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Spontaneous orientation-tuning driven by the strain variation in self-assembled ZnO-SrRuO₃ heteroepitaxy

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Heteroepitaxial ZnO and SrRuO₃ were grown on SrTiO₃ (111) substrates and formed a self-assembled wurtzite-perovskite nanostructure. Spontaneous orientation-tuning of the SrRuO₃ pillars was observed, with the growth direction changing from [111]_{SRO} to [011]_{SRO} as the film thickness increased, which is attributed to a misfit strain transition from the biaxial strain imposed by the SrTiO₃ substrate to the vertical strain provided by the ZnO matrix. The [011]-SrRuO₃ and [0001]-ZnO combination presents a favorable matching in the nanocomposite films, resulting in higher charge carrier mobility. This vertically integrated configuration and regulation on the crystallographic orientations are expected to be employed in designing multi-functional nanocomposite systems for applications in electronic devices. © 2015 AIP Publishing LLC.

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As advanced control of nanomaterials synthesis developed over the past decades, heterostructures of the functional phases can be achieved to manipulate the interplay of lattice, charge, orbital, and spin degrees of freedom, which offers tremendous opportunities for next-generation electronic devices.^{1–3} For the heterostructure configurations, three main types have attracted widespread attention: nanodots/particles, multilayer or superlattice structures, and vertical architectures in terms of the different tunable dimensions.^{4–6} Specifically, the vertical heteroepitaxy with high interface-to-volume ratio has drawn a continuous spotlight and has been used to tune the functionalities due to their abundant structural freedom.^{7–9} The structure modification induces unique physical and chemical interaction phenomena in materials. Therefore, the design and control of the structure is a fundamental topic often addressed in the nanocomposite systems.

Various factors from thermodynamics and kinetics affect the evolution of the heterostructures, such as the elastic energy, crystal structure, interface energy, growth parameters, etc.^{10–12} Elastic strain energy for epitaxial thin films, commonly induced by the epitaxial constraints along the heterointerfaces due to lattice mismatch, has been demonstrated to be a vital factor controlling the growth of heterostructures. The elastic strain in the heteroepitaxial films can be divided into horizontal strain in biaxial in-plane directions and vertical strain in the out-of-plane direction. The in-plane strain has been systematically demonstrated to significantly

influence the structure and properties of epitaxial thin films.^{13,14} Meanwhile, the vertical strain originated from the columnar hetero-interfaces begins to control the growth behaviors of the component phases with increasing the film thickness.^{15,16} The significant effects on nanostructure growth triggered by the strains from the in-plane and out-of-plane directions, respectively, are quite complicated, which depends on the different structural information in the nanocomposite films.

In order to study the competition between the strain imposed by the substrate and the strain mediated by the matrix in vertical heteroepitaxy, we chose self-assembled (ZnO)_{0.67}:(SrRuO₃)_{0.33} nanocomposites epitaxially grown on SrTiO₃(111) as a model system. The integrated ZnO-SrRuO₃ (SRO) self-assembled nanostructures have interesting electron transport behavior and dynamic photoresponse performance driven by the visible light. However, the micro-structure and the underlining growth mechanisms on this wurtzite-perovskite orientation combination have yet to be addressed, which is a prerequisite to understand the coupling mechanism among electricity, optics, and elasticity. Here, we focus on the micro-structure, strain environment and the heterointerface structure of the wurtzite-perovskite system at an atomic scale. Specifically, a spontaneous optimized orientation-tuning in the nanocomposite is explored, and a relationship between the structure and the related properties has been built.

Self-assembled ZnO-SRO nanostructures were grown on (111)-oriented SrTiO₃ (STO) substrates at 800 °C by using pulsed laser deposition operating at 10 Hz in an oxygen pressure of 100 mTorr. The volume fraction of ZnO:SRO is

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67:33 in this system. SRO is an orthorhombic distorted perovskite structure (S.G. $Pbnm$ 62), having a pseudocubic lattice $a_{pc} = 3.941 \text{ \AA}$.¹⁷ The following indexing and discussion for SRO were referred to the pseudo-cubic unit cell for simplicity. ZnO is a hexagonal wurtzite structure (S.G. $P6_3mc$ 186) with the lattice of $a_H = b_H = 3.249 \text{ \AA}$, $c_H = 5.205 \text{ \AA}$.¹⁸ The subscripts $_H$ and $_{pc}$ represent the hexagonal and pseudocubic structures, respectively. Cross-sectional as well as plane-view samples for TEM (Transmission Electron Microscopy) studies were prepared by standard ion milling technique. TEM investigation was carried out using an FEI Tecnai F20 equipped with a high-angle annular dark-field (HAADF) detector.

Figure 1 gives the low magnification morphologies of the ZnO-SRO nanostructure on an (111)-oriented STO substrate from both plane-view and cross-sectional directions. In the “Z”-dependent contrast Scanning TEM (STEM) images, the SrRuO_3 phase containing heavy atoms of Sr (38) and Ru (44) exhibited much brighter contrast while the ZnO phase containing relative light atoms presented darker contrast. SRO nanostructures were homogeneously distributed in the ZnO matrix and grew vertically through the whole composite film (Figs. 1(a) and 1(c)). It demonstrates that the ZnO matrix grew along $[0001]$ direction while SRO nanostructures orientated along $[011]$ when grown on the (111)STO substrate, which is analyzed from the corresponding selected area electron diffraction pattern (EDP) in Fig. 1(d). ZnO grown as the matrix has the best in-plane atomic matching with the (111) STO substrate, that is, $\text{ZnO}[0001](1-100)_H \parallel \text{STO}[111](0-11)_{pc}$ with a lattice mismatch of $\sim 2\%$. The SRO domains rotated every 60° interval around STO $[111]$ axis in the film plane to derive SRO variants, which all matched the $[0001]$ -ZnO hexagonal patterns (red hexagon) with the same relationship. Three SRO $[011]$ -oriented domains marked by the green rectangles in Fig. 1(b) were identified by the crystallographic analysis. Thus, the orientation relationships are obtained as: $\text{ZnO}[0001](1-100)_{(11-20)_H} \parallel \text{SRO}[011](0-11)_{(100)_{pc}}$ [Fig. 1(b)], and

$\text{ZnO}(0002)_H \parallel \text{SRO}(011)_{pc} \parallel \text{STO}(111)$ [Fig. 1(d)]. The existence of nonperiodic additional weak spots in the EDPs besides the main phases is likely due to the presence of some small crystal nucleus formed during the film growth process. It is interesting to note that SRO variants prefer to elongate along in-plane $\langle 100 \rangle$ directions rotating every 60° , as shown in the inset zoom-in STEM image in Fig. 1(a), which also can be confirmed by the plane-view high-resolution TEM (HRTEM) analysis (see Fig. S1).¹⁹

It is worth noting that the crystal orientation of the SRO nanopillars in this $\sim 300 \text{ nm}$ -thick composite film was $[011]_{pc}$ rather than $[111]_{pc}$ direction. This is quite unexpected when grown on (111) STO substrate since both of them are perovskite structures. Previous reports suggest that a simple “cube-on-cube” hetero-epitaxial growth, typical in perovskite nanocomposite systems, would have occurred.^{6,20} In addition, the ZnO serves as the matrix while the perovskite SRO is the nanopillar, which is contrasted with other works.²¹ More detailed microstructural features are needed to be investigated deeply to reveal the growth behavior of SRO nanopillars and the heterointerface structure in the present system.

The cross-sectional heterointerface structure along the STO $[0-11]$ direction from the film bottom to top is shown in Fig. 2. The SRO nanopillar went through an orientation transition from $[111]$ to $[011]$ as the film grew thicker while the ZnO matrix maintained its $[0001]_H$ growth direction through the whole film. Typically, SRO presents an $[011]$ growth direction at the upper part of the film (Fig. 2(a)). Combined with the corresponding Fast Fourier Transaction (FFT) patterns (Figs. 2(c) and 2(d)), a good epitaxial relationship between the SRO nanopillar and the ZnO matrix can be identified: $\text{ZnO}(0002)(11-20)_H \parallel \text{SRO}(011)(100)_{pc}$, which is consistent with the EDP analysis. A well-defined two-phase heterointerface was observed, and no secondary phase was found at the boundaries. This is the most common type of interface bonding observed in the wurtzite-perovskite nanostructure system. Then, a coexistent area with both $[011]$ and

FIG. 1. (a) STEM Z-contrast image of ZnO-SRO nanostructures in plane-view sample and the corresponding diffraction patterns showing the in-plane rotation of SRO domains (b). The top inset in (a) is the higher magnification image showing clearly two-phase structures. The inset of histogram displays the statistical distribution of the lateral size (c) Cross-sectional STEM image show a vertical architecture of the composite films and corresponding selected area diffraction patterns (d).

FIG. 2. (a) The high resolution ZnO-SRO interface near the bottom of the film along $[0-11]\text{SRO}/[01-10]\text{ZnO}$ direction exhibiting the orientation tuning area of SRO phase. (b) The bottom triple interface of ZnO-SRO-STO substrate along the same cross-sectional direction shows a (111) -oriented SRO phase induced by STO substrate. (c)–(g) are the corresponding FFT of selected areas from (a) and (b), illustrating the orientations of different phases. Particularly, (d)–(f) are the FFT images of SRO nanopillars from top to bottom positions.

