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2010 Nanotechnology 21 165302

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Nanotechnology **21** (2010) 165302 (5pp) doi:10.1088/0957-4484/21/16/165302

Self-assembled Bi interconnections produced by on-film formation of nanowires for *in situ* device fabrication

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Received 14 January 2010, in final form 2 March 2010
Published 26 March 2010
Online at stacks.iop.org/Nano/21/165302

Abstract

We fabricated Bi nanowire interconnections between two pre-patterned electrodes using a combination of on-film formation of nanowires (OFF–ON) and self-assembly. Bi nanowires were found to grow laterally from a multilayer structure with a Cr (or SiO_2) overlayer on top of a Bi thin film through thermal annealing to relieve vertically stored compressive stress. A Bi nanobridge with a diameter of 192 nm was formed between two Cr electrodes and was highly ohmic according to I–V measurements. A high transverse magnetoresistance of 123% was also observed at 300 K. Our results indicate that self-assembled lateral nanowire growth can be utilized as an easy means for fabricating a variety of nanowire devices without the use of catalysts or complex patterning processes.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Nanowires and nanotubes have attracted growing interest as one-dimensional candidate structures for bottom-up implementation of ever-shrinking nanoelectronics. The bottom-up approach offers much promise for future integrated circuits in that the conventional top-down approaches will face a scaling limit beyond a certain feature size [1, 2]. The self-assembly of various structures is a critical path for success in the bottom-up approach. However, the method struggles with precise control in position and shape because bottom-up grown structures are generally irregular in character. A nanobridge is a typical nanostructure that can be used as a fundamental lateral interconnect, and these structures ideally can be produced by self-assembly methods. A variety of approaches have been pursued for nanobridge or nanowire interconnection, including SiO_2 patterning and cracking [1], self-assembly on Si substrates by a thermal evaporation method [3, 4], field emission induced growth using a scanning tunneling microscopy tip [5], electric field assisted growth [6, 7], bridging from each patterned catalyst island [8], selective bridging on granular films [9], and patterning by e-beam lithography [10]. However, none of these methods are simple or cost effective.

Bismuth (Bi) is intrinsically a semimetal with interesting properties, such as a highly anisotropic Fermi surface, low carrier concentration, small effective mass ($0.001m_0$–$0.26m_0$) [11], and long mean free path ($\sim$1.35 μm) of the carriers [12]. Due to these attractive attributes, Bi has been extensively investigated, particularly in order to understand quantum transport phenomena, finite size effects, and magnetoresistance effects [13–15]. High quality Bi nanowires are essential for studying the properties of Bi-based one-dimensional structures in depth.

Zhang *et al* [16] synthesized Bi nanowires in porous anodic alumina using a high pressure liquid injection technique, which was similar to their prior work but employed a surfactant to lower the pressure required for filling the pores. A vapor phase preparation technique was also developed in order to prepare a Bi nanowire array, making possible small nanowires with diameters as small as 7 nm [17]. Furthermore, the native oxide layer etching needed to fabricate Bi nanowire devices, which is crucial to making ohmic contacts on the nanowire devices, was found to be challenging.

In this paper we introduce a self-assembled interconnection of nanostructures produced by an on-film formation

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of nanowires (OFF–ON) method in order to form a highly ohmic Bi nanobridge. Here, OFF–ON is a stress induced nanowire growth technique activated by a thermodynamic driving force caused by compressive stress accumulated in a film, arising from a difference in thermal expansion between the film and the substrate [12, 18]. Transport properties are subsequently presented for the individual two-probe Bi nanobridge devices that were fabricated without a surface oxide etching process. This is the first demonstration of the feasibility of Bi nanodevices fabricated by self-assembly.

2. Experimental details

A Bi thin film was first deposited on a thermally oxidized Si(100) substrate at a rate of 40 Å s^{-1} by radio frequency (RF) sputtering at 300 K. The sputter system was kept in an ultrahigh vacuum (UHV) of 10^{-6} Torr before deposition, and sputtering was performed under an Ar gas pressure of 2 mTorr for 180 s. For the lateral growth of Bi nanowires, we sputtered a thin Cr (or SiO_2) layer on top of the Bi film. The Bi thin films were subsequently put into a custom-made vacuum furnace for thermal annealing in order to grow Bi nanowires by the OFF–ON method. Annealing was undertaken under an UHV of 10^{-6} Torr at 250 °C for 5–10 h. In the layer structure used, Bi nanowires cannot be grown vertically due to the difficulty of punching through the top Cr (or SiO_2) layer. Instead, laterally grown Bi nanobridges were formed almost exclusively across the two distant nanopatterns, which had Cr electrodes on top. The current–voltage (I–V) characteristics and the variation in the transverse magnetoresistance (MR) for a 600 nm diameter Bi nanobridge grown thus were analyzed using a Keithley 236 Source-Measure Unit and 2182 Multimeter with a Physical Property Measurement System (PPMS).

3. Results and discussion

The motivation for this research was a desire to laterally grow Bi nanowires between confined regions using a bilayer structure of Cr (or SiO_2)/Bi films to eliminate the need for a metal catalyst, required in previous approaches to define such interconnecting structures, and eliminate post-patterning of electrodes.

Figure 1 schematically illustrates the process of growth of the Bi nanowire between Cr (or SiO_2)/Bi patterns by the on-film formation of nanowires (OFF–ON) method. The schematic diagram shows the origin and driving force for the spontaneous growth of Bi nanowires. Without an overlayer, annealing a Bi thin film induces compressive stress acting in the plane, which originates from a mismatch in thermal expansion of the film and the substrate. This mismatch is attributed to the large difference in thermal expansion coefficients between Bi (13.4×10^{-6} °C^{-1}) and SiO_2/Si ((0.5×10^{-6} °C^{-1})/(2.4×10^{-6} °C^{-1})), and acts as a thermodynamic driving force for spontaneous nanowire growth. Bi nanowires grow normal to the plane to relieve the compressive stress through atomic diffusion. The situation becomes different, however, if the Bi film is capped with a Cr (or SiO_2) overlayer.

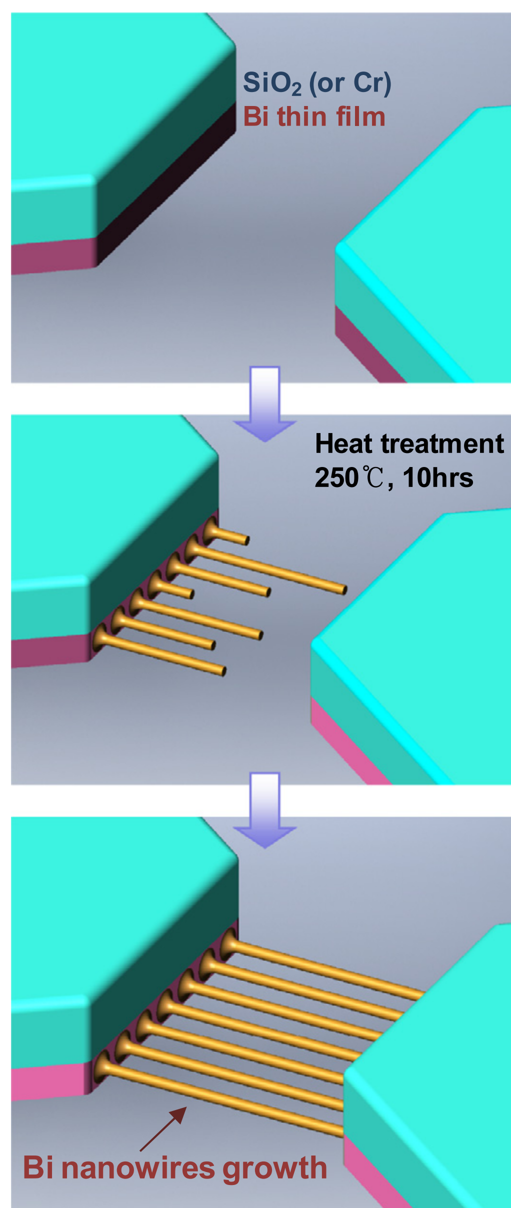

Figure 1. Schematic pictures of Bi nanobridge formation. Step 1: pre-patterned structures of a Cr/Bi bilayer on a thermally oxidized Si substrate. Step 2: initiation of lateral Bi nanowire growth caused by a thermal expansion mismatch between Cr, Bi, and the substrate during annealing at 250 °C for 10 h. Step 3: formation of Bi nanobridges.

In this case, the Bi film is sandwiched between two layers with low thermal expansion coefficients, leading to a second compressive stress that acts perpendicular to the plane. Taking into account that thermal expansion is a three-dimensional phenomenon and that the Bi film thickness (∼600 nm) is much smaller than the planar area (1.25 × 1.25 cm^2), this out-of-plane stress should be overwhelming its in-plane counterpart. Moreover, the overlayer (Cr or SiO_2) blocks any stress-relieving atomic diffusion normal to the plane. Therefore, Bi nanowires should grow laterally with diameters larger than those of vertical nanowires since more accumulated stress should be released in a reduced dimension. This process is depicted in the middle and bottom pictures of figure 1.

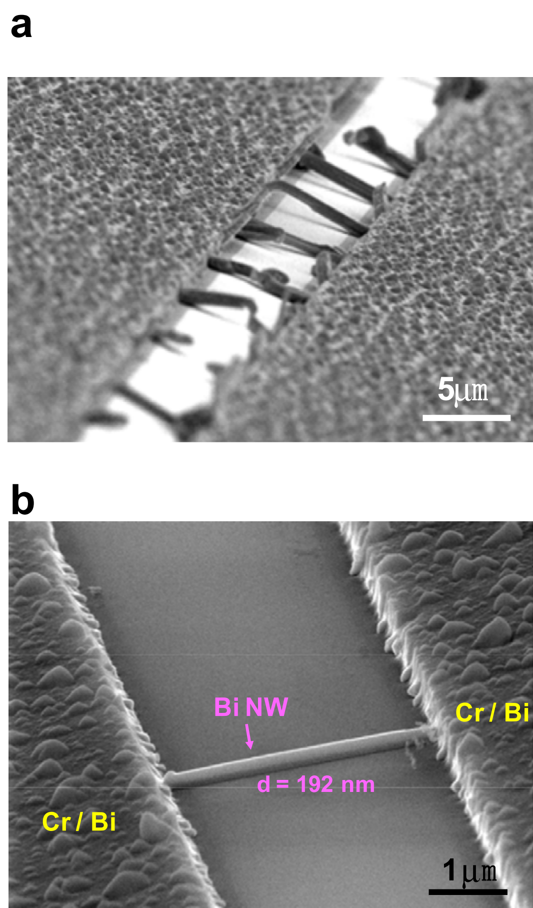

Figure 2. Scanning electron microscope (SEM) images of (a) multiple nanobridges formed between two adjacent electrode patterns and (b) a Bi nanobridge ($d = 192$ nm) formed between two Cr/Bi electrodes with 4 μm spacing.

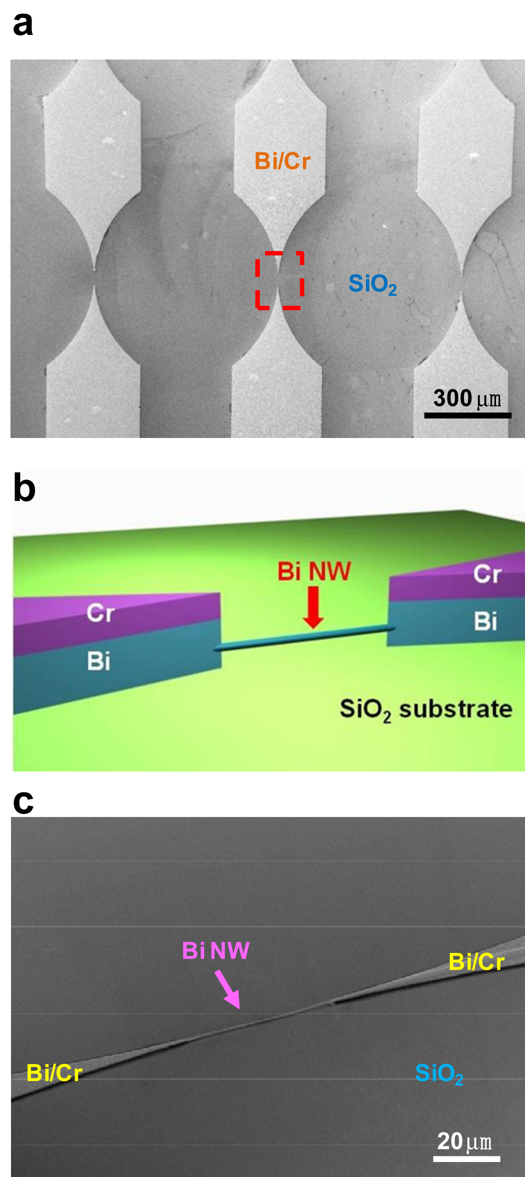

Figure 3. (a) Scanning electron microscope (SEM) image of patterns for the growing Bi nanobridge. (b) Schematic and (c) SEM images of an individual Bi nanobridge.

We did indeed observe lateral growth of Bi nanowires. Figures 2(a) and (b) exhibit representative field emission scanning electron microscope (FE-SEM) images of the Bi nanobridges grown. Figure 2(a) presents multiple nanobridges formed through a 1 μm deep, 4 μm wide trench. All the nanobridges appeared to have a cylindrical shape with a uniform diameter over the entire length. The Bi nanobridges were synthesized under optimized conditions (250 °C, 10 h) and were found to laterally connect the two electrode patterns, which were 5 μm apart. Thus, it is evident that Bi nanobridges can be directly formed in a self-assembly manner across the pre-patterned electrodes on a thermally oxidized Si substrate. This is done using only a single sputtering and annealing step without the use of metal catalysts or metal gas. The nanobridges tended to form on the sidewalls of the Cr/Bi patterns during annealing in a vacuum furnace (10^{-6} Torr), and the Cr overlayer acts as a Bi diffusion barrier in the vertical direction, as mentioned above. With increased annealing time, the nanowires grow longer and come into contact with the opposing vertical surface to form nanobridges (see figure 2(a)). Due to this dependence of the nanowire length on the annealing time, the nanobridges appeared to form with a slightly higher yield when annealed for 10 h, compared to 5 h annealing, even at the fixed temperature of 250 °C. In a previous study, we found that the diameter of a Bi nanowire depends on the mean grain size of the as-grown film, which in turn is determined by the thickness of the film. This result indicates that the Bi nanowire diameter is strongly coupled to the grain size of the film. The smallest diameter of a lateral Bi nanowire observed was 190 nm, which was achieved by tuning the thickness of the Bi thin film grown at room temperature. Owing to the larger accumulated stress (explained above), this diameter is twice as large as the smallest value (98 nm) for universal vertical Bi nanowires.

For transport measurements on an individual Bi nanobridge, the pattern shape was controlled. The patterns with sharp tips were fabricated to connect an individual Bi nanobridge to two electrodes at both ends (see figure 3(a)). The patterns can restrict the nanowire growth position to the sharp tips because the compressive stress concentrates

