Supplementary Materials: A Mixed-Valence Tetra-Nuclear Nickel Dithiolene Complex: Synthesis, Crystal Structure, and the Lability of Its Nickel Sulfur Bonds

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1) 1H-, 13C-NMR and Mass spectra of ligand precursor compounds **1** to **3** (Figures S1–S7).
2) IR spectrum of **3** (Figure S8).
3) Mass spectrum of **4** (Figure S9).
4) IR spectra of **4** (Figure S10).
5) UV-Vis spectra of **4** (Figure S11).
6) Voltammograms of **4** (Figures S12 and S13).
7) Magnetic properties/calculations (Figures S14 and S15).
8) Crystallography (Figure S16 and Tables S1–S5).

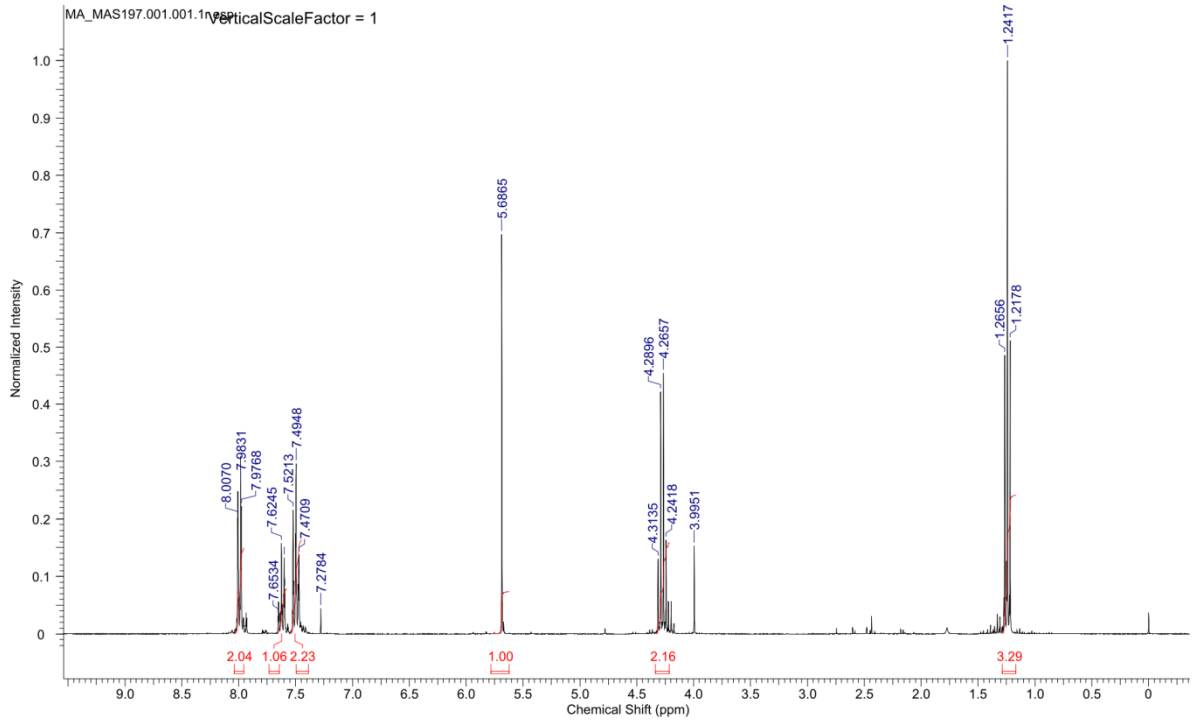

Figure S1. ^{1}H NMR of ethyl 2-bromo-3-oxo-3-phenylpropanoate (**1**) in $CDCl_3$ at 25 °C.

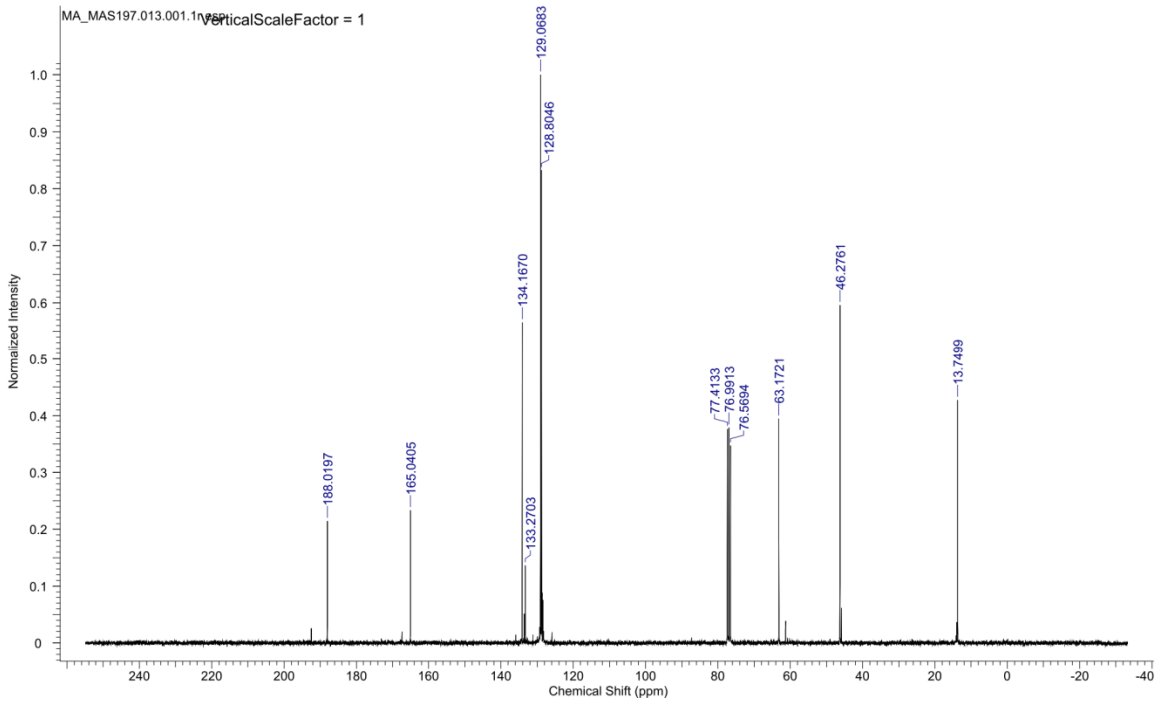

Figure S2. ^{13}C NMR of **1** in $CDCl_3$ at 25 °C.

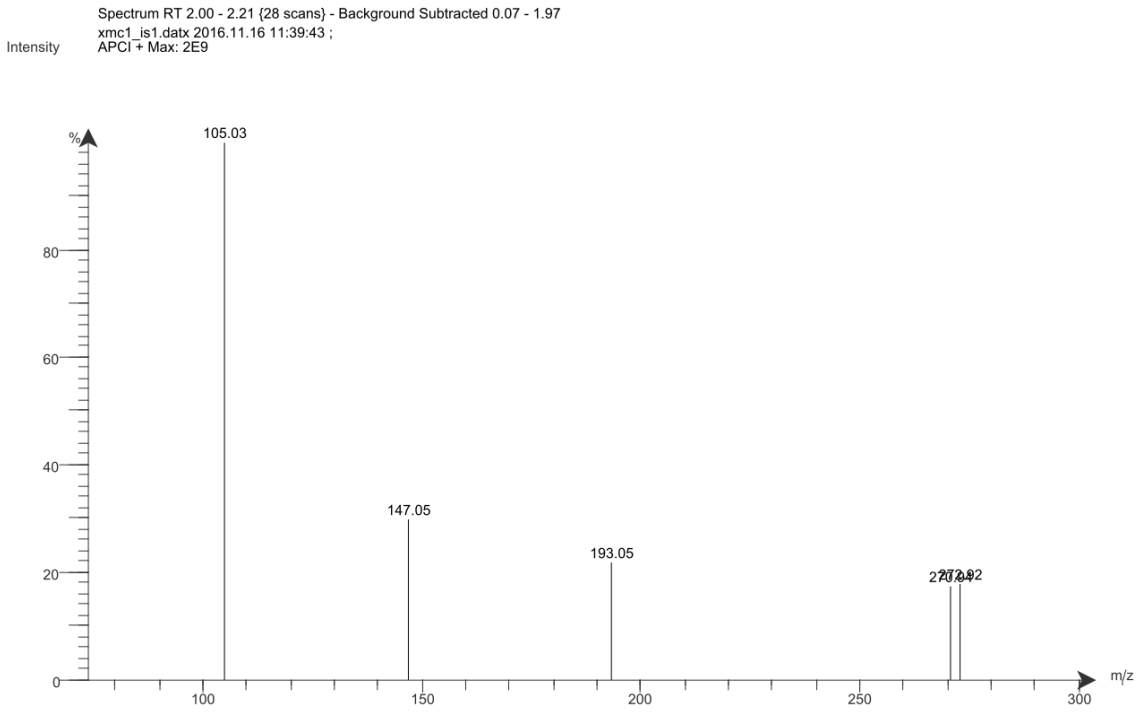

Figure S3. APCI mass spectrum of 1 in methanol.

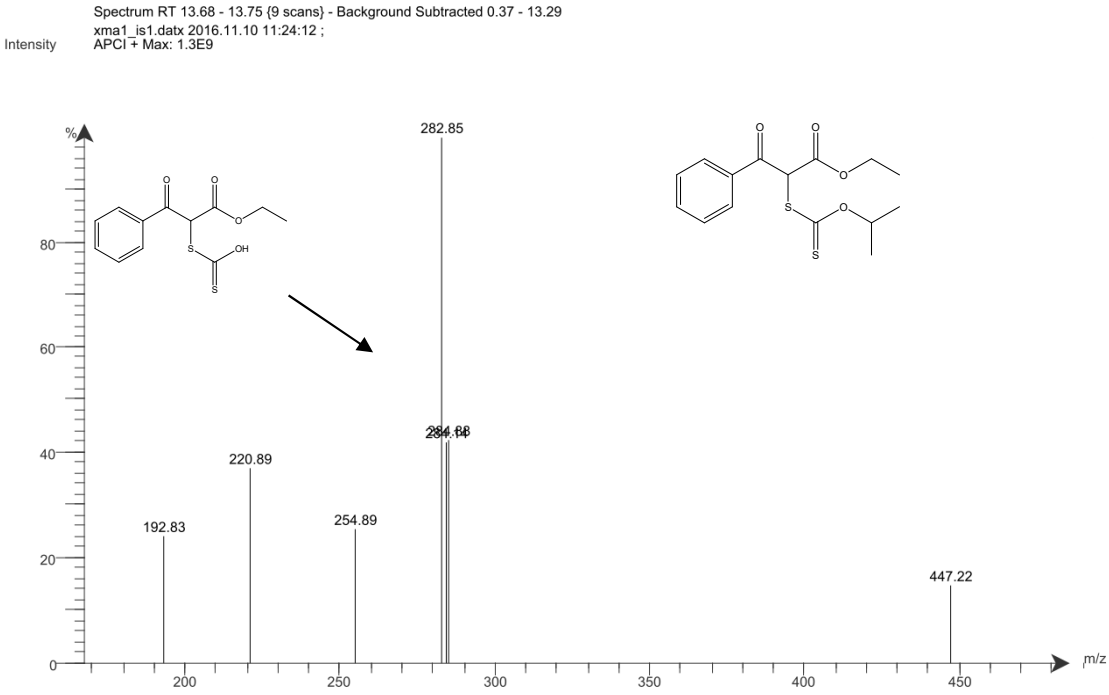

Figure S4. APCI mass spectrum of **2** in methanol.

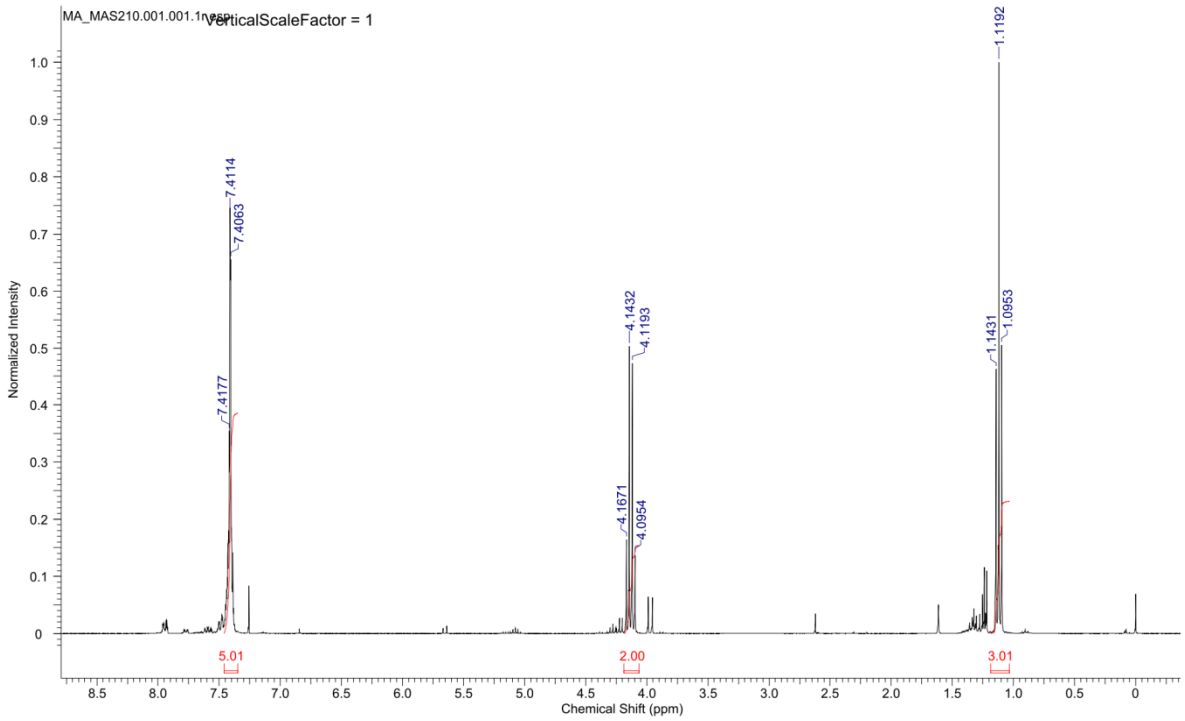

Figure S5. ^{1}H NMR of **3** (*ecpdt*) in $CDCl_3$ at 25 °C.

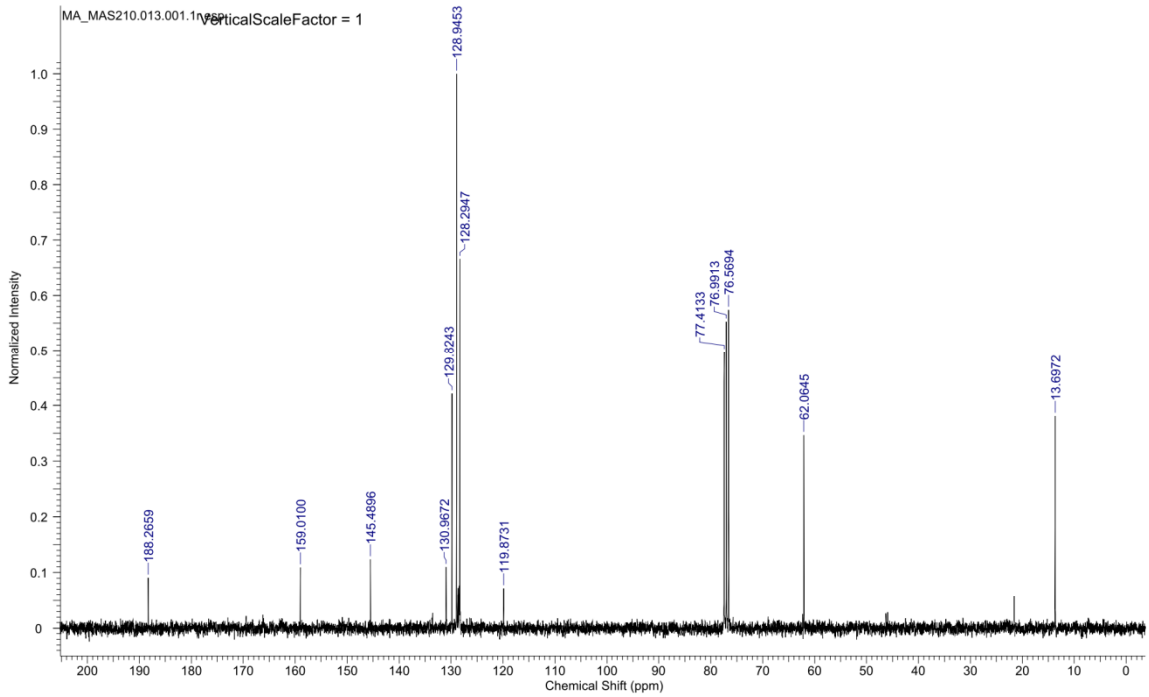

Figure S6. ^{13}C NMR of **3** in $CDCl_3$ at 25 °C.

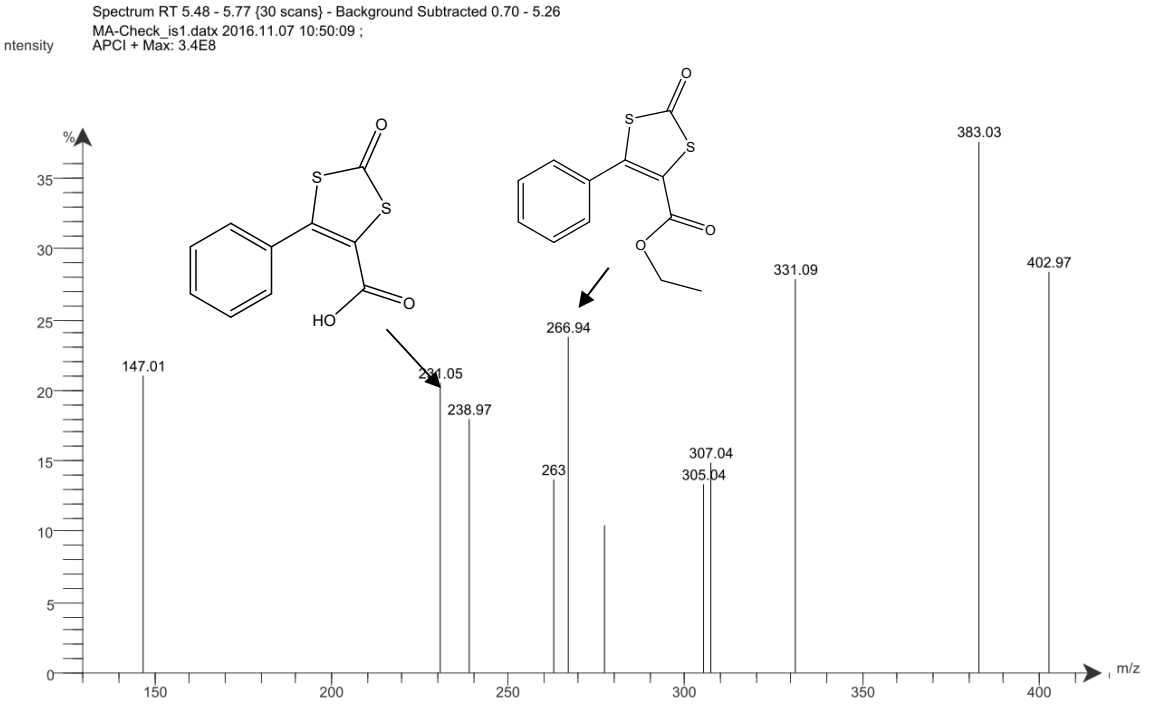

Figure S7. APCI mass spectrum of **3** in MeOH at 25 °C.

