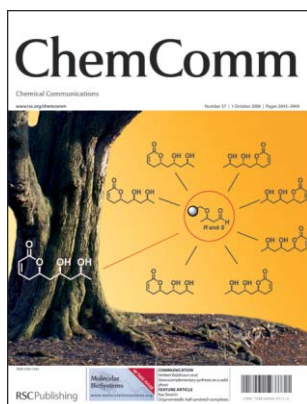


## IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (37) 3845-3944 (2006)



### Cover

See Herbert Waldmann *et al.*, page 3868. Enantioselective allylations applied to two resin-bound aldehydes were the basis for the synthesis of a diastereomer library derived from the natural product cryptocarya diacetate. Image reproduced by permission of Ana B. Garcia, Torben Leßmann, Jayant D. Umarye, Victor Mamane, Stefan Sommer and Herbert Waldmann, *Chem. Commun.*, 2006, 3868.

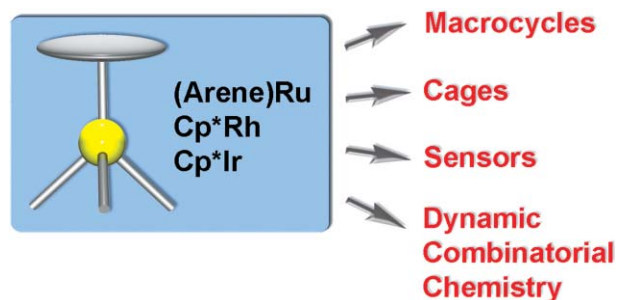
## FEATURE ARTICLE

3859

### Supramolecular chemistry with organometallic half-sandwich complexes

Kay Severin

Organometallic half-sandwich complexes can be employed for the construction of metallamacrocycles and coordination cages, to study the adaptive behavior of dynamic combinatorial libraries and to generate indicator displacement assays for the detection of biologically interesting analytes.



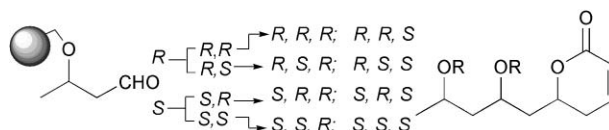
## COMMUNICATIONS

3868

### Stereocomplementary synthesis of a natural product-derived compound collection on a solid phase

Ana B. Garcia, Torben Leßmann, Jayant D. Umarye, Victor Mamane, Stefan Sommer and Herbert Waldmann\*

Enantiocomplementary allylation of solid phase-bound aldehydes gives rise to a natural product-derived compound collection, including all stereoisomers of cryptocarya diacetate.



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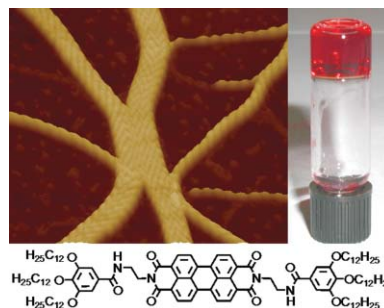
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### Functional organogels from highly efficient organogelator based on perylene bisimide semiconductor

Xue-Qing Li, Vladimir Stepanenko, Zhijian Chen, Paulette Prins, Laurens D. A. Siebbeles and Frank Würthner\*

A new n-type semiconducting perylene bisimide dye has been synthesized that gels a broad variety of organic solvents to afford well-defined nano- and mesoscopic helical fibers and bundles.

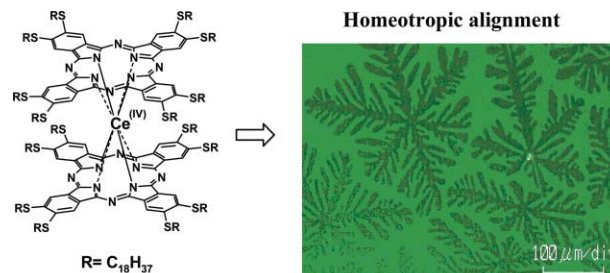


3874

### A new double-decker phthalocyanine mesogen

Fabien Nekelson, Hirosato Monobe and Yo Shimizu\*

A new double-decker cerium phthalocyanine complex shows an unusual valence state of the central lanthanide ion and, interestingly, its hexagonal columnar (Colh) mesophase demonstrates a strong tendency towards homeotropic alignment on glass substrates.

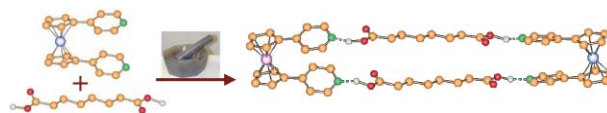


3877

### Solid-state preparation of hybrid organometallic–organic macrocyclic adducts with long chain dicarboxylic acids

Dario Braga,\* Stefano Luca Giaffreda and Fabrizia Grepioni\*

The supramolecular macrocyclic adducts of general formula  $\{[\text{Fe}(\eta^5\text{-C}_5\text{H}_4\text{-C}_5\text{H}_4\text{N})_2][\text{HOOC}(\text{CH}_2)_n\text{COOH}]\}_2$  with  $n = 4$  (adipic acid),  $n = 6$  (suberic acid),  $n = 7$  (azelaic acid) and  $n = 8$  (sebacic acid) have been obtained quantitatively by kneading powdered samples of the crystalline organometallic and organic reactants with drops of MeOH (for  $n = 4, 6$  and  $7$ ) and by direct crystallization from MeOH for  $n = 8$ .

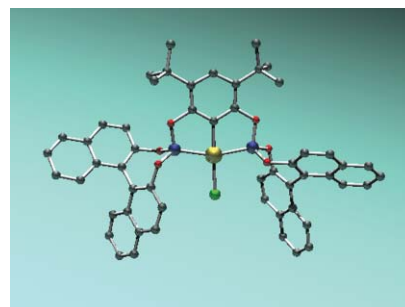


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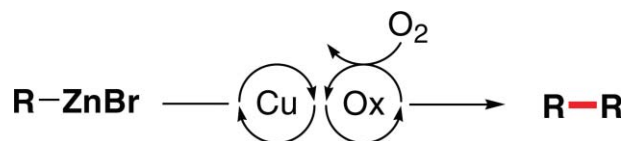
### Chiral palladium bis(phosphite) PCP-pincer complexes via ligand C–H activation

R. Angharad Baber, Robin B. Bedford,\* Michael Betham, Michael E. Blake, Simon J. Coles, Mairi F. Haddow, Michael B. Hursthouse, A. Guy Orpen, Lukasz T. Pilarski, Paul G. Pringle\* and Richard L. Wingad

The first synthesis of bis(phosphite) PCP-pincer complexes via ligand C–H activation is reported; the chiral palladium complexes produced show promising activity in the asymmetric allylation of aldehydes.



3883

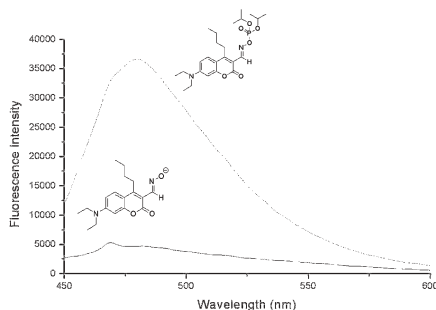


### Copper catalyzed oxidation of organozinc halides

Xianbin Su, David J. Fox, David T. Blackwell, Kiyotaka Tanaka and David R. Spring\*

A wide range of organozinc substrates may be oxidized in the presence of catalytic copper to give carbon–carbon bonds in high yield. The new methodology was applied in the total synthesis of the *Amaryllidaceae* alkaloid buflavine.

3886

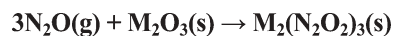


### Detection of chemical warfare simulants by phosphorylation of a coumarin oximate

Karl J. Wallace, Ruth I. Fagbemi, Frantz J. Folmer-Andersen, Jeroni Morey, Vincent M. Lynth and Eric V. Anslyn\*

The detection of chemical warfare simulants is attained by the PET mechanism that gives an “off–on” fluorescent response with a half-life of approximately 50 ms upon phosphorylation of a reactive oximate functionality.