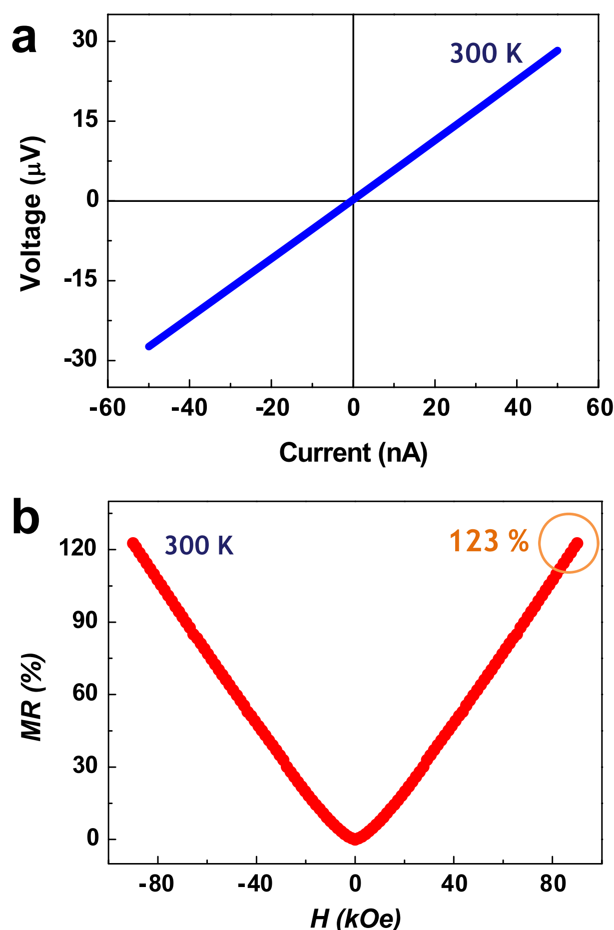

Figure 4. (a) I–V curve recorded at room temperature. The resistivity is 3.6×10^{-4} Ω cm. (b) Transverse magnetoresistance (MR) for a 600 nm diameter Bi nanobridge, reaching 123%.

at the tips, and the tips are in a high energy state due to the high surface-to-volume ratio. Figure 3(b) schematically shows the Bi nanobridge formed between two sharp tips of Cr/Bi electrode patterns facing each other. We confirmed that the Bi nanobridge spanned from tip to tip through the OFF–ON process, as shown in figure 3(c). A typical I–V characteristic for this Bi nanobridge is shown in figure 4(a). From the linearity of the I–V characteristic, it is seen that contacts between Cr electrodes and a Bi nanobridge are highly ohmic at 300 K, resulting in a nanobridge resistivity (ρ) of 3.6×10^{-4} Ω cm. The resistivity of the individual Bi nanobridge appears to be constant, although it was measured by the two-point method, wherein avoiding a contact resistance contribution is difficult. Recollecting that Bi nanowires have an oxide layer on their outer surfaces, this constant resistivity of the Bi nanobridge indicates that contacts to the self-assembled nanobridge are well made even without the oxide layer etching process. Figure 4(b) shows the variation in transverse magnetoresistance (MR) for a 600 nm diameter Bi nanobridge. Here, MR is defined as $\Delta R(H)/R(0) = [R(H) - R(0)]/R(0)$, where $R(0)$ is the zero-field resistance, and $R(H)$ the resistance at a given magnetic field, H. In this work, a large transverse MR = 123% was observed at 300 K, as shown in figure 4(b). The transverse MR is caused by distortion of the electron trajectory by an applied magnetic field and tends to be inversely proportional to the carrier density [16]. Unlike normal metals, the carrier concentration in semimetallic Bi is very low and becomes even lower in high quality Bi nanowires, leading to a large MR. These results indicate that the Bi nanobridges fabricated by the combination of OFF–ON and self-assembly are high quality nanowires capable of good contact with metal electrodes. To our knowledge, this is the first successful demonstration of self-assembled Bi nanobridge synthesis without the aid of position-controlling catalysts. Further studies on an individual Bi nanobridge will be necessary to investigate the transport properties, such as the temperature-dependent mobility and carrier concentration, in more detail.

4. Conclusions

We demonstrated *in situ* Bi nanowire device fabrication by self-assembled interconnection using on-film formation of nanowires (OFF–ON). A Bi thin film was first deposited on a thermally oxidized Si(100) substrate by radio frequency (RF) sputtering. For the lateral growth of Bi nanowires, we sputtered a thin Cr (or SiO_2) layer on top of the Bi film. Through thermal annealing, Bi nanowires grow laterally to relieve a vertically stored large compressive stress. We fabricated a self-assembled Bi nanobridge (d = 192 nm) device *in situ* using OFF–ON through annealing at 250 °C for 10 h. From I–V measurements taken for the Bi nanobridge device, contacts to the nanobridge were found to be highly ohmic. The quality of the Bi nanobridge was also proved by the high MR of 123% obtained from transverse MR measurements. These results manifest the possibility of self-assembled nanowire interconnection between various nanostructures for a variety of applications and provide a simple device fabrication method for investigating transport properties for nanowires without complex patterning and etching processes.

Acknowledgments

This work was supported by Priority Research Centers Program (2009-0093823) through the National Research Foundation of Korea (NRF), and by a grant from the 'Center for Nanostructured Materials Technology', under the '21st Century Frontier R&D Programs' of the Ministry of Education, Science, and by a grant from the Fundamental R&D Program for Core Technology of Materials funded by the Ministry of Knowledge Economy, Republic of Korea, and by the Seoul Research and Business Development Program (10816).

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