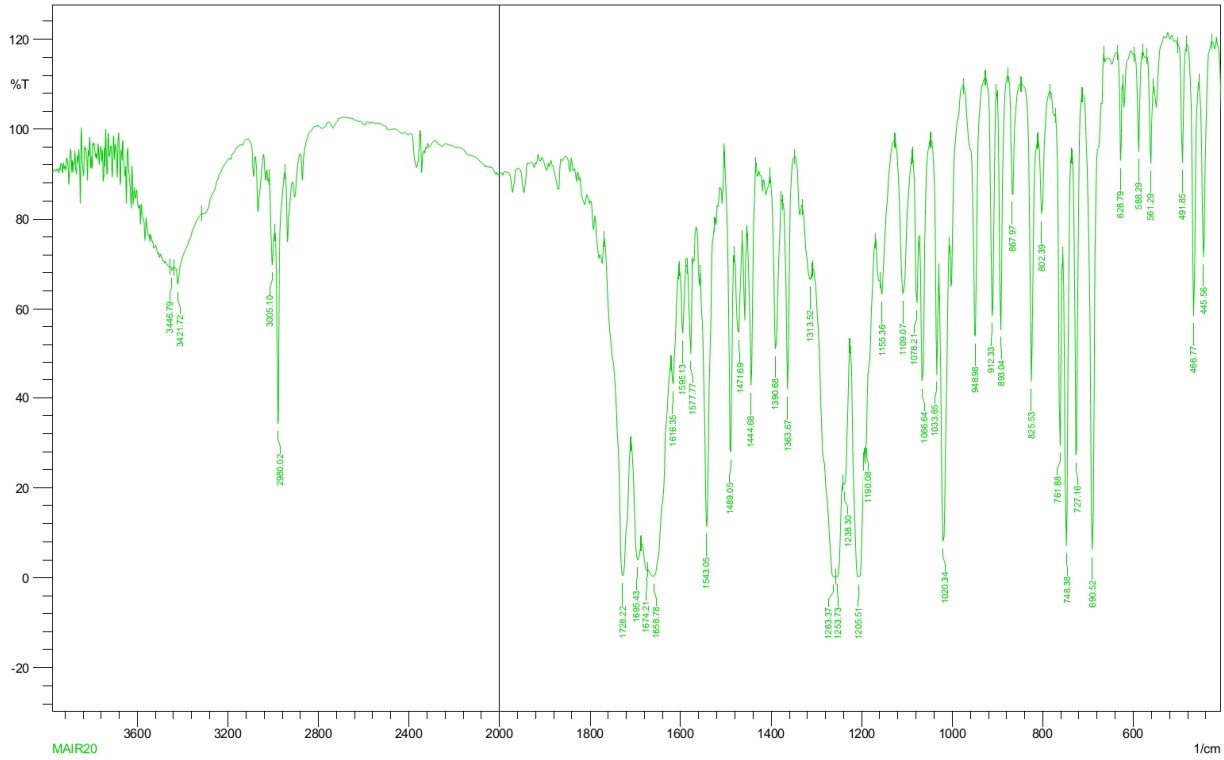

Figure S8. FT-IR spectrum of **3** in KBr plate.

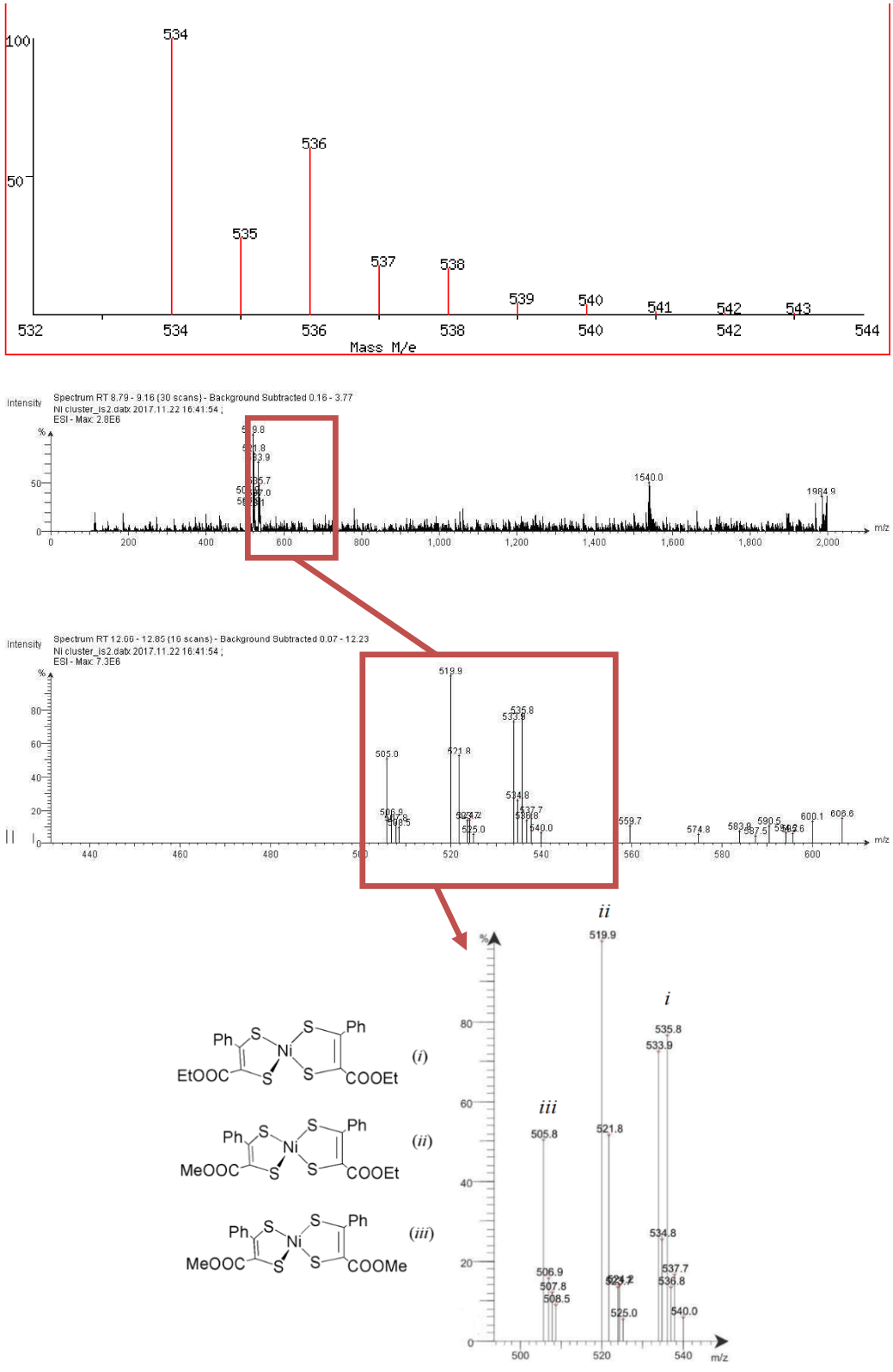

Figure S9. The ESI(−)-MS of **4** in methanol in the negative ion mode.

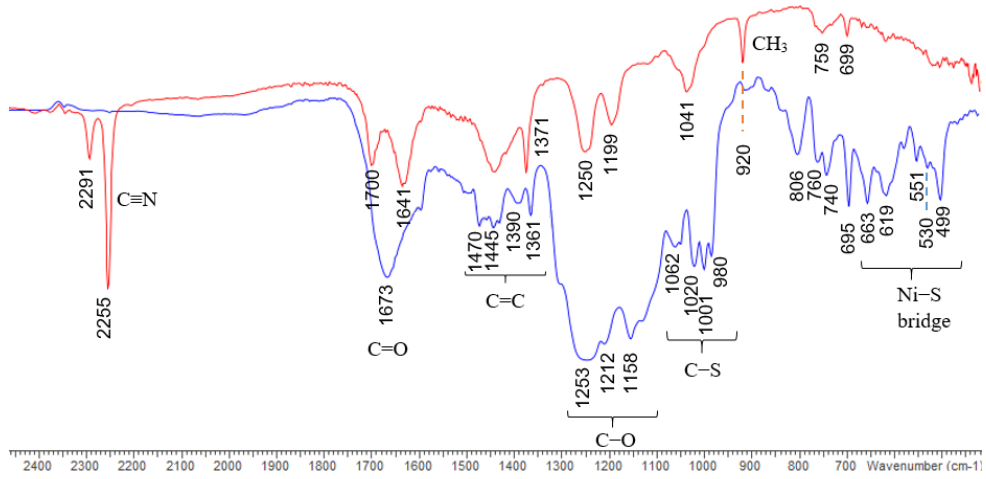

Figure S10. IR spectra of **4** in acetonitrile (red spectrum, top) and in solid-state (blue spectrum, bottom) (KBr pellet).

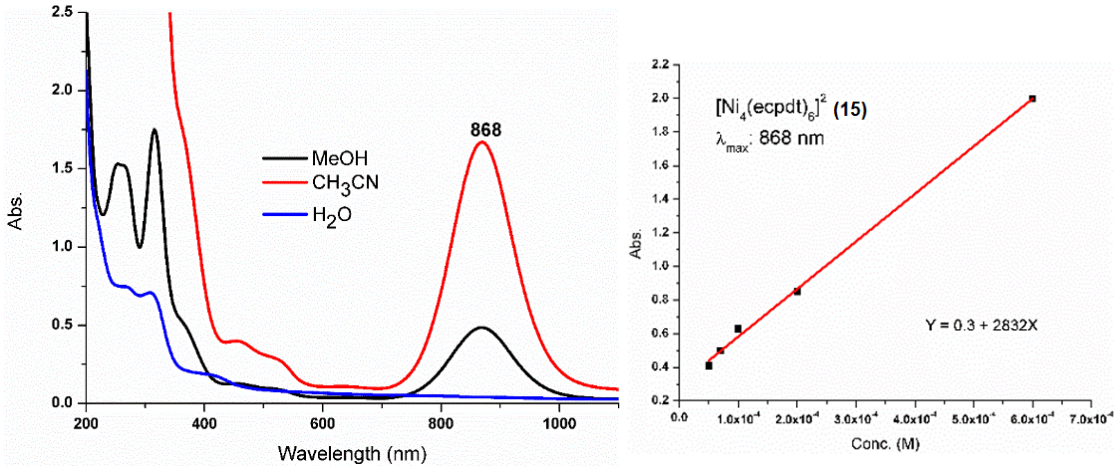

Figure S11. Left: UV-vis-NIR spectra of a light brown solution of **4** in acetonitrile, methanol, and water (5.16×10^{-4} M each); Right: Extinction coefficient diagram at 868 nm in acetonitrile.

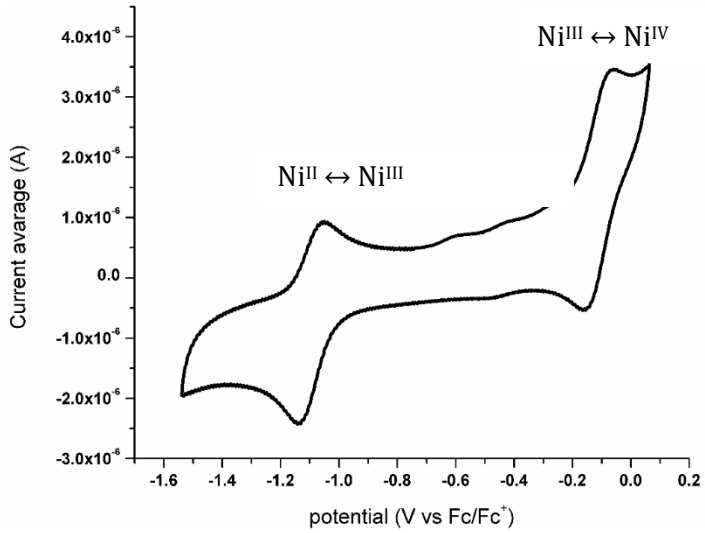

Figure S12. Cyclic voltammogram of **4** (1 mM) in acetonitrile containing 0.1 M of Bu_4NPF_6 as electrolyte (scan rate: 0.1 Vs^{-1}). The peak potentials were recorded vs. internal reference $[Fc]/[Fc]^+$ at 298 K.

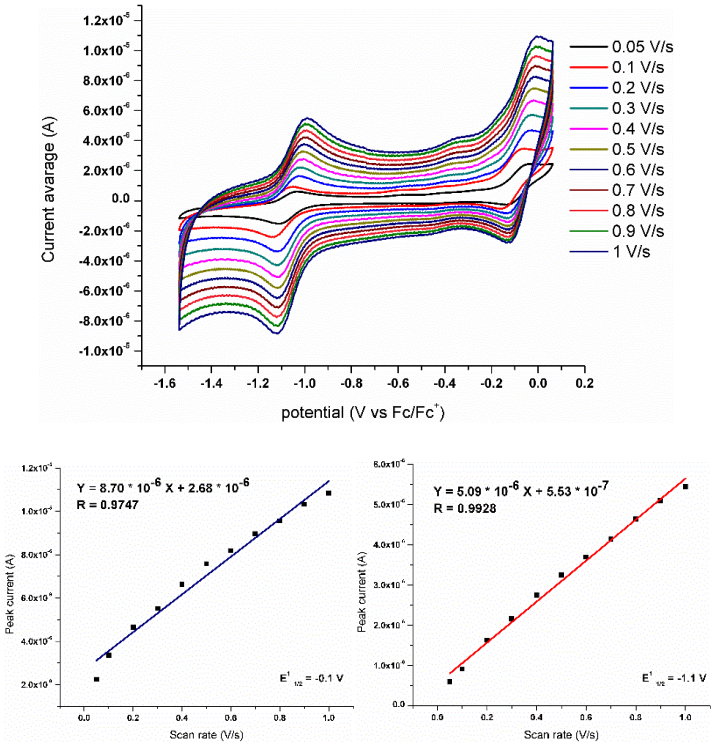

Figure S13. Cyclic voltammogram of **4** (1 mM) in acetonitrile containing 0.1 M of Bu_4NPF_6 as electrolyte at different scan rates. The peak potentials were recorded vs. internal reference Fc/Fc^+ at 298 K (left); anodic peak current vs. scan rate plots for **4** with the linear fit (right).

Magnetic Properties:

1) MSB method:

The magnetic susceptibility is calculated using the following questions:

$$\chi_{mol} = \frac{M \times C \times l \times (R - R_0)}{m \times 10^9} = 69.21 \times 10^{-4} \text{ mol}^{-1}$$

$\chi P = \chi mol - \chi D$

χD = 12 (S) + 6 (C=C) + 6 (Ph) + 18 (C=C ring) + 36 C (Ph) + 6 (COOR) + 12 (O) + 12 C (C=C) + 6 C (COOR) + 6 (C=C-Ar) + 6 (C=O) + 2 (K+) + 2 (CH_3CN) + 2 (Et_2O) = - 843.92 × 10^{-6} emu mol^{-1}

χP = [6921 – (- 843.92)] × 10^{-6} = 7764.92 × 10^{-6} emu mol^{-1}

$$\mu_{eff} = \frac{3 \times k \times T \times x\, p}{N_L \times \mu B^2} = 9.64865698 \times 10^{-23} / 5.17977615 \times 10^{-23} = 1.86 \text{ BM}$$

X_{mol} = molar susceptibility
μ_{eff} = magnetic moment
C = calibration constant 1.0304 g^{-1} cm^{-1}
M = molecular weight
l = length of sample in tube (cm) = 1.4
T = temperature [K]
R_0 = display of balance with empty tube = −41
R = display of balance with sample filled tube = +86

k = Boltzmann constant 1.380662 × 10^{-23}
N_L = 6.0225 × 10^{23} mol^{-1}
μ_B = Bohr magneton 9.274 × 10^{-24} JT^{-1} (T = Tesla)

2) Evans' method:

The magnetic moment was determined with an NMR experiment in CD_3CN at 300 K with a Bruker Avance II-300 MHz spectrometer and a 5 mm *Wilmad* Coaxial insert NMR tube. The magnetic moment (μ_{eff}) was calculated using the following equation:

$$\mu_{eff} = \frac{A}{\sqrt{{}^{c}/_{T \times \Delta ppm}}} = \frac{0.0381}{\sqrt{{}^{3.55 \cdot 10^{-3} mol/L}/_{300K \times 0.0651 ppm}}} = 2.826 \text{ BM}$$

c = concentration of **4** in mol/L (3.55·10^{-3} mol/L)
T = Temperature [K]
Δ_{ppm} = difference of chemical shift by paramagnetic substances (Figure S14-15)
A = 0.0381; constant determined by calibration with a paramagnetic substances with known μ_{eff} ($Cu(NO_3)_2$).

In this method, the NMR tube was filled with 0.5 mL of 3.55 × 10^{-3} M solution of **4** using a mixture of CD_3CN (475 µL) and *tert*-Butanol (25 µL) as the solvent. The capillary tube was filled with 0.5 mL of the same solvent mixture. Then, the capillary was placed into the NMR tube and the spectrum measured at 300 K. The difference of the chemical shift of the methyl protons of *t*-BuOH due to the presence of a paramagnetic substance was found to be 0.0651 (Figure S14). For calibration, we used the same method employing 14.07 × 10^{-3} M of $Cu(NO_3)_2$ as a known paramagnetic compound. The difference in the chemical shift of the methyl protons of *t*-BuOH was found to be 0.1216 (Figure S15). Value A in the above equation was calculated to be 0.0381 with the known μ_{eff} value of the paramagnetic substance (1.94 BM) (S = ½).[E. A. Boudreaux and D. J. Miller, *Inorganic and Nuclear Chemistry Letters*, 1966, **2**, 59–61.]

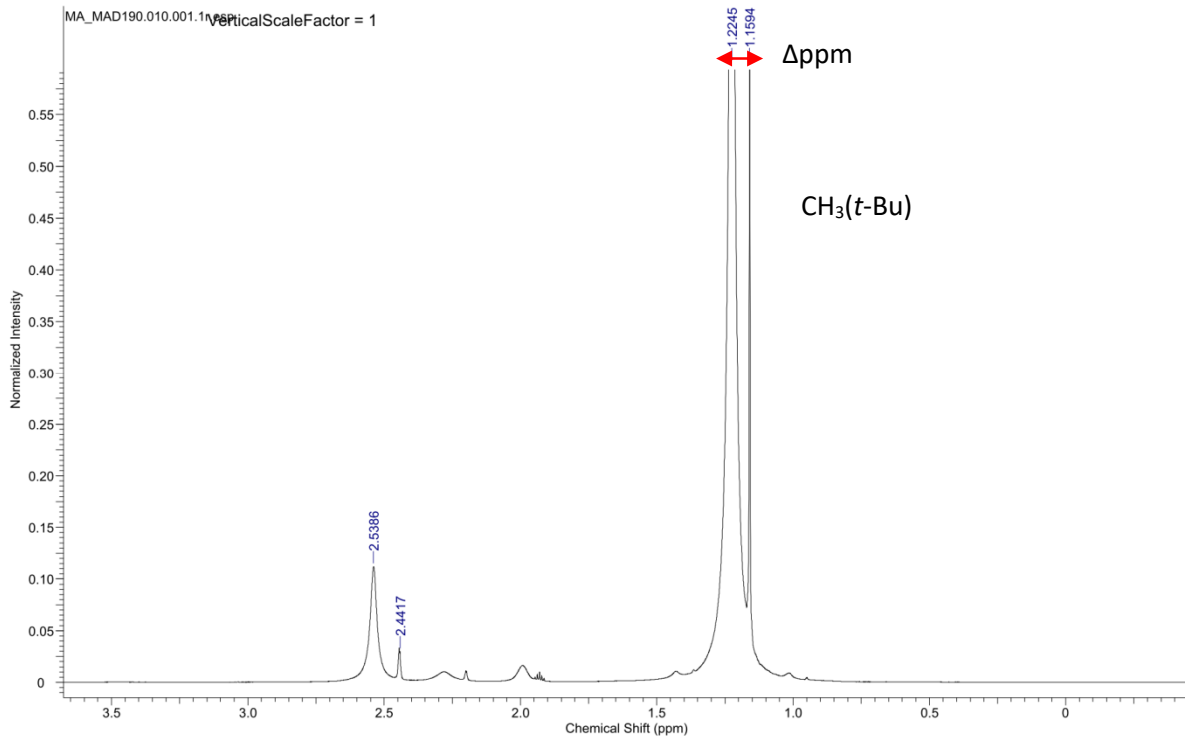

Figure S14. ^{1}H NMR for measuring the magnetic moment of the Ni cluster in CD_3CN at 25 °C (475 µL CD_3CN+ 22 µL *t*-BuOH).

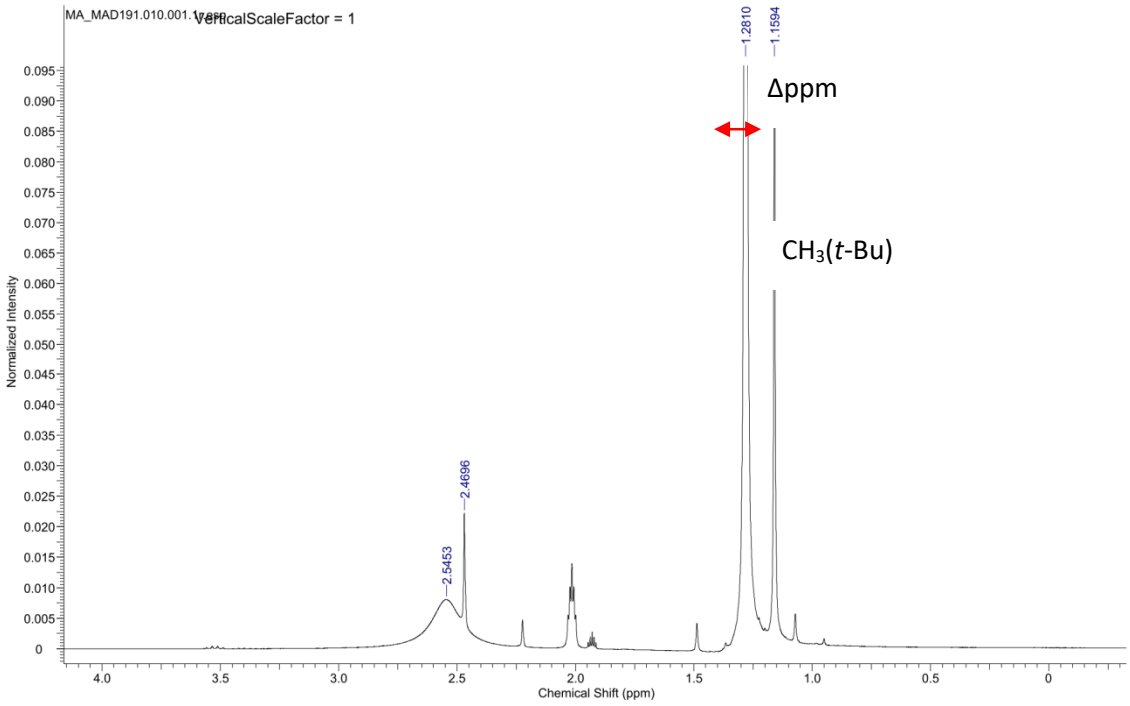

Figure S15. ^{1}H NMR for measuring the magnetic moment of the $Cu(NO_3)_2$ in CD_3CN at 25 °C (475 µL CD_3CN+ 22 µL *t*-BuOH).

Crystallography:

Table S1. General crystallographic data of **3** and **4·2CH$_3$CN·2Et$_2$O** at 170 K.

	3	**4·2CH$_3$CN·2Et$_2$O**
CCDC number	1985587	1985588
Formula	$C_{12}H_{10}O_3S_2$	$C_{68}H_{60}K_2N_2Ni_4O_{14}S_{14}$
Mw	266.32	1889.09
Crystal system	Monoclinic	Monoclinic
Space group	*C*2/*c*	*P*2$_1$/*n*
a [Å]	30.240(6)	13.588(3)
b [Å]	3.9413(8)	19.062(4)
c [Å]	20.418(4)	16.764(3)
β [°]	107.77(3)	100.11(3)
Z	8	2
V [Å^3]	2317.5(9)	4274.8(15)
d_{calc} [g/cm^3]	1.527	1.469
μ [mm^{-1}]	0.451	1.316
F(0000)	1104	1948
Theta range for data collection [°]	3.994 to 25.678	1.632 to 25.684
reflns collected / unique reflns / R_{int}	7984 / 2175 /	32961 / 8052 / 0.0937
Data / restraints / parameters	2175 / 0 / 155	8052 / 245 / 538
R_1 / wR_2 (I>2σ(I))	0.0501 / 0.1103	0.0568 / 0.1466
R_1 / wR_2 (all data)	0.0944 / 0.1292	0.1006 / 0.1668
GOF [F^2]	0.940	0.921
residual density [e·Å^{-3}]	0.281 / −0.378	1.672 / −0.714

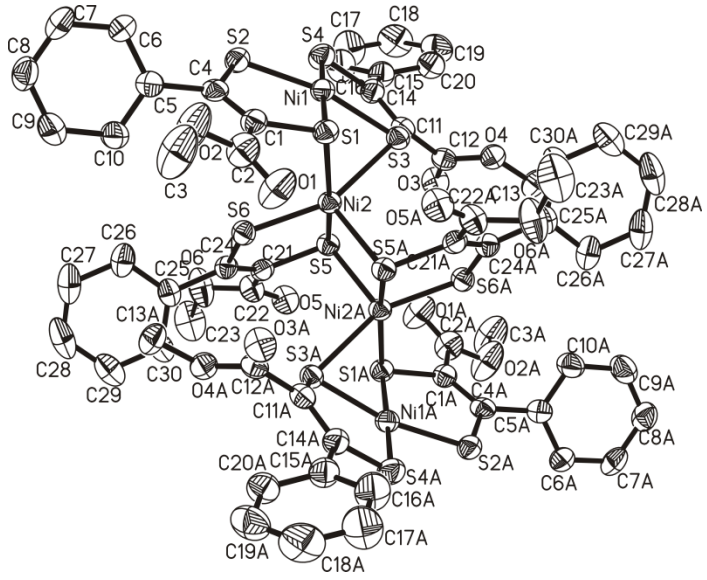

Figure S16. Molecular structure of the di-anion of **4·CH$_3$CN·Et$_2$O** shown with 50% probability thermal ellipsoids (the two potassium counter cations, disordered minor occupancies, H atoms and lattice solvents are omitted for clarity reasons).

Table S2. Complete list of bond lengths [Å] and angles [°] for **4·2CH₃CN·2Et₂O**.

