White-Light Emission

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Bright White-Light Emission from a Single Organic Compound in the Solid State**

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Dedicated to Helmut Schwarz on the occasion of his 70th birthday

Abstract: *White-light-emitting materials and devices have attracted enormous interest because of their great potential for various lighting applications. We herein describe the light-emitting properties of a series of new difunctional organic molecules of remarkably simple structure consisting of two terminal 4-pyridone push–pull subunits separated by a polymethylene chain. They were found to emit almost "pure" white light as a single organic compound in the solid state, as well as when incorporated in a polymer film. To the best of our knowledge, they are the simplest white-light-emitting organic molecules reported to date.*

Despite an active search for white-light-emitting materials and devices,[1] most of the materials reported so far rely on the combination of different emitters (red/blue/green or blue/orange) to cover the visible range from 400 to 700 nm, which may cause problems such as color aging or different requirements for device fabrication.[2] Compared to the multicomponent white-light sources, direct emission of white light from a single-component entity may be expected to avoid the drawbacks of combined emitters, thus giving rise to white light of higher quality.[3,4] Therefore, the search for new molecules that can directly emit white light is clearly of interest and importance. In molecular species, single-component white-light emission has been generated from two families of compounds: a) metal complexes[3] and b) purely organic molecules.[4] White-light-emitting single-organic molecules are scarce and, to date, only a few have been reported. Such is the case for a system composed of benzo[*a*]xanthene and benzo[*b*]xanthene derivatives,[4a] for organic molecules containing complementary emitting units between which energy transfer is blocked because of excited-state intramolecular proton transfer,[4b,c] for an organic molecule based on π-conjugated carbazole-substituted phenylenynes,[4d] for an organic dye that displays panchromatic emission,[4e] and for electroluminescent white-light-emitting organic molecules.[4f] However, these molecules have complicated structures and require intricate synthetic strategies, features that may hinder their practical use. Thus, achieving white-light emission from small molecules with simple structures is an exciting challenge in synthetic chemistry as well as in photochemistry and photophysics.

Herein, we report the light-emission properties of new difunctional organic molecules of simple structure and easy access that are based on two terminal push–pull moieties separated by a polymethylene chain: the molecules **D1a**–**D1e** containing two 4-pyridone units and the compounds **D2**–**D4** bearing different but related head groups, which were studied for comparison purposes. The corresponding monofunctional molecules **M1**–**M4** were also examined as reference compounds (Scheme 1).

The mono- and difunctional compounds **M1** and **D1** were synthesized by direct alkylation at the nitrogen site of 4-hydroxypyridine with the corresponding halo- or α,ω-dihaloalkanes through a slight modification of a known reaction.[5] Compounds **M2** and **D2** were prepared from 4-(4-hydroxyphenyl)pyridine[6] through a slight modification of a known reaction.[7] Compounds **M3**, **M4**, **D3**, and **D4** were obtained according to a modified literature procedure.[8] Alkylation of 4-hydroxyquinoline, 9(10*H*)-acridone, and 4-(4-hydroxyphenyl)pyridine gave the *N*-substituted products directly. However, alkylation of 4-hydroxypyridine can produce either *N*-substituted pyridones or *O*-substituted pyridines depending on the specific alkylating agent and conditions.[9–11] In the present study, the alkylation of 4-hydroxypyridine with α,ω-dibromoalkanes produced a mixture of N- and O-alkylation products. The symmetrical di-*N*-substituted pyridone is the dominant product. The unsymmetrical by-product, where the central chain is connected to the nitrogen site of one terminal heterocyclic group and to the oxygen atom of the other one, can be easily separated. After purification, the purity of the symmetrical di-*N*-substituted pyridone can be increased to 99.8% (as judged by integration of the ^{1}H NMR spectrum). Detailed synthetic, isolation, and purification procedures for all the compounds are described in the Supporting Information.

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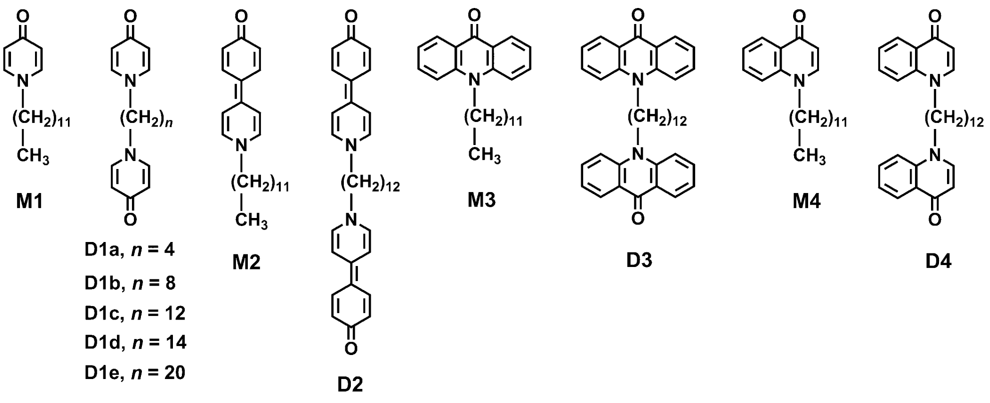

Scheme 1. Structures of the monofunctional **M** and difunctional **D** compounds investigated.

The absorption and emission spectra of the monosubstituted **M1**–**M4** (see Figure S1 in the Supporting Information) and those of the disubstituted **D1**–**D4** compounds (see Figure 1 for **D1c** and **D2**; see Figure S2 in the Supporting Information for **D3** and **D4**) in dilute solution are similar. **D1a**–**D1e** have the same head group and differ only by the length of the alkane chain. After an initial exploration of the photophysical properties, **D1c** was chosen as the representative molecule, and the same dodecyl alkane chain was used in **D2**–**D4**. The absorption spectra of all compounds **M1**–**M4** and **D1**–**D4** display intense bands at wavelengths below 500 nm. These absorptions are assigned to intraligand $^1\pi$-π^* transitions of the molecules, as confirmed by DFT computational studies (see below). Compounds **M1**–**M4** and **D1**–**D4** all emit blue light in dilute methanol.

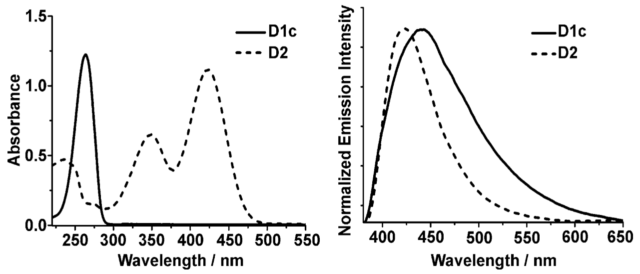

Figure 1. Absorption (left) and emission (right) spectra of solutions (5×10^{-5} M in methanol) of the disubstituted compounds **D1c** (excited at 280 nm) and **D2** (excited at 339 nm).

In the solid state, in contrast to the solutions, the monosubstituted **M1** and disubstituted **D1** compounds display remarkably different room-temperature photoluminescence (PL) spectra, as shown in Figure 2 for **M1** and **D1c** (see Figure S3 in the Supporting Information for the PL spectra of **M3, D3**, **M4**, and **D4**). Whereas **M1** emits blue light and shows emission bands centered around 450 nm, **D1** emits bluish-white to white light. Most remarkably, **D1c** displays a strong white-light emission, which corresponds to a very broad band that covers the whole visible range extending from 400 to 750 nm as well as a band at 560 nm and a shoulder at 445 nm (a deconvolution of the two bands is shown in Figure S4 in the Supporting Information). Similar luminescence properties are shown by the other **D1** compounds, **D1a**, **D1b**, **D1d**, and **D1e** (see Figure S5 in the Supporting Information). The shoulder at short wavelength is reminiscent of the emission band from the monofunctional **M1** and may thus be assigned to emission from the terminal groups of the difunctional **D1c**. One may hypothesize that the high-energy emissions are due to intrachromophore π-π* transitions and that the low-energy ones result from excited stacked groups covering a range from intermolecular excimers to excited oligomers (see also the crystal structure, Figure 6; for simplicity, when the term excimer is used for our data, it is meant to describe the actual white-light-emitting entity).[12–15]

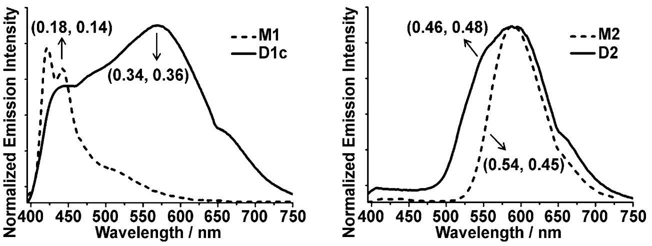

Figure 2. Emission spectra of molecules **M1**, **D1c**, **M2**, and **D2** in the solid state with CIE coordinates under excitation at 365 nm.

Further information to this end, was provided by the emission spectra of **M1** and **D1c** at different concentrations (see Figures S6 and S7 in the Supporting Information). In both cases, a new emission band appeared at 480 nm as the concentration was increased. This band may be assigned to an excimer emission in solution and not involving the excited oligomers formed in the solid state.

To probe the origin of the long-wavelength emissions, the photoluminescence excitation spectra of **D1c** were measured in the solid state for emissions at 445 and 560 nm (see Figure S8 in the Supporting Information). The emissions at the two wavelengths are similar, thus suggesting that they should have a common excitation pathway, and excluding the possibility of the formation of a new fluorophore. Accordingly, the long-wavelength emissions of **D1c** should result from excimers. The monosubstituted molecule **M1** also shows an excimer emission at 523 nm, but its intensity is much weaker and narrower, thereby resulting in the emission of blue light instead of white light. This may be due to a different packing of the pyridone moieties in **M1** compared to **D1c** (see below).

Time-dependent profiles of the monomer and excimer emission of **D1c** and **D2** were measured in the solid state. The lifetime of the monomer emission (445 nm) of **D1c** reveals a biexponential decay feature with $\tau = 7.8$ and 20.9 ns. Such lifetimes are characteristic of ligand-centered (LC) emissions and may originate from π-π* transitions as well as intraligand charge transfer (ILCT). The time evolution of the excimer emission (560 nm) shows a different biexponential decay, with longer lifetimes of $\tau = 46.1$ and 103.4 ns. Similar results have been obtained for the decay of the monomer and excimer emission of **D2**, with the biexponential decays having lifetimes of $\tau = 2.4$ and 14.6 ns and of $\tau = 19.6$ and 63.8 ns, respectively.

Compounds **M2** and **D2** both show in the solid state a broad band centered around 590 nm, in the region of the long-wavelength region of the emission of **D1c**. In addition, **D2** also gives a weak emission at short wavelength (410 nm). As in the case of **D1c**, the latter high-energy emission of **D2** may be assigned to intrachromophore π-π* transitions, whereas the low-energy band (590 nm) would again result from excimer emission in both compounds. The fact that compound **M2** already displays strong excimer emission may be attributed to the larger size of the chromophoric terminal group, as the more extended push–pull head group allows for more efficient stacking, thus resulting in excimer emission even as a monofunctional entity. In contrast, **M3**, **M4**, **D3**, and **D4** do not show excimer emission in the solid state (see Figure S3 in the Supporting Information). So one may speculate that the difference in the relative disposition of the molecules in the solid state resulting from the different shapes of the head groups (and to a lesser extent from their different polarity) have a large impact on the photophysical properties. Thus, **D1** and **D2**, which contain the elongated, prolate push–pull moieties (pyridone and pyridinylidenecyclohexa-2,5-dienone, respectively), might pack in the solid state so as to give excimer emission, whereas **D3** and **D4**, which contain larger and laterally extended oblate terminal groups (quinolinone and acridinone), are less prone to give an arrangement suitable for excimer emission (see Figure S9 in the Supporting Information). The more commensurate size between the head groups and the hydrocarbon spacer chain may allow for better stacking in the cases of **D1** and **D2** than in the cases of **D3** and **D4**.

The striking difference between **D1c** and **D2** is that the emission spectrum of the former extends over the whole range of wavelengths, whereas that of the latter presents very weak emission between 400 and 500 nm (Figure 2).

To further investigate the formation of excimers in the elongated push–pull molecules, the emission spectra of **D1c** and **D2** were also measured in polymer films containing increasing amounts of the two compounds. These films were prepared by spin-coating a mixture of solutions of **D1c** (or **D2**) and polymethylmethacrylate (PMMA) in dichloromethane. In the case of **D1c**, the film formed at a low dopant concentration of 5 wt% at 298 K exhibited the blue light emission of isolated **D1c** molecules (Figure 3), similar to the emission in solution (Figure 1). Increasing the concentration in the range 5–58 wt% resulted in a low-energy structureless emission developing at about 550 nm, thereby finally yielding a spectrum similar to that of **D1c** in the solid state at high concentration (Figure 3; see also the chromaticity coordinates in Figure S10 and the excitation spectra in Figure S11 in the Supporting Information). The observation of such a concentration-dependent emission may be attributed to excimer emission caused by denser packing of molecules at increased concentrations, as in the crystal lattice of **D1c** (see below). To gain more information about the origin of the emission, a film containing 50% **D1c** dopant was investigated by scanning electron microscopy (SEM). The images obtained showed the presence of microcrystalline inclusions (see Figure S12 in the Supporting Information), thus indicating that the emission was due to solid-state material dispersed in the film.

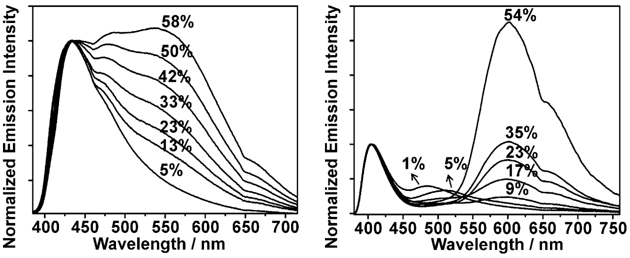

Figure 3. Emission spectra of **D1c** (left) and **D2** (right) at increasing concentrations in PMMA films under excitation at 365 nm. The percentages of **D1c** (from 5% to 58%) and **D2** (from 1% to 54%) (by weight) in PMMA increase from the bottom to the top emission curves (as indicated). The CIE values for the emission of **D1c** are, for example, (0.28, 0.33) at 50% and (0.30, 0.34) at 58% dopant (see Figure S10 in the Supporting Information) and for **D2** emission (0.51, 0.39) at 54% dopant. For photographs of the white-light-emitting films see Figure S14 in the Supporting Information.

Similarly, in the case of **D2**, as the concentration increases from 1% to 54 wt%, in addition to the weak emission observed at 410 nm, a much more intense low-energy structureless emission develops at about 600 nm (Figure 3). Again such a concentration-dependent emission likely arises from intermolecular excimer emission.[12–14] The fact that compound **M2** itself displays excimer emission may be due to the fact that the more-extended push–pull head group allows for more efficient stacking, thereby resulting in excimer emission even as a monofunctional entity.

In a further experiment, a sample of **D1c** was melted (107 °C) and its emission spectra were recorded as it cooled down to room temperature (25 °C; Figure 4). In the molten state, **D1c** showed strong high-energy emission at 445 nm, assigned to intramolecular π-π* transitions, with very weak low-energy excimer emission. In the melt, the polar pyridone units are in a disordered state and cannot form excimers effectively, thus emitting blue light. The intensity of the excimer emission developed progressively on cooling from 107 °C to 25 °C over 0–45 min, until the solidified sample again produced bright white light. One may expect that on the way to the solid state, the arrangement of the chromophores changes from nondirectional to directional alignment, becoming more and more organized, so that the pyridone units can form excimers effectively. The excitation spectra of **D1c** in the molten state for emissions at 445 and 560 nm are similar (see Figure S13 in the Supporting Information).

The other four **D1** molecules with the same head group as **D1c** present similar white-light emission in the solid state. **D1a**, **D1b**, and **D1e** emit bluish white light with CIE

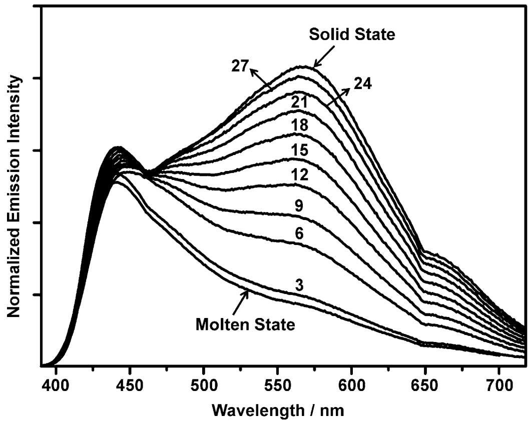

Figure 4. Emission spectra of **D1c** on cooling from the molten state (107 °C) to the solid state (25 °C) under excitation at 365 nm. Emission curves from the bottom to the top were measured at the various times (indicated in minutes) on progressive cooling from the melt.

coordinates of (0.28, 0.33), (0.28, 0.35), and (0.28, 0.37), respectively. **D1c** and **D1d** emit white light with CIE coordinates of (0.34, 0.36) and (0.33, 0.34), respectively, very close to pure white light (CIE: 0.33, 0.33). The five molecules **D1a**–**D1e** differ by the length of the alkyl chain connecting the two *N*-substituted pyridones, which may lead to different packing arrangements[16] and result in slightly different arrangements of the polar pyridone chromophores, thus producing slightly different white-light emissions. They give high emission quantum yields ranging from 30 % to 33 % (see Table S2 in the Supporting Information). The emission properties of the molecules **D1c** and **D2** under excitation at 365 nm are illustrated in Figure 5, together with the corresponding 1931 Commission Internationale de L'Eclairage (CIE) diagrams. Importantly, excitation at different wavelengths (350–420 nm) gives the same white-light emission. Thus, emission of bright-white light from a single organic molecule can be achieved under irradiation in the near-UV region and into the visible range. Notably and of potential practical interest, the emission from the PMMA film at 58 % **D1c** doping (Figure 3) is even flatter over the wide 425–600 nm range than that of the solid **D1c** itself, with a CIE coefficient of (0.30, 0.34).

