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Article / Book Information

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Title(English)	Studies on effect of stress and film thickness on domain structure in (100)/(001)-oriented epitaxial tetragonal Pb(Zr,Ti)O 3 films
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（博士課程）
Doctoral Program

論 文 要 約

THESIS SUMMARY

系・コース： Department of, Graduate major in	材料 系 材料 コース	申請学位（専攻分野）： Academic Degree Requested	博士 （ 工学 ） Doctor of
学生氏名： Student's Name	一ノ瀬 大地	指導教員（主）： Academic Supervisor(main)	舟窪 浩
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Ferroelectric films have been widely investigated for various applications including nonvolatile ferroelectric random access memory devices (FeRAM) and microelectromechanical systems (MEMS). The ferroelectric and piezoelectric properties are strongly affected by the film orientation and its domain structure. The domain structure largely changes by applying an electric field in the ferroelectric and the piezoelectric material, and its domain structure strongly affect the ferroelectric and the piezoelectric property. Therefore, the investigation of the domain structure and its control can be a guide for designing the domain structure and characteristics of the ferroelectric material. Domain structure is strongly affected by strain and film thickness, so that it is important to investigate the relationship between domain structure and strain. The relationship between ferroelectric domain structure and strain is experimentally and theoretically studied by using a typical piezoelectric material such as $Pb(Zr,Ti)O_3$. Many of experimental studies are carried out for films under compressive strain. For example, domain structures on a $SrTiO_3$ substrate have been extensively studied from thin films of several nanometers to thick films of micrometer order, and analysis of domain structure has been conducted. However, compressive strain occurs in the $Pb(Zr,Ti)O_3$ film on the $SrTiO_3$ substrate under the film growth temperature, so the analysis is advanced only in the domain structure for the film grown under compressive strain. Meanwhile, film on Si substrates used in practical applications such as MEMS are prepared under tensile strain, so the domain structure on Si substrate is expected to be different from these on $SrTiO_3$ substrate. Therefore, the investigation of the domain structure of the films prepared under tensile strain is also important from a practical point of view. However, most of studies on the domain structure are carried out in thin films grown on under tensile strain, and there are few reports on research. In addition, recently there are reports emphasizing the importance of the domain structure of the films grown under tensile stress. However, research on a ferroelectric domain structure of the films grown under tensile stress is not sufficient at present. Therefore, a comprehensive investigation was carried out on the domain structure grown under tensile stress using a typical piezoelectric material, $Pb(Zr,Ti)O_3$ thin film.

The objective of this research is to investigate the domain structure of PZT film grown under tensile strain. Author mainly investigated the influence of strain and film thickness on domain structure of PZT film grown under tensile strain.

The thesis consists of the following sessions. The experimental set up including $Pb(Zr,Ti)O_3$ films growth and characterization techniques are described in Chapter 1.

The effect of misfit strain on the domain structure was systematically investigated using {100}-oriented epitaxial tetragonal PZT films with various Zr/(Zr+Ti) ratio grown on $(100)KTaO_3$ substrate in Chapter 2. The formation process of domain structure under tensile strain and compressive strain was investigated in Chapter 3. And then, the domain structure before and after applying an electric field was investigated in view of the domain structure under where compressive and tensile strain in Chapter 4.

The film thickness dependence of the domain structure under tensile strain was investigated in Chapters 5 and 6. I investigated mainly thin films below 90 nm in thickness in Chapter 5 And a thicker film up to a thickness of 5300 nm was investigated in Chapter 6.

In Chapter 2, the effect of misfit strain to the domain structure was systematically investigated using {100}-oriented epitaxial tetragonal PZT films with various Zr/(Zr+Ti) ratio grown on $(100)KTaO_3$ substrate. In general, studies on the influence of misfit strain have been carried out by adjusting the lattice parameter using different kinds of the substrate. However, systematic investigation is impossible by this method. In Chapter 2, systematic investigation on the influence of misfit strain was carried out by changing the composition of the PZT films grown on $(100)KTaO_3$ substrates. By using this method, it is possible to systematically investigate the domain structure formed under from compressive to tensile strain. The domain structure occurs is composed of *a*/*c*-domain in case of compressive strain, whereas the domain structure is composed of *a*/*a*-domain and *a*/*c*-domain in case of tensile strain. Many theoretical predictions are predicted to be composed of *a*/*c*-domain regardless of tensile and compressive strain. However, this result showed that not only *a*/*c*-domain but also *a*/*a*-domain is formed for the film prepared under tensile strain. The volume fraction of the domain can be controlled continuously by misfit strain and follows the following area mismatch equation: $a_{PT}^2 V_c + a_{PT} c_{PT}(1 - V_c) = a_{sub.}^2$. The domain volume fraction under compressive strain follows the above equation, but the domain volume fraction for the films under tensile strain does not follow above equation. Details are given in Chapters 3 and 4.

In Chapter 3, the formation process of domain structure under tensile strain and compressive strain was investigated. The domain structure where compressive strain occurs is changed from paraelectric phase to ferroelectric *c*-domain and then to *a/c*-domain as decreasing temperature. On the other hand, the domain structure where tensile strain occurs is changed from paraelectric phase to ferroelectric *a*/*a*-domain and *a/c* + *a*/*a*-domain as decreasing temperature. The formation process is different under tensile and compressive strain, there is a difference in the domain structure that is ultimately composed due to the difference in formation process. In addition, the influence of the cooling rate was also investigated during the cooling process. The influence of cooling rate was not noticeable in the domain structure under compressive strain, but the effect of cooling rate was observed in the domain structure under tensile strain. It is influenced kinetically in the domain structure under tensile strain.

In Chapter 4, the domain structure before and after voltage application was investigated for the films prepared under compressive and tensile strain. The domain structure of the film grown under tensile strain composed of *a*/*c*-domain and *a*/*a*-domain, and their lattice parameters are distorted compared to *a*/*c*-domain one for the film formed under compressive strain. The change in the domain volume fraction before and after the application of an electric field under compressive strain was several %, while that of under tensile strain was about 30%. The volume fraction after application of an electric field for the film grown under tensile strain well agrees with the area mismatch equation. By forming *a*/*a*-domain at the first, it is considered that following *a*/*c*-domain formation becomes difficult and it is not sufficiently relaxed. It is considered that it changed to a more stable state by applying an electric field.

In Chapter 5, the film thickness dependence of the domain structure of the film grown under tensile strain was investigated. I investigated mainly thin films below 90 nm in Chapter 5. A thin $PbTiO_3$ film below 90 nm in thickness grown on $KTaO_3$ substrate forms an *a*/*a*-domain. Lattice parameters and tilting angle increase with increasing a film thickness, and these parameters keep constant value in the case of reaching bulk value of *a*-axis one. The tilting angle increase with increasing of *c*/*a* ratio following equation: $\alpha = \tan^{-1}(c/a) - 45°$. Domain width of *a*/*a*-domain increase with increasing film thickness, showing the similar behavior followed by Kittel's law, but the thin film thickness is slightly larger due to the influence of *c*/*a* ratio change with film thickness. The phase transition temperature show the the maximum value of 660 °C in case of 30 nm thick film, and the phase transition temperature above 30 nm gradually decreases and approaches to the bulk phase transition temperature due to relaxation of strain. When the film thickness is below 30 nm, it tends to decrease gradually with decreasing film thickness, it may be the effect of general size effect and this is not reported for the films consist of *a*/*a*-domain.

In Chapter 6, the film thickness dependence of the domain structure under tensile strain was investigated. A thick film up to 5300 nm was investigated. In addition, the domain structures of the films grown under tensile and compression strain were compared and comprehensively investigated. The domain structure under compressive strain forms a *c*-domain in the thin film and changes to *a*/*c*-domain, then *a'*/*a'*-domain + *a'*/*c'*-domain as the film thickness increases. On the other hand, the domain structure of the film grown under tensile strain is *a'*/*a'*-domain + *a'*/*c'*-domain + *a''*/*c''*-domain, and then *a'*/*a'*-domain + *a'*/*c'*-domain. The combination of *a'*/*a'*-domain + *a'*/*c'*-domain was formed without a difference in domain structure under compressive and tensile strain in a film thickness of more than 2100 nm.