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Second harmonic component of focused sound observed with a concave receiver

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To develop a measurement method for acoustic nonlinearity parameter B/A using an acoustic microscope, the second harmonic component of a focused Gaussian beam observed with a concave receiver, with an aperture large enough to cover the whole beam, set within the post focal region has been investigated. The theory based on the successive approximation solution for the Khokhlov-Zabolotskaya-Kuznetsov equation is validated through the experiment employing a 1.9-MHz focused ultrasound and a concave receiver. Both the fundamental and second harmonic amplitudes attain the maxima at the position where the multiple reflection signal produced between the source and receiver is well detected by the concave receiver. The phase of the second harmonic component relative to the fundamental is the same as that of the axial pressure in particular at this position. These are true even in the case with the insertion of a different liquid layer. The generation of the second harmonic, which corresponds to the linear and nonlinear acoustic properties of the different liquid, is observed with the concave receiver. This suggests that the B/A measurement utilizing the axial pressure of the focused beam may be extended to the data obtained with the concave receiver.

Keywords: Nonlinear acoustics, Focused ultrasound, Second harmonic, Acoustic microscope, Nonlinearity parameter, Finite amplitude method

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1. INTRODUCTION

The present author and his colleagues have analyzed the nonlinearly generated second harmonic component in a focused sound including the problem of its spatial distribution.¹⁻⁵⁾ Based on the finite-amplitude method,⁶⁾ utilizing the second harmonic sound in focused ultrasound, a measurement for the nonlinearity parameter B/A in liquidlike media was also presented and experimented, where the on-axis acoustic pressure beyond the sample was observed at the focal region with a point receiver.⁷⁾ Small volume samples, for example, of 0.5 ml were measured using a 1.9-MHz focusing source.⁸⁾

On the other hand, acoustic microscopes have been applied to various acoustic characterizations and measurements, where high-frequency focused sounds are employed to obtain the high spatial resolution. Although the nonlinear phenomena must take place in the acoustic microscope, the phenomena are never used for the acoustical measurement. The techniques for obtaining nonlinear acoustic imaging⁹⁾ and measuring the nonlinear property¹⁰⁾ of the medium have ever been intended utilizing the second harmonic component in the focused sound of the acoustic microscope. In these proposal, however, the diffraction, which plays an important role in the nonlinear propagation of focused sounds, has scarcely been taken into account regarding to the nonlinear distortion process. By virtue of this, the obtained data do not necessarily correspond to the nonlinear property of the sample medium. Since the diffraction effect is taken into considerations, in

contrast, in the B/A measurement mentioned above, the result is expected to be more accurate in principle, and therefore the measurement may be extended to the acoustic microscope. The acoustic microscope usually detects the whole sound beam using a receiver with a finite aperture, which differs from the point receiver employed in the experiment of the aforementioned B/A measurement. It is difficult to detect the acoustic pressure at a point in the beam of the microscope, because the radius of the focusing source is as small as a few millimeters at most. Thus the above measurement method cannot be immediately applied to the acoustic microscope.

The present article theoretically and experimentally examines the properties of the fundamental and second harmonic components in the focused sound detected by a concave receiver with the aperture as wide as the beam at the post focal region, supposing the receiver is set vertical to the axis of the focusing source. The successive approximation solution for the governing equation is adopted for the theoretical analysis, where the Gaussian beam is assumed for the focused sound. For convenience, the experiment to validate the theory and to confirm the possibility for measuring the B/A is conducted at relatively low frequency of 1.9 MHz.

It has already been reported that the nonlinear phenomena associated with a large amplitude focused sound enhance the resolution of acoustic microscope,¹¹⁾ and give rise to an anomalous phase shift of the sound in the center of the beam.¹²⁾ Since this paper treats the moderate amplitude to which the successive approximation is available, the present focusing field is free from such phenomena.

2. FREE FIELD DETECTED BY CONCAVE RECEIVER

2.1 Theory

Suppose that a spherical focusing source with the focal length D emits a sinusoidal sound of angular frequency ω along the z axis as shown in Fig. 1, where the medium is water. The sound source is assumed to be a Gaussian source with the on-source pressure amplitude of $p_0 \exp(-\xi r^2)$. The focused sound is detected by a concave receiver whose curvature and aperture radii and d and a, respectively. Taking into the consideration the harmonic components up to the second order, the acoustic pressure at the point (r, z) is assumed to be

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Fig. 1 Geometry and notation.

 $p_{1w}(r, z) \exp(-j\omega\tau) + p_{2w}(r, z) \exp(-j2\omega\tau)$. Here τ is the retarded time $t - z/c_w$, where t is the time, and c_w is the sound speed in water. Then the fundamental and second harmonic components in the output voltage of the concave receiver are approximately proportional to the quantities,

$$P_{1w} = \int_{0}^{a} p_{1w}(r, z) \exp\left(-j\frac{k_{w}r^{2}}{2d}\right) r dr, \qquad (1)$$

$$P_{2w} = \int_0^a p_{2w}(r, z) \exp\left(-j \frac{k_w r^2}{d}\right) r dr , \qquad (2)$$

where k_w is the wavenumber of the fundamental in water ω/c_w . Since the proportion factor, which should depend primarily on the receiver sensitivity, is not significant in the discussion that follows, the output voltage is assumed to be $P_w(z) = P_{1w}(z) \times$ $\exp(-j\omega\tau) + P_{2w}(z) \exp(-j2\omega\tau)$ for simplicity. Using the successive approximation solution for the Khokhlov-Zabolotskaya-Kuznetsov equation,³⁾

$$p_{1w}(r, z) = -j \frac{k_w p_0 e^{-\alpha_w z}}{h(z)} \exp\left[j \frac{k_w r^2}{2z} \left\{1 + j \frac{k_w}{h(z)}\right\}\right],$$
(3)

$$p_{2w}(r, z) = j \frac{\beta_{w} k_{w}^{3} p_{0}^{2} e^{-4\alpha_{w} z}}{2\rho_{w} c_{w}^{2} h(z)} \exp\left[j \frac{k_{w} r^{2}}{z} \left\{1 + j \frac{k_{w}}{h(z)}\right\}\right]$$
$$\cdot \int_{0}^{z} \frac{e^{2\alpha_{w} z'}}{h(z')} dz', \qquad (4)$$

the following relations are derived.

$$P_{1w}(z) = -\frac{k_w p_0}{2g(z)h(z)} e^{-\alpha_w z} [e^{jg(z)a^2} - 1], \quad (5)$$

$$P_{2w}(z) = \frac{\beta_{w}k_{w}^{3}p_{0}^{2}e^{-4\alpha_{w}z}}{8\rho_{w}c_{w}^{2}g(z)h(z)}[e^{j2g(z)a^{2}}-1]$$

$$\cdot \int_{0}^{z} \frac{e^{2\alpha_{w}z'}}{h(z')}dz', \qquad (6)$$

where $h(z) = 2\xi z - jk_w(1 - z/D), \quad g(z) = k_w[1 + jk_w/D]$

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 $h(z)]/2z - k_w/2d$, α_w is the attenuation coefficient in water at the fundamental frequency, ρ_w is the density of water, and β_w is the acoustic nonlinearity parameter $1 + (B/A)_w/2$ of water.

Comparing the ratio of Eqs. (3) and (5) with the ratio of Eqs. (4) and (6), one obtains

$$\frac{P_{2w}(z)}{P_{1w}(z)} = \frac{1}{2} \frac{e^{j2q(z)a^2} - 1}{e^{jg(z)a^2} - 1} \frac{p_{2w}(0, z)}{p_{1w}(0, z)}$$
$$\approx \frac{1}{2} \frac{p_{2w}(0, z)}{p_{1w}(0, z)}. \quad (a \gg 0)$$
(7)

Consequently the distortion ratio of the received waveform becomes half that of the waveform of the axial pressure at arbitrary z when the aperture diameter 2a is larger than the width of focused beam. The phase parameter $\phi_w(z) = \angle p_{2w}(0, z) - 2 \angle p_{1w}(0, z) + \pi/2$ was defined to express the phase delay of the zero-crossing of the axial second harmonic pressure relative to that of the axial fundamental pressure. The phase parameter for the signal detected by the concave receiver, $\Phi_w(z) = \angle P_{2w}(z) - 2 \angle P_{1w}(z) + \pi/2$, is also defined for expressing the phase delay of the zero-crossing of P_{2w} relative to that of P_{1w} . They both depend on the range z. Their difference is derived from Eqs. (3)-(6) as

$$\Phi_{w}(z) - \phi_{w}(z) = \angle [e^{j^{2}g(z)a^{2}} - 1]$$

-2\angle [e^{jg(z)a^{2}} - 1] + \angle g(z) + \frac{\pi}{2}
\approx \angle g(z) - \frac{\pi}{2}. (a\approx 0) (8)

When the aperture radius is large so that the concave receiver detects whole focused beam, both $\exp[jg(z)a^2]$ and $\exp[j2g(z)a^2]$ vanish because $\operatorname{Im}[g(z)a^2]$ is much larger than unity.

2.2 Experiment

The burst wave emitted from a concave PZT transducer of frequency $\omega/2\pi = 1.9$ MHz and focal length D=85 mm was experimentally observed with a concave PZT transducer with aperture radius a=20 mm, curvature radius d=42 mm and the thickness resonant frequency of 4 MHz. The sound source is approximated as the Gaussian source with the coefficient $\xi = 2300 \text{ m}^{-2.7}$ Figure 2(a) compares the measured z dependence of P_{1w} and P_{2w} with calculated ones obtained from Eqs. (3) and (4). The acoustic properties of water as $\rho_w = 1000 \text{ kg/m}^3$, $c_w = 1483 \text{ m/s}$, $\alpha_w = 0.1 \text{ Np/m}$ and $(B/A)_w = 5.0$ are



Fig. 2 Range dependence of received sound in free field. (a) Harmonic amplitudes. (b) Phase parameter.

supposed in the calculation. The experimental value shows good correspondence with the calculated value.

The distortion factor of the axial pressure does not strongly depend on z beyond the focus.⁴⁾ Then Eq. (7) means that neither does the distortion ratio of the concave receiver output. Hence both the fundamental and second harmonic amplitudes vary with z almost in the same manner within the experimental range. However, since the focused beam spherically diverges beyond the focal region, the shape of the wavefront fits the concave surface near the range z = D + d, where the concave receiver most effectively detects both the fundamental and second harmonic components so that their amplitudes attain the maxima. This range at which the maximum amplitudes are obtained is named z_w here. Figure 2(b) compares the observed phase parameter with the calculated phase parameter shown by a solid curve. Large errors may be contained in the measured phase parameter because of extremely frequency dependent sensitivity of the experimental receiver with the resonance frequency at 4 MHz, to

which the frequency of second harmonic sound is close. Nevertheless the measured result for the z dependence of the phase parameter agrees well with the calculated. The measurement error is, therefore, rather small. At the range $z=z_w$ where the maximum amplitude is attained, the calculated axial phase parameter ϕ_w shown by a dot curve coincides with the value Φ_w of the solid curve.

These can be explained well by the behavior of g(z) in Eqs. (5) and (6) whose real part is the most sensitive to z. Where the shape of the wavefront fits the concave surface, the real part of g(z) in the denominator of Eqs. (5) and (6) almost vanishes and then |g(z)| takes the minimum to make $\angle g(z) = \pi/2$. Then the amplitudes of P_{1w} and P_{2w} take the maxima, and the relation $\Phi_w = \phi_w$ holds due to Eq. (8).

From the above discussion, it is clarified that the detection of signal P_w at $z=z_w$ brings the information both for amplitudes and phase parameter similar to those of the axial pressure. This suggests that, if the focused sound is detected at the range where the amplitude attains the maximum, the same procedure as the B/A measurement utilizing the axial pressure becomes applicable.

Since the acoustic wave reflected from the receiver set at $z = z_w$ traces back to the source and is reflected again as were it radiated from the source, the wavefront of the wave arriving at the receiver fits the receiver curvature again. By the repetition of this phenomenon, the multiple reflection takes place between the source and receiver. Hence, when the concave receiver is set at $z = z_w$, the receiver gets the multiple reflection of the burst wave as shown in Fig. 3. In other words, the receiver can be correctly set at the position of $z = z_w$ by adjusting the receiver location to observe the multiple reflection well. In the following experiment, since the range and direction of the receiver can be adjusted by using such mechanical stages as an x - y - z stage, a gonio stage, and a rotary stage, it is easy to find the position to get the multiple reflection signals. Here the fundamental and second harmonic components, of course, of the first burst of the reflection signals as shown in Fig. 3 are of interest.

An acoustic beam is, in general, expressed by the superposition of various plane waves with different complex amplitudes and propagation directions.¹³⁾ The concave receiver located at $z=z_w$ attains high sensitivity to obtain high S/N ratio, because it



Fig. 3 Observed multiple reflection signals.



Fig. 4 Plane wave obtained with triple transition.¹⁶

detects all the plane waves. This is an advantage of the concave receiver. When a planar receiver^{10,14}) is employed instead of the concave receiver, the receiver output is significantly reduced because only a portion of the plane waves, whose wavefront fits the planar surface, can be detected. In addition, the second harmonic sounds detected by the planar receiver includes the portion generated by the mutual interaction with other plane waves, which are different from the detected plane waves. By virtue of this, the relation of the detected fundamental and second harmonic sounds is not always easy to interpret. This may cause the discrepancy of the measured results from the theoretical prediction as seen in the references.^{10,15)} In order to avoid the disadvantage of the planar receiver, the use of the reflected acoustic wave has previously been dem-As illustrated in Fig. 4, the planar onstrated.¹⁶⁾ receiver is set such that the wave, reflected at the receiver and once more at the focusing source, becomes a planar wave. Then even the planar receiver gets high sensitivity. However, since the amplitude of the reflected wave depends on the reflection coefficients both at the source and receiver, and further the acoustic wave transmits three times

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a sample which would be inserted at the focus illustrated as the point F in Fig. 4, the process of nonlinear distortion is rather complicated. The application to the B/A measurement, therefore, is inappropriate. The use of concave receiver is more useful.

3. FIELD WITH DIFFERENT LIQUID LAYER

3.1 Theory

Suppose that a different liquid layer of the thickness L is set vertical to the axis such that the center of the layer locates at z=D. The density, sound speed, and acoustic nonlinearity parameter in the layer are denoted by ρ , c, and $\beta = 1 + (B/A)/2$, respectively. The attenuation coefficient in the liquid layer is assumed to be α_1 at the fundamental frequency and α_2 at the second harmonic frequency. The focused beam transmitting through the sample is detected by the concave receiver. Then the fundamental and second harmonic components of the acoustic pressure averaged on the receiver are defined as follows:

$$P_{1s} = \int_{0}^{a} p_{1s}(r, z) \exp\left(-j \frac{k_{w} r^{2}}{2d}\right) r dr , \qquad (9)$$

$$P_{2s} = \int_{0}^{a} p_{2s}(r, z) \exp\left(-j\frac{k_{\pi}r^{2}}{d}\right) r dr . \qquad (10)$$

Applying the fundamental and second harmonic pressure at an arbitrary point beyond the different liquid layer;

$$p_{1s}(r, z) = -j \frac{k_{w} T_{11} T_{01}}{h(z_{s})} p_{0} \exp[-\alpha_{w}(z-L) - \alpha_{1}L] \cdot \exp\left[j \frac{k_{w} r^{2}}{2z_{s}} \left\{1 + j \frac{k_{w}}{h(z_{s})}\right\}\right], \quad (11)$$

$$p_{2s}(r, z) = j \frac{k_{w}^{3} p_{0}^{2}}{2\rho_{w} c_{w}^{2} h(z_{s})} \exp[-4\alpha_{w}(z-L) - \alpha_{2}L]$$

$$\cdot \exp\left[j \frac{k_{w}r^{2}}{z_{s}} \left\{1 + j \frac{k_{w}}{h(z_{s})}\right\}\right]$$

$$\cdot \left(\beta_{w} T_{12} T_{02} \int_{0}^{z_{1}} \frac{e^{2\alpha_{w}z'}}{h(z')} dz'$$

$$+ \beta T_{11}^{2} T_{02} \frac{\rho_{w} c_{w}^{3}}{\rho c^{3}}$$

$$\cdot e^{2\alpha_{w}z_{1}} \int_{0}^{L} \frac{\exp[(\alpha_{2} - 2\alpha_{1})z']}{h(z_{1} + cz'/c_{w})} dz'$$

$$+ \beta_{w} T_{11}^{2} T_{01}^{2} \exp[2\alpha_{w} z_{1} + (\alpha_{2} - 2\alpha_{1})L]$$

$$\cdot \int_{0}^{z-z_{0}} \frac{e^{2\alpha_{w}z'}}{h(z' + z_{1} + cL/c_{w})} dz'\right), \quad (12)$$

the fundamental and second harmonic components of the receiver output is derived as follows:

$$P_{1s}(z) = -\frac{k_{w}T_{11}T_{01}}{2g(z_{a})h(z_{a})}p_{0}$$

$$\cdot \exp[-\alpha_{w}(z-L) - \alpha_{1}L][e^{jg(z_{a})a^{2}} - 1], \quad (13)$$

$$P_{2s}(z) = \frac{k_{w}^{3}p_{0}^{2}}{8\rho_{w}c_{w}^{2}g(z_{a})h(z_{a})}$$

$$\cdot \exp[-4\alpha_{w}(z-L) - \alpha_{2}L][e^{j2g(z_{a})a^{2}} - 1]$$

$$\cdot \left(\beta_{w}T_{12}T_{02}\int_{0}^{z_{1}}\frac{e^{2\alpha_{w}z'}}{h(z')}dz' + \beta T_{11}^{2}T_{02}\frac{\rho_{w}c_{w}^{3}}{\rho c^{3}}e^{2\alpha_{w}z_{1}}$$

$$\cdot \int_{0}^{L}\frac{\exp[(\alpha_{2} - 2\alpha_{1})z']}{h(z_{1} + cz'/c_{w})}dz' + \beta_{w}T_{11}^{2}T_{01}^{2}$$

$$\cdot \exp[2\alpha_{w}z_{1} + (\alpha_{2} - 2\alpha_{1})L]$$

$$\cdot \int_{0}^{z-z_{0}}\frac{e^{2\alpha_{w}z'}}{h(z' + z_{1} + cL/c_{w})}dz'\right), \quad (14)$$

where $z_a = z + (c/c_w - 1)L$, $z_I = D - L/2$, and $z_0 = D + L/2$. The T_{In} and T_{On} are the transmission coefficients of the sound pressure for the *n*-th harmonic component (n=1, 2) at the interfaces of $z = z_I$ and $z = z_0$, respectively: That is

$$T_{\rm In} = \frac{2\rho c(1+jn\alpha_{\rm w}/k_{\rm w})}{\rho_{\rm w} c_{\rm w}(1+j\alpha_{\rm n}/nk) + \rho c(1+jn\alpha_{\rm w}/k_{\rm w})}, \quad (15)$$
$$= \frac{2\rho_{\rm w} c_{\rm w}(1+j\alpha_{\rm n}/nk)}{2\rho_{\rm w} c_{\rm w}(1+j\alpha_{\rm n}/nk)}, \quad (16)$$

$$T_{0n} = \frac{2\rho_{\rm w}c_{\rm w}(1+j\alpha_n/nk)}{\rho_{\rm w}c_{\rm w}(1+j\alpha_n/nk) + \rho c(1+jn\alpha_{\rm w}/k_{\rm w})} \,. \tag{16}$$

By the manner similar to that dealing with the free field, one obtains

$$\frac{P_{2s}(z)}{P_{1s}(z)} = \frac{1}{2} \frac{e^{j_{2g}(z_{a})a^{2}} - 1}{e^{j_{g}(z_{a})a^{2}} - 1} \frac{p_{2s}(0, z)}{p_{1s}(0, z)}$$
$$\approx \frac{1}{2} \frac{p_{2s}(0, z)}{p_{1s}(0, z)}, \quad (a \gg 0)$$
(17)

$$\Phi_{s}(z) - \phi_{s}(0, z) = \angle [e^{j_{2}g(z_{\mathbf{k}})a^{2}} - 1] - 2 \angle [e^{j_{g}(z_{\mathbf{k}})a^{2}} - 1] + \angle g(z_{\mathbf{k}}) + \frac{\pi}{2}$$
$$\approx \angle g(z_{\mathbf{k}}) - \frac{\pi}{2} . \quad (a \gg 0)$$
(18)

3.2 Experiment

Using the aforementioned source and receiver, the experiment to detect the beam transmitting through a 20-mm thick benzyl alcohol layer was carried out. The benzyl alcohol was filled in the cubic case made from brass, which had the 17-mm diameter circular windows on both sides covered with a polyvinyliden



Fig. 5 Range dependence of received sound with inserted sample layer. (a) Harmonic amplitudes. (b) Phase parameter.

chloride film of 10- μ m thickness as the window material. Then the layer was set such that the center located at z=85 mm. Figures 5(a) and 5(b) compare the experimental results for the z dependence of the amplitudes of P_{1s} and P_{2s} and the phase parameter $\Phi_{s}(z) = \angle P_{2s}(z) - 2 \angle P_{1s}(z) + \pi/2$ with the calculated ones. The acoustic property of the benzyl alcohol assumed in the calculation is $\rho =$ 1045 kg/m³, c=1540 m/s, $\alpha_1=0.38$ Np/m, $\alpha_2=4\alpha_1$, and B/A=10.2. The experimental result agrees well with the calculated result both in amplitude and phase. Even in this case, both the amplitudes of the fundamental and second harmonic attain the maxima near $z=z_w$.

When P_{1w} of Eq. (5) is compared with P_{1s} of Eq. (13), the z dependence of P_{1w} is almost determined by the term $\{\exp[jg(z)a^2]-1\}/g(z)h(z)$ in the regime not far from $z=z_w$, while the z dependence of P_{1s} is determined by $\{\exp[jg(z_a)a^2]-1\}/g(z_a)h(z_a)$. The difference is only that z for P_{1w} has been replaced with z_a for P_{1s} . If P_{1w} takes the maximum at $z=z_w$, thus, P_{1s} attains the maximum J. Acoust. Soc. Jpn. (E) 16, 1 (1995)





at $z_a = z_w$, that is $z = z_w - (c/c_w - 1)L$. This predicts that, when the sound speed in the liquid layer differs from that in water, the wave fronts of focused beam must fit the concavity of the receiver at the position shifted toward the source from $z = z_w$ by $(c/c_w - 1)L$ due to refraction of beam. The position to obtain the maximum amplitude in the case with the different liquid layer is called z_s here. In the case having a benzyl alcohol layer whose sound speed is close to water, it holds that $z_s = z_w$.

The calculated value of the axial phase parameter Φ_s is shown by the dotted line in Fig. 5(b). The value of Φ_s at $z=z_s$ turns to be almost the same as the value of ϕ_s . Since Eqs. (17) and (18) hold in the field with a different liquid layer as do so Eqs. (7) and (8) in the free field, both the fundamental and the second harmonic amplitudes take the maxima

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simultaneously at a certain range from the concave receiver. The phase parameter at this range also equals to that of axial pressure.

The harmonic amplitudes and phase parameter of the output voltage of the concave receiver set at the maximum positions $(z = z_w \text{ and } z = z_s)$ measured at various transmitting sound levels are shown in Figs. 6(a) and 6(b) for the cases with and without the insertion of the benzyl alcohol layer. The plots with the solid and hollow circles indicate the experimental results for the case without and with the layer, respectively. The abscissa indicates the input voltage of an rf power amplifier to drive the experimental focusing source, to which the source pressure p_0 is proportional. According to the calculation using Eqs. (5), (6), (13), (14), (15) and (16), the insertion of the benzyl alcohol layer must reduce the fundamental amplitude by 3% and enhance the second harmonic amplitude by 32%. Further, the phase parameter must keep almost constant accompanying only 0.08 rad increase. The experimental results agree well with the prediction within the small amplitude range. The increase of the second harmonic component is primarily caused by the large nonlinearity parameter of the benzyl alcohol. Consequently, the observation using a concave receiver may be employed for measuring the B/A of an inserted layer. The detail of the B/Ameasurement will be discussed in the subsequent paper.

4. CONCLUSION

In order to develop the B/A measurement method using acoustic microscope, the fundamental and nonlinearly generated second harmonic components detected by a concave receiver were examined for the cases without and with an inserted liquid layer. There exists the receiver position where the amplitudes of both the fundamental and second harmonic in the receiver output attain the maxima. Their amplitude ratio and phase parameter are in the simple relation with those of the axial pressure at this position. The experimental results for the case of the benzyl alcohol sample agreed well with the theoretical prediction. Hence the B/A measurement utilizing the axial pressure may be extended to the case using a concave receiver. The detail of the B/A measurement will be discussed in the subsequent paper.

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