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Supporting Information

Magnetic cations doped into a Double Perovskite Semiconductor

Lun Jin[1]*, Danrui Ni[1], Xin Gui[1], Daniel B. Straus[1], Qiang Zhang[2] and Robert J. Cava[1]*

[1]Department of Chemistry, Princeton University, Princeton, NJ, 08544, USA

[2]Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, TN, 37831, USA

*E-mails of corresponding authors: ljin@princeton.edu; rcava@princeton.edu

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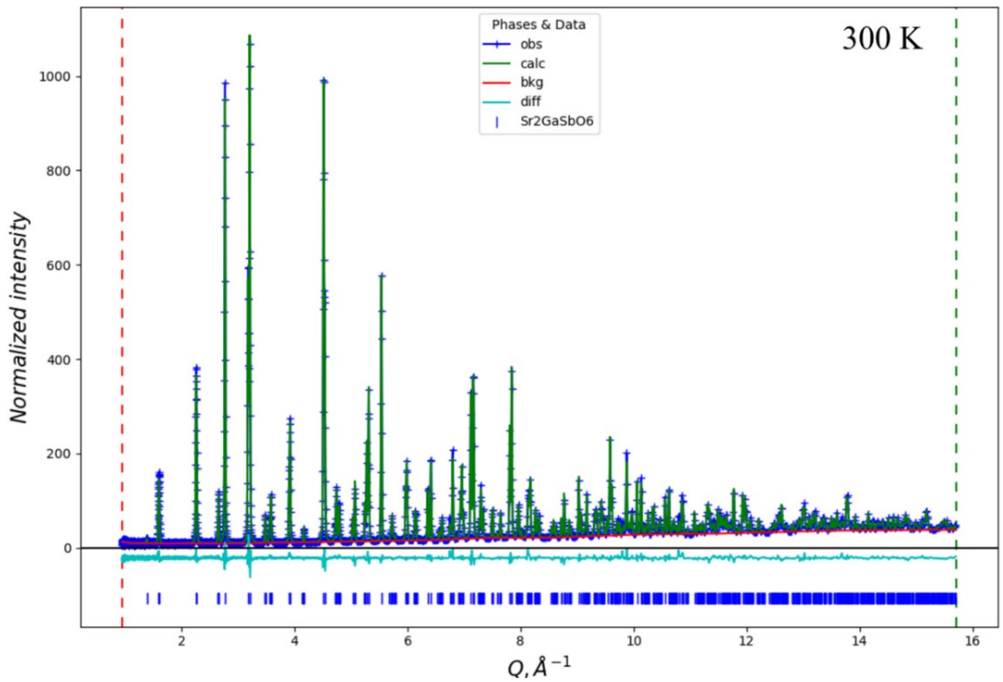
1. **Rietveld co-refinement of ambient temperature neutron and X-ray powder diffraction data**



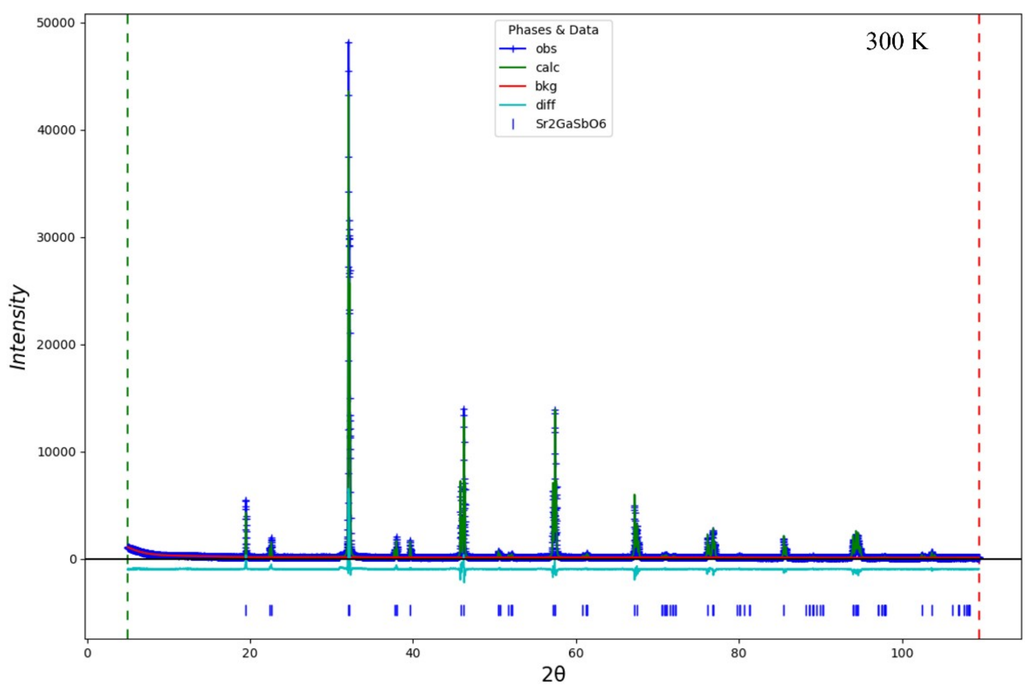


Figure S1. Observed, calculated and difference plots from the Rietveld co-refinement of Sr_2GaSbO_6 (space group *I*4/*m*) against neutron (top) and X-ray (bottom) powder diffraction data collected at 300 K.

Atoms	*x*/*a*	*y*/*b*	*z*/*c*	S.O.F.	U_{iso} equiv. (Å^2)
Sr1	0	0.5	0.25	1	0.00628
Ga1	0	0	0	1	0.00068
Sb1	0.5	0.5	0	1	0.00503
O1	0	0	0.2495(3)	1	0.00967
O2	0.2243(3)	0.2771(3)	0	1	0.00773

Sr_2GaSbO_6 space group *I*4/*m* (#87)
Formula weight: 462.72 g mol^{-1}, Z = 2
a = 5.54042(4) Å, *c* = 7.90283(4) Å, Volume = 242.587(5) Å^3
Radiation source: time of flight neutrons and Cu K-α
Temperature: 300 K
wR = 7.303%; *GOF* = 3.01

Table S1. Structural parameters and crystallographic positions from the co-refinement of neutron and X-ray powder diffraction data collected from Sr_2GaSbO_6 at 300 K.

