

The role of solitons on the properties of electron transport through the CNT/ $t$ -PA/CNT system—a Green's function approach

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2007 J. Phys.: Condens. Matter 19 116211

(<http://iopscience.iop.org/0953-8984/19/11/116211>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 129.252.86.83

The article was downloaded on 28/05/2010 at 16:36

Please note that [terms and conditions apply](#).

# The role of solitons on the properties of electron transport through the CNT/*t*-PA/CNT system—a Green's function approach

H Milani Moghaddam<sup>1,2,3,5</sup>, S A Ketabi<sup>3,4</sup> and N Shahtahmasebi<sup>1,3</sup>

<sup>1</sup> Department of Physics, Ferdowsi University of Mashhad, Mashhad, Iran

<sup>2</sup> Department of Physics, University of Mazandaran, Babolsar, Iran

<sup>3</sup> Nanotechnology Research Center of the Ferdowsi University of Mashhad, Mashhad, Iran

<sup>4</sup> School of Physics, Damghan University of Basic Sciences, Damghan, Iran

E-mail: [hossainmilani@yahoo.com](mailto:hossainmilani@yahoo.com)

Received 30 November 2006, in final form 7 February 2007

Published 5 March 2007

Online at [stacks.iop.org/JPhysCM/19/116211](http://stacks.iop.org/JPhysCM/19/116211)

## Abstract

Using a tight-binding model and methods based on Green's function theory and Löwdin's partitioning technique, we numerically investigate the effects of the coupling strength, the role of the solitons and the presence of a linear applied voltage drop across the molecule on the electronic transmission through a system in which *trans*-polyacetylene (*t*-PA) as the molecule is sandwiched between two semi-infinite single-walled carbon nanotubes (CNT). We rely on *Landauer* formalism as the basis for studying the conductance properties of this system. Our calculations show that the solitons play an important role in this system, causing a large enhancement in the conductance. Also, our results suggest that the conductance is sensitive to the CNT/molecule coupling strength, and the presence of an applied voltage considerably increases the conductance.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

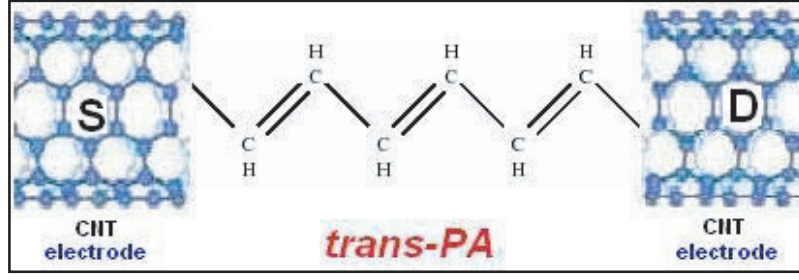
The study and manipulation of matter on the nanometre scale is a thriving area of research, with profound implications for technology (e.g. nanoelectronics, nanostructured materials, nanobiology). One of the most attractive research areas has been focused on the use of molecular system as electronic devices. So devices have been designed in such way that a molecule is sandwiched between two electrodes (metallic or organic). Considering the immense progress in the field of nanofabrication, attempts have also been made towards other

<sup>5</sup> Author to whom any correspondence should be addressed.

structures, such as CNTs, as conducting metallic reservoirs. In some recent works, simple molecular wires have been connected to two semi-infinite CNTs [1–3]. The significantly improved switching characteristics of short organic FETs with metallic CNT electrodes over those with metal electrodes are attributed to the excellent electrostatics attainable with a nanotube electrode geometry [4, 5]. The most important basic question in this context, therefore, is: what determines the conductance of such a molecule if it is used as a current-carrying element bridging two reservoirs of differing electron chemical potential? This question has an immediate relevance for experiments in which such conductance is measured, using either (i) scanning probe tips for individual molecules adsorbed on surfaces [6, 7] for molecular wires adsorbed at step edges [8] embedded in self-assembled monolayers [9, 10] or (ii) (macroscopic) electrodes obtained by nanolithography [11, 12] or from a mechanically controllable break junction [13]. So far, many theoretical works have been developed and models have been proposed to represent the molecular system and the reservoirs. In most model systems, the molecular wires connected to two semi-infinite surfaces [13, 14] or to two semi-infinite ‘rods’ [15] or to clusters [16, 17]. Within a framework equivalent to the latter model, calculations have been extended to the Hartree–Fock level for a molecule attached to gold clusters [18]. Also, density-functional theory has been applied to a molecular wire (described by atomic pseudo-potentials) connected to two jellium surfaces [19]. These theoretical studies show some of the important parameters that affect the conductance of the system. First, they have shown the importance of the electronic and chemical interactions between the ends of molecular wire and the reservoirs. Second, they show that, in the limit of a small applied voltage and away from the Coulomb blockade regime, the transport is dominated by charge carrier tunnelling inside the highest occupied molecular orbital (HOMO)–lowest unoccupied molecular orbital (LUMO) gap of the molecule. This gap is a crucial parameter for control of the conductance of the wire. The smaller gap gives rise to the larger tunnelling transmission.

Following these interests and towards the modelling of a full carbon transistor, we present a model in which a *t*-PA molecule is connected to two semi-infinite armchair CNTs. The possibility of a semiconductor-to-metal transition and the capability for becoming a very conductive material have been the reasons for the experimental and theoretical investigations on the physical properties of *t*-PA in comparison with other conducting polymers [20]. Several studies of the structural, electronic and conduction properties of *t*-PA have been published [21–23]. In *t*-PA, as a conjugated polymer, due to the Peierls transition, there is a strong coupling of the  $\pi$ -electrons occupying the HOMO and LUMO states to the bond-alternation pattern. This coupling means that the low-lying states of a charged molecule (via which any net transport of charge through the molecule must proceed) involve an intimate coupling of electronic and lattice degrees of freedom, to produce excitations such as solitons [20, 24]. These coupled excitations can be thought of as conspiring to lower the energy gap locally around a charge carrier when it is introduced into the system. The polaronic and solitonic phenomena have been studied for decades in bulk or thin-film samples of conducting polymers [20, 25].

We investigate numerically the conductance properties of *t*-PA molecules in the CNT/*t*-PA/CNT structure (figure 1), where the nanocontacts are considered to be the armchair ( $l, l$ ) single-walled carbon nanotubes. In general, there are  $M$  atomic positions over the interfacial end-atoms of the tubes. For an armchair where ( $l, l$ ) CNT topology is imposed, the number of carbon sites at the interface is  $M = 2l$ . In addition, we make further studies of the conductance properties of the systems in the presence of the solitons, and show its effect on the energy gap of the molecule. The model and the description of the methods for investigating the conductance properties of the molecular wire are introduced in section 2. The results and discussion are presented in section 3, followed by a summary in section 4.



**Figure 1.** A schematic representation of the CNT/*t*-PA/CNT structure, as described in the text.

## 2. Methodology

The most commonly used computational schemes for calculating the (coherent) conductance  $g$  are the *Landauer* theory [26] and the Green's function formalism [27–29].

The conductance  $g$  at zero temperature is simply proportional to the transmission coefficient,  $T(E)$ , for injected electrons at the Fermi energy:

$$g = g_0 T(E); \quad g_0 = \frac{2e^2}{h}. \quad (1)$$

The transmission coefficient can be calculated from the knowledge of the molecular energy levels, the nature and the geometry of the contacts. Using the Fisher–Lee relation [30], the conductance can be expressed as,

$$g = g_0 |G_{1N}(E)|^2 \Delta_1(E + eV) \Delta_N(E), \quad (2)$$

where  $G_{1N}(E)$  is the  $(1, N)$  element of the retarded Green's function matrix of the molecule.  $\Delta_1 (= \Delta_N)$ , the spectral density of the left (right) CNT electrode, is related to the semi-infinite left (right) electrode Green's function matrix,  $g^{L(R)}$ . This is minus the imaginary part of the CNT self-energies (per spin):

$$\Sigma^\alpha = \Lambda^\alpha - i\Delta^\alpha = \sum_{m_\alpha, m'_\alpha} (t_c)_{m_\alpha} (t_c)_{m'_\alpha} g_{m_\alpha, m'_\alpha}^\alpha; \quad \alpha = L(R). \quad (3)$$

$m_{L(R)}$  and  $m'_{L(R)}$  run over interfacial end-atoms of the CNTs.  $t_c$  is the coupling strength of the CNT/molecule, and the single-particle Green's function  $g_{m_\alpha, m'_\alpha}^\alpha$  can be expressed as [31],

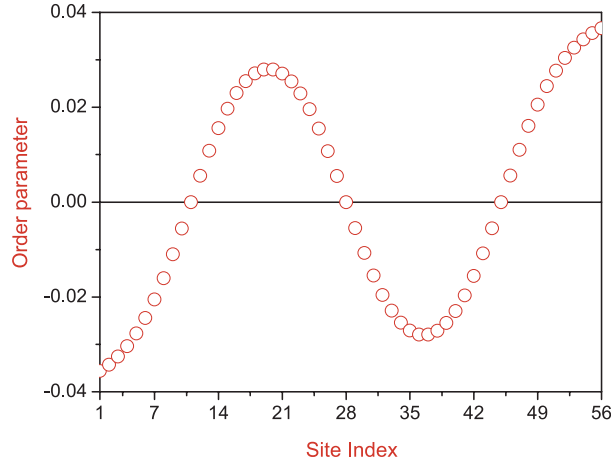
$$g_{n_y, n'_y}(E) = \frac{1}{2\ell} \sum_{j=1}^{2\ell} \varphi_j(n_y) \tilde{G}^j(E) \varphi_j(n'_y), \quad (4)$$

where  $\varphi_j(n_y) = \exp(ik_y^j n_y a)$ , with  $k_y^j a = \pi j \ell$ ,  $1 \leq j \leq 2\ell$  and  $a$  is the lattice space. We assume the  $x$  direction to be parallel to the tube (and to the transport direction) and  $y$  to be the finite transverse coordinate.  $\tilde{G}(E)$  is the surface Green's function, and is given by [31],

$$\tilde{G}^j(E) = \frac{1}{2t_{\text{CNT}}} \frac{E - \varepsilon}{2t_{\text{CNT}}} \left( 1 + i \frac{\sin\left(\frac{q_{\beta^*}^j a}{2}\right)}{\sqrt{\left(\frac{E - \varepsilon}{2t_{\text{CNT}}}\right)^2 - \sin^2\left(\frac{\pi j}{\ell}\right)}} \right); \quad \beta^* = \text{sgn}(E - \varepsilon) \quad (5)$$

and

$$\cos\left(\frac{q_{\beta^*}^j a}{2}\right) = \frac{-1}{2} \cos\left(\frac{\pi j}{\ell}\right) - \frac{\beta}{2} \sqrt{\left(\frac{E - \varepsilon}{2t_{\text{CNT}}}\right)^2 - \sin^2\left(\frac{\pi j}{\ell}\right)} \quad (6)$$



**Figure 2.** The order parameter  $\phi_n$  for the molecule with  $N = 56$  carbon atoms and three solitons.

in which  $\varepsilon$  and  $t_{\text{CNT}}$  are the on-site energy and the hopping integral between the nearest-neighbours, respectively. The molecular electronic Green's function is introduced as self-energy corrections into the bare 'molecule' Green's function,

$$G^{-1} = G^{\text{M-1}} - \Sigma^{\text{L}} - \Sigma^{\text{R}}, \quad (7)$$

where  $G^{\text{M-1}}$  and  $\Sigma^{\text{L(R)}}$  are the bare 'molecule' retarded Green's function and the left (right) CNT self-energy, respectively. The bare 'molecule' retarded Green's function operator is given by,

$$G^{\text{M}} = (1z - H_{\text{SSH}})^{-1}; \quad z = E + is \quad s \rightarrow 0 \quad (8)$$

in which  $H_{\text{SSH}}$  represents the well-known SSH Hamiltonian [32] of the bare molecule, 1 stands for the identity matrix, and  $z$  is the complex number whose real part,  $E$ , is the energy at which the transfer occurs.

It has already been shown that the main contribution to the electronic properties comes from the  $\pi$ -electrons [33, 34], such that one may only consider the electronic part of the well-known SSH Hamiltonian. The electronic part of  $H_{\text{SSH}}$  is given as follow [20]:

$$H_{\text{SSH}} = - \sum_n t_{n+1,n} (c_{n+1}^+ c_n + c_n^+ c_{n+1}), \quad (9)$$

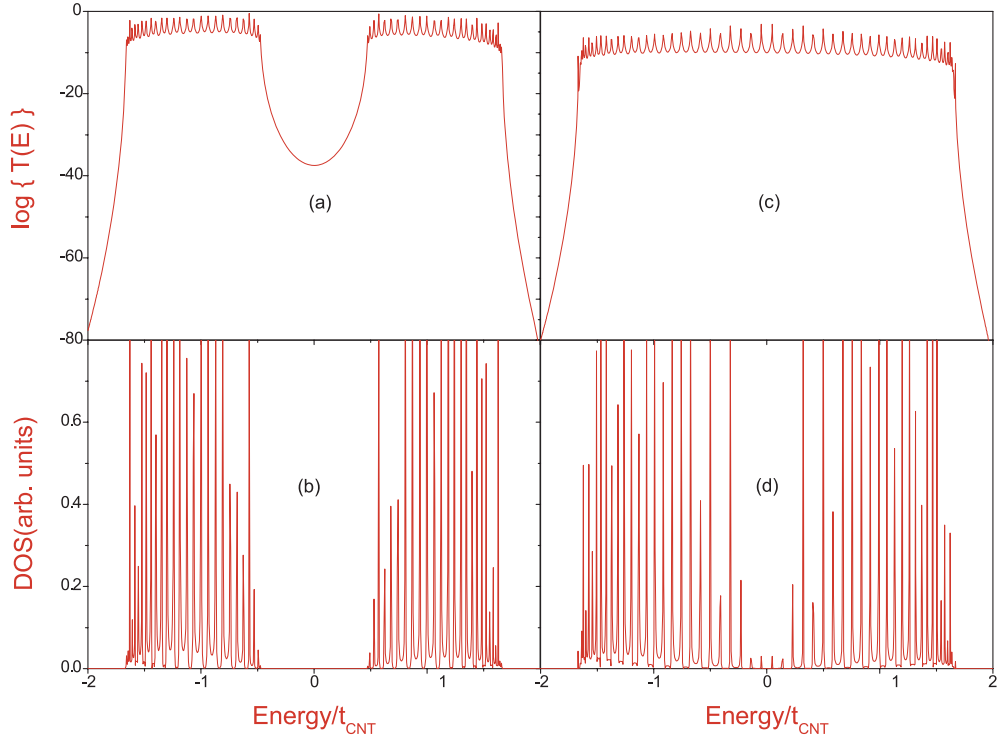
in which  $t_{n+1,n}$  is the nearest-neighbour transfer integral and is given as,

$$t_{n+1,n} = t_0 + \alpha(u_n - u_{n+1}), \quad (10)$$

where  $t_0$  is the hopping integral of an undimerized chain,  $\alpha$  is the electron-phonon coupling constant, and  $u_n$  is the displacement of the  $n$ th carbon atom from its equilibrium position. The reference energy is chosen such that the carbon atom on-site energy is zero. For perfectly dimerized *trans-PA*,  $u_n$  is given as  $u_n = (-1)^n u_0$ . Using the parameters of [32] we set  $\alpha = 4.1 \text{ eV } \text{\AA}^{-1}$ ,  $u_0 \approx 0.04 \text{ \AA}$  and  $t_0 = 2.5 \text{ eV}$ .

In *t-PA*, solitons (geometrical defects) correspond to phonon field configurations and may effectively affect the electronic conduction through the polymer. In the presence of solitons the displacements of carbon atoms are determined in such a way as to minimize the total energy and to give the optimum shape for the soliton. Accordingly, the displacements of the carbon atoms are modelled as [35, 36],

