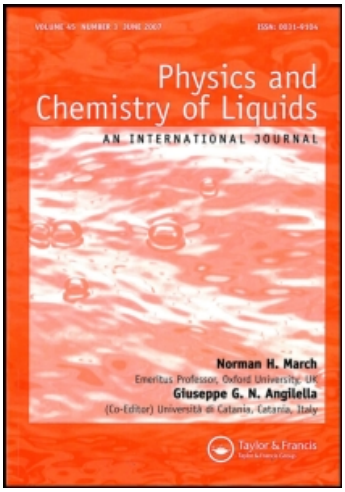

Physics and Chemistry of Liquids
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646857

Heuristic treatments of tricritical behaviour as observed in ^{4}He-^{3}He mixtures and in polymeric assemblies

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Online publication date: 14 April 2010

To cite this Article March, N. H. and Zhang, Z. D.(2010) 'Heuristic treatments of tricritical behaviour as observed in ^{4}He-^{3}He mixtures and in polymeric assemblies', Physics and Chemistry of Liquids, 48: 2, 279 — 284
To link to this Article: DOI: 10.1080/00319100903296121
URL: http://dx.doi.org/10.1080/00319100903296121

Physics and Chemistry of Liquids
Vol. 48, No. 2, April 2010, 279–284

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Heuristic treatments of tricritical behaviour as observed in ^{4}He–^{3}He mixtures and in polymeric assemblies

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(*Received 24 August 2009; final version received 29 August 2009*)

Tricritical behaviour in three dimensions is accessible to experiment in, for example, ^{4}He–^{3}He mixtures and some polymeric assemblies. Here, by largely heuristic arguments, which appeal however to some specific model Hamiltonians, we indicate how one can understand the gist of the experimental results near the tricritical point in the systems cited above. We point out that the spin fluctuations at the tricritical point are much weaker than those at the critical point, which leads to the conclusion that the tricritical behaviours can be described well by the mean-field exponents with logarithmic corrections, at $d = 3$.

Keywords: tricritical behaviour; tricritical exponents; ^{4}He–^{3}He mixtures; polymeric assemblies; Ising model

1. Experimental background and outline

In early experimental studies of ^{4}He–^{3}He mixtures, the superfluid transition, termed the λ-point, at 2.1 K for pure ^{4}He, was found to be lowered by the addition of ^{3}He. The result is a λ-line, which goes back at least to Abraham *et al.* [1]. Some years later, Walters and Fairbank [2] observed experimentally that below around 0.87 K the mixture separates into two coexisting liquid phases. We have redrawn in Figure 1 the results of Graf *et al.* [3]: the dashed line separates the normal fluid from the superfluid and the intersection shown marks the tricritical point. Other well-studied experimental systems exhibiting a tricritical point are some binary mixtures involving liquid sulphur: e.g. benzene or triphenylmethane plus sulphur. For the latter mixture, we have redrawn in Figure 2 the experimental phase diagram measured by Larkin *et al.* [4]. This second class of mixtures chosen here is, at least partially, motivated by the recent work in this Journal involving disordered sulphur by Angilella and March [5], sulphur together with silicon representing two of the sparse number of existing one – component glasses. Some further discussion of the structure factor of liquid sulphur is given in the book by March and Tosi [6], rings and chains being

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ISSN 0031–9104 print/ISSN 1029–0451 online

DOI: 10.1080/00319100903296121
http://www.informaworld.com

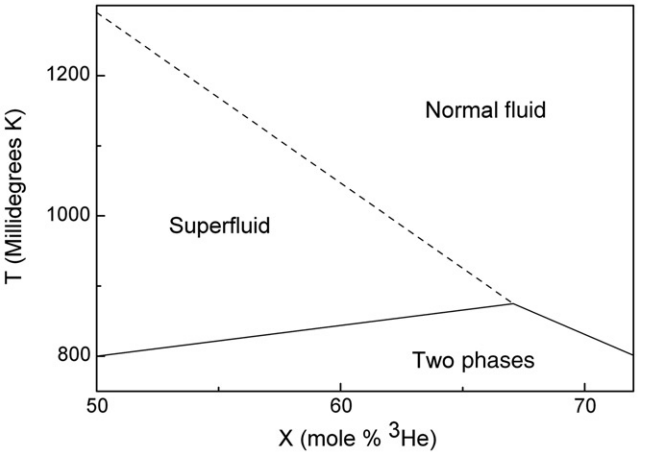

Figure 1. Phase diagram of ^{4}He–^{3}He liquid mixtures. This figure was redrawn according to Figure 2 of Graf *et al*. [3]. The dashed line represents the onset of superfluidity, while the solid line is for the phase separation and the intersection marks the tricritical point.

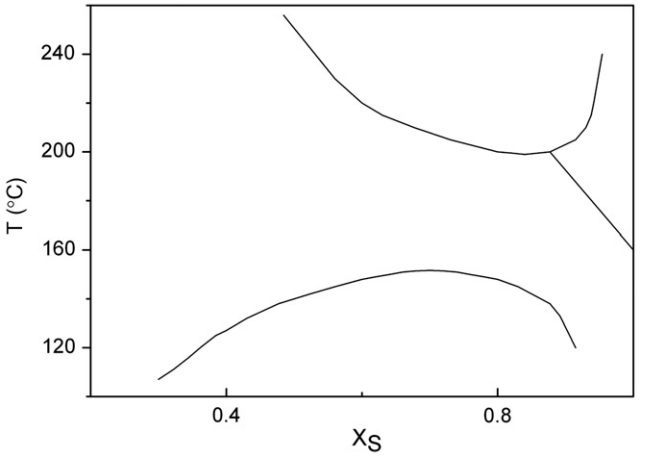

Figure 2. Phase diagram for the mixture of sulphur with triphenylmethane. This figure was redrawn after Figure 8 of Larkin *et al*. [4].

featured there. We shall return to polymeric chains below, in discussing model interpretations related to the phase diagram of the mixture shown in Figure 2.

The outline of this article then is as follows. Section 2 summarises some heuristic considerations, beginning with models based on polymer chains, going back to de Gennes [7]. Various theoretical treatments will be referred to, including some controversial matters which seem now to be resolved through applications of the renormalisation group and also field theoretical treatments of models. Section 3 contains first a short summary and then some proposals for future work which should prove fruitful. In the appendix, reference is made to three model Hamiltonians which already have had an impact on this area of tricriticality.

2. Heuristic treatments appropriate to the tricritical point

We shall begin by recalling in general terms what is set out in more detail in the appendix. de Gennes [7] led the way in relation to tricritical behaviour involving polymer chains (compare Figure 2) by exposing an analogy with magnetic systems. This can be formalised into a d-dimensional Hamiltonian set out in Equation (A1), which involves intimately an n-component continuous spin variable S_α (x). This work by de Gennes [7] was closely followed by the study of Stephen and McCauley [8] who employed the Wilson classical spin model to determine the tricritical exponents α, γ and η, by expansion in ϵ related to dimensionality by $d = 3 - \epsilon$. Their results read, for an n-component classical field:

$$\alpha = \frac{1}{2} + \frac{1}{2}\epsilon + O(\epsilon^2), \tag{1}$$

$$\gamma = 1 + \frac{5}{8}\frac{(n+2)(n+4)}{(3n+22)^2}\epsilon^2 + O(\epsilon^3), \tag{2}$$

and

$$\eta = \frac{1}{12}\frac{(n+2)(n+4)}{(3n+22)^2}\epsilon^2 + O(\epsilon^3). \tag{3}$$