$[111]$ orientations come next when the bottom zone of the film with the thickness of about 30–50 nm was approached (Fig. 2(e)). Finally, $[111]$ -oriented SRO, matching well with the (111) STO substrate, turned into the dominant growth at the bottom of the film within 30 nm, as shown in Fig. 2(b). The FFT patterns corresponding to the areas squared in Figs. 2(a) and 2(b) are also given in Figs. 2(c)–2(g), respectively, which reveal clear spontaneous orientation transitions of the SRO phase from $[111]$, coexistent to $[011]$ orientations along the growth direction from the bottom to top in the wurtzite-perovskite composite film.

The spontaneous orientation tuning of the SRO nanopillars in the ZnO-SRO/STO(111) system brings different heterointerfaces and complex strain fields between SRO and ZnO phases, which is a key factor to explore the origin of the orientation transition process. Figures 3(a) and 3(b) are the cross-sectional HRTEM images of a ~ 40 nm-thick ZnO-SRO thin film grown on the (111) STO substrate. Such a thin composite film was designed deliberately to reveal its initial growth characteristic. The two phase separation was distinguished, and the SRO nanopillars were epitaxially grown on the STO substrate, as shown in Fig. 3(a). A zoom-in image in the vicinity of the right side SRO pillar is given in Fig. 3(b). ZnO matrix maintains its $[0001]_{\text{H}}$ orientation while $[111]$ -orientated SRO nanopillar exhibits cube-on-cube growth on (111) STO perovskite substrate, which is consistent with the previous results, revealed in Fig. 2(b). The Geometric Phase Analysis (GPA) method was carried out based on the HRTEM of the heterogeneous interfaces, which is an image processing technology used for mapping lattice displacement and has been successfully applied to characterize interface or defect structures such as misfit dislocations and their associated strain fields.²² GPA strain maps of the heterointerface shown in Fig. 3(b) were performed from in-plane (x axis) and out-of-plane (y axis) directions. For the

FIG. 3. (a) A cross-sectional TEM image along STO $[0-11]$ direction showing cube-on-cube growth of SRO on STO(111) substrate in the very thin composite film. (b) The enlargement of the SRO-ZnO-STO triple interface at the right side of (a) and the image was rotated for applying GPA method. (c) and (d) GPA graphs of (b) along the x, y axes give the distortion map ϵ_{xx} , ϵ_{yy} , respectively. (e) A typical high resolution cross-section TEM image of the ZnO-SRO interface along $[0-11]\text{SRO}/[01-10]\text{ZnO}$ direction and the corresponding FFT showing at the upper corner. (f) GPA lattice distortion map, ϵ_{yy} , of (a) along the growth direction. (g)–(i) profiles extracted from GPA maps of (c), (d), and (f), respectively.

in-plane distortion map (ϵ_{xx}) in Fig. 3(c), the relative uniform color contrast was revealed with a small lattice misfit of $\sim 1.0\%$ between the two phases and the substrate (Fig. 3(g)). This suggests that a strong in-plane strain constrained by the substrate is imposed on the composite film system, resulting in the epitaxial growth of [111]-oriented SRO and c-axis oriented ZnO phases. However, such a combination may lead to a large mismatch between SRO and ZnO, as shown in the out-of plane map (ϵ_{yy}) in Fig. 3(d). The lattice distortion and misfit planes in the fine SRO nanopillar were resolved clearly and the profile extracted across the SRO-ZnO hetero-interface (Fig. 3(h)) shows a $\sim 12\%$ misfit along the $[111]_{\text{SRO}}/[0001]_{\text{ZnO}}$ (y axis) direction. Consequently, decreasing such a large misfit strain and forming a stable vertical bonding in the system is necessary in the nanocomposite system. Fig. 3(e) is a typical cross-sectional HRTEM image of the SRO(011)-ZnO(0002) interface, which is determined to be the favorable vertical heterointerface bonding in the relative thicker wurtzite-perovskite composite films. Figure 3(f) presents the corresponding lattice distortion map (ϵ_{yy}) obtained by GPA method and no obvious lattice distortion was observed in SRO phase. Additionally, about 6.9% lattice deformation extracted from the profile in Fig. 3(i) is demonstrated between the $\text{SRO}(011)_{\text{pc}}$ and $\text{ZnO}(0002)_{\text{H}}$ planes, which is close to the theoretical calculated mismatch of $\sim 7.3\%$. Thus, the favorable combination with a small lattice mismatch between the two components of SRO and ZnO formed and the mismatch strain significantly relaxed with increased film thickness.

The epitaxial strains due to the mismatch between the component phases and the substrate play a key role in determining the growth behavior of the nanostructures. In view of the thickness variation of a vertical nanocomposite film, the epitaxial strain originates from two aspects: laterally from the substrate and vertically from the component phases, which are the in-plane strain and the out-of plane (vertical) strain, respectively. The structure models illustrated schematically in Fig. 4 show the atomic arrangement of the heterointerfaces in the ZnO-SRO/(111)STO system and the related growth mechanism.

At the initial growth stage, composite thin films are strongly constrained by the STO (111) substrate, which controls the growth of the nanostructures by the in-plane strain originated from the lattice mismatch. Therefore, (111)-SRO nanopillars grow epitaxially on a (111) perovskite substrate and display a cube-on-cube orientation relationship, as shown in Figs. 4(a) and 4(c). However, the vertical combination bonding between ZnO(0002) and SRO (111) planes (Fig. 4(a) right) is not an optimized matching, in which a large mismatch of about 12% is present as shown in the ϵ_{yy} strain field map (Fig. 3(d)).

As the film grew thicker, the elastic energy contribution from the substrate sharply weakened when the thickness exceeded a critical value (~ 30 nm) by forming misfit dislocations. The calculated in-plane strain relaxation curves based on the Matthews and Blakeslee theory are shown in detail in Fig. S2.¹⁹ On the other hand, the vertical strain from the heterointerfaces of the composite film became dominant when the thickness exceeded ~ 30 nm. As a result, the followed SRO species deposited on the ZnO vertical walls tuned into the [011] orientation to match the [0001]-ZnO, forming a lower misfit heterointerface, as shown in Fig. 4(d). Finally, an interface with a relative small lattice mismatch of around 7% between SRO and ZnO formed when the film grew thicker, in which the vertically aligned SRO nanopillars grew along the [011] direction with their (011) planes paralleling to ZnO(0002) (Figs. 4(b) and 4(e)).

Furthermore, the electron transport behavior in the ZnO-SrRuO₃ films system was investigated. Figure 5 shows the resistivity ρ of the $\text{ZnO}_{0.67}\text{:SRO}_{0.33}$ film as a function of temperature with various film thicknesses. Interestingly, the trend of the resistivity change is increased initially and then decreased to a relatively stable value (200-nm and 300-nm films) relative to the film thickness. In general, structural defects are not only scatterers for charge carriers but also recombination centers for electrons and holes. Therefore, a well-matched interface is more favorable for the charge carrier transportation in the heterostructures, leading to a lower resistivity. Thus, the 200- and 300 nm-thick film with a large

FIG. 4. (a) Cube-on-cube epitaxial growth of SRO(111) on STO(111) substrate and the interface models of SRO(111)-ZnO(0002) matching hetero-interface. The labels of the colored atoms are given at the right corner. (b) The heterointerface model of SRO(011)-ZnO(0002) matching interface with a small lattice mismatch. (c)–(e) Three-dimensional model for SRO-ZnO films with different strain-control growth models on STO(111) substrate.

FIG. 5. Resistivity temperature dependence in the ZnO-SRO nanocomposite with different thicknesses.

portion of optimized interface structure ((011)SRO-(0001)ZnO, Fig. 3(e)) exhibited lower resistivity than the 50 nm-thick film, which had mainly heterointerfaces (111)SRO-(0001)ZnO with larger misfit (Fig. 3(b)). For the 150 nm-thick film, mismatched 111-orientation and mixed-orientations occupied a large portion of the SRO nanopillars. Also, Fig. 2(a) shows that the structure in the mixed-orientation areas was relatively disordered and that many defects exist in the transition zone. In this case, the charge carrier transport may be impeded by the large portion of mismatched interface in the nanocomposite, which can be attributed to the scattering mechanisms limiting the mobility.²³ Therefore, the highest resistivity was revealed in the 150 nm film. The investigation of the wurtzite-perovskite nanocomposites on the STO(111) substrate gives detailed microstructural information to deepen the understanding of all wurtzite-perovskite systems by combining the previous reports on the STO(001) substrates.

In summary, we demonstrated an attractive nanostructure configuration with a spontaneous orientation-tuning character and different heterointerface structures in self-assembled ZnO-SrRuO₃ nanocomposite films grown on (111)-oriented SrTiO₃ substrates. The substrate-controlled biaxial strain dominates the [111]-oriented growth behavior of SRO at the initial stage. The vertical elastic strain from ZnO matrix later became a driving force in controlling the orientation-tuning process and lead to a stable vertical heterostructures of SRO(011)-ZnO(0002) with higher charge carrier mobility. The plentiful combination forms and the heterointerface structures may provide plentiful choices to

manipulate degrees of freedom while leading to exciting functionalities in the complex transition metal oxides.

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