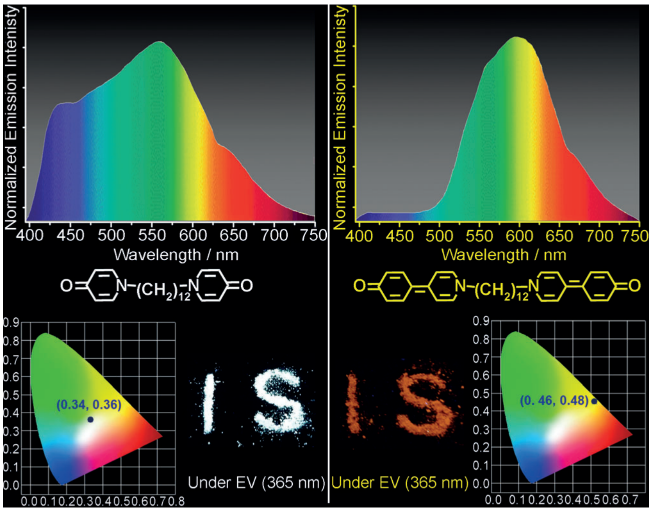

Figure 5. Top: Solid-state photoluminescent spectra of **D1c** and **D2** under excitation at 365 nm; bottom: chromaticity coordinates (CIE) and photographs of the name of our Institute ISIS written with the white-light-emitting **D1c** (left; CIE: 0.34, 0.36) and the orange-light emitting **D2** (right; CIE: 0.46, 0.48) molecules. The optical photographs were taken under irradiation at 365 nm. The emission curves have been superimposed on the spectrum of white light.

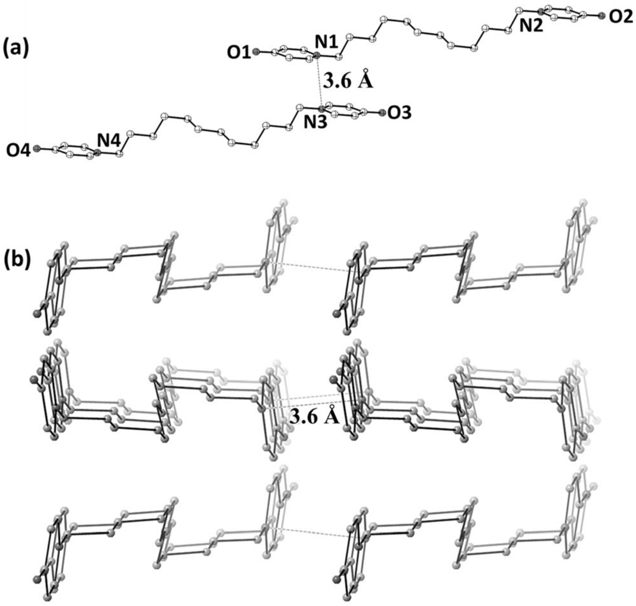

Figure 6. X-ray solid-state molecular structure of the white-light-emitting molecule **D1c**: a) illustration of the relative disposition and intermolecular interactions of two molecules. b) side view showing the packing and the interaction between adjacent units in each stack of molecules. The solvent molecules are omitted for clarity.

To relate the optical properties to the molecular arrangement in the solid state, more complete structural data are desirable. Fortunately, single crystals of the white-light-emitting molecule **D1c** could be grown on cooling a hot aqueous solution (see the Supporting Information). Single-crystal X-ray structural analysis confirmed the alkylation of 4-hydroxypyridine to give a symmetrical di-*N*-substituted pyridone molecule. The asymmetric unit contains half a centrosymmetric **D1c** molecule and one molecule of water. In the crystal lattice, the ligands are packed through intermolecular stacking between two planar pyridones as well as hydrogen bonding between the carbonyl oxygen atom and the water molecules. Pairs of planar pyridones are in contact, separated by about 3.6 Å, and in a shifted head-to-tail geometry (Figure 6). Most importantly, the nitrogen sites, which represent the positive end of the electric dipole (about 7.9 D) of the polar 4-pyridone groups, are located on top of each other and, therefore, interact in a repulsive manner. We recall that an excimer is formed by the stabilization of a dimer structure in the excited state at a geometry for which the intermolecular interactions are repulsive in the ground state.[13,17] Emission thus occurs from the minimum of the excited state to a repulsive region of the ground state. Therefore, the energy change is smaller than in the isolated chromophore, and the emission from the excimer is red-shifted (see Figure S15 in the Supporting Information). The special intermolecular positioning of the pyridone groups in the solid state of **D1c**, together with the excited oligomer character enhance these features and may be considered as the major factors for the broad white-light emission. The slight differences in the emission properties along the **D1a**–**D1e** series may result from small changes in the arrangement through a contribution from the packing of the hydrocarbon chains to generate the final disposition. The shortest distance between two hydrocarbon chains is 3.96 Å (see also Figure S16 in the Supporting Information).

