RESEARCH ARTICLE | SEPTEMBER 26 2023

Enhanced resistive switching characteristics of conductive bridging memory device by a Co–Cu alloy electrode ⊘

Calvin Xiu Xian Lee ⁽[®]); Putu Andhita Dananjaya ^{(®}); Mun Yin Chee ^{(®}); Han Yin Poh ^{(®}); Funan Tan ^{(®}); Jia Rui Thong ^{(®}); Lingli Liu ^{(®}); Gerard Joseph Lim ^{(®}); Yuanmin Du ^{(®}); Juan Boon Tan ^{(®}); Wen Siang Lew **≤** ^{(®})



Appl. Phys. Lett. 123, 133504 (2023) https://doi.org/10.1063/5.0160380



Articles You May Be Interested In

A functional integral formalism for quantum spin systems

J. Math. Phys. (July 2008)

Modes selection in polymer mixtures undergoing phase separation by photochemical reactions *Chaos* (June 1999)

Spreading of a surfactant monolayer on a thin liquid film: Onset and evolution of digitated structures *Chaos* (March 1999)

27 September 2023 08:37:32





Enhanced resistive switching characteristics of conductive bridging memory device by a Co-Cu alloy electrode

Cite as: Appl. Phys. Lett. **123**, 133504 (2023); doi: 10.1063/5.0160380 Submitted: 1 June 2023 · Accepted: 8 September 2023 · Published Online: 26 September 2023

Calvin Xiu Xian Lee,^{1,2} D Putu Andhita Dananjaya,¹ Mun Yin Chee,¹ Han Yin Poh,^{1,2} Funan Tan,¹ C I Jia Rui Thong,^{1,2} Lingli Liu,¹ C Gerard Joseph Lim,¹ Rui Thong,^{1,2} Lingli Liu,¹ C Gerard Joseph Lim,¹ Rui Yuanmin Du,¹ D Juan Boon Tan,² And Wen Siang Lew^{1,a)}

AFFILIATIONS

¹School of Physical and Mathematical Sciences, Nanyang Technological University, 21 Nanyang Link, Singapore 637371 ²GlobalFoundries, 60 Woodlands Industrial Park D Street 2, Singapore 738406

^{a)}Author to whom correspondence should be addressed: wensiang@ntu.edu.sg

ABSTRACT

One of the main challenges in the development of conductive bridging random access memory (CBRAM) is the large stochastic nature of ion movement that ultimately leads to large parameter variability. In this study, the resistive switching variability of CBRAM devices is significantly improved by employing Co–Cu alloy as the active electrode. By comparing with $Pt/Ta_2O_5/Co$ devices, the $Co_{70}Cu_{30}$ alloy exhibited lower forming voltage (<2 V), lower SET voltage (<0.70 V), and faster response time (~70 ns). The filament stability indicated by the distribution of SET/RESET voltage and high resistance state/low resistance state variation was significantly improved. Our experimental results suggest the formation of Co filaments, and the proposed mechanism is governed by the galvanic effect. In addition, a comparison between $Co_{70}Cu_{30}$ and $Co_{30}Cu_{70}$ alloys highlights that the relative proportion between Co and Cu plays an essential role in the device performance. A physical model based on different electrochemical activities of the alloys has been proposed to explain the filament formation and the improved switching uniformity in the $Co_{70}Cu_{30}$ alloy. This study not only develops a CBRAM with enhanced performance but also advances the implementation of suitable alloy systems for the application of such devices.

Published under an exclusive license by AIP Publishing. https://doi.org/10.1063/5.0160380

With a simple metal-insulator-metal (MIM) structure, resistive random access memory (ReRAM) has attracted great attention in recent years, with the strong technological demand to develop next-generation memory devices,1-4 reconfigurable logic circuits,5,6 and neuromorphic computing applications.⁷⁻⁹ Among the various types of resistive switching devices, a typical conductive bridge random access memory (CBRAM) consists of an electrochemically active electrode (AE) and an inert electrode (IE), separated by a thin solid electrolyte layer.^{10,11} The reversible resistive switching mechanism of CBRAM is based on electrochemical metallization (ECM), which involves the nucleation and migration of metal ions (e.g., Ag^{12,13} or Cu¹⁴⁻¹⁶) through the solid electrolyte under an external electric field to form conductive filaments.^{17,18} Due to its high k-dielectric constant and compatibility with the complementary metal oxide semiconductor (CMOS) fabrication process, Ta2O5 is considered as one of the most promising solid electrolytes for CBRAM.^{19–21} Among the various pure metal-based electrochemically active electrodes, Cu is a preferred material due to its abundance, low cost, and relatively low solubility with the electrolyte layer.^{22–24} More

recently, Co-based CBRAMs have been reported, with the advantages in low operating current, excellent retention, and CMOS-compatible fabrication capability.^{25–27} Nonetheless, the pure metal-based CBRAM still exhibits large switching variability and performance degradation due to its large stochasticity of the ion movement, which inhibits further technological development of this memory technology.^{23,28,29} The stochastic nature of the conductive filament formation and dissolution in CBRAM plays a significant role in the device performance.^{30,31} In comparison, electrode alloying provides a new degree of freedom for controlling electrochemical activity, nucleation sites, ion migration, and filament formation.^{32–35} By employing an Au–Ag alloy electrode, the forming voltage, resistive switching stability, and speed were reported to be significantly enhanced due to the galvanic effect between Au and Ag.33 ³⁵ Using the Ag-Cu alloy electrode, the switching performance was also improved through the formation of Cu filaments, with the composition identified by transmission electron microscopy (TEM).^{36,37} However, there has not yet been an investigation of how the Co-Cu alloy influences the resistive switching properties of CBRAM.

In this work, we investigated the resistive switching behavior of the MIM device structures consisting of $Pt/Ta_2O_5/AE$, where AE is varied with different active electrode materials, namely, Co, Cu, $Co_{70}Cu_{30}$, and $Co_{30}Cu_{70}$. The $Co_{70}Cu_{30}$ alloy electrode CBRAM exhibits reduced electroforming voltage along with smaller SET/ RESET voltages variation, improved resistance states uniformity, and faster response time. The morphological analysis of the Co–Cu alloy surface revealed that the resistive switching improvement can be attributed to the preferential oxidation of Co within the Co–Cu alloy system, also known as the galvanic effect. A physical model based on the galvanic effect is proposed to explain the origin of the different resistive switching performances.

The fabrication of the devices begins with inert electrode patterning onto a SiO₂ substrate via UV lithography. Following a Ti (2 nm) adhesion layer deposition, Pt (10 nm) inert electrode was deposited via direct current (DC) magnetron sputtering with 20 sccm Ar gas flow rate at a working pressure of 2 mTorr and sputtering power 50 W. The patterned Pt inert electrode is formed via a liftoff process. Subsequently, the active electrode patterning was performed, followed by a Ta2O5 (7 nm) electrolyte layer and Co-Cu alloy (50 nm) active electrode deposition. The electrolyte layer is deposited via radio frequency (RF) magnetron sputtering with 20 sccm Ar gas flow rate at a working pressure of 2 mTorr, utilizing a 2 in. ceramic target of Ta₂O₅, while the Co-Cu alloys are deposited via DC magnetron sputtering with 20 sccm Ar gas flow rate at a working pressure of 2 mTorr. The composition of the Co-Cu alloys was obtained by co-sputtering the 2 in. Co and Cu targets at varying power levels (Co70Cu30/144 W: 28 W and Co30Cu70/61 W: 67 W) with a fixed deposition time of 400 s to achieve the desired deposition rates. A Pt capping layer (5 nm) was deposited to protect the device. The chamber atmosphere during all the sputtering processes of all the films consisted of 2 mTorr Ar gas and the target-to-substrate distance was consistently maintained at 200 mm. The fabrication process is finalized by performing a liftoff process to obtain the Pt/Ta2O5/Co-Cu alloy/Pt CBRAM structure, as illustrated in Fig. 1(a).

Figure 1(b) shows the optical image of the fabricated CBRAM device, where a $10 \times 10 \ \mu m^2$ cell size of the experimental devices is formed. X-ray photoelectron spectroscopy (XPS) characterization determined the nominal mole or atomic fraction (at. %) of the Co–Cu alloy electrodes as Co: Cu = 70%: 30% for the Co-rich alloy, and Co: Cu = 30%: 70% for the Cu-rich alloy. The XPS spectra are presented in the supplementary material as Fig. S1, where Kratos Axis Supra XPS system was used for analysis. To perform the electrical characterization of the Pt/Ta₂O₅/Co–Cu alloy CBRAM devices, a Keithley 4200A semiconductor parameter analyzer was utilized by grounding the inert electrode and biasing the active electrode. In addition, Pt/Ta₂O₅/Ag–Cu alloy CBRAMs are also fabricated as a reference.

