

EPJ B

Condensed Matter
and Complex Systems

EPJ.org
your physics journal

Eur. Phys. J. B (2016) 89: 102

DOI: [10.1140/epjb/e2016-70013-y](https://doi.org/10.1140/epjb/e2016-70013-y)

An efficient coarse-grained approach for the electron transport through large molecular systems under dephasing environment

Daijiro Nozaki, Raul Bustos-Marín, Carlos J. Cattena,
Gianaurelio Cuniberti and Horacio M. Pastawski

edp sciences



 Springer

An efficient coarse-grained approach for the electron transport through large molecular systems under dephasing environment

Daijiro Nozaki^{1,2,3,4,a}, Raul Bustos-Marín^{5,6}, Carlos J. Cattena⁵,
Gianaurelio Cuniberti^{1,2,3}, and Horacio M. Pastawski⁵

¹ Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, 01062 Dresden, Germany

² Dresden Center for Computational Materials Science (DCCMS), TU Dresden, 01062 Dresden, Germany

³ Center for Advancing Electronics Dresden (cfAED), TU Dresden, 01062 Dresden, Germany

⁴ Lehrstuhl für Theoretische Physik, Universität Paderborn, Paderborn, Germany

⁵ Instituto de Física Enrique Gaviola and Facultad de Matemática Astronomía y Física, Universidad Nacional de Córdoba, Ciudad Universitaria, 5000 Córdoba, Argentina

⁶ Facultad de Ciencias Químicas, Universidad Nacional de Córdoba, Ciudad Universitaria, 5000 Córdoba, Argentina

Received 9 January 2016 / Received in final form 24 February 2016

Published online 18 April 2016 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2016

Abstract. Dephasing effects in electron transport in molecular systems connected between contacts average out the quantum characteristics of the system, forming a bridge to the classical behavior as the size of the system increases. For the evaluation of the conductance of the molecular systems which have sizes within this boundary domain, it is necessary to include these dephasing effects. These effects can be calculated by using the D’Amato-Pastawski model. However, this method is computationally demanding for large molecular systems since transmission functions for all pairs of atomic orbitals need to be calculated. To overcome this difficulty, we develop an efficient coarse-grained model for the calculation of conductance of molecular junctions including decoherence. By analyzing the relationship between chemical potential and inter-molecular coupling, we find that the chemical potential drops stepwise in the systems with weaker inter-unit coupling. Using this property, an efficient coarse-grained algorithm which can reduce computational costs considerably without losing the accuracy is derived and applied to one-dimensional organic systems as a demonstration. This model can be used for the study of the orientation dependence of conductivity in various phases (amorphous, crystals, and polymers) of large molecular systems such as organic semiconducting materials.

1 Introduction

The study of charge transport through molecules became an active research field in the last few decades. The most widely used methods for calculation of conductance of nanostructures and molecules connected between contacts or electrodes are based on Landauer’s approach combined with Green’s function formalism [1,2]. In a standard application, this method only takes into account for coherent tunneling which is identified with the conductance. However, as the size of molecular structures reaches the nanoscale, the effect of inelastic tunneling, dephasing and disorder begins to be influential. Thus, for the integration of molecular devices, such effects have to be taken into account. This requires to expand the study of electron transport through the large molecular systems such as DNAs and organic semiconductors between contacts where dephasing phenomena play essential roles.

One of major sources of dephasing is the interaction of traveling electron with vibrational degree of free-

dom, i.e. electron-phonon (e-ph) interaction. In order to incorporate such dephasing and inelasticity mechanisms, various methods such as density matrix approaches, kinetic equations, and scattering matrix methods have been developed [3–24]. Among them, D’Amato-Pastawski (DP) model is suitable for the analysis of the relationship between molecular structures and its electrical response under dephasing environment and weakly inelastic processes [3–5]. In the DP model, local scattering processes are introduced which, in full analogy to the Büttiker’s description of voltage probes in a multiterminal device [25–27], must be complemented with fictitious reservoirs or “probes” that reinject locally each particle lost from the coherent beam. At small voltage bias, these last allow to impose the current conservation providing a form of Markov equation relating the reservoirs from which the net steady state current is calculated. This results in an effective transmittance between the actual external reservoirs which adds an additional correction term to the coherent tunneling component. Although the DP model was applied to a series of organic

