Supporting Information

First principles approach and Experimental Exploration of a New Double Perovskite Phase $Sr_2(In_{0.33}Sn_{0.33}Sb_{0.33})_2O_6$: Evaluation of Structural, Optical, and Dielectric Properties

Besma BELGACEM [a], Nabil NASRI[a], Mouna BEN YAHIA [b], Abderrazek OUESLATI [c], Rached BEN HASSEN [a]

[a] Laboratory of Materials and Environment for Sustainable Development LR18ES10, ISSBAT, University of Tunis El Manar, Tunis, Tunisia
[b] Institute Charles Gerhardt of Montpellier (ICGM), Univ. Montpellier, CNRS, ENSCM, Montpellier, France
[c] Laboratory of Spectroscopic Characterization and Optical Materials, Faculty of Sciences, University of Sfax, B.P. 1171, Sfax 3000, Tunisia

Table S1: Lattice parameters of the different calculated models compared to those of experimental data (The relative energetic stabilities of these various models are presented).

				cell parameters						
Models		**Stacking (Wyckoff site)**	**Energy (meV/f.u)**	**a**	**Δa/a %**	**b**	**Δb/b %**	**c**	**Δc/c %**	**□**
	Exp. 1			**5.4502**		**5.7206**		**9.7006**		**123.53**
O6 ($P2_1/c$)	**Model 1**	**Sn/In (2d) Sn/Sb (2a)**	**0.00**	**5.7900**	**6.2**	**5.8235**	**1.8**	**10.0500**	**3.6**	**125.3**
	Model 2	**Sn/In (2d) Sn/Sb (2a)**	**4**	**5.7835**	**6.1**	**5.8263**	**1.9**	**10.0443**	**3.5**	**125.2**
	Model3	**Sn/In (2d) Sn/Sb (2a)**	**0.3**	**5.7838**	**6.1**	**5.8262**	**1.8**	**10.0446**	**3.5**	**125.2**
	Exp. 2			**17.1850**		**5.6955**		**9.9037**		**125.2**
O9 ($P2_1/c$)	**Model 4**	**In(2a) In(2d) Sn/Sb(4e) Sn/Sb(4e)**	**417**	**17.3758**	**1.1**	**5.7719**	**1.3**	**8.1321**	**17.9**	**125.5**
	Model 5	**In(2a) In(2d) Sn/Sb(4e) Sn/Sb(4e)**	**199**	**17.2984**	**0.7**	**5.7796**	**1.5**	**8.1318**	**17.9**	**125.5**
	Model 6	**In/Sn(4e) Sb/Sn(4e) In(2a) Sb(2d)**	**95**	**17.2691**	**0.5**	**5.7833**	**1,5**	**9.9396**	**0.4**	**125.4**
	Model 7	**In/Sn(4e) Sb/Sn(4e) Sb(2a) In(2d)**	**186**	**17.2984**	**0.7**	**5.7796**	**1,5**	**8.1318**	**17.9**	**125.3**
	Model 8	**In(4e) Sb(4e) Sn(2a) Sn(2d)**	**94**	**17.2815**	**0.6**	**5.7762**	**1,4**	**9.9534**	**0.5**	**125.4**

To reproduce the disorder, models 1 to 3 correspond to supercells created by tripling the experimental O6 unit cell (Exp.1), with space group $P2_1/c$, along the *a* direction. The atoms were then placed on the Wyckoff sites indicated in Table S1. Each model features a different

distribution of Sn and In atoms on the 2d site, and Sn and Sb atoms on the 2a site, while respecting the experimental occupancy.
Models 4 to 8 are based on a second experimental model, also monoclinic (space group $P2_1/c$), corresponding to the O9 structure with a lattice parameter three times larger than that of the O6 structure, and presenting full or partial occupancy of the 2a, 2d, and 4e sites by the metalloid atoms. For example, model 5 shows full occupancy of the 2a and 2d sites by In, while Sn and Sb share the two remaining 4e sites, which corresponds to the experimental proposal. Model 8, fully ordered, corresponds to the placement of In on one 4e site, Sb on the other 4e site, and Sn on the 2a and 2d sites. The construction scheme and the tested models are shown below.

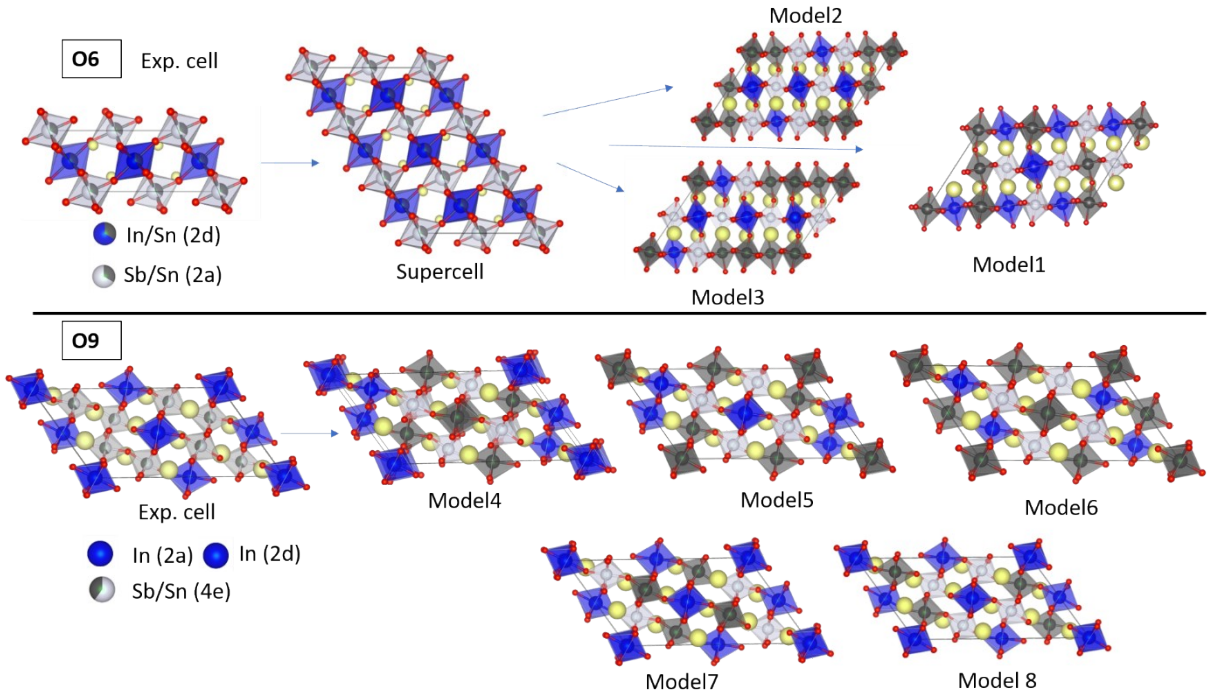

Fig. S1 The construction scheme of the supercells, as well as the tested models.

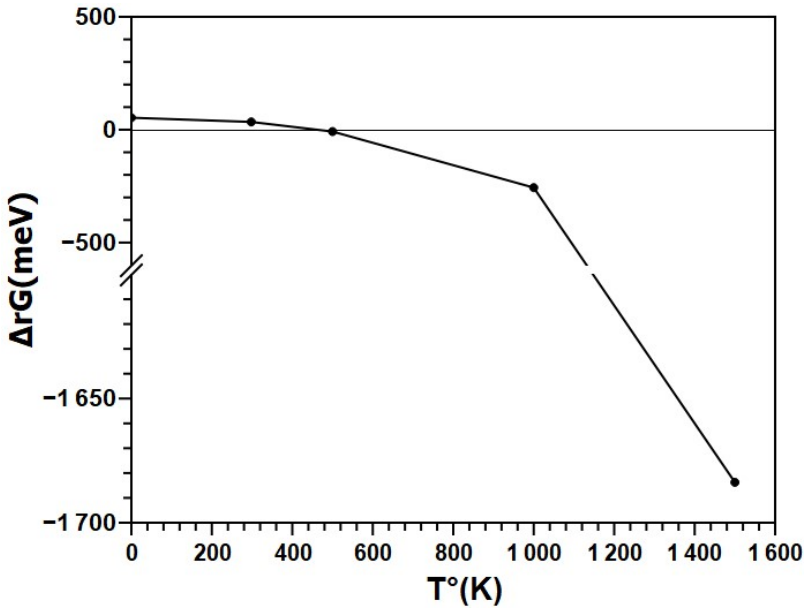

Fig. S2 Free energy in meV as a function of temperature in K° associated with the formation of the new phase according to the reaction:

