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Luminescent Liquids

Solvent-Free Luminescent Organic Liquids**

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A prospective research scenario of organic electronics[1] utilizes noncovalent interactions to assemble optoelectronically active molecules[2] to attain improved performance in devices, such as field-effect transistors[3] and solar cells,[4] because it may enable simple and cheap manufacture as well as easy defect annealing. In order to compete with inorganic materials, utmost care is needed for organic substances, from molecular design to self-organization, fabrication into devices in a predictable way, and finally to end-use applications with improved performance and longevity.[4,5] Therefore, alternative and qualitatively different approaches should be considered in this direction.

Softening of the optoelectronic functional materials is one such example towards printable organic electronics. The formulation of solvent-free organic materials, such as ionic liquids,[6] ionic liquids that contain nanoparticles,[7] or organic chromophores,[8] is an emerging and challenging area which aims to find replacements for self-assembled organic semiconductors.[9] Recently, uncharged room-temperature organic liquids have been introduced as new functional liquids by isolating the π-core through the use of low-viscosity organic chains. Room-temperature, solvent-free organic liquids, such as phthalocyanines,[10] porphyrins,[11] carbazoles[12] and fullerenes[13] have been reported as proof of this concept. However, it is still premature to deliver excellent end-use performance and meaningful applications. The use of organic liquids at room temperature is expected to provide several benefits, for example, nonvolatility, processing under solvent-free conditions, tunable optoelectronic functions, a high density of electronically active π-conjugated moieties, or the ability to act as solvent/matrix for other organic or inorganic components. Furthermore, the liquid materials are tractable, and can be easily treated and used on a bulk scale.

The linear π-conjugated molecule oligo(*p*-phenylenevinylene) (OPV) was chosen as the functional core moiety. OPV has been widely studied in organic optoelectronics because it has excellent stability and emission characteristics, as well as self-assembly properties.[2,14] Herein, we report the synthesis of a series of room-temperature liquid OPVs (**1**–**4**, Figure 1 a), as well as their use as a solvent/matrix and a blue-emitting component for the preparation of liquid inks that emit white light. This study paves the way to light-emitting liquids which can be painted onto various surfaces that have different geometries.

Room temperature, solvent-free, liquid OPVs were synthesized by substituting two different OPV cores with low-viscosity hydrocarbons, such as branched aliphatic chains (Figure 1 a). For example, the complex viscosities (η^*) of the branched alkyl bromide uncoupled (**2b**) and coupled to benzaldehyde (**2a**) (Scheme S1 in the Supporting Information) are 1.03 and 0.01 Pa s, respectively, at an angular frequency of $\omega = 10\,\mathrm{rad\,s^{-1}}$ (Figure S1 in the Supporting Information). The molecular design strategy, which includes the position of the alkyl chain substituent and the extent of chain branching (Figure 1 a) are extremely important to tune the physical features of the liquid. The targeted branched chain, coupled OPV derivatives **1**–**4** were obtained as pale yellow viscous fluids at room temperature, whereas the reference molecules **5** and **6** were solids. All of the OPV derivatives were unambiguously identified by ^{1}H NMR spectroscopy and MALDI-TOF mass spectrometry. The ^{1}H NMR spectrum (Figure S2 in the Supporting Information) and thermogravimetric analysis (TGA, Figure S3 in the Supporting Information) of the fluids indicates the absence of residual solvent in the bulk materials, hence the fluid-like behavior is an inherent property of the single bulk component.

