Research Article

Preparation of Compensation Ions Codoped $\text{SrTiO}_3:\text{Pr}^{3+}$ Red Phosphor with the Sol-Gel Method and Study of Its Luminescence Enhancement Mechanism

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Received 30 June 2014; Revised 14 November 2014; Accepted 20 November 2014; Published 14 December 2014

Academic Editor: Jung Y. Huang

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$\text{SrTiO}_3:\text{Pr}^{3+}$ is the most representative titanate matrix red phosphor for field emission display (FED). The red luminous efficiency of $\text{SrTiO}_3:\text{Pr}^{3+}$ will be greatly improved after the compensation ions codoping, so $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor has been a research focus at home and abroad. $\text{SrTiO}_3:\text{Pr}^{3+}$, $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Mg}^{2+}$, and $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$ phosphors are synthesized by a new sol-gel method. Crystal structure, spectral characteristics, and luminescence enhancement mechanism of the sample were studied by XRD and PL spectra. The results showed that after co-doped, $\text{SrTiO}_3:\text{Pr}^{3+}$ phosphor is single SrTiO_3 cubic phase, the main emission front is located at 614 nm, corresponding to Pr^{3+} ions $1D^2 \rightarrow 3H^4$ transition emission. $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Mg}^{2+}$ and $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$ phosphor luminescence intensity is enhanced, but the main luminescence mechanism is not changed. Acceptor impurity $x = \text{Mg}^{2+}, \text{Al}^{3+}$ will replace Ti bit after being doped into the crystal lattice to form X_{Ti}^- charge compensation corresponding defect centers Pr_{Sr}^+ to reduce the demand of Sr^{2+} or Ti^{3+} vacancy. While Sr-doped Pr will make lattice distortion and transition energy of 4f-5d is very sensitive to crystal electric field changes around Pr atom. Doping different impurities will make electric field distribution around the ion have a different change. It increases energy transfer of 4f-5d transition and improves the luminous intensity of $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor.

1. Introduction

Field emission display (FED) is a new display technology, with its high quality, low cost, large area of attractive advantages, achieving broad development prospects. Compared with cathode-ray tubes (CRT), FED has the high image quality. At the same time, FED has the thinness of liquid crystal display (LCD) and the large area characteristics of plasma display (PDP). FED has a considerable advantage in luminous efficiency, brightness, viewing angle, and power consumption way. In addition, FED also has a high resolution, fast response, high temperature resistant harsh, antivibration shock, weak electromagnetic radiation, and low production cost and is easy to implement digital display and so on.

Research found that $\text{SrTiO}_3:\text{Pr}^{3+}$ [1–3] is the most representative for FED display titanate matrix red phosphor in the perovskite structure. $\text{SrTiO}_3:\text{Pr}^{3+}$ phosphor generates red

light when photoluminescence and cathode-ray excitation. And the red light's coordinates is $x = 0.680$, $y = 0.311$, and it is very close to the American NTSC system providing an ideal red light. However, $\text{SrTiO}_3:\text{Pr}^{3+}$ material has a problem that Pr^{3+} ion luminous efficiency is low, limiting its application. Study found that compensation ion doping can greatly improve $\text{SrTiO}_3:\text{Pr}^{3+}$ material luminous intensity. For example, in $\text{SrTiO}_3:\text{Pr}^{3+}$ material synthesis process, the luminous intensity will increase nearly 200 times more when adding $\text{Al}(\text{OH})_3$ or Ga_2O_3 . Domestic Zhang et al. [4] studied Al^{3+} -doping of $\text{SrTiO}_3:0.2\%\text{Pr}^{3+}:x \text{ Al}$ ($x = 0 \sim 0.35$) to improve the performance of red phosphors [5] luminescence. When $x = 0.26$, its luminous intensity reaches the maximum; luminous intensity at this time is probably 20 times before. Yamamoto and Okamoto [6] used high temperature solid method to produce Al^{3+} -doped $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor and characterized and analyzed $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor. They have found generated strontium aluminate

TABLE 1: Calculated Mulliken population analysis for $\text{SrTiO}_3:\text{Pr}^{3+}$, $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Mg}^{2+}$, and $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$ samples.

$\text{SrTiO}_3:\text{Pr}^{3+}$			Mulliken bond population $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Mg}^{2+}$			$\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$		
Bond	Population	Length (nm)	Bond	Population	Length (nm)	Bond	Population	Length (nm)
Sr-O	-0.03	0.2760	Sr-O	-0.04	0.2765	Sr-O	-0.04	0.2764
Ti-O	0.62	0.1952	Ti-O	0.65	0.1933	Ti-O	0.68	0.1919
Pr-O	0.12	0.2700	Pr-O	0.15	0.2608	Pr-O	0.15	0.2609
—	—	—	Ti*-O*	0.66	0.1902	Ti*-O*	0.70	0.1893
—	—	—	Mg-O	-0.10	0.2015	Al-O	0.40	0.1910

Ti* and O* are the atoms that are near the impurity atoms (Al and Mg).

in the experiment, as well as proposing charge compensation theory. SrO layer disappears in SrTiO_3 lattice, degree of crystallinity is improved, and luminous enhancement has also been improved to some extent.

In this experiment, we use the sol-gel method to produce compensating ions Mg^{2+} , Al^{3+} codoped $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor. Sol-Gel method has the product's high uniformity, high purity, low firing temperature and the reaction easy control and so forth. Therefore, the project produces compensating ions doped $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor by the sol-gel method, improving the luminous intensity and persistence time by compensation ions doping method.

2. Experiment Details

In this experiment, $\text{Sr}(\text{NO}_3)_2$ and $\text{Ti}(\text{OC}_4\text{H}_9)_4$ are used as the precursor; $\text{CH}_3\text{CH}_2\text{OH}$ is the solvent and CH_3COOH is the stabilizer. In the process of sol configuration, we select Pr^{3+} , Al^{3+} , and Mg^{2+} to be doping elements to make $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor. Weigh moderate amount of $\text{Sr}(\text{NO}_3)_2$, $\text{Pr}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, and the impurities ($\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ or $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$) to a beaker marked A, and then add 15 mL of pure water to dissolve them and stir for about 20 min to manufacture solution A. Then, corresponding molar ratio of $\text{Ti}(\text{OC}_4\text{H}_9)_4$ and H_2O is 40 : 1; volume ratio of CH_3COOH and H_2O is 1.4 : 1; the amount of $\text{CH}_3\text{CH}_2\text{OH}$ is 10 mL; aging time and temperature are 1.5 h/32°C, and the sol is turned into gel; dry the gel in 180°C for 8 h and the gel is turned into light yellow powder. The light yellow powder finally is annealed in air at 950°C for 1 hour and the $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor has been made.

In this experiment, the crystal structure and lattice constant of the $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor are investigated by powder X-ray diffractometer (XRD). Photoluminescence (PL) spectrum is taken by Hitachi F4500 luminescence spectrometer. And wavelength of the excitation light is 350 nm, the gap width is 5 nm, and scan range is from 525 nm to 650 nm.

The virtual crystal approximation (VCA) method is carried out to establish undoped SrTiO_3 model and Al, Mg codoped $\text{SrTiO}_3:\text{Pr}^{3+}$ nanophosphors valence-bond model using the CASTEP software package [7]. For example, to $\text{SrTiO}_3:0.2\%\text{Pr}^{3+}, 25\%\text{Al}^{3+}$, we set Sr and Ti atoms as mixture atoms in the crystal using the VCA method. When Pr^{3+} replaces Sr^{2+} , the relative concentration of Pr^{3+} is 0.2% and Sr^{2+} is 99.8%. And when Al^{3+} replaces Ti^{4+} , the relative concentration of Al^{3+} is 25% and Ti^{4+} is 75%. The interaction between nuclei and electrons is approximated with Vanderbilt ultrasoft pseudopotential [8] and the Perdew and Wang 91 parametrization [9] is taken as the exchange-correlation potential, which is the precise method used for the calculation of electronic structure at present [10]. From the electronic structure point of view (including the Band Structure, DOS, Mulliken population analysis) give a reasonable explanation for compensating ions Al, Mg, Li codoped $\text{SrTiO}_3:\text{Pr}^{3+}$ system enhancement mechanism of luminescence. We use $\text{Pm}3\text{m}(\text{O}_{\text{h}1})$ as space group to establish the crystal cell; crystal cell parameters are $a = b = c = 3.9051 \text{ \AA}$, $\alpha = \beta = \gamma = 90^\circ$. Plane wave basis with kinetic energy cutoff of 380 eV is used to represent wave functions. And the Brillouin Zone integration is approximated using the special k-points sampling scheme of Monkhorst-Pack [11] and $6 \times 6 \times 6$ k-points grids are used.

3. Theoretical Calculations

After compensating ion Al codoping, $\text{SrTiO}_3:\text{Pr}^{3+}$ becomes a direct band gap semiconductor from an indirect band gap semiconductor, and the result is shown in Figure 1. Table 1 gives the calculated Mulliken population analysis for the prepared $\text{SrTiO}_3:\text{Pr}^{3+}$, $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Mg}^{2+}$, and $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$ samples. When Pr^{3+} is single doped, the bond lengths of Sr-O and Ti-O are 0.2760 and 0.1952 nm, and the corresponding bond populations are -0.03 and 0.62, which indicates that the Sr-O band is an ionicity and the Ti-O band is a high degree of covalency. As we have seen, the bond length of Pr-O is 0.2700 nm, which is 0.006 nm less than Sr-O, and a positive population value is 0.12, which obviously indicates that there exists strong interaction between Pr-O and the Pr-O's ionicity is weaker than Sr-O. In the case of $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Mg}^{2+}$, considerable electron charge density near the Mg atom is redistributed. It is obvious that the calculated bond length of Ti*-O* reduces to 0.1902 nm, the

FIGURE 1: The band structure of different concentrations of Al codoped $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$.

bond population of Ti^*-O^* increases to 0.66, and the bond population of $\text{Ti}-\text{O}$ also increases to 0.65. At the same time, the bond population of $\text{Pr}-\text{O}$ in $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Mg}^{2+}$ is bigger than $\text{Pr}-\text{O}$ band in $\text{SrTiO}_3:\text{Pr}^{3+}$. In the case of $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$, the bond length of Ti^*-O^* decreases to 0.1893 nm, smaller than Ti^*-O^* in $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$. Meanwhile, the bond population of Ti^*-O^* increases to 0.70 and the bond population of $\text{Ti}-\text{O}$ also increases to 0.68. As we all know, the calculated positive bond population in Mulliken population analysis indicates that there is strong attraction interaction between two atoms [12]. The larger the bond population is, the stronger the interaction is [12]. So, after codoping Mg^{2+} and Al^{3+} to $\text{SrTiO}_3:\text{Pr}^{3+}$, the interactions of $\text{Ti}-\text{O}$ and $\text{Pr}-\text{O}$ band are enhanced, and the interactions of $\text{Ti}-\text{O}$ and $\text{Pr}-\text{O}$ band in $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$ are stronger than those in $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Mg}^{2+}$.

In prepared $\text{SrTiO}_3:\text{Pr}^{3+}, \text{SrTiO}_3:\text{Pr}^{3+}, \text{Mg}^{2+}$, and $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$, SrTiO_3 lattice absorbed ultraviolet leading to electron transition from O 2p occupied valence bands to Ti 3d unoccupied conduction bands. Based on the above discussion, the $\text{Ti}-\text{O}$ and $\text{Pr}-\text{O}$ band is enhanced and the corresponding bond lengths decrease by Al(Mg) codoping. So, the charge transfer efficiency is improved, which

indirectly indicates that the energy transfer efficiency is improved. Therefore, the luminous efficiency of $\text{SrTiO}_3:\text{Pr}^{3+}, \text{Al}^{3+}$ is improved.

The band gap increases from 3.2 eV to 4.25 eV after being 25% Al^{3+} -doped. It introduced the impurity levels about -7 eV of the valence band and it is shown in Figure 1. As the doping concentration of Al increases, the energy band tends to degeneration and becomes more curved, the electronic effective mass becomes smaller, and it is more benefiting to transfer.

Figure 2 shows the DOS. It can be seen that the DOS peak is moved to right in the top of valence band. Fermi level enters into the valence band because the DOS peak of O p-stated electron is moved to left, and the contribution of O p electrons to the conduction band weakens or even disappears. The DOS peak is moved to left of Sr s-stated and d-stated electron, while p-stated is moved to right. The DOS peak of Ti d-stated electron is lessened and shifted to right. The contribution of Al, Mg of s-stated and p-stated electron is significant to the conduction band. Thus, the second impurity atoms will exclude the contribution of Ti d-stated electron to the conduction band, and the greater the doping concentration, the more apparent.

FIGURE 2: Continued.

FIGURE 2: The DOS of different compensation ions codoped $\text{SrTiO}_3:\text{Pr}^{3+}$.

FIGURE 3: (a) PL of samples Pr^{3+} single doped $\text{SrTiO}_3:\text{Pr}^{3+}$ (A: Pr^{3+} 0.1%mol; B: Pr^{3+} 0.2%mol; C: Pr^{3+} 0.5%mol; D: Pr^{3+} 1%mol; E: Pr^{3+} 3%mol). (b) PL of samples Mg^{2+} codoped $\text{SrTiO}_3:\text{Pr}^{3+}$ (A: Pr^{3+} 0.2%mol, Mg^{2+} 2%mol; B: Pr^{3+} 0.2%mol, Mg^{2+} 4%mol; C: Pr^{3+} 0.2%mol, Mg^{2+} 6%mol; D: Pr^{3+} 0.2%mol, Mg^{2+} 8%mol; E: Pr^{3+} 0.2%mol, Mg^{2+} 10%mol; F: Pr^{3+} 0.2%mol, Mg^{2+} 12%mol). (c) PL of samples Al^{3+} codoped $\text{SrTiO}_3:\text{Pr}^{3+}$ (A: Pr^{3+} 0.2%mol, Al^{3+} 0%mol; B: Pr^{3+} 0.2%mol, Al^{3+} 1%mol; C: Pr^{3+} 0.2%mol, Al^{3+} 5%mol; D: Pr^{3+} 0.2%mol, Al^{3+} 10%mol; E: Pr^{3+} 0.2%mol, Al^{3+} 15%mol; F: Pr^{3+} 0.2%mol, Al^{3+} 20%mol; G: Pr^{3+} 0.2%mol, Al^{3+} 25%mol; H: Pr^{3+} 0.2%mol, Al^{3+} 30%mol).

4. Results and Discussion

4.1. Luminescent Properties. Figure 3(a) shows the PL excitation spectra of the first group of samples Pr^{3+} single doped $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor. As we can see from Figure 3(a), the luminescence peak appears at the wavelength of 610 nm; the luminescence peak can be attributed to $1D^2 \rightarrow 3H^4$ transition of Pr^{3+} . Experimental results show that the sample

of $\text{SrTiO}_3:0.002\text{molPr}^{3+}$ luminous intensity is the highest, which is consistent with other related experiments. Generally, samples' luminescence intensity is not high enough; charge compensation principle described in the relevant literature can explain this phenomenon. Therefore, we introduce compensation ions codoping. Figure 3(b) shows the PL excitation spectra of the second group of samples Mg^{2+} codoped $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor and Figure 3(c) shows, obviously,

FIGURE 4: PL of different samples (A: Pr^{3+} 0.2%mol; B: Pr^{3+} 0.2%mol, Mg^{2+} 10%mol; C: Pr^{3+} 0.2%mol, Al^{3+} 25%mol).

the third groups of samples Al^{3+} codoped $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor. Experimental results show that samples have a luminescence peak which appears at the wavelengths of 611 nm and 614 nm, and the optimum ratio of the samples is Pr^{3+} 0.2%mol; Mg^{2+} 10%mol, Pr^{3+} 0.2%mol; Al^{3+} 25%mol, Pr^{3+} 0.2%mol.

Next, we choose separately the maximum luminous intensity of samples in the first three sets of samples to compare with each other, and the result is shown in Figure 4. Experimental results show that different compensation ions codoping does not cause the change of the position of the luminescence peak. And the position of the luminescence peak ranges between 610 nm and 615 nm; however, different compensation ions codoping will improve luminous intensity of the luminescence peak of $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor in varying degrees. Luminous intensity of Al^{3+} codoped $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor at the luminescence peak is max and increases by appropriately 2 times compared with the sample of Pr^{3+} single doping. Thus, trivalent Al has a better luminous enhancement function. Finally, we have researched the annealing temperature of Al^{3+} codoped $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphor, and Figure 5 shows the experimental result. As can be seen from Figure 5, the best annealing temperature is 950°C.

When there is only one impurity Pr^{3+} in SrTiO_3 crystal, Pr^{3+} replaces Sr^{2+} to generate positively charged impurity defect centers Pr_{Sr}^+ , which can deter electron transitions of the intra-4f from the $1\text{D}^2 \rightarrow 3\text{H}^4$ of Pr^{3+} ions [1, 13]. In the sample preparation process, it will produce some oxygen vacancies O^{2-} to neutralize with the positive charge generated by Pr_{Sr}^+ . The amount of Ti^{4+} vacancies or Sr^{2+} which is possibly generated by O^{2-} oxygen vacancies is rare, so excess positive charge compensation mechanism of Pr_{Sr}^+ induced

FIGURE 5: PL of different annealing temperature (A: Pr^{3+} 0.2%mol, Al^{3+} 25%mol, 890°C; B: Pr^{3+} 0.2%mol, Al^{3+} 25%mol, 950°C; C: Pr^{3+} 0.2%mol, Al^{3+} 25%mol, 1010°C).

by Pr^{3+} mostly is compensated by the oxygen vacancies O^{2-} ; the effect is not very obvious. Due to these reasons, luminous intensity of $\text{SrTiO}_3:\text{Pr}^{3+}$ materials without codoping is not high. Thus, we choose to introduce the second compensation ions codoping to mainly compensate the positive charge posted by Pr_{Sr}^+ . When the acceptor impurity X is mixed into the crystal lattice, it will replace Ti to form X_{Ti}^- charge compensation pair corresponding to Pr_{Sr}^+ , which reduces the demand of Sr^{2+} or Ti^{3+} vacancy. Meanwhile, when Sr is doped to Pr, it will cause lattice distortion. The energy generated by the 4f-5d transition is very sensitive to the crystal electric field changes around the Pr ion. After the incorporation of different acceptor impurities, it will change the crystal electric field distribution, thereby weakening or promoting 4f-5d transition energy transfer.

Obviously, as we see in Figure 6, after Mg codoping, there is a certain influence on the electric field around Pr^{3+} , but it is not obvious. When Al is codoped, there is a very distinct effect on the electric field around Pr^{3+} . Figure 6 is the Electron Density Difference of samples.

4.2. XRD. Figures 7(a) and 7(b), respectively, show the two sets of samples $\text{SrTiO}_3:\text{Pr}^{3+}$; $\text{SrTiO}_3:\text{Pr}^{3+}$, Mg^{2+} and $\text{SrTiO}_3:\text{Pr}^{3+}$, Al^{3+} XRD patterns. It can be seen from the figure that the overall XRD diffraction peak indicates that SrTiO_3 crystal grows along the (110) crystal orientation. Experimental diffraction peak coincides with the standard SrTiO_3 sample card (pdf number: 35-0734). The diffraction angles of the samples increase after Al(Mg) codoping as shown in Table 2. However, there exists a impurity peaks. Based on JADE software, the impure peak is caused by TiO_2 , which shows that a bit of TiO_2 exists in the crystal lattice and it causes the lattice changes. And the intensity of the

FIGURE 6: The Electron Density Difference of samples.

FIGURE 7: (a) XRD patterns of the first set of samples (A: Pr^{3+} 0.2%mol; B: Pr^{3+} 0.2%mol, Mg^{2+} 10%mol; C: Pr^{3+} 0.2%mol, Al^{3+} 25%mol). (b) XRD patterns of the second set of samples (A: Pr^{3+} 0.2%mol, Al^{3+} 1%mol; B: Pr^{3+} 0.2%mol, Al^{3+} 15%mol; C: Pr^{3+} 0.2%mol, Al^{3+} 30%mol).

TABLE 2: The first set of samples (110) diffraction peak characteristic values and parameters.

Number of samples	Diffraction angle
Pr^{3+} 0.2% mol	32.16
Pr^{3+} 0.2% mol, Mg^{2+} 10% mol	32.18
Pr^{3+} 0.2% mol, Al^{3+} 25% mol	32.3
Pr^{3+} 0.2% mol, Al^{3+} 1% mol	32.24
Pr^{3+} 0.2% mol, Al^{3+} 15% mol	32.26
Pr^{3+} 0.2% mol, Al^{3+} 30% mol	32.2

diffraction peaks is different: the samples' diffraction peak intensity of Al codoping is max, and the samples' diffraction peak intensity of Mg codoped is min. Based on the JADE software, we get two parts of the sample data shown in Table 2.

As we can see from Table 2, the diffraction angle has a slight increase from Pr^{3+} single-doped to Al^{3+} codoped. This phenomenon indicates that a part of Al and Mg at least is homogeneously incorporated into the SrTiO_3 lattice.

5. Conclusions

In a word, Mg^{2+} , Al^{3+} -doped $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphors and different annealing temperatures $\text{SrTiO}_3:\text{Pr}^{3+}:\text{Al}^{3+}$ are synthesized by the sol-gel method and the luminescence enhancement mechanism of compensation ions codoping is investigated. It can be seen from the experimental results that the compensation ions codoping can significantly improve $\text{SrTiO}_3:\text{Pr}^{3+}$ red phosphors' luminous intensity. When the excitation light is 350 nm and the gap width is 5 nm, the position of the luminescence peak ranges between 610 nm and 615 nm. Trivalent Al luminescence enhancement is the

best and divalent Mg also has enhancement, but the effect is not as Al^{3+} . And the optimal annealing temperature is 950°C .

X-ray diffraction (XRD) patterns show that the impurity ions are incorporated into the SrTiO_3 crystal lattice uniformly. Experimental diffraction peak coincides with the standard sample SrTiO_3 card (pdf number: 35-0734). There exists some impure peaks in experimental results, based on JADE software; this impure peak produces TiO_2 . In comprehensive experimental results, the impurity ions are incorporated into the crystal lattice and cause some difference but did not cause changes in SrTiO_3 main phase structure.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

This work is supported by the Natural Science Basic Research Plan of China (Program no. 61306009). The authors are also thankful for the support of the Natural Science Basic Research Plan of Shaan Xi Province of China (Program no. 2013KJXX-24).

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