Progress Report

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CRITICAL SCATTERING OF LASER LIGHT
BY THIN FLUID FILMS

AEC Contract AT(11-1)-2203

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Contract Period: February 1, 1972 - January 31, 1973

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Three areas of research have been initiated during this contract period:

Area (1)

The development of internal reflection spectroscopic techniques for the purpose of measuring the critical parameters and the effects of wall forces of thin fluid films at the surface of an internal reflection element.

Area (2)

The development of a closely spaced Fabry-Perot interferometer with flats separated by a thin fluid film for the purpose of studying the critical behavior of the fluid and the effects of wall forces and confinement.

Area (3)

The development of techniques to study critical phenomena in fluids through the observation of scattered laser light from suspended Brownian particles.

(This latter area of research was not explicitly described in the original contract proposal and as a consequence no contract funds were expended in this area other than to make certain modifications in the correlation spectrometer-computer and temperature controller. These devices are also to be used in areas (1) and (2) which were explicitly described.)

Accomplishments to date in Area (1): Internal Reflection Spectroscopy

The design and construction of the spectrometer is complete including the vacuum system, high pressure system, scattering cell, internal reflection prism, and the temperature controller. The internal reflection prism has just been received and glued to the scattering cell window. The system is now ready for use. CO_2 has been chosen as the fluid for the initial experiments.

In the construction stage it was necessary to develop a double light beam chopper with low vibration and wind noise. Its purpose is to gate the photon counter for incident and exit beam comparison. A pressure and volume adjustment -- a plunger -- has been included on the scattering cell and operates to our satisfaction in preliminary tests. The system is mounted on a vibration-isolation bench and it is easily adjusted to be level for purposes of reducing

gravitational field effects. The appropriate electronics were constructed for connecting the photon counting system to a printer. Experiments (in Area 3) with the fine temperature controller demonstrate that it is capable of controlling the temperature of a system to $\pm .001^{\circ}\text{C}$ with a resolution of $.0001^{\circ}\text{C}$. The complete temperature control system includes an outer coarse controller and an inner fine controller.

It is anticipated that initial measurements of the turbidity of CO_2 will be made by the end of the contract period. These data will be interpreted in terms of the isothermal compressibility and the correlation length as functions of $(T-T_C)$. Gravitational field effects will be searched for by varying the angle of incidence of the laser beam with respect to the surface of the internal reflection prism. In this way different layer thicknesses of fluid will be probed at the IRE-fluid interface close to the critical point. These measurements will be made, of course, after the proper level adjustments have been completed.

Accomplishments to Date in Area (2): Fabry-Perot Interferometer

The design and construction of the closely spaced Fabry-Perot system is essentially complete except for the Fabry-Perot flats. These flats have just been received and must be sent out for their high reflectance coatings. This system is similar but separate from the system designed for the internal reflection element (IRE) in all respects but differs in the design of the scattering cell. The system is mounted on the same vibration-isolation bench and is arranged to use the same high pressure filling system, electronics and vacuum pump.

The scattering cell consists of shut-off valves, an adjustable pressure-volume control plunger, and two high reflectance optically flat windows. A mechanical adjuster for tuning the interferometer was constructed and tested with a separate pair of flats under high pressure conditions (≈ 1100 psi). After a number of trials and modifications it is now possible to tune the Fabry-Perot mechanically to our satisfaction from outside the vacuum enveloped. The piezoelectric transducers which will work in tandem with the mechanical adjusters have been purchased but they have not yet been installed or tested in the interferometer.

On receipt of the Fabry-Perot flats, the system will be assembled and tested using CO_2 as the critical fluid. In this device both the turbidity and

correlation spectrum will be measured as a function of the flat spacing and the temperature. It is anticipated that some initial results will be obtained by the end of this contract period. This system may also be used to measure the critical opalescence in the binary system nitroethane-isooctane as an aid to the experiments in Area (3). The scattering from this binary mixture is very low at the critical point because the two components have almost the same refractive index. The Fabry-Perot is expected to be more sensitive to the effects of light scattering than the usual type scattering cell.

Accomplishments to Date in Area (3): Light Scattering from Brownian Particles

A third method of studying critical phenomena in fluids is through the analysis of the correlation spectrum of scattered laser light from Brownian particles suspended in the fluid. A correlation spectrometer has been designed and constructed. It has been in almost constant use in the study of the binary mixture. See the enclosed preprint [1]. This particular mixture was chosen because the refractive indices of the two components are very closely matched and as a consequence the scattered light from the Brownian particles is not masked by the critical opalescence in the region of the critical point. After trying many different types of particles it was found that of these only teflon microspheres could be successfully suspended in the fluid without ensuing coagulation.

The anomalous shear viscosity was obtained from the correlation spectra as a function of temperature in the region of the critical point. The results agree rather well with a recent theory by Perl and Ferrell [2].

There have been a number of interesting observations not mentioned in the preprint. As the temperature is lowered through the critical temperature, the scattered light from the suspended particles decreases quite suddenly to almost zero. In the experiment the laser beam was located just above the level at which the interface between the two separate phases appears, i.e., is the isooctane

[1] Light Scattering Investigations of Brownian Motion in a Critical Mixture, by K. B. Lyons, R. C. Mockler, and W. J. O'Sullivan. Submitted for publication to Physical Review Letters.

[2] R. Perl and R. A. Ferrell, Phys. Rev. Lett. 29, 51 (1972).

rich phase. The particles are apparently swept out of the top phase just as the interface forms as a result of lowering the temperature through its critical value. In effect it rains. This result is quite reproducible. When using a second sample of teflon balls obtained from a different commercial source, the teflon balls were observed to rain up when passing down through the critical temperature. The scattered intensity increased drastically and the teflon balls are observed to increase their concentration in the upper isooctane rich layer. This result is also quite reproducible. These results suggest that a kind of nucleation is occurring very rapidly and very close to the critical point. They further suggest that the surfactants used in the two different preparations of teflon balls are different: One of the surfactants gives the teflon balls a greater affinity for nitroethane and the other surfactant gives the balls a greater affinity for isooctane.

This work suggests that laser light scattering from Brownian particles will be useful in the study of critical phenomena in fluids and may also have utility in the study of nucleation processes and wall-fluid interactions.

New mixtures have been made with considerable care taken in their purification. The elimination of extraneous particulate matter was attained through the distillation process, membrane filters and an ultra centrifuge. The elimination of the extraneous particulate matter was quite successful which improves the possibility of observing the correlation spectrum of the critical opalescence of the pure fluid mixture without teflon balls. This would provide a better value of the correlation length as a function of temperature which is needed for the determination of the anomalous viscosity.

The purified mixtures display a shift in the critical temperature of about 0.3°C , from the value obtained in the original experiments. These new mixtures also show that the mixtures are not quite critical; there is an excess of nitroethane.

Viscometer and density measurements have been made for the mixtures from the critical temperature to about 25°C above the critical temperature. This will also allow a better determination of the anomalous viscosity.

The following work is underway and in some cases will be completed by the end of the contract period. The experiments on the anomalous viscosity of

the nitroethane-isooctane mixture will be redone with purified liquids and with a number of different teflon ball sizes. A more exact determination of the critical concentration will be made. A critical comparison will be made between the viscosity measured using the teflon Brownian particles and the viscosity measured using oscillating disc and capillary flow methods. The purpose of this comparison is to study the effect of macroscopic velocity gradients on the measured viscosity in the region of the critical point.

With regard to the "nucleation" phenomena, we should seek to determine the various surfactants that are put into commercial teflon suspension in water which is the form in which these teflon microspheres are purchased. With this information we may be able to make some inferences about the different affinities for the fluid components of our two different samples of teflon balls. It may be of interest to deliberately add certain known surfactants for further studies.

Publications Intended for this Contract Period

1. K. B. Lyons, R. C. Mockler, and W. J. O'Sullivan, "Light Scattering Investigations of Brownian Motion in a Critical Mixture." Submitted for publication to Phys. Rev. Letters.
2. K. B. Lyons, R. C. Mockler, and W. J. O'Sullivan, "Light Scattering from Suspended Particles in a Critical Mixture," to be submitted for publication in the Bulletin of the American Physical Society, New York Meeting, December, 1972.
3. A detailed writeup of the work done on the scattering of laser light from Brownian particles suspended in the critical mixture nitroethane-isooctane will be submitted to Physical Review for publication.

Summary of Effort

The Co-Principal Investigators devoted 100 percent time to the project during the period June 1, 1972 to August 31, 1972. They devoted 15 percent time to the project during the period February 1, 1972 to May 31, 1972, and will continue to devote this percentage of time to the project through January 31, 1973.

Other Current Support

None.

Financial Statement

University of Colorado

Contract No. AT(11-1)-2203

For the Period February 1, 1972 to January 31, 1973

1.	Estimated Total Project Costs for the Current Period	\$38,675
2.	Amount Chargeable to AEC for Current Period	38,675
3.	Cumulative Support Costs	-0-
4.	Estimated Total Cumulative Support Costs	\$38,675
5.	Cumulative Support Ceiling	\$38,675
6.	Anticipated Difference Between Estimated Total Cumulative Support Costs and Cumulative Support Ceiling	\$ -0-

Light Scattering Investigation of Brownian
Motion in a Critical Mixture*

by

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Abstract

The diffusion coefficient of Brownian particles in the binary mixture nitroethane-isooctane close to the critical phase transition has been measured using autocorrelation spectroscopy. The results support a logarithmic divergence in the shear viscosity. In the region $0.01 < T - T_c < 0.5^\circ\text{C}$, the data fit the curve $\Delta\eta/\eta_0 = (0.057 \pm 0.003) \ln Q_D \xi$, in agreement with a recent theory, where ξ is the correlation length, η_0 is the background viscosity, and Q_D is a fitting parameter.

To date, the only experimental investigation of Brownian motion in a binary liquid near the critical point has been performed by Bal'tsevich, Martynets, and Matizen^{1,2} (BMM) who applied microphotography to follow individual particles. In this letter we present the first results of a study of Brownian motion near the critical point of a binary mixture using a photon autocorrelator. These data represent a significant statistical improvement over the earlier results since the light scattering process effectively averages over a large particle population.

It has been shown theoretically³⁻⁶ and experimentally⁷⁻⁹ that the viscosity of a binary mixture exhibits an anomaly at the critical mixing point. The theories predict a power-law divergence^{4,6} or at most a logarithmic divergence^{3,5} depending on whether the treatment is classical or based on scaling laws. The phenomenon has been investigated experimentally by the oscillating disc⁸ and capillary flow⁷ methods, as well as by analysis of anomalous Rayleigh linewidth in scattering from critical fluctuations^{3,10}. Except for the latter, which measures a q -dependent viscosity and from which precise data are difficult to obtain³, these methods have the disadvantage of applying a macroscopic disturbance to the system in the process of measurement, which could change the results.

Indeed, it has long been known experimentally¹¹ and has recently been shown theoretically^{4,12,13} that a macroscopic velocity gradient can significantly affect the usual viscosity measurements in the critical region, even as far as 0.2°C from T_c . In the view of the present authors, it thus seems highly doubtful whether capillary measurements can be accurate at $T - T_c \lesssim 0.1^{\circ}\text{C}$ or can even be considered to yield order of magnitude measurements closer than a few hundredths of a degree from the critical point¹⁴. The oscillating disc technique usually

operates with a much lower gradient, so the results of Kuskova and Matizen⁸ might be valid within 0.01°K ¹⁵. This will be considered in more detail in a forthcoming publication.

The Brownian motion of particles in a critical mixture should reflect the anomalous viscosity^{1,2} without involving a macroscopic disturbance during measurement. The effective viscosity, η , is obtained from the diffusion coefficient, D , by the equation due to Einstein:

$$D = k_B T / 3\pi d \eta, \quad (1)$$

where d is the diameter of the particles, k_B is Boltzmann's constant, and T is the temperature. In the microphotography studies the observed anomaly was much larger than that to be expected from the results of other viscosity measurements on the same systems. The difference was assumed to be caused by the long correlation length, ξ , which invalidates the derivation of Eq. (1) for $\xi \geq d$. However, in the systems in question this inequality is true only, for $T - T_c \leq 0.01^\circ\text{C}$. Since the "anomalous mobility" found by BMM seemed to be observable^{1,2} as far as 0.1°C from the critical point, their explanation might not be entirely correct; rather, part of the discrepancy may be due to the effect of velocity gradients on the viscosity data with which they compared their results.

The use of autocorrelation spectroscopy to investigate Brownian motion has been discussed by Clark, Lunacek and Benedek¹⁶. If the particles are monodisperse, the correlation function is given by

$$g(\tau) = A e^{-2\tau/\tau_c} + B \quad (2)$$

where τ_c is a function of particle diameter, d , and

$$\tau_c(d) = (DK^2)^{-1} = 3\pi d \eta / k_B T K^2, \quad (3)$$

where K is the scattering momentum transfer, and A and B are constants determined by factors such as count rate and scattering geometry. If one has a distribution of particle sizes specified by a distribution function, $f(d)$, then the spectrum takes the form

$$g(\tau) = \int_0^\infty dx f(x) \left[A e^{-2\tau/\tau_c(x)} + B \right] / \int_0^\infty dx f(x) \quad (4)$$

In this work we assumed a Gaussian distribution of inverse particle size (i.e. a Gaussian distribution of τ_c) of the form

$$f(d) = D_0 \exp \left[- (d^{-1} - d_0^{-1})^2 w^2 \right] \quad (5)$$

and performed a least squares analysis with w and d_0 as adjustable parameters. D_0 is a normalization constant. Using this technique we were able to fit the spectrum typically to within 0.2% out to $\tau \sim 5\tau_c(d_0)$, where $\tau_c(d_0)$ is given by Eq. (2).

The system chosen for this investigation was nitroethane-isooctane (2,2,4-trimethylpentane)¹⁷⁻¹⁹ which opalesces only weakly at the transition, due to the near perfect match of the refractive indices of the components. This choice was made to prevent the critical opalescence from masking the Brownian scattering. The critical parameters of this system were located by observation of the meniscus position as a function of temperature and concentration in a series of samples. We determined the critical concentration to be $C_c = 47.8 \pm 0.4$ wt% nitroethane, and the critical temperature to be $T_c = 30.8 \pm 0.4^\circ\text{C}$. The location of these parameters by visual observations is difficult since the meniscus is very hard to see, hence the rather large uncertainties. The values of T_c and C_c agree with previous investigations¹⁷⁻¹⁹. The particles used were Teflon microspheres manufactured by Dupont Co., and fractionated centrifugally in our laboratory. The Stokes diameter of the particles was measured

by introducing them into pure nitroethane and performing the autocorrelation spectroscopy. Using available data²⁰ on the viscosity of nitroethane we found the mean particle size, from Eq. (5), to be $d_0 = 0.31 \mu\text{m}$, and the fractional width determined from the least squares (w/d_0) to be 17%. The measurements in pure nitroethane and in the mixture were performed at the same scattering angle, which was $59.7^\circ \pm 1^\circ$.

The $0.31 \mu\text{m}$ particles had a slow settling rate, but it was significant enough to require that the sample be agitated approximately every two days to keep the particle distribution at the scattering volume from changing. It was then necessary to wait at least seven hours to allow larger, extraneous dust to settle out. We waited more than ten hours before beginning any measurements, and they were then very reproducible for a period of two days, with no evidence of significant coagulation during this period. The sample was agitated in this manner eight times during taking of the data shown in Fig. 1.

The temperatures were measured differentially by an uncalibrated Platinum sensor and an AC Kelvin double bridge. Resolution was $\pm 0.0001^\circ\text{C}$ and stability at each point was held to $\pm 0.003^\circ\text{C}$. The absolute temperature was known only to 0.3°K . The temperature gradient along the entire sample, of length 2 cm, including laser heating, is estimated to be less than 0.005°C .

The results are displayed in Fig. 1, graphed as absolute viscosity, with relative error bars as determined by standard techniques from the least squares procedure. As the transition is approached from above, the observed viscosity shows a logarithmic divergence (see Fig. 2), which seems to round off $T - T_c \leq 0.01^\circ\text{C}$, but remains valid for $0.01 < T - T_c < 7^\circ\text{C}$. Below the transition it falls very sharply, probably due to the rapidly changing concentration of the phases. (The scattering volume is in the isooctane-rich phase.) In the region $T - T_c \leq 0.01^\circ\text{C}$ it is hard to say whether the rounding effect

is real, indicating a non-divergent viscosity, or possibly caused by impurities, by temperature gradients, or by an error in the critical concentration. The latter is a distinct possibility since the critical concentration has been reported only to one figure accuracy¹⁷⁻¹⁹, and there is considerable room for error in our determination.

As mentioned, BMM observed a drastic reduction in the mobility of their particles near the critical point^{1,2}. In their study of the phenol-water system², where the Brownian particles were "incidental dust particles," they observed the diffusion constant to decrease by a factor of 4. In later work¹ on the methanol-cyclohexane system they used fractionated glass-dust particles with $d \approx 0.23 \mu\text{m}$ and found that the diffusion constant dropped by a factor of 2 at the transition. In both cases the observed change in "effective viscosity" was several times that found by more standard viscometric techniques, with the discrepancy still evident as far as 0.1°C from the transition. In marked contrast to these results, our data shows a decrease in mobility of only 26% at the transition.

The results in the two systems investigated by BMM led Gitterman et al²¹ to postulate a theory of "trapping" and "entrainment" of the particles by concentration fluctuations. In both of these systems, as well as the system treated in this work, it is possible to assume that the particles might interact more favorably with one component than with the other, resulting in an energy "well" at appropriate concentration fluctuations. The Teflon particles could interact more favorably with the polar nitroethane molecules; glass particles might interact more with methanol than with cyclohexane; and "incidental dust" particles might do anything. However, the large "anomalous mobility" observed by BMM^{1,2} was significant for $T - T_c$ as large as 0.1°C , where $\xi \sim 0.1d$, in which case it

seems unreasonable that any actual entrapment could be occurring. Indeed, the result of the theory gives $\eta \sim (T-T_c)^{-1.33}$ while their data gave $\eta \sim (T-T_c)^{-0.2}$. The explanation advanced by BMM, on the other hand, supposed the existence of a correlated layer of fluid at the surface of the ball, of thickness approximately ξ . They calculated the extra diameter needed to explain their results, and find it compares well with $\xi(T)$. Therefore, this explanation seems more plausible.

It should be the case, however, that such a "correlated layer" could be important only if the composition were substantially altered near the surface of the particle. This implies a strong interaction with the fluid components, and therefore should be specific to the system studied. It is significant in this regard that the more polar system² (phenol-water) showed a much larger effect, even though the particles used in that investigation were several times larger than in methanol-cyclohexane. We see, then, that our data need not be viewed as a contradiction of the results of BMM. Rather, it would seem that in our system the effects of a correlated layer, if present, are small, since we observed only a 35% increase in the viscosity, similar to that obtained for the methanol-cyclohexane system in the oscillating disc measurements of Kuskova and Matizen.⁸ Further evidence for this viewpoint is given by the logarithmic dependence of our data, as seen in Fig. 2, in agreement with a recent theory of the anomalous shear viscosity due to Perl and Ferrell³. That theory predicts

$$\Delta\eta/\eta_0 = A \ln Q_D \xi \quad (6)$$

where ξ is the correlation length, Q_D is a Debye cutoff parameter, and $A = 0.054$. If we assume that $\xi \sim (T-T_c)^{-5/8}$, the results shown in Fig. 2, for the data in the range $0.01 < T-T_c < 7^\circ\text{C}$ give $A=0.093 \pm 0.004$. On the other hand, if we fit the data for $0.01 < T-T_c < 0.5^\circ\text{C}$ ²², we obtain

$A = 0.057 \pm 0.003$, in good agreement with the theory³. The agreement with the theory for the shear viscosity in the critical region definitely lends credence to the view that the anomaly we have observed includes little if any effect of a surface layer. The data of BMM showed a definite power law dependence, so, since we observed a logarithmic dependence, it seems reasonable that the mechanisms could be different.

We plan to continue this investigation to obtain data on the dependence of the effect on the size of the particles. If the correlation length is causing significant errors in the measured viscosity, those errors should be greatly magnified for smaller particles. This should allow a more definite conclusion as to the problems outlined above. We also plan investigation of the effect of small changes in concentration, with a view to defining the critical parameters more precisely. The role of impurities is unknown to this point and will bear investigation. Data at higher temperatures is needed to improve the extrapolation of the background, since an extrapolation from $T - T_c > 7^\circ\text{C}$ is not considered entirely satisfactory⁹.

With these reservations we conclude that our data supports a logarithmic divergence in the shear viscosity and that the "anomalous mobility" observed by earlier workers was not important in our work. The technique shows promise of further elucidating critical phenomena.

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*Research supported in part by the U. S. Atomic Energy Commission under Contