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$SnCl_4$-Mediated Reactions of Cyclopropyl Alkyl Ketones with α-Keto Esters

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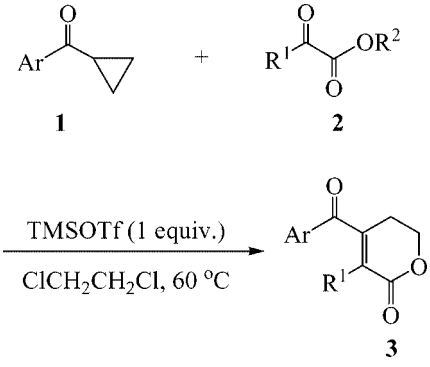
Keywords: Cyclopropyl alkyl ketones / Lewis acids / α-Keto esters / Spiro-γ-lactones

This paper reports the full details of Lewis acid $SnCl_4$-mediated reactions of cyclopropyl alkyl ketones with α-keto esters under mild conditions. In general, it was found that four products were formed in the $SnCl_4$-mediated reactions of cyclopropyl alkyl ketones. However, by controlling the aldol reaction process, the corresponding spiro-γ-lactone products can be exclusively obtained in moderate to good yields with good stereoselectivities via the sequential nucleophilic ring-opening reaction of cyclopropane by H_2O, aldol type reaction and subsequent cyclic transesterification mediated by the Lewis acid. The mechanism was further confirmed by introducing a substituent at the other α-position of the cyclopropyl alkyl ketone to produce two or three other products without the formation of spiro-γ-lactone products under the same reaction conditions. In summary, $SnCl_4$-mediated reactions of cyclopropyl alkyl ketones with α-keto esters afforded a new general method for the synthesis of spiro-γ-lactones in moderate to good yields with good stereoselectivities.

Introduction

Cyclopropane derivatives, as versatile building blocks, have been more than laboratory curiosities for quite some time.[1] In order to activate strained three-membered rings, electron donating or withdrawing substituents are generally involved in their reactions to make polar processes more favorable. In general, cyclopropanes involved in synthetically useful reactions frequently contain two activating groups.[2] However, ring-opening reactions of monoactivated cyclopropane derivatives are generally sluggish. So far, the only reported examples have required severe conditions either assisted by strong nucleophiles such as the iodide anion[3] and strong Lewis acids such as $TiCl_4$,[4] or assisted by the β-effect of the silicon atom of a trimethylsilyl group.[5] In addition, Bu_3SnH mediated free-radial process and SmI_2-promoted electron transfer process have been also reported in this transformation.[6] More recently, cycloaddition processes involving simple cyclopropyl ketones catalyzed by nickel(0) complexes such as $Ni(cod)_2$ have also been developed by Kurosawa and Montgomery, respectively.[7] Our specific aim is to develop new methods for ring-opening reactions of simple monoactivated cyclopropane derivatives under mild conditions in the presence of easily handled and available Lewis acid catalysts.

On the basis of this concept, we recently developed a cascade process involving the ring-opening of monoactivated cyclopropyl aryl ketones **1** by H_2O, followed by a transesterification reaction with α-keto esters **2** and an aldol type reaction mediated by a Lewis acid. The cascade process provided an efficient synthetic protocol for the preparation of 5,6-dihydropyran-2-ones **3**,[8] which are an important class of compounds because they are skeletal motifs in many natural products possessing important biological activities (Scheme 1).[9] A natural extension of this work is to explore the possibility of replacing the corresponding cyclopropyl aryl ketones with cyclopropyl alkyl ketones in the reactions to get more specific 5,6-dihydropyran-2-one derivatives. A short communication on $SnCl_4$-mediated reactions of cyclopropyl alkyl ketones with α-keto esters to afford a novel method for the syntheses of 1,6-dioxaspiro[4.4]non-3-en-2-ones with high stereoselectivities in moderate to good yields by controlling the aldol reaction process has been previously published.[10] In this paper, we wish to report the full details of the Lewis acid-mediated reactions of cyclopropyl alkyl ketones with α-keto esters along with the



Scheme 1. Lewis acid TMSOTf-mediated reaction of cyclopropyl aryl ketones **1** with α-keto esters **2** to give the corresponding 5,6-dihydropyran-2-ones **3**.

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examination of the reaction mechanism on the basis of introducing a substituent at another α-position of the cyclopropyl alkyl ketone.

Results and Discussion

Initially, we attempted to examine the reaction of cyclopropyl hexyl ketone, a widely available cyclopropyl alkyl ketone, with methyl benzoylformate (**2a**) in the presence of the Lewis acid TMSOTf (1.0 equiv.). Surprisingly, it was found that the reaction not only afforded the expected product 5,6-dihydropyran-2-one **3a** in 21% yield, but also gave three unexpected additional products, namely 9-hexylidene-1,9-dihydro-2*H*-3-oxafluoren-4-one (**4a**), 5-hexylidene-4-(2-hydroxyethyl)-3-phenyl-*5H*-furan-2-one (**5a**), and spiro-γ-lactone compound (**6a**) in 15, 45, and 16% yield, respectively (Scheme 2). Their structures were determined on the basis of NMR spectroscopic and other analytic data. In order to confirm the structure of the spiro-γ-lactone product **6a**, we attempted to determine its structure by X-ray crystal diffraction with a solid spiro-γ-lactone derivative. Fortunately, a single crystal of the spiro-γ-lactone **6b** was obtained from the reaction of cyclopropyl benzyl ketone **1b** with **2a** under identical conditions (Scheme 2). The ORTEP drawing of **6b** is shown in Figure 1.[11]

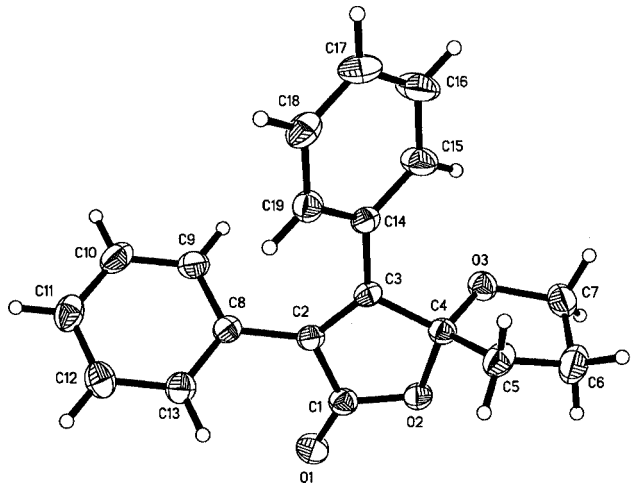
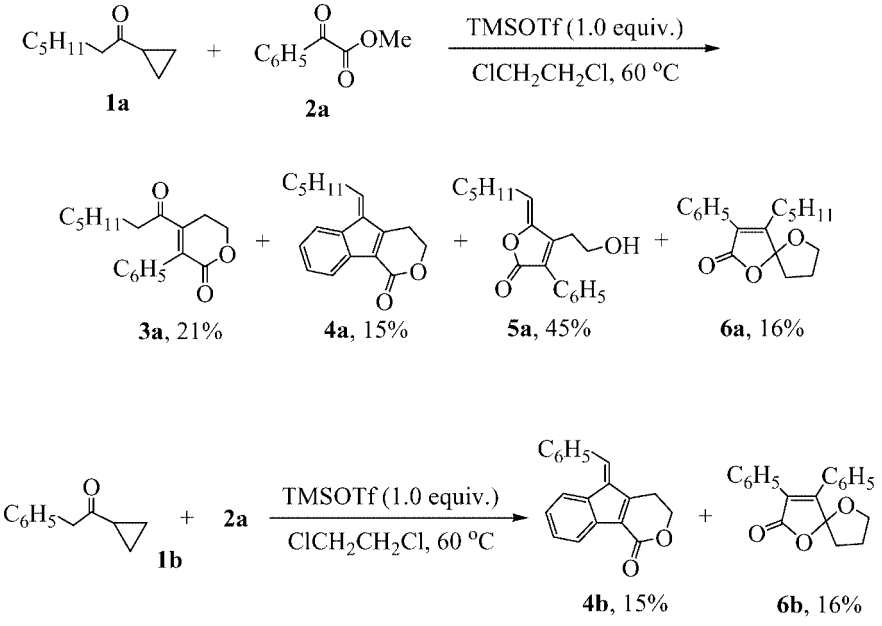


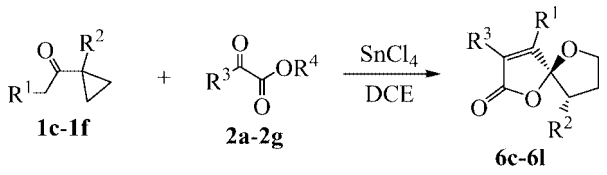
Figure 1. ORTEP drawing of **6b**.

