Synthesis of Spiro[benzazepine-2,4′-piperidine]

J. Cossy,* C. Poitevin† and D. Gomez Pardo

Laboratoire de Chimie Organique, Associé au CNRS, ESPCI, 10 rue Vauquelin, 75231, Paris Cedex 05, France

J.-L. Peglion* and A. Dessinges

Institut de Recherches Servier, 11 rue des Moulineaux, 92150 Suresnes Cedex, France

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Interest in spiroheterocycles derives from the varied biological activity of this class of compounds.[1] As part of our program in heterocyclic chemistry, we needed to synthesize spiro[benzazepine-2,4′-piperidines] of type **A**. Two strategies were conceived to generate this target. The first would involve a Heck reaction as the key step from aryl bromide **B**, while the second would employ an aldol condensation on compound **C**. (Scheme 1).

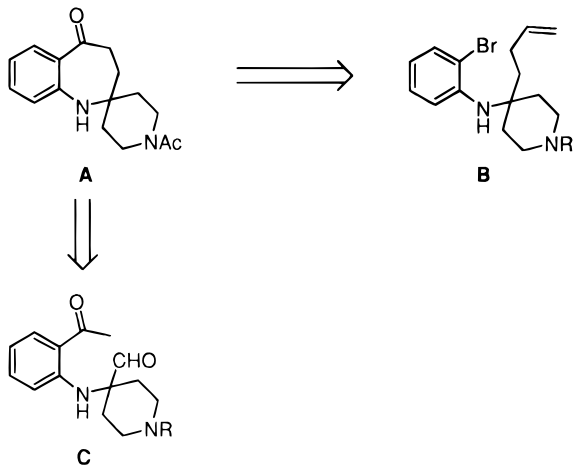
Recently, great attention has been focused on the Heck[2] reaction because of its technical simplicity and tolerance to a variety of functional groups. The intramolecular Heck[3] reaction has been used to build spiroheterocycles and tetrasubstituted carbon centers.[4]

The synthesis of **B** (R = Boc) was planned from the *N*-Boc-4-piperidone **1** in two steps (Scheme 2). After condensation of the *N*-Boc-4-piperidone **1** with *o*-bromoaniline **2**, a solution of the imine **3** in toluene was treated with homoallylmagnesium bromide. Unfortunately the corresponding amine **4** could not be isolated.

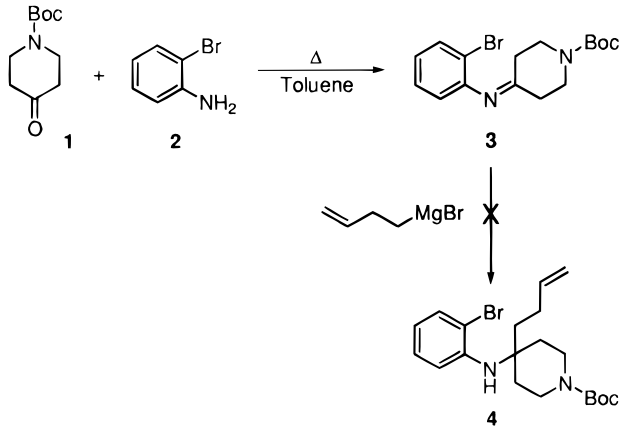
A second approach based on a Heck reaction and an intramolecular aldol condensation is outlined in a retrosynthetic scheme (Scheme 3).

In contrast to homoallylmagnesium bromide, allylmagnesium bromide reacted readily with imine **3** in toluene (Scheme 4) and led to the amine **5** [allylmagnesium bromide (1 equiv), rt, 72% yield]. After deprotection of **5** (HCl/EtOH, reflux, 2 h) and treatment of the resulting diamine **6** with acetic anhydride (1.1 equiv) in the presence of DMAP (1 equiv) and Et_3N (1.1 equiv), the monoprotected amine **7** was obtained from **2**, with an overall yield of 47%. When the Heck reaction was applied to **7** [$Pd(OAc)_2$ (0.11 equiv), PPh_3 (0.22 equiv), Et_3N (2.2 equiv)], **8′** was the only observed product when the crude reaction mixture was analyzed by ^1H NMR. After purification by flash chromatography on silica gel, an isomerization occurred and two regioisomers **8** and **8′** were obtained in a ratio of 10/90. However, when the mixture **8**/**8′** was treated with an excess of acetyl chloride (16 equiv) in refluxing chloroform for 20 h, the more thermodynamically stable *N*,*N*-diacetylated spiro[benz-

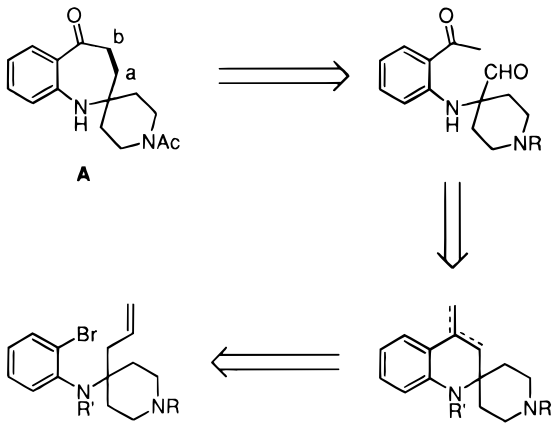
Scheme 1



Scheme 2



Scheme 3



* To whom the correspondence should be addressed. Phone: (+33) 1 40 79 44 29. Fax: (+33) 1 40 79 44 25. E-mail: janine.cossy@espci.fr.

† Present address: Institut de Recherches Servier, 11 rue des Moulineaux − 92150 Suresnes Cedex − France.

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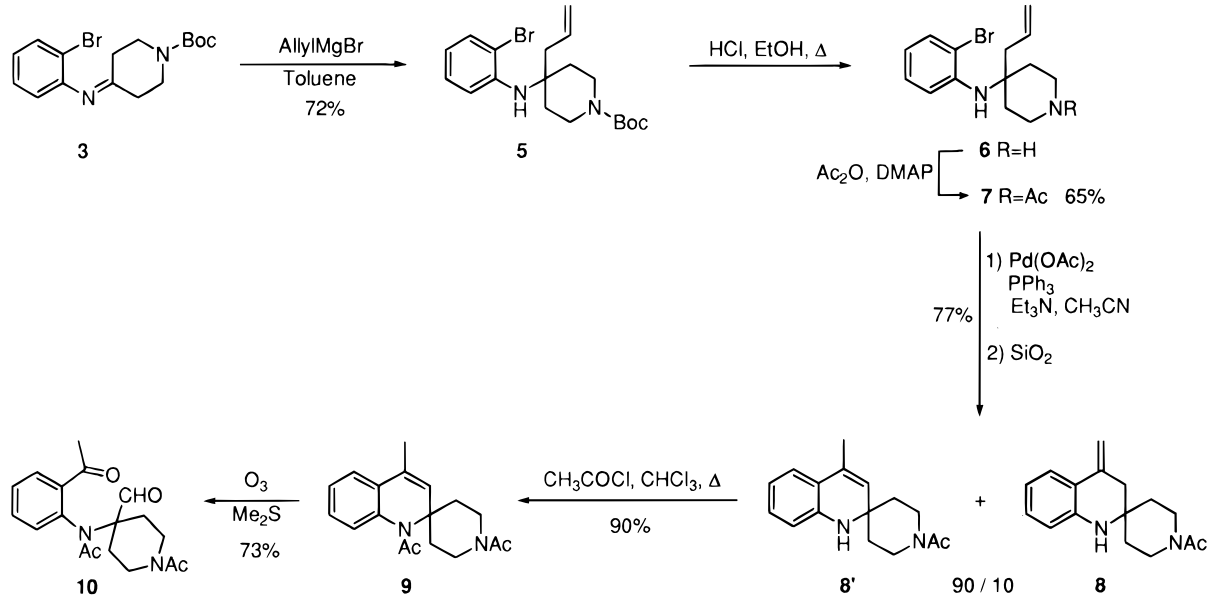
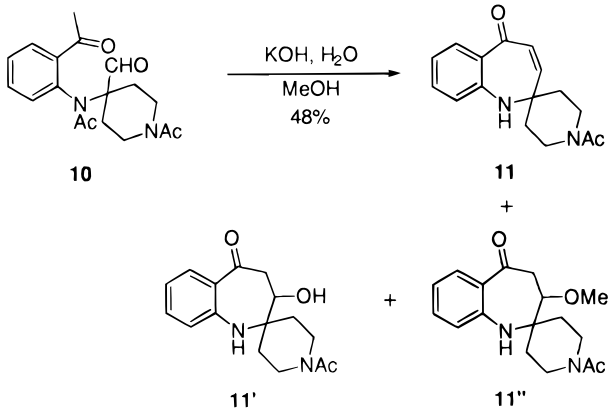
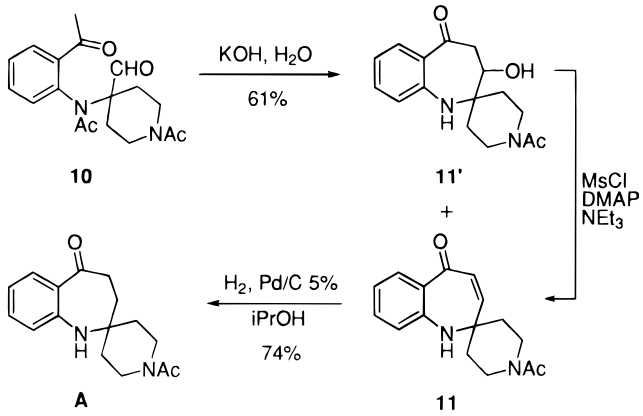
Scheme 4



Scheme 5



Scheme 6



azepine-2,4′-piperidine] **9**, was the only isolated product (90%). The unsaturated protected compound **9** was ozonolyzed and the keto aldehyde **10** was isolated after Me_2S reduction of the ozonide (73% yield).

The construction of the seven-membered ring of **A** was achieved from **10** by using an intramolecular aldol condensation (Scheme 5). When the keto aldehyde **10** was treated with an aqueous methanolic solution of KOH, three products, **11** (aldolisation-crotonisation product), **11′** (aldolisation product),[5] and **11″** (addition of methylate on **11**), were isolated in a 8/1/1 ratio (48% yield). Furthermore, concommitently to the aldolisation a desacetylation of the *N*-arylamine occurred during the process.

When the reaction was performed in THF in the presence of an aqueous solution of KOH, only the two products **11** and **11′** were isolated in a ratio of 8/2 (61% yield). We point out that **11′** could be transformed to **11** when compound **11′** was treated with mesyl chloride (2.9 equiv) in the presence of DMAP (0.09 equiv) and an excess of Et_3N (4.65 equiv) in CH_2Cl_2 at room temperature (Scheme 6). Compound **11** was obtained from **10** with an overall yield of 61%. After hydrogenation of **11** (Pd/C, 5%; H_2, 1 atm; iPrOH), the desired spiro[benzazepine-2,4′-piperidine] **A** was isolated (74% yield).

This synthesis, employing an aldol condensation, is an efficient entry to spiro[benzazepine-2,4′-piperidine] **A**. The target compound was obtained in nine steps from the *N*-Boc-piperidinone **1** with an overall yield of 11%.

Experimental Section

General. ^1H NMR and ^{13}C NMR spectra were recorded at 300 and 75 MHz, respectively, using $CDCl_3$ or CD_3OD as solvents. Chemical shifts are reported as δ values in ppm relative to internal standard tetramethylsilane. Only major diagnostic IR spectra absorption bands are reported. Mass spectra were obtained by GC/MS with electron impact ionization on a 5971 Hewelt Packard instrument at 70 eV; only selected ions are reported. Uncorrected melting points were taken using a Kofler bank. Moisture sensitive reactions were conducted in oven-dried glassware under a nitrogen atmosphere. Analytical TLC was performed on Merck silica gel 60F-254 plates. Flash column chromatography was accomplished on Merck Kieselgel 60 (230−400 mesh). Solvents such as chloroform, dichloroethane, toluene, or acetonitrile were purified by distillation under N_2 from CaH_2, and ethanol was distilled from Mg.

1-*tert*-Butoxycarbonyl-4-allyl-4-(2-bromophenylamino)piperidine (5). A solution of 2-bromoaniline **2** (8.2 g, 47.7 mmol, 1.0 equiv) and *N*-*tert*-butoxycarbonyl-4-piperidone **1** (9.1 g, 45.7 mmol, 1.0 equiv) in benzene (12 mL) was heated at reflux, with azeotropical removal of water, during 72 h. After cooling and concentration, the crude ketimine **3** was obtained as an oil. To

(5) **11′** was converted into **11** under basic conditions (KOH, H_2O, MeOH).

a solution of **3** in anhydrous toluene (100 mL) was added, during 90 min, a solution of allylmagnesium bromide (45.3 mL, 45.3 mmol, 1.0 equiv, 1 M in diethyl ether). The mixture was stirred during 24 h. The reaction mixture was poured into a saturated aqueous NH_4Cl solution (50 mL). After extraction with diethyl ether (3 × 500 mL), the organic phase was dried over $MgSO_4$ and filtered. The solvent was removed in vacuo to afford an oil which was purified by flash columm chromatography on silica gel (EtOAc/cyclohexane 15/85) to give **5** as a yellow oil (13.0 g; 32.8 mmol, 72% yield). R_f 0.68 (EtOAc/cyclohexane 15/85); IR (neat) 3400, 1600 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.42 (dd, J = 7.9 and 1.5 Hz, 1H), 7.11 (ddd, J = 8.0, 7.9 and 1.5 Hz, 1H), 6.87 (dd, J = 8.2 and 1.4 Hz, 1H), 6.55 (ddd, J = 8.0, 7.9 and 1.4 Hz, 1H), 5.81−5.63 (m, 1H), 5.10−4.95 (m, 2H), 4.21 (s, 1H), 3.95−3.65 (m, 2H), 3.22−3.08 (m, 2H), 2.55−2.48 (m, 2H), 2.09−1.96 (m, 2H), 1.70−1.44 (m, 11H); ^{13}C NMR ($CDCl_3$) δ 154.6, 142.6, 132.7, 132.6, 127.9, 118.4, 118.0, 114.3, 111.9, 79.3, 54.2, 42.6, 39.2, 34.9, 28.3; EI MS *m*/*z* (relative intensity) 396 ($M^{+\bullet}$, 2), 394 ($M^{+\bullet}$, 2), 355 (44), 353 (44), 299 (99), 297 (100), 238 (34), 236 (33), 226 (64), 224 (65), 82 (33), 57 (72).

1-Acetyl-4-allyl-4-(2-bromophenylamino)piperidine (7). A solution of **5** (31.5 g, 79.5 mmol, 1.0 equiv) in absolute ethanol (60 mL) was treated with a 2.5 N ethanolic hydrochloric acid solution (40 mL). The solution was heated at reflux for 2 h. The solvent was removed in vacuo. The remaining residue was treated with an aqueous solution of sodium hydroxide (1 N) to pH 10, extracted with CH_2Cl_2 (2 × 75 mL). The organic phase was dried over $MgSO_4$ and removal of the solvent in vacuo afforded the crude diamine **6** (20.0 g, 67.9 mmol) as a brown oil. A solution of **6** (20.0 g, 67.8 mmol, 1.0 equiv) in CH_2Cl_2 (100 mL) containing DMAP (6.2 g, 69.0 mmol, 1.0 equiv), Ac_2O (7.0 mL, 74.2 mmol, 1.1 equiv), and Et_3N (10.4 mL, 74.6 mmol, 1.1 equiv) was stirred for 16 h at room temperature. After removal of the solvent in vacuo, the remaining solid was purified by flash column chromatography on silica gel (EtOAc/cyclohexane/MeOH 75/15/10) to give **7** as a yellow oil (17.6 g, 52.2 mmol, 65% yield). R_f 0.75 (EtOAc/cyclohexane/MeOH 75/15/10); IR (neat) 1680 (broad) cm^{-1}; ^1H NMR (CD_3OD) δ 7.42 (dd, J = 8.1 and 1.5 Hz, 1H), 7.19−7.01 (m, 1H), 6.96 (dd, J = 8.1 and 1.5 Hz, 1H), 6.57 (ddd, J = 8.1, 7.7 and 1.5 Hz, 1H), 5.75−5.59 (m, 1H), 5.05−4.86 (m, 2H), 4.17−4.05 (m, 1H), 3.68−3.54 (m, 1H), 3.40−3.22 (m, 1H), 3.08−2.94 (m, 1H), 2.55−2.41 (m, 2H), 2.15−1.92 (m, 5H), 1.70−1.42 (m, 2H); ^{13}C NMR (CD_3OD) δ 171.7, 144.4, 134.3, 134.2, 129.6, 119.9, 119.4, 116.4, 113.3, 55.8, 43.6, 43.5, 38.7, 36.7, 36.3, 21.6; EI MS *m*/*z* (relative intensity) 338 ($M^{+\bullet}$, 4), 336 ($M^{+\bullet}$, 4), 297 (50), 295 (52), 238 (96), 236 (100), 124 (17), 82 (18); HRMS calcd for $C_{16}H_{21}BrN_2O$ 336.0837, found 336.0831.

1-Acetyl-4′-methylidene-3′,4′-dihydrospiro[piperidine-4,2′(1′*H*)-quinoline] (8) and 1-acetyl-4′-methylspiro[piperidine-4,2′(1′*H*)-quinoline] (8′). To a solution of **7** (8.8 g, 26.0 mmol, 1.0 equiv) in anhydrous acetonitrile (200 mL) was added triethylamine (8.0 mL, 57.4 mmol, 2.2 equiv), triphenylphosphine (1.50 g, 5.72 mmol, 0.22 equiv), and palladium acetate (0.64 g, 2.85 mmol, 0.11 equiv). The reaction mixture was heated at 80 °C for 72 h. After cooling, palladium was filtered off and removal of acetonitrile in vacuo afforded a brown residue which was purified by flash column chromatography on silica gel (EtOAc/cyclohexane/MeOH 75/15/10). A mixture of **8** and **8′** (5.15 g, 20.11 mmol, 77% yield) in 10/90 ratio was obtained. R_f 0.51 (EtOAc/cyclohexane/MeOH 75/15/10). **8′** "*endo*": IR (neat) 3350, 1680 cm^{-1}; ^1H NMR ($CDCl_3$) δ 6.93 (dd, J = 7.7 and 1.3 Hz, 1H), 6.85 (ddd, J = 7.3, 7.3 and 1.5 Hz, 1H), 6.51 (ddd, J = 7.3, 7.7 and 1.1 Hz, 1H), 6.39 (dd, J = 7.7 and 1.5 Hz, 1H), 5.29 (s, 1H), 4.33 (s broad, 1H), 3.65−3.15 (m, 4H), 1.94 (s, 3H), 1.89 (s, 3H), 1.72−1.40 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 168.8, 142.7, 130.5, 128.4, 124.1, 123.7, 121.6, 117.3, 113.2, 51.4, 41.7, 38.7, 37.9, 36.9, 21.3, 18.6; EI MS *m*/*z* (relative intensity) 256 ($M^{+\bullet}$, 33), 213 (40), 185 (13), 171 (24), 170 (100), 169 (32), 158 (22), 157 (59), 143 (11), 115 (11); HRMS calcd for $C_{16}H_{20}N_2O$ 256.1576, found 256.1575. **8** "*exo*" (always mixed with **8′**): ^1H NMR ($CDCl_3$) δ 7.67−7.30 (m, 4H), 5.39 (s, 1H), 4.75 (s, 1H), 4.33 (s broad, 1H), 3.65−3.15 (m, 4H), 2.41 (s, 2H), 1.94 (s, 3H), 1.72−1.40 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 168.8, 142.1, 131.9, 131.1, 124.5, 117.7, 115.1, 108.0, 51.4, 42.3, 41.2, 37.4, 35.8, 35.6, 21.3; EI MS *m*/*z* (relative intensity) 256 ($M^{+\bullet}$, 46), 213 (24), 170 (90), 169 (100), 157 (52), 143 (11), 115 (11).

1,1′-Diacetyl-4′-methylspiro[piperidine-4,2′(1′*H*)-quinoline] (9). To a solution of **8** and **8′** (1.0 g, 4.0 mmol, 1.00 equiv) in anhydrous $CHCl_3$ (10 mL) was added dropwise acetyl chloride (4.5 mL, 63.3 mmol, 16.0 equiv), and the reaction mixture was stirred and heated at 70 °C during 20 h. After cooling to 0 °C, an aqueous sodium hydroxide solution (5 N) was added dropwise to the reaction mixture with vigorous stirring. The resulting mixture was stirred at room temperature for 20 min and then extracted with diethyl ether (2 × 150 mL). The etheral phases were dried over $MgSO_4$ and concentrated in vacuo to afford an oil which was purified by flash column chromatography on silica gel (EtOAc/cyclohexane/MeOH 75/15/10) to give **9** (1.1 g, 3.6 mmol, 90% yield). R_f 0.35 (EtOAc/cyclohexane/MeOH 75/15/10); IR ($CHCl_3$) 1680 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.28−7.13 (m, 3H), 7.10−7.03 (m, 1H), 5.77 (s, 1H), 3.90−3.79 (m, 1H), 3.65−3.20 (m, 3H), 2.80−2.61 (m, 1H), 2.25−2.08 (m, 1H), 2.02 (s, 6H), 1.88 (s, 3H), 1.65−1.48 (m, 1H), 1.45−1.29 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 173.0, 168.8, 138.0, 133.6, 131.3, 130.6, 127.2, 126.6, 126.3, 123.3, 59.0, 43.5, 38.1, 34.8, 33.6, 26.4, 21.2, 17.7; EI MS *m*/*z* (relative intensity) 298 ($M^{+\bullet}$, 100), 255 (47), 196 (48), 170 (67), 169 (44), 150 (61), 130 (48), 124 (46), 115 (37).

***N*-(1-Acetyl-4-formylpiperidin-4-yl)-*N*-(2-acetylphenyl)-acetamide (10).** A solution of **9** (6.2 g, 20.8 mmol, 1.0 equiv) in absolute ethanol (150 mL) was treated with ozone at −78 °C. After 1 h, dimethyl sulfide (50 mL, 680 mmol, 32.7 equiv) was added. The reaction mixture was warmed to room temperature and concentrated in vacuo. The resulting oil was purified by flash column chromatography on silica gel (EtOAc/cyclohexane/MeOH 75/15/10) to give **10** as an oil (5.0 g, 15.2 mmol, 73% yield). R_f 0.33 (EtOAc/cyclohexane/MeOH 75/15/10); IR (neat) 1725, 1685, 1635 cm^{-1}; ^1H NMR ($CDCl_3$) (mixture of rotamers) δ 9.59−9.48 (m, 1H), 7.90−7.82 (m, 1H), 7.65−7.50 (m, 2H), 7.29−7.11 (m, 1H), 4.36−4.17 (m, 1H), 3.85−3.42 (m, 2H), 3.28−3.02 (m, 1H), 2.77−2.31(m, 4H), 2.09−1.78 (m, 4H), 1.69 (s, 3H), 1.62−1.35 (m, 1H), 1.20−1.00 (m, 1H); ^{13}C NMR ($CDCl_3$) (mixture of rotamers) δ 199.0, 198.8, 197.6, 171.0, 170.9, 168.7, 168.6, 137.3, 137.1, 136.0, 133.2, 133.1, 131.9, 131.6, 130.6, 130.4, 129.6, 65.1, 65.0, 43.2, 41.7, 38.0, 36.4, 30.9, 30.1, 29.4, 29.1, 28.1, 23.0, 21.0, 20.9; EI MS *m*/*z* (relative intensity) 312 ($M^{+\bullet} - H_2O$, 3), 269 (12), 184 (30), 162 (100), 120 (51), 108 (54); HRMS calcd for $C_{18}H_{22}N_2O_4$ 330.1586, found 330.1584.

