

The dataset contains:

Analytical characterisation of the complexes (CHN microanalysis)

X-ray Crystallographic data:

- Structure of (S)-L¹iPr (CCDC 2091639)
- Structure of (R)-L¹tBu (CCDC 2091640)
- Structure of (S)-L¹tBu (CCDC 2091641)
- Structure of (R)-L¹tBu·(S)-L¹tBu (CCDC 2091642)
- Structure of [Fe((S)-L¹Ph)₂][BF₄]₂·MeCN (CCDC 2091644)
- Structure of [Fe((R)-L¹Ph)₂][ClO₄]₂·MeCN (CCDC 2091645)
- Structure of [Fe((S)-L¹Ph)₂][ClO₄]₂·MeCN (CCDC 2091646)
- Structure of [Fe((R)-L¹Ph)₂][FeCl₄]₂·Et₂O (CCDC 2091647)
- Structure of [Fe((R)-L¹Ph)((S)-L¹Ph)][BF₄]₂·1.5MeCN (CCDC 2091648)
- Structure of [Fe((R)-L¹Ph)₂][Fe((S)-L¹Ph)₂][ClO₄]₄·2MeCN (CCDC 2091649)
- Structure of [Fe((R)-L¹iPr)((S)-L¹iPr)][ClO₄]₂·2MeCN (CCDC 2091658)
- Structure of [Fe((S)-L¹tBu)₂][ClO₄]₂·Me₂CO (CCDC 2091659)
- Structure of [Fe((S)-L¹tBu)(NCMe)₂(OH₂)][ClO₄]₂·H₂O·0.25MeCN·0.25Et₂O (CCDC 2091660)
- Structure of [Fe((S)-L²tBu)₂][ClO₄]₂·Me₂CO (CCDC 2091661)

X-ray powder diffraction data (measured and simulated).

¹H NMR spectra (raw data and plotted spectra).

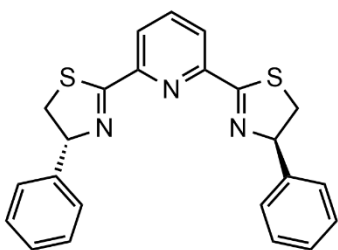
Electrospray mass spectra.

Solid state magnetic susceptibility measurements (raw and processed data).

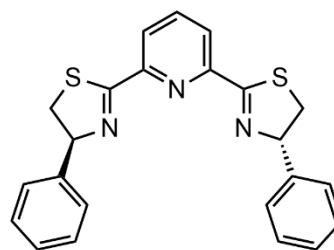
Solution magnetic susceptibility measurements (raw spectra and calculated $\chi_{\text{M}}T$ values).

DFT calculations (*SPARTAN* files).

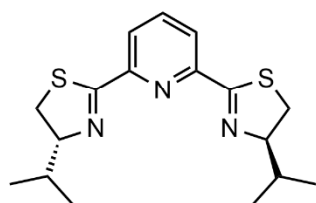
Ligands referred to in this study



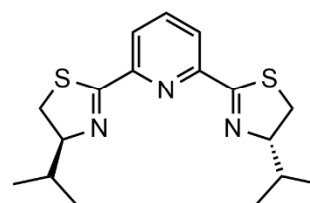
2,6-Bis(4-(*R*)-phenyl-4,5-dihydrothiazol-2-yl)pyridine
 $C_{23}H_{19}N_3S_2$
(*R*)- L^1Ph



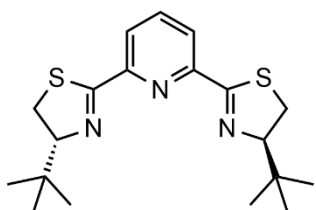
2,6-Bis(4-(*S*)-phenyl-4,5-dihydrothiazol-2-yl)pyridine
 $C_{23}H_{19}N_3S_2$
(*S*)- L^1Ph



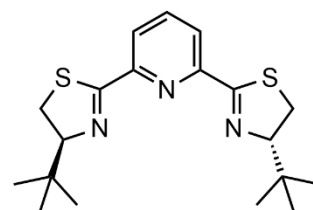
2,6-Bis(4-(*R*)-isopropyl-4,5-dihydrothiazol-2-yl)pyridine
 $C_{17}H_{23}N_3S_2$
(*R*)- L^1iPr



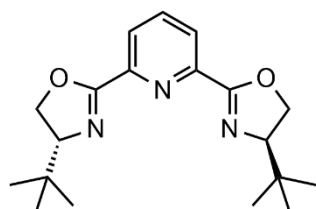
2,6-Bis(4-(*S*)-isopropyl-4,5-dihydrothiazol-2-yl)pyridine
 $C_{17}H_{23}N_3S_2$
(*S*)- L^1iPr



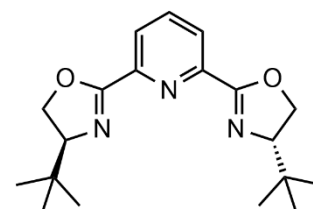
2,6-Bis(4-(*R*)-*tert*butyl-4,5-dihydrothiazol-2-yl)pyridine
 $C_{19}H_{27}N_3S_2$
(*R*)- L^1tBu



2,6-Bis(4-(*S*)-*tert*butyl-4,5-dihydrothiazol-2-yl)pyridine
 $C_{19}H_{27}N_3S_2$
(*S*)- L^1tBu

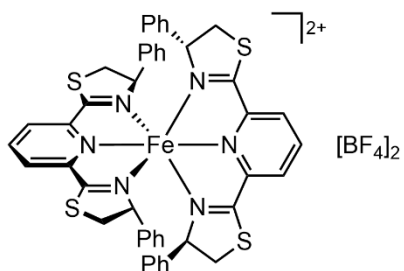


2,6-Bis(4-(*R*)-*tert*butyl-4,5-dihydrooxazol-2-yl)pyridine
 $C_{19}H_{27}N_3O_2$
(*R*)- L^2tBu

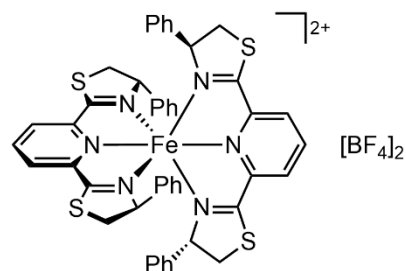


2,6-Bis(4-(*S*)-*tert*butyl-4,5-dihydrooxazol-2-yl)pyridine
 $C_{19}H_{27}N_3O_2$
(*S*)- L^2tBu

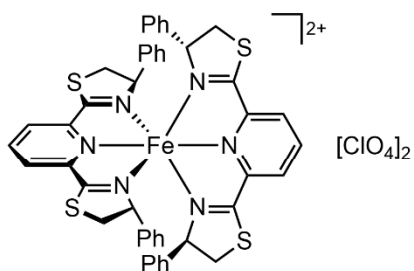
Complexes referred to in this study



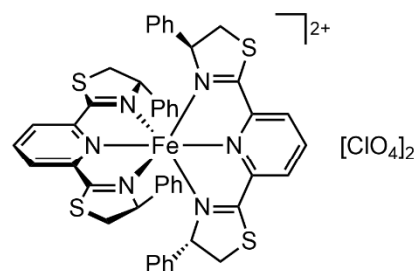
Bis[2,6-bis((*R*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II) ditetrafluoroborate
 $C_{46}H_{38}B_2F_8FeN_6S_4$
[Fe((*R*)-*L*¹Ph)₂][BF₄]₂



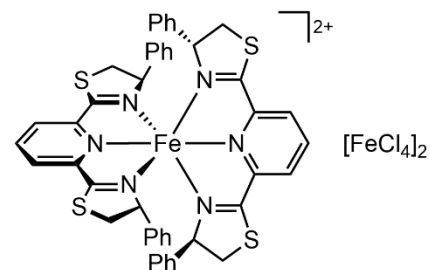
Bis[2,6-bis((*S*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II) ditetrafluoroborate
 $C_{46}H_{38}B_2F_8FeN_6S_4$
[Fe((*S*)-*L*¹Ph)₂][BF₄]₂



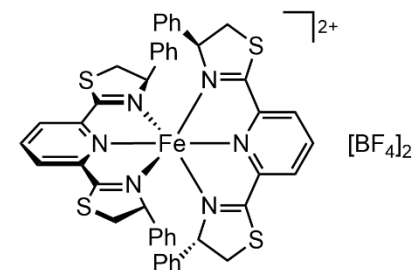
Bis[2,6-bis((*R*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II) diperchlorate
 $C_{46}H_{38}Cl_2FeN_6O_8S_4$
[Fe((*R*)-*L*¹Ph)₂][ClO₄]₂



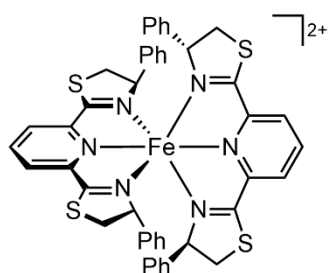
Bis[2,6-bis((*S*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II) diperchlorate
 $C_{46}H_{38}Cl_2FeN_6O_8S_4$
[Fe((*S*)-*L*¹Ph)₂][ClO₄]₂



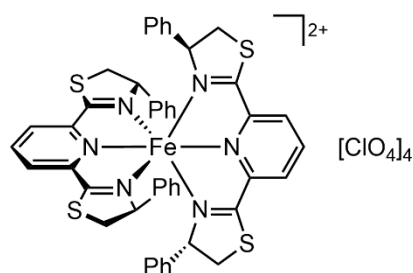
Bis[2,6-bis((*R*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II) di[tetrachloroferrate(III)]
 $C_{46}H_{38}Cl_8Fe_3N_6S_4$
[Fe((*R*)-*L*¹Ph)₂][FeCl₄]₂



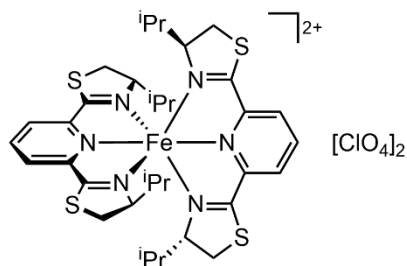
[2,6-Bis((*R*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]-
[2,6-bis((*S*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II) ditetrafluoroborate
 $C_{46}H_{38}B_2F_8FeN_6S_4$
[Fe((*R*)-*L*¹Ph)][(S)-*L*¹Ph][BF₄]₂



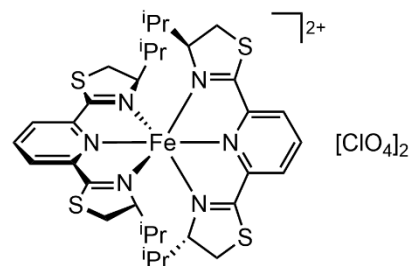
Bis[2,6-bis((*R*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]iron(II)-
bis[2,6-bis((*S*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]iron(II) diperchlorate
 $C_{92}H_{76}Cl_4Fe_2N_{12}O_{16}S_8$
[Fe((*R*)-*L*¹Ph)₂][Fe((*S*)-*L*¹Ph)₂][ClO₄]₄



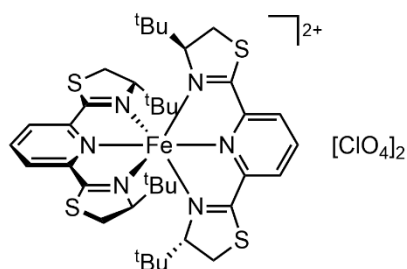
Complexes referred to in this study (continued)



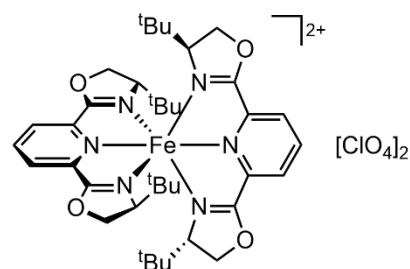
Bis[2,6-bis((*S*)-4-*isopropyl*-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II) diperchlorate
 $C_{34}H_{46}Cl_2FeN_6O_8S_4$
 $[Fe((S)-L^1iPr)_2][ClO_4]_2$



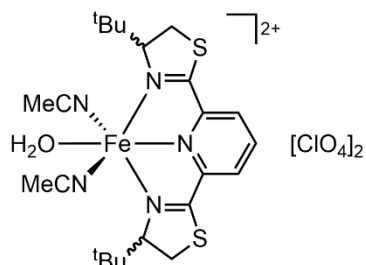
[2,6-Bis((*R*)-4-*isopropyl*-4,5-dihydrothiazol-2-yl)pyridine]-
[2,6-bis((*S*)-4-*isopropyl*-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II) diperchlorate
 $C_{34}H_{46}Cl_2FeN_6O_8S_4$
 $[Fe((R)-L^1iPr)((S)-L^1iPr)][ClO_4]_2$



Bis[2,6-bis((*S*)-4-*tert*butyl-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II) diperchlorate
 $C_{38}H_{54}Cl_2FeN_6O_8S_4$
 $[Fe((S)-L^1tBu)_2][ClO_4]_2$

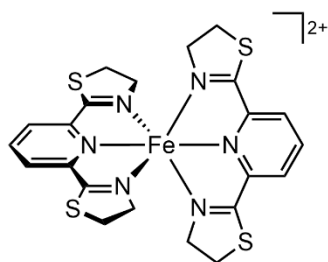


Bis[2,6-bis((*S*)-4-*tert*butyl-4,5-dihydrooxazol-2-yl)pyridine]-
iron(II) diperchlorate
 $C_{38}H_{54}Cl_2FeN_6O_{12}$
 $[Fe((S)-L^2tBu)_2][ClO_4]_2$

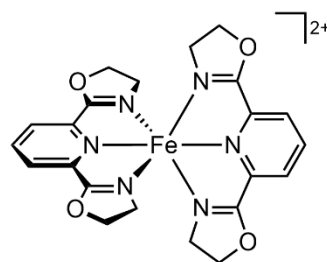


rac-[2,6-Bis(4-*tert*butyl-4,5-dihydrothiazol-2-yl)pyridine]bis(acetonitrile)-
(aqua)iron(II) diperchlorate
 $C_{23}H_{35}Cl_2FeN_5O_9S_2$
rac- $[Fe(L^1tBu)(NCMe)_2(OH_2)][ClO_4]_2$

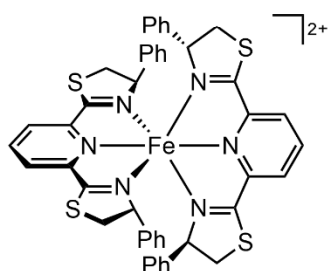
Complex cations studied by DFT calculations



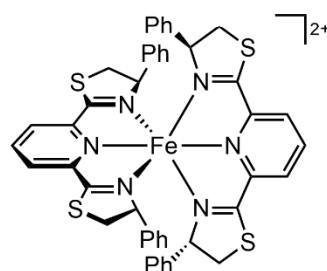
Bis[2,6-bis(4,5-dihydrothiazol-2-yl)pyridine]iron(II)
 $C_{22}H_{22}FeN_6S_4$
 $[Fe(L^1H)_2]^{2+}$



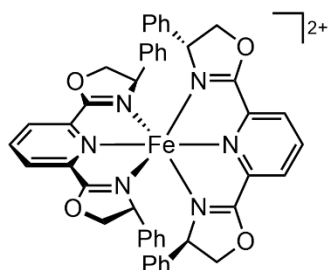
Bis[2,6-bis(4,5-dihydrooxazol-2-yl)pyridine]iron(II)
 $C_{22}H_{22}FeN_6O_4$
 $[Fe(L^2H)_2]^{2+}$



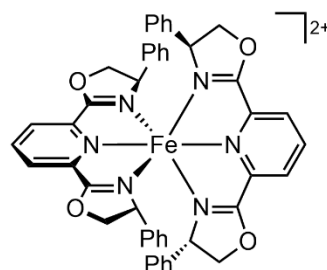
Bis[2,6-bis((*R*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]-
 iron(II)
 $C_{46}H_{38}FeN_6S_4$
 $[Fe((R)-L^1Ph)_2]^{2+}$



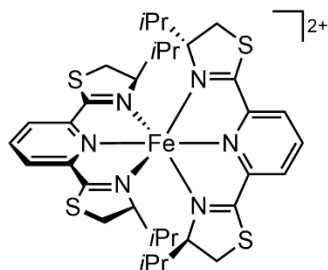
[2,6-Bis((*R*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]-
 [2,6-bis((*S*)-4-phenyl-4,5-dihydrothiazol-2-yl)pyridine]-
 iron(II)
 $C_{46}H_{38}FeN_6S_4$
 $[Fe((R)-L^1Ph)((S)-L^1Ph)]^{2+}$



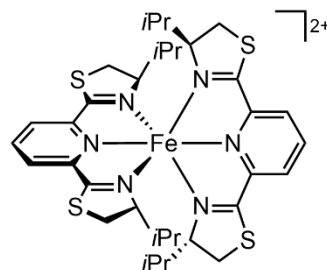
Bis[2,6-bis((*R*)-4-phenyl-4,5-dihydrooxazol-2-yl)pyridine]-
 iron(II)
 $C_{46}H_{38}FeN_6O_4$
 $[Fe((R)-L^2Ph)_2]^{2+}$



[2,6-Bis((*R*)-4-phenyl-4,5-dihydrooxazol-2-yl)pyridine]-
 [2,6-bis((*S*)-4-phenyl-4,5-dihydrooxazol-2-yl)pyridine]-
 iron(II)
 $C_{46}H_{38}FeN_6O_4$
 $[Fe((R)-L^2Ph)((S)-L^2Ph)]^{2+}$

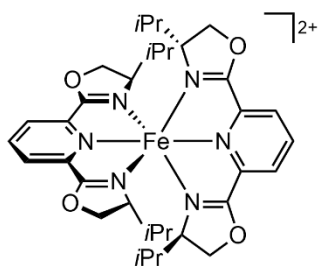


Bis[2,6-bis((*S*)-4-isopropyl-4,5-dihydrothiazol-2-yl)pyridine]-
 iron(II)
 $C_{34}H_{46}FeN_6S_4$
 $[Fe((R)-L^1iPr)_2]^{2+}$

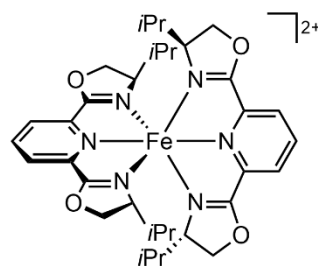
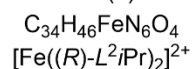


[2,6-Bis((*R*)-4-isopropyl-4,5-dihydrothiazol-2-yl)pyridine]-
 [2,6-bis((*S*)-4-isopropyl-4,5-dihydrothiazol-2-yl)pyridine]-
 iron(II)
 $C_{34}H_{46}FeN_6S_4$
 $[Fe((R)-L^1iPr)((S)-L^1iPr)]^{2+}$

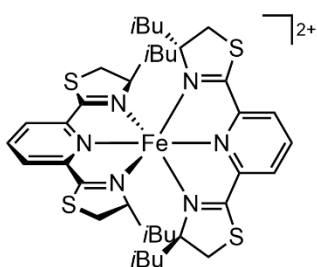
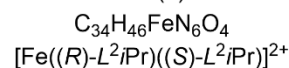
Complex cations studied by DFT calculations (continued)



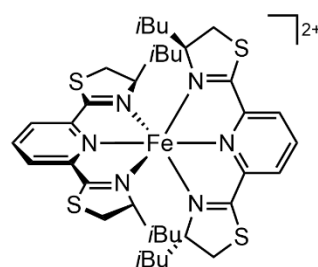
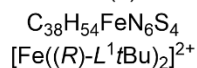
Bis[2,6-bis((*S*)-4-*isopropyl*-4,5-dihydrooxazol-2-yl)pyridine]-
iron(II)



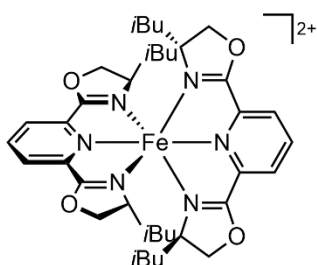
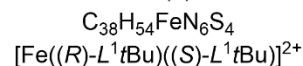
[2,6-Bis((*R*)-4-*isopropyl*-4,5-dihydrooxazol-2-yl)pyridine]-
[2,6-bis((*S*)-4-*isopropyl*-4,5-dihydrooxazol-2-yl)pyridine]-
iron(II)



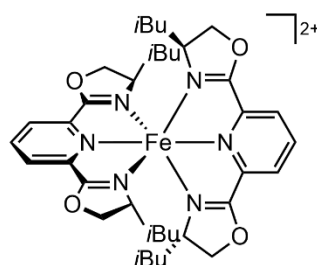
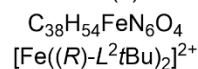
Bis[2,6-bis((*S*)-4-*tertbutyl*-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II)



[2,6-Bis((*R*)-4-*tertbutyl*-4,5-dihydrothiazol-2-yl)pyridine]-
[2,6-bis((*S*)-4-*tertbutyl*-4,5-dihydrothiazol-2-yl)pyridine]-
iron(II)



Bis[2,6-bis((*S*)-4-*tertbutyl*-4,5-dihydrooxazol-2-yl)pyridine]-
iron(II)



[2,6-Bis((*R*)-4-*tertbutyl*-4,5-dihydrooxazol-2-yl)pyridine]-
[2,6-bis((*S*)-4-*tertbutyl*-4,5-dihydrooxazol-2-yl)pyridine]-
iron(II)