In our previous communication,[10] we introduced a substituent at the α-position of the carbonyl group in the cyclopropyl alkyl ketone and successfully synthesized a series of cyclopropyl ketones **1c**–**1f** with a substituent R^2 at the cyclopropyl moiety in moderate to good yields (see the Supporting Information). The reactions of all these substituted cyclopropyl alkyl ketones with α-keto esters in the presence of $SnCl_4$ (1.0 equiv.) produced the corresponding spiro-γ-lactone products **6c**–**6l** with *trans* stereoselectivities (*S**,*R**) as the sole products, in which R^1, R^2, and R^3 could be either aromatic or aliphatic groups, without the formation of the other three products (Scheme 3). In addition, the stereochemistry of these products was determined by X-ray crystal diffraction.[10] In order to elucidate the scope and limitations of Lewis acid-mediated reaction of cyclopropyl alkyl ketones with α-keto esters, other types of cyclopropyl alkyl ketones were examined along with a detailed investigation of the reaction mechanism.

Previously, Ranfaing and Pittman independently reported the rearrangement reactions of simple cyclopropyl ketones **1** in concd. sulfuric acid to produce oxolan-2-ylium ions **7**, 2,3-dihydro-furans **8**, and γ-hydroxy ketones **9** as hydrolysis products (Scheme 4).[12] On the basis of our results above and previous investigations, we propose that all of the four products in the reaction of cyclopropyl hexyl ketone with methyl benzoylformate (**2a**) shown in Scheme 2 are formed via the same hydrolysis intermediate **9a**, which



Scheme 2. Reaction of cyclopropyl alkyl ketones **1a** and **1b** with α-keto ester **2a** mediated by TMSOTf.



6c: 72%, R^1 = nPr, R^2 = Ph, R^3 = Ph

6d: 85%, R^1 = nPr, R^2 = Ph, R^3 = *p*-ClC_6H_4

6e: 77%, R^1 = nPr, R^2 = Ph, R^3 = *p*-FC_6H_4

6f: 66%, R^1 = nPr, R^2 = Ph, R^3 = *p*-MeC_6H_4

6g: 41%, R^1 = nPr, R^2 = Ph, R^3 = *p*-$MeOC_6H_4$

6h: 74%, R^1 = nPr, R^2 = Ph, R^3 = EtO_2C

6i: 55%, R^1 = nPr, R^2 = Ph, R^3 = Me

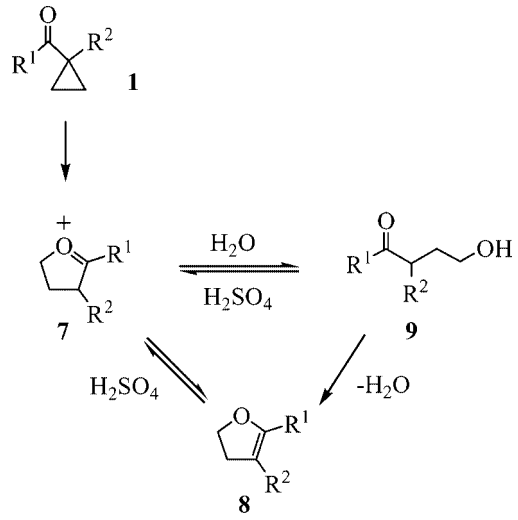
6j: 63%, R^1 = nPr, R^2 = Me, R^3 = Ph

6k: 67%, R^1 = H, R^2 = Ph, R^3 = Ph

6l: 72%, R^1 = Ph, R^2 = Ph, R^3 = Ph

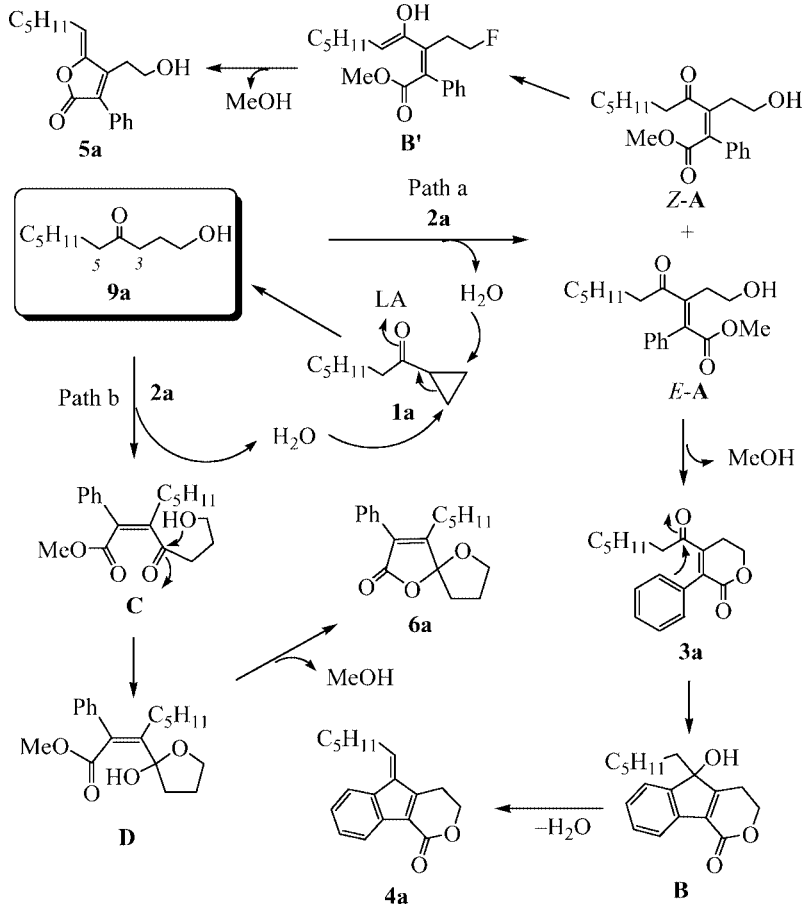
Scheme 3. Synthesis of spiro-γ-lactones from substituted cyclopropyl alkyl ketones and α-keto esters.

is formed from cyclopropyl hexyl ketone and ambient H_2O in the presence of a Lewis acid.



Scheme 4. Rearrangement of cyclopropyl ketones in concd. sulfuric acid.

Therefore, a comprehensive mechanistic picture can be provided on the basis of previous investigations and our own results. As shown in Scheme 5, in the presence of a Lewis acid, cyclopropyl hexyl ketone **1a** reacts with ambient H_2O to give the corresponding adduct γ-hydroxy ketone **9a**. Starting from the hydrolysis intermediate **9a**, if an aldol type reaction occurs at the C-3 position (Path a), intermedi-



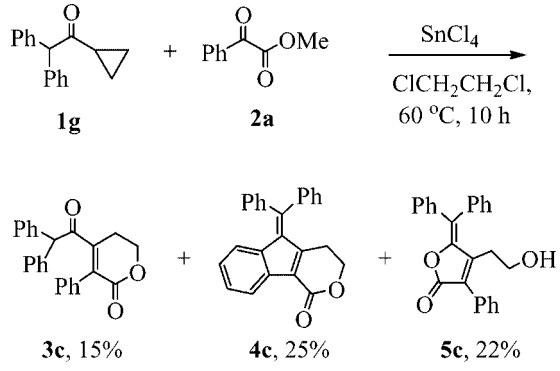
Scheme 5. Proposed mechanism for the reaction of cyclopropyl hexyl ketone with methyl benzoylformate.

ate **A** could be formed as a mixture of *E*- and *Z*-isomers. Thus, product **9a** can be formed after an intramolecular transesterification process via intermediate *E*-**A**.[8] The highly conjugated product *Z*-**4a** can be formed by dehydration of intermediate **B**, which itself can be derived from a Bradsher-type cyclization reaction of **3a**. On the other hand, product *Z*-**5a** can be formed from Z-**A** via the enolate intermediate **B′** after an intramolecular transesterification. Alternatively, if the aldol-type reaction takes place at the C-5 position in the γ-hydroxy ketone **9a**, another intermediate **C**, would be formed along with regeneration of an equivalent of H_2O, which can react with **1a** to initiate the next reaction cycle. Intramolecular nucleophilic attack by the terminal hydroxy group at the ketone group can take place, leading to the corresponding cyclic intermediate **D** containing a hemiacetal hydroxy group. From intermediate **D**, the corresponding product 1,6-dioxaspiro[4.4]non-3-en-2-one **6a** can be formed via intramolecular transesterification. Overall, the formation of the spiro-γ-lactone product **6a** is a cascade reaction involving the nucleophilic ring-opening reaction of the cyclopropane by H_2O, an aldol-type reaction and an intramolecular transesterification reaction in the presence of a Lewis acid. Moreover, the ambient water is required to initiate the process, but additional H_2O is not necessary because the H_2O is regenerated in situ in the subsequent aldol-type reaction.

Another plausible mechanism for the formation of the key intermediate is that Lewis acid-catalyzed rearrangements of cyclopropyl ketones take place to give the corresponding dihydrofurans, which equilibrate with the oxolan-2-ylium inos **7**. Subsequently, the latter then undergo hydrolysis to furnish the corresponding key intermediates **9** in Scheme 5, although at the present stage, we can not determine which is the dominant reaction mechanism in the process.

In Scheme 3, all the substrates with a substituent at the cyclopropyl position adjacent to the carbonyl group could give the corresponding intermediates **9** containing a substituent R^2 at the C-3 position. This substituent would prohibit the reaction shown in Path a because the formation of the corresponding thermodynamically favored intermediate *E*-**A** with the release of water is prohibited, which is necessary for the initiation of the reaction shown in Path a. Therefore, when R^2 in Scheme 3 is not a hydrogen atom, all of the reactions can take place along Path b to give the corresponding spiro-γ-lactone products exclusively as the sole products.