3889

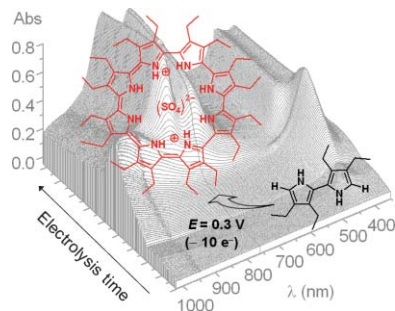


### The remarkable reaction of N<sub>2</sub>O with a binary component lanthanide oxide mixture

M. T. Harrington, E. Brennan, J. C. Wenger and M. A. Morris\*

Recent work challenges concepts about the reactivity of N<sub>2</sub>O to metal oxide surfaces particularly the lanthanide oxides.

3891



### Electrochemical synthesis of cyclo[8]pyrrole

Christophe Bucher,\* Charles H. Devillers, Jean-Claude Moutet, Jacques Pécaut and Jonathan L. Sessler\*

The synthesis of cyclo[8]pyrrole, an expanded porphyrin macrocycle, has been achieved in high yield through a straightforward and efficient electrochemical oxidative bipyrrrole C(2)–C(2)' coupling process.

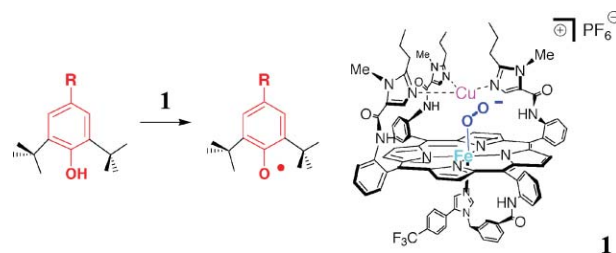


3894

**Single-turnover intermolecular reaction between a Fe<sup>III</sup>-superoxide-Cu<sup>I</sup> cytochrome *c* oxidase model and exogenous Tyr244 mimics**

James P. Collman,\* Richard A. Decréau and Christopher J. Sunderland

An Fe<sup>III</sup>-superoxide-Cu<sup>I</sup> cytochrome *c* oxidase model reacts intermolecularly with hindered phenols leading to phenoxy radicals, as was observed in the enzyme. Evidence for the formation of an Fe<sup>IV</sup>-oxo is presented.

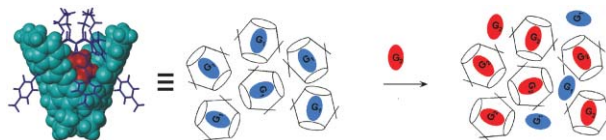


3897

**Guest exchange in dimeric capsules of a tetraurea calix[4]arene in the solid state**

Marat A. Ziganshin, Ludmila S. Yakimova, Khasan R. Khayarov, Valery V. Gorbachuk,\* Myroslav O. Vysotsky and Volker Böhmer\*

A solid powder of hydrogen bonded dimers of a tetraurea calix[4]arene is able to exchange the encapsulated guest in contact with the vapor of a second guest.

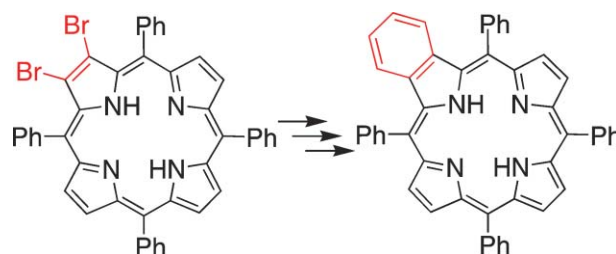


3900

**Benzoporphyrins via an olefin ring-closure metathesis methodology**

Lijuan Jiao, Erhong Hao, Frank R. Fronczek, M. Graça H. Vicente and Kevin M. Smith\*

5,10,15,20-Tetraphenylporphyrin can be converted efficiently into mono-, di-, or tri-benzoporphyrins using a reaction sequence of (1) vicinal bromination, (2) Suzuki allylation, (3) ring-closure olefin metathesis and (4) oxidation.

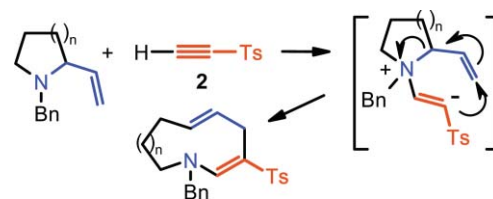


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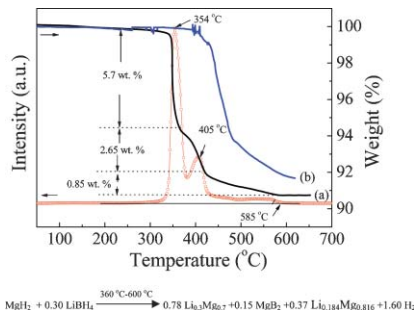
**Ring-expansion of tertiary cyclic  $\alpha$ -vinylamines by tandem conjugate addition to (*p*-toluenesulfonyl)ethyne and formal 3-aza-Cope rearrangement**

Mitchell H. Weston, Katsumasa Nakajima, Masood Parvez and Thomas G. Back\*

Cyclic  $\alpha$ -vinylamines undergo conjugate addition to the acetylenic sulfone **2**, followed by aza-Cope rearrangement, to afford functionalized medium and large-ring amines under remarkably mild conditions.



3906

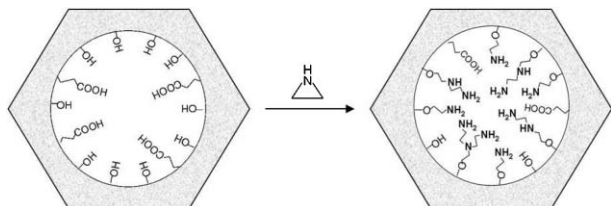


### A new dehydrogenation mechanism for reversible multicomponent borohydride systems—The role of Li–Mg alloys

X. B. Yu, D. M. Grant and G. S. Walker\*

Magnesium destabilized the  $\text{LiBH}_4$  resulting in complete dehydrogenation of the borohydride phase and the formation of a Li–Mg alloy, prior to the appearance of  $\text{MgB}_2$ .

3909

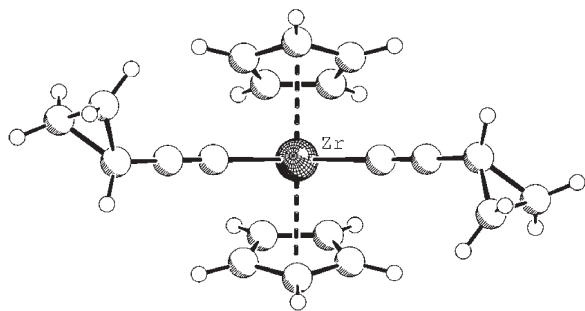


### Amino-functionalization of large-pore mesoscopically ordered silica by a one-step hyperbranching polymerization of a surface-grown polyethylenimine

Jessica M. Rosenholm, Antti Penninkangas and Mika Lindén\*

A simple method for surface functionalization of large-pore mesoporous silica by hyperbranching polymerization resulting in a high loading of amine groups is presented.

3912

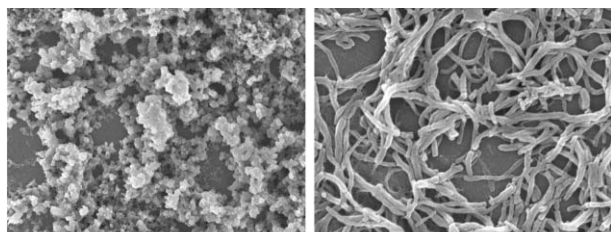


### Stereo-electronic interaction in complex molecules: cyclopropyl conjugation with Lewis acidic centres across connecting carbon–carbon triple bonds

Gereon Dierker, Gerald Kehr, Roland Fröhlich, Gerhard Erker\* and Stefan Grimme\*

The characteristic bisected conformation observed of bis(cyclopropylethynyl) Group 4 metallocenes in the crystal supports a DFT calculated weak electronic conjugation of the bent metallocene core with the cyclopropyl substituent across the connecting carbon–carbon triple bond.

3915



Without initiator

With initiator

### A general synthetic route to nanofibers of polyaniline derivatives

Henry D. Tran and Richard B. Kaner\*

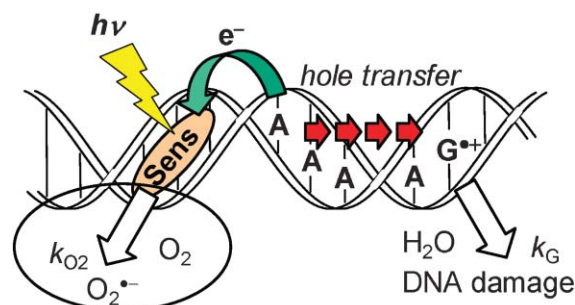
Nanofibrous mats of a wide variety of polyaniline derivatives can be synthesized without the need for templates or functional dopants by simply introducing an initiator into the reaction mixture of a rapidly mixed reaction between monomer and oxidant.

3918

### Effects of reaction rate of radical anion of a photosensitizer with molecular oxygen on the photosensitized DNA damage

Kiyohiko Kawai,\* Yasuko Osakada, Mamoru Fujitsuka and Tetsuro Majima\*

The reaction rate of the radical anion of the photosensitizer was demonstrated to be critically important in determining the efficiency of photosensitized DNA damage.

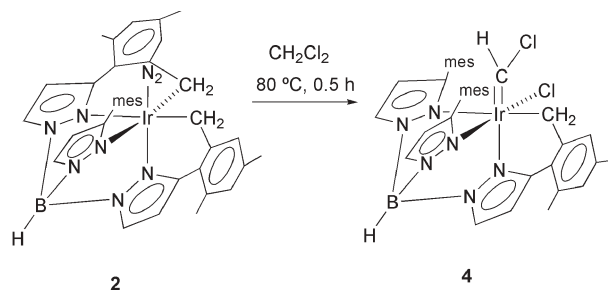


3921

### Unusual fragmentation of $\text{CH}_2\text{Cl}_2$ by an Ir(III) centre bonded to a doubly metalated $\text{Tp}^{\text{Ms}}$ Ligand ( $\text{Tp}^{\text{Ms}}$ = hydrotris(3-mesitylpyrazol-1-yl)borate)

Jorge A. López, Kurt Mereiter, Margarita Paneque, Manuel L. Poveda, Oracio Serrano, Swiatoslaw Trofimenko and Ernesto Carmona\*

The iridium complex **2** cleaves a molecule of  $\text{CH}_2\text{Cl}_2$  to yield the coordinated chlorocarbene  $=\text{C}(\text{H})\text{Cl}$ , and chloride ligands of **4**, plus a H atom that adds across one of the Ir- $\text{CH}_2$  bonds of **3** to regenerate an *o*- $\text{CH}_3$  (Ms) group in **4**.

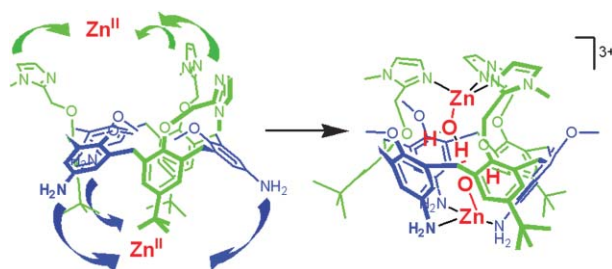


3924

### Encapsulation of a $(\text{H}_3\text{O}_2)^-$ unit in the aromatic core of a calix[6]arene closed by two Zn(II) ions at the small and large rims

David Coquière, Jérôme Marrot and Olivia Reinaud\*

The calix[6]arene aza-ligand allows the simultaneous binding of one Zn(II) ion at the small rim and a second one at the large rim, thus stabilizing a  $\text{Zn}_2(\text{H}_3\text{O}_2)$  unit in its hydrophobic cavity.

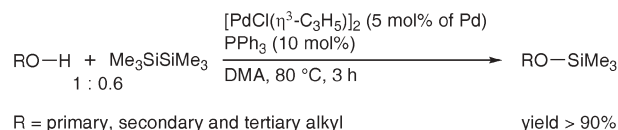


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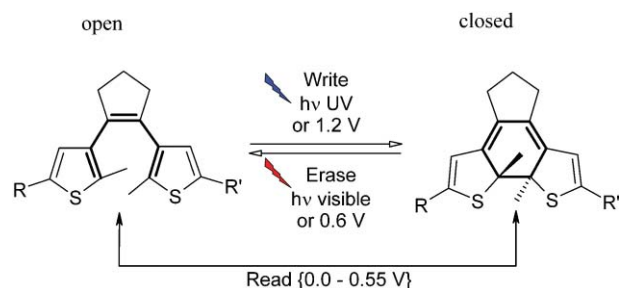
### Palladium-catalyzed silylation of alcohols with hexamethyldisilane

Eiji Shirakawa,\* Koji Hironaka, Hidehito Otsuka and Tamio Hayashi\*

An atom-economical way of the trimethylsilylation of alcohol is presented, where both of the two trimethylsilyl groups of hexamethyldisilane are transferred to alcohols with the aid of a palladium catalyst.



3930

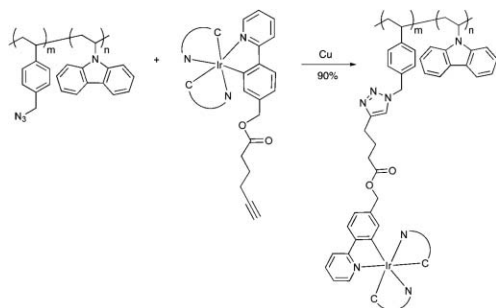


### Photo- and electrochromism of diarylethene modified ITO electrodes—towards molecular based read–write–erase information storage

Jetsuda Areephong, Wesley R. Browne, Nathalie Katsonis and Ben L. Feringa\*

Molecular memory devices based on dithienylethene switch modified ITO electrodes undergo reversible ring opening/closing both photo- and electrochemically with non-destructive electrochemical readout.

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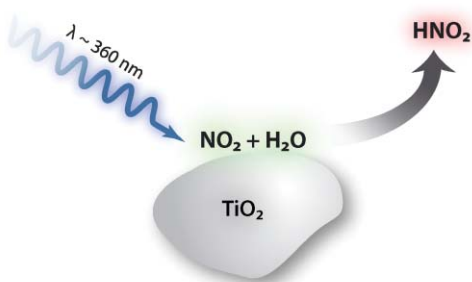


### Functionalization of polymers with phosphorescent iridium complexes *via* click chemistry

Xian-Yong Wang, Alpay Kimyonok and Marcus Weck\*

We report a simple yet highly efficient route to prepare polymers with a variety of pendant iridium complexes as potential materials in organic light-emitting diodes by employing click chemistry.

3936

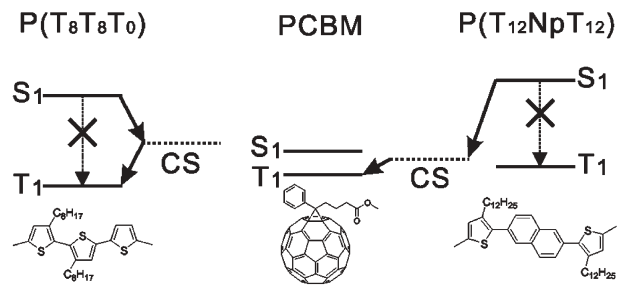


### Reduction of $\text{NO}_2$ to nitrous acid on illuminated titanium dioxide aerosol surfaces: implications for photocatalysis and atmospheric chemistry

R. Joel Gustafsson, Alexander Orlov, Paul T. Griffiths, R. Anthony Cox and Richard M. Lambert\*

$\text{TiO}_2$ , a component of atmospheric mineral aerosol, catalyses the reduction of  $\text{NO}_2$  to nitrous acid ( $\text{HONO}$ ) when present as an aerosol and illuminated with near UV light under conditions pertinent to the troposphere.

3939



### Radical ion pair mediated triplet formation in polymer–fullerene blend films

Hideo Ohkita,\* Steffan Cook, Yeni Astuti, Warren Duffy, Martin Heeney, Steve Tierney, Iain McCulloch, Donal D. C. Bradley and James R. Durrant\*

For films of high ionisation potential polythiophenes blended with a fullerene derivative, efficient triplet formation was still observed, though the polymer fluorescence was strongly quenched, and is assigned to formation *via* geminate charge recombination of bound radical ion pairs.



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