ) and the frequency of applied electric field (ϖ) for nanowires with (a) a single and (b) multiple planar defects, having the same parameters as in Figure 2, parts a and b, respectively.

the perfect shape of quantum steps in the periodic case (orange line), which is originated by adding the constant single-channel trace around each E_v, as shown in eqs 3 and 4. For example, a cross section of four identical atoms has eigenvalues $E_1 = -2|t|$, $E_2 = E_3 = 0$, and $E_4 = 2|t|$ and then, Trace(E) of a periodic nanowire with this cross section has steps of single height at $E = \pm 4|t|$ and double height at $E = \pm 2|t|$. In general, the nonuniform step heights in the dc conductivity spectrum, as observed in nanowires of gold and nickel,[2] could be related to the degeneracy of E_v, i.e., a three-dimensional character of nanowires. For the case of defects, their presence diminishes the conductivity and softens the edge of steps. Also, notice that the spectra for the electron case have a mirror symmetry with respect to $E = 0$, because the nanowire structure is bipartite for the electronic Hamiltonian given by eq 1. For the phonon case, we observe that the optical vibration modes are more sensitive to the presence of defects. When quasi-periodically arranged defects are introduced, both spectra of electron and of phonon, the green lines in parts b and b′ of Figure 2, become spiky with a significantly reduced average valor.

The electrical conductance (g) is defined as $g(\mu,\varpi,T) = \sigma(\mu,\varpi,T)\Omega_\perp/\Omega_{||}$, where $\Omega_{||}$ is the volume of parallel subsystem. Parts a and b of Figure 3 show the *ac* electrical conductance at $T = 0$ versus μ and ϖ for the same nanowires with defects of parts a and b of Figure 2, respectively. Observe that when an oscillating electric field is applied, the quantized steps in units of $g_0 = 2e^2/h$ are quickly destroyed and g shows a general diminishing behavior as ϖ grows. Nevertheless, nanowires with quasi-periodic defects have a less reduced g for high values of ϖ, showing resonance peaks at several energies. A detailed study of the quasi-periodicity effects on these *ac* resonance peaks is currently carried out.[17]

On the other hand, the lattice thermal conductance is defined as $K(T) = \kappa(T)\Omega_\perp/\Omega_{||}$ and it can be normalized by the quantum of thermal conductance[18] [$K_0 = \pi k_B^2 T/(6\hbar)$] for a single mode at $T_0 = \hbar_0\omega_0/k_B$, i.e., $K_0 = \pi k_B\omega_0/6$. In Figure 4a, the temperature dependence of K is shown for linear chains of 433494438 atoms without defects (open orange circles) and with single (open blue triangles) and quasi-periodically arranged defects (open green squares). A magnification of the low-temperature region is illustrated in

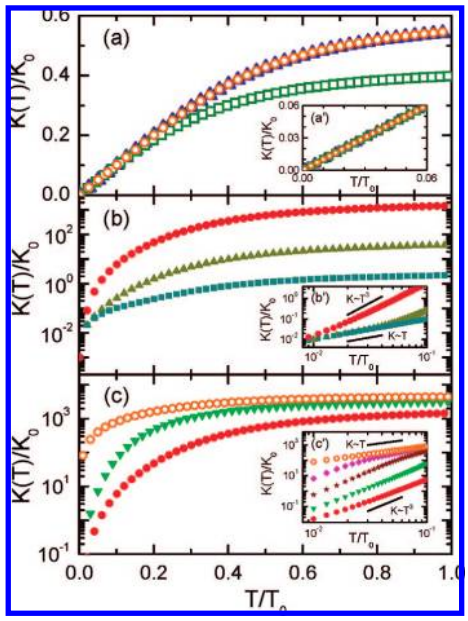

Figure 4. Lattice thermal conductance (K) as a function of temperature (T) for (a) chains without (open orange circles), with single (open blue triangles), and multiple (open green square) defects; (b) periodic nanowires with $\beta = \alpha$ and cross sections of 3 × 3 (cyan solid squares), 14 × 14 (yellow solid triangles), and 90 × 90 (red solid circles) atoms; and (c) periodic nanowires with a cross section of 90 × 90 and $\beta = \alpha$ (red solid circles), $\beta = 0.1\alpha$ (green solid triangles) and $\beta = 0$ (open orange circles). Insets: The corresponding low-temperature behaviors of K. Cases of $\beta = 0.01\alpha$ (wine solid stars) and $\beta = 0.001\alpha$ (magenta solid rhombuses) are also presented in (c′).

the inset, Figure 4a′, where a unique linear dependence is observed for the three cases, since the lattice thermal conduction at low temperatures is mainly through long-wavelength acoustic phonons and they are very lightly affected by the presence of defects. Moreover, Figure 4b shows $K(T)$ of periodic nanowires with cross sections of 3 × 3 (cyan solid squares), 14 × 14 (yellow solid triangles), and 90 × 90 (red solid circles) atoms. In its inset, Figure 4b′, a log−log plot for the low-temperature region is presented, where a power-law temperature dependence (T^d) is observed. There is a transition from $d = 1$ to $d = 3$, and the transition temperature diminishes when the cross section increases. In fact, at the zero-temperature limit, all the nanowires have linear temperature dependence in the lattice thermal conductance. This transition has been observed in silicon nanowires.[19] It is worth mentioning that the umklapp phonon dispersion process is not considered in eq 9 and that the reduction of lattice thermal conductance at high-temperature region is absent in this model.

Finally, Figure 4c shows $K(T)$ of periodic nanowires with a cross section of 90 × 90 atoms and $\beta = \alpha$ (red solid circles), $\beta = 0.1\alpha$ (green solid triangles), and $\beta = 0$ (open orange circles). A log−log plot for the low-temperature region is illustrated in the inset, Figure 4c′, where two additional curves of $K(T)$ are presented for $\beta = 0.01\alpha$ (wine solid stars) and $\beta = 0.001\alpha$ (magenta solid rhombuses). Observe that for $\beta = 0$, $K(T)$ is 8100 times the value of a single periodic chain (see Figure 4a) and in general, $K(T)$ diminishes when β grows, due to the interference between conducting channels in the nanowire. Indeed, as shown in Figure 4c′, the nanowire with $\beta = 0$ has a one-dimensional (1D) behavior ($K \sim T$) and those with $\beta > 0.01\alpha$ have an almost three-dimensional (3D) behavior ($K \sim T^3$). The transition between 1D and 3D behaviors seems to occur around $\beta = 0.001\alpha$; i.e., a small coupling between conducting channels could induce a qualitative change of $K(T)$ in the low-temperature region.

Conclusions. We have presented a comparative study of the electrical and lattice thermal conductivities by means of the Kubo−Greenwood formula. Despite the well-known similarity in Green's functions of electrons and of phonons within tight-binding type models,[8] the response of both conductivities to the presence of defects is qualitatively different mainly due to their fermionic and bosonic nature. In the case of phonons, the long-wavelength acoustic vibration modes are essentially unaffected by even multiple defects and they are the main responsible of the low-temperature lattice thermal conductivity. However, the dc electrical conductivity at low temperature is carried out by electrons around the Fermi energy and these electrons are strongly scattered by defects due to their relatively short wavelength. This difference in the scattering process is even enhanced when the system has a few transport channels, such is the case of nanowires.

Two main results obtained in this study are the nonuniformly quantized dc electrical conductance and the transition from $d = 1$ to $d = 3$ in the power-law temperature dependence (T^d) of the lattice thermal conductance at low-temperature region. Both results have been partially verified in real nanowires, revealing its three-dimensional character. It would be worth mentioning that actually the exponent d is the dimensionality of system;[20] for example, the quantum of thermal conductance has a linear temperature dependence for a 1D periodic system, which can be obtained by using eqs 9 and 10.

Concerning the renormalization plus convolution method, it is capable to evaluate the Kubo−Greenwood formula in nanowires of a wide range of lengths and even containing planar defects. The convolution theorem expressed in its summation form (see eq 5) is particularly useful for nanowires due to the reduced number of eigenvalues in the sum. In fact, the convolution theorem offers an unambiguous manner to understand the quantum steps in the dc electrical conductance spectrum of periodic nanowires, by adding rectangular ballistic conductance spectra for each given eigenvalue of the perpendicular subsystem. Finally, it is important to stress that this method can also be applied to noncubically structured lattices; for example, honeycomb atomic lattices can be mapped to a squarelike one by considering that each new lattice point represents four atoms. This example could be particularly important for the study of carbon nanotubes.

Acknowledgment. This work has been partially supported by CONACyT-58938, UNAM- IN111507, and IN114008. Computations were performed at Bakliz and Kanbalam of DGSCA, UNAM.

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NL801829W