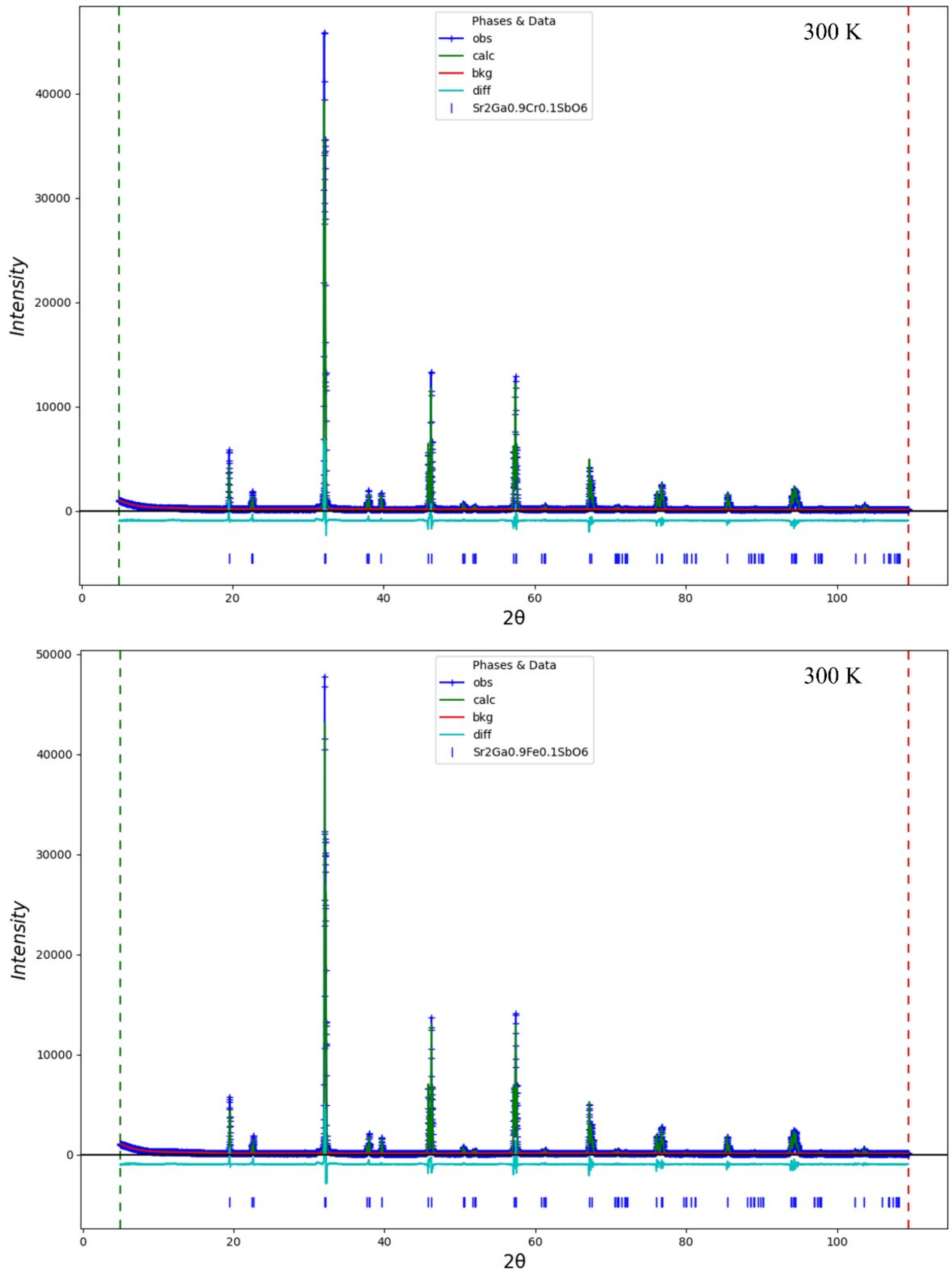


Figure S2. Observed, calculated and difference plots from the Rietveld co-refinement of $Sr_2Ga_{0.9}Cr_{0.1}SbO_6$ (top) and $Sr_2Ga_{0.9}Fe_{0.1}SbO_6$ (bottom) in space group *I*4/*m* against X-ray powder diffraction data collected at 300 K.

Table S2 Selected bond lengths for the double perovskites from the Rietveld co-refinement of neutron and X-ray powder diffraction data.

Undoped			**Doped**		
Sr_2GaSbO_6			**$Sr_2Ga_{0.9}Cr_{0.1}SbO_6$**		
Sr(1)	O(1)	2.770(1) (Å) × 4	**Sr(1)**	O(1)	2.772(1) (Å) × 4
	O(2)	2.932(1) (Å) × 4		O(2)	2.929(1) (Å) × 4
	O(2)	2.641(1) (Å) × 4		O(2)	2.645(1) (Å) × 4
Ga(1)	O(1)	1.982(1) (Å) × 2	**Ga/Cr(1)**	O(1)	1.972(1) (Å) × 2
	O(2)	1.972(1) (Å) × 4		O(2)	1.973(1) (Å) × 4
Sb(1)	O(1)	1.971(1) (Å) × 2	**Sb(1)**	O(1)	1.980(1) (Å) × 2
	O(2)	1.968(1) (Å) × 4		O(2)	1.969(1) (Å) × 4
			$Sr_2Ga_{0.9}Fe_{0.1}SbO_6$		
			Sr(1)	O(1)	2.773(1) (Å) × 4
				O(2)	2.935(1) (Å) × 4
				O(2)	2.643(1) (Å) × 4
			Ga/Fe(1)	O(1)	1.969(1) (Å) × 2
				O(2)	1.974(1) (Å) × 4
			Sb(1)	O(1)	1.987(1) (Å) × 2
				O(2)	1.970(1) (Å) × 4

2. Scanning Electron Microscope Characterization

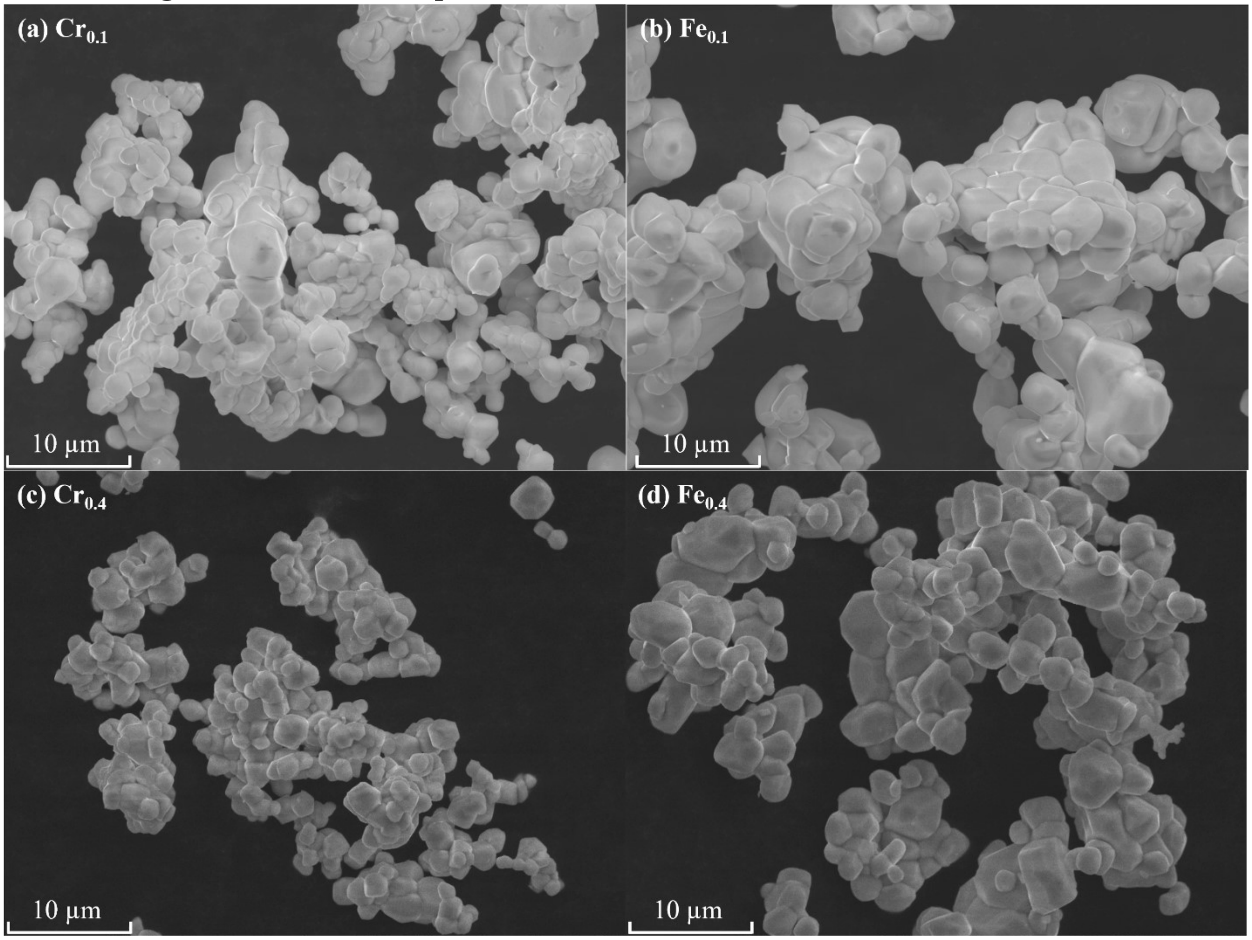


Figure S3. The FEI XL30 field-emission gun scanning electron microscope (SEM) photos of particle morphology of (a)-(b) $Sr_2Ga_{0.9}M_{0.1}SbO_6$ (M = Cr/Fe), (c)-(d) $Sr_2Ga_{0.6}M_{0.4}SbO_6$ (M = Cr/Fe).

3. Magnetization data for the $Sr_2Ga_{1-x}M_xSbO_6$ series (M = Cr/Fe) phases

Table S3. The Curie constant and Weiss temperature extracted from the fitting of paramagnetic susceptibility to the Curie-Weiss law, the observed effective moment per formula unit, and the calculated effective moment per formula unit predicted based on the spin-only formula for each composition of $Sr_2Ga_{1-x}M_xSbO_6$ series (M = Cr/Fe).

Composition	Curie constant C (cm^3 K mol^{-1})	Observed effective moment per formula unit $\mu_{eff.obs}$ (μ_B/f.u.)	Calculated effective moment per formula unit $\mu_{eff.cal}$ (μ_B/f.u.)	Weiss temperature θ (K)
$Sr_2Ga_{1-x}M_xSbO_6$				
M = Cr^{3+}				
x = 0.1	0.165(3)	1.149	1.225	0.35(5)
x = 0.2	0.365(1)	1.709	1.732	−2.76(2)
x = 0.3	0.482(2)	1.964	2.121	−2.98(4)
x = 0.4	0.719(1)	2.398	2.449	−8.69(3)
M = Fe^{3+}				
x = 0.1	0.364(2)	1.706	1.871	−16.33(27)
x = 0.2	0.789(4)	2.512	2.646	−44.57(35)
x = 0.3	1.284(3)	3.205	3.240	−80.70(64)
x = 0.4	1.642(1)	3.624	3.742	−108.66(36)

