



Zirconia quantum dots for a nonvolatile resistive random access memory device*

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Abstract: We propose a nonvolatile resistive random access memory device by employing nanodispersion of zirconia (ZrO₂) quantum dots (QDs) for the formation of an active layer. The memory devices comprising a typical sandwich structure of Ag (top)/ZrO₂ (active layer)/Ti (bottom) are fabricated using a facile spin-coating method. The optimized device exhibits a high resistance state/low resistance state resistance difference (about 10 Ω), a good cycle performance (the number of cycles larger than 100), and a relatively low conversion current (about 1 μA). Atomic force microscopy and scanning electron microscope are used to observe the surface morphology and stacking state of the ZrO₂ active layer. Experimental results show that the ZrO₂ active layer is stacked compactly and has a low roughness (Ra=4.49 nm) due to the uniform distribution of the ZrO₂ QDs. The conductive mechanism of the Ag/ZrO₂/Ti device is analyzed and studied, and the conductive filaments of Ag ions and oxygen vacancies are focused on to clarify the resistive switching memory behavior. This study offers a facile approach of memristors for future electronic applications.

Key words: Zirconia quantum dot; Resistive switching; Memory device; Spin coating

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1 Introduction

In the information age, given the continuous developments of computer technology and the universal applications of smartphones, the means of storing information is particularly important (Han et al., 2017, 2018; Jiang et al., 2017; Craig, 2018; Wan et al., 2018; Vescio et al., 2019). However, in the architecture adopted in traditional computers,

computing and storing functions are separated and performed by a central processing unit (CPU) and the memory, respectively (Liu et al., 2017; Zhou et al., 2018; Wang et al., 2019). While the speed and capacity of the CPUs and memory are increasing rapidly, increases of bus speed for transferring data and instructions have been quite limited. Memristors or resistance random access memory (RRAM) can be used to not only store data but also implement logic calculations (Pan et al., 2014). Therefore, RRAM can effectively solve the problem of the speed mismatch between a CPU and memory, and improve information processing efficiency. Strukov et al. (2008) proved the existence of resistive switching memory through a physical experiment, which initiated an upsurge in the RRAM research. Memristors have demonstrated that they can realize high resistance

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state (HRS) (“Off” state) and low resistance state (LRS) (“On” state) conversion during operation. If the HRS or “Off” state in the logic is regarded as “0,” the LRS or “On” state in the logic is regarded as “1,” then the RRAM can realize information storage in a binary form (Sun et al., 2018b). Therefore, in recent years, memristors have been considered the most promising candidates for the new generation of non-volatile storage devices (Chua, 2011; Kadhim et al., 2018).

Memristors can be prepared from a wide variety of sources, such as metal oxides (Liang et al., 2015; Lyu et al., 2015; Sun et al., 2018a; Yu et al., 2018), perovskites (Yan et al., 2016; Zhang et al., 2017), two-dimensional layered materials (Han et al., 2017; Zhao et al., 2017), and biological material (Sun et al., 2018b). Metal-oxide structures have been extensively studied due to their low cost and simple preparation process, and zirconia (ZrO_2) has emerged as a prospective candidate for the active functional layer in memory devices (Emelyanov et al., 2019). ZrO_2 has many advantages, such as a high dielectric constant, stable chemical properties, nontoxicity, and simple composition (Panda and Tseng, 2013; Xia et al., 2018). Therefore, ZrO_2 has been widely applied to the preparation of a variety of optical and electronic devices (Panda and Tseng, 2013; He et al., 2019a). Recently, due to the various advantages of ZrO_2 , RRAM devices based on a ZrO_2 active layer have been proposed (Emelyanov et al., 2019). However, the preparation methods for RRAM devices are complex, as they usually involve a magnetron sputtering method (Wang et al., 2011; Emelyanov et al., 2019). As far as we know, using a simple spin-coating method for the preparation of an Ag/ ZrO_2 /Ti sandwich structure has rarely been reported. In this work, aqueous nanodispersion of ZrO_2 quantum dots (QDs), prepared using a hydrothermal method coupled with alkaline hydrogen peroxide (AHP) process, is used for the formation of an active layer in the memristor of the memory devices fabricated in a typical Ag/ ZrO_2 /Ti sandwich structure. The optimized device shows an HRS/LRS resistance difference (about 10 Ω), a good cycle performance (the number of cycles larger than 100), and a relatively low conversion current (about 1 μA) at room temperature. In this work, we demonstrate the potential applications of ZrO_2 QDs as an

active layer in the fabrication of high-performance memory devices through a simple spin-coating method.

2 Experiments

2.1 Preparation of aqueous nanodispersion of ZrO_2 QDs

Aqueous nanodispersion of ZrO_2 QDs was synthesized using a conventional hydrothermal method coupled with an AHP process (He et al., 2019b). Zirconium carbonate basic (ZCB, CAS:57219-64-4, ZrO_2 content >40%), sodium hydroxide (NaOH, 99.99%), hydrogen peroxide (H_2O_2 , 30%), and nitric acid as sources for reaction were purchased from the Aladdin Co., Ltd. (Shanghai of China), to prepare the aqueous ZrO_2 nanodispersion. Deionized water was deputed using a Smart-S30 water purification system (Hitech). For a typical experiment, 20 g NaOH, 20 g ZCB, and 40 g deionized water were weighed and heated in a hydrothermal kettle at 110 $^\circ C$ for 6 h. All the products (including solids and liquids) were moved from the hydrothermal kettle to a 250-mL round bottom flask, and then 85 mL deionized water was added to dilute them. Finally, 5.1 mL H_2O_2 was added at 50 $^\circ C$ for 5 h under vigorous stirring. Subsequently, the excess alkali solution was removed using a decantation process and the wet solids were washed repeatedly with deionized water to remove the excess ions. Finally, the wet solids were added to a diluted nitric acid solution (1.5 mol/L) to turn them into transparent and stable aqueous ZrO_2 nanodispersion after stirring.

2.2 Preparation of Ag/ ZrO_2 /Ti sandwich structural memristors

Fig. 1 shows the manufacturing process of the Ag/ ZrO_2 /Ti sandwich structure. First, titanium foils (1 cm \times 3 cm \times 0.1 mm) were cleaned with alcohol in an ultrasound bath. Then, the titanium foil was fixed on a spin coater (KW-4A), and aqueous nanodispersion of ZrO_2 QDs with a 1% solid content was pipetted onto the titanium foil with a pipette. The spin coater was set to 5 s (500 r/min) for the first cycle and 20 s (1500 r/min) for the second cycle. The titanium foil loaded with the ZrO_2 nanoparticle layer was then

dried in a vacuum oven at 90°C for 6 h. Then, Ag was sputtered on the top of the ZrO₂ QD layer using a metal shadow mask to form the sandwich structure.

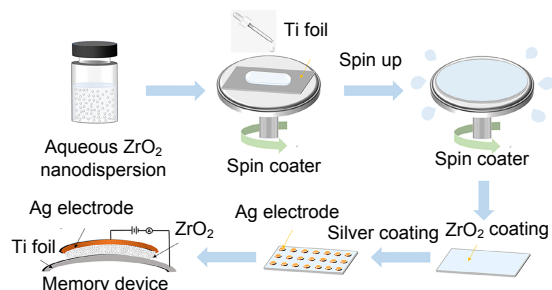


Fig. 1 Manufacturing process of the Ag/ZrO₂/Ti device

2.3 Characterization

The hydrated particle size of the ZrO₂ QDs in the aqueous solution was tested by dynamic light scattering (DLS, Malvern Nano ZS90, England). The density of the stacked layers and coating morphology of the ZrO₂/Ti foil were examined using scanning electron microscope (SEM, Hitachi S4800). The surface morphology of the ZrO₂/Ti foil was also investigated by atomic force microscopy (AFM, Bruker

Dimension Icon, Germany) with tapping-mode pattern. The magnitude and morphology of the ZrO₂ nanoparticles were obtained using transmission electron microscope (TEM, Hitachi HT-7700). The lattice structure and size of the ZrO₂ QDs were determined by high-resolution TEM (HRTEM, Hitachi H-9500). The particle size of the ZrO₂ QDs in the TEM was statistically analyzed using the Nano Measurer software. The X-ray diffraction (XRD) patterns of the samples were measured using an XRD-6000 diffractometer (Shimadzu Inc.). The current-voltage (*I-V*) curves of the Ag/ZrO₂/Ti devices were measured with a CHI 660B electrochemical workstation (CH Instruments).

3 Results and discussion

ZrO₂ QDs prepared using the hydrothermal method and AHP treatment are characterized (Fig. 2). According to the DLS results (Fig. 2a), the *z*-average particle diameter of the aqueous ZrO₂ nanodispersion is 14.52 nm and the PDI value is 0.370, showing that the AHP-treated ZrO₂ QDs have good dispersibility

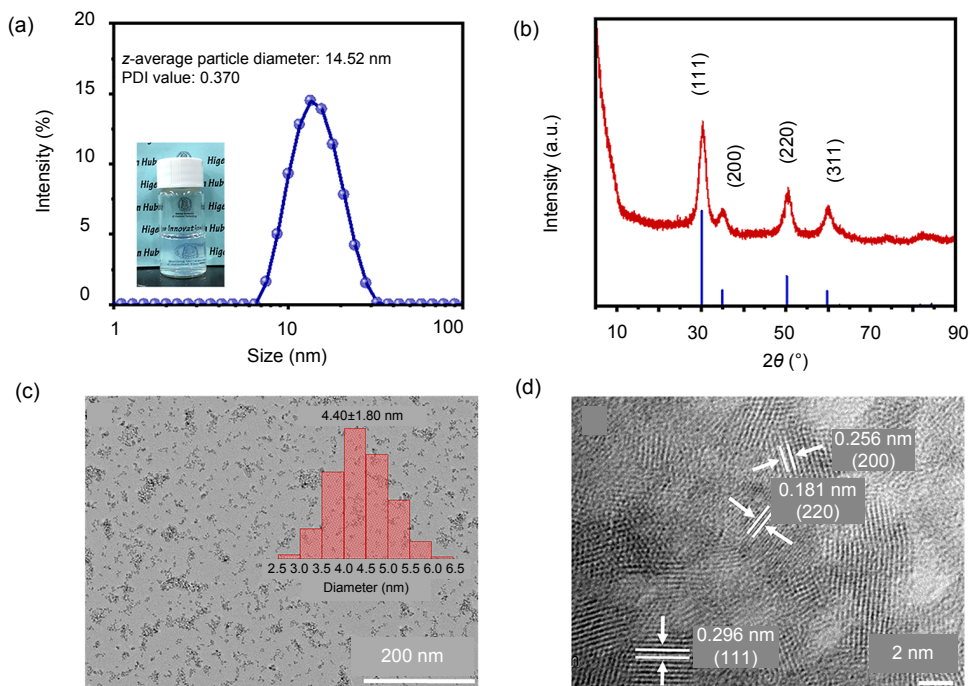


Fig. 2 Characterization of ZrO₂ QDs: (a) DLS results of aqueous ZrO₂ nanodispersion (inset: ZrO₂ nanodispersion with 1% solid content); (b) XRD curve of the ZrO₂ nanopowder; (c) morphologies and structural characteristic of aqueous ZrO₂ nanodispersion (inset: particle size distributions of single ZrO₂ particles); (d) HRTEM of ZrO₂ QDs

in water. Fig. 2b shows that the phase of hydrophilic ZrO₂ QDs is the single cubic phase (JCPDS: 49-1642) given their XRD patterns. The diffraction peaks are observed at 30.12°, 34.96°, 50.22°, and 59.74°, corresponding to the crystal planes of (111), (200), (220), and (311), respectively. The ZrO₂ QDs with cubic phase have the highest density and the most stable structure in the three crystal phases of ZrO₂, and are an ideal choice for optoelectronic applications (He et al., 2019b). Fig. 2c shows a TEM image of the aqueous ZrO₂ nanodispersion, which has an average particle size of 4.40 nm. It can be seen from the TEM image that the ZrO₂ QDs do not reach a monodisperse state, but mainly a small agglomeration formed by several particles. This corresponds to the DLS results that a small agglomeration formed mainly by 2–5 ZrO₂ QDs was obtained. Using the inorganic zirconium source as a raw material to prepare the ZrO₂ QDs, the growth of the hydroxyl group on the surface of the ZrO₂ is promoted only by the oxidation of H₂O₂. It is not easy to obtain such a grade of aqueous ZrO₂ nanodispersion without adding other surfactants or modifiers. At the same time, the surface of the ZrO₂ QDs is treated by H₂O₂, which can increase the number of defect sites. The proportion of defect sites in the layer is related to the performance of the memristors. However, in this preparation method, factors such as surfactants or modifiers that can influence the performance of the memristors are avoided. In Fig. 2d, different lattice lengths can be observed to determine different crystal planes, in which (111) corresponds to 0.296 nm, (220) corresponds to 0.181 nm, and (200) corresponds to 0.256 nm. This corresponds to the XRD results, demonstrating that the crystal phase of ZrO₂ QDs is the cubic phase.

Figs. 3a and 3b show the surface structure of the ZrO₂-coated titanium foil. The hydrophilic ZrO₂ coating clearly shows a film formed by stacking a dense oxide on the titanium sheet. The thickness of the ZrO₂ coating is observed by SEM at around 500 nm (Fig. 3c). Fig. 3d depicts a three-dimensional AFM image of the ZrO₂ coating on the titanium foil. The average surface roughness (Ra) of the ZrO₂ surface coating is 4.49 nm, which is close to the size of a single ZrO₂ QD.

Fig. 4a shows the consecutive *I-V* curves of the Ag/ZrO₂/Ti sandwich structure, which was examined

at a scanning rate of 0.1 V/ms from 0→1.5→0→-1.5→0 V. To avoid permanent electrical breakdown of the Ag/ZrO₂/Ti device, the compliance current is secured at 0.5 A. Results show that the device has a quick response even in very short time and a relatively low switching current (about 1 μA). At the same time, it is found that the device exhibits a very typical resistive switching performance. As the forward voltage decreases backswept, the device transfers from LRS to HRS. Fig. 4b shows a resistive switching behavior in a single cyclic sweep, which exhibits good “Set” and “Reset” performances. Considering the practical application of memristors, a stable cycle performance and a large HRS/LRS resistance difference are necessary.

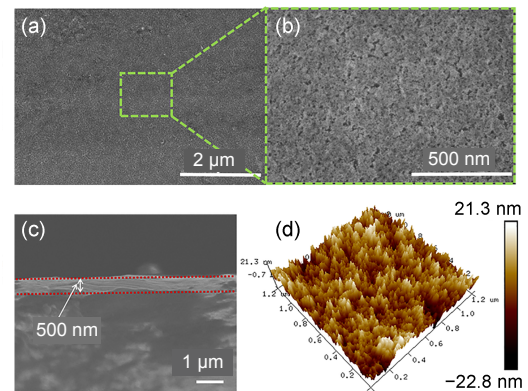


Fig. 3 SEM images (a and b), the cross sectional SEM image (c), and an AFM image (d) of ZrO₂ coating on the titanium foil

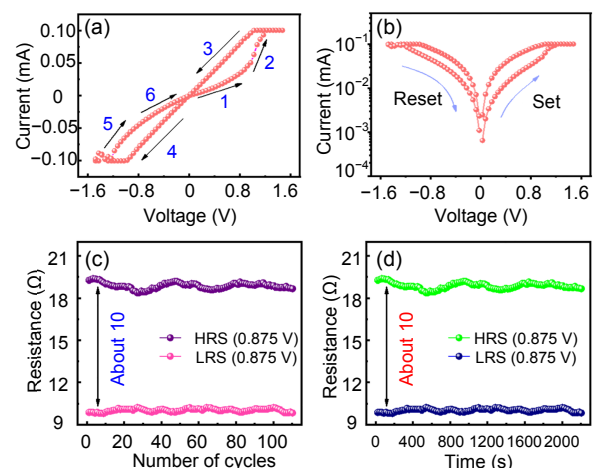


Fig. 4 *I-V* curves of a single cycle when the voltage changes from 0→1.5→0→-1.5→0 V (a and b), and resistance curves vs. the number of cycles and time under a positive voltage of 0.875 V (c and d)

The resistance difference is obtained by contrasting the current density of the LRS (“On” state) with that of the HRS (“Off” state) at a specific voltage (0.875 V). Resistance memories of both the HRS and LRS are calculated (Figs. 4c and 4d). Results show that the HRS/LRS resistance difference is about 10 Ω . It is also shown that the HRS/LRS resistance difference remains constant under a long cycle (the number of cycles larger than 100). The above results show that the Ag/ZrO₂/Ti device has a stable performance.

To realize the resistive switching mechanism in the devices, the curve is secured at a log-log scale (Figs. 5a and 5b). Figs. 5a and 5b show the I - V curves in the negative and positive voltage sections of the Ag/ZrO₂/Ti device, respectively. Fig. 5a represents the charges distributed through the Ag anode into the ZrO₂ active layer, leading to the conduction mechanism (Sleiman et al., 2012). The slope of this partial linear portion is 1.036 (close to 1), indicating Ohmic conductance. As the applied voltage increases, the log I vs. log V has a linear relationship and the slope is 2.29 (Fig. 5a), indicating $I \propto V^m$. Therefore, the conduction performance of the Ag/ZrO₂/Ti device follows the regulation of classical trap-controlled space charge limited conduction (SCLC) (Siddiqui et al., 2017). It shows that the charge transfer happens between the Ag electrode and the ZrO₂ layer. This can be demonstrated by the formation of the Ag conductive filaments (Kadhim et al., 2018).

In the LRS of the positive part, the I - V curve is well secured through the Ohmic conductance performance with a slope of about 0.99. SCLC in semiconductors and their corresponding space charge effects are important basic concepts. The SCLC model generally contains three regimes: low voltage Ohmic region ($I \propto V$), Child’s square region ($I \propto V^2$), and current steep increase region ($I \propto V^m$, $m > 2$). The formula for the SCLC model is expressed as follows (Vishwanath and Kim, 2016):

$$J \propto V^{m+1} / t^{2m+1}, \quad (1)$$

where J , V , t , and m are the current density of ZrO₂, bias voltage, thickness, and fitting index, respectively. When $m=0$, Eq. (1) represents the Ohmic conductance behavior. If $m=1$, it becomes a Child’s square law, which is given by (Vishwanath and Kim, 2016; Li et al., 2017)

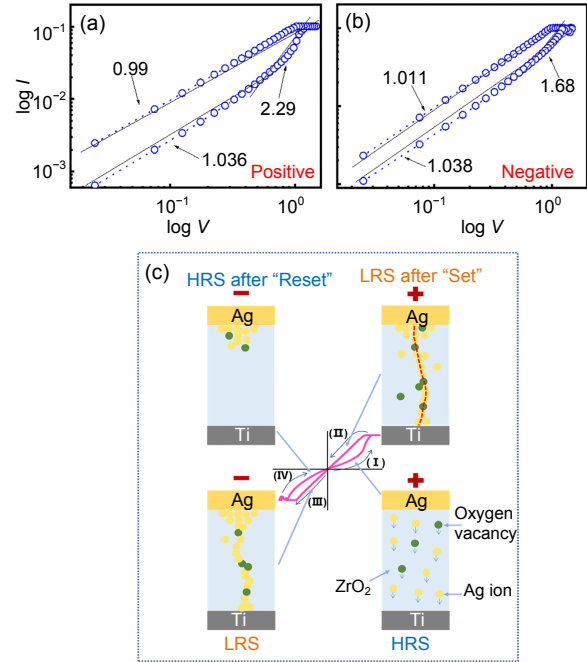


Fig. 5 I - V curves of the Ag/ZrO₂/Ti structure device (a and b) and the speculative model of Ag ion and oxygen vacancy filaments (c)

$$J \propto \frac{9}{8} \varepsilon_0 \varepsilon_r \mu \frac{V^2}{t^3}, \quad (2)$$

where μ , ε_0 , and ε_r are the carrier mobility, vacuum dielectric constant, and relative dielectric constant, respectively. In this work, Ag atoms in the Ag electrode of the Ag/ZrO₂/Ti devices are ionized into Ag ions by the application of an electric field. This behavior can be described as $\text{Ag}^+ + e^- \rightarrow \text{Ag}$ (Sun et al., 2014; Zhou et al., 2017).

At the same time, the possible formation of conductive filaments is analyzed. The Ag ions can move along the electric field direction in the applied electric field (Fig. 5c). When the Ag ions and oxygen vacancies accumulate to a certain extent, the conductivity of the ZrO₂ active layer is greatly increased, because the Ag ions and oxygen vacancies act as conductive filaments, and this process corresponds to the “Set” process of resistance switching (Wu et al., 2017). After the “Set” process, the device retains the LRS (“On” state) until a large voltage of opposite polarity ($< V_{\text{Reset}}$) dissolves the Ag and oxygen vacancy filaments, with this route corresponding to “Reset” of the Ag/ZrO₂/Ti device, following which the Ag ions and oxygen vacancies are moved back to

the Ag electrode. Therefore, the Ag/ZrO₂/Ti device can be reversibly switched by connecting and disconnecting conducting filaments.

4 Conclusions

In this work, nonvolatile resistive random access memory devices using ZrO₂ QDs as an active layer have been devised via a simple spin-coating process. The resistive switching memory devices based on Ag (top)/ZrO₂ (active layer)/Ti (bottom) showed an HRS/LRS resistance difference (about 10 Ω), a good cycle performance (the number of cycles larger than 100), and a relatively low conversion current (about 1 μA). This result revealed that the Ag ions and oxygen vacancies are organized as conducting filaments inside the ZrO₂ coating, and provided a new approach to nonvolatile memory devices based on a ZrO₂ active layer for further electronic applications.

Compliance with ethics guidelines

Xiang-lei HE, Rui-jie TANG, Feng YANG, Mayameen S. KADHIM, Jie-xin WANG, Yuan PU, and Dan WANG declare that they have no conflict of interest.

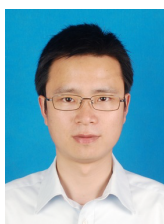
References

- Chua L, 2011. Resistance switching memories are memristors. *Appl Phys A*, 102(4):765-783. <https://doi.org/10.1007/s00339-011-6264-9>
- Craig J, 2018. Cybersecurity research—essential to a successful digital future. *Engineering*, 4(1):9-10. <https://doi.org/10.1016/j.eng.2018.02.006>
- Emelyanov AV, Nikiruy KE, Demin VA, et al., 2019. Yttria-stabilized zirconia cross-point memristive devices for neuromorphic applications. *Microelectron Eng*, 215: 110988. <https://doi.org/10.1016/j.mee.2019.110988>
- Han PD, Sun B, Li J, et al., 2017. Ag filament induced non-volatile resistive switching memory behaviour in hexagonal MoSe₂ nanosheets. *J Coll Interf Sci*, 505:148-153. <https://doi.org/10.1016/j.jcis.2017.05.082>
- Han WB, Chen XG, Li SF, et al., 2018. A novel non-volatile memory storage system for I/O-intensive applications. *Front Inform Technol Electron Eng*, 19(10):1291-1302. <https://doi.org/10.1631/FITEE.1700061>
- He XL, Tang RG, Pu Y, et al., 2019a. High-gravity-hydrolysis approach to transparent nanozirconia/silicone encapsulation materials of light emitting diodes devices for healthy lighting. *Nano Energy*, 62:1-10. <https://doi.org/10.1016/j.nanoen.2019.05.024>
- He XL, Wang Z, Wang D, et al., 2019b. Sub-kilogram-scale synthesis of highly dispersible zirconia nanoparticles for hybrid optical resins. *Appl Surf Sci*, 491:505-516. <https://doi.org/10.1016/j.apsusc.2019.06.187>
- Jiang H, Belkin D, Savel'ev SE, et al., 2017. A novel true random number generator based on a stochastic diffusive memristor. *Nat Commun*, 8(1):882. <https://doi.org/10.1038/s41467-017-00869-x>
- Kadhim MS, Yang F, Sun B, et al., 2018. A resistive switching memory device with a negative differential resistance at room temperature. *Appl Phys Lett*, 113(5):053502. <https://doi.org/10.1063/1.5037191>
- Li XM, Tao L, Chen ZF, et al., 2017. Graphene and related two-dimensional materials: structure-property relationships for electronics and optoelectronics. *Appl Phys Rev*, 4(2):021306. <https://doi.org/10.1063/1.4983646>
- Liang L, Li K, Xiao C, et al., 2015. Vacancy associates-rich ultrathin nanosheets for high performance and flexible nonvolatile memory device. *J Am Chem Soc*, 137(8): 3102-3108. <https://doi.org/10.1021/jacs.5b00021>
- Liu X, Lu YT, Yu J, et al., 2017. ONFS: a hierarchical hybrid file system based on memory, SSD, and HDD for high performance computers. *Front Inform Technol Electron Eng*, 18(12):1940-1971. <https://doi.org/10.1631/FITEE.1700626>
- Lyu MJ, Liu YW, Zhi YD, et al., 2015. Electric-field-driven dual vacancies evolution in ultrathin nanosheets realizing reversible semiconductor to half-metal transition. *J Am Chem Soc*, 137(47):15043-15048. <https://doi.org/10.1021/jacs.5b10212>
- Pan F, Gao S, Chen C, et al., 2014. Recent progress in resistive random access memories: materials, switching mechanisms, and performance. *Mater Sci Eng R Rep*, 83:1-59. <https://doi.org/10.1016/j.mser.2014.06.002>
- Panda D, Tseng TY, 2013. Growth, dielectric properties, and memory device applications of ZrO₂ thin films. *Thin Sol Film*, 531:1-20. <https://doi.org/10.1016/j.tsf.2013.01.004>
- Siddiqui GU, Rehman MM, Choi KH, 2017. Resistive switching phenomena induced by the heterostructure composite of ZnSnO₃ nanocubes interspersed ZnO nanowires. *J Mater Chem C*, 5(22):5528-5537. <https://doi.org/10.1039/c7tc01105a>
- Sleiman A, Mabrook MF, Nejm RR, et al., 2012. Organic bistable devices utilizing carbon nanotubes embedded in poly (methyl methacrylate). *J Appl Phys*, 112(2):024509.

