Supplementary Materials

Multifunctional materials based on double-perovskite organic-inorganic hybrid $(CH_3NH_3)_2[KCr(CN)_6]$ showing switchable dielectric, magnetic and semiconducting behaviour

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CAPTIONS OF FIGURES

CAPTIONS OF TABLES

Table S1. Crystal data and structure refinement for **MACr**.

Compound:	**MACr**
Formula	$(CH_3NH_3)_2[KCr(CN)_6]$
Formula weight	311.36
T (K)	300(2)
λ [Å]	0.71073
Crystal system	monoclinic
Space group	*C*2/*c*
a (Å)	13.995(3)
b (Å)	8.052(4)
c (Å)	13.996(4)
β (°)	108.68(2)
V (Å^3)	1494.1(9)
Z	4
μ (Mo K_α) (mm^{-1})	1.04
Crystal size (mm)	0.47×0.27×0.15
θ Range (°)	3.1 to 36.8
Index ranges	--16 ≤ h ≤ 16 -9 ≤ k ≤ 9 -16 ≤ l ≤ 13
Absorption correction	analytical
T_{min}, T_{max}	0.736, 0.880
No. of measured, independent and observed [$I > 2\sigma(I)$] reflections	7450, 1388, 1328
R_{int}	0.051
Goodness-of-fit on F^2	1.13
Final R_1, wR_2 indices [$F^2 > 2\sigma(F^2)$]	R_1 = 0.034, wR_2 = 0.085

Table S2. Geometric parameters (Å,°) for **MACr**.

$(CH_3NH_3)^+$	
N4—C4	1.465(4)
$[KCr(CN)_6]^{2-}$	
Cr1—C1	2.0683(19)
Cr1—C1^{vi}	2.0683(19)
Cr1—C2^{vi}	2.0737(18)
Cr1—C2	2.0738(18)
Cr1—C3^{vi}	2.0831(19)
Cr1—C3	2.0831(19)
N1—C1	1.142(3)
N1—K1^{viii}	2.831(2)
N2—C2	1.144(2)
N3—C3	1.143(2)
N3—K1^{vii}	2.8935(18)
K1—N1^{i}	2.831(2)
K1—N1^{ii}	2.831(2)
K1—N2	2.8680(18)
K1—N2^{iii}	2.8681(18)
K1—N3^{iv}	2.8935(19)
K1—N3^{v}	2.8935(18)
C1—Cr1—C1^{vi}	180.0
C1—Cr1—C2^{vi}	90.04(7)
C1—Cr1—C2	89.96(7)
C1^{vi}—Cr1—C2	90.04(7)
C1^{vi}—Cr1—C2^{vi}	89.96(7)
C1—Cr1—C3	90.82(8)
C1—Cr1—C3^{vi}	89.18(8)
C1^{vi}—Cr1—C3	89.18(8)
C1^{vi}—Cr1—C3^{vi}	90.82(8)
C2^{vi}—Cr1—C2	180.0
C2—Cr1—C3	89.17(7)
C2^{vi}—Cr1—C3	90.83(7)
C3^{vi}—Cr1—C3	180.0
C2^{vi}—Cr1—C3^{vi}	89.17(7)
C1—N1—K1^{viii}	130.56(16)
C2—N2—K1	138.01(14)
C3—N3—K1^{vii}	152.88(16)
N1—C1—Cr1	178.41(18)
N2—C2—Cr1	179.00(17)
N3—C3—Cr1	177.62(16)
C2—Cr1—C3^{vi}	90.83(7)
N1^{i}—K1—N1^{ii}	87.41(11)
N1^{i}—K1—N2	97.28(6)
N1^{ii}—K1—N2	75.91(6)
N1^{i}—K1—N2^{iii}	75.91(6)
N1^{ii}—K1—N2^{iii}	97.28(6)
N1^{i}—K1—N3^{iv}	152.04(6)
N1^{ii}—K1—N3^{iv}	85.07(7)
N1^{i}—K1—N3^{v}	85.07(7)
N1^{ii}—K1—N3^{v}	152.04(6)
N2—K1—N2^{iii}	170.74(8)
N2—K1—N3^{iv}	106.87(5)
N2^{iii}—K1—N3^{iv}	78.40(5)
N2—K1—N3^{v}	78.40(5)
N2^{iii}—K1—N3^{v}	106.87(5)
N3^{iv}—K1—N3^{v}	113.06(9)

Symmetry codes: (i) x-1/2, y-1/2, z; (ii) -x+1/2, y-1/2, -z+1/2; (iii) -x, y, -z+1/2; (iv) -x+1/2, y+1/2, -z+1/2; (v) x-1/2, y+1/2, z; (vi) -x+1/2, -y+1/2, -z; (vii) x+1/2, y-1/2, z; (viii) x+1/2, y+1/2, z.

