Supporting Information

An expeditious FeCl₃-catalyzed cascade 1,4-conjugate addition/annulation/1,5-H shift sequence for modular access of all-pyranomoiety-substituted chromenes

Xinwei He,a,* Ruxue Li,a Pui Ying Choy,b,c Jiahui Duan,a Zhenzhen Yin,a Keke Xu,a Qiang Tang,a Rong-Lin Zhong,b Yongjia Shang,a,* and Fuk Yee Kwongb,c,*

aKey Laboratory of Functional Molecular Solids, Ministry of Education, College of Chemistry and Materials Science, Anhui Normal University, Wuhu 241000, P.R. China
bState Key Laboratory of Synthetic Chemistry and Department of Chemistry, The Chinese University of Hong Kong, New Territories, Shatin, Hong Kong SAR, P.R. China
cShenzhen Center of Novel Functional Molecules, Shenzhen Municipal Key Laboratory of Chemical Synthesis of Medicinal Organic Molecules, CUHK Shenzhen Research Institute, No. 10. Second Yuexing Road, Shenzhen 518507, P.R. China

Email: xinweihe@mail.ahnu.edu.cn, shyj@mail.ahnu.edu.cn, fykwong@cuhk.edu.hk

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S1
1. General consideration

Unless otherwise specified, all reagents and starting materials were purchased from commercial sources and used as received without purification. The solvents were used directly without purification unless stated. The dried acetonitrile used in screening was distilled from calcium hydride under nitrogen using standard procedures. All cascade reactions were performed in a resealable screw-capped Schlenk flask (approximately 20 mL volume) in the presence of Teflon-coated magnetic stirrer bar (4.5 mm × 12 mm). Thin layer chromatography was conducted on precoated silica gel 60 F254 plates. Flash chromatography was performed using 200-300 mesh silica gel with the indicated solvent system according to standard techniques. Melting points were measured on an uncorrected Melting Point instrument. The $^1$H and $^{13}$C NMR spectra were recorded on a 400 MHz and 100 MHz NMR spectrometers, unless otherwise specified. Chemical shifts (δ) in parts per million were reported relative to the residual signals of chloroform (7.26 ppm for $^1$H and 77.0 ppm for $^{13}$C), and all $^{13}$C NMR were recorded with proton broadband decoupling and indicated as $^{13}$C($^1$H) NMR. Multiplicities are described as s (singlet), d (doublet), t (triplet), q (quartet), or m (multiplet), and the coupling constants ($J$) are reported in Hertz (Hz). HRMS analysis with a quadrupole time-of-flight mass spectrometer yielded ion mass/charge (m/z) ratios in atomic mass units.
## 2. Reaction optimization

### Table S1. Optimization of reaction conditions

| entry | Catalyst (mol%) | solvent  | temp./ °C | yield/%b | E:Z |
|-------|-----------------|----------|-----------|----------|-----|
| 1     | FeCl$_3$ (50)   | MeCN     | 80        | 82       | 4:1 |
| 2     | Sc(OTf)$_3$ (50)| MeCN     | 80        | 71       | 4:1 |
| 3     | CuI (50)        | MeCN     | 80        | 59       | 4:1 |
| 4     | AgNO$_3$ (50)   | MeCN     | 80        | 49       | 4:1 |
| 5     | CuBr$_2$ (50)   | MeCN     | 80        | trace    | 4:1 |
| 6     | ZnI$_2$ (50)    | MeCN     | 80        | 45       | 4:1 |
| 7     | Fe(acac)$_3$ (50)| MeCN | 80        | 53       | 4:1 |
| 8     | --              | MeCN     | 80        | trace    | -   |
| 9c    | FeCl$_3$ (50)   | MeCN     | 80        | 72       | 4:1 |
| 10d   | FeCl$_3$ (50)   | MeCN     | 80        | 76       | 4:1 |
| 11    | FeCl$_3$ (20)   | MeCN     | 80        | 83       | 4:1 |
| 12    | FeCl$_3$ (10)   | MeCN     | 80        | 65       | 4:1 |
| 13    | FeCl$_3$ (20)   | DCE      | 80        | nd       | -   |
| 14    | FeCl$_3$ (20)   | THF      | 80        | nd       | -   |
| 15    | FeCl$_3$ (20)   | DMF      | 80        | 31       | 4:1 |
| 16    | FeCl$_3$ (20)   | EtOH     | 80        | 10       | 4:1 |
| 17    | FeCl$_3$ (20)   | toluene  | 80        | 15       | 4:1 |
| 18    | FeCl$_3$ (20)   | MeCN     | rt        | nr       | -   |
| 19    | FeCl$_3$ (20)   | MeCN     | 60        | 32       | 9:5 |
| 20    | FeCl$_3$ (20)   | MeCN     | 100       | 72       | >20:1 |
| 21    | FeCl$_3$ (20)   | MeCN     | 120       | 84       | >20:1 |
| 22e   | FeCl$_3$ (20)   | MeCN     | 120       | 84       | >20:1 |
| 23f   | FeCl$_3$ (20)   | MeCN     | 120       | 77       | >20:1 |
| 24g   | FeCl$_3$ (20)   | MeCN     | 120       | 64       | >20:1 |
| 25h   | FeCl$_3$ (20)   | MeCN     | 120       | 64       | >20:1 |

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*aReaction conditions: 2-(3-phenyl-1-(pyrrolidin-1-yl)prop-2-yn-1-yl)phenol (1a) (0.3 mmol), (E)-3-(methylthio)-1-phenyl-3-(phenylamino)prop-2-en-1-one (2a) (0.2 mmol), and catalyst (50 mol%) in undried solvent (2 mL) at indicated temperature for 12 h. Isolated yields. 1.0 equivalent of 1a was used. 2.0 equivalent of 1a was used. For 18 h. For 8 h. For 5 h. Dried MeCN was used. nd = not detected. nr = no reaction.
3. **General procedures for the synthesis of propargylamines 1**

![Chemical structure](attachment:structure.png)

To a 25 mL round-bottom flask equipped with a magnetic stir bar were added pyrrolidine (1.2 mmol), aldehyde (1.0 mmol), acetylene (1.2 mmol), copper(I) iodide (10 mol%) and toluene (3 mL). The mixture was degassed and backfilled with nitrogen, and then stirred in an oil bath preheated to 100 °C for 5 h (monitored by TLC). After the reaction completed (as determined using TLC), the reaction mixture was cooled to room temperature, diluted with CH$_2$Cl$_2$ (10 mL) and filtered through a thin pad of silica gel. The filter cake was washed with CH$_2$Cl$_2$, and the combined filtrate was concentrated in vacuum. The crude product was purified by flash column chromatography on silica gel to afford the corresponding propargylamines.

4. **General procedures for the synthesis of N,S-keteneacetals 2**

![Chemical structure](attachment:structure2.png)

A mixture of acetophenone (0.24 mL, 2.0 mmol), sodium hydride (96 mg, 4.0 mmol), and N,N-dimethylformamide (5 mL) was stirred at room temperature for 30 minutes. Then, aryl isothiocyanates was added dropwise at 0 °C in an ice-water bath, and stirring was continued at room temperature for 1 h. Iodomethane (0.18 mL, 2.4 mmol) was then added dropwise, stirring for another 1 h. Upon completion of the reaction, the reaction mixture was extracted with CH$_2$Cl$_2$ (3 × 10 mL), and washed with brine. The organic layers were combined, dried over Na$_2$SO$_4$, filtered, and then evaporated under vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:40, v/v) as the elution solvent to give corresponding N,S-keteneacetals.
5. General procedures for the synthesis of alkenyl-iminochromene 3

A mixture of propargylamines 1 (0.3 mmol), N,S-keteneacetals 2 (0.2 mmol), and FeCl$_3$ (20-50 mol%) were added under air atmosphere to a resealable screw-capped Schlenk tube. Undried acetonitrile (2 mL) was then added. The tube sealed with a Teflon-coated cap and the resulting mixture was stirred in an oil bath preheated to 120 °C for 12 h (monitored by TLC). Upon completion of the reaction, the reaction mixture was cooled to room temperature, extracted with CH$_2$Cl$_2$ (3 × 10 mL), and washed with brine. The organic layers were combined, dried over Na$_2$SO$_4$, filtered, and then evaporated under a vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:20, v/v) as the elution solvent to give desired products 3.

6. Large scale synthesis of compound 3aa

A mixture of 1a (3.75 mmol), 2a (2.5 mmol), and FeCl$_3$ (20 mol%) were added under air atmosphere to a resealable screw-capped Schlenk tube. Undried acetonitrile (10 mL) was then added. The tube sealed with a Teflon-coated cap and the resulting mixture was stirred in an oil bath preheated to 120 °C for 12 h (monitored by TLC). Upon completion of the reaction, the reaction mixture was cooled to room temperature, extracted with CH$_2$Cl$_2$ (3 × 20 mL), and washed with brine. The organic layers were combined, dried over Na$_2$SO$_4$, filtered, and then evaporated under a vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:20, v/v) as the elution solvent to give
desired product 3aa in 69% yield (736 mg).

7. **General procedures for the synthesis of aryl-iminochromene 5**

A mixture of 4 (0.3 mmol), N,S-keteneacetics 2a (0.2 mmol), and FeCl₃ (20 mol%) were added under air atmosphere to a resealable screw-capped Schlenk tube. Undried acetonitrile (2 mL) was then added. The tube sealed with a Teflon-coated cap and the resulting mixture was stirred in an oil bath preheated to 120 °C for 12 h (monitored by TLC). Upon completion of the reaction, the reaction mixture was cooled to room temperature, extracted with CH₂Cl₂ (3 × 10 mL), and washed with brine. The organic layers were combined, dried over Na₂SO₄, filtered, and then evaporated under a vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:20, v/v) as the elution solvent to give desired products 5.

8. **General procedures for the synthesis of compound 6aq**

A mixture of 1a (0.3 mmol), 2q (0.2 mmol), and FeCl₃ (20 mol%) were added under air atmosphere to a resealable screw-capped Schlenk tube. Dried acetonitrile (2 mL) was then added. The tube sealed with a Teflon-coated cap and the resulting mixture was stirred in an oil bath preheated to 120 °C for 5 h (monitored by TLC). Upon completion of the reaction, the reaction mixture was cooled to room temperature, extracted with CH₂Cl₂ (3 × 10 mL), and washed with brine. The organic layers were
combined, dried over Na$_2$SO$_4$, filtered, and then evaporated under a vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:20, v/v) as the elution solvent to give desired product 6a in 46% yield (35 mg).

9. General procedures for the synthesis of compound 6ia

![Chemical structures and reaction scheme]

A mixture of 1i (0.3 mmol), 2a (0.2 mmol), and FeCl$_3$ (20 mol%) were added under air atmosphere to a resealable screw-capped Schlenk tube. Undried THF (2 mL) was then added. The tube sealed with a Teflon-coated cap and the resulting mixture was stirred in an oil bath preheated to 80 °C for 12 h (monitored by TLC). Upon completion of the reaction, the reaction mixture was cooled to room temperature, extracted with CH$_2$Cl$_2$ (3 × 10 mL), and washed with brine. The organic layers were combined, dried over Na$_2$SO$_4$, filtered, and then evaporated under a vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:20, v/v) as the elution solvent to give desired product 6ia in 70% yield (64 mg).

10. General procedures for the synthesis of compound 8

![Chemical structures and reaction scheme]

**Method A**: To a mixture of phenyl((Z)-2-(phenylimino)-4-((E)-styril)-2H-chromen-3-yl)methanone (3aa) (84 mg, 0.2 mmol) and I$_2$ (152 mg, 0.6 mmol) in undried ethanol (2 mL) under air atmosphere to a resealable screw-capped Schlenk tube. The tube sealed
with a Teflon-coated cap and the resulting mixture was stirred in an oil bath preheated to 80 °C for 6 h (monitored by TLC). Upon completion of the reaction, the reaction mixture was cooled to room temperature, and the solvent was removed under reduced pressure. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:20, v/v) as the elution solvent to give desired product 8 in 92% yield (65 mg).

**Method B:** A mixture of 1a (0.3 mmol), 2r (0.2 mmol), and FeCl$_3$ (50 mol%) were added under air atmosphere to a resealable screw-capped Schlenk tube. Undried acetonitrile (2 mL) was then added. The tube sealed with a Teflon-coated cap and the resulting mixture was stirred in an oil bath preheated to 120 °C for 5 h (monitored by TLC). Upon completion of the reaction, the reaction mixture was cooled to room temperature, extracted with CH$_2$Cl$_2$ (3 × 10 mL), and washed with brine. The organic layers were combined, dried over Na$_2$SO$_4$, filtered, and then evaporated under a vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:20, v/v) as the elution solvent to give desired product 8 in 78% yield (55 mg).

11. **General procedures for the synthesis of compound 9**

To a solution of 4-vinyliminochromone 3 (0.2 mmol), in THF (2 mL) was added solid sodium borohydride (5-10 equiv.) and the mixture was stirred in an oil bath preheated to 80 °C under air atmosphere for 12 h (monitored by TLC). Upon completion of the reaction, the reaction mixture was cooled to room temperature, extracted with CH$_2$Cl$_2$ (3 × 10 mL), and washed with brine. The combined organic layers were dried over Na$_2$SO$_4$, filtered and concentrated under reduced pressure. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:20, v/v) as the elution solvent to give desired product 9.
12. General procedures for deuterium-labelling experiment

A mixture of propargylamines 1a (0.15 mmol), N,S-keteneacetals 2a (0.1 mmol), and FeCl₃ (20 mol%) were added under air atmosphere to a resealable screw-capped Schlenk tube. Undried acetonitrile (2 mL) and ethanol-d₆ (20 μL) was then added. The tube sealed with a Teflon-coated cap and the resulting mixture was stirred in an oil bath preheated to 120 °C for 12 h (monitored by TLC). Upon completion of the reaction, the reaction mixture was cooled to room temperature, extracted with CH₂Cl₂ (3 × 10 mL), and washed with brine. The organic layers were combined, dried over Na₂SO₄, filtered, and then evaporated under a vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:20, v/v) as the elution solvent to give desired products 3aa-d in 65% yield.
13. Characterization data for all compounds

Phenyl(\(Z\)-2-(phenylimino)-4-((\(E\))-styryl)-2\(H\)-chromen-3-yl)methanone  (Scheme 2, compound 3aa)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
\(R_f\) = 0.6) to afford a yellow solid in 84% yield (72 mg); mp 143-144 °C, \(^1\)H NMR (400 MHz, CDCl\(_3\)) 
\(\delta\) 8.08–7.97 (m, 2H), 7.68 (d, \(J = 8.0\) Hz, 1H), 7.58–7.53 (m, 1H), 7.47–7.40 (m, 3H), 7.36–7.30 
(m, 4H), 7.30–7.26 (m, 3H), 7.23–7.18 (m, 1H), 7.17–7.14 (m, 1H), 7.13–7.09 (m, 2H), 7.08–7.04 
(m, 1H), 6.95 (dd, \(J = 24.4\) Hz, 16.4 Hz, 2H); \(^{13}\)C\(\{^1\)H\} NMR (100 MHz, CDCl\(_3\)) \(\delta\) 194.0, 152.8, 147.6, 
145.5, 139.8, 139.2, 136.7, 135.8, 133.5, 131.2, 129.3, 129.0, 128.8, 128.7, 128.4, 127.0, 
125.7, 124.1, 123.9, 123.1, 119.7, 119.4, 116.4; HRMS (ESI-TOF) \(m/z\): [M+H]\(^+\) Calcd for 
C\(_{30}\)H\(_{22}\)NO\(_2\) 428.1645; Found 428.1641.

(\(Z\)-6-Chloro-2-(phenylimino)-4-((\(E\))-styryl)-2\(H\)-chromen-3-yl)(phenyl)methanone  
(Scheme 2, compound 3ba)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
\(R_f\) = 0.5) to afford a yellow solid in 72% yield (66 mg); mp 196-197 °C, \(^1\)H NMR (400 MHz, CDCl\(_3\)) 
\(\delta\) 8.08–7.97 (m, 2H), 7.63 (d, \(J = 2.4\) Hz, 1H), 7.60–7.55 (m, 1H), 7.48–7.43 (m, 2H), 7.38 (dd, \(J = 
8.8\) Hz, 2.4 Hz, 1H), 7.36–7.31 (m, 4H), 7.31–7.26 (m, 3H), 7.14–7.04 (m, 4H), 6.92 (dd, \(J = 50.0\) 
Hz, 16.4 Hz, 2H); \(^{13}\)C\(\{^1\)H\} NMR (100 MHz, CDCl\(_3\)) \(\delta\) 193.5, 151.2, 146.9, 145.2, 139.8, 138.7, 136.5, 
135.6, 133.7, 130.9, 129.8, 129.2, 129.2, 128.8, 128.5, 127.1, 125.3, 124.4, 123.1, 120.9, 119.0, 
117.7; HRMS (ESI-TOF) \(m/z\): [M+H]\(^+\) Calcd for C\(_{30}\)H\(_{21}\)ClNO\(_2\) 462.1255; Found 462.1249.

(\(Z\)-6-Bromo-2-(phenylimino)-4-((\(E\))-styryl)-2\(H\)-chromen-3-yl)(phenyl)methanone  
(Scheme 2, compound 3ca)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
\(R_f\) = 0.4) to afford a yellow solid in 69% yield (70 mg); mp 180-181 °C, \(^1\)H NMR (400 MHz, CDCl\(_3\))
δ 8.05–7.98 (m, 2H), 7.76 (d, J = 2.4 Hz, 1H), 7.59–7.54 (m, 1H), 7.52 (dd, J = 8.8 Hz, 2.4 Hz, 1H), 7.48–7.43 (m, 2H), 7.37–7.32 (m, 4H), 7.31–7.26 (m, 3H), 7.13–7.06 (m, 3H), 7.04 (d, J = 8.8 Hz, 1H), 6.92 (dd, J = 50.8 Hz, 16.4 Hz, 2H); 13C{1H} NMR (100 MHz, CDCl₃) δ 193.4, 151.7, 146.8, 145.1, 139.8, 138.7, 136.5, 133.8, 133.7, 129.8, 129.3, 129.2, 128.8, 128.5, 128.2, 127.1, 124.4, 123.1, 121.4, 119.0, 118.0, 116.5; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C₃₀H₂₁BrNO₂ 506.0750; Found 506.0745.

((Z)-8-Methyl-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3da)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.7) to afford a yellow solid in 58% yield (51 mg); mp 156–157 °C, ¹H NMR (400 MHz, CDCl₃) δ 8.04 (d, J = 7.2 Hz, 2H), 7.58–7.51 (m, 2H), 7.45 (t, J = 7.6 Hz, 2H), 7.37–7.31 (m, 3H), 7.31–7.28 (m, 4H), 7.28–7.26 (m, 1H), 7.18–7.13 (m, 2H), 7.12–7.04 (m, 2H), 6.96 (dd, J = 19.6 Hz, 16.4 Hz, 2H), 2.27 (s, 3H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 194.1, 150.9, 147.8, 145.7, 140.3, 139.1, 136.8, 135.9, 133.5, 132.6, 129.3, 128.9, 128.8, 128.7, 128.3, 127.0, 125.8, 124.1, 123.4, 123.3, 123.2, 120.1, 119.1, 15.5; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C₃₁H₂₄NO₂ 442.1802; Found 442.1811.

((Z)-7-Chloro-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3ea)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.5) to afford a yellow solid in 76% yield (70 mg); mp 156–157 °C, ¹H NMR (400 MHz, CDCl₃) δ 8.02 (d, J = 7.6 Hz, 2H), 7.65–7.49 (m, 3H), 7.45 (d, J = 7.6 Hz, 2H), 7.32–7.30 (m, 4H), 7.29–7.26 (m, 2H), 7.19–7.15 (m, 2H), 7.11–7.05 (m, 3H), 6.92 (dd, J = 22.4 Hz, 16.4 Hz, 2H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 193.6, 153.1, 146.8, 145.1, 139.6, 139.2, 136.9, 136.6, 135.6, 133.7, 132.0, 129.3, 129.2, 128.7, 128.5, 127.0, 126.6, 124.4, 124.2, 123.2, 123.0, 119.2, 118.2, 116.7; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C₃₃H₂₄ClNO₂ 462.1255; Found 462.1250.

((Z)-6-Chloro-4-((E)-4-chlorostyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3fa)
This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, R_f = 0.7) to afford a yellow solid in 78% yield (77 mg); mp 184-185 °C, _1^H NMR (400 MHz, CDCl_3) δ 8.08–7.91 (m, 2H), 7.62–7.53 (m, 2H), 7.45 (t, J = 8.0 Hz, 2H), 7.38 (d, J = 8.8 Hz, 1H), 7.32 – 7.27 (m, 4H), 7.26 – 7.22 (m, 2H), 7.15 – 7.01 (m, 4H), 6.86 (dd, J = 42.4 Hz, 16.4 Hz, 2H). _13^C{^1}H NMR (100 MHz, CDCl_3) δ 193.4, 151.2, 146.8, 145.1, 138.4, 136.5, 135.1, 134.0, 133.8, 131.0, 130.0, 129.2, 129.0, 128.8, 128.5, 128.2, 125.2, 124.4, 123.0, 120.7, 119.5, 117.7; HRMS (ESI-TOF) m/z: [M+H]^+ Calcd for C_{30}H_{20}Cl_2NO_2 469.0866; Found 469.0859.

(Z)-6,8-Di-tert-butyl-4-((E)-4-methylstyril)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3a)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, R_f = 0.6) to afford a yellow solid in 53% yield (59 mg); mp 146-147 °C, _1^H NMR (400 MHz, CDCl_3) δ 8.11–7.98 (m, 2H), 7.56–7.48 (m, 2H), 7.47–7.41 (m, 3H), 7.26–7.16 (m, 4H), 7.14–7.08 (m, 2H), 7.04–6.91 (m, 2H), 6.90–6.83 (m, 3H), 2.33 (s, 3H), 1.31 (s, 9H), 1.09 (s, 9H); _13^C{^1}H NMR (100 MHz, CDCl_3) δ 194.2, 147.6, 145.6, 141.7, 139.0, 136.9, 133.5, 133.3, 129.4, 128.6, 128.5, 127.1, 126.9, 126.4, 123.2, 121.4, 120.4, 119.4, 118.8, 34.7, 34.6, 31.4, 29.4, 21.3; HRMS (ESI-TOF) m/z: [M+H]^+ Calcd for C_{39}H_{40}NO_2 554.3051; Found 554.3052.

(Z)-6-Chloro-4-((E)-4-methoxystyril)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3a)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, R_f = 0.5) to afford a yellow solid in 82% yield (81 mg); mp 175-176 °C, _1^H NMR (400 MHz, CDCl_3) δ 8.03–7.97 (m, 2H), 7.63 (d, J = 2.4 Hz, 1H), 7.58–7.53 (m, 1H), 7.47–7.42 (m, 2H), 7.39–7.35 (m, 1H), 7.31–7.25 (m, 4H), 7.10–7.04 (m, 4H), 6.93 (d, J = 16.4 Hz, 1H), 6.86–6.81 (m, 2H), 6.71 (d, J = 16.4 Hz, 1H), 3.80 (s, 3H); _13^C{^1}H NMR (100 MHz, CDCl_3) δ 193.7, 160.5, 151.2, 147.0,
145.2, 139.3, 139.0, 136.6, 129.2, 129.1, 128.7, 128.5, 128.4, 125.3, 124.3, 123.0, 121.0, 117.6, 116.5, 114.2, 55.3; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C_{31}H_{23}ClNO_{3} 492.1361; Found 492.1353.

**((Z)-4-((E)-4-Methoxystyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3ia)**

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, R_f = 0.4) to afford a yellow solid in 65% yield (59 mg); mp 145-146 °C, ^1H NMR (400 MHz, CDCl_3) δ 8.10–7.94 (m, 2H), 7.69 (d, J = 8.0 Hz, 1H), 7.57–7.52 (m, 1H), 7.48–7.38 (m, 3H), 7.30–7.27 (m, 2H), 7.26–7.13 (m, 4H), 7.11–7.03 (m, 3H), 6.94 (d, J = 16.4 Hz, 1H), 6.86–6.76 (m, 3H), 3.80 (s, 3H); ^13C{^1H} NMR (100 MHz, CDCl_3) δ 194.2, 160.4, 152.8, 147.7, 145.6, 140.1, 138.8, 136.8, 133.5, 131.1, 129.3, 128.6, 128.4, 128.3, 125.7, 124.0, 123.8, 123.1, 119.6, 117.3, 116.3, 114.1, 55.3; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C_{31}H_{24}NO_{3} 458.1751; Found 458.1756.

**((Z)-4-((E)-4-Methylstyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3ja)**

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, R_f = 0.6) to afford a yellow solid in 68% yield (60 mg); mp 172-173 °C, ^1H NMR (400 MHz, CDCl_3) δ 8.10–7.98 (m, 2H), 7.69 (d, J = 8.0 Hz, 1H), 7.57–7.53 (m, 1H), 7.48–7.40 (m, 3H), 7.29 (t, J = 7.8 Hz, 2H), 7.26–7.15 (m, 4H), 7.15–7.10 (m, 4H), 7.07 (d, J = 7.6 Hz, 1H), 6.93 (dd, J = 34.0 Hz, 16.4 Hz, 2H), 2.34 (s, 3H); ^13C{^1H} NMR (100 MHz, CDCl_3) δ 194.1, 152.8, 147.7, 145.6, 140.0, 139.2, 136.8, 133.5, 133.1, 131.2, 129.4, 129.3, 128.7, 128.4, 127.0, 125.8, 124.1, 123.9, 123.1, 119.5, 118.6, 116.3, 21.3; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C_{31}H_{24}NO_{2} 442.1802; Found 442.1809.

**((Z)-6-Methyl-4-((E)-4-methylstyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3ka)**
This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.5) to afford a yellow solid in 65% yield (59 mg); mp 173–174 °C, ¹H NMR (400 MHz, CDCl₃) δ 8.11–7.94 (m, 2H), 7.57–7.52 (m, 1H), 7.47–7.39 (m, 3H), 7.30–7.27 (m, 2H), 7.26–7.20 (m, 3H), 7.16–7.08 (m, 4H), 7.08–7.01 (m, 2H), 6.90 (dd, J = 30.0 Hz, 16.4 Hz, 2H), 2.37 (s, 3H), 2.33 (s, 3H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 194.2, 150.8, 147.9, 145.7, 140.0, 139.1, 139.1, 136.8, 133.4, 133.4, 133.2, 131.9, 129.4, 129.3, 128.6, 128.5, 126.9, 125.7, 124.0, 123.1, 119.2, 118.8, 116.0, 21.3, 20.9; HRMS (ESI-TOF) m/z: [M+H]⁺ Calcd for C₃₂H₂₆NO₄ 456.1958; Found 456.1955.

