

Experimental determination of the tricritical crossover exponent*

N. Giordano[†]

Department of Engineering and Applied Science, Yale University, New Haven, Connecticut 06520

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It is shown how the tricritical crossover exponent ϕ can be determined directly from experimentally accessible quantities. The analysis of previously published data gives $\phi = 1.95 \pm 0.08$ for ^3He - ^4He mixtures, and $\phi = 1.95 \pm 0.11$ for the antiferromagnet dysprosium aluminum garnet. These results are in good agreement with the present theory which predicts $\phi = 2$.

I. INTRODUCTION

There has been considerable interest recently in the behavior of systems near tricritical points.¹ In analogy with the theory of critical points,² the values of the various tricritical exponents are of prime importance. One tricritical exponent which plays an important role in the theory is the crossover exponent ϕ .³⁻⁵ The exponent ϕ describes the shape of the phase boundaries in field space, and also determines how exponents are "renormalized" depending upon the path used to approach the tricritical point.⁵ Although ϕ has been estimated from experimental data by indirect means such as from the ratio of two other exponents,⁶ its value has yet to be determined directly. The reason for this is that until now it has not proved possible to separate the contribution containing ϕ from the regular or nonsingular contribution in various thermodynamic quantities. In this paper I point out how this separation can be made and thus show how ϕ can be extracted *directly* from experimentally accessible quantities. This makes possible the first direct experimental determination of ϕ .

II. THEORY

The phase diagram in field space near a tricritical point is shown in Fig. 1.⁵ Here T is the temperature and H is the nonordering field. In antiferromagnets, H is the internal magnetic field while in ^3He - ^4He mixtures it is the chemical potential difference between the two isotopes. The phase boundary separates the ordered phase which is present for small T and H , from the disordered phase. In antiferromagnets these two phases are the antiferromagnetic and paramagnetic states, while in helium mixtures they are the superfluid and normal fluid phases. For temperatures less than the tricritical temperature (T_t) the phase transition is first order while for $T > T_t$ it is second order. The phase boundary for $T > T_t$ is a line of critical points and is often referred to as the λ line.

According to both scaling^{4,5,7} and renormalization-group⁸ theories, the "fundamental" fields for describing tricritical behavior are the scaling fields g_1 and g_2 . Although it has not yet proved possible to determine the scaling fields for any real system, it is known that the scaling fields are analytic functions of H and T ,^{8,9} and that one of them is parallel to the phase boundary at the tricritical point while the other is not. In the following we will take g_2 to be the tangential scaling field (see Fig. 1). The theories predict that near the tricritical point the singular part of the free energy obeys the functional equation⁴

$$F_s(l^\phi g_1, l g_2) = l^{\phi(2-\alpha_t)} F_s(g_1, g_2), \quad (1)$$

where α_t is a susceptibility exponent and l is an arbitrary (positive) constant.

As shown by Riedel,⁵ and Hankey, Stanley and Chang,⁷ the equation of the λ line in the g_1 - g_2 plane follows directly from (1). On the λ line F_s is singular,^{5,6} and from (1) we see that if F_s is singular at the point (g_1, g_2) then it must also be singular at

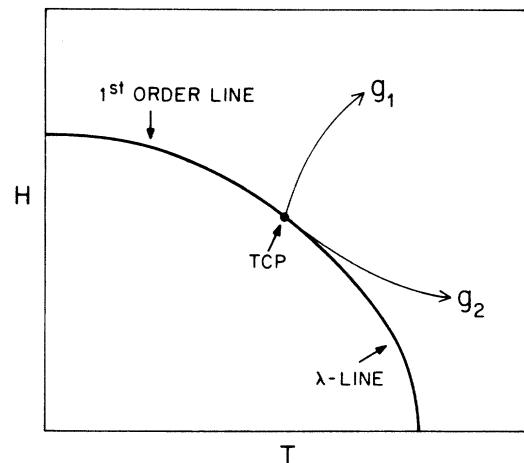


FIG. 1. Tricritical-point phase diagram with the tricritical point (TCP) as indicated. The heavy line is the phase boundary. Also shown is one possible orientation of g_1 and g_2 .

