VOLUME 16, NUMBER 23

trices 4×4 and 47×47 , respectively, after due reduction for rotational symmetry around the cylinder). The small bump occurring around $\mu/kT \simeq 3$ for N = 5 is readily changed into a high narrow peak for N = 10 at $\mu/kT \simeq 3.6$ with the corresponding values $p/kT \simeq 0.74$ and $\rho \simeq 0.175$ (maximum density of this system is $\rho = 0.20$). This clearly demonstrates that this system will (in the limit $N \rightarrow \infty$) display a phase transition. No definite conclusion about its exact nature can be drawn here. It appears, however, that the peak of Fig. 2 is much sharper than the corresponding one observed by Runnels³ for first-neighbor exclusions only; therefore, the transition reported here could well be of the first-order type. This view is confirmed to some extent by approximate calculations for this same system based on the Rushbrooke-Scoins theorem⁴ (which is nearly equivalent to the Kikuchi method): Including all configurations based on groups of five sites and

less, we found a first-order phase transition with the following features:

$$\rho_{\text{fluid}}^{* \simeq 0.160, \rho_{\text{solid}}^{* \simeq 0.192, (p/kT)^{*} \simeq 0.738,}$$

$$(\mu/kT)^{* \simeq 3.64, S_{\text{fusion}}^{\simeq 0.78k,}$$

in good agreement with the position of the peak of Fig. 2.

A detailed account of this work will be published soon. We are much indebted to Professor I. Prigogine for his constant interest in this research.

²D. A. Gaunt and M. E. Fisher, J. Chem. Phys. <u>43</u>, 2840 (1965).

³L. K. Runnels, Phys. Rev. Letters <u>15</u>, 581 (1965).

MEASUREMENTS OF THE ULTRASONIC ATTENUATION IN LIQUID He⁴ AT LOW TEMPERATURES*

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We wish to report some preliminary measurements of the attenuation of ultrasound, $\alpha(\omega, T)$, in liquid He⁴ at low temperatures and at frequencies of 30, 90, and 150 Mc/sec, where ω is the angular frequency and T the absolute temperature. Interest in this work arose because of the disagreement among the various theoretical explanations advanced to explain the attenuation and because the reported experimental results failed to support any of the theories.

Landau and Khalatnikov¹ made a power series expansion of the dispersion relation, which for small momenta can be written $\epsilon(p) = c p (1 - \gamma p^2)$, where c is the velocity of sound. They used the available experimental results to evaluate the constants and pointed out that γ was positive; consequently, energy and momentum could not be conserved in a three-phonon process. They concluded, therefore, that the lowest order process that could contribute to the attenuation was a four-phonon process. For this latter mechanism, Khalatnikov² showed that α varied as ωT^6 . Others³⁻⁵ assumed the three-phonon process and found that α varies as ωT^4 . Kawasaki⁶ justified this assumption by pointing out that if the energy uncertainty $(\delta \epsilon)$ arising from the finite lifetime (τ) of the thermal phonons satisfied the inequality $\delta \epsilon = \hbar/$ $\tau \ge 3\gamma \bar{p}^2 \hbar \omega$, then the three-phonon process could take place. The average thermal phonon momentum \bar{p} is 3kT/c. Recently Kwok, Martin, and Miller⁷ and Pethick and ter Haar⁸ recalculated the attenuation arising from a threephonon mechanism by taking into account the finite phonon lifetime and found⁸

$$\alpha = \frac{\pi^3}{60} \frac{(\mu+1)^2}{\rho} \frac{\omega(kT)^4}{\hbar^3 c^6} \times \left[\arctan\omega\tau - \arctan(\frac{3}{2}\gamma\overline{\rho}\omega\tau)\right], \tag{1}$$

where ρ is the density and $u = (\rho/c)\partial c/\partial \rho$. At temperatures below 500 mdeg, $\omega \tau \gg 1$ for the frequencies used in our experiments and one can then replace $\arctan \omega \tau$ by $\pi/2$. In the one limit where $\frac{3}{2}\gamma \bar{p}\omega\tau < 1$, one recovers the previously mentioned expression for the threephonon process, $\alpha \propto \omega T^4$; in the other limit where $\frac{3}{2}\gamma \bar{p}^2\omega\tau > 1$, one obtains an expression

¹D. M. Burley, Proc. Phys. Soc. (London) <u>77</u>, 451 (1961).

⁴See, for example, H. N. V. Temperley, Proc. Phys. Soc. (London) <u>80</u>, 813 (1962).

for the attenuation which is independent of frequency. Recently Khalatnikov and Chernikova⁹ derived an expression [Eq. (3) of their paper] which agrees within a factor $\pi/2$ with Eq. (1) in the limits $\gamma = 0$, $\omega \tau \gg 1$.

The experiments reported here show that the attenuation varies nearly as T^4 but that the frequency dependence does not follow a simple power law. In fact, the experimental attenuation appears to become independent of frequency at the higher frequencies. These experiments are qualitatively explained by Eq. (1) and support the three-phonon process as the major contribution to the attenuation in the temperature range 120-450 mdeg.