((Z)-6-Bromo-4-((E)-4-methylstyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3la)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.6) to afford a yellow solid in 68% yield (71 mg); mp 180-181 °C, ¹H NMR (400 MHz, CDCl₃) δ 8.04–7.95 (m, 2H), 7.76 (d, J = 1.6 Hz, 1H), 7.58–7.54 (m, 1H), 7.51 (d, J = 8.8 Hz, 1H), 7.44 (d, J = 8.0 Hz, 2H), 7.31–7.27 (m, 2H), 7.26–7.20 (m, 2H), 7.13 (s, 1H), 7.11 (s, 1H), 7.10–7.01 (m, 4H), 6.87 (dd, J = 59.2, 16.4 Hz, 2H), 2.34 (s, 3H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 193.5, 151.7, 146.9, 145.2, 139.7, 139.5, 138.8, 136.5, 133.5, 133.7, 132.9, 129.5, 129.2, 128.7, 128.5, 128.2, 127.0, 124.3, 123.0, 121.4, 118.0, 117.9, 116.5, 21.3; HRMS (ESI-TOF) m/z: [M+H]⁺ Calcd for C₃₁H₂₃BrNO₂ 520.0907; Found 520.0900.

((Z)-8-Bromo-4-((E)-4-methylstyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3ma)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.6) to afford a yellow solid in 74% yield (77 mg); mp 201-202 °C, ¹H NMR (400 MHz, CDCl₃) δ 8.00 (d, J = 6.8 Hz, 2H), 7.63 (dd, J = 12.0 Hz, 7.6 Hz, 2H), 7.54 (t, J = 7.6 Hz, 1H), 7.44 (t, J = 7.6
Hz, 2H), 7.37–7.26 (m, 4H), 7.21 (d, J = 8.0 Hz, 2H), 7.15–7.02 (m, 4H), 6.88 (dd, J = 38.4 Hz, 16.4 Hz, 2H), 2.32 (s, 3H); \(^{13}\)C\(^{1}\)H NMR (100 MHz, CDCl\(_3\)) \(\delta\) 193.6, 149.6, 146.1, 144.5, 139.5, 139.4, 136.6, 134.6, 133.6, 133.0, 129.4, 129.4, 129.2, 128.7, 128.3, 128.3, 127.0, 124.9, 124.8, 124.4, 124.3, 121.3, 118.3, 110.2, 21.3; HRMS (ESI-TOF) m/z: [M+H]\(^+\) Calcd for C\(_{31}\)H\(_{23}\)BrNO\(_2\) 520.0907; Found 520.0900.

\((-\text{Z})\)-4-((\text{E})-2-Methoxystyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3na)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, \(R_f = 0.6\)) to afford a yellow solid in 75% yield (69 mg); mp 161-162 °C; \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 8.12–8.00 (m, 2H), 7.74 (d, \(J = 7.6\) Hz, 1H), 7.60–7.54 (m, 1H), 7.49–7.40 (m, 3H), 7.33–7.27 (m, 4H), 7.25–7.18 (m, 2H), 7.17–7.10 (m, 3H), 7.07 (t, \(J = 7.6\) Hz, 1H), 7.03–6.98 (m, 1H), 6.90 (d, \(J = 7.2\) Hz, 1H), 6.83 (d, \(J = 8.4\) Hz, 1H), 3.73 (s, 3H); \(^{13}\)C\(^{1}\)H NMR (100 MHz, CDCl\(_3\)) \(\delta\) 194.1, 157.5, 152.8, 147.8, 145.7, 140.7, 137.0, 134.7, 133.4, 131.1, 130.1, 129.4, 128.6, 128.4, 127.5, 126.0, 125.0, 124.0, 123.8, 123.1, 120.6, 120.3, 119.6, 116.3, 111.0, 55.4; HRMS (ESI-TOF) m/z: [M+H]\(^+\) Calcd for C\(_{31}\)H\(_{24}\)NO\(_3\) 458.1751; Found 458.1760.

\((-\text{Z})\)-4-((\text{E})-2-Fluorostyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3oa)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, \(R_f = 0.7\)) to afford a yellow solid in 84% yield (75 mg); mp 115-116 °C; \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 8.09–7.99 (m, 2H), 7.71–7.66 (m, 1H), 7.59–7.54 (m, 1H), 7.48–7.41 (m, 3H), 7.36–7.31 (m, 1H), 7.30–7.27 (m, 2H), 7.25–7.18 (m, 2H), 7.17–7.09 (m, 4H), 7.08–6.98 (m, 4H); \(^{13}\)C\(^{1}\)H NMR (100 MHz, CDCl\(_3\)) \(\delta\) 193.9, 160.6 (d, \(J_{CF} = 250.5\) Hz), 152.8, 147.5, 145.5, 139.9, 136.8, 133.6, 131.9 (d, \(J_{CF} = 2.8\) Hz), 131.2, 130.3 (d, \(J_{CF} = 8.3\) Hz), 128.9 (d, \(J_{CF} = 86.9\) Hz), 129.1, 128.4, 128.1 (d, \(J_{CF} = 3.0\) Hz), 125.8, 124.3, 124.3, 124.0 (d, \(J_{CF} = 21.3\) Hz), 123.8, 123.7, 123.1, 122.5, 122.4, 119.3, 116.4, 116.0, 115.8; HRMS (ESI-TOF) m/z: [M+H]\(^+\) Calcd for C\(_{30}\)H\(_{21}\)FNO\(_2\) 446.1551; Found 446.1545.

\((-\text{Z})\)-4-((\text{E})-3-Methylstyrlyl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3pa)
This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
Rf = 0.6) to afford a yellow solid in 79% yield (70 mg); mp 143-144 °C; ¹H NMR (400 MHz, CDCl₃) 
δ 8.14–7.94 (m, 2H), 7.69 (dd, J = 8.0 Hz, 1.6 Hz, 1H), 7.59–7.53 (m, 1H), 7.49–7.40 (m, 3H),
7.33–7.27 (m, 2H), 7.26–7.16 (m, 3H), 7.16–7.12 (m, 3H), 7.12–7.05 (m, 3H), 6.96 (dd, J = 22.0
Hz, 16.4 Hz, 2H), 2.33 (s, 3H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 194.0, 152.8, 147.6, 145.6, 139.9,
139.4, 138.4, 136.8, 135.8, 133.5, 131.2, 129.8, 129.3, 128.7, 128.7, 128.6, 128.4, 127.7, 125.8,
124.2, 124.1, 123.9, 123.1, 119.5, 119.5, 116.3, 21.3; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for
C₃₁H₂₄NO₂ 442.1802; Found 442.1798.

(Z)-4-((E)-3-Bromostyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone
(Scheme 2, compound 3qa)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
Rf = 0.7) to afford a yellow solid in 71% yield (72 mg); mp 158-159 °C; ¹H NMR (400 MHz, CDCl₃) 
δ 8.10–7.96 (m, 2H), 7.63 (dd, J = 8.0 Hz, 1.6 Hz, 1H), 7.59–7.55 (m, 1H), 7.50–7.41 (m, 4H),
7.41–7.37 (m, 1H), 7.33–7.26 (m, 2H), 7.26–7.20 (m, 2H), 7.19–7.14 (m, 2H), 7.14–7.03 (m, 3H),
6.91 (dd, J = 19.2 Hz, 16.4 Hz, 2H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 193.7, 152.8, 147.4, 145.4,
139.3, 137.9, 137.6, 136.7, 133.6, 131.8, 131.3, 130.2, 129.6, 129.2, 128.7, 128.4, 125.7, 125.6,
124.2, 123.9, 123.1, 122.9, 121.2, 119.3, 116.4; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for
C₃₀H₂₁BrNO₂ 506.0750; Found 506.0747.

(Z)-6-Bromo-4-((E)-2-(cyclohex-1-en-1-yl)vinyl)-2-(phenylimino)-2H-chromen-3-
yl)(phenyl)methanone (Scheme 2, compound 3ra)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
Rf = 0.4) to afford a yellow solid in 72% yield (73 mg); mp 180-181 °C; ¹H NMR (400 MHz, CDCl₃) 
δ 8.02–7.96 (m, 2H), 7.70 (d, J = 2.4 Hz, 1H), 7.60–7.55 (m, 1H), 7.51–7.43 (m, 3H), 7.31–7.26
(m, 1H), 7.26–7.22 (m, 1H), 7.08–7.03 (m, 3H), 7.00 (d, J = 8.8 Hz, 1H), 6.35 (dd, J = 165.6 Hz,
16.0 Hz, 2H), 5.81 (d, J = 4.4 Hz, 1H), 2.16–2.09 (m, 2H), 2.08–2.02 (m, 2H), 1.69–1.62 (m, 2H),
1.62–1.56 (m, 2H); 13C{1H} NMR (100 MHz, CDCl3) δ 193.8, 151.7, 147.0, 145.3, 143.4, 139.5, 136.7, 135.7, 133.6, 129.2, 128.8, 128.6, 128.4, 128.3, 124.2, 123.0, 121.5, 117.9, 116.3, 114.7, 26.1, 23.9, 22.0, 22.0; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C30H25BrNO2 510.1063; Found 510.1057.

(Z)-4-((E)-Oct-1-en-1-yl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3a)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.5) to afford a yellow liquid in 53% yield (46 mg); 1H NMR (400 MHz, CDCl3) δ 8.04–7.97 (m, 2H), 7.59–7.54 (m, 2H), 7.48–7.43 (m, 2H), 7.42–7.37 (m, 1H), 7.30–7.27 (m, 1H), 7.25–7.23 (m, 1H), 7.20–7.16 (m, 1H), 7.13–7.02 (m, 4H), 6.30–6.16 (m, 1H), 6.13–5.98 (m, 1H), 2.06 (d, J = 6.8 Hz, 2H), 1.27–1.20 (m, 4H), 1.19–1.12 (m, 4H), 0.85 (d, J = 6.8 Hz, 3H); 13C{1H} NMR (100 MHz, CDCl3) δ 194.0, 152.7, 147.7, 145.6, 142.7, 140.3, 136.7, 133.4, 131.0, 129.3, 128.6, 128.4, 128.2, 125.6, 123.9, 123.7, 123.0, 121.1, 119.6, 116.2, 33.5, 31.5, 28.5, 28.5, 22.5, 14.0; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C30H30NO2 436.2271; Found 436.2268.

Phenyl((Z)-2-(phenylimino)-4-((E)-2-(4'-propyl-[1,1'-biphenyl]-4-yl)vinyl)-2H-chromen-3-yl)methanone (Scheme 2, compound 3ta)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.5) to afford a yellow solid in 76% yield (83 mg); mp 181–182 °C; 1H NMR (400 MHz, CDCl3) δ 8.04 (d, J = 7.2 Hz, 2H), 7.73–7.68 (m, 1H), 7.59–7.54 (m, 2H), 7.54–7.47 (m, 4H), 7.46–7.43 (m, 2H), 7.43–7.35 (m, 2H), 7.32–7.26 (m, 3H), 7.25–7.19 (m, 2H), 7.16 (d, J = 8.0 Hz, 1H), 7.15–7.05 (m, 3H), 6.98 (dd, J = 16.4 Hz, 10.4 Hz, 2H), 2.63 (t, J = 7.6 Hz, 2H), 1.73–1.63 (m, 2H), 0.97 (t, J = 7.6 Hz, 3H); 13C{1H} NMR (100 MHz, CDCl3) δ 194.0, 152.8, 147.7, 145.6, 142.7, 140.3, 136.7, 133.4, 131.0, 129.3, 128.6, 128.4, 128.2, 125.6, 123.9, 123.7, 123.0, 121.1, 119.6, 116.2, 33.5, 31.5, 28.5, 28.5, 22.5, 14.0; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C39H32NO2 546.2271; Found 546.2268.

(4-Methoxyphenyl)((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Scheme 2, compound 3ab)
This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, R$_f$ = 0.4) to afford a yellow solid in 82% yield (75 mg); mp 123-124 °C. $^1$H NMR (400 MHz, CDCl$_3$) δ 8.06–7.93 (m, 2H), 7.68 (dd, $J$ = 8.0 Hz, 1.6 Hz, 1H), 7.45–7.39 (m, 1H), 7.36–7.30 (m, 4H), 7.29–7.26 (m, 3H), 7.22–7.17 (m, 1H), 7.16–7.13 (m, 1H), 7.13–7.09 (m, 2H), 7.08–7.04 (m, 1H), 7.03–6.88 (m, 4H), 3.85 (s, 3H); $^{13}$C{$_1^H$} NMR (100 MHz, CDCl$_3$) δ 192.5, 163.9, 152.8, 147.6, 145.7, 139.4, 139.0, 135.9, 131.7, 131.0, 129.8, 129.0, 128.9, 128.7, 128.4, 127.0, 125.7, 124.0, 123.8, 123.1, 119.8, 119.5, 116.3, 113.9, 55.5; HRMS (ESI-TOF) m/z: [M+H]$^+$ Calcd for C$_{31}$H$_{24}$NO$_3$ 458.1751; Found 458.1758.

