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Formation of Acridones by Ethylene Extrusion in the Reaction of Arynes with β-Lactams and Dihydroquinolinones

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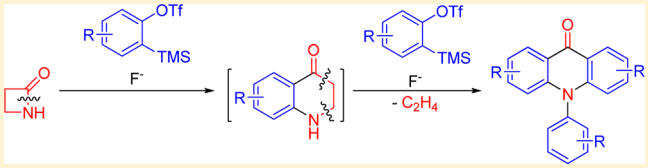
Supporting Information

ABSTRACT: *N*-Unsubstituted β-lactams react with a molecule of aryne by insertion into the amide bond to form a 2,3-dihydroquinolin-4-one, which subsequently reacts with another molecule of aryne to form an acridone by extrusion of a molecule of ethylene. 2,3-Dihydroquinolin-4-ones react under the same reaction conditions to afford identical results. This is the first example of ethylene extrusion in aryne chemistry.



INTRODUCTION

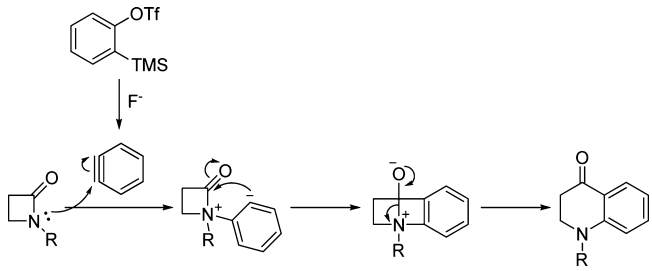
Benzyne is a highly reactive intermediate, which was first proposed by Wittig in 1942[1] and confirmed by Roberts in 1956.[2] Since the discovery of the Kobayashi aryne precursor,[3] various nucleophiles have been shown to react with arynes by nucleophilic addition to open the strained, triple bond of the aryne.[4] When the nucleophile is tethered to an electrophile, the nucleophilic addition can trigger subsequent electrophilic trapping of the aryl anion, leading to a formal σ-bond cleavage.[5] Amide functionality is one such tethered nucleophile−electrophile pair, where the nitrogen and the carbonyl serve as the nucleophile and the electrophile, respectively. Although amides typically undergo simple NH arylation,[4,6] amides with more electrophilic carbonyl groups, including trifluoroacetamides, trifluoromethanesulfinamides,[7] ureas,[8] and DMF,[9] have been shown to undergo carbonyl−nitrogen cleavage. Another class of substrates that could potentially exhibit such reactivity is a strained or twisted amide,[10] where the poor n−π conjugation makes the amide behave more like an independent amine and ketone. To the best of our knowledge, the reactivity of such amides toward arynes has received little attention.[11] We wish to report our initial results in this interesting area.

The substrates we have chosen to study are β-lactams. The angular strain of β-lactams renders poor conjugation of the nitrogen to the carbonyl. Thus, β-lactams have a stronger C=O double bond and a more basic nitrogen than regular amides.[12] We envisioned that the nitrogen atom of the β-lactam should exhibit greater nucleophilicity toward arynes than normal amides, thus leading to eventual C(O)−N bond cleavage to afford dihydroquinolinones (Scheme 1). While this outcome proved correct, we have observed some interesting subsequent chemistry, which we now report.

RESULTS AND DISCUSSION

Initial Discovery. We initiated our study using the *N*-unsubstituted β-lactam **1a** as the starting material (Scheme 2).

Scheme 1. Originally Anticipated Reaction of a β-Lactam with an Aryne



Stirring lactam **1a** with 1.0 equiv of the parent aryne precursor **2a** in the presence of 2.0 equiv of CsF as the fluoride source afforded three products in addition to unreacted **1a**: the simple *N*-arylation product **1b**, product **3a′** resulting from C(O)−N bond insertion and subsequent *N*-arylation, and acridone **4a**. Much to our surprise, not only was the originally anticipated product **3a** not observed, but also dihydroquinolinone **3a′** was identified as only a minor product by GC−MS analysis. The major product of this reaction was acridone **4a**. It thus appeared that the initial insertion products **3a** and/or **3a′** were also reactive toward arynes, if not even more so than lactam **1a**, and thus served merely as intermediates, eventually leading to acridone **4a**. For this to happen, however, the *C*2−*C*3 unit of the dihydroquinolinone **3a** and/or **3a′** must have been lost as a molecule of ethylene during the course of the reaction. It is very rare that aryne reactions lead to the extrusion of a neutral molecule.[13] To the best of our knowledge, this is the first example of ethylene extrusion in aryne chemistry.

To test our hypothesis that 2,3-dihydroquinolin-4-ones **3a**/**3a′** are reactive with arynes, pure **3a** from a commercial source

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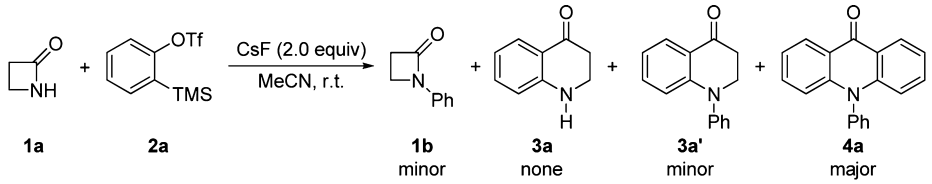
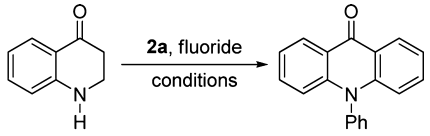
Scheme 2. Initial Results Leading to an Acridone from a β-Lactam



was subjected to our standard aryne reaction conditions. We thus found that as long as sufficient aryne was present, acridone **4a** was indeed formed under quite mild conditions, regardless of the fluoride source or the solvent used (Table 1). Thus, the intermediacy of dihydroquinolinone **3a** during the generation of acridone **4a** from β-lactam **1a** is confirmed.

Table 1. Formation of an Acridone from a 2,3-Dihydroquinolin-4(1*H*)-one[a]

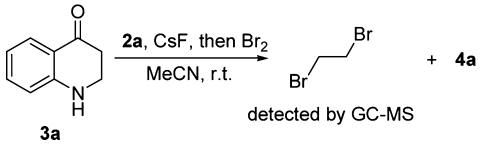


entry	equiv of **2a**	fluoride source (equiv)	conditions	yield[b] (%)
1	2	CsF (4)	MeCN, rt, 1 d	65
2	2.4	CsF (4.8)	MeCN, rt, 1 d	77
3	2.4	CsF (4.8)	THF, 65 °C, 1 d	72
4	2.4	TBAF (4.8)	THF, rt, 1 d	59
5	2.4	TBAT (4.8)	toluene, rt, 1 d	50

[a]All reactions were carried out on a 0.25 mmol scale in 4 mL of solvent. [b]Isolated yield of acridone **4a**.

To gain further evidence for the mechanism, an experiment was carried out to trap the extruded ethylene (the *C*2−*C*3 unit of the 2,3-dihydroquinolin-4-one). Thus, the reaction was repeated in a sealed vessel, and bromine was injected into the vessel after 10 h. GC−MS analysis of the crude reaction mixture revealed the presence of a large quantity of 1,2-dibromoethane, which supports the generation of ethylene in this reaction (Scheme 3).

Scheme 3. Ethylene Trapping



Scope of the β-Lactams. Encouraged by these findings, we first studied the scope of the reaction between various β-lactams and arynes. Although we have suggested that the reaction proceeds through the intermediacy of a dihydroquinolinone (such as **3a** and/or its *N*-arylated product **3a′**), all attempts to isolate such an intermediate have thus far been unsuccessful, even when using a 1:1 stoichiometry of the β-lactam and the aryne precursor. The best yields of acridone **4** have been achieved by employing 3.5 equiv of the aryne precursor (Figure 1) for the *N*-unsubstituted β-lactam **1a** or 2.4 equiv for the *N*-substituted β-lactams **1b** and **1c**. As seen in Table 2, compounds **4b** and **4c** can be obtained in reasonable yields from the symmetrical aryne precursors **2b** and **2c** (entries 2 and 3), respectively. The aryne derived from **2e** is known to be attacked preferentially by nucleophiles at the *meta* position (with respect to the OMe group) for both electronic and steric reasons.[4,14] In our studies, compound **4d** was formed in a surprisingly high yield as a single regioisomer (entry 4). *N*-Substituted β-lactams have also been examined in this reaction (entries 5−7). However, the *N*-phenyl lactam **1b** proved unreactive under our standard reaction conditions, and the *N*-allyl lactam **1c** was only marginally reactive, affording no more than a trace of the desired product **4e**. The *N*-benzyl lactam **1d** was slightly more reactive, affording compound **4f** in a 13% isolated yield (entry 7). It is worth pointing out that the yields of these three reactions did not improve very much even when the reactions were performed at a higher temperature. We also examined one α,α-disubstituted β-lactam **1e** (entry 8). In this case, the anticipated chemistry would require the extrusion of an olefin much larger than ethylene. Gratifyingly, we were able to identify the desired product **4a** in a 30% yield, indicating that extrusion of a molecule as large as 4-methylenehepta-1,6-diene is possible.

Scope of the Dihydroquinolinones. Because of the limited availability of β-lactams and the fact that incorporation of three molecules of aryne results in limited variability in the substitution pattern of the acridone product, we felt that the use of 2,3-dihydroquinolin-4-ones (series **3**) as the starting material would be more synthetically useful. Thus, we next focused our efforts on studying the reaction of dihydroquinolinones **3** with arynes.

We first examined *N*-unsubstituted substrates (Table 3). As shown previously, we have had preliminary success in the reaction of **3a** with **2a** (cf. entry 2, Table 1). Expanding the scope of the dihydroquinolinones **3** revealed that alkyl, ether, and chloride substituents are well tolerated, affording the corresponding acridones in good to excellent yields (entries 1−3). However, 6-fluorodihydroquinolinone (**3e**) proved unreactive (entry 4), presumably because of the electron-withdrawing nature of the fluoride. Different aryne precursors

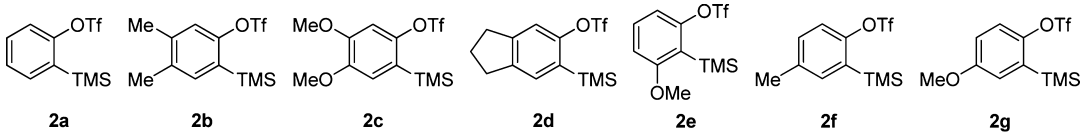


Figure 1. Aryne precursors.

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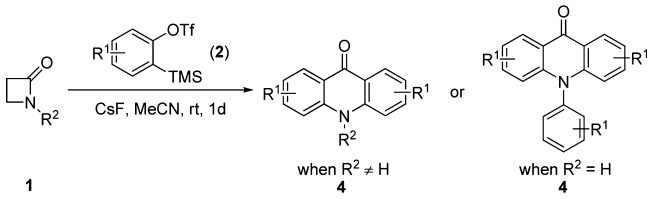
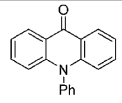
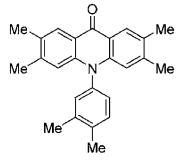
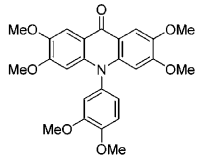
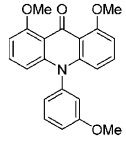
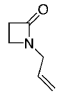
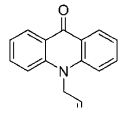
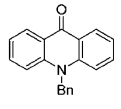
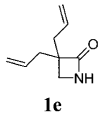
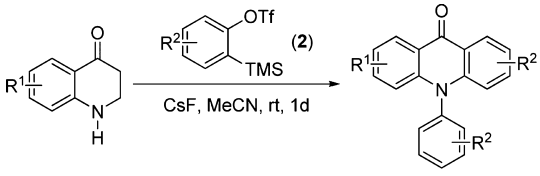
Table 2. Scope of the β-Lactams[a]

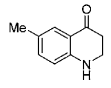
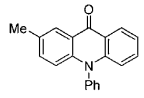
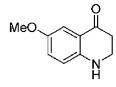
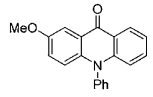
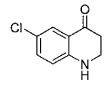
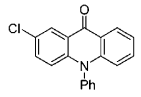
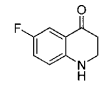
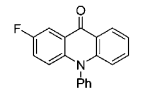
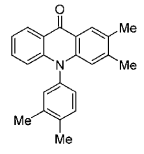
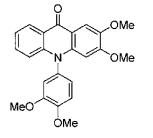
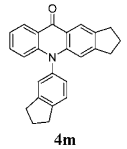
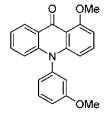


entry	β-lactam	aryne precursor	product	yield[b] (%)
1	 **1a**	**2a**	 **4a**	50
2	**1a**	**2b**	 **4b**	45
3	**1a**	**2c**	 **4c**	40
4	**1a**	**2e**	 **4d**	83
5	 **1b**	**2a**	**4a**	nr[c,d]
6		**2a**		trace[d,e]
7	 **1d**	**2a**	 **4f**	13[d]
8	 **1e**	**2a**	**4a**	30

[a]All reactions were carried out on a 0.25 mmol scale in 4 mL of MeCN with 3.5 equiv of the aryne precursor and 7 equiv of CsF. [b]Isolated yield. [c]All lactam starting material was recovered. [d]2.4 equiv of **2a** and 4.8 equiv of CsF were employed. [e]Detected by GC–MS.

Table 3. Scope of the *N*-Unsubstituted 2,3-Dihydroquinolin-4-ones[a]



entry	dihydroquinolinone	aryne precursor	product	yield[b] (%)
1	 **3b**	**2a**	 **4g**	88
2	 **3c**	**2a**	 **4h**	75
3	 **3d**	**2a**	 **4i**	80
4	 **3e**	**2a**	 **4j**	trace[c]
5	**3a**	**2b**	 **4k**	73
6	**3a**	**2c**	 **4l**	85
7	**3a**	**2d**	 **4m**	69
8	**3a**	**2e**	 **4n**	90

[a]All reactions were carried out on a 0.25 mmol scale with 2.4 equiv of **2** and 4.8 equiv of CsF in 4 mL of MeCN. [b]Isolated yield. [c]Detected by GC–MS.

(cf. Figure 1) have also been shown to react smoothly (entries 5–8), and compound **4n** has been obtained as a single regioisomer in a 90% yield (entry 8).

N-Substituted 2,3-dihydroquinolin-4-ones were next examined (Table 4). Compared with the results in Table 3, the yields using *N*-substituted 2,3-dihydroquinolin-4-ones are noticeably lower. Thus, the *N*-methyl dihydroquinolinone **3f** reacted with 1.2 equiv of benzyne precursor **2a** to afford acridone **4o** in a 63% yield (entry 1), a 14% drop from the corresponding *N*-unsubstituted precursor **3a** (cf. entry 2, Table 1). Similarly, substrates **3g–3i** were all smoothly transformed into the corresponding acridones **4p–4r** in moderate yields (entries 2–

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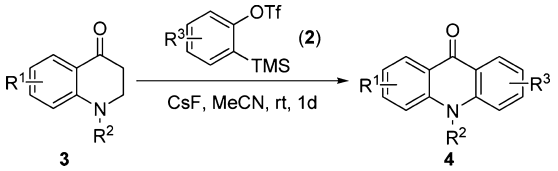
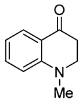
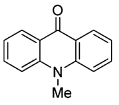
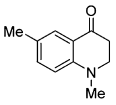
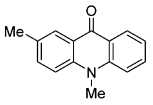
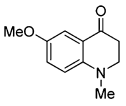
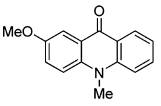
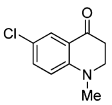
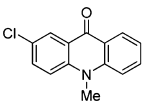
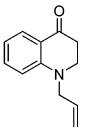
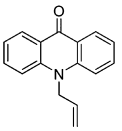
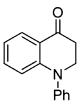
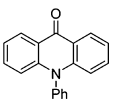
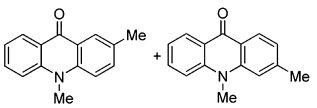
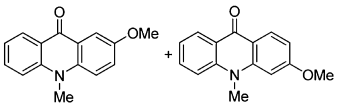
Table 4. Scope of the *N*-Substituted 2,3-Dihydroquinolin-4-ones[a]



3 **4**

entry	dihydroquinolinone	aryne precursor	product	yield[b] (%)
1	 **3f**	**2a**	 **4o**	63
2	 **3g**	**2a**	 **4p**	62
3	 **3h**	**2a**	 **4q**	55
4	 **3i**	**2a**	 **4r**	50
5	 **3j**	**2a**	 **4e**	48
6	 **3a'**	**2a**	 **4a**	trace[c]
7	**3f**	**2f**	 **4s + 4s'**	57[d] (1:1)
8	**3f**	**2g**	 **4t + 4t'**	60[d] (1:2)

[a]All reactions were carried out on a 0.25 mmol scale with 1.2 equiv of **2** and 2.4 equiv of CsF in 4 mL of MeCN.. [b]Isolated yield. [c]Detected by GC–MS. [d]Inseparable mixtures of regioisomers. The ratios were obtained by ^{1}H NMR spectroscopy. No attempts were made to identify the major isomer.

4). Other than a methyl group on the nitrogen, an allyl group can also be tolerated as seen in the reaction of substrate **3j**, which afforded acridone **4e** in a 48% yield (entry 5). However, placing a phenyl group on the nitrogen resulted in much lowered reactivity, as dihydroquinolinone **3a'** afforded only a trace of acridone **4a**, as detected by GC–MS (entry 6), indicating that the nucleophilicity and/or steric hindrance of the nitrogen is crucial to the reaction. This chemistry has also

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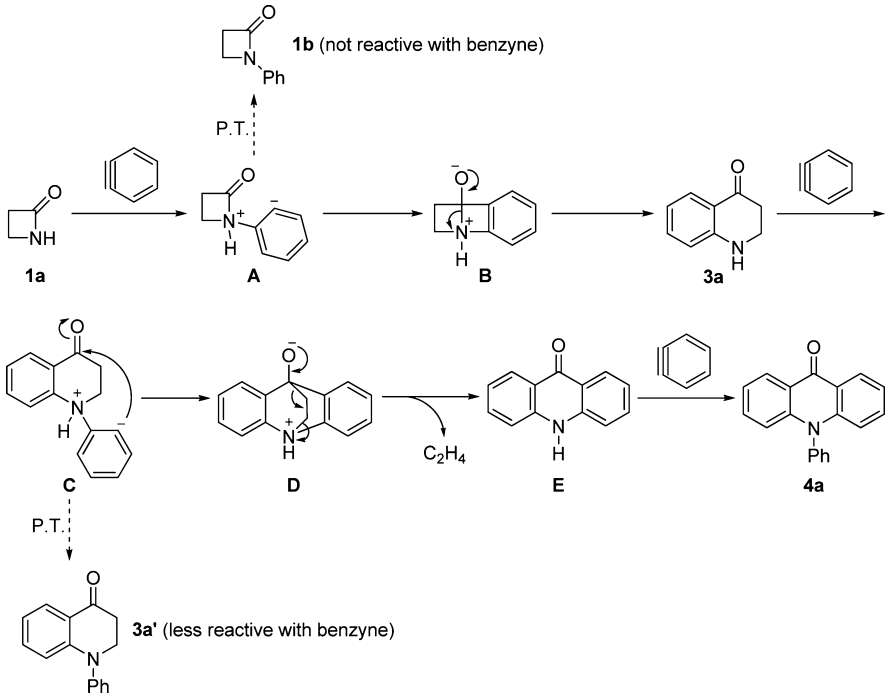
Scheme 4. Mechanistic Pathway

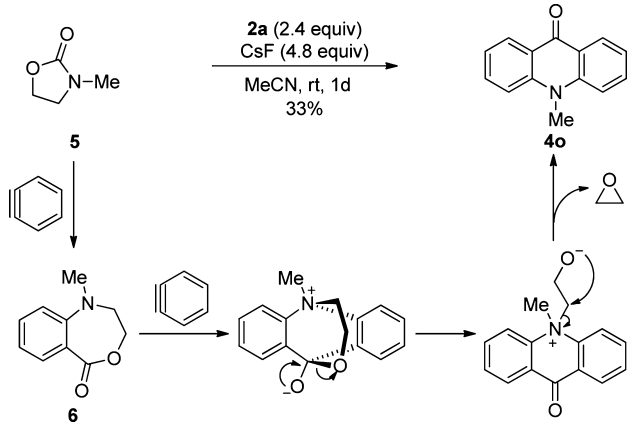


been extended to substituted aryne precursors. Thus, silyl triflates **2f** and **2g** reacted with dihydroquinolinone **3f** to afford the desired products in 57 and 60% yields, respectively (entries 7 and 8). Not surprisingly, since these two precursors are neither electronically nor sterically biased, mixtures of two regioisomers were obtained.

Mechanistic Discussion. On the basis of the above results, we propose the following mechanistic picture for this overall process (Scheme 4). First, the nitrogen atom of the *β*-lactam (**1a**) reacts with one molecule of the aryne to form intermediate **A**. Although **A** could undergo a simple proton transfer to afford lactam **1b** (as shown in Scheme 2), this is apparently a minor route and a nonproductive one with respect to the formation of acridone **4a**, since lactam **1b** does not readily react with arynes (see entry 5, Table 2). Thus, the aryl anion of intermediate **A** apparently nucleophilically adds to the carbonyl, and the resulting highly strained intermediate **B** collapses to furnish dihydroquinolinone **3a** with release of the ring strain. Compound **3a** presumably then reacts with a second molecule of aryne to afford intermediate **C**. Once again proton transfer from **C** to form dihydroquinolinone **3a′** is apparently a minor and nonproductive route (see entry 6, Table 4). In a fashion similar to the conversion of **A** to **B**, intermediate **C** most likely cyclizes to **D**, and subsequent extrusion of ethylene either by the arrow-pushing sequence described in Scheme 4 or by a retro-Diels–Alder process leads to the acridone **E**. Finally, acridone **E** undergoes NH arylation by a third molecule of aryne to afford the final major product acridone **4a**. In other words, the formation of acridone **4a** from lactam **1a** proceeds through the intermediacy of compounds **3a** and **E**, and compounds **1b** and **3a′** are much less important intermediates en route to acridone **4a**.

Possibilities for the Extrusion of Small Molecules Other than Ethylene. Inspired by the finding that dihydroquinolinone **3a** apparently reacts with an aryne to afford structures like intermediate **D** (Scheme 4), we were prompted to investigate other substrates that might afford a similar intermediate. It has been shown that a 5-membered ring urea can undergo C(O)–N cleavage upon reaction with arynes to afford products similar to compound **3**.[8] Thus, we examined the reaction of the cyclic carbamate 3-methyloxazolidin-2-one (**5**) with benzyne generated from **2a**. Gratifyingly, we were able to isolate acridone **4o** in a 33% yield (Scheme 5). This reaction

Scheme 5. Extrusion of Ethylene Oxide in the Reaction of Benzyne with 3-Methyl-2-oxazolidinone



is quite interesting on its own, because mechanistically, the first C(O)–N cleavage presumably results in a seven-membered ring intermediate **6**, whose subsequent reaction with benzyne must apparently extrude a molecule of ethylene oxide. Again, to the best of our knowledge, such a process is unprecedented in aryne chemistry.

■ CONCLUSIONS

In summary, we have demonstrated that the reaction of *β*-lactams or 2,3-dihydroquinolin-4-ones with arynes could afford respectable yields of acridones through the extrusion of

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ethylene. This chemistry speaks for the tendency of intermediates like **A** and **C** to readily undergo intramolecular nucleophilic cyclization rather than the seemingly easier proton transfer process. Further study has suggested that the extrusion of molecules larger than ethylene, such as 4-methylenehepta-1,6-diene and an epoxide, are also possible in aryne processes.

EXPERIMENTAL SECTION

General Information. The solvent THF was distilled over Na/benzophenone, and dichloromethane was distilled over CaH_2. Anhydrous MeCN, DMF, and DCE were used as received. The aryne precursors were used as received. Silica gel for column chromatography was supplied as 230−400 mesh from a commercial source. Powdered CsF and TBAF (1 M in THF solution) were used as received and stored in a desiccator.

All melting points are uncorrected. The ^{1}H and ^{13}C NMR spectra were recorded and are referenced to the residual solvent signals (7.26 ppm for ^{1}H and 77.2 ppm for ^{13}C in $CDCl_3$, 2.05 ppm for ^{1}H and 30.19 ppm for ^{13}C in acetone-d_6). A QTOF analyzer was used for all of the HRMS measurements.

***β*-Lactams.** Compound **1a** was commercially available and was used as received. The rest were prepared as follows.

1-Phenylazetidin-2-one (1b).[15] To a suspension of 1.8 mL (20 mmol) of aniline and 3.3 g (24 mmol) of K_2CO_3 in 20 mL of DCM at 0 °C was added dropwise 2.5 mL (24 mmol) of 3-bromopropanoyl chloride. The mixture was stirred at 0 °C for minutes and allowed to warm up to room temperature for another 3 h. The reaction was quenched with water and extracted with EtOAc three times. The combined organic layers were evaporated, and the residue was recrystallized in a hot solution of 1:1 petroleum ether/EtOAc to afford 3.42 g (ca. 15 mmol, ∼75% as is) of 3-bromo-*N*-phenylpropanamide as white crystals. This solid was then dissolved in DMF and cooled to 0 °C. To this solution was added 1.57 g (16.5 mmol) of sodium *tert*-butoxide in one portion, and the mixture was allowed to warm up to room temperature gradually. The reaction was quenched with water after 3 h and extracted with EtOAc. The combined organic layers were evaporated, and the residue was recrystallized from a hot solution of 1:1 petroleum ether/EtOAc to afford 1.76 g (60% overall yield) of 1-phenylazetidin-2-one as a red solid: mp 78−80 °C (lit[16] 78−79 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.53−7.32 (m, 5 H), 3.72 (t, J = 6.4 Hz, 1 H), 3.63 (t, J = 4.8 Hz, 1 H), 3.12 (t, J = 4.8 Hz, 1 H), 2.95 (t, J = 6.4 Hz, 1 H).

1-Allylazetidin-2-one (1c). The above procedure was applied to 1.14 g (20 mmol) of allylamine and 2.5 mL (24 mmol) of 3-bromopropanoyl chloride, followed by 1.52 g (16 mmol) of sodium *tert*-butoxide to afford 1.22 g (55% overall yield) of lactam **1c** as a colorless oil: ^{1}H NMR (400 MHz, $CDCl_3$) δ 5.80−5.70 (m, 1 H), 5.22 (d, J = 5.2 Hz, 1 H), 5.19 (s, 1 H), 3.82 (d, J = 6.0 Hz, 2 H), 3.22 (t, J = 4.0 Hz, 2 H), 2.94 (t, J = 4.0 Hz, 2 H).

1-Benzylazetidin-2-one (1d). The above procedure was applied to 2.14 g (20 mmol) of benzylamine and 2.5 mL (24 mmol) of 3-bromopropanoyl chloride, followed by 1.57 g (16.5 mmol) of sodium *tert*-butoxide to afford 1.87 g (58% overall yield) of lactam **1d** as a colorless oil: ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.39−7.23 (m, 5 H), 4.38 (s, 2 H), 3.14 (t, J = 3.9 Hz, 2 H), 2.95 (t, J = 3.9 Hz, 2 H).

3,3-Diallylazetidin-2-one (1e).[17] A mixture of 0.71 g (10 mmol) of azetidin-2-one (**1a**), 2.25 g (15 mmol) of *tert*-butyldimethylsilyl chloride, and 2.08 mL (15 mmol) of triethylamine in 20 mL of DCM was stirred for 12 h at room temperature. The mixture was then washed with water, and the aqueous phase was extracted with EtOAc three times. The combined organic layers were washed with brine, dried over Na_2SO_4, filtered, and concentrated to afford a colorless oil consisting of *N*-*tert*-butyldimethylsilylazetidin-2-one and residual TBSCl. This mixture was dissolved in THF, cooled to −78 °C under an N_2 atmosphere, and charged with 6.67 mL (1.8 M THF solution) of LDA. After being stirred for 2 h at −78 °C, 1.04 mL (12 mmol) of allyl bromide was added, and the mixture was gradually warmed up to room temperature for another 12 h. The reaction was quenched with water and extracted with EtOAc. The combined organic layers were washed with brine, dried over Na_2SO_4, filtered, and concentrated. The residue was purified by column chromatography (5:1 petroleum ether/EtOAc) to afford 0.52 g (2.3 mmol) of 3-allyl-1-(*tert*-butyldimethylsilyl)azetidin-2-one. This intermediate was treated with the above allylation procedure again to afford 0.3 g (1.1 mmol) of 3,3-diallyl-1-(*tert*-butyldimethylsilyl)azetidin-2-one. This product was dissolved in 10 mL of methanol, and 0.334 g (2.2 mmol) of CsF was added. After being stirred for 2 h, the mixture was quenched with water and extracted with EtOAc. The combined organic layers were washed with brine, dried over Na_2SO_4, filtered, and concentrated to afford 0.15 g (1.0 mmol, 10% overall yield from **1a**) of 3,3-diallylazetidin-2-one as a red oil: ^{1}H NMR (400 MHz, $CDCl_3$) δ 6.32 (s, 1 H), 5.85−5.74 (m, 2 H), 5.12 (d, J = 6.4 Hz, 2 H), 5.08 (s, 2 H), 3.07 (s, 2 H), 2.39 (d of ABq, J_{AB} = 14.1 Hz, J_{AX} = 6.7 Hz, J_{BX} = 7.9 Hz, 2 H), 2.31 (d of ABq, J_{AB} = 14.1 Hz, J_{AX} = 6.7 Hz, J_{BX} = 7.9 Hz, 2 H).

***N*-Unsubstituted 2,3-Dihydroquinolin-4-ones.** Compounds **3a** and **3e** were commercially available and used as received. The remaining dihydroquinolinones were prepared as follows.

6-Methyl-2,3-dihydroquinolin-4(1*H*)-one (3b).[15,18] Following the procedure described above for the synthesis of compound **1b**, 2.14 g (20 mmol) of *p*-toluidine, 2.5 mL (24 mmol) of 3-bromopropanoyl chloride, and 1.57 g (16.5 mmol) of sodium *tert*-butoxide were employed to obtain 1.92 g of *N*-(*p*-tolyl)azetidin-2-one. To a solution of 1.61 g (10 mmol) of 1-(*p*-tolyl)azetidin-2-one in 20 mL of DCE at 0 °C was added 2 mL (22 mmol) of TfOH. The mixture was allowed to warm up to room temperature and stirred for 2 h. The reaction was quenched with aq $NaHCO_3$ and extracted by EtOAc three times. The combined organic layers were washed with brine, dried over Na_2SO_4, filtered, and concentrated. The residue was purified by column chromatography (2:1 petroleum ether/EtOAc) to afford 1.19 g (40% overall yield from *p*-toluidine) of dihydroquinolinone **3b** as a yellow solid: mp 80−82 °C (lit[16] 82−84 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.65 (s, 1 H), 7.13 (dd, J = 8.4, 1.6 Hz, 1 H), 6.59 (d, J = 8.4 Hz, 1 H), 4.26 (s, 1 H), 3.55 (td, J = 8.0, 1.6 Hz, 2 H), 2.68 (t, J = 6.8 Hz, 2 H), 2.24 (s, 3 H).

6-Methoxy-2,3-dihydroquinolin-4(1*H*)-one (3c). Following the procedure described above for the synthesis of compound **1b**, 2.46 g (20 mmol) of *p*-anisidine, 2.5 mL (24 mmol) of 3-bromopropanoyl chloride, and 1.62 g (17 mmol) of sodium *tert*-butoxide were employed to obtain 2.3 g of *N*-(4-methoxyphenyl)azetidin-2-one. Next, 1.77 g (10 mmol) of *N*-(4-methoxyphenyl)azetidin-2-one was treated with 2 mL (22 mmol) of TfOH as described above to yield 1.28 g (47% overall yield from 4-methoxyaniline) of dihydroquinolinone **3c** as a yellow solid: mp 110−112 °C (lit[16] 113−114 °C); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.32 (d, J = 2.8 Hz, 1 H), 6.98 (dd, J = 8.8, 2.8 Hz, 1 H), 6.64 (d, J = 8.8 Hz, 1 H), 4.17 (s, 1 H), 3.77 (s, 3 H), 3.54 (t, J = 6.0 Hz, 2 H), 2.69 (t, J = 6.0 Hz, 2 H).

6-Chloro-2,3-dihydroquinolin-4(1*H*)-one (3d). Following the procedure described above for the synthesis of compound **1b**, 2.55 g (20 mmol) of 4-chloroaniline, 2.5 mL (24 mmol) of 3-bromopropanoyl chloride, and 1.57 g (16.5 mmol) of sodium *tert*-butoxide were employed to obtain 2.18 g of *N*-(4-chlorophenyl)-azetidin-2-one. Next, 1.82 g (10 mmol) of *N*-(4-chlorophenyl)-azetidin-2-one was treated with 2 mL (22 mmol) of TfOH as described above to yield 1.12 g (37% overall yield from 4-chloroaniline) of dihydroquinolinone **3d** as a yellow solid: mp 123−125 °C (lit[16] 125−126 °C); ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.81 (d, J = 2.7 Hz, 1 H), 7.23 (dd, J = 8.4, 2.7 Hz, 1 H), 6.63 (d, J = 8.4 Hz, 1 H), 4.40 (s, 1 H), 3.58 (td, J = 7.8, 1.5 Hz, 2 H), 2.69 (t, J = 7.5 Hz, 2 H).

***N*-Substituted 2,3-Dihydroquinolin-4-ones.** Compound **3a′** was commercially available and used as received. The other *N*-substituted dihydroquinolinones were prepared as follows.

1-Methyl-2,3-dihydroquinolin-4(1*H*)-one (3f). To an oven-dried vial was added 0.367 g (2.5 mmol) of 2,3-dihydroquinolin-4(1*H*)-one (**3a**) and 5 mL of DMF, followed by 0.15 g (3.75 mmol, 60% dispersed in mineral oil) of NaH. The mixture was stirred under a N_2 atmosphere at room temperature for 2 h and charged with 0.31 mL (5 mmol) of MeI. The vial was then capped and heated in an 80 °C oil bath for 12 h. After being quenched with water, the mixture was

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extracted with EtOAc three times. The combined organic layers were washed with brine, dried over Na_2SO_4, filtered, and concentrated. The residue was purified by column chromatography (2:1 petroleum ether/EtOAc) to afford 0.173 g (1.07 mmol, 43% yield) of dihydroquinolinone **3f** as a yellow oil: ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.90 (dd, J = 8.0, 1.2 Hz, 1 H), 7.40 (td, J = 4.4, 1.6 Hz, 1 H), 6.77−6.70 (m, 2 H), 3.47 (t, J = 7.2 Hz, 2 H), 2.99 (s, 3 H), 2.74 (t, J = 7.2 Hz, 2 H).

1,6-Dimethyl-2,3-dihydroquinolin-4(1*H*)-one (3g). The above procedure used for the synthesis of dihydroquinolinone **3f** was applied to 0.402 g (2.5 mmol) of dihydroquinolinone **3b**, 0.15 g (3.75 mmol,) of NaH, followed by 0.31 mL (5 mmol) of MeI to afford 0.197 g (45% overall yield) of compound **3g** as a yellow oil: ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.72 (s, 1 H), 7.23 (dd, J = 8.0, 1.2 Hz, 1 H), 6.65 (d, J = 7.6 Hz, 1 H), 3.42 (t, J = 7.2 Hz, 2 H), 2.95 (s, 3 H), 2.72 (t, J = 7.2 Hz, 2 H), 2.25 (s, 3 H).

6-Methoxy-1-methyl-2,3-dihydroquinolin-4(1*H*)-one (3h). The above procedure used to synthesize compound **3f** was applied to 0.442 g (2.5 mmol) of dihydroquinolinone **3c**, 0.15 g (3.75 mmol) of NaH, followed by 0.31 mL (5 mmol) of MeI to afford 0.205 g (43% overall yield) of compound **3h** as a yellow oil: ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.41 (d, J = 2.4 Hz, 1 H), 7.07 (dd, J = 8.1, 2.4 Hz, 1 H), 6.70 (d, J = 8.1 Hz, 1 H), 3.79 (s, 3 H), 3.39 (t, J = 7.5 Hz, 2 H), 2.94 (s, 3 H), 2.73 (t, J = 7.5 Hz, 2 H).

6-Chloro-1-methyl-2,3-dihydroquinolin-4(1*H*)-one (3i). The above procedure used to synthesize compound **3f** was applied to 0.454 g (2.5 mmol) of dihydroquinolinone **3d**, 0.15 g (3.75 mmol) of NaH, followed by 0.31 mL (5 mmol) of MeI to afford 0.200 g (41% overall yield) of compound **3i** as a yellow oil: ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.83 (d, J = 2.7 Hz, 1 H), 7.31 (dd, J = 9.0, 2.7 Hz, 1 H), 6.65 (d, J = 9.0 Hz, 1 H), 3.46 (t, J = 7.2 Hz, 2 H), 2.97 (s, 3 H), 2.72 (t, J = 7.2 Hz, 2 H).

1-Allyl-2,3-dihydroquinolin-4(1*H*)-one (3j). The above procedure used to synthesize compound **3f** was applied to 0.367 g (2.5 mmol) of dihydroquinolinone **3a**, 0.15 g (3.75 mmol) of NaH, followed by 0.605 g (5 mmol) of allyl bromide to afford 0.182 g (39% overall yield) of compound **3i** as a yellow oil: ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.90 (dd, J = 8.0, 1.6 Hz, 1 H), 7.36 (td, J = 4.4, 1.6 Hz, 1 H), 6.73−6.70 (m, 2 H), 5.90−5.82 (m, 1 H), 5.28−5.20 (m, 2 H), 3.99 (d, J = 5.2 Hz, 2 H), 3.52 (t, J = 7.2 Hz, 2 H), 2.72 (t, J = 7.2 Hz, 2 H).

3-Methyloxazolidin-2-one (5). This compound was commercially available and used as received.

General Procedures for Aryne Reactions Affording Acridones. Compounds **4a**−**4d** were prepared according to the following procedure (representative procedure for β-lactams where 3.5 equiv of arynes were used): to an oven-dried vial were added 0.875 mmol of aryne precursor, 0.25 mmol of β-lactam, 4 mL of MeCN, and 0.266 g (1.75 mmol) of CsF, sequentially. A nitrogen atmosphere was not required, except that a balloon of nitrogen was attached to the reaction vial for the ventilation of ethylene. The reaction was allowed to stir for 24 h before being quenched with aq Na_2CO_3 and extracted with EtOAc. The combined organic layers were washed with brine, dried over Na_2SO_4, filtered, and concentrated. The residue was purified by column chromatography (petroleum ether/EtOAc) to afford the desired products.

10-Phenylacridin-9(10*H*)-one (4a). The representative procedure was employed to afford 33.9 mg (0.13 mmol, 50% yield) of **4a** as a yellow solid: mp 271−273 °C (lit[19] 276 °C); R_f = 0.38 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.58 (dd, J = 8.0, 1.2 Hz, 2 H), 7.71 (t, J = 8.0 Hz, 2 H), 7.65 (t, J = 8.0 Hz, 1 H), 7.49 (td, J = 8.0, 1.6 Hz, 2 H), 7.37 (d, J = 7.6 Hz, 2 H), 7.27 (t, J = 8.0 Hz, 2 H), 6.75 (d, J = 8.4 Hz, 2 H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 178.3, 143.3, 139.2, 133.4, 131.3, 130.2, 129.8, 127.5, 122.0, 121.7, 117.0; HRMS (APCI) calcd for $C_{19}H_{14}NO$ (M + H) 272.1070, found 272.1076.

10-(3,4-Dimethylphenyl)-2,3,6,7-tetramethylacridin-9(10*H*)-one (4b). The representative procedure was employed to afford 39.9 mg (0.11 mmol, 45% yield) of **4b** as a yellow solid: mp 306−308 °C; R_f = 0.37 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.31 (s, 2 H), 7.42 (d, J = 8.0 Hz, 1 H), 7.06−7.04 (m, 2 H), 6.53 (s, 2 H), 2.45 (s, 3 H), 2.37 (s, 3 H), 2.35 (s, 6 H), 2.23 (s, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.7, 143.2, 141.9, 140.0, 138.1, 137.0, 132.0, 130.8, 130.4, 127.3, 127.0, 120.2, 117.3, 21.0, 20.2, 19.9, 19.3; HRMS (ESI) calcd for $C_{25}H_{26}NO$ (M + H) 356.2009, found 356.2012.

10-(3,4-Dimethoxyphenyl)-2,3,6,7-tetramethoxyacridin-9-(10*H*)-one (4c). The representative procedure was employed to afford 45.1 mg (0.10 mmol, 40% yield) of **4c** as a brown solid: mp 256−257 °C; R_f = 0.25 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.92 (s, 2 H), 7.14 (d, J = 8.0 Hz, 1 H), 6.98 (dd, J = 8.0, 1.2 Hz, 1 H), 6.85 (d, J = 1.6 Hz, 1 H), 6.18 (s, 2 H), 4.04 (s, 3 H), 4.01 (s, 6 H), 3.87 (s, 3 H), 3.68 (s, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 175.2, 154.0, 151.0, 149.9, 145.7, 139.2, 132.0, 122.3, 115.4, 112.4, 112.3, 106.4, 98.4, 56.51, 56.49, 56.3, 56.1; HRMS (ESI) calcd for $C_{25}H_{26}NO_7$ (M + H) 452.1704, found 452.1708.

1,8-Dimethoxy-10-(3-methoxyphenyl)acridin-9(10*H*)-one (4d). The representative procedure was employed to afford 74.9 mg (0.21 mmol, 83% yield) of **4d** as a brown solid: mp 250−253 °C; R_f = 0.11 (pure EtOAc); ^{1}H NMR (400 MHz, acetone-d_6) δ 7.65 (t, J = 8.0 Hz, 1 H), 7.39 (t, J = 8.4 Hz, 2 H), 7.25 (d, J = 8.0 Hz, 1 H), 6.96−6.94 (m, 2 H), 6.79 (d, J = 8.4 Hz, 2 H), 6.27 (d, J = 8.4 Hz, 2 H), 3.96 (s, 6 H), 3.87 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 179.2, 163.1, 161.6, 145.7, 142.1, 134.1, 123.7, 122.8, 120.6, 116.4, 115.0, 110.0, 104.8, 57.1, 56.3; HRMS (ESI) calcd for $C_{22}H_{20}NO_4$ (M + H) 362.1387, found 362.1389.

Compounds **4f**−**4n** were prepared according to the following procedure (representative procedure for *N*-substituted β-lactams/*N*-unsubstituted 2,3-dihydroquinolin-4-ones where 2.4 equiv of arynes were used): the general procedure used above for the synthesis of compound **4a** was applied to 0.6 mmol of aryne precursor, 0.25 mmol of *N*-substituted β-lactam/*N*-unsubstituted 2,3-dihydroquinolin-4-one starting material, 4 mL of MeCN, and 0.182 g (1.2 mmol) of CsF to afford the desired products.

10-Benzylacridin-9(10*H*)-one (4f). The representative procedure was employed to afford 9.3 mg (0.03 mmol, 13% yield) of **4f** as a brown solid: mp 178−180 °C (lit[20] 176−179 °C); R_f = 0.37 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.60 (dd, J = 8.0, 1.2 Hz, 2 H), 7.64 (td, J = 8.0, 1.6 Hz, 2 H), 7.38−7.15 (m, 9 H), 5.61 (s, 2 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 178.5, 142.8, 135.7, 134.3, 129.5, 128.1, 125.9, 122.8, 121.9, 115.4, 109.5, 51.1; HRMS (ESI) calcd for $C_{20}H_{16}NO$ (M + H) 286.1226, found 286.1229.

2-Methyl-10-phenylacridin-9(10*H*)-one (4g). The representative procedure was employed to afford 62.7 mg (0.22 mmol, 88% yield) of **4g** as a yellow solid: mp 220−221 °C; R_f = 0.38 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.57 (d, J = 7.2 Hz, 1 H), 8.36 (s, 1 H), 7.71−7.63 (m, 3 H), 7.46 (t, J = 7.6 Hz, 1 H), 7.35−7.29 (m, 3 H), 7.23 (t, J = 7.6 Hz, 1 H), 6.73 (d, J = 8.8 Hz, 1 H), 6.66 (d, J = 8.4 Hz, 1 H), 2.44 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 178.1, 143.1, 141.4, 139.2, 134.9, 133.2, 131.3, 131.2, 130.2, 129.6, 127.4, 126.6, 121.82, 121.77, 121.4, 116.9, 116.8, 20.9; HRMS (ESI) calcd for $C_{20}H_{16}NO$ (M + H) 286.1226, found 286.1233.

2-Methoxy-10-phenylacridin-9(10*H*)-one (4h). The representative procedure was employed to afford 56.4 mg (0.19 mmol, 75% yield) of **4h** as a yellow solid: mp 158−159 °C; R_f = 0.24 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.58 (d, J = 7.2 Hz, 1 H), 7.96 (d, J = 1.2 Hz, 1 H), 7.69−7.63 (m, 3 H), 7.47 (t, J = 7.6 Hz, 1 H), 7.36 (d, J = 8.8 Hz, 2 H), 7.24 (d, J = 7.6 Hz, 1 H), 7.13 (dd, J = 7.6, 1.2 Hz, 1 H), 6.76 (d, J = 8.4 Hz, 1 H), 6.72 (d, J = 8.4 Hz, 1 H), 3.93 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.7, 154.8, 142.8, 139.2, 138.2, 133.1, 131.2, 130.2, 129.7, 127.3, 124.2, 122.5, 121.4, 121.2, 118.7, 116.8, 106.2, 56.0; HRMS (ESI) calcd for $C_{20}H_{16}NO_2$ (M + H) 302.1176, found 302.1182.

2-Chloro-10-phenylacridin-9(10*H*)-one (4i). The representative procedure was employed to afford 61.1 mg (0.20 mmol, 80% yield) of **4i** as a yellow solid: mp 228−230 °C (lit[21] 229−230 °C); R_f = 0.45 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.52 (d, J = 8.4 Hz, 1 H), 8.49 (d, J = 2.8 Hz, 1 H), 7.73−7.67 (m, 3 H), 7.51 (td, J = 8.0, 1.2 Hz, 1 H), 7.42−7.37 (m, 3 H), 7.26 (t, J = 7.6 Hz, 1 H), 6.76 (d, J = 8.4 Hz, 1 H), 6.71 (d, J = 8.4 Hz, 1 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.1, 143.2, 141.7, 138.8, 133.7, 133.5, 131.4,

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130.0, 127.6, 127.4, 126.5, 122.7, 122.1, 121.8, 118.8, 117.6, 117.1; HRMS (ESI) calcd for $C_{19}H_{13}ClNO$ (M + H) 306.0680, found 306.0688.

10-(3,4-Dimethylphenyl)-2,3-dimethylacridin-9(10*H*)-one (4k). The representative procedure was employed to afford 59.8 mg (0.18 mmol, 73% yield) of **4k** as a yellow solid: mp 245−247 °C; R_f = 0.37 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.56 (d, J = 8.0 Hz, 1 H), 8.31 (s, 1 H), 7.46−7.41 (m, 2 H), 7.21 (t, J = 8.0 Hz, 1 H), 7.08−7.05 (m, 2 H), 6.77 (d, J = 8.4 Hz, 1 H), 6.58 (s, 1 H), 2.43 (s, 3 H), 2.36 (s, 3 H), 2.35 (s, 3 H), 2.23 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.9, 143.7, 143.3, 142.0, 139.8, 138.2, 136.8, 132.8, 132.0, 130.8, 127.3, 127.2, 127.0, 121.9, 121.1, 120.0, 117.4, 117.3, 117.0, 20.9, 20.1, 19.9, 19.3; HRMS (ESI) calcd for $C_{23}H_{22}NO$ (M + H) 328.1696, found 328.1704.

10-(3,4-Dimethoxyphenyl)-2,3-dimethoxyacridin-9(10*H*)-one (4l). The representative procedure was employed to afford 83.2 mg (0.21 mmol, 85% yield) of **4l** as a yellow solid: mp 234−235 °C; R_f = 0.62 (pure EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.51 (dd, J = 8.0, 1.2 Hz, 1 H), 7.86 (s, 1 H), 7.43 (td, J = 8.0, 1.2 Hz, 1 H), 7.21 (t, J = 8.0 Hz, 1 H), 7.11 (d, J = 8.4 Hz, 1 H), 6.95 (dd, J = 8.4, 1.6 Hz, 1 H), 6.84 (d, J = 1.6 Hz, 1 H), 6.79 (d, J = 8.8 Hz, 1 H), 6.18 (s, 1 H), 4.00 (s, 3 H), 3.95 (s, 3 H), 3.86 (s, 3 H), 3.66 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.5, 154.5, 151.0, 149.8, 145.7, 143.0, 139.7, 132.5, 131.7, 127.0, 122.2, 121.4, 121.3, 116.8, 115.7, 112.5, 112.4, 106.4, 98.5, 56.4, 56.32, 56.28, 56.0; HRMS (ESI) calcd for $C_{23}H_{22}NO_5$ (M + H) 392.1492, found 392.1490.

5-(2,3-Dihydro-1*H*-inden-5-yl)-2,3-dihydro-1*H*-cyclopenta[*b*]acridin-10(5*H*)-one (4m). The representative procedure was employed to afford 60.6 mg (0.17 mmol, 69% yield) of **4m** as a yellow solid: mp 176−178 °C; R_f = 0.48 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.57 (d, J = 8.0 Hz, 1 H), 8.41 (s, 1 H), 7.51−7.43 (m, 2 H), 7.24−7.21 (m, 1 H), 7.15 (s, 1 H), 7.07 (d, J = 8.0 Hz, 1 H), 6.79 (d, J = 8.0 Hz, 1 H), 6.66 (s, 1 H), 3.10−2.98 (m, 6 H), 2.87 (t, J = 7.6 Hz, 2 H), 2.24 (t, J = 7.6 Hz, 2 H), 2.08 (t, J = 7.6 Hz, 2 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 178.2, 151.5, 147.6, 145.8, 143.4, 142.9, 138.5, 137.5, 132.8, 127.7, 127.3, 126.6, 125.9, 121.9, 121.8, 121.1, 121.0, 117.1, 112.4, 33.8, 33.2, 33.0, 32.0, 26.0, 25.8; HRMS (ESI) calcd for $C_{25}H_{22}NO$ (M + H) 352.1696, found 352.1705.

1-Methoxy-10-(3-methoxyphenyl)acridin-9(10*H*)-one (4n). The representative procedure was employed to afford 74.6 mg (0.22 mmol, 90% yield) of **4n** as a pale white solid: mp 221−222 °C; R_f = 0.52 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.52 (d, J = 7.6 Hz, 1 H), 7.56 (t, J = 8.0 Hz, 1 H), 7.41 (t, J = 7.6 Hz, 1 H), 7.32 (t, J = 8.4 Hz, 1 H), 7.20 (t, J = 7.2 Hz, 1 H), 7.13 (d, J = 8.0 Hz, 1 H), 6.91 (d, J = 7.2 Hz, 1 H), 6.84 (s, 1 H), 6.66 (t, J = 9.6 Hz, 2 H), 6.33 (d, J = 8.4 Hz, 1 H), 4.01 (s, 3 H), 3.82 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.9, 161.9, 161.6, 145.7, 142.2, 140.7, 133.4, 132.8, 131.8, 127.4, 123.6, 122.0, 121.7, 116.5, 115.5, 115.3, 112.7, 109.3, 102.9, 56.4, 55.7; HRMS (ESI) calcd for $C_{21}H_{18}NO_3$ (M + H) 332.1281, found 332.1285.

Compounds **4e**, **4o**−**4r**, and the inseparable mixtures of **4s** + **4s′** and **4t** + **4t′** were prepared according to the following procedure (representative procedure for *N*-substituted 2,3-dihydroquinolin-4-ones where 1.2 equiv of arynes were used): the general procedure used above for the synthesis of acridone **4a** was applied to 0.3 mmol of aryne precursor, 0.25 mmol of *N*-substituted 2,3-dihydroquinolin-4-one, 4 mL of MeCN, and 0.091 g (0.6 mmol) of CsF to afford the desired product.

10-Allylacridin-9(10*H*)-one (4e). The representative procedure was employed to afford 28.2 mg (0.12 mmol, 48% yield) of **4e** as a yellow solid: mp 131−132 °C (lit[22] 132−134 °C); R_f = 0.36 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.55 (d, J = 7.6 Hz, 2 H), 7.69 (t, J = 7.6 Hz, 2 H), 7.39 (d, J = 8.8 Hz, 2 H), 7.28 (t, J = 7.6 Hz, 2 H), 6.18−6.09 (m, 1 H), 5.31 (d, J = 10.8 Hz, 1 H), 5.10 (d, J = 17.2 Hz, 1 H), 4.96 (s, 2 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 178.4, 142.4, 134.1, 130.8, 127.6, 122.7, 121.4, 117.6, 115.2, 49.5; HRMS (ESI) calcd for $C_{16}H_{14}NO$ (M + H) 236.1070, found 236.1067.

10-Methylacridin-9(10*H*)-one (4o). The representative procedure was employed to afford 32.9 mg (0.16 mmol, 63% yield) of **4o** as a yellow solid: mp 201−203 °C (lit[23] 201−203 °C); R_f = 0.22 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.53 (d, J = 7.2 Hz, 2 H), 7.68 (t, J = 6.4 Hz, 2 H), 7.46 (d, J = 8.0 Hz, 2 H), 7.25 (t, J = 6.4 Hz, 2 H), 3.82 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 178.2, 142.7, 133.9, 127.9, 122.6, 121.4, 114.9, 33.8; HRMS (ESI) calcd for $C_{14}H_{12}NO$ (M + H) 210.0913, found 210.0918.

2,10-Dimethylacridin-9(10*H*)-one (4p). The representative procedure was employed to afford 34.6 mg (0.16 mmol, 62% yield) of **4p** as a yellow solid: mp 149−151 °C (lit[24] 153 °C); R_f = 0.25 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.53 (d, J = 8.0 Hz, 1 H), 8.31 (s, 1 H), 7.66 (t, J = 7.2 Hz, 1 H), 7.49−7.43 (m, 2 H), 7.36 (d, J = 8.8 Hz, 1 H), 7.22 (d, J = 7.6 Hz, 1 H), 3.80 (s, 3 H), 2.44 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 178.1, 142.6, 140.8, 135.3, 133.7, 131.0, 127.9, 127.2, 122.6, 122.5, 121.1, 114.9, 114.8, 33.7, 20.8; HRMS (ESI) calcd for $C_{15}H_{14}NO$ (M + H) 224.1070, found 224.1075.

2-Methoxy-10-methylacridin-9(10*H*)-one (4q). The representative procedure was employed to afford 32.9 mg (0.14 mmol, 55% yield) of **4q** as a yellow solid: mp 139−141 °C (lit[25] 138 °C); R_f = 0.12 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.54 (d, J = 8.0 Hz, 1 H), 7.92 (d, J = 2.8 Hz, 1 H), 7.66 (td, J = 7.6, 1.2 Hz, 1 H), 7.46−7.42 (m, 2 H), 7.31 (dd, J = 9.2, 3.2 Hz, 1 H), 7.23 (d, J = 7.6 Hz, 1 H), 3.92 (s, 3 H), 3.82 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.6, 154.5, 142.2, 137.5, 133.7, 127.9, 124.5, 123.3, 121.9, 121.0, 116.7, 114.8, 106.8, 55.9, 33.8; HRMS (ESI) calcd for $C_{15}H_{14}NO_2$ (M + H) 240.1019, found 240.1021.

2-Chloro-10-methylacridin-9(10*H*)-one (4r). The representative procedure was employed to afford 30.5 mg (0.13 mmol, 50% yield) of **4r** as a yellow solid: mp 171−173 °C; R_f = 0.21 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.44 (d, J = 7.6 Hz, 1 H), 8.39 (s, 1 H), 7.68 (t, J = 8.0 Hz, 1 H), 7.53 (dd, J = 9.2, 2.0 Hz, 1 H), 7.43 (d, J = 8.8 Hz, 1 H), 7.36 (d, J = 9.2 Hz, 1 H), 7.23 (d, J = 7.2 Hz, 1 H), 3.79 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.0, 142.4, 140.9, 134.2, 133.8, 127.8, 127.3, 126.8, 123.3, 122.4, 121.7, 116.7, 115.0, 34.0; HRMS (ESI) calcd for $C_{14}H_{11}ClNO$ (M + H) 244.0524, found 244.0523.

2,10-Dimethylacridin-9(10*H*)-one and 3,10-dimethylacridin-9(10*H*)-one (4s + 4s′). The representative procedure was employed to afford 31.8 mg (0.14 mmol, 57% total yield) of **4s** + **4s′** as a yellow solid: R_f = 0.25 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.54−8.51 (m, 2 H), 8.39 (d, J = 8.0 Hz, 1 H), 8.30 (s, 1 H), 7.65 (t, J = 7.6 Hz, 2 H), 7.47 (dd, J = 8.4, 2.0 Hz, 1 H), 7.43 (d, J = 8.8 Hz, 2 H), 7.35 (d, J = 8.8 Hz, 1 H), 7.22 (d, J = 10.0 Hz, 3 H), 7.05 (d, J = 8.0 Hz, 1 H), 3.79 (s, 3 H), 3 78 (s, 3 H), 2.48 (s, 3 H), 2.43 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 178.1, 177.9, 144.8, 142.8, 142.7, 142.6, 140.8, 135.3, 133.7, 131.0, 127.8, 127.4, 123.1, 123.0, 122.7, 122.6, 122.5, 121.2, 121.0, 120.7, 114.85, 114.82, 33.73, 33.71, 22.8, 20.8; HRMS (ESI) calcd for $C_{15}H_{14}NO$ (M + H) 224.1070, found 224.1072.

2-Methoxy-10-methylacridin-9(10*H*)-one and 3-methoxy-10-methylacridin-9(10*H*)-one (4t + 4t′). The representative procedure was employed to afford 35.9 mg (0.15 mmol, 60% total yield) of **4t** + **4t′** as a yellow solid: R_f = 0.18 (2:1 petroleum ether/EtOAc); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.53 (d, J = 8.0 Hz, 1 H), 8.49 (d, J = 7.6 Hz, 0.5 H), 8.43 (d, J = 8.8 Hz, 0.5 H), 7.91 (d, J = 2.8 Hz, 1 H), 7.67−7.61 (m, 1.5 H), 7.44−7.38 (m, 2.5 H), 7.30 (dd, J = 9.2, 3.2 Hz, 1 H), 7.25−7.21 (m, 1.5 H), 6.81 (dd, J = 8.8, 2.0 Hz, 0.5 H), 6.72 (s, 0.5 H), 3.91 (s, 3 H), 3.90 (s, 1.5 H), 3.81 (s, 3 H), 3.72 (s, 1.5 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.6, 177.2, 164.3, 154.5, 144.5, 142.8, 142.2, 137.5, 133.6, 133.4, 127.8, 124.5, 124.4, 123.2, 122.7, 121.8, 121.0, 117.2, 116.7, 114.7, 106.8, 98.0, 56.0, 55.7, 33.9, 33.8; HRMS (ESI) calcd for $C_{15}H_{14}NO_2$ (M + H) 240.1019, found 240.1020.

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■ ASSOCIATED CONTENT

Supporting Information

Full ^{1}H and ^{13}C NMR spectra. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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