Table S3. Selected bond lengths and the shortest M···K distances (Å) (where M: Cr^{3+}, Co^{3+}, Fe^{3+}) for **MACr**, **MACo**[2], **MAFe**[1].

	M—C	K—N	C—N	M· · · K	M—C	K—N	C—N	M···K
	LT – phase				HT – phase			
MACr	2.068(3) 2.074(2) 2.083(2)	2.831(2) 2.868(2) 2.894(2)	1.142(3) 1.143(2) 1.144(2)	5.695(2)				
MAFe[1]	1.938(2) 1.937(2) 1.948(2)	2.823(2) 2.854(2) 2.889(2)	1.147(2) 1.151(2) 1.151(2)	5.414(5)	1.930(2)	2.832(2)	1.083(2)	5.739(8)
MACo[2]	1.897(2) 1.901(2) 1.907(3)	2.819(2) 2.854(2) 2.885(2)	1.153(2) 1.153(2) 1.154(2)	5.536(2)	1.862(11)	2.716(12)	1.116(16)	5.694(6)

Table S4. Hydrogen-bond geometry (Å, °) for **MACr**.

D—H···A	D—H	H···A	D···A	D—H···A
N4—H4*C*···N1	0.90(3)	2.13(3)	2.952(3)	152(3)
N4—H4*A*···N2^{ix}	0.79(3)	2.24(3)	2.968(3)	154(3)
N4—H4*B*···N3^{iv}	0.83(4)	2.43(4)	3.155(3)	146(3)

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

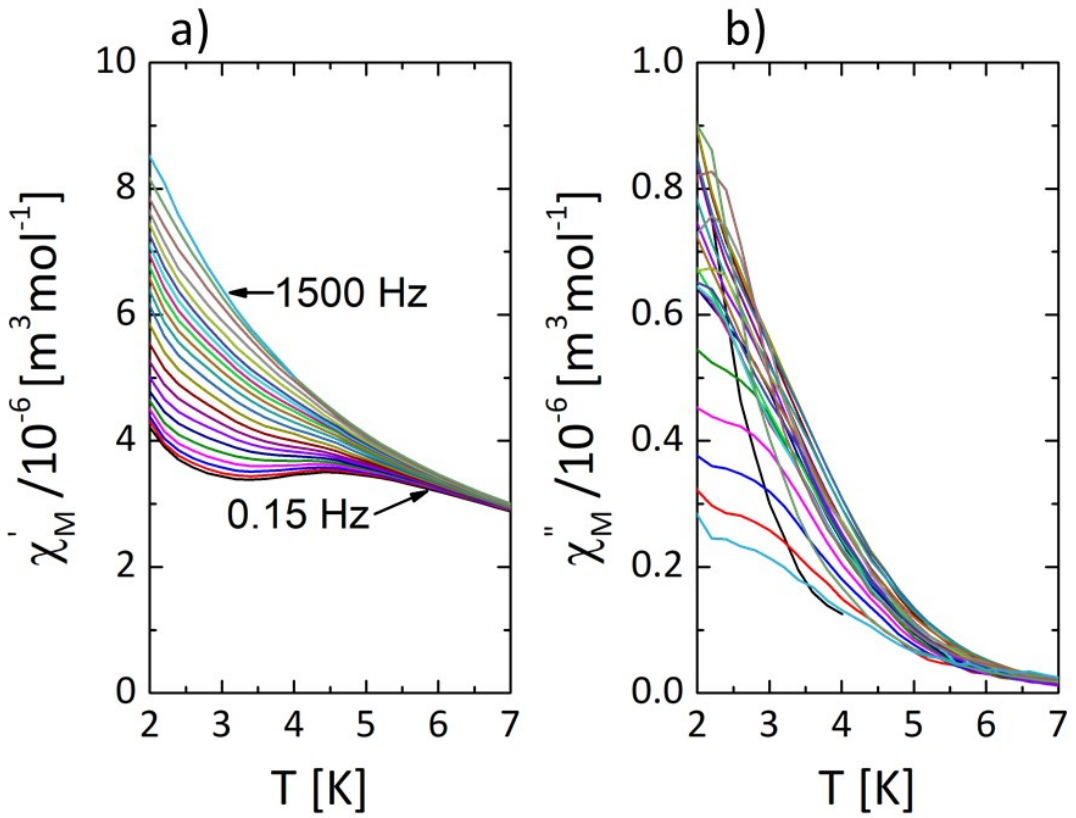

Fig. S1. Temperature dependence of the AC susceptibility components for **MACr** at B_{DC} = 0.35 T for a set of frequencies f = 0.1 – 1500 Hz.

Table S5. Temperature dependence of AC susceptibility parameters for **MACr** at B_{DC} = 0.35 T.

T/K	R(χ′) /%	R(χ″) /%	χ_S	χ_{LF}	α_{LF}	τ_{LF} / s	χ_{HF}	α_{HF}	τ_{HF} /10^{-3} s	x_{LF}	x_{HF}
1.8	0.75	4.9	4.5(1)	7.3(11)	.32(13)	3.6(27)	11.0(10)	.39(2)	5.2(5)	.42	.58
2.0	0.40	2.6	4.0(1)	6.2(2)	.22(4)	1.39(13)	9.6(1)	.41(1)	5.0(2)	.40	.60
2.2	0.33	1.7	3.7(1)	5.6(1)	.18(3)	0.98(4)	8.8(1)	.43(1)	4.9(2)	.38	.62
2.4	0.17	1.5	3.4(1)	4.9(1)	.10(1)	0.74(1)	8.0(1)	.46(1)	4.8(1)	.33	.67
2.6	0.17	1.9	3.3(1)	4.4(1)	.03(1)	0.61(1)	7.4(1)	.48(1)	4.6(1)	.27	.73
2.8	0.18	2.1	3.2(1)	4.1(1)	.01(1)	0.55(1)	7.0(1)	.48(1)	4.4(1)	.25	.75
3.2	0.24	2.8	3.2(1)	3.8(1)	.00	0.44(1)	6.2(1)	.50(1)	4.4(2)	.21	.79
3.6	0.30	3.7	3.2(1)	3.6(1)	.00	0.39(2)	5.6(1)	.52(1)	4.9(5)	.18	.82
4.0	0.27	4.7	3.3(1)	3.6(1)	.00	0.39(2)	5.1(1)	.51(2)	5.1(6)	.17	.83
4.4	0.19	4.9	3.4(1)	3.6(1)	.00	0.41(3)	4.7(1)	.53(2)	4.9(6)	.16	.84
5.0	0.15	6.6	3.4(1)	3.5(4)	.00	0.46(5)	4.1(7)	.53(3)	6.2(10)	.16	.84
5.6	0.17	11	3.3(1)	3.4(5)	.00	0.55(12)	3.8(1)	.55(5)	7.1(23)	.14	.86
6.2	0.13	16	3.1(1)	3.2(1)	.01	0.90(50)	3.4(1)	.57(6)	9.3(42)	.18	.82

[a] Obtained by a two-set Debye model; χ in units of 10^{-6} m^3 mol^{-1}.

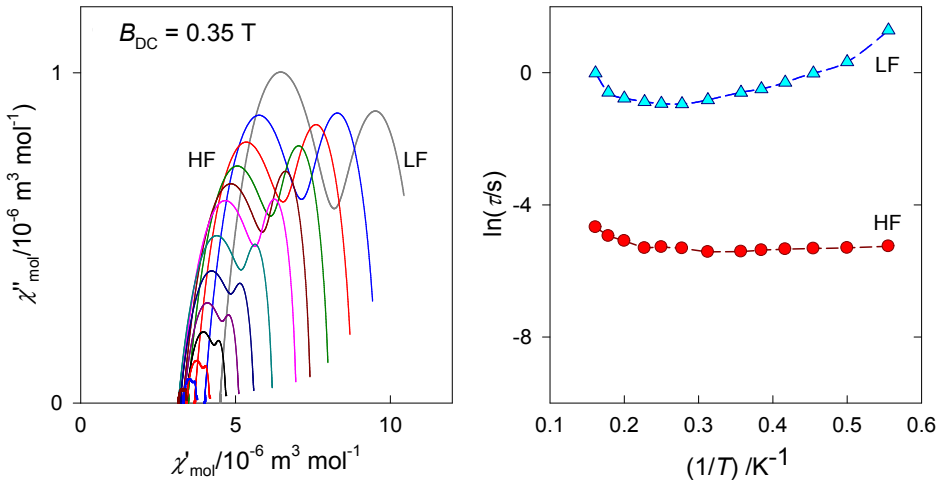

Fig. S2. Argand diagram (left) and the Arrhenius-like plot for **MACr** based upon the fitted data.