

Quantitative Prediction of the Electro-Mechanical Response in Organic Crystals

Alessandro Landi* Andrea Peluso Alessandro Troisi

Dr. Alessandro Landi, Prof. Andrea Peluso

Dipartimento di Chimica e Biologia

Università di Salerno, I-84084 Fisciano, Salerno, Italy

Email Address:alelandi1@unisa.it

Prof. Alessandro Troisi

Department of Chemistry, University of Liverpool, Liverpool L69 3BX, U.K.

Keywords: *Flexible electronics, charge mobility, electro-mechanical properties, strain*

Organic semiconductors' inherent flexibility makes them appealing for advanced applications such as wearable electronics, e-skins or pressure sensors and can even be used to enhance their intrinsic electronic properties. Unfortunately, these applications for organic materials are currently hindered by the lack of a quantitative understanding of the interplay between their electrical and mechanical properties. In this work, we fill this gap by presenting an accurate methodology able to predict quantitatively the effects of external deformation on the charge transport properties of any organic semiconductors. Three prototypical materials are investigated, showing that the experimental variation of charge carrier mobility with strain is fully reproduced, even in a wide range of deformations applied along different crystal axes. Our results point out that the intrinsic electro-mechanical response of the materials varies by orders of magnitude within the class of organic semiconductors, a difference rationalized observing that the mobility trend is primarily influenced by transfer integrals' variation, rather than by a modification of the crystal phonons. In light of its robustness, accuracy and low computational cost, this protocol represents an ideal tool to quantify the electro-mechanical response in new organic compounds, thus establishing a reliable route for a full exploitation of strain engineering in advanced technologies.

In the last years, increasing attention has been devoted to one of the key features of organic semiconductors, i.e. their inherent flexibility, and to the effects of external deformations on their electrical performances. Indeed, many advanced applications require either materials with a controlled yet marked electro-mechanical response (e.g. pressure sensors[1, 2] and e-skin, a bionic device that can mimic the skin of human beings[3]) or, conversely, materials preserving their electrical performances under mechanical deformation, e.g. in flexible displays or in bioelectronic applications[2, 4], such as bio-integrated circuits[4, 5]. Moreover, inducing strain is a potential mechanism to increase device performances [6], as already demonstrated in the past for inorganic semiconductors[7, 8], where increase of hole and electron mobility (μ) higher than 100% have been reported for silicon transistors under strain. Finally, comparing relative trends in charge mobility as a function of the applied mechanical deformation represents a unique opportunity to validate the models describing charge transport in organic materials, since systematic errors of theories and experiments on the absolute value of the mobility become unimportant.[9] Despite all these opportunities, electro-mechanical properties are not currently exploited in real devices because the relation between the deformation applied and the material performances is still unclear. Indeed, conflicting results about either the magnitude and the direction of the strain mostly affecting the charge mobility have been reported in the literature.

To compare the data reported in different works, we resort to the dimensionless strain factor

$$g_i^j = \frac{\mu_i(\epsilon_j) - \mu_i^0}{\epsilon_j \times \mu_i^0} \times 100, \quad (1)$$

where μ_i^0 is the mobility measured along direction i in the unstrained crystal, while $\mu_i(\epsilon_j)$ is the mobility for the crystal under strain ϵ_j induced along the direction j . This latter quantity is given by $\epsilon_j = (L_j - L_j^0)/L_j^0$, L_j and L_j^0 being the axial length in presence and absence of mechanical deformation, respectively. Notably, several recent experimental studies dealing with the same system, i.e. highly purified rubrene single crystals[10, 11, 12, 13, 14], reported a very wide range of g values, from ≈ 0 to ≈ 250 . To further complicate this picture, while in some experiments[11] it has been found that compression along the principal π -stacking direction (a axis, **Figure 1**) results in a lesser mobility increase than compression along the orthogonal b axis, a recent work[12] reports on the contrary a strong change in mobility when deformation is applied along a axis, while strain along b seems to have no effects at all on the measured mobility.

This broad range of conflicting experimental findings could have several causes, such as the presence of residual defects (grain boundaries, micro-cracks, dislocation) in the crystal or a partial stress relief in the elastomeric substrate over which the crystal is laminated[12]. All these issues prevent the access to the intrinsic electro-mechanical properties of the material and thus represent a significant hindrance for the systematic implementation of strain engineering in advanced technologies. In this respect, the development of a reliable computational protocol could be of great help to overcome these difficulties, since it allows both to separate the intrinsic material response from extrinsic effects in the interpretation of experimental results and could also be used to identify new promising materials. Unfortunately, even though several theoretical works[11, 15, 16] have been performed, a consensus is yet to be reached. For example, some works[11, 15] predict a similar increase in mobility with compressive strain applied either along or perpendicularly to a axis for rubrene single crystals, thus lacking the anisotropy reported in experimental works [10, 11, 12]; on the other hand, another paper[16] predicts variations at odds with experimental findings (mobility decreasing with compression), unless ad-hoc corrections (e.g. extrinsic energetic disorder) are introduced. All these works face the challenge of the high computational cost of phonons for crystals with hundreds of atoms in the unit cell.[16, 17] In this contribution, we tackle all these difficulties which hinder the full exploitation of electro-mechanical properties of organic materials, by developing a fast computational protocol capable to quantitatively predict the mobility variations induced in organic crystals by directional mechanical strains.

Charge carrier mobility is evaluated in the framework of the Transient Localization Theory (TLT) developed by Fratini and Ciuchi[18, 19] which has shown a high level of predictive power[17, 20, 21], leading to results in agreement with other theoretical methods [22, 23, 24] at a much lower computational cost. The TLT builds up on the widely accepted evidence[25] that, in organic materials, the transfer integrals J_{ij} between two neighbouring molecules i and j undergo large fluctuations $\sigma_{ij} = \sqrt{\langle (J_{ij} - \langle J_{ij} \rangle)^2 \rangle}$, giving rise to a dynamic disorder which causes a localization of the instantaneous eigenstates of the lattice. This effect is quantified by evaluating the transfer integrals, the phonons, the local electron-phonon coupling, and the non-local electron-phonon coupling. For the last term we find useful to define the quantity ∇J_{ij} , which is the numerical derivative of the transfer integral with respect to the Cartesian displacement of an atom and measures how sensitive the transfer integral are to deformation in any direction.[26, 27] More in-depth details about the methodology are given in the Supporting Information.

Application of this methodology to orthorhombic rubrene crystal leads to a mobility value along the π -stacking a axis ($15.3 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$) in excellent agreement with experimental reports for highly purified rubrene single crystals [28, 29], thus ensuring the reliability of our approach. We have then turned our attention to the effect on charge mobility of uniaxial strain, which corresponds to any experimental setup preventing a molecular rearrangement along the cell vectors normal to strain, e.g. a clamped or laterally confined crystal, such as in ref. [11]. Uniaxial strain has been introduced by appropriately rescaling the corresponding crystal axis and the molecular positions along the same direction; then the atomic coordinates have been reoptimized while the lattice parameters are held fixed. Our computations on rubrene crystals under strain up to $\pm 1\%$ predict that compression either along a or along b axis results in an increase in mobility, the latter being higher in magnitude (**Figure 1**). This difference is easily rationalized by observing that compression either along a or b results in a similar increase of transfer integrals, but the latter leads to a decrease of the dynamic disorder. Moreover, compression along a leads to a decrease in the intermolecular distances between the molecules along the π -stacking direction, which, as shown in ref. [20], have a trivial effect on the calculated mobility (i.e. the mobility quadratically depends on the intermolecular distances).[19, 20]. Our predictions are in very good agreement with experimental results by Matta et al.[11], inasmuch as we obtain $g_a^a = -7.9\%$ and $g_a^b = -15.6\%$, to be compared with $g_a^a = -6\%$ and $g_a^b = -21\%$ in ref. [11].

So far, our analysis has been performed on a limited range of applied deformation, since rubrene single crystals show fractures for strains larger than $\pm 0.4\%$ [14]; more in general, the vast majority of organic semiconductors have never been analyzed under strain larger than 3% [30]. A remarkable exception is TIPS, for which charge mobility has been measured under both compressive and tensile strain up to 16.7% [31], the widest deformation range reported in the literature to date, showing outstanding

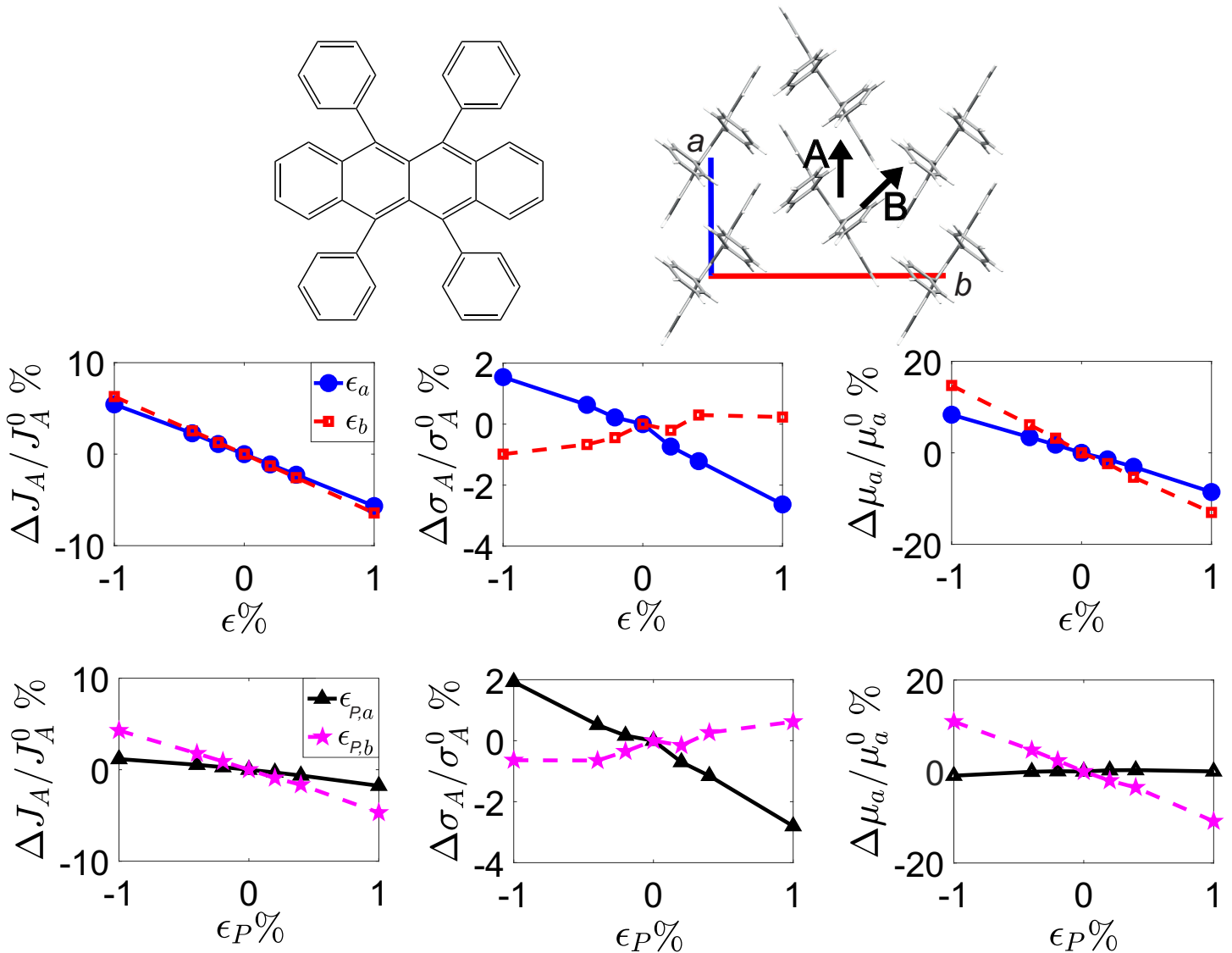


Figure 1: Top: molecular structure of rubrene (left) and pictorial representation of the symmetry-independent charge transfer paths in the ab plane of rubrene crystals (right). Middle: relative variation of J_A (left) σ_A (middle) and μ_a (right) as a function of uniaxial strain applied along the a axis (blue full line with dots) or b axis (red dashed line with squares). Bottom: relative variation of J_A (left) σ_A (middle) and μ_a (right) as a function of uniaxial stress ϵ_P applied along the a axis (black full line with triangles) or b axis (magenta dashed line with stars). J_B , σ_B and μ_b follow a very similar trend (see Supporting Information).

monotonic increase in the charge carrier mobility under compression, while only a little decrease is observed under elongation. Our predicted relative variation of the mobility with strain is in very good agreement with experimental data in ref. [31]: our computations predict a monotonic increase in mobility up to 1979% (experimental 1854%) under 16% compression and a decrease up to 42% (experimental 97%) under 16% elongation. Notably, on the basis of our computed transfer integrals, hopping approaches such as Marcus theory are unable to reproduce the experimental trend, since the mobility is proportional to the square of the transfer integral value and the non-monotonic trend in the transfer integral variation (**Figure 2**) would reflect in a non-monotonic variation of mobility (e.g. 75% decrease with 1% elongation but 358% increase with 5% elongation, see Supporting Information), in clear contradiction with experimental reports [31]. This result confirms that comparing relative trends in charge mobility under the effect of different mechanical deformations is an ideal approach to highlight the main qualitative differences between theories.

Our method also allows the computation of the mobility changes when one applies uniaxial *stress* (ϵ_P), i.e. a uniaxial deformation resulting in a lateral rearrangement of the crystal in the directions perpendic-

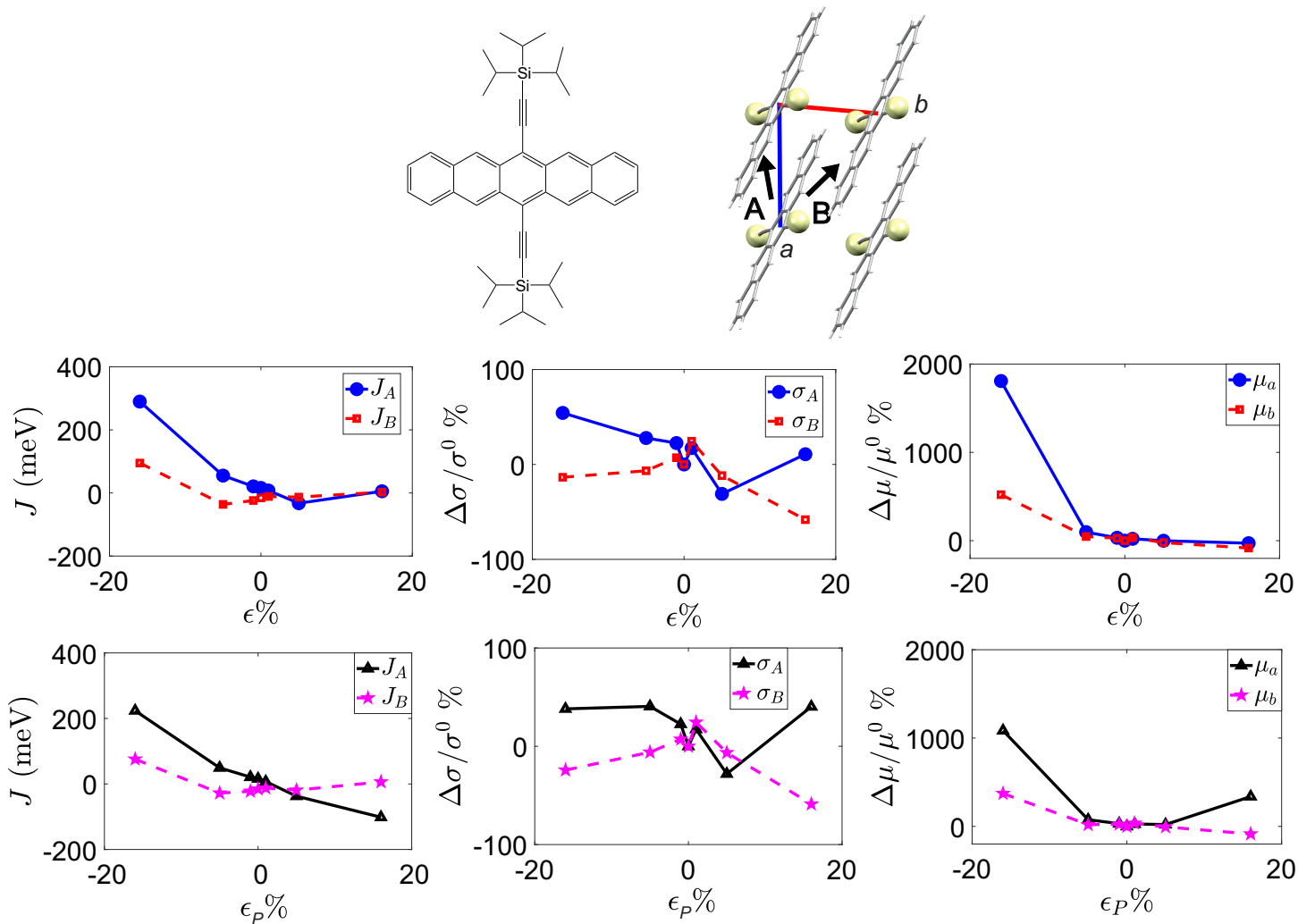


Figure 2: Top: molecular structure of 6,13-bis(triisopropylsilylethynyl)pentacene (TIPS, left) and pictorial representation of the symmetry-independent charge transfer paths in the ab plane of TIPS crystals (right). Middle: variation of J (left) and relative variation of σ (middle) and μ (right) as a function of uniaxial strain applied along the a axis. Bottom: variation of J (left) and relative variation of σ (middle) and μ (right) as a function of uniaxial stress ϵ_P applied along the a axis. J has been shown instead of $\Delta J/J^0$ to highlight the changes in the transfer integral sign. The predicted mobility value for the unstrained system along the π -stacking a axis is $1.56 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, in line with experimental reports [31, 32].

ular to the induced *strain*; this corresponds to any experimental setups where the crystal is laminated above a substrate without any lateral confinement, thus free to rearrange along all spatial directions, as in some works by Frisbie group[33, 34]. In our computations, the uniaxial stress has been introduced by rescaling cell axes and molecular positions along the three lattice directions accordingly to their Poisson ratio ν , defined as the negative of the ratio of transverse strain ϵ_j to axial strain ϵ_i , i.e. $\nu_{i,j} = -\frac{d\epsilon_j}{d\epsilon_i}$. For rubrene, as shown in **Figure 1**, while compression along b axis still results in a mobility enhancement, even though lower in magnitude with respect to the uniaxial strain case, the impact of deformation along a axis is strikingly different, inasmuch as it has almost no effect on the mobility.

Notably, uniaxial stress has a strong impact also on the computed mobility of TIPS: not only it results in a smaller mobility increase with compression but also causes an opposite variation with elongation (**Figure 2**), because of the strikingly different transfer integral values. Thus, our predictions suggest that if lateral deformation is allowed (i.e. the crystals are not clamped), TIPS-based devices display better performances when strained, irrespective of the strain verse (i.e. both under elongation and under compression, the latter resulting in a bigger improvement).