The fluid was evaluated by differential scanning calorimetry (DSC) and rheology analyses. As shown in Figure 1 b, the cooling traces of the DSC thermogram indicate that **1**–**4** have relatively low glass transition temperatures (T_g) between −43 to −55 °C (Table S1 in the Supporting Information), which shows that over a wide range of temperature, until decomposition at around 300 °C (estimated by TGA), these compounds exist as solvent-free fluids. The molar heat capacities (C_{mol}) of **1**–**4** at the glass transition temperature are between 639.4 (**1**) and 679.5 J mol^{-1} K^{-1} (**4**, Table S1 in the Supporting

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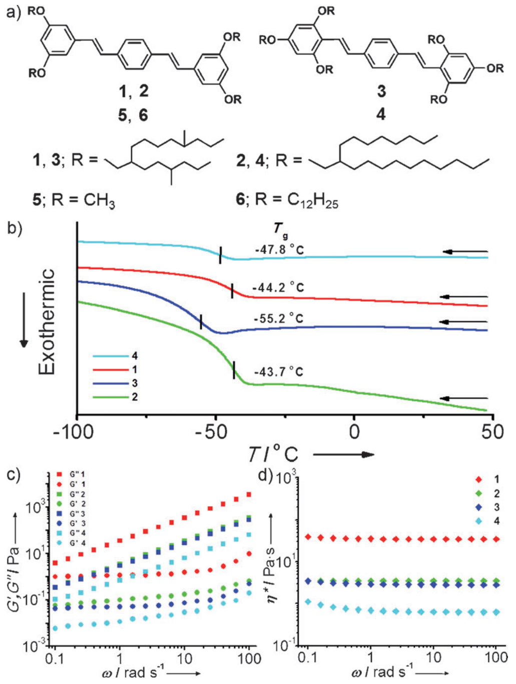

Figure 1. a) Chemical structures of OPVs **1**–**6**. b) The cooling trace of DSC thermograms of **1** (red), **2** (green), **3** (dark blue), and **4** (light blue) showing the glass transition temperatures (T_g). c) Variation of storage modulus (G', circles) and loss modulus (G'', squares) versus angular frequency on double-logarithmic scale and d) complex viscosity (η^*) versus angular frequency on double-logarithmic scale for **1** (red), **2** (green), **3** (dark blue), and **4** (light blue); shear strain (γ) = 0.1.

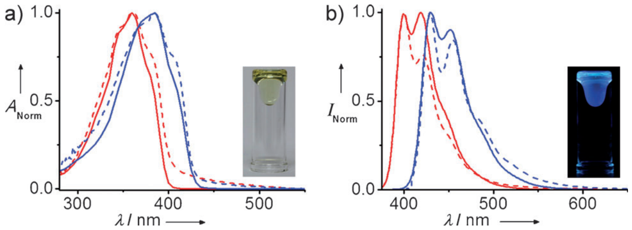

Figure 2. Comparison of the normalized a) absorption and b) emission spectra of solvent-free **2** (red) and **4** (blue) in bulk-liquid state (dashed line) supported on quartz plate and dichloromethane solution (solid line). $c = 5 \times 10^{-5}$ M, $l = 1$ mm, $\lambda_{ex} = 360$ nm for **2** and $\lambda_{ex} = 385$ nm for **4**. Insets show images of **2** under a) visible and b) UV light (365 nm).

