Supplemental Material for Electron-phonon scattering and thermoelectric transport in p-type PbTe from first principles

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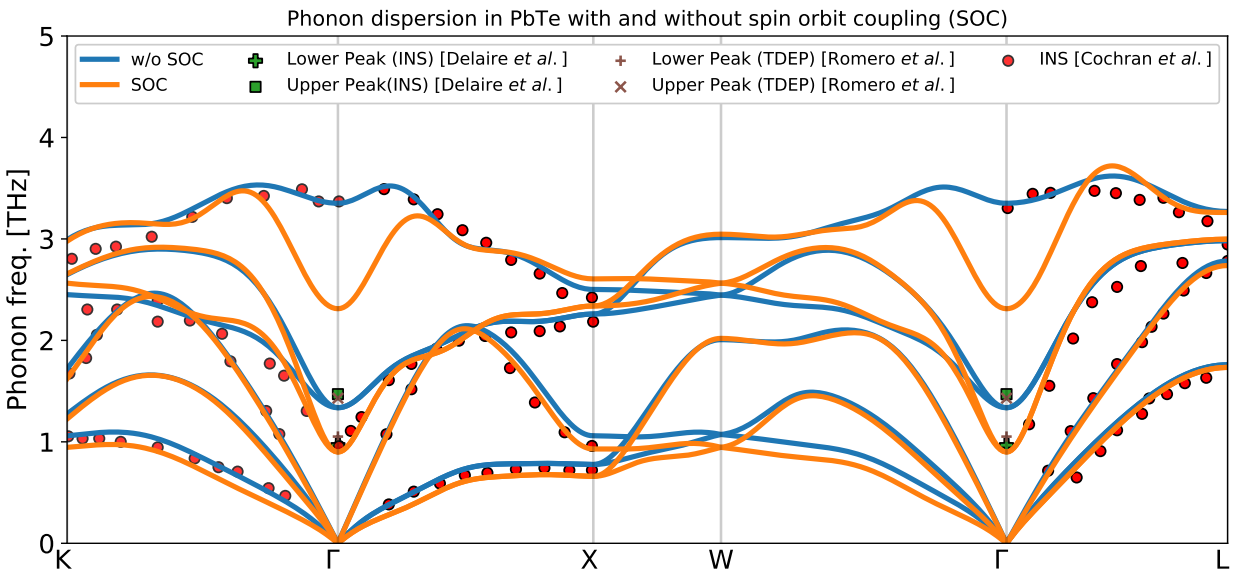

FIG. S1. Phonon dispersion of PbTe calculated using density functional perturbation theory and the local density approximation with and without spin orbit coupling (SOC) (blue and orange lines, respectively). The inelastic neutron scattering (INS) measurements from Cochran *et al.* [1] are represented by red circles. The experimental upper and lower transverse optical (TO) peaks at the Γ point from INS measurements of Delaire *et al.* [2] are represented by square and plus green symbols, respectively. The upper and lower TO peaks at Γ calculated using the temperature dependent effective potential (TDEP) method by Romero *et al.* [3] are shown in plus and cross brown symbols, respectively.

TABLE S1. Electron-phonon matrix elements of PbTe between the valence band maximum at one of the L points, (0.5, 0.5, 0.5), and the valence band maxima along all twelve Σ directions in the first Brillouin zone. TA_1 and TA_2 (TO_1 and TO_2) denote transverse acoustic (optical) modes. LA and LO stand for longitudinal acoustic and optical modes, respectively.

$\boldsymbol{k}$ = L (0.5, 0.5, 0.5)		ACOUSTIC					
λ (phonon mode)		TA_1		TA_2		LA	
$\boldsymbol{k'}$ = Σ (crystal coordinates)	$\boldsymbol{k}-\boldsymbol{k'}$	$\Xi^{\lambda}_{L\Sigma}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{L\Sigma}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{L\Sigma}$ [eV/Å]	ω_λ [meV]
(0.375, 0.1875, 0.1875)	(0.125, 0.3125, 0.3125)	0.00537522	3.6463	0.37363814	4.0493	0.97415839	8.149
(0.1875, -0.1875, 0.0)	(0.3125, 0.6875, 0.5)	0.05228076	4.637	0.00464080	4.9137	0.01242659	8.4383
(0.1875, 0, -0.1875)	(0.3125, 0.5, 0.6875)	0.03328585	4.637	0.01087413	4.9137	0.01320310	8.4383
(0, 0.1875, -0.1875)	(0.5, 0.3125, 0.6875)	0.03547255	4.637	0.00560566	4.9137	0.00301144	8.4383
(-0.1875, -0.1875, -0.375)	(0.6875, 0.6875, 0.875)	0.00733268	3.6463	0.39484111	4.0493	0.98467095	8.149
(0.1875, 0.1875, 0.375)	(0.3125, 0.3125, 0.125)	0.00733283	3.6463	0.39484157	4.0493	0.98467001	8.149
(0, -0.1875, 0.1875)	(0.5, 0.6875, 0.3125)	0.03547254	4.637	0.00560589	4.9137	0.00301140	8.4383
(-0.1875, 0, 0.1875)	(0.6875, 0.5, 0.3125)	0.03328604	4.637	0.01087384	4.9137	0.01320311	8.4383
(-0.1875, 0.1875, 0)	(0.6875, 0.3125, 0.5)	0.05228073	4.637	0.00464071	4.9137	0.01242656	8.4383
(-0.375, -0.1875, -0.1875)	(0.875, 0.6875, 0.6875)	0.00537521	3.6463	0.37363751	4.0493	0.97415931	8.149
(-0.1875, -0.375, -0.1875)	(0.6875, 0.875, 0.6875)	0.01181774	3.6463	0.39040430	4.0493	0.98785567	8.149
(0.1875, 0.375, 0.1875)	(0.3125, 0.125, 0.3125)	0.01181775	3.6463	0.39040485	4.0493	0.98785463	8.149

$\boldsymbol{k}$ = L (0.5, 0.5, 0.5)		OPTICAL					
λ (phonon mode)		TO_1		TO_2		LO	
$\boldsymbol{k'}$ = Σ (crystal coordinates)	$\boldsymbol{k}-\boldsymbol{k'}$	$\Xi^{\lambda}_{L\Sigma}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{L\Sigma}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{L\Sigma}$ [eV/Å]	ω_λ [meV]
(0.375, 0.1875, 0.1875)	(0.125, 0.3125, 0.3125)	0.01827590	9.7718	0.78818891	11.2738	1.48043333	12.9856
(0.1875, -0.1875, 0.0)	(0.3125, 0.6875, 0.5)	0.25536692	10.8167	0.01418282	10.8682	0.44962780	12.5526
(0.1875, 0, -0.1875)	(0.3125, 0.5, 0.6875)	0.21352907	10.8167	0.01501669	10.8682	0.45891137	12.5526
(0, 0.1875, -0.1875)	(0.5, 0.3125, 0.6875)	0.21143557	10.8167	0.02228118	10.8682	0.47998768	12.5526
(-0.1875, -0.1875, -0.375)	(0.6875, 0.6875, 0.875)	0.00498288	9.7718	0.74945308	11.2738	1.45550417	12.9856
(0.1875, 0.1875, 0.375)	(0.3125, 0.3125, 0.125)	0.00498263	9.7718	0.74945453	11.2738	1.45550228	12.9856
(0, -0.1875, 0.1875)	(0.5, 0.6875, 0.3125)	0.21143557	10.8167	0.02228084	10.8682	0.47998774	12.5526
(-0.1875, 0, 0.1875)	(0.6875, 0.5, 0.3125)	0.21352909	10.8167	0.01501637	10.8682	0.45891138	12.5526
(-0.1875, 0.1875, 0)	(0.6875, 0.3125, 0.5)	0.25536681	10.8167	0.01418275	10.8682	0.44962781	12.5526
(-0.375, -0.1875, -0.1875)	(0.875, 0.6875, 0.6875)	0.01827586	9.7718	0.78818746	11.2738	1.48043555	12.9856
(-0.1875, -0.375, -0.1875)	(0.6875, 0.875, 0.6875)	0.02274992	9.7718	0.77350308	11.2738	1.48435689	12.9856
(0.1875, 0.375, 0.1875)	(0.3125, 0.125, 0.3125)	0.02274979	9.7718	0.77350474	11.2738	1.48435518	12.9856

TABLE S2. Electron-phonon matrix elements of PbTe between the one of the valence band maxima at Σ, (0.1875, 0.375, 0.1875), and the valence band maxima along all twelve Σ directions in the first Brillouin zone.

$\boldsymbol{k}$ = Σ (0.1875, 0.375, 0.1875)		ACOUSTIC					
λ (phonon mode)		TA_1		TA_2		LA	
$\boldsymbol{k'}$ = Σ (crystal coordinates)	$\boldsymbol{k}-\boldsymbol{k'}$	$\Xi^{\lambda}_{\Sigma\Sigma}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{\Sigma\Sigma}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{\Sigma\Sigma}$ [eV/Å]	ω_λ [meV]
(0.375, 0.1875, 0.1875)	(0.1875, -0.1875, 0)	0.00556926	3.2914	0.00531492	6.2902	0.58447356	8.993
(0.1875, -0.1875, 0.0)	(0, -0.5625, -0.1875)	0.28898093	6.0652	0.01928725	6.7176	0.85409967	9.224
(0.1875, 0, -0.1875)	(0, -0.375, -0.375)	0.01370947	3.0408	0.01370947	3.0408	0.15071472	6.7821
(0, 0.1875, -0.1875)	(-0.1875, -0.1875, -0.375)	0.02585733	3.2914	0.04348656	6.2902	0.55983524	8.993
(-0.1875, -0.1875, -0.375)	(-0.375, -0.5625, -0.5625)	0.30149586	6.0652	0.01983353	6.7176	0.88193285	9.224
(0.1875, 0.1875, 0.375)	(0, -0.1875, 0.1875)	0.06585004	3.2914	0.01084862	6.2902	0.56919059	8.993
(0, -0.1875, 0.1875)	(-0.1875, -0.5625, 0)	0.29923702	6.0652	0.02055871	6.7176	0.83472230	9.224
(-0.1875, 0, 0.1875)	(-0.375, -0.375, 0)	0.01266830	3.0408	0.01266830	3.0408	0.18127715	6.7821
(-0.1875, 0.1875, 0)	(-0.375, -0.1875, -0.1875)	0.03718070	3.2914	0.02296725	6.2902	0.55348535	8.993
(-0.375, -0.1875, -0.1875)	(-0.5625, -0.5625, -0.375)	0.27741139	6.0652	0.02482761	6.7176	0.86185377	9.224
(-0.1875, -0.375, -0.1875)	(-0.375, -0.75, -0.375)	0.01546990	4.3318	0.01171215	5.2648	1.50217061	7.0542
(0.1875, 0.375, 0.1875)	(0, 0, 0)	0	0	0	0	0	0

