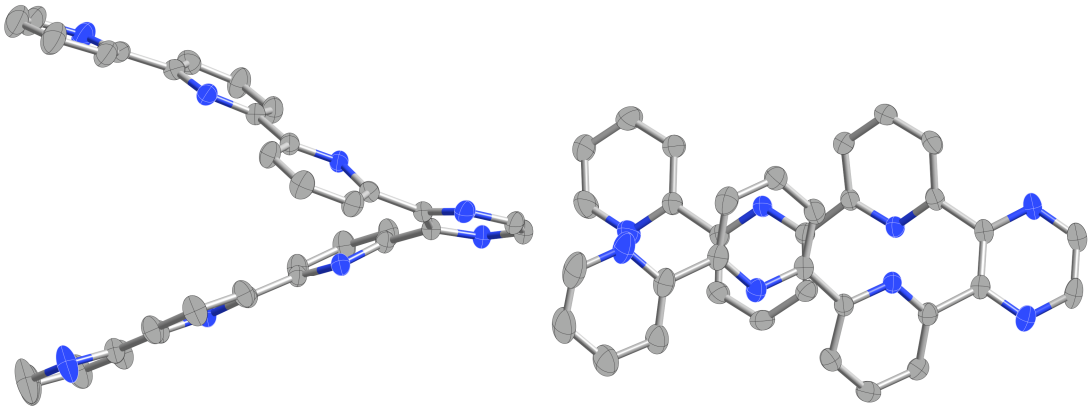
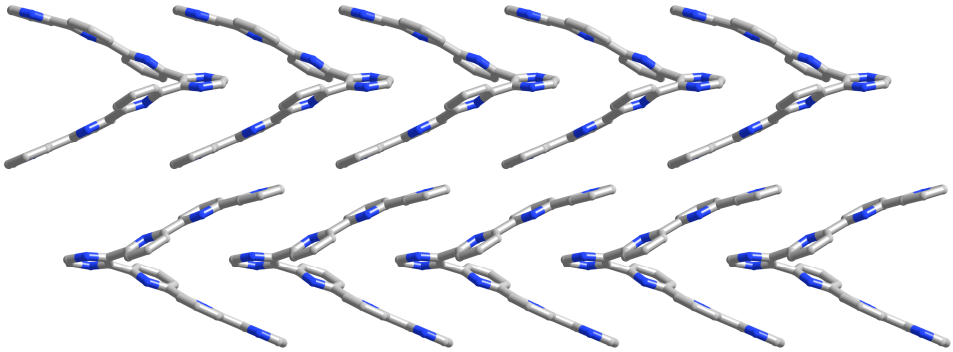
SUPPLEMENTARY INFORMATION

1. Details of the crystal lattice of ligand L

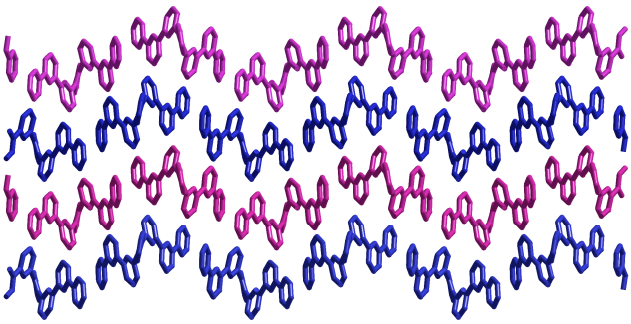
Figure S1



(a) Orthogonal views of a ligand molecule, showing how planarity would involve an impossible overlap of aromatic rings.



(b) A partial view of adjacent, antiparallel but homochiral columns of ligand molecules forming undulating sheets parallel to the *ab* plane.



(c) A partial cross section of several undulating sheets (shown in different colours) of columns of ligand molecules, the chirality alternating from one sheet to the next.

2. Preliminary structural study of the Cu(II) complex 5, $[Cu_2L(NO_3)_2(H_2O)_2](NO_3)_2$.

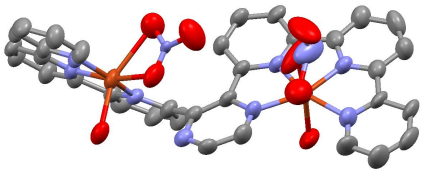


Figure S2 -The cation present in complex **5** – the unbound pyrazine-N is shown in a darker shade of blue than are the other N-donors

3. Magnetic susceptibility of the Cu(II) complex 5.

Taking into account the dimeric structure, the spin Hamiltonian is defined as $H = -J\sum S_1 S_2$ and the magnetic data were analysed using Bleaney-Bowers equation:[62]

$$\chi_m = \frac{N\beta^2 g^2}{3kT}\left[1+\frac{1}{3}\exp\left(\frac{-2J}{kT}\right)\right]^{-1}$$

the J value (singlet-triplet energy gap) characterizes intradimer interactions. The molecular field model was added to obtain other intermolecular magnetic interactions:[62]

$$\chi_m^{corr} = \frac{\chi_m}{1-\frac{2zJ'\chi_m}{Ng^2\beta^2}}$$

where zJ' is the molecular exchange parameter, z is number of the nearest neighbours. Other symbols have their usual meaning: N is the Avogadro number, g is the spectroscopic splitting factor, ® is the Bohr magneton, and k is the Boltzmann constant. The best fit parameters obtained were: g = 2.06, J = -0.87±0.02 cm^{-1}, zJ' = -0.06±0.01 cm^{-1}, R = 4.08×10^{-6} and they confirm only weak intramolecular antiferromagnetic interactions between the Cu(II) ions.

4. IR spectra

6-bromo-2,2':6',2"-terpyridine C:

IR (KBr) ν = $\nu(C\text{-}H)_{py}$ 3055, 3048, 3039 2995; $\gamma(C\text{-}H)_{py}$ overtones 2003-1602; $\nu(C{=}C)_{py}$ 1584, 1578, 1563, 1550, 1474, 1435; $\nu(C{=}N)_{py}$ 1421, 1379, 1265; $\rho(C\text{-}H)_{py}$ 1154, 1131, 1085, 1071, 1062, 1042; $\gamma(C\text{-}H)_{py}$ 993, 986, 846, 838, 785, 774, 744, 678, 631; ν(C-Br) 420 cm^{-1}.

2,3-bis(6-(6-(pyridin-2-yl)pyridin-2-yl)pyridin-2-yl)pyrazine L:

IR (KBr) ν = $\nu(C\text{-}H)_{py}$ 3057, 3045, 2998; $\gamma(C\text{-}H)_{py}$ overtones 1984-1611; $\nu(C{=}C)_{py}$ 1587, 1578, 1568, 1562, 1475, 1457; $\nu(C{=}N)_{py}$ 1428, 1398, 1266; $\rho(C\text{-}H)_{py}$ 1114, 1078, 1038; $\gamma(C\text{-}H)_{py}$ 990, 855, 815, 775, 747, 733, 633 cm^{-1}.

$[Mn_2\mathbf{L}_2](ClO_4)_4$ (**1**):

IR (KBr) ν = $\nu(C\text{-}H)_{py}$ 3076; $\gamma(C\text{-}H)_{py}$ overtones 2014-1688; $\nu(C{=}C)_{py}$ 1596, 1572, 1493, 1478, 1463; $\nu(C{=}N)_{py}$ 1449, 1419, 1378, 1248; δ(OClO) 1143, 1113, 1087; $\rho(C\text{-}H)_{py}$ 1179, 1009, 940; $\gamma(C\text{-}H)_{py}$ 817, 780, 686, 665, 636; ν(ClO) 626 cm^{-1}.

$[Zn_2\mathbf{L}_2](CF_3SO_3)_4$ (**2**):

IR (KBr) ν = $\nu(C\text{-}H)_{py}$ 3089; $\gamma(C\text{-}H)_{py}$ overtones 1931-1627; $\nu(C{=}C)_{py}$ 1599, 1579, 1573, 1537, 1495, 1481, 1465; $\nu(C{=}N)_{py}$ 1456, 1424, 1407, 1381; $\nu(SO_3)$ 1258, 1032; $\nu(CF_3)$ 1229, 1164; $\rho(C\text{-}H)_{py}$ 1088; $\gamma(C\text{-}H)_{py}$ 1014, 817, 780, 761, 640 cm^{-1}.

$[Fe_2\mathbf{L}_2F_2(H_2O)](BF_4)_2\cdot 1.5H_2O$ (**3**):

IR (KBr) ν = $\nu(C\text{-}H)_{py}$ 3078; $\gamma(C\text{-}H)_{py}$ overtones 1750-1632; $\nu(C{=}C)_{py}$ 1600, 1577, 1569, 1497, 1482, 1462; $\nu(C{=}N)_{py}$ 1451, 1416, 1374, 1248; $\rho(C\text{-}H)_{py}$ 1162, 1059; $\nu(BF_4)$ 1084, 522; $\gamma(C\text{-}H)_{py}$ 890, 816, 779, 723, 653 cm^{-1}.

$[Co_2(\mathbf{L})_2F_2(H_2O)](BF_4)_2$ (**4**):

IR (KBr) ν = $\nu(C\text{-}H)_{py}$ 3204, 3089; $\gamma(C\text{-}H)_{py}$ overtones 1845-1696; $\nu(C{=}C)_{py}$ 1599, 1574, 1568, 1531, 1478, 1462; $\nu(C{=}N)_{py}$ 1442, 1419, 1407, 1383; $\rho(C\text{-}H)_{py}$ 1125, 1048; $\nu(BF_4)$ 1085, 522; $\gamma(C\text{-}H)_{py}$ 993, 867, 816, 777, 738, 630 cm^{-1}.

$[Cu_2\mathbf{L}(NO_3)_2(H_2O)_2](NO_3)_2$ (**5**):

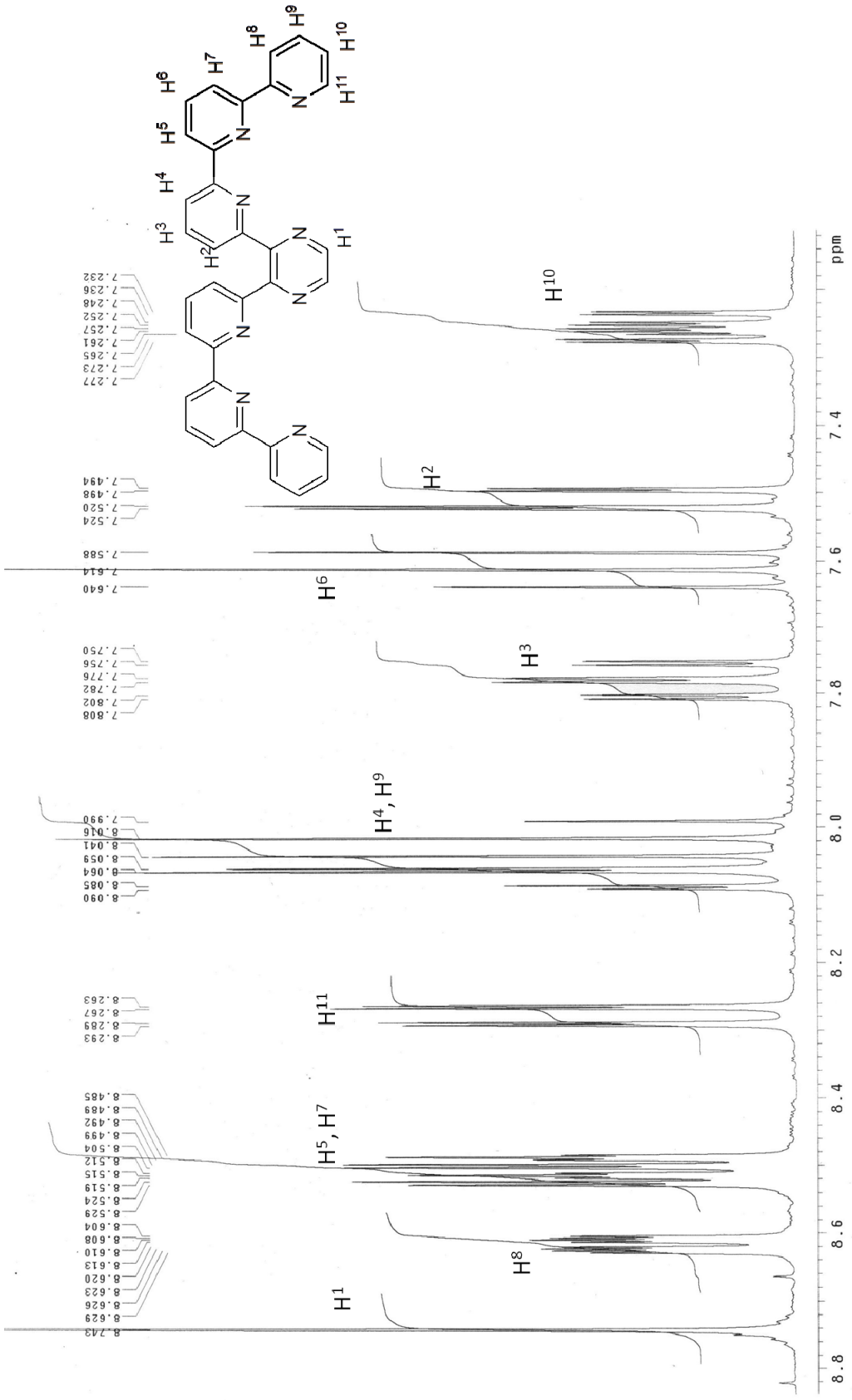
IR (KBr): ν = ν(O-H) 3413; $\nu(C\text{-}H)_{py}$ 3065; $\nu(C{=}C)_{py}$ 1599, 1577, 1569, 1563, 1461; $\nu(C{=}N)_{py}$ 1426, 1277, 1257, 1246; $\nu(NO_3^-)$ 1383, 1375, 1354; $\rho(C\text{-}H)_{py}$ 1193, 1140, 1085, 1043, 1005; $\gamma(C\text{-}H)_{py}$ 818, 780, 684, 658, 651 cm^{-1}.

$[Cd_2\mathbf{L}(NO_3)_4]$ (**6**):

IR (KBr): ν = $\nu(C\text{-}H)_{py}$ 3070; $\nu(C{=}C)_{py}$ 1593, 1578, 1567, 1462; $\nu(C{=}N)_{py}$ 1447, 1425, 1248; $\nu(NO_3^-)$ 1384, 1351; $\rho(C\text{-}H)_{py}$ 1179, 1009, 940; $\gamma(C\text{-}H)_{py}$ 817, 780, 686, 665, 636; 626 cm^{-1}.

5. NMR spectra of L

^{1}H NMR:



^{13}C NMR: