Influence of inserted sample on second harmonic component in a finite-amplitude focused sound

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The influence of a liquid sample layer on the nonlinearly generated second harmonic component is examined by computations and experiments. The sample with plane surfaces is inserted in the focal region normal to the axis, whose acoustical properties are different from those of surrounding medium (water). The theory is based on the quasi-linear parabolic approximation, valid for the case of weak distortion and small concavity. A linear transmission through the boundaries is also applied for the second harmonics. The analysis for a source with a Gaussian profile excitation shows that, in the case of a lossless sample, the second harmonic component generated at the sample can be the greater part of the second harmonic sound propagating in the post focal region. In the case of a lossy sample, a newly introduced quantity C_s evaluated from the waveform at the post focal region corresponds well to the sample properties excepting the linear absorption. The experimental results suggest the usefulness of the quantity C_s .

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1. INTRODUCTION

A spherical focusing source is widely used in order to get a high lateral resolution for various applications, including acoustical microscopy and biomedical ultrasonics. In these focusing sources, the radiated sound of an initially sinusoidal wave can distort with the propagation because of the nonlinear properties primarily due to the elastic nonlinearity of the medium. The second harmonic component is readily found because its amplitude is the greatest among higher harmonic components. The present paper treats the nonlinearly generated second harmonic sound in a focused sound field.

Previous experiments have shown that the higher harmonics are also focused into the focal point, in other words they also reach maximum near the focus.¹⁾ One may think that this phenomenon can occur because the nonlinear effect is accentuated in

the vicinity of the focus where the sound amplitude attains the maximum. However, an exact physical interpretation is needed to account for diffraction effects.²⁾ Lucas and Muir gave an analytical method for the second harmonic sound everywhere in the nonlinear focusing field taking into consideration the diffraction effect.³⁾ The present authors and Muir showed good agreement between the experiments and calculations using the theory of Lucas and Muir.⁴⁾ Based on this analysis, it was found that the phase reversal due to the diffraction of the sound while propagating at the focal region causes second harmonic component generated at the pre-focal region to be cancelled by those generated at the post-focal region. Therefore the second harmonic component generated near the focal point can be the dominant contributor to the propagating second harmonic sound.⁴⁾ This fact also means that the nonlinear effect is, without doubt, the greatest at the focal region. This can be applied to measure the nonlinear characteristics of small samples immersed in the focal region.

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In a previous paper,⁵⁾ the present authors proposed a method to selectively detect the second harmonic component generated at the focal region separately from the second harmonic components generated at other regions, and the experimental results using a liquid sample showed the validity of this method. However the previous paper only treated a liquid sample whose nonlinearity parameter β differed from surrounding medium (water) for simplicity. The present paper investigates the influence of more realistic liquid samples whose density ρ and sound speed c also differ from those of water. The sound source is assumed to have a radially shaded velocity excitation whose profile is characterized by a Gaussian distribution. This type of source facilitates the field calculations and makes a simple radiation field without pressure peaks and nulls due to source-edge diffractions. The discussion also takes linear absorption into account.

2. THEORETICAL FORMULATION

Suppose that a sinusoidal sound wave with angular frequency ω radiates from the spherical focusing source with the geometrical focal length D , as shown in Fig. 1. The sound speed, density, linear absorption coefficient, and parameter of nonlinearity of the medium are denoted by c , ρ , α , and β respectively. The absorption coefficient is assumed to be the value at the fundamental angular frequency ω . Cylindrical coordinates r and z are used, as shown in the figure. Supposing that a particle velocity $u(r, z)$ is defined by a velocity potential $\phi(r, z)$ as $u(r, z) = \nabla \phi(r, z)$ in uniform media, $\phi(r, z)$ satisfies the following nonlinear equation.⁶⁾

$$\left(1 + \frac{2\alpha}{\omega k} \frac{\partial}{\partial t}\right) \nabla^2 \phi - \frac{1}{c^2} \frac{\partial^2 \phi}{\partial t^2} = \frac{1}{c^2} \frac{\partial}{\partial t} \left[(V\phi)^2 + \frac{\beta-1}{c^2} \left(\frac{\partial \phi}{\partial t} \right)^2 \right], \quad (1)$$

where t is the time, and k is the wavenumber ω/c . Note that Eq. (1) premises the frequency squared law of the absorption coefficient. An infinitely broad liquid layer set by the insertion of a sample, whose sound speed, density, absorption coefficient, and parameter of nonlinearity are c_L , ρ_L , α_L and β_L respectively, is centered on the plane $z = D$ perpendicular to the axis as shown in Fig. 1. Then, Eq. (1) containing c_L , ρ_L , α_L , and β_L instead of c , ρ , α , and β respectively, is assumed to be satisfied in this sample.

Fig. 1 Geometry and notation.

By the perturbation formulation valid for cases involving weak distortion, the equations for the velocity potential of the fundamental component $\phi_1(r, z)$ and the velocity potential of the second harmonic component $\phi_2(r, z)$ are given by

$$\left(1 + \frac{2\alpha}{\omega k} \frac{\partial}{\partial t}\right) \nabla^2 \phi_1 - \frac{1}{c^2} \frac{\partial^2 \phi_1}{\partial t^2} = 0 \quad (2)$$

and

$$\begin{aligned} &\left(1 + \frac{2\alpha}{\omega k} \frac{\partial}{\partial t}\right) \nabla^2 \phi_2 - \frac{1}{c^2} \frac{\partial^2 \phi_2}{\partial t^2} \\ &= \frac{1}{c^2} \frac{\partial}{\partial t} \left[(V\phi_1)^2 + \frac{\beta-1}{c^2} \left(\frac{\partial \phi_1}{\partial t} \right)^2 \right]. \end{aligned} \quad (3)$$

The fundamental velocity potential $\phi_1(r, z)$ is described in complex notation by

$$\phi_1(r, z) = q_1(r, z) \exp \left[-j\omega \left(t - \frac{z}{c} \right) \right]. \quad (4)$$

Assuming small linear absorption, i.e. $\alpha \ll k$, and applying the parabolic approximation appropriate for the case of low concavity,⁷⁾ Eq. (2) is reduced to

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial q_1}{\partial r} \right) + j2k \frac{\partial q_1}{\partial z} + j2\alpha k q_1 = 0. \quad (5)$$

After use of a Hankel transform

$$\tilde{q}_1(s, z) = \int_0^\infty q_1(r, z) J_0(sr) r dr,$$

Eq. (5) becomes

$$\frac{d\tilde{q}_1}{dz} + \left(\alpha + j \frac{s^2}{2k} \right) \tilde{q}_1 = 0.$$

The use of the inverse Hankel transform on the solution of the above equation leads to the solution of Eq. (5), which is

$$\begin{aligned} q_1(r, z) = &-j \frac{k}{z-d} \exp \left[j \frac{kr^2}{2(z-d)} - \alpha(z-d) \right] \\ &\cdot \int_0^\infty q_1(x, d) J_0 \left(\frac{krx}{z-d} \right) \exp \left[j \frac{kx^2}{2(z-d)} \right] x dx, \end{aligned} \quad (6)$$

where d is the z coordinate of an arbitrary plane perpendicular to the axis, and x is the integral variable. Hence $q_1(r, d)$ means the complex amplitude of the fundamental velocity potential given on the plane $z=d$.

The velocity potential of the second harmonic component $\phi_2(r, z)$ is described in complex notation by

$$\phi_2(r, z) = q_2(r, z) \exp \left[-j2\omega \left(t - \frac{z}{c} \right) \right]. \quad (7)$$

Using the parabolic approximation again, the equation for the complex amplitude of the second harmonic radiation $q_2(r, z)$ becomes

$$\begin{aligned} \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial q_2}{\partial r} \right) + j4k \frac{\partial q_2}{\partial z} + j16\alpha k q_2 \\ = j \frac{\beta k^3}{c} q_1^2. \end{aligned} \quad (8)$$

Applying the Hankel transform on both sides of the above equation, one derives

$$\frac{d\tilde{q}_2}{dz} + \left(4\alpha + j \frac{s^2}{4k} \right) \tilde{q}_2 = \frac{\beta k^3}{4} \tilde{q}_1^2.$$

Inversely transforming the solution of this equation, one finally obtains the solution of Eq. (8).

$$\begin{aligned} q_2(r, z) \\ = -j \frac{2k}{z-d} \exp \left[j \frac{kr^2}{z-d} - 4\alpha(z-d) \right] \\ \cdot \int_0^\infty q_1(x, d) J_0 \left(\frac{2krx}{z-d} \right) \exp \left[j \frac{kx^2}{z-d} \right] x dx \\ + j \frac{\beta k^5}{2c} \exp [-4\alpha(z-d)] \int_{z'=0}^z \int_{y=0}^\infty \int_{x=0}^\infty \int_{r'=0}^\infty \\ \cdot \frac{q_1(x, d) q_1(y, d)}{(z-z')(z'-d)^2} J_0 \left(\frac{kr'x}{z'-d} \right) J_0 \left(\frac{kr'y}{z'-d} \right) \\ \cdot J_0 \left(\frac{2kr r'}{z'-d} \right) \exp \left[j \frac{k(x^2+y^2)}{2(z'-d)} \right] \exp [2\alpha(z'-d)] \\ \cdot \exp \left[j \frac{k}{z-z'} \left(r^2 + \frac{z-d}{z'-d} r'^2 \right) \right] \\ \cdot r' xy dr' dx dy dz', \end{aligned} \quad (9)$$

where $q_2(r, d)$ is the amplitude of the second harmonic given on the plane $z=d$, and y and r' are the integral variables.

For simplicity suppose that a source whose radial shading of surface velocity is given by a Gaussian density function. Then one imposes boundary conditions available for the case of low source concavity, presented at $z=0$ instead of those at the spherical surface of the source.

$$\begin{aligned} q_1(r, 0) &= \frac{u_0}{jk} \exp \left[- \left(A + j \frac{k}{2D} \right) r^2 \right], \\ q_2(r, 0) &= 0, \end{aligned} \quad (10)$$

where A is the coefficient that determines the source distribution, and u_0 is the velocity amplitude of the source at $r=0$.

With regard to the boundary between water and liquid sample, even though transmission of finite-amplitude bounded beam has not been analyzed, it has been shown that the second harmonic component in finite amplitude sound radiated from a plane piston reflects and transmits at the boundary as predicted in the linear theory, just as the fundamental component.^{8,9)} Thus, the boundary condition that both the particle velocity and the sound pressure are continuous in the transition through the boundary may be adopted here. A normally incident sound for the case of small concavity ($D\sqrt{A} > 3$) is assumed. Employing the velocity-potential transmission ratios¹⁰⁾ for the angular frequencies ω and 2ω at the boundary $z=z_1$ between two dissipative fluid media;

$$\begin{aligned} T_{11} &= \frac{2\rho c_L \left(1 + j \frac{\alpha}{k} \right)}{\rho c \left(1 + j \frac{\alpha_L}{k_L} \right) + \rho_L c_L \left(1 + j \frac{\alpha}{k} \right)}, \\ T_{12} &= \frac{2\rho c_L \left(1 + j \frac{2\alpha}{k} \right)}{\rho c \left(1 + j \frac{2\alpha_L}{k_L} \right) + \rho_L c_L \left(1 + j \frac{2\alpha}{k} \right)}, \end{aligned}$$

and those at the boundary $z=z_2$ for the angular frequencies ω and 2ω ;

$$\begin{aligned} T_{01} &= \frac{2\rho_L c \left(1 + j \frac{\alpha_L}{k_L} \right)}{\rho c \left(1 + j \frac{\alpha_L}{k_L} \right) + \rho_L c_L \left(1 + j \frac{\alpha}{k} \right)}, \\ T_{02} &= \frac{2\rho_L c \left(1 + j \frac{2\alpha_L}{k_L} \right)}{\rho c \left(1 + j \frac{2\alpha_L}{k_L} \right) + \rho_L c_L \left(1 + j \frac{2\alpha}{k} \right)}, \end{aligned}$$

the condition at both boundaries are provided by

$$\begin{aligned} q_1(r, z_{1+}) &= T_{11} q_1(r, z_{1-}), \\ q_2(r, z_{1+}) &= T_{12} q_2(r, z_{1-}), \\ q_1(r, z_{2+}) &= T_{01} q_1(r, z_{2-}), \end{aligned} \quad (11)$$

and

$$q_2(r, z_{2+}) = T_{02} q_2(r, z_{2-}).$$

Here $z = z_{i+}$ and z_{i-} ($i = 1, 2$) imply the right and left planes adjacent to the boundary plane $z = z_i$, and $k_L = \omega/c_L$. Though multiple reflections, which would occur in the sample layer, are ignored in Eq. (11), there is no disadvantage in many practical situations when short cw pulses are employed.

Applying the above conditions, the use of given values of $q_1(r, 0)$ and $q_2(r, 0)$ leads to the values of $q_1(r, z)$ and $q_2(r, z)$ in the region before the sample ($z < z_1$).

$$q_1(r, z) = -\frac{u_0 \exp[-\alpha z]}{h(z)} \exp \left[j \frac{kr^2}{2z} \left\{ 1 + j \frac{k}{h(z)} \right\} \right], \quad (12)$$

$$q_2(r, z) = \frac{\beta k^2 u_0^3 \exp[-4\alpha z]}{4ch(z)} \exp \left[j \frac{kr^2}{z} \left\{ 1 + j \frac{k}{h(z)} \right\} \right] \cdot \int_0^z \frac{\exp[2\alpha z']}{h(z')} dz', \quad (13)$$

where $h(z) = \{(2A - jk(1/z - 1/d))z\}$. One evaluates the values of $q_1(r, z_{1+}) = T_{11}q_1(r, z_{1-})$ and $q_2(r, z_{1+}) = T_{12}q_2(r, z_{1-})$ from Eqs. (12) and (13) respectively, by putting $z = z_1$. Then q_1 and q_2 in the sample ($z_1 < z < z_2$) are given by

$$q_1(r, z) = -\frac{T_{11}u_0}{h(z_a)} \exp[-\{\alpha z_1 + \alpha_L(z - z_1)\}] \cdot \exp \left[j \frac{kr^2}{2z_a} \left\{ 1 + j \frac{k}{h(z_a)} \right\} \right], \quad (14)$$

$$q_2(r, z) = \frac{k^2 u_0^3}{4ch(z_a)} \exp[-4\{\alpha z_1 + \alpha_L(z - z_1)\}] \cdot \exp \left[j \frac{kr^2\{h(z_1) + jk\}}{h(z_a)z_1} \right] \cdot \left[\beta T_{12} \int_0^{z_1} \frac{\exp(2\alpha z')}{h(z')} dz' + \beta_L T_{11}^2 \left(\frac{c}{c_L} \right)^3 \cdot \exp(2\alpha z_1) \int_0^{z-z_1} \frac{\exp(2\alpha_L z')}{h(z_1 + c_L z'/c)} dz' \right], \quad (15)$$

where $z_a = c_L z/c + (1 - c_L/c)z_1$. Regarding the values of q_1 and q_2 in the region after the sample ($z > z_2$), $q_1(r, z_{2+}) = T_{01}q_1(r, z_{2-})$ and $q_2(r, z_{2+}) = T_{02}q_2(r, z_{2-})$ are used for the known values which are obtained from Eqs. (14) and (15) by putting $z = z_2$, respectively. Hence,

$$q_1(r, z) = -\frac{T_{11}T_{01}u_0}{h(z_b)} \exp[-\{\alpha(z - L) + \alpha_L L\}] \cdot \exp \left[j \frac{kr^2}{2z_b} \left\{ 1 + j \frac{k}{h(z_b)} \right\} \right], \quad (16)$$

$$q_2(r, z) = \frac{k^2 u_0^3}{4ch(z_b)} \exp[-4\{\alpha(z - L) + \alpha_L L\}]$$

$$\cdot \exp \left[j \frac{kr^2\{h(z_1) + jk\}}{h(z_b)z_1} \right] \cdot \left[\beta T_{12}T_{02} \int_0^{z_1} \frac{\exp(2\alpha z')}{h(z')} dz' + \beta_L T_{11}^2 T_{02} \left(\frac{c}{c_L} \right)^3 \exp(2\alpha z_1) \cdot \int_0^L \frac{\exp(2\alpha_L z')}{h(z_1 + c_L z'/c)} dz' + \beta T_{11}^2 T_{01}^2 \cdot \exp[2(\alpha z_1 + \alpha_L L)] \cdot \int_0^{z-z_2} \frac{\exp(2\alpha z')}{h(z' + z_1 + c_L L/c)} dz' \right], \quad (17)$$

where $z_b = z + (c_L/c - 1)L$.

3. SOUND FIELD ANALYSIS

The parameters of the source employed for a standard model are the fundamental frequency $\omega/2\pi = 2$ MHz, the focal length $D = 85$ mm, and the Gaussian distribution coefficient $A = 1,250 \text{ m}^{-2}$. Then the absorption coefficient in water $\alpha = 0.1$ neper/m.

The axial distribution of $Q_s = q_{20} \sin \theta / q_{10}$ and $Q_e = q_{20} \cos \theta / q_{10}$ for the case of a lossless liquid sample with thickness $L = 20$ mm, but whose sound speed c_L differs from that of water c , is shown in Fig. 2. The axial values of Q_s and Q_e for the sample with the same thickness, but whose density ρ_L differs from that of water ρ , are also shown in Fig. 3. Here we define as $q_{10} = |q_1|$, $q_{20} = |q_2|$, and the phase parameter $\theta = \angle q_2 - 2\angle q_1$ relates the phases of the fundamental and second harmonic components. The values of Q_s and Q_e in Figs. 2 and 3 are normalized so that the q_{20}/q_{10} at the focal point for the case with no samples is unity. Note that, while the transmission ratios have been derived through the

Fig. 2 Axial values of Q_s and Q_e for the case with a sample of various c_L

Fig. 3 Axial values of Q_s and Q_c for the case with a sample of various ρ_L .

boundary condition of continuous sound pressure, the reflected sound is neglected in these calculations. Then each velocity potential should be discontinuous at the boundary even if $\rho_L = \rho$. However the values Q_s and Q_c , which correspond to the amplitude ratios of the velocity potentials of the second harmonic and fundamental, are almost continuous. While Q_s in the post focal region changes as c_L and ρ_L change, Q_c is almost constant. Furthermore, Q_c nearly vanishes at $z=2D$ ($=170$ mm). This is due to the fact that, while Q_s corresponds to the magnitude of the second harmonic component generated in the inserted sample, Q_c corresponds to the second harmonic component generated in the region outside the sample.

The value of the phase parameter θ at the point $z=2D$ on the axis varies as shown in Fig. 4, where any one of three lossless-sample parameters, c_L/c , ρ_L/ρ and β_L/β , changes within the range of 0.75 to 1.25. The value θ remains at about $\pi/2$ rad in this extent. This means that the second harmonic component generated in lossless samples, whose phase parameter results in $\pi/2$ rad, dominates in the second harmonic sound at $z=2D$, because the second harmonic components generated before and after the sample cancel each other. This behavior is similar to the cancellation performed by the second harmonic sounds generated at the pre- and post-focal regions in the case without samples.⁵⁾

The axial values of Q_s and Q_c , in the case that only the absorption coefficient α_L of the lossy sample with thickness $L=20$ mm changes, are shown in Fig. 5. The values of Q_s and Q_c are normalized so that $q_{20}/q_{10}=1$ at the focus when $\alpha_L/\alpha=1$. The changes of Q_s and Q_c at the axial point $z=2D$ according to the change of α_L are also

Fig. 4 Axial phase parameter θ at $z=2D$ for the case with a sample of various c_L , ρ_L and β_L .

Fig. 5 Axial values of Q_s and Q_c for the case with a sample of various α_L .

Fig. 6 Dependence of axial values Q_s and Q_c at $z=2D$ on absorption coefficient of sample α_L .

shown in Fig. 6. Since the absorption coefficient is supposed to be proportional to the square of the frequency, the second harmonic sound is more attenuated. Consequently the amplitude ratio q_{20}/q_{10} and Q_s decrease as α_L increases. On the other hand, the second harmonic component generated before the sample, which needs longer propagation to reach the point $z=2D$ than that generated after the sample does, is more affected by α_L . Then the large absorption weakens the cancellation effect of the second harmonic components. Therefore Q_c increases as α_L increases.

Now we suppose that the sample is located in front of a plane piston source which is radiating an idealized plane wave instead of a focused sound. Let the position of the piston surface be $z=z_{1-}$, that is just before the sample. Assuming that $k \gg \alpha$ and $k_L \gg \alpha_L$ for simplicity, the transmission ratios are approximated as

$$T_{11} \simeq T_{12} = \frac{2\rho c_L}{\rho c + \rho_L c_L} (=T_1),$$

$$T_{01} \simeq T_{02} = \frac{2\rho_L c}{\rho c + \rho_L c_L} (=T_0).$$

The fundamental component at $z=z_{2+}$ (right after the sample) obtained by solving the equation derived by neglecting the r -dependent term of Eq. (5) is

$$q_1(z_{2+}) = T_1 T_0 q_1(z_{1-}) \exp(-\alpha_L L),$$

and the fundamental component at the same point under the condition without sample is

$$q_{1W}(z_{2+}) = q_1(z_{1-}) \exp(-\alpha L).$$

Therefore the change of the fundamental component due to the insertion of the sample is expressed by

$$R_f = \frac{q_1(z_{2+})}{q_{1W}(z_{2+})} = T_1 T_0 \exp[-(\alpha_L - \alpha)L]. \quad (18)$$

On the other hand, the second harmonic component right after the sample is given by the solution of Eq. (8) neglecting the r -dependent term, which is

$$q_2(z_{2+}) = \frac{\beta_L k_L^2 T_1^2 T_0 q_1^2(z_{1-})}{8c_L} \cdot \frac{1 - \exp(-2\alpha_L L)}{\alpha_L} \exp(-2\alpha_L L)$$

$$\simeq \frac{\beta_L k_L^2 T_1^2 T_0 q_1^2(z_{1-})}{4c_L} L \exp(-3\alpha_L L).$$

($\alpha_L L \ll 1$)

For the case without sample,

$$q_{2W}(z_{2+}) \simeq \frac{\beta k^2 q_1^2(z_{1-})}{4c} L \exp(-3\alpha L).$$

Hence the ratio R_s becomes

$$R_s = \frac{q_2(z_{2+})}{q_{2W}(z_{2+})} = T_1^2 T_0 \left(\frac{\beta_L}{\beta} \right) \left(\frac{c}{c_L} \right)^3 \cdot \exp[-3(\alpha_L - \alpha)L]. \quad (19)$$

In order to eliminate the absorption coefficient α_L in Eqs. (18) and (19), the following quantity C_{s0} is introduced.

$$C_{s0} = \frac{R_s}{R_f^3} = \frac{1}{T_1 T_0^2} \left(\frac{\beta_L}{\beta} \right) \left(\frac{c}{c_L} \right)^3. \quad (20)$$

Returning to the case of a focusing source, since $q_{20} \sin \theta$ at the post focal region is proportional to the magnitude of the second harmonic component generated in the sample, the change of $q_{20} \sin \theta$ at $z=2D$ for instance is supposed to have a physical meaning similar to the aforementioned R_s . The change of q_{10} at $z=2D$ corresponds to R_f because of a linear propagation of the fundamental after the sample. Then the change of the value, $q_{20} \sin \theta / q_{10}^3 (=Q_s/q_{10}^2)$ at $z=2D$ is similar to C_{s0} . Consequently the value of Q_s/q_{10}^2 is expected to be substantially independent of α_L . The value of Q_s/q_{10}^2 at $z=2D$ is named C_s . An example of C_s obtained by using Eqs. (16) and (17) for the case that $L=20$ mm and $\alpha_L/\alpha=1$ is shown by the solid curves in Fig. 7, where each of the three parameters c_L , ρ_L and β_L , is varied from the corresponding value of water. The dot curves in the figure indicate the values C_{s0} for the case of plane piston. They agree well each other. Therefore C_s changes in the same manner as that of C_{s0} for plane waves according to the variation of acoustic properties of the sample.

Fig. 7 Values of C_s (—) and C_{s0} (···) for samples with various c_L , ρ_L and β_L .

Fig. 8 Dependence of value C_s (—) on thickness L and absorption coefficient α_L/α of sample, and the relation to value of C_{s0} (---).

This can happen because the second harmonic component observed at $z=2D$ reflects the sample characteristics.

The dependence of C_s on L and α_L/α obtained by using Eqs. (16), (17) and (20) is shown in Fig. 8, where each of the three parameters, c_L , ρ_L and β_L , is set to be 0.75 or 1.25 times the respective value of water. Since the multiple reflection in the sample is ignored in the present formulation, the C_s in Fig. 8 is not necessarily unity when $L=0$. This contradiction is not serious so far as a sample of relatively large L is concerned. The C_s changes with increasing L until C_s tends to saturate near the value of C_{s0} , represented by dot straight lines in the figure. For the L larger than the focal length, the sample sticks out of the focal region, and C_s almost remains constant. The focal region length $\Delta z (=10AD^2/k)$ is 11 mm for the present standard model. Since the value C_s is weakly dependent on the absorption coefficient α_L , C_s is the quantity having the property similar to C_{s0} of Eq. (20). The value of C_s is influenced by c_L rather than ρ_L and β_L as seen from Eqs. (17) and (20).

4. EXPERIMENT

The block diagram of the experimental apparatus is shown in Fig. 9. The arrangement and the experimental procedure are almost same as those in Ref. 5). The transducer, whose resonance frequency and radius a are 2 MHz and 40 mm respectively, radiated cw pulses of 20 μ s in duration. A focused beam was formed in degassed fresh water by use of an acoustic lens with radius of 40 mm and focal

Fig. 9 Block diagram of measurement system.

Table 1 Acoustic properties of samples.

Liquid sample	c_L (m/s)	ρ_L (kg/m ³)	α_L (/m)	β_L
Benzyl alcohol	1,540	1,045	0.38	6.1
<i>n</i> -Butyl alcohol	1,628	810	0.34	6.35
Methyl alcohol	1,121	791	0.13	5.8
Glycerine	1,923	1,261	10.8	5.4

length of 170 mm attached to the transducer. The thickness of the focal region became approximately 40 mm.⁴⁾ The temperature was kept constant at 20°C. A miniature PVDF hydrophone (product of Medicoteknisk Institut, Denmark) with diameter of 1 mm was set 170 mm forward from the focal point, and 5 mm off the axis. The received waveform was observed on an oscilloscope.

Table 1 shows the sound speed c_L , the density ρ_L , the absorption coefficient α_L and the nonlinearity parameter β_L of experimental liquid samples at 20°C found in Refs. 11) and 12). The sample thickness was set to be 30 mm. The experimental source amplitude was adjusted to avoid shock formations even in the case of methyl alcohol sample, where the greatest distortion was expected because of the lowest sound speed. Then the fundamental amplitude at $z=2D$ was 206.8 dB *re* 1 μ Pa. This condition led to the pressure amplitude

Table 2 Measured and calculated values of C_s .

Liquid sample	Meas.	Cal.	C_{s0}
Benzyl alcohol	1.52	1.44	1.56
<i>n</i> -Butyl alcohol	3.41	2.90	3.14
Methyl alcohol	4.13	3.90	4.14
Glycerine	1.03	0.82	0.78

ratio of the second harmonic and fundamental components at $z=2D$ to be 0.33 and 0.086 respectively in the experiment with and without the methyl alcohol sample. Thus the quasi-linear analysis discussed above is appropriate.

Two sets of the values q_{10} , q_{20} and θ were obtained through the Fourier transform of the pressure waveforms observed under the conditions with and without a sample, then C_s was evaluated. The result is shown in Table 2. The table also shows the calculated values of C_s and C_{s0} . The experimental sound source was not a Gaussian source with an infinite radius. In this case, using the boundary condition

$$q_1(r, 0) = \frac{u(r)}{jk} \exp \left[-j \frac{kr^2}{2D} \right], \quad (0 \leq r \leq a)$$

$$= 0 \quad (a < r)$$

instead of Eq. (10), the fundamental component in the post focal region ($z > z_2$) becomes

$$q_1(r, z) = -\frac{T_1 T_0}{z_b} \exp[-\{\alpha(z-L) + \alpha_L L\}]$$

$$\cdot \exp \left[j \frac{kr^2}{2z_b} \right] \int_0^a u(x) dx$$

$$\cdot \exp \left[j \frac{kx^2}{2} \left(\frac{1}{z_b} - \frac{1}{D} \right) \right] J_0 \left(\frac{krx}{z_b} \right) x dx. \quad (21)$$

The second harmonic component in this region is given as follows:

$$q_2(r, z) = \frac{T_1 T_0 k^2}{4\pi c z_b} \exp[-4\{\alpha(z-L) + \alpha_L L\}]$$

$$\cdot \exp \left[j \frac{kr^2}{z_b} \right] \int_{\xi=0}^{\pi} \int_{y=0}^{\alpha} \int_{x=0}^a$$

$$\cdot \left[\beta \int_0^{z_1} \frac{\exp(2\alpha z')}{z'} \exp \left(j \frac{G}{z'} \right) dz' \right.$$

$$\left. + T_1 \beta_L \left(\frac{c}{c_L} \right)^4 \exp(2\alpha z_1) \int_0^L \frac{\exp(2\alpha_1 z')}{z' + c z_1 / c_L} \right]$$

$$\cdot \exp \left(j \frac{G}{z' + c z_1 / c_L} \right) dz' + T_1 T_0 \beta$$

$$\cdot \exp [2(\alpha z_1 + \alpha_L L)] \int_0^{z-z_2} \frac{\exp(2\alpha z')}{z' + z_1 + c L / c_L} dz'$$

$$\cdot \exp \left(j \frac{G}{z' + z_1 + c L / c_L} \right) dz' \Big] u(x) u(y)$$

$$\cdot \exp \left[-\frac{k}{2D} (x^2 + y^2) \right] \exp \left(j \frac{kF}{4z_b} \right)$$

$$\cdot J_0 \left(\frac{kr\sqrt{F}}{z_b} \right) xy dx dy d\xi \quad (22)$$

where $G = k(x^2 + y^2 + 2xy \cos \xi) / 4z$ and $F = x^2 + y^2 - 2xy \cos \xi$. The $u(r)$ is the source velocity amplitude, for which the experimentally obtained approximate expression $u(r) = u_0 \exp(-720r^2)$ is employed here. The values of C_s in Table 2 were computed by using Eqs. (21) and (22). The measured values of C_s do not agree well with the calculated values because of the complicated sound field due to source edge diffractions, and a rough approximation of $u(r)$ for the computation. However the measured values show the tendency similar to those of the calculated values of C_s and C_{s0} . Therefore the values of C_s may express the influence of the sample properties excluding the linear absorption on the nonlinearly generated second harmonic component. A better agreement would be attained by a completely furnished Gaussian source. Since an ideal Gaussian source makes the value C_{s0} closely approximate the value C_s , the influence of the sample value of c_L , ρ_L and β_L on the second harmonic generation would be simply described by C_{s0} . Then, when c_L and ρ_L are known, the relative value of the nonlinearity parameter of the sample β_L / β may be evaluated.

5. CONCLUSION

The second harmonic component nonlinearly generated in a focused sound field with the insertion of a liquid sample, whose linear and nonlinear properties are different from those of surrounding medium (water), was examined by numerical computations and experiments.

The analysis for the focusing source with a Gaussian profile excitation shows that, in the case of a lossless sample, the second harmonic component generated in the inserted sample accounts for the greater part of the second harmonic sound at $z=2D$. This is due to the fact that the second harmonic component newly generated after the

sample cancels the second harmonic component propagating through the sample. In the case of a lossy sample, since the second harmonic component generated in front of the sample is affected by the absorption in the sample more than the component generated after the sample is, the cancellation of second harmonic sound is weakened. However the value of C_s , that is the ratio of the values $q_{20} \sin \theta / q_{10}^3$ for the cases with and without a sample, is independent of the absorption coefficient of the sample, and is determined only by the sound speed, density and nonlinearity parameter.

The experiment carried out in the focused beam in water shows that the value C_s tends to change according to the variation of samples as predicted in the theory.

The cancellation of the second harmonic sound discussed in the present article is peculiar to spherically focused sounds. This cannot be expected in cylindrically focused sounds.¹⁸⁾

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