

TABLE I. Comparison between our previous measurement and those of Wallenstein, Paisner, and Schawlow for $v' = 62$, $J' = 27$ level.

	g_J	g_1
Our previous measurement (Ref. 2)	-1.6 ± 0.2	3.5 ± 0.5
Wallenstein, Paisner, and Schawlow (Ref. 1)	-2.6 ± 0.2	7.5 ± 0.8

comes not only from the $v' = 62$, $J = 27$ level but also from four other levels because the 5017-Å line coincides with the following transitions: 62-0 $P(12)$; 62-0 $R(26)$; 64-0 $R(79)$; 67-0 $R(49)$; (70 ± 1) -0 $R(54 \pm 1)$. The g_J and g_1 values of the levels excited by these transitions have been measured by the technique of resonances in a modulated light beam. The details of the experiment and the complete results will be published elsewhere. However, we extract from these results the content of Table II.

The comparison between Table I and Table II leads to the following conclusions: Our previous measurement was correct and is confirmed. The measurement of Wallenstein, Paisner, and Schawlow is also correct. But it refers to the $v' = 64$, $J' = 40$ level and not to the $v' = 62$, $J' = 27$. To support this conclusion, we present the following arguments: In our experiment, the laser was oscillating in a single mode in order to excite mainly one level and a spectroscopic study of the fluores-

TABLE II. The results of our present experiment.

	g_J	g_1
$v' = 62$, $J' = 27$	-1.82 ± 0.06	3.65 ± 0.4
$v' = 64$, $J' = 40$	-2.7 ± 0.3	7.5 ± 1.7

cence permits an unambiguous assignment of this level. On the contrary, Wallenstein, Paisner, and Schawlow never studied the fluorescence spectrum but decided on their assignment after a precise comparison between the wavelength of their dye laser and that of an Ar^+ laser oscillating at 5017 Å. This comparison is clearly insufficient, the 62-0 $R(26)$ and the 64-0 $R(39)$ absorption lines lying within 3 GHz. Moreover the fluorescence intensities that they provide are of the same order. Therefore we think that this controversy has at least the interest of pointing out the efficiency of the technique of resonances in a modulated light beam.

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Roton Second Sound*

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Second-sound propagation is considered, both when collisions between elementary excitations do and when they do not conserve total number. Simple expressions are derived for the velocity in each case. Assuming number conservation in roton-roton collisions, the velocity of roton second sound is found to be $3kT/p_0$, where p_0 is the momentum at the roton minimum.

Recent experiments by Dynes, Narayanamurti, and Andres¹⁻³ have convincingly demonstrated the existence of roton second sound in superfluid He⁴. In their experiments a pulse of phonons was generated by electrically heating a metallic film in contact with the liquid. The detector was a bolometer placed a few millimeters away. At

high temperatures (e.g., 1 K) each pulse generated gave one pulse at the detector. The propagation velocity was close to the second-sound velocity given by Landau's formula⁴

$$c_2^2 = TS^2/\rho_n C, \quad (1)$$

where S , C , and ρ_n are the entropy, specific

heat, and normal-fluid density, respectively. At a liquid pressure of 24 atm and at temperatures around 0.6 K two pulses were detected by the bolometer. The pulse that arrived first had a velocity close to the velocity of ordinary (first) sound and was produced by phonon excitations which had traveled without scattering from the generator to the detector. The slower pulse had a velocity somewhat greater than that of second sound at 1 K and is believed to be roton second sound. This is a collective excitation similar to ordinary second sound, but involving only excitations from the roton part of the spectrum. It is possible for there to be a collective excitation of the rotons uncoupled to the phonons because, under the conditions of the experiment, the rotons interact strongly among themselves (collision time τ_{RR}) but weakly with the phonons (collision time $\tau_{PR} \gg \tau_{RR}$). Roton second sound was first predicted by Khalatnikov and Chernikova,^{5,6} who carried out detailed calculations of τ_{RR} and τ_{PR} to determine the conditions under which it could be observed. Their result for the velocity was

$$c_{2R}^2 = TS_R^2 / \rho_{nR} C_R, \quad (2)$$

where the quantities S_R , C_R , and ρ_{nR} have the same meaning as in Eq. (1), but only the roton contribution to each quantity is considered. In this Letter I make some general comments on the characteristics of second-sound propagation, and consider the validity of the Khalatnikov-Chernikova result for the velocity of roton second sound.

The second sound velocity can be derived by starting from the Boltzmann equation for the excitations. This is

$$n_p = (\dot{n}_p)_{c_{011}} - \vec{v}_p \cdot \nabla n_p, \quad (3)$$

where n_p and \vec{v}_p are the number and group velocity of excitations of momentum \vec{p} , and the first term on the right-hand side is the contribution to \dot{n}_p from collisions between excitations. I consider only solutions of (3) which are sufficiently slowly varying in space and time that n_p always corresponds to a distribution which is in local equilibrium. The most general form of n_p is then

$$n_p = \{ \exp[(\epsilon_p - \vec{p} \cdot \vec{V} + \Delta\mu) / k(T + \Delta T)] - 1 \}^{-1}, \quad (4)$$

where \vec{V} , $\Delta\mu$, and ΔT are the local drift velocity, change in chemical potential, and change in temperature, respectively. $\Delta\mu$ will be zero unless the number of excitations is conserved in collisions between excitations. If one now looks for wave solutions of Eq. (3) in which \vec{V} , $\Delta\mu$, and

ΔT all vary as $\exp[i(Kz - \Omega t)]$, one can use the conservation laws of energy, number, and momentum to derive the following equations:

$$c[A_{200}(\Delta T/T) + A_{100}\Delta\mu] - A_{111}V/3 = 0, \quad (5)$$

$$c[A_{100}(\Delta T/T) + A_{000}\Delta\mu] - A_{011}V/3 = 0, \quad (6)$$

$$A_{111}(\Delta T/T) + A_{011}\Delta\mu - cA_{020}V = 0, \quad (7)$$

where $c \equiv \Omega/K$ is the wave velocity, and

$$A_{\alpha\beta\gamma} = \int \epsilon_p^\alpha p^\beta v_p^\gamma \bar{n}_p (\bar{n}_p + 1) d\tau_p; \quad (8)$$

\bar{n}_p is the equilibrium distribution function for the excitations.

In the standard theory of second sound⁷ it is assumed that collisions between excitations do not conserve number. Then $\Delta\mu = 0$ and only Eqs. (5) and (7) should be considered. The resultant velocity is thus

$$c_\mu^2 = A_{111}^2 / 3A_{200}A_{020}. \quad (9)$$

It is straightforward to show that $A_{111} = 3kT^2S$, $A_{200} = kT^2C$, and $A_{020} = 3kT\rho_n$. Thus Eq. (9) is equivalent to the standard result, Eq. (1). The subscript μ is added to the velocity to emphasize that in this case second sound is a wave at constant (and zero) chemical potential.

At 1 K and above it is certainly correct to set $\Delta\mu = 0$ because there are many collisions between excitations which change the overall number of excitations. However, in considering roton second sound one must not make this assumption. Roton second sound can only exist when collisions between rotons and phonons can be neglected. Thus, the important conservation laws are those pertaining to roton-roton scattering. Now this scattering is almost entirely of the type

$$R + R' \rightarrow R'' + R'''$$

and thus conserves number. Hence to calculate the velocity one should solve Eqs. (5)–(7) with nonzero $\Delta\mu$. The result is⁸ a velocity c_N for “constant number” waves, given by

$$c_N^2 = \frac{A_{011}^2 A_{200} - 2A_{011} A_{111} A_{100} + A_{111}^2 A_{000}}{3A_{020}(A_{200}A_{100} - A_{100}^2)}. \quad (10)$$

For the case of an arbitrary dispersion law for the excitations the velocities c_N and c_μ are different. In all cases $c_N > c_\mu$, because from (9) and (10)

$$c_N^2 - c_\mu^2 = \frac{(A_{011}A_{200} - A_{100}A_{111})^2}{3A_{020}A_{200}(A_{200}A_{000} - A_{100}^2)}. \quad (11)$$

Note that $A_{200}A_{000}$ is always greater than A_{100}^2 . For free particles with $\epsilon = p^2/2m$, evaluation of

the A coefficients gives

$$c_N = c_\mu = \frac{5}{3}^{1/2} (kT/m)^{1/2}. \quad (12)$$

For a gas of phonons, one has $\epsilon = cp$, and one finds

$$c_N = c_\mu = c/\sqrt{3}. \quad (13)$$

However, for a roton gas with dispersion law

$$\epsilon = \Delta + (p - p_0)^2/2m, \quad (14)$$

one obtains for low temperatures ($kT \ll \Delta$, $mkT \ll p_0^2$)

$$c_N = 3kT/p_0, \quad c_\mu = 3^{1/2}kT/p_0. \quad (15)$$

Thus the result c_N for the velocity of roton second sound differs from the Khalatnikov-Chernikova result c_μ by a factor of $\sqrt{3}$.

One can understand why there is a difference between c_N and c_μ for rotons but not for free particles or phonons, as follows. When number is conserved the collective excitation that is traditionally called second sound can alternatively be viewed as an ordinary sound wave propagating in the gas of excitations. Hence one might guess that

$$c_N = (B_{SN}/\rho_n)^{1/2}, \quad (16)$$

where B_{SN} , the bulk modulus of the excitation gas at constant entropy and number, is defined by

$$B_{SN} \equiv -V(\partial P/\partial V)_{S,N}.$$

The pressure of the gas is

$$P = \frac{1}{3} \int p v_p n_p d\tau_p. \quad (17)$$

It is straightforward to verify that the guess (16) is in agreement with the expression (10) for c_N in terms of the $A_{\alpha\beta\gamma}$ coefficients. This in turn leads to the conjecture that the velocity of ordinary second sound (i.e., second sound at constant and zero chemical potential) should be given by the surprisingly simple formula

$$c_\mu^2 = B_{S\mu}/\rho_n, \quad (18)$$

$$B_{S\mu} = -V(\partial P/\partial V)_{S,\mu=0}. \quad (19)$$

It is straightforward to show by use of thermodynamics that this is equivalent to the Landau formula (1) and to the expression (9) in terms of the $A_{\alpha\beta\gamma}$ coefficients. Consider now the derivatives involved in B_{SN} and $B_{S\mu}$. We may imagine two contributions to the total pressure change δP resulting from a volume change δV at constant entropy and chemical potential. The volume is

first changed by δV at constant S and N , and a pressure change δP_1 results. In general, a change $\delta\mu$ in the chemical potential will be produced. The second contribution δP_2 now arises from the change in pressure at constant V and S when $\delta\mu$ is reduced to zero as a result of number-nonconserving collisions between excitations. It is δP_2 which gives the difference between B_{SN} and $B_{S\mu}$. Now for a gas of free particles we can write

$$P = \frac{2}{3} \int \epsilon_p n_p d\tau_p. \quad (20)$$

Hence, the pressure is completely determined by the internal energy which cannot be changed by collisions, whether or not these collisions are number conserving. Thus $\delta P_2 = 0$ and B_{SN} and $B_{S\mu}$ are equal for free particles. Similarly, for a gas of phonons,

$$P = \frac{1}{3} \int \epsilon_p n_p d\tau_p.$$

and so again B_{SN} and $B_{S\mu}$ are the same. For rotons, however, the pressure is not just a function of the internal energy and so δP_2 is nonzero and B_{SN} is greater than $B_{S\mu}$.

The present theory predicts that the velocity of roton sound should be $(19 \text{ K}^{-1})T \text{ m sec}^{-1}$ at 24 atm, whereas the Khalatnikov-Chernikova theory gives $(11 \text{ K}^{-1})T \text{ m sec}^{-1}$. The experiments performed to date are not sufficiently accurate to confirm definitely one velocity or the other.⁹ More accurate measurements would be very worthwhile since if agreement with theory were obtained this would provide some confirmation of current views on roton-roton interactions.

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⁴Equation (1) and the subsequent discussion assume that $\rho_s \approx \rho$. This is a good approximation at 1 K and below.

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⁷See, for example, J. Wilks, *Liquid and Solid Helium* (Clarendon, Oxford, England, 1967).

⁸There exists one other solution to these equations;

this has $c=0$.

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Possible "New" Quantum Systems

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Systems of spin-aligned hydrogen isotopes are studied. They are shown to exhibit even more extreme "quantum" behavior than the helium isotopes. Spin-aligned hydrogen is predicted to be a gas at *all* temperatures and its Bose-Einstein condensation and possible superfluidity are discussed. Spin-aligned deuterium is predicted to show critical behavior strongly influenced by quantum mechanics. The preparation of spin-aligned hydrogen (in the presence of large magnetic fields) is also discussed.

Recently Dugan and Etters¹ and Etters, Dugan, and Palmer² have studied the ground-state properties of spin-aligned isotopes of hydrogen, which we denote by $H\uparrow$, $D\uparrow$, and $T\uparrow$. They used a Morse-potential approximation to the accurate pair potential for two hydrogen atoms in the $b^3\Sigma_u^+$ state calculated by Kolos and Wolniewicz.³ DEP pointed out that systems composed of these spin-aligned atoms were strongly quantum mechanical systems analogous to those composed of the helium isotopes and they briefly discussed their possible superfluidity.

Here we discuss further these spin-aligned systems which may conceivably be prepared in magnetic fields of the order of 10^5 G. Our main point is that the study of these systems should yield exciting and fundamental new information about the properties of quantum systems. Since $H\uparrow$ and $D\uparrow$ are lighter and more weakly interacting than ^4He or ^3He , they may be expected to exhibit even more extreme quantum behavior than helium does. The present work utilizes recent studies of quantum systems within the context of the quantum theorem of corresponding states (QTCS).⁴⁻⁶ We analyze the expected superfluid properties and the quantum-mechanically driven phase transitions which may occur in these systems.^{4,5} We also discuss briefly the feasibility of preparing spin-aligned systems.

We shall begin with a discussion of the K-W po-

tential for the $b^3\Sigma_u^+$ state of H_2 (Fig. 1). We have also pictured a Lennard-Jones (L-J) potential chosen to fit the well depth and position of the minimum of the K-W potential. Although DEP fit the K-W potential with a Morse potential, we believe that the L-J potential also gives a reasonable fit. Moreover, it represents the long-range r^{-6} part of the potential better, which is important for these weakly bound systems. In addition, it allows us to make contact with the wide range of calculations which already exist for L-J systems.^{4,5}

We now recall briefly the QTCS.^{7,4} It applies to a class of systems with a pair potential of the form $v(r) = \epsilon v^*(r/\sigma)$ where ϵ is the coupling constant, σ is the "collision diameter," and $v^*(x)$ is the same dimensionless function for each system in this class. For the L-J potential, $v^*(x) = 4(x^{-12} - x^{-6})$. We introduce the quantum parameter⁴ $\eta \equiv \hbar^2/m\epsilon\sigma^2$, the reduced temperature $T^* = k_B T/\epsilon$, the reduced volume $V^* = V/N\sigma^3$, and the reduced Helmholtz free energy $F^* = F/N\epsilon$. The QTCS states that for a one-component system, $F^* = F^*(T^*, V^*, \eta)$, where F^* depends *only* on the form of $v^*(x)$ and on whether the particles obey *Bose-Einstein* or *Fermi-Dirac* statistics.^{7,4} Values of ϵ , σ , η , and other useful quantities are given in Table I for $H\uparrow$, $D\uparrow$, $T\uparrow$, and other light elements. Heavier spin-aligned atoms such as $Li\uparrow$ and $N\uparrow$ are not expected to show significant