Figures 1(c)–1(f) show DC–voltage (I–V) curves of the CBRAM devices, with the compliance current (I_{CC}) set at 1 mA. The resolution of the voltage sweeping is 5 mV per step. All the CBRAM devices in the pristine state show an initial resistance value of $\sim 10^8 \Omega$ with a 0.2 V read voltage, indicating a good consistency between the Ta₂O₅ films. An electroforming process (0 V \rightarrow +3 V) is required before the device can be reversibly switched between a high resistance state (HRS) and a low resistance state (LRS). A bipolar switching characteristic has been observed for all the devices. When a positive set voltage (V_{SET}) is applied, the device switches from HRS to LRS, and when a

negative reset voltage (V_{RESET}) is applied, the device switches from LRS to HRS. Among the pure metal-based CBRAM devices, a higher electroforming voltage ($V_{Forming}$) is observed for the Co-based device [Fig. 1(c)], compared to that of the Cu-based device [Fig. 1(d)]. This can be ascribed to the different diffusion capability of Cu and Co ions into the Ta₂O₅ insulator matrix, to form conductive bridges between the active and inert electrodes. For the following resistive switching operations (100 switching cycles), although relative smaller V_{SET} and V_{RESET} values have been observed for the Cu-based device [Fig. 1(d)], a wider switching distribution is observed, which indicates a great disadvantage in contrast to that of the Co-based device [Fig. 1(c)]. The distribution of V_{SET} , V_{RESET} , and $V_{Forming}$ for different devices is shown in Fig. S2.

By comparing with the pure metal-based devices, an enhancement of the device performance has been observed, when a $Co_{70}Cu_{30}$ alloy is selected as the active electrode material. Figure 1(e) shows I–V curves of a Pt/Ta₂O₅/Co₇₀Cu₃₀ device, with $V_{Forming}$ at ~2 V, which is significantly reduced compared to that of a Co-based device with $V_{Forming}$ at ~2.6 V [Fig. 1(c)]. More interestingly, an even more stable switching characteristic is shown for the $Co_{70}Cu_{30}$ alloy-based device, with V_{SET} and V_{RESET} varying in a much smaller range. For comparison on the resistive switching performance of the Co–Cu alloy, another device with $Co_{30}Cu_{70}$ as the AE has also been fabricated, with the I–V curves shown in Fig. 1(f). A decreasing of the Co percentage results in a further reduction of the $V_{Forming}$ [Fig. S2(d)]. However, a degradation of the device performance is observed where both V_{SET} and V_{RESET} shift into a wider range from cycle to cycle, and an unstable switching characteristic is presented.

Figures 2(a) and 2(b) show the cumulative probability distribution of the V_{SET}/V_{RESET} and the resistance states during 100 continuous DC switching cycles for the CBRAM devices shown in Figs. 1(c)-1(f). A boxplot of the resistance of the HRS and LRS of each CBRAM device is shown in Fig. S3. For the Co₇₀Cu₃₀ alloy-based device, both V_{SET} and V_{RESET} and the resistance states (HRS and LRS) show narrow distributions, indicating significant enhancements of the resistive switching. The V_{RESET} and V_{SET} ranges are -0.94 to -0.51 V and 0.44 to 0.88 V, respectively, which are much narrower than those of the other devices. The average/standard deviation (SD) of the operating voltages is -0.65 V/0.07 V (V_{RESET}) and 0.68 V/0.07 V (V_{SET}). With the Cu-based device as an example, V_{RESET} ranges from -0.99 to -0.18 V (SD, 0.19 V), and V_{SET} ranges from 0.18 to 1.22 V (SD, 0.23 V). The SD of the V_{RESET} and V_{SET} is reduced by 69.9% and 63.2%, respectively, for the Co70Cu30 alloy-based device, in comparison with that of Cu. Similarly, for the HRS/LRS distribution [Fig. 2(b)], a much narrower distribution is also observed for the $Co_{70}Cu_{30}$ alloy-based device. Although the other CBRAM devices typically show a larger memory window (or resistance switching ratio), the large variation of the resistance values (HRS) had a great disadvantage of these devices for memory storage applications. Among the alloy electrode CBRAMs, the resistive switching properties of Ag-Cu alloy-based CBRAM have been extensively investigated, making it suitable as a reference stack for comparison.^{7,36-38} Between the fabricated Ag-Cu alloy-based CBRAM and Co-Cu alloy-based CBRAM, the Co₇₀Cu₃₀ alloy also shows the advantage on the uniformity of switching operations (see Figs. S4 and S5 for more details). Qiao et al.³⁶ reported that Cu filaments are formed in the Ag-Cu alloy-based CBRAMs, which is attributed to the galvanic effect. The nature of the conductive filaments





FIG. 1. (a) Schematic illustration of the CBRAM device structure where the electrolyte layer (Ta₂O₅) is sandwiched between the active electrode (AE) and inert electrode (IE). (b) Optical image of a fabricated Co–Cu alloy-based CBRAM device. I–V measurements of CBRAM devices with different active electrode: (c) Pt/Ta₂O₅/Co, (d) Pt/Ta₂O₅/Cu, (e) Pt/Ta₂O₅/Co₃₀Cu₇₀. The plots represent 101 cycles of sweeping loops, with the red and blue curves representing the typical SET and RESET operations, and the black curve representing the initial forming and the first RESET after the forming operations, respectively.

FIG. 2. Cumulative probability of (a) V_{SET} and V_{RESET} and (b) HRS/LRS over 100 continuous DC sweeping cycles for CBRAM devices with different active electrodes, Co, Cu, Co₇₀Cu₃₀, and Co₃₀Cu₇₀, respectively. HRS—high resistance state, LRS—low resistance state.



FIG. 3. (a) SET and (b) RESET response time characteristics of $Pt/Ta_2O_5/Co$ (orange), $Pt/Ta_2O_5/Cu$ (blue), $Pt/Ta_2O_5/Co_{70}Cu_{30}$ (black), and $Pt/Ta_2O_5/Co_{30}Cu_{70}$ (green) device. For SET and RESET responses, a 1.5 V/5 μ s pulse and a -1.6 V/10 μ s pulse are used, respectively. The red lines represent the input voltages. Inset provides the enlarged region of the current response for different active electrodes.

formed across the device is essential in the resistive switching performance for a CBRAM device.

Figure 3 shows the typical SET/RESET response time measurements of the CBRAM devices. READ pulses (0.2 V/10 μ s) are applied before and after each SET/RESET operation to indicate the resistance states. The response time is obtained from the delay between the response current and the input voltage. The schematic diagrams at the top of Figs. 3(a) and 3(b) illustrate the voltage pulses used for SET and RESET operations, respectively, while the lower portion of Figs. 3(a) and 3(b) reveals the current change of the Co-Cu alloy CBRAM. A comparison of the SET/RESET response time for different devices is listed in Table I. Under a SET voltage pulse of $1.5 \text{ V}/5 \mu \text{s}$, the SET response time is \sim 280 ns; under a RESET voltage pulse of -1.6 V/10 μ s, the RESET response time is ~620 ns. An improvement has also been observed for the Co70Cu30 alloy-based device, with the SET/ RESET response times significantly reduced by up to 75%/51% (~70 ns/~480 ns), which is much faster than the Cu-based device (Table I). While for the pure Co electrode-based device, much longer response time is observed. The faster SET/RESET response time renders the Co₇₀Cu₃₀ alloy-based CBRAM devices to be more competitive in terms of data processing and storage application.

To gain more insight into the filaments formed in the Co–Cu alloy-based CBRAM devices, the temperature dependent resistance measurement is carried out for Co, Cu, and Co–Cu alloy CBRAM devices (Fig. S6). With each device at LRS, the R-T plot can be fitted by

 $\ensuremath{\mathsf{TABLE}}$ I. Comparison of SET/RESET response time of Co, Cu, and Co–Cu alloy CBRAM.

Active electrode (AE)	Electrolyte	Inert electrode	SET response time (ns)	RESET response time (ns)
Со	Ta ₂ O ₅	Pt	130	920
Cu	Ta ₂ O ₅	Pt	280	620
Co30Cu20	Ta ₂ O ₅	Pt	220	410
Co ₇₀ Cu ₃₀	Ta ₂ O ₅	Pt	70	480

$$R(T) = R_0 [1 + \alpha (T - T_0)], \qquad (1)$$

where R_0 is the reference resistance at temperature of T_0 and α is the temperature coefficient of resistance. The fitting results show a positive coefficient of 7.37×10^{-4} and 7.17×10^{-4} K⁻¹ for Co and Co₇₀Cu₃₀ alloy CBRAM, respectively. This is consistent with the report by Yang et al.,³⁹ where a coefficient of 8.6×10^{-4} K⁻¹ is obtained for the Co nanofilament. As for the Cu and Co30Cu70 alloy CBRAM, the fitting results indicate a positive coefficient of 3.09×10^{-3} and 1.01×10^{-3} K⁻¹, respectively. Both coefficient values agree with the values reported in literatures where the temperature coefficient of Cu nanofilament ranges from 1.45×10^{-3} to 4.5×10^{-3} K⁻¹ (Refs. 40 and 41). In a Co-Cu alloy system, the standard electrode potential for Co²⁺/Co is -0.28 V, which is substantially smaller than that of Cu²⁺/Cu (0.34 V) and Co dissolve preferentially before Cu in the Co-Cu alloy.⁴² The galvanic effect occurs between Co and Cu clusters where Co atoms will be oxidized into Co²⁺ ions by Cu ions.^{43,44} Under the influence of an external electric field, the Co^{2+} ions migrate across the electrolyte, and Co filaments are formed.

In addition to the galvanic effect, the interface roughness between AE and the electrolyte also affects the filament formation process.^{45,46} Near the ridges of the metal/electrolyte interface, the electrical field is much stronger than the surrounding regions, resulting in an increased propensity for filament formation. The surface and interfacial roughness of the different AEs were investigated using atomic force microscopy (AFM) and x-ray reflectivity (XRR). Figure 4 shows the interfacial roughness of the Co, Cu, Co70Cu30, and Co30Cu70 films with Ta₂O₅ film, and the AFM measurements are shown in Fig. S7. The surface and interfacial roughness results are summarized in Table II. Among all the films scanned, Cu has the highest roughness, from which an abrupt interface may be formed between Cu and Ta2O5. For Co and Co-Cu alloys, a typically smaller roughness is observed, with Co₇₀Cu₃₀ the smoothest. The Co₇₀Cu₃₀ film has the smallest average (R_a), root mean square (R_q), and interfacial (σ) values, that is, 0.30, 0.38, and 0.216 nm, respectively, which are much smaller than that of Co ($R_a = 0.39 \text{ nm}$; $R_g = 0.49 \text{ nm}$; $\sigma = 0.373 \text{ nm}$). Nevertheless, even with a reduced roughness, the Co₇₀Cu₃₀ alloy-based device shows a smaller $V_{Forming}$ compared to the Co-based device [Figs. 1(d) and 1(e)].



FIG. 4. XRR measurement and fitting of (a) Pt/Ta₂O₅/Co, (b) Pt/Ta₂O₅/Cu, (c) Pt/Ta₂O₅/Co₇₀Cu₃₀, and (d) Pt/Ta₂O₅/Co₃₀Cu₇₀ for interfacial roughness between Ta₂O₅ and TE.

In other words, the galvanic effect leads to the observed decrease in $V_{\rm Forming}$ between $\rm Co_{70}Cu_{30}$ and Co-based devices.

While a lower $V_{Forming}$ is observed for the Co₃₀Cu₇₀ alloy-based device, a more stable and uniform switching is obtained for the Co₇₀Cu₃₀ alloy-based device. This difference can be explained by considering the corrosion potential that varies logarithmically with the atomic percentage of Cu. The corrosion potential can be expressed as

$$E_{corr} = k - b_c \log[S_{\theta}], \qquad (2)$$

where S_{θ} is the volume proportion and *k* and b_c are Tafel constants.⁴⁷ In a Co–Cu alloy system, due to the electrode potential difference between Co and Cu, as the Cu concentration increases, the corrosion rate of Co–Cu alloy increases. The acceleration of corrosion rate may lead to more nucleation sites; consequently, more filaments are formed. It has been reported that the randomness of filament

 TABLE II. Comparison of average, root mean square surface roughness, and interfacial roughness.

Active electrode (AE)	Average surface roughness, R _a (nm)	Root mean square surface roughness, R _q (nm)	Interfacial roughness, σ (nm)
Со	0.39	0.49	0.373
Cu	0.44	0.56	0.449
Co30Cu20	0.40	0.50	0.248
Co ₇₀ Cu ₃₀	0.30	0.38	0.216

formation typically has a poorer stability of switching performance.^{38,48} By comparing with a higher Cu concentration of the Co₃₀Cu₇₀ alloy CBRAM, fewer yet more robust Co filaments tend to be formed for the Co₇₀Cu₃₀ alloy CBRAM as there are sufficient Co nucleation sites for Co filament formation. Based on this, a physical model illustrating the mechanism of Cu and Co filament formation is shown in Fig. 5. The higher corrosion reaction rate, coupled with increased roughness (see Table II), leads to the formation of a greater number of but less robust Cu filaments [Fig. 5(a), Co₃₀Cu₇₀], in contrast to that shown in Fig. 5(b) for the Co₇₀Cu₃₀ alloy-based device where Co filaments formation are present. The relative concentration between Co and Cu determines the corrosion rate and filament composition, which ultimately acts as an important role in the resistive switching characteristics. According to the proposed model, it is expected that optimizing the relative concentration of Cu in the Co-Cu alloy holds the potential to enhance the performance of the device.

In conclusion, the resistive switching characteristics of Co–Cu alloy electrode CBRAM devices have been investigated. By utilizing Co₇₀Cu₃₀ alloy as the active electrode, reduced $V_{Forming}$ (~2 V), smaller V_{SET}/V_{RESET} distribution, narrower resistance states (HRS and LRS) variation, and shorter SET/RESET response time (~70 ns/~480 ns) have been realized. These enhanced switching performances are attributed to the formation of Co conductive filaments through a galvanic effect, where the electrochemical oxidation of Co atoms takes precedence over Cu atoms due to their respective electrode potentials. Additionally, a comparative analysis involving Co₃₀Cu₇₀ alloy underscores the significance of appropriate alloy composition in



FIG. 5. Schematic diagrams of filament formation in different Co–Cu alloy-based CBRAM devices. (a) SET process of $Co_{30}Cu_{70}$ alloy CBRAM where the elemental composition of CFs are mainly Cu atoms due to the lack of Co atoms at the metal/ electrolyte interface. (b) SET process of $Co_{70}Cu_{30}$ alloy CBRAM where the elemental composition of the CFs is mainly Co atoms. (c) Exchange of electrons between Cu and Co due to the galvanic effect.

determining CBRAM performance. The proposed approach and underlying mechanism not only develop a CBRAM with enhanced performance but also suggest that optimizing the alloy composition based on corrosion potential can achieve more uniform switching and larger memory window characteristics. This technique hold promises as a strategy for developing high-performance memory devices for application in high-speed and lower-power electronics.

See the supplementary material for the results of XPS analysis of Co–Cu alloy films, the boxplot distribution of $V_{\rm SET}$ and $V_{\rm RESET}$ for size random devices with different Co–Cu alloy AEs, the HRS and LRS boxplot comparison for each device stack with different AEs, the I–V curves of Ag–Cu alloy-based devices, the boxplot distribution of $V_{\rm SET}$ and $V_{\rm RESET}$ for Ag–Cu alloy-based devices, the resistance-temperature plot of Co–Cu alloy-based devices, and the AFM analysis of Co–Cu alloy films.

This work was supported by a RIE2020 ASTAR AME IAF-ICP (Grant No. 11801E0030) and EDB-IPP: Economic Development Board–Industrial Postgraduate Program (Grant No. REQ0165097).

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Calvin Xiu Xian Lee: Conceptualization (equal); Data curation (equal); Investigation (equal); Methodology (equal); Visualization

(equal); Writing – original draft (equal). Putu Andhita Dananjaya: Conceptualization (equal); Investigation (supporting); Writing original draft (equal); Writing - review & editing (equal). Mun Yin Chee: Data curation (equal); Investigation (supporting); Writing review & editing (equal). Han Yin Poh: Data curation (equal); Writing review & editing (equal). Funan Tan: Investigation (equal); Visualization (equal); Writing - review & editing (equal). Jia Rui Thong: Data curation (supporting); Investigation (supporting); Resources (supporting). Lingli Liu: Data curation (supporting); Investigation (supporting). Gerard Joseph Lim: Writing - review & editing (supporting). Yuanmin Du: Investigation (equal); Methodology (supporting); Writing - review & editing (equal). Juan Boon Tan: Funding acquisition (equal); Supervision (equal); Writing - review & editing (equal). Wen Siang Lew: Conceptualization (equal); Funding acquisition (equal); Project administration (equal); Resources (equal); Supervision (equal); Writing - review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its supplementary material.

REFERENCES

- ¹K. H. Kim, S. Gaba, D. Wheeler, J. M. Cruz-Albrecht, T. Hussain, N. Srinivasa, and W. Lu, Nano Lett. **12**, 389 (2012).
- ²G. Zhu, W. Chen, D. Wang, H. Xie, Z. Zhao, P. Gao, J. Schutt-Aine, and W. Y. Yin, IEEE Trans. Electron Devices 66, 1747 (2019).
- ³A. Chen, Solid State Electron. **125**, 25 (2016).
- ⁴S. C. W. Chow, P. A. Dananjaya, J. M. Ang, D. J. J. Loy, J. R. Thong, S. W. Hoo, E. H. Toh, and W. S. Lew, Appl. Surf. Sci. 608, 155233 (2023).
- ⁵J. J. Yang, D. B. Strukov, and D. R. Stewart, Nat. Nanotechnol. 8, 13 (2013).
- ⁶Q. Xia, W. Robinett, M. W. Cumbie, N. Banerjee, T. J. Cardinali, J. J. Yang, W. Wu, X. Li, W. M. Tong, D. B. Strukov, G. S. Snider, G. Medeiros-Ribeiro, and R. S. Williams, Nano Lett. 9, 3640 (2009).
- ⁷H. Yeon, P. Lin, C. Choi, S. H. Tan, Y. Park, D. Lee, J. Lee, F. Xu, B. Gao, H. Wu, H. Qian, Y. Nie, S. Kim, and J. Kim, Nat. Nanotechnol. **15**, 574 (2020).
- ⁸M. Y. Chee, P. A. Dananjaya, G. J. Lim, Y. Du, and W. S. Lew, ACS Appl. Mater. Interfaces 14, 35959 (2022).
- ⁹J. M. Ang, P. A. Dananjaya, S. C. W. Chow, G. J. Lim, C. S. Seet, and W. S. Lew, Nanotechnology **34**, 185202 (2023).
- ¹⁰D. Jana, S. Roy, R. Panja, M. Dutta, S. Z. Rahaman, R. Mahapatra, and S. Maikap, Nanoscale Res. Lett. **10**, 188 (2015).
- ¹¹Y. M. Sun, C. Song, J. Yin, L. L. Qiao, R. Wang, Z. Y. Wang, X. Z. Chen, S. Q. Yin, M. S. Saleem, H. Q. Wu, F. Zeng, and F. Pan, Appl. Phys. Lett. **114**, 193502 (2019).
- ¹²M. Saadi, P. Gonon, C. Vallée, F. Jomni, E. Jalaguier, and A. Bsiesy, J. Mater. Sci. 31, 13487 (2020).
- ¹³H. Wang, Y. Du, Y. Li, B. Zhu, W. R. Leow, Y. Li, J. Pan, T. Wu, and X. Chen, Adv. Funct. Mater. 25, 3825 (2015).
- ¹⁴D. Jana, S. Chakrabarti, S. Z. Rahaman, and S. Maikap, Nanoscale Res. Lett. 10, 392 (2015).
- ¹⁵H. J. Kim, T. H. Park, K. J. Yoon, W. M. Seong, J. W. Jeon, Y. J. Kwon, Y. Kim, D. E. Kwon, G. S. Kim, T. J. Ha, S. G. Kim, J. H. Yoon, and C. S. Hwang, Adv. Funct. Mater. **29**, 1806278 (2019).
- ¹⁶W. Banerjee and H. Hwang, Adv. Electron. Mater. 6, 2000488 (2020).
- ¹⁷P. A. Dananjaya, D. J. J. Loy, S. C. W. Chow, and W. S. Lew, ACS Appl. Electron. Mater. 1, 2076 (2019).
- ¹⁸Q. Liu, J. Sun, H. Lv, S. Long, K. Yin, N. Wan, Y. Li, L. Sun, and M. Liu, Adv. Mater. 24, 1844 (2012).
- ¹⁹ J. Yu, X. Xu, T. Gong, Q. Luo, D. Dong, P. Yuan, L. Tai, J. Yin, X. Zhu, X. Wu, H. Lv, and M. Liu, Nanoscale Res. Lett. **14**, 111 (2019).
- ²⁰A. Saleem, F. M. Simanjuntak, S. Chandrasekaran, S. Rajasekaran, T. Y. Tseng, and T. Prodromakis, Appl. Phys. Lett. **118**, 112103 (2021).
- ²¹G. Baek, S. Yang, and T. Kim, Microelectron. Eng. 215, 110987 (2019).