^a e-mail: daijiro.nozaki@gmail.com

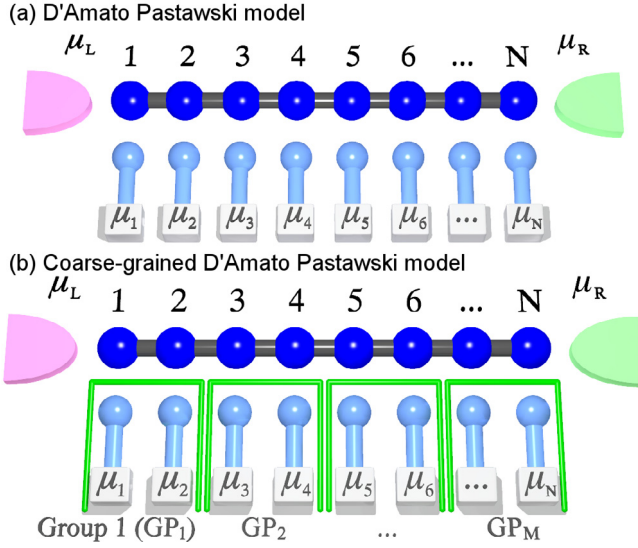


Fig. 1. A schematic model of (a) D'Amato-Pastawski (DP) model and (b) coarse-grained D'Amato-Pastawski (CGDP) model. In both cases, the molecules having N sites are coupled between two contacts. In order to describe dephasing processes, fictitious scattering probes (Büttiker probes) are attached homogeneously. In the coarse-grained model, the condition of zero current is imposed not to each single scattering probe but to each group of scattering probes.

molecular wires [7] and could successfully demonstrate the well-known tunneling-to-hopping transition [8,9,28], it is still too demanding for the study of electron transport through large molecular systems such as organic semiconducting materials or assemblies of nanoparticles connected via organic molecules.

In this article, we investigate the relationship between tight-binding parameters of one-dimensional (1D) molecular wires and the drop of chemical potential along the wires embedded between contacts. After analyzing the stepwise drop of chemical potential along the 1D chains with different inter-unit coupling, we derive a coarse-grained algorithm that solves the DP model enabling to calculate conductance of large molecular systems connected between two contacts under dephasing environment efficiently with the minimum loss of the accuracy. We show the condition for the application of the coarse-grained model and present that the effective transmission including dephasing for the molecular wires can be calculated efficiently only if the inter unit coupling is weak enough. We demonstrate the coarse-grained DP (CGDP) model first using an 1D chain with toy model basis and then apply to organic molecules as a practical application.

2 Method

Figure 1 shows tight-binding scheme of (a) the DP model and (b) the proposed CGDP model. The central molecular system having N sites is connected between two contacts. In both models, effective Hamiltonians are identical defined as $H_{\text{eff}} = H_M + \Sigma_L + \Sigma_R + \sum_{m=1}^N \Sigma_{B,m}$, where

H_M is the Hamiltonian matrix for the central molecule, $\Sigma_{L/R}$ is the self-energy term for left/right contact, the last term $\Sigma_{B,m}$ is the self-energy correction due to the m th dephasing probes defined as $\Sigma_{B,m} = -i\gamma_{e\text{-ph}}/2$ is the dephasing parameter set from electron-phonon coupling. Hence, the effective Green's function in orthogonal basis is given by:

$$G_{\text{eff}}^{R/A}(E) = [(E \pm i\delta)I - H_{\text{eff}}]^{-1}. \quad (1)$$

The CG model starts from the DP model described in detail in reference [3]. Here we first give a brief derivation of DP model for completeness. In the DP model (Fig. 1a), N phenomenological scattering probes connected to external reservoirs are introduced in order to describe the incoherent events that affect the transport properties of the system.

The net current in each scattering probe is determined by the chemical potentials $\mu_L = \mu_0$, μ_i ; $i = 1, 2, \dots, N$, and $\mu_R = \mu_{N+1}$, if the transmission and reflection probabilities $T_{i,j}$ and $R_{i,i}$ are given. $T_{i,j}$ is the transmission probability from j th to i th probe obtained from a generalized form of the Fisher-Lee relation as $T_{i,j} = \text{Tr}[T_{B,i} G_{\text{eff}}^{R} T_{B,j} G_{\text{eff}}^A]$, where the term $T_{B,m}$ is broadening function defined as $T_{B,m} \equiv i[\Sigma_{B,m} - \Sigma_{B,m}^\dagger]$, and $R_{i,i}$ is the reflection probability at i th channel. Taking $\mu_L > \mu_R$, the condition of no current flow at each i th channel implies following relations:

$$(1 - R_{i,i})(\mu_i - \mu_R) - \sum_{j=1(j \neq i)}^N T_{i,j}(\mu_j - \mu_R) - T_{i,L}(\mu_L - \mu_R) = 0, \quad \forall i = 1, 2, \dots, N. \quad (2)$$