K1—O1^{i}	2.591 (5)	C15—C16	1.382 (10)
K1—O3	2.651 (5)	C15—C20	1.403 (9)
K1—O5	2.676 (5)	C16—C17	1.391 (10)
K1—O7	2.744 (8)	C16—H16	0.9500
K1—N1	2.831 (8)	C17—C18	1.339 (12)
K1—S5	3.378 (2)	C17—H17	0.9500
K1—C22	3.511 (6)	C18—C19	1.389 (11)
K1—C35'	3.53 (3)	C18—H18	0.9500
Ni1—S4	2.1464 (18)	C19—C20	1.412 (9)
Ni1—S2	2.1482 (17)	C19—H19	0.9500
Ni1—S1	2.1487 (17)	C20—H20	0.9500
Ni1—S3	2.1725 (17)	C21—C24	1.369 (8)
Ni1—Ni2	2.6747 (11)	C21—C22	1.488 (8)
Ni2—S5	2.1758 (16)	C23—H23A	0.9800
Ni2—S6	2.1793 (16)	C23—H23B	0.9800
Ni2—S1	2.3125 (16)	C23—H23C	0.9800
Ni2—S3	2.3509 (16)	C23'—H23D	0.9800
Ni2—S5^{i}	2.3554 (16)	C23'—H23E	0.9800
Ni2—Ni2^{i}	2.9187 (16)	C23'—H23F	0.9800
S1—C1	1.745 (6)	C24—C25	1.501 (8)
S2—C4	1.711 (6)	C25—C30	1.383 (9)
S3—C11	1.749 (6)	C25—C26	1.395 (9)
S4—C14	1.729 (6)	C26—C27	1.388 (10)
S5—C21	1.748 (6)	C26—H26	0.9500
S6—C24	1.712 (6)	C27—C28	1.373 (12)
O1—C2	1.215 (8)	C27—H27	0.9500
O2—C2	1.338 (8)	C28—C29	1.396 (11)
O2—C3	1.522 (13)	C28—H28	0.9500
O2—C3'	1.593 (17)	C29—C30	1.357 (9)
O3—C12	1.204 (7)	C29—H29	0.9500
O4—C12	1.337 (7)	C30—H30	0.9500
O4—C13	1.448 (7)	C31—C32	1.473 (13)
O5—C22	1.211 (7)	C32—H32A	0.9800
O6—C22	1.335 (7)	C32—H32B	0.9800
O6—C23'	1.483 (11)	C32—H32C	0.9800
O6—C23	1.516 (16)	O7—C35	1.420 (17)

N1—C31	1.219 (13)	O7—C33'	1.422 (18)
C1—C4	1.366 (8)	O7—C33	1.474 (18)
C1—C2	1.475 (9)	O7—C35'	1.487 (19)
C3—H3A	0.9800	C33—C34	1.63 (2)
C3—H3B	0.9800	C33—H33A	0.9900
C3—H3C	0.9800	C33—H33B	0.9900
C3'—H3A1	0.9800	C34—H34A	0.9800
C3'—H3A2	0.9800	C34—H34B	0.9800
C3'—H3A3	0.9800	C34—H34C	0.9800
C4—C5	1.495 (8)	C35—C36	1.64 (2)
C5—C6	1.385 (8)	C35—H35A	0.9900
C5—C10	1.398 (9)	C35—H35B	0.9900
C6—C7	1.388 (9)	C36—H36A	0.9800
C6—H6	0.9500	C36—H36B	0.9800
C7—C8	1.369 (10)	C36—H36C	0.9800
C7—H7	0.9500	C33'—C34'	1.67 (2)
C8—C9	1.361 (10)	C33'—H33C	0.9900
C8—H8	0.9500	C33'—H33D	0.9900
C9—C10	1.403 (9)	C34'—H34D	0.9800
C9—H9	0.9500	C34'—H34E	0.9800
C10—H10	0.9500	C34'—H34F	0.9800
C11—C14	1.364 (8)	C35'—C36'	1.60 (2)
C11—C12	1.490 (8)	C35'—H35C	0.9900
C13—H13A	0.9800	C35'—H35D	0.9900
C13—H13B	0.9800	C36'—H36D	0.9800
C13—H13C	0.9800	C36'—H36E	0.9800
C14—C15	1.478 (9)	C36'—H36F	0.9800
O1^{i}—K1—O3	82.86 (18)	O4—C13—H13C	109.5
O1^{i}—K1—O5	84.33 (16)	H13A—C13—H13C	109.5
O3—K1—O5	135.41 (15)	H13B—C13—H13C	109.5
O1^{i}—K1—O7	108.6 (2)	C11—C14—C15	123.7 (5)
O3—K1—O7	115.7 (2)	C11—C14—S4	120.8 (5)
O5—K1—O7	108.9 (2)	C15—C14—S4	115.4 (4)
O1^{i}—K1—N1	155.3 (2)	C16—C15—C20	118.2 (6)
O3—K1—N1	105.8 (2)	C16—C15—C14	121.1 (6)
O5—K1—N1	73.2 (2)	C20—C15—C14	120.6 (6)

O7—K1—N1	88.7 (3)	C15—C16—C17	121.5 (8)
O1^{i}—K1—S5	85.83 (12)	C15—C16—H16	119.2
O3—K1—S5	79.58 (10)	C17—C16—H16	119.2
O5—K1—S5	56.99 (10)	C18—C17—C16	120.4 (8)
O7—K1—S5	159.6 (2)	C18—C17—H17	119.8
N1—K1—S5	73.4 (2)	C16—C17—H17	119.8
O1^{i}—K1—C22	95.65 (16)	C17—C18—C19	120.8 (7)
O3—K1—C22	125.56 (15)	C17—C18—H18	119.6
O5—K1—C22	16.48 (13)	C19—C18—H18	119.6
O7—K1—C22	116.1 (2)	C18—C19—C20	119.5 (7)
N1—K1—C22	60.3 (2)	C18—C19—H19	120.2
S5—K1—C22	46.21 (10)	C20—C19—H19	120.2
O1^{i}—K1—C35'	112.2 (5)	C15—C20—C19	119.6 (7)
O3—K1—C35'	92.8 (5)	C15—C20—H20	120.2
O5—K1—C35'	131.5 (5)	C19—C20—H20	120.2
O7—K1—C35'	23.4 (5)	C24—C21—C22	127.7 (5)
N1—K1—C35'	90.8 (5)	C24—C21—S5	119.0 (5)
S5—K1—C35'	159.6 (5)	C22—C21—S5	113.3 (4)
C22—K1—C35'	135.6 (5)	O5—C22—O6	123.5 (6)
S4—Ni1—S2	90.62 (7)	O5—C22—C21	124.1 (6)
S4—Ni1—S1	176.32 (7)	O6—C22—C21	112.2 (5)
S2—Ni1—S1	91.82 (6)	O5—C22—K1	38.8 (3)
S4—Ni1—S3	92.50 (7)	O6—C22—K1	131.2 (4)
S2—Ni1—S3	167.10 (7)	C21—C22—K1	99.8 (4)
S1—Ni1—S3	84.52 (6)	O6—C23—H23A	109.5
S4—Ni1—Ni2	120.45 (6)	O6—C23—H23B	109.5
S2—Ni1—Ni2	110.99 (6)	H23A—C23—H23B	109.5
S1—Ni1—Ni2	56.03 (4)	O6—C23—H23C	109.5
S3—Ni1—Ni2	56.90 (5)	H23A—C23—H23C	109.5
S5—Ni2—S6	90.24 (6)	H23B—C23—H23C	109.5
S5—Ni2—S1	164.00 (6)	O6—C23'—H23D	109.5
S6—Ni2—S1	92.80 (6)	O6—C23'—H23E	109.5
S5—Ni2—S3	94.06 (6)	H23D—C23'—H23E	109.5
S6—Ni2—S3	156.26 (6)	O6—C23'—H23F	109.5
S1—Ni2—S3	77.09 (6)	H23D—C23'—H23F	109.5
S5—Ni2—S5^{i}	99.91 (6)	H23E—C23'—H23F	109.5
S6—Ni2—S5^{i}	99.79 (6)	C21—C24—C25	124.6 (5)

S1—Ni2—S5^{i}	95.06 (6)	C21—C24—S6	120.4 (5)
S3—Ni2—S5^{i}	102.43 (5)	C25—C24—S6	115.0 (4)
S5—Ni2—Ni1	113.74 (5)	C30—C25—C26	119.0 (6)
S6—Ni2—Ni1	106.42 (5)	C30—C25—C24	121.8 (5)
S1—Ni2—Ni1	50.40 (4)	C26—C25—C24	119.1 (6)
S3—Ni2—Ni1	50.73 (4)	C27—C26—C25	119.6 (7)
S5^{i}—Ni2—Ni1	136.58 (5)	C27—C26—H26	120.2
S5—Ni2—Ni2^{i}	52.65 (4)	C25—C26—H26	120.2
S6—Ni2—Ni2^{i}	98.07 (5)	C28—C27—C26	120.4 (7)
S1—Ni2—Ni2^{i}	141.97 (6)	C28—C27—H27	119.8
S3—Ni2—Ni2^{i}	103.09 (5)	C26—C27—H27	119.8
S5^{i}—Ni2—Ni2^{i}	47.25 (4)	C27—C28—C29	119.7 (7)
Ni1—Ni2—Ni2^{i}	152.41 (4)	C27—C28—H28	120.2
C1—S1—Ni1	103.8 (2)	C29—C28—H28	120.2
C1—S1—Ni2	112.4 (2)	C30—C29—C28	119.9 (7)
Ni1—S1—Ni2	73.57 (6)	C30—C29—H29	120.1
C4—S2—Ni1	104.3 (2)	C28—C29—H29	120.1
C11—S3—Ni1	102.3 (2)	C29—C30—C25	121.3 (7)
C11—S3—Ni2	117.9 (2)	C29—C30—H30	119.3
Ni1—S3—Ni2	72.38 (5)	C25—C30—H30	119.3
C14—S4—Ni1	103.5 (2)	N1—C31—C32	175.3 (11)
C21—S5—Ni2	104.7 (2)	C31—C32—H32A	109.5
C21—S5—Ni2^{i}	107.23 (19)	C31—C32—H32B	109.5
Ni2—S5—Ni2^{i}	80.09 (6)	H32A—C32—H32B	109.5
C21—S5—K1	99.0 (2)	C31—C32—H32C	109.5
Ni2—S5—K1	146.21 (7)	H32A—C32—H32C	109.5
Ni2^{i}—S5—K1	115.46 (6)	H32B—C32—H32C	109.5
C24—S6—Ni2	105.2 (2)	C35—O7—C33	102.4 (16)
C2—O1—K1^{i}	168.6 (5)	C33'—O7—C35'	120.3 (18)
C2—O2—C3	117.3 (8)	C35—O7—K1	127.9 (13)
C2—O2—C3'	109.9 (11)	C33'—O7—K1	123.6 (14)
C12—O3—K1	133.7 (4)	C33—O7—K1	112.4 (11)
C12—O4—C13	116.1 (5)	C35'—O7—K1	109.5 (15)
C22—O5—K1	124.7 (4)	O7—C33—C34	84.9 (12)
C22—O6—C23'	116.5 (8)	O7—C33—H33A	114.5
C22—O6—C23	111.6 (13)	C34—C33—H33A	114.5
C31—N1—K1	134.3 (8)	O7—C33—H33B	114.5

