Experimental Study on Phase Velocity of Ultrasonic Waves Decaying in Space and in Time

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Modified system of high-resolution Bragg reflection was used to observe both the spatial and temporal sound waves simultaneously in the relaxation region of liquid furan (C_4H_4O). The spatial phase velocity at 100 MHz was shown to be higher than the temporal velocity by 7 m/s at 20 °C. The result is in good agreement with the theoretical prediction, providing an experimental evidence of the difference between them.

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The phase velocity of sound in a medium with some absorption mechanism is somewhat ill defined since it is impossible to determine both the angular frequency and the wave number simultaneously. A sound wave generated by a constant source propagates and decays *in space*, and therefore the wave number loses its strict meaning, having a distribution with a width proportional to the absorption. On the other hand, reverberation of sound in an acoustic resonator would decay *in time* while staying at the same wave number, but with its frequency being distributed. In this regard, two different definitions of phase velocity are given: one relevant to spatial decay and another to temporal decay. Among the ultrasonic techniques currently used, pulse method or interferometer observes the former; Brillouin scattering, the latter. Theoretical analysis of the complex equation of dispersion has predicted that the velocity of spatial wave should have a value higher than that of temporal wave.[1,2] No experiment has been made, however, to provide clear evidence of the difference between them, although some workers have emphasized the importance of this problem[3] and suggested the possibility of its detection.[4]

A few years ago, we established a high-resolution Bragg-reflection technique[5,6] useful for the measurement of spatial sound in a frequency range up to 1.5 GHz. After a recent modification of the electronic system, this technique is capable of yielding either of the two phase velocities. In this work, we made an experiment in the relaxation region of liquid furan (C_4H_4O) and succeeded in observing the difference between them.

The underlying principle and theoretical basis of this technique have been given previously.[5,6] The modified experimental system used in this work is schematically shown in Fig. 1. An ultrasonic wave is excited by a ZnO-film transducer evaporated on the surface of a quartz rod, and a laser beam is introduced into the test liquid. If the angle of incidence with respect to the sound wave front is close to the Bragg angle appropriate to the sound frequency, a portion of the incident light is scattered and detected in an optical heterodyne system. The wave number of the sound causing the scattering should be related with the angle of incidence through the following equation of momentum conservation

$$2q \sin\theta = k, \tag{1}$$

where q and k are wave numbers of the light and sound, respectively. A combination of tracking generator (TG) and spectrum analyzer (SA) is used for excitation of the sound and reception of the photobeat current. To observe the spatial wave, frequency of SA/TG is fixed at ω and the angle of incidence is swept around the Bragg angle by rotating the table on which the scattering

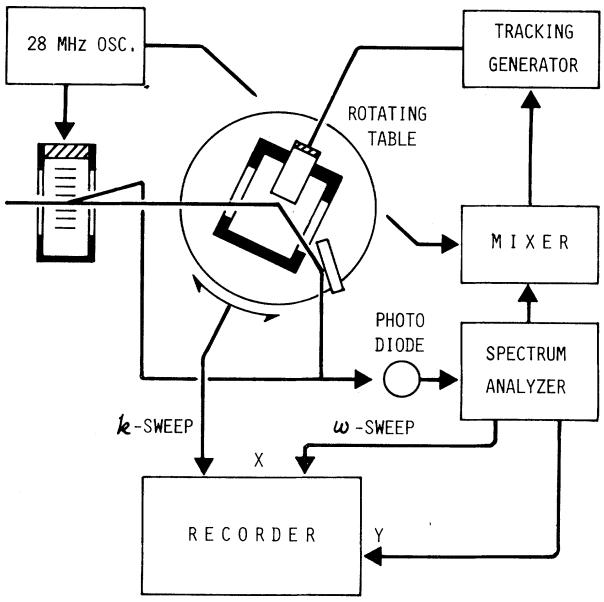

FIG. 1. Modified system of high-resolution Bragg reflection.

cell is mounted. The beat signal recorded as a function of the angular sweep is associated with the k spectrum of the spatial sound. The peak of the curve gives k', center of the k spectrum, from which the spatial phase velocity is obtained:

$$v_s = \omega/k' = \omega/(2q \sin\theta'), \quad (2)$$

where θ' is the peak angle. To observe the temporal wave, the rotating table is locked at θ and, instead, SA/TG is swept. The recorded curve gives the ω spectrum of sound at fixed value of k. The temporal phase velocity is obtained by

$$v_t = \omega'/k = \omega'/(2q \sin\theta), \quad (3)$$

where ω' is the center of the ω spectrum.

An experiment was made in furan at 20 °C near 100 MHz. This liquid is a suitable medium for the present purposes since it has strong dispersion originating from vibrational relaxation,[7] and hence a nonnegligible difference between v_s and v_t is expected to be observed in the relaxing region. The obtained spectra are shown in Fig. 2. The upper curve is appropriate to the k spectrum at $\omega/2\pi = 100.926$ MHz. The peak angle, given to be $\theta' = 1°31'10''$, yields $v_s = 1205$ m/s. Then, the table is fixed at the above angle, $\theta = \theta'$; and SA/TG is swept around 100 MHz. The lower curve of Fig. 2 shows the ω spectrum thus obtained. If

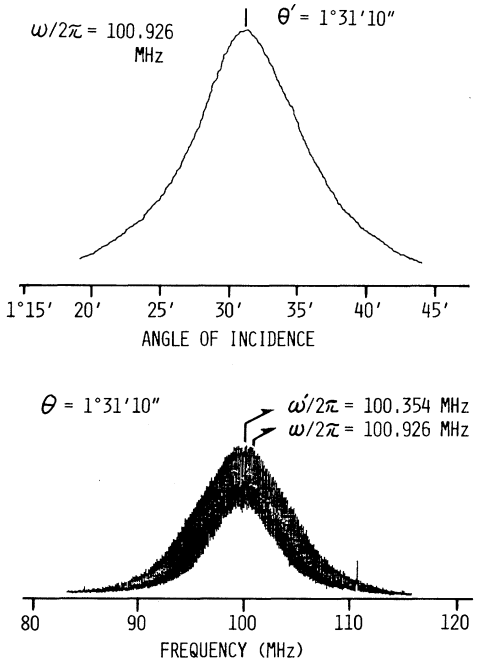

FIG. 2. Recorded curves of k spectrum (upper) and ω spectrum (lower).

v_t were equal to v_s, the spectrum should have taken maximum at 100.926 MHz. Because of the difference, however, the peak is shifted down to $\omega'/2\pi = 100.354$ MHz, giving $v_t = 1198$ m/s.

One of the problems to be kept in mind throughout the experiment is the frequency characteristics of the electronic and/or ultrasonic system. The most remarkable one is that of the ultrasonic transducer. The resonance of ZnO film itself is heavily damped by mechanical load of quartz rod and perfectly flat within the range of sweep. The rod works as an acoustic resonator, however, and sharp peaks occur at every frequency of half wavelength as shown in the lower curve of Fig. 2. The interval is 150 kHz, in agreement with the value estimated from longitudinal velocity and length of the rod, 6000 m/s and 2 cm, respectively. These resonances would not have harmful effects on the experiment except that the spectrum obtained is sampled discretely. Nevertheless, the most troublesome one is the frequency dependence in insertion loss of the transducer viewed from the generator, which may shift the apparent peak of the spectrum. We accomplished almost ideal flatness by carefully adjusting a three-stub tuner placed between the generator and the transducer. From a technical point of view, this requisition is surely the most difficult and limits the frequency range of this experiment.

Markham, Beyer, and Lindsay[1] discussed the problem of spatial decay and temporal decay theoretically and derived dispersion equations for both types of waves. When a single relaxation process is the only mechanism of absorption in a medium, the secular equation characterizing the propagation of sound is given by

$$\omega^{*2}(\omega^*/v_\infty^2 - i\omega_r/v_0^2) = k^{*2}(\omega^* - i\omega_r),$$

where stars indicate a complex number, v_0 and v_∞ are the limiting velocities at lower and higher frequencies, respectively, and ω_r is the relaxation frequency of adiabatic compressibility, corresponding to the inflection point of the curve of $(\alpha/\omega^2)(v_0/v)$ vs $\ln\omega$. This equation gives a proper description of dispersion in furan because more than 98% of total absorption at 100 MHz is due to the relaxation. For the spatial wave with absorption coefficient α, one sets $\omega^* = \omega$ and $k^* = k' - i\alpha$. Then, v_s is derived as a function of ω by

$$v_s^2\left[1 + \left(\frac{\alpha}{k'}\right)^2\right] = v_\infty^2 - \frac{v_\infty^2 - v_0^2}{1 + (\omega/\omega_1)^2}, \quad (4)$$

where $\omega_1 = (v_\infty/v_0)\omega_r$. For the temporal wave, $k^* = k$ and $\omega^* = \omega' + i\Gamma$, Γ being the decay rate.

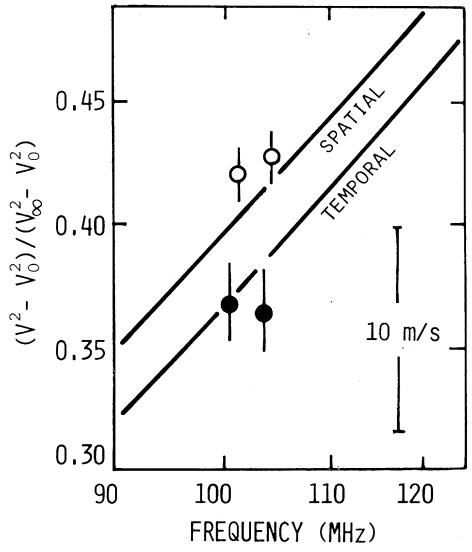

FIG. 3. Values of v_s and v_t observed in the relaxation region of liquid furan. The solid lines represent the theoretical curves of Eqs. (4) and (5).

Hence, v_t is given as a function of ω' by

$$v_t^2\left[1-\left(\frac{\Gamma}{\omega'}\right)^2\right] = v_\infty^2 - \frac{(v_\infty^2 - v_0^2)(1-\Gamma/\omega_2)}{(1-\Gamma/\omega_2)^2 + (\omega'/\omega_2)^2}, \quad (5)$$

where $\omega_2 = (v_\infty/v_0)^2\omega_r$. Note that v_s and v_t are close to the same value, v_0 or v_∞, outside the relaxation region.

Ultrasonic relaxation of furan has been investigated in the previous work[7] and all the values required for numerical computation of these equations are available. The characteristic values at 20 °C are $\omega_r/2\pi = 109$ MHz, $v_0 = 1154$ m/s, $v_\infty = 1272$ m/s. The quantity α/k' has been measured at each frequency and Γ/ω' is approximately set equal to α/k'. Figure 3 shows the curves of Eqs. (4) and (5) normalized with respect to $v_\infty^2 - v_0^2$. They are in good agreement with the experimental values of v_s and v_t indicated by open and closed circles, respectively. Although the accuracy in v_s is better than 0.1%, v_t may include error somewhat larger than 0.2% because of the discrete character of the recorded spectrum. The overall accuracy of this experiment is, however, sufficient to detect the difference between v_s and v_t.

We emphasize here that v_t in this work should not be confused with the phase velocity obtained by spontaneous Brillouin scattering. The Brillouin shift is not identical with ω' because of the antisymmetric term of Brillouin component. This problem is inherent in the difference between *thermally driven* temporal wave and *freely oscillating* temporal wave, as has been discussed by Montrose, Solovyer, and Litovitz,[8] and by Nichols and Carome.[2] The Brillouin phase velocity may even be smaller than v_t.

[1]J. L. Markham, R. T. Beyer, and R. B. Lindsay, Rev. Mod. Phys. 23, 353 (1951).
[2]W. H. Nichols and E. F. Carome, J. Chem. Phys. 49, 1000 (1968).
[3]G. I. A. Stegeman, W. S. Gornall, V. Volterra, and B. P. Stoicheff, J. Acoust. Soc. Am. 49, 979 (1971).
[4]R. D. Mountain and T. A. Litovitz, J. Acoust. Soc. Am. 42, 516 (1967).
[5]K. Takagi, P.-K. Choi, and K. Negishi, J. Acoust. Soc. Am. 62, 354 (1977).
[6]K. Takagi and K. Negishi, Ultrasonics 16, 259 (1978).
[7]K. Takagi and K. Negishi, J. Appl. Phys. Jpn. 16, 1319 (1977).
[8]C. J. Montrose, V. A. Solovyev, and T. A. Litovitz, J. Acoust. Soc. Am. 43, 117 (1968).