The following Communications have been judged by at least two referees to be "very important papers" and will be published online at www.angewandte.org soon:

*J. K. Jabor, R. Stößer, N. H. Thong, B. Ziemer, M. Meisel**
Unexpected Reactions of $Ag(NCCH_3)_3[(V_2O_3)_2(RPO_3)_4F]$ with H_2 and NO.

J. Barluenga, P. Moriel, C. Valdés, F. Aznar*
***N*-Tosylhydrazones as Reagents for Cross-Coupling Reactions: A Novel Route for the Synthesis of Polysubstituted Olefins**

C. Wang, Y. Hou, J. Kim, S. Sun**
A General Strategy for Synthesizing FePt Nanowires and Nanorods

*J. Ge, Y. Hu, Y. Yin**
Highly Tunable Superparamagnetic Colloidal Photonic Crystals

*J. R. Johnson, N. Fu, E. Arunkumar, W. M. Leevy, S. T. Gammon, D. Piwnica-Worms, B. D. Smith**
Squaraine Rotaxanes: Superior Substitutes for Cy-5 in Molecular Probes for Near-Infrared Fluorescence Cell Imaging

*K. E. Augustyn, J. C. Genereux, J. K. Barton**
Distance-independent DNA Charge Transport across an Adenine Tract

News

Books

Highlights

Crown jewels: The judicious use of weakly coordinating anions has recently led to the first direct syntheses of alkali-metal complexes of siloxane-based crown ethers and cryptands. These findings have implications in the areas of host–guest chemistry, inorganic ring transformations, and ring-opening polymerization.

Silicon Crown Ethers

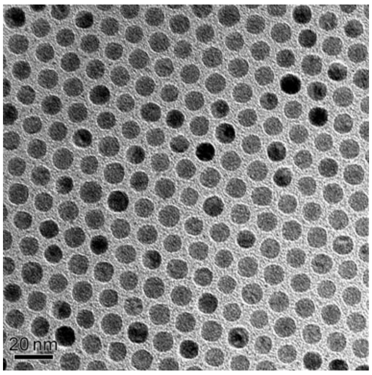
Minireviews

Leading the way to highly efficient and generally applicable enantioselective Morita–Baylis–Hillman (MBH) reactions (see scheme; EWG: electron-withdrawing group) is the use of bifunctional organocatalysis. Tethering both Lewis base (LB) and Lewis acid (LA) units to a chiral rigid backbone often generates an efficient catalyst for the MBH reaction. Cinchona alkaloids, binol, and ureas are excellent starting points for further catalyst development.

Morita–Baylis–Hillman Reaction

Reviews

Nanostructures

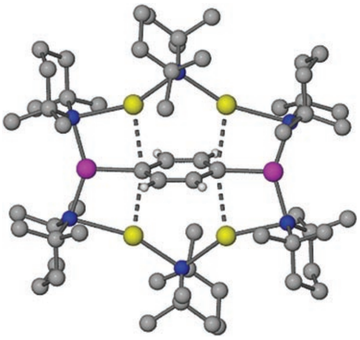


All the same: The synthesis of monodisperse nanocrystals is of key importance for many future applications, because their physical properties depend strongly on their dimensions. Mechanistic studies reveal that monodisperse nanocrystals are produced when a burst of nucleation is combined with a subsequent diffusion-controlled growth process. This Review summarizes recent progress made on the synthesis of monodisperse spherical nanocrystals (TEM image: iron oxide nanoparticles).

Communications

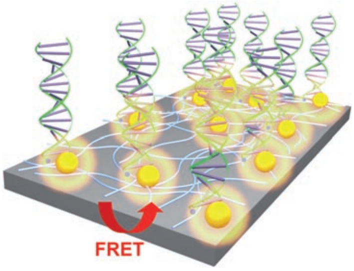
Inverse-Crown Complexes

Inside out approach: Twofold deprotonation of benzene by a sodium monoalkyl bisamido manganate(II) reagent derived from BuNa, 2,2,6,6-tetramethylpiperidine, and $Mn(CH_2SiMe_3)_2$ has produced the first inverse-crown complex in which the transition-metal atoms are incorporated in the host (see X-ray structure, blue N, green Na, purple Mn). Variable-temperature magnetization measurements show that the complex is antiferromagnetic.



DNA Microarrays

Expand on that: Signal-amplifying DNA chips have been fabricated by a light-directed on-chip DNA synthesis on a thin film of a photostable conjugated polyoxadiazole derivative. The fluorescence signal from a hexachlorofluorescein-labeled target DNA was increased by a large amount through efficient Förster energy transfer from the conjugated polymer to the dye after DNA–DNA hybridization when compared with the signal from a control slide.



