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Supplementary Information

Modulation of GeSe, As_2Se_3 Motifs to Optimise GeAsSe OTS Performance and Its Mechanism

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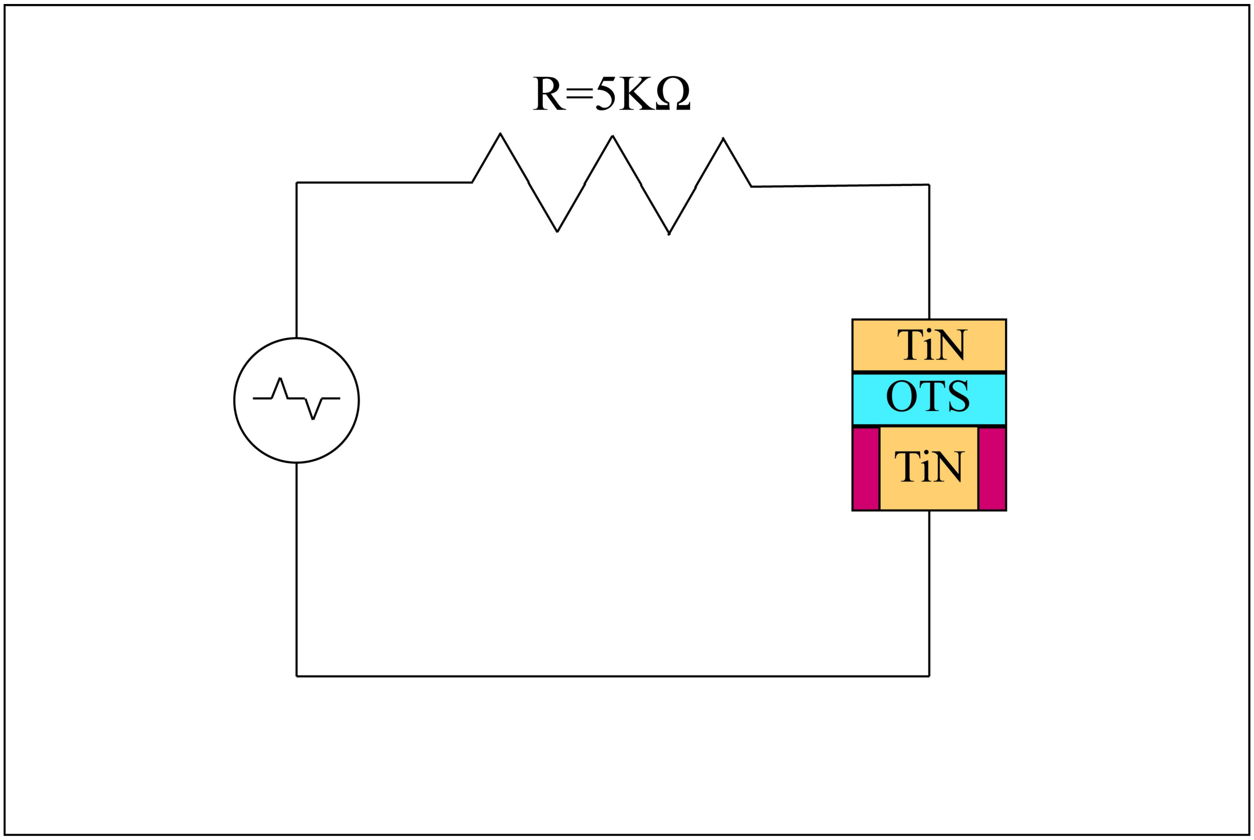

Fig.S1 Circuit diagram for DC testing. To prevent device damage from excessive current, a 5 KΩ resistor has been integrated into the test circuit. Test results are obtained by acquiring signals from both side of the device.

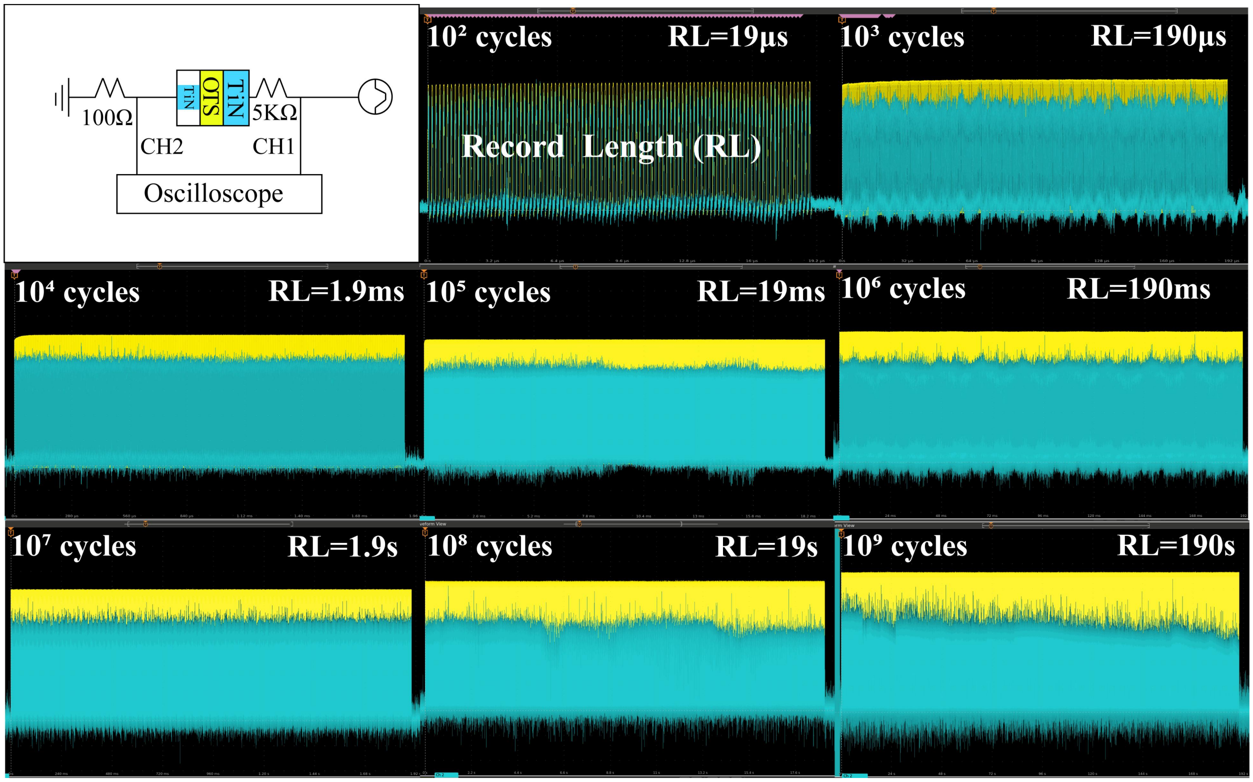

Fig.S2 Endurance of unannealed As_2Se_3 40 at.% GAS devices. The endurance was measured using 2.5V and 3.5V square pulses with 20 ns rising and falling edges, 100 ns pulse width and 50 ns interval. The dynamic response of the switch under a given number of square pulses is measured. The yellow curve corresponds to the applied voltage pulse and the blue curve corresponds to the measured dynamic response of the resistor in series with the device. The on-currents can be obtained from the dynamic response curve. Clearly the devices can be successfully turned on and off with each pulse. After a certain number of square pulse runs, the DC I-V curves of these devices were obtained, from which the off-currents were obtained, as shown in **Fig.1d**. In this case, the As_2Se_3 40at. % device can work well for up to 10^9 cycles of pulse operation.

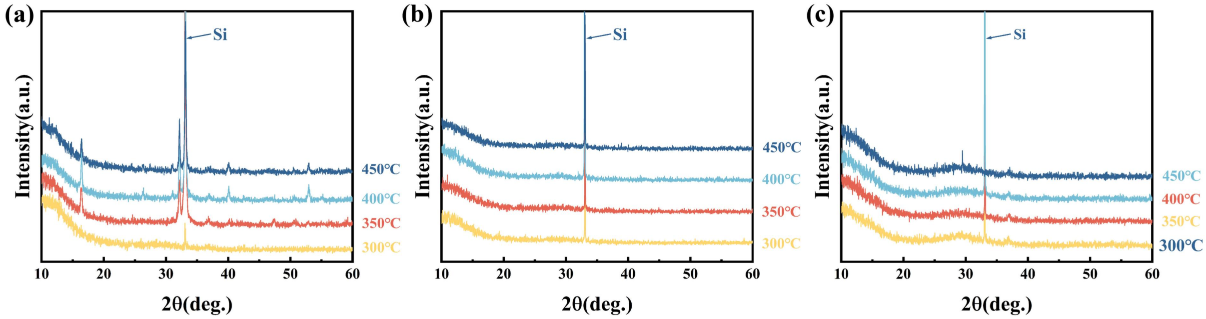

Fig.S3 X-ray diffraction (XRD) results of (a) GeSe, (b) $Ge_{26}As_{15}Se_{59}$, (c) $Ge_{18.5}As_{18}Se_{63.5}$ films at different temperatures. Fig.S3 a represents the XRD results of GeSe samples annealed at 300℃-450℃ for 3minutes. Clearly there are crytallization peaks at 16° and 32° at 350℃, indicating that the crystallization temperature of amorphous GeSe is between 300℃ and 350℃.

Fig.S3 b and **Fig.S3 c** represent XRD results of GeAsSe samples processed at 300, 350, 400 and 450℃ for 3 minutes. The results revealed that the GeAsSe films did not crystallize, thereby substantiating the efficacy of our methodology employed in enhancing the thermal stability of the material.

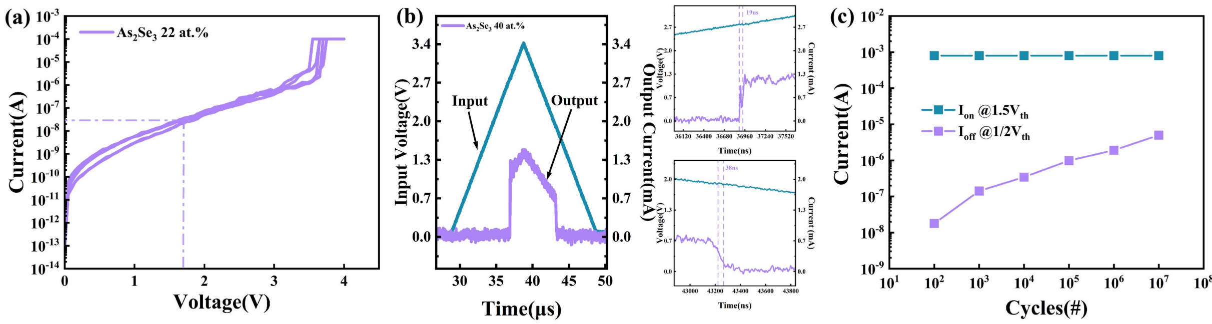

Fig.S4 Electrical performance of As_2Se_3 22 at.% devices after annealing. Fig.S4 a depicts the direct current (DC) test outcomes obtained under the circuit configuration depicted in **Figure.S1**, along with the device's leakage current. Following annealing, the device exhibited switching speeds of 19 ns and 38 ns, respectively (**Fig.S4 b**). Additionally, the endurance test demonstrated that the device's cycle count increased and I_{off} decreased after annealing, reaching 10^7 cycles (**Fig.S4 c**).

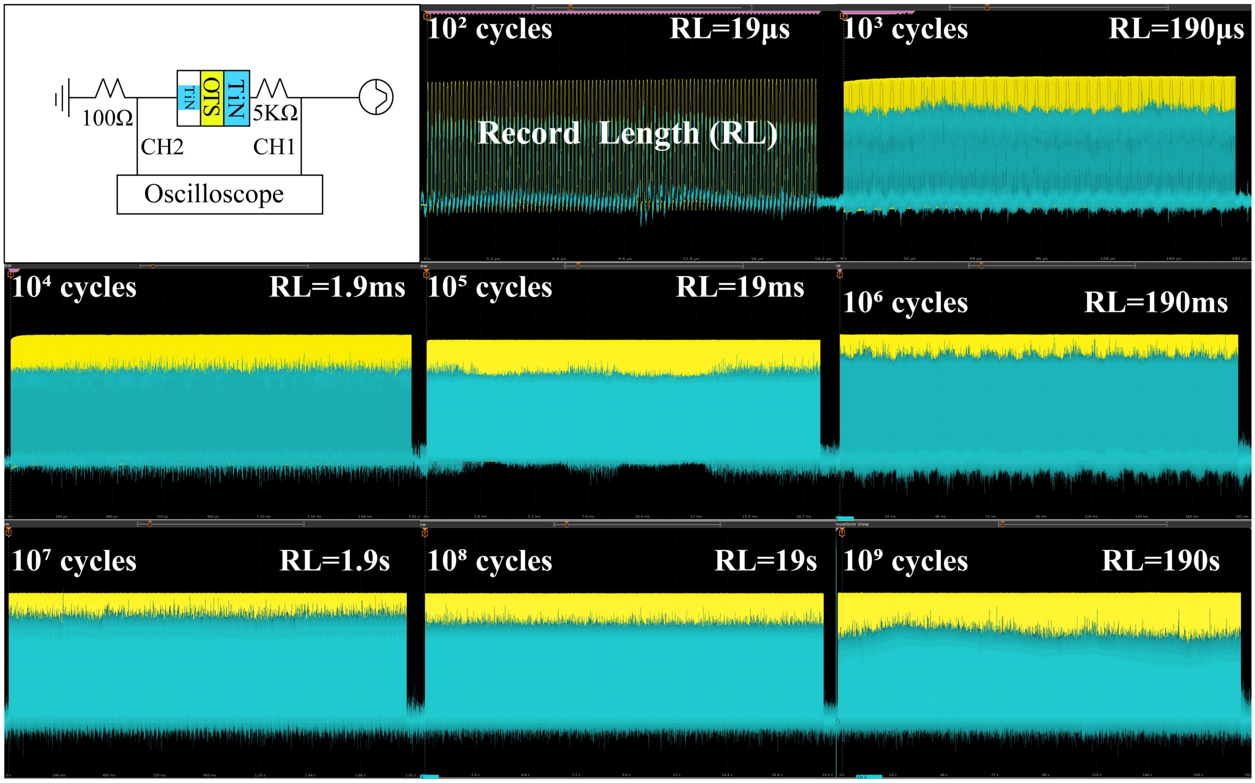

Fig.S5 Endurance of annealed As_2Se_3 40 at.% GAS devices. The endurance was measured using 2.5V and 3.5V square pulses with 20 ns rising and falling edges, 100 ns pulse width and 50 ns interval. As_2Se_3 40at. % device can successfully turned on and off for over 10^9 cycles, as shown in **Fig.2d**.