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Rayleigh-Brillouin spectrum of compressed He, Ne, and Ar. II. The hydrodynamic region

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The Rayleigh-Brillouin spectra of He, Ne, and Ar are reported for the hydrodynamic region. The experiments show that light scattering may be used to determine thermodynamic and transport properties of gases as a function of density and thus provide another method of probing intermolecular potentials. In addition, the transition from the hydrodynamic to the kinetic regime is examined in terms of generalized hydrodynamics.

Since the mid 1960's, there has been extensive theoretical and experimental work on the Rayleigh-Brillouin spectra of fluids. For molecular fluids this has included both the kinetic and the hydrodynamic regime (see for example, Refs. 1 to 4). For the rare gases most of the work has centered around the low-density region (see, for example, Refs. 5 and 6), presumably because workers thought that "nothing new" would appear in the hydrodynamic region, and because the kinetic region was interesting as it provided a test of the Boltzmann equation.[7] It is the purpose of this article to show that Brillouin spectroscopy has advanced to the point where thermodynamic and transport properties may be extracted from the spectra in the hydrodynamic region, and as a result information obtained concerning molecular interactions. This is to be contrasted with the kinetic region where the spectra are relatively insensitive to the molecular force law.[6,8] In part I we reported on the Rayleigh-Brillouin spectra of He, Ne, and Ar and analyzed the data in terms of scaling. There the major stress was on the results for low densities. Here we analyze the same data but restrict ourselves primarily to the hydrodynamic region. To avoid repetition the reader is referred to Refs. 8 and 9 for the experimental details.

Figure 1 is a plot of the digital data for helium at several densities. (The density in amagat units is the true density divided by the density at STP.) Additional results for neon and argon have been presented in part I. Except at the lowest pressure shown in the figure, one is clearly in the hydrodynamic region. As we wish to analyze the spectra using the hydrodynamic theory of Mountain,[10] it is evident that one problem which must be faced is the determination of the boundary between the region where one may use the hydrodynamic theory with confidence in its validity and the low-density or kinetic region. The low-density observations play a role in this and are the reason for showing an example of the low-density results in Fig. 1. The decrease of the width of the components with increasing density is evident in the figure. The instrumental width [full width at half maximum (FWHM)] shown in the figure makes a major contribution to the width at the highest density. As in part I we will average the data, such as those shown in Fig. 1, by superimposing two orders of the interferometer and by folding about the laser line or zero frequency. With about 100 channels in the interval from zero frequency to just beyond the Brillouin peak, the composite spectra then consist of some 400 data points in that interval. This is the form in which we wish to compare our results with theory. For further details on data manipulation the reader is referred to part I.

Starting from the linearized Navier-Stokes equation, Mountain[10] has derived an expression for the spectrum $S(k,\omega)$ which is proportional to the real part of

$$n(k,\omega) = \frac{-\omega^2 + i\omega(a+b)k^2 + abk^4 + C_0^2(1-1/\gamma)k^2}{-i\omega^3 - \omega^2(a+b)k^2 + i\omega(C_0^2k^2 + abk^4) + aC_0^2k^4/\gamma}, \tag{1}$$

where $a = \kappa/\rho_0 C_v$ and $b = (\frac{4}{3}\eta_s + \eta_B)/\rho_0$. Here, ω is the frequency, ρ_0 is the mean mass density, C_v is the specific heat at constant volume, γ is the specific heat ratio, C_0 is the adiabatic sound velocity, κ is the thermal conductivity, and η_s and η_B are the shear and bulk viscosity. For the densities used in the present experiments one can set the bulk viscosity to zero. In the extreme hydrodynamic limit (a and b small) this yields the well-known Rayleigh-Brillouin triplet with widths $\Delta\omega$, FWHM for the central and Brillouin components given by $\Gamma_c = 2(a/\gamma)k^2$ and $\Gamma_B = (a + b - a/\gamma)k^2$, and a ratio of intensities $I_c/2I_B$ given by $\gamma - 1$, the Landau-Placzek relationship. Using known values of the parameters we generated theoretical spectra from Eq. (1) which were then convoluted with

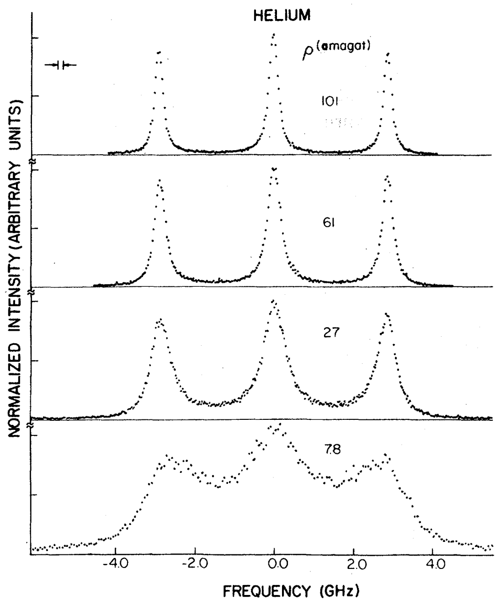

FIG. 1. A plot of the experimental points for the Rayleigh-Brillouin spectrum of He at several densities.

the experimentally determined instrumental profile. Allowance was made both for overlapping with the adjacent order of the interferometer and for the spread in wave vector arising from the finite cone of observation. The theoretical curves agreed with the experimental points within the statistical fluctuations in the observations. Therefore, the natural step to take was to invert the process and to try to determine the various parameters from the data. Although many authors have used the Brillouin component to determine the velocity of sound we belive this is the first attempt at extracting the maximum amount of information from the complete spectrum, although Cannell and Benedek[11] have taken a step in this direction with Xe near its critical point.

Examination of Eq. (1) reveals that the combinations of thermodynamic quantities that may in principle be determined are ak^2, bk^2, C_0k, and γ. We will assume that the scattering angle and the index of refraction are known so that k may be calculated with precision. Thus, the adjustable parameters are C_0, γ, a and b, or equivalently C_0, γ, the uniformity parameter $y = (\sqrt{2}/3) \times \rho_0 v_0/\eta k$, and the Eucken ratio $\kappa/\eta C_v$. (The uniformity parameter is widely used in kinetic discussions of the Rayleigh-Brillouin spectrum. Roughly speaking it is $1/kl$, where l is the mean free path. Thus $y \gg 1$ corresponds to the hydrodynamic regime.) Here v_0 is the mean molecular speed $(k_BT/m)^{1/2}$. From the point of view of precisely measuring these constants, the experiments must be regarded as rudimentary. One would want to improve the experimental technique before subjecting the data to a complicated least-squares analysis. We have carried out a simpler analysis of the spectra based on the fact that the spectra have certain easily identifiable characteristics.

The velocity of sound may be determined by adjusting C_0 in the theory until the calculated position of the Brillouin peak agrees with the experimental position. One does not use the usual Brillouin equation $\omega_B = kC_0$, as it is exact only in the absence of any overlap of the tail of the central component with the Brillouin component.

The value of γ may be determined using an algorithm to simulate the Landau-Placzek ratio. We computed the ratio of the integrated intensity from $\omega = 0$ to one-half the Brillouin frequency ω_B to the integrated intensity from $\frac{1}{2}\omega_B$ to one-half the spectral free range for both the experimental and theoretical curves, and we adjusted γ in the theory until the two ratios agreed. Here one risks making a systematic error. The very weak transmission by a Fabry-Perot interferometer, between orders, contributes to the results. Unfortunately, we did not measure the complete instrumental profile and so were forced to construct one artifically using the measured portion plus a constant background of about 10^{-4} times the peak height at maximum transmission. The value of 10^{-4} is the value calculated from the reflectivity of the plates. There is a systematic difference of about $\frac{1}{2}\%$ between the values of γ which are deduced with and without the background transmission of the interferometer. Experience gained after these experiments were completed indicates that the measured instrumental background may have contributions both from light scattered within the interferometer and around the interferometer in addition to the "theoretical" transmission at the minimum. The ideal situation is one in which the complete instrumental profile is obtained under identical conditions of illumination as the Rayleigh-Brillouin spectrum.

Table I lists the specific-heat ratios and the velocity of sound for He, Ne, and Ar at a number of densities as determined from the Rayleigh-Brillouin spectra. The calculated values of C_0 and γ included in Table I were determined from pressure-volume-temperature (PVT) data. There are two common ways of using PVT data to calculate C_0 and γ. A massive netting of the PVT surface over a wide range of temperature and pressure with an empirical 16-parameter equation of state has been carried out for He, Ne, and Ar by several authors.[12,13(a),13(b),14(a)] At

TABLE I. Specific heat ratios and velocity of sound for He, Ne, and Ar. Values in parentheses are last significant figures.

He

ρ(amagat)	Y	γ(PVT)[a]	γ(Expt)	Diff (%)	C_0(PVT)[a] (m/sec)	C_0(Expt)	Diff (%)	T(°C)
101.2	19.5(5)	1.652	1.66(5)	+0.8	1062	1061	−0.1	22
93.5	18.0(6)	1.653	1.65(2)	−0.1	1058	1058	±0.0	22
85.5	16.5(0)	1.655	1.66(3)	+0.5	1054	1053	−0.1	22
77.5	14.9(3)	1.656	1.65(8)	+0.1	1050	1047	−0.3	22
69.0	13.3(9)	1.657	1.66(9)	+0.7	1046	1043	−0.3	22
60.8	11.8(2)	1.658	1.66(3)	+0.3	1041	1043	+0.2	22
44.1	8.5(8)	1.660	1.67(0)	+0.6	1033	1036	+0.3	22
35.8	6.9(4)	1.662	1.65(9)	−0.2	1029	1033	+0.4	22
Mean error				+0.3			+0.1	

Ne

ρ(amagat)	Y	γ(PVT)	γ(Expt)	Diff (%)	C_0(PVT)	C_0(Expt)	Diff (%)	T(°C)
72.6	19.5(5)	1.697	1.67(2)	−1.5	473.8	467.9	−1.3	24
67.0	18.0(6)	1.695	1.68(5)	−0.6	472.0	465.3	−0.8	24
61.2	16.5(1)	1.692	1.69(2)	0.0	470.1	467.2	−0.6	24
55.3	14.9(3)	1.690	1.70(3)	+0.8	468.3	465.3	−0.6	24
49.5	13.3(9)	1.687	1.69(3)	+0.4	466.5	460.6	−1.0	24
43.7	11.8(2)	1.685	1.67(7)	−0.5	464.7	461.4	−0.7	24
31.7	8.5(8)	1.680	1.69(4)	+0.8	461.0	459.8	−0.3	24
25.6	6.9(4)	1.677	1.68(7)	+0.6	459.2	462.0	+0.6	24
Mean error				0.0			−0.6	

Ar

ρ(amagat)	Y	γ(PVT)[b]	γ(Expt)	Diff (%)	C_0(PVT)[b]	C_0(PVT)[b]	Diff (%)	T(°C) (±0 − 1)
37.9	19.5(5)	1.790	1.79(0)	±0.0	325.3	324.6	+0.2	24
34.9	18.0(6)	1.780	1.78(8)	−0.4	324.8	322.7	+0.6	24
31.4	16.5(0)	1.768	1.76(7)	+0.1	324.4	321.5	+0.9	23
28.4	14.8(5)	1.759	1.75(8)	+0.1	324.0	322.9	+0.1	23
25.5	13.3(9)	1.750	1.74(9)	+0.1	323.7	322.5	+0.4	23
22.5	11.8(2)	1.739	1.73(9)	±0.0	323.3	319.3	+1.2	23
16.2	8.5(8)	1.719	1.71(9)	±0.0	322.6	321.7	+0.3	23
13.1	6.9(4)	1.709	1.70(9)	±0.0	322.2	320.8	+0.4	24
Mean error				±0.0			+0.5	

[a] Reference 12.

[b] Reference 13(a).

best for He and Ar these calculations yield γ and C_0 accurate to 1 and $\frac{1}{2}$%, respectively, over the range of densities explored in the present experiments. No calculations of γ and C_0 have been reported for Ne, presumably because the experimental data base is not sufficient to determine the parameters of the equation of state with high precision. (The reader is reminded that specific heats are basicially second derivatives of the equation of state and, therefore, require extreme precision in PVT measurements.) We attempted to determine $C_p = T(\partial S/\partial T)_p$ from Yendall,[14(b)] but concluded the numbers tabled were not accurate enough to yield values better than 3% at best. The second common way differs from the first only in that the equation of state is assumed to have the virial form. Here one is "netting" only pressure-volume (PV) isotherms to determine temperature-dependent coefficients, rather than the entire PVT surface simultaneously. Apparently such data handling can lead to systematic errors in derived quantities such as C_p, C_v, etc. Using the precise PVT data of Ar the two methods do yield the same results [compare Refs. 13(a) and 13(b) with 13(c)]. For He our values of γ calculated from virial coefficients agree within 1–2% at worst with the values calculated by McCarty.[12] Thus for Ne, for which a preferred analysis is not possible, we resorted to a calculation of γ and C_0 in terms of the virial

TABLE II. Virial coefficients and their derivatives for Ne.

B	$\partial B/\partial T$	$\partial^2 B/\partial T^2$	C	$\partial C/\partial T$	$\partial^2 C/\partial T^2$
11.31	1.961×10^{-2}	-9.180×10^{-5}	236.3	-0.1865	1.064×10^{-2}

coefficients and their temperature derivatives (see, for example, Hirschfelder, Curtiss, and Bird[15]). For the virial coefficeints we have used those determined by the van der Waals laboratory as tabulated by Dymond and Smith.[16] In particular, we used those coefficients determined from a third- or fourth-order fit of the PVT data over a range of densities slightly larger than the range of density encountered in the Brillouin spectra of Ne. The values of the virial coefficients and their derivatives were calculated from a least-squares fit in Taylor-series form expanded about the temperature at which the spectra were recorded. The χ^2 and error in the coefficients were used to judge the appropriate order of polynomial for use in the Taylor expansion. The values of B, C, $\partial B/\partial T$, $\partial^2 B/\partial T^2$, $\partial C/\partial T$, and $\partial^2 C/\partial T^2$ so determined are given in Table II. We have not put an error limit on these calculated values but suspect that ±2% in γ and 1% in C_0 represent upper limits.

Returning to Table I, we see there is a systematic (mean) percentage difference between the calculated and our measured values of γ of 0.3%, 0.0%, and 0.0% for He, Ne, and Ar, respectively, while the random errors (standard deviation) appear to be about ±0.5%, ±0.8%, and ±0.2%. The latter agree well with our estimate of $\pm\frac{1}{2}\%$. The agreement is better than to be expected, since the calculated (PVT) values for He and Ar have an error of the order of ±1% at the highest densities used [see Ref. 13(a)]. Clark and Katz[17] have measured experimentally the specific-heat ratio for Ar and He. For Ar at 38 amagat units of density their results are 1.9% higher than the PVT or our value. For He their γ is 1.2% higher than our value, or 2.0% higher than the PVT value at the highest density. These differences are outside the estimated error limits. The reason for this is unclear at the moment.

For the velocity of sound the systematic (mean) errors are +0.1%, −0.6%, and +0.5% for He, Ne, and Ar, respectively. The random errors ±0.2%, ±0.8%, and ±0.6% are roughly consistent with our estimate of $\pm\frac{1}{2}\%$. Van Itterbeek *et al.*[18] have measured the velocity of sound in Ar and compared it with the velocity determined from the PVT data of Michels *et al.*[19] These are in complete agreement with each other, with our measurements, and with the calculation of Hilsenrath.[13(b)]

Overall, the agreement between the values of γ and C_0 determined from PVT data and from the Brillouin spectra is satisfactory, and it is evident that Brillouin spectroscopy can be used to measure γ and C_0 with some precision. With improvements, mainly to the interferometer, we estimate that an accuracy of 0.1% could be obtained. This would be considerably better than values obtainable from PVT data.

The transport coefficients determine the widths or, equivalently, the heights of the components. Thus, one can use the heights to determine the uniformity parameter y and the ratio of the heights to determine the Eucken ratio E. Two experimental factors limit the accuracy to about 10% for these quantities in the hydrodynamic region. First, the apparatus was arranged to prevent overlapping of the orders at low densities where the spectrum is broader. This does not give the optimum resolution for the high-density experiments, and consequently the instrumental contribution to the line widths is significant and limits the precision obtainable. The second and more serious limitation arises from the irregular scanning of the interferometer, which we believe is inherent in a Fabry-Perot interferometer that uses piezoelectric materials. Typically, one adjusts the interferometer such that the peak heights of injected laser light are equal for two or three orders. This does not guarantee that the finesse is constant at some other frequency. It was not unusual to observe considerable (~5%) variation in the Brillouin peak heights at the high densities, i.e., far outside the variation in the intensity of the laser (< 1%). This we attribute to irregularities in the finesse of the interferometer, arising from the piezoelectric crystals. A pressure-scanned spherical interferometer would probably be free of such difficulties, but one would need a set of interferometers for the rare gases. There is little merit in reporting values of y and E accurate to only 10%. However, with improvements it should be possible to obtain an accuracy of 1% or better for the transport coefficients. That would be worthwhile, particularly if one were interested in covering a wide range of temperatures and pressures.

Figures 2, 3, and 4 illustrate how well the computed theoretical profiles reproduce the

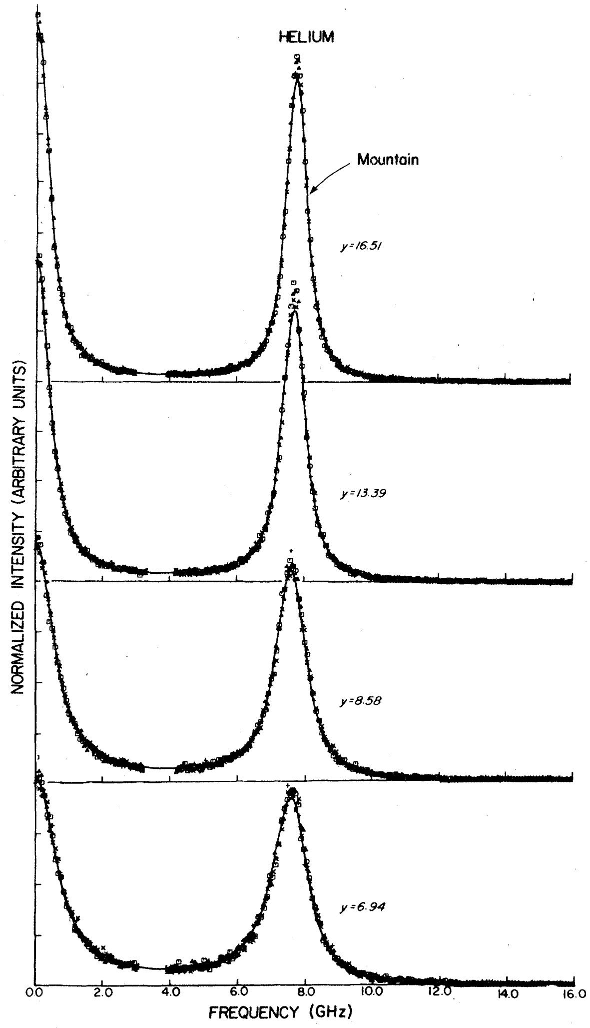

FIG. 2. A comparison of the experimental points with the computed hydrodynamic spectrum for He at several densities or y values. For clarity the experimental points have been omitted in places.

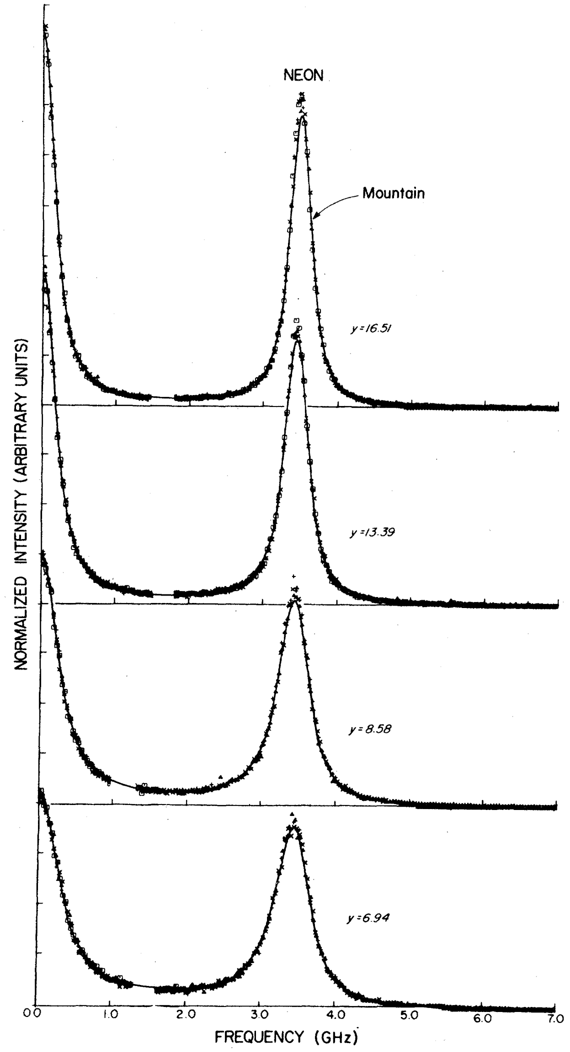

FIG. 3. A comparison of the experimental points with the computed hydrodynamic spectrum for Ne at several densities or y values. For clarity the experimental points have been omitted in places.

experimental curves. The "best-fit" values of C_0 and γ were used in the calculations while the values of the viscosity and thermal conductivity were taken from Sengers[20] and references therein. The density dependence of the thermal conductivity of He does not appear to have been measured. We used the low-density results of Kannuluik and Karman.[21] Except for the irregularity in the amplitude of the Brillouin component mentioned above, the agreement between experiment and theory is excellent. Overall, the present experiments show that an apparatus designed specifically to measure the Rayleigh-Brillouin spectra of gasses in the hydrodynamic domain will permit the precise determination of the density and temperature dependence of the adiabatic sound velocity, the specific heat ratio, the thermal conductivity, and the shear viscosity. [Here we assume that C_v and ρ_0, as required for a and b in Eq. (1), have been measured.] The availability of data on the density dependence of the transport coefficients might well stimulate further theoretical work which, at the moment, is limited to hard-sphere calculations.[15]

In this last section, the question of the value of y above which the hydrodynamic equations can be used to analyze data is explored. As this is a question of degree depending upon the precision of the experiment, the answer cannot be an exact value of y. The approach taken is to examine the low-density data and to compare the experimental

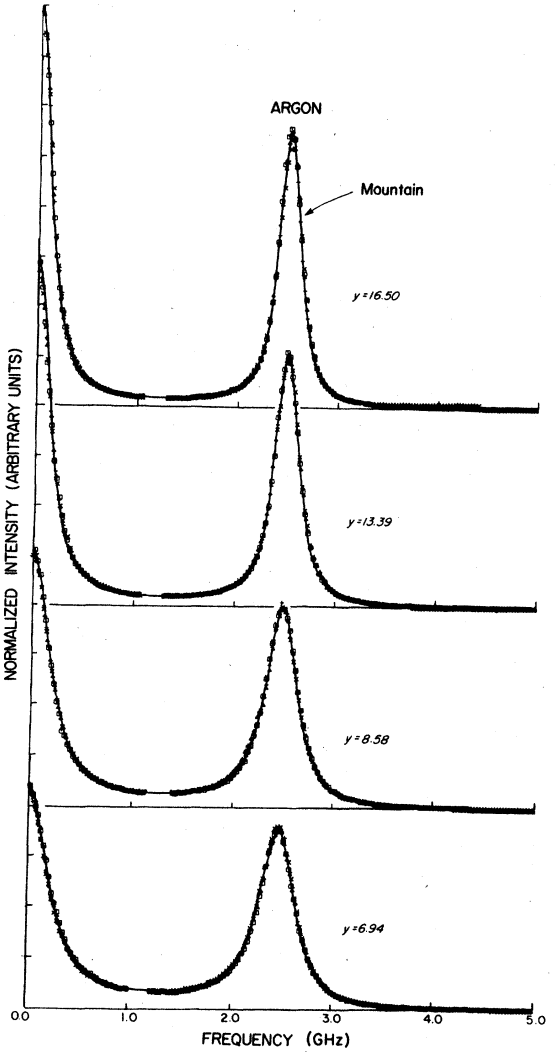

FIG. 4. A comparison of the experimental points with the computed hydrodynamic spectrum for Ar at several densities or y values. For clarity the experimental points have been omitted in places.

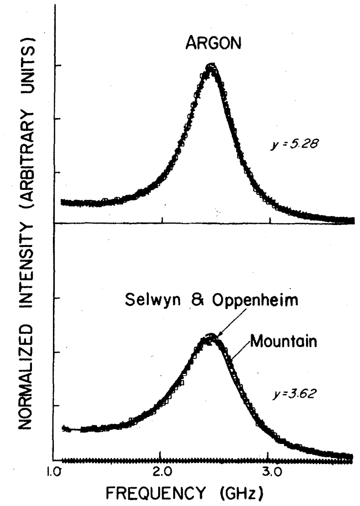

FIG. 5. A comparison of the experimental points for Ar in the neighborhood of the Brillouin component with the hydrodynamic and generalized hydrodynamic curves. The y values were chosen to show small departures from a purely hydrodynamic behavior.

results both with the hydrodynamic theory and with curves predicted by generalized hydrodynamics. In an attempt to extend hydrodynamic theory to higher ω and k values, Selwyn and Oppenheim[22] have given a formal expression for the spectrum in terms of certain correlation functions. In a second paper,[23] they work out the spectrum explicitly for an ideal Maxwell gas. To avoid complications arising from non-ideality in the comparison between theories and experiment, we use the results for Ar, since the velocity of sound at low pressures, given in Table I, are closest to the ideal-gas value for this gas. For the hydrodynamic calculations "accepted values" of the thermodynamic variables were used. For the generalized hydrodynamic calculations, essentially the same constants were used (Ar is nearly ideal in the range of pressures covered in the comparison), except that the thermal conductivity was omitted. It is well known that a Maxwell gas has a fixed Eucken ratio so that it is reasonable to utilize only the viscosity, the better-known transport coefficient.

Figure 5 shows the computed curves and the experimental points in the neighborhood of the Brillouin peak at y equal to 5.28 and 3.62. The differences between the various curves at lower frequency are too small to display. In Fig. 6

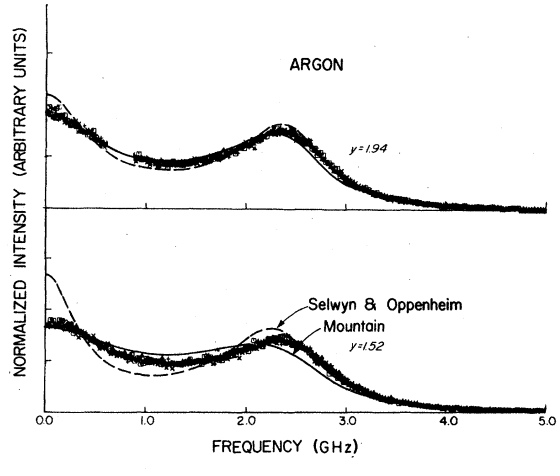

FIG. 6. A comparison of the experimental Rayleigh-Brillouin spectrum of Ar with hydrodynamic and generalized hydrodynamic curves. The y values correspond to the kinetic or nonhydrodynamic regime.

the lower frequency part is included for the two lowest values of y, 1.94 and 1.52. The difference between the two theoretical curves becomes smaller than the statistical fluctuation in the data at y equal to 6.94 and greater. Figures 5 and 6 show that differences between the two theories exist, but they rapidly decrease as one progresses toward y equal to 5 or 6. Thus we are justified in using hydrodynamic equations to measure the thermodynamic parameters at y equal to 6.94 and higher, as we have done in the first part of the paper. The same conclusion was reached by Clark.[24]

Some further comments may be made concerning Figs. 5 and 6. As the density decreases the Brillouin peak moves toward lower frequencies. Physically this occurs both because of the pulling which arises from the overlap with the central component and because the damping (width) of the Brillouin component increases; a damped oscillator has a lower frequency than the corresponding free oscillator. This feature is displayed by both theoretical curves and by the experimental spectra. However, as Selwyn and Oppenheim have pointed out, the position of the Brillouin peak for generalized hydrodynamics is at a higher frequency than that predicted by ordinary hydrodynamics. This can be seen in the figures. The important point to note is that the *experimental* Brillouin positions agree with those predicted by generalized hydrodynamics except at the lowest value of y. Thus our observations support the theoretical predictions of Selwyn and Oppenheim.

It must be remembered that generalized hydrodynamics retain only the next-highest terms in k and ω of an infinite series. At best it can hope to extend the range of hydrodynamics to values of y slightly lower than 6 or 7. The fact that smaller discrepancies occur at low y values, between the observed spectra and the curve computed for hydrodynamics, than for generalized hydrodynamics indicates that many more terms must be retained which somehow almost cancel, leaving the true spectrum close to that predicted by hydrodynamics alone. At still lower values of y it is known that hydrodynamic theory diverges rapidly from experimental profiles.

In part I we showed that the S6R model solution[2] gave an adequate description of the spectrum at low y values (< 6). Within the precision of our calcomp plotter the S6R theory duplicates the generalized hydrodynamic behavior at y equal to 3.62 and 5.28. As the S6R also follows the experimental curves at lower y values, it appears that it is a reasonable theory to use to extract thermodynamic parameters from spectra when the components begin to overlap. Experimentally one can skirt such problems at low densities by reducing the scattering angle. One drawback of the S6R theory its "ideal hydrodynamic" character. Mean-field effects have been included in a hard-sphere calculation by Furtardo *et al.*,[25] an area we intend to examine in a future publication. In summary, we have shown that thermodynamic parameters and transport coefficients may be determined from Brillouin spectroscopy of the compressed rare gases and have explored quantitatively the region of applicability of hydrodynamic theory.

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