As a reasonable proposal, according to the mechanism shown in Scheme 5 if we introduce a substituent at one of the other α-positions of the cyclopropyl alkyl ketone, it would produce three products without the formation of a spiro-γ-lactone. Therefore, we further examined the reaction of methyl benzoylformate (**2a**) with the cyclopropyl alkyl ketone **1g** substituted in α-position (C-5) in the presence of $SnCl_4$ (1.0 equiv.) (Scheme 6). As expected, this reaction produced three products, namely 4-diphenylacetyl-3-phenyl-5,6-dihydropyran-2-one (**3c**), 9-benzhydrylidene-1,9-dihydro-2*H*-3-oxafluoren-4-one (**4c**), and 5-benzhydrylidene-4-(2-hydroxyethyl)-3-phenyl-5*H*-furan-2-one (**5c**) in 15, 25 and 22% yield, respectively. We did not detect the corresponding spiro-γ-lactone product in the reaction mixture. The structures of products **4c** and **5c** were further confirmed by X-ray crystallographic analyses. Their ORTEP drawings are shown in Figure 2 and Figure 3, respectively.[13] Lewis acid effects were also examined in the presence of TMSOTf, $TiCl_4$, $Sn(OTf)_2$, and $BF_3{\cdot}OEt_2$ under otherwise identical conditions in a similar manner as those reported before.[10] We found that $SnCl_4$ is the best catalyst in this reaction.



Scheme 6. Reaction of cyclopropyl diphenylmethyl ketone with methyl benzoylformate mediated by $SnCl_4$.

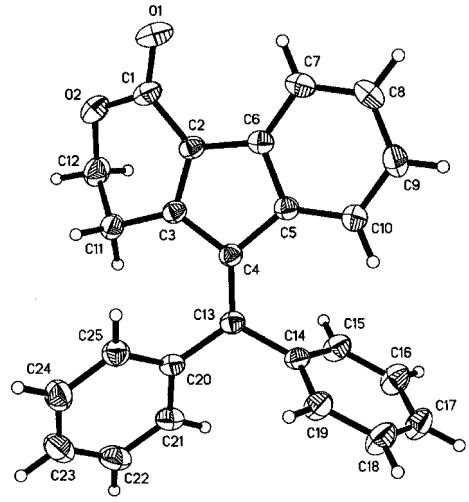


Figure 2. ORTEP drawing of **4c**.

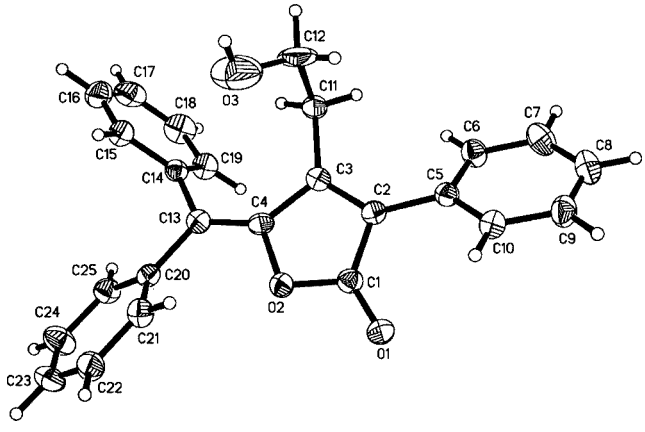
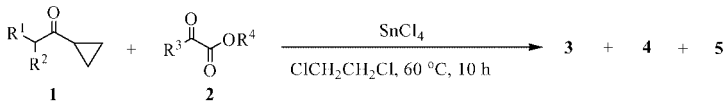


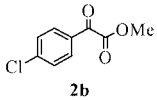
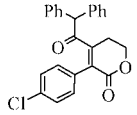
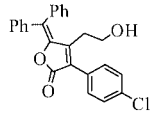
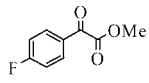
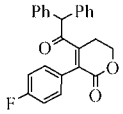
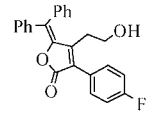
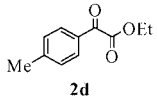
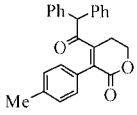
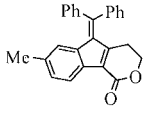
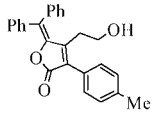
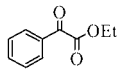
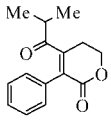
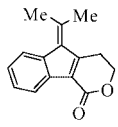
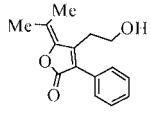
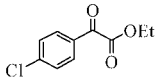
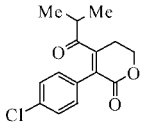
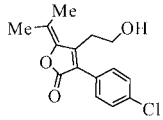
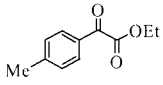
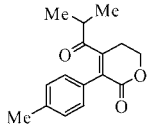
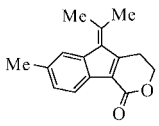
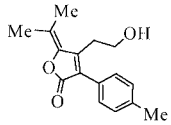
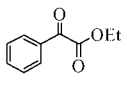
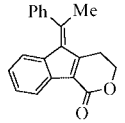
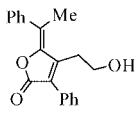
Figure 3. ORTEP drawing of **5c**.

Moreover, the above reaction was found to be quite general under identical conditions. The reactions of other cyclopropyl alkyl ketones **1g**–**i** bearing a substituent at the α-position (C-5) with several α-keto esters also underwent the same reaction under standard conditions to give the corresponding products **3**, **4**, and **5** without the formation of spiro-γ-lactones (Table 1, entries 1–7). As for the series of aryl α-keto esters, an electronic effect was clearly observed. In general, aryl α-keto esters having an electron withdrawing group on the aromatic ring produced the corresponding 5,6-dihydropyran-2-ones **3** and 5*H*-furan-2-ones **5** in moderate yields, but the corresponding highly conjugated products **4** were not formed (Table 1, entries 1, 2 and 5). For the aryl α-keto ester **2d** bearing an electron donating substituent (methyl group) on the aromatic ring, the corresponding Bradsher-type cyclization products **4f** and **4i** were obtained in the highest yields among the three products (Table 1, entries 3 and 6). As a reaction sequence, product

Table 1. $SnCl_4$-mediated reactions of substituted cyclopropyl alkyl ketones **1g**–**1i** with various α-keto esters.

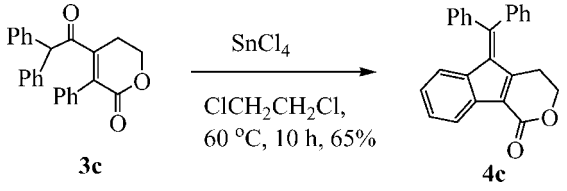


1 **2**

Entry	R^1, R^2	α-Keto ester	**3**	**4**	**5**
1	Ph, Ph **1g**	 **2b**	 **3d**, 58%	—	 **5d**, 16%
2	Ph, Ph **1g**	 **2c**	 **3e**, 38%	—	 **5e**, 32%
3	Ph, Ph **1g**	 **2d**	 **3f**, 17%	 **4f**, 44%	 **5f**, 29%
4	Me, Me **1h**	 **2a**	 **3g**, 12%	 **4g**, 37%	 **5g**, trace
5	Me, Me **1h**	 **2b**	 **3h**, 42%	—	 **5h**, trace
6	Me, Me **1h**	 **2d**	 **3i**, trace	 **4i**, 29%	 **5i**, trace
7	Ph, Me **1i**	 **2a**	—	 **4j** (*Z*/*E* = 1/2), 63%	 **5j**, trace

[a] Isolated yields.

4 was proved to be derived from the corresponding product **3** in the presence of a Lewis acid via a Bradsher-type or intramolecular Friedel–Crafts reaction by the control experiment shown in Scheme 7.[14] Therefore, the results above are consistent with the nature of a Bradsher-type or intramolecular Friedel–Crafts reaction. The reaction of 1-phenylethyl cyclopropyl ketone **1i** with methyl benzoylformate (**2a**) under the same reaction conditions gave mainly product **4j** (*Z*:*E* = 1:2) in 64% yield and a trace of the 5*H*-furan-2-one **5j**, presumably because the Bradsher-type reaction is facilitated, in this case, in the presence of $SnCl_4$ due to steric effects and the stability of the related cationic intermediate (Table 1, entry 7). The C–C double bond configuration of **4** and **5** was determined by NOESY spectra (see the Supporting Information).



Scheme 7. Control experiment for the transformation of **3c** to **4c** in the presence of $SnCl_4$ (1.0 equiv.) in which 18% of **3c** was recovered.