1′-Acetyl-3-hydroxy-3,4-dihydrospiro[5*H*-1-benzazepine-2(1*H*),4′-piperidin]-5-one (11′) and 1′-Acetyl-3-methoxy-3,4-dihydrospiro[5*H*-1-benzazepine-2(1*H*),4′-piperidin]-5-one (11″). A solution of **10** (0.27 g, 0.81 mmol, 1.0 equiv) in methanol (2 mL) was added to potassium hydroxide (0.09 g, 1.66 mmol, 2.05 equiv) in water (3 mL) at 0 °C. After 4 h, The reaction mixture was concentrated in vacuo. The remaining residue was extracted with CH_2Cl_2 (2 × 30 mL), the organic phase was dried over $MgSO_4$, and the solvent was removed in vacuo. The resulting yellow solid was purified by flash column chromatography on silica gel (EtOAc/cyclohexane/MeOH 75/15/10) to give **11** as a yellow solid (0.104 g, 0.38 mmol, 48% yield), **11′** as an amorphous solid (0.02 g, 0.07 mmol, 8% yield), and **11″** as an oil (0.02 g, 0.06 mmol, 7% yield).

Analytical data for 11″: R_f 0.45 (EtOAc/cyclohexane/MeOH 75/15/10); IR (neat) 3400, 1640−1620, 1580 cm^{-1}; ^1H NMR ($CDCl_3$) (mixture of rotamers) δ 7.75−7.60 (m, 1H), 7.32−7.28 (m, 1H), 6.89−6.78 (m, 1H), 6.77−6.69 (m, 1H), 4.24−3.84 (m, 2H), 3.25−3.35 (m, 3H), 3.12−2.90 (m, 4H), 2.00−1.50 (m, 9H); ^{13}C NMR ($CDCl_3$) (mixture of rotamers) δ 198.7, 198.4, 168.9, 146.7, 133.4, 133.3, 131.1, 128.5, 120.1, 119.9, 87.8, 87.0, 59.0, 58.9, 57.5, 57.4, 45.4, 44.7, 42.0, 37.1, 37.0, 34.5, 33.7, 30.7, 29.0, 21.2.

Analytical data for 11′: R_f 0.35 (EtOAc/cyclohexane/MeOH 75/15/10); IR ($CHCl_3$) 3320, 1705, 1620 (broad) cm^{-1}; ^1H NMR ($CDCl_3$) (mixture of rotamers) δ 7.75 (d, J = 9.0 Hz, 1H), 7.37 (ddd, J = 9.0, 8.8 and 1.8 Hz, 1H), 6.95 (ddd, J = 9.0, 8.8 and 1.8 Hz, 1H), 6.83 (d, J = 8.8 Hz, 1H), 4.30−3.93 (m, 3H), 3.63−3.40 (m, 1H), 3.29−2.89 (m, 4H), 2.50 (s, 1H); 2.05−1.97 (m, 3H), 1.95−1.55 (m, 4H); ^{13}C NMR ($CDCl_3$) (mixture of rotamers) δ 199.1, 198.6, 169.0, 146.1, 146.0, 133.4, 129.1, 128.9, 127.4, 120.2, 120.1, 119.9, 119.8, 77.2, 76.7, 58.7, 49.9, 41.9, 41.8, 37.0, 36.9, 33.1, 32.3, 31.1, 30.4, 21.2; EI MS *m*/*z* (relative intensity) 288 ($M^{+\bullet}$, 7), 259 (65), 200 (100), 188 (12), 182 (15), 170 (9), 158 (9), 91 (9).