$$u_n = (-1)^n u_0 \prod_m \tanh \left[ \frac{(n-m)a}{\xi} \right], \quad (11)$$



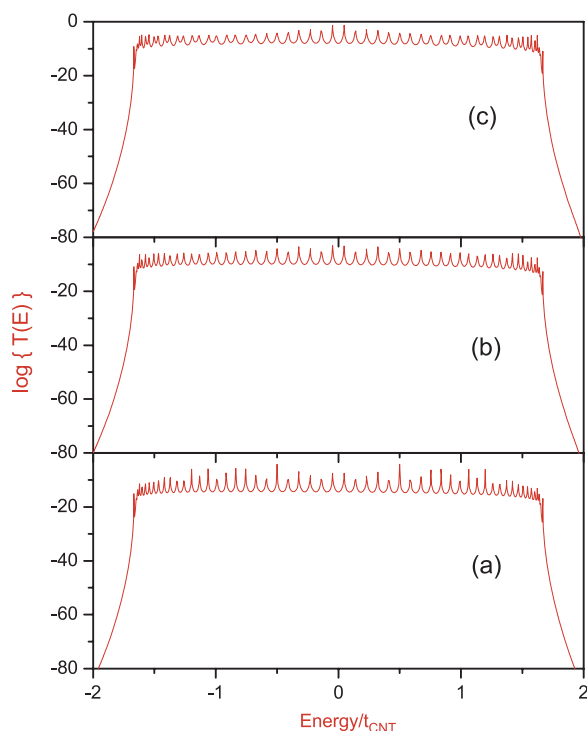
**Figure 3.** Panels (a) and (c) show the logarithm of the transmission coefficient versus the dimensionless parameter  $energy/t_{CNT}$  for the CNT/*t*-PA/CNT system with  $t_{CNT} = 3$  eV,  $t_c = 0.3$  eV for  $N = 56$  carbon atoms in a *t*-PA molecule, without a soliton and with three solitons, respectively. The CNTs are considered to be armchair (5, 5). Corresponding to plots (a) and (c), the electronic DOS is illustrated in panels (b) and (d), respectively.

where  $2\xi = 14a$ , and where  $a = 1.22$  Å (lattice constant) is the width of a soliton.  $(ma)$  is the location of the  $m$ th soliton centre on the chain. Defining the order parameter  $\varphi_n = -(-1)^n u_n$ , it is easy to observe the different soliton distributions along the *t*-PA chain, since  $\varphi_n = 0$  means a soliton has been located at the  $n$ th site. The order parameter  $\varphi_n$  for three solitons is illustrated in figure 2. Using partitioning techniques [27, 37], we may rewrite the  $(1, N)$  element of the retarded Green's function matrix of the molecule as follow:

$$G_{1N}(0) = \begin{bmatrix} E_1 + i\Delta & t_{1,2} & 0 & \cdots & 0 & 0 & 0 \\ t_{2,1} & E_2 & t_{2,3} & \cdots & 0 & 0 & 0 \\ 0 & t_{3,2} & E_3 & \cdots & 0 & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & 0 & \cdots & E_{N-2} & t_{N-2,N-1} & 0 \\ 0 & 0 & 0 & \cdots & t_{N-1,N-2} & E_{N-1} & t_{N-1,N} \\ 0 & 0 & 0 & \cdots & 0 & t_{N,N-1} & E_N + i\Delta \end{bmatrix}_{1N}^{-1}. \quad (12)$$

Also, the electronic density of states (DOS) of the system is given as [37],

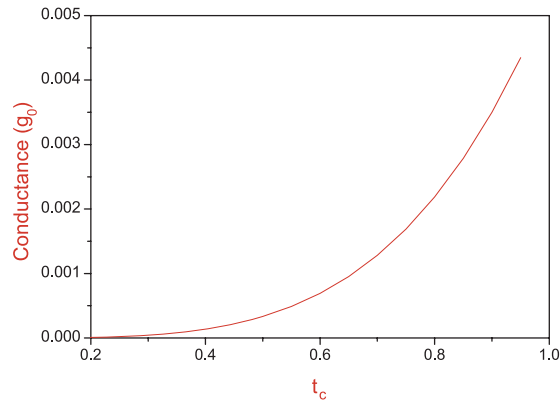
$$\rho_n(E) = -\frac{1}{\pi} \text{Im} \{G_{nn}(E)\}. \quad (13)$$



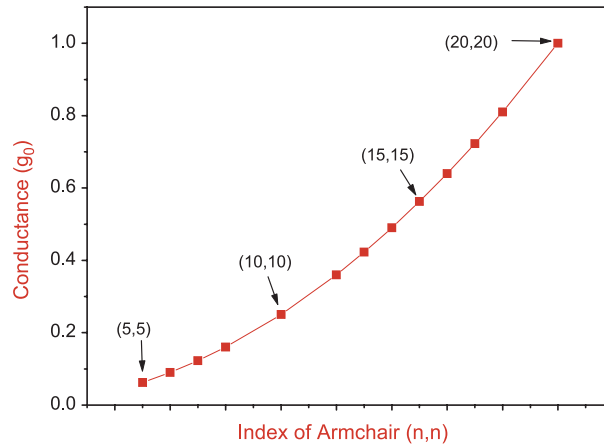
**Figure 4.** Plots (a)–(c) show the logarithm of transmission coefficient versus the dimensionless parameter  $energy/t_{CNT}$  for the CNT/*t*-PA/CNT system with  $t_{CNT} = 3$  eV,  $t_c = 0.1, 0.3, 0.5$  eV for  $N = 56$  carbon atoms and three solitons. The CNTs are considered to be armchair (5, 5).

### 3. Results and discussion

Based on the formalism described in section 2, we have investigated the electronic conduction properties of the CNT/*t*-PA/CNT structure for several typical *t*-PA molecules with different features. Here we focus on the dependence of the electron conduction on the presence of solitons in the *t*-PA chain. Figure 3 illustrates the electronic density of states (DOS) of the CNT/*t*-PA/CNT system in the absence and in the presence of solitons (panels (b) and (d), respectively). The plots show that the presence of solitons induces electronic states within the bandgap and causes a large enhancement in the conductance of the system. Also, the electronic transmission probability through the system is shown in panels (a) and (c), which correspond to panels (b) and (d), respectively. In these figures the CNT/molecule coupling strength,  $t_c$ , is fixed. Similar calculations have been performed for different values of  $t_c$ . Our results suggest that any increase in  $t_c$  gives rise to a considerable enhancement in the conductance of the system. Figure 4 illustrates this aspect of the electronic transmission clearly. Figures 5 and 6 illustrate the conductance of the molecular wire for some selected armchair nanotubes with different tube diameters as the electrode. We find that, as the tube radius ( $R$ ) increases, the conductance of the system considerably increases too. From the most experimentally observed carbon nanotube sizes, there is a tiny gap in the armchair nanotube types which arises from curvature effects [38]. Thus any increase in the tube diameter give rises to a decrease in the gap as  $1/R^2$  [39] and causes a large enhancement in the conductance of the system.



**Figure 5.** The differential conductance (in units of  $g_0$ ) versus  $t_c$ , the CNT/molecule coupling strength in the CNT/*t*-PA/CNT system with  $t_{\text{CNT}} = 3$  eV for  $N = 56$  carbon atoms and three solitons. The CNTs are considered to be armchair (5, 5).



**Figure 6.** The differential conductance (in units of  $g_0$ ) versus the tube diameter of electrodes with  $t_{\text{CNT}} = 3$  eV,  $t_c = 0.3$  eV and  $N = 56$  carbon atoms.

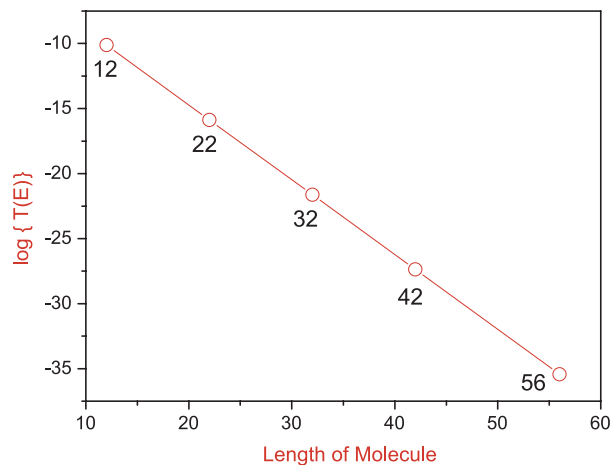
In figure 7 we have shown the effect of the selected lengths of the molecule on the conductance of the system. From this figure we see that, when the length of the molecule increases, the conductance largely decreases. As is well known, our results indicate that the decrease in the conductance follows an exponential law, as shown by *Magoga and Joachim* [14].

In all of the above calculations, no voltage drop was considered across the molecule. However, in order to study the behaviour of the system in the presence of an applied voltage, we consider a linear voltage drop along the *t*-PA molecule, i.e. the electric potential between the electrodes (CNTs) varying linearly with distance. This assumption is a good approximation to the computational potential profile through the molecule between the two electrodes [40]. Thus the on-site energies of the molecule are functions of the bias,

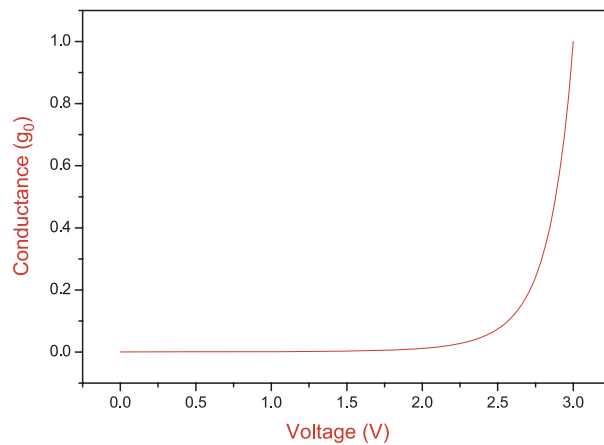
$$V(j) = -\left(\frac{eV_0}{N-1}\right)(j-1); \quad j = 1, \dots, N. \quad (14)$$

The results of these studies are presented in figure 8. These results suggest that any increase in the value of the electric potential between the molecule ends gives rise to an increase in the





**Figure 7.** A plot of the logarithm of the transmission coefficient as a function of length of the *t*-PA molecule with  $t_{\text{CNT}} = 3$  eV,  $t_c = 0.3$  eV.



**Figure 8.** The differential conductance (in units of  $g_0$ ) versus the voltage drop along the *t*-PA molecule for the CNT/*t*-PA/CNT system. Here, for armchair (5, 5) nanotubes as electrodes,  $t_{\text{CNT}} = 3$  eV,  $t_c = 0.3$  eV,  $V_0 = 2$  V and  $N = 56$  carbon atoms are set. In the presence of solitons, the conductance–voltage curve is steeper.

conductance of the molecular wire. This is due to the fact that the voltage drop makes the molecular orbitals wider, and thus electron tunnelling from site to site will be easier.

#### 4. Summary

In brief, we have studied in detail the role of solitons and the CNT/molecule coupling strength on the conductance of the CNT/*t*-PA/CNT structure. We have applied some well-known approaches and methods based on Green's function theory and Löwdin's partitioning technique, *Landauer* formalism and the SSH Hamiltonian model to investigate the electron conduction through the CNT/*t*-PA/CNT structure. Our results show that (i) the presence of solitons in the *t*-PA chain induces electronic states within the bandgap of the molecule, which give rise to a large enhancement in the conductance and (ii) the conductance is sensitive to the CNT/*t*-PA coupling, the length of the *t*-PA molecule and the CNT's chirality. In addition, the presence

of the linear voltage drop along the molecule considerably increases the conductance of the system.

## References

- [1] Qi P *et al* 2004 *J. Am. Chem. Soc.* **126** 11774–5 (Preprint [cond-mat/0408421](#))
- [2] Smith B W *et al* 2001 *Fall Meeting Symposium Z; MRS Proc.* **706** 1–6
- [3] Gutierrez R *et al* 2003 *Preprint cond-mat/0301620*
- [4] Liang G C, Ghosh A W, Paulsson M and Datta S 2004 *Phys. Rev. B* **69** 115302
- [5] Guo J *et al* 2003 *IEEE Trans. Nanotechnol.* **2** 329
- [6] Joachim C, Gimzewski J K, Schlittler R R and Chavy C 1995 *Phys. Rev. Lett.* **74** 2102
- [7] Sautet P 1997 *Chem. Rev.* **97** 1097  
Wolkow R A 1999 *Annu. Rev. Phys. Chem.* **50** 413
- [8] Langlais V J *et al* 1999 *Phys. Rev. Lett.* **83** 2809
- [9] Bumm L A *et al* 1996 *Science* **271** 1705
- [10] Bumm L A *et al* 1999 *J. Phys. Chem. B* **103** 8122
- [11] Zhou C, Kong J and Dai H 2000 *Phys. Rev. Lett.* **84** 5604
- [12] Porath D, Bezryadin A, de Vries S and Dekker C 2000 *Nature* **403** 635
- [13] Kergueris C *et al* 1999 *Phys. Rev. B* **59** 12505
- [14] Magoga M and Joachim C 1998 *Phys. Rev. B* **56** 4722  
Magoga M and Joachim C 1999 *Phys. Rev. B* **59** 16011
- [15] Emberly E G and Kirczenow G 1998 *Phys. Rev. B* **58** 10911  
Emberly E G and Kirczenow G 1998 *Phys. Rev. Lett.* **81** 5205  
Emberly E G and Kirczenow G 1999 *Nanotechnology* **10** 285  
Emberly E G and Kirczenow G 1999 *J. Phys.: Condens. Matter* **11** 6911
- [16] Hall L E, Reimers J R, Hush N S and Silverbrook K 2000 *J. Chem. Phys.* **112** 1510
- [17] Datta S *et al* 1997 *Phys. Rev. Lett.* **79** 2530  
Tian W 1997 *Physica E* **1** 304  
Tian W 1998 *J. Chem. Phys.* **109** 2874  
Xue Y 1999 *Phys. Rev. B* **59** R7852
- [18] Yaliraki S N *et al* 1999 *J. Chem. Phys.* **111** 6997
- [19] Di Ventura M, Pantelides S T and Lang N D 2000 *Phys. Rev. Lett.* **84** 979
- [20] Heeger A J, Kivelson S, Schrieffer J R and Su W P 1988 *Rev. Mod. Phys.* **60** 781
- [21] Phillport S R, Baeriswyl D, Bishop A R and Lomdahl P S 1987 *Phys. Rev. B* **35** 7533
- [22] Stafstrom S 1991 *Phys. Rev. B* **43** 9158
- [23] Hjort M and Stafstrom S 2000 *Phys. Rev. B* **62** 5245
- [24] Lu Y 1988 *Solitons and Polarons in Conducting Polymers* (Singapore: World Scientific)
- [25] Bredas J L, Cornil J and Heeger A J 1996 *Adv. Mater.* **8** 447  
Knapfer M *et al* 2000 *Phys. Rev. B* **61** 1662
- [26] Buttiker M, Imry Y, Landauer R and Pinhas S 1985 *Phys. Rev. B* **31** 6207
- [27] Mujica V, Kemp M and Ratner M A 1994 *J. Chem. Phys.* **101** 6849–55
- [28] Larade B and Bratkovsky A M 2003 *Phys. Rev. B* **68** 235305
- [29] Calev Y *et al* 2004 *Isr. J. Chem.* **44** 133
- [30] Fisher D S and Lee P A 1981 *Phys. Rev. B* **23** R6851–4
- [31] Cuniberti G *et al* 2002 *Chem. Phys.* **281** 465–76
- [32] Su W P, Schrieffer J R and Heeger A J 1980 *Phys. Rev. B* **22** 2099
- [33] Shahtahmasebi N and Ketabi S A 2002 *Indian J. Phys. A* **76** 245
- [34] Chavy C, Joachim C and Altibelli A 1993 *Chem. Phys. Lett.* **214** 569
- [35] Agranovich V M and Maradudim A A 1986 *Solitons* (Amsterdam: North-Holland)
- [36] Stafstrom S and Chao K A 1984 *Phys. Rev. B* **30** 2098
- [37] Mujica V, Kemp M and Ratner M A 1994 *J. Chem. Phys.* **101** 6856–64
- [38] Wildoer J W G *et al* 1998 *Nature* **391** 59
- [39] Kim P, Odom T W, Huang J L and Lieber C M 1999 *Phys. Rev. Lett.* **82** 1225  
Odom T W *et al* 1998 *Nature* **391** 59
- [40] Liang G C, Ghosh A W, Paulsson M and Datta S 2004 *Phys. Rev. B* **69** 115302  
Paulsson M, Zahid F and Datta S 2002 *Preprint cond-mat/0208183*