The role of H_2O in the process was confirmed by introducing 4 Å molecular sieves to the reaction mixture to remove traces of water. When 60 mg of 4-Å molecular sieves was introduced to a 0.2 mmol scale reaction mixture containing cyclopropyl ketone **1c** with methyl benzoylformate (**2a**), the reaction became sluggish and gave only a trace of product **6c** after one day at 40 °C in the presence of $SnCl_4$. Hence, indicating that H_2O plays a key role in the initiation step of the reaction sequence.[10]

In conclusion, we have found an $SnCl_4$-mediated reaction of cyclopropyl alkyl ketones with α-keto esters which gives different products depending on the substituent at the α-positions of the cyclopropyl alkyl ketone. In general, four products were formed in the reaction. By controlling the aldol process, the corresponding spiro-γ-lactones can be exclusively obtained in moderate to good yields with good stereoselectivities via a cascade reaction of nucleophilic ring-opening of cyclopropane by H_2O, aldol type reaction and subsequent cyclic transesterification mediated by a Lewis acid. Efforts are underway to further elucidate the reaction mechanism and to understand the scope and limitations of this process.

Experimental Section

General Methods: Melting points are uncorrected. ^1H and ^{13}C NMR spectra were recorded using a Bruker AM-300 spectrometer at 300 and 75 MHz, respectively. Mass spectra were recorded by EI and ESI methods and HRMS was measured using a Kratos Analytical Concept mass spectrometer (EI), IonSpec 4.7 Tesla FTMS (MALDI). Some of the compounds reported in this paper gave satisfactory CHN microanalyses with a Carlo–Erba 1106 analyzer. Commercially obtained reagents were used without further purification. All the reactions were monitored by TLC with Yinlong GF254 silica gel coated plates. Flash column chromatography was carried out using 300–400 mesh silica gel at increased pressure.

General Procedure for the Preparation of 1,6-Dioxaspiro[4.4]non-3-en-2-ones from Cyclopropyl Alkyl Ketones 1c–1f and α-Keto Esters: In an argon atmosphere, a mixture of cyclopropyl ketone **1** (0.4 mmol), α-keto ester **2** (0.2 mmol), and $SnCl_4$ (0.2 mmol) were dissolved in 1,2-dichloroethane (DCE, 3.0 mL) and the reaction mixture was heated to 40 °C until **2** was consumed. The reaction mixture was cooled to room temp. and then quenched by the addition of aqueous $NaHCO_3$ solution. The reaction mixture was extracted using dichloromethane (3 × 15 mL) and dried with anhydrous $MgSO_4$. The solvent was then removed under reduced pressure and the residue was purified by a silica gel column chromatography using petroleum ether/ethyl acetate as an eluent to give the corresponding 1,6-dioxaspiro[4.4]non-3-en-2-one.

General Procedure for the Reaction of Cyclopropyl Alkyl Ketones 1g–1i and α-Keto Esters: In an argon atmosphere, a mixture of cyclopropyl ketone **1** (0.2 mmol), α-keto ester **2** (0.2 mmol), and $SnCl_4$ (0.2 mmol) in 1,2-dichloroethane (3.0 mL) was heated at 60 °C for 10 h. The reaction mixture was cooled to room temp. and then quenched by the addition of aqueous $NaHCO_3$ solution. The reaction mixture was extracted using dichloromethane (3 × 15 mL) and dried with anhydrous $MgSO_4$. The solvent was then removed under reduced pressure and the residue was purified by a silica gel column chromatography using petroleum ether/ethyl acetate.

4-Heptanoyl-3-phenyl-5,6-dihydropyran-2-one (3a): This compound was obtained as a red oil, yield 12 mg, 21%. IR (CH_2Cl_2): $\tilde{\nu}$ = 1007, 1086, 1116, 1161, 1200, 1257, 1317, 1400, 1445, 1467, 1720, 2858, 2929 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.81 (t, J = 7.5 Hz, 3 H, CH_3), 0.96–1.09 (m, 4 H, CH_2), 1.14–1.37 (m, 4 H, CH_2), 1.98 (t, J = 7.2 Hz, 2 H, CH_2), 2.77 (t, J = 6.3 Hz, 2 H, CH_2), 4.54 (t, J = 6.3 Hz, 2 H, OCH_2), 7.27–7.31 (m, 2 H, Ar), 7.36–7.40 (m, 3 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 13.9, 22.3, 23.5, 26.3, 28.4, 31.2, 42.0, 66.3, 128.4, 129.2, 129.9, 130.6, 133.3, 150.6, 164.4, 206.1 ppm. MS (EI): *m*/*z* (%) = 286 (23) [M^+], 257 (21), 229 (94), 173 (23), 155 (13), 128 (43), 115 (35), 43 (100). HRMS (EI) calcd. $(C_{18}H_{22}O_3)^+$: 286.1569; found: 286.1569.

(*Z*)-9-Hexylidene-1,9-dihydro-2*H*-3-oxafluoren-4-one (*Z*-4a): This compound was obtained as a yellow solid, yield 8.0 mg, 15%, m.p. 84–86 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1072, 1137, 1173, 1311, 1412, 1463, 1637, 1719, 2854, 2924, 2955 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.94 (t, J = 7.2 Hz, 3 H, CH_3), 1.38–1.45 (m, 4 H, CH_2), 1.62–1.71 (m, 2 H, CH_2), 2.82–2.90 (m, 4 H, CH_2), 4.57 (t, J = 6.3 Hz, 2 H, OCH_2), 6.63 (t, J = 7.8 Hz, 1 H, CH), 7.26–7.36 (m, 2 H, Ar), 7.72 (d, J = 7.8 Hz, 1 H, Ar), 7.97 (d, J = 7.2 Hz, 1 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 14.0, 21.8, 22.5, 28.8, 30.1, 31.7, 67.0, 121.8, 123.7, 125.4, 125.8, 128.0, 134.1, 136.0, 138.9, 143.0, 151.1, 163.1 ppm. MS (EI): *m*/*z* (%) = 268 (68) [M^+], 239 (5), 215 (10), 198 (100), 168 (96), 152 (46), 105 (52). HRMS (EI) calcd. $(C_{18}H_{20}O_2)^+$: 268.1463; found: 268.1463.

5-Hexylidene-4-(2-hydroxyethyl)-3-phenyl-5*H*-furan-2-one (5a): This compound was obtained as a red oil, yield 26 mg, 45%. IR (CH_2Cl_2): $\tilde{\nu}$ = 1003, 1057, 1446, 1666, 1763, 2858, 2928, 2954, 3424 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.90 (t, J = 6.3 Hz, 3 H, CH_3), 1.25–1.35 (m, 4 H, CH_2), 1.47–1.51 (m, 2 H, CH_2), 2.38–2.45 (m, 2 H, CH_2), 2.88 (t, J = 6.6 Hz, 2 H, CH_2), 3.85 (t, J = 6.6 Hz, 2 H, $HOCH_2$), 5.48 (t, J = 8.1 Hz, 1 H, CH), 7.37 (m, 3 H, Ar), 7.54–7.56 (m, 2 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 13.9, 22.4, 26.3, 28.4, 28.7, 31.4, 61.2, 114.6, 128.0, 128.5, 128.7, 128.9, 129.6, 147.5, 149.1, 169.2 ppm. MS (EI): *m*/*z* (%) = 286 (14) [M^+], 269 (1), 241 (3), 216 (15), 188

(13), 173 (13), 129 (14), 115 (54), 55 (100). HRMS (EI) calcd. $(C_{18}H_{22}O_3)^+$: 286.1569; found: 286.1561.

4-Pentyl-3-phenyl-1,6-dioxaspiro[4.4]non-3-en-2-one (6a): This compound was obtained as a pale oil, yield 9 mg, 16%. IR (CH_2Cl_2): $\tilde{\nu}$ = 1003, 1095, 1171, 1215, 1262, 1331, 1446, 1458, 1494, 1763, 2871, 2956 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.86 (t, J = 7.2 Hz, 3 H, CH_3), 1.22–1.29 (m, 4 H, CH_2), 1.54–1.59 (m, 2 H, CH_2), 2.16–2.40 (m, 5 H, CH_2), 2.51–2.56 (m, 1 H, CH_2), 4.09–4.16 (m, 1 H, OCH_2), 4.26–4.32 (m, 1 H, OCH_2), 7.38–7.47 (m, 5 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 13.8, 22.1, 24.5, 26.2, 27.3, 32.0, 34.5, 70.5, 113.8, 128.4, 128.7, 128.8, 128.9, 129.9, 159.4, 170.0 ppm. MS (EI): *m/z* (%) = 286 (11) [M^+], 241 (3), 215 (35), 187 (18), 141 (24), 115 (36), 71 (66), 57 (100), 43 (91). HRMS (EI) calcd. $(C_{18}H_{22}O_3)^+$: 286.1569; found: 286.1551.

(*Z*)-9-Benzylidene-1,9-dihydro-2*H*-3-oxafluoren-4-one (*Z*-4b): This compound was obtained as a red solid, yield 17 mg, 20%, m.p. 201–203 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1048, 1065, 1091, 1151, 1173, 1708, 1718 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.97 (t, J = 6.3 Hz, 2 H, CH_2), 4.61 (t, J = 6.3 Hz, 2 H, OCH_2), 7.04 (dt, J = 0.9 Hz, J = 7.5 Hz, 1 H, Ar), 7.26–7.31 (m, 1 H, Ar), 7.45–7.50 (m, 4 H, CH, Ar), 7.60–7.63 (m, 3 H, Ar), 7.93 (d, J = 7.5 Hz, 1 H, 2 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 21.9, 67.0, 121.7, 123.1, 125.8, 126.6, 128.7, 128.8, 129.5, 129.6, 133.2, 135.1, 137.4, 138.2, 139.3, 151.4, 162.8 ppm. MS (EI): *m/z* (%) = 274 (75) [M^+], 244 (17), 229 (29), 215 (100), 202 (9), 189 (10), 107 (31). $C_{19}H_{14}O_2$ (274.3133): calcd. C 83.19, H 5.14%; found C 83.46, H 5.25%.

3,4-Diphenyl-1,6-dioxaspiro[4.4]non-3-en-2-one (6b): This compound was obtained as a white solid, yield 8 mg, 14%, m.p. 138–140 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1001, 1060, 1107, 1166, 1264, 1294, 1316, 1351, 1445, 1489, 1763, 2887, 3059 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 1.98–2.13 (m, 2 H, CH_2), 2.20–2.44 (m, 2 H, CH_2), 4.12–4.23 (m, 1 H, OCH_2), 4.32–4.42 (m, 1 H, OCH_2), 7.27–7.44 (m, 10 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 24.5, 34.8, 70.6, 113.7, 128.3, 128.6, 128.7, 128.9, 129.0, 129.3, 129.5, 129.7, 130.9, 154.7, 169.7 ppm. MS (EI): *m/z* (%) = 292 (28) [M^+], 248 (15), 222 (9), 205 (9), 178 (100), 147 (14), 42 (40). $C_{19}H_{16}O_3$ (292.3285): calcd. C 78.06, H 5.52%; found C 77.82, H 5.58%.

3,6-Diphenyl-4-propyl-1-oxaspiro[4.4]non-3-en-2-one 6c: This compound was obtained as a white solid, yield 48 mg, 72%, m.p. 126–128 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1003, 1090, 1103, 1130, 1174, 1235, 1322, 1454, 1496, 1763, 2899, 2933, 2965 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.95 (t, J = 7.5 Hz, 3 H, CH_3), 1.57–1.67 (m, 2 H, CH_2), 2.37–2.51 (m, 2 H, CH_2), 2.61–2.70 (m, 1 H, CH_2), 2.72–2.83 (m, 1 H, CH_2), 3.58–3.65 (m, 1 H, CH), 4.18–4.27 (m, 1 H, OCH_2), 4.42–4.49 (m, 1 H, OCH_2), 7.16–7.19 (m, 2 H, Ar), 7.22–7.36 (m, 8 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 14.5, 21.0, 28.6, 29.6, 51.4, 68.8, 113.4, 127.9, 128.3, 128.55, 128.6, 128.7, 128.9, 129.7, 131.7, 134.1, 157.8, 169.1 ppm. MS (ESI): *m/z* (%) = 335 (100) [$(M + 1)^+$]. $C_{22}H_{22}O_3$ (334.4083): calcd. C 79.02, H 6.63%; found C 78.86, H 6.60%.

3-(4-Chlorophenyl)-9-phenyl-4-propyl-1,6-dioxaspiro[4.4]non-3-en-2-one (6d): This compound was obtained as a white solid, yield 63 mg, 85%, m.p. 125–127 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1091, 1176, 1213, 1233, 1267, 1322, 1454, 1494, 1741, 1759, 2873, 2927, 2964 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.95 (t, J = 7.5 Hz, 3 H, CH_3), 1.53–1.72 (m, 2 H, CH_2), 2.37–2.51 (m, 2 H, CH_2), 2.58–2.68 (m, 1 H, CH_2), 2.75–2.83 (m, 1 H, CH_2), 3.57–3.64 (m, 1 H, CH), 4.18–4.27 (m, 1 H, OCH_2), 4.42–4.48 (m, 1 H, OCH_2), 7.12 (d, J = 8.4 Hz, 2 H, Ar), 7.22–7.28 (m, 5 H, Ar), 7.31 (d, J = 8.4 Hz, 2 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 14.5, 20.9, 28.6, 29.5, 51.5, 68.9, 113.5, 127.9, 128.1, 128.3, 128.6, 128.8, 130.0, 130.5, 134.0, 134.6, 158.4, 168.8 ppm. MS (ESI): *m/z* (%) = 371 (35), 369 (100) [$(M + H)^+$]. HRMS (MALDI) calcd. $(C_{22}H_{21}ClO_3+H)^+$: 369.1252; found: 369.1258.

3-(4-Fluorophenyl)-9-phenyl-4-propyl-1,6-dioxaspiro[4.4]non-3-en-2-one (6e): This compound was obtained as a white solid, yield 54 mg, 77%, m.p. 126–128 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1002, 1102, 1130, 1160, 1175, 1234, 1323, 1510, 1603, 1760, 2965 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.89 (t, J = 7.5 Hz, 3 H, CH_3), 1.44–1.69 (m, 2 H, CH_2), 2.29–2.47 (m, 2 H, CH_2), 2.51–2.61 (m, 1 H, CH_2), 2.66–2.81 (m, 1 H, CH_2), 3.51–3.57 (m, 1 H, CH), 4.12–4.20 (m, 1 H, OCH_2), 4.36–4.42 (m, 1 H, OCH_2), 6.93–6.99 (m, 2 H, Ar), 7.07–7.12 (m, 2 H, Ar), 7.17–7.23 (m, 5 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 14.5, 20.9, 28.6, 29.5, 51.4, 68.9, 113.4, 115.5 ($J_{C\text{-}F}$ = 21.5 Hz), 125.6, 127.6, 127.9, 128.3, 128.8 ($J_{C\text{-}F}$ = 17.2 Hz), 130.6 ($J_{C\text{-}F}$ = 8.6 Hz), 134.1, 157.9, 163.1 ($J_{C\text{-}F}$ = 247.3 Hz), 169.0 ppm. MS (EI): *m/z* (%) = 352 (1) [M^+], 200 (1), 161 (1), 147 (2), 133 (6), 118 (100), 91 (11). $C_{22}H_{21}FO_3$ (352.1475): calcd. C 74.98, H 6.01%; found C 74.96, H 5.99%.

9-Phenyl-4-propyl-3-*p*-tolyl-1,6-dioxaspiro[4.4]non-3-en-2-one (6f): This compound was obtained as a pale-yellow solid, yield 46 mg, 66%, m.p. 120–122 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1002, 1104, 1129, 1174, 1235, 1322, 1455, 1514, 1761, 2925, 2962 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.95 (t, J = 7.2 Hz, 3 H, CH_3), 1.55–1.76 (m, 2 H, CH_2), 2.32 (s, 3 H, CH_3), 2.36–2.52 (m, 2 H, CH_2), 2.61–2.70 (m, 1 H, CH_2), 2.71–2.86 (m, 1 H, CH_2), 3.57–3.64 (m, 1 H, CH), 4.17–4.25 (m, 1 H, OCH_2), 4.41–4.47 (m, 1 H, OCH_2), 7.08 (d, J = 8.1 Hz, 2 H, Ar), 7.14 (d, J = 8.1 Hz, 2 H, Ar), 7.21–7.28 (m, 5 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 14.5, 21.0, 21.2, 28.6, 29.5, 51.3, 68.8, 113.3, 126.7, 127.8, 128.3, 128.6, 128.9, 129.0, 131.5, 134.2, 138.5, 157.1, 169.2 ppm. MS (EI): *m/z* (%) = 348 (1) [M^+], 261 (1), 203 (1), 158 (1), 143 (3), 129 (5), 118 (100), 103 (4), 91 (11). HRMS (MALDI) calcd. $(C_{23}H_{24}O_3+Na)^+$: 371.1618; found: 371.1621.

3-(4-Methoxyphenyl)-9-phenyl-4-propyl-1,6-dioxaspiro[4.4]non-3-en-2-one (6g): This compound was obtained as a white solid, yield 30 mg, 41%. m.p. 153–155 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1033, 1104, 1129, 1173, 1251, 1292, 1456, 1513, 1608, 1759, 2853, 2926, 2961 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.96 (t, J = 7.5 Hz, 3 H, CH_3), 1.56–1.73 (m, 2 H, CH_2), 2.35–2.50 (m, 2 H, CH_2), 2.61–2.71 (m, 1 H, CH_2), 2.75–2.84 (m, 1 H, CH_2), 3.57–3.63 (m, 1 H, CH), 3.79 (s, 3 H, OCH_3), 4.17–4.26 (m, 1 H, OCH_2), 4.42–4.48 (m, 1 H, OCH_2), 6.86 (d, J = 8.7 Hz, 2 H, Ar), 7.15 (d, J = 8.7 Hz, 2 H, Ar), 7.22–7.28 (m, 5 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 14.5, 21.0, 28.6, 29.6, 51.3, 55.2, 68.8, 113.3, 113.8, 122.0, 127.8, 128.3, 128.9, 130.0, 131.0, 134.2, 156.4, 159.7, 169.4 ppm. MS (ESI): *m/z* (%) = 365 (100) [$(M+1)^+$]. $C_{23}H_{24}O_4$ (364.4343): calcd. C 75.80, H 6.64%; found C 75.56, H 6.62%.

Ethyl 2-Oxo-9-phenyl-4-propyl-1,6-dioxaspiro[4.4]non-3-ene-3-carboxylate (6h): This compound was obtained as a white solid, yield 49 mg, 74%, m.p. 90–92 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1027, 1050, 1066, 1107, 1140, 1213, 1275, 1317, 1353, 1374, 1455, 1655, 1721, 1782, 2875, 2904, 2968 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 1.05 (t, J = 7.5 Hz, 3 H, CH_3), 1.29 (t, J = 7.2 Hz, 3 H, CH_3), 1.67–1.78 (m, 2 H, CH_2), 2.44–2.53 (m, 2 H, CH_2), 2.69–2.77 (m, 1 H, CH_2), 2.90–3.00 (m, 1 H, CH_2), 3.56–3.63 (m, 1 H, CH), 4.15–4.28 (m, 1 H, OCH_2), 4.24 (q, J = 7.2 Hz, 2 H, OCH_2), 4.39–4.45 (m, 1 H, OCH_2), 7.22–7.30 (m, 5 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 13.9, 14.6, 21.8, 29.2, 30.2, 51.5, 61.3, 69.3, 112.6, 122.8, 128.0, 128.5, 128.9, 133.5, 160.7, 164.9, 173.0 ppm. MS (ESI): *m/z* (%) = 331 (100) [$(M + H)^+$]. $C_{19}H_{22}O_5$ (330.1467): calcd. C 69.07, H 6.71%; found C 68.99, H 6.76%.

3-Methyl-9-phenyl-4-propyl-1,6-dioxaspiro[4.4]non-3-en-2-one (6i): This compound was obtained as a pale oil, yield 30 mg, 55%. IR (CH_2Cl_2): $\tilde{\nu}$ = 1010, 1034, 1058, 1103, 1269, 1295, 1320, 1455, 1682, 1763, 2873, 2930, 2963 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 1.01 (t, J = 7.2 Hz, 3 H, CH_3), 1.58–1.78 (m, 2 H, CH_2), 1.70 (s, 3 H, CH_3), 2.29–2.49 (m, 3 H, CH_2, CH_2), 2.46–2.79 (m, 1 H, CH_2), 3.46–3.53 (m, 1 H, CH_2), 4.11–4.35 (m, 1 H, OCH_2), 4.35–4.41 (m, 1 H, OCH_2), 7.20–7.29 (m, 5 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 8.8, 14.4, 21.0, 28.1, 30.2, 50.9, 68.5, 113.7, 126.9, 127.7, 127.72, 128.3, 129.0, 134.5, 156.4, 171.1 ppm. MS (ESI): m/z (%) = 273 (100) [(M + H)$^+$]. HRMS (MALDI) calcd. $(C_{17}H_{20}O_3+Na)^+$: 295.1305; found: 295.1316.

9-Methyl-3-phenyl-4-propyl-1,6-dioxaspiro[4.4]non-3-en-2-one (6j): This compound was obtained as a white solid, yield 34 mg, 63%, m.p. 112–114 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1031, 1042, 1105, 1356, 1447, 1470, 1743, 2896, 2930, 2963 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.90 (t, J = 7.2 Hz, 3 H, CH_3), 1.04 (d, J = 6.6 Hz, 3 H, CH_3), 1.23–1.62 (m, 2 H, CH_2), 2.01–2.12 (m, 1 H, CH_2), 2.24–2.47 (m, 2 H, CH_2), 2.48–2.56 (m, 2 H, CH_2), 4.03–4.09 (m, 1 H, OCH_2), 4.11–4.30 (m, 1 H, OCH_2), 7.36–7.47 (m, 5 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 11.8, 14.5, 21.0, 28.2, 31.5, 40.1, 69.0, 114.5, 128.4, 128.7, 128.9, 129.9, 131.0, 158.4, 170.0 ppm. MS (ESI): m/z (%) = 273 (100) [(M + H)$^+$]. HRMS (MALDI) calcd. $(C_{17}H_{20}O_3+Na)^+$: 295.1305; found: 295.1318.

3,9-Diphenyl-1,6-dioxaspiro[4.4]non-3-en-2-one (6k): This compound was obtained as a white solid, yield 39 mg, 67%, m.p. 115–117 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1004, 1044, 1060, 1111, 1175, 1232, 1270, 1307, 1326, 1356, 1450, 1494, 1759, 2897, 2961, 3028, 3069, 3088 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.46–2.55 (m, 1 H, CH_2), 2.72–2.87 (m, 1 H, CH_2), 3.55–3.62 (m, 1 H, CH), 4.18–4.27 (m, 1 H, OCH_2), 4.42–4.49 (m, 1 H, OCH_2), 7.20–7.36 (m, 8 H, Ar), 7.65–7.69 (m, 2 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 29.9, 52.8, 68.8, 111.3, 127.4, 127.8, 128.3, 128.5, 128.8, 129.1, 129.7, 133.9, 135.6, 141.9, 168.4 ppm. MS (EI): m/z (%) = 292 (1) [M$^+$], 265 (1), 247 (1), 189 (1), 145 (1), 118 (100), 102 (17), 91 (14). HRMS (MALDI) calcd. $(C_{19}H_{16}O_3+Na)^+$: 315.0992; found: 315.0999.

3,4,9-Triphenyl-1,6-dioxaspiro[4.4]non-3-en-2-one (6l): This compound was obtained as a white solid, yield 53 mg, 72%, m.p. 125–127 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1001, 1060, 1107, 1166, 1264, 1294, 1316, 1351, 1445, 1489, 1763, 2887, 3059 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.35–2.44 (m, 1 H, CH_2), 2.77–2.85 (m, 1 H, CH_2), 3.31–3.38 (m, 1 H, CH), 4.22–4.30 (m, 1 H, OCH_2), 4.52–4.58 (m, 1 H, OCH_2), 7.09–7.31 (m, 10 H, Ar), 7.36–7.48 (m, 5 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$, TMS): δ = 29.6, 51.7, 69.0, 113.2, 127.8, 128.2, 128.3, 128.7, 128.89, 128.9, 128.91, 129.00, 129.2, 129.9, 130.5, 130.9, 134.1, 153.3, 168.8 ppm. MS (ESI): m/z (%) = 369 (100) (M+1)$^+$]. HRMS (MALDI) calcd. $(C_{25}H_{20}O_3+H)^+$: 369.1485; found: 369.1483.

4-(Diphenylacetyl)-3-phenyl-5,6-dihydropyran-2-one (3c): This compound was obtained as a red oil, yield 11 mg, 15%. IR (CH_2Cl_2): $\tilde{\nu}$ = 1086, 1153, 1204, 1258, 1319, 1398, 1452, 1493, 1723, 2922 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.44 (t, J = 6.3 Hz, 2 H, CH_2), 4.23 (t, J = 6.3 Hz, 2 H, CH_2), 4.49 (s, 1 H, CH), 6.77–6.80 (m, 4 H, Ar), 7.16–7.21 (m, 6 H, Ar), 7.32–7.35 (m, 2 H, Ar), 7.35–7.44 (m, 3 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 28.3, 62.9, 66.2, 127.6, 128.7, 128.9, 129.1, 129.6, 129.8, 130.2, 133.5, 136.0, 151.4, 164.0, 202.6 ppm. MS (EI): m/z (%) = 368 (10) [M$^+$], 353 (100), 338 (45), 322 (20), 294 (30), 279 (45), 167 (100). HRMS (EI) calcd. $(C_{25}H_{20}O_3)^+$: 368.1412; found: 368.1413.

9-Benzhydrylidene-1,9-dihydro-2*H*-3-oxafluoren-4-one (4c): This compound was obtained as a yellow solid, yield 17.3 mg, 25%, m.p. 220–222 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1177, 1409, 1443, 1716 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.14 (t, J = 6.3 Hz, 2 H, CH_2), 4.25 (t, J = 6.3 Hz, 2 H, CH_2), 6.63–6.66 (m, 1 H, Ar), 6.88–6.93 (m, 1 H, Ar), 7.20–7.50 (m, 11 H, Ar), 7.99 (d, J = 7.8 Hz, 1 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 25.6, 67.3, 121.4, 123.2, 125.4, 127.7, 128.2, 128.4, 128.7, 129.8, 129.9, 130.7, 131.0, 135.6, 136.6, 138.3, 141.2, 141.6, 149.9, 155.2, 163.6 ppm. MS (CI): m/z (%) = 351 (29) [M+1$^+$]. HRMS (EI) calcd. $(C_{25}H_{18}O_2)^+$: 350.1307; found: 350.1307.

5-Benzhydrylidene-4-(2-hydroxyethyl)-3-phenyl-5*H*-furan-2-one (5c): This compound was obtained as a yellow solid, yield 16 mg, 22%, m.p. 155–157 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1002, 1048, 1153, 1444, 1491, 1753, 3447 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.33 (t, J = 6.9 Hz, 2 H, CH_2), 3.22 (t, J = 6.9 Hz, 2 H, OCH_2), 7.18–7.42 (m, 15 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 30.0, 61.1, 128.0, 128.54, 128.57, 128.6 (2), 128.9, 129.1, 129.2, 129.7, 130.9, 131.0, 131.1, 137.5, 138.1, 145.4, 147.9, 168.7 ppm. MS (EI): m/z (%) = 368 (33) [M$^+$], 367 (100), 337 (39), 291 (11), 215 (24), 165 (95), 105 (49). HRMS (EI) calcd. $(C_{25}H_{20}O_3)^+$: 368.1412; found: 368.1425.

