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Ripplon Spectroscopic Study on Liquid Surface under Temperature Gradient
定常熱非平衡状態におけるリプロン伝搬の観察

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1. Introduction

Ripplon is a thermally excited capillary wave, whose propagation can be observed with a ripplon light scattering spectroscopy[1]. Up to now, we have applied this technique for the investigation of the dynamic molecular properties at the liquid surface and the liquid-liquid interface in the thermodynamic equilibrium state. Recently, a number of theoretical and experimental studies have been carried out on the phonon propagation in bulk liquid under non-equilibrium state with a temperature gradient[2], while few works have been reported on the liquid surface in the non-equilibrium. In this study, we applied our theoretical model of phonon propagation under temperature gradient to the ripplon. In the theory, we easily take all the actual experimental conditions into account. In the experiment, we observed the propagation of the ripplons on the liquid surface under a large temperature gradient in the range of 50 to $100\,\mathrm{K/cm}$ with the ripplon spectroscopic technique.

2. Theory

In the thermodynamically non-equilibrium state, the number density of the ripplons propagating from the higher temperature region to the lower is higher than that of the ripplons propagating to the opposite direction, since ripplons are generated by thermal fluctuations $k_B T$. Therefore, intensities of the Stokes and anti-Stokes components of the power spectrum are not equal, and the spectrum becomes asymmetric. Here, the asymmetric parameter ε is defined as

$$\varepsilon = \frac{I_{st} - I_{as}}{I_{st} + I_{as}} \tag{1}$$

where I_{st} and I_{as} are the intensity of the Stokes and anti-Stokes component, respectively. The intensity of each component is calculated by a simple theoretical description, which is originally proposed in the study of the Brillouin scattering with a temperature gradient[3] and applied to this study. The idea of the theory is that ripplon is generated with a probability proportional to the local temperature at the excited point, and propagates into the scattering region with a spatial attenuation. We can calculate the intensities of the ripplon propagating from the colder region to the scattering region, I_{cold}, and from the hotter region to the scattering region, I_{hot}, as

$$I_{cold} \propto \int_{-b}^{0} k_B T(x) \exp(2\alpha x)\, dx \tag{2}$$

$$I_{hot} \propto \int_{0}^{b} k_B T(x) \exp(-2\alpha x)\, dx \tag{3}$$

where α is the spatial attenuation constant of the ripplon[4], $2b$ is the gap distance between the boundary giving the high and low temperatures, k_B is the Boltzmann constant, and $T(x)$ is the temperature profile of the practical experimental condition. One of I_{cold} and I_{hot} corresponds to I_{st} and the other to I_{as}, which can be determined from the asymmetric ripplon spectrum. In this calculation, the effect of the ripplon reflected by the cell boundaries is ignored. Note that when the temperature gradient is uniform and the distance between the boundary cell is much larger than the attenuation length, which means no boundary effect, eqs. (2) and (3) give the same asymmetric parameter, which has been represented in the number of theoretical predictions[5], expressed as

$$\varepsilon = \frac{1}{2\alpha} \hat{k} \cdot \frac{\nabla T}{T_0} \tag{4}$$

where ∇T is the temperature gradient, and T_0 is the temperature at the scattering point.

3. Experimentals

The experimental setup is shown in Fig. 1. By switching the optical path with a flip mount, the incident light can enter symmetrically with the local light, and we can easily switch the position of the Stokes and anti-Stokes components, which cancels out the frequency response of the detecting system.

The substrate of the sample liquid is pure water. The surface tension of the pure liquid becomes, however, smaller as the temperature increases. The surface tension gradient induced by a temperature gradient drives the Marangoni convection. When insoluble molecules are spread on the pure liquid surface with a temperature gradient, adsorbed molecular layer expands and suppresses the Marangoni flow. As a result, the surface tension is kept constant in any region, and the mechanically stable state can be achieved. In our experiment, we used water covered with ethyl myrystate monomolecular film as a sample liquid.

4. Results and discussions

Figure 2 shows typical examples of non-equilibrium spectra. The solid curves are fitting curves, which give experimental values of asymmetric parameters. Figure 3 shows the comparison of the experimental values with a theoretically predicted one from eq. (2) and (3), expressed as dashed lines, and one from eq. (4), expressed as solid lines, at each wavenumber. Ripplon with lower wavenumber has a longer attenuation length, and the effect of the boundaries is more active. Details will be commented in our presentation, and experimental results well described and well agreed with our theoretical estimation.

References

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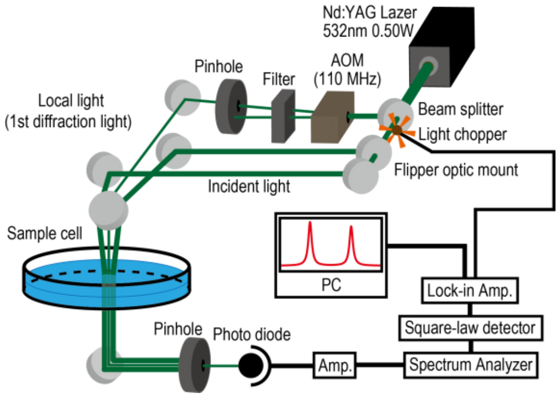

Fig.1 Schemaic view of the Ripplon Spectroscopy technique.

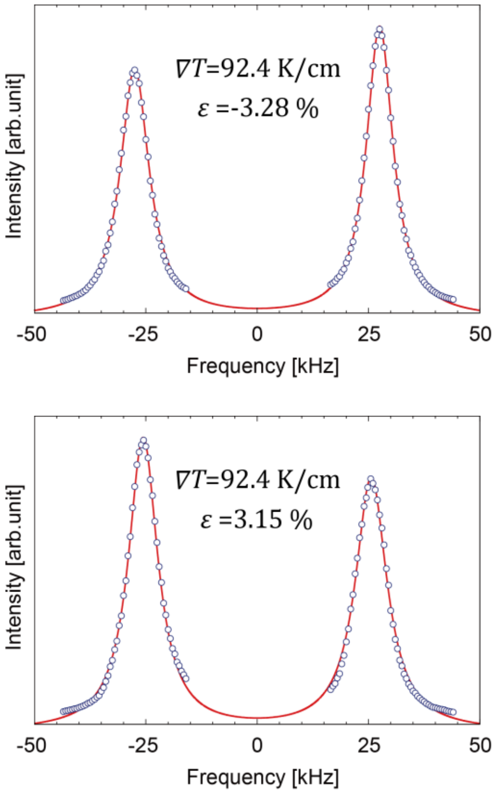

Fig. 2 Typical examples of spectra with a temperature gradient. The wavenumber is $8.27 \times 10^4\ \mathrm{m}^{-1}$

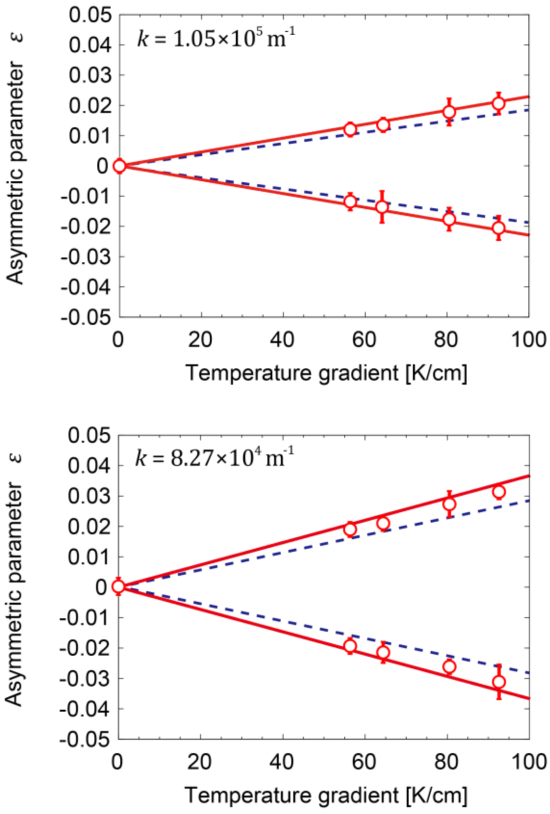

Fig. 3 Comparison of the experimental data with the theoretical estimation using eqs. (2), (3), and (4).