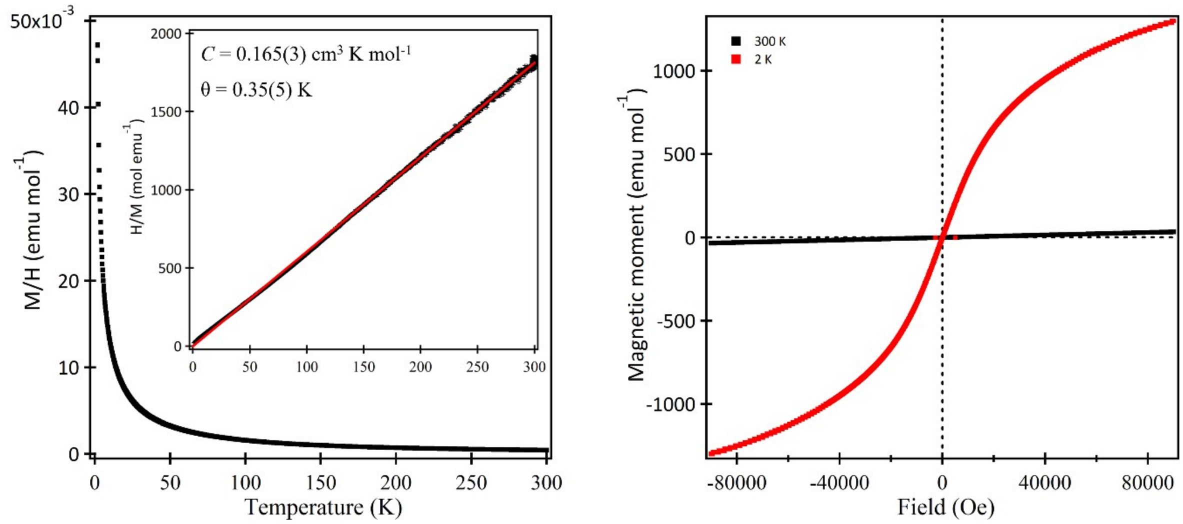


Figure S4. The magnetization data collected from $Sr_2Ga_{0.9}Cr_{0.1}SbO_6$.

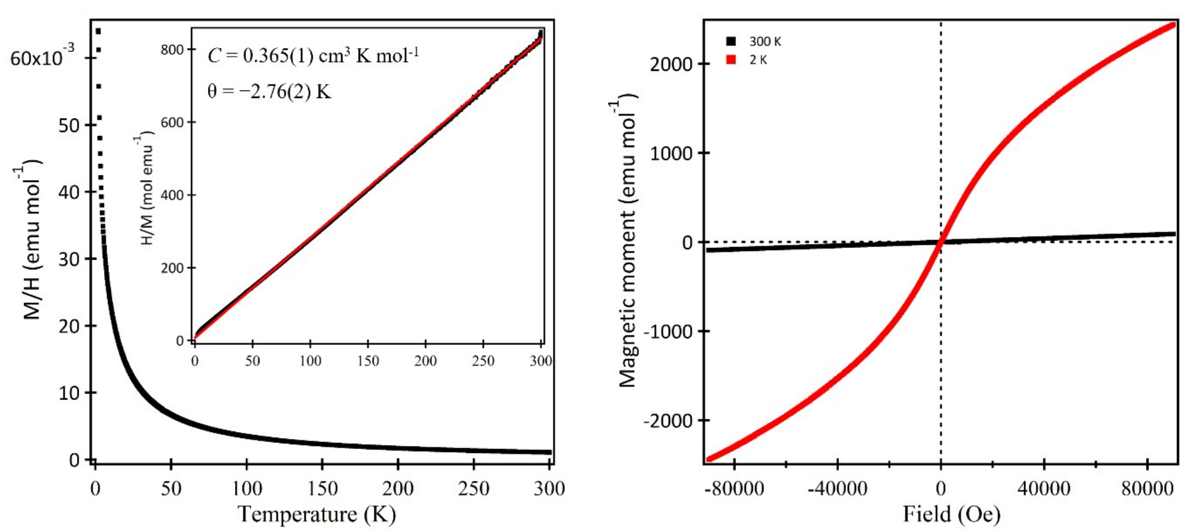


Figure S5. The magnetization data collected from $Sr_2Ga_{0.8}Cr_{0.2}SbO_6$.

