Resistive switching memories in MoS₂ nanosphere assemblies

Xu, Xiao-Yong; Yin, Zong-You; Xu, Chun-Xiang; Dai, Jun; Hu, Jing-Guo

2014

Xu, X.-Y., Yin, Z.-Y., Xu, C.-X., Dai, J., & Hu, J. G. (2014). Resistive switching memories in MoS₂ nanosphere assemblies. *Applied Physics Letters*, 104(3), 033504-.

<https://hdl.handle.net/10356/80057>

<https://doi.org/10.1063/1.4862755>

© 2014 AIP Publishing LLC. This paper was published in *Applied Physics Letters* and is made available as an electronic reprint (preprint) with permission of AIP Publishing LLC. The paper can be found at the following official DOI: [<http://dx.doi.org/10.1063/1.4862755>]. One print or electronic copy may be made for personal use only. Systematic or multiple reproduction, distribution to multiple locations via electronic or other means, duplication of any material in this paper for a fee or for commercial purposes, or modification of the content of the paper is prohibited and is subject to penalties under law.

Downloaded on 26 Apr 2023 11:24:57 SGT

Resistive switching memories in MoS₂ nanosphere assemblies

Xiao-Yong Xu, Zong-You Yin, Chun-Xiang Xu, Jun Dai, and Jing-Guo Hu

Citation: [Applied Physics Letters](#) **104**, 033504 (2014); doi: 10.1063/1.4862755

View online: <http://dx.doi.org/10.1063/1.4862755>

View Table of Contents: <http://scitation.aip.org/content/aip/journal/apl/104/3?ver=pdfcov>

Published by the [AIP Publishing](#)

The advertisement features a photograph of the Model PS-100 probe station, which is a complex mechanical device with various components like a probe head, sample stage, and base. The background of the ad is a gradient of blue. On the left, the text "NEW" is in orange, and "Model PS-100" is in large, bold, dark blue. Below it, "Preconfigured Tabletop Probe Station" is in a smaller, dark blue font. On the right, the "Lake Shore CRYOTRONICS" logo is shown, with "Lake Shore" in a large, white, serif font and "CRYOTRONICS" in a smaller, white, sans-serif font. Below the logo, the tagline "An affordable solution for a wide range of research" is written in a white, italicized, serif font.

NEW
Model PS-100
Preconfigured Tabletop
Probe Station

Lake Shore
CRYOTRONICS

*An affordable solution for
a wide range of research*

Resistive switching memories in MoS₂ nanosphere assemblies

Xiao-Yong Xu,^{1,2,3,a)} Zong-You Yin,³ Chun-Xiang Xu,^{2,a)} Jun Dai,² and Jing-Guo Hu^{1,a)}

¹School of Physics Science and Technology, Yangzhou University, Yangzhou 225002, China

²State Key Laboratory of Bioelectronics and School of Electronic Science and Engineering, Southeast University, Nanjing 210096, China

³School of Materials Science and Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore, Singapore 639798

(Received 14 November 2013; accepted 5 January 2014; published online 21 January 2014)

A resistive switching memory device consisting of reduced graphene oxide and indium tin oxide as top/bottom two electrodes, separated by dielectric MoS₂ nanosphere assemblies as the active interlayer, was fabricated. This device exhibits the rewritable nonvolatile resistive switching with low SET/RESET voltage (~ 2 V), high ON/OFF resistance ratio ($\sim 10^4$), and superior electrical bistability, introducing a potential application in data storage field. The resistance switching mechanism was analyzed in the assumptive model of the electron tunneling across the polarized potential barriers. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4862755>]

The resistance switching (RS) phenomenon basing on the electrically stimulated change of the resistance in a metal–insulator–metal (MIM) sandwich structure has recently attracted a great deal of attention due to potential application for nonvolatile random-access memory (NVRAM).^{1–3} In the resistive memory cells, the resistance switches between a high resistance state (HRS) and a low resistance state (LRS) under the regulation of the formation and elimination of conductive pathways induced by the external electric field.⁴ Developing nanoscale RS memory cells with high ON/OFF resistance ratio, fast switching speed, and low power consumption is one key step for achieving the application of ultrahigh density NVRAM.⁵ So far, the RS characteristic has been observed in many semiconducting and insulating materials including binary transition metal oxides,^{6–8} perovskite oxides,^{9–11} chalcogenides,^{12,13} sulfides,¹⁴ amorphous silicon,¹⁵ and organic materials.^{16,17} Accordingly, various models have been suggested, including the metal-insulator phase transition,^{11,18} the ferroelectric polarization,^{19,20} and the conductive bridge constructed by the migration of localized metal atoms or defects,^{8,21} and the RS mechanisms are still being debated.^{1,2} Recently, molybdenum disulphide (MoS₂) has aroused increasing interests for various applications in electronic and optoelectronic devices, such as field-effect transistors (FET),^{22,23} integrated circuits,²⁴ phototransistors,^{25,26} and so on. Moreover, the different conductive properties like metal, insulator even superconductivity have been observed, respectively, in MoS₂,^{27,28} although the natures of charge transport still remain elusive.²⁹ More recently, the MoS₂/graphene heterostructure memory devices have been constructed by using monolayer MoS₂ as the channel and graphene as electrodes in the FET geometry, and the gate voltage-modulated nature of electronic states in the two-dimensional (2D) ultrathin MoS₂ nanosheets has been demonstrated.^{30,31} And Yin *et al.*³² reported recently a sandwich-structured memory diode by using the mixture of 2D MoS₂ and graphene oxide (GO) nanosheets as the

channel material, which showed that the MoS₂ nanosheets introduced in the mixed MoS₂-GO film might facilitate the oxygen migration of GO during device operation but the really active component was still GO. Herein, in the MoS₂ nanospheres-based diode, we found that the MoS₂ channel conductivity depends directly on the bias voltage, resulting significantly in the bipolar RS effect with low switching voltage (~ 2 V), high ON/OFF resistance ratio ($\sim 10^4$), and superior electrical bistability. It is worth noting that the nanoparticle assembly system was proposed in 2009 by Kim *et al.*³³ to be promising for RS devices in the context of studying extensively the thin film-based RS devices; however, there are few following reports on the RS behavior in particle systems. In the present MoS₂ nanosphere assemblies, a model of electron tunneling across the polarization potential barriers modulated by the external electric field is proposed as one possible interpretation for the RS behavior. This work devotes to deepen understanding of the RS mechanism and to expand nonvolatile memory devices based on the MoS₂ nanosphere assemblies.

The diode involving MoS₂ nanospheres, reduced graphene oxide (rGO), and indium tin oxide (ITO) as the top and bottom electrodes was fabricated. The MoS₂ nanosphere assembly layer (around 200 nm) was deposited onto ITO glass by a hydrothermal route. Typically, 6 mg sodium molybdate (Na₂MoO₄·2H₂O) and 12 mg thioacetamide (C₂H₅NS) were dissolved in 40 ml deionized water to form a transparent mixture. This solution was transferred to a 100 ml Teflon-lined stainless steel autoclave. Then, a cleansed ITO glass was immersed in the above mixture with a leaning angle against the wall of the autoclave chamber and the autoclave was heated at 200 °C for 20 h in an electric oven. As shown in the scanning electron microscopy (SEM) image of as-synthesized sample in Fig. 1(a), a mass of MoS₂ nanoparticles assemble in the ITO substrate and the amplified SEM image of several particles around the edge in the inset show that the MoS₂ nanoparticles are shaped like spheres with average diameter of 100 nm. The energy dispersive X-ray spectrum (EDX) shown in Fig. 1(b) indicates that the grown nanospheres are composed of characteristic Mo and S

^{a)}Electronic addresses: xxyy@yzu.edu.cn, xcxsu@seu.edu.cn, and jghu@yzu.edu.cn

FIG. 1. (a) SEM image, (b) EDX spectrum, and (c) XRD pattern of MoS₂ nanospheres fabricated on ITO. The inset in (a) shows the amplified SEM image of several nanospheres around the edge.

elements, whereas some other detected elements including Si, In, Ca, and O should come from the ITO glass. Fig. 1(c) presents the XRD pattern of sample, where the standard pattern of hexagonal MoS₂ phase (JCPDS card No. 37-1492) is also inset as a reference. Except two dominated diffraction peaks of 29.8 and 34.7° originated from ITO, the other detected peaks can be assigned to the (004), (104), (105), and (110) planes in the hexagonal MoS₂ phase.³⁴ The indistinction of the (002) peak corresponding to the *c*-plane of layered MoS₂ is reasonable for the present MoS₂ nanospheres. Fig. 2(a) shows the X-ray photoelectron spectroscopy (XPS) survey spectrum of the as-synthesized MoS₂/ITO film, the characteristic peaks of chemically coordinated Mo and S elements are detected in addition to some peaks of inevitable C and O elements and In element from ITO glass. The amplified XPS spectra of Mo 3d and S 2p regions in Figs. 2(b) and 2(c) identify the strong Mo 3d_{5/2} and S 2p_{3/2} bands located, respectively, at 229.2 and 162.4 eV, indicating that Mo⁴⁺ and S²⁻ are the dominant oxidation states.³⁵

To fabricate a MIM device structure without the forming of short circuits, we employed an rGO sheet as the top electrode to cover on the pressed MoS₂ nanosphere assemblies. A great number of studies have demonstrated the good conductivity of high temperature annealing-reduced graphene oxide films,^{36,37} and the excellent performances of rGO electrodes used in various electronic devices, such as solar cells,³⁸ FETs,³⁷ and electronic storage,³⁹ have also been reported recently. Herein, using, respectively, rGO and ITO as top and bottom electrodes, we constructed a MoS₂ nanospheres-based diode device, and the assembly procedure is schematically shown in Fig. S1.⁴⁶ Briefly, the GO film was prepared first via spin-coating the GO aqueous solution on a SiO₂/Si substrate similar to the previous method.⁴⁰ Then, the highly reduced GO film was obtained by high temperature (1000°C) annealing and was transferred onto the as-grown MoS₂/ITO by a normal transfer route.⁴¹ Finally,

the adherent the poly(methyl methacrylate) (PMMA) on rGO was removed by using the ultraviolet (UV) irradiation and the developer of isopropyl alcohol (IPA):methyl isobutyl ketone (MIBK) basing on a process proposed by Liu *et al.*³⁹

Fig. 3(a) shows the current–voltage (I–V) characteristic of the as-fabricated rGO/MoS₂/ITO diode, and its schematic was illustrated in right inset. At low voltage, the device exhibits HRS. The current gradually increases with a negatively increased voltage (stage 1). The resistance retains the HRS until the voltage approaches the switching threshold of about −2.0 V, at which the current jumps abruptly from 4.2×10^{-5}

FIG. 2. (a) XPS survey spectrum and high-resolution XPS spectra in (b) Mo 3d and (c) S 2p regions of MoS₂ nanospheres fabricated on ITO.

FIG. 3. (a) Typical I-V curve and schematic illustration (inset) of the rGO/MoS₂/ITO memory device. (b) Retention tests of the ON and OFF states with the ON/OFF ratio of around 10^4 at -1 V.

to the maximum limited current of 0.01 A in our semiconductor characterization system (Keithley 4200) (stage 2), indicating that the resistance switches from HRS to LRS). This SET operation from conductivity OFF to ON states represents a “writing” process in the memory device. The ON state with the high conductivity can remain in the subsequent negative sweep (stage 3) and reverse sweep of positive voltage less than 1.6 V (stage 4), indicating that the written data are of the non-volatile nature in the memory window of -2.0 – 1.6 eV including zero voltage. Interestingly, once the positive sweep

voltage reaches more than 1.6 V, the LRS can be switched back to the HRS (stage 5). This RESET operation from the conductivity ON to OFF states acts as the erasing function of storage data. After the RESET, the device remains in the OFF state in the subsequent sweep of positive voltage less than 6 V (stage 6) and can be reprogrammed to the ON state in the next sweep. The repeatable “SET-RESET” operation was further demonstrated by sweeping the voltage through multiple consecutive cycles in Fig. S2. The capacity of voltage regulating resistance state indicates the achievement of non-volatile memory with erasable and rewritable functions. In addition, we carried out the model-fitting for the experimental I-V characteristics of the device in HRS and LRS, respectively. In the HRS, the I-V data were fitted by the thermionic emission (TE) model⁴² with Eq. (S1) and the space-charge-limited current (SCLC) model³⁹ with Eq. (S2) in Figs. S3(a) and S3(b), respectively. A more fitting linear relation was observed in the plot of $\ln(I)$ vs. $V^{1/2}$, indicating that the conduction mechanism probably arises from the TE model, and the charge injection and accumulation in dielectric nanospheres may be dominant in the OFF state. And, the I-V characteristic in the LRS could be fitted well by an ohmic conduction model⁴³ with Eq. (S3) in Fig. S3(c), indicating definitely that the good ohmic conductive channels are established in the ON state. A constant voltage of 1.0 V was applied to investigate the retention property of the memory cell at room temperature, as shown in Fig. 3(b). Both the ON and OFF states were stable during the test period of 5.5×10^3 s, and the high ON/OFF resistance ratio of $\sim 10^4$ did not decline obviously. The strong retention ability with high ON/OFF ratio between the HRS and LRS is crucial both to preserve persistently the stored information and to restrict the misreading probability in the practical application of memory devices.

Note that the MoS₂ film-based device does not exhibit significantly the RS phenomenon, as shown in Fig. S4, which is in agreement with some recently reported findings.^{31,32} Thus, we consider that the unique shape of nanospheres may play a crucial role in the carrier-transport process, and a general model of electron tunneling across the polarization potential barriers⁴⁴ in dielectric particle networks was proposed accordingly to understand the electrically controlled RS effect observed in our experiment. Fig. 4 shows the schematic, illustrating the electric-field-modulated change of conductive pathway. For the nanospheres assembling network film, the physical contact between the adjacent spheres results in massive grain boundaries and high junction

FIG. 4. Model schematic of the electron tunneling across the junction barriers modulated by electric-field-induced polarization with SET and RESET processes.

barriers, which dominate the electron transport and make the whole diode be in the initial HRS, as shown in Fig. 4(a). Setting the applied electric field to a relatively low value, the charge will gradually inject and accumulate in individual sphere cells following the above mentioned TE model due to the difficulty of electron tunneling in HRS, thus resulting in the formation of polarized domains. Moreover, the increased charge density in spheres reduces the junction-barrier height, allowing easier electron tunneling and transportation, i.e., the conductive pathway is formed along the boundaries of the spheres and the diode is set into the LRS, as illustrated in Fig. 4(b). More importantly, because of the existence of bound charges and internal polarization fields, the conductive pathway can always survive until a reversely applied electric field is high enough to rupture the polarized domains, as shown in Fig. 4(c). Once the polarization charges accumulated near surface of spheres are relaxed, the junction barriers between spheres will resume the natural height, hindering the electron tunneling and transportation. At this point, the conductive pathway is destroyed, as shown in Fig. 4(d), and the diode is reset into HRS. But, it is worth noting that the electric-field-induced polarized domains in tunnel barriers have a certain direction, which is contrary to that of the applied first electric field. Since the vectorial polarization is a collective phenomenon like magnetization, the polarization fields, and bound charges induced by the applied first electric field in dielectric nanospheres cannot indeed be released in full in one operation cycle, as shown in Fig. 4(d). Thus, the reverse RESET voltage can reset the device to the HRS rather than to the original ground state. Fig. S5(a) shows the amplified I-V curve of Fig. 3(a), and we can identify the difference between the initial and the reset OFF states, which is highlighted by a red circle. This indicates that the reset HRS is not the ground state and it may remain still some formerly polarized domains or Schottky barriers, which make the reset device less easy to occur the polarization reversal and resistance-state switching during the last sweep of positive voltage less than 6 V. However, if setting first positive voltage, the polarized domains in opposite direction will grow and remain in the whole operation cycle, then the converse “SET–RESET” process can be manipulated, as shown in Fig. S5(b). Thus, the vectorial polarization induced by the SET field affects the conduction behavior of device in the whole sweeping cycle including in the HRS after RESET operation. In a word, the electric-field-induced polarized domains with varying junction barriers in the nanosphere networks may have an important effect on the formation and rupture of conductive pathway in the bipolar RS behavior. Indeed, it should be noted that the nanoparticles/nanospheres as building blocks have shown the promising applicability for many electronic nanodevices, but the fantastic electron-transport property of their assemblies is still an open issue and needs more researches.^{33,45} The present interpretation of observed electrical behavior of nanosphere assemblies as of memristor is not conclusive and should be explored more accurately by using more profound theories in the future.

In conclusion, we have demonstrated that the MoS₂ nanosphere assemblies fabricated by a simple hydrothermal method can be employed as the active layer for the

nonvolatile memory device. The device exhibits the sustainable conductivity bistability (HRS and LRS) and the typical bipolar RS with low switching voltage (~ 2 V) and high ON/OFF resistance ratio ($\sim 10^4$). A possible model of the electron tunneling across the junction barriers modulated by electric-field-induced polarization was proposed to explain the observed RS behavior, indicating that the unusual electrical properties in the nanosphere or nanoparticle assemblies can expand further the application of relevant nanodevices.

This work was supported by National Natural Science Foundation of China (Nos. 11104240 and 60725413), “973” Program (Nos. 2011CB302004 and 2013CB932903), and Jiangsu Government Scholarship for Overseas Studies in 2013.

- ¹R. Waser and M. Aono, *Nature Mater.* **6**, 833 (2007).
- ²J. J. Yang, M. D. Pickett, X. M. Li, D. A. A. Ohlberg, D. R. Stewart, and R. S. Williams, *Nat. Nanotechnol.* **3**, 429 (2008).
- ³A. Chanthbouala, V. Garcia, R. O. Cherifi, K. Bouzehouane, S. Fusil, X. Moya, S. Xavier, H. Yamada, C. Deranlot, N. D. Mathur, M. Bibes, A. Barthélémy, and J. Grollier, *Nature Mater.* **11**, 860 (2012).
- ⁴L. H. Wang, W. Yang, Q. Q. Sun, P. Zhou, H. L. Lu, S. J. Ding, and D. W. Zhang, *Appl. Phys. Lett.* **100**, 063509 (2012).
- ⁵M. H. R. Lankhorst, B. W. S. M. M. Ketelaars, and R. A. M. Wolters, *Nature Mater.* **4**, 347 (2005).
- ⁶K. Nagashima, T. Yanagida, K. Oka, M. Taniguchi, T. Kawai, J. S. Kim, and B. H. Park, *Nano Lett.* **10**, 1359 (2010).
- ⁷J. Qi, M. Olmedo, J. Ren, N. Zhan, J. Zhao, J. G. Zheng, and J. L. Liu, *ACS Nano* **6**, 1051 (2012).
- ⁸L. He, Z. M. Liao, H. C. Wu, X. X. Tian, D. S. Xu, G. L. W. Cross, G. S. Duesberg, I. V. Shvets, and D. P. Yu, *Nano Lett.* **11**, 4601 (2011).
- ⁹K. Szot, W. Speier, G. Bihlmayer, and R. Waser, *Nature Mater.* **5**, 312 (2006).
- ¹⁰A. Asamitsu, Y. Tomioka, H. Kuwahara, and Y. Tokura, *Nature* **388**, 50 (1997).
- ¹¹D. S. Kim, Y. H. Kim, C. E. Lee, and Y. T. Kim, *Phys. Rev. B* **74**, 174430 (2006).
- ¹²M. Wuttig, *Nature Mater.* **4**, 265 (2005).
- ¹³Y. Li, Y. P. Zhong, L. Xu, J. J. Zhang, X. H. Xu, H. J. Sun, and X. S. Miao, *Sci. Rep.* **3**, 1619 (2013).
- ¹⁴Z. M. Liao, C. Hou, Q. Zhao, D. S. Wang, Y. D. Li, and D. P. Yu, *Small* **5**, 2377 (2009).
- ¹⁵S. H. Jo, K. H. Kim, and W. Lu, *Nano Lett.* **9**, 870 (2009).
- ¹⁶T. Lee and Y. Chen, *MRS Bull.* **37**, 144 (2012).
- ¹⁷Z. M. Liu, A. A. Yasseri, J. S. Lindsey, and D. F. Bocian, *Science* **302**, 1543 (2003).
- ¹⁸M. J. Rozenberg, I. H. Inoue, and M. J. Sanchez, *Appl. Phys. Lett.* **88**, 033510 (2006).
- ¹⁹E. Y. Tsybmal and H. Kohlstedt, *Science* **313**, 181 (2006).
- ²⁰H. Kohlstedt, N. A. Pertsev, J. R. Contreras, and R. Waser, *Phys. Rev. B* **72**, 125341 (2005).
- ²¹D. Lee, D. Seong, I. Jo, F. Xiang, R. Dong, S. Oh, and H. Hwang, *Appl. Phys. Lett.* **90**, 122104 (2007).
- ²²K. Lee, H. Kim, M. Lotya, J. Coleman, G. Kim, and G. Duesberg, *Adv. Mater.* **23**, 4178 (2011).
- ²³H. Qiu, L. Pan, Z. Yao, J. Li, Y. Shi, and X. Wang, *Appl. Phys. Lett.* **100**, 123104 (2012).
- ²⁴B. Radisavljevic, M. B. Whitwick, and A. Kis, *ACS Nano* **5**, 9934 (2011).
- ²⁵Z. Yin, H. Li, H. Li, L. Jiang, Y. Shi, Y. Sun, G. Lu, Q. Zhang, X. Chen, and H. Zhang, *ACS Nano* **6**, 74 (2012).
- ²⁶W. Choi, M. Y. Cho, A. Konar, J. H. Lee, G. B. Cha, S. C. Hong, S. Kim, J. Kim, D. Jena, J. Joo, and S. Kim, *Adv. Mater.* **24**, 5832 (2012).
- ²⁷G. Eda, T. Fujita, H. Yamaguchi, D. Voiry, M. Chen, and M. Chhowalla, *ACS Nano* **6**, 7311 (2012).
- ²⁸J. T. Ye, Y. J. Zhang, R. Akashi, M. S. Bahramy, R. Arita, and Y. Iwasa, *Science* **338**, 1193 (2012).
- ²⁹H. Qiu, T. Xu, Z. Wang, W. Ren, H. Nan, Z. Ni, Q. Chen, S. Yuan, F. Miao, F. Song, G. Long, Y. Shi, L. Sun, J. Wang, and X. Wang, *Nat. Commun.* **4**, 2642 (2013).

- ³⁰M. S. Choi, G. H. Lee, Y. J. Yu, D. Y. Lee, S. H. Lee, P. Kim, J. Hone, and W. J. Yoo, *Nat. Commun.* **4**, 1624 (2013).
- ³¹S. Bertolazzi, D. Krasnozhan, and A. Kis, *ACS Nano* **7**, 3246 (2013).
- ³²Z. Y. Yin, Z. Y. Zeng, J. Q. Liu, Q. Y. He, P. Chen, and H. Zhang, *Small* **9**, 727 (2013).
- ³³T. H. Kim, E. Y. Jang, D. J. Lee, K. J. Lee, J. T. Jang, J. S. Choi, S. H. Moon, and J. Cheon, *Nano Lett.* **9**, 2229 (2009).
- ³⁴Y. G. Li, H. L. Wang, L. M. Xie, Y. Y. Liang, G. S. Hong, and H. J. Dai, *J. Am. Chem. Soc.* **133**, 7296 (2011).
- ³⁵W. J. Zhou, Z. Y. Yin, Y. P. Du, X. Huang, Z. Y. Zeng, Z. X. Fan, H. Liu, J. Y. Wang, and H. Zhang, *Small* **9**, 140 (2013).
- ³⁶H. A. Becerril, J. Mao, Z. Liu, R. M. Stoltenberg, Z. Bao, and Y. Chen, *ACS Nano* **2**, 463 (2008).
- ³⁷S. P. Pang, H. N. Tsao, X. L. Feng, and K. Müllen, *Adv. Mater.* **21**, 3488 (2009).
- ³⁸Q. Liu, Z. F. Liu, X. Y. Zhang, N. Zhang, L. Y. Yang, S. G. Yin, and Y. S. Chen, *Appl. Phys. Lett.* **92**, 223303 (2008).
- ³⁹J. Q. Liu, Z. Y. Yin, X. H. Cao, F. Zhao, L. H. Wang, W. Huang, and H. Zhang, *Adv. Mater.* **25**, 233 (2013).
- ⁴⁰X. Zhou, X. Huang, X. Qi, S. Wu, C. Xue, F. Boey, Q. Yan, P. Chen, and H. Zhang, *J. Phys. Chem. C* **113**, 10842 (2009).
- ⁴¹K. S. Kim, Y. Zhao, H. Jang, S. Y. Lee, J. M. Kim, K. S. Kim, J. Ahn, P. Kim, J. Choi, and B. Hong, *Nature* **457**, 706 (2009).
- ⁴²S. M. Sze, *Physics of Semiconductor Devices* (Wiley, New York, 1981).
- ⁴³X. Zhuang, Y. Chen, G. Liu, P. Li, C. Zhu, E. Kang, K. Noeh, B. Zhang, J. Zhu, and Y. Li, *Adv. Mater.* **22**, 1731 (2010).
- ⁴⁴X. T. Li, D. J. Bergman, and D. Stroud, *Europhys. Lett.* **69**, 1010 (2005).
- ⁴⁵M. Chen, L. F. Hu, J. X. Xu, M. Y. Liao, L. M. Wu, and X. S. Fang, *Small* **7**, 2449 (2011).
- ⁴⁶See supplementary material at <http://dx.doi.org/10.1063/1.4862755> for the fabrication procedure, the SEM image, and the original data and the model-fitting data of I-V measurement of the rGO/MoS₂ nanospheres/ITO memory device.