Evolution of the structural and magnetic properties of $\text{Sr}_{2-x}\text{Ca}_x\text{CrSbO}_6$

Electronic Supporting Information

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Table S1 – Structural parameters of $\text{Sr}_{1.75}\text{Ca}_{0.25}\text{CrSbO}_6$, $\text{Sr}_{1.25}\text{Ca}_{0.75}\text{CrSbO}_6$ and $\text{Sr}_{0.75}\text{Ca}_{1.25}\text{CrSbO}_6$ at room temperature, as refined from powder x-ray diffraction data collected on I11 (Space Group $P2_1/n$)

		$\text{Sr}_{1.75}\text{Ca}_{0.25}\text{CrSbO}_6$	$\text{Sr}_{1.25}\text{Ca}_{0.75}\text{CrSbO}_6$	$\text{Sr}_{0.75}\text{Ca}_{1.25}\text{CrSbO}_6$
$a/\text{\AA}$		5.565817(6)	5.52108(9)	5.49252(13)
$b/\text{\AA}$		5.543283(7)	5.52028(10)	5.49971(13)
$c/\text{\AA}$		7.831500(9)	7.81132(8)	7.76498(17)
$\beta/^\circ$		90.0031(5)	89.993(4)	90.1360(19)
$V/\text{\AA}^3$		241.6245(5)	238.073(6)	234.558(9)
Sr/Ca	x	0.0013(2)	0.9987(7)	0.9995(5)
$4e$	y	-0.00652(9)	0.01593(8)	0.0264(1)
	z	0.74997(6)	0.2492(9)	0.2504(6)
	$U_{\text{iso}}/\text{\AA}^2$	0.00993(4)	0.01127(6)	0.01159(9)
	Sr Occupancy	0.88	0.635	0.385
	Ca Occupancy	0.12	0.365	0.615
Cr/Sb1	$U_{\text{iso}}/\text{\AA}^2$	0.00278(2)	0.00336(2)	0.00376(3)
$2d (\frac{1}{2} 0 0)$	Cr Occupancy	0.987(1)	0.953(1)	0.955(1)
	Sb Occupancy	0.013(1)	0.047(1)	0.045(1)
Cr/Sb2	$U_{\text{iso}}/\text{\AA}^2$	0.00278(2)	0.00336(2)	0.00376(3)
$2c (\frac{1}{2} 0 \frac{1}{2})$	Cr Occupancy	0.023(1)	0.067(1)	0.065(1)
	Sb Occupancy	0.977 (1)	0.933(1)	0.935(1)
O1	x	0.5464(4)	0.0389(12)	0.075(1)
$4e$	y	0.0014(6)	0.4932(5)	0.4875(6)
	z	0.74994(34)	0.2475(11)	0.2514(9)
	$U_{\text{iso}}/\text{\AA}^2$	0.00741(25)	0.0071(5)	0.0077(4)
O2	x	0.7393(6)	0.7201(23)	0.7290(13)
$4e$	y	0.2642(6)	0.2831(21)	0.2856(15)
	z	0.0236(10)	0.0355(19)	0.0261(11)
	$U_{\text{iso}}/\text{\AA}^2$	0.00741(25)	0.0071(5)	0.0077(4)
O3	x	0.7643(5)	0.2274(20)	0.2091(15)
$4e$	y	0.7631(6)	0.2332(22)	0.2113(14)
	z	0.0210(10)	0.9628(22)	0.9648(13)
	$U_{\text{iso}}/\text{\AA}^2$	0.00741(25)	0.0071(5)	0.0077(4)
O (all) occupancy		1.00	0.9950	0.9950

Table S2 – Structural parameters of $\text{Sr}_{0.50}\text{Ca}_{1.50}\text{CrSbO}_6$, $\text{Sr}_{0.25}\text{Ca}_{1.75}\text{CrSbO}_6$ and $\text{Ca}_2\text{CrSbO}_6$ at room temperature, as refined from powder x-ray diffraction data collected on I11 (Space Group $\text{P2}_1/\text{n}$)

		$\text{Sr}_{0.50}\text{Ca}_{1.50}\text{CrSbO}_6$	$\text{Sr}_{0.25}\text{Ca}_{1.75}\text{CrSbO}_6$	$\text{Ca}_2\text{CrSbO}_6$
$a/\text{\AA}$		5.463425(33)	5.437808(30)	5.407974(12)
$b/\text{\AA}$		5.496490(32)	5.492989(30)	5.494512(11)
$c/\text{\AA}$		7.74688(5)	7.72160(5)	7.691100(17)
$\beta/^\circ$		90.0447(15)	90.0712(12)	90.11100(20)
$V/\text{\AA}^3$		232.6361(24)	230.6427(22)	228.5343(8)
Sr/Ca	x	0.98978(29)	0.99273(32)	0.99167(16)
$4e$	y	0.03094(8)	0.03653(7)	0.04340(7)
	z	0.25095(14)	0.25045(12)	0.25023(9)
	$U_{\text{iso}}/\text{\AA}^2$	0.0092(1)	0.00876(9)	0.00668(7)
	Sr Occupancy	0.255	0.125	0
	Ca Occupancy	0.745	0.875	1
Cr/Sb1	$U_{\text{iso}}/\text{\AA}^2$	0.00337(2)	0.00350(2)	0.00335(2)
$2d (\frac{1}{2} 0 0)$	Cr Occupancy	0.941(1)	0.954(1)	0.952(1)
	Sb Occupancy	0.059(1)	0.046(1)	0.048(1)
Cr/Sb2	$U_{\text{iso}}/\text{\AA}^2$	0.00337(2)	0.00350(2)	0.00335(2)
$2c (\frac{1}{2} 0 \frac{1}{2})$	Cr Occupancy	0.079(1)	0.056(1)	0.058(1)
	Sb Occupancy	0.921(1)	0.944(1)	0.942(1)
O1	x	0.0641(6)	0.0857(5)	0.08743(30)
$4e$	y	0.4830(4)	0.4806(4)	0.47652(30)
	z	0.2444(8)	0.2507(5)	0.25043(25)
	$U_{\text{iso}}/\text{\AA}^2$	0.00636(26)	0.00656(22)	0.0072(4)
O2	x	0.7067(7)	0.7065(5)	0.70787(33)
$4e$	y	0.2937(6)	0.2938(5)	0.29601(32)
	z	0.0408(9)	0.0384(7)	0.04261(31)
	$U_{\text{iso}}/\text{\AA}^2$	0.00636(26)	0.00656(22)	0.0068(4)
O3	x	0.2178(6)	0.2116(5)	0.20344(33)
$4e$	y	0.2180(6)	0.2116(5)	0.21001(33)
	z	0.9586(9)	0.9591(7)	0.95676(32)
	$U_{\text{iso}}/\text{\AA}^2$	0.00636(26)	0.00656(22)	0.0084(5)
O (all) occupancy		0.995	1.00	1.00

Table S3 – Bond lengths (Å) in $\text{Sr}_{1.75}\text{Ca}_{0.25}\text{CrSbO}_6$, $\text{Sr}_{1.25}\text{Ca}_{0.75}\text{CrSbO}_6$ and $\text{Sr}_{0.75}\text{Ca}_{1.25}\text{CrSbO}_6$ at room temperature.

	$\text{Sr}_{1.75}\text{Ca}_{0.25}\text{CrSbO}_6$	$\text{Sr}_{1.25}\text{Ca}_{0.75}\text{CrSbO}_6$	$\text{Sr}_{0.75}\text{Ca}_{1.25}\text{CrSbO}_6$
Sr/Ca – O1	2.532(3)	2.556(7)	2.347(7)
Sr/Ca – O1	2.741(3)	2.644(3)	2.570(3)
Sr/Ca – O1	2.828(3)	2.894(3)	2.993(3)
Sr/Ca – O1	3.035(3)	2.971(7)	3.162(7)
Sr/Ca – O2	2.589(6)	2.437(16)	2.520(8)
Sr/Ca – O2	2.696(6)	2.707(16)	2.689(10)
Sr/Ca – O2	2.828(6)	2.779(16)	2.694(10)
Sr/Ca – O2	2.995(6)	3.175(16)	3.130(8)
Sr/Ca – O3	2.596(6)	2.489(17)	2.409(7)
Sr/Ca – O3	2.717(6)	2.635(16)	2.700(13)
Sr/Ca – O3	2.807(6)	2.836(16)	2.722(13)
Sr/Ca – O3	2.987(6)	3.128(18)	3.231(7)
<Sr/Ca – O>	2.779	2.771	2.764
Cr/Sb1 – O1	1.975(3)	1.984(9)	1.976(7)
Cr/Sb1 – O2	1.988(3)	1.999(7)	2.022 (7)
Cr/Sb1 – O3	1.979(3)	2.002(7)	1.994(6)
<Cr/Sb1-O>	1.981	1.995	1.997
Cr/Sb2 – O1	1.974(3)	1.946(9)	1.995(7)
Cr/Sb2 – O2	1.961(3)	1.957(8)	1.910(7)
Cr/Sb2 – O3	1.969(3)	1.974(8)	1.979(6)
<Cr/Sb2 – O>	1.968	1.959	1.961

Table S4 – Bond lengths (Å) in $\text{Sr}_{0.50}\text{Ca}_{1.50}\text{CrSbO}_6$, $\text{Sr}_{0.25}\text{Ca}_{1.75}\text{CrSbO}_6$ and $\text{Ca}_2\text{CrSbO}_6$ at room temperature.

