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SUPPLEMENTARY INFORMATION

The role of structural order and stiffness in the simultaneous enhancement of optical contrast and thermal stability in phase change materials

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Table S1 Parameters of fitted spectrum for GeTe and $Ge_2Sb_2Te_5$ films with different T_a.

Parameters	$Ge_2Sb_2Te_5$ Cub. 250 °C	$Ge_2Sb_2Te_5$ Hex. 350 °C	GeTe Rhom. 250 °C	GeTe Cub. 350 °C
A (eV)	148.9	206.4	200.9	241.4
C (eV)	2.36	1.67	1.21	1.17
E_0 (eV)	1.87	1.83	1.85	1.82
E_g (eV)	0.51	0.4	0.64	0.63
ε_{hf} (eV)	4.15	5.24	4.91	5.78

Table S2 Parameters of fitted Raman spectra for $Ge_2Sb_2Te_5$ and GeTe at T_a = 250 °C and T_a = 350 °C.

		$Ge_2Sb_2Te_5$ Cub. 250 °C			$Ge_2Sb_2Te_5$ Hex. 350 °C			GeTe Rhom. 250 °C			GeTe Cub. 350 °C		
Peak assignment	Peak identity	Raman shift (cm^{-1})	FWHM (cm^{-1})	Peak area (%)	Raman shift (cm^{-1})	FWHM (cm^{-1})	Peak area (%)	Raman shift (cm^{-1})	FWHM (cm^{-1})	Peak area (%)	Raman shift (cm^{-1})	FWHM (cm^{-1})	Peak area (%)
P_2	Octahedral Ge	95	25.7	26.1	95	23.9	27.8	95	23.7	27.2	95	22.9	29.7
P_3	$GeTe_{4-n}Ge_n$ (n = 0) corner shared tetrahedra	112	26.5	14.1	113	20.2	12.9	112	23.0	21.1	112	19.5	20.0
P_4	Distorted octahedral Ge + defective octahedral Ge	126	35.6	19.9	132	26.2	19.3	127	30.1	23.7	128	29.1	19.6
P_5	Defective octahedral Ge							156	28.3	8.1	156	25.7	8.9
P_6	$GeTe_{4-n}Ge_n$ (n = 1, 2) edge-sharing tetrahedra	181	42.0	13.7	186	35.3	14.2	180	39.2	6.7	183	26.8	7.0
P_7	$GeTe_{4-n}Ge_n$ (n = 2) corner shared tetrahedra	236	71.3	10.4	233	49.9	10.5	231	85.1	13.2	183	26.8	7.0
P_{11}	Sb-Te vibrations in cubic Sb_2Te_3 units	150	34.4	15.7									
P_{12}	Mode of hexagonal Sb_2Te_3 phase				159	25.2	15.5						

Table S3 Parameters of fitted Raman spectra for GeTe at as-deposited (as-dep.), T_a = 250 °C and T_a = 350 °C.

		GeTe Amor. As-dep.			GeTe Rhom. 250 °C			GeTe Cub. 350 °C		
Peak assignment	Peak identity	Raman shift (cm^{-1})	FWHM (cm^{-1})	Peak area (%)	Raman shift (cm^{-1})	FWHM (cm^{-1})	Peak area (%)	Raman shift (cm^{-1})	FWHM (cm^{-1})	Peak area (%)
P_1	Symmetric bending mode of $GeTe_4$	91	20.2	5.9						
P_2	Octahedral Ge				95	23.7	27.2	95	22.9	29.7
P_3	$GeTe_{4-n}Ge_n$ (n = 0) corner shared tetrahedra	112	34.3	27.5	112	23.0	21.1	112	19.5	20.0
P_4	Distorted octahedral Ge + defective octahedral Ge	129	56.2	23.1	127	30.1	23.7	128	29.1	19.6
P_5	Defective octahedral Ge	156	42.4	15.7	156	28.3	8.1	156	25.7	8.9
P_6	$GeTe_{4-n}Ge_n$ (n = 1, 2) edge-sharing tetrahedra	190	53.7	14.2	180	39.2	6.7	183	26.8	7.0
P_7	$GeTe_{4-n}Ge_n$ (n = 2) Corner shared tetrahedra	236	65.6	13.6	231	85.1	13.2	183	26.8	7.0

Table S4 Parameters of fitted Raman spectra for $GeTe_4$ at as-deposited (as-dep.), T_a = 250 °C and T_a = 350 °C.

		$GeTe_4$ Amor. As-dep.			$GeTe_4$ Rhom. 250 °C			$GeTe_4$ Cub. 350 °C		
Peak assignment	Peak identity	Raman shift (cm^{-1})	FWHM (cm^{-1})	Peak area (%)	Raman shift (cm^{-1})	FWHM (cm^{-1})	Peak area (%)	Raman shift (cm^{-1})	FWHM (cm^{-1})	Peak area (%)
P_1	Symmetric bending mode of $GeTe_4$	91	20.6	8.5						
P_2	Octahedral Ge				94	19.3	11.4	95	24.3	20.3
P_3	$GeTe_{4-n}Ge_n$ (n = 0) corner shared tetrahedra	112	25.2	13.5	112	9.8	23.1	112	10.5	22.0
P_4	Distorted octahedral Ge + defective octahedral Ge	123	14.3	15.3						
P_5	Defective octahedral Ge				154	47.4	10.7	156	53.5	17.2
P_7	$GeTe_{4-n}Ge_n$ (n = 2) Corner shared tetrahedra	217	104.4	21.6	228	80.1	7.1	221	71.5	12.3
P_8	Vibrations of short disordered Te-Te chains	149	33.1	41.0						
P_9	c-$GeTe_4$				119	9.8	30.3	119	9.3	16.4
P_{10}	Crystalline Te vibrations				135	13.5	17.4	133	12.7	11.7

Table S5 Parameters of fitted spectrum for $GeTe_4$ films at T_a = 250 °C and T_a = 350 °C.

Parameters	$GeTe_4$ Cub. 250°C	$GeTe_4$ Cub. 350°C
A (eV)	138.5	112.0
C (eV)	1.46	2.03
E_0 (eV)	1.92	1.85
E_g (eV)	0.67	0.63
ε_{hf} (eV)	4.73	4.36

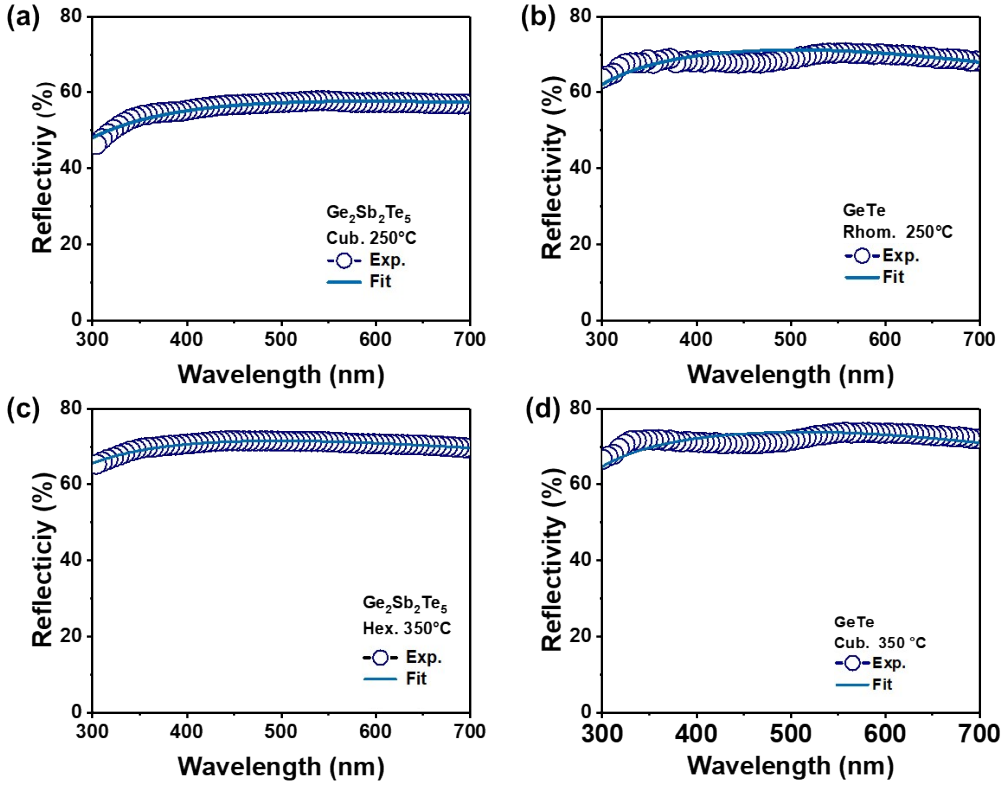

Fig. S1 The experimental and fitted reflectance spectra for (a) $Ge_2Sb_2Te_5$ and (b) GeTe films at T_a = 250 °C. The experimental and fitted reflectance spectra for (c) $Ge_2Sb_2Te_5$ and (d) GeTe films T_a = 350 °C.

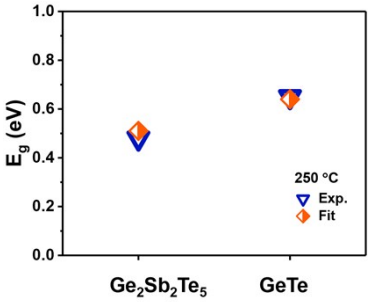

Fig. S2 The experimental and fitted optical bandgap for $Ge_2Sb_2Te_5$ and GeTe films at T_a = 250 °C.

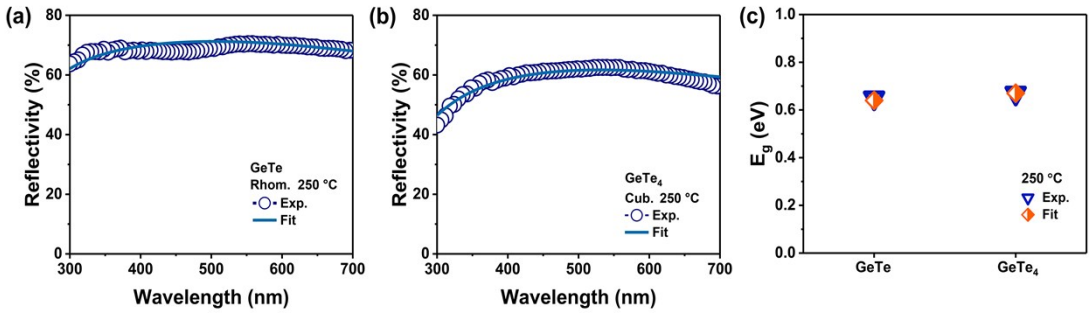

Fig. S3 The experimental and fitted reflectance spectrum for GeTe (a) and $GeTe_4$ (b) films at T_a = 250 °C. (c) The experimental and fitted optical bandgap for GeTe and $GeTe_4$ films at T_a = 250 °C.

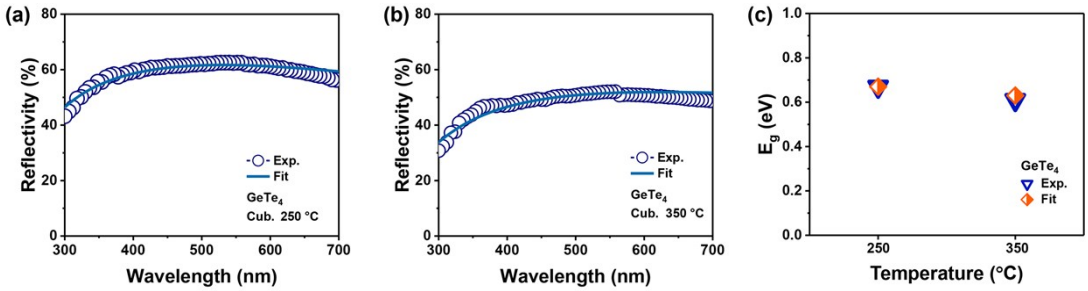

Fig. S4 The experimental and fitted reflectance spectrum for $GeTe_4$ films at T_a = 250 °C (a) and T_a = 350 °C (b). (c) The experimental and fitted optical bandgap for $GeTe_4$ films at T_a = 250 °C and T_a = 350 °C.