**Current-Induced Crystallisation in Heusler Alloy Films**

**for Memory Potentiation in Neuromorphic Computation**

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Figure S1 shows the development of giant magnetoresistive (GMR) behaviour by an applied current of 50 µA for 1 s up to 45 times. The noise decreases with increasing the current application, proving the Co2FeAl0.5Si0.5 (CFAS) Heusler-alloy films in the GMR pillar device. Similar behaviour has been observed in over 12 devices.

(a) (b)

***R*/*R*min (%)**

(c)  (d)

(e) (f)

**Figure S1: GMR curves of a CFAS/Ag/CFAS GMR device with the diameter of 150 nm × 100 nm.** | Normalised GMR curves measured under an applied field of ±1 kOe (a) before and after the current crystallisation by an applied current of 50 µA for 1 s (b) once, (c) 3, (d) 10, (e) 20 and (f) 45 times. Red and blue curves correspond to the field sweep from negative to positive and *vice versa*.