Polymer Folding

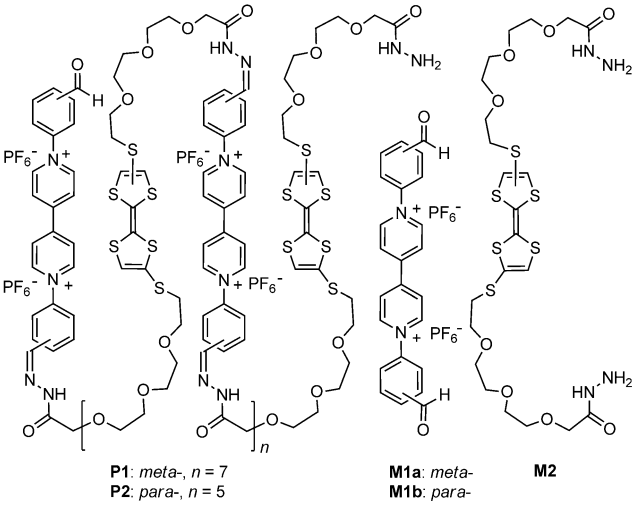
Deutsche Ausgabe: DOI: 10.1002/ange.201410757
Internationale Ausgabe: DOI: 10.1002/anie.201410757

Quadruple Switching of Pleated Foldamers of Tetrathiafulvalene–Bipyridinium Alternating Dynamic Covalent Polymers**

Lan Chen, Hui Wang, Dan-Wei Zhang, Yaming Zhou,* and Zhan-Ting Li**

Abstract: *Two dynamic covalent polymers **P1** and **P2** were prepared by alternately linking electron-rich tetrathiafulvalene (TTF) and electron-deficient bipyridinium ($BIPY^{2+}$) through hydrazone bonds. In acetonitrile, the polymers were induced by intramolecular donor–acceptor interactions to form pleated foldamers, which unfolded upon oxidation of the TTF units to the radical cation $TTF^{\bullet+}$. Reduction of the $BIPY^{2+}$ units to $BIPY^{\bullet+}$ led to the formation of another kind of pleated secondary structures, which are stabilized by intramolecular dimerization of the $BIPY^{\bullet+}$ units. The diradical dicationic cyclophane cyclobis(paraquat-p-phenylene) ($CBPQT^{2(\bullet+)}$) could further force the folded structures to unfold by including the $BIPY^{\bullet+}$ units of the polymers. Upon oxidation of the $BIPY^{\bullet+}$ units of the cyclophane and polymers to $BIPY^{2+}$, the first folded state was regenerated. Switching or conversion between the four conformational states was confirmed by UV/Vis spectroscopic experiments.*

Foldamers are linear molecules or macromolecules that fold into discrete secondary structures stabilized by noncovalent forces.[1–6] Foldamers are ideally suited for the design of responsive molecular systems owing to the dynamic nature of their reversible folding process. Early studies on the conformational switching of foldamers mainly involved changes in temperature or solvent composition.[3] Since then, many examples, including the use of changes in the pH value,[7] donor–acceptor interactions,[8,9] guest complexation,[10] metal coordination,[11] light,[12] and redox reactions[13] as external stimuli, have been reported. Recently, several folding motifs have been applied as key components for the construction of tunable supramolecular devices or machines.[4g,14,15] The incorporation of different driving forces into one backbone may lead to multiresponsive conformation-switching systems: higher-order structures that could potentially be useful in the design of new smart molecular materials. However, this possibility has not been addressed. Recently, it was demonstrated that the homodimerization of conjugated radical cations is highly orthogonal.[16,17] Herein, we describe how the folding–unfolding process of two tetrathiafulvalene (TTF)–bipyridinium ($BIPY^{2+}$) alternating dynamic covalent polymers **P1** and **P2** can be modulated in four different ways by donor–acceptor interactions and radical-cation dimerization.



Polymers **P1** and **P2** were conveniently prepared from dialdehyde **M1a** or **M1b** and the di(acylhydrazine) **M2** in MeCN through the formation of acylhydrazones: a well-established dynamic covalent approach for the generation of dynamic polymers.[18] Polymers **P1** and **P2** were soluble in MeCN. Their average degree of polymerization was determined to be approximately 8 and 6, respectively, by ^{1}H NMR spectroscopy in [D_3]MeCN on the basis of the relative integrals of the signals for the aldehyde hydrogen atom and the α hydrogen atoms of the $BIPY^{2+}$ units (see the Supporting Information).

As compared to that of the control **M2**, the signal of the TTF hydrogen atoms of **P1** and **P2** in [D_3]MeCN shifted upfield by 0.17 and 0.12 ppm, respectively (see the Supporting Information), thus suggesting that these signals were shielded by neighboring $BIPY^{2+}$ units. Furthermore, the degree of shifting was concentration-independent. This result may be regarded as the first evidence for backbone folding driven by intramolecular donor–acceptor interactions between TTF and $BIPY^{2+}$ units.[19]

UV/Vis spectra were then recorded for **P1** and **P2** in MeCN. Both polymers exhibited the typical absorption band of TTF–$BIPY^{2+}$ charge-transfer (CT) complexation (Figure 1). This absorption was centered at 820 and 740 cm,[19,20] respectively. The molar absorption coefficient

[*] L. Chen, Dr. H. Wang, Prof. Dr. D.-W. Zhang, Prof. Dr. Y. Zhou, Prof. Dr. Z.-T. Li
Department of Chemistry, Collaborative Innovation Center of Chemistry for Energy Materials (iChEM), Fudan University
220 Handan Road, Shanghai 200433 (China)
E-mail: zhangdw@fudan.edu.cn
ztli@fudan.edu.cn

[**] We are grateful to the Ministry of Science and Technology of China (2013CB834501), the Ministry of Education (IRT1117) and Research Fund for the Doctoral Program of China, the Science and Technology Commission of Shanghai Municipality (13M1400200), and the National Natural Science Foundation of China (21472023, 21432004) for financial support.

Supporting information for this article is available on the WWW under http://dx.doi.org/10.1002/anie.201410757.

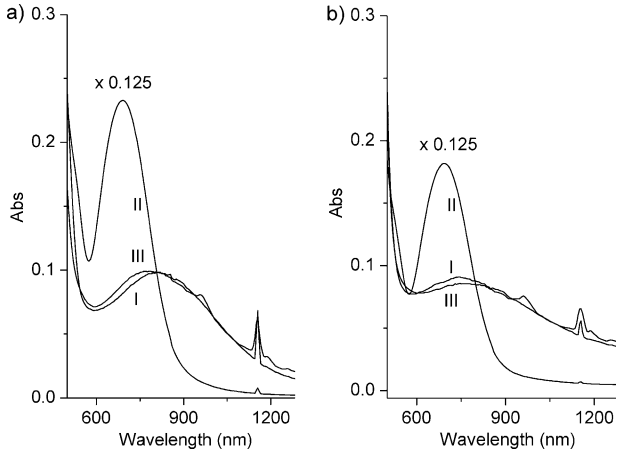


Figure 1. UV/Vis absorption spectra of polymers a) **P1** and b) **P2** in MeCN ([TTF] = [$BIPY^{2+}$] = 0.50 mM) at 25 °C; I: pure solution; II: after the addition of $Fe(ClO_3)_4$ (1.0 equiv, 0.50 mM); III: after the further addition of ascorbic acid (1.0 equiv, 0.50 mM).

(ε) corresponding to [TTF] and [$BIPY^{2+}$] was calculated to be 210 and 228 $L\,mol^{-1}\,cm^{-1}$, respectively. Upon dilution of the solutions from 1.3 to 0.3 mM, the CT absorbance decreased linearly (see the Supporting Information). That is, the Beer–Lambert law was observed, but the maximum absorption wavelength did not shift. For both polymers, this linear relationship was also time-independent within 1 week, thus implying that their hydrazone bonds did not break or exchange at room temperature. At even lower concentrations, the CT band was too weak to be determined accurately. These results suggest that CT complexation occurred intramolecularly through backbone folding. We thus proposed a pleated folding pattern for the complexation (Figure 2, Folded A), because this pattern enables the maximum number of donor–acceptor interactions.[9] An increase in temperature weakened the CT absorption: At 60 °C, the CT absorption of **P1** and **P2** was weakened by 35 and 40 %, respectively (see the Supporting Information). The addition of the electron-rich macrocyclic polyether bis-1,5-dinaphtho[38]crown-10 (1.0 mM, 2.0 equiv relative to [$BIPY^{2+}$]) to the solution of **P1** or **P2** in MeCN did not cause any change in the CT absorption band of the polymer, thus indicating that the folded conformation of both polymers was quite stable and the electron-rich cyclophane was not able to break the folding process through encapsulation of the $BIPY^{2+}$ units of the polymers.

The addition of 1 equivalent of $Fe(ClO_4)_3$ relative to [TTF] to the solution of **P1** and **P2** caused single-electron oxidation of the TTF units to the radical cation $TTF^{\bullet+}$, as indicated by the appearance of a strong absorption band at around 690 nm and the disappearance of the CT absorption band (Figure 1).[21] The formation of $TTF^{\bullet+}$ was further supported by electron paramagnetic resonance (EPR) spectroscopy. An EPR signal was observed for monomeric $TTF^{\bullet+}$[22] and was nearly identical for samples of **P1**, **P2**, and monomer **M2** in MeCN (see the Supporting Information). The UV/Vis spectra of **P1** and **P2** did not show the absorption band of the dimer $(TTF^{\bullet+})_2$,[17] even at the saturation concentration (about 1.0 mM; see the Supporting Information). Moreover, for both polymers, the strength of the EPR signal of monomeric $TTF^{\bullet+}$ was linearly related to the concentration. These results all indicated that the two polymers adopted unfolded, flexible conformations (Figure 2, Unfolded A). The fact that the stacking of the $TTF^{\bullet+}$ units in both polymers did not occur intra- or intermolecularly may be rationalized by considering the electrostatic repulsion of the $BIPY^{2+}$ units, because both inter- and intramolecular stacking of the $TTF^{\bullet+}$ units would shorten the distance between the two different cationic species.

The addition of ascorbic acid (1.0 equiv) to the above solutions of **P1** and **P2** regenerated neutral TTF,[23] and thus recovered the Folded A state: UV/Vis spectra of the resulting solutions showed a CT absorption band of comparable strength to that of the original polymer prior to treatment with $Fe(ClO_4)_3$, accompanied by the disappearance of the absorption band of $TTF^{\bullet+}$ (Figure 1).

The $BIPY^{2+}$ units of **P1** and **P2** were further reduced to $BIPY^{\bullet+}$ in MeCN with an excess of zinc dust. The UV/Vis spectrum of **P1** displayed two absorption bands at around 450 and 650 nm (Figure 3 a), which could be assigned to the $BIPY^{\bullet+}$ monomer.[17] The spectrum also showed a band at around 985 nm. This band was not generated by monomeric **M1 a** with the same $BIPY^{\bullet+}$ concentration (see the Supporting Information). Thus, this band could be attributed to the

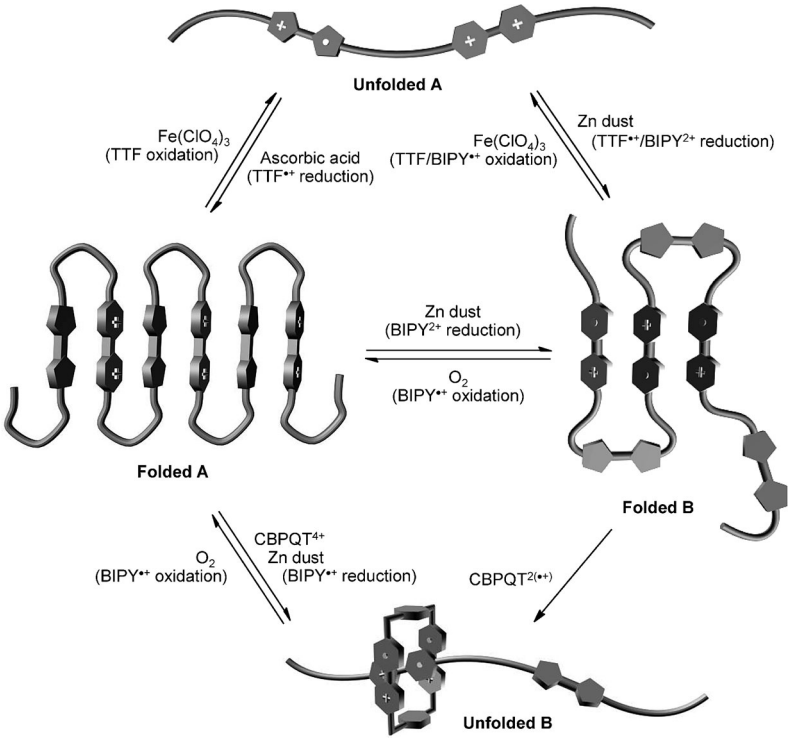


Figure 2. Multiply tuned folding–unfolding processes of polymers **P1** and **P2**, as driven by donor–acceptor interactions and the dimerization of conjugated radical cations.

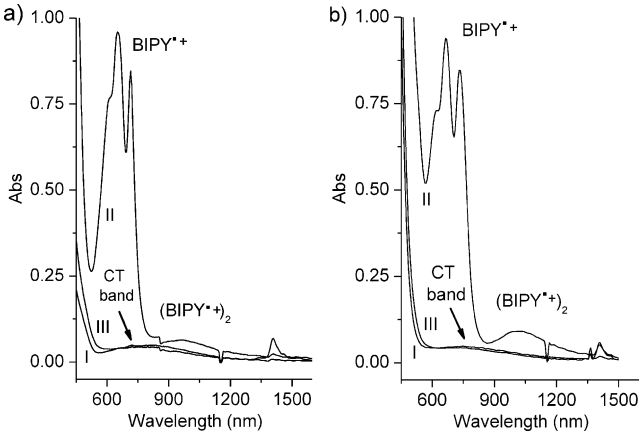


Figure 3. UV/Vis absorption spectra of polymers a) **P1** and b) **P2** in MeCN ([$BIPY^{2+}$] = 0.25 mM) at 25 °C; I: pure solution; II: after the addition of excess Zn dust; III: after exposure of the solutions to air for 2 h.

formation of the radical-cation dimer $(BIPY^{\bullet+})_2$.[24] A similar absorption band of $(BIPY^{\bullet+})_2$, at around 1045 nm, was also observed for **P2** (Figure 3b). UV/Vis dilution experiments (0.8–0.1 mM) showed that the $(BIPY^{\bullet+})_2$ absorption was linearly concentration-dependent (see the Supporting Information), which supports the hypothesis that the dimerization took place intramolecularly. Thus, it is reasonable to propose that the polymers incorporating $BIPY^{\bullet+}$ formed another kind of pleated foldamer (Figure 2, Folded B). For both polymers, this linear relationship was time-independent within 12 h, thus implying that their hydrazone bonds were also stable at room temperature when the viologen units existed as radical cations. Since dimerized *N*,*N*′-diphenyl-substituted $BIPY^{\bullet+}$ units are connected by a flexible linker of 36 atoms that incorporates a TTF unit, the formation of this folded state reflects the strength of the intramolecular dimerization of the $BIPY^{\bullet+}$ units. The $BIPY^{\bullet+}$ signal of **P1** and **P2** in the EPR spectra in MeCN was also substantially weaker than that of the corresponding controls **M1a** and **M1b** at the same concentration (see the Supporting Information). This result further supports the hypothesis that the $BIPY^{\bullet+}$ units of the polymers had partially dimerized into EPR-silent $(BIPY^{\bullet+})_2$. The $BIPY^{\bullet+}$ radical cation could be reversibly oxidized by oxygen in air to $BIPY^{2+}$. This process led to the recovery of the donor–acceptor-interaction-driven foldamers (Figure 2, Folded A), as evidenced by UV/Vis spectroscopy, which showed the regeneration of CT absorption bands of comparable strength to those observed for the original polymers (Figure 3).

The $TTF^{\bullet+}$ and $BIPY^{2+}$ units of **P1** and **P2** in the state of Unfolded A (Figure 2) could be reduced simultaneously with an excess of Zn dust to TTF and $BIPY^{\bullet+}$, respectively. The UV/Vis spectra of the resulting solutions were nearly identical to those recorded after the treatment of solutions of the polymers in the state of Folded A with Zn dust (see the Supporting Information), thus suggesting that the Unfolded A state could be converted into the Folded B state directly. The addition of 1.0 equivalent of $Fe(ClO_4)_3$ led to the conversion of the Folded B state into the Folded A state for both samples, whereas the addition of 2.0 equivalents of $Fe(ClO_4)_3$ led to the conversion of the Folded B state into the Unfolded A state. Both processes were supported by UV/Vis spectroscopy (see the Supporting Information).

Addition of the electron-deficient cyclophane cyclobis(paraquat-*p*-phenylene) ($CBPQT^{4+}$ $4PF_6^-$; 1.0 equiv relative to [TTF], 0.25 mM) to a solution of **P1** or **P2** in MeCN caused a slight increase in the CT absorption band of both polymers, at around 820 and 740 nm, respectively. In contrast, the addition of paraquat ($2PF_6^-$; 2 equiv, 0.50 mM) did not produce any observable increase in the CT absorption. It has been established that, owing to electrostatic repulsion, the $CBPQT^{4+}$ ring is not able to move over a $BIPY^{2+}$ unit incorporated in a linear molecule.[25] Thus, the slight enhancement in the CT absorption of **P1** and **P2** in the presence of $CBPQT^{4+}$ might be attributed to the encapsulation of part of the TTF unit at the end of the polymer backbone. The two mixtures were further treated with an excess of Zn dust to reduce the $BIPY^{2+}$ units of the polymer and $CBPQT^{4+}$ to $BIPY^{\bullet+}$. UV/Vis spectra of the two resulting solutions exhibited the absorption, at around 1012 and 1050 nm for **P1** and **P2**, respectively, of dimer $(BIPY^{\bullet+})_2$ (see the Supporting Information). As compared to that of the corresponding polymer solution that contained no $CBPQT^{\bullet+}$ (Figure 3), the absorbance was enhanced by 289 and 203%, respectively. In contrast, the addition of radical-cationic paraquat (0.50 mM) led to only 9 and 16% enhancement of the $(BIPY^{\bullet+})_2$ absorption (see the Supporting Information). At the studied concentration, a solution of $CBPQT^{2(\bullet+)}$ or radical-cationic paraquat itself did not lead to the appearance of any absorption band of the dimer $(BIPY^{\bullet+})_2$. Thus, the remarkable enhancement of the $(BIPY^{\bullet+})_2$ absorption by the addition of $CBPQT^{2(\bullet+)}$ could be ascribed to the threading of the $BIPY^{\bullet+}$ units of the polymers into the cavity of $CBPQT^{2(\bullet+)}$. The enhancement was magnified upon further addition of the cyclophane to a concentration of 0.5 mM (see the Supporting Information). Above this concentration, intermolecular dimerization of the $BIPY^{\bullet+}$ units of the cyclophane occurred, as indicated by the appearance of the $(BIPY^{\bullet+})_2$ absorption band at around 1100 nm in the UV/Vis spectrum (see the Supporting Information). These observations indicate that threading complexation took place by stronger intermolecular $BIPY^{\bullet+}$ dimerization between the polymer and the cyclophane, which led to the formation of another unfolded state (Figure 2, Unfolded B), even though the complexation could not reach saturation owing to the solubility limit. As expected, when the solutions were exposed to air, the $BIPY^{\bullet+}$ units of the polymers and the cyclophane were oxidized by oxygen to $BIPY^{2+}$, and the state of Folded A was regenerated (see the Supporting Information). The adding of $CBPQT^{2(\bullet+)}$ to the solution of the polymers of the Folded B state afforded similar results, which were supported by UV/Vis spectroscopic experiments. Because the Unfolded B state was driven by intermolecular $BIPY^{\bullet+}$ dimerization, an increase in the concentration of the components would facilitate its formation.

We have demonstrated that tetrathiafulvalene–bipyridinium alternating dynamic covalent polymers can adopt two pleated folded conformations and two unfolded, flexible

conformations. The key for controlling the formation of four different conformational states of the polymers is the introduction of two different noncovalent forces, that is, a donor–acceptor interaction between the tetrathiafulvalene and bipyridinium units and orthogonal dimerization of their respective radical cations. The four folded and unfolded states can be tuned or switched by reversible regulation of the redox states of the two aromatic segments. This system is the first example of multiple tuning of the ordered conformations of long flexible backbones. Further investigations will address the introduction of photocontrollable segments and chiral units. We first plan to develop a multiply repeated conformational-switching motif for the creation of new smart structures. Such a motif would also avoid the formation of by-products observed with the oxidation–reduction approach. In a second study, we will examine the formation of new artificial secondary structures to emulate the elegance of natural peptides and proteins.

Keywords: dynamic covalent polymers · donor–acceptor interactions · foldamers · radical cations · viologens

Zitierweise: *Angew. Chem. Int. Ed.* **2015**, *54*, 4028–4031
Angew. Chem. **2015**, *127*, 4100–4103

[1] *Foldamers: Structure, Properties, and Applications* (Eds.: S. Hecht, I. Huc), Wiley-VCH, Weinheim, **2007**, p. 456.
[2] a) S. H. Gellman, *Acc. Chem. Res.* **1998**, *31*, 173–180; b) H. Yin, A. D. Hamilton, *Angew. Chem. Int. Ed.* **2005**, *44*, 4130–4163; *Angew. Chem.* **2005**, *117*, 4200–4235; c) V. Azzarito, K. Long, N. S. Murphy, A. J. Wilson, *Nat. Chem.* **2013**, *5*, 161–173.
[3] D. J. Hill, M. J. Mio, R. B. Prince, T. S. Hughes, J. S. Moore, *Chem. Rev.* **2001**, *101*, 3893–4011.
[4] a) I. Huc, *Eur. J. Org. Chem.* **2004**, 17–29; b) B. Gong, *Chem. Eur. J.* **2001**, *7*, 4336–4342; c) Z.-T. Li, J.-L. Hou, C. Li, *Acc. Chem. Res.* **2008**, *41*, 1343–1353; d) H. Juwarker, J.-M. Suk, K.-S. Jeong, *Chem. Soc. Rev.* **2009**, *38*, 3316–3325; e) Q. Gan, Y. Wang, H. Jiang, *Curr. Org. Chem.* **2011**, *15*, 1293–1301; f) P. Prabhakaran, G. Priya, G. J. Sanjayan, *Angew. Chem. Int. Ed.* **2012**, *51*, 4006–4008; *Angew. Chem.* **2012**, *124*, 4079–4081; g) D.-W. Zhang, X. Zhao, J.-L. Hou, Z.-T. Li, *Chem. Rev.* **2012**, *112*, 5271–5316.
[5] a) X. Li, D. Yang, *Chem. Commun.* **2006**, 3367–3379; b) X. Li, Y.-D. Wu, D. Yang, *Acc. Chem. Res.* **2008**, *41*, 1428–1438.
[6] Y. Zhao, K. Cho, L. Widanapathirana, S. Zhang, *Acc. Chem. Res.* **2013**, *46*, 2763–2772.
[7] a) C. Dolain, V. Maurizot, I. Huc, *Angew. Chem. Int. Ed.* **2003**, *42*, 2738–2740; *Angew. Chem.* **2003**, *115*, 2844–2846; b) E. Kolomiets, V. Berl, J.-M. Lehn, *Chem. Eur. J.* **2007**, *13*, 5466–5479; c) H.-Y. Hu, J.-F. Xiang, Y. Yang, C.-F. Chen, *Org. Lett.* **2008**, *10*, 1275–1278.
[8] A. Das, S. Ghosh, *Angew. Chem. Int. Ed.* **2014**, *53*, 2038–2054; *Angew. Chem.* **2014**, *126*, 2068–2084.
[9] a) R. S. Lokey, B. L. Iverson, *Nature* **1995**, *375*, 303–305; b) S. Ghosh, S. Ramakrishnan, *Angew. Chem. Int. Ed.* **2004**, *43*, 3264–3268; *Angew. Chem.* **2004**, *116*, 3326–3330; c) S. Ghosh, S. Ramakrishnan, *Angew. Chem. Int. Ed.* **2005**, *44*, 5441–5447; *Angew. Chem.* **2005**, *117*, 5577–5583; d) X. Zhao, M.-X. Jia, X.-K. Jiang, L.-Z. Wu, Z.-T. Li, G.-J. Chen, *J. Org. Chem.* **2004**, *69*, 270–279; e) K. Liu, X. Zheng, A. Z. Samuel, S. G. Ramkumar, S. Ghosh, X. Tan, D. Wang, Z. Shuai, S. Ramakrishnan, D. Liu, X. Zhang, *Langmuir* **2013**, *29*, 14438–14443.
[10] a) Y.-X. Xu, X. Zhao, X.-K. Jiang, Z.-T. Li, *J. Org. Chem.* **2009**, *74*, 7267–7273; b) Y. Wang, J. Xiang, H. Jiang, *Chem. Eur. J.* **2011**, *17*, 613–619; c) K. P. McDonald, Y. Hua, S. Lee, A. H. Flood, *Chem. Commun.* **2012**, *48*, 5065–5075; d) C. Sun, C. Ren, Y. Wei, B. Qin, H. Zeng, *Chem. Commun.* **2013**, *49*, 5307–5309; e) L. Cera, C. A. Schalley, *Chem. Sci.* **2014**, *5*, 2560–2567.
[11] R. M. Meudtner, M. Ostermeier, R. Goddard, C. Limberg, S. Hecht, *Chem. Eur. J.* **2007**, *13*, 9834–9840.
[12] a) Z. Yu, S. Hecht, *Angew. Chem. Int. Ed.* **2011**, *50*, 1640–1643; *Angew. Chem.* **2011**, *123*, 1678–1681; b) A. Khan, C. Kaiser, S. Hecht, *Angew. Chem. Int. Ed.* **2006**, *45*, 1878–1881; *Angew. Chem.* **2006**, *118*, 1912–1915; c) Y. Wang, F. Bie, H. Jiang, *Org. Lett.* **2010**, *12*, 3630–3633; d) S. Lee, Y. Hua, A. H. Flood, *J. Org. Chem.* **2014**, *79*, 8383–8396; e) Y. Hua, A. H. Flood, *J. Am. Chem. Soc.* **2010**, *132*, 12838–12840; f) Z. Yu, S. Hecht, *Chem. Eur. J.* **2012**, *18*, 10519–10524; g) Z. Yu, S. Hecht, *Angew. Chem. Int. Ed.* **2013**, *52*, 13740–13744; *Angew. Chem.* **2013**, *125*, 13985–13989; h) Z. Yu, S. Weidner, T. Risse, S. Hecht, *Chem. Sci.* **2013**, *4*, 4156–4167.
[13] D. Siebler, M. Linseis, T. Gasi, L. M. Carrella, R. F. Winter, C. Förster, K. Heinze, *Chem. Eur. J.* **2011**, *17*, 4540–4551.
[14] D.-W. Zhang, X. Zhao, Z.-T. Li, *Acc. Chem. Res.* **2014**, *47*, 1961–1970.
[15] K.-D. Zhang, X. Zhao, G.-T. Wang, Y. Liu, Yi. Zhang, H.-J. Lu, X.-K. Jiang, Z.-T. Li, *Angew. Chem. Int. Ed.* **2011**, *50*, 9866–9870; *Angew. Chem.* **2011**, *123*, 10040–10044.
[16] D.-W. Zhang, J. Tian, L. Chen, L. Zhang, Z.-T. Li, *Chem. Asian J.* **2015**, *10*, 56–68.
[17] a) M. Schmittel, K. Mahata, *Angew. Chem. Int. Ed.* **2008**, *47*, 5284–5286; *Angew. Chem.* **2008**, *120*, 5364–5366; b) C.-H. Wong, S. C. Zimmerman, *Chem. Commun.* **2013**, *49*, 1679–1695.
[18] a) J.-M. Lehn, *Chem. Eur. J.* **1999**, *5*, 2455–2463; b) S. J. Rowan, S. J. Cantrill, G. R. L. Cousins, J. K. M. Sanders, J. F. Stoddart, *Angew. Chem. Int. Ed.* **2002**, *41*, 898–952; *Angew. Chem.* **2002**, *114*, 938–993; c) P. T. Corbett, J. Leclaire, L. Vial, K. R. West, J.-L. Wietor, J. K. M. Sanders, S. Otto, *Chem. Rev.* **2006**, *106*, 3652–3711.
[19] a) D. Philp, A. M. Z. Slawin, N. Spencer, J. F. Stoddart, D. J. Williams, *J. Chem. Soc. Chem. Commun.* **1991**, 1584–1586; b) Z.-T. Li, P. C. Stein, J. Becher, D. Jensen, P. Mörk, N. Svenstrup, *Angew. Chem. Int. Ed. Engl.* **1995**, *34*, 2524–2528; *Angew. Chem.* **1995**, *107*, 2719–2723.
[20] a) Y. Liu, A. H. Flood, J. F. Stoddart, *J. Am. Chem. Soc.* **2004**, *126*, 9150–9151; b) Y. Liu, A. H. Flood, R. M. Moskowitz, J. F. Stoddart, *Chem. Eur. J.* **2005**, *11*, 369–385; c) S. Nygaard, B. W. Laursen, T. S. Hansen, A. D. Bond, A. H. Flood, J. O. Jeppesen, *Angew. Chem. Int. Ed.* **2007**, *46*, 6093–6097; *Angew. Chem.* **2007**, *119*, 6205–6209; d) E. H. Witlicki, S. S. Andersen, S. W. Hansen, J. O. Jeppesen, E. W. Wong, L. Jensen, A. H. Flood, *J. Am. Chem. Soc.* **2010**, *132*, 6099–6107; e) S. S. Andersen, A. I. Share, B. La Cour Poulsen, M. Kørner, T. Duedal, C. R. Benson, S. W. Hansen, J. O. Jeppesen, A. H. Flood, *J. Am. Chem. Soc.* **2014**, *136*, 6373–6384.
[21] A. Credi, M. Montalti, V. Balzani, S. J. Langford, F. M. Raymo, J. F. Stoddart, *New J. Chem.* **1998**, *22*, 1061–1065.
[22] A. Y. Ziganshina, Y. H. Ko, W. S. Jeon, K. Kim, *Chem. Commun.* **2004**, 806–807.
[23] I. Hwang, A. Y. Ziganshina, Y. H. Ko, G. Yun, K. Kim, *Chem. Commun.* **2009**, 416–418.
[24] a) C. Zhou, J. Tian, J.-L. Wang, D.-W. Zhang, X. Zhao, Y. Liu, Z.-T. Li, *Polym. Chem.* **2014**, *5*, 341–345; b) L. Zhang, T.-Y. Zhou, J. Tian, H. Wang, D.-W. Zhang, X. Zhao, Y. Liu, Z.-T. Li, *Polym. Chem.* **2014**, *5*, 4715–4721.
[25] H. Li, Y.-L. Zhao, A. C. Fahrenbach, S.-Y. Kim, W. F. Paxton, J. F. Stoddart, *Org. Biomol. Chem.* **2011**, *9*, 2240–2250.

Received: November 4, 2014
Published online: January 30, 2015

