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Tuning Crystallization Pathways in Ge-Rich GST Alloys: The Influence of Nitrogen and Hydrogen. Supplément

Jacopo Remondina, Eloïse Rahier, Minh Anh Luong, Nicolas Ratel-Ramond, Sijia Ran, David Grosso, Cristian Mocuta, Alain Portavoce, Yannick Le Friec, Daniel Benoit, Elisa Petroni, Alain Claverie, Magali Putero

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Supporting Information

Tuning Crystallization Pathways in Ge-rich GST Alloys: The Influence of Nitrogen and Hydrogen

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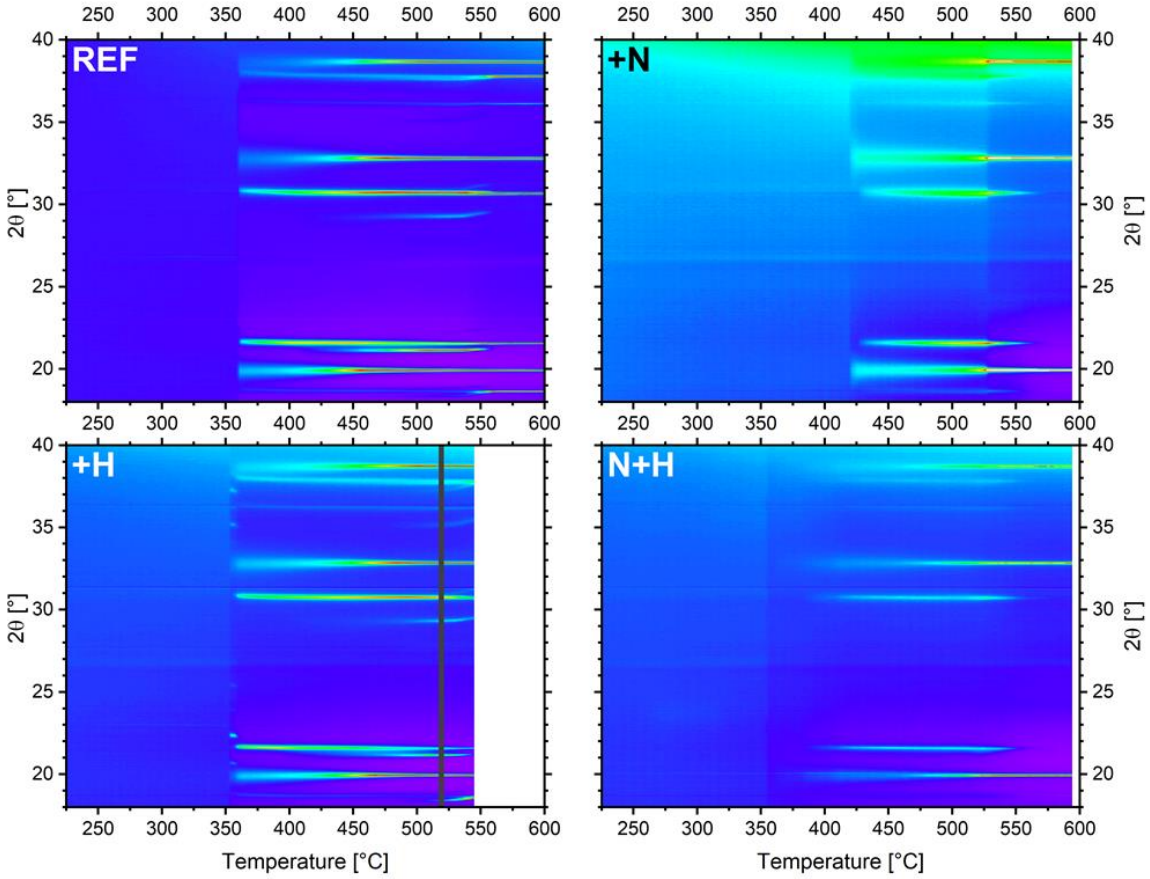


Figure S1. Contour plots of the full 2θ range XRD patterns (λ = 1.13 Å) recorded during *in situ* ramp annealing (2°C/min) on REF (top-left), GGST_N (top-right), GGST_H (bottom-left) and GGST_NH (bottom-right) samples. The vertical black line in GGST_H (bottom-left panel) corresponds to a beam shutdown.

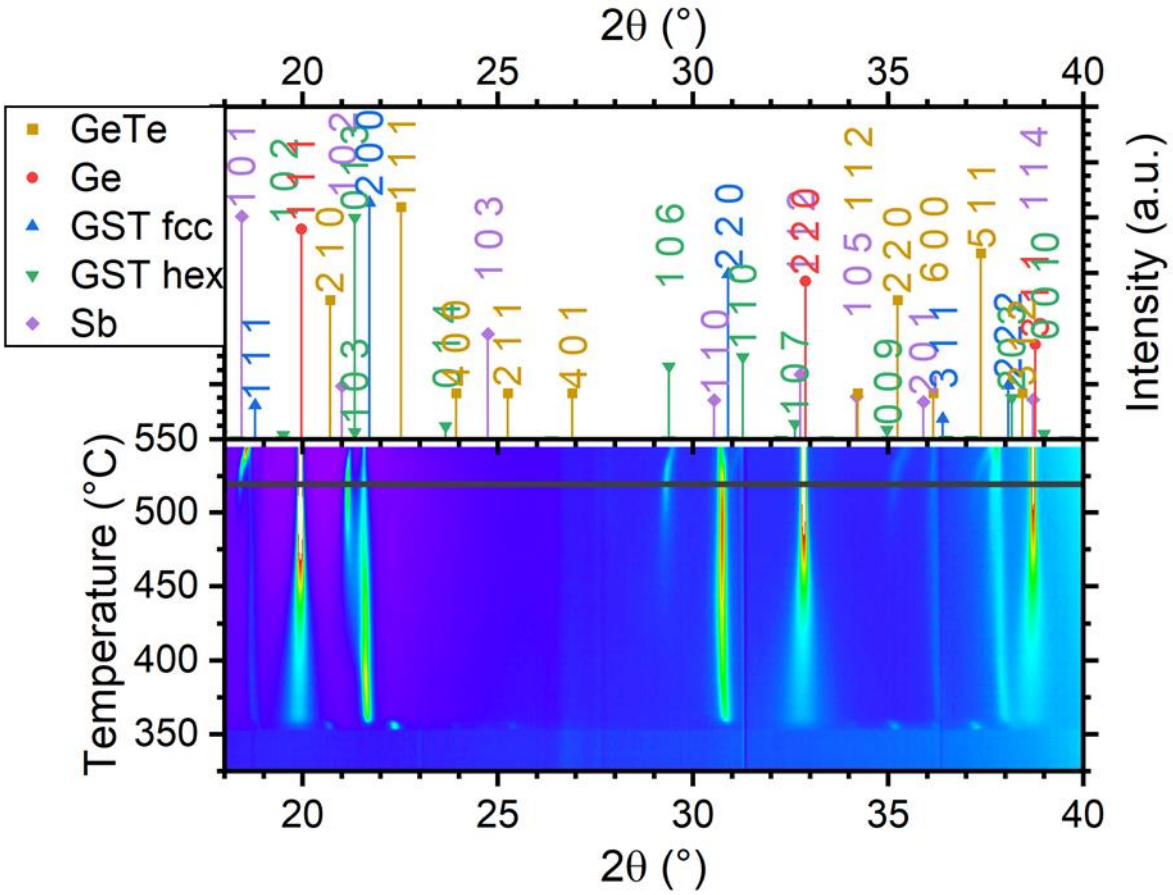


Figure S2. Peak attribution (top) for all the detected XRD peaks during the annealing of H-doped sample (the other samples contain a subset of the phases detected on this one). The labels above the dots indicate the Miller index of the peak, while the colors and the symbols shape the phase (black squares = GeTe Pnma; red circles = cubic Ge; blue up arrow = GST fcc; green down arrow = GST hex; purple diamond = Sb hex). (bottom): Contour plots of the full 2θ range XRD patterns (λ = 1.13 Å) recorded during in situ ramp annealing (2°C/min) on GGST_H sample, already shown in Fig. S1.

