

Supporting Information

Formation of cyclopenta[*c*]quinolines through visible-light-induced photoredox cascade bis-annulations of 1,7-enynes with sulfoxonium ylides

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1. General information

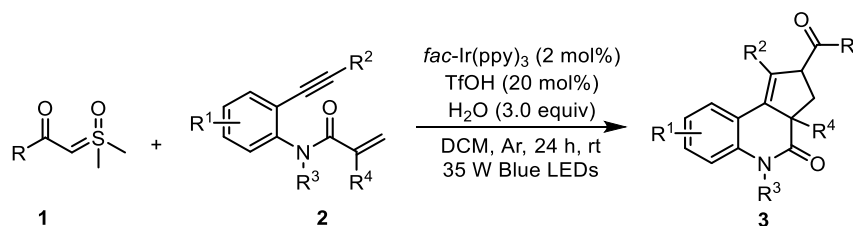
The reactions via general procedure were carried out under an atmosphere of argon unless otherwise noted. Column chromatography was performed using silica gel (200-300 mesh). ^1H , ^{13}C and ^{19}F NMR spectra were recorded on Bruker-AV (400, 100 and 376 MHz, respectively) instrument internally referenced to tetramethylsilane (TMS) or chloroform signals. Mass spectra were measured on Agilent 5977 GC-MS instrument (EI). High-resolution mass spectra (ESI) were obtained with the Thermo Scientific LTQ Orbitrap XL mass spectrometer. Melting points were measured with a YUHUA X-5 melting point instrument and were uncorrected. The structures of known compounds were further corroborated by comparing their ^1H NMR, ^{13}C NMR data and HRMS data with those in literature. A commercially available blue LED (35W, HIPAR30, luminous flux is not less than 3200 lm) was purchased from Shenzhen Jing Feng Times Lighting Technology Co., Ltd as the reaction light source. All irradiation reactions were carried out in borosilicate glass vessel. The distance from the light source to the irradiation vessel is around 4-5 cm. Unless otherwise noted, all photocatalysts and other reagents were obtained from commercial suppliers and used without further purification.

2. General procedure of the reactions

Reaction device diagram: Reaction set-up for irradiation of mixture with 35 W blue LEDs and the homemade insulation attached to the apparatus was used to maintain the reaction temperature at 25-35 °C (**Figure S1**).



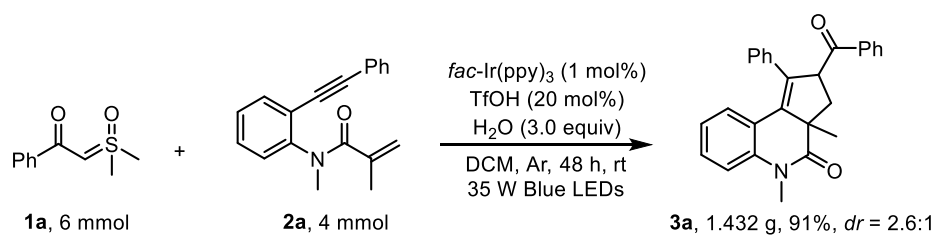
Figure S1. reaction setup (35 W blue LED)



General Procedure A: A 10 mL reaction vessel was charged with *fac*-Ir(ppy)₃ (2.6 mg, 0.004 mmol, 2 mol%), 1,7-enynes (**2**, 0.2 mmol, 1.0 equiv) and sulfoxonium ylides (**1**, 0.3 mmol, 1.5 equiv). The atmosphere was exchanged by applying vacuum and backfilling with Ar (this process was conducted for three times). Then, TfOH (20 mol%, 3.6 μL), H₂O (3.0 equiv, 10.8 μL) and DCM (dry, 2 mL) were added. The resulting mixture was stirred for 24 h under irradiation with a 35 W blue LEDs. After cooling to room temperature, the crude reaction mixture extracted with dichloromethane (3×10 mL). The extracts were combined, dried over sodium sulfate, and filtered, and the volatiles were removed under reduced pressure. Column chromatography was performed using silica gel (200-300 mesh) to give the pure products **3**.

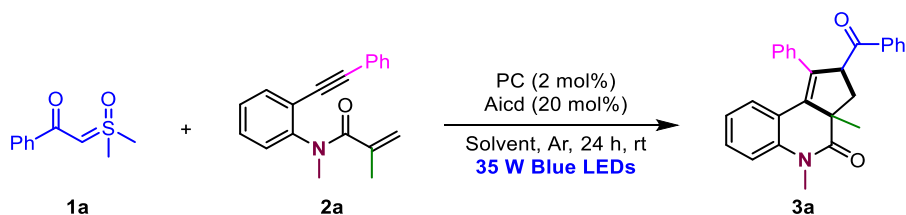
General Procedure B: A 10 mL reaction vessel was charged with *fac*-Ir(ppy)₃ (2.6 mg, 0.004 mmol, 2 mol%), 1,7-enynes (**2**, 0.2 mmol, 1.0 equiv) and sulfoxonium ylides (**1**, 0.6 mmol, 3.0 equiv). The atmosphere was exchanged by applying vacuum and backfilling with Ar (this process was conducted for three times). Then, TfOH (20 mol%, 3.6 μL), H₂O (3.0 equiv, 10.8 μL) and DCM (dry, 2 mL) were added. The resulting mixture was stirred for 24 h under irradiation with a 35 W blue LEDs. After cooling to room temperature, the crude reaction mixture extracted with dichloromethane (3×10 mL). The extracts were combined, dried over sodium sulfate, and filtered, and the volatiles were removed under reduced pressure. Column chromatography was performed using silica gel (200-300 mesh) to give the pure products **3**.

Scale-up experiment:

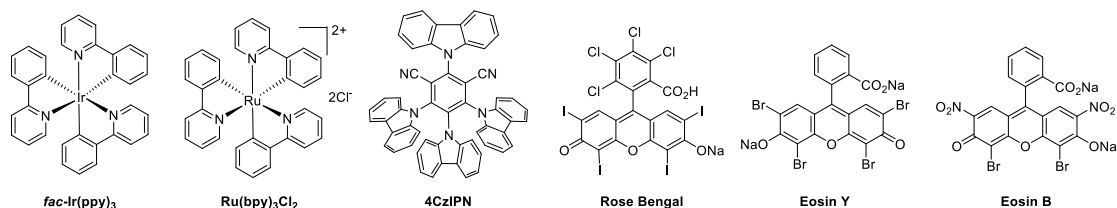


4 mmol scale reaction: A 100 mL reaction vessel was charged with *N*-methyl-*N*-(2-(phenylethynyl)phenyl)methacrylamide (1.101 g, 4 mmol), 2-(dimethyl(oxo)- λ^6 -sulfaneylidene)-1-phenylethan-1-one (1.177 g, 6 mmol) and *fac*-Ir(ppy)₃ (26.2 mg, 0.04 mmol, 1 mol%), followed by the addition of DCM (40 mL). The atmosphere was exchanged by applying vacuum and backfilling with Ar (this process was conducted for three times). Then, H₂O (3.0 equiv, 216 μ L) and TfOH (20 mol%, 72 μ L) were added. The resulting mixture was stirred for 48 hours under irradiation with a 35 W blue LEDs. The reaction was monitored by TLC. The mixture extracted with dichloromethane (3 \times 50 mL). The extracts were combined, dried over sodium sulfate, and filtered, and the volatiles were removed under reduced pressure. Column chromatography was performed using silica gel (petroleum ether/acetone = 8:1) to give product **3a** (1.432 g, 91% yield, *dr* = 2.6:1).

3. Optimization of reaction conditions^a



Entry	PC	Acid	Solvent	Yield (%) ^b
1	<i>fac</i> -Ir(ppy) ₃	-	DCM	71 (<i>dr</i> = 1.7:1)
2	<i>fac</i> -Ir(ppy) ₃	-	MeCN	44 (<i>dr</i> = 1:2.6)
3	<i>fac</i> -Ir(ppy) ₃	-	Toluene	26 (<i>dr</i> = 1.6:1)
4	<i>fac</i> -Ir(ppy) ₃	-	Acetone	62 (<i>dr</i> = 1.6:1)
5	<i>fac</i> -Ir(ppy) ₃	-	THF	<10
6	<i>fac</i> -Ir(ppy) ₃	-	DMF	25 (<i>dr</i> = 1.6:1)
7	<i>fac</i> -Ir(ppy) ₃	-	DCE	67 (<i>dr</i> = 1.7:1)
8	<i>fac</i> -Ir(ppy) ₃	-	DCM (dry)	trace
9	<i>fac</i> -Ir(ppy) ₃	-	DCM (dry) + H ₂ O (2 equiv)	57 (<i>dr</i> = 1.3:1)
10	<i>fac</i> -Ir(ppy) ₃	-	DCM (dry) + H ₂ O (3 equiv)	69 (<i>dr</i> = 1.4:1)
11	<i>fac</i> -Ir(ppy) ₃	-	DCM (dry) + H ₂ O (4 equiv)	65 (<i>dr</i> = 1.4:1)
12	<i>fac</i> -Ir(ppy) ₃	-	DCM (dry) + H ₂ O (5 equiv)	52 (<i>dr</i> = 1.3:1)
13	<i>fac</i> -Ir(ppy) ₃	-	DCM (dry) + H ₂ O (10 equiv)	50 (<i>dr</i> = 1.4:1)
14	<i>fac</i> -Ir(ppy) ₃	-	DCM: H ₂ O (1:9)	23 (<i>dr</i> = 1:10)
15	<i>fac</i> -Ir(ppy) ₃	-	DCM: H ₂ O (1:7)	<10
16	4CzIPN	-	DCM (dry) + H ₂ O (3 equiv)	61 (<i>dr</i> = 1.6:1)
17	Rose Bengal	-	DCM (dry) + H ₂ O (3 equiv)	trace
18	Eosin Y	-	DCM (dry) + H ₂ O (3 equiv)	ND
19	Eosin B	-	DCM (dry) + H ₂ O (3 equiv)	ND
20	Ru(bpy)₃Cl₂	-	DCM (dry) + H ₂ O (3 equiv)	trace
21	<i>fac</i> -Ir(ppy) ₃	TFA	DCM (dry) + H ₂ O (3 equiv)	70 (<i>dr</i> = 1.7:1)
22	<i>fac</i> -Ir(ppy) ₃	H ₂ SO ₄	DCM (dry) + H ₂ O (3 equiv)	86 (<i>dr</i> = 2.7:1)
23	<i>fac</i> -Ir(ppy) ₃	TfOH	DCM (dry) + H ₂ O (3 equiv)	93 (<i>dr</i> = 2.3:1)
24	-	TfOH	DCM (dry) + H ₂ O (3 equiv)	ND
25 ^c	<i>fac</i> -Ir(ppy) ₃	TfOH	DCM (dry) + H ₂ O (3 equiv)	ND

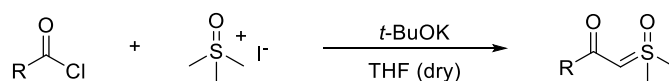


^a Reaction conditions: **1a** (0.3 mmol, 1.5 equiv), **2a** (0.2 mmol, 1.0 equiv) and photocatalyst (PC, 0.004, 2 mol %) in solvent (2 mL) at (25-30 °C) under Ar atmosphere and 35 W blue LEDs

irradiation for 24 h. ^b Yield and diastereoselectivity ratio (*cis/trans*) of isolated product were given.^c
No light.

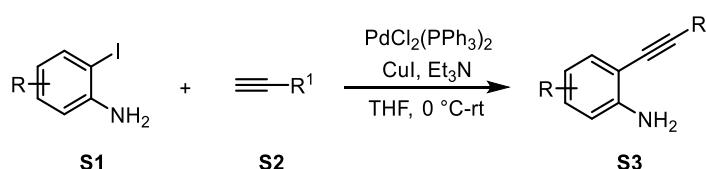
4. Procedures for the preparation of sulfoxonium ylides and 1,7-enynes

4.1 General Procedure for preparation of sulfoxonium ylides ^[1]:

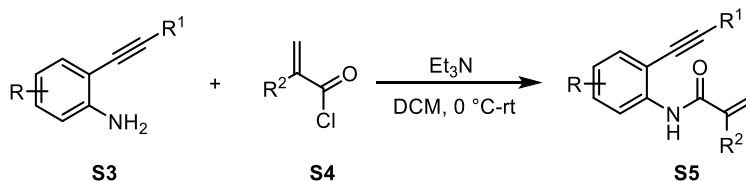


To a stirred solution of potassium tert-butoxide (3.0 g, 27.2 mmol) in THF (dry, 30 mL) was added trimethylsulfoxonium iodide (5.0 g, 20.6 mmol) at rt. The resulting mixture is refluxed for 2 h. Then reaction mixture is cooled to 0 °C, followed by addition of acyl chlorides (7 mmol) in THF (7 mL). The reaction was allowed to rt and stirred for 3 h. Next, the solvent was evaporated and water (15 mL) and dichloromethane (20 mL) were added to the resulting slurry. The layers were separated and the aqueous layer was washed with dichloromethane (3×50 mL) and the organic layers were combined. The organic solution was dried over anhydrous sodium sulphate (Na₂SO₄), filtered over a sintered funnel and evaporated to dryness. The crude product was purified by flash chromatography over silica gel using DCM/MeOH (95:5) to afford the corresponding sulfoxonium ylides.

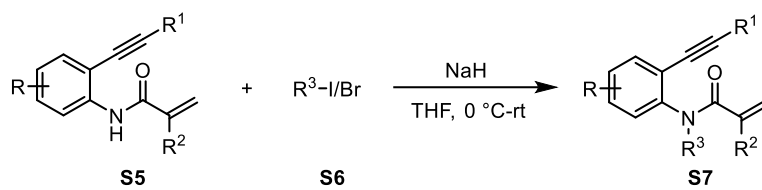
4.2 General Procedure for preparation of 1,7-enynes ^[2]:



(a) To a solution of aryl iodide **S1** (12 mmol, 1.0 equiv), PdCl₂(PPh₃)₂ (2 mol%), CuI (4 mol%) and Et₃N (3.0 equiv) in THF (dry, 20 mL) at rt under argon. After stirring for 5 minutes, the corresponding phenylacetylene **S2** (24 mmol, 2.0 equiv) was added at 0 °C. Then the reaction mixture was warmed to rt and stirred for 12 h. Upon full consumption of the starting material, the reaction mixture was extracted with NH₄Cl aqueous solution and DCM (3×20 mL). The combined organic layers were dried over Na₂SO₄, filtered and concentrated under reduced pressure. The crude product was purified by flash column chromatography (SiO₂; gradient eluent: hexane/ethyl acetate = 30:1) to provide the product **S3**.



(b) To a solution of **S3** (1.0 equiv) and Et₃N (1.5 equiv) in DCM (20 mL). The corresponding methacryloyl chloride **S4** (1.2 equiv) was added neat at 0 °C. The mixture was warmed to rt and stirred for 5 h. Then the reaction was quenched by H₂O (20 mL), and the aqueous layer was extracted with DCM (3×20 mL). The combined organic layers were dried over Na₂SO₄, filtered and concentrated under reduced pressure. The crude product was used for the next step without purification.

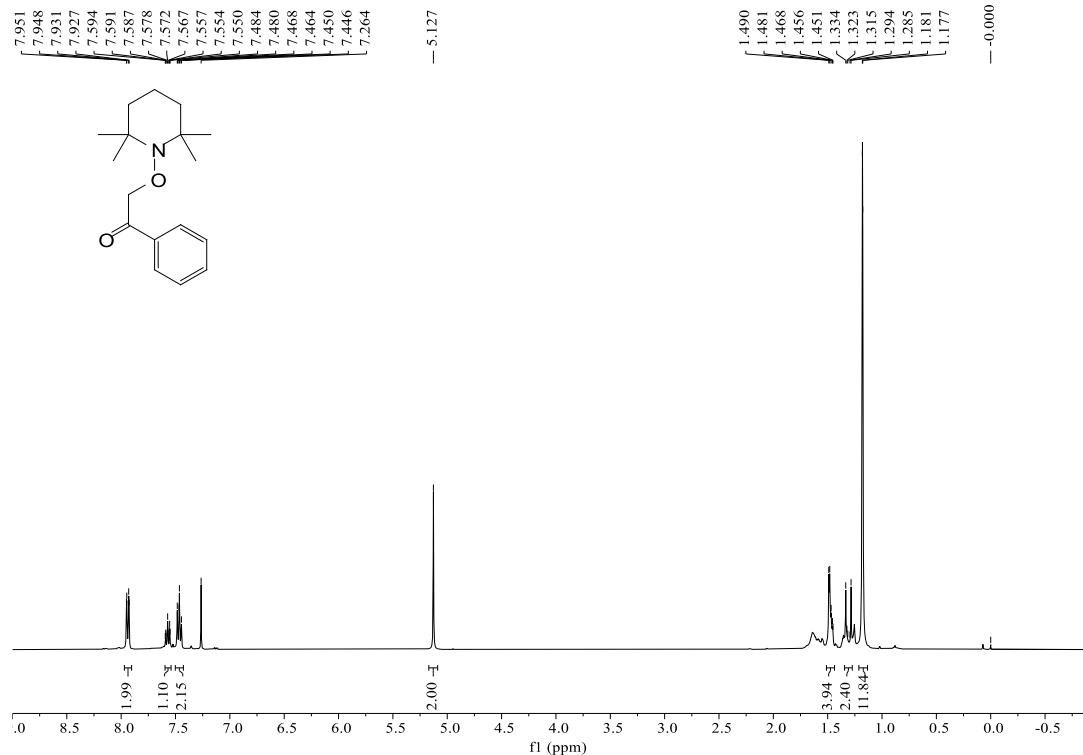
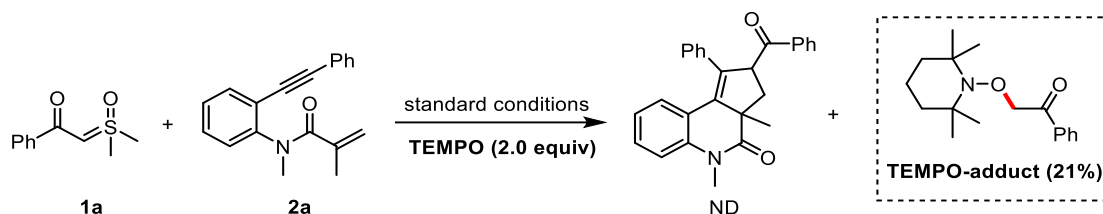


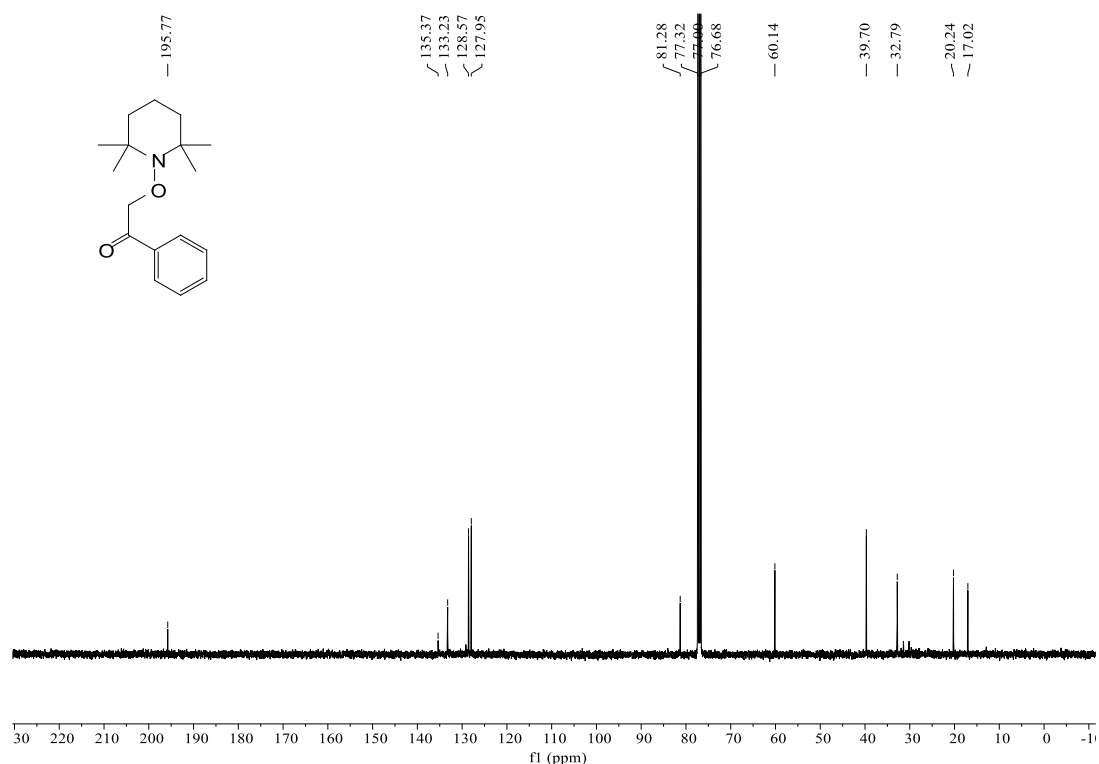
(c) To a solution of **S5** (1.0 equiv) in THF (dry, 20 mL) was added in portions NaH (2.0 equiv) at 0 °C. After stirring for 15 min at the same temperature, MeI or R-Br (1.5 equiv) was added. The mixture was warmed to rt and stirred for 6 h. Then the reaction was quenched by H₂O (20 mL), and the aqueous layer was extracted with EtOAc (3×20 mL). The combined organic layers were dried over Na₂SO₄, filtered and concentrated under reduced pressure. The crude product was purified by a flash column chromatography on silica gel using ethyl acetate and hexane as eluent.

5. Mechanistic studies

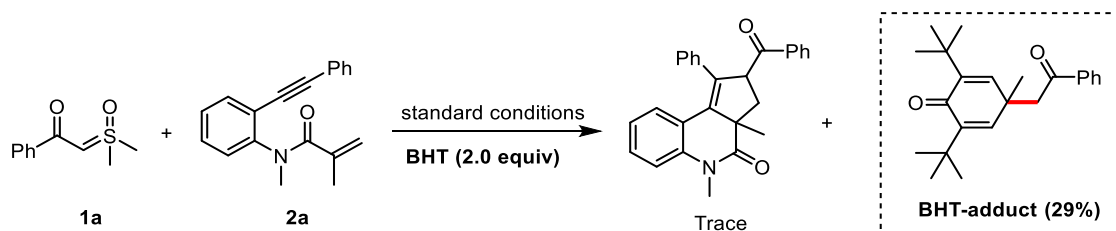
5.1 Radical trapping experiments

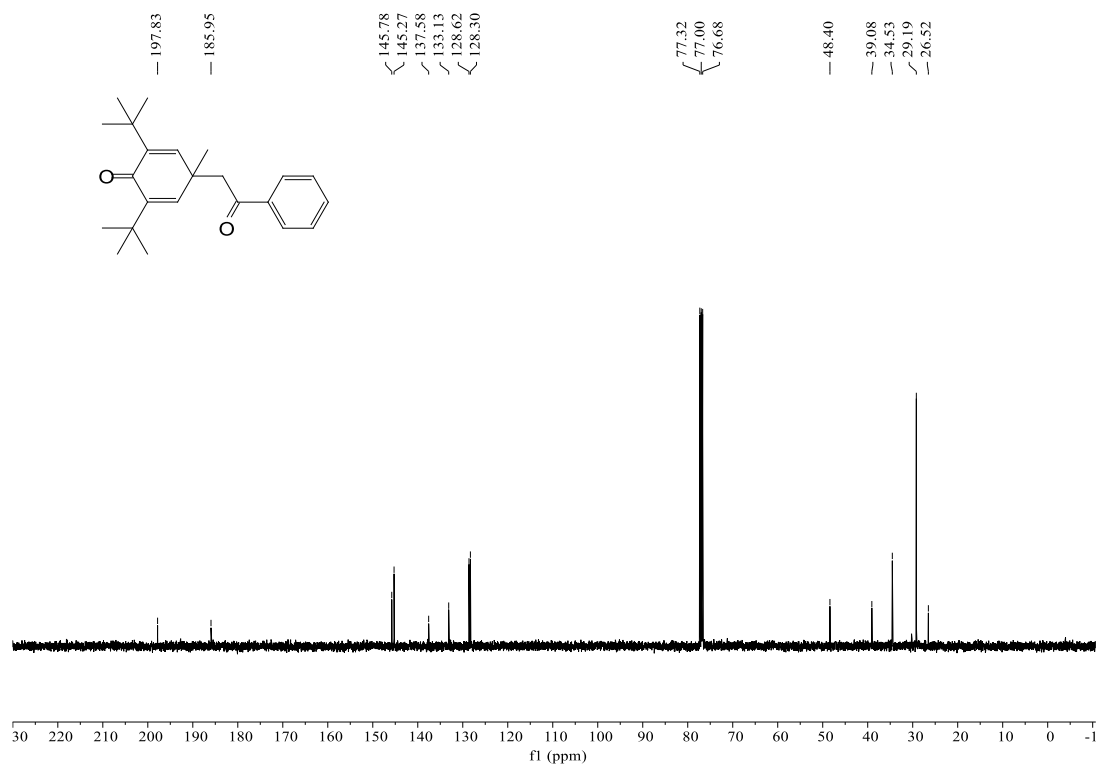
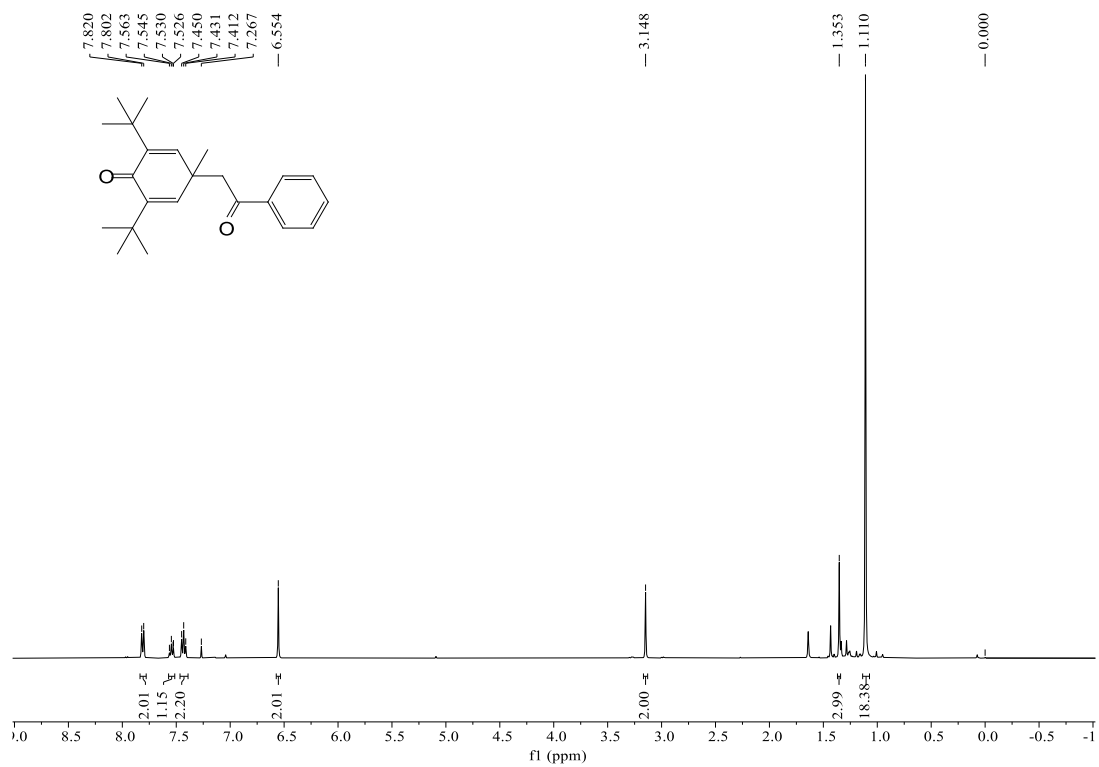
(a) The following reaction was carried out under standard conditions: A 10 mL reaction vessel was charged with *fac*-Ir(ppy)₃ (2.6 mg, 0.004 mmol), **1a** (58.9 mg, 0.3 mmol), **2a** (55.0 mg, 0.2 mmol) and 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO) (62.5 mg, 0.4 mmol, 2.0 equiv). The atmosphere was exchanged by applying vacuum and backfilling with Ar (this process was conducted for three times). Then, TfOH (20 mol%, 3.6 μ L), H₂O (3.0 equiv, 10.8 μ L) and DCM (dry, 2 mL) were added. The resulting mixture was stirred for 24 h under irradiation with a 35 W blue LEDs. After cooling to room temperature, the crude reaction mixture extracted with dichloromethane (3 \times 10 mL). The extracts were combined, dried over sodium sulfate, and filtered, and the volatiles were removed under reduced pressure. Column chromatography was performed using silica gel (200-300 mesh) (petroleum ether/ethyl acetate = 100:1) to give the TEMPO-trapped product (21%). The formation of product **3a** was completely suppressed.





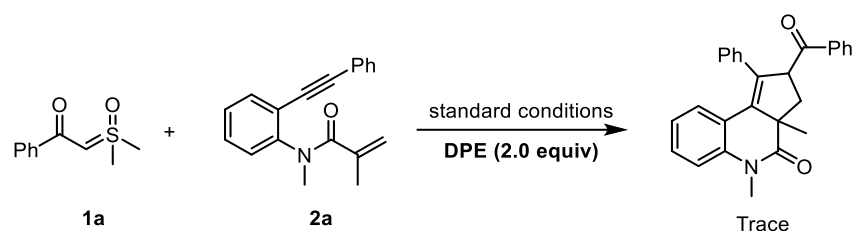
(b) The following reaction was carried out under standard conditions: The following reaction was carried out under standard conditions: A 10 mL reaction vessel was charged with *fac*-Ir(ppy)₃ (2.6 mg, 0.004 mmol), **1a** (58.9 mg, 0.3 mmol), **2a** (55.0 mg, 0.2 mmol) and hydroxytoluene (BHT) (88.1 mg, 0.4 mmol, 2.0 equiv). The atmosphere was exchanged by applying vacuum and backfilling with Ar (this process was conducted for three times). Then, TfOH (20 mol%, 3.6 μ L), H₂O (3.0 equiv, 10.8 μ L) and DCM (dry, 2 mL) were added. The resulting mixture was stirred for 24 h under irradiation with a 35 W blue LEDs. After cooling to room temperature, the crude reaction mixture extracted with dichloromethane (3 \times 10 mL). The extracts were combined, dried over sodium sulfate, and filtered, and the volatiles were removed under reduced pressure. Column chromatography was performed using silica gel (200-300 mesh) (petroleum ether/ethyl acetate = 50:1) to give the BHT-trapped product (29%). The formation of product **3a** was not observed.





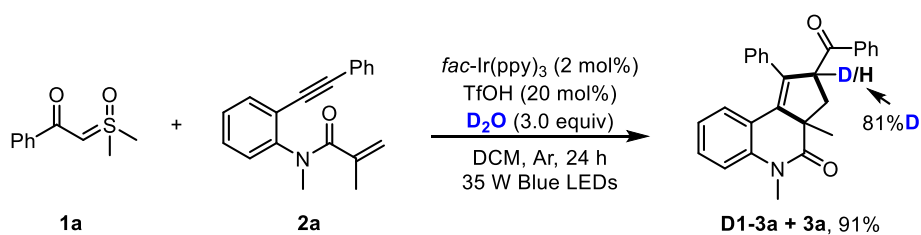
(c) The following reaction was carried out under standard conditions: The following reaction was carried out under standard conditions: A 10 mL reaction vessel was charged with *fac*-Ir(ppy)₃ (2.6 mg, 0.004 mmol), **1a** (58.9 mg, 0.3 mmol), **2a** (55.0 mg, 0.2 mmol) and 1,1-Diphenylethylene (DPE)

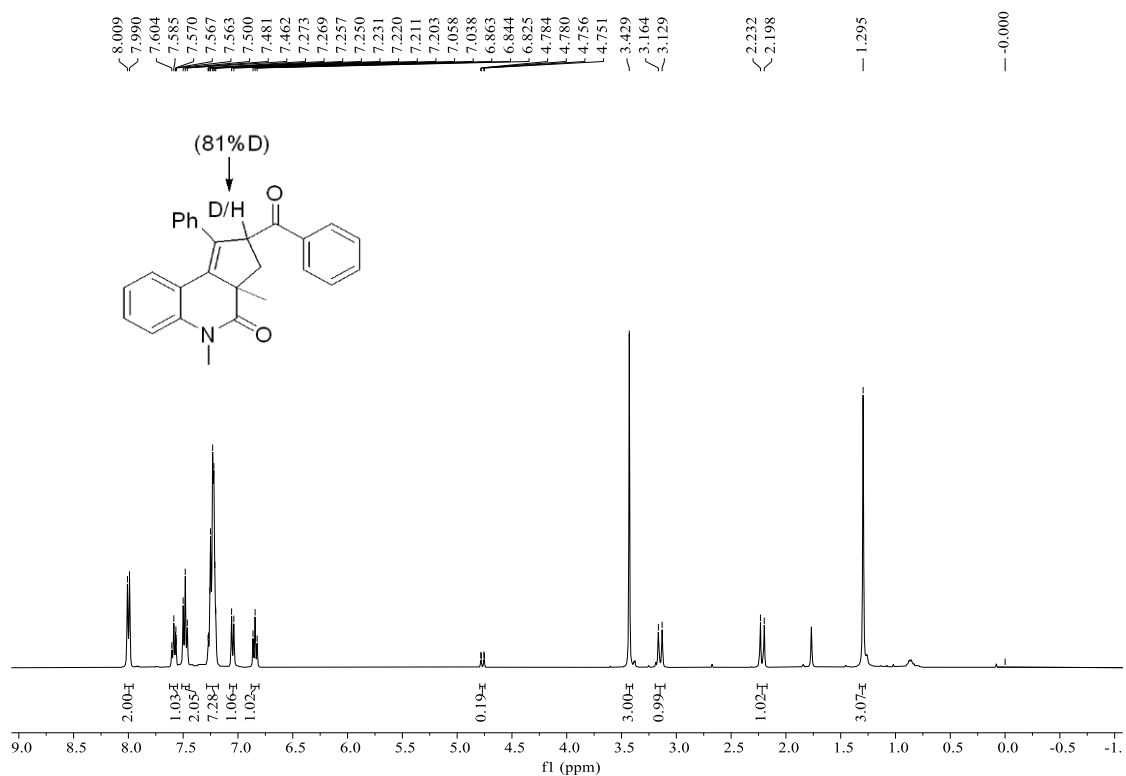
(71.0 μL , 0.4 mmol, 2.0 equiv). The atmosphere was exchanged by applying vacuum and backfilling with Ar (this process was conducted for three times). Then, TfOH (20 mol%, 3.6 μL), H_2O (3.0 equiv, 10.8 μL) and DCM (dry, 2 mL) were added. The resulting mixture was stirred for 24 h under irradiation with a 35 W blue LEDs. After cooling to room temperature, the crude reaction mixture extracted with dichloromethane (3×10 mL). The extracts were combined, dried over sodium sulfate, and filtered, and the volatiles were removed under reduced pressure. The crude residues were analyzed by GC-MS and HRMS. DPE-trapped product was not detected by GC-MS and the formation of product **3a** was not observed.



5.2 H/D exchange experiment

The following reaction was carried out under standard conditions: The following reaction was carried out under standard conditions: A 10 mL reaction vessel was charged with *fac*-Ir(ppy)₃ (2.6 mg, 0.004 mmol), **1a** (58.9 mg, 0.3 mmol) and **2a** (55.0 mg, 0.2 mmol). The atmosphere was exchanged by applying vacuum and backfilling with Ar (this process was conducted for three times). Then, TfOH (20 mol%, 3.6 μL), D_2O (3.0 equiv, 10.8 μL) and DCM (dry, 2 mL) were added. The resulting mixture was stirred for 24 h under irradiation with a 35 W blue LEDs. After cooling to room temperature, the crude reaction mixture extracted with dichloromethane (3×10 mL). The extracts were combined, dried over sodium sulfate, and filtered, and the volatiles were removed under reduced pressure. Column chromatography was performed using silica gel (200-300 mesh) (petroleum ether/acetone = 8:1) to give product **D1-3a + 3a** (91%, *dr* = 2.3:1).

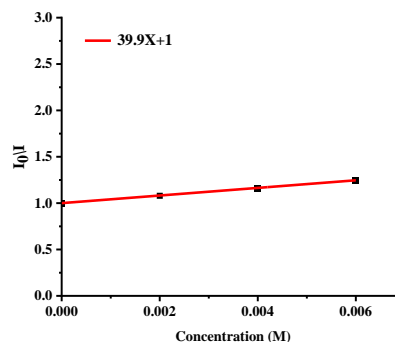
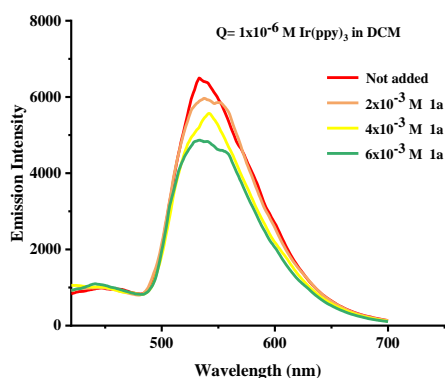




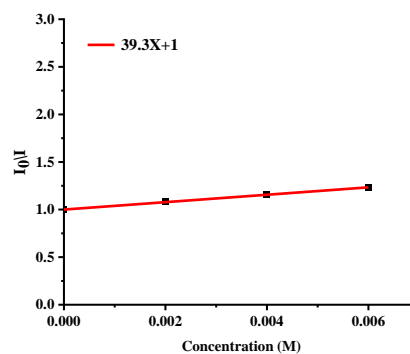
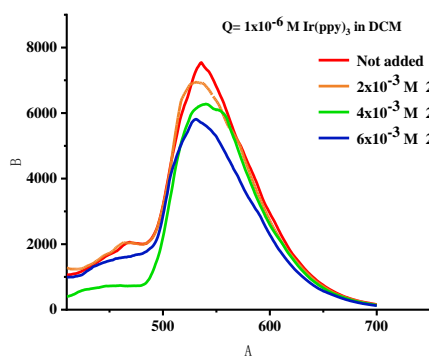
5.3 Stern–Volmer Quenching

Formulation solution: N-methyl-N-(2-(phenylethynyl)phenyl)methacrylamide (**2a**, 55 mg) was dissolved in DCM in a 5 mL volumetric flask to set the concentration to be 0.2 M. 2-(dimethyl(oxo)-λ⁶-sulfaneylidene)-1-phenylethan-1-one (**1a**, 196 mg) was dissolved in DCM in a 5 mL volumetric flask to set the concentration to be 0.2 M. Photocatalyst *fac*-Ir(ppy)₃ (1.6 mg) was dissolved in DCM (25.0 mL) to set the concentration to be 0.1 mM.

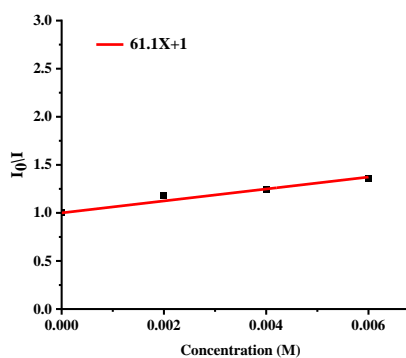
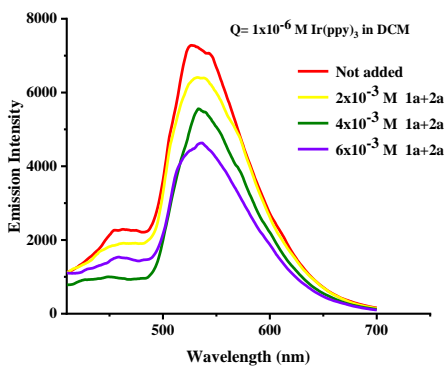
Experimental procedure: The resulting 0.1 mM solution (20 μL) was added to cuvette to obtain different concentrations of catalyst solution. This solution was then diluted to a volume of 2.0 mL by adding DCM to prepare a 1.0 μM solution. The resulting mixture was sparged with argon for 2 minutes and then irradiated at 390 nm. Fluorescence emission spectra were recorded (3 trials per sample). Into this solution, 20.0 μL of a N-methyl-N-(2-(phenylethynyl)phenyl)methacrylamide solution was successively added and uniformly stirred, and the resulting mixture was bubbled with argon for 2 minutes and irradiated at 390 nm. Fluorescence emission spectra of 0 μL, 20.0 μL, 40.0 μL, 60.0 μL fluorescence intensity. Follow this method and make changes to the amount to obtain the Stern–Volmer relationship in turn. The results were shown in the following figures.



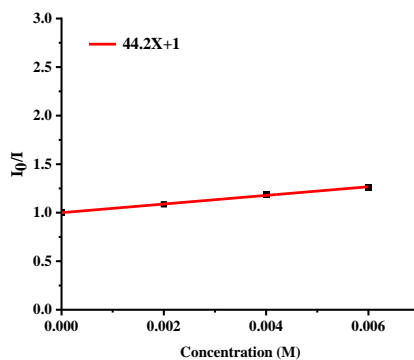
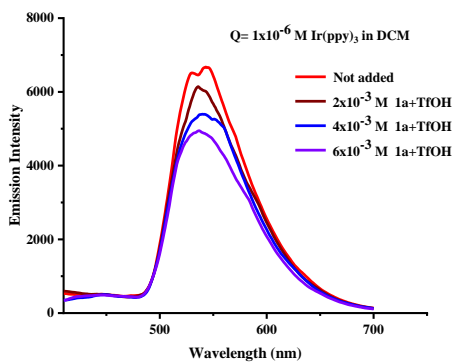
Emission quenching of *fac*-Ir(ppy)₃ with 2-(dimethyl(oxo)-λ⁶-sulfaneylidene)-1-phenylethan-1-one (**1a**) in DCM



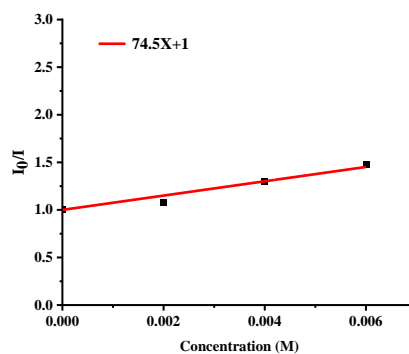
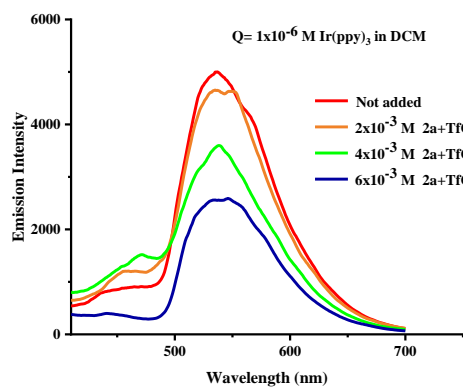
Emission quenching of *fac*-Ir(ppy)₃ with N-methyl-N-(2-(phenylethynyl)phenyl)methacrylamide (**2a**) in DCM



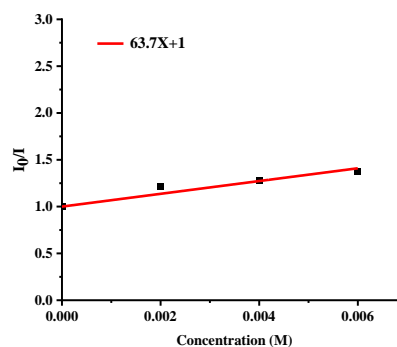
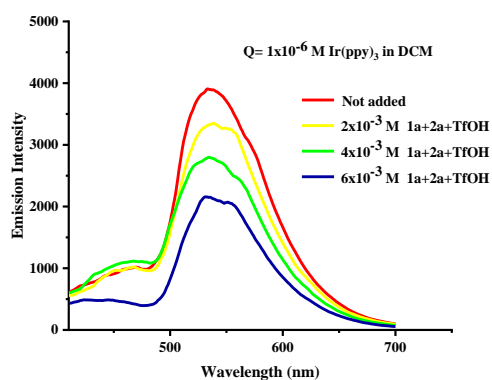
Emission quenching of *fac*-Ir(ppy)₃ with N-methyl-N-(2-(phenylethynyl)phenyl)methacrylamide (**2a**) and 2-(dimethyl(oxo)-λ⁶-sulfaneylidene)-1-phenylethan-1-one (**1a**) in DCM



Emission quenching of *fac*-Ir(ppy)₃ with 2-(dimethyl(oxo)-λ⁶-sulfaneylidene)-1-phenylethan-1-one (**1a**) and TfOH (20 mol%) in DCM



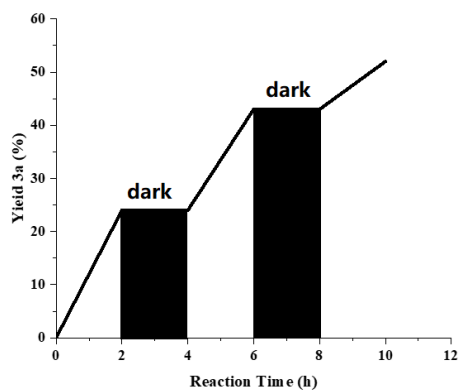
Emission quenching of *fac*-Ir(ppy)₃ with N-methyl-N-(2-(phenylethynyl)phenyl)methacrylamide (**2a**) and TfOH (20 mol%) in DCM



Emission quenching of *fac*-Ir(ppy)₃ with N-methyl-N-(2-(phenylethynyl)phenyl)methacrylamide (**2a**), 2-(dimethyl(oxo)-λ⁶-sulfaneylidene)-1-phenylethan-1-one (**1a**) and TfOH (20 mol%) in DCM

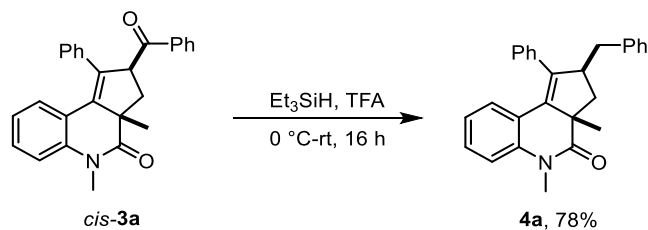
5.4 Light on/off experiment

Conducted the relationship of products with light on-off under standard conditions. Subsequent samples (each 20 μL) taken at regular time intervals and determined by GC with dodecane as the internal standard.

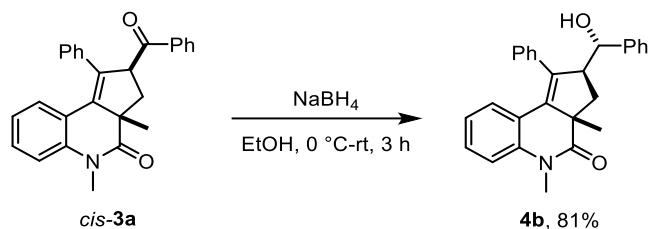


Plot of light on-off experiments

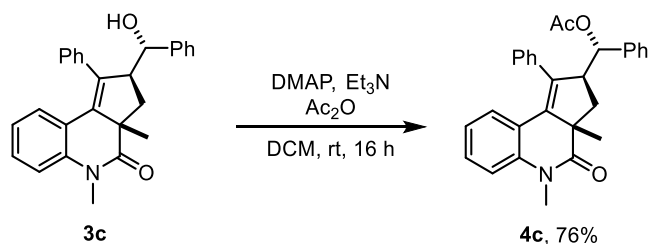
6. Late-stage derivation and application



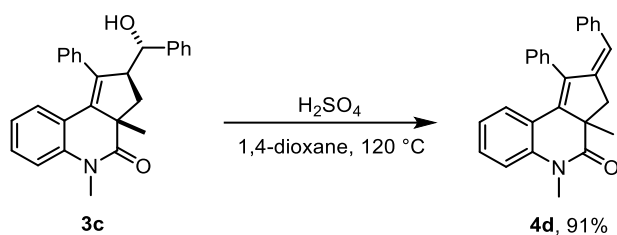
(a) Under argon atmosphere, to a stirred solution of the product *cis*-**3a** (78.6 mg, 0.2 mmol, 1.0 equiv) in TFA (3.6 mL) at 0 °C was dropwise added Et₃SiH (0.6 mmol, 3.0 equiv). After stirring for 10 min, the mixture was slowly raised to room temperature and stirred at same temperature for 16 h. The solvent was removed by vacuum evaporation. The crude product was purified by flash column chromatography (petroleum ether/ ethyl acetate = 20:1) to afford product **4a** (58.4 mg, 78% yield).



(b) To a stirred solution of product *cis*-**3a** (78.6 mg, 0.2 mmol, 1.0 equiv) in anhydrous EtOH at 0 °C were added NaBH₄ (0.6 mmol, 3.0 equiv), and the mixture was stirred at room temperature for 12 h. [3] The reactions were stopped by the addition of sat. aqueous NaHCO₃, and diluted by EtOAc (3 x 20 mL). The combined organic phases were dried over Na₂SO₄ and then filtered. The organic solvent was evaporated under reduced pressure. The crude product was purified by flash column chromatography (petroleum ether/ethyl acetate 3:1) to afford the alcohol product **4b** (64.1 mg, 81% yield, *dr* > 20:1).



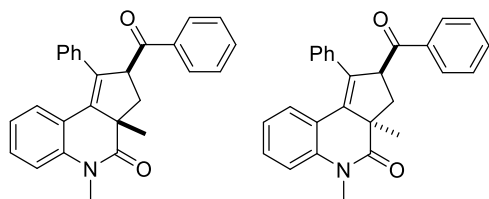
(c) To a stirred solution of the alcohol **3c** (39.5 mg, 0.1 mmol, 1.0 equiv) in DCM at room temperature were added triethylamine (0.2 mmol, 2.0 equiv) and acetic anhydride (Ac₂O) (0.2 mmol, 2.0 equiv), and the mixture was stirred for 12 h. [4] The reactions were stopped by the addition of 10% HCl (1 mL) and the acetate produced was extracted with DCM (3 x 20 mL). The combined organic phases were dried over Na₂SO₄ and then filtered. The organic solvent was evaporated under reduced pressure. The crude product was purified by flash column chromatography (petroleum ether/ethyl acetate 6:1) to afford the desired product **4c** (33.2 mg, 76% yield, *dr* > 20:1).



(d) To a stirred solution of the alcohol **3c** (79.0 mg, 0.2 mmol, 1.0 equiv) in 1,4-dioxane (2 mL) was added 2-3 drops H_2SO_4 , and the mixture was stirred at 120 °C for 6 h. ^[4] The mixture was quenched by sat. aqueous NaHCO_3 , and diluted by EtOAc. The organic phase was collected, and the aqueous phase was extracted with EtOAc. The combined organic layers were dried over anhydrous Na_2SO_4 , filtered, and concentrated under reduced pressure. The crude product was purified by flash column chromatography (petroleum ether/ethyl acetate 15:1) to afford product **4d** (68.6 mg, 91% yield), respectively.

7. Characterization data of all products

2-benzoyl-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3a)



Following the general procedure A, *cis*-**3a** (50.9 mg) as a white solid (mp 55-57 °C) and *trans*-**3a** (22.2 mg) as a white solid (mp 146-148 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3a**: 93%, *dr*: 2.3:1.

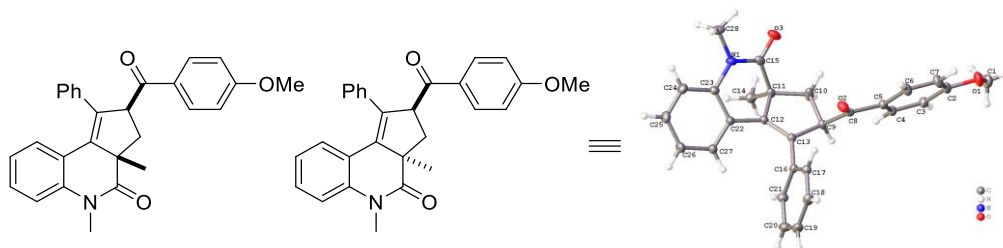
Data of *cis*-**3a**

¹H NMR (400 MHz, CDCl₃) δ 8.03 – 7.97 (m, 2H), 7.57 (t, *J* = 7.3 Hz, 1H), 7.47 (t, *J* = 7.6 Hz, 2H), 7.29 – 7.17 (m, 8H), 7.04 (d, *J* = 8.5 Hz, 1H), 6.83 (t, *J* = 7.5 Hz, 1H), 4.81 – 4.74 (m, 1H), 3.42 (s, 3H), 3.21 – 3.11 (m, 1H), 2.27 – 2.18 (m, 1H), 1.30 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.0, 175.3, 139.9, 137.9, 136.9, 136.7, 136.2, 133.2, 128.8, 128.7, 128.64, 128.61, 128.34, 128.30, 127.50, 127.48, 122.5, 121.0, 114.9, 57.2, 54.4, 38.9, 29.9, 24.8. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₃NNaO₂⁺ 416.1621; Found 416.1634.

Data of *trans*-**3a**

¹H NMR (400 MHz, CDCl₃) δ 7.93 – 7.87 (m, 2H), 7.51 (t, *J* = 7.4 Hz, 1H), 7.40 (t, *J* = 7.7 Hz, 2H), 7.28 – 7.15 (m, 6H), 7.12 – 7.02 (m, 2H), 6.81 (t, *J* = 7.5 Hz, 1H), 5.28 (t, *J* = 8.9 Hz, 1H), 3.41 (s, 3H), 2.75 – 2.66 (m, 1H), 2.63 – 2.54 (m, 1H), 1.34 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.4, 174.5, 140.1, 137.3, 136.9, 136.5, 136.0, 133.0, 128.8, 128.5, 128.4, 128.3, 127.9, 127.4, 122.2, 120.8, 115.2, 54.5, 53.6, 39.9, 29.9, 23.7. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₃NNaO₂⁺ 416.1621; Found 416.1636.

2-(4-methoxybenzoyl)-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3b)



trans-**3b**, CCDC No. 2369006

Following the general procedure A, *cis*-**3b** (52.3 mg) as a white solid (mp 69-71 °C) and *trans*-**3b** (16.3 mg) as a white solid (mp 228-230 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 7:1). Total isolated yield of **3b**: 81%, *dr*: 3.2:1.

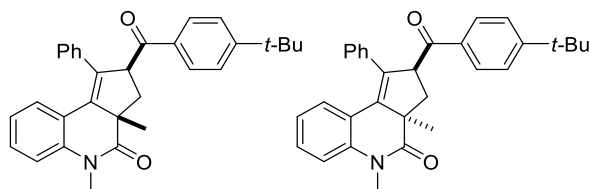
Data of *cis*-**3b**

^1H NMR (400 MHz, CDCl_3) δ 8.01 – 7.95 (m, 2H), 7.27 – 7.18 (m, 7H), 7.04 (d, $J = 8.1$ Hz, 1H), 6.98 – 6.92 (m, 2H), 6.83 (t, $J = 7.5$ Hz, 1H), 4.76 – 4.69 (m, 1H), 3.87 (s, 3H), 3.43 (s, 3H), 3.18 – 3.09 (m, 1H), 2.24 – 2.17 (m, 1H), 1.30 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 200.5, 175.4, 163.6, 139.9, 137.7, 137.1, 136.8, 131.0, 129.3, 128.7, 128.6, 128.4, 128.3, 127.5, 122.5, 121.1, 114.9, 113.8, 56.8, 55.5, 54.4, 39.1, 29.9, 24.9. HRMS (ESI) m/z : $[\text{M}+\text{H}]^+$ Calcd for $\text{C}_{28}\text{H}_{26}\text{NNaO}_3^+$ 424.1907; Found 424.1905.

Data of *trans*-**3b**

^1H NMR (400 MHz, CDCl_3) δ 7.93 – 7.88 (m, 2H), 7.27 – 7.15 (m, 6H), 7.12 – 7.02 (m, 2H), 6.92 – 6.86 (m, 2H), 6.81 (t, $J = 7.6$ Hz, 1H), 5.25 – 5.17 (m, 1H), 3.85 (s, 3H), 3.42 (s, 3H), 2.72 – 2.63 (m, 1H), 2.60 – 2.52 (m, 1H), 1.33 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 198.8, 174.6, 163.5, 140.2, 137.6, 136.4, 136.2, 130.8, 129.9, 128.8, 128.4, 127.9, 127.5, 127.4, 122.3, 121.0, 115.2, 113.7, 55.4, 54.2, 53.6, 40.1, 29.9, 23.7. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{28}\text{H}_{26}\text{NNaO}_3^+$ 446.1727; Found 446.1733.

2-(4-(*tert*-butyl)benzoyl)-3*a*,5-dimethyl-1-phenyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3c**)



Following the general procedure A, *cis*-**3c** (58.4 mg) as a white solid (mp 78-80 °C) and *trans*-**3c** (24.3 mg) as a yellow solid (mp 172-174 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3c**: 92%, *dr*: 2.4:1.

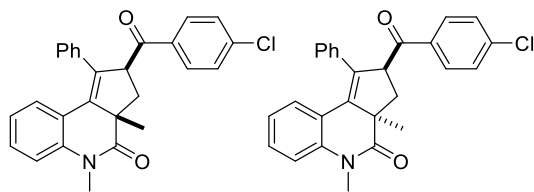
Data of *cis*-**3c**

^1H NMR (400 MHz, CDCl_3) δ 7.97 – 7.91 (m, 2H), 7.52 – 7.46 (m, 2H), 7.27 – 7.17 (m, 7H), 7.04 (d, $J = 8.5$ Hz, 1H), 6.83 (t, $J = 7.5$ Hz, 1H), 4.77 – 4.71 (m, 1H), 3.42 (s, 3H), 3.19 – 3.09 (m, 1H), 2.25 – 2.17 (m, 1H), 1.34 (s, 9H), 1.28 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 201.6, 175.4, 157.1, 139.9, 137.8, 137.1, 136.8, 133.7, 128.8, 128.71, 128.67, 128.4, 128.3, 127.5, 125.6, 122.5, 121.2, 114.9, 57.2, 54.4, 39.1, 35.1, 31.1, 29.9, 24.8. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{31}\text{H}_{31}\text{NNaO}_2^+$ 472.2247; Found 472.2266.

Data of *trans*-**3c**

^1H NMR (400 MHz, CDCl_3) δ 7.86 (d, $J = 8.3$ Hz, 2H), 7.43 (d, $J = 8.3$ Hz, 2H), 7.28 – 7.16 (m, 7H), 7.12 – 7.04 (m, 2H), 6.82 (t, $J = 7.5$ Hz, 1H), 5.25 (t, $J = 8.9$ Hz, 1H), 3.42 (s, 3H), 2.70 – 2.62 (m, 1H), 2.61 – 2.54 (m, 1H), 1.33 (s, 12H). ^{13}C NMR (100 MHz, CDCl_3) δ 199.9, 174.6, 156.8, 140.2, 137.5, 136.5, 136.2, 134.3, 128.8, 128.4, 127.9, 127.5, 127.4, 125.5, 122.3, 121.0, 115.2, 54.6, 53.7, 40.1, 35.1, 31.0, 29.9, 23.8. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{28}\text{H}_{26}\text{NNaO}_3^+$ 472.2247; Found 472.2269.

2-(4-chlorobenzoyl)-3*a*,5-dimethyl-1-phenyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3d**)



Following the general procedure A, **cis-3d** (54.9 mg) as a white solid (mp 61-63 °C) and **trans-3d** (20.4 mg) as a yellow solid (mp 202-204 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3d**: 88%, *dr*: 2.7:1.

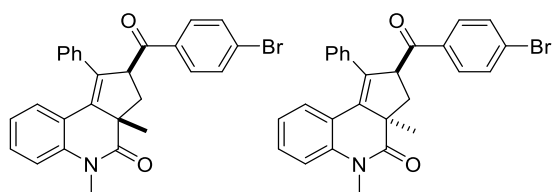
Data of **cis-3d**

¹H NMR (400 MHz, CDCl₃) δ 7.95 – 7.89 (m, 2H), 7.44 (d, *J* = 8.5 Hz, 2H), 7.28 – 7.20 (m, 7H), 7.04 (d, *J* = 8.1 Hz, 1H), 6.83 (t, *J* = 7.5 Hz, 1H), 4.76 – 4.68 (m, 1H), 3.42 (s, 3H), 3.19 – 3.10 (m, 1H), 2.23 – 2.16 (m, 1H), 1.30 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.7, 175.1, 139.8, 139.7, 138.1, 136.6, 136.5, 134.5, 130.1, 128.93, 128.85, 128.6, 128.34, 128.27, 127.6, 122.5, 120.9, 114.9, 57.2, 54.4, 38.7, 29.9, 24.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₂ClNNaO₂⁺ 450.1231; Found 450.1234.

Data of **trans-3d**

¹H NMR (400 MHz, CDCl₃) δ 7.87 – 7.80 (m, 2H), 7.41 – 7.35 (m, 2H), 7.29 – 7.17 (m, 6H), 7.12 – 7.04 (m, 2H), 6.82 (t, *J* = 7.5 Hz, 1H), 5.19 (t, *J* = 8.9 Hz, 1H), 3.42 (s, 3H), 2.74 – 2.65 (m, 1H), 2.60 – 2.52 (m, 1H), 1.33 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 199.3, 174.4, 140.2, 139.5, 137.0, 136.8, 135.9, 135.2, 129.8, 129.0, 128.9, 128.5, 127.9, 127.6, 127.5, 122.3, 120.8, 115.3, 54.8, 53.7, 39.8, 29.9, 23.8. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₂ClNNaO₂⁺ 450.1231; Found 450.1257.

2-(4-bromobenzoyl)-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (**3e**)



Following the general procedure A, **cis-3e** (54.5 mg) as a white solid (mp 139-141 °C) and **trans-3e** (30.0 mg) as a yellow solid (mp 223-225 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3e**: 80%, *dr*: 2.6:1.

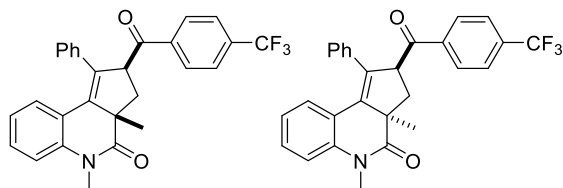
Data of **cis-3e**

¹H NMR (400 MHz, CDCl₃) δ 7.87 – 7.82 (m, 2H), 7.64 – 7.58 (m, 2H), 7.28 – 7.18 (m, 7H), 7.05 (d, *J* = 8.2 Hz, 1H), 6.84 (t, *J* = 7.5 Hz, 1H), 4.73 – 4.67 (m, 1H), 3.43 (s, 3H), 3.18 – 3.09 (m, 1H), 2.23 – 2.15 (m, 1H), 1.29 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 201.0, 175.2, 139.9, 138.1, 136.6, 136.5, 135.0, 132.0, 130.2, 128.9, 128.6, 128.5, 128.4, 128.3, 127.6, 122.5, 120.9, 114.9, 57.2, 54.4, 38.8, 29.9, 24.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₂BrNNaO₂⁺ 494.0726; Found 494.0724.

Data of **trans-3e**

^1H NMR (400 MHz, CDCl_3) δ 7.79 – 7.72 (m, 2H), 7.55 (d, J = 8.5 Hz, 2H), 7.29 – 7.17 (m, 6H), 7.07 (t, J = 8.3 Hz, 2H), 6.82 (t, J = 7.6 Hz, 1H), 5.23 – 5.14 (m, 1H), 3.42 (s, 3H), 2.76 – 2.65 (m, 1H), 2.62 – 2.51 (m, 1H), 1.33 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 199.5, 174.4, 140.2, 137.0, 136.8, 135.9, 135.6, 131.9, 129.9, 129.0, 128.5, 128.3, 127.9, 127.6, 127.5, 122.4, 120.8, 115.3, 54.8, 53.7, 39.8, 30.0, 23.8. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{27}\text{H}_{22}\text{BrNNaO}_2^+$ 494.0726; Found 494.0750.

2-(4-trifluoromethyl)-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3f)



Following the general procedure A, *cis*-**3f** (65.3 mg) as a white solid (mp 81-83 °C) and *trans*-**3f** (25.1 mg) as a yellow solid (mp 178-180 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3f**: 98%, *dr*: 2.6:1.

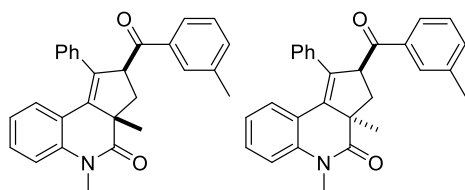
Data of *cis*-**3f**

^1H NMR (400 MHz, CDCl_3) δ 8.09 (d, J = 8.1 Hz, 2H), 7.74 (d, J = 8.1 Hz, 2H), 7.30 – 7.19 (m, 7H), 7.06 (d, J = 8.2 Hz, 1H), 6.85 (t, J = 7.6 Hz, 1H), 4.76 (d, J = 10.6 Hz, 1H), 3.43 (s, 3H), 3.23 – 3.13 (m, 1H), 2.26 – 2.17 (m, 1H), 1.30 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 201.1, 175.1, 139.9, 139.0, 138.4, 136.5, 136.3, 134.5 (q, J = 32.7 Hz), 129.02, 128.98, 128.6, 128.4, 128.3, 127.7, 125.7 (q, J = 3.7 Hz), 123.5 (d, J = 272.6 Hz), 122.6, 120.9, 115.0, 57.6, 54.5, 38.6, 29.9, 25.0. ^{19}F NMR (376 MHz, CDCl_3) δ -63.1. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{28}\text{H}_{22}\text{F}_3\text{NNaO}_2^+$ 484.1495; Found 484.1518.

Data of *trans*-**3f**

^1H NMR (400 MHz, CDCl_3) δ 7.97 (d, J = 8.1 Hz, 2H), 7.67 (d, J = 8.1 Hz, 2H), 7.30 – 7.18 (m, 6H), 7.12 – 7.05 (m, 2H), 6.83 (t, J = 7.6 Hz, 1H), 5.22 (t, J = 8.9 Hz, 1H), 3.43 (s, 3H), 2.78 – 2.68 (m, 1H), 2.63 – 2.54 (m, 1H), 1.35 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 199.7, 174.4, 140.2, 139.6, 137.0, 136.7, 135.8, 134.2 (q, J = 32.7 Hz), 129.1, 128.7, 128.6, 127.9, 127.7, 127.5, 125.6 (q, J = 3.7 Hz), 123.5 (q, J = 272.6 Hz), 122.4, 120.7, 115.3, 55.3, 53.8, 39.6, 30.0, 23.9. ^{19}F NMR (376 MHz, CDCl_3) δ -63.1. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{28}\text{H}_{22}\text{F}_3\text{NNaO}_2^+$ 484.1495; Found 484.1518.

3a,5-dimethyl-2-(3-methylbenzoyl)-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3g)



Following the general procedure A, **cis-3g** (51.1 mg) as a white solid (mp 45-47 °C) and **trans-3g** (18.2 mg) as a yellow solid (mp 124-126 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3g**: 85%, *dr*: 2.8:1.

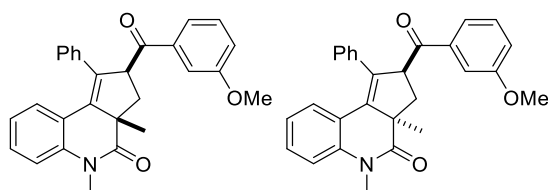
Data of **cis-3g**

¹H NMR (400 MHz, CDCl₃) δ 7.83 – 7.76 (m, 2H), 7.42 – 7.33 (m, 2H), 7.28 – 7.18 (m, 7H), 7.04 (d, *J* = 8.6 Hz, 1H), 6.84 (t, *J* = 7.5 Hz, 1H), 4.80 – 4.72 (m, 1H), 3.43 (s, 3H), 3.21 – 3.11 (m, 1H), 2.41 (s, 3H), 2.26 – 2.18 (m, 1H), 1.29 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.1, 175.4, 139.9, 138.5, 137.8, 137.0, 136.8, 136.3, 134.0, 129.2, 128.8, 128.6, 128.5, 128.4, 128.3, 127.5, 125.9, 122.5, 121.1, 114.9, 57.3, 54.4, 39.0, 29.9, 24.9, 21.3. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₂⁺ 430.1778; Found 430.1800.

Data of **trans-3g**

¹H NMR (400 MHz, CDCl₃) δ 7.74 – 7.67 (m, 2H), 7.37 – 7.30 (m, 2H), 7.30 – 7.16 (m, 7H), 7.12 – 7.04 (m, 2H), 6.82 (t, *J* = 7.5 Hz, 1H), 5.26 (t, *J* = 8.9 Hz, 1H), 3.42 (s, 3H), 2.72 – 2.64 (m, 1H), 2.62 – 2.55 (m, 1H), 2.37 (s, 3H), 1.34 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.6, 174.6, 140.2, 138.4, 137.4, 137.0, 136.6, 136.1, 133.8, 128.9, 128.9, 128.4, 127.9, 127.5, 127.4, 125.6, 122.3, 121.0, 115.2, 54.6, 53.7, 40.1, 29.9, 23.8, 21.3. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₂⁺ 430.1778; Found 430.1809.

2-(3-methoxybenzoyl)-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[*c*]quinolin-4-one (**3h**)



Following the general procedure A, **cis-3h** (59.8 mg) as a colorless liquid and **trans-3h** (24.9 mg) as a white solid (mp 158-160 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 6:1). Total isolated yield of **3h**: 86%, *dr*: 2.4:1.

Data of **cis-3h**

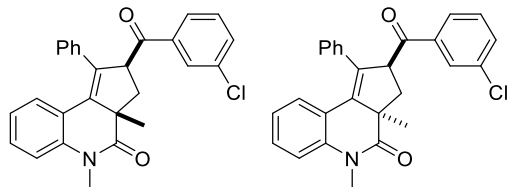
¹H NMR (400 MHz, CDCl₃) δ 7.57 – 7.52 (m, 2H), 7.37 (t, *J* = 8.2 Hz, 1H), 7.27 – 7.19 (m, 7H), 7.15 – 7.10 (m, 1H), 7.05 (d, *J* = 8.1 Hz, 1H), 6.84 (t, *J* = 7.5 Hz, 1H), 4.78 – 4.72 (m, 1H), 3.86 (s, 3H), 3.43 (s, 3H), 3.19 – 3.10 (m, 1H), 2.26 – 2.19 (m, 1H), 1.30 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 201.8, 175.3, 159.9, 139.9, 137.9, 137.6, 136.9, 136.7, 129.6, 128.8, 128.6, 128.3, 127.5, 122.5, 121.3, 121.1, 119.8, 114.9, 112.8, 57.4, 55.4, 54.4, 39.0, 29.9, 24.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₃⁺ 446.1727; Found 446.1757.

Data of **trans-3h**

¹H NMR (400 MHz, CDCl₃) δ 7.74 – 7.67 (m, 2H), 7.37 – 7.30 (m, 2H), 7.30 – 7.16 (m, 7H), 7.12 – 7.04 (m, 2H), 6.82 (t, *J* = 7.5 Hz, 1H), 5.26 (t, *J* = 8.9 Hz, 1H), 3.42 (s, 3H), 2.72 – 2.64 (m, 1H), 2.62 – 2.55 (m, 1H), 2.37 (s, 3H), 1.34 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.6, 174.6, 140.2, 138.4, 137.4, 137.0, 136.6, 136.1, 133.8, 128.9, 128.9, 128.4, 127.9, 127.5, 127.4, 125.6, 122.3,

121.0, 115.2, 54.6, 53.7, 40.1, 29.9, 23.8, 21.3. HRMS (ESI) m/z : $[M+Na]^+$ Calcd for $C_{28}H_{25}NNaO_3^+$ 446.1727; Found 446.1757.

2-(3-chlorobenzoyl)-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3i)



Following the general procedure A, *cis*-**3i** (58.0 mg) as a white solid (mp 53-55 °C) and *trans*-**3i** (20.7 mg) as a white solid (mp 135-137 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3i**: 92%, *dr*: 2.8:1.

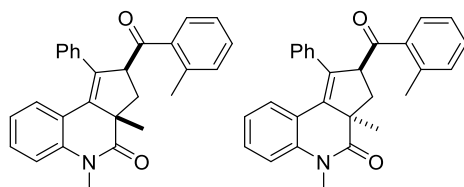
Data of *cis*-**3i**

1H NMR (400 MHz, $CDCl_3$) δ 7.97 (s, 1H), 7.84 (d, $J = 7.8$ Hz, 1H), 7.55 (d, $J = 8.4$ Hz, 1H), 7.41 (t, $J = 7.9$ Hz, 1H), 7.22 (s, 7H), 7.05 (d, $J = 8.2$ Hz, 1H), 6.84 (t, $J = 7.5$ Hz, 1H), 4.74 – 4.67 (m, 1H), 3.43 (s, 3H), 3.21 – 3.11 (m, 1H), 2.24 – 2.17 (m, 1H), 1.29 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 200.7, 175.2, 139.9, 138.2, 137.8, 136.6, 136.4, 135.1, 133.2, 130.0, 128.9, 128.8, 128.6, 128.4, 128.3, 127.6, 126.8, 122.6, 120.9, 115.0, 57.4, 54.4, 38.7, 29.9, 24.9. HRMS (ESI) m/z : $[M+Na]^+$ Calcd for $C_{27}H_{22}ClNNaO_2^+$ 450.1231; Found 450.1232.

Data of *trans*-**3i**

1H NMR (400 MHz, $CDCl_3$) δ 7.84 (s, 1H), 7.77 (d, $J = 7.4$ Hz, 1H), 7.52 – 7.47 (m, 1H), 7.35 (t, $J = 7.9$ Hz, 1H), 7.29 – 7.18 (m, 6H), 7.07 (t, $J = 7.2$ Hz, 2H), 6.82 (t, $J = 7.6$ Hz, 1H), 5.24 – 5.14 (m, 1H), 3.43 (s, 3H), 2.76 – 2.66 (m, 1H), 2.61 – 2.53 (m, 1H), 1.34 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 199.2, 174.4, 140.2, 138.4, 136.9, 136.9, 135.9, 134.9, 133.0, 129.9, 129.0, 128.51, 128.49, 127.9, 127.6, 127.5, 126.5, 122.4, 120.8, 115.3, 55.0, 53.8, 39.7, 30.0, 23.9. HRMS (ESI) m/z : $[M+Na]^+$ Calcd for $C_{27}H_{22}ClNNaO_2^+$ 450.1231; Found 450.1259.

3a,5-dimethyl-2-(2-methylbenzoyl)-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3j)



Following the general procedure A, *cis*-**3j** (53.0 mg) as a white solid (mp 46-48 °C) and *trans*-**3j** (14.7 mg) as a pale yellow solid (mp 140-142 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3j**: 83%, *dr*: 3.6:1.

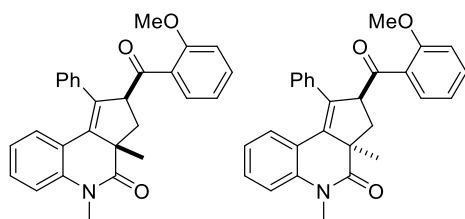
Data of *cis*-**3j**

^1H NMR (400 MHz, CDCl_3) δ 7.62 (d, $J = 7.7$ Hz, 1H), 7.39 (t, $J = 7.5$ Hz, 1H), 7.31 – 7.20 (m, 9H), 7.04 (d, $J = 8.0$ Hz, 1H), 6.84 (t, $J = 7.5$ Hz, 1H), 4.72 – 4.64 (m, 1H), 3.42 (s, 3H), 3.11 – 3.01 (m, 1H), 2.56 (s, 3H), 2.21 – 2.13 (m, 1H), 1.34 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 205.7, 175.4, 139.9, 138.8, 138.0, 137.5, 137.1, 136.8, 132.1, 131.6, 128.9, 128.8, 128.6, 128.4, 128.4, 127.6, 125.7, 122.5, 121.1, 114.9, 59.9, 54.3, 38.6, 29.9, 25.0, 21.4. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{28}\text{H}_{25}\text{NNaO}_2^+$ 430.1778; Found 430.1798.

Data of *trans*-**3i**

^1H NMR (400 MHz, CDCl_3) δ 7.58 (d, $J = 7.6$ Hz, 1H), 7.35 – 7.29 (m, 1H), 7.26 – 7.22 (m, 2H), 7.19 (s, 5H), 7.13 (d, $J = 7.5$ Hz, 1H), 7.08 – 7.03 (m, 2H), 6.83 – 6.77 (m, 1H), 5.12 (dd, $J = 9.6$, 7.8 Hz, 1H), 3.43 (s, 3H), 2.82 – 2.73 (m, 1H), 2.59 – 2.50 (m, 1H), 2.05 (s, 3H), 1.31 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 205.1, 174.6, 140.3, 139.0, 138.1, 137.7, 136.6, 136.0, 131.7, 131.1, 128.9, 128.4, 128.1, 127.7, 127.5, 125.4, 122.3, 120.8, 115.2, 57.4, 53.6, 39.8, 29.9, 23.8, 20.3. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{28}\text{H}_{25}\text{NNaO}_2^+$ 430.1778; Found 430.1790.

2-(2-methoxybenzoyl)-3*a*,5-dimethyl-1-phenyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3k**)



Following the general procedure A, *cis*-**3k** (43.1 mg) as a pale yellow solid (mp 83-85 °C) and *trans*-**3k** (17.9 mg) as a pale yellow solid (mp 45-47 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 5:1). Total isolated yield of **3k**: 72%, *dr*: 2.4:1.

Data of *cis*-**3k**

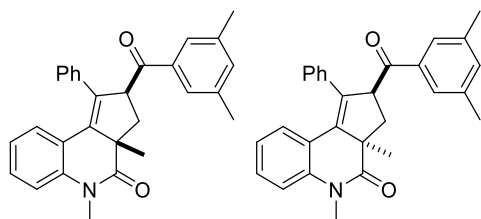
^1H NMR (400 MHz, CDCl_3) δ 7.82 – 7.77 (m, 1H), 7.51 – 7.45 (m, 1H), 7.29 – 7.19 (m, 7H), 7.06 – 6.99 (m, 2H), 6.95 (d, $J = 8.5$ Hz, 1H), 6.83 (t, $J = 7.5$ Hz, 1H), 4.82 – 4.75 (m, 1H), 3.84 (s, 3H), 3.42 (s, 3H), 3.07 – 2.97 (m, 1H), 2.30 – 2.21 (m, 1H), 1.29 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 204.4, 175.8, 158.7, 140.0, 137.9, 137.4, 137.2, 133.8, 130.8, 128.7, 128.6, 128.5, 128.2, 128.0, 127.3, 122.5, 121.3, 120.7, 114.9, 111.6, 61.8, 55.5, 53.9, 38.8, 29.9, 24.7. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{28}\text{H}_{25}\text{NNaO}_3^+$ 446.1727; Found 446.1757.

Data of *trans*-**3k**

^1H NMR (400 MHz, CDCl_3) δ 7.43 – 7.37 (m, 1H), 7.26 – 7.13 (m, 7H), 7.09 – 7.02 (m, 2H), 6.93 (d, $J = 8.4$ Hz, 1H), 6.86 (t, $J = 7.5$ Hz, 1H), 6.79 (t, $J = 7.5$ Hz, 1H), 5.32 – 5.24 (m, 1H), 3.94 (s, 3H), 3.42 (s, 3H), 2.73 – 2.64 (m, 1H), 2.63 – 2.55 (m, 1H), 1.29 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 174.9, 157.9, 140.3, 138.3, 136.2, 136.1, 133.1, 130.1, 129.9, 128.7, 128.2, 128.2, 127.5, 127.3, 122.2, 121.1, 120.7, 115.2, 111.2, 58.7, 55.5, 53.2, 40.0, 29.9, 23.5. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{28}\text{H}_{25}\text{NNaO}_3^+$ 446.1727; Found 446.1755.

2-(3,5-dimethylbenzoyl)-3*a*,5-dimethyl-1-phenyl-2,3,3*a*,5-tetrahydro-4*H*-

cyclopenta[*c*]quinolin-4-one (**3l**)



Following the general procedure A, **cis-3l** (52.9 mg) as a white solid (mp 68-70 °C) and **trans-3l** (23.0 mg) as a white solid (mp 165-167 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3l**: 90%, *dr*: 2.3:1.

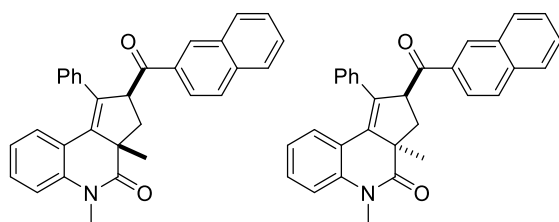
Data of **cis-3l**

¹H NMR (400 MHz, CDCl₃) δ 7.60 (s, 2H), 7.28 – 7.16 (m, 8H), 7.04 (d, *J* = 8.2 Hz, 1H), 6.83 (t, *J* = 7.5 Hz, 1H), 4.77 – 4.70 (m, 1H), 3.42 (s, 3H), 3.20 – 3.10 (m, 1H), 2.36 (s, 6H), 2.25 – 2.18 (m, 1H), 1.28 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.3, 175.4, 139.9, 138.3, 137.8, 137.1, 136.8, 136.3, 134.9, 128.8, 128.7, 128.4, 128.3, 127.5, 126.5, 122.5, 121.1, 114.9, 57.4, 54.4, 39.1, 29.9, 24.9, 21.2. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₉H₂₇NNaO₂⁺ 444.1934; Found 444.1957.

Data of **trans-3l**

¹H NMR (400 MHz, CDCl₃) δ 7.49 (s, 2H), 7.25 – 7.14 (m, 8H), 7.11 – 7.02 (m, 2H), 6.80 (t, *J* = 7.5 Hz, 1H), 5.24 (t, *J* = 8.9 Hz, 1H), 3.40 (s, 3H), 2.70 – 2.61 (m, 1H), 2.60 – 2.53 (m, 1H), 2.33 (s, 6H), 1.33 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.8, 174.6, 140.2, 138.2, 137.5, 137.0, 136.5, 136.2, 134.8, 128.8, 128.4, 127.9, 127.5, 127.4, 126.2, 122.3, 121.0, 115.2, 54.5, 53.7, 40.1, 29.9, 23.9, 21.2. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₉H₂₇NNaO₂⁺ 444.1934; Found 444.1958.

2-(2-naphthoyl)-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[*c*]quinolin-4-one (**3m**)



Following the general procedure A, **cis-3m** (60.2 mg) as a pale yellow solid (mp 43-45 °C) and **trans-3m** (23.2 mg) as a pale yellow solid (mp 152-154 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3m**: 94%, *dr*: 2.6:1.

Data of **cis-3m**

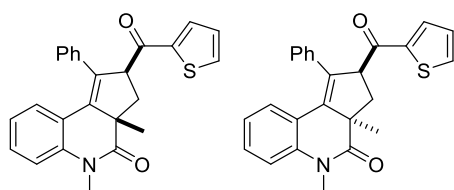
¹H NMR (400 MHz, CDCl₃) δ 8.48 (s, 1H), 8.12 – 8.06 (m, 1H), 7.96 – 7.86 (m, 3H), 7.64 – 7.52 (m, 2H), 7.30 – 7.18 (m, 7H), 7.05 (d, *J* = 8.2 Hz, 1H), 6.85 (t, *J* = 7.5 Hz, 1H), 4.98 – 4.91 (m, 1H), 3.44 (s, 3H), 3.30 – 3.19 (m, 1H), 2.35 – 2.27 (m, 1H), 1.33 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.0, 175.4, 139.9, 138.0, 137.0, 136.8, 135.6, 133.5, 132.4, 130.6, 129.6, 128.8, 128.7, 128.6,

128.6, 128.40, 128.36, 127.7, 127.6, 126.8, 124.3, 122.5, 121.1, 114.9, 57.3, 54.5, 39.1, 30.0, 24.9. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₃₁H₂₅NNaO₃⁺ 466.1778; Found 466.1801.

Data of *trans*-**3m**

¹H NMR (400 MHz, CDCl₃) δ 8.48 (s, 1H), 7.99 – 7.90 (m, 2H), 7.85 (t, *J* = 7.5 Hz, 2H), 7.63 – 7.53 (m, 2H), 7.30 – 7.24 (m, 4H), 7.22 – 7.05 (m, 5H), 6.83 (t, *J* = 7.5 Hz, 1H), 5.44 (t, *J* = 8.9 Hz, 1H), 3.44 (s, 3H), 2.81 – 2.72 (m, 1H), 2.70 – 2.63 (m, 1H), 1.38 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.4, 174.6, 140.3, 137.5, 136.7, 136.1, 135.5, 134.4, 132.5, 130.0, 129.6, 128.9, 128.51, 128.46, 128.0, 127.7, 127.5, 126.7, 124.2, 122.3, 121.0, 115.3, 54.6, 53.8, 40.2, 30.0, 23.9. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₃₁H₂₅NNaO₂⁺ 466.1778; Found 466.1802.

3*a*,5-dimethyl-1-phenyl-2-(thiophene-2-carbonyl)-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3n**)



Following the general procedure A, *cis*-**3n** (45.5 mg) as a white solid (mp 142-144 °C) and *trans*-**3n** (16.8 mg) as a white solid (mp 154-156 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 7:1). Total isolated yield of **3n**: 78%, *dr*: 2.7:1.

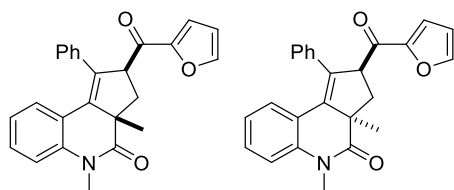
Data of *cis*-**3n**

¹H NMR (400 MHz, CDCl₃) δ 7.70 – 7.63 (m, 2H), 7.27 – 7.15 (m, 7H), 7.13 – 7.09 (m, 1H), 7.04 (d, *J* = 8.2 Hz, 1H), 6.82 (t, *J* = 7.5 Hz, 1H), 4.66 – 4.60 (m, 1H), 3.43 (s, 3H), 3.18 – 3.08 (m, 1H), 2.33 – 2.24 (m, 1H), 1.36 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 194.8, 175.4, 144.0, 139.9, 138.2, 136.5, 136.5, 134.3, 132.7, 128.8, 128.6, 128.4, 128.3, 127.6, 122.5, 121.0, 114.9, 58.5, 54.5, 39.1, 29.9, 25.1. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₂₅H₂₁NNaO₂S⁺ 422.1185; Found 422.1209.

Data of *trans*-**3n**

¹H NMR (400 MHz, CDCl₃) δ 7.78 – 7.73 (m, 1H), 7.60 – 7.55 (m, 1H), 7.29 – 7.17 (m, 7H), 7.12 – 7.04 (m, 3H), 6.82 (t, *J* = 7.5 Hz, 1H), 5.06 – 4.98 (m, 1H), 3.43 (s, 3H), 2.86 – 2.75 (m, 1H), 2.63 – 2.54 (m, 1H), 1.33 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 193.1, 174.5, 143.7, 140.3, 137.0, 137.0, 135.7, 133.9, 132.3, 129.0, 128.5, 128.0, 128.0, 127.6, 127.5, 122.3, 120.8, 115.3, 56.5, 53.7, 40.0, 29.9, 23.7. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₂₅H₂₁NNaO₂S⁺ 422.1185; Found 422.1215.

2-(furan-2-carbonyl)-3*a*,5-dimethyl-1-phenyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3o**)



Following the general procedure B, **cis-3o** (45.5 mg) as a pale yellow solid (mp 138-140 °C) and **trans-3o** (16.8 mg) as a white solid (mp 175-177 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 7:1). Total isolated yield of **3o**: 78%, *dr*: 2.7:1.

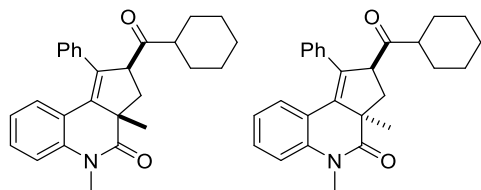
Data of **cis-3o**

¹H NMR (400 MHz, CDCl₃) δ 7.70 – 7.63 (m, 2H), 7.27 – 7.15 (m, 7H), 7.13 – 7.09 (m, 1H), 7.04 (d, *J* = 8.2 Hz, 1H), 6.82 (t, *J* = 7.5 Hz, 1H), 4.66 – 4.60 (m, 1H), 3.43 (s, 3H), 3.18 – 3.08 (m, 1H), 2.33 – 2.24 (m, 1H), 1.36 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 194.8, 175.4, 144.0, 139.9, 138.2, 136.5, 136.5, 134.3, 132.7, 128.8, 128.6, 128.4, 128.3, 127.6, 122.5, 121.0, 114.9, 58.5, 54.5, 39.1, 29.9, 25.1. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₅H₂₁NNaO₂S⁺ 422.1185; Found 422.1209.

Data of **trans-3o**

¹H NMR (400 MHz, CDCl₃) δ 7.78 – 7.73 (m, 1H), 7.60 – 7.55 (m, 1H), 7.29 – 7.17 (m, 7H), 7.12 – 7.04 (m, 3H), 6.82 (t, *J* = 7.5 Hz, 1H), 5.06 – 4.98 (m, 1H), 3.43 (s, 3H), 2.86 – 2.75 (m, 1H), 2.63 – 2.54 (m, 1H), 1.33 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 193.1, 174.5, 143.7, 140.3, 137.0, 137.0, 135.7, 133.9, 132.3, 129.0, 128.5, 128.0, 128.0, 127.6, 127.5, 122.3, 120.8, 115.3, 56.5, 53.7, 40.0, 29.9, 23.7. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₅H₂₁NNaO₂S⁺ 422.1185; Found 422.1215.

2-(cyclohexanecarbonyl)-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[*c*]quinolin-4-one (**3p**)



Following the general procedure A, **cis-3p** (43.5 mg) as a white solid (mp 134-136 °C) and **trans-3p** (12.4 mg) as a white solid (mp 146-148 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 9:1). Total isolated yield of **3p**: 70%, *dr*: 3.5:1.

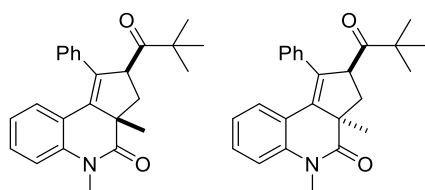
Data of **cis-3p**

¹H NMR (400 MHz, CDCl₃) δ 7.29 – 7.19 (m, 4H), 7.16 – 7.08 (m, 3H), 7.02 (d, *J* = 8.2 Hz, 1H), 6.83 – 6.76 (m, 1H), 4.13 – 4.06 (m, 1H), 3.41 (s, 3H), 2.98 – 2.89 (m, 1H), 2.50 – 2.41 (m, 1H), 2.11 – 2.02 (m, 1H), 1.89 – 1.76 (m, 4H), 1.71 – 1.63 (m, 1H), 1.49 – 1.36 (m, 1H), 1.35 – 1.17 (m, 7H). ¹³C NMR (100 MHz, CDCl₃) δ 215.6, 175.5, 139.8, 137.5, 137.3, 136.9, 128.7, 128.38, 128.36, 128.2, 127.5, 122.4, 121.0, 114.8, 60.2, 54.2, 51.4, 37.5, 29.9, 28.4, 28.4, 25.7, 25.59, 25.56, 25.1. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₉NNaO₂⁺ 422.2091; Found 422.2092.

Data of **trans-3p**

¹H NMR (400 MHz, CDCl₃) δ 7.31 – 7.21 (m, 6H), 7.08 – 7.03 (m, 2H), 6.83 – 6.77 (m, 1H), 4.48 – 4.40 (m, 1H), 3.43 (s, 3H), 2.58 – 2.50 (m, 1H), 2.44 – 2.37 (m, 1H), 2.32 (d, *J* = 11.9 Hz, 1H), 1.70 – 1.65 (m, 1H), 1.60 (d, *J* = 9.3 Hz, 3H), 1.26 (s, 3H), 1.21 – 1.01 (m, 6H). ¹³C NMR (100 MHz, CDCl₃) δ 214.3, 174.7, 140.2, 137.2, 136.7, 136.0, 128.9, 128.6, 128.0, 127.7, 127.5, 122.3, 120.7, 115.2, 58.9, 53.4, 49.7, 38.7, 29.9, 29.0, 28.0, 25.7, 25.5, 23.3. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₉NNaO₂⁺ 422.2091; Found 422.2092.

3a,5-dimethyl-1-phenyl-2-pivaloyl-2,3,3a,5-tetrahydro-4H-cyclopenta[*c*]quinolin-4-one (**3q**)



Following the general procedure A, *cis*-**3q** (40.2 mg) as a white solid (mp 192-194 °C) and *trans*-**3q** (9.8 mg) as a pale yellow solid (mp 103-105 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 10:1). Total isolated yield of **3q**: 67%, *dr*: 4.1:1.

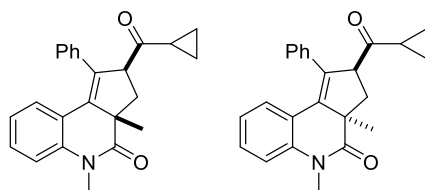
Data of *cis*-**3q**

¹H NMR (400 MHz, CDCl₃) δ 7.26 – 7.15 (m, 4H), 7.09 – 7.04 (m, 2H), 7.03 – 6.96 (m, 2H), 6.75 (t, *J* = 7.5 Hz, 1H), 4.38 – 4.30 (m, 1H), 3.38 (s, 3H), 3.01 – 2.90 (m, 1H), 1.92 – 1.86 (m, 1H), 1.34 (s, 3H), 1.06 (s, 9H). ¹³C NMR (100 MHz, CDCl₃) δ 217.5, 175.5, 139.6, 138.3, 137.8, 136.9, 128.5, 128.4, 128.3, 127.8, 127.5, 122.4, 121.1, 114.7, 56.5, 54.5, 44.6, 38.4, 29.9, 26.0, 25.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₅H₂₇NNaO₂⁺ 396.1934; Found 396.1933.

Data of *trans*-**3q**

¹H NMR (400 MHz, CDCl₃) δ 7.28 – 7.20 (m, 4H), 7.20 – 7.14 (m, 2H), 7.05 – 7.00 (m, 1H), 6.92 – 6.87 (m, 1H), 6.76 (t, *J* = 7.3 Hz, 1H), 4.75 (t, *J* = 8.5 Hz, 1H), 3.40 (s, 3H), 2.44 (d, *J* = 8.5 Hz, 2H), 1.29 (s, 3H), 1.03 (s, 9H). ¹³C NMR (100 MHz, CDCl₃) δ 215.6, 174.6, 140.1, 138.2, 137.0, 136.5, 128.6, 128.5, 128.4, 128.2, 127.4, 122.2, 120.8, 115.1, 53.7, 53.6, 44.4, 41.1, 29.9, 25.9, 24.2. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₅H₂₇NNaO₂⁺ 396.1934; Found 396.1926.

2-(cyclopropanecarbonyl)-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[*c*]quinolin-4-one (**3r**)



Following the general procedure A, *cis*-**3r** (49.2 mg) as a white solid (mp 104-106 °C) and *trans*-**3r** (6.6 mg) as a white solid (mp 119-121 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 10:1). Total isolated yield of **3r**: 78%, *dr*: 7.4:1.

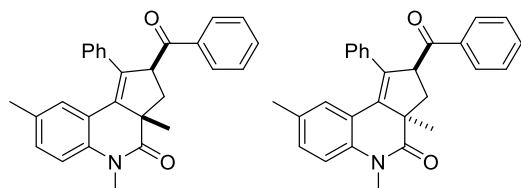
Data of *cis*-**3r**

¹H NMR (400 MHz, CDCl₃) δ 7.30 – 7.20 (m, 6H), 7.18 – 7.14 (m, 1H), 7.03 (d, *J* = 8.2 Hz, 1H), 6.81 (t, *J* = 7.5 Hz, 1H), 4.12 – 4.05 (m, 1H), 3.41 (s, 3H), 3.06 – 2.96 (m, 1H), 2.35 – 2.26 (m, 1H), 2.06 – 1.97 (m, 1H), 1.29 (s, 3H), 1.19 – 1.07 (m, 2H), 1.04 – 0.95 (m, 1H), 0.94 – 0.85 (m, 1H). ¹³C NMR (100 MHz, CDCl₃) δ 212.5, 175.4, 139.9, 137.5, 136.9, 136.7, 128.8, 128.6, 128.3, 128.1, 127.6, 122.4, 121.0, 114.9, 62.7, 54.1, 37.7, 29.9, 24.7, 20.8, 12.0, 11.6. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₄H₂₃NNaO₂⁺ 380.1621; Found 380.1621.

Data of *trans*-3r

^1H NMR (400 MHz, CDCl_3) δ 7.31 – 7.23 (m, 6H), 7.21 – 7.16 (m, 1H), 7.08 (d, $J = 8.2$ Hz, 1H), 6.85 (t, $J = 7.5$ Hz, 1H), 4.39 – 4.31 (m, 1H), 3.45 (s, 3H), 2.82 – 2.71 (m, 1H), 2.50 – 2.40 (m, 1H), 1.89 – 1.80 (m, 1H), 1.28 (s, 3H), 0.90 – 0.83 (m, 1H), 0.75 – 0.63 (m, 1H), 0.50 – 0.35 (m, 2H). ^{13}C NMR (100 MHz, CDCl_3) δ 211.0, 174.8, 140.2, 137.0, 136.8, 135.5, 129.1, 128.5, 128.1, 127.8, 127.6, 122.4, 120.7, 115.3, 61.1, 53.7, 37.8, 29.9, 23.2, 18.4, 11.5, 11.0. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{25}\text{H}_{27}\text{NNaO}_2^+$ 380.1621; Found 380.1615.

2-benzoyl-3a,5,8-trimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3s)



Following the general procedure A, *cis*-3s (52.8 mg) as a pale yellow solid (mp 64-66 °C) and *trans*-3s (18.9 mg) as a pale yellow solid (mp 50-52 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of 3s: 88%, *dr*: 2.8:1.

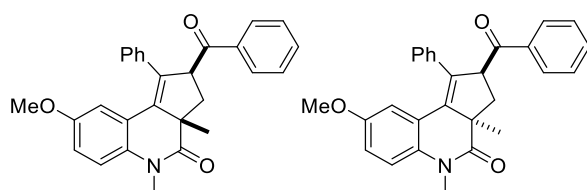
Data of *cis*-3s

^1H NMR (400 MHz, CDCl_3) δ 8.04 – 7.98 (m, 2H), 7.61 – 7.55 (m, 1H), 7.47 (t, $J = 7.6$ Hz, 2H), 7.29 – 7.18 (m, 5H), 7.09 – 7.02 (m, 2H), 6.93 (d, $J = 8.3$ Hz, 1H), 4.81 – 4.73 (m, 1H), 3.40 (s, 3H), 3.20 – 3.09 (m, 1H), 2.25 – 2.18 (m, 1H), 2.09 (s, 3H), 1.28 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 202.0, 175.1, 138.1, 137.6, 136.7, 136.4, 136.2, 133.2, 131.9, 129.4, 128.8, 128.7, 128.64, 128.56, 128.2, 127.5, 120.9, 114.8, 57.1, 54.4, 38.9, 29.8, 24.8, 20.4. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{28}\text{H}_{25}\text{NNaO}_2^+$ 430.1778; Found 430.1777.

Data of *trans*-3s

^1H NMR (400 MHz, CDCl_3) δ 7.91 (d, $J = 7.7$ Hz, 2H), 7.53 (t, $J = 7.3$ Hz, 1H), 7.42 (t, $J = 7.6$ Hz, 2H), 7.29 – 7.15 (m, 6H), 7.09 – 7.02 (m, 1H), 6.98 – 6.88 (m, 2H), 5.27 (t, $J = 8.9$ Hz, 1H), 3.39 (s, 3H), 2.72 – 2.62 (m, 1H), 2.63 – 2.53 (m, 1H), 2.07 (s, 3H), 1.33 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 200.5, 174.4, 137.9, 137.0, 136.9, 136.8, 136.1, 133.0, 131.7, 129.5, 128.5, 128.4, 128.3, 127.93, 127.89, 127.4, 120.7, 115.1, 54.6, 53.7, 40.0, 29.9, 23.8, 20.4. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{28}\text{H}_{25}\text{NNaO}_2^+$ 430.1778; Found 430.1774.

2-benzoyl-8-methoxy-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3t)



Following the general procedure A, *cis*-3t (50.8 mg) as a white solid (mp 189-191 °C) and *trans*-3t (20.3 mg) as a pale yellow solid (mp 100-102 °C) were obtained after flash chromatography

(Petroleum ether/Acetone = 6:1). Total isolated yield of **3t**: 84%, *dr*: 2.5:1.

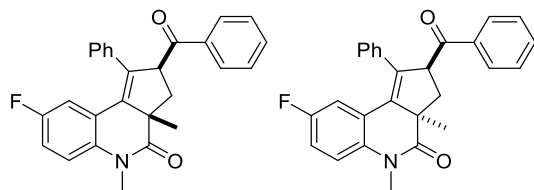
Data of *cis*-**3t**

¹H NMR (400 MHz, CDCl₃) δ 8.04 – 7.97 (m, 2H), 7.58 (t, *J* = 7.4 Hz, 1H), 7.47 (t, *J* = 7.6 Hz, 2H), 7.31 – 7.20 (m, 5H), 6.96 (d, *J* = 8.9 Hz, 1H), 6.83 – 6.73 (m, 2H), 4.83 – 4.74 (m, 1H), 3.44 (s, 3H), 3.40 (s, 3H), 3.21 – 3.11 (m, 1H), 2.28 – 2.20 (m, 1H), 1.31 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 201.9, 174.8, 154.6, 138.4, 137.2, 136.7, 136.2, 133.5, 133.2, 128.72, 128.66, 128.6, 128.3, 127.6, 121.7, 116.0, 115.5, 112.3, 57.2, 55.1, 54.3, 38.9, 29.9, 25.0. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₃⁺ 446.1727; Found 446.1730.

Data of *trans*-**3t**

¹H NMR (400 MHz, CDCl₃) δ 7.94 – 7.88 (m, 2H), 7.53 (t, *J* = 7.4 Hz, 1H), 7.42 (t, *J* = 7.7 Hz, 2H), 7.30 – 7.14 (m, 5H), 6.97 (d, *J* = 9.0 Hz, 1H), 6.82 – 6.78 (m, 1H), 6.61 (d, *J* = 2.9 Hz, 1H), 5.31 – 5.23 (m, 1H), 3.44 (s, 3H), 3.39 (s, 3H), 2.72 – 2.64 (m, 1H), 2.63 – 2.55 (m, 1H), 1.35 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.4, 174.0, 154.6, 137.7, 137.0, 136.9, 136.1, 133.9, 133.0, 128.5, 128.41, 128.39, 128.0, 127.5, 121.7, 116.3, 115.3, 111.7, 55.0, 54.6, 53.5, 40.0, 30.0, 23.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₃⁺ 446.1727; Found 446.1727.

2-benzoyl-8-fluoro-3*a*,5-dimethyl-1-phenyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3u**)



Following the general procedure A, *cis*-**3u** (49.1 mg) as a white solid (mp 62-64 °C) and *trans*-**3u** (17.6 mg) as a pale yellow solid (mp 135-137 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3u**: 81%, *dr*: 2.8:1.

Data of *cis*-**3u**

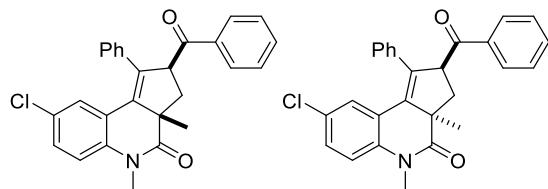
¹H NMR (400 MHz, CDCl₃) δ 7.99 (d, *J* = 8.2 Hz, 2H), 7.62 – 7.55 (m, 1H), 7.52 – 7.44 (m, 2H), 7.24 (s, 5H), 7.01 – 6.89 (m, 3H), 4.83 – 4.73 (m, 1H), 3.41 (s, 3H), 3.21 – 3.10 (m, 1H), 2.28 – 2.19 (m, 1H), 1.30 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 201.8, 174.9, 158.1 (d, *J* = 242.5 Hz), 138.7, 137.1 (d, *J* = 2.1 Hz), 136.3 (d, *J* = 2.3 Hz), 136.21, 136.18, 133.4, 128.8, 128.60, 128.58, 128.0, 122.7 (d, *J* = 8.3 Hz), 116.2 (d, *J* = 8.3 Hz), 115.4 (d, *J* = 23.0 Hz), 114.9 (d, *J* = 23.9 Hz), 57.4, 54.2, 39.0, 30.2, 25.0. ¹⁹F NMR (376 MHz, CDCl₃) δ -120.3. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₂FNNaO₂⁺ 434.1527; Found 434.1526.

Data of *trans*-**3u**

¹H NMR (400 MHz, CDCl₃) δ 7.90 (d, *J* = 7.7 Hz, 2H), 7.54 (t, *J* = 7.4 Hz, 1H), 7.42 (t, *J* = 7.6 Hz, 2H), 7.23 (d, *J* = 4.3 Hz, 5H), 7.04 – 6.92 (m, 2H), 6.80 – 6.74 (m, 1H), 5.28 (t, *J* = 8.9 Hz, 1H), 3.40 (s, 3H), 2.73 – 2.64 (m, 1H), 2.63 – 2.54 (m, 1H), 1.34 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.2, 174.1, 157.9 (d, *J* = 242.1 Hz), 138.9, 136.8, 136.6 (d, *J* = 2.5 Hz), 135.7 (d, *J* = 2.1 Hz), 135.4, 133.1, 128.6, 128.6, 128.4, 127.9, 127.8, 122.4 (d, *J* = 8.3 Hz), 116.4 (d, *J* = 8.3 Hz), 115.4

(d, $J = 22.9$ Hz), 113.9 (d, $J = 23.8$ Hz), 54.5, 53.4, 39.9, 30.2, 23.8. HRMS (ESI) m/z : $[M+Na]^+$ Calcd for $C_{27}H_{22}FNNaO_2^+$ 434.1527; Found 434.1524.

2-benzoyl-8-chloro-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3v)



Following the general procedure A, *cis*-**3v** (45.2 mg) as a white solid (mp 127-129 °C) and *trans*-**3v** (18.1 mg) as a white solid (mp 125-127 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3v**: 74%, *dr*: 2.5:1.

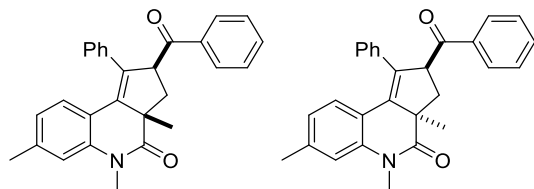
Data of *cis*-**3v**

1H NMR (400 MHz, $CDCl_3$) δ 7.99 (d, $J = 7.7$ Hz, 2H), 7.59 (t, $J = 7.3$ Hz, 1H), 7.48 (t, $J = 7.6$ Hz, 2H), 7.22 (s, 5H), 7.15 (d, $J = 8.3$ Hz, 1H), 7.04 (d, $J = 1.9$ Hz, 1H), 6.85 – 6.79 (m, 1H), 4.81 – 4.73 (m, 1H), 3.41 (s, 3H), 3.19 – 3.08 (m, 1H), 2.27 – 2.18 (m, 1H), 1.29 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.8, 175.2, 141.0, 137.7, 136.8, 136.4, 136.1, 134.4, 133.4, 129.3, 128.7, 128.54, 128.46, 127.7, 122.5, 119.4, 115.3, 57.2, 54.3, 38.9, 30.0, 24.9. HRMS (ESI) m/z : $[M+Na]^+$ Calcd for $C_{27}H_{22}ClNNaO_2^+$ 450.1231; Found 450.1228.

Data of *trans*-**3v**

1H NMR (400 MHz, $CDCl_3$) δ 7.92 – 7.85 (m, 2H), 7.56 – 7.50 (m, 1H), 7.46 – 7.39 (m, 2H), 7.24 – 7.14 (m, 5H), 7.05 (d, $J = 2.0$ Hz, 1H), 7.00 (d, $J = 8.3$ Hz, 1H), 6.82 – 6.77 (m, 1H), 5.30 – 5.23 (m, 1H), 3.40 (s, 3H), 2.73 – 2.64 (m, 1H), 2.62 – 2.54 (m, 1H), 1.33 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 200.3, 174.4, 141.3, 138.1, 136.8, 135.7, 135.5, 134.4, 133.2, 128.60, 128.55, 128.41, 128.39, 127.8, 127.7, 122.3, 119.3, 115.6, 54.4, 53.5, 39.9, 30.0, 23.9. HRMS (ESI) m/z : $[M+Na]^+$ Calcd for $C_{27}H_{22}ClNNaO_2^+$ 450.1231; Found 450.1228.

2-benzoyl-3a,5,7-trimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3w)



Following the general procedure A, *cis*-**3w** (54.4 mg) as a pale yellow solid (mp 47-49 °C) and *trans*-**3w** (18.1 mg) as a white solid (mp 187-189 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3w**: 89%, *dr*: 3.0:1.

Data of *cis*-**3w**

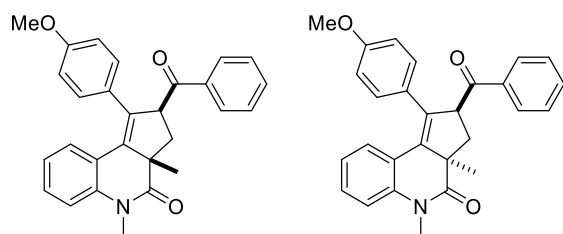
1H NMR (400 MHz, $CDCl_3$) δ 8.03 – 7.97 (m, 2H), 7.60 – 7.54 (m, 1H), 7.47 (t, $J = 7.6$ Hz, 2H), 7.26 – 7.17 (m, 5H), 7.13 (d, $J = 7.8$ Hz, 1H), 6.86 (s, 1H), 6.66 (d, $J = 7.8$ Hz, 1H), 4.79 – 4.71 (m,

1H), 3.42 (s, 3H), 3.19 – 3.07 (m, 1H), 2.34 (s, 3H), 2.27 – 2.17 (m, 1H), 1.29 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.1, 175.4, 139.8, 138.9, 138.0, 136.9, 136.3, 135.8, 133.2, 128.7, 128.6, 128.3, 128.1, 127.4, 123.3, 118.3, 115.7, 57.2, 54.4, 38.9, 29.9, 24.9, 21.7. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₂⁺ 430.1778; Found 430.1790.

Data of *trans*-**3w**

¹H NMR (400 MHz, CDCl₃) δ 7.91 (d, *J* = 7.4 Hz, 2H), 7.52 (t, *J* = 7.4 Hz, 1H), 7.41 (t, *J* = 7.6 Hz, 2H), 7.27 – 7.15 (m, 5H), 6.99 (d, *J* = 7.8 Hz, 1H), 6.87 (s, 1H), 6.64 (d, *J* = 7.8 Hz, 1H), 5.32 – 5.19 (m, 1H), 3.41 (s, 3H), 2.71 – 2.62 (m, 1H), 2.62 – 2.54 (m, 1H), 2.35 (s, 3H), 1.33 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.6, 174.7, 140.2, 139.0, 137.0, 136.7, 136.3, 136.2, 133.0, 128.5, 128.4, 127.9, 127.3, 127.3, 123.1, 118.1, 116.0, 54.6, 53.7, 40.0, 29.9, 23.8, 21.8. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₂⁺ 430.1778; Found 430.1774.

2-benzoyl-1-(4-methoxyphenyl)-3a,5-dimethyl-2,3,3a,5-tetrahydro-4H-cyclopenta[*c*]quinolin-4-one (**3x**)



Following the general procedure A, *cis*-**3x** (47.2 mg) as a pale yellow solid (mp 37-39 °C) and *trans*-**3x** (16.3 mg) as a yellow solid (mp 73-75 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 7:1). Total isolated yield of **3x**: 75%, *dr*: 2.9:1.

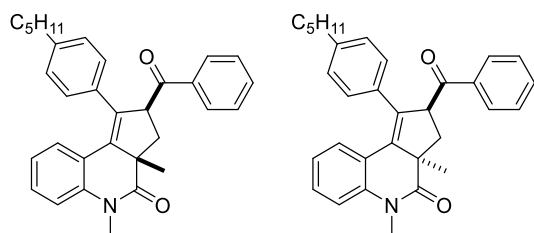
Data of *cis*-**3x**

¹H NMR (400 MHz, CDCl₃) δ 8.04 – 7.98 (m, 2H), 7.61 – 7.55 (m, 1H), 7.48 (t, *J* = 7.6 Hz, 2H), 7.34 – 7.29 (m, 1H), 7.27 – 7.22 (m, 1H), 7.17 (d, *J* = 8.8 Hz, 2H), 7.07 – 7.02 (m, 1H), 6.90 – 6.84 (m, 1H), 6.74 (d, *J* = 8.8 Hz, 2H), 4.77 – 4.69 (m, 1H), 3.75 (s, 3H), 3.42 (s, 3H), 3.21 – 3.08 (m, 1H), 2.26 – 2.15 (m, 1H), 1.27 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.1, 175.3, 158.9, 139.8, 136.9, 136.4, 136.2, 133.2, 129.8, 128.8, 128.7, 128.64, 128.61, 128.3, 122.4, 121.3, 114.9, 113.7, 57.2, 55.1, 54.2, 38.9, 29.9, 24.7. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₃⁺ 446.1727; Found 446.1733.

Data of *trans*-**3x**

¹H NMR (400 MHz, CDCl₃) δ 7.91 (d, *J* = 7.4 Hz, 2H), 7.52 (t, *J* = 7.4 Hz, 1H), 7.41 (t, *J* = 7.6 Hz, 2H), 7.27 – 7.15 (m, 5H), 6.99 (d, *J* = 7.8 Hz, 1H), 6.87 (s, 1H), 6.64 (d, *J* = 7.8 Hz, 1H), 5.32 – 5.19 (m, 1H), 3.41 (s, 3H), 2.71 – 2.62 (m, 1H), 2.62 – 2.54 (m, 1H), 2.35 (s, 3H), 1.33 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.6, 174.7, 140.2, 139.0, 137.0, 136.7, 136.3, 136.2, 133.0, 128.5, 128.4, 127.9, 127.3, 127.3, 123.1, 118.1, 116.0, 54.6, 53.7, 40.0, 29.9, 23.8, 21.8. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₃⁺ 446.1727; Found 446.1737.

2-benzoyl-3a,5-dimethyl-1-(4-pentylphenyl)-2,3,3a,5-tetrahydro-4H-cyclopenta[*c*]quinolin-4-one (**3y**)



Following the general procedure A, **cis-3y** (64.9 mg) as a white solid (mp 35-37 °C) and **trans-3y** (23.2 mg) as a pale yellow solid (mp 96-98 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 7:1). Total isolated yield of **3y**: 95%, *dr*: 2.8:1.

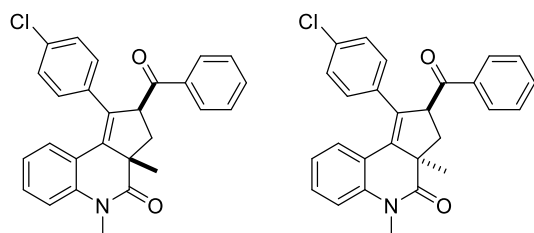
Data of **cis-3y**

¹H NMR (400 MHz, CDCl₃) δ 8.00 (d, *J* = 7.6 Hz, 2H), 7.57 (t, *J* = 7.3 Hz, 1H), 7.47 (t, *J* = 7.6 Hz, 2H), 7.31 (d, *J* = 7.7 Hz, 1H), 7.24 (t, *J* = 7.8 Hz, 1H), 7.15 (d, *J* = 7.8 Hz, 2H), 7.03 (t, *J* = 8.6 Hz, 3H), 6.85 (t, *J* = 7.5 Hz, 1H), 4.76 (d, *J* = 11.1 Hz, 1H), 3.42 (s, 3H), 3.21 – 3.08 (m, 1H), 2.53 (t, *J* = 7.8 Hz, 2H), 2.26 – 2.15 (m, 1H), 1.64 – 1.51 (m, 2H), 1.28 (s, 7H), 0.87 (t, *J* = 6.7 Hz, 4H). ¹³C NMR (100 MHz, CDCl₃) δ 202.0, 175.3, 142.3, 139.8, 137.2, 136.8, 136.2, 133.7, 133.2, 128.7, 128.64, 128.61, 128.4, 128.35, 128.29, 122.4, 121.2, 114.9, 57.1, 54.3, 38.8, 35.5, 31.4, 30.9, 29.9, 24.7, 22.4, 14.0. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₃₂H₃₃NNaO₂⁺ 486.2404; Found 486.2408.

Data of **trans-3y**

¹H NMR (400 MHz, CDCl₃) δ 7.90 (d, *J* = 7.7 Hz, 2H), 7.52 (t, *J* = 7.5 Hz, 1H), 7.41 (t, *J* = 7.6 Hz, 2H), 7.29 – 7.22 (m, 1H), 7.18 – 7.11 (m, 3H), 7.05 (d, *J* = 8.3 Hz, 1H), 7.00 (d, *J* = 7.8 Hz, 2H), 6.83 (t, *J* = 7.5 Hz, 1H), 5.25 (t, *J* = 8.9 Hz, 1H), 3.42 (s, 3H), 2.73 – 2.64 (m, 1H), 2.60 – 2.47 (m, 3H), 1.60 – 1.51 (m, 2H), 1.32 (s, 3H), 1.31 – 1.24 (m, 4H), 0.87 (t, *J* = 6.9 Hz, 4H). ¹³C NMR (100 MHz, CDCl₃) δ 200.6, 174.6, 142.2, 140.1, 137.4, 136.9, 135.9, 133.1, 133.0, 128.7, 128.5, 128.4, 127.7, 127.4, 122.2, 121.1, 115.2, 54.6, 53.6, 39.9, 35.6, 31.5, 30.7, 29.9, 23.8, 22.5, 14.0. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₃₂H₃₃NNaO₂⁺ 486.2404; Found 486.2403.

2-benzoyl-1-(4-chlorophenyl)-3a,5-dimethyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (**3z**)



Following the general procedure A, **cis-3z** (60.3 mg) as a pale yellow solid (mp 47-49 °C) and **trans-3z** (20.1 mg) as a white solid (mp 137-139 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3z**: 94%, *dr*: 3.0:1.

Data of **cis-3z**

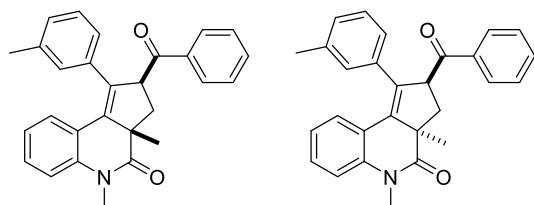
¹H NMR (400 MHz, CDCl₃) δ 8.04 – 7.96 (m, 2H), 7.63 – 7.56 (m, 1H), 7.49 (t, *J* = 7.7 Hz, 2H), 7.30 – 7.15 (m, 6H), 7.05 (d, *J* = 8.2 Hz, 1H), 6.87 (t, *J* = 7.3 Hz, 1H), 4.78 – 4.67 (m, 1H), 3.42 (s,

3H), 3.23 – 3.10 (m, 1H), 2.28 – 2.17 (m, 1H), 1.27 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 201.8, 175.0, 139.9, 138.7, 135.9, 135.4, 135.2, 133.4, 133.3, 130.0, 129.0, 128.70, 128.66, 128.5, 128.2, 122.6, 120.6, 115.0, 57.1, 54.3, 38.9, 29.9, 24.8. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₂₇H₂₂CINNaO₂⁺ 450.1231; Found 450.1236.

Data of *trans*-**3z**

¹H NMR (400 MHz, CDCl₃) δ 7.95 – 7.87 (m, 2H), 7.60 – 7.51 (m, 1H), 7.44 (t, *J* = 7.7 Hz, 2H), 7.32 – 7.24 (m, 1H), 7.18 (s, 4H), 7.12 – 7.04 (m, 2H), 6.85 (t, *J* = 7.5 Hz, 1H), 5.24 (t, *J* = 8.9 Hz, 1H), 3.41 (s, 3H), 2.72 – 2.55 (m, 2H), 1.33 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.2, 174.4, 140.2, 137.5, 136.7, 135.8, 134.6, 133.3, 133.2, 129.3, 129.2, 128.73, 128.66, 128.4, 127.4, 122.4, 120.6, 115.4, 54.4, 53.7, 40.0, 29.9, 23.8. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₂₇H₂₂CINNaO₂⁺ 450.1231; Found 450.1233.

2-benzoyl-3*a*,5-dimethyl-1-(*m*-tolyl)-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3aa**)



Following the general procedure A, *cis*-**3aa** (53.9 mg) as a pale yellow solid (mp 121-123 °C) and *trans*-**3aa** (18.6 mg) as a yellow liquid were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3aa**: 89%, *dr*: 2.9:1.

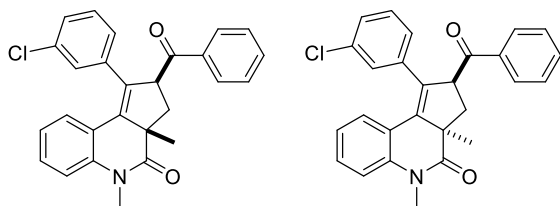
Data of *cis*-**3aa**

¹H NMR (400 MHz, CDCl₃) δ 8.02 – 7.95 (m, 2H), 7.60 – 7.54 (m, 1H), 7.51 – 7.44 (m, 2H), 7.28 – 7.21 (m, 2H), 7.14 – 6.99 (m, 5H), 6.87 – 6.80 (m, 1H), 4.80 – 4.70 (m, 1H), 3.43 (s, 3H), 3.19 – 3.09 (m, 1H), 2.28 – 2.15 (m, 4H), 1.30 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.0, 175.4, 139.8, 137.9, 137.7, 137.1, 136.7, 136.4, 133.2, 129.1, 128.72, 128.68, 128.6, 128.4, 128.3, 128.2, 125.8, 122.5, 121.1, 114.9, 57.3, 54.4, 38.9, 29.9, 24.9, 21.3. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₂⁺ 430.1778; Found 430.1786.

Data of *trans*-**3aa**

¹H NMR (400 MHz, CDCl₃) δ 7.94 – 7.86 (m, 2H), 7.56 – 7.49 (m, 1H), 7.42 (t, *J* = 7.6 Hz, 2H), 7.28 – 7.22 (m, 1H), 7.15 – 7.01 (m, 5H), 6.97 (d, *J* = 7.4 Hz, 1H), 6.85 – 6.78 (m, 1H), 5.30 – 5.21 (m, 1H), 3.41 (s, 3H), 2.74 – 2.64 (m, 1H), 2.61 – 2.52 (m, 1H), 2.19 (s, 3H), 1.33 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.5, 174.6, 140.2, 137.9, 137.5, 137.0, 136.3, 135.9, 133.0, 128.8, 128.51, 128.48, 128.4, 128.3, 128.2, 127.5, 125.0, 122.2, 121.0, 115.2, 54.5, 53.6, 39.9, 29.9, 23.7, 21.3. HRMS (ESI) m/z: [M+Na]⁺ Calcd for C₂₈H₂₅NNaO₂⁺ 430.1778; Found 430.1782.

2-benzoyl-1-(3-chlorophenyl)-3*a*,5-dimethyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3ab**)



Following the general procedure A, **cis-3ab** (61.6 mg) as a white solid (mp 127-129 °C) and **trans-3ab** (20.6 mg) as a white solid (mp 155-157 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3ab**: 96%, *dr*: 3.0:1.

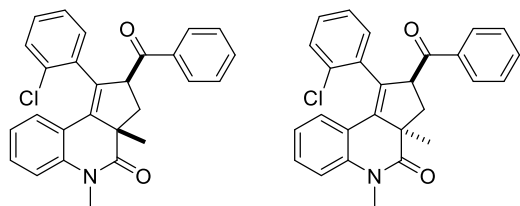
Data of **cis-3ab**

¹H NMR (400 MHz, CDCl₃) δ 8.00 (d, *J* = 7.8 Hz, 2H), 7.66 – 7.40 (m, 3H), 7.32 – 7.10 (m, 6H), 7.06 (d, *J* = 8.2 Hz, 1H), 6.87 (t, *J* = 7.6 Hz, 1H), 4.73 (d, *J* = 11.3 Hz, 1H), 3.43 (s, 3H), 3.22 – 3.06 (m, 1H), 2.24 (d, *J* = 13.9 Hz, 1H), 1.28 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 201.7, 175.0, 139.9, 139.2, 138.7, 136.0, 135.2, 134.1, 133.4, 129.6, 129.2, 128.73, 128.70, 128.6, 128.3, 127.7, 127.1, 122.6, 120.5, 115.1, 57.2, 54.4, 39.0, 29.9, 24.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₃ClNNaO₂⁺ 450.1231; Found 450.1239.

Data of **trans-3ab**

¹H NMR (400 MHz, CDCl₃) δ 7.91 (d, *J* = 7.7 Hz, 2H), 7.56 (t, *J* = 7.4 Hz, 1H), 7.44 (t, *J* = 7.6 Hz, 2H), 7.32 – 7.21 (m, 3H), 7.19 – 7.10 (m, 3H), 7.10 – 7.02 (m, 2H), 6.85 (t, *J* = 7.6 Hz, 1H), 5.24 (t, *J* = 8.9 Hz, 1H), 3.42 (s, 3H), 2.72 – 2.54 (m, 2H), 1.34 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.0, 174.3, 140.2, 138.1, 138.0, 136.7, 135.5, 134.2, 133.3, 129.8, 129.3, 128.6, 128.4, 127.8, 127.6, 127.4, 126.3, 122.4, 120.4, 115.4, 54.4, 53.7, 39.9, 29.9, 23.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₃ClNNaO₂⁺ 450.1231; Found 450.1240.

2-benzoyl-1-(2-chlorophenyl)-3a,5-dimethyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (**3ac**)



Following the general procedure A, **cis-3ac** (50.0 mg) as a white solid (mp 133-135 °C) and **trans-3ac** (19.3 mg) as a white solid (mp 138-140 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3ac**: 81%, *dr*: 2.6:1.

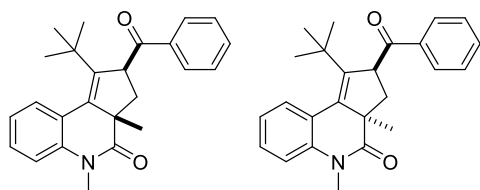
Data of **cis-3ac**

¹H NMR (400 MHz, CDCl₃) δ 7.87 (d, *J* = 7.7 Hz, 2H), 7.62 – 7.54 (m, 1H), 7.53 – 7.47 (m, 1H), 7.37 (t, *J* = 7.7 Hz, 2H), 7.28 – 7.14 (m, 4H), 7.07 – 6.98 (m, 2H), 6.81 – 6.74 (m, 1H), 4.94 – 4.87 (m, 1H), 3.42 (s, 3H), 3.27 – 3.17 (m, 1H), 2.44 – 2.30 (m, 1H), 1.41 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 201.0, 175.2, 140.0, 139.3, 136.2, 135.3, 134.1, 133.1, 132.3, 129.4, 128.94, 128.91, 128.5, 128.4, 126.8, 126.4, 122.5, 120.7, 114.8, 56.3, 54.2, 38.1, 29.9, 25.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₃ClNNaO₂⁺ 450.1231; Found 450.1234.

Data of *trans*-**3ac**

¹H NMR (400 MHz, CDCl₃) δ 7.91 – 7.84 (m, 2H), 7.52 – 7.46 (m, 1H), 7.41 – 7.34 (m, 2H), 7.34 – 7.29 (m, 1H), 7.25 – 7.20 (m, 1H), 7.18 – 7.06 (m, 3H), 7.03 (d, *J* = 8.2 Hz, 1H), 6.79 – 6.73 (m, 1H), 6.68 (d, *J* = 7.8 Hz, 1H), 5.47 (t, *J* = 11.7 Hz, 1H), 3.41 (s, 3H), 2.83 – 2.72 (m, 1H), 2.61 – 2.50 (m, 1H), 1.42 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.5, 174.4, 140.0, 139.3, 136.9, 135.4, 135.0, 133.0, 132.8, 132.1, 129.2, 129.0, 128.9, 128.4, 128.3, 127.2, 127.1, 122.5, 120.2, 115.0, 60.3, 53.1, 39.6, 29.8, 24.3. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₃CINNaO₂⁺ 450.1231; Found 450.1236.

2-benzoyl-1-(tert-butyl)-3*a*,5-dimethyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3ad**)



Following the general procedure A, *cis*-**3ad** (35.4 mg) as a pale yellow solid (mp 129-131 °C) and *trans*-**3ad** (15.4 mg) as a white solid (mp 181-183 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 10:1). Total isolated yield of **3ad**: 68%, *dr*: 2.3:1.

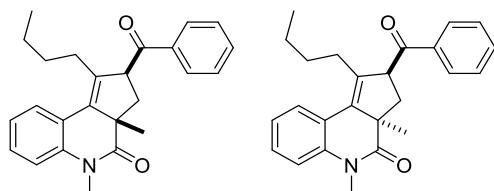
Data of *cis*-**3ad**

¹H NMR (400 MHz, CDCl₃) δ 8.06 – 8.00 (m, 2H), 7.76 – 7.71 (m, 1H), 7.64 – 7.58 (m, 1H), 7.55 – 7.49 (m, 2H), 7.35 – 7.29 (m, 1H), 7.15 – 7.09 (m, 1H), 7.03 (d, *J* = 8.2 Hz, 1H), 4.93 – 4.86 (m, 1H), 3.38 (s, 3H), 3.12 – 3.02 (m, 1H), 1.86 – 1.79 (m, 1H), 1.15 (s, 9H), 1.08 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.6, 176.4, 146.6, 139.5, 136.3, 135.9, 133.2, 130.5, 128.8, 128.5, 128.3, 124.4, 122.2, 114.6, 55.6, 54.6, 37.7, 34.1, 31.5, 30.1, 24.1. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₅H₂₇NNaO₂⁺ 396.1934; Found 396.1948.

Data of *trans*-**3ad**

¹H NMR (400 MHz, CDCl₃) δ 8.01 – 7.95 (m, 2H), 7.64 – 7.59 (m, 1H), 7.58 – 7.53 (m, 1H), 7.50 – 7.43 (m, 2H), 7.33 – 7.27 (m, 1H), 7.12 – 7.06 (m, 1H), 7.04 (d, *J* = 8.1 Hz, 1H), 4.82 – 4.75 (m, 1H), 3.38 (s, 3H), 2.82 – 2.76 (m, 1H), 2.10 – 2.01 (m, 1H), 1.13 (s, 9H), 0.99 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.3, 174.4, 147.3, 139.4, 136.2, 135.2, 132.8, 128.9, 128.6, 128.5, 127.6, 126.0, 121.7, 114.7, 55.0, 54.1, 36.2, 33.9, 31.0, 30.3, 24.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₅H₂₇NNaO₂⁺ 396.1934; Found 396.1940.

2-benzoyl-1-butyl-3*a*,5-dimethyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3ae**)



Following the general procedure A, *cis*-**3ae** (45.0 mg) as a yellow liquid and *trans*-**3ae** (14.0 mg)

as a colorless liquid were obtained after flash chromatography (Petroleum ether/Acetone = 10:1). Total isolated yield of **3ae**: 79%, *dr*: 3.2:1.

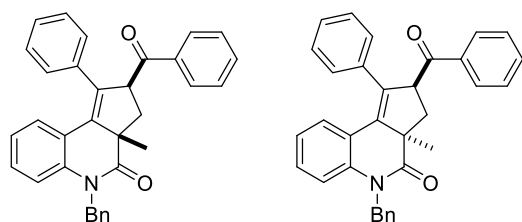
Data of *cis*-**3ae**

¹H NMR (400 MHz, CDCl₃) δ 8.06 – 7.99 (m, 2H), 7.65 – 7.48 (m, 4H), 7.36 – 7.29 (m, 1H), 7.14 (t, *J* = 7.5 Hz, 1H), 7.07 (d, *J* = 8.2 Hz, 1H), 4.69 – 4.61 (m, 1H), 3.39 (s, 3H), 3.15 – 3.05 (m, 1H), 2.67 – 2.56 (m, 1H), 2.40 – 2.31 (m, 1H), 2.11 – 2.02 (m, 1H), 1.42 – 1.31 (m, 1H), 1.30 – 1.13 (m, 6H), 0.77 (t, *J* = 7.0 Hz, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.0, 175.7, 139.6, 139.2, 136.3, 135.1, 133.3, 128.7, 128.6, 128.1, 127.8, 122.7, 122.1, 114.8, 53.9, 53.8, 38.1, 29.93, 29.90, 28.1, 25.2, 22.7, 13.7. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₅H₂₇NNaO₂⁺ 396.1934; Found 396.1943.

Data of *trans*-**3ae**

¹H NMR (400 MHz, CDCl₃) δ 8.02 (d, *J* = 7.1 Hz, 2H), 7.63 – 7.56 (m, 1H), 7.53 – 7.41 (m, 3H), 7.32 (t, *J* = 7.8 Hz, 1H), 7.15 – 7.04 (m, 2H), 4.81 (t, *J* = 8.8 Hz, 1H), 3.37 (s, 3H), 2.66 – 2.55 (m, 2H), 2.47 – 2.37 (m, 1H), 2.21 – 2.11 (m, 1H), 1.59 – 1.27 (m, 5H), 1.22 (s, 3H), 0.88 (t, *J* = 7.1 Hz, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 201.3, 175.1, 140.1, 139.8, 137.0, 135.2, 133.2, 128.7, 128.5, 128.3, 126.8, 122.6, 121.9, 115.1, 53.1, 52.8, 39.5, 30.6, 29.9, 27.2, 24.2, 22.9, 13.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₅H₂₇NNaO₂⁺ 396.1934; Found 396.1937.

2-benzoyl-5-benzyl-3a-methyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[*c*]quinolin-4-one (**3af**)



Following the general procedure A, *cis*-**3af** (54.4 mg) as a yellow solid (mp 47-49 °C) and *trans*-**3af** (23.6 mg) as a yellow solid (mp 59-61 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 10:1). Total isolated yield of **3af**: 83%, *dr*: 2.3:1.

Data of *cis*-**3af**

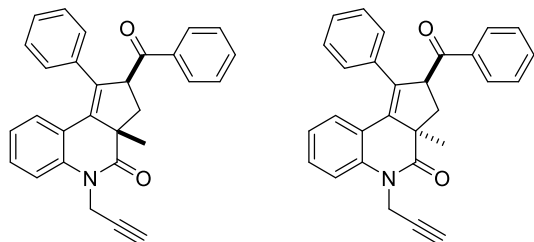
¹H NMR (400 MHz, CDCl₃) δ 8.02 (d, *J* = 7.3 Hz, 2H), 7.58 (t, *J* = 7.4 Hz, 1H), 7.48 (t, *J* = 7.6 Hz, 2H), 7.36 – 7.17 (m, 12H), 7.11 – 7.05 (m, 1H), 6.91 (d, *J* = 8.2 Hz, 1H), 6.78 (t, *J* = 7.5 Hz, 1H), 5.65 (d, *J* = 16.3 Hz, 1H), 4.87 – 4.75 (m, 2H), 3.31 – 3.19 (m, 1H), 2.31 – 2.21 (m, 1H), 1.44 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.0, 175.4, 139.3, 137.8, 137.1, 136.7, 136.2, 133.3, 128.79, 128.77, 128.71, 128.69, 128.6, 128.44, 128.36, 127.6, 127.1, 126.0, 122.6, 121.2, 115.8, 57.2, 54.5, 46.5, 38.8, 25.0. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₃₃H₂₇NNaO₂⁺ 492.1934; Found 492.1938.

Data of *trans*-**3af**

¹H NMR (400 MHz, CDCl₃) δ 7.93 (d, *J* = 7.5 Hz, 2H), 7.54 (t, *J* = 7.4 Hz, 1H), 7.43 (t, *J* = 7.6 Hz, 2H), 7.34 (t, *J* = 7.5 Hz, 2H), 7.29 – 7.17 (m, 8H), 7.09 (t, *J* = 7.3 Hz, 2H), 6.93 (d, *J* = 8.5 Hz, 1H), 6.77 (t, *J* = 7.5 Hz, 1H), 5.64 (d, *J* = 16.4 Hz, 1H), 5.30 (t, *J* = 8.9 Hz, 1H), 4.79 (d, *J* = 16.3 Hz, 1H), 2.86 – 2.75 (m, 1H), 2.68 – 2.59 (m, 1H), 1.48 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.4,

174.7, 139.6, 137.6, 137.1, 136.9, 136.5, 136.0, 133.1, 128.9, 128.8, 128.6, 128.5, 128.4, 128.0, 127.5, 127.1, 126.1, 122.4, 121.1, 116.1, 54.8, 53.8, 46.6, 39.7, 24.0. HRMS (ESI) m/z : $[M+Na]^+$ Calcd for $C_{33}H_{27}NNaO_2^+$ 492.1934; Found 492.1930.

2-benzoyl-3a-methyl-1-phenyl-5-(prop-2-yn-1-yl)-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3ag)



Following the general procedure A, **cis-3ag** (51.2 mg) as a white solid (mp 42-44 °C) and **trans-3af** (18.9 mg) as a white solid (mp 90-92 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 10:1). Total isolated yield of **3ag**: 84%, *dr*: 2.7:1.

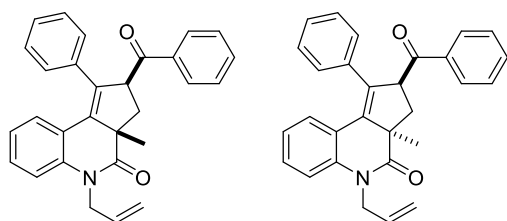
Data of **cis-3ag**

1H NMR (400 MHz, $CDCl_3$) δ 8.02 – 7.96 (m, 2H), 7.58 (t, $J = 7.4$ Hz, 1H), 7.51 – 7.44 (m, 2H), 7.28 – 7.17 (m, 8H), 6.90 – 6.82 (m, 1H), 5.02 (dd, $J = 17.6, 2.5$ Hz, 1H), 4.78 (dd, $J = 11.3, 2.0$ Hz, 1H), 4.52 (dd, $J = 17.6, 2.5$ Hz, 1H), 3.23 – 3.11 (m, 1H), 2.26 – 2.18 (m, 2H), 1.32 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 201.9, 174.5, 138.2, 137.4, 137.2, 136.6, 136.2, 133.3, 128.8, 128.7, 128.6, 128.5, 128.3, 127.6, 122.9, 121.1, 115.3, 78.6, 71.9, 57.1, 54.3, 38.6, 32.1, 24.6. HRMS (ESI) m/z : $[M+Na]^+$ Calcd for $C_{29}H_{23}NNaO_2^+$ 440.1621; Found 440.1632.

Data of **trans-3ag**

1H NMR (400 MHz, $CDCl_3$) δ 7.90 (dd, $J = 8.4, 1.4$ Hz, 2H), 7.53 (t, $J = 7.4$ Hz, 1H), 7.42 (t, $J = 7.7$ Hz, 2H), 7.31 – 7.15 (m, 7H), 7.11 (d, $J = 7.7$ Hz, 1H), 6.89 – 6.80 (m, 1H), 5.28 (dd, $J = 9.6, 8.2$ Hz, 1H), 5.07 (dd, $J = 17.6, 2.5$ Hz, 1H), 4.44 (dd, $J = 17.6, 2.5$ Hz, 1H), 2.76 – 2.66 (m, 1H), 2.64 – 2.54 (m, 1H), 1.36 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 200.3, 173.7, 138.6, 137.7, 136.9, 136.1, 135.9, 133.1, 128.9, 128.6, 128.5, 128.4, 127.9, 127.63, 127.56, 122.7, 121.0, 115.6, 78.7, 71.9, 54.6, 53.6, 39.7, 32.3, 23.6. HRMS (ESI) m/z : $[M+Na]^+$ Calcd for $C_{33}H_{27}NNaO_2^+$ 440.1621; Found 440.1628.

5-allyl-2-benzoyl-3a-methyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (3ah)



Following the general procedure A, **cis-3ah** (50.7 mg) as a yellow solid (mp 49-51 °C) and **trans-3ah** (23.1 mg) as a white solid (mp 105-107 °C) were obtained after flash chromatography

(Petroleum ether/Acetone = 10:1). Total isolated yield of **3ah**: 83%, *dr*: 2.2:1.

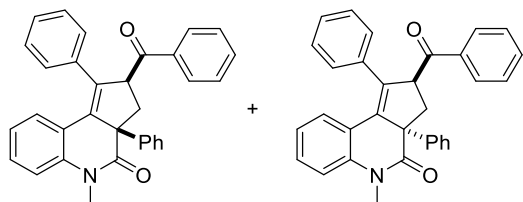
Data of *cis*-**3ah**

¹H NMR (400 MHz, CDCl₃) δ 8.00 (d, *J* = 7.1 Hz, 2H), 7.58 (t, *J* = 7.3 Hz, 1H), 7.48 (t, *J* = 7.6 Hz, 2H), 7.29 – 7.16 (m, 7H), 7.02 (d, *J* = 8.3 Hz, 1H), 6.82 (t, *J* = 7.5 Hz, 1H), 5.99 – 5.86 (m, 1H), 5.18 (t, *J* = 14.3 Hz, 2H), 5.01 – 4.91 (m, 1H), 4.79 (dd, *J* = 11.4, 2.0 Hz, 1H), 4.32 – 4.22 (m, 1H), 3.19 (dd, *J* = 14.0, 11.3 Hz, 1H), 2.21 (dd, *J* = 13.9, 2.1 Hz, 1H), 1.35 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 202.0, 174.9, 139.2, 137.9, 136.9, 136.7, 136.2, 133.3, 132.4, 128.74, 128.70, 128.67, 128.6, 128.4, 128.3, 127.5, 122.5, 121.1, 115.9, 115.5, 57.2, 54.4, 45.1, 38.7, 24.9. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₉H₂₅NNaO₂⁺ 442.1778; Found 442.1789.

Data of *trans*-**3ah**

¹H NMR (400 MHz, CDCl₃) δ 7.94 – 7.87 (m, 2H), 7.53 (t, *J* = 7.4 Hz, 1H), 7.45 – 7.38 (m, 2H), 7.28 – 7.16 (m, 6H), 7.10 (dd, *J* = 7.8, 1.6 Hz, 1H), 7.04 (d, *J* = 8.2 Hz, 1H), 6.83 – 6.76 (m, 1H), 6.00 – 5.87 (m, 1H), 5.31 – 5.12 (m, 3H), 5.02 – 4.90 (m, 1H), 4.30 – 4.19 (m, 1H), 2.78 – 2.68 (m, 1H), 2.63 – 2.52 (m, 1H), 1.38 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.4, 174.1, 139.5, 137.4, 136.9, 136.5, 136.0, 133.1, 132.5, 128.8, 128.5, 128.42, 128.39, 128.0, 127.51, 127.48, 122.3, 121.0, 115.94, 115.88, 54.7, 53.7, 45.3, 39.7, 23.8. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₃₃H₂₇NNaO₂⁺ 442.1778; Found 442.1784.

2-benzoyl-5-methyl-1,3*a*-diphenyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (**3aj**)

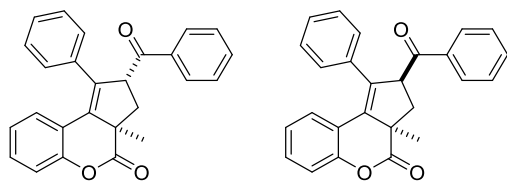


Following the general procedure A, *cis*-**3aj** + *trans*-**3aj** (54.7 mg) as a yellow solid (mp 165-167 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3aj**: 60%, *dr*: 4.6:1.

Data of *cis*-**3aj** + *trans*-**3aj**

¹H NMR (400 MHz, CDCl₃) δ 7.93 (d, *J* = 7.5 Hz, 2H), 7.54 (t, *J* = 7.4 Hz, 1H), 7.43 (t, *J* = 7.6 Hz, 2H), 7.34 (t, *J* = 7.5 Hz, 2H), 7.29 – 7.17 (m, 8H), 7.09 (t, *J* = 7.3 Hz, 2H), 6.93 (d, *J* = 8.5 Hz, 1H), 6.77 (t, *J* = 7.5 Hz, 1H), 5.64 (d, *J* = 16.4 Hz, 1H), 5.30 (t, *J* = 8.9 Hz, 1H), 4.79 (d, *J* = 16.3 Hz, 1H), 2.86 – 2.75 (m, 1H), 2.68 – 2.59 (m, 1H), 1.48 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 200.4, 174.7, 139.6, 137.6, 137.1, 136.9, 136.5, 136.0, 133.1, 128.9, 128.8, 128.6, 128.5, 128.4, 128.0, 127.5, 127.1, 126.1, 122.4, 121.1, 116.1, 54.8, 53.8, 46.6, 39.7, 24.0. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₃₃H₂₇NNaO₂⁺ 492.1934; Found 492.1930.

2-benzoyl-3*a*-methyl-1-phenyl-3,3*a*-dihydrocyclopenta[*c*]chromen-4(2*H*)-one (**3ak**)



Following the general procedure A, **cis-3ak** (32.0 mg) as a yellow solid (mp 38-40 °C) and **trans-3ak** (22.8 mg) as a white solid (mp 156-158 °C) were obtained after flash chromatography (Petroleum ether/Acetone = 8:1). Total isolated yield of **3ak**: 72%, *dr*: 1:1.4.

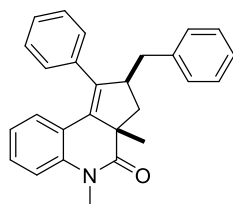
Data of **cis-3ak**

¹H NMR (400 MHz, CDCl₃) δ 7.98 (d, *J* = 8.0 Hz, 2H), 7.64 – 7.56 (m, 1H), 7.53 – 7.44 (m, 2H), 7.26 (d, *J* = 1.6 Hz, 6H), 7.20 (d, *J* = 7.8 Hz, 1H), 7.12 (d, *J* = 8.4 Hz, 1H), 6.92 (t, *J* = 7.6 Hz, 1H), 4.87 – 4.79 (m, 1H), 3.25 – 3.15 (m, 1H), 2.33 – 2.23 (m, 1H), 1.44 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 201.4, 173.1, 151.3, 138.01, 136.97, 136.0, 134.5, 133.5, 129.7, 128.8, 128.7, 128.6, 128.4, 128.0, 127.7, 124.1, 118.6, 116.7, 57.1, 53.4, 38.1, 24.8. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₆H₂₀NNaO₃⁺ 403.1305; Found 403.1313.

Data of **trans-3ak**

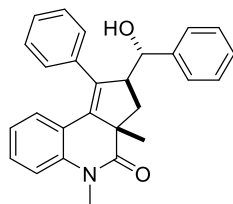
¹H NMR (400 MHz, CDCl₃) δ 7.93 – 7.86 (m, 2H), 7.54 (t, *J* = 7.2 Hz, 1H), 7.47 – 7.38 (m, 2H), 7.31 – 7.19 (m, 6H), 7.17 – 7.06 (m, 2H), 6.91 (t, *J* = 7.6 Hz, 1H), 5.29 (t, *J* = 8.8 Hz, 1H), 2.85 – 2.74 (m, 1H), 2.66 – 2.56 (m, 1H), 1.46 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 199.8, 172.2, 151.5, 139.0, 136.6, 135.2, 133.3, 129.8, 128.64, 128.60, 128.4, 128.0, 127.7, 126.9, 124.0, 118.6, 117.0, 54.4, 52.5, 39.0, 23.8. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₆H₂₀NNaO₃⁺ 403.1305; Found 403.1309.

(2*S*,3*aS*)-2-benzyl-3*a*,5-dimethyl-1-phenyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (4a)



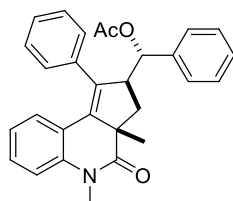
¹H NMR (400 MHz, CDCl₃) δ 7.41 – 7.36 (m, 2H), 7.36 – 7.32 (m, 1H), 7.31 – 7.24 (m, 5H), 7.22 – 7.14 (m, 4H), 7.07 (dd, *J* = 7.7, 1.6 Hz, 1H), 6.99 (d, *J* = 8.2 Hz, 1H), 6.81 – 6.76 (m, 1H), 3.38 (s, 3H), 3.29 – 3.21 (m, 2H), 2.72 – 2.64 (m, 1H), 2.63 – 2.55 (m, 1H), 1.93 (dd, *J* = 14.2, 2.2 Hz, 1H), 1.35 (s, 3H). ¹³C NMR (101 MHz, CDCl₃) δ 175.5, 143.5, 140.5, 139.7, 137.6, 134.8, 128.9, 128.8, 128.7, 128.5, 128.4, 128.3, 127.3, 126.0, 122.3, 121.6, 114.8, 53.4, 52.0, 41.2, 37.4, 30.0, 27.2. HRMS (ESI) *m/z*: [M+Na]⁺ Calcd for C₂₇H₂₅NNaO⁺ 402.1828; Found 402.1819.

(2*S*,3*aS*)-2-((*S*)-hydroxy(phenyl)methyl)-3*a*,5-dimethyl-1-phenyl-2,3,3*a*,5-tetrahydro-4*H*-cyclopenta[*c*]quinolin-4-one (4b)



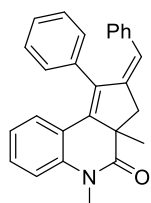
^1H NMR (400 MHz, CDCl_3) δ 7.44 – 7.22 (m, 11H), 7.21 – 7.15 (m, 1H), 6.95 (d, $J = 8.2$ Hz, 1H), 6.80 (t, $J = 7.6$ Hz, 1H), 4.92 (d, $J = 7.2$ Hz, 1H), 3.59 – 3.51 (m, 1H), 3.33 (s, 3H), 2.79 – 2.68 (m, 1H), 2.00 (s, 1H), 1.75 (dd, $J = 14.7, 3.2$ Hz, 1H), 0.97 (s, 3H). ^{13}C NMR (101 MHz, CDCl_3) δ 175.5, 142.5, 141.6, 139.4, 137.8, 137.1, 129.3, 128.4, 128.4, 128.2, 127.7, 127.5, 127.4, 127.0, 122.3, 121.5, 114.7, 78.3, 58.3, 53.4, 35.7, 30.0, 26.0. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{27}\text{H}_{25}\text{NNaO}_2^+$ 418.1778; Found 418.1772.

(S)-((2S,3aS)-3a,5-dimethyl-4-oxo-1-phenyl-3,3a,4,5-tetrahydro-2H-cyclopenta[c]quinolin-2-yl)(phenyl)methyl acetate (4c)



^1H NMR (400 MHz, CDCl_3) δ 7.41 – 7.25 (m, 11H), 7.23 – 7.16 (m, 1H), 6.96 (d, $J = 8.2$ Hz, 1H), 6.81 (t, $J = 7.6$ Hz, 1H), 5.87 (d, $J = 9.3$ Hz, 1H), 3.78 – 3.65 (m, 1H), 3.34 (s, 3H), 2.73 (dd, $J = 14.7, 10.5$ Hz, 1H), 1.68 – 1.62 (m, 1H), 1.52 (s, 3H), 1.13 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 175.1, 170.1, 141.4, 139.4, 139.2, 137.8, 137.2, 129.0, 128.6, 128.4, 128.2, 128.0, 127.6, 127.3, 127.1, 122.4, 121.5, 114.7, 79.4, 54.6, 53.3, 35.6, 30.1, 26.5, 20.8. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{29}\text{H}_{27}\text{NNaO}_3^+$ 460.1883; Found 460.1886.

(Z)-2-benzylidene-3a,5-dimethyl-1-phenyl-2,3,3a,5-tetrahydro-4H-cyclopenta[c]quinolin-4-one (4d)



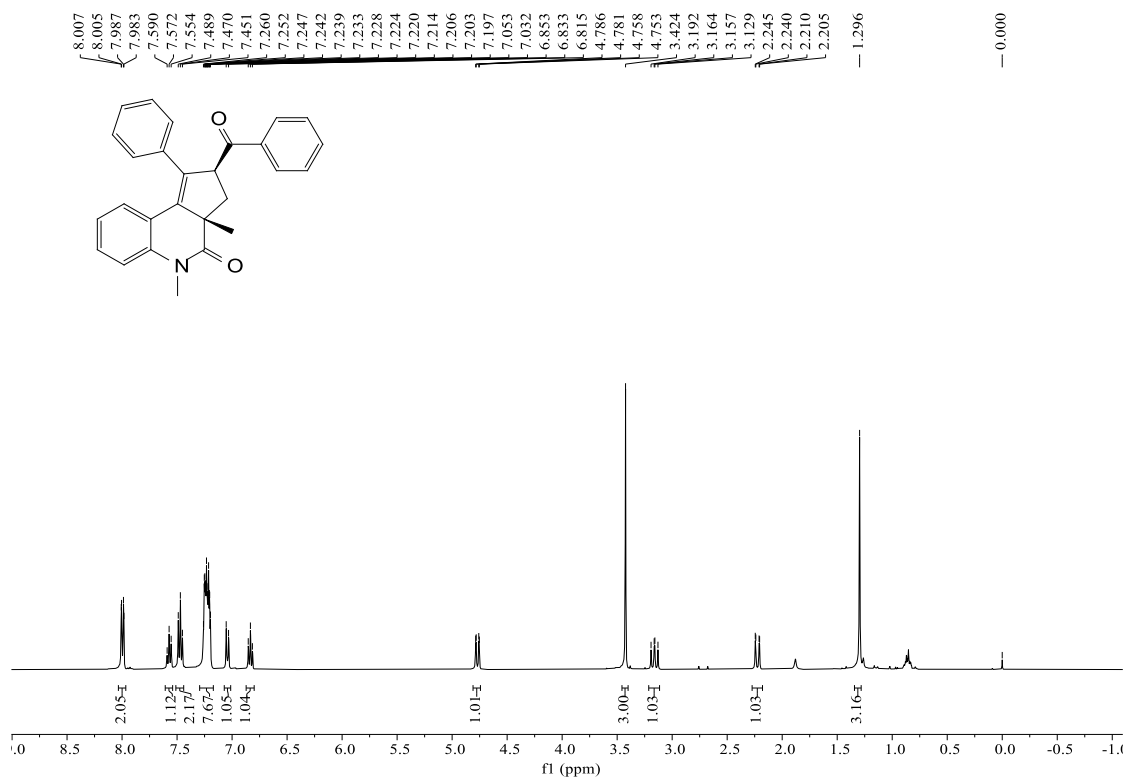
^1H NMR (400 MHz, CDCl_3) δ 7.49 – 7.36 (m, 5H), 7.31 (t, $J = 7.6$ Hz, 2H), 7.27 – 7.14 (m, 4H), 7.02 (d, $J = 8.3$ Hz, 1H), 6.82 (dd, $J = 7.7, 1.6$ Hz, 1H), 6.75 (t, $J = 7.5$ Hz, 1H), 6.11 (s, 1H), 3.76 (dd, $J = 17.3, 2.9$ Hz, 1H), 3.44 (s, 3H), 3.12 (dd, $J = 17.3, 1.9$ Hz, 1H), 1.33 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 174.3, 146.5, 142.3, 140.9, 139.7, 137.7, 134.9, 129.4, 128.8, 128.6, 128.3, 128.3, 127.7, 127.3, 126.4, 123.0, 122.4, 121.3, 115.0, 52.5, 40.7, 30.1, 26.8. HRMS (ESI) m/z : $[\text{M}+\text{Na}]^+$ Calcd for $\text{C}_{27}\text{H}_{23}\text{NNaO}^+$ 400.1672; Found 400.1665.

8. References

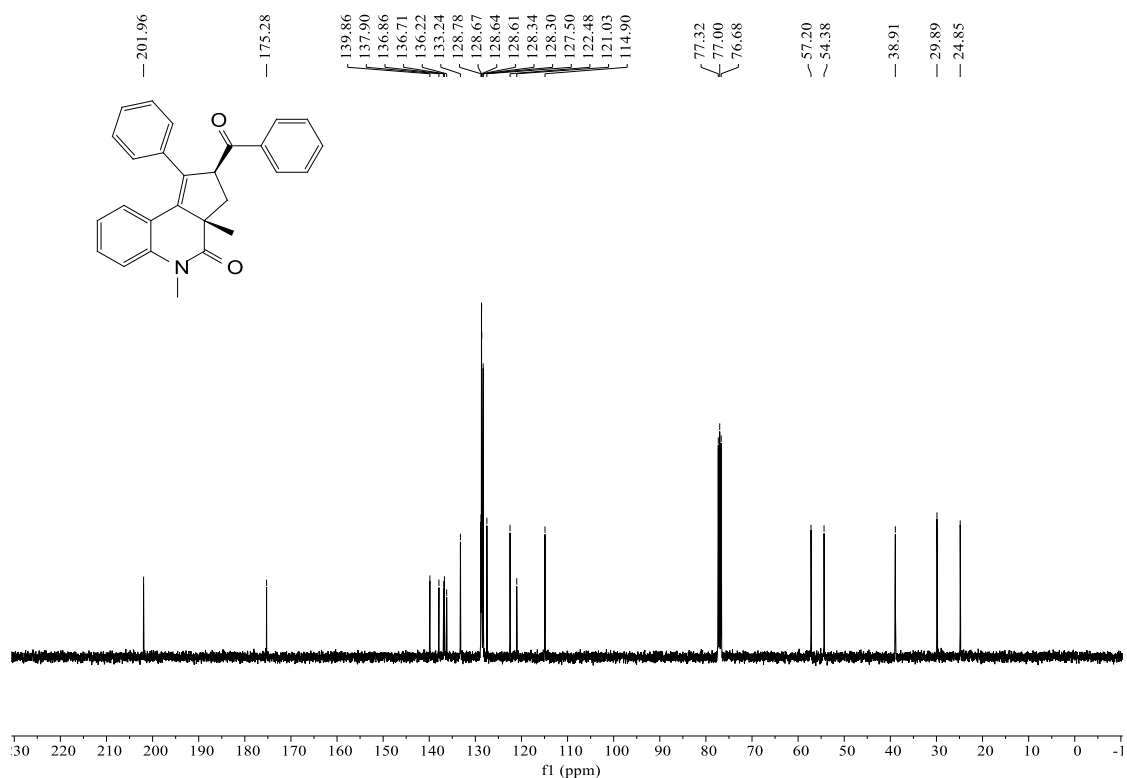
- [1] (a) A. Garay Talero, B. Martins and A. Burtoloso, *Org. Lett.*, 2018, **20**, 7206-7211; (b) S. Zhu, K. Shi, H. Zhu, Z. Jia, X. Xia, D. Wang and L. Zou, *Org. Lett.*, 2020, **22**, 1504-1509.
- [2] (a) M. Hu, J. H. Fan, Y. Liu, X. H. Ouyang, R. J. Song and J. H. Li, *Angew. Chem., Int. Ed.*, 2015, **54**, 9577-9580; (b) C. Wu, J. Liao and S. Ge, *Angew. Chem., Int. Ed.*, 2019, **58**, 8882-8886; (c) Y. Liu, J.-L. Zhang, R.-J. Song, P.-C. Qian and J.-H. Li, *Angew. Chem., Int. Ed.*, 2014, **53**, 9017-9020.
- [3] S. Ghosh, Z.-W. Qu, S. Pradhan, A. Ghosh, S. Grimme and I. Chatterjee, *Angew. Chem., Int. Ed.*, 2022, **61**, e202115272.
- [4] Z. Qu, T. Tian, Y. Tan, X. Ji, G.-J. Deng and Huang, H. *Green Chem.*, 2022, **24**, 7403-7409.

9. Copies of ^1H , ^{13}C and ^{19}F NMR spectra of all products

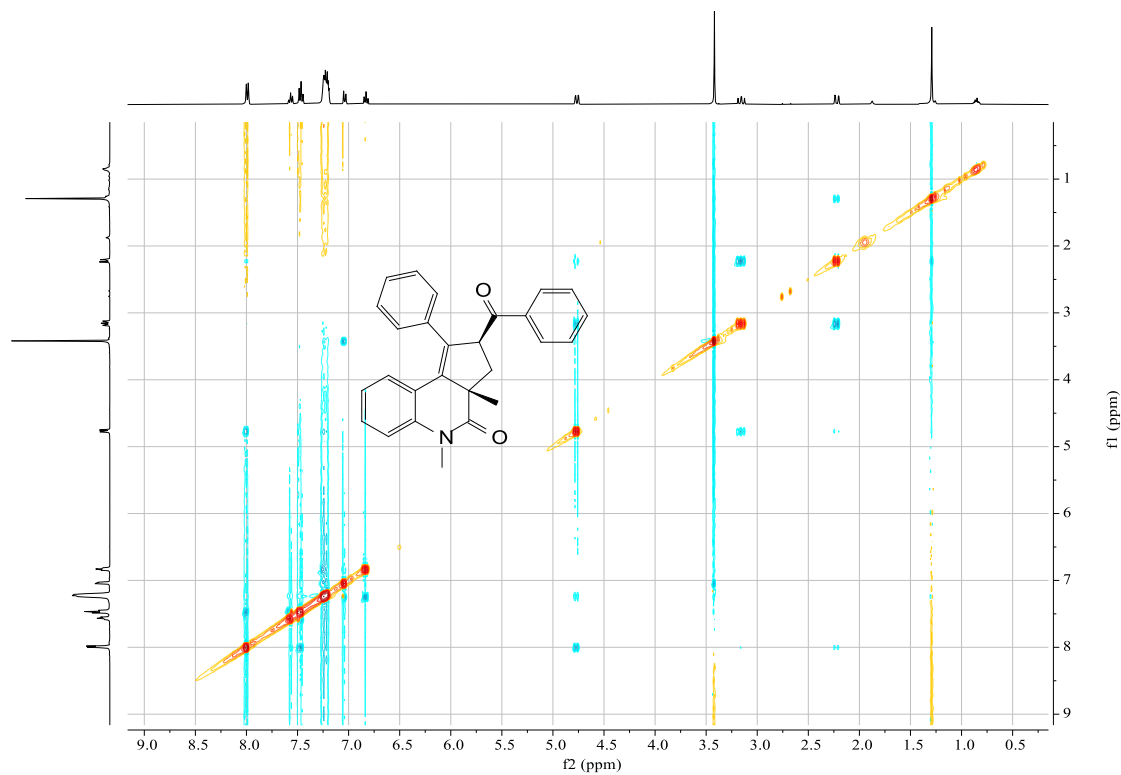
^1H NMR spectra of *cis*-3a (400 MHz, CDCl_3)



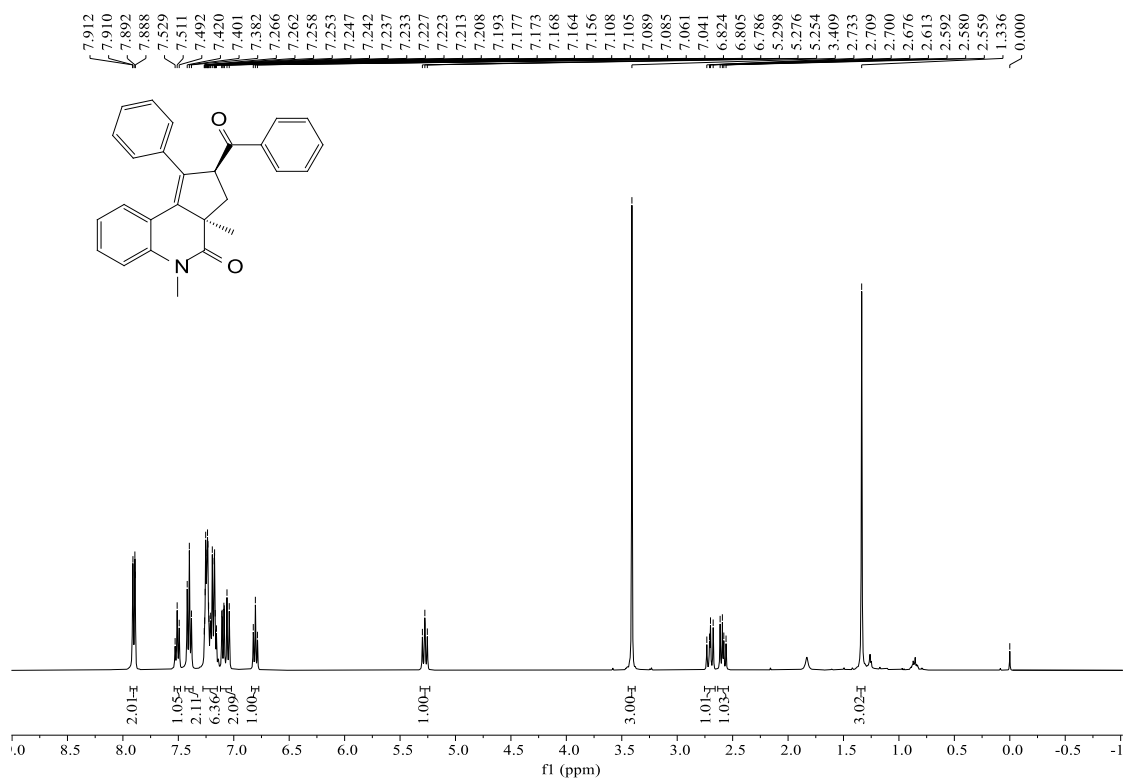
^{13}C NMR spectra of *cis*-3a (100 MHz, CDCl_3)



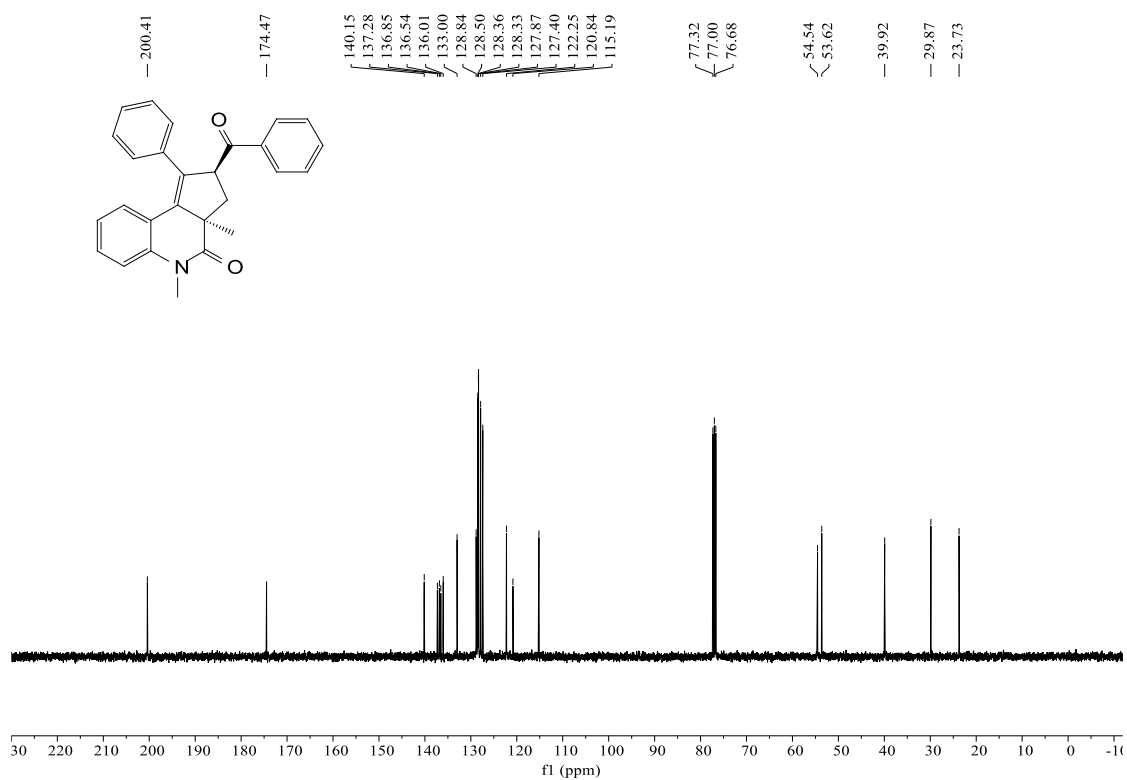
NOE spectra of *cis*-3a



¹H NMR spectra of *trans*-3a (400 MHz, CDCl₃)

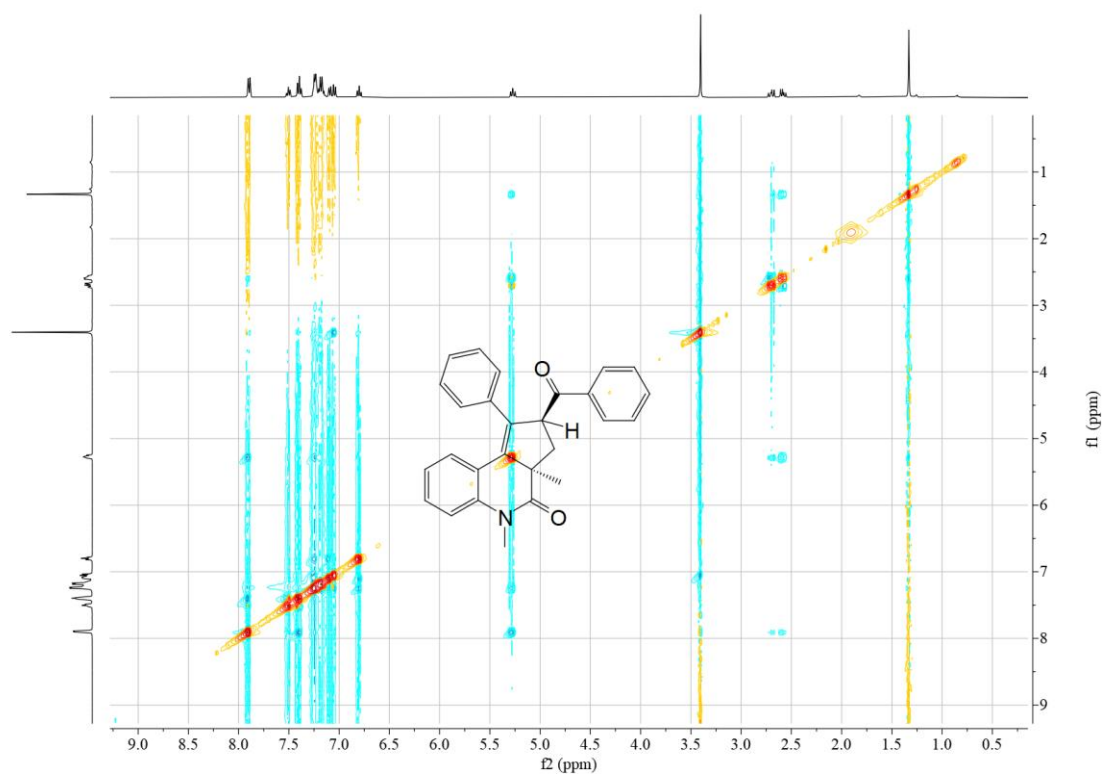


¹³C NMR spectra of *trans*-3a (100 MHz, CDCl₃)

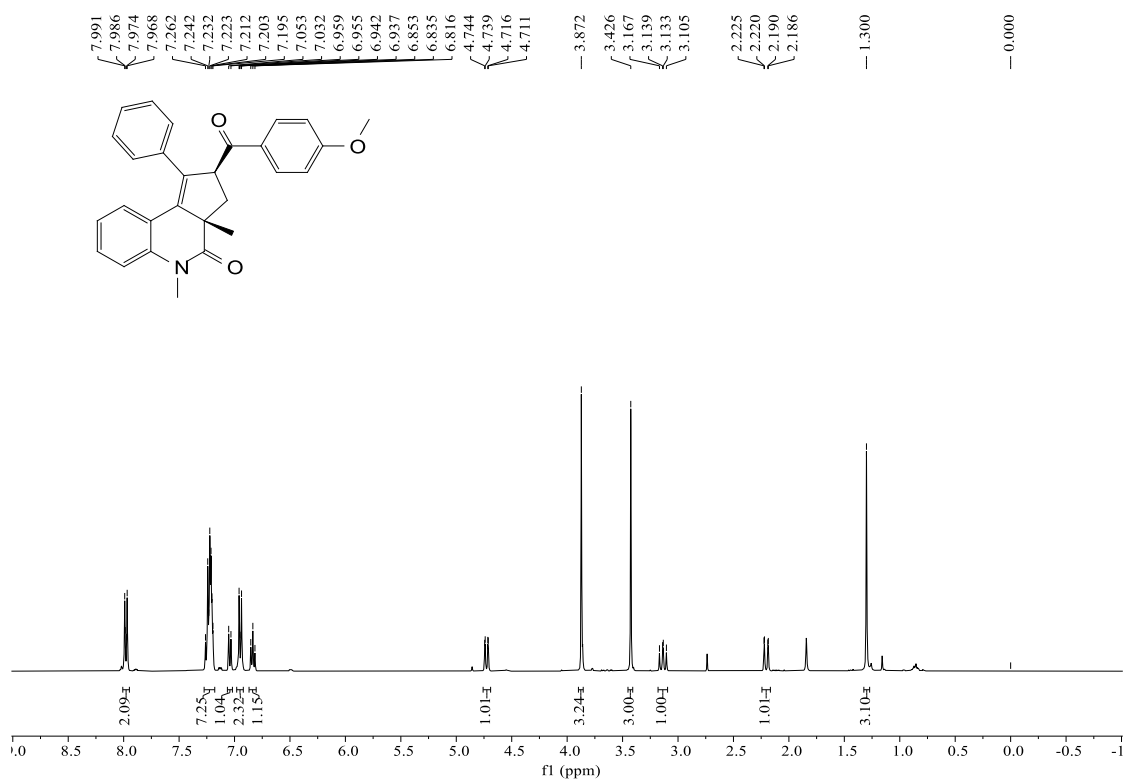


NOE spectra of *cis*-3a

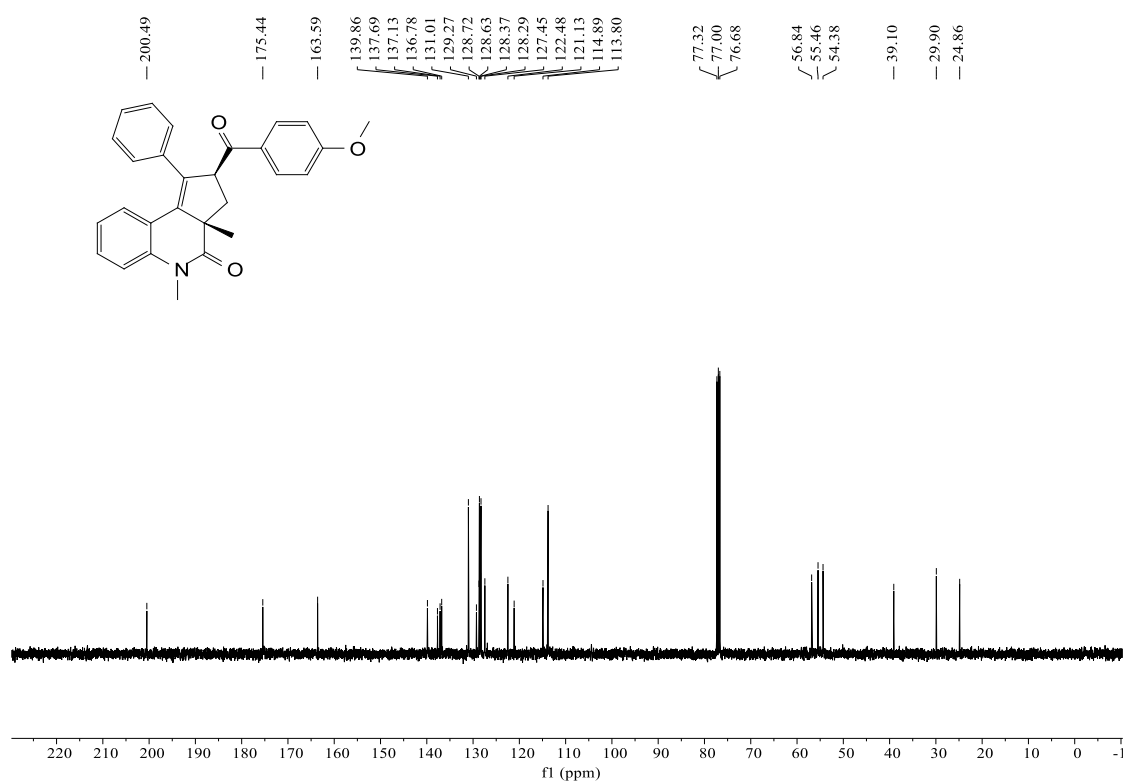




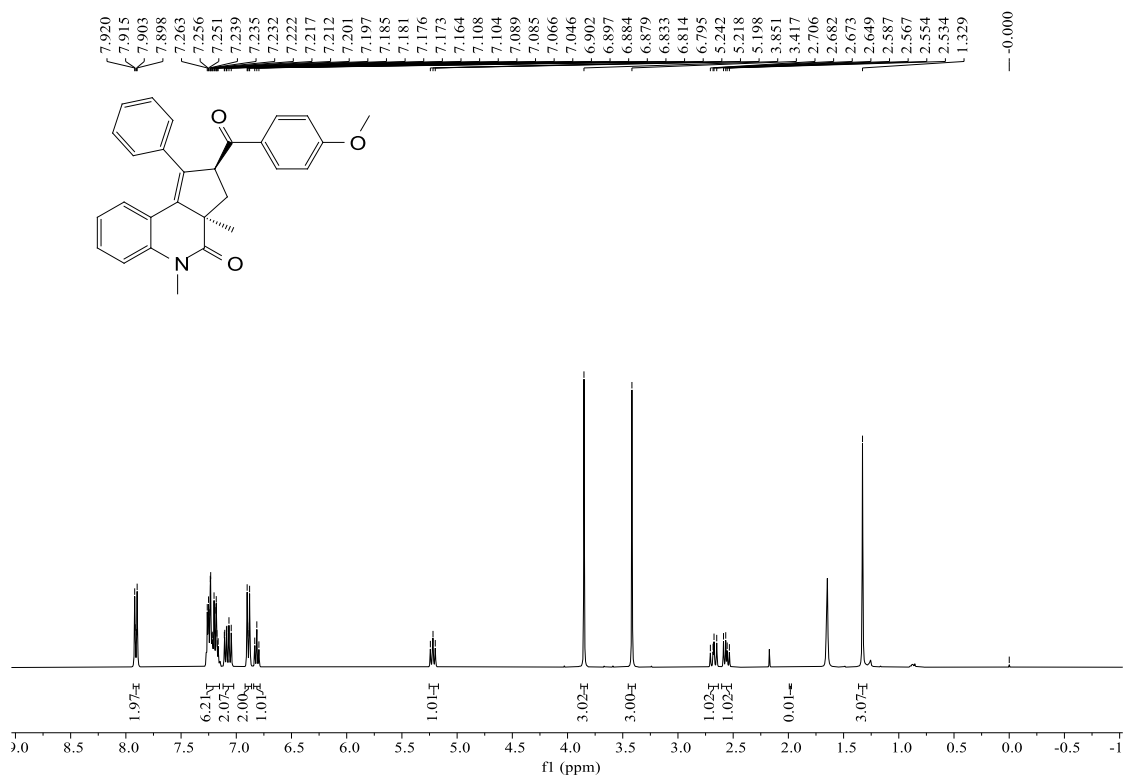
¹H NMR spectra of *cis*-3b (400 MHz, CDCl₃)



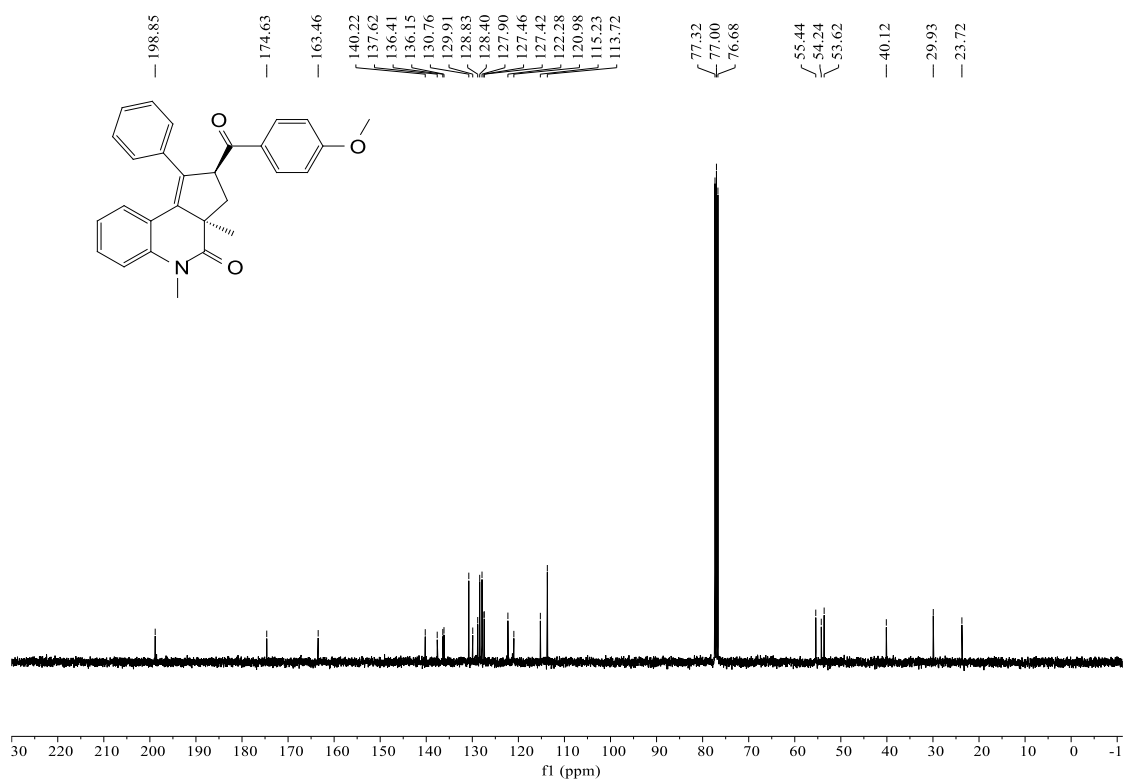
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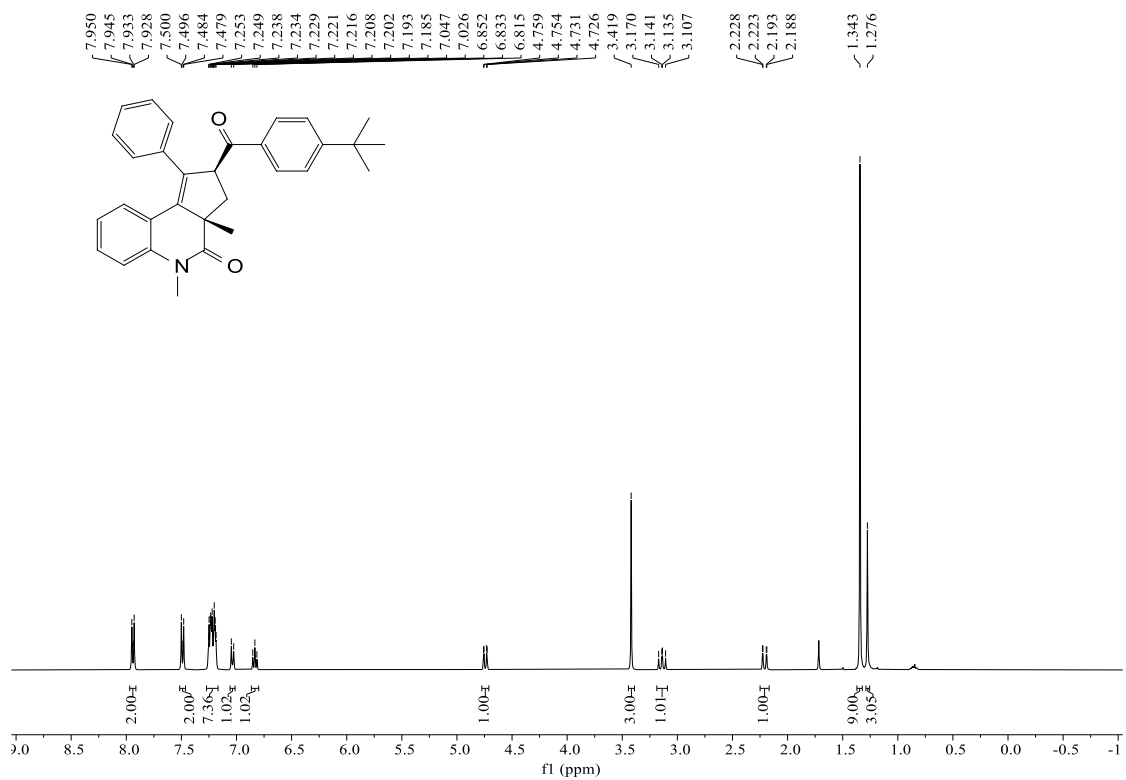
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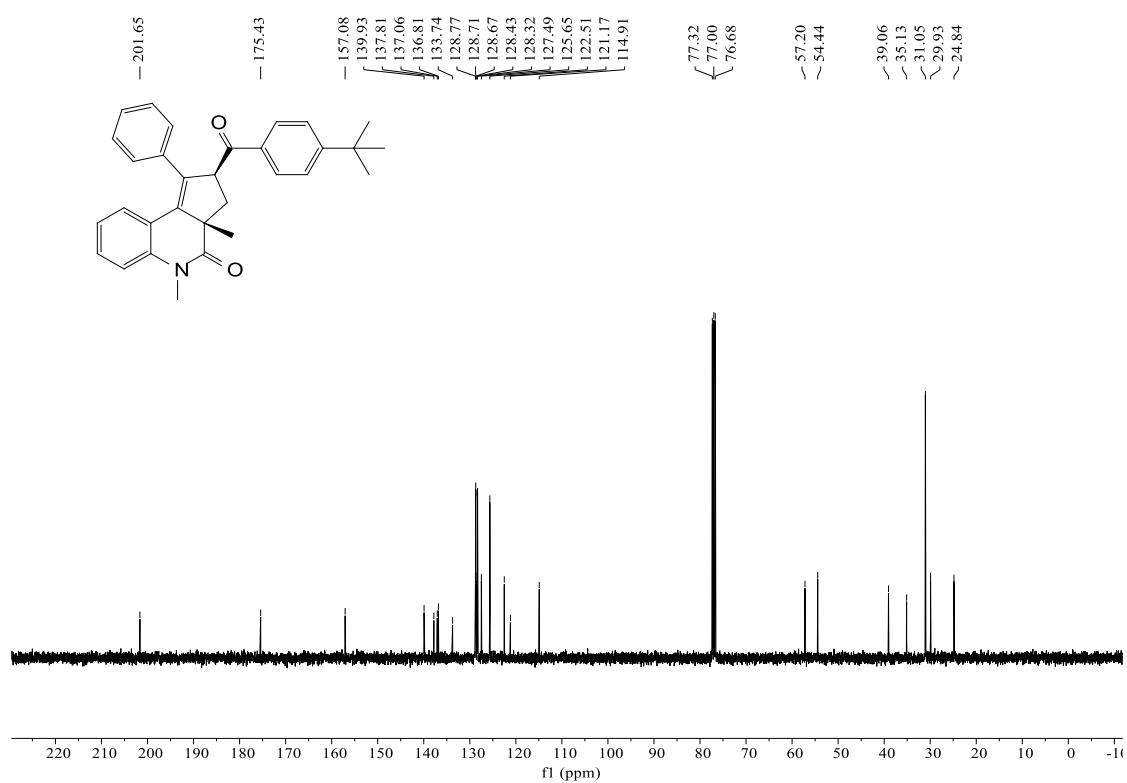
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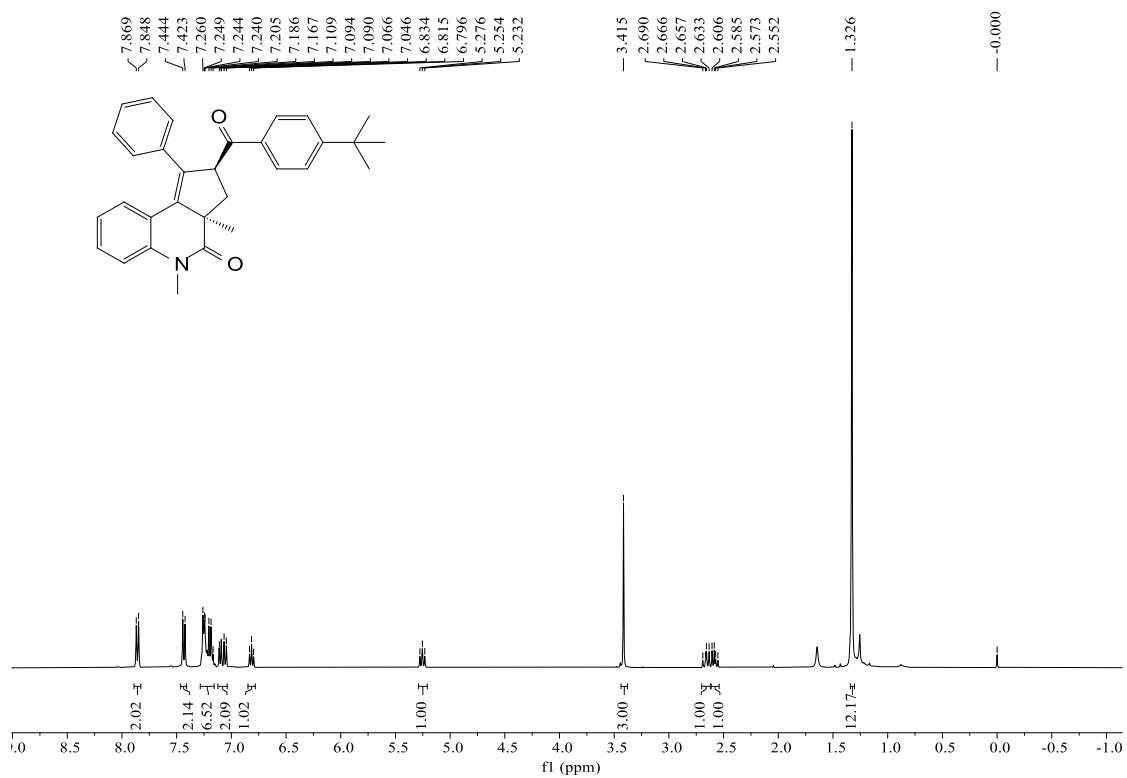
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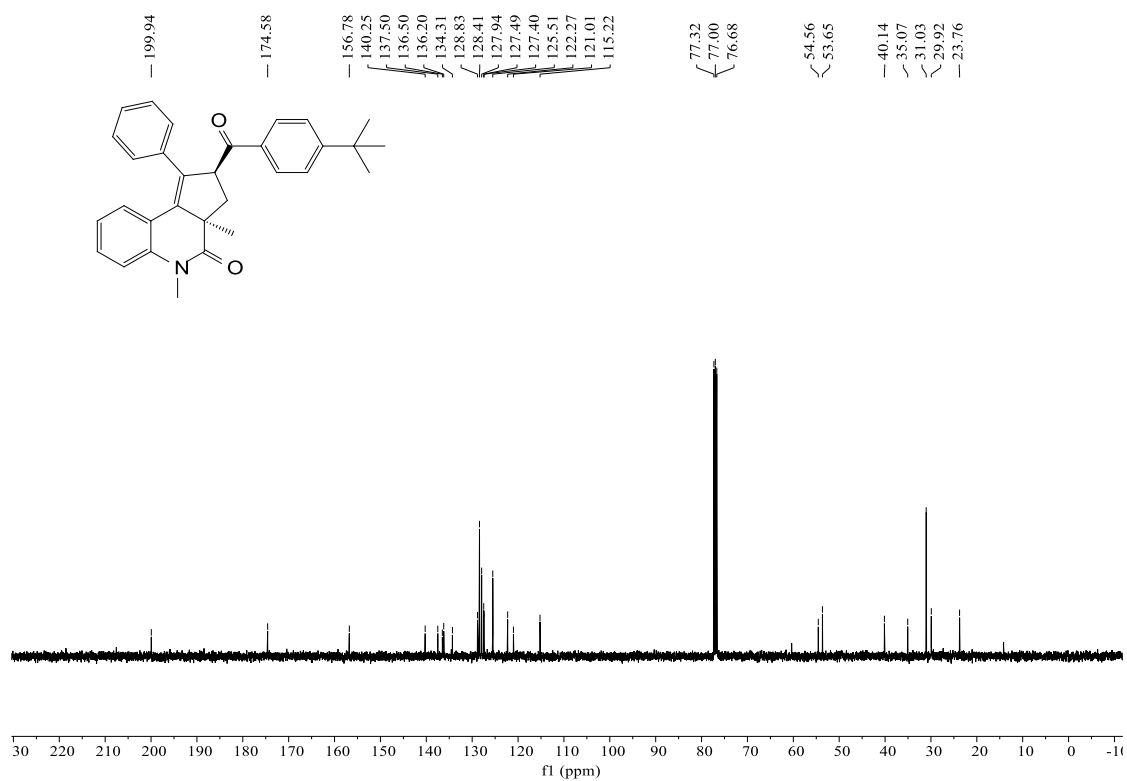
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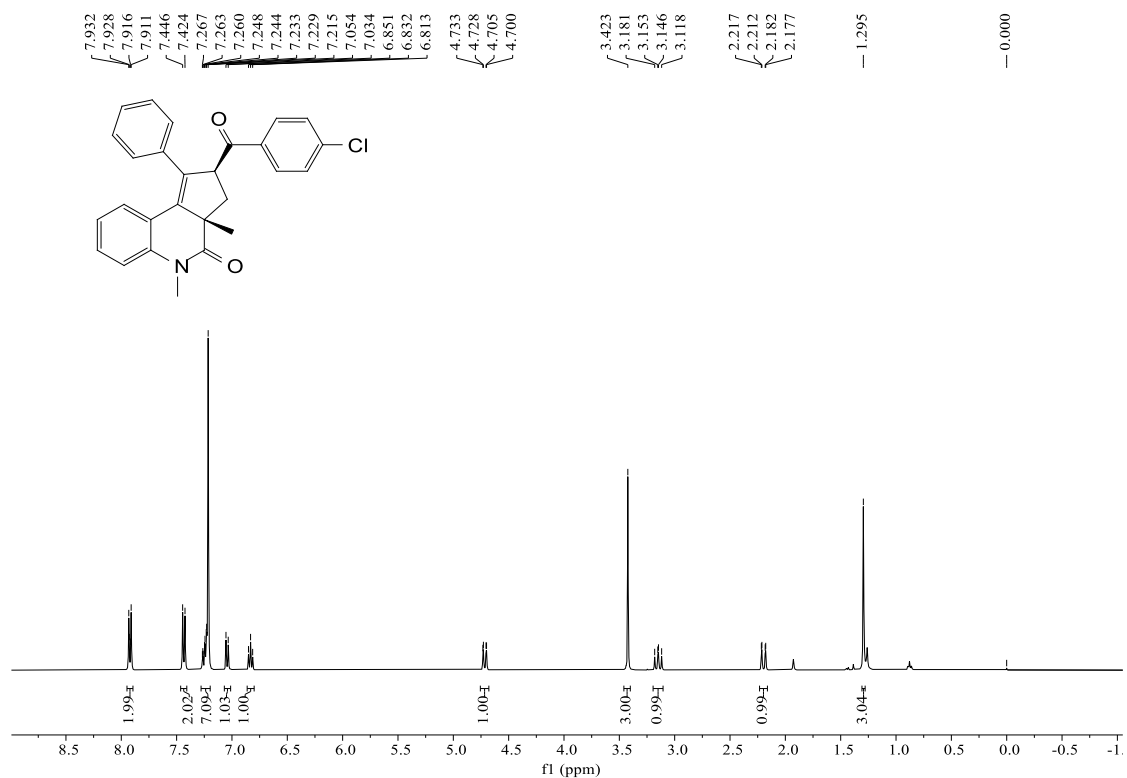
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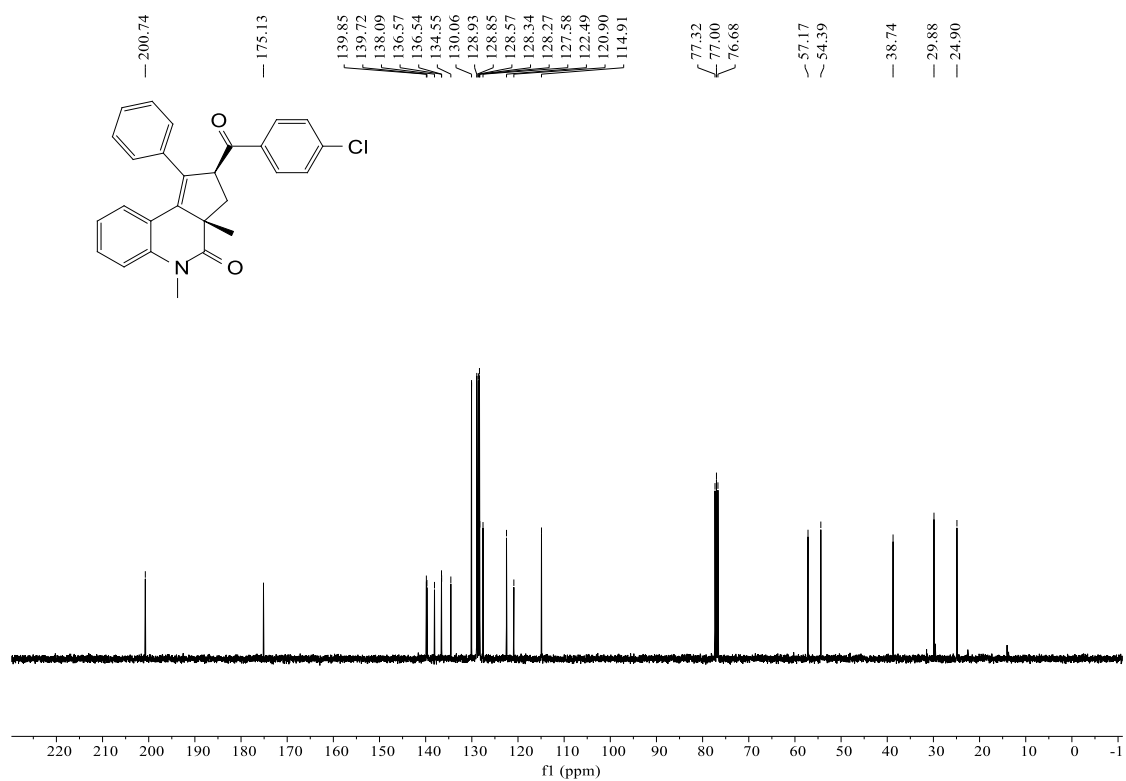
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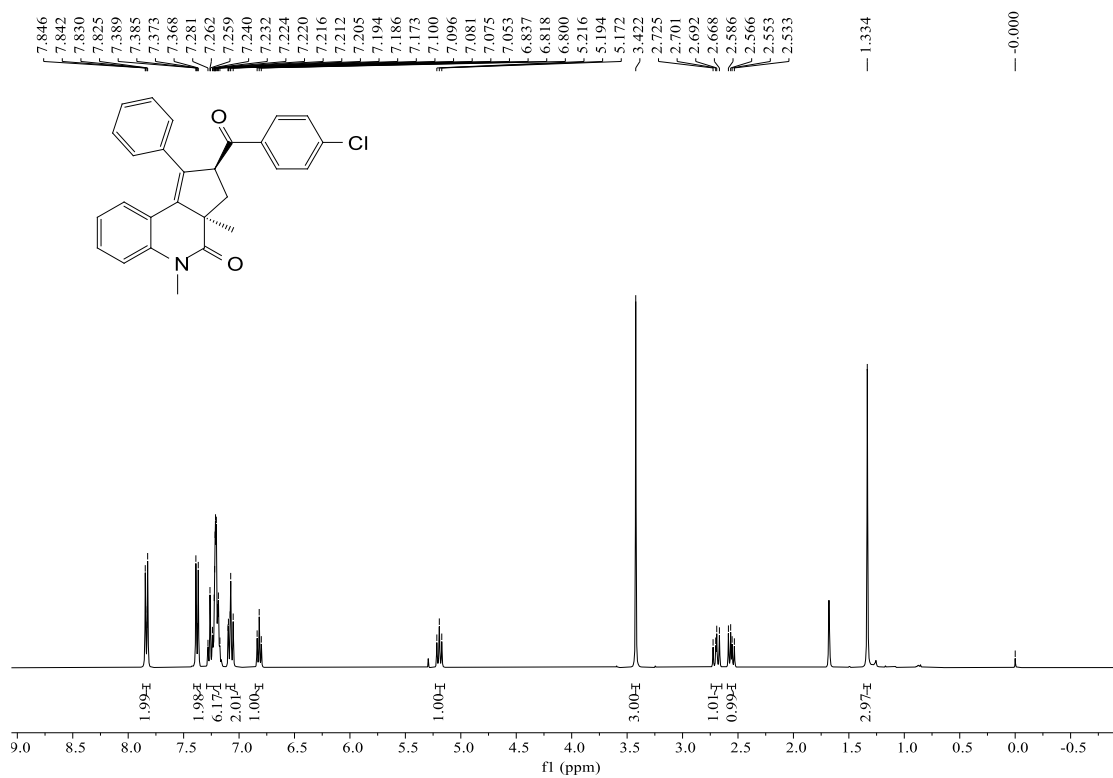
¹H NMR spectra of *cis*-3d (400 MHz, CDCl₃)



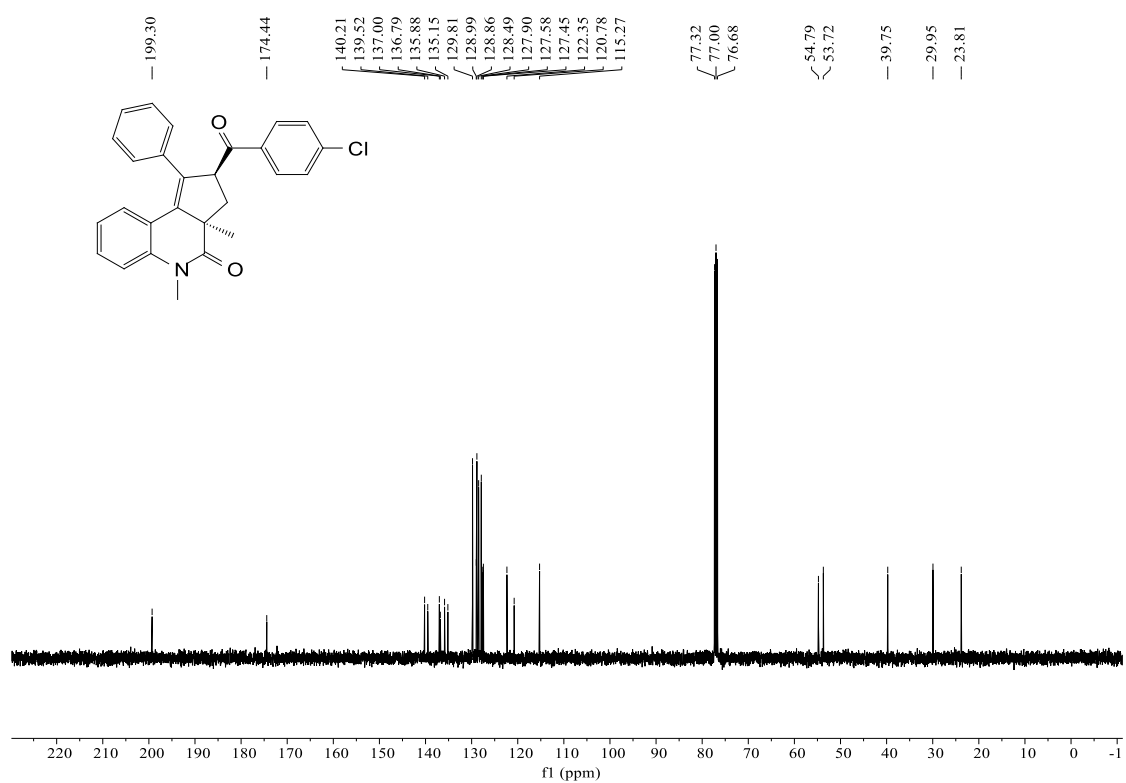
¹³C NMR spectra of *cis*-3d (100 MHz, CDCl₃)



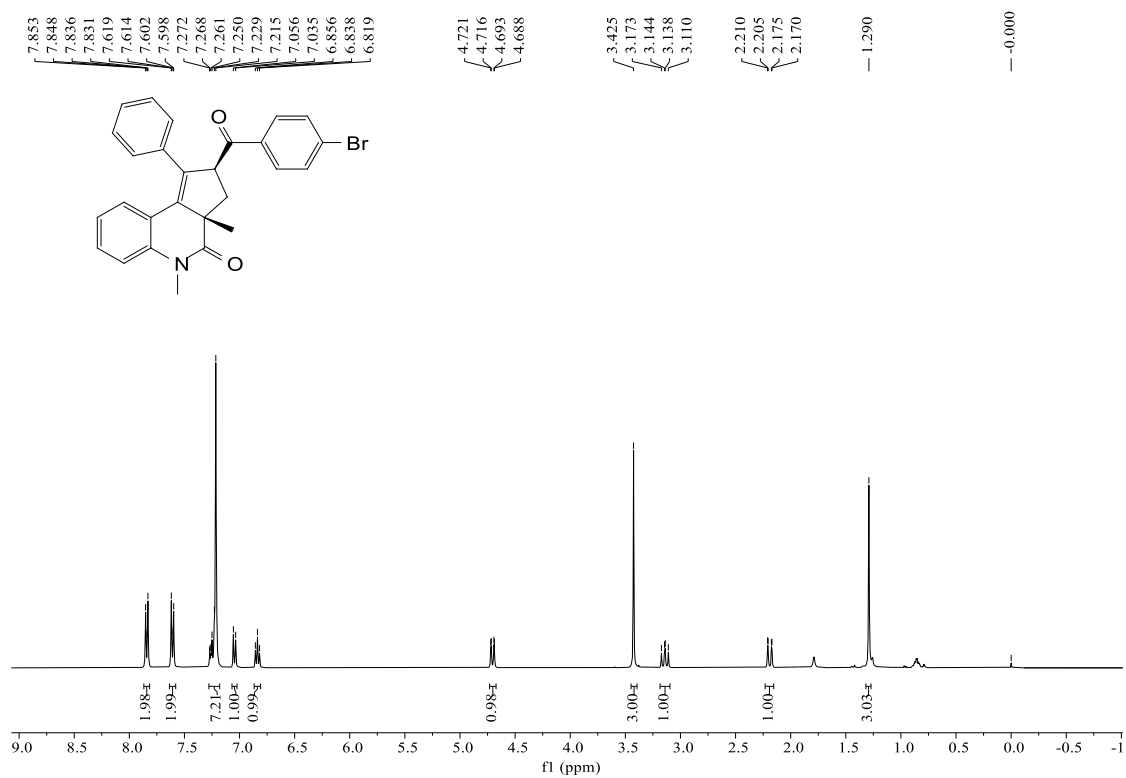
¹H NMR spectra of *trans*-3d (400 MHz, CDCl₃)



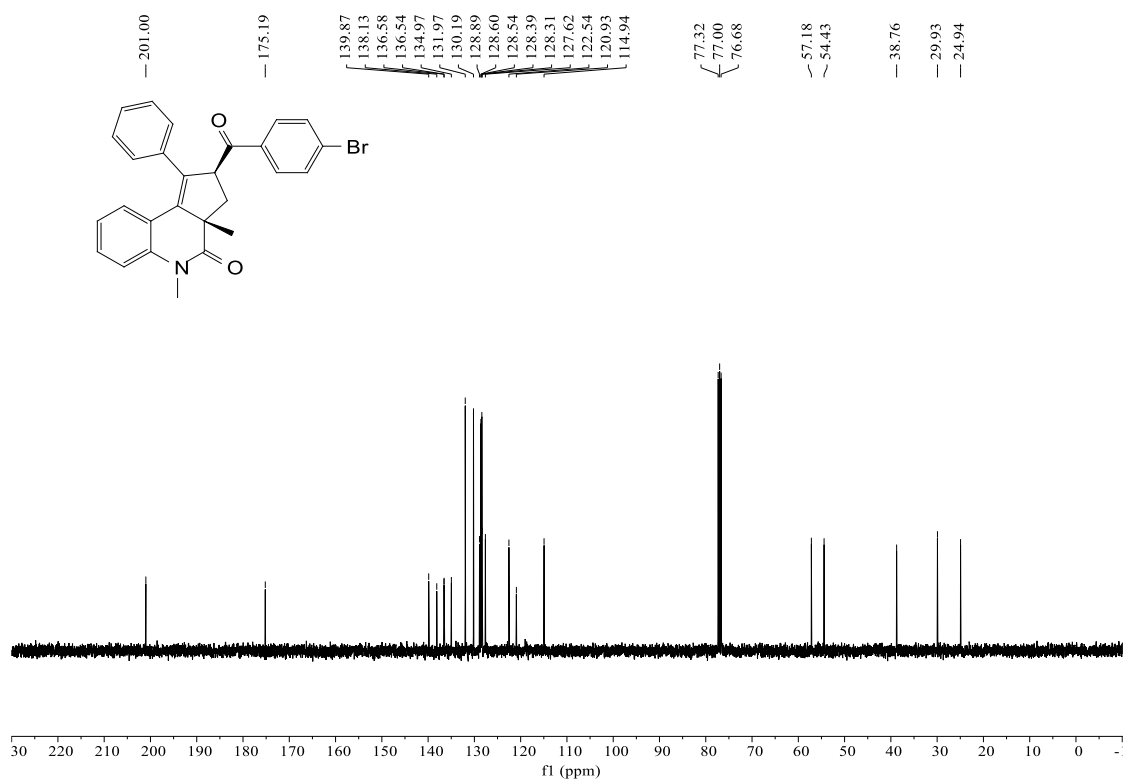
¹³C NMR spectra of *trans*-3d (100 MHz, CDCl₃)



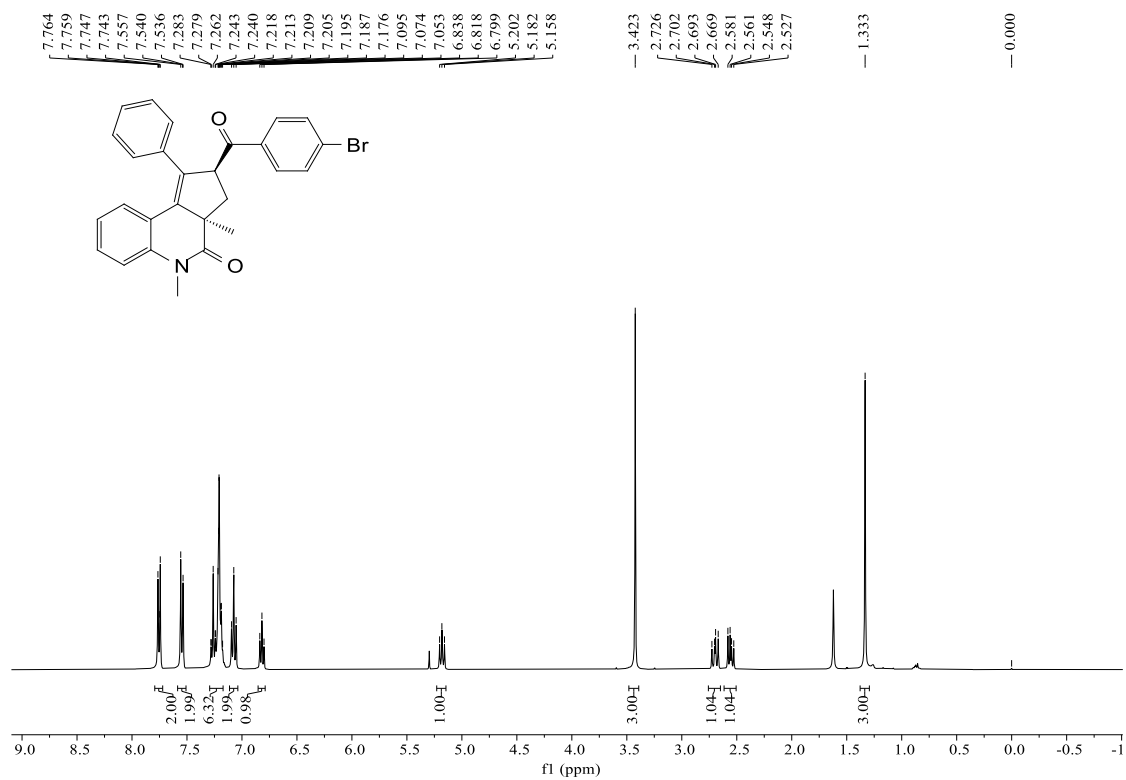
¹H NMR spectra of *cis*-3e (400 MHz, CDCl₃)



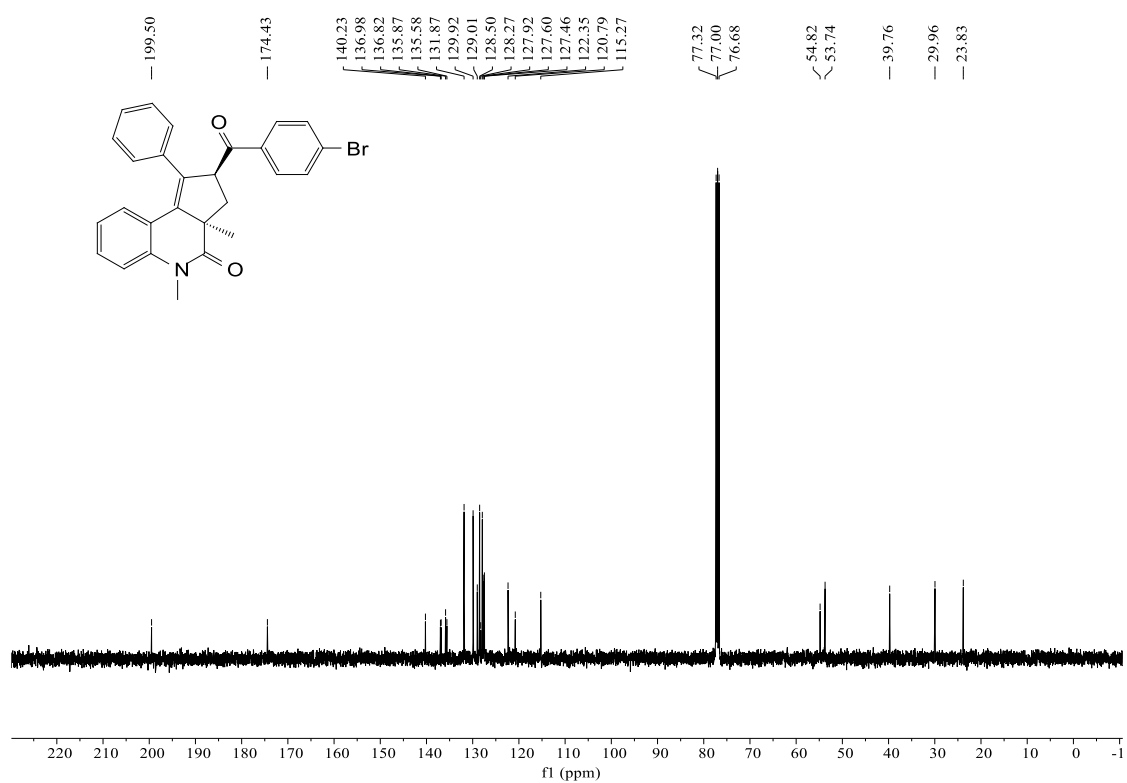
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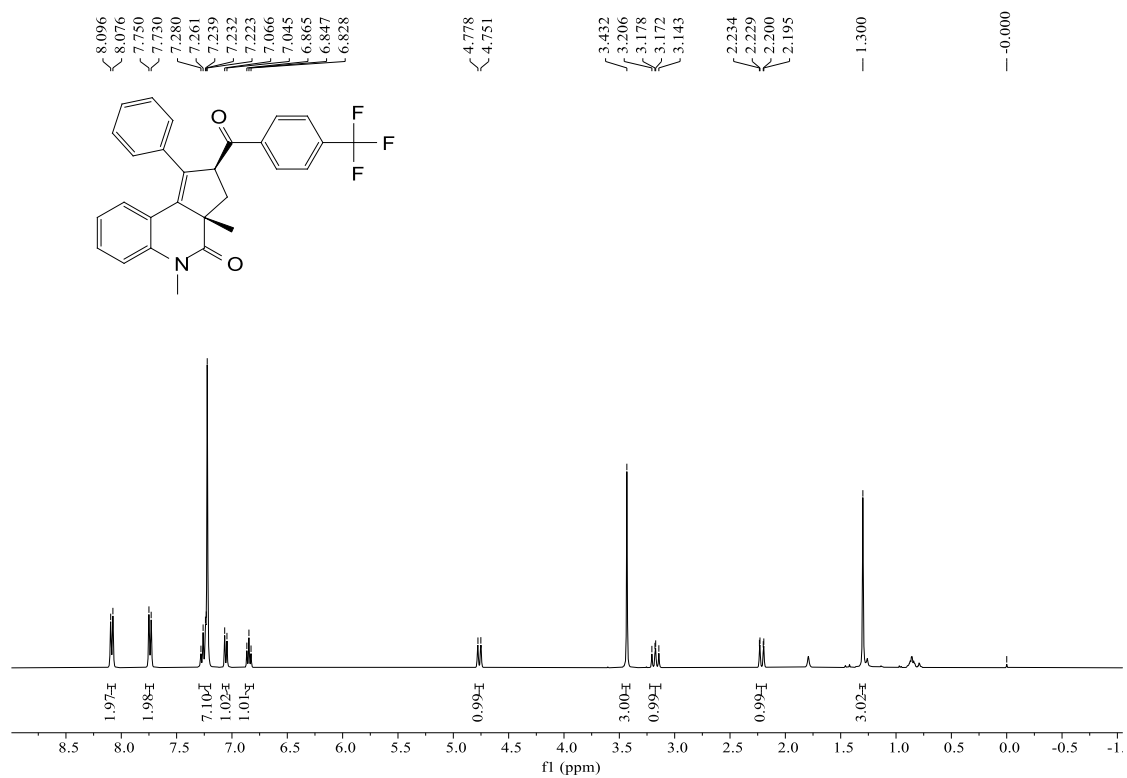
¹H NMR spectra of *trans*-3e (400 MHz, CDCl₃)



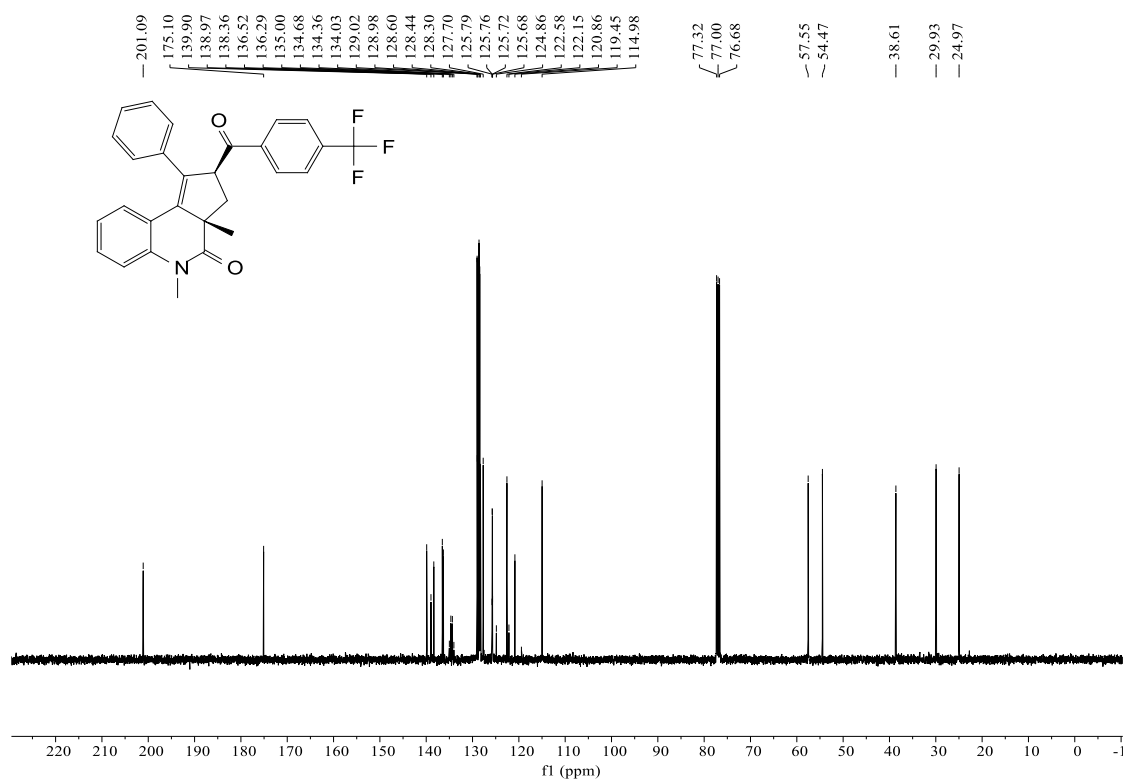
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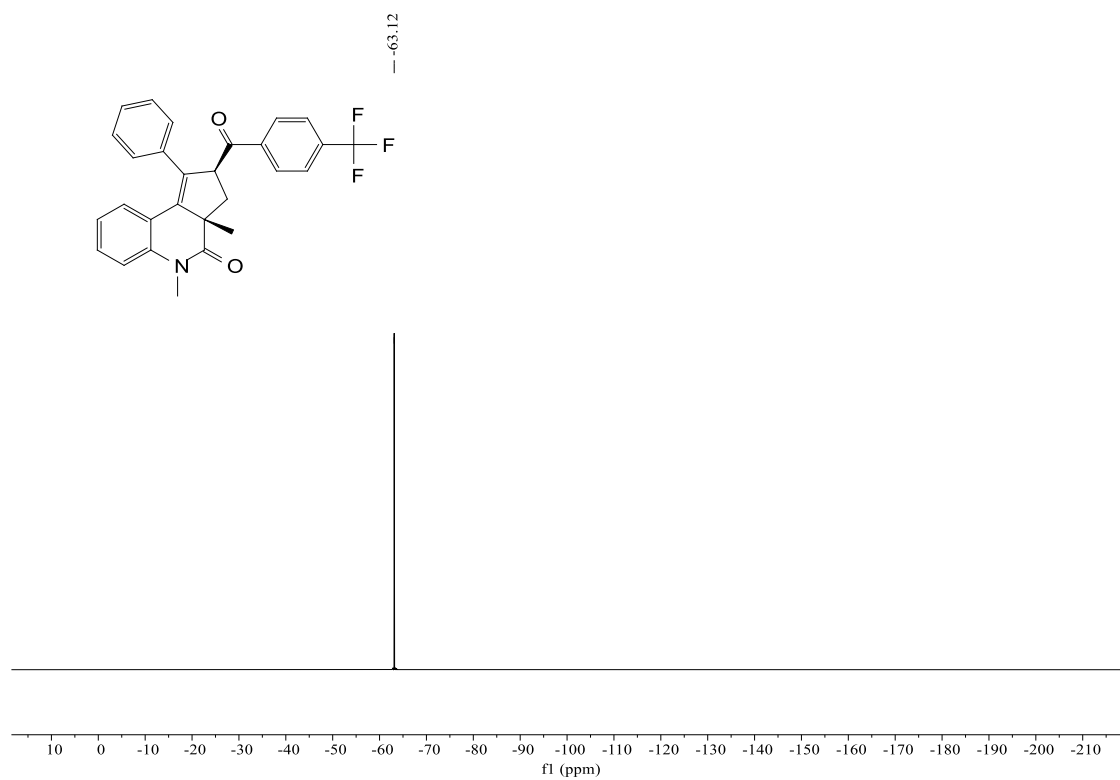
¹H NMR spectra of *cis*-3f (400 MHz, CDCl₃)



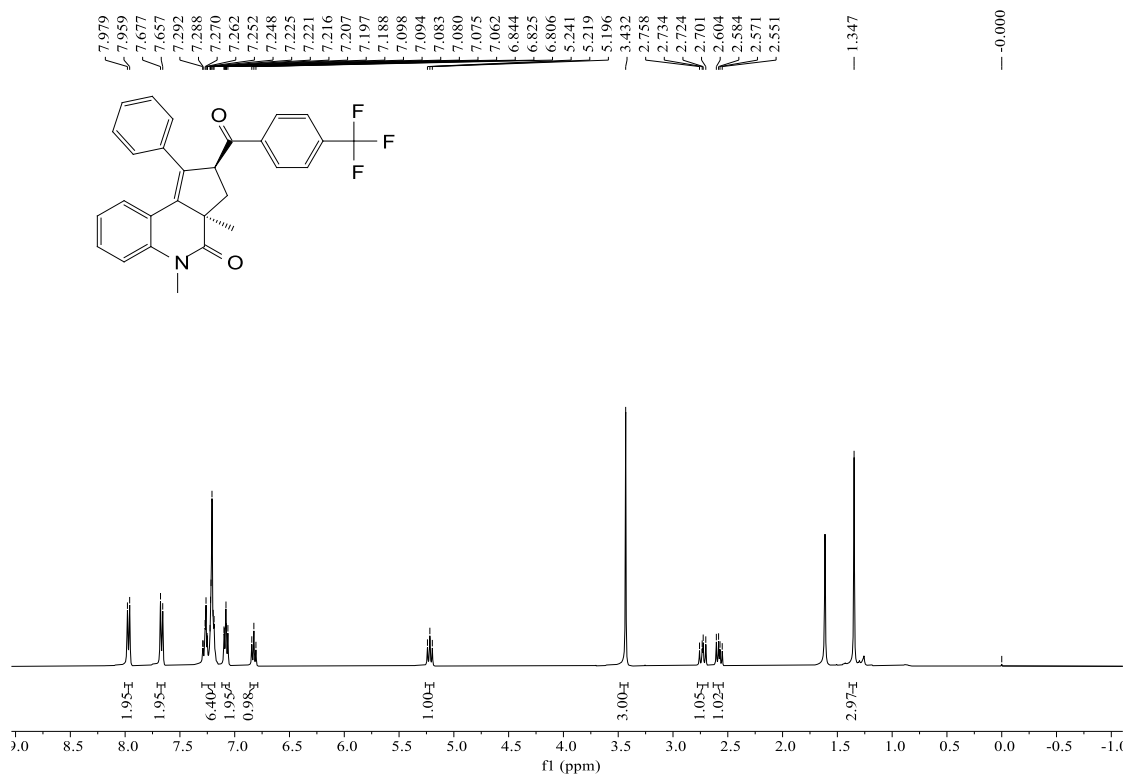
¹³C NMR spectra of *cis*-3f (100 MHz, CDCl₃)



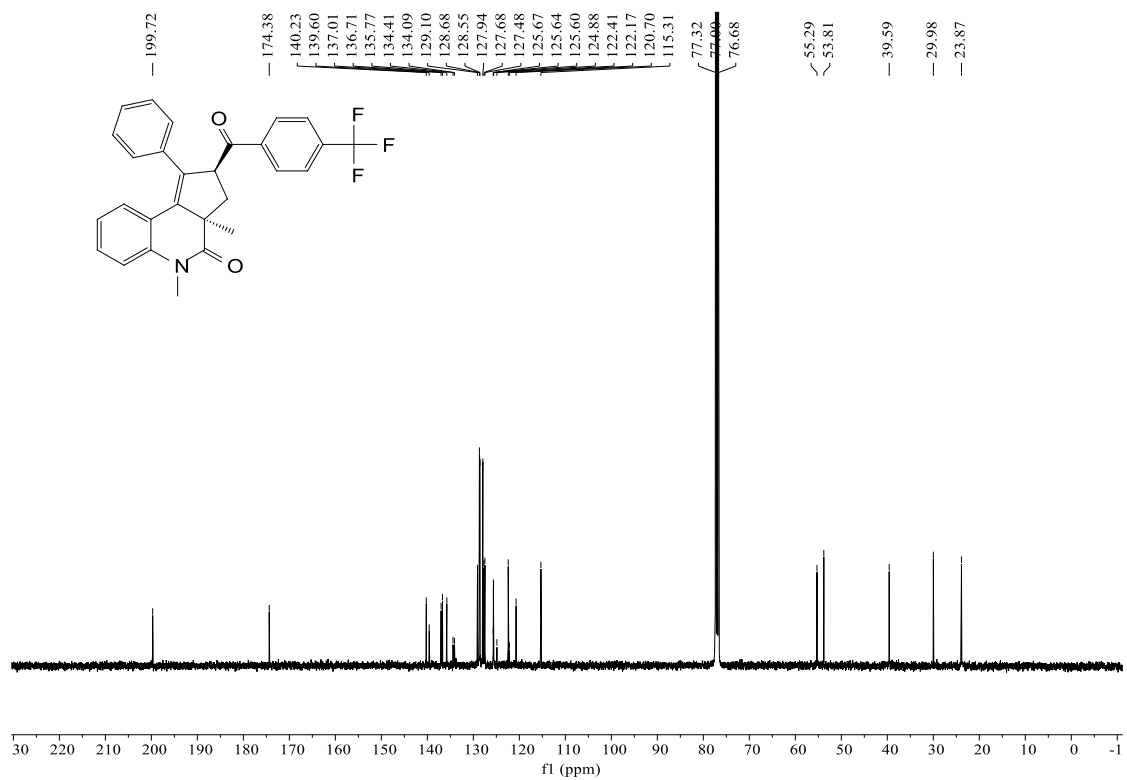
¹⁹F NMR spectra of *cis*-3f (376 MHz, CDCl₃)



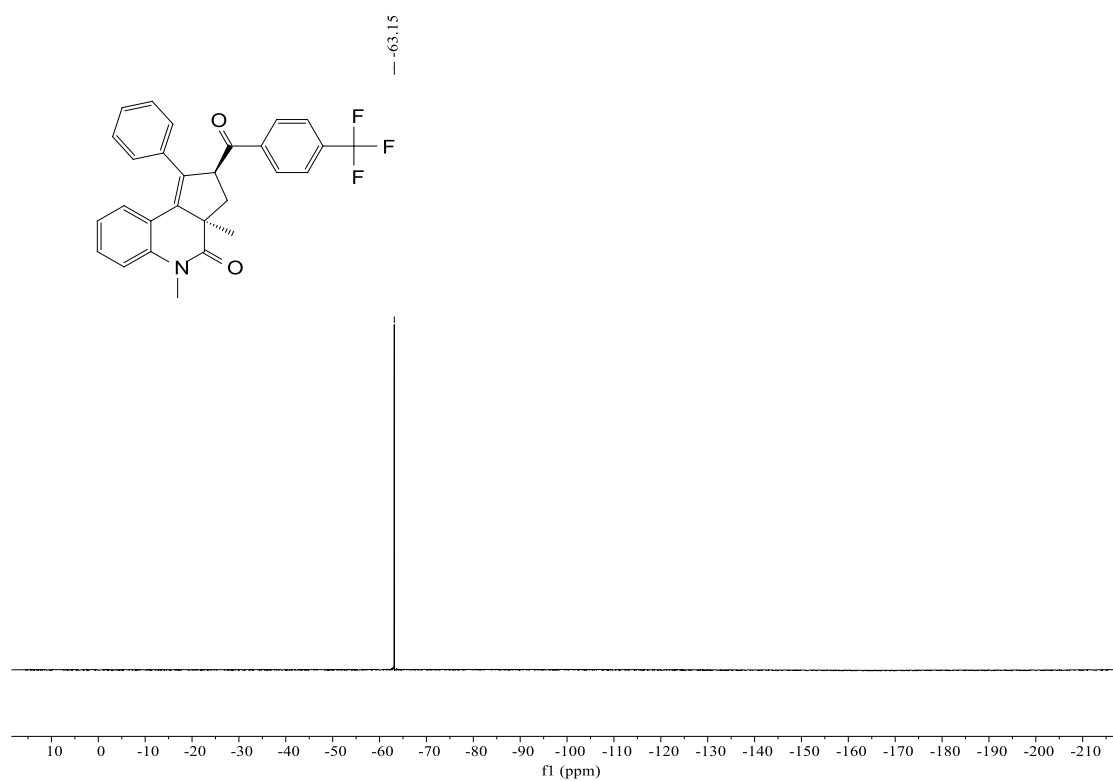
¹H NMR spectra of *trans*-3f (400 MHz, CDCl₃)



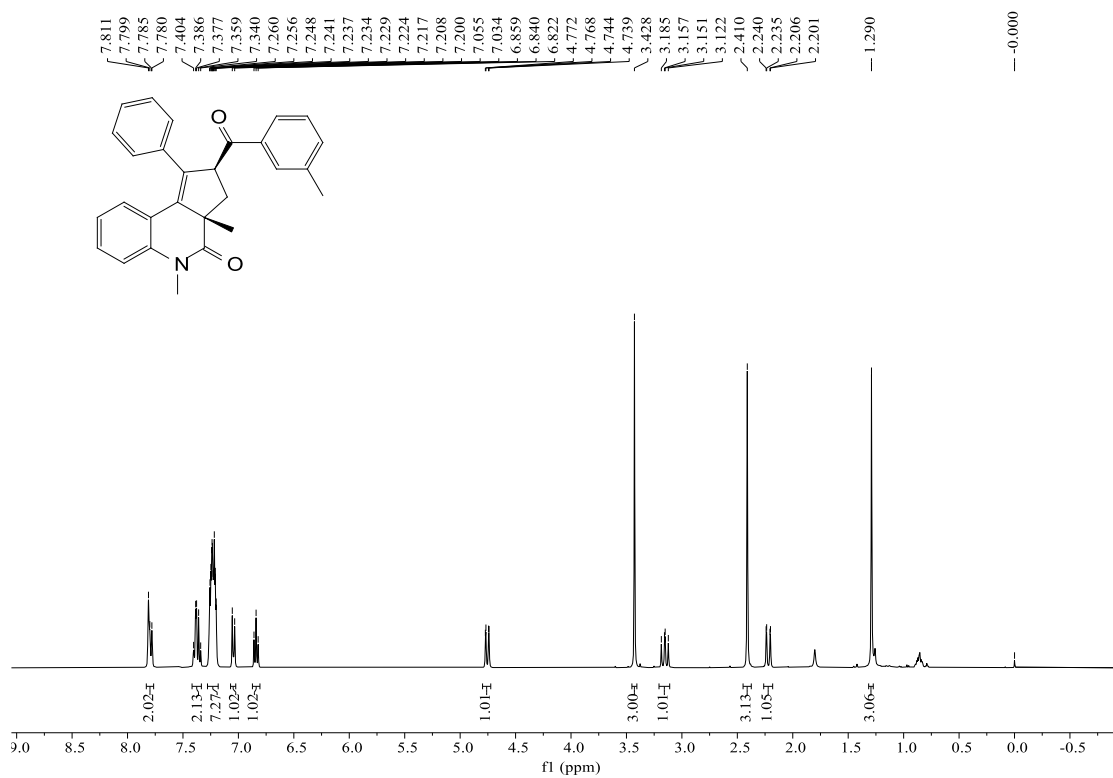
¹³C NMR spectra of *trans*-3f (100 MHz, CDCl₃)



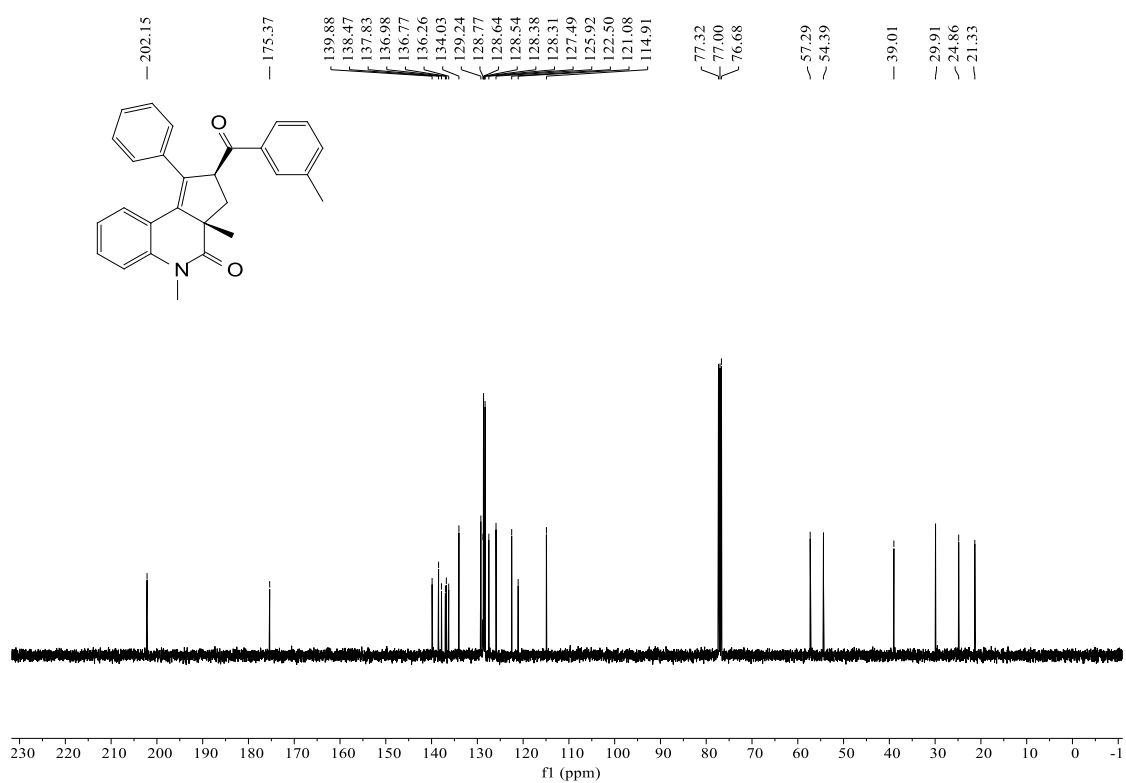
¹⁹F NMR spectra of *trans*-3f (376 MHz, CDCl₃)



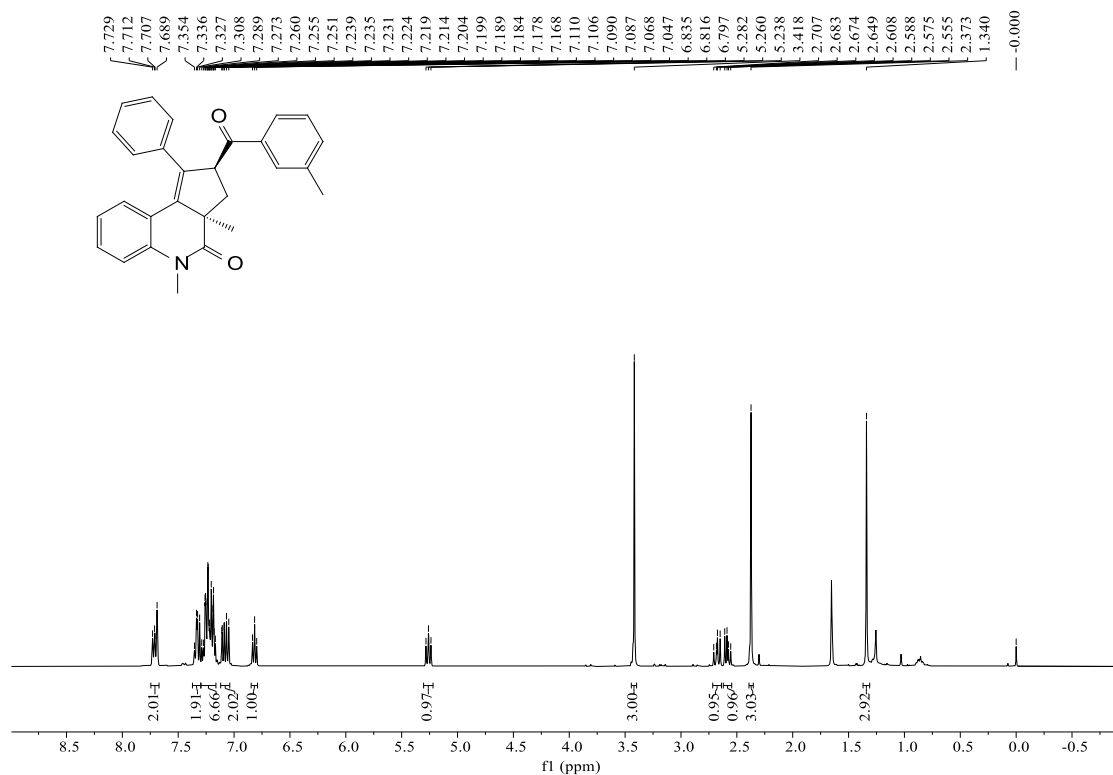
^1H NMR spectra of *cis*-3g (400 MHz, CDCl_3)



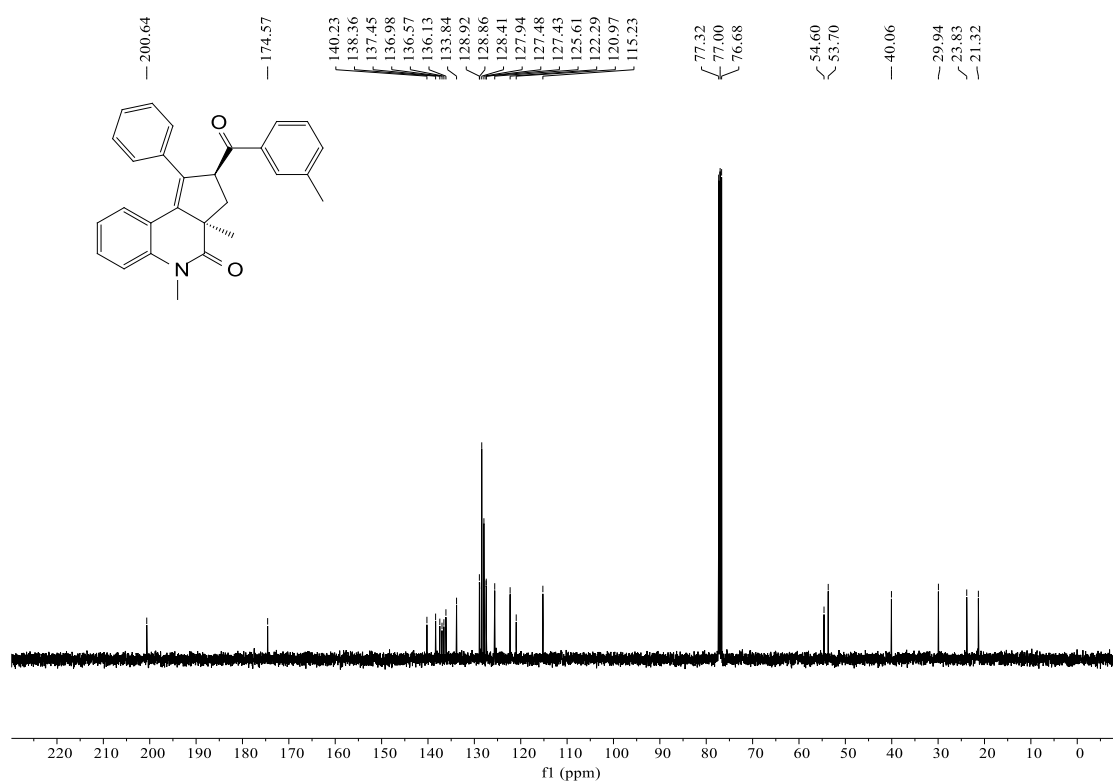
^{13}C NMR spectra of *cis*-3g (100 MHz, CDCl_3)



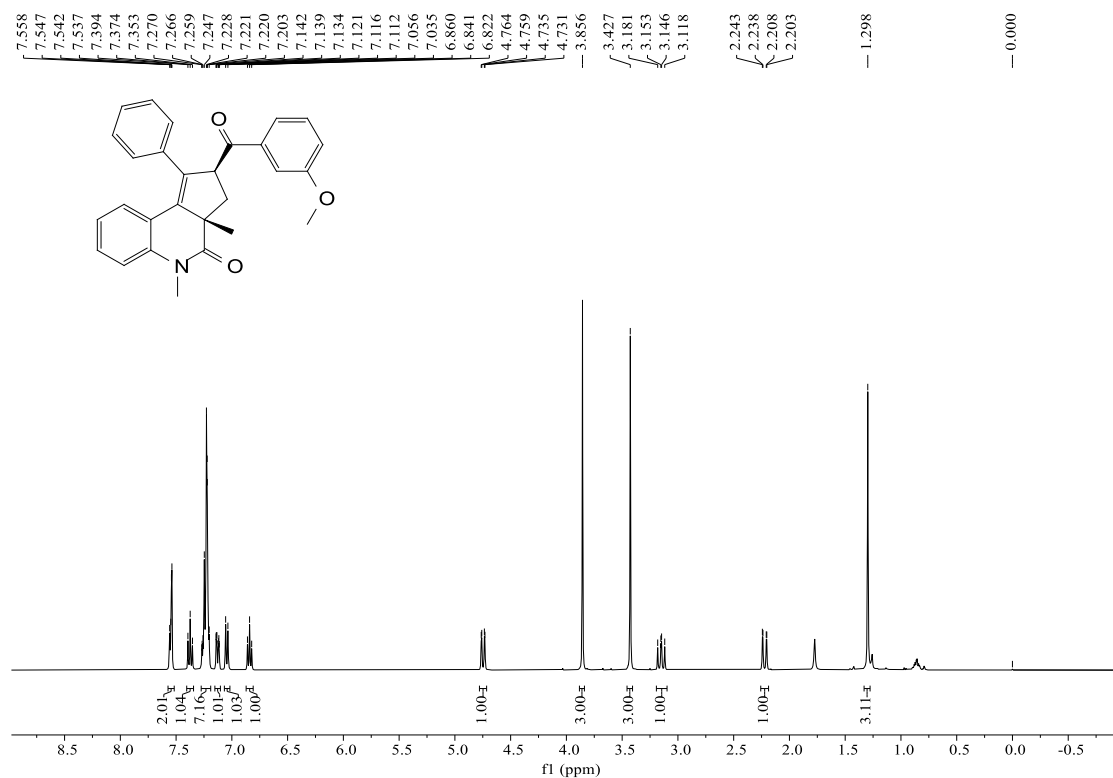
¹H NMR spectra of *trans*-3g (400 MHz, CDCl₃)



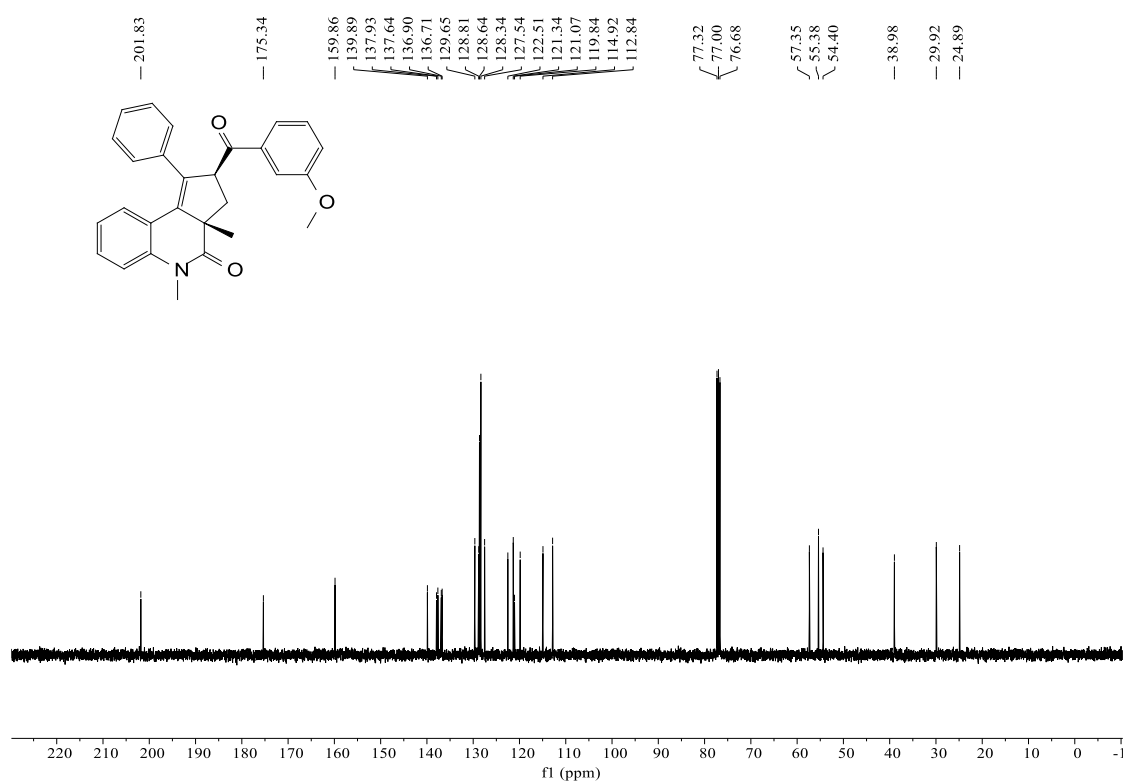
¹³C NMR spectra of *trans*-3g (100 MHz, CDCl₃)



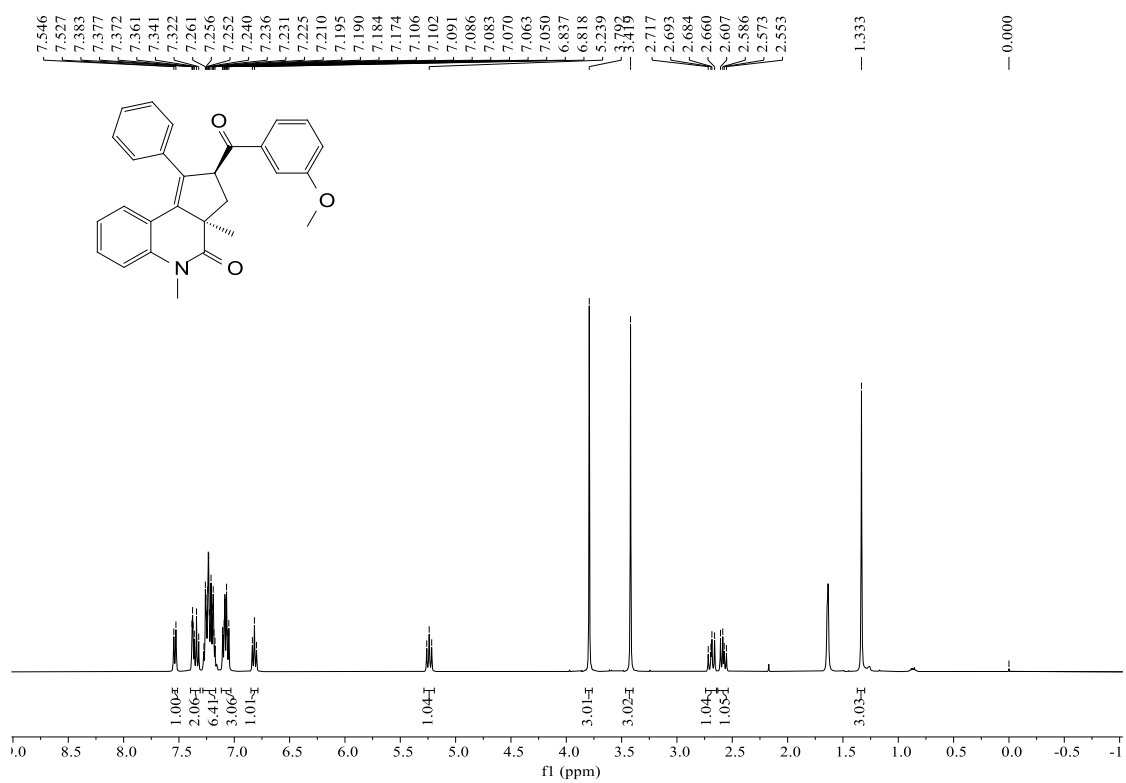
¹H NMR spectra of *cis*-3h (400 MHz, CDCl₃)



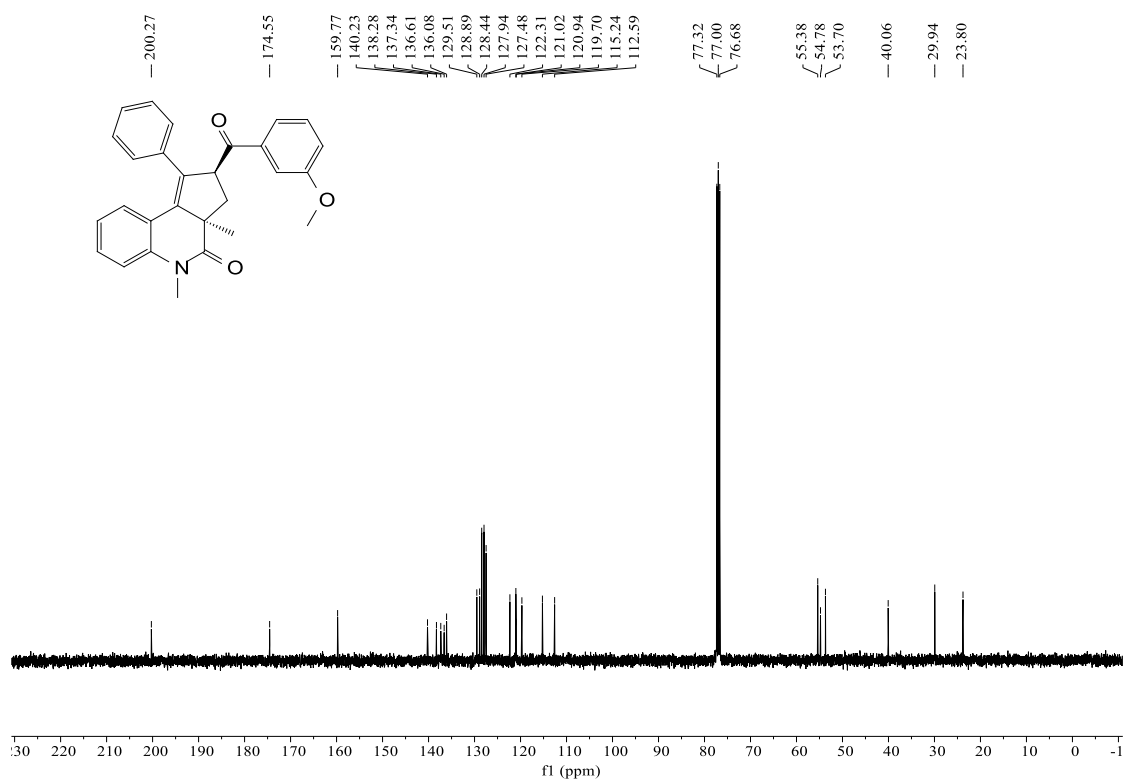
¹³C NMR spectra of *cis*-3h (100 MHz, CDCl₃)



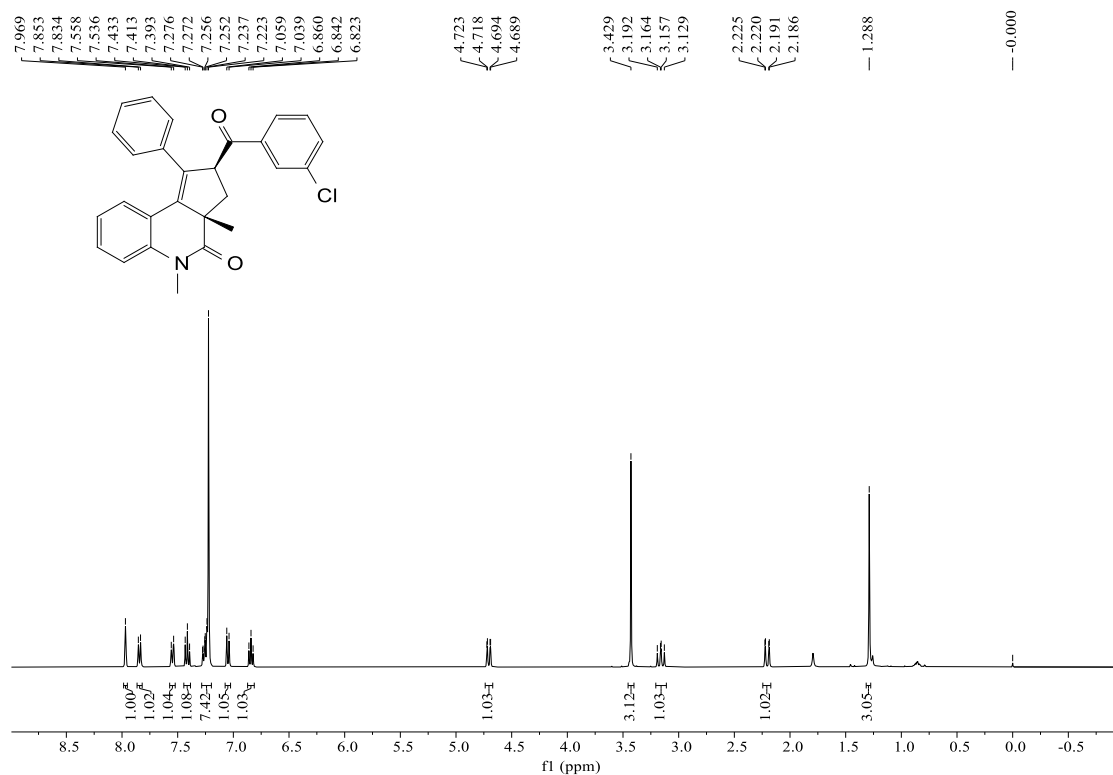
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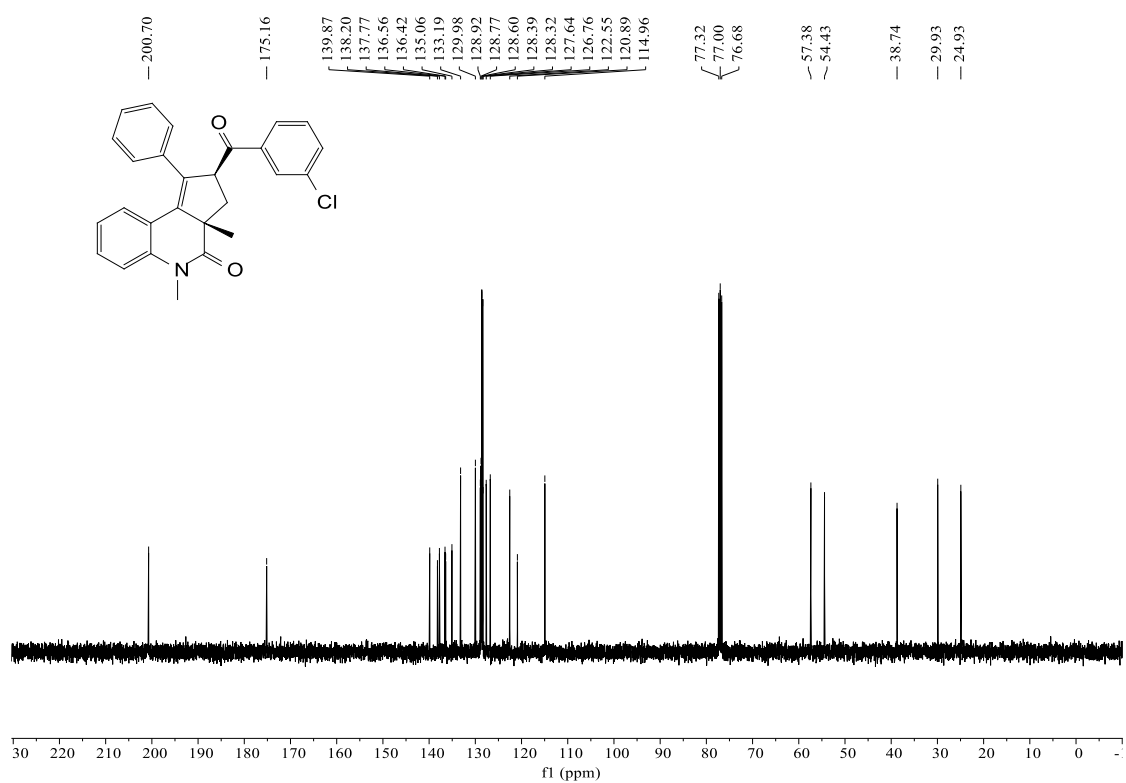
¹³C NMR spectra of *trans*-3h (100 MHz, CDCl₃)



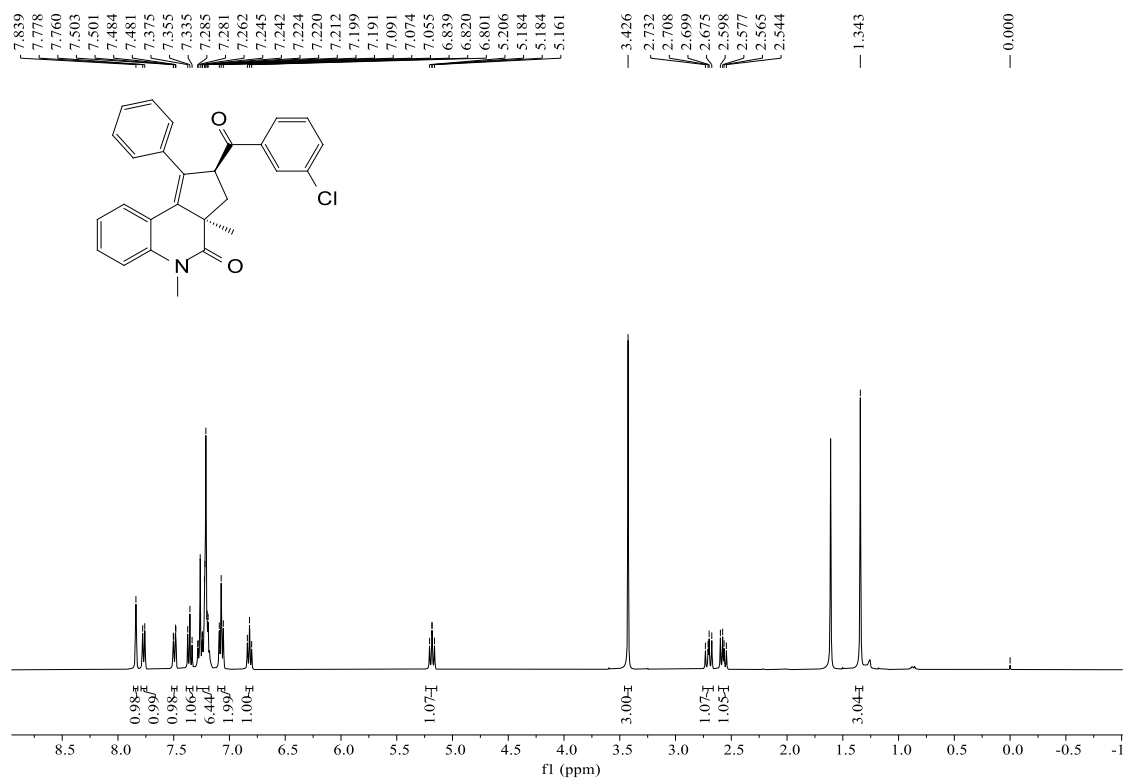
¹H NMR spectra of *cis*-3i (400 MHz, CDCl₃)



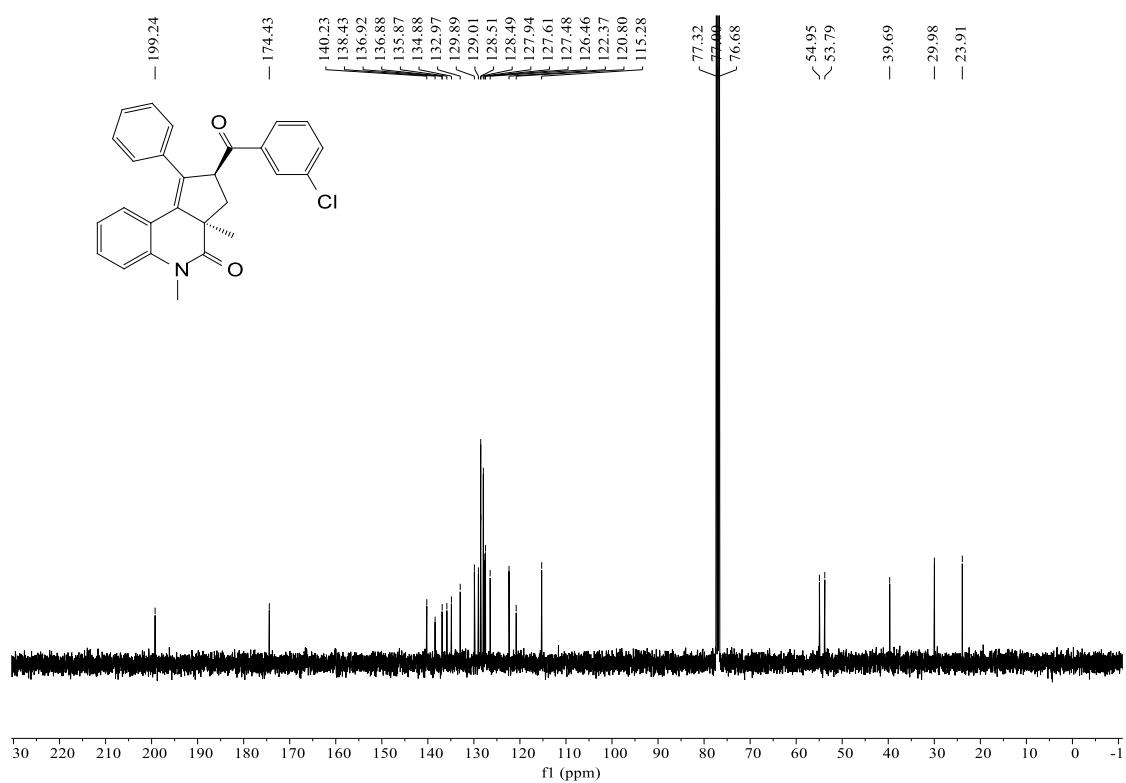
¹³C NMR spectra of *cis*-3i (100 MHz, CDCl₃)



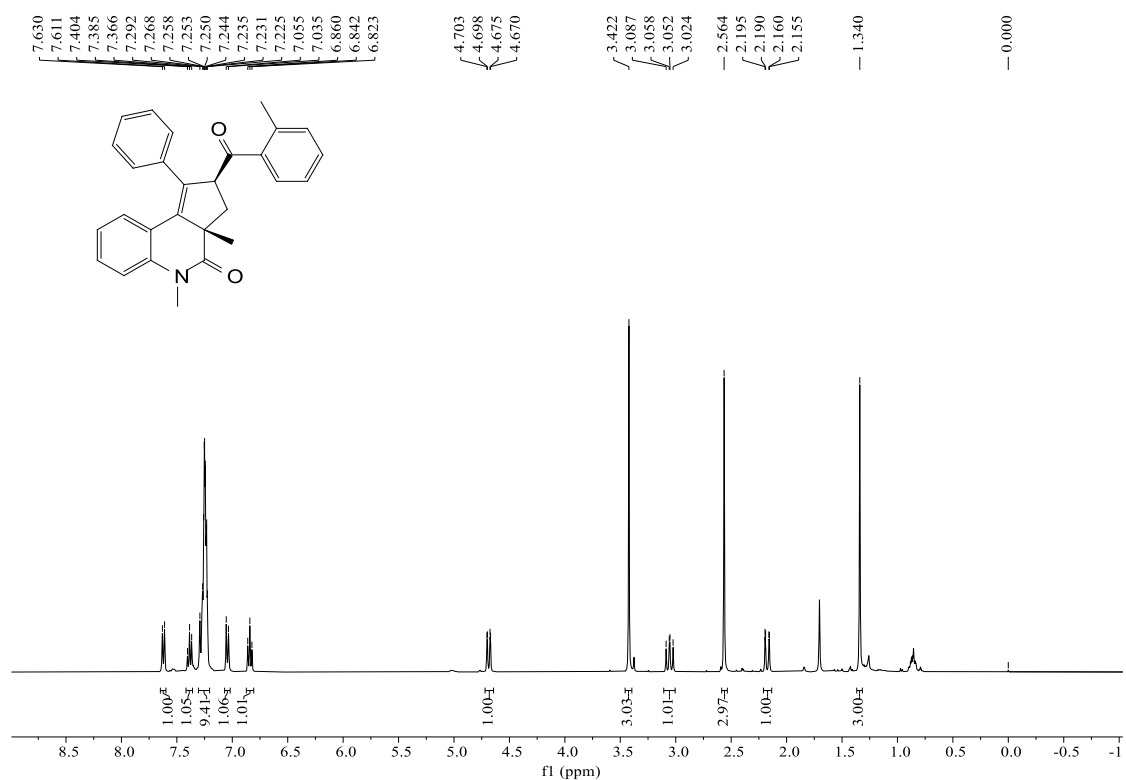
¹H NMR spectra of *trans*-3i (400 MHz, CDCl₃)



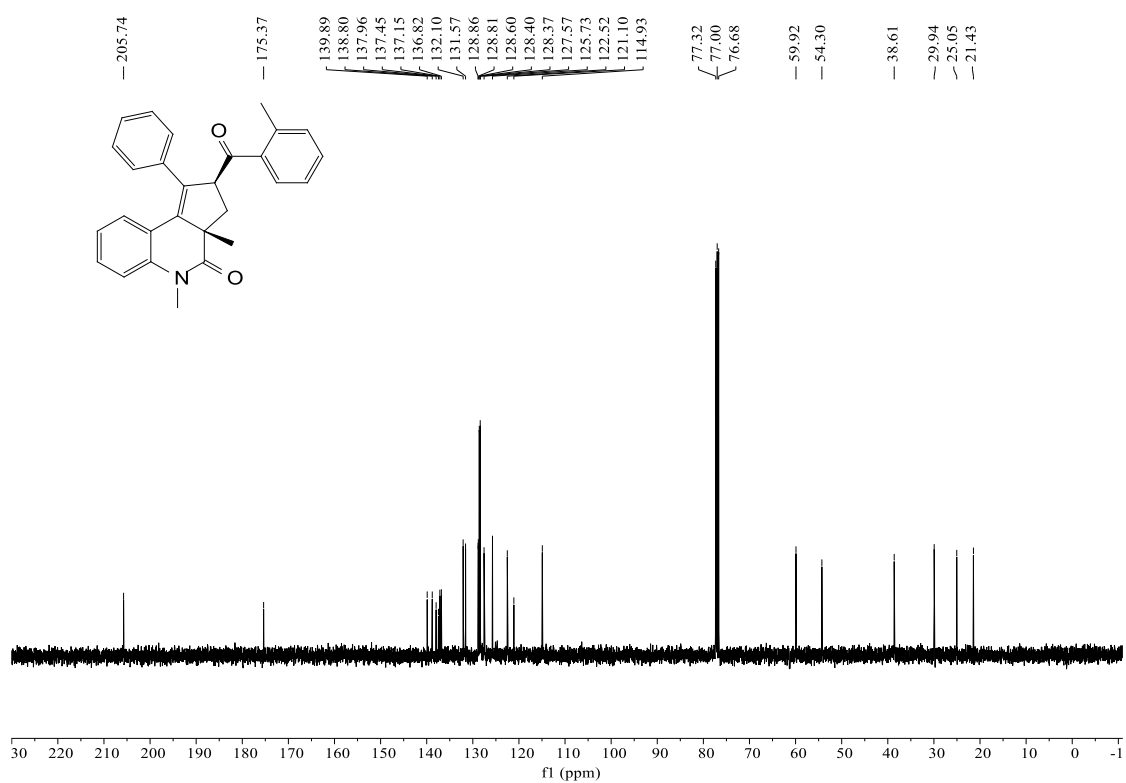
¹³C NMR spectra of *trans*-3i (100 MHz, CDCl₃)



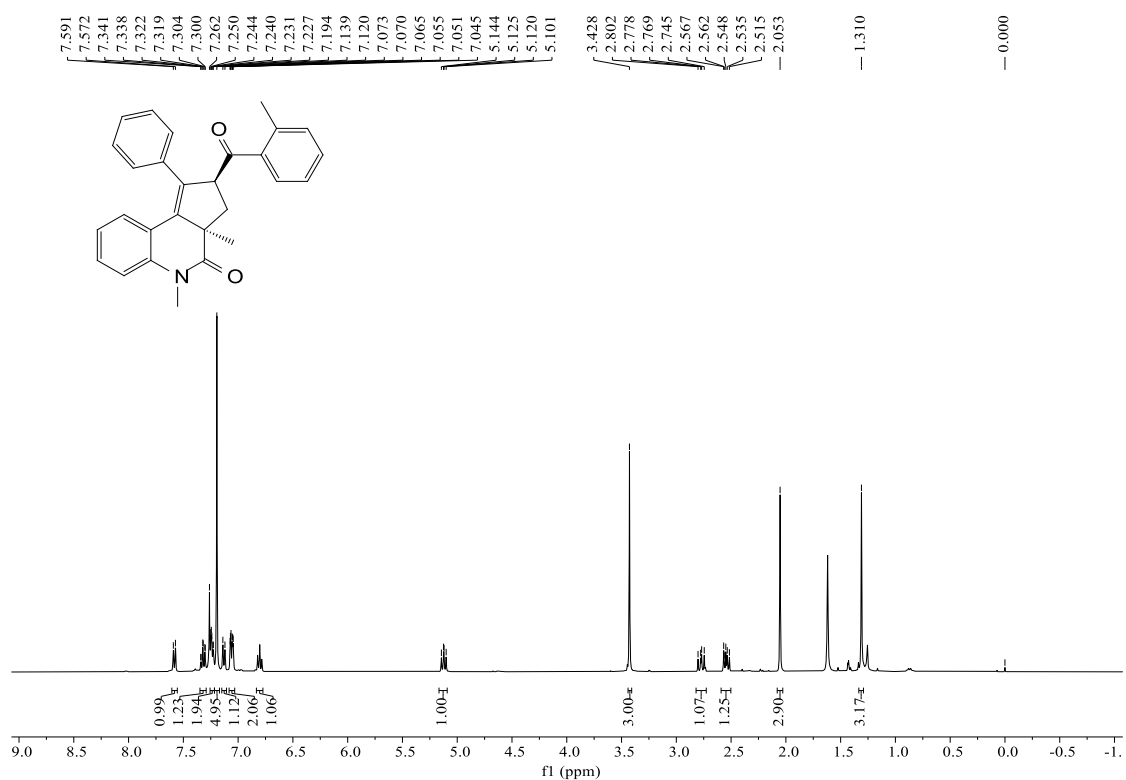
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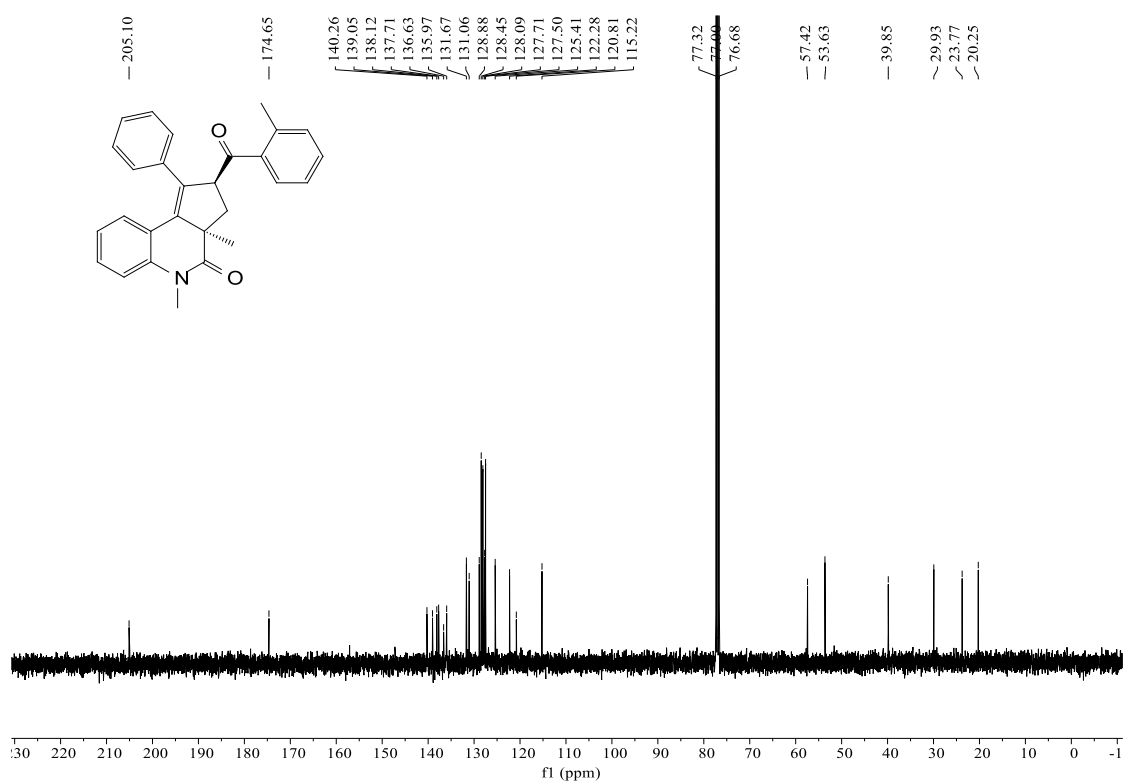
¹³C NMR spectra of *cis*-3j (100 MHz, CDCl₃)



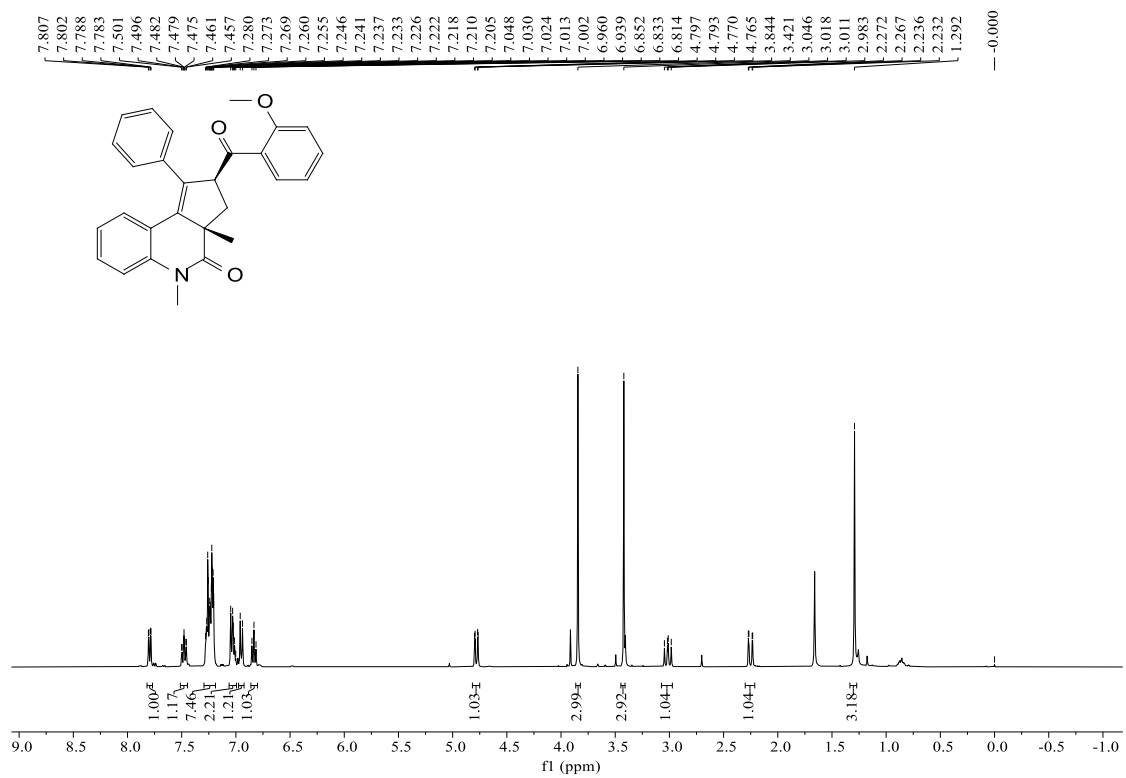
¹H NMR spectra of *trans*-3j (400 MHz, CDCl₃)



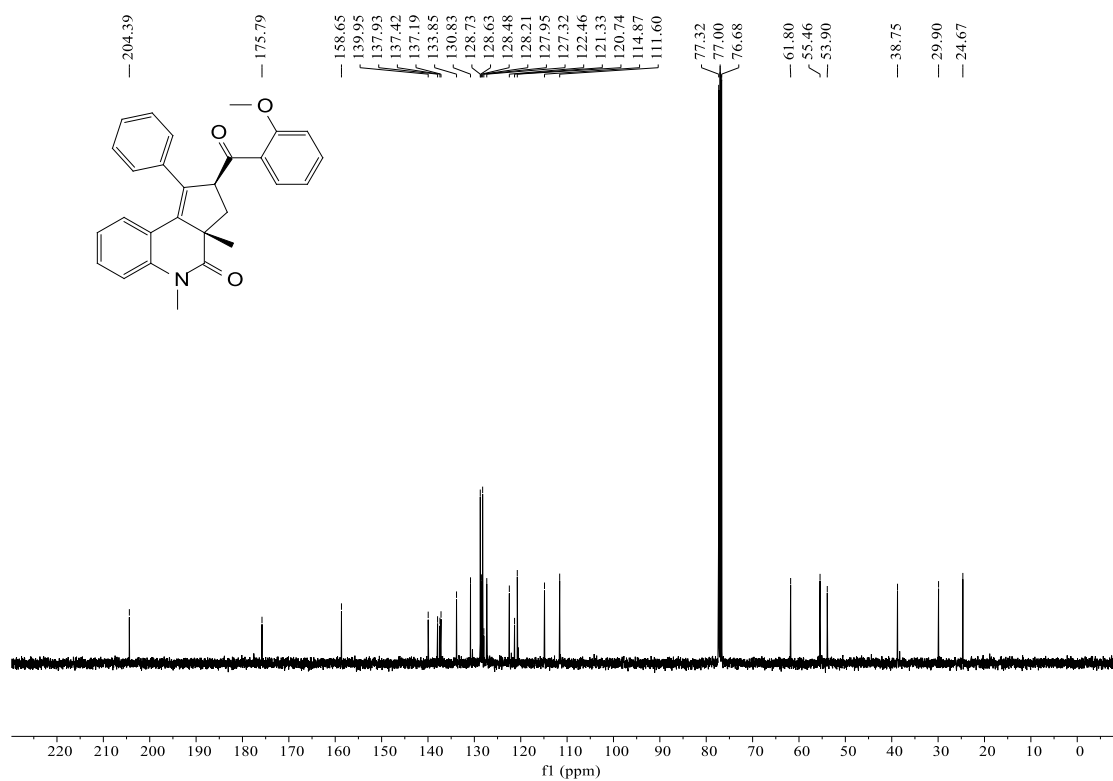
¹³C NMR spectra of *trans*-3j (100 MHz, CDCl₃)



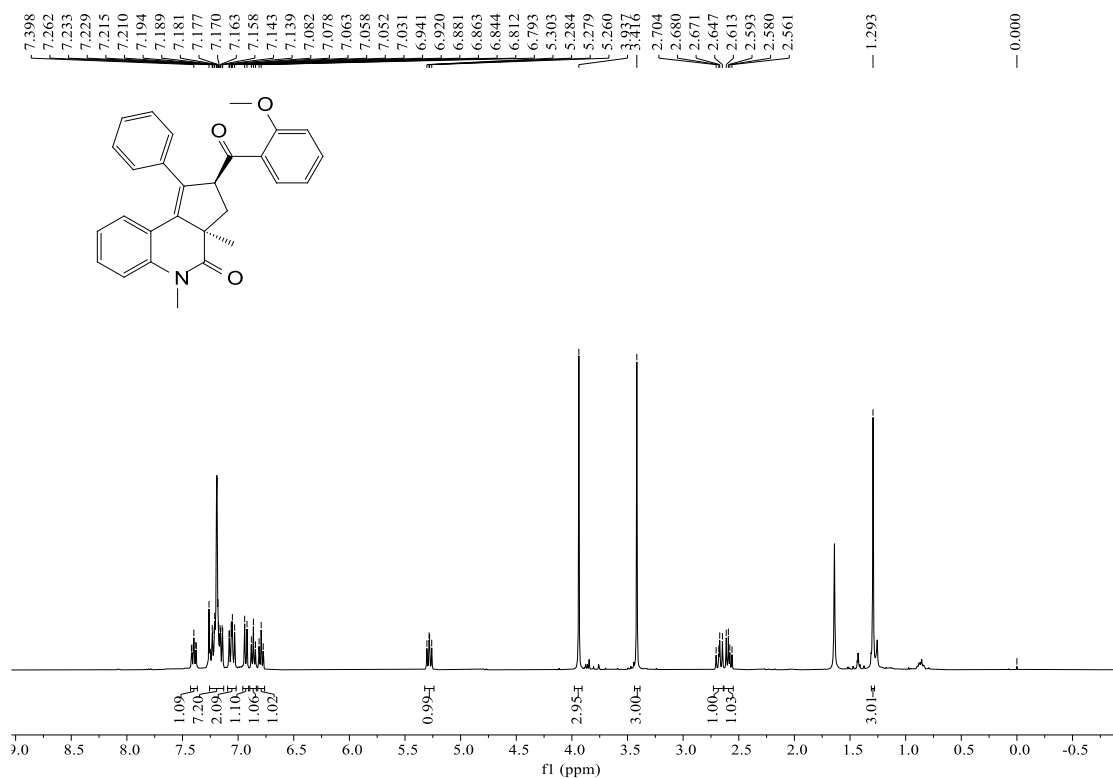
¹H NMR spectra of *cis*-3k (400 MHz, CDCl₃)



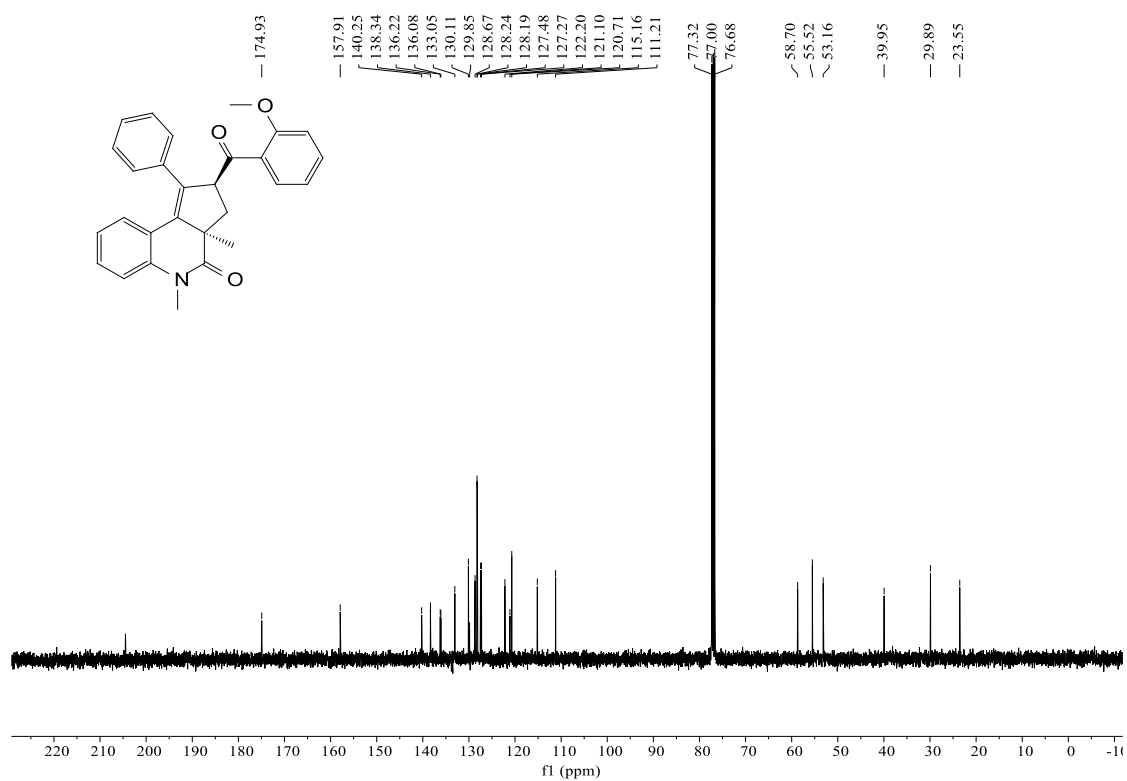
¹³C NMR spectra of *cis*-3k (100 MHz, CDCl₃)



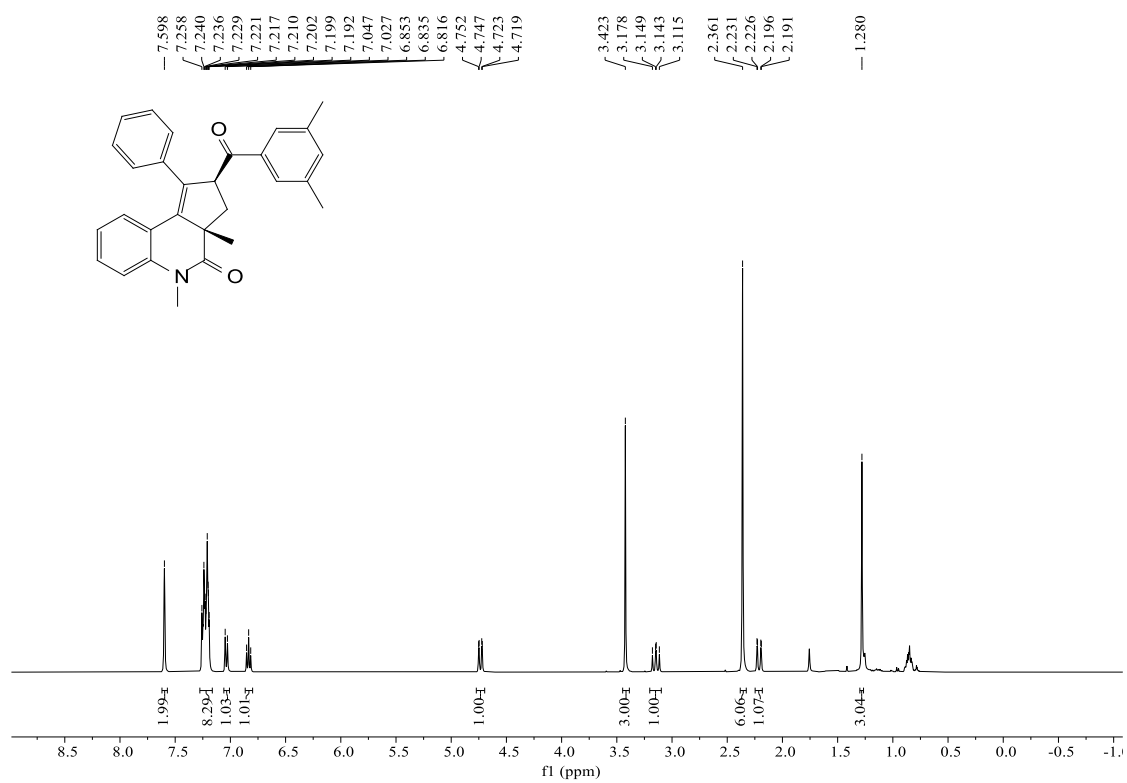
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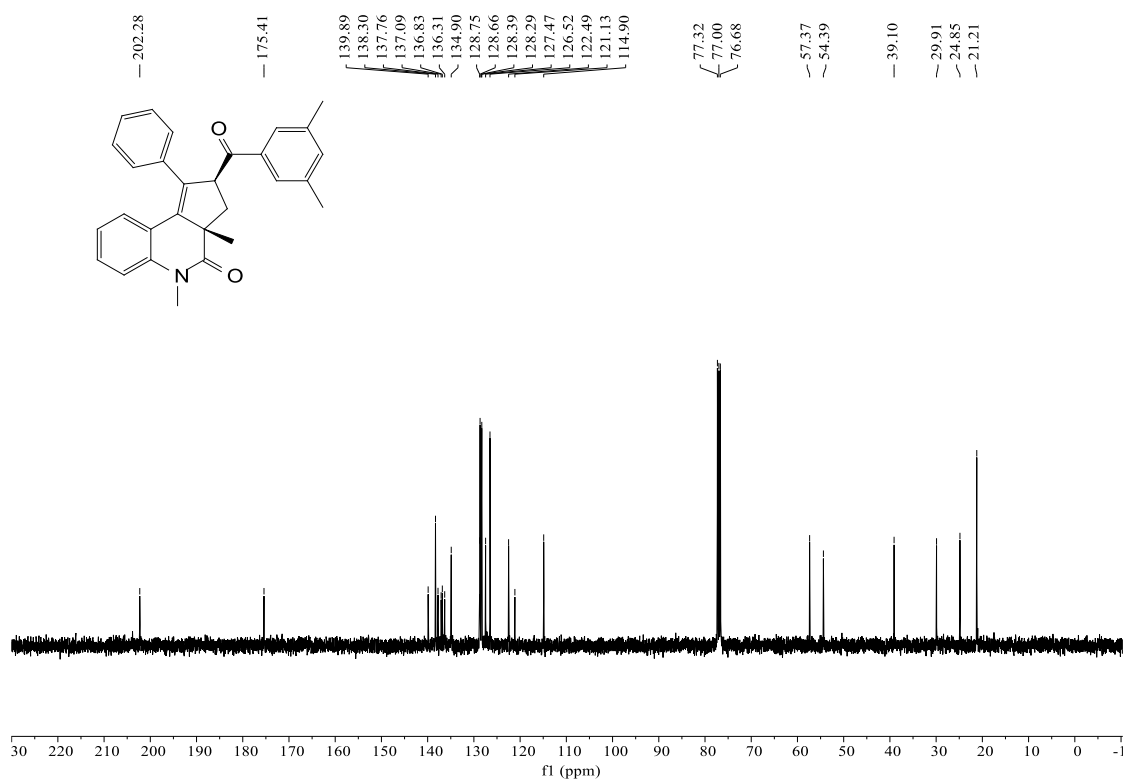
¹³C NMR spectra of *trans*-3k (100 MHz, CDCl₃)



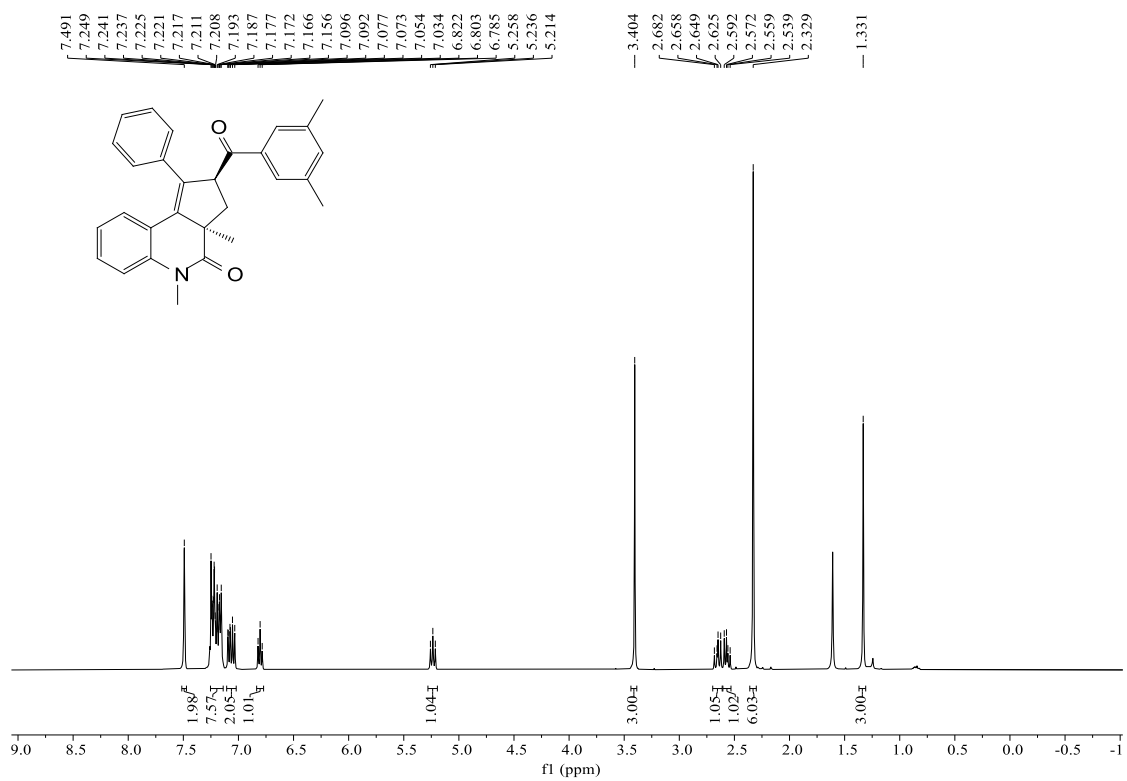
¹H NMR spectra of *cis*-3l (400 MHz, CDCl₃)



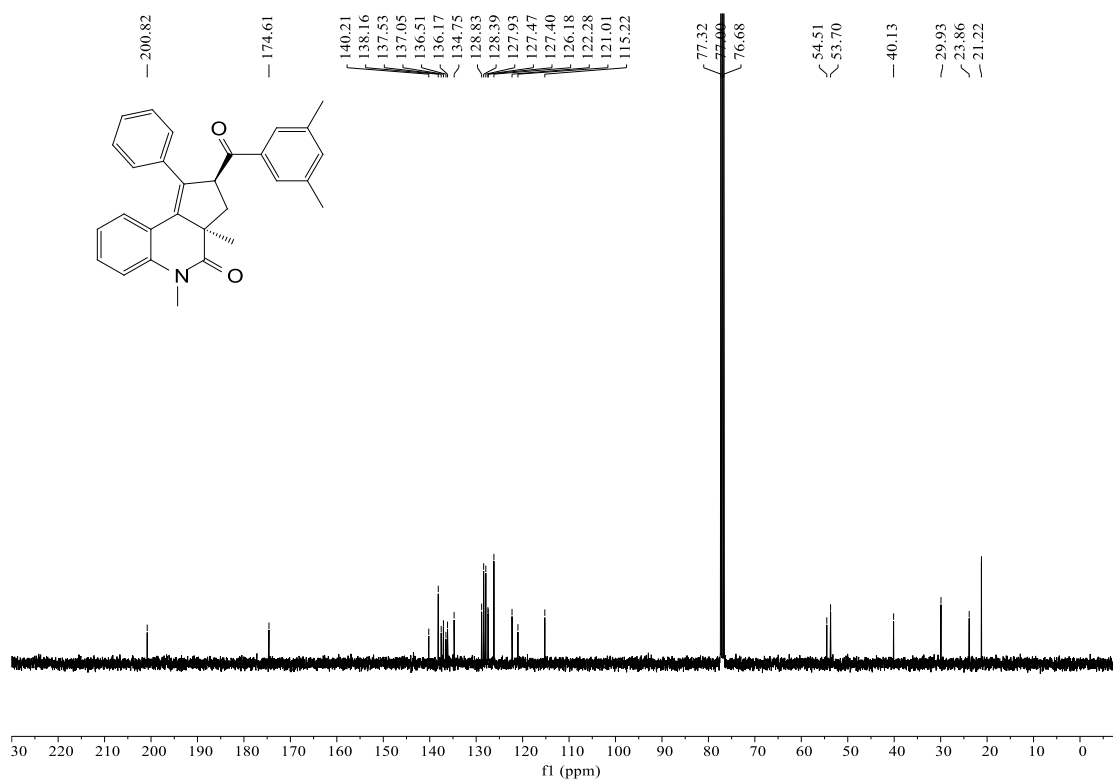
¹³C NMR spectra of *cis*-3l (100 MHz, CDCl₃)



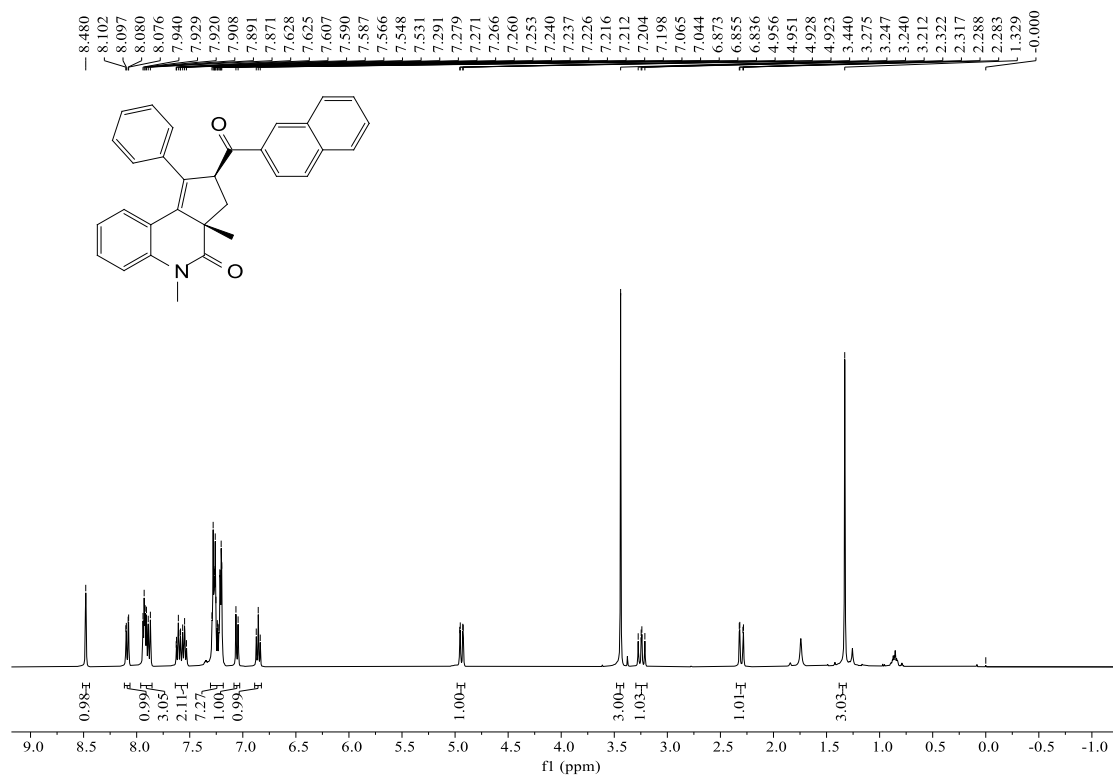
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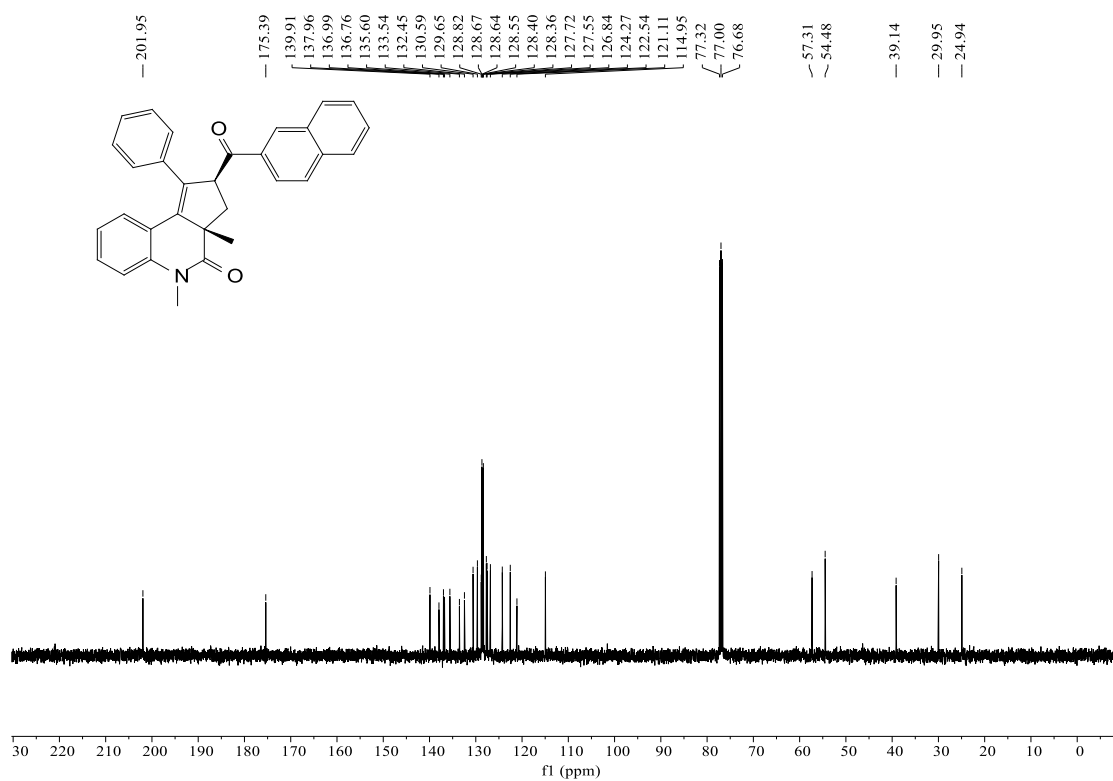
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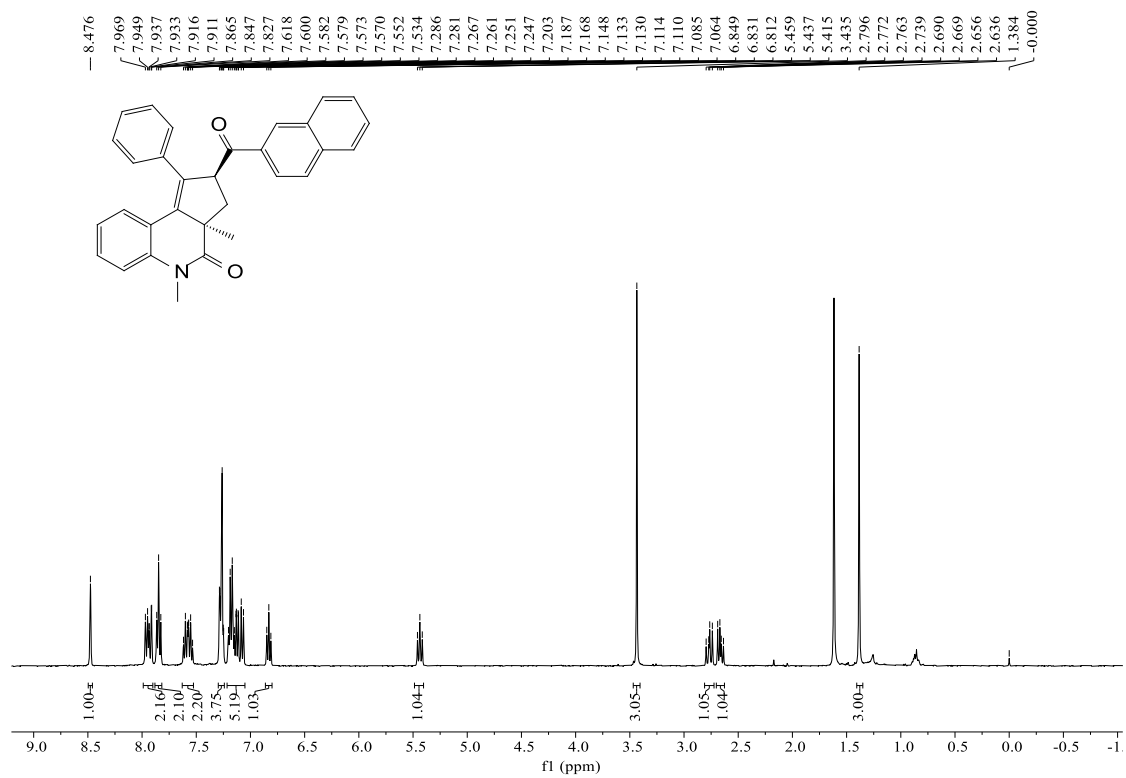
¹H NMR spectra of *cis*-3m (400 MHz, CDCl₃)



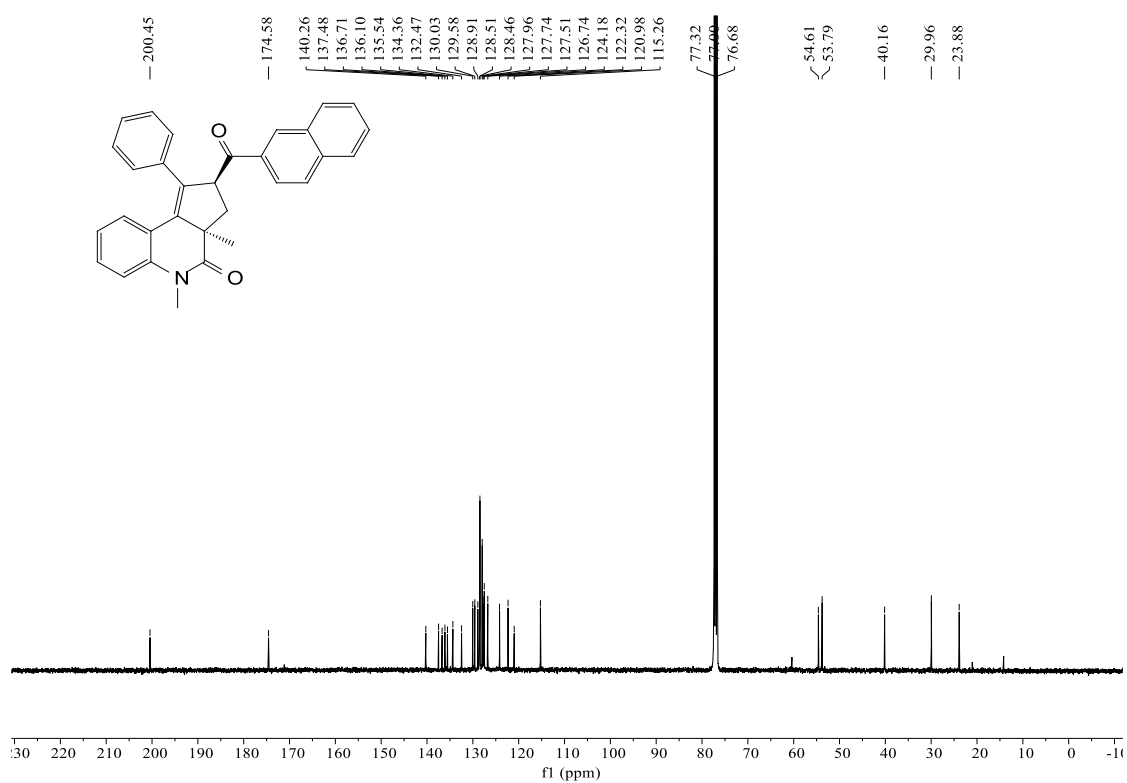
¹³C NMR spectra of *cis*-3m (100 MHz, CDCl₃)



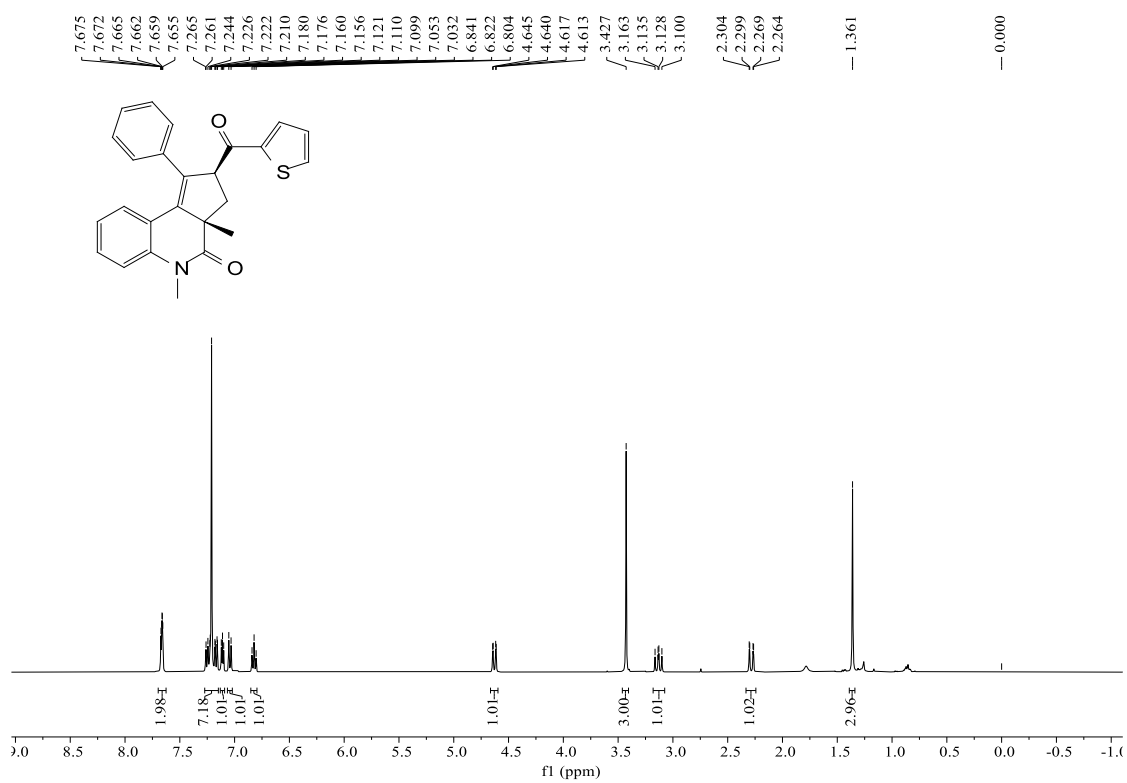
¹H NMR spectra of *trans*-3m (400 MHz, CDCl₃)



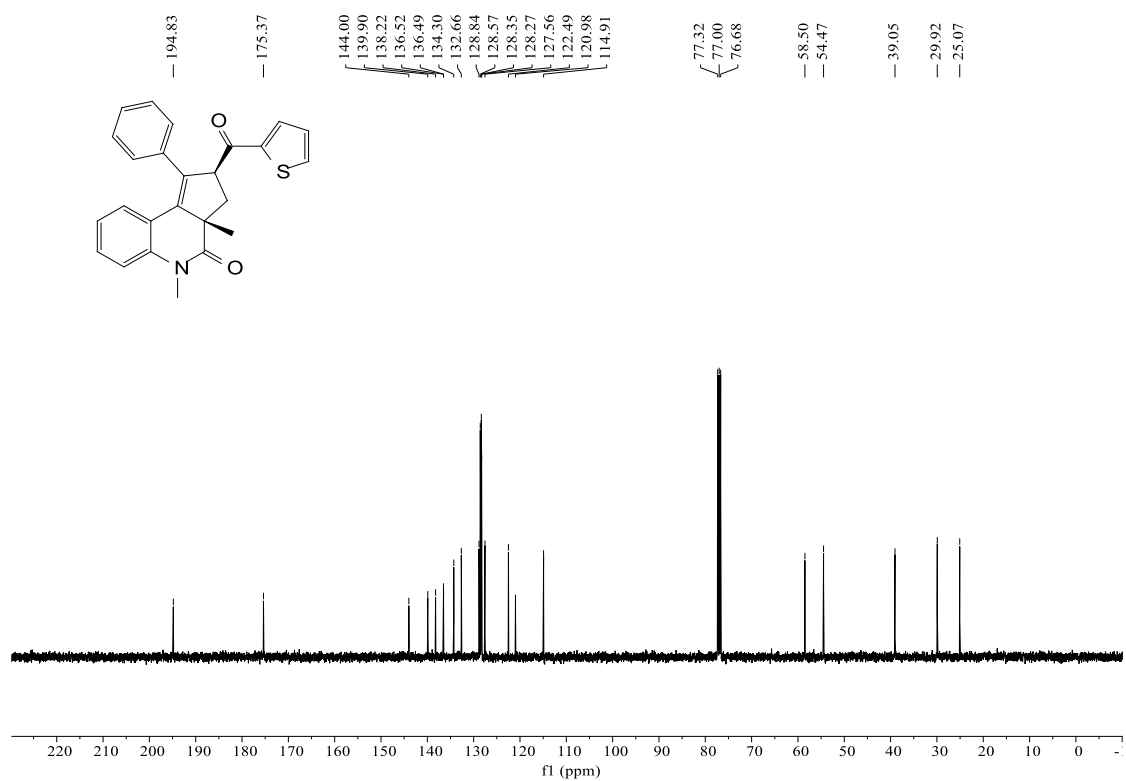
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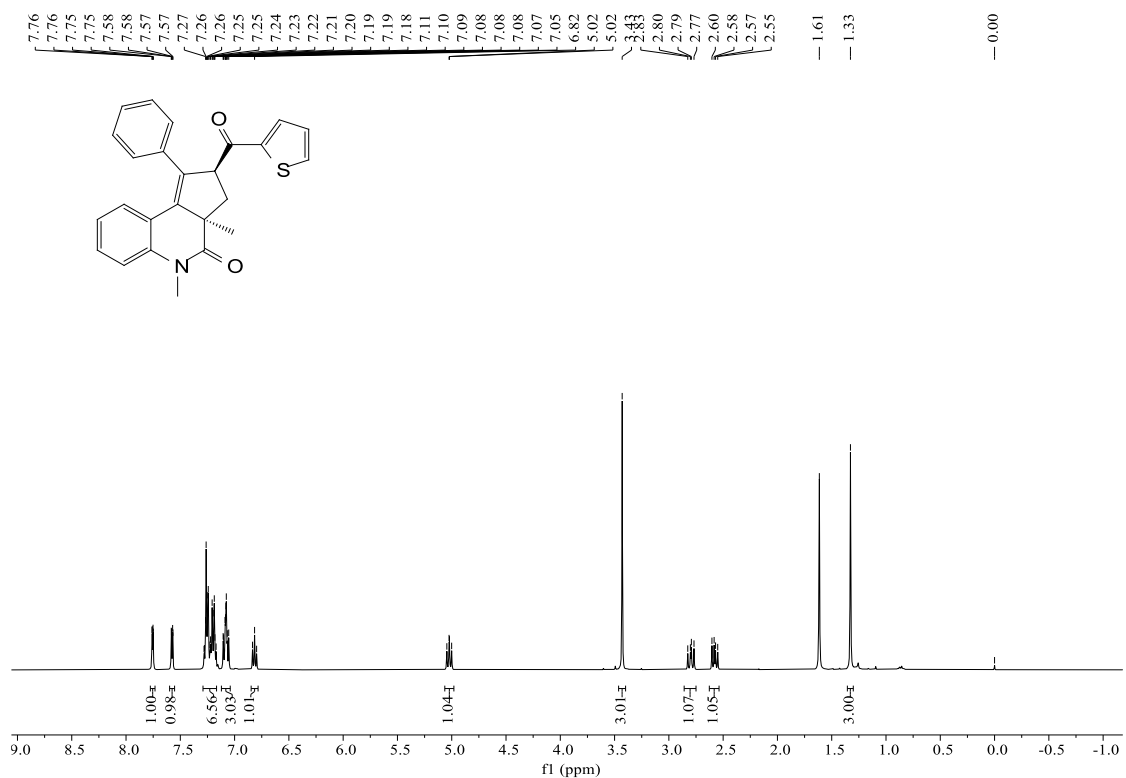
¹H NMR spectra of *cis*-3n (400 MHz, CDCl₃)



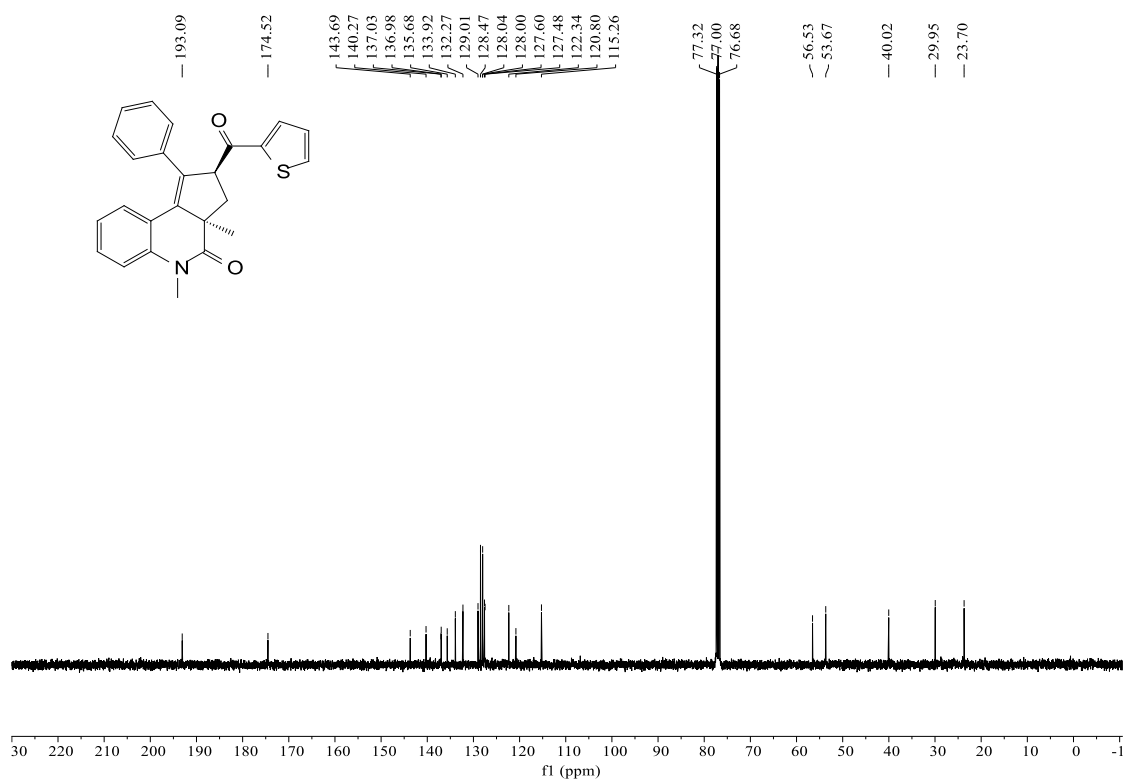
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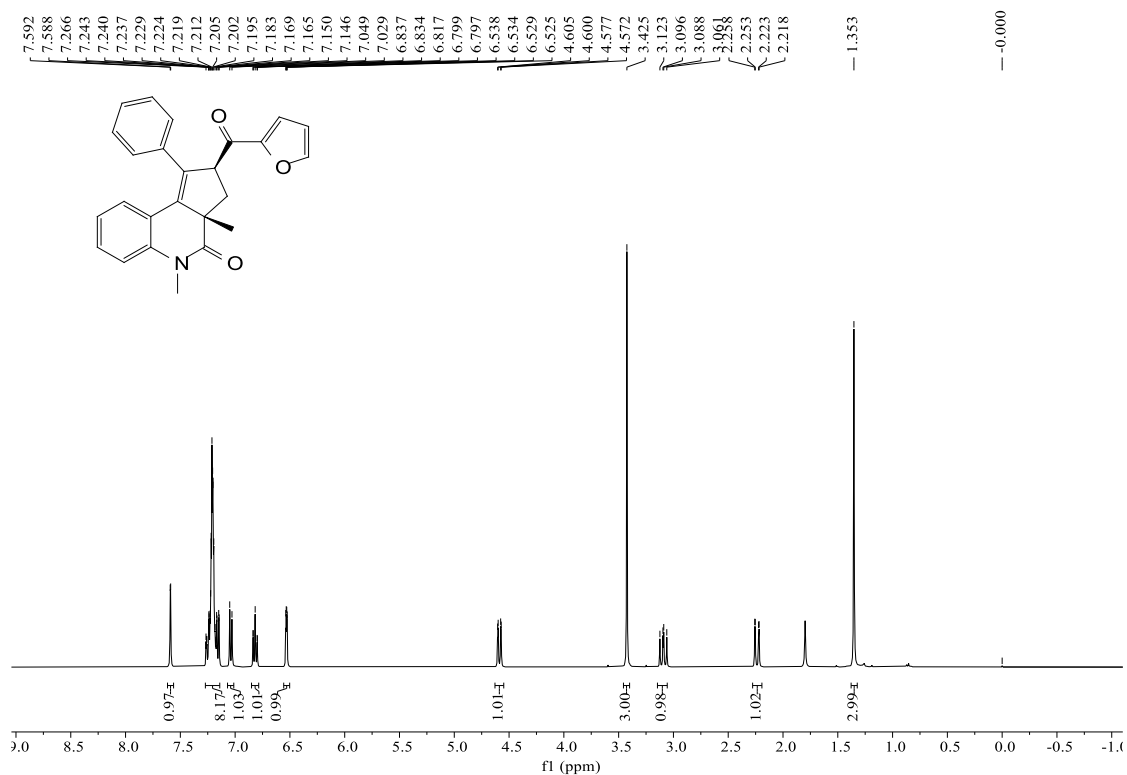
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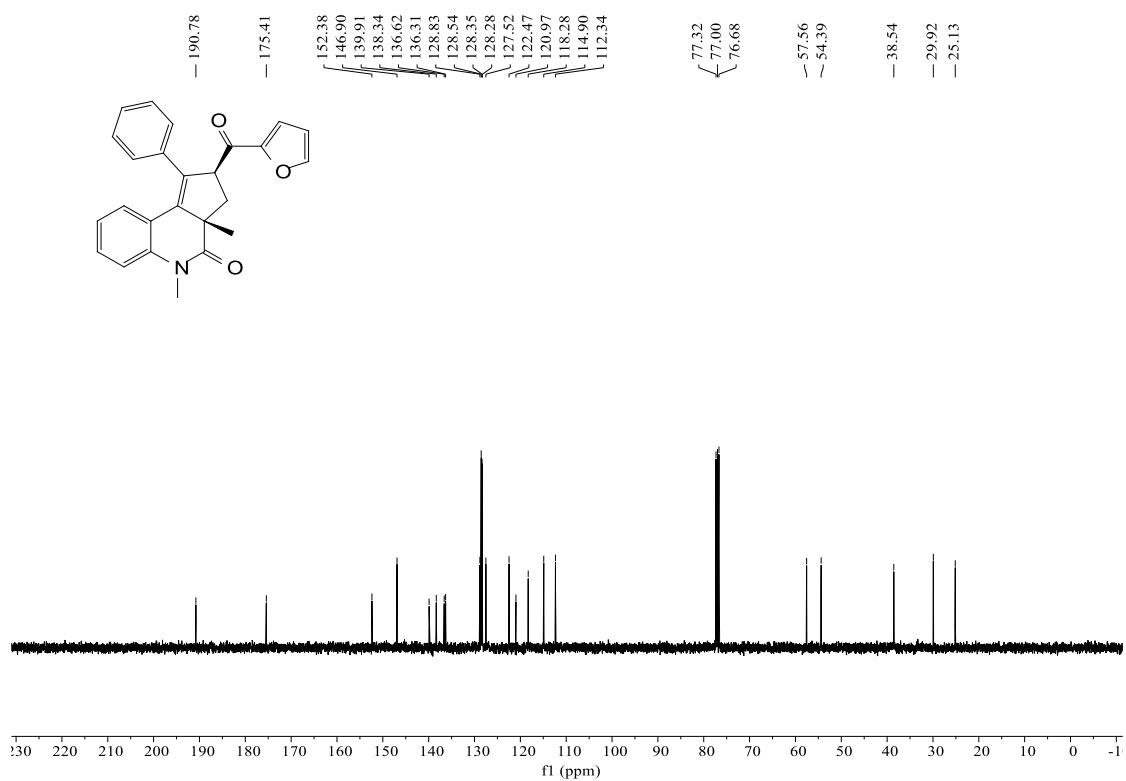
¹³C NMR spectra of *trans*-3n (100 MHz, CDCl₃)



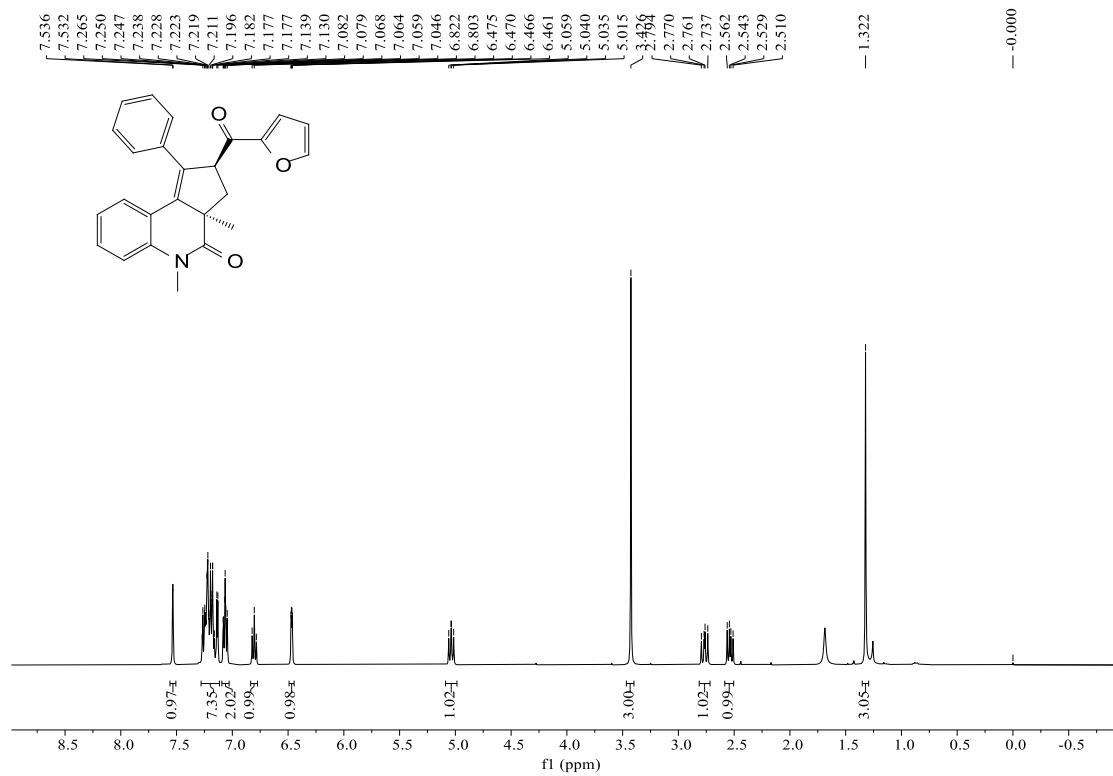
¹H NMR spectra of *cis*-3o (400 MHz, CDCl₃)



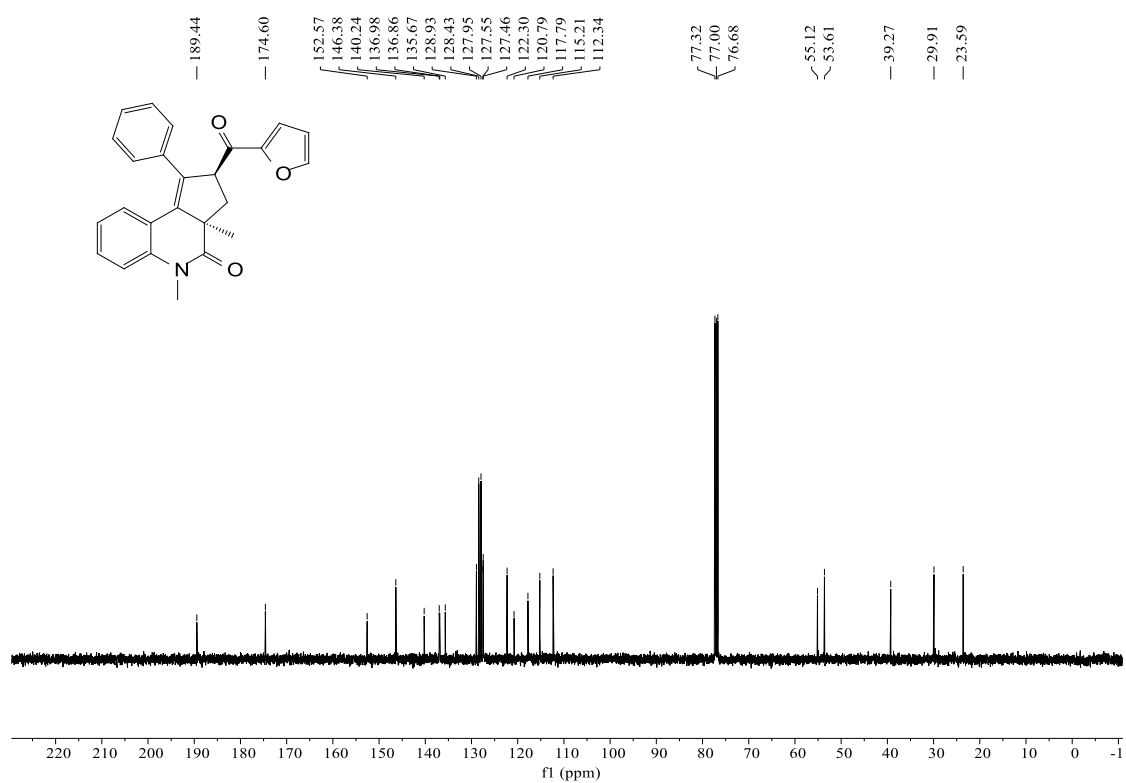
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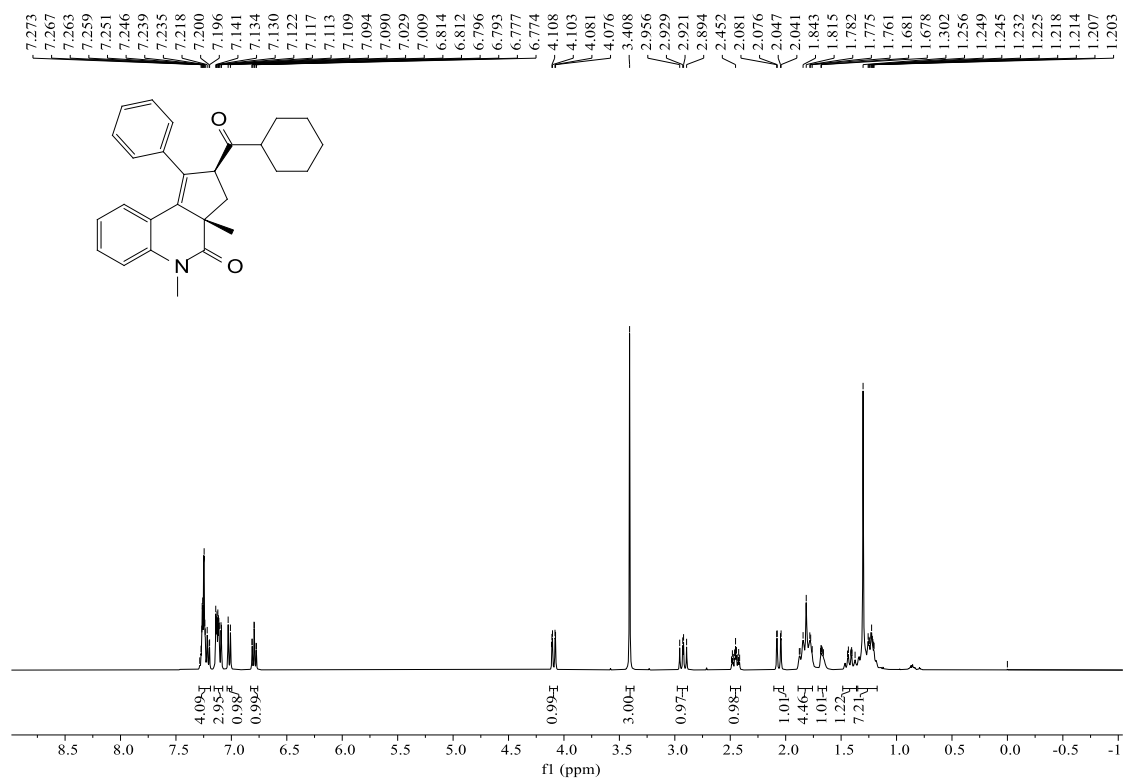
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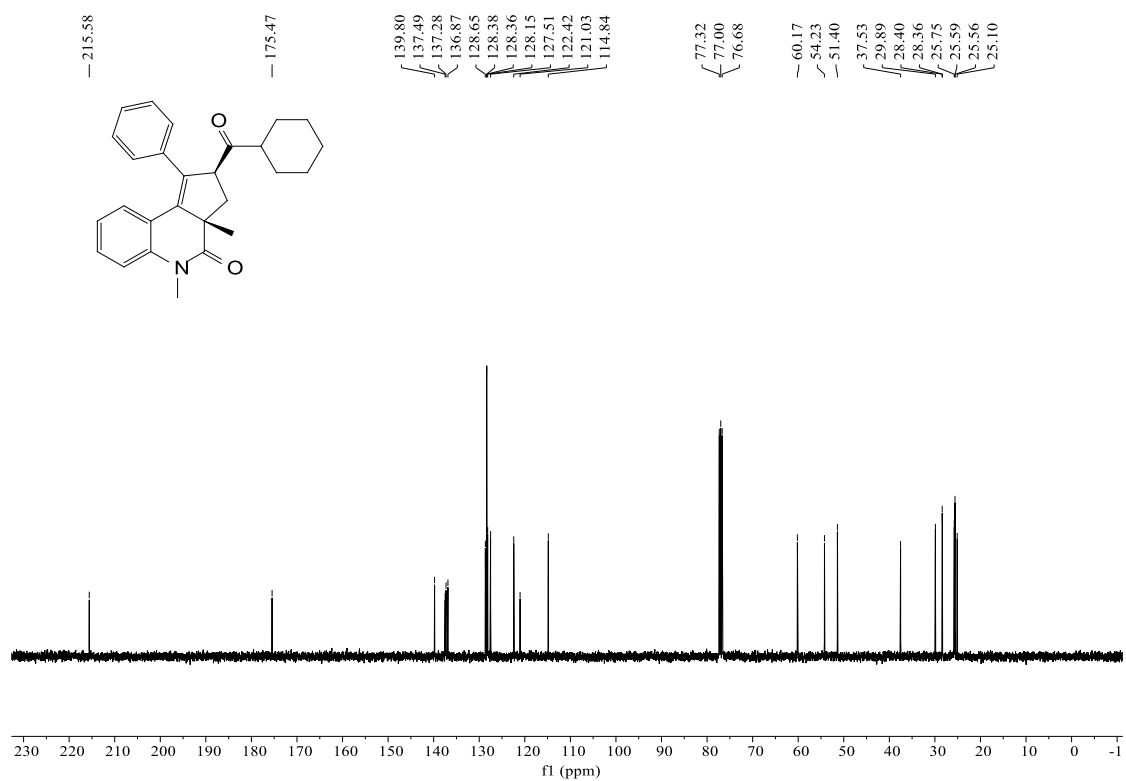
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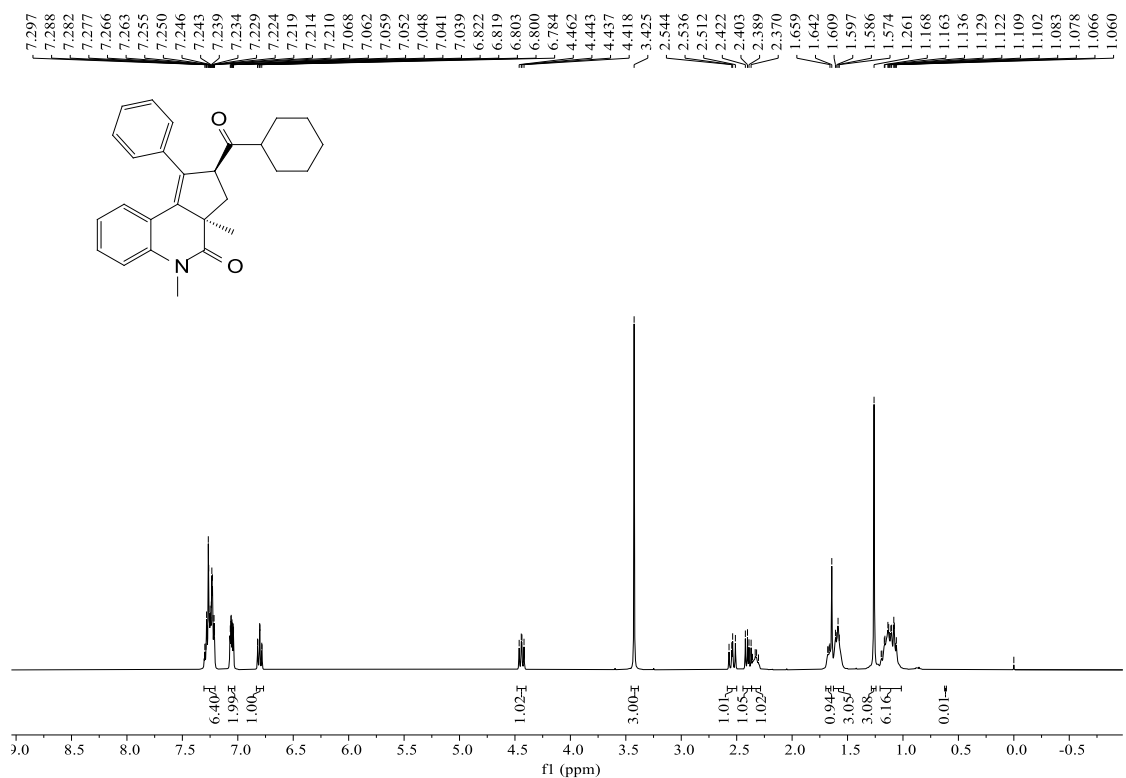
¹H NMR spectra of *cis*-3p (400 MHz, CDCl₃)



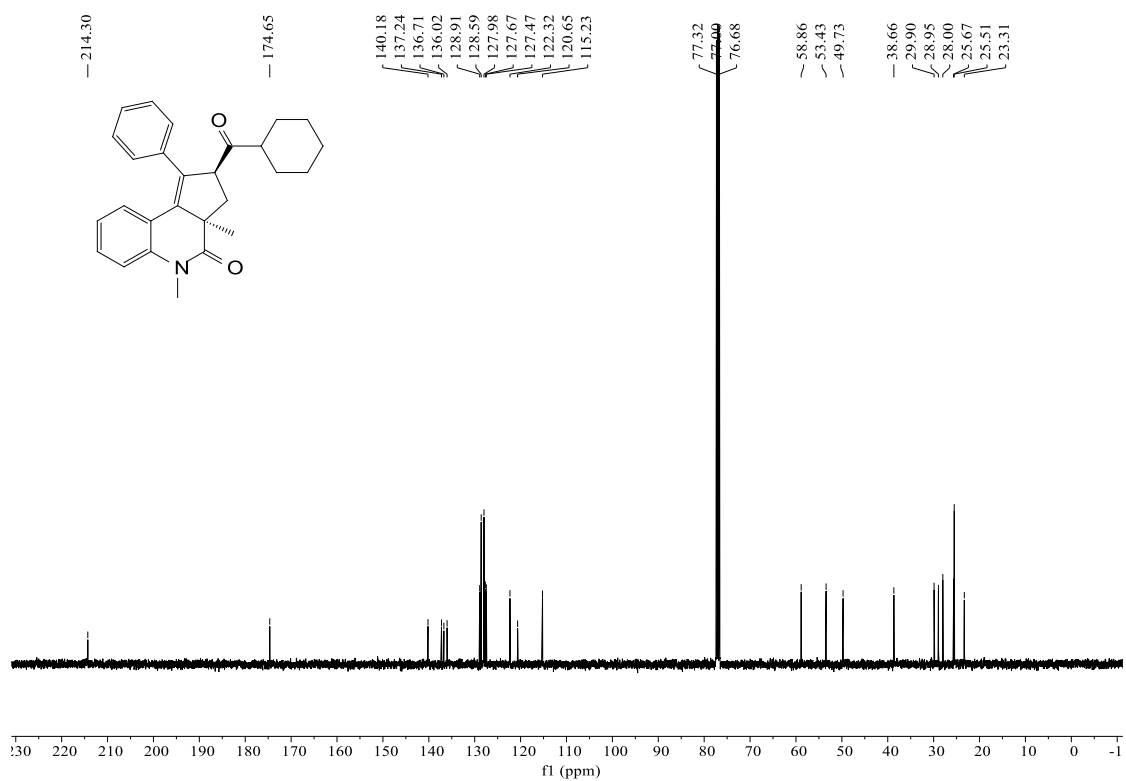
¹³C NMR spectra of *cis*-3p (100 MHz, CDCl₃)



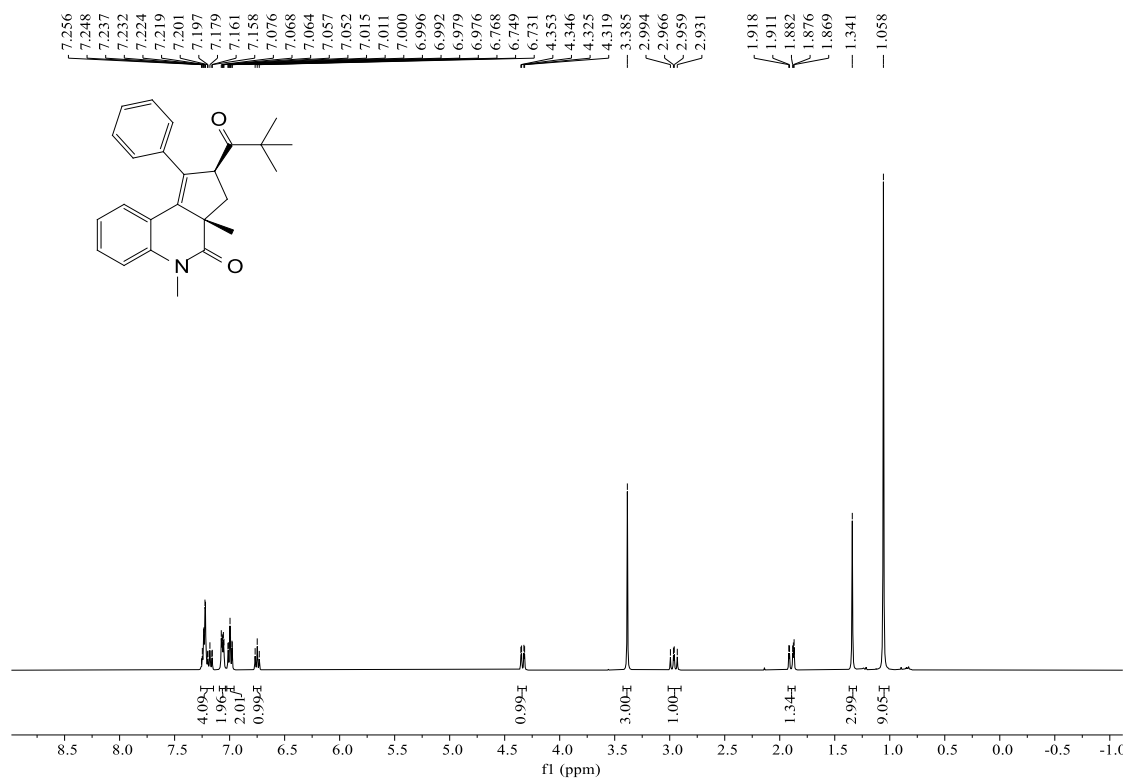
¹H NMR spectra of *trans*-3p (400 MHz, CDCl₃)



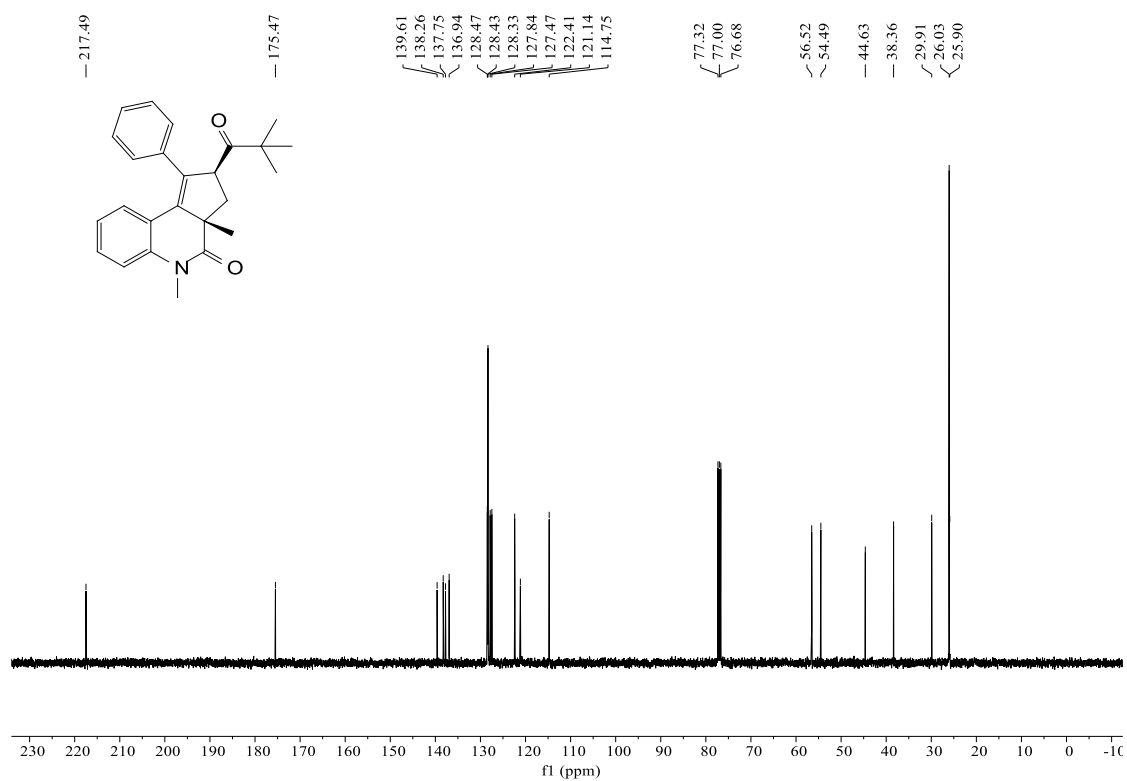
¹³C NMR spectra of *trans*-3p (100 MHz, CDCl₃)



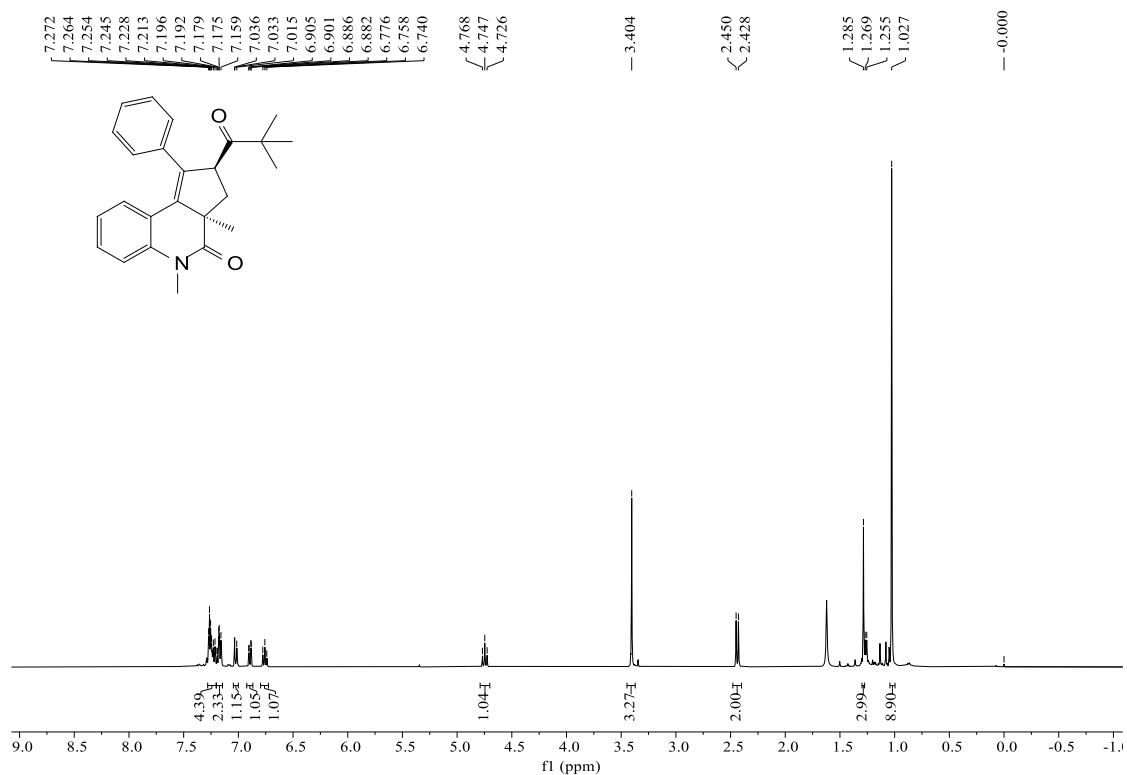
¹H NMR spectra of *cis*-3q (400 MHz, CDCl₃)



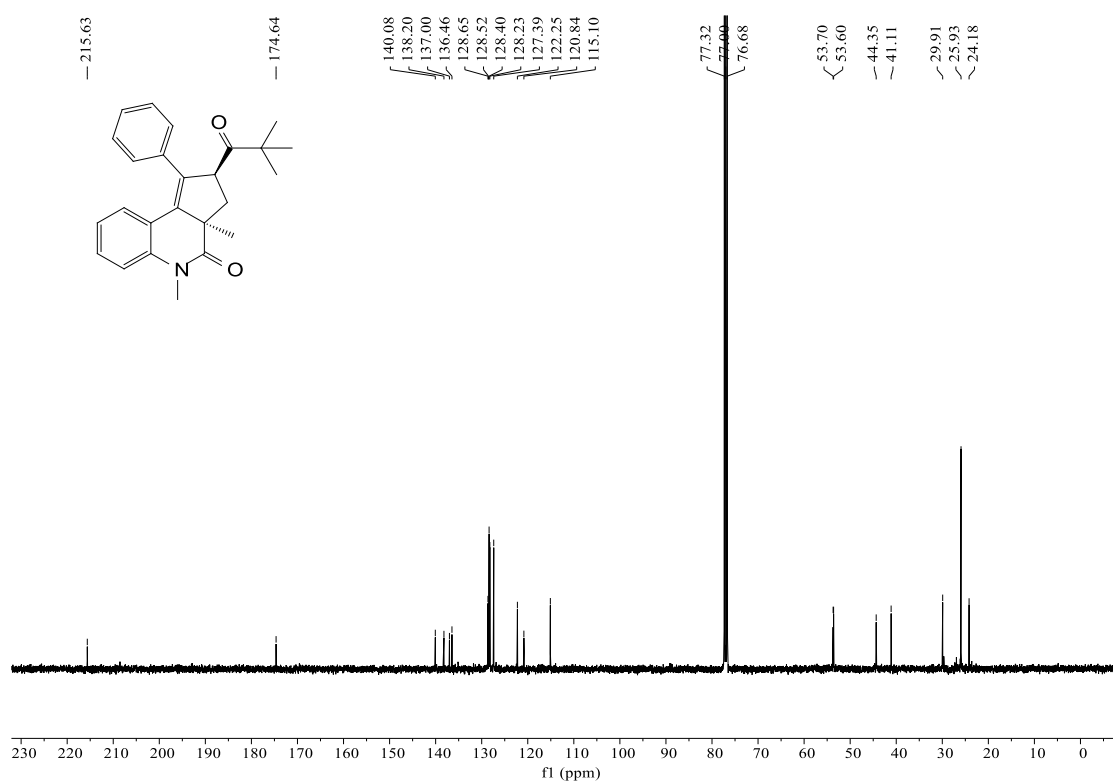
¹³C NMR spectra of *cis*-3q (100 MHz, CDCl₃)



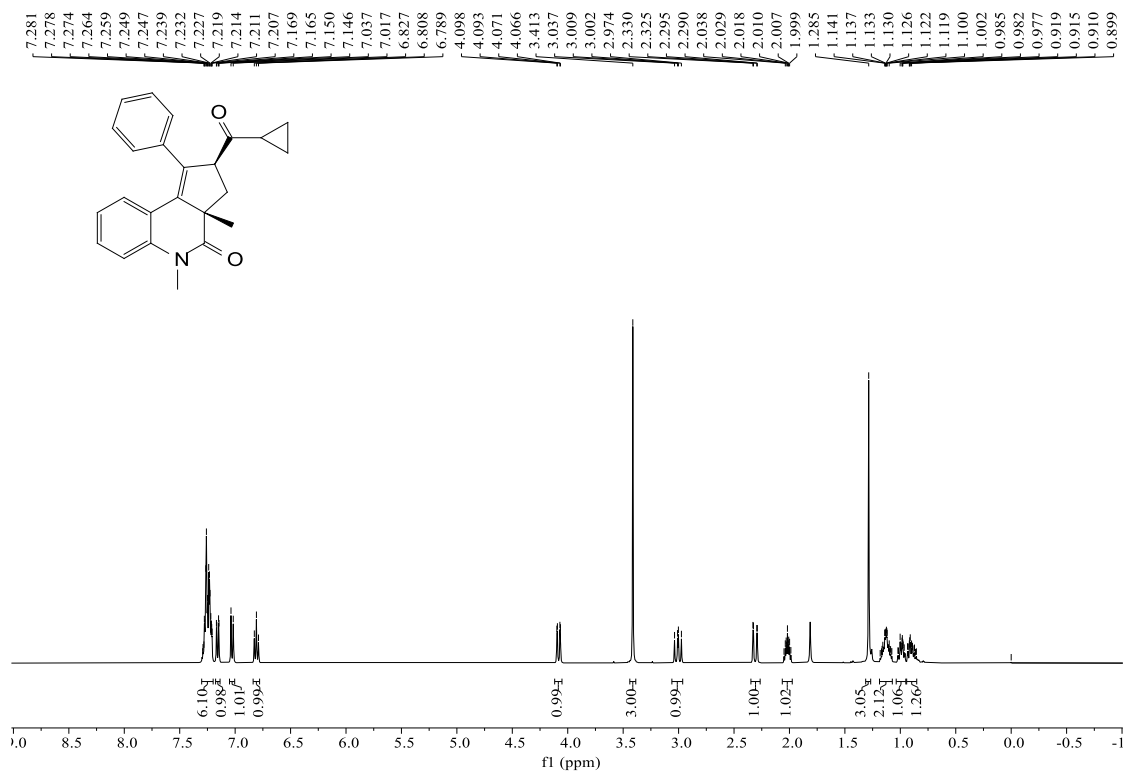
¹H NMR spectra of *trans*-3q (400 MHz, CDCl₃)



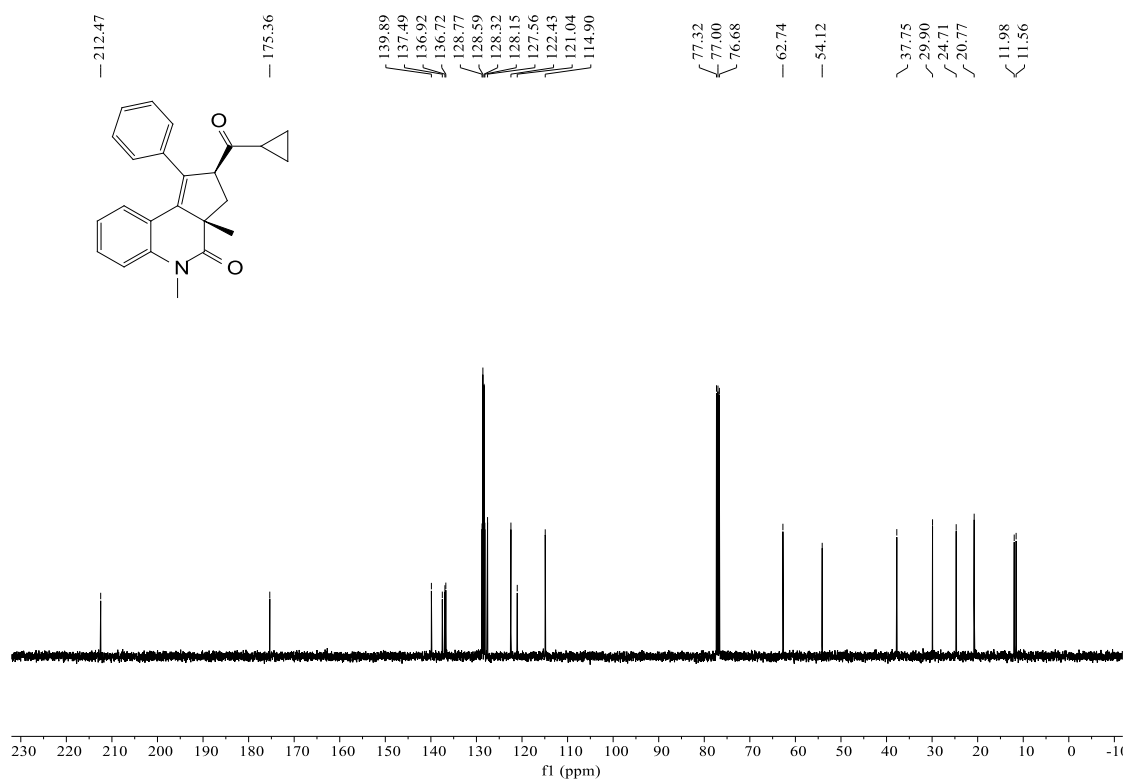
¹³C NMR spectra of *trans*-3q (100 MHz, CDCl₃)



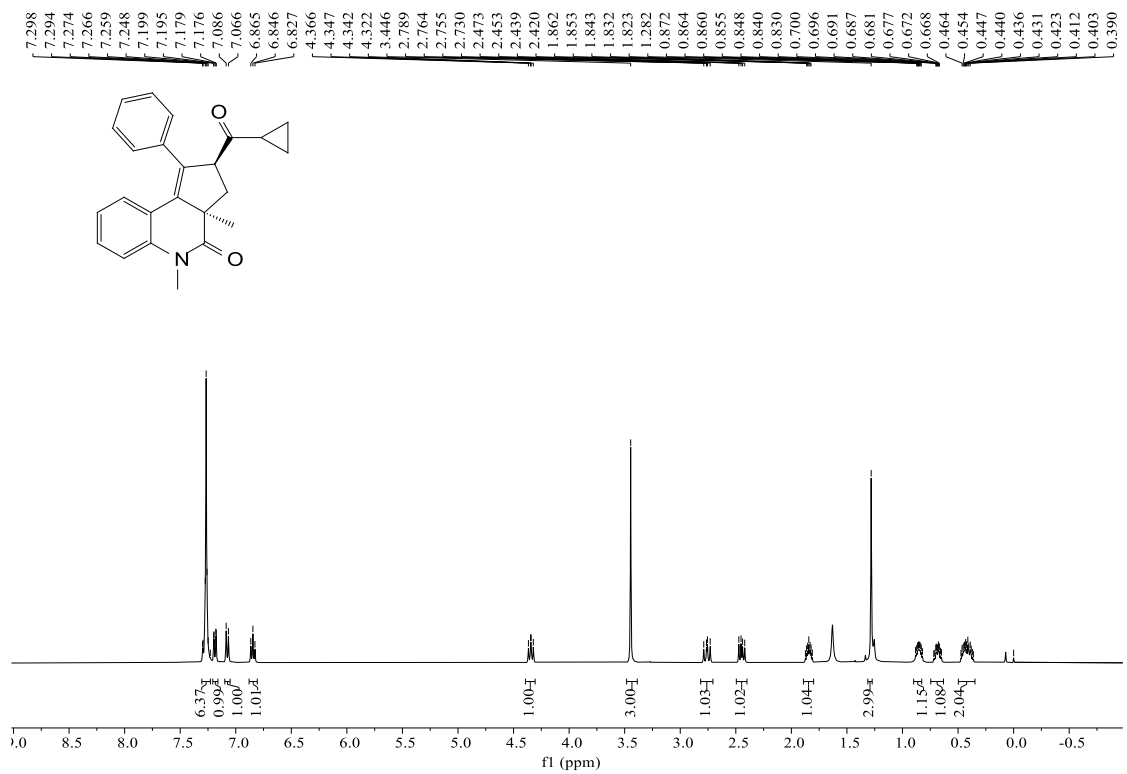
¹H NMR spectra of *cis*-3r (400 MHz, CDCl₃)



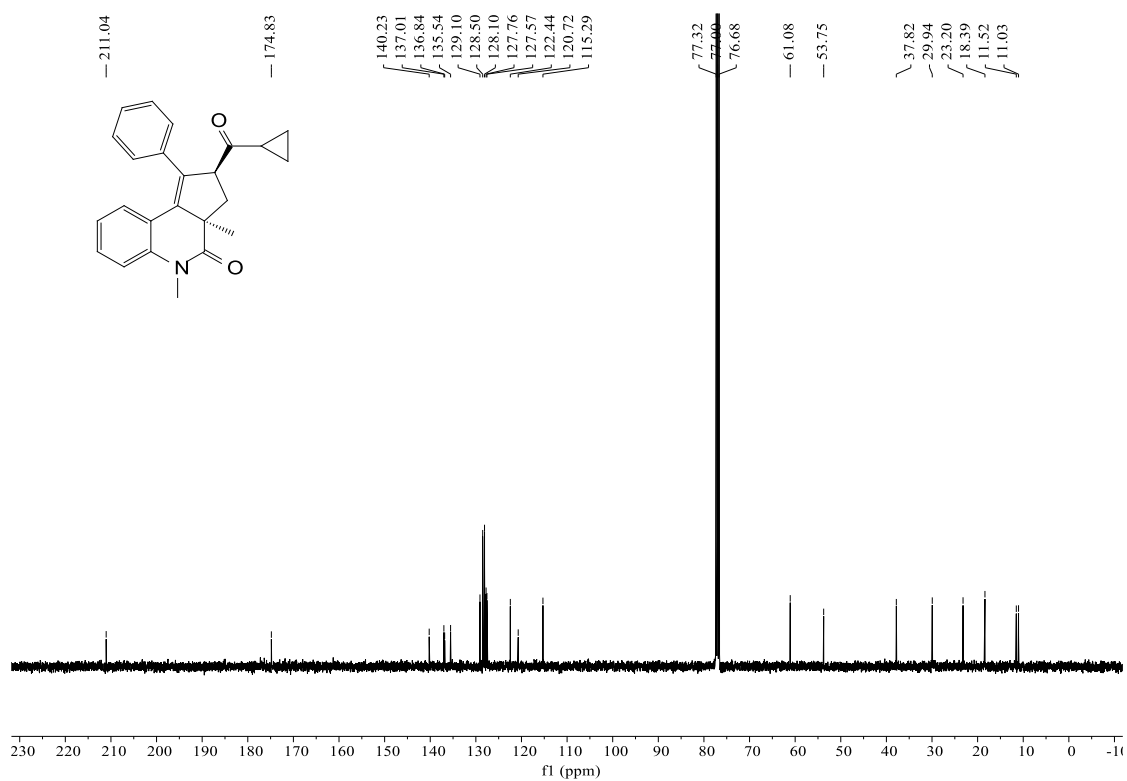
¹³C NMR spectra of *cis*-3r (100 MHz, CDCl₃)



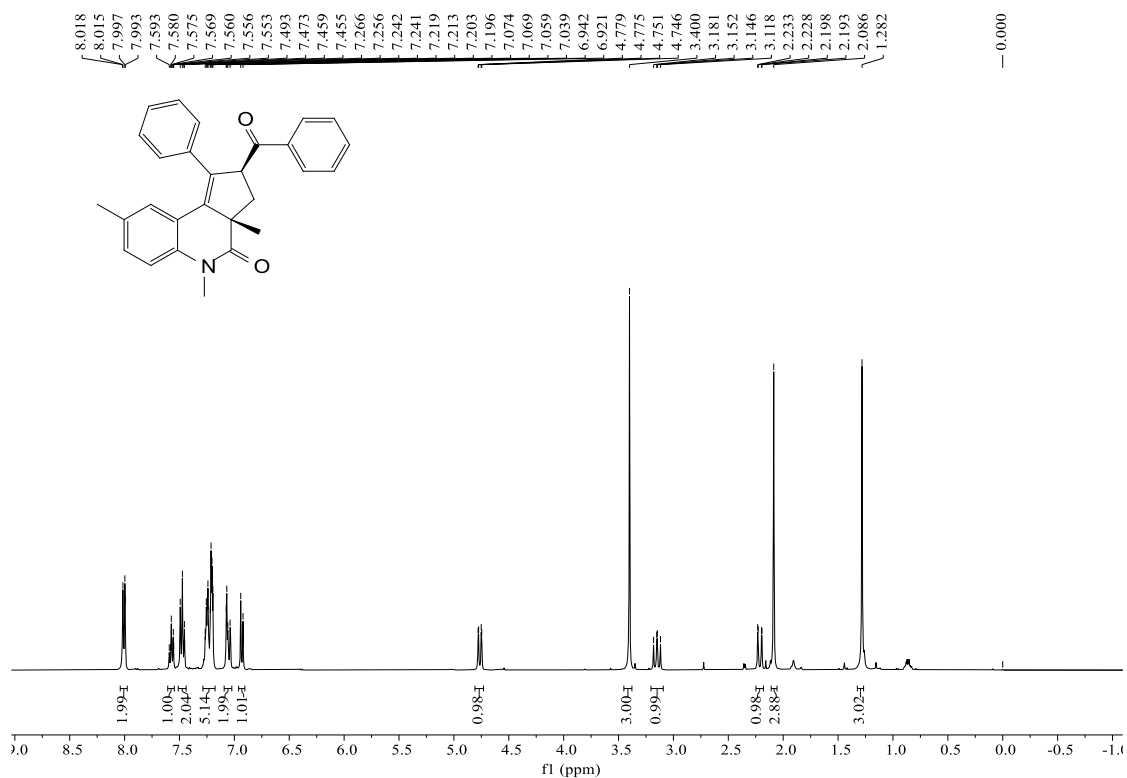
¹H NMR spectra of *trans*-3r (400 MHz, CDCl₃)



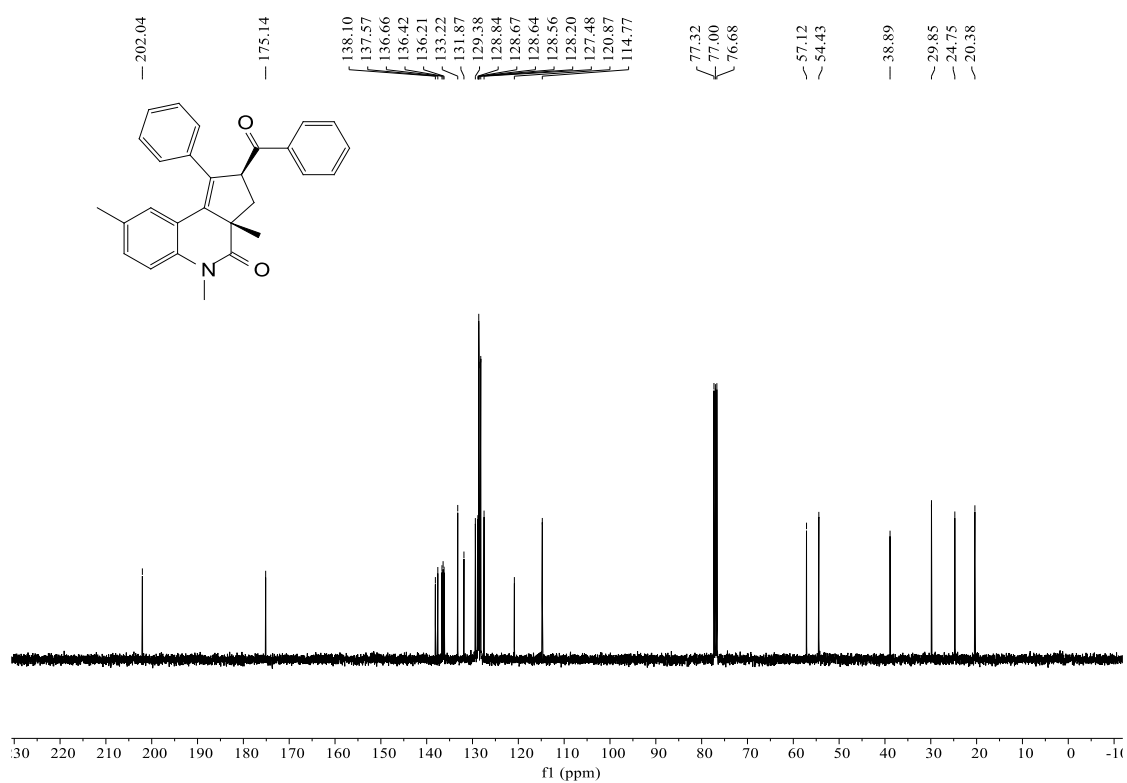
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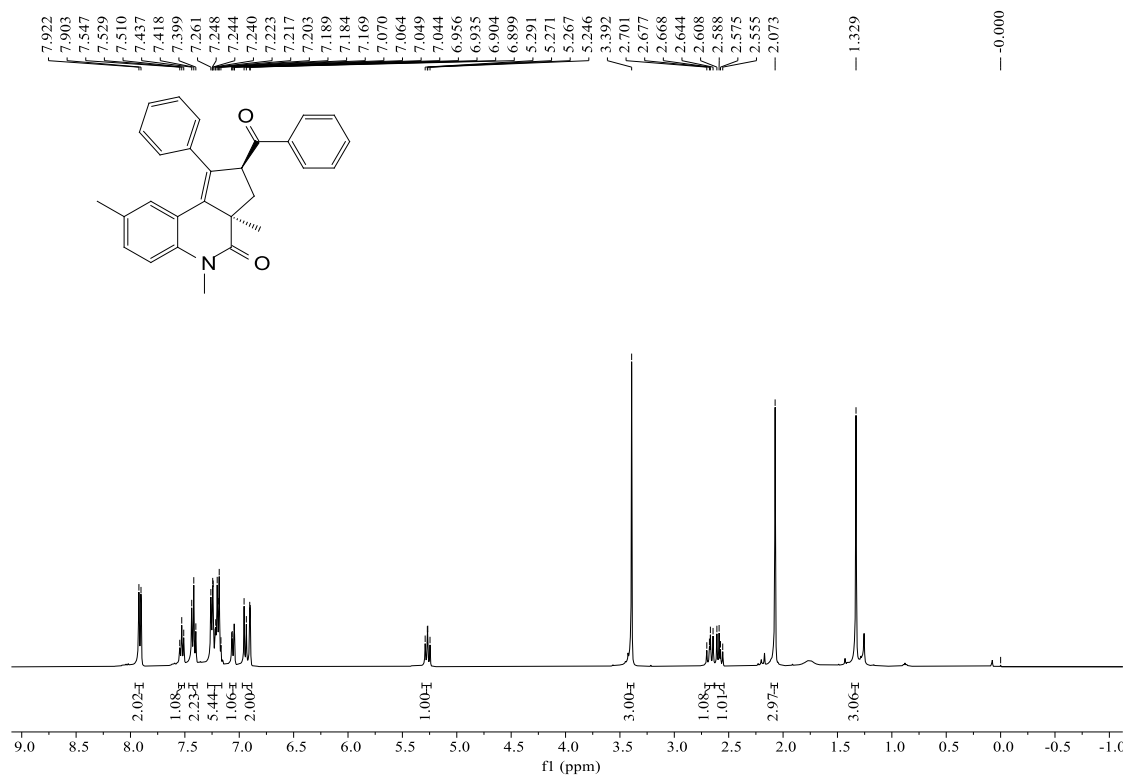
¹H NMR spectra of *cis*-3s (400 MHz, CDCl₃)



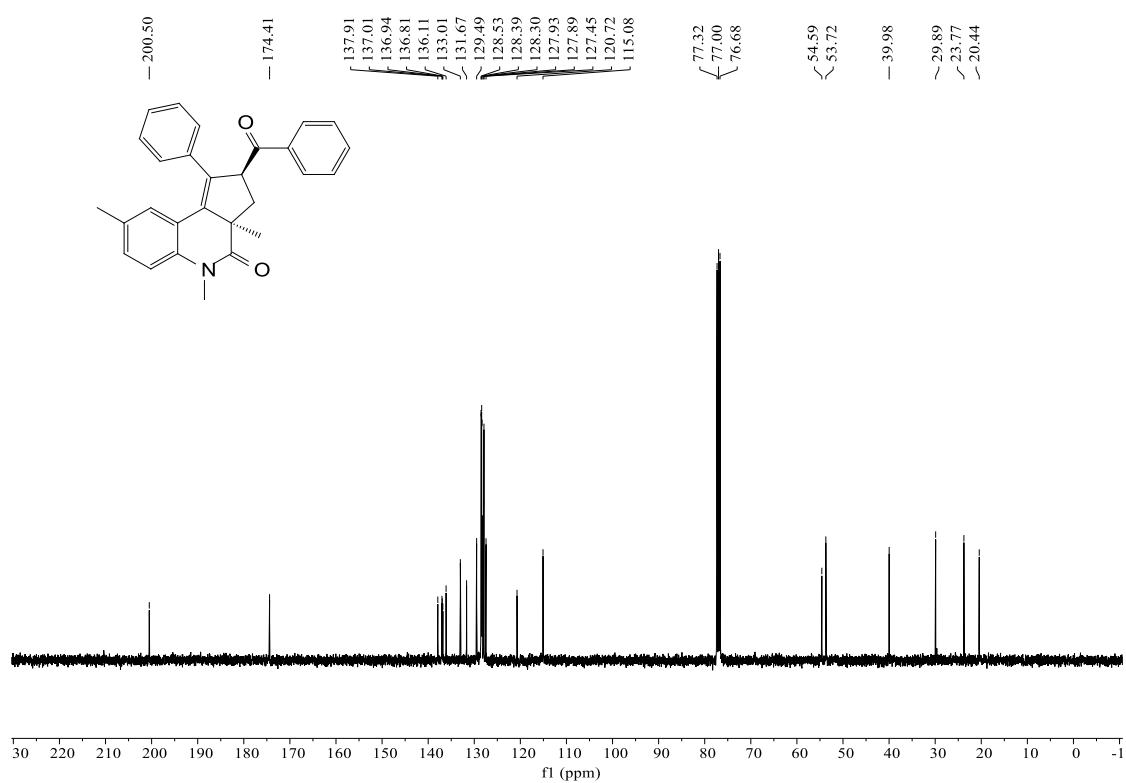
¹³C NMR spectra of *cis*-3s (100 MHz, CDCl₃)



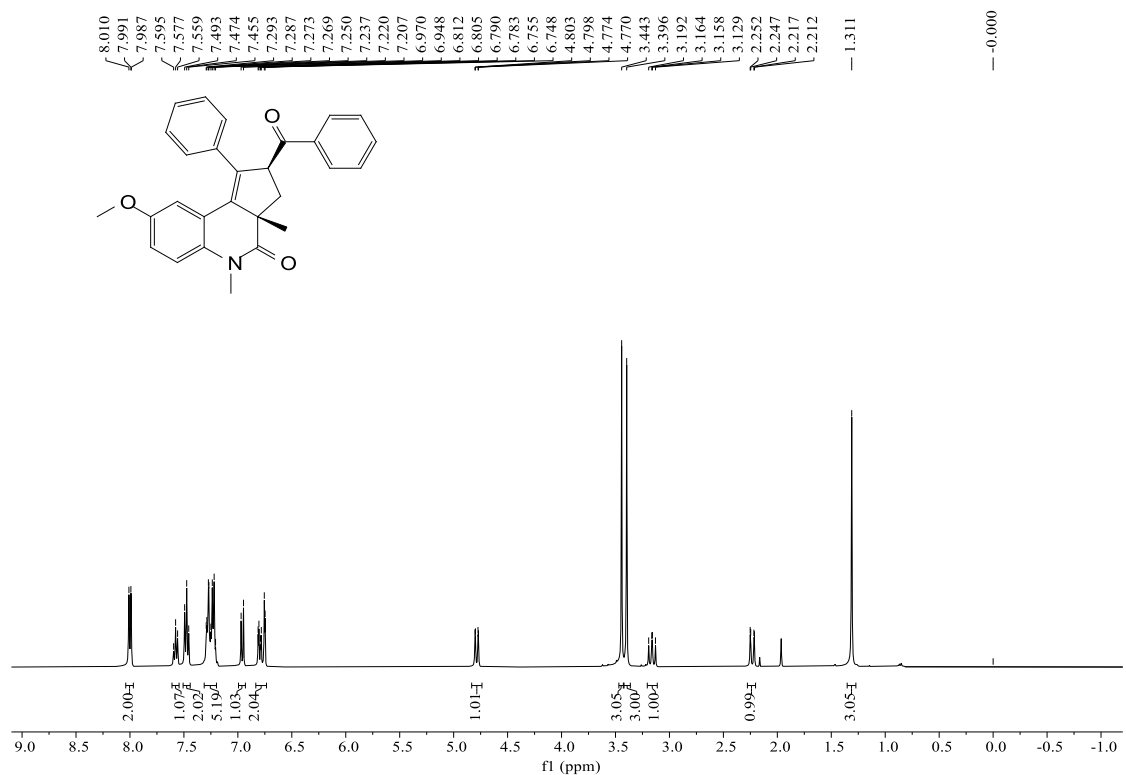
¹H NMR spectra of *trans*-3s (400 MHz, CDCl₃)



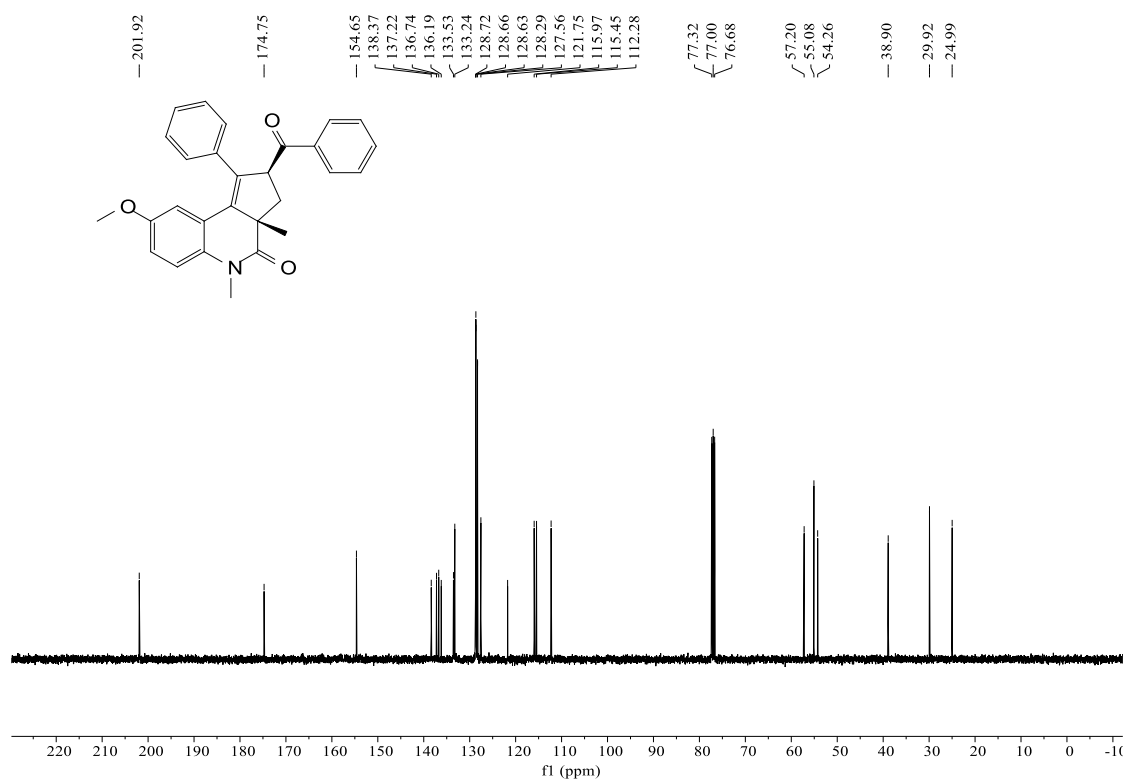
¹³C NMR spectra of *trans*-3s (100 MHz, CDCl₃)



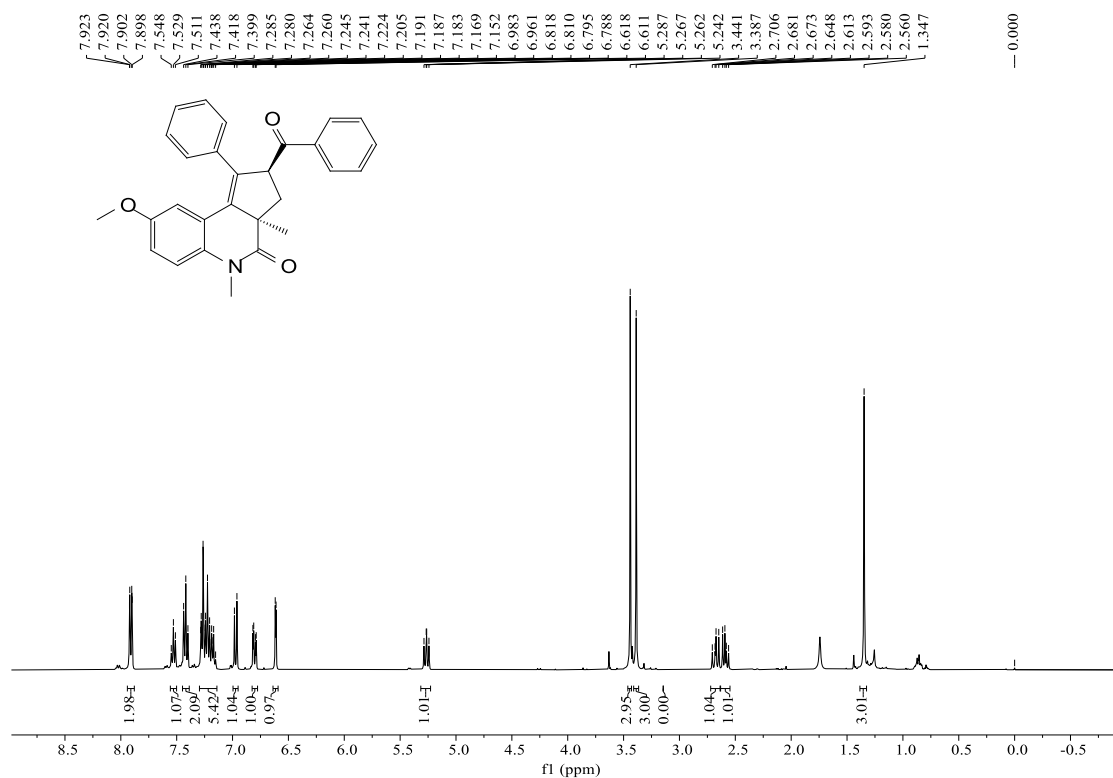
¹H NMR spectra of *cis*-3t (400 MHz, CDCl₃)



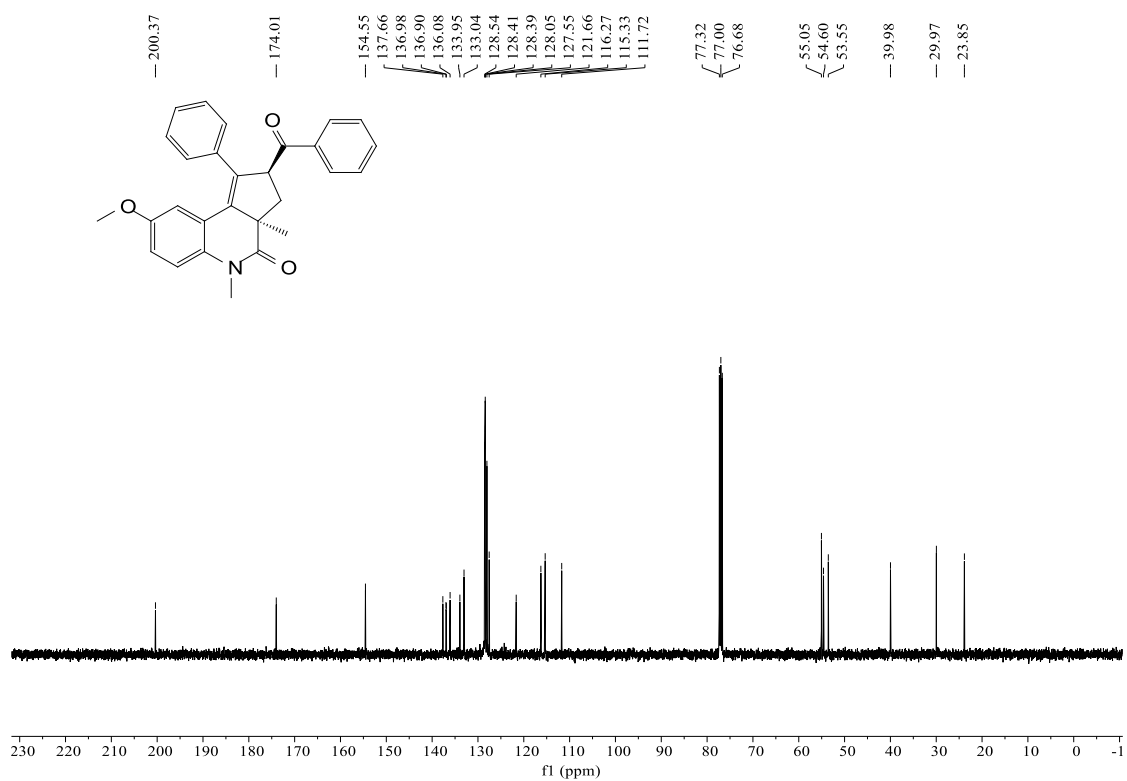
¹³C NMR spectra of *cis*-3t (100 MHz, CDCl₃)



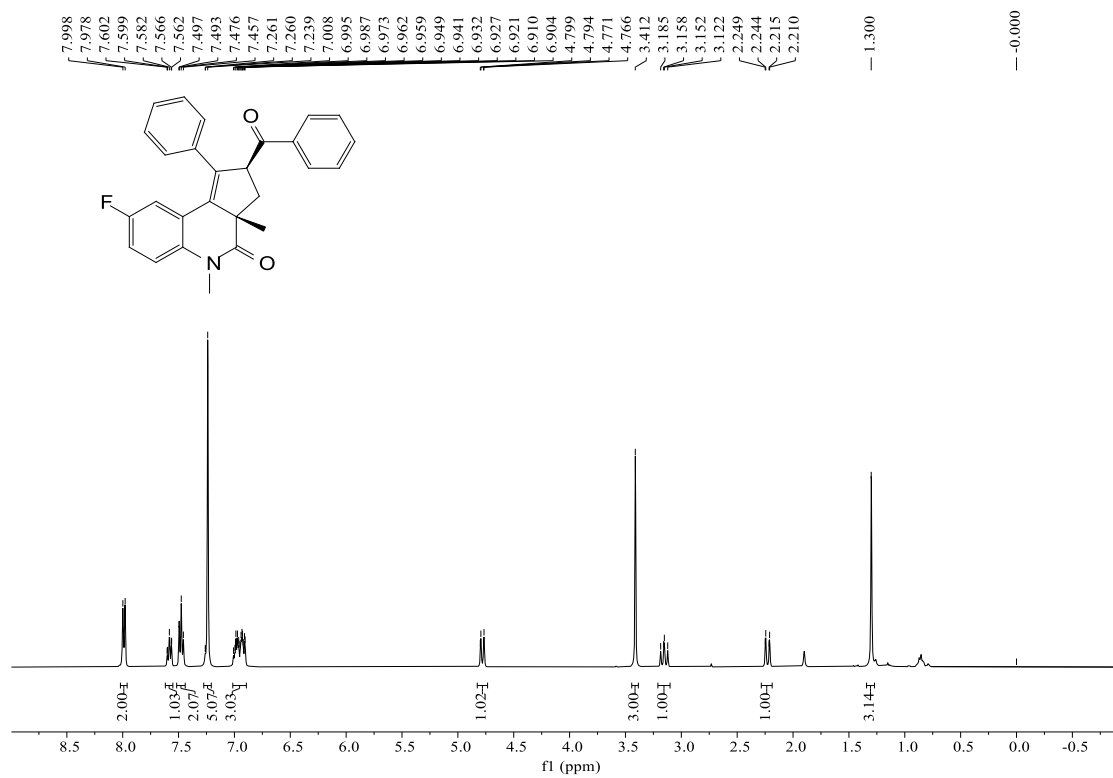
¹H NMR spectra of *trans*-3t (400 MHz, CDCl₃)



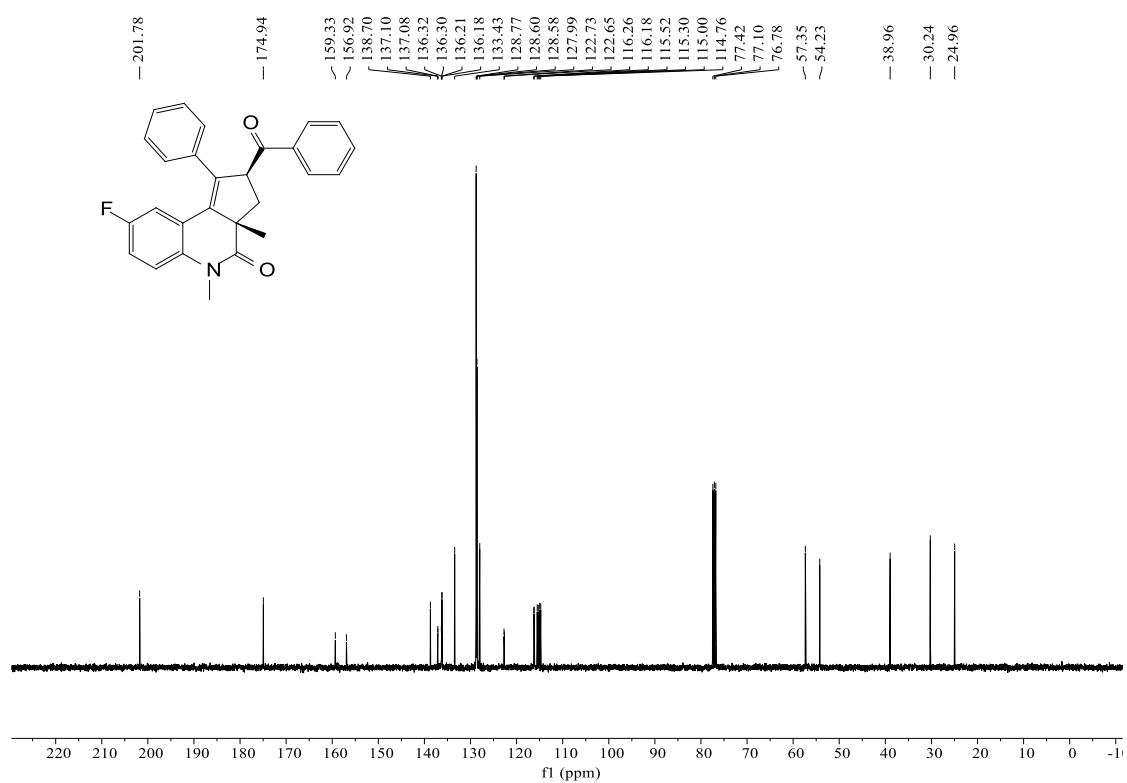
¹³C NMR spectra of *trans*-3t (100 MHz, CDCl₃)



¹H NMR spectra of *cis*-3u (400 MHz, CDCl₃)



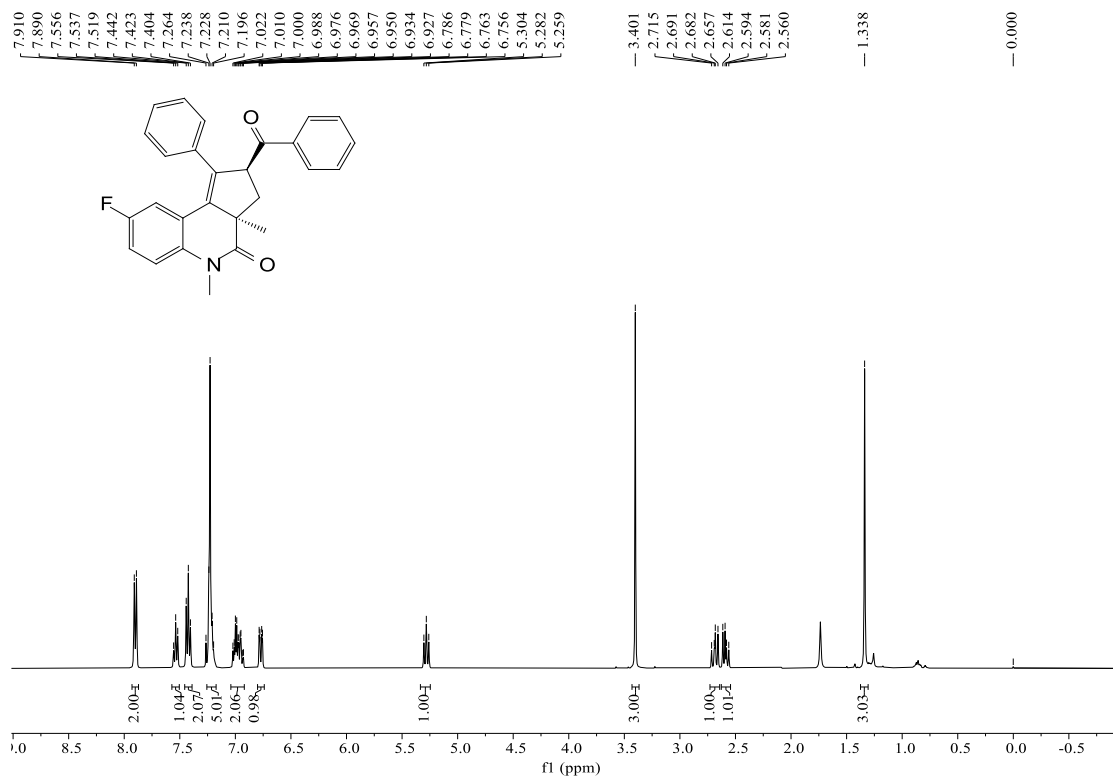
¹³C NMR spectra of *cis*-3u (100 MHz, CDCl₃)



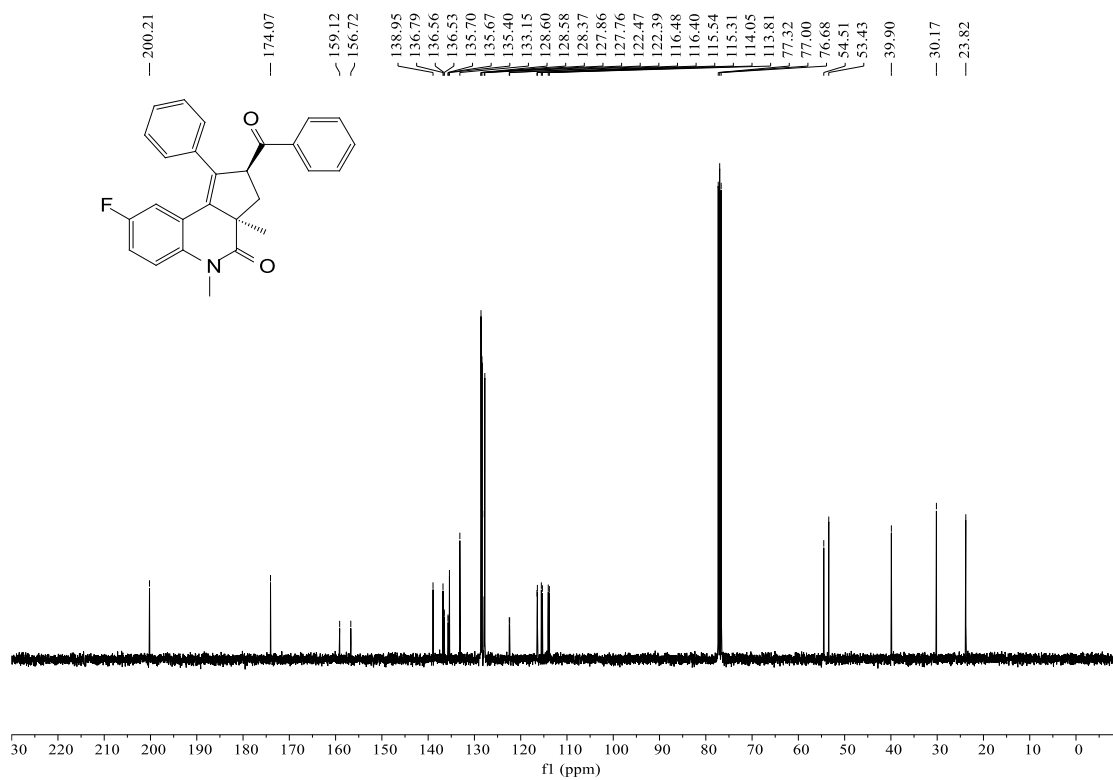
¹⁹F NMR spectra of *cis*-3u (376 MHz, CDCl₃)



¹H NMR spectra of *trans*-3u (400 MHz, CDCl₃)



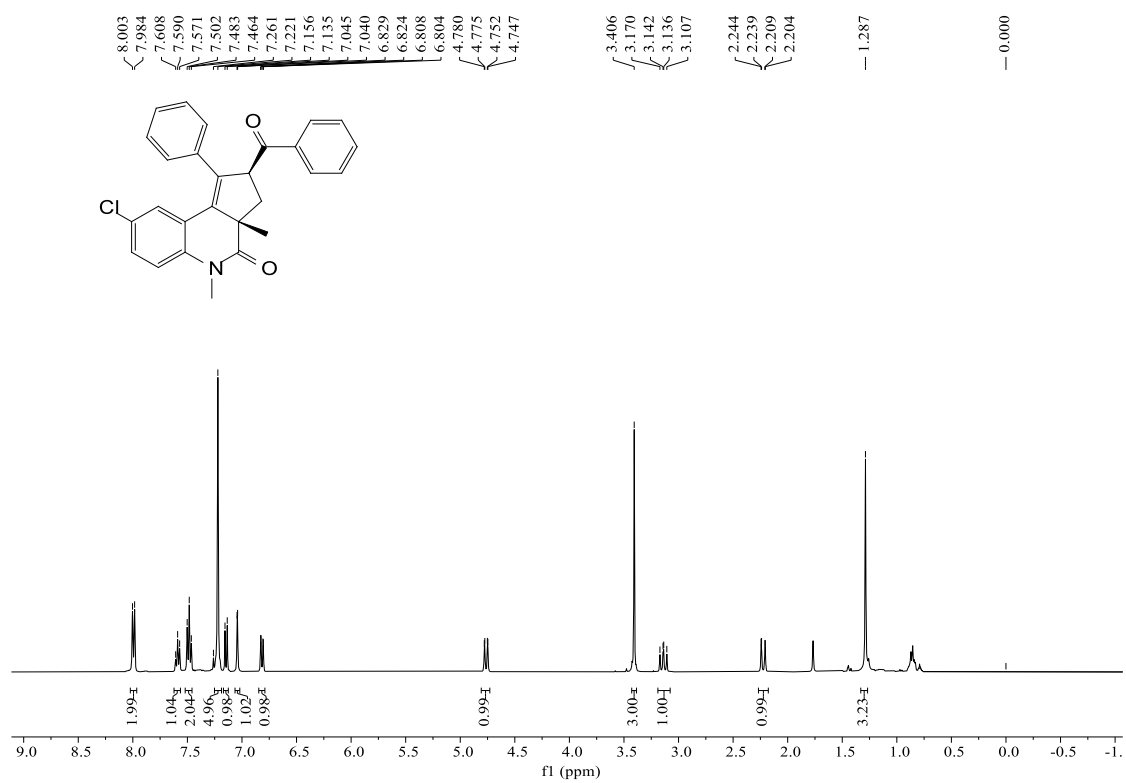
¹³C NMR spectra of *trans*-3u (100 MHz, CDCl₃)



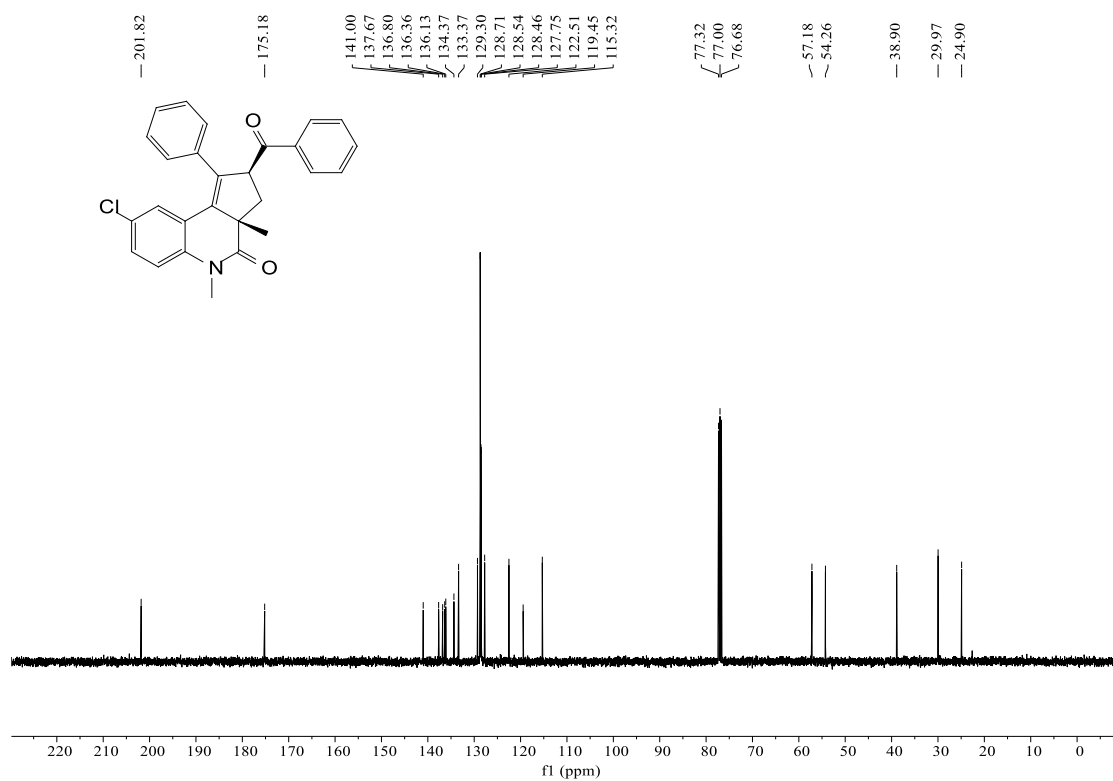
¹⁹F NMR spectra of *trans*-3u (376 MHz, CDCl₃)



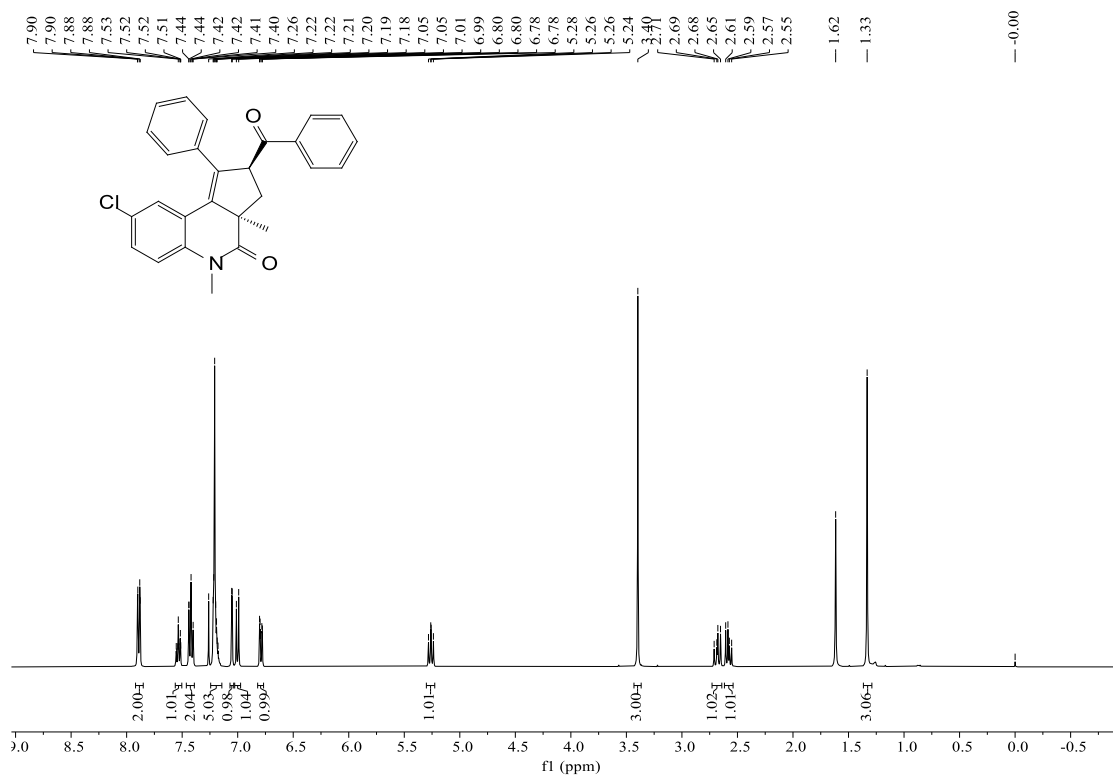
^1H NMR spectra of *cis*-3v (400 MHz, CDCl_3)



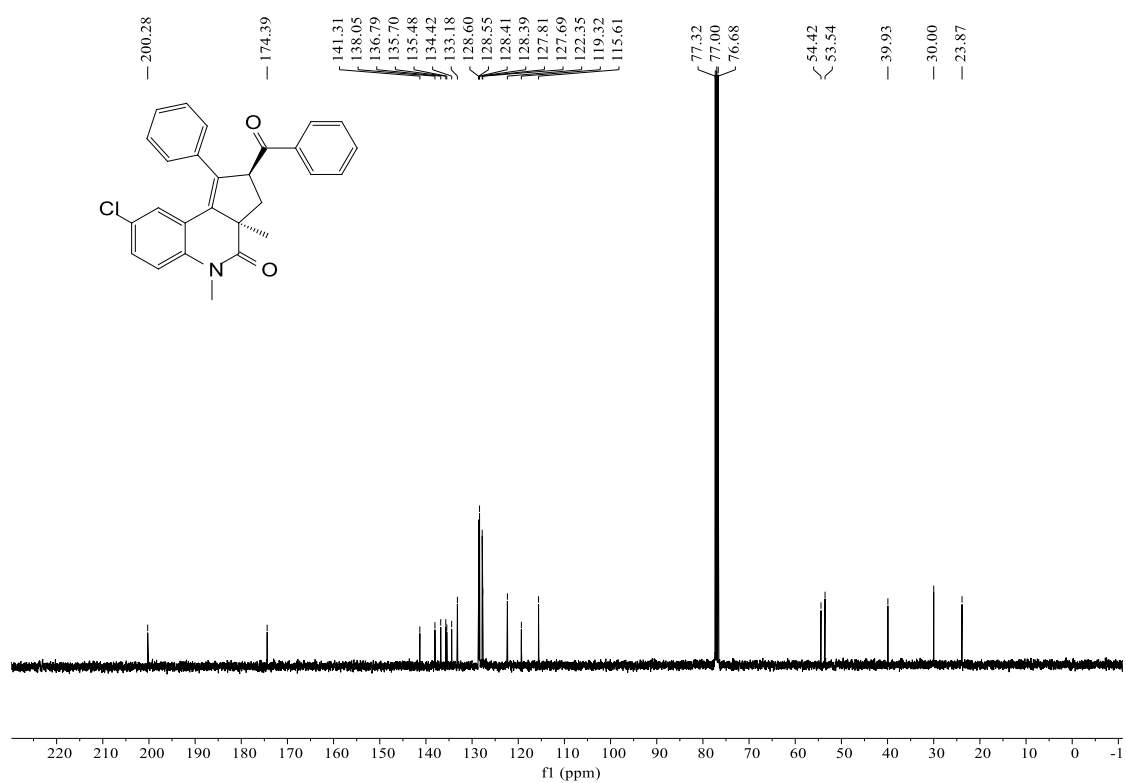
^{13}C NMR spectra of *cis*-3v (100 MHz, CDCl_3)



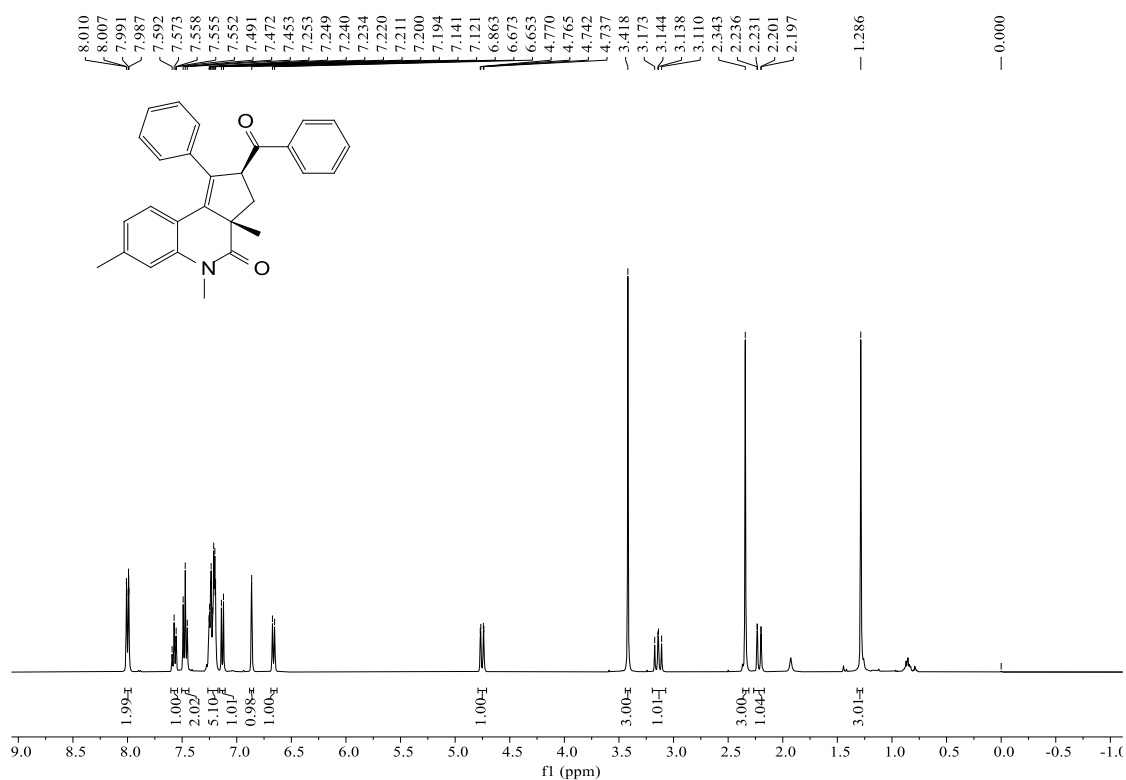
¹H NMR spectra of *trans*-3v (400 MHz, CDCl₃)



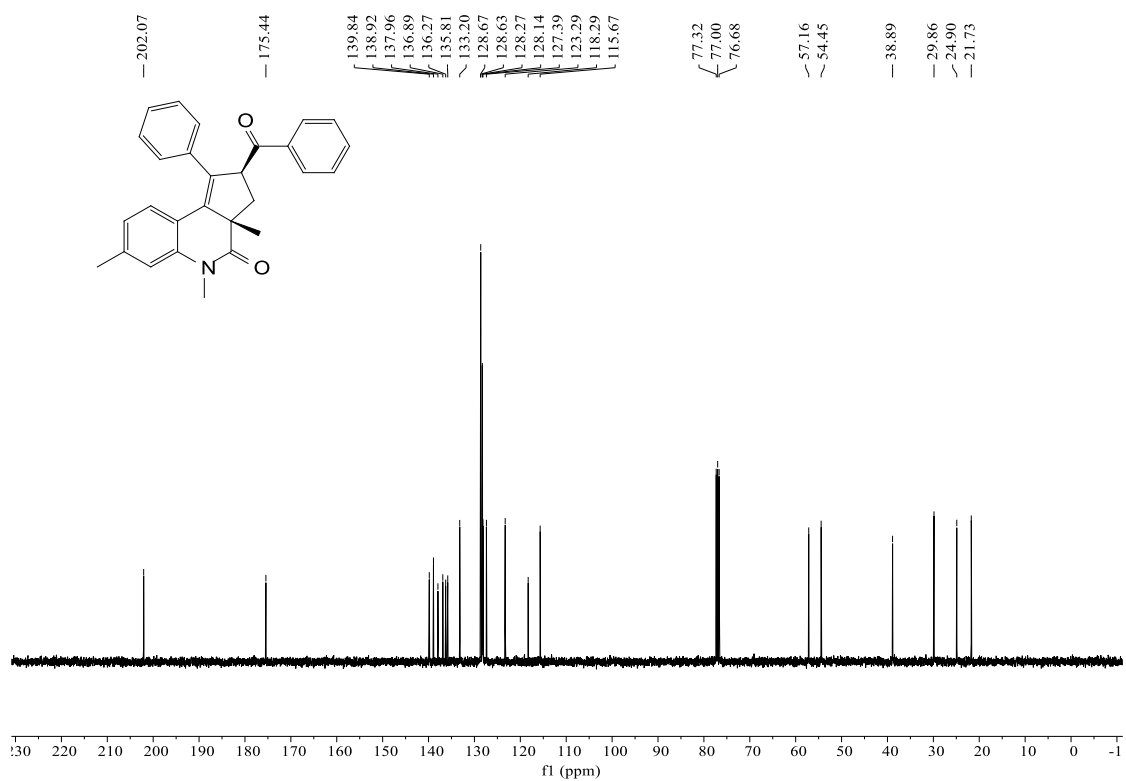
¹³C NMR spectra of *trans*-3v (100 MHz, CDCl₃)



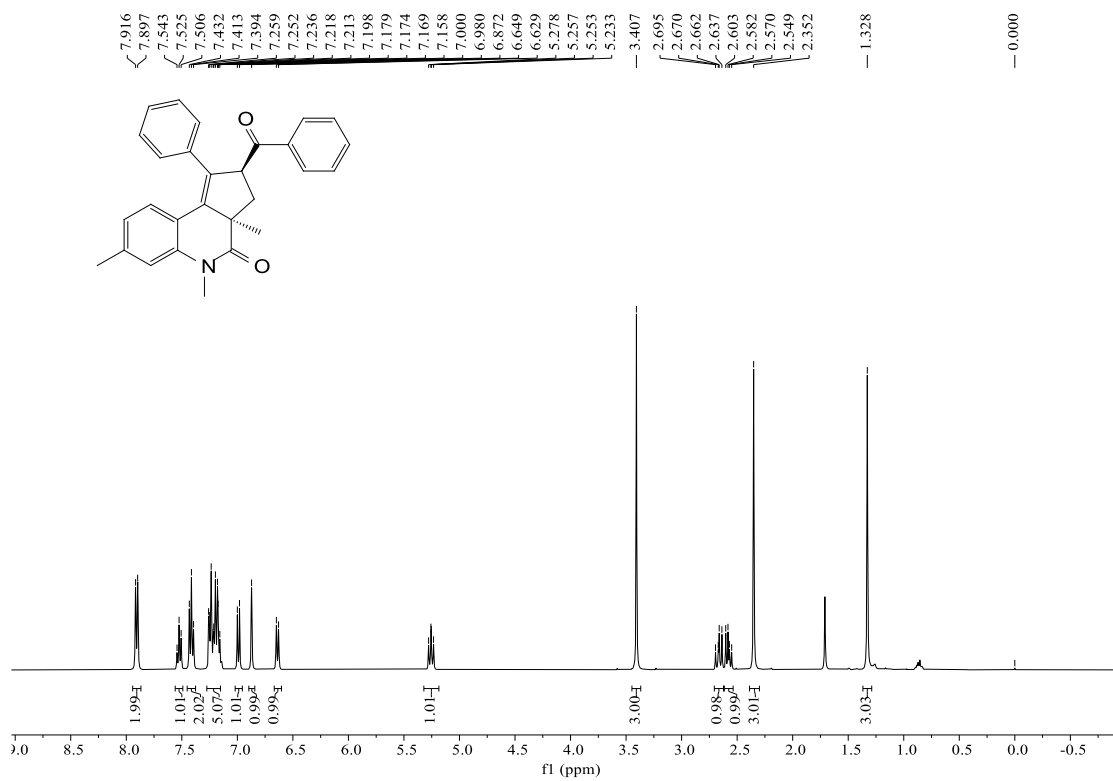
¹H NMR spectra of *cis*-3w (400 MHz, CDCl₃)



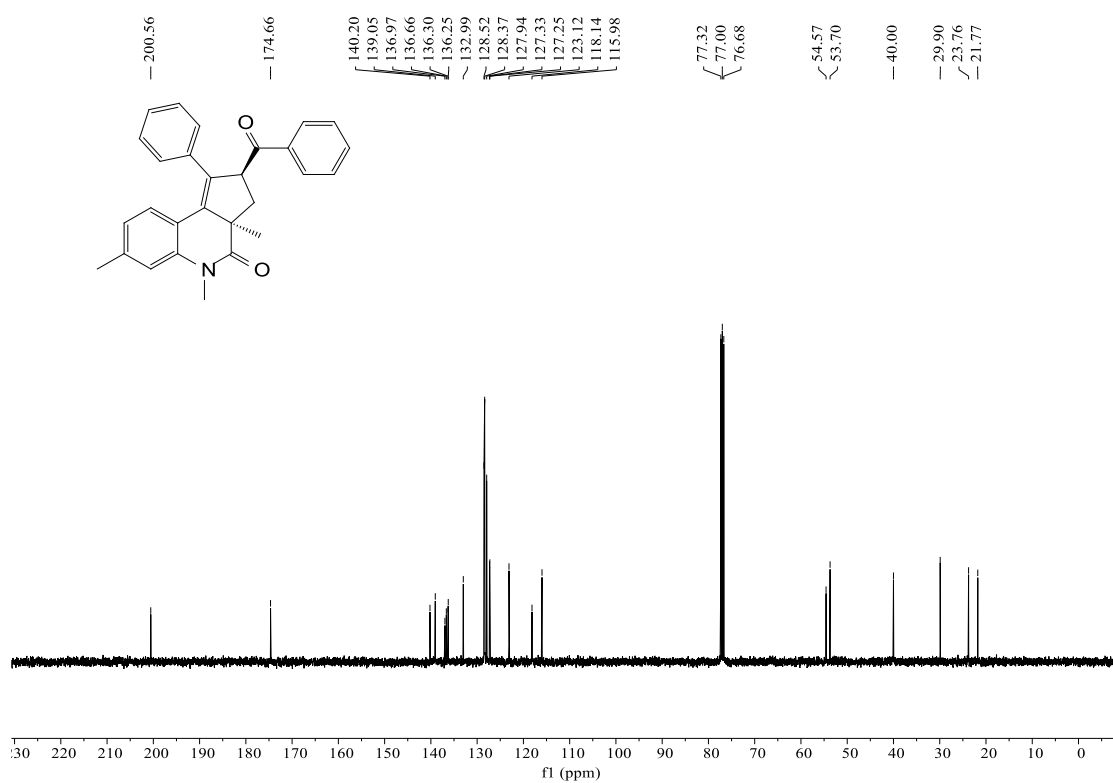
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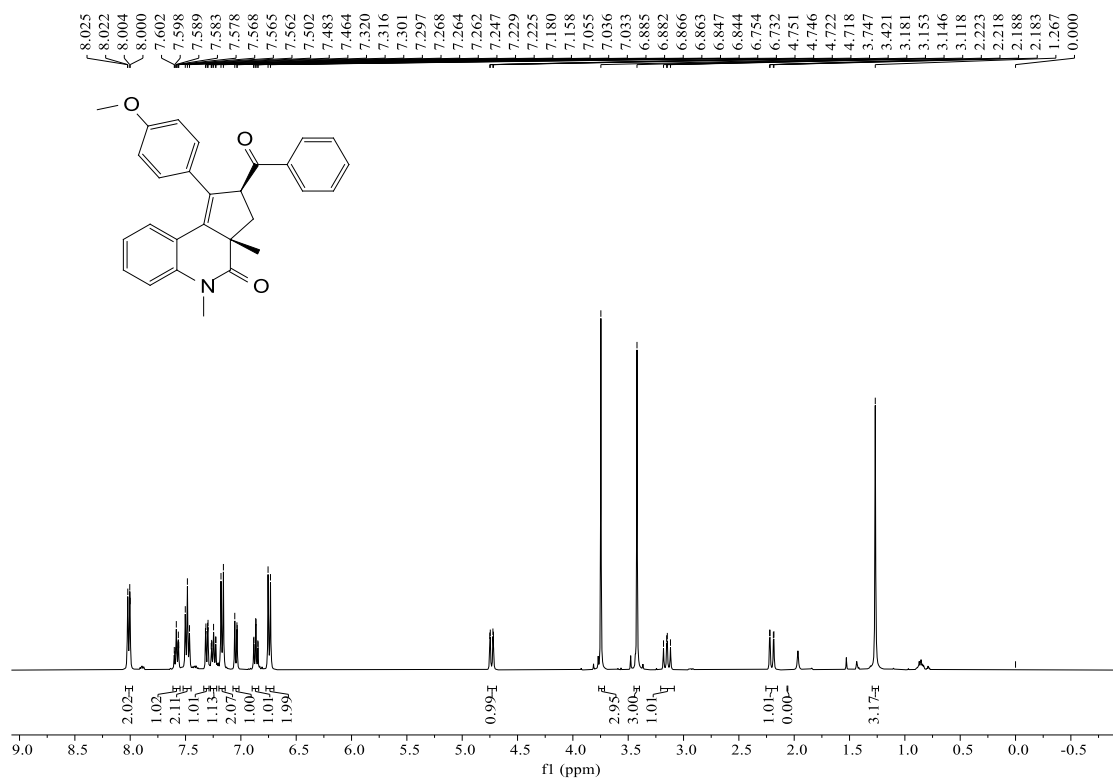
¹H NMR spectra of *trans*-3w (400 MHz, CDCl₃)



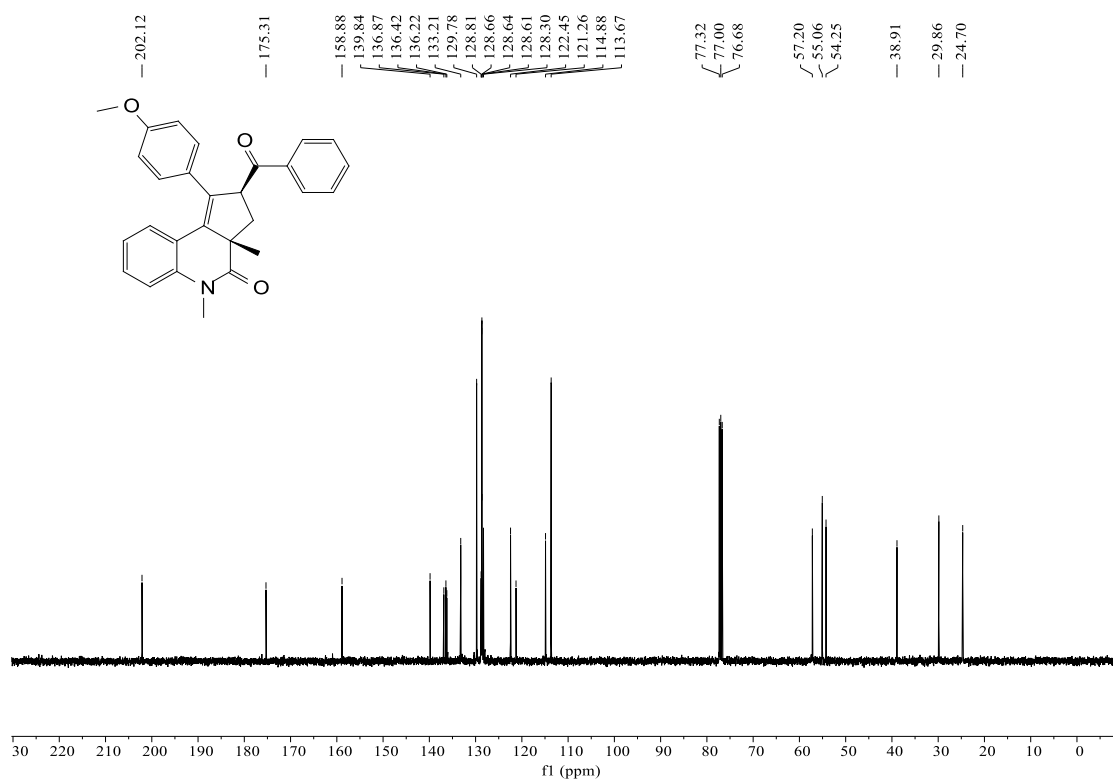
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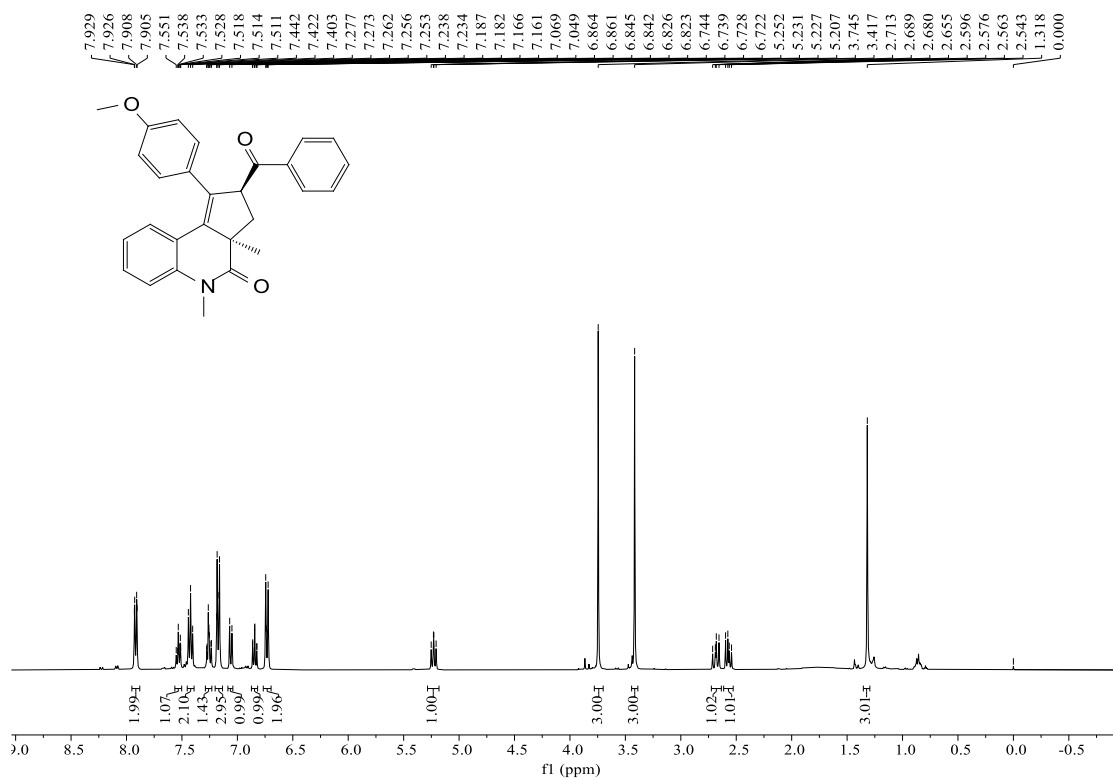
¹H NMR spectra of *cis*-3x (400 MHz, CDCl₃)



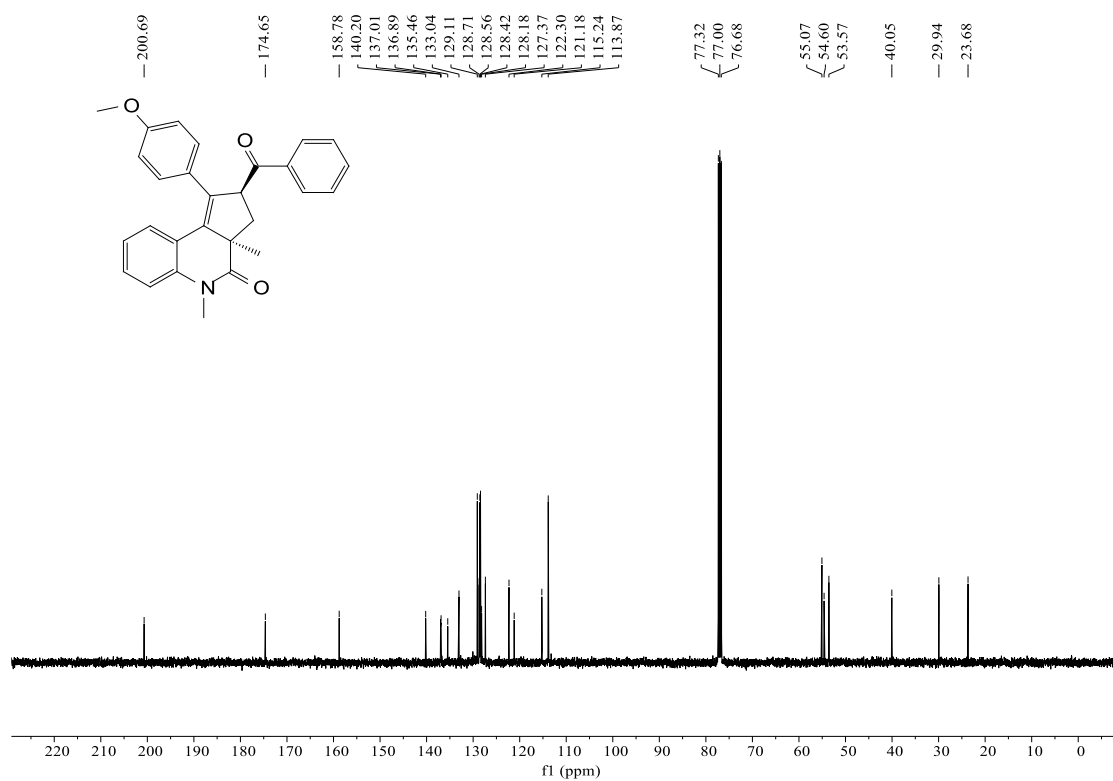
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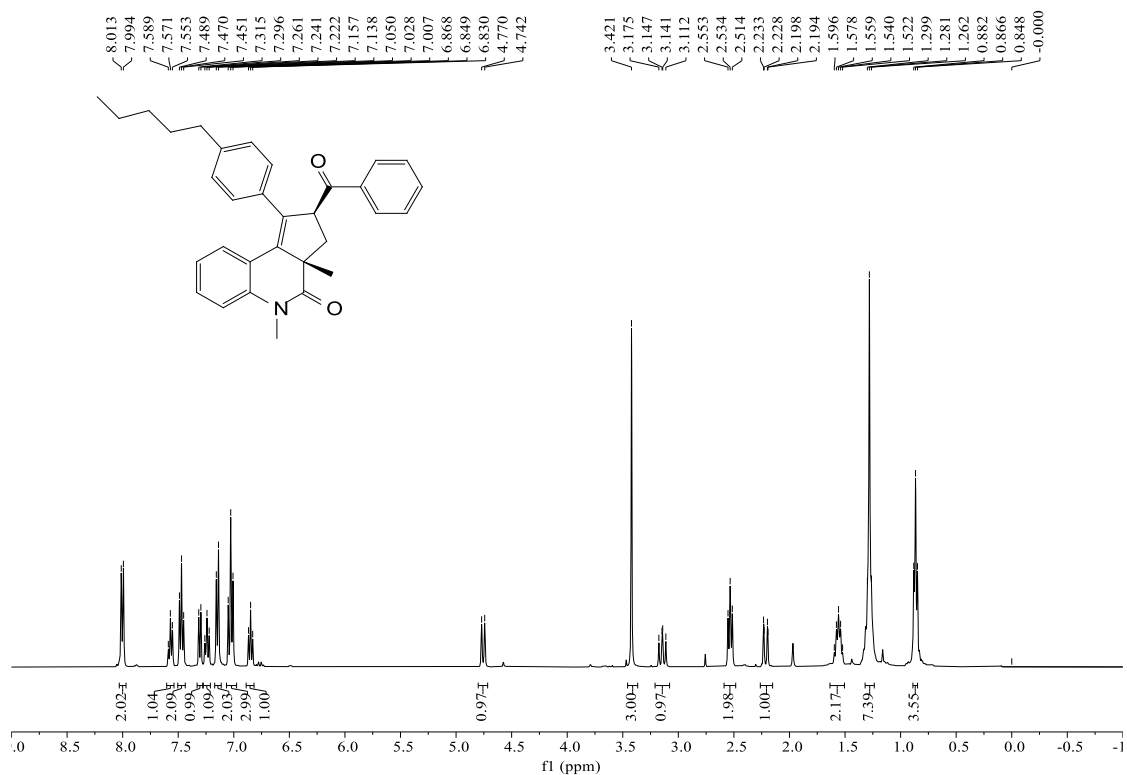
¹H NMR spectra of *trans*-3x (400 MHz, CDCl₃)



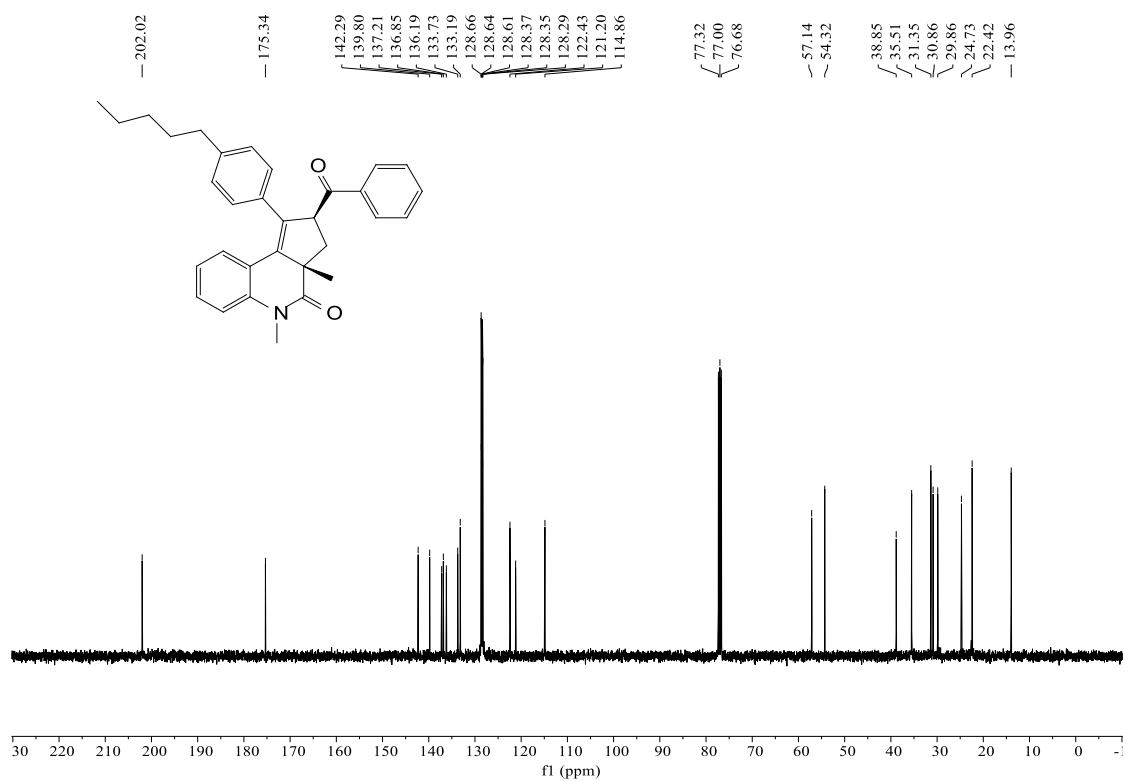
¹³C NMR spectra of *trans*-3x (100 MHz, CDCl₃)



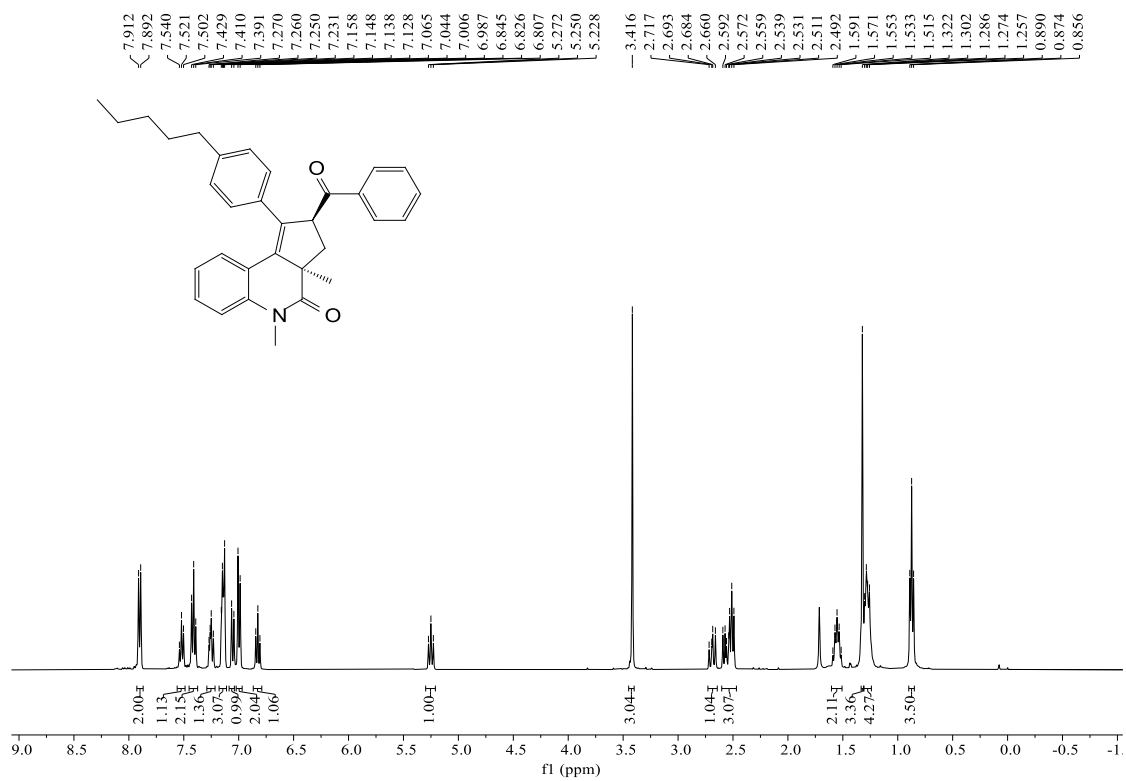
¹H NMR spectra of *cis*-3y (400 MHz, CDCl₃)



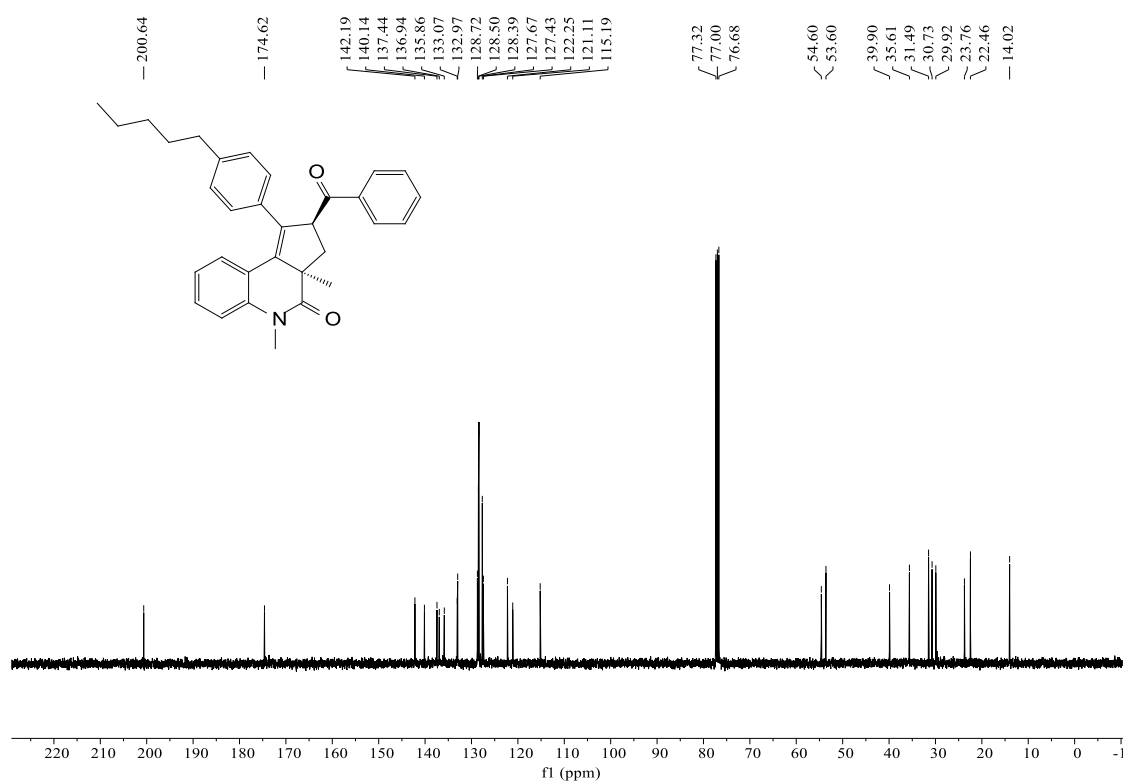
¹³C NMR spectra of *cis*-3y (100 MHz, CDCl₃)



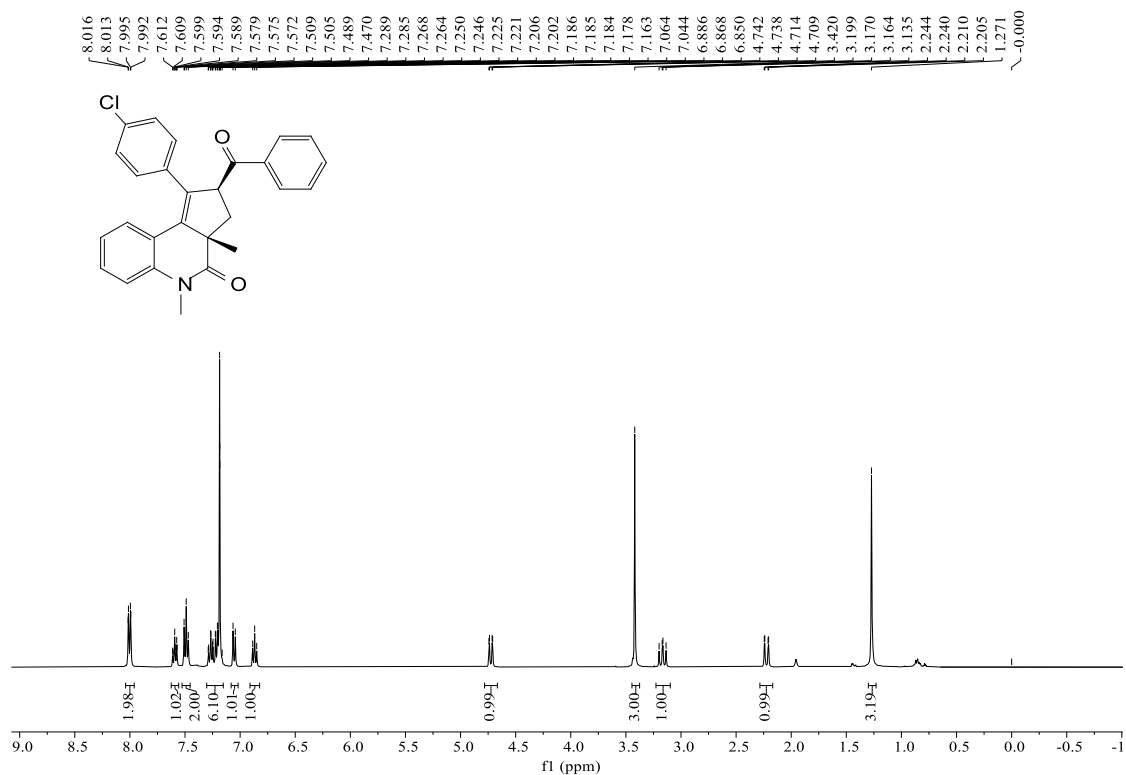
¹H NMR spectra of *trans*-3y (400 MHz, CDCl₃)



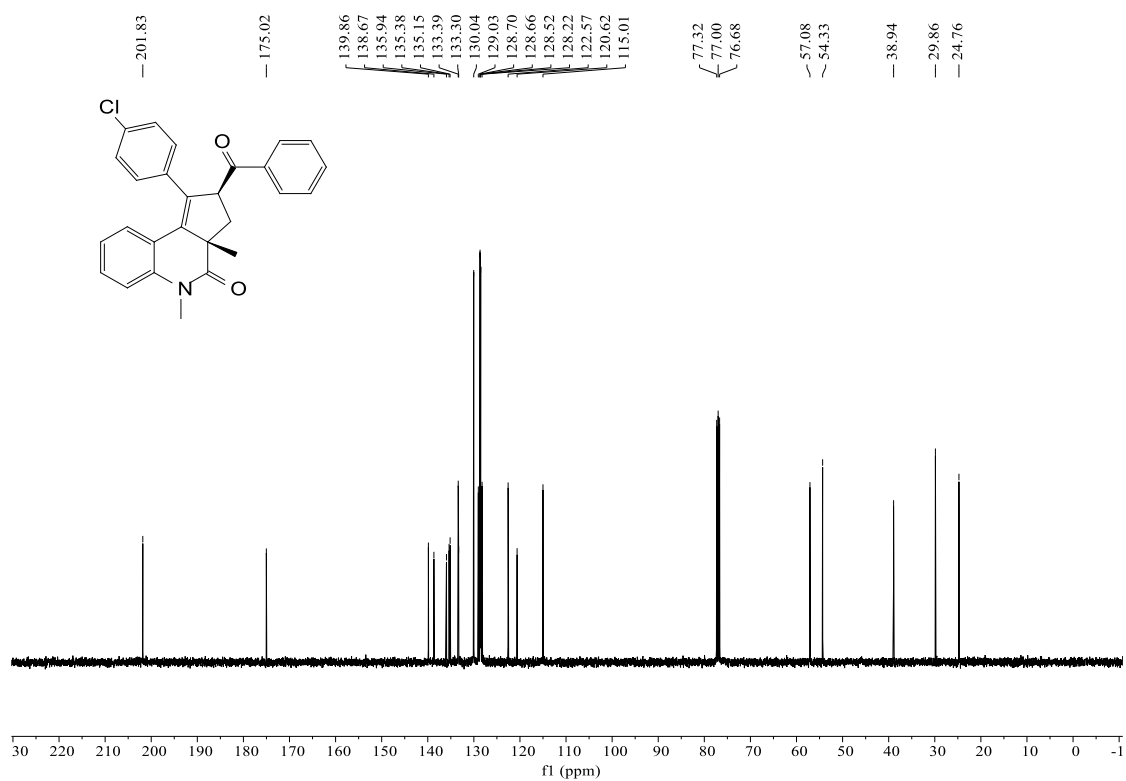
¹³C NMR spectra of *trans*-3y (100 MHz, CDCl₃)



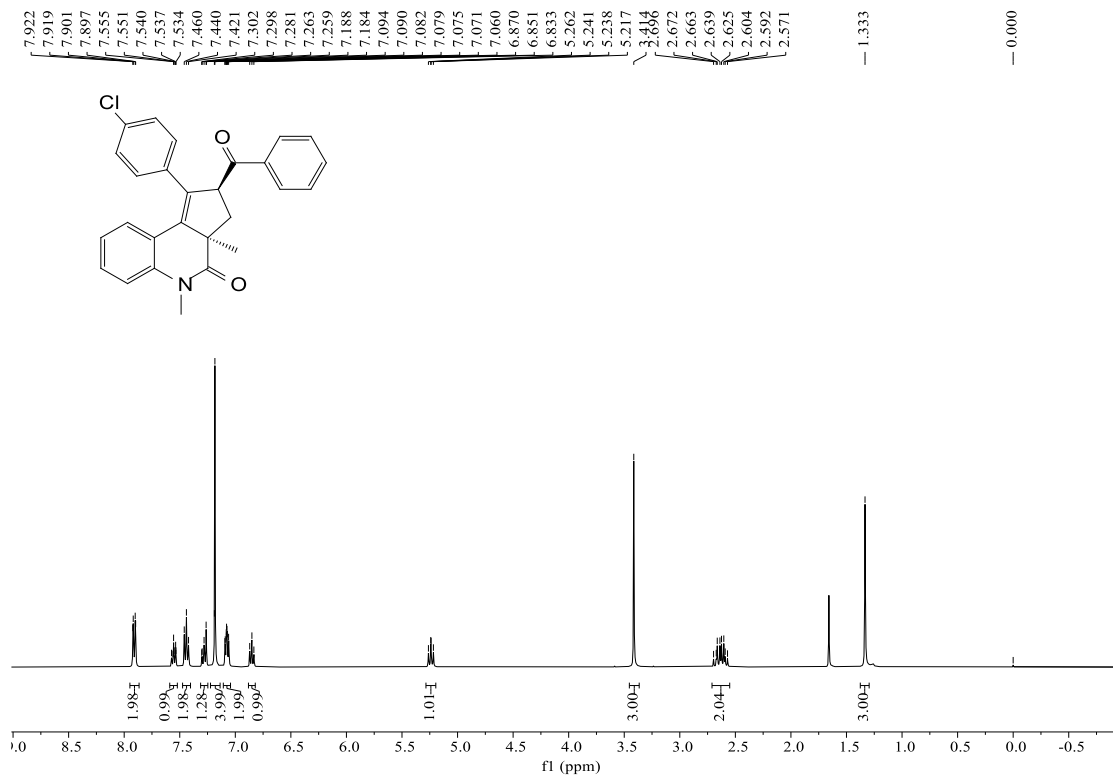
¹H NMR spectra of *cis*-3z (400 MHz, CDCl₃)



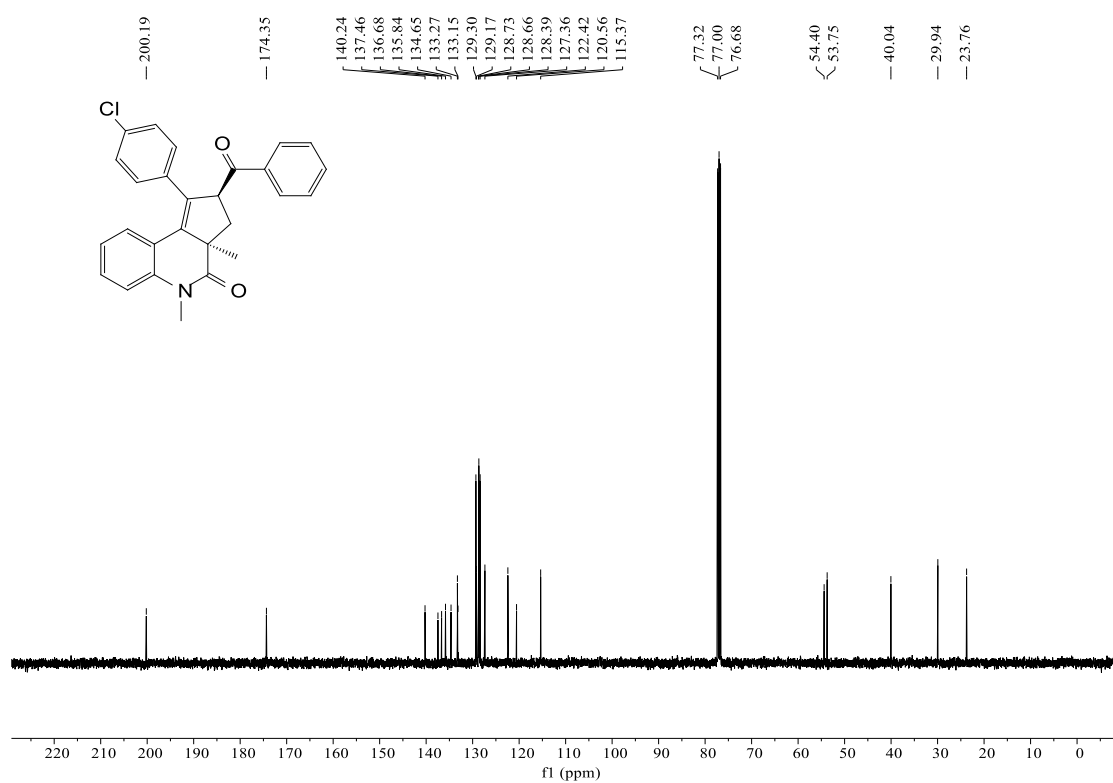
¹³C NMR spectra of *cis*-3z (100 MHz, CDCl₃)



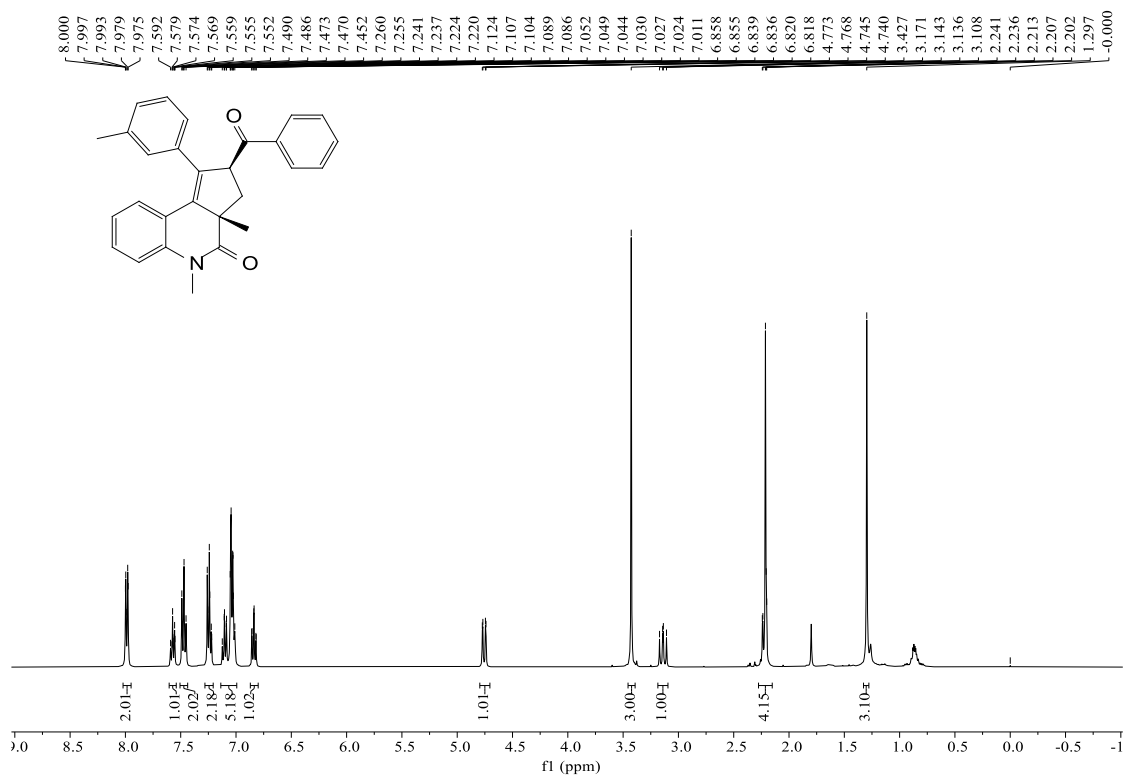
¹H NMR spectra of *trans*-3z (400 MHz, CDCl₃)



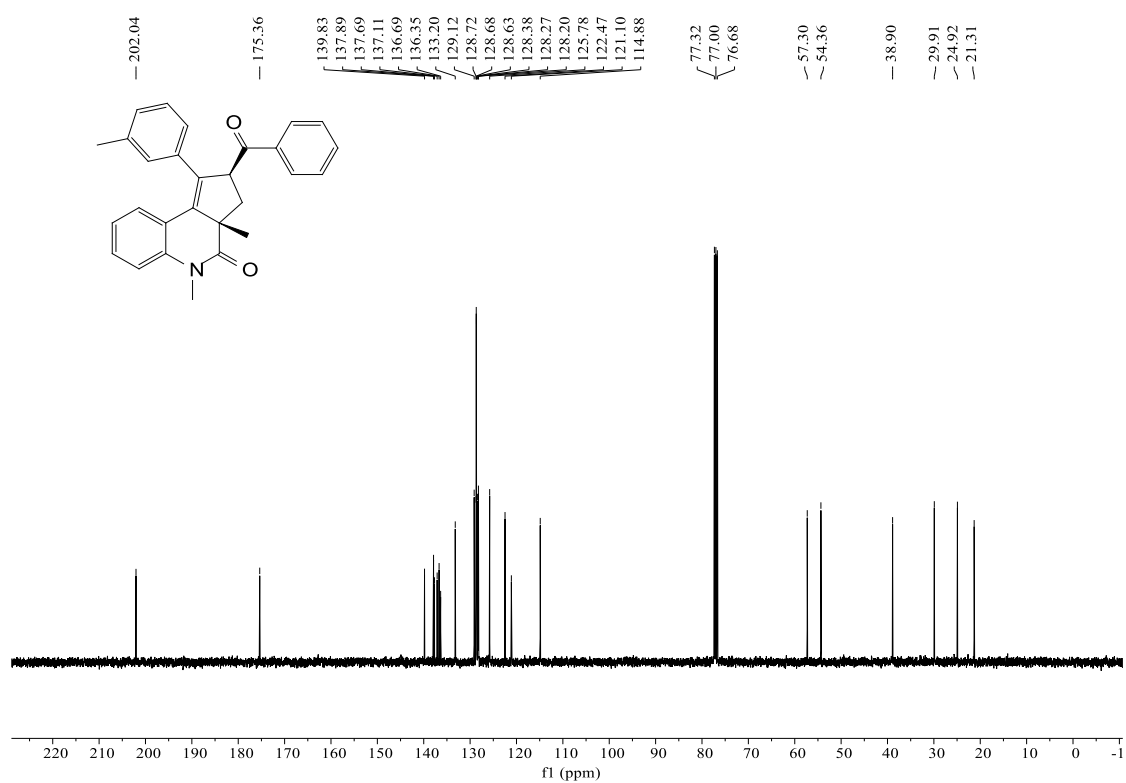
¹³C NMR spectra of *trans*-3z (100 MHz, CDCl₃)



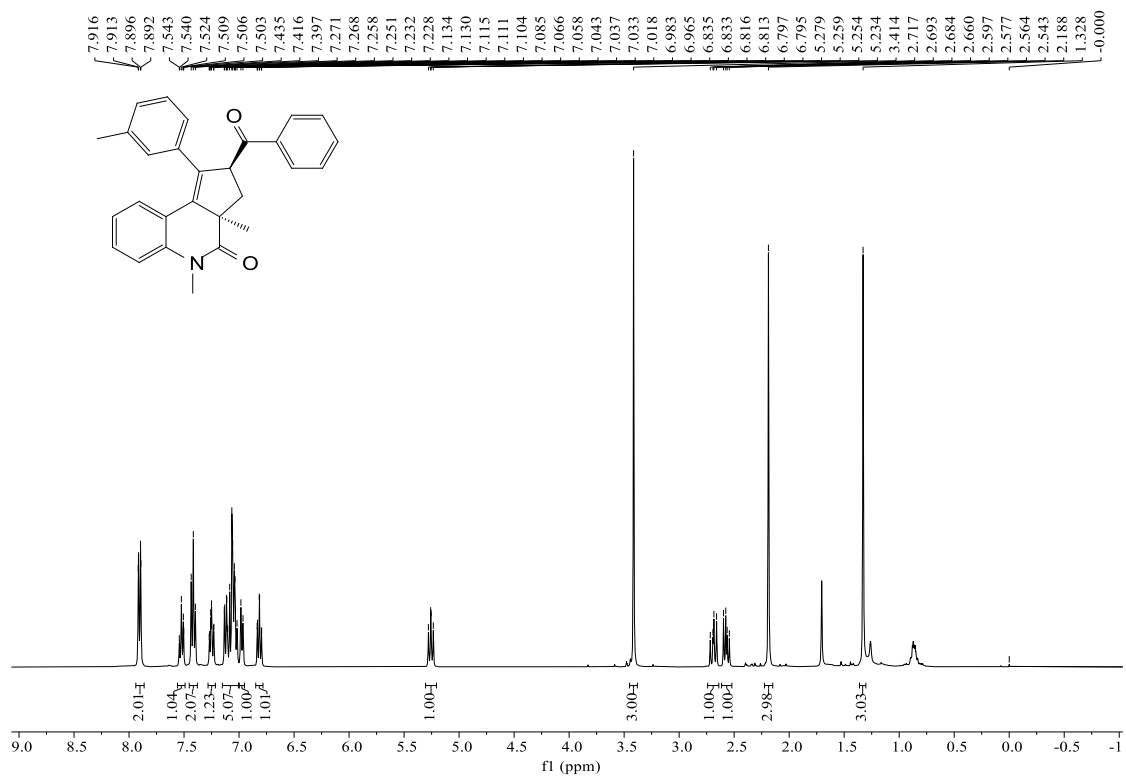
¹H NMR spectra of *cis*-3aa (400 MHz, CDCl₃)



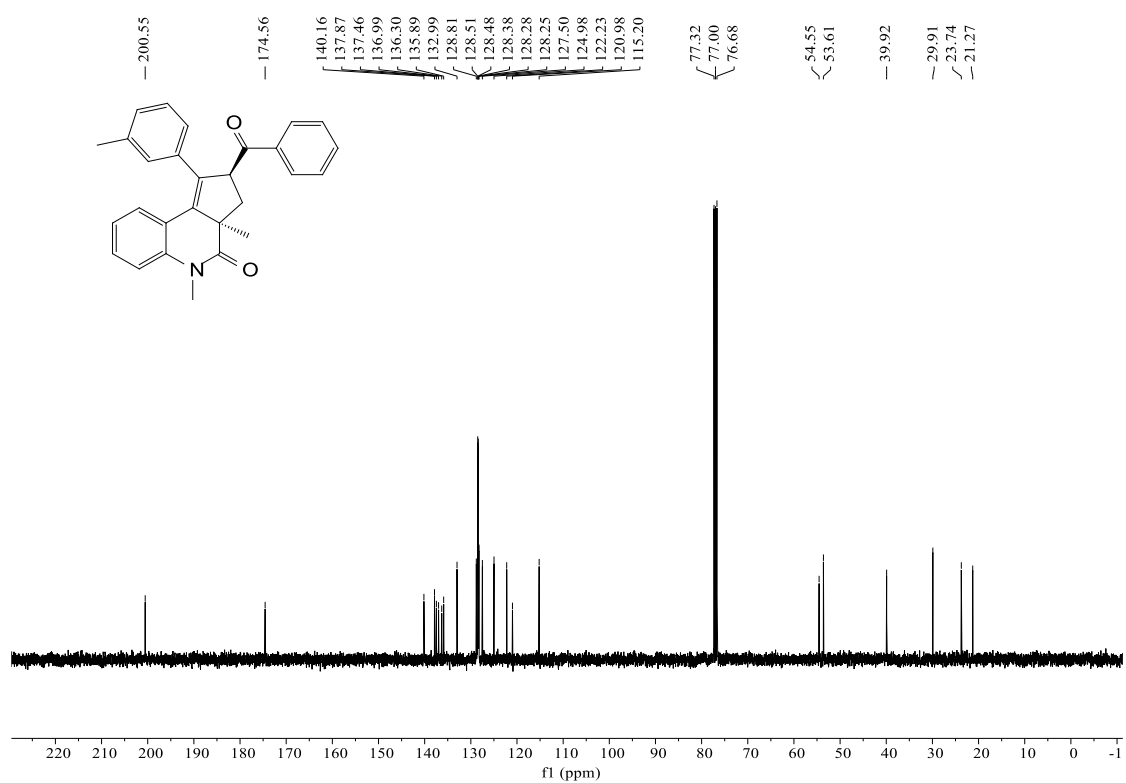
¹³C NMR spectra of *cis*-3aa (100 MHz, CDCl₃)



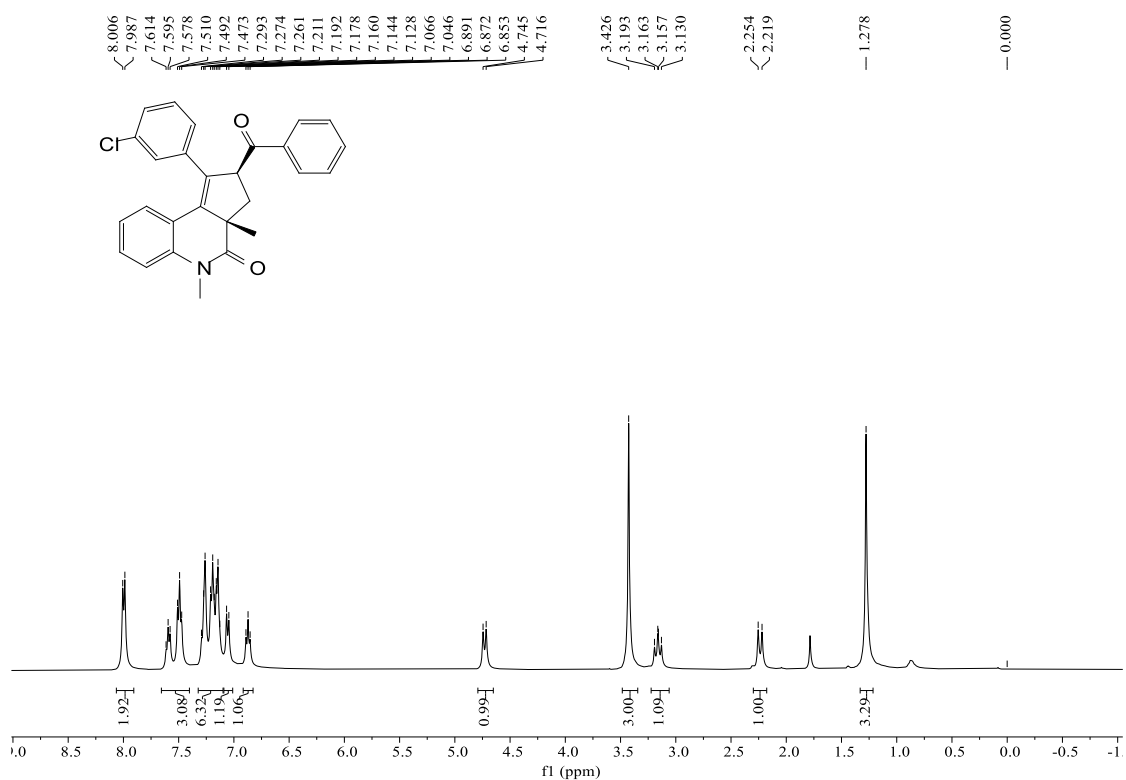
¹H NMR spectra of *trans*-3aa (400 MHz, CDCl₃)



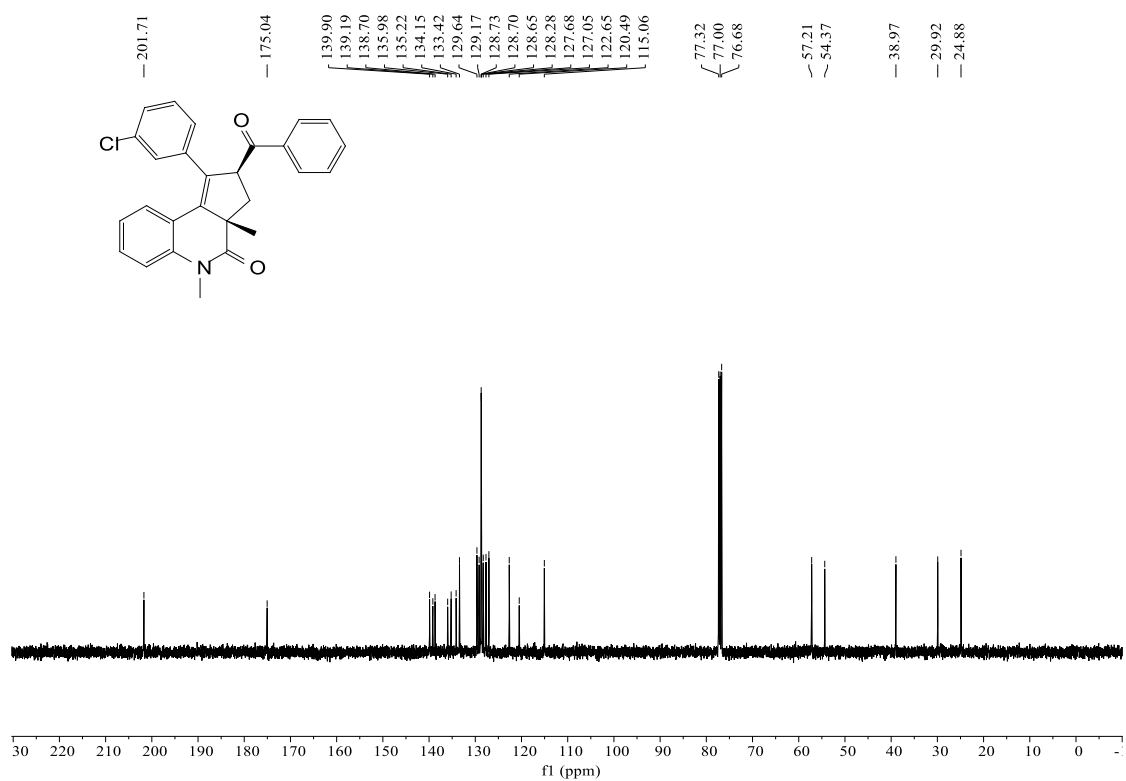
¹³C NMR spectra of *trans*-3aa (100 MHz, CDCl₃)



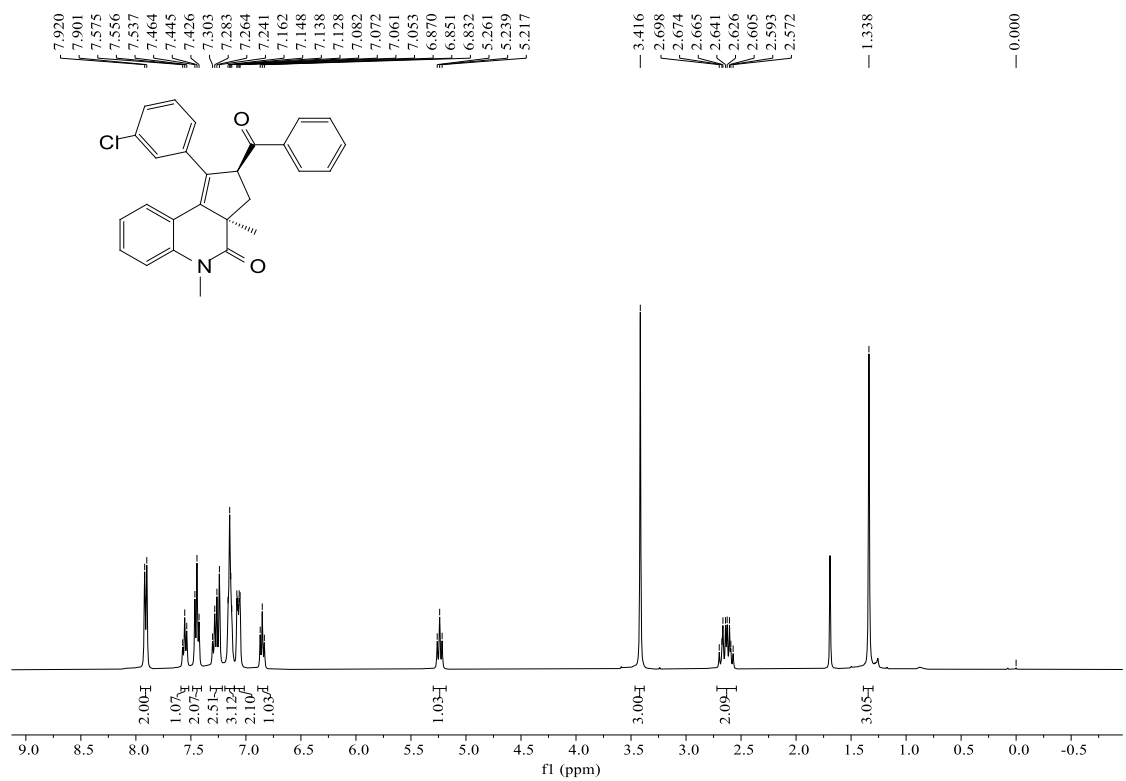
¹H NMR spectra of *cis*-3ab (400 MHz, CDCl₃)



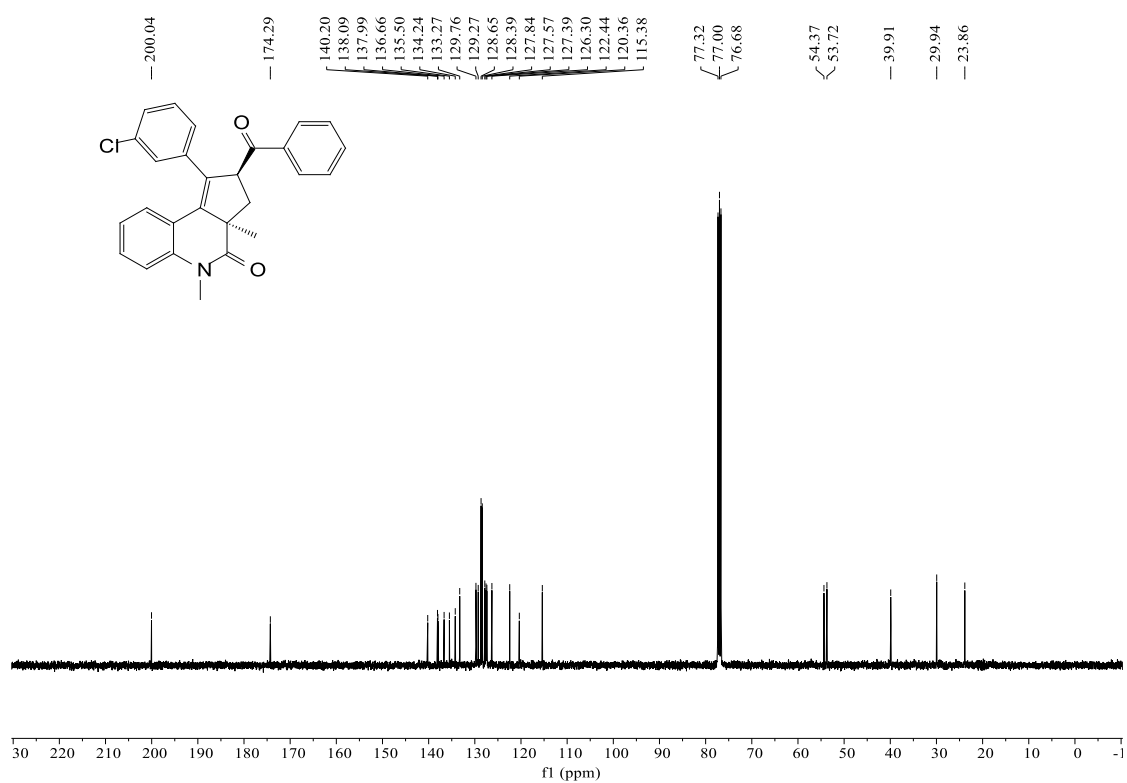
¹³C NMR spectra of *cis*-3ab (100 MHz, CDCl₃)



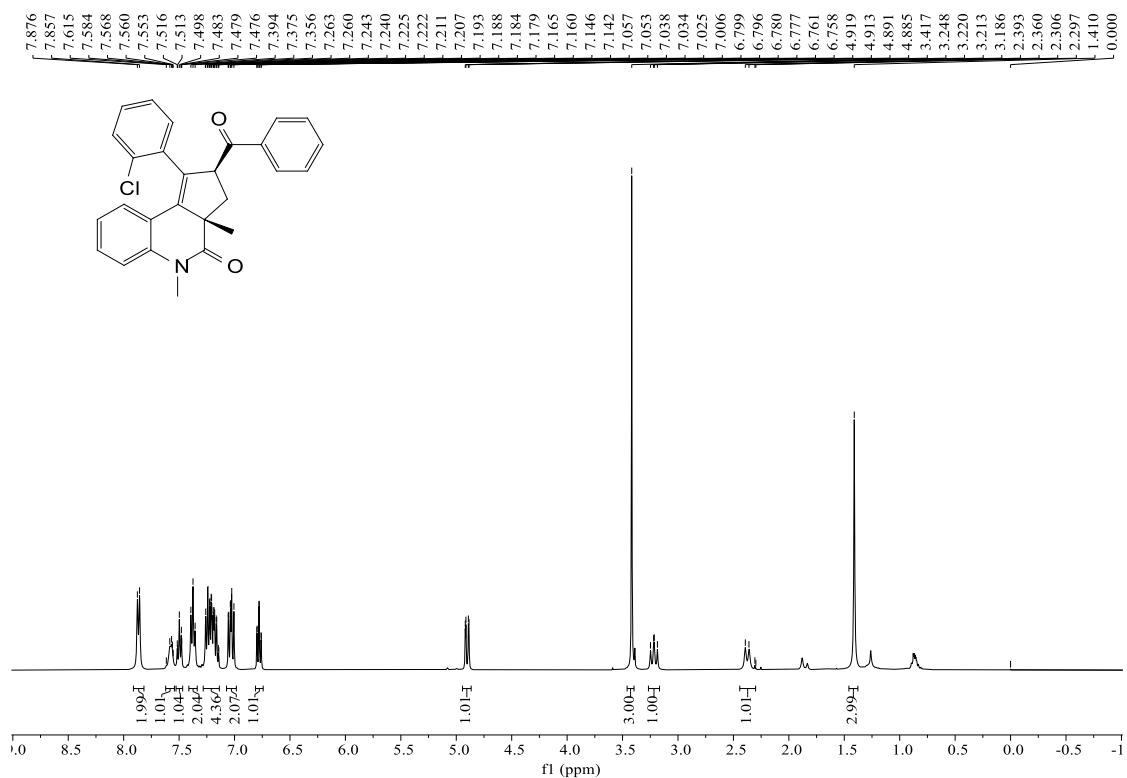
¹H NMR spectra of *trans*-3ab (400 MHz, CDCl₃)



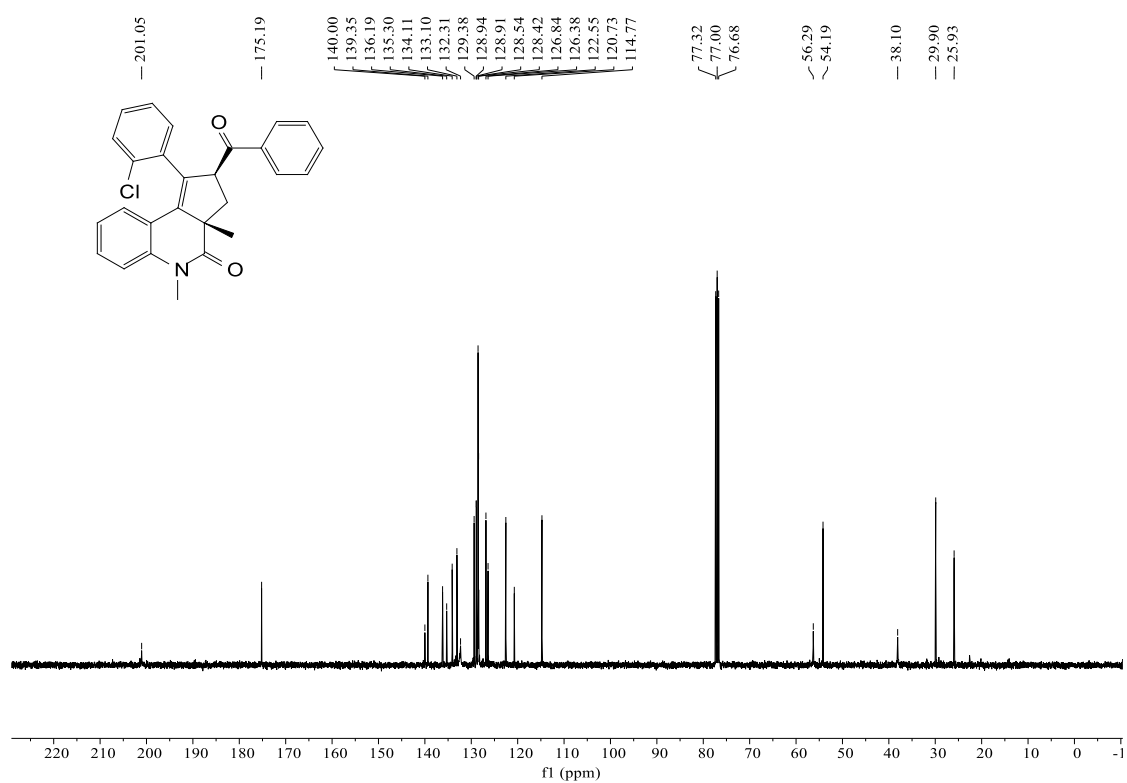
¹³C NMR spectra of *trans*-3ab (100 MHz, CDCl₃)



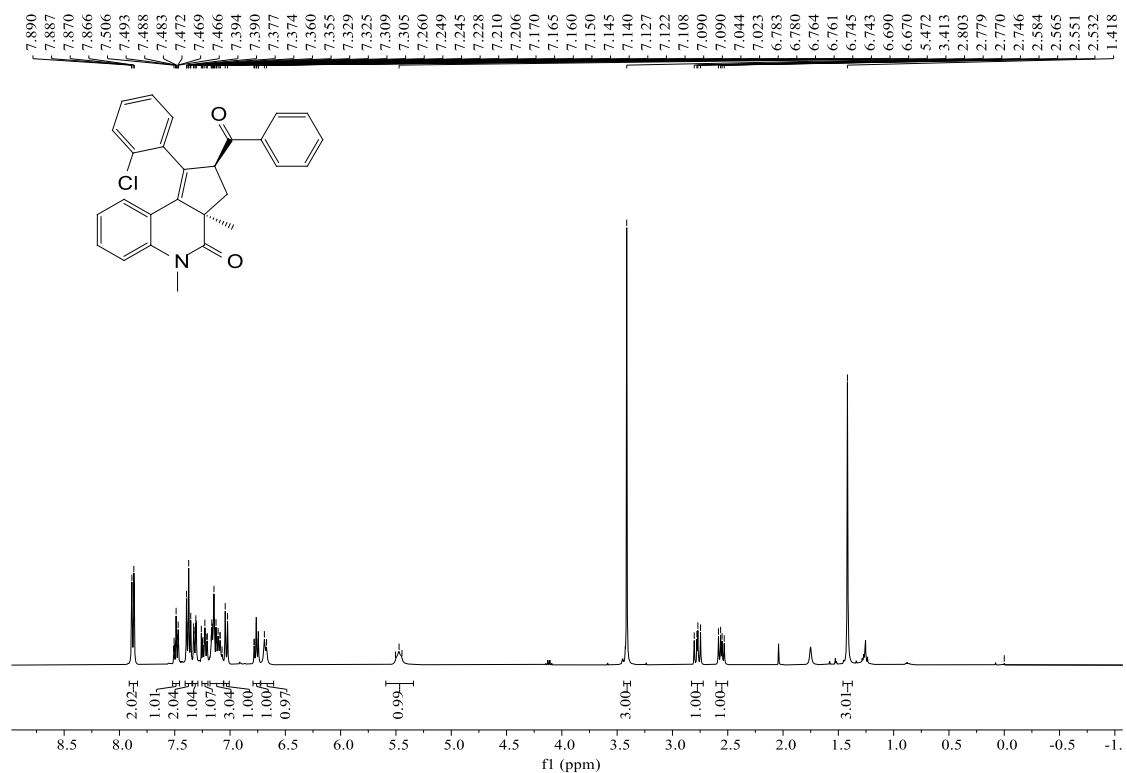
¹H NMR spectra of *cis*-3ac (400 MHz, CDCl₃)



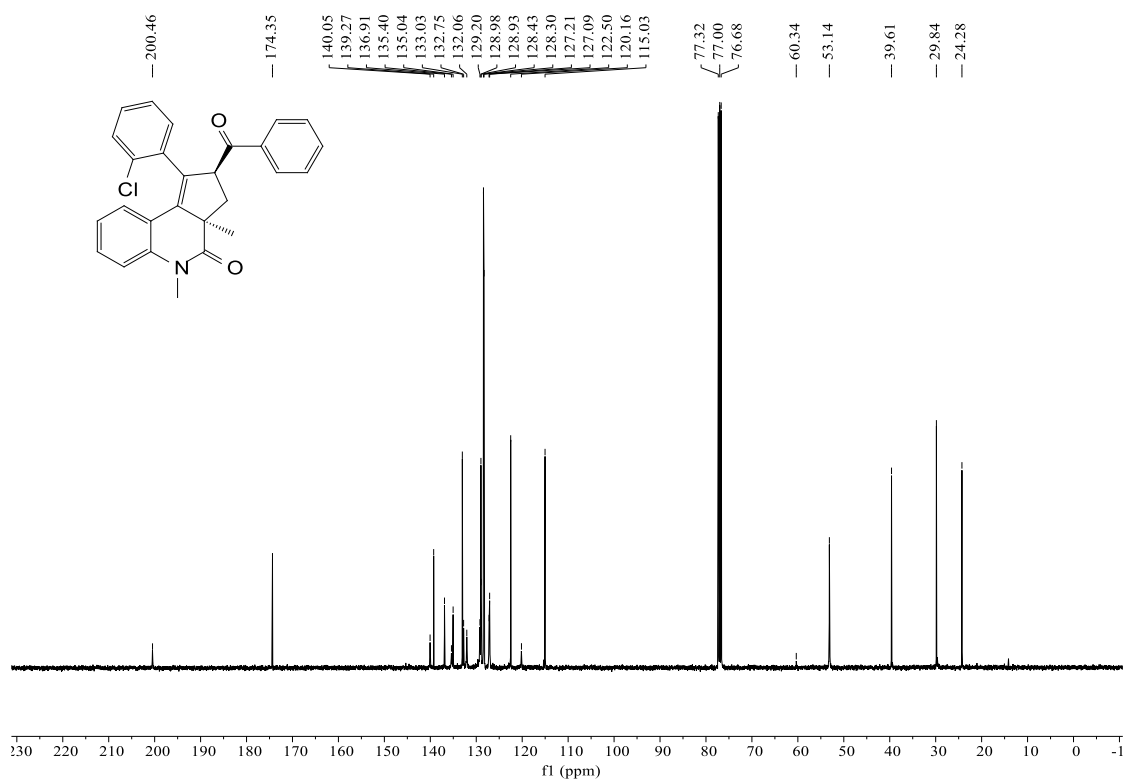
¹³C NMR spectra of *cis*-3ac (100 MHz, CDCl₃)



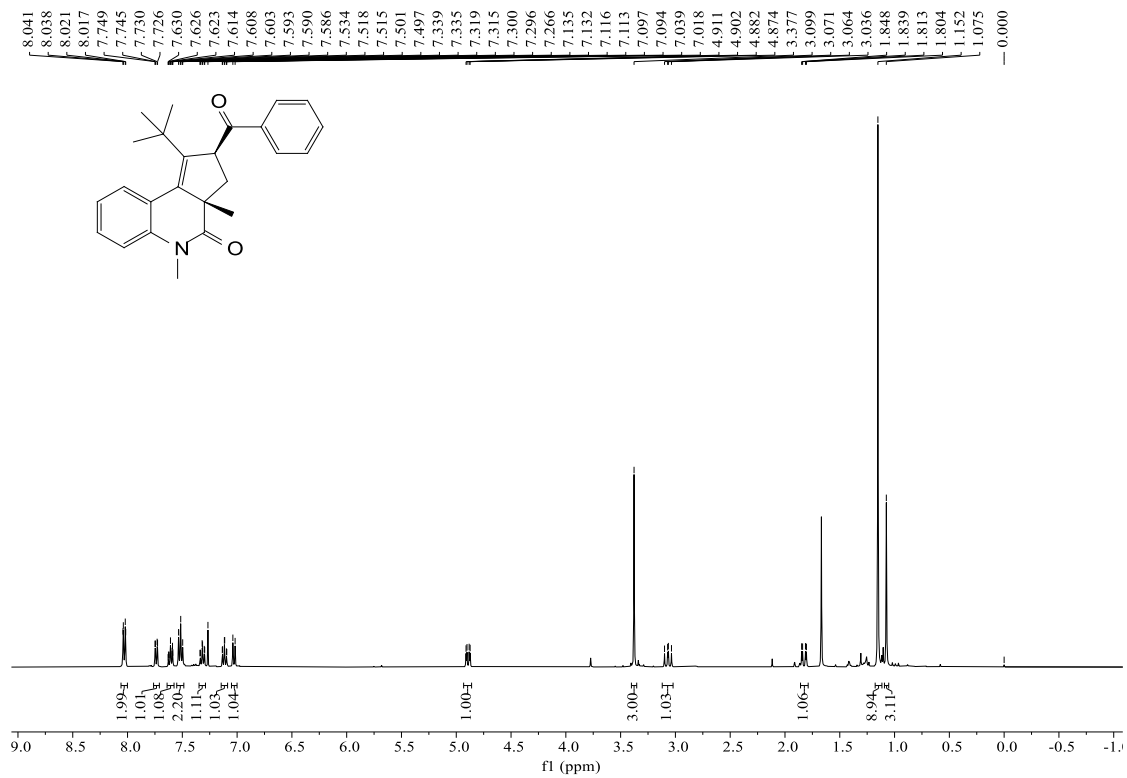
¹H NMR spectra of *trans*-3ac (400 MHz, CDCl₃)



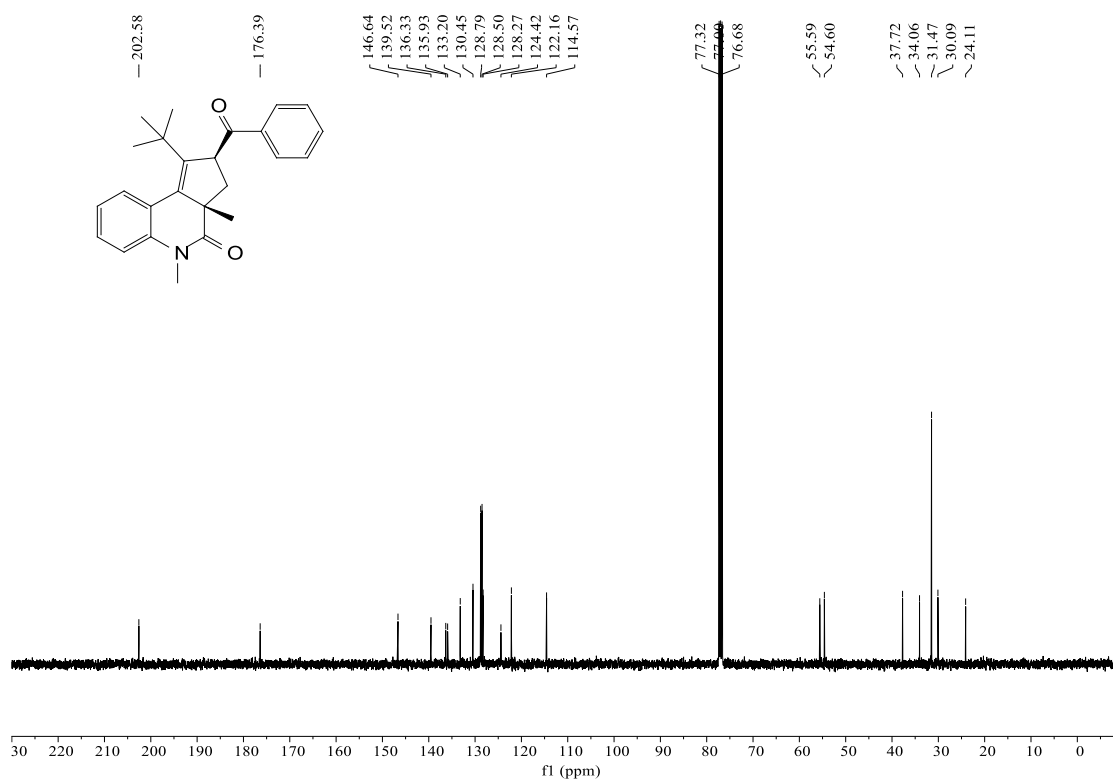
¹³C NMR spectra of *trans*-3ac (100 MHz, CDCl₃)



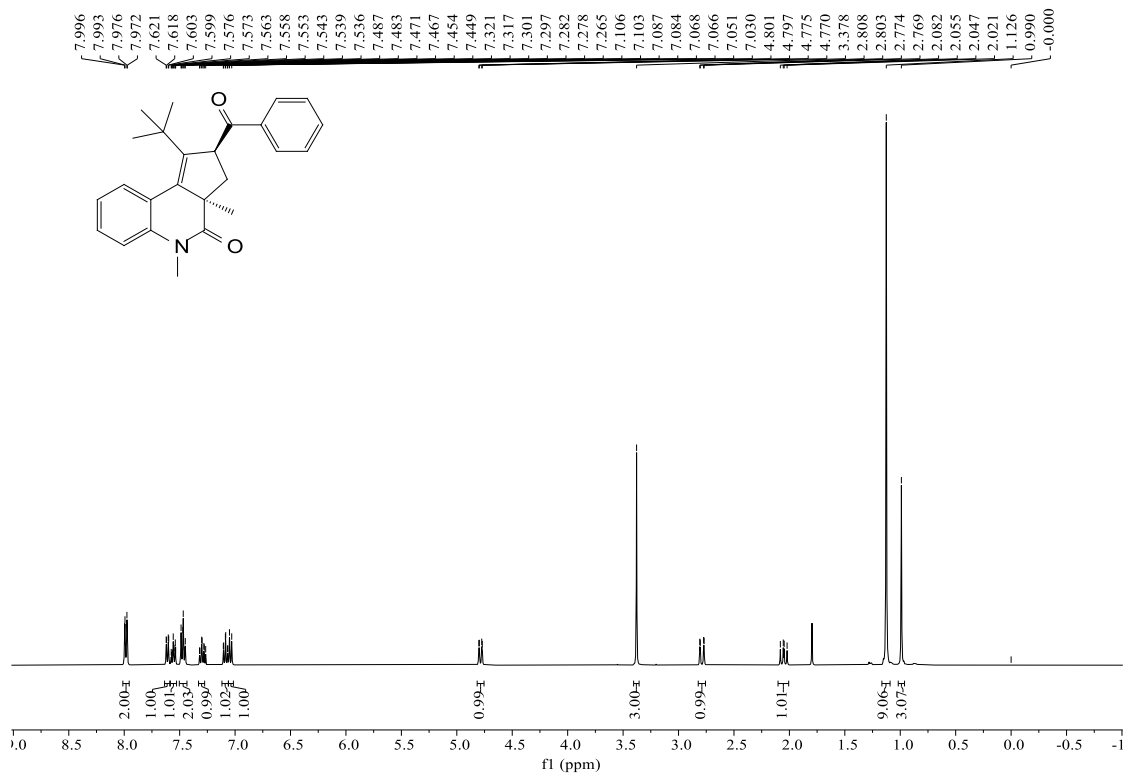
¹H NMR spectra of *cis*-3ad (400 MHz, CDCl₃)



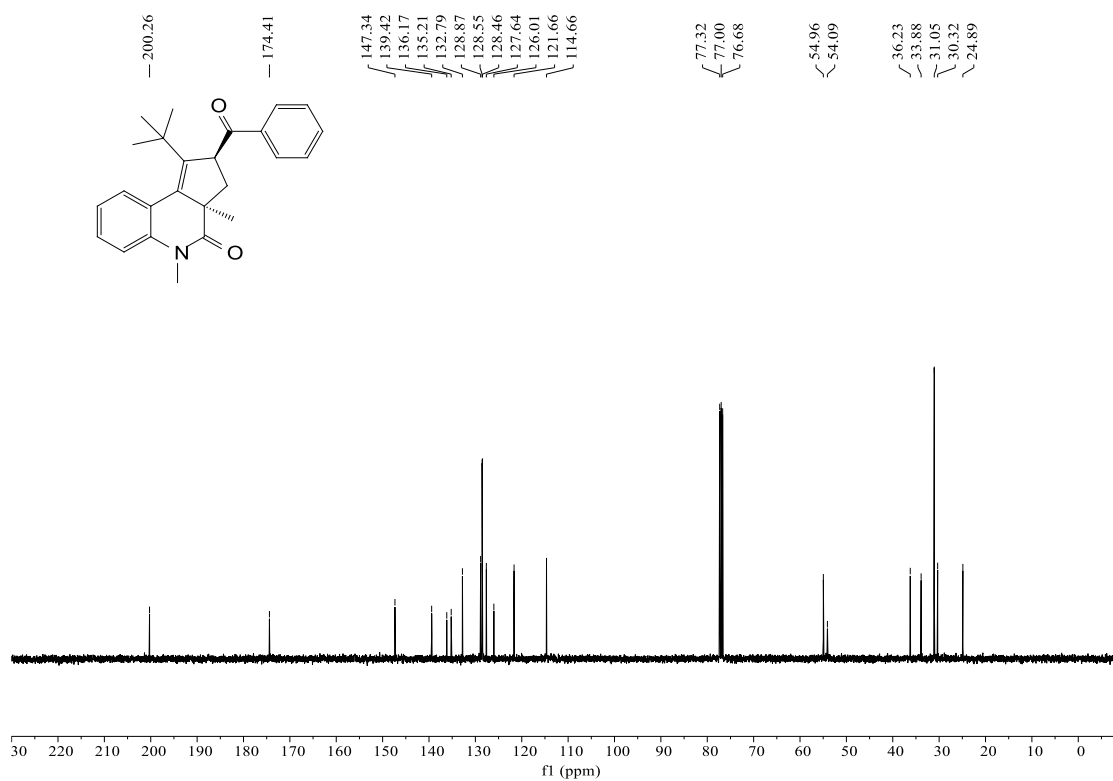
¹³C NMR spectra of *cis*-3ad (100 MHz, CDCl₃)



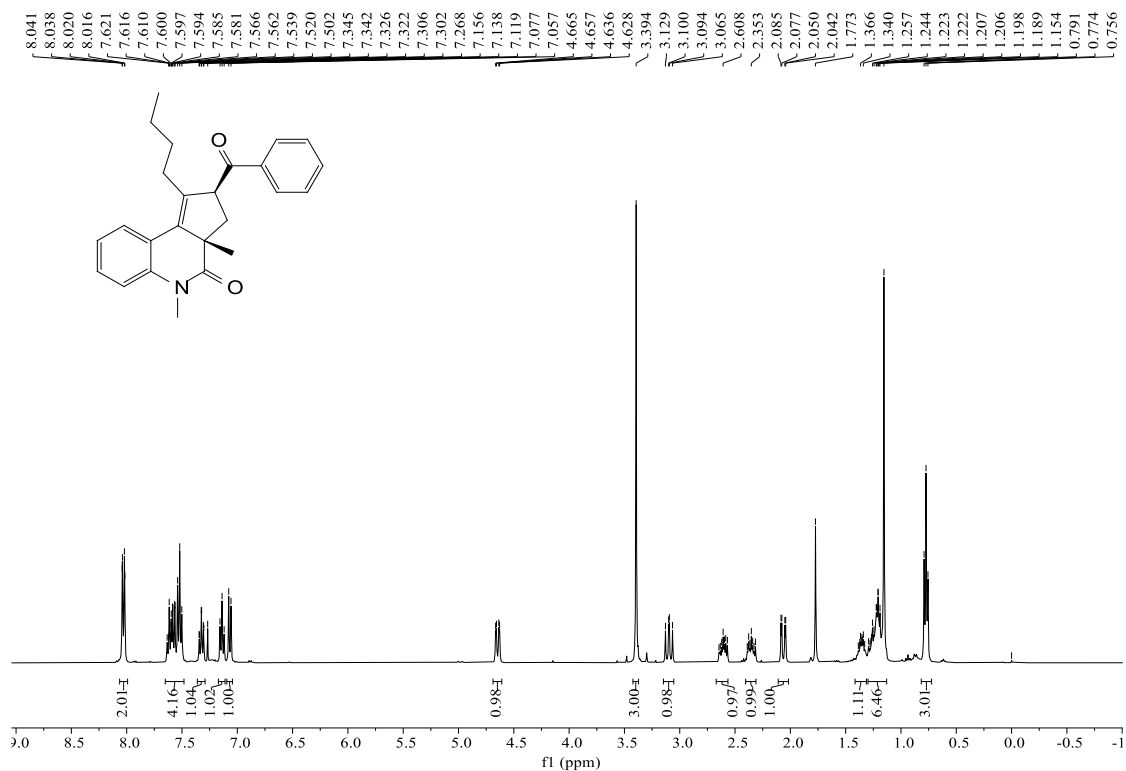
^1H NMR spectra of *trans*-3ad (400 MHz, CDCl_3)



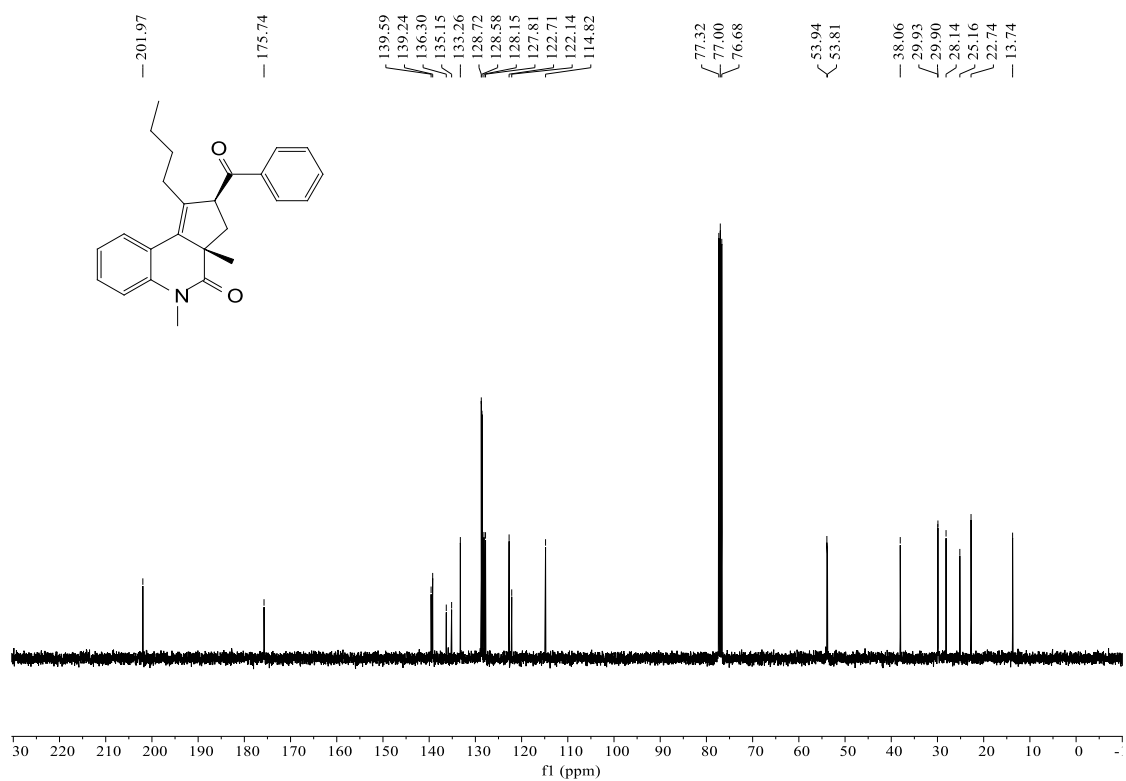
^{13}C NMR spectra of *trans*-3ad (100 MHz, CDCl_3)



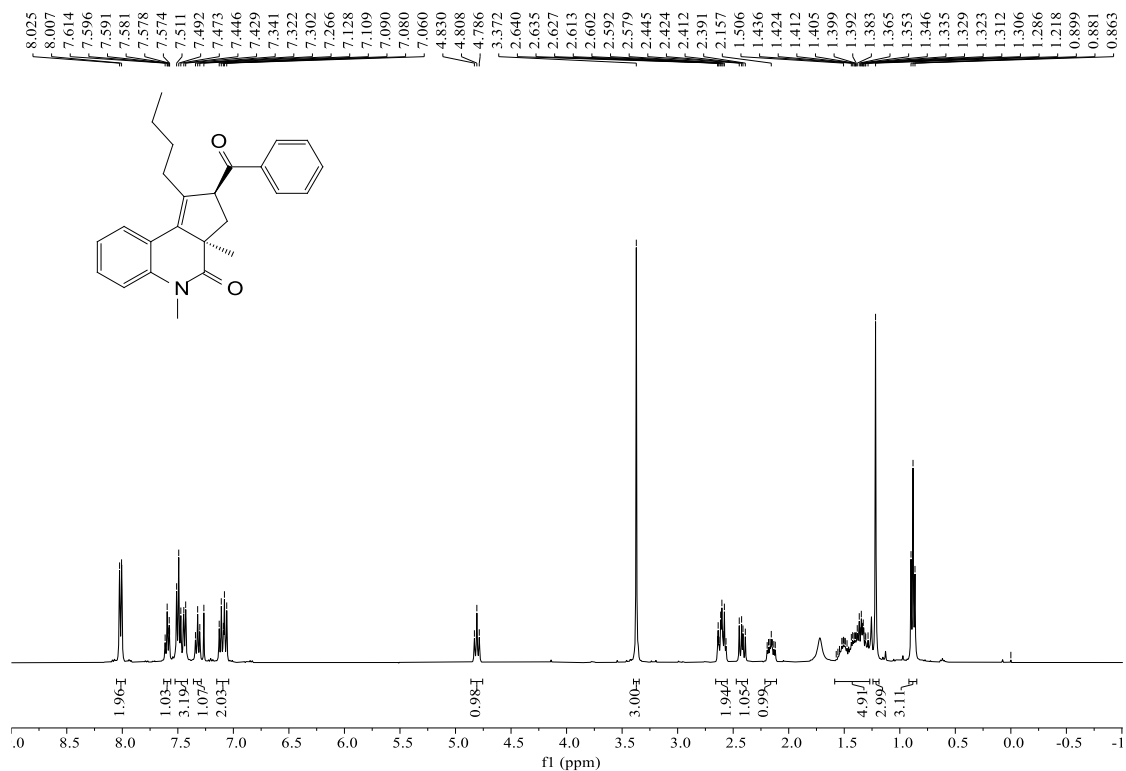
¹H NMR spectra of *cis*-3ae (400 MHz, CDCl₃)



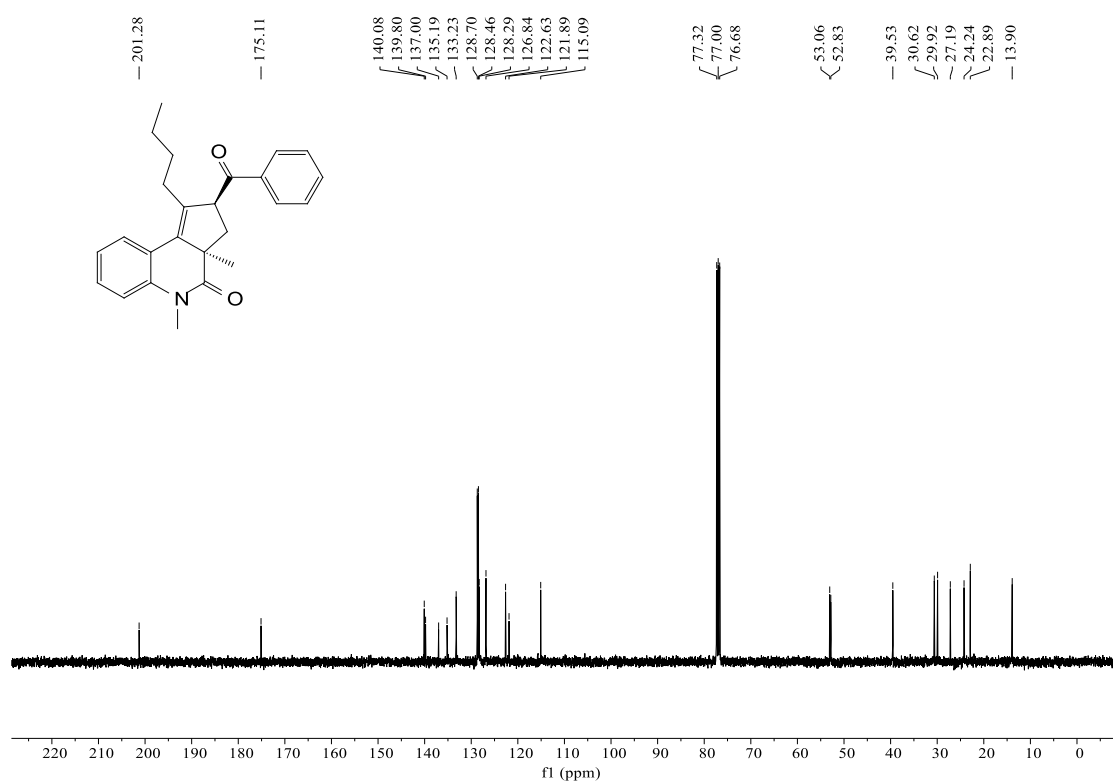
¹³C NMR spectra of *cis*-3ae (100 MHz, CDCl₃)



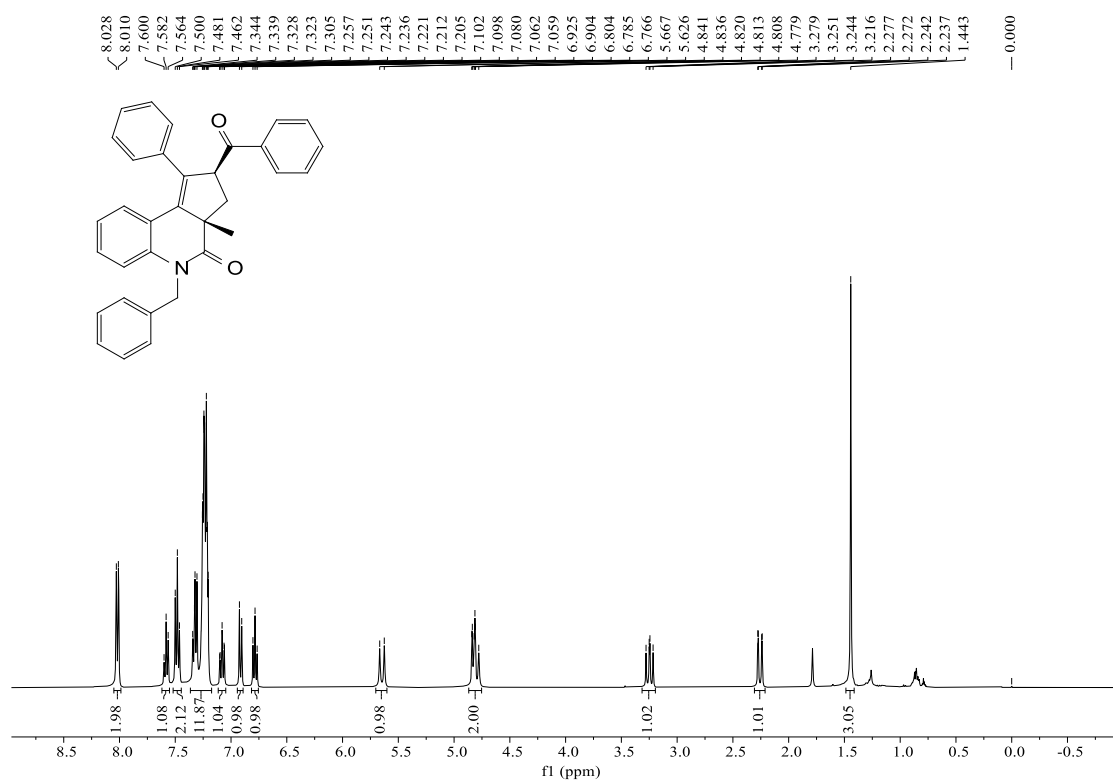
¹H NMR spectra of *trans*-3ae (400 MHz, CDCl₃)



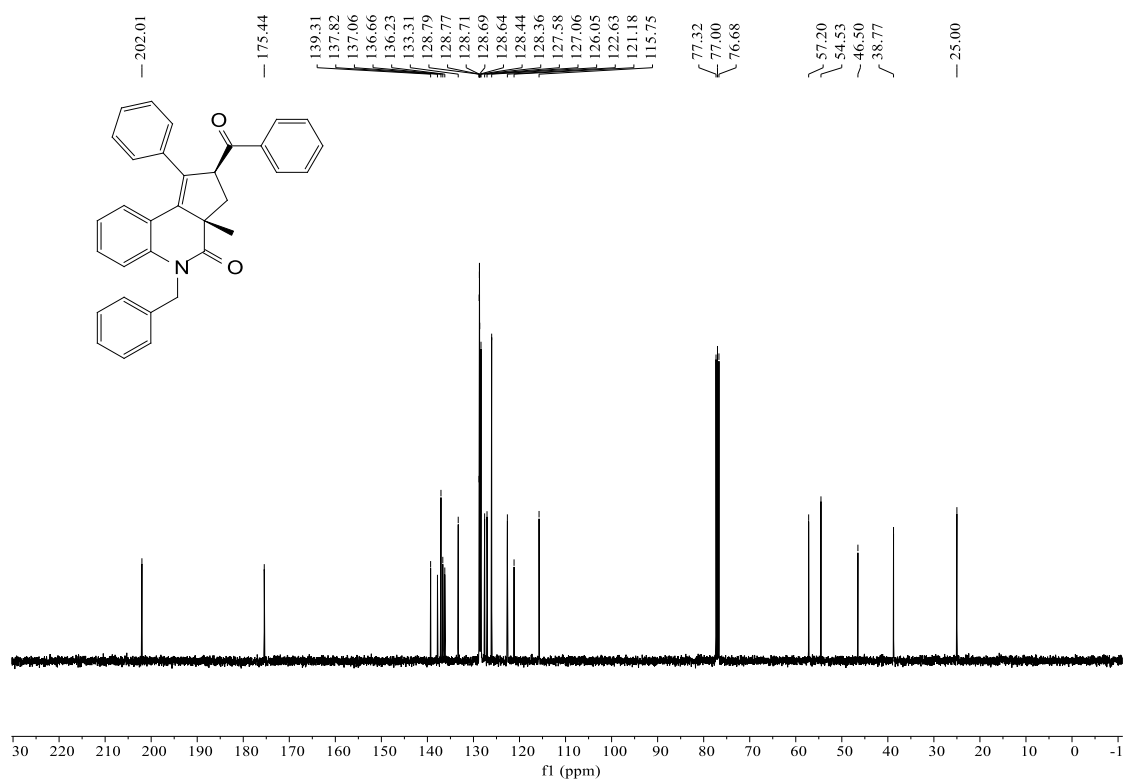
¹³C NMR spectra of *trans*-3ae (100 MHz, CDCl₃)



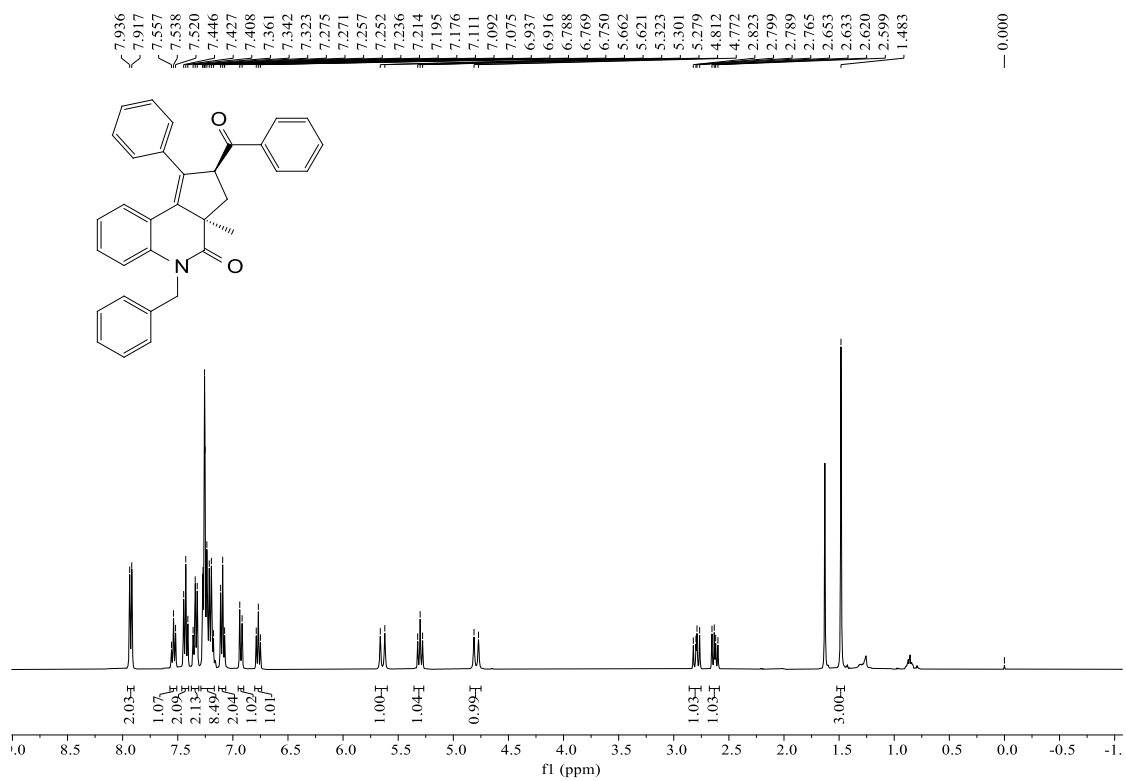
¹H NMR spectra of *cis*-3af (400 MHz, CDCl₃)



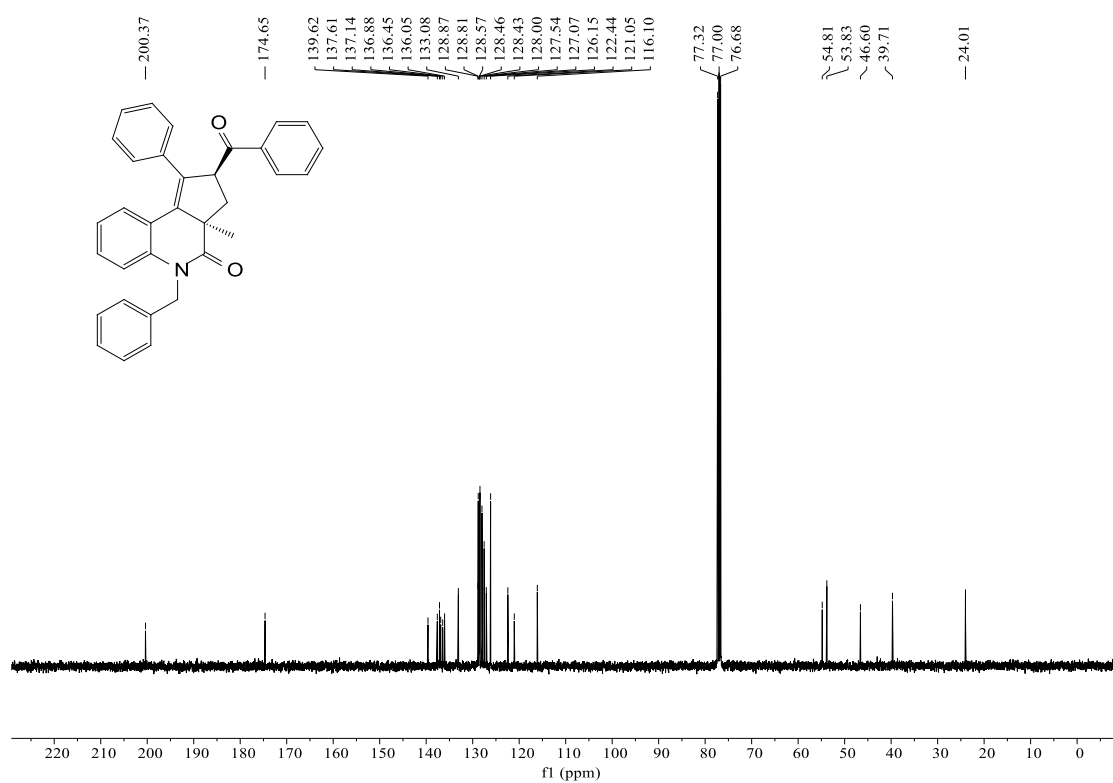
¹³C NMR spectra of *cis*-3af (100 MHz, CDCl₃)



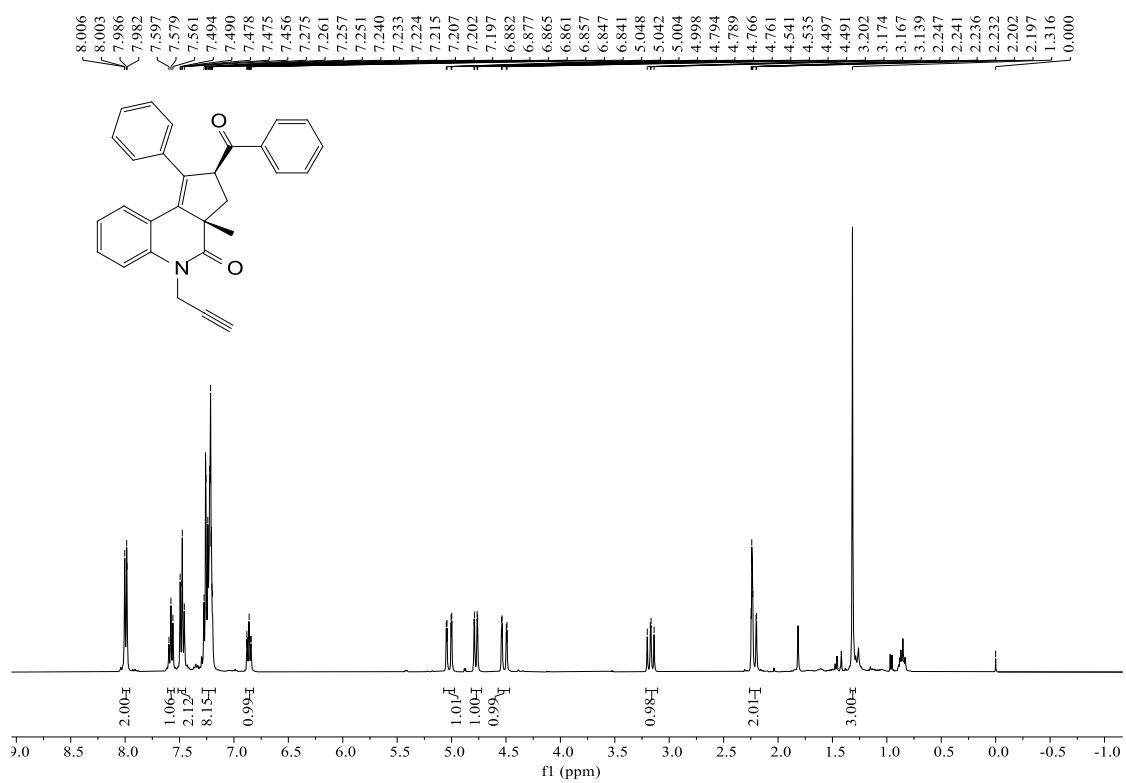
¹H NMR spectra of *trans*-3af (400 MHz, CDCl₃)



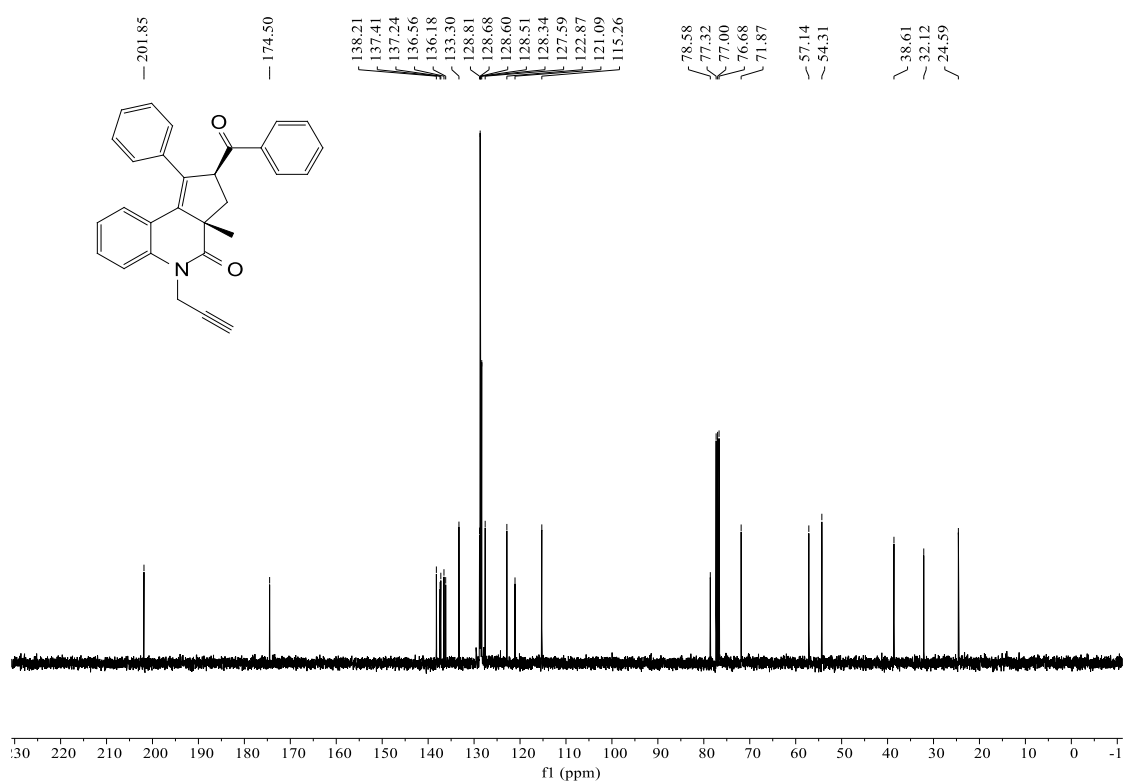
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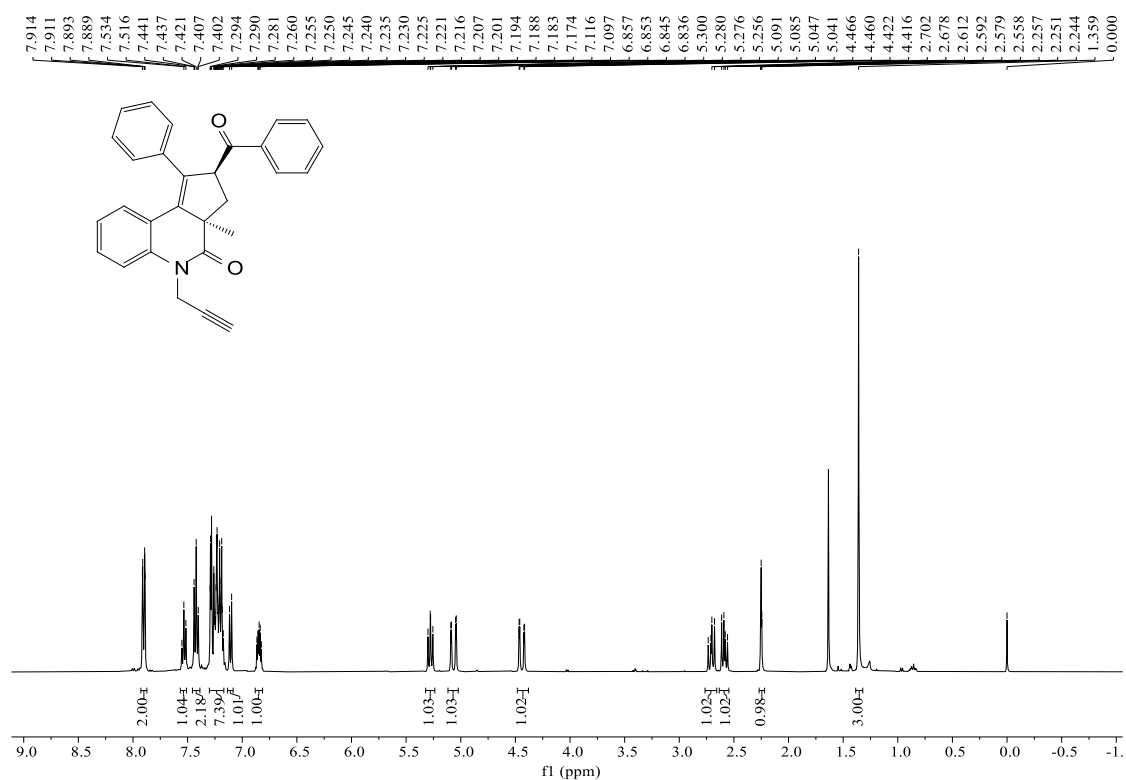
^1H NMR spectra of *cis*-3ag (400 MHz, CDCl_3)



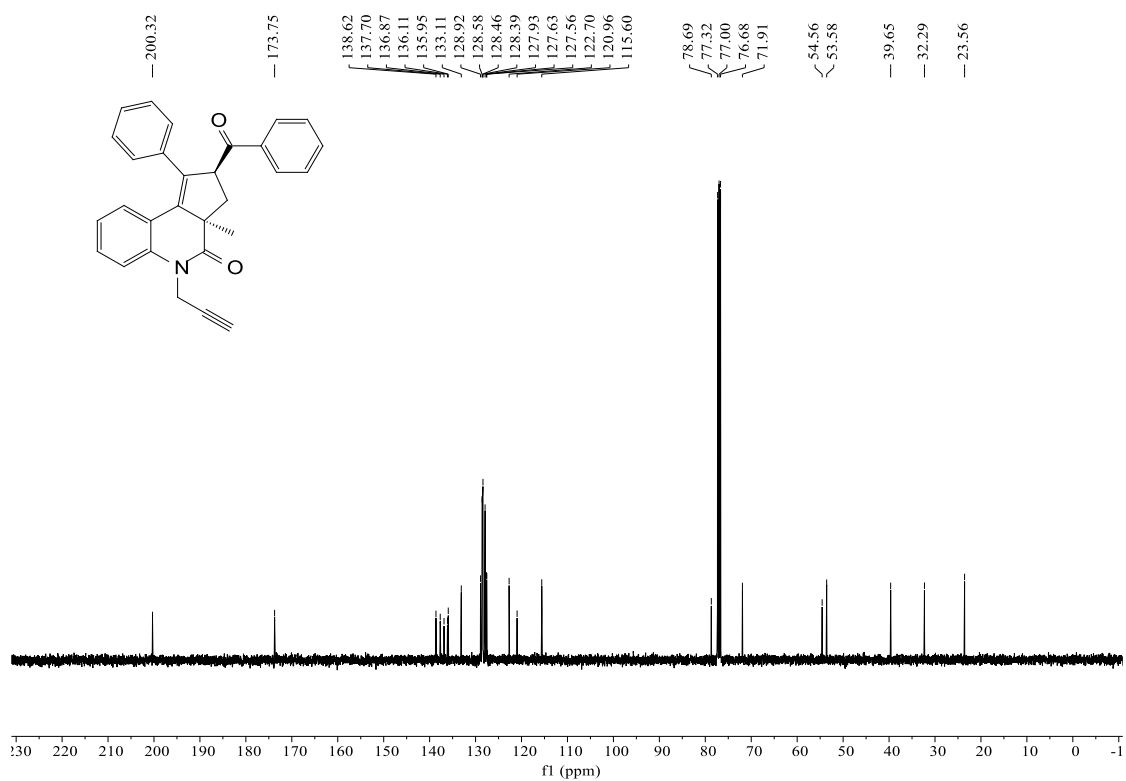
^{13}C NMR spectra of *cis*-3ag (100 MHz, CDCl_3)



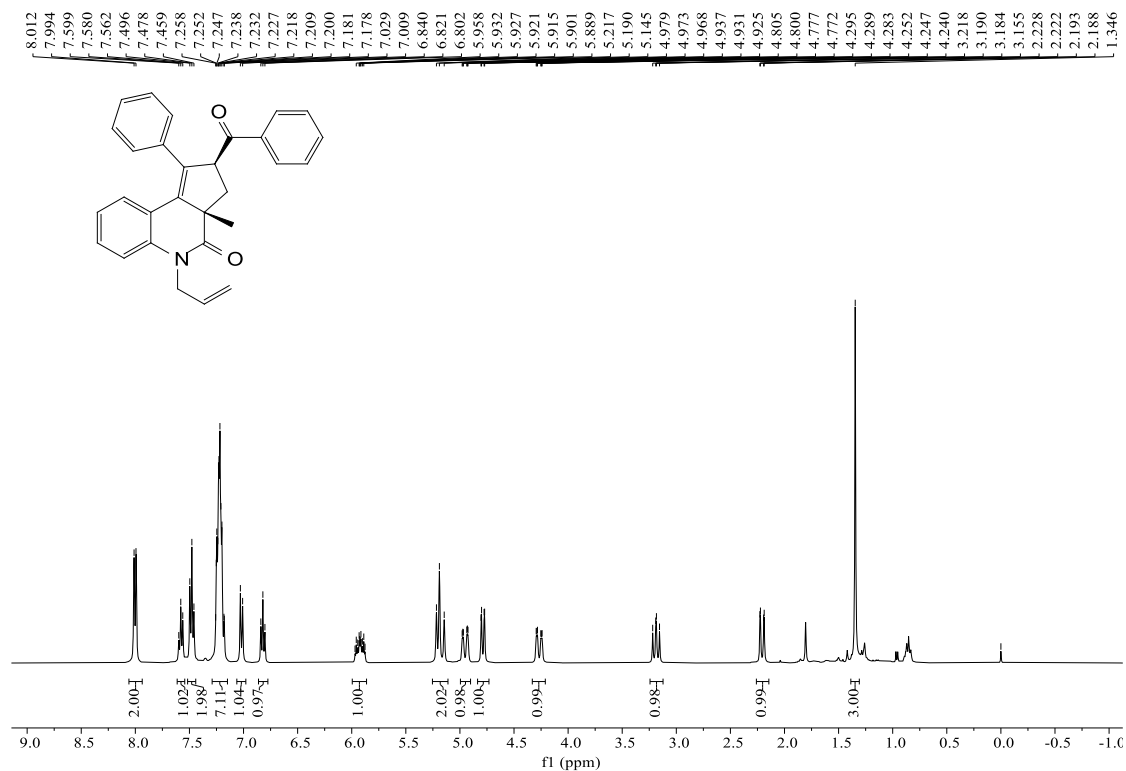
¹H NMR spectra of *trans*-3ag (400 MHz, CDCl₃)



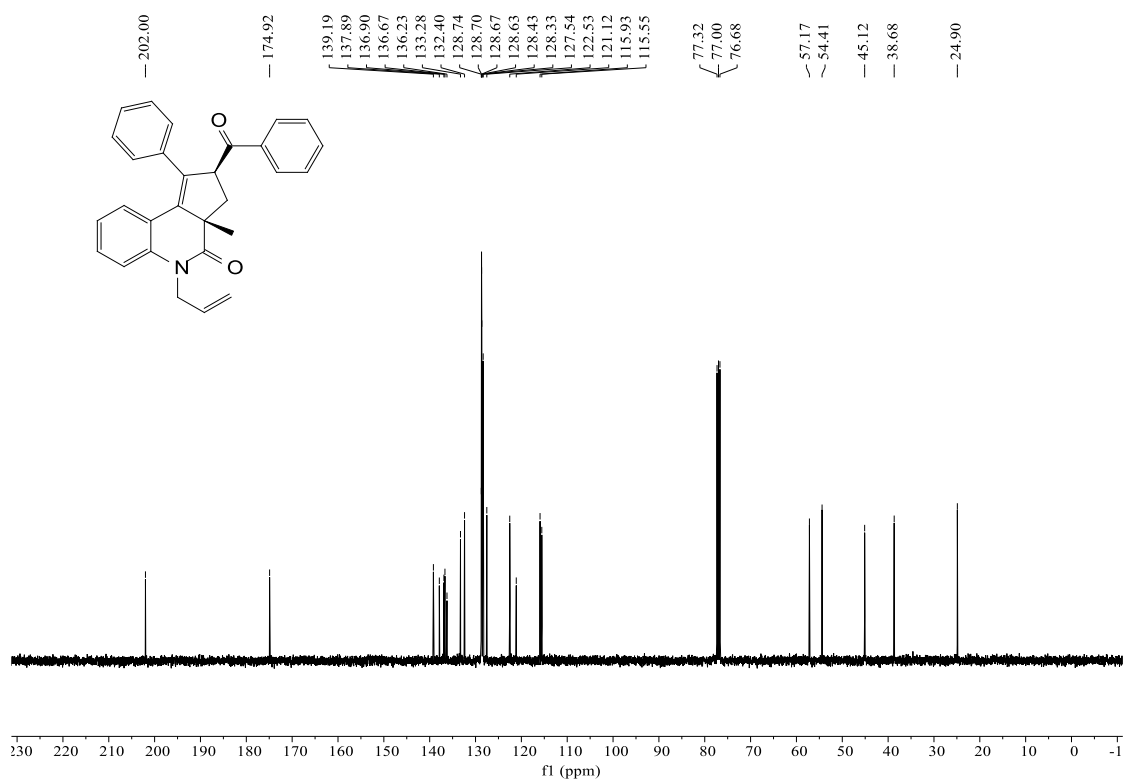
¹³C NMR spectra of *trans*-3ag (100 MHz, CDCl₃)



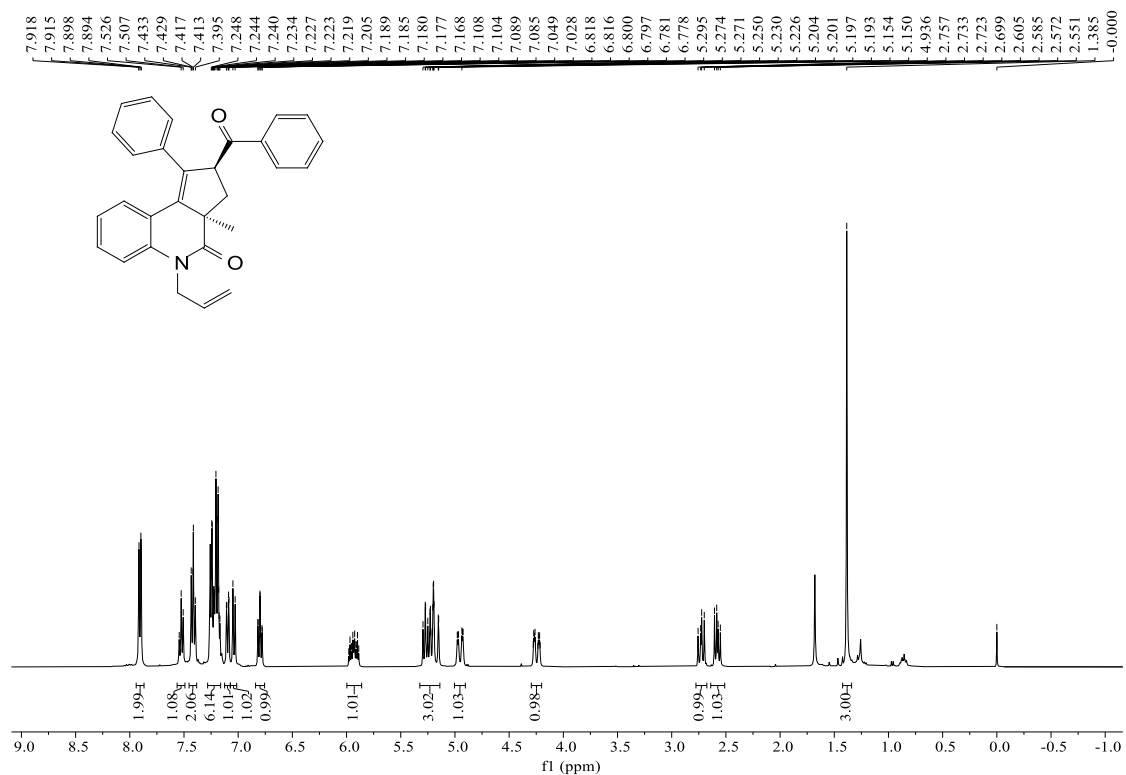
¹H NMR spectra of *cis*-3ah (400 MHz, CDCl₃)



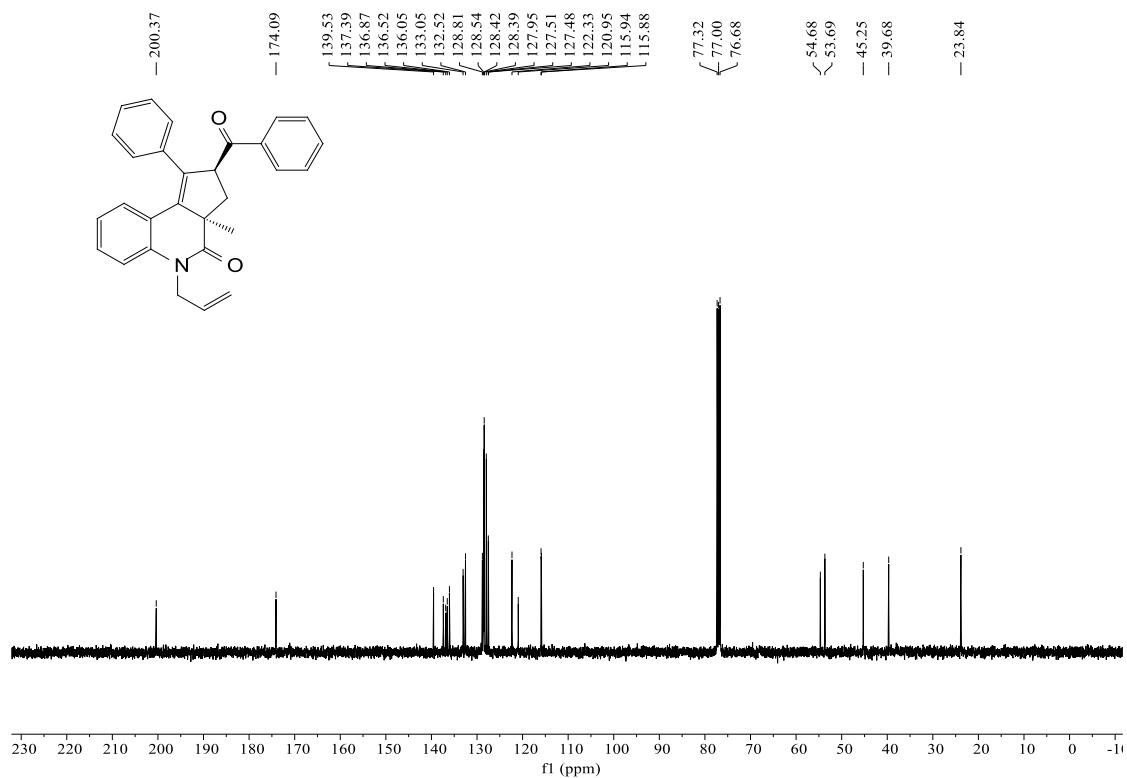
¹³C NMR spectra of *cis*-3ah (100 MHz, CDCl₃)



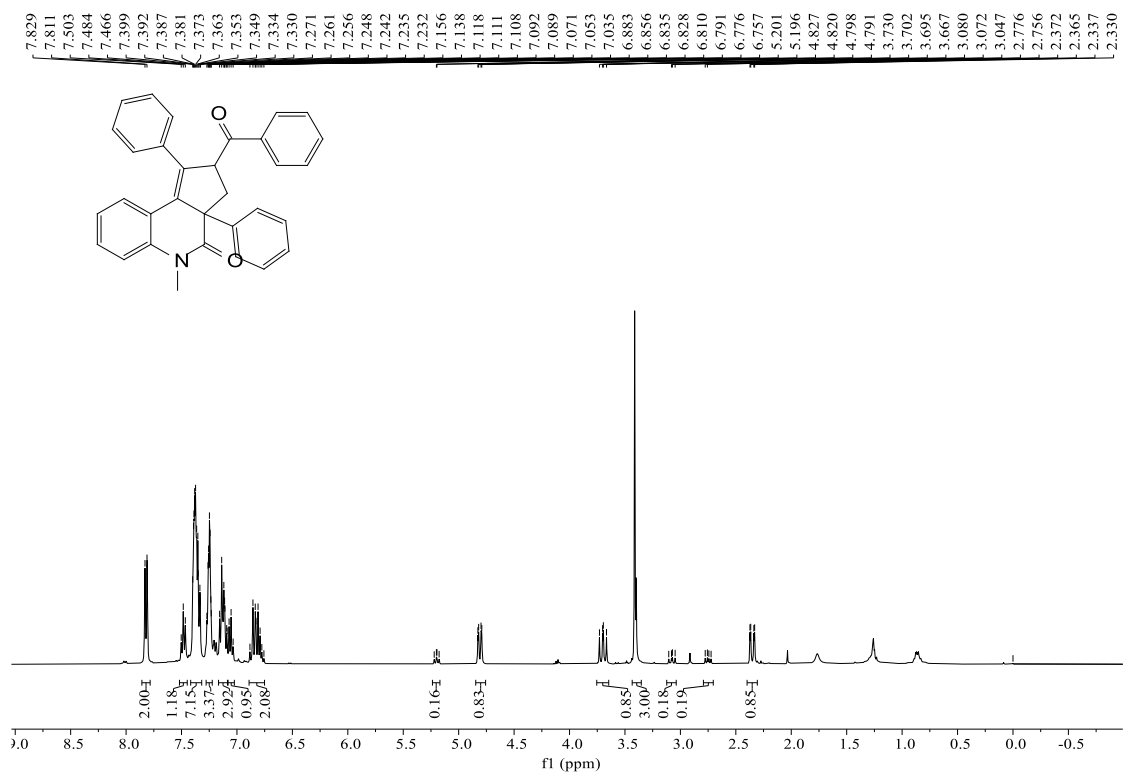
¹H NMR spectra of *trans*-3ah (400 MHz, CDCl₃)



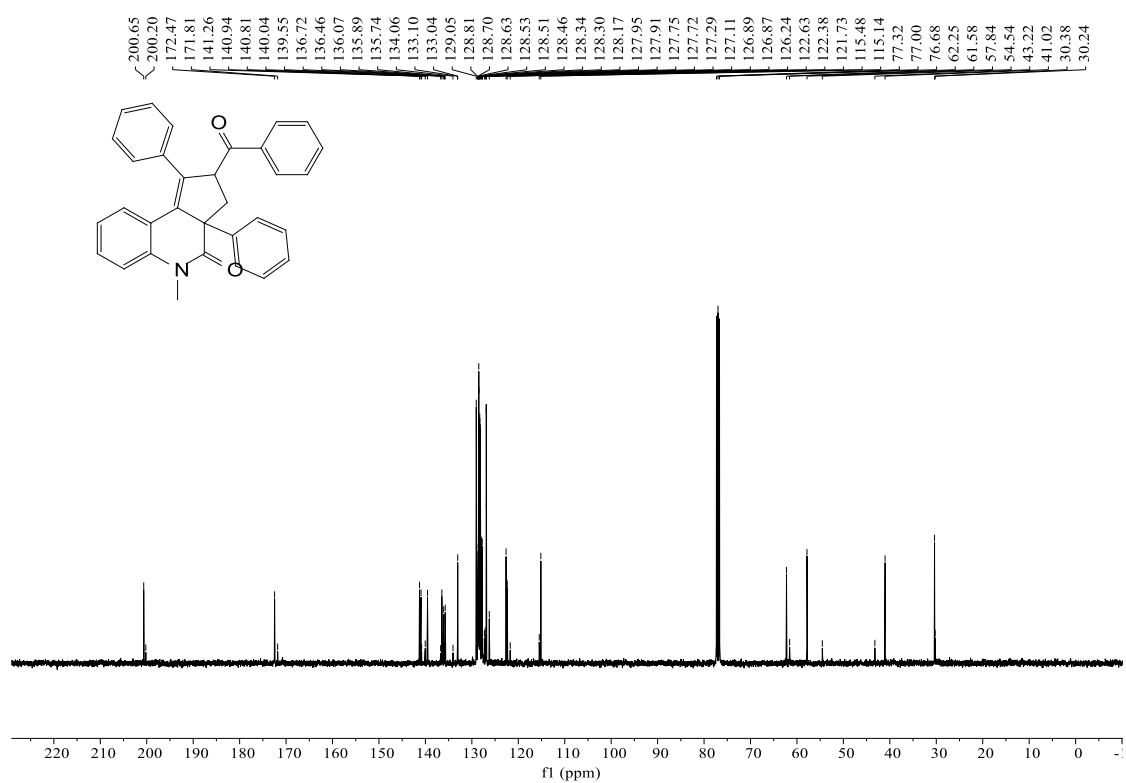
¹³C NMR spectra of *trans*-3ah (100 MHz, CDCl₃)



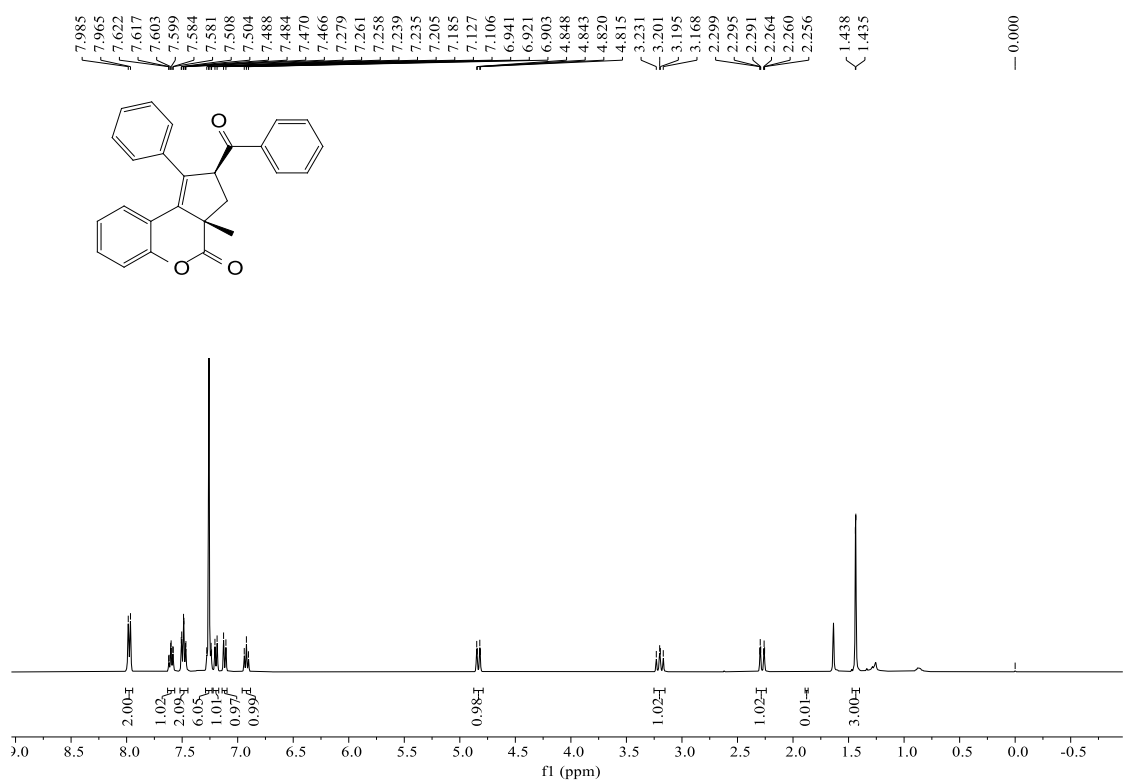
¹H NMR spectra of *trans*-3aj + *cis*-3aj (400 MHz, CDCl₃)



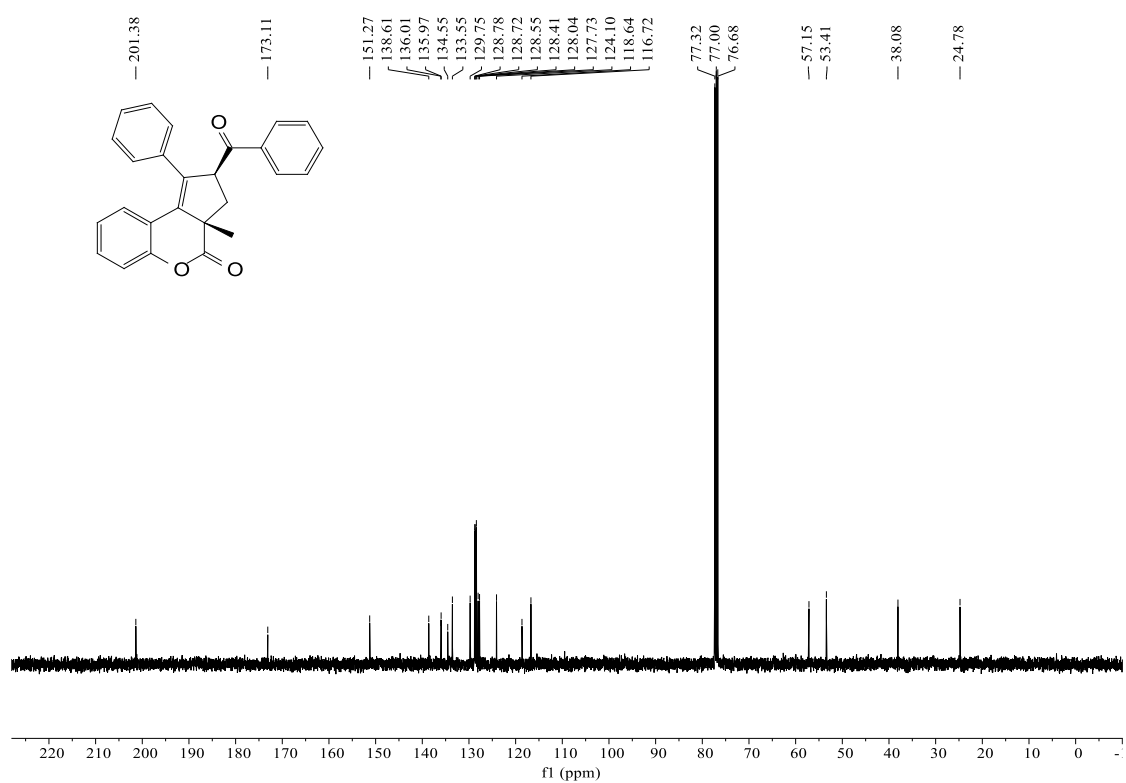
¹³C NMR spectra of *trans*-3aj + *cis*-3aj (100 MHz, CDCl₃)



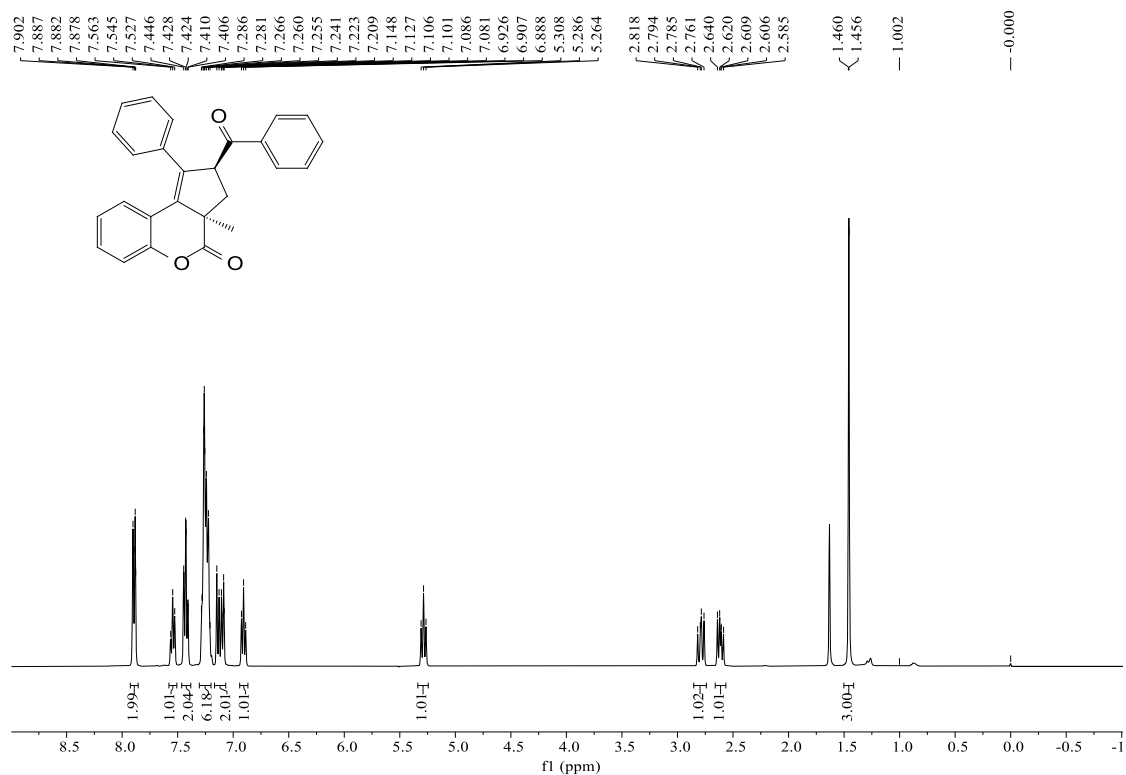
¹H NMR spectra of *cis*-3ak (400 MHz, CDCl₃)



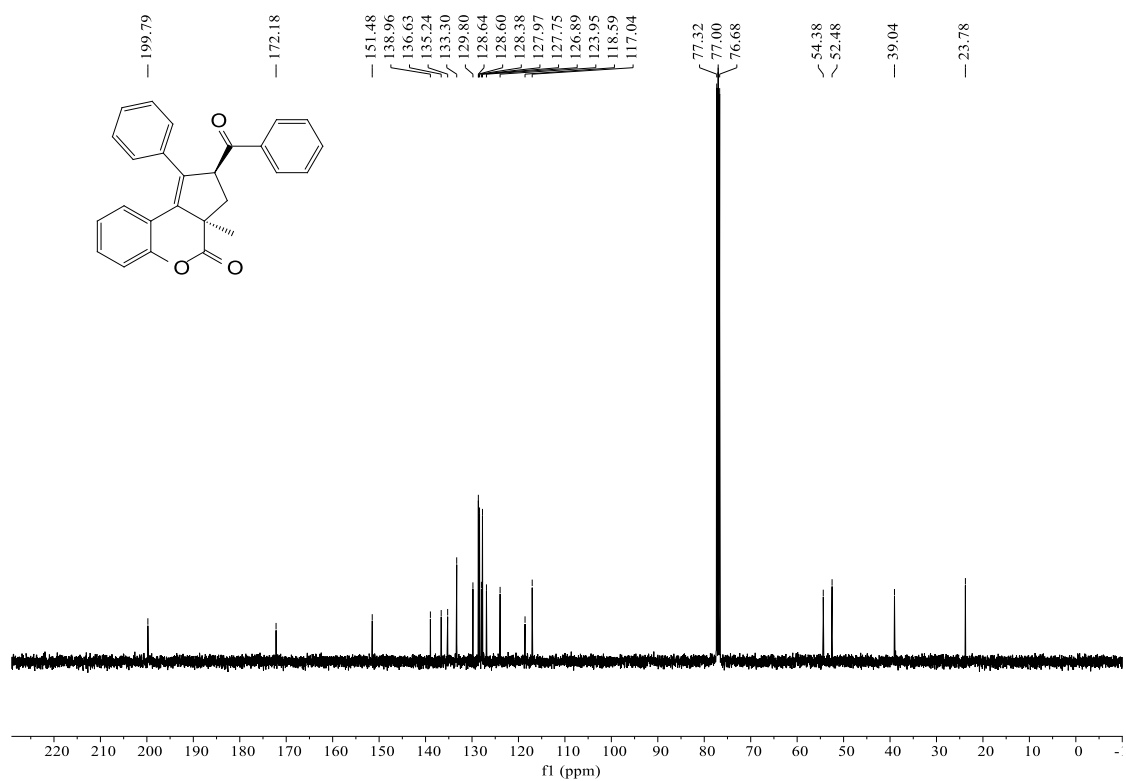
¹³C NMR spectra of *cis*-3ak (100 MHz, CDCl₃)



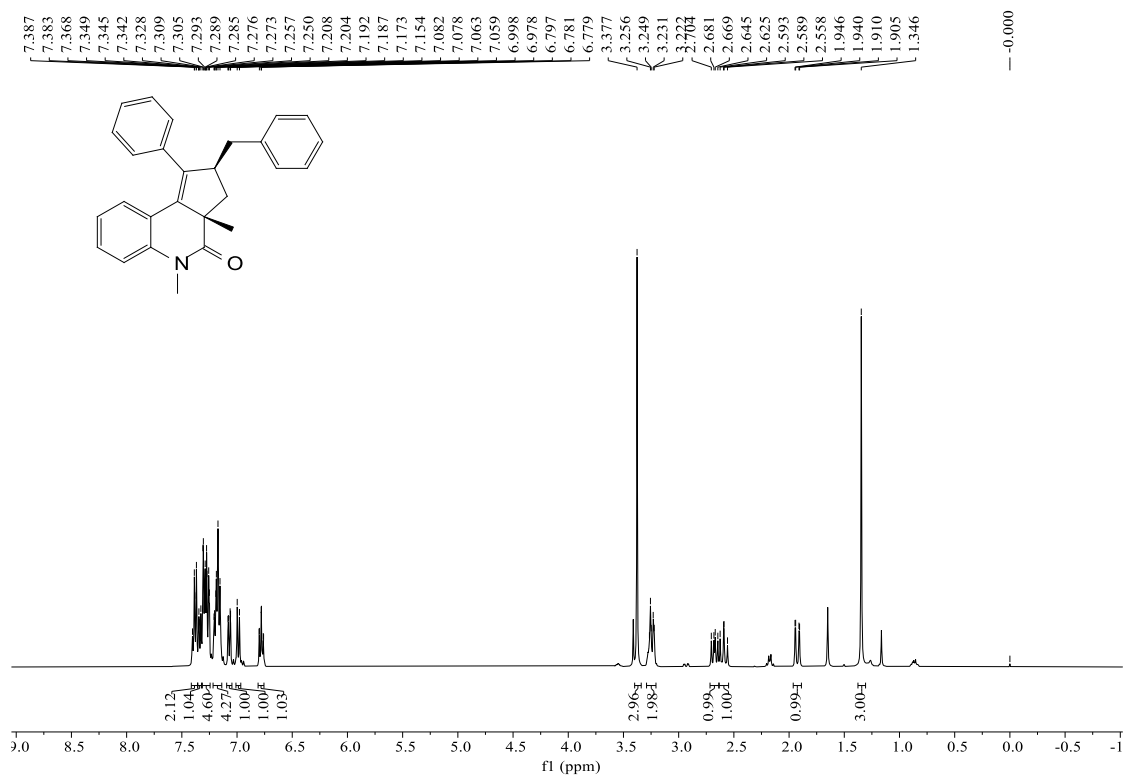
¹H NMR spectra of *trans*-3ak (400 MHz, CDCl₃)



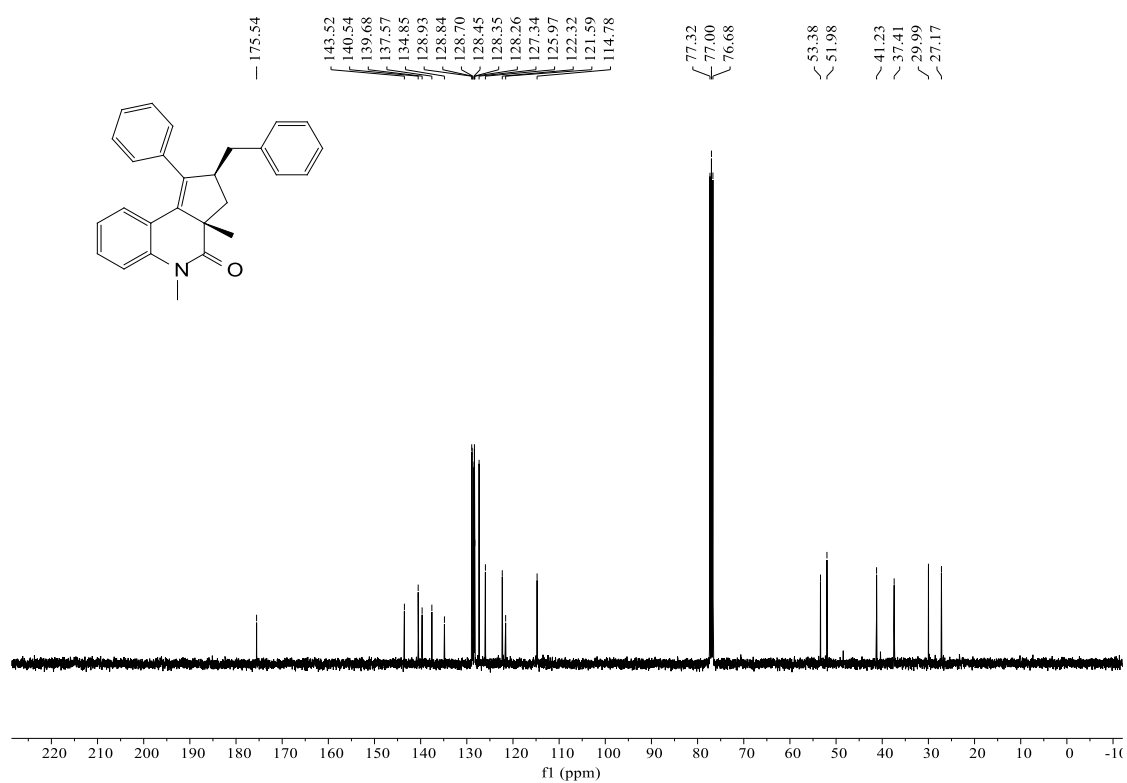
¹³C NMR spectra of *trans*-3ak (100 MHz, CDCl₃)



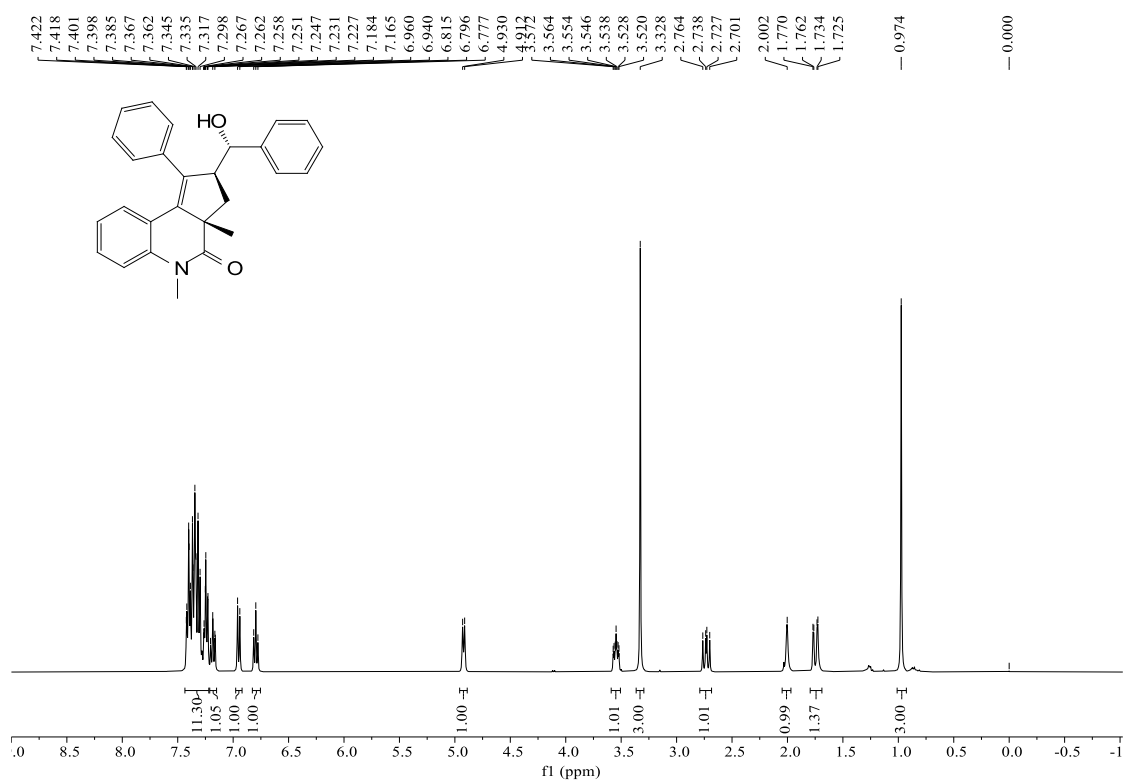
¹H NMR spectra of 4a (400 MHz, CDCl₃)



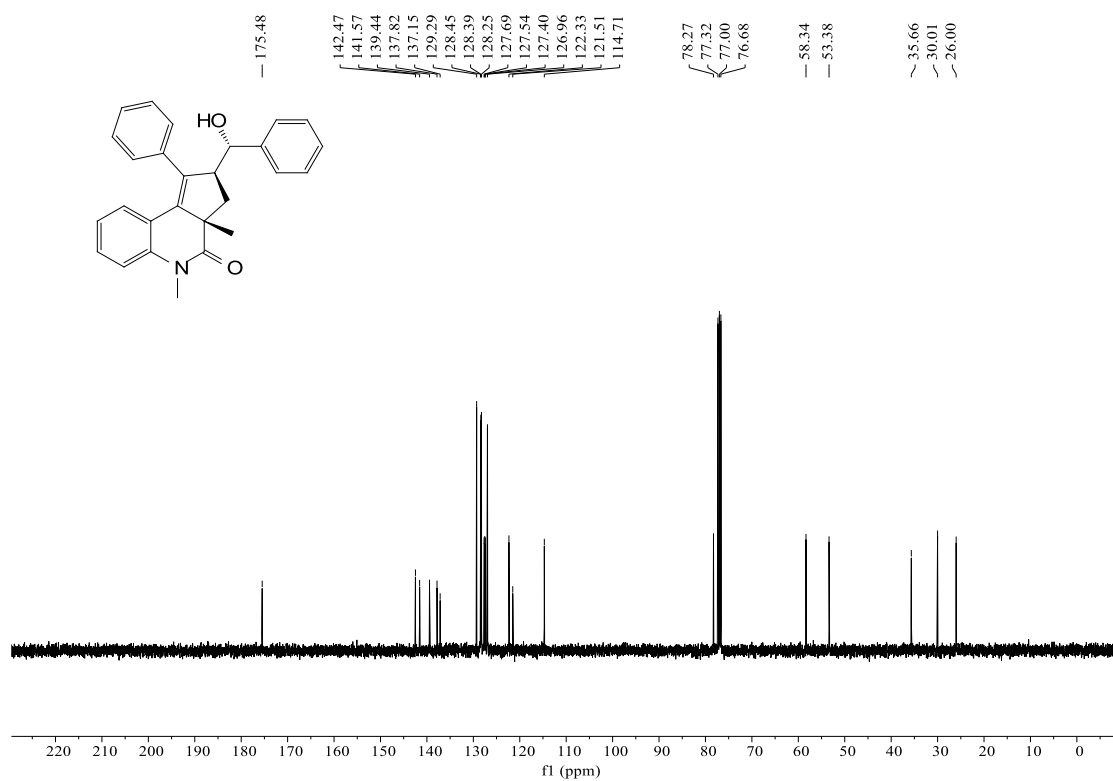
¹³C NMR spectra of 4a (100 MHz, CDCl₃)



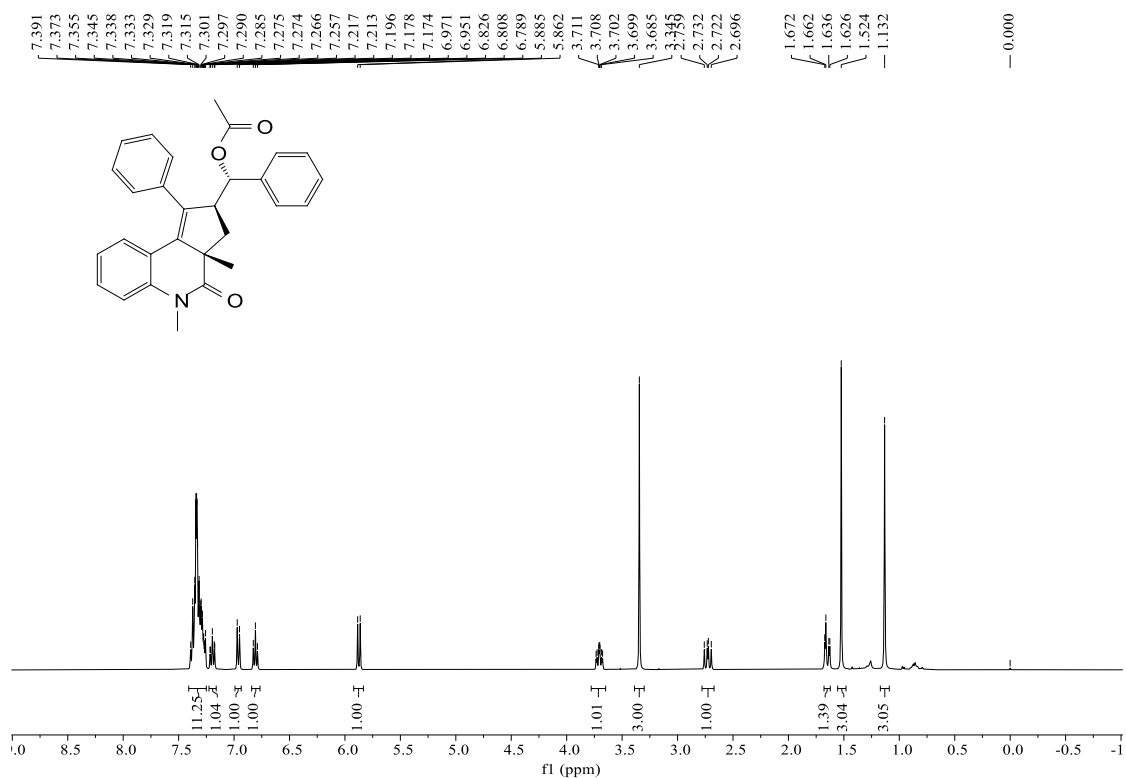
¹H NMR spectra of 4b (400 MHz, CDCl₃)



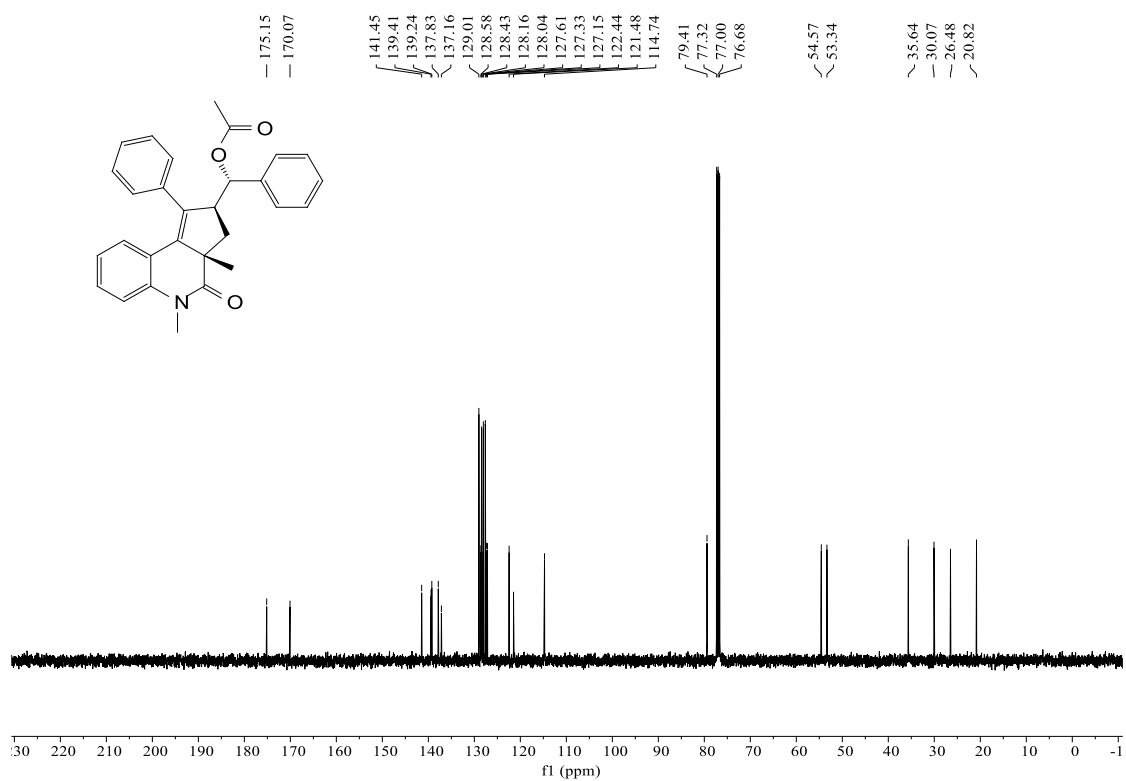
¹³C NMR spectra of 4b (100 MHz, CDCl₃)



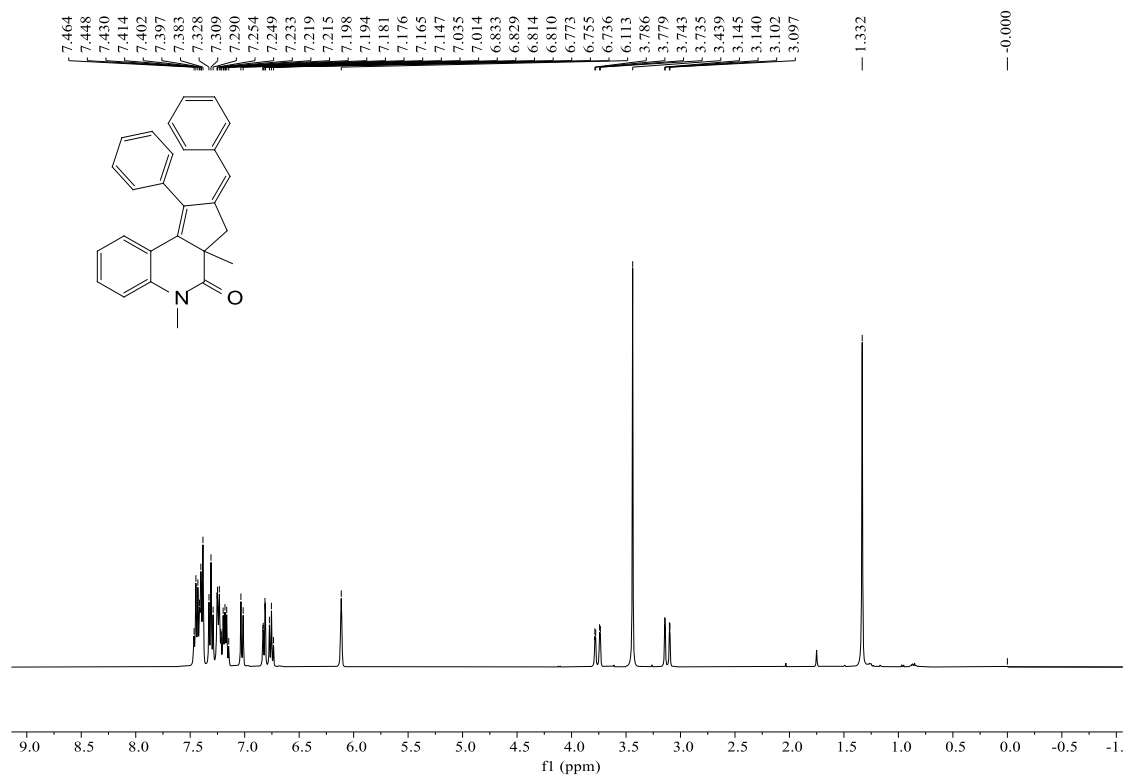
¹H NMR spectra of 4c (400 MHz, CDCl₃)



¹³C NMR spectra of 4c (100 MHz, CDCl₃)



¹H NMR spectra of 4d (400 MHz, CDCl₃)



¹³C NMR spectra of 4d (100 MHz, CDCl₃)

