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Dispersion of the Velocity of Sound in Xenon in the Critical Region*

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The sound velocity in xenon on the critical isochore was determined in Brillouin scattering measurements at scattering angles of 40° and 90°. The sound dispersion calculated from our data, ultrasonic data, and other Brillouin data agrees with the Kawasaki theory for a particular choice of parameters, but the dispersion in the Brillouin velocities is 2 to 3 times smaller than the theory predicts when values obtained for the parameters from other experiments are used.

The spectrum of light scattered by a fluid contains a pair of inelastic (Brillouin) components shifted symmetrically from the incident frequency by $\Delta\nu_B = vq/2\pi$, where $q = 2K_0 \sin\frac{1}{2}\theta$ (K_0 is the wave vector of the incident light in the medium and θ is the scattering angle). Near the critical point the sound velocity in the low-frequency limit, v_0, should approach zero as $(T-T_c)^{\alpha/2}$, where α is the exponent which describes the critical divergence in the specific heat c_v. For nonzero frequencies, however, the velocity may approach a nonzero limiting value as $T \to T_c$ due to dispersion associated with the coupling of sound waves to the critical density fluctuations, as first proposed by Fixman.[1]

The presence of a dispersion in the sound velocity of a simple fluid near the critical point was first reported many years ago by Chynoweth and Schneider,[2] whose measurements in xenon revealed a small but definite dispersion in the frequency range 0.25 to 1.25 MHz for $(T-T_c) \lesssim 2°C$. A large dispersion in the sound velocity was observed in recent Brillouin scattering experiments[3] in CO_2 (in the frequency range 420 to 800 MHz), and the temperature at which this dispersion occurred agreed qualitatively with the theory of Kadanoff and Swift,[4] which predicts a strongly temperature-dependent dispersion at a frequency $\omega_R = \chi\xi^{-2}$, where χ is the thermal diffusivity ($\chi \equiv \lambda/\rho c_p$, where λ, ρ, and c_p are the thermal conductivity, density, and constant-pressure specific heat, respectively) and ξ is the correlation length. The comparison of the observed dispersion with the Kadanoff-Swift prediction was made using values for χ and ξ determined in independent measurements of the Rayleigh linewidth.[5] Quantitative comparison of the Brillouin and ultrasonic data for CO_2 was complicated by the effects of molecular degrees of freedom.

Sound velocities. —We have performed Brillouin scattering measurements on xenon on the critical isochore at scattering angles of 40° and 90° with a Spectra-Physics model 119 stable, single-frequency, 6328-Å laser and a Tropel model 240 piezoelectrically scanned, 5-cm, confocal Fabry-Perot interferometer. For the temperature ranges investigated, the Brillouin shift frequency ranged from 158 to 189 MHz for $\theta = 40°$, and from 302 to 364 MHz for $\theta = 90°$. The sample cell geometry and thermostat are described in Henry, Swinney, and Cummins.[6] The sample used in the present experiment, filled with gas from a different lot from that used for our other xenon sample,[6] has a critical temperature of (16.59 ± 0.02)°C, in good agreement with the value of (16.606 ± 0.020)°C obtained for our other sample.

Sound velocities determined from our data are shown in Fig. 1 along with the Brillouin scattering results of Cannell and Benedek[7] for $\theta = 170°$ (the Brillouin shift frequency for these data, also obtained with 6328-Å laser excitation, ranged from 436 to 576 MHz), and the ultrasonic data of Garland, Eden, and Mistura[8] at 0.55 and 5 MHz. (To avoid confusion the data points are shown only for the $\theta = 90°$ data.) The associated attenuation data are discussed in a separate publication by Garland, Eden, and Mistura.[8] Cannell and Benedek, whose data were obtained with a high-resolution, high-contrast tandem interferometer, are also publishing a detailed analysis of their spectra separately.[7]

In this Letter we analyze the dispersion in the velocity data of Fig. 1, plus additional ultrasonic data at 1 and 3 MHz, in terms of recent theoretical predictions by Kawasaki.[9] The sound velocity dispersion was computed using the 0.55-MHz velocity data as v_0 for $(T - T_c) \geqslant 3$°C, but closer to the critical point the 0.55-MHz velocity data exhibited significant dispersion (e.g., 2.6% at $T - T_c = 0.5$°C; see Fig. 1), and hence, for this region it was necessary to compute v_0 from specific-heat data using the expression[10,11]

$$v_0^2 = MT/[c_v(1 - c_v/c_p)], \tag{1}$$

where $M = (\partial P/\partial T)_v^2/\rho^2$ was chosen so that $v_0 = v(0.55 \text{ MHz})$ at $T - T_c = 3$°C.

Kawasaki theory. —Kawasaki[9] has used his extended mode-mode coupling theory to investigate

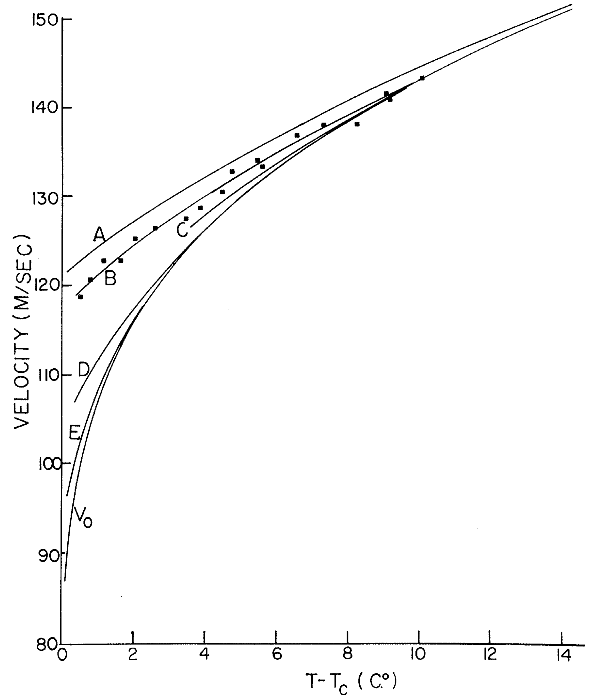

FIG. 1. The velocity of sound in xenon on the critical isochore. Brillouin data: A, $\theta = 170°$ (Cannell and Benedek, Ref. 7); B, $\theta = 90°$, and C, $\theta = 40°$ (present experiment. Ultrasonic data: D, 5 MHz, and E, 0.55 MHz (Garland, Eden, and Mistura, Ref. 8). Static velocity v_0: computed from thermodynamic data for $T - T_c < 3$°C; from 0.55-MHz data for $T - T_c \geqslant 3$°C. The data points shown are for the $\theta = 90°$ Brillouin data.

the coupling of the sound mode to other modes, and he has obtained the following closed expression for the sound velocity dispersion[12]:

$$\frac{v(\omega) - v_0}{v_0} = \frac{k_B T^3 (1 - \eta/2)^2}{2\pi^3 v_0^2 \rho^3 c_v^2 \xi} \left(\frac{\partial P}{\partial T}\right)_v^2 \times \left(\frac{\partial \xi^{-1}}{\partial T}\right)_s^2 I\left(\frac{\omega}{\omega_R}\right), \tag{2}$$

where $I(\omega/\omega_R)$ is given by

$$I\left(\frac{\omega}{\omega_R}\right) = \int_0^\infty \frac{x^2}{(1 + x^2)^2} \left[\frac{1}{1 + 4(\omega/\omega_R)^{-2} K^2(x)}\right] dx,$$

with

$$K(x) = \tfrac{3}{4}[1 + x^2 + (x^3 - x^{-1}) \arctan x].$$

In these expressions $v(\omega)$ is the sound velocity measured at frequency ω (which for Brillouin scattering is the Brillouin shift frequency $\Delta\omega_B$), k_B is Boltzmann's constant, and η is the exponent which describes the departure from Ornstein-Zernike behavior ($\eta = 0.06 \pm 0.06$). In the mode-

mode coupling theory the thermal diffusivity has the same critical behavior as the reciprocal of the correlation length: $\chi \sim \xi^{-1}$. Since $\xi = \xi_0 \epsilon^{-\nu}$ [where $\epsilon \equiv (T-T_c)/T_c$], we can write $\chi = \chi_0 \epsilon^{\nu}$ and thereby obtain $\omega_R \equiv \chi \xi^{-2} = \chi_0 \xi_0^{-2} \epsilon^{3\nu}$.

Immediately after Kawasaki reported his result for the sound velocity dispersion, Barmatz tested the dispersion formula (2) with his ultrasonic data for the sound velocity in He^4 in the range 1.5-50 kHz.[13] For that comparison ξ_0 and an overall multiplicative constant were taken as adjustable parameters. The predicted temperature dependence of the velocity was found to agree well with the data, but the theory was found to underestimate the magnitude of the dispersion.

For our comparison of the Kawasaki theory with the xenon velocity data it is convenient to rewrite Eq. (2) using the thermodynamic relations,

$$\frac{T(\partial P/\partial T)_v^{\,2}}{v_0^{\,2}\rho^3 c_v^{\,2}} = \frac{1-c_v/c_p}{\rho c_v},$$

$$(\partial \xi^{-1}/\partial T)_s = (\partial \xi^{-1}/\partial T)_\rho,$$

where the latter expression is an approximation valid over the range of temperatures of interest here. We now define a "reduced dispersion" (D_R) which depends only on experimentally determined quantities:

$$D_R \equiv \frac{[v(\omega)-v_0]/v_0}{F(T)}, \tag{3}$$

with

$$F(T) = \frac{k_B T^2 (1-\eta/2)^2}{2\pi^2 \rho c_v \xi}\left(1-\frac{c_v}{c_p}\right)\left(\frac{\partial \xi^{-1}}{\partial T}\right)_\rho^2. \tag{4}$$

Now the prediction of the Kawasaki theory is that the reduced dispersion is equal to the function $I(\omega/\omega_R)$. The D_R and the scaled frequency ω/ω_R are both functions of temperature and frequency, but in such a way that, according to the theory, all the data at different temperatures and frequencies should fall on a single universal curve.

Data analysis.—Recent Rayleigh linewidth measurements by Henry, Swinney, and Cummins[6] on xenon have determined χ_0, ν, and ξ_0 and, as mentioned previously, the specific heats c_v and c_p are known for xenon,[10,11,14] so the Kawasaki theory can be compared directly with the velocity data with no adjustable parameters. The velocity data extend three orders of magnitude in ω and over more than one order of magnitude in ϵ, so these data should serve as a thorough test of the theory.

The experimental results for the reduced dispersion are compared with Kawasaki's theory [the integral for $I(\omega/\omega_R)$ was evaluated numerically] for two sets of the parameters χ_0, ν, ξ_0 in Fig. 2[15] (for these plots we have taken $\eta = 0$). In Fig. 2(a) the values of the parameters are those determined in measurements of the Rayleigh linewidth[6]: $\chi_0 = 6.94\times10^{-4}$ cm^2/sec, $\nu = 0.751$, and $\xi_0 = 0.606$ Å (a later best-fit value of ξ_0 to the linewidth data was slightly lower, $\xi_0 = 0.58$ Å). The ultrasonic data are seen to be in reasonable agreement with theory, but the Brillouin scattering values for the reduced dispersion are approximately 5 times smaller than the theoretical values.

The parameters χ_0, ν, and ξ_0 were all varied over a large range (ν from 0.40 to 0.76) in an attempt to improve the fit of the data to the theory. Both ω_R and $F(T)$ have a temperature dependence which is sensitive to the value of ν: $\omega_R \sim \epsilon^{3\nu}$, and $F(T)$ is a monotone increasing function for $\nu \gtrsim 0.6$,

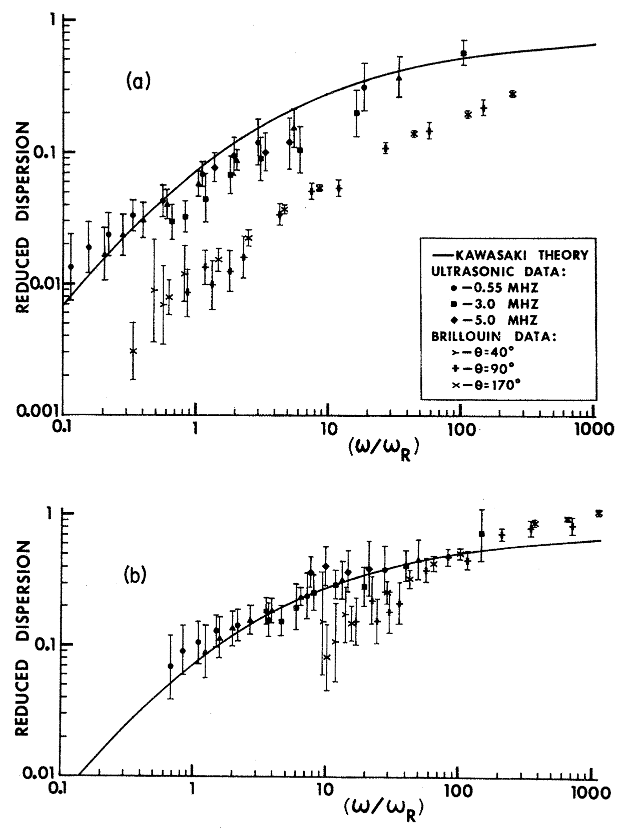

FIG. 2. The reduced sound dispersion in xenon, scaled according to Kawasaki's theory: (a) with parameters from Rayleigh linewidth measurements (Ref. 6), $\chi_0 = 6.94\times10^{-4}$ cm^2/sec, $\nu = 0.751$, and $\xi_0 = 0.606$ Å; (b) with the parameters which give the best overall fit to the data, $\chi_0 = 0.64\times10^{-4}$ cm^2/sec, $\nu = 0.54$, and $\xi_0 = 2.94$ Å.

and monotone decreasing for $\nu \lesssim 0.6$.[16] The result for the set of parameters which produced the best agreement between the theory and all the velocity data is shown in Fig. 2(b). Here the Brillouin and ultrasonic data are both in fair agreement with the theory, and the values used for ν and ξ_0, $\nu = 0.54$ and $\xi_0 = 2.94$ Å, agree fairly well with those obtained from measurements of the angular dependence of the linewidth and the scattering intensity for xenon[17]; however, this value for ν and the value used for χ_0, $\chi_0 = 0.64 \times 10^{-4}$ cm^2/sec, are in complete disagreement with the results from Rayleigh linewidth measurements of the thermal diffusivity.[6]

In the mode-mode coupling theory[4,9] the critical behavior of both χ and ξ is characterized by the exponent ν, but the exponent $\gamma - \psi$ obtained for χ in Rayleigh linewidth measurements, $\gamma - \psi = 0.751 \pm 0.004$, is significantly greater than the generally accepted value for ν, $\nu = 0.64 \pm 0.03$ (see discussion in Ref. 6). However, Garland, Eden, and Mistura[8] have derived Kawasaki's results for the sound dispersion[18] and attenuation without the assumption that χ and ξ are characterized by the same exponent. In this case ω_R is given by $\omega_R = \chi_0 \xi_0^{-2} \epsilon^{\gamma - \psi + 2\nu}$. We have calculated D_R[19] with $\gamma - \psi \neq \nu$, using values obtained for the parameters from other experiments: $\chi_0 = 6.94 \times 10^{-4}$ cm^2/sec, $\gamma - \psi = 0.751$, $\eta = 0.06$, $\xi_0 = 1.40$ Å,[17] and $\nu = 0.64$; for these parameters we have $\omega_R = (3.54 \times 10^{12}) \epsilon^{2.03}$ sec^{-1}. The resultant plot of the reduced dispersion is similar to Fig. 2(a); however, for this case D_R for the Brillouin data is 2 to 3 rather than 5 times smaller than the theory predicts.

The good agreement between the ultrasonic velocity data and the Kawasaki theory is paralleled by a similar agreement between the theory and the sound attenuation data of Garland, Eden, and Mistura[8]; on the other hand, while we find that the Brillouin sound-velocity dispersion is 2 to 3 times smaller than the theory predicts when reasonable values are used for the thermal diffusivity, Garland, Eden, and Mistura[8] found that the Brillouin sound attenuation data of Cannell and Benedek,[7] when plotted as a function of the scaled frequency $\omega/2\omega_R$, fall on the same curve as the ultrasonic attenuation data. In their comparison of the theory with experiment, Garland, Eden, and Mistura took $F(T)$ as a constant, $F(T) = 0.30$ [in terms of their B, $F(T) = B/2\pi$], and $\omega_R = (2.69 \times 10^{12}) \epsilon^2$ sec^{-1}. This value "2" for the critical exponent for ω_R is obtained if, for example, the mode-mode coupling result $\gamma - \psi = \nu$ is combined with the assumption that $\nu = \frac{2}{3}$ (see Ref. 8), but for this value of ν, $F(T)$ has a significant temperature dependence.[16] On the other hand, we have found that if $\nu = 0.62$, then $F(T)$ is essentially independent of temperature over the temperature range of interest, and, moreover, this value of ν, together with the parameters $\chi_0 = 7.58 \times 10^{-4}$ cm^2/sec, $\gamma - \psi = 0.76$, $\eta = 0.12$, and $\xi_0 = 1.68$ Å,[17] yields $F \approx 0.30$ and $\omega_R = (2.69 \times 10^{12}) \epsilon^2$ sec^{-1}, as in Ref. 8. This set of parameters, in good agreement with other experiments, is nearly the same as those used in our data analysis discussed in the preceding paragraph. Thus, by following the Garland, Eden, and Mistura data-analysis procedure, we again find that the dispersion in the ultrasonic velocities is in agreement with the theory, but the dispersion for the Brillouin velocity data is 2 to 3 times smaller than the theory predicts.

Discussion. —Although we have shown that there exists a set of parameters which brings both the ultrasonic and Brillouin velocity data into agreement with the Kawasaki theory, it is disturbing that this requires a thermal diffusivity which is in complete disagreement with that obtained in Rayleigh linewidth measurements. For reasonable values of the thermal diffusivity and the other parameters, the ultrasonic velocity data and both the ultrasonic and Brillouin attenuation data all appear to be in good agreement with the theory, while the observed dispersion in the Brillouin velocities is one-half to one-third that predicted by the theory. Three possible explanations of this discrepancy have been proposed[20]:

(1) Perhaps the high-frequency data do not scale with ω/ω_R. On the other hand, the Brillouin data do fall on a single curve, so apparently ω_R is the relevant scale frequency.

(2) As Mountain and others have pointed out, the information obtained from Brillouin scattering experiments (where q is real and ω is complex) is not necessarily to be interpreted in the same way as the information obtained from ultrasonic experiments (where ω is real and q is complex).[21] However, the difference between the results of the two types of experiments is expected to be significant only when the sound dispersion and attenuation are large, while for the data presented here the largest discrepancy between the ultrasonic and Brillouin results occurs for the smallest values of the dispersion, $[v(\omega) - v_0]/v_0 \lesssim 0.04$.

(3) The range of scaled frequencies ω/ω_R for which the discrepancy between the ultrasonic and

Brillouin data is most serious, $0.1 \lesssim \omega/\omega_R \lesssim 100$, corresponds to different temperature ranges for the two sets of data: $0.1 \lesssim T-T_c \lesssim 0.8$°C for the ultrasonic data, and $0.8 \lesssim T-T_c \lesssim 10$°C for the Brillouin data, where the $T-T_c \approx 0.8$°C crossover temperature corresponds to $\omega/\omega_R \sim 0.4$ for the ultrasonic data and to $\omega/\omega_R \sim 80$ for the Brillouin data. Thus if the static sound velocities v_0 were correct for $T-T_c \lesssim 0.8$°C but were high by an amount increasing to several percent at the higher temperatures, then this would explain the observed discrepancy; however, the 0.55-MHz sound velocity data of Garland, Eden, and Mistura, used for v_0 for $T-T_c \geq 3$°C, are believed to be accurate within 1% (see Refs. 8 and 15).

Note added in proof. —The third explanation above seems most likely, since values for v_0 calculated using the Habgood and Schneider[11] data for $(\partial P/\partial T)_v$ in Eq. (1) are approximately 5.5% lower than the values used in the above analysis. The Brillouin dispersion calculated using these lower values for v_0 is in reasonable agreement with the Kawasaki theory.

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[10]The data used for the constant-volume specific heat c_v were those of C. Edwards, J. A. Lipa, and M. J. Buckingham, Phys. Rev. Lett. 20, 496 (1968). We thank Dr. M. Barmatz for sending us tables of these data.

[11]Values of the specific heat ratio c_v/c_p (which was small but non-negligible over the temperature range of the sound-velocity measurements—e.g., $c_v/c_p < 0.02$ for $T-T_c < 3$°C, and $c_v/c_p < 0.1$ for $T-T_c < 20$°C) were obtained by combining the results of Habgood and Schneider with those of Michels *et al.*: H. W. Habgood and W. G. Schneider, Can. J. Chem. 32, 164 (1954); A. Michels, T. Wassenaar, G. J. Wolkers, and J. Dawson, Physica (Utrecht) 22, 17 (1956).

[12]See Kawasaki, footnote 15 in the first paper in Ref. 9.

[13]M. Barmatz, Phys. Rev. Lett. 24, 651 (1970).

[14]The c_v data of Ref. 10 extend from near the critical point to $T-T_c = 6.4$°C; for larger values of $T-T_c$ the c_v data of Ref. 11 were used.

[15]The error bars in Fig. 2 reflect only the uncertainty in $v(\omega)$ and not that in v_0. The assumed uncertainties in the Brillouin data were 2 m/sec for $\theta = 40°$, 1.5 m/sec for $\theta = 90°$, and 1 m/sec for $\theta = 170°$ (Dr. Cannell has informed us that the absolute values of his velocities are actually uncertain by 1.5 m/sec). For the ultrasonic data the uncertainty was 1% for most of the data, but ranged up to 5% or more when the attenuation was large (private communication).

[16]For example, if $T-T_c$ is increased from 0.1°C to 10°C, then $F(T)$ increases by a factor of 8.2 for $\nu = 0.75$ and increases a factor of 2.6 for $\nu = \frac{2}{3}$, while $F(T)$ decreases by a factor of 2.3 for $\nu = 0.54$.

[17]We computed the correlation-length coefficient ξ_0 for different values of the exponent ν using the expression $\xi_0 = \xi\epsilon^{\nu}$ along with the result $\xi = 182$ Å at $\epsilon = 5\times10^{-4}$; this value for ξ was observed both in Rayleigh linewidth measurements by Henry, Swinney, and Cummins, Ref. 6, and in scattering intensity measurements by M. Giglio and G. B. Benedek, Phys. Rev. Lett. 23, 1145 (1969).

[18]In the notation of Ref. 8, the sound dispersion is given by $[v(\omega)-v_0]/v_0 = (1-\gamma)\Delta'(\omega)/2c_p$, where $\Delta'(\omega)$ is the real part of $\Delta(\omega)$.

[19]The theoretical result for D_R, the integral $I(\omega/\omega_R)$, is fairly insensitive to the form of $K(x)$: For $(\omega/\omega_R) < 100$, the Fixman form of $K(x)$, $K(x) = x^2(1+x^2)$, yields values for I which are within 16% of the values obtained using Kawasaki's $K(x)$

[20]For all the data presented in this paper, the inequalities $\omega \ll v\xi^{-1}$ and $q\xi \ll 1$ are both satisfied.

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