Supporting Information

Table S1. Selected geometric parameters, distances and angles (Å, º), for **1**

Structural data taken from Battaglia *et al*., (entry JOGSON in the CSD) with due atom renaming according to Figure **1** in the main text.

Cu1—O1B	1.9461 (6)	N1B—C5B	1.3667 (4)
Cu1—O1W	2.2724 (7)	N2A—C11A	1.3750 (4)
Cu1—N1A	2.0192 (6)	N2A—C10A	1.3390 (4)
Cu1—N2A	2.0116 (6)	O1D—C1D	1.3357 (4)
Cu1—O2B^{i}	1.9365 (6)	O2D—C1D	1.2238 (4)
O1B—C1B	1.2565 (4)	N1D—C5D	1.3766 (4)
O2B—C1B	1.2813 (4)	O1C—N1C	1.2169 (4)
N1A—C1A	1.3105 (4)	O2C—N1C	1.2335 (4)
N1A—C12A	1.3697 (4)	O3C—N1C	1.2425 (4)
O1B—Cu1—O1W	89.24 (2)	O1B—C1B—O2B	124.58 (2)
O1B—Cu1—N1A	92.04 (2)	O1B—C1B—C2B	117.90 (2)
O1B—Cu1—N2A	173.66 (2)	O2B—C1B—C2B	117.44 (2)
O1B—Cu1—O2B^{i}	94.60 (2)	N1B—C5B—C4B	120.63 (2)
O1W—Cu1—N1A	99.31 (2)	N1B—C5B—C6B	121.11 (2)
O1W—Cu1—N2A	93.98 (2)	N2A—C10A—C9A	120.56 (2)
O1W—Cu1—O2B^{i}	94.78 (2)	N2A—C11A—C12A	116.31 (2)
N1A—Cu1—N2A	82.06 (2)	N2A—C11A—C7A	121.79 (2)
O2B^{i}—Cu1—N1A	164.49 (2)	N1A—C12A—C4A	121.26 (2)
O2B^{i}—Cu1—N2A	90.57 (2)	N1A—C12A—C11A	118.17 (2)
Cu1—O1B—C1B	132.28 (2)	O1D—C1D—C2D	116.09 (2)
Cu1^{i}—O2B—C1B	126.44 (2)	O2D—C1D—C2D	124.23 (2)
Cu1—N1A—C1A	130.25 (2)	O1D—C1D—O2D	119.65 (2)
Cu1—N1A—C12A	111.11 (2)	N1D—C5D—C4D	122.51 (2)

C1A—N1A—C12A	118.63 (2)	N1D—C5D—C6D	120.07 (2)
Cu1—N2A—C11A	112.14 (2)	O2C—N1C—O3C	119.63 (2)
Cu1—N2A—C10A	128.68 (2)	O1C—N1C—O2C	120.82 (2)
C10A—N2A—C11A	119.16 (2)	O1C—N1C—O3C	119.55 (2)
N1A—C1A—C2A	122.63 (2)		

Symmetry code: (i) -*x*, *y*, -*z*+1/2.

Table S2: Selected geometric parameters, distances and angles (Å, º), for 2. (Data provided by a queezed refinement of an incomplete model)

Cu1—O1B^{i}	1.9442 (13)	C9A—C10A	1.395 (3)
Cu1—O2B	1.9486 (13)	C9A—H9A	0.93
Cu1—N1A	2.0117 (16)	C10A—H10A	0.93
Cu1—N2A	2.0186 (16)	C11A—C12A	1.426 (3)
Cu1—N1C	2.2563 (15)	O1B—C1B	1.269 (2)
N1A—C1A	1.328 (3)	O2B—C1B	1.263 (2)
N1A—C12A	1.357 (3)	N1B—C5B	1.387 (3)
N2A—C10A	1.331 (3)	N1B—H1BA	0.90
N2A—C11A	1.362 (3)	N1B—H1BB	0.90
C1A—C2A	1.399 (3)	C1B—C2B	1.481 (3)
C1A—H1A	0.93	C2B—C7B	1.396 (3)
C2A—C3A	1.359 (4)	C2B—C3B	1.396 (3)
C2A—H2A	0.93	C3B—C4B	1.375 (3)
C3A—C4A	1.395 (4)	C3B—H3B	0.93
C3A—H3A	0.93	C4B—C5B	1.395 (3)
C4A—C12A	1.403 (3)	C4B—H4B	0.93
C4A—C5A	1.440 (3)	C5B—C6B	1.398 (3)
C5A—C6A	1.345 (4)	C6B—C7B	1.373 (3)
C5A—H5A	0.93	C6B—H6B	0.93
C6A—C7A	1.437 (3)	C7B—H7B	0.93
C6A—H6A	0.93	N1C—C2C	1.329 (3)
C7A—C8A	1.402 (4)	N1C—C1C	1.327 (3)
C7A—C11A	1.398 (3)	C1C—C2C^{ii}	1.384 (3)
C8A—C9A	1.373 (4)	C1C—H1C	0.93
C8A—H8A	0.93	C2C—H2C	0.93
O1B^{i}—Cu1—O2B	96.27 (6)	N2A—C10A—H10A	118.8
O1B^{i}—Cu1—N1A	164.80 (6)	C9A—C10A—H10A	118.8
O2B—Cu1—N1A	90.62 (6)	N2A—C11A—C7A	123.1 (2)
O1B^{i}—Cu1—N2A	89.63 (6)	N2A—C11A—C12A	116.33 (17)
O2B—Cu1—N2A	170.05 (6)	C7A—C11A—C12A	120.59 (19)

N1A—Cu1—N2A	81.82 (7)	N1A—C12A—C4A	123.0 (2)
O1B^{i}—Cu1—N1C	93.15 (6)	N1A—C12A—C11A	116.69 (17)
O2B—Cu1—N1C	87.55 (6)	C4A—C12A—C11A	120.29 (19)
N1A—Cu1—N1C	100.69 (6)	C1B—O1B—Cu1^{i}	123.39 (12)
N2A—Cu1—N1C	100.16 (6)	C1B—O2B—Cu1	135.32 (12)
C1A—N1A—C12A	118.31 (17)	C5B—N1B—H1BA	115.1
C1A—N1A—Cu1	129.04 (14)	C5B—N1B—H1BB	107.5
C12A—N1A—Cu1	112.63 (13)	H1BA—N1B—H1BB	130.7
C10A—N2A—C11A	118.25 (18)	O2B—C1B—O1B	124.58 (17)
C10A—N2A—Cu1	129.34 (15)	O2B—C1B—C2B	117.34 (16)
C11A—N2A—Cu1	112.42 (13)	O1B—C1B—C2B	118.09 (16)
N1A—C1A—C2A	122.0 (2)	C7B—C2B—C3B	118.20 (18)
N1A—C1A—H1A	119.0	C7B—C2B—C1B	121.50 (17)
C2A—C1A—H1A	119.0	C3B—C2B—C1B	120.30 (17)
C3A—C2A—C1A	119.6 (2)	C4B—C3B—C2B	120.72 (19)
C3A—C2A—H2A	120.2	C4B—C3B—H3B	119.6
C1A—C2A—H2A	120.2	C2B—C3B—H3B	119.6
C2A—C3A—C4A	120.2 (2)	C3B—C4B—C5B	120.65 (19)
C2A—C3A—H3A	119.9	C3B—C4B—H4B	119.7
C4A—C3A—H3A	119.9	C5B—C4B—H4B	119.7
C3A—C4A—C12A	116.8 (2)	N1B—C5B—C4B	120.4 (2)
C3A—C4A—C5A	124.9 (2)	N1B—C5B—C6B	120.8 (2)
C12A—C4A—C5A	118.2 (2)	C4B—C5B—C6B	118.70 (19)
C6A—C5A—C4A	121.0 (2)	C7B—C6B—C5B	120.17 (19)
C6A—C5A—H5A	119.5	C7B—C6B—H6B	119.9
C4A—C5A—H5A	119.5	C5B—C6B—H6B	119.9
C5A—C6A—C7A	121.8 (2)	C6B—C7B—C2B	121.26 (19)
C5A—C6A—H6A	119.1	C6B—C7B—H7B	119.4
C7A—C6A—H6A	119.1	C2B—C7B—H7B	119.4
C8A—C7A—C11A	117.1 (2)	C2C—N1C—C1C	115.99 (17)
C8A—C7A—C6A	124.9 (2)	C2C—N1C—Cu1	121.23 (13)
C11A—C7A—C6A	118.0 (2)	C1C—N1C—Cu1	122.77 (13)
C9A—C8A—C7A	119.8 (2)	N1C—C1C—C2C^{ii}	121.92 (18)
C9A—C8A—H8A	120.1	N1C—C1C—H1C	119.0

C7A—C8A—H8A	120.1	C2C^{ii}—C1C—H1C	119.0
C8A—C9A—C10A	119.5 (2)	N1C—C2C—C1C^{ii}	122.08 (19)
C8A—C9A—H9A	120.3	N1C—C2C—H2C	119.0
C10A—C9A—H9A	120.3	C1C^{ii}—C2C—H2C	119.0
N2A—C10A—C9A	122.3 (2)		

Symmetry codes: (i) -*x*+1, *y*, -*z*-1/2; (ii) -*x*+1, -*y*+2, -*z*.

Figures

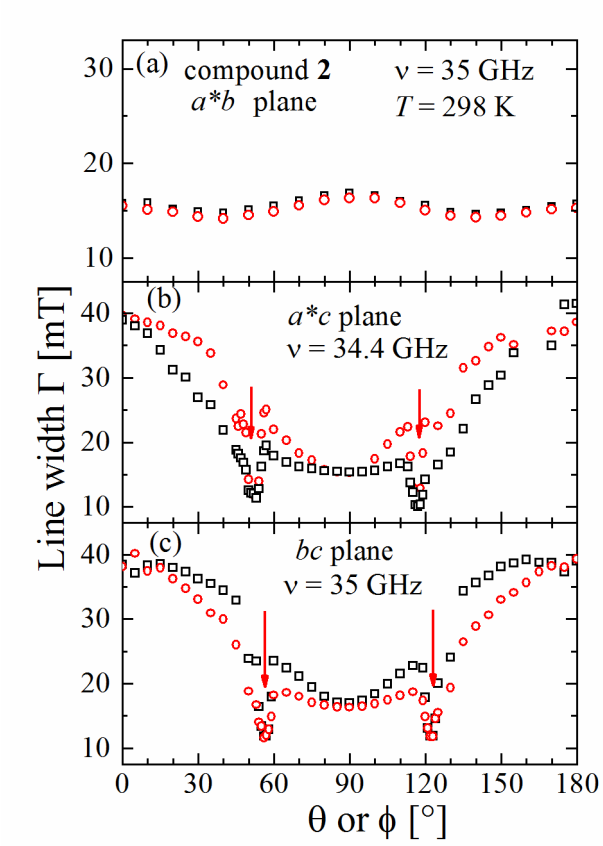

Figure S1: Angular variation of the line width Γ in the crystal planes *a*b*, *a*c* and *bc* at Q-band and 298 K, for compound **2**. Symbols are experimental values; the arrows indicate the angle around which the collapse of the resonances occurs.

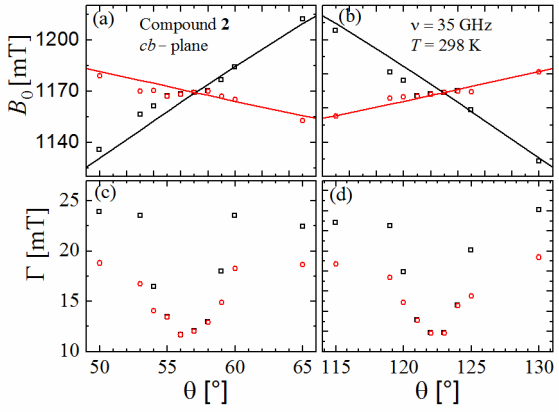

Figure S2: (a) and (b) position of the ±1 ↔ 0 EPR transitions, (c) and (d) linewidths of signals in compound **2** around the magic angles in the *cb*−plane [indicated with green circle in Fig. 6(c)]. Symbols are experimental values; the solid lines are obtained from a global fit of Eq. (4) to the data.

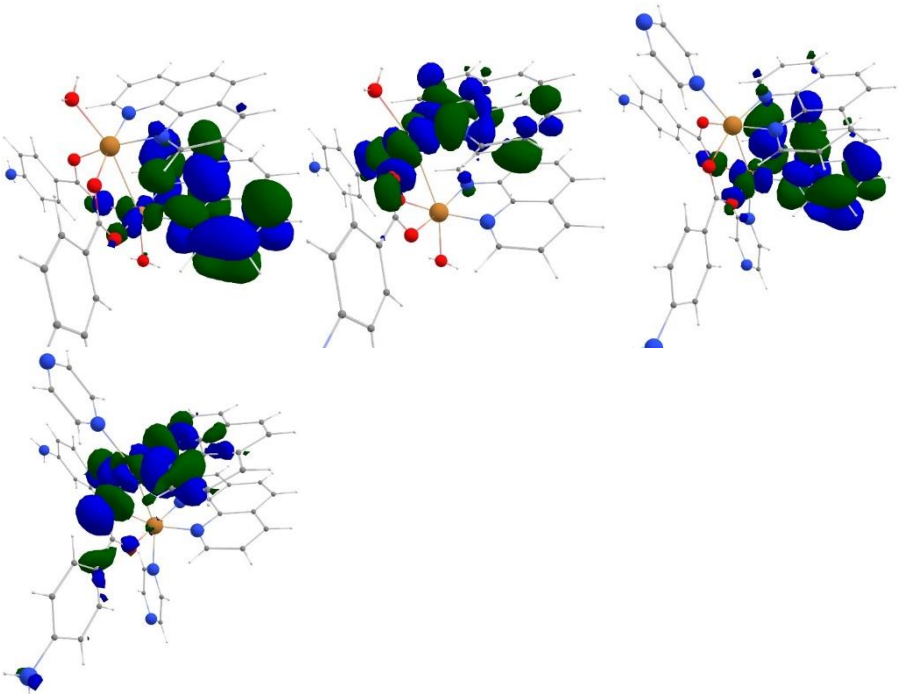

Figure S3: Localized SOMOs of compounds **1** and **2** in BS state (contour value 0.03).

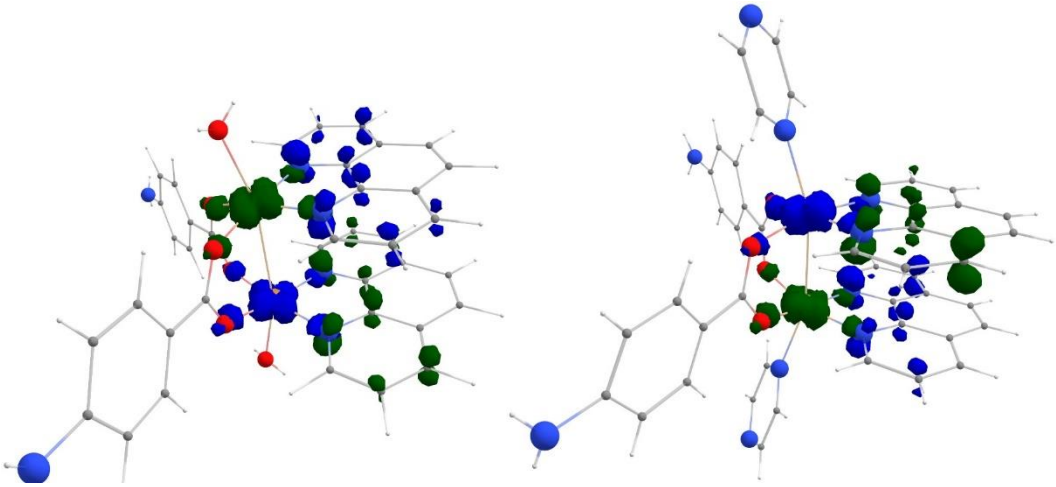

Figure S4: Spin densities calculated for the BS states of compounds **1** and **2** (blue surface is used for positive spin densities and green surface is used for negative spin densities, contour value is 0.01).