C4—C1—C2	127.3 (5)	C34—C33—H33B	114.5
C4—C1—S1	119.3 (4)	H33A—C33—H33B	111.6
C2—C1—S1	113.3 (4)	C33—C34—H34A	109.5
O1—C2—O2	122.2 (6)	C33—C34—H34B	109.5
O1—C2—C1	124.2 (6)	H34A—C34—H34B	109.5
O2—C2—C1	113.5 (6)	C33—C34—H34C	109.5
O2—C3—H3A	109.5	H34A—C34—H34C	109.5
O2—C3—H3B	109.5	H34B—C34—H34C	109.5
H3A—C3—H3B	109.5	O7—C35—C36	85.0 (12)
O2—C3—H3C	109.5	O7—C35—H35A	114.5
H3A—C3—H3C	109.5	C36—C35—H35A	114.5
H3B—C3—H3C	109.5	O7—C35—H35B	114.5
O2—C3'—H3A1	109.5	C36—C35—H35B	114.5
O2—C3'—H3A2	109.5	H35A—C35—H35B	111.6
H3A1—C3'—H3A2	109.5	C35—C36—H36A	109.5
O2—C3'—H3A3	109.5	C35—C36—H36B	109.5
H3A1—C3'—H3A3	109.5	H36A—C36—H36B	109.5
H3A2—C3'—H3A3	109.5	C35—C36—H36C	109.5
C1—C4—C5	125.2 (5)	H36A—C36—H36C	109.5
C1—C4—S2	120.4 (4)	H36B—C36—H36C	109.5
C5—C4—S2	114.3 (4)	O7—C33'—C34'	83.7 (12)
C6—C5—C10	119.4 (6)	O7—C33'—H33C	114.7
C6—C5—C4	121.1 (5)	C34'—C33'—H33C	114.7
C10—C5—C4	119.4 (5)	O7—C33'—H33D	114.7
C5—C6—C7	120.1 (6)	C34'—C33'—H33D	114.7
C5—C6—H6	119.9	H33C—C33'—H33D	111.8
C7—C6—H6	119.9	C33'—C34'—H34D	109.5
C8—C7—C6	120.5 (6)	C33'—C34'—H34E	109.5
C8—C7—H7	119.7	H34D—C34'—H34E	109.5
C6—C7—H7	119.7	C33'—C34'—H34F	109.5
C9—C8—C7	120.1 (6)	H34D—C34'—H34F	109.5
C9—C8—H8	119.9	H34E—C34'—H34F	109.5
C7—C8—H8	119.9	O7—C35'—C36'	84.5 (13)
C8—C9—C10	120.8 (7)	O7—C35'—K1	47.1 (11)
C8—C9—H9	119.6	C36'—C35'—K1	87.7 (15)
C10—C9—H9	119.6	O7—C35'—H35C	114.6
C5—C10—C9	119.0 (6)	C36'—C35'—H35C	114.6

C5—C10—H10	120.5	K1—C35'—H35C	151.5
C9—C10—H10	120.5	O7—C35'—H35D	114.6
C14—C11—C12	122.0 (5)	C36'—C35'—H35D	114.6
C14—C11—S3	119.6 (5)	K1—C35'—H35D	70.6
C12—C11—S3	118.2 (4)	H35C—C35'—H35D	111.7
O3—C12—O4	123.0 (6)	C35'—C36'—H36D	109.5
O3—C12—C11	124.2 (6)	C35'—C36'—H36E	109.5
O4—C12—C11	112.8 (5)	H36D—C36'—H36E	109.5
O4—C13—H13A	109.5	C35'—C36'—H36F	109.5
O4—C13—H13B	109.5	H36D—C36'—H36F	109.5
H13A—C13—H13B	109.5	H36E—C36'—H36F	109.5

Symmetry code: (i) −*x*+1, −*y*+1, −*z*+1.

Table S3. Hydrogen-bond geometry [Å and °] of **4·2CH$_3$CN·2Et$_2$O**.

Donor–H···Acceptor	[ARU]	D - H	H···A	D···A	D - H···A
C8 –H8 ···O4	[2545]	0.95	2.55	3.420(8)	152
C32–H32A ···N1	[3766]	0.98	2.45	3.276(13)	142
C32 –H32B ···S2$_{intra}$	[3666]	0.98	2.82	3.727(9)	153

Translation of ARU-Code to CIF and Equivalent Position Code

[3666] = 1−x,1−y,1−z
[3766] = 2−x,1−y,1−z
[2545] = 1/2−x,−1/2+y,1/2−z

Table S4. Atomic coordinates (× 10^4) and equivalent isotropic displacement parameters (Å^2 ×10^3) for **4·2CH$_3$CN·2Et$_2$O**. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	*y*	*z*	U_{eq}
K1	0.71016 (11)	0.44706 (8)	0.29867 (9)	47 (3)
Ni1	0.69418 (5)	0.38751 (4)	0.65910 (4)	34 (2)
Ni2	0.56826 (5)	0.47859 (4)	0.57311 (4)	30 (1)
S1	0.57039 (11)	0.43544 (8)	0.70232 (8)	35 (1)
S2	0.80032 (11)	0.44076 (8)	0.74891 (9)	38 (1)
S3	0.58250 (11)	0.35685 (7)	0.55581 (8)	33 (1)
S4	0.81342 (12)	0.34155 (9)	0.60819 (10)	43 (1)

S5	0.60600 (10)	0.50403 (7)	0.45559 (8)	32 1)
S6	0.61603 (11)	0.58272 (7)	0.61768 (8)	34 1)
O1	0.4664 (4)	0.5381 (3)	0.7878 (3)	66 (2)
O2	0.5951 (4)	0.5608 (4)	0.8873 (3)	79 (2)
O3	0.6331 (3)	0.3283 (2)	0.3424 (3)	49 (1)
O4	0.5258 (3)	0.2675 (2)	0.4024 (2)	40 (1)
O5	0.6917 (3)	0.5831 (2)	0.3326 (3)	48 (1)
O6	0.7837 (4)	0.6551 (3)	0.4222 (3)	61 (1)
N1	0.8701 (6)	0.4804 (5)	0.4253 (6)	108 (3)
C1	0.6271 (4)	0.4932 (3)	0.7773 (3)	36 (1)
C2	0.5549 (5)	0.5331 (4)	0.8156 (4)	52 (2)
C3	0.5247 (12)	0.5983 (18)	0.9345 (15)	86 (4)
C3'	0.5331 (19)	0.6283 (17)	0.905 (2)	84 (4)
C4	0.7290 (4)	0.4959 (3)	0.7958 (3)	35 (1)
C5	0.7878 (4)	0.5483 (3)	0.8510 (3)	37 (1)
C6	0.8593 (5)	0.5273 (4)	0.9158 (4)	45 (2)
C7	0.9153 (5)	0.5770 (4)	0.9647 (4)	51 (2)
C8	0.9032 (5)	0.6469 (4)	0.9475 (4)	51 (2)
C9	0.8342 (6)	0.6686 (4)	0.8833 (4)	57 (2)
C10	0.7749 (5)	0.6198 (3)	0.8337 (4)	50 (2)
C11	0.6560 (4)	0.3273 (3)	0.4868 (3)	36 (1)
C12	0.6051 (4)	0.3092 (3)	0.4032 (4)	36 (1)
C13	0.4745 (5)	0.2455 (4)	0.3233 (4)	47 (2)
C14	0.7559 (4)	0.3167 (3)	0.5121 (4)	39 (1)
C15	0.8205 (5)	0.2792 (3)	0.4638 (4)	45 (2)
C16	0.9166 (5)	0.3019 (4)	0.4617 (5)	62 (2)
C17	0.9801 (6)	0.2644 (5)	0.4207 (6)	74 (2)
C18	0.9480 (6)	0.2060 (5)	0.3799 (6)	73 (2)
C19	0.8520 (6)	0.1808 (4)	0.3794 (5)	64 (2)
C20	0.7874 (5)	0.2173 (4)	0.4223 (4)	49 (2)
C21	0.6617 (4)	0.5868 (3)	0.4686 (3)	34 (1)
C22	0.7108 (5)	0.6083 (3)	0.3999 (4)	41 (1)
C23	0.809 (3)	0.6942 (18)	0.3498 (14)	67 (4)
C23'	0.8457 (18)	0.6734 (13)	0.3607 (11)	63 (3)
C24	0.6610 (4)	0.6221 (3)	0.5396 (3)	32 (1)
C25	0.6978 (4)	0.6959 (3)	0.5561 (4)	38 (1)
C26	0.7638 (5)	0.7103 (4)	0.6278 (4)	48 (2)

C27	0.7959 (5)	0.7786 (4)	0.6450 (5)	59 (2)
C28	0.7642 (6)	0.8317 (4)	0.5913 (6)	66 (2)
C29	0.6960 (6)	0.8173 (4)	0.5208 (5)	61 (2)
C30	0.6633 (5)	0.7507 (3)	0.5047 (4)	48 (2)
C31	0.8755 (7)	0.5017 (6)	0.4945 (6)	86 (2)
C32	0.8899 (6)	0.5303 (5)	0.5774 (5)	68 (2)
O7	0.8460 (7)	0.4334 (5)	0.1964 (5)	122 (3)
C33	0.8310 (19)	0.4859 (12)	0.1308 (12)	134 (4)
C34	0.887 (2)	0.5397 (11)	0.1998 (16)	149 (5)
C35	0.9519 (14)	0.4301 (12)	0.2171 (17)	137 (4)
C36	0.935 (2)	0.3450 (12)	0.2207 (19)	149 (5)
C33'	0.9134 (17)	0.4874 (13)	0.183 (2)	132 (4)
C34'	0.809 (2)	0.5308 (14)	0.146 (2)	138 (5)
C35'	0.875 (2)	0.3585 (12)	0.192 (2)	138 (4)
C36'	0.964 (2)	0.3788 (18)	0.2649 (19)	143 (5)

Table S5. Anisotropic displacement parameters (Å^2 x 10^3) for **4·2CH$_3$CN·2Et$_2$O**. The anisotropic displacement factor exponent takes the form: −2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12].

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
K1	0.0463 (8)	0.0514 (9)	0.0440 (8)	−0.0016 (6)	0.0112 (6)	−0.0034 (6)
Ni1	0.0374 (4)	0.0315 (4)	0.0321 (4)	0.0034 (3)	0.0024 (3)	−0.0004 (3)
Ni2	0.0333 (4)	0.0296 (4)	0.0264 (4)	0.0016 (3)	0.0039 (3)	0.0007 (3)
S1	0.0384 (8)	0.0354 (8)	0.0292 (7)	0.0008 (6)	0.0038 (6)	0.0019 (6)
S2	0.0381 (8)	0.0381 (8)	0.0365 (8)	0.0019 (6)	0.0008 (6)	−0.0026 (6)
S3	0.0360 (8)	0.0291 (7)	0.0325 (7)	0.0012 (6)	0.0051 (6)	−0.0010 (6)
S4	0.0370 (8)	0.0446 (9)	0.0467 (9)	0.0075 (7)	0.0030 (7)	−0.0059 (7)
S5	0.0338 (7)	0.0316 (7)	0.0300 (7)	0.0011 (5)	0.0060 (5)	0.0003 (6)
S6	0.0385 (8)	0.0328 (8)	0.0295 (7)	−0.0007 (6)	0.0048 (6)	−0.0020 (6)
O1	0.047 (3)	0.095 (4)	0.054 (3)	0.013 (3)	0.003 (2)	−0.026 (3)
O2	0.055 (3)	0.122 (5)	0.060 (3)	0.004 (3)	0.009 (2)	−0.050 (3)
O3	0.060 (3)	0.052 (3)	0.038 (2)	−0.005 (2)	0.017 (2)	−0.002 (2)
O4	0.039 (2)	0.041 (2)	0.038 (2)	0.0007 (18)	0.0064 (17)	0.0002 (18)
O5	0.060 (3)	0.050 (3)	0.036 (2)	−0.005 (2)	0.017 (2)	0.000 (2)
O6	0.072 (3)	0.057 (3)	0.064 (3)	−0.027 (2)	0.042 (3)	−0.018 (2)
N1	0.093 (5)	0.108 (6)	0.102 (5)	0.012 (5)	−0.039 (5)	−0.017 (5)
C1	0.041 (3)	0.036 (3)	0.029 (3)	0.002 (2)	0.004 (2)	−0.001 (2)

C2	0.057 (5)	0.061 (5)	0.040 (4)	0.003 (3)	0.011 (3)	−0.009 (3)
C3	0.068 (6)	0.131 (9)	0.060 (7)	0.002 (7)	0.014 (5)	−0.052 (6)
C3'	0.060 (7)	0.140 (9)	0.056 (8)	−0.005 (7)	0.023 (6)	−0.054 (7)
C4	0.049 (4)	0.033 (3)	0.022 (3)	0.005 (2)	0.003 (2)	0.004 (2)
C5	0.041 (3)	0.038 (3)	0.034 (3)	0.005 (3)	0.008 (2)	0.000 (3)
C6	0.046 (4)	0.045 (4)	0.040 (3)	0.000 (3)	−0.006 (3)	0.001 (3)
C7	0.039 (4)	0.067 (5)	0.041 (4)	0.001 (3)	−0.005 (3)	−0.010 (3)
C8	0.049 (4)	0.052 (4)	0.049 (4)	0.000 (3)	0.000 (3)	−0.018 (3)
C9	0.071 (5)	0.037 (4)	0.060 (5)	−0.002 (3)	0.005 (4)	−0.007 (3)
C10	0.068 (4)	0.038 (4)	0.041 (4)	0.005 (3)	−0.004 (3)	−0.004 (3)
C11	0.043 (3)	0.029 (3)	0.037 (3)	0.002 (2)	0.008 (3)	0.000 (2)
C12	0.039 (3)	0.031 (3)	0.039 (3)	0.006 (2)	0.010 (3)	−0.001 (2)
C13	0.044 (4)	0.056 (4)	0.036 (3)	−0.003 (3)	−0.002 (3)	−0.007 (3)
C14	0.037 (3)	0.038 (3)	0.040 (3)	0.002 (3)	0.004 (3)	−0.003 (3)
C15	0.039 (4)	0.047 (4)	0.051 (4)	0.004 (3)	0.010 (3)	−0.004 (3)
C16	0.049 (4)	0.066 (5)	0.077 (5)	0.002 (4)	0.022 (4)	−0.013 (4)
C17	0.059 (5)	0.074 (6)	0.096 (7)	0.005 (4)	0.034 (4)	−0.018 (5)
C18	0.057 (5)	0.080 (6)	0.092 (6)	0.020 (4)	0.037 (4)	−0.003 (5)
C19	0.075 (6)	0.051 (5)	0.067 (5)	0.017 (4)	0.019 (4)	−0.008 (4)
C20	0.047 (4)	0.048 (4)	0.052 (4)	0.007 (3)	0.013 (3)	−0.005 (3)
C21	0.030 (3)	0.037 (3)	0.033 (3)	0.000 (2)	0.002 (2)	0.004 (2)
C22	0.047 (4)	0.038 (3)	0.040 (4)	−0.004 (3)	0.017 (3)	−0.001 (3)
C23	0.071 (8)	0.076 (8)	0.069 (6)	−0.014 (7)	0.056 (7)	−0.019 (6)
C23'	0.069 (7)	0.063 (7)	0.071 (6)	−0.014 (6)	0.052 (5)	−0.012 (5)
C24	0.035 (3)	0.033 (3)	0.028 (3)	0.001 (2)	0.005 (2)	−0.002 (2)
C25	0.039 (3)	0.033 (3)	0.043 (3)	−0.005 (2)	0.013 (3)	−0.007 (3)
C26	0.038 (4)	0.056 (4)	0.050 (4)	−0.009 (3)	0.012 (3)	−0.017 (3)
C27	0.047 (4)	0.062 (5)	0.070 (5)	−0.019 (4)	0.021 (4)	−0.031 (4)
C28	0.062 (5)	0.042 (4)	0.105 (7)	−0.018 (4)	0.041 (5)	−0.027 (4)
C29	0.073 (5)	0.037 (4)	0.076 (5)	−0.004 (3)	0.024 (4)	0.001 (4)
C30	0.055 (4)	0.034 (4)	0.056 (4)	−0.006 (3)	0.015 (3)	−0.002 (3)
C31	0.064 (5)	0.096 (6)	0.091 (5)	0.007 (4)	−0.003 (5)	−0.003 (5)
C32	0.054 (4)	0.082 (6)	0.067 (4)	0.011 (4)	0.011 (4)	0.008 (4)
O7	0.122 (5)	0.134 (6)	0.128 (6)	−0.010 (5)	0.070 (5)	−0.021 (5)
C33	0.136 (7)	0.141 (8)	0.139 (8)	−0.008 (7)	0.064 (7)	−0.019 (6)

C34	0.151 (10)	0.150 (9)	0.160 (10)	−0.015 (9)	0.062 (9)	−0.025 (9)
C35	0.132 (7)	0.151 (8)	0.140 (7)	−0.007 (7)	0.057 (7)	−0.017 (7)
C36	0.136 (11)	0.160 (9)	0.152 (11)	0.008 (9)	0.028 (10)	−0.019 (10)
C33'	0.132 (7)	0.140 (8)	0.141 (8)	−0.007 (6)	0.066 (7)	−0.021 (7)
C34'	0.142 (10)	0.144 (10)	0.141 (11)	0.000 (9)	0.066 (10)	−0.013 (10)
C35'	0.137 (7)	0.146 (8)	0.143 (8)	0.000 (7)	0.058 (7)	−0.022 (7)
C36'	0.141 (10)	0.143 (11)	0.153 (11)	0.000 (9)	0.049 (8)	−0.027 (9)