First-Principles Understanding of Hole Mobility and Intrinsic Transport Mechanisms in Sn(II) Oxides

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Supplementary Information

Materials	Direct band gap (eV)		Indirect band gap (eV)	
	PBE0	HSE06	PBE0	HSE06
SnO	3.29	2.64	1.24	0.81
c-$K_2Sn_2O_3$	2.53	2.15	2.50	2.10
r-$K_2Sn_2O_3$	2.36	1.75	-	-
r-$Rb_2Sn_2O_3$	2.37	1.76	-	-
$TiSnO_3$	3.37	2.68	2.94	2.26

Table 1: Direct and indirect band gaps computed with hybrid functionals (PBE0 and HSE06).

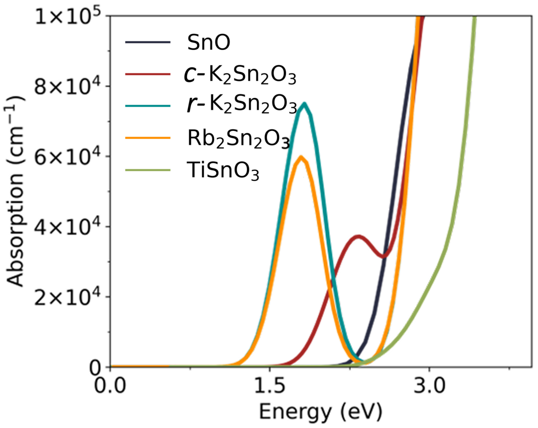

Fig. S1: Band–band optical absorption for the different Sn(II) oxides computed with HSE06.

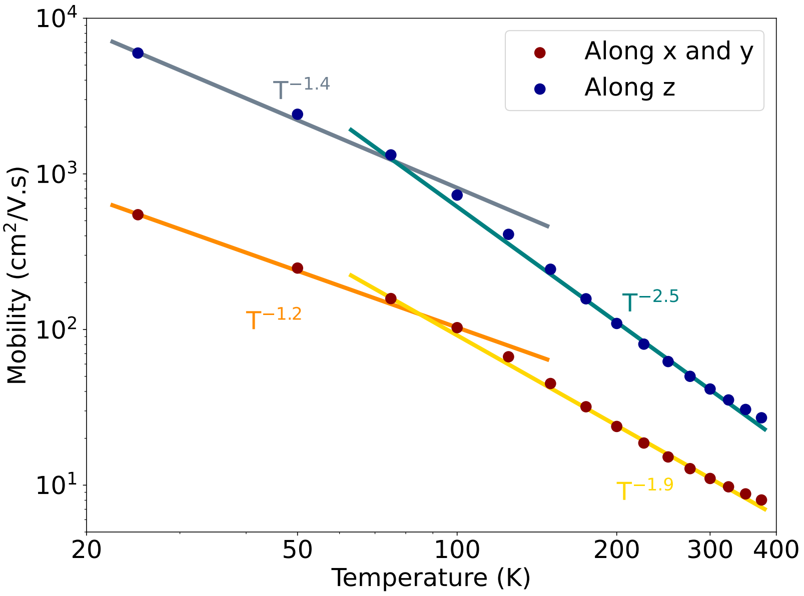

Fig. S2: IBTE hole drift mobility in SnO as a function of the temperature showing the change in the temperature trend around 100K.

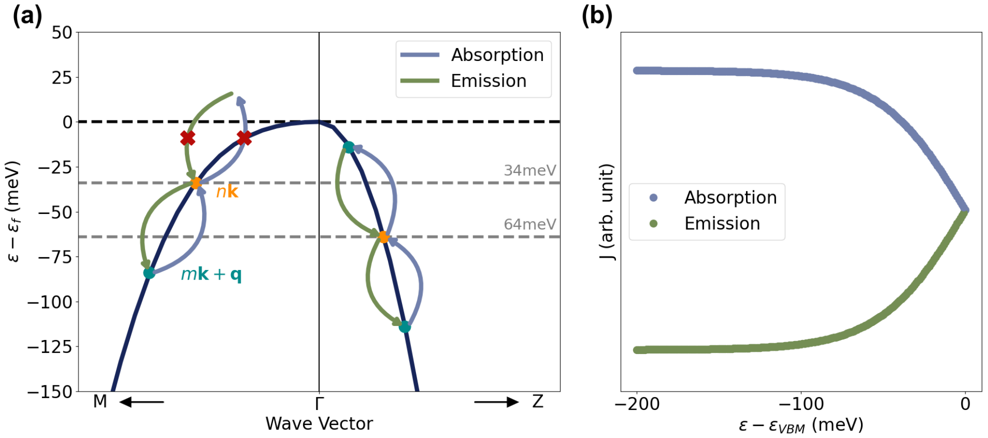

Fig. S3: **(a)** Schematic view of the different transitions possible from a initial state $n\mathbf{k}$ (in orange) of 34 and 64 meV to a final state $m\mathbf{k}+\mathbf{q}$ (in cyan) due to a phonon of 50 meV in SnO. The absorption and emission phenomena are in blue and green, respectively. The conservation of momentum is not taken into account in this schematic view. **(b)** Model of the behaviour of the parameter J as a function of the energy due to the absorption and emission of a phonon of a specific energy (following the Einstein model) in a single parabolic band p-type crystal.

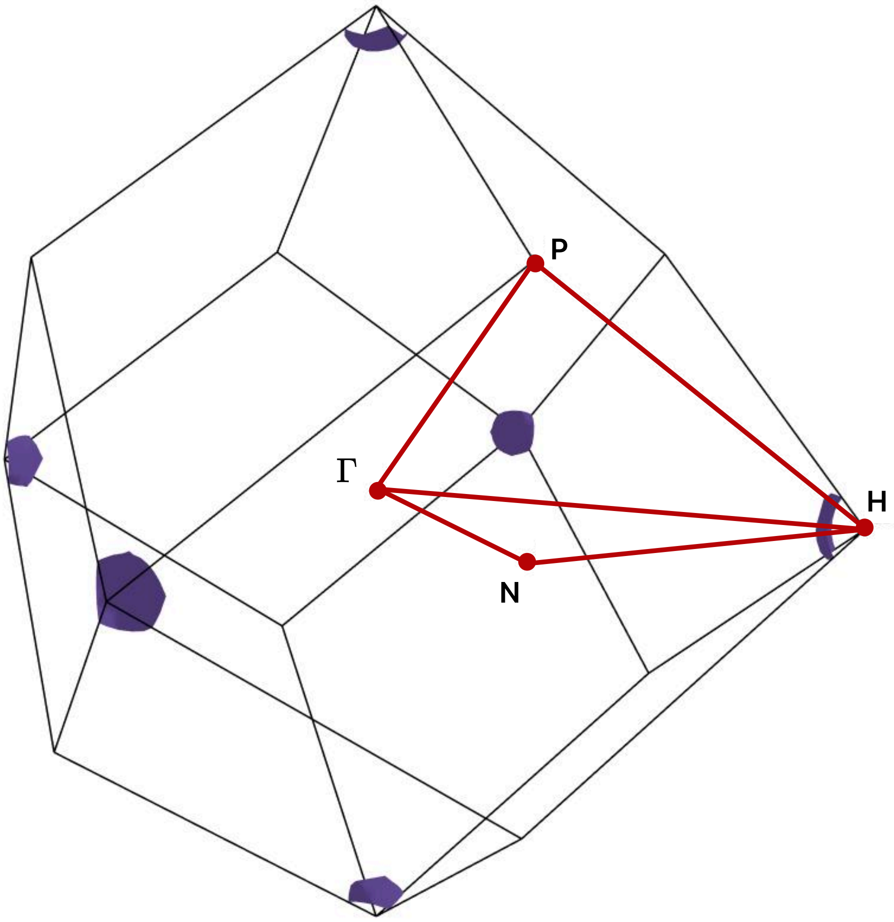

Fig. S4: Fermi surface of c-$K_2Sn_2O_3$ at an energy close to the VBM showing the different pockets that allow intervalley transitions.

