1. Introduction

High-performance surface acoustic wave (SAW) devices are receiving much attention for high-frequency modules. Internet of things devices are rapidly spreading in our society, causing a massive increase in the data volume. To realize the processing of this massive amount of data in real time, fifth-generation (5G) communication systems with high-frequency bands of 3.7, 4.5, and 28 GHz is required. Thus, SAW devices that operate at high-frequency bands are indispensable for 5G communication systems. Surface acoustic wave devices consist of metal interdigitated transducers (IDTs) with a pattern pitch of several micrometers on a piezoelectric substrate. An electric signal incident on the piezoelectric substrate generates strain in the substrate, leading to excitation of the SAW via the piezoelectric effect between the electrodes. The SAW propagates from the input side to the output side of the IDTs parallel to the surface of the substrate.[1, 2] The SAW characteristic is expressed by the relationship $f = v / \lambda$, where $f$ is the selected SAW frequency; $v$ is the SAW phase velocity, determined by the piezoelectric substrate; and $\lambda$ is the wavelength of the excited SAW, determined as twice the pitch of the IDT pattern.[3, 4] Thus, the SAW performance depends on the piezoelectric substrate material and the IDT pattern pitch. The requirements for 5G SAW devices are (1) a large SAW velocity ($v \geq 6,000$ m/s), (2) a small temperature coefficient of frequency (TCF of $\leq 0$ ppm/°C) to reduce frequency fluctuation caused by temperature change, and (3) a large electromechanical coupling factor ($K^2 \geq 10\%$).[5] Nowadays, the pitch of the IDT pattern has reached the practical limits of lithography, thus varying the piezoelectric substrate materials is key for varying the SAW characteristic. The materials LiTaO$_3$ (LT) and LiNbO$_3$ (LN) have widely been used for 4G and
pre-4G SAW substrates, but the conventional thick LT or LN substrates do not meet the requirements of 5G SAW. Among the SAW propagation modes, leaky SAWs (LSAWs) and longitudinal LSAWs (LLSAWs) possess higher phase velocities than the Rayleigh-type SAWs used in conventional SAW devices. These LSAWs and LLSAWs can be excited using thin LT or LN substrates, where several studies have reported that the SAWs derived with thin LT or LN acquire the large velocities of LSAW or LLSAW. However, the $K^2$ of these reported SAW devices is too small because the thin LT or LN substrates were bonded to a non-piezoelectric material as a supporting substrate. Recently, we proposed thin-LT/quartz and thin-LN/quartz combined substrates, exploiting the fact that quartz is a piezoelectric single crystal and thus has a small TCF and a large $K^2$ value. In addition, the LT/quartz or LN/quartz bonding technique we have developed uses an amorphous SiO$_2$ or Al$_2$O$_3$ intermediate layer to improve the bonding reliability, where the amorphous films provide an active surface on the substrate (Fig. 1). For 5G SAW devices, the bonding strength and SAW characteristic of the proposed substrate can be improved by reducing the residual stress of the amorphous layer. Previously-reported simulation results predict that a thin-LT (2-$\mu$m-thick)/quartz combined substrate exhibits both a large $K^2$ (12%) and a small TCF (0 ppm/$^\circ$C). Corresponding measurements have shown that a thin-LT/quartz structure exhibits a larger $K^2$ and a smaller TCF than a conventional single LT substrate. Moreover, the quality factor of the LLSAW was improved by the Al$_2$O$_3$ intermediate layer inserted thin-LN/quartz structure, while the intermediate layers have a negligible effect on the phase velocity and $K^2$. However, the actual measured TCF value in that work was less than the calculated value of the simulations, where the disparity of results is presumed to be caused by residual stress in the bonding interface.

In this report, we studied various deposition methods of a low-residual-stress amorphous intermediate layer to optimize the bonding strength and SAW characteristics of LT/quartz and LN/quartz bonding. Amorphous films of SiO$_2$ or Al$_2$O$_3$ were deposited on LT substrates by three various deposition methods: ion beam sputtering (IBS), electron cyclotron resonance (ECR) sputtering, and atomic layer deposition (ALD). Next, the residual stress of the wafers were determined by the curvature of the substrates. Furthermore, thin-LN/quartz substrates bonded using amorphous Al$_2$O$_3$ films were also investigated from viewpoint of the bonding strength.

2. Experimental Procedure

2.1 Sample preparation

A double-side-polished LT substrate with a thickness of 0.21 mm was used for the residual stress evaluation. The LT substrate was diced into chips with dimensions 20 $\times$ 20 mm$^2$, whereupon ultrasonic cleaning using acetone, isopropyl alcohol, and de-ionized water at room temperature was carried out to remove organic contaminants from the substrate surface. Next, cleaning with an SC-1 solution (i.e., mixture of ammonia and hydrogen peroxide solutions) was performed at 80$^\circ$C. Then, an amorphous film was deposited on the substrate, where the film comprised either SiO$_2$ or Al$_2$O$_3$ deposited via IBS (M820, Hakuto), ECR sputtering, or ALD. The ALD process for SiO$_2$ used a different system (ALD-SiO$_2$; AT-400, Anric Technologies) and set of parameters than that used for Al$_2$O$_3$ (SUNALE R-150, Picosun). The intended thickness of the amorphous films was 100 nm excepting the ALD-SiO$_2$ sample (50 nm thick). The amorphous film deposition via IBS and ECR sputtering was conducted at room temperature, while the ALD-Al$_2$O$_3$ film was grown at 300$^\circ$C using a trimethylaluminum precursor and an H$_2$O oxidant; and the ALD-SiO$_2$ film was grown at 185$^\circ$C using a tris(dimethylamino)silane precursor and an O$_3$ oxidant. We ultimately studied six combinations of test samples, including IBS-SiO$_2$; IBS-Al$_2$O$_3$; ECR-SiO$_2$; ECR-Al$_2$O$_3$; ALD-SiO$_2$; and ALD-Al$_2$O$_3$.

2.2 Measurement of thin film residual stress

Before and after the amorphous film deposition, the warpage of each LT substrate was measured by a contact profilometer (KLA-Tencor Profiler P-15, KLA-Tencor) to
determine its curvature before \((R_1)\) and after \((R_2)\) deposition. The residual stress in the plane of the thin film could then be calculated by the Stoney formula,[16] given by

\[
\sigma = \frac{1}{6R_2} \left( \frac{1}{6R_1} \right) \frac{t_s^2}{t_f} \frac{E_s}{1-v_s},
\]

where \(\sigma\) is the thin film residual stress; \(E_s\) is the Young’s modulus of the substrate; \(v_s\) is the Poisson’s ratio of the substrate; and \(t_s\) and \(t_f\) are the substrate and film thicknesses. To simplify the calculation herein, a Young’s modulus of 230 GPa and a Poisson’s ratio of 0.25 were used for the LT substrate.[16] The actual thickness of each deposited film \((t_f)\) was measured by scanning electron microscopy (SEM; SU-8240, HITACHI) and ellipsometry (UVISEL, HORIBA JOBIN YVON).

2.3 Bonding of LN/quartz
To investigate the effect that residual stress in the amorphous film had on the bonding strength, 3 inch-diameter substrates of LN and quartz were used. Organic and SC-1 cleaning processes were performed on each substrate, whereupon a low-residual-stress amorphous Al\(_2\)O\(_3\) film (50 nm thick) was deposited on the quartz substrate as an active layer by either IBS, ECR sputtering, or ALD. The bonding surfaces of each substrate were treated using vacuum ultraviolet (VUV) light with a wavelength of 172 nm in the presence of oxygen gas (UER 20-172, Ushio, Inc.) at room temperature for 10 min. The light intensity and oxygen pressure in the chamber were 10 mW/cm\(^2\) and 7.0 \times 10\(^3\) Pa, respectively. This VUV treatment caused the surface to become hydrophilic following removal of the surface contaminants (Fig. 1-ii).[17, 18] After VUV treatment, bonding of the LN and quartz combined with the Al\(_2\)O\(_3\) film was performed using a bonding instrument (SB6e, SUSS MicroTec AG). The samples were bonded at 200°C with an applied pressure of 5 MPa for 10 min. Finally, the bonding strengths were measured using a tensile tester (MODEL-1307R, AIKOH ENGINEERING).

3. Result and Discussion
3.1 Evaluation of residual stress of the amorphous films
The warpage of the LT substrates without an amorphous film and those with various amorphous films is summarized in Fig. 2. The warpage of substrates coated with the Al\(_2\)O\(_3\) films was less than that of substrates coated with the SiO\(_2\) films for all deposition methods. In addition, the IBS-SiO\(_2\), IBS-Al\(_2\)O\(_3\), ECR-SiO\(_2\), and ECR-Al\(_2\)O\(_3\) samples exhibited convex upward warping; while the ALD-SiO\(_2\) and ALD-Al\(_2\)O\(_3\) samples exhibited convex downward warping. Thus, it was found that the direction of the warpage is a function of film formation method. Figure 3 summarizes and compares the results of the residual stresses as calculated from the warpage measurement and film thickness, where the ALD-Al\(_2\)O\(_3\) film exhibits the smallest residual stress of 127.3 MPa. This result suggests that the ALD-Al\(_2\)O\(_3\) film is useful for reducing residual stress.

The residual stress \(\sigma\) can be expressed with the thermal...
stress $\sigma_{th}$ and the intrinsic stress $\sigma_i$ using the relation

$$\sigma = \sigma_{th} + \sigma_i.$$  \hspace{1cm} (2)

The thermal stress is caused by a mismatch of the coefficient of thermal expansion (CTE) between the substrate and the film materials. Thermal stress can be calculated by

$$\sigma_{th} = E(\alpha_i - \alpha_s)(T_s - T_a)$$ \hspace{1cm} (3)

$$\sigma_{th} = E(\alpha_i - \alpha_s)\Delta T'$$ \hspace{1cm} (4)

$$\Delta T' = T_s - T_a$$ \hspace{1cm} (5)

where $E$ is the elastic modulus of the film; $T_s$ and $T_a$ are the substrate temperatures during deposition and measurement, respectively; and $\alpha_s$ and $\alpha_i$ are the CTE values of the substrate and the film, respectively. The parameters used in the calculation and the calculated values are summarized in Table 1. The thermal stress of the Al$_2$O$_3$ was larger than that of SiO$_2$. These results indicate that the intrinsic stresses are affected by the growth method rather than the thermal stress.[19–21] The contribution of thermal stress was thus considered to be low in these samples. Among the samples with deposited Al$_2$O$_3$ films, the LT substrate with the ALD-Al$_2$O$_3$ film exhibited the least amount of warpage. Thin film deposition via physical vapor deposition methods such as IBS and ECR sputtering are formed on one side of the substrate at a time. Thus, the substrate warpage occurs after the deposition and is a function of deposition condition and film quality (Fig. 4(a)). However, thin film deposition by ALD is formed on both sides of the substrate simultaneously because the step coverage of the ALD process is quite high.[22] The actual thicknesses of the ALD-Al$_2$O$_3$ film on back and front sides of the LT substrate were 95.6 and 94.6 nm, respectively. Therefore, the same amount of residual stress was applied at both the front and back sides of the LT substrate, which neutralized the residual stresses and reduced the warpage, as shown in Fig. 4(b). However, the residual stress of the ALD-SiO$_2$ film was not similarly reduced because the deposited film lacked uniformity (Supplementary Table S1), and thus its residual stress was not neutralized.

### Table 1 Residual Stress Calculation Parameters and results.

| Film material | CTE, $\alpha_{\times 10^6}/K$ | Elastic modulus, $E$ [/GPa] | Thermal stress, $\sigma_{th}$ [MPa] |
|---------------|-----------------|----------------------------|---------------------------------|
| Al$_2$O$_3$   | 4.2             | 171.0                      | $2.0 \times \Delta T'$         |
| SiO$_2$       | 0.5             | 70.0                       | $1.1 \times \Delta T'$         |
| LT            | 16.1 ($\alpha_s$) |                            |                                 |

![Fig. 4](image_url) Mechanism of residual stress generation during film deposition in (a) IBS and ECR sputtering, and (b) ALD.

![Fig. 5](image_url) Tensile strength of LN/quartz bonded substrate without (nothing) and with an amorphous Al$_2$O$_3$ film deposited via various methods.

### 3.2 Bonding results

To investigate the relationship between the residual stress and the bonding strength, the LN/quartz bonding strength combined with the Al$_2$O$_3$ films, which have lower residual stresses than the SiO$_2$ films, were measured. The bonded sample without an amorphous film was also evaluated as a reference, and the bonding results are shown in Fig. 5. The LN/quartz could not be bonded every time without the amorphous film, while the bonding succeeded when combined with the amorphous Al$_2$O$_3$ film. It is because the single crystal surfaces, especially quartz surface, were difficult to activated by the surface treatment by providing the active surface using the AIB method as previously reported.[11, 12] The amorphous Al$_2$O$_3$ films pre-
pared by each deposition method can promote hydrophilization of the quartz surface for the bonding process.[23] Finally, the sample combined with the film exhibiting the lowest residual stress (i.e., ALD-Al₂O₃) achieved the maximum tensile strength of 3.7 MPa. This result indicates that the ALD-Al₂O₃ film improves both the bonding strength and the SAW performance. However, the IBS-Al₂O₃ samples exhibited a greater residual stress and bonding strength than the ECR-Al₂O₃ samples. It is assumed that the bonding strength also depends on other parameters such as film density and crystallinity.[24]

4. Conclusion

We investigated the effect that the deposition method for a low-residual-stress amorphous film had on the bonding strength and SAW characteristics of LT or LN/quartz SAW substrates toward 5G communications. In this report, we evaluated the amorphous Al₂O₃ or SiO₂ films deposited by IBS, ECR sputtering, and ALD. It was found that the residual stresses of substrates deposited with amorphous Al₂O₃ films were smaller than those with SiO₂ films for all deposition methods. The differences of the intrinsic stress depend on the film growth conditions. The ALD-Al₂O₃ film was formed simultaneously on both the back and front sides of the LT substrate, and exhibited the smallest residual stress of 127.3 MPa. The maximum bonding strength of 3.7 MPa in the LN/quartz substrate was also achieved with the ALD-Al₂O₃ film. In addition, the Al₂O₃ intermediate layer has the positive effect on the chrematistics of the LLSAW as we previously reported.[14] This result indicates that the ALD-Al₂O₃ film is a promising amorphous film for 5G SAW devices.

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**Supplemental Data: Low-residual Stress Amorphous Film for LiTaO$_3$/quartz or LiNbO$_3$/quartz Bonding toward 5G Surface Acoustic Wave Devices**

| Sample | Thickness of front side nm | Thickness of back side nm |
|--------|-----------------------------|--------------------------|
| ALD-Al$_2$O$_3$ | 95.6 | 94.6 |
| ALD-SiO$_2$ | 70.4 | 64.5 |
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