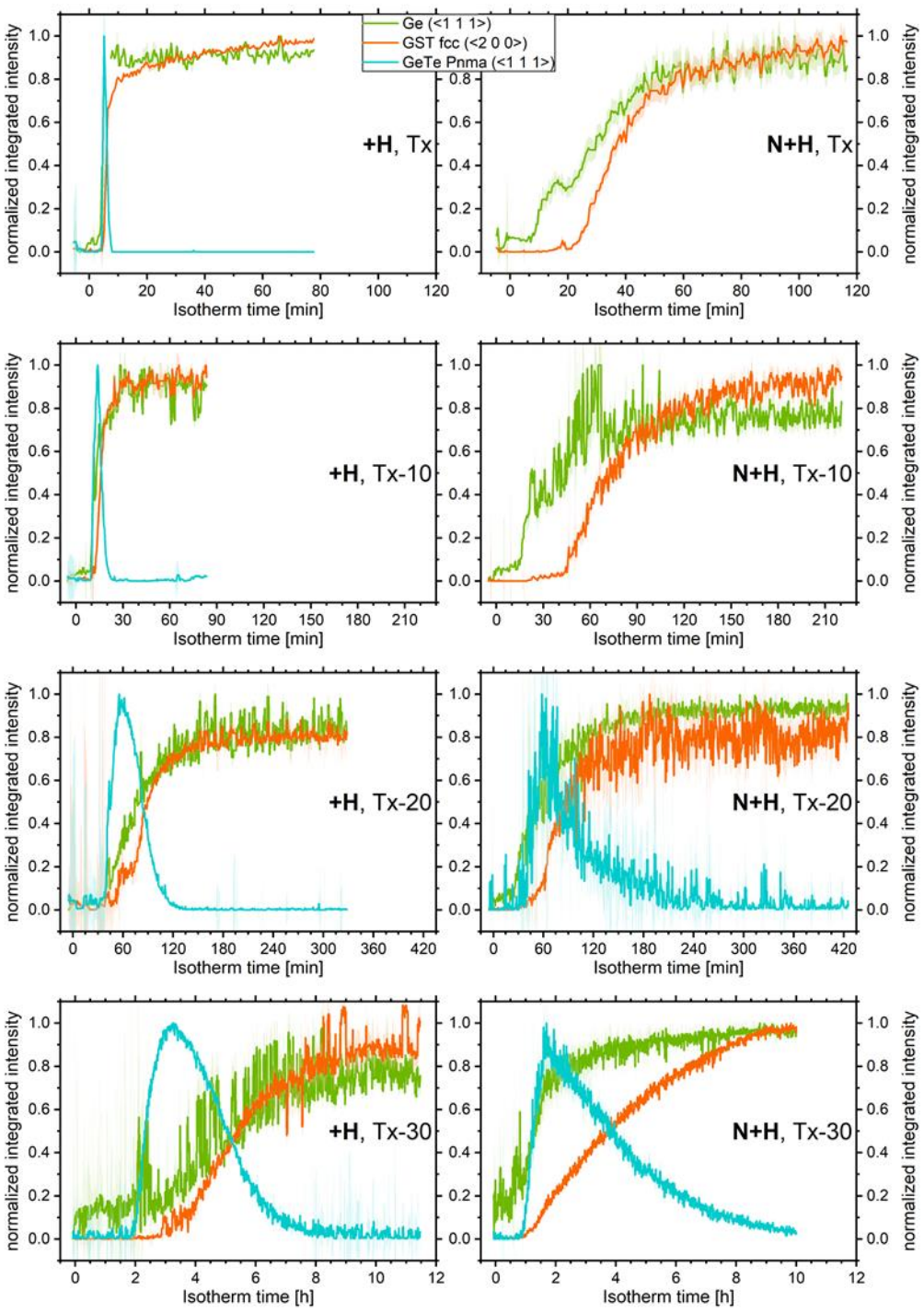


Figure S3. Normalized XRD integrated intensities collected on H-doped (left) and H+N doped (right) samples at Tx_{Ge} (top row), Tx_{Ge}-10 (second row), Tx_{Ge}-20 (third row) and Tx_{Ge}-30 (bottom row) (Tx_{Ge} denote the crystallization temperature of Ge in each sample) for the Ge (1 1 1) (green), GST cubic (2 0 0) (orange) and GeTe Pnma (cyan) reflections**.** The shaded curves correspond to error bars.