1′-Acetylspiro[5*H*-1-benzazepine-2(1*H*),4′-piperidin]-5-one (11). A solution of **10** (5.0 g, 15.2 mmol, 1.0 equiv) in THF (5 mL) was added to potassium hydroxide (1.7 g, 30.4 mmol, 2.0 equiv) in water (5 mL) at 0 °C. After 4 h, the resulting precipitate was filtered, washed with cold water, and then dried over P_2O_5 to afford **11** (2.0 g, 7.4 mmol) as a yellow solid. The filtrate was extracted with methylene chloride (2 × 50 mL), the organic layer was dried over $MgSO_4$, and the solvent removed in vacuo to afford a solid (1.0 g, 3.5 mmol) which was dissolved in methylene chloride (25 mL). To this solution were added anhydrous triethylamine (2.3 mL, 16.1 mmol, 4.6 equiv) and DMAP (0.04 g, 0.32 mmol, 0.09 equiv) at 0 °C, and then mesyl chloride (0.74 mL, 9.56 mmol, 2.90 equiv) was added under vigorous stirring. After 1 h, the solvent was removed in vacuo, and the remaining solid was purified to afford **11** as a yellow solid (0.50 g, 1.85 mmol, 61% yield). R_f 0.40 (EtOAc/cyclohexane/MeOH 75/15/10). IR (CCl_4) 3300, 1710, 1680−1530 (broad) cm^{-1}; ^1H NMR ($CDCl_3$) δ 8.05 (dd, J = 8.1 and 1.8 Hz, 1H), 7.34 (ddd, J = 8.1, 7.0 and 1.8 Hz, 1H), 6.92 (ddd, J = 8.1, 7.0 and 1.1 Hz, 1H), 6.77 (dd, J = 8.1 and 1.1 Hz, 1H), 6.39 (d, J = 12.1 Hz, 1H), 6.33 (d, J = 12.1 Hz, 1H), 4.30 (s, 1H), 3.82−3.57 (m, 2H), 3.47−3.36 (m, 2H), 2.08 (s, 3H), 2.02−1.77 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 189.9, 169.0, 145.4, 145.2, 134.6, 133.0, 131.2, 123.6, 120.2, 119.9, 53.8, 42.3, 37.2, 35.7, 35.2, 21.3; EI MS m/z (relative intensity) 270 ($M^{+\bullet}$, 100), 227 (38), 198 (25), 184 (16), 172 (19), 156(18), 143(18); HRMS calcd for $C_{16}H_{18}N_2O_2$ 270.1368, found 270.1368.

1′-Acetyl-3,4-dihydrospiro[5*H*-1-benzazepine-2(1*H*),4′-piperidin]-5-one (A). A suspension of enone **11** (2.0 g, 7.4 mmol, 1.0 equiv) in isopropyl alcohol (180 mL) was hydrogenated over palladium on charcoal 5% (0.2 g) under 1 atm. After completion of the reaction, the palladium was filtered off and the crude product was purified by flash column chromatography on silica gel (EtOAc/cyclohexane/MeOH 75/15/10), to furnish **A** as a white solid (1.5 g, 5.5 mmol, 74% yield): mp 153−154 °C (ethanol); R_f 0.40 (EtOAc/cyclohexane/MeOH 75/15/10); IR (Nujol) 3326, 1668, 1626 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.72 (d, J = 8.1 Hz, 1H), 7.21 (dd, J = 7.7 and 7.7 Hz, 1H), 6.84 (dd, J = 7.7 and 7.7 Hz, 1H), 6.78 (d, J = 7.7 Hz, 1H), 4.28 (s, 1H), 3.62−3.21 (m, 4H), 2.82−2.60 (m, 2H), 1.98 (s, 3H), 1.95−1.80 (m, 2H), 1.70−1.50 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 201.8, 168.7, 147.3, 133.5, 129.3, 127.4, 121.8, 120.7, 55.2, 42.4, 39.0, 37.5, 37.2, 37.0, 35.8, 21.2; EI MS m/z (relative intensity) 272 ($M^{+\bullet}$, 100), 229 (26), 186 (49), 185 (46), 120 (26), 95 (23). Anal. Calcd for $C_{16}H_{20}N_2O_2$: C, 70.56; H, 7.40; N, 10.29. Found C, 70.33; H, 7.13; N, 10.11.

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Supporting Information Available: Copies of ^1H NMR for the compounds **5**, **7**, **8′**, **9**, **10**, **11**, **11′**, **11″** (8 pages). This material is contained in libraries on microfiche, immediately follows this article in the microfilm version of the journal, and can be ordered from the ACS; see any current masthead page for ordering information.

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