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Detection of Laser Light Scattered by Materials near a Phase Transition

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DETECTION OF LASER LIGHT SCATTERED BY MATERIALS NEAR A PHASE TRANSITION

Final Report

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Using light scattering techniques we have measured the correlation function of a binary liquid near the critical point with a precision of 0.2%. The data cover a range of $0.18 < k\xi < 26$ in terms of the scaling variable $k\xi$ and yield the critical exponent values $\gamma = 1.240 \pm 0.007$, $\nu = 0.625 \pm 0.003$ and $\eta = 0.016 \pm 0.007$. The results indicate that binary liquids belong to the same universality class as the Ising model.

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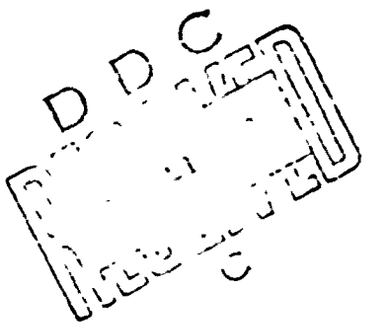

Abstract

A summary is presented of a study of critical fluctuations using laser light scattering techniques under contract N00014-67-A-0239-0014 and N00014-76-C-0304 with ONR. Highlights of the accomplishments are experimental verification of exponential decay of critical fluctuations and of the hypothesis of dynamical scaling, experimental observation of an effect of a viscosity anomaly on the decay rate of the fluctuations and experimental evidence for the principle of universality of the critical correlation function.

Table of Contents

I. Introduction

This report summarizes the research and accomplishments conducted at the University of Maryland under contract N00014-67-A-0239-0014 from August 1970 till August 1975 and under contract N00014-76-C-0304 from August 1975 till August 1976 with the Office of Naval Research.

The research was concerned with studies of fluctuations in materials near a critical point phase transition using laser light scattering technique The research was conducted under the supervision of Professors C.O. Alley and J.V. Sengers as co-principal investigators with Professor R.F. Chang as their main collaborator on this project. The experiments were conducted by Professor Chang in the quantum electronics laboratory of Professor Alley. Professor Sengers supervised the interpretation of the experimental results. During the course of the project the investigators were assisted by two postdoctoral associates, namely Dr. C. Bendjaballah (1972-1973) and Dr. A.J. Bray (1974-1976) and by three graduate students, namely P.H. Keyes, M. Wigdor and H. Burstyn.

II. Summary of Accomplishments

Critical point phase transitions occur in a large variety of different kinds of physical systems such as ferromagnets near the Curie point, antiferromagnets near the Néel point, alloys exhibiting order-disorder transitions (e.g. ammonium chloride), binary liquid mixtures near the critical point of mixing and fluids near the gas-liquid critical point. These diverse physical systems have a number of phenomena in common near their respective critical points. One important common feature is the existence of large thermal fluctuations in all these systems near the critical point.

The main purpose of the research was to study the nature of these critical fluctuations. The fluctuations may be characterized by a correlation length which diverges at the critical point. This implies that in the vicinity of a critical point the correlation length becomes much larger than the range of intermolecular forces. Hence, the fluctuations near the critical point become insensitive to the details of these interactions. It is believed, therefore, that the critical fluctuation behavior in the various different systems has an essential similarity, referred to as the principle of universality of critical behavior.

The existance of diverse physical systems displaying analogous critical phenomena provides the experimenter with the opportunity to choose both the best material and the best technique for studying specific aspects of critical phenomena. The size of the fluctuations in magnetic solids far from the critical point can be readily measured with neutron scattering (such experiments were conducted simultaneously by Professor Minkiewicz and are continued by Professor Lynn at the University of Maryland). However, this technique has serious limitations very close to the critical point because the fluctuations extend over distances much larger than the wavelengths of the neutrons. Moreover, very close to the critical point all experiments on solids are limited by lattice strains which result from impurities, vacancies, etc. On the other hand, fluctuations in gases and liquids, which continuously "anneal" themselves can be studied accurately by laser light scattering techniques. However, experiments near the gas-liquid critical point are severely affected by multiple scattering. Hence, experiments in binary liquids near the critical point, where multiple scattering can be minimized by refractive index matching, seemed to offer the best opportunity for studying the fluctuations in the close vicinity of the critical point. Hence, it was decided to measure the critical fluctuations in the binary liquid

3-methylpentane-nitroethane. The wisdom of this choice has been confirmed by the experimental results as we shall see below.

Here we give a brief survey of the results of the research. For the details of our accomplishments we refer to the publications and reports listed in Section III.

When the experiments were initiated, Professor Koreman made a theoretical analysis of what type of information could be obtained from the application of sophisticated photon counting techniques near the critical point of fluids.[1] At the same time Professor Sengers made an analysis of the nature of the transport processes and the decay rate of the fluctuations near the critical point of fluids on the basis of existing experimental data.[2,3] In particular, he found that the decay rate data could indeed be described in terms of the modern concepts for treating critical point phenomena provided the decay rate was separated in an anomalous and a regular part.

The correlation length, determining the spatial extent of the critical fluctuations, can be measured by determining the intensity of scattered light as a function of the scattering angle. The decay rate can be obtained from the frequency distribution of the scattered light. A theory for the decay rate of the critical fluctuations was originally developed by Kawasaki at Temple University and Ferrell at the University of Maryland. This theory related the decay rate to the correlation length and the shear viscosity of the system. In the literature it was claimed that this theory gave an adequate account of the experimentally observed decay rate. However, due to the difficulty of obtaining accurate light scattering data, and hence accurate data for the correlating length, the theory was always applied using adjustable parameters for the temperature dependence of the correlation length. In our opinion, in order for a crucial test of the theory, it was imperative that the correlation length and the decay rate of the critical fluctuations

both be measured simultaneously for the same physical system. This was the purpose of our first series of experiments completed in 1972 and reported in references 4,5,6. These experiments showed for the first time that the theoretical expressions derived by Kawasaki and Ferrell in fact were insufficient to account for the experimentally observed decay rate of the critical fluctuations. This result was caused, in part, by effects from anomalous viscosity behavior on the decay rate and it led to considerable activity by a number of theoretical investigators at the University of Maryland as well as elsewhere and several refined formulations were proposed yielding improved agreement with the experimental data.[6]

These theories suggested that also the assumption of exponential decay of the fluctuations might break down close to the critical point. With an improved photon counting detection procedure we started to investigate the nature of the time dependence of the critical fluctuations more accurately and we were able to verify the exponential nature of this decay up to very close to the critical point.[7]

Nevertheless, a number of questions remained unanswered. First, although the new refined theories led to an improved agreement between theory and experiment, the various theories (sometimes referred to as mode-coupling and decoupled-mode theories) yielded, in fact, predictions different from each other. However, it was impossible to discriminate between these theories based on the existing experimental capability. Secondly, the long range character of the critical correlation function is characterized by an exponent η which appeared to be a very elusive property to be measured experimentally.[10] Finally, there remained the nagging question whether the difference between the critical exponents deduced experimentally for fluid systems and the theoretical values calculated for the Ising model indicated a small but

fundamental difference between fluid systems and lattice systems. It was evident that in order to answer these questions satisfactorily, considerably more accurate light scattering intensity measurements would be required. This goal has been the object of our research during the past three years and we are pleased that we have succeeded in obtaining light scattering data with a precision of 0.2% which represents an improvement of the prec by one order of magnitude.[11]

When data are to be obtained with such precision, many small effects, previously ignored, must be taken into account. Most important is a precise assessment of the effect of double scattering which was studied extensively.[9]

The new experimental results are reported in the Appendix of this report. The interpretation of these results was greatly facilitated by the fact that Dr. Bray at the University of Maryland had been able to develop an accurate theoretical formulation for the correlation function of the Ising model. When the data were interpreted in terms of this theoretical correlation function, it appeared that the critical exponents deduced from our data for the binary liquid were extremely close to a recent calculation of these exponents for the Ising model. For details we refer to the Appendix. We thus have obtained at the conclusion of the present phase of the project strong experimental evidence that the critical correlation function is indeed universal and the same in fluids and solids. This is the first time that such experimental information for the correlation function of fluids has been obtained. We may confidently expect that our further experimental work on the critical fluctiations in liquids may yield valuable information for fluids and solid systems alike.

III. List of publications and technical reports.

1. "Photon Counting Correlation Functions and the Critical Region," V. Lorenman, Phys. Rev. A2, 449-458 (1970).

2. "Scaling of the Thermal Conductivity near the Gas-Liquid Critical Point," J. V. Sengers and P. H. Keyes, Phys. Rev. Letters 26, 70-73 (1971).

3. "Transport Properties of Fluids Near Critical Points," J. V. Sengers, in "Critical Phenomena," Proc. Intern. School "Enrico Fermi," Course LI, M. S. Green, ed. (Academic Press, New York, 1971), pp.445-507.

4. "Dynamics of Concentration Fluctuations near the Critical Mixing Point of a Binary Fluid," R. F. Chang, P. H. Keyes, J. V. Sengers and C. O. Alley, Phys. Rev. Letters 27, 1706-1709 (1971).

5. "Non-local Effects in the Diffusion Coefficient of a Binary Fluid Near the Critical Mixing Point," R. F. Chang, P. H. Keyes, J. V. Sengers and C.O. Alley, Ber. Bunsenges, physik. Chemie 76, 260-267 (1972).

6. "Concentration Fluctuations in the Critical Region of the Binary Liquid 3-Methylpentane-Nitroethane," P. H. Keyes, R. F. Chang, J. V. Sengers and C. O. Alley, Technical Report No. 73-018 (Department of Physics and Astronomy, University of Maryland, College Park, Maryland, 1972), 105 pp.

7. "Autocorrelation Function of Scattered Light for Binary Fluid near the Critical Mixing Point", C. Bendjaballah, Opt. Comm. 9, 279-282 (1973).

8. "The Three-Particle Collision Term in the Generalized Boltzmann Equation," J. V. Sengers, Acta Physica Austriaca, Suppl. X, 177-208 (1973).

9. "Double Scattering Correction in the Interpretation of Rayleigh Scattering Data near the Critical Point", A. J. Bray and R. F. Chang, Phys. Rev. A12, 2594-2605 (1975).

10. "Progress Report on the Measurement of the Critical Exponent η of a Binary Liquid Mixture Near Its Critical Mixing Point", R. F. Chang and H. Burstyn, Technical Report No. 76-040 (Department of Physics and Astronomy, University of Maryland, College Park, Maryland, 1975), 21 pp.

11. "Experimental Determination of the Critical Correlation Function for a Binary Liquid Mixture: Evidence for Universality", R. F. Chang, H. Burstyn, J. V. Sengers, and A. J. Bray, Technical Report No. 77-[illegible] (Department of Physics and Astronomy, University of Maryland, College Park, Maryland, 1976), submitted to Phys. Rev. Letters.

APPENDIX

Experimental Determination of the Critical Correlation Function for a Binary Liquid Mixture: Evidence for Universality

Experimental Determination of the Critical Correlation Function for a Binary Liquid Mixture: Evidence for Universality*

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Abstract

Using light scattering techniques we have measured the correlation function of a binary liquid near the critical point with a precision of 0.2%. The data cover a range of $0.18 < k\xi < 26$ in terms of the scaling variable $k\xi$ and yield the critical exponent values $\gamma = 1.240 \pm 0.007$, $\nu = 0.625 \pm 0.003$ and $\eta = 0.016 \pm 0.007$. The results indicate that binary liquids belong to the same universality class as the Ising model.

Near a second order phase transition the intensity of scattered radiation for scattering vector $\underset{\sim}{k}$ is proportional to the Fourier transform $\chi(k)$ of the order parameter correlation function. Sufficiently close to the critical temperature T_c one expects the scaling form

$$\chi(k) = \Gamma\, t^{-\gamma} g(k\xi) \qquad (1)$$

where $t = (T-T_c)/T_c$, γ the susceptibility exponent, ξ the correlation length and $g(x)$ the correlation scaling function[1].

According to the universality hypothesis, systems having the same basic symmetries are expected to have identical critical exponents and scaling functions, and are said to belong to the same universality class.[2] It is widely assumed that universality classes for homogeneous, isotropic systems may be assigned according to the spatial dimensionality d and the number of components n of the order parameter. This assumption implies that fluids near the gas-liquid critical point and binary liquids near the consolute point should belong to the same universality class ($d = 3$, $n = 1$) as the three-dimensional Ising model. The critical exponents for this universality class have been calculated using high-temperature series expansion techniques[3,4] and renormalization group techniques[5,6]. The various calculations yield numerically similar results although some small unresolved discrepancies do exist.

During the past years the values reported for the critical exponents of gases[7] as well as binary liquids[8] appeared to deviate from those attributed to the Ising model. However, recent experiments of Hocken and Moldover on Xe, SF_6 and CO_2 extremely close to the gas-liquid critical point indicated that the exponents do approach Ising like values[9]. In addition, recent co-existence curve data obtained by Greer for the isobutyric acid and water

mixture showed good agreement between the experimental and the Ising value for the exponent β [10].

In order to further investigate the hypothesis of universality, with particular emphasis on that for the critical correlation function, we have obtained a series of accurate light scattering measurements for the binary liquid 3-methylpentane-nitroethane. The measurements enable us to determine the critical exponents and the correlation scaling function. The values obtained for the critical exponents are very close to those obtained from the Ginzburg-Landau-Wilson formulation of the Ising model by Baker *et al.*[6] indicating that binary liquid mixtures do indeed belong to the same universality class as the Ising model.

Using photon counting techniques the scattered intensity at a scattering angle of 90° was measured as a function of temperature. Data points taken in the range $1 \times 10^{-6} < t < 3 \times 10^{-3}$, corresponding to $0.18 < k\xi < 26$, were used in the analysis. In order to obtain accurate absolute scattering data each scattering intensity reading was normalized with respect to the incident intensity measured with the same photodetector. The r.m.s. uncertainty of each data point is roughly 0.2%, determined from twenty consecutive readings collected at 100-second counting intervals at a fixed temperature. The temperature was stabilized to within 0.2 millidegrees. The concentration of the mixture was within 0.4% of its critical value. Experimental details will be presented elsewhere.

When the precision of the data is a fraction of one percent, many small effects, previously ignored, must be taken into account. Therefore, the data are corrected for the following effects: (1) turbidity, (2) double scattering[11], (3) temperature dependence of the intensity prefactor $T\rho^{-1}(\partial\epsilon/\partial c)^2$ where ρ is the density, ϵ the dielectric constant and c the concentration

(order parameter), and (4) entropy fluctuations, Brillouin scattering[12] and cell wall scattering. These effects were either measured or estimated from known physical properties of the mixture. The magnitudes of the turbidity and double scattering corrections are, respectively, about 4% and 1% at $k\xi = 10$, 2% and 0.4% at $k\xi = 3$, and become progressively smaller as $k\xi$ decreases. The temperature dependence of the prefactor causes it to deviate from its value at T_c by 0.1% at $k\xi = 1$ and 2% at $k\xi = 0.18$. The extraneous contributions from entropy fluctuations and Brillouin scattering are estimated to be roughly two-thirds of the scattering intensity at $T-T_c = 10$ K and are nearly constant in the temperature range of the experiment.

Past determinations of the exponent η from scattering experiments have been difficult, partly due to the lack of a scaling function $g(x)$ of sufficient accuracy[13]. This deficiency may be remedied by using a "truncated Fisher-Langer" approximant recently proposed by one of the authors[14]. This reproduces to high accuracy the theoretically known Ising model correlation functions in 2 and $(4-\epsilon)$ dimensions. To facilitate the analysis we use a form linearized in η. A numerical check confirms that the linearization is valid for the small values of η considered here. The reciprocal of the scaling function is given by:

$$g^{-1}(x) = 1 + x^2\left(1 + \frac{x^2}{9}\right)^{-\eta/2}\left\{1 + \eta\left[s_2(x) - \frac{w}{2}\ln\left(\frac{1 + x^2/9}{1 + x^2/36}\right)\right]\right\} \qquad (2)$$

where $s_2(x)$ is given by

$$s_2(x) = \frac{x^2}{9}\int_1^\infty \frac{du}{u(u^2 + x^2/9)}\,[1 - F(u)] \qquad (3)$$

Here $F(u)$ is the spectral function [14, 15], truncated so as to vanish for $u < 2$, which is derived from the Fisher-Langer scaling function[16]

$$g_{FL}(x) = (C_1/x^{2-\eta})(1 + C_2/x^{(1-\alpha)/\nu} + C_3/x^{1/\nu}), \text{ for } x >> 1,$$

through the relation $\mathrm{Im}\, g_{FL}^{-1}(i|x|) = [\sin(\pi\eta/2)/C_1]\ |x|^{2-\eta}\ F(|x|/3)$. The spectral function may be "fine-tuned" by adding a constant contribution of strength $w \geq 0$ in the range $1<u<2$. This contribution leads to the final term in the square brackets in Eq. (2).

The data analysis is carried out by means of a non-linear least squares fitting procedure using five free parameters: an overall proportionality constant I_o, the critical exponents ν and η, the correlation length amplitude ξ_o, and an extraneous intensity contribution ΔI. The best fit values for ΔI turn out to be very close to the estimates discussed above. Preliminary analysis indicated that the best fit values of η and ν are close to 0.015 and 0.625 respectively. Subsequently for the final analysis we used an $s_2(x)$ evaluated with $\nu = 5/8$ and $\alpha = 1/8$, the latter derived from the hyperscaling relation $2 - \alpha = 3\nu$, and the values $\eta = 1/54 = 0.0185$, $C_2 = 1.773$ and $C_3 = -2.745$ derived from the ε-expansion to $O(\varepsilon^2)$ [17]. The use of these values leads to a spectral function of the expected shape[14]. Since the best fit values of the critical exponents were found to be insensitive to the choice of w within a physically reasonable range ($0 \leq w \lesssim F(2) \sim 0.17$), the value $w=0$ was used in the final analysis. The closeness of the best fit values of ν and η to those used in calculating $s_2(x)$ supports a posteriori the self-consistency of the analysis.

Eight experimental runs were available for this analysis, the results of which are presented in Table I. The values for γ and η_4, which are the basic exponents calculated by Baker et al.[6] were deduced from ν and η using the scaling law $\gamma = \nu(2 - \eta)$ and the definition $\eta_4 = \eta - 2 + 1/\nu$. The first six runs were obtained with the scattering beam located at the level where the

meniscus would appear. In the last two runs this position was varied by 2 and 5 mm from the earlier position. The results appear to be independent of the level of the beam confirming the absence of gravity effects. The average values and r.m.s. errors of the critical exponents from the eight runs are

$$\nu = 0.625 \pm 0.003 \qquad \gamma = 1.240 \pm 0.007 \qquad (4)$$
$$\eta = 0.016 \pm 0.007 \qquad \eta_4 = -0.384 \pm 0.010$$

When an extended scaling correction factor $(1 + Ct^{\Delta_1})$ with $\Delta_1 = 0.5$ [6,18] was included in the analysis, we obtained $C = 0.3 \pm 0.3$ with insignificant shifts in the exponent values indicating that the data are within the range of asymptotic simple scaling.

Our results are rather similar to the exponent values calculated by Baker et al. from the Ginzburg-Landau-Wilson model using the Callan-Symanzik equation[6]:

$$\nu = 0.627 \pm 0.01 \qquad \gamma = 1.2410 \pm 0.002 \qquad (5)$$
$$\eta = 0.021 \pm 0.02 \qquad \eta_4 = -0.3843 \pm 0.003$$

The data were also compared with the predictions of high temperature series expansions [3,4] by fixing ν and η at 0.638 and 0.041 and using an appropriate scaling function [14]. For comparison, two deviation plots of a representative experimental run (run no. 3) are presented in Fig. 1. Whereas the upper plot with $\nu = 0.625$ and $\eta = 0.017$ does not show any significant deviations, small but noticeable systematic deviations do appear in the lower plot with $\nu = 0.638$ and $\eta = 0.041$. Our data thus seems to support the Ising exponents as computed by Baker et al.[6].

Our exponent γ is also in good agreement with the value of this exponent found by Hocken and Moldover for gases asymptotically near the critical point[9]. However, unlike gases where the asymptotic behavior is not approached

unless $t = 10^{-5}$, we find that for binary liquids the range of asymptotic behavior extends to $t = 10^{-3}$ in agreement with Greer's analysis of the coexistence curve of a binary liquid[10].

In conclusion we note that our analysis determines both the exponents and the correlation scaling function. The close agreement between our results and those of Baker et al. indicates that binary liquids and the Ising model do belong to the same universality class. Moreover, unlike gases this universal behavior is observable in a temperature range that is readily accessible experimentally.

The authors are indebted to R. A. Ferrell, R. W. Gammon, M. S. Green, S. C. Greer, P. C. Hohenberg and C. M. Knobler for stimulating discussions and valuable remarks, to C. O. Alley for his encouraging support and to H. J. M. Boots of the University of Leiden for his assistance in the analysis of the data. The collaboration with Dr. Boots was supported by NATO research grant 1065.

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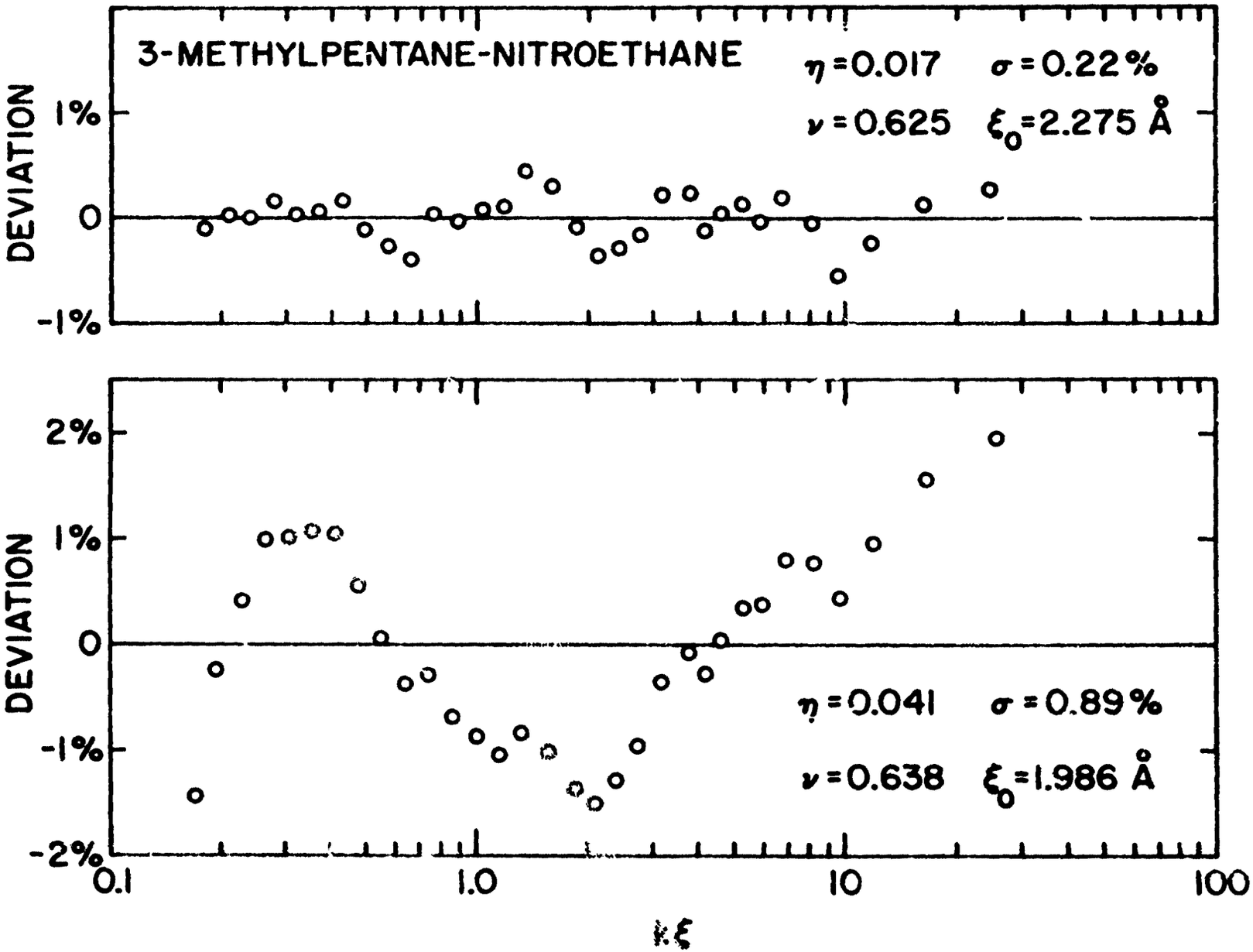
TABLE I. EXPERIMENTAL CORRELATION FUNCTION PARAMETERS

Run	ν	η	ξ_0(Å)	γ	η_4	r.m.s. error of fit
1	0.6218	0.0288	2.324	1.2257	-0.3630	0.24%
2	0.6295	0.0200	2.196	1.2463	-0.3913	0.23%
3	0.6252	0.0169	2.275	1.2398	-0.3835	0.22%
4	0.6227	0.0175	2.315	1.2344	-0.3765	0.12%
5	0.6250	0.0058	2.319	1.2460	-0.3937	0.20%
6	0.6278	0.0122	2.274	1.2479	-0.3949	0.15%
7	0.6250	0.0108	2.313	1.2432	-0.3891	0.16%
8	0.6237	0.0158	2.317	1.2376	-0.3809	0.18%

FIGURE CAPTION

Fig. 1 Percentage deviations between the measured scattering intensity and the theoretical prediction using two different choices for ν and η.


3-METHYLPENTANE-NITROETHANE
η = 0.017 σ = 0.22%
ν = 0.625 ξ0 = 2.275 Å
DEVIATION
1%
0
-1%
2%
1%
0
-1%
-2%
η = 0.041 σ = 0.89%
ν = 0.638 ξ0 = 1.986 Å
0.1
1.0
10
100
k ξ