3-(4-Chlorophenyl)-4-diphenylacetyl-5,6-dihydropyran-2-one (3d): This compound was obtained as a pale yellow solid, yield 47 mg, 58%, m.p. 133–135 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1091, 1154, 1203, 1267, 1399, 1452, 1492, 1722 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.51 (t, J = 6.0 Hz, 2 H, CH_2), 4.31 (t, J = 6.0 Hz, 2 H, CH_2), 4.60 (s, 1 H, CH), 6.88–6.90 (m, 4 H, Ar), 7.25–7.34 (m, 8 H, Ar), 7.42 (d, J = 8.7 Hz, 2 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 28.3, 63.1, 66.2, 127.7, 128.2, 128.8, 128.9, 129.1, 130.0, 131.5, 131.6, 135.9, 151.9, 163.8, 202.7 ppm. MS (ESI): m/z (%) = 404 (1), 402 (1) [M$^+$], 356 (1), 235 (1), 167 (100), 152 (37), 63 (3). HRMS (EI) calcd. $(C_{25}H_{19}O_3Cl)^+$: 402.1023; found: 402.1033.

5-Benzhydrylidene-3-(4-chlorophenyl)-4-(2-hydroxyethyl)-5*H*-furan-2-one (5d): This compound was obtained as a pale yellow solid, yield 12.7 mg, 16%, m.p. 140–142 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1042, 1092, 1154, 1209, 1266, 1444, 1491, 1586, 1752, 3466 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.39 (t, J = 7.2 Hz, 2 H, CH_2), 3.30 (t, J = 7.2 Hz, 2 H, OCH_2), 7.26–7.47 (m, 14 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 29.9, 60.9, 104.7, 128.0, 128.2, 128.6, 128.7, 128.9, 129.2, 129.9, 130.6, 130.9, 131.0, 135.0, 137.3, 138.1, 145.2, 148.4, 168.5 ppm. MS (EI): m/z (%) = 404 (6) 402 (13) [M$^+$], 370 (27), 280 (6), 249 (7), 167 (73), 139 (15), 84 (100). HRMS (EI) calcd. $(C_{25}H_{19}O_3Cl)^+$: 402.1023, found: 402.1026.

4-(Diphenylacetyl)-3-(4-fluorophenyl)-5,6-dihydropyran-2-one (3e): This compound was obtained as a pale yellow oil, yield 29 mg, 38%. IR (CH_2Cl_2): $\tilde{\nu}$ = 1084, 1158, 1225, 1257, 1278, 1495, 1509, 1599, 1723, 3064 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.50 (t, J = 6.0 Hz, 2 H, CH_2), 4.31 (t, J = 6.0 Hz, 2 H, CH_2), 4.59 (s, 1 H, CH), 6.87–6.91 (m, 4 H, Ar), 7.13–7.27 (m, 8 H, Ar), 7.36–7.41 (m, 2 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 28.3, 63.1, 66.2, 115.6 ($J_{C\text{-}F}$ = 21.8 Hz), 127.7, 128.7 ($J_{C\text{-}F}$ = 6.9 Hz), 128.9, 129.3, 132.1, 132.2, 135.9, 151.7, 163.4 ($J_{C\text{-}F}$ = 249.0 Hz), 164.0, 202.6 ppm. MS (ESI): m/z (%) = 386 (1) [M$^+$], 340 (1), 218 (2), 182 (5), 167 (100), 152 (13), 146 (4), 105 (7). HRMS (EI) calcd. $(C_{25}H_{19}O_3F)^+$: 386.1318, found: 386.1326.

5-Benzhydrylidene-3-(4-fluorophenyl)-4-(2-hydroxyethyl)-5*H*-furan-2-one (5e): This compound was obtained as a pale yellow solid, yield 25 mg, 32%, m.p. 53–55 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1044, 1160, 1230, 1266, 1444, 1492, 1508, 1595, 1759, 2927 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.39 (t, J = 7.2 Hz, 2 H, CH_2), 3.30 (t, J = 6.6 Hz, 2 H, OCH_2), 7.09–7.15 (m, 2 H, Ar), 7.32–7.52 (m,

12 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 29.9, 60.9, 115.8 ($J_{C\text{-}F}$ = 21.8 Hz), 125.7 ($J_{C\text{-}F}$ = 3.5 Hz), 128.0, 128.6 ($J_{C\text{-}F}$ = 9.8 Hz), 128.9, 129.1, 130.1, 130.9, 131.0, 131.2, 131.3, 137.4, 138.1, 145.3, 148.0, 162.9 ($J_{C\text{-}F}$ = 248.4 Hz), 168.7 ppm. MS (EI): *m*/*z* (%) = 386 (100) [M$^+$], 355 (28), 309 (6), 249 (6), 233 (10), 165 (80), 133 (15). HRMS (EI) calcd. $(C_{25}H_{19}O_3F)^+$: 386.1318, found: 386.1355.

4-Diphenylacetyl-3-*p*-tolyl-5,6-dihydropyran-2-one (3f): This compound was obtained as a red oil, yield 32 mg, 42%. IR (CH_2Cl_2): $\tilde{\nu}$ = 1085, 1153, 1185, 1203, 1265, 1319, 1398, 1452, 1494, 1723, 2923 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.43 (s, 3 H, CH_3), 2.50 (t, *J* = 6.0 Hz, 2 H, CH_2), 4.28 (t, *J* = 6.0 Hz, 2 H, CH_2), 4.62 (s, 1 H, CH), 6.86–6.89 (m, 4 H, Ar), 7.23–7.31 (m, 10 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 21.4, 28.3, 62.7, 66.2, 127.6, 128.6, 128.7, 129.1, 129.6, 130.1, 130.5, 136.1, 139.8, 150.9, 164.3, 202.9 ppm. MS (EI): *m*/*z* (%) = 382 (1) [M$^+$], 336 (1), 262 (3), 215 (12), 187 (7), 167 (100), 128 (10). HRMS (EI) calcd. $(C_{26}H_{22}O_3)^+$: 382.1569, found: 382.1574.

9-Benzhydrylidene-7-methyl-1,9-dihydro-2*H*-3-oxafluoren-4-one (4f): This compound was obtained as a red solid, yield 13.3 mg, 18%, m.p. 240–242 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1171, 1405, 1716 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.10 (s, 3 H, CH_3), 2.12 (t, *J* = 6.0 Hz, 2 H, CH_2), 4.24 (t, *J* = 6.0 Hz, 2 H, CH_2), 6.41 (d, *J* = 0.6 Hz, 1 H, Ar), 7.04 (dd, *J* = 0.6 Hz, *J* = 7.5 Hz, 1 H, Ar), 7.20–7.26 (m, 2 H, Ar), 7.34–7.50 (m, 8 H, Ar), 7.85 (d, *J* = 8.1 Hz, 1 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 21.6, 25.6, 67.3, 121.0, 124.2, 128.2, 128.35, 128.4, 128.6, 129.67, 129.7, 130.7, 130.9, 135.0, 135.8, 135.9, 136.7, 141.2, 141.6, 149.2, 154.6, 163.7 ppm. MS (CI): *m*/*z* (%) = 364 (26) [M$^+$], 363 (100), 304 (23), 278 (27), 229 (11), 155 (27), 91 (74), 44 (75). HRMS (MALDI) calcd. $(C_{26}H_{20}O_2+H)^+$: 365.1536, found: 365.1526.

5-Benzhydrylidene-4-(2-hydroxyethyl)-3-*p*-tolyl-5*H*-furan-2-one (5f): This compound was obtained as a pale yellow solid, yield 22 mg, 29%, m.p. 127–129 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1042, 1152, 1207, 1266, 1444, 1491, 1511, 1600, 1752, 3469 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.37 (s, 3 H, CH_3), 2.40 (t, *J* = 7.2 Hz, 2 H, CH_2), 3.30 (t, *J* = 7.2 Hz, 2 H, OCH_2), 7.21–7.46 (m, 14 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 21.3, 30.0, 61.1, 126.8, 128.0, 128.2, 128.5, 129.0 (2), 129.04, 129.3, 130.8, 131.0, 131.1, 137.5, 138.2, 139.0, 145.5, 147.3, 168.8 ppm. MS (EI): *m*/*z* (%) = 382 (4) [M$^+$], 363 (3), 350 (4), 229 (2), 165 (16), 129 (7), 105 (14), 84 (100), 44 (69). HRMS (EI) calcd. $(C_{26}H_{22}O_3)^+$: 382.1569, found: 382.1578.

4-Isobutyryl-3-phenyl-5,6-dihydropyran-2-one (3g): This compound was obtained as a red oil, yield 6 mg, 12%. IR (CH_2Cl_2): $\tilde{\nu}$ = 1086, 1152, 1179, 1208, 1258, 1313, 1399, 1445, 1466, 1725, 1970 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.84 (d, *J* = 6.6 Hz, 6 H, CH_3), 2.15 (sept, *J* = 6.6 Hz, 1 H, CH_3), 2.78 (t, *J* = 6.0 Hz, 2 H, CH_2), 4.56 (t, *J* = 6.0 Hz, 2 H, OCH_2), 7.30–7.39 (m, 5 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 17.5, 27.3, 39.7, 66.2, 128.3, 129.1, 129.8, 133.2, 150.9, 164.1, 209.8 ppm. MS (CI): *m*/*z* (%) = 244 (100) [M$^+$], 229 (30), 201 (22), 183 (15), 173 (53), 128 (30). HRMS (EI) calcd. $(C_{15}H_{16}O_3)^+$: 244.1099, found: 244.1099.

