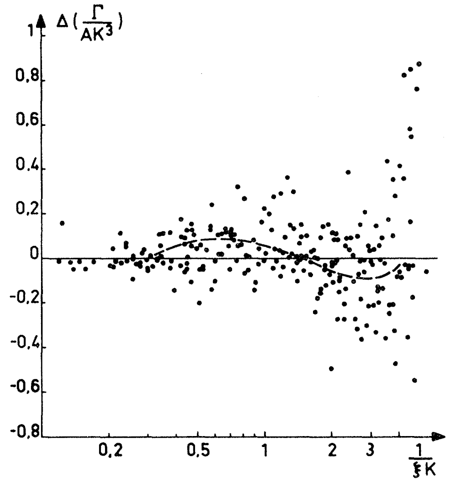

FIG. 2. Deviation of the experimental points from the best-fit curve in the range $0.5 \leq 1/\xi K \leq 4$.

quality factor. This can be done by studying the deviation of each experimental point from the "best fit" theoretical curve as a function of $1/\xi K$. The experimental points should be statistically distributed on both sides of a "true" theoretical curve. On the other hand we find that the experimental points are not statistically distributed around the theoretical curve in the region $0.5 \leq 1/\xi K \leq 4$. This is shown in Fig. 2 where schematically the dotted line shows the behavior of the experimental points around the calculated curve. The fact that this region is mostly responsible for the low quality factor is confirmed by the fact that a fit using only the points outside and on both sides of the region $0.5 \leq 1/\xi K \leq 4$ gives a quality factor of 0.876.

As a final remark we note that, using Kawasaki's expression $A = k_B T/16\eta^*$ and our experimental value $A = 1.51 \times 10^{-13}$ cm^3 sec^{-1}, one gets

$$\eta^* = 1.97 \times 10^{-2} \text{ stokes},$$

which is in excellent agreement with the static value determined by Arcovito et al.[11] using a capillary-flow viscosimeter ($\eta \simeq 1.9 \times 10^{-2}$ stokes).

Thus we may conclude that Kawasaki's expression gives a complete and fairly accurate description of the behavior of the spectrum of the light scattered by a binary mixture in the hydrodynamical and nonhydrodynamical regimes. There is, however, at least in a limited region, a small but significant discrepancy between theory and experiment. We feel that, because of their great number and overall accuracy, our experiments could be compared with an even more refined theory; for instance, one which would consider $\eta \neq 0$, $\eta^* = \eta^*(\xi, K)$, or second-order terms.

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TEST OF A PARAMETRIC EQUATION OF STATE AND CALCULATION OF GRAVITY EFFECTS AT THE GAS-LIQUID CRITICAL POINT

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The effect of gravity on measurements of the sound velocity and specific heat near the gas-liquid critical point is calculated in detail, using the "linear model" parametric equation of state. It is found that the linear model is consistent with the best available experimental data, both in gravity-free and gravity-dominated regions, with a choice of the exponent α equal to 0.06 ± 0.02.

One of the most difficult problems encountered in the study of critical phenomena is the analysis of the "rounding" of the transition, which occurs in all practical experiments.[1] This rounding may be caused by inhomogeneities, finite-sample effects, impurities, or nonequilibrium behavior, and it is often difficult to separate the different effects, or to estimate their magnitude. We report here the detailed calculation of one such effect, the density inhomogeneity induced by gravity near the gas-liquid critical point.[1,2] The analysis is tractable because the effect is caused by a precisely known force, whose strength can effectively be varied by changing the sample height. Because of the divergence of the compressibility, the effect is largest close to T_c. However, it is also most difficult to calculate there, since the density distribution present in the sample depends in a complicated way on the unknown equation of state.[2,3]

Recently, a convenient parametric form was proposed for "scaled"[4] equations of state, and a simple "linear model" (LM) was introduced.[5] This model gives a satisfactory description of experimental data on magnetic systems and fluids, using a small number of parameters. We have used the LM to carry out the first realistic quantitative calculations of the gravity effect in fluids. We have first tested the LM in a region where gravity is negligible, and find that it is consistent with the best presently available data on the specific heat C_v[2,6] and the sound velocity u,[7] but only if the exponent α[4] is equal to 0.06 ± 0.02. Once the value of α is fixed, the behavior of C_v and u is entirely determined in the gravity-dominated region, where the LM is also found to be consistent with the data, within the experimental uncertainties.

The equation of state in the LM may be written in the form[5]

$$[\mu(\rho,T)-\mu_0(T)]/\mu_c \equiv a\theta(1-\theta^2)r^{\beta\delta}, \tag{1a}$$

$$(\rho-\rho_c)/\rho_c \equiv k\theta r^{\beta}, \tag{1b}$$

$$(T-T_c)/T_c \equiv t \equiv (1-b^2\theta^2)r, \tag{1c}$$

where μ is the chemical potential, ρ the density, and T the temperature, and μ_c, ρ_c, and T_c are their respective critical values. The critical exponents β and δ have their usual definitions,[4] and the variables r and θ are defined by Eq. (1). The constants a and k are determined[8] from experiment,[9] and b is given by the "minimization" condition[5] $b^2=(\delta-3)/(\delta-1)(1-2\beta)$. In view of the scaling relations[4] $\beta(\delta+1)=2-\alpha=\gamma+2\beta$, only two of the critical exponents are independent; of these, we shall take β from experiment[9] and leave α as an undetermined parameter. The Helmholtz free energy is

$$A(\rho,T)=F(\rho,T)+A_0(T)+\mu_0(T)\rho, \tag{2}$$

where $A_0(T)$ and $\mu_0(T)$ are regular functions of T with $\mu_0(T_c)=\mu_c$. The singular part of A may be written as $F=f(\theta)r^{2-\alpha}$, with $f(\theta)$ determined[5] by integrating the relation $\mu=(\partial A/\partial\rho)_T$.

For a given sample height h, there is a characteristic temperature interval $t_0(h)$ (to be determined below), outside of which gravity effects are negligible. Thus, for $|t|\gg t_0(h)$, the measured specific heat ρC_v along the critical isochore is ρC_μ, or in magnetic notation,[5] C_H. We have,[5] for $\rho=\rho_c$,

$$\rho C_v/T=A^{\gtrless}|t|^{-\alpha}+C_0, \quad t\gtrless 0, \tag{3}$$

where expressions for the constants $A^{\gtrless}$ are given in Ref. 5. It is important to note that in "simple" scaling theories,[4,5,9] C_0 is regular at T_c, i.e., there is no additional discontinuous term in C_v apart from the term $A^{\gtrless}|t|^{-\alpha}$. Thus the value of the "jump" $C_v(-t)-C_v(t)$ at a point away from T_c $[|t|\gg t_0(h)]$ depends only on $A^{\gtrless}$ and α, and may be used within the linear model to fix the value of the remaining unknown parameter, the exponent α. By performing such a fit for He^4[6] and Xe,[2] we have found the values $\alpha=0.07$ and $\alpha=0.05$, respectively, with an uncertainty no larger than ± 0.005![10]

The zero-frequency sound velocity u and adiabatic compressibility κ_s are found from the thermodynamic relation[11]

$$u^2=(\rho\kappa_s)^{-1}=(\rho\kappa_T)^{-1} + (T/\rho^2)(\partial P/\partial T)_\rho{}^2 C^{-1}, \tag{4}$$

with gravity effects still neglected. As T approaches T_c, $(\rho\kappa_T)^{-1}$ goes to zero and $(T/\rho^2)(\partial P/\partial T)_\rho{}^2$ approaches a constant, so that $u^2\propto C_v{}^{-1}$.[11,7] We have calculated C_v and u in the whole (ρ, T) plane from the LM, and have compared the values with experimental data[6,7] in He^4. The results for u are shown in Fig. 1, where the theory has been normalized to the data at $t=3.9\times10^{-3}$, by adjusting the constants d^2A_0/dT^2 and $d^2\mu_0/dT^2$ [see Eq. (2)] for the best fit. The agreement between experiment and theory is satisfactory for T not too close to T_c. Similar agreement is obtained with the C_v data,[6] using the same parameters. The small discrepancies, which appear in Fig. 1 at $t=9.3\times10^{-4}$ for $\rho>\rho_c$, are probably not within the error of the experimental determination of $\rho-\rho_c$. They must await a more careful study, however, before they may be attributed to breakdown of the LM. For the points closest to T_c, the disagreement is more pronounced, but, as shown below, the gravitational corrections are important in this region.

In a gravitational field, the chemical potential μ of an isothermal fluid in equilibrium varies with the vertical height z according to the rela-

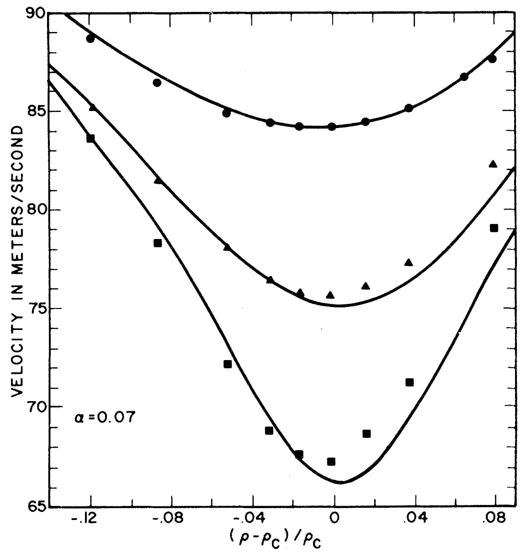

FIG. 1. Sound velocity versus reduced density for He[4]. The data of Ref. 6 at ≈1.8 kHz are denoted by closed circles, triangles, and squares, corresponding to $t = 3.9\times10^{-3}$, 9.3×10^{-4}, and 1.4×10^{-4}, respectively. The solid lines represent the zero-frequency velocity calculated from the linear model without gravity corrections.

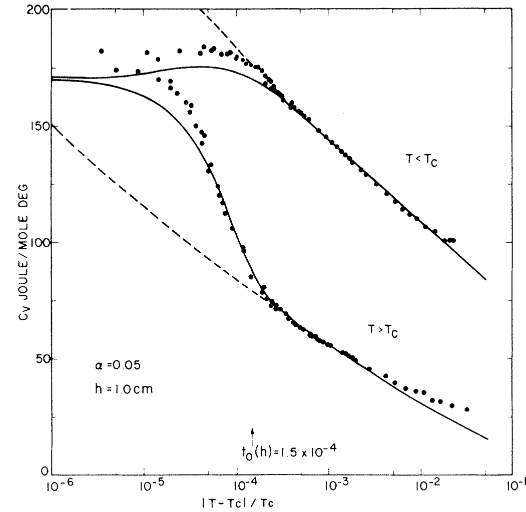

FIG. 2. Specific heat versus reduced temperature in Xe for $\bar{\rho}=\rho_c$. The solid line is the gravity average $\tilde{C}_v$ in the linear model. The points are the experimental data of Ref. 2. The dashed line represents the theoretical C_v in the absence of gravity.

tion $d\mu = g dz$, where g is the acceleration of gravity. Choosing the origin of z to be the height at which $\mu=\mu_0(T)$ (i.e., $\rho=\rho_c$), we may integrate this relation and combine it with Eq. (1a) to obtain

$$z = -h_0 a\theta(1-\theta^2)r^{\beta\delta}, \tag{5}$$

with $h_0 \equiv \mu_c/g$. For a sample of height h, a characteristic temperature interval $t_0(h)$, at which gravity effects become important, may be estimated from Eqs. (5) and (1) as $t_0(h)\equiv(h/ah_0)^{1/\beta\delta}$. For example, in He[4], with $h=0.5$ cm, $t_0 = 2.5 \times 10^{-4}$.

The density distribution in the sample may be obtained in principle by solving Eqs. (5) and (1c) for $\theta(z,t)$, and inserting this expression into Eq. (1b) to obtain $\rho(z,t)$. The location of $z=0$ with respect to the sample dimensions may be obtained by integrating $\rho(z,t)$ throughout the sample volume to obtain the average density $\bar{\rho}$. Similarly, the local values of any other thermodynamic function follow from the appropriate parametric form, with $r = t/(1-b^2\theta^2)$ and $\theta=\theta(z,t)$. For simplicity, we shall restrict the discussion of gravity effects to the case where $\bar{\rho}$ is ρ_c, and the sample has a uniform cross section.[12] Then from the symmetry properties of Eqs. (5) and (1), it may be seen that the origin of z always occurs at the geometric center of the sample.

As noted by previous authors,[2,3] the "average" specific heat $\tilde{C}_v$ is not the average of the local C_v, but rather the derivative of the average entropy.[13] Thus, for $\bar{\rho}=\rho_c$[12] in a cylindrical container of height h,

$$\tilde{C}_v = T\frac{d}{dT}\tilde{S}(T) = T\frac{d}{dT}\int_{-h/2}^{h/2} S(z,T)dz, \tag{6}$$

where $S(z,T)$ is the local entropy. The differentiation may be performed under the integral sign by expressing S as a function of $\theta(z,t)$ and differentiating at constant z. We have calculated $\tilde{C}_v$ numerically from Eq. (6) for the case of Xe, with $\alpha = 0.05$ (see above). The results for $h=1$ cm are shown in Fig. 2, where they are compared with the data of Ref. 2. It is seen that the shape is accurately reproduced; the maximum is correctly predicted to occur at a temperature $t_m = -4.5\times10^{-5} \approx -0.3t_0$, which is below T_c; and the absolute value of $\tilde{C}_v$ at T_c is correctly given, to within the accuracy of the experiment.[14] In particular, we consider it significant that the value of α which was fitted to the jump in C_v in the gravity-free region also yields a value consistent with experiment at T_c. For $\alpha=\frac{1}{8}$, for instance, the LM would predict a value $\tilde{C}_v = 279$ J/mole K at T_c, and a jump $\Delta C_v = 142$ J/mole K at $|t| = 10^{-3}$, both of which are quite far from the experimental values (see Fig. 2). Thus the linear model self-consistently chooses the value $\alpha = 0.05 \pm 0.01$ in Xe. It is also worth noting that although the same exponent α was used above and below T_c, an "apparent exponent," determined by a power-law fit to the solid line in Fig. 2 over a limited range of t, would be less than α for $T < T_c$, and greater than α for $T > T_c$.

We have also calculated $\tilde{C}_v$ in He^4 and Ar and find theoretical curves which are, in appropriate reduced units, quite similar to those in Fig. 2. Comparison with the He^4 data of Moldover[6] reveals certain disagreements in the gravity-affected region very close to T_c, which we attribute to experimental errors, uncertainties in the value of T_c, and a departure from ideal cylindrical geometry. In the case of Ar, when we compare our theory with the data quoted by Berestov, Giterman, and Malyshenko,[3] we find serious disagreements in the shape of the curve, as well as the value of the maximum in $\tilde{C}_v$ and the magnitude of the gravity correction away from T_c. We believe that these discrepancies are due to systematic experimental errors. The theory of Berestov, Giterman, and Malyshenko[3] is based on an equation of state which is known to be inaccurate near T_c, and yields results for the gravity effect which are not quantitatively correct.

In order to find the "average sound velocity" $\tilde{u}$, as measured by a resonance experiment under gravity, we must calculate the normal modes of the resonator, with a given local sound velocity $u(z)$. To do this, we have solved the wave equation numerically by a method devised for us by Wasserstrom.[15] The results for the lowest radial mode[7] of a cylindrical resonator are shown in Fig. 3, along with experimental data[7] in He^4. The general shape of $\tilde{u}$ vs t is similar to the inverse of $\tilde{C}_v$, except that the minimum occurs above T_c.[16] Since no adjustable parameters were used in Fig. 3 [the parameters of the LM were determined entirely from data in gravity-free regions[6,7,9]], the results are encouraging, but more extensive and precise data are needed very close to T_c before the LM can be tested critically in the gravity region. In addition, nonlocal effects,[13] leading to dispersion,[7] are not entirely negligible in this range and must be accounted for in detail.

We have also calculated $\tilde{u}$ in CO_2 for a height of 4.26 cm, as used by Feke, Fritsch, and Carome,[11] and find that the gravity corrections are appreciable in those experiments [$(\tilde{u}-u)/u \approx 6\%$ at $T-T_c = 0.02$ K], and may partly explain the small value of α quoted by these authors.

In conclusion, we have used the LM to carry out the first realistic calculations of gravity effects near the gas-liquid critical point. The best

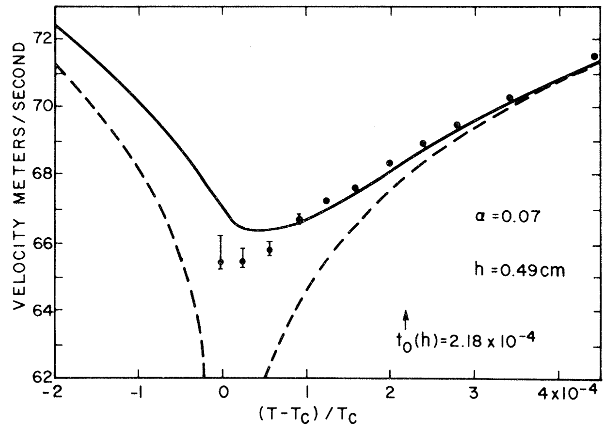

FIG. 3. Sound velocity versus reduced temperature for $\tilde{\rho} = \rho_c$ in He^4. The solid circles represent the data of Ref. 7, taken at ≈ 1.8 kHz. No dispersion corrections have been made. The solid line represents the gravity-averaged sound velocity $\tilde{u}$, calculated from the linear model. Note the linear temperature scale.

presently available data on C_v and u are consistent with the LM in He^4 and Xe, in both gravity-free and gravity-dominated regions, with an exponent $\alpha = 0.06 \pm 0.02$. The value $\alpha = \frac{1}{8}$, on the other hand, is inconsistent with the LM, although it is by no means ruled out by the experimental data themselves.[2,6,7] More accurate experiments, perhaps as a function of sample height, are needed to test the LM further, in order to determine whether the slight disagreements between experiment and theory found by our analysis are significant. If they are, the model will have to be modified, and the exponent α may then take on a value different from 0.06. We emphasize, however, that even in this case the parametric form can still be used for the modified equation of state, and the present methods may be applied to a calculation of gravity effects. Adequate account of such effects is of course crucial to any accurate analysis of experiments very near the critical point.

We wish to thank Dr. E. Wasserstrom for devising a very elegant and convenient numerical solution of the wave equation. We are grateful to Miss J. Seery and Mrs. Z. Wasserman for considerable help in programming, to V. Chirba for aid in the analysis, and to Professor D. Litster, Dr. J. M. H. Levelt Sengers, and Professor M. Buckingham for fruitful discussions and important preprints.

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[9]M. Vicentini-Missoni, J. M. H. Levelt Sengers, and M. S. Green, J. Res. Natl. Bur. Std. (U.S.) 73A, 563 (1969). Unless noted otherwise, we shall use the values quoted in this work for all experimental constants.

[10]Using the values of the constants given in Ref. 9, we have estimated α from the jump in C_v for that model and find $\alpha \approx 0.035$ for Xe, He^4, and CO_2. This value is close to the value $\alpha \approx 0.04$-0.06 obtained in Ref. 9 by a least-squares analysis of PVT data, leaving α as a free parameter.

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[12]The general case, $\tilde{\rho} \neq \rho_c$, involves more complicated algebra and will be treated in a later publication.

[13]Throughout this work we neglect all nonlocal effects caused by the finite correlation length ξ [see Ref. 2]. For thermodynamic measurements these effects may be shown to be negligible in the experimental temperature range. For the sound velocity measurements, nonlocal effects are not entirely negligible very near T_c, even at the lowest frequencies used (see Ref. 7 and Fig. 3). A more thorough study of these effects will be carried out in a later publication.

[14]Professor Buckingham informs us that the data were obtained from heating curves, so that we expect nonequilibrium behavior to lead to experimental points which, very close to T_c, are too high. In addition, an increase in the value of T_c by 3×10^{-3} K ($\delta t \approx 10^{-5}$) would improve the agreement considerably for $T > T_c$. Finally, we note that the theoretical value of $\tilde{C}_v$ at T_c can be made to fit experiment exactly by choosing $\beta = 0.333$.

[15]E. Wasserstrom, to be published.

[16]For the lowest radial mode, the minimum of u is at $t_m = 4.2\times10^{-5} \approx 0.2t_0(h)$ (see Fig. 3). For the other modes, the value of t_m varies up to about $0.5t_0(h)$.

BALLOONING MODES IN AXISYMMETRIC TOROIDAL CONFIGURATIONS

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The stability of ballooning modes is investigated in the neighborhood of the magnetic axis of an axisymmetric toroidal configuration of the Tokamak type. It is shown that the stability criterion for these modes is the same as that for localized modes.

A purely poloidal configuration of the multipole type with closed magnetic field lines and an average magnetic well may become unstable when the ratio β of the kinetic pressure to the magnetic pressure exceeds a critical value.[1] The unstable perturbations have larger amplitude in the regions of unfavorable curvature of the field lines than in the regions of favorable curvature and are called ballooning modes. On the other hand, for toroidal configurations with rotational transform (Tokamak, Stellarator) the magnetohydrodynamic stability problem is usually studied with the so-called localized criterion.[2-4] The corresponding perturbations are much more localized near a magnetic surface than the ballooning modes, and in the case of an axisymmetric discharge having magnetic surfaces with circular cross sections it is found[5,6] that their stability is almost independent of β.

The purpose of the present work is to derive a stability criterion against ballooning modes for axisymmetric toroidal configurations with current parallel to the magnetic field lines to provide a rotational transform. Since all perturbations are easily stabilized by shear of the field lines, we restrict our calculations to the neighborhood of the magnetic axis where shear is negligible.

In cylindrical coordinates r, φ, z, the axisymmetric equilibrium magnetic field is

$$\vec{B} = \frac{T(r,z)}{r}\vec{e}_\varphi - \frac{1}{r}\vec{e}_\varphi \times \nabla\psi(r,z), \tag{1}$$