Transparency Enhancement for $SrVO_3$ by $SrTiO_3$ Mixing: A First-Principles Study

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Supplemental Materials

A. Magnetism and potential choices for V

We justify the choice of U = 4 eV for V. Figure S1 shows the energy-dependent complex dielectric function spectra ε_1 and ε_2 for $SrVO_3$, and Figure S2 shows the band structures calculated with different parameter sets. In Figure S1, there are curves from six variations, ferromagnetic (FM) or non-magnetic (NM) with PBE potentials and U = 0 or 4 eV, or using the Heyd-Scuseria-Ernzerhof hybrid functional (HSE06). In agreement with previous calculations[1], FM/U = 0 matches the experimental measurements well. Comparing FM/U = 0 and NM/U = 0, the lack of consideration of magnetization has a small influence on the complex dielectric functions, consistent with previous calculations[1]. It red-shifts (lower) the rise of ε_2 at ~2.8 eV by 0.2 eV. On the other hand, within FM, the introduction of the GGA+U scheme with U = 4 eV red-shifts the rise of ε_2 greatly, by 0.8 eV, while within NM, it merely blue-shifts (higher) the rise by 0.2 eV, overlapping with the FM/U = 0 curve. Generally, GGA+U widens the band gap for semiconductors and insulators, and blue-shifts the overall complex dielectric function curves, as in the NM case. Further calculations reveal that for FM, U = 0 results in the magnetic moment of V^{4+} being 0.5 μ_B, and a value of U beyond 2 eV increases the effective on-site Coulomb repulsion and raises the magnetic moment to 1 μ_B. At U = 4 eV, there is a larger separation between the spin-up and spin-down channels of the V-t_{2g} bands around the Fermi energy level (E_F) as shown in Figure S2(a) and (b). Each channel then gets closer to the adjacent bands, i.e. the lower-lying O-p and higher-lying V-e_g bands, facilitating interband transitions. GGA+U leads to little change of the NM band structure, because there is no such spin channel separation occurring in the FM case. Only NM/U = 4 eV is shown in Figure S2(c), similar to FM/U = 0 in Figure S2(a). The FM and NM HSE06 curves in Figure S1 deviate greatly from the experimental observations. In short, considering the correlated nature of $SrVO_3$, small magnetic moments, and agreement of complex dielectric functions with the experimental measurements, we used NM/U = 4 eV for all calculations in this work.

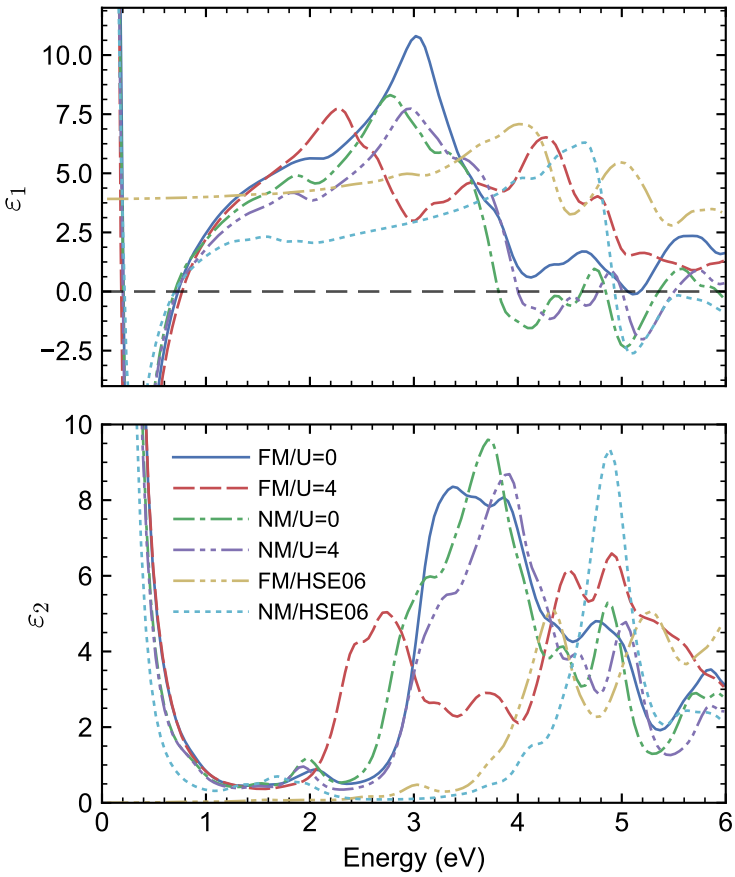

Figure S1. Calculated complex dielectric function spectra ε_1 and ε_2 of $SrVO_3$ with different parameter settings. FM stands for the ferromagnetic configuration for V^{4+} and NM for non-magnetic. The first four cases were calculated with PBE potentials, and last two with HSE06.

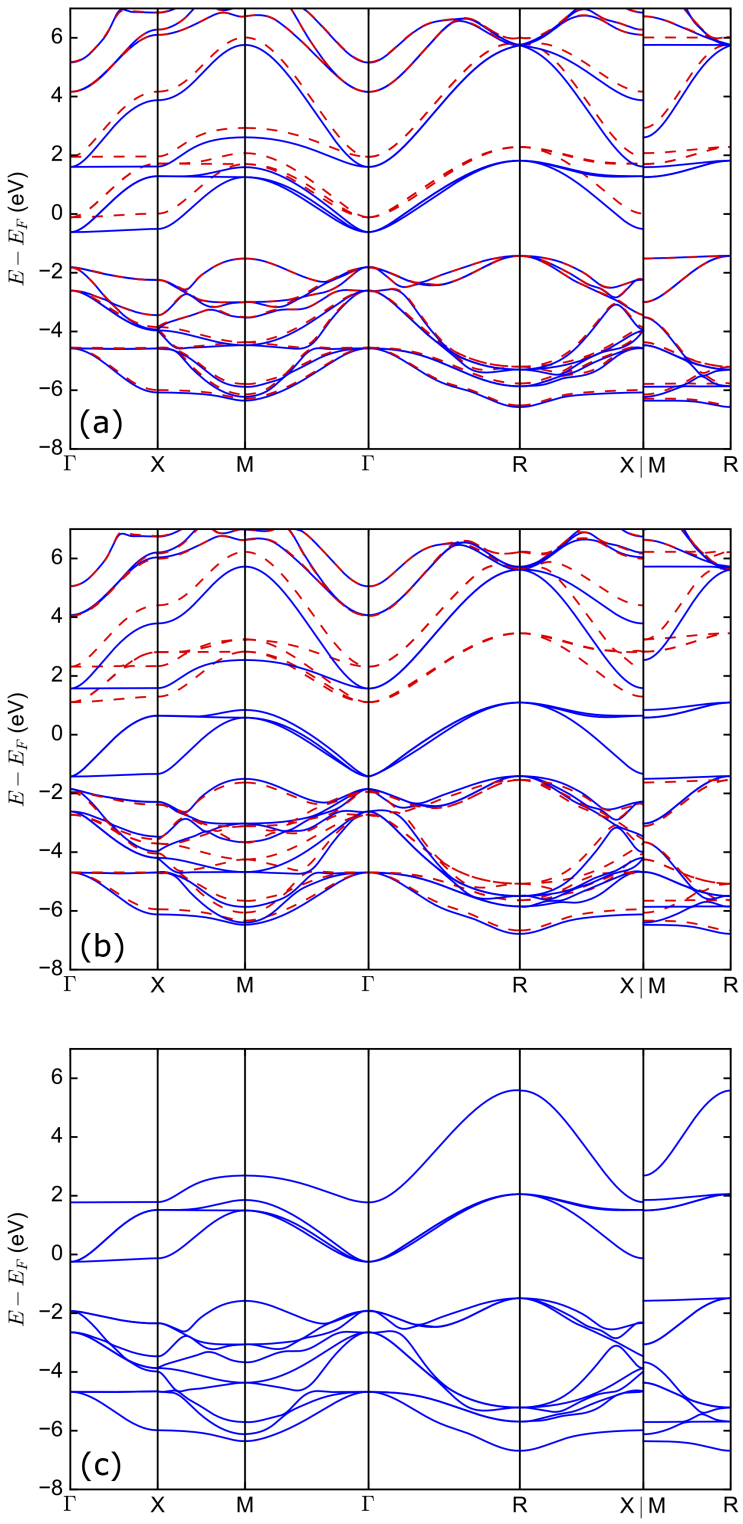

Figure S2. Calculated band structures of $SrVO_3$ with (a) FM/U = 0, (b) FM/U = 4 eV, (c) NM/U = 4 eV, all with PBE potentials. FM stands for the ferromagnetic configuration for V^{4+} and NM for non-magnetic. In cases of FM, the solid blue and dashed red curves stand for the spin-up and spin-down channels, respectively.

B. Cluster expansion

Below we provide detailed information of the cluster set used in the fitting process.

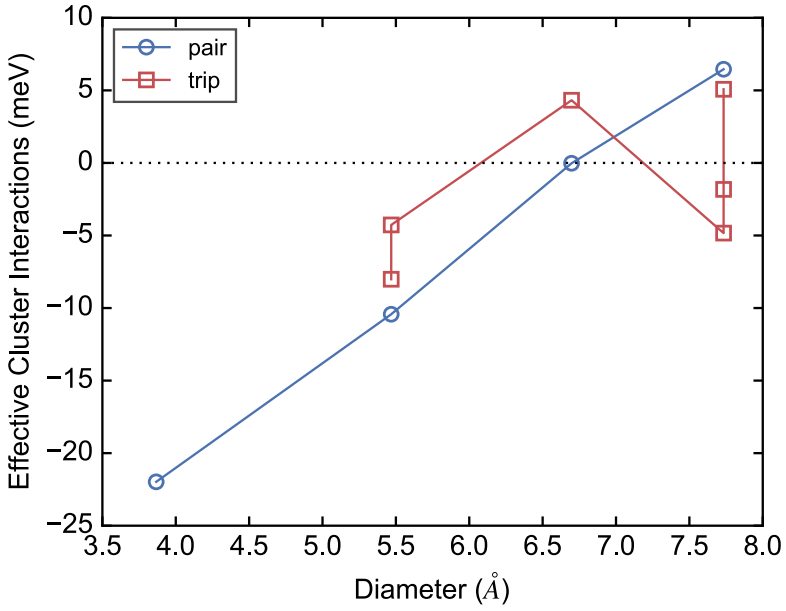

Figure S3. Effective cluster interactions (ECIs) of $SrTi_{1-x}V_xO_3$. ECI values have been multiplied by their multiplicities. Blue circles and red squares triangles indicate pair and triplet interactions. Connecting lines are a guide to the eye.

Table S1. Cluster sets and effective cluster interactions (ECIs) of $SrTi_{1-x}V_xO_3$.

Type	Index	Diameter (Å)	Multiplicity	Coordinates	ECI (meV)
empty	1	0.00	1		25.777
point	1	0.00	1	[[1.0, 1.0, 1.0]]	8.794
pair	1	3.87	3	[[1.0, 1.0, 1.0], [1.0, 0.0, 1.0]]	-7.326
pair	2	5.47	6	[[1.0, 1.0, 1.0], [1.0, 0.0, 0.0]]	-1.738
pair	3	6.70	4	[[1.0, 1.0, 1.0], [0.0, 0.0, 0.0]]	-0.006
pair	4	7.73	3	[[1.0, 1.0, 1.0], [1.0, 1.0, -1.0]]	2.152
trip	1	5.47	12	[[1.0, 1.0, 1.0], [1.0, 1.0, 0.0], [1.0, 0.0, 0.0]]	0.668

trip	2	5.47	8	[[1.0, 1.0, 1.0], [0.0, 1.0, 0.0], [1.0, 0.0, 0.0]]	0.534
trip	3	6.70	24	[[1.0, 1.0, 1.0], [1.0, 1.0, 0.0], [0.0, 0.0, 0.0]]	-0.180
trip	4	7.73	3	[[1.0, 1.0, 1.0], [1.0, 1.0, 0.0], [1.0, 1.0, -1.0]]	1.612
trip	5	7.73	12	[[1.0, 1.0, 1.0], [1.0, 0.0, 0.0], [1.0, 1.0, -1.0]]	0.152
trip	6	7.73	12	[[1.0, 1.0, 1.0], [0.0, 0.0, 0.0], [1.0, 1.0, -1.0]]	-0.423

Volumes of random solid solutions deviate from linearity slightly with down-bowing behavior.

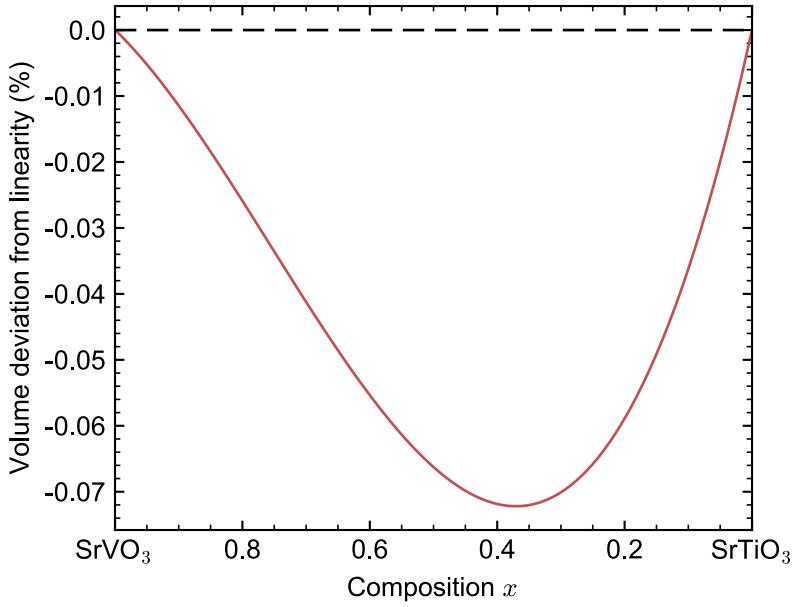

Figure S4. Volume deviations from linearity of $SrTi_{1-x}V_xO_3$. Curves indicate CE-fitted values of the random solid solution configurations.

C. Special quasi-random structure (SQS) prototypes

Here we provide the ionic configurations of the SQS's of $SrTi_{1-x}V_xO_3$ at x = 8/9, 7/9, …, 1/9. Each SQS is a cubic supercell with a lattice constant of 11.6 Å of 3 × 3 × 3 5-atom unit cells. The 27 exchangeable sites are listed for Ti and V. Coordinates of the other ions are provided at the end.

Table S2. Ionic configurations of the SQS's of $SrTi_{1-x}V_xO_3$ at x = 8/9, 7/9, …, 1/9.

x = 8/9:

	Fractional Coordinates				Fractional Coordinates		
Ion	**u_1**	**u_2**	**u_3**	**Ion**	**u_1**	**u_2**	**u_3**
Ti	0.333	0.333	1.000				
Ti	1.000	0.333	0.333				
Ti	0.667	0.333	0.333				
V	1.000	1.000	1.000	V	1.000	0.667	0.333
V	0.333	1.000	1.000	V	0.333	0.667	0.333
V	0.667	1.000	1.000	V	0.667	0.667	0.333
V	1.000	0.333	1.000	V	1.000	1.000	0.667
V	0.667	0.333	1.000	V	0.333	1.000	0.667
V	1.000	0.667	1.000	V	0.667	1.000	0.667
V	0.333	0.667	1.000	V	1.000	0.333	0.667
V	0.667	0.667	1.000	V	0.333	0.333	0.667
V	1.000	1.000	0.333	V	0.667	0.333	0.667
V	0.333	1.000	0.333	V	1.000	0.667	0.667
V	0.667	1.000	0.333	V	0.333	0.667	0.667
V	0.333	0.333	0.333	V	0.667	0.667	0.667

x = 7/9:

	Fractional Coordinates				Fractional Coordinates		
Ion	**u_1**	**u_2**	**u_3**	**Ion**	**u_1**	**u_2**	**u_3**
Ti	0.333	1.000	1.000				
Ti	0.667	0.333	1.000				
Ti	0.333	0.667	1.000				
Ti	0.667	0.667	1.000	V	0.667	0.333	0.333
Ti	0.333	0.333	0.333	V	1.000	0.667	0.333
Ti	0.333	1.000	0.667	V	0.333	0.667	0.333
V	1.000	1.000	1.000	V	0.667	0.667	0.333
V	0.667	1.000	1.000	V	1.000	1.000	0.667
V	1.000	0.333	1.000	V	0.667	1.000	0.667
V	0.333	0.333	1.000	V	1.000	0.333	0.667
V	1.000	0.667	1.000	V	0.333	0.333	0.667
V	1.000	1.000	0.333	V	0.667	0.333	0.667
V	0.333	1.000	0.333	V	1.000	0.667	0.667
V	0.667	1.000	0.333	V	0.333	0.667	0.667
V	1.000	0.333	0.333	V	0.667	0.667	0.667

x = 6/9:

	Fractional Coordinates				Fractional Coordinates		
Ion	**u_1**	**u_2**	**u_3**	**Ion**	**u_1**	**u_2**	**u_3**
Ti	0.333	1.000	1.000				
Ti	1.000	0.333	1.000				
Ti	1.000	0.667	1.000				
Ti	0.667	0.667	1.000	V	0.333	0.333	0.333
Ti	0.333	1.000	0.333	V	0.667	0.333	0.333
Ti	0.667	1.000	0.333	V	0.667	0.667	0.333
Ti	1.000	0.333	0.333	V	1.000	1.000	0.667
Ti	1.000	0.667	0.333	V	0.333	1.000	0.667
Ti	0.333	0.667	0.333	V	0.667	1.000	0.667
V	1.000	1.000	1.000	V	1.000	0.333	0.667
V	0.667	1.000	1.000	V	0.333	0.333	0.667
V	0.333	0.333	1.000	V	0.667	0.333	0.667
V	0.667	0.333	1.000	V	1.000	0.667	0.667
V	0.333	0.667	1.000	V	0.333	0.667	0.667
V	1.000	1.000	0.333	V	0.667	0.667	0.667

$x = 5/9$:

	Fractional Coordinates				Fractional Coordinates		
Ion	**u_1**	**u_2**	**u_3**	**Ion**	**u_1**	**u_2**	**u_3**
Ti	1.000	1.000	1.000				
Ti	0.333	1.000	1.000				
Ti	0.667	1.000	1.000				
Ti	1.000	0.333	1.000	V	0.333	1.000	0.333
Ti	0.333	0.333	1.000	V	1.000	0.333	0.333
Ti	1.000	0.667	1.000	V	0.333	0.333	0.333
Ti	0.667	0.667	1.000	V	0.667	0.333	0.333
Ti	0.667	1.000	0.333	V	1.000	0.667	0.333
Ti	0.333	0.667	0.333	V	0.333	1.000	0.667
Ti	0.667	0.667	0.333	V	0.667	1.000	0.667
Ti	1.000	1.000	0.667	V	1.000	0.333	0.667
Ti	0.333	0.667	0.667	V	0.333	0.333	0.667
V	0.667	0.333	1.000	V	0.667	0.333	0.667
V	0.333	0.667	1.000	V	1.000	0.667	0.667
V	1.000	1.000	0.333	V	0.667	0.667	0.667

$x = 4/9$:

	Fractional Coordinates				Fractional Coordinates		
Ion	**u_1**	**u_2**	**u_3**	**Ion**	**u_1**	**u_2**	**u_3**