$$SrSnO_3 + Sr_2SbInO_6 \rightarrow 3/2\ Sr_2(Sn_{0.33}Sb_{0.33}In_{0.33})_2O_6$$

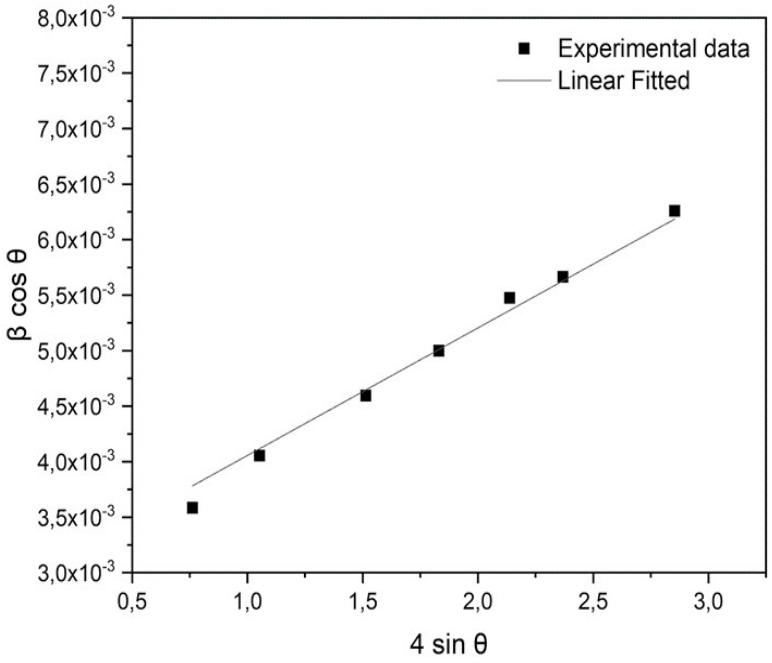

Fig. S3 Williamson-Hall (W-H) plot of $Sr_2(Sn_{0.33}Sb_{0.33}In_{0.33})_2O_6$.

Table S2: EDX analysis showing the atomic percent of all evaluated elements of the $Sr_2(Sn_{0.33}Sb_{0.33}In_{0.33})_2O_6$ ceramic

	Sr	Sn	In	Sb	O	C
Nominal composition	20	6.666	6.666	6.666	60	
Experimental composition	16.1	8.1	8.3	8.3	54.8	4.5
Absolute error (%)	0.3	1.2	0.8	1.0	1.5	0.6

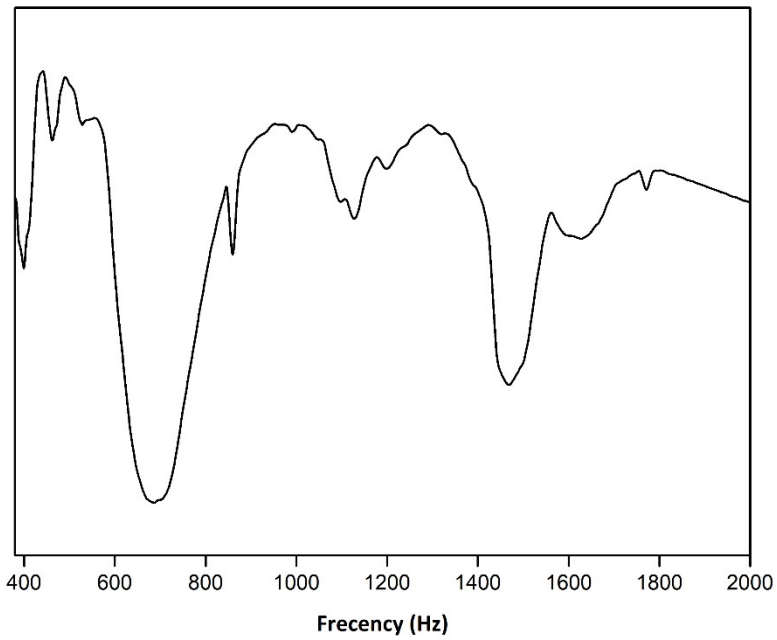

Fig. S4 Experimental IR spectrum of the $Sr_2(Sn_{0.33}Sb_{0.33}In_{0.33})_2O_6$ within the

Range of 400 to 2000 cm^{-1}.

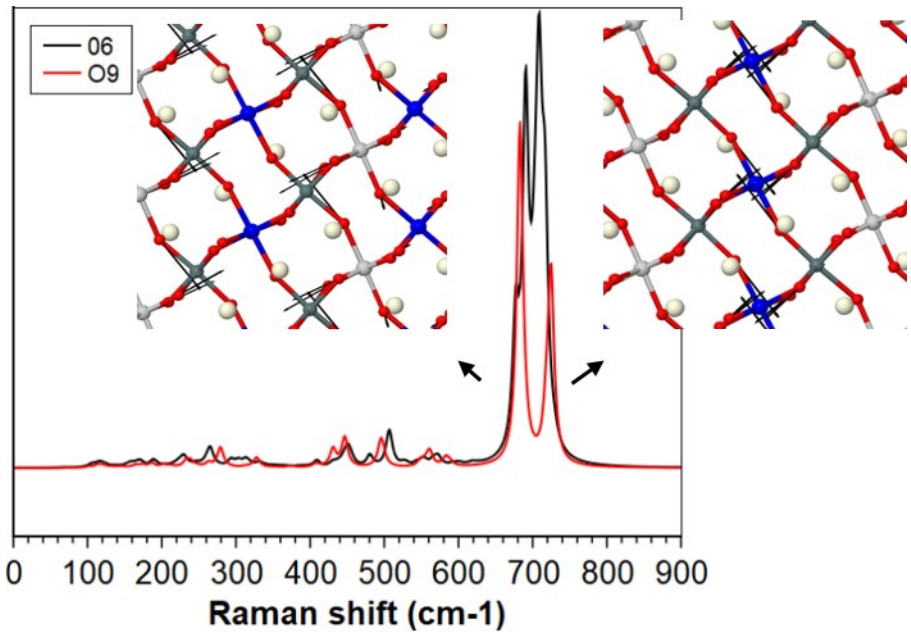

Fig. S5: Calculated Raman spectrum of the most stable O9 model (model 8) compared to the O6 model (model 1). Schematics of the vibrational modes associated with the most intense stretches are shown in (a) and (b). The region above 700 cm^{-1} is dominated by In–O and Sn–O bond stretching, while the region between 650–700 cm^{-1} primarily corresponds to the stretching of oxygen atoms bonded to Sb and Sn.

Table S3 Fitting parameters of the complex impedance plots at various temperatures for $Sr_2(Sn_{0.33}Sb_{0.33}In_{0.33})_2O_6$ ceramic

T(K)	R(Ω)	C(F)	Q(F)	α
573	$2.0733E^{7}$	$4.038E^{-11}$	$5.000E^{-10}$	0.63837
593	$1.0958E^{7}$	$4.029E^{-11}$	$5.990E^{-10}$	0.64186
613	$6.0656E^{6}$	$4.022E^{-11}$	$7.010E^{-10}$	0.64558
633	$3.545E^{6}$	$4.038E^{-11}$	$9.178E^{-10}$	0.63694
653	$2.2286E^{6}$	$4.030E^{-11}$	$1.160E^{-9}$	0.63232
673	$1.2354E^{6}$	$4.509E^{-11}$	$5.460E^{-9}$	0.502