ISSN 1477-0520 CODEN OBCRAK 3(2) 189-376 (2005)

# In this issue.

Synthesis and stereochemistry of musacins

Relative and absolute stereochemistry of several musacins unequivocally assigned. See Kinoshita et al. page 295.





#### Cover

See John M. Sanderson, pp. 201–212. The illustration depicts protein-bound lipid molecules in the x-ray crystal structure of the mitochondrial membrane protein Cytochrome C Oxidase (from pdb entry 1v54). The image was prepared using the program PyMol (DeLano Scientific, San Carlos, CA, USA. http://www.pymol.org).

Image reproduced by permission of John M. Sanderson

© John M. Sanderson 2005

# **PERSPECTIVE**

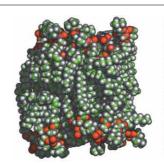
201



# Peptide-lipid interactions: insights and perspectives

John M. Sanderson\*

Studies of peptide interactions with membrane lipids allow elucidation of the roles of lipid ordering and curvature strain in processes such as budding, fusion and antimicrobial activity.





 $\Delta G$ Tm  $\Delta H$ 

2π/Q δ

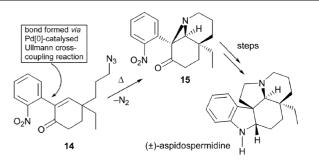
## **COMMUNICATIONS**

# 213

Exploiting the palladium[0]-catalysed Ullmann cross-coupling reaction in natural products chemistry: application to a total synthesis of the alkaloid (±)-aspidospermidine

Martin G. Banwell\* and David W. Lupton

Azide 14, available through the title cross-coupling process, has been converted, via the ring-fused aziridine 15, into the alkaloid aspidospermidine.



#### **EDITORIAL STAFF**

#### Managing editor

Caroline Potter

#### Assistant editors

Suzanne Abbott, James Crow

#### Crystallographic data editor

Kirsty Anderson

#### Publishing assistant

Emma Crisp

#### Team leader, serials production

Michelle Canning

#### Technical editors

Susan Askey, David Barden, Nicola Burton, Christopher Incles, Michael Spencelayh, Joanna Stevens

#### Editorial secretaries

Sonya Spring, Julie Thompson, Rebecca Gotobed

#### Publisher

Janet Dean

Organic & Biomolecular Chemistry (print: ISSN 1477-0520; electronic: ISSN 1477-0539) is published 24 times a year by the Royal Society of Chemistry, Thomas Graham House, Science Park, Milton Road, Cambridge, UK CB4 0WF.

All orders, with cheques made payable to the Royal Society of Chemistry, should be sent to Portland Customer Services, Commerce Way, Colchester, Essex, CO2 8HP. Tel +44 (0) 1206 226050; E-mail sales@rscdistribution.org

2005 Annual (print + electronic) subscription price: £2400; USS3960. 2005 Annual (electronic) subscription price: £2160; USS3656. Customers in Canada will be subject to a surcharge to cover GST. Customers in the EU subscribing to the electronic version only will be charged VAT.

If you take an institutional subscription to any RSC journal you are entitled to free, site-wide web access to that journal, You can arrange access via internet Protocol (IP) address at www.rsc.org/ip. Customers should make payments by cheque in sterling payable on a UK clearing bank or in US dollars payable on a US clearing bank. Periodicals postage paid at Rahway NJ, USA, and at additional mailing offices. Airfreight and mailing in the USA by Mercury Airfreight International Ltd., 365 Blair Road, Avenel, NJ 07001,

US Postmaster: send address changes to Organic & Biomolecular Chemistry, c/o Mercury Airfreight International Ltd., 365 Blair Road, Avenel, NJ 07001. All despatches outside the UK by Consolidated

PRINTED IN THE UK

Advertisement sales: Tel +44 (0) 1223 432243; Fax +44 (0) 1223 426017; E-mail advertising@rsc.org

# **Organic & Biomolecular Chemistry**

An international journal of synthetic, physical and biomolecular organic chemistry

#### www.rsc.org/obc

Organic & Biomolecular Chemistry brings together molecular design, synthesis, structure, function and reactivity in one journal. It publishes fundamental work on synthetic, physical and biomolecular organic chemistry as well as all organic aspects of: chemical biology, medicinal chemistry, natural product chemistry, supramolecular chemistry, macromolecular chemistry, theoretical chemistry, and catalysis.

# **EDITORIAL BOARD**

#### Chair

Professor Ben Feringa, Groningen

Professor Chris Abell, Cambridge Professor Varinder Aggarwal, Bristol Professor Donna Blackmond,

London

Professor Thomas Carell, Munich Professor François Diederich, ETH Professor Andrew Hamilton, Yale Professor Karl Jørgensen, Aarhus Professor Laura Kiessling,

Wisconsin-Madison Professor Shu Kobayashi, Tokyo Professor K C Nicolaou, Scripps; UC-San Diego

Professor Jay Siegel, Zürich Professor Itamar Willner, Jerusalem Professor Peter Wipf, Pittsburgh

#### International advisory board

Roger Alder (Bristol, UK) Vincenzo Balzani (Bologna, Italy) Barry Carpenter (Cornell, USA) Andre Charette (Montreal, Canada) Peter Chen (ETH, Switzerland) Jonathan Ellman (Berkeley, USA) Kurt Faber (Graz, Austria) Malcolm Forbes (North Carolina,

Sam Gellman (Wisconsin, USA) Jan Kihlberg (Umea, Sweden) Philip Kocienski (Leeds, UK) Steven V Ley (Cambridge, UK) Manuel Martín Lomas, (Seville,

Zhang Li-He (Beijing, China)

# Associate editor for North

# America

Professor Peter Wipf Department of Chemistry, University of Pittsburgh Pittsburgh, PA 15260, USA

Tel +1 412 624 8606

E-mail pwipf@pitt.edu

Michael Martinelli (Amgen, USA). Keiji Maruoka (Kyoto, Japan) E W 'Bert' Meijer (Eindhoven, The Netherlands) Eiichi Nakamura (Tokyo, Japan)

Eiichi Nakamura (Tokyo, Japan) Ryoji Noyori (Nagoya, Japan) Mark Rizzacasa (Melbourne, Australia)

Alanna Schepartz (Yale, USA) Oliver Seitz (Berlin, Germany) Kevan Shokat (UC San Francisco; UC Berkeley) Steve Street (Pfizer, UK)

Suzanne Walker (Harvard, USA) Jon Waltho (Sheffield, UK) James D White (Oregon, USA) Henry N. C. Wong (Hong Kong, China) Sam Zard (Ecole Polytechnique, France)

# **INFORMATION FOR AUTHORS**

Full details of how to submit material for publication in Organic & Biomolecular Chemistry are given in Instructions for Authors on our Web site http://www.rsc.org/authors. Correspondence on editorial matters should be addressed to: Dr Caroline V Potter, Managing Editor, Organic Publications, The Royal Society of Chemistry, Thomas Graham House, Science Park, Milton Road, Cambridge, UK, CB4 OWF.
El +444 (0) 1223 432137; Fax +44 (0) 1223 420247 E-mall obc@rsc.org

Authors may reproduce/republish portions of their published contribution without seeking permission from the RSC, provided that any such republication is accompanied by an acknowledgement in the form: (Original citation) — Reproduced by permission of the Royal Society of Chemistry

© The Royal Society of Chemistry, 2005. Apart from fair dealing for the purposes of research or private study for non-commercial purposes, or criticism or review, as permitted under the

Copyright, Designs and Patents Act 1988 and the Copyright and Related Rights Regulations 2003, this publication may only be reproduced, stored or transmitted, in any form or by any means, with the prior permission in writing of the Publishers or in the case of reprographic reproduction in accordance with the terms of licences issued by the Copyright Licensing Agency in the UK. US copyright law is applicable to users in the USA. The Royal Society of Chemistry takes reasonable care in the preparation of this publication but does not accept liability for the consequences of any errors or omissions.

