Ruthenium-Catalyzed Oppenauer-Type Oxidation of 3β-Hydroxy Steroids. A Highly Efficient Entry into the Steroidal Hormones with 4-En-3-one Functionality

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Oxidation of 5-unsaturated 3β-hydroxy steroids **1** to the corresponding 4-en-3-one derivatives **2** can be performed efficiently by acetone at reflux in the presence of a catalytic system consisting of either $(PPh_3)_3RuCl_2$ (**3**) and K_2CO_3 or $[(C_4Ph_4COHOCC_4Ph_4)(\mu\text{-}H)][(CO)_4Ru_2]$ (**4**). The reaction proceeds via a ruthenium-catalyzed dehydrogenation of **1** and subsequent hydrogen transfer to acetone with concomitant double bond migration.

Introduction

Steroid hormones are vital regulators of the lives of mammals. These molecules control a variety of body functions, such as reproduction (male and female sexual hormones), carbohydrate metabolism (glucocorticoids), ion transport (mineralocorticoids), etc.[1] Since their applications in human and veterinary medicine are numerous and since a substantial proportion of modern birth-control methods relies on "The Pill", there is a continuing need for efficient, large-scale industrial production of both natural hormones and their analogues.

In spite of enormous progress in synthetic methodology[2,3] and total synthesis of steroids[3,4] in the last two decades, most industrially viable procedures are still using semisynthetic routes[1,5,6] starting from naturally occurring steroids—diosgenin, stigmasterol, sitosterol, cholesterol, etc.[1,5] Here, microbiological processes[1,5] often play a crucial role, bringing about substantial shortcuts that cannot be attained by classical organic chemistry methods. On the other hand, the advent of powerful transition metal catalysts, which has had a dramatic impact on organic chemistry in general, has not yet been fully appreciated in steroid chemistry.[7]

A typical feature of major steroidal hormones, such as testosterone, progesterone, cortisol, and aldosterone, is the enone moiety in the A-ring (cf. **2** in Scheme 1 and Figure 1).[1] The latter functionality can be constructed by oxidation of a suitable industrial precursor,[1,8] such as 3β-hydroxypregn-5-en-20-one. This is a standard step in the bulk production of steroidal hormones.[5] Simple though it may seem, this step is, however, beset by serious problems. Thus, for instance, standard procedures based on Cr(VI) reagents prove to be capricious in this particular case and hardly applicable by industry on a large scale.[1,8,9] The classical Oppenauer oxidation[10] using $(i\text{-}PrO)_3Al$ and a large excess of cyclohexanone, which had been introduced into steroid chemistry in the early days,[1,11] still appears to be the best choice, in spite of considerable efforts[8] aimed at developing a more efficient alternative. With this method, however, the resulting enone is always contaminated with the products of aldol condensation of cyclohexanone, for whose separation steam distillation has to be used.[8,11] Furthermore, the need for a high-boiling solvent (toluene or xylene) further increases the energy cost.[12−14] The former problem has been partly solved by replacing cyclohexanone with 1-methyl-4-piperidone;[15] being an amino ketone, the latter reagent gives rise to amine-containing aldol byproducts, which can be separated from the steroid by acid extraction. However, this still represents considerable inconvenience on the bulk scale, in addition to the higher cost of 1-methyl-4-piperidone (compared to cyclohexanone). Hence, there is clearly a need for a more efficient (preferably catalytic) method that would work at a lower temperature. Such a method would solve one of the long-standing problems in steroid chemistry and would be of considerable commercial interest.

In the last few years we have developed a mild, ruthenium-catalyzed procedure (with complexes **3** and **4**)

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(1) Fieser, L. F.; Fieser, M. *Steroids*; Reinhold: New York, 1959.
(2) (a) Kočovský, P.; Tureček, F.; Hájíček, J. *Synthesis of Natural Products: Problems of Stereoselectivity*; CRC Press: Boca Raton, FL, 1986; Vols. I and II. (b) Smith, M. B. *Organic Synthesis*; McGraw-Hill: New York, 1994.
(3) Corey, E. J.; Cheng, X.-M. *The Logic of Chemical Synthesis*; J. Wiley: New York, 1989; pp 23 and 385.
(4) Taub, D. In *The Total Synthesis of Natural Products*; ApSimon, J., Ed.; J. Wiley: New York, 1973; Vol. 2, p 641.
(5) (a) Lednicer, D.; Mitscher, L. A. *The Organic Chemistry of Drug Synthesis*; J. Wiley: New York, 1977; Vol. 1, p 155; (b) 1980; Vol. 2, p 135; (c) 1984; Vol. 3, p 81; (d) 1980; Vol. 4, p 65.
(6) Freid, J.; Edwards, J. A. *Organic Reactions in Steroid Chemistry*; Van Nostrand: New York, 1972.
(7) For recent examples of application of both transition and nontransition metals in steroid chemistry, see, e.g.: (a) Urbanos, F.; Fernandez-Baeza, J.; Chaudret, B. *J. Chem. Soc., Chem. Commun.* **1991**, 1739. (b) Halcrow, M. A.; Urbanos, F.; Chaudret, B. *Organometallics* **1993**, *12*, 955. (c) Urbanos, F.; Halcrow, M. A.; Fernandez-Baeza, J.; Dahan, F.; Labroue, D.; Chaudret, B. *J. Am. Chem. Soc.* **1993**, *115*, 3484. (d) Horiguchi, Y.; Nakamura, E.; Kuwajima, I. *J. Am. Chem. Soc.* **1989**, *111*, 6257. (e) KoËovsky, P.; Bains, R. S. *J. Org. Chem.* **1994**, *59*, 5439. (f) Kočovský, P.; Šrogl, J.; Pour, M.; Gogoll, A. *J. Am. Chem. Soc.* **1994**, *116*, 186.
(8) Rasmusson, G. H.; Arth, G. E. In *Organic Reactions in Steroid Chemistry*; Fried, J., Edwards, J. A., Eds.; Van Nostrand: New York, 1972; Vol. 1, p 222.
(9) The problem is that of the instability of the primarily generated 5-en-3-one toward further oxidation.[1,8]
(10) Oppenauer, R. V. *Rec. Trav. Chem.* **1937**, *56*, 137.
(11) Djerassi, C. *Org. React.* **1951**, *6*, 207.
(12) Another drawback is the need of stoichiometric quantities of $(i\text{-}PrO)_3Al$.
(13) 17α-Hydroxypregnan-20-one derivatives easily isomerize to D-homosteroids in the presence of Lewis acids or bases,[8,11,14] so that application of the Oppenauer oxidation in corticoid chemistry requires protection of the 17α-hydroxyl.
(14) For a mechanistic overview of the D-homo rearrangement, see: Kirk, D. N.; Hartshorn, M. P. *Steroid Reaction Mechanisms*; Elsevier: Amsterdam, 1968; p 294.
(15) (a) Keana, J. F. W.; Reich, R. *Synth. Commun.* **1972**, *2*, 323. (b) Kirk, D. N.; Rajagopalan, M. S.; Varley, M. J. *J. Chem. Soc., Perkin Trans. 1* **1983**, 2225.

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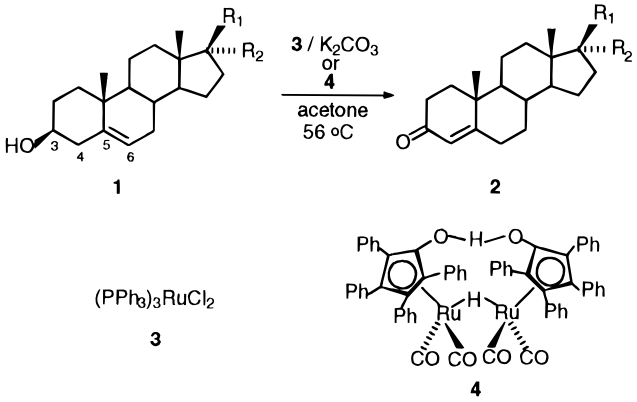
Scheme 1



for the dehydrogenation of primary and secondary alcohols.[16] The method has recently been significantly improved[17] and is now an efficient catalytic version of the Oppenauer oxidation. Herein we report the application of this new protocol to the oxidation of steroidal 5-en-3β-ols **1** to the corresponding 4-en-3-ones **2** (Scheme 1).

Results and Discussion

In order to develop milder and more efficient methods for the synthesis of biologically important substrates, such as steroidal hormones (Figure 1), we investigated the oxidation of a series of steroids (cholestanes, androstanes, and pregnanes) under our present catalytic conditions. The oxidations were carried out in acetone at reflux (56 °C) employing either ruthenium complex **3** and K_2CO_3 or complex **4** as depicted in Scheme 1.

Reaction of steroid alcohols **1** with acetone in the presence of 0.25−1 mol % of the catalyst afforded steroidal 4-en-3-ones **2** in good isolated yields (Table 1). When complex **3** was employed as catalyst, a catalytic amount of K_2CO_3 was added. We recently discovered that it is essential to add a small amount of water (~0.6%) to the acetone when using this catalytic system (K_2CO_3/**3**).[17] The mechanism of this reaction, which is described elsewhere,[16,17] involves a hydrogen transfer from the alcohol substrate to acetone via ruthenium alkoxide intermediates.

The procedure resulted in a highly selective reaction that tolerates the presence of double bonds, ketone and ester groups, tertiary hydroxyl groups, and protected corticoid side chains. Except for one case (**1g** → **2g**, Table 1, entry 11), the NMR spectrum of the crude product showed the presence of only one compound in essentially quantitative yield. The pure oxidized steroids **2** were easily isolated by simple evaporation of the solvent followed by flash chromatography.

In a previously described ruthenium-catalyzed oxidation of alcohols with MnO_2 as the oxidant, cholesterol (**1a**) gave a moderate yield of **2b**.[16c] Attempts to employ this method for the oxidation of the other steroids **1b**−**g** were unsuccessful and gave only low conversion. Also, the recently reported ruthenium−cobalt-catalyzed aerobic oxidation of alcohols[18] led to discouraging results. Thus, under these aerobic conditions stigmasterol **1b** was preferentially oxidized at the C-5 double bond. By contrast, with the catalytic Oppenauer-type oxidation in the present paper, 4-en-3-one was the sole product obtained in high yield from sigmasterol **1b** (Table 1, entries 2 and 3).

No problems were encountered in the oxidation of pregnenolone (**1c**). Again, clean reactions were observed, and progesterone **2c** was isolated in high yield (Table 1, entries 4 and 5).

In the androstanes series, compound **1d** showed a low reactivity with complex **3**/K_2CO_3 as the catalytic system. However, it was cleanly converted to the testosterone benzoate (**2d**) when complex **4** was employed as catalyst

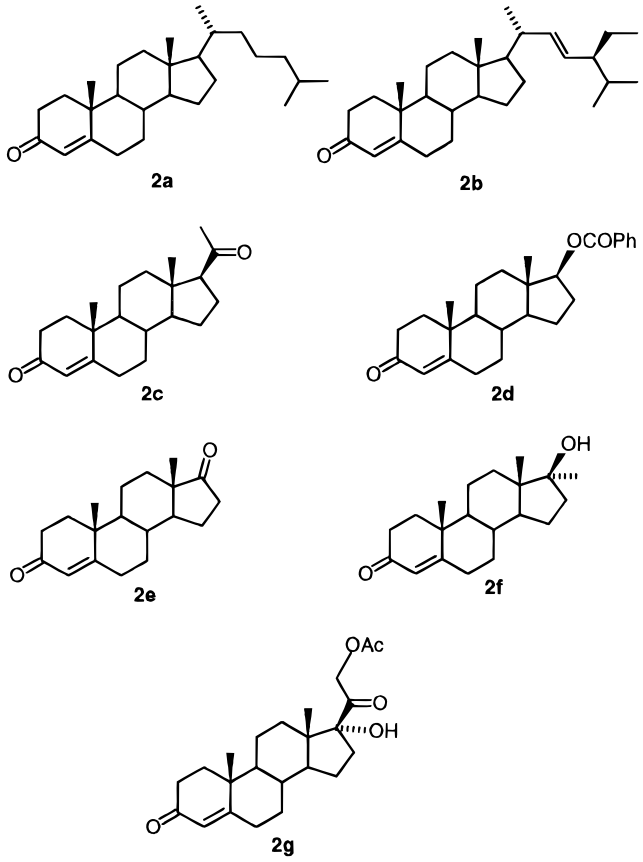


Figure 1.

Table 1. Ruthenium-Catalyzed Oxidation of Steroidal 5-En-3β-ols to 4-En-3-ones (1 → 2)[a]

entry	product	catalyst (mol%)	time (h)	yield (%)
1	**2a**	**4** (0.5)	22	74
2	**2b**	**3** (1)[b]	24	80
3	**2b**	**4** (0.5)	16	89
4	**2c**	**3** (0.5)[b]	24	81
5	**2c**	**4** (0.25)	12	90
6	**2d**	**4** (0.5)	20	87
7	**2e**	**3** (0.5)[b]	24	75
8	**2e**	**4** (0.25)	24	81
9	**2f**	**3** (1)[b]	18	72
10	**2f**	**4** (0.5)	18	93
11	**2g**	**4** (0.5)	24	67[c]

[a] In refluxing acetone. [b] In the presence of K_2CO_3. [c] In this case the isomeric 5,6-unsaturated ketone was also isolated in 28% yield.

(16) (a) Bäckvall, J. E.; Chowdhury, R. L.; Karlsson, U.; Wang, G.-Z. In *Perspectives in Coordination Chemistry*; Williams, A. F., Floriani, C., Merbach, A. E., Eds.; Verlag Hel. Chim. Acta: Basel, 1992; p 463. (b) Wang, G.-Z.; Bäckvall, J. E. *J. Chem. Soc., Chem. Commun.* **1992**, 337. (c) Karlsson, U.; Wang, G.-Z.; Bäckvall, J. E. *J. Org. Chem.* **1994**, *59*, 1196. (d) Wang, G.-Z.; Andreasson, U.; Bäckvall, J. E. *J. Chem. Soc., Chem. Commun.* **1994**, 1037. (e) Bäckvall, J. E.; Chowdhury, R. L.; Karlsson, U. *J. Chem. Soc., Chem. Commun.* **1991**, 473.

(17) Almeida, M. L. S.; Beller, M.; Wang, G.-Z.; Bäckvall, J. E. *Chem. Eur. J.*, in press.

(18) Murahashi, S. I.; Naota, T.; Hirai, N. *J. Org. Chem.* **1993**, *58*, 7318.

(Table 1, entry 6). The alcohols **1e** and **1f** were easily oxidized to androstenedione (**2e**) and 17α-methyltestosterone (**2f**), respectively, in good yields and clean manner with either of the catalytic systems **3**/K_2CO_3 or **4** (Table 1, entries 7−10).

Finally, the corticoid substrate **1g** reacted slowly and was mainly recovered when using the catalytic system with complex **3**. In the presence of catalyst **4**, this compound was converted to a mixture of cortexolone acetate (**2g**) and the isomeric 5,6-unsaturated ketone. These two isomers were easily separated by chromatography, and the product **2g** was obtained in good yield (Table 1, entry 11). It is important to mention that, under our reaction conditions, no rearrangement to the 6-membered D-ring[13] was observed.

In summary, a mild and efficient procedure was developed for the catalytic oxidation of steroids. Acetone, which is inexpensive and unreactive toward most organic functional groups, acts both as solvent and as hydrogen acceptor in this homogeneous hydrogen transfer reaction. Employing this method, only a catalytic amount (≤1 mol %) of the ruthenium complex is necessary. Moreover, under these mild conditions, no side reactions or D-homosteroid rearrangement occur and no decomposition products were detected. Thus, the procedure described offers significant advantages over previous oxidation methods.

Experimental Section

General Methods. Melting points were determined on a Kofler block and are uncorrected. NMR spectra were recorded in $CDCl_3$ using a Varian 300 spectrometer, ^1H at 300 MHz and ^{13}C at 75.4 MHz with chloroform-d_1 (δ 7.26, ^1H; δ 77.0, ^{13}C) as internal standard. Yields are given for isolated product showing one spot on a chromatographic plate and no impurities detectable in the NMR spectrum. The identity of the products was checked by comparing their mp, IR and NMR spectra, and TLC behavior with those of authentic samples. All reactions were performed in oven-dried glassware, under an atmosphere of nitrogen. Solvents and solutions were transferred by syringe−septum and cannula techniques. Acetone (Baker, grade reagent) was used without further purification and was degassed by bubbling through a stream of nitrogen for 10 min prior to cannula transfer. The steroids **1a**−**g** are commercially available from Sigma, Merck, and Steraloids. The ruthenium complex **3** was purchased from Strem Chem, and complex **4** was prepared according to a literature procedure.[19]

General Procedure for Ruthenium-Catalyzed Oxidation of Steroids by Acetone. The substrate, catalyst **3**, and K_2CO_3, or the substrate and catalyst **4**, were weighed into a two-necked round bottom flask equipped with a condenser and a magnetic stirring bar. The reaction system was purged with nitrogen for 10 min, and then acetone (containing 0.6% of water when using complex **3**) was added via a cannula. The resulting solution was refluxed (56 °C) with stirring under nitrogen for the indicated time, until TLC showed the starting material to be consumed. The reaction mixture was cooled and the solvent evaporated. The resulting crude mixture was then chromatographed on silica, using the solvent system indicated, allowing isolation of the products.

Cholest-4-en-3-one (2a). Oxidation of cholest-5-en-3β-ol (**1a**, 387 mg, 1 mmol) with **4** (5.4 mg, 0.0050 mmol) in acetone (5 mL) for 22 h gave 286 mg (74%) of the product after flash chromatography (CH_2Cl_2/ether, 20:1). The spectral data of **2a** agree with those reported:[20] ^1H NMR δ 5.753 (s, 1H), 2.224−2.483 (several m, 4H), 2.048−1.951 (m, 2H), 1.876−1.745 (m, 2H), 1.748−1.2191 (several m, 12H), 1.203 (s, 3H), 0.913 (d, J = 6Hz, 3H), 0.864 (dd, J = 9, 3 Hz, 6H), 1.187−0.943 (several m, 8H), 0.713 (s, 3H); ^{13}C NMR δ 199.66, 171.69, 123.72, 56.07, 55.84, 53.78, 42.36, 39.60, 39.47, 38.58, 36.09, 35.73, 35.67, 35.59, 33.97, 32.94, 32.02, 28.16, 27.99, 24.16, 23.79, 22.81, 22.55, 21.00, 18.62, 17.36, 11.93.

(24*R*)-24-Ethylcholesta-4,22-dien-3-one (2b). Oxidation of (24*R*)-24-ethylcholesta-5,22-dien-3β-ol (**1b**, 413 mg, 1.0 mmol) with **4** (5.4 mg, 0.0050 mmol) in acetone (10 mL) for 16 h gave 366 mg (89%) of the product after flash chromatography (CH_2Cl_2/ether, 20:1). The spectral data of **2b** agree with those reported:[21] ^1H NMR δ 5.712 (s, 1H), 5.139 (dd, J = 15, 8.4 Hz, 1H), 5.008 (dd, J = 15, 8.4 Hz, 1H), 2.423−2.263 (several m, 4H), 2.048−1.980 (m, 3H), 1.848−1.371 (several m, 10H), 1.284−0.888 (several m, 8H), 1.175 (s, 3H), 1.009 (d, J = 6.5 Hz, 3H), 0.848−0.770 (complex m, 9H), 0.719 (s, 3H); ^{13}C NMR δ 199.64, 171.66, 138.12, 129.40, 123.73, 55.95, 55.85, 53.80, 42.24, 40.48, 39.49, 38.59, 35.67, 35.58, 33.97, 32.93, 32.01, 31.86, 28.87, 25.40, 24.23, 21.15, 21.10, 20.99, 18.97, 17.37, 12.25, 12.13.

Pregn-4-ene-3,20-dione (2c). Oxidation of 3-β-hydroxypregn-5-en-20-one (**1c**, 119 mg, 0.376 mmol) with **3** (1.8 mg, 0.0019 mmol) and K_2CO_3 (5.2 mg, 0.0376 mmol) in acetone (3 mL, containing 0.6% of water) for 24 h gave 96 mg (81%) of the product after flash chromatography (CH_2Cl_2/ether, 10:1). The product was characterized by comparison with an authentic sample:[22] ^1H NMR δ 5.738 (s, 1H), 2.562−2.517 (m, 1H), 2.481−2.146 (several m, 5H), 2.128 (s, 3H), 2.093−2.015 (m, 2H), 1.897−1.838 (m, 1H), 1.764−0.954 (complex m, 11H), 1.192 (s, 3H), 0.673 (s, 3H); ^{13}C NMR δ 209.21, 199.35, 170.84, 123.91, 63.47, 56.00, 53.62, 43.89, 38.64, 38.54, 35.70, 35.53, 33.92, 32.74, 31.86, 31.45, 24.33, 22.81, 20.99, 17.34, 13.30.

17β-(Benzoyloxy)androst-4-en-3-one (2d). Oxidation of 17β-(benzoyloxy)androst-5-en-3β-ol (**1d**, 100 mg, 0.253 mmol) with **4** (1.4 mg, 0.0013 mmol) in acetone (4 mL) for 20 h gave 87 mg (87%) of the product after flash chromatography (CH_2Cl_2/ether, 10:1). The spectral data of **2d** agree with those reported:[23] ^1H NMR δ 8.035 (dd, J = 7.5, 1.5 Hz, 2H), 7.555 (tt, J = 7.5, 1.5 Hz, 1H), 7.437 (t, J = 7.5 Hz, 2H), 5.735 (s, 1H), 4.848 (t, J = 7.8 Hz, 1H), 2.430−2.252 (several m, 5H), 2.064−1.992 (m, 1H), 1.905−1.845 (m, 2H), 1.762−1.555 (several m, 5H), 1.490−1.363 (m, 2H), 1.310−1.048 (several m, 3H), 1.201 (s, 3H), 1.035−0.910 (m, 1H), 0.980 (s, 3H); ^{13}C NMR δ 199.44, 170.91, 166.43, 132.80, 130.57, 129.47, 128.30, 123.93, 82.92, 53.66, 50.24, 42.84, 38.58, 36.68, 35.66, 35.38, 33.90, 32.71, 31.46, 27.63, 23.58, 20.51, 17.38, 12.26.

Androst-4-ene-3,17-dione (2e). Oxidation of 3β-hydroxyandrost-5-en-17-one (**1e**, 200 mg, 0.69 mmol) with **4** (2 mg, 0.0017 mmol) in acetone (4 mL) for 24 h gave 162 mg (81%) of the product after flash chromatography (CH_2Cl_2/ether, 10:1). The spectral data of **2e** agree with those reported:[24] ^1H NMR δ 5.731 (s, 1H), 2.508−2.284 (several m, 5H), 2.148−1.328 (several m, 10H), 1.197 (s, 3H), 1.309−0.929 (several m, 4H), 0.901 (s, 3H); ^{13}C NMR δ 220.37, 199.26, 170.27, 124.09, 53.75, 50.77, 47.45, 38.58, 35.70, 35.64, 35.08, 33.86, 32.51, 31.21, 30.69, 21.70, 20.26, 17.33, 13.66.

17β-Hydroxy-17-methylandrost-4-en-3-one (2f). Oxidation of 17β-hydroxy-17-methylandrost-5-en-3β-ol (**1f**, 200 mg, 0.66 mmol) with **4** (3.6 mg, 0.0033 mmol) in acetone (5 mL) for 18 h gave 186 mg (93%) of the product after flash chromatography (CH_2Cl_2/ether, 5:1). The spectral data of **2f** agree with those reported:[25] ^1H NMR δ 5.711 (s, 1H), 2.421−2.236 (several m, 4H), 2.057−1.984 (m, 1H), 0.855−1.870 (several m, 15H), 1.198 (s, 3H), 1.187 (s, 3H), 0.891 (s, 3H); ^{13}C NMR δ 199.56, 171.29, 123.80, 81.43, 53.74, 50.08, 45.29, 38.80, 38.62, 36.41, 35.68, 33.92, 32.80, 31.64, 31.36, 25.77, 23.17, 20.62, 17.36, 13.86.

(19) Shvo, Y.; Menashe, N. *Organometallics* **1991**, *10*, 3885.
(20) Parish, E. J.; Honda, H.; Chitrakorn, S.; Livant, P. *Lipids* **1991**, *26*, 675.
(21) Takatsuto, S.; Ikekawa, N. *J. Chem. Soc., Perkin Trans. 1* **1986**, 2269.
(22) Duddeck, H.; Rosenbaum, D.; Hani, M.; Elgamal, A.; Fayez, M. B. E. *Magn. Reson. Chem.* **1987**, *25*, 25.
(23) Gravestock, M. B.; Morton, D. R.; Boots, S. G.; Johnson, W. S. *J. Am. Chem. Soc.* **1980**, *102*, 800.
(24) Kirk, D. N.; Toms, H. C.; Douglas, C.; White, K. A.; Smith, K. E.; Latif, S.; Hubbard, R. W. P. *J. Chem. Soc., Perkin Trans. 2* **1990**, 1567.
(25) Hayamizu, K.; Ishii, T.; Yanagisawa, M.; Kamo. O. *Magn. Reson. Chem.* **1990**, *28*, 250.

21-Acetoxy-17α-hydroxypregn-4-en-3,20-dione (2g). Oxidation of 21-acetoxy-3β,17α-dihydroxypregn-5-en-20-one (**1g**, 150 mg, 0.384 mmol) with **4** (2.1 mg, 0.0019 mmol) in acetone (7−10 mL) for 24 h gave 100 mg (67%) of the product and 42 mg (28%) of the isomeric β,γ-unsaturated ketone after flash chromatography (CH_2Cl_2/ether, 3:1). The product was characterized by comparison with an authentic sample:[26] ^{1}H NMR δ 5.719 (s, 1H), 5.078 and 4.835 (AB-system, J = 17.7 Hz, 2H), 2.762−2.666 (m, 2H), 2.431−2.240 (several m, 4H), 2.157 (s, 3H), 2.063−1.992 (m, 1H), 1.878−1.318 (complex m, 11H), 1.175 (s, 3H), 1.109−0.931 (m, 2H), 0.704 (s, 3H); ^{13}C NMR δ 205.26, 199.70, 171.20, 170.66, 123.86, 89.91, 67.96, 53.29, 50.43, 48.21, 38.56, 35.67, 35.61, 34.74, 33.88, 32.77, 32.00, 30.02, 23.58, 20.61, 20.52, 17.34, 14.49.

(26) Hofmeister, H.; Annen K.; Laurent, H.; Wiechert, R. *Lieb. Ann. Chem.* **1987**, 423.

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