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Measurements of acoustic properties for thin films

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A measurement method for determining thin-film acoustic properties, such as characteristic acoustic impedance, sound velocity, density, and stiffness constant, is developed with a simple measurement principle and high measurement accuracy. The acoustic properties are determined from a maximum reflection loss and a center frequency obtained through a frequency response of the reflection loss for an acoustic transmission line composed of a sapphire/film/water system by using the acoustic pulse mode measurement system in the UHF range. The determination of the acoustic properties is demonstrated for sputtered fused quartz film, low-expansion borosilicate glass films, and chalcogenide glass films of evaporated As_2S_3 and As_2Se_3, within the measurement accuracy around 1–2%. It is also found that the acoustic properties of thin films are generally different from those of bulk materials, depending on the fabrication techniques and conditions.

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INTRODUCTION

Thin films prepared by the techniques of vacuum deposition, sputtering, chemical vapor deposition, and so on, have currently occupied an important role in the fabrication of thin-film devices, such as integrated semiconductors, integrated optic, and surface acoustic wave devices. Detailed data on thin-film characteristics are required to design the thin-film devices. In general, the characteristics of thin films should be considered more or less different from those of bulk materials, depending on the preparation methods and conditions. A great variety of materials are employed as thin films in electrical devices, and relevant properties of each thin-film material, such as structural, mechanical, optical, electrical, and magnetic properties, have been widely investigated. However, little investigation on thin-film acoustic properties, namely, elastic constant, sound velocity, and characteristic acoustic impedance, has been made, because the measurement methods of determining acoustic properties for thin films with a high accuracy have not been completely established as compared to those for bulk materials.[1–8]

Studies on the elastic properties of thin films started in the 1950s and especially the tensile strength and Young's modulus of evaporated or electrodeposited metal films, such as gold and silver, were evaluated by the tensile test.[9–12] Ultrasonic techniques were also applied for determining the sound velocity and elastic constant of metal films by the measurement of a delayed time in the ultrasonic low-frequency pulse echo method,[13] and a resonant frequency in the vibrating reed method.[14,15] Subsequently, the ultrasonic high-frequency pulse mode method was used to evaluate the acoustic properties of sputtered or evaporated glass films with a quarter-wavelength thickness, from the measurement of reflection coefficients in acoustic transmission lines at microwave frequencies.[16–18] An acousto-optic technique of using Brillouin scattering phenomena in thin-film optical waveguides can be used to measure the acoustic properties of glass films deposited on fused silica substrates.[19] Recently, material constants for AlN films epitaxially grown on sapphire substrates have been determined by computer fitting the dispersion curves of the velocity and electromechanical coupling factor measured for surface acoustic waves.[20] Nonscanning reflection acoustic microscopes, which have been established to measure acoustic properties of bulk materials,[21–24] also can be expected to determine the acoustic properties of thin films formed on substrates.

In this paper, a simple and very accurate measurement method for determining the acoustic properties of thin films is described. The method includes a technique of reflection loss measurements in the UHF range for an acoustic transmission line composed of a sapphire/thin-film/water system in which the thickness of the film is a quarter wavelength. This method is established by introducing the acoustic pulse mode measurement system previously proposed. The acoustic-property measurement technique can directly determine the characteristic acoustic impedance, sound velocity, density, and elastic constant for a thin film from a maximum reflection loss and a center frequency measured in a frequency response of the reflection loss. The measured results are presented for some typical films such as a rf sputtered fused quartz film, low-expansion borosilicate glass films, and vacuum-evaporated chalcogenide glass films.

MEASUREMENT PRINCIPLE

An acoustic transmission line, as shown in Fig. 1, is considered for describing the measurement principle of the acoustic properties of thin films. In this transmission line, a thin film with a characteristic acoustic impedance of Z_f is formed between reference media I and II. It is assumed that the acoustic medium I has a very high characteristic acoustic impedance of Z_1 and the medium II has a very low characteristic acoustic impedance of Z_2, and the acoustic loss in each medium, including thin film, can be neglected. The acoustic input impedance Z_{in}, which is the impedance looking from a-a' boundary, is obtained from the transmission line theory as follows:

$$Z_{in} = Z_f \frac{Z_2 + jZ_f \tan \beta l}{Z_f + jZ_2 \tan \beta l}, \qquad (1)$$

where $\beta = 2\pi/\lambda = 2\pi f/v$, β is the phase constant, f the

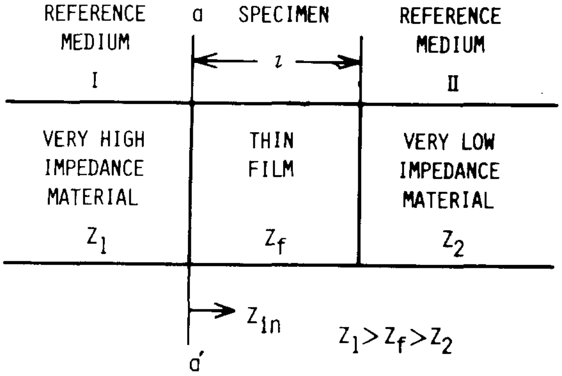

FIG. 1. Acoustic transmission line for determining acoustic properties of thin films.

acoustic frequency, v the sound velocity of the film, λ the acoustic wavelength in the film, and l the film thickness. When the sound wave is incident from the medium I, the reflection coefficient Γ is given at the a-a' boundary as follows:

$$\Gamma = (Z_{in} - Z_1)/(Z_{in} + Z_1). \quad (2)$$

Reflection loss RL is defined as

$$\begin{aligned} \mathrm{RL} &= -10 \log|\Gamma|^2, \\ &= -20 \log|(Z_{in} - Z_1)/(Z_{in} + Z_1)| \text{ (dB)}. \end{aligned} \quad (3)$$

The reflection loss represented by Eq. (3) is markedly dependent on the frequency. The magnitude of the reflection loss becomes maximum when $\beta l = (2n-1)\pi/2$; that is, $l = (2n-1)\lambda/4$, where $n = 1,2,\ldots$. In this paper, we take $n = 1$ so that $l = \lambda/4$. Here, a Z-cut sapphire and water are taken as the reference media I and II, respectively, in order to compare with the experiments, because the acoustic properties of these materials are well known. Characteristic acoustic impedances of Z-cut sapphire and water (20 °C) are 44.56×10^6 and 1.480×10^6 kg m^{-2} s^{-1}, respectively. Typical frequency responses of the reflection losses calculated are shown in Fig. 2, where the frequency is normalized by a frequency corresponding to $l = \lambda/4$. In this figure, thin-film characteristic acoustic impedance Z_f is taken as a parameter. At a frequency of $\lambda/4$, the reflection loss gives a maximum. When $l = \lambda/4$, Eq. (1) is $Z_{in} = Z_f^2/Z_2$, so that Eq. (3) is rewritten as follows:

$$\mathrm{RL_{max}} = -20 \log \left| \frac{Z_f^2 - Z_1 Z_2}{Z_f^2 + Z_1 Z_2} \right| \text{ (dB)}. \quad (4)$$

In the present method, the magnitude of the reflection loss $\mathrm{RL_{max}}$ and the center frequency f_0 at which the maximum reflection loss is given are used to determine the acoustic properties of thin film. With the measurement of the magnitude of the maximum reflection loss $\mathrm{RL_{max}}$, the characteristic acoustic impedance Z_f of a thin film can be determined by Eq. (4), if the characteristic acoustic impedances Z_1 and Z_2 of the reference materials are well known. The velocity v for a thin film can be calculated from the relation of $v = f_0\lambda = 4f_0 l$. The density ρ of a thin film can be determined by calculation from the definition of $Z_f = \rho v$. Furthermore, the stiffness constant c for the longitudinal velocity is calculated from the relation of $c = \rho v^2$.

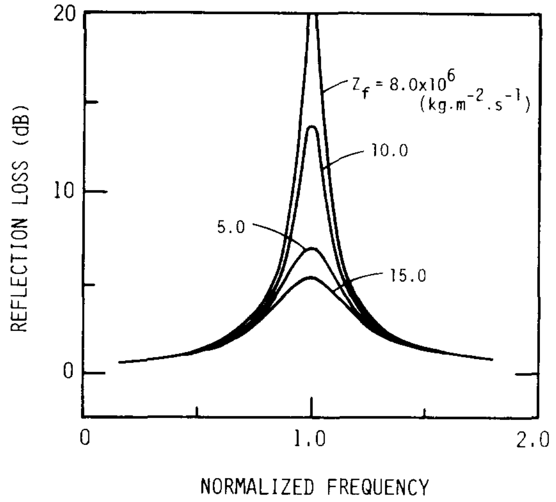

FIG. 2. Theoretical results of the frequency response of reflection loss in an acoustic transmission line composed of a sapphire/film/water system as a parameter of the characteristic acoustic impedance of the films.

Thus, the acoustic properties of thin films can all be determined with simple calculations only if the maximum reflection loss and the corresponding center frequency are measured.

PREPARATION OF THIN FILMS

A fused quartz (SiO_2), two kinds of low-expansion borosilicate glasses of E6 glass (Ohara Optical Glass Mfg. Co., Ltd.) and Pyrex glass (Corning Co., Ltd., No. 7740), vitreous As_2Se_3 and As_2S_3 are taken as thin-film materials to demonstrate the measurement method of determining the thin-film acoustic properties proposed here.

Thin-film samples with a film thickness corresponding to a quarter wavelength for an operating frequency are fabricated on one surface of Z-cut sapphire rods, 6 mm in length, (as shown in Fig. 3). Fused quartz and borosilicate glasses are deposited on the sapphire rods by planar magnetron rf sputtering under the following sputtering conditions: a mixture of Ar(80%) and O_2(20%), a gas pressure of 3×10^{-3} Torr, a distance of 60 mm between targets and substrates, a substrate temperature of 220 °C, an input rf power of 500 W, and a deposition rate of 1.2–1.5 μm/h. Chalcogenide glass As_2S_3 and As_2Se_3 films are deposited on the rods by heating a Mo-wire-wound fused quartz crucible with weighted evaporation sources under the following evaporation conditions: a substrate temperature of 25 °C, a pressure of 3–10×10^{-6} Torr, and a deposition rate of 1.0–1.5 μm/min. These chal-

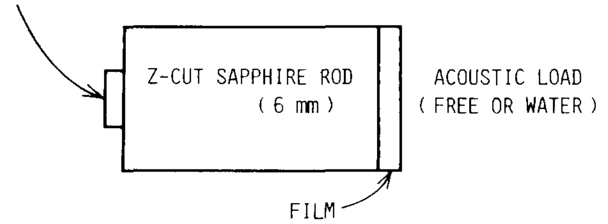

FIG. 3. Sample configuration for determining acoustic properties of films.

cogenide glasses are examined to be amorphous by x-ray analysis. Film thicknesses are measured by an interference microscope.

On the opposite end surface of the sapphire rods, ZnO piezoelectric film transducers with an electrode, 1 mm in diameter, are fabricated by dc sputtering to generate and detect longitudinal acoustic waves. The thickness of the ZnO film is about 5.7 μm and the center frequency of the transducers is around 450 MHz.

MEASUREMENTS

In order to measure the frequency response of the reflection loss in the acoustic transmission line for determining the acoustic properties of thin-film materials, the acoustic pulse mode measurement system[25] developed by the present authors is used. The block diagram of the system is shown in Fig. 4, and it has a wide dynamic range of more than 90 dB and a very high accuracy. The system is fundamentally composed of a combination of spectrum analyzer and tracking generator. A cw output signal of a tracking generator tuned to a spectrum analyzer is converted into a pulsed rf signal by an rf pulse modulator. The rf pulse signal with a pulse width of 0.7 μs, short enough to prevent the interference of acoustic waves in a sapphire rod, is applied to a sample. A signal pulse to be measured, which is the first acoustic echo reflected from the sapphire/water interface shown in Fig. 3, is substracted by an rf gate from a train of pulse echoes, and the peak power is detected by the spectrum analyzer. The pulsed video output of the spectrum analyzer is converted into a conventional video output by a peak-holding circuit. The output is recorded as a frequency response of the insertion loss into a digital wave memorizer which is linked to a computer. Two frequency responses of insertion losses are measured in a 50-Ω system for two different conditions: the film surface is free or in distilled water, respectively. The difference between these two insertion losses corresponds to a reflection loss at the sapphire/film interface in the acoustic transmission line of the sapphire/film/water system. The frequency response of a reflection loss calculated by the computer is displayed on an oscilloscope or an X-Y plotter.

From the frequency response of the reflection loss obtained in such a way, both the magnitude of maximum reflection loss $\mathrm{RL}_{\max}$ and the center frequency f_0 are determined. Figure 5 shows the experimental results of the frequency response of the reflection loss measured in a frequency range of 250 to 750 MHz for a $\mathrm{As_2Se_3}$ film with a thickness of 1.18 μm. The maximum reflection loss of 16.1 dB is measured at a center frequency of 467.8 MHz. By substituting the value of $\mathrm{RL}_{\max}$ into Eq. (4), the characteristic acoustic impedance Z_f for this film is determined to be $Z_f = 9.54\times 10^6$ kg m^{-2} s^{-1}. The longitudinal velocity v_l from the relation of $v_l = 4f_0 l$ is determined to $v_l = 2.21\times 10^3$ m s^{-1}. Then, the density ρ and the stiffness constant c_{11} are calculated to be 4.32×10^3 kg m^{-3} and 2.11×10^{10} N m^{-2}, respectively. The experimental results for all films are given in Table I, together with the bulk material constants including those for a Z-cut sapphire and water as reference materials.

In the table, all the values measured for $\mathrm{SiO_2}$ films are in good agreement with those for the bulk. For Pyrex glass, the measured values of the characteristic acoustic impedance, longitudinal velocity, density, and stiffness constant of the films are higher by 5.0, 2.3, 2.7, and 7.5% than those of the bulk, respectively. For E6 glass, they are also higher by 4.8, 2.6, 2.3, and 7.6%, respectively. On the other hand, for $\mathrm{As_2Se_3}$ films the impedance, velocity, density, and stiffness constant of the films are lower by 8.6, 1.8, 6.9, and 10.2%, respectively, and for $\mathrm{As_2S_3}$ films they are lower by 10.5, 7.7,

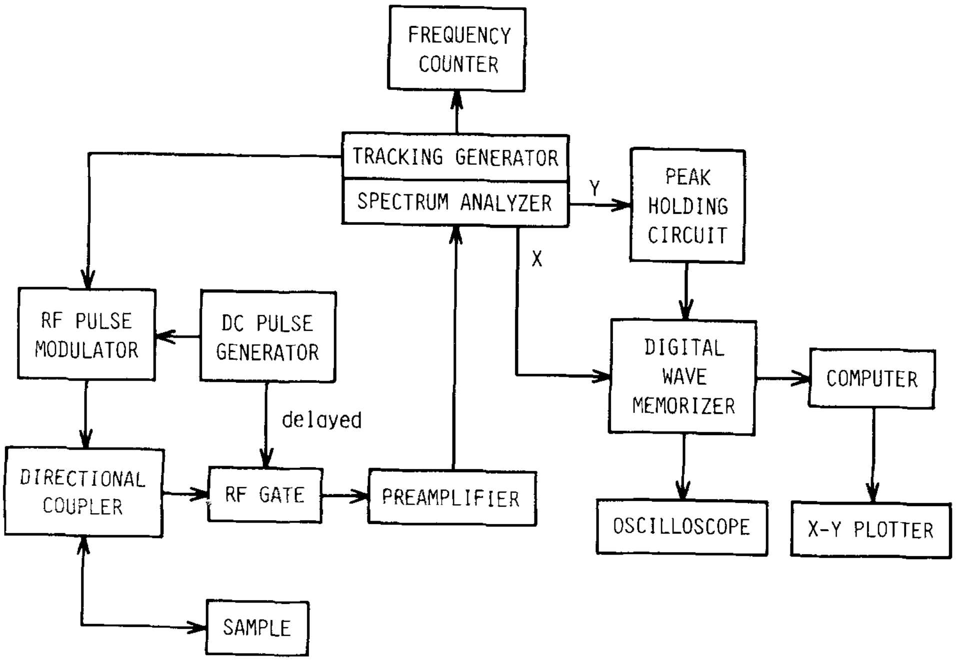

FIG. 4. Basic block diagram of the acoustic pulse mode measurement system for determining acoustic properties of films.

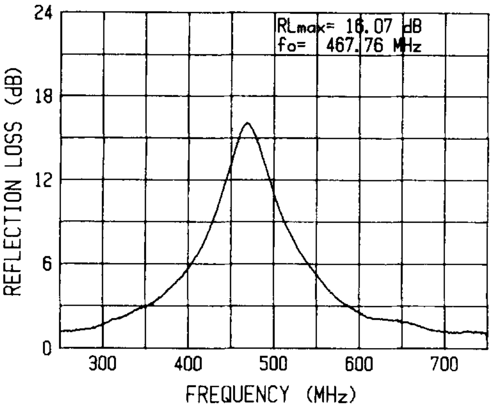

FIG. 5. Experimental results of the frequency response of reflection loss for an evaporated amorphous As_2Se_3 film (1.18 μm).

3.1, and 17.3%, respectively. These results prove that the changes of the acoustic properties by the reformation of the bulk to the film may be caused by the variations of density, composition, or structure, depending strongly on the preparation methods and conditions.

DISCUSSION

The effect of the acoustic loss is examined for each material in the acoustic transmission line. When the acoustic loss is taken into consideration, the characteristic acoustic impedance for each material must be represented as a complex quantity of $\dot{Z}$ as follows[26]:

$$\dot{Z} = \rho\dot{v} = (\rho\dot{c})^{1/2}, \tag{5}$$

where

$$\dot{c} = c + j\omega\frac{\alpha}{4.343}\left(\frac{c}{\rho}\right)^{1/2}\left(\frac{c}{\omega^2}\right). \tag{6}$$

$\dot{c}$ is the complex stiffness constant, α the attenuation constant (dB/m), and $\omega = 2\pi f$. In this case, the frequency response of the reflection loss differs slightly from that in the case of a no-loss transmission line. The center frequency f_0', giving a maximum reflection loss $\mathrm{RL}_{\max}$ (f_0'), and the complex characteristic acoustic impedance $\dot{Z}_f$, vary with the magnitude of the acoustic loss in thin film. The complex impedance of the film is given as

$$\dot{Z}_f = \left(Z_f^2 + j\frac{\alpha}{4.343}\cdot\frac{Z_f^3}{\rho\omega}\right)^{1/2}. \tag{7}$$

Thus, the film acoustic properties of a set of Z_f, v, ρ, c, and α should be appropriately determined by fitting the calculated curves of the frequency response of the reflection loss to the corresponding experimental curves with considering moderate acoustic losses.

To examine numerically the effect of the acoustic losses on the present measurement accuracy, the quantity of the variation of characteristic acoustic impedance is now defined as $|[Z_f(f_0) - \dot{Z}_f(f_0')]/Z_f(f_0)| \times 100(\%)$, where $Z_f(f_0)$ is the impedance obtained by neglecting the acoustic loss, while $\dot{Z}_f(f_0')$ is the complex impedance obtained by assuming the loss of α.

The calculated results as a function of the attenuation factor (dB/m) at 1 GHz are shown in Fig. 6 as a parameter of the characteristic acoustic impedance Z_f of the film. The values of 13.14×10^6 (bulk SiO_2), 8.26×10^6 (bulk vitreous As_2S_3), and 5.53×10^6 kg m^{-2} s^{-1} (As-S film determined in the experiment[18]) are taken as a parameter of acoustic impedance. In the figure, some bulk attenuation factors (dB/m) at 1 GHz are marked by arrows: Z-cut sapphire[26] of 1.8×10; SiO_2 (Ref. 26) of 1.5×10^3; vitreous As_2S_3 (Ref. 27) of 1.5×10^4; and water[28] of 2.2×10^5. In the calculations, acoustic attenuation is taken into account as α is proportional to f^2. It can be seen by the calculated results that the variation of characteristic acoustic impedance is not almost affected by the attenuation factor up to 10^4 dB/m at 1 GHz. In a range higher than 10^4 dB/m at 1 GHz, the variation

TABLE I. Measured film acoustic properties and bulk properties.

Sample	Characteristic acoustic impedance (10^6 kg m^{-2} s^{-1})		Longitudinal velocity (10^3 m s^{-1})		Density (10^3 kg m^{-3})		Stiffness constant (10^{10} N m^{-2})		Ref.
SiO_2	13.12	(13.14)	5.96	(5.97)	2.20	(2.20)	7.81	(7.85)	a
7740	13.09	(12.47)	5.72	(5.59)	2.29	(2.23)	7.49	(6.97)	b
E6	12.31	(11.75)	5.53	(5.39)	2.23	(2.18)	6.81	(6.33)	c
As_2Se_3	9.54	(10.44)	2.21	(2.25)	4.32	(4.64)	2.11	(2.35)	d
As_2S_3	7.39	(8.26)	2.39	(2.59)	3.09	(3.19)	1.77	(2.14)	e
Z-cut Al_2O_3	—	(44.56)	—	(11.18)	—	(3.986)	—	(49.81)	f
Water (20 °C)	—	(1.480)	—	(1.483)	—	(0.9982)	—	(0.2195)	g,h

Bulk constants in parentheses.
[a] W. P. Mason, *Physical Acoustics and the Properties of Solids* (McGraw-Hill, New York, 1958), p. 17.
[b] Technical data from Corning Co. Ltd.
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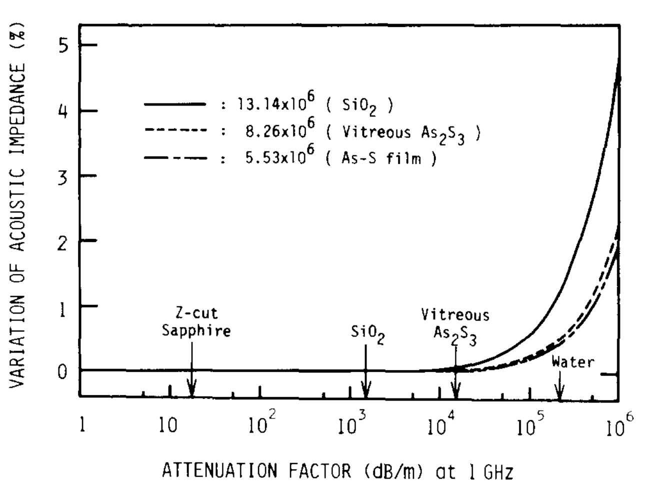

FIG. 6. Theoretical consideration of the effect of acoustic losses in the media on the determination of characteristic acoustic impedance. The parameter is the characteristic acoustic impedance of the film.

increases gradually with the value of the attenuation factor. Assuming tentatively that the acoustic loss in a unit of dB/m at 1 GHz increases enormously by one order of magnitude by reforming from the bulk to the film for each material, we can see from the figure that the variation for SiO_2 is less than 0.1%, and even that for As_2S_3 is less than 0.4%. As a result, it can be said that the normal acoustic losses in the media have little effect on the determination of the acoustic properties of films by the measurement method proposed here.

Other main factors affecting the accuracy of values determined in the present measurement method may be taken up as follows:

(a) measurement error of the pulse mode measurement system;

(b) temperature dependence of the acoustic properties of water as an acoustic load;

(c) measurement error of film thickness.

For (a), the measurement error of the system can be estimated to be less than about 0.1 dB.[17] The measurement error decreases as the maximum reflection loss RL_{max} increases. Namely, when RL_{max} is 5 dB, the error is at most 2%; but, if RL_{max} is more than 20 dB, the error is less than 0.5%. Factor (b) is not a major contribution to the error, because the physical values for water in determining the acoustic properties of the films are used for the temperature at which experiments are performed. For (c), the error of the film thickness measured by means of an interference microscope may be about ± 300 Å. The latest factor is considered to be a main factor for determining the measurement accuracy of this method. The overall measurement accuracy of this method can be estimated to within about 1–2%.

CONCLUSION

This paper has described the measurement method for determining the acoustic properties of thin films by the technique of reflection loss measurement in an acoustic transmission line. Experiments on the measurement of the acoustic properties of sputtered SiO_2 film, two low-expansion borosilicate glass films, and evaporated chalcogenide glass films of As_2S_3 and As_2Se_3 formed in the acoustic transmission line of Z-cut-sapphire/film/water system, have been demonstrated by using the acoustic pulse mode measurement system. The acoustic properties of the characteristic acoustic impedance, longitudinal velocity, density, and stiffness constant have been determined from a maximum reflection loss RL_{max} and a center frequency f_0 in the measured frequency response of reflection loss at the interface of the sapphire/film with a known film thickness. It has been found by experiment that the acoustic properties of films fabricated by the conventional deposition techniques such as evaporation and sputtering, in general, vary from those of bulk materials.

The present measurement system should be appropriately applied to film materials with a range of characteristic acoustic impedances from 4×10^6 to 20×10^6 kg m^{-2} s^{-1}. The method developed here with a simple measurement principle and high measurement accuracy is considered very useful for the direct determination of the acoustic properties of thin film.

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