A decade or so later, Duplantier [9] discussed tricritical polymer chains for $d \leq 3$. There had been some early controversy (see e.g. [10]). In [9], the tricritical exponent ν is written as $\nu = \frac{1}{2} + O(\epsilon^2)$ where $d = 3 - \epsilon$. Duplantier adds the scaling relation $\nu = (2 - \eta)^{-1}\gamma$ and then for $\epsilon = 0$ corresponding to $d = 3$ concludes that $\nu = 1/2$, $\eta = 0$, $\gamma = 1$, the later two values agree with Equations (3) and (2) given above by Stephen and McCauley when $\epsilon = 0$. Hager [11] then performed ϵ-expansions of tricritical exponents ν and η up to ϵ^3. The conclusion seems clear: the Wilson renormalisation group plus field theory treatments (e.g. [12–14]) are leading to the correct tricritical exponents, with only logarithmic corrections to mean-field results for $d = 3$.

3. Summary and proposed directions for further study

We have focussed first on two experimentally determined phase diagrams concerned with tricritical points. Figure 1 shows the case of ^{4}He–^{3}He mixtures, in which the λ-point of ^{4}He is lowered by mixing with ^{3}He. As a second example, we have chosen a mixture involving sulphur, one of the reasons being the current interest in single – component glasses: of which sulphur and silicon are two but a few presently known disordered assemblies of this kind. Thus in Figure 2, we have selected the mixture of sulphur with triphenylmethane. Section 2 then summarises some models which have been proposed, especially related to Figure 2 in which polymeric chains are anticipated, for tricritical exponents. After some early controversy, briefly referred to in Section 2, the essence of Equations (2) and (3) for tricritical exponents, going back at least to Stephen and McCauley [8] is confirmed. As to further directions, refined treatments using density functional theory would seem of ongoing interest for ^{4}He–^{3}He mixtures especially.

But, to conclude, it remains of considerable interest to give simple, first principles explanations as to why, for critical exponents, the crossover dimension above which

mean-field values become appropriate is at $d=4$, whereas for the tricriticality discussed in the present article, the mean-field exponents are already appropriate, though now with logarithmic corrections, at $d=3$. Note that for critical exponents of an ordinary second-order transition, the recent theoretical work gives $\alpha=0$, $\beta=3/8$, $\gamma=5/4$, $\delta=13/3$, $\eta=1/8$ and $\nu=2/3$ for the 3D Ising model [15], whereas the mean-field values are $\alpha=0$, $\beta=1/2$, $\gamma=1$, $\delta=3$, $\eta=0$ and $\nu=1/2$. It has been found that the tricriticality exists in various magnetic systems with diluted mixed spins [16–19], four-spin interaction [20], strongly competing anisotropy [19,21–25] and/or competing magnetic fields [26].

We try to understand this issue as follows: first, we inspect the mean-field theory itself. The mean-field theory treats all the models the same, by dealing with every spin, bond, etc. in an average manner and neglecting all spin fluctuations in the order parameter in which nearby parts of the system, while remaining interrelated, do something different from the average. Therefore, the mean-field theory can be valid at the region, far from the critical point for $d\leq 3$, where fluctuations are weaker. There is a characterised temperature region $|t|=|T-T_c|$, within which the mean-field theory loses its validity since there are strong fluctuations near the critical point. It is estimated that the $|t|$ is in order of 10^{-2} for magnetic systems, but about 10^{-10} for superconductivity. Thus the mean-field theory, such as Bardeen–Cooper–Schrieffer (BCS) theory [27,28], works quite well for the critical behaviour of superconductivity. For the tricriticality in the ^{4}He–^{3}He mixtures, the characterised temperature region $|t|$ should be much smaller than it is near the critical point. This is the first reason why the mean-field theory works in the present topic for $d=3$. Secondly, the first-order phase separation and the onset of superfluidity of second order are two phase transitions known to occur in liquid ^{4}He–^{3}He mixtures. At the tricritical point, the mixture separates into two coexisting liquid phases so that in principle, the system at this point possesses the characters of both the first-order and the second-order phase transitions. The emerging of the first-order phase transition would certainly suppress the spin fluctuations at the critical point of the second-order phase transition. It was understood that the tricritical transition differs from an ordinary second-order transition by the existence of an additional density with critical fluctuations [29] and that the tricritical point can be characterised as the simultaneous instability point of the system to two types of critical fluctuations [14]. Thirdly, in the systems with magnetic mixtures, the spin number of at least one of the mixtures should be larger than 1/2. It is known that a system with higher spins would behave more classically, which can be better described by the mean-field theory, because high spins suppress the spin fluctuations (a similar analysis could be done also for the effect of high anisotropy). From the analysis above, the spin fluctuations at the tricritical point are much weaker than those at the critical point so that the tricritical behaviours can be described well by the mean-field exponents with logarithmic corrections, at $d=3$.

Acknowledgements

N.H. March wishes to acknowledge that his contribution to this article was brought to fruition during a visit to ICTP, Trieste. He is grateful to Prof V.E. Kravtsov for the very stimulating atmosphere in which this work was carried out, and also for his generous hospitality. N.H. March also acknowledges on-going affiliation with the University of Antwerp (UA) plus

partial financial support through BOF-NOI, made possible by Profs D. Lamoen, and C. Van Alsenoy. Z.D. Zhang acknowledges the support of the National Natural Science Foundation of China (under grant numbers 50831006 and 10674139).

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Appendix

Three Hamiltonians relevant to tricriticality

We summarise in this appendix three Hamiltonians that have proved valuable in discussing tricritical points.

The first of these stemmed from an analogy between polymer chains, such as exist in the sulphur mixture shown in Figure 2 of the main text, and magnetic systems, which was pointed out by de Gennes [7]. This was employed in the study of Stephen [30] and that referred to above [8], and forms a basis for treating critical and tricritical behaviour in d dimensions. The effective Hamiltonian reads

$$H = \int d^d x \left[\frac{r_0}{2} \sum_{\alpha=1}^{n} S_\alpha^2(x) + \frac{1}{2} \sum_{\alpha=1}^{n} |\nabla S_\alpha(x)|^2 + \frac{u_4}{4!} \left(\sum_{\alpha=1}^{n} S_\alpha^2(x) \right)^2 + \frac{u_6}{6!} \left(\sum_{\alpha=1}^{n} S_\alpha^2(x) \right)^3 + \cdots \right]. \quad \text{(A1)}$$

In the above Equation (A1), $S_\alpha(x)$ is an n-component continuous spin variable. de Gennes noted that as $n \to \infty$ the Hamiltonian (A1) describes a self-avoiding random walk: a polymer chain with excluded volume (see also [31]).

The second Hamiltonian of interest goes back at least to Blume–Emery–Griffiths (BEG) [32]. The total Hamiltonian reads

$$H = -J \sum_{\langle i,j \rangle}^{N} S_i S_j - K \sum_{\langle i,j \rangle} S_i^2 S_j^2 + \Delta \sum_i S_i^2 - N(zK_{33} + \mu_3). \quad \text{(A2)}$$

This has become of wider interest recently in the different context of negative specific heat (see e.g. [33]). There, the BEG Hamiltonian was also utilised to compare the relation between the magnetisation and internal energy of the itinerant systems Fe and Ni.

The final Hamiltonian referred to in this appendix is that proposed by Ashkin and Teller [34] (see also [35]). It is shown in [35] that the four-component statistics model of Ashkin and Teller is equivalent to a special case of a staggered eight-vertex model [36], while the eight-vertex model is equivalent to an asymmetrical Ashkin–Teller model. This has been utilised in the context of tricriticality by Riedel and Wegner [14] who also refer incidentally to a generalisation of the BEG model in relation to the ^{4}He–^{3}He mixtures displayed in the present Figure 1.