

SUPPLEMENTARY MATERIAL TO  
**Synthesis and mechanism of formation of hybrid structures  
comprising 2-oxochromene, thiazole and hydrazilidenechromene  
fragments**

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**7-(2-Methoxyphenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-  
b]chromene-6,8-dione (2a)**

This compound was obtained as a white solid, yield 0.35 g (74%), mp 193–  
195°C. IR: 3057, 3001, 2958, 2870, 2837 (C–H), 1725 (O–C=O), 1665 (C=O),  
1611, 1568, 1502, 1489 (C=C), 1227 (=C–O–C). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>,  $\delta$ /  
ppm): 1.03 (s, 3H, CH<sub>3</sub>), 1.15 (s, 3H, CH<sub>3</sub>), 2.26 (q,  $J$  = 16.2 Hz, 2H, CH<sub>2</sub>), 2.65–  
2.58 (m, 2H, CH<sub>2</sub>), 3.69 (s, 3H, OCH<sub>3</sub>), 5.04 (s, 1H, CH), 6.79–6.75 (m, 1H, Ar),  
6.91 (dd, 1H,  $J$  = 7.5, 1.2 Hz, Ar), 7.15 (td, 1H,  $J$  = 7.8, 1.7 Hz, Ar), 7.26 (s, 1H,  
Ar), 7.37–7.27 (m, 2H, Ar), 7.52 (dd, 1H,  $J$  = 5.0, 3.2 Hz, Ar), 7.89–7.86 (m, 1H,  
Ar). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>,  $\delta$  / ppm): 162.45, 160.62, 157.96, 154.46,  
152.63, 132.48, 131.77, 129.09, 128.46, 124.02, 122.17, 120.60, 116.80, 113.91,  
113.33, 111.15, 105.08, 55.41, 50.76, 40.94, 31.19, 31.18, 29.35, 26.92; calcd for  
C<sub>25</sub>H<sub>22</sub>O<sub>5</sub>: C, 74.61; H, 5.51, found: C, 75.21; H, 5.28.

**7-(2,4-Dimethoxyphenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-6H,8H-chromeno-  
[4,3-b]chromene-6,8-dione (2b)**

This compound was obtained as a white solid, yield 0.33 g (69%), mp 201–  
202°C. IR: 3082, 2962, 2935, 2873 (C–H), 1734 (O–C=O), 1688 (C=O), 1630,  
1611, 1599, 1584, 1557 (C=C), 1223 (=C–O–C). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>,  $\delta$ /  
ppm): 1.03 (s, 3H, CH<sub>3</sub>), 1.15 (s, 3H, CH<sub>3</sub>), 2.25 (q,  $J$  = 8 Hz,  $J$  = 8 Hz, 2H, CH<sub>2</sub>),  
2.59 (m, 2H, CH<sub>2</sub>), 3.64 (s, 3H, OCH<sub>3</sub>), 3.74 (s, 3H, OCH<sub>3</sub>), 4.95 (s, 1H, CH), 6.32

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(d, 1H,  $J = 2.4$  Hz, Ar), 6.46 (dd, 1H,  $J = 8.3, 2.3$  Hz, Ar), 7.32–7.20 (m, 2H, Ar), 7.43 (d, 1H,  $J = 8.3$  Hz, Ar), 7.52 (t, 1H,  $J = 7.8$  Hz, Ar), 7.86 (d, 1H,  $J = 7.8$  Hz, Ar).  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ,  $\delta$  / ppm): 162.40, 160.73, 160.11, 158.82, 154.35, 152.58, 137.03, 132.95, 131.71, 124.04, 122.17, 121.73, 116.77, 113.95, 113.33, 105.29, 104.40, 98.97, 55.35, 50.78, 40.93, 32.19, 30.67, 29.41, 26.87, 20.33; calcd for  $\text{C}_{26}\text{H}_{24}\text{O}_6$ : C, 72.21; H, 5.59, found: C, 72.85; H, 5.82.

*7-(4-Methoxyphenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromene-6,8-dione (2c)*

This compound was obtained as a white solid, yield 0.33 g (69%), mp 290–291°C. IR: 3074, 2958, 2906, 2908, 2873 (C–H), 1706 (O–C=O), 1646 (C=O), 1634, 1607, 1511, 1483 (C=C), 1225 (=C–O–C).  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ,  $\delta$  / ppm): 1.13 (s, 3H,  $\text{CH}_3$ ), 1.23 (s, 3H,  $\text{CH}_3$ ), 2.32 (s, 2H,  $\text{CH}_2$ ), 2.54–2.81 (m, 2H,  $\text{CH}_2$ ), 3.84 (s, 3H,  $\text{OCH}_3$ ), 4.98 (s, 1H, CH), 6.62–6.77 (m, 1H, Ar), 6.75–6.92 (m, 1H, Ar), 7.18 (d, 1H,  $J = 1.9$  Hz, Ar), 7.26 (s, 1H, Ar), 7.35 (t, 2H,  $J = 8.0$  Hz, Ar), 7.57 (t, 1H,  $J = 7.4$  Hz, Ar), 7.85 (d, 1H,  $J = 7.9$  Hz, Ar).  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ,  $\delta$  / ppm): 168.81, 160.16, 160.56, 154.01, 152.63, 150.73, 141.15, 138.77, 136.89, 132.24, 124.23, 122.34, 119.70, 116.94, 115.01, 114.00, 113.70, 106.63, 55.97, 50.73, 40.89, 33.01, 29.03, 27.70, 20.63; calcd for  $\text{C}_{25}\text{H}_{22}\text{O}_5$ : C, 74.61; H, 5.51, found: C, 74.15; H, 5.27.

*7-(3,4-Dimethoxyphenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromene-6,8-dione (2d)*

This compound was obtained as a white solid, yield 0.33 g (69%), mp 223–224°C. IR: 3098, 3008, 2954, 2925, 2866, 2833 (C–H), 1700 (O–C=O), 1648 (C=O), 1615, 1567, 1503, 1470 (C=C), 1208 (=C–O–C).  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ,  $\delta$  / ppm): 1.13 (s, 3H,  $\text{CH}_3$ ), 1.26 (s, 3H,  $\text{CH}_3$ ), 2.16 (dd,  $J = 16.2$  Hz,  $J = 8.8$  Hz, 2H,  $\text{CH}_2$ ), 2.46 (s, 2H,  $\text{CH}_2$ ), 3.74 (s, 3H,  $\text{OCH}_3$ ), 3.87 (s, 3H,  $\text{OCH}_3$ ), 5.08 (s, 1H, CH), 6.71 (d, 1H,  $J = 2.6$  Hz, Ar), 6.78 (dd, 1H,  $J = 8.3, 2.3$  Hz, Ar), 7.32–7.25 (m, 2H, Ar), 7.48 (d, 1H,  $J = 8.3$  Hz, Ar), 7.52 (t, 1H,  $J = 7.8$  Hz, Ar), 8.01 (d, 1H,  $J = 7.8$  Hz, Ar).  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ,  $\delta$  / ppm): 161.48, 161.19, 160.05, 158.52, 155.36, 153.36, 133.06, 132.92, 125.24, 122.84, 120.68, 117.85, 115.82, 113.67, 110.25, 108.82, 106.07, 101.93, 56.58, 53.05, 43.54, 32.92, 31.08, 28.24, 26.43, 24.93; calcd for  $\text{C}_{26}\text{H}_{24}\text{O}_6$ : C, 72.21; H, 5.59, found: C, 72.48; H, 5.43.

*7-(3-Methoxy-4-hydroxyphenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromene-6,8-dione (2e)*

This compound was obtained as a white solid, yield 0.32 g (69%), mp 246–248°C. IR: 3365 (O–H), 3170, 3049, 2958, 2873, 2835 (C–H), 1688 (O–C=O), 1648 (C=O), 1624, 1513, 1489, 1473, 1434 (C=C), 1244 (=C–O–C).  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ,  $\delta$  / ppm): 1.10 (s, 3H,  $\text{CH}_3$ ), 1.16 (s, 3H,  $\text{CH}_3$ ), 2.30 (dd, 2H,  $J = 4.7$ ,

1.9 Hz, CH<sub>2</sub>), 2.64–2.70 (m, 2H, CH<sub>2</sub>), 3.91 (s, 3H, OCH<sub>3</sub>), 4.88 (s, 1H, CH), 5.50 (s, 1H, OH), 6.62 (dt, 1H, *J* = 8.3, 2.0 Hz, Ar), 6.74 (dd, 1H, *J* = 8.1, 1.9 Hz, Ar), 7.12 (d, 1H, *J* = 2.0 Hz, Ar), 7.26 (d, 1H, *J* = 1.9 Hz, Ar), 7.32–7.34 (m, 2H, Ar), 7.55 (d, 1H, *J* = 7.9 Hz, Ar), 7.85 (dd, 1H, *J* = 7.9, 1.9 Hz, Ar). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>,  $\delta$ /ppm): 161.81, 160.76, 153.67, 152.55, 146.07, 144.61, 134.79, 132.11, 124.22, 122.37, 120.24, 116.89, 115.25, 114.09, 113.78, 112.51, 106.98, 95.34, 55.97, 50.74, 40.83, 32.85, 32.32, 29.16, 27.52; calcd for C<sub>25</sub>H<sub>22</sub>O<sub>6</sub>: C, 72.76; H, 5.30; found: C, 72.54; H, 5.94.

*2-(7-(2-Methoxyphenyl)-10,10-dimethyl-6-oxo-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromen-8-ylidene)hydrazine -1-carbothioamide (3a)*

This compound was obtained as a yellow solid, yield 0.13 g (45%) (*method 1*) or 0.21 g (72%) (*method 2*), mp 274–276°C. IR; 3458, 3346 (N–H, NH<sub>2</sub>), 3147 (N–H), 2957, 2921, 2837 (C–H), 1736 (O–C=O), 1675 (C=N), 1629, 1609, 1559, 1495 (C=C), 1096 (C=S). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>,  $\delta$ /ppm): 1.04 (s, 3H, CH<sub>3</sub>), 1.18 (s, 3H, CH<sub>3</sub>), 2.08–2.27 (m, 2H, CH<sub>2</sub>), 2.54 (s, 2H, CH<sub>2</sub>), 3.97 (s, 3H, OCH<sub>3</sub>), 5.49 (s, 1H, CH), 6.23 (s, 1H, NH<sub>2</sub>), 8.25 (s, 1H, NH), 8.50 (s, 1H, NH<sub>2</sub>), 6.23 (s, 1H, Ar), 6.88 (dd, 2H, *J* = 15.7, 7.8 Hz, Ar), 7.02–7.15 (m, 2H, Ar), 7.21 – 7.27 (m, 2H, Ar), 7.44–7.57 (m, 1H, Ar), 7.85 (dt, 1H, *J* = 8.0 Hz, Ar). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>,  $\delta$ /ppm): 178.71, 161.97, 160.81, 156.72, 154.02, 152.46, 145.18, 139.32, 131.89, 129.37, 128.28, 124.08, 122.36, 121.71, 116.76, 113.95, 113.27, 111.83, 105.96, 57.06, 40.13, 36.77, 31.07, 29.66, 27.76, 27.36; calcd for C<sub>26</sub>H<sub>25</sub>O<sub>4</sub>N<sub>3</sub>S: C, 65.67; H, 5.30; S, 6.74; N, 8.84; found: C, 66.31; H, 5.84; S, 6.57; N, 8.13.

*2-(7-(2,4-Dimethoxyphenyl)-10,10-dimethyl-6-oxo-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromen-8-ylidene)hydrazine -1-carbothioamide (3b)*

This compound was obtained as a light yellow solid, yield 0.15 g (51%) (*method 1*) or 0.24 g (83%) (*method 2*), mp 233–235°C. IR; 3502, 3439 (N–H, NH<sub>2</sub>), 3155 (N–H), 3068, 2968, 2929, 2831 (C–H), 1721 (O–C=O), 1677 (C=N), 1630, 1611, 1582, 1499 (C=C), 1094 (C=S). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>,  $\delta$ /ppm): 1.03 (s, 3H, CH<sub>3</sub>), 1.17 (s, 3H, CH<sub>3</sub>), 2.13–2.28 (m, 2H, CH<sub>2</sub>), 2.47–2.69 (m, 2H, CH<sub>2</sub>), 3.73 (s, 3H, OCH<sub>3</sub>), 3.93 (s, 3H, OCH<sub>3</sub>), 5.37 (s, 1H, CH), 6.24 (s, 1H, NH<sub>2</sub>), 8.16 (s, 1H, NH), 8.52 (s, 1H, NH<sub>2</sub>), 6.30–6.45 (m, 3H, Ar), 6.98 (d, 1H, *J* = 8.4 Hz, Ar), 7.19–7.30 (m, 2H, Ar), 7.52 (ddd, 2H, *J* = 8.9, 7.5, 1.7 Hz, Ar), 7.85 (ddd, 1H, *J* = 7.9, 5.9, 1.6 Hz, Ar). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>,  $\delta$ /ppm): 178.84, 162.39, 160.07, 159.72, 157.64, 153.85, 152.49, 152.43, 146.30, 131.78, 129.94, 125.10, 124.04, 122.31, 116.74, 114.01, 110.00, 106.31, 100.12, 56.90, 55.26, 40.90, 40.13, 32.22, 31.06, 29.68, 27.32; calcd for C<sub>27</sub>H<sub>27</sub>O<sub>5</sub>N<sub>3</sub>S: C, 64.14; H, 5.38; S, 6.34; N, 8.31; found: C, 63.75; H, 5.85; S, 6.48; N, 8.65.

*2-(7-(4-Methoxyphenyl)-10,10-dimethyl-6-oxo-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromen-8-ylidene)hydrazine-1-carbothioamide (3c)*

This compound was obtained as a light yellow solid, yield 0.16 g (54%) (*method 1*) or 0.20 g (69%) (*method 2*), mp 248–249°C. IR; 3506, 3485 (N–H, NH<sub>2</sub>), 3074 (N–H), 3003, 2957, 2904 (C–H), 1719 (O–C=O), 1675 (C=N), 1619, 1607, 1565, 1509 (C=C), 1092 (C=S). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>,  $\delta$ /ppm): 1.05 (s, 3H, CH<sub>3</sub>), 1.15 (s, 3H, CH<sub>3</sub>), 2.18 (dd,  $J$  = 16.4 Hz,  $J$  = 32 Hz, 2H, CH<sub>2</sub>), 2.51 (s, 2H, CH<sub>2</sub>), 3.95 (s, 3H, OCH<sub>3</sub>), 5.42 (s, 1H, CH), 6.21 (s, 1H, NH<sub>2</sub>), 8.19 (s, 1H, NH), 8.50 (s, 1H, NH<sub>2</sub>), 6.92 (s, 1H, Ar), 6.88 (dd, 2H,  $J$  = 15.7, 7.8 Hz, Ar), 7.02–7.15 (m, 2H, Ar), 7.21–7.27 (m, 2H, Ar), 7.44–7.57 (m, 1H, Ar), 7.88 (dd, 1H,  $J$  = 8.0, 1.6 Hz, Ar), 6.92–8.03 (m, 8H, Ar). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>,  $\delta$ /ppm): 178.85, 165.61, 164.52, 158.41, 152.52, 152.28, 145.71, 135.95, 132.80, 127.61, 126.91, 124.85, 124.36, 116.93, 116.61, 114.01, 105.80, 104.18, 97.11, 55.26, 46.11, 45.52, 39.79, 35.49, 30.07, 29.43; calcd for C<sub>26</sub>H<sub>25</sub>O<sub>4</sub>N<sub>3</sub>S: C, 65.67; H, 5.30; S, 6.74; N, 8.84, found: C, 66.31; H, 5.84; S, 6.57; N, 8.13.

*2-(7-(3,4-Dimethoxyphenyl)-10,10-dimethyl-6-oxo-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromen-8-ylidene)hydrazine-1-carbothioamide (3d)*

This compound was obtained as a light yellow solid, yield 0.14 g (48%) (*method 1*) or 0.21 g (75%) (*method 2*), mp 283–285°C. IR; 3504, 3452 (N–H, NH<sub>2</sub>), 3145 (N–H), 3090, 2975, 2931, 2850 (C–H), 1724 (O–C=O), 1675 (C=N), 1638, 1615, 1580, 1503 (C=C), 1093 (C=S). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>,  $\delta$ /ppm): 1.07 (s, 3H, CH<sub>3</sub>), 1.21 (s, 3H, CH<sub>3</sub>), 2.14 (dd,  $J$  = 16.2 Hz,  $J$  = 34 Hz, 2H, CH<sub>2</sub>), 2.48 (s, 2H, CH<sub>2</sub>), 3.74 (s, 3H, OCH<sub>3</sub>), 3.87 (s, 3H, OCH<sub>3</sub>), 5.42 (s, 1H, CH), 6.24 (s, 1H, NH<sub>2</sub>), 8.19 (s, 1H, NH), 8.50 (s, 1H, NH<sub>2</sub>), 6.38–6.45 (m, 3H, Ar), 6.68–6.91 (m, 1H, Ar), 7.08–7.20 (m, 2H, Ar), 7.36 (ddd, 2H,  $J$  = 8.9, 7.5, 1.7 Hz, Ar), 7.98 (dd, 1H,  $J$  = 7.9, 1.6 Hz, Ar). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>,  $\delta$ /ppm): 178.31, 163.39, 160.75, 156.62, 153.65, 146.67, 144.37, 138.30, 136.18, 135.08, 129.28, 126.92, 122.31, 120.81, 120.05, 118.13, 116.35, 113.64, 102.24, 67.32, 58.69, 41.62, 40.36, 36.18, 31.44, 29.16, 27.39; calcd for C<sub>27</sub>H<sub>27</sub>O<sub>5</sub>N<sub>3</sub>S: C, 64.14; H, 5.38; S, 6.34; N, 8.31, found: C, 63.43; H, 5.82; S, 6.48; N, 8.15.

*2-(7-(3-Methoxy-4-hydroxyphenyl)-10,10-dimethyl-6-oxo-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromen-8-ylidene)hydrazine-1-carbothioamide (3e)*

This compound was obtained as a yellow solid, yield 0.13 g (44%) (*method 1*) or 0.23 g (81%) (*method 2*), mp 263–264°C. IR; 3487, 3347 (N–H, NH<sub>2</sub>), 3148 (N–H), 2963, 2923, 2878 (C–H), 1719 (O–C=O), 1675 (C=N), 1628, 1611, 1563, 1511 (C=C), 1094 (C=S). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>,  $\delta$ /ppm): 1.12 (s, 3H, CH<sub>3</sub>), 1.21 (s, 3H, CH<sub>3</sub>), 2.10–2.353 (m, 2H, CH<sub>2</sub>), 2.59 (s, 2H, CH<sub>2</sub>), 3.83 (s, 3H, OCH<sub>3</sub>), 4.93 (s, 1H, CH), 5.49 (s, 1H, OH), 6.13 (s, 1H, NH<sub>2</sub>), 6.92 (s, 1H, NH), 8.55 (s, 1H, NH<sub>2</sub>), 6.75–6.84 (m, 2H, Ar), 6.95–7.03 (m, 1H, Ar), 7.29–7.36 (m,

2H, Ar), 7.55 (ddd, 1H,  $J = 8.4, 7.4, 1.6$  Hz, Ar), 7.84 (dd, 1H,  $J = 7.8, 1.6$  Hz, Ar).  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ,  $\delta$ /ppm): 175.86, 165.39, 161.70, 158.39, 152.66, 146.58, 140.33, 136.15, 135.38, 131.91, 126.68, 124.23, 122.39, 120.73, 116.84, 114.39, 111.01, 102.49, 100.20, 66.76, 55.96, 40.10, 36.85, 31.26, 29.79, 27.52; calcd for  $\text{C}_{26}\text{H}_{25}\text{O}_5\text{N}_3\text{S}$ : C, 63.53; H, 5.13; S, 6.52; N, 8.55, found: C, 63.12; H, 5.85; S, 6.58; N, 8.85.

*7-(2-Methoxyphenyl)-10,10-dimethyl-8-(2-(4-(2-oxo-2H-chromen-3-yl)thiazol-2-yl)hydrazilidene)-8,9,10, 11-tetrahydro-6H,7H-chromeno[4,3-b]chromene-6-one (5a)*

This compound was obtained as a yellow solid, yield 0.26 g (97%), mp 275–277°C. IR: 3354 (N–H), 3269, 3182, 3068, 2966, 2963, 2914, 2831 (C–H), 1729 (O–C=O), 1675 (C=N), 1624, 1609, 1576, 1513 (C=C).  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ,  $\delta$ /ppm): 1.05 (s, 3H,  $\text{CH}_3$ ), 1.25 (s, 3H,  $\text{CH}_3$ ), 2.11–2.48 (m, 2H,  $\text{CH}_2$ ), 2.54 (d, 2H,  $J = 16.6$  Hz,  $\text{CH}_2$ ), 3.67 (s, 3H,  $\text{OCH}_3$ ), 5.17 (s, 1H, CH), 6.95 (t, 1H,  $J = 8.0$  Hz, Ar), 6.77 (d, 1H,  $J = 8.3$  Hz, Ar), 7.18 (t, 1H,  $J = 8.0$  Hz, Ar), 7.34 (dt, 4H,  $J = 14.6, 8.1$  Hz, Ar), 7.63 (ddd, 4H,  $J = 44.2, 19.1, 8.4$  Hz, Ar), 7.91 (s, 1H, H-5<sub>(thiazol)</sub>), 8.51 (s, 1H, H-4<sub>(chromen)</sub>), 10.92 (s, 1H, NH).  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ,  $\delta$ /ppm): 168.40, 160.59, 160.44, 144.77, 139.05, 138.47, 137.51, 135.51, 135.34, 133.68, 131.70, 131.67, 129.03, 128.54, 128.29, 124.02, 122.21, 121.76, 120.70, 120.05, 118.8, 117.81, 117.48, 116.78, 116.40, 110.92, 110.39, 109.91, 103.78, 55.10, 49.15, 40.60, 40.33, 32.63, 29.89, 27.50, 26.92; calcd for  $\text{C}_{37}\text{H}_{29}\text{O}_6\text{N}_3\text{S}$ : C, 69.04; H, 4.54; S, 4.98; N, 6.53, found: C, 68.59; H, 4.92; S, 5.16; N, 6.78.

*7-(2,4-Dimethoxyphenyl)-10,10-dimethyl-8-(2-(4-(2-oxo-2H-chromen-3-yl)thiazol-2-yl)hydrazilidene)-8,9,10, 11-tetrahydro-6H,7H-chromeno[4,3-b]chromene-6-one (5b)*

This compound was obtained as a yellow solid, yield 0.21 g (79%), mp 244–245°C. IR: 3490 (N–H), 3419, 3358, 3294, 3267, 3176, 3062, 2963, 2931 (C–H), 1727 (O–C=O), 1675 (C=N), 1626, 1607, 1505, 1491 (C=C).  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ,  $\delta$ /ppm): 1.03 (s, 3H,  $\text{CH}_3$ ), 1.24 (s, 3H,  $\text{CH}_3$ ), 2.14–2.64 (m, 4H,  $\text{CH}_2$ ), 3.62 (s, 3H,  $\text{OCH}_3$ ), 3.75 (s, 3H,  $\text{OCH}_3$ ), 5.11 (s, 1H, CH), 6.32 (d, 1H,  $J = 2.4$  Hz, Ar), 6.48 (dt, 1H,  $J = 8.4, 1.8$  Hz, Ar), 7.27–7.39 (m, 4H, Ar), 7.43–7.63 (m, 4H, Ar), 7.69 (dd, 1H,  $J = 7.9, 1.6$  Hz, Ar), 7.86 (dd, 1H,  $J = 8.0, 1.6$  Hz, Ar), 7.92 (s, 1H, H-5<sub>(thiazol)</sub>), 8.50 (s, 1H, H-4<sub>(chromen)</sub>), 10.58 (s, 1H, NH).  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ,  $\delta$ /ppm): 168.38, 160.13, 158.84, 152.94, 139.62, 139.08, 137.13, 135.40, 135.38, 134.04, 132.44, 131.71, 130.67, 130.14, 129.07, 125.99, 124.97, 124.01, 122.17, 120.99, 118.80, 116.76, 116.39, 114.15, 113.36, 110.31, 109.46, 103.67, 98.81, 56.78, 55.25, 50.02, 40.30, 38.48, 32.02, 31.17, 29.94, 26.79; calcd for  $\text{C}_{38}\text{H}_{31}\text{O}_7\text{N}_3\text{S}$ : C, 67.74; H, 4.64; S, 4.76; N, 6.24, found: C, 68.27; H, 4.82; S, 4.58; N, 6.51.

*7-(4-Methoxyphenyl)-10,10-dimethyl-8-(2-(4-(2-oxo-2H-chromen-3-yl)thiazol-2-yl)hydrazilidene)-8,9,10,11-tetrahydro-6H,7H-chromeno[4,3-b]chromene-6-one (5c)*

This compound was obtained as a yellow solid, yield 0.24 g (85%), mp 281–283°C. IR; 3499 (N–H), 3425, 3327, 2954, 3076, 3059 (C–H), 1733, 1717 (O–C=O), 1667 (C=N), 1624, 1611, 1588, 1513 (C=C). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>,  $\delta$ / ppm): 1.01 (s, 3H, CH<sub>3</sub>), 1.28 (s, 3H, CH<sub>3</sub>), 2.37 (d,  $J$  = 16.2 Hz, 1H, CH<sub>2</sub>), 2.49 (m, 3H, CH<sub>2</sub>), 3.72 (s, 3H, OCH<sub>3</sub>), 5.23 (s, 1H, CH), 6.95 (t, 1H,  $J$  = 8.0 Hz, Ar), 6.77 (d, 1H,  $J$  = 8.3 Hz, Ar), 7.18 (t, 1H,  $J$  = 8.0 Hz, Ar), 7.34 (dd, 4H,  $J$  = 44.8, 20.4, 8.2, Ar), 7.63 (dd, 4H,  $J$  = 44.2, 19.1, 8.4 Hz, Ar), 6.83–7.95 (m, 12H, Ar), 7.93 (s, 1H, H-5<sub>(thiazol)</sub>), 8.50 (s, 1H, H-4<sub>(chromen)</sub>), 10.93 (s, 1H, NH). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>,  $\delta$ / ppm): 168.87, 160.85, 160.07, 159.72, 157.63, 153.85, 152.49, 152.43, 145.30, 134.72, 132.96, 131.78, 129.94, 125.53, 125.09, 124.84, 124.01, 122.31, 122.15, 116.70, 114.01, 111.81, 111.46, 110.03, 106.81, 106.01, 104.25, 100.12, 98.94, 56.60, 55.30, 40.90, 40.13, 32.22, 31.06, 29.67, 27.32; calcd for C<sub>37</sub>H<sub>29</sub>O<sub>6</sub>N<sub>3</sub>S: C, 69.04; H, 4.54; S, 4.98; N, 6.53, found: C, 69.45; H, 4.85; S, 4.58; N, 6.45.

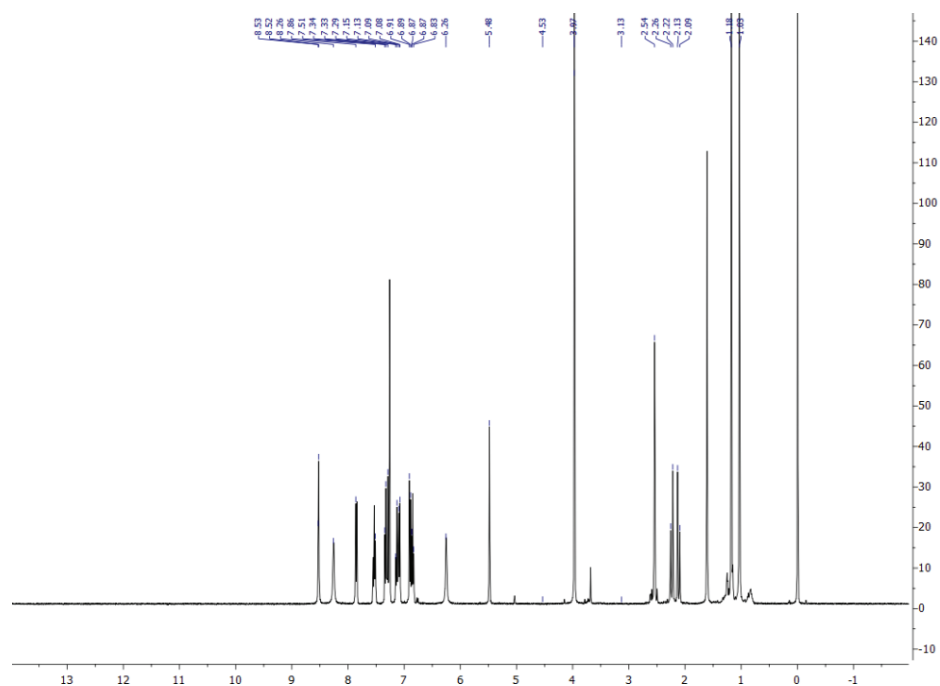
*7-(3,4-Dimethoxyphenyl)-10,10-dimethyl-8-(2-(4-(2-oxo-2H-chromen-3-yl)thiazol-2-yl)hydrazilidene)-8,9,10,11-tetrahydro-6H,7H-chromeno[4,3-b]chromene-6-one (5d)*

This compound was obtained as a yellow solid, yield 0.21 g (75%), mp 244–245°C. IR; 3489 (N–H), 3421, 3360, 3280, 3176, 3058, 2985, 2896 (C–H), 1723 (O–C=O), 1677 (C=N), 1625, 1615, 1599, 1511 (C=C). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>,  $\delta$ / ppm): 1.02 (s, 3H, CH<sub>3</sub>), 1.23 (s, 3H, CH<sub>3</sub>), 2.25–2.61 (m, 4H, CH<sub>2</sub>), 3.61 (s, 3H, OCH<sub>3</sub>), 3.73 (s, 3H, OCH<sub>3</sub>), 5.09 (s, 1H, CH), 6.31 (d, 1H,  $J$  = 2.2 Hz, Ar), 6.46–6.53 (m, 1H, Ar), 7.24–7.37 (m, 4H, Ar), 7.50–7.58 (m, 3H, Ar), 7.67 (dd,  $J$  = 7.8, 1.9 Hz, 1H), 7.84 (dd,  $J$  = 8.0, 2.0 Hz, 1H), 7.90 (s, 1H, H-5<sub>(thiazol)</sub>), 8.49 (s, 1H, H-4<sub>(chromen)</sub>), 10.61 (s, 1H, NH). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>,  $\delta$ / ppm): 168.36, 161.16, 160.12, 154.94, 158.83, 154.60, 153.28, 152.92, 152.52, 139.35, 138.95, 137.83, 134.03, 133.94, 132.39, 131.58, 129.05, 124.94, 123.98, 122.15, 120.99, 118.79, 116.73, 116.35, 114.14, 110.33, 110.00, 109.45, 103.63, 98.80, 55.23, 55.06, 52.68, 40.28, 38.47, 32.00, 31.14, 29.94, 26.82; calcd for C<sub>38</sub>H<sub>31</sub>O<sub>7</sub>N<sub>3</sub>S: C, 67.74; H, 4.64; S, 4.76; N, 6.24, found: C, 67.25; H, 5.12; S, 5.13; N, 6.10.

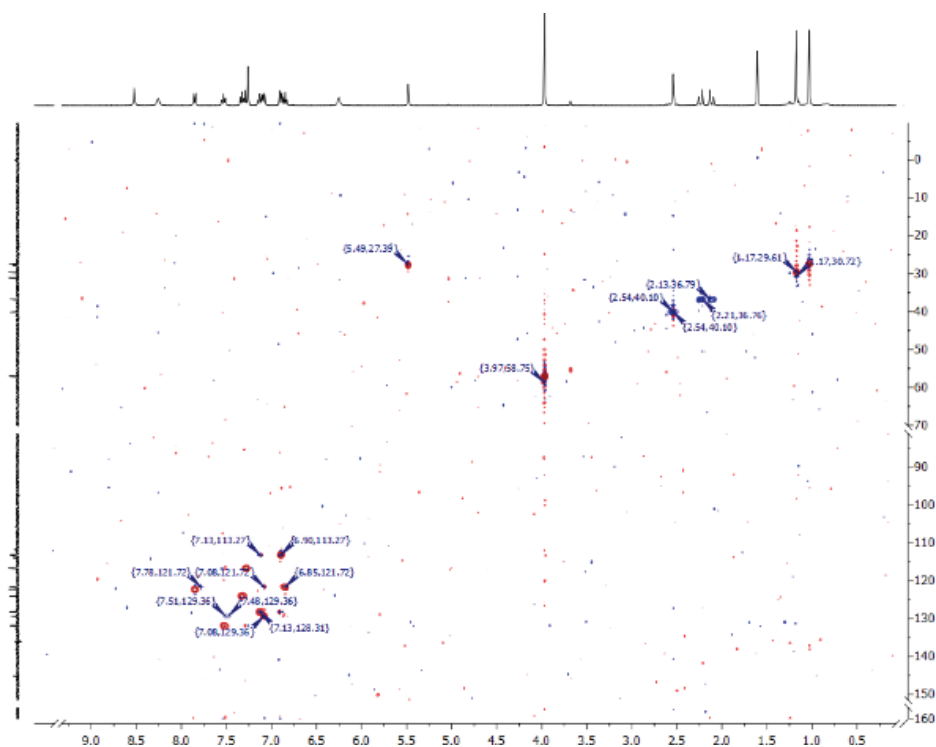
*7-(3-Methoxy-4-hydroxyphenyl)-10,10-dimethyl-8-(2-(4-(2-oxo-2H-chromen-3-yl)thiazol-2-yl)hydrazilidene)-8,9,10,11-tetrahydro-6H,7H-chromeno[4,3-b]chromene-6-one (5e)*

This compound was obtained as an orange solid, yield 0.20 g (71%), mp 285–286°C. IR; 3506 (O–H), 3419 (N–H), 3347, 3167, 2954, 2933, 2873 (C–H), 1704

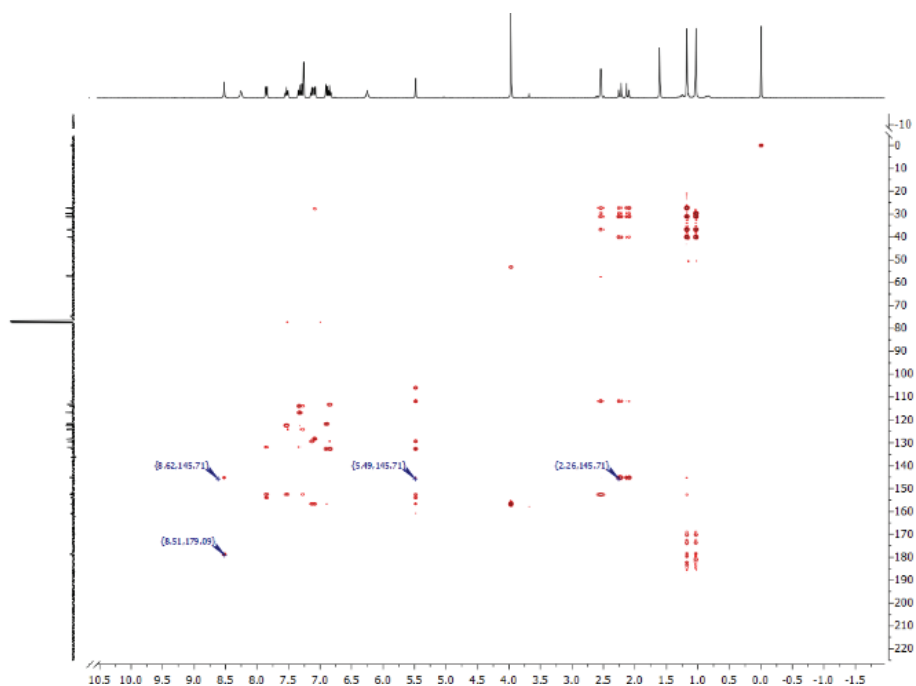
(O=C=O), 1675 (C=N), 1624, 1609, 1576, 1513 (C=C).  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ,  $\delta$ / ppm): 1.01 (s, 3H,  $\text{CH}_3$ ), 1.21 (s, 3H,  $\text{CH}_3$ ), 2.33 (d,  $J = 16.4$  Hz, 1H,  $\text{CH}_2$ ), 2.38 (m, 3H,  $\text{CH}_2$ ), 3.62 (s, 3H,  $\text{OCH}_3$ ), 5.23 (s, 1H, CH), 6.03 (s, 1H, OH), 6.35 (d, 1H,  $J = 2.4$  Hz, Ar), 6.51–6.54 (m, 1H, Ar), 7.24–7.40 (m, 4H, Ar), 7.51–7.56 (m, 3H, Ar), 7.68 (dd,  $J = 7.9, 2.1$  Hz, 1H), 7.91 (dd,  $J = 8.0, 2.0$  Hz, 1H), 7.89 (s, 1H, H-5<sub>(tiazol)</sub>), 8.56 (s, 1H, H-4<sub>(chromen)</sub>), 10.74 (s, 1H, NH).  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ,  $\delta$ / ppm): 168.83, 161.06, 160.37, 159.72, 157.83, 156.85, 155.99, 155.43, 148.32, 135.74, 133.06, 131.75, 130.04, 126.53, 125.39, 124.95, 124.22, 122.30, 121.95, 116.50, 114.67, 112.31, 110.86, 110.03, 108.01, 106.21, 104.55, 102.12, 100.94, 57.30, 55.35, 41.93, 40.03, 32.42, 31.96, 30.32, 28.67; calcd for  $\text{C}_{37}\text{H}_{29}\text{O}_7\text{N}_3\text{S}$ : C, 67.36; H, 4.43; S, 4.86; N, 6.37, found: C, 67.82; H, 4.86; S, 5.14; N, 6.48.



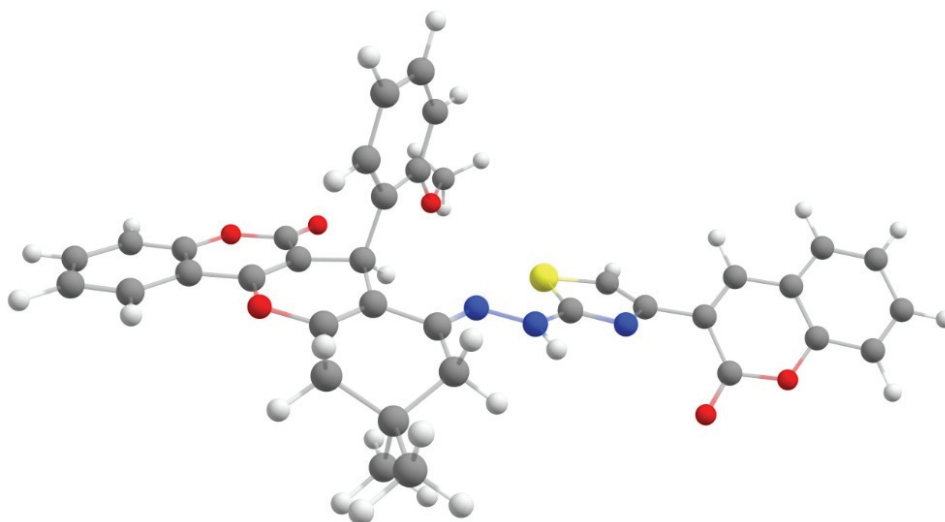
**Figure S-1.**  $^1\text{H}$  NMR spectrum of 2-(7-(2-methoxyphenyl)-10,10-dimethyl-6-oxo-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromene -8-ylidene)hydrazine-1-carbothioamide (**3a**),  $\text{CDCl}_3$



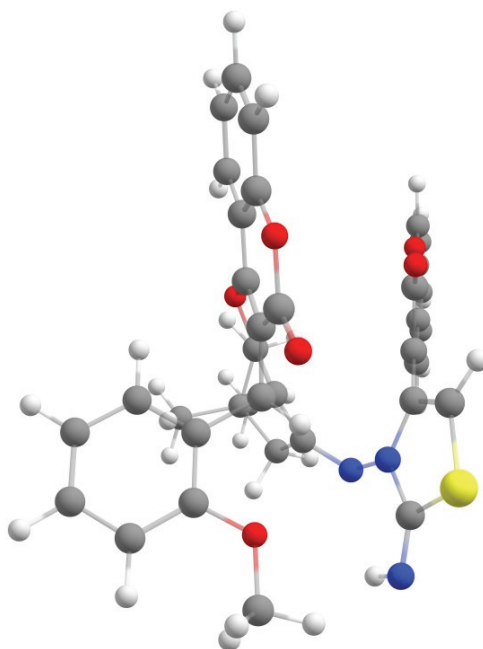
**Figure S-2.** HSQC spectra of 2-(7-(2-methoxyphenyl)-10,10-dimethyl-6-oxo-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b] chromene-8-ylidene)hydrazine-1-carbothioamide (**3a**),  $\text{CDCl}_3$



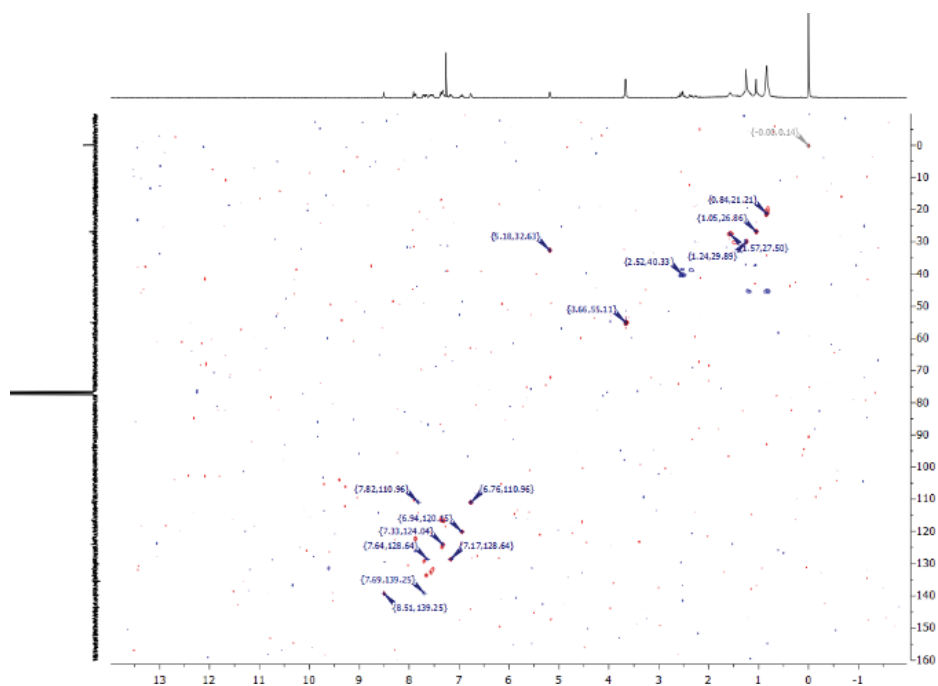
**Figure S-3.** HMBC spectra of 2-(7-(2-methoxyphenyl)-10,10-dimethyl-6-oxo-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b] chromene-8-ylidene)hydrazine-1-carbothioamide (**3a**),  $\text{CDCl}_3$ .



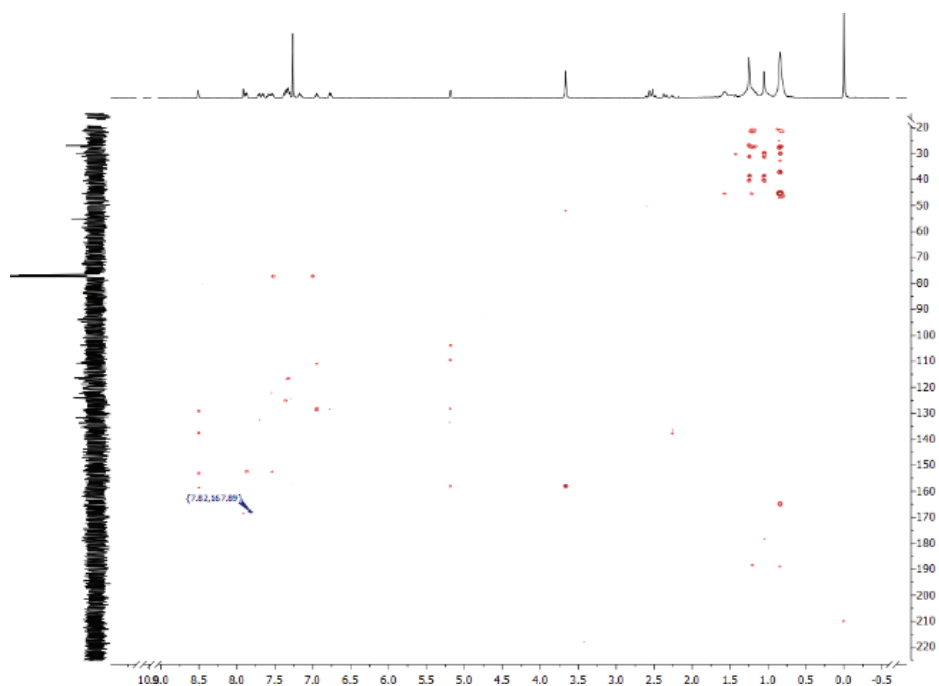
**Figure S-4.** Optimized geometry of 7-(2-methoxyphenyl)-10,10-dimethyl-8-(2-(4-(2-oxo-2H-chromen-3-yl)thiazol-2-yl)hydrazino)-8,9,10,11-tetrahydrochromeno[4,3-b]chromen-6(7H)-one (**5'a**)



**Figure S-5.** Optimized geometry of 8-((2-imino-4-(2-oxo-2H-chromen-3-yl)thiazol-3(2H)-yl)imino)-7-(2-methoxyphenyl)-10, 10-dimethyl-8,9,10,11-tetrahydrochromeno[4,3-b]chromen-6(7H)-one (**6a**)



**Figure S-6.** HSQC spectra of 10,10-dimethyl-8-(2-(4-(2-oxo-2H-chromen-3-yl)thiazol-2-yl)hydrazilidene)-7-(2-methoxyphenyl)-8,9,10,11-tetrahydro-6H,7H-chromeno[4,3-b]chromene-6-one (**5a**), CDCl<sub>3</sub>



**Figure S-7.** HMBC spectra of 10,10-dimethyl-8-(2-(4-(2-oxo-2H-chromen-3-yl)thiazol-2-yl)hydrazilidene)-7-(2-methoxyphenyl)-8,9,10,11-tetrahydro-6H,7H-chromeno[4,3-b]chromene-6-one (**5a**), CDCl<sub>3</sub>

**Table S-I.** Theoretically calculated and experimentally obtained key signals of the <sup>1</sup>H NMR spectra of the probable reaction products of the reaction 2-(7-aryl-10,10-dimethyl-6-oxo-7,9,10,11-tetrahydro-6H,8H -chromeno[4,3-b]chromen-8-ylidene)hydrazine-1-carbothioamide (3a-e) with 3-bromoacetyl coumarin

| Compound | Calc (vacuum)               | Calc (CDCl <sub>3</sub> )    | Exp (CDCl <sub>3</sub> )       |
|----------|-----------------------------|------------------------------|--------------------------------|
| 5a       | 5,7079 H <sub>52</sub> (CH) | 5,6405 H <sub>52</sub> (CH)  |                                |
|          | 6,787 H <sub>62</sub> (CH)  | 6,9343 H <sub>62</sub> (CH)  |                                |
|          | 8,065 H <sub>63</sub> (CH)  | 8,4178 H <sub>63</sub> (CH)  | 5,17 H <sub>52</sub> (CH)      |
|          | 9,8601 H <sub>61</sub> (NH) | 9,9276 H <sub>61</sub> (NH)  | 7,91 H <sub>62(68)</sub> (CH)  |
|          | 6,1523 H <sub>52</sub> (CH) | 6,0532 H <sub>52</sub> (CH)  | 8,53 H <sub>63(69)</sub> (CH)  |
| 5'a      | 6,8684 H <sub>68</sub> (CH) | 7,0554 H <sub>68</sub> (CH)  | 10,61 H <sub>61(67)</sub> (NH) |
|          | 7,7973 H <sub>69</sub> (CH) | 8,1128 H <sub>69</sub> (CH)  |                                |
|          | 8,27 H <sub>67</sub> (NH)   | 8,46 H <sub>67</sub> (NH)    |                                |
|          | 5,6635 H <sub>54</sub> (CH) | 5,5913 H <sub>54</sub> (CH)  |                                |
| 5b       | 6,7941 H <sub>63</sub> (CH) | 6,9449 H <sub>63</sub> (CH)  |                                |
|          | 8,0588 H <sub>64</sub> (CH) | 8,4164 H <sub>64</sub> (CH)  | 5,09 H <sub>54(53)</sub> (CH)  |
|          | 9,8933 H <sub>62</sub> (NH) | 9,9645 H <sub>62</sub> (NH)  | 7,87 H <sub>63(69)</sub> (CH)  |
|          | 6,0381 H <sub>53</sub> (CH) | 5,9312 H <sub>53</sub> (CH)  | 8,48 H <sub>64(70)</sub> (CH)  |
| 5'b      | 6,883 H <sub>69</sub> (CH)  | 7,0674 H <sub>69</sub> (CH)  | 10,62 H <sub>62(68)</sub> (NH) |
|          | 7,7966 H <sub>70</sub> (CH) | 8,1104 H <sub>70</sub> (CH)  |                                |
|          | 8,2601 H <sub>68</sub> (NH) | 8,4557 H <sub>68</sub> (NH)  |                                |
|          | 5,6446 H <sub>52</sub> (CH) | 5,5869 H <sub>52</sub> (CH)  |                                |
| 5c       | 6,7958 H <sub>62</sub> (CH) | 6,9445 H <sub>62</sub> (CH)  |                                |
|          | 8,0576 H <sub>63</sub> (CH) | 8,4179 H <sub>63</sub> (CH)  | 5,23 H <sub>52</sub> (CH)      |
|          | 9,9938 H <sub>61</sub> (NH) | 10,0602 H <sub>61</sub> (NH) | 7,93 H <sub>62(68)</sub> (CH)  |
|          | 5,4408 H <sub>52</sub> (CH) | 5,3825 H <sub>52</sub> (CH)  | 8,50 H <sub>63(69)</sub> (CH)  |
| 5'c      | 6,8813 H <sub>68</sub> (CH) | 7,051 H <sub>68</sub> (CH)   | 10,93 H <sub>61(67)</sub> (NH) |
|          | 7,8019 H <sub>69</sub> (CH) | 8,1115 H <sub>69</sub> (CH)  |                                |
|          | 8,1723 H <sub>67</sub> (NH) | 8,3877 H <sub>67</sub> (NH)  |                                |
|          | 5,6328 H <sub>54</sub> (CH) | 5,5729 H <sub>54</sub> (CH)  |                                |
| 5d       | 6,7292 H <sub>63</sub> (CH) | 6,8766 H <sub>63</sub> (CH)  |                                |
|          | 8,0065 H <sub>64</sub> (CH) | 8,3662 H <sub>64</sub> (CH)  | 5,14 H <sub>54(53)</sub> (CH)  |
|          | 8,0065 H <sub>62</sub> (NH) | 9,9669 H <sub>62</sub> (NH)  | 7,91 H <sub>63(69)</sub> (CH)  |
|          | 5,3841 H <sub>53</sub> (CH) | 5,3232 H <sub>53</sub> (CH)  | 8,49 H <sub>64(70)</sub> (CH)  |
| 5'd      | 6,9191 H <sub>69</sub> (CH) | 7,0928 H <sub>69</sub> (CH)  | 10,62 H <sub>62(68)</sub> (NH) |
|          | 7,7911 H <sub>70</sub> (CH) | 8,106 H <sub>70</sub> (CH)   |                                |
|          | 8,1827 H <sub>68</sub> (NH) | 8,4012 H <sub>68</sub> (NH)  |                                |
|          | 5,6262 H <sub>53</sub> (CH) | 5,5702 H <sub>53</sub> (CH)  |                                |
| 5e       | 6,7496 H <sub>62</sub> (CH) | 6,8976 H <sub>62</sub> (CH)  |                                |
|          | 8,0277 H <sub>63</sub> (CH) | 8,3876 H <sub>63</sub> (CH)  | 5,23 H <sub>54(53)</sub> (CH)  |
|          | 9,956 H <sub>61</sub> (NH)  | 10,024 H <sub>61</sub> (NH)  | 7,89 H <sub>63(69)</sub> (CH)  |
|          | 5,3814 H <sub>52</sub> (CH) | 5,3242 H <sub>52</sub> (CH)  | 8,56 H <sub>64(70)</sub> (CH)  |
| 5'e      | 6,9406 H <sub>68</sub> (CH) | 7,1132 H <sub>68</sub> (CH)  | 10,74 H <sub>62(68)</sub> (NH) |
|          | 7,8108 H <sub>69</sub> (CH) | 8,125 H <sub>70</sub> (CH)   |                                |
|          | 8,179 H <sub>67</sub> (NH)  | 8,3972 H <sub>67</sub> (NH)  |                                |

21.