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ANGEWANDTE CHEMIE International Edition (ISSN 1433-7851) is published weekly by Wiley-VCH, PO Box 191161, 69451 Weinheim, Germany. Air freight and mailing in the USA by Publications Expediting Inc., 200 Meacham Ave., Elmont, NY 11003. Periodicals postage paid at Jamaica, NY 11431. US POSTMASTER: send address changes to *Angewandte Chemie*, Wiley-VCH, 111 River Street, Hoboken, NJ 07030. Annual subscription price for institutions: US$ 5685/5168 (valid for print and electronic / print or electronic delivery); for individuals who are personal members of a national chemical society prices are available on request. Postage and handling charges included. All prices are subject to local VAT/sales tax.

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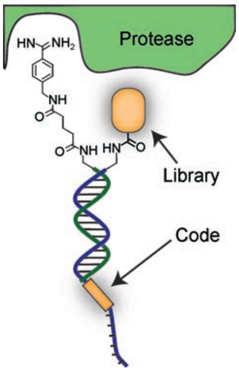


Maturing is easy to do: Annealing benzamidine–oligonucleotide conjugates with a library of DNA-encoded compounds allows the affinity capture of pharmacophores that are capable of binding to exosites adjacent to the primary substrate-binding pocket of the serine protease trypsin. Selected conjugates show an improvement in IC_{50} values of several orders of magnitude compared with the starting benzamidine.

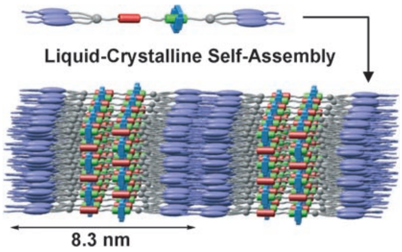
DNA-Encoded Library

S. Melkko, Y. Zhang, C. E. Dumelin, J. Scheuermann, D. Neri* **4671 – 4674**

Isolation of High-Affinity Trypsin Inhibitors from a DNA-Encoded Chemical Library

---

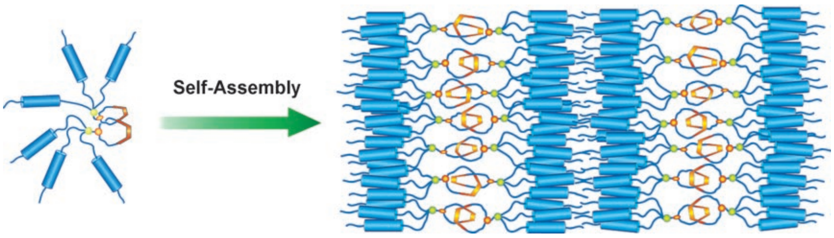
Entering a new phase: Mesogenic stoppers (purple) at the ends of the rod section of a switchable donor–acceptor [2]rotaxane induce the formation of a smectic A liquid-crystalline (LC) phase over a wide temperature range. The bistable [2]rotaxane which contains a tetracationic cyclophane (blue), a tetrathiafulvalene unit (green), and a 1,5-dioxynaphthalene unit (red) self-assembles into a LC phase with a layer spacing of about 8 nm (see picture).



Liquid Crystals

I. Aprahamian, T. Yasuda, T. Ikeda, S. Saha, W. R. Dichtel, K. Isoda, T. Kato,* J. F. Stoddart* **4675 – 4679**

A Liquid-Crystalline Bistable [2]Rotaxane

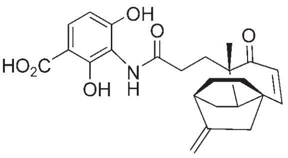


Getting organized: A free [2]catenane and its copper complex having forklike mesogens form nanosegregated liquid-crystalline (LC) structures over wide temperature ranges. The ordered arrangement of the free [2]catenane in the LC smectic A phase exerts a significant effect on the conformation of the two macrocycles, which is fixed in the ordered assembled state (see picture).

Supramolecular Chemistry

E. D. Baranoff, J. Voignier, T. Yasuda, V. Heitz,* J.-P. Sauvage,* T. Kato* **4680 – 4683**

A Liquid-Crystalline [2]Catenane and Its Copper(I) Complex



1

Two birds with one stone: Platencin (**1**) is a novel and potent broad-spectrum Gram-positive antibiotic. Whereas platensimycin is a selective inhibitor of FabF, platencin exerts its activity by a novel mode of action by dual inhibition of FabH and FabF.