The copper sonic cell was constructed as a bomb; it was filled at room temperature with purified helium to a pressure of 500 psi and sealed off. In this way all problems arising from film flow or thermal conductivity through bulk He II were avoided. The cell was thermally bonded to a potassium chrome alum refrigerator pill. Carbon resistance thermometers were fastened to both the refrigerator and the cell; in addition, a cerium magnesium nitrate magnetic thermometer was fastened to the cell only. The thermometers were calibrated in situ against the vapor pressure of liquid He³ over the temperature range 600-1600 mdeg at the beginning and end of the experiment. Within the error of the temperature measurement (±2 mdeg) the calibration remained unchanged. The low temperatures were reached by adiabatic demagnetization from 500 mdeg, obtained with a He³ cryostat.

Freely vibrating, coaxially plated, 30-Mc/ sec transducers were employed for both the transmitter and receiver. The transducers, clamped only at the rim, were separated by a fused-quartz spacer 1.022 cm in length. The rise time of the pulse was about 10 μ sec and the width about 30 μ sec, which is to be compared with the one-way transit time through the He of about 50 μ sec. No detectable change in the warming rate (0.3 mdeg/min with no)applied rf power) was observed at the repetition rate of 10 cps used in this experiment. The total insertion loss from transmitter to receiver was 40 dB at 30 Mc/sec. With rf pulses of 5 mW peak power, the signal-to-noise ratio was 70 dB at the low temperatures. Even at very low power levels, it was observed that the attenuation became amplitude-dependent; consequently, all measurements were carefully checked so as not to exceed the critical power level. The attenuation was measured relative to a comparison pulse which initially was obtained from a separate oscillator. In the later measurements a single oscillator was used to eliminate any errors arising from relative amplitude drifts in the oscillators. A "wave guide beyond cutoff" attenuator with an accuracy of 0.01 dB was used for all measurements.

After demagnetization the temperature of the cell was followed for a period of about one hour to be certain of temperature equilibrium. During this time interval the temperature change was never more than 6 mdeg; as a consequence several measurements of the pulse height established an accurate reference with which to compare changes in attenuation. The cell was then heated and after temperature equilibrium had been re-established the pulse height was remeasured. This procedure was followed until the signal disappeared into the noise or until finite-amplitude effects precluded further measurements.

Figures 1, 2, and 3 show the attenuation as



FIG. 1. Ultrasonic attenuation at 30 Mc/sec as a function of temperature.

a function of temperature for 30, 90, and 150 Mc/sec, respectively. Between approximately 120 and 150 mdeg no change in pulse height was observed for the 90- and 150-Mc/sec runs within the reproducibility of the data $(\pm 0.1 \text{ dB})$. This observation, coupled with the theoretically supported assumption that $\alpha \propto T^n$, where $n \ge 4$, allows us to assign to the lowest temperature point a value of zero attenuation with an error of at most 0.1 dB. Thus, the data plotted in Figs. 2 and 3 can be considered as absolute attenuation measurements. In the lowtemperature region where the data can be fitted to $\alpha = AT^n$, *n* equals 4.45 and 4.05 for 90and 150-Mc/sec data, respectively. The reference level for the 30-Mc/sec data was not established as accurately as the level for the other frequencies because of an experimental mishap; however, these data are reliable within the stated error $(\pm 0.1 \text{ dB})$ as relative attenuation measurements. By adding 0.6 dB to all the data in Fig. 1, which is not an unreasonable amount, the curvature exhibited at low temperatures would be removed and the data would lie on a straight line with slope corresponding



FIG. 2. Ultrasonic attenuation at 90 Mc/sec as a function of temperature.

to n = 3.99. The attenuation measured at the minimum, ca. 2°K, is 5.5 dB/cm. If this value is scaled as ω^2 to 12 Mc/sec, one obtains 0.89 dB/cm. This value is 0.24 dB/cm higher than the value measured by Chase,¹⁰ but is within the quoted combined experimental errors.

Two additional high-temperature runs at 30 Mc/sec were also made. These runs were normalized by adjusting the value at the minimum in the attenuation curve ($T \simeq 1.9^{\circ}$ K) to coincide with that for the low-temperature run. One high-temperature run was made at 90 Mc/sec and normalized to the low-temperature run.

The maximum frequency used in this experiment is a factor of 10 greater than any used in previously reported ultrasonic studies in liquid He⁴ and the minimum is a factor of 2 greater. Thus, it is not possible to make a meaningful comparison between these results and those of other workers,^{11,12} particularly those of Jeffers and Whitney,¹³ who find that $\alpha \propto \omega^{3/2}T^3$ in the frequency range 1-12 Mc/sec. In conclusion we can say that the temperature



FIG. 3. Ultrasonic attenuation at 150 Mc/sec as a function of temperature.

dependence of the attenuation is close enough to T^4 to support firmly the three-phonon process as the principal attenuation mechanism between 120-450 mdeg. The data are not yet complete enough to draw detailed conclusions about the frequency dependence except to say that the attenuation is becoming independent of frequency as the frequency is raised. This observation is in qualitative agreement with Eq. (1), which is based on the three-phonon process.

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i Teor. Fiz. 19, 637, 709 (1949).

²I. M. Khalatnikov, Zh. Eksperim. i Teor. Fiz. 44, 769 (1963) [translation: Soviet Phys.-JETP 17, 519 (1963)].

³T. O. Woodruff, Phys. Rev. <u>127</u>, 682 (1962).

⁴K. Dransfeld, Phys. Rev. <u>127</u>, 17 (1962).

⁵K. Kawasaki and H. Mori, Progr. Theoret. Phys. (Kyoto) 28, 784 (1962).

⁶K. Kawasaki, Progr. Theoret. Phys. (Kyoto) 26, 795 (1961).

⁷P. C. Kwok, P. C. Martin, and P. B. Miller, Solid State Commun. 3, 181 (1965); and private communication.

⁸C. J. Pethick and D. ter Haar, to be published; and private communication.

⁹I. M. Khalatnikov and D. M. Chernikova, Zh. Eksperim. i Teor. Fiz.- Pis'ma Redakt. 2, 566 (1965) [translation: JETP Letters 2, 353 (1965)].

¹⁰C. E. Chase, Proc. Roy. Soc. (London) A220, 116 (1950).

¹¹C. E. Chase and M. A. Herlin, Phys. Rev. <u>97</u>, 1447 (1955).

¹²K. Dransfeld, J. A. Newell, and J. Wilks, Proc. Roy. Soc. (London) A243, 500 (1958).

¹³W. A. Jeffers, Jr., and W. M. Whitney, Phys. Rev. 139, A1082 (1965).

LOCALIZED MAGNETIC MOMENTS IN METALS*

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The electronic properties of an otherwise nonmagnetic metal into which has been dissolved a dilute concentration of magnetic impurities have been obtained starting from the Anderson Hamiltonian¹:

$$\mathcal{K} = \sum_{\sigma} \mathcal{E}_{d\sigma} d_{\sigma}^{\dagger} d_{\sigma} + U \hat{n}_{\dagger} \hat{n}_{\dagger} + \sum_{\vec{k},\sigma} \mathcal{E}_{\vec{k},\sigma} C_{\vec{k},\sigma}^{\dagger} C_{\vec{k}\sigma} + \sum_{\vec{k}\sigma} V_{\vec{k}} (d_{\sigma}^{\dagger} C_{\vec{k}\sigma} + C_{\vec{k}\sigma}^{\dagger} d_{\sigma}), \qquad (1)$$

where $V_{\vec{k}}^* = V_{\vec{k}}^*$, d_{σ}^{\dagger} is a creation operator for an electron in the single *d* orbital, $C_{\vec{k}\sigma}^{\dagger}^{\dagger}$ is a creation operator for a conduction electron of wave vector \vec{k} , and where

$$\mathcal{E}_{d\sigma} = \mathcal{E}_{d} - g\sigma H,$$

$$\mathcal{E}_{\overline{\mathbf{k}}\sigma} = \mathcal{E}_{\overline{\mathbf{k}}} - \sigma H,$$

$$\mathcal{E}_{d} = \epsilon_{\mathbf{F}} - \xi U, \quad \xi \neq (0, \frac{1}{2}, 1).$$
(2)

In this work the interaction U is treated exactly, in contrast to the more usual Hartree-Fock approach. If we define Δ by

$$\Delta = \pi V_k \frac{2}{F} \rho(\epsilon_F), \qquad (3)$$

then we shall consider the case $U \gg \Delta$. Even in this case, however, a perturbation expansion in powers of Δ/U is not valid for $\Delta \gg T$. If we define the Green's function for the d electron by

$$G_{d}^{\sigma}(\omega) \equiv i \int_{0}^{\infty} e^{i\omega t} \langle [d_{\sigma}(t), d_{\sigma}^{\dagger}]_{+} \rangle dt, \qquad (4)$$

then for $|\omega - \epsilon_F| \ll T$, $G_d^{\sigma}(\omega)$ may be expanded in the form

$$G_{d}^{\sigma}(\epsilon_{\mathbf{F}}) = \sum_{m=1}^{\infty} B_{m}^{\sigma}(n^{-\sigma},\xi) \left(\frac{\Delta}{U}\right)^{m} + \sum_{m=0}^{\infty} h_{m}\left(n^{\sigma},\xi,\frac{\Delta}{U}\right) \ln^{m} \left\{\frac{2D\eta}{\pi T}\right\}, \quad (5)$$

where $\ln \eta = 0.577 \cdots$ and where D, considered to be of order U, is introduced as a lower bound on the conduction-electron energies in order to obtain convergent integrals when V_k and $\rho(\epsilon_k)$ are treated as constants, and where $n^{\sigma} = \langle d_{\sigma}^{\dagger} d_{\sigma} \rangle$. The function $h_m(n^{-\sigma},\xi,\Delta/U)$ may be expanded

1042

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