Process Integration of High-Performance Piezoelectric Micromachined Ultrasonic Transducer Using Epitaxial PZT Thin Film

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論文内容要約

In recent years, a piezoelectric micromachined ultrasonic transducer (pMUT) based on a flexurally-suspended membrane has been widely applied for health care devices in the range of 1–15 MHz of working frequency, for instance, ultrasonic imaging, ultrasonic care, etc.. The pMUT structure composes of two main layers, namely the piezoelectric thin film and elastic layer such as silicon or silicon dioxide. The control of fundamental properties, such as piezoelectric coefficient ($e_{31,f}$) and dielectric constant (ε_{r33}), of piezoelectric material is required to achieve the higher performance because these properties influence on sensing and transmitting sensitivities of pMUT device. In transmission mode, the larger deformation of membrane by the higher value of $e_{31,f}$ can provide the larger sound pressure at the same driving voltage. As a result, the transmitting sensitivity is decided by $e_{31,f}$ of piezoelectric material. In sensing part, the deflection of membrane is generated when receiving an acoustic wave. According on the piezoelectric effect, an output voltage is formed. In this case, the performance in sense mode is considered by *g* factor which is determined by $e_{31,f}/\varepsilon_0\varepsilon_{r33}$ where ε_0 corresponds to the vacuum permittivity. In order to consider the contribution of both transmission/ sense modes from the viewpoint of piezoelectric material, a performance evaluation which can know as figure of merit (FOM) of pMUT is determined by $e_{31,f}^2 / \varepsilon_0\varepsilon_{r33}$. This expression indicates that an ideal piezoelectric material which provides a large piezoelectricity and a low dielectric property will improve the performance of pMUT. But, the trade-off relationship among the piezoelectric and dielectric constants generally exists. In fact, the permittivity of piezoelectric material normally increases with increasing of the piezoelectricity. This relationship limits the total performance of pMUT in terms of the transmitter and receiver. A highly (001)–oriented (known as *c*–axis) lead zirconate titanate ($Pb(Zr,Ti)O_3$, PZT) with its unique characterizations was recently reported. The monocrystalline PZT thin film shows large piezoelectric coefficient and relatively small dielectric constant, i.e. $e_{31,f} = -10 \sim -14$ C/m^2 and $\varepsilon_{r33} = 200 \sim 300$, respectively, and promises an excellent performance pMUT by considering FOM of material.

However, there is a lack of the deep investigation for process integration of high–performance pMUT using epitaxial PZT thin film. It means that the large rooms for further improvement and design innovation still have necessary to create the next generation of excellent performance pMUT devices.

In addition, the FOM of material which considers only the effect of piezoelectric material on the performance of transducer is insufficient because the high–performance pMUT device can be also increased due

to the improving structure from changing the conventional structure with its fully–clamp boundary. Definitions of FOMs are employed to compare the relative performances of different structures based on different materials. Thus, the determination of FOM strongly depends on the specific application. For high frequency application of pMUT, the transmit sensitivity can be evaluated by pMUT velocity which equals to relationship between displacement-amplitude and frequency. The performance of ultrasound transmission is defined by f x $\delta_{dynamic}$ [MHz x nm] where $\delta_{dynamic}$ is the dynamic displacement at the center frequency, f, of pMUT. The receiver sensitivity is related to ($e_{31,f}$ / $\varepsilon_0\varepsilon_{r33}$). Considering the contribution of both material and structure parameters for high frequency applications, the FOM value is determined by (f x $\delta_{dynamic}$) x ($e_{31,f}$ / $\varepsilon_0\varepsilon_{r33}$) or a simple equation as (f x $\delta_{dynamic}$) x ($e_{31,f}$ /ε_{r33}). This expression is utilized to determine the high performance of pMUT based on epitaxial PZT film in this research compared to current devices.

The following summarizes each chapter and gives out the corresponding conclusion.

Chapter 2 Application of high (001) –oriented epitaxial PZT thin film to partially-etched pMUT

In this chapter, the investigations of the epitaxial growth as well as the repeatability of highly (001)-oriented PZT thin film on Si substrate using suitable oxide buffer layer $SrRuO_3$/$LaSrCoO_3$/YSZ (Y_2O_3: 8 mol%) were carried out. Although the oxide buffer layers and some fabrication parameters were reported, the effects of these factors on the growth and repeatability of preferably (001)-oriented PZT on Si substrate are still insufficiently studied.

The effects of fabrication conditions were deeply investigated to obtain its high reproducibility of the epitaxial growth success in this chapter. In fact, the better crystallinity of the buffer layer which induced by the thicker thickness of SRO layer was proved as one of the main causes to create the high (001)-orientation of epitaxial PZT thin film.

Next, according on the prediction using finite element method, the partially-clamped pMUT structure using monocrystalline PZT thin film was studied and prepared because this structure helped the membrane to vibrate easily and promised to enlarge the displacement sensitivity.

Under applying the excited voltage to determine the electric/ mechanical sensitivity, it was found the important information about the fragile structure of pMUT using monocrystalline PZT thin film after several measuring. In fact, the crack problem caused unreliable results and has reported for the first time. Micro-cracking and mechanical fractures are considered as the main causes to degrade their electrical and mechanical properties and thus reduce the usability. However, relatively few discussions have been reported to crack phenomena in the monocrystalline PZT thin film.

Chapter 3 Fibered–epitaxial PZT thin film for robust pMUT

In this chapter, to solve the fragile structure of pMUT based on monocrytalline PZT thin film, a potential piezoelectric material with its well-balance properties between high FOM value and mechanical robustness was proposed. The crack generation in pMUT structure using highly (001)–oriented PZT thin film could be explained by the existence of larger residual stress in the thin film. At the growth temperature, misfit and intrinsic stresses mainly contribute to stresses. During the cooling process, stresses such as thermal stress or phase transformation stress might be added in the thin film. To reduce crack formation from residual stress, the growth of PZT thin film at lower epitaxial temperature was carried out in the range of 500 ~ 550^{o}C. The proposed PZT thin film shown intermediate features between the polycrystalline and monocrystalline thin films. The fact that the grain boundaries and in-plane epitaxial relationship were confirmed by scanning electron microscopy and X-ray diffraction analyses, respectively. Thus, this proposed film was called as "fibered-epitaxial PZT thin film. It had a smaller ε_{r33} value of ~500 and reasonably high $e_{31,f}$ of −10 ~ −11 C/m^2 even without poling treatment in comparison to these properties of the polycrystalline case. From this result, FOM of material becomes high i.e. 22 ~ 27 GPa.

Mechanical characterizations of pMUT structures using fibered-epitaxial and monocrystalline thin films were shown by the visual observations of crack generations when applying driving voltages. As the result, the mechanical fragility such as mechanical breaks or crack generation was not shown in the fibered-epitaxial thin film after the actuation test. On the other hand, the pMUT device based on monocrystalline thin film PZT was observed with many cracks. One of the main reasons to easily crack is probably explained by a single crystal of thin film because it is able to cleave along this crystal direction. These results again verified the mechanical fragility of monocrystalline PZT thin film as a brittle material.

Probably the existences of the grain boundaries and/ or a little difference of the crystalline orientation in the in-plane orientation of fibered-epitaxial PZT inhibit the crack propagation as "crack stopper". It indicated the promising ability of fibered-epitaxial PZT thin film to improve mechanical robustness. From the viewpoints of the reliability and practical applications, fibered-epitaxial PZT thin film is a potential candidate for the mechanical robustness and flexibility of designs. This original study in this chapter successfully introduced the fibered-epitaxial PZT thin film with well-balanced properties and a reasonably-high FOM for pMUT because of its smaller dielectric constant and good mechanical robustness.

Chapter 4 Island pMUT based on monocrystalline PZT family thin film

In this chapter, to solve the crack problems of pMUT structure based on monocrystalline PZT, improving structures with supported materials were proposed to increase the mechanical robustness from the viewpoint of structure. The modified structures were studied to maintain the high FOM ($e_{31,f}^2/\varepsilon_0\varepsilon_{r33}$) of highly (001)−oriented thin film PZT family due to the great potential of the piezoelectric and relatively small dielectric constant, while their robustnesses were still obtained by the improving designs.

On pMUT with partially−etched PZT film, the propagation of cracks usually found out at the corner of partially-etched structure. It implied that the high residual stress was stored at these corners after fabrication process. In order to remove the crack generation, the unexpected PZT area which did not contribute to the actuation had eliminated. An island pMUT structure was considered with including only the motional part of piezoelectric thin film without its neighboring area. It is the first time that an effective solution has introduced to solve the mechanically fragile structure, besides the analysis and estimation studies in the previous literature.

This new designs provided the relatively linear relationship for both higher vibration and robustness of pMUT with increasing the external voltage. Moreover, the presentation of the transduction ratio of electrical/ mechanical energy or vice versa using island pMUT by electromechanical coupling coefficient (k^2) was evaluated. In fact, k^2 of these modified structures increased to twice that of conventional structure. FOM ((f x $\delta_{dynamic}$) x ($e_{31,f}$ /ε_{r33})) of island pMUT structure using monocrystalline thin film PZT family, 60 [(MHz x nm) x (C/m^2)], was higher value than the current devices.

Chapter 5 Epitaxial growth of metallic buffer layer structure and highly (001)−oriented PZT family thin film on Si

In this chapter, metallic buffer layer structure for epitaxially growing monocrystalline thin film $Pb(Mn_{1/3},Nb_{2/3})O_3$−$Pb(Zr,Ti)_{O3}$(PMnN−PZT) on the silicon substrate was introduced. To obtain the better performance of island pMUT structure, it requires the high precision of etching PZT, for instance the use of dry etching. But, the oxide buffer layer structure is not suitable as an etch stop for both wet etching and dry etching processes. It can be explained by the quite similar characterizations between PZT and its oxide buffer layer. From this view, a metallic bottom layer which has ability to act as an etch stop is potential. Thus, "metallic buffer layer structure" for epitaxially growing PMnN−PZT thin film was worth studying.

The first layer of metallic buffer structure, Iridium (Ir) was successfully developed on a (100)YSZ/ (100) silicon. A lattice mismatch between Ir and YSZ layers could minimized to 0.4% for epitaxial growth along <100> direction by alignment of 4 unit cells of Ir (lattice constant = 0.5139 nm) on 3 unit cells of YSZ (lattice constant = 0.384 nm). Moreover, the deposition pressure of sputtering method was also studied to improve the crystalline orientation of (100)-oriented Ir on (100) YSZ/ (100) Si. At 0.3 Pa of the optimal pressure, dominantly (100)−oriented Ir thin film was achieved, whereas the (111) peak could not be observed. After that, a preferential (100) oriented thin film Pt was epitaxially grown on the (100) orientation of the Ir layer. It was contributed to not only the small mismatch 2.1% but also their same face centered cubic (known as *fcc*) crystal structures. On the other hand, a SRO layer was still used as a bottom electrode for this buffer structure to avoid the fatigue of PZT-based thin film. After that, the successful growth with epitaxial structure of the PMnN-PZT thin film on the metallic buffer layer was shown. The fact that the intensity ratio between (400) and (004) peak of PMnN−PZT reached more than 90%.

To consider the usability of metallic buffer layer structure in comparison with oxide buffer layer structure, fundamental characterizations of piezoelectric PMnN−PZT thin film on them were evaluated. On metallic buffer structure, the properties of thin film were $e_{31,f}$= −10 to −12 C/m^2 and ε_{r33}= ~250 for piezoelectric constant and relative dielectric constant, respectively. These values are quite similar to that of thin film on oxide buffer layer.

All these discussions and results in this thesis are strongly necessary to break-out of the limitations for integrating epitaxial PZT thin film into pMUT device. According on these successful investigations, the next generation of pMUT device using epitaxial PZT thin film is expected to show its larger sensitivity and better performance for wider applications.