

# Generation and observation of GHz ultrasonic waves on liquid surfaces and a liquid/liquid interface by transient reflecting grating method

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We optically generated and observed GHz ultrasonic waves on liquid surfaces and a liquid/liquid interface for the first time using the transient reflecting grating (TRG) method. Four kinds of samples were used for measurement of the surfaces; two types of black ink, an aqueous solution of crystal violet (CV aq solution) and ethanol aqueous solutions of CV. A cyclohexane/CV aq solution interface was used for measurement of the liquid/liquid interface. The 0.7 GHz and 0.6 GHz ultrasonic waves were generated on the liquid surfaces and the liquid/liquid interface, respectively. These ultrasonic waves resemble longitudinal acoustic waves partly localized at the surface, and they cause surface displacement. The amplitudes of TRG signals from surfaces of the CV mixed solvent solutions were found to be greatly affected by both the viscosity and surface tension. This result indicated the viscosity and interface tension must be considered as perturbations in the elastic theory. The strong dependence on interface tension showed the high selectivity of the TRG signals for the interface. © 1999 American Institute of Physics. [S0021-9606(99)70244-8]

## INTRODUCTION

Liquid surfaces have attracted much attention because they play important roles in basic processes in various fields of science and technology, e.g., evaporation, condensation, energy transfer, mass transfer, and photoionization.<sup>1-3</sup> However, experimental research on liquid surfaces is still in an early stage because it is difficult to measure them.<sup>4</sup> Microscopic clarifications of the structures, roughness, properties such as elasticity, viscosity, and interface tension, and solute-solvent interaction of the liquid surface and the liquid/liquid interface are highly desirable. The static structures of liquid surfaces have been proposed based on results from second harmonic generation and sum frequency generation,<sup>5-7</sup> but the dynamic processes on liquid surfaces are still unknown. As a method to investigate and analyze the collective molecular dynamics on liquid surfaces, the quasi-elastic laser scattering method has been reported.<sup>8-11</sup> It allows the measurement of capillary waves on liquid surfaces and liquid/liquid interfaces with a frequency up to the MHz region. However, it is still insufficient to observe fast phenomena of intra- and intermolecules around liquid surfaces and liquid/liquid interfaces. Although much effort has been made, there still remain many fundamental problems to be clarified.

As for solute molecules in bulk solution, the fast phe-

nomena of bulk molecules such as molecular dynamics of translation, vibration, and rotation, solute-solvent effect, and isomerization have been discussed using many methods. Ultrasonic spectroscopy is one method to investigate the mechanism of the absorption or the relaxation of the ultrasonic waves in a substance using a spectrum of the sound velocity or the absorption coefficient vs the frequency of the ultrasonic waves. For liquid samples, ultrasonic waves with a frequency between the MHz and GHz region are mainly used. The dissociation reaction of micelles and monomers in a surfactant solution and the local rotational dynamics of polymer chains in a polymer solution have been investigated with this method.<sup>12-14</sup> Furthermore, the hydration number of ions in an electrolyte solution has been determined from the adiabatic compressibility obtained by measuring sound velocity in the solution.<sup>15-17</sup> Thus, we focus on the application of this method to liquid surfaces and liquid/liquid interfaces. Therefore, techniques to induce and observe ultrasonic waves on liquid surfaces and liquid/liquid interfaces with the frequency from the MHz to GHz region are highly desirable.

Waves on a liquid surface have been investigated for a long time, beginning more than two hundred years ago from an interest in the waves at sea. Early casual interest gave way to serious studies in the 1960s which used mechanical methods. Mann and Hansen<sup>18</sup> induced and observed waves using a ripple generator and ripple transducer, while Davies and Vose<sup>19</sup> induced waves with an electromagnetic vibrator and observed them stroboscopically. Still in the 1960s Lu-

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cassen first discussed longitudinal waves, and Sohl *et al.*<sup>20</sup> developed an apparatus to investigate longitudinal waves by introducing an electrocapillarity method. This technique was followed by Miyano<sup>21</sup> and the waves were induced by electric fields and observed with a laser beam. Hajiloo<sup>22</sup> generated waves on liquid surfaces by a laser beam and this technique was easily extended to liquid/liquid interfaces as well. However, these techniques were not able to generate surface waves with a frequency between the MHz and GHz region.

To measure ultrasonic waves with such a high frequency on solid surfaces and at solid/liquid interfaces, we and other few groups have developed a transient reflecting grating (TRG) technique.<sup>23–26</sup> Fayer and co-workers<sup>27,28</sup> and Nelson and co-workers<sup>28</sup> presented detailed theoretical analysis of the method. This technique was used to generate and detect GHz surface acoustic waves successfully. Elastic and thermal properties of thin films and multilayer thin films were estimated by this technique.<sup>29–33</sup> Some chemical or physical applications were also made for measurements of ultrafast carrier dynamics.<sup>34–38</sup> This method has advantages of providing noncontact, nondestructive, and interface-selective measurements, and high temporal resolution. Moreover, it can be applied to various types of samples because it utilizes nonradiative relaxation processes.

Therefore, as we have demonstrated on solid surfaces and solid/liquid interfaces, the optical generation and observation of GHz ultrasonic waves will be a powerful tool for an investigation of liquid surfaces and liquid/liquid interfaces.

In this study, we apply the TRG method to the liquid surfaces and the liquid/liquid interface and to investigate the possible use as a technique to generate ultrasonic waves on liquid surfaces and at liquid/liquid interfaces.

## PRINCIPLE

The TRG method uses optical interference fringes on surfaces.<sup>22–33</sup> Two temporally coincident laser pulses (pump pulses) with the same wavelength produce optical interference fringes on and under the sample surface. These optical fringes cause spatial modulations of the refractive index and surface displacement. Increases in photoexcited species and in temperature are the main causes of the refractive index modulation. The modulation of the surface displacement is caused by thermal expansion. All these modulations work as a transient reflecting grating (TRG) which causes a third pulse beam (probe beam) to be diffracted. A TRG is considered as a corrugated grating superimposed on the reflectivity grating. For convenience, a TRG is classified into three types; a population grating formed by the density distribution of photoexcited species, a thermal grating formed by the temperature distribution resulting from nonradiative relaxation and heat generation, and an acoustic grating formed by thermal expansion that generally generates counterpropagating surface acoustic waves for solids. For liquids, the acoustic grating is due to a longitudinal acoustic wave localized at the interface. Both the spacing and direction of the grating are optically controllable. The optical fringe spacing  $\Lambda$  is given by

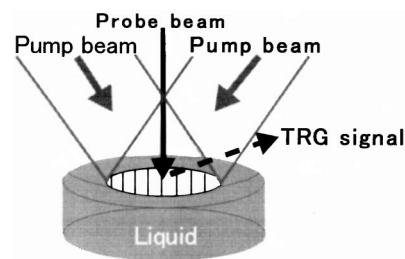


FIG. 1. The optical arrangement to observe the liquid surfaces and the liquid/liquid interface.

$$\Lambda = \lambda/2 \sin \theta, \quad (1)$$

where  $\lambda$  is the optical wavelength and  $2\theta$  is the incident angle of the pump pulses. Thus,  $\Lambda$  is tunable over a range from sub- $\mu\text{m}$  to more than 100  $\mu\text{m}$  with a visible laser source.

## EXPERIMENT

The experimental setup of the TRG system was the same as described previously,<sup>31–33</sup> except for the arrangement to observe liquid surfaces and liquid/liquid interfaces (Fig. 1). A mirror was placed between the lenses and the liquid sample on the stages, and consequently both pump and probe pulses were incident on the liquid surface and the liquid/liquid interface. A mode-locked Q-switched Nd:YAG laser (Quantronix, model 416) was used as a light source. The pulse train wavelength was 532 nm with a repetition rate of 1 kHz and a pulsewidth of 80 ps in full-width at half-maximum. The pulse was separated into pump and probe pulses using a partial reflective mirror. The pump beam was further divided into two beams by a half-mirror. The two pump pulses were crossed and irradiated onto the same spot of the sample surface so as to coincide in time to form an interference pattern. The probe pulse was irradiated at the center of the spot after passing through a computer-controlled optical delay line. The reflecting diffracted light was monitored with a photomultiplier (PMT) connected to a flexible light guide, whose entrance was placed at one of the first order reflecting diffraction spots. The output signal from the PMT was gated and averaged over one millisecond with a boxcar integrator before an analog-to-digital transformation for computer storage. The spot diameters of the pump and probe beams were 60  $\mu\text{m}$  and 40  $\mu\text{m}$ , respectively. The fringe spacing was typically 2.3–2.4  $\mu\text{m}$ . The intensity of the pump and probe pulse was less than 3, 0.1  $\mu\text{J}/\text{pulse}$ , respectively.

We used four liquid samples for measurements of the surfaces: black ink 1, “art pen ink” (Rotring, Art.R598 317); black ink 2, “drawing ink” (Rotring, Art.R591. 017); an aqueous solution of crystal violet (CV; Kanto Chemical Co., Inc.: special grade); and CV ethanol aqueous solutions. Thin layer chromatography showed that the first three samples were a colloidal solution, a mixed-dye aqueous solution, and a single-dye aqueous solution, respectively. A cyclohexane (Kanto Chemical Co., Inc.; special grade)/CV aq solution interface was used for measurement of the liquid/liquid interface. All the reagents were used as purchased

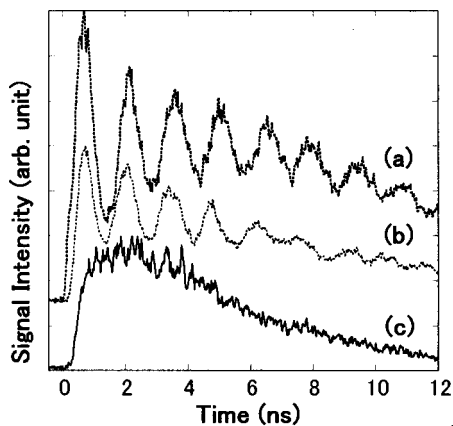


FIG. 2. (a), (b), and (c) are the TRG signals from the surfaces of the CV aqueous solution (36 mM), and black inks 2, and 1, respectively. Pump power, 2.9  $\mu\text{J}/\text{pulse}$ . Grating spacing, 2.3  $\mu\text{m}$ .

without further purification. The two black inks were diluted by purified water (from a Millipore Milli-Q system) and solutions were prepared in the range of 10–100 % w/w. The CV aq solutions were prepared in the range of 4.4–36 mM. The CV ethanol aqueous solutions were prepared at 10 mM and those solvents were prepared by mixing ethanol into purified water with ratios of 100%, 50%, 20%, 5%, and 0%. The optical absorption length of these samples was in the range of 5–10  $\mu\text{m}$ . About 5  $\text{mL}$  of these samples were added to a glass measurement cell which had a depth of 1 cm. As for the liquid/liquid interface, about 5  $\text{mL}$  of 10 mM CV aq solution were added before the addition of the same volume of cyclohexane to the solution.

## RESULTS AND DISCUSSION

The TRG signals from the surface of black ink 1, 2, and CV aq solution are shown in Fig. 2. The signal of black ink 1 has neither sharp peaks nor vibrations, but rather a gentle slope after the maximum value. Signals of black ink 2 and the CV aq solution have the largest and the sharpest first peak, and both show vibrations. This is the first observation of TRG signals with vibrations from liquid surfaces, which are very similar to those from solid surfaces.

In order to analyze these signals, we introduced the formula below, which was derived from the empirical equation for solid surfaces,<sup>39</sup>

$$S(t) = A[\exp(-t/\tau_{\text{th}}) - r \exp(-t/\tau_{\text{ac}})\cos(2\pi f_{\text{ac}}t)]^2 + A_{\text{el}}\exp(-2t/\tau_{\text{el}}), \quad (2)$$

where  $A$  and  $A_{\text{el}}$  are constants,  $r$  is the ratio of the acoustic effect to the thermal effect of each contribution to grating amplitude.  $\tau_{\text{th}}$ ,  $\tau_{\text{ac}}$ , and  $\tau_{\text{el}}$  are attenuation time constants for the temperature, the acoustic wave, and the excited species, respectively.  $f_{\text{ac}}$  is the frequency of the vibrations.

We calculated  $f_{\text{ac}}$  and  $\tau_{\text{ac}}$  of two TRG signals from black ink 2 and the CV aq solution by fitting with formula (1), and we also calculated each ultrasonic velocity  $V_{\text{ac}}$  as the product of  $f_{\text{ac}}$  and the fringe spacing  $\Lambda$ . These results are summarized in Table I. Both values of  $V_{\text{ac}}$  seem to be a little larger than that of the sound velocity in bulk water (1.48 km/s) at

TABLE I. The experimental frequencies of the vibrations  $f_{\text{ac}}$  and the attenuation time constants  $\tau_{\text{ac}}$  of the two TRG signals obtained by fitting formula (1), and the velocity  $V_{\text{ac}}$  calculated as the product of  $f_{\text{ac}}$  and the fringe spacing  $\Lambda$ .

Sample	$f_{\text{ac}}$ (GHz)	$\tau_{\text{ac}}$ (ns)	$V_{\text{ac}}$ (km/s)
Black ink 2	0.705	2.75	1.62
CV aq (36 mM)	0.687	3.37	1.57

20 °C. However, the discrepancies are within a reasonable range because the densities of black ink 2 and CV aq solution are 0.993  $\text{g}/\text{cm}^3$ , and 0.978  $\text{g}/\text{cm}^3$ , respectively, and the sound velocity is proportional to the (density)<sup>-1/2</sup>. Each attenuation time constant  $\tau_{\text{ac}}$  decreases with increasing viscosity (black ink 2, 1.3 cP; CV aq, 0.93 cP). The frequency of the TRG vibrations is much larger than that of a capillary wave of the same wavelength, which is experimentally observed at 1.7–1.8 MHz.<sup>40</sup> Therefore, we concluded that we had generated and observed GHz ultrasonic waves on liquid surfaces using the TRG method. Higher frequency vibrations can be also easily generated because  $\Lambda$  is controllable by changing the incident angle  $\theta$ .

As for the TRG signal from black ink 1, the reason why this signal shows no vibrations is not clear. However, we can presume that because of the unique behavior of carbon particles, something like a flow induced by heat from the carbon particles diminishes the TRG oscillations.<sup>41</sup>

Here, we discuss how the TRG oscillations on liquid surfaces are generated. Elastic theory shows that the surface is deformed so as to satisfy the boundary condition when an acoustic wave adjacent to the surface is generated.<sup>29,30,32,33</sup> On the other hand, such an acoustic wave also causes modulation of the refractive index.<sup>27,28</sup> Nelson and co-workers<sup>33</sup> showed that the oscillations of TRG signals at the surface are mainly caused by the surface displacement modulation and not by the refractive index modulation in the case of the surface measurement. In addition to the report, we found from diffraction theory considerations that the refractive index modulation was a minor component of the signal.<sup>31,42</sup> Therefore, we can see that the TRG oscillations in the surface signal consist of the modulation of the surface deformation as the main component and that of the refractive index as the minor component, both of which are induced by acoustic waves localized near the surface. The oscillating signal contains information in the  $\Lambda/2$  region from the surface, that is, 1  $\mu\text{m}$  in this case, because the acoustic waves which causes the surface displacement are localized in the surface region of the waves' half-wavelength.<sup>43</sup>

We investigated the pump power dependence of the TRG signals from surfaces of black ink 2 and CV aq solution. We examined the measured magnitude of the average intensities of the first peaks of each signal. The results of the CV aq solution are shown in Fig. 3. The figure represented that the TRG signals were proportional to the square of the pump power. According to the TRG theory on solid surfaces, the surface displacement is directly proportional to the temperature change. In turn, the temperature change is directly proportional to the pump power. Since diffracted light is

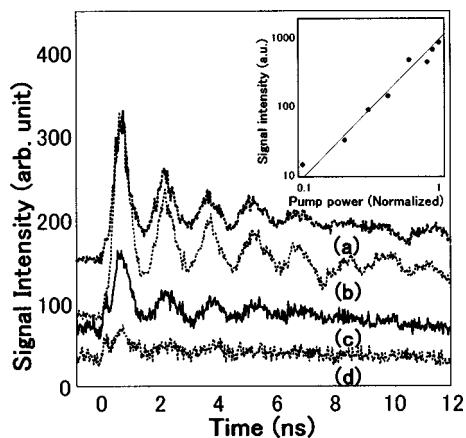


FIG. 3. The pump power dependence of the intensities of the TRG signals from the surface of the CV aqueous solution (20 mM). Grating spacing, 2.4  $\mu\text{m}$ . The pump power intensities of signals (a), (b), (c), and (d) are 1.1, 0.78, 0.39, and 0.26  $\mu\text{J}/\text{pulse}$ , respectively. The averaged intensity of the TRG signal from the surface of the CV solution is proportional to the square of the pump power.

generally proportional to the square of surface displacement, the TRG signal is proportional to the square of the pump power. The agreement between the theory and this experimental result indicates that the TRG theory can be fundamentally applied to liquid surfaces just as with solid surfaces.

We consider what influences the signal amplitude, that is, surface displacement. The surface displaces due to the applied force to the surface. Referring to the elastic theory of liquids<sup>44</sup> and the theory of capillary waves,<sup>45</sup> four forces are raised as the applied forces to the surface, that is, elastic force, viscosity, surface tension and gravity. In this case, gravity can be negligible. Although mixture of solvents and addition of solutes will cause other physical properties changes, we can suppose that the influences are included in the signal through these forces change. The viscosity and surface tension dependence of the TRG signals from surfaces of the CV ethanol aqueous solutions was investigated. It is well known that the viscosity and the surface tension are greatly influenced by the portion of ethanol in water.<sup>46</sup> The relationship between the viscosity and surface tension of ethanol water mixtures at 25  $^{\circ}\text{C}$  is shown in Fig. 4(A). The solvents were prepared by mixing ethanol into water at ratios of 100%, 50%, 20%, 5%, and 0%. The CV solutions were prepared at 10 mM with each solvent. The results are shown in Fig. 4(B). Amplitude of each signal is greatly affected. We can suppose that the elastic force of the ethanol aqueous solutions changes monotonically with the ethanol concentration because the sound velocity of the solution changes monotonically. But the signal amplitude did not change monotonically with the ethanol concentration. Therefore we can see that the signal dependence is regarded due to the viscosity and/or surface tension, though it is not clear how much each of them affects the TRG signal. Furthermore, TRG signals were different in waveform at 0%, 5%, and 100%, where the surface tension changes while the viscosity is fairly constant. Though it is too much to say that these signals depends only on the surface tension, this dependence

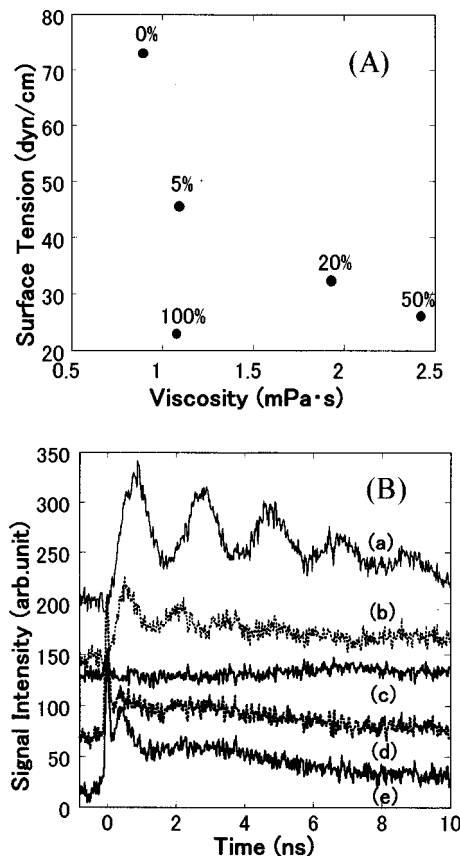


FIG. 4. (A) The relationship between the viscosity and the surface tension of ethanol–water mixtures. The percentages above the circles are the volume ratio of ethanol. (B) The TRG signals from the surfaces of the CV ethanol aqueous solutions (10 mM). Grating spacing, 2.4  $\mu\text{m}$ . Pump power, 1.8  $\mu\text{J}/\text{pulse}$ . The ratios of ethanol in the solution (a), (b), (c), (d), and (e) are 100%, 50%, 20%, 5%, and 0%, respectively.

implies the high selectivity of the TRG signals for the interface. For more exact analysis, we must solve the elastic equation including viscosity and surface tension as perturbations, and we are now in progress. In addition to the viscosity and surface tension, we suppose that the absorption of the acoustic wave by solutes will affect the signal waveform for measurements at the specified wavelength. The information will give some information on the solute–solvent interaction at the surface or interface.

As shown in Fig. 3, the first peak of the TRG signal splits as the intensity of the pump power decreases. As shown in Fig. 4(B), the first sharp peak of the signal diminishes as the ratios of ethanol in the solvents of the CV solutions change. These peaks seem to reflect the dynamics of the relaxation of the electronically photoexcited state of CV molecules on the surface. Thus, it would be possible to clarify this dynamics and to investigate the solute–solvent effect on the surfaces by applying a femtosecond pulsed-laser to the surface of the CV aqueous solution.

As for liquid/liquid interfaces, we also applied the TRG method to the cyclohexane/CV aq solution interface. The results are shown in Fig. 5. The signal at the liquid/liquid interface shows oscillations like the liquid surfaces did. The frequency of these oscillations was calculated as 0.6 GHz. The pump power dependence of this signal was investigated.

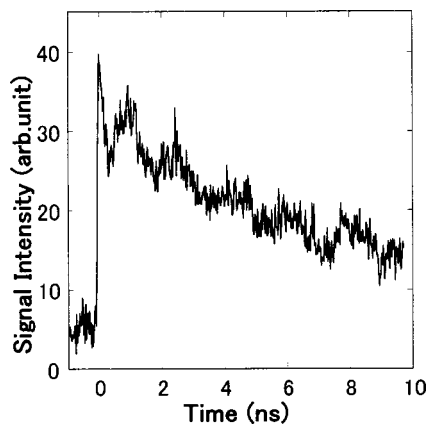


FIG. 5. The TRG signal from the cyclohexane/CV aqueous solution (10 mM) interface. Grating spacing,  $2.3 \mu\text{m}$ . Pump power,  $0.23 \mu\text{J/pulse}$ . Probe power,  $0.035 \mu\text{J/pulse}$ .

The signal is proportional to the square of the pump power. Thus, we conclude that we can generate and observe ultrasonic waves at the liquid/liquid interface as well as at liquid surfaces.

As shown before, it is easy to generate ultrasonic waves of various frequencies with the TRG method by changing the interference fringe spacing. By analyzing the fringe spacing dependence of the TRG signals, it is expected that we can apply the TRG method to ultrasonic measurements of interfaces and to ultrasonic spectroscopy in order to investigate the relaxation phenomena of molecules at interfaces. It is also expected that we can investigate fast phenomena on a surface and at a liquid/liquid interface microscopically, such as the solute-solvent effect by using a femtosecond pulsed laser.

## CONCLUSIONS

The 0.7 GHz ultrasonic wave on liquid surfaces and 0.6 GHz ultrasonic wave at a liquid/liquid interface were successfully generated and observed using the TRG method for the first time. These waves were considered to have a surface displacement normal to the surface and were considered to include information about the surface region to a penetration depth of  $1 \mu\text{m}$ . It is expected that this method will be a powerful tool to observe and analyze fast phenomena microscopically on liquid surfaces and liquid/liquid interfaces.

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