Ti	1.000	1.000	1.000				
Ti	0.333	1.000	1.000				
Ti	1.000	0.333	1.000				
Ti	0.667	0.333	1.000	V	0.667	1.000	1.000
Ti	0.667	0.667	1.000	V	0.333	0.333	1.000
Ti	1.000	0.333	0.333	V	1.000	0.667	1.000
Ti	0.333	0.667	0.333	V	0.333	0.667	1.000
Ti	0.333	1.000	0.667	V	1.000	1.000	0.333
Ti	0.667	1.000	0.667	V	0.333	1.000	0.333
Ti	1.000	0.333	0.667	V	0.667	1.000	0.333
Ti	0.333	0.333	0.667	V	0.333	0.333	0.333
Ti	0.667	0.333	0.667	V	0.667	0.333	0.333
Ti	1.000	0.667	0.667	V	1.000	0.667	0.333
Ti	0.333	0.667	0.667	V	0.667	0.667	0.333
Ti	0.667	0.667	0.667	V	1.000	1.000	0.667

x = 3/9:

	Fractional Coordinates				**Fractional Coordinates**		
Ion	**u_1**	**u_2**	**u_3**	**Ion**	**u_1**	**u_2**	**u_3**
Ti	1.000	1.000	1.000				
Ti	0.333	0.333	1.000				
Ti	1.000	0.667	1.000				
Ti	0.667	0.667	1.000	Ti	1.000	0.667	0.667
Ti	0.333	1.000	0.333	Ti	0.333	0.667	0.667
Ti	1.000	0.333	0.333	Ti	0.667	0.667	0.667
Ti	0.333	0.333	0.333	V	0.333	1.000	1.000
Ti	1.000	0.667	0.333	V	0.667	1.000	1.000
Ti	0.333	0.667	0.333	V	1.000	0.333	1.000
Ti	0.667	0.667	0.333	V	0.667	0.333	1.000
Ti	0.333	1.000	0.667	V	0.333	0.667	1.000
Ti	0.667	1.000	0.667	V	1.000	1.000	0.333
Ti	1.000	0.333	0.667	V	0.667	1.000	0.333
Ti	0.333	0.333	0.667	V	0.667	0.333	0.333
Ti	0.667	0.333	0.667	V	1.000	1.000	0.667

x = 2/9:

	Fractional Coordinates				**Fractional Coordinates**		
Ion	**u_1**	**u_2**	**u_3**	**Ion**	**u_1**	**u_2**	**u_3**
Ti	1.000	1.000	1.000				
Ti	0.333	1.000	1.000				

Ti	0.667	1.000	1.000				
Ti	1.000	0.333	1.000	Ti	1.000	0.333	0.667
Ti	0.333	0.333	1.000	Ti	0.333	0.333	0.667
Ti	1.000	0.667	1.000	Ti	0.667	0.333	0.667
Ti	0.333	0.667	1.000	Ti	1.000	0.667	0.667
Ti	0.667	0.667	1.000	Ti	0.333	0.667	0.667
Ti	0.667	1.000	0.333	Ti	0.667	0.667	0.667
Ti	0.333	0.333	0.333	V	0.667	0.333	1.000
Ti	1.000	0.667	0.333	V	1.000	1.000	0.333
Ti	0.333	0.667	0.333	V	0.333	1.000	0.333
Ti	0.667	0.667	0.333	V	1.000	0.333	0.333
Ti	1.000	1.000	0.667	V	0.667	0.333	0.333
Ti	0.333	1.000	0.667	V	0.667	1.000	0.667

$x = 1/9$:

	Fractional Coordinates				**Fractional Coordinates**		
Ion	$\mathbf{u_1}$	$\mathbf{u_2}$	$\mathbf{u_3}$	**Ion**	$\mathbf{u_1}$	$\mathbf{u_2}$	$\mathbf{u_3}$
Ti	1.000	1.000	1.000				
Ti	0.333	1.000	1.000				
Ti	1.000	0.333	1.000				
Ti	0.333	0.333	1.000	Ti	0.667	0.667	0.333
Ti	0.667	0.333	1.000	Ti	1.000	1.000	0.667
Ti	1.000	0.667	1.000	Ti	0.333	1.000	0.667
Ti	0.333	0.667	1.000	Ti	1.000	0.333	0.667
Ti	0.667	0.667	1.000	Ti	0.333	0.333	0.667
Ti	0.333	1.000	0.333	Ti	0.667	0.333	0.667
Ti	0.667	1.000	0.333	Ti	1.000	0.667	0.667
Ti	1.000	0.333	0.333	Ti	0.333	0.667	0.667
Ti	0.333	0.333	0.333	Ti	0.667	0.667	0.667
Ti	0.667	0.333	0.333	V	0.667	1.000	1.000
Ti	1.000	0.667	0.333	V	1.000	1.000	0.333
Ti	0.333	0.667	0.333	V	0.667	1.000	0.667

Other ions:

	Fractional Coordinates				**Fractional Coordinates**		
Ion	$\mathbf{u_1}$	$\mathbf{u_2}$	$\mathbf{u_3}$	**Ion**	$\mathbf{u_1}$	$\mathbf{u_2}$	$\mathbf{u_3}$
Sr	0.167	0.167	0.167	O	1.000	1.000	0.167
Sr	0.500	0.167	0.167	O	0.333	1.000	0.167
Sr	0.833	0.167	0.167	O	0.667	1.000	0.167
Sr	0.167	0.500	0.167	O	1.000	0.333	0.167

Sr	0.500	0.500	0.167	O	0.333	0.333	0.167
Sr	0.833	0.500	0.167	O	0.667	0.333	0.167
Sr	0.167	0.833	0.167	O	1.000	0.667	0.167
Sr	0.500	0.833	0.167	O	0.333	0.667	0.167
Sr	0.833	0.833	0.167	O	0.667	0.667	0.167
Sr	0.167	0.167	0.500	O	1.000	1.000	0.500
Sr	0.500	0.167	0.500	O	0.333	1.000	0.500
Sr	0.833	0.167	0.500	O	0.667	1.000	0.500
Sr	0.167	0.500	0.500	O	1.000	0.333	0.500
Sr	0.500	0.500	0.500	O	0.333	0.333	0.500
Sr	0.833	0.500	0.500	O	0.667	0.333	0.500
Sr	0.167	0.833	0.500	O	1.000	0.667	0.500
Sr	0.500	0.833	0.500	O	0.333	0.667	0.500
Sr	0.833	0.833	0.500	O	0.667	0.667	0.500
Sr	0.167	0.167	0.833	O	1.000	1.000	0.833
Sr	0.500	0.167	0.833	O	0.333	1.000	0.833
Sr	0.833	0.167	0.833	O	0.667	1.000	0.833
Sr	0.167	0.500	0.833	O	1.000	0.333	0.833
Sr	0.500	0.500	0.833	O	0.333	0.333	0.833
Sr	0.833	0.500	0.833	O	0.667	0.333	0.833
Sr	0.167	0.833	0.833	O	1.000	0.667	0.833
Sr	0.500	0.833	0.833	O	0.333	0.667	0.833
Sr	0.833	0.833	0.833	O	0.667	0.667	0.833
O	1.000	0.167	1.000	O	0.167	1.000	1.000
O	0.333	0.167	1.000	O	0.500	1.000	1.000
O	0.667	0.167	1.000	O	0.833	1.000	1.000
O	1.000	0.500	1.000	O	0.167	0.333	1.000
O	0.333	0.500	1.000	O	0.500	0.333	1.000
O	0.667	0.500	1.000	O	0.833	0.333	1.000
O	1.000	0.833	1.000	O	0.167	0.667	1.000
O	0.333	0.833	1.000	O	0.500	0.667	1.000
O	0.667	0.833	1.000	O	0.833	0.667	1.000
O	1.000	0.167	0.333	O	0.167	1.000	0.333
O	0.333	0.167	0.333	O	0.500	1.000	0.333
O	0.667	0.167	0.333	O	0.833	1.000	0.333
O	1.000	0.500	0.333	O	0.167	0.333	0.333
O	0.333	0.500	0.333	O	0.500	0.333	0.333
O	0.667	0.500	0.333	O	0.833	0.333	0.333
O	1.000	0.833	0.333	O	0.167	0.667	0.333
O	0.333	0.833	0.333	O	0.500	0.667	0.333

