

Transmission line method for the measurement of the acoustic nonlinearity parameter in biological liquids at very high frequencies

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Nonlinear wave propagation in a system consisting of a liquid specimen held between two SiO₂ buffer rods is studied as a new method of determining the acoustic nonlinearity parameter of liquids at very high frequencies. Since the sign of the nonlinearity parameter of SiO₂ is opposite that for liquids, the system can be viewed as a transmission line containing regions of both positive and negative nonlinearity. The nonlinear characterization curve is employed in which the second harmonic component output of the receiving transducer is plotted as a function of propagation distance in liquids between the two SiO₂ buffer rods. The nonlinearity parameter is determined by measuring the dip position at which the transducer output becomes zero in the curve. Measurements are reported for water and aqueous solutions of dextrose and dextran in the fundamental frequency range 100–200 MHz. © 1997 Acoustical Society of America. [S0001-4966(97)05811-6]

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INTRODUCTION

For studies of ultrasonic tissue characterization, a bioultrasonic spectroscopy system and method operating in the VHF and UHF ranges has been developed.¹ Bulk linear acoustic properties of velocity, attenuation coefficient, impedance, and density can be determined simultaneously using an ultrasonic transmission line in which the specimen is sandwiched between two synthetic silica glass (SiO₂) buffer rods having ZnO film ultrasonic transducers on their outer ends, and with distilled water employed as the reference medium. Measurements of some liquids and biological media such as the bovine tissues of liver, heart muscle, and fat have been reported in the frequency range from 10 to 500 MHz, revealing the significantly different properties from those measured at the low megahertz diagnostic medical frequencies.^{1,2} We have taken a strong interest in nonlinear propagation phenomena in liquids and liquidlike media such as tissues at these high frequencies, as the acoustic nonlinearity parameter B/A has been extensively investigated for tissue characterization at low megahertz medical frequencies,^{3–11} and have desired to accumulate basic data of biological media with the nonlinear acoustic parameter as well as with the linear acoustic parameters in this frequency range, for a new research area of tissue characterization.

The broadband operation of ZnO film ultrasonic transducers fabricated on SiO₂ buffer rods, using radio frequency (rf) tone burst pulses, is taken as the great advantage for measurements of frequency characteristics of the linear acoustic parameters. In order to apply the transmission line methodology to measure the acoustic nonlinearity parameter of liquids and liquidlike media, such as tissues, using the finite amplitude method, some problems result from the non-

linear generation in the two SiO₂ buffer rods, for which the nonlinearity parameter has a negative value, in contradistinction to the nonlinearity parameters of liquids/tissues, and most of other solid materials, reported as positive values.^{12–14} In this paper, nonlinear wave propagation in the SiO₂/liquid–specimen/SiO₂ system, in the fundamental frequency range 100–200 MHz, is investigated theoretically and experimentally for an input fundamental-frequency ultrasonic power of about 10 mW. A new method of measuring the nonlinearity parameter of liquids is proposed, taking notice that the sign of the nonlinearity parameter of SiO₂ is different from that of liquids. Measurements are reported for water and 25% weight-fractional aqueous solutions of dextrose and dextran.

I. NONLINEAR PROPAGATION

A. Transducer output

A schematic view of the ultrasonic wave propagation is shown in Fig. 1 for the SiO₂/liquid–specimen/SiO₂ ultrasonic transmission line, where a liquid specimen is inserted between two SiO₂ buffer rods with ultrasonic transducers attached on their outer ends. As all materials are inherently nonlinear, generation of harmonics not only in the liquid but also in the SiO₂ buffer rods must be considered. In our analysis, only the second harmonic component is taken into account and all higher harmonics are ignored. The transmitting transducer is excited by electrical rf pulses at the fundamental frequency f_1 and emits ultrasonic rf pulses into the SiO₂ buffer rod. The receiving transducer detects ultrasonic rf pulses having the fundamental (f_1) and the second harmonic ($f_2 = 2f_1$) components.

In order to describe nonlinear wave propagation in this system, we consider the particle velocities for the fundamental and second harmonic components. Assuming finite, but moderate amplitude wave propagation within the discontinu-

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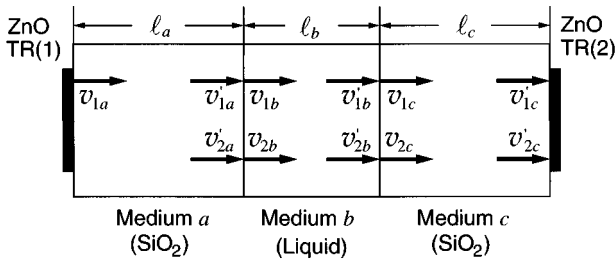


FIG. 1. A model of wave propagation in the SiO₂/liquid-specimen/SiO₂ ultrasonic transmission line, where v_1 and v_2 represents the particle velocities for the fundamental and second harmonic components.

ity distance, the second harmonic particle velocity $v_2(z)$ at the distance z from a fundamental sound source with the particle velocity $v_1(0)$ can be, in general, given by^{3,15,16}

$$v_2(z) = \frac{\beta \pi f_1}{2c_0^2} v_1^2(0) \times \frac{\exp(-\alpha_2 z) - \exp(-2\alpha_1 z)}{2\alpha_1 - \alpha_2} \text{DIFF}(z), \quad (1)$$

where β is the nonlinearity parameter in the medium, c_0 is the infinitesimal wave velocity, α_1 and α_2 are the attenuation coefficients at f_1 and f_2 , respectively, and $\text{DIFF}(z)$ is the diffraction factor. The nonlinearity parameters for liquids, β_L , and isotropic solids, β_S , are defined by¹⁴

$$\beta_L = 2 + \frac{B}{A}, \quad (2)$$

and

$$\beta_S = -\frac{3C_{11} + C_{111}}{C_{11}}, \quad (3)$$

where B/A is the ratio of the coefficient of the second degree term to that of the first degree term of the Taylor's series expansion representation of the equation of state relating the pressure and density,¹⁷ and C_{11} and C_{111} are the second- and third-order elastic constants, respectively. For simplicity, to derive the particle velocities detected by the receiving transducer, we neglect the diffraction effect as $\text{DIFF}(z) = 1$. In Fig. 1, each medium $i (= a, b, c)$ containing ultrasonic propagation loss is featured with the complex acoustic impedance Z_i and the medium by length l_i . The transmission coefficient T_{ij} for the particle velocity from medium i to medium j , where i and $j = a, b, c$ is defined as

$$T_{ij} = \frac{2Z_i}{Z_i + Z_j}. \quad (4)$$

The particle velocity of the fundamental component, v'_{1c} , received by the transducer following transmission through the three media is simply obtained as

$$v'_{1c} = v_{1a} T_{ab} T_{bc} \exp[-(\alpha_{1a} l_a + \alpha_{1b} l_b + \alpha_{1c} l_c)], \quad (5)$$

where v_{1a} is the arbitrary amplitude of the particle velocity at the transmitting transducer. The particle velocity of the second harmonic component at the receiving transducer can be obtained as

$$v'_{2c} = T_{ab} T_{bc} K_a v_{1a}^2 \text{ATT}_a \exp[-(\alpha_{2b} l_b + \alpha_{2c} l_c)] + T_{bc} K_b [T_{ab} v_{1a} \exp(-\alpha_{1a} l_a)]^2 \text{ATT}_b \times \exp(-\alpha_{2c} l_c) + K_c \{T_{ab} T_{bc} v_{1a} \times \exp[-(\alpha_{1a} l_a + \alpha_{1b} l_b)]\}^2 \text{ATT}_c, \quad (6)$$

where $K_i = \beta_i \pi f_1 / c_{0i}^2$ and ATT_i is defined by

$$\text{ATT}_i = \frac{\exp(-\alpha_{2i} l_i) - \exp(-2\alpha_{1i} l_i)}{2\alpha_{1i} - \alpha_{2i}}, \quad (7)$$

and $i = a, b, c$.

The fundamental frequency component, Eq. (5), is the same representation as that for the infinitesimal amplitude wave propagation. In Eq. (6), describing the particle velocity of the second harmonic v'_{2c} , the first term corresponds to the propagation of the second harmonic component generated in medium a , and the second term that in medium b , and the third term that in medium c , respectively.