$\boldsymbol{k}$ = Σ (0.1875, 0.375, 0.1875)		OPTICAL					
λ (phonon mode)		TO_1		TO_2		LO	
$\boldsymbol{k'}$ = Σ (crystal coordinates)	$\boldsymbol{k}-\boldsymbol{k'}$	$\Xi^{\lambda}_{\Sigma\Sigma}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{\Sigma\Sigma}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{\Sigma\Sigma}$ [eV/Å]	ω_λ [meV]
(0.375, 0.1875, 0.1875)	(0.1875, -0.1875, 0)	0.01644423	9.2675	0.04541800	12.0267	2.00242601	13.6257
(0.1875, -0.1875, 0.0)	(0, -0.5625, -0.1875)	0.11142206	12.1425	0.02798880	12.3347	0.54873280	13.3546
(0.1875, 0, -0.1875)	(0, -0.375, -0.375)	0.04378253	9.3552	0.04378253	9.3552	0.09164763	11.642
(0, 0.1875, -0.1875)	(-0.1875, -0.1875, -0.375)	0.05847350	9.2675	0.06860462	12.0267	2.02428529	13.6257
(-0.1875, -0.1875, -0.375)	(-0.375, -0.5625, -0.5625)	0.02312252	12.1425	0.01530666	12.3347	0.53863574	13.3546
(0.1875, 0.1875, 0.375)	(0, -0.1875, 0.1875)	0.07251402	9.2675	0.02006236	12.0267	2.04872690	13.6257
(0, -0.1875, 0.1875)	(-0.1875, -0.5625, 0)	0.04269751	12.1425	0.06438753	12.3347	0.51529728	13.3546
(-0.1875, 0, 0.1875)	(-0.375, -0.375, 0)	0.02457348	9.3552	0.02457348	9.3552	0.14937458	11.642
(-0.1875, 0.1875, 0)	(-0.375, -0.1875, -0.1875)	0.05800702	9.2675	0.04978086	12.0267	2.02926247	13.6257
(-0.375, -0.1875, -0.1875)	(-0.5625, -0.5625, -0.375)	0.07535207	12.1425	0.05475274	12.3347	0.56246126	13.3546
(-0.1875, -0.375, -0.1875)	(-0.375, -0.75, -0.375)	0.05114207	10.3939	0.02298381	11.0684	0.27293883	12.5984
(0.1875, 0.375, 0.1875)	(0, 0, 0)	0.02623423	4.6112	0.02623423	4.6112	0.02623423	4.6112

TABLE S3. Electron-phonon matrix elements of PbTe between the one of the valence band maxima at Σ, (0.1875, 0.375, 0.1875), and the valence band maxima at all four L points in the first Brillouin zone.

$\boldsymbol{k}$ = Σ (0.1875, 0.375, 0.1875)		ACOUSTIC					
λ (phonon mode)		TA_1		TA_2		LA	
$\boldsymbol{k'}$ = L (crystal coordinates)	$\boldsymbol{k-k'}$	$\Xi^{\lambda}_{\Sigma L}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{\Sigma L}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{\Sigma L}$ [eV/Å]	ω_λ [meV]
(0.5, 0.5, 0.5)	(0.3125, 0.125, 0.3125)	0.010283	3.6463	0.343957	4.0493	0.959388	8.1490
(0.0, 0.0, -0.5)	(-0.1875, -0.375, -0.6875)	0.027108	4.6370	0.007845	4.9137	0.015433	8.4383
(0.0, -0.5, 0.0)	(-0.1875, -0.875, -0.1875)	0.012120	3.6463	0.394212	4.0493	0.910912	8.1490
(-0.5, 0.0, 0.0)	(-0.6875, -0.375, -0.1875)	0.025131	4.6370	0.037676	4.9137	0.053357	8.4383
$\boldsymbol{k}$ = Σ (0.1875, 0.375, 0.1875)		OPTICAL					
λ (phonon mode)		TO_1		TO_2		LO	
$\boldsymbol{k'}$ = L (crystal coordinates)	$\boldsymbol{k-k'}$	$\Xi^{\lambda}_{\Sigma L}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{\Sigma L}$ [eV/Å]	ω_λ [meV]	$\Xi^{\lambda}_{\Sigma L}$ [eV/Å]	ω_λ [meV]
(0.5, 0.5, 0.5)	(0.3125, 0.125, 0.3125)	0.019530	9.7718	0.760915	11.2738	1.467691	12.9856
(0.0, 0.0, -0.5)	(-0.1875, -0.375, -0.6875)	0.181667	10.8167	0.034469	10.8682	0.471954	12.5526
(0.0, -0.5, 0.0)	(-0.1875, -0.875, -0.1875)	0.008135	9.7718	0.753037	11.2738	1.466387	12.9856
(-0.5, 0.0, 0.0)	(-0.6875, -0.375, -0.1875)	0.203825	10.8167	0.050496	10.8682	0.502266	12.5526

TABLE S4. Electron-phonon matrix elements of PbTe between the valence band maximum at one of the L points, (0.5, 0.5, 0.5), and the valence band maxima along all four L points in the first Brillouin zone.

$\boldsymbol{k}$ = L (0.5, 0.5, 0.5)		ACOUSTIC					
λ (phonon mode)		TA_1		TA_2		LA	
$\boldsymbol{k'}$ = L (crystal coordinates)	$\boldsymbol{k-k'}$	Ξ^{λ}_{LL} [eV/Å]	ω_λ [meV]	Ξ^{λ}_{LL} [eV/Å]	ω_λ [meV]	Ξ^{λ}_{LL} [eV/Å]	ω_λ [meV]
(0.5, 0.5, 0.5)	(0, 0, 0)	0	0	0	0	0	0
(0.0, 0.0, -0.5)	(-0.5, -0.5, -1)	7.89E-10	2.9861	7.89E-10	2.9861	7.89E-10	4.0823
(0.0, -0.5, 0.0)	(-0.5, -1, -0.5)	2.26E-10	2.9861	2.26E-10	2.9861	3.62E-11	4.0823
(-0.5, 0.0, 0.0)	(-1, -0.5, -0.5)	1.68E-10	2.9861	1.68E-10	2.9861	8.46E-11	4.0823
$\boldsymbol{k}$ = L (0.5, 0.5, 0.5)		OPTICAL					
λ (phonon mode)		TO_1		TO_2		LO	
$\boldsymbol{k'}$ = L (crystal coordinates)	$\boldsymbol{k-k'}$	Ξ^{λ}_{LL} [eV/Å]	ω_λ [meV]	Ξ^{λ}_{LL} [eV/Å]	ω_λ [meV]	Ξ^{λ}_{LL} [eV/Å]	ω_λ [meV]
(0.5, 0.5, 0.5)	(0, 0, 0)	1.52E-10	4.6112	1.52E-10	4.6112	1.52E-10	4.6112
(0.0, 0.0, -0.5)	(-0.5, -0.5, -1)	3.74E-10	9.4853	3.74E-10	9.4853	4.31E-09	10.3983
(0.0, -0.5, 0.0)	(-0.5, -1, -0.5)	9.60E-11	9.4853	9.60E-11	9.4853	4.58E-09	10.3983
(-0.5, 0.0, 0.0)	(-1, -0.5, -0.5)	1.47E-10	9.4853	1.47E-10	9.4853	4.69E-09	10.3983

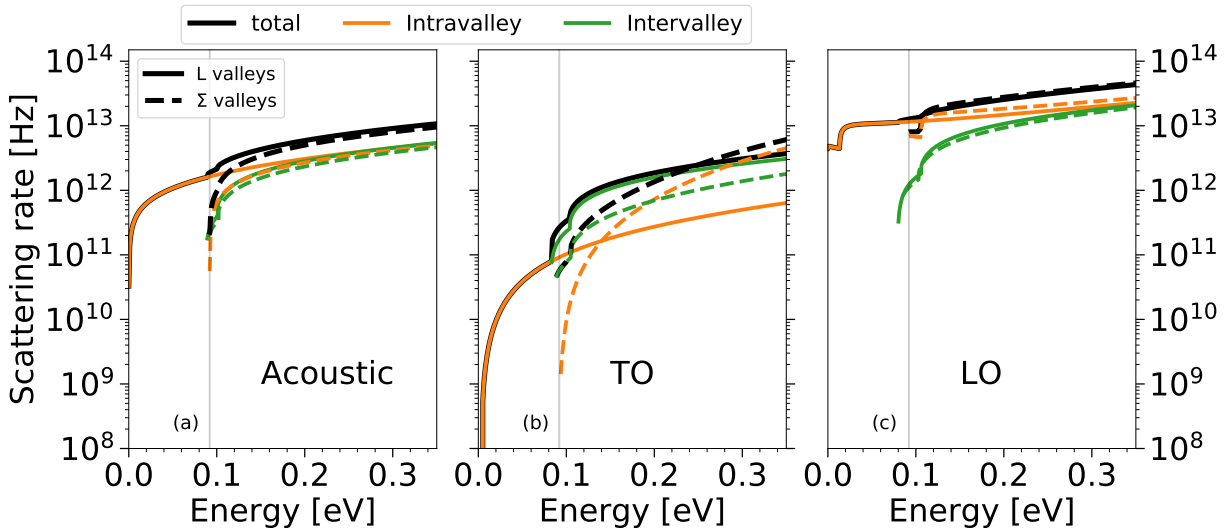

FIG. S2. Electron-phonon scattering rates at 300 K for the valence bands of PbTe versus hole energy (from the valence band maxima at L corresponding to zero energy and below). The scattering rates are calculated using the electronic band structure at 300 K and resolved by (a) acoustic, (b) transverse optical (TO) and (c) longitudinal optical (LO) phonon modes. Solid (dashed) lines refer to the scattering rates for the L (Σ) valleys. The intravalley and intervalley contributions to the total scattering rate (black lines) are given by orange and green lines, respectively. The gray vertical line corresponds to the energy difference between the L and Σ valleys at 300 K.

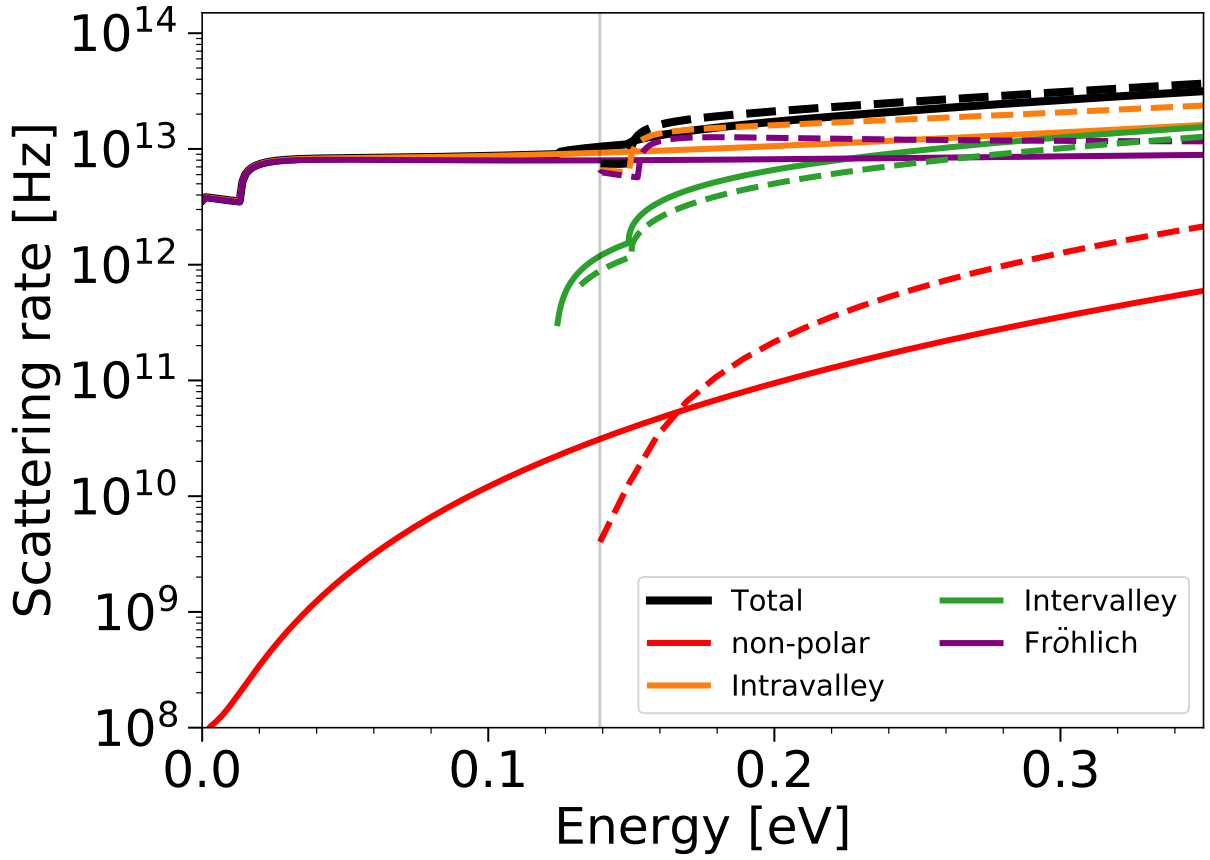

FIG. S3. The breakdown of the different contributions to the scattering rate due to longitudinal optical phonons. The polar Fröhlich and non-polar intravalley contributions are shown in purple and red, respectively. Orange line shows the total intravalley scattering rate whose electron-phonon matrix element is determined as the coherent sum of the polar and non-polar intravalley matrix elements. The intervalley and total scattering rates are given in green and black, respectively. Solid (dashed) lines refer to the scattering rates calculated for the L (Σ) valleys.

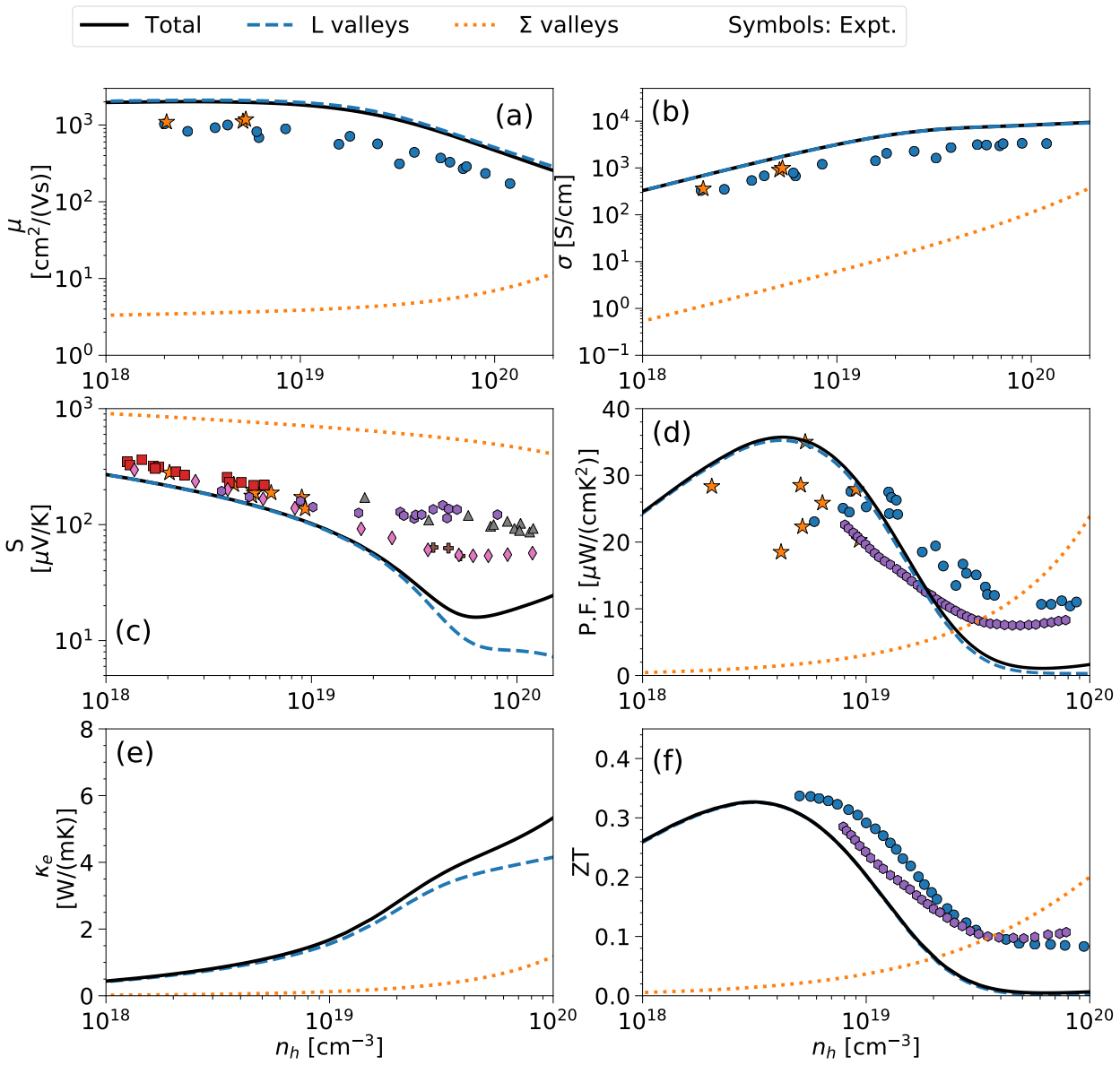

FIG. S4. Room temperature thermoelectric transport parameters of p-type PbTe without the temperature renormalization of the electronic band structure: (a) mobility (μ), (b) conductivity (σ), (c) Seebeck coefficient (S), (d) power factor (PF), (e) electrical thermal conductivity (κ_e) and (f) figure of merit (ZT) as a function of doping concentration n_h. Solid black lines show the results obtained using our model and the electronic band structure at 0 K. Dashed blue and dotted orange lines represent the contributions from the L and Σ valleys, respectively. Symbols represent the same experimental measurements as mentioned in the caption of Fig. 4 in the paper.