- ²²H. Abbas, J. Li, and D. Ang, Micromachines 13, 725 (2022).
- ²³W. Banerjee, Electronics 9, 1029 (2020).
- ²⁴J. Woo and H. Hwang, ECS J. Solid State Sci. Technol. 5, Q98 (2016).
- ²⁵A. Belmonte, T. Witters, A. Covello, G. Vereecke, A. Franquet, V. Spampinato, S. Kundu, M. Mao, H. Hody, G. S. Kar, J. Radhakrishnan, L. Goux, G. L. Donadio, P. Kumbhare, A. Redolfi, R. Delhougne, L. Nyns, and W. Devulder, in 2019 IEEE International Electron Devices Meeting (IEDM) (IEEE, 2019), pp. 35.8.1–35.8.4.
- ²⁶J. Radhakrishnan, A. Belmonte, L. Nyns, W. Devulder, G. Vereecke, G. L. Donadio, P. Kumbhare, R. Delhougne, M. Houssa, G. S. Kar, and L. Goux, Appl. Phys. Lett. **117**, 151902 (2020).
- ²⁷Y. J. Choi, S. Bang, T.-H. Kim, K. Hong, S. Kim, S. Kim, B.-G. Park, and W. Y. Choi, ACS Appl. Electron. Mater. 5, 1834 (2023).
- ²⁸S. Yu and P. Y. Chen, IEEE Solid-State Circuits Mag. 8, 43 (2016).
- ²⁹F. Pan, S. Gao, C. Chen, C. Song, and F. Zeng, Mater. Sci. Eng. R 83, 1 (2014).
- ³⁰D. S. Jeong, R. Thomas, R. S. Katiyar, J. F. Scott, H. Kohlstedt, A. Petraru, and C. S. Hwang, Rep. Prog. Phys. **75**, 076502 (2012).
- ³¹L. Chen, Y. Xu, Q. Q. Sun, H. Liu, J. J. Gu, S. J. Ding, and D. W. Zhang, IEEE Electron Device Lett. **31**, 356 (2010).
- ³²S. K. Vishwanath, H. Woo, and S. Jeon, Appl. Phys. Lett. **112**, 253503 (2018).
- ³³H. Woo, S. K. Vishwanath, and S. Jeon, ACS Appl. Mater. Interfaces 10, 8124 (2018).
- ³⁴L. Goux, K. Sankaran, G. Kar, N. Jossart, K. Opsomer, R. Degraeve, G. Pourtois, G.-M. Rignanese, C. Detavernier, S. Clima, Y.-Y. Chen, A. Fantini, B. Govoreanu, D. J. Wouters, M. Jurczak, L. Altimime, and J. A. Kittl, in 2012 Symposium on VLSI Technology (IEEE, 2012), pp. 69–70.

- ³⁵W. Devulder, K. Opsomer, F. Seidel, A. Belmonte, R. Muller, B. De Schutter, H. Bender, W. Vandervorst, S. Van Elshocht, M. Jurczak, L. Goux, and C. Detavernier, ACS Appl. Mater. Interfaces 5, 6984 (2013).
- ³⁶L. Qiao, Y. Sun, C. Song, S. Yin, Q. Wan, J. Liu, R. Wang, F. Zeng, and F. Pan, J. Phys. Chem. C 124, 11438 (2020).
- ³⁷Y. T. Tseng, I. C. Chen, T. C. Chang, J. C. Huang, C. C. Shih, H. X. Zheng, W. C. Chen, M. H. Wang, W. C. Huang, M. C. Chen, X. H. Ma, Y. Hao, and S. M. Sze, Appl. Phys. Lett. **113**, 053501 (2018).
- ³⁸J. Wang, G. Cao, K. Sun, J. Lan, Y. Pei, J. Chen, and X. Yan, Nanoscale 14, 1318 (2022).
- ³⁹Z. Yang, Q. Zhan, X. Zhu, Y. Liu, H. Yang, B. Hu, J. Shang, L. Pan, B. Chen, and R. W. Li, EPL 108, 58004 (2014).
- ⁴⁰G. Schindler, G. Steinlesberger, M. Engelhardt, and W. Steinhögl, Solid State Electron. 47, 1233 (2003).
- ⁴¹Q. Huang, C. M. Lilley, M. Bode, and R. S. Divan, in 2008 8th IEEE Conference on Nanotechnology (IEEE-NANO, 2008), p. 549.
- ⁴²S. Yang, B. Zhang, Q. Zhang, R. Wang, X. Yu, C. Wang, and Y. Liu, ECS J. Solid State Sci. Technol. 8, P416 (2019).
- ⁴³R. Nafria, A. Genç, M. Ibáñez, J. Arbiol, P. Ramírez De La Piscina, N. Homs, and A. Cabot, Langmuir 32, 2267 (2016).
- ⁴⁴X. Xia, Y. Wang, A. Ruditskiy, and Y. Xia, Adv. Mater. **25**, 6313 (2013).
- ⁴⁵M. Al-Mamun, S. W. King, and M. Orlowski, ECS J. Solid State Sci. Technol. 8, N220 (2019).
- ⁴⁶T. A. De Assis, F. Borondo, R. M. Benito, and R. F. S. Andrade, Phys. Rev. B 78, 235427 (2008).
- ⁴⁷J. Idrac, G. Mankowski, G. Thompson, P. Skeldon, Y. Kihn, and C. Blanc, Electrochim. Acta 52, 7626 (2007).
- ⁴⁸J. Park, H. Jeon, H. Kim, W. Jang, H. Seo, and H. Jeon, RSC Adv. 4, 61064 (2014).