B. Numerical calculation

Using Eq. (6), we made numerical calculations of the relative transducer outputs at the three fundamental frequencies 120, 150, and 180 MHz, for the second harmonic components in order to know the behavior of the SiO₂/water-specimen/SiO₂ system. The linear and nonlinear acoustic parameters of SiO₂ and water used in the calculations are shown in Table I. The length of the SiO₂ buffer rods is taken as 8 mm.

The results of the calculations are shown in Fig. 2. The relative transducer outputs vary with the length of the liquid medium l_b (propagation distance) between the two SiO₂ buffer rods and reach zero values at 577, 559, and 543 μm for the second harmonic frequencies of 240, 300, and 360 MHz, respectively. The dips result from the different signs of the nonlinearity parameters in SiO₂ and water (see Table I). It can be interpreted, therefore, that, before the dip position, the second harmonic components generated in the two SiO₂ buffer rods make greater contributions to the second harmonic component transducer output, whereas, beyond the dip position, those in the water with 180° phase difference in

TABLE I. Linear and nonlinear acoustic parameters used in the numerical calculations.

Sample	Velocity [m/s]	Attenuation α/f^2 [$\times 10^{-15} \text{ s}^2/\text{m}$]	Density [$\times 10^3 \text{ kg/m}^3$]	β	Temperature [°C]
water	1496.7 (Ref. 18)	21.90 (Ref. 19)	0.9971 (Ref. 20)	7.11 (Ref. 21)	25
SiO ₂	5954 (Ref. 22)	0.15 (Ref. 22)	2.202 (Ref. 22)	-11.58 (Refs. 12-14) ^a	23

^aTemperature is 27 °C.

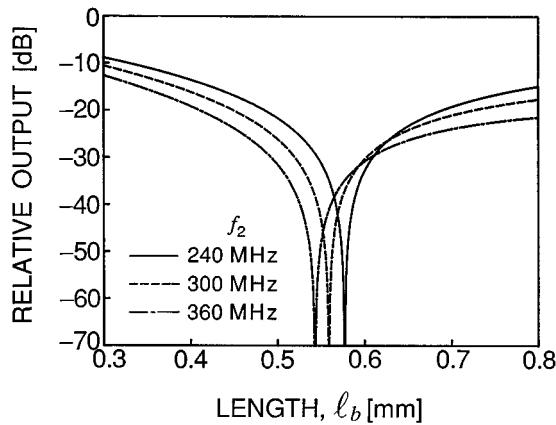


FIG. 2. Calculated receiving transducer outputs of the second harmonic components versus propagation distance in the SiO₂/water/SiO₂ system. The dip positions are 577, 559, and 543 μm for the second harmonic frequencies of 240, 300, and 360 MHz, respectively.

the nonlinearity parameter contribute more. And, as the frequency increases, the dip position appears at lesser values of liquid length. This is mainly due to the frequency dependence of attenuation of water. In this SiO₂/water/SiO₂ system, the ultrasonic waves are mostly attenuated by the water.

$$\beta_L = - \frac{c_{0b}^2 ATT_a \{ \exp[-(\alpha_{2a}l_a + \alpha_{2b}z_d)] + T_{ab}(2 - T_{ab}) \exp[-2(\alpha_{1a}l_a + \alpha_{1b}z_d)] \}}{c_{0a}^2 T_{ab} ATT_b \exp[-(2\alpha_{1a}l_a + \alpha_{2a}l_a)]} \beta(\text{SiO}_2). \quad (9)$$

From these equations, we propose a new method of determining the nonlinearity parameter of the liquid in the space between the buffer rods by measuring the dip position z_d , with the premeasured/known linear acoustic parameters of the velocity, attenuation, and impedance, as well as with the known linear and nonlinear parameters of SiO₂ employed as the reference medium.

II. EXPERIMENTS

A. Measurement system

A block diagram of the measurement system is shown in Fig. 3. Measurements are carried out in rf pulse mode operation, and a signal generator and two bandpass filters, necessary for the nonlinear experiments, are introduced additionally to the pulse mode measurement system used in the bioultrasonic spectroscopy system.¹ This system consists of six main parts: the signal generator of electrical rf tone burst pulses, a pair of ultrasonic plane wave devices including the liquid specimen, the mechanical system for alignment of the two ultrasonic devices and for translation to record the nonlinear characterization curve, the rf receiver, the temperature measurement system, and the computer to control the entire system and to process the measured data.

An rf pulse is passed through the bandpass filter tuned to the fundamental frequency used in experiments, in order to

Thus as the second harmonic component transducer output exhibits a characteristic curve featured with a dip at a particular propagation distance, the location of the dip in the nonlinear characterization curve can be employed for measurement of the nonlinearity parameter.

C. Determination of the nonlinearity parameter

As the media a and c are the same materials, SiO₂ with the negative nonlinearity parameter $\beta(\text{SiO}_2)$, and the medium b is the liquid specimen in which the propagation distance is alterable; in Eq. (6) the second harmonic component transducer outputs for the two SiO₂ buffer rods and liquid are canceled out at a certain propagation distance in the liquid $l_b = z_d$, so that v_{2c}' becomes zero. Then, the nonlinearity parameter of the liquid specimen, β_L , is expressed as

$$\beta_L = - \frac{c_{0b}^2 ATT_a \exp[-(\alpha_{2b}z_d + \alpha_{2c}l_c)]}{c_{0a}^2 T_{ab} ATT_b \exp[-(2\alpha_{1a}l_a + \alpha_{2c}l_c)]} \beta(\text{SiO}_2) - \frac{c_{0b}^2 T_{ab} T_{bc} ATT_c \exp[-2(\alpha_{1a}l_a + \alpha_{1b}z_d)]}{c_{0c}^2 T_{ab} ATT_b \exp[-(2\alpha_{1a}l_a + \alpha_{2c}l_c)]} \beta(\text{SiO}_2). \quad (8)$$

Furthermore, when the length of two SiO₂ buffer rods are equal, i.e., $l_a = l_c$, the above equation can be rewritten as

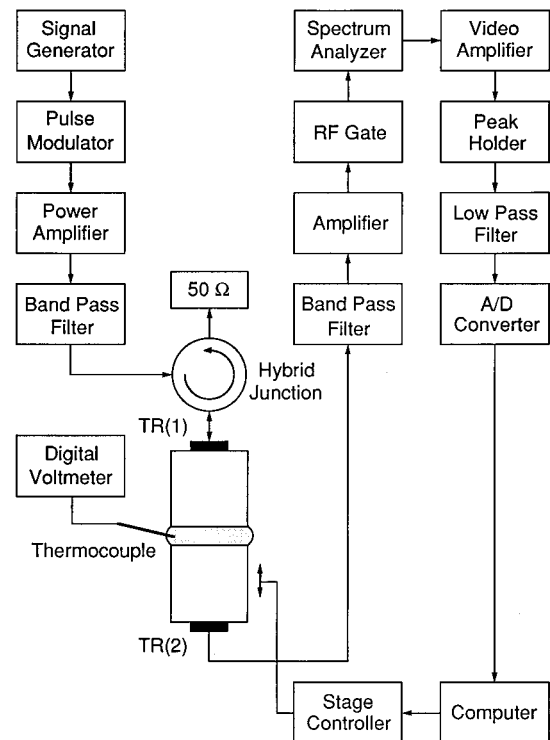


FIG. 3. Block diagram of the measurement system.

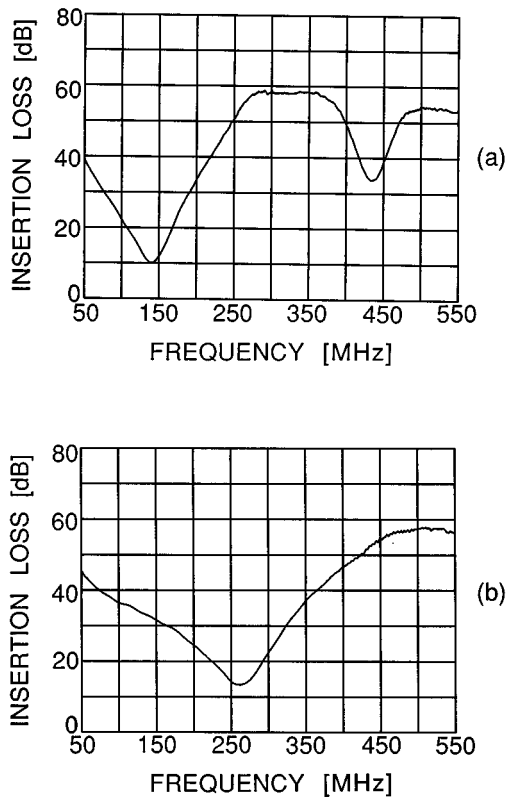


FIG. 4. Frequency characteristics of the insertion loss of the transmitting and receiving ultrasonic devices. (a) Transmitter; (b) receiver.

eliminate sufficiently its harmonics. The ultrasonic rf pulse wave, excited by the transmitting transducer TR(1), propagates through the two SiO₂ buffer rods and the liquid specimen, and the harmonics are generated in both media. The receiving transducer TR(2) detects a train of distorted ultrasonic rf pulse waves and converts them to electrical signals. The second bandpass filter selects the frequency components to be observed. The signals are amplified and the desired rf pulse signals are extracted by the rf gate circuit, and then detected with the spectrum analyzer. The A/D converted signals for the fundamental and second harmonic components are recorded as a function of the distance z by changing the spacing length between the two SiO₂ buffer rods by a z -axis translation stage with a stepping motor with a resolution of 0.1 μm . The temperature of the liquid specimen is measured by an inserted copper–constantan thermocouple.

The transducers used are of ZnO piezoelectric films fabricated by dc sputtering on the cylindrical SiO₂ (T-4040, Toshiba Ceramics Co.) rods 8 mm long and with diameters of 8 mm for the transmitter and 20 mm for the receiver, in order to support the liquid specimen in the gap between the two rods. The transmitting and receiving transducers were designed to have the film thicknesses of 19 μm around the center frequency of 150 MHz and of 9.5 μm around 300 MHz, respectively, having the same transducer diameter of 2.6 mm. Figure 4 shows the frequency characteristics of the insertion loss of the two devices. This transmitter can be used to excite ultrasonic waves at fundamental frequencies from 100 to 200 MHz, and the receiver can detect the corresponding fundamental and second harmonic components.

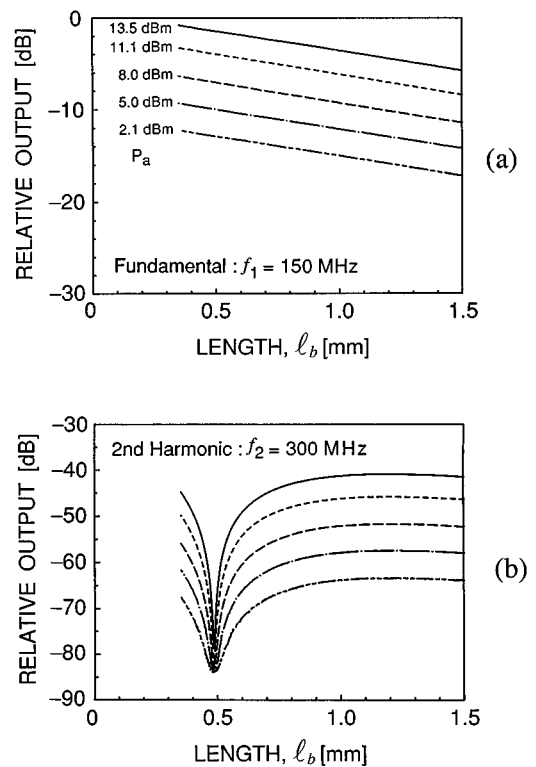


FIG. 5. Receiving transducer outputs for water as the liquid between the buffer rods versus propagation distance as a function of the acoustic power at the transmitting transducer. The dip position is 482 μm .

B. Results and discussions

1. Water

To confirm the nonlinear wave propagation measurement method described above, experiments were first conducted at 150 MHz for water for which the linear and nonlinear acoustic properties are well known.^{18–21} Figure 5 shows the results of the receiving transducer output obtained as a function of acoustic power P_a at the transmitter. Measurements using the pulse width of 300 ns were carried out with the propagation distance larger than 0.3 mm. Figure 5(a) shows the results for the fundamental frequency and Fig. 5(b) for the second harmonic frequency. Both the transducer outputs at 150 and 300 MHz increase in proportion to the ultrasonic power used in experiments. The transducer output at 150 MHz decreases monotonically with the length of propagation distance in the liquid. The transducer output at 300 MHz becomes zero at the same propagation distance, as predicted in the calculated curves of Fig. 2. The dip distance was measured to be 482 μm .

Next, similar experiments were carried out for two other fundamental frequencies, 120 and 180 MHz, with the ultrasonic power of 8 dBm. Figure 6 shows the results, together with those at 150 MHz, reflecting the frequency dependence of attenuation. The dips were observed clearly at the different propagation distances, viz., at 496, 482, and 464 μm for the fundamental frequencies of 120, 150, and 180 MHz, respectively, as theoretically predicted above.

From the dip positions obtained above, the nonlinear parameters for water were calculated, using Eq. (9) with the

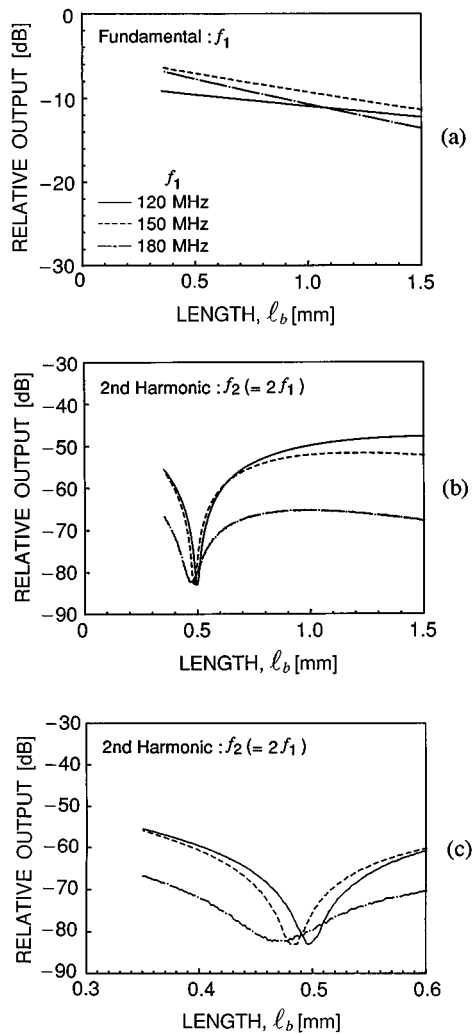


FIG. 6. Receiving transducer outputs for water versus propagation distance as a function of the fundamental frequency. The dip positions are 496, 482, and 464 μm for the second harmonic frequencies of 240, 300, and 360 MHz, respectively.

relevant acoustic parameters of water and SiO_2 given in Table I, and compared with published data measured at 9.9 MHz,²¹ as shown in Table II. Slightly different β_W values of 8.34–8.44 were obtained, depending on the frequencies employed, but approximately 20% larger than the published data.

2. Aqueous solutions of dextrose and dextran

Twenty-five percent weight-fractional aqueous solutions of dextrose and dextran of different molecular weights were

TABLE II. Determined nonlinearity parameter for water.

f_1	β_W
120 MHz	8.36
150 MHz	8.34
180 MHz	8.44
9.9 MHz	7.11 (Ref. 21) ^a

^a $B/A = 5.11$ at 25 °C.

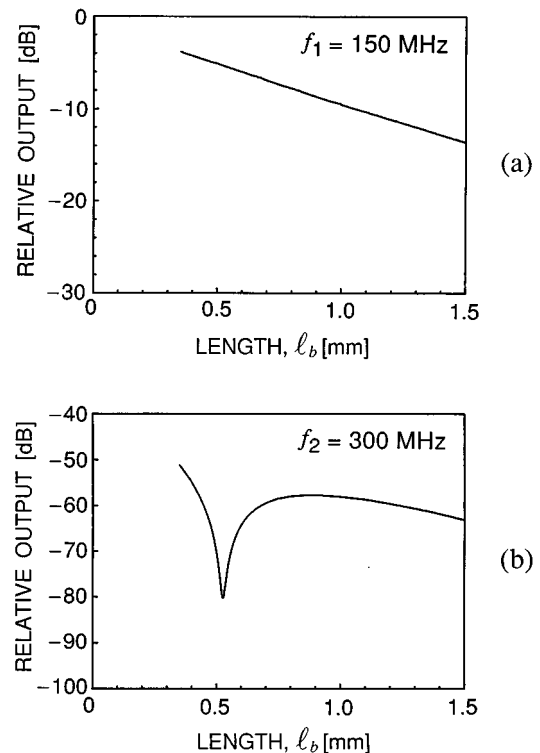


FIG. 7. Receiving transducer outputs for 25% aqueous dextrose solution versus propagation distance. The dip position is 526 μm .

studied at the fundamental frequency of 150 MHz. The molecular weights were 180 Daltons for the dextrose and $(2-3) \times 10^5$ Daltons for the dextran solutes. The measured results of the transducer outputs are shown in Fig. 7 for 25% aqueous dextrose solution and in Fig. 8 for 25% aqueous dextran solution. It is seen from these data that the 25% dextrose aqueous solution has the linear acoustic properties of lesser attenuation, and the slightly larger dip position in the nonlinear characterization curve. To determine their nonlinear parameters, we also measured the linear acoustic parameters of velocity, attenuation, and impedance,¹ and calculated the nonlinearity parameters, comparing them with the published data,⁵ as shown in Table III. The nonlinearity parameter of 9.18 for the 25% aqueous dextrose solution is smaller than the 9.56 value for the 25% aqueous dextran solution. Both values are approximately 20% larger than the published data.

The relative nonlinearity parameter, $\beta_R (= \beta_L / \beta_W)$, obtained by dividing the measured values by the nonlinearity parameter value of water, β_W , actually obtained in the same system at the same frequency, might lead to more reasonable results based on the two following reasons: (1) water is one of the better known and most physically and chemically stable media and one of the most important constituents of biological media; and (2) measurement errors due to the total diffraction effects in this system might be experimentally minimized because the velocities of the liquids are close to the velocity of water, as previously introduced in the bioultrasonic spectroscopy system.¹ For the 25% aqueous solutions of dextrose and dextran, we obtain 1.10 and 1.15, respectively, as given in Table III.

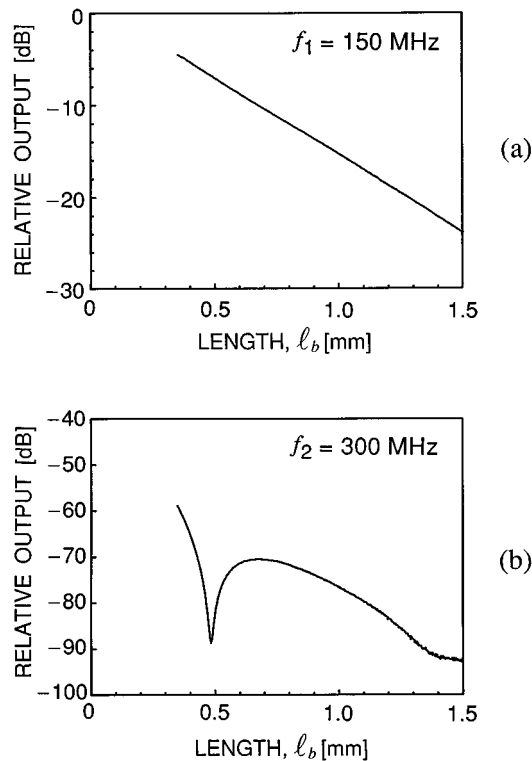


FIG. 8. Receiving transducer outputs for 25% aqueous dextran solution versus propagation distance. The dip position is 487 μm .

From these experiments, we could find out the first new information that the nonlinearity parameter is dependent on the molecular weight when these very high frequencies are utilized, although it was reported that the nonlinearity parameter in the dextrose and dextran aqueous solutions is insensitive to molecular weight and nearly the same for the same concentration, obtained by the finite amplitude and thermodynamic methods.⁵

C. Measurement accuracy

Accuracy of the nonlinearity parameters measured for the 25% aqueous solutions of dextrose and dextran with this prototype system, carried out in an air-conditioned room but wherein the temperature of specimen is measured independently, is discussed using Eq. (9), for which the diffraction effect is ignored. The measurement accuracy depends on the measurement of the dip position z_d in the nonlinear characterization curve, the measurement of the linear acoustic parameters of velocity, attenuation, and impedance for the aqueous solutions, the values of the linear and nonlinear acoustic parameters for the SiO_2 buffer rods as the reference

medium, and the measurement of the length of the buffer rods. We now assume sufficient accuracy for the necessary values for the SiO_2 rods. The length of buffer rods can be easily measured with better than $\pm 0.002\%$ by a length gauging system having a resolution of $\pm 0.1 \mu\text{m}$. The reproducibility of about $\pm 0.5 \mu\text{m}$ in dip measurement corresponds to the error of $\pm 0.1\%$ in β . On the other hand, the measurement accuracies of the linear acoustic parameters for the aqueous solutions are estimated to be $\pm 0.05\%$ for velocity, $\pm 1\%$ for attenuation, and $\pm 1\%$ for impedance in this study, which contribute to the errors in β of $\pm 0.1\%$, $\pm 0.02\%$, and $\pm 0.6\%$, respectively. It can be understood that the measurement error of the acoustic impedance has the greatest influence on the β determination. From these considerations, the measurement accuracy in the present measurements, neglecting diffraction effects, is estimated to be around $\pm 0.8\%$.

The significant differences between the previously published values of β and those measured in this study for water and the aqueous solutions might be attributed to neglect of the diffraction effects in the mathematical treatment. This suggests that further theoretical and experimental studies on the diffraction correction are very important. As the relative measurement error is now estimated to be $\pm 0.8\%$ in β , the difference of 4% obtained between the two 25% aqueous solutions of dextrose and dextran is meaningful and it is considered to result from the different molecular weights.

III. CONCLUDING REMARKS

Nonlinear wave propagation in the system of SiO_2 /liquid-specimen/ SiO_2 has been investigated theoretically and experimentally in the frequency range of the fundamental from 100 to 200 MHz, taking notice that the sign of the nonlinearity parameter of SiO_2 is different from that of liquids. It has been observed that, in the nonlinear characterization curve which records the second harmonic component output of the receiving transducer as a function of propagation distance in the liquid specimen between two SiO_2 buffer rods, there exists a dip at a certain distance z_d where the receiver transducer output becomes zero. A new method of determining the nonlinearity parameter of liquids, by measuring the dip position and the acoustic parameters of velocity, attenuation, and impedance with reference to SiO_2 with well-known linear and nonlinear acoustic parameters, has been developed; measurements have been made with water and aqueous solutions of dextrose and dextran.

Further studies on the effect of diffraction on the determination of the nonlinearity parameters will complete the method, and the following problems will have to be investigated at these very high frequencies: effect of velocity dis-

TABLE III. Determined linear and nonlinear acoustic parameters for aqueous solutions of dextrose and dextran. $\beta_w = 8.34$ at fundamental frequency of 150 MHz.

Sample	Velocity [m/s]	Attenuation $\alpha [\times 10^3 \text{ neper/m}]$		Density [$\times 10^3 \text{ kg/m}^3$]	β_L	β_L/β_w	β_L (Publ.)
		(150 MHz)	(300 MHz)				
25% Dextrose	1623	0.847	3.31	1.06	9.18	1.10	7.96 (Ref. 5)
25% Dextran	1587	1.34	4.18	1.12	9.56	1.15	8.05 (Ref. 5) ^a

^aConcentration is 24%.

persion on measurements, measurement limitation for media with very high ultrasonic losses, and modification of the method suitable for semi-solid biological tissues having a predetermined thickness. This method will be able to yield more accurate measurements than the conventional finite amplitude and thermodynamic methods because absolute wave amplitude measurements are replaced by length measurements.

Bioultrasonic research in the VHF and UHF ranges will be initiated in the near future with the bioultrasonic spectroscopy system with both functions of measuring the linear and nonlinear acoustic properties of biological media.

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