4-((Z)-2-(Phenylimino)-4-((E)-styryl)-2H-chromene-3-carbonyl)benzonitrile (Scheme 2, compound 3ac)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, R$_f$ = 0.6) to afford a yellow solid in 92% yield (83 mg); mp 155-156 °C. $^1$H NMR (400 MHz, CDCl$_3$) δ 8.18–8.02 (m, 2H), 7.81–7.72 (m, 2H), 7.69 (dd, $J$ = 8.0, 1.6 Hz, 1H), 7.50–7.44 (m, 1H), 7.39–7.33 (m, 4H), 7.32–7.26 (m, 3H), 7.25–7.21 (m, 1H), 7.20–7.17 (m, 1H), 7.14–7.02 (m, 3H), 6.95 (s, 2H); $^{13}$C{$_1^H$} NMR (100 MHz, CDCl$_3$) δ 192.5, 152.8, 147.4, 145.0, 141.0, 139.9, 139.8, 135.5, 132.6, 131.7, 129.4, 129.3, 128.8, 128.5, 127.6, 127.0, 125.9, 124.5, 124.1, 123.1, 119.2, 119.2, 118.0, 116.5; HRMS (ESI-TOF) m/z: [M+H]$^+$ Calcd for C$_{31}$H$_{22}$N$_2$O$_2$ 453.1598; Found 453.1603.
[1,1′-Biphenyl]-4-yl((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Scheme 2, compound 3ad)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
\( R_f = 0.5 \)) to afford a yellow solid in 92% yield (93 mg); mp 136-137 °C, \(^1\)H NMR (400 MHz, CDCl\(_3\)) \( \delta \) 8.19–8.04 (m, 2H), 7.73–7.65 (m, 3H), 7.65–7.60 (m, 2H), 7.49–7.42 (m, 3H), 7.42–7.38 (m, 1H), 7.38–7.32 (m, 3H), 7.32–7.27 (m, 4H), 7.24–7.20 (m, 1H), 7.20–7.06 (m, 4H), 6.98 (dd, \( J = 28.4 \) Hz, 16.4 Hz, 2H); \(^{13}\)C\(\{^1\)H\}\) NMR (100 MHz, CDCl\(_3\)) \( \delta \) 193.5, 152.8, 147.6, 146.2, 145.6, 139.9, 139.8, 139.3, 135.9, 135.5, 131.2, 129.9, 129.0, 129.0, 128.9, 128.8, 128.7, 128.4, 128.2, 127.4, 127.3, 127.0, 125.8, 124.1, 123.9, 123.1, 119.7, 119.5, 116.4; HRMS (ESI-TOF) \( m/z \): [M+H]+ Calcd for C\(_{36}\)H\(_{26}\)NO\(_2\) 504.1958; Found 504.1956.

((Z)-2-(Phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)(4-(trifluoromethyl)phenyl)methanone (Scheme 2, compound 3ae)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
\( R_f = 0.7 \)) to afford a yellow solid in 76% yield (75 mg); mp 147-148 °C, \(^1\)H NMR (400 MHz, CDCl\(_3\)) \( \delta \) 8.12 (d, \( J = 8.0 \) Hz, 2H), 7.76–7.66 (m, 3H), 7.49–7.43 (m, 1H), 7.37–7.32 (m, 4H), 7.31–7.26 (m, 3H), 7.25–7.16 (m, 2H), 7.13–7.04 (m, 3H), 6.96 (dd, \( J = 20.8 \) Hz, 16.4 Hz, 2H); \(^{13}\)C\(\{^1\)H\}\) NMR (100 MHz, CDCl\(_3\)) \( \delta \) 192.9, 152.8, 147.4, 145.2, 140.6, 139.7, 139.5, 135.6, 134.7, 134.4, 130.4 (d, \( J_{CF} = 227.5 \) Hz), 129.4, 129.2, 128.6 (d, \( J_{CF} = 30.2 \) Hz), 128.0, 127.0, 125.8 (q, \( J_{CF} = 3.5 \) Hz), 124.9, 124.2 (d, \( J_{CF} = 32.2 \) Hz), 123.1, 119.3, 119.3, 116.5; HRMS (ESI-TOF) \( m/z \): [M+H]+ Calcd for C\(_{31}\)H\(_{21}\)FNO\(_2\) 496.1519; Found 496.1525.

Benzo[d][1,3]dioxol-5-yl((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Scheme 2, compound 3af)
This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
$R_f = 0.5$) to afford a yellow solid in 89% yield (84 mg); mp 168-169 °C, $^1$H NMR (400 MHz, CDCl$_3$) 
$\delta$ 7.68 (dd, $J = 8.0$ Hz, 1.6 Hz, 1H), 7.62 (dd, $J = 8.4$ Hz, 2.0 Hz, 1H), 7.51 (d, $J = 1.6$ Hz, 1H), 7.45–
7.40 (m, 1H), 7.39–7.35 (m, 2H), 7.35–7.30 (m, 3H), 7.30–7.27 (m, 2H), 7.22–7.17 (m, 1H), 7.17–
7.11 (m, 3H), 7.10–7.05 (m, 1H), 6.97 (dd, $J = 34.8$ Hz, 16.4 Hz, 2H), 6.82 (d, $J = 8.2$ Hz, 1H), 6.03
(s, 2H); $^{13}$C($^1$H) NMR (100 MHz, CDCl$_3$) $\delta$ 192.0, 152.8, 152.3, 148.3, 147.6, 145.6, 139.5, 139.1,
135.9, 131.6, 131.1, 129.0, 128.7, 128.4, 127.0, 126.3, 125.7, 124.0, 123.8, 123.1, 119.7, 119.4,
116.3, 108.8, 108.1, 101.9; HRMS (ESI-TOF) $m/z$: [M+H]$^+$ Calcd for C$_{31}$H$_{22}$NO$_4$ 472.1543; Found
472.1538.

(3-Fluorophenyl)((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone
(Scheme 2, compound 3ag)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
$R_f = 0.5$) to afford a yellow solid in 90% yield (80 mg); mp 101-102 °C, $^1$H NMR (400 MHz, CDCl$_3$) 
$\delta$ 7.80 (d, $J = 7.6$ Hz, 1H), 7.73–7.66 (m, 2H), 7.47–7.41 (m, 2H), 7.40–7.32 (m, 4H), 7.31–7.27
(m, 3H), 7.26–7.19 (m, 2H), 7.17 (d, $J = 8.0$ Hz, 1H), 7.13–7.05 (m, 3H), 6.96 (dd, $J = 22.4$ Hz, 16.4
Hz, 2H); $^{13}$C($^1$H) NMR (100 MHz, CDCl$_3$) $\delta$ 192.7, 162.9 (d, $J_{CF} = 246.4$ Hz), 152.8, 147.4, 145.3,
140.3, 139.5, 138.8, 135.7, 130.4 (d, $J_{CF} = 7.5$ Hz), 130.3 (d, $J_{CF} = 224.6$ Hz), 128.8, 128.5, 127.0,
125.8, 125.0 (d, $J_{CF} = 2.8$ Hz), 124.1 (d, $J_{CF} = 28.6$ Hz), 123.1, 120.7, 120.4, 119.5, 119.3, 116.4,
115.6 (d, $J_{CF} = 30.3$ Hz); HRMS (ESI-TOF) $m/z$: [M+H]$^+$ Calcd for C$_{30}$H$_{21}$FNO$_2$ 446.1551; Found
446.1554.

(3-Methoxyphenyl)((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone
(Scheme 2, compound 3ah)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
$R_f = 0.4$) to afford a yellow solid in 76% yield (70 mg); mp 99-100 °C, $^1$H NMR (400 MHz, CDCl$_3$) 
$\delta$ 7.68 (dd, $J = 8.0$ Hz, 1.6 Hz, 1H), 7.64–7.54 (m, 2H), 7.45–7.40 (m, 1H), 7.38–7.31 (m, 5H),
7.31–7.26 (m, 3H), 7.23–7.18 (m, 1H), 7.17–7.05 (m, 5H), 6.97 (dd, $J = 26.8$ Hz, 16.4 Hz, 2H),
3.83 (s, 3H); $^{13}$C($^1$H) NMR (100 MHz, CDCl$_3$) $\delta$ 193.8, 159.9, 152.8, 147.6, 145.6, 139.8, 139.2,
138.1, 135.9, 131.2, 129.7, 129.0, 128.8, 128.7, 128.4, 127.0, 125.7, 124.1, 123.9, 123.1, 122.3,
120.2, 119.7, 119.4, 116.3, 113.0, 55.4; HRMS (ESI-TOF) $m/z$: [M+H]$^+$ Calcd for C$_{31}$H$_{24}$NO$_3$
(3-Bromophenyl)((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Scheme 2, compound 3ai)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.6) to afford a yellow solid in 65% yield (66 mg); m.p. 149-150 °C, ¹H NMR (400 MHz, CDCl₃) δ 8.30–8.03 (m, 1H), 7.94 (d, J = 8.0 Hz, 1H), 7.77–7.62 (m, 2H), 7.48–7.43 (m, 1H), 7.42–7.34 (m, 3H), 7.31–7.27 (m, 2H), 7.25–7.20 (m, 1H), 7.69 (dd, J = 22.8 Hz, 16.4 Hz, 2H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 192.5, 152.8, 147.4, 145.3, 140.4, 139.6, 138.6, 136.3, 135.7, 131.9, 131.4, 130.3, 129.2, 128.5, 128.0, 127.8, 127.0, 125.8, 124.3, 124.0, 123.1, 123.0, 119.4, 119.3, 116.4; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C₃₀H₂₁BrNO₅ 506.0750; Found 506.0752.

((Z)-2-(Phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)(o-tolyl)methanone (Scheme 2, compound 3aj)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.7) to afford a yellow solid in 74% yield (65 mg); mp 180-181 °C, ¹H NMR (400 MHz, CDCl₃) δ 7.78 (dd, J = 8.0 Hz, 1.6 Hz, 1H), 7.67 (dd, J = 8.0 Hz, 1.6 Hz, 1H), 7.44–7.39 (m, 1H), 7.39–7.33 (m, 4H), 7.33–7.27 (m, 4H), 7.25–7.17 (m, 3H), 7.17–7.11 (m, 3H), 7.10–7.05 (m, 1H), 6.97 (dd, J = 31.6 Hz, 17.2 Hz, 2H), 2.63 (s, 3H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 195.7, 152.8, 147.5, 145.5, 140.4, 139.2, 139.0, 136.5, 135.9, 132.1, 132.0, 131.0, 130.6, 130.4, 129.0, 128.7, 128.4, 127.0, 125.8, 125.6, 124.1, 123.8, 123.3, 119.9, 119.6, 116.3, 21.5; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C₃₁H₂₄NO₅ 442.1802; Found 442.1796.

(2-Chlorophenyl)((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Scheme 2, compound 3ak)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20,
Rf = 0.6) to afford a yellow solid in 78% yield (72 mg); mp 174-175 °C, 1H NMR (400 MHz, CDCl3) δ 8.00–7.88 (m, 1H), 7.70 (dd, J = 8.0 Hz, 1.6 Hz, 1H), 7.48–7.42 (m, 2H), 7.41–7.38 (m, 3H), 7.37–7.32 (m, 3H), 7.32–7.27 (m, 3H), 7.22–7.18 (m, 1H), 7.15 (dd, J = 8.4 Hz, 1.2 Hz, 1H), 7.13–7.06 (m, 3H), 7.06–7.00 (m, 2H); 13C{1H} NMR (100 MHz, CDCl3) δ 192.1, 152.9, 147.3, 145.5, 140.6, 139.3, 136.6, 135.9, 133.4, 132.9, 131.7, 131.3, 131.1, 129.7, 129.0, 128.7, 128.5, 127.1, 126.8, 126.2, 124.1, 123.8, 123.2, 119.9, 119.6, 116.4; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C30H21ClNO2 462.1255; Found 462.1264.

Phenyl ((Z)-4-((E)-styryl)-2-(o-tolylimino)-2H-chromen-3-yl)methanone (Scheme 2, compound 3a)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.6) to afford a yellow solid in 60% yield (52 mg); mp 162–163 °C, 1H NMR (400 MHz, CDCl3) δ 8.12–8.00 (m, 2H), 7.70 (dd, J = 8.0 Hz, 1.6 Hz, 1H), 7.59–7.54 (m, 1H), 7.48–7.44 (m, 2H), 7.43–7.39 (m, 1H), 7.37–7.34 (m, 2H), 7.33–7.26 (m, 3H), 7.23–7.19 (m, 1H), 7.17–7.09 (m, 2H), 7.07–6.99 (m, 2H), 6.99–6.93 (m, 1H); 13C{1H} NMR (100 MHz, CDCl3) δ 194.0, 152.9, 147.1, 144.3, 139.7, 139.2, 136.8, 135.9, 133.6, 131.3, 131.2, 130.1, 129.3, 129.0, 128.7, 127.0, 125.8, 125.7, 123.9, 123.8, 121.3, 119.7, 119.4, 116.4, 18.0; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C31H24NO2 442.1802; Found 442.1799.

((Z)-2-((2-Bromophenyl)imino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3am)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.5) to afford a yellow solid in 65% yield (66 mg); mp 163–164 °C, 1H NMR (400 MHz, CDCl3) δ 8.15–8.03 (m, 2H), 7.73–7.68 (m, 1H), 7.58–7.54 (m, 1H), 7.51 (dd, J = 8.0 Hz, 1.6 Hz, 1H), 7.48–7.41 (m, 3H), 7.36–7.27 (m, 5H), 7.26–7.18 (m, 2H), 7.14–7.07 (m, 2H), 7.01 (dd, J = 29.6 Hz, 16.4 Hz, 2H), 6.93–6.87 (m, 1H); 13C{1H} NMR (100 MHz, CDCl3) δ 193.8, 152.7, 148.8, 145.2, 140.8, 139.6, 136.6, 135.8, 133.7, 132.6, 131.3, 129.7, 129.1, 128.8, 128.7, 128.7, 128.1, 127.5, 127.0, 125.8, 124.7, 124.1, 123.0, 119.5, 119.3, 116.9, 116.5; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C30H23BrNO2 506.0750; Found 506.0744.
(Z)-2-((4-Chlorophenyl)imino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3an)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.7) to afford a yellow solid in 72% yield (66 mg); mp 187-188 °C, 1H NMR (400 MHz, CDCl3) δ 8.09–7.96 (m, 2H), 7.70 (dd, J = 8.0 Hz, 1.6 Hz, 1H), 7.59–7.54 (m, 1H), 7.49–7.43 (m, 3H), 7.35–7.31 (m, 3H), 7.31–7.27 (m, 2H), 7.26–7.19 (m, 3H), 7.17 (dd, J = 8.4 Hz, 1.2 Hz, 1H), 7.10–7.04 (m, 2H), 6.96 (dd, J = 27.2 Hz, 16.4 Hz, 2H); 13C{1H} NMR (100 MHz, CDCl3) δ 193.9, 152.6, 148.1, 144.1, 140.2, 139.5, 136.7, 135.8, 133.7, 131.4, 129.3, 129.2, 129.1, 128.7, 128.5, 128.5, 127.0, 125.8, 124.6, 124.1, 119.6, 119.4, 116.3; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C30H21ClNO2 462.1255; Found 462.1260.

(Z)-2-((4-Methoxyphenyl)imino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone (Scheme 2, compound 3ao)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.4) to afford a yellow solid in 70% yield (64 mg); mp 160-161 °C, 1H NMR (400 MHz, CDCl3) δ 8.05–7.88 (m, 2H), 7.65 (dd, J = 8.4 Hz, 2.0 Hz, 1H), 7.55–7.50 (m, 1H), 7.42 (d, J = 8.0 Hz, 3H), 7.36–7.26 (m, 4H), 7.25–7.08 (m, 5H), 7.03–6.89 (m, 2H), 6.88–6.72 (m, 2H); 13C{1H} NMR (100 MHz, CDCl3) δ 194.1, 156.6, 152.9, 146.7, 139.0, 138.4, 136.9, 135.9, 133.5, 131.0, 129.3, 129.2, 128.9, 128.7, 128.7, 127.2, 127.0, 125.7, 125.2, 123.8, 119.8, 119.6, 116.3, 113.6, 55.4; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C31H24NO3 458.1751; Found 458.1759.

3-Methyl-1-((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)butan-1-one (Scheme 2, compound 3ap)
This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
Rf = 0.5) to afford a yellow solid in 60% yield (48 mg); mp 100-101 °C, 1H NMR (400 MHz, CDCl3) 
δ 7.64 (dd, J = 8.0 Hz, 1.6 Hz, 1H), 7.53–7.49 (m, 2H), 7.43–7.38 (m, 3H), 7.37–7.33 (m, 3H), 7.28–7.27 (m, 1H), 7.26–7.24 (m, 1H), 7.19–7.10 (m, 3H), 7.02 (s, 2H), 2.74 (d, J = 6.4 Hz, 2H), 2.35–2.25 (m, 1H), 0.94 (d, J = 6.4 Hz, 6H); 13C{1H} NMR (100 MHz, CDCl3) δ 203.3, 152.5, 147.0,
145.5, 139.2, 137.9, 135.8, 131.1, 131.0, 129.1, 128.9, 128.5, 127.0, 125.8, 124.2, 123.8, 123.3,
119.5, 119.4, 116.2, 52.2, 23.7, 22.7; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C28H26NO2 408.1958; Found 408.1960.

Phenyl(4-phenyl-2-(phenylamino)-4H-chromen-3-yl)methanone (Scheme 3, compound 5aa)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
Rf = 0.6) to afford a white solid in 88% yield (71 mg); mp 188–190 °C, 1H NMR (400 MHz, CDCl3) 
δ 13.36 (s, 1H), 7.53 (d, J = 7.6 Hz, 2H), 7.46–7.38 (m, 3H), 7.37–7.32 (m, 2H), 7.25–7.17 (m, 4H),
7.16–7.11 (m, 3H), 7.12–7.04 (m, 3H), 6.86–6.81 (m, 2H), 5.03 (s, 1H); 13C NMR (100MHz, CDCl3) δ 194.4, 160.0, 148.4, 146.7, 141.3, 137.2, 129.1, 129.0, 128.9, 128.4, 128.0, 127.6, 126.9, 126.4,
126.3, 125.2, 124.6, 122.4, 116.2, 89.9, 42.3; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C28H22NO2 404.1645; Found 404.1648.

(6-Methyl-2-(phenylamino)-4-(o-tolyl)-4H-chromen-3-yl)(phenyl)methanone (Scheme 3, compound 5ba)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
Rf = 0.5) to afford a white solid in 82% yield (70 mg); mp 101-102 °C, 1H NMR (400 MHz, CDCl3) 
δ 13.30 (s, 1H), 7.57 – 7.50 (m, 2H), 7.47 – 7.28 (m, 5H), 7.20 (t, J = 7.4 Hz, 1H), 7.10 – 7.01 (m, 3H), 7.02 – 6.98 (m, 3H), 6.97 – 6.92 (m, 1H), 6.93 – 6.82 (m, 2H), 5.23 (s, 1H), 2.19 (s, 3H), 1.74 (s, 3H); 13C{1H} NMR (100 MHz, CDCl3) δ 195.1, 160.1, 146.0, 145.8, 141.8, 137.4, 134.8, 134.0,
130.3, 129.2, 128.8, 128.7, 128.3, 128.3, 126.6, 126.5, 126.1, 126.0, 124.5, 122.5, 116.1, 90.4,
37.9, 20.8, 19.0; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C30H26NO2 432.1958; Found 432.1965.

(6-Chloro-4-phenyl-2-(phenylamino)-4H-chromen-3-yl)(phenyl)methanone (Scheme 3, compound 5ca)
This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.6) to afford a white solid in 75% yield (65 mg); mp 178-179 °C, 1H NMR (400 MHz, CDCl₃) δ 13.28 (s, 1H), 7.50–7.46 (m, 2H), 7.46–7.37 (m, 3H), 7.36–7.30 (m, 2H), 7.25–7.18 (m, 1H), 7.19–7.12 (m, 4H), 7.13–7.04 (m, 2H), 6.82–6.77 (m, 2H), 4.97 (s, 1H); 13C NMR (100 MHz, CDCl₃) δ 194.5, 159.7, 146.9, 146.0, 141.0, 137.0, 130.1, 129.2, 129.1, 128.1, 128.7, 128.6, 128.1, 127.7, 126.8, 126.6, 126.3, 124.8, 122.6, 117.6, 89.3, 42.3; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C₂₈H₂₁ClNO₂ 438.1255; Found 438.1248.

(6-Bromo-4-phenyl-2-(phenylamino)-4H-chromen-3-yl)(phenyl)methanone (Scheme 3, compound 5da)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.6) to afford a white solid in 70% yield (65 mg); mp 183–184 °C, 1H NMR (400 MHz, CDCl₃) δ 13.27 (s, 1H), 7.51–7.47 (m, 2H), 7.46–7.38 (m, 3H), 7.37–7.28 (m, 3H), 7.27–7.19 (m, 2H), 7.19–7.15 (m, 2H), 7.15–7.07 (m, 3H), 7.01 (d, J = 8.6 Hz, 1H), 6.84–6.76 (m, 2H), 4.97 (s, 1H); 13C NMR (100 MHz, CDCl₃) δ 194.5, 159.6, 147.5, 146.0, 141.0, 137.0, 131.6, 130.6, 129.2, 129.1, 129.0, 128.6, 128.1, 126.8, 126.6, 126.3, 124.8, 122.6, 118.0, 117.6, 89.3, 42.2; HRMS (ESI-TOF) m/z: [M+H]+ Calcd for C₂₈H₂₁BrNO₂ 482.0750; Found 482.0745.

(4-(4-Chlorophenyl)-2-(phenylamino)-4H-chromen-3-yl)(phenyl)methanone (Scheme 3, compound 5ea)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, Rf = 0.6) to afford a white solid in 70% yield (61 mg); mp 170–171 °C, 1H NMR (400 MHz, CDCl₃) δ 13.32 (s, 1H), 7.54–7.49 (m, 2H), 7.48–7.39 (m, 3H), 7.38–7.32 (m, 2H), 7.25–7.17 (m, 4H), 7.14 (d, J = 8.0 Hz, 1H), 7.11–7.04 (m, 4H), 6.77–6.69 (m, 2H), 5.02 (s, 1H); 13C NMR (100 MHz, CDCl₃) δ 194.4, 159.9, 148.3, 145.2, 141.2, 137.1, 132.0, 129.1, 129.1, 128.9, 128.5, 128.2, 127.9, 126.3, 126.3, 125.3, 124.7, 122.5, 116.4, 89.5, 41.8; HRMS (ESI-TOF) m/z: [M+H]+ Calcd
for C\textsubscript{28}H\textsubscript{21}ClNO\textsubscript{2} 438.1255; Found 438.1263.

(4-\{4-bromophenyl\}-2-\{phenylamino\}-4\textsubscript{H}-chromen-3-yl\}(phenyl)methanone (Scheme 3, compound 5fa)

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This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, \(R_f = 0.6\)) to afford a white solid in 68% yield (65 mg); mp 155-156 °C, \(^1\)H NMR (400 MHz, CDCl\textsubscript{3}) \(\delta\) 13.31 (s, 1H), 7.50 (d, \(J = 7.8\) Hz, 2H), 7.42 (q, \(J = 7.4\) Hz, 3H), 7.35 (t, \(J = 7.4\) Hz, 2H), 7.22 (d, \(J = 8.2\) Hz, 4H), 7.18 (d, \(J = 7.6\) Hz, 2H), 7.13 (d, \(J = 8.1\) Hz, 1H), 7.07 (d, \(J = 4.2\) Hz, 2H), 6.66 (d, \(J = 8.3\) Hz, 2H), 5.00 (s, 1H); \(^{13}\)C NMR (100 MHz, CDCl\textsubscript{3}) \(\delta\) 194.4, 159.9, 148.3, 145.7, 141.2, 137.0, 131.5, 129.2, 129.1, 128.9, 128.6, 128.2, 127.9, 126.3, 126.2, 125.3, 124.7, 122.5, 120.1, 116.4, 89.4, 41.8; HRMS (ESI-TOF) \(m/z\): [M+H]\(^+\) Calcd for C\textsubscript{28}H\textsubscript{21}BrNO\textsubscript{2} 482.0750; Found 482.0746.

(4-\{4-Fluorophenyl\}-2-\{phenylamino\}-4\textsubscript{H}-chromen-3-yl\}(phenyl)methanone (Scheme 3, compound 5ga)

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This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, \(R_f = 0.6\)) to afford a white solid in 62% yield (52 mg); mp 158-159 °C, \(^1\)H NMR (400 MHz, CDCl\textsubscript{3}) \(\delta\) 13.30 (s, 1H), 7.52–7.48 (m, 2H), 7.45–7.32 (m, 5H), 7.20 (m, 4H), 7.13 (d, \(J = 8.0\) Hz, 1H), 7.09–7.05 (m, 2H), 5.02 (s, 1H); \(^{13}\)C NMR (100 MHz, CDCl\textsubscript{3}) \(\delta\) 194.5, 162.5, 161.3 (d, \(J_{CF} = 243.4\) Hz), 148.3, 142.6 (d, \(J_{CF} = 3.1\) Hz), 141.3, 137.1, 129.1, 128.9, 128.4 (d, \(J_{CF} = 8.0\) Hz), 128.1, 127.8, 126.6, 126.3, 125.3, 124.7, 122.5, 116.4, 115.2 (d, \(J_{CF} = 21.3\) Hz), 89.8, 41.6; HRMS (ESI-TOF) \(m/z\): [M+H]\(^+\) Calcd for C\textsubscript{28}H\textsubscript{21}FNO\textsubscript{2} 422.1551; Found 422.1556.
(4-(3-Chlorophenyl)-2-(phenylamino)-4\textit{H}-chromen-3-yl)(phenyl)methanone (Scheme 3, compound 5\textit{a})

![Chemical structure image]

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, \(R_f = 0.6\)) to afford a white solid in 60\% yield (52 mg); mp 143-144 °C, \(^1\text{H}\) NMR (400 MHz, CDCl\(_3\)) \(\delta\) 13.29 (s, 1H), 7.50–7.47 (m, 2H), 7.47–7.37 (m, 3H), 7.36–7.30 (m, 2H), 7.25–7.20 (m, 1H), 7.18–7.13 (m, 3H), 7.13–7.09 (m, 2H), 7.04 (d, \(J = 1.1\) Hz, 2H), 6.82–6.77 (m, 2H), 4.99 (s, 1H); \(^{13}\text{C}\) NMR (100MHz, CDCl\(_3\)) \(\delta\) 194.6, 159.5, 148.6, 146.2, 141.1, 136.9, 132.8, 129.9, 129.2, 129.1, 128.5, 128.1, 126.8, 126.5, 126.3, 125.5, 125.4, 124.8, 122.6, 116.6, 89.5, 41.9; HRMS (ESI-TOF) \(m/z\): [M+H]\(^+\) Calcd for C\(_{28}\)H\(_{21}\)ClNO\(_4\) 438.1255; Found 438.1258.

(2-(Ethylamino)-4-(phenylethynyl)-4\textit{H}-chromen-3-yl)(phenyl)methanone (Scheme 3, compound 6\textit{a})

![Chemical structure image]

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, \(R_f = 0.4\)) to afford a yellow solid in 46\% yield (35 mg). mp 114-115 °C, \(^1\text{H}\) NMR (400 MHz, CDCl\(_3\)) \(\delta\) 11.35 (d, \(J = 6.0\) Hz, 1H), 7.62–7.57 (m, 2H), 7.47–7.42 (m, 3H), 7.33–7.27 (m, 2H), 7.26–7.20 (m, 5H), 7.17–7.09 (m, 2H), 4.81 (s, 1H), 3.62 (dq, \(J = 7.2\) Hz, 5.6 Hz, 2H), 1.37 (d, \(J = 7.2\) Hz, 3H); \(^{13}\text{C}\)\(^{1}\text{H}\) NMR (100 MHz, CDCl\(_3\)) \(\delta\) 191.9, 162.9, 149.0, 141.8, 131.5, 128.5, 128.7, 128.3, 128.1, 127.8, 126.5, 125.1, 123.8, 123.4, 116.3, 92.4, 86.0, 81.5, 35.8, 29.3, 15.4; HRMS (ESI-TOF) \(m/z\): [M+H]\(^+\) Calcd for C\(_{26}\)H\(_{22}\)NO\(_2\) 380.1645; Found 380.1647.

(4-((4-Methoxyphenylethynyl)-2-(phenylamino)-4\textit{H}-chromen-3-yl)(phenyl)methanone (Scheme 3, compound 6\textit{ia})

![Chemical structure image]
This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
$R_f = 0.7$) to afford a white solid in 70% yield (64 mg); mp 133-134 °C, $^1$H NMR (400 MHz, CDCl$_3$) 
$\delta$ 13.40 (s, 1H), 7.70–7.64 (m, 2H), 7.53–7.50 (m, 1H), 7.50–7.48 (m, 2H), 7.48–7.46 (m, 2H), 7.44–7.39 (m, 2H), 7.33–7.27 (m, 2H), 7.24–7.16 (m, 4H), 7.15–7.12 (m, 1H), 6.80–6.75 (m, 2H), 4.92 (s, 1H), 3.78 (s, 3H); $^{13}$C{$^1$H} NMR (100 MHz, CDCl$_3$) $\delta$ 193.2, 160.3, 159.3, 148.8, 141.2, 137.0, 132.9, 129.3, 129.1, 128.7, 128.4, 128.2, 126.6, 125.4, 124.8, 123.7, 122.6, 116.5, 115.3, 113.7, 90.4, 87.9, 81.8, 55.2, 29.3; HRMS (ESI-TOF) m/z: [M+H]$^+$ Calcd for C$_{31}$H$_{24}$NO$_3$ 458.1751; Found 458.1743.

(E)-3-Benzoyl-4-styryl-2H-chromen-2-one (Scheme 5, compound 8)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
$R_f = 0.3$) to afford a white solid in 78% yield (55 mg); mp 150–151 °C, $^1$H NMR (400 MHz, CDCl$_3$) 
$\delta$ 7.97–7.87 (m, 2H), 7.84 (dd, $J$ = 8.0 Hz, 1.6 Hz, 1H), 7.66–7.61 (m, 1H), 7.59–7.55 (m, 1H), 7.47–7.41 (m, 3H), 7.40–7.33 (m, 2H), 7.32–7.30 (m, 4H), 7.03 (dd, $J$ = 18.0 Hz, 16.4 Hz, 2H); 
$^{13}$C{$^1$H} NMR (100 MHz, CDCl$_3$) $\delta$ 192.9, 159.0, 153.4, 148.8, 141.1, 136.3, 135.3, 134.1, 132.7, 129.6, 129.3, 128.9, 128.8, 127.2, 126.3, 124.7, 124.2, 119.1, 118.6, 117.5; HRMS (ESI-TOF) m/z: [M+H]$^+$ Calcd for C$_{24}$H$_{17}$O$_3$ 353.1172; Found 353.1181.

(Z)-N,2,4-Triphenyl-1,4-dihydro-2H,5H-pyrano[3,4-c]chromen-5-imine (Scheme 5, compound 9aa)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
$R_f = 0.7$) to afford a yellow solid in 51% yield (44 mg); mp 191-192 °C, $^1$H NMR (400 MHz, CDCl$_3$) 
$\delta$ 7.51–7.48 (m, 2H), 7.48–7.45 (m, 1H), 7.42–7.37 (m, 5H), 7.37–7.31 (m, 4H), 7.31–7.27 (m, 2H), 7.26–7.25 (m, 1H), 7.21–7.17 (m, 1H), 7.13–7.10 (m, 1H), 7.08–6.99 (m, 3H), 6.21 (s, 1H), 4.77 (dd, $J$ = 10.4 Hz, 4.0 Hz, 1H), 3.05 (dd, $J$ = 17.6 Hz, 4.4 Hz, 1H), 2.96 (dd, $J$ = 17.6 Hz, 10.4 Hz, 1H); $^{13}$C{$^1$H} NMR (100 MHz, CDCl$_3$) $\delta$ 152.4, 146.7, 146.0, 141.7, 139.8, 138.1, 130.2, 128.9, 128.5, 128.4, 128.0, 127.8, 127.7, 126.0, 125.7, 123.6, 122.8, 122.8, 119.8, 115.9, 74.4, 68.2, 31.5; HRMS (ESI-TOF) m/z: [M+H]$^+$ Calcd for C$_{30}$H$_{24}$NO$_2$ 430.1802; Found 430.1809.
(Z)-N-(4-Chlorophenyl)-2,4-diphenyl-1,4-dihydro-2H,5H-pyranono[3,4-c]chromen-5-imine (Scheme 5, compound 9an)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
R_f = 0.7) to afford a yellow solid in 55% yield (51 mg); mp 179-180 °C, \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 7.50 (d, \(J = 7.8\) Hz, 3H), 7.44–7.37 (m, 6H), 7.35–7.30 (m, 3H), 7.25–7.21 (m, 3H), 7.15 (d, \(J = 8.2\) Hz, 1H), 6.97 (d, \(J = 8.2\) Hz, 2H), 6.19 (s, 1H), 4.76 (dd, \(J = 10.4\) Hz, 4.0 Hz, 1H), 3.03 (dd, \(J = 18.0\) Hz, 4.4 Hz, 1H), 2.96 (dd, \(J = 17.6\) Hz, 10.4 Hz, 1H); \(^{13}\)C\({}^1\)H NMR (100 MHz, CDCl\(_3\)) \(\delta\) 152.6, 147.6, 144.9, 142.0, 139.0, 130.7, 129.2, 129.0, 128.9, 128.8, 128.5, 128.2, 126.3, 125.9, 124.6, 124.2, 123.3, 120.1, 116.3, 74.7, 68.6, 31.9; HRMS (ESI-TOF) \(m/z\): [M+H]^+ Calcd for C\(_{30}\)H\(_{23}\)ClNO\(_2\) 464.1412; Found 464.1414.

(Z)-2-(3-Bromophenyl)-N,4-diphenyl-1,4-dihydro-2H,5H-pyranono[3,4-c]chromen-5-imine (Scheme 5, compound 9qa)

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
R_f = 0.7) to afford a yellow solid in 50% yield (51 mg); mp 192-193 °C, \(^1\)H NMR (500 MHz, CDCl\(_3\)) \(\delta\) 7.56–7.55 (m, 1H), 7.48–7.46 (m, 3H), 7.44–7.36 (m, 2H), 7.35–7.31 (m, 3H), 7.30–7.28 (m, 3H), 7.25–7.19 (m, 2H), 7.12–7.10 (m, 1H), 7.05–7.02 (m, 1H), 7.01-6.98 (m, 2H), 6.20 (s, 1H), 4.73 (dd, \(J = 10.8\) Hz, 4.0 Hz, 1H), 3.04 (dd, \(J = 17.6\) Hz, 4.0 Hz, 1H), 2.89 (dd, \(J = 18.0\) Hz, 10.8 Hz, 1H); \(^{13}\)C\({}^1\)H NMR (100 MHz, CDCl\(_3\)) \(\delta\) 152.3, 146.5, 145.9, 144.1, 139.5, 137.7, 130.8, 130.3, 130.1, 129.0, 128.8, 128.4, 128.1, 127.9, 125.6, 124.5, 123.6, 122.8, 122.6, 119.6, 116.0, 74.4, 67.6, 31.5; HRMS (ESI-TOF) \(m/z\): [M+H]^+ Calcd for C\(_{30}\)H\(_{23}\)BrNO\(_2\) 508.0907; Found 508.0914.

Phenyl[(Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl]methanone (Scheme 4, compound 3aa-d)
This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:20, 
$R_f = 0.6$) to afford a yellow solid in 65% yield (27 mg); mp 138-140 °C, 
$^1$H NMR (400 MHz, CDCl$_3$) 
$\delta$ 8.03 (d, $J = 7.0$ Hz, 2H), 7.69 (d, $J = 7.8$ Hz, 1H), 7.56 (t, $J = 7.4$ Hz, 1H), 7.46-7.41 (m, 3H), 7.31-7.28 (m, 6H), 7.20-7.14 (m, 2H), 7.11-7.04 (m, 3H), 7.01-6.90 (m, 1H); 
$^{13}$C($^1$H) NMR (100 MHz, CDCl$_3$) $\delta$ 194.1, 152.9, 147.7, 145.6, 139.9, 139.8, 139.7, 139.2, 139.1, 136.8, 135.9, 133.6, 131.3, 129.4, 129.1, 128.9, 128.8, 128.5, 127.1, 125.8, 124.2, 124.0, 123.2, 119.8, 119.5, 116.6; 
HRMS (ESI-TOF) $m/z$: [M+H]$^+$ Calcd for C$_{30}$H$_{21}$DNO$_2$ 429.1708; Found 429.1706.
14. X-ray crystallographic data of compound 3aa

The purified compound 3aa is dissolved in a mixed solvent of ethyl acetate and petroleum ether, and placed in a dark cabinet to slowly evaporate. After several days, a colourless bulk crystal was obtained. The X-ray crystal-structure determinations were obtained on a Bruker Smart CCDC APEX-2 diffractometer (graphite-monochromated Mo Kα radiation, λ=0.71073 nm) at 296(2) K.

Figure S1. ORTEP drawing of compound 3aa (30% probability for the thermal ellipsoid).

Table S2. Crystal data and structure refinement for compound 3aa.

| Property                        | Value                        |
|---------------------------------|------------------------------|
| CCDC number                     | 2184624                      |
| Identification code             | 20200616h                    |
| Empirical formula               | C30 H21 N O2                 |
| Formula weight                  | 427.48                       |
| Temperature                     | 298.15 K                     |
| Wavelength                      | 0.71073 Å                    |
| Crystal system                  | Monoclinic                   |
| Space group                     | P2_1/n                       |
| Unit cell dimensions            | a = 13.493(16) Å, α = 90°    |
|                                | b = 9.8466(12) Å, β = 102.184(4)° |
|                                | c = 17.378(2) Å, γ = 90°     |
| Volume                          | 2256.9(5) Å³                 |
| Z                               | 4                            |
| Density (calculated)            | 1.258 g/cm³                  |
| Absorption coefficient          | 0.078 mm⁻¹                   |
| F(000)                          | 896.0                        |
| Crystal size                    | 0.15 x 0.11 x 0.1 mm³        |
| Theta range for data collection | 5.962 to 49.998°             |
| Index ranges                    | -16<=h<=15, -11<=k<=11, -20<=l<=20 |
| Reflections collected           | 44868                        |
| Independent reflections         | 3960 [R(int) = 0.1377, R(sigma) = 0.0596] |
| Max. and min. transmission      | 0.746 and 0.703              |
| Data / restraints / parameters  | 3960 / 691 / 489             |
| Goodness-of-fit on F²           | 1.017                        |
| Final R indices [I>2sigma(I)]   | R1 = 0.0688, wR2 = 0.1680    |
| Final R indices (all data)      | R1 = 0.1615, wR2 = 0.2281    |
| Largest diff. peak and hole     | 0.20 and -0.22 e.Å³          |
15. X-ray crystallographic data of compound 5ga

The purified compound 5ga is dissolved in a mixed solvent of ethyl acetate and petroleum ether, and placed in a dark cabinet to slowly evaporate. After several days, a colourless bulk crystal was obtained. The X-ray crystal-structure determinations were obtained on a Bruker Smart CCDC APEX-2 diffractometer (graphite-monochromated Mo Kα radiation, λ=0.71073 nm) at 296(2) K.

Figure S2. ORTEP drawing of compound 5ga (30% probability for the thermal ellipsoid).

Table S3. Crystal data and structure refinement for compound 5ga.

| Property                              | Value                                      |
|---------------------------------------|--------------------------------------------|
| CCDC number                           | 2184624                                    |
| Identification code                   | 20210930a                                  |
| Empirical formula                     | C28 H20 F N O2                             |
| Formula weight                        | 421.45                                     |
| Temperature                           | 300.00 K                                   |
| Wavelength                            | 0.71073 Å                                  |
| Crystal system                        | Triclinic                                  |
| Space group                           | P-1                                        |
| Unit cell dimensions                  | a = 9.5747(19) Å, α= 101.581(6)°.          |
|                                      | b = 9.6132(18) Å, β= 95.065(7)°.           |
|                                      | c = 12.847(3) Å, γ= 111.403(6)°.           |
| Volume                                | 1061.5(4) Å³                              |
| Z                                      | 2                                          |
| Density (calculated)                  | 1.319 g/cm³                                |
| Absorption coefficient                | 0.089 mm⁻¹                                 |
| F(000)                                | 440.0                                      |
| Crystal size                          | 0.13 × 0.12 × 0.11 mm³                      |
| Theta range for data collection       | 5.192 to 55.336°.                          |
| Index ranges                          | -12<=h<=12, -12<=k<=12, -16<=l<=16         |
| Reflections collected                 | 16838                                      |
| Independent reflections               | 4695 [R(int) = 0.1059, R(sigma) = 0.1007]  |
| Goodness-of-fit on F²                 | 0.995                                      |
| Max. and min. transmission            | 0.746 and 0.703                            |
| Data / restraints / parameters        | 4695 / 0 / 290                             |
| Final R indices [l>2sigma(l)]         | R1 = 0.0686, wR2 = 0.1612                 |
| Final R indices (all data)            | R1 = 0.1646, wR2 = 0.2094                  |
| Largest diff. peak and hole           | 0.24 and -0.22 e. Å³                       |
16. X-ray crystallographic data of compound 9aa

The purified compound 9aa is dissolved in a mixed solvent of ethyl acetate and petroleum ether, and placed in a dark cabinet to slowly evaporate. After several days, a colourless bulk crystal was obtained. The X-ray crystal-structure determinations were obtained on a Bruker Smart CCD APEX-2 diffractometer (graphite-monochromated Mo Kα radiation, λ=0.71073 nm) at 296(2) K.

![ORTEP drawing of compound 9aa](image)

**Figure S3.** ORTEP drawing of compound 9aa (30% probability for the thermal ellipsoid).

**Table S4.** Crystal data and structure refinement for compound 9aa.

| Property                        | Value                        |
|---------------------------------|------------------------------|
| CCDC number                     | 2184629                      |
| Identification code             | mo_20201230a_0m_a            |
| Empirical formula               | C30 H23 N O2                 |
| Formula weight                  | 429.49                       |
| Temperature                     | 300.01 K                     |
| Wavelength                      | 0.71073 Å                    |
| Crystal system                  | Triclinic                    |
| Space group                     | P-1                          |
| Unit cell dimensions            | a = 4.8767(3) Å, α = 89.426(3)° |
|                                 | b = 9.4727(7) Å, β = 89.496(3)° |
|                                 | c = 24.3972(19) Å, γ = 77.542(3)° |
| Volume                          | 1100.42(14) Å³               |
| Z                               | 2                            |
| Density (calculated)            | 1.296 g/cm³                  |
| Absorption coefficient          | 0.081 mm⁻¹                   |
| F(000)                          | 452.0                        |
| Crystal size                    | 0.12 × 0.11 × 0.1 mm³        |
| Theta range for data collection | 6.644 to 55.05°.             |
| Index ranges                    | -6≤h≤6, -12≤k≤12, -31≤l≤31   |
| Reflections collected           | 48049                        |
| Independent reflections         | 5071 [R(int) = 0.0607, R(sigma) = 0.0333] |
| Max. and min. transmission      | 0.746 and 0.701              |
| Data / restraints / parameters  | 5071 / 77 / 384              |
| Goodness-of-fit on F²           | 1.016                        |
| Final R indices [I>2sigma(I)]   | R1 = 0.0673, wR2 = 0.1576    |
| R indices (all data)            | R1 = 0.1128, wR2 = 0.1837    |
| Largest diff. peak and hole     | 0.39 and -0.43 e.Å³          |
17. Computational Details

Geometry optimizations were performed by DFT calculation with the B3PW91-D3 functional, where the Stuttgart-Dresden-Bonn basis sets were used for Fe with the effective core potentials employed for representing core electrons and the 6-31G(d) basis sets were used for all other atoms. We performed single-point calculations using 6-311+G(d,p) basis sets for non-metal atoms. Solvation effects of acetonitrile were evaluated with the PCM method. All these calculations were carried out with Gaussian16 program.

![Figure S4. DFT energy profile of possible reaction mechanism from intermediate C to D](image)

| Coordination       | C     | 2.045003 | 0.640617 | -0.460577 |
|--------------------|-------|----------|----------|-----------|
| C, B3PW91-D3/BSII=-3303.730947 | C     | 1.814749 | 1.427638 | 1.865820  |
| C -2.796217        | -0.949539 | 2.927755 | N 3.372506 | 0.421403 | -0.264421 |
| C -1.716185        | -1.567923 | 2.305096 | H 3.642123 | 0.745323 | 0.684215 |
| C -0.886355        | -0.848803 | 1.442061 | C -0.711475 | 2.216671 | -0.492934 |
| C -1.125554        | 0.517505  | 1.201165 | C -1.176964 | 3.054231 | -1.389181 |
| C -2.224298        | 1.110503  | 1.833147 | O 0.160283 | -1.450248 | 0.805121 |
| C -3.058991        | 0.396536  | 2.685293 | H 0.195298 | -2.377144 | 1.079907 |
| H -3.427277        | -1.523752 | 3.600986 | S 1.301709 | 0.272077 | -2.028387 |
| H -1.514223        | -2.623969 | 2.481329 | C 2.703935 | 0.402576 | -3.182815 |
| C -0.217040        | 1.343658  | 0.358361 | H 3.263930 | -0.530793 | -3.267385 |
| H -2.394118        | 2.169703  | 1.668269 | H 2.262435 | 0.657451 | -4.150264 |
| H -3.893119        | 0.893376  | 3.171833 | H 3.374575 | 1.207278 | -2.873312 |
| C 1.257247         | 1.185775  | 0.548233 | H -1.242031 | 2.703606 | -2.422234 |

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| Atoms | x    | y    | z    |
|-------|------|------|------|
| C     | 4.114992 | -0.678978 | -0.755377 |
| C     | 5.477345 | -0.499873 | -1.010329 |
| C     | 3.532377 | -1.938554 | -0.938923 |
| C     | 6.247704 | -1.566997 | -1.463921 |
| H     | 5.916857 | 0.482684  | -0.861009 |
| C     | 4.304859 | -2.993418 | -1.416471 |
| H     | 2.480572 | -2.074545 | -0.708230 |
| C     | 5.663111 | -2.814548 | -1.680677 |
| H     | 7.306267 | -1.418561 | -1.660626 |
| H     | 3.844938 | -3.966924 | -1.567081 |
| H     | 6.262810 | -3.643386 | -2.046752 |
| C     | -1.606105 | 4.437870 | -1.134681 |
| C     | -0.734492 | 5.498328 | -1.418073 |
| C     | -2.875789 | 4.734777 | -0.614855 |
| H     | -1.111738 | 6.811785 | -1.175832 |
| H     | 0.250208 | 5.282989 | -1.827675 |
| C     | -3.250485 | 6.052794 | -0.370832 |
| H     | -3.568385 | 3.926491 | -0.409409 |
| C     | -2.369903 | 7.100868 | -0.648083 |
| H     | -0.419824 | 7.625111 | -1.400601 |
| H     | -4.238536 | 6.264483 | 0.031127 |
| H     | -2.666017 | 8.129504 | -0.459790 |
| C     | 2.101068 | 0.233319 | 2.726005 |
| C     | 1.065428 | -0.614287 | 3.127116 |
| C     | 3.407329 | -0.023336 | 3.145177 |
| C     | 1.338890 | -1.742422 | 3.922217 |
| H     | 0.048361 | -0.402445 | 2.809510 |
| C     | 3.683522 | -1.147948 | 3.917997 |
| H     | 4.204288 | 0.647926 | 2.838595 |
| C     | 2.650999 | -2.001745 | 4.305921 |
| H     | 0.526878 | -2.374937 | 4.238678 |
| H     | 4.706621 | -1.359492 | 4.217719 |
| H     | 2.867909 | -2.878433 | 4.910971 |
| O     | 2.057168 | 2.567096  | 2.341676  |
| Fe    | 1.999191 | 4.543714  | 2.016782  |
| Cl    | 3.596004 | 5.494788  | 3.097591  |
| Cl    | 0.112923 | 5.325618  | 2.689922  |
| Cl    | 2.226446 | 4.931380  | -0.085732 |

**TS$$\text{C}_{D}$, B3PW91-D3/BSII−**

| Atoms | x    | y    | z    |
|-------|------|------|------|
| C     | -1.611265 | -0.869520 | 4.048050 |
| C     | -0.700221 | -1.424319 | 3.152083 |
| C     | -0.461375 | -0.751450 | 1.963750 |
| C     | -1.059334 | 0.461626  | 1.630057 |
| C     | -1.969819 | 0.998922  | 2.547702 |

| C     | C     | H     | C     | H     | C     | H     | C     | H     |
|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| 3.596004 | 5.494788 | 3.097591 | -0.869520 | 4.048050 | -1.424319 | 3.152083 | -0.751450 | 1.963750 | 0.461626 | 1.630057 | 0.998922 | 2.547702 |

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| C | 1.643856 | 4.415087 | 0.268402 | C | 6.064549 | -1.675784 | -1.440462 |
| C | 0.212667 | 4.976465 | -2.062090 | H | 5.781322 | 0.457268 | -1.413872 |
| H | 0.187609 | 2.839165 | -2.370411 | C | 4.152346 | -2.998652 | -0.784292 |
| C | 1.355060 | 5.732860 | -0.071057 | H | 2.388750 | -1.884174 | -0.173026 |
| H | 2.233534 | 4.186257 | 1.151693 | C | 5.481256 | -2.929784 | -1.221625 |
| C | 0.640549 | 6.013513 | -1.236551 | H | 7.104147 | -1.613544 | -1.772878 |
| H | -0.333911 | 5.194129 | -2.974717 | C | 3.700430 | -3.964245 | -0.585831 |
| H | 1.701386 | 6.542928 | 0.565073 | H | 6.048414 | -3.840872 | -1.382526 |
| H | 0.419162 | 7.043404 | -1.502093 | C | -2.492019 | 4.027889 | -1.510084 |
| O | 2.937051 | 1.797097 | -0.364209 | C | -2.874791 | 4.818872 | -2.601976 |
| Fe | 3.887889 | 2.093890 | | C | -2.793789 | 4.475412 | -0.215441 |
| Cl | 3.968604 | 4.185718 | | C | -3.519901 | 6.037363 | -2.405583 |
| Cl | 1.212298 | 1.687896 | 0.076737 | H | -2.658544 | 4.476840 | -3.612137 |
| Cl | 5.861410 | 1.205873 | -1.613244 | C | -3.436623 | 5.692720 | -0.020813 |
| C-is1, B3PW91-D3/BSII=-3303.717636 | | | | C | -3.799392 | 6.481614 | -1.114002 |
| C | -1.379097 | -1.088223 | 3.182831 | C | -3.806483 | 6.640058 | -3.263676 |
| C | -0.605537 | -1.554842 | 2.133348 | C | -3.670532 | 6.019688 | 0.989421 |
| C | -0.252935 | -0.740698 | 1.017840 | H | -4.307242 | 7.429994 | -0.959981 |
| C | -0.753532 | 0.609709 | 1.078151 | C | 1.016871 | 3.247458 | 2.053818 |
| C | -1.533617 | 1.072801 | 2.156743 | C | 0.157331 | 4.223559 | 1.542047 |
| C | -1.842337 | 0.236196 | 3.210413 | C | 1.141373 | 3.089550 | 3.437625 |
| H | -1.614683 | -1.754955 | 4.010630 | C | -0.577991 | 5.028570 | 2.409150 |
| H | -0.233572 | -2.583002 | 2.128024 | H | 0.067044 | 4.360361 | 0.468634 |
| C | -0.273996 | 1.540894 | 0.046378 | C | 0.381150 | 3.871603 | 4.300278 |
| C | -1.832970 | 2.118190 | 2.170289 | C | 1.820469 | 2.334954 | 3.822144 |
| H | -2.427084 | 0.601010 | 4.050151 | C | -0.478695 | 4.844058 | 3.787617 |
| C | 1.212298 | 1.687896 | 0.076737 | H | -1.231391 | 5.794757 | 2.001084 |
| C | 1.956074 | 0.784459 | -0.644070 | H | 0.460268 | 3.724768 | 5.374133 |
| C | 1.860130 | 2.404276 | 1.165620 | H | -1.066471 | 5.460929 | 4.462782 |
| O | 3.080402 | 2.330217 | 1.397420 | O | 0.455902 | -1.209770 | 0.032496 |
| N | 3.307752 | 0.620140 | -0.577222 | Fe | -0.024627 | -3.024714 | -0.287612 |
| H | 3.660600 | 1.248402 | 0.164034 | Cl | -2.187163 | -3.047417 | -0.674534 |
| C | -1.034913 | 2.134701 | -0.867809 | Cl | 0.555350 | -4.376168 | 1.319692 |
| C | -1.799730 | 2.757110 | -1.745802 | C | 1.062478 | -3.455904 | -2.220040 |
| H | -0.163159 | 0.795453 | -2.014274 | S | 1.1212705 | -0.090184 | -2.009988 |
| S | 1.137065 | -0.090184 | -2.009988 | C | 1.572376 | 1.166926 | -3.345953 |
| H | 2.655484 | 1.258106 | -3.455756 | C | -0.202624 | 1.503961 | 2.316345 |
| H | 1.131873 | 0.803483 | -4.278095 | C | -0.143019 | -0.713347 | 1.163828 |
| H | 1.135903 | 2.135494 | -3.054816 | C | -0.582389 | 0.625220 | 1.218745 |
| C | -1.930562 | 2.303937 | -2.733943 | C | -1.119209 | 1.133906 | 2.402217 |
| C | 4.004251 | -0.595883 | -0.796429 | C | -1.197934 | 0.333797 | 3.539581 |
| C | 5.337143 | -0.518124 | -1.232470 | H | -0.780734 | -1.608803 | 4.376781 |
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| Cl | 2.231777  | -3.730503 | 1.695403 |
| TS_C3a/b, B3PW91-D3/BSII=−2865.0193129 |   |   |   |
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| C  | 5.051917  | -1.547799 | -1.931886 |
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| H  | 6.010697  | -0.184801 | -3.297491 |
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| H  | -1.785446 | 6.427303  | 2.225458 |

**D**, B3PW91-D3/BSII=−2865.0872217

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H 1.127087 -0.503590 -0.081951
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H 1.127088 1.009617 0.791699

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18. $^1$H, and $^{13}$C NMR spectra for all compounds

Phenyl(\(\text{(Z)}\)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Figure 2, compound 3aa)

$^1$H NMR Spectra of compound 3aa

$^{13}$C NMR Spectra of compound 3aa
(Z)-6-Chloro-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone
(Figure 2, compound 3ba)

1H NMR Spectra of compound 3ba

13C NMR Spectra of compound 3ba
((Z)-6-Bromo-2-(phenylimino)-4-(E-styryl)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3ca)

$^1$H NMR Spectra of compound 3ca

$^{13}$C NMR Spectra of compound 3ca
((Z)-8-Methyl-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone
(Figure 2, compound 3da)

$^1$H NMR Spectra of compound 3da

$^{13}$C NMR Spectra of compound 3da
((Z)-7-Chloro-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone
(Figure 2, compound 3ea)

$^{1}H$ NMR Spectra of compound 3ea

$^{13}C$ NMR Spectra of compound 3ea
((Z)-6-Chloro-4-((E)-4-chlorostyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3fa)

\[ \text{1H NMR Spectra of compound 3fa} \]

\[ \text{13C NMR Spectra of compound 3fa} \]
(Z)-6,8-Di-tert-butyl-4-((E)-4-methylstyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3ga)

$\begin{align*}
&\text{1H NMR Spectra of compound 3ga} \\
&\text{13C NMR Spectra of compound 3ga}
\end{align*}$
(Z)-6-Chloro-4-((E)-4-methoxystyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3ha)

$^1$H NMR Spectra of compound 3ha

$^{13}$C NMR Spectra of compound 3ha
((Z)-4-((E)-4-Methylstyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3ja)

{\textbf{1}^1\text{H} NMR Spectra of compound 3ja}

{\textbf{13}^C NMR Spectra of compound 3ja}
((Z)-6-Methyl-4-((E)-4-methylstyril)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3ka)

$^1$H NMR Spectra of compound 3ka

$^{13}$C NMR Spectra of compound 3ka
((Z)-6-Bromo-4-((E)-4-methylstyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3la)

$^1$H NMR Spectra of compound 3la

$^{13}$C NMR Spectra of compound 3la
((Z)-8-Bromo-4-((E)-4-methylstyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3ma)

**1H NMR Spectra of compound 3ma**

**13C NMR Spectra of compound 3ma**
(2Z)-4-[(E)-2-Methoxystyril]-2-(phenylimino)-2H-chromen-3-yl](phenyl)methanone
(Figure 2, compound 3na)

\[ \text{\(1^1H\) NMR Spectra of compound 3na} \]

\[ \text{\(13^C\) NMR Spectra of compound 3na} \]
((Z)-4-((E)-2-Fluorostyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone
(Figure 2, compound 3oa)
(Z)-4-((E)-3-Methylstyrlyl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3pa)

^1H NMR Spectra of compound 3pa

^13C NMR Spectra of compound 3pa
((Z)-4-((E)-3-Bromostyryl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone
(Figure 2, compound 3qa)

$^1$H NMR Spectra of compound 3qa

$^{13}$C NMR Spectra of compound 3qa
(Z)-6-Bromo-4-((E)-2-(cyclohex-1-en-1-yl)vinyl)-2-(phenylimino)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3ra)

\[ \text{\( ^{1}H \) NMR Spectra of compound 3ra} \]

\[ \text{\( ^{13}C \) NMR Spectra of compound 3ra} \]
((Z)-4-(((E)-Oct-1-en-1-yl)-2-phenylimino)-2H-chromen-3-yl)(phenyl)methanone
(Figure 2, compound 3sa)

$^1$H NMR Spectra of compound 3sa

$^{13}$C NMR Spectra of compound 3sa
Phenyl((Z)-2-(phenylimino)-4-((E)-2-(4'-propyl-[1,1'-biphenyl]-4-yl)vinyl)-2H-chromen-3-yl)methanone (Figure 2, compound 3ta)

$^1$H NMR Spectra of compound 3ta

$^{13}$C NMR Spectra of compound 3ta
(4-Methoxyphenyl)((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone
(Figure 2, compound 3ab)

$^{1}H$ NMR Spectra of compound 3ab

$^{13}C$ NMR Spectra of compound 3ab
4-((Z)-2-(Phenylimino)-4-((E)-styril)-2H-chromene-3-carbonyl)benzonitrile (Figure 2, compound 3ac)

**1H NMR Spectra of compound 3ac**

**13C NMR Spectra of compound 3ac**
[1,1'-Biphenyl]-4-yl[(Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl]methanone (Figure 2, compound 3ad)

$^1$H NMR Spectra of compound 3ad

$^{13}$C NMR Spectra of compound 3ad
((Z)-2-(Phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)(4-(trifluoromethyl)phenyl)methanone (Figure 2, compound 3ae)

$^{1}H$ NMR Spectra of compound 3ae

$^{13}C$ NMR Spectra of compound 3ae
Benzo[d][1,3]dioxol-5-yl((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Figure 2, compound 3af)

$^1$H NMR Spectra of compound 3af

$^{13}$C NMR Spectra of compound 3af
(3-Fluorophenyl)((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Figure 2, compound 3ag)

$^{1}$H NMR Spectra of compound 3ag

$^{13}$C NMR Spectra of compound 3ag
(3-Methoxyphenyl)((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Figure 2, compound 3ah)

$^1$H NMR Spectra of compound 3ah

$^{13}$C NMR Spectra of compound 3ah
(3-Bromophenyl)((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Figure 2, compound 3ai)

$^{13}C$ NMR Spectra of compound 3ai

$^1H$ NMR Spectra of compound 3ai
((Z)-2-(Phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)(o-tolyl)methanone  (Figure 2, compound 3aj)

$^1$H NMR Spectra of compound 3aj

$^{13}$C NMR Spectra of compound 3aj
(2-Chlorophenyl)((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)methanone (Figure 2, compound 3ak)
Phenyl((Z)-4-((E)-styryl)-2-(o-tolylimino)-2H-chromen-3-yl)methanone (Figure 2, compound 3a)

$^1$H NMR Spectra of compound 3a

$^{13}$C NMR Spectra of compound 3a
((Z)-2-((2-Bromophenyl)imino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone
(Figure 2, compound 3am)
((Z)-2-((4-Chlorophenyl)imino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone
(Figure 2, compound 3an)
((Z)-2-((4-Methoxyphenyl)imino)-4-((E)-styryl)-2H-chromen-3-yl)(phenyl)methanone (Figure 2, compound 3ao)

$\text{H NMR Spectra of compound 3ao}$

$\text{C NMR Spectra of compound 3ao}$
3-Methyl-1-((Z)-2-(phenylimino)-4-((E)-styryl)-2H-chromen-3-yl)butan-1-one (Figure 2, compound 3ap)

$^1$H NMR Spectra of compound 3ap

$^{13}$C NMR Spectra of compound 3ap
Phenyl(4-phenyl-2-(phenylamino)-4H-chromen-3-yl)methanone (Figure 3, compound 5aa)

$^1$H NMR Spectra of compound 5aa

$^{13}$C NMR Spectra of compound 5aa

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(6-Methyl-2-(phenylamino)-4-(o-tolyl)-4H-chromen-3-yl)(phenyl)methanone  (Figure 3, compound 5ba)

$^1$H NMR Spectra of compound 5ba

NMR Spectra of compound 5ba
(6-Chloro-4-phenyl-2-(phenylamino)-4H-chromen-3-yl)(phenyl)methanone (Figure 3, compound 5ca)

**1H NMR Spectra of compound 5ca**

**13C NMR Spectra of compound 5ca**
(6-Bromo-4-phenyl-2-(phenylamino)-4H-chromen-3-yl)(phenyl)methanone (Figure 3, compound 5da)

\[\text{1H NMR Spectra of compound 5da}\]

\[\text{13C NMR Spectra of compound 5da}\]
(4-(4-Chlorophenyl)-2-(phenylamino)-4H-chromen-3-yl)(phenyl)methanone (Figure 3, compound 5ea)

$^1$H NMR Spectra of compound 5ea

$^{13}$C NMR Spectra of compound 5ea
(4-(4-bromophenyl)-2-(phenylamino)-4H-chromen-3-yl)(phenyl)methanone (Figure 3, compound 5fa)

**1H NMR Spectra of compound 5fa**

**13C NMR Spectra of compound 5fa**
(4-(4-Fluorophenyl)-2-(phenylamino)-4H-chromen-3-yl)(phenyl)methanone (Figure 3, compound 5ga)

1H NMR Spectra of compound 5ga

13C NMR Spectra of compound 5ga
(4-(3-Chlorophenyl)-2-(phenylamino)-4\textit{H}-chromen-3-yl)(phenyl)methanone (Figure 3, compound 5ha)

\[\text{\textit{H} NMR Spectra of compound 5ha}\]

\[\text{\textit{C} NMR Spectra of compound 5ha}\]
(2-(Ethylamino)-4-(phenylethynyl)-4H-chromen-3-yl)(phenyl)methanone (Figure 3, compound 6aq)

$\text{H NMR Spectra of compound 6aq}$

$\text{C NMR Spectra of compound 6aq}$
(4-((4-Methoxyphenyl)ethynyl)-2-(phenylamino)-4'H-chromen-3-yl)(phenyl)methanone (Figure 3, compound 6ia)

\[
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\]
(E)-3-Benzyol-4-styryl-2H-chromen-2-one (Figure 5, compound 8)

$^1$H NMR Spectra of compound 8

$^{13}$C NMR Spectra of compound 8
(Z)-N,2,4-Triphenyl-1,4-dihydro-2H,5H-pyrano[3,4-c]chromen-5-imine (Figure 5, compound 9aa)

$^1$H NMR Spectra of compound 9aa

$^{13}$C NMR Spectra of compound 9aa
(Z)-N-(4-Chlorophenyl)-2,4-diphenyl-1,4-dihydro-2H,5H-pyrano[3,4-c]chromen-5-imine (Figure 5, compound 9an)

$^1$H NMR Spectra of compound 9an

$^{13}$C NMR Spectra of compound 9an
(Z)-2-(3-Bromophenyl)-N,4-diphenyl-1,4-dihydro-2H,5H-pyrano[3,4-c]chromen-5-imine (Figure 5, compound 9qa)

$^{1}H$ NMR Spectra of compound 9qa

$^{13}C$ NMR Spectra of compound 9qa
Phenyl(\(Z\))-2-(phenylimino)-4-((E)-styril)-2H-chromen-3-yl)methanone (Scheme 4, compound 3aa-d)

\[ \text{Ph} \quad \text{H} \quad \text{N} \quad \text{O} \quad \text{Ph} \quad \text{D} \]

\[ \begin{align*}
1^H \text{ NMR Spectra of compound 3aa-d} \\
13C \text{ NMR Spectra of compound 3aa-d}
\end{align*} \]
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