

Measurement of the long-range correlation length of SF₆ very near the critical point*

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Results are reported of measurements of the long-range correlation length ξ , made along the critical isochore. The measurements were made using two different light-scattering techniques and cover the temperature range $4.0 \times 10^{-6} \leq T/T_c - 1 \leq 3.1 \times 10^{-2}$. All of the results are accurately fit by the expression $\xi = 2.016(T/T_c - 1)^{-0.621}$ Å. Measurements of the isothermal compressibility κ_T , in the temperature range $1.5 \times 10^{-4} \leq T/T_c - 1 \leq 3.1 \times 10^{-2}$ are also reported and yield the result $\kappa_T = 1.327 \times 10^{-9} (T/T_c - 1)^{-1.223}$ cm²/dyn. From these measurements the value of the exponent η may be deduced as 0.03 ± 0.03 . The measured values of ξ are used to interpret existing measurements of the Rayleigh linewidth in terms of mode-mode coupling theories. Experimental results are also presented which indicate that stirring may be used to eliminate gravitationally induced density gradients near the critical point without affecting the fluid's critical properties.

INTRODUCTION

As a pure fluid is brought into the vicinity of its gas-liquid critical point the spatial range over which spontaneous density fluctuations are correlated grows very large. This effect was first treated theoretically by Ornstein and Zernike,¹ who found that for large $|\vec{r} - \vec{r}'|$ the equal-time density-density correlation function $G(\vec{r}, \vec{r}') \equiv \langle \delta\rho(\vec{r})\delta\rho(\vec{r}') \rangle$ was proportional to $\rho e^{-|\vec{r} - \vec{r}'|/\xi} / R^2 |\vec{r} - \vec{r}'|$, where $\delta\rho(\vec{r})$ is the difference between the density at the point \vec{r} , and the average density ρ , and the brackets denote an ensemble average. The long-range correlation length ξ increases without limit as the critical point is approached, while by assumption the direct correlation range R remains small even at the critical point.

The angular distribution of the intensity of scattered light corresponding to the Ornstein-Zernike form of the correlation function is given by

$$I(\vec{q}) = A \kappa_T \sin^2 \phi / (1 + q^2 \xi^2), \quad (1)$$

where ϕ is the angle between the incident electric field and the wave vector of the scattered light, and \vec{q} is the scattering wave vector whose magnitude is given by $2k_0 \sin(\theta/2)$, where k_0 is the wave vector of the incident light *in the fluid*, and θ is the scattering angle. The prefactor A is $(\pi^2 k_B T / \lambda_0^4) (\rho \partial n^2 / \partial \rho)_T$, where λ_0 is the vacuum wavelength of the incident light, ρ is the fluid density, and n the refractive index. It is known on theoretical grounds that the Ornstein-Zernike form of the correlation function is not rigorously correct,² and Fisher and Burford have calculated more exact forms for the angular dependence of the scattered intensity for the three-dimensional Ising model.³ The more exact correlation function differs from the Ornstein-Zernike form in

that it decays asymptotically at large r as $r^{-(1+\eta)}$, where the new exponent η is ~ 0.056 for the three-dimensional Ising model.³ The more exact scattering function calculated in Ref. 3 differs only very slightly from the Ornstein-Zernike result numerically and so I have used the simpler form of Eq. (1) to analyze my data. This is also justified on experimental grounds since direct observation has confirmed the correctness of Eq. (1) to within a few percent for pure fluids.^{4,5}

DIFFERENTIAL INTENSITY MEASUREMENTS

Accurate determination of the magnitude and temperature dependence of the correlation range by direct measurement of the angular dependence of the scattered intensity has proved to be rather difficult since ξ is only a few Å well away from the critical point but increases to many thousands of Å near the critical point. This difficulty may be overcome by using a very accurate differential technique⁶ to measure directly the difference in the intensities of light scattered in the forward direction and in the backward direction. The ratio of the difference $I_F - I_B$ to the forward scattered intensity I_F is a direct measure of ξ . The apparatus used for these measurements has been described previously,⁶ and the interested reader is referred there for practical details. For the present experiments the angle for forward scattering was set to 8.5° and that for backward scattering to 171.5° , as measured in the fluid. The cell was filled with SF₆ of 99.99% purity, and the density was adjusted to be within 0.1% of the critical density by measuring the meniscus height as a function of temperature and releasing gas from the cell as required. The meniscus height measurements yielded the value 45.500 K for the critical temperature T_c , and this was confirmed

to within 0.5 mK by observation of the reappearance of the meniscus upon cooling. The cell temperature was controlled by immersing it in circulating isopropyl alcohol, the temperature of which was controlled using a thermistor sensor and a combined proportional and integral feedback control system to heat the alcohol. The cell was further isolated from the room by an outer aluminum box controlled to within 0.1 K by a commercial circulator. A second thermistor and sensing electronics was used to evaluate the performance of this controller, and it was found to maintain the interior of the cell constant in temperature and isothermal to within 50 μ K for a period of 72 h. No longer duration test was made because there was no indication of drift. All measurements to be described were made at the critical density for temperatures greater than T_c .

Since the method used to take and analyze data was somewhat different from that presented in Ref. 6, I will describe it briefly. The quantity measured at either angle is not the actual intensity, but a current proportional to it. The proportionality constants include effects such as cell window transmittances, collection solid angles, etc., and in general the proportionality constants are not identical for the two optical systems used to collect the forward and backward scattered light. The constants were adjusted to be equal by raising the cell to temperatures greater than $T_c + 25$ K and adjusting the collection solid angles of the two optical systems so as to have a measured intensity difference independent of temperature. Since at these temperatures the

scattered intensity is temperature dependent, but the correlation length is so short it produces no measurable asymmetry in the scattered intensity, this adjustment resulted in equal proportionality constants for the two collection systems. This adjustment was periodically checked throughout the course of data taking, and the measured intensity difference at $T_c + 25$ K typically remained stable to within 0.03% of the intensity measured by either collection system alone. The small residual asymmetry was attributed to stray elastically scattered light. At any given temperature the quantity measured was the ratio of the intensity difference $I_F - I_B$ to the forward scattered intensity I_F , which is a direct measure of the correlation length at that temperature. This method of calibrating the two optical systems eliminated the need for the more involved data analysis carried out in Ref. 6. In this manner correlation range measurements were made over the temperature range $1.5 \times 10^{-4} \leq \epsilon \leq 3.1 \times 10^{-2}$. These measurements were made with the cell in thermal equilibrium, and the data were analyzed using Eq. (1).

The values of ξ determined in this manner are shown as open circles in Fig. 1. Weighting each point equally by percent, the best fit to these data is $\xi = 72.46(T - T_c)^{-0.6215}$, which may also be expressed as $\xi = 2.015\epsilon^{-0.6215} \text{ \AA}$, where $\epsilon = (T/T_c - 1)$. These results can be compared to those of Puglielli and Ford⁷ who, using a different technique which will be discussed below, measured ξ in the range $1.2 \times 10^{-4} \leq \epsilon \leq 1.4 \times 10^{-3}$ and found $\xi = (72 \pm 11)(T - T_c)^{-0.67 \pm 0.07} \text{ \AA}$ or $\xi = (1.5 \pm 0.23) \times \epsilon^{-0.67 \pm 0.07} \text{ \AA}$. Although my value for the prefactor

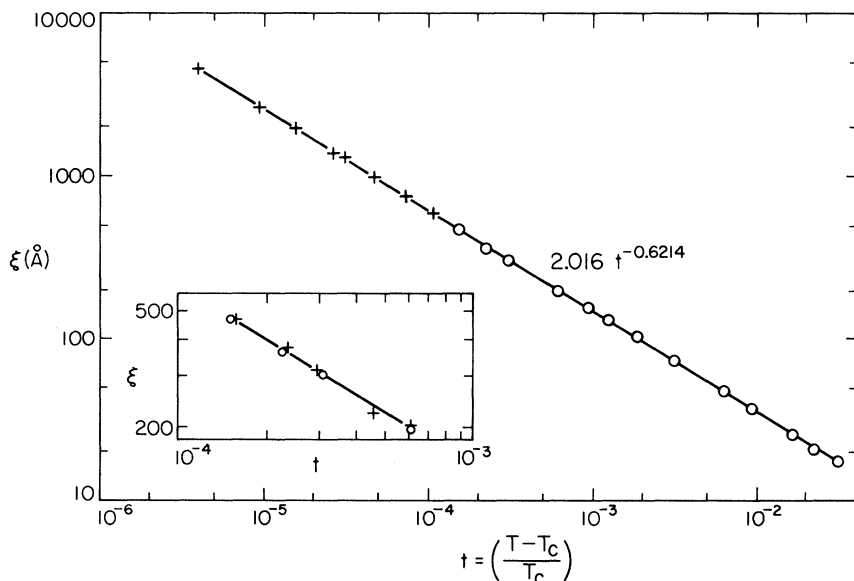


FIG. 1. Long-range correlation length of SF_6 as a function of temperature on the critical isochore. The circles are the results of differential intensity measurements, and the crosses are values determined from turbidity and compressibility measurements. The inset compares the results in the temperature range where both methods could be used.

lies well outside their error bars for the prefactor in the expression involving ϵ , the actual measured values of ξ agree to within 15% throughout the entire range where they were able to make measurements. The discrepancy is due to the fact that in computing the error bars for the prefactor ξ_0 in the expression $\xi = \xi_0 \epsilon^{-\nu}$ they did not include the effect of errors in ν . Inclusion of these errors results in the more reasonable value of $(1.5 \pm 0.6) \times \epsilon^{-0.67 \pm 0.07}$. From a practical viewpoint, expressions of the form $\xi_0 \epsilon^{-\nu}$ are a poor representation of data in the critical region ($\epsilon \lesssim 10^{-2}$) because the point $\epsilon = 1$ lies well outside the region of interest. For this reason a given set of data can be represented quite well by a large range of choices for ξ_0 and ν , the choices being highly correlated. This effect is much less noticeable if expressions of the form $\xi'_0 (T - T_c)^{-\nu}$ are used, since $T - T_c = 1$ K lies well within the experimentally accessible region for room temperature experiments. Consequently I have presented my results in both forms.

In attempting to extend these measurements to values of ϵ less than 1.5×10^{-4} two major experimental difficulties were encountered. The first difficulty arises because the fluid becomes such an intense scatterer that a noticeable fraction of the light collected by either optical system is the result of two or more scatterings. Since the intensity of this multiply scattered light does not have the same angular distribution as does that of the singly scattered light, it makes measurements very difficult to interpret.⁸ The second difficulty arises because extremely large values of the isothermal compressibility κ_T are assumed by a pure fluid in the vicinity of its critical point; and if maintained at constant and uniform temperature the fluid eventually reaches an equilibrium state in which the density varies quite rapidly with height in the cell. The equilibrium density profile is that which generates the excess hydrostatic pressure required to support the fluid in the earth's gravitational field. Any measurement which determines the properties of a fluid sample of finite height must of necessity sample a range of fluid densities and this can result in substantial errors.⁹ Since optical measurements can be made in volumes of very small vertical dimension, they might seem to be insensitive to this problem, and indeed light scattering has proved a powerful tool for the study of such systems. Unfortunately, in the immediate vicinity of the critical point the gravitationally induced density gradients result in refractive index gradients so strong as to cause the incident and scattered light beams to bend sharply downward. Experimenters have occasionally attempted to eliminate density grad-

ients by stirring the fluid, but the validity of this procedure has never been clearly established.

THE EFFECTS OF STIRRING

In the course of making the measurements described above I noticed that to within experimental error of ~1% the same results were always obtained when the fluid had been thoroughly stirred by convection or when it had been allowed to reach thermal equilibrium. The measurements in both cases were made in the center of the cell, which is the height where maximum opalescence occurred and also the position where the meniscus reappeared upon cooling. In order to make a more stringent test of these observations the cell was allowed to reach thermal equilibrium at a temperature 12 mK above T_c as evidenced by the scattered intensity vs height in the cell becoming time independent. Accurate measurements of the forward and backward scattered intensities were made and the fluid was then stirred by heating the cell a few degrees and cooling to $T_c + 0.012$ K in the interval of a few minutes. This caused rapid convection which ceased after a few more minutes and the fluid was left in a state where the density appeared quite uniform throughout most of the cell. The only visible density gradients occurred at the extreme top and bottom of the cell, the central 1.6-cm portion of the 2-cm-high cell being uniform. The intensity measurements were then repeated and found to agree to within 0.6% with those made at equilibrium. I estimate that at this temperature ~10% of the collected light was due to multiple scattering, and this was apparently not changed by stirring.

The stirring caused by convection left the fluid in a state of uniform density throughout most of the cell, and thus the temperature of the fluid in that portion of the cell must have been non-uniform to the extent required to generate the excess pressure needed to support the fluid. Since $(\partial P / \partial T)_V$ is well behaved throughout the critical region,¹⁰ being $\sim 8 \times 10^5$ dyn/cm² K, the required temperature gradient is 0.9 mK/cm. Since the incident beam was less than 0.05 cm in diam the scattering volume was isothermal to within 45 μ K. The exact temperature profile assumed by the fluid upon the cessation of convection would appear to be difficult to calculate, but it should be noted that the results obtained after stirring were always the same and did not depend on the amount by which the cell temperature was raised or the speed with which it was lowered provided only that sufficient convection occurred to leave the density uniform. On the basis of these observations I used convective

stirring to eliminate refractive index gradients in making turbidity measurements for values of ϵ less than 1.5×10^{-4} .

SCATTERED INTENSITY AND TURBIDITY MEASUREMENTS

The problem of multiple scattering is more fundamental than that of density gradients, but one quantity which can be measured accurately even in the presence of severe multiple scattering is τ , the attenuation per unit length suffered by the incident beam in passing through the fluid. As Puglielli and Ford have shown, τ departs from simple power law behavior near the critical point, and this departure can be used to measure ξ .⁷ As shown in Ref. 7, the integral of Eq. (1) over all scattering angles yields

$$\tau = \pi A \kappa_T f(\alpha), \quad (2)$$

where

$$f(\alpha) = \left(\frac{2\alpha^2 + 2\alpha + 1}{\alpha^3} \right) \ln(1 + 2\alpha) - \frac{2(1 + \alpha)}{\alpha^2}, \quad (3)$$

with $\alpha = 2(k_0 \xi)^2$. Since $f(\alpha)$ is very nearly constant for small α , τ exhibits the same power law behavior as does κ_T well away from the critical point. Puglielli and Ford relied on this fact and used the behavior of τ for $T - T_c \geq 0.5$ K to obtain a best power law fit to κ_T and extrapolated this fit to obtain κ_T for $T - T_c < 0.5$ K. Then, by measuring τ in the range $0.038 \leq T - T_c \leq 0.45$ K, they were able to obtain ξ from Eq. (2) in the same range. However, κ_T can in fact be directly and accurately measured to within $T - T_c = 0.048$ K by measuring the forward scattered intensity. This reduces the range over which the values of κ_T must be obtained by extrapolation, and thus improves considerably the accuracy with which ξ can be determined by this method.

In order to measure the actual values of κ_T by measuring the photocurrent caused by the forward scattered light, the constant of proportionality between the photocurrent and the forward scattered intensity must be determined. This was achieved in the following manner. The photocurrent was measured at several different temperatures in the range $0.6 \leq T - T_c \leq 10.0$ K, and the quantity A was evaluated at each of these temperatures, using the Lorentz-Lorenz relationship and the measured value¹¹ of n to evaluate $(\rho \partial n^2 / \partial \rho)_T^2$. Since $q^2 \xi^2 \ll 1$ for forward scattering in this temperature range, these measurements determine the ratio of κ_T at any two temperatures studied. The apparatus for measuring τ , which is described in Ref. 6, was then used to determine the differences in the val-

ues of τ for these same temperatures. For each pair of temperatures thus studied, the measured ratio of κ_T and the measured difference of τ were used in Eq. (2) to compute the actual value of κ_T and τ at that temperature. In so doing the values of ξ already determined by the differential intensity measurements were used to evaluate $f(\alpha)$ at each temperature. It should be noted that in this temperature range α is quite small and $f(\alpha)$ is essentially constant and independent of ξ . For this reason the values of κ_T and τ determined in this manner are almost completely independent of the previous measurements of ξ . Having thus determined the constant of proportionality between the measured photocurrent and the forward scattered intensity, it was possible to measure κ_T in the temperature range $0.048 \leq T - T_c \leq 10.0$ K, where there was no indication of multiple scattering. These measurements were made with the fluid in thermal equilibrium. The resulting values are accurately fit by the expression $\kappa_T = 1.529 \times 10^{-6} (T - T_c)^{-1.223}$ cm²/dyn or $\kappa_T = 1.327 \times 10^{-9} \times (T/T_c - 1)^{-1.223}$ cm²/dyn, the rms deviation of the data points from the fit being 0.8%. The value of the exponent γ is in excellent agreement with the results of Puglielli and Ford⁷ who found $\gamma = 1.225$, and with those of Feke *et al.*,¹² who found $\gamma = 1.235$. An independent determination of the magnitude of κ_T has been made by Puglielli and Ford, and another value can be deduced from the PVT measurements of MacCormack and Schneider.¹⁰ By way of comparison my value for κ_T at $\epsilon = 4.56 \times 10^{-3}$ lies 1.4% below that determined from Ref. 10 and 10% above that of Ref. 7. This is the largest value of ϵ for which the PVT measurements were made and should yield the most accurate result, measurements nearer T_c being more affected by density gradients.

The measurements used to determine the constant of proportionality between the photocurrent and the forward scattered intensity also served to determine the actual values of τ for several temperatures in the range $T - T_c \geq 0.6$ K. These results were used to calibrate the apparatus for measuring τ so as to be able to measure directly the actual value of τ at any temperature desired rather than merely the difference in τ at any two temperatures. Measurements of τ were then made for a number of temperatures in the range $4.0 \times 10^{-6} \leq \epsilon \leq 6.1 \times 10^{-4}$, the fluid being allowed to reach thermal equilibrium for $\epsilon \geq 1.5 \times 10^{-4}$ and being thoroughly stirred by convection for $\epsilon < 1.5 \times 10^{-4}$. For each of these temperatures the value of κ_T was calculated from the best fit to the measured values of κ_T , and Eq. (2) was then used to obtain ξ from τ and κ_T . The resulting values of ξ are shown as crosses in Fig. 1. The values ob-

tained for $\epsilon < 1.5 \times 10^{-4}$ depend on extrapolating the best fit to κ_T . In the temperature range $1.5 \times 10^{-4} \leq \epsilon \leq 6.1 \times 10^{-4}$ the results can be compared directly with values obtained by the differential intensity method as shown in the inset in Fig. 1. The best fit to the values of ξ determined from the measurements of τ and κ_T is $\xi = 72.34(T - T_c)^{-0.6216}$ or $\xi = 2.011\epsilon^{-0.6216}$ Å, in essentially perfect agreement with the result determined by the differential intensity measurements. The best fit to the combined data is $\xi = 72.44(T - T_c)^{-0.6214}$ Å or $2.016 \times \epsilon^{-0.6214}$ Å.

In the interest of clarity the actual sequence of calibrations and measurements performed is summarized here. First, the differential intensity apparatus was calibrated so as to yield a direct measurement of ξ at any temperature desired, and ξ was measured in the temperature range $1.5 \times 10^{-4} \leq \epsilon \leq 3.1 \times 10^{-2}$. Second, both the apparatus for measuring the forward scattered intensity I_F , and that for measuring the turbidity τ , were simultaneously calibrated, so that the actual value of τ could be measured directly at any temperature, and so that the actual value of κ_T could be determined at any temperature directly from the value of I_F measured at that temperature. Third, κ_T was determined from measurements of I_F in the temperature range $1.5 \times 10^{-4} \leq \epsilon \leq 3.1 \times 10^{-2}$. Fourth, τ was measured in the temperature range $4.0 \times 10^{-6} \leq \epsilon \leq 6.1 \times 10^{-4}$, the fluid having been stirred by convection in the range $4.0 \times 10^{-6} \leq \epsilon < 1.5 \times 10^{-4}$. These were the only measurements made for which the fluid was not in thermal equilibrium. Fifth, values of ξ were deduced at each temperature where τ had been measured, using Eqs. (2) and (3). The values of κ_T required to calculate ξ in this manner were obtained from the best fit to the measured values of κ_T . This is an extrapolation in the temperature range $4.0 \times 10^{-6} \leq \epsilon < 1.5 \times 10^{-4}$.

In fitting both the data for κ_T and ξ all data points were weighted equally by percent. The value of T_c used was 45.500 K and had already been determined by meniscus height measurements. The statistical errors in the exponents ν and γ computed using the deviations of the individual data points from the fit are too small to be realistic and do not take account of possible systematic errors. My experience in making these measurements leads me to feel that reasonable errors for ν and γ are $\sim \pm 0.01$, and that the accuracy of measuring the actual magnitudes of ξ and κ_T is a few percent.

If the Ornstein-Zernike form of the correlation function is correct, the exponents ν and γ describing the divergences of ξ and κ_T , respectively, are related by $2\nu = \gamma$, which is just allowed by the

assigned errors. If the modified correlation function proposed by Fisher² is used, the value of the exponent η can be deduced from the relation $(2 - \eta)\nu = \gamma$ and the measured values of ν and γ , the result being $\eta = 0.03 \pm 0.03$. A reanalysis of the data using the first-order approximate form of the scattering function given by Fisher and Burford³ resulted in no significant change in the measured values of ξ . The greatest change occurred for the point closest to T_c and increased the measured value of ξ by only 1.5%, an effect equal to that which would be caused by a temperature measuring error of 30 μ K. Within the context of the Ornstein-Zernike formulation the direct correlation range R can be temperature dependent although not divergent, and my results do show R to be weakly divergent. It should be noted however, that over the entire temperature range of this experiment R increases by only 10% as $T \rightarrow T_c$.

ANALYSIS OF RAYLEIGH LINEWIDTHS

The measured values of ξ in combination with other data may be used to check mode-mode coupling theories^{13, 14} which relate the static and dynamic properties of critical systems. This has already been done in the case of sound propagation¹⁵ using preliminary values of ξ , and the results of an analysis of Rayleigh linewidth measurements will be presented here. The analysis depends on extracting the critical part¹⁶ of the linewidth Γ^c from the measured value Γ since it is the critical part which is predicted by the theories. I will use the approach and notation of Swinney and Henry,¹⁷ and the interested reader is referred to Ref. 17 for a much broader and more detailed treatment. The measured Rayleigh linewidth may be written as

$$\Gamma = (\Lambda_B q^2 / \rho C_p)(1 + q^2 \xi^2) + \Gamma^c (C_p^c / C_p), \quad (4)$$

where Λ_B is the background thermal conductivity, C_p the constant pressure heat capacity, and $C_p^c \simeq C_p - C_v$ is the critical part of the heat capacity. The theoretical prediction concerning Γ^c may be written

$$\Gamma^c = \frac{k_B T q^2}{6\pi\eta_s \xi} K_0(q\xi)H(q\xi), \quad (5)$$

where η_s is the shear viscosity and

$$K_0(x) = \frac{3}{4} [1 + x^2 + (x^3 - x^{-1}) \tan^{-1} x]. \quad (6)$$

The function $H(q\xi)$ describes corrections to the basic theory due to including wave-vector dependence in the viscosity, frequency dependence in the viscosity, higher-order "vertex" corrections, and the more exact form of the correlation func-

tion due to Fisher and Burford.³ It may be written as the product of four separate functions of $q\xi$, one for each effect. Thus theory predicts that $(\Gamma^c/q^3)/(k_B T/6\pi\eta_s) \equiv \Gamma^*$ is a function only of $q\xi$. Recently Oxtoby and Gelbart¹⁸ have shown that by including the background value of the Rayleigh linewidth in the calculations, the critical parts of both the linewidth and the shear viscosity may be calculated, thus strengthening the theory considerably. They also obtain a correction factor due to the wave vector dependence of the viscosity which is only half as large as that calculated by Kawasaki and Lo,¹⁹ being 3% for $q\xi \ll 1$ and 11.5% for $q\xi = 10$. They do not find this correction to be strictly a function of $q\xi$, the result depending primarily on $q\xi$ but also very weakly on ξ .¹⁸ I have used their results to calculate Γ^* as a function of $q\xi$ for SF₆ on the critical isochore. In doing so I have ignored the very slight dependence on ξ mentioned above. I have also ignored both "vertex" corrections and the effect of frequency dependence in the viscosity, and I have used the Ornstein-Zernike form of the correlation function since I find the rather small value of 0.03 for η . All of these effects together contribute no more than 5% to the calculated values of Γ^* . In order to obtain experimental values of Γ^* from the measured values of Γ , it is necessary to know $\Lambda^B(T)$, $C_p(T)$, $C_p^c(T)$, $\eta_s(T)$, and $\xi(T)$. I used²⁰ $\Lambda^B(T) = (3.29 + 2.23\epsilon) \times 10^3$ erg/cm sec K and calculated

$$C_p^c \cong C_p - C_v = \frac{T}{\rho} \kappa_T \left(\frac{\partial P}{\partial T} \right)_v^2$$

using my own values of κ_T and the value¹⁰ 8.28×10^5 dyn/cm² K for $(\partial P/\partial T)_v$. The values of C_v listed in Ref. 15 were used to calculate C_p from C_p^c . The values of $\eta_s(T)$ were obtained as the sum of a background contribution,¹⁷ $\eta_s^B = (374 + 131\epsilon)$ μ P, and a critical contribution obtained from the theory.¹⁸ The critical contribution rises to 22% of the background value at $T - T_c = 0.001$ K. At this temperature the calculated value of η is 456 μ P which may be compared with the value 425 μ P obtained by Wu and Webb.²¹ The values of $\xi(T)$ were computed from the best fit to my combined data. Two

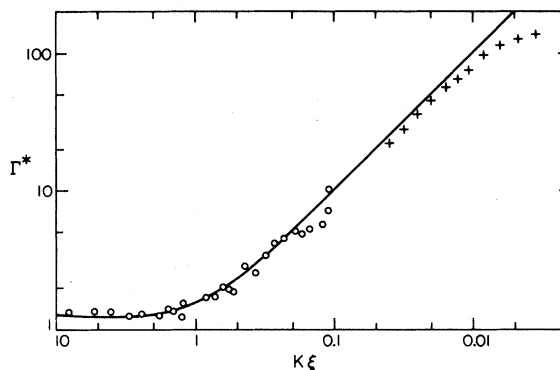


FIG. 2. Reduced Rayleigh linewidth for SF₆ as a function of $K\xi$ on the critical isochore. The solid curve shows the theoretical result. The data of Lim *et al.* are shown as circles, and those of Feke *et al.* as crosses.

sets of experimental data for Γ were analyzed in this way, the data of Lim *et al.*²² taken at a scattering angle of 90° ($q = 1.53 \times 10^5$ cm⁻¹) and the data of Feke *et al.*,¹² taken with $\theta = 4.43^\circ$ ($q = 8.38 \times 10^3$ cm⁻¹). The results are shown in Fig. 2, together with the theoretical curve. The theory is in excellent agreement with the data of Lim *et al.*, shown as open circles, but is systematically larger than the data of Feke *et al.*, shown as crosses, by from 12% to a factor of 2.1, the disagreement being worst for the smallest values of $q\xi$. Such a large discrepancy could be at least partially due to neglect in the theory of terms involving three or more modes.¹⁸ Similar disagreement is evident in the case of xenon,^{17, 18} but is not so marked, perhaps because the data do not extend to such small values of $q\xi$. For the smallest value of $q\xi$ the background contribution to the linewidth, which has been subtracted to find Γ^c , is 80% of the total linewidth. An error of 40% in the estimate of the background would account for the discrepancy as well.

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