DFT/TDDFT calculations were performed on the white-light-emitting molecule **D1c** as well as on **D2** to gain insight into the optical features of the excimer formation. To avoid the complexity of periodic structures, **D1c** was truncated into the 4-pyridone fragment from the crystal structure for the computation. The ground-state geometries were optimized for both molecules and their Frontier Molecular Orbitals (FMOs) are displayed in Figure 7. In addition, similar computational studies were performed on the representative fragments of **D3** and **D4** (see Figure S17 in the Supporting Information). The calculations also gave the dipole moments of the chromophoric groups (see Scheme S1 in the Supporting Information).

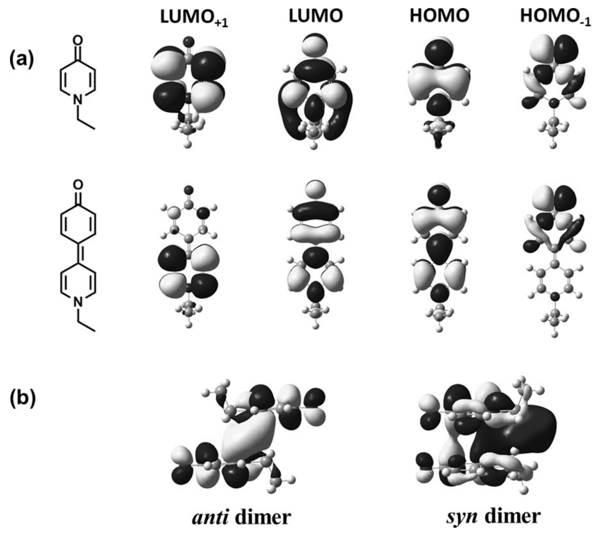

Figure 7. a) Frontier molecular orbitals of **D1c** and **D2** in the electronic ground state. b) LUMO orbitals for the *anti* (head-to-tail, as in the crystal structure) and *syn* arrangements of dimers of two 4-pyridone moieties in **D1c** at an interplanar distance of 3.6 Å.

As shown in Figure 7a, the electron distribution in the HOMO$_{-1}$ and the LUMO$_{+1}$ in compounds **D1c** and **D2** localizes at the side of the oxygen atom and of the nitrogen atom, respectively. This electron transfer from nitrogen to oxygen represents the electron-donor/electron-acceptor distribution in the conjugated N-C-C-O fragment. The photophysical properties and energies of the FMOs of **D1c** and **D2** are collected in Table S3 in the Supporting Information. The theoretical values for the $S_0 \rightarrow S_1$ ($^1\pi \rightarrow \pi^*$) transitions compare well with the experimental absorption bands.

Excimer formation in **D1c** was investigated by exploring several relative dispositions of two 4-pyridone groups, keeping their geometry frozen. Several dimer structures were obtained by rotating one of the molecules along the N-N axis or by changing the interplanar distance. Among those, Figure 7b displays the LUMO energies of the *anti* dimer and *syn* dimer at an interplanar distance of 3.6 Å. In the dimer, these LUMO energies have been split, but the HOMOs are little affected. Moreover, the *anti* dimer is calculated to be more stable than the *syn* dimer by 11.5 kcal mol^{-1}, which corresponds to the head-to-tail orientation found in the crystal structure of **D1c** (Figure S18 in the Supporting Information depicts the effect of the intermolecular interactions on the FMO energies of the *anti* dimer as a function of the distance between the two units). At a moderate spacing (4.0 Å), the interaction between the LUMOs of the pyridone groups is already substantial and the energy gap between HOMO and LUMO ($\Delta E_{\text{(LUMO-HOMO)}}$) is reduced. At shorter intermolecular separations, this interaction increases and the $\Delta E_{\text{(LUMO-HOMO)}}$ value becomes smaller, so that the excimer formation observed in the *anti* dimer leads to emission at longer wavelengths, as observed. These theoretical results are thus in good agreement with the experimental data. Excited states extending over more than two stacked groups may contribute to emission at longer wavelengths than that of the excimer.[12]

In conclusion, we synthesized and investigated a series of difunctional molecules bearing polar moieties at the ends of polymethylene chains. Their photophysical properties in the solid state depend on the nature of the head groups, which determine their emission properties. The introduction of 4-pyridone moieties as head groups gave a novel class of particularly simple difunctional organic molecules [pyridone]-$(CH_2)_n$-[pyridone] (**D1**) displaying white-light emission with high quantum yields in the solid state. The striking feature results from the remarkable simultaneous occurrence of emission of the isolated molecule type at short wavelengths and very broad emission from excited oligomers at longer wavelengths, with relative emission intensities such as to yield a plateau-type band covering a wide domain in the visible range and thus generating bright and almost pure white light. Such emission from structurally simple organic molecules is remarkable and promises potential implementation in future applications. Of particular significance in this respect is the possibility to prepare white-light-emitting polymeric films. Finally, one may note that these properties are, in effect, of collective supramolecular nature, as they result from the special intermolecular positioning of the molecules and their mutual interaction.