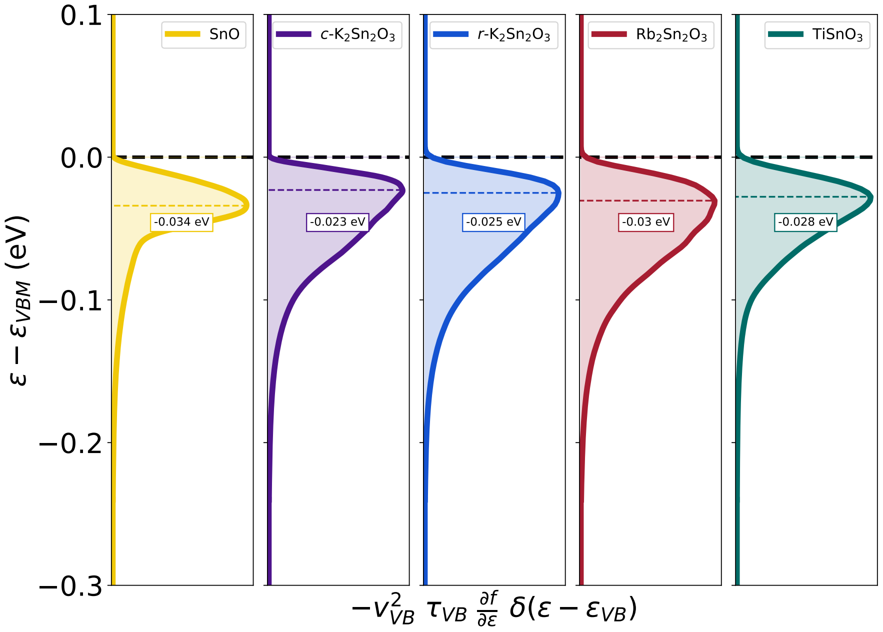

Fig. S5: The function $-v_{\mathrm{VB}}^2\ \tau_{\mathrm{VB}}\ \frac{\partial f}{\partial \varepsilon}$, convoluted with $\delta(\varepsilon - \varepsilon_{\mathrm{VB}})$ in the valence band at 300K for the different materials studied in this work. In this expression, v_{VB} and τ_{VB} stand for the carrier velocity and lifetime in the valence band (VB), $\frac{\partial f}{\partial \varepsilon}$ is the derivative of the Fermi-Dirac distribution function with respect to the energy and δ the Dirac delta function. By integrating this function, the relaxation time approximation (RTA) hole mobility is obtained. The energy corresponding to the maximum of this curve is used for the phonon analysis.