 ⊕The paper used in this publication meets the requirements of ANSI/NISO Z39.48-1992 (Permanence of Paper).

Royal Society of Chemistry; Registered Charity No. 207890

# **COMMUNICATIONS**

#### 216

New and simple one-step cobalt-catalyzed preparation of functionalized arylstannanes from the corresponding aryl bromides or iodides

Corinne Gosmini\* and Jacques Périchon

A variety of functionalized arylstannanes are obtained in moderate to excellent yields by a one-step chemical procedure from corresponding halides and tributylstannyl chloride *via* cobalt catalysis.

## 218

# 7

# Synthesis of selectively deuterated fulvenes and indenes from enediynes

Scott W. Peabody, Boris Breiner, Serguei V. Kovalenko, Satish Patil and Igor V. Alabugin\*

A facile enediyne  $\rightarrow$  fulvene  $\rightarrow$  indene transformation provides a route to all possible isotopomers of substituted fulvenes and indenes.

# 222



# A new synthesis of difluoromethanesulfonamides—a novel pharmacophore for carbonic anhydrase inhibition

Nicholas A. Boyle, W. Richard Chegwidden and G. Michael Blackburn\*

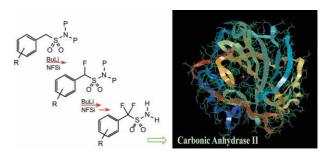
A novel, efficient synthesis of carboxydifluoromethanesulfonamide makes available a new range of inhibitors for carbonic anhydrase.

#### 225

# Synthesis of $\alpha$ -fluoro- and $\alpha$ , $\alpha$ -difluoro-benzenemethane-sulfonamides: new inhibitors of carbonic anhydrase

G. Michael Blackburn\* and Hasan Türkmen

Direct fluorination of arenemethanesulfonamide anions under mild conditions and in high yield has been accomplished using *N*-fluorobisbenzenesulfonimide.



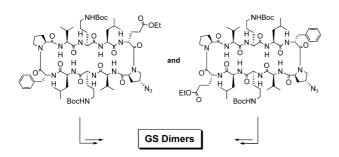
## **ARTICLES**

# 227

Phospholidines incorporating a  $\beta$  *N*-sulfonylaminoalcohol moiety: first observed selectivity of phosphorus heterocycle aminolysis in the presence of water

Frédéric Dujols, Laurence Marty and Michel Mulliez\*

The good sulfonamide leaving group enables selective phosphorylation of primary aliphatic amines.



# Synthesis and biological evaluation of gramicidin S dimers

Gijsbert M. Grotenbreg, Martin D. Witte, Peter A. V. van Hooft, Emile Spalburg, Philipp Reiß, Daan Noort, Albert J. de Neeling, Ulrich Koert, Gijsbert A. van der Marel, Herman S. Overkleeft\* and Mark Overhand\*

The design, synthesis and biological evaluation of a collection of dimeric gramicidin S analogues.

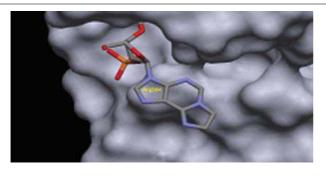
239

# Imidazolinium salts as catalysts for the aza-Diels-Alder reaction

Václav Jurčík and René Wilhelm\*

Easily accessible imidazolinium salts act as organocatalysts for the aza-Diels-Alder reaction, giving the products in up to 95% yield.

245

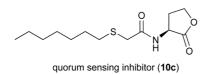


# Adenophostin A and analogues modified at the adenine moiety: synthesis, conformational analysis and biological

Charles N. Borissow, Steven J. Black, Michael Paul, Stephen C. Tovey, Skarlatos G. Dedos, Colin W. Taylor and Barry V. L. Potter\*

New biologically active structurally modified mimics of the potent agonist adenophostin A were synthesized and studied conformationally by NMR and molecular modelling giving insight into the binding of adenophostin to the IP3 receptor.