Chemical potentials μ_i , $i = 1, 2, \dots, N$, are determined by imposing the current conservation in equation (2) for all scattering probes simultaneously. Therefore, they are given by:

$$(\mu_i - \mu_R) = \sum_{j=1(j \neq i)}^N W_{i,j}^{-1} T_{j,0}(\mu_L - \mu_R), \quad \forall i = 1, 2, \dots, N, \quad (3)$$

where the Markov matrix W is defined as $W_{i,j} = (1 - R_{i,i})\delta_{i,j} - T_{i,j}(1 - \delta_{i,j})$. The dephased component is included as an additional correction term to coherent component. Therefore, the effective current within DP model is obtained by $I = \frac{e}{h} \sum_{i=0}^N T_{R,i}(\mu_i - \mu_R) = \frac{e}{h} T_{\text{eff}} \delta\mu$, where $\delta\mu \equiv (\mu_L - \mu_R)$ and the effective transmission is given by:

$$T_{\text{eff}} = T_{L,R} + \sum_{i,j=1}^N T_{R,i} W_{i,j}^{-1} T_{j,L}. \quad (4)$$

In order to obtain effective conductance by DP model, all transmission probabilities, T_{ij} for all pairs of scattering probes needs to be calculated. However, this process requires large computational time since the number of the terms T_{ij} increases quadratically. For overcoming this

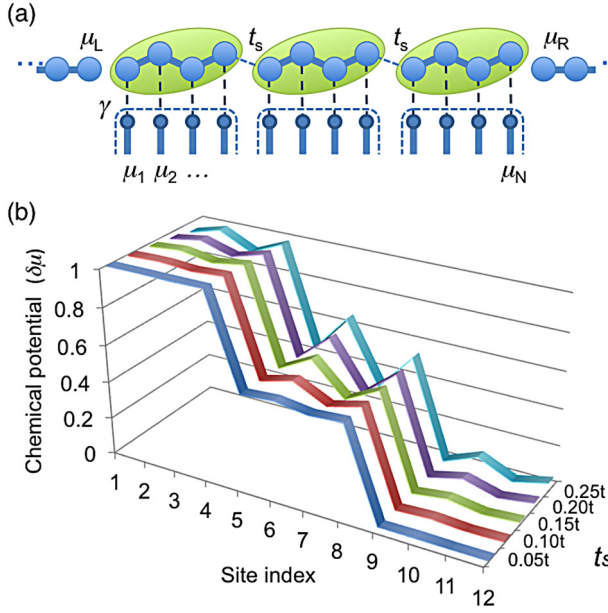


Fig. 2. Application of the DP and the CGDP model to 1D systems: (a) 1D polymer with inter-unit coupling (t_s) is coupled between contacts. Each monomer has 4 sites and the polymer consists of 3 monomers. Thus, the number of whole sites is set to $N = 12$. The fictitious probe is connected to each site. γ_{e-ph} is the dephasing strength. The transfer integrals within the monomer unit is set to t . For the CGDP model, 4 probes are grouped. (b) Chemical potentials across the 1D molecule with different inter-unit coupling calculated within conventional DP model.

difficulty, we propose a coarse-grained version of the DP model (see Fig. 1b).

Before proceeding to the derivation of the coarse-grained version of the DP model, we examine the chemical potential drop along the one-dimensional (1D) tight-binding systems coupled between contacts using DP model in order to get an insight into the speeding up of the DP model. Figure 2 presents the test system where a polymeric molecule having three unit cells is coupled between contacts. The inter-unit couplings are set to t_s (Fig. 2a). For the simplicity, we assume that the monomer unit consists of four sites and that intra-molecular transfer integrals are set to t with nearest neighbor approximation. The bias voltage is set to small enough. The coupling between left/right lead and the molecular system is set to $V_{L/R} = 0.2t$. All on-site energies are set to zero. The left/right contacts are described by 1D tight-binding chains within Newns-Anderson model where those self-energy terms are given by $\Sigma_{L/R}(E) = V_{L/R}^2 e^{i\theta}/t_{L/R}$ and $\cos\theta = E/2t_{L/R}$. The transfer integral in the left/right lead, $t_{L/R}$, is set to t .

Figure 2b presents the drop of chemical potential across the 1D system in Figure 2a with different inter-unit coupling t_s , which was calculated by using the DP model. We can clearly see that the stepwise drop of the chemical potential for weakly coupled 1D systems. This result implies that the Büttiker probes can be grouped in

case of the molecular systems such as non-polymeric organic semiconductors having the weak inter-unit coupling. Next we exploit this feature for the coarse-grained model.