Antibiotics

H. Jayasuriya,* K. B. Herath, C. Zhang, D. L. Zink, A. Basilio, O. Genilloud, M. T. Diez, F. Vicente, I. Gonzalez, O. Salazar, F. Pelaez, R. Cummings, S. Ha, J. Wang, S. B. Singh* **4684 – 4688**

Isolation and Structure of Platencin: A FabH and FabF Dual Inhibitor with Potent Broad-Spectrum Antibiotic Activity

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2nd September Issue

Biomimetric Synthesis
Y. Lu

Carbene Carbonylation
T. T. Tidwell

Repairing DNA Damage
H.-A. Wagenknecht

ACIEFS 45 (34) 5583–5716 (2006) · ISSN 1433-7851 · Vol. 45 · No. 34 · August 25, 2006

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Although *Angewandte Chemie* is owned by the German Chemical Society (Gesellschaft Deutscher Chemiker, GDCh) and is published by Wiley-VCH in a charming small town in southwest Germany, it is international in every other respect. Authors and referees from around the globe contribute to its success. Most of the articles are submitted from China (20%), USA (16%), and Japan (13%) - only then comes Germany (12%). Most of the referee reports come from Germany and the USA, but Japan and Western Europe are also well represented.

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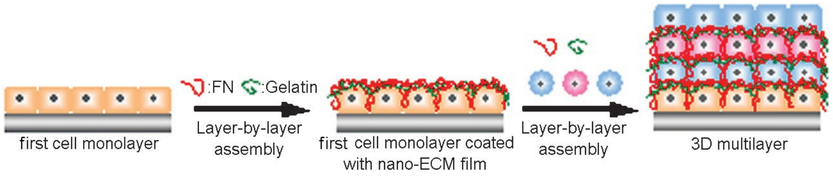
Cellular Multilayers

M. Matsusaki, K. Kadowaki, Y. Nakahara, M. Akashi* **4689 – 4692**

Fabrication of Cellular Multilayers with Nanometer-Sized Extracellular Matrix Films



Layer it on: Cellular multilayers were fabricated by preparing nanometer-sized extracellular matrix (ECM) films (6-nm thick) with fibronectin (FN) and gelatin on the surface of each cell layer. The four-layer cellular architecture was well organized and self-standing. Xenogenic human bilayer architectures similar to blood vessels were prepared by fabrication of the nanofilms on cell surfaces.

Azaspiracid (1)

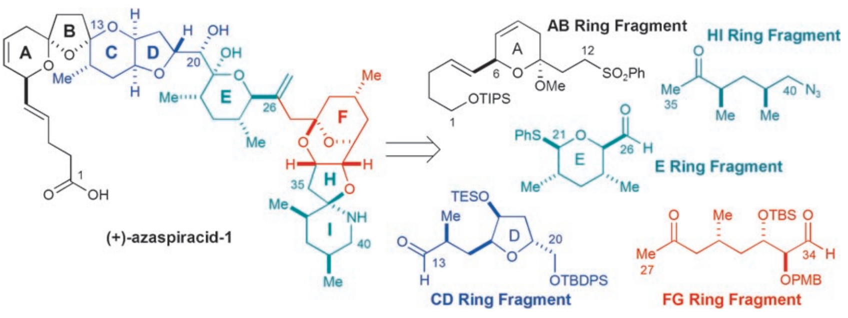
D. A. Evans,* L. Kværnø, J. A. Mulder, B. Raymer, T. B. Dunn, A. Beauchemin, E. J. Olhava, M. Juhl, K. Kagechika **4693 – 4697**

Total Synthesis of (+)-Azaspiracid-1. Part I: Synthesis of the Fully Elaborated ABCD Aldehyde

Azaspiracid (2)

D. A. Evans,* T. B. Dunn, L. Kværnø, A. Beauchemin, B. Raymer, E. J. Olhava, J. A. Mulder, M. Juhl, K. Kagechika, D. A. Favor **4698 – 4703**

Total Synthesis of (+)-Azaspiracid-1. Part II: Synthesis of the EFGHI Sulfone and Completion of the Synthesis



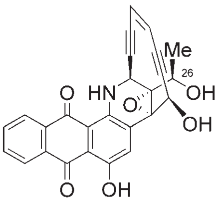
Has aspirations: The total synthesis of (+)-azaspiracid-1 has been realized. The ABCD fragment was synthesized in 20 linear steps and 16% overall yield through the enantioselective syntheses of the AB sulfone and the CD aldehyde, their coupling by a sulfone anion addition, and a thermodynamically controlled bis(spiroketalization) event. The E, FG, and HI ring fragments were synthesized enantioselectively and then coupled by using aldol methodology. Finally, a complex addition of the anion of the anomeric EFGHI sulfone to the ABCD aldehyde completed the synthesis in 26 linear steps and 2.7% yield.

Uncialamycin

K. C. Nicolaou,* H. Zhang, J. S. Chen, J. J. Crawford, L. Pasunoori **4704 – 4707**

Total Synthesis and Stereochemistry of Uncialamycin

Unseal the secrets of uncialamycin: The total synthesis of the C26 epimers of the newly discovered enediyne natural product allows its stereochemical assignment and further biological investigations of this potent antibiotic. Key steps involve the addition of an acetylide to a pyridinium species, an intramolecular formation of the enediyne, and Hauser annulation to form the anthraquinone moiety.



Uncialamycin

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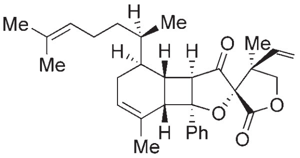
Contents

Biyouyanagin A

K. C. Nicolaou,* D. Sarlah,
D. M. Shaw ________ **4708–4711**

Total Synthesis and Revised Structure of Biyouyanagin A



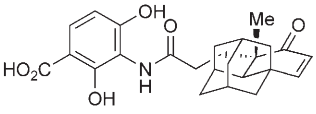
biyouyanagin A

It all adds up: A 12-step total synthesis of biyouyanagin A, an inhibitor of HIV replication, has revealed its structure, rendered it available for biological investigations, and allows the synthesis of analogues. The convergent synthesis involves two cascade sequences and a remarkably selective [2+2] cycloaddition reaction to forge the cyclobutane ring of the target molecule in the ultimate step.

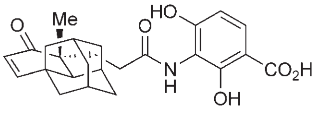
Platensimycin

K. C. Nicolaou,* T. Lister, R. M. Denton,
A. Montero, D. J. Edmonds **4712–4714**

Adamantaplatensimycin: A Bioactive Analogue of Platensimycin



(−)-**1**



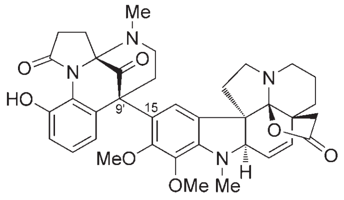
(+)-**1**

Adamantly pursuing the bugs: The syntheses of the two enantiomers of adamantaplatensimycin ((−)-**1** and (+)-**1**), a novel analogue of the newly discovered antibiotic platensimycin, have been achieved. Potent antibacterial action against methicillin-resistant *Staphylococcus aureus* and vancomycin-resistant *Enterococcus faecium* has been revealed for the (−) enantiomer.

Haplophytine

K. C. Nicolaou,* U. Majumder,
S. P. Roche, D. Y.-K. Chen* _ **4715–4718**

Construction of the “Left Domain” of Haplophytine



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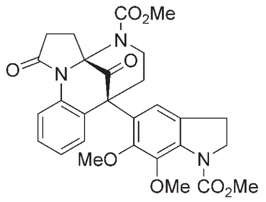


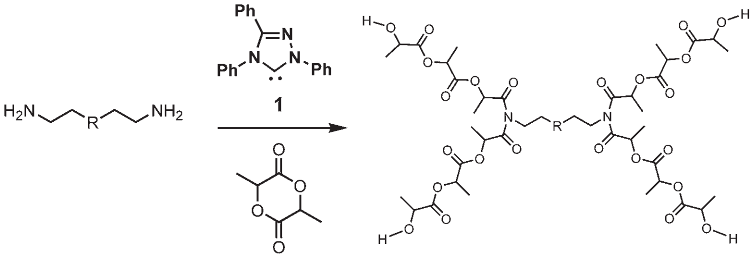
2

Left of the middle: Synthesis of the “left” structural domain (**2**) of haplophytine (**1**) features a stereoselective construction of its sterically congested carbon–carbon bond (C9′–C15) and an efficient cascade sequence involving a skeletal rearrangement of a presumed epoxide intermediate.

Block Copolymers

O. Coulembier, M. K. Kiesewetter,
A. Mason, P. Dubois, J. L. Hedrick,*
R. M. Waymouth* ________ **4719–4721**

A Distinctive Organocatalytic Approach to Complex Macromolecular Architectures



Branch manager: A one-step approach to branched copolymers is described. Primary amines are found to function as bifunctional initiators for ring-opening polymerization in the presence of **1** to promote the polymerization of two chains, thereby enabling the facile introduction of branch points in block copolymers.

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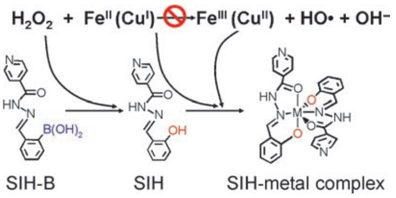
Bioinorganic Chemistry

Y. Wei, M. Guo* 4722–4725

Hydrogen Peroxide Triggered Prochelator Activation, Subsequent Metal Chelation, and Attenuation of the Fenton Reaction

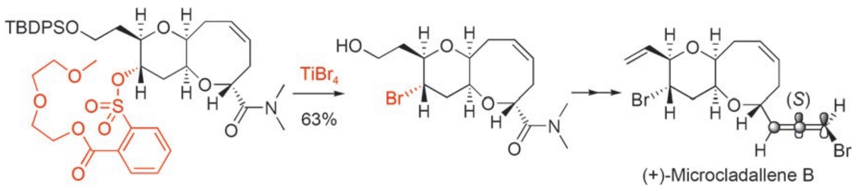
Fenton antidote: The prochelator 2-boronobenzaldehyde isonicotinoyl hydrazone (SIH-B) reacts with H_2O_2 to yield the active chelator salicylaldehyde isonicotinoyl hydrazone (SIH), which sequesters copper and iron ions (see scheme; M = Fe, Cu). This process attenuates both Fe- and Cu-promoted Fenton reactions under physiologically relevant conditions.



Natural Products Synthesis

J. Park, B. Kim, H. Kim, S. Kim, D. Kim* 4726–4728

Substrate-Controlled Asymmetric Total Synthesis of (+)-Microcladallene B with a Bromination Strategy Based on a Nucleophile-Assisting Leaving Group

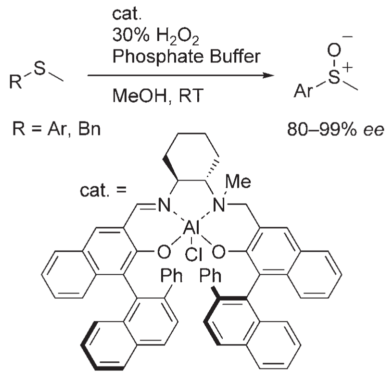


The formidable challenge of incorporating a bromine substituent stereoselectively at an sp^3 center was met through application of a nucleophile-assisting leaving group (see scheme). A further highlight of the highly stereo-, regio-, and chemoselective synthesis of (+)-microcladallene B was a dianion alkylation to provide the required α,α′-*anti* substrate for ring-closing metathesis. TBDPS = *tert*-butyldiphenylsilyl.

Asymmetric Catalysis

T. Yamaguchi, K. Matsumoto, B. Saito, T. Katsuki* 4729–4731

Asymmetric Oxidation Catalysis by a Chiral Al(salalen) Complex: Highly Enantioselective Oxidation of Sulfides with Aqueous Hydrogen Peroxide



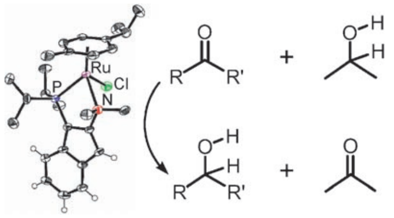
Not afraid to get wet: Aluminum(salalen) complexes were found to be water compatible and therefore suitable catalysts for asymmetric oxidation with aqueous hydrogen peroxide. A variety of sulfides underwent the title reaction in the presence of the aluminum(salalen) complex shown to the give the desired sulfoxides with high to excellent enantioselectivity (see scheme; Bn = benzyl).

Transfer Hydrogenation

R. J. Lundgren, M. A. Rankin, R. McDonald, G. Schatte, M. Stradiotto* 4732–4735

A Formally Zwitterionic Ruthenium Catalyst Precursor for the Transfer Hydrogenation of Ketones that Does Not Feature an Ancillary Ligand N–H Functionality

Fast and furious: Even without the assistance of an N–H donor ligand, the Ru complex shown in the scheme is a remarkably active precatalyst for the transfer hydrogenation of ketones in basic *i*PrOH (R,R′ = alkyl or aryl), providing near-quantitative yields within minutes and exhibiting consistently high turnover frequencies (10^4 to $10^5\,h^{-1}$) for a diverse range of substrates.



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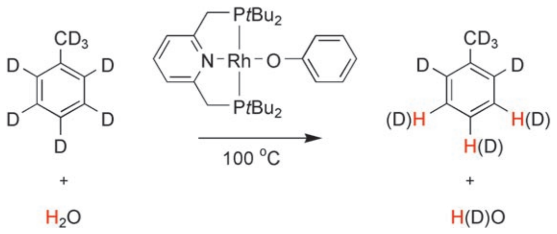
Contents

C–H Bond Activation

S. M. Kloek, D. M. Heinekey,*
K. I. Goldberg* ———— **4736–4738**

C–H Bond Activation by Rhodium(I) Hydroxide and Phenoxide Complexes



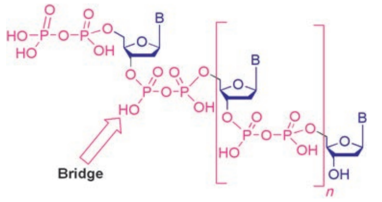
Rhodium(I) complexes of the form $[(PNP)Rh(OR)]$ ($R=H$, CH_2CF_3, C_6H_5; PNP = 2,6-bis[(di-*tert*-butylphosphino)methyl]pyridine) have been prepared. Upon thermolysis in $[D_6]$benzene, the hydroxide and trifluoroethoxide complexes undergo benzene activation. $[(PNP)Rh(OC_6H_5)]$ is an active catalyst for the H/D exchange between D_2O and benzene and between H_2O and $[D_8]$toluene (see scheme), for which exchange occurs selectively at the *meta* and *para* positions.

Modified Oligodeoxynucleotides

Y. Ahmadibeni, K. Parang* — **4739–4743**

Synthesis and Evaluation of Modified Oligodeoxynucleotides Containing Diphosphodiester Internucleotide Linkages

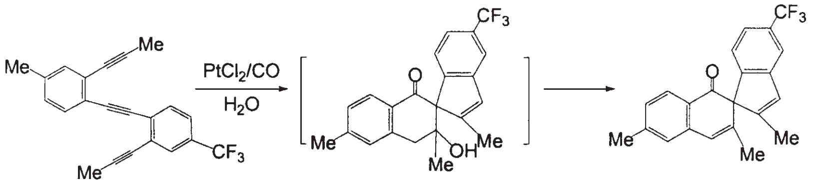
Build a bridge: Oligodeoxynucleotides (ODNs) containing diphosphodiester bridges were synthesized by a solid-phase synthesis strategy. Modified ODNs formed stable duplexes with complementary modified and unmodified nucleic acid sequences. The modified oligomers were resistant to degradation by DNase I and 3′-exonuclease I under conditions in which unmodified ODNs were degraded. B = T, A, G, or C.



Synthetic Methods

H.-K. Chang, S. Datta, A. Das, A. Odedra, R.-S. Liu* ———— **4744–4747**

$PtCl_2$-Catalyzed Hydrative Cyclization of Trialkyne Functionalities to Form Bicyclic Spiro Ketones

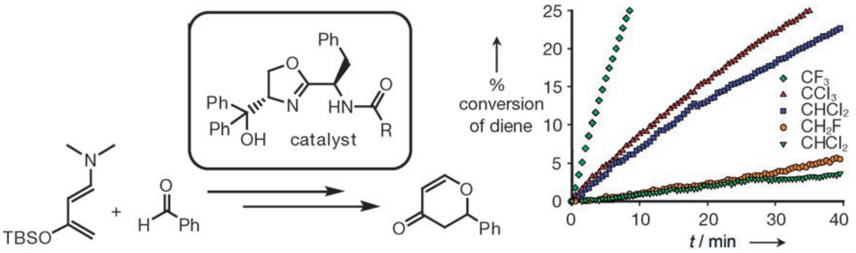


Triple rounds: A regioselective hydrative cyclization of triynes has been developed to give bicyclic β-hydroxy spiro ketones, which undergo subsequent dehydration to give the β,γ-unsaturated ketones (see scheme). Model reactions suggest that this platinum catalysis includes two selective hydrations, an alkyne insertion, and an aldol condensation.

Asymmetric Catalysis

K. H. Jensen, M. S. Sigman* **4748–4750**

Systematically Probing the Effect of Catalyst Acidity in a Hydrogen-Bond-Catalyzed Enantioselective Reaction



Defining the relationship: The effect of catalyst acidity has been systematically probed by using a modular oxazoline catalyst in a hetero-Diels–Alder reaction catalyzed by hydrogen bonding. Linear free energy relationships were observed between the catalyst acidity and both the reaction rate and enantioselectivity (see picture).

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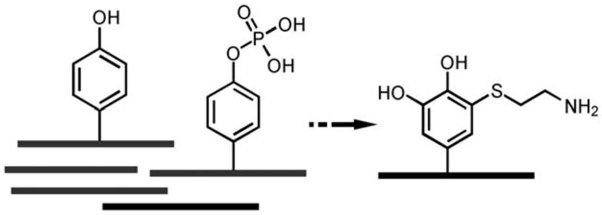
Protein Phosphorylation

S. Li,* D. Zeng ______ **4751 – 4753**

Chemoenzymatic Enrichment of Phosphotyrosine-Containing Peptides

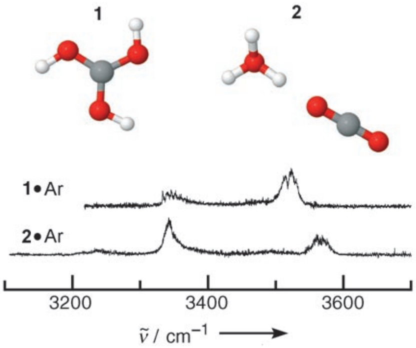


Change the Tyr: A tyrosinase-assisted chemoenzymatic method was used to modify and enrich phosphotyrosine-containing peptides from a peptide mixture. This approach could be used to study protein tyrosine phosphorylation, and represents a promising alternative to immunoaffinity purification by using anti-phosphotyrosine antibodies.

Carbenium Ions

H.-S. Andrei, S. A. Nizkorodov,
O. Dopfer* ______ **4754 – 4756**

IR Spectra of Protonated Carbonic Acid and Its Isomeric $H_3O^+ \cdot CO_2$ Complex

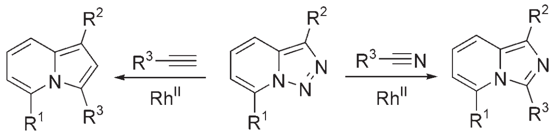


Complex detection: Gas-phase protonated carbonic acid, $C(OH)_3^+$ (**1**), and its isomeric $H_3O^+ \cdot CO_2$ complex (**2**) were spectroscopically identified for the first time by resonant IR photodissociation spectroscopy of their Ar-tagged adducts (see picture). This identification represents an important step toward understanding the precise role of protonated carbonic acid in CO_2 hydrolysis and related processes.

Annulated Heterocycles

S. Chuprakov, F. W. Hwang,
V. Gevorgyan* ______ **4757 – 4759**

Rh-Catalyzed Transannulation of Pyridotriazoles with Alkynes and Nitriles

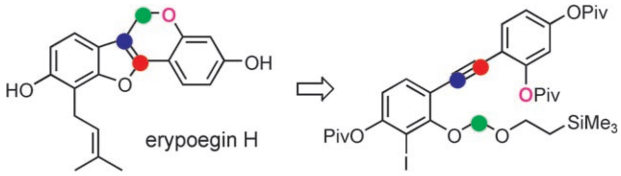


Changing the rings: A variety of N-fused pyrrolo- and imidazopyridines can be readily formed by a direct Rh-catalyzed transannulation of pyridotriazoles with alkynes and nitriles, respectively (see scheme). Substituted pyridotriazoles can also serve as stable precursors for Rh carbenoids, the preparation of which does not require special precautions or slow-addition techniques.

Natural Products

A. Fürstner,* E. K. Heilmann,
P. W. Davies ______ **4760 – 4763**

Total Synthesis of the Antibiotic Erypoegin H and Cognates by a $PtCl_2$-Catalyzed Cycloisomerization Reaction



Fighting back: A concise route to the pterocarpene derivative erypoegin H, a natural product endowed with considerable activity against a range of methicillin-resistant *Staphyllococcus aureus* strains and vancomycin-resistant enterococci, has been developed. The key step in the synthesis is a $PtCl_2$-catalyzed carboalkoxylation reaction of an alkyne (see retrosynthesis; Piv = pivaloyl).

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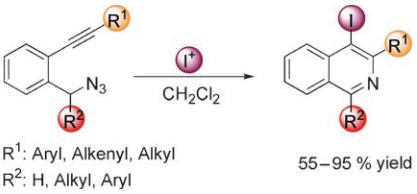
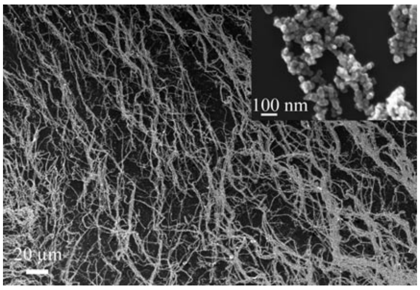
Synthetic Methods



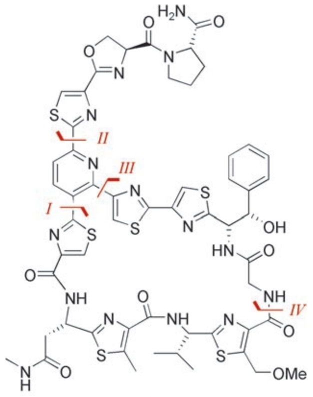
The I's have it: A series of 2-alkynyl benzyl azides have been smoothly converted into 1,3,4-trisubstituted isoquinolines in moderate to excellent yields by an iodonium-mediated synthesis (see scheme). Depending on the structure of the substrate, iodine, Barluenga reagent (Py_2IBF_4/HBF_4), or *N*-iodosuccinimide has been employed as the I^+ source.

Nanoparticle Aggregates



In the field: In the presence of a time-varied magnetic field, electrostatically stabilized magnetite nanoparticles aggregate into microsized fibrous suprastructures (see image). The process is irreproducible for sterically stabilized colloids. It is inferred that the phenomenon results from the co-effects of magnetic moment interaction and electrostatic interaction that are connected by the time-varied magnetic field.

Total Synthesis

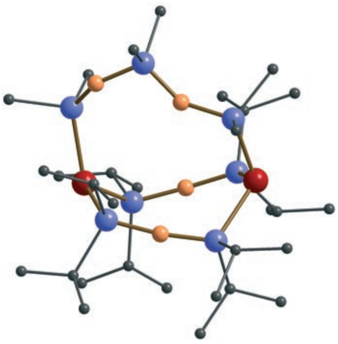


1

When one door closes, another opens: In the synthesis of the thiazolyl peptide GE2270 A (**1**), the bonds labeled *I* and *II* at the pyridine core were established by two consecutive cross-coupling reactions. Amide bond formation (*IV*) and subsequent intramolecular Stille reaction (*III*) were more effective than the originally conceived connection strategy (*III* before *IV*). GE2270 A (**1**) was prepared with an overall yield of 4.8 % in 20 steps along the longest linear sequence.

Cage Compounds

Modeled on organics: The inorganic cryptand [$P_2\{O(Si\mathit{i}Pr_2)_2\}_2\{SiMe_2(OSiMe_2)_2\}$] (see figure; C black, O orange, P red, Si blue) can be synthesized by a stepwise lithiation/silylation process. A first coordination compound of this ligand was obtained from its reaction with $Li[Al(OR_F)_4]$.



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On the path to chiral polysilanes, the synthesis and first structural characterization of the enantiomerically pure tetrasilane (*R*)-**3** was achieved. A new synthetic strategy was used, by which the cleavage of a silicon–phenyl bond in the substrate (*R*)-**1** generated as intermediate the enantiomerically enriched lithiosilane **2**.

Chiral Silanes

C. Strohmann,* C. Däschlein, M. Kellert, D. Auer ______ **4780 – 4782**

A Highly Enantiomerically Enriched Lithiosilane by Selective Cleavage of a Silicon–Phenyl Bond with Lithium

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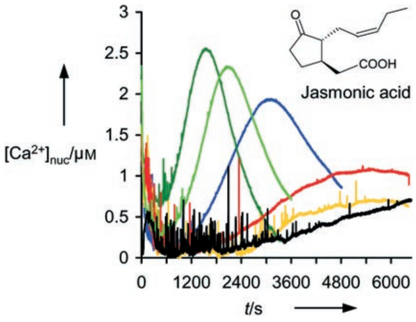
A three-class society: Jasmonates, a group of oxylipin phytohormones differentially induce changes in intracellular Ca^{2+} concentrations in plant cells (see diagram). Structure–activity analysis revealed that jasmonates fall into three distinct classes: 1) compounds inducing Ca^{2+} changes in both the cytosol and the nucleus 2) compounds inactive on either compartment, and 3) compounds acting selectively on the nucleus.



Signal Transduction

A. Walter, C. Mazars, M. Maitrejean, J. Hopke, R. Ranjeva, W. Boland, A. Mithöfer* ______ **4783 – 4785**

Structural Requirements of Jasmonates and Synthetic Analogues as Inducers of Ca^{2+} Signals in the Nucleus and the Cytosol of Plant Cells

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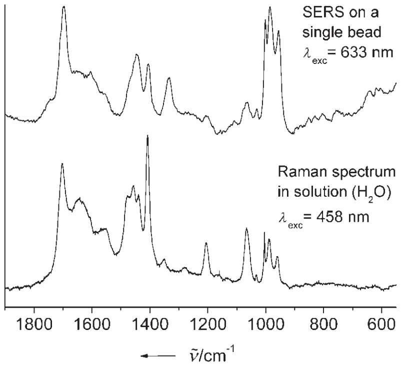

Sensitive and specific: By using surface-enhanced Raman scattering (SERS) microspectroscopy, solid-phase-bound compounds can be detected directly and marker-free in a few seconds on a single polystyrene bead. No significant contributions of the matrix are observed, as demonstrated by reference Raman spectra.

Analytical Methods

C. Schmuck,* P. Wich, B. Küstner, W. Kiefer, S. Schlücker* ______ **4786 – 4789**

Direct and Label-Free Detection of Solid-Phase-Bound Compounds by Using Surface-Enhanced Raman Scattering Microspectroscopy

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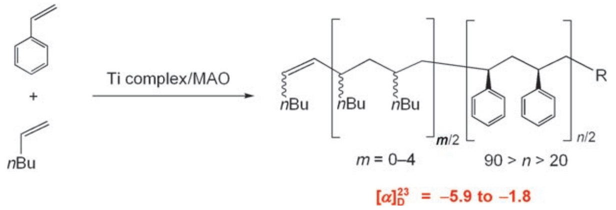

Tactful: Titanium catalyst precursors in both optically active, enantiomeric forms have been obtained diastereoselectively, and after activation with methylaluminoxane (MAO) they were used for the stereospecific oligomerization of styrene. The isotactic polystyrenes with a degree of polymerization of up to 45 show measurable optical activity, thus corroborating that enantiomorphic site control is operating in these homogeneous catalysts.

Polymerization Catalysis

VIP

K. Beckerle, R. Manivannan, B. Lian, G.-J. M. Meppelder, G. Raabe, T. P. Spaniol, H. Ebeling, F. Pelascini, R. Mülhaupt, J. Okuda* ______ **4790 – 4793**

Stereospecific Styrene Enchainment at a Titanium Site within a Helical Ligand Framework: Evidence for the Formation of Homochiral Polystyrene

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Supporting information is available on the WWW (see article for access details).

A video clip is available as Supporting Information on the WWW (see article for access details).

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