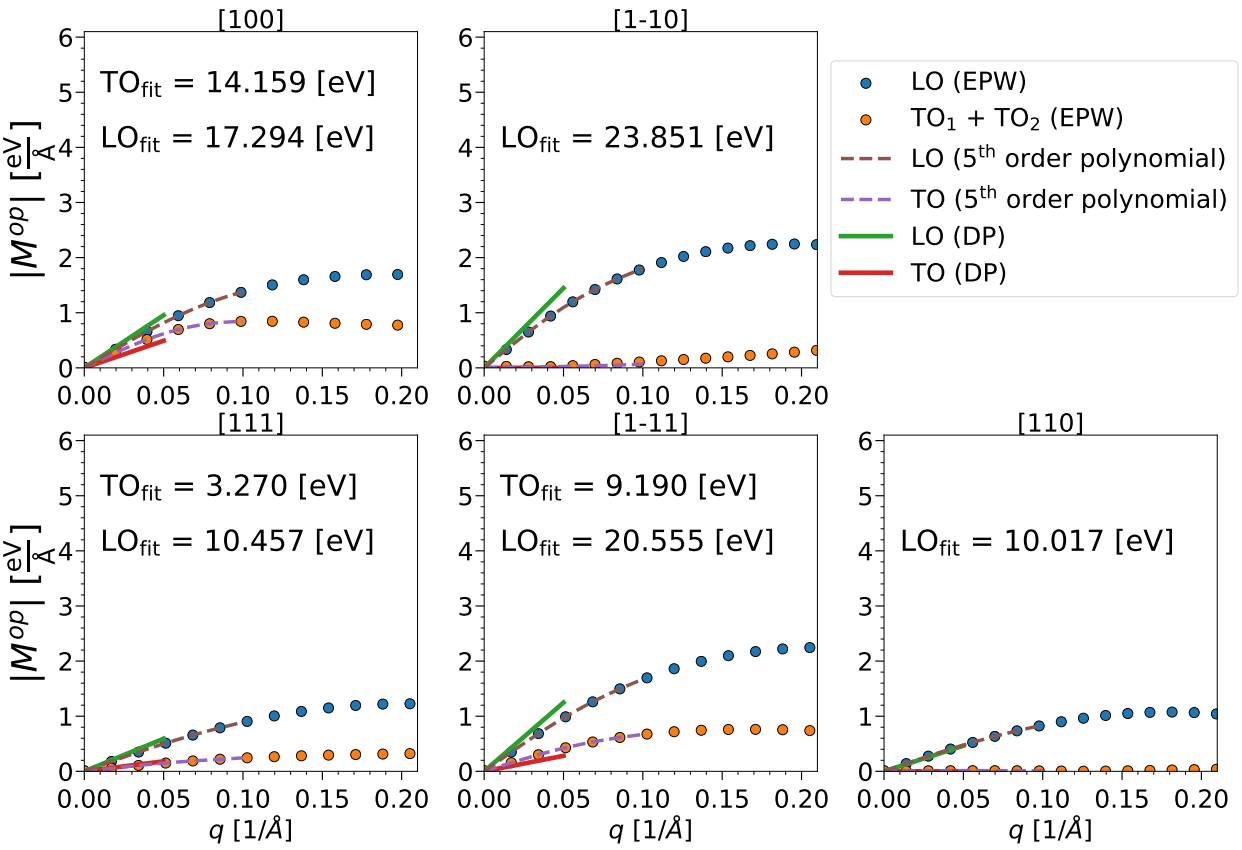

FIG. S5. Electron-phonon matrix elements due to optical modes between the states Σ (0.1875, 0.375, 0.1875) and $\Sigma+\boldsymbol{q}$ as a function of the phonon wave vector $\boldsymbol{q}$ whose directions are indicated at the top of each panel. Orange and blue dots represent the matrix elements calculated using density functional perturbation theory and electron-phonon Wannier (EPW) approach for transverse and longitudinal acoustic modes, respectively. Dashed lines are the 5th order polynomial fits to the EPW matrix elements. Red (green) lines are obtained using the computed values of deformation potentials (DP) given in Table III of the main paper for transverse (longitudinal) optical modes in the long wavelength limit.

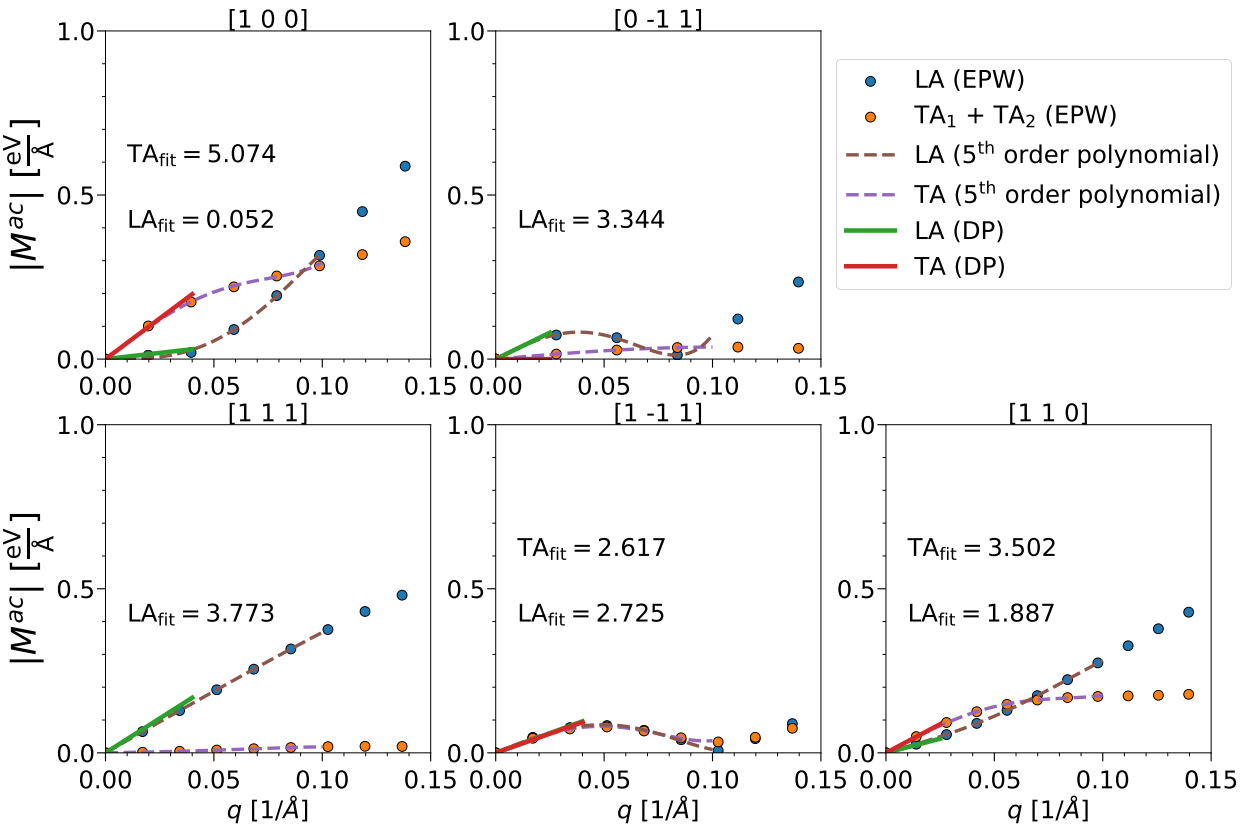

FIG. S6. Electron-phonon matrix elements due to acoustic modes between the states L (0.5, 0,5, 0,5) and L+$\boldsymbol{q}$ as a function of the phonon wave vector $\boldsymbol{q}$ whose directions are indicated at the top of each panel. Orange and blue dots represent the matrix elements calculated using density functional perturbation theory and electron-phonon Wannier (EPW) approach for transverse and longitudinal acoustic modes, respectively. Dashed lines are the 5th order polynomial fits to the EPW matrix elements. Red (green) lines are obtained using the computed values of deformation potentials (DP) given in Table III of the main paper for transverse (longitudinal) acoustic modes in the long wavelength limit.

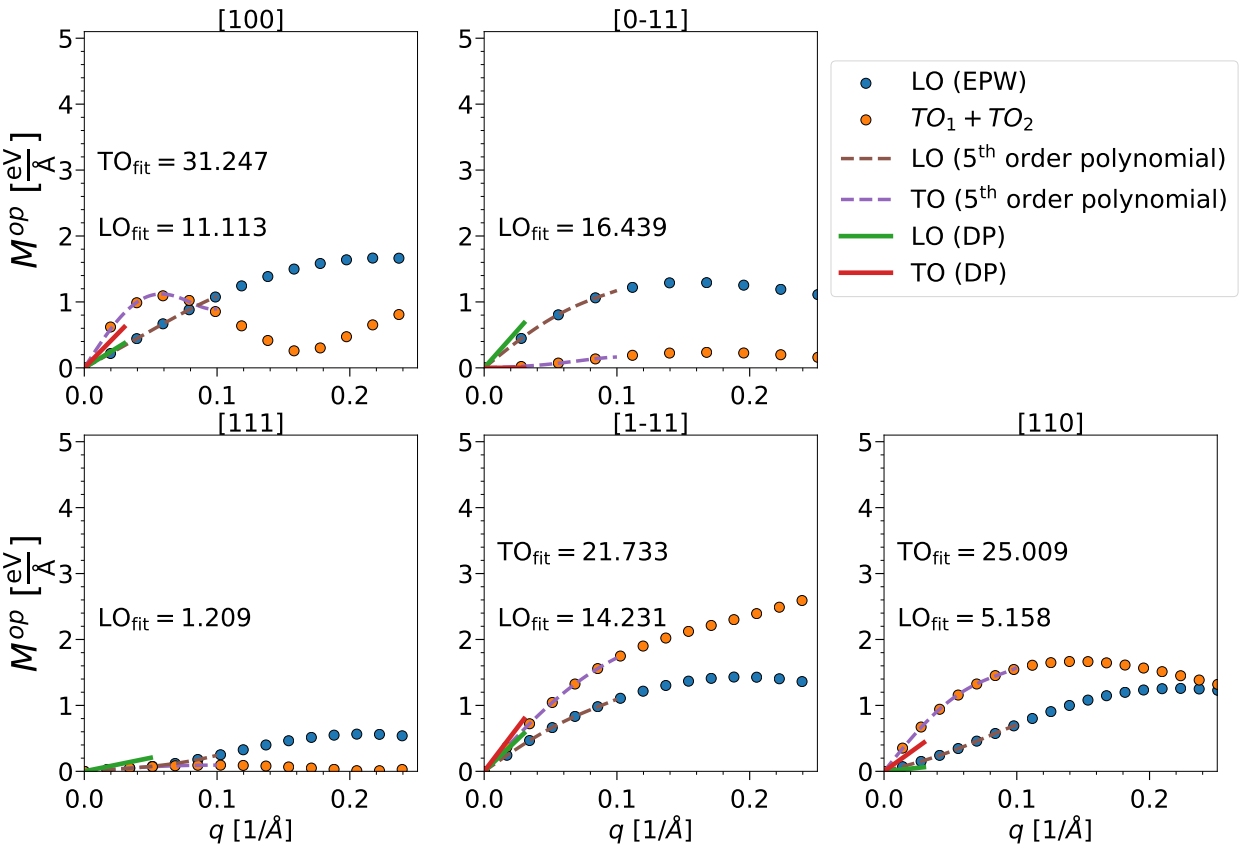

FIG. S7. Electron-phonon matrix elements due to optical modes between the states L and L+$\boldsymbol{q}$ as a function of the phonon wave vector $\boldsymbol{q}$ whose directions are indicated at the top of each panel. Orange and blue dots represent the matrix elements calculated using density functional perturbation theory and electron-phonon Wannier (EPW) approach for transverse and longitudinal acoustic modes, respectively. Dashed lines are the 5^{th} order polynomial fits to the EPW matrix elements. Red (green) lines are obtained using the computed values of deformation potentials (DP) given in Table III of the main paper for transverse (longitudinal) optical modes in the long wavelength limit.

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