

Elastic, anelastic, and piezoelectric coefficients of langasite: resonance ultrasound spectroscopy with laser-Doppler interferometry

著者	宇田 聡
journal or publication title	IEEE Transactions on Ultrasonics, Ferroelectrics and Frequency Control
volume	50
number	5
page range	553-560
year	2003
URL	http://hdl.handle.net/10097/47247

doi: 10.1109/TUFFC.2003.1201468

Elastic, Anelastic, and Piezoelectric Coefficients of Langasite: Resonance Ultrasound Spectroscopy with Laser-Doppler Interferometry

Hirotsugu Ogi, Nobutomo Nakamura, Keiji Sato, Masahiko Hirao, and Satoshi Uda

Abstract—This paper presents advanced techniques to determine all independent elastic-stiffness coefficients C_{ij} , the associated internal friction Q_{ij}^{-1} , and piezoelectric coefficients e_{ij} of monocrystal langasite ($\text{La}_3\text{Ga}_5\text{SiO}_{14}$) using a single rectangular parallelepiped specimen. Langasite's crystal structure belongs to the trigonal system with point group 32, thus six independent C_{ij} , two e_{ij} , and two dielectric coefficients ϵ_{ij} . All of the elastic and piezoelectric coefficients affect the mechanical resonance frequencies of the solid specimen, and measuring them very accurately permits one to determine the C_{ij} and e_{ij} with known density, dimensions, and ϵ_{ij} . We developed a piezoelectric tripod to support the specimen upward and measured the free-vibration resonance frequencies with minimum load from its own weight. This weak and stable acoustic coupling ensures high accuracy of the frequency measurement better than 10^{-5} , being enough to determine the reliable coefficients. Our C_{ij} fall in the range of results measured with previous (conventional) methods. Our e_{11} is smaller than the reported values by 1.2–13%, and e_{14} is larger than those by 44–97%. For the internal friction measurement, we used a solenoid coil to vibrate the specimen without any contact. The longitudinal-wave internal friction considerably exceeds the shear-wave internal friction, which can be interpreted as phonon-phonon interactions.

I. INTRODUCTION

LANGASITE attracts many researchers because of its large piezoelectric coefficients and less-temperature-dependent elastic constants. These properties are especially suitable for surface-acoustic-wave (SAW) filters [1]–[8]¹ and langasite is replacing quartz in a wide variety of electric devices. Langasite belongs to crystals with 32 point-group symmetry that show six independent elastic-stiffness coefficients C_{ij} , two piezoelectric coefficients e_{ij} , and two dielectric coefficients ϵ_{ij} . Besides, six independent internal friction Q_{ij}^{-1} will exist. Optimization of an electric device requires a complete set of the material coefficients, and significant efforts have been made for measuring them

[2]–[8]. The dielectric coefficients are available from low-frequency capacitance measurements [9], but measuring all of the elastic and piezoelectric coefficients presents a formidable task as demonstrated by Smith and Welsh [10]. Conventional methods used in previous studies [2]–[9] involve many independent measurements on many crystals in many orientations; the pulse-echo measurements or rod-resonance measurements coupled with the resonance-antiresonance measurements of electric impedance. One then must solve a set of labyrinthine equations. Various errors thus easily occur and are associated with the use of different specimens, crystal's misorientation, resonance frequency shifts by attaching electrodes, and so on. Concerning the internal friction tensor Q_{ij}^{-1} of langasite, only one study appears by Ledbetter *et al.* [11].

Here, we propose an advanced methodology that determines all the elastic, anelastic, and piezoelectric coefficients from a single rectangular-parallelepiped specimen. We note that the mechanical resonance frequencies of a piezoelectric solid depend on the C_{ij} and e_{ij} , and that resonant ultrasound spectroscopy (RUS) can detect them, from which an inverse calculation yields the needed coefficients. In the past, this approach was used to determine only the C_{ij} of metals [12]–[15], composites [16]–[18], and intermetallic compounds [19]. For quartz, Ohno [20] determined the C_{ij} , neglecting the e_{ij} and ϵ_{ij} . For lithium niobate (showing much larger piezoelectric coefficients than quartz), Dunn *et al.* [21] found that e_{ij} and ϵ_{ij} considerably affect the resonance frequencies and suggested that accurate frequency measurements would deduce the e_{ij} as well. For langasite, the e_{ij} 's contribution to the frequencies is in the order of 10^{-2} at most. To deduce them accurately, the resonance-frequency measurements must be done with an accuracy of 10^{-4} or better. Even more important is the mode matching between the observed and calculated resonances. Mode mismatch is fatal to obtain these less contributing coefficients. Therefore, obtaining the reliable e_{ij} along with the C_{ij} requires precise frequency measurements and correct mode identification. We realize this by using a piezoelectric tripod to detect free oscillation of the specimen and by incorporating laser interferometry into the resonance measurement to scan the surface displacement for identifying the vibrating modes. For internal friction, we used a contactless free-decay method to deduce Q_{ij}^{-1} .

Manuscript received March 28, 2002; accepted November 25, 2002.

H. Ogi, N. Nakamura, K. Sato, and M. Hirao are with the Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan (e-mail: ogi@me.es.osaka-u.ac.jp).

S. Uda is with the Microelectronics Laboratories, Mitsubishi Materials Corp., Yokoze, Chichibu, Saitama 368-8503, Japan.

¹Many other studies appear in 1998–2000 *Proceedings of the IEEE Ultrasonic Symposium*.

TABLE I
DIMENSIONS L_i (MM), MASS DENSITY ρ (10^3 KG/M³), ELASTIC-STIFFNESS COEFFICIENTS C_{ij} (GPA), AND PIEZOELECTRIC COEFFICIENTS e_{ij} (C/M²) OF THE FIVE SPECIMEN CRYSTALS.

	Specimen A	Specimen B	Specimen C	Specimen D	Specimen E	Average
L_1	2.966	2.97	8.032	10.024	9.965	—
L_2	3.745	5.768	9.814	10.043	10.048	—
L_3	4.012	4.012	6.022	13.764	14.406	—
ρ	5.723	5.730	5.713	5.731	5.723	5.724±0.007
C_{11}	189.4	190.6	190.4	188.7	188.7	189.5±0.9
C_{33}	261.0	262.8	263.3	263.2	262.6	262.6±0.9
C_{44}	53.35	53.95	53.37	53.50	53.33	53.50±0.26
C_{66}^*	42.10	42.28	42.10	42.05	41.94	42.09±0.12
C_{12}	105.2	106.0	106.2	104.6	104.8	105.4±0.7
C_{13}	96.80	97.12	98.28	96.77	96.83	97.16±0.64
C_{14}	14.19	14.47	14.28	14.18	14.11	14.25±0.14
e_{11}	-0.389	-0.387	-0.384	-0.405	-0.422	-0.397±0.02
e_{14}	0.183	0.208	0.22	0.209	0.194	0.203±0.01

$$*C_{66} = (C_{11} - C_{12})/2.$$

II. MATERIALS

We used five oriented rectangular parallelepiped crystals (specimen A to E). Using Archimedes's method and distilled water as a standard, we determined the mass density ρ . Their dimensions and mass density are given in Table I. The material coefficients in contracted notation can be written as:

$$[C_{ij}] = \begin{bmatrix} C_{11} & C_{12} & C_{13} & C_{14} & 0 & 0 \\ C_{12} & C_{11} & C_{13} & -C_{14} & 0 & 0 \\ C_{13} & C_{13} & C_{33} & 0 & 0 & 0 \\ C_{14} & -C_{14} & 0 & C_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{44} & C_{14} \\ 0 & 0 & 0 & 0 & C_{14} & (C_{11} - C_{12})/2 \end{bmatrix}, \quad (1)$$

$$[e_{ij}] = \begin{bmatrix} e_{11} & -e_{11} & 0 & e_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & -e_{14} & -e_{11} \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}, \quad (2)$$

and

$$[\varepsilon_{ij}] = \begin{bmatrix} \varepsilon_{11} & 0 & 0 \\ 0 & \varepsilon_{11} & 0 \\ 0 & 0 & \varepsilon_{33} \end{bmatrix}. \quad (3)$$

III. MEASUREMENT

A. RUS/Laser Technique

The RUS usually measures mechanical resonance frequencies of a solid specimen by sandwiching it between two piezoelectric transducers for the transmission and detection of acoustic vibration. This setup restricts the specimen's displacements and raises the frequencies from those at ideal free vibrations. To minimize this influence, we use a piezoelectric tripod consisting of two pinducers for generation and detection of vibration, and one only for support (Fig. 1). No external force was applied to the specimen, except for the specimen weight, and no coupling

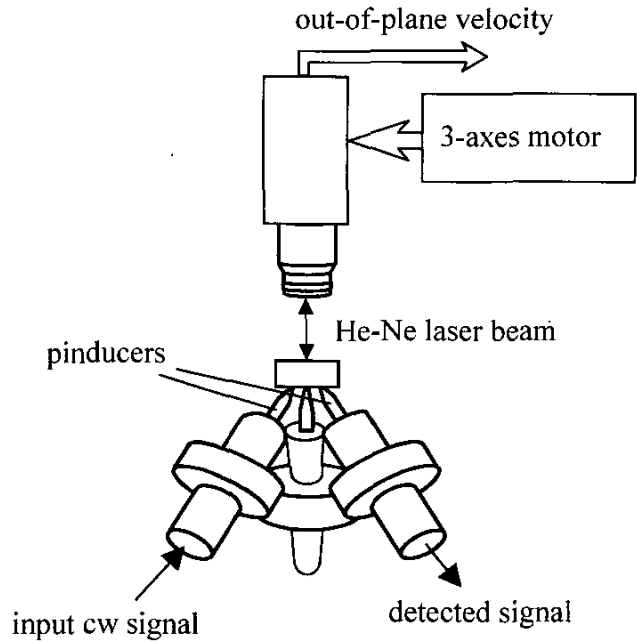


Fig. 1. Measurement setup of the RUS/laser combination.

agent was used. A frequency scan detected all the resonance peaks in a frequency band as shown in Fig. 2. We measured them at a constant temperature of $30 \pm 0.02^\circ\text{C}$ three times for each specimen. Owing to the weak coupling, reproducibility among the independent measurements was better than 10^{-5} .

After finishing the series of measurements (including the internal-friction measurement described below), we deposited 100-nm aluminum film on a specimen surface for mode identification with laser interferometry. (This deposition was needed because langasite is a transparent material.) A He-Ne laser beam was focused on the vibrating specimen surface with a $15 \mu\text{m}$ focal diameter. The reflected beam entered the Doppler interferometer, which detected the normal component of the velocity at the focal

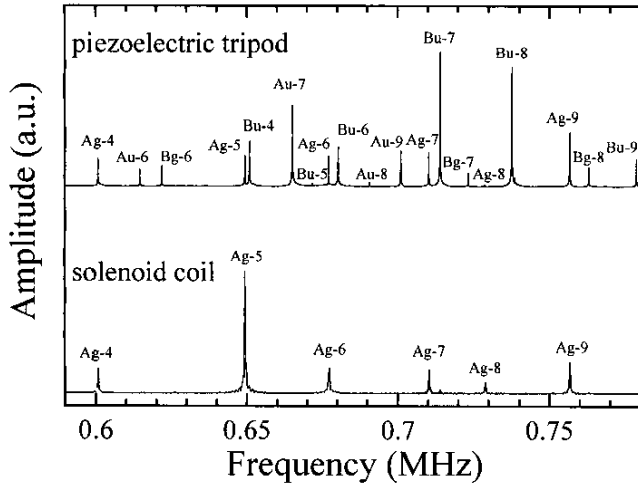


Fig. 2. Resonance spectrum of Specimen B measured by the piezoelectric tripod and solenoid coil.

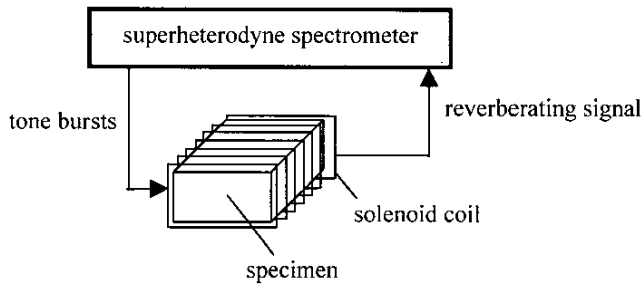


Fig. 3. Measurement setup of the dynamic electromagnetic-field technique.

point. The velocity was easily converted into the normal displacement because of harmonic oscillation. Depositing aluminum shifted so slightly the resonance frequencies that the modes are clearly identified after the deposition.

B. Dynamic Electromagnetic Field Technique

Not significant, but the piezoelectric tripod still causes energy leakage into the touching pinducers and as-measured internal friction contains a background. The best way to measure internal friction is to introduce vibration sources inside the specimen in a noncontact way using the piezoelectric effect of langasite itself. We used a solenoid coil to excite and detect the free vibrations by means of the dynamic electromagnetic field. Johnson *et al.* [22] first adopted this approach for studying temperature dependence of internal friction of langatate ($\text{La}_3\text{Ga}_{5.5}\text{Ta}_{0.5}\text{O}_{14}$), an isomorph of langasite, but not for the internal-friction tensor Q_{ij}^{-1} .

Fig. 3 shows our measurement setup. The specimen was inserted in a solenoid coil, which was loose and never constrained the specimen. We put a thin polymer sheet between the specimen and coil wires. The specimen and sheet are in contact at a few points; but, because of the large acoustic-impedance mismatch and no applied force,

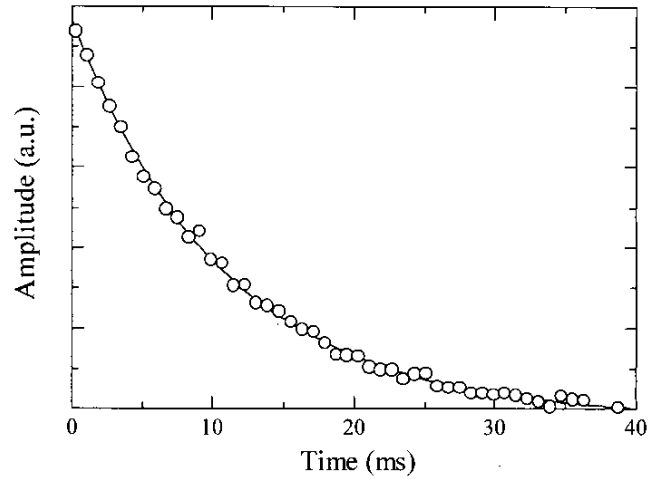


Fig. 4. Free decay of the reverberating amplitude after the excitation. Open circles denote measurements and solid line denotes the fitted exponential function.

they are acoustically noncontacting. We drove the coil with high-power radio-frequency (rf) bursts to induce oscillating electric field near the specimen surfaces and to excite the free vibration through the converse piezoelectric effect. After the excitation, the same coil received the vibration through the piezoelectric effect. We processed the received signals with an analog superheterodyne spectrometer to extract the signal amplitude at the tone-bursts frequency [23]. A frequency scan provided the resonance peaks (Fig. 2). We obtained the amplitude decay of vibration after exciting the coil at a resonance frequency and determined the attenuation coefficient α , which leads to internal friction via $Q^{-1} = \alpha/\pi f$. Fig. 4 exemplifies the measured amplitude decay and a fitted exponential function. Due to the frequency limit of our instrument, we measured internal friction of the smaller specimens A to C.

We did not use this noncontact method to determine the C_{ij} and e_{ij} through the resonance frequencies because of much fewer peaks than those measured by the piezoelectric tripod. For example, only A_g vibration modes were observed in Fig. 2. The detectable modes depend on the geometrical relationship between the crystal orientation and solenoid coil, that is, on the electric-field direction. Other configurations detected only B_g vibration modes. But, we failed to detect A_u and B_u modes with the solenoid coil. (Mode notation will be shown in Section IV.)

IV. INVERSE CALCULATION

A. C_{ij} and e_{ij}

The governing equations showing the interconnection between elastic and electric properties are

$$T_{ij} = C_{ijkl}S_{kl} - e_{kij}E_k, \quad (4)$$

$$D_i = e_{ikl}S_{kl} + \epsilon_{ij}E_j, \quad (5)$$

$$S_{kl} = \frac{1}{2} \left(\frac{\partial u_k}{\partial x_l} + \frac{\partial u_l}{\partial x_k} \right), \quad (6)$$

where T_{ij} is a component of the stress tensor. E_i , D_i , and u_i denote electric field, electric flux density, and displacement along the x_i axis, respectively. The electric field can be divided into a rotational component and an irrotational component (or quasistatic field). In the megahertz-frequency region, the rotation component is negligible and the quasistatic electric field dominates, which is expressed by the electric potential ϕ [24]:

$$E_k = -\frac{\partial\phi}{\partial x_k}. \quad (7)$$

(As will be shown later, this quasistatic approximation actually causes trivial errors in the resonance frequency calculation.) Substituting (1)–(4) into the equation of motion

$$-\rho\omega^2 u_i = \frac{\partial T_{ij}}{\partial x_j}, \quad (8)$$

with the boundary conditions for the stresses and electric field would lead to the free-vibration resonance frequencies ω of the system. However, analytical solutions for the displacements and electric potential are unavailable for rectangular-parallelepiped piezoelectric specimens. Eer Nisse [25] then derived the Lagrangian of a vibrating piezoelectric material as:

$$L = \frac{1}{2} \int_v \left(S_{ij} C_{ijkl} S_{kl} - \frac{\partial\phi}{\partial x_m} \varepsilon_{mn} \frac{\partial\phi}{\partial x_n} + 2 \frac{\partial\phi}{\partial x_m} e_{mkl} S_{kl} - \rho\omega^2 u_i u_i \right) dV. \quad (9)$$

The stationary point of the Lagrangian gives the resonance modes ($\delta L = 0$). Ohno [20] used approximation for the displacements and electric potential in terms of linear combinations of the basis functions Ψ consisting of the normalized Legendre polynomials:

$$u_i(x_1, x_2, x_3) = \sum_k a_k^i \Psi_k^i(x_1, x_2, x_3), \quad (10)$$

$$\phi(x_1, x_2, x_3) = \sum_k a_k^\phi \Psi_k^\phi(x_1, x_2, x_3). \quad (11)$$

Here

$$\Psi_k(x_1, x_2, x_3) = \sqrt{\frac{8}{L_1 L_2 L_3}} \bar{P}_l(2x_1/L_1) \bar{P}_m(2x_2/L_2) \bar{P}_n(2x_3/L_3). \quad (12)$$

\bar{P}_λ denotes the normalized Legendre polynomial of degree λ , and L_i denotes the edge length along the x_i axis of the rectangular parallelepiped. The Lagrangian minimization with a Rayleigh-Ritz approach [12], [13] determines the resonance frequencies together with the associated sets of expansion coefficients a_k^i . An oriented rectangular-parallelepiped crystal with 32 point-group symmetry has four vibration groups labeled A_g , B_g , A_u , and B_u , according to the deformation symmetry as tabulated by Ohno [20]. Choosing proper combinations of basis functions thus reduces the calculation time. While following

Ohno's calculation [20] (forward calculation), we implemented a least-squares-fitting procedure for the calculated and measured resonance frequencies to deduce the C_{ij} and e_{ij} .

In this inverse-calculation, comparison of the measurements with the calculations must be made on correctly the same resonance mode (otherwise, resultant material coefficients are physically meaningless). However, this has never been an easy task and mode mismatches easily occurred because nearly 100 resonance peaks are observed and some of them appear at very close frequencies. To overcome this difficulty, we pay special attention to the expansion coefficients a_k^i . They tell us the displacement distribution on a specimen surface, a fingerprint of an individual mode. Thus, comparison between the measured and computed displacement distributions leads us to unmistakable mode identification.

One cannot separately determine the e_{ij} and ε_{ij} from mechanical spectroscopy because their ratios affect the resonance frequencies. Fortunately, the ε_{ij} can be obtained by the capacitance measurements with good accuracy. Actually, several researchers [2], [3], [5]–[8] reached close ε_{ij} within a 2% range, while there are a lot of discrepancies for other coefficients (see Table II). For this reason, we fixed the ε_{ij} at averaged values over the previous studies.

B. Q_{ij}^{-1}

We neglect the piezoelectric effect for the calculation of internal friction tensor because their contributions to the resonance vibration are much smaller than those of the elastic stiffness coefficients. Internal friction Q_{ij}^{-1} can be considered as the ratio of imaginary-to-real part of the complex elastic stiffness \tilde{C}_{ij} [14], or:

$$\tilde{C}_{ij} = C_{ij} (1 + jQ_{ij}^{-1}). \quad (13)$$

Determination of all independent C_{ij} and their companion internal friction Q_{ij}^{-1} permits one to predict unmeasurable mechanical loss of any ultrasonic modes and then to select less-lossy mode, propagation direction, and surface orientation for designing acoustic devices. Calculation of the internal-friction tensor is based on a reasonable assumption that the square of the complex frequency can be expressed by a linear combination of complex elastic-stiffness coefficients:

$$\tilde{f}_p^2 = \sum_q \frac{\partial f_p^2}{\partial C_q} \tilde{C}_q = \sum_q 2f_p b_{pq} C_q (1 + jQ_q^{-1}), \quad (14)$$

Here, \tilde{f}_p denotes the complex frequency of the p th resonance and q denotes the matrix-notation subscripts (i.e., $q = 11, 12, 13, \dots$). $b_{pq} (= \partial f_p / \partial C_q)$ means the sensitivity of the elastic stiffness C_q to the resonance frequency f_p , which is obtainable in the inverse calculation [13]. However, we can define a particular modulus C_p and inter-

TABLE II
ELASTIC-STIFFNESS COEFFICIENTS (GPA), PIEZOELECTRIC COEFFICIENTS (C/M²), DIELECTRIC COEFFICIENTS NORMALIZED BY THAT IN VACUUM, AND THE INTERNAL FRICTION TENSOR.

	Present	[8]	[7]	[6]	[5]	[4]	[3]	[2]	Present Q_{ij}^{-1} (10 ⁻⁴)
C_{11}	189.5	188.5	189	189.5	189.3	190.2	188.9	190.9	1.73
C_{33}	262.6	261.7	268	259.9	262.4	262.1	262.2	261.9	0.95
C_{44}	53.5	53.71	53.3	53.91	53.84	53.82	53.9	52.4	0.26
C_{66}	42.09	42.21	42.4	42.4	42.16	42	42.2	43.2	0.27
C_{13}	97.16	96.88	102	97.86	95.28	91.9	96.8	104.2	1.65
C_{14}	14.25	14.15	14.4	14.64	14.93	14.7	14.3	15.2	0.16
e_{11}	-0.397	-0.402	-0.438	-0.428	-0.431	—	-0.44	-0.45	—
e_{14}	0.203	0.13	0.104	0.114	0.108	—	0.07	0.077	—
ϵ_{11}	19.04*	19.62	19.06	19.07	18.97	—	18.86	18.99	—
ϵ_{33}	50.51*	49.41	51.06	50.3	52	49	49.1	49.32	—

*Averages of the previously reported values.

nal friction Q_p^{-1} constructing the complex resonance frequency as:

$$\tilde{f}_p^2 = \text{const.} \tilde{C}_p = \text{const.} C_p (1 + jQ_p^{-1}) = f_p^2 (1 + jQ_p^{-1}). \quad (15)$$

Comparison of (14) with (15) leads to:

$$Q_p^{-1} = \frac{2 \sum_q b_{pq} C_q Q_q^{-1}}{f_p} \quad \text{and} \quad \frac{2 \sum_q b_{pq} C_q}{f_p} = 1. \quad (16)$$

The quantities f_p and Q_p^{-1} (peak width) correspond to the measured resonance frequencies and internal friction, respectively, and b_{pq} are obtainable as a result of the inverse calculation. Then, we can deduce the Q_q^{-1} ($= Q_{ij}^{-1}$) with a standard least-squares procedure.

V. RESULTS

Fig. 5 shows examples of measured and computed displacement figures on a vibrating crystal surface. Excellent agreement allowed us to make unambiguous mode identification. We identified more than 60 peaks for each specimen and entered them into the inverse calculation. Fig. 6 plots the differences between the measured and calculated resonance frequencies after the inverse calculation for all specimens. It also includes plots from purely elastic calculation neglecting e_{ij} and ϵ_{ij} . The piezoelectric effect raises the frequencies, that is, piezoelectric stiffening. Correlation coefficient between the measurements and calculations was 0.99999 and rms error between them was 0.065%. This agreement indicates the validity of the quasistatic approximation made in the analysis. Table I shows the determined elastic and piezoelectric coefficients. Despite various specimen dimensions, the resulting coefficients are close with each other. Averages over the five specimens are our final results, which are compared with previously reported values in Table II.

Fig. 7 shows the measured internal friction (Q_p^{-1}). The measurements by the piezoelectric tripod exceeded those by the contactless method using the solenoid coil, especially at lower frequencies. This implies that the RUS internal friction contains energy loss caused by contacting. In Table II, we show the six components of Q_{ij}^{-1} , which were deduced from all measurements for the three specimens by the solenoid coil.

VI. DISCUSSION

A. C_{ij} and e_{ij}

Possible errors arising in the present method are frequency-measurement error ($\sim 0.001\%$), dimensions and density error ($< 0.02\%$), crystal's misorientation error (less than 1°), and calculation error ($< 0.065\%$). Among them, we consider that the last issue dominates the accuracy of the resulting coefficients. The calculation error originates from the approximation for the displacements and electric potential in the inverse calculation. (Increasing the number of basis functions can reduce this error.) Therefore, a coefficient that contributes less to the resonance frequencies than the calculation error would not be determined on a rigorous basis. We calculated the normalized contributions of the coefficients to the resonance frequencies. Among all the coefficients, e_{14} showed the smallest contribution, 0.45% on average. (This is probably the main reason that previously reported e_{14} widely ranges.) However, this is still larger than the calculation error by a factor 7, indicating that reliable coefficients are obtainable. Using the contribution and the calculation error for each resonance mode, we estimated possible errors included in the resulting coefficients. The largest error occurs in e_{14} by 4%, which is smaller than the standard deviation among the five specimens ($\sim 7\%$) in Table I. Thus, differences of the resulting coefficients among the five specimens occur due to the difference of the crystal.

It has been pointed out that such an inverse calculation is much affected by the initial guess to start the itera-

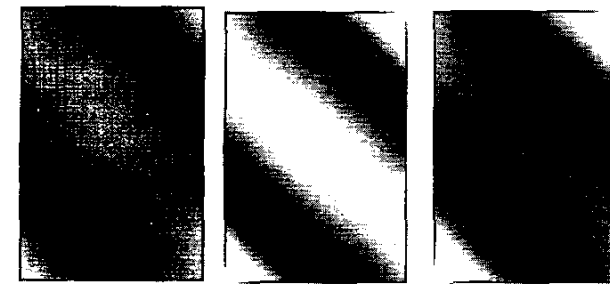
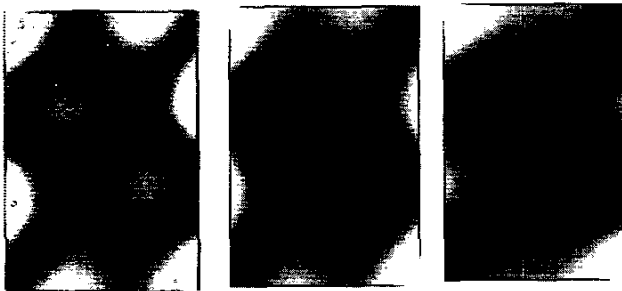
(a) A_g -4 (195 kHz)(b) A_u -6 (240 kHz)(c) B_u -12 (321 kHz)(d) B_g -12 (345 kHz)

Fig. 5. Measured (left) and computed (middle and right) displacement figures on the x face of Specimen E. The frequencies shown in parentheses are measured ones. Bright area implies large displacement amplitude and black area zero amplitude, that is, nodal lines. The maximum displacement amplitude was about 1 nm. In the computations, we used the coefficients taken from Kaminskii *et al.* [2] (middle) and $C_{11} = 200$, $C_{33} = 220$, $C_{12} = 70$, $C_{13} = 80$, $C_{44} = 47$, and $C_{14} = 20$ GPa; $e_{11} = -0.5$, $e_{22} = 0.01$ C/m² (right), which are far away from the true values.

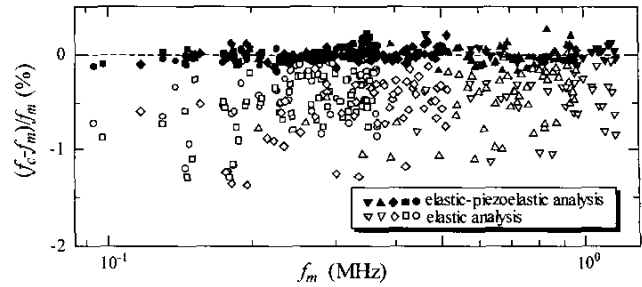


Fig. 6. Differences between the measured (f_m) and calculated (f_c) resonance frequencies of the five specimens. Taking the piezoelectric effect into consideration, the calculations agree with the measurements with 0.065% on average. (triangles pointing up A, triangles pointing down B, diamonds C, squares D, circles E)

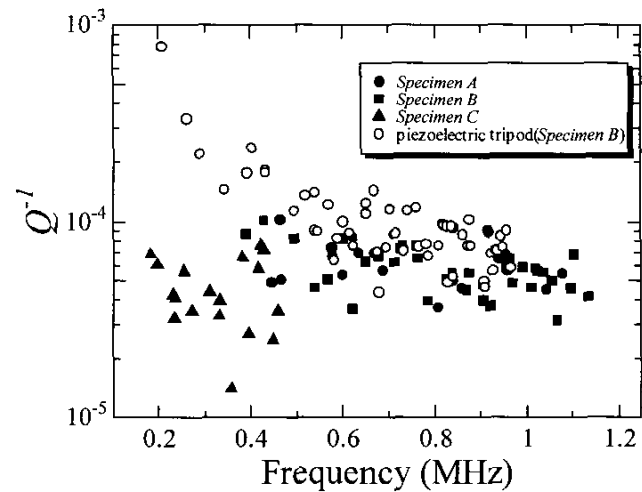


Fig. 7. Internal friction measured by the contactless free-decay method. Open circles denote the peak-width measurements by the piezoelectric tripod.

tion calculations, especially when the modes are not identified [12]–[15]. However, because we can correctly identify the observed modes by referring to the displacement figures, the resulting coefficients are insensitive to the initial guess. Indeed, we always reached exactly the same answer from any previously reported values as an initial guess. Furthermore, the inverse calculation provided the correct answer even with an unreasonable initial guess, listed in the caption of Fig. 5. This is because the displacement-distribution patterns hardly were affected by the material coefficients as demonstrated in Fig. 5. They are unique for individual modes like fingerprints. However, we failed to obtain the true values without referring to the displacement figures, even beginning with a good initial guess by Kaminskii *et al.* [2]. Thus, the correct mode identification is the key for the successful determination of the material coefficients.

Concerning the elastic-stiffness coefficients, our results fall within the previous error limits. For the piezoelectric coefficients, however, our results considerably differ from

the widely ranging previous results; e_{11} is smaller by 1.2–13% and e_{14} is larger by 44–97% than the existing results. We are confident about our results, because we removed the error sources of the conventional measurements; coupling agent and excess load for transduction, electrodes on the specimen surfaces, and use of many specimens oriented in various directions. Moreover, the pulse-echo measurements are vulnerable to noises and waveform change caused by many factors.

B. Q_{ij}^{-1}

Obviously, Q_{11}^{-1} and Q_{33}^{-1} exceed Q_{44}^{-1} and Q_{66}^{-1} by a factor of about 5. We repeated the determination of Q_{ij}^{-1} three times and found that they varied by 20% at most. Thus, this difference is meaningful and indicates that longitudinal-wave attenuation is larger than shear-wave attenuation, a contrary trend to metals where dislocations' anelastic movement causes larger shear-wave Q^{-1} [14], [15].

Possible origin of internal friction of such a brittle material is phonon-phonon interactions. Acoustic waves break an equilibrium state of phonons due to the lattice anharmonicity. The scattered thermal-mode phonons subsequently equilibrate by interacting with a low-frequency-mode (acoustic) phonon and other thermal-mode phonons, during which energy loss arises. Such energy loss can be expressed by the relaxation time τ of an acoustic phonon to relax to a thermal phonon with angular frequency of the acoustic wave ω (Akhieser result) [26]. At room temperature and in the kilohertz frequency region ($\omega\tau \ll 1$), it reduces:

$$Q_{phon}^{-1} = C\omega\tau, \quad (17)$$

where C is a constant depending on the mass density, sound velocity, and the lattice anharmonicity. This theory agreed with measurements of germanium, quartz, and silicon as summarized by Mason [26]. Especially, germanium shows the longitudinal/shear-modes internal-friction ratio about 5, very close to ours on langasite. Usually, the thermal relaxation time for longitudinal waves is about twice that for shear waves because of the difference of the associated volume change [26]. Our Q_{ij}^{-1} then can be interpreted as phonon-phonon interactions.

VII. CONCLUSIONS

We developed the RUS/laser hybrid technique to perform the simultaneous determination of the elastic and piezoelectric coefficients of langasite from a single specimen. Accuracy of the frequency measurement was better than 0.001%. We succeeded in correctly identifying all of the observed resonance peaks by measuring the displacement distributions on a surface using laser-Doppler interferometry. The measured coefficients are independent of the specimen dimensions.

Our elastic coefficients are consistent with those reported previously using conventional measurement methods. Our piezoelectric coefficients were considerably different from those reported in the past.

Our elastic and piezoelectric coefficients are insensitive to the initial guess for iteration owing to correct mode identification.

We measured internal friction using a contactless, free-decay method and deduced the internal friction tensor. Longitudinal-wave internal friction was larger than shear-wave internal friction by a factor of about 5, which was interpreted as phonon-phonon interactions.

ACKNOWLEDGMENTS

We thank Professor I. Ohno (Ehime University) for giving us valuable comments on the free-vibration groups of a trigonal rectangular parallelepiped. K. Yaegawa and Y. Kawasaki (Osaka University) helped us considerably on measurements.

REFERENCES

- [1] T. Sato, M. Murota, and Y. Shimizu, "Characteristics of Rayleigh and leaky surface acoustic wave propagating on a $\text{La}_3\text{Ga}_5\text{SiO}_{14}$ substrate," *Jpn. J. Appl. Phys.*, vol. 37, pp. 2914–2917, 1998.
- [2] A. Kaminskii, I. Silvestrova, S. Sarkisov, and G. Denisenko, "Investigation of trigonal $(\text{La}_{1-x}\text{Nd}_x)_3\text{Ga}_5\text{SiO}_{14}$ crystals," *Phys. State Solids (a)*, vol. 80, pp. 607–620, 1983.
- [3] A. Ilyaev, B. Umarov, L. Shabanova, and M. Dubovik, "Temperature dependence of electromechanical properties of LGS crystals," *Phys. State Solids (a)*, vol. 98, pp. K109–K114, 1986.
- [4] I. Silvestrova, Y. Pisarevskii, P. Senyushchenkov, and A. Krupnyi, "Temperature dependence of the properties of $\text{La}_3\text{Ga}_5\text{SiO}_{14}$ single crystal," *Sov. Phys.—Solid State*, vol. 28, pp. 1613–1614, 1986, through J. Gualtieri, J. Kosinski, A. Ballato, "Piezoelectric materials for acoustic wave applications," *IEEE Trans. Ultrason., Ferroelect., Freq. Contr.*, vol. 41, pp. 53–59, 1994.
- [5] S. Sakharov, P. Senushencov, A. Medvedev, and Y. Pisarevsk, "New data on temperature stability and acoustical losses of langasite crystals," in *Proc. IEEE Ultrason. Symp.*, 1995, pp. 647–652.
- [6] K. Inoue and K. Sato, "Propagation characteristics of surface acoustic waves on langasite," *Jpn. J. Appl. Phys.*, vol. 37, pp. 2909–2913, 1998.
- [7] A. Bungo, C. Jian, K. Yamaguchi, Y. Sawada, S. Uda, and Y. Pisarevsky, "Analysis of surface acoustic wave properties of the rotated Y-cut langasite substrate," *Jpn. J. Appl. Phys.*, vol. 38, pp. 3239–3243, 1999.
- [8] D. Malocha, M. Cunha, E. Adler, R. Smythe, S. Frederick, M. Chou, R. Helmbold, and Y. Zhou, "Recent measurements of material constants versus temperature for langatate, langanite and langasite," in *IEEE/EIA Int. Freq. Contr. Symp. Exhib.*, 2000, pp. 200–205.
- [9] *IEEE Standard on Piezoelectricity*, ANSI/IEEE Std., pp. 176–1978, 1979.
- [10] R. Smith and F. Welsh, "Temperature dependence of the elastic, piezoelectric, and dielectric constants of lithium tantalate and lithium niobate," *J. Appl. Phys.*, vol. 42, pp. 2219–2230, 1971.
- [11] H. Ledbetter, S. Kim, and M. Lei, personal communication, Jan. 2002.
- [12] I. Ohno, "Free vibration of a rectangular parallelepiped crystal and its application to determination of elastic constants of orthorhombic crystals," *J. Phys. Earth*, vol. 24, pp. 355–379, 1976.
- [13] A. Migliori, J. Sarrao, W. Visscher, T. Bell, M. Lei, Z. Fisk, and R. Leisure, "Resonant ultrasonic spectroscopy techniques

for measurement of the elastic moduli of solids," *Physica B*, vol. 183, pp. 1–24, 1993.

- [14] H. Ledbetter, C. Fortunko, and P. Heyliger, "Elastic constants and internal friction of polycrystalline copper," *J. Mater. Res.*, vol. 10, pp. 1352–1353, 1995.
- [15] H. Ogi, H. Ledbetter, S. Kim, and M. Hirao, "Contactless mode-selective resonance ultrasound spectroscopy: Electromagnetic acoustic resonance," *J. Acoust. Soc. Amer.*, vol. 106, pp. 660–665, 1999.
- [16] H. Ledbetter, C. Fortunko, and P. Heyliger, "Orthotropic elastic constants of a boron-aluminum fiber-reinforced composite: An acoustic-resonance-spectroscopy study," *J. Appl. Phys.*, vol. 78, pp. 1542–1546, 1995.
- [17] H. Ogi, K. Takashima, H. Ledbetter, M. Dunn, G. Shimoike, M. Hirao, and P. Bowen, "Elastic constants and internal friction of an SiC-fiber-reinforced Ti-alloy-matrix crossply composite: Measurement and theory," *Acta Materialia*, vol. 47, pp. 2787–2796, 1999.
- [18] H. Ogi, M. Dunn, K. Takashima, and H. Ledbetter, "Elastic properties of a SiCf/Ti unidirectional composite: Acoustic resonance measurements and micromechanics predictions," *J. Appl. Phys.*, vol. 87, pp. 2769–2774, 2000.
- [19] K. Tanaka and M. Koiwa, "Single-crystal elastic constants of intermetallic compounds," *Intermetallics*, vol. 4, pp. S29–S39, 1996.
- [20] I. Ohno, "Rectangular parallelepiped resonance method for piezoelectric crystals and elastic constants of alpha-quartz," *Phys. Chem. Minerals*, vol. 17, pp. 371–378, 1990.
- [21] M. Dunn, H. Ledbetter, and P. Heyliger, "Free vibration of piezoelectric crystals: Application to the determination of elastic and piezoelectric constants," in *Engineering Mechanics*. Washington, DC: ASME, 1995, pp. 758–761.
- [22] W. Johnson, S. Kim, and D. Lauria, "Anelastic loss in langatate," in *Proc. IEEE/EIA Int. Freq. Contr. Symp. Exhib.*, 2000, pp. 186–190.
- [23] M. Hirao, H. Ogi, and H. Fukuoka, "Resonance EMAT system for acoustoelastic stress evaluation in sheet metals," *Rev. Sci. Instrum.*, vol. 64, pp. 3198–3205, 1993.
- [24] B. A. Auld, *Acoustic Fields and Waves in Solids*. vol. 1, New York: Wiley-Interscience, 1973, p. 298.
- [25] E. P. Eer Nisse, "Variational method for electroelastic vibration analysis," *IEEE Trans. Sonics Ultrason.*, vol. SU-14, pp. 153–160, 1967.
- [26] W. Mason, "Effect of impurities and phonon process on the ultrasonic attenuation of germanium, crystal quartz, and silicon," in *Physical Acoustics*. vol. IIIB, W. Mason, Ed. New York: Academic, 1965, pp. 235–286.



Hirotsugu Ogi was born in Kyoto, Japan, on November 2, 1967. He studied at Osaka University, Osaka, Japan, from 1987 to 1993; and earned B.S., M.S., and Ph.D. degrees in mechanical science and engineering at Osaka University, in 1991, 1993, and 1997, respectively.

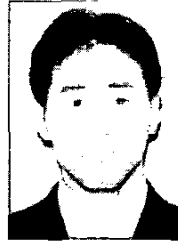
He joined the Graduate School of Engineering Science, Osaka University, in 1993 as a research associate and the Materials Science & Engineering Laboratory, National Institute of Standards and Technology as a guest researcher between 1998 and 1999; he has been an associate professor since 2000.

He is a member of the Acoustical Society of America. His research fields include acoustic-resonance studies for elastic and anelastic behaviors of materials, piezoelectric materials, composites and thin films; and nondestructive evaluation of materials. He was given the 1995 Outstanding Paper Award from the American Society of Nondestructive Testing.

He is a member of the Acoustical Society of America. His research fields include acoustic-resonance studies for elastic and anelastic behaviors of materials, piezoelectric materials, composites and thin films; and nondestructive evaluation of materials. He was given the 1995 Outstanding Paper Award from the American Society of Nondestructive Testing.



Nobutomo Nakamura was born in Okayama, Japan, on February 24, 1979. He is currently a graduated student at Osaka University, Osaka, Japan. He earned a B.S. degree in mechanical science and engineering at Osaka University in 2000.



Keiji Sato was born in Kumamoto, Japan, on June 24, 1976. He is currently a graduated student at Osaka University, Osaka, Japan. He earned B.S. and M.S. degrees in mechanical science and engineering at Osaka University in 2000 and 2002, respectively.



Masahiko Hirao was born in Wakayama, Japan, in October 1951. He studied mechanical engineering at Osaka University, Osaka, Japan, and received B.S., M.S., and Dr.Eng. degrees from Osaka University, in 1974, 1976, and 1982, respectively.

He joined the Graduate School of Engineering Science, Osaka University, in 1977 as a research associate, where, after a postdoctoral year at Cornell University, Ithaca, NY, he is a Professor of the Division of Mechanical Science.

His research interests are in the general area of elastic waves and ultrasonic noncontact characterization of materials. He is principally studying elastic constants and internal friction with electromagnetic acoustic resonance (EMAR) for industrial applications as well as for physical acoustic of materials. He has authored or coauthored over 160 technical publications including receiving two best paper awards. He serves on the Editorial Board of *Research in Nondestructive Evaluation* (ASNT).



Satoshi Uda was born in Aichi, Japan, on May 29, 1955. He majored in geology at the University of Tokyo and received B.S. and M.S. degrees in 1979 and 1981, respectively. He entered Mitsubishi Materials Corp., Japan in 1981 and started the research on crystal growth. He entered the graduate school of Engineering, Department of Materials Science and Engineering, Stanford University, Stanford, CA in 1987 and received M.S. and Ph.D. degrees in 1989 and 1992, respectively. He is currently involved in the growth of oxide crystals and their applications relating to the piezoelectric materials and nonlinear optical materials at Mitsubishi Materials Corp. He also has been a visiting professor at Tokyo Institute of Technology since 1999.

He is currently involved in the growth of oxide crystals and their applications relating to the piezoelectric materials and nonlinear optical materials at Mitsubishi Materials Corp. He also has been a visiting professor at Tokyo Institute of Technology since 1999.