	$\text{Sr}_{0.50}\text{Ca}_{1.50}\text{CrSbO}_6$	$\text{Sr}_{0.25}\text{Ca}_{1.75}\text{CrSbO}_6$	$\text{Ca}_2\text{CrSbO}_6$
Sr/Ca – O1	2.452(4)	2.313(3)	2.3057(18)
Sr/Ca – O1	2.518(2)	2.491(2)	2.4355(17)
Sr/Ca – O1	3.038(4)	3.095(2)	3.1532(18)
Sr/Ca – O1	3.039(2)	3.160(3)	3.1575(17)
Sr/Ca – O2	2.336(6)	2.370(4)	2.357(2)
Sr/Ca – O2	2.669(5)	2.663(4)	2.612(2)
Sr/Ca – O2	2.715(6)	2.675(5)	2.682(2)
Sr/Ca – O2	3.324(6)	3.309(4)	3.347(2)
Sr/Ca – O3	2.406(5)	2.389(4)	2.362(2)
Sr/Ca – O3	2.589(5)	2.618(4)	2.607(2)
Sr/Ca – O3	2.783(6)	2.722(5)	2.693(2)
Sr/Ca – O3	3.251(5)	3.285(4)	3.337(2)
<Sr/Ca – O>	2.760	2.758	2.754
Cr/Sb1 – O1	2.013(6)	1.984(4)	1.9819(19)
Cr/Sb1 – O2	1.995(4)	1.988(3)	2.0037 (18)
Cr/Sb1 – O3	1.979(4)	1.977(3)	2.0030(18)
<Cr/Sb1-O>	1.996	1.983	1.996
Cr/Sb2 – O1	1.927(6)	1.993(4)	1.987(2)
Cr/Sb2 – O2	1.988(4)	1.980(3)	1.9651(18)
Cr/Sb2 – O3	1.981(4)	1.983(3)	1.9650(18)
<Cr/Sb2 – O>	1.965	1.985	1.972

Table S5 – Key bond angles (°) in $\text{Sr}_{1.75}\text{Ca}_{0.25}\text{CrSbO}_6$, $\text{Sr}_{1.25}\text{Ca}_{0.75}\text{CrSbO}_6$ and $\text{Sr}_{1.25}\text{Ca}_{0.75}\text{CrSbO}_6$ at room temperature.

	$\text{Sr}_{1.75}\text{Ca}_{0.25}\text{CrSbO}_6$	$\text{Sr}_{1.25}\text{Ca}_{0.75}\text{CrSbO}_6$	$\text{Sr}_{0.75}\text{Ca}_{1.25}\text{CrSbO}_6$
O1 – Cr/Sb1 – O2	90.09(25)	93.3(4)	93.44(28)
O1 – Cr/Sb1 – O3	90.70(25)	94.3(6)	90.8(4)
O2 – Cr/Sb1 – O3	90.97(11)	91.5(9)	93.4(5)
<O – Cr/Sb1 – O>	90.59	93.03	92.55
O1 – Cr/Sb2 – O2	90.05(25)	93.7(4)	91.95(29)
O1 – Cr/Sb2 – O3	90.41(25)	93.6(6)	90.63(33)
O2 – Cr/Sb2 – O3	90.41(11)	93.8(9)	91.6(5)
<O – Cr/Sb2 – O>	90.29	93.7	91.39
Cr/Sb1–O1–Cr/Sb2	164.95(14)	167.2(4)	155.72(35)
Cr/Sb1–O2–Cr/Sb2	167.8(4)	158.5(8)	162.5(4)
Cr/Sb1–O3–Cr/Sb2	168.6(4)	160.9(10)	156.03(34)
<Cr/Sb1–O– Cr/Sb2>	167.12	162.2	158.08

Table S6 – Key bond angles (°) in $\text{Sr}_{0.50}\text{Ca}_{1.50}\text{CrSbO}_6$, $\text{Sr}_{0.25}\text{Ca}_{1.75}\text{CrSbO}_6$ and $\text{Ca}_2\text{CrSbO}_6$ at room temperature.

	$\text{Sr}_{0.50}\text{Ca}_{1.50}\text{CrSbO}_6$	$\text{Sr}_{0.25}\text{Ca}_{1.75}\text{CrSbO}_6$	$\text{Ca}_2\text{CrSbO}_6$
O1 – Cr/Sb1 – O2	91.12(23)	91.85(18)	91.68(8)
O1 – Cr/Sb1 – O3	92.95(24)	90.02(17)	90.33(9)
O2 – Cr/Sb1 – O3	91.33(14)	90.32(12)	90.48(8)
<O – Cr/Sb1 – O>	91.80	90.73	90.83
O1 – Cr/Sb2 – O2	92.18(21)	90.65(17)	90.51(9)
O1 – Cr/Sb2 – O3	90.72(25)	91.31(18)	91.20(9)
O2 – Cr/Sb2 – O3	93.65(15)	91.99(12)	90.93(8)
<O – Cr/Sb2 – O>	92.18	91.32	90.88
Cr/Sb1–O1–Cr/Sb2	158.79(19)	152.19(14)	151.40(9)
Cr/Sb1–O2–Cr/Sb2	153.16(31)	153.83(24)	152.46(12)
Cr/Sb1–O3–Cr/Sb2	156.32(34)	154.72(24)	152.55 (12)
<Cr/Sb1–O– Cr/Sb2>	156.09	153.58	152.14

Table S7 – Compositional analysis of the phases present in each sample

Nominal composition	Impurity composition	Impurity percentage (mol %)	Adjusted perovskite composition
$\text{Sr}_2\text{CrSbO}_6$	n/a	0	$\text{Sr}_2\text{CrSbO}_6$
$\text{Sr}_{1.75}\text{Ca}_{0.25}\text{CrSbO}_6$	$\text{Sr}_{0.84}\text{Ca}_{1.16}\text{Sb}_2\text{O}_7$	0.91	$\text{Sr}_{1.76}\text{Ca}_{0.24}\text{Cr}_{1.01}\text{Sb}_{0.99}\text{O}_6$
$\text{Sr}_{1.5}\text{Ca}_{0.5}\text{CrSbO}_6$	$\text{Sr}_{0.61}\text{Ca}_{1.39}\text{Sb}_2\text{O}_7$	1.81	$\text{Sr}_{1.55}\text{Ca}_{0.45}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_6$ and $\text{Sr}_{1.33}\text{Ca}_{0.67}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_6$
$\text{Sr}_{1.25}\text{Ca}_{0.75}\text{CrSbO}_6$	$\text{Sr}_{0.41}\text{Ca}_{1.59}\text{Sb}_2\text{O}_7$	2.59	$\text{Sr}_{1.27}\text{Ca}_{0.73}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_{5.97}$
SrCaCrSbO_6	$\text{Sr}_{0.29}\text{Ca}_{1.71}\text{Sb}_2\text{O}_7$	4.16	$\text{Sr}_{1.03}\text{Ca}_{0.97}\text{Cr}_{1.04}\text{Sb}_{0.95}\text{O}_{5.94}$
$\text{Sr}_{0.75}\text{Ca}_{1.25}\text{CrSbO}_6$	$\text{Sr}_{0.28}\text{Ca}_{1.72}\text{Sb}_2\text{O}_7$	2.56	$\text{Sr}_{0.77}\text{Ca}_{1.23}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_{5.97}$
$\text{Sr}_{0.5}\text{Ca}_{1.5}\text{CrSbO}_6$	$\text{Sr}_{0.26}\text{Ca}_{1.74}\text{Sb}_2\text{O}_7$	2.33	$\text{Sr}_{0.51}\text{Ca}_{1.49}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_{5.97}$
$\text{Sr}_{0.25}\text{Ca}_{1.75}\text{CrSbO}_6$	$\text{Sr}_{0.19}\text{Ca}_{1.81}\text{Sb}_2\text{O}_7$	1.16	$\text{Sr}_{0.25}\text{Ca}_{1.75}\text{Cr}_{1.01}\text{Sb}_{0.99}\text{O}_6$
$\text{Ca}_2\text{CrSbO}_6$	$\text{Ca}_2\text{Sb}_2\text{O}_7$	1.02	$\text{Ca}_2\text{Cr}_{1.01}\text{Sb}_{0.99}\text{O}_6$

Figure captions

Figure S1 - Observed (red crosses) and calculated (green line) neutron powder diffraction profiles of $\text{Sr}_2\text{CrSbO}_6$ at 25 K. Data were collected on D2b and fitted using the space group $I\bar{1}$. Reflection markers for $\text{Sr}_2\text{CrSbO}_6$ are shown in black.