253



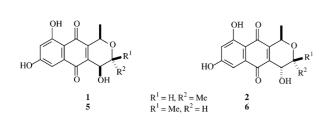
# Rational design and synthesis of new quorum-sensing inhibitors derived from acylated homoserine lactones and natural products from garlic

Tobias Persson, Thomas H. Hansen, Thomas B. Rasmussen, Mette E. Skindersø, Michael Givskov and John Nielsen\*

A parallel approach to novel scaffolds was developed, which leads to discovery of potent quorum-sensing inhibitors in Pseudomonas aeruginosa.

263





# Syntheses in enantiopure form of four diastereoisomeric naphthopyranquinones derived from aphid insect pigments

Rachna Aggarwal, Robin G. F. Giles,\* Ivan R. Green, Francois J. Oosthuizen and C. Peter Taylor

The first assembly of the monochiral naphthopyranquinones 1, 2, 5 and **6** is reported, from either (R)- or (S)-lactate.

# Total synthesis of the Fusarium toxin equisetin

Lisa T. Burke, Darren J. Dixon,\* Steven V. Ley and Félix Rodríguez

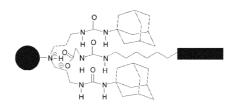
A short stereoselective synthesis of the Fusarium toxin equisetin, a potent inhibitor of HIV-1 integrase enzyme, using an IMDA key step is described.

#### 281

# The chirality of dendrimer-based supramolecular complexes

Maarten A. C. Broeren, Bas F. M. de Waal, Joost L. J. van Dongen, Marcel H. P. van Genderen and E. W. Meijer\*

Complexes between guest molecules containing Boc-protected L-phenylalanine and several urea-adamantyl dendrimers were prepared and characterized. The mobility of the end groups was investigated by optical rotation experiments.



## 286



Theoretical (DFT, GIAO-NMR, NICS) study of carbocations (M+H)+, dications (M<sup>2+</sup>) and dianions  $(M^{2-})$  from dihydro-dicyclopenta[ef,kl]heptalene (dihydro-azupyrene), dihydro-dicyclohepta[ed,gh]pentalene, and related bridged [14]annulenes

Takao Okazaki and Kenneth K. Laali\*

Annulene monocations, dications, and dianions derived from  ${\it dihydrodicyclopenta} [\it ef,kl] {\it heptalenes}, {\it dihydrodicyclohepta} [\it ed,gh]$ pentalenes, and the related bridged [14]annulenes were studied by DFT at the B3LYP/6-31G(d) and 6-31+G(d,p) levels.







# 295

# Synthesis and stereochemistry of musacins isolated from Streptomyces griseoviridis (FH-S 1832)

Toshihiko Ueki and Takamasa Kinoshita\*

The stereoselective synthesis of musacins, new secondary metabolites with anthelminthic and antiviral activities, is described.

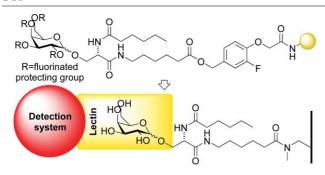
# 303



# Azidocryptands-synthesis, structure, and complexation properties

Christina Tönshoff, Klaus Merz and Götz Bucher\*

Will an aryl azide serve as a ligand to alkali cations? The answer is yes. Structural aspects and complexation properties of azido-functionalized cryptands are described.



# Solid-phase synthesis of serine-based glycosphingolipid analogues for preparation of glycoconjugate arrays

Fredrik K. Wallner, Henrik A. Norberg, Annika I. Johansson, Mickael Mogemark and Mikael Elofsson\*

Glycoconjugate arrays were prepared with galactosylceramide analogues obtained by solid-phase synthesis using a fluorinated linker and <sup>19</sup>F-NMR spectroscopy as monitoring technique.

#### 316

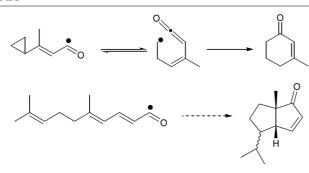


# α,β-Unsaturated and cyclopropyl acyl radicals, and their ketene alkyl radical equivalents. Ring synthesis and tandem cyclisation reactions

Christopher J. Hayes, Nicola M. A. Herbert, Nicole M. Harrington-Frost and Gerald Pattenden\*

Substituted α,β-unsaturated acyl and cyclopropyl acyl radical intermediates undergo separate cyclisations via their equivalent ketene alkyl radical species leading to a range of interesting ring structures.

## 328



# Vinylcyclopropylacyl and polyeneacyl radicals. Intramolecular ketene alkyl radical additions in ring synthesis

Benoît De Boeck, Nicola M. A. Herbert, Nicole M. Harrington-Frost and Gerald Pattenden\*

Substituted conjugated polyene acyl and vinylcyclopropyl acyl radical intermediates undergo separate rearrangement and intramolecular cyclisations via their ketene alkyl radical equivalents, leading to cyclohexenones, cyclopentenones and diquinanes.

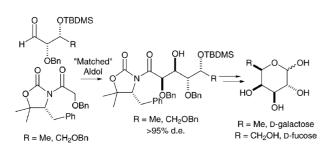
# 340

# Tandem cyclisations involving $\alpha$ -ketenyl alkyl radicals. New syntheses of the natural triguinanes pentalenene and modhephene

Benoît De Boeck, Nicole M. Harrington-Frost and Gerald Pattenden\*

New syntheses of the triquinanes pentalenene and modhephene, based on tandem cyclisations involving  $\alpha$ -ketene alkyl radical intermediates derived from  $\alpha,\beta$ -unsaturated acyl precursors, are described.

# 348



# A SuperQuat glycolate aldol approach to the asymmetric synthesis of hexose monosaccharides

Stephen G. Davies,\* Rebecca L. Nicholson and Andrew D. Smith

The asymmetric synthesis of the hexose monosaccharides D-galactose, D-fucose, D-idose, D-6-deoxyidose, D-talose and D-6-deoxytalose via an iterative double diastereoselective aldol approach is described.



# Molecular mechanism of dielectrically controlled optical resolution (DCR)

Kenichi Sakai,\* Rumiko Sakurai, Toshio Akimoto and Noriaki Hirayama\*

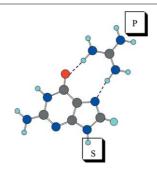
A molecular mechanism of dielectrically controlled optical resolution (DCR) was disclosed based on crystal structures of diastereomeric salts.

366

Modelling protein-RNA interactions: an electron density study of the guanidinium and formate complexes with RNA bases

Isabel Rozas,\* Ibon Alkorta and José Elguero

Complexes formed by double interactions between RNA bases and the guanidinium and formate cations have been explored theoretically.



# ReSourCe Lighting your way through the publication process

ReSourCe is a website designed to help authors and referees track the progress of their articles by providing user-friendly, rapid access to an extensive range of online services.

#### ReSourCe enables authors to:

- access submission information and tools
- submit manuscripts electronically
- track their manuscript through the peer review and publication process
- check and update their contact details
- view the history of articles previously submitted
- collect their free PDF reprints
- link to published articles

#### ReSourCe enables referees to:

- download articles for review
- report on articles
- monitor status of articles previously reviewed
- check and update their contact details
- check and update their research profile
- access refereeing guidelines and tools

"The web site is fantastic. I find it very user-friendly and clear. Congratulations"

Professor Vivian W. W. Yam, The University of Hong Kong

To register with ReSourCe, visit: www.rsc.org/resource



## **AUTHOR INDEX**

Aggarwal, Rachna, 263 Akimoto, Toshio, 360 Alabugin, Igor V., 218 Alkorta, Ibon, 366 Banwell, Martin G., 213 Black, Steven J., 245 Blackburn, G. Michael, 222, 225 Borissow, Charles N., 245 Boyle, Nicholas A., 222 Breiner, Boris, 218 Broeren, Maarten A. C., 281 Bucher, Götz, 303 Burke, Lisa T., 274 Chegwidden, W. Richard, 222 Davies, Stephen G., 348 De Boeck, Benoît, 328, 340 de Neeling, Albert J., 233 de Waal, Bas F. M., 281 Dedos, Skarlatos G., 245 Dixon, Darren J., 274 Dujols, Frédéric, 227 Elguero, José, 366

Elofsson, Mikael, 309 Giles, Robin G. F., 263 Givskov, Michael, 253 Gosmini, Corinne, 216 Green, Ivan R., 263 Grotenbreg, Gijsbert M., 233 Hansen, Thomas H., 253 Harrington-Frost, Nicole M., 316, 328 340 Hayes, Christopher J., 316 Herbert, Nicola M. A., 316, 328 Hirayama, Noriaki, 360 Johansson, Annika I., 309 Jurčík Václav 239 Kinoshita, Takamasa, 295 Koert, Ulrich, 233 Kovalenko, Serguei V., 218 Laali, Kenneth K., 286 Ley, Steven V., 274 Lupton, David W., 213 Marty, Laurence, 227 Meijer, E. W., 281

Merz, Klaus, 303 Mogemark, Mickael, 309 Mulliez, Michel, 227 Nicholson, Rebecca L., 348 Nielsen, John, 253 Noort, Daan, 233 Norberg, Henrik A., 309 Okazaki, Takao, 286 Oosthuizen, François J., 263 Overhand Mark 233 Overkleeft, Herman S., 233 Patil, Satish, 218 Pattenden, Gerald, 316, 328, 340 Paul, Michael, 245 Peabody, Scott W., 218 Périchon, Jacques, 216 Persson, Tobias, 253 Potter, Barry V. L., 245 Rasmussen, Thomas B., 253 Reiß, Philipp, 233 Rodríguez, Félix, 274 Rozas, Isabel, 366

Sakai, Kenichi, 360 Sakurai, Rumiko, 360 Sanderson, John M., 201 Skindersø, Mette E., 253 Smith, Andrew D., 348 Spalburg, Emile, 233 Taylor, C. Peter, 263 Taylor, Colin W., 245 Tönshoff, Christina, 303 Tovey, Stephen C., 245 Türkmen, Hasan, 225 Ueki, Toshihiko, 295 van der Marel, Gijsbert A., 233 van Dongen, Joost L. J., 281 van Genderen, Marcel H. P., 281 van Hooft, Peter A. V., 233 Wallner, Fredrik K., 309 Wilhelm, René, 239 Witte, Martin D., 233

#### **COPIES OF CITED ARTICLES**

The Library and Information Centre (LIC) of the RSC offers a first class Document Delivery Service for items in Chemistry and related subjects. Contact the LIC, The Royal Society of Chemistry, Burlington House, Piccadilly, London W1V 0BN, UK; Tel: +44 (0) 20 7437 8656; Fax: +44 (0) 20 7287 9798; E-mail: library@rsc.org

This service is only available from the LIC in London and not the RSC in Cambridge.

#### FREE E-MAIL ALERTS

Contents lists in advance of publication are available on the web via www.rsc.org/obc – or take advantage of our free e-mail alerting service (www.rsc.org/ej\_alert) to receive notification each time a new list becomes available.

## ADVANCE ARTICLES AND ELECTRONIC JOURNAL

Free site-wide access to Advance Articles and the electronic form of this journal is provided with a full-rate institutional subscription. See www.rsc.org/ejs for more information.

\* Indicates the author for correspondence: see article for details.



Electronic supplementary information is available on www.rsc.org/esi: see article for further information.

## **FORTHCOMING ARTICLES**

## The golden gate to catalysis

Anja Hoffmann-Röder and Norbert Krause (DOI: 10.1039/b416516k)

Stereoselective oxidation of protected inositol derivatives catalyzed by inositol dehydrogenase from *Bacillus subtilis* Richard Daniellou, Christopher P. Phenix, Pui Hang Tam, Michael C. Laliberte and David R. J. Palmer (**DOI**: 10.1039/b417757f)

Subtle factors are important: radical formation and transmetallation in reactions of butyl cuprates with cyclohexyl iodide Steven H. Bertz, Jason Human, Craig A. Ogle and Paul Seagle (DOI: 10.1039/b416612d)

Antioxidant activity of olive phenols: mechanistic investigation and characterization of oxidation products by mass spectrometry

Marjolaine Roche, Claire Dufour, Nathalie Mora and Olivier Dangles (DOI: 10.1039/b416101g)

Role of the peri-effect in synthesis and reactivity of highly substituted naphthaldehydes: a novel backbone amide linker for solid-phase synthesis

Michael Pittelkow, Ulrik Boas, Mikkel Jessing, Knud J. Jensen, and Jørn B. Christensen (DOI: 10.1039/b412971g)

Synthesis and metal complexation properties of Ph-DTPA and Ph-TTHA: novel radionuclide chelating agents for use in nuclear medicine

Sébastien G. Gouin, Jean-François Gestin, Laurence Monrandeau, Fabienne Segat-Dioury, Jean Claude Meslina and David Deniaud (**DOI**: 10.1039/b413758b)

Citations reported with a DOI instead of page numbers (e.g. A. N. Author, Org. Biomol. Chem., 2005, DOI: 10.1039/b417644h) can be easily located from the article finder at the bottom of each journal homepage (e.g. www.rsc.org/obc) or from http://xlink.rsc.org/?DOI=xxxxxxxx where xxxxxxxx is replaced by the last eight characters of the DOI (e.g. http://xlink.rsc.org/?DOI=b417644h).





CWads/05020408-colour

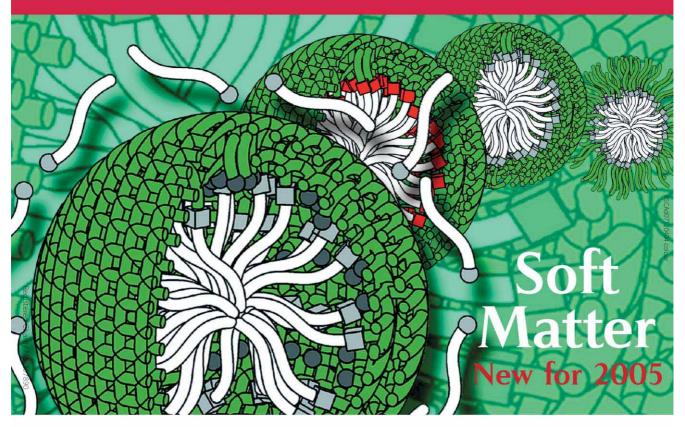
Visit the gateway for free access to chemical biology primary literature plus the latest news, reviews and funding opportunities

ROYAL SOCIETY OF CHEMISTRY advancing the chemical sciences

Further details Sales & Customer Care Dept Royal Society of Chemistry · Thomas Graham House Science Park · Milton Road · Cambridge · св4 оw ғ · u к T +44(0)1223 432360 · F +44(0)1223 426017 · E sales@rsc.org Or visit our websites: www.rsc.org and www.chemsoc.org Registered Charity No. 207890



A new journal providing a forum for the communication of generic science underpinning the properties and applications of soft matter.



Soft Matter will publish high quality interdisciplinary research into soft materials, with a particular focus on the interface between chemistry and physics.

Main research areas will include:

- (Bulk) soft matter assemblies
- Soft nanotechnology and self-assembly
- Biological aspects of soft matter
- Surfaces, interfaces, and interactions
- Building blocks/synthetic methodology
- Theory, modelling, and simulation

Find out more, and submit at:

www.softmatter.org

