

Phase conversion and interface growth in phase-separated ^3He - ^4He liquid mixtures

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We have developed a method for measuring the transmission coefficient of a sound propagating through the interface in phase-separated ^3He - ^4He liquid mixtures. The method and the results are described with discussions by examining the phase-conversion process of ^3He quasiparticles driven to flow across the interface. From the data, we have determined the kinetic growth coefficient of the interface, $\xi(T, P, \omega)$, as a function of temperature, pressure, and frequency. The temperature range of the present investigation is about 2–100 mK at the pressure mainly around 1 bar with sound frequency 9.64, 14.4, and 32.4 MHz. The main specific features observed for the kinetic growth coefficient are, as follows: (i) there is a maximum at some temperature $T_m(\omega)$ depending on the frequency, (ii) above $T_m(\omega)$, ξ decreases with the increase of temperature as $\propto \omega^{5/2}T^{-3}$, and (iii) below $T_m(\omega)$, ξ becomes frequency independent and diminishes as a cube of temperature, T^3 .

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I. INTRODUCTION

First-order phase transitions are widespread in nature. Some phase transitions and interfacial boundaries exist at very low temperatures down to absolute zero. Such transitions and interfaces, having unique properties compared with high-temperature classical systems, have been studied intensively in helium.¹⁻⁵

The kinetics of phase transitions and interfacial dynamics depend strongly on $\omega\tau$, where τ is the relaxation time and ω is the frequency of the process. In this respect helium systems are very convenient since the relaxation time, depending drastically on temperature, can be varied by many orders of the magnitude. The kinetics of phase transitions within the linear Onsager approach is described in terms of the kinetic growth coefficient ξ which connects the mass flow across the interface and the imbalance in chemical potentials $\Delta\mu$ between the adjacent phases. Experiments in pure ^4He with the vapor-liquid⁶ and liquid-solid^{7,8} interfaces have revealed unusual behavior in ξ compared with classical interfaces. In this paper we have turned to studying the phase transition in a ^3He - ^4He liquid mixture.

Below ~ 0.9 K, the liquid ^3He - ^4He mixtures decompose into the ^3He -dilute phase (d phase) and the ^3He -concentrated phase (c phase). The interface between the coexisting d and c phases provides us a unique system as a spontaneously existing boundary between two quantum liquids, one being superfluid and the other being normal or superfluid at sufficiently ultralow temperatures. The interface has been studied from various viewpoints such as the interfacial tension,⁹ the wetting phenomena,¹⁰ the Helmholtz-Kelvin instability,¹¹ and the growth properties.¹² In this paper, we will give the results of our systematic study of the sound transmission across the interface. Then, we employ the relation between the sound transmission coefficient and the kinetic growth coefficient in order to extract information on the growth dynamics of the interface in the nonequilibrium situation.

At low temperatures below ~ 100 mK, the c phase is almost pure liquid ^3He , the Fermi temperature being ~ 1.6 K.

On the contrary, the d phase represents a solution of a finite concentration of ^3He in superfluid ^4He down to absolute zero. The Fermi temperature of the saturated d phase is approximately 0.4 K. At low enough temperatures the d phase behaves as a Fermi liquid of ^3He quasiparticles moving in the background of superfluid ^4He . So, at such low temperatures, the interface presently studied is that between two different degenerate Fermi liquids. The size of their Fermi spheres has a significant difference since

$$p_{Fc} \simeq 2.3 \times p_{Fd}, \quad (1)$$

where p_{Fc} and p_{Fd} represent the magnitude of the Fermi momentum in the c and d phases, respectively.

The saturated concentration of ^3He in the d phase, $x_s(T, P)$, is known to be sensitive to the pressure,¹³ changing within 0.067–0.095 in the present temperature range studied. So a pressure modulation or sound wave injected onto the interface can produce a phase conversion between the c and d phases. This results in a flow of one phase into the other across the interface, entailing the motion of the interface. We call this motion of the interfacial boundary the interface growth. That is, we borrow the terminology from ^4He superfluid-crystal growth,¹⁴ keeping in view some common features in the sound transmission. Thus, studying a response of the interface on the pressure modulation, we can obtain information about the kinetic growth properties of the interface. In the phase conversion we can discern two stages. The first is a mass flow driven by the pressure modulation. At low enough temperatures, it represents practically a flow of ^3He quasiparticles alone. At the second stage the quasiparticles incoming into the d phase from the c phase should be converted to the quasiparticles of the d phase and vice versa. At this stage one may expect that a significant difference in the Fermi momentum between the phases as Eq. (1) plays a crucial role, especially in the very-low-temperature region where ^3He quasiparticles flow ballistically across the interface.

Our present experiment covers a wide temperature range approximately 2–100 mK with the sound of 9.64, 14.4, and 32.4 MHz. In the temperature behavior of the kinetic growth coefficient, we have observed a crossover between the hydrodynamic and ballistic regimes.

II. EXPERIMENT

The method of injecting an acoustic wave onto the interface has widely been applied in the course of studying the kinetic growth properties and phase-conversion phenomena for the vapor-liquid⁶ and liquid-solid^{7,15,16} interfaces in pure ⁴He. The transmission of a sound wave through the interface depends strongly on the interface mobility.

The mobility of the interface is usually discussed in terms of the kinetic growth coefficient ξ . For a single-component system, in general, ξ depends on temperature T and frequency ω —i.e., $\xi = \xi(T, \omega)$. The mass conservation implies¹⁴

$$J = \rho_1(v_1 - \dot{\zeta}) = \rho_2(v_2 - \dot{\zeta}), \quad (2)$$

where J is the mass flow across the interface per unit time and unit area, $\dot{\zeta}$ is the velocity of the interfacial boundary, and $v_{1,2}$ and $\rho_{1,2}$ are the fluid velocities due to sound and the densities of the phases, respectively. By introducing ξ with the equation

$$v_1 - v_2 = \xi \delta P, \quad (3)$$

we have

$$\dot{\zeta} = v_1 + \frac{\rho_2}{\rho_1 - \rho_2} \xi \delta P = v_2 + \frac{\rho_1}{\rho_1 - \rho_2} \xi \delta P \quad (4)$$

and

$$J = \xi \left(\frac{\rho_1 \rho_2}{\rho_1 - \rho_2} \right) \delta P. \quad (5)$$

Here δP is the pressure deviation from phase-equilibrium pressure $P(T)$ due to sound perturbation. From Eqs. (3)–(5) we recognize that, if $\xi=0$, the mass flow J vanishes in spite of the phase imbalance as for the case of two immiscible liquids where any phase growth is absent—i.e., $\dot{\zeta}=v_1=v_2$. On the other hand, if ξ does not vanish, we have a finite magnitude of mass flow J and a finite difference in v_1-v_2 . That is, we have the growth of one of the phases. In essence, ξ thus introduced determines the rate of phase conversion and is called the kinetic growth coefficient of the interface.

The kinetic growth coefficient is known to be related to the sound transmission and reflection coefficients. For the oblique incidence of sound from phase 1 onto phase 2, the transmission coefficient $\tau_{1 \rightarrow 2}$, defined by the ratio of the sound pressure amplitudes of the transmitted and incident waves, is given by

$$\begin{aligned} \tau_{1 \rightarrow 2} = & 2Z_2 \cos \vartheta_1 \left(1 - i\omega\sigma_i \frac{\rho_1^2 \rho_2^2}{(\rho_1 - \rho_2)^2} \xi \frac{\sin \vartheta_1 \sin \vartheta_2}{Z_1 Z_2} \right) \\ & \times \left\{ Z_1 \cos \vartheta_2 + Z_2 \cos \vartheta_1 - \frac{1}{4} \frac{i\omega\sigma_i}{u_1 u_2} \sin 2\vartheta_1 \sin 2\vartheta_2 \right. \\ & + Z_1 Z_2 \xi \left[1 - \frac{i\omega\sigma_i}{2Z_1 Z_2} \frac{\rho_1 \rho_2}{(\rho_1 - \rho_2)^2} \left(\frac{\rho_1}{u_1} \sin 2\vartheta_1 \sin \vartheta_2 \right. \right. \\ & \left. \left. + \frac{\rho_2}{u_2} \sin 2\vartheta_2 \sin \vartheta_1 \right) \right] \right\}^{-1}. \quad (6) \end{aligned}$$

Here ϑ_1 is the incident angle, ϑ_2 is the refraction angle of the transmitted wave, and σ_i is the interfacial surface tension. The acoustic impedance of the phase denotes a product of density and sound velocity—i.e., $Z_{1,2} = \rho_{1,2} u_{1,2}$.

The case of the phase-separated ³He-⁴He liquid mixtures is considered in Ref. 17. In mixtures such as a two-component system the growth coefficient depends on the temperature, pressure, and frequency—i.e., $\xi = \xi(T, P, \omega)$. For normal incidence, the sound transmission from the c phase to the d phase, $\tau_{c \rightarrow d}$, reads

$$\tau_{c \rightarrow d} = \frac{2Z_d}{Z_c + Z_d + Z_c Z_d \xi}, \quad (7)$$

where Z_c and Z_d are the acoustic impedance of the c and d phases, respectively.

Provided the response of the interface on the pressure modulation is slow—i.e., the phase conversion from one phase to another is not so efficient and the interface mobility is extremely small—the sound transmission will be the same as for the case of two immiscible liquids with

$$\tau_{0,c \rightarrow d} = \frac{2Z_d}{Z_c + Z_d}. \quad (8)$$

On the contrary, if the phase conversion between the phases occurs at an infinite rate and the interface has a high mobility, the system is able to retain the phase equilibrium state and to smooth the total magnitude of the pressure modulation at the interface down to zero. This means that a sound wave incident onto the interface from one phase should not cause any pressure modulation at the interface and, thus, in the other phase. Hence, in this ideal case, the sound transmission into the other phase vanishes. Thus the coefficients of sound transmission and reflection at the interface may give certain information on the rate of conversion processes between two adjacent phases.

A. Principle of the method

The sound attenuation in the c phase is very large and that in the d phase also is not so small. Therefore, a new idea is required as to how to extract the information on the sound transmission coefficient across the interface from the sound transmitted through the bulk liquids. That is, it is necessary to separate the bulk contribution from the transmitted signal. For this purpose, we have developed¹² a way to move the position of the interface very slowly between two transducers with an almost constant velocity.

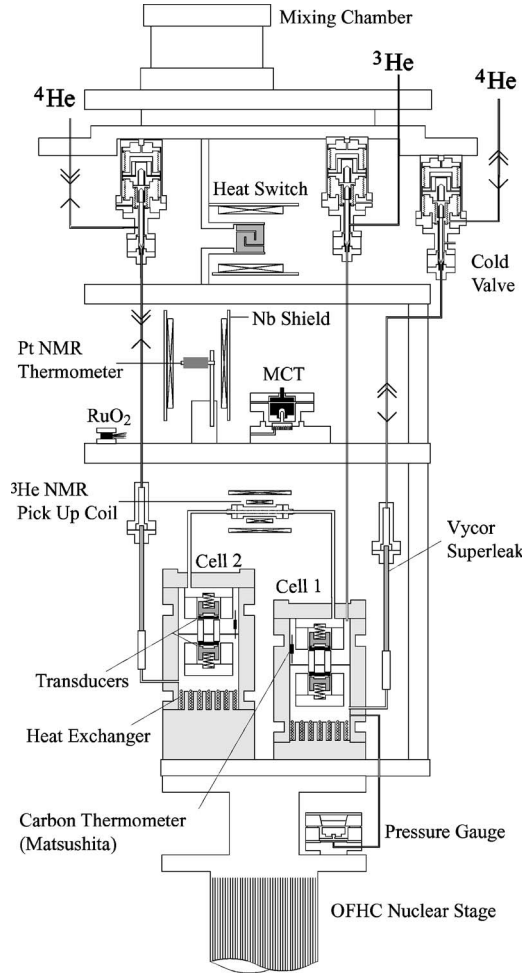


FIG. 1. Schematic drawing of the principal part of our cryostat.

As can be seen in the lower part of Fig. 1, the details of which will be explained in Sec. II B, we have two sound cells, cell 1 and cell 2, which are of almost the same inner diameter. They are connected with each other by a tube. Each cell contains a pair of transducers and has a Vycor superleak line connected to the bottom side of the cell. A ^3He line equipped with a cold valve¹⁸ is connected to one of the cells (cell 1 in Fig. 1) at the top side.

First, pure ^4He is condensed into the cells through the superleak lines. Then, ^3He is condensed through the ^3He line and at the same time ^4He flows out through the superleak lines. After obtaining an appropriate ratio of c and d phases, we close the cold valve in the ^3He line in order to keep constant the total amount of ^3He in the cells. We stabilize the temperature under desired pressure which can be controlled by adding or pouring ^4He out through the superleak lines.

As superfluid ^4He flows into cell 1 through its superleak line, the interface in cell 1 moves up. At the same time superfluid ^4He flows out from cell 2 through its superleak line and the interface in cell 2 shifts down. The process can also be done in a reversed way. The flow-controlling system outside of the cryostat is designed so as to realize the motion of the interface with almost constant velocity under nearly constant pressure.

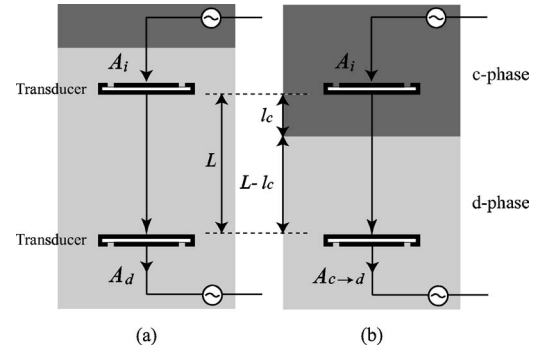


FIG. 2. Illustration of our method for obtaining the transmission coefficient through the interface, $\tau_{c \rightarrow d}$, with the sound transmitted from the upper transducer to the lower one. The rf voltage applied to the transmitter transducer is A_i , and A_d or $A_{c \rightarrow d}$ is a signal induced at the receiver transducer. (a) The interface lies far above the upper transducer. (b) The interface lies between the transducers.

Let us consider, as is shown in Fig. 2, one of the cells in which the sound is transmitted from the upper transducer and received with the lower one separated by the distance L . We start moving the interface from the position far above the upper transducer [Fig. 2(a)]. As we have only the saturated d phase between the transducers, we can write the following relation between the rf voltage applied to the transmitter transducer, A_i , and that induced at the receiver transducer, A_d , as

$$A_d = \frac{a_{up}(\omega)}{a_{low}(\omega)} A_i \tau_{t \rightarrow d} \tau_{d \rightarrow t} \exp[-\alpha_d L] \exp[-i\omega L/u_d]. \quad (9)$$

Here, u_d and α_d are the velocity and attenuation coefficients of sound in the d phase, respectively. The transmission coefficient of sound from the transducer to the d phase is denoted with $\tau_{t \rightarrow d}$ and that from the d phase to the transducer with $\tau_{d \rightarrow t}$. The notations $a_{up}(\omega)$ and $a_{low}(\omega)$ represent the conversion factors from the rf voltage signals, A_i and A_d , to the sound signals in the corresponding transducers, A_{up} and A_{low} , respectively. Thus, for the situation of Fig. 2(a) we may have

$$A_{up} = a_{up}(\omega) A_i \text{ and } A_{low} = a_{low}(\omega) A_d. \quad (10)$$

When the interface moves down and enters into the region between the transducers [Fig. 2(b)], the rf voltage from the receiver transducer, $A_{c \rightarrow d}$, is expressed as

$$A_{c \rightarrow d} = \frac{a_{up}(\omega)}{a_{low}(\omega)} A_i \tau_{t \rightarrow c} \tau_{c \rightarrow d} \tau_{d \rightarrow t} \exp[-\alpha_c l_c] \exp[-\alpha_d (L - l_c)] \\ \times \exp[-i\omega \{l_c/u_c + (L - l_c)/u_d\}], \quad (11)$$

where $\tau_{c \rightarrow d}$ is the transmission coefficient of sound through the interface. This is the quantity we want to obtain. In Eq. (11), l_c is the distance between the upper transducer and the interface as shown in Fig. 2(b), u_c and α_c are the velocity and the attenuation coefficients of sound in the c phase, respectively, and $\tau_{t \rightarrow c}$ is the transmission coefficient from the transducer to the c phase.

The transmission coefficient $\tau_{c \rightarrow d}$ is, in general, a complex-valued quantity with the modulus and argument

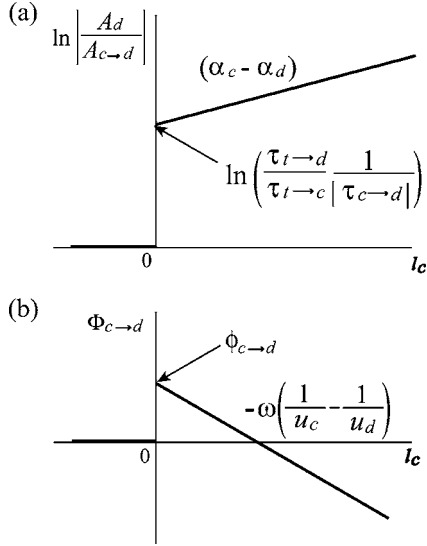


FIG. 3. Schematic illustrations for Eq. (14) in (a) and for Eq. (15) in (b). Here l_c is the thickness of the c phase under the transmitter transducer. The region $l_c < 0$ means that the interface is above the upper transducer.

$$\tau_{c \rightarrow d} = |\tau_{c \rightarrow d}| \exp[i\phi_{c \rightarrow d}]. \quad (12)$$

With Eqs. (9), (11), and (12), we obtain

$$\frac{A_d}{A_{c \rightarrow d}} = \left| \frac{A_d}{A_{c \rightarrow d}} \right| \exp[i\Phi_{c \rightarrow d}], \quad (13)$$

where

$$\ln \left| \frac{A_d}{A_{c \rightarrow d}} \right| = \ln \left(\frac{\tau_{t \rightarrow d}}{\tau_{t \rightarrow c}} \frac{1}{|\tau_{c \rightarrow d}|} \right) + (\alpha_c - \alpha_d) l_c \quad (14)$$

and

$$\Phi_{c \rightarrow d} = \phi_{c \rightarrow d} - \omega l_c \left(\frac{1}{u_c} - \frac{1}{u_d} \right). \quad (15)$$

Equations (14) and (15) are schematically illustrated in Figs. 3(a) and 3(b), where $\ln|A_d/A_{c \rightarrow d}|$ and $\Phi_{c \rightarrow d}$ are plotted as a function of l_c , respectively. As can be seen from Fig. 3(a), the magnitude of the transmission coefficient $|\tau_{c \rightarrow d}|$ is obtained from the intercept of the straight line of slope $(\alpha_c - \alpha_d)$ with the ordinate $l_c = 0$. Thus, the contribution from the interface and that from the bulk liquids become nicely separated.

From Eq. (14), one may recognize another important advantageous aspect of the present method; namely, signal A_d is always utilized as a reference to $A_{c \rightarrow d}$ for obtaining $|\tau_{c \rightarrow d}|$. This provides a critical check for our measurement as a whole. In principle, the information on the interface dynamics can also be obtained by measuring the reflection of sound from the interface. In that case, however, one does not have such reference quantity.

B. Experimental setup

The principal part of our cryostat is schematically shown in Fig. 1. The two sound cells are fastened to a copper

nuclear stage which is almost the same as in Refs. 4 and 19. The heat exchanger is about 14 g of sintered silver powder in each cell. The effective volume of each cell for the bulk liquids is about 10 cm^3 . Each cell contains crystal transducers in pairs separated with a sapphire spacer of $L \sim 5 \text{ mm}$ in thickness. The parallelism between the spacer's faces is better than 2×10^{-4} . The transducers in cell 1 are quartz of the fundamental frequency around 10 MHz, and those in cell 2 are LiNbO_3 of around 15 MHz. The nominal diameter of the active electrode of each transducer is about 4 mm.

The thermometry is fulfilled with a ^3He melting-curve thermometer (MCT), a Pt-NMR thermometer, and a commercial RuO_2 thermometer (calibrated down to 50 mK; SI, Inc.). The thermometers are attached to a thick copper plate fastened tightly to the nuclear stage. Carbon resistance thermometers (Matsushita 47 Ω and 100 Ω) are put inside of the cells for monitoring stabilization of the temperature in the liquids during measurements. The final check of our thermometry is made by observing the superfluid transition of the phase-separated c phase with the NMR measurement in the similar way as described in Ref. 19. For this purpose, the tube connecting the cells, Cu-Ni tube of 1.8 mm i.d., is crossed at the middle part with a Stycast-1266 tube around which the ^3He -NMR pickup coil is wound. Using the Greywall's temperature scale for the MCT and the superfluid transition temperature of ^3He ,²⁰ we find that the accuracy of our thermometry keeps within 3% in the whole range of the temperature presently studied.

The pressure of the liquids in the cell is measured with a Straty-Adams-type strain gauge.²¹ Its construction can be found in Ref. 22. The cold valves in the ^4He superleak lines are used to avoid any pressure disturbance in the course of the transfer of liquid helium every 4 days.

C. Procedure and arguments on the accuracy

A block diagram of our sound measuring system is shown in Fig. 4. We apply the method of the two-channel phase-sensitive detection²³ (PSD) with a pulse-modulated rf voltage controlled by a computer. This enables us to obtain simultaneously two components, cosine and sine, of the sound signal. The amplitude and phase of the signal are calculated by using these both components. Then, we obtain the attenuation coefficient and the change of velocity.

