JACS

JOURNAL OF THE AMERICAN CHEMICAL SOCIETY

2001

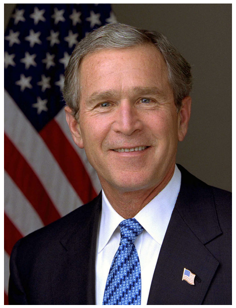
Literature Review

Philipp Gritsch

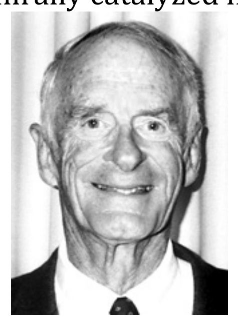
12 March 2015

AK Gaich – Leibniz University Hannover

Introduction



Chirally catalyzed hydrogenation and oxidation reactions



William S. Knowles

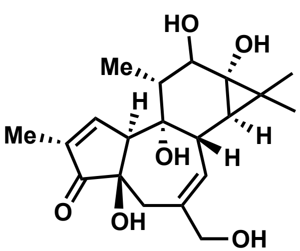
Ryoji Noyori

K. Barry Sharpless

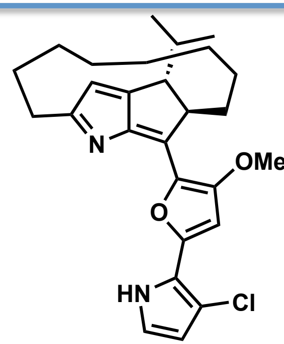
In JACS:

- 2833 Articles / 73 with topic „Total Synthesis“
  - In Total Synthesis: 5 by Larry Overman , 4 by Dale Boger, Samuel Danishefsky and Amos B. Smith III
  - Overall: Barry Trost published 14 articles

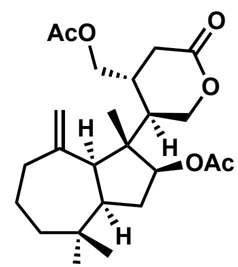
Total Syntheses



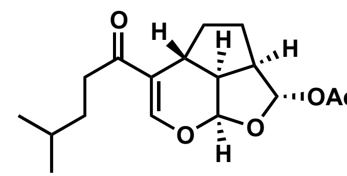
(+)-Phorbol (1)



Roseophiline (2)



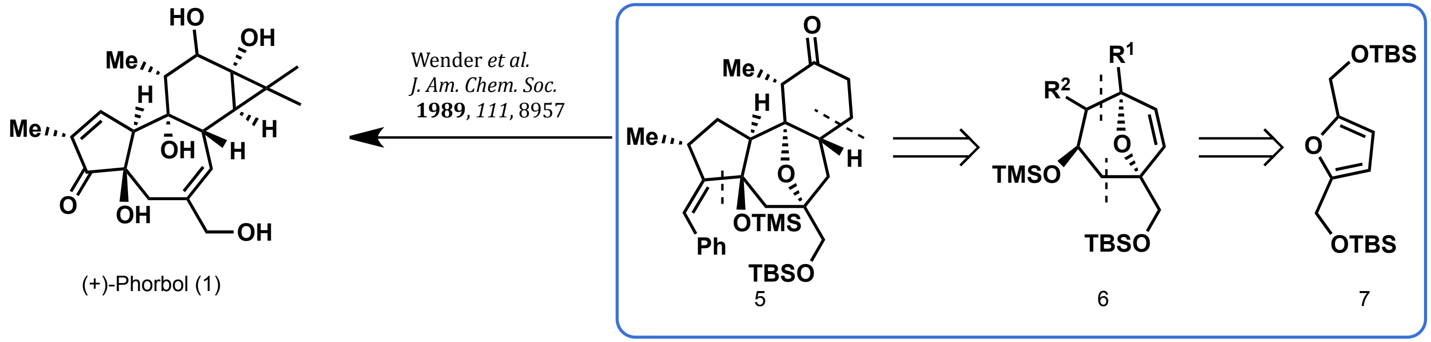
(+)-Shahamin K (3)

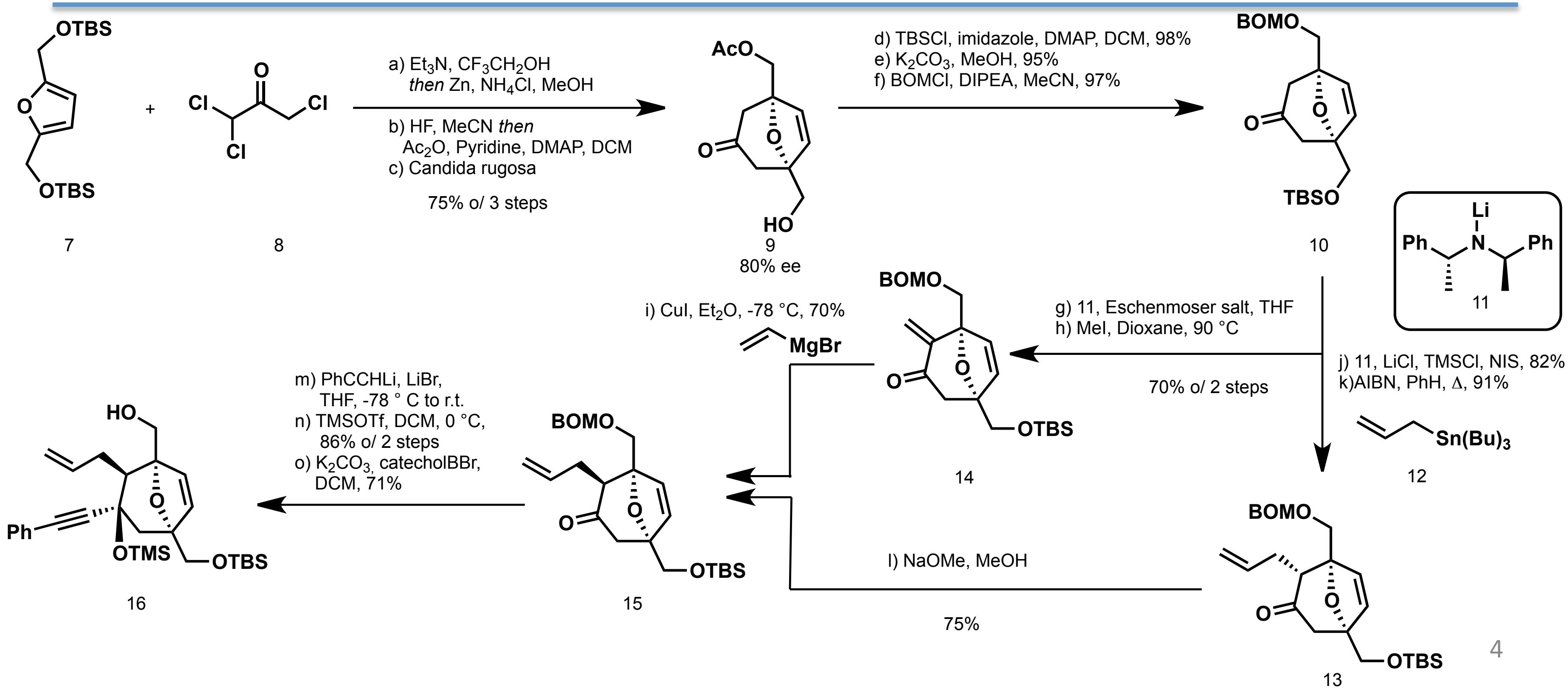


Euplotin A (4)

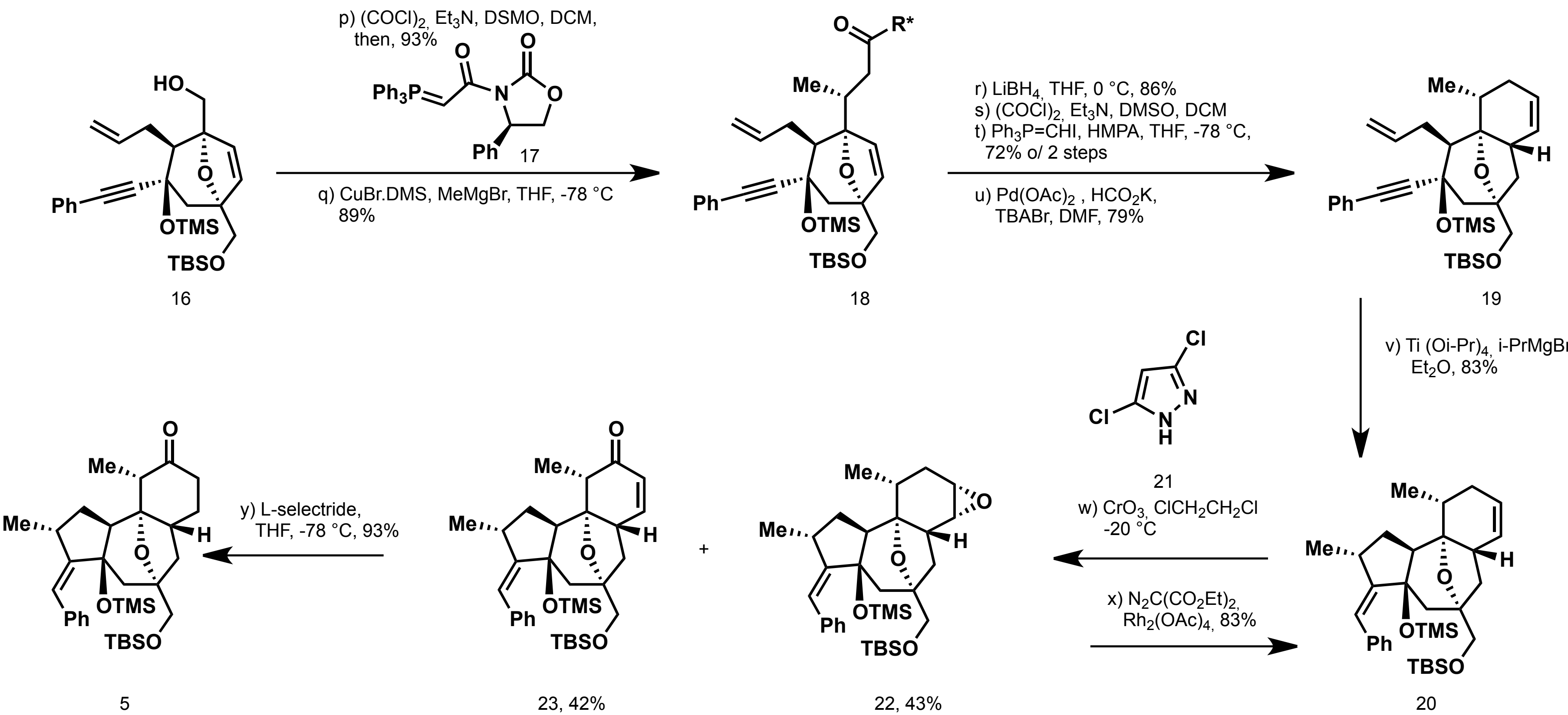
Formal Synthesis of (+)–Phorbol

Lee , K.; Cha, J.K. *J. Am. Chem. Soc.* **2001**, *123*, 5590-5591





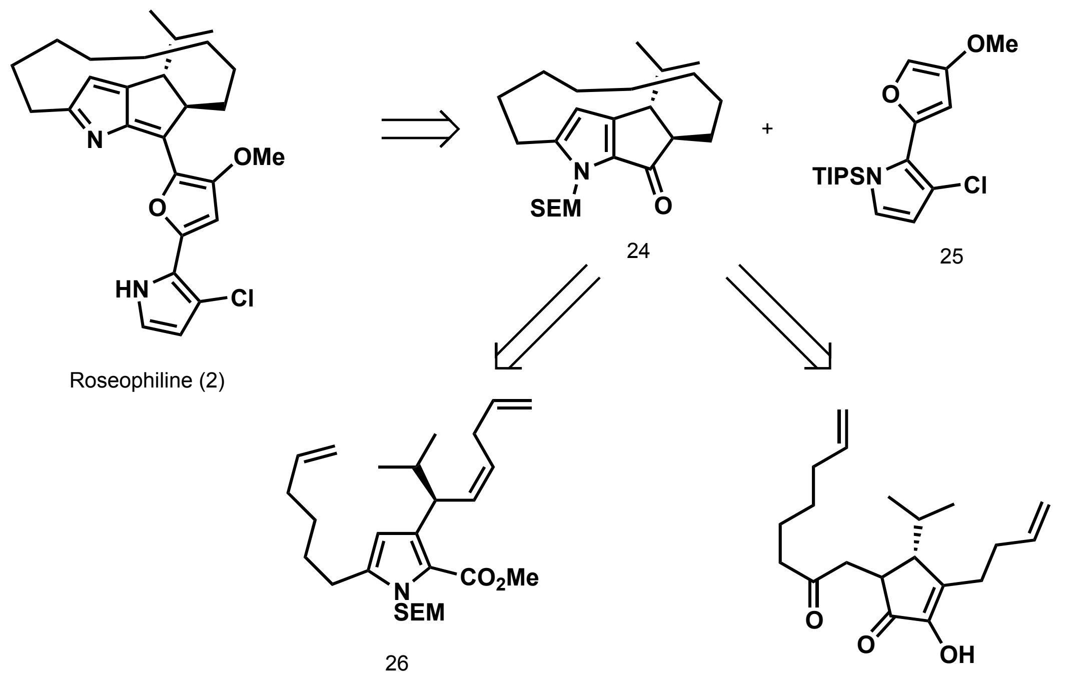
Formal Synthesis of (+)–Phorbol



Asymmetric synthesis of Roseophiline

Harrington, P.E. Tius, M.A. *J. Am. Chem. Soc.* **2001**, *123*, 8509 – 8514

Boger, D.L.; Hong, J. *J. Am. Chem. Soc.* **2001**,*123*, 8515 - 8519



Roseophiline (2)

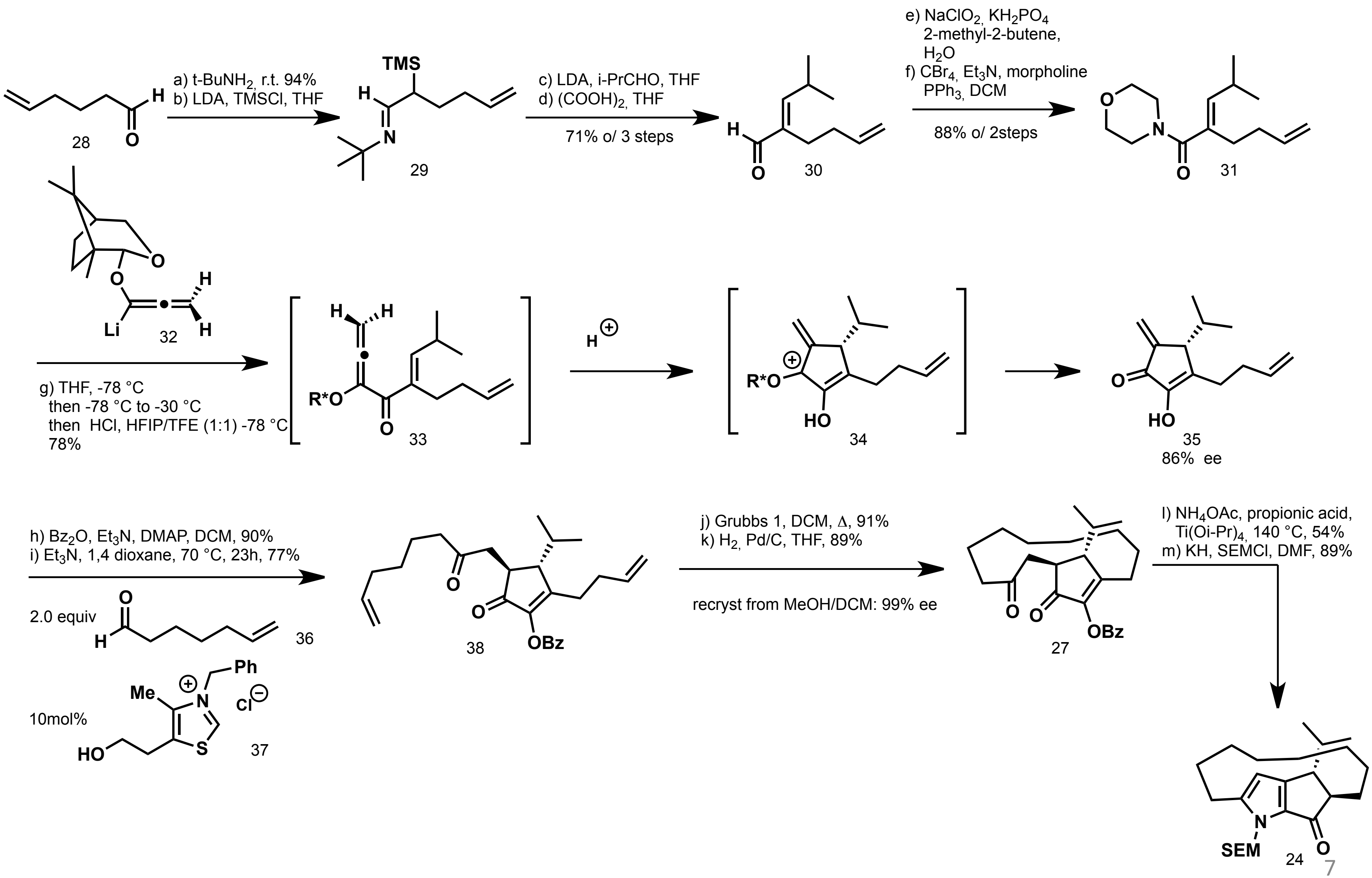
24

25

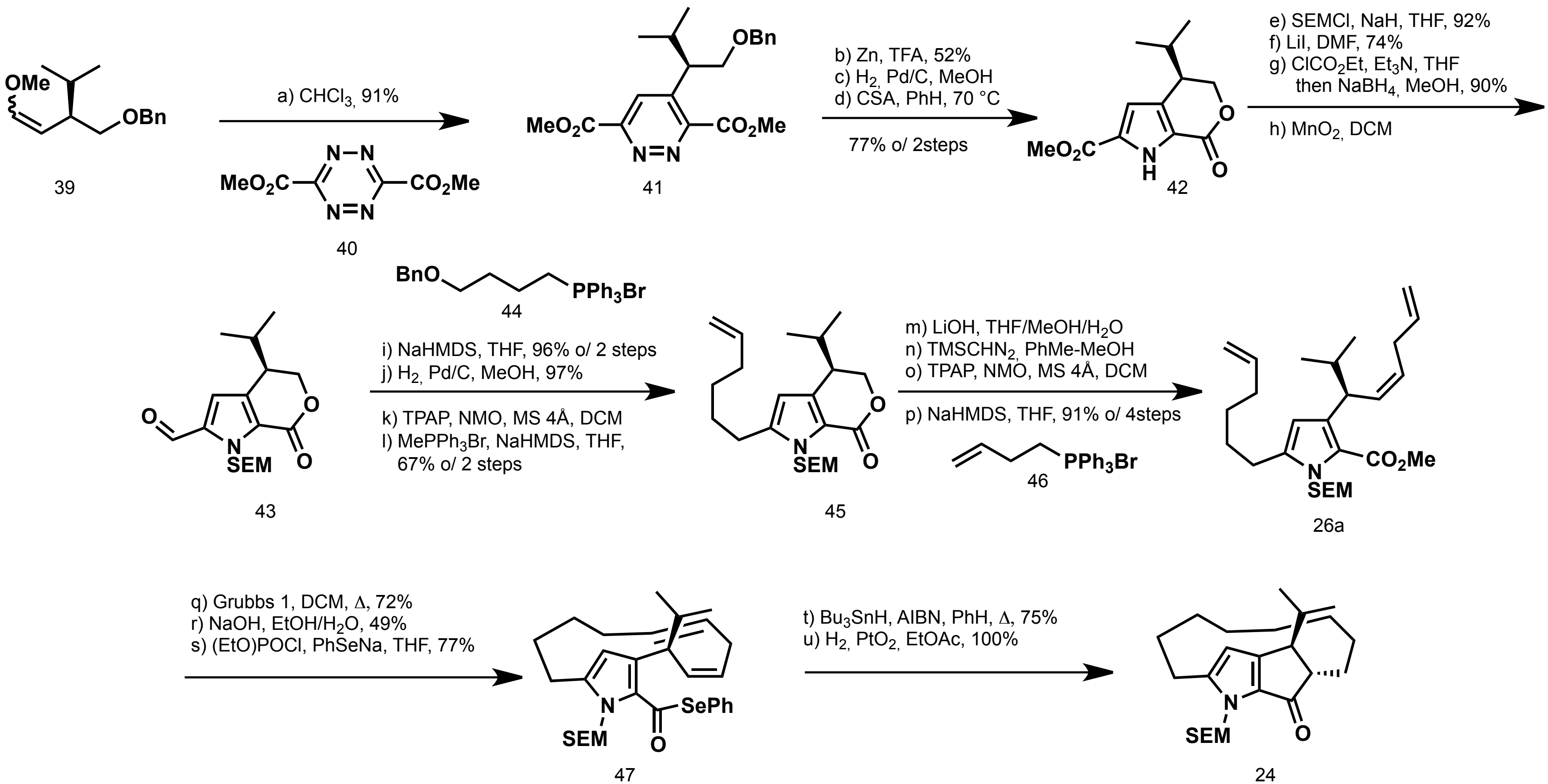
26
Hong and Boger
ent compound
inverse electron demand hetero-Diels-Alder
radical alkene cyclization
26 steps, 1.8%

27
Harrington and Tius
asymmetric cyclopentannelation
Knorr pyrrole synthesis
17 steps, 7.0% overall

Roseophiline: Harrington and Tius

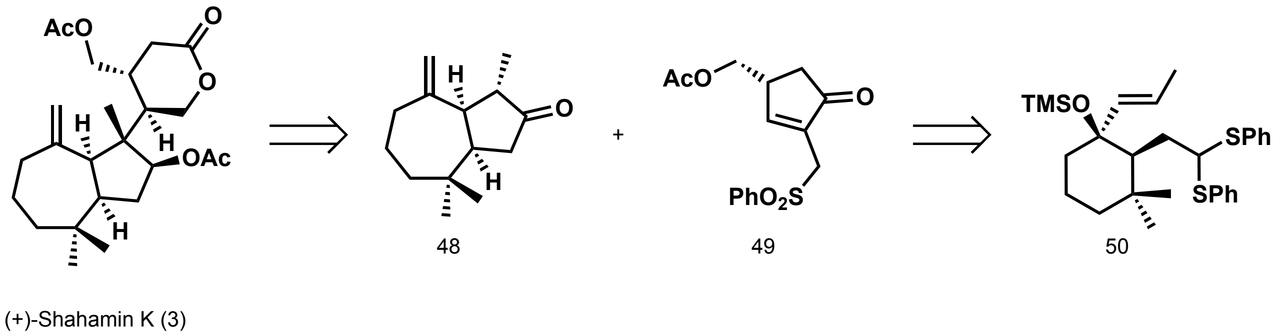


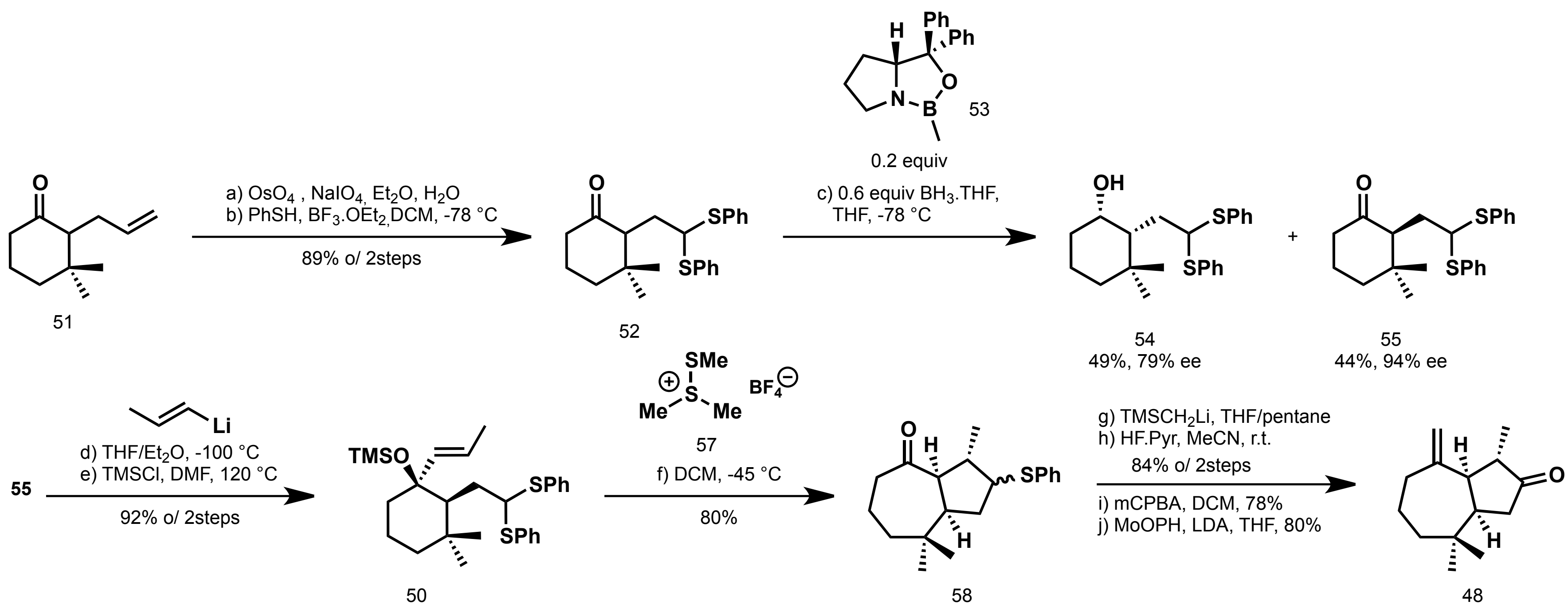
Roseophiline: Boger and Hong



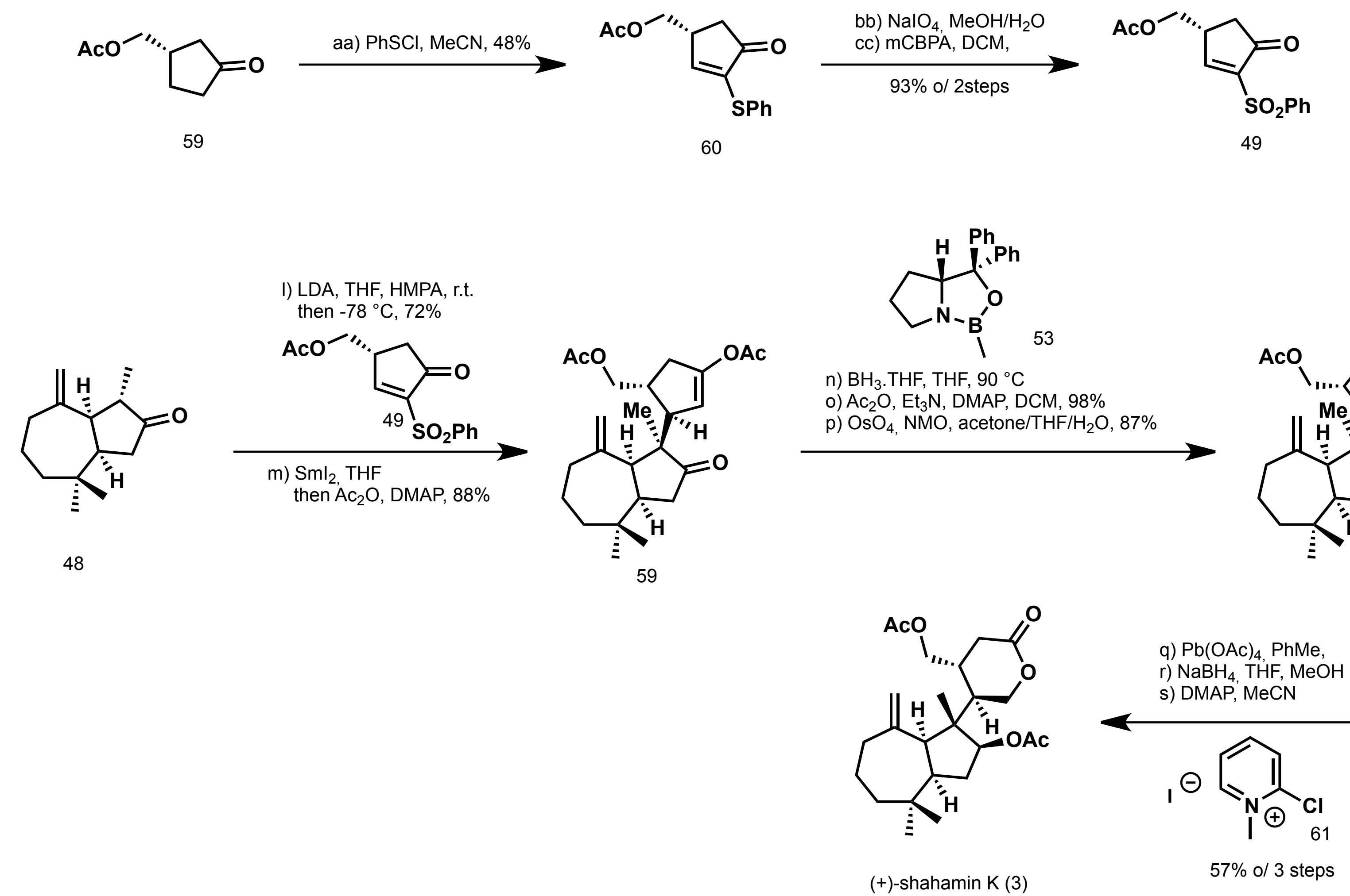
Enantioselective Total Synthesis of Shahamin K

- Lebsack, A.D.; Overman, L.E.; Valentekovich, R.J. *J. Am. Chem. Soc.* **2001**, *123*, 4851-4852



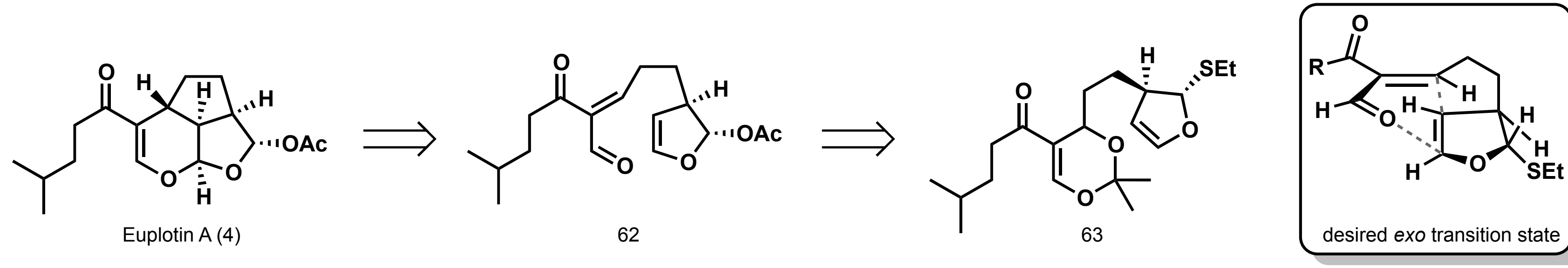


Enantioselective Total Synthesis of Shahamin K

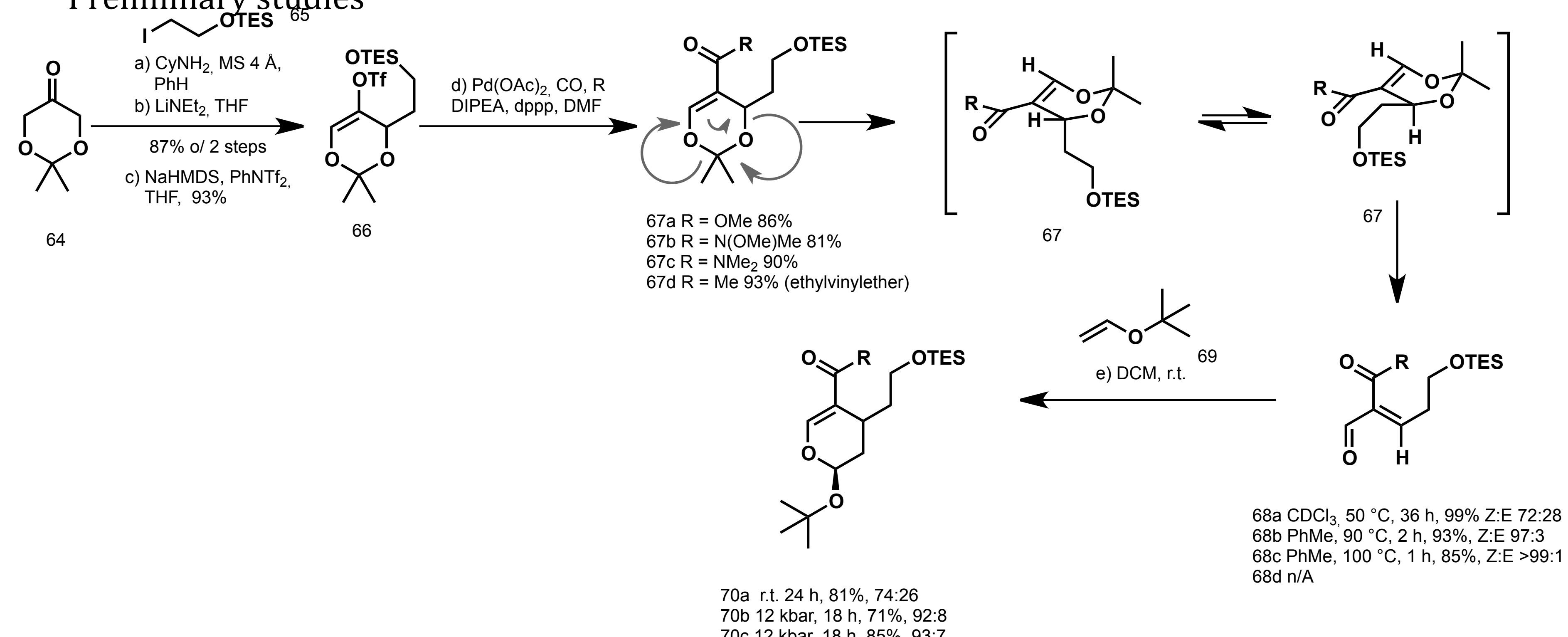


Synthesis of (*Z*)-2-Acyl-2-enals: Application in the Total Synthesis of Cytotoxin (±)-Euplotin

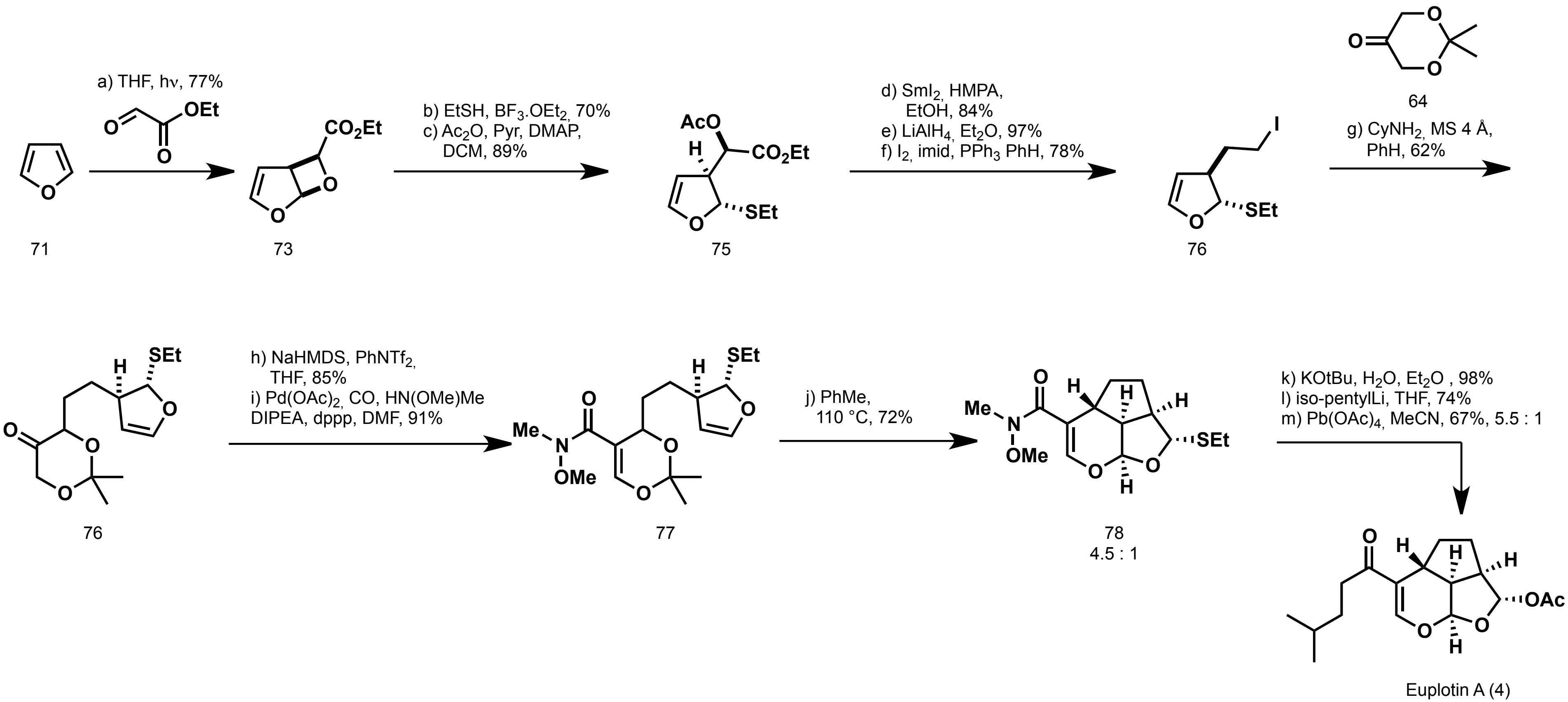
Aungst, R.A.Jr.; Funk, R.L. *J. Am. Chem. Soc.* **2001**, *123*, 9455 – 9456



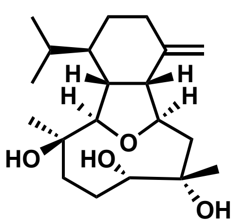
Preliminary studies



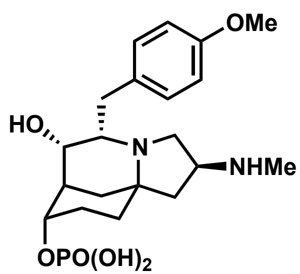
Synthesis of (*Z*)-2-Acyl-2-enals: Application in the Total Synthesis of Cytotoxin (±)-Euplotin



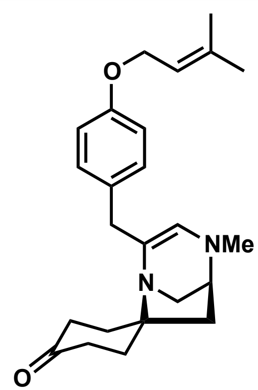
Keysteps



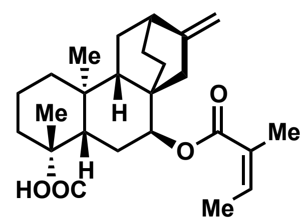
sclerophytin A (79)



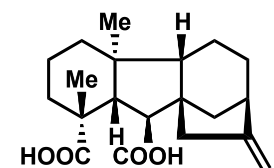
FR901483 (80)



TAN1251C (81)



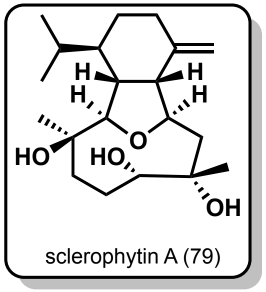
Gummiferolic Acid (82)



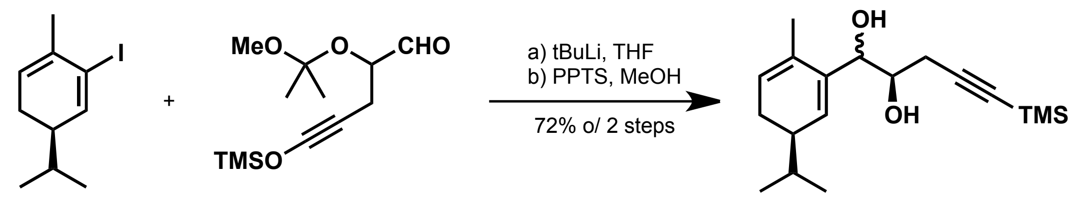
gibberellin A_{12} (83)

General Approach for Cladellin Diterpenes

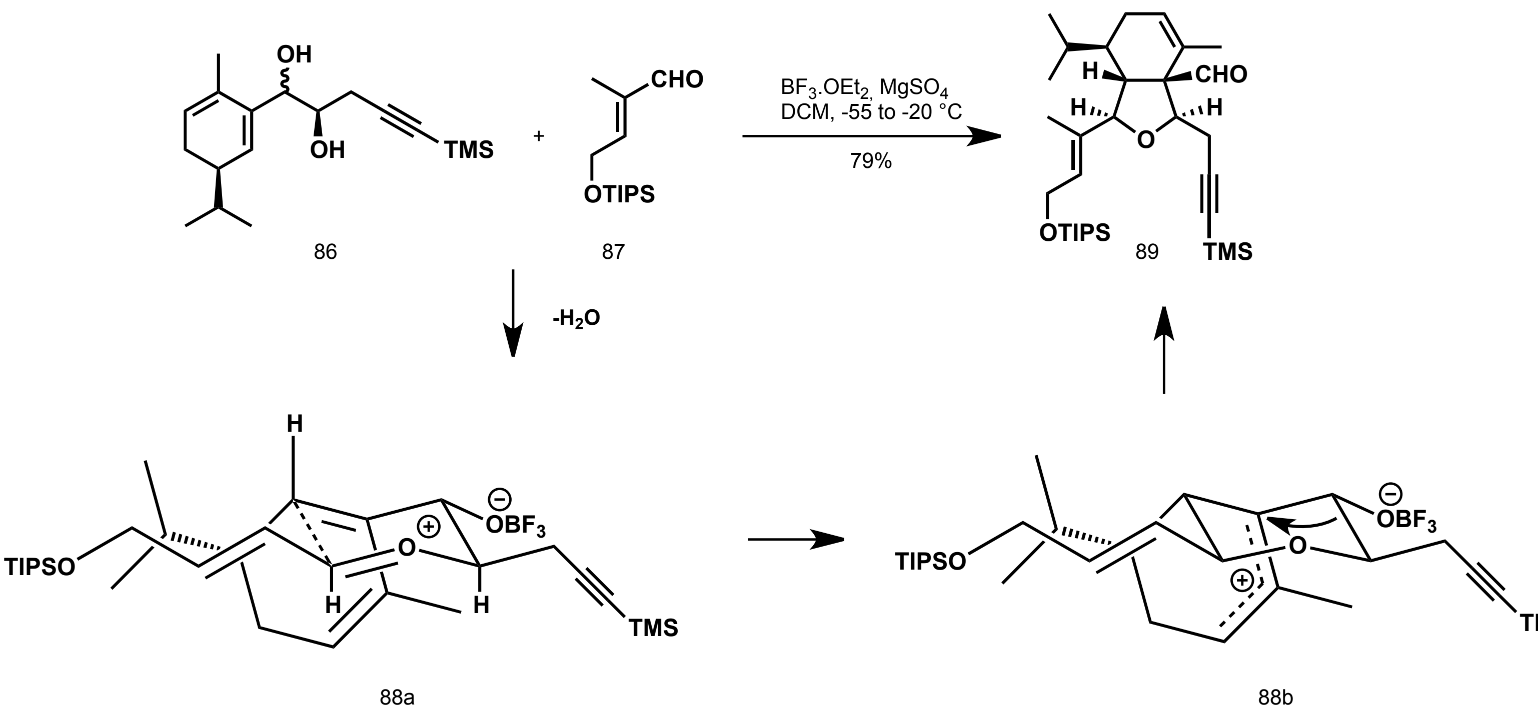
MacMillan, D.W.C.; Overman, L.E.; Pennington, L.D. *J. Am. Chem. Soc.* **2001**, *123*, 9033-9044



sclerophytin A (79)

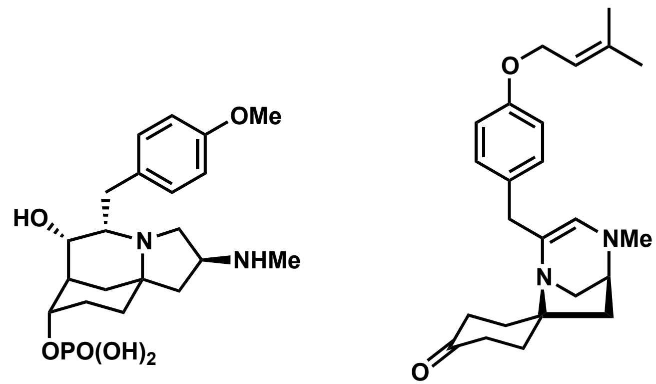


84 85 86



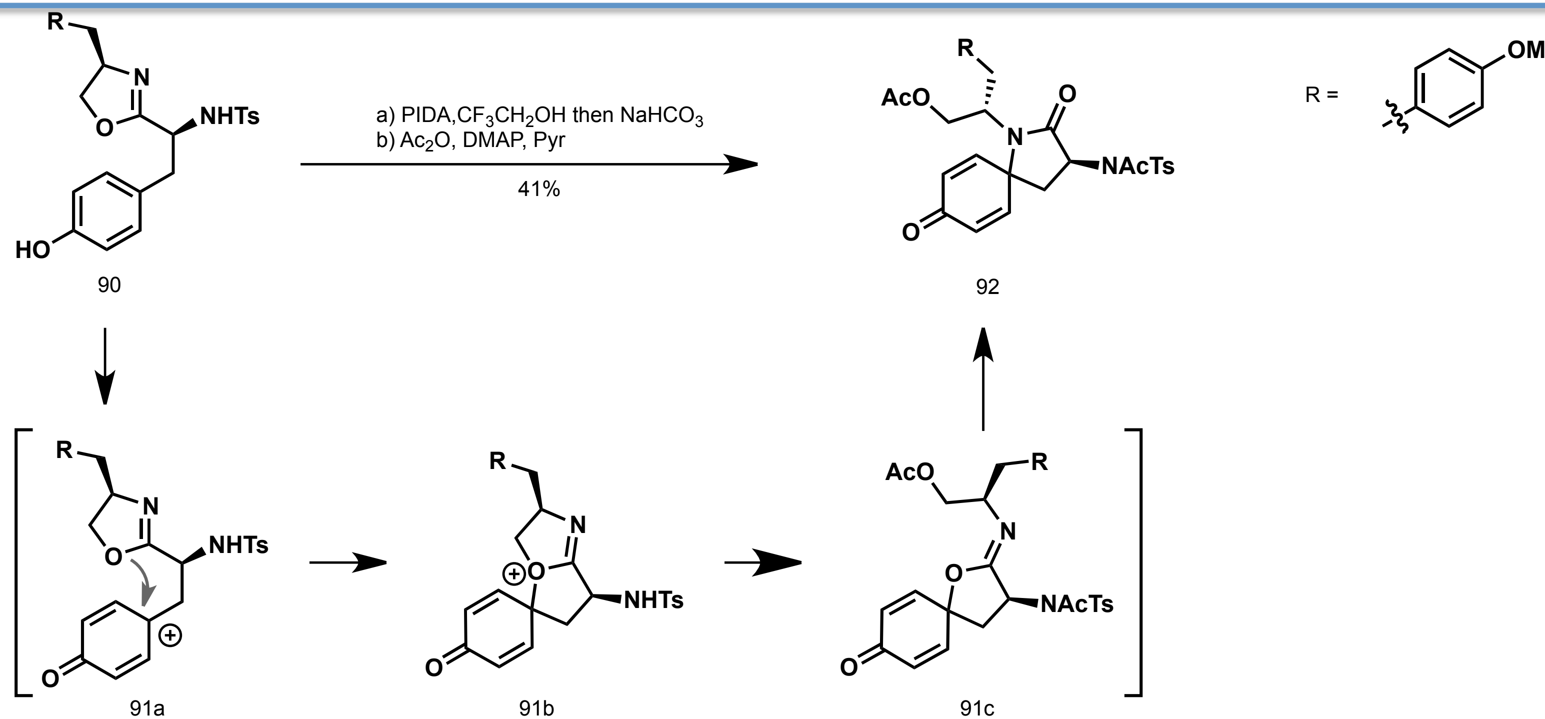
Total Synthesis of Tricyclic Azaspirane Derivatives of Tyrosine: FR901483 and TAN 1251C

- Ousmer, M.; Braun, N.A.; Claude, B.; Perrin, M. Ciufolini, M.A. *J. Am. Chem. Soc.* **2001**, *123*, 7534-7538



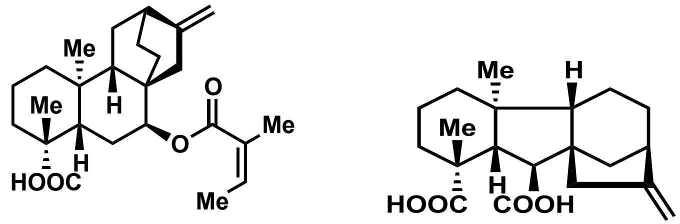
FR901483 (80)

TAN1251C (81)



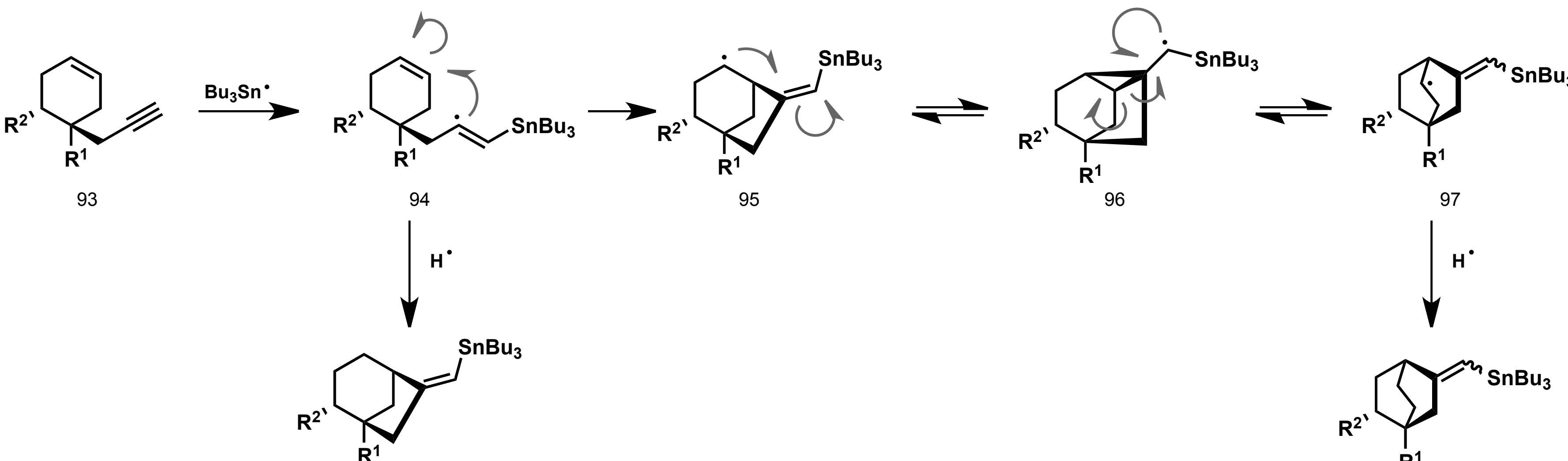
Radical Cyclization of Processes of Cyclic Enynes

- Toyota, M.; Yokota, M.; Ihara, M. *J. Am. Chem. Soc.* **2001**, *123*, 1856 - 1861

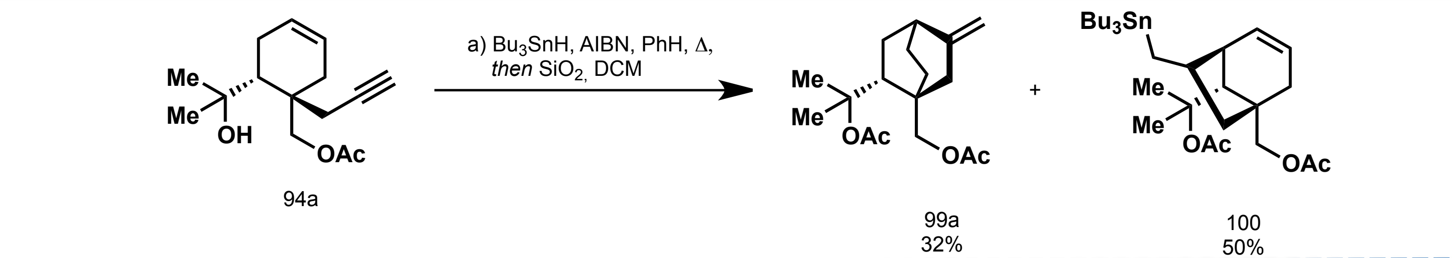


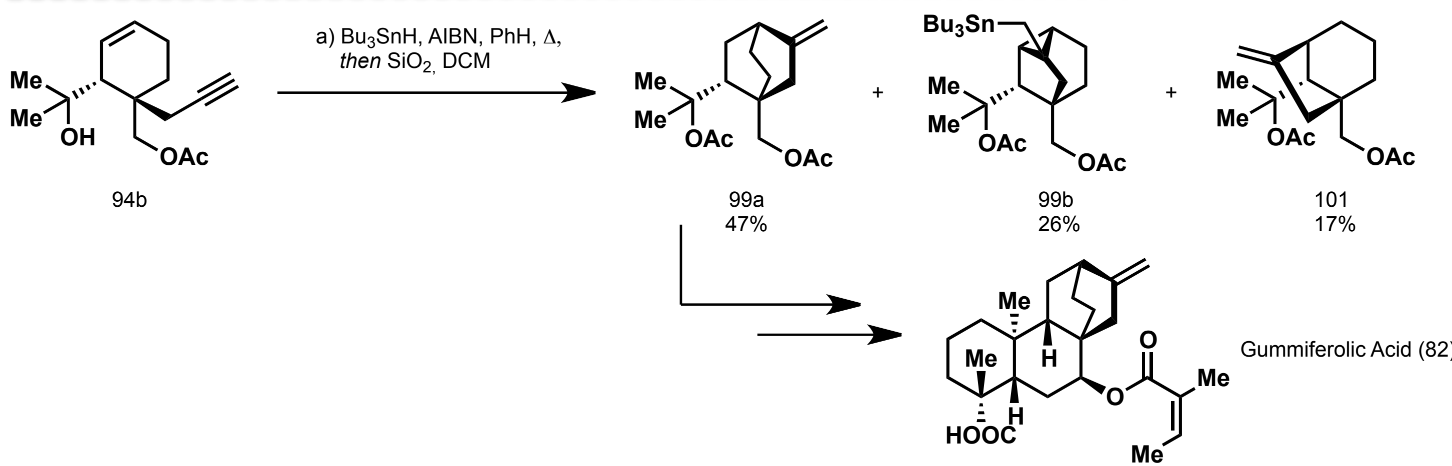
Gummiferolic Acid (82)

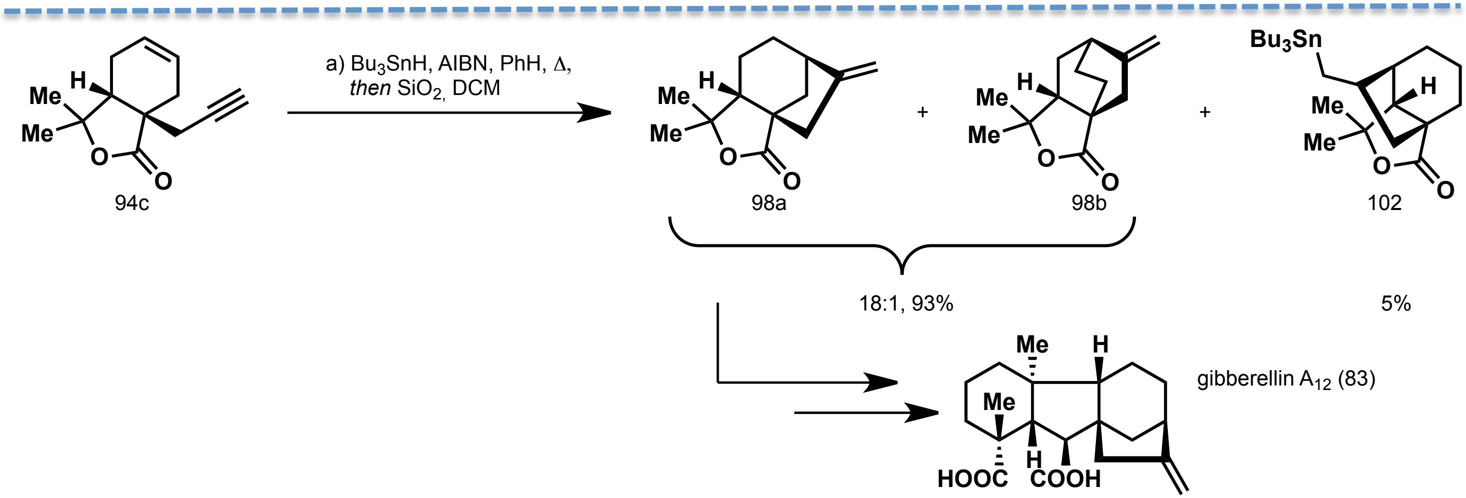
gibberellin A_{12} (83)



Radical Cyclization of Processes of Cyclic Enynes



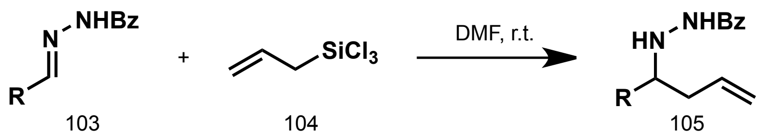




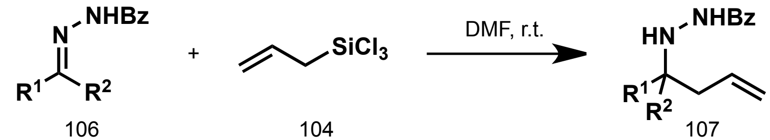
Methodologies

Addition of Allyltrichlorosilanes to Benzoylhydrazones

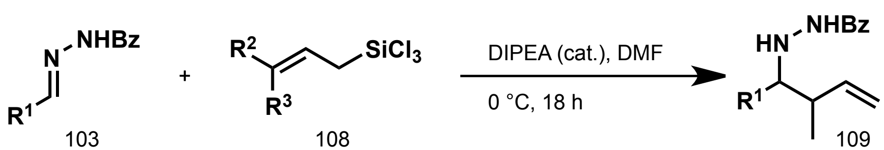
Hirabayashi, R.; Ogawa, C.; Sugiura, M.; Kobayashi, S. *J. Am. Chem. Soc.* **2001**, *123*, 12095-12096



Entry	R	Time [h]	yield
1	Ph	1	96
2	(*E*)-PhCH=CH	1	90
3	$Ph(CH_2)_2$	15	77
4	*n*-pentane	13	76
5	*i*-Bu	1	73
6	Cyc-Hex	15	74
7	*t*-Bu	7	77



Entry	R^1	R^2	Time [h]	yield
1	Ph	Me	3	95
2	*P*-MeOPh	Me	3	95
3	*M*-NO_2Ph	Me	3	90
4	2-naphthyl	Me	3	96
5	Me	Me	3	60
6	$Ph(CH_2)_2$	Me	0.3	81
7	$-(CH_2)_5-$		2	62
8	Ph	*n*Pr	3	87



Z : R^2 = H, R^3 = Me
E : R^2 = Me, R^3 = H

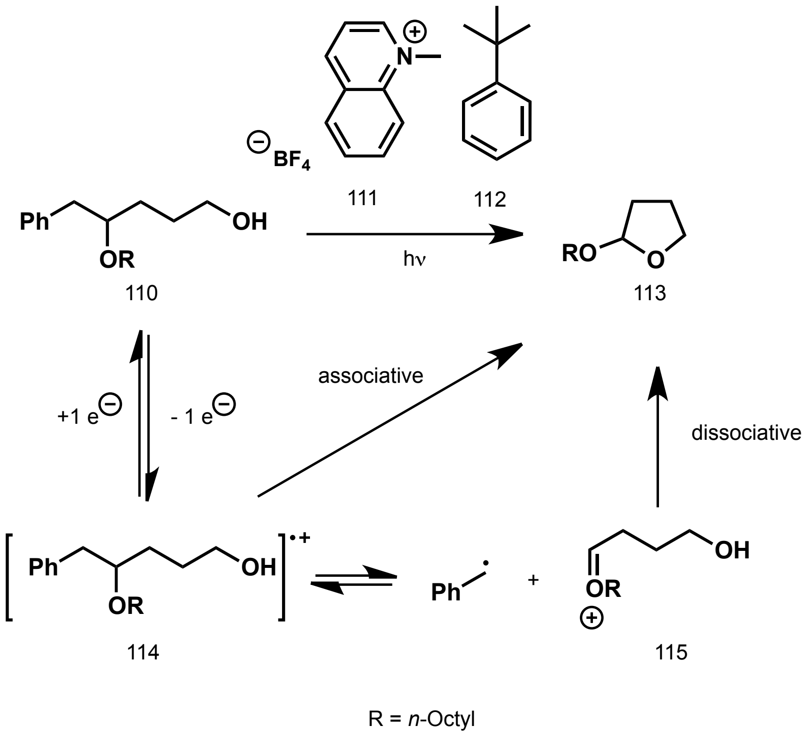
Entry	R^1	Crotylsilane	yield	syn/*anti*
1	Ph	*Z*	79	1/99
2	Ph	*E*	59	78/22
3	(*E*)-PhCH=CH	*Z*	80	3/97
4	(*E*)-PhCH=CH	*E*	82	95/5
5	$Ph(CH_2)_2$	*Z*	65	9/91
6	$Ph(CH_2)_2$	*E*	66	92/8

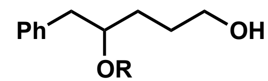
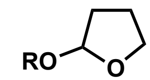
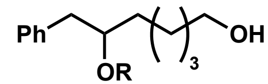
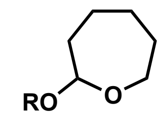
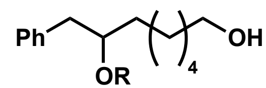
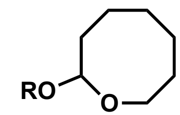
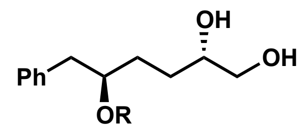
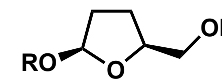
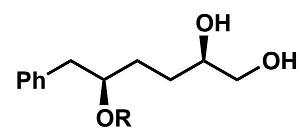
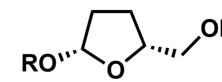
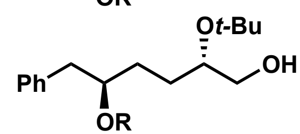
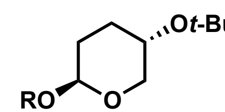
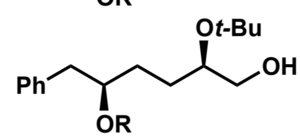
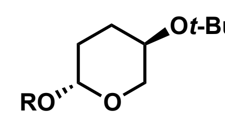
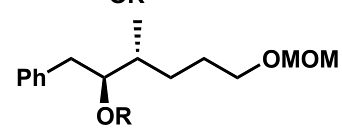
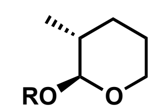
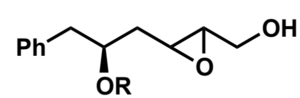
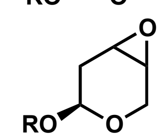
Entry	R^1	Crotylsilane	yield	syn/*anti*
7	*n*-pentane	*Z*	65	7/93
8	*n*-pentane	*E*	67	93/7
9	*i*-Bu	*Z*	65	7/93
10	*i*-Bu	*E*	68	94/6
11	Cyc-Hex	*Z*	61	5/95
12	Cyc-Hex	*E*	48	55/45

Electron Transfer Initiated Cyclizations

Kumar, V.S.; Floreancig, P.E.

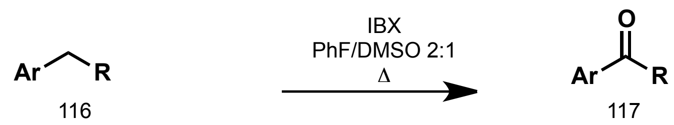
J. Am. Chem. Soc. **2001**, *123*, 3842 - 3843



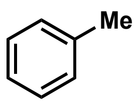
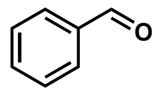
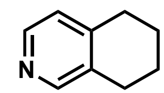
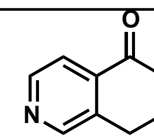
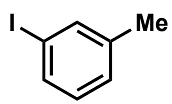
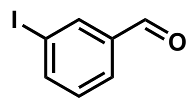
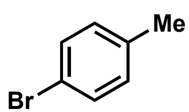
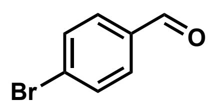
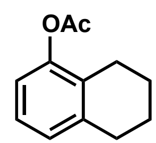
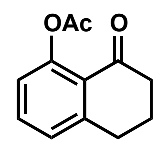
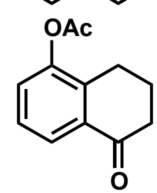
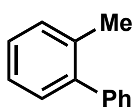
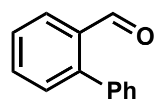
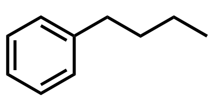
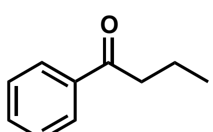
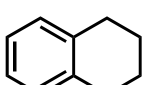
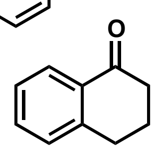
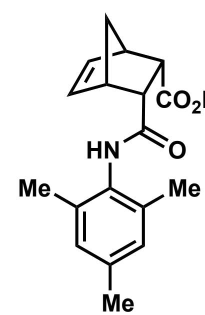
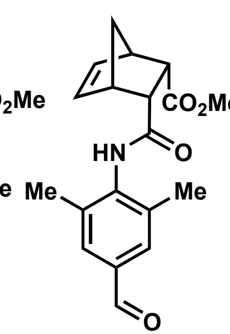
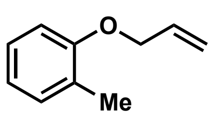
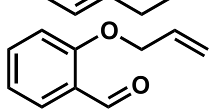
entry	Substrate	Product	Yield [%]	*dr*
1			74	
2			82	
3			55	
4			74	10:1
5			92	1.2:1
6			84	1.7:1
7			82	1.1:1
8			67	1.4:1
9			78	2.6:1

Selective Oxidation at Carbon Adjacent to Aromatic Systems with IBX

Nicolaou, K.C.; Baran, P.S.; Zhong, Y-L. *J. Am. Chem. Soc.* **2001**, *123,* 3183 - 3185



R= Alkyl, H
Substituents on Aryl: Halogenes, Aryl, Alkyl, MeO, Amines

Entry	Substrate	Product	Conditons	Yield [%]	Entry	Substrate	Product	Conditions	Yield [%]
1			12 h/85 °C 3.0 equiv	85	8			24 h/85 °C 3.0 equiv	70
2			16 h/85 °C 4.0 equiv	72					
3			16 h/80 °C 3.0 equiv	73	9		 	24 h/85 °C 3.0 equiv 1:1	70
4			24 h/90 °C 3.0 equiv	78					
5			8 h/80 °C 3.0 equiv	72					
6			12 h/80 °C 3.0 equiv	70	10			24 h/90 °C 3.0 equiv	75
7			12 h/80 °C 3.0 equiv	80					