Information). As the viscosity decreases, the molar heat capacity increases, and this can be correlated with the fluidity of **1**–**4**. The loss modulus G'' is higher than the storage modulus G' over the measured angular frequency range of $\omega = 0.1$–100 rad s^{-1} and shear strain of $\gamma = 0.1$. This result clearly indicates that the target molecules **1**–**4** behave as Newtonian liquids (Figure 1 c). The value of η^* was found to vary significantly from 34.6 (**1**) to 0.64 Pa s (**4**) at $\omega = 10$ rad s^{-1} (Figure 1 d, see also Table S1 in the Supporting Information). This result shows that a (2,4,6)-substitution pattern on both sides of the phenyl moieties and swallow-tailed aliphatic chains are more effective than a (3,5)-substitution pattern and hyper-branched chains to attain low-viscosity OPV liquids. In other words, together with the spectroscopic results discussed below (Figure 2, see also Figure S6 in the Supporting Information), this molecular design can efficiently disturb intermolecular π–π interactions of the OPV core by steric hindrance via soft tail attachment (Figure S4 in the Supporting Information) and as a consequence, low-viscosity organic liquids are produced. Comparing the physical properties of the liquids **1**–**4** with the two solid reference compounds **5** and **6** (Figure 1 a, see also Table S2 in the Supporting Information), in which the OPV core has methoxy and dodecyloxy substituents, respectively, stresses the importance of the branched aliphatic side chains for tuning the intermolecular interactions. In compound **6**, π–π interactions between the aryl groups and van der Waals forces between the linear alkyl chains contribute to the assembly and solid characteristics. In such cases, the melting point stays at high temperatures, 171 and 64 °C for **5** and **6**, respectively. Furthermore, X-ray diffraction (XRD) analyses of the liquid samples showed only two halos that correspond to the average distance between the disordered OPV core (13.6–18.2 Å) and the molten aliphatic chains (4.6–4.9 Å, Figure S5a in the Supporting Information). The XRD results point to no regular molecular ordering in the densely dispersed OPV cores in liquids **1**–**4**. The isolation of the OPV core by branched hydrocarbon chains thus enables the OPVs to be liquid at room temperature, and basic ^{1}H NMR, TGA, DSC, XRD, and rheology experiments lead to the conclusion that fluidity is an inherent characteristic of these compounds (Table S1 in the Supporting Information).

Comparison of the absorption and emission spectra of **1**–**4** in the solvent-free liquid state and in homogeneous solution (Figure 2, see also Figures S6 and S7 in the Supporting Information) indicates that in the solvent-free liquid state, π–π interactions between the cores are efficiently inhibited by the introduction of branched alkyl chains. The slightly broader absorption and emission spectra in the solvent-free state relative to monomers in dichloromethane solution illustrate that the OPV core is in an almost monomerically dispersed and highly dense state, but probably with small clustering. The optical inhomogeneity of the solvent-free liquid leads to light scattering, which appears as a tail that extends to longer wavelengths (Figure 2 a).[15a] In Figure 2 b, the spectral peak that corresponds to the 0–0 vibronic band ($\lambda_{max} = 400$ nm (**2**), 430 nm (**4**)) of the $S_1 \rightarrow S_0$ transition of the fluorescence spectra in solution and solvent-free states are almost identical, whereas the 1–0 band ($\lambda_{max} = 418$ nm (**2**), 453 nm (**4**)) has a lower intensity in the solvent-free liquids, which indicates a difference in the stabilization of the excited states in the solvent-free versus solution states.[15b] The insets of Figure 2 a and b show images of **2** at room temperature under visible and UV light ($\lambda_{ex} = 365$ nm), respectively, which shows the viscous and blue emissive characteristics in the liquid state. All of the liquids **1**–**4** emit blue light in the solvent-free bulk state with an absolute fluorescence quantum yield of 45–48 % (Table S2 in the Supporting Information). To have an insight into our design strategy, the refractive index (RI) was also evaluated by using 589.3 nm light. The RI values ranged from 1.537 (**1**) to 1.520 (**4**, Table S1 in the Supporting Information). The observed RI values are close to those of amorphous solids of distyrylbenzene derivatives with bulky substituents.[16] Interestingly, even if the trend in RI has only a small variation, it follows the trend of decreasing viscosity from **1** to **4**, and clearly shows the importance of the extent of OPV core density and isolation in tuning the viscosity and RI values. To understand how the core isolation affects the optoelectronic properties, transient photoconductivity experiments were carried out (Figure S8 in the Supporting Information).

Organic materials that emit white light are particularly important because durable materials with improved color purity and stability are in high demand for the development of large-area devices through low-cost manufacturing technology.[17] Except for a few examples,[18] in most cases of solution-[19] or gel-based[20] white-light emission, color stability and purity cannot be retained in solvent-free conditions, such as coated thin films or xerogels, whereas in the case of solid samples, processing becomes a tedious task. This situation demands suitable functional materials that have better emissive features in solvent-free conditions as well as facile and effortless processing characteristics. Liquid OPVs **1**–**4** can be used as solvents or matrices for other organic or inorganic components to obtain useful hybrid materials. For instance, 30 mg of green-emitting tris(8-hydroxyquinolinato)aluminium (Alq3) and 2 mg of orange-emitting 5,6,11,12-tetraphenylnaphthacene (rubrene) were dissolved in 100 mg of **2** by simply blending with a spatula. A larger amount of the solutes could be dissolved into the OPV matrix by heating or dissolving in a suitable solvent and then evaporating the solvent. This interesting finding motivated us to tune the emission properties of liquid OPVs to achieve white-emitting liquids. As indicated in Figure 3 a, pastes that emit white light were prepared by blending liquid OPVs with Alq3 and rubrene in a watch glass with a spatula. The supporting video file (see the Supporting Information) also clearly demonstrates the method adopted for the preparation of the pastes within a minute. We used **2** and **4** for preparing white-emitting composites, which are referred to as composite **I** and **II**, respectively. Blending **2** with Alq3 and rubrene in the molar ratio of 1:1.65:0.23 and annealing the sample at 40 °C for 2 min led to comparatively good white emission with a Commission Internationale de l'Éclairage (CIE) 1931 chromaticity diagram[21a] coordinate value of (0.33, 0.34) in the solvent-free state. A reasonably large spectral width that ranges from 400 to 700 nm, which matches the standard spectral width of white emission,[21] was observed (Figure 3 b). A possible advantage of our liquid composites is the reduced interface resistance between the components in the solvent-free liquid matrix, which presumably leads to partial energy transfer between the matrix host and the guest molecules to give pure white emission.[21b] The photoexcited carrier decay of OPV samples is very fast and the major component (ca. 80 %) has a lifetime

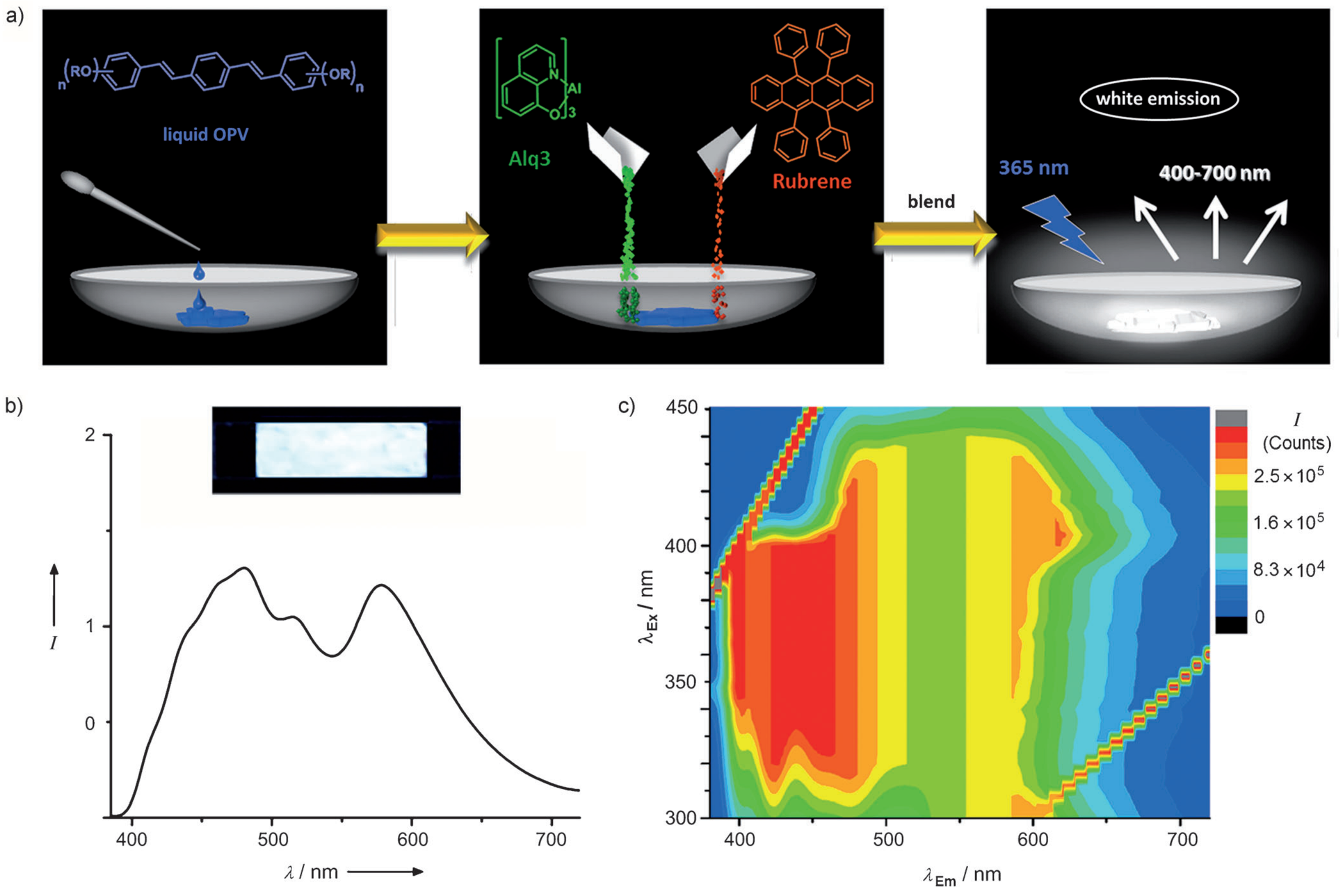

Figure 3. a) Illustration of the preparation of solvent-free white-emitting liquid composite. b) Emission spectrum of composite **I** ($\lambda_{ex} = 360$ nm) supported on quartz plate; inset shows the image of the white emission from composite **I**. c) Fluorescence matrix scan of composite **I**.

of 600 ps. The lifetime of the composite **I** was much shorter than that of composite **2** (Figure S9 in the Supporting Information). This result supports the possibility of energy transfer between OPV component and the dopants. The molar ratio between the three components was found to be significant in obtaining pure white emission (Figure S10 in the Supporting Information). Furthermore, annealing of the sample changes the interface characteristics towards improved brightness of the white emission. Fluorescence matrix scan measurements of composite **I**, which has a CIE coordinate value of (0.33, 0.35, $\lambda_{ex} = 360$ nm), exhibited broad emission from 400–700 nm (Figure 3c), and the emission can be tuned by using different excitation wavelengths (Figure S11 in the Supporting Information). An important feature of composites **I** and **II** is that white emission can be obtained across a wide range of excitation wavelengths from 350–430 nm (Figure 3c, see also Figure S12 in the Supporting Information). The achievement of white emission by low-energy excitation above 400 nm is energetically favored and can reduce the thermal energy loss. It will save electric power consumption and prolong device lifetime. Composites **I** and **II** have an absolute quantum yield of 35 % and 37 %, respectively. These values indicate that, unlike other materials that emit white light,[19,20] the quantum yield is not reduced to a large extent relative to the parent liquid samples. Composites **I** and **II** maintain their strongly emissive features, which is a good indication of the practical demonstration of composites **I** and **II** as white light emitting liquid materials.

DSC and rheology analyses of the white-light-emitting pastes that were prepared by blending the components (Figure S13 in the Supporting Information) clearly indicates that the fluidity is retained. Composites **I** and **II** have glass transition temperatures of −45.1 and −45.9 °C, respectively. Hence, there was no considerable change in T_g upon mixing the liquid OPVs with the solid dopants. Even in the presence of Alq3 and rubrene, the viscosity remains 3.2 Pa s in the case of composite **II**, and the material retains the Newtonian fluidity (Figure S13b,c in the Supporting Information). In order to compare with the solvent-free white emission, we carried out fluorescence studies of the OPVs in a solution of dichloromethane. These experiments revealed that a significantly different molar ratio is needed to achieve white emission in homogenous solution (Figure S14 in the Supporting Information). The different molar ratios that are required to produce white light emission in the homogenous solution versus the solvent-free state could be attributed to the difference in the (average) donor–acceptor distance[16] and the extent of direct excitation of the components. It should be noted that drop-casted films of the white-emitting dichloromethane solutions did not exhibit white emission, because a different molar ratio is needed in the solvent-free state (Figure S15i in the Supporting Information). This is probably a serious issue when samples are integrated into a device surface from volatile solutions with simultaneous solvent evaporation to get white-emitting thin films.[22] Control experiments with a solid composite of **6**, Alq3, and rubrene (Figure S15ii in the Supporting Information) as well as a composite of liquid **2** and Alq3 (Figure S15iii in the Supporting Information) revealed the advantage of the liquid-host matrix and three emissive components in the present system to obtain a feasible white-emitting liquid composite with color purity.

To efficiently address the advantage of the current system, as shown in Figure 4, we carried out experiments to tune the emission from blue-white (i) to pure white (ii) and further to red-white (iii). However, for practical demonstration of the liquid composite, it is of particular importance to have tunable white emission, such as blue-white (cool), pure white, and yellow-white (warm) colors, which can find applications according to requirements.[23] This was obtained from composite **I** when the molar ratio of the components (**2**/Alq3/rubrene) was slightly varied (Figure 4a–c). As a realistic demonstration, the lower viscosity value of the white-emitting liquid composite **II** enabled it to be used as ink for a roller-ball pen to write directly on various surfaces, which include paper and solid surfaces. The writing is then readable under a 365 nm UV lamp. Figure 4d shows an image of text written on paper with white-emitting ink. As another application, the composite material was used as a paint to coat large areas of various surfaces. As the viscosity of composite **II** is 3.2 Pa s, a paintbrush was suitable for coating the surfaces. Figure 4e shows an image of white emission from a 5 × 5 cm^2 area of the composite that was painted onto a phenol-formaldehyde-type black surface by using a brush. The applicability of the composites as a tractable material has been realized, as white-light emission was obtained by coating composite **II** onto a commercially available UV-LED (375 nm). As shown in

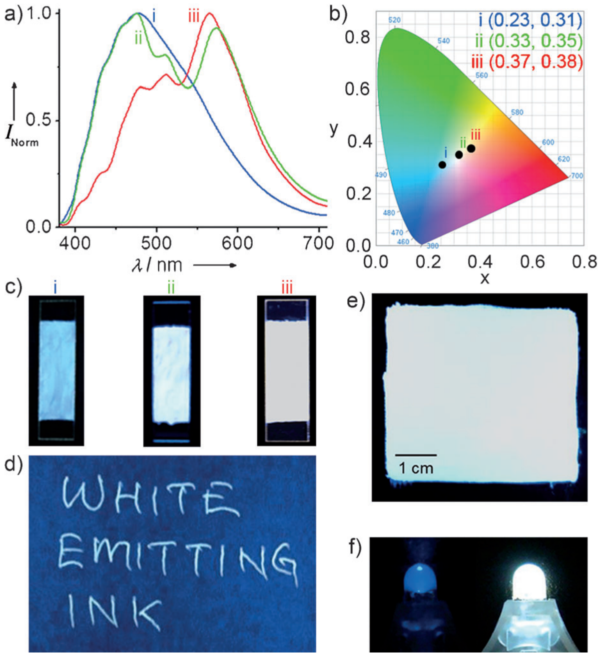

Figure 4. a) Normalized emission spectra of composite **I** with molar ratios of **2**/Alq3/rubrene of 1.0:1.50:0.05 (blue), 1.0:1.65: 0.23 (green), and 1.0:1.65:0.30 (red) supported on a quartz plate ($\lambda_{ex} = 360$ nm) with corresponding b) CIE coordinate values and c) images. Images of d) text written on paper with composite **II** in a rollerball pen, e) 5 × 5 cm^2 area coated with composite **II** using a brush and exposed under UV light (365 nm), and f) commercially available UV-LED (375 nm) before (left) and after (right) coating with composite **II**.

Figure 4 f, bright white emission was observed when a thin layer of composite **II** was applied onto the UV-LED. This experiment shows the feasibility of the composites as a cost effective white-emitting material. Hence, as well as being a white-emitting liquid, our composite liquids have solved a longstanding problem with solution-[19,22a] or gel-based[20,22b] white-emissive materials, that is, self-quenching in the dried, solvent-free state on substrate surfaces and in devices.

In conclusion, blue-emitting, environmental friendly, low-viscosity, room-temperature liquids that contain an OPV core have been synthesized and fully characterized as solvent-free functional organic liquids. This study demonstrates the ability of a highly dense but monomerically dispersed π-core system as an emissive liquid matrix to accommodate various guest dopants to tune the optical properties. The liquid matrix has been used to prepare liquid composites that emit white light by using a blue-emitting OPV liquid blended with green- and orange-emitting solid dopants under solvent-free conditions. The paintable composite can be applied to various surfaces over a large area to get white emission with desirable CIE coordinate values. Our demonstration introduces a clean strategy towards efficient organic liquids for advanced optoelectronic applications. Apart from the overtedious self-assembly process of organic semiconductors, our method offers plenty of room for designing the intrinsic characteristics and facile fabrication of emissive organic liquids. The polymerization of the liquid surface by mixing, UV light, or with thermally polymerizable monomers in combination with the composite to avoid stickiness and produce nonsticky films for white-emitting displays is under development in our laboratory. We believe that our luminescent organic liquids coupled with recent advances in printable electronic technology will definitely offer an alternative for the next generation organic devices, such as flexible, large area, white-emitting displays.

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[1] a) A. P. H. J. Schenning, E. W. Meijer, *Nature* **2002**, *419*, 353–354; b) S. R. Forrest, *Nature* **2004**, *428*, 911–918.
[2] F. J. M. Hoeben, P. Jonkheijm, E. W. Meijer, A. P. H. J. Schenning, *Chem. Rev.* **2005**, *105*, 1491–1546.
[3] T. Sekitani, T. Someya, *Adv. Mater.* **2010**, *22*, 2228–2246.
[4] a) R. F. Service, *Science* **2011**, *332*, 293; b) V. Shrotriya, *Nat. Photonics* **2009**, *3*, 447–449.
[5] J. Hachmann, R. Olivares-Amaya, S. Atahan-Evrenk, C. Amador-Bedolla, R. S. Sánchez-Carrera, A. Gold-Parker, L. Vogt, A. M. Brockway, A. Aspuru-Guzik, *J. Phys. Chem. Lett.* **2011**, *2*, 2241–2251.
[6] a) M. Armand, F. Endres, D. R. MacFarlane, H. Ohno, B. Scrosati, *Nat. Mater.* **2009**, *8*, 621–629; b) A. W. Perriman, A. P. S. Brogan, H. Cölfen, N. Tsoureas, G. R. Owen, S. Mann, *Nat. Chem.* **2010**, *2*, 622–626.
[7] S. C. Warren, M. J. Banholzer, L. S. Slaughter, E. P. Giannelis, F. J. DiSalvo, U. B. Wiesner, *J. Am. Chem. Soc.* **2006**, *128*, 12074–12075.
[8] a) M. Wathier, M. W. Grinstaff, *J. Am. Chem. Soc.* **2008**, *130*, 9648–9649; b) K. Y. Yung, A. J. Schadock-Hewitt, N. P. Hunter, F. V. Bright, G. A. Baker, *Chem. Commun.* **2011**, *47*, 4775–4777.
[9] a) H. Akamatu, H. Inokuchi, Y. Matsunaga, *Nature* **1954**, *173*, 168–169; b) S. S. Babu, H. Möhwald, T. Nakanishi, *Chem. Soc. Rev.* **2010**, *39*, 4021–4035.
[10] E. M. Maya, J. S. Shirk, A. W. Snow, G. L. Roberts, *Chem. Commun.* **2001**, 615–616.
[11] a) S. Maruyama, K. Sato, H. Iwahashi, *Chem. Lett.* **2010**, *39*, 714–716; b) A. Nowak-Król, D. Gryko, D. T. Gryko, *Chem. Asian J.* **2010**, *5*, 904–909.
[12] S. Hirata, K. Kubota, H. H. Jung, O. Hirata, K. Goushi, M. Yahiro, C. Adachi, *Adv. Mater.* **2011**, *23*, 889–893.
[13] T. Michinobu, T. Nakanishi, J. P. Hill, M. Funahashi, K. Ariga, *J. Am. Chem. Soc.* **2006**, *128*, 10384–10385.
[14] a) P. Jonkheijm, P. van der Schoot, A. P. H. J. Schenning, E. W. Meijer, *Science* **2006**, *313*, 80–83; b) S. S. Babu, K. K. Kartha, A. Ajayaghosh, *J. Phys. Chem. Lett.* **2010**, *1*, 3413–3424; c) S. Yagai, S. Kubota, T. Iwashima, K. Kishikawa, T. Nakanishi, T. Karatsu, A. Kitamura, *Chem. Eur. J.* **2008**, *14*, 5246–5257; d) D. Oelkrug, A. Tompert, J. Gierschner, H.-J. Egelhaaf, M. Hanack, M. Hohloch, E. Steinhuber, *J. Phys. Chem. B* **1998**, *102*, 1902–1907.
[15] a) J. C. Sancho-García, J. L. Brédas, D. Beljonne, J. Cornil, R. Martínez Álvarez, M. Hanack, L. Poulsen, J. Gierschner, H.-G. Mack, H.-J. Egelhaaf, D. Oelkrug, *J. Phys. Chem. B* **2005**, *109*, 4872–4880; b) J. Gierschner, H.-G. Mack, L. Lüer, D. Oelkrug, *J. Chem. Phys.* **2002**, *116*, 8596–8609.
[16] H.-J. Egelhaaf, J. Gierschner, D. Oelkrug, *Synth. Met.* **2002**, *127*, 221–227.
[17] K. T. Kamtekar, A. P. Monkman, M. R. Bryce, *Adv. Mater.* **2010**, *22*, 572–582.
[18] S. Park, J. E. Kwon, S. H. Kim, J. Seo, K. W. Chung, S.-Y. Park, D.-J. Jang, B. M. Medina, J. Gierschner, S. Y. Park, *J. Am. Chem. Soc.* **2009**, *131*, 14043–14049.
[19] X. Zhang, S. Rehm, M. M. Safont-Sempere, F. Würthner, *Nat. Chem.* **2009**, *1*, 623–629.
[20] C. Giansante, G. Raffy, C. Schäfer, H. Rahma, M.-T. Kao, A. G. L. Olive, A. D. Guerzo, *J. Am. Chem. Soc.* **2011**, *133*, 316–325.
[21] a) M. C. Gather, A. Köhnen, K. Meerholz, *Adv. Mater.* **2011**, *23*, 233–248; b) S. Sepeai, M. M. Salleh, M. Yahaya, A. A. Umar, *Thin Solid Films* **2009**, *517*, 4679–4683.
[22] R. Abbel, R. van der Weegen, W. Pisula, M. Surin, P. Leclère, R. Lazzaroni, E. W. Meijer, A. P. H. J. Schenning, *Chem. Eur. J.* **2009**, *15*, 9737–9746.
[23] T. Kitabatake, T. Uchikoshi, F. Munakata, Y. Sakka, N. Hirosaki, *J. Ceram. Soc. Jpn.* **2010**, *118*, 1–4.