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CRITICAL PHENOMENA 1965-1972: QUESTIONS AND ANSWERS*

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ABSTRACT: Recent advances on the field of Critical Phenomena are discussed. We restrict the discussion to three main problems: a) What is the nature of the singularities in the thermodynamic properties near the critical point? b) How do the exponents which express these singularities depend on the physical nature of the system? and c) Which transport properties are anomalous near the critical point?.

INTRODUCTION

When I was invited to give a talk on Critical Phenomena, I thought that the best thing to do would be to evaluate the main advances that were made in answering the questions that were raised at the 1965 Critical Phenomena Conference held at Washington, D.C.[1]. I believe that at this conference the relevant problems in this field were exhibited in a unified way and precise questions were stated. These questions are the basic material for this paper.

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At that time the re-interest in the study of Critical Phenomena came first from the fact that widely different systems or phenomena manifest similar behaviour as one approaches the critical point; to mention a few, vapor-liquid transition, mixtures, magnetism, superconductivity, superfluidity. Although such similarities could be understood through the Classical theories of 2nd order transitions[2] (Van der Waals, mean field type theories) we still were puzzled by the singular behaviour of analogous properties in different systems.

We wanted to know if there was a common reason which could explain such behaviour. We also knew that the classical theories failed to predict the experimental data[3] from a quantitative point of view and we felt too that some fundamental fact underlaid these experimental discrepancies.

Of course, to support this last conjecture we had at that time the Onsager solution to the two dimensional Ising model[4] and also all the numerical results achieved by the group at King's College, London[5], using series summation methods. Both gave significant differences from the classical theories. In addition to these reasons which motivated our interest, we had also such spectacular features of critical phenomena as critical opalescence, which at that time were only partially understood[6].

In re-reading the introduction I wrote for the Proceedings of the 1965 Conference I felt that we could regroup all the questions into three main questions and three subsidiary ones. There were also other questions raised at that meeting; however, these became spurious due to experimental mistakes or other reasons and therefore we will not mention them here.

The first question is a very broad one: what is the nature of the singularities in the thermodynamic and other quantities near the critical point? In other words, are the singularities of the exponent type, logarithmic or other type of singular behaviour?

The second question has to do with the physical nature of the exponents: how do the exponents which express these singularities depend on the physical nature of the system? By this question we mean whether the exponents depend on the specific model we choose, say, a Heisenberg or an Ising Model, or on the specific form of the intermolecular forces. Are the exponents affected by taking square lattices instead of triangular ones for the purpose of our calculations? What is the role of the space dimension? What is the order of magnitude of these effects? Are there any specific quantum effects on Critical Phenomena? At the 1965 meeting there was a paper[7] showing differences between the critical behaviour of liquid helium and that of xenon. In a few words, do the exponents remain constant for all systems or not? If they do, what is the fundamental reason underlying this fact? If they are not the same for all systems, what would be the most adequate scheme for calculating them?

The third question is: which transport properties are anomalous near the critical point and why? This question should be taken in a very broad sense. Here we are concerned with all aspects of non-equilibrium phenomena near the critical point.

The subsidiary questions I have chosen on the basis of personal interest and also because they are still open questions. The first one is: How does the pair correlation function behave near the critical point? Here we are interested in the deviations from the Ornstein-Zernike theory. I might say that I have been a bit biased choosing this question because this is my personal area of research on Critical Phenomena.

The second question is: What is the experimental behaviour of the specific heat of various systems near the critical point? This question was formulated in 1965 in a somewhat incorrect way; that is, is the logarithmic behaviour of the specific heat universal for all critical systems? However we now know that this question was wrongly posed. The third question is: How wide ranging are the analogies among critical phenomena?

We now come to the answers or progress made towards an answer to all these questions.

II. NATURE OF THE SINGULARITIES NEAR THE CRITICAL POINT

The most significant development in this area was the concept of *scaling*. Nowadays the idea of scaling can be stated in a very simple way, that is: *The Gibbs potential is a generalized homogeneous function near the critical point*. This brings about a unification of the behaviour of all thermodynamic properties near the critical point, not only along certain lines but through out all the thermodynamic phase space. Scaling is associated with the names of Widom[8], Domb and Hunter[9] and Kadanoff[10]. These authors arrived at this idea by means of different arguments. Widom reached the idea of scaling by continuing his phenomenological studies on the nature of the thermodynamical properties near the critical point. Domb and Hunter noticed certain characteristics of their series expansions which could be reinterpreted to imply scaling behaviour. Finally, Kadanoff gave a heuristic argument based on the Ising model. Instead of considering the interaction between the sites on this lattice he assumed an interaction between cells. However he preserved the form of the Hamiltonian and then asked for the Gibbs potential to have the same functional relationship with respect to the new Hamiltonian parameters except for a multiplying constant. With this and some other physical assumptions he was led to scaling.

The most important achievements of scaling were first to produce relationships among the exponents and secondly, to make specific predictions concerning the form of the equations of state near the critical point. One of the significant features of this second point was the reduction of the two dimensional thermodynamic data to a one dimensional relationship.

This was done by Kouvel[11] et al and Fisher[12] and later by myself, Levelt-Sengers and Vicentini Missoni[13] for a number of liquid vapor systems as well, and with Josephs[14] for magnetic systems.

However this concept of scaling is not a complete answer to the question of what is the nature of the singularities, specially when we are concerned with a neighborhood of the critical point. More recently a number of authors have attempted to extend the idea of scaling to find what are the next terms in an asymptotic expansion in which scaling is the most important term. I must emphasize that this is not a pedantic problem but one which has to be understood for true analysis of experimental data, specially for that related to weak divergences. We might mention some experimental work concerned with these higher order terms as that of Wallace and Meyer[15] on the density dependence of chemical potential difference between the liquid and gas phases on the critical isotherm of H_e^3 and H_e^4 as well as that of Levelt-Sengers[16] et al, on the coexistent curve of CO_2. From the theoretical point of view we might mention a very important paper in this direction by Griffiths and Wheeler[17], where it was emphasized that near the critical point the behaviour of the system is characterized by a principal direction in the space of the intensive variables ("fields"), which can be identified in a single component system with the tangent to the coexistence curve at the critical point. Green, Cooper and Levelt-Sengers[18] proposed an expansion for the thermodynamic properties in the critical region which extended beyond the range of ordinary scaling. Their work was based on a generalization of the Josephson[19]-Schofield[20] parametric representation of thermodynamic scaling by introducing a new critical exponent and relating it to the existing ones by means of the invariance principle stated in the work by Griffiths and Wheeler. Another important paper is that by Mermin and Rehr[21] in which they argue that the conclusion that Green, Cooper and Levelt-Sengers arrived at in their paper about the singularity of the derivate of the coexistence curve-diameter, could be reached without making any reference to the equation of state and showing that this follows directly from the Griffith and Wheeler hypothesis. We could mention other developments on the nature of the singularities, yet I think we have mentioned the more relevant ones.

III. CRITICAL EXPONENTS AND THE PHYSICAL NATURE OF THE SYSTEMS

One of the most exciting features of critical phenomena is how similar quite different systems seem to behave near their critical point, for instance, liquid helium near its λ point behaves like argon near its critical point.

Two different lines of thought have developed in order to answer this question. One makes the assumption of universality and the other does not make any reference to this concept. The concept of universality states that within a good approximation, completely different systems have the same critical behaviour. In a concrete way we would say that all critical exponents assume the same values for different systems. However this is not entirely correct; Griffiths[22] pointed out that there are various causes for the variation of the critical exponents such as: lattice dimensionality, the symmetry of the order parameter, the range of interaction, etc. Nonetheless if the parameters in the Hamiltonian do not produce a basic change such as those mentioned above, this "restricted" universality is a good working hypothesis. In other words we could restate the concept of universality as follows: *All systems of a similar type have the same behaviour near the critical point.*

One of the most prominent exponents of the idea of universality is Kadanoff[23]. He expresses this idea by making use of the "reduction hypothesis" which suggests that a product of any two nearby fluctuating local quantities is expected to behave as a linear combination of all other local variables. In other words only a finite number of local fluctuating variables are necessary to describe critical phenomena. The coefficients of the expansion of these products in terms of the relevant variables will describe an algebra. He later shows a scheme of how this reduction algebra can determine the exponents. We may hope that inherent symmetry properties of each system will introduce enough constraints on the algebra that the values of the exponents are determined. Until now, this has been only done in the case of the two-dimensional Ising model. Therefore from Kadanoff's point of view the changes from one set of values of the critical exponents to another, happens in a discrete way because they are caused by a change in the symmetry of the system.

A theoretical study which favors the ideas of universality is the work of Wortis[24] and associates, in which a very extensive series expansions of the properties of a system was performed. This system was characterized by a Hamiltonian with two parameters in which one could go continuously from a Heisenberg to an Ising model. They found that the values of the

the equilibrium correlation function with no attempt to solve the problem of the equilibrium critical phenomena. We must name a number of authors in this connection. In particular the concept of dynamical scaling is associated with the names of Halperin and Hohenberg[31] who extended the concept of scaling to the time correlation function which provided valuable information about the transport coefficients. However this theory is not capable of predicting the magnitude of the thermal conductivity exponent nor the magnitude of the Botch-Fixman coefficient or the value of the critical region line width exponent.

Another approach is the mode-mode coupling theory, which succeeds in predicting the values of the critical-point exponents for the transport coefficients and shows which of these are expected to diverge. Many authors have contributed in formulating this theory, the first one was Fixman[32] he was followed by Kawasaki[33], Kawasaki and Tanaka[34], Deutch and Zwanzig[35], Mountain and Zwanzig[36], Villain[37], Ferrell[38], and Kadanoff[39a] and Swift[39b]. Experimentally both theories have come up with very important confirmations. I should mention an extensive study of the inelastic neutron scattering of rubidium manganese fluoride[40] which provided the time correlation function for this composite near its Curie point. This was in very good agreement with the prediction of dynamic scaling among other things. Another experimental technique which has been of great importance in the study of Critical Phenomena is the inelastic scattering of light. This technique was made possible by the existence of the laser and the combination of optic and electronic means. The first successful realization of this technique was reported in 1965[41] and since then the temporal nature of the fluctuations responsible for critical opalescence are now pretty well understood due to the work of Swinney and Cummins[42], Benedek[43], Ford[44], Sengers[45] among others. These results are much in agreement with the theory of Kawasaki (1970). Just recently one contradiction with respect to the existing theory was resolved in favor of theory; that is, the difference in the width of the Rayleigh line above and below the critical point of sulphur hexafluoride. Large differences in these two measurements which no theory could explain were thought to exist, but later on these disappeared through new experiments[46] and theoretical explanations proposed by Sengers[47]. We can conclude that this question is in a very satisfactory state and we could not ask for better answers to it.

exponent would only change when the basic symmetry of the Hamiltonian was changed and not otherwise. Although in their method as one approached the change in symmetry the exponent started to change continuously, they adscribe this ambiguity to their method. A counter example to this configuration was given by Baxter[25] who solved a rather peculiar model of a phase transition (the eight-vertex model) in which the exponents dependend continuously on a parameter in the Hamiltonian*. Although this is a very special case of a phase transition this would support the other conflicting view point which states that the behaviour of critical phenomena varies from system to system and would change accordingly with the parameters included in the Hamiltonian characterizing the system. The first to take this point of view were Migdal[26] and Polyakov[27] which by using Quantum Field theoretical methods (Diagrammatic approach) were able to give a proof of scaling as well as diagrammatic expressions for the critical exponents. This idea of Migdal and Polyakov was further pursued by Jona-LaSinio[28] and Di Castro[29] using another Quantum Field theoretical approach, the Renormalization Group method. They manage, by making appropriate assumptions about the analytic behaviour of the vertex function, to predict singularities of the exponent form, where the exponents are given as derivates of the Vertex function.

All the previous arguments are of a formal character. They are able to predict scaling, the exponent form of the singularities, yet they do not come out with a numerical value for the exponents. However, Wilson[30] has recently produced a "tour de Force" by actually calculating these exponents by means of a very sophisticated use of the "Renormalization Group" which agree very well with the results predicted by the experiment. As you can see this question is still a controversial one and the answer is up in the air. To my mind this is one of the most interesting questions in Critical Phenomena, and where exciting things will be happening in the coming years.

IV. TRANSPORT PROPERTIES NEAR THE CRITICAL POINT

Let us now refer to the third question which is perhaps one of the most significant successes since 1965 because in a way we have answered nearly all the questions which were unknown to us at that time. This came about by the combination of well-known principles of non-equilibrium Statistical Mechanics combined with a phenomenological theory (Scaling theory) of

* Kadanoff and Wegner[54] have shown that the Baxter model is the limiting case when the exponents depend continuously on a parameter.

V. SUBSIDIARY QUESTIONS

The first of these questions, as we mentioned before, has to do with the deviations from the Ornstein-Zernike theory of critical opalescence. This theory was proposed in the early days of this century and explained qualitatively the major features of critical opalescence. However we have reasons to believe that this is not the truly correct theory. The Ornstein-Zernike theory predicts a pair correlation function of the Yukawa form $G(r) = [\exp(-kr)]/r$. However we expect that there is a small difference in the exponent of r of the order of .06 to .09 which Fisher denoted by η. That is, we expect a form $G(r) = [\exp(-kr)]/[r^{d-2+\eta}]$ where d is the dimension of the space. If η is not zero I don't believe there are any temperatures and densities for which the Ornstein-Zernike theory could stand under a sufficiently critical analysis of the data. One piece of evidence that we have that η is not zero, are the Buckingham-Gunton inequalities[48]. In 1965 we thought we knew that η was different from zero, but being more critical about it we could say that we did not know that η differed from zero, and is still an open question to actually measure η. Apparently it has been measured for rubidium manganese fluoride[49] and I hope someone will complete a measurement for Neon near its critical point. I might say that in exponents, small exponents are open questions; exponents of the order of .05 to .09 are very hard to measure. Feynman once made a statement which we cannot accept, but has a lot of truth in it: "exponents less than a quarter are zero".

The second question has to do with the experimental behaviour of the specific heats near critical points. In 1965 the evidence seemed to be that all the singularities in the specific heat at constant volume were logarithmic of the type found for Helium, yet closer examination indicated that these singularities were not logarithmic but of the exponent type of behaviour, where the exponents were also of the order of .05 to .07. However there are experimental difficulties, which become very great near the critical point, in order to determine the value of such exponents. This is still an open experimental question.

We finally come to the last subsidiary question. One aspect of critical phenomena on which we were very much interested in 1965 was the analogies among the various types of transitions. We knew that the specific heat at constant volume of liquid Helium near its λ point seemed to behave like the specific heat at constant volume of liquid Argon near its critical point, but we did not know too much of the detail of the analogy between a λ-point and a liquid-vapor transition. I can mention two experiments in this respect in which the order parameter for liquid Helium was measured near its λ-point. The order parameter is, in this case, the condensate wave function. Josephson has pointed out that this order parameter is almost, but not exactly the square root of the superfluid density. Tyson and Douglass[50] on one hand, and Clow and Reppy[51] on the other, measured the superfluid density near the λ-point and found that the behaviour of the square root of the superfluid density with respect to the reduced temperature, had an exponent very close to 1/3 which is the same as the exponent for the order parameter in magnetism (the magnetization), and that of a liquid-vapor transition (the difference between the liquid and gas densities). I should also mention the work of Ferrell[52] who showed that beyond the Bardeen-Cooper-Schrieffer[53] theory of superconductivity which is in the category of the mean field theories, there are, in certain special systems, phenomena which are different from that predicted by the Bardeen-Cooper-Schrieffer theory and can be understood as being analogous to the non-classical behaviour of magnetic and liquid-vapor systems.

This is a very personal review of what has happened from the point of view of one person, but I hope I have not been too biased.

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RESUMEN

Se discuten los últimos adelantos en el campo de los Fenómenos Críticos. Restringimos la discusión a tres problemas principales: a) ¿Cuál es la naturaleza de las singularidades en las propiedades termodinámicas cerca del punto crítico? b) ¿Cómo dependen los exponentes que expresan estas singularidades en la naturaleza física del sistema? y c) ¿Cuáles propiedades de transporte son anómalas cerca del punto crítico?.