Electronic Supplementary Information

Electrocatalytic hydrogen production by dinuclear cobalt(II) compounds containing redox-active diamidate ligands: A combined experimental and theoretical study

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Table of Contents

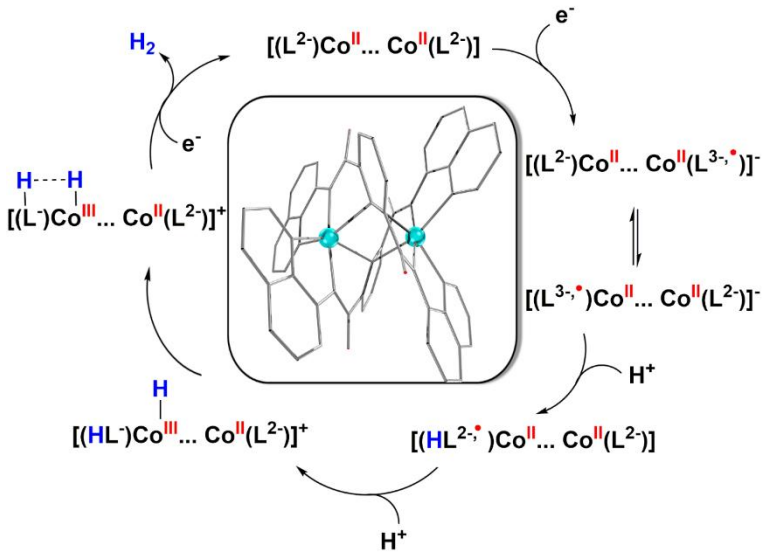

Fig. S1 Electrocatalytic hydrogen evolution proposed cycle induced by the Co-based dimeric complexes.

NMR spectroscopy

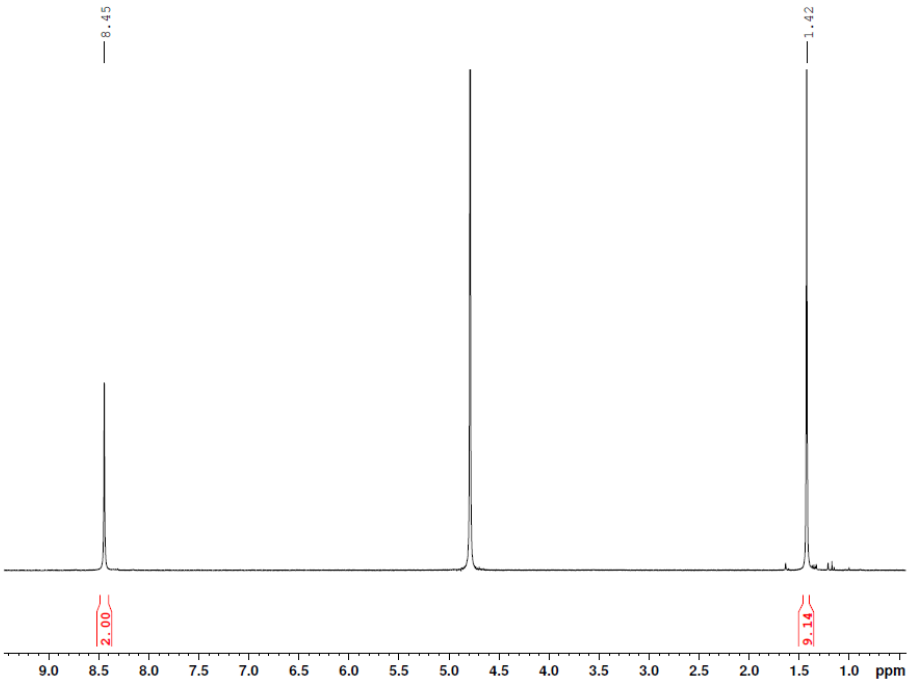

Fig. S2 ^{1}H NMR spectrum of 4-*tert*-butylpyridine-2,6-dicarboxylic acid in D_2O

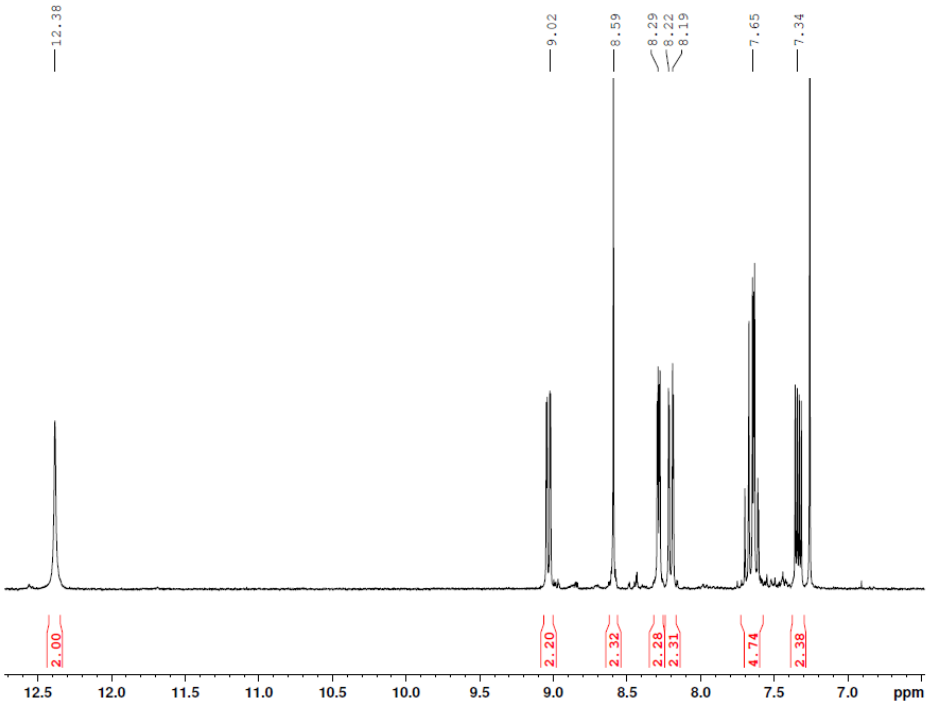

Fig. S3 ^{1}H NMR spectrum of H_2L^{Bu} in $CDCl_3$

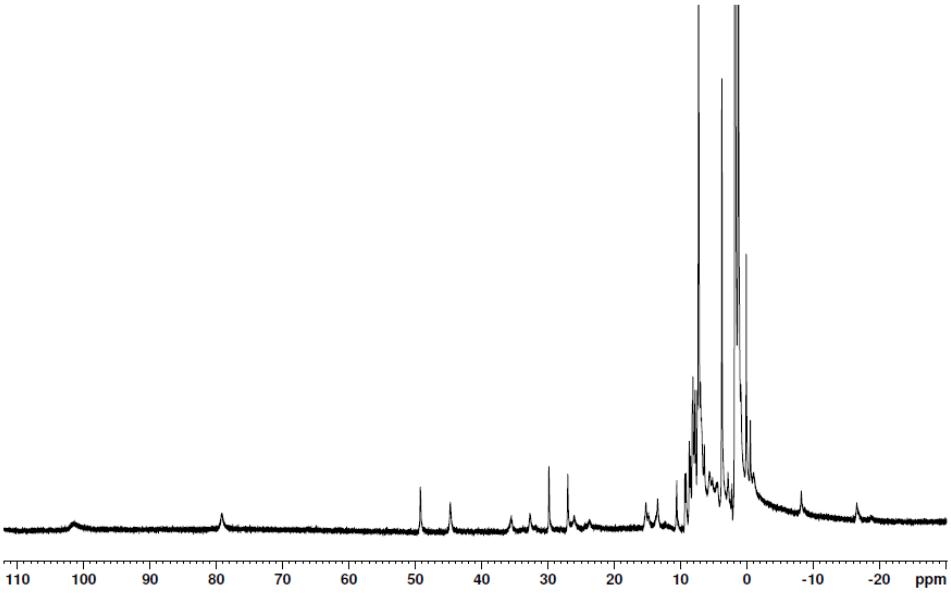

Fig. S4 ^{1}H NMR spectrum of **1** in $CDCl_3$

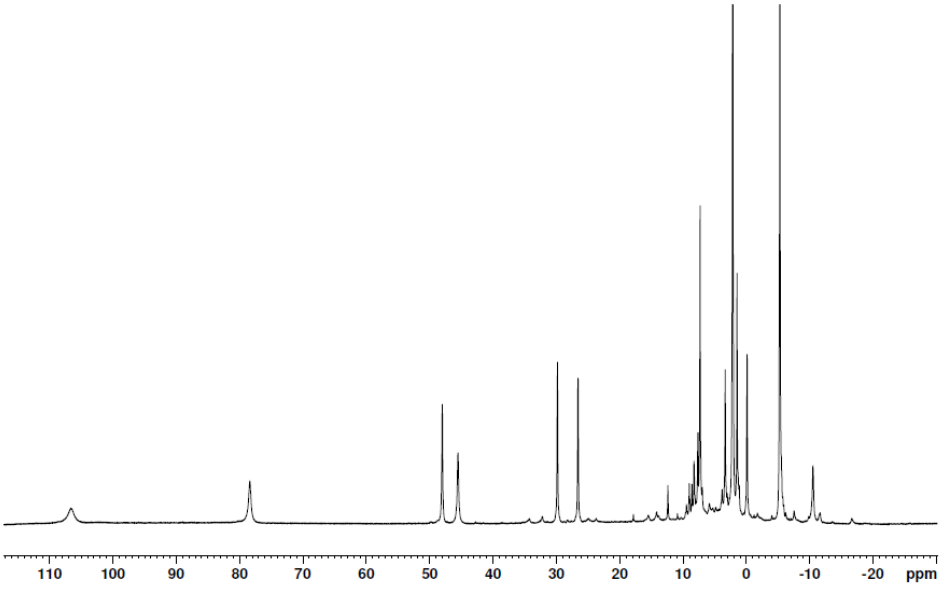

Fig. S5 ^{1}H NMR spectrum of **2** in $CDCl_3$

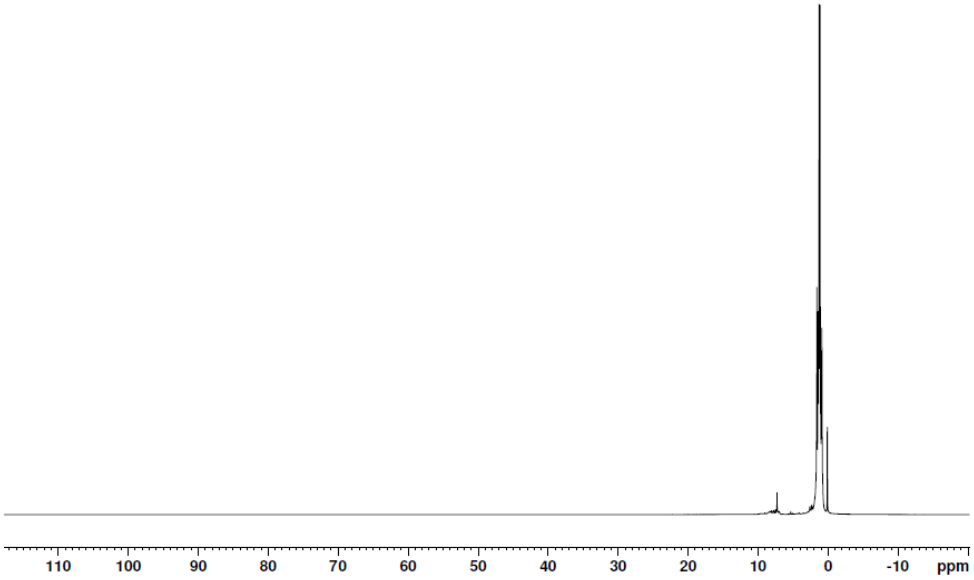

Fig. S6 ^{1}H NMR spectrum of **3** in $CDCl_3$. Compound **3** shows no paramagnetic shift.

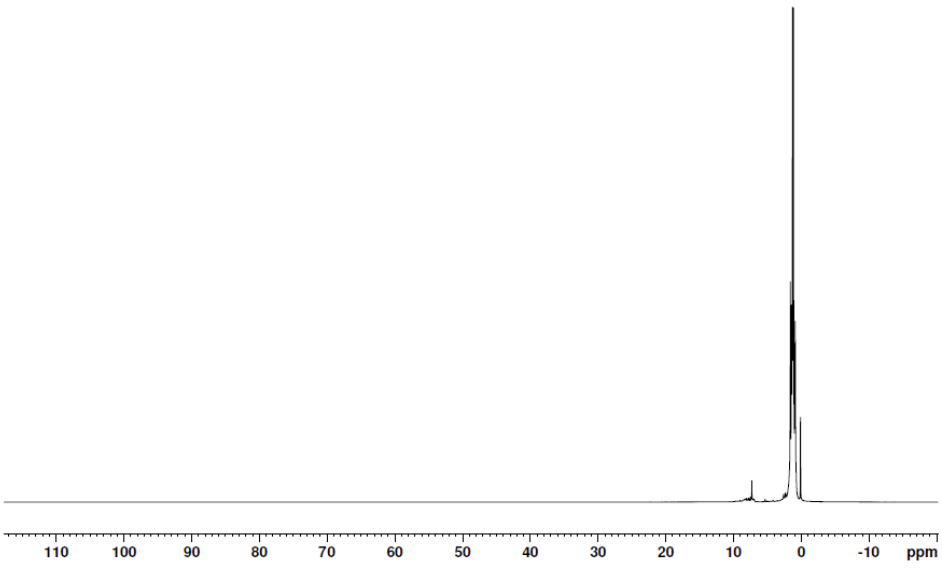

Fig. S7 ^{1}H NMR spectrum of **4** in $CDCl_3$. Compound **4** shows no paramagnetic shift.

Mass spectrometry

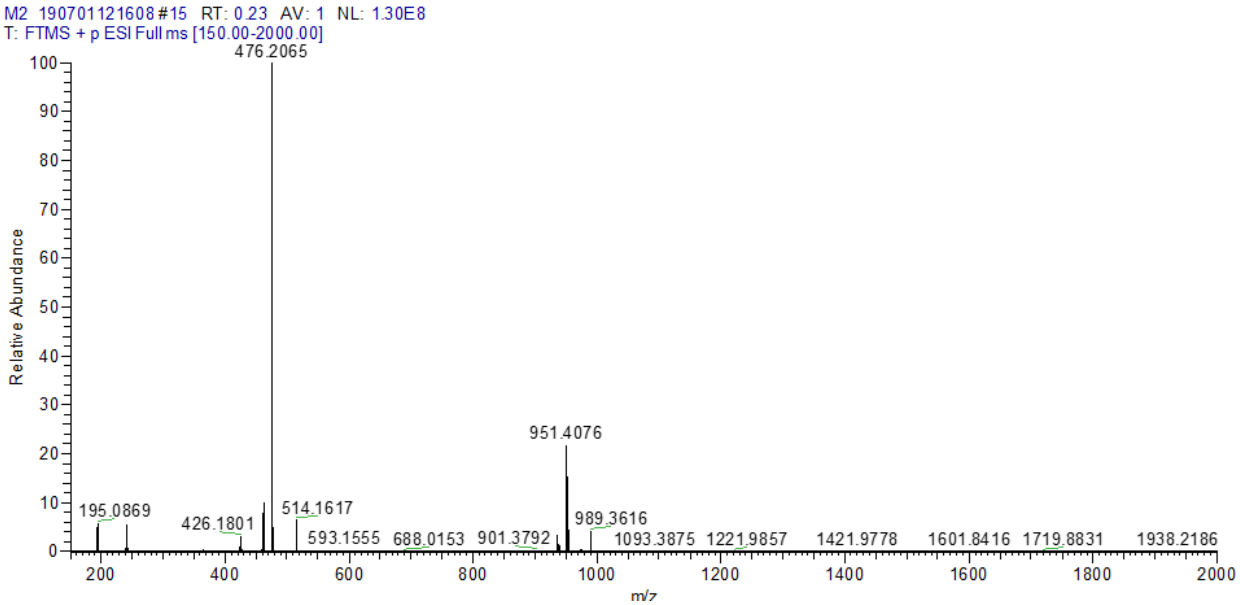

Fig. S8 Mass spectrum of H_2L^{Bu} in CH_3OH containing a drop of CH_3COOH

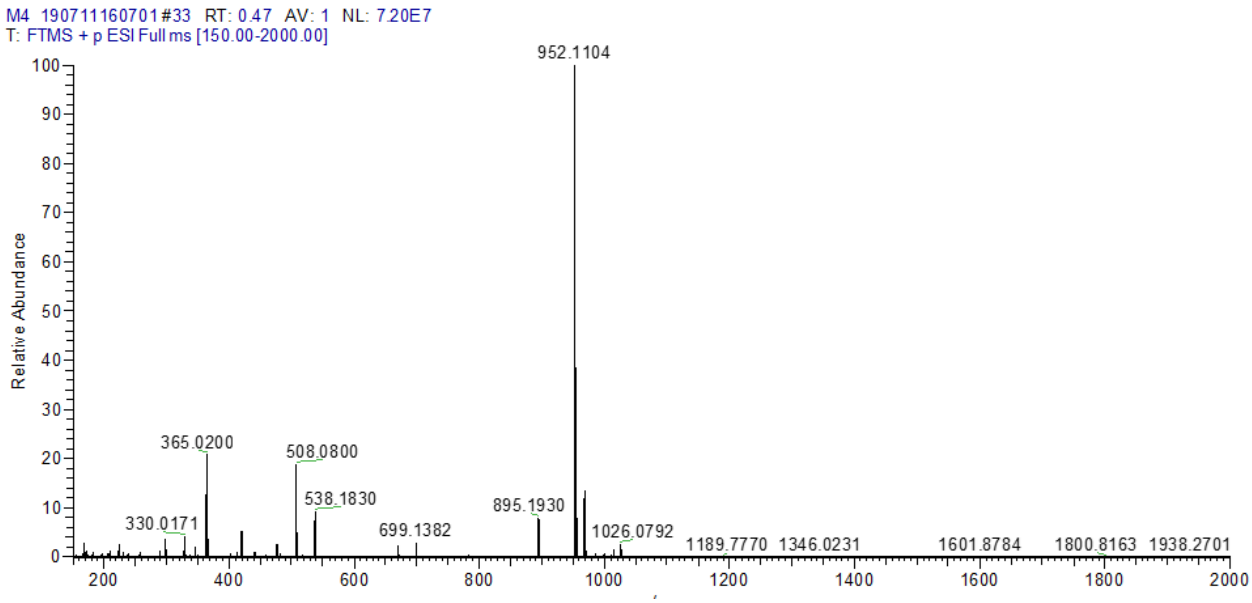

Fig. S9 Mass Spectrum of **1** in CH_3OH

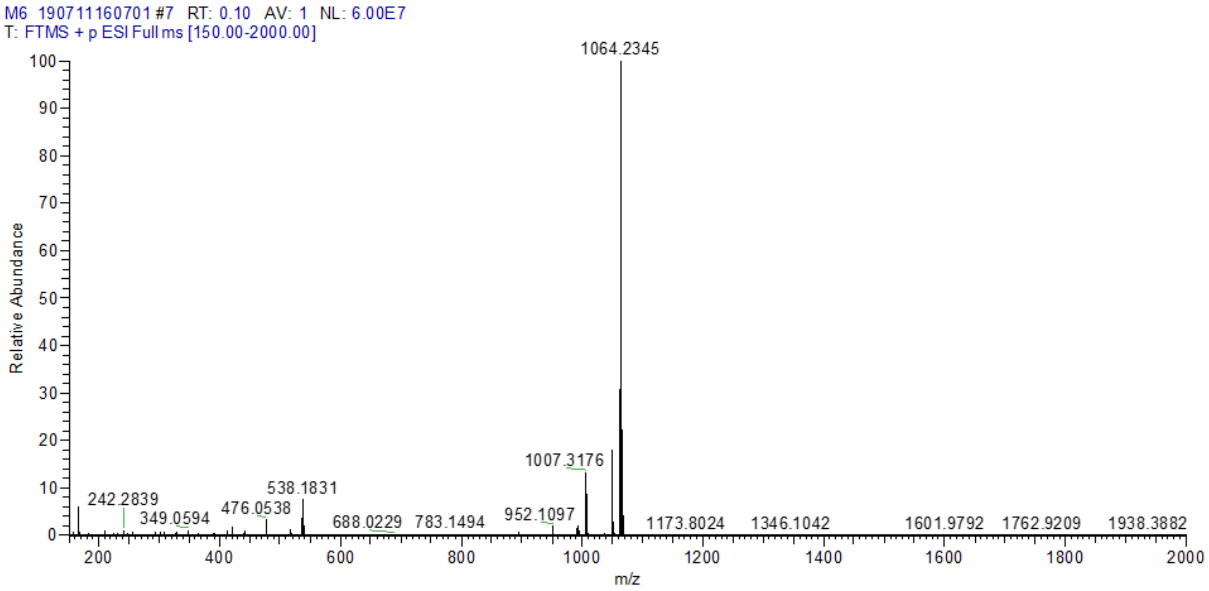

Fig. S10 Mass spectrum of **2** in CH_3OH

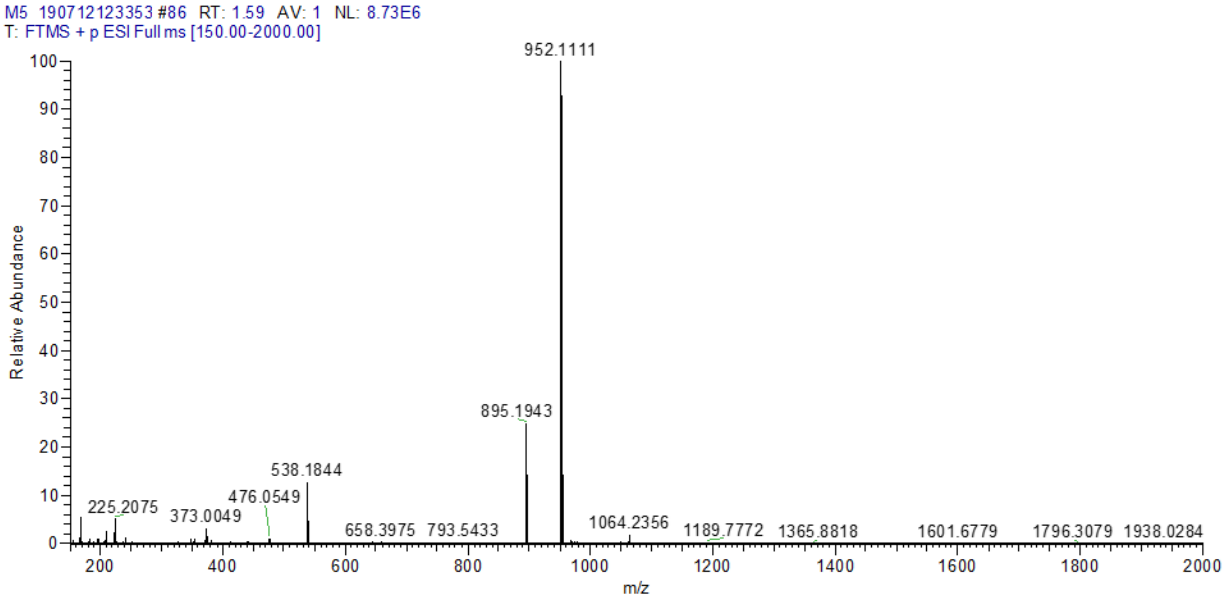

Fig. S11 Mass spectrum of **3** in CH_3OH

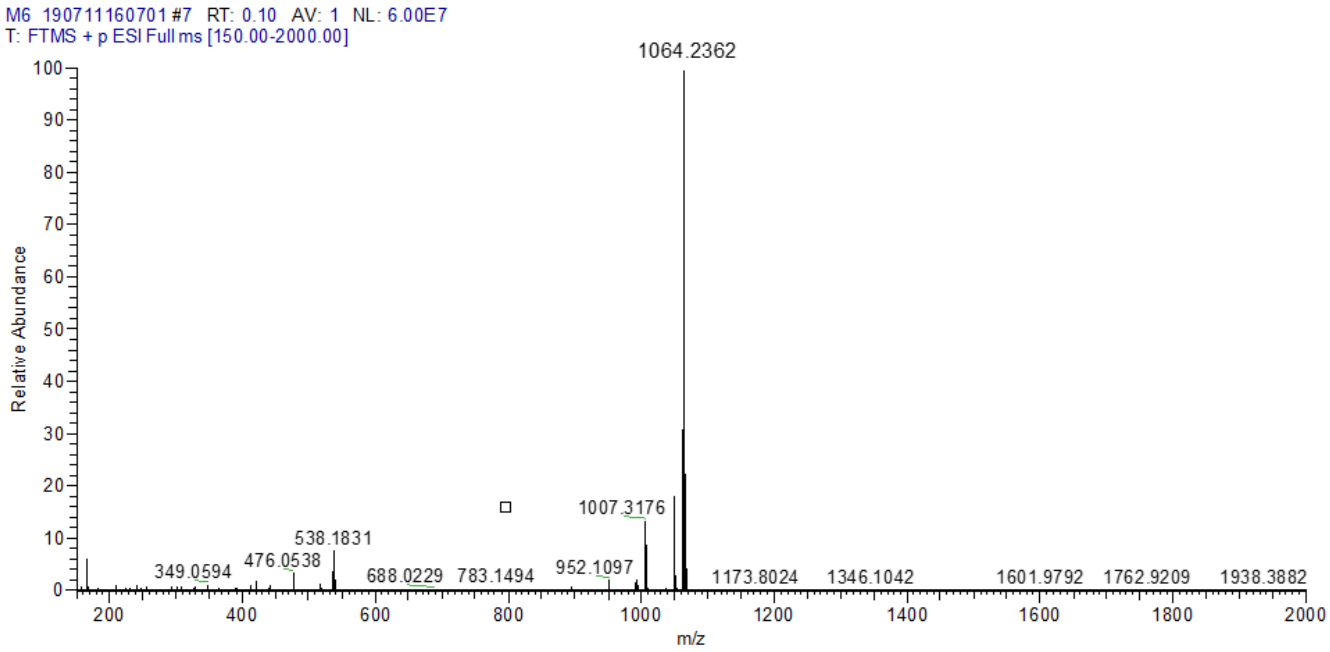

Fig. S12 Mass spectrum of **4** in CH_3OH

Electronic spectroscopy

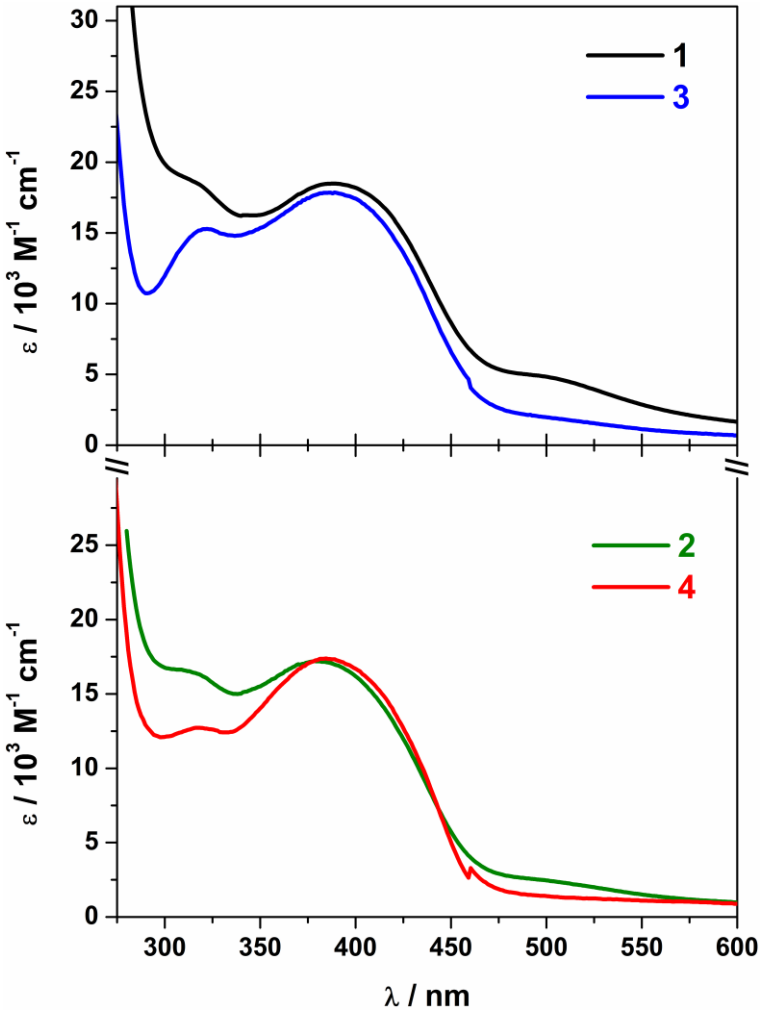

Fig. S13 Overlay of the electronic spectra of **1** and **3** (top) and **2** and **4** (bottom) recorded in DMF solution at ambient temperature.

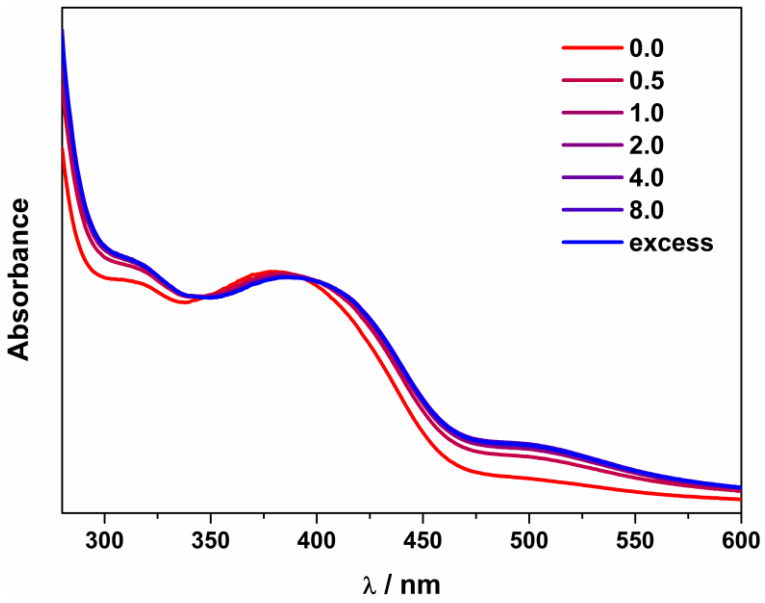

Fig. S14 Overlay of electronic spectra of **2** in DMF with sequential addition of AcOH.

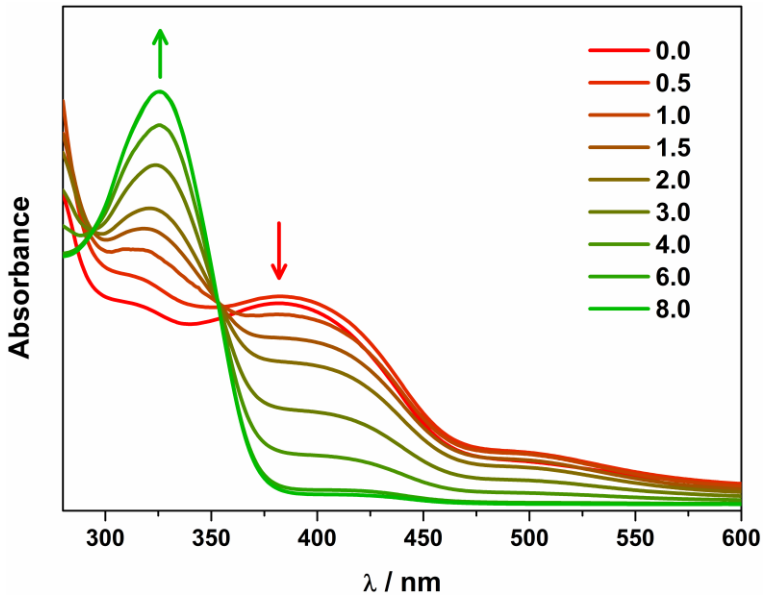

Fig. S15 Overlay of electronic spectra of **2** in DMF with sequential addition of TsOH.

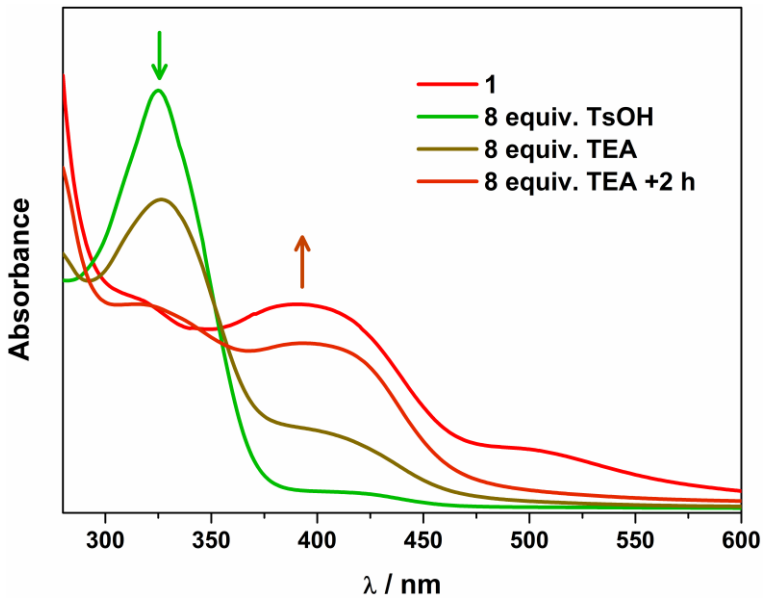

Fig. S16 Overlay of electronic spectrum of the dicobalt **1** in DMF, formation of the monocobalt species upon treatment with 8 equiv. of *p*-toluenesulfonic acid (TsOH), and regeneration of **1** with 8 equiv. of triethylamine (TEA), and the spectrum 2 h after neutralization with TEA.

Crystallography

X-Ray Crystallographic Data Collection and Refinement of 1 and 4. Suitable single crystal was selected and mounted onto a rubber loop using Fomblin oil. Single-crystal X-ray diffraction data of **1**–**4** were recorded on a Bruker Apex CCD diffractometer (λ (MoK_{α}) = 0.71073 Å) at 150 K equipped with a graphite monochromator. Structure solution and refinement were carried out with SHELXS-97[1] and SHELXL-97[2] using the WinGX software package.[3] Data collection and reduction were performed using the Apex2 software package. Corrections for incident and diffracted beam absorption effects were applied using empirical absorption corrections.[4] All the non-H atoms were refined anisotropically. The positions of hydrogen atoms were calculated based on stereochemical considerations using the riding model. Final unit cell data and refinement statistics for **1**–**4** are collated in Table S1. The crystallographic data for **1**–**4** can be obtained free of charge from the Cambridge Crystallographic Data Centre, 12, Union Road, Cambridge CB2 1EZ; fax:(+44) 1223-336-033, deposit@ccdc.cam.ac.uk.

Table S1 Salient crystallographic bond lengths (Å) in **1**–**4**

	1	**2**	**3**	**4**
Co(1)–N(1)	2.430(1)	2.266(3)	-	-
Co(1)–N(2)	2.010(9)	2.029(3)	1.977(2)	1.982(5)
Co(1)–N(3)	2.126(1)	2.150(3)	2.009(3)	2.016(5)
Co(1)–N(4)	2.021(8)	2.038(3)	1.975(2)	1.979(4)
Co(1)–N(5)	2.110(8)	2.101(3)	2.001(3)	2.024(5)
Co(1)–N(6)	2.402(8)	-	-	-
Co(2)–N(1)	2.402(8)	-	1.975(2)	1.989(5)
Co(2)–N(6)	2.430(1)	2.301(3)	1.981(2)	1.991(5)
Co(2)–N(7)	2.010(9)	1.994(3)	1.890(2)	1.895(4)
Co(2)–N(8)	2.126(1)	2.137(3)	1.941(2)	1.953(6)
Co(2)–N(9)	2.021(8)	2.025(3)	1.882(2)	1.907(4)
Co(2)–N(10)	2.110(8)	2.088(4)	1.940(2)	1.950(5)

Table S2 Salient crystallographic bond angles (°) in **1**–**4**

	1	**2**	**3**	**4**
N(1)–Co(1)–N(2)	74.5(3)	75.2(1)	-	-
N(1)–Co(1)–N(3)	152.5(3)	151.5(1)	-	-
N(1)–Co(1)–N(4)	96.8(3)	117.1(1)	-	-
N(1)–Co(1)–N(5)	81.8(3)	85.5(1)	-	-
N(1)–Co(1)–N(6)	97.2(3)	-	-	-
N(2)–Co(1)–N(3)	79.2(3)	77.8(1)	83.10(9)	89.9(2)
N(2)–Co(1)–N(4)	171.2(4)	157.3(1)	133.01(9)	131.6(2)
N(2)–Co(1)–N(5)	100.5(4)	121.9(1)	120.14(9)	122.9(2)
N(2)–Co(1)–N(6)	103.8(3)	-	-	-
N(3)–Co(1)–N(4)	109.3(4)	91.4(1)	123.71(9)	120.1(2)
N(3)–Co(1)–N(5)	111.0(3)	101.2(1)	117.67(9)	121.9(2)
N(3)–Co(1)–N(6)	81.5(3)	-	-	-
N(4)–Co(1)–N(5)	79.1(4)	79.5(1)	83.50(9)	82.8(2)
N(4)–Co(1)–N(6)	75.6(3)	-	-	-
N(5)–Co(1)–N(6)	154.4(3)	-	-	-
N(6)–Co(2)–N(7)	74.5(3)	76.4(1)	82.79(9)	82.2(1)
N(6)–Co(2)–N(8)	152.5(3)	154.2(1)	166.4(1)	164.9(2)
N(6)–Co(2)–N(9)	96.8(3)	111.4(1)	96.64(9)	100.8(2)
N(6)–Co(2)–N(10)	81.8(3)	87.3(1)	88.62(9)	86.9(2)
N(6)–Co(2)–N(1)	97.2(3)	-	98.29(9)	99.7(2)
N(7)–Co(2)–N(8)	79.2(3)	78.9(1)	83.6(1)	83.6(2)
N(7)–Co(2)–N(9)	171.2(4)	157.0(1)	175.7(1)	176.1(2)
N(7)–Co(2)–N(10)	100.5(4)	123.3(1)	92.05(9)	94.3(2)
N(7)–Co(2)–N(1)	103.8(3)	-	102.06(9)	99.2(2)
N(8)–Co(2)–N(9)	109.3(4)	94.4(1)	96.8(1)	93.1(2)
N(8)–Co(2)–N(10)	111.0(3)	100.2(1)	90.3(1)	88.8(2)
N(8)–Co(2)–N(1)	81.5(3)	-	86.04(9)	87.8(2)
N(9)–Co(2)–N(10)	79.1(4)	79.4(1)	83.6(1)	83.5(2)
N(9)–Co(2)–N(1)	75.6(3)	-	82.27(9)	82.8(2)
N(10)–Co(2)–N(1)	154.4(3)	-	164.93(9)	165.7(2)

Electrochemistry

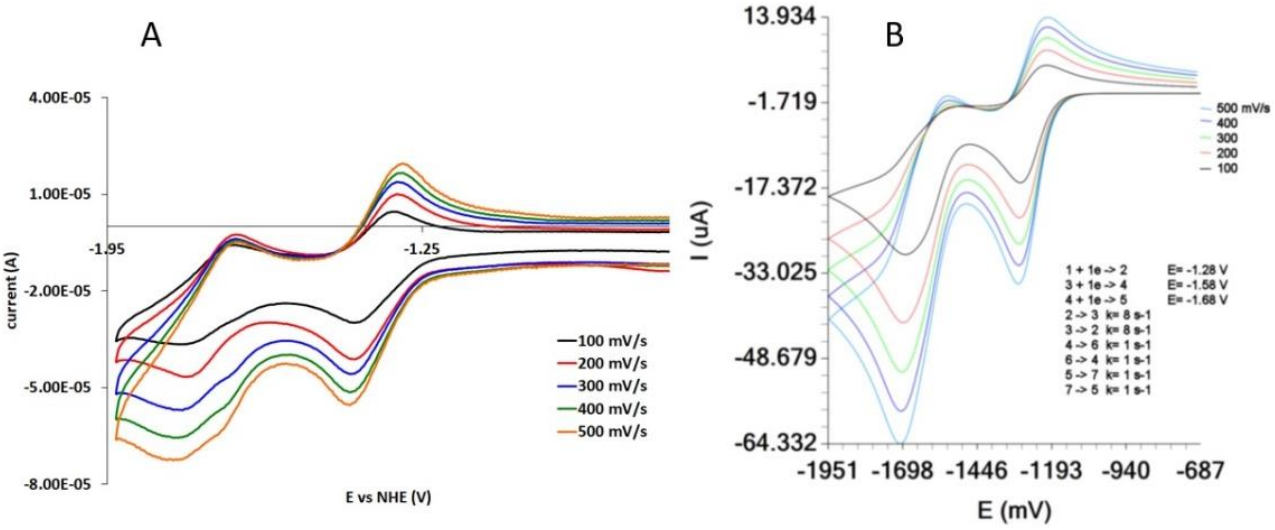

Fig. S17 (a) Overlay of voltammograms of **1** in 95:5 (v/v) DMF/H_2O solution containing 0.1 M [N(*n*-Bu)$_4$]ClO_4 as supporting electrolyte at multiple scan rates, 100–500 mV s^{-1}. At scan rates above 100 mV s^{-1}, the current ratio (i_c/i_a) increases for the wave at –1.28 V and the cathodic peak at –1.72 V broadens and splits into two peaks. This behavior suggests that the cathodic peak at –1.31 V is associated with the cathodic peak at –1.72 V. (b) The corresponding simulations[5] of the reduction processes at incremental scan rates 100–500 mV s^{-1} of **1**. Simulation parameters are shown in the figure: Numbers 1–7 were used to label the various electrochemical and chemical species. The overall electrochemical process traverses the following sequence showing entities 1–4 given in parenthesis:

1. $[Co^{II}{}_2(\mu\text{-}L^{2-})_2]^0$ (1) + $e^- \rightarrow [Co^{II}Co^{I}(\mu\text{-}L^{2-})_2]^{1-}$ (2)
2. $[Co^{II}Co^{I}(\mu\text{-}L^{2-})_2]^{1-}$ (2) $\rightarrow [Co^{II}{}_2(\mu\text{-}L^{3-\bullet})(\mu\text{-}L^{2-})]^{1-}$ (3)
3. $[Co^{II}{}_2(\mu\text{-}L^{3-\bullet})(\mu\text{-}L^{2-})]^{1-}$ (3) + $e^- \rightarrow [Co^{II}Co^{I}(\mu\text{-}L^{3-\bullet})(\mu\text{-}L^{2-})]^{2-}$ (4)

The first and third processes in the above sequence represent outer sphere electron transfer (*i.e.* one-electron reduction) whereas the second process describes an intramolecular electron transfer defined as moving an electron from the metal ion to the ligand. The former are quantified by the reduction potential, *E* (in volts) taken from the experimental voltammogram, and the latter are defined by the electron transfer rate constant, *k* (per second).

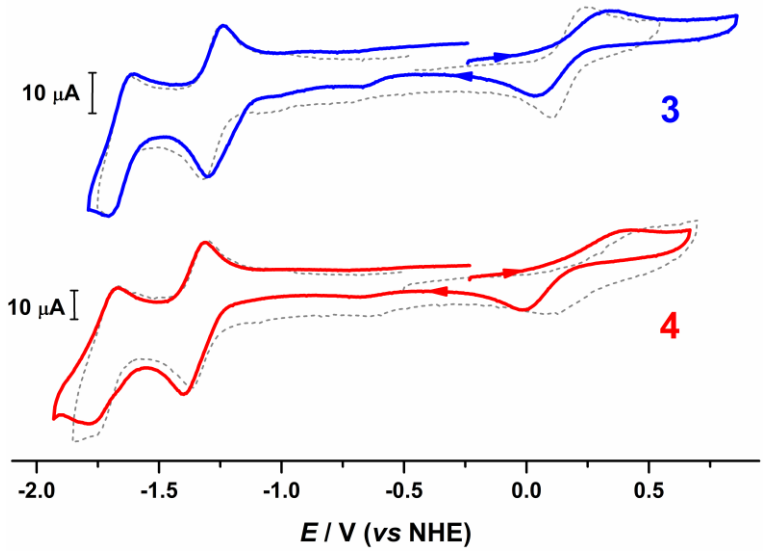

Fig. S18 Overlay of the cyclic voltammogram of **3** (blue line) with **1** (dashed line), and **4** (red line) with **2** (dashed line) recorded in 95:5 (v/v) DMF/H_2O solutions containing 0.1 M [N(*n*-Bu)$_4$]ClO_4 as supporting electrolyte at scan rate of 100 mV s^{-1}.

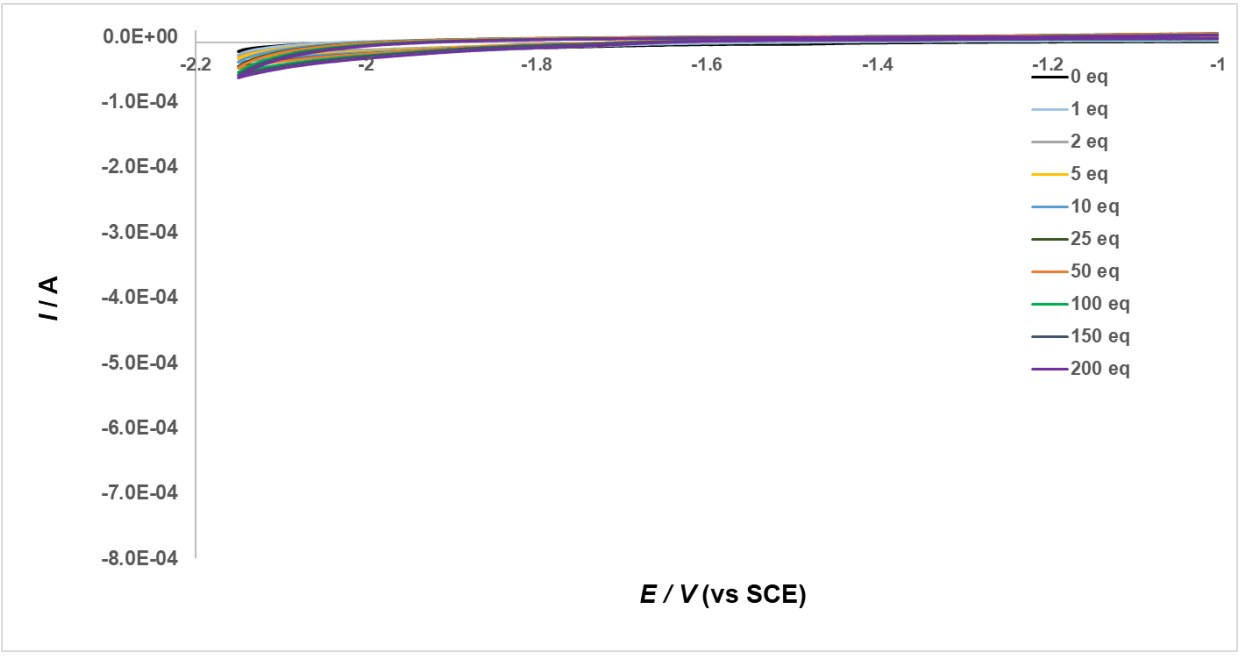

Fig. S19 Cyclic voltammograms in the presence of acid (and absence of catalyst) recorded in 95:5 (v/v) DMF/H_2O solutions containing 0.1 M [N(*n*-Bu)$_4$]ClO_4 as supporting electrolyte at scan rate of 100 mV s^{-1}.

Theoretical calculations

The program package ORCA was used for all calculations.[6] With the exception of neutral **1**, the input geometry for all molecules were generated using ArgusLab from crystallographic coordinates of **1**.[7] The geometries of all molecules were fully optimized by a spin-unrestricted DFT method employing the BP86 functional.[8] Split-valence basis sets with one set of polarization functions (def2-SVP) were used for all atoms.[9] A scalar relativistic correction was applied using the zeroth-order regular approximation (ZORA) method.[10] The RI approximation combined with the appropriate Ahlrichs auxiliary basis set was used to speed up the calculations.[11] The conductor like screening model (COSMO) was used for all calculations.[12] The self-consistent field calculations were tightly converged (1×10^{-8} E_h in energy, 1×10^{-7} E_h in the density charge, and 1×10^{-7} in the maximum element of the DIIS[13] error vector). The geometry was converged with the following convergence criteria: change in energy $<10^{-5}$ E_h, average force $<5 \times 10^{-4}$ E_h $Bohr^{-1}$, and the maximum force 10^{-4} E_h $Bohr^{-1}$. The geometry search for all complexes was carried out in redundant internal coordinates without imposing geometry constraints. Where feasible the stability of all solutions was checked by performing frequency calculations: no negative frequencies were observed. Some numerical frequency calculations failed to compute based on the size and flexibility of these molecules at this level of theory. Single point calculations were performed on optimized coordinates using the B3LYP functional[14] and triple-ζ-quality basis sets with two sets of polarization functions (def2-TZVPP) for all atoms.[15] The RIJCOSX algorithm was used to speed the calculation of Hartree–Fock exchange. The broken symmetry (BS) approach was used to estimate the exchange coupling constants in these compounds.[16] We adopt the following notation: the given system was divided into two fragments. The notation BS(*m*,*n*) refers then to a broken symmetry state with *m* unpaired α-spin electrons essentially on fragment 1 and *n* unpaired β-spin electrons localized on fragment 2. In most cases, fragments 1 and 2 correspond to the metal and the ligands, respectively. In this notation the standard high-spin, open-shell solution is written as BS(*m* + *n*,0). The BS(*m*,*n*) notation refers to the initial guess to the wavefunction. The variational process

does, however, have the freedom to converge to a solution of the form BS(m – n,0) in which effectively the nβ-spin electrons pair up with $n < m$α-spin electrons on the partner fragment. Such a solution is then a standard $M_S \approx (m - n)/2$ spin-unrestricted Kohn-Sham solution. The exchange coupling constant, J, was calculated on the broken-symmetry solution using eq. S1,[17] and assuming the spin-Hamiltonian eq. S2 is valid.

$$J = \frac{E_{HS} - E_{BS}}{\langle \hat{S}^2 \rangle_{HS} - \langle \hat{S}^2 \rangle_{BS}} \quad \text{(S1)}$$

$$\hat{H} = -2J\hat{S}_A \cdot \hat{S}_B \quad \text{(S2)}$$

As explained elsewhere,[18] the nature of the solution is investigated from the corresponding orbital transformation (COT) which, from the corresponding orbital overlaps, displays whether the system should be described as a spin-coupled or a closed-shell solution. Corresponding orbitals and density plots were obtained using *Molekel*.[19]

Table S3 Hydrogen atom optimized coordinates of **1** $M_S = 7$

Co	-0.00015209887595	0.00014813107817	0.00001213847443
Co	0.00017868633890	0.00010296699600	-3.18341784406734
N	-1.74836323200291	0.49420969128104	-1.61314653547326
N	-1.03071378752830	-1.71152234510329	-0.21815066372174
N	1.21960955724582	-1.34507927843424	1.10607797413722
N	1.60699259733536	0.49203077071203	-4.46002786735966
N	-0.10093811745450	2.01897349119598	-3.16479699171359
N	1.80425052995576	-0.21351873482775	-1.57024734480535
N	0.29612099973728	-1.97583468920530	-2.96527905716963
N	-1.64197836114625	-0.77497854824423	-4.28949748844130
N	-1.29577203762143	1.07026953969866	1.27663233670817
N	0.86691447764995	1.82613566334610	-0.01858367528463
O	-3.13073987605814	-2.63246331726728	-0.66333581190826
O	-1.25556451732174	3.87848750083478	-2.38411334311435
O	1.88289054449102	-3.63124240502538	-2.52011068588075
O	2.64600884404244	3.10122005951067	-0.79924690797187
C	2.33899406617870	-1.10212311439648	1.74238084559069
H	2.64645290500594	-0.04433619056627	1.80831939627247
C	3.13523474635887	-2.12045737508863	2.28865802184089
H	4.07588380829770	-1.85505608865207	2.79191016535679
C	2.71561166631205	-3.40613769990870	2.19051262317910
H	3.32099058606560	-4.22231303268541	2.61833853336368
C	1.53245764162416	-3.72222380688307	1.54755420116902
C	0.79383756334795	-2.64587973641245	0.99716218762926
C	-0.41986890561563	-2.87549677554185	0.28386932815929
C	-0.86104676534203	-4.19365777250296	0.13501218841670
H	-1.79546109985606	-4.38708105805217	-0.40073164599385
C	-0.11118076046020	-5.23954135289350	0.72504491980890
H	-0.49337201165561	-6.26658407331602	0.60345414636609
C	1.02654861051414	-5.03566446888328	1.40990022619602
H	1.59284641311611	-5.87027805645667	1.85069129800618
C	-2.30146506384357	-1.71723576677204	-0.68723262832346
C	-2.71248682868065	-0.37916220881482	-1.21499432359418
C	-4.08202911914488	-0.03956829821519	-1.18474144649344
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C	-4.44055544354908	1.23354709020399	-1.57407676685025
H	-5.49767046991971	1.54450303571919	-1.55719044585828
C	-3.45759879172943	2.13450852814337	-2.00084741067841
H	-3.67946037642155	3.16474290500154	-2.30948114858600
C	-2.13594921255086	1.71725509012618	-2.00689416169633
C	-1.08349176103799	2.65886490011827	-2.53669816204436
C	0.94877814007504	2.68455200859612	-3.78609736233794
C	1.20118275030721	4.04441121013832	-3.79710042160353
H	0.50767043437589	4.69556602774188	-3.25291826677323
C	2.31669342086027	4.57290892083210	-4.46422200432194
H	2.48112589589854	5.66147834139901	-4.43763446495650
C	3.18613092849110	3.76752719345891	-5.13357022040751
H	4.06463431823624	4.18447645006907	-5.65172020028996
C	2.99535967337911	2.35723736030919	-5.15607006101664
C	1.85012754551562	1.83329994237153	-4.47483163181617
C	3.85179352420309	1.45771615081328	-5.79238828930503
H	4.74639865591629	1.84302309067494	-6.30813265270763
C	3.56727856845192	0.12468708409322	-5.77731896401406
H	4.21478454768486	-0.61047777688918	-6.27675238725429
C	2.42347735366096	-0.31778041795667	-5.10295154231712
H	2.16950840046588	-1.39094381055899	-5.06709459060085
C	-2.58280059308363	-0.12158519364183	-4.92579451800949
H	-2.46023547696628	0.97331216593126	-4.99438850749456
C	-3.70849546880257	-0.75703006553689	-5.47208246410569
H	-4.47569701521824	-0.15268012727430	-5.97782373902891
C	-3.81360908922993	-2.10536840691744	-5.37395422537045
H	-4.68588749138505	-2.62721100862869	-5.80244655859673
C	-2.84191776100271	-2.85074175564055	-4.73100271210870
C	-1.74720314282365	-2.13962673037044	-4.18059905846104
C	-0.71414897942932	-2.81684924045884	-3.46731210433057
C	-0.81180799104237	-4.20344687691587	-3.31847257546831
H	-0.02345192122432	-4.73829922060884	-2.77860983477221
C	-1.90523840233141	-4.88211251421206	-3.90851653831091
H	-1.94640824101801	-5.97737914715976	-3.78557911423838
C	-2.87796564465988	-4.25778970611447	-4.59336645378803
H	-3.72088588010584	-4.81149277514285	-5.03495999828904
C	1.46766101167989	-2.46809101292925	-2.49620033714880
C	2.36007891812983	-1.38969579558888	-1.96842290486880
C	3.75520153649716	-1.60085977049568	-1.99867494413825
H	4.11707948546634	-2.58356309783009	-2.32874440404432
C	4.57424373358670	-0.56234633646628	-1.60932456862703

H	5.67006252850606	-0.68038663513650	-1.62618615756566
C	4.01160243727572	0.64652114541192	-1.18254018519477
H	4.61127908373968	1.51415735390611	-0.87574916238586
C	2.63094211172918	0.76761474672457	-1.17649536406623
C	2.01969029119987	2.04065381838804	-0.64666694001533
C	0.15240249770373	2.84317629549531	0.60272765394346
C	0.44040478243394	4.19594572338745	0.61374837895581
H	1.33098013783941	4.53120573654274	0.07033815658256
C	-0.38742221591562	5.11156730369539	1.28088936216156
H	-0.12190202926661	6.17978601996773	1.25471253155924
C	-1.49912281985206	4.70084961036058	1.95021966228083
H	-2.15068034993635	5.42221412439633	2.46891820583010
C	-1.86337864334275	3.32512085400285	1.97270136007362
C	-1.00635019067696	2.40231901135564	1.29145350874748
C	-2.99913606012379	2.82246276919138	2.60901046039272
H	-3.67797934571120	3.52052168752180	3.12531492715117
C	-3.24718790145684	1.48217318506377	2.59392372877471
H	-4.12740761898328	1.05064797669964	3.09237788429579
C	-2.36026995332105	0.63516138186588	1.91954790419562
H	-2.53664839598120	-0.45347805517037	1.88257593136796

Table S4 Geometry optimized coordinates of **1** $M_S = 7$

Co	0.13991796931363	0.10424766342127	0.10655861212545
Co	2.12017683645541	0.64043712323305	1.96728233752734
C	1.69342778745937	0.98578516122531	5.00220605369427
H	2.04434784889165	-0.05548516865465	5.07738780743931
C	1.08809289804919	1.64169669348962	6.10189685679421
H	0.96710167934317	1.10363915521586	7.05317160438555
C	0.65390656323828	2.95271046493565	5.95241045195033
H	0.17424362075009	3.48676474965382	6.78765592622003
C	0.82888079521784	3.62289377637127	4.70805306282997
C	1.43767142110332	2.87356861713874	3.64643594048612
C	1.62471716589496	3.45360138828504	2.33876980713323
C	1.26142633664398	4.79840097096974	2.15281398717736
H	1.43658285042227	5.25113980470919	1.17061196569153
C	0.66358561135943	5.52956658622971	3.20740195869044
H	0.36299837926112	6.57162455620145	3.01874591436518
C	0.43287867881990	4.96751004240490	4.45853984819499
H	-0.04576213340884	5.54588595760431	5.26307287928440
C	2.49853867717623	2.97066480984651	0.13922101972315
C	2.88414662254003	1.78831191849638	-0.71329635571584
C	3.76401851305582	1.96932022175505	-1.79120094475083
H	4.04730228883383	2.99939076867922	-2.04758522146400
C	4.28371160409814	0.83961141935768	-2.43789684364595
H	5.01771772930450	0.94814572571728	-3.25116990624949
C	3.85182659829696	-0.42996538989804	-2.03007430179859
H	4.20950860175391	-1.36545616867581	-2.48105925488128
C	2.86211060180125	-0.52275292725943	-1.03980660870018
C	2.22064939001384	-1.85640982550952	-0.75252966643342
C	0.21452456456875	-2.76784229482540	0.23615640846332
C	0.62235007148083	-4.10423075635779	0.39100724301800
H	1.63273987042665	-4.37700129670217	0.06810586239221
C	-0.25056614604522	-5.05449999081595	0.97371179911914
H	0.11166729332456	-6.08500356389978	1.11140530844572
C	-1.53552459772640	-4.72101202363690	1.38906406849680
H	-2.19604592254684	-5.46803417227846	1.85383054220958
C	-2.00991840908093	-3.39111226037688	1.20535994436735
C	-1.13341997814186	-2.42022332556932	0.61546094430882
C	-3.30966340969189	-2.94456797124988	1.58022692161899
H	-4.00705587160609	-3.65559300763404	2.05050603206488
C	-3.68443994409353	-1.62691145053004	1.34832931582807
H	-4.68179688615165	-1.25794253587813	1.62637881887190
C	-2.75647225682506	-0.74183756960478	0.74626988155869
H	-3.01209733535095	0.31195937777741	0.55185633634803
N	2.41192694999038	0.56514582257927	-0.36794915832960
N	2.11232545906786	2.57453730297819	1.38888811730502
N	1.84785318280319	1.57777104966399	3.81850307795127
N	-1.52562517668935	-1.12391145566609	0.40568527988120
N	0.97680415128171	-1.70161703374241	-0.20639482395050
O	2.61566124260814	4.12203606945986	-0.31175914631175
O	2.80653466223729	-2.90478451030107	-1.06844193021298
C	0.47385698105990	0.26662248460732	-2.94617043889681
H	0.58485813519016	-0.82866535576563	-2.89901807984922
C	0.78298073600943	0.99084005685892	-4.12353402993041
H	1.13989910416879	0.45170077734942	-5.01273257898034

C	0.63351449575195	2.37217234437799	-4.12706936090658
H	0.87333115003473	2.96285110576825	-5.02524437255507
C	0.16398895068300	3.04074385829774	-2.96029938302621
C	-0.10853998499627	2.22688085522761	-1.81045155647143
C	-0.55161284965464	2.81417200685028	-0.56955842663553
C	-0.78628896210555	4.19975397321394	-0.54066112271713
H	-1.16086073929534	4.64198961692319	0.38930225718232
C	-0.51766442120222	4.99395901488140	-1.68153032293000
H	-0.68387629872475	6.08036863929778	-1.61532467481743
C	-0.03883038183809	4.44692659206440	-2.86759722085254
H	0.18215622452349	5.08083590445737	-3.73945223781675
C	-1.20305468212125	2.25543152951033	1.68711982075829
C	-1.09327541510856	1.12149388221592	2.67449799354925
C	-2.00316064569537	1.04158150458383	3.74017046173863
H	-2.69752618369671	1.88198952171716	3.88068635203624
C	-2.02579386890007	-0.11924536614731	4.52568500782056
H	-2.76369917009819	-0.23469384109757	5.33453084521983
C	-1.09062729431506	-1.12981045474176	4.26300307016850
H	-1.03682086956430	-2.07010667569415	4.82869910556184
C	-0.12321450529761	-0.91650092327157	3.26907260267143
C	1.02346534495962	-1.88559188278360	3.12902892254890
C	3.25340906929522	-1.99613188272604	2.21002758409076
C	3.44703580699717	-3.38789496319099	2.24337935654054
H	2.63142444249623	-4.01474457020689	2.62031392336363
C	4.65785089980365	-3.95277255727497	1.77598204465561
H	4.76575725936135	-5.04865449308785	1.78986064714034
C	5.69964748762622	-3.16978617018584	1.28955173453027
H	6.62858843567684	-3.62574310621259	0.91581740721477
C	5.56656122741731	-1.75231410796955	1.27852819863944
C	4.34396798455615	-1.16998998241835	1.75291971468275
C	6.57196465091842	-0.85554607839787	0.81575133322966
H	7.51807807464747	-1.26399500171772	0.42723316812706
C	6.35321074058488	0.51552408951556	0.85982694458247
H	7.11464110797217	1.22812248334756	0.51213412715667
C	5.11909636490094	1.00085887875091	1.35755921773959
H	4.90106935355766	2.07997459195248	1.40009870853318
N	-0.14731192017314	0.17387976699944	2.46390363867891
N	-0.65384567804206	1.92050548072327	0.48159713837738
N	0.05841355509196	0.86721458512567	-1.83108682645899
N	4.15039702936817	0.18613734787541	1.77490750974003
N	2.10114216406744	-1.29908067582590	2.52709714506740
O	-1.79459139941707	3.29905003235349	2.00992793810534
O	0.92145204399703	-3.03242533446379	3.59601893987540

Table S5 Geometry optimized coordinates of **1** $M_S = 3$

Co	0.16759385088010	0.11254040219401	0.14205236970456
Co	2.10675627086388	0.64198675461556	1.96520473020169
C	2.13290880754813	0.48257336640764	4.93870886246855
H	2.31780687050591	-0.59194844225280	4.79689971441205
C	1.96608373753925	1.04205363840050	6.22958880643384
H	2.02573042986841	0.38321367282393	7.10742681663235
C	1.72719090131274	2.40376655560444	6.36504712759453
H	1.58779075491730	2.85579657045893	7.35957366541785
C	1.66571387893731	3.23213844027208	5.20699391136255
C	1.84391169533949	2.58447632697884	3.94182497544146
C	1.80967980047170	3.31616202202576	2.70934572740409
C	1.60035805022908	4.70511882097427	2.75670440010785
H	1.57123291927932	5.26252447667579	1.81374994134914
C	1.41635907891316	5.33916005699502	4.00902195148287
H	1.23743509148482	6.42574708805208	4.02267577603909
C	1.44295831295846	4.63872426338307	5.21466131349238
H	1.29122197855182	5.15451605926299	6.17447408660680
C	2.20481742702677	2.95321964592302	0.32224990371558
C	2.57003611646871	1.78852540787069	-0.54231180396931
C	3.45601312607916	1.94881819459190	-1.60740519416192
H	3.69242837486534	2.98195440441128	-1.90054023641888
C	4.06821102203562	0.82679279336176	-2.19279747028792
H	4.80454132263145	0.94043958286452	-3.00132431439599
C	3.70842556096789	-0.44379886593235	-1.71153744249528
H	4.11718465755440	-1.37818121396039	-2.12227462171375
C	2.72692118515327	-0.56877004235156	-0.72806464452643
C	2.07906349152933	-1.88922283141421	-0.46273135948669
C	-0.16845292835130	-2.65711441253779	0.12374097210542
C	-0.00432638580300	-4.05262138998630	0.12523740222470
H	0.99930498566862	-4.46395408770976	-0.03113187283910

C	-1.12817829052149	-4.88593513791410	0.34052270086358
H	-0.97468740629429	-5.97661729000738	0.35222992503207
C	-2.41033363065334	-4.37661995260309	0.54412229551551
H	-3.26714075491782	-5.04539731226856	0.71429197044023
C	-2.61794368970664	-2.96773997882671	0.53353779734797
C	-1.48240219946632	-2.12000301842854	0.32530657988709
C	-3.87528088347524	-2.32188866769746	0.71390304882932
H	-4.77594574221551	-2.93095950095968	0.88762569867826
C	-3.95265249913015	-0.93591294258263	0.66451731784658
H	-4.91053866758795	-0.41262528359010	0.79472452762431
C	-2.77930610747982	-0.17237430699721	0.44916089904951
H	-2.80879399845784	0.92605274599033	0.40758898101644
N	2.16335189630498	0.53527976204229	-0.10169864927462
N	1.99034074887330	2.50958559989172	1.60472923960302
N	2.06197107488705	1.23086908665869	3.83511960620116
N	-1.58494032878156	-0.74877488164836	0.29748106800402
N	0.78706707916222	-1.67794028489048	-0.04767815451792
O	2.22656015686631	4.12804414981481	-0.06939521967877
O	2.61849225349121	-2.97745595778337	-0.70749385220998
C	0.28233463282853	-0.38650449978815	-2.79042211195115
H	0.56061828192263	-1.41525055783000	-2.52102699717762
C	0.22774536794572	0.03672313242292	-4.14128359511354
H	0.46873610206089	-0.68460281307914	-4.93485419341287
C	-0.12281083055880	1.34731668743871	-4.43952048206006
H	-0.16560287048748	1.69623894802744	-5.48315447944148
C	-0.43662224850519	2.25477576840137	-3.38653019486092
C	-0.35331456532890	1.74475628833619	-2.05047546343327
C	-0.65217727694444	2.56341979566790	-0.91188041101027
C	-1.04129925284784	3.89681955940973	-1.12429196133145
H	-1.26699195913739	4.52077135208791	-0.25248384078455
C	-1.11619357928706	4.39734731362093	-2.44620531611762
H	-1.41321440989438	5.44759368773801	-2.59101808734573
C	-0.82407477266460	3.61372155735535	-3.56206526557804
H	-0.88768736463589	4.02914375217261	-4.57894136368507
C	-0.91060651728477	2.35125037827387	1.51180841200496
C	-0.78725892067478	1.24562640637654	2.51122427320294
C	-1.68461421031561	1.14944637115935	3.57470913645120
H	-2.33035413800675	2.02106510641846	3.75692902335628
C	-1.79421558342942	-0.04967530301165	4.30106733555303
H	-2.52910201121301	-0.15710958953653	5.11175657834465
C	-0.93328386491557	-1.10667736289619	3.95796133034635
H	-0.92861330893832	-2.07209565876662	4.48459709507366
C	0.03499519035467	-0.92953677425185	2.96966223063433
C	1.17512287703003	-1.88675463400677	2.83951912265057
C	3.55329647321398	-1.72211778447178	2.28902030879793
C	3.97818266142266	-3.05181379538832	2.45063546159890
H	3.22848714945913	-3.81790730560950	2.67803853647784
C	5.35068640095851	-3.36656121458048	2.30426706862961
H	5.66148037874727	-4.41601887347123	2.42364069879892
C	6.31361257944740	-2.40170215003128	2.01152200862160
H	7.37335200152237	-2.67416717080264	1.89704361802129
C	5.92286742809026	-1.04115401283671	1.85624355585279
C	4.53398886924310	-0.71818203412686	1.99528578446364
C	6.80684077829673	0.04021747965005	1.57293567751110
H	7.88250513282010	-0.16001309046832	1.44733020608684
C	6.30550064905322	1.33113009715287	1.46243579789043
H	6.96540851951432	2.18401138686687	1.25055082107576
C	4.91687992732411	1.56272255454098	1.61863311289667
H	4.48932301539107	2.57172077617474	1.53211282997967
N	0.10860194687538	0.22894655357009	2.20672084646270
N	-0.50451904098565	1.89031479176366	0.28279351625033
N	0.01054992842716	0.44611012834730	-1.78280400201763
N	4.06246579217877	0.56696641303894	1.86518158856033
N	2.27514787867863	-1.21089655044994	2.37169151124059
O	-1.42125653815064	3.45005316247458	1.76837841252950
O	1.12519390507556	-3.06563235928645	3.21792259179696

Table S6 Geometry optimized coordinates of [**1**]$^{-}$ $M_S = 2$

Co	0.33704605554840	0.09265831558647	-0.15951669816447
Co	1.95252590135380	0.58093243330441	2.25328697445881
C	2.14585596607874	0.70276347143258	5.36476288010277
H	2.39589189704527	-0.36861505751182	5.29585286255753
C	1.89002330142672	1.33069800628732	6.60860398092077
H	1.94565705209877	0.74215982901729	7.53680593760983
C	1.57075357812083	2.68302338925865	6.62917825392869

H	1.36045698762477	3.20005778944028	7.57947371253792
C	1.50190777630974	3.42234101597971	5.41065893932932
C	1.77253755517176	2.70056137163956	4.19488065443293
C	1.71073732384772	3.34745470706897	2.90069973590961
C	1.38115910475208	4.72015726237064	2.88151063549238
H	1.31970691219141	5.21970200165854	1.91010119901587
C	1.11931457806930	5.41705616050606	4.08189123337462
H	0.84726854902310	6.48281622275328	4.01450823138315
C	1.17295535214805	4.80312158793085	5.33202032657435
H	0.94929770680893	5.35954874480652	6.25545758615486
C	2.13623801234009	3.04225807997623	0.54120001820669
C	2.61761357048397	2.02502859489719	-0.46990348958664
C	3.67099799444013	2.41212473885758	-1.31309014739340
H	3.93231967513338	3.48018307481036	-1.32022022501488
C	4.38709749518571	1.44932124411490	-2.04595019017448
H	5.23095111573148	1.74330135875504	-2.68837772307088
C	4.03713446765054	0.10103177191435	-1.87722923890458
H	4.58424741684229	-0.73384430750185	-2.33761677697361
C	2.91237160053356	-0.21923367051812	-1.11499580781830
C	2.44279273685128	-1.62817656467363	-0.92813856214976
C	0.47653424548038	-2.68684012636205	0.09244563267035
C	0.88381419892780	-4.02826287553864	0.20106288310381
H	1.88745089322663	-4.29878238800547	-0.14535571115552
C	-0.00940797281935	-4.97437216589493	0.75922670355480
H	0.32986827373579	-6.01758824000412	0.86471487391294
C	-1.29398291202066	-4.63225734374036	1.18233268377545
H	-1.96702782522249	-5.38740148394057	1.61600357912515
C	-1.75187768603291	-3.28882340905737	1.05291417935697
C	-0.84468097232002	-2.32427354953577	0.51666090970120
C	-3.05374974600148	-2.82231622044774	1.40623946868164
H	-3.78508399450123	-3.52525639291278	1.83475419358024
C	-3.38110191235584	-1.48760228041840	1.19088730071630
H	-4.37937981142915	-1.10056004592997	1.44226222724691
C	-2.42280322863343	-0.59516182791146	0.65829465498793
H	-2.65379356485449	0.46356765237182	0.47925334885801
N	2.15772973596350	0.73741432892616	-0.46456101928164
N	1.96322039622263	2.52511727071573	1.80812518259954
N	2.08934781182595	1.37077845325080	4.21297377633250
N	-1.17138850401314	-0.98682017854664	0.36032532749641
N	1.19033314219456	-1.59879741724252	-0.36322305288517
O	2.09434637221838	4.24081822411687	0.21330523954090
O	3.11320747538341	-2.62462035388567	-1.24882109063606
C	0.20977427865946	-0.22020874034124	-3.25671871056160
H	0.44091029031887	-1.28029684189858	-3.06050821866505
C	0.20174353793079	0.31126893538169	-4.56966759380482
H	0.42628494750654	-0.34529751423755	-5.42358980944656
C	-0.08866929020074	1.65796121922040	-4.75070117697241
H	-0.09935658050193	2.10417198943833	-5.75840277421189
C	-0.37102038930667	2.48837402533208	-3.62536937638633
C	-0.33271839900877	1.86445748704994	-2.32896508509390
C	-0.58159495780103	2.61746342737481	-1.11739384274972
C	-0.87479204549329	3.99102611641836	-1.26167505669123
H	-1.05611547347891	4.57447570166783	-0.35471102350149
C	-0.91483671476790	4.59008887603943	-2.54018002095034
H	-1.13695968346990	5.66780899665414	-2.60191551930169
C	-0.67117290504194	3.87521618927688	-3.71169006701325
H	-0.69332410442944	4.36484987328945	-4.69803074872388
C	-0.88281127311150	2.43259959673194	1.27132705299836
C	-0.92348675859495	1.43325578248728	2.40618997137453
C	-2.05909029216476	1.45237690989439	3.23112649159245
H	-2.73400986801787	2.31314275470753	3.12199798291887
C	-2.33176782585950	0.37573845968705	4.09355712086943
H	-3.23355829852376	0.37332804570938	4.72437987305530
C	-1.45560021068458	-0.71996138113579	4.07389124115059
H	-1.61979223544420	-1.64369940570893	4.64758883792448
C	-0.28398794390865	-0.64019392577026	3.31867782666812
C	0.72809297885470	-1.74246192491776	3.28930185270987
C	2.97830242536041	-2.01617528356386	2.34404537461519
C	3.16305939626104	-3.40854786778252	2.40672492820238
H	2.34786904792280	-4.02743415038984	2.79851967546352
C	4.38453214602251	-3.96097348894131	1.95181662271386
H	4.50916535802809	-5.05570093897570	1.98324297913254
C	5.42690408645989	-3.17255663560167	1.46393212427901
H	6.36452028645884	-3.62896778289258	1.11136500756203
C	5.28603331958735	-1.75525604062043	1.41773020501150
C	4.04585380556695	-1.19453458660382	1.85205593802584
C	6.28891968545359	-0.83903245763756	0.97989487313786
H	7.25596972630851	-1.22404098245386	0.62039603774089
C	6.03050770693863	0.52694476073154	1.02276277654030

H	6.78766638788030	1.25781550988426	0.70312989166490
C	4.77345110064363	1.00082160271723	1.46320588389389
H	4.53743327220719	2.07279991804058	1.50014398061516
N	0.02407802097740	0.45635619639969	2.53766692878730
N	-0.48562023082264	1.89351184701285	0.06580487388706
N	-0.05092077076380	0.53410733971881	-2.18912713876425
N	3.78771554060452	0.16610456356485	1.83586425180482
N	1.86538446160833	-1.27362837182000	2.67616743637902
O	-1.34881274864968	3.57002593228245	1.46118960779352
O	0.52406309469943	-2.87545493758810	3.75759292030406

Table S7 Geometry optimized coordinates of [**1**]$^{-}$ M_S = 4

Co	0.17644761237003	0.11894509780317	0.14622582936535
Co	2.08382156774160	0.63276118237697	1.94109673773178
C	1.77427999367638	0.86294852499006	4.94549345035433
H	2.11863889548733	-0.18149783883868	4.98064963799547
C	1.22598508569920	1.50180813242535	6.08262980572676
H	1.14327610179404	0.94084487700124	7.02566069874688
C	0.79388114840363	2.81986492558100	5.98808525092199
H	0.35139919884152	3.33518766621756	6.85538249565928
C	0.92446408725218	3.52007761921797	4.75185589434370
C	1.47842505073444	2.79009310175771	3.64859304273583
C	1.64188515207422	3.39749172649942	2.35227997559291
C	1.29058809999747	4.75392140841615	2.20563604739116
H	1.44298966025770	5.22609940338117	1.22889284986484
C	0.73340416784051	5.46106240611548	3.29506328125436
H	0.43318220063589	6.50966913925188	3.13955422505899
C	0.53927178857352	4.87400630978446	4.54458122935122
H	0.09267108216957	5.43979116464877	5.37689115539606
C	2.52647366220729	2.95667734903690	0.15081750559386
C	2.91310290050911	1.80012091103361	-0.72732512387432
C	3.78452686773708	1.98820926702115	-1.81340260770294
H	4.06234449516784	3.01932172832249	-2.07187112773132
C	4.30221563344741	0.85845709131525	-2.46462789512821
H	5.03399794424931	0.97086962520740	-3.28035945631623
C	3.87420833643702	-0.41559431360725	-2.06195360764703
H	4.23690474193077	-1.34871322137650	-2.51395400080237
C	2.87991123328795	-0.51137561895501	-1.07387478915463
C	2.21648149388173	-1.82764951870749	-0.78283757099990
C	0.20933112001685	-2.70813755198358	0.22379241899160
C	0.58656523690448	-4.05792434257134	0.37007515023960
H	1.59665242493340	-4.34758539433213	0.06175694543248
C	-0.32060033530691	-4.98744457149357	0.92763523475485
H	0.01493706768537	-6.02754855681546	1.06845810862431
C	-1.61038607131204	-4.62888605166782	1.31783178332590
H	-2.29687509454990	-5.36610887272144	1.76210778628159
C	-2.05146652912888	-3.28751200351666	1.13991419045473
C	-1.13379505720599	-2.33406154311017	0.58712787966903
C	-3.35136138191636	-2.80874195277438	1.48253678326782
H	-4.08041890013900	-3.50254138167672	1.93046034880475
C	-3.68052875876289	-1.47867862769189	1.24451225522334
H	-4.67697787572593	-1.08481104638474	1.49360189984689
C	-2.71614209741651	-0.61005313541881	0.68063508929023
H	-2.93861861725019	0.45017865912043	0.48546104345283
N	2.44105544497770	0.57517573100877	-0.39604525766255
N	2.09993160764156	2.53128257414378	1.37925039270770
N	1.87805703837021	1.48003627207395	3.76334635529035
N	-1.47754394996666	-1.01881355800711	0.38035257549133
N	0.99370596704583	-1.65180520682030	-0.19537038177166
O	2.67880754289951	4.12804925994233	-0.24726050773713
O	2.75466710780915	-2.89562025944091	-1.13444524633846
C	0.44488786608655	0.12197628073034	-2.86884209582126
H	0.55977493710975	-0.96948173673798	-2.78032174044859
C	0.71584248912287	0.80132939202031	-4.08006379788918
H	1.04790097892849	0.22482584217710	-4.95630424348336
C	0.56674172745198	2.18252902145585	-4.14087028308633
H	0.78509783376849	2.73605956516494	-5.06808800635792
C	0.12465620410607	2.89781418205702	-2.98806813340609
C	-0.11135162629101	2.13017789156101	-1.79992592463953
C	-0.54266170399730	2.75420368512918	-0.57530861961315
C	-0.78640751178944	4.14174928514208	-0.58669826627437
H	-1.14596294699841	4.61023021913487	0.33618267365114
C	-0.54099619162037	4.89203592270733	-1.75914009732950
H	-0.70565171888102	5.98113504945687	-1.72784605568474
C	-0.08675793413787	4.30431370565085	-2.93940973730961

H	0.11288246725758	4.90926088186291	-3.83761658370500
C	-1.22305818717786	2.23348355680243	1.67982469131717
C	-1.12804539283540	1.12268277622891	2.68714887933339
C	-2.03644212544880	1.04857412853899	3.75670157592942
H	-2.72721608607938	1.89145245213579	3.90034367023554
C	-2.05895537095431	-0.11433091833993	4.54206503224563
H	-2.79924715387792	-0.22920818800892	5.34979455051669
C	-1.12452409249440	-1.12824879115347	4.28357598521373
H	-1.07767087733574	-2.07086671305603	4.84611205385300
C	-0.14842249806411	-0.91050405345505	3.29636675542126
C	1.01087434098376	-1.85477084440436	3.14947073753186
C	3.23239589936894	-1.93721697010815	2.21189160410278
C	3.45924796378998	-3.32710161562527	2.25179107304041
H	2.65233769511045	-3.97304136298241	2.61419560250952
C	4.69232770510896	-3.85449274062385	1.80549485033005
H	4.82821182146482	-4.94829009995342	1.81377960617654
C	5.72752361304069	-3.04264866454107	1.34362598251755
H	6.67470624424157	-3.47545436555823	0.98591145365585
C	5.56044628034432	-1.62940560362851	1.33197952035425
C	4.30911647088156	-1.08571283839405	1.77463337361906
C	6.55232501938847	-0.69865478106905	0.90085759779651
H	7.51856346082012	-1.07547190656104	0.52945521985484
C	6.28796749313350	0.66532132907987	0.95538300007844
H	7.03714828903274	1.40433033338904	0.63488107910613
C	5.02964682516548	1.11450948458172	1.42135544366781
H	4.77869788445607	2.18535415151855	1.46677066228707
N	-0.18019956550505	0.17667826803188	2.48987860263018
N	-0.62826798326641	1.88821957083495	0.49726527528686
N	0.06387549202667	0.76737181778254	-1.76027682620446
N	4.06616505073306	0.26697490587574	1.80033174628507
N	2.06392798167700	-1.26225204489876	2.50678103306637
O	-1.84952350851530	3.27682010808372	1.94963604959262
O	0.95894834866258	-2.99604615381782	3.64869724867896

Table S8 Geometry optimized coordinates of [**1**]$^{-}$ M_S = 6

Co	0.23145237515463	0.13099222862278	0.20239260152159
Co	2.02941944499297	0.62000352804135	1.89384084825000
C	1.67936086064262	0.97727956786509	4.95564791814285
H	2.00516631035345	-0.07324900199268	5.01192474423205
C	1.10023874244754	1.63543210714206	6.06800996119687
H	0.97014272209255	1.08801297189532	7.01365805641082
C	0.69650629788234	2.95831123625512	5.93835349151338
H	0.23339735489089	3.49411992788082	6.78257436459612
C	0.87649159354271	3.63927517861577	4.69847383763211
C	1.45609806690387	2.88452707186680	3.62217206084751
C	1.64618826900595	3.47583666309835	2.31541275313721
C	1.30512298766261	4.83282467429246	2.14731826670674
H	1.47505033297276	5.28512441602262	1.16357100099175
C	0.73547945136597	5.56268279062553	3.21512590602134
H	0.44840679424516	6.61188357010905	3.03908435336444
C	0.51036311616042	4.99457357582276	4.46729534365272
H	0.05143515767757	5.57456049318955	5.28310648502682
C	2.49460850206463	2.99301392244935	0.11149454509305
C	2.88806395216993	1.80294380910741	-0.72738955651139
C	3.76681084543519	1.98307385622597	-1.80548396634301
H	4.03456146316152	3.01709597552117	-2.06373536753203
C	4.30196382189595	0.85749052146885	-2.44765670109056
H	5.03625091517505	0.96966615307145	-3.26083720921974
C	3.88041059446171	-0.41328348377193	-2.03138460609528
H	4.24594168000839	-1.35077458781112	-2.47230040720109
C	2.89026255020554	-0.51611672114019	-1.04344448258009
C	2.26727452755431	-1.86045807542175	-0.76178915279446
C	0.25480288768984	-2.77982236518248	0.19419035556659
C	0.64455033703932	-4.12944576810524	0.31309721664713
H	1.65323353920960	-4.40082773254355	-0.01731360924903
C	-0.24057868906545	-5.08032584108013	0.87035165584942
H	0.10830291145431	-6.11965233555383	0.98342603408249
C	-1.52359744505414	-4.74221008981960	1.29591911941869
H	-2.19327947372241	-5.49287524611972	1.74331855714873
C	-1.97942260970200	-3.40177279551550	1.15347794782858
C	-1.09073140275705	-2.42369583340232	0.59006542361637
C	-3.27296398490258	-2.94724870814730	1.54650597862799
H	-3.97670070347765	-3.66183101325493	2.00308069042444
C	-3.62968852628264	-1.61860456814218	1.35159694579111
H	-4.62022000193304	-1.24283253513760	1.64747553809906

C	-2.69215439126147	-0.72974490621141	0.77076314042835
H	-2.93062485433194	0.33314147658230	0.60578982511333
N	2.42403356152467	0.57293563226802	-0.37194621827267
N	2.10784298861646	2.60334559059392	1.35815911232803
N	1.83521252619218	1.57804418058782	3.77504661953329
N	-1.46433367368718	-1.11732694866193	0.41964814339684
N	1.02540530104080	-1.71864230038835	-0.21968370822374
O	2.62023648926256	4.14576024218513	-0.34735339513940
O	2.86435857077992	-2.90360992340592	-1.09380729661405
C	0.49427867515160	0.25311908507334	-2.88398401340329
H	0.63039857637110	-0.83807258805051	-2.81444596913373
C	0.78720484150943	0.96352858159264	-4.07385415753182
H	1.16103698200667	0.41737608159458	-4.95275257083240
C	0.60704672132035	2.34088413824982	-4.10279960519817
H	0.83786719485970	2.92298488570830	-5.00967308579399
C	0.12105640012031	3.01965920661860	-2.94668412737246
C	-0.13094570569639	2.21786646170991	-1.78163337737022
C	-0.59042938164771	2.81889129437511	-0.54837719873395
C	-0.85140073286555	4.20387074718928	-0.54444371512951
H	-1.22872328854028	4.65048766680800	0.38275330110062
C	-0.59862326936328	4.98040095662737	-1.69823105717880
H	-0.78065315487265	6.06624180966433	-1.65000492820373
C	-0.11306811365109	4.42160624128987	-2.87882478118868
H	0.09663432109115	5.04489529358067	-3.76212389316125
C	-1.21603050305806	2.28201127934735	1.71660403873379
C	-1.10168114561434	1.14076712032020	2.69580705678253
C	-2.00478166911477	1.06293765492849	3.76639961947413
H	-2.68511673079724	1.91452695337887	3.90982213610148
C	-2.04040462376844	-0.09983201591151	4.54930446452197
H	-2.77619622523509	-0.21021866193941	5.36141607647839
C	-1.11667601782236	-1.11848533593356	4.27482168128531
H	-1.06792144898990	-2.06458961256754	4.83153917913291
C	-0.14816695433806	-0.91534466603262	3.28113278273444
C	0.98375356599320	-1.90244947508041	3.14310589502188
C	3.21793541998006	-2.01291957900963	2.24584654512583
C	3.42322543341318	-3.40632612751749	2.29330200549234
H	2.60422307723588	-4.03146634617523	2.66672523960232
C	4.63929073253214	-3.96406893251730	1.83746860298159
H	4.75432379565300	-5.06025662610259	1.85689993109061
C	5.68095590656314	-3.17767615481159	1.34965912697193
H	6.61437477892617	-3.63106246417325	0.98117499170988
C	5.53663673717222	-1.76231585765267	1.31942543827719
C	4.30668608538020	-1.18102979863528	1.78142800143433
C	6.53537278684277	-0.86270375953939	0.84293725986388
H	7.48374009015617	-1.27034633655750	0.45708955332223
C	6.30315420365146	0.50689460582036	0.86555743592985
H	7.05703654123547	1.22062639600662	0.50180525473877
C	5.06445734595665	0.98937153794897	1.35503156290992
H	4.83302563457857	2.06636023560477	1.37907890013319
N	-0.15983035139816	0.18228299835393	2.47416863416724
N	-0.67606905379543	1.94657863824083	0.51157736172458
N	0.06840886455287	0.86322920319597	-1.77640131859416
N	4.09982060669539	0.17259285164760	1.78180065114271
N	2.06477388831322	-1.32655832634809	2.54655200633669
O	-1.81460152329446	3.32547238811284	2.04540499367111
O	0.86909859984281	-3.04928222903030	3.61967090546123

Table S9 Geometry optimized coordinates of [**1**–H] $M_S = 2$

Co	0.64013110105157	0.12096637177657	-0.00524150797123
Co	2.24697658440843	-0.01071350473449	2.28959009884051
C	1.66530990095667	6.01927647294273	2.95964184029282
C	1.02982365671418	6.38259303122110	4.15867121428377
C	0.94809070300719	5.43817720414275	5.18108179321513
C	1.46248760796175	4.12278811947932	5.01056653520775
C	2.06188207800690	3.80025366943426	3.73477185089589
C	2.49782080583759	2.45925150738125	3.45310047451274
C	2.51580342283329	1.55612644486338	4.54696475410474
C	1.95087815718183	1.87616145361512	5.79652331089184
C	1.39315965064725	3.13067724103118	6.02603546326806
C	3.08623930424093	2.47982187423636	1.04211331359470
C	3.36609688357600	1.51387214258621	-0.05217150270219
C	4.48072312330391	1.72933825920564	-0.87945630602499
C	4.98911110285215	0.67687952946575	-1.65545281645121
C	4.35243495397628	-0.57552010165892	-1.57888033225144
C	3.17725871974023	-0.69975107830072	-0.83978162585030

C	2.38194828710732	-1.97240672883922	-0.81068523353063
C	0.16317891329169	-2.64255183512463	-0.00542451043896
C	0.27543254659544	-4.04264728585302	0.00120433958079
C	-0.80936070677700	-4.82128755710376	0.47507236911374
C	-2.00147666604950	-4.25064183780348	0.91678428541771
C	-2.16342027868755	-2.83545896571544	0.88903851921633
C	-1.06629537433455	-2.04372995960806	0.42839123939297
C	-3.34129454845605	-2.13586730400251	1.28467185400115
C	-3.38408756178685	-0.74997926925436	1.18394819234313
C	-2.25139890337508	-0.03966580840402	0.72076709986144
N	2.65461585712618	0.33996041890436	-0.09699785994715
N	2.73874765683573	1.87503272024394	2.20632705483305
N	2.15846147532419	4.78904331445115	2.79373336233164
N	-1.11280293161067	-0.66088593862838	0.38083697824701
N	1.11704412515556	-1.70233672798545	-0.34878773593338
O	3.32185642387680	3.71567081999922	0.85212908160049
O	2.84774748105699	-3.06581328540327	-1.16634224023984
C	0.76500124362224	0.14666853086414	-3.11130619449164
C	0.95619740682930	0.78605848898174	-4.36079722548845
C	0.94635127820585	2.17353868888783	-4.41677387474251
C	0.74483670519424	2.93643969543717	-3.22868364315270
C	0.56453802026211	2.20591679433355	-2.00417840421046
C	0.37096287492901	2.88425931157702	-0.74204116255337
C	0.34277657937508	4.29288399545653	-0.76383996952871
C	0.51763818345992	5.00273129703255	-1.97551403710335
C	0.72020891214145	4.35862901151838	-3.19169104675350
C	-0.06960089342328	2.54669654732476	1.61032272593229
C	-0.32184366413846	1.48835286568365	2.66099042418737
C	-1.43357030562512	1.69099019429923	3.49822562878024
C	-1.93464690821488	0.63781059774274	4.27727088792213
C	-1.32047236857147	-0.61724587409267	4.16395025645224
C	-0.15711685571347	-0.73237925225626	3.39909730035450
C	0.56585935777564	-2.03632029568032	3.24644015969821
C	2.72656143979899	-2.73240297226456	2.32652418113836
C	2.61591997230710	-4.13045576697709	2.28332653544003
C	3.72936140923813	-4.89343773933603	1.85215382696839
C	4.94522374471182	-4.30838227170572	1.50008011515887
C	5.10526968238719	-2.89514208588176	1.58578024827679
C	3.97532528383873	-2.12138973260956	1.99175280809741
C	6.30726284043197	-2.17890774970369	1.31396399306557
C	6.33798234522052	-0.79917871984861	1.48623644823344
C	5.17151512743210	-0.10887170360553	1.89145810780263
N	0.39140523223092	0.32460714042092	2.71485416211856
N	0.24252260912238	2.05064950104729	0.37222336261908
N	0.57615536551109	0.83732997000620	-1.98773610105200
N	4.01025968549693	-0.74406793463420	2.10880520657388
N	1.77289940699029	-1.79094374899432	2.65113251119314
O	-0.29828479585731	3.73757974462506	1.90843860894584
O	0.09821120695614	-3.11978931127635	3.62304677978564
H	1.79593215777685	6.71256918841877	2.11621878364976
H	0.62314398197346	7.39567481585673	4.27771585889647
H	0.47860401699984	5.69795727489133	6.14231482279588
H	2.95999570093919	0.56193521005826	4.37695275267527
H	1.94425927670468	1.11393421990880	6.59025234581011
H	0.91719602531809	3.37835759102315	6.98578306650472
H	4.96517633028408	2.71470569809257	-0.83304998206531
H	5.88375675532007	0.82030666493235	-2.27854530447249
H	4.70851740763270	-1.46816081535110	-2.11256247362091
H	1.20947456851209	-4.49526432895113	-0.35199644358515
H	-0.69830041311927	-5.91707681324564	0.49521228288953
H	-2.82643813715568	-4.87703408247352	1.28708348993551
H	-4.20813513819173	-2.70268990275252	1.65815811091919
H	-4.28501614396118	-0.18688109398093	1.46734999383735
H	-2.25191772533035	1.05606926755971	0.63226739365933
H	0.77192628298009	-0.95133335777999	-3.01404501165170
H	1.11113841249901	0.17793218302130	-5.26376781465407
H	1.09631936232083	2.70096894622683	-5.37203871537788
H	0.17758708022698	4.81895704781889	0.18060309324143
H	0.49423627501363	6.10384936228043	-1.94348033144465
H	0.86338995696811	4.92594071831737	-4.12371854164924
H	-1.91093837110006	2.67968685893943	3.45805520466875
H	-2.81861319458623	0.78236461722341	4.91605667361499
H	-1.68721895614052	-1.52604253985145	4.66125016066107
H	1.66718599998772	-4.59469609900975	2.57617386142247
H	3.62315697321218	-5.98804647843575	1.79106898461058
H	5.79274325493995	-4.92394127044504	1.16415306235282
H	7.20274837445980	-2.72626644112971	0.98147305470240
H	7.25687147240239	-0.22433092928314	1.30191762756385
H	5.16344121970955	0.98190907944820	2.02384516961605

H	2.69175585188987	4.45605228973945	1.86234210211980

Table S10 Geometry optimized coordinates of [**1**–H] isomer $M_S = 2$

Co	0.88453998682083	0.28418647010174	0.09872909209367
Co	2.24069122936415	0.34486891605695	2.25334321463009
C	2.73492306724965	0.23370529900781	5.63495639528127
C	1.94636979173315	0.67000945465649	6.70205140434718
C	1.44492324845565	1.97632496778788	6.71484686446894
C	1.67952627795822	2.85251592189859	5.60313788497036
C	2.38790739575685	2.34130731372035	4.47287321595983
C	2.44824865616613	3.04315267036194	3.22543510114563
C	2.04152791735163	4.39731356300981	3.24545524683153
C	1.42979037285230	4.94667507859641	4.38959325393413
C	1.19850088689282	4.18649101937214	5.53587189010366
C	3.02312982767886	2.90158971522799	0.88206676194709
C	3.33238088788863	1.93238270145116	-0.24266522643138
C	4.41135720641247	2.24131735233863	-1.08972005025690
C	4.94248046943733	1.25988594865702	-1.94100305263503
C	4.40457214750379	-0.03497925115300	-1.88516875118669
C	3.27585076537490	-0.25963027902140	-1.09369388830183
C	2.65266863121359	-1.61591086469224	-0.92941308803572
C	0.62541180480079	-2.47990328628129	0.16122464935829
C	0.82519812373896	-3.86844777026584	0.13026474735211
C	-0.18201601131857	-4.72256334984536	0.64713190221788
C	-1.37630690587316	-4.23458574688269	1.17302696549063
C	-1.62655385908259	-2.83120530876031	1.18367142559243
C	-0.60669969838944	-1.96512409518043	0.67976772978284
C	-2.82558629667729	-2.21236406096724	1.64149883120312
C	-2.97089893984990	-0.83389831645253	1.53278645379510
C	-1.91398576866738	-0.05119133275806	1.01169464705226
N	2.69884123566209	0.73418814032187	-0.34563981140348
N	2.77195802392849	2.28391009543588	2.08779224917425
N	2.94254962946976	1.06300790354988	4.56790856644934
N	-0.74817257743543	-0.59417638429357	0.63960345183602
N	1.48072256240075	-1.47485670575409	-0.23710330146810
O	3.16215872551582	4.11609018938581	0.67211953921612
O	3.17549510419426	-2.65921464124935	-1.34783826287229
C	0.34371892961259	-0.22333997250283	-2.79971204882298
C	0.23544898845847	0.22073681189414	-4.14017364877778
C	0.16148196288590	1.58291298570546	-4.40312222979875
C	0.16879176120248	2.51796388060559	-3.32748676253643
C	0.26036306153450	1.98141763608595	-1.99923893998870
C	0.24095281146764	2.82370311173732	-0.83324084034561
C	0.17090268731247	4.21424566999467	-1.03867513273125
C	0.10774261007082	4.74079080947086	-2.35179480599280
C	0.09628854526325	3.93010133966752	-3.48336969410513
C	-0.08584800984651	2.74872189273573	1.54589304059764
C	-0.22710468038066	1.83872746159465	2.74307489719097
C	-1.24722067100386	2.13993587727311	3.66307709904089
C	-1.63602200798371	1.19169424862122	4.62061706344106
C	-1.02999637971338	-0.07465612620094	4.59164658247345
C	0.03332849933772	-0.29031528472648	3.71457110613287
C	0.75257266865957	-1.60130758514083	3.60229143375017
C	2.80541430701067	-2.37308390426634	2.49946060780757
C	2.73521143603020	-3.76731388546290	2.64501065288372
C	3.83088138213037	-4.56112886050443	2.22244465044607
C	4.98975305534972	-4.00545242291463	1.68193231506751
C	5.11120756277330	-2.59068758666485	1.56446942261549
C	4.00091548472142	-1.78947967192239	1.97238817674573
C	6.26664078799174	-1.89880267559190	1.09575563178026
C	6.28456971269036	-0.50771945519028	1.10588483165352
C	5.14628357656650	0.21142986480785	1.53512370880643
N	0.49578960542069	0.69004532813586	2.86822117901123
N	0.29262451277378	2.12941372710784	0.37732746421389
N	0.36513772573040	0.63410965544457	-1.77647503077218
N	4.01365281301116	-0.40514177318953	1.91864526374335
N	1.85424469265299	-1.41727275103455	2.80184169663020
O	-0.46589517720696	3.92701782780767	1.65596725805366
O	0.37424585283191	-2.64228684010816	4.15612040900535
H	3.20484715076401	-0.75593962328064	5.58069525342292
H	1.75534269225837	-0.02169745225710	7.53392776813090
H	0.85524532752561	2.34385847526958	7.56674602474622
H	2.13705507878884	4.97609542782499	2.32228598996723
H	1.07151616327454	5.98569805394274	4.34272930442447
H	0.64602730115125	4.59924086545698	6.39311662252237

H	4.83469939781077	3.25255977756786	-1.01031829678099
H	5.79565818744193	1.48966322126767	-2.59667523927947
H	4.81376372099135	-0.89036714050682	-2.44058347345014
H	1.76131063226677	-4.25742928516281	-0.28705857610631
H	-0.00428982314065	-5.80950756094219	0.63401497442109
H	-2.13938741843499	-4.91786056223186	1.57378413736736
H	-3.63211375956457	-2.83459377753357	2.05875943657433
H	-3.89426684665716	-0.33119850708131	1.85357984933985
H	-2.00286256712748	1.03841219688278	0.90601511202677
H	0.42631371355829	-1.29230566003320	-2.55126373825613
H	0.22068532638164	-0.52029050908500	-4.95220575905486
H	0.09408164699382	1.95556316385752	-5.43723167491797
H	0.15970204004039	4.87596742325467	-0.16872108137774
H	0.06370364569065	5.83489453135504	-2.47158239813248
H	0.03692101557585	4.36095884196799	-4.49419252069204
H	-1.73586148903671	3.11775951483131	3.55507376046486
H	-2.43605641034233	1.41879204077874	5.34085377511093
H	-1.33007906846625	-0.90978230446688	5.23986395696656
H	1.82968631942796	-4.20780328092537	3.07831099315803
H	3.75506471132721	-5.65530718696738	2.32201841000071
H	5.82352674858030	-4.64348712211050	1.35392926151593
H	7.13926964238783	-2.47247713786104	0.74778293489754
H	7.17056167553667	0.05206029930588	0.77402806765648
H	5.12807099359561	1.30997603449966	1.54978513029426
H	3.63520391351680	0.79949311473239	3.85308412217763

Table S11 Geometry optimized coordinates of [**1**–H] M_S = 4

Co	0.48114244301666	0.12856689190092	0.03414768982638
Co	2.25724721007410	-0.03065425562602	2.18962571193821
C	1.72608222311594	6.03622242592765	2.84402580825486
C	1.09846438008306	6.44497219301983	4.02674768516864
C	0.97668308859863	5.53177298402178	5.07719334892683
C	1.45119280344884	4.19064206589100	4.94558433838463
C	2.04496136546910	3.81593124287138	3.68315365160165
C	2.43719767226400	2.45410436614225	3.43931583498890
C	2.38684986407248	1.56607697966778	4.54824521545931
C	1.84325442078987	1.93725591437567	5.78476241689192
C	1.34336947559183	3.22841063030711	5.98209385461887
C	3.18606817217184	2.45080489177574	1.06973311281240
C	3.34458334961500	1.50720404646971	-0.05756624153794
C	4.21711354861842	1.79600547466100	-1.12657522547425
C	4.57024352095299	0.77538137143523	-2.01288064670354
C	4.01659110617144	-0.51460907468435	-1.83567655600101
C	3.04727983557077	-0.69849116659819	-0.85112011634564
C	2.24514974740698	-1.97326857402055	-0.75562700844716
C	0.01557467691849	-2.62493607385628	0.01361524547099
C	0.12588353157340	-4.02521532390129	0.03877307085907
C	-0.96409440153828	-4.79976496889846	0.50546732059690
C	-2.16464769637368	-4.22606160326933	0.92036901437681
C	-2.32752584500583	-2.81202511075154	0.87087124812831
C	-1.22440452714899	-2.02326373355364	0.41614589539094
C	-3.51077979989651	-2.10851470323168	1.24334659812202
C	-3.55270728109898	-0.72315121406983	1.13649245393580
C	-2.41397674844829	-0.01620267872671	0.68123720167454
N	2.70170382252638	0.29855082712404	0.01863247733163
N	2.70864843954404	1.86519504016577	2.20072429960194
N	2.17346579395772	4.77249816127708	2.71293031175008
N	-1.27771449263360	-0.64409136374296	0.35181591166326
N	0.97919338485472	-1.68978053312695	-0.31536364032664
O	3.57949902888509	3.65459929515074	0.93237425606801
O	2.71043220016231	-3.07335265689956	-1.08772291106703
C	0.81199812523122	0.10253006544305	-2.96340589944196
C	1.15707792212638	0.72621039948912	-4.18495469428929
C	1.26422460687930	2.11102938400756	-4.24031591295536
C	1.00963630119014	2.89091686927285	-3.07310917928333
C	0.67017800587422	2.17802265775125	-1.87580943984992
C	0.40678408051018	2.86332031095801	-0.63715557299077
C	0.45401007720160	4.27147318144541	-0.64121317282927
C	0.79563563532619	4.96784669746871	-1.82439000851290
C	1.07770018522119	4.31163129149000	-3.02088101582476
C	-0.16202248108715	2.48547958657043	1.68464224993577
C	-0.39100508784785	1.38380673464103	2.68496079179765
C	-1.46631150731962	1.52433807692015	3.57569997352739
C	-1.92022265706438	0.41944528802701	4.31077128495340
C	-1.28309713250130	-0.81562801456609	4.11709231241895

C	-0.14672270157208	-0.86996001293975	3.30707938813374
C	0.63859655929456	-2.13590119788181	3.14307817925542
C	2.85662528858688	-2.73774866553691	2.29279466301243
C	2.80329089115649	-4.13777632186388	2.24598949891810
C	3.96182393511236	-4.85901177566262	1.86324653628008
C	5.16724864470008	-4.22875459212393	1.55866932552795
C	5.26757600269291	-2.81002895643388	1.64335829002137
C	4.09466104392720	-2.07909036546632	2.00801850182453
C	6.44634692052540	-2.04729086025469	1.39920351608214
C	6.41789046198564	-0.66587813590501	1.55457103217107
C	5.21407535784685	-0.01977116514821	1.92564243795739
N	0.33669051816121	0.22623692077311	2.63637693850716
N	0.15743654921715	2.01421825576475	0.43952018525709
N	0.59027060530628	0.80696174509500	-1.84703615645361
N	4.07962299773585	-0.70418459266945	2.12246863499382
N	1.84767654792530	-1.83605647736925	2.57033953639948
O	-0.37764385321263	3.66846991978433	2.01115591860492
O	0.22836801679347	-3.24072592870725	3.52036786602050
H	1.88866087880488	6.69930397385679	1.98299742048491
H	0.72218706125549	7.47309352912235	4.11715230687782
H	0.50751150334309	5.83272904449849	6.02605874944989
H	2.77113412154211	0.54255169603057	4.40172639797600
H	1.80029316822635	1.19692861356441	6.59773100205574
H	0.87847830599484	3.51935093659818	6.93520404543754
H	4.63089784977906	2.81145236559538	-1.18708042956581
H	5.27669440372045	0.97026760645229	-2.83374956259201
H	4.27182901017746	-1.36855888939860	-2.47789502119913
H	1.06703244799027	-4.48209438294356	-0.28936638304754
H	-0.85017978756559	-5.89462630631335	0.54430913012965
H	-2.99430926805605	-4.84918076013347	1.28571359018364
H	-4.38295804743006	-2.67289389228048	1.60818991313424
H	-4.45668104117428	-0.15908202051902	1.40787297200864
H	-2.40708957218195	1.07999423078566	0.59188876371986
H	0.71913957410721	-0.99090922894314	-2.87745754153897
H	1.33844487737339	0.10429623452690	-5.07351172558088
H	1.53885121475415	2.61948103309565	-5.17772240636489
H	0.22515922736099	4.80187835020829	0.28859734840544
H	0.84202546616145	6.06799086886865	-1.78849229967901
H	1.34974959335422	4.87148068957576	-3.92829058807417
H	-1.95373228672405	2.50825866103582	3.61323687705741
H	-2.78202647229614	0.51143110557365	4.98841139079593
H	-1.60420232912606	-1.75030864060047	4.59763352527757
H	1.86102479658892	-4.63956647857275	2.49470440550430
H	3.89906861483093	-5.95669733628955	1.80070643134335
H	6.05005941745786	-4.81156121453772	1.25698918595711
H	7.37223314216295	-2.56117423699045	1.09759303725899
H	7.31729396501702	-0.05608362149138	1.38814976142960
H	5.15958211439757	1.06977777955892	2.05569679027140
H	2.77319620892060	4.44379570446608	1.85063452677445

Table S12 Geometry optimized coordinates of [**1**–H] M_S = 6

Co	0.41965174634216	0.17963504660903	0.07252811837610
Co	2.17978814692646	0.02720302102466	2.14591426133500
C	1.71113836891932	6.07768984133600	2.86402512319769
C	1.05523867404702	6.45913337602738	4.03669567295189
C	0.89991987693519	5.52047864319039	5.06380546467208
C	1.37606153618924	4.18243722612551	4.91392019297191
C	2.00692010853952	3.83377206214601	3.66226007258214
C	2.40805649340603	2.47889765658053	3.40694295740254
C	2.34285772082400	1.56915468041066	4.49956579683852
C	1.75235406828799	1.91183867414761	5.72262225911980
C	1.23736886464801	3.19596260381074	5.92489784198743
C	3.19026927851144	2.49522107642453	1.04527389999070
C	3.34660919264001	1.56321081991419	-0.10057143199258
C	4.18679441593908	1.89233558736259	-1.18300901568334
C	4.49595844804704	0.90626521018450	-2.12430572762426
C	3.95349896354714	-0.38697436216550	-1.97155843087180
C	3.03087883762905	-0.61092269997249	-0.94490079842291
C	2.27481680957689	-1.92058061928137	-0.84747425491909
C	0.12973555113737	-2.69096751069731	-0.00366136711408
C	0.32064953180377	-4.08323328089964	-0.04037280774461
C	-0.69355213897262	-4.94987165462381	0.43574473725835
C	-1.90040921492973	-4.47332082948127	0.94001775879784
C	-2.15310458227115	-3.07217567832786	0.95634795657297
C	-1.13848386687926	-2.18584775060072	0.46505011420050

C	-3.35470450975663	-2.47471890532511	1.43504097660190
C	-3.51046510245434	-1.09441659366084	1.39063527808803
C	-2.46386001545520	-0.29601080238090	0.87025141681249
N	2.71857960205783	0.35334795586364	-0.04144235442108
N	2.70258270794504	1.90138814868137	2.16421125440068
N	2.15947062955015	4.81173155786316	2.71408045829611
N	-1.31751069269433	-0.82527997766065	0.43531803519804
N	1.03317739956580	-1.70541509605817	-0.33028112674852
O	3.58666697352347	3.69729534269316	0.92074576208855
O	2.78789229727757	-2.97814027747692	-1.24772876019335
C	0.70221095333782	0.15953842172071	-2.98586933034142
C	1.12778669490106	0.75587952286826	-4.19492828506898
C	1.36945678869131	2.12520261185615	-4.22660049376526
C	1.17168922758252	2.91203481118918	-3.05488279342610
C	0.73860684451526	2.22228888886363	-1.87240992630640
C	0.53335241896671	2.92569756477779	-0.62772360941877
C	0.72688311930365	4.32041619458450	-0.62364943705672
C	1.15915409678657	4.99051791527423	-1.79146002983994
C	1.38811901503206	4.31756494351896	-2.98933709649181
C	-0.19057956563808	2.62325044482034	1.66495367568532
C	-0.41863730177282	1.54220707810475	2.68541421778251
C	-1.44177244663205	1.73314723907606	3.63075759826781
C	-1.89751640168931	0.65594952157551	4.40042040646198
C	-1.31548279743261	-0.60847638191799	4.19586182082140
C	-0.22635063326895	-0.71503344957874	3.33171942934143
C	0.51023579311092	-2.01093728459859	3.15820988603691
C	2.69019833904277	-2.70123972381529	2.27961150116934
C	2.58781038487779	-4.09930240357899	2.25706497394977
C	3.71542959288640	-4.86725855358516	1.87077892661737
C	4.93771793881533	-4.28532823992985	1.54023623005341
C	5.09089521429524	-2.87034851239554	1.60801921813995
C	3.94928865487940	-2.09234729924991	1.97575518649563
C	6.29558958224972	-2.15530330956313	1.34974811899852
C	6.32345389082249	-0.77326837014583	1.50262584149145
C	5.14903272679861	-0.08021281753877	1.88173181385201
N	0.26003982116685	0.35294418254645	2.61919052819434
N	0.20255897135627	2.11790045327858	0.45276311438367
N	0.52537953781684	0.86925924136924	-1.86741959909638
N	3.98943679838674	-0.71824871379467	2.08352248129513
N	1.71608744237682	-1.76131458545640	2.55645676770055
O	-0.45096869082838	3.80764109661605	1.94526025573518
O	0.07078477531397	-3.09694629486404	3.55742275112768
H	1.89469055812815	6.75972785971389	2.02246738671980
H	0.67886728752317	7.48594871821410	4.14055008639766
H	0.40551280213799	5.80070483888191	6.00595998867987
H	2.75366219146356	0.55640925604469	4.35042839078081
H	1.68973865951758	1.15761268843777	6.52138301596143
H	0.74000053759193	3.46577650361860	6.86786073487066
H	4.59644016510849	2.91040602045444	-1.22147570137143
H	5.16691210234886	1.13258763438031	-2.96690140299206
H	4.18501585059549	-1.21821934521130	-2.65126764308839
H	1.27333831197041	-4.46367389363338	-0.42785694249718
H	-0.50911847439182	-6.03580961242415	0.41547048891575
H	-2.66725173727350	-5.16163050984184	1.32510695327592
H	-4.15214602400381	-3.11896993367066	1.83706634041938
H	-4.42947794087446	-0.61116428848602	1.75204177225831
H	-2.54694360487233	0.80129131880070	0.81764948851717
H	0.51463468202754	-0.92324670391188	-2.90963010602146
H	1.27001486469993	0.12942580522147	-5.08731800669974
H	1.71239537850092	2.61584627602285	-5.15110712829541
H	0.53436042913085	4.86483374568061	0.30657261436756
H	1.32237797633792	6.07856574706051	-1.74265987565824
H	1.72930120948169	4.85272146833985	-3.88822635761424
H	-1.88589202681054	2.73673333729809	3.68160109133046
H	-2.71767836749995	0.78968256634808	5.12165176655280
H	-1.64644422529588	-1.52217770464515	4.70843627002431
H	1.63437011486750	-4.56428213596262	2.53186357410930
H	3.61250402073930	-5.96286745323905	1.82686003574179
H	5.79495407618084	-4.90328262400194	1.23488504026401
H	7.19802203498841	-2.70626865464596	1.04273105376255
H	7.24505615170520	-0.19982253469863	1.32820228371849
H	5.13893940427948	1.01124750447624	2.00946589555476
H	2.76891901724290	4.50316492556785	1.86365466622010

Table S13 Geometry optimized coordinates of [**1**–H] isomer $M_S = 6$

Co	0.07007910250384	0.56251876364720	0.05884506089634
Co	2.03618021795095	0.52152518999102	1.49895469804698
C	3.46467007797541	-0.33933680445438	5.35381195416956
C	2.99507281175222	-0.19812971248737	6.66653118481407
C	2.29766119699412	0.95814693855983	7.02801612108462
C	2.04931864240661	1.99264967637966	6.07736139649716
C	2.52169515406164	1.78375704882558	4.73521799368670
C	2.29312880449388	2.72609082877808	3.67860724253078
C	1.64791108063873	3.93407264802179	4.03555571412662
C	1.20597318384961	4.15614971466234	5.35444682461708
C	1.37566529526278	3.20760582383336	6.36637602145120
C	3.08117795293323	3.30997688687732	1.49938248270023
C	3.22790460210743	2.73610508845795	0.11692358733100
C	3.75227233399105	3.45262687293989	-0.96863024352768
C	3.90562388655214	2.79726657033072	-2.20192453271446
C	3.52994090474087	1.44693461049817	-2.31581779036237
C	2.96238657161339	0.81075716004509	-1.20475880429904
C	2.50193541733259	-0.62488377757685	-1.22189566410627
C	1.13107887874827	-2.16667678832896	0.04890134465542
C	1.86779022214169	-3.35476141971228	-0.09657787316612
C	1.34876746727245	-4.57615185654694	0.38911296007287
C	0.11167525405981	-4.64511752150669	1.02616669597900
C	-0.68722414687424	-3.47299629064654	1.15393116928874
C	-0.18038638969390	-2.23799849320743	0.63061872058886
C	-1.97084245696701	-3.43795085865835	1.77242121612801
C	-2.67929571779776	-2.24417073905089	1.82203132884003
C	-2.11658948526269	-1.07895206434773	1.25369720216498
N	2.81572545710261	1.45964059083691	-0.02627268877179
N	2.65874161520600	2.35099835299856	2.40298739676871
N	3.22094894300210	0.63821979856287	4.45652445661020
N	-0.90685925064425	-1.07100207191611	0.67605928666105
N	1.57958236628439	-0.88013165212094	-0.22121301312513
O	3.36255789323705	4.49146097773979	1.73274668361844
O	2.91664590545392	-1.41899231781349	-2.07285070212451
C	-0.08034301803985	-0.36874498312023	-2.89124561247952
C	0.20850386145821	-0.20659540080037	-4.26782559587981
C	0.52244491992352	1.05922870060266	-4.74734952810850
C	0.54080920784959	2.17358047153100	-3.85964946431221
C	0.22939972011857	1.91264472770178	-2.48401068466574
C	0.24784811759129	2.96865747580089	-1.49826754453050
C	0.56601346282534	4.26905093101766	-1.92967798838790
C	0.87658575717644	4.51474949693363	-3.29003944494756
C	0.87310526261237	3.50399148543120	-4.24679443058549
C	-0.18744619762707	3.39894892485075	0.84386819444425
C	-0.53349080190200	2.64604121317141	2.10451411148419
C	-1.27007593730078	3.28262071402747	3.11358349881456
C	-1.61996664762904	2.55998746040895	4.26363101866703
C	-1.15923634335208	1.24495957086781	4.39161922019097
C	-0.36509012966737	0.69415772464508	3.37112132145943
C	0.23969793588128	-0.66743878932522	3.58017291579213
C	2.14576939717052	-1.99114719395300	2.94841197652526
C	1.78519177730667	-3.20123031603143	3.57557032136698
C	2.64749685319073	-4.32221236046535	3.52859659774702
C	3.87713563641149	-4.28911565278424	2.87811200143545
C	4.30166973018924	-3.08333686387744	2.25096852336878
C	3.43797486199350	-1.93510793275904	2.29764574596549
C	5.53908089715335	-2.93321749161364	1.56179701163039
C	5.87398011711995	-1.71372636228054	0.98305292826193
C	4.97319480934481	-0.62999609007779	1.09905601114620
N	-0.09987144451772	1.36599536586341	2.22193802621135
N	-0.02803196414499	2.54870165074951	-0.21072046518615
N	-0.06548917583170	0.65252208640288	-2.03453107686504
N	3.80163625666335	-0.74380741165499	1.73046585467164
N	1.39747027559243	-0.83021692077010	2.87128384362357
O	-0.12770112358339	4.63972454424565	0.86369802899306
O	-0.29291438850214	-1.46073251340090	4.37753008791003
H	4.00462975028789	-1.22011408290786	4.98543097428538
H	3.18269315298393	-1.00218730687659	7.39062713777392
H	1.92919542394041	1.08390800454700	8.05728180742254
H	1.47777502706795	4.68553580656302	3.25484344173718
H	0.69384303393033	5.10328767829987	5.58385845720514
H	1.00692354891685	3.38828343252143	7.38640731695620
H	4.03090085201826	4.50287045093569	-0.80655531384416
H	4.31285204274979	3.33432389604323	-3.07086037039910
H	3.64384818714727	0.87158270572027	-3.24458955429516
H	2.85153985012614	-3.31518813786135	-0.57810078217421
H	1.95648952299138	-5.48724775335566	0.28299325793077
H	-0.26790426681667	-5.59748416237551	1.42471908732087
H	-2.38394953201645	-4.36004085776466	2.20906503527037

H	-3.66618065978529	-2.18594400194700	2.30130103417024
H	-2.65163881558217	-0.11654917284091	1.28097965933781
H	-0.30082680458886	-1.36210712714774	-2.46919996095227
H	0.19633212504967	-1.08398896549449	-4.92967354732486
H	0.76885919929856	1.21435570258388	-5.80975295911764
H	0.58005205103076	5.07313729560911	-1.18414380826628
H	1.13690004581395	5.54187595502899	-3.59063672150466
H	1.12391497328372	3.71366491443148	-5.29781779617456
H	-1.55343269817322	4.33186625659432	2.95397215469066
H	-2.21965050356346	3.02549048020919	5.06011693515286
H	-1.36073845571855	0.61811818452034	5.27088626049768
H	0.81885589726414	-3.24257047780873	4.08871748370444
H	2.31997114394456	-5.25294018150361	4.01726762797673
H	4.53029676184194	-5.17436613677053	2.84369409504707
H	6.22083686161000	-3.79562920281026	1.49198716375612
H	6.82238330472958	-1.57772717982588	0.44433513720614
H	5.20097582322090	0.35683507417884	0.66552430021873
H	3.48890936709326	0.51755800755557	3.45914314749961

Table S14 Geometry optimized coordinates of [H-**1**–H]$^{+}$ M_S = 2

Co	0.57370505389818	0.03500098701429	0.38203953360381
Co	2.45914672580022	-0.22086929772808	2.31080674606866
C	2.05102699540748	6.06909646011481	2.18430107452124
C	1.44529349505207	6.72971840704286	3.26812970359379
C	1.27374819694893	6.01992505515018	4.45043048751641
C	1.65254284024219	4.65285005880524	4.55449172196737
C	2.20738851477684	4.00957536364387	3.38521871955751
C	2.50139922250807	2.58889773888115	3.38662097091200
C	2.34908369477351	1.95497492566514	4.63253502812858
C	1.86920754111282	2.59292201209822	5.79448807606055
C	1.48386542018577	3.92302921650841	5.76052591176379
C	3.26689269456645	2.17305464661719	1.07719167403274
C	3.43514447204078	1.09307977051225	0.06563865449833
C	4.46818800344950	1.17861093037846	-0.87696053140109
C	4.79028642098242	0.05224769217380	-1.65039451720410
C	4.02431594043714	-1.11282083184435	-1.49102069379900
C	2.93434294325876	-1.10235995220473	-0.61719303973387
C	1.95296802440811	-2.23621990875490	-0.60185877126317
C	-0.42182720639379	-2.52671980606406	-0.11042491967033
C	-0.58700938259384	-3.90095260002431	-0.33628501593409
C	-1.88138079179108	-4.47076812119883	-0.24267157071429
C	-3.01037973040447	-3.70922759025007	0.05064069263857
C	-2.88053417116286	-2.30719022420075	0.26883252925902
C	-1.57041725755269	-1.73337870340138	0.19938197106862
C	-3.95911953615258	-1.41713943773123	0.53741580145945
C	-3.70848622851303	-0.05963592702952	0.69745886615950
C	-2.37991969910551	0.42356619054050	0.62520597669602
N	2.64525677059210	-0.01949654694421	0.17828753888778
N	2.78340799268429	1.76304996961429	2.27402287203812
N	2.40234007806547	4.78566049077252	2.27782465990391
N	-1.34125472696490	-0.39050394860958	0.40651482351443
N	0.74636768739111	-1.78156451177236	-0.14527535512716
O	3.67197509637998	3.32863777886770	0.73855321387260
O	2.23009521067902	-3.36294397414272	-1.03096107148088
C	0.71530875495366	0.17136644597807	-2.60321123674453
C	0.78687636651764	0.87459532522100	-3.83071148065467
C	0.69523671122288	2.26054283294538	-3.83412622189451
C	0.51745580614380	2.96160556742231	-2.60577052650979
C	0.47288530601453	2.17407070970413	-1.41033233253849
C	0.27901030083460	2.77612360475801	-0.12117211264935
C	0.08063976385989	4.16615669591126	-0.05584198115760
C	0.13347455960792	4.94204363155244	-1.24112111955444
C	0.35962040690976	4.37348406917385	-2.49195645227627
C	-0.12101995122088	2.13749618949871	2.19239013410200
C	-0.27240999526584	0.86768818606201	2.98230464478943
C	-1.37316775934229	0.73883070094549	3.83585493997768
C	-1.78229560649360	-0.53474380527416	4.26541693665171
C	-1.05120546187669	-1.65336297040304	3.83717442111863
C	0.10683285220025	-1.46095662049615	3.08166782763796
C	1.02538968455830	-2.59594621981841	2.74534873698141
C	3.38384369829475	-2.82134637570955	2.11539134426163
C	3.52211418913171	-4.20394861670090	1.92879647600467
C	4.80831867034484	-4.74115711050848	1.67261148898680
C	5.95349210998491	-3.94788289632876	1.62503067613415
C	5.85095152739251	-2.54343239053491	1.84014746599468

C	4.54745330204394	-1.99509635983064	2.06178154862314
C	6.94435446465623	-1.63207843019814	1.87293824803643
C	6.71329125434153	-0.28793067594657	2.14040509258210
C	5.39051825538401	0.17575202122504	2.33623125450475
N	0.50829917012673	-0.21592193889488	2.65259029814453
N	0.27905026362743	1.85613204603051	0.91631617471863
N	0.57463428994961	0.80434938923931	-1.43618225352113
N	4.33823578878883	-0.64839324467849	2.26855225217254
N	2.23736088982543	-2.08868391528091	2.36001001295432
O	-0.46537452892848	3.24176135338124	2.64836392901798
O	0.70318531692518	-3.78332770923975	2.84829013957851
H	2.28043359941334	6.56721173251753	1.23074614459303
H	1.14796663881915	7.78272197731871	3.17685710371619
H	0.83629337601554	6.50994554716157	5.33370503028313
H	2.58073664976683	0.88035737413819	4.66042931288896
H	1.77306307708370	2.00691401537269	6.72045837472312
H	1.05533563133263	4.42320971493975	6.64069255776092
H	5.03522474134093	2.11725445451494	-0.93294234881085
H	5.62814839526615	0.08343120987180	-2.36221853007439
H	4.20629911653629	-2.03797797123987	-2.05666981430652
H	0.29296236444092	-4.50773523371111	-0.57660902503776
H	-1.99093751007501	-5.55240483349940	-0.41363567709594
H	-4.00586242008592	-4.17342115589813	0.10972011865283
H	-4.98445092538790	-1.81272219845985	0.60184958179495
H	-4.52360889034244	0.65287755267589	0.88626229663639
H	-2.15835545681023	1.49218417584270	0.75597828591873
H	0.77310528351606	-0.92662328654251	-2.56685404907121
H	0.90819632299938	0.30799383407505	-4.76468542828899
H	0.74468301906365	2.82593723405201	-4.77745018213432
H	-0.13265331811672	4.61961282754767	0.91807551645329
H	-0.02347250219250	6.02918568471386	-1.16470679285094
H	0.39624799111809	4.99412992731405	-3.39906824536291
H	-1.92967107147328	1.65500856297135	4.07825478467390
H	-2.67098361166731	-0.65331649235640	4.90245086815794
H	-1.31456796844890	-2.68710676814215	4.10265366493976
H	2.63067660910891	-4.83942786491174	1.97728315378763
H	4.89861286775176	-5.82566572590586	1.50836460506277
H	6.94103680162740	-4.39254373141012	1.43451384471370
H	7.96611304768483	-2.00490877862337	1.70333276358988
H	7.54114387902967	0.43243430749968	2.19854872195964
H	5.18178396996140	1.23300553086256	2.54566617602427
H	2.98822962499008	4.25985143421377	1.47786681413783
H	2.61164044721399	-0.41403250760808	3.72639070969726

Table S15 Geometry optimized coordinates of [H-**1**–H]$^{+}$ M_S = 4

Co	0.41797674727103	0.02205036062719	0.27359949135251
Co	2.43138244576550	-0.27833777534680	2.27142796297242
C	2.39639236437167	6.06040061943340	1.79866367060252
C	1.73372823817968	6.80530313839765	2.79167447136984
C	1.36648377112993	6.15015693463993	3.96022910826116
C	1.61837612338165	4.76122932842488	4.14282762429651
C	2.23837485369826	4.03407759565150	3.05750353546094
C	2.42440324862477	2.59687560176416	3.13929531321173
C	2.13179712132106	2.03669871669596	4.39673398790192
C	1.58156057675501	2.75242262849126	5.47728017751258
C	1.27822066237092	4.09743529831276	5.34939843846664
C	3.30501598697593	2.00691407707659	0.90516401362058
C	3.45306662021278	0.86482998974518	-0.04356478681014
C	4.43179837327246	0.89424354405093	-1.04620593898424
C	4.65829510742708	-0.26488032374536	-1.80410590641819
C	3.84496848093680	-1.38791913677694	-1.58619441886752
C	2.80575881627797	-1.30899867106085	-0.65082500800536
C	1.76919917773065	-2.40717707799228	-0.57535427855119
C	-0.59017448927336	-2.63741184175118	-0.00435098944645
C	-0.71527318722485	-4.03368740762992	-0.04999111249949
C	-1.96675604176347	-4.64716106685022	0.21119654933619
C	-3.10368305816258	-3.90369970461652	0.50851638315609
C	-3.02548761067347	-2.48037750476312	0.55040126412042
C	-1.76122575949402	-1.85301346214358	0.29601735379006
C	-4.12721799713892	-1.62515951291425	0.83248914746560
C	-3.95035075758610	-0.24582596860531	0.84335799312513
C	-2.66369190027684	0.28543612906723	0.58919175566945
N	2.64390014949047	-0.21348249934835	0.14827629095672
N	2.74977544824470	1.69484859805072	2.10130504082250
N	2.61803642217640	4.75633422357684	1.96040528566812

N	-1.60745313899394	-0.49066364460580	0.33689442812341
N	0.56328512377010	-1.88617352547055	-0.18478118402334
O	3.76705631939579	3.12159767948279	0.50707943051223
O	2.04679603130330	-3.56423146567584	-0.90931276587222
C	1.06305318007798	0.69914211765825	-2.61995932052901
C	1.37003579977553	1.57556858495167	-3.68858138887521
C	1.26692574023072	2.94716122037120	-3.49209634449312
C	0.85887905030942	3.45386433347976	-2.22381920362964
C	0.60737866935417	2.49448985805288	-1.18898632404952
C	0.21411086089355	2.90943967234048	0.13623499255778
C	-0.01612676101508	4.27802274646623	0.36038675608930
C	0.21150039640451	5.21796925103656	-0.67573097156753
C	0.66428959976211	4.83647301645095	-1.93555405949166
C	-0.30767204281356	2.01564931647713	2.32674735909266
C	-0.40935050826917	0.66236978760761	2.99794779261220
C	-1.56476322157996	0.42012020794188	3.75547150481257
C	-1.92480156181959	-0.89299143712269	4.09039994898024
C	-1.08461117813018	-1.93654929294728	3.68389617585547
C	0.11549029086656	-1.62822193401704	3.03773174640686
C	1.09618810747229	-2.71284831763900	2.70480451112007
C	3.45438855880137	-2.83769705710459	2.06118587014032
C	3.64072632728071	-4.20822507916682	1.83454278670419
C	4.94613306862216	-4.69263756590419	1.56837258331404
C	6.06387704797193	-3.86002645654638	1.55395961406486
C	5.91197303857286	-2.46718706175762	1.81135186443937
C	4.58777898512922	-1.97077748379821	2.03397189630998
C	6.97236148035428	-1.52059220132725	1.88844754683893
C	6.69095745247667	-0.19573309256056	2.20115614206620
C	5.35047778232177	0.21650544962299	2.38915715831877
N	0.46763092459076	-0.34735253351430	2.69411944498997
N	0.10121258375000	1.86736482950972	1.03761457027644
N	0.70570861733899	1.14678236622926	-1.41431344401137
N	4.32834150027572	-0.63924909892388	2.27113869836826
N	2.28190862922761	-2.15136494223322	2.32301281099122
O	-0.68808672104392	3.04478224938354	2.91163521666456
O	0.82804059338729	-3.91438064741584	2.79910492867286
H	2.76385224268329	6.50217305776069	0.86132121920540
H	1.54124464604913	7.87628480898186	2.64287154327931
H	0.87436517803552	6.70252524558303	4.77550873102378
H	2.30318711065757	0.95464876981990	4.49420257763997
H	1.36351187235177	2.21614486682192	6.41245532330936
H	0.79382355744441	4.66130595866317	6.15905635237827
H	5.02287530898099	1.81106989567930	-1.17066672626792
H	5.45546213171542	-0.28898482075150	-2.56204069825093
H	3.95304272014225	-2.32625915689380	-2.14835562588022
H	0.17291589370446	-4.63297712227117	-0.27947425543796
H	-2.03008770607083	-5.74552738734069	0.17891355402742
H	-4.06672592855103	-4.39602230356251	0.70988918560493
H	-5.11507841542070	-2.06772603768990	1.03453659795694
H	-4.78775969449501	0.43607060946025	1.04812151413685
H	-2.48074609432663	1.37228947973275	0.60284808312128
H	1.10637602381352	-0.39457331955766	-2.74454476729781
H	1.67063299655064	1.15843146455253	-4.66001495607212
H	1.48343950876153	3.65071264263798	-4.31081699498934
H	-0.37034367442365	4.59137320910286	1.34930337934542
H	0.01916060988116	6.28292799253647	-0.47109468591210
H	0.84527412590670	5.58147057629261	-2.72449308103564
H	-2.19485020953062	1.28724552602406	3.99719299454759
H	-2.85642232109950	-1.09829587693878	4.63769466241170
H	-1.29437721939806	-2.99916465458456	3.87257111505940
H	2.77311375810048	-4.87666155073233	1.86203654652950
H	5.07413477457023	-5.76795835340585	1.37226649834543
H	7.06728815885217	-4.26524662848470	1.35837917906337
H	8.00857920280782	-1.85144281471540	1.71931749472083
H	7.49236524459694	0.54967298968020	2.29971218818626
H	5.10192633032641	1.25852407704644	2.62730536570546
H	3.18200773969713	4.14303395801801	1.20915427703416
H	2.58291205261347	-0.41231343025854	3.69521064117417

Table S16 Geometry optimized coordinates of [H-**1**–H] $M_S = 3$

Co	0.54357429974794	0.02207901195405	0.43512669926188
Co	2.45769797288607	-0.24239685680845	2.29209260765820
C	2.32987208541796	6.09542530899329	2.08529013663604
C	1.64490996061581	6.79926106998122	3.08794258600045
C	1.26322442593413	6.10205278417746	4.23222068847661

C	1.51959927867171	4.71070816854186	4.37314797921863
C	2.18474308198890	4.02821329705526	3.28085925788003
C	2.37568763423838	2.58805307650582	3.30614104577610
C	1.96697378260241	1.95738823702464	4.50388498280388
C	1.38199207770893	2.62985391742826	5.59034449308081
C	1.12092818666503	3.99072090319485	5.52883993607585
C	3.32096717249500	2.13342381282117	1.07169851651798
C	3.38630723546152	1.09024739368148	0.04066013728739
C	4.32627113114196	1.19034593386571	-0.99852464937859
C	4.61680309139624	0.07619344082527	-1.79216002035775
C	3.89670561695787	-1.11196943015273	-1.54818895657046
C	2.88372895598736	-1.12880503484379	-0.59550365043956
C	1.90976944972958	-2.26208730364334	-0.55908782015518
C	-0.46496459478678	-2.53605968794731	-0.06577226284829
C	-0.63655945733068	-3.91258401462325	-0.28652364825514
C	-1.93012469500960	-4.47776739163328	-0.17924137696656
C	-3.05661817535971	-3.71502260773172	0.12555842988506
C	-2.92150664843981	-2.31406905891755	0.34327391900460
C	-1.61134426491335	-1.74153668727268	0.25724901625778
C	-3.99326673730571	-1.42305892289141	0.63648096225420
C	-3.73476737456160	-0.06864732000781	0.80455431393804
C	-2.40638543500418	0.41226954126355	0.71442264479303
N	2.60897753246335	-0.04238213790629	0.22549121711144
N	2.74063642949351	1.74916971577531	2.24181998288403
N	2.57186582712407	4.78615367398282	2.21362391130373
N	-1.37370250849912	-0.40225718662265	0.46915005439087
N	0.70004616806862	-1.79562988621331	-0.11119600668650
O	3.88038196191277	3.25807656818639	0.80976626290390
O	2.16719028057170	-3.39623155013815	-0.98723923145870
C	0.78065311677766	0.22955251824827	-2.52633966091270
C	0.95042271737944	0.96588405796783	-3.72418598380605
C	0.90898016575052	2.35399671332343	-3.69253004119107
C	0.68388814470888	3.02704527033109	-2.45588421562429
C	0.54012316976550	2.20577325277405	-1.29175160603734
C	0.30432773132461	2.77253602380094	0.00762778588198
C	0.15830114793263	4.16918718212995	0.11115998947261
C	0.31016529243158	4.97621235478304	-1.04235218421442
C	0.58086364064072	4.43987402627613	-2.30077814923041
C	-0.21034961010898	2.05196220607966	2.27920985623891
C	-0.29078696256398	0.76759563557700	3.03197955244497
C	-1.34525805541565	0.58424895424011	3.93450358350860
C	-1.72535438225738	-0.70258192937068	4.33897804009432
C	-0.98332195623603	-1.78996886965179	3.83504937926222
C	0.12285486045087	-1.56593463290892	3.02385896210278
C	1.07101392826589	-2.66665588663940	2.69284693102624
C	3.44048627255642	-2.82850855486023	2.10206356269789
C	3.60794908432113	-4.21217310126507	1.92335840424538
C	4.90053737773085	-4.72445946218147	1.66020663692165
C	6.03139508219003	-3.91012826636297	1.59266381896764
C	5.90210372152830	-2.50676935059359	1.79427304316525
C	4.59110817363615	-1.98003054141978	2.02912460364124
C	6.97709528013849	-1.57189559961020	1.79278752457210
C	6.72114669635346	-0.22928415719370	2.03954261097104
C	5.39200223528020	0.20946870724312	2.25368596595902
N	0.48660023206928	-0.29612787210618	2.57676962979310
N	0.24188828159518	1.82316957539479	1.00282262378908
N	0.59273906720686	0.83305779899934	-1.35022995761698
N	4.35695890837644	-0.63748095837322	2.22330836099322
N	2.28432772817386	-2.12251151378155	2.34654070116870
O	-0.62076190298465	3.13589156772834	2.73450965477350
O	0.79595196560109	-3.87044020600882	2.78977920189438
H	2.70275751950023	6.56824622345501	1.16484521612529
H	1.43509470472780	7.87091174326688	2.96752630882422
H	0.74087097870799	6.62053393384863	5.05113185526353
H	2.10775888449379	0.86516041354639	4.53648852067970
H	1.08990659522852	2.04548382434346	6.47592732004473
H	0.60459499357129	4.51984746219204	6.34297555062115
H	4.86240075419716	2.14418999184052	-1.09378253171853
H	5.38575287600706	0.12361185793197	-2.57665576894336
H	4.05049268689186	-2.03759830364994	-2.12176423726243
H	0.24300483007035	-4.51889699643084	-0.52801651665753
H	-2.04279621596388	-5.56057281047169	-0.34487401614384
H	-4.05227493376362	-4.17765358511817	0.20018670276593
H	-5.01760800731126	-1.81833957034302	0.72063950267482
H	-4.54312293415460	0.64446681178401	1.01996770897648
H	-2.17850778181559	1.47679067577174	0.86356215609566
H	0.80479738717190	-0.87003330670656	-2.51731138258373
H	1.11309972416877	0.42172372283661	-4.66557171348026
H	1.03943366152040	2.94361421438609	-4.61334168436530

H	-0.08277083405862	4.60024490190344	1.08870270644588
H	0.20325581050517	6.06751389194176	-0.93451667302157
H	0.69810508272971	5.08789683454692	-3.18211070708783
H	-1.89125033196699	1.49054325153528	4.23403462439645
H	-2.57800473607310	-0.85979706538545	5.01479010536344
H	-1.19656861302309	-2.83690494293339	4.09581931449699
H	2.72653929179314	-4.86017656945226	1.98175448678482
H	5.01016642287437	-5.80891361614306	1.50247449245639
H	7.02548720519490	-4.33579493615522	1.39044596569809
H	8.00272288073909	-1.92718898737798	1.60730112411244
H	7.53323020653162	0.51129644666032	2.06295584112622
H	5.16546194350132	1.26645770322767	2.44278506116245
H	3.22124401911076	4.19821558859872	1.47667734362204
H	2.61245151320391	-0.39923817292411	3.73411296429133

Table S17 Geometry optimized coordinates of [H-**1**–H] $M_S = 5$

Co	0.49599663839466	0.04488243456984	0.36755374642400
Co	2.43331768685962	-0.28863008144067	2.26372354938361
C	2.40745738553551	6.00527130410393	1.55699753928386
C	1.69898861781993	6.78362663190748	2.47744897853596
C	1.26084133966402	6.18204036212906	3.66022784356846
C	1.50319894062928	4.79995408949310	3.92221205788194
C	2.18413118768727	4.02533582630921	2.90557406063711
C	2.36636730855113	2.59836498249095	3.06288497665651
C	1.96108933647535	2.06789599252219	4.31517384922835
C	1.35609832353810	2.82872817273596	5.32301429530435
C	1.08789056601975	4.18012467605137	5.12635754186739
C	3.45079105550536	1.95957578090024	0.94011428405681
C	3.55802606276315	0.83508323363491	-0.01733468642537
C	4.58850952896621	0.81526889293136	-0.96989583530945
C	4.80815446217365	-0.34552722980204	-1.72239166200341
C	3.93898448938536	-1.43964826435444	-1.54509339420591
C	2.86597186591133	-1.32367299112382	-0.65814700829993
C	1.79468664660888	-2.38541191821517	-0.64080114562396
C	-0.55305390424735	-2.60894736673536	-0.06086500902476
C	-0.68693333816802	-4.00347763688036	-0.16290841037634
C	-1.93213205445120	-4.62573535682331	0.10001592315144
C	-3.06216168816189	-3.89713861059240	0.45598394459537
C	-2.97895638429123	-2.47826522819028	0.55546507282279
C	-1.72087631628774	-1.83821150704677	0.29315782950232
C	-4.07145565527334	-1.63660856706755	0.90749312289953
C	-3.89415093267412	-0.26005352087323	0.97178606190832
C	-2.61579085506868	0.28293323402610	0.69623577311072
N	2.69591448513836	-0.22038504753084	0.13521989168126
N	2.75732337352959	1.67505190652006	2.07724379415883
N	2.61599751744579	4.69462871473949	1.78950392639948
N	-1.56870480570361	-0.47964819914500	0.37911072765635
N	0.60064137795009	-1.86028232637550	-0.22142490637694
O	4.08153936329967	3.03280611674003	0.65580880994054
O	2.03797769827310	-3.53595584750255	-1.03434866299086
C	1.16151643170005	0.80522547482167	-2.52266267539449
C	1.45088575201473	1.71076273982420	-3.57175729316878
C	1.26405963305297	3.07193288469792	-3.36486741084708
C	0.79350575304829	3.54038571636709	-2.10350643873795
C	0.57033656303232	2.55290035817289	-1.08633083555931
C	0.13286820061758	2.92706617261441	0.23883650566193
C	-0.17889679814532	4.27997202800663	0.47291530651354
C	0.01913227628357	5.24244248357290	-0.54572254592552
C	0.51736593757840	4.90551193316256	-1.80221979490605
C	-0.39838974290939	1.98477704175474	2.40016887446548
C	-0.39950451342611	0.64623278922465	3.09189653662859
C	-1.46862914394456	0.39355672915837	3.96713863023449
C	-1.80214804516901	-0.91910129894329	4.31812144336917
C	-1.01136173252105	-1.96306318816174	3.80362456242486
C	0.11722219299375	-1.65250436412049	3.04752936039739
C	1.08245206791384	-2.73395317838477	2.66727007700686
C	3.43590582481681	-2.86101758360054	2.02719886851171
C	3.60952638374957	-4.22970413621598	1.76880307212978
C	4.90917194882288	-4.72025316008166	1.49400853901652
C	6.03691935946330	-3.89871366119509	1.49970381630797
C	5.89935777055878	-2.51025924907824	1.78547733044348
C	4.58101064438269	-2.00499669971126	2.01711062739171
C	6.96859123448258	-1.57233346270707	1.87614979128699
C	6.69697177580305	-0.25018301958777	2.20719260236554
C	5.35958341699339	0.17362296801824	2.39417088289837

N	0.44522613717110	-0.36481102527216	2.67954778119811
N	0.07480247699640	1.87461050959574	1.12353578819626
N	0.74513814276262	1.21693505600657	-1.32375705890155
N	4.33173463195485	-0.67536729543733	2.26563629798690
N	2.27448762291618	-2.17049439235869	2.30814133148283
O	-0.91033350151351	2.98063361446443	2.94364163014083
O	0.81186733331952	-3.93961205014916	2.74026678025202
H	2.81586808603514	6.39834628703993	0.61616278991500
H	1.51046879309716	7.84595509464363	2.26935867352233
H	0.71877228190576	6.76879391768743	4.41719825395806
H	2.12272024537203	0.98916468930325	4.45561393082517
H	1.06090528430741	2.33298023527697	6.25982374131969
H	0.55641521465308	4.77735399671466	5.88121395853946
H	5.22479233071181	1.70763944647046	-1.04438877542205
H	5.63996545188632	-0.39936411687153	-2.44065093681716
H	4.02822786337873	-2.37840459689760	-2.10952854256302
H	0.19838885032131	-4.59064015472679	-0.43046581516620
H	-1.99565054952585	-5.72260841387007	0.02555509019752
H	-4.02059830519294	-4.39611710269175	0.66514104629007
H	-5.05131156754589	-2.09066832275028	1.12446758557843
H	-4.72224261424197	0.41268866557278	1.23706835593340
H	-2.43537921029792	1.36888680619603	0.74591124966883
H	1.27166109108721	-0.28238463026462	-2.65665080230697
H	1.80750568652753	1.32242657841800	-4.53649551218531
H	1.46657989640832	3.79764346502933	-4.16825408818880
H	-0.55656637547422	4.56232869629726	1.46181768474755
H	-0.22150768097953	6.29389004121238	-0.32377283232769
H	0.67933347884168	5.67100212396108	-2.57622433076936
H	-2.06161318148192	1.26626533176621	4.27447279148092
H	-2.67127341560630	-1.13002959079011	4.95870301264018
H	-1.20367204571120	-3.02771221549384	3.99795691233964
H	2.73188128557436	-4.88514969282910	1.77731459184617
H	5.02679175838737	-5.79215119240334	1.27177219231329
H	7.03694489010947	-4.30953876244847	1.29595521662620
H	8.00171025983546	-1.90980973442650	1.69964369181204
H	7.50386295795042	0.48911268202154	2.31000978947512
H	5.11678527919987	1.21724668297981	2.63037914808059
H	3.29388500426766	4.09318537882578	1.16327488873529
H	2.56368408260145	-0.38922862851623	3.70121820101263

Table S18 Geometry optimized coordinates of [**1**–H···H]$^{+‡}$ M_S = 2

Co	0.49309010074510	0.09464188627227	0.30422404681364
Co	2.22974699663876	-0.03511154764261	2.40095319121789
C	4.27232700843987	0.77553074312709	6.09776555789205
C	4.32549079079266	1.62489401490844	7.21455583916070
C	3.74022861951069	2.88768356782984	7.11929536487159
C	3.12036077565470	3.33034171480302	5.91942221588452
C	3.09727801246093	2.42341804406898	4.79512321095521
C	2.52240975172493	2.78279494636731	3.52822389077801
C	1.98855084781377	4.08431172107124	3.44916405924851
C	1.99365963141511	4.97389137110705	4.54918432912634
C	2.54702171537135	4.62122696795103	5.77343105562968
C	2.90690468301382	2.46897340188544	1.20336386625881
C	3.18348208804161	1.41809259135322	0.16318721257564
C	4.21831816220746	1.61350989971298	-0.75719262600276
C	4.69438458184531	0.52037855524234	-1.50204550895261
C	4.08466326288993	-0.73332672142570	-1.33053187572860
C	2.97083052907285	-0.84048246876433	-0.49420917785369
C	2.11381410272282	-2.07311259285799	-0.47774757989731
C	-0.26114543110173	-2.55408739317739	-0.10436631695663
C	-0.31221534915716	-3.94347668111107	-0.28841031343843
C	-1.56551638437839	-4.60410959409952	-0.24515701350111
C	-2.76185720346288	-3.91943225768405	-0.04152354065453
C	-2.74886096502125	-2.50417316286684	0.12387881377507
C	-1.48327273597473	-1.83685148897226	0.09881080224607
C	-3.90311411863817	-1.68710250239057	0.29827819729372
C	-3.76355516614856	-0.30932123026611	0.41267112500306
C	-2.47230414796480	0.27136982375591	0.38168200874191
N	2.52394904872116	0.22350554766340	0.25519151019739
N	2.58725990036012	1.90960010153773	2.43411289185555
N	3.68648003194779	1.19609030154137	4.96795774191174
N	-1.36961345570606	-0.47161037678796	0.25046642926757
N	0.84721592525599	-1.72500714280175	-0.09251480997750
O	3.11617192533880	3.65939175780370	0.95517513692891
O	2.52298188300763	-3.18165939250693	-0.84429205530904

C	0.67774127702899	-0.06449006533714	-2.67404611857578
C	0.77399845044202	0.51768974805812	-3.96138151919597
C	0.70261383790742	1.89839466087498	-4.09765809202981
C	0.51222515606288	2.71602686848300	-2.94669587694151
C	0.42163663682973	2.04630175715081	-1.68315279053532
C	0.20147539690937	2.76773265656044	-0.46444420405082
C	0.07028541480030	4.16357989140227	-0.52629464204183
C	0.17311288325293	4.82439214496773	-1.77593657792201
C	0.39220358888610	4.13525561374676	-2.96583650328084
C	-0.29837591584781	2.34606146013037	1.88351534215544
C	-0.52191558004595	1.14656696371209	2.76066922807263
C	-1.66565363500897	1.10932485708308	3.56413743822867
C	-2.13388339873789	-0.11801215550185	4.06424510352593
C	-1.41420104672897	-1.28260593410707	3.75558096546182
C	-0.21235714910107	-1.17636620996375	3.05378093407507
C	0.69944120878928	-2.34952789846945	2.88348850495945
C	3.07619343965959	-2.68617482482740	2.42169964728558
C	3.17013399301773	-4.08679121189614	2.38967568996751
C	4.43667153005177	-4.69236641423487	2.20650297867973
C	5.60690722926536	-3.94817905476897	2.06209706206243
C	5.55020735916276	-2.52674138140357	2.10947922826858
C	4.26966016503738	-1.91131746096169	2.29514591154572
C	6.67052315345610	-1.65652651216272	1.98188199285335
C	6.48769347408786	-0.28053480010490	2.05214355335464
C	5.18994307630983	0.24852340415487	2.24551161875526
N	0.24631168702262	0.02332791562157	2.55242995746484
N	0.13756023303040	1.94363916814052	0.65159191191105
N	0.52069433050287	0.68003728532403	-1.57633659767281
N	4.11158635479227	-0.54106047964540	2.35853936135715
N	1.94882505010491	-1.89970601242635	2.54483846531083
O	-0.63546381990902	3.48848371843415	2.22852028090037
O	0.34338389651132	-3.51740467869227	3.07286006129472
H	4.68065144196925	-0.24476954344463	6.08825901941731
H	4.81142620769831	1.28245235027401	8.13750115339133
H	3.75757530321356	3.56874342303732	7.98452249162893
H	1.54232410196452	4.40870462637609	2.50659853679744
H	1.54001154020615	5.96706594381639	4.41806392824178
H	2.55764353803595	5.31731420564734	6.62443772275201
H	4.66603348030905	2.61538361081210	-0.81387196868423
H	5.53645207394800	0.64399011307631	-2.19883215480364
H	4.40351328713123	-1.63653937730674	-1.86989303403135
H	0.62210193110735	-4.49084521833314	-0.45725441078413
H	-1.58666709216242	-5.69623759981725	-0.37957512470724
H	-3.72213651069919	-4.45509436270197	-0.01659413418408
H	-4.89870110228585	-2.15611300160466	0.32790335907209
H	-4.63870720670396	0.34493940785080	0.52947350602441
H	-2.33080286884643	1.35832408855198	0.47054497994566
H	0.73301316235796	-1.15407800183390	-2.53371698803792
H	0.90591423823976	-0.13679802303528	-4.83426495545427
H	0.78193091444543	2.37097049231790	-5.08881343407002
H	-0.11750681588619	4.71922000478888	0.39826839706603
H	0.07323682385579	5.92035038788483	-1.79775363839926
H	0.46598193618334	4.66914916948606	-3.92463841971647
H	-2.20486630739821	2.05645643272030	3.70831659593030
H	-3.05626987723004	-0.16629326166367	4.66104845707282
H	-1.71823271731422	-2.28665756174531	4.08397031433995
H	2.25464397962339	-4.68010019589707	2.49281119524476
H	4.49071260153305	-5.79119692859465	2.16839181739157
H	6.57680130774550	-4.44301596199014	1.90815239363427
H	7.67330307019735	-2.08376439400937	1.82791136296019
H	7.33442797854111	0.41356362604623	1.95762147230469
H	5.01687177257112	1.33218278776859	2.30285882772296
H	3.50412016096691	0.52029783549870	4.18431959557428
H	2.13694558570089	-0.13008614096453	3.84061176475442

Table S19 Geometry optimized coordinates of [**1**–H···H]$^{+‡}$ M_S = 4

Co	0.49309010074510	0.09464188627227	0.30422404681364
Co	2.22974699663876	-0.03511154764261	2.40095319121789
C	4.27232700843987	0.77553074312709	6.09776555789205
C	4.32549079079266	1.62489401490844	7.21455583916070
C	3.74022861951069	2.88768356782984	7.11929536487159
C	3.12036077565470	3.33034171480302	5.91942221588452
C	3.09727801246093	2.42341804406898	4.79512321095521
C	2.52240975172493	2.78279494636731	3.52822389077801
C	1.98855084781377	4.08431172107124	3.44916405924851

C	1.99365963141511	4.97389137110705	4.54918432912634
C	2.54702171537135	4.62122696795103	5.77343105562968
C	2.90690468301382	2.46897340188544	1.20336386625881
C	3.18348208804161	1.41809259135322	0.16318721257564
C	4.21831816220746	1.61350989971298	-0.75719262600276
C	4.69438458184531	0.52037855524234	-1.50204550895261
C	4.08466326288993	-0.73332672142570	-1.33053187572860
C	2.97083052907285	-0.84048246876433	-0.49420917785369
C	2.11381410272282	-2.07311259285799	-0.47774757989731
C	-0.26114543110173	-2.55408739317739	-0.10436631695663
C	-0.31221534915716	-3.94347668111107	-0.28841031343843
C	-1.56551638437839	-4.60410959409952	-0.24515701350111
C	-2.76185720346288	-3.91943225768405	-0.04152354065453
C	-2.74886096502125	-2.50417316286684	0.12387881377507
C	-1.48327273597473	-1.83685148897226	0.09881080224607
C	-3.90311411863817	-1.68710250239057	0.29827819729372
C	-3.76355516614856	-0.30932123026611	0.41267112500306
C	-2.47230414796480	0.27136982375591	0.38168200874191
N	2.52394904872116	0.22350554766340	0.25519151019739
N	2.58725990036012	1.90960010153773	2.43411289185555
N	3.68648003194779	1.19609030154137	4.96795774191174
N	-1.36961345570606	-0.47161037678796	0.25046642926757
N	0.84721592525599	-1.72500714280175	-0.09251480997750
O	3.11617192533880	3.65939175780370	0.95517513692891
O	2.52298188300763	-3.18165939250693	-0.84429205530904
C	0.67774127702899	-0.06449006533714	-2.67404611857578
C	0.77399845044202	0.51768974805812	-3.96138151919597
C	0.70261383790742	1.89839466087498	-4.09765809202981
C	0.51222515606288	2.71602686848300	-2.94669587694151
C	0.42163663682973	2.04630175715081	-1.68315279053532
C	0.20147539690937	2.76773265656044	-0.46444420405082
C	0.07028541480030	4.16357989140227	-0.52629464204183
C	0.17311288325293	4.82439214496773	-1.77593657792201
C	0.39220358888610	4.13525561374676	-2.96583650328084
C	-0.29837591584781	2.34606146013037	1.88351534215544
C	-0.52191558004595	1.14656696371209	2.76066922807263
C	-1.66565363500897	1.10932485708308	3.56413743822867
C	-2.13388339873789	-0.11801215550185	4.06424510352593
C	-1.41420104672897	-1.28260593410707	3.75558096546182
C	-0.21235714910107	-1.17636620996375	3.05378093407507
C	0.69944120878928	-2.34952789846945	2.88348850495945
C	3.07619343965959	-2.68617482482740	2.42169964728558
C	3.17013399301773	-4.08679121189614	2.38967568996751
C	4.43667153005177	-4.69236641423487	2.20650297867973
C	5.60690722926536	-3.94817905476897	2.06209706206243
C	5.55020735916276	-2.52674138140357	2.10947922826858
C	4.26966016503738	-1.91131746096169	2.29514591154572
C	6.67052315345610	-1.65652651216272	1.98188199285335
C	6.48769347408786	-0.28053480010490	2.05214355335464
C	5.18994307630983	0.24852340415487	2.24551161875526
N	0.24631168702262	0.02332791562157	2.55242995746484
N	0.13756023303040	1.94363916814052	0.65159191191105
N	0.52069433050287	0.68003728532403	-1.57633659767281
N	4.11158635479227	-0.54106047964540	2.35853936135715
N	1.94882505010491	-1.89970601242635	2.54483846531083
O	-0.63546381990902	3.48848371843415	2.22852028090037
O	0.34338389651132	-3.51740467869227	3.07286006129472
H	4.68065144196925	-0.24476954344463	6.08825901941731
H	4.81142620769831	1.28245235027401	8.13750115339133
H	3.75757530321356	3.56874342303732	7.98452249162893
H	1.54232410196452	4.40870462637609	2.50659853679744
H	1.54001154020615	5.96706594381639	4.41806392824178
H	2.55764353803595	5.31731420564734	6.62443772275201
H	4.66603348030905	2.61538361081210	-0.81387196868423
H	5.53645207394800	0.64399011307631	-2.19883215480364
H	4.40351328713123	-1.63653937730674	-1.86989303403135
H	0.62210193110735	-4.49084521833314	-0.45725441078413
H	-1.58666709216242	-5.69623759981725	-0.37957512470724
H	-3.72213651069919	-4.45509436270197	-0.01659413418408
H	-4.89870110228585	-2.15611300160466	0.32790335907209
H	-4.63870720670396	0.34493940785080	0.52947350602441
H	-2.33080286884643	1.35832408855198	0.47054497994566
H	0.73301316235796	-1.15407800183390	-2.53371698803792
H	0.90591423823976	-0.13679802303528	-4.83426495545427
H	0.78193091444543	2.37097049231790	-5.08881343407002
H	-0.11750681588619	4.71922000478888	0.39826839706603
H	0.07323682385579	5.92035038788483	-1.79775363839926
H	0.46598193618334	4.66914916948606	-3.92463841971647
H	-2.20486630739821	2.05645643272030	3.70831659593030

H	-3.05626987723004	-0.16629326166367	4.66104845707282
H	-1.71823271731422	-2.28665756174531	4.08397031433995
H	2.25464397962339	-4.68010019589707	2.49281119524476
H	4.49071260153305	-5.79119692859465	2.16839181739157
H	6.57680130774550	-4.44301596199014	1.90815239363427
H	7.67330307019735	-2.08376439400937	1.82791136296019
H	7.33442797854111	0.41356362604623	1.95762147230469
H	5.01687177257112	1.33218278776859	2.30285882772296
H	3.50412016096691	0.52029783549870	4.18431959557428
H	2.13694558570089	-0.13008614096453	3.84061176475442

Table S20 Geometry optimized coordinates of [**1**–H···H]‡ M_S = 3

Co	-0.13169111780863	0.10852808400173	-0.30141965380886
Co	2.27653858244548	-0.06200031444556	2.38460783966599
C	4.13377332494163	0.70618291972115	6.19684374271784
C	4.25721556615262	1.58868825736748	7.28239535591822
C	3.78137616254528	2.89127755951545	7.13546898194977
C	3.21002539343807	3.34084357465007	5.91421434445409
C	3.12286353856059	2.39989704374415	4.81913601506794
C	2.59906296452461	2.76719389812893	3.53251673431415
C	2.16979640371737	4.10357396895160	3.41205899982506
C	2.23191650553510	5.02461734601882	4.48257172071412
C	2.74613923043347	4.66838437230894	5.72239135937442
C	3.04286556680699	2.40277994903886	1.23285573627216
C	3.11098729875507	1.37391149904962	0.13870326748037
C	3.74370068913855	1.68107365308157	-1.07296268683203
C	3.89263743136771	0.66628314684624	-2.02785146631058
C	3.34601913282329	-0.59795054671425	-1.76240649276653
C	2.62538863924369	-0.80179870328057	-0.57428589739857
C	1.80990338798477	-2.07187537597808	-0.40167531806866
C	-0.51752556064375	-2.65969367320746	0.05659495205343
C	-0.38365817833073	-4.04079890158992	0.25666408531906
C	-1.50939214403087	-4.82413249900914	0.61623405410393
C	-2.77494366769631	-4.26991523479955	0.77059419236320
C	-2.96306911994916	-2.87405150658188	0.55131411534683
C	-1.82820136479649	-2.07576756267675	0.18675090446050
C	-4.21239701398008	-2.20508275993166	0.67793493091914
C	-4.29596998071531	-0.83507351331049	0.45183140860421
C	-3.12758160548795	-0.12256103570274	0.09622271862783
N	2.55717201260544	0.16179735500453	0.38126426723155
N	2.61050746598013	1.88162403325807	2.44331499918106
N	3.59684001359724	1.13345508276946	5.04709276250208
N	-1.93960476904134	-0.72211601991538	-0.03055045070024
N	0.49473045083179	-1.74371678162063	-0.21252159070274
O	3.41896309018188	3.55941104980261	1.01283844568221
O	2.32747919042310	-3.18669878728450	-0.51437415150137
C	0.54483897778781	0.40345304216390	-3.23360357624015
C	1.01985555072885	1.13170716254676	-4.34820867535017
C	1.27771082669048	2.49117153341376	-4.20877639130444
C	1.06844988976230	3.13294278623966	-2.95572495806280
C	0.59835029216532	2.31718704557141	-1.87281212122172
C	0.38107436788397	2.87071708444121	-0.55933780288076
C	0.57854970103989	4.24842054682921	-0.38740042749504
C	1.03750134682991	5.05133204533052	-1.46231712907366
C	1.29452014095490	4.52049213306443	-2.72082740229778
C	-0.23730296589700	2.24358206588830	1.71765977363604
C	-0.56506398892241	0.97466096318625	2.48479880343385
C	-1.86240807026876	0.84909091972836	3.01121641585761
C	-2.31960919890902	-0.39623706648809	3.46364908446902
C	-1.45290438398344	-1.49313651437129	3.38356107168895
C	-0.14206383844268	-1.28541941654165	2.94360020068915
C	0.84166938407483	-2.41640153160836	2.89522729639712
C	3.22571120699702	-2.68173309220364	2.45517285182217
C	3.37838627922061	-4.07636741013560	2.48052903434043
C	4.67765062071080	-4.63333694304413	2.40320719774375
C	5.82356810969412	-3.84466241929434	2.30934376869249
C	5.70671840896883	-2.42644327816358	2.30614719039544
C	4.39174934107237	-1.86002209564938	2.38515559226076
C	6.79829007671902	-1.51433561068534	2.23638551574417
C	6.55595808159872	-0.14642667367829	2.26483414420465
C	5.22797610400275	0.33286198251295	2.36331731731320
N	0.31411474808954	-0.06514172738609	2.53077064954230
N	0.02573579188052	1.93703463279418	0.41279293407420
N	0.34324474836664	0.97758830565230	-2.04361047694640
N	4.17732118512881	-0.49775713042373	2.41856596078322

N	2.05896970089661	-1.94298665044528	2.48479561074455
O	-0.34879867604051	3.35572988783100	2.24771374299156
O	0.54507684835959	-3.57789183014873	3.19364443621195
H	4.44767113027424	-0.34685296564302	6.22925105816282
H	4.70645097908324	1.24032521820735	8.22159874918873
H	3.84878736770751	3.60017962217967	7.97553561277728
H	1.76689742847292	4.43067894523554	2.45226219539671
H	1.86077311428317	6.04628540082826	4.31528216709035
H	2.80830880281585	5.38844352731734	6.55097208519939
H	4.12114788683308	2.70349602830369	-1.20848664433305
H	4.41680784029312	0.86045767365496	-2.97508547803280
H	3.41738099013032	-1.42851968676018	-2.47905768056496
H	0.61038448742050	-4.49055745052207	0.15049763704758
H	-1.36373305825503	-5.90117621137593	0.78843596952638
H	-3.63691768997726	-4.89096112653821	1.05530744157555
H	-5.10678791084217	-2.78386819689099	0.95619530250027
H	-5.25117993430862	-0.29832297726751	0.54107531168603
H	-3.15179304422910	0.96337043962760	-0.08778465196666
H	0.32202165013791	-0.67309448095366	-3.30277473325914
H	1.17104654050283	0.61664933646585	-5.30750923674917
H	1.64078858493889	3.08438654254336	-5.06224505271179
H	0.37232546158279	4.68597032900572	0.59547847034601
H	1.20058680359386	6.12449607026437	-1.28156011958718
H	1.65671402222571	5.15463109901216	-3.54331746605966
H	-2.50912127957706	1.73778927440489	3.00063502145858
H	-3.34445361877634	-0.51220362129644	3.84435640976529
H	-1.73368487386494	-2.51745971954081	3.66590858762256
H	2.48500352687461	-4.70612066664318	2.55384992183506
H	4.77903808848630	-5.72953959821478	2.41068258061847
H	6.82107690975473	-4.30238574959715	2.23880208407927
H	7.82540715695453	-1.90343698574845	2.16249803675093
H	7.37835866728092	0.58082619502480	2.21403810020675
H	5.01540883519050	1.41020724142399	2.39601168319151
H	3.34119285499448	0.44071104582410	4.28656339864515
H	2.14347378728487	-0.19453251946866	3.82980400337055

Table S21 Geometry optimized coordinates of [**1**–H···H]‡ M_S = 5

Co	0.39837373206761	0.08265374113620	0.24232421264616
Co	2.28019067735376	-0.12840128802058	2.30715422138677
C	4.00219884212342	1.08426360315175	6.11424467057916
C	3.87796209181428	2.03104874578633	7.13087969217305
C	3.27356613295933	3.26648985164022	6.85337333327493
C	2.79906047864524	3.56511384739943	5.53643775325600
C	2.94476586338752	2.56244967677424	4.51436551071433
C	2.51255709780799	2.78552685749476	3.17178463875144
C	1.97615590507977	4.05649628231040	2.87790608935382
C	1.81828944636236	5.04086558613669	3.86968109399310
C	2.21358136833440	4.80904493437286	5.18636178301211
C	3.14649965248829	2.21224728612639	0.95366797163699
C	3.35812988554294	1.07604481497492	-0.01149636460235
C	4.36584556193949	1.16455507717385	-0.97885333929252
C	4.70442709625225	0.01613346812121	-1.71518231190971
C	3.96498552299269	-1.15876985475729	-1.51268311478008
C	2.87560770558904	-1.13863904599205	-0.63171789066758
C	1.89305619030675	-2.28249684402976	-0.61879610468414
C	-0.45464938372135	-2.65014018689033	-0.10295013045571
C	-0.51260069810025	-4.04989811273484	-0.20662458788051
C	-1.73207090969915	-4.73259140796722	0.02260410407383
C	-2.90644433045037	-4.06096366266278	0.34937679806149
C	-2.89945954649204	-2.63960740344172	0.44664690095152
C	-1.67022630517528	-1.93860086161973	0.21039247348238
C	-4.04048733063048	-1.85100385890937	0.77090789240131
C	-3.93384506041969	-0.46709582491626	0.82869281169763
C	-2.68122136971943	0.14022692638713	0.56722358746651
N	2.60878388494809	-0.04797130124989	0.14912666933034
N	2.67868586286632	1.79541603491554	2.18174192056173
N	3.56135850790314	1.37170583222297	4.85854632399501
N	-1.59095540190730	-0.57121279815164	0.27926132755168
N	0.65428674004611	-1.83714851637671	-0.24082754079213
O	3.48447708890824	3.35796841478053	0.62220839733198
O	2.23172341145208	-3.41888113580486	-0.98261744467691
C	1.01889903278704	0.45118067056884	-2.73918554323329
C	1.38927098271325	1.21692906008056	-3.87143213397587
C	1.34065778200798	2.60377211090042	-3.79721651856538
C	0.92116165839802	3.23762214299633	-2.59204535405272

C	0.60355705814490	2.38204106088293	-1.48528667720398
C	0.20227718609477	2.92570276744932	-0.20939615251145
C	0.02619566003999	4.31885326112277	-0.11346585082405
C	0.33123804032466	5.15465745666731	-1.21540874464706
C	0.78999363842840	4.64645198577308	-2.42731546811288
C	-0.43954976792543	2.25231671867755	2.03220839987959
C	-0.53514438812073	0.99533474660139	2.85629215097989
C	-1.64320917421462	0.87997847291907	3.70871262512681
C	-2.05037739832235	-0.37776270790284	4.17338854520385
C	-1.30095598720220	-1.50165410095398	3.78459179645687
C	-0.13026669646218	-1.31684592610543	3.05000884462211
C	0.79159083038437	-2.47047790340720	2.79681422054050
C	3.14976940170286	-2.76970447757705	2.25376510328410
C	3.25352038581262	-4.16803790723159	2.16242047980895
C	4.52834935451402	-4.75692384220265	1.99094969558106
C	5.69828730828335	-3.99965559814253	1.92732304212044
C	5.63098073107064	-2.58168997935393	2.03550381458064
C	4.34004811022277	-1.98139651687349	2.18527260048538
C	6.75278285971276	-1.70211809720056	2.01407614672021
C	6.55371975942719	-0.33335735547163	2.15027313401003
C	5.24353137180573	0.18347549221848	2.27865658623789
N	0.28055977683592	-0.08364814224235	2.59360829371662
N	0.04440050284291	1.97691167581016	0.78139076693597
N	0.65301298095885	1.01788672923964	-1.58869257406327
N	4.16423826023808	-0.61612674168666	2.28384754449587
N	2.01657274963026	-1.99813611049315	2.41325616167631
O	-0.89730987882889	3.32623351567022	2.45666732798017
O	0.45688680546159	-3.64900861846286	2.97528394926801
H	4.44686960444799	0.08989413252917	6.25507278888966
H	4.25575242205407	1.79401254372708	8.13498696292112
H	3.16393136147069	4.02586700428441	7.64182996106180
H	1.67803262871944	4.27005264592070	1.84843609425473
H	1.36723340799765	6.00456062702773	3.59115993710144
H	2.08959558724235	5.57903265772115	5.96273700288330
H	4.88505211342129	2.12675224720654	-1.08439110398620
H	5.53249930682654	0.03966585701981	-2.43968228031227
H	4.15859912862514	-2.09329647413805	-2.05788390501365
H	0.40863991676081	-4.59049758603067	-0.45189156747097
H	-1.74013668547824	-5.83126508484644	-0.05215546155797
H	-3.84293330048098	-4.60818090166020	0.53547676443812
H	-5.00079801916544	-2.35163798160448	0.97201998594393
H	-4.80027712270434	0.16420093556666	1.07249736648376
H	-2.55773316893840	1.23482434950767	0.59871327519307
H	1.02153482156314	-0.65005319423833	-2.76300646301107
H	1.70257654675480	0.70420975989756	-4.79231777069530
H	1.61482535305475	3.22486971308044	-4.66441516036419
H	-0.34594274726436	4.73215528755254	0.83138199569217
H	0.20558330606768	6.24251404335517	-1.09806136756036
H	1.03871008530533	5.31207948986611	-3.26739749204188
H	-2.20843012065787	1.80237544466664	3.90411306358003
H	-2.94890208896645	-0.48507105611341	4.79881172127029
H	-1.55869717771416	-2.53315010333286	4.06283293670202
H	2.33945541750251	-4.76901218049947	2.21932617358575
H	4.59181597567384	-5.85285477582366	1.90352592543450
H	6.67849359931279	-4.48192974593067	1.79675615982201
H	7.76511095697350	-2.11794526532008	1.89424741953168
H	7.39876174470286	0.36964097413956	2.14379461310625
H	5.05974789598972	1.26275652701039	2.36020258612692
H	3.41966868513418	0.60251899134901	4.16298783141991
H	2.24015285214677	-0.18888724454058	3.76332306910844

Table S22 Hydrogen atom optimized coordinates of **3**

Co	-0.00041702744629	0.00030030192130	-0.00159893009189
Co	0.00041925559423	-0.00003394871987	4.01763098044622
N	1.50114603189948	-0.09522025805663	1.28635074180019
N	0.20160695712638	-1.87861705735261	-0.01168720906047
N	-1.39078218107777	-0.35809017790109	-1.30697948456695
N	-0.39001923216603	-1.74804594715225	3.18179681254102
N	-1.47077708551069	0.25036091437564	1.29327782008912
N	0.33907210733731	1.80034081622207	3.27992023687971
N	1.36868832145204	-1.06421291427761	5.03341780493110
N	-0.09550057514626	1.87830239320932	-0.08625298079778
N	-1.41154030877385	0.94655578212103	5.07328352041001
N	1.29740233299026	0.21730334544154	-1.42669094541169
O	-1.94480878541843	-3.03463407039253	2.01752328295971

O	1.84487533409837	-3.47742470966071	0.30737794708640
O	1.75723973756810	3.21576400464489	2.08116285342548
O	-1.49433076502315	3.66948518589935	0.35647702157758
C	-1.50312613011576	-1.70384550465963	-1.57617802627642
C	2.08425842329105	-1.31333456705851	1.24032812356292
C	-2.11044626943219	-0.65887288539331	2.05060531791192
C	2.09465206609875	0.85848075174000	2.03750658957614
C	0.88326069580571	2.48190205402345	-0.87284644937988
C	1.35003575253350	2.11820060084328	2.43252630418533
C	1.37548459408535	-2.37976946672607	4.64404696190151
C	1.37155268309759	-2.36484233892517	0.46447895076095
C	-1.44815842076317	-1.97704658979109	2.38758790930549
C	0.43504131214484	-2.77727530210455	3.65447955585606
C	-2.20006845892655	0.48315178537365	-1.90150116189960
H	-2.06285130074408	1.55302969270528	-1.68369309741217
C	-0.52176141852834	2.77100143949198	3.80595009306892
C	-0.61465117771566	-2.56381127249262	-0.90608437926356
C	-1.16546508244831	2.48920911432836	0.45110045199999
C	-1.97377047050086	1.50133817342524	1.21521650088422
C	1.21209423949295	3.81269059207463	-0.95342420370480
H	0.64045075765372	4.54694039177997	-0.37521899026494
C	1.89344041419778	-0.69462052595496	-2.17477080683578
H	1.59448574811984	-1.74172772963883	-2.02380511711279
C	-2.46669761513698	3.11426187097557	5.28612336175735
C	1.62572659722655	1.54061997965137	-1.60559921904587
C	3.25300075356431	-1.60870468264972	1.89278041096720
H	3.62350341799856	-2.64131719468168	1.81812775069366
C	0.46291884104930	-4.08965523325378	3.22544459861085
H	-0.24574511061316	-4.41170966242010	2.45432921732836
C	-0.59435393396605	4.10368902759942	3.45734602964866
H	0.11986227497193	4.50145399136834	2.72918450433252
C	2.28414153829780	4.19938819555336	-1.77920512079169
H	2.55092360773050	5.26687118097853	-1.81637743195689
C	-2.49116012104457	4.47372935072373	4.90976150306147
H	-3.26227206261910	5.13079524401208	5.34019028906414
C	-2.44546876069454	-2.19813782177160	-2.49177308940524
C	-1.47193778769588	2.27831333792119	4.74817684279922
C	-1.58176815949470	4.94036643333531	4.02276107655316
H	-1.60272633788267	5.99619971138465	3.71298223734629
C	-3.14353556759130	1.86918072289134	1.85404048866118
H	-3.45935535633655	2.91845088812518	1.76470312252197
C	1.35710611756108	-5.00974329841770	3.81320202459044
H	1.33155207013287	-6.05305990623762	3.46403132421901
C	-2.31411817189805	0.44357442338658	5.90438950624165
H	-2.22489871806293	-0.63218995401692	6.12893015945670
C	2.88462354516705	-0.37245592138395	-3.11753026425411
H	3.33700729422352	-1.18208097958601	-3.70592123312041
C	-3.32521618433490	-0.35932518775435	2.66262298781396
H	-3.82134538862559	-1.15028290095127	3.24286885030739
C	3.33210827765609	0.64210684203390	2.62425111303721
H	3.78816650719379	1.47312887885371	3.18160116152150
C	-3.21217369277245	0.05547531250014	-2.77915616515722
H	-3.87407270456067	0.80959978959723	-3.22712201278343
C	2.22339480827088	-0.65167481851955	5.95364430320412
H	2.16542003404415	0.41202954051584	6.23760883439241
C	-3.39482279745608	2.53643213471912	6.16956843626042
H	-4.18119943909022	3.17185528258471	6.60709490164997
C	-3.84390667134362	0.90942130864640	2.56394642123807
H	-4.78944708790173	1.16683070542865	3.06425141261659
C	2.65685977637098	1.93369638936100	-2.49442109326385
C	-0.63691773958247	-3.90809145465642	-1.18673154183472
H	0.06683674462032	-4.57409361540541	-0.67525811994193
C	-2.44051695771428	-3.58138460464201	-2.76173914708298
H	-3.16810883407825	-3.98082345784968	-3.48455694699141
C	-3.32721705569294	-1.26609820182275	-3.07525213886656
H	-4.09872772775247	-1.62344440754046	-3.77596121626829
C	3.91718132449398	-0.60069579943460	2.56220610413156
H	4.87994037821987	-0.79269779394007	3.05840476932311
C	2.29164516088692	-3.29226183680527	5.20525045579293
C	-1.55540738978977	-4.40010735427942	-2.13435137411726
H	-1.55503506216339	-5.47867354933172	-2.35400946088696
C	2.23487782113259	-4.63937497576475	4.77466021658787
H	2.92954946718702	-5.36699774350878	5.22181020908118
C	3.27355982121225	0.92828688434981	-3.25584302490507
H	4.07067942472743	1.20574625047412	-3.96418489479519
C	2.97510286439734	3.31046054453353	-2.54239282236444
H	3.78970227266693	3.64381718949016	-3.20291297500541
C	3.16040976058086	-1.49953572316387	6.53677884091703
H	3.85275609544102	-1.09762265349497	7.29079711829135

C	3.20933408959738	-2.79987582458184	6.14965049217599
H	3.94447932548921	-3.49302816754930	6.58882830255685
C	-3.32143138858930	1.21103865384213	6.47031293796354
H	-4.03849826247775	0.73699258721547	7.15618297704079

Table S23 Geometry optimized coordinates of **3** $M_S = 4$

Co	0.02743308883813	-0.02051565872627	0.18593370047365
Co	-0.00684617184650	-0.04617345091731	3.88749160295933
N	1.52873006467137	-0.22768436423352	1.45768044026255
N	0.12458800995194	-1.91858187321925	0.12366175474315
N	-1.30419358700945	-0.24475571192371	-1.20843941070780
N	-0.45488695156062	-1.85412114922468	3.25268646874241
N	-1.50188852486438	0.17071723412489	1.42711429993198
N	0.44322462203627	1.77099292356759	3.28514415611119
N	1.45772980233318	-0.98345723435422	4.87700160113515
N	-0.06829985743940	1.87804137598516	0.14554027417831
N	-1.49808590388452	0.87127549651310	4.85826259024249
N	1.38952586715683	0.22060873628350	-1.17591271234612
O	-1.98187901878640	-3.19844183006587	2.06885649075383
O	1.58040717250234	-3.69429668941272	0.55585906034737
O	1.99073967836562	3.13454142946355	2.15053204569311
O	-1.53417065378580	3.64807071851533	0.56618970497603
C	-1.56635184025060	-1.57443617016843	-1.45530262393386
C	2.05717093723057	-1.48179542987572	1.37751059008462
C	-2.19764545362546	-0.80320701797615	2.06981826505890
C	2.21111545445555	0.73909494422802	2.12558235666540
C	0.85208831081966	2.49290950848198	-0.68521744746428
C	1.52114793449127	2.04534255079946	2.48803674376772
C	1.47850885706513	-2.32399945167549	4.56962734570371
C	1.23567279069353	-2.51181071249369	0.65481521680014
C	-1.51684698640099	-2.11474439989153	2.43016715154771
C	0.42846046116058	-2.82628303215527	3.72031070077145
C	-1.96064667921556	0.70105664646408	-1.88644730834740
H	-1.70184307650190	1.74736463707402	-1.67642698498932
C	-0.45341174556611	2.73333845550734	3.74744096453442
C	-0.77735158607899	-2.52309163079551	-0.73445457730918
C	-1.19148927479496	2.46443924337575	0.65862816885201
C	-2.02850568786443	1.42592777838190	1.35054986669350
C	1.05347632656845	3.86285551028337	-0.90207263652020
H	0.42677359060191	4.58394155673217	-0.36552138054886
C	2.06040795524430	-0.71700978979273	-1.85116064744017
H	1.79589035399338	-1.76550470003490	-1.66030470079316
C	-2.57217011865696	3.06726779973891	5.04136137284369
C	1.65709054636344	1.55319671014882	-1.40013605735874
C	3.30162779799189	-1.80409477132510	1.92744356285451
H	3.63452715287093	-2.84894906545891	1.85004696125079
C	0.42663807819060	-4.19352834967405	3.40947825421159
H	-0.37286391609531	-4.58393991712397	2.76943223158114
C	-0.45104764042936	4.10441233111800	3.45444620661824
H	0.36016348939909	4.50665165641146	2.83664762967490
C	2.06166174587456	4.28287052676673	-1.80565017282278
H	2.21440674610620	5.36260848096224	-1.95471735408491
C	-2.54102652043832	4.45055295291716	4.69805561573810
H	-3.34443255618644	5.11236680418292	5.05386780937129
C	-2.55293956573665	-1.98879892672857	-2.40824441588330
C	-1.51857474251860	2.21553963933198	4.56814730872360
C	-1.49399691053299	4.94135617992479	3.92668293899328
H	-1.46938876543996	6.00831097294756	3.65910761907092
C	-3.28467153503326	1.74192822154167	1.87672607376557
H	-3.61620287879850	2.78754468955723	1.80388055949971
C	1.45562011037991	-5.04121636111770	3.89302121198441
H	1.43173598538026	-6.10468875575854	3.61193953930371
C	-2.47522537299215	0.32989811750092	5.59317848336202
H	-2.40297501202206	-0.75236997752983	5.78582489919991
C	3.07377943624020	-0.37725916852235	-2.78006572602530
H	3.60079417094266	-1.18522567007468	-3.30572725423370
C	-3.50567634320208	-0.56912342915882	2.52776580010937
H	-4.04852422476780	-1.40409525492533	2.99238867143452
C	3.50965112804676	0.50001538211875	2.60706703158098
H	4.04253044698534	1.32984033576045	3.09204581293393
C	-2.95305176286190	0.37285094216670	-2.84179767410825
H	-3.46811748610610	1.18734580672310	-3.36918641908630
C	2.42167427348652	-0.45620323180374	5.63919840511388
H	2.35003563973636	0.62370152358417	5.84475541730726
C	-3.59453343970237	2.46135112268047	5.82376329983348
H	-4.41764427011365	3.08337731103642	6.20868946697579
C	-4.05605378748175	0.71681003288529	2.44022519019149

H	-5.06479947103420	0.92049492686183	2.82749652794551
C	2.66382984818215	1.97904656342289	-2.32660153292950
C	-0.97496310727505	-3.89027020358538	-0.97104768335643
H	-0.36063653727083	-4.61798223272791	-0.42919209585074
C	-2.74292674524808	-3.38625756696540	-2.60760314647545
H	-3.50132183622035	-3.72818043614851	-3.32691689547477
C	-3.25570436406843	-0.95913192860683	-3.09623613910793
H	-4.02744692920782	-1.22948813702808	-3.83378720679884
C	4.06146113101397	-0.78532022208301	2.51724526662320
H	5.06190388528397	-0.99355015946794	2.92314233467897
C	2.51833865843362	-3.18653569267289	5.05381915709182
C	-1.96328993883695	-4.29904782416920	-1.90132027019912
H	-2.11350367200101	-5.37685812032239	-2.06588234262482
C	2.48813041787769	-4.56521159984956	4.69264114250065
H	3.28097363165712	-5.23527011814335	5.05667942386993
C	3.38187188461396	0.95772398419436	-3.01119176451195
H	4.16987563716970	1.23692648189588	-3.72797103783406
C	2.85736093000348	3.37879193821980	-2.50522740244772
H	3.63135041004546	3.72949247201908	-3.20338504566493
C	3.48156727184830	-1.23559995897496	6.15593733643261
H	4.24898686947274	-0.75906674452452	6.78254704580728
C	3.52666420911320	-2.59550390633866	5.86524979411174
H	4.33861011517060	-3.22635617861865	6.25942257266082
C	-3.54925547812713	1.09792860548575	6.09767980623482
H	-4.32762499817851	0.60991294847924	6.70151872476436

Table S24 Geometry optimized coordinates of **3** $M_S = 2$

Co	0.03150310733656	-0.01591702716815	0.21013233944931
Co	-0.00911880299257	-0.03793596078882	3.19822574685610
N	1.60686536984820	-0.17890984941793	1.50134829951632
N	0.18469434916300	-1.91196876781962	0.19253741549188
N	-1.29398004665538	-0.30486030487298	-1.16388093019316
N	-0.64178181498173	-1.84371890479578	3.08497819706116
N	-1.57738718903915	0.15759712738567	1.45996374545146
N	0.60159575000196	1.77995366940594	3.13805266721825
N	1.31600935313018	-0.79995104508320	4.40377033064359
N	-0.10535166217529	1.88161843861430	0.19368871944111
N	-1.36469825817241	0.68238364975481	4.39676273320707
N	1.38946654931620	0.26750279979170	-1.13549334826455
O	-2.26974715339067	-3.25808470986366	2.16472071119356
O	1.58320547396868	-3.68801333288748	0.78812511210462
O	2.22417245726245	3.23664838438261	2.27512942700337
O	-1.48609086042593	3.67334608581026	0.78331211269515
C	-1.54850760615837	-1.64470026164311	-1.35521527210173
C	2.16389010538746	-1.42200236168707	1.35265336573473
C	-2.39207313014838	-0.87559481274108	1.84270897378411
C	2.39460256676658	0.86174960212527	1.91852848841135
C	0.84268164640382	2.52353633348325	-0.58515323306879
C	1.72198717037317	2.11796663588899	2.42335951157020
C	1.22949605764326	-2.17465798278980	4.47151339110533
C	1.27951619744190	-2.49103146570009	0.77782219679667
C	-1.75204289222284	-2.15052432957930	2.34189154063844
C	0.14624372351116	-2.77717721072254	3.75345404574324
C	-1.97210683660223	0.61332746529435	-1.85752812120684
H	-1.71268902697671	1.66639817806984	-1.68249895864813
C	-0.21234361454888	2.68843017749919	3.80954746781269
C	-0.73420203001617	-2.55862851131818	-0.61614906973790
C	-1.20070044356535	2.47214484451322	0.76580987299561
C	-2.11196624115528	1.41053877601066	1.31236637068821
C	1.06465194198620	3.89927814877385	-0.74018380490379
H	0.43530923892308	4.60461104092646	-0.18577090954774
C	2.06977911932950	-0.65264562746181	-1.82478438656742
H	1.79555233957698	-1.70426867450763	-1.66529151090887
C	-2.26539968218423	2.81076711193215	5.23368859602828
C	1.66371695275960	1.60567365707013	-1.31150279560300
C	3.51220168613182	-1.66300026445209	1.62926564809592
H	3.87414250108809	-2.69606398652195	1.52821959669203
C	0.01757102156772	-4.17354401894360	3.77779460627634
H	-0.80510170532220	-4.63911808631044	3.22339951087487
C	-0.10593881946498	4.08588564994237	3.86206159141065
H	0.71822327281285	4.57466552640407	3.33036413170704
C	2.09993842111741	4.34337496312200	-1.60047904606279
H	2.26720522803816	5.42658772627749	-1.70063532721758
C	-2.12938030353093	4.22941932386007	5.25833099070118
H	-2.86321079236117	4.83081117729400	5.81488958929950
C	-2.55639282109863	-2.10012361717334	-2.26547030332881
C	-1.29876531486147	2.05621310818464	4.49564272255335

C	-1.07158275391115	4.83330862279643	4.58296491563184
H	-0.97432567114600	5.92965147922118	4.60362382908478
C	-3.46241657691776	1.66979843021541	1.56056727441825
H	-3.80531948836393	2.70959036410228	1.46265270527884
C	0.95963769610738	-4.94940251409492	4.49974538399408
H	0.84569948985262	-6.04432567018420	4.49745395874511
C	-2.33140938553916	0.01644466706696	5.03913120498495
H	-2.32455067110733	-1.07916640741057	4.93675786721075
C	3.10328012232981	-0.28729606482238	-2.72121021077692
H	3.63797982377352	-1.07946472149620	-3.26300450522492
C	-3.77274935856064	-0.69364972259820	1.97540729705787
H	-4.37985095956638	-1.57056252588410	2.24213079622263
C	3.77443814928439	0.69985067044959	2.08120532215769
H	4.36082397771683	1.58223638504789	2.37482196107066
C	-2.98319275407592	0.24325956053940	-2.77730043371227
H	-3.51761614032069	1.03324754335801	-3.32255532941902
C	2.28288755699049	-0.16181148125752	5.07290534823757
H	2.29219508390538	0.93574033275180	4.99321918688233
C	-3.29274911877616	2.07510806444577	5.89199072928454
H	-4.05721299695174	2.61465394274439	6.47243984112636
C	-4.31716061430544	0.59672639846356	1.85601591919671
H	-5.39381032164339	0.76423331740532	2.00851024825570
C	2.69644601466090	2.05624942613674	-2.19598893482275
C	-0.93426745723265	-3.93558350271016	-0.78965148332327
H	-0.30862498198632	-4.63816251889191	-0.22747456042666
C	-2.74410639407196	-3.50527087049495	-2.40309927206784
H	-3.51831978384886	-3.87983248615342	-3.08841350737969
C	-3.28220293840773	-1.09995919497573	-2.97327363836555
H	-4.07041291706417	-1.40217005633987	-3.68046244864227
C	4.34242046723729	-0.58011784304454	1.95717539754431
H	5.41798673779905	-0.73210676520363	2.13171914315636
C	2.17284073941063	-2.95832696764183	5.20911424058491
C	-1.94355146050553	-4.38485786280106	-1.67765234431377
H	-2.09421537868252	-5.46914236259630	-1.79168220134527
C	2.01533870198001	-4.37489924615008	5.20336450249892
H	2.73151902370958	-4.99812016451287	5.75891001134082
C	3.42401166977973	1.05346533071938	-2.89814293105145
H	4.23063498691518	1.35186678818411	-3.58584267097239
C	2.90483495312832	3.46009103833940	-2.31654290268012
H	3.69850632434431	3.83064972224094	-2.98147632633252
C	3.24814196170782	-0.86296116994015	5.83494715170231
H	4.02417140084422	-0.29358391523245	6.36586931156837
C	3.20138792446884	-2.25146103432089	5.89690034333316
H	3.94885562169795	-2.81388673023728	6.47770983185417
C	-3.31709800290960	0.68753434359975	5.80186223284968
H	-4.09151685458427	0.09602688359929	6.31049499669141

The structure of **1** was optimized using the GGA functional BP86 as this gives better metal-ligand and intraligand bond distances than hybrid functionals. The optimization was carried out assuming two low-spin Co(II) ions (M_S = 3) and two high-spin Co(II) ions (M_S = 7). The former was the more stable structure as the high-spin variant was a sizeable 24.8 kcal mol^{-1} higher in energy. However, the optimized bond distances in the high-spin structure are best match the experimental values with the variation not exceeding 0.06 Å (Table S25). The crystallographic symmetry is largely preserved in the optimized structures. Furthermore, this is consistent with the room temperature magnetic moment for **1** of 4.8 μ_B, which results from uncoupling of symmetry-equivalent magnetic orbitals at room temperature. The disparity is a consequence of the known functional dependence on spin states,[20] where the low-spin state is favored by the BP86 functional as it is devoid of Hartree-Fock (HF) exchange. The introduction of 20% HF exchange in the B3LYP hybrid functional results in a re-ordering of the relative energies with the M_S = 7 structured preferred over the M_S = 3 structure by 11.1 kcal mol^{-1} (Table S27). The salient difference between the optimized and crystallographic structures is the separation of the Co centers. In both optimized structures this distance is *ca*. 0.4 Å shorter than observed experimentally, and therefore wholly a consequence of lattice forces in the crystal packing. This highlights a large degree of flexibility in these pentadentate ligands as they intertwine around two metal ions.[21]

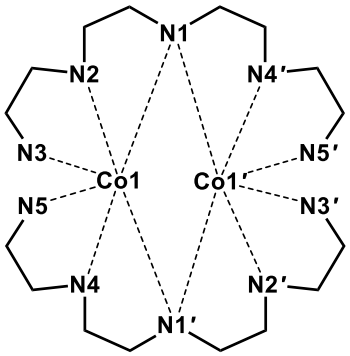
The electronic structure of **1** is calculated using the broken-symmetry method using the aforementioned B3LYP hybrid functional, the large def2-TZVPP basis set for all atoms, and applying ZORA to account for relativistic effects. The BS(3,3) calculation positions three spins on one redox center and three of opposing spin on the other to give a net S = 0 ground state brought about by antiferromagnetic coupling between the two spin centers (Table S26). For **1**, the metal d orbitals are the highest occupied (HOMO) and therefore the site for the unpaired spins giving two coupled high-spin Co(II) ions. The strength of the coupling, J, is estimated using the difference in energies of the high-spin (parallel alignment of spins) and broken-symmetry (anti-parallel alignment of spins) according the eq. S1, assuming the Heisenberg-Dirac-van Vleck Hamiltonian

(eq. S2) is valid.[17] The estimated coupling constant of –5.5 cm^{-1} for the optimized structure and a slightly large coupling of –11.7 cm^{-1} for the crystallographic structure are commensurate with the 4.80 μ_B magnetic moment that approximately equates to two pairs of uncoupled spins at room temperature. This is reflected in the MO scheme showing three pairs of symmetry matched magnetic orbitals with weak overlap integrals, S < 0.1 (Fig. S20). Each singly occupied orbital (SOMO) has >80% Co d-character with the remaining composed of the first coordination sphere nitrogen donors of L^{2-}. The Mulliken spin density analysis reflects the degree of covalency produces by the high-spin Co(II) ion as opposed the corresponding low-spin center (Fig. S21). For the BS(3,3) solution, +2.7 spins are located on Co1 with a counterbalancing –2.7 spins on Co2. These values are below the expected three spins for a high-spin Co(II) ion with 0.3 spins transferred to the first coordination sphere for each Co center through the σ-bonds. The ligand spin density is confined to the terminal nitrogen atoms from the amido and quinoline groups of L^{2-}, and visually absent on the bridging pyridine groups. This is the result of cancellation of positive spin deferred from Co1 with the negative spin derived from Co2 in addition to the longer bonds to the bridging pyridine groups (~2.4 Å *cf.* ~2.1 Å for the terminal *N*-donors). In the corresponding BS(1,1) calculation, there is 0.95 spins on each Co center of opposing sign, with only 0.05 spins distributed on the *N*-donor atoms of the first coordination sphere. The BS(2,2) calculation converged to the BS(1,1) solution, as evidenced by the distribution of spin density. This also reveals an inability for the calculation to distinguish one Co center from the other to produce a solution with mixed spin states. The BS solution is *ca*. 6 kcal mol^{-1} less favorable that the high-spin (M_S = 5) solution (Table S26).

The larger *J*-value for the crystallographic structure of **1** stem from the greater intermetal separation between high-spin Co(II) ions that reduces electronic repulsion leading to a 21.7 kcal mol^{-1} stabilization of the crystallographic structure over the optimized structure. The spin ladder was calculated on the crystallographic coordinates and shows the S = 0 ground state resides 304 cm^{-1} below the first excited state which results from flipping one spin on the second Co ion to give an S

= 1 species (Table S26). The next excited state is generated by a second spin flip to give an S = 2 species requiring a further 184 cm^{-1} of energy (Fig. S22).

Table S25 Comparison of experimental and geometry optimized[a] bond lengths (Å) in **1**



	Exptl	M_S **= 7**	M_S **= 3**
Co(1)–N(1)	2.402(8)	2.366	2.055
Co(1)–N(1′)	2.430(1)	2.376	2.070
Co(1)–N(2)	2.010(9)	2.017	1.906
Co(1)–N(3)	2.126(1)	2.084	1.961
Co(1)–N(4)	2.021(8)	2.015	1.904
Co(1)–N(5)	2.110(8)	2.091	1.960
Co(1′)–N(1)	2.430(1)	2.355	2.069
Co(1′)–N(1′)	2.402(8)	2.368	2.055
Co(1′)–N(2′)	2.010(9)	2.019	1.906
Co(1′)–N(3′)	2.126(1)	2.093	1.960
Co(1′)–N(4′)	2.021(8)	2.019	1.904
Co(1′)–N(5′)	2.110(8)	2.089	1.959
Co(1)···Co(1′)	3.183(1)	2.770	2.714
Total energy[b]	+34.7	+24.8	0.0

[a] Optimized at the BP86/def2-SVP level; $M_S = 2S + 1$. [b] Total energy relative to the most stable structure, in kcal mol^{-1}.

Table S26 Comparison of energies for calculated ground state electronic configurations[a] for **1**

	Geometry optimized[b]			Crystallographic structure[c]			Configuration[g]
	Total energy[d]	***J***[e]	**Relative energy**[f]	**Total energy**[d]	***J***[e]	**Relative energy**[f]	
BS(3,3)							
HS	–151627.06815			–151628.00376			↑↑↑ ↑↑↑
BS	–151627.07430	–5.5	0.0	–151628.01685	–11.7	0.0	↑↑↑ ↓↓↓
BS(2,2)							
HS	–151626.82181			–151627.65811			↑↑↑ ↑↑↓
BS	–151626.59327	+383.8	+11.1	–151627.32424	+561.9	+16.0	↑↑↓ ↑↓↓
BS(1,1)							
HS	–151626.56755			–151627.29730			↑↑↓ ↑↑↓
BS	–151626.59317	–204.6	+11.1	–151627.32073	–189.0	+16.1	↑↑↓ ↑↓↓

[a] Based on broken symmetry calculations at the B3LYP/ZORA-def2-TZVPP level; HS = high-spin; BS = broken-symmetry. [b] Optimized at the BP86/def2-SVP level for M_S = 7. [c] Hydrogen atom optimization at the BP86/def2-SVP level for M_S = 7. [d] In eV. [e] Exchange coupling constant calculated using eq. S1, in cm^{-1}. [f] Total energy relative to the most stable structure, in kcal mol^{-1}. [g] Spin configuration for each high-spin Co(II) center.

Table S27 Comparison of energies for calculated ground state electronic configurations[a] for **1**

	M_S = 7 optimized [b]			M_S = 3 optimized [c]		
	Total energy [d]	***J*** [e]	**Relative energy** [f]	**Total energy** [d]	***J*** [e]	**Relative energy** [f]
BS(3,3)						
HS	–151627.06815			–151624.74694		
BS	–151627.07430	–5.5	0.0	–151624.93812	–169.3	+49.3
BS(1,1)						
HS	–151626.56755			–151626.15270		
BS	–151626.59317	–204.6	+11.1	–151626.59107	–2699.3	+11.1

[a] Based on broken symmetry calculations at the B3LYP/ZORA-def2-TZVPP level; HS = high-spin; BS = broken-symmetry. [b] Optimized at the BP86/def2-SVP level for M_S = 7. [c] Optimized at the BP86/def2-SVP level for M_S = 3. [d] In eV. [e] Exchange coupling constant calculated using eq. S1, in cm^{-1}. [f] Total energy relative to the most stable structure, in kcal mol^{-1}.

Table S28 Calculation of spin ladder for **1**[a]

	Total energy / eV	**Relative energy / kcal mol^{-1}**	***J*** **/ cm^{-1}** [b]	**Electronic configuration** [c]	**Spin state**
M_S = 1	–151628.01685	0.0	–11.7	↑↑↑ ↓↓↓	S = 0
M_S = 3	–151627.67515	+7.9	+304.3	↑↑↑ ↑↓↓	S = 1
M_S = 5	–151627.65569	+8.3	+488.3	↑↑↑ ↑↑↓	S = 2

[a] Based on broken symmetry calculations at the B3LYP/ZORA-def2-TZVPP level on hydrogen atom optimized crystallographic coordinates. [b] Exchange coupling constant calculated using eq. S1. [c] Spin configuration for each high-spin Co(II) center.

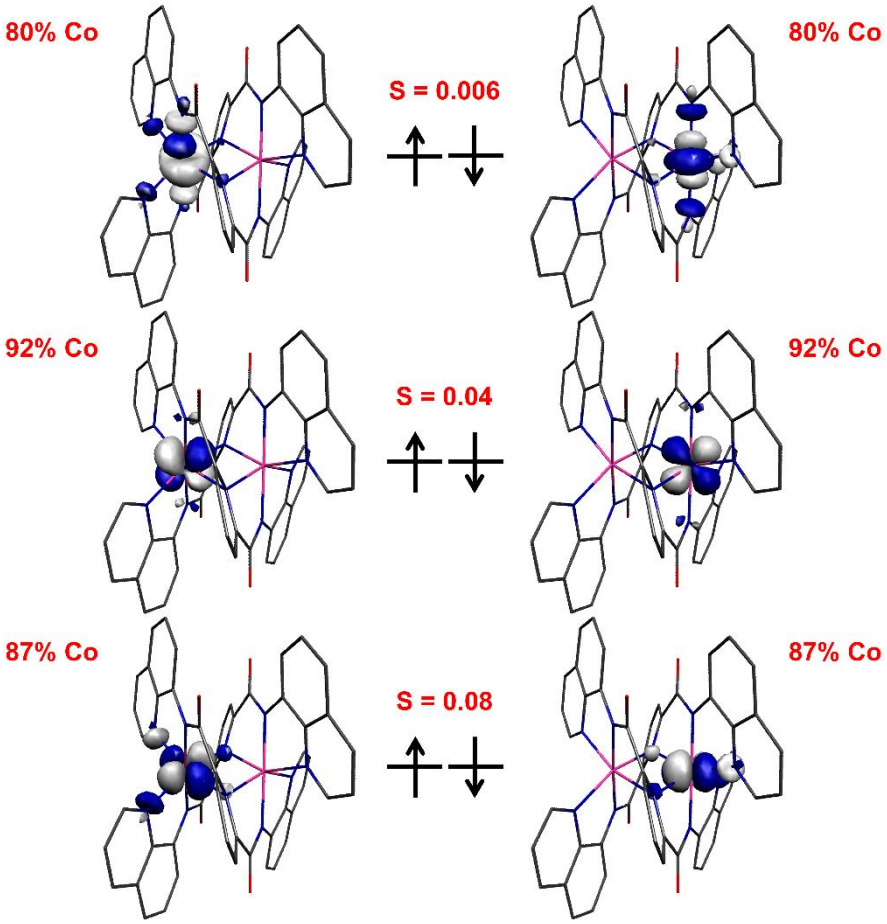


Fig. S20 Qualitative MO scheme depicting the corresponding pairs of magnetic orbitals in **1** as derived from a BS(3,3) DFT calculation.

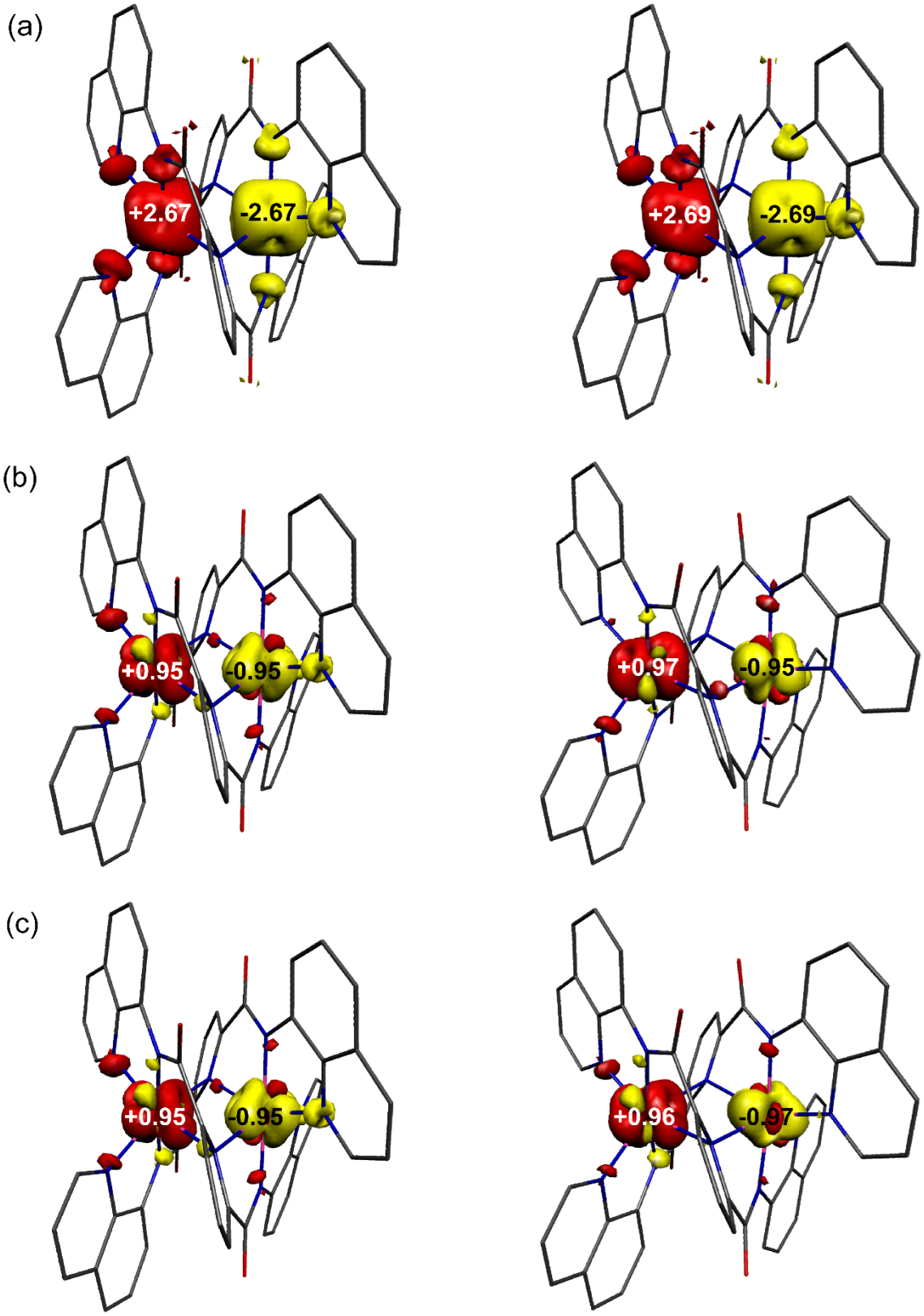


Fig. S21 Mulliken spin population analyses for the (a) BS(3,3), (b) BS(2,2), and (c) BS(1,1) solutions on the geometry optimized M_S = 7 (left) and crystallographic (right) structures from B3LYP/ZORA-def2-TZVPP calculations (red: α-spin; yellow: β-spin).

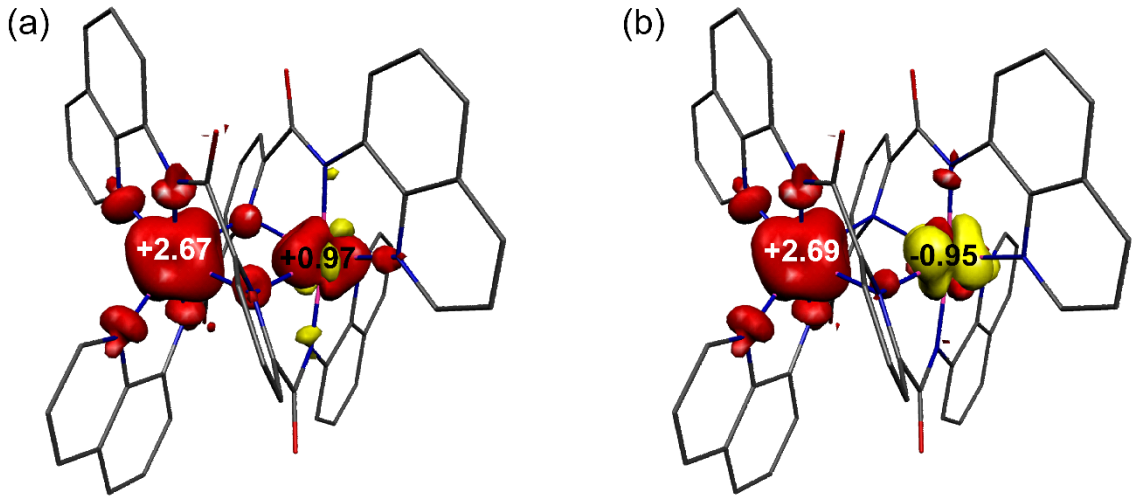


Fig. S22 Mulliken spin population analyses for the (a) S = 2, and (b) S = 1 states for the spin ladder for **1** from B3LYP/ZORA-def2-TZVPP calculations (red: α-spin; yellow: β-spin).

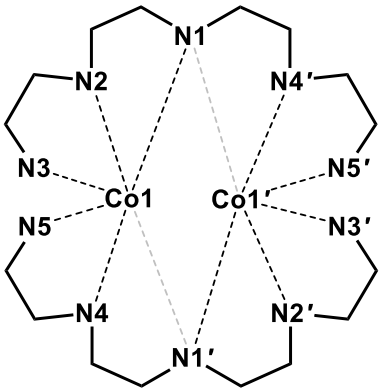
One-electron reduction to give [**1**]$^{-}$ produced a molecule with same equivalency of the Co ion as in neutral **1**. This is borne out in the optimized bond distances irrespective on the multiplicity used in the calculation where the first coordination sphere *N*-donors are the same for each Co center (Table S29). The optimization was carried out starting with the crystallographic coordinates of **1** and optimized on M_S = 2, M_S = 4 and M_S = 6 spin multiplicities for the formally $Co^{II}Co^{I}$ mixed-valent species, which encompasses the three spin state permutations for combining high-spin (S = $^3/_2$) and low-spin (S = $^1/_2$) Co(II) with a high-spin (S = 1) and low-spin (S = 0) Co(I) ion. The M_S = 2 optimized structure – both low-spin Co ions – was favored over the structure bearing at least one high-spin center by 18.3 kcal mol^{-1}. Considering the impact of HF exchange on the relative energy for **1**, it is important to note that low-spin solution is also preferred when comparing energies calculated on the optimized coordinates with the B3LYP hybrid functional (Table S30). The single-point and BS(2,1) solutions for the M_S = 2 optimized structure is at a minimum 6 kcal mol^{-1} more stable than any other configuration, which is outside the generally accepted error for DFT of 3–5 kcal mol^{-1}.[22] The salient effect of increasing the spin multiplicity is the concomitant decrease in the Co···Co separation from 2.944 Å to 2.517 Å in the M_S = 6 optimized structure. The higher spin multiplicities both contain six-coordinate Co centers with both pyridine groups of the ligand bridging the metal ions. In the M_S = 2 optimized structure, the distance to the pyridine nitrogen atoms is beyond that considered for bonding, and therein each are five-coordinate. For each of these Co centers, the four equatorial bonds are markedly shorter (*ca*. 0.12 Å) than the "apical" *N*-donor from the pendant quinoline group. This reflects the spin state at the Co ion where the d_{z^2} orbital is partially occupied in a low-spin Co(II) center with pseudo-octahedral geometry and the $d_{x^2-y^2}$ is unoccupied; both of these constitute the σ* MOs in this complex. The loss of the sixth *N*-donor atom stabilizes the d_{z^2} orbital relative to the $d_{x^2-y^2}$ orbital and therein preference for the low-spin configuration.

The most noticeable consequence of the one-electron reduction of **1** is that this electron is deposited in ligand-centered orbitals to create a ligand radical. Given the two ligands are identical, the

unpaired electron is shared equally over both ligands and primarily localized to their bridging pyridine moiety (Fig. S23). The electronic structure is best represented by the $[Co^{II}_2(\mu\text{-}L^{3-\bullet})(\mu\text{-}L^{2-})]^-$ ↔ $[Co^{II}_2(\mu\text{-}L^{2-})(\mu\text{-}L^{3-\bullet})]^-$ limiting resonance forms. The storage of reducing equivalents on ligands has been scrutinized previously in related HER catalysts.[23,24,25] In this case, the ligand-centered reduction boosts the field strength aiding stabilization of the low-spin configuration at the metal ions. Importantly for all the calculations carried out on the three different optimized structures of [**1**]$^-$ all gave a ligand radical rather than a $Co^{II}Co^{I}$ mixed-valence species. This outcome is independent of the amount HF exchange included in the functional, which was assessed by varying the HF content from 0% to 50% in 10% increments using the B3LYP hybrid functional (a similar investigation was undertaken for [**1**–H] with the data compiled in Table S33). Despite the equivalence of the Co(II) ions in the M_S = 2 optimized structure, the BS(4,1) solution yielded inequivalent spin states with Co1 high-spin and Co1′ low-spin, a phenomenon not observed neither with the M_S = 4 and M_S = 6 optimized structures nor neutral **1** where the Co ions are both +II and high-spin (Table S30). The specified spin state is achieved by flipping one of the three unpaired electrons on Co1′ in order to give a net $S = {}^1/_2$ at this center. This is reflected in the Mulliken spin population analysis comparing the single-point calculation of each of the optimized structures, where +0.88 spins are located on each Co ion in the M_S = 2 structure that have the same appearance as the SOMO which is primarily comprised by the d_{z^2} orbital (Fig. S23). In contrast, the spin density map of the Co1′ ion in the M_S = 4 optimized structure bears +0.91 spins that results from cancellation of α- and β-spins in the three singly occupied d orbitals of the high-spin Co(II) ion. In the spin density plot of the M_S = 2 structure, there is –0.76 spins located over both ligands, which is less than the expected value on account of bond covalency specifically between the Co center and the "apical" quinoline group. The close proximity of metal- and ligand-based spins produces a greater overlap of these magnetic orbitals and significantly stronger antiferromagnetic interaction (J = –808 cm^{-1}) than that seen in the intramolecular coupling in **1** (J = –5.5 cm^{-1}). The competing bond covalency and bond polarization reaches its zenith in the spin density distribution in the M_S =

6 optimized structure, where +0.55 spins are pushed from each Co center to the first coordination sphere offset by –0.65 spins localized to the pyridine groups through bond polarization (*i.e.* ligand radical) to give a net –0.10 spins on each ligand (Fig. S23)

Table S29 Comparison of geometry optimized[a] bond distances (Å) in [**1**]$^{-}$



	M_S = 2	M_S = 4	M_S = 6
Co(1)–N(1)	1.955	2.371	2.306
Co(1)–N(2)	1.993	1.980	2.057
Co(1)–N(3)	2.113	2.021	2.117
Co(1)–N(4)	1.905	1.975	2.053
Co(1)–N(5)	1.926	2.017	2.116
Co(1)–N(1′)		2.373	2.309
Co(1′)–N(1)		2.374	2.307
Co(1′)–N(1′)	1.953	2.365	2.300
Co(1′)–N(2′)	1.904	1.978	2.053
Co(1′)–N(3′)	1.927	2.021	2.121
Co(1′)–N(4′)	1.995	1.980	2.056
Co(1′)–N(5′)	2.117	2.020	2.120
Co(1)···Co(1′)	2.944	2.669	2.517
Total energy[b]	0.0	+18.3	+19.9

[a] Optimized at the BP86/def2-SVP level; $M_S = 2S + 1$. [b] Total energy relative to most stable structure, in kcal mol^{-1}.

Table S30 Comparison of energies for calculated ground state electronic configurations[a] for [**1**]$^{-}$

		SP	BS(2,1)		BS(4,1)		BS(6,1)	
			HS	BS	HS	BS	HS	BS
M_S = 2	E_{tot} / eV	–151629.88745	–151629.65414	–151629.88746	–151629.00567	–151629.26007	–151628.30582	–151628.63742
	J / cm^{-1} [b]			–808.4		–470.4		–418.4
	E_{rel} / kcal mol^{-1} [c]	0.0		0.0		+14.5		+28.8
	Configuration [d]	↑ ↓ ↑	↑ ↑ ↑	↑ ↓ ↑	↑↑↑ ↑ ↑	↑↑↑ ↓ ↑	↑↑↑ ↑ ↑↑↑	↑↑↑ ↓ ↑↑↑
M_S = 4	E_{tot} / eV	–151629.22963	–151629.24794	–151629.29606	–151629.36303	–151629.24786	–151629.18638	–151629.36313
	J / cm^{-1} [b]			–191.9		+189.2		–224.7
	E_{rel} / kcal mol^{-1} [c]	+15.2		+13.6		+14.7		+12.1
	Configuration [d]	↑↑↑ ↓ ↑↑↓	↑↑↑ ↓ ↑↑↓	↑↑↑ ↓ ↑↓↓	↑↑↑ ↑ ↑↑↓	↑↑↑ ↓ ↑↑↓	↑↑↑ ↑ ↑↑↑	↑↑↑ ↓ ↑↑↑
M_S = 6	E_{tot} / eV	–151629.62676	–151629.21717	–151628.76290	–151629.62676	–151629.42950	–151629.29919	–151629.62673
	J / cm^{-1} [b]			+1601.6		+265.4		–403.6
	E_{rel} / kcal mol^{-1} [c]	+6.0		+25.9		+10.6		+6.0
	Configuration [d]	↑↑↑ ↓ ↑↑↑	↑↑↑ ↓ ↑↑↓	↑↑↑ ↓ ↑↓↓	↑↑↑ ↓ ↑↑↑	↑↑↑ ↓ ↑↑↓	↑↑↑ ↑ ↑↑↑	↑↑↑ ↓ ↑↑↑

[a] Based on broken symmetry calculations at the B3LYP/ZORA-def2-TZVPP level; SP = single point; HS = high-spin; BS = broken-symmetry. [b] Exchange coupling constant calculated using eq. S1. [c] Total energy relative to the BS(2,1) solution for M_S = 2 optimized structure. [d] Spin configuration at each redox center: Co1, red; L_2, green; Co2 blue.

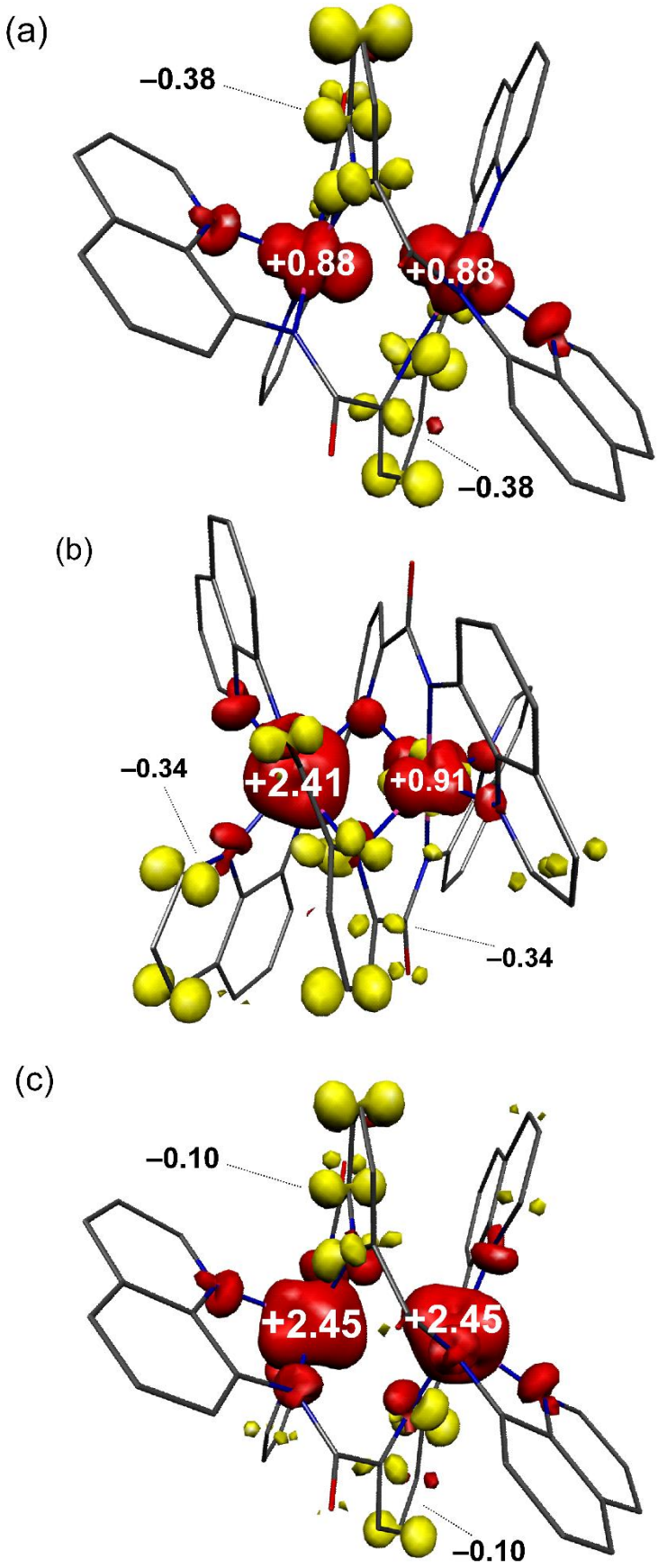


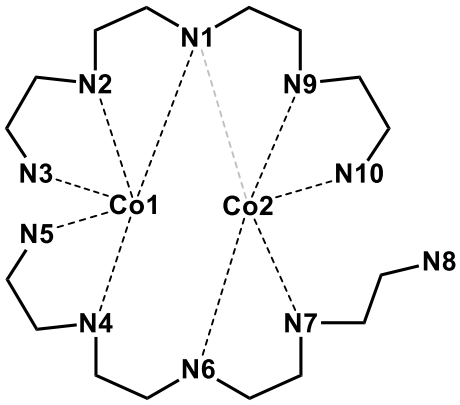
Fig. S23 Mulliken spin population analyses for the (a) M_S = 2, (b) M_S = 4, and (c) M_S = 6 spin configurations for [**1**]$^-$ from B3LYP/ZORA-def2-TZVPP calculations (red: α-spin; yellow: β-spin).

Protonation of the ligand enables creates a vacant coordination site at one of the Co ions. This only occurs for the reduced complex, [**1**]$^{-}$, as the weaker acetic acid cannot protonate neutral **1**. A stronger acid, such as *p*-toluenesulfonic acid is able to cleave [$Cu^{II}_2(\mu\text{-}L)_2$] giving a monocopper complex with a protonated pendent quinoline group, [$Cu^{II}(HL)$]$^{+}$.[26] The long Co–N_{qn} bond distance in [**1**]$^{-}$ represents the most likely protonation site in this complex; this protonated species is denoted [**1**–H]. The loss of a nitrogen donor (N8) renders the two Co ions inequivalent, with the second cobalt center identified as losing one quinoline group and therefore maximally five-coordinate if both pyridine groups are retained as bridging ligands. A range of electronic and structural possibilities were assessed at the BP86/def2-SVP level of theory (Table S31). Three multiplicities of M_S = 2, 4 and 6 were optimized based on coordinates generated using molecular mechanics software. This isomer position the protonated quinoline – denoted quinolinium – as projected away from the metal ion based on electrostatic repulsion. The resultant structure produced an intraligand hydrogen-bond between the amide carbonyl and the quinolinium. The addition of this interaction provides this isomer an additional 6.6 kcal mol^{-1} stabilization over the one where the quinolinium proton is adjacent the Co ion (designated "isomer 2" in Table S31). Each optimized structure both Co centers are five-coordinate with only one pyridine (N1) retained in the bridging mode, specifically the group on the protonated ligand, HL^{-}. The Co1 center adopts a geometry best described as trigonal bipyramidal whereas the Co2 center the geometry is closer to square pyramidal where the ventral side presents an opening to further coordination.

As noted above, the increase in spin multiplicity results in a decrease of the intermetal separation though the changes are much less pronounced for [**1**–H] (Table S31). The comparison of total energies for the possible electronic permutations reveals the no preference for the spin state of Co1 (Table S32). This is can be seen in the <3 kcal mol^{-1} difference between the M_S = 2, M_S = 4 and M_S = 6 solution for each of the respective optimized structures. On the other hand, there is no calculation that produces a high-spin configuration for Co2; this is invariably low-spin as could be expected given its coordination number of 5 and approximate square pyramidal geometry when

compared to the result for [**1**]$^{-}$. The ligand radical identified in [**1**]$^{-}$ is retained in [**1**–H], though the distribution of the unpaired spin has shifted entirely to the quinolinium group (Fig. S24). This is the same outcome in the analogous reduced complexes with Ni(II) and Cu(II) central ions along the pathway to hydrogen generation,[26,27] as well as related Co systems from *N*-donor ligands.[23] The broken-symmetry solution with unpaired spins on each Co ion and the ligands is independent of the amount HF exchange included in the functional, which was assessed by varying the HF content from 0% to 50% in 10% increments using the B3LYP hybrid functional (Table S33). This is because HF exchange varies the degree of localization within the complex, *i.e.* covalency, but not the degree of bond polarization that gives rise to π-ligand radicals. Increasing the HF content in the functional leads to more α-spin deposited on the Co ions.

Table S31 Comparison of geometry optimized[a] bond distances (Å) in [**1**–H]



	Isomer 1			**Isomer 2**	
	M_S **= 2**	M_S **= 4**	M_S **= 6**	M_S **= 2**	M_S **= 6**
Co(1)–N(1)	2.028	2.227	2.308	1.921	2.314
Co(1)–N(2)	2.006	1.956	1.987	1.958	2.107
Co(1)–N(3)	2.109	2.003	2.062	1.977	2.001
Co(1)–N(4)	1.916	1.917	2.023	1.887	2.007
Co(1)–N(5)	1.958	1.947	2.039	1.931	2.100
Co(2)–N(1)	2.446	2.240	2.276		2.408
Co(2)–N(2)					2.265
Co(2)–N(6)	1.933	1.988	2.004	1.882	1.953
Co(2)–N(7)	1.951	1.949	1.946	2.017	2.134
Co(2)–N(9)	1.877	1.890	1.893	1.886	2.029
Co(2)–N(10)	1.918	1.944	1.953	1.954	2.184
Co(1)···Co(2)	2.805	2.797	2.724	2.547	2.437
Co(2)···H				2.170	2.440
Total energy[b]	0.0	+7.8	+21.1	+6.6	+17.8

[a] Optimized at the BP86/def2-SVP level; $M_S = 2S + 1$. [b] Total energy relative to most stable structure, in kcal mol^{-1}.