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[1] a) G. M. Farinola, R. Ragni, *Chem. Soc. Rev.* **2011**, *40*, 3467–3482; b) B. W. D'Andrade, S. R. Forrest, *Adv. Mater.* **2004**, *16*, 1585–1595; c) C. Ulbricht, B. Beyer, C. Friebe, A. Winter, U. S. Schubert, *Adv. Mater.* **2009**, *21*, 4418–4441; d) S. C. F. Kui, P. K. Chow, G. S. M. Tong, S. L. Lai, G. Cheng, C. C. Kwok, K. H. Low, M. Y. Ko, C. M. Che, *Chem. Eur. J.* **2013**, *19*, 69–73; e) H. C. Su, H. F. Chen, F. C. Fang, C. C. Liu, C. C. Wu, K. T. Wong, Y. H. Liu, S. M. Peng, *J. Am. Chem. Soc.* **2008**, *130*, 3413–3419; f) B. H. Zhang, G. P. Tan, C. S. Lam, B. Yao, C. L. Ho, L. H. Liu, Z. Y. Xie, W. Y. Wong, J. Q. Ding, L. X. Wang, *Adv. Mater.* **2012**, *24*, 1873–1877; g) V. K. Praveen, C. Ranjith, N. Armaroli, *Angew. Chem.* **2014**, *126*, 373–376; *Angew. Chem. Int. Ed.* **2014**, *53*, 365–368; h) B. D'Andrade, M. E. Thompson, S. R. Forrest, *Adv. Mater.* **2002**, *14*, 147–151.

[2] a) G. L. Tu, C. Y. Mei, Q. G. Zhou, Y. X. Cheng, Y. H. Geng, L. X. Wang, D. G. Ma, X. B. Jing, F. S. Wang, *Adv. Funct. Mater.* **2006**, *16*, 101–106; b) B. P. Yan, C. C. C. Cheung, S. C. F. Kui, H. F. Xiang, V. A. L. Roy, S. J. Xu, C. M. Che, *Adv. Mater.* **2007**, *19*, 3599–3603; c) B. W. D'Andrade, R. J. Holmes, S. R. Forrest, *Adv. Mater.* **2004**, *16*, 624–628.

[3] a) P. Coppo, M. Duati, V. N. Kozhevnikov, J. W. Hofstraat, L. De Cola, *Angew. Chem.* **2005**, *117*, 1840–1844; *Angew. Chem. Int. Ed.* **2005**, *44*, 1806–1810; b) A. H. Shelton, I. V. Sazanovich, J. A. Weinstein, M. D. Ward, *Chem. Commun.* **2012**, *48*, 2749–2751; c) H. J. Bolink, F. De Angelis, E. Baranoff, C. Klein, S. Fantacci, E. Coronado, M. Sessolo, K. Kalyanasundaram, M. Gratzel, M. K. Nazeeruddin, *Chem. Commun.* **2009**, 4672–4674; d) D. Sykes, I. S. Tidmarsh, A. Barbieri, I. V. Sazanovich, J. A. Weinstein, M. D. Ward, *Inorg. Chem.* **2011**, *50*, 11323–11339; e) G. L. Law, K. L. Wong, H. L. Tam, K. W. Cheah, W. T. Wong, *Inorg. Chem.* **2009**, *48*, 10492–10494; f) G. J. He, D. Guo, C. He, X. L. Zhang, X. W. Zhao, C. Y. Duan, *Angew. Chem.* **2009**, *121*, 6248–6251; *Angew. Chem. Int. Ed.* **2009**, *48*, 6132–6135; g) V. Adamovich, J. Brooks, A. Tamayo, A. M. Alexander, P. I. Djurovich, B. W. D'Andrade, C. Adachi, S. R. Forrest, M. E. Thompson, *New J. Chem.* **2002**, *26*, 1171–1178.

[4] a) Y. Yang, M. Lowry, C. M. Schowalter, S. O. Fakayode, J. O. Escobedo, X. Xu, H. Zhang, T. J. Jensen, F. R. Fronczek, I. M. Warner, R. M. Strongin, *J. Am. Chem. Soc.* **2006**, *128*, 14081–14092; b) S. Park, J. E. Kwon, S. H. Kim, J. Seo, K. Chung, S. Y. Park, D. J. Jang, B. M. Medina, J. Gierschner, S. Y. Park, *J. Am. Chem. Soc.* **2009**, *131*, 14043–14049; c) K. C. Tang, M. J. Chang, T. Y. Lin, H. A. Pan, T. C. Fang, K. Y. Chen, W. Y. Hung, Y. H. Hsu, P. T. Chou, *J. Am. Chem. Soc.* **2011**, *133*, 17738–17745; d) Y. Liu, M. Nishiura, Y. Wang, Z. M. Hou, *J. Am. Chem. Soc.* **2006**, *128*, 5592–5593; e) P. Nandhikonda, D. Heagy, *Chem. Commun.* **2010**, *46*, 8002–8004; f) J. Y. Li, D. Liu, C. Ma, O. Lengyel, C. S. Lee, C. H. Tung, S. T. Lee, *Adv. Mater.* **2004**, *16*, 1538–1541; g) for white-light emission from a two-component micellar system, see X. Zhang, D. Görl, F. Würthner, *Chem. Commun.* **2013**, *49*, 8178–8180.

[5] S. K. Freeman, W. F. Ringk, P. E. Spoerri, *J. Am. Chem. Soc.* **1947**, *69*, 858–859.

[6] A. Das, J. C. Jeffery, J. P. Maher, J. A. McCleverty, E. Schatz, M. D. Ward, G. Wollermann, *Angew. Chem.* **1992**, *104*, 1554–1556; *Angew. Chem. Int. Ed. Engl.* **1992**, *31*, 1515–1518.

[7] V. Diemer, H. Chaumeil, A. Defoin, P. Jacquesb, C. Carre, *Tetrahedron Lett.* **2005**, *46*, 4737–4740.

[8] R. M. Acheson, E. C. Constable, *J. Chem. Soc. Chem. Commun.* **1980**, 1065–1066.

[9] L. Johnson, Y. Kitahara, T. J. R. Weakley, J. F. W. Keana, *Tetrahedron Lett.* **1993**, *34*, 5555–5558.

[10] H. Vorbrüggen, K. Krolikiewicz, *Chem. Ber.* **1984**, *117*, 1523–1541.

[11] A molecule containing three 4-hydroxypyridine/4-pyridone groups was prepared in the course of an exploration of potential multivalent ligands for luminescent rare-earth metal cations, see Q. Y. Yang, K. Li, J. Luo, M. Pan, C. Y. Su, *Chem. Commun.* **2011**, *47*, 4234–4236.

[12] For a study of emission from an excited oligomer of stacked anthracene molecules in the solid state, see T. Hinoue, Y. Shigenoi, M. Sugino, Y. Mizobe, I. Hisaki, M. Miyata, N. Tohnai, *Chem. Eur. J.* **2012**, *18*, 4634–4643.

[13] For a presentation of excimers, see B. Valeur, *Molecular Fluorescence: Principles and Applications*, Wiley-VCH, Weinheim, **2002**, pp. 94–98.

[14] For studies on excimers, see a) A. D. Q. Li, W. Wang, L. Q. Wang, *Chem. Eur. J.* **2003**, *9*, 4594–4601; b) H. Yoo, J. Yang, A. Yousef, M. R. Wasielewski, D. Kim, *J. Am. Chem. Soc.* **2010**, *132*, 3939–3944; c) J. M. Giaimo, J. V. Lockard, L. E. Sinks, A. M. Scott, T. M. Wilson, M. R. Wasielewski, *J. Phys. Chem. A* **2008**, *112*, 2322–2330; d) F. Würthner, C. Thalacker, S. Diele, C. Tschierske, *Chem. Eur. J.* **2001**, *7*, 2245–2253.

[15] For intramolecular excimers, see also a) K. Zachariasse, W. Kühnle, *Z. Phys. Chem. Neue Folge* **1976**, *101*, 267; b) M. A. Winnik, *Chem. Rev.* **1981**, *81*, 491–524; c) F. C. De Schryver, P. Collart, J. Vandendriessche, R. Goedeweeck, A. Swinnen, M. Van der Auweraer, *Acc. Chem. Res.* **1987**, *20*, 159–166; d) F. M. Winnik, *Chem. Rev.* **1993**, *93*, 587–614.

[16] A. Neelakantan, R. Stine, J. K. Maranas, *Macromolecules* **2003**, *36*, 3721–3731.

[17] D. Kim, J. L. Bredas, *J. Am. Chem. Soc.* **2009**, *131*, 11371–11380.