Unlike rubrene and TIPS, some systems are predicted to have mobility along the π -stacking direction very little dependent on either stress or strain, a very appealing feature for their exploitation in flex-

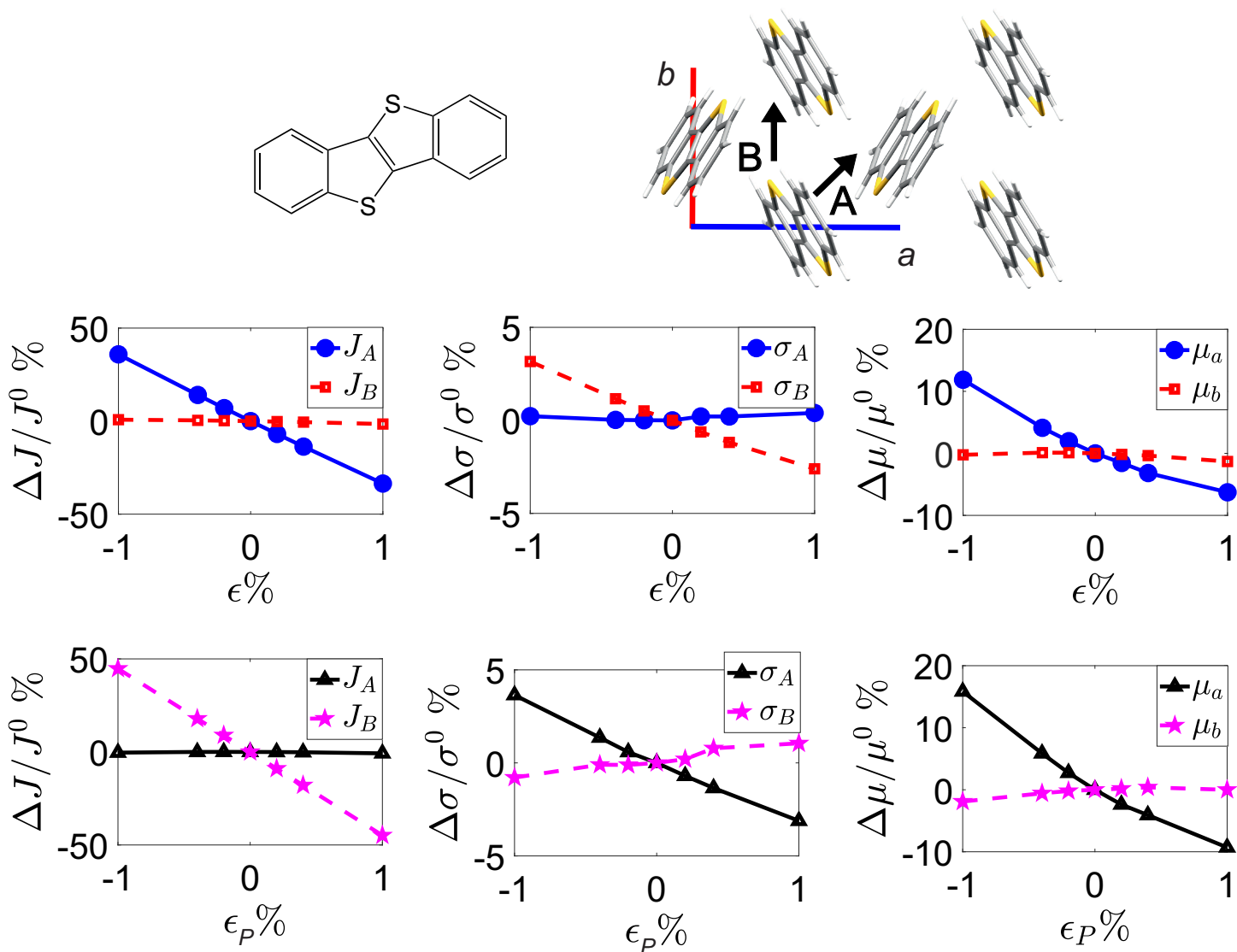


Figure 3: Top: molecular structure of [1]benzothieno[3,2-b][1]benzothiophene (BTBT, left) and pictorial representation of the symmetry-independent charge transfer paths in the ab plane of BTBT crystals (right). Middle: relative variation of J (left), σ (middle) and μ (right) as a function of uniaxial strain applied along the b axis. Bottom: relative variation of J (left), σ (middle) and μ (right) as a function of uniaxial stress ϵ_P applied along the b axis. The predicted mobility value for the unstrained system along the π -stacking b axis is $\mu_b = 2.90 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$.

ible electronics, where indeed a preservation of electrical performances under mechanical deformation is required. As an example, we here consider BTBT, where μ_a , i.e. the mobility perpendicular to the principal π -stacking axis b , displays a steep variation with deformations, while μ_b shows only negligible changes. Moreover, very similar trends are obtained either under uniaxial *strain* or under uniaxial *stress*, despite the non negligible Poisson ratio elements of this material[15]. This behaviour can represent a significant advantage for advanced applications, inasmuch as different experimental setups can be combined in the same device obtaining a similar electro-mechanical response.

Our results point out that electro-mechanical properties vary by orders of magnitude (g ranging from 0 to 2000%) within the class of organic semiconductors, and this is due to the different intrinsic response of the materials, not to device artifacts or other extrinsic effects, as longly disputed in the literature[12, 16]. This finding highlights that there is plenty of room for the identification of new compounds either very sensitive or insensitive to deformation for applications in cutting-edge technologies such as wearable electronics[4], tactile sensors[2], or e-skins[3].

More importantly, our work provides the tools for such quest, by rationalizing the origin of the different electro-mechanical behaviour. Indeed, a common trend emerges for all the materials studied in this

work: the mobility is primarily influenced by the changes in J , while σ determines the absolute value of the mobility but has a minor impact on its relative variation, probably because σ varies slowly with the deformation. Moreover, σ itself appears to be only slightly influenced by the phonons variation (see Supporting Information) but rather it follows the trend in $|\nabla\mathbf{J}|$. If we completely neglect the changes in phonons (i.e. assuming constant phonons for all the range of the investigated strain) we would reproduce the change in mobility within 20% difference. These observations, combined with the small dependence of the results on the inclusion of local electron-phonon coupling highlight that the electro-mechanical behaviour of a new material can be quickly estimated by looking at the transfer integral variation (which can be computed in a few minutes) and, if suitable for the desired application, it can be further investigated evaluating $|\nabla\mathbf{J}|$ while assuming constant phonons over all the strain range to get a reasonable guess of the mobility change. This strategy could also be used for a possible future screening of large materials databases.

In conclusion, in this work we have shown that it is possible to rationalize and predict quantitatively the electro-mechanical responses of organic semiconductors. Application of our protocol to three prototypical compounds allowed to reproduce, for the first time in the literature, all the experimental trends reported in a wide range of strain, being able to capture also the impact of deformations applied along different directions, without the need of introducing any external corrections. Our results show that strongly different intrinsic electro-mechanical responses can be found across various organic semiconductors, indicating that new promising materials for advanced technologies that fully exploit the materials' softness are likely to emerge. When the origin of the mobility changes under mechanical stress is analysed, a common trend is found, where the deformations seem to influence primarily the electronic rather than the vibrational structure of the material. This finding suggests a strategy for the identification of new materials that are either highly sensitive or highly insensitive to mechanical deformations and rationalizes the origin of their different behaviour.

1 Methods

The structures of the molecules studied in this work have been retrieved from the Cambridge Structural Database[35]; 1244265, 975935 and 1570910 are the CSD codes for rubrene, BTBT and TIPS, respectively. Experimental Poisson ratio elements for rubrene has been extracted from ref. [36]. Poisson ratio elements from MD simulations have been extracted from ref.s [37] and [15] for TIPS and BTBT, respectively. Electronic structure, normal modes, and vibrational frequencies have been calculated by employing the self-consistent charge density-functional tight-binding (DFTB) method as implemented in the DFTB+ software package.[38, 39]. The 3-ob-1 Slater-Koster set of parameters have been employed for rubrene and BTBT, while the matsci-0-3 set has been used for TIPS. The atomic positions of the unit cell have been optimized keeping the experimental lattice values fixed, using the conjugate gradient method with a force-threshold criterion of 10^{-8} Hartree per Bohr radius. We have used periodic boundary conditions, employing a $2 \times 2 \times 1$ Monkhorst-Pack k -point sampling scheme, as suggested in previous works[16, 40], and Lennard-Jones dispersion corrections [41, 42]. As shown in the Supporting Information, a higher number of k -points or the use of 3ob-freq Slater-Koster files lead to very similar results. Since DFTB is known to severely underestimate transfer integrals because of the minimal basis set used [26, 43], they have been evaluated at the B3LYP/3-21g* level of theory as implemented in Gaussian 16 [44]. The freely distributed code described in ref. [45] has been used for mobility calculations in the framework of Transient Localization Theory.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

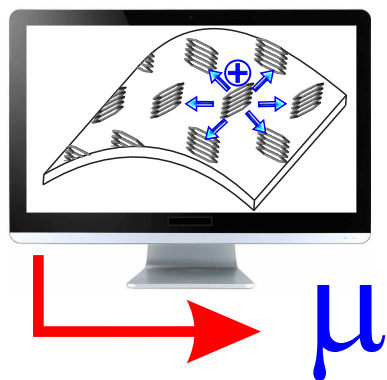
The authors thank Prof. V. Podzorov (Rutgers) and Dr. T. Nematiram (Liverpool) for fruitful discussions. A.L. and A.P. acknowledge the financial support of Università degli Studi di Salerno. A.T. acknowledges the support of EPSRC.

References

- [1] T. Someya, T. Sekitani, S. Iba, Y. Kato, H. Kawaguchi, T. Sakurai, *Proc. Natl. Acad. Sci.* **2004**, *101*, 9966.
- [2] Y. Qian, X. Zhang, L. Xie, D. Qi, B. K. Chandran, X. Chen, W. Huang, *Adv. Mater.* **2016**, *28*, 9243.
- [3] M. Zhu, M. U. Ali, C. Zou, W. Xie, S. Li, H. Meng, *Front. Phys.* **2020**, *16*, 13302.
- [4] T. Q. Trung, N.-E. Lee, *Adv. Mater.* **2017**, *29*, 1603167.
- [5] A. Chortos, Z. Bao, *Mater. Today* **2014**, *17*, 321 .
- [6] Y. Li, J. Wan, D.-M. Smilgies, R. Miller, R. L. Headrick, *Phys. Rev. Research* **2020**, *2*, 033294.
- [7] S. Bedell, A. Khakifirooz, D. Sadana, *MRS Bull.* **2014**, *39*, 131.
- [8] C. K. Maiti, L. K. Bera, S. S. Dey, D. Nayak, N. B. Chakrabarti, *Sol. Stat. Electr.* **1997**, *41*, 1863 .
- [9] T. Nematiram, A. Troisi, *J. Chem. Phys.* **2020**, *152*, 190902.
- [10] T. Morf, T. Mathis, B. Batlogg, Unusual anisotropic response of the charge carrier mobility to uniaxial mechanical strain in rubrene crystals, **2016**, ArXiv:1606.06875v2.
- [11] M. Matta, M. J. Pereira, S. M. Gali, D. Thuau, Y. Olivier, A. Briseno, I. Dufour, C. Ayela, G. Wantz, L. Muccioli, *Mater. Horiz.* **2018**, *5*, 41.
- [12] H. H. Choi, H. T. Yi, J. Tsurumi, J. J. Kim, A. L. Briseno, S. Watanabe, J. Takeya, K. Cho, V. Podzorov, *Adv. Sci.* **2019**, *7*, 1901824.
- [13] M. A. Reyes-Martinez, A. J. Crosby, A. L. Briseno, *Nat. Commun.* **2015**, *6*, 6948.
- [14] H. Wang, L. Deng, Q. Tang, Y. Tong, Y. Liu, *IEEE Electron Device Letters* **2017**, *38*, 1598.
- [15] S. M. Gali, C. Quarti, Y. Olivier, J. Cornil, L. Truffandier, F. Castet, L. Muccioli, D. Beljonne, *J. Mater. Chem. C* **2019**, *7*, 4382.
- [16] M. T. Ruggiero, S. Ciuchi, S. Fratini, G. D'Avino, *J. Phys. Chem. C* **2019**, *123*, 15897.
- [17] T. F. Harrelson, V. Dantanarayana, X. Xie, C. Koshnick, D. Nai, R. Fair, S. A. Nuñez, A. K. Thomas, T. L. Murrey, M. A. Hickner, J. K. Grey, J. E. Anthony, E. D. Gomez, A. Troisi, R. Faller, A. J. Moulé, *Mater. Horiz.* **2019**, *6*, 182.
- [18] S. Ciuchi, S. Fratini, D. Mayou, *Phys. Rev. B* **2011**, *83*, 081202.
- [19] S. Ciuchi, S. Fratini, *Phys. Rev. B* **2012**, *86*, 245201.
- [20] S. Fratini, S. Ciuchi, D. Mayou, G. T. De Laissardière, A. Troisi, *Nat. Mater.* **2017**, *16*, 998.
- [21] T. Nematiram, D. Padula, A. Landi, A. Troisi, *Adv. Funct. Mater.* **2020**, *30* 2001906.
- [22] S. Giannini, A. Carof, M. Ellis, H. Yang, O. G. Ziogos, S. Ghosh, J. Blumberger, *Nat. Commun.* **2019**, *10*, 3843.
- [23] S. Fratini, S. Ciuchi, *Phys. Rev. Res.* **2020**, *2* 013001.
- [24] A. Landi, *J. Phys. Chem. C* **2019**, *123*, 18804.
- [25] A. Troisi, G. Orlandi, *Phys. Rev. Lett.* **2006**, *96*, 086601.
- [26] A. Landi, A. Troisi, *J. Phys. Chem. C* **2018**, *122*, 18336.

- [27] A. Landi, A. Troisi, A. Peluso, *J. Mater. Chem. C* **2019**, *7*, 9665.
- [28] W. Xie, K. A. McGarry, F. Liu, Y. Wu, P. P. Ruden, C. J. Douglas, C. D. Frisbie, *J. Phys. Chem. C* **2013**, *117*, 11522.
- [29] X. Ren, M. J. Bruzek, D. A. Hanifi, A. Schulzetenberg, Y. Wu, C.-H. Kim, Z. Zhang, J. E. Johns, A. Salleo, S. Fratini, A. Troisi, C. J. Douglas, C. D. Frisbie, *Adv. Electron. Mater.* **2017**, *3*, 1700018.
- [30] T. Kubo, R. Häusermann, J. Tsurumi, J. Soeda, Y. Okada, Y. Yamashita, N. Akamatsu, A. Shishido, C. Mitsui, T. Okamoto, S. Yanagisawa, H. Matsui, J. Takeya, *Nat. Commun.* **2016**, *7*, 11156.
- [31] Y. Park, K. S. Park, B. Jun, Y.-E. K. Lee, S. U. Lee, M. M. Sung, *Chem. Mater.* **2017**, *29*, 4072.
- [32] J. Wade, F. Steiner, D. Niedzialek, D. T. James, Y. Jung, D.-J. Yun, D. D. C. Bradley, J. Nelson, J.-S. Kim, *J. Mater. Chem. C* **2014**, *2*, 10110.
- [33] Y. Wu, A. R. Chew, G. A. Rojas, G. Sini, G. Haugstad, A. Belianinov, S. V. Kalinin, H. Li, C. Risko, J.-L. Brédas, A. Salleo, C. D. Frisbie, *Nat. Commun.* **2016**, *7* 10270.
- [34] Z. Zhang, G. Yu, J. Garcia-Barriocanal, Z. Xie, C. D. Frisbie, *ACS Appl. Mater. & Interfaces* **2020**, *12*, 40607.
- [35] C. R. Groom, I. J. Bruno, M. P. Lightfoot, S. C. Ward, *Acta Cryst. B* **2016**, *72*, 171.
- [36] Y. Zhang, D. R. Manke, S. Sharifzadeh, A. L. Briseno, A. Ramasubramaniam, K. J. Koski, *Appl. Phys. Lett.* **2017**, *110*, 071903.
- [37] X. Zheng, H. Geng, Y. Yi, Q. Li, Y. Jiang, D. Wang, Z. Shuai, *Adv. Funct. Mater.* **2014**, *24*, 5531.
- [38] B. Hourahine, B. Aradi, V. Blum, F. Bonafé, A. Buccheri, C. Camacho, C. Cevallos, M. Y. Deshaye, T. Dumitrică, A. Dominguez, S. Ehlert, M. Elstner, T. van der Heide, J. Hermann, S. Irle, J. J. Kranz, C. Köhler, T. Kowalczyk, T. Kubař, I. S. Lee, V. Lutsker, R. J. Maurer, S. K. Min, I. Mitchell, C. Negre, T. A. Niehaus, A. M. N. Niklasson, A. J. Page, A. Pecchia, G. Penazzi, M. P. Persson, J. Řezáč, C. G. Sánchez, M. Sternberg, M. Stöhr, F. Stuckenberg, A. Tkatchenko, V. W.-z. Yu, T. Frauenheim, *J. Chem. Phys.* **2020**, *152*, 124101.
- [39] M. Elstner, D. Porezag, G. Jungnickel, J. Elsner, M. Haugk, T. Frauenheim, S. Suhai, G. Seifert, *Phys. Rev. B* **1998**, *58*, 7260.
- [40] Y. Yi, V. Coropceanu, J.-L. Brédas, *J. Chem. Phys.* **2012**, *137*, 164303.
- [41] A. K. Rappe, C. J. Casewit, K. S. Colwell, W. A. Goddard, W. M. Skiff, *J. Am. Chem. Soc.* **1992**, *114*, 10024.
- [42] A. Landi, R. Borrelli, A. Capobianco, A. Velardo, A. Peluso, *J. Phys. Chem. C* **2018**, *122*, 25849.
- [43] A. Kubas, F. Gajdos, A. Heck, H. Oberhofer, M. Elstner, J. Blumberger, *Phys. Chem. Chem. Phys.* **2015**, *17*, 14342.
- [44] M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, G. Scalmani, V. Barone, G. A. Petersson, H. Nakatsuji, X. Li, M. Caricato, A. V. Marenich, J. Bloino, B. G. Janesko, R. Gomperts, B. Mennucci, H. P. Hratchian, J. V. Ortiz, A. F. Izmaylov, J. L. Sonnenberg, D. Williams-Young, F. Ding, F. Lipparini, F. Egidi, J. Goings, B. Peng, A. Petrone, T. Henderson, D. Ranasinghe, V. G. Zakrzewski, J. Gao, N. Rega, G. Zheng, W. Liang, M. Hada, M. Ehara, K. Toyota, R. Fukuda, J. Hasegawa, M. Ishida, T. Nakajima, Y. Honda, O. Kitao, H. Nakai, T. Vreven, K. Throssell, J. A. Montgomery, Jr., J. E. Peralta, F. Ogliaro, M. J. Bearpark, J. J. Heyd, E. N. Brothers, K. N. Kudin, V. N. Staroverov, T. A. Keith, R. Kobayashi, J. Normand,

Table of Contents



We show that it is possible to fully rationalize the relation between mechanical deformation and electronic properties of organic molecular semiconductors through a suitable and accessible theory, achieving quantitative agreement with experiments across orders-of-magnitude differences. Our protocol provides the tools to identify rapidly new compounds sensitive/insensitive to mechanical deformation for advanced applications such as flexible, wearable, e.g. like e-skins devices with tactile sensing capabilities.

K. Raghavachari, A. P. Rendell, J. C. Burant, S. S. Iyengar, J. Tomasi, M. Cossi, J. M. Millam, M. Klene, C. Adamo, R. Cammi, J. W. Ochterski, R. L. Martin, K. Morokuma, O. Farkas, J. B. Foresman, D. J. Fox, Gaussian 16 Revision C.01, **2016**, Gaussian Inc. Wallingford CT.

[45] T. Nematiram, S. Ciuchi, X. Xie, S. Fratini, A. Troisi, *J. Phys. Chem. C* **2019**, *123*, 6989.