

Supplemental Materials:

Laser Ablation of Ga–Sb–Te Thin Films Monitored with Quadrupole Ion Trap Time-of-Flight Mass Spectrometry

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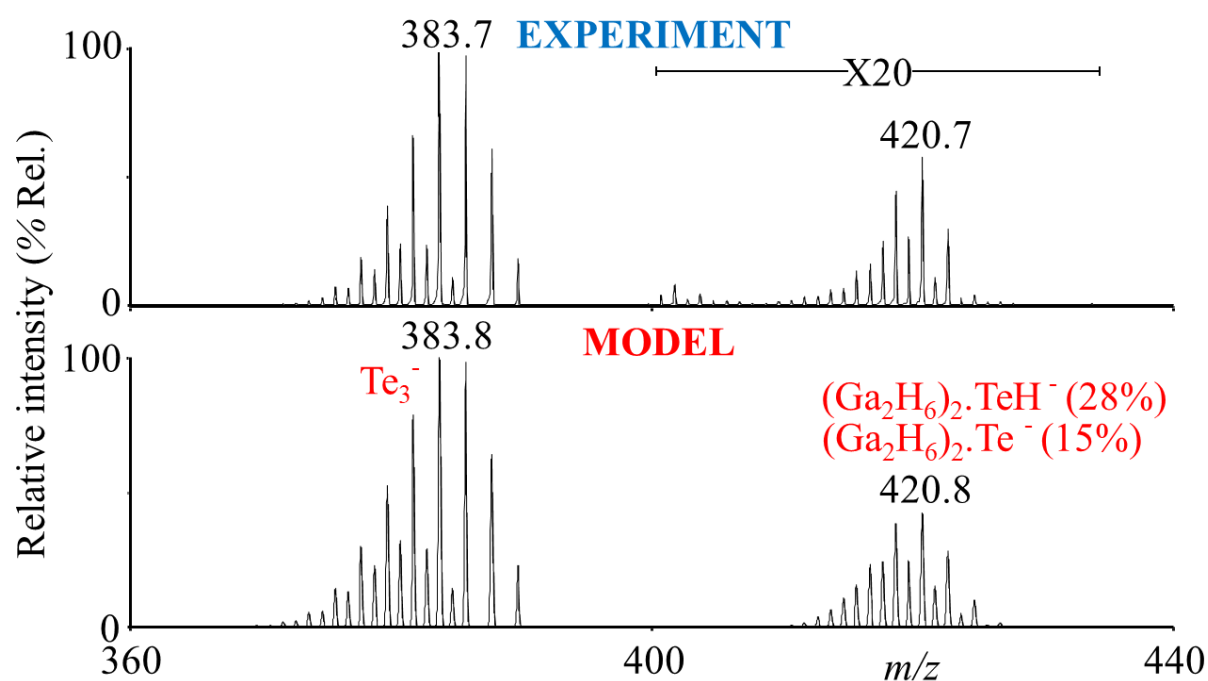


FIGURE S1 Comparison of the experimental and theoretical mass spectra recorded via LA for thin film **F** coated with trehalose. Conditions: negative ion mode, laser energy 140 a.u., m/z range 400-434 was magnified 20 times, 2000 profiles, and 100% intensity corresponds to 320 mV

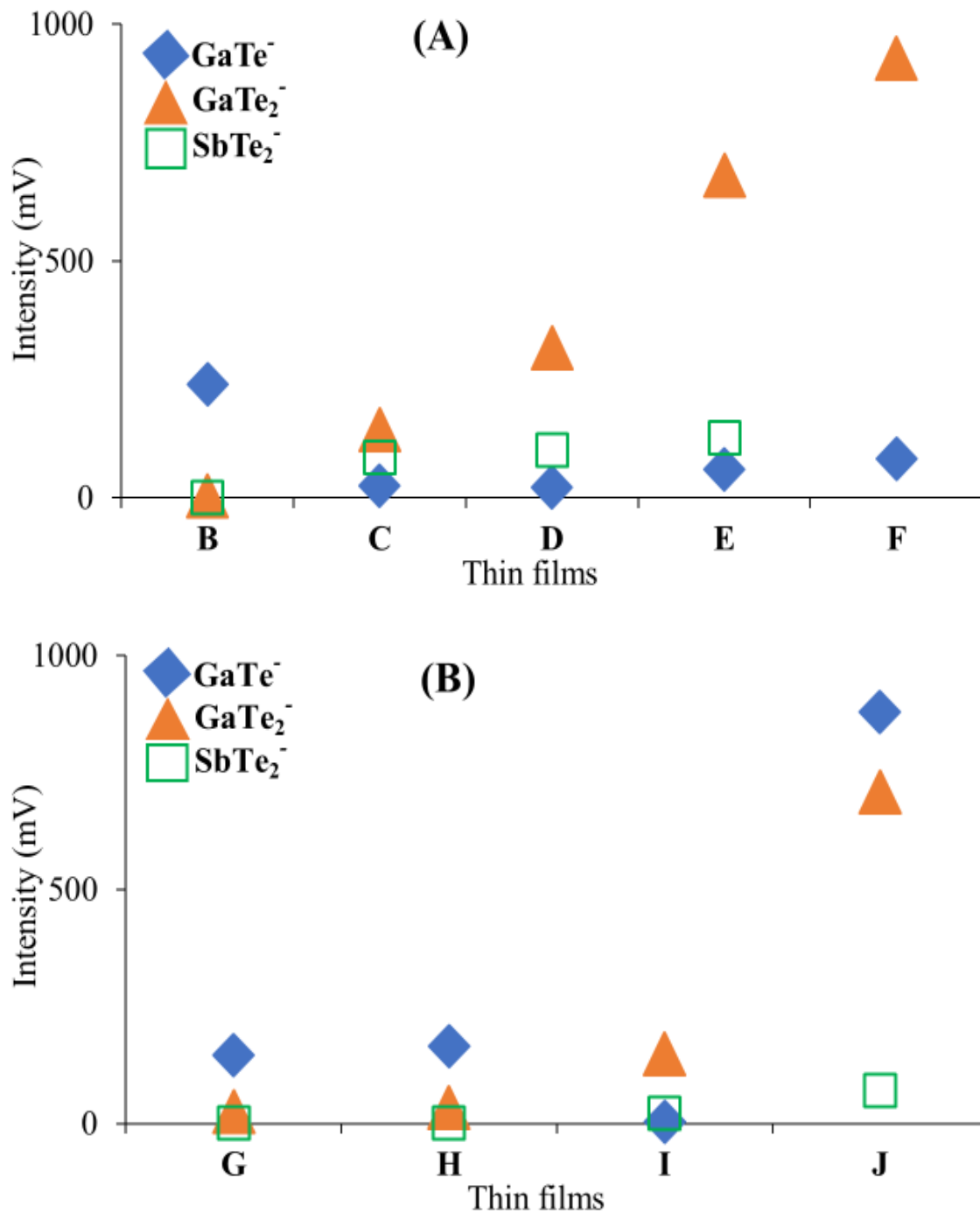


FIGURE S2 The variation in peaks intensities for selected clusters for: (A) the line 1 chemical composition of thin films (B-F), and (B) the line 2 chemical composition of thin films (G-J), which were shown in Figure 1. Conditions: negative ion mode, laser energy 150 a.u., and 2000 profiles

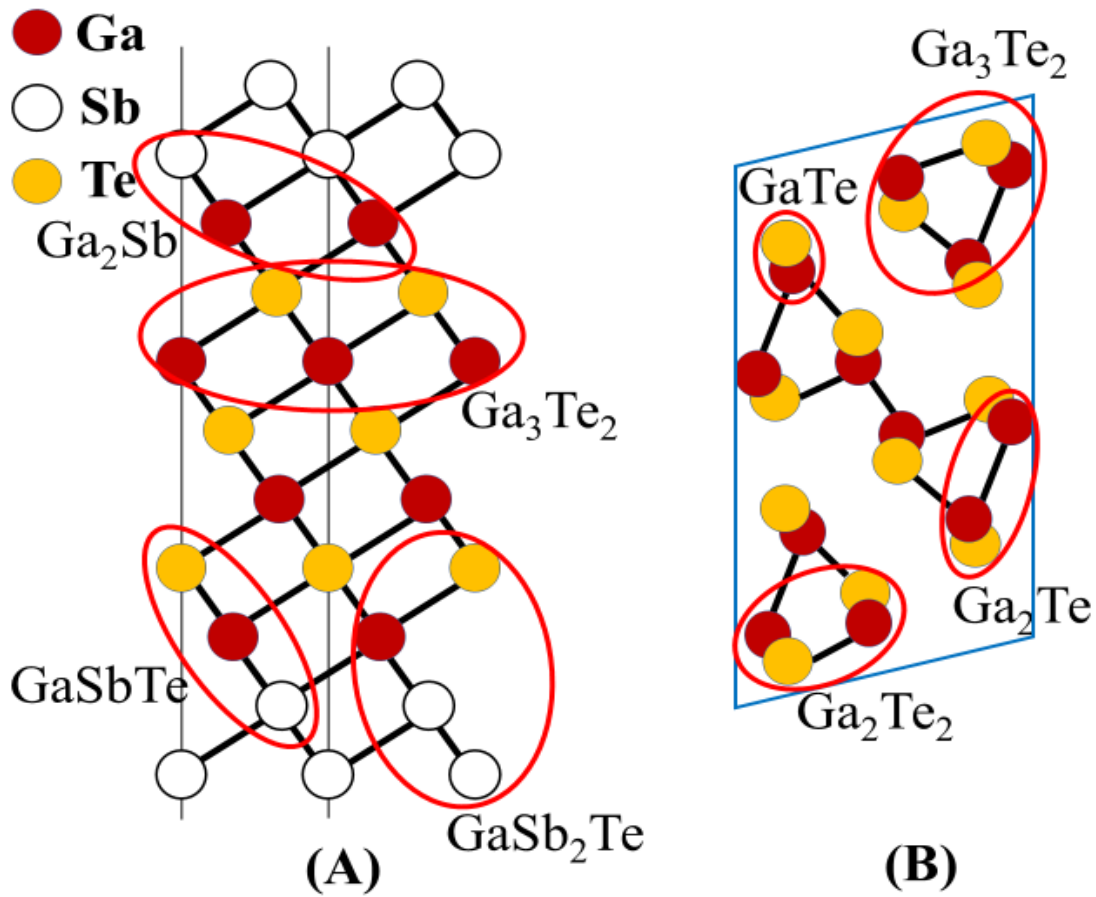


FIGURE S3 Structures of (A) the crystalline Ga₄Sb₆Te₃ phase change material, as adapted from Baratella et al.,³³ and (B) the structure of GaTe binary phase change alloy, as adapted from Bouzid et al.²³ Analogies between the possible fractions of the phase change material and selected clusters from the fabricated thin films are marked with ellipses

TABLE S1 (A) Overview of $\text{Ga}_x\text{Sb}_y\text{Te}_z$ clusters formed via LA from individual Ga-Sb-Te thin films (A-F) in both positive and negative ion modes

Thin film	Ga_x	Sb_y	Te_z	Ga_xSb_y	Ga_xTe_z	Sb_yTe_z	$\text{Ga}_x\text{Sb}_y\text{Te}_z$
A		Sb^- Sb_2^- Sb_3^-		GaSb_2^-			
B	Ga_2^+	Sb^+ Sb_3^+	Te^- Te_2^-	GaSb^+	GaTe^- GaTe_2^-	$\text{SbTe}^{+/-}$ SbTe_2^- Sb_3Te^+	
C	Ga_2^+	Sb^+	Te^- Te_2^-		GaTe^- GaTe_2^-	$\text{SbTe}^{+/-}$ Sb_2Te^+ Sb_3Te^+	
D	Ga_2^+	Sb^+ Sb_9^+	$\text{Te}^{+/-}$ $\text{Te}_2^{+/-}$	$\text{GaSb}_8^{+/-}$	GaTe^- GaTe_2^- Ga_2Te^+	$\text{SbTe}_2^{+/-}$ Sb_2Te^+ Sb_3Te^+ SbTe_2^- SbTe_3^-	GaSbTe^+
E	$\text{Ga}_2^{+/-}$	Sb^+ $\text{Sb}_9^{+/-}$	$\text{Te}^{+/-}$ $\text{Te}_2^{+/-}$ Te_3^-	GaSb_8^- Ga_2Sb^+ $\text{Ga}_2\text{Sb}_{15}^-$ $\text{Ga}_3\text{Sb}_{14}^-$ $\text{Ga}_4\text{Sb}_{13}^-$ Ga_6Sb_5^- Ga_7Sb_4^-	$\text{GaTe}^{+/-}$ $\text{GaTe}_2^{+/-}$ GaTe_3^- GaTe_4^- Ga_2Te^+ Ga_2Te_2^-	SbTe^+ Sb_2Te^+ Sb_3Te^+ $\text{SbTe}_2^{+/-}$ SbTe_3^-	GaSbTe^+
F			$\text{Te}^{+/-}$ $\text{Te}_2^{+/-}$ $\text{Te}_3^{+/-}$ $\text{Te}_4^{+/-}$		$\text{GaTe}^{+/-}$ $\text{GaTe}_2^{+/-}$ $\text{GaTe}_3^{+/-}$ GaTe_4^- Ga_2Te^+ Ga_2Te_2^+		

Remarks: The symbols $^+$ and $^-$ mean that clusters were observed in the positive and negative ion modes, respectively, whereas $^{+/-}$ means that clusters were detected in both the positive and negative ion modes. The clusters highlighted in ‘**BOLD**’ are the most intensive peaks of binary and ternary clusters. Some minor (low-intensity) peaks of “oxidized and hydrogenated or oxidized/hydrogenated” clusters observed are not mentioned here.

TABLE S1 (B) Overview of $\text{Ga}_x\text{Sb}_y\text{Te}_z$ clusters formed via LA from individual Ga-Sb-Te thin films (**G-K**) in both positive and negative ion modes

Thin film	Ga_x	Sb_y	Te_z	Ga_xSb_y	Ga_xTe_z	Sb_yTe_z	$\text{Ga}_x\text{Sb}_y\text{Te}_z$
G	Ga_2^+	Sb^+	Te^-		GaTe^-	$\text{SbTe}^{+/-}$	
		Sb_2^-	Te_2^-		GaTe_2^-	Sb_2Te^-	
		Sb_3^-				SbTe_2^-	
		Sb_4^+					
H	Ga_2^+	Sb^+	Te^-		GaTe^-	$\text{SbTe}^{+/-}$	GaSbTe^+
			Te_2^-		GaTe_2^-	Sb_2Te^+	GaSbTe_2^-
	Ga_3^-				GaTe_3^-	Sb_3Te^+	$\text{Ga}_2\text{SbTe}_2^-$
					Ga_2Te^+	SbTe_2^-	
					Ga_2Te_2^-	SbTe_3^-	
						Sb_2Te_2^-	
I	Ga_2^+		$\text{Te}^{+/-}$		GaTe^-	SbTe^+	GaSbTe^+
			Te_2^-		GaTe_2^-	Sb_2Te^+	GaSbTe_2^-
					GaTe_3^-	Sb_3Te^+	GaSbTe_3^-
					Ga_2Te^-	SbTe_2^-	
					Ga_2Te_2^-	SbTe_3^-	
J	Ga_2^+	Sb^+	$\text{Te}^{+/-}$		$\text{GaTe}^{+/-}$	SbTe^+	GaSbTe^+
			$\text{Te}_2^{+/-}$		$\text{GaTe}_2^{+/-}$	Sb_2Te^+	GaSbTe_2^+
					$\text{GaTe}_3^{+/-}$	Sb_3Te^+	GaSb_2Te^+
					Ga_2Te^+	$\text{SbTe}_2^{+/-}$	$\text{GaSb}_2\text{Te}_2^+$
					$\text{Ga}_2\text{Te}_2^{+/-}$	SbTe_3^-	
					Ga_3Te^+		
					Ga_3Te_2^+		
K			$\text{Te}^{+/-}$				
			$\text{Te}_2^{+/-}$				
			$\text{Te}_3^{+/-}$				
			$\text{Te}_4^{+/-}$				
			$\text{Te}_5^{+/-}$				

Remarks: The symbols $^+$ and $^-$ mean that clusters were observed in the positive and negative ion modes, respectively, whereas $^{+/-}$ means that clusters were detected in both the positive and negative ion modes. The clusters highlighted in ‘**BOLD**’ are the most intensive peaks of binary and ternary clusters. Some minor (low-intensity) peaks of “oxidized and hydrogenated or oxidized/hydrogenated” clusters observed are not mentioned here.