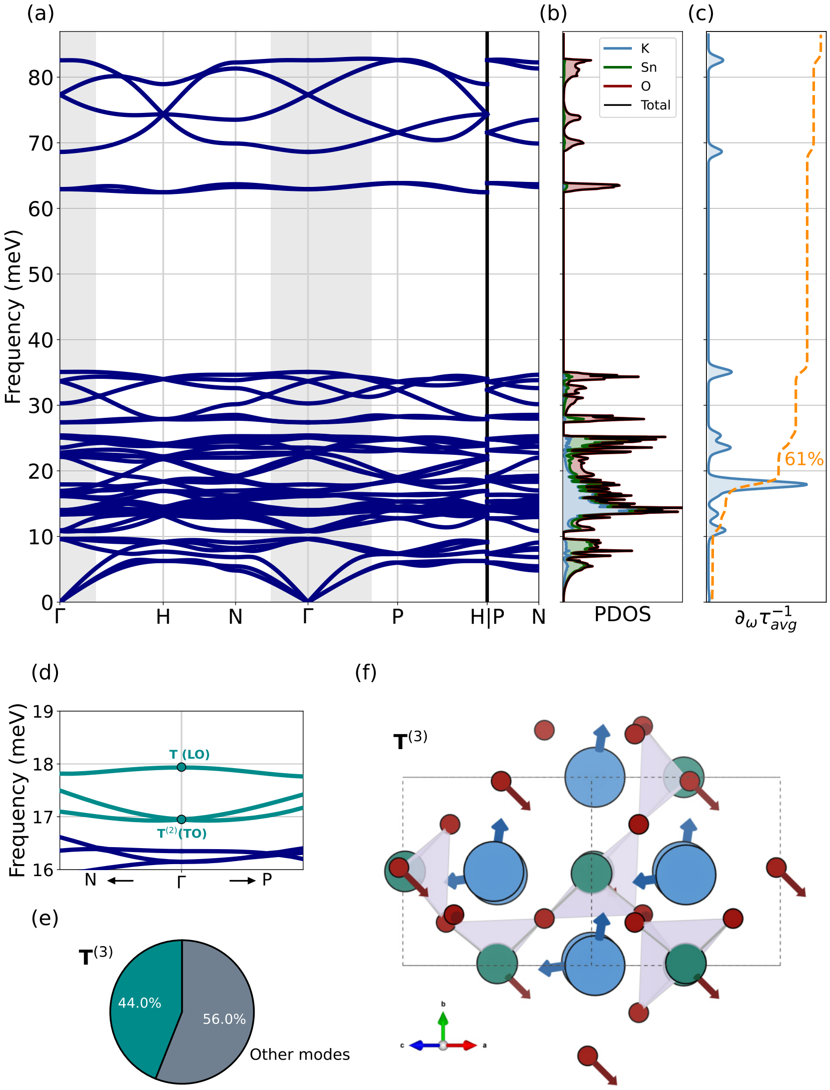

Fig. S6: **(a)** c-$K_2Sn_2O_3$ phonon dispersion computed using the PBEsol functional. The grey areas around Γ show the regions relevant (due to energy conservation) for the scattering, as expressed in Eq. (2). **(b)** PDOS with the contribution of K, Sn and O atoms in blue, green and red, respectively. **(c)** Spectral decomposition of the hole scattering rates at 300K. The scattering rates are computed at an energy of 23 meV from the VBM, corresponding to the energy for which the maximum number of states participate in hole transport. **(d)** Phonon dispersion at Γ illustrating the most important modes for hole scattering with **(e)** their relative importance and **(f)** the corresponding atomic displacements. K, Sn and O atoms are in blue, dark green and red, respectively.

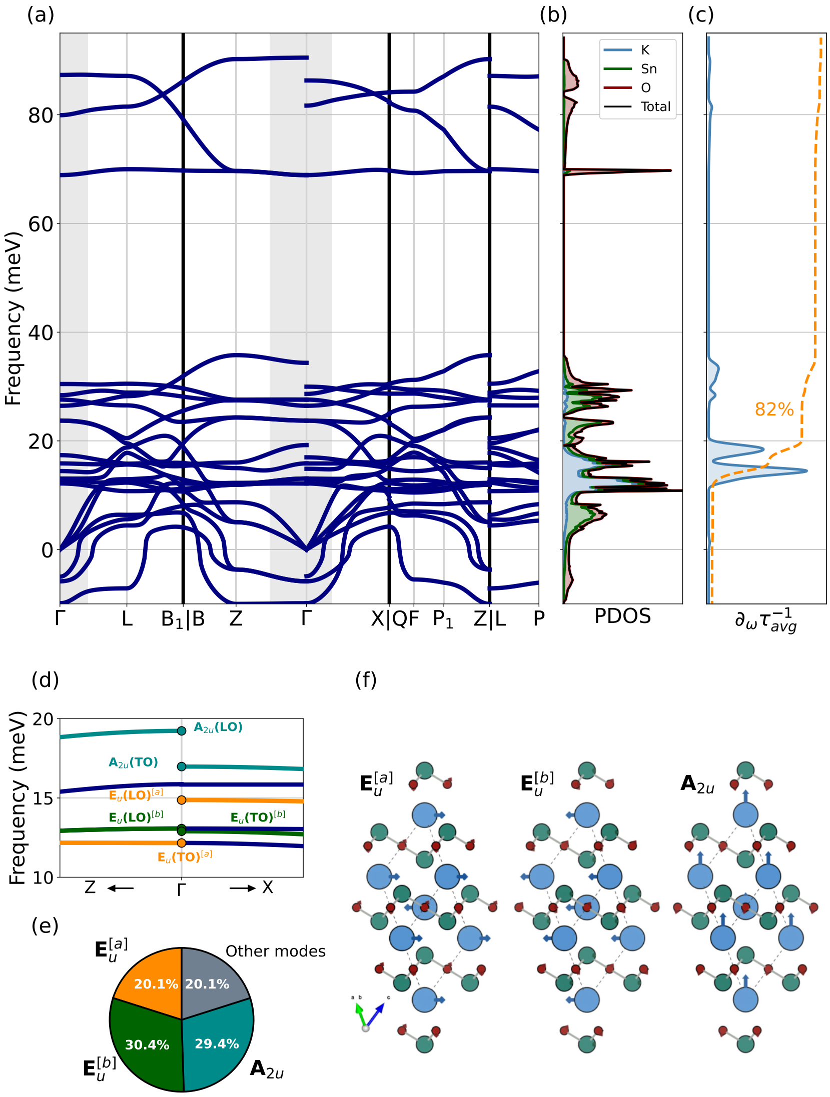

Fig. S7: **(a)** r-K$_2$Sn$_2$O$_3$ phonon dispersion computed using the PBEsol functional. The grey areas around Γ show the regions relevant (due to energy conservation) for the scattering, as expressed in Eq. (2). **(b)** PDOS with the contribution of K, Sn and O atoms in blue, green and red, respectively. **(c)** Spectral decomposition of the hole scattering rates at 300K. The scattering rates are computed at an energy of 25 meV from the VBM, corresponding to the energy for which the maximum number of states participate in hole transport. **(d)** Phonon dispersion at Γ illustrating the most important modes for hole scattering with **(e)** their relative importance and **(f)** the corresponding atomic displacements. K, Sn and O atoms are in blue, dark green and red, respectively.

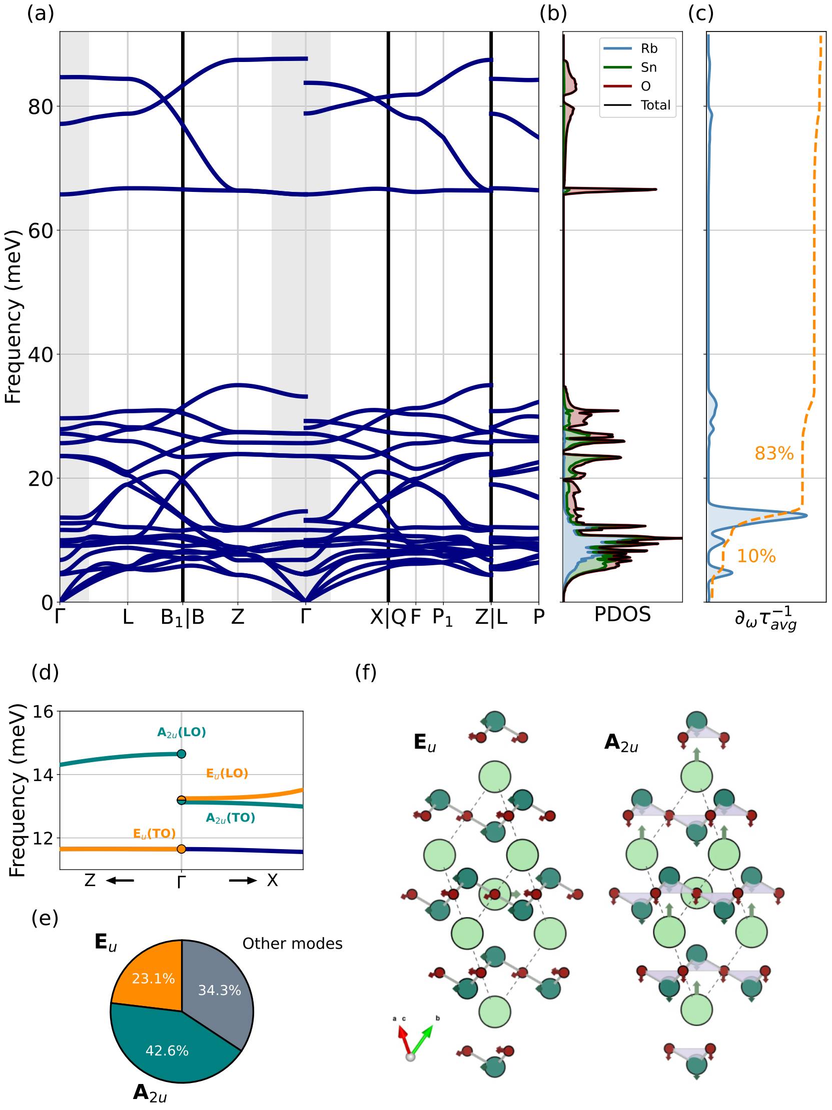

Fig. S8: **(a)** $Rb_2Sn_2O_3$ phonon dispersion computed using the PBEsol functional. The grey areas around Γ show the regions relevant (due to energy conservation) for the scattering, as expressed in Eq. (2). **(b)** PDOS with the contribution of Rb, Sn and O atoms in blue, green and red, respectively. **(c)** Spectral decomposition of the hole scattering rates at 300K. The scattering rates are computed at an energy of 30 meV from the VBM, corresponding to the energy for which the maximum number of states participate in hole transport. **(d)** Phonon dispersion at Γ illustrating the most important modes for hole scattering with **(e)** their relative importance and **(f)** the corresponding atomic displacements. Rb, Sn and O atoms are in lightgreen, dark green and red, respectively.

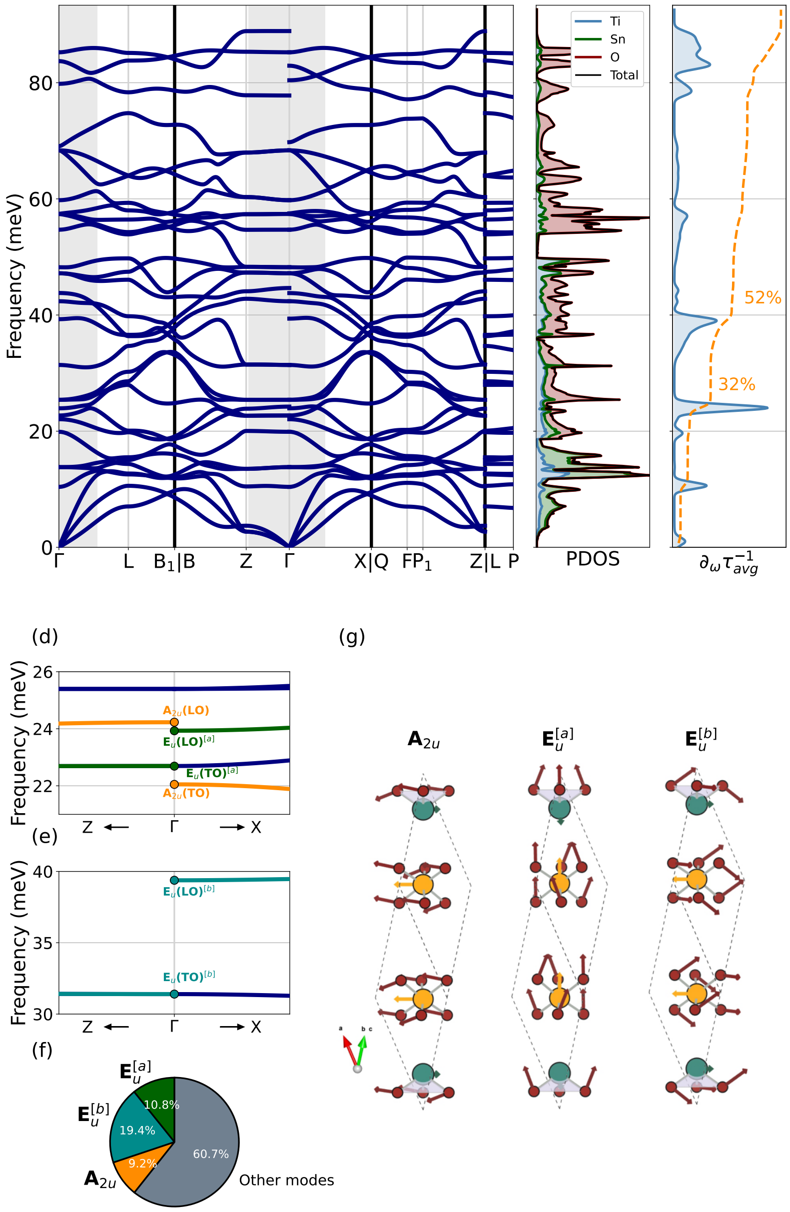

Fig. S9: **(a)** $TiSnO_3$ phonon dispersion computed using the PBEsol functional. The grey areas around Γ show the regions relevant (due to energy conservation) for the scattering, as expressed in Eq. (2). **(b)** PDOS with the contribution of Ti, Sn and O atoms in blue, green and red, respectively. **(c)** Spectral decomposition of the hole scattering rates at 300K. The scattering rates are computed at an energy of 28 meV from the VBM, corresponding to the energy for which the maximum number of states participate in hole transport. **(d)** and **(d)** Phonon dispersions at Γ illustrating the most important modes for hole scattering with **(f)** their relative importance and **(g)** the corresponding atomic displacements. Ti, Sn and O atoms are in orange, dark green and red, respectively.