Figure S2 - Observed (red crosses) and calculated (green line) neutron powder diffraction profiles of nominal $\text{Sr}_{1.5}\text{Ca}_{0.5}\text{CrSbO}_6$ collected at room temperature on D2b. The data were refined using a two-phase model where both perovskite phases are of the space group $P2_1/n$. Reflection markers are shown top to bottom for $\text{Sr}_{0.61}\text{Ca}_{1.39}\text{Sb}_2\text{O}_7$ (blue), $P2_1/n$ $\text{Sr}_{1.33}\text{Ca}_{0.67}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_6$ (red) and $P2_1/n$ $\text{Sr}_{1.55}\text{Ca}_{0.45}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_6$ (black).

Figure S3 - Observed (red crosses) and calculated (green line) neutron powder diffraction profiles of nominal $\text{Sr}_{1.5}\text{Ca}_{0.5}\text{CrSbO}_6$ collected at 25 K on D2b. The data were refined using a two-phase model where both perovskite phases are of the space group $P2_1/n$. Reflection markers are shown top to bottom for $\text{Sr}_{0.61}\text{Ca}_{1.39}\text{Sb}_2\text{O}_7$ (blue), $P2_1/n$ $\text{Sr}_{1.33}\text{Ca}_{0.67}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_6$ (red) and $P2_1/n$ $\text{Sr}_{1.55}\text{Ca}_{0.45}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_6$ (black).

Figure S4 - Observed (red crosses) and calculated (green line) neutron powder diffraction profiles of nominal $\text{Sr}_{1.5}\text{Ca}_{0.5}\text{CrSbO}_6$ collected at 2 K on D2b. The data were refined using a two-phase model where both perovskite phases are of the space group $P2_1/n$. Reflection markers are shown top to bottom for $\text{Sr}_{0.61}\text{Ca}_{1.39}\text{Sb}_2\text{O}_7$ (blue), $P2_1/n$ $\text{Sr}_{1.33}\text{Ca}_{0.67}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_6$ (red) and $P2_1/n$ $\text{Sr}_{1.55}\text{Ca}_{0.45}\text{Cr}_{1.02}\text{Sb}_{0.98}\text{O}_6$ (black).

Figure S5 - Observed (red crosses) and calculated (green line) neutron powder diffraction profiles of SrCaCrSbO_6 at room temperature. Data were collected on D2b and fitted using the space group $P2_1/n$. Reflection markers are shown top to bottom for $\text{Sr}_{0.29}\text{Ca}_{1.71}\text{Sb}_2\text{O}_7$ (red) and SrCaCrSbO_6 (black).

Figure S6 - Observed (red crosses) and calculated (green line) neutron powder diffraction profiles of SrCaCrSbO_6 at 25 K. Data were collected on D2b and fitted using the space group $P2_1/n$. Reflection markers are shown top to bottom for $\text{Sr}_{0.29}\text{Ca}_{1.71}\text{Sb}_2\text{O}_7$ (red) and SrCaCrSbO_6 (black).

Figure S7 - Observed (red crosses) and calculated (green line) neutron powder diffraction profiles of SrCaCrSbO_6 at 2 K. Data were collected on D2b and fitted using the space group

$P2_1/n$. Reflection markers are shown top to bottom for $\text{Sr}_{0.29}\text{Ca}_{1.71}\text{Sb}_2\text{O}_7$ (blue), magnetic SrCaCrSbO_6 (red) and structural SrCaCrSbO_6 (black).

Figure S8 - Observed (red crosses) and calculated (green line) neutron powder diffraction profiles of SrCaCrSbO_6 at 2 K. Data were collected on D1b and fitted using the space group $P2_1/n$. Reflection markers are shown top to bottom for magnetic SrCaCrSbO_6 (blue), $\text{Sr}_{0.29}\text{Ca}_{1.71}\text{Sb}_2\text{O}_7$ (red) and structural SrCaCrSbO_6 (black).

Figure S9 - Observed (red crosses) and calculated (green line) synchrotron x-ray diffraction profiles of a) $\text{Sr}_{1.75}\text{Ca}_{0.25}\text{CrSbO}_6$ and b) $\text{Ca}_2\text{CrSbO}_6$ collected at room temperature on I11 and fitted using the space group $P2_1/n$. Reflection markers are shown top to bottom for $\text{Sr}_{1-x}\text{Ca}_x\text{Sb}_2\text{O}_7$ (red) and structural $\text{Sr}_{1-x}\text{Ca}_x\text{CrSbO}_6$ (black). Inset is a close-up of the XRD pattern between 10 and 22°.

Figure S1

Figure S2

Figure S3

Figure S4

Figure S5

Figure S6

Figure S7

Figure S8

a)

b)

Figure S9