Universität Konstanz

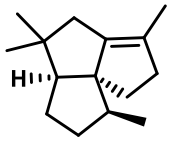
Organic Letters 2014 & 2015

Heiko Rebmann

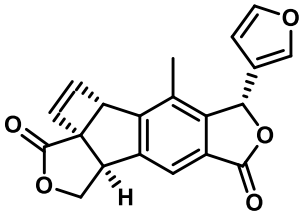
04.10.2017

Organic LETTERS

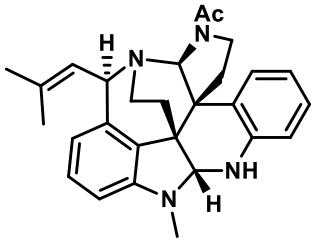
Overview – Total syntheses



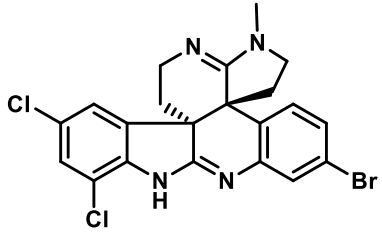
(±)-Panaginsene
revised structure
Lee



(±)-Salvileucalin C
Ding

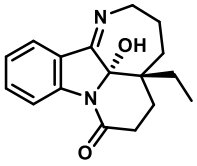


(±)-Communesin F
formal synthesis
Stoltz

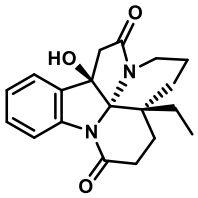


(±)-Perophoramidine
formal synthesis
Stoltz

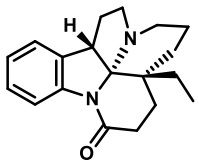
Dai:



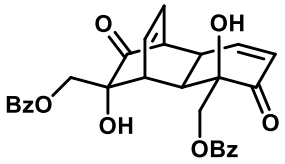
(±)-Mersicarpine



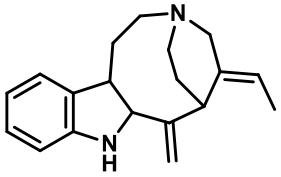
(±)-Leuconodine B



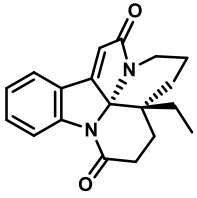
(±)-Leuconodine D



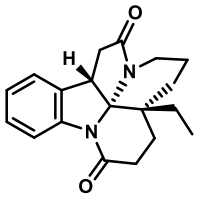
(±)-Grandifloracin
Stoltz



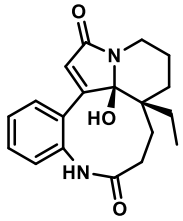
(±)-Subincanadine E
Zhai



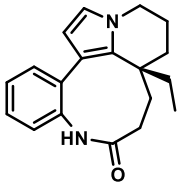
(±)-Melodinine E



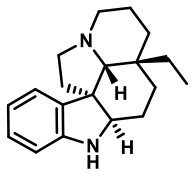
(±)-Leuconoxine



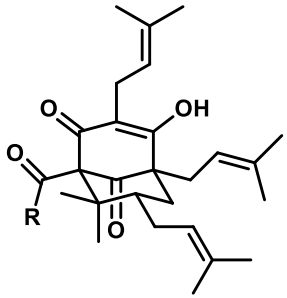
(±)-Leuconolam



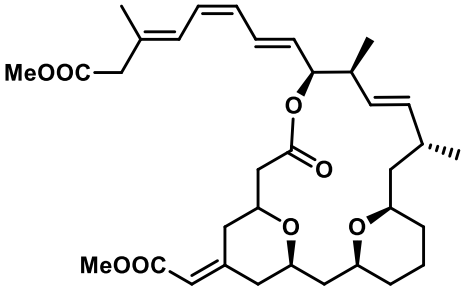
(±)-Rhazinilam



(±)-Aspidospermidine
formal synthesis

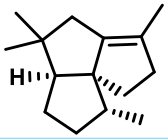


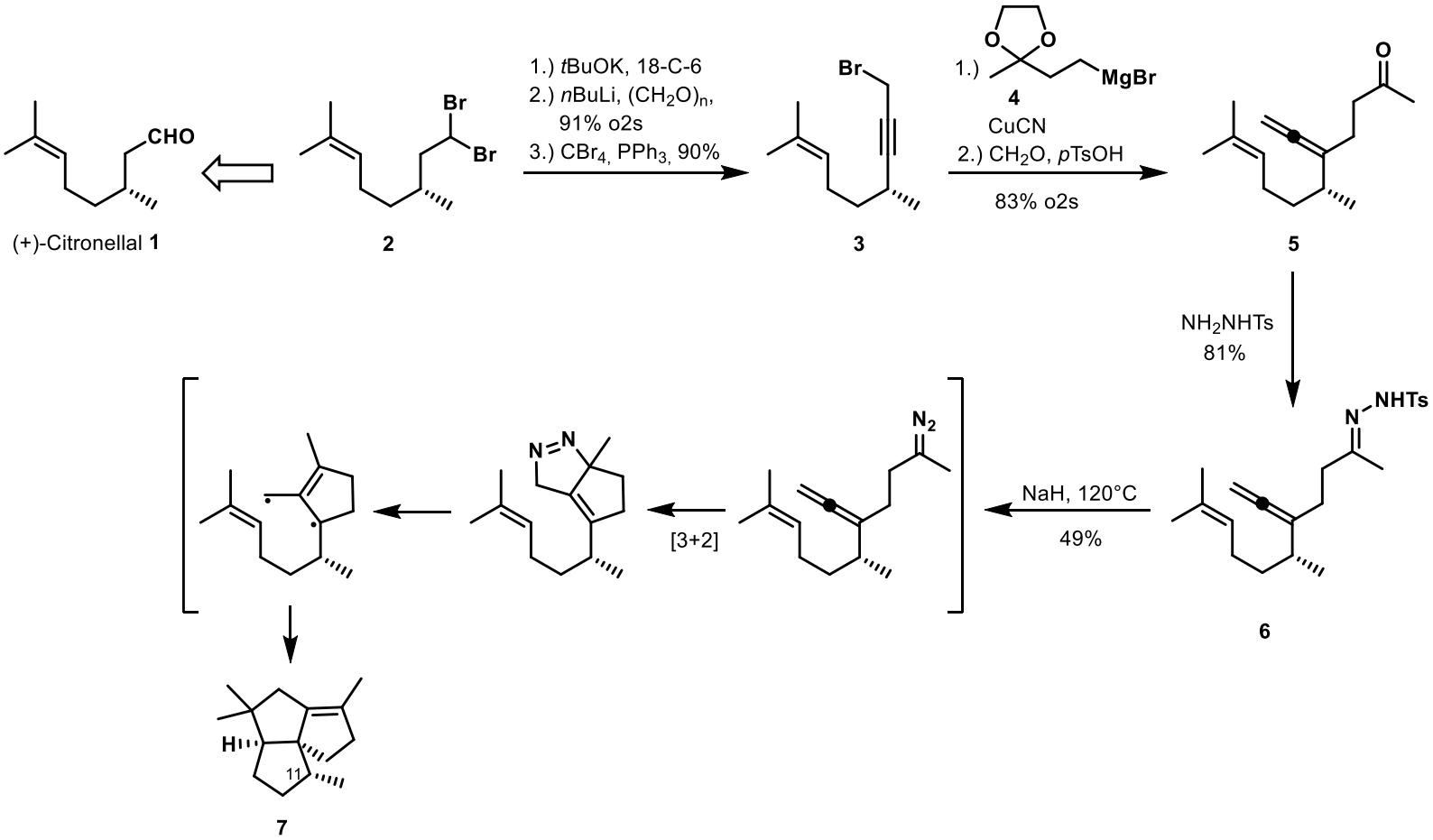
R = Ph (**–**)-Nemorosone
R = *i*Pr (+)-Secohyperforin
Shair



(**–**)-Exiguolide
Song

Panaginsene





(+)-Citronellal **1** **2** **3** **4** **5** **6** **7**

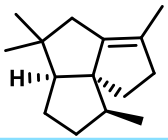
proposed structure of Panaginsene

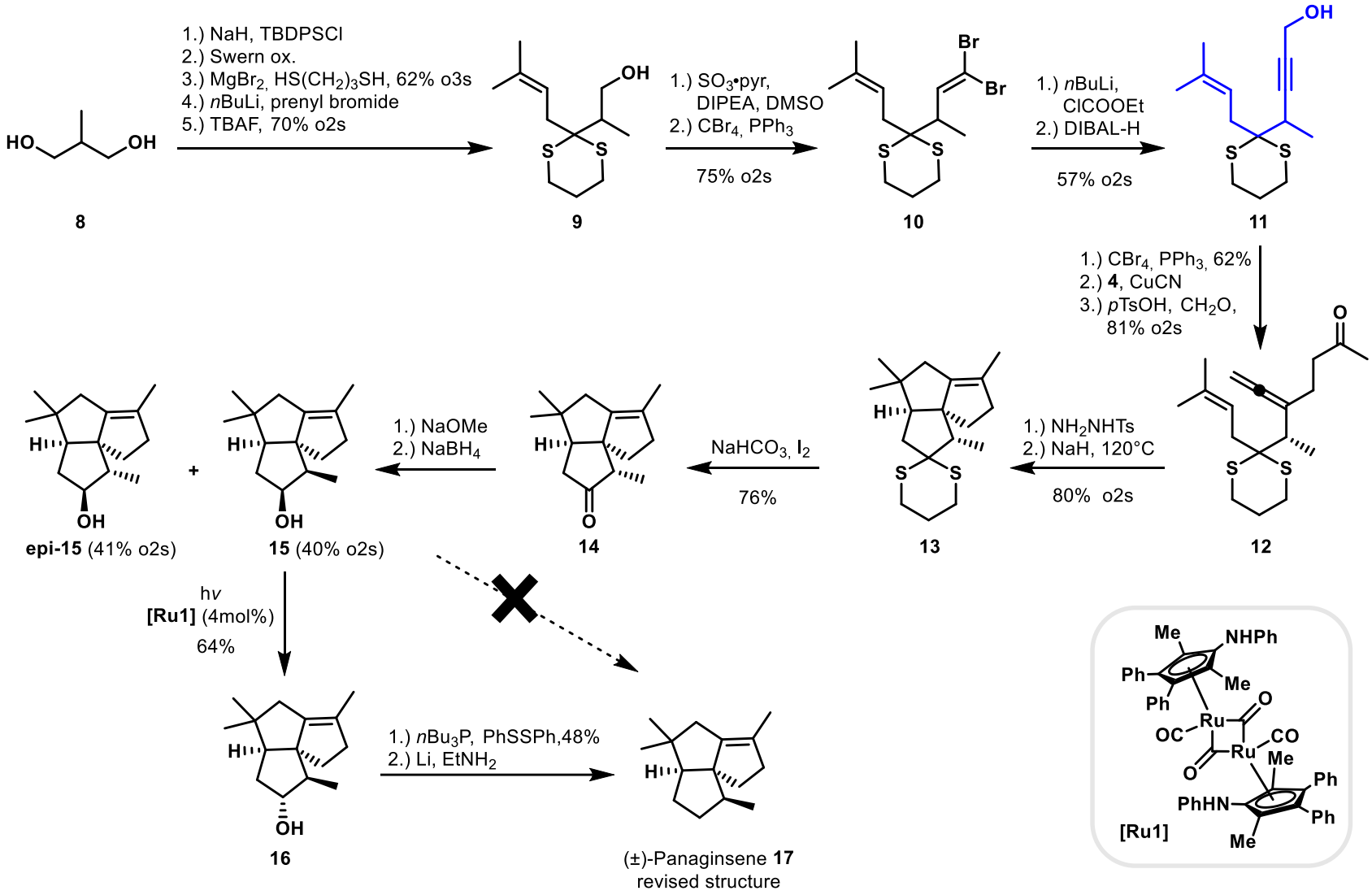
BUT: spectral data not identical with natural product

→ 11-*epi*-Panaginsene

Geum, S.; Lee, H.-Y., *Org. Lett.* **2014,** *16* (9), 2466-2469.

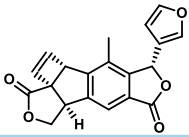
Panaginsene

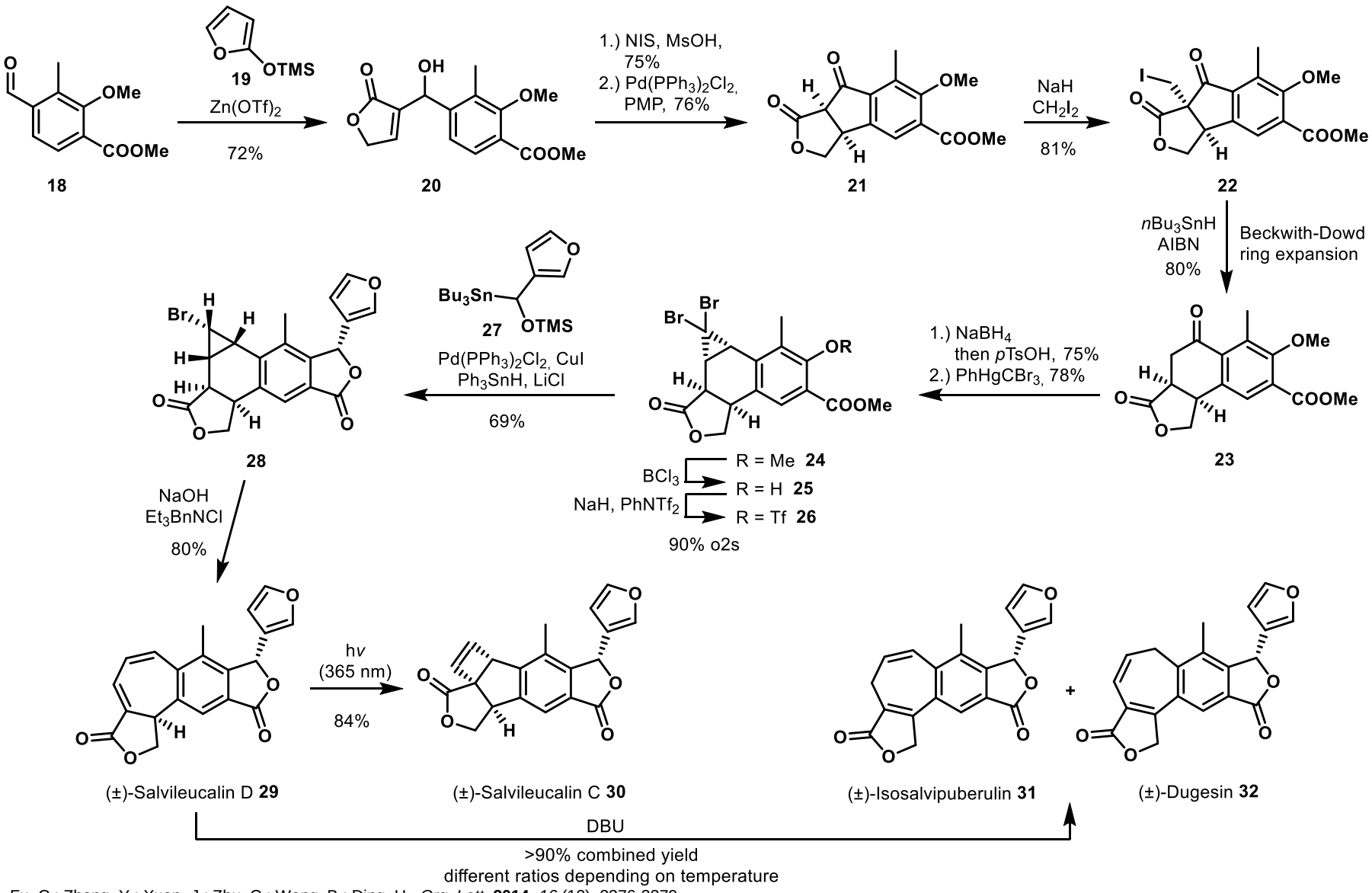




Geum, S.; Lee, H.-Y., *Org. Lett.* **2014,** *16* (9), 2466-2469.

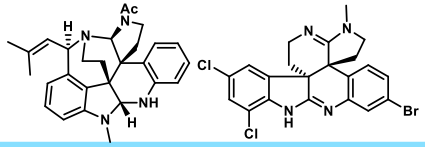
(±)-Salvileucalin C

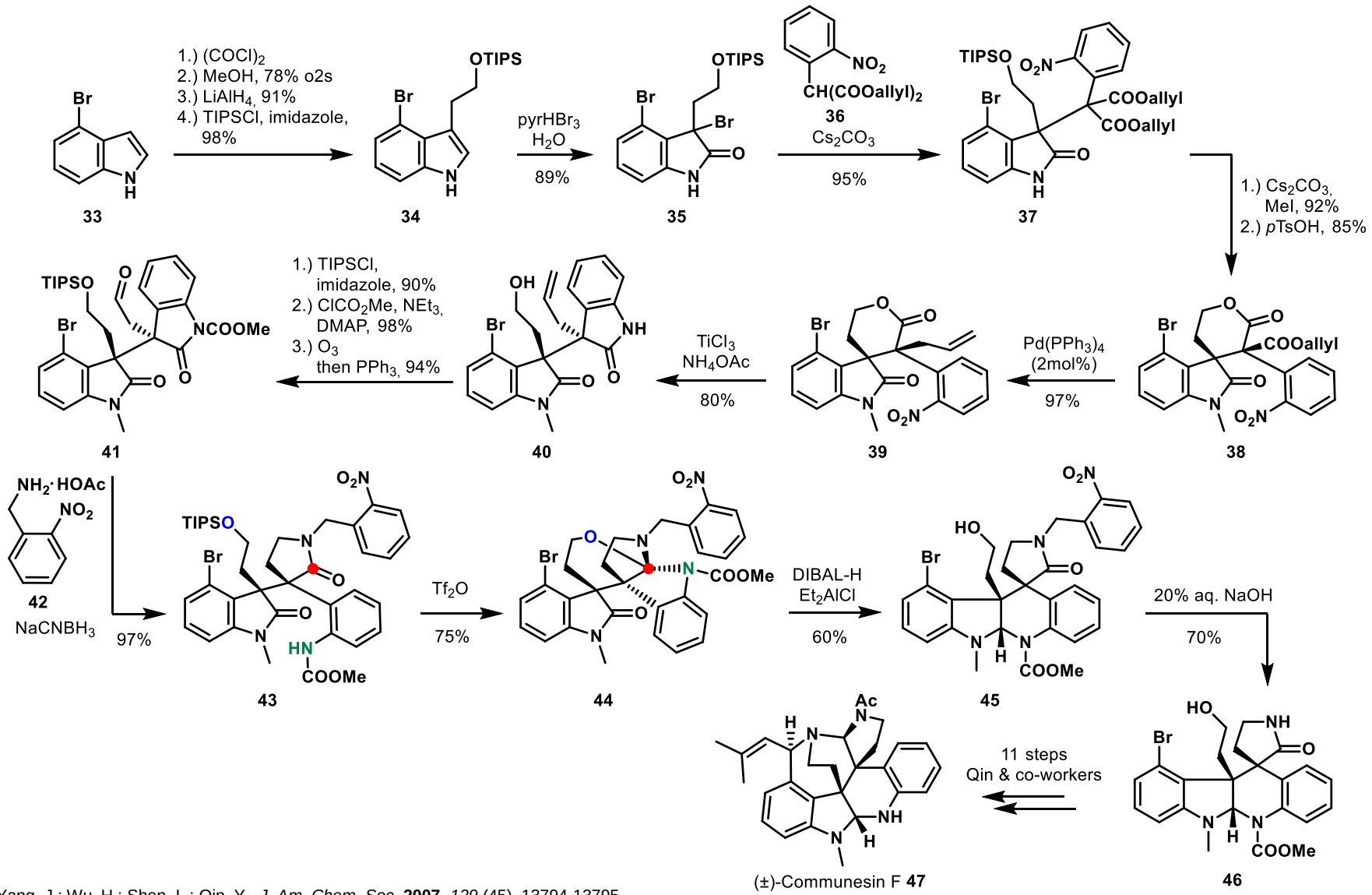




Fu, C.; Zhang, Y.; Xuan, J.; Zhu, C.; Wang, B.; Ding, H., *Org. Lett.* **2014,** *16* (12), 3376-3379.

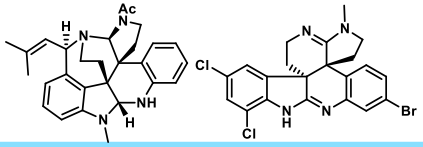
(±)-Communesin F & (±)-Perophoramidine

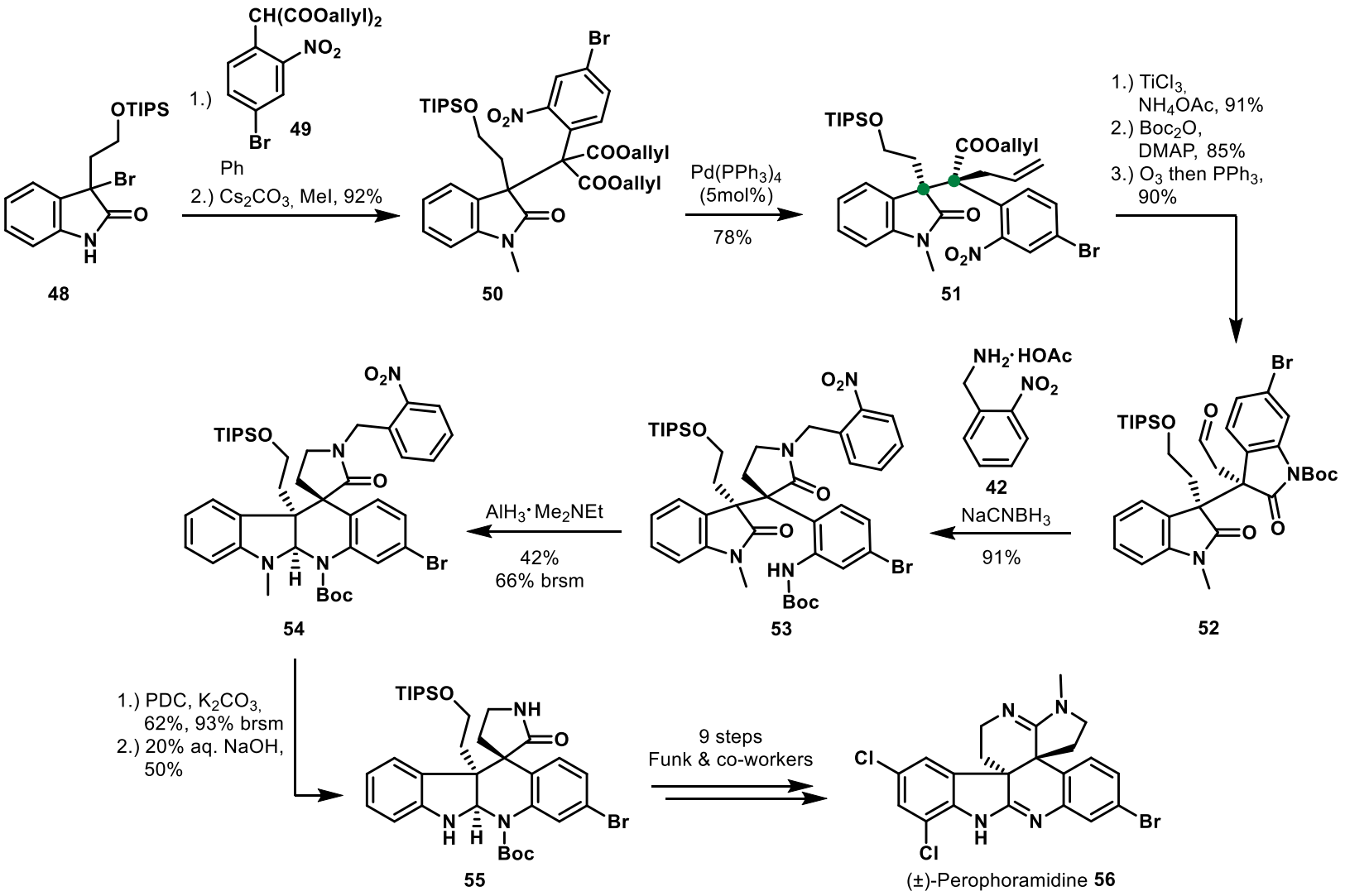




Yang, J.; Wu, H.; Shen, L.; Qin, Y., *J. Am. Chem. Soc.* **2007,** *129* (45), 13794-13795.
Han, S.-J.; Vogt, F.; Krishnan, S.; May, J. A.; Gatti, M.; Virgil, S. C.; Stoltz, B. M., *Org. Lett.* **2014,** *16* (12), 3316-3319.

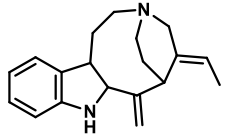
(±)-Communesin F & (±)-Perophoramidine

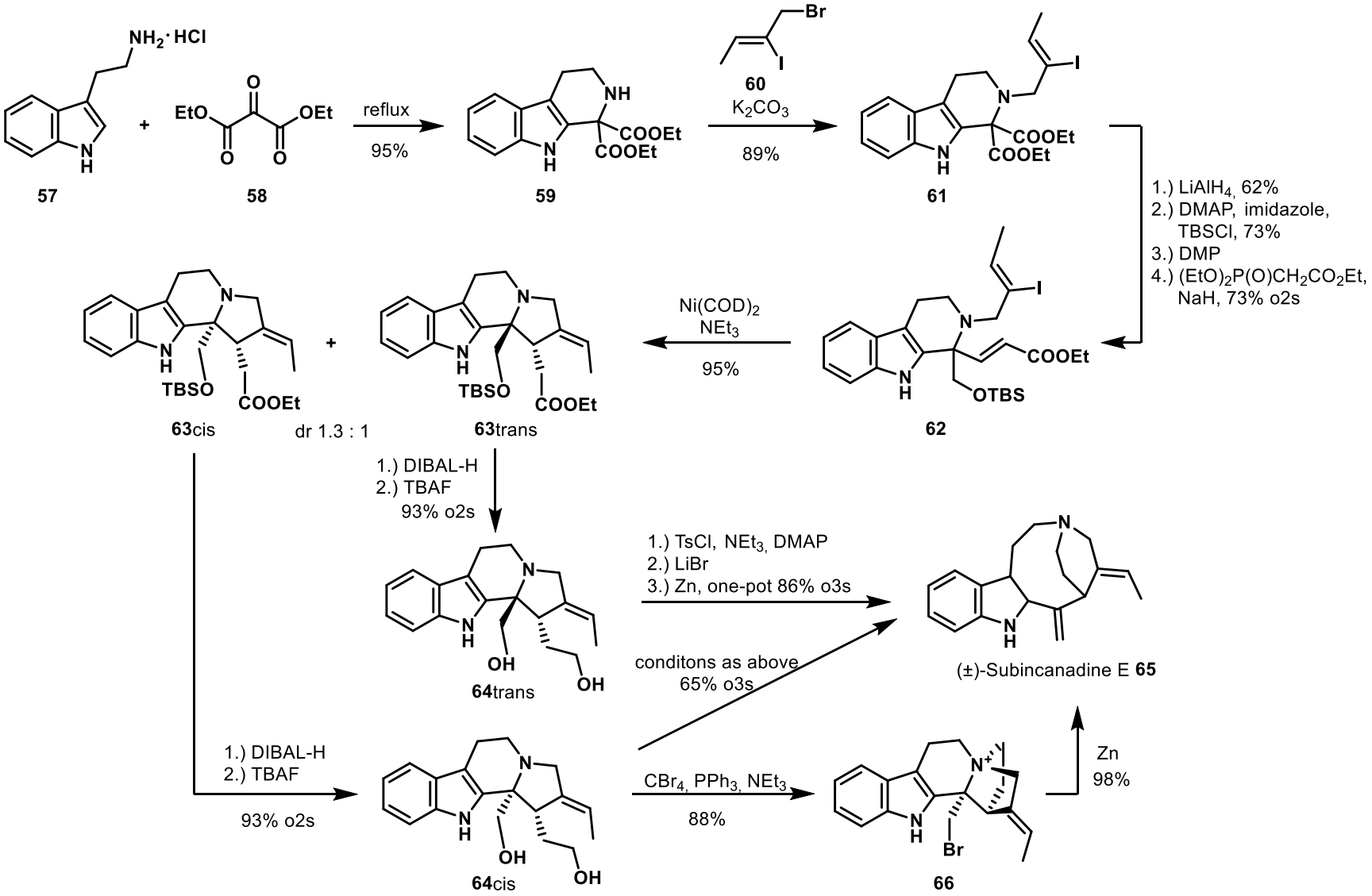




Fuchs, J. R.; Funk, R. L., *J. Am. Chem. Soc.* **2004,** *126* (16), 5068-5069.
Han, S.-J.; Vogt, F.; Krishnan, S.; May, J. A.; Gatti, M.; Virgil, S. C.; Stoltz, B. M., *Org. Lett.* **2014,** *16* (12), 3316-3319.

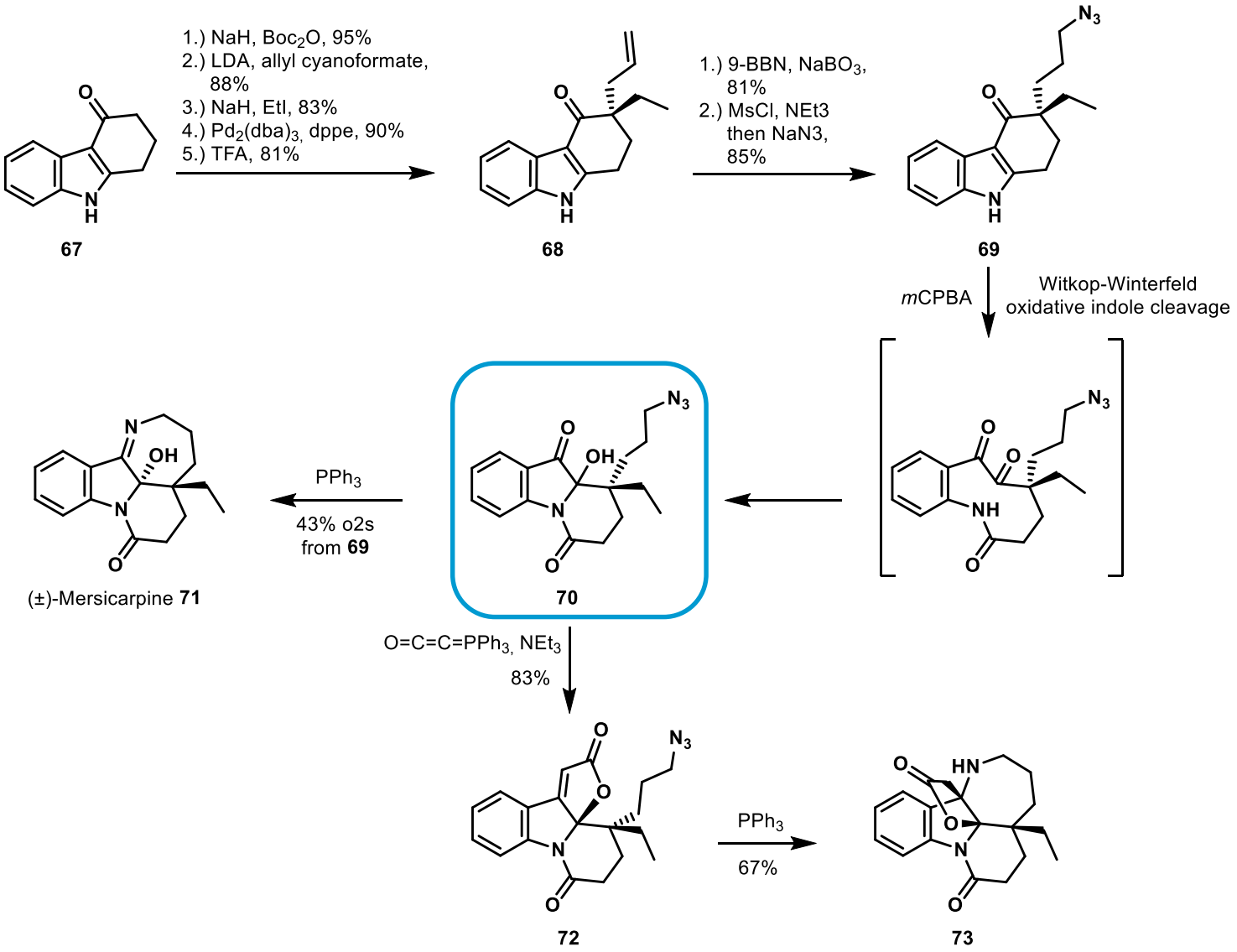
(±)-Subincanadine E





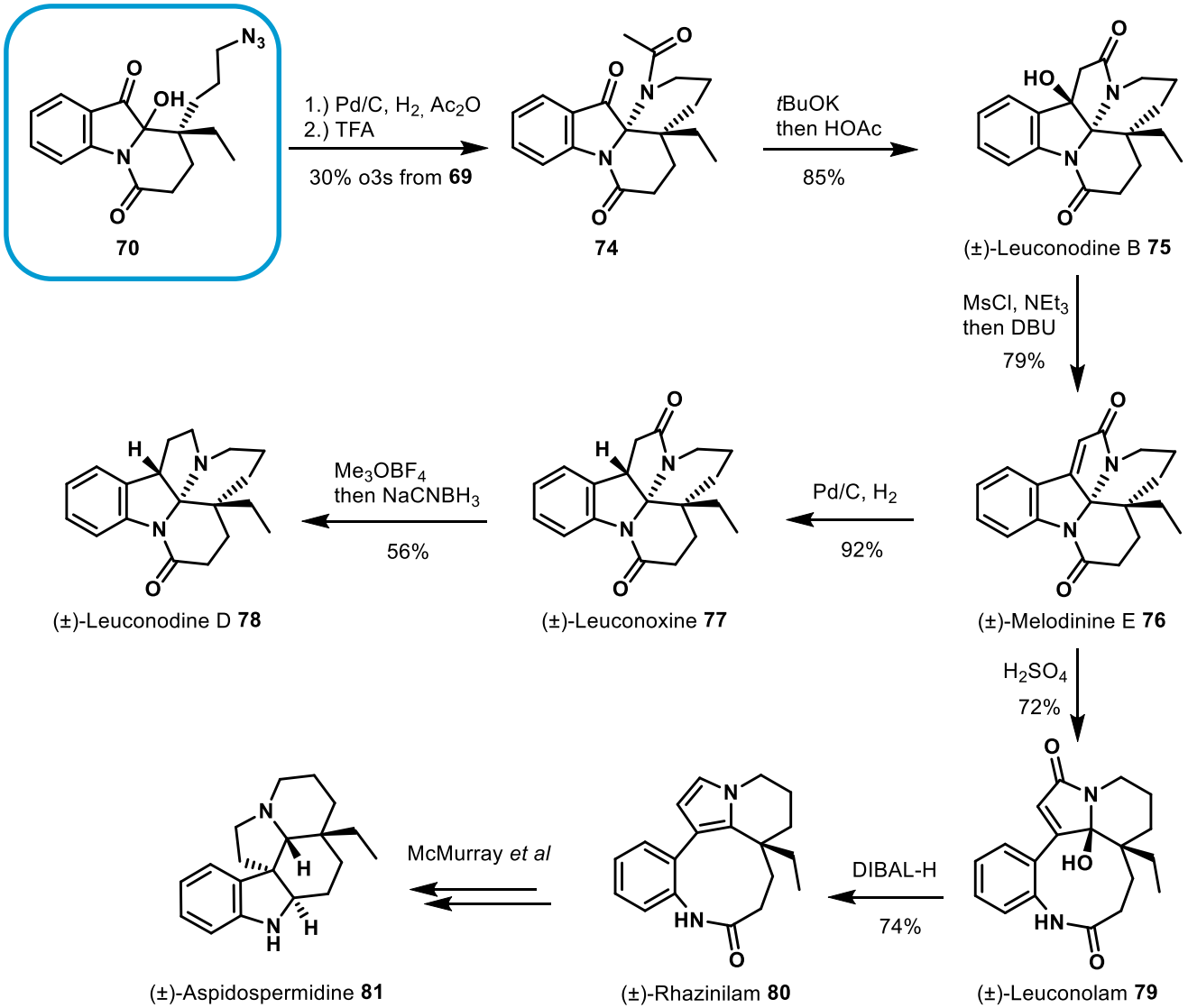
Tian, J.; Du, Q.; Guo, R.; Li, Y.; Cheng, B.; Zhai, H., *Org. Lett.* **2014,** *16* (12), 3173-3175.

Monoterpene indole alkaloids via functional-group-pairing strategy



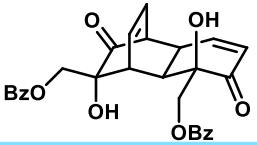
Yang, Y.; Bai, Y.; Sun, S.; Dai, M., *Org. Lett.* **2014,** *16* (23), 6216-6219.

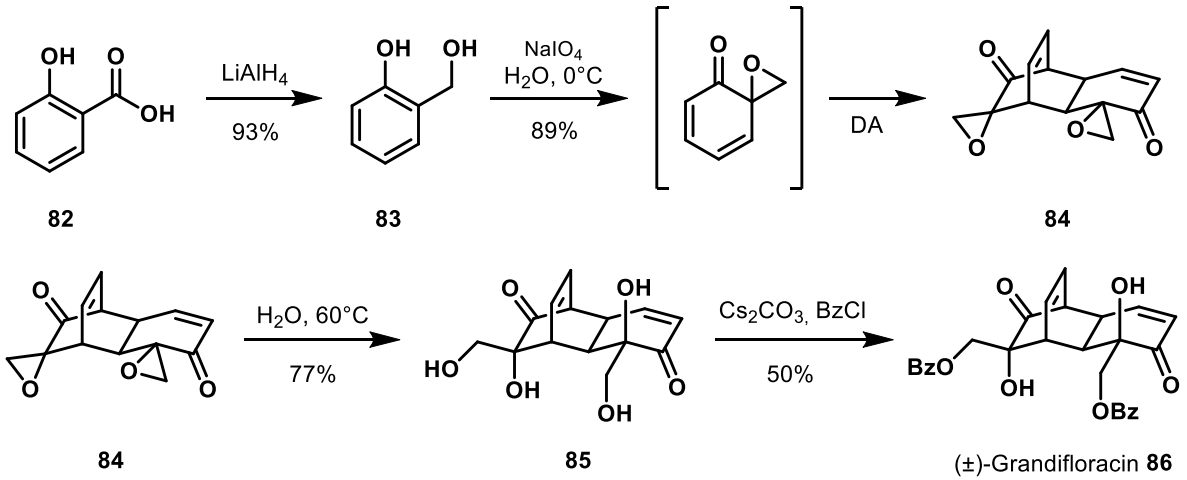
Monoterpene indole alkaloids via functional-group-pairing strategy



Yang, Y.; Bai, Y.; Sun, S.; Dai, M., *Org. Lett.* **2014,** *16* (23), 6216-6219.
McMurray, L.; Beck, E. M.; Gaunt, M. J., *Angew. Chem. Int. Ed.* **2012,** *51* (37), 9288-9291.

(±)-Grandifloracin

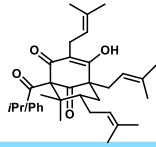


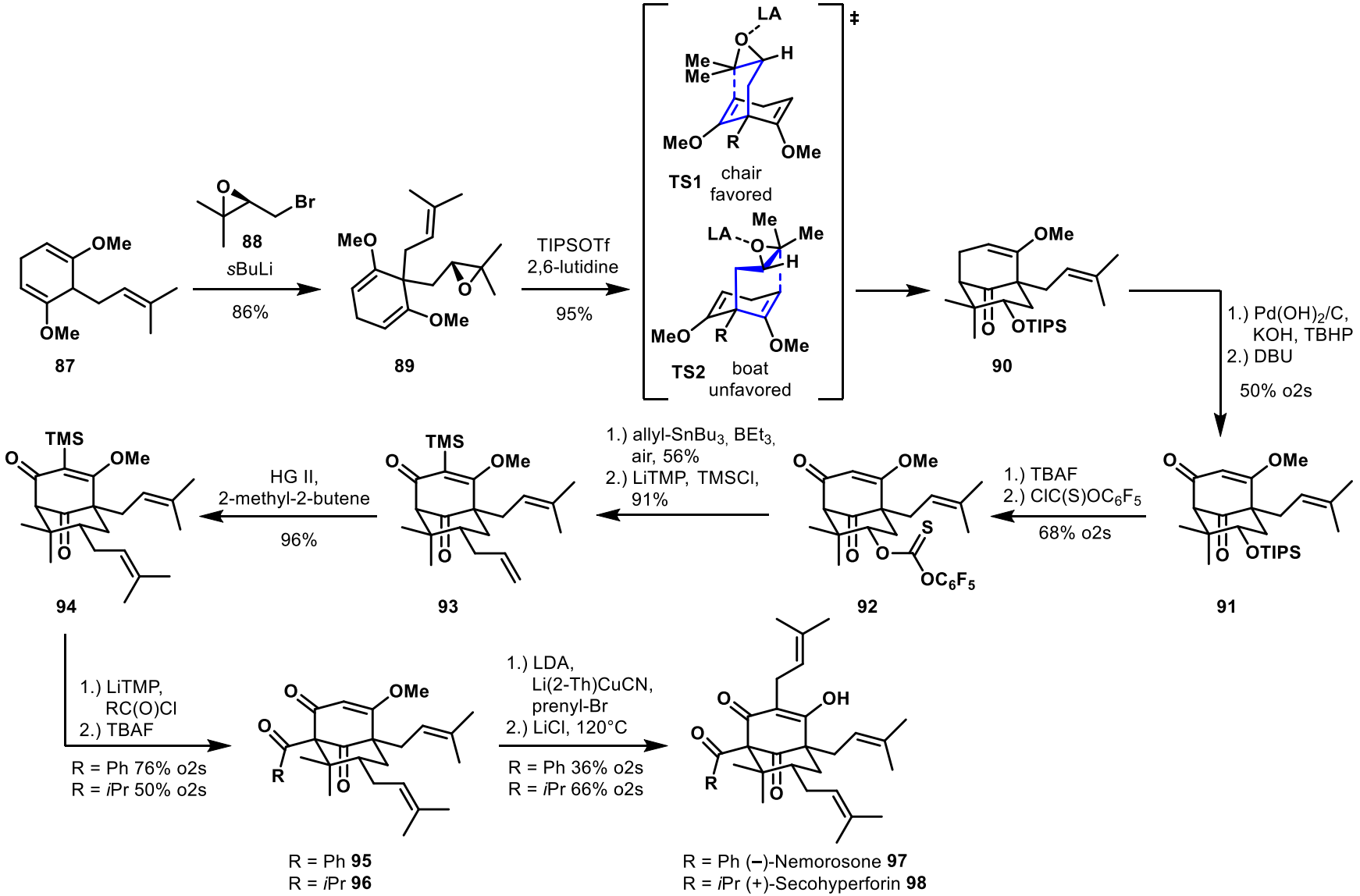


also possible in telescoped synthesis with only one purification in 52% overall yield

Bergner, M.; Duquette, D. C.; Chio, L.; Stoltz, B. M., *Org. Lett.* **2015,** *17* (12), 3008-3010.

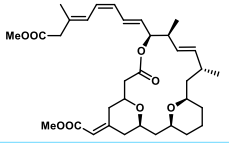
(–)-Nemorosone & (+)-Secohyperforin

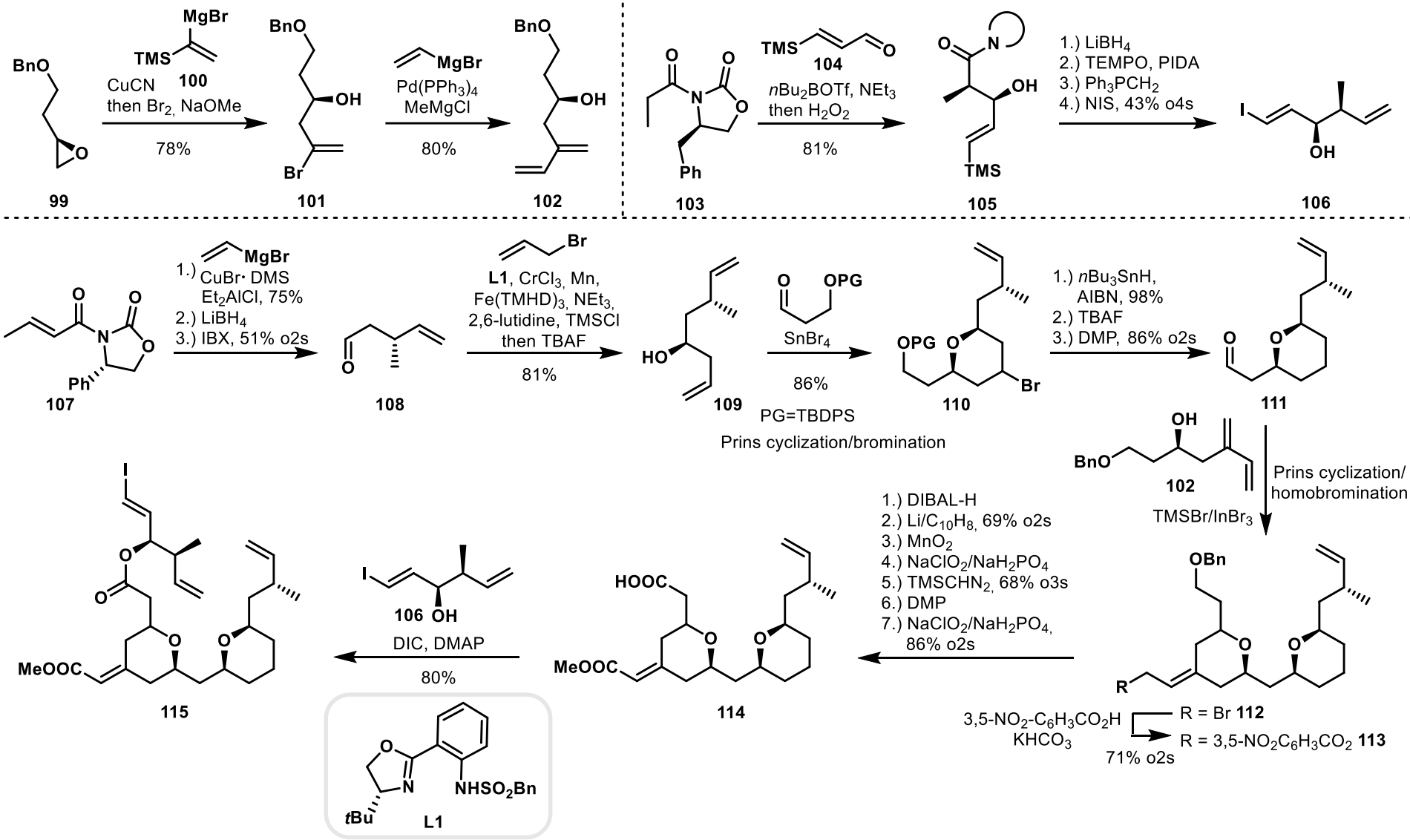




Sparling, B. A.; Tucker, J. K.; Moebius, D. C.; Shair, M. D., *Org. Lett.* **2015,** *17* (14), 3398-3401.

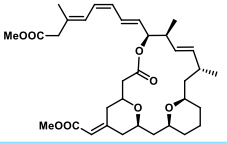
(–)-Exiguolide

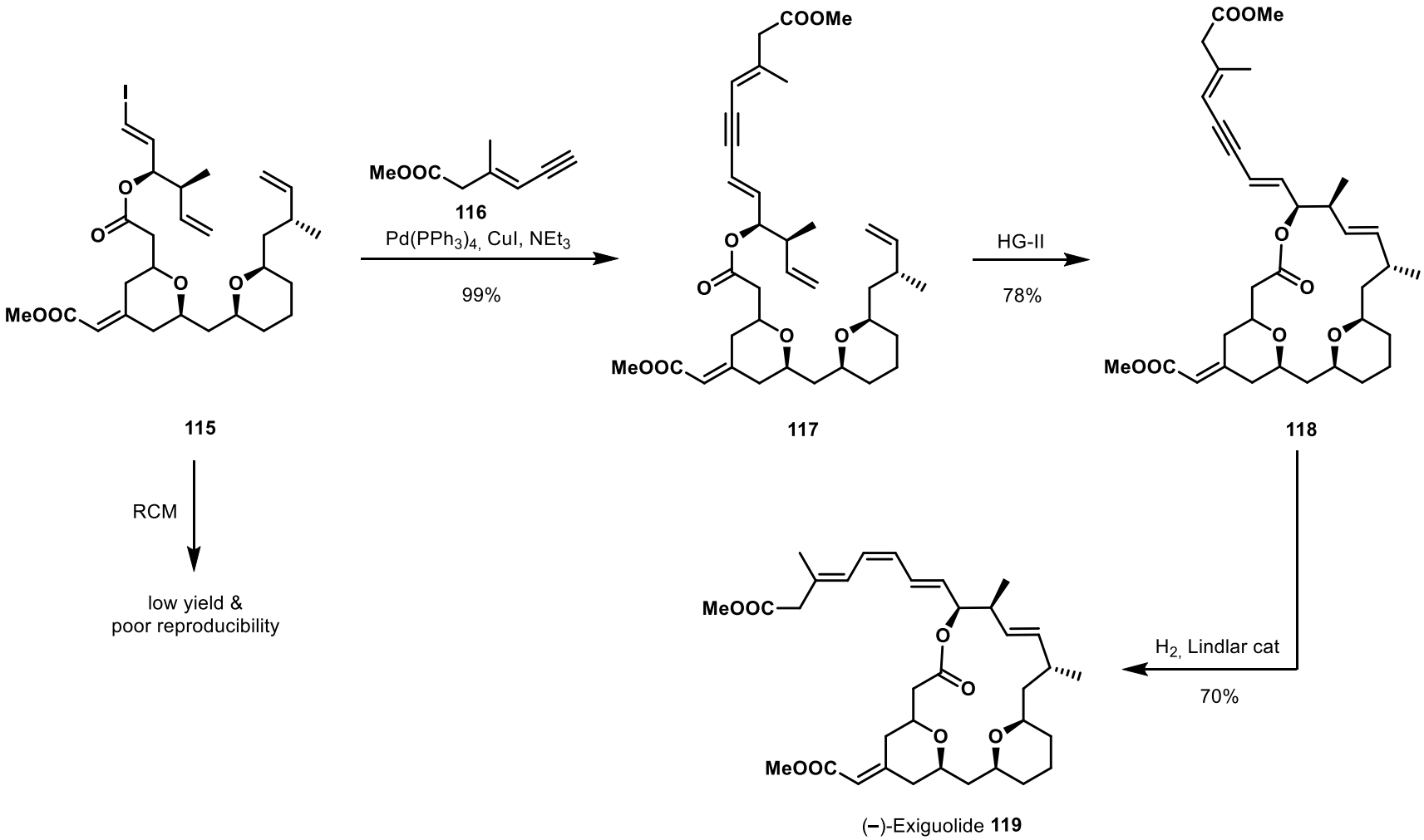




Zhang, Z.; Xie, H.; Li, H.; Gao, L.; Song, Z., *Org. Lett.* **2015,** *17* (19), 4706-4709.

(–)-Exiguolide

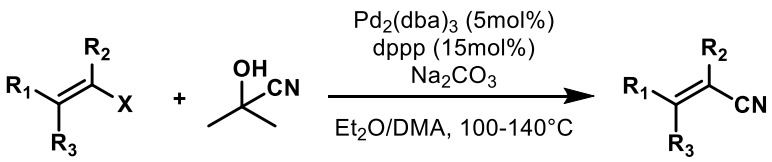




Zhang, Z.; Xie, H.; Li, H.; Gao, L.; Song, Z., *Org. Lett.* **2015,** *17* (19), 4706-4709.

Methodology

Chemoselective Palladium-Catalyzed Cyanation of Vinyl Halides



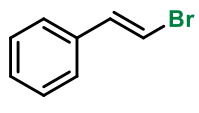
X = I, Br, Cl

R_1, R_2, R_3 = aryl, alkyl, H

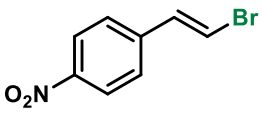
14 examples

dppp = $Ph_2P(CH_2)_3PPh_2$

substrates



116 79%



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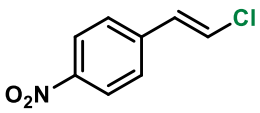

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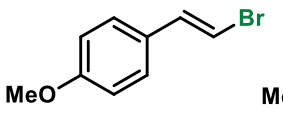

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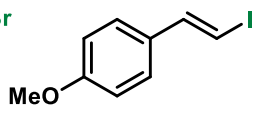

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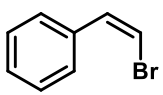

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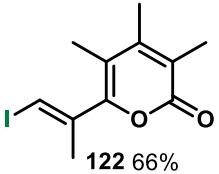

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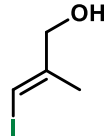

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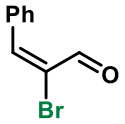

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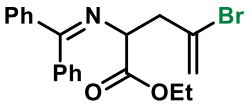

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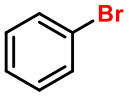
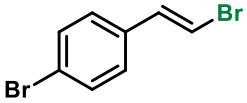


126 no reaction



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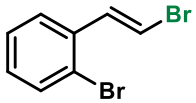
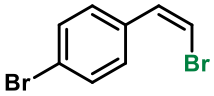


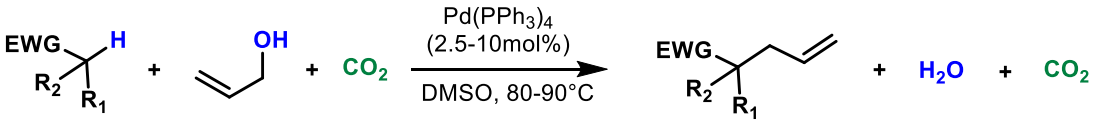
128 68%



129 67%

Powell, K. J.; Han, L.-C.; Sharma, P.; Moses, J. E., *Org. Lett.* **2014,** *16* (8), 2158-2161.

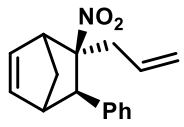
Activation of Allylic Alcohols with Carbon Dioxide



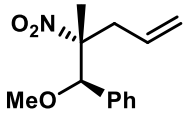
EWG = NO_2, CN, CHO

48 examples

selected examples



130 94%
95:5 dr



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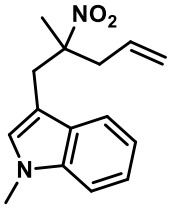
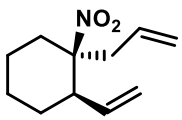
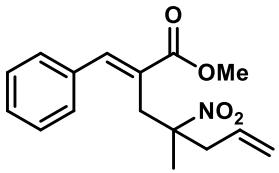
88:12 dr



132 86%



133 90%
85:15 dr



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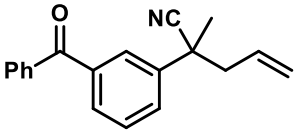

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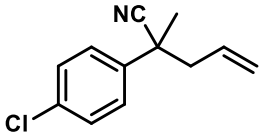

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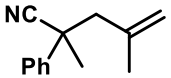

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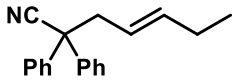

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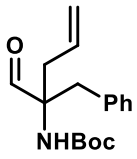

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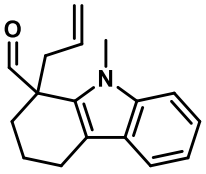

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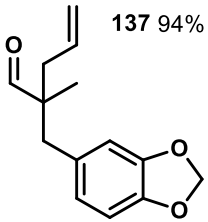

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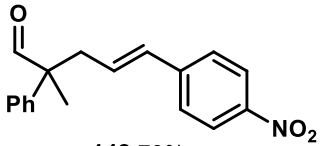
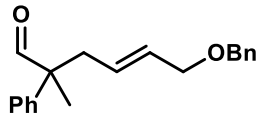


142 79%



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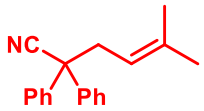
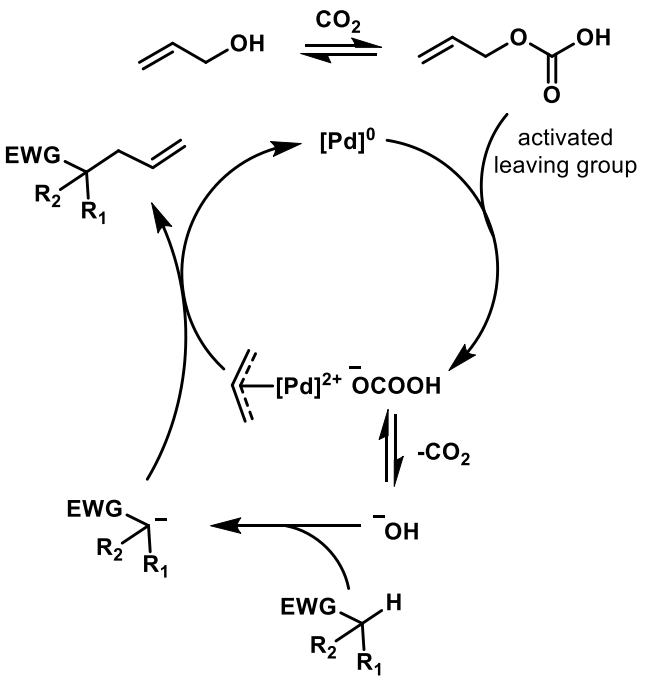


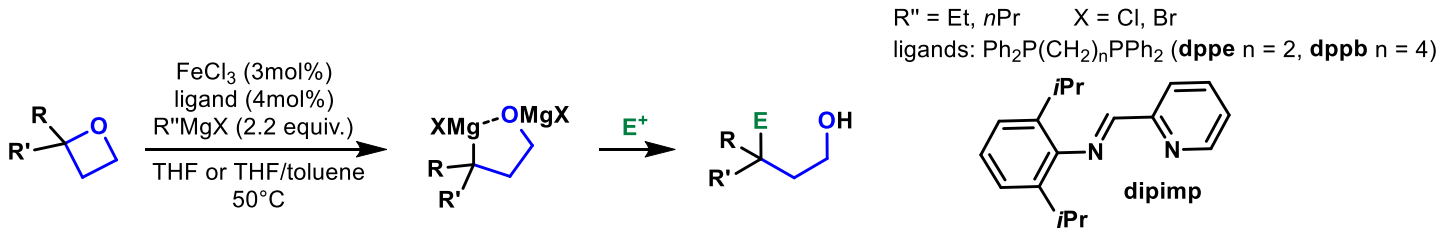
144 0%

mechanism



Lang, S. B.; Locascio, T. M.; Tunge, J. A., *Org. Lett.* **2014,** *16* (16), 4308-4311.

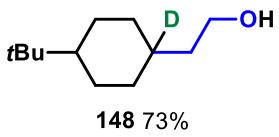
Iron-Catalyzed Reductive Magnesiation of Oxetanes



examples:

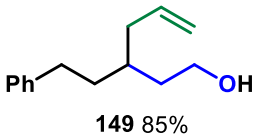
E = H_2O/D_2O

143 R = Ph, R' = H	>99%
144 R = CH_2CH_2Ph, R' = H	>99%
145 R = Me, R' = Ph	>99%
146 R = R' = Ph	91%
147 R = Et, R' = CH_2OPh	7%

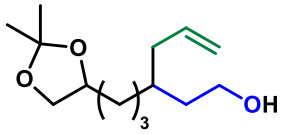


148 73%

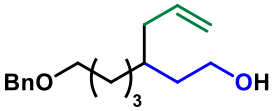
E = allyl bromide



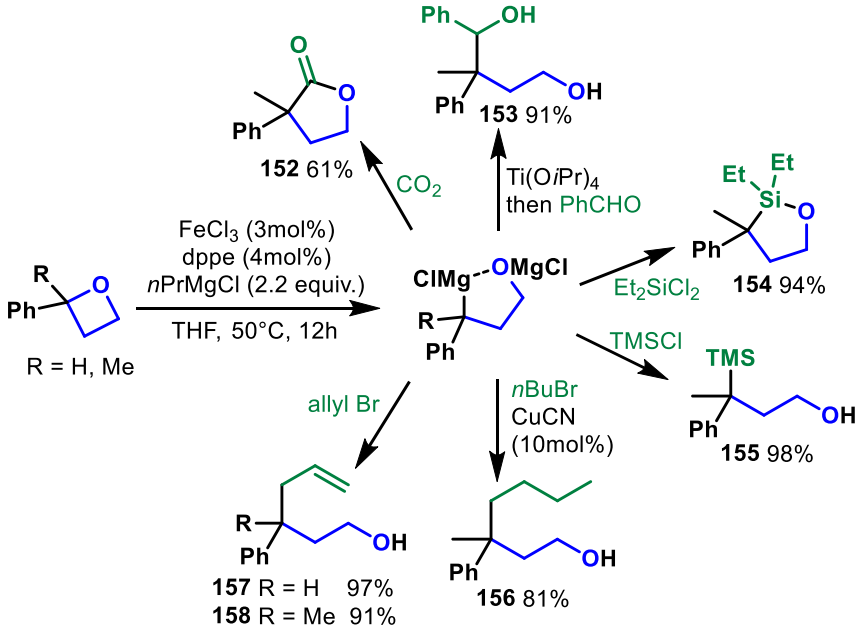
149 85%



150 87%



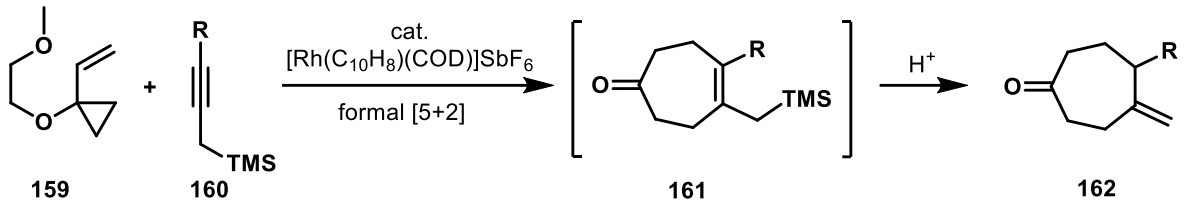
151 84%

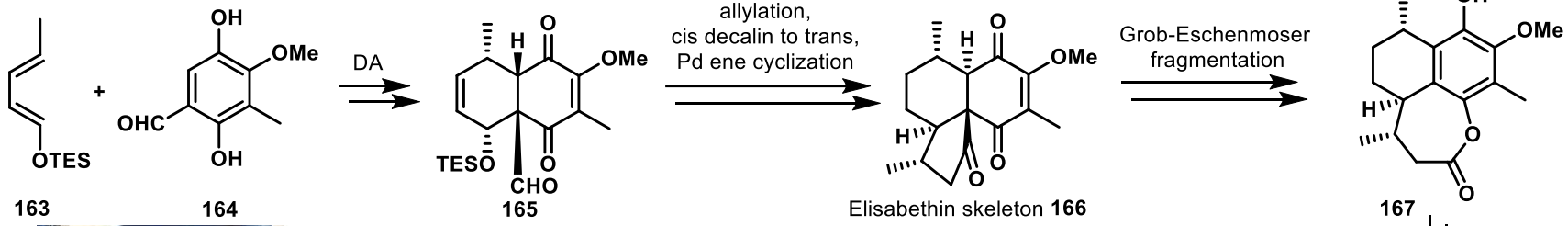


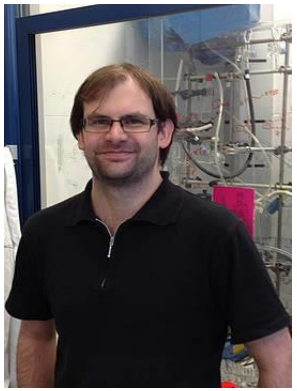
Sugiyama, Y.-k.; Heigozono, S.; Okamoto, S., *Org. Lett.* **2014,** *16* (24), 6278-6281.

Some familiar faces…

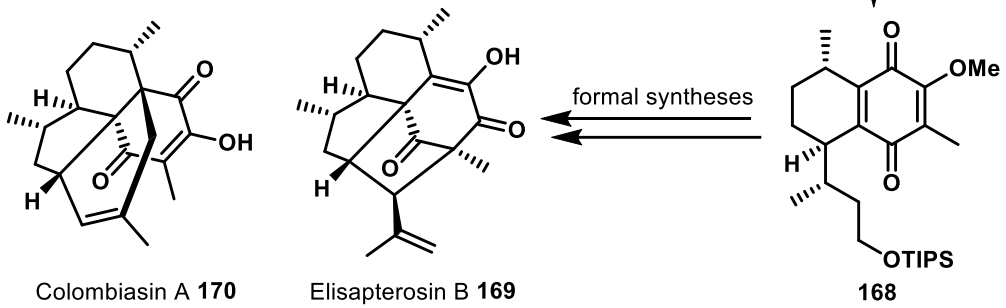
Propargyltrimethylsilanes as Allene Equivalents in Transition Metal-Catalyzed [5 + 2] Cycloadditions







A Short Access to the Skeleton of Elisabethin A and Formal Syntheses of Elisapterosin B and Colombiasin A



Wender, P. A.; Inagaki, F.; Pfaffenbach, M.; Stevens, M. C., *Org. Lett.* **2014,** *16* (11), 2923-2925.
Preindl, J.; Leitner, C.; Baldauf, S.; Mulzer, J., *Org. Lett.* **2014,** *16* (16), 4276-4279.

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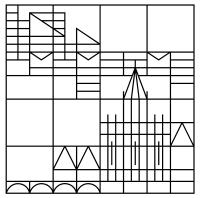

Thank you
for your attention!