If N scattering probes are divided into M groups as Figure 1b, then, equation (2) can be written in the following form as:

$$\begin{aligned} & \sum_{m \in GP_I} (1 - R_{m,m})(\mu_m - \mu_R) - \sum_{m,n \in GP_I, m \neq n} T_{m,n}(\mu_n - \mu_R) \\ & - \sum_{m \in GP_I, n \in GP_J, m \neq n} T_{m,n}(\mu_n - \mu_R) \\ & - \sum_{m \in GP_I} T_{m,L}(\mu_L - \mu_R) = 0, \quad \forall I = 1, 2, \dots, M. \end{aligned} \quad (5)$$

In the coarse-grained model, the condition of zero current is imposed not to *each single scattering probe* as equation (2) but to *each group of scattering probes*. Then, the effective conductance in the coarse-grained model, \tilde{T}_{eff} , is derived from this condition. Apparently, this change of imposition of zero current from the individual scattering probes in equation (2) to the group of the scattering probes seems to give the different effective conductance from the one obtained from equation (4). However, hereafter we show if we use the stepwise profile of the chemical potentials of the fictitious probes for the weakly coupled 1D systems, the effective conductance \tilde{T}_{eff} in the coarse-grained model is the same as the one obtained from the conventional DP model, T_{eff} .

Since the chemical potentials of the intra-unit are nearly identical for weakly coupled case as shown in Figure 2b, all chemical potentials μ_m in group I are the same:

$$\mu_m = \mu_I; m \in GP_I; I = 1, 2, \dots, M. \quad (6)$$

Thus, equation (5) can be simplified to the following form:

$$\begin{aligned} & (\tilde{R}_{I,I})(\mu_I - \mu_R) - \sum_{J=1, I \neq J}^M \tilde{T}_{I,J}(\mu_J - \mu_R) \\ & - \tilde{T}_{I,L}(\mu_L - \mu_R) = 0, \quad \forall I = 1, 2, \dots, M, \end{aligned} \quad (7)$$

where $\tilde{R}_{I,I} = \sum_{m \in GP_I} (1 - R_{m,m}) + \sum_{m,n \in GP_I, m \neq n} T_{m,n}$ and $\tilde{T}_{I,J} = \sum_{m \in GP_I, n \in GP_J, I \neq J} T_{m,n}$. Equation (7) can be regarded as coarse-grained version of equation (2). Therefore, chemical potentials μ_I satisfying both the restriction of equation (6) and current conservation requirement simultaneously for all coarse grained scattering probes in equation (7) are given by:

$$(\mu_I - \mu_R) = \left[\sum_{J=1(J \neq I)}^M \tilde{W}_{I,J}^{-1} \tilde{T}_{J,0} \right] \delta\mu, \quad \forall I = 1, 2, \dots, M. \quad (8)$$

The Markov matrix \tilde{W} in coarse-grained form is defined as $\tilde{W}_{I,J} = \tilde{R}_{I,I} \delta_{I,J} - \tilde{T}_{I,J}(1 - \delta_{I,J})$. Finally, the effective current within CGDP model is obtained by:

$$I = \frac{e}{h} \sum_{I=0}^M \tilde{T}_{R,I}(\mu_I - \mu_R) = \frac{e}{h} \tilde{T}_{\text{eff}} \delta\mu,$$

where \tilde{T}_{eff} is the coarse-grained version of the effective transmission in equation (4) given by:

$$\tilde{T}_{\text{eff}} = T_{L,R} + \sum_{i,j=1}^M \tilde{T}_{R,I} \tilde{W}_{I,J}^{-1} \tilde{T}_{J,L}. \quad (9)$$

Using this coarse-grained model, the size of Markov matrix W in the conventional DP model can be dramatically reduced from N^2 to M^2 without substantial loss in accuracy. Therefore, computational time can be reduced to $\frac{M^2}{N^2}$ of the original DP model. Note that this coarse-grained model does not reduce the size of the Hamiltonian matrices itself as shown in equation (1). Therefore the bottleneck of the computation is the inversion in equation (1) with $\mathcal{O}(N^3)$. The approach to reduce the size of original Hamiltonian itself is given elsewhere [28]. CGDP model guarantees $\tilde{T}_{\text{eff}} = T_{\text{eff}}$ if the condition in equation (6) is satisfied i.e. for the molecular system with weak inter-unit couplings. Another merit is that we do not need to calculate all transmissions $T_{i,j}$ for all pairs of the probes but only need the transmissions $\tilde{T}_{I,J}$ for all pairs of the group of the probes, which can be calculated at once by $\tilde{T}_{I,J} = \text{Tr}[\tilde{I}_{B,I} G_{\text{eff}}^R \tilde{I}_{B,J} G_{\text{eff}}^A]$, where the term $\tilde{I}_{B,I}$ is broadening function of the I th group of the probes defined as $\tilde{I}_{B,I} \equiv i \sum_{m \in I} [\Sigma_{B,m} - \Sigma_{B,m}^\dagger]$.

3 Applications

3.1 Application to toy model

For checking the performance of the coarse-grained model, we applied the CGDP model to 1D systems and compared the calculated chemical potential along the systems, the effective conductance, and computational time with the conventional DP model in toy model basis. First, the chemical potential profiles along the 1D system in Figure 2a are calculated by using both DP and CGDP model for strong (Fig. 3a) and weak (Fig. 3b) inter-unit couplings. We grouped four Büttiker probes as one effective probe as shown in Figure 2a, thus the CGDP model has three phenomenological probes. As expected from the analysis in Figure 2 and subsequent derivation of the coarse-grained model, the chemical potential of the weakly coupled system calculated by the CGDP model shows a very good agreement with the one calculated by the conventional DP model (see Fig. 3b). This result supports the grouping of Büttiker probes and the simplification of the DP model.

Next, we considered an 1D system having 96 sites (24 unit cells) and applied both models to this system. Then we examined how the effective transmission at the Fermi energy obtained by the CGDP model dissociates from the reference value calculated from the conventional DP model depending on the inter-unit couplings. In Figure 4a, the relative error $\frac{|\tilde{T}_{\text{eff}}(E_F) - T_{\text{eff}}(E_F)|}{T_{\text{eff}}(E_F)}$ as a function of inter-unit coupling t_s/t is displayed. We clearly see small

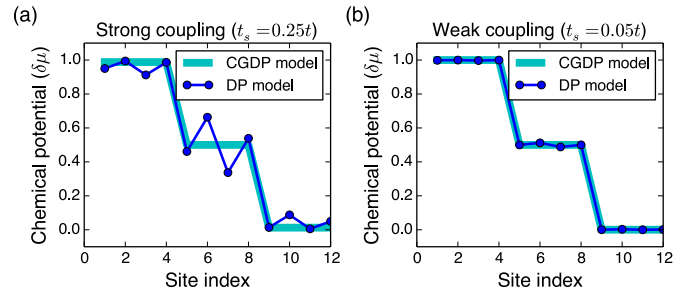


Fig. 3. Comparison of the calculated chemical potentials of Figure 2a between DP model and CGDP model for (a) weak and (b) strong inter-unit couplings. The chemical potential drop obtained by the CGDP model in weaker inter-unit couplings shows a good agreement with the conventional one.

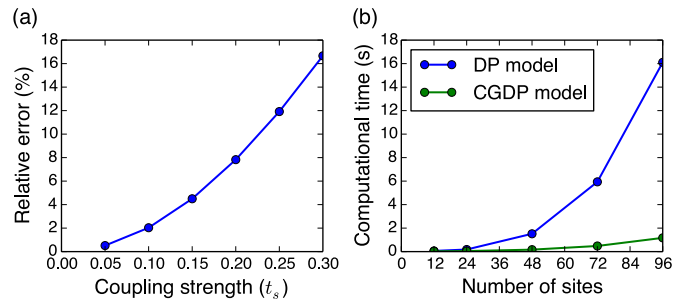


Fig. 4. Performance of CGDP model: (a) relative error $|T_{\text{eff}} - \tilde{T}_{\text{eff}}|/T_{\text{eff}}$ of CGDP model as a function of inter-unit coupling t_s in the systems in a 1D chain having 96 sites (24 units and 4 sites in each unit cell), (b) comparison of computational time as a function of the number of sites calculated by DP and CGDP model. The employment of CGDP model in weakly coupled system gives small errors with short time.

difference between two methods for smaller inter-unit couplings. Thus, the CGDP method is suitable for the calculation of transmission for the weakly coupled systems such as stacks of organic molecules where the transfer integrals for intermolecular pairs of atomic orbitals is much smaller than that of the intramolecular ones.

In order to evaluate the computational time reduction with the CGDP model, we compared computational times for CGDP model and conventional DP model. In Figure 4b, the comparison of computational time of these models as a function of chain length is shown. In the CGDP model, every four Büttiker probes is grouped into one group as shown in Figure 2a. Although the computational time by CGDP model also increases quadratically, still computational time for the CGDP model is significantly reduced from the conventional one. From these analyses, we conclude that CGDP model applied to weakly coupled systems can reduce computational time considerably with small relative error.

3.2 Practical applications to organic molecules

To demonstrate the convenience of the coarse-grained model with practical atomistic structures and also to give a warning to the improper application of the

Table 1. Comparison of the performance between DP model and CGDP model. The effective conductance at the Fermi energy in the weakly coupled systems shown in Figure 5a with different number of stacked naphthalenes are compared with computational time. The relative error $\frac{|\tilde{T}_{\text{eff}}(E_F) - T_{\text{eff}}(E_F)|}{T_{\text{eff}}(E_F)}$, computational times, and the time gain due to the coarse-grained model are followed. The CGDP model can reduce computational time significantly without essential loss of accuracy for the weakly coupled systems.

N	Effective transmission $T(E_F)$		Relative error (%)	Time (s)		Gain
	DP	CGDP		DP	CGDP	
1	3.3209e-09	3.3210e-09	2.168e-02	372.85	0.316	1181
2	3.4879e-09	3.4879e-09	1.434e-04	1036.89	0.636	1631
4	3.7104e-09	3.7105e-09	8.085e-04	4476.10	2.342	1912
8	3.8131e-09	3.8132e-09	1.925e-03	37814.44	17.129	2208
12	1.9426e-10	1.9409e-10	8.766e-02	113729.27	51.627	2203

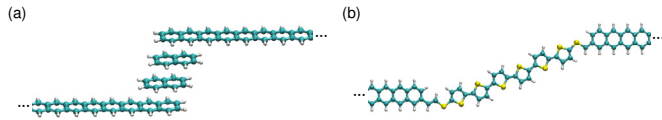


Fig. 5. Relaxed structures of (a) naphthalene stacks and (b) polythiophene oligomers between GNR leads.

coarse-grained model to the strongly interconnected systems, we applied both DP model and CGDP model to two types of systems; stacks of organic molecules with weak inter-unit coupling and polymeric systems with strong inter-unit coupling. Then we analyze the applicable scope and limitation of the CGDP model by comparing effective transmissions at the Fermi energies, relative errors $\frac{|\tilde{T}_{\text{eff}}(E_F) - T_{\text{eff}}(E_F)|}{T_{\text{eff}}(E_F)}$, and computational times. For the weakly coupled systems, we consider stacks of naphthalene molecules where molecules are weakly bound with van der Waals interactions, while for the strongly coupled systems we consider polythiophenes where monomers are covalently connected.

For the determination of the stable configurations of the molecular systems between conductive graphene nanoribbon (GNR) contacts, conjugate-gradient geometry optimization of the extended molecule (stacks of molecules plus one unit cell of the GNR on each side) was performed using the density-functional tight-binding method (DFTB). This DFT-parametrized semi-empirical approach has been shown to be computationally highly efficient. First, we relaxed the stacks of organic molecules or poly-thiophenes with periodic boundary conditions. Then the structurally optimized unit cells of the molecules were then placed between semi-infinite GNRs, and the extended molecules were relaxed again to take into account local structural modifications resulting from the interaction between the molecule and the electrodes. We use the parameterization of reference [29] for the carbon and hydrogen atoms and reference [30] for the strongly coupled systems using carbon, hydrogen, and sulfur atoms. Figure 5 shows the relaxed structures of the stack and polymers between GNR leads.

After the relaxation of the molecular junctions we calculated the effective conductance of the systems between the pair of GNR leads and compared the effective conductance at the Fermi energy in the CGDP model with conventional DP model. The Fermi energy was estimated to

be -4.49 eV, which is the Fermi energy of the GNR leads. Table 1 summarizes the effective conductance of naphthalene stacks between GNR leads at the Fermi energy calculated by the CGDP model and DP model with computation times and the relative errors. As expected from the theory and toy model analysis in Figures 3 and 4, the coarse-grained model can calculate the conductance with small computational time and small error. Therefore, the CGDP model can be used as a cost-efficient mean to estimate the conductance including dephasing effect of weakly coupled systems such as stacks of organic semiconducting materials.

Next, we applied the same models to the polythiophenes having strong inter-unit couplings. Table 2 presents the same contents as Table 1 but applied for the polythiophenes. We can see that the calculation time is largely reduced due to the CGDP model, while the relative errors are large for all systems. This is typical violation of the CGDP model. The large relative errors originates from the non-stepwise drop of the chemical potential along the polymers as discussed in the toy model in Figure 3a. Thus, the CGDP model should not be applied for the polymeric systems with strong inter unit coupling because of the large relative errors, or should be used keeping the possible errors in mind¹.