O	0.667	0.833	0.333	O	0.833	0.667	0.333
O	1.000	0.167	0.667	O	0.167	1.000	0.667
O	0.333	0.167	0.667	O	0.500	1.000	0.667
O	0.667	0.167	0.667	O	0.833	1.000	0.667
O	1.000	0.500	0.667	O	0.167	0.333	0.667
O	0.333	0.500	0.667	O	0.500	0.333	0.667
O	0.667	0.500	0.667	O	0.833	0.333	0.667
O	1.000	0.833	0.667	O	0.167	0.667	0.667
O	0.333	0.833	0.667	O	0.500	0.667	0.667
O	0.667	0.833	0.667	O	0.833	0.667	0.667

Here we show the bond lengths of Ti-O and bond angles O-Ti-O in SQS's of $SrTi_{1-x}V_xO_3$ with different compositions x for completeness.

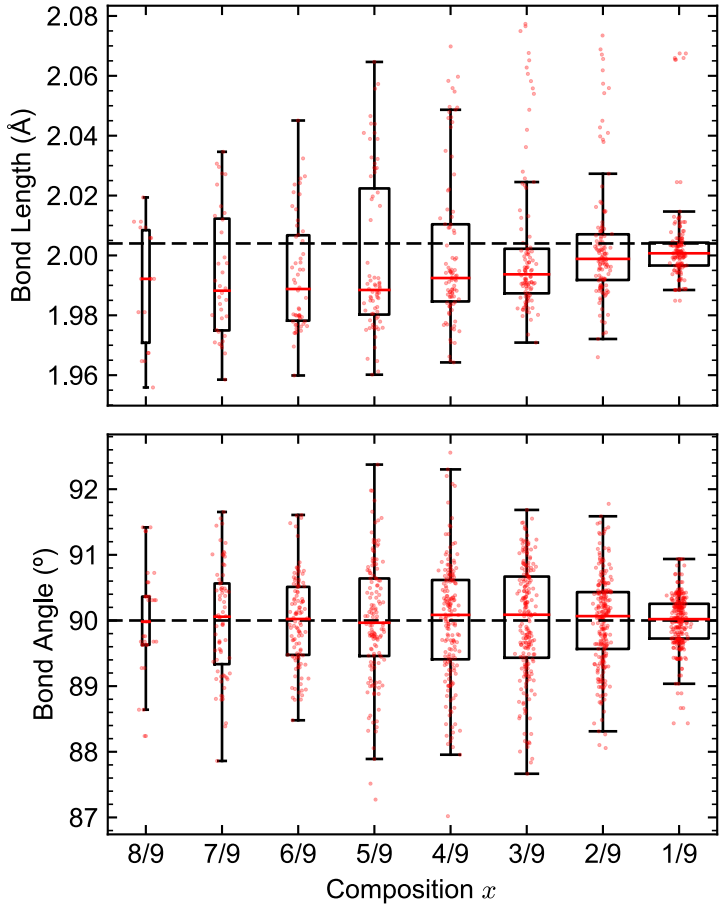

Figure S5. Bond lengths of Ti-O, and bond angles of O-Ti-O in SQS's of $SrTi_{1-x}V_xO_3$ with different compositions x. Each box extends from the lower to upper quartile values, with a line at the median. The whiskers extend from the box to show the range of the data. Widths of the box are scaled with respect to numbers of data points. The slightly horizontally jittered red dots are actual values. The dashed lines correspond to values in the end member $SrTiO_3$.