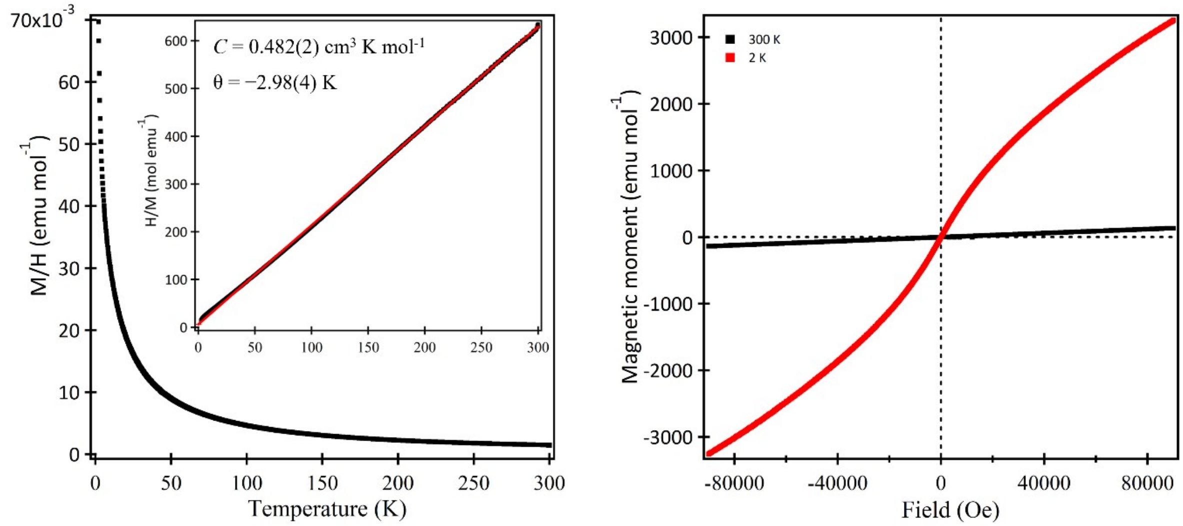


Figure S6. The magnetization data collected from $Sr_2Ga_{0.7}Cr_{0.3}SbO_6$.

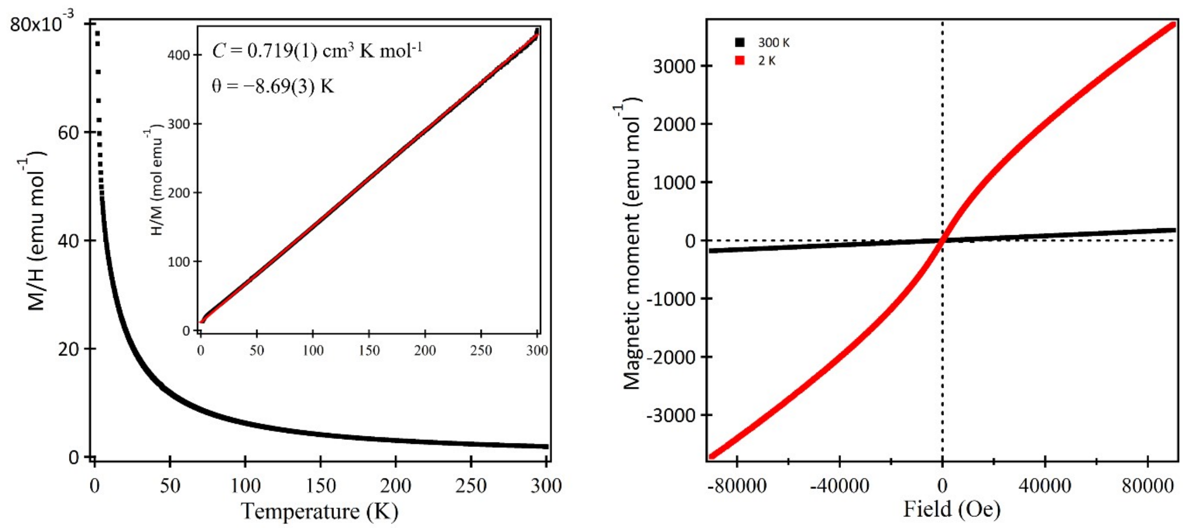


Figure S7. The magnetization data collected from $Sr_2Ga_{0.6}Cr_{0.4}SbO_6$.

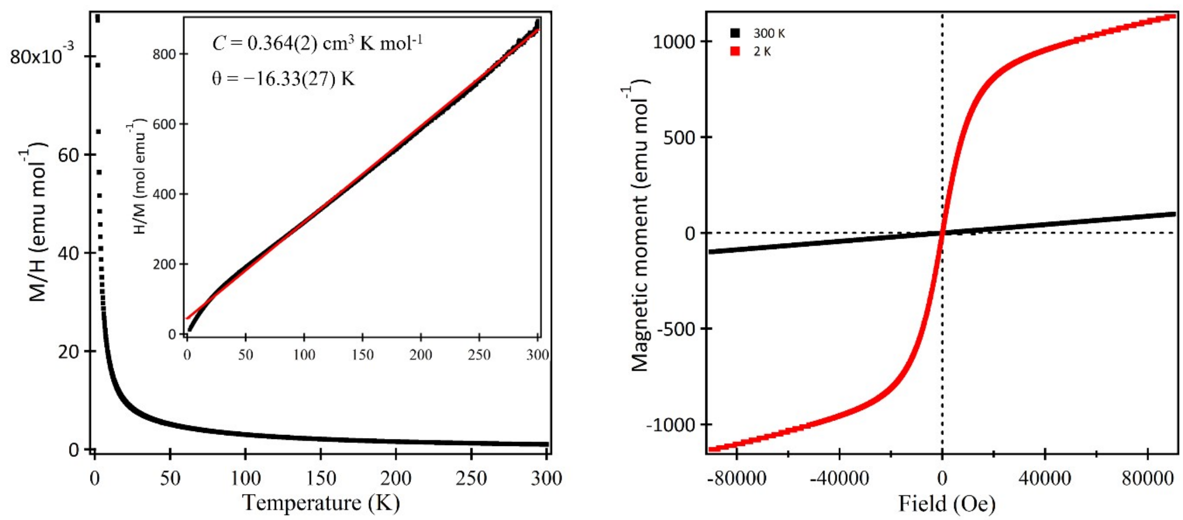


Figure S8. The magnetization data collected from $Sr_2Ga_{0.9}Fe_{0.1}SbO_6$.

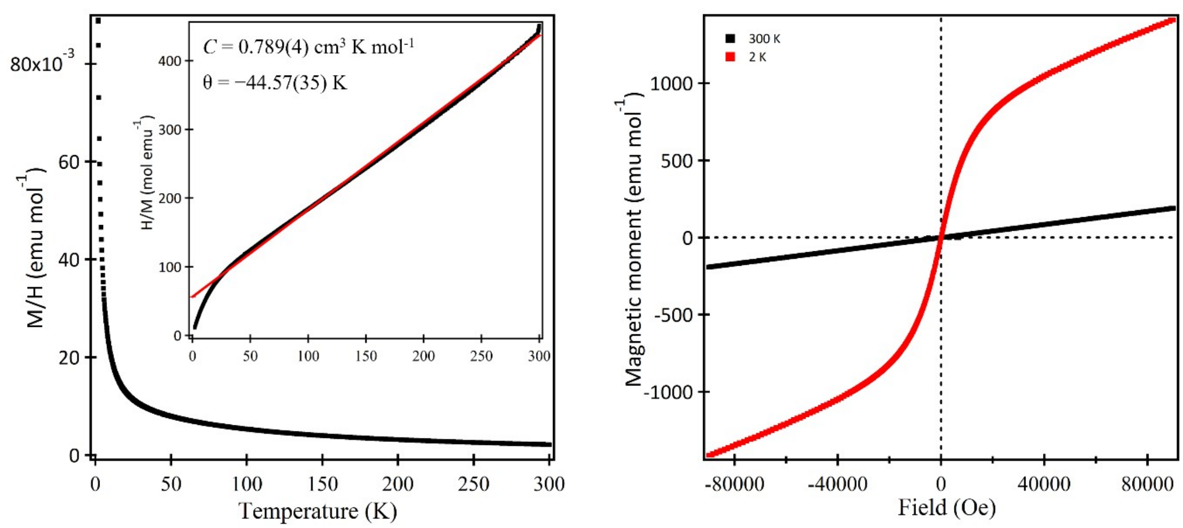


Figure S9. The magnetization data collected from $Sr_2Ga_{0.8}Fe_{0.2}SbO_6$.

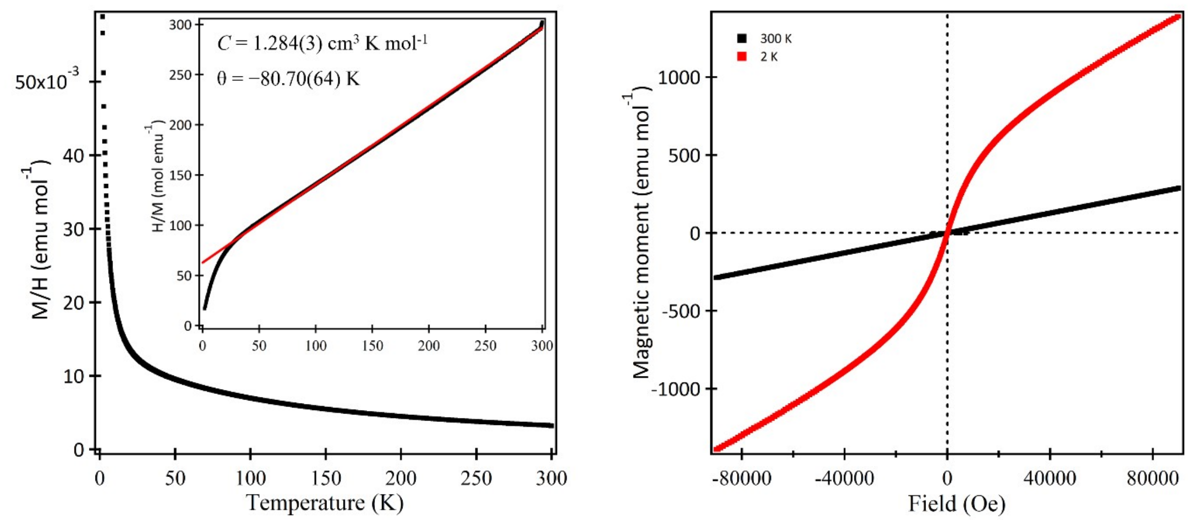


Figure S10. The magnetization data collected from $Sr_2Ga_{0.7}Fe_{0.3}SbO_6$.

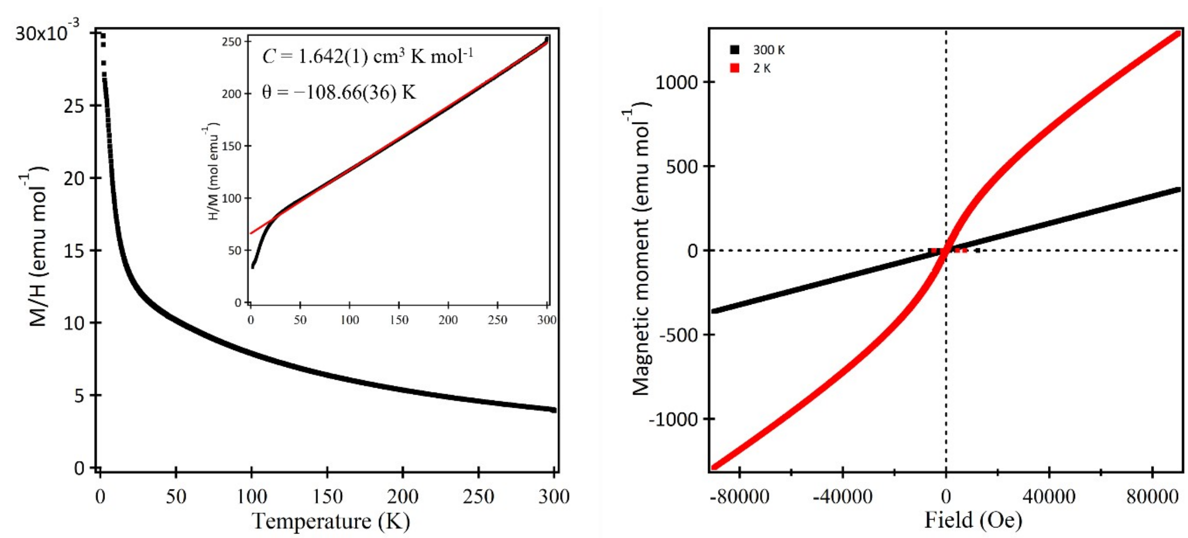


Figure S11. The magnetization data collected from $Sr_2Ga_{0.6}Fe_{0.4}SbO_6$.

4. The calculated band gaps for all the double perovskites prepared in this study

Table S4. The calculated band gaps from Tauc plot by using both indirect and direct transition equation for all the double perovskites prepared in this study.

Composition	Direct band gap (eV)	Indirect band gap (eV)
$Sr_2Ga_{1-x}M_xSbO_6$		
undoped	3.83	3.52
M = Cr^{3+}		
$x = 0.1$	3.80	3.16
$x = 0.2$	3.76	2.88
$x = 0.3$	3.33	2.55
$x = 0.4$	3.47	2.86
M = Fe^{3+}		
$x = 0.1$	3.51	3.24
$x = 0.2$	3.43	3.13
$x = 0.3$	3.33	2.97
$x = 0.4$	3.27	2.85