Table S32 Comparison of energies for calculated ground state electronic configurations[a] for [**1**–H]

		M_S = 2	BS(2,1)		BS(4,1)		M_S = 4	M_S = 6
			HS	BS	HS	BS		
M_S = 2	E_{tot} / eV	–151642.16581	–151641.98287	–151642.16579	–151641.53790	–151641.62032	–151641.98298	
	J / cm^{-1} [b]			–649.7		–107.1		
	E_{rel} / kcal mol^{-1} [c]	0.0		0.0		+12.6	+4.2	
	Configuration [d]	↑ ↓ ↑	↑ ↑ ↑	↑ ↓ ↑	↑↑↑ ↑ ↑	↑↑↑ ↓ ↑	↑ ↑ ↑	
M_S = 4	E_{tot} / eV		–151642.10864	–151642.16285	–151641.81834	–151641.86920	–151642.10863	–151641.81832
	J / cm^{-1} [b]			–206.4		–67.7		
	E_{rel} / kcal mol^{-1} [c]			+0.1		+6.9	+1.3	+8.0
	Configuration [d]		↑ ↑ ↑	↑ ↓ ↑	↑↑↑ ↑ ↑	↑↑↑ ↓ ↑	↑ ↑ ↑	↑↑↑ ↑ ↑
M_S = 6	E_{tot} / eV		–151641.81132	–151641.82252	–151642.02144	–151642.02764	–151641.81132	–151642.02149
	J / cm^{-1} [b]			–44.9		–8.3		
	E_{rel} / kcal mol^{-1} [c]			+7.9		+3.2	+8.2	+3.3
	Configuration [d]		↑ ↑ ↑	↑ ↓ ↑	↑↑↑ ↑ ↑	↑↑↑ ↓ ↑	↑ ↑ ↑	↑↑↑ ↑ ↑

[a] Based on broken symmetry calculations at the B3LYP/ZORA-def2-TZVPP level; HS = high-spin; BS = broken-symmetry. [b] Exchange coupling constant calculated using eq. S1. [c] Total energy relative to the BS(2,1) solution for M_S = 2 optimized structure. [d] Spin configuration at each redox center: Co1, red; HL, green; Co2 blue.

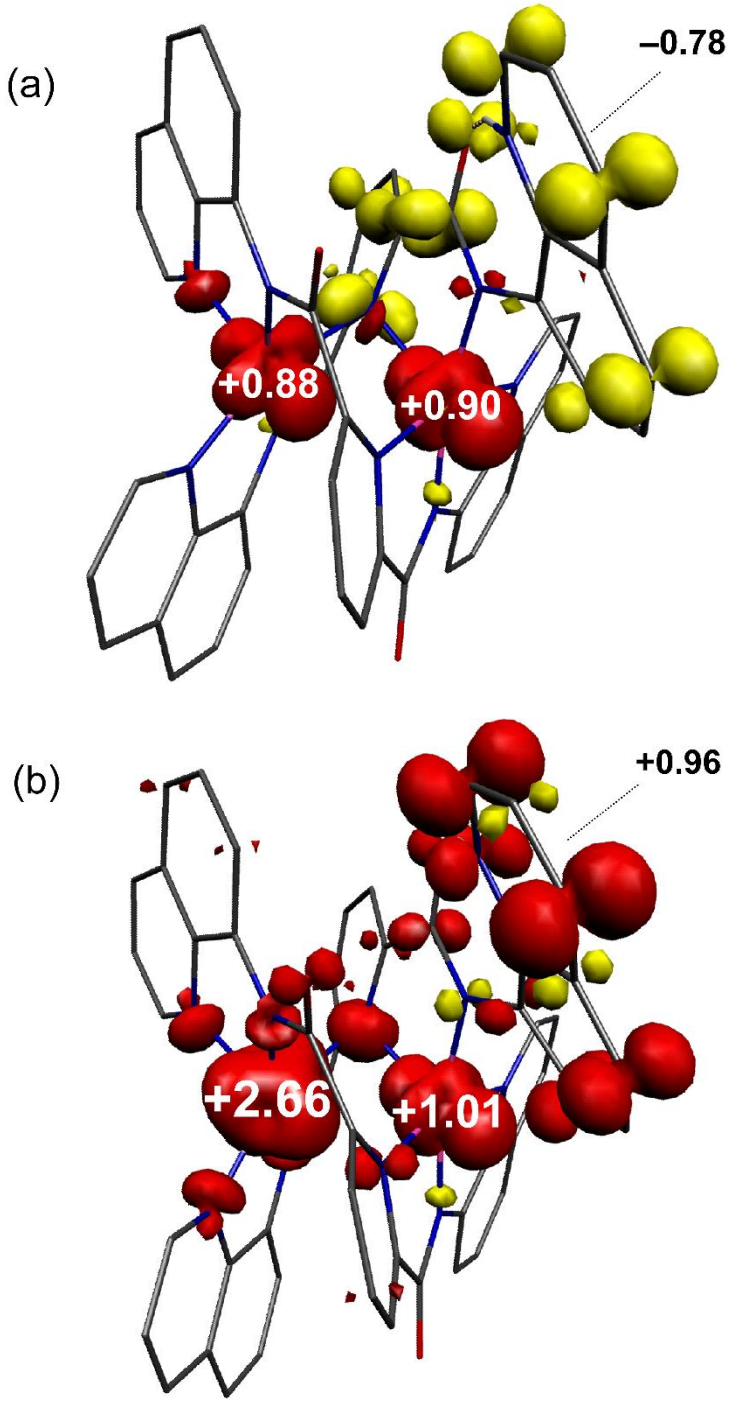


Fig. S24 Mulliken spin population analyses for the (a) M_S = 2, and (b) M_S = 6 spin configurations for [**1**–H] from B3LYP/ZORA-def2-TZVPP calculations (red: α-spin; yellow: β-spin).

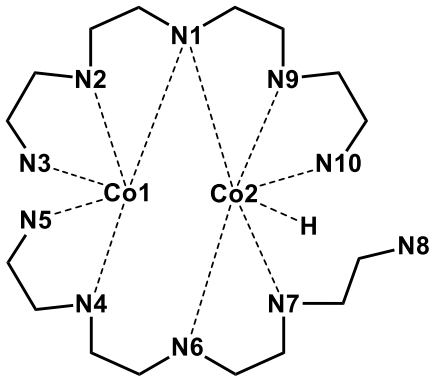
Table S33 Effect of Hartree-Fock exchange on electronic structure of [**1**–H] [a]

HF content	ρ_{Co1} [b]	ρ_{Co2} [b]	ρ_{L} [c]	J / cm^{-1} [d]
10%	+0.792	+0.673	–0.465	–1070.66
20% [e]	+0.879	+0.898	–0.777	–646.69
30%	+0.918	+0.966	–0.884	–510.16
40%	+0.934	+0.987	–0.922	–427.92
50%	+0.943	+0.995	–0.938	–334.90

[a] Based on a BS(2,1) calculation at the B3LYP/ZORA-def2-TZVPP level where HF% was specified for each calculation. [b] Calculated Mulliken spin density at the Co ion. [c] Calculated spin density on the ligands, which is the net difference in α-spin delocalized to the first coordination sphere nitrogen donor atoms due to covalency and the β-spin distributed in the π orbital system from bond polarization. [d] Exchange coupling constant calculated using eq. S1, and the difference in energy between the high-spin $S = {}^{3}/_{2}$ (ferromagnetic) and the BS $S = {}^{1}/_{2}$ (antiferromagnetic) solutions. [e] HF exchange content of the B3LYP functional.

The release of a quinoline group from the coordination sphere triggered by its protonation creates an opening for a subsequent protonation of the Co center. The resultant monocationic complex, denoted [H-**1**–H]$^{+}$, has a protonated quinoline on the same side as the protonated Co ion. Upon protonation, a Co^{III}–H is formed where one electron is supplied by the metal ion and the other from the ligand. The structure was optimized for M_S = 2 and M_S = 4 states which accounts for an adjacent low-spin and high-spin Co(II) ion, respectively. For both structures, each Co ion is six-coordinate with both pyridine groups bridging the metal centers (Table S33). The different spin state at Co1 leads to a lengthening of the Co–N distances in the M_S = 4 optimized structure, whereas there is a negligible change for the bond distances about Co2. The Co–H distance is identical in both optimized structures suggesting a low-spin Co(III) center in both and that the higher multiplicity only impacts the Co(II) ion. This distance is in the range observed for structurally characterized Co^{III}–H complexes.[28] Interestingly the Co···Co distance increases by ~0.15 Å for the high-spin structure, which is the opposite trend seen for [**1**]$^{-}$ and [**1**–H], and only observed for the optimized structures of **1**. Single-point calculations of each optimized structure reveals the high-spin M_S = 4 structure is energetically favored by 2.3 kcal mol^{-1} over the low-spin M_S = 2 structure though this difference in within the error for DFT (Table S35). The calculation shows no change to the spin state at the Co(III) center underscoring the ligand field strength of the hydride enforcing the low-spin d^6 configuration. The electronic structure is best visualized by the Mulliken spin density plot for both configurations with the unpaired spins found on Co2 and the quinolinium group in [**1**–H] now reflecting their diamagnetic state in [H-**1**–H]$^{+}$ (Fig. S25). The two solutions differ only in the α-spin located at the adjacent Co(II) center as +0.82 and +2.64 spins for the M_S = 2 and M_S = 4 species, respectively. Protonation of the Co ion requires a two-electron transfer to form a Co–H bond. With the second electron derived from the ligand, the ligand is oxidized to the HL^{-} form which in turn reduces its field strength and thereby leading to stabilization of the high-spin Co(II) ion as observed in **1**.

Table S34 Comparison of geometry optimized bond distances (Å) in [H-**1**–H]^{z}



	z = 1+		z = 0	
	M_S = 2	M_S = 4	M_S = 3	M_S = 5
Co(1)–N(1)	2.082	2.242	2.077	2.228
Co(1)–N(2)	1.921	2.022	1.912	2.024
Co(1)–N(3)	1.974	2.049	1.962	2.073
Co(1)–N(4)	1.899	1.968	1.904	1.997
Co(1)–N(5)	1.962	2.090	1.964	2.130
Co(1)–N(6)	2.285	2.449	2.166	2.349
Co(2)–N(1)	2.150	2.135	2.082	2.146
Co(2)–N(6)	1.981	2.010	1.992	2.033
Co(2)–N(7)	2.011	2.006	2.012	1.999
Co(2)–N(9)	1.882	1.880	1.889	1.889
Co(2)–N(10)	1.928	1.931	1.941	1.937
Co(2)–H	1.437	1.438	1.459	1.447
Co(1)···Co(2)	2.709	2.852	2.680	2.731
Total energy[b]	0.0	+14.9	0.0	+21.2

[a] Optimized at the BP86/def2-SVP level; $M_S = 2S + 1$. [b] Total energy relative to most stable structure, in kcal mol^{-1}.

The pathway to generating H_2 is either chemical with protonation of the hydride ligand followed by reduction of the complex to give neutral **1**, where the proton is derived from the quinolinium group as a stronger acid in the DMF/water solvent mixture than acetic acid.[29] Alternatively, the [H-**1**–H]$^+$ complex can be reduced at the electrode to produce the formally Co^{II}–H species whose pK_a or "hydricity" is more amenable to protonation from the acid.[25,30–32] Structures for the [H-**1**–H] species were optimized with M_S = 3 and M_S = 5 states, where the former would comprise ferromagnetically coupled low-spin Co(II) ions and the latter a coupling of a high-spin Co(II) with the low-spin Co(II) ion that bears the hydride ligand (Table S34). The BP86/def2-SVP calculation prefers the low-spin configuration by 21.2 kcal mol^{-1}, though the inclusion of HF exchange reverses this ordering to stabilize the high-spin configuration by 4.8 kcal mol^{-1} (Table S35). This energy gap is greater than that recorded for the [H-**1**–H]$^+$ complex but still within the error for DFT.[22] It was anticipated that a Co(II)–H unit would be formed with reduction of the Co(III) center, however, the low-spin d^6 configuration is energetically preferred to a ligand-centered reduction, and therein the excess electron is found on both ligands primarily on the bridging pyridine groups as in [**1**]$^-$ (Fig. S25). The strong field hydride destabilizes the e_g orbitals of the Co(III) center, specifically the Co–H σ* MO which is composed of predominantly the d_{z^2} orbital, above that of the π* MOs of the ligand and therefore the electron is not added to the Co(III) ion. As a consequence, the Co–H bond length is largely unchanged compared with [H-**1**–H]$^+$ (Table S34). The net S = 0 and S = 1 spin ground state calculated for the M_S = 3 and M_S = 5 optimized structures results from strong antiferromagnetic coupling between an unpaired spin on the Co(II) ion with the ligand radical, as reflected in the large exchange coupling constants of –1222 and –370 cm^{-1}, respectively (Table S35). This magnetic interaction also serve to stabilize the ligand radical as they are significantly larger than the exchange coupling between the more distance Co ions. There is insufficient experimental data to speculate as to whether the [H-**1**–H]$^+$ complex is reduced prior to H_2 elimination and regeneration of neutral **1**. However, the change in spin state at the adjacent Co(II)

center may serve to facilitate the H_2 production by modulating the pK_a of the terminal hydride ligand. It has been shown that high-spin states in [Co(porphyrin)] species increase the pK_a of the Co-hydride.[32] The change in pK_a is not as significant as reducing the Co(III) d^6 ion to a Co(II) d^7 entity,[31] as the effect will be more subtle given the ~2.7 Å separation of the Co ions in this system. However, this system exploits the synergism between these two Co ions in a similar fashion to the bimetallic [Fe_2] and [NiFe] active sites of native hydrogenase enzymes.[33]

Table S35 Comparison of energies for calculated ground state electronic configurations[a] for [H-**1**–H]^z

[H-**1**–H]$^+$	M_S = 2	M_S = 4	E_{rel} / kcal mol^{-1} [b]
E_{tot} / eV	–151654.24060	–151654.33999	–2.3
Configuration [c]	↑ • •	↑↑↑ • •	

[H-**1**–H]	BS(1,1)	BS(3,1)	E_{rel} / kcal mol^{-1} [b]
E_{tot} / eV	–151657.18488 (J = –1222.4 cm^{-1})[d]	–151657.39105 (J = –369.8 cm^{-1})[d]	–4.8
Configuration [c]	↑ ↓ •	↑↑↑ ↓ •	

[a] Based on B3LYP/ZORA-def2-TZVPP level; BS = broken-symmetry. [b] $E_{rel} = E_{LS} - E_{HS}$; LS = low-spin; HS = high-spin. [c] Spin configuration at each redox center: Co1, red; HL, green; Co2 blue; dot = no spin. [d] Exchange coupling constant calculated using eq. S1.

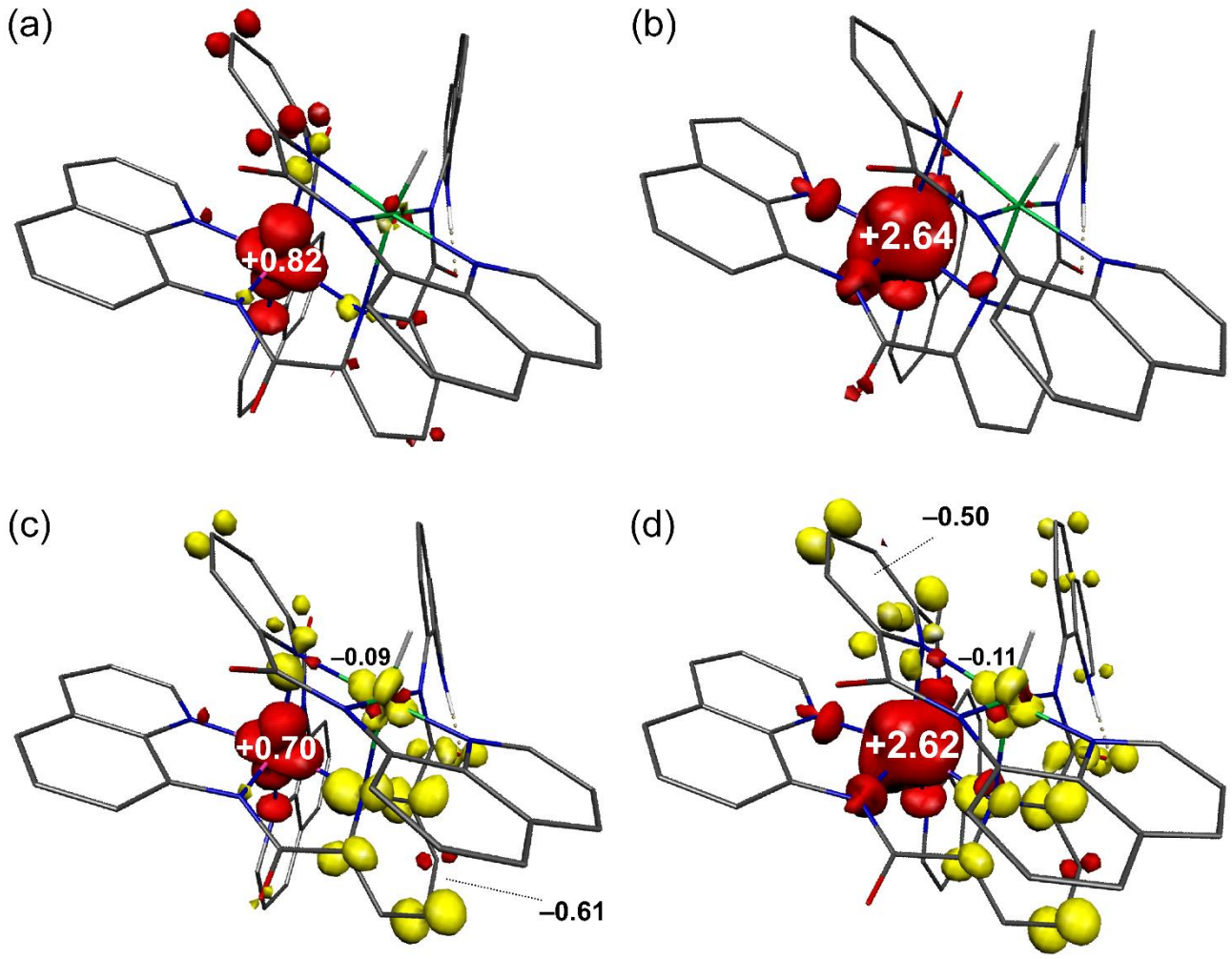


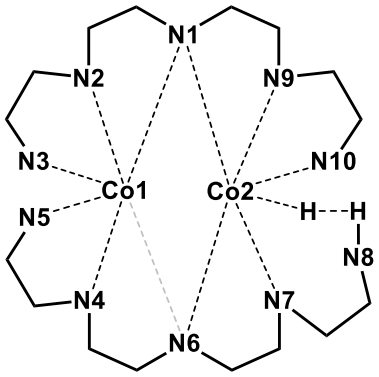
Fig. S25 Mulliken spin population analyses for the (a) M_S = 2 and (b) M_S = 4 states of [H-**1**–H]$^+$, and (c) BS(1,1) and (d) BS(3,1) solutions for [H-**1**–H] from B3LYP/ZORA-def2-TZVPP calculations (red: α-spin; yellow: β-spin. Atom color: Co^{II}, pink; Co^{III}, green).

The completion of the mechanism sees the quinolinium group relay its proton to the terminal hydride ligand. The position of the quinolinium relative to the Co-hydride presents a barrier to this last stage of the mechanism as the quinolinium needs to rotate about its amide linkage to position the proton in order to facilitate the heterolytic formation of a H–H bond. The transition state that represents the proximity of the proton and hydride atoms is optimized for the monocationic [H-**1**–H]$^{+}$ and neutral species [H-**1**–H] whose electronic structure has been assessed as containing a Co^{III}–H (Table S35). This proposed transition state is effectively a measure of the energy barrier to rotation of the quinolinium group that requires breaking of the intraligand hydrogen-bond. For [**1**–H] this was calculated at 6.6 kcal mol^{-1} (Table S36). Here, the energy barrier is roughly the same, and found in the range of 9–12 kcal mol^{-1} for the low-spin optimized structures, and a significantly smaller <4 kcal mol^{-1} for the high-spin optimized structures (Table S36). It should be noted that this is the net result from energy expended to break the intraligand hydrogen bond which is offset energy gained from establishing the H···H interaction that precedes H_2 formation. The structural changes to applied to [H-**1**–H]$^{+}$ (M_S = 2 optimized) in forming the proposed transition state [**1**–H···H]$^{+}$ sees a 0.01 Å lengthening of the Co–H bond and a concomitant 0.03 Å shortening of the Co–N_{py} bond that is *trans* to the hydride ligand. The nucleophilic hydride resides 1.553 Å away from the electrophilic proton in this proposed transition state and in the range for such interactions for the so-named dihydrogen bond.[34] The Co–N distances about the neighboring Co(II) ion are invariant, and the Co···Co separation increases slightly by 0.015 Å. Conversely, the neutral variant, [**1**–H···H] (M_S = 3 optimized) sees a 0.01 Å shortening of the Co–H bond and 0.03 Å lengthening of the Co–N_{py} bond trans to it. Again there is negligible change at the adjacent Co(II) center but the Co ions are 0.1 Å further apart. The structures with an adjacent high-spin Co(II) ion, which were slightly favored as the Co hydride complexes, show similar bond dimensions about the Co(III) center. For the neutral intermediate, there is a 0.01 Å elongation of the Co–H bond accompanied by a 0.04 Å shortening of the *trans* Co–N_{py} bond. The M_S = 4 optimized [**1**–H···H]$^{+}$

exhibits the greatest structural rearrangement with the pyridine groups of both ligands no longer bridging the metal ions. This results in a four-coordinate Co(II) ion that is tetrahedral geometry as a high-spin center, reminiscent to mixed-valent monocationic **3** produced from the chemical oxidation of **1** (*vide infra*). It can be inferred that such a structure is not truly representative of the transition state as such a rearrangement at the Co(II) center would require a prohibitive amount of reorganization energy. The Co(III) ion is six-coordinate with comparable geometry to the low-spin $M_S = 2$ optimized structure (Table S36).

The production of H_2 and reconstitution of the neutral **1** catalyst occurs in three steps: proton transfer from the quinolinium to the hydride, re-coordination of the quinoline to the Co center, and reduction of the Co(III) to Co(II). For [**1**–H···H]$^+$, this begins with protonation of the hydride to release H_2, and is followed by either coordination of the quinoline than reduction of the Co(III) ion to form **1**, or the reduction of the Co(III) species precedes the ligation by quinoline. Neither the experimental nor computational data allow the exact sequence to be ascertained. Alternatively, the [H-**1**–H]$^+$ can be first reduced to the neutral Co-hydride species, [H-**1**–H], which is followed by proton transfer to the hydride, quinoline coordination and an internal electron transfer from the reduced ligand to the Co center to give the +II oxidation state found for **1**. In both scenarios it can be suggested that the spin state change occurs after coordination of the quinoline to give the six-coordinate Co center as the isotropic distribution of *N*-donor atoms (in this case octahedral) promotes a high-spin configuration. This is evidenced by the contrasting geometries for each Co(II) ion in [**1**–H] where the low- and high-spin configurations for the trigonal bipyramidal Co(II) center are isoenergetic whereas the square pyramidal Co(II) center is strictly low-spin (Table S32).

Table S36 Comparison of geometry optimized bond distances (Å) in the [**1**–H···H]^{z} transition state



	z = 1+		z = 0	
	M_S = 2	M_S = 4	M_S = 3	M_S = 5
Co(1)–N(1)	2.036		2.086	2.216
Co(1)–N(2)	1.915	1.969	1.909	2.001
Co(1)–N(3)	1.970	2.004	1.958	2.072
Co(1)–N(4)	1.896	1.957	1.907	1.996
Co(1)–N(5)	1.948	2.008	1.965	2.094
Co(1)–N(6)	2.263		2.156	2.360
Co(2)–N(1)	2.181	2.035	2.125	2.184
Co(2)–N(6)	1.990	1.968	2.006	2.021
Co(2)–N(7)	1.978	1.973	1.966	1.969
Co(2)–N(9)	1.891	1.896	1.895	1.891
Co(2)–N(10)	1.949	1.950	1.950	1.946
Co(2)–H	1.446	1.457	1.448	1.458
H···H	1.553	1.431	1.549	1.476
Co(1)···Co(2)	2.726	3.612	2.780	2.802
Relative energy [b]	+8.9	+3.6	+11.7	+3.9

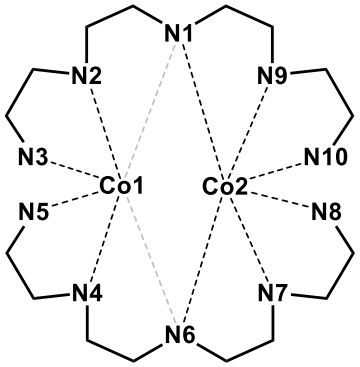
[a] Optimized at the BP86/def2-SVP level; $M_S = 2S + 1$. [b] Total energy relative to the corresponding structure of [H-**1**–H]^{z} (z = 1+, 0) in Table S34, in kcal mol^{-1}.

Oxidation of **1** and **2** yields mixed-valent monocationic species **3** and **4**. As there is no difference in the electronic structure between these pairs of compounds that differ only in the addition of a *tert*-butyl group on the periphery of the ligand, calculations were restricted to the slightly smaller compound **3**. The structure was optimized starting from the crystallographic coordinates with a multiplicity $M_S = 4$ consistent with three unpaired electrons for the high-spin Co(II) ion determined by room temperature magnetic measurements. The result varied little from the experimental data, with the salient change a shortening is the intermetallic distance by 0.3 Å in the optimized structure (Table S37). A similar shift in the Co···Co separation was observed for the optimized structure of **1** (*vide supra*), which is ascribed to the absence of lattice forces present in the crystalline material and the degree of flexibility of the ligands to accommodate intermolecular interactions. The average Co–N distance for Co2 is consistent with a low-spin Co(III) center, and the adjacent tetrahedral Co(II) ion is clearly high-spin giving rise to the aforementioned magnetic moment. The degree of tetrahedralization is lowered slightly in the optimized structure as shortening the Co···Co leads to a departure of the *N*,*N*′-chelates about the Co(II) ion from their mutually orthogonal alignment in the crystal structure. However, the $M_S = 4$ optimized structure is favored by 16.1 kcal mol^{-1} over the crystallographic structure, underscoring the energetic input of packing forces in the latter.

As noted previously, the BP86 pure functional is bias toward low-spin states, and this is the case here for the $M_S = 2$ optimized structure for the monocation in **3**. In order to accommodate a low-spin configuration at the Co(II) center, the structure reverts back to the edge-sharing bioctahedron topology with the pyridine groups returning to a bridging mode (Table S37). Re-establishing a six-coordinate Co(II) center permits the switch to the low-spin configuration for this d^7 ion. The Co–N bond distances is the Co(II) ion reveal an effective Jahn-Teller distortion along the Co···Co vector and long $Co–N_{py}$ bonds that eliminate any orbital degeneracy. This is in contrast to the neighboring low-spin $S = 0$ Co(III) ion where the first coordination sphere bonds are the roughly equivalent and mostly unchanged

from the M_S = 4 optimized structure. Although the M_S = 2 optimized structure in no way matches the experimental reality, it is 7.4 kcal mol^{-1} more stable than the corresponding M_S = 4 optimized structure.

Table S37 Comparison of experimental and geometry optimized[a] structural metrics in **3**



	Experimental	**M_S = 4 optimized**	**M_S = 2 optimized**
Co(1)–N(1)			2.347
Co(1)–N(2)	1.977(2)	1.967	1.927
Co(1)–N(3)	2.009(3)	2.002	1.947
Co(1)–N(4)	1.975(2)	1.968	1.919
Co(1)–N(5)	2.001(3)	2.001	1.948
Co(1)–N(6)			2.349
Co(2)–N(1)	1.975(2)	1.901	2.043
Co(2)–N(6)	1.981(2)	1.941	2.045
Co(2)–N(7)	1.890(2)	1.902	1.902
Co(2)–N(8)	1.941(2)	1.941	1.931
Co(2)–N(9)	1.882(2)	1.978	1.903
Co(2)–N(10)	1.940(2)	1.979	1.933
Co(1)···Co(2)	4.019(1)	3.702	2.988
α [b]	87.2(1)	73.9	
Total energy [c]	+23.5	+7.4	0.0

[a] Optimized at the BP86/def2-SVP level; distances in angstrom; angles in degrees. [b] Dihedral between two chelating *N*,*N*′-donor ligands coordinated to Co(1). [c] Total energy relative to the most stable structure, in kcal mol^{-1}.

The true electronic structure is decided by comparing total energies calculated with our B3LYP/ZORA-def2-TZVPP protocol applied consistently across this series of compounds (Table S38). The results show the $S = {}^{3}/_{2}$ calculation on the $M_S = 4$ optimized structure as the most stable solution. This is 11.4 kcal mol^{-1} more stable than the corresponding spin-doublet ground state calculated on the $M_S = 2$ optimized structure, and 15.1 kcal mol^{-1} preferable to a spin-doublet ground state for the mixed coordination number of the $M_S = 4$ optimized structure. It is important to note that none of the calculations positioned any unpaired spins at the Co(III) center for any of the optimized or crystallographic geometries. This confirms a low-spin $S = 0$ state is the preferred configuration for a six-coordinate Co(III) center. The electronic structure is best visualized using Mulliken spin population analysis which shows +2.67 spins located at the four-coordinate Co(II) center with +0.33 delocalized on to its first coordination sphere as seen in the high-spin Co(II) ions in **1** (Fig. S26). There is no trace of spin density on the neighboring Co(III) center underpinning the preference for a low-spin configuration for an octahedral d^6 metal ion.

Table S38 Comparison of energies for calculated ground state electronic configurations[a] for **3**

		M_S = 4 optimized	M_S = 2 optimized
M_S = 2	E_{tot} / eV	–151622.91222	–151623.07514
	ρ_{Co1} [b]	+0.954	+0.872
	E_{rel} / kcal mol^{-1} [c]	+15.1	+11.4
	Configuration [d]	↑ • •	↑ • •
M_S = 4	E_{tot} / eV	–151623.56853	
	ρ_{Co1} [b]	+2.674	
	E_{rel} / kcal mol^{-1} [c]	0.0	
	Configuration [d]	↑↑↑ • •	

[a] Based on B3LYP/ZORA-def2-TZVPP level; $M_S = 2S + 1$. [b] Calculated Mulliken spin density at the Co1 center. [c] Energy relative to the most stable configuration: the M_S = 4 calculation on the M_S = 4 optimized structure. [d] Spin configuration at each redox center: Co1, red; L, green; Co2 blue; dot = no spin. [d] Exchange coupling constant calculated using eq. S1.

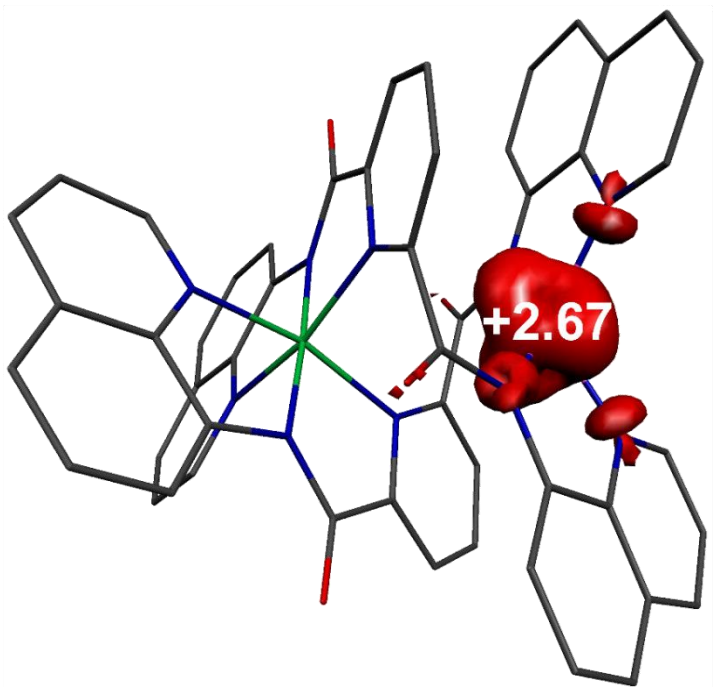


Fig. S26 Mulliken spin population analysis for **3** obtained from B3LYP/ZORA-def2-TZVPP calculations (red: α-spin. Atom color: Co^{II}, pink; Co^{III}, green).

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