9-Isopropylidene-1,9-dihydro-2*H*-3-oxafluoren-4-one (4g): This compound was obtained as a yellow solid, yield 17 mg, 37%, m.p. 194–196 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1045, 1058, 1089, 1157, 1177, 1314, 1415, 1464, 1713 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.28 (s, 3 H, CH_3), 2.45 (s, 3 H, CH_3), 2.95 (t, *J* = 6.3 Hz, 2 H, CH_2), 4.39 (t, *J* = 6.3 Hz, 2 H, OCH_2), 7.17–7.22 (m, 2 H, Ar), 7.63 (d, *J* = 7.2 Hz, 1 H, Ar), 7.97 (dd, *J* = 6.3 Hz, *J* = 1.2 Hz, 1 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 26.0, 26.5, 27.2, 66.6, 121.6, 123.7, 125.55, 125.6, 126.8, 134.6, 135.3, 137.7, 149.5, 153.6, 163.6 ppm. MS (CI): *m*/*z* (%) = 226 (100) [M$^+$], 196 (52), 181 (14), 167 (38), 152 (40), 83 (28), 44 (48). $C_{15}H_{14}O_2$ (226.2705): calcd. C 79.62, H 6.24; found C 79.40, H 6.27.

3-(4-Chlorophenyl)-4-isobutyryl-5,6-dihydropyran-2-one (3h): This compound was obtained as a red oil, yield 23.5 mg, 42%, m.p. 85–87 °C. IR (KBr): $\tilde{\nu}$ = 1007, 1058, 1074, 1088, 1154, 1402, 1682, 1721, 2969 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$, TMS): δ = 0.88 (d, *J* = 6.6 Hz, 6 H, CH_3), 2.15 (sept, *J* = 6.6 Hz, 1 H, CH_3), 2.78 (t, *J* = 6.0 Hz, 2 H, CH_2), 4.56 (t, *J* = 6.0 Hz, 2 H, OCH_2), 7.23–7.38 (m, 4 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 17.6, 27.4, 39.9, 66.2, 128.5, 128.7, 131.2, 131.6, 135.4, 151.5, 163.9, 209.5 ppm. MS (CI): *m*/*z* (%) = 280 (17), 278 (55) [M$^+$], 243 (70), 235 (75), 207 (100), 163 (30), 128 (98), 43 (38). $C_{15}H_{15}ClO_3$ (278.7305): calcd. C 64.64, H 5.42; found C 64.39, H 5.41.

9-Isopropylidene-7-methyl-1,9-dihydro-2*H*-3-oxafluoren-4-one (4i): This compound was obtained as a yellow solid, yield 14 mg, 29%, m.p. 200–202 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1049, 1082, 1167, 1313, 1411, 1465, 1713 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.37 (s, 3 H, CH_3), 2.43 (s, 3 H, CH_3), 3.06 (s, 3 H, CH_3), 3.04 (t, *J* = 6.0 Hz, 2 H, CH_2), 4.48 (t, *J* = 6.0 Hz, 2 H, OCH_2), 7.13 (d, *J* = 7.8 Hz, 1 H, Ar), 7.53 (s, 1 H, Ar), 7.91 (d, *J* = 7.5 Hz, 1 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 22.0, 25.9, 26.5, 27.2, 66.7, 121.3, 124.7, 125.7, 127.5, 134.7, 135.2 (2), 135.6, 148.7, 152.8, 163.9 ppm. MS (EI): *m*/*z* (%) = 240 (100) [M$^+$], 225 (8), 210 (50), 181 (23), 152 (18), 115 (5). HRMS (EI) calcd. $(C_{16}H_{16}O_2)^+$: 240.1150, found: 240.1142.

9-(1-Phenylethylidene)-1,9-dihydro-2*H*-3-oxafluoren-4-one (*E*-4j): This compound was obtained as a yellow solid, yield 24 mg, 42%, m.p. 138–140 °C. IR (CH_2Cl_2): $\tilde{\nu}$ = 1049, 1078, 1166, 1185, 1202, 1314, 1411, 1442, 1465, 1594, 1711, 1765, 3052 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$, TMS): δ = 1.94 (t, *J* = 6.0 Hz, 2 H, CH_2), 2.84 (s, 3 H, CH_3), 4.15 (t, *J* = 6.0 Hz, 2 H, OCH_2), 7.24–7.45 (m, 7 H, Ar), 7.85 (d, *J* = 7.2 Hz, 1 H, Ar), 8.06–8.08 (m, 1 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 25.7, 26.3, 67.0, 121.9, 123.9, 125.9, 126.9, 127.7, 128.0, 128.5, 128.7, 128.9, 135.4, 138.4, 143.5, 149.9, 153.8, 163.9 ppm. MS (CI): *m*/*z* (%) = 288 (11) [M$^+$], 270 (5), 227 (10), 215 (12), 131 (21), 105 (42), 81 (87), 43 (100). HRMS (EI) calcd. $(C_{20}H_{16}O_2)^+$: 288.1150, found: 288.1155.

9-(1-Phenylethylidene)-1,9-dihydro-2*H*-3-oxafluoren-4-one (*Z*-4j): This compound was obtained as a yellow solid, yield 12 mg, 21%, m.p. 118–120 °C. ^{1}H NMR (300 MHz, $CDCl_3$, TMS): δ = 2.53 (s, 3 H, CH_3), 3.14 (t, *J* = 6.3 Hz, 2 H, CH_2), 4.52 (t, *J* = 6.3 Hz, 2 H, OCH_2), 6.07 (d, *J* = 8.1 Hz, 1 H, Ar), 6.72–6.77 (m, 1 H, Ar), 7.07–7.12 (m, 1 H, Ar), 7.19–7.26 (m, 1 H, Ar), 7.39–7.43 (m, 1 H, Ar), 7.89 (d, *J* = 8.1 Hz, 1 H, Ar) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 25.8, 27.0, 66.9, 121.3, 123.3, 125.3, 127.1, 127.2, 127.6, 128.6, 129.1, 136.0, 136.3, 137.6, 143.3, 149.3, 153.4, 163.4 ppm.

Supporting Information (see also the footnote on the first page of this article): ^{1}H and ^{13}C NMR spectra for the reaction products and the X-ray crystal data for compounds **6b**, **4c**, and **5c**.

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[10] Y.-H. Yang, M. Shi, *Org. Lett.* **2006**, *8*, 1709–1712. The X-ray crystal structure of **6c** is reported in this reference.

[11] The crystal data of **6b** has been deposited in CCDC with number 287397. Empirical formula: $C_{19}H_{16}O_3$; formula weight: 292.32; crystal size: 0.483 × 0.275 × 0.228; crystal color, habit: colorless, prismatic; crystal system: monoclinic; lattice type: primitive; lattice parameters: a = 28.780(3) Å, b = 6.3521(8) Å, c = 16.3252(19) Å, α = 90°, β = 96.739(2)°, γ = 90°, V = 2963.8(6) Å^3; space group: $C2/c$; Z = 8; D_{calc} = 1.310 g/cm^3; $F(000)$ = 1232; $R1$ = 0.0458, $wR2$ = 0.1010. Diffractometer: Rigaku AFC7R. CCDC-287397 (for **6b**), -29384 (for **4c**) and -297742 (for **5c**) contain the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.

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[13] The crystal data of **4c** has been deposited in CCDC with number 293784. Empirical formula: $C_{25}H_{18}O_2$; formula weight: 350.39; crystal size: 0.506 × 0.275 × 0.260; crystal color, habit: colorless, prismatic; crystal system: monoclinic; lattice type: primitive; lattice parameters: a = 9.8111(12) Å, b = 18.704(2) Å, c = 9.8784(12) Å, α = 90°, β = 94.422(2)°, γ = 90°, V = 1807.4(4) Å^3; space group: $P21/c$; Z = 4; D_{calc} = 1.288 g/cm^3; $F(000)$ = 736; $R1$ = 0.0493, $wR2$ = 0.1204. Diffractometer: Rigaku AFC7R. The crystal data of **5c** has been deposited in CCDC with number 297742. Empirical formula: $C_{25}H_{20}O_3$; formula weight: 368.41; crystal size: 0.303 × 0.150 × 0.139; crystal color, habit: colorless, prismatic; crystal system: orthorhombic; lattice type: primitive; lattice parameters: a = 6.2042(6) Å, b = 13.2809(12) Å, c = 22.759(2) Å, α = 90°, β = 90°, γ = 90°, V = 1875.3(3) Å^3; space group: $P2(1)2(1)2(1)$; Z = 4; D_{calc} = 1.298 g/cm^3; $F(000)$ = 768; $R1$ = 0.0654, $wR2$ = 0.1746. Diffractometer: Rigaku AFC7R.

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