sions.

published).

fluctuations in the superradiating process known to us are restricted to the cases of few atoms or a single mode of the radiation field. The interesting work of Bonifacio and Preparata³ is of the latter type, and one sees in it the periodic exchange of energy between atoms and field which should be expected in a cavity. The principal

*Research partially supported by the National Science Foundation.

[†]A preliminary report of this research was made at the Rochester Symposium on the Electromagnetic Interactions of Two-Level Atoms, 1970 (unpublished).

¹R. H. Dicke, Phys. Rev. <u>93</u>, 99 (1954).

²M. Dillard and H. Robl, Phys. Rev. <u>184</u>, 312 (1969).

³R. Bonifacio and G. Preparata, Phys. Rev. (to be

PHYSICAL REVIEW A

VOLUME 2, NUMBER 4

OCTOBER 1970

Critical Isotherm of He³

Barnie Wallace, Jr. and Horst Meyer

Department of Physics, Duke University, Durham, North Carolina 27706 (Received 27 April 1970)

The critical isotherm of He³ has been measured over a density range $|\Delta\rho| = |\rho - \rho_c|/\rho_c$ up to 0.6, and the chemical potential change $\mu - \mu_c$ was obtained as a function of the density change $\rho - \rho_c$. The departure from perfect antisymmetry about ρ_c , which can be represented by an expression of the form $|\Delta\rho|^{\epsilon}$ with $\epsilon = 6.2$, is compared with theoretical suggestions.

Recently Missoni, Sengers, and Green¹ have reported success in scaling the thermodynamic properties near the critical point for a number of fluids and magnets. They found that the change in chemical potential

 $\Delta \mu = \left[\mu(\rho, T) - \mu(\rho_c, T) \right] / P_c V_c$

was antisymmetric in the change in density $\Delta \rho = (\rho - \rho_c)/\rho_c$ for $|\Delta \rho| < 0.30$ for the fluids He⁴, Xe, and CO₂ when $-0.01 < t = (T - T_c)/T_c < 0.03$. Along the critical isotherm, $\Delta \mu \propto \Delta \rho |\Delta \rho|^{6-1}$, where $\delta \simeq 4.0 - 4.6$ for all fluids. We have reported² agreement for He³ with their scaled equation of state for $|\Delta \rho| < 0.25$, and in the present note we wish to describe the departure from antisymmetry for $0.25 < |\Delta \rho| < 0.6$.

Cooper, Sengers, and Green³ (CSG) have proposed a modification of the scaling ideas to extend the interpretation over an enlarged region about the critical point and also to include systems lacking an intrinsic or known symmetry. Cooper and Green⁴ found for an ideal Bose gas that the simple scaling form appears as the first term in an expansion about the critical point for a nonclassical equation of state. The corresponding Helmholtz energy F was expressed as a function of t and reduced variable $x = t |\Delta\rho|^{-1/\beta}$. Generalizing this result to physical systems of no known symmetry, CSG assert that the free energy in the critical region is given by an expression of the form

drawback of the Bialynicka-Birula⁴ treatment is

the restriction to times sufficiently close to the

have concentrated on small numbers of atoms. We would like to thank our colleagues, especially

instant of peak emission, while Dillard and Robl²

G. S. Agarwal and L. Mandel, for several discus-

⁴Z. Bialvnicka-Birula, Phys. Rev. D 1, 400 (1970).

⁶J. H. Eberly and N. E. Rehler, Phys. Letters <u>29A</u>,

142 (1969); N. E. Rehler, Ph. D. dissertation, Univer-

⁵N. E. Rehler and J. H. Eberly (unpublished).

⁷L. Mandel, Phys. Rev. 136, B1221 (1964).

sity of Rochester (unpublished).

$$F(\rho, t) = F_0(\rho, t) + \left| \Delta \rho \right|^{\delta + 1} \sum_{s=1}^{\infty} \Delta \rho^{s-1} f_s(x) , \qquad (1)$$

where the $f_s(x)$ are regular functions. Since one has

$$\mu(\rho, t) = \left(1 + \rho \frac{\partial}{\partial \rho}\right) F(\rho, t) , \qquad (2)$$

we immediately see that along the critical isotherm where the asymmetric $\Delta \mu$ term varies as $\Delta \rho$ $\times |\Delta \rho|^{\delta-1}$, the first correction which gives a symmetric term is of the order $\Delta \rho |\Delta \rho|^{\delta-1}$ with $\epsilon = \delta + 1$.

Griffiths⁵ has discussed two asymmetric latticegas models. For his first model, the leading symmetric term for the chemical potential change $\Delta \mu$ as a function of $\Delta \rho$ along the critical isotherm has an exponent $\epsilon = 2\delta$. A more recent model, which is a "decorated" system, gives $\epsilon = 2\delta - \beta^{-1}$. We have made extensive P, ρ , T measurements near the critical point of He³ for solutions of 10 and 250-ppm He⁴ impurities using dielectric-constant techniques. The 250-ppm work,² which will be reported elsewhere in detail, describes the cryostat, experimental procedures, and errors. We found no significant differences between the two mixtures, and we report here our findings on He³(10-ppm He⁴) where we have extended our density measurements along the critical isotherm to $|\Delta \rho|=0.6$ so as to compare our results with the suggestions of CSG and Griffiths.

The coexistence-curve data from 17 isotherms were analyzed, and we obtained $\rho_c = 0.04138$ $\pm 0.0002 \text{g/cm}^3$, $\beta = 0.361 \pm 0.005$, and $T_c = 3.3098$ \pm 0.0005 °K. The *P*-versus- ρ relation for three isotherms 3.3095, 3.3098, and 3.3101 $^\circ K$ was then measured over large density ranges and μ $-\mu_c$ was obtained by graphical integration using the expression $d\mu = V dP$ along an isotherm. The systematic correction of the results caused by the slight density dependence⁶ of the dielectric constant of He³ was found to be negligibly small, as discussed previously.² The large number of data makes a tabulation impractical in this note. The original $P-\rho-T$ data, together with the calculated $\Delta\mu$ -versus- $\Delta\rho$ isotherms, will be submitted to the National Auxiliary Publications Service, together with the results described in Ref. 2. The tabulations are also available from the authors.

For T = 3.3098 °K, a plot of $|\Delta \mu|$ versus $|\Delta \rho|$ on logarithmic scales gave within experimental error



FIG. 1. Logarithmic plot of the reduced change in chemical potential $|\Delta \mu|$ versus the reduced density change $|\Delta \rho|$ in He³ for three isotherms. The temperature T = 3.3098 °K is believed to be the critical one. For convenience, the range $0.16 < |\Delta \rho| < 0.22$, which shows a continuation of the straight line for T=3.3098 °K has been omitted from this figure.



FIG. 2. Logarithmic plot of $|\Delta \mu|$ versus $|\Delta \rho|$ for $|\Delta \rho| > 0.22$ on the critical isotherm of He³, showing the departure from antisymmetry between the gas side $(\rho < \rho_c)$ and the liquid side $(\rho > \rho_c)$.

a straight line for $|\Delta \rho| < 0.25$ (Fig. 1), thus reinforcing our conclusion that this was the true critical isotherm. From this density range, where there is no evidence of a symmetric term in $\Delta \mu$, we obtained $\delta = 4.16 \pm 0.1$. This value is in good agreement with the theoretical prediction by Kiang⁷ based on the liquid-droplet model, namely, $\delta = 4.24$ for He³.

For $0.25 < |\Delta\rho|$, the first-order correction becomes increasingly significant, and the experimental results (Fig. 2) could be well represented by the expression

$$\Delta \mu = A \Delta \rho \left(\left| \Delta \rho \right|^{6-1} - B \Delta \rho \left| \Delta \rho \right|^{6-2} \right) , \qquad (3)$$

where A = 2.42, B = 0.45, and $\epsilon = 6.18 \pm 0.5$, and we note that $|\Delta \mu|_{gas} > |\Delta \mu|_{1iq}$ for a given $|\Delta \rho|$. For clarity we plot in Fig. $3 \Delta \Delta \mu = |\Delta \mu|_{gas} - |\Delta \mu|_{1iq}$



FIG. 3. The difference in chemical potential between gas and liquid on the critical isotherm in He³ (present work) and He⁴ plotted on a logarithmic scale versus $|\Delta \rho|$.

versus $\Delta \rho$ on a logarithmic scale, where it can be seen that within the experimental error a straight line is obtained. Since the theoretically suggested exponent ϵ is that for the first symmetric term only in a power series in $\Delta \rho$, it may be assumed that comparison between experiment and theory can only be made for $\Delta\Delta\mu \ll \langle |\Delta\mu| \rangle_{av}$. The difficulty is, of course, that for small $\Delta \Delta \mu$, the experimental scatter is large, preventing an accurate determination of the exponent ϵ . One might expect that, as $\Delta \Delta \mu$ increases, more terms in the series will become important, and a logarithmic plot of $\Delta\Delta\mu$ versus $\Delta \rho$ would show some curvature. This does not appear to be the case in our experiments up to $\Delta\Delta\mu/\langle |\Delta\mu| \rangle_{av} \simeq 0.3$. Hence possibly, the straight line we obtain in Fig. 3 implies that only the first symmetric term in the above-mentioned series is contributing and that the measured slope is not an average representing several terms in the expansion series. Although we are not able to justify from first principles that $|\Delta \mu_{gas}|$ should be larger than $|\Delta \mu_{1ig}|$, we note that this is so for a fluid obeying the classical van der Waals equation. Here one finds $\delta = 3$ and $\epsilon = \delta + 1 = 4$. Higher-order terms for the correction from antisymmetry around ρ_c become important for $|\Delta \rho| \ge 0.35$.

¹M. Vicentini-Missoni, J. M. H. Levelt Sengers, and M. S. Green, Phys. Rev. Letters <u>22</u>, 389 (1969).

 $^2B.$ Wallace and H. Meyer, this issue, Phys. Rev. A $\underline{2},$ 1563 (1970).

³M. J. Cooper, J. M. H. Levelt Sengers, and M. S. Green (unpublished).

⁴M. J. Cooper and M. S. Green, Phys. Rev. <u>176</u>, 302 (1968).

⁵R. B. Griffiths (private communication).

We also examined the He⁴ data of Roach, ⁸ and calculated $\Delta \mu$ versus $\Delta \rho$ along his "critical" isotherm (T = 5.1929 °K), and over the density range that extended up to $|\Delta \rho| = 0.45$. These He⁴ results are plotted in Fig. 3, where we note the disagreement with the data for He³. This discrepancy is not understood, especially as other scaled properties are nearly the same. However, the slope ϵ is about 7 ± 1.5 and is consistent with that for He³.

We conclude that for both helium isotopes, the exponent ϵ that describes the departure from perfect antisymmetry of $\Delta\mu$ versus $\Delta\rho$ along the critical isotherm lies between the two extreme theoretical suggestions, namely, $\delta + 1$ and 2δ . It is fairly consistent with the suggestion $2\delta - 1/\beta = 5.6$. The data on He³, more abundant than that on He⁴, suggest a power of about $\delta + 2$.

ACKNOWLEDGMENTS

The authors are grateful to Dr. R. B. Griffiths, Dr. M. J. Cooper, and Dr. J. M. H. Levelt Sengers for stimulating conversations and correspondence. This work was supported by a grant from the National Science Foundation.

⁶E. C. Kerr and R. H. Sherman, *Proceedings of the Eleventh InternationalConference on Low Temperature Physics, St. Andrews, Scotland, 1968 edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (University of St. Andrews Printing Dept., St. Andrews, Scotland, 1969), Vol. 1, p. 236.*

⁷C. S. Kiang, Phys. Rev. Letters <u>24</u>, 47 (1970). ⁸P. Roach, Phys. Rev. <u>170</u>, 213 (1968).

PHYSICAL REVIEW A

VOLUME 2, NUMBER 4

OCTOBER 1970

ERRATA

Stimulated Electric Polarization and Photon Echoes. C. V. Heer and R. H. Kohl [Phys. Rev. A 1, 693 (1970)]. Although Eq. (22) is correct for $U(\bar{t}, 0)$, it must be changed for $U(t, t_0)$ to

$$U(t, t_0) = \sum_{\substack{n, n' \\ \alpha}} |n| e^{-iE_n t/\hbar} e^{-i\ell_{n\alpha} t} B(n\alpha) B^*(n'\alpha)$$
$$\times e^{i\ell_{n'\alpha} t_0} e^{iE_n t_0/\hbar} (n')$$

 $= e^{-i(H_0/\hbar + \Delta)t} e^{-i\xi(t-t_0)} e^{i(H_0/\hbar + \Delta)t_0}$

This change in form is important for the study of the shape of the echo and the time dependence of the echo is dependent on $e^{-i\omega t}e^{-i\Delta(t-t_3-t_2+t_1)}$. Echo intensity and polarization depend on $\xi(t-t_0)$ and the results reported in the paper are unchanged.

Hyperfine Structure of the Ground State of 3 He⁺ by the Ion-Storage Exchange-Collision Technique, H. A. Schuessler, E. N. Fortson, and H. G. Dehmelt [Phys. Rev. <u>187</u>, (1969)]. The following corrections should be noted: