Multi-dimensional Nanostructured Oxide Devices

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1. Introduction

The construction of nano-structures represents an important technology in investigations concerning novel quantum physics and in the fabrication of highly integrated devices. In the semiconductor and metal devices fields, the physical properties of nano-structures has been investigated using lithography techniques and has led to the construction of devices including LSI. Transition metal oxides exhibit a broad spectrum of characteristics including ferromagnetism, ferroelectricity and superconductivity based on phase transition. Recent studies have suggested that the ground state of transition metal oxides tends to be intrinsically inhomogeneous, including nano scale novel electric phases. Therefore, preparing low dimensional confined nano-space is quite interesting to design high performance functional oxide devices. A novel process to fabricate integrated functional oxide nano-box arrays, nano-wire and nano-dot is developed. This method can fabricate highly integrated 3D functional oxide nanostructures with sub-10-nm resolution and high aspect ratio and have a great tuneability in feature shapes, sizes and materials through different combination of imprint molds and target materials of physical vapor deposition [1-5]. Sidewall deposition using pulsed laser deposition onto nano-scale integrated resist templates prepared by nano-imprint lithography enables us to prepare Au, Mo, ZnO, (Fe,Zn)₃O₄ hollow nano box whose wall thickness can be systematically controlled from 100nm down to several ten nm [6,7]. Here, we introduce the recent progress in multi-dimensional nanostructured functional oxide and their unique physical properties and functionalities.

2. Experimental set up

Figures 1(a)-(f) show schematics of the process for fabricating of integrated functional oxide (or metal) 3D nano-box arrays or 1D nano-wire arrays. First bilayer resist was spin-coated on substrate for UV-NIL process (step (a)). A typical process of ultraviolet UV- NIL, a low-viscosity and photocurable resist is pressed by a quartz mold and physically deformed following the topology of the mold. The resist is cured by exposure to UV light and finally producing a uniform and rigid polymer network. During UV-NIL process, a top layer filled into inside of hole-patterns and polymerized (step (b)). The imprinted patterns on top layers were transferred to bottom layers by using a RIE (reactive ion etching) process (step (c)). The residual top layer was removed by CF_4 plasma and the bottom layer was removed by following O_2 plasma. Functional oxide (or metal) was deposited by pulsed laser deposition (or a RF sputtering [6]) method, which could control the sidewall thickness (step (d)). Ion milling process removed top and bottom layers except sidewalls (step (e)). To remove residual polymers, a cleaning process was applied (step (f)), and post annealing was conducted for oxide crystallization.



Figure 1 Schematic process to fabricate the integrated 3D nano-box arrays.

In the case of the construction of oxide nano-dot, Mo 3D nano-box was used as template for epitaxial oxide growth in selected nano area by pulsed laser deposition technique [7].

3. Results and Discussion

3-1: 3D Nano Box array :

Figure 2 shows SEM images of square Mo 3D nano-box with 60nm window (Fig. 2(a)) and ZnO semiconductor 3D nano-box with 60nm window (Fig. 2(b)). Remarkably, the widow size of 60nm could be fabricated much smaller than that of original mold size (250nm) by controlling etching step. The wall thickness of nano-box could be systematically controlled by deposition time from 50nm down to 10nm beyond the limitation of top down nano-fabrication. The resulting ZnO nano-box structures have a *c*-axis orientation after post annealing, and show an intense cathode luminescence peak around 400 nm at RT.



Figure 2 SEM images of (a) Mo metal nano-box array and (b) ZnO metal oxide nano-box array.

3-2: 1D Nano wire array :

To construct 1D nano-wire structure, the template mold was changed to a line and space pattern. Figure 3 shows SEM images of organic resist line pattern (Fig. 3(a) and ferromagnetic semiconductor (Fe,Mn)₃O₄ wire array after crystallization (Fig.3(b). The width of resulting (Fe,Mn)₃O₄ nano-wire is as small as 35nm shown in the inset of Fig. 3(b), and exhibited magnetoresistive properties at room temperature.



Figure 3 SEM images of (a) organic resist wire pattern, (b) $(Fe,Mn)_3O_4$ nano-wires (The inset shows the cross sectional profile).

3-3: 0D Nano dot array :

Toward much lower dimensional nano-oxide, the Mo nano box was applied as nano template for epitaxial oxide deposition [7]. Figure 4 shows extremely small epitaxial nano-dot structures of ferromagnetic semiconductor oxide (Fe,Mn)₃O₄ beyond the limitation of top down nano-process, which was directly grown on sapphire substrates at the deposition temperature of 350°C in a pulsed laser deposition method.



Figure 4 SEM images of epitaxial (Fe,Mn) $_{3}O_{4}$ metal oxide nano-dot array grown on Al $_{2}O_{3}$ (0001) substrate.

3-4: Physical property of ferromagnetic oxide nano-dot diode.

Nano-size hetero-structured devices, such as nano-FET and nano-diode using functional oxides, would be interesting as advanced spintronic nano-devices. Spin injection via Schottky contacts (Schottky diode) is one of the most important techniques for spintronic device applications. In (Fe,Zn)₃O₄ degenerated semiconductor thin films grown on n-type semiconducting Nb doped SrTiO₃ (Nb-STO) substrate, when a magnetic field is applied to the ferromagnetic diode, the exchange energy Δ_{ex} increases to $(\Delta_{ex} + 2\mu_B B)$ in the ferromagnetic layer by a Zeeman splitting effect (μ_B : Bohr magnetron, B: magnetic field), and the majority (down) spin increases. As a result, magnetic field tunable diode could be constructed. For fabrication of the nano-dot diodes, we used the epitaxial oxide growth into Mo nano-box mentioned above [2, 7].

Fig. 5 shows the magnetic field dependence of the junction magnetoresistance (MR) of a FZO/Nb-STO ferromagnetic nano-dot diode with a size of 300×300 nm² at 100 K. With an increasing magnetic field from 0 to 8T, the junction MR increased systematically from 1.00 (0T) to 1.08 (8T).



Figure 5 Magnetic field dependence of the junction MR of the $(Fe,Zn)_3O_4/Nb$ doped SrTiO₃ nano-dot diode $(300\times300 \text{ nm}^2)$ at 100 K. Inset shows the magnetic field vs. {I(B)/I(0)}-1 for the calculation of *P*.

According to the developed thermionic emission theory for ferromagnetic metal-semiconductor contacts by introducing the electron spin-split conduction band model, as proposed by Ziese *et al.*, spin polarization was estimated that *P* of FZO was enhanced from *P*=0.74 in a conventional film to *P*=0.89 in a nano-dot diode whose size of 300×300 nm² [5].

4. Conclusions

The nanofabrication technique used here will enable us to developmentally construct multi-dimensional nanostructured oxide devices such as a nano-dot TMR device with a half metallic ferromagnetic oxide like $(Fe,Zn)_3O_4$, and it will open up superior functionalities for macro-size film devices.

References

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