paraelectric phase and the characteristic temperature $T_0$ are obtained from Fig. 9(a); $C_0=1.81\times10^6$ m/F·°C & $T_0=160$°C[7]. As the measured value of $P_s$ at $T_c=163$°C is $5.33\times10^{-2}$ C/m$^2$ [7], the values of $\xi$ & $\zeta$ are obtained by putting above values of $C_0, T_0, T_c$ & $P_s$ at $T_c$ into eq. (19) & eq. (25); $\xi=-5.65\times10^9$ m$^5$/F·C$^2$ & $\zeta=1.13\times10^{12}$ m$^9$/F·C$^4$. On the other hand, the applied electric field dependence of the relative permittivity at T=15°C & 120°C measured by the authors is expressed as a dotted line in Fig. 10[7]. By putting above values of $C_0, T_0, \xi$ & $\zeta$ into eq. (26), the applied electric field dependence of the relative permittivity is shown as a solid line in Fig. 10. The measured value follows to the calculated value approximately and then the suitability of eq. (26) is confirmed. Here, the solid lines at T=150°C & 155°C in Fig. 10 are merely the calculated curve from eq. (26). Moreover, by putting the previous values into eq. (24) & eq. (23), the temperature dependence of $1/\epsilon_r$ & $P_s$ are obtained like a solid line in Fig. 9(a) & (b). From Fig. 9(a), it is found that the measured value in ferroelectric phase coincides almost with the calculated value, and then it is predicted that the temperature dependence of $P_s$ should be like a solid line in Fig. 9(b).

4. Dielectric Loss Tangent

The electric field E and the dielectric constant in ferroelectric phase are given by eq. (3) and eq. (4), respectively. The polarization P in ferroelectric phase written here is expressed as the sum of spontaneous polarization $P_s$ and induced polarization $P_E$, namely

$$P = P_s + P_E \tag{35}$$

When the electric field $Ee^{j\omega t}$ with angular frequency $\omega$ is applied to the sample, the polarization induced in the sample by the field is expressed to be $P_E=P_0e^{j(\omega t-\delta_1)}$, where $\delta_1$ is the phase delayed from the phase of the applied field, and $P_0$ is the magnitude of the induced polarization.

As the coefficients u, g, $\xi$ & $\zeta$ in eq. (3) & eq. (4) must be complex number, the coefficients u*, g*, $\xi^*$ & $\zeta^*$ should be substituted for those. Then, when the field $Ee^{j\omega t}$ is impressed in the sample, the eq. (3), eq. (4) & eq. (35) must be written respectively as follows;

$$Ee^{j\omega t} = (u^* + g^*p)P^* + \xi^*P^{*3} + \zeta^*P^{*5} \tag{36}$$

$$1/(\epsilon^* - \epsilon_0) = u^* + g^*p + 3\xi^*P^{*2} + 5\zeta^*P^{*4} \tag{37}$$

$$P^* = P_s + P^*_E = P_s + P_0e^{j(\omega t-\delta_1)} \tag{38}$$

By putting P* given by eq. (38) into P* in eq. (36), the eq. (36) is expressed as follows;

$$\begin{aligned} Ee^{j\omega t} &= (u^* + g^*p)P_s + \xi^*P_s^3 + \zeta^*P_s^5 + P_0e^{j(\omega t-\delta_1)}(u^* + g^*p + 3\xi^*P_s^2 \\ &+ 5\zeta^*P_s^4) + P_0^2e^{j2(\omega t-\delta_1)}(3\xi^*P_s + 10\zeta^*P_s^3) + P_0^3e^{j3(\omega t-\delta_1)}(\xi^* + 10\zeta^*P_s^2) \\ &+ P_0^4e^{j4(\omega t-\delta_1)}.\, 5\zeta^*P_s + P_0^5e^{j5(\omega t-\delta_1)}\zeta^* \end{aligned}$$

Provided that the phases of u*, g*, $\xi^*$ & $\zeta^*$ are all $\delta_1$, furthermore, as $P_s$ in ferroelectric phase exists under E=0 and then $P_0=0$, the $P_s$ satisfies the equation derived from the eq. (36)

$$u^* + g^*p + \xi^*P_s^2 + \zeta^*P_s^4 = 0 \tag{39}$$

The above equation becomes the following form by putting A into $P_0/P_s$, namely $A=P_0/P_s$, and using the eq. (39);

$$
\begin{aligned}
Ee^{j\omega t} = A\ \{&(2\xi P_s^3 + 4\zeta P_s^5)e^{j\omega t} + A(3\xi P_s^3 + 10\zeta P_s^5)e^{j(2\omega t-\delta_1)} \\
&+ A^2(\xi P_s^3 + 10\zeta P_s^5)e^{j(3\omega t-2\delta_1)} + A^3 5\zeta P_s^5 e^{j(4\omega t-3\delta_1)} \\
&+ A^4\zeta P_s^5 e^{j(5\omega t-4\delta_1)}\}
\end{aligned}
$$

Where since A is the ratio of the induced polarization to the spontaneous polarization, the relationship $A<<1$ holds good. By using this condition and neglecting the higher term than the second one in the right hand side of the above equation, it is clear that the phase of the left hand side of the above equation is equal to that of the right hand side. Furthermore, when the complex dielectric constant is expressed to be $\epsilon=\epsilon_1-j\epsilon_2$, the loss tangent ($\tan\delta$) is shown to be $\tan\delta=\epsilon_2/\epsilon_1$. On the other hand, the reciprocal dielectric constant in paraelectric phase is expressed to be $1/(\epsilon^* - \epsilon_0) = u^* + g^*p = (u + gp)e^{j\delta_1}$ from eq. (37) because of including no higher terms than the second power of P*. In general, because of $\epsilon_1 >> \epsilon_0$ for ferroelectrics, the loss tangent in paraelectric phase is shown to be $\tan\delta=\epsilon_2/\epsilon_1 \doteqdot \tan\delta_1$ from above relationship. Therefore, the loss tangent in paraelectric phase corresponds to that in normal dielectrics.

By the above two facts, it is confirmed to be appropriate that phases of the coefficients u*, g*, ξ* & ζ* have been determined to be all $\delta_1$.

The complex dielectric constant $\epsilon^*$ in ferroelectric phase can be obtained by substituting eq. (35) & eq. (39) for eq. (37) as follows;

$$
\begin{aligned}
\frac{1}{\epsilon^* - \epsilon_0} &= (2\xi P_s^2 + 4\zeta P_s^4)e^{j\delta_1} + A(6\xi P_s^2 + 20\zeta P_s^4)e^{j\omega t} \\
&+ A^2(3\xi P_s^2 + 30\zeta\ P_s^4)\, e^{j(2\omega t-\delta_1)} + A^3\cdot 20\zeta P_s^4\, e^{j(3\omega t-2\delta_1)} \\
&+ A^4\cdot 5\zeta P_s^4\, e^{j(4\omega t-3\delta_1)}
\end{aligned}
$$

By comparing the real & imaginary parts of the right hand side with those of the left hand side with those of the left hand side of the above equation, the following relationships can be obtained;

From the real part,

$$
\begin{aligned}
\frac{\epsilon_1 - \epsilon_0}{(\epsilon_1 - \epsilon_0)^2 + \epsilon_2^2} &= 2(\xi + 2\zeta P_s^2)P_s^2 \cos\delta_1 + 2A(3\xi + 10\zeta P_s^2)P_s^2 \cos\omega t \\
&+ 3A^2(\xi + 10\zeta P_s^2)P_s^2 \cos 2(\omega t - \delta_1/2) + 0(A^3),
\end{aligned}
$$

where $0(A^3)$ stands for the small quantity including the higher terms than the third power of A ($A<<1$).

From the imaginary part,

$$
\begin{aligned}
\frac{\epsilon_2}{(\epsilon_1 - \epsilon_0)^2 + {\epsilon_2}^2} &= 2(\xi + 2\zeta P_s^2)P_s^2 \sin\delta_1 + 2A(3\xi + 10\zeta P_s^2)P_s^2 \sin\omega t \\
&+ 3A^2(\xi + 10\zeta P_s^2)P_s^2 \sin 2(\omega t - \delta_1/2) + O'(A^3)
\end{aligned}
$$

, where $O'(A^3)$ stands for the small quantity as well as $O(A^3)$. By putting above relationships into $\tan\delta = \epsilon_2/\epsilon_1$, the loss tangent in ferroelectric phase can be obtained as follows;

$$
\tan\delta \doteqdot \tan\delta_1 + \frac{3\xi + 10\zeta P_s^2}{(\xi + 2\zeta P_s^2)\cos\delta_1} A \sin\omega t + O''(A^2)
$$

, where $O''(A^2)$ stands for the small quantity. Then the root mean square of **tan δ** with time can be obtained as follows;