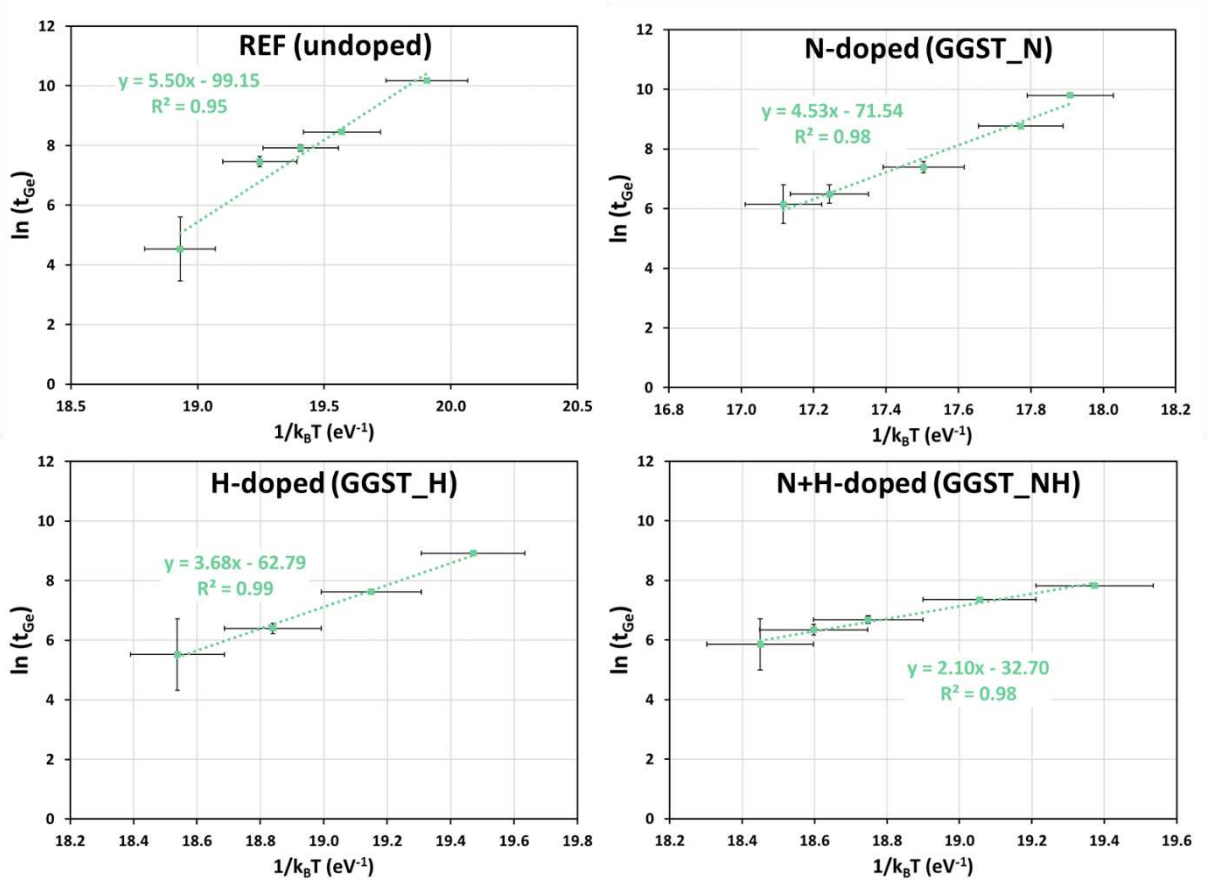


Figure S4. Variations of the Ge incubation time $\ln(t_{Ge})$ as a function of the inverse of temperature for undoped and doped GGST layers, for all isothermal annealing at different temperatures below Tx_{Ge}. The slope of the linear fit of these datasets allows to determine the activation energy (E_a) for the onset of crystallization.

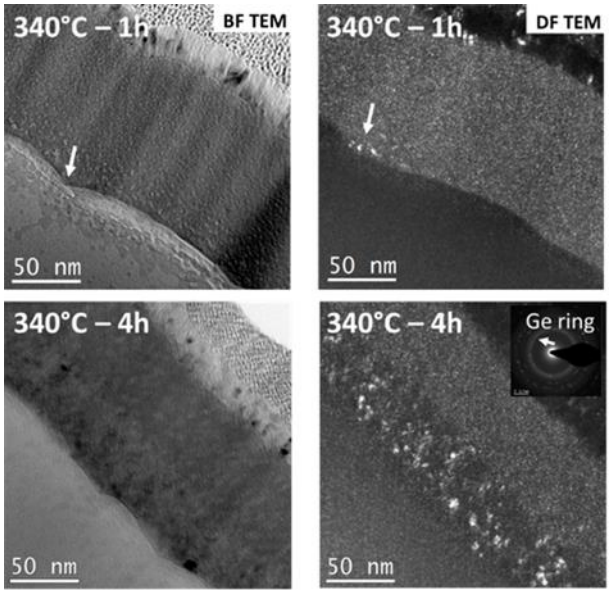


Figure S5. Left: TEM images (BF on the left, DF on the right) collected on the (N+H)-doped sample (GGST_NH) during *in situ* isothermal annealing at 340°C during 1 hour (top) and 4 hours (bottom). The top images show the onset of Ge crystallization, that nucleates at the GGST/SiN interface; the bottom images show that Ge first crystallizes in the bottom part of the layer.

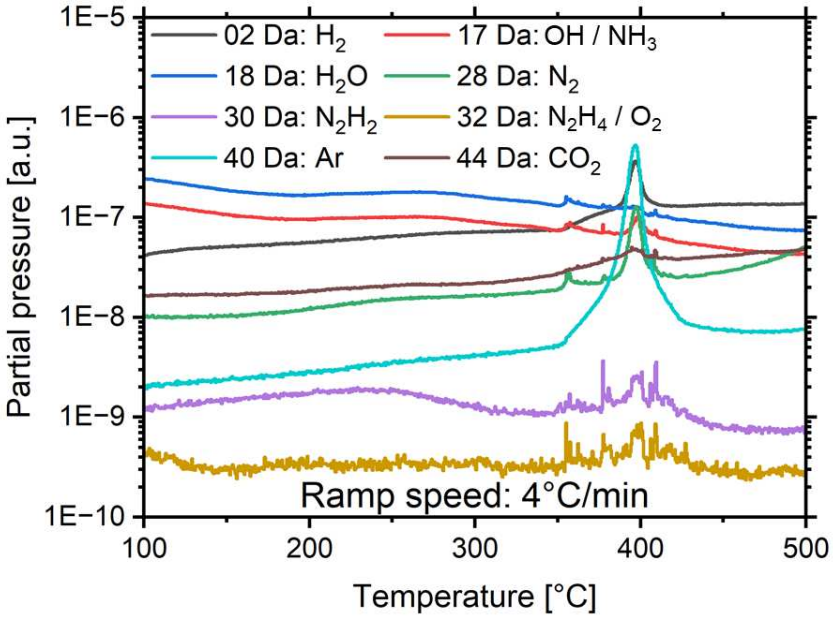


Figure S6. Thermal Desorption Spectroscopy experiments performed on H+N sample. The legend shows the selected mass, in Dalton/Atomic Mass Units, for each trend. The mass has been chosen to record the partial pressure of the H_2^+ ions (2 Da, black line), OH^+ ions (17 Da, red line, corresponding also to NH_3^+ ion), H_2O^+ ions (18 Da, blue line), N_2^+ ions (28 Da, green), $N_2H_2^+$ ions (30 Da, purple line), $N_2H_4^+$ ions (32 Da, yellow line, corresponding also to the O_2^+ ion), Ar^+ ion (40 Da, cyan line), CO_2^+ ion (44 Da, brown line).

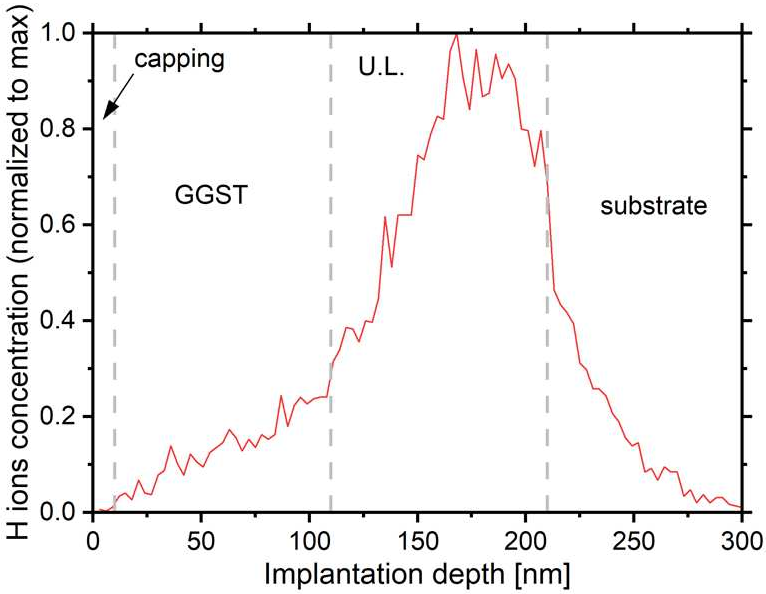


Figure S7. TRIM simulation of H implantation using an accelerating energy of 18 keV. Dashed vertical lines denotes the interfaces between the capping layer (20 nm of TiN), the GGST (100 nm) layer, the underlayer (100 nm-thick SiN), and the Si substrate.

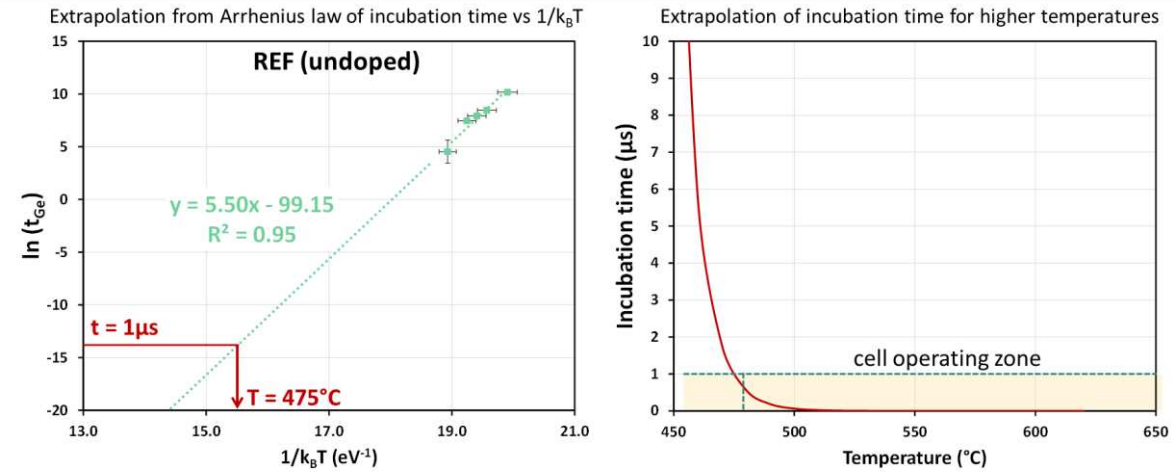


Figure S8. Example of extrapolation for sample REF (undoped GGST). Left: extrapolation from Arrhenius law (see Fig. S4) of incubation time as a function of $1/k_BT$. Right: Extrapolation of incubation time for higher temperatures with equation $\boldsymbol{t = exp\left(\frac{5.5}{k_BT} - 99.15\right)}$, showing that when the temperature exceeds 475°C in the case of undoped GGST, Ge crystallization would be triggered in less than 1 µs, as it happens in real GGST-based PCRAM memory cell.