3.3 Practical application for isotropy analysis

To demonstrate the usability of the coarse-grained model, we applied the CGDP model to organic semiconducting materials in order to investigate the isotropy of dephased electron transport through them. As an example we focused on the pentacene-derivative (TMS) [31,32]. We relaxed semiconducting organic crystals using DFTB+ program and extracted a piece of organic crystal ($3 \times 3 \times 3$) and attached a pair of gold leads to calculate the effective conductance between the pair of gold leads. Figure 6 shows the extracted molecular structure of the organic crystal

¹ If the linearity of $\mu_i - \mu_R$ with the position of site i holds in the strong coupling regime, it is possible to describe the effective current as $I = \frac{2e}{h} [T_{LR}(\mu_L - \mu_R) + \sum_i T_{iR}(\mu_i - \mu_R)]$, where the appropriate expression for $\mu_i - \mu_R$ can be directly used. The transmission between fictitious probes $T_{i,j}$ need to be evaluated anyway but one can avoid the inversion of the W matrix speeding up the calculation.

Table 2. Comparison of the performance between DP model and CGDP model. The table has the same structure as Table 1. The systems of interest are polythiophene, one of those are shown in Figure 5b. The CGDP model can reduce computational time significantly, while it gives relative errors over 10%.

N	Effective transmission $T(E_F)$		Relative error (%)	Time (s)		Gain
	DP	CGDP		DP	CGDP	
2	4.8714e-03	5.8118e-03	19.304	562.42	0.430	1309
4	1.5982e-03	1.8383e-03	15.019	1268.39	0.937	1354
6	1.0267e-03	1.2003e-03	16.910	2862.00	1.962	1459
8	7.7654e-04	9.2408e-04	19.000	5172.72	3.722	1390
10	6.7352e-04	8.0776e-04	19.931	9134.71	7.533	1213

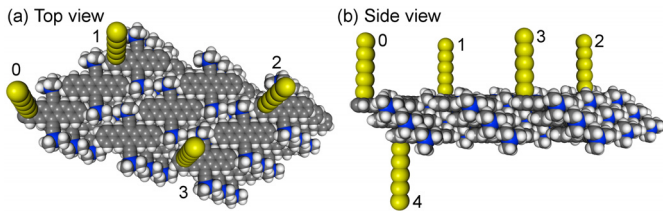


Fig. 6. A modeled structure for the calculation of anisotropy of conductance in pentacene derivatives. The pentacene derivatives are relaxed in advance using period boundary conditions. 27 monomers stacked in three layers are extracted from the relaxed structures and the gold leads are attached for the evaluation of anisotropy of conductance.

Table 3. Orientation dependence of effective conductance of organic crystal calculated by CGDP model. The effective conductance at the Fermi energy (set to $E = -4.5$ eV) of piece of organic crystal in Figure 6 between two leads are compared.

Case	Pairs	$\tilde{T}_{\text{eff}} (2e^2/h)$
1	0-1	8.541e-05
2	0-2	5.897e-06
3	0-3	6.212e-06
4	0-4	1.508e-04

after the relaxation. we compared the effective conductance between different pair of gold leads to investigate the orientation dependence of the conductance.

Table 3 summarizes the effective conductance at the Fermi energy (set to $E = -4.5$ eV) between pairs of gold leads calculated by the CGDP model. The effective conductance between 0th and 4th leads is the highest among 4 pairs of leads. This is easily expected from the shortest distance and strong π - π stacking. On the other hand, in spite of the long distance, the reduction of the effective transmission between 0th and 2nd leads is small. This is due to the decoherence assisted transport as discussed ever in many other studies (for instance, [12,28]). This is how we can apply the CGDP model for the study of the orientation dependence of conductivity in the large molecular systems in various phases (amorphous, crystals, and polymers) with moderate computational cost. Although the original DP model cannot be applied for large systems because of computational costs, the CGDP model allows one to investigate influence of the structural or environmental factors such as disorders, defects, and doping, to the transport [33].

4 Conclusion

In summary, for the study of electron transport through large molecules coupled between contacts under dephasing condition, we have derived the efficient coarse-grained model starting from D'Amato-Pastawski model. Using the stepwise chemical potential profile in weakly coupled molecular wires, the effective conductance including dephasing effect could be calculated efficiently reducing computational time considerably without essential loss of accuracy. This model would give an opportunity to investigate the fundamental charge transporting process through the nanostructures such as organic semiconducting devices and DNAs, where dephasing effect plays an important role.

We gratefully acknowledge support from the German Research Foundation (DFG) within the Cluster of Excellence ‘‘Center for Advancing Electronics Dresden’’ (cfAED). We also gratefully acknowledge for the funds from the European Union (ERDF) and the Free State of Saxony via TP A2 (MolFunc) of the cluster of excellence European Center for Emerging Materials and Processes Dresden (ECEMP), from the European Union (ERDF) and the Free State of Saxony via the ESF project 080942409 InnovaSens. We acknowledge the Center for Information Services and High Performance Computing (ZIH) at the Dresden University of Technology for computational resources. The work at Cordoba received the support of CON-ICET, FoNCyT and SeCyT-UNC.

References

1. Y. Imry, R. Landauer, Rev. Mod. Phys. **71**, S306 (1999)
2. S. Datta, *Electronic Transport in Mesoscopic Systems* (Cambridge University Press, Cambridge, 1995)
3. J.L. D'Amato, H.M. Pastawski, Phys. Rev. B **41**, 7411 (1990)
4. H.M. Pastawski, L.E.F. Foa Torres, E. Medina, Chem. Phys. **281**, 257 (2002)
5. H.M. Pastawski, E. Medina, Rev. Mex. Fis. **47S1**, 1 (2001)
6. R. Golizadeh-Mojarad, S. Datta, Phys. Rev. B **75**, 081301 (2007)
7. J. Qi, N. Edirisinghe, M.G. Rabbani, M.P. Antram, Phys. Rev. B **87**, 085404 (2013)
8. D. Nozaki, Y. Girard, K. Yoshizawa, J. Phys. Chem. C **112**, 17408 (2008)
9. D. Nozaki, C. Gomes da Rocha, H.M. Pastawski, G. Cuniberti, Phys. Rev. B **85**, 155327 (2012)

10. J. Maassen, F. Zahid, H. Guo, Phys. Rev. B **80**, 125423 (2009)
11. M. Zilly, O. Ujsághy, D.E. Wolf, Eur. Phys. J. B **68**, 237 (2009)
12. M. Zilly, O. Ujsághy, M. Woelki, D.E. Wolf, Phys. Rev. B **85**, 075110 (2012)
13. C.J. Cattena, R.A. Bustos-Marín, H.M. Pastawski, Phys. Rev. B **82**, 144201 (2010)
14. M. Mardaani, H. Rabani, A. Esmaceli, Solid State Commun. **151**, 928 (2011)
15. T. Stegmann, M. Zilly, O. Ujsághy, D.E. Wolf, Eur. Phys. J. B **85**, 264 (2012)
16. M. Žnidarič, M. Horvat, Eur. Phys. J. B **86**, 67 (2013)
17. A.K. Felts, W.T. Pollard, R.A. Friesner, J. Phys. Chem. **99**, 2929 (1995)
18. D. Segal, A. Nitzan, M. Ratner, W.B. Davis, J. Phys. Chem. B **104**, 2790 (2000)
19. N. Sergueev, D. Roubtsov, H. Guo, Phys. Rev. Lett. **95**, 146803 (2005)
20. E.G. Petrov, V. May, P. Hänggi, Chem. Phys. **296**, 251 (2004)
21. E.G. Petrov, V. May, J. Phys. Chem. A **105**, 10176 (2001)
22. X. Li, Y. Yan, Phys. Rev. B **65**, 155326 (2001)
23. X.Y. Yu, H.Y. Zhang, P. Han, X.Q. Li, Y. Yan, J. Chem. Phys. **117**, 2180 (2002)
24. T. Stegmann, O. Ujsághy, D.E. Wolf, Eur. Phys. J. B **87**, 30 (2014)
25. M. Büttiker, Phys. Rev. B **32**, 1846 (1985)
26. M. Büttiker, IBM J. Res. Dev. **32**, 317 (1988)
27. M. Kilgour, D. Segal, J. Chem. Phys. **143**, 024111 (2015)
28. C.J. Cattena, L.J. Fernández-Alcázar, R.A. Bustos-Marín, D. Nozaki, H.M. Pastawski, J. Phys.: Condens. Matter **26**, 345304 (2014)
29. E. Rauls, J. Elsner, R. Gutierrez, T. Frauenheim, Solid State Commun. **111**, 459 (1999)
30. T.A. Niehaus, M. Elster, Th. Frauenheim, S. Suhai, J. Mol. Struct. Theochem **541**, 185 (2001)
31. J.E. Anthony, D.L. Eaton, S.R. Parkin, Org. Lett. **4**, 15 (2002)
32. R.C. Haddon, X. Chi, M.E. Itkis, J.E. Anthony, D.L. Eaton, T. Siegrist, C.C. Mattheus, T.T.M. Palstra, J. Phys. Chem. B **106**, 8288 (2002)
33. K.S. Kumar, R.R. Pasula, S. Lim, C.A. Nijhuis, Adv. Mater. **28**, 1824 (2016)