The experiment is performed in sequence from (i) to (v). The arguments are given in (vi), (vii), and (viii) on the basic numerical values of various physical quantities necessary to obtain the growth coefficient ξ from the transmitted sound signal.

(i) We condense pure ^4He through the superleak lines. The transmission of sound in each cell is measured within the temperature range from about 30 mK to 1 K under the saturated vapor pressure (svp). The observed temperature dependence of the sound velocity, $u_4(T, P = \text{svp}, 9.64 \text{ MHz})$, is in a good agreement with the data published in Ref. 24 at 12 MHz by taking the absolute value at zero temperature as $u_4(T=0, P=0) = 238.3 \text{ m/s}$ from Ref. 25. Thus, the resolution of our system for the phase variation is such that we can detect a relative change of the sound velocity within limits of about 10^{-5} .

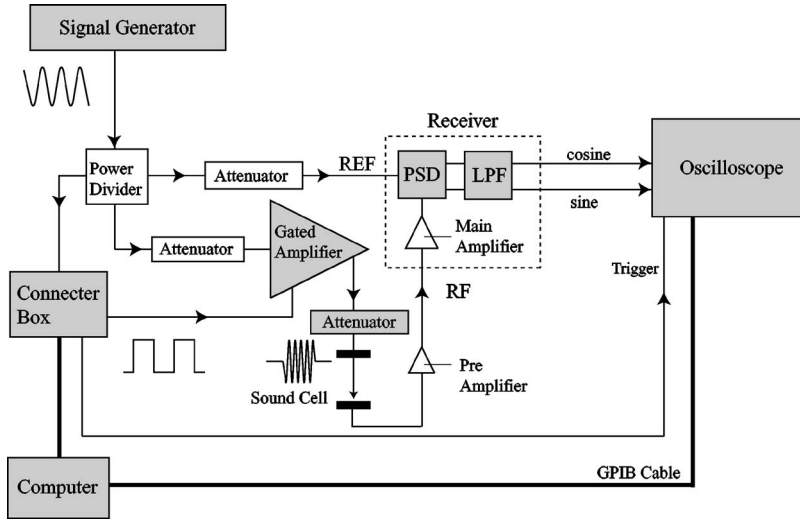


FIG. 4. Block diagram of our sound measuring system.

Consider the same situation as in Fig. 2(a) with pure liquid ^4He instead of a ^3He - ^4He mixture. The rf voltage from the receiver transducer, A_4 , is expressed as

$$A_4 = \frac{a_{up}(\omega)}{a_{low}(\omega)} A_i \tau_{t \rightarrow d} \tau_{d \rightarrow t} \exp[-\alpha_4 L] \exp[-i\omega L/u_4]. \quad (16)$$

This corresponds to Eq. (9) for the d phase in which subscript 4 is substituted by d . Within the resolution of our measuring system as a whole with the present spacer of the thickness $L \sim 5$ mm, we can detect any significant dependence of the signal on temperature only above about 150 mK because the lower limit of our attenuation measurement is about 0.01 Np/cm. Employing analogous arguments made in Ref. 24 as to how to determine the absolute value of α_4 , we may safely assume a sufficient smallness of α_4 at ~ 30 mK in order to have $\alpha_4(T \sim 30 \text{ mK}, P \sim 1 \text{ bar}, \omega)L = 0$. Accordingly, we can put

$$\exp[-\alpha_4(T \sim 30 \text{ mK}, P \sim 1 \text{ bar}, \omega)L] = 1. \quad (17)$$

This enables us to determine the factor

$$\frac{a_{up}(\omega)}{a_{low}(\omega)} \tau_{t \rightarrow d} \tau_{d \rightarrow t} \quad (18)$$

in Eq. (16) since A_i is known and A_4 is measured at ~ 30 mK.

As is described in Sec. II A, the measurement of the sound transmission through the saturated d phase is crucial for the present investigation. In order to obtain the absolute value of α_d with Eq. (9), it is necessary to fix the magnitude of a factor

$$\frac{a_{up}(\omega)}{a_{low}(\omega)} \tau_{t \rightarrow d} \tau_{d \rightarrow t}. \quad (19)$$

For this purpose, we use the magnitude determined in Eq. (18) since it is reasonable to assume within the satisfactory accuracy

$$\tau_{t \rightarrow d} = \tau_{t \rightarrow 4} \quad \text{and} \quad \tau_{d \rightarrow t} = \tau_{4 \rightarrow t}, \quad (20)$$

This is due to the weak dependence of the sound velocity on the ^3He concentration and the large difference of the sound velocities in helium and transducer material.

(ii) When the saturated d phase is entirely between the transducers as in Fig. 2(a), we make various checks to determine the width and amplitude of the pulse-modulated rf voltage applied to the transmitter transducer, τ_{pulse} and $|A_i|$, appropriate for the present experiment. First, the amplitude of the receiver signal $|A_d|$ is plotted as a function of τ_{pulse} with the various constant values of $|A_i|$ larger than 4 mV. It is found that $|A_d|$ does not show any appreciable dependence on τ_{pulse} when τ_{pulse} exceeds 6 μs . So, in the present study, we always use the pulse width of $\tau_{pulse} = 7 \mu\text{s}$.

Then, the heating effect due to the input of the sound power is checked for this pulse width at various constant temperatures above ~ 14 mK. The repetition rate is always one pulse per second. It is found that we have no detectable heating effect when $|A_i|$ does not exceed ~ 3 V. So, in the present study, the magnitude $|A_i|$ is always between 4 mV and 1 V.

The restraints mentioned above restrict the upper limit of the attenuation coefficient measurable with the present system.

(iii) As the attenuation coefficient varies with the temperature, pressure, and frequency, the magnitude $|A_i|$ must be changed according to the situation. So the measuring system must have a good linear behavior in the wide ranges. Such checks are made for various attenuators and amplifiers.

(iv) For the present aim to obtain the transmission coefficient with high accuracy, it was very important to realize the condition of the normal incidence of sound onto the interface [see Eq. (6)]. The cryostat is mounted on air springs which act not only to reduce the effect from the external vibrations but also to have a good horizontal positioning of the interface. That positioning is carefully controlled, and a satisfactory normal incidence is achieved within about 0.04° . It is monitored continuously by using digital gauges combined with display units (Sony Precision Technology, Inc.) to keep almost the same condition during the whole experiment.

(v) After finishing all the procedures (i)–(iv), the measurement is performed by starting from the situation of Fig. 2(a) and then by moving the interface as shown in Fig. 2(b). We stop the motion at the position of the interface far enough below the upper transducer, usually within the range about 0.1–2 mm, depending on the magnitude of the attenuation coefficient in the c phase. Then, the process is reversed—i.e., from Fig. 2(b) to Fig. 2(a). The velocity of the interface is about 1 $\mu\text{m/s}$ throughout the present experiment.

(vi) As will be discussed minutely in Sec. III B, the value of l_c is determined with Eq. (15). For this purpose, it is necessary to have the values of $u_c(T, P)$ and $u_d(T, P)$.

Within our accuracy it is sufficient to replace u_d with u_4 as a difference between them is negligible; i.e., the fractional variation in the sound velocity is smaller than $\sim 5 \times 10^{-4}$ within the temperature range of the present experiment.^{26,27} The evaluation of $u_d(T, P)$ can be made with the help of Ref. 28, according to which we can neglect the temperature variation of sound velocity and take the value

$$u_d(T, P = 1 \text{ bar}) = 246 \text{ m/s} \quad (21)$$

for the experiments performed at the pressure around 1 bar.

As the c phase below about 100 mK represents almost pure ^3He , we can safely replace $u_c(T, P)$ with $u_3(T, P)$, the velocity of first sound in liquid ^3He . Referring to the experimental and calculated results in Refs. 29 and 30, we also neglect the temperature dependence of sound velocity and take the value

$$u_c(T, P = 1 \text{ bar}) = 201 \text{ m/s} \quad (22)$$

throughout the present study made at a pressure of around 1 bar.

(vii) In order to obtain the growth coefficient ξ from the transmission coefficient $\tau_{c \rightarrow d}$ of Eq. (7), it is necessary to know the values of acoustic impedance,

$$Z_c(T, P) = \rho_c(T, P)u_c(T, P), \quad Z_d(T, P) = \rho_d(T, P)u_d(T, P). \quad (23)$$

Here, ρ_c and ρ_d are the density of the coexisting c and d phases, respectively. The value of $\rho_c(T, P)$ can be replaced with $\rho_3(T, P)$, the density of pure liquid ^3He ,²⁹ as is mentioned above. The value of $\rho_d(T, P, x_s)$ is calculated straightforwardly since we have an empirical expression for the molar volume of the d phase, $V_d(T, P, x)$, as will be mentioned in Sec. III A with Eq. (25).

(viii) Since the c phase in the present temperature range is almost pure liquid ^3He , we expect the crossover of a propagating sound between the hydrodynamic first-sound and zero-sound modes. The acoustic impedance for the zero-sound mode is discussed in Ref. 31 by defining the generalized acoustic impedance as a ratio of the momentum flux to the mass flux and density. Then, one may have $Z_c = \rho_c u_{0c}$ for the longitudinal zero sound to a fairly good approximation. Here, u_{0c} is the zero-sound velocity in the c phase. Thus, we can use the expression for the sound transmission coefficient as Eqs. (7) and (8) regardless of the type of sound modes.

The difference of sound velocities between u_c and u_{0c} is estimated with the formula given for pure liquid ^3He .³² We

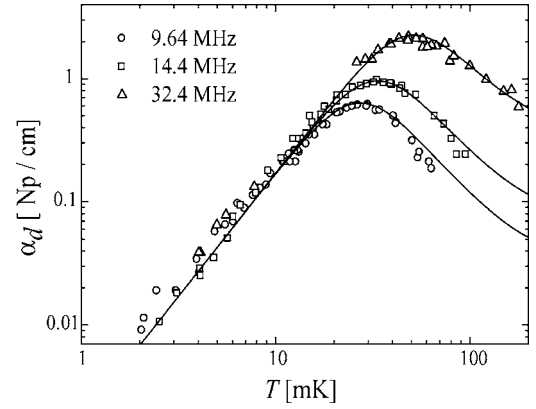


FIG. 5. The attenuation coefficient of the first-sound mode in the saturated d phase as a function of the temperature at a pressure of ~ 1 bar. The solid curves are the fitting with Eq. (24), $\alpha_d(T, P = 1 \text{ bar}, x_s, \omega)$, in which the fitting parameter τ_d is taken according to Eq. (27).

have the estimation as $(u_{0c} - u_c)/u_c \approx 0.03$ for the pressure $P = 1$ bar. In the following, we neglect this small difference and use the value of Eq. (22) for the sound velocity in the c phase at a pressure of around 1 bar throughout the present whole temperature range.

The transmission coefficient through the interface can also be obtained by transmitting the sound from the lower transducer to the upper one. This yields $\tau_{d \rightarrow c}$. We made such measurements and confirmed that both $\tau_{c \rightarrow d}$ and $\tau_{d \rightarrow c}$ give the same result for ξ as it should. See, for example, Ref. 12. In this paper, we present results based mainly on the data of $\tau_{c \rightarrow d}$.

III. RESULTS FOR α_d , α_c , AND $\tau_{c \rightarrow d}$

We have performed experiments with sound of 9.64, 14.4, and 32.4 MHz. The absolute value of the attenuation coefficient in the saturated d phase is determined from Eq. (9) with the procedure described in Sec. II C. In addition to the measurements performed at every beginning of each run in order to displace the position of the interface at constant temperature, the measurement is also made by sweeping the temperature when the interface is always kept far above the upper transducer. A comparison between the results of two measurements provides a convincing check of our measurements.

A. Results for α_d

The results under a constant pressure of ~ 1 bar are shown in Fig. 5 in which α_d is plotted as a function of temperature for the three frequencies studied. As can be seen from the figure, α_d shows a maximum at the temperature $T_{d,m}(\omega)$ which depends on the frequency. Below $T_{d,m}(\omega)$, α_d decreases with the temperature in proportion to T^2 and the frequency dependence disappears.

The attenuation of first sound in the d phase was theoretically discussed in Ref. 33. The theoretical expression for the

attenuation coefficient of the d phase of ^3He molar concentration x is given as²⁷

$$\alpha_d(T, P, x, \omega) = \frac{2}{3} \frac{P_F(T, P, x) m_4}{n_4(T, P, x) m^*(P) u_d^2(T, P, x) m^*(P) u_d(T, P, x)} \times \left(1 + \alpha_{BBP}(T, P, x) + \frac{m^*(P) - m_3}{m_4} \right)^2 \times \frac{\omega^2 \tau_d(T, P, x)}{1 + \omega^2 \tau_d^2(T, P, x)}. \quad (24)$$

Here, P_F is usually called the Fermi pressure,³⁴ which can be calculated as accurately as one wants, and $m^* = m^*(P)$ is the effective mass of ^3He quasiparticles in the dilute limit $x \rightarrow 0$. The quantities m_3 and m_4 are the bare mass of ^3He and ^4He , respectively. The parameter α_{BBP} is usually called the Bardeen-Baym-Pines (BBP) parameter³⁵ whose meaning is to relate the molar volumes of the d phase $V_d(T, P, x)$ and pure ^4He phase $V_{40}(T, P)$ as

$$V_d(T, P, x) = V_{40}(T, P)[1 + \alpha_{BBP}(T, P, x)x]. \quad (25)$$

With Avogadro's number N_A , the number density of ^4He , n_4 , is expressed as

$$n_4 = n - n_3 \text{ with } n \equiv N_A/V_d \text{ and } n_3 = nx. \quad (26)$$

The empirical formulas for $\alpha_{BBP}(T, P, x)$, $m^*(P)$, and the saturated ^3He concentration $x_s(T, P)$ can be found in Ref. 36. That of $V_{40}(T, P)$ is given in Ref. 22. For $u_d(T, P = 1 \text{ bar}, x_s)$, we use the value of Eq. (21). The time τ_d , which appears in the expression for first viscosity, has a meaning of the relaxation time for ^3He quasiparticles in the d phase due to ^3He - ^3He collisions.

Then, the numerical calculation of the attenuation coefficient in the saturated d phase, $\alpha_d(T, P, x_s(T, P), \omega)$, can be made straightforwardly with Eq. (24) in which we take τ_d as a fitting parameter, neglecting its dependence on $x = x_s(T, P)$. This assumption can be justified because a relative change of $x_s(T, 1 \text{ bar})$ within the temperature range 0–100 mK is about 9%.³⁶ The results at the pressure $P = 1 \text{ bar}$ are shown in Fig. 5 with the solid curves for which the fitting parameter τ_d is taken as

$$\tau_d(T, P = 1 \text{ bar}, x_s) T^2 = 12 \times 10^{-12} [\text{s K}^2]. \quad (27)$$

As can be seen from Fig. 5, the overall agreement is quite good. Furthermore, the magnitude of $\tau_d T^2$ in Eq. (27) determined from the best fitting seems reasonable compared with the data published—for example,

$$\begin{aligned} \tau_d(T, P \sim 0, x = 0.050) T^2 &= 24 \times 10^{-12} [\text{s K}^2] \quad (\text{Ref. 37}) \\ &17.8 \times 10^{-12} [\text{s K}^2] \quad (\text{Ref. 38}), \end{aligned}$$

$$\begin{aligned} \tau_d(T, P \sim 0, x = 0.013) T^2 &= 20 \times 10^{-12} [\text{s K}^2] \\ &15.0 \times 10^{-12} [\text{s K}^2], \end{aligned} \quad (28)$$

(Refs. 37 and 38) and

$$\tau_d(T, P \sim 0, x = 0.050) T^2 = 20 \times 10^{-12} [\text{s K}^2]. \quad (29)$$

(Ref. 27). We believe that such reasonable agreements support the accuracy of our measurements as a whole.

The existence of $T_{d,m}(\omega)$ in the α_d vs T plot has been reported by several groups.^{26,39–41} We believe, however, that the present data given in Fig. 5 are the first which convincingly show the T^2 dependence of α_d in a wide temperature range below $T_{d,m}(\omega)$ —i.e., in the region $\omega \tau_d \gg 1$.

B. Results for α_c and $\tau_{c \rightarrow d}$

The transmission coefficient $\tau_{c \rightarrow d}$ is obtained from Eqs. (12)–(15) with the procedures described in Secs. II A and II C. The examples of actual data are shown in Figs. 6 and 7. In each figure, the magnitudes of $\ln|A_d/A_{c \rightarrow d}|$ and $\Phi_{c \rightarrow d}$ are plotted in (a) and (b), respectively, as a function of time t , during which the position of the interface is moved with an almost constant velocity. In the plot of $\Phi_{c \rightarrow d}$, we take the origin of the axis, $\Phi_{c \rightarrow d} = 0$, for the receiver signal obtained when we have only the saturated d phase between the transducers [see Fig. 2(a)]. As can be seen from Figs. 6(a) and 7(a), in the region between $\sim t_1$ and $\sim t_2$, $\ln|A_d/A_{c \rightarrow d}|$ shows a curious deviation from the linear t dependence observed for $t > t_2$ or from a zero value for $t < t_1$. Similar behavior is also seen when the interface movement is reversed. We believe that the appearance of such behavior may be due to some complicated motion of the interface when it starts leaving or touching the transducer surface. In particular, a nonuniform and nonsimultaneous break of the interface from the transducer surface is very likely.

On the other hand, as can be seen from Figs. 6(b) and 7(b), $\Phi_{c \rightarrow d}$ approaches a zero value rather smoothly. So we may safely take

$$\phi_{c \rightarrow d} = 0 \quad (30)$$

in Eq. (15). That is, the transmission coefficient $\tau_{c \rightarrow d}$ proves to be a real quantity [see Eq. (12)]. Then, Eq. (15) is rewritten as

$$\Phi_{c \rightarrow d} = -\omega l_c \left(\frac{1}{u_c} - \frac{1}{u_d} \right). \quad (31)$$

Thus, we can obtain the value of $l_c(t)$ from that of $\Phi_{c \rightarrow d}(t)$.

We fix the origin of the l_c axis as illustrated in Figs. 6(b) and 7(b). First, we estimate the time $t = t_0$ at which the interface would arrive at the transducer surface as if there were no interaction between the interface and transducer surface mentioned above; i.e., we assume that the interface continues its steady motion for $t < t_2$. So, as is shown in Figs. 6(b) and 7(b), t_0 is fixed as the time moment corresponding to the intersection point of the horizontal line $\Phi_{c \rightarrow d} = 0$ with the extrapolated straight line of $\Phi_{c \rightarrow d}(t)$ obtained by a fit in the region $t > t_2$. Then, we take $l_c(t_0) = 0$. From the figures, it is recognized that t_0 thus fixed nearly coincides with t_2 .

Then, we can convert Figs. 6(a) and 7(a) into the figures in which $\ln|A_d/A_{c \rightarrow d}|$ is plotted versus l_c , as is shown in Figs. 6(c) and 7(c), respectively. The value of $l_c(t)$ in the region $t_1 \leq t \leq t_2$ is obtained according to the extrapolated lines of $\Phi_{c \rightarrow d}(t)$ mentioned above; i.e., in the region $t_1 \leq t \leq t_0$, we

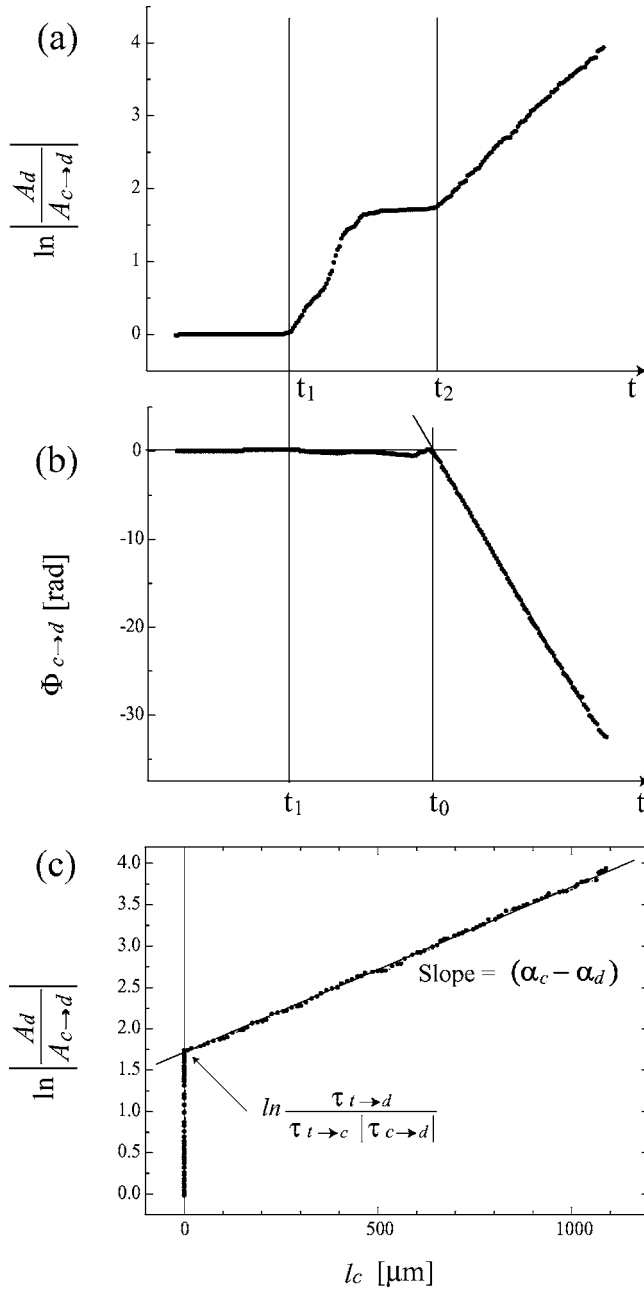


FIG. 6. Example of the actual data taken at $T \approx 5.5$ mK under a pressure of ~ 1 bar with the sound of 9.64 MHz. (a) Logarithm of amplitude ratio $\ln|A_d/A_{c \rightarrow d}|$ is plotted as a function of time, t . (b) The phase change $\Phi_{c \rightarrow d}$ is plotted as a function of time t . The time t_0 is determined from the intersection point between the fitted t -linear line and the horizontal line $\Phi_{c \rightarrow d} = 0$. (c) Logarithm of amplitude ratio $\ln|A_d/A_{c \rightarrow d}|$ shown in (a) is plotted as a function of the c -phase thickness l_c .

take $l_c(t) = 0$. Note that plots such as Figs. 6(c) and 7(c) can be meaningful only in the region $l_c \geq 0$ since we have no actual experimental information on the interface position above the upper transducer, which is formally related to the region $l_c < 0$ as illustrated in Fig. 3. One may recognize that the characteristic features of Figs. 6(c) and 7(c) resemble the schematic illustration given in Fig. 3(a) with the exception of the curious behavior around $l_c = 0$. One can see from Figs.

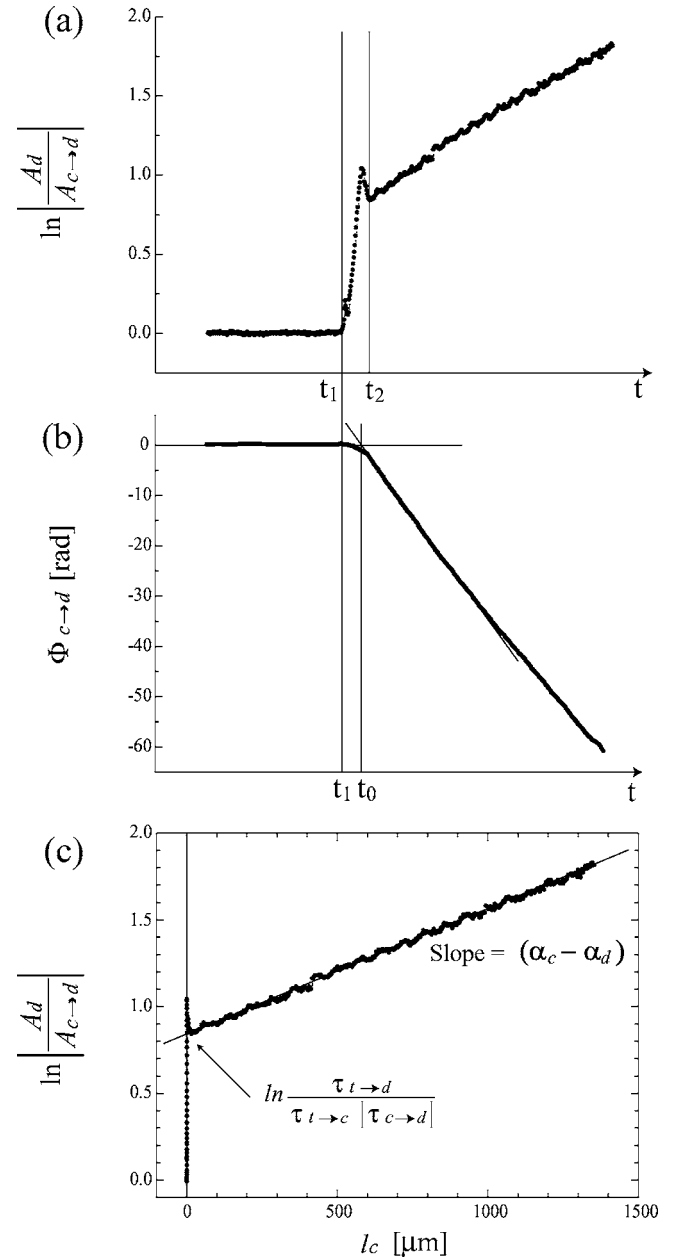


FIG. 7. Same as Fig. 6 for the sound of 14.4 MHz at $T \approx 36$ mK under a pressure of ~ 1 bar.

6(c) and 7(c) that such curious behavior of $\ln|A_d/A_{c \rightarrow d}|$ has almost negligible effect for determining the intersection of the straight line continued from the region $l_c > 0$ with the ordinate $l_c = 0$.

As is noted for Eq. (14) and Fig. 3(a), the slope of the straight line fitted for the region $l_c > 0$ in Figs. 6(c) and 7(c) gives the value of $(\alpha_c - \alpha_d)$. Since we already have the data of α_d as is described in the preceding section, we can determine α_c from the slope. Thus the obtained $\alpha_c(T, P \sim 1 \text{ bar}, \omega)$ is shown in Fig. 8 as a function of temperature. As can be seen from the figure, the results for the c -phase attenuation coefficient may be summarized as follows:

(i) α_c shows a maximum at the temperature $T_{c,m}(\omega)$ which depends on the frequency.

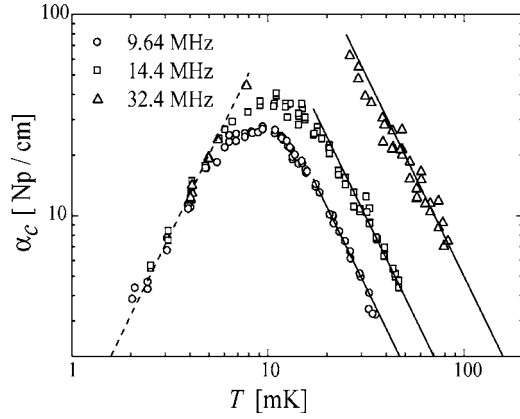


FIG. 8. The sound attenuation coefficient in the saturated c phase as a function of the temperature at a pressure of ~ 1 bar. The values of $\alpha_c(T, P \sim 1 \text{ bar}, \omega)$ are obtained from the slope of the fitting line as is described in the text for Eq. (14) and Figs. 3(a), 6(c), and 7(c). The solid lines represent Eq. (32) with $B_1(P \sim 1 \text{ bar}) = 1.2 \times 10^{-18} [\text{s}^2 \text{K}^2 \text{Np cm}^{-1}]$. The dotted line expresses Eq. (33) with $B_0(P \sim 1 \text{ bar}) = 0.8 \times 10^6 [\text{K}^{-2} \text{Np cm}^{-1}]$.

(ii) Above $T_{c,m}(\omega)$, α_c decreases with the increase of the temperature as

$$\alpha_c(T, P \sim 1 \text{ bar}, \omega) = B_1 \omega^2 / T^2. \quad (32)$$

(iii) Below $T_{c,m}(\omega)$, α_c diminishes as the temperature decreases,

$$\alpha_c(T, P \sim 1 \text{ bar}, \omega) = B_0 T^2, \quad (33)$$

and the frequency dependence vanishes.

One may easily recognize that the features observed are the same as those reported for the sound attenuation coefficient in pure liquid ^3He .⁴² Such an observation is quite reasonable as the c phase below about 100 mK represents in essence pure liquid ^3He . Under simplest assumptions, $T_{c,m}(\omega)$ may correspond to

$$\omega \tau_c(T = T_{c,m}(\omega), P \sim 1 \text{ bar}) = 1, \quad (34)$$

where $\tau_c(T, P)$ is the relaxation time of ^3He quasiparticles in the c phase and can be identified with that appearing in the first viscosity coefficient. With the fitting lines plotted in Fig. 8, we obtain

$$\tau_c(T, P \sim 1 \text{ bar}) T^2 = 1.22 \times 10^{-12} [\text{s K}^2]. \quad (35)$$

As is mentioned in Sec. II C (viii), we always use the value u_c given by Eq. (22) throughout the whole temperature range of the present experiment performed under a pressure of ~ 1 bar in the procedure for determining the value l_c with Eq. (31) in order to obtain the results shown in Figs. 6(c) and 7(c); i.e., we do not distinguish a small difference in velocities of the first- and zero-sound modes.

The magnitude of $\tau_c T^2$, thus obtained, seems reasonable as compared with those for pure liquid ^3He derived from the sound attenuation⁴² and those from the viscosity which give the relaxation time $\tau_{\eta,3}$.⁴³ After Ref. 42, above $T_{3,m}(\omega)$ in the region of the first-sound mode, one has

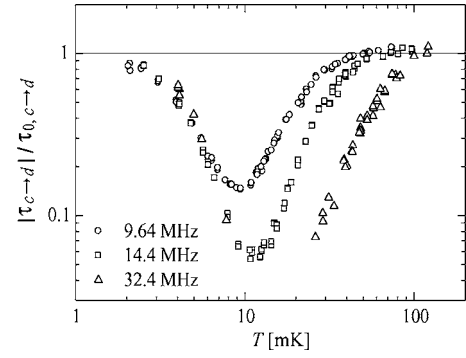


FIG. 9. The sound transmission coefficient through the interface, $|\tau_{c \rightarrow d}(T, P \sim 1 \text{ bar}, \omega)|$, versus temperature. Note that the data are normalized by the standard acoustic-mismatch coefficient $\tau_{0,c \rightarrow d}(T, P = 1 \text{ bar})$.

$$\tau_1(T, P = 0.32 \text{ atm}) T^2 = 1.5 \times 10^{-12} [\text{s K}^2], \quad (36)$$

and below $T_{3,m}(\omega)$ —i.e., in the region of the zero-sound mode—one obtains

$$\tau_0(T, P = 0.32 \text{ atm}) T^2 = 1.1 \times 10^{-12} [\text{s K}^2]. \quad (37)$$

In general, τ_1 and τ_0 are different. However, we neglect this difference, taking some average, since this difference is small and the use of single time τ_c instead of two ones should not give large errors. From Ref. 43 for the viscosity data, one finds

$$\tau_{\eta,3}(T, P = 0) T^2 = 1.24 \times 10^{-12} [\text{s K}^2],$$

$$\tau_{\eta,3}(T, P = 3 \text{ bar}) T^2 = 1.15 \times 10^{-12} [\text{s K}^2]. \quad (38)$$

We believe that such reasonable agreement shows well the reliableness of the experimental method presently developed.

To obtain value $|\tau_{c \rightarrow d}|$ with Eq. (14), it is necessary to fix the ratio $\tau_{t \rightarrow d} / \tau_{t \rightarrow c}$. Although $\tau_{c \rightarrow d}$ is treated like a real quantity according to Eqs. (30) and (31), we keep the notation of the absolute value for convenience of referring to Eq. (14). This ratio can be written as

$$\frac{\tau_{t \rightarrow d}}{\tau_{t \rightarrow c}} = \left(\frac{2Z_d}{Z_t + Z_d} \right) \times \left(\frac{2Z_c}{Z_t + Z_c} \right)^{-1}, \quad (39)$$

where Z_t is the acoustic impedance of the transducer. As $Z_t \gg Z_c, Z_d$, Eq. (39) is evaluated as

$$\frac{\tau_{t \rightarrow d}}{\tau_{t \rightarrow c}} \simeq \frac{Z_d}{Z_c} = \frac{\rho_d u_d}{\rho_c u_c} \simeq 2.08 \text{ at } P = 1 \text{ bar}. \quad (40)$$

Here we used approximations (vi) and (vii) discussed in Sec. II C.

The transmission coefficient of the interface, thus determined at pressure ~ 1 bar, $|\tau_{c \rightarrow d}(T, P \sim 1 \text{ bar}, \omega)|$, is plotted in Fig. 9 as a function of temperature. In the figure, $|\tau_{c \rightarrow d}|$ is normalized by the usual acoustic-mismatch coefficient $\tau_{0,c \rightarrow d}$ in order to represent distinctly the effect of the phase conversion. We emphasize that $|\tau_{c \rightarrow d}| / \tau_{0,c \rightarrow d} < 1$ means an existence of the phase conversion induced by the sound pressure wave. The value of $\tau_{0,c \rightarrow d}(T, 1 \text{ bar}) \simeq 1.35$ is obtained with the same approximations used for Eq. (40).

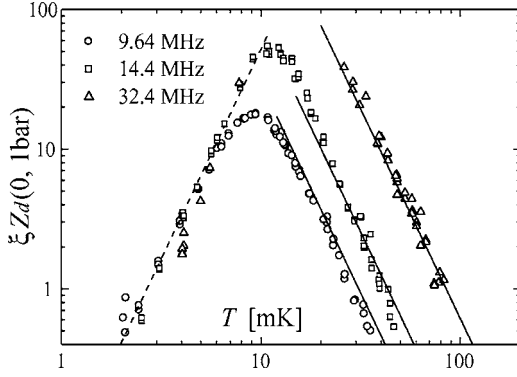


FIG. 10. The temperature dependence of kinetic growth coefficient $\xi(T, P \sim 1 \text{ bar}, \omega)$. The data are shown in the dimensionless units of Eq. (41). The solid lines represent Eq. (42) with $A_1(P \sim 1 \text{ bar}) = 1.04 \times 10^{-24} [\text{s}^{5/2} \text{K}^3]$. The dotted line expresses Eq. (43) with $A_0(P \sim 1 \text{ bar}) = 5.2 \times 10^7 [\text{K}^{-3}]$.

From Fig. 9, the most interesting observations are that $|\tau_{c \rightarrow d}|/\tau_{0, c \rightarrow d}$ has a frequency-dependent minimum at some temperature $T_{\tau, \min}(\omega)$ and that the reduction at this point is almost a factor of 10 or much more depending on the frequency. In the following section, we will derive the kinetic growth coefficient of the interface from the data shown in Fig. 9 and give various discussions on the results.

IV. RESULTS FOR $\xi(T, P, \omega)$ AND DISCUSSIONS

A. Results for $\xi(T, P \sim 1 \text{ bar}, \omega)$

From the data of $|\tau_{c \rightarrow d}(T, P \sim 1 \text{ bar}, \omega)|$ described in the preceding section, we derive the kinetic growth coefficient of the interface, $\xi(T, P \sim 1 \text{ bar}, \omega)$, by using Eq. (7). For this derivation, we use the numerical values of Eq. (40) for Z_d/Z_c and $Z_d(T, 1 \text{ bar}) = Z_d(0, 1 \text{ bar}) = 3.54 \times 10^3 \text{ g cm}^{-2} \text{ s}^{-1}$. The results are shown in Fig. 10, in which, for convenient comparison, the dimensionless quantity

$$\xi(T, P \sim 1 \text{ bar}, \omega) \times Z_d(0, 1 \text{ bar}) \quad (41)$$

is plotted as a function of temperature. We take $Z_d(0, 1 \text{ bar})$ as a typical magnitude for the acoustic impedance of the present system at $\sim 1 \text{ bar}$. As can be recognized from Eq. (7), the inaccuracy of the value of ξ thus derived is enhanced as $|\tau_{c \rightarrow d}|/\tau_{0, c \rightarrow d}$ approaches unity. So, in Fig. 10, the plot is shown for ξZ_d down to ~ 0.5 .

As can be seen from the figure, ξ shows a maximum at the temperature $T_m(\omega)$ corresponding to $T_{\tau, \min}(\omega)$ in Fig. 9—i.e., at $\approx 9 \text{ mK}$ for 9.64 MHz, $\approx 10 \text{ mK}$ for 14.4 MHz, and $\approx 15 \text{ mK}$ for 32.4 MHz. The most prominent features are as follows.

(i) Above $T_m(\omega)$, the growth coefficient is frequency dependent and falls with an increase of the temperature as

$$\xi(T, P \sim 1 \text{ bar}, \omega) Z_d(0, 1 \text{ bar}) = A_1 \omega^{5/2} T^{-3}. \quad (42)$$

(ii) Below $T_m(\omega)$, the growth coefficient becomes frequency independent and also diminishes as the temperature is lowered:

$$\xi(T, P \sim 1 \text{ bar}, \omega) Z_d(0, 1 \text{ bar}) = A_0 T^3. \quad (43)$$

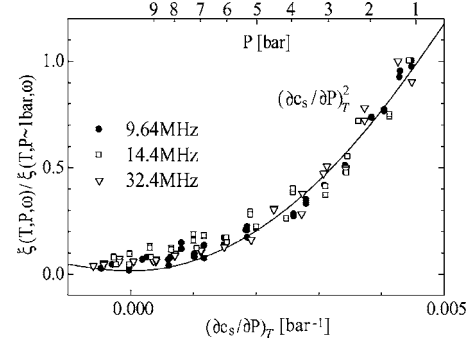


FIG. 11. The dependence of ξ on the phase-separation pressure at the fixed temperature $\sim 30 \text{ mK}$ for a pressure above $\sim 1 \text{ bar}$. The data are reduced to the growth coefficient at $T \sim 30 \text{ mK}$ and $P \sim 1 \text{ bar}$ and given as a function of concentration slope $(\partial c_s / \partial P)_T$. The solid curve is the fitted one proportional to $(\partial c_s / \partial P)_T^2$. For convenience in recognizing the pressure dependence of $(\partial c_s / \partial P)_T$, the corresponding phase-separation pressure P is given at the upper horizontal axis.

(iii) The values of $T_m(\omega)$ nearly coincide with those of $T_{c, m}(\omega)$ in Fig. 8 at the same frequency.

These characteristic features strongly suggest that we observe a crossover between the hydrodynamic and ballistic regimes for the kinetic growth coefficient determined from the sound transmission within the frequency range presently studied. In the following sections, we will discuss what kind of kinetic processes can be involved in the phase-conversion and crossover phenomena at the interface and how we can understand the observed dependence of ξ on the temperature and frequency. Before starting the discussion, we will present the pressure dependence of ξ at a fixed temperature.

B. Results for $\xi(T \sim 30 \text{ mK}, P, \omega)$

Besides the temperature dependence of $\tau_{c \rightarrow d}$ at a pressure of around 1 bar, we have measured the dependence of $\tau_{c \rightarrow d}$ on the phase-separation pressure $P(c_s, T = \text{const})$ at a fixed temperature of about 30 mK. Here, c denotes the mass concentration of ^3He in the d phase and c_s corresponds to x_s . From the data, we obtain the dependence of ξ on the slope $(\partial c_s / \partial P)_T$ shown in Fig. 11. Note that ξ increases in proportion to $(\partial c_s / \partial P)_T^2$. This fact unambiguously evidences that ξ determined with the present method really reflects the interface dynamics as a result of the phase conversion induced by the sound pressure. The empirical formula³⁶ of $x_s(T, P)$ gives $(\partial c_s(T, P) / \partial P)_T \approx dc_s(0, P) / dP$ within the present temperature range. That is, the observed dependence of ξ on the slope $(\partial c_s / \partial P)_T$ does not imply any extra temperature dependence of ξ in the present study.

C. Arguments on the interfacial mode

The existence of phase conversion between the adjacent phases may imply possible excitations of interfacial wave, which could affect the interface mobility. For the present system, such a wave may be called a phase-separation wave. The damping coefficient of the wave may be given as

$$\gamma = 2(\eta_c + \eta_d)\omega^{4/3}/(\rho_c + \rho_d)^{1/3}\sigma_i^{2/3}, \quad (44)$$

where σ_i is the interface-tension coefficient and η_c and η_d are the viscosity coefficient of the c and d phases, respectively. The characteristic features of the present system are that σ_i is so small as 0.02 erg/cm^2 and that within the present temperature range η_c is so large, as can be recognized from the magnitude of $\tau_c T^2$, Eq. (35). Furthermore, our sound frequencies are rather high. So, for the present experimental situation, we have $\gamma \gg \omega$; i.e., the interfacial wave is a highly damped mode. In addition, we have a good normal incidence of sound onto the interface. Furthermore, inequality $\omega\sigma_i/(u_1 u_2) \ll Z_{c,d}$ is well satisfied for the present system. Thus, we can neglect the terms with the interfacial tension in Eq. (6) even for large incidence angles. On the whole, we may conclude that the interfacial mode does not concern the present investigation on ξ .

D. Discussions on the region $T < T_m$

We start from the very striking T^3 dependence of ξ in the temperature region $T < T_m$. At such sufficiently low temperatures, as can be recognized from Figs. 5 and 8, the relaxation time of ^3He quasiparticles is much longer than $\tau_d(T_{d,m})$ and $\tau_c(T_{c,m})$. So we may consider the situation in which we have the ballistic flow of ^3He quasiparticles across the interface driven by the pressure modulation. For the coexisting c and d phases, we have a requirement of the equality of the chemical potentials as

$$\mu_{Fc} = \mu_{Fd}, \quad (45)$$

where μ_{Fc} and μ_{Fd} are the chemical potential per particle of ^3He in the c and d phases, respectively. On the other hand, we have a potential jump U in the Fermi energies of quasiparticles belonging to the different phases as

$$E_{Fc} = E_{Fd} + U. \quad (46)$$

Here E_{Fc} and E_{Fd} are the Fermi energy of the c and d phases, respectively. However, the presence of the potential step has almost negligible effect on the flow of ^3He quasiparticles with Fermi momenta since the quantum-mechanical transmission coefficient through such a potential does not much differ from unity.⁴⁴

As an illustration, we consider the flow from the c phase to the d phase. As usual for degenerate Fermi systems, it is reasonable to consider quasiparticles lying near the Fermi surface of each phase within the range of thermal broadening, $\pm k_B T$. Suppose that a quasiparticle of momentum $\mathbf{p}_{1,c}$ is driven to flow across the interface from the c phase to the d phase. The phase-conversion phenomenon for this quasiparticle means the process in which it collides with the d -phase quasiparticles and finally relaxes to a d -phase quasiparticle in thermal equilibrium with the surrounding bulk liquids. That is, in order to have phase conversion, we must have collisions among quasiparticles. So it seems very reasonable that ξ falls with a decrease of the temperature, since the lower the temperature, the lesser the frequency of collisions.

With the physical picture mentioned above, we try to understand the temperature dependence observed in Eq. (43). Consider the n -particle collision specified by

$$(\mathbf{p}_{1,c}) + (\mathbf{p}_{1,d}) + \cdots + (\mathbf{p}_{n-1,d}) \rightarrow (\mathbf{p}'_{1,d}) + \cdots + (\mathbf{p}'_{n,d}), \quad (47)$$

where subscript d means the d -phase quasiparticle. The law of momentum conservation is expressed as

$$\mathbf{p}_{1,c} + \sum_{i=1}^{n-1} \mathbf{p}_{i,d} = \sum_{i=1}^n \mathbf{p}'_{i,d}. \quad (48)$$

For the magnitude of the momentum, we may write

$$|\mathbf{p}_{1,c}| \simeq p_{Fc} \text{ and } |\mathbf{p}_{i,d}| \simeq |\mathbf{p}'_{i,d}| \simeq p_{Fd}. \quad (49)$$

As is noted in Sec. I with Eq. (1), there is a large difference between p_{Fc} and p_{Fd} . In such a situation, the two-particle collision cannot be effective for the phase-conversion process since it requires some definite angle of scattering to fulfill the conservation of momentum, Eq. (48), under the restriction of Eq. (49). Note that the law of energy conservation does not impose any significant restraints on the scheme of the collision process because of Eq. (46). It affects the temperature dependence of the relaxation time of ^3He quasiparticles. That is, due to the Pauli exclusion principle, the n -particle collision of Eq. (47) may give the relaxation time $\tau_{(n)}$ as⁴⁵

$$\tau_{(n)} \propto T^{-2(n-1)}, \quad (50)$$

when we consider quasiparticles near the Fermi surface within the range $\pm k_B T$.

Since the two-particle collision seems ineffective for the phase-conversion phenomena in the present system, we consider, as a natural step, the next-higher-order process—i.e., the three-particle collision which may not be strongly affected by the restrictions due to Eqs. (48) and (49). From Eq. (50) with $n=3$, we have

$$\tau_{(3)} \propto T^{-4}. \quad (51)$$

From the dimensional point of view, $\tau_{(3)}$ should be aE_F^3/T^4 . Here a is a quantity associated with the amplitude of the three-fermion interaction.

As is noted above, the d -phase quasiparticle converted from the incoming c phase one must finally be in thermal equilibrium with the surrounding bulk liquids for completion of the phase conversion. Such a process of thermal relaxation implies that the three-particle collision mentioned above is followed by subsequent two-particle collisions through the bulk liquids. This is the stage of thermal conduction through the bulk liquids and is governed by the magnitude of the thermal conductivity, κ . From the kinetic theory of gases, we have the order-of-magnitude estimate

$$\kappa \propto C v_F^2 \tau, \quad (52)$$

where C is the specific heat, v_F is the Fermi velocity, and τ is the relaxation time of the particle. For degenerate bulk Fermi liquids, we may take $\tau_{(2)}$ for τ in Eq. (52) and C is proportional to T . The same arguments can be applied for the opposite case in which a quasiparticle passes from the d phase to the c phase.

Therefore, we may conclude that the growth coefficient of the interface in the ballistic regime is, as a whole, proportional to

$$\tau_{(3)}^{-1} \kappa \propto T^4 T^{-1} = T^3. \quad (53)$$

This result agrees with the temperature dependence observed in Eq. (43).

E. Discussions on the region $T > T_m$

The transmission of sound across the superfluid-normal interface of ^3He - ^4He liquid mixtures is theoretically discussed in Ref. 17 within the framework of hydrodynamic approximation. Even in the region $T > T_m$, we may safely consider the c phase to be almost pure liquid ^3He within the temperature range presently studied. As is discussed in Ref. 17, in the d phase we have not only a first-sound mode but also a second-sound one. Furthermore, there is a diffusive mode of ^3He which will decay at the diffusion length defined as

$$\delta_D(\omega) = \sqrt{D/\omega}, \quad (54)$$

where D denotes the mass diffusion coefficient of ^3He atoms in the d phase. Then, the kinetic growth coefficient is given as¹⁷

$$\xi = \sqrt{u_{2d}^2 - i\omega D\rho} \frac{\partial \mathcal{Z}}{\partial c} \left(\frac{\partial c_s(T, P)}{\partial P} \right)_T, \quad (55)$$

where u_{2d} is the second-sound velocity in the d phase and \mathcal{Z} is the thermodynamic potential conjugated to the mass concentration c . Neglecting the entropy term, we have the expression for u_{2d} as⁴⁶

$$u_{2d}^2 = \frac{c^2 \rho_s}{\rho_n} \frac{\partial \mathcal{Z}}{\partial c}, \quad (56)$$

where ρ_s and ρ_n are the density of the superfluid and normal fluid components, respectively.

In the hydrodynamic limit $\omega\tau_d \ll 1$, the attenuation coefficient of second sound α_{2d} is given as⁴⁷

$$\alpha_{2d} = \frac{2}{3} \frac{\omega^2}{u_{2d}^3 \rho_n} (\eta_{3d} + \text{diffusion and heat conduction terms}). \quad (57)$$

Here, η_{3d} is the ^3He viscosity in the d phase. If we take the viscosity term alone, we can estimate the magnitude of α_{2d} from α_d of Eq. (24) with the approximation $\rho_n = n_3 m^*$ as

$$\alpha_{2d} \approx \alpha_d (u_d/u_{2d})^3 (n_4/n_3) (m^*/m_4). \quad (58)$$

For example, taking the values of $u_{2d}(100 \text{ mK}) \sim 13 \text{ m/s}$ from Ref. 48 and our $\alpha_d(100 \text{ mK}, 9.64 \text{ MHz}) \sim 0.1 \text{ Np/cm}$, we have $\alpha_{2d}(100 \text{ mK}, 9.64 \text{ MHz}) \sim 1.6 \times 10^4 \text{ Np/cm}$. The estimation of the magnitude of $\alpha_{2d}(T, 9.64 \text{ MHz})$ may also be made by using Eq. (57) and the experimental results in Ref. 48 performed at 25 kHz. For example, taking the value $\alpha_{2d}(100 \text{ mK}, 25 \text{ kHz}) \sim 0.3 \text{ Np/cm}$,⁴⁸ we have $\alpha_{2d}(100 \text{ mK}, 9.64 \text{ MHz}) \sim 5 \times 10^4 \text{ Np/cm}$.

Therefore, the frequencies of sound in the present study seem too high to consider the second-sound mode as a well-defined propagating mode. So we may safely neglect the term of u_{2d}^2 in the argument of square root of Eq. (55). Then, the dependence of ξ of Eq. (55) on the temperature and frequency may be given as

$$(\omega D)^{1/2} \propto (\omega \tau_{(2)})^{1/2} \propto \omega^{1/2} T^{-1}. \quad (59)$$

This does not agree with the observed result of Eq. (42). Furthermore, Eq. (55) gives the growth coefficient as a complex-valued quantity, which means a complex-valued transmission coefficient $\tau_{c \rightarrow d}$. Such behavior also does not agree with our observation (Sec. III B).

On the other hand, as is noted in Sec. IV B with Fig. 11, ξ behaves at a fixed temperature in proportion to $(\partial c_s / \partial P)_T^2$. This behavior is in accordance with Eq. (55). In this regard, though qualitatively and partly, the results shown in Fig. 11 may support the procedure of deriving Eq. (55). Note that the $(\partial c_s / \partial P)_T^2$ dependence in that equation results from the assumption of the phase equilibrium at the interface.¹⁷

We now try to find the reason of the disagreement between Eqs. (42) and (59) with respect to the dependence on the temperature and frequency. At first, without the sound injection, we have an equilibrium state at temperature T and the overall pressure $P = P_0$ as shown in Fig. 12(a). Then, suppose that an extra pressure δP is applied to the interface. The situation considered in Ref. 17 for this condition is illustrated in Fig. 12(b). As is mentioned above, it is assumed that one has the thermodynamic equilibrium at the interface; i.e., the ^3He concentration on the d -phase side changes from $x_s(T, P_0)$ to $x_s(T, P_0 + \delta P)$ only at the interface and decreases toward its magnitude in the bulk d -phase $x_s(T, P_0)$ with the characteristic length $\delta_D(\omega)$.

We think, however, that the real situation may be such as illustrated in Fig. 12(c); i.e., we have a finite region of about δ beside the interface in which the equilibrium ^3He concentration $x_s(T, P_0 + \delta P)$ is accomplished—i.e., the phase conversion is completed. Then, the growth rate of the interface with the sound of frequency ω may be proportional roughly to $\omega\delta$ —i.e.,

$$\xi \propto \omega\delta. \quad (60)$$

One recognizes that, if one takes $\delta_D(\omega)$ as δ , one obtains

$$\xi \propto \omega\delta_D(\omega) = \sqrt{\omega D}, \quad (61)$$

which coincides with the prefactor of Eq. (55) without the term due to the second-sound mode. So it seems reasonable to assume that δ is proportional to $\delta_D(\omega)$; i.e., the finite part of the region of $\delta_D(\omega)$ completes the phase conversion.

As is discussed in the preceding section, in order to complete the phase conversion, we need, first, the flow of ^3He quasiparticles and, second, the process to realize the thermal equilibrium with the surrounding bulk liquids. In the hydrodynamic region, the flow is a diffusion one. This fact may be taken into account if we assume $\delta \propto \delta_D(\omega)$ as mentioned above. The second process of thermal relaxation may be important to determine how large the part of the region $\delta_D(\omega)$ which completes the phase conversion. This aspect is not

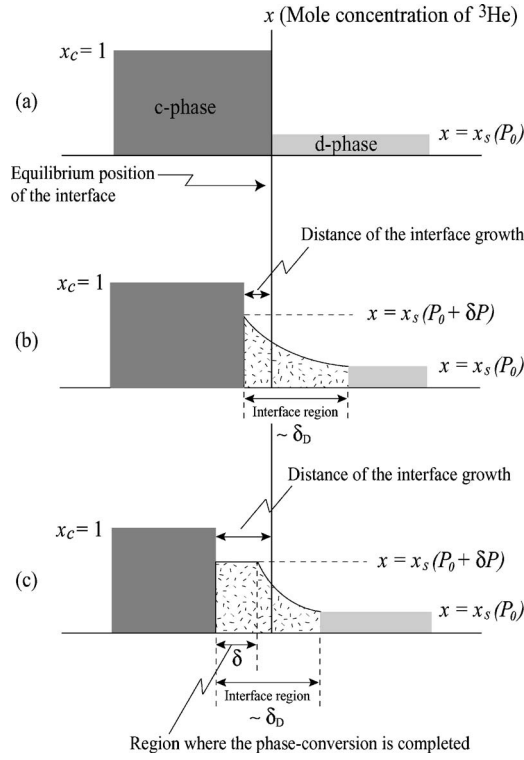


FIG. 12. Schematic illustration of the region near the interface. (a) The equilibrium interface at the overall pressure $P = P_0$. The ${}^3\text{He}$ concentration of the d phase is $x_s(T, P_0)$ and that of the c phase is $x_c = 1$. (b) The interface region considered in Ref. 17 when extra pressure δP is applied to the interface due to incident sound wave. The thermodynamically equilibrium value of ${}^3\text{He}$ concentration $x_s(T, P_0 + \delta P)$ is achieved only at the interface on the d -phase side. (c) The condition for the pressure is the same as in (b). Here, however, we have a finite region of about δ in which the concentration becomes $x_s(T, P_0 + \delta P)$.

fully considered in Ref. 17. Since we have both the diffusive mode and the propagating first-sound mode in the interface region, it may be not much unreasonable to assume that the second process in the hydrodynamic region is concerned with the attenuation process of the first-sound mode. Then, Eq. (60) may be modified as

$$\xi \propto \omega \delta_D(\omega) \alpha_{int}, \quad (62)$$

where α_{int} is the attenuation coefficient of first sound in the interface region. It seems reasonable to assume

$$\alpha_{int} \propto \omega^2 \tau_{int} \text{ with } \tau_{int} \sim \tau_{(2)} \propto T^{-2} \quad (63)$$

in the hydrodynamic limit. Here, τ_{int} is the relaxation time of ${}^3\text{He}$ quasiparticles in the interface region. Then, from Eq. (62) we may conclude that the growth coefficient in the hydrodynamic regime is proportional to

$$\omega \omega^{-1/2} T^{-1} \omega^2 T^{-2} = \omega^{5/2} T^{-3}. \quad (64)$$

This result agrees with the observed dependence of ξ on the temperature and frequency of Eq. (42).

The agreement mentioned above may suggest the direction of the improvement for the theoretical considerations made in Ref. 17. In order to obtain the formula of ξ as Eq. (62), it seems necessary to take into account an entanglement between the diffusive mode and the first-sound mode which coexist in the interface region. At present, we have not succeeded to formulate the situation shown in Fig. 12(c).

The observed fact (iii) mentioned in Sec. IV A can be understood if we approximate τ_{int} by

$$\frac{1}{\tau_{int}} \approx \frac{1}{\tau_c} + \frac{1}{\tau_d}. \quad (65)$$

Since τ_d is always much longer than τ_c , τ_c may be most effective for the process in the interface region.

The extension of the present study into the higher-temperature region seems interesting since the higher the temperature, the contribution of ${}^4\text{He}$ quasiparticles to the mass flow across the interface would become the more noticeable. The phase conversion of ${}^4\text{He}$ quasiparticles means the conversion process between quasiparticles of the Bogoliubov type⁴⁹ in the d phase and those of the Zharkov-Silin type⁵⁰ in the c phase. It would be worthwhile to see how such a process affects the interface mobility.

V. SUMMARY

We have carried out a systematic study of the sound transmission across the interface between the ${}^3\text{He}$ -dilute and ${}^3\text{He}$ -concentrated phases of a separated ${}^3\text{He}$ - ${}^4\text{He}$ liquid mixture. The interface represents a new enticing object for investigating the phase transition kinetics in a quantum liquid mixture. To obtain the interface transmission coefficient, we have developed an original experimental method based on displacing the interface position between two transducers with a uniform velocity at constant temperature and pressure.

The measurements of the sound transmission coefficient, performed at various temperatures, pressures, and frequencies, have revealed an essential discrepancy with the standard acoustic-mismatch theory at sufficiently low temperatures. We have observed an anomalous temperature- and frequency-dependent behavior of the sound transmission coefficient. It shows a frequency-dependent minimum at the temperature $T_{\tau_{min}}(\omega)$ and the reduction at this minimum point is almost a factor of 10 or much more depending on the frequency. The results are interpreted in terms of the kinetic growth coefficient of the interface, ξ . The prominent features observed for ξ are as follows.

(i) The kinetic growth coefficient $\xi(T, P, \omega)$ shows a frequency-dependent maximum at $T_m(\omega)$ which corresponds to $T_{\tau_{min}}(\omega)$.

(ii) Above $T_m(\omega)$, ξ is frequency dependent and falls with an increase of the temperature as $\xi \propto \omega^{5/2} T^{-3}$.

(iii) Below $T_m(\omega)$, ξ becomes frequency independent and also diminishes as $\xi \propto T^3$ as the temperature is lowered.

(iv) For the same frequencies, the values $T_m(\omega)$ nearly coincide with the temperatures $T_{c,m}(\omega)$ at which the sound attenuation coefficient of the c phase shows a maximum.

(v) The measurement of the phase-separation pressure dependence at a fixed temperature of about 30 mK shows that ξ increases in proportion to $(\partial c_s / \partial P)_T^2$.

The crossover observed in the temperature dependence of ξ [(i)] is important. Taking into account (iv), it may be reasonable to consider the temperature region $T < T_m(\omega)$ as the ballistic one for ^3He quasiparticles. Our discussion on item (iii), given in Sec. IV D, is made from this point of view. First, we note that the collision process among quasiparticles is necessary in order to realize and complete the phase-conversion process. Then, it is physically very reasonable that ξ decreases with a reduction of the collisional frequency. That is, without collisions we have no phase conversion. Second, we note that a large difference in the Fermi momentum between the c and d phases, as is given by Eq. (1), is crucial. Two-particle collisions cannot be effective, and we must consider higher-order three-particle collisions. Then, taking into account the subsequent process of thermal relaxation necessary for completion of the phase conversion, we can arrive at the T^3 dependence of ξ in agreement with the observation. We believe that the present result (iii) is the first case of experimental observations in liquid ^3He - ^4He mixtures, which shows the importance of three-particle collisions among ^3He quasiparticles.

Observation (v) is also very important. First, it unambiguously shows that ξ determined with the present method reflects the phase conversion induced by the sound pressure at the interface. Second, the observed $(\partial c_s / \partial P)_T^2$ dependence is consistent with the theoretical prediction made in Ref. 17, Eq. (55). In the theory, that dependence is associated with a tendency of the system towards phase equilibrium at the interface [Fig. 12(b)]. However, the temperature and frequency behavior for the prefactor of Eq. (55), $(\omega D)^{1/2} \propto \omega^{1/2} T^{-1}$, on the neglect of the contribution from the second-sound mode, does not agree with observation (ii). So we propose the situation of Fig. 12(c) instead of Fig. 12(b). We have not yet succeeded in formulating the situation of Fig. 12(c). Assuming the relation of Eq. (62) for the expression of δ in Fig. 12(c), we arrive at Eq. (64) in agreement with observation (ii). It is a future problem to investigate the validity of the speculation, Eq. (62), from a reasonable theoretical formulation, which would also give the transmission coefficient $\tau_{c \rightarrow d}$ and the growth coefficient ξ as real-valued quantities.

Throughout the discussions in Secs. IV D and IV E we consider that the phase-conversion process is completed when the phase-converted region sets in thermal equilibrium with the surrounding bulk liquids. Then, we obtain the dependence of ξ on the temperature and frequency which agrees with the ones observed in both the ballistic and hydrodynamic regimes. So it seems that the transmitted sound detects the interface growth as a region where the phase conversion is completed in the sense mentioned above, at least within the frequency range presently studied.

Here we would like to mention the important aspect of the present results in connection with those of nucleation phenomenon in supersaturated ^3He - ^4He superfluid mixtures.⁴ In Ref. 4 it is observed that the critical supersaturation of superfluid ^3He - ^4He mixtures is almost independent of temperature below about 10 mK. Furthermore, in this tempera-

ture region, the phase separation completes very quickly when the nucleation starts. On the other hand, the present sound experiments reveal that the interface mobility is very small in this low-temperature region. In order to compromise these apparently contradicting aspects, it seems necessary to assume the occurrence of nucleation of the c phase at greatly many nucleation centers. Then, nucleation seeds easily coalesce to form large enough droplets, much larger than the mean free path of ^3He quasiparticles. That is, the present sound experiment strongly supports homogeneous nucleation in the experiment of Ref. 4.

The interesting aspect of the extension of the present study into the higher-temperature region is noted in Sec. IV E. An extension into the lower-temperature region in the ballistic regime also seems very interesting in connection with the Andreev reflection phenomena.⁵¹ The existence of the Andreev reflection is a general effect for the boundary between the normal fluid and the Cooper-paired superfluid. The effect is well known and was studied originally for the normal metal-superconductor (N-S) configuration. We may think of the analog of Andreev's original N-S boundary for the interface between the d and c phases provided the c phase becomes a superfluid state. In fact, from the standpoint of Fermi ^3He quasiparticles the interface represents a boundary separating normal states in the d phase and Cooper-paired states in the superfluid c phase. Since Fermi excitations in the superfluid state have an energy gap in the spectrum, Fermi excitations in the normal state, whose energy is smaller than the energy gap, must be reflected due to impossibility of satisfying the energy conservation law. This affects the transmission of Fermi quasiparticles across the interface and, correspondingly, should modify the sound transmission and reflection coefficients. Due to different character of the energy gap in the A and B phases, we expect different behavior of the sound coefficients in these superfluid states. In this respect, it would be worthwhile to investigate how the Andreev reflection phenomena affect the sound coefficients.

Finally, we wish to note advantageous aspects of the present experimental way of sound measurement. First, as is described in Sec. II C, we start the measurements with pure liquid ^4He and then condense ^3He to prepare a phase-separated mixture. So the absolute accuracy of the sound attenuation measurements can be referred to the accuracy of measurements in pure liquid ^4He . The good agreement of our $\alpha_d(T, P, x_s, \omega)$ with the calculated curves based on the theoretical formula in a wide temperature range, as is shown in Sec. III A, supports the accuracy of our measurements. Second, as is noted in Sec. II A for the measurement of the transmission coefficient $\tau_{c \rightarrow d}$, we always utilize the signal A_d transmitted through the saturated d phase. This fulfills the role of a crucial check of the measurements with phase-separated mixtures at every stage. The satisfactory agreement of our $\alpha_c(T, P, \omega)$ with the data published for pure liquid ^3He evidences the reliableness of our present method as a whole.

In conclusion, we have presented a method and enticing results which will trigger further experimental and theoretical studies of the interfacial dynamics in phase-separated ^3He - ^4He liquid mixtures.

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