Provided for non-commercial research and education use.
Not for reproduction, distribution or commercial use.

This article appeared in a journal published by Elsevier. The attached copy is furnished to the author for internal non-commercial research and education use, including for instruction at the authors institution and sharing with colleagues.

Other uses, including reproduction and distribution, or selling or licensing copies, or posting to personal, institutional or third party websites are prohibited.

In most cases authors are permitted to post their version of the article (e.g. in Word or Tex form) to their personal website or institutional repository. Authors requiring further information regarding Elsevier's archiving and manuscript policies are encouraged to visit:

http://www.elsevier.com/copyright

Author's personal copy

Thin Solid Films 518 (2010) S35–S37

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Study of the effects of growth temperature and time on the alignment of Si quantum dots on hafnium oxide coated single wall carbon nanotubes

Mario Olmedo [a], Alfredo A. Martinez-Morales [a], Gang Liu [b], Emre Yengel [c], Cengiz S. Ozkan [c], Chun Ning Lau [b], Mihrimah Ozkan [a], Jianlin Liu [a,*]

[a] *Department of Electrical Engineering, University of California, Riverside, CA 92521, United States*
[b] *Department of Physics and Astronomy, University of California, Riverside, CA 92521, United States*
[c] *Department of Mechanical Engineering, University of California, Riverside, CA 92521, United States*

ARTICLE INFO

Available online 19 October 2009

PACS:
64.75.Yz

Keywords:
Self-assembly
Carbon Nanotubes
Quantum dots

ABSTRACT

The alignment of quantum dots (QDs) has been a complicated exercise of combining lithography with deposition or growing QD superlattices. In this work, we demonstrate the aligned growth of epitaxial Si QDs on 1-dimensional (1-D) HfO_2 ridges. These ridges were created by the growth of HfO_2 thin film on single wall carbon nanotubes (SWCNT) parallel to the substrate surface. The reason for the QD alignment on the HfO_2 thin film is in the strain that is built at the top of the 1-D ridges, which favors the formation of Si seeds over the surrounding flat HfO_2 area. Periodic alignment of Si QDs on the 1-D HfO_2 ridge was observed. A study over the effects of different growth conditions, such as growth temperature, and growth time is presented. The size of the QDs is found to be directly related to the size of the ridge.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

The study of the growth of QDs on a semiconducting or insulating surface has been of interest from many scientists in different fields. Ge dots on Si substrates showed potential applications in the electronic and optoelectronic fields [1], GaAs dots have shown laser emission [2,3], Si dots can be used for charge storage [4,5], etc. Most of these QDs are randomly distributed and how to control the location of these dots were intensively pursued, such as using photolithographic patterns to facilitate QD alignment, using the step of the substrate as the alignment pattern, or doing QD superlattices. Here we present a technique that uses a bottom up approach to align QDs on an oxide surface that requires no lithographic patterns on the surface. We use CNTs to be our template and the HfO_2 as the top surface for the alignment of Si QDs.

The template consists of SWCNTs on a SiO_2 surface covered by a HfO_2 thin film formed by atomic layer deposition (ALD). The SWCNTs provide their 1-D shape and nanometer scale diameter, to form a ridge in the HfO_2 thin film. This ridge is close to the dimensions of the CNT which enables applications based on this structure to go beyond the complementary metal oxide semiconductor (CMOS) ultimate limit. Our experiment followed the technique of selective area epitaxy [6,7] which was reported on the alignment of Ge QDs on Si ridges at a much larger scale. That experiment started by fabricating a patterned template via top down photolithography, followed by a self assembled growth of QDs. The self assembly mechanism requires a smooth substrate surface so that during growth the adatoms will be able to migrate to the pre-patterned lowest energy spots. Single crystal surfaces have been commonly applied for the alignment of QDs via the selective epitaxial growth. Now with the demonstration of QD alignment on an oxide surface we will be able to use aligned QDs for possible nonvolatile nanocrystal memory applications. The ALD technique which is used to cover the CNTs can produce thin oxide films with smooth conformal polycrystalline surfaces for subsequent QD self assembly [8].

2. Experiment

We first grew SWCNTs on SiO_2/Si substrates using a chemical vapor deposition technique [9,10]. The CNT covered substrates were then subjected to an UV cleaning for 5 min to make the surface of the substrate hydrophilic. This time interval was adjusted to ensure good water adhesion to the surface as well as leaving the CNTs unperturbed. The samples were then introduced to a Cambridge Nanotech Savannah 100 atomic layer deposition chamber. The system ran at a partial pressure of 3×10^{-1} Torr, and a deposition time of 5 s was used for both precursor and source. The temperature was kept at a constant 250 °C. For Si QD deposition a custom built gas source molecular beam epitaxy (GSMBE) system was employed. This system points the source, in this case disilane (Si_2H_6), directly at the substrate ensuring direct impingement. The chamber base pressure was in the order of 10^{-8} Torr, while growth pressure was in the range of 10^{-5} Torr. All the samples were heated via a Ta heating coil behind the substrate; while a thermocouple situated between the heater and substrate was used to measure the

* Corresponding author.
E-mail address: jianlin@ee.ucr.edu (J. Liu).

0040-6090/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.tsf.2009.10.050

Author's personal copy

desired temperature. The disilane gas flow was controlled using a mass flow controller (MFC) UFC 1660. Atomic force microscopy (AFM) characterization was carried out using a Veeco multimode AFM.

3. Results

Fig. 1(a) shows the schematic of a cross sectional view of a QD grown on top of HfO_2 covered CNTs. The QDs are drawn preferentially to the 1-D HfO_2 ridge formed by the CNTs underneath the film. This preference does not stop other QDs to be formed on the surrounding flat surface. This event can be controlled by tuning the magnitude of the length of Si adatom migration on HfO_2 and the proximity of adjacent CNTs. Fig. 1(b) shows a cross sectional SEM image of aligned Si QDs on top of a HfO_2 covered CNT. The HfO_2 thickness is 6 nm and the ridge height was measured to be 1 nm to 1.2 nm by AFM characterization. Si QDs are clearly observed to align at the top of the 1-D HfO_2 ridge, while some others are also seen on the rest of the surrounding oxide. The Si QDs along the CNT are larger, demonstrating that a greater quantity of Si adatoms preferentially settled on the 1-D ridge. We think that if the quantity of CNTs is increased, selective growth of Si QDs only on the ridges formed by the CNTs will be evident. A reference sample, which has gone through the same substrate temperature cycle process but without the introduction of disilane was prepared, showing that the HfO_2 was stable at the high growth temperatures without forming HfO_2 grains. This result is shown elsewhere [11]. The SEM image can also confirm that the HfO_2 layer is highly conformal on the SiO_2 surface. The CNT surface however is not chemically active [12] due to its lack of dangling bonds so fewer layers of oxide are deposited over the ridge than the rest of the film.

Fig. 2(a)–(d) shows AFM images of the samples with different disilane growth time. The formation of a QD starts with a Si atom seed. From this seed more Si atoms attach to form a QD. High strain areas like the CNT ridges have been known to attract Si seed in patterned HfO_2 films [10]. As the growth temperature increases, the dot density increases. This is reasonable because as the first Si QDs forms, they relax the strain that initially produced them. This point of relaxation is decided by the size of the ridge itself, since it defines the size of the QD base. Then secondary QDs form elsewhere around the ridge increases the density. As these secondary QDs also relax the surface of the ridge,

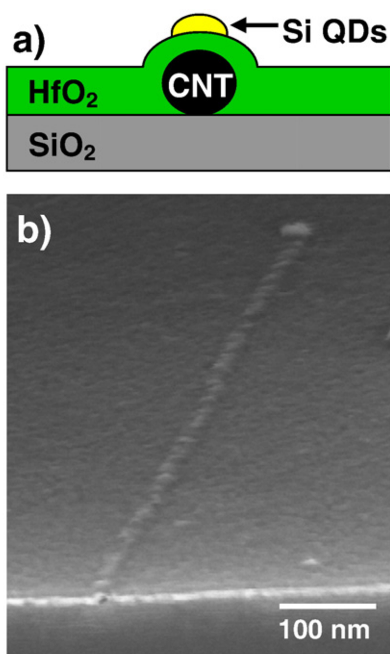

Fig. 1. QDs on a HfO_2 covered CNT. (a) Schematic of a cross sectional view of the structure. (b) SEM cross sectional image shows aligned Si QDs on a HfO_2 covered CNT.

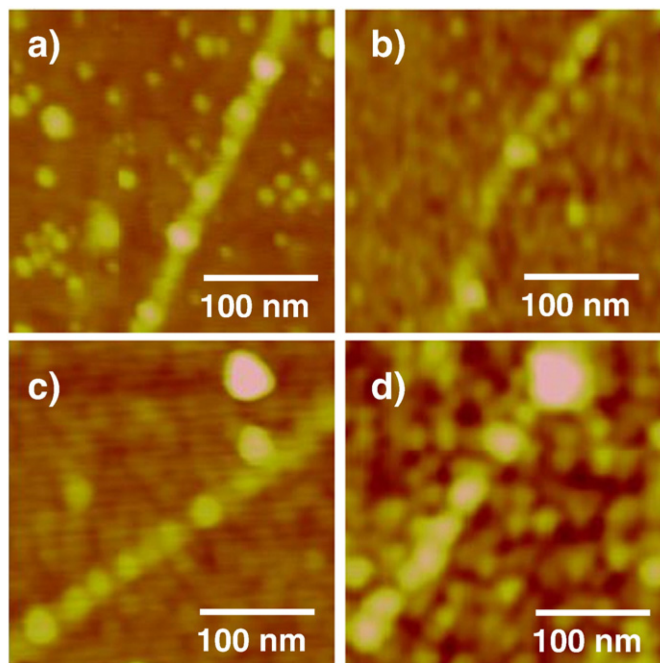

Fig. 2. AFM images of four Si QD alignment samples that have different disilane growth times: a) 3 min, b) 5 min, c) 7 min, d) 8 min.

growth in other parts of the substrate is accelerated. Further increase of growth time reduces the QD density as the dots on the 1-D ridge start to merge with each other.

Fig. 3(a)–(d) shows the changes in QD morphology due to growth temperatures. It is noticed that the change in temperature has an indistinguishable effect on QD density. Since growth temperature normally changes the adatom diffusion length [13,14], for the temperature range between 640 °C and 680 °C for which there is alignment, the diffusion length is large enough for Si adatoms to reach preferential nucleation sites on 1-D HfO_2 ridges on CNTs and form QDs.

Oxide thickness is also one of the deciding factors for QD alignment. Surface strain on ALD grown films changes depending on their thickness on CNTs. The samples with increased oxide thickness are shown in Fig. 4(a)–(c). With an increase in oxide thickness there is a clearer

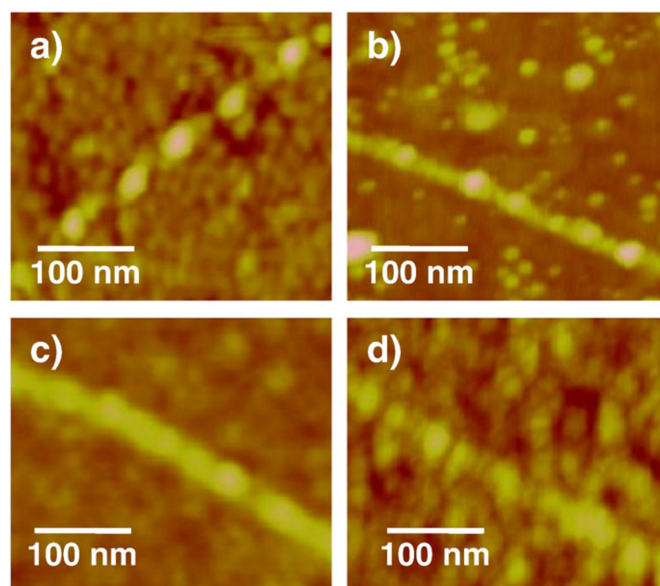

Fig. 3. AFM images of four Si QD alignment samples that have different growth temperatures: a) 640 °C, b) 650 °C, c) 670 °C, d) 680 °C.

Author's personal copy

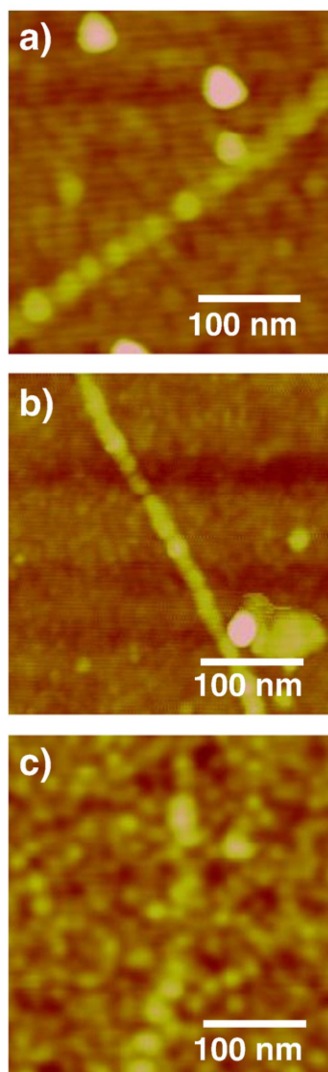

Fig. 4. AFM images of three Si QD alignment samples that have different HfO_2 thickness: a) 6 nm, b) 7 nm, c) 7.6 nm.

formation of QDs around the CNT ridge. The decrease in preferentiality of nucleation on 1-D ridges is due to the relaxation of the strain in the CNT ridge. With less selectivity toward the ridge, the Si adatoms randomly settle on the flat areas of the sample. It takes 6 nm of HfO_2 to fully cover these randomly grown CNTs even though ALD is conformal to most surfaces. This is due to the CNT's inertness to the H_2O precursor [12]. Therefore the thickness of the HfO_2 layer is needed to be thicker than the CNTs themselves to completely cover the CNT and have a smooth surface for Si adatom migration. The window for alignment starts close to 6 nm, and after 1 to 2 nm depending on the size of the ridge sharper Si growth outside the CNT starts to become evident.

In summary, we have studied the growth time and temperature effect on aligned Si QDs on HfO_2 covered CNTs. Alignment has been shown to be evident and HfO_2 is proved to be a viable surface for Si adatom migration. We found that growth time changes the density of Si QDs and that temperature is also crucial due to its effect on the diffusion length. We observed that the QD base size is dictated by the size of the ridge, which in turn is caused by the size of the CNT. In addition, the oxide thickness can be tuned to control the strain in the ridges. With this structure as the base for future transistor technologies such as nanocrystal memory, the goal to go beyond CMOS ultimate limit is a closer possibility.

Acknowledgements

The authors acknowledge the financial and program support of the Focus Center Research Program (FCRP) on FENA and the National Science Foundation (NSF) (DMR 0807232). CSO acknowledges support by NSF CMMI Award 0800680, CSO and MO acknowledge support from the NSF Center for Hierarchical Manufacturing (CHM). CNL and GL acknowledge support by NSF CAREER DMR/0748910 and ONR/DMEA Award H94003-07-2-0703.

References

[1] O.P. Pchelyakov, Y.B. Bolkhovityanov, A.V. Dvurechenskii, A.I. Nikiforov, A.I. Yakimov, B. Voigtlander, Think Solid Films 367 (2000) 75.
[2] M. Asada, Y. Miyamoto, Y. Suematsu, IEEE J. Quantum Electron. 22 (1986) 1915.
[3] N. Kirstaedter, O. Schmidt, N. Ledenstov, D. Bimberg, V. Ustinov, A. Egorov, A. Zhukov, M. Maximov, P. Kop'ev, Zh.I. Alferov, Appl. Phys. Lett. 69 (1996) 1226.
[4] D.W. Kim, T. Kim, S.K. Banerjee, IEEE Trans. Electron Devices 50 (2003) 1823.
[5] J.J. Lee, X. Wang, W. Bai, N. Lu, D.L. Kwong, IEEE Trans. Electron Devices 50 (2003) 2067.
[6] G. Jin, J.L. Liu, S.G. Thomas, Y.H. Luo, K.L. Wang, B.Y. Nguyen, Appl. Phys. Lett. 75 (1999) 2752.
[7] G. Jin, J.L. Liu, K.L. Wang, Appl. Phys. Lett. 76 (2000) 3591.
[8] S.S. Coffee, J.G. Ekerdt, J. Appl. Phys. 102 (2007) 4912.
[9] S. Han, X. Liu, C. Zhou, J. Am. Chem. Soc. 127 (2005) 5294.
[10] P. Jarillo-Herrero, J.A. Van Dam, L.P. Kouwenhoven, Nature 439 (2006) 953.
[11] M. Olmedo, A. Martinez-Morales, G. Liu, E. Yengel, C.S. Ozkan, C.N. Lau, M. Ozkan, J.L. Liu, Appl. Phys. Lett. 94 (2009) 123109.
[12] A. Javey, J. Guo, D.B. Farmer, Q. Wang, E. Yenilmez, R.G. Gordon, M. Lundstrom, H. Dai, Nano Lett. 4 (2004) 1319.
[13] J. Tershoff, C. Teichert, M.G. Lagally, Phys. Rev. Lett. 76 (1996) 1675.
[14] Q. Xie, A. Madhukar, P. Chen, N.P. Kobayashi, Phys. Rev. Lett. 75 (1995) 2542.