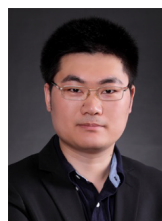
- <https://doi.org/10.1063/1.4737599>
- Strukov DB, Snider GS, Stewart DR, et al., 2008. The missing memristor found. *Nature*, 453(7191):80-83. <https://doi.org/10.1038/nature06932>
- Sun B, Li HW, Wei LJ, et al., 2014. Hydrothermal synthesis and resistive switching behaviour of $\text{WO}_3/\text{CoWO}_4$ core-shell nanowires. *Cryst Eng Comm*, 16(42):9891-9895. <https://doi.org/10.1039/C4CE01442A>
- Sun B, Zhu SH, Mao SS, et al., 2018a. From dead leaves to sustainable organic resistive switching memory. *J Coll Interf Sci*, 513:774-778. <https://doi.org/10.1016/j.jcis.2017.12.007>
- Sun B, Zhang XJ, Zhou GD, et al., 2018b. A flexible non-volatile resistive switching memory device based on ZnO film fabricated on a foldable PET substrate. *J Coll Interf Sci*, 520:19-24. <https://doi.org/10.1016/j.jcis.2018.03.001>
- Vescio G, Martín G, Crespo-Yepes A, et al., 2019. Low-power, high-performance, non-volatile inkjet-printed HfO_2 -based resistive random access memory: from device to nanoscale characterization. *ACS Appl Mater Interf*, 11(26):23659-23666. <https://doi.org/10.1021/acsami.9b01731>
- Vishwanath SK, Kim J, 2016. Resistive switching characteristics of all-solution-based $\text{Ag}/\text{TiO}_2/\text{Mo}$ -doped In_2O_3 devices for non-volatile memory applications. *J Mater Chem C*, 4(46):10967-10972. <https://doi.org/10.1039/c6tc03607d>
- Wan T, Qu B, Du HW, et al., 2018. Digital to analog resistive switching transition induced by graphene buffer layer in strontium titanate based devices. *J Coll Interf Sci*, 512:767-774. <https://doi.org/10.1016/j.jcis.2017.10.113>
- Wang SY, Tsai CH, Lee DY, et al., 2011. Improved resistive switching properties of $\text{Ti}/\text{ZrO}_2/\text{Pt}$ memory devices for RRAM application. *Microelectron Eng*, 88(7):1628-1632. <https://doi.org/10.1016/j.mee.2010.11.058>
- Wang ZR, Li C, Song WH, et al., 2019. Reinforcement learning with analogue memristor arrays. *Nat Electron*, 2(3):115-124. <https://doi.org/10.1038/s41928-019-0221-6>
- Wu Y, Wei Y, Huang Y, et al., 2017. Capping CsPbBr_3 with ZnO to improve performance and stability of perovskite memristors. *Nano Res*, 10(5):1584-1594. <https://doi.org/10.1007/s12274-016-1288-2>
- Xia Y, Zhang C, Wang JX, et al., 2018. Synthesis of transparent aqueous ZrO_2 nanodispersion with a controllable crystalline phase without modification for a high-refractive-index nanocomposite film. *Langmuir*, 34(23):6806-6813. <https://doi.org/10.1021/acs.langmuir.8b00160>
- Yan XB, Li YC, Zhao JH, et al., 2016. Roles of grain boundary and oxygen vacancies in $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ films for resistive switching device application. *Appl Phys Lett*, 108(3):033108. <https://doi.org/10.1063/1.4940198>
- Yu YM, Yang F, Mao SS, et al., 2018. Effect of anodic oxidation time on resistive switching memory behavior based on amorphous TiO_2 thin films device. *Chem Phys Lett*, 706:477-482. <https://doi.org/10.1016/j.cplett.2018.06.063>
- Zhang YY, Yang T, Yan XB, et al., 2017. A metal/ $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3/\text{SiO}_2/\text{Si}$ single film device for charge trapping memory towards a large memory window. *Appl Phys Lett*, 110(22):223501. <https://doi.org/10.1063/1.4984220>
- Zhao H, Dong ZP, Tian H, et al., 2017. Atomically thin femtojoule memristive device. *Adv Mater*, 29(47):1703232. <https://doi.org/10.1002/adma.201703232>
- Zhou GD, Sun B, Zhou AK, et al., 2017. A larger nonvolatile bipolar resistive switching memory behaviour fabricated using eggshells. *Curr Appl Phys*, 17(2):235-239. <https://doi.org/10.1016/j.cap.2016.09.018>
- Zhou J, Li PG, Zhou YH, et al., 2018. Toward new-generation intelligent manufacturing. *Engineering*, 4(1):11-20. <https://doi.org/10.1016/j.eng.2018.01.002>



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