$(l^{\phi}g_1, l^{\phi}g_2)$. As l is arbitrary, this gives a parametric representation of the λ line. Elimination of l gives^{5,7,10,11}

$$g_1 = Dg_2^{\phi}, \quad (2)$$

where D is a constant. From (2) the equation of the λ line in H - T space can be derived. Defining $t = (T - T_t)/T_t$ and $h = (H - H_t)/H_t$, where H_t is the tricritical value of H , and using the analyticity of g_1 and g_2 we have

$$g_1 = h - \left(\frac{dh}{dt}\right)_t t + at^2 + bh^2 + cth + \dots, \quad (3)$$

$$g_2 = t + eh + \dots,$$

where $(dh/dt)_t$ is the slope of the phase boundary at the tricritical point in the appropriate units and a , b , c , and e are (unknown) constants. Inserting (3) into (2) gives for the equation of the λ line to lowest order in t

$$h_{\lambda} = \left(\frac{dh}{dt}\right)_t t + D't^{\phi} + a't^2, \quad (4)$$

where D' and a' are given by

$$D' = D \left[e \left(\frac{dh}{dt}\right)_t + 1 \right]^{\phi}, \quad (5)$$

$$a' = -a - c \left(\frac{dh}{dt}\right)_t - b \left(\frac{dh}{dt}\right)_t^2.$$

In (4) we have dropped terms of order t^3, t^4, \dots , which arise from the expansion of g_1 , and also terms of order $t^{2\phi-1}, t^{3\phi-2}, \dots$ and $t^{\phi+1}, t^{\phi+2}, \dots$, which arise from the expansion of g_2 . In the following we will be concerned only with cases in which $\phi > \frac{3}{2}$ and thus these terms can all be safely neglected in (4).

The last term in (4) is the regular or nonsingular contribution. The present theories^{8,12} predict $\phi = 2$, and hence the "singular" and nonsingular terms in (4) are predicted to be of the same order in t . However, experimental data in conjunction with (4) cannot be used to test this prediction for the following reason. Given data for h_{λ} over only a finite range of t , it is impossible to distinguish in (4) between the case $\phi = 2$, and the case $\phi \neq 2$ with $D' \ll a'$. The fact that experimental data for h_{λ} can be fit to (4) with $\phi = 2$ may only indicate that $D' \ll a'$, and therefore it is not possible to determine ϕ from the shape of the λ line alone. This fact has been noted previously by Riedel, Meyer, and Behringer.⁶

I would like to point out that it is possible to use *additional* experimental data to separate the singular and nonsingular contributions in (4). Let M denote the nonordering density (the magnetization in an antiferromagnet and the ^3He concentration in helium mixtures) and let M_t be its value at

the tricritical point. Assuming only that $\phi > 1$ and that the exponent $\beta_u \leq 1$ (which has been found experimentally to be the case for the systems we will be considering), it is shown in the Appendix that the equation of the path $M = M_t$ has the form⁶

$$g_1 = E g_2^{\phi}, \quad (6)$$

where the effects of the regular part of the free energy have been *included*. From (6) and (3) the equation of this path in h - t space is

$$h_{M_t} = \left(\frac{dh}{dt}\right)_t t + E't^{\phi} + a't^2, \quad (7)$$

where E' is defined in analogy with D' and we have again dropped all terms of order higher than t^2 . The crucial feature is that the regular contributions in (4) and (7) are equal, and thus the difference $h_{M_t} - h_{\lambda}$ is free of regular contributions to order t^2 . From (4) and (7) we find

$$h_{M_t} - h_{\lambda} = (E' - D')t^{\phi}. \quad (8)$$

Since the quantity $h_{M_t} - h_{\lambda}$ can be readily measured, (8) can be used to determine ϕ .

I should also note that there is another combination of quantities which can be used to determine ϕ . From (1) it can be shown^{5,7} that the equation for the first-order line has the same form as (2). This leads to a relation analogous to (4); we find

$$h_1 = \left(\frac{dh}{dt}\right)_t t + F|t|^{\phi} + a't^2, \quad (9)$$

where h_1 denotes the value of h at the first-order line. Since the regular contributions in (4) and (9) are equal, it is possible in principle to separate it from the singular terms. There are, however, several complications which would be encountered in the use of (9) which do not arise when (8) is used. First, since t is positive in (4), but negative in (9), the difference $h_{\lambda} - h_1$ is given by

$$h_{\lambda} - h_1 = 2 \left(\frac{dh}{dt}\right)_t |t| + (D' - F)|t|^{\phi}. \quad (10)$$

Thus the slope of the phase boundary $(dh/dt)_t$ must be estimated before (10) can be used to determine ϕ . Second, (10) requires the comparison of measurements made at different values of T (but at the same value of $|t|$). Because of these complications, it would appear that the use of (10) is intrinsically less accurate than the use of (8) in the determination of ϕ . However, (10) may be useful in cases where the path $M = M_t$ cannot be determined.

III. EXPERIMENTAL RESULTS

The data tabulated by Goellner, Behringer, and Meyer¹³ and Riedel, Meyer, and Behringer⁶ has

been used to estimate $\Delta_{X_t} - \Delta_\lambda$ as a function of T for ^3He - ^4He mixtures.¹⁴ Here and below we use the conventional notation for helium mixtures and denote the nonordering field and density by Δ and X , respectively (i.e., $H - \Delta$ and $M - X$). Δ_{X_t} is the value of Δ along the path $X = X_t$ and Δ_λ is its value on the λ line. [The reduced quantities $(\Delta_{X_t} - \Delta_t)/\Delta_t$ and $(\Delta_\lambda - \Delta_t)/\Delta_t$ cannot be estimated since Δ_t is not known.⁶] A plot of $(\Delta_{X_t} - \Delta_\lambda)^{1/2}$ vs T is shown in Fig. 2.¹⁵ The data are plotted in this way so as to test the theoretical prediction^{8,12} $\phi = 2$, and we see that the results are in good agreement with this prediction. Even though the data correspond to fairly large values of t , we note that the scaling region in helium mixtures has been found to extend up to $T \approx 1.25$ K,⁶ and therefore (8) should be valid up to approximately this temperature. It is interesting that the data seem to follow the asymptotic behavior out to at least 1.40 K. This may be due to partial cancellation of the higher-order terms in (4) and (7) when the difference (8) is taken. A least-squares fit to the data shown in Fig. 2 with the value of T_t constrained to lie within the limits determined in Ref. 13 gives $\phi = 1.95 \pm 0.08$ where the quoted uncertainty is the standard error.¹⁶ This is in good agreement with the value $1/\phi = 0.48 \pm 0.03$ determined by indirect means.⁶

Fig. 3 shows a plot of $(h_{M_t} - h_\lambda)^{1/2}$ vs T for dysprosium aluminum garnet with H along the [110] direction. The data are taken from the work of Giordano and Wolf,^{1,17} and are all from within the scaling region. From Fig. 3 we see that the data are quite consistent with the theory although the scatter is somewhat larger than in Fig. 2. It should be pointed out however that in terms of

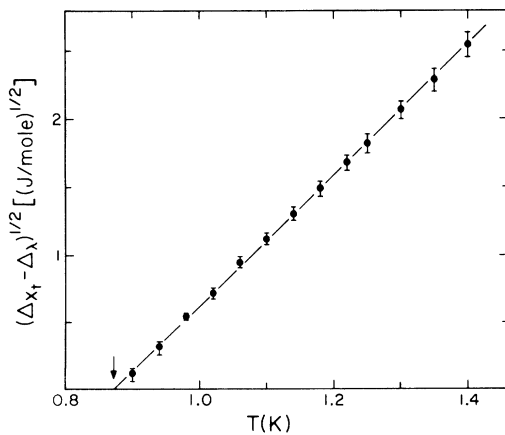


FIG. 2. Plot of $(\Delta_{X_t} - \Delta_\lambda)^{1/2}$ vs T for ^3He - ^4He mixtures. The arrow indicates the value of T_t , 0.872 K, as determined in Ref. 13.

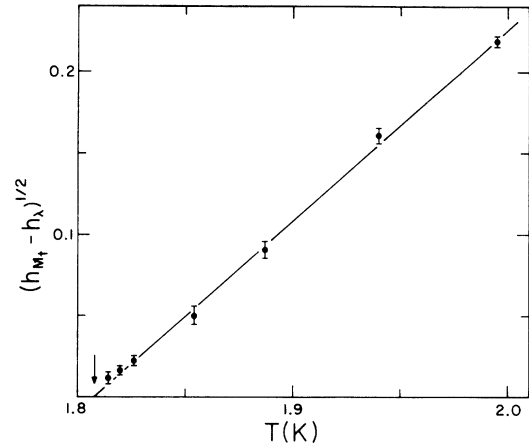


FIG. 3. Plot of $(h_{M_t} - h_\lambda)^{1/2}$ vs T for dysprosium aluminum garnet with H along [110]. The arrow indicates the value of T_t , 1.808 K, as determined in Ref. 1.

reduced temperature units, t , all of the data in Fig. 3 lie within the range covered by only the first two points in Fig. 2. A fit to the data in Fig. 3 with the value of T_t constrained to be within the limits¹⁸ determined in Ref. 1 gives $\phi = 1.95 \pm 0.11$.

In summary, it has been shown how the tricritical crossover exponent ϕ can be determined directly from experimental data. Previously published data for ^3He - ^4He mixtures and dysprosium aluminum garnet have been used to estimate ϕ , and the results for both systems are in good agreement with the theory. The method of analysis is quite straightforward and should prove useful in the analysis of other tricritical-point experiments.

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APPENDIX

It will now be shown that the path $M = M_t$ in the g_1 - g_2 plane is given by (6). Let M_s denote the singular part of M . Differentiating both sides of (1) with respect to H and setting the arbitrary parameter l equal to $|g_2|^{-1}$ gives

$$\begin{aligned} |g_2|^{-\phi(2-\alpha_t)} M_s(g_1, g_2) &= |g_2|^{-\phi} \frac{\partial g_1}{\partial H} F_{s1}(g_1/|g_2|^\phi, \pm 1) \\ &+ |g_2|^{-1} \frac{\partial g_2}{\partial H} F_{s2}(g_1/g_2^\phi, \pm 1), \end{aligned} \quad (\text{A1})$$

where the subscripts 1 and 2 on F_s denotes differentiation with respect to the first or second argu-

ment, and the argument ± 1 indicates $g_2 \gtrless 0$. Rearranging and using the scaling relation⁴ $\beta_u = \phi(1 - \alpha_t)$ gives

$$M_s(g_1, g_2) = |g_2|^{\beta_u} \left(\frac{\partial g_1}{\partial H} F_{s_1}(g_1/|g_2|^\phi, \pm 1) + |g_2|^{\phi-1} \frac{\partial g_2}{\partial H} F_{s_2}(g_1/|g_2|^\phi, \pm 1) \right). \quad (\text{A2})$$

Inherent in scaling theory and hence (1) is the assumption⁴ $\phi > 1$, so just as is the case for the equation of state near an ordinary critical point,¹⁹ the second term in the large parentheses in (A2) will become negligible near the tricritical point. However, this term will be included in the following, so that we can estimate the functional dependences of the correction terms to (6).²⁰

To get an expression for M , the regular part of M must be added to (A2). The regular part of M can be expanded about the tricritical point in a power series in g_1 and g_2 . Sufficiently near the tricritical point we need keep only the terms linear in g_1 and g_2 ; we have

$$M - M_t = Ag_1 + Bg_2 + |g_2|^{\beta_u} \left[\frac{\partial g_1}{\partial H} F_{s_1} \left(\frac{g_1}{|g_2|^\phi} \right) + |g_2|^{\phi-1} \frac{\partial g_2}{\partial H} F_{s_2} \left(\frac{g_1}{|g_2|^\phi} \right) \right], \quad (\text{A3})$$

where we have condensed the notation for $F_{s_1}(x, \pm 1)$ to $F_{s_1}(x)$, etc., for F_{s_2} . From (3) $\partial g_1/\partial H$ and $\partial g_2/\partial H$ are analytic functions of h and t , so to lowest order we can write

$$\begin{aligned} \frac{\partial g_1}{\partial H} &= a_0 + a_1 g_1 + a_2 g_2, \\ \frac{\partial g_2}{\partial H} &= b_0 + b_1 g_1 + b_2 g_2. \end{aligned} \quad (\text{A4})$$

The equation for the path $M = M_t$ in the g_1 - g_2 plane is found from the solution of (A3) with $M = M_t$. It will now be shown that this solution is given by

$$g_1 = E |g_2|^\phi (1 + R). \quad (\text{A5})$$

R is a correction term and we will show that R is of order g_2^ϵ with $\epsilon > 0$. Substituting (A4) and (A5) into (A3) with $M = M_t$ gives

$$\begin{aligned} 0 &= a_0 |g_2|^{\beta_u} F_{s_1}(E + ER) + Bg_2 + AE |g_2|^\phi (1 + R) \\ &\quad + b_0 |g_2|^{\beta_u + \phi - 1} F_{s_2}(E + ER) \\ &\quad + a_1 |g_2|^{\beta_u} g_2 F_{s_1}(E + ER), \end{aligned} \quad (\text{A6})$$

where we have dropped terms of higher order in g_2 , which as will be seen can be neglected in the following. Away from the critical line, F_{s_1} and F_{s_2} can be expanded to give

$$\begin{aligned} 0 &= |g_2|^{\beta_u} [a_0 F_{s_1}(E) + CR] + Bg_2 + AE |g_2|^\phi (1 + R) \\ &\quad + b_0 |g_2|^{\beta_u + \phi - 1} [F_{s_2}(E) + C'R] \\ &\quad + |g_2|^{\beta_u} g_2 [a_1 F_{s_1}(E) + C''R], \end{aligned} \quad (\text{A7})$$

where C , C' , and C'' are constants.

As the solution for R depends upon the relative values of β_u and ϕ , we will for simplicity discuss several possible cases separately. If $1 > \beta_u > 0$ and $\beta_u + \phi > 2$, then we need consider only the first two terms in (A7). (A7) is then satisfied to order g_2 when

$$F_{s_1}(E) = 0, \quad (\text{A8})$$

$$R = -(B/C) |g_2|^{1-\beta_u}. \quad (\text{A9})$$

(A8) determines the value of E ,²¹ and from (A9) we see that R vanishes as $g_2 \rightarrow 0$.

Next, consider the case $\beta_u = 1$, $2 > \phi > 1$. Now only the last term in (A7) can be neglected. (A7) is then satisfied to order g_2^ϕ when

$$F_{s_1}(E) = -B/a_0, \quad (\text{A10})$$

$$R = -\{[AE + b_0 F_{s_2}(E)]/C\} |g_2|^{\phi-\beta_u}. \quad (\text{A11})$$

(A10) determines E , while (A11) shows that once again R vanishes as $g_2 \rightarrow 0$.

Similarly, one can show that for $\beta_u = 1$, $\phi > 1$, the solution to (A7) is given by (A5). It is interesting that although the regular part of the free energy can affect the value of E in (A5), to lowest order in g_2 it does not affect the form of the result. The presence of R in (A5) will lead to terms of order $t^{\phi+\epsilon}$ in (7). Since as we have shown, $\epsilon > 0$, these terms will be negligible as $t \rightarrow 0$.

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¹See, for example, N. Giordano and W. P. Wolf, Phys. Rev. Lett. **35**, 799 (1975), and references contained therein.

²See for example, H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Oxford U.P., New York, 1971).

³We use here the notation proposed in Ref. 4. Note that ϕ is related to the crossover exponent ϕ_t defined by Riedel (Ref. 5) by $\phi = 1/\phi_t$.

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- ¹¹Here and in the following all logarithmic corrections will be ignored. These corrections appear to be negligible in all experiments to date, suggesting that they will be negligible here as well. Although the logarithmic corrections for tricritical behavior have been considered in detail in Ref. 8 and Ref. 10, the logarithmic corrections to the shape of the λ line have not yet been calculated. See Ref. 10 for a discussion of this point.
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- ¹³G. Goellner, R. Behringer, and H. Meyer, J. Low Temp. Phys. 13, 113 (1973).
- ¹⁴Riedel, Meyer, and Behringer (Ref. 6) have also analyzed these data, and have found that the paths $\Delta_\lambda - \Delta_t$ and $\Delta_{X_t} - \Delta_t$ can both be described by an equation analogous to (4) with $\phi = 2$. Thus the difference $\Delta_{X_t} - \Delta_\lambda$ must also be consistent with (8) with $\phi = 2$. The purpose of the present analysis is to set quantitative error limits on the value of ϕ as determined from these data.
- ¹⁵The results for $\Delta_{X_t} - \Delta_\lambda$ are derived from the tables of Δ as a function of X and T given in Refs. 6 and 13. As the data in these tables have been smoothed, care is required in the assignment of the uncertainty in $\Delta_{X_t} - \Delta_\lambda$. The authors of Refs. 6 and 13 estimate a $\pm 7\%$ uncertainty in the values of $(\partial\Delta/\partial X)_t$ used in the analysis. As we are concerned here with differences in Δ at constant T , a corresponding uncertainty of $\pm 7\%$ has been assigned to the values of $\Delta_{X_t} - \Delta_\lambda$, in addition to the uncertainties which arise from errors in X_t and the location of the λ line. This 7% uncertainty is the major contribution to the error bars shown in Fig. 2, except for $T < 1.02$ K, where the contributions from all three sources are approximately equal.
- ¹⁶The value of T_t permitted in the fit was 0.872 ± 0.003 K (see Ref. 13). G. Ahlers and D. S. Greywall [Phys. Rev. Lett. 29, 849 (1972)] and P. Leiderer, D. R. Watts, and W. W. Webb [Phys. Rev. Lett. 33, 483 (1974)] have reported slightly lower values of T_t . Ahlers and Greywall give $T_t = 0.867$ K, and Leiderer, Watts, and Webb report $T_t = 0.866 \pm 0.001$ K. If these values of T_t are used in the fit the value of ϕ is increased by 0.05 and 0.06, respectively.
- ¹⁷N. Giordano and W. P. Wolf, in Proceedings of the 21st Conference on Magnetism and Magnetic Materials—1975, AIP Conf. Proc. 29, 459 (1976).
- ¹⁸The value T_t permitted in the fit was 1.808 ± 0.002 K.
- ¹⁹M. E. Fisher, in *Critical Phenomena, Proceedings of the Enrico Fermi Summer School of Physics, Varenna, 1970* (Academic, New York, 1971).
- ²⁰One also expects that the correction terms to (1) will yield corrections to (A2) of the same order in g_2 as the second term in the brackets. For the purposes of the present work, it can be assumed that all of these terms have been absorbed into F_{32} .
- ²¹Just as in the analogous scaling functions for an ordinary critical point (see Ref. 19), one can show that $F_{s1}(x)$ diverges to $\pm\infty$ as $x \rightarrow \pm\infty$. Since the branch of F_{s1} for $g_2 > 0$ is continuous (M is continuous for $g_2 > 0$), this guarantees that a solution to (A8) exists.