Bibliography

[1] J. A. Luceño-Sánchez, A. M. Díez-Pascual, and R. Peña Capilla, "Materials for Photovoltaics: State of Art and Recent Developments", *Int. J. Mol. Sci.* **20**, 1–42 (2019).

[2] S. Fawzy, A. I. Osman, J. Doran, and D. W. Rooney, "Strategies for mitigation of climate change: a review", *Environ. Chem. Lett.* (2020).

[3] J. Barber, "Photosynthetic energy conversion: natural and artificial", *Chem. Soc. Rev.* **38**, 185–196 (2009).

[4] Y. Firdaus, V. M. Le Corre, S. Karuthedath, W. Liu, A. Markina, W. Huang, S. Chattopadhyay, M. M. Nahid, M. I. Nugraha, Y. Lin, et al., "Long-range exciton diffusion in molecular non-fullerene acceptors", *Nat. Commun.* **11**, 1–10 (2020).

[5] A. Classen, C. L. Chochos, L. Lüer, V. G. Gregoriou, J. Wortmann, A. Osvet, K. Forberich, I. McCulloch, T. Heumüller, and C. J. Brabec, "The role of exciton lifetime for charge generation in organic solar cells at negligible energy-level offsets", *Nat. Energy* **5**, 711–719 (2020).

[6] D. Amarasinghe Vithanage, A. Devižis, V. Abramavičius, Y. Infahsaeng, D. Abramavičius, R. C. I. MacKenzie, P. Keivanidis, A. Yartsev, D. Hertel, J. Nelson, V. Sundström, and V. Gulbinas, "Visualizing charge separation in bulk heterojunction organic solar cells", *Nat. Commun.* **4**, 2334 (2013).

[7] "Charge transport in organic semiconductors", H. Bässler and A. Köhler, in *Unimolecular and supramolecular electronics I*, pages 1–65 (Springer, 2011).

[8] V. Coropceanu, X.-K. Chen, T. Wang, Z. Zheng, and J.-L. Brédas, "Charge-transfer electronic states in organic solar cells", *Nat. Rev. Mater.* **4**, 689–707 (2019).

[9] P. Peumans, S. Uchida, and S. Forrest, "Efficient bulk heterojunction photovoltaic cells using small-molecular-weight organic thin films", *Nature* **425**, 158–162 (2003).

[10] C. J. Brabec, M. Heeney, I. McCulloch, and J. Nelson, "Influence of blend microstructure on bulk heterojunction organic photovoltaic performance", *Chem. Soc. Rev.* **40**, 1185–1199 (2011).

[11] L. Lu, T. Zheng, Q. Wu, A. M. Schneider, D. Zhao, and L. Yu, "Recent Advances in Bulk Heterojunction Polymer Solar Cells", *Chem. Rev.* **115**, 12666–12731 (2015).

[12] H. Kang, G. Kim, J. Kim, S. Kwon, and H. Kim, "Bulk-Heterojunction Organic Solar Cells: Five Core Technologies for Their Commercialization", *Adv. Mater.* **28**, 7821–7861 (2016).

[13] S. Rafiquea, S. M. Abdullah, K. Sulaiman, and M. Iwamoto, "Fundamentals of bulk heterojunction organic solar cells: An overview of stability/degradation issues and strategies for improvement", *Renew. Sust. Energ. Rev.* **84**, 43–53 (2018).

[14] C. Yan, S. Barlow, Z. Wang, H. Yan, A. K.-Y. Jen, S. R. Marder, and X. Zhan, "Non-fullerene acceptors for organic solar cells", *Nat. Rev. Mater.* **3**, 18003 (2018).

[15] K. Weng, L. Ye, L. Zhu, J. Xu, J. Zhou, X. Feng, G. Lu, S. Tan, F. Liu, and Y. Sun, "Optimized active layer morphology toward efficient and polymer batch insensitive organic solar cells", *Nat. Commun.* **11**, 1–9 (2020).

[16] Q. Liu, Y. Jiang, K. Jin, J. Qin, J. Xu, W. Li, J. Xiong, J. Liu, Z. Xiao, K. Sun, S. Yang, X. Zhang, and L. Ding, "18% Efficiency organic solar cells", *Sci. Bull.* **65**, 272–275 (2020).

[17] O. A. Abdulrazzaq, V. Saini, S. Bourdo, E. Dervishi, and A. S. Biris, "Organic Solar Cells: A Review of Materials, Limitations, and Possibilities for Improvement", *Particul. Sci. Technol.* **31**, 427–442 (2013).

[18] S. E. Shaheen, R. Radspinner, N. Peyghambarian, and G. E. Jabbour, "Fabrication of bulk heterojunction plastic solar cells by screen printing", *Appl. Phys. Lett.* **79**, 2996–2998 (2001).

[19] S. H. Park, A. Roy, S. Beaupre, S. Cho, N. Coates, J. S. Moon, D. Moses, M. Leclerc, K. Lee, and A. J. Heeger, "Bulk heterojunction solar cells with internal quantum efficiency approaching 100%", *Nat. Photonics* **3**, 297–302 (2009).

[20] D. Baran, R. S. Ashraf, D. A. Hanifi, M. Abdelsamie, N. Gasparini, J. A. Röhr, S. Holliday, A. Wadsworth, S. Lockett, M. Neophytou, et al., "Reducing the efficiency–stability–cost gap of organic photovoltaics with highly efficient and stable small molecule acceptor ternary solar cells", *Nat. Mater.* **16**, 363–369 (2017).

[21] G. Chamberlain, "Organic solar cells: A review", *Solar Cells* **8**, 47 – 83 (1983).

[22] Z. M. Beiley and M. D. McGehee, "Modeling low cost hybrid tandem photovoltaics with the potential for efficiencies exceeding 20%", *Energy Environ. Sci.* **5**, 9173–9179 (2012).

[23] M. Scharber and N. Sariciftci, "Efficiency of bulk-heterojunction organic solar cells", *Prog. Polym. Sci.* **38**, 1929 – 1940 (2013), Topical issue on Conductive Polymers.

[24] S. Holliday, R. S. Ashraf, A. Wadsworth, D. Baran, S. A. Yousaf, C. B. Nielsen, C.-H. Tan, S. D. Dimitrov, Z. Shang, N. Gasparini, et al., "High-efficiency and air-stable P3HT-based polymer solar cells with a new non-fullerene acceptor", *Nat. Commun.* **7**, 1–11 (2016).

[25] Z. Jianqi, Z. Lingyun, and W. Zhixiang, "Toward Over 15% Power Conversion Efficiency for Organic Solar Cells: Current Status and Perspectives", *Small Methods* **1**, 1700258 (2017).

[26] J. Hou, O. Inganäs, R. H. Friend, and F. Gao, "Organic solar cells based on non-fullerene acceptors", *Nat. Mater.* **17**, 119–128 (2018).

[27] R. Zhou, Z. Jiang, C. Yang, J. Yu, J. Feng, M. A. Adil, D. Deng, W. Zou, J. Zhang, K. Lu, et al., "All-small-molecule organic solar cells with over 14% efficiency by optimizing hierarchical morphologies", *Nat. Commun.* **10**, 1–9 (2019).

[28] J. Xiong, K. Jin, Y. Jiang, J. Qin, T. Wang, J. Liu, Q. Liu, H. Peng, X. Li, A. Sun, et al., "Thiolactone copolymer donor gifts organic solar cells a 16.72% efficiency", *Sci. Bull.* **64**, 1573–1576 (2019).

[29] Y. Cui, H. Yao, J. Zhang, T. Zhang, Y. Wang, L. Hong, K. Xian, B. Xu, S. Zhang, J. Peng, et al., "Over 16% efficiency organic photovoltaic cells enabled by a chlorinated acceptor with increased open-circuit voltages", *Nat. Commun.* **10**, 1–8 (2019).

[30] Y. Cui, H. Yao, L. Hong, T. Zhang, Y. Tang, B. Lin, K. Xian, B. Gao, C. An, P. Bi, W. Ma, and J. Hou, "Organic photovoltaic cell with 17% efficiency and superior processability", *Nat. Sci. Rev.* **7**, 1239–1246 (2019).

[31] L. Perdigón-Toro, H. Zhang, A. Markina, J. Yuan, S. M. Hosseini, C. M. Wolff, G. Zuo, M. Stolterfoht, Y. Zou, F. Gao, D. Andrienko, S. Shoaee, and D. Neher, "Barrierless Free Charge Generation in the High-Performance PM6:Y6 Bulk Heterojunction Non-Fullerene Solar Cell", *Adv. Mater.* **32**, 1906763 (2020).

[32] A. Wadsworth, Z. Hamid, J. Kosco, N. Gasparini, and I. McCulloch, "The Bulk Heterojunction in Organic Photovoltaic, Photodetector, and Photocatalytic Applications", *Adv. Mater.* **32**, 2001763 (2020).

[33] Y. Lin, M. I. Nugraha, Y. Firdaus, A. D. Scaccabarozzi, F. Aniés, A.-H. Emwas, E. Yengel, X. Zheng, J. Liu, W. Wahyudi, E. Yarali, H. Faber, O. M. Bakr, L. Tsetseris, M. Heeney, and T. D. Anthopoulos, "A Simple n-Dopant Derived from Diquat Boosts the Efficiency of Organic Solar Cells to 18.3%", *ACS Energy Lett.* **5**, 3663–3671 (2020).

[34] M. A. Green, E. D. Dunlop, J. Hohl-Ebinger, M. Yoshita, N. Kopidakis, and X. Hao, "Solar cell efficiency tables (version 56)", *Prog Photovolt* **28**, 629–638 (2020).

[35] S. C. Price, A. C. Stuart, L. Yang, H. Zhou, and W. You, "Fluorine substituted conjugated polymer of medium band gap yields 7% efficiency in polymer- fullerene solar cells", *J. Am. Chem. Soc.* **133**, 4625–4631 (2011).

[36] K. H. Hendriks, G. H. Heintges, V. S. Gevaerts, M. M. Wienk, and R. A. Janssen, "High-Molecular-Weight Regular Alternating Diketopyrrolopyrrole-based Terpolymers for Efficient Organic Solar Cells", *Angew. Chem.* **125**, 8499–8502 (2013).

[37] S.-H. Liao, H.-J. Jhuo, Y.-S. Cheng, and S.-A. Chen, "Fullerene derivative-doped zinc oxide nanofilm as the cathode of inverted polymer solar cells with low-bandgap polymer (PTB7-Th) for high performance", *Adv. Mater.* **25**, 4766–4771 (2013).

[38] M. Zhang, X. Guo, W. Ma, H. Ade, and J. Hou, "A large-bandgap conjugated polymer for versatile photovoltaic applications with high performance", *Adv. Mater.* **27**, 4655–4660 (2015).

[39] L. Huo, T. Liu, X. Sun, Y. Cai, A. J. Heeger, and Y. Sun, "Single-junction organic solar cells based on a novel wide-bandgap polymer with efficiency of 9.7%", *Adv. Mater.* **27**, 2938–2944 (2015).

[40] H. Bin, L. Gao, Z.-G. Zhang, Y. Yang, Y. Zhang, C. Zhang, S. Chen, L. Xue, C. Yang, M. Xiao, et al., "11.4% Efficiency non-fullerene polymer solar cells with trialkylsilyl substituted 2D-conjugated polymer as donor", *Nat. Commun.* **7**, 1–11 (2016).

[41] J. Zhao, Y. Li, G. Yang, K. Jiang, H. Lin, H. Ade, W. Ma, and H. Yan, "Efficient organic solar cells processed from hydrocarbon solvents", *Nat. Energy* **1**, 1–7 (2016).

[42] Y. Jin, Z. Chen, S. Dong, N. Zheng, L. Ying, X.-F. Jiang, F. Liu, F. Huang, and Y. Cao, "A Novel Naphtho [1, 2-c: 5, 6-c] Bis ([1, 2, 5] Thiadiazole)-Based Narrow-Bandgap π-Conjugated Polymer with Power Conversion Efficiency Over 10%", *Adv. Mater.* **28**, 9811–9818 (2016).

[43] C. Sun, F. Pan, H. Bin, J. Zhang, L. Xue, B. Qiu, Z. Wei, Z.-G. Zhang, and Y. Li, "A low cost and high performance polymer donor material for polymer solar cells", *Nat. Commun.* **9**, 1–10 (2018).

[44] H. Fu, Z. Wang, and Y. Sun, "Polymer Donors for High-Performance Non-Fullerene Organic Solar Cells", *Angew. Chem. Inter. Ed.* **58**, 4442–4453 (2019).

[45] K. Jin, Z. Xiao, and L. Ding, "D18, an eximious solar polymer!", *J. Semicond.* **42**, 010502–010502 (2021).

[46] J. Qin, L. Zhang, C. Zuo, Z. Xiao, Y. Yuan, S. Yang, F. Hao, M. Cheng, K. Sun, Q. Bao, et al., "A chlorinated copolymer donor demonstrates a 18.13% power conversion efficiency", *J. Semicond.* **42**, 010501–010501 (2021).

[47] G. Zhang, J. Zhao, P. C. Y. Chow, K. Jiang, J. Zhang, Z. Zhu, J. Zhang, F. Huang, and H. Yan, "Nonfullerene Acceptor Molecules for Bulk Heterojunction Organic Solar Cells", *Chem. Rev.* **118**, 3447–3507 (2018).

[48] G. Yu, J. Gao, J. C. Hummelen, F. Wudl, and A. J. Heeger, "Polymer photovoltaic cells: enhanced efficiencies via a network of internal donor-acceptor heterojunctions", *Science* **270**, 1789–1791 (1995).

[49] R. Giridharagopal and D. S. Ginger, "Characterizing Morphology in Bulk Heterojunction Organic Photovoltaic Systems", *J. Phys. Chem. Lett.* **1**, 1160–1169 (2010).

[50] A. Karki, J. Vollbrecht, A. L. Dixon, N. Schopp, M. Schrock, G. N. M. Reddy, and T.-Q. Nguyen, "Understanding the High Performance of over 15% Efficiency in Single-Junction Bulk Heterojunction Organic Solar Cells", *Adv. Mater.* **31**, 1903868 (2019).

[51] T. Shan, Y. Zhang, Y. Wang, Z. Xie, Q. Wei, J. Xu, M. Zhang, C. Wang, Q. Bao, X. Wang, et al., "Universal and versatile morphology engineering via hot fluorous solvent soaking for organic bulk heterojunction", *Nat. Commun.* **11**, 1–11 (2020).

[52] H. Oberhofer, K. Reuter, and J. Blumberger, "Charge transport in molecular materials: An assessment of computational methods", *Chem. Rev.* **117**, 10319–10357 (2017).

[53] J. Kirkpatrick, V. Marcon, J. Nelson, K. Kremer, and D. Andrienko, "Charge mobility of discotic mesophases: a multiscale quantum/classical study", *Phys. Rev. Lett.* **98**, 227402 (2007).

[54] J. Nelson, J. J. Kwiatkowski, J. Kirkpatrick, and J. M. Frost, "Modeling charge transport in organic photovoltaic materials", *Acc. Chem. Res.* **42**, 1768–1778 (2009).

[55] R. C. I. MacKenzie, J. M. Frost, and J. Nelson, "A numerical study of mobility in thin films of fullerene derivatives", *J. Chem. Phys.* **132**, 064904 (2010).

[56] V. Ruehle, J. Kirkpatrick, and D. Andrienko, "A multiscale description of charge transport in conjugated oligomers", *J. Chem. Phys.* **132**, 134103 (2010).

[57] V. Ruehle, A. Lukyanov, F. May, M. Schrader, T. Vehoff, J. Kirkpatrick, B. Baumeier, and D. Andrienko, "Microscopic simulations of charge transport in disordered organic semiconductors", *J. Chem. Theory Comput.* **7**, 3335–3345 (2011).

[58] "Simulations of morphology and charge transport in supramolecular organic materials", D. Andrienko, in *Supramolecular Materials for Opto-Electronics*, edited by N. Koch, RSC Smart Materials Series, chapter 9, pages 309–362 (Royal Society of Chemistry, 2015).

[59] N. L. Doltsinis, J. Bachmann, T. Koch, C. Schwermann, and T. Winands, "Trendbericht Theoretische Chemie 2017: Optimierung organischer Solarzellen mit Multiskalensimulationen", *Nachr. Chem.* **66**, 320–324 (2018).

[60] D. T. Gillespie, "A general method for numerically simulating the stochastic time evolution of coupled chemical reactions", *J. Chem. Phys.* **22**, 403 – 434 (1976).

[61] D. T. Gillespie, "Exact stochastic simulation of coupled chemical reactions", *J. Phys. Chem.* **81**, 2340–2361 (1977).

[62] H. Qian, Z. Wang, W. Yue, and D. Zhu, "Exceptional Coupling of Tetrachloroperylene Bisimide: Combination of Ullmann Reaction and C-H Transformation", *J. Am. Chem. Soc.* **129**, 10664–10665 (2007).

[63] K. Ditte, W. Jiang, T. Schemme, C. Denz, and Z. Wang, "Innovative Sensitizer DIPBI Outperforms PCBM", *Adv. Mater.* **24**, 2104–2108 (2012).

[64] T. Winands, M. Böckmann, T. Schemme, P.-M. T. Ly, D. H. de Jong, Z. Wang, C. Denz, A. Heuer, and N. L. Doltsinis, "P3HT:DIPBI bulk heterojunction solar cells: morphology and electronic structure probed by multiscale simulation and UV/vis spectroscopy", *Phys. Chem. Chem. Phys.* **18**, 6217–6227 (2016).

[65] S. J. Marrink, H. J. Risselada, S. Yefimov, D. P. Tieleman, and A. H. De Vries, "The MARTINI force field: coarse grained model for biomolecular simulations", *J. Phys. Chem. B* **111**, 7812–7824 (2007).

[66] S. J. Marrink and D. P. Tieleman, "Perspective on the Martini model", *Chem. Soc. Rev.* **42**, 6801–6822 (2013).

[67] T. A. Wassenaar, K. Pluhackova, R. A. Böckmann, S. J. Marrink, and D. P. Tieleman, "Going backward: a flexible geometric approach to reverse transformation from coarse grained to atomistic models", *J. Chem. Theory Comput.* **10**, 676–690 (2014).

[68] M. E. Casida, *Time-Dependent Density Functional Response Theory for Molecules*, volume 1 (D. P. Chong, World Scientific, Singapore, 1995).

[69] M. Böckmann, T. Schemme, D. H. de Jong, C. Denz, A. Heuer, and N. L. Doltsinis, "Structure of P3HT crystals, thin films, and solutions by UV/Vis spectral analysis", *Phys. Chem. Chem. Phys.* **17**, 28616–28625 (2015).

[70] M. Böckmann and N. L. Doltsinis, "Can Excited Electronic States of Macromolecules with Extended Pi-Systems be Reliably Predicted? A Case Study on P3HT", *Front. Mater.* **2**, 25 (2015).

[71] C. Schwermann and N. L. Doltsinis, "Exciton transfer free energy from Car-Parrinello molecular dynamics", *Phys. Chem. Chem. Phys.* **22**, 10526–10535 (2020).

[72] T. Koch, J. Bachmann, T. Lettmann, and N. L. Doltsinis, "Multiscale modelling of charge transport in P3HT:DIPBI bulk heterojunction organic solar cells", *Phys. Chem. Chem. Phys.* (2021), *submitted for publication.*

[73] E. W. Dijkstra, "A Note on Two Problems in Connexion with Graphs", *Numer. Math.* **1**, 269–271 (1959).

[74] "Quantenmechanische Berechnungen zu Ladungstransferprozessen in organischen Halbleitern", F. Lengers, Bachelor thesis, Westfälische Wilhelms-Universität Münster, 2014.

[75] "Ab-initio-Berechnungen von Ladungstransferintegralen in organischen Solarzellen", T. Lettmann, Master thesis, Westfälische Wilhelms-Universität Münster, 2016.

[76] J. Sanning, L. Stegemann, P. R. Ewen, C. Schwermann, C. G. Daniliuc, D. Zhang, N. Lin, L. Duan, D. Wegner, N. L. Doltsinis, and C. A. Strassert, "Colour-tunable

asymmetric cyclometalated Pt(ii) complexes and STM-assisted stability assessment of ancillary ligands for OLEDs", *J. Mater. Chem. C* **4**, 2560–2565 (2016).

[77] J. Ren, M. Cnudde, D. Brünink, S. Buss, C. G. Daniliuc, L. Liu, H. Fuchs, C. A. Strassert, H.-Y. Gao, and N. L. Doltsinis, "On-Surface Reactive Planarization of Pt(II) Complexes", *Angew. Chem. Int. Ed.* **58**, 15396–15400 (2019).

[78] "Kinetische Monte Carlo Simulation der Ladungsträger in organischen Solarzellen", J. Bachmann, Master thesis, Westfälische Wilhelms-Universität Münster, 2016.

[79] N. L. Doltsinis, "Aktuelle Aspekte der Theoretischen Chemie und Physik: Ab initio Molekulardynamik", (2014).

[80] W. Kolsos, "Adiabatic Approximation and Its Accuracy", *Adv. Quant. Chem.* **5**, 99 – 133 (1970).

[81] W. Kutzelnigg, "The adiabatic approximation I. The physical background of the Born-Handy ansatz", *Mol. Phys.* **90**, 909–916 (1997).

[82] M. Born and R. Oppenheimer, "Zur Quantentheorie der Molekeln", *Ann. Phys.* **389**, 457–484 (1927).

[83] D. Marx and J. Hutter, *Ab Initio Molecular Dynamics: Basic Theory and Advanced Methods*, volume 1 (Cambridge University Press, Cambridge, 1 edition, 2009).

[84] T. D. Kühne, "Second generation Car-Parrinello molecular dynamics", *WIREs Comput. Mol. Sci.* **4**, 391–406 (2014).

[85] M. P. Allen and D. J. Tildesley, *Computer Simulation of Liquids* (Oxford University Press, Oxford, 2 edition, 2017).

[86] W. Ritz, "Über eine neue Methode zur Lösung gewisser Variationsprobleme der mathematischen Physik.", *J. für die Reine und Angew. Math.* **135**, 1–61 (1909).

[87] J. C. Slater, "The Theory of Complex Spectra", *Phys. Rev.* **34**, 1293–1322 (1929).

[88] D. R. Hartree, "The Wave Mechanics of an Atom with a Non-Coulomb Central Field. Part I. Theory and Methods", *Math. Proc. Camb. Philos. Soc.* **24**, 89 (1928).

[89] V. Fock, "Näherungsmethode zur Lösung des quantenmechanischen Mehrkörperproblems", *Z. Phys.* **61**, 126–148 (1930).

[90] A. Szabo and N. S. Ostlund, *Modern Quantum Chemistry: Introduction to Advanced Electronic Structure Theory* (Dover Publications, Inc., Mineola, first edition, 1996).

[91] P. Hohenberg and W. Kohn, "Inhomogeneous Electron Gas", *Phys. Rev. B* **136**, 864–871 (1964).

[92] W. Kohn and L. J. Sham, "Self-Consistent Equations Including Exchange and Correlation Effects", *Phys. Rev. A* **140**, 1133–1138 (1965).

[93] J. Hutter, Dichtefunktionaltheorie, 2004.

[94] D. M. Ceperley and B. J. Alder, "Ground State of the Electron Gas by a Stochastic Method", *Phys. Rev. Lett.* **45**, 566–569 (1980).

[95] S. H. Vosko, L. Wilk, and M. Nusair, "Accurate spin-dependent electron liquid correlation energies for local spin density calculations: a critical analysis", *Can. J. Phys,* **58**, 1200–1211 (1980).

[96] E. G. Moroni, G. Kresse, J. Hafner, and J. Furthmüller, "Ultrasoft pseudopotentials applied to magnetic Fe, Co, and Ni: From atoms to solids", *Phys. Rev. B* **56**, 15629–15646 (1997).

[97] U. von Barth and L. Hedin, "A local exchange-correlation potential for the spin polarized case. i", *J. Phys. C* **5**, 1629–1642 (1972).

[98] A. D. Becke, "Density-functional exchange-energy approximation with correct asymptotic behavior", *Phys. Rev. A* **38**, 3098–3100 (1988).

[99] C. Lee, W. Yang, and R. G. Parr, "Development of the Colle-Salvetti correlation-energy formula into a functional of the electron density", *Phys. Rev. B* **37**, 785–789 (1988).

[100] J. P. Perdew, K. Burke, and M. Ernzerhof, "Generalized Gradient Approximation Made Simple", *Phys. Rev. Lett.* **77**, 3865–3868 (1996).

[101] C. Adamo and V. Barone, "Toward reliable density functional methods without adjustable parameters: The PBE0 model", *J. Chem. Phys.* **110**, 6158–6170 (1999).

[102] A. D. Becke, "Density-functional thermochemistry. III. The role of exact exchange", *J. Chem. Phys.* **98**, 5648–5652 (1993).

[103] T. Van Voorhis and G. E. Scuseria, "A novel form for the exchange-correlation energy functional", *J. Chem. Phys.* **109**, 400–410 (1998).

[104] Y. Zhao, N. E. Schultz, and D. G. Truhlar, "Exchange-correlation functional with broad accuracy for metallic and nonmetallic compounds, kinetics, and noncovalent interactions", *J. Chem. Phys.* **123**, 161103 (2005).

[105] P. Hao, J. Sun, B. Xiao, A. Ruzsinszky, G. I. Csonka, J. Tao, S. Glindmeyer, and J. P. Perdew, "Performance of meta-GGA Functionals on General Main Group Thermochemistry, Kinetics, and Noncovalent Interactions", *J. Chem. Theory Comput.* **9**, 355–363 (2013).

[106] D. J. Tozer and N. C. Handy, "Improving virtual Kohn-Sham orbitals and eigenvalues: Application to excitation energies and static polarizabilities", *J. Chem. Phys.* **109**, 10180–10189 (1998).

[107] M. E. Casida, C. Jamorski, K. C. Casida, and D. R. Salahub, "Molecular excitation energies to high-lying bound states from time-dependent density-functional response theory: Characterization and correction of the time-dependent local density approximation ionization threshold", *J. Chem. Phys.* **108**, 4439–4449 (1998).

[108] T. Yoshihiro, T. Takao, Y. Susumu, Y. Takeshi, and H. Kimihiko, "A long-range-corrected time-dependent density functional theory", *J. Chem. Phys.* **120**, 8425–8433 (2004).

[109] H. Iikura, T. Tsuneda, T. Yanai, and K. Hirao, "A long-range correction scheme for generalized-gradient-approximation exchange functionals", *J. Chem. Phys.* **115**, 3540–3544 (2001).

[110] T. Yanai, D. P. Tew, and N. C. Handy, "A new hybrid exchange-correlation functional using the Coulomb-attenuating method (CAM-B3LYP)", *Chem. Phys. Lett.* **393**, 51–57 (2004).

[111] O. A. Vydrov and G. E. Scuseria, "Assessment of a long-range corrected hybrid functional", *J. Chem. Phys.* **125**, 234109 (2006).

[112] T. M. Henderson, A. F. Izmaylov, G. Scalmani, and G. E. Scuseria, "Can short-range hybrids describe long-range-dependent properties?", *J. Chem. Phys.* **131**, 044108 (2009).

[113] A. Kubas, F. Hoffmann, A. Heck, H. Oberhofer, M. Elstner, and J. Blumberger, "Electronic couplings for molecular charge transfer: Benchmarking CDFT, FODFT, and FODFTB against high-level ab initio calculations", *J. Chem. Phys.* **140**, 104105 (2014).

[114] Q. Wu and T. Van Voorhis, "Extracting electron transfer coupling elements from constrained density functional theory", *J. Chem. Phys.* **125**, 164105 (2006).

[115] T. Van Voorhis, T. Kowalczyk, B. Kaduk, L.-P. Wang, C.-L. Cheng, and Q. Wu, "The Diabatic Picture of Electron Transfer, Reaction Barriers, and Molecular Dynamics", *Annu. Rev. Phys. Chem.* **61**, 149–170 (2010).

[116] H. Oberhofer and J. Blumberger, "Electronic coupling matrix elements from charge constrained density functional theory calculations using a plane wave basis set", *J. Chem. Phys.* **133**, 244105 (2010).

[117] F. L. Hirshfeld, "Bonded-atom fragments for describing molecular charge densities", *Theor. Chem. Acc.* **44**, 129–138 (1977).

[118] R. Eisenschitz and F. London, "Über das Verhältnis der van der Waalsschen Kräfte zu den homöopolaren Bindungskräften", *Z. Phys.* **60**, 491–527 (1930).

[119] S. Grimme, "Semiempirical GGA-Type Density Functional Constructed with a Long-Range Dispersion Correction", *J. Comput. Chem.* **27**, 1787–1799 (2006).

[120] S. Grimme, J. Antony, S. Ehrlich, and H. Krieg, "A consistent and accurate ab initio parametrization of density functional dispersion correction (DFT-D) for the 94 elements H-Pu", *J. Chem. Phys.* **132**, 154104 (2010).

[121] S. Grimme, S. Ehrlich, and L. Goerigk, "Effect of the damping function in dispersion corrected density functional theory", *J. Comp. Chem.* **32**, 1456–1465 (2011).

[122] E. Caldeweyher, C. Bannwarth, and S. Grimme, "Extension of the D3 dispersion coefficient model", *J. Chem. Phys.* **147**, 034112 (2017).

[123] A. Koide, "A new expansion for dispersion forces and its application", *J. Phys. B* **9**, 3173 (1976).

[124] A. D. Becke and E. R. Johnson, "A density-functional model of the dispersion interaction", *J. Chem. Phys.* **123**, 154101 (2005).

[125] "Time-Dependent Density Functional Theory", N. L. Doltsinis, in *Computational Nanoscience: Do It Yourself*, pages 357–373 (J. Grotendorst and S. Blügel and D. Marx, Jülich, NIC Series, 31 edition, 2006).

[126] E. Runge and E. K. U. Gross, "Density-Functional Theory for Time-Dependent Systems", *Phys. Rev. Lett.* **52**, 997–1000 (1984).

[127] M. E. Casida, "Time-dependent density-functional theory for molecules and molecular solids", *J. Mol. Struc.: THEOCHEM* **914**, 3–18 (2009).

[128] T. A. Niehaus, T. Hofbeck, and H. Yersin, "Charge-transfer excited states in phosphorescent organo-transition metal compounds: a difficult case for time dependent density functional theory?", *RSC Adv.* **5**, 63318–63329 (2015).

[129] A. Dreuw and M. Head-Gordon, "Failure of Time-Dependent Density Functional Theory for Long-Range Charge-Transfer Excited States: The Zincbacteriochlorin–Bacteriochlorin and Bacteriochlorophyll–Spheroidene Complexes", *J. Am. Chem. Soc.* **126**, 4007–4016 (2004).

[130] J. Autschbach, "Computing chiroptical properties with first-principles theoretical methods: Background and illustrative examples", *Chirality* **21**, 116–152 (2009).

[131] I. Warnke and F. Furche, "Circular dichroism: electronic", *WIREs Comput. Mol. Sci.* **2**, 150–166 (2012).

[132] A. Cotton, "Absorption inégale des rayons circulaires droit et gauche dans certains corps actifs.", *C. R. Acad. Sci.* **120**, 989–991 (1895).

[133] A. Cotton, "Recherches sur l'absorption et la dispersion de la lumiére par les milieux doués du pouvoir rotatoire", *Ann. Chim. Phys.* **8**, 347–432 (1896).

[134] J. Autschbach, T. Ziegler, S. J. A. van Gisbergen, and E. J. Baerends, "Chiroptical properties from time-dependent density functional theory. I. Circular dichroism spectra of organic molecules", *J. Chem. Phys.* **116**, 6930–6940 (2002).

[135] J. Autschbach and T. Ziegler, "Calculating molecular electric and magnetic properties from time-dependent density functional response theory", *J. Chem. Phys.* **116**, 891–896 (2002).

[136] S. Wilde, D. Ma, T. Koch, A. Bakker, D. Gonzalez-Abradelo, L. Stegemann, C. G. Daniliuc, H. Fuchs, H. Gao, N. L. Doltsinis, L. Duan, and C. A. Strassert, "Toward Tunable Electroluminescent Devices by Correlating Function and Submolecular Structure in 3D Crystals, 2D-Confined Monolayers, and Dimers", *ACS Appl. Mater. Interfaces* **10**, 22460–22473 (2018).

[137] V. Barone, J. Bloino, M. Biczysko, and F. Santoro, "Fully Integrated Approach to Compute Vibrationally Resolved Optical Spectra: From Small Molecules to Macrosystems", *J. Chem. Theory Comput.* **5**, 540–554 (2009).

[138] J. Franck and E. G. Dymond, "Elementary processes of photochemical reactions", *Trans. Faraday Soc.* **21**, 536–542 (1926).

[139] E. Condon, "A Theory of Intensity Distribution in Band Systems", *Phys. Rev.* **28**, 1182–1201 (1926).

[140] V. Barone, J. Bloino, and M. Biczysko, "Vibrationally-resolved electronic spectra in GAUSSIAN 09", *GAUSSIAN 09 Revision A.02* (2009).

[141] D. Porezag, T. Frauenheim, T. Köhler, G. Seifert, and R. Kaschner, "Construction of tight-binding-like potentials on the basis of density-functional theory: Application to carbon", *Phys. Rev. B* **51**, 12947–12957 (1995).

[142] M. Gaus, Q. Cui, and M. Elstner, "DFTB3: Extension of the Self-Consistent-Charge Density-Functional Tight-Binding Method (SCC-DFTB)", *J. Chem. Theory Comput.* **7**, 931–948 (2011).

[143] P. Koskinen and V. Mäkinen, "Density-functional tight-binding for beginners", *Comp. Mater. Sci.* **47**, 237–253 (2009).

[144] J. C. Slater and G. F. Koster, "Simplified LCAO Method for the Periodic Potential Problem", *Phys. Rev.* **94**, 1498–1524 (1954).

[145] Format of the v1.0 Slater-Koster files, `http://www.dftb.org/fileadmin/DFTB/public/misc/slakoformat.pdf`, Accessed: 2019-08-01.

[146] J. F. Janak, "Proof that $\frac{\partial E}{\partial n_i} = \epsilon$ in density-functional theory", *Phys. Rev. B* **18**, 7165–7168 (1978).

[147] M. Elstner, P. Hobza, T. Frauenheim, S. Suhai, and E. Kaxiras, "Hydrogen bonding and stacking interactions of nucleic acid base pairs: A density-functional-theory based treatment", *J. Chem. Phys.* **114**, 5149–5155 (2001).

[148] C. Köhler, G. Seifert, and T. Frauenheim, "Density functional based calculations for Fen (n≤32)", *Chem. Phys.* **309**, 23–31 (2005).

[149] T. A. Niehaus, D. Heringer, B. Torralva, and T. Frauenheim, "Importance of electronic self-consistency in the TDDFT based treatment of nonadiabatic molecular dynamics", *Eur. Phys. J. D* **35**, 467–477 (2005).

[150] T. A. Niehaus, S. Suhai, F. Della Sala, P. Lugli, M. Elstner, G. Seifert, and T. Frauenheim, "Tight-binding approach to time-dependent density-functional response theory", *Phys. Rev. B* **63**, 085108 (2001).

[151] J. J. Kranz, M. Elstner, B. Aradi, T. Frauenheim, V. Lutsker, A. D. Garcia, and T. A. Niehaus, "Time-Dependent Extension of the Long-Range Corrected Density Functional Based Tight-Binding Method", *J. Chem. Theory Comput.* **13**, 1737–1747 (2017).

[152] F. Jensen, *Introduction to Computational Chemistry* (John Wiley & Sons, New York, 2 edition, 2007).

[153] W. J. Hehre, R. F. Stewart, and J. A. Pople, "Self-Consistent Molecular-Orbital Methods. I. Use of Gaussian Expansions of Slater-Type Atomic Orbitals", *J. Chem. Phys.* **51**, 2657–2664 (1969).

[154] M. J. S. Dewar and W. Thiel, "Ground states of molecules. 38. The MNDO method. Approximations and parameters", *J. Am. Chem. Soc.* **99**, 4899–4907 (1977).

[155] M. J. S. Dewar, E. G. Zoebisch, E. F. Healy, and J. J. P. Stewart, "Development and use of quantum mechanical molecular models. 76. AM1: a new general purpose quantum mechanical molecular model", *J. Am. Chem. Soc.* **107**, 3902–3909 (1985).

[156] J. J. P. Stewart, "Optimization of parameters for semiempirical methods I. Method", *J. Comp. Chem.* **10**, 209–220 (1989).

[157] J. Ridley and M. Zerner, "An intermediate neglect of differential overlap technique for spectroscopy: Pyrrole and the azines", *Theor. Chim. Acta* **32**, 111–134 (1973).

[158] J. D. Da Motta Neto and M. C. Zerner, "New parametrization scheme for the resonance integrals (H$\mu\nu$) within the INDO/1 approximation. Main group elements", *Int. J. Quant. Chem.* **81**, 187–201 (2001).

[159] J. C. Slater, "Atomic Shielding Constants", *Phys. Rev.* **36**, 57–64 (1930).

[160] J. B. Collins, P. von R. Schleyer, J. S. Binkley, and J. A. Pople, "Self-consistent molecular orbital methods. XVII. Geometries and binding energies of second-row molecules. A comparison of three basis sets", *J. Chem. Phys.* **64**, 5142–5151 (1976).

[161] T. H. Dunning and P. J. Hay, *Gaussian Basis Sets for Molecular Calculations*, volume 3 (H. F. Schaefer, Plenum, New York, 1977).

[162] E. R. Davidson and D. Feller, "Basis set selection for molecular calculations", *Chem. Rev.* **86**, 681–696 (1986).

[163] R. Ditchfield, W. J. Hehre, and J. A. Pople, "Self–Consistent Molecular–Orbital Methods. IX. An Extended Gaussian–Type Basis for Molecular–Orbital Studies of Organic Molecules", *J. Chem. Phys.* **54**, 724–728 (1971).

[164] F. Bloch, "Über die Quantenmechanik der Elektronen in Kristallgittern", *Z. Phys.* **52**, 555–600 (1929).

[165] L. Brillouin, "Les électrons libres dans les métaux et le role des réflexions de Bragg", *J. Phys. Radium* **1**, 377–400 (1930).

[166] D. Vanderbilt, "Soft self-consistent pseudopotentials in a generalized eigenvalue formalism", *Phys. Rev. B* **41**, 7892–7895 (1990).

[167] K. Laasonen, A. Pasquarello, R. Car, C. Lee, and D. Vanderbilt, "Car-Parrinello molecular dynamics with Vanderbilt ultrasoft pseudopotentials", *Phys. Rev. B* **47**, 10142–10153 (1993).

[168] N. Troullier and J. L. Martins, "Efficient pseudopotentials for plane-wave calculations", *Phys. Rev. B* **43**, 1993–2006 (1991).

[169] H. Oberhofer and J. Blumberger, "Charge constrained density functional molecular dynamics for simulation of condensed phase electron transfer reactions", *J. Chem. Phys.* **131**, 064101 (2009).

[170] L. Verlet, "Computer 'Experiments' on Classical Fluids. I. Thermodynamical Properties of Lennard-Jones Molecules", *Phys. Rev.* **159**, 98–103 (1967).

[171] R. Car and M. Parrinello, "Unified Approach for Molecular Dynamics and Density-Functional Theory", *Phys. Rev. Lett.* **55**, 2471–2474 (1985).

[172] P. Tangney, "On the theory underlying the Car-Parrinello method and the role of the fictitious mass parameter", *J. Chem. Phys.* **124**, 044111 (2006).

[173] G. Sutmann, *Classical Molecular Dynamics*, volume 10, pages 215–220 (John von Neumann Institute for Computing, Jülich, NIC Series, 2002).

[174] J. E. Jones and S. Chapman, "On the determination of molecular fields. I. From the variation of the viscosity of a gas with temperature", *Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character* **106**, 441–462 (1924).

[175] J. E. Jones, "On the Determination of Molecular Fields. II. From the Equation of State of a Gas", *Proceedings of the Royal Society of London Series A* **106**, 463–477 (1924).

[176] P. P. Ewald, "Die Berechnung optischer und elektrostatischer Gitterpotentiale", *Ann. Phys.* **369**, 253–287 (1921).

[177] J. W. Cooley and J. W. Tukey, "An algorithm for the machine calculation of complex Fourier series", *Math. Comp.* **19**, 297–301 (1965).

[178] H. Berendsen and W. van Gunsteren, "Practical Algorithms for Dynamic Simulation", *Soc. Italiana di Fisica* **1**, 43–65 (1986).

[179] P. H. Hünenberger, *Thermostat Algorithms for Molecular Dynamics Simulations*, pages 105–149 (Springer, Berlin, Heidelberg, 2005).

[180] G. Bussi, D. Donadio, and M. Parrinello, "Canonical sampling through velocity rescaling", *J. Chem. Phys.* **126**, 014101 (2007).

[181] H. C. Andersen, "Rattle: A 'velocity' version of the shake algorithm for molecular dynamics calculations", *J. Chem. Phys.* **52**, 24–34 (1983).

[182] S. Nosé, "A molecular dynamics method for simulations in the canonical ensemble", *Mol. Phys.* **52**, 255–268 (1984).

[183] W. G. Hoover, "Canonical dynamics: Equilibrium phase-space distributions", *Phys. Rev. A* **31**, 1695–1697 (1985).

[184] H. J. C. Berendsen, J. P. M. Postma, W. F. van Gunsteren, A. DiNola, and J. R. Haak, "Molecular dynamics with coupling to an external bath", *J. Chem. Phys.* **81**, 3684–3690 (1984).

[185] H. A. Posch, W. G. Hoover, and F. J. Vesely, "Canonical dynamics of the Nosé oscillator: Stability, order, and chaos", *Phys. Rev. A* **33**, 4253–4265 (1986).

[186] P. Siders and R. A. Marcus, "Quantum effects for electron-transfer reactions in the 'inverted region'", *J. Am. Chem. Soc.* **103**, 748–752 (1981).

[187] S. H. Lin, C. H. Chang, K. K. Liang, R. Chang, Y. J. Shiu, J. M. Zhang, T.-S. Yang, M. Hayashi, and F. C. Hsu, *Ultrafast Dynamics and Spectroscopy of Bacterial Photosynthetic Reaction Centers*, chapter 1-2, pages 1–88 (John Wiley & Sons, Ltd, 2002).

[188] "Electron Transfer Reactions in Chemistry: Theory and Experiment", R. A. Marcus, in *Nobel Lectures: Chemistry 1991–1995*, edited by B. G. Malmström (World Scientific, Singapore, 1997).

[189] R. A. Marcus, "Electron transfer reactions in chemistry. Theory and experiment", *Rev. Mod. Phys.* **65**, 599 (1993).

[190] C. Greco, A. Melnyk, K. Kremer, D. Andrienko, and K. Ch. Daoulas, "Generic Model for Lamellar Self-Assembly in Conjugated Polymers: Linking Mesoscopic Morphology and Charge Transport in P3HT", *Macromol.* **52**, 968–981 (2019).

[191] S. F. Nelsen, S. C. Blackstock, and Y. Kim, "Estimation of inner shell Marcus terms for amino nitrogen compounds by molecular orbital calculations", *J. Am. Chem. Soc.* **109**, 677–682 (1987).

[192] A. Miller and E. Abrahams, "Impurity Conduction at Low Concentrations", *Phys. Rev.* **120**, 745–755 (1960).

[193] B. Movaghar, M. Grünewald, B. Ries, H. Bässler, and D. Würtz, "Diffusion and relaxation of energy in disordered organic and inorganic materials", *Phys. Rev. B* **33**, 5545–5554 (1986).

[194] P. M. Borsenberger, L. Pautmeier, and H. Bässler, "Charge transport in disordered molecular solids", *J. Chem. Phys.* **94**, 5447 (1991).

[195] W. F. Pasveer, J. Cottaar, C. Tanase, R. Coehoorn, P. A. Bobbert, P. W. M. Blom, D. M. de Leeuw, and M. A. J. Michels, "Unified Description of Charge-Carrier Mobilities in Disordered Semiconducting Polymers", *Phys. Rev. Lett.* **94**, 206601 (2005).

[196] N. Vukmirović and L.-W. Wang, "Charge Carrier Motion in Disordered Conjugated Polymers: A Multiscale Ab Initio Study", *Nano Lett.* **9**, 3996–4000 (2009).

[197] M. Panzer and C. Frisbie, "High Carrier Density and Metallic Conductivity in Poly(3-hexylthiophene) Achieved by Electrostatic Charge Injection", *Adv. Func. Mater.* **16**, 1051–1056 (2006).

[198] A. S. Dhoot, G. M. Wang, D. Moses, and A. J. Heeger, "Voltage-Induced Metal-Insulator Transition in Polythiophene Field-Effect Transistors", *Phys. Rev. Lett.* **96**, 246403 (2006).

[199] J. Yuen, R. Menon, N. Coates, E. Namdas, S. Cho, S. Hannahs, D. Moses, and A. Heeger, "Nonlinear transport in semiconducting polymers at high carrier densities", *Nat. Mater.* **8**, 572–575 (2009).

[200] A. Kronemeijer, E. Huisman, I. Katsouras, P. Van Hal, T. Geuns, P. Blom, S. Van Der Molen, and D. De Leeuw, "Universal scaling in highly doped conducting polymer films", *Phys. Rev. Lett.* **105**, 156604 (2010).

[201] K. Asadi, A. J. Kronemeijer, T. Cramer, J. A. L. Koster, P. W. M. Blom, and D. M. de Leeuw, "Polaron hopping mediated by nuclear tunnelling in semiconducting polymers at high carrier density", *Nat. Commun.* (2013).

[202] H. Grabert and U. Weiss, "Quantum Tunneling Rates for Asymmetric Double-Well Systems with Ohmic Dissipation", *Phys. Rev. Lett.* **54**, 1605–1608 (1985).

[203] U. Weiss, *Quantum Dissipative Systems*, volume 13 (World Scientific, 2nd edition, 2008).

[204] M. P. A. Fisher and A. T. Dorsey, "Dissipative Quantum Tunneling in a Biased Double-Well System at Finite Temperatures", *Phys. Rev. Lett.* **54**, 1609–1612 (1985).

[205] R. Egger, C. H. Mak, and U. Weiss, "Quantum rates for nonadiabatic electron transfer", *J. Chem. Phys.* **100**, 2651–2660 (1994).

[206] A. Leggett, S. Chakravarty, A. Dorsey, M. Fisher, A. Garg, and W. Zwerger, "Dynamics of the dissipative two-state system", *Rev. Mod. Phys.* **59**, 1–85 (1987).

[207] J. Jortner, M. Bixon, T. Langenbacher, and M. E. Michel-Beyerle, "Charge transfer and transport in DNA", *Proc. Natl. Acad. Sci.* **95**, 12759–12765 (1998).

[208] H. Oberhofer, K. Reuter, and J. Blumberger, "Charge transport in molecular materials: An assessment of computational methods", *Chem. Rev.* **117**, 10319–10357 (2017).

[209] R. S. Mulliken, "Molecular compounds and their spectra. II", *J. Am. Chem. Soc.* **74**, 811–824 (1952).

[210] N. S. Hush, "Intervalence-transfer absorption. Part 2. Theoretical considerations and spectroscopic data", *Prog. Inorg. Chem* **8**, 12 (1967).

[211] E. F. Valeev, V. Coropceanu, D. A. da Silva Filho, S. Salman, and J.-L. Brédas, "Effect of Electronic Polarization on Charge-Transport Parameters in Molecular Organic Semiconductors", *J. Am. Chem. Soc.* **128**, 9882–9886 (2006).

[212] N. Gillet, L. Berstis, X. Wu, F. Gajdos, A. Heck, A. de la Lande, J. Blumberger, and M. Elstner, "Electronic Coupling Calculations for Bridge-Mediated Charge Transfer Using Constrained Density Functional Theory (CDFT) and Effective Hamiltonian Approaches at the Density Functional Theory (DFT) and Fragment-Orbital Density Functional Tight Binding (FODFTB) Level", *J. Chem. Theory Comput.* **12**, 4793–4805 (2016).

[213] V. Rühle, C. Junghans, A. Lukyanov, K. Kremer, and D. Andrienko, "Versatile Object-Oriented Toolkit for Coarse-Graining Applications", *J. Chem. Theory Comput.* **5**, 3211–3223 (2009).

[214] J. Kirkpatrick, "An approximate method for calculating transfer integrals based on the ZINDO Hamiltonian", *Int. J. Quantum Chem.* **108**, 51–56 (2008).

[215] I. Kondov, M. Čížek, C. Benesch, H. Wang, and M. Thoss, "Quantum Dynamics of Photoinduced Electron-Transfer Reactions in Dye-Semiconductor Systems: First-Principles Description and Application to Coumarin 343 TiO2", *J. Phys. Chem. C* **111**, 11970–11981 (2007).

[216] Z. Futera and J. Blumberger, "Electronic Couplings for Charge Transfer across Molecule/Metal and Molecule/Semiconductor Interfaces: Performance of the Projector Operator-Based Diabatization Approach", *J. Phys. Chem. C* **121**, 19677–19689 (2017).

[217] X. Jiang, Z. Futera, M. E. Ali, F. Gajdos, G. F. von Rudorff, A. Carof, M. Breuer, and J. Blumberger, "Cysteine Linkages Accelerate Electron Flow through Tetra-Heme Protein STC", *J. Am. Chem. Soc.* **139**, 17237–17240 (2017).

[218] B. Baumeier, J. Kirkpatrick, and D. Andrienko, "Density-functional based determination of intermolecular charge transfer properties for large-scale morphologies", *Phys. Chem. Chem. Phys.* **12**, 11103–11113 (2010).

[219] P. Löwdin, "On the Non-Orthogonality Problem Connected with the Use of Atomic Wave Functions in the Theory of Molecules and Crystals", *J. Chem. Phys.* **18**, 365–375 (1950).

[220] K. Nishimoto and N. Mataga, "Electronic Structure and Spectra of Some Nitrogen Heterocycles", *Z. Phys. Chem.* **12**, 335–338 (1957).

[221] R. S. Mulliken, C. A. Rieke, D. Orloff, and H. Orloff, "Formulas and Numerical Tables for Overlap Integrals", *J. Chem. Phys.* **17**, 1248–1267 (1949).

[222] H. Oberhofer and J. Blumberger, "Revisiting electronic couplings and incoherent hopping models for electron transport in crystalline C_{60} at ambient temperatures", *Phys. Chem. Chem. Phys.* **14**, 13846–13852 (2012).

[223] P. Ren and J. W. Ponder, "Polarizable Atomic Multipole Water Model for Molecular Mechanics Simulation", *J. Phys. Chem. B* **107**, 5933–5947 (2003).

[224] M. Schwarze, K. S. Schellhammer, K. Ortstein, J. Benduhn, C. Gaul, A. Hinderhofer, L. Perdigón Toro, R. Scholz, J. Kublitski, S. Roland, M. Lau, C. Poelking, D. Andrienko, G. Cuniberti, F. Schreiber, D. Neher, K. Vandewal, F. Ortmann, and K. Leo, "Impact of molecular quadrupole moments on the energy levels at organic heterojunctions", *Nat. Commun.* **10**, 2466 (2019).

[225] H. D. Herce, A. E. Garcia, and T. Darden, "The electrostatic surface term: (I) Periodic systems", *J. Chem. Phys.* **126**, 124106 (2007).

[226] T. Darden, D. York, and L. Pedersen, "Particle mesh Ewald: An N log(N) method for Ewald sums in large systems", *J. Chem. Phys.* **98**, 10089–10092 (1993).

[227] C. Poelking and D. Andrienko, "Long-Range Embedding of Molecular Ions and Excitations in a Polarizable Molecular Environment", *J. Chem. Theory Comput.* **12**, 4516–4523 (2016).

[228] S. I. Pekar, "Local quantum states of electrons in an ideal ion crystal", *Zh. Eksp. Tear. Fiz.* **16**, 341–348 (1946).

[229] S. I. Pekar, *Untersuchungen über die Elektronentheorie der Kristalle.* (Akademieverlag, 1954).

[230] D. Dunlap, P. Parris, and V. Kenkre, "Charge-Dipole Model for the Universal Field Dependence of Mobilities in Molecularly Doped Polymers", *Phys. Rev. Lett.* **77**, 542–545 (1996).

[231] B. Thole, "Molecular polarizabilities calculated with a modified dipole interaction", *Chem. Phys.* **59**, 341–350 (1981).

[232] C. Hättig and B. A. Heß, "Calculation of orientation-dependent double-tensor moments for Coulomb-type intermolecular interactions", *Mol. Phys.* **81**, 813–824 (1994).

[233] C. M. Breneman and K. B. Wiberg, "Determining atom-centered monopoles from molecular electrostatic potentials. The need for high sampling density in formamide conformational analysis", *J. Comput. Chem.* **11**, 361–373 (1990).

[234] A. J. Stone, *The Theory of intermolecular forces* (Clarendon Press, Oxford, 1997).

[235] A. Stone and M. Alderton, "Distributed multipole analysis", *Mol. Phys.* **56**, 1047–1064 (1985).

[236] A. J. Stone, "Distributed Multipole Analysis: Stability for Large Basis Sets", *J. Chem. Theory Comput.* **1**, 1128–1132 (2005).

[237] P. T. van Duijnen and M. Swart, "Molecular and Atomic Polarizabilities: Thole's Model Revised", *J. Phys. Chem. A* **102**, 2399–2407 (1998).

[238] J. Applequist, J. R. Carl, and K.-K. Fung, "Atom dipole interaction model for molecular polarizability. Application to polyatomic molecules and determination of atom polarizabilities", *J. Am. Chem. Soc.* **94**, 2952–2960 (1972).

[239] V. Rühle, A. Lukyanov, F. May, M. Schrader, T. Vehoff, J. Kirkpatrick, B. Baumeier, and D. Andrienko, VOTCA-CTP, Charge Transport Simulations, User Manual Version 1.5, 2019.

[240] J. Cottaar and P. A. Bobbert, "Calculating charge-carrier mobilities in disordered semiconducting polymers: Mean field and beyond", *Phys. Rev. B* **74**, 115204 (2006).

[241] A. A. Markov, "An Example of Statistical Investigation of the Text Eugene Onegin Concerning the Connection of Samples in Chains", *Sci Context* **19**, 591–600 (2006).

[242] P. Gagniuc, *Markov Chains: From Theory to Implementation and Experimentation* (Wiley, 2017).

[243] A. B. Bortz, M. H. Kalos, and J. L. Lebowitz, "A new algorithm for Monte Carlo simulation of Ising spin systems", *J. Comp. Phys.* **17**, 10–18 (1975).

[244] "Charge-transport simulations in organic semiconductors", F. May, PhD dissertation, Johannes Gutenberg-Universität Mainz, 2012.

[245] K. Dill and S. Bromberg, *Molecular driving forces: statistical thermodynamics in biology, chemistry, physics, and nanoscience* (Garland Science, 2010).

[246] V. Kažukauskas, M. Pranaitis, V. Čyras, L. Sicot, and F. Kajzar, "Negative mobility dependence on electric field in poly(3-alkylthiophenes) evidenced by the charge extraction by linearly increasing voltage method", *Thin Solid Films* **516**, 8988 – 8992 (2008).

[247] A. Devizis, K. Meerholz, D. Hertel, and V. Gulbinas, "Ultrafast charge carrier mobility dynamics in poly(spirobifluorene-*co*-benzothiadiazole): Influence of temperature on initial transport", *Phys. Rev. B* **82**, 155204 (2010).

[248] A. Lukyanov and D. Andrienko, "Extracting nondispersive charge carrier mobilities of organic semiconductors from simulations of small systems", *Phys. Rev. B* **82**, 193202 (2010).

[249] K. Seki and M. Tachiya, "Electric field dependence of charge mobility in energetically disordered materials: Polaron aspects", *Phys. Rev. B* **65**, 014305 (2001).

[250] J. J. M. van der Holst, M. A. Uijttewaal, B. Ramachandhran, R. Coehoorn, P. A. Bobbert, G. A. de Wijs, and R. A. de Groot, "Modeling and analysis of the three-dimensional current density in sandwich-type single-carrier devices of disordered organic semiconductors", *Phys. Rev. B* **79**, 085203 (2009).

[251] M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, G. Scalmani, V. Barone, B. Mennucci, G. A. Petersson, H. Nakatsuji, M. Caricato, X. Li, H. P. Hratchian, and D. J. Fox et. al., Gaussian 09 Revision D.01, 2009, Gaussian Inc. Wallingford CT.

[252] CPMD Version 3.15.1, 2014, IBM Research Division MPI Festkörperforschung, Stuttgart.

[253] B. Aradi, B. Hourahine, and T. Frauenheim, "DFTB+, a Sparse Matrix-Based Implementation of the DFTB Method", *J. Phys. Chem. A* **111**, 5678–5684 (2007).

[254] J. J. P. Stewart, "Optimization of parameters for semiempirical methods II. Applications", *J. Comp. Chem.* **10**, 221–264 (1989).

[255] J. J. P. Stewart, "Optimization of parameters for semiempirical methods. III Extension of PM3 to Be, Mg, Zn, Ga, Ge, As, Se, Cd, In, Sn, Sb, Te, Hg, Tl, Pb, and Bi", *J. Comp. Chem.* **12**, 320–341 (1991).

[256] M. Elstner, D. Porezag, G. Jungnickel, J. Elsner, M. Haugk, T. Frauenheim, S. Suhai, and G. Seifert, "Self-consistent-charge density-functional tight-binding method for simulations of complex materials properties", *Phys. Rev. B* **58**, 7260–7268 (1998).

[257] T. Niehaus, M. Elstner, T. Frauenheim, and S. Suhai, "Application of an approximate density-functional method to sulfur containing compounds", *J. Mol. Struct. THEOCHEM* **541**, 185–194 (2001).

[258] M. Gaus, A. Goez, and M. Elstner, "Parametrization and Benchmark of DFTB3 for Organic Molecules", *J. Chem. Theory Comput.* **9**, 338–354 (2013).

[259] M. Gaus, X. Lu, M. Elstner, and Q. Cui, "Parameterization of DFTB3/3OB for Sulfur and Phosphorus for Chemical and Biological Applications", *J. Chem. Theory Comput.* **10**, 1518–1537 (2014).

[260] M. Kubillus, T. Kubař, M. Gaus, J. Řezáč, and M. Elstner, "Parameterization of the DFTB3 Method for Br, Ca, Cl, F, I, K, and Na in Organic and Biological Systems", *J. Chem. Theory Comput.* **11**, 332–342 (2015).

[261] S. Dapprich, I. Komáromi, K. Byun, K. Morokuma, and M. J. Frisch, "A new ONIOM implementation in Gaussian98. Part I. The calculation of energies, gradients, vibrational frequencies and electric field derivatives.", *J. Mol. Struct. THEOCHEM* **461-462**, 1 – 21 (1999).

[262] T. Vreven, K. S. Byun, I. Komáromi, S. Dapprich, J. A. Montgomery, K. Morokuma, and M. J. Frisch, "Combining Quantum Mechanics Methods with Molecular Mechanics Methods in ONIOM", *J. Chem. Theory Comput.* **2**, 815–826 (2006).

[263] M. Knipper, J. Parisi, K. Coakley, C. Waldauf, C. J. Brabec, and V. Dyakonov, "Impedance Spectroscopy on Polymer-Fullerene Solar Cells", *Z. Naturforsch.* **62a**, 490–494 (2007).

[264] W. Lee, S. Chuang, H. Chen, W. Su, and C. Lin, "Exploiting optical properties of P3HT:PCBM films for organic solar cells with semitransparent anode", *Thin Solid Films* **518**, 7450–7454 (2010).

[265] A. Bondi, "van der Waals Volumes and Radii", *J. Chem. Phys.* **68**, 441–451 (1964).

[266] J. W. Ponder, C. Wu, P. Ren, V. S. Pande, J. D. Chodera, M. J. Schnieders, I. Haque, D. L. Mobley, D. S. Lambrecht, R. A. DiStasio, M. Head-Gordon, G. N. I. Clark, M. E. Johnson, and T. Head-Gordon, "Current Status of the AMOEBA Polarizable Force Field", *J. Phys. Chem. B* **114**, 2549–2564 (2010).

[267] T. Koch, Charge Transport Package, `https://github.com/tobiaskoch1988/charge_transport_package`, Accessed: 2020-04-01.

[268] M. J. Abraham, B. Hess, D. v. d. Spoel, and E. Lindahl, GROMACS User Manual version 5.0.4, 2020.

[269] D. Van Der Spoel, E. Lindahl, B. Hess, G. Groenhof, A. E. Mark, and H. J. C. Berendsen, "GROMACS: Fast, flexible, and free", *J. Comput. Chem.* **26**, 1701–1718 (2005).

[270] O. Tange, "GNU Parallel - The Command-Line Power Tool", *;login: The USENIX Magazine* **36**, 42–47 (2011).

[271] M. Parrinello and A. Rahman, "Polymorphic transitions in single crystals: A new molecular dynamics method", *J. Appl. Phys.* **52**, 7182–7190 (1981).

[272] J. Bachmann, Molecular Plane Analyser, `https://github.com/jimbach/molecular_plane_analyzer`, Accessed: 2020-03-01.

[273] J. R. Reynolds and T. A. Skotheim, *Handbook of Conducting Polymers. Conjugated Polymers: Theory, Synthesis, Properties, and Characterization*, volume 2 (CRC Press Taylor & Francis Group, Boca Raton, 3nd edition, 2007).

[274] T. Helgaker and P. Jorgensen, "An electronic Hamiltonian for origin independent calculations of magnetic properties", *J. Chem. Phys.* **95**, 2595–2601 (1991).

[275] J. Tomasi, B. Mennucci, and R. Cammi, "Quantum Mechanical Continuum Solvation Models", *Chem. Rev.* **105**, 2999–3094 (2005).

[276] W. M. Skid, "UFF, a Full Periodic Table Force Field for Molecular Mechanics and Molecular Dynamics Simulations", *J. Am. Chem. Soc.* **114**, 10024–10035 (1992).

[277] C. G. Broyden, "The convergence of a class of double-rank minimization algorithms 1. general considerations", *IMA J. Appl. Math.* **6**, 76–90 (1970).

[278] R. Fletcher, "A new approach to variable metric algorithms", *Comput. J.* **13**, 317–322 (1970).

[279] S. Goedecker, M. Teter, and J. Hutter, "Separable dual-space Gaussian pseudopotentials", *Phys. Rev. B* **54**, 1703–1710 (1996).

[280] C. Hartwigsen, S. Goedecker, and J. Hutter, "Relativistic separable dual-space Gaussian pseudopotentials from H to Rn", *Phys. Rev. B* **58**, 3641–3662 (1998).

[281] M. Krack, "Pseudopotentials for H to Kr optimized for gradient-corrected exchange-correlation functionals", *Theor. Chem. Acc.* **114**, 145–152 (2005).

[282] D. Alberga, A. Perrier, I. Ciofini, G. F. Mangiatordi, G. Lattanzi, and C. Adamo, "Morphological and charge transport properties of amorphous and crystalline P3HT and PBTTT: insights from theory", *Phys. Chem. Chem. Phys.* **17**, 18742–18750 (2015).

[283] M. Abadi, A. Agarwal, P. Barham, E. Brevdo, Z. Chen, C. Citro, G. S. Corrado, A. Davis, J. Dean, M. Devin, S. Ghemawat, I. Goodfellow, A. Harp, G. Irving, M. Isard, Y. Jia, R. Jozefowicz, L. Kaiser, M. Kudlur, J. Levenberg, D. Mané, R. Monga, S. Moore, D. Murray, C. Olah, M. Schuster, J. Shlens, B. Steiner, I. Sutskever, K. Talwar, P. Tucker, V. Vanhoucke, V. Vasudevan, F. Viégas, O. Vinyals, P. Warden, M. Wattenberg, M. Wicke, Y. Yu, and X. Zheng, TensorFlow: Large-Scale Machine Learning on Heterogeneous Systems, 2015.

[284] M. Hultell and S. Stafström, "Impact of ring torsion dynamics on intrachain charge transport in conjugated polymers", *Phys. Rev. B* **79**, 014302 (2009).

[285] D. Raithel, L. Simine, S. Pickel, K. Schötz, F. Panzer, S. Baderschneider, D. Schiefer, R. Lohwasser, J. Köhler, M. Thelakkat, M. Sommer, A. Köhler, P. J. Rossky, and R. Hildner, "Direct observation of backbone planarization via side-chain alignment in single bulky-substituted polythiophenes", *Proc. Natl. Acad. Sci.* **115**, 2699–2704 (2018).

[286] V. Lemaur, J. Cornil, R. Lazzaroni, H. Sirringhaus, D. Beljonne, and Y. Olivier, "Resilience to Conformational Fluctuations Controls Energetic Disorder in Conjugated Polymer Materials: Insights from Atomistic Simulations", *Chem. Mater.* **31**, 6889–6899 (2019).

[287] A. A. Kocherzhenko, S. Patwardhan, F. C. Grozema, H. L. Anderson, and L. D. A. Siebbeles, "Mechanism of Charge Transport along Zinc Porphyrin-Based Molecular Wires", *J. Am. Chem. Soc.* **131**, 5522–5529 (2009).

[288] K. F. Freed and J. Jortner, "Multiphonon Processes in the Nonradiative Decay of Large Molecules", *J. Chem. Phys.* **52**, 6272–6291 (1970).

[289] P. F. Barbara, T. J. Meyer, and M. A. Ratner, "Contemporary Issues in Electron Transfer Research", *J. Phys. Chem.* **100**, 13148–13168 (1996).

[290] V. Rühle, J. Kirkpatrick, and D. Andrienko, "A multiscale description of charge transport in conjugated oligomers", *J. Chem. Phys.* **132**, 134103 (2010).

[291] J. Sjöqvist, J. Maria, R. A. Simon, M. Linares, P. Norman, K. P. R. Nilsson, and M. Lindgren, "Toward a Molecular Understanding of the Detection of Amyloid Proteins with Flexible Conjugated Oligothiophenes", *J. Phys. Chem. A* **118**, 9820–9827 (2014).

[292] "Morphology and charge transport in P3HT: A theorist's perspective", C. Poelking, K. Daoulas, A. Troisi, and D. Andrienko, in *P3HT Revisited–From Molecular Scale to Solar Cell Devices*, pages 139–180 (Springer, 2014).

[293] N. Hush, "Distance Dependence of Electron Transfer Rates", *Coord. Chem. Rev.* **64**, 135–157 (1985).

[294] R. Marcus and N. Sutin, "Electron transfers in chemistry and biology", *Biochim. Biophys. Acta* **811**, 265–322 (1985).

[295] G. L. Closs and J. R. Miller, "Intramolecular Long-Distance Electron Transfer in Organic Molecules", *Science* **240**, 440–447 (1988).

[296] K. J. Lee, Y. Xiao, E. S. Kim, F. Mathevet, L. Mager, O. Cregut, F. Fages, J.-C. Ribierre, J. W. Wu, and A. D'Aléo, "Donor-Acceptor Distance-Dependent Charge Transfer Dynamics Controlled by Metamaterial Structures", *ACS Photonics* **6**, 2649–2654 (2019).

[297] S. Neumann and O. S. Wenger, "Fundamentally Different Distance Dependences of Electron-Transfer Rates for Low and High Driving Forces", *Inorg. Chem.* **58**, 855–860 (2019).

[298] C. Creutz and N. Sutin, "Vestiges of the inverted region for highly exergonic electron-transfer reactions", *J. Am. Chem. Soc.* **99**, 241–243 (1977).

[299] Y. Choulis, S. A.and Kim, J. Nelson, D. D. C. Bradley, M. Giles, M. Shkunov, and I. McCulloch, "High ambipolar and balanced carrier mobility in regioregular poly(3-hexylthiophene)", *Appl. Phys. Lett.* **85**, 3890–3892 (2004).

[300] A. J. Mozer and N. S. Sariciftci, "Negative electric field dependence of charge carrier drift mobility in conjugated, semiconducting polymers", *Chem. Phys. Lett.* **389**, 438–442 (2004).

[301] C. Poelking and D. Andrienko, "Effect of Polymorphism, Regioregularity and Paracrystallinity on Charge Transport in Poly(3-hexylthiophene) [P3HT] Nanofibers", *Macromol.* **46**, 8941–8956 (2013).

[302] "Morphology and Charge Transport in P3HT: A Theorist's Perspective", C. Poelking, K. Daoulas, A. Troisi, and D. Andrienko, in *P3HT Revisited – From Molecular Scale to Solar Cell Devices*, edited by S. Ludwigs, pages 139–180 (Springer Berlin Heidelberg, Berlin, Heidelberg, 2014).

[303] F. Laquai, D. Andrienko, R. Mauer, and P. W. M. Blom, "Charge Carrier Transport and Photogeneration in P3HT:PCBM Photovoltaic Blends", *Macro. Rap. Comm.* **36**, 1001–1025 (2015).

[304] M. L. Jones, D. M. Huang, B. Chakrabarti, and C. Groves, "Relating Molecular Morphology to Charge Mobility in Semicrystalline Conjugated Polymers", *J. Phys. Chem. C* **120**, 4240–4250 (2016).

[305] M. Polkehn, H. Tamura, and I. Burghardt, "Impact of charge-transfer excitons in regioregular polythiophene on the charge separation at polythiophene-fullerene heterojunctions", *J. Phys. B: At. Mol. Opt. Phys* **51**, 014003 (2018).

[306] J. C. Hummelen, B. W. Knight, F. LePeq, F. Wudl, J. Yao, and C. L. Wilkins, "Preparation and Characterization of Fulleroid and Methanofullerene Derivatives", *J. Org. Chem.* **60**, 532–538 (1995).

[307] M. M. Wienk, J. M. Kroon, W. J. H. Verhees, J. Knol, J. C. Hummelen, P. A. van Hal, and R. A. J. Janssen, "Efficient Methano[70]fullerene/MDMO-PPV Bulk Heterojunction Photovoltaic Cells", *Angew. Chem. Int. Ed.* **42**, 3371–3375 (2003).

[308] T. Weil, T. Vosch, J. Hofkens, K. Peneva, and K. Müllen, "The Rylene Colorant Family–Tailored Nanoemitters for Photonics Research and Applications", *Angew. Chem. Int. Ed.* **49**, 9068–9093 (2010).

[309] Y. Li, C. Wang, C. Li, S. Di Motta, F. Negri, and Z. Wang, "Synthesis and Properties of Ethylene-Annulated Di(perylene diimides)", *Org. Lett.* **14**, 5278–5281 (2012).

[310] Y. Lin, Y. Wang, J. Wang, J. Hou, Y. Li, D. Zhu, and X. Zhan, "A Star-Shaped Perylene Diimide Electron Acceptor for High-Performance Organic Solar Cells", *Adv. Mater.* **26**, 5137–5142 (2014).

[311] L. Ye, K. Sun, W. Jiang, S. Zhang, W. Zhao, H. Yao, Z. Wang, and J. Hou, "Enhanced Efficiency in Fullerene-Free Polymer Solar Cell by Incorporating Fine-designed Donor and Acceptor Materials", *ACS Appl. Mater. & Interfaces* **7**, 9274–9280 (2015).

[312] D. Meng, D. Sun, C. Zhong, T. Liu, B. Fan, L. Huo, Y. Li, W. Jiang, H. Choi, T. Kim, J. Y. Kim, Y. Sun, Z. Wang, and A. J. Heeger, "High-Performance Solution-Processed Non-Fullerene Organic Solar Cells Based on Selenophene-Containing Perylene Bisimide Acceptor", *J. Am. Chem. Soc.* **138**, 375–380 (2016).

[313] T. A. Shastry, P. E. Hartnett, M. R. Wasielewski, T. J. Marks, and M. C. Hersam, "Ternary Polymer-Perylenediimide-Carbon Nanotube Photovoltaics with High Efficiency and Stability under Super-Solar Irradiation", *ACS Energy Lett.* **1**, 548–555 (2016).

[314] C. Zhan and J. Yao, "More than Conformational Twisting or Coplanarity: Molecular Strategies for Designing High-Efficiency Nonfullerene Organic Solar Cells", *Chem. Mater.* **28**, 1948–1964 (2016).

[315] H. Lin, S. Chen, H. Hu, L. Zhang, T. Ma, J. Y. L. Lai, Z. Li, A. Qin, X. Huang, B. Tang, and H. Yan, "Reduced Intramolecular Twisting Improves the Performance of 3D Molecular Acceptors in Non-Fullerene Organic Solar Cells", *Adv. Mater.* **28**, 8546–8551 (2016).

[316] P. E. Hartnett, H. S. S. R. Matte, N. D. Eastham, N. E. Jackson, Y. Wu, L. X. Chen, M. A. Ratner, R. P. H. Chang, M. C. Hersam, M. R. Wasielewski, and T. J. Marks, "Ring-fusion as a perylenediimide dimer design concept for high-performance non-fullerene organic photovoltaic acceptors", *Chem. Sci.* **7**, 3543–3555 (2016).

[317] H. Zhong, C.-H. Wu, C.-Z. Li, J. Carpenter, C.-C. Chueh, J.-Y. Chen, H. Ade, and A. Jen, "Rigidifying Nonplanar Perylene Diimides by Ring Fusion Toward Geometry-Tunable Acceptors for High-Performance Fullerene-Free Solar Cells", *Adv. Mater.* **28**, 951–958 (2016).

[318] L. Yang, W. Gu, L. Lv, Y. Chen, Y. Yang, P. Ye, J. Wu, L. Hong, A. Peng, and H. Huang, "Triplet Tellurophene-Based Acceptors for Organic Solar Cells", *Angew. Chem. Inter. Ed.* **57**, 1096–1102 (2018).

[319] N. Liang, X. Zhu, Z. Zheng, D. Meng, G. Liu, J. Zhang, S. Li, Y. Li, J. Hou, B. Hu, and Z. Wang, "Tuning Charge Generation Process of Rylene Imide-Based Solar Cells via Chalcogen-Atom-Annulation", *Chem. Mater.* **31**, 3636–3643 (2019).

[320] D. Meng, H. Fu, C. Xiao, X. Meng, T. Winands, W. Ma, W. Wei, B. Fan, L. Huo, N. L. Doltsinis, Y. Li, Y. Sun, and Z. Wang, "Three-Bladed Rylene Propellers with Three-Dimensional Network Assembly for Organic Electronics", *J. Am. Chem. Soc.* **138**, 10184–10190 (2016).

[321] G. Liu, T. Koch, Y. Li, N. L. Doltsinis, and Z. Wang, "Nanographene Imides Featuring Dual-Core Sixfold [5]Helicenes", *Angew. Chem. Int. Ed.* **58**, 178–183 (2019).

[322] H. Hoppe and N. S. Sariciftci, "Morphology of polymer/fullerene bulk heterojunction solar cells", *J. Mater. Chem.* **16**, 45–61 (2006).

[323] M. Scharber, D. Mühlbacher, M. Koppe, P. Denk, C. Waldauf, A. Heeger, and C. Brabec, "Design Rules for Donors in Bulk-Heterojunction Solar Cells-Towards 10% Energy-Conversion Efficiency", *Adv. Mater.* **18**, 789–794 (2006).

[324] V. Coropceanu, J. Cornil, D. A. da Silva Filho, Y. Olivier, R. Silbey, and J.-L. Brédas, "Charge Transport in Organic Semiconductors", *Chem. Rev.* **107**, 926–952 (2007).

[325] K. Maturová, S. S. van Bavel, M. M. Wienk, R. A. J. Janssen, and M. Kemerink, "Morphological Device Model for Organic Bulk Heterojunction Solar Cells", *Nano Lett.* **9**, 3032–3037 (2009).

[326] J.-L. Brédas, J. E. Norton, J. Cornil, and V. Coropceanu, "Molecular Understanding of Organic Solar Cells: The Challenges", *Acc. Chem. Res.* **42**, 1691–1699 (2009).

[327] R. Noriega, J. Rivnay, K. Vandewal, F. Koch, N. Stingelin, P. Smith, M. Toney, and A. Salleo, "A general relationship between disorder, aggregation and charge transport in conjugated polymers", *Nat. Mater.* **12**, 1038–1044 (2013).

[328] Y. Huang, E. J. Kramer, A. J. Heeger, and G. C. Bazan, "Bulk Heterojunction Solar Cells: Morphology and Performance Relationships", *Chem. Rev.* **114**, 7006–7043 (2014).

[329] J. Song, M. Zhang, M. Yuan, Y. Qian, Y. Sun, and F. Liu, "Morphology Characterization of Bulk Heterojunction Solar Cells", *Small Methods* **2**, 1700229 (2018).

[330] L. Meng, Y. Zhang, X. Wan, C. Li, X. Zhang, Y. Wang, X. Ke, Z. Xiao, L. Ding, R. Xia, H.-L. Yip, Y. Cao, and Y. Chen, "Organic and solution-processed tandem solar cells with 17.3% efficiency", *Science* **361**, 1094–1098 (2018).

[331] R. J. Kline, M. D. McGehee, E. N. Kadnikova, J. Liu, J. M. J. Fréchet, and M. F. Toney, "Dependence of Regioregular Poly(3-hexylthiophene) Film Morphology and Field-Effect Mobility on Molecular Weight", *Macromol.* **38**, 3312–3319 (2005).

[332] P. Schilinsky, U. Asawapirom, U. Scherf, M. Biele, and C. J. Brabec, "Influence of the Molecular Weight of Poly(3-hexylthiophene) on the Performance of Bulk Heterojunction Solar Cells", *Chem. Mater.* **17**, 2175–2180 (2005).

[333] H. A. Bronstein and C. K. Luscombe, "Externally Initiated Regioregular P3HT with Controlled Molecular Weight and Narrow Polydispersity", *J. Am. Chem. Soc.* **131**, 12894–12895 (2009).

[334] C. Scharsich, R. H. Lohwasser, M. Sommer, U. Asawapirom, U. Scherf, M. Thelakkat, D. Neher, and A. Köhler, "Control of aggregate formation in poly(3-hexylthiophene) by solvent, molecular weight, and synthetic method", *J. Polym. Sci. Pol. Phys.* **50**, 442–453 (2011).

[335] F. P. V. Koch, J. Rivnay, S. Foster, C. Müller, J. M. Downing, E. Buchaca-Domingo, P. Westacott, L. Yu, M. Yuan, M. Baklar, Z. Fei, C. Luscombe, M. A. McLachlan, M. Heeney, G. Rumbles, C. Silva, A. Salleo, J. Nelson, P. Smith, and N. Stingelin, "The impact of molecular weight on microstructure and charge transport in semicrystalline polymer semiconductors poly(3-hexylthiophene), a model study", *Prog. Polym. Sci.* **38**, 1978 – 1989 (2013).

[336] A. G. Dixon, R. Visvanathan, N. A. Clark, N. Stingelin, N. Kopidakis, and S. E. Shaheen, "Molecular weight dependence of carrier mobility and recombination rate in neat P3HT films", *J. Polym. Sci. Pol. Phys.* **56**, 31–35 (2018).

[337] A. Sharenko, D. Gehrig, F. Laquai, and T.-Q. Nguyen, "The Effect of Solvent Additive on the Charge Generation and Photovoltaic Performance of a Solution-Processed Small Molecule:Perylene Diimide Bulk Heterojunction Solar Cell", *Chem. Mater.* **26**, 4109–4118 (2014).

[338] F. C. Krebs, "Fabrication and processing of polymer solar cells: A review of printing and coating techniques", *Sol. Energy Mater. Sol. Cells* **93**, 394–412 (2009).

[339] X. Zhang, C. Zhan, and J. Yao, "Non-Fullerene Organic Solar Cells with 6.1% Efficiency through Fine-Tuning Parameters of the Film-Forming Process", *Chem. Mater.* **27**, 166–173 (2015).

[340] G. Zhang, R. C. Huber, A. S. Ferreira, S. D. Boyd, C. K. Luscombe, S. H. Tolbert, and B. J. Schwartz, "Crystallinity Effects in Sequentially Processed and Blend-Cast Bulk-Heterojunction Polymer/Fullerene Photovoltaics", *J. Phy. Chem. C* **118**, 18424–18435 (2014).

[341] P. Westacott, J. R. Tumbleston, S. Shoaee, S. Fearn, J. H. Bannock, J. B. Gilchrist, S. Heutz, J. de Mello, M. Heeney, H. Ade, J. Durrant, D. S. McPhail, and N. Stingelin, "On the role of intermixed phases in organic photovoltaic blends", *Energy Environ. Sci.* **6**, 2756–2764 (2013).

[342] A. Gadisa, J. R. Tumbleston, D.-H. Ko, M. Aryal, R. Lopez, and E. T. Samulski, "The role of solvent and morphology on miscibility of methanofullerene and poly(3-hexylthiophene)", *Thin Solid Films* **520**, 5466 – 5471 (2012).

[343] S. A. Hawks, J. C. Aguirre, L. T. Schelhas, R. J. Thompson, R. C. Huber, A. S. Ferreira, G. Zhang, A. A. Herzing, S. H. Tolbert, and B. J. Schwartz, "Comparing Matched Polymer:Fullerene Solar Cells Made by Solution-Sequential Processing and Traditional Blend Casting: Nanoscale Structure and Device Performance", *J. Phys. Chem. C* **118**, 17413–17425 (2014).

[344] M. Reichenberger, S. Baderschneider, D. Kroh, S. Grauf, J. Köhler, R. Hildner, and A. Köhler, "Watching Paint Dry: The Impact of Diiodooctane on the Kinetics of Aggregate Formation in Thin Films of Poly(3-hexylthiophene)", *Macromol.* **49**, 6420–6430 (2016).

[345] L. J. Richter, D. M. DeLongchamp, and A. Amassian, "Morphology Development in Solution-Processed Functional Organic Blend Films: An In Situ Viewpoint", *Chem. Rev.* **117**, 6332–6366 (2017).

[346] C. Liu, K. Huang, W.-T. Park, M. Li, T. Yang, X. Liu, L. Liang, T. Minari, and Y.-Y. Noh, "A unified understanding of charge transport in organic semiconductors: the importance of attenuated delocalization for the carriers", *Mater. Horiz.* **4**, 608–618 (2017).

[347] G. Juška, K. Arlauskas, and M. Viliūnas, "Extraction current transients: New method of study of charge transport in microcrystalline silicon", *Phys. Rev. Lett.* **84**, 4946–4949 (2000).

[348] A. J. Mozer, N. S. Sariciftci, A. Pivrikas, R. Österbacka, G. Juška, L. Brassat, and H. Bässler, "Charge carrier mobility in regioregular poly(3-hexylthiophene) probed by transient conductivity techniques: A comparative study", *Phys. Rev. B* **71**, 035214 (2005).

[349] A. Tsumura, H. Koezuka, and T. Ando, "Macromolecular electronic device: Field-effect transistor with a polythiophene thin film", *Appl. Phys. Lett.* **49**, 1210–1212 (1986).

[350] C. R. Singh, G. Gupta, R. Lohwasser, S. Engmann, J. Balko, M. Thelakkat, T. Thurn-Albrecht, and H. Hoppe, "Correlation of charge transport with structural order in highly ordered melt-crystallized poly(3-hexylthiophene) thin films", *Inc. J. Polym. Sci., Part B: Polym. Phys.* **51**, 943–951 (2013).

[351] Z. Bao, A. Dodabalapur, and A. Lovinger, "Soluble and processable regioregular poly(3-hexylthiophene) for thin film field-effect transistor applications with high mobility", *Appl. Phys. Lett.* **69**, 4108–4110 (1996).

[352] H. Sirringhaus, N. Tessler, and R. H. Friend, "Integrated Optoelectronic Devices Based on Conjugated Polymers", *Science* **280**, 1741–1744 (1998).

[353] H. Sirringhaus, P. Brown, R. Friend, M. Nielsen, K. Bechgaard, B. Langeveld-Voss, A. Spiering, R. Janssen, E. Meijer, P. Herwig, and D. De Leeuw, "Two-dimensional charge transport in self-organized, high-mobility conjugated polymers", *Nature* **401**, 685–688 (1999).

[354] J. Chang, J. Clark, N. Zhao, H. Sirringhaus, D. Breiby, J. Andreasen, M. Nielsen, M. Giles, M. Heeney, and I. McCulloch, "Molecular-weight dependence of interchain polaron delocalization and exciton bandwidth in high-mobility conjugated polymers", *Phys. Rev. B* **74**, 115318 (2006).

[355] R. J. Kline, M. D. McGehee, and M. F. Toney, "Highly oriented crystals at the buried interface in polythiophene thin-film transistors", *Nat. Mater.* **5**, 222–228 (2006).

[356] A. Zen, M. Saphiannikova, D. Neher, J. Grenzer, S. Grigorian, U. Pietsch, U. Asawapirom, S. Janietz, U. Scherf, I. Lieberwirth, and G. Wegner, "Effect of Molecular Weight on the Structure and Crystallinity of Poly(3-hexylthiophene)", *Macromol.* **39**, 2162–2171 (2006).

[357] P. Kohn, S. Huettner, H. Komber, V. Senkovskyy, R. Tkachov, A. Kiriy, R. Friend, U. Steiner, W. Huck, J.-U. Sommer, and M. Sommer, "On the Role of Single Regiodefects and Polydispersity in Regioregular Poly(3-hexylthiophene): Defect Distribution, Synthesis of Defect-Free Chains, and a Simple Model for the Determination of Crystallinity", *J. Am. Chem. Soc.* **134**, 4790–805 (2012).

[358] A. Ballantyne, L. Chen, J. Dane, T. Hammant, F. Braun, M. Heeney, W. Duffy, I. McCulloch, D. Bradley, and J. Nelson, "The Effect of Poly(3-hexylthiophene) Molecular Weight on Charge Transport and the Performance of Polymer:Fullerene Solar Cells", *Adv. Funct. Mater.* **18**, 2373–2380 (2008).

[359] D. Cheung, D. McMahon, and A. Troisi, "Computational Study of the Structure and Charge-Transfer Parameters in Low-Molecular-Mass P3HT", *J. Phys. Chem. B* **113**, 9393–401 (2009).

[360] N. Rolland, J. F. Franco-Gonzalez, R. Volpi, M. Linares, and I. V. Zozoulenko, "Understanding morphology-mobility dependence in PEDOT:Tos", *Phys. Rev. Mater.* **2**, 045605 (2018).

[361] M. Brinkmann and P. Rannou, "Molecular Weight Dependence of Chain Packing and Semicrystalline Structure in Oriented Films of Regioregular Poly(3-hexylthiophene) Revealed by High-Resolution Transmission Electron Microscopy", *Macromol.* **42**, 1125–1130 (2009).

[362] F. Steiner, C. Poelking, D. Niedzialek, D. Andrienko, and J. Nelson, "Influence of orientation mismatch on charge transport across grain boundaries in tri-isopropylsilylethynyl (TIPS) pentacene thin films", *Phys. Chem. Chem. Phys.* **19**, 10854–10862 (2017).

[363] V. V. Korolkov, A. Summerfield, A. Murphy, D. B. Amabilino, K. Watanabe, T. Taniguchi, and P. H. Beton, "Ultra-high resolution imaging of thin films and single strands of polythiophene using atomic force microscopy", *Nat. Commun.* **10**, 1–8 (2019).

[364] R. Noriega, "Efficient Charge Transport in Disordered Conjugated Polymer Microstructures", *Macro. Rap. Comm.* **39**, 1800096 (2018).

[365] S. A. Mollinger, A. Salleo, and A. J. Spakowitz, "Anomalous Charge Transport in Conjugated Polymers Reveals Underlying Mechanisms of Trapping and Percolation", *ACS Cent. Sci.* **2**, 910–915 (2016).

[366] S. A. Mollinger, B. A. Krajina, R. Noriega, A. Salleo, and A. J. Spakowitz, "Percolation, Tie-Molecules, and the Microstructural Determinants of Charge Transport in Semicrystalline Conjugated Polymers", *ACS Macro Lett.* **4**, 708–712 (2015).

[367] Y. Kim, S. Cook, S. Tuladhar, S. Choulis, J. Nelson, J. R. Durrant, D. Bradley, M. Giles, I. McCulloch, C.-S. Ha, and M. Ree, "A strong regioregularity effect in self-organizing conjugated polymer films and high-efficiency polythiophene:fullerene solar cells", *Nat. Mater.* **5**, 197–203 (2006).

[368] R. Mauer, M. Kastler, and F. Laquai, "The Impact of Polymer Regioregularity on Charge Transport and Efficiency of P3HT:PCBM Photovoltaic Devices", *Adv. Func. Mater.* **20**, 2085–2092 (2010).

[369] P. R. Berger and M. Kim, "Polymer solar cells: P3HT:PCBM and beyond", *J. Renew. Sustain. Energy* **10**, 013508 (2018).

[370] V. Mihailetchi, H. Xie, B. de Boer, L. Koster, and P. Blom, "Charge Transport and Photocurrent Generation in Poly(3-hexylthiophene): Methanofullerene Bulk-Heterojunction Solar Cells", *Adv. Funct. Mater.* **16**, 699–708 (2006).

[371] E. von Hauff, V. Dyakonov, and J. Parisi, "Study of field effect mobility in PCBM films and P3HT:PCBM blends", *Sol. Energ. Mat. Sol. Cells.* **87**, 149 – 156 (2005).

[372] S. Foster, F. Deledalle, A. Mitani, T. Kimura, K.-B. Kim, T. Okachi, T. Kirchartz, J. Oguma, K. Miyake, J. R. Durrant, S. Doi, and J. Nelson, "Electron Collection as a Limit to Polymer:PCBM Solar Cell Efficiency: Effect of Blend Microstructure on Carrier Mobility and Device Performance in PTB7:PCBM", *Adv. Energy Mater.* **4**, 1400311 (2014).

[373] A. Lv, S. R. Puniredd, J. Zhang, Z. Li, H. Zhu, W. Jiang, H. Dong, Y. He, L. Jiang, Y. Li, W. Pisula, Q. Meng, W. Hu, and Z. Wang, "High Mobility, Air Stable, Organic Single Crystal Transistors of an n-Type Diperylene Bisimide", *Adv. Mater.* **24**, 2626–2630 (2012).

[374] P. H. Wöbkenberg, D. D. Bradley, D. Kronholm, J. C. Hummelen, D. M. de Leeuw, M. Cölle, and T. D. Anthopoulos, "High mobility n-channel organic field-effect transistors based on soluble C60 and C70 fullerene derivatives", *Synth. Met.* **158**, 468 – 472 (2008).

[375] F. C. Grozema, P. T. van Duijnen, Y. A. Berlin, M. A. Ratner, and L. D. A. Siebbeles, "Intramolecular Charge Transport along Isolated Chains of Conjugated Polymers: Effect of Torsional Disorder and Polymerization Defects", *J. Phys. Chem. B* **106**, 7791–7795 (2002).

[376] O. J. Sandberg, S. Dahlström, M. Nyman, S. Wilken, D. Scheunemann, and R. Österbacka, "Impact of a Doping-Induced Space-Charge Region on the Collection of Photogenerated Charge Carriers in Thin-Film Solar Cells Based on Low-Mobility Semiconductors", *Phys. Rev. Applied* **12**, 034008 (2019).

[377] T. Kirchartz, T. Agostinelli, M. Campoy-Quiles, W. Gong, and J. Nelson, "Understanding the Thickness-Dependent Performance of Organic Bulk Heterojunction Solar Cells: The Influence of Mobility, Lifetime, and Space Charge", *J. Phys. Chem. Lett.* **3**, 3470–3475 (2012).

[378] M. Stolterfoht, A. Armin, B. Philippa, and D. Neher, "The Role of Space Charge Effects on the Competition between Recombination and Extraction in Solar Cells with Low-Mobility Photoactive Layers", *J. Phys. Chem. Lett.* **7**, 4716–4721 (2016).

[379] G. Dibb, M.-A. Muth, T. Kirchartz, S. Engmann, H. Hoppe, G. Gobsch, M. Thelakkat, N. Blouin, S. Tierney, M. Carrasco-Orozco, J. Durrant, and M. Al-Hashimi, "Influence of doping on charge carrier collection in normal and inverted geometry polymer:fullerene solar cells", *Sci. Rep.* **3**, 3335 (2013).

[380] A. Spies, M. List, T. Sarkar, and U. Würfel, "On the Impact of Contact Selectivity and Charge Transport on the Open-Circuit Voltage of Organic Solar Cells", *Adv. Energy Mater.* **7**, 1601750 (2017).

[381] S. Wilken, O. J. Sandberg, D. Scheunemann, and R. Österbacka, "Watching Space Charge Build Up in an Organic Solar Cell", *Solar RRL* **4**, 1900505 (2020).

[382] P. Kordt, T. Speck, and D. Andrienko, "Finite-size scaling of charge carrier mobility in disordered organic semiconductors", *Phys. Rev. B* **94**, 014208 (2016).

[383] N. P. Holmes, S. Ulum, P. Sista, K. B. Burke, M. G. Wilson, M. C. Stefan, X. Zhou, P. C. Dastoor, and W. J. Belcher, "The effect of polymer molecular weight on P3HT:PCBM nanoparticulate organic photovoltaic device performance", *Sol. Energy Mater. Sol. Cells* **128**, 369 – 377 (2014).

[384] X. Shen, W. Hu, and T. P. Russell, "Measuring the Degree of Crystallinity in Semicrystalline Regioregular Poly(3-hexylthiophene)", *Macromol.* **49**, 4501–4509 (2016).

[385] B. A. Collins, J. R. Tumbleston, and H. Ade, "Miscibility, Crystallinity, and Phase Development in P3HT/PCBM Solar Cells: Toward an Enlightened Understanding of Device Morphology and Stability", *J. Phys. Chem. Lett.* **2**, 3135–3145 (2011).

[386] N. Tombros, A. Veligura, J. Junesch, M. H. Guimarães, I. J. Vera-Marun, H. T. Jonkman, and B. J. Van Wees, "Quantized conductance of a suspended graphene nanoconstriction", *Nature Phys.* **7**, 697–700 (2011).

[387] R. Yamada, H. Kumazawa, T. Noutoshi, S. Tanaka, and H. Tada, "Electrical Conductance of Oligothiophene Molecular Wires", *Nano Lett.* **8**, 1237–1240 (2008).

[388] R. Yamada, H. Kumazawa, S. Tanaka, and H. Tada, "Electrical Resistance of Long Oligothiophene Molecules", *Appl. Phys. Exp.* **2**, 025002 (2009).

[389] V. Stehr, R. F. Fink, M. Tafipolski, C. Deibel, and B. Engels, "Comparison of different rate constant expressions for the prediction of charge and energy transport in oligoacenes", *WIREs Comput. Mol. Sci.* **6**, 694–720 (2016).

[390] L. Ye, S. Zhang, W. Zhao, H. Yao, and J. Hou, "Highly Efficient 2D-Conjugated Benzodithiophene-Based Photovoltaic Polymer with Linear Alkylthio Side Chain", *Chem. Mater.* **26**, 3603–3605 (2014).

[391] L. Huo, S. Zhang, X. Guo, F. Xu, Y. Li, and J. Hou, "Replacing Alkoxy Groups with Alkylthienyl Groups: A Feasible Approach To Improve the Properties of Photovoltaic Polymers", *Angew. Chem. Inter. Ed.* **50**, 9697–9702 (2011).

[392] S. Zhang, M. A. Uddin, W. Zhao, L. Ye, H. Y. Woo, D. Liu, B. Yang, H. Yao, Y. Cui, and J. Hou, "Optimization of side chains in alkylthiothiophene-substituted benzo[1,2-b:4,5-b']dithiophene-based photovoltaic polymers", *Polym. Chem.* **6**, 2752–2760 (2015).

[393] S. Rajaram, R. Shivanna, S. K. Kandappa, and K. S. Narayan, "Nonplanar Perylene Diimides as Potential Alternatives to Fullerenes in Organic Solar Cells", *J. Phys. Chem. Lett.* **3**, 2405–2408 (2012).

[394] J. Yuan, Y. Zhang, L. Zhou, G. Zhang, H.-L. Yip, T.-K. Lau, X. Lu, C. Zhu, H. Peng, P. A. Johnson, M. Leclerc, Y. Cao, J. Ulanski, Y. Li, and Y. Zou, "Single-Junction Organic Solar Cell with over 15% Efficiency Using Fused-Ring Acceptor with Electron-Deficient Core", *Joule* **3**, 1140–1151 (2019).

[395] X. Cui, C. Xiao, T. Winands, T. Koch, Y. Li, L. Zhang, N. L. Doltsinis, and Z. Wang, "Hexacene Diimides", *J. Am. Chem. Soc.* **140**, 12175–12180 (2018).

[396] F. Würthner, C. R. Saha-Möller, B. Fimmel, S. Ogi, P. Leowanawat, and D. Schmidt, "Perylene Bisimide Dye Assemblies as Archetype Functional Supramolecular Materials", *Chem. Rev.* **116**, 962–1052 (2016).

[397] L. Lin, H. Geng, Z. Shuai, and Y. Luo, "Theoretical insights into the charge transport in perylene diimides based n-type organic semiconductors", *Org. Electron.* **13**, 2763 – 2772 (2012).

[398] Z. Ma, T. Winands, N. Liang, D. Meng, W. Jiang, N. L. Doltsinis, and Z. Wang, "A C2-symmetric triple [5]helicene based on N-annulated triperylene hexaimide for chiroptical electronics", *Sci. China Chem.* (2019).

[399] Y. Jin, Z. Chen, M. Xiao, J. Peng, B. Fan, L. Ying, G. Zhang, X.-F. Jiang, Q. Yin, Z. Liang, F. Huang, and Y. Cao, "Thick Film Polymer Solar Cells Based on Naphtho[1,2-c:5,6-c]bis[1,2,5]thiadiazole Conjugated Polymers with Efficiency over 11%", *Adv. Energy Mater.* **7**, 1700944 (2017).

[400] W. Zhao, S. Li, H. Yao, S. Zhang, Y. Zhang, B. Yang, and J. Hou, "Molecular Optimization Enables over 13% Efficiency in Organic Solar Cells", *J. Am. Chem. Soc.* **139**, 7148–7151 (2017).

[401] Y. Cui, H. Yao, B. Gao, Y. Qin, S. Zhang, B. Yang, C. He, B. Xu, and J. Hou, "Fine-Tuned Photoactive and Interconnection Layers for Achieving over 13% Efficiency in a Fullerene-Free Tandem Organic Solar Cell", *J. Am. Chem. Soc.* **139**, 7302–7309 (2017).

[402] Y. Yang, Z.-G. Zhang, H. Bin, S. Chen, L. Gao, L. Xue, C. Yang, and Y. Li, "Side-Chain Isomerization on an n-type Organic Semiconductor ITIC Acceptor Makes 11.77% High Efficiency Polymer Solar Cells", *J. Am. Chem. Soc.* **138**, 15011–15018 (2016).

[403] S. R. Peurifoy, E. Castro, F. Liu, X.-Y. Zhu, F. Ng, S. Jockusch, M. L. Steigerwald, L. Echegoyen, C. Nuckolls, and T. J. Sisto, "Three-Dimensional Graphene Nanostructures", *J. Am. Chem. Soc.* **140**, 9341–9345 (2018).

[404] S. V. Bhosale, C. H. Jani, and S. J. Langford, "Chemistry of naphthalene diimides", *Chem. Soc. Rev.* **37**, 331–342 (2008).

[405] M. Al Kobaisi, S. V. Bhosale, K. Latham, A. M. Raynor, and S. V. Bhosale, "Functional Naphthalene Diimides: Synthesis, Properties, and Applications", *Chem. Rev.* **116**, 11685–11796 (2016).

[406] S. Katsuta, K. Tanaka, Y. Maruya, S. Mori, S. Masuo, T. Okujima, H. Uno, K.-i. Nakayama, and H. Yamada, "Synthesis of pentacene-, tetracene- and anthracene bisimides using double-cyclization reaction mediated by bismuth(iii) triflate", *Chem. Commun.* **47**, 10112–10114 (2011).

[407] W. Jiang, L. Ye, X. Li, C. Xiao, F. Tan, W. Zhao, J. Hou, and Z. Wang, "Bay-linked perylene bisimides as promising non-fullerene acceptors for organic solar cells", *Chem. Commun.* **50**, 1024–1026 (2014).

[408] N. J. Schuster, D. W. Paley, S. Jockusch, F. Ng, M. L. Steigerwald, and C. Nuckolls, "Electron Delocalization in Perylene Diimide Helicenes", *Angew. Chem. Int. Ed.* **55**, 13519–13523 (2016).

[409] Y. Zhong, B. Kumar, S. Oh, M. T. Trinh, Y. Wu, K. Elbert, P. Li, X. Zhu, S. Xiao, F. Ng, M. L. Steigerwald, and C. Nuckolls, "Helical Ribbons for Molecular Electronics", *J. Am. Chem. Soc.* **136**, 8122–8130 (2014).

[410] T. J. Sisto, Y. Zhong, B. Zhang, M. T. Trinh, K. Miyata, X. Zhong, X.-Y. Zhu, M. L. Steigerwald, F. Ng, and C. Nuckolls, "Long, Atomically Precise Donor-Acceptor Cove-Edge Nanoribbons as Electron Acceptors", *J. Am. Chem. Soc.* **139**, 5648–5651 (2017).

[411] W. Fan, T. Winands, N. L. Doltsinis, Y. Li, and Z. Wang, "A Decatwistacene with an Overall 170° Torsion", *Angew. Chem. Int. Ed.* **56**, 15373–15377 (2017).

[412] Y. Huang, Y. Mai, U. Beser, J. Teyssandier, G. Velpula, H. van Gorp, L. A. Straasø, M. R. Hansen, D. Rizzo, C. Casiraghi, R. Yang, G. Zhang, D. Wu, F. Zhang, D. Yan, S. De Feyter, K. Müllen, and X. Feng, "Poly(ethylene oxide) Functionalized Graphene Nanoribbons with Excellent Solution Processability", *J. Am. Chem. Soc.* **138**, 10136–10139 (2016).

[413] D. Meng, G. Liu, C. Xiao, Y. Shi, L. Zhang, L. Jiang, K. K. Baldridge, Y. Li, J. S. Siegel, and Z. Wang, "Corannurylene Pentapetalae", *J. Am. Chem. Soc.* **141**, 5402–5408 (2019).

[414] T. Hosokawa, Y. Takahashi, T. Matsushima, S. Watanabe, S. Kikkawa, I. Azumaya, A. Tsurusaki, and K. Kamikawa, "Synthesis, Structures, and Properties of Hexapole Helicenes: Assembling Six [5]Helicene Substructures into Highly Twisted Aromatic Systems", *J. Am. Chem. Soc.* **139**, 18512–18521 (2017).

[415] M. C. Tamargo and D. O. Cowan, "Triplet exciplex formation in the external heavy-atom effect", *J. Am. Chem. Soc.* **104**, 1107–1109 (1982).

[416] G. Lin, T. Luo, L. Yuan, W. Liang, and H. Xu, "High Performance n-Type and Ambipolar Small Organic Semiconductors for Organic Field-Effect Transistors", *Prog. Chem.* **29**, 1316 (2017).

[417] T. Siegrist, C. Kloc, J. H. Schön, B. Batlogg, R. C. Haddon, S. Berg, and G. A. Thomas, "Enhanced Physical Properties in a Pentacene Polymorph", *Angew. Chem. Int. Ed.* **40**, 1732–1736 (2001).

[418] A. Troisi and G. Orlandi, "Charge-transport regime of crystalline organic semiconductors: Diffusion limited by thermal off-diagonal electronic disorder", *Phys. Rev. Lett.* **96**, 86601–86604 (2006).

[419] B. Bransden, *Quantum Mechanics (2nd Edition)* (Prentice Hall, 2000).

[420] D. Griffiths, *Introduction to quantum mechanics* (Pearson Prentice Hall, Upper Saddle River, NJ, 2005).

[421] G. Liu, C. Xiao, F. Negri, Y. Li, and Z. Wang, "Dodecatwistarene Imides with Zigzag-Twisted Conformation for Organic Electronics", *Angew. Chem. Int. Ed.* **59**, 2008–2012 (2020).

[422] S. Liu, C. Li, Y. Xu, Z. Li, H. Huang, N. Fu, J. Shao, B. Zhao, H. Huang, and W. Huang, "Optoelectronic properties and aggregation effects on the performance of planar versus contorted pyrene-cored perylenediimide dimers for organic solar cells", *Dyes Pigment.* **173**, 107976 (2020).

[423] C. Brückner and B. Engels, "A theoretical description of charge reorganization energies in molecular organic P-type semiconductors", *J. Comput. Chem.* **37**, 1335–1344 (2016).

[424] H. Geng, Y. Niu, Q. Peng, Z. Shuai, V. Coropceanu, and J.-L. Brédas, "Theoretical study of substitution effects on molecular reorganization energy in organic semiconductors", *J. Chem. Phys.* **135**, 104703 (2011).

[425] S. Wilde, L. Stegemann, C. G. Daniliuc, T. Koch, N. L. Doltsinis, and C. A. Strassert, "Studie über den Einfluss des Fluorierungsgrades an einem tetradentaten C^N*N^C-Luminophor auf die photophysikalischen Eigenschaften seiner Platin(II)-Komplexe und deren Aggregation", *Z. Naturforsch. B* **73**, 849–863 (2018).

[426] W. Lu, B.-X. Mi, M. C. W. Chan, Z. Hui, C.-M. Che, N. Zhu, and S.-T. Lee, "Light-Emitting Tridentate Cyclometalated Platinum(II) Complexes Containing σ-Alkynyl Auxiliaries: Tuning of Photo- and Electrophosphorescence", *J. Am. Chem. Soc.* **126**, 4958–4971 (2004).

[427] T. Fleetham, J. Ecton, Z. Wang, N. Bakken, and J. Li, "Single-Doped White Organic Light-Emitting Device with an External Quantum Efficiency Over 20%", *Adv. Mater.* **25**, 2573–2576 (2013).

[428] A. Salehi, X. Fu, D.-H. Shin, and F. So, "Recent Advances in OLED Optical Design", *Adv. Funct. Mater.* **29**, 1808803 (2019).

[429] P. Tyagi, R. Srivastava, L. I. Giri, S. Tuli, and C. Lee, "Degradation of organic light emitting diode: Heat related issues and solutions", *Synth. Met.* **216**, 40 – 50 (2016).

[430] H. Yersin and W. J. Finkenzeller, *Triplet Emitters for Organic Light-Emitting Diodes: Basic Properties*, chapter 1, pages 1–97 (John Wiley & Sons, Ltd, 2008).

[431] H. Yersin, A. F. Rausch, R. Czerwieniec, T. Hofbeck, and T. Fischer, "The triplet state of organo-transition metal compounds. Triplet harvesting and singlet harvesting for efficient OLEDs", *Coord. Chem. Rev.* **255**, 2622 – 2652 (2011).

[432] P. R. Ewen, J. Sanning, T. Koch, N. L. Doltsinis, C. A. Strassert, and D. Wegner, "Spectroscopic mapping and selective electronic tuning of molecular orbitals in phosphorescent organometallic complexes – a new strategy for OLED materials", *Beilstein J. Nanotechnol.* **5**, 2248–2258 (2014).

[433] J. Sanning, P. R. Ewen, L. Stegemann, J. Schmidt, C. G. Daniliuc, T. Koch, N. L. Doltsinis, D. Wegner, and C. A. Strassert, "Rastertunnelspektroskopisch gesteuertes Design massgeschneiderter tiefblauer Triplettemitter", *Angew. Chem.* **127**, 798–803 (2015).

[434] J. Sanning, P. R. Ewen, L. Stegemann, J. Schmidt, C. G. Daniliuc, T. Koch, N. L. Doltsinis, D. Wegner, and C. A. Strassert, "Scanning-Tunneling-Spectroscopy-Directed Design of Tailored Deep-Blue Emitters", *Angew. Chem. Int. Ed.* **54**, 786–791 (2015).

[435] R. Siebert, C. Hunger, J. Guthmuller, F. Schlütter, A. Winter, U. S. Schubert, L. González, B. Dietzek, and J. Popp, "Direct Observation of Temperature-Dependent Excited-State Equilibrium in Dinuclear Ruthenium Terpyridine Complexes Bearing Electron-Poor Bridging Ligands", *J. Phys. Chem. C* **115**, 12677–12688 (2011).

[436] M. Jäger, L. Freitag, and L. González, "Using computational chemistry to design Ru photosensitizers with directional charge transfer", *Coord. Chem. Rev.* **304-305**, 146 – 165 (2015).

[437] T. Koch, C. Höppener, and N. L. Doltsinis, "Conformation-dependent phosphorescence emission of individual mononuclear ruthenium-(ii)-bisterpyridine complexes", *Phys. Chem. Chem. Phys.* **20**, 24921–24926 (2018).

[438] R. C. Evans, P. Douglas, and C. J. Winscom, "Coordination complexes exhibiting room-temperature phosphorescence: Evaluation of their suitability as triplet emitters in organic light emitting diodes", *Coord. Chem. Rev.* **250**, 2093–2126 (2006).

[439] S. I. Bokarev, O. S. Bokareva, and O. Kühn, "A theoretical perspective on charge transfer in photocatalysis. The example of Ir-based systems", *Coord. Chem. Rev.* **304-305**, 133 – 145 (2015).

[440] I. Maisuls, C. Wang, M. E. Gutierrez Suburu, S. Wilde, C.-G. Daniliuc, D. Brünink, N. L. Doltsinis, S. Ostendorp, G. Wilde, J. Kösters, U. Resch-Genger, and C. A. Strassert, "Ligand-controlled and nanoconfinement-boosted luminescence employing Pt(ii) and Pd(ii) complexes: from color-tunable aggregation-enhanced dual emitters towards self-referenced oxygen reporters", *Chem. Sci.* **12**, 3270–3281 (2021).

[441] V. W.-W. Yam and A. S.-Y. Law, "Luminescent d8 metal complexes of platinum(II) and gold(III): From photophysics to photofunctional materials and probes", *Coord. Chem. Rev.* **414**, 213298 (2020).

[442] C. Adachi, "Third-generation organic electroluminescence materials", *Jpn. J. Appl. Phys* **53**, 060101 (2014).

[443] C. Bizzarri, E. Spuling, D. M. Knoll, D. Volz, and S. Bräse, "Sustainable metal complexes for organic light-emitting diodes (OLEDs)", *Coord. Chem. Rev.* **373**, 49–82 (2018).

[444] H. Kaur, S. Sundriyal, V. Pachauri, S. Ingebrandt, K.-H. Kim, A. L. Sharma, and A. Deep, "Luminescent metal-organic frameworks and their composites: Potential future materials for organic light emitting displays", *Coord. Chem. Rev.* **401**, 213077 (2019).

[445] "Photochemistry and photophysics of coordination compounds: platinum", J. G. Williams, in *Photochemistry and Photophysics of Coordination Compounds II*, pages 205–268 (Springer, 2007).

[446] G. Li, X. Zhao, T. Fleetham, Q. Chen, F. Zhan, J. Zheng, Y.-F. Yang, W. Lou, Y. Yang, K. Fang, Z. Shao, Q. Zhang, and Y. She, "Tetradentate Platinum(II)

Complexes for Highly Efficient Phosphorescent Emitters and Sky Blue OLEDs", *Chem. Mater.* **32**, 537–548 (2020).

[447] D. A. K. Vezzu, J. C. Deaton, J. S. Jones, L. Bartolotti, C. F. Harris, A. P. Marchetti, M. Kondakova, R. D. Pike, and S. Huo, "Highly Luminescent Tetradentate Bis-Cyclometalated Platinum Complexes: Design, Synthesis, Structure, Photophysics, and Electroluminescence Application", *Inorg. Chem.* **49**, 5107–5119 (2010).

[448] L. Paterson, F. May, and D. Andrienko, "Computer aided design of stable and efficient OLEDs", *J. Appl. Phys.* **128**, 160901 (2020).

[449] T.-Y. Chu and O.-K. Song, "Hole mobility of N, N'-bis (naphthalen-1-yl)-N, N'-bis (phenyl) benzidine investigated by using space-charge-limited currents", *Appl. Phys. Lett.* **90**, 203512 (2007).

[450] C. A. Strassert, C.-H. Chien, M. D. Galvez Lopez, D. Kourkoulos, D. Hertel, K. Meerholz, and L. De Cola, "Switching On Luminescence by the Self-Assembly of a Platinum(II) Complex into Gelating Nanofibers and Electroluminescent Films", *Angew. Chem. Int. Ed.* **50**, 946–950 (2011).

[451] "Elektronenstrukturrechnungen an metall-organischen Verbindungen", T. Koch, Master thesis, Westfälische Wilhelms-Universität Münster, 2015.

[452] K. Tonigold and A. Groß, "Adsorption of small aromatic molecules on the (111) surfaces of noble metals: A density functional theory study with semiempirical corrections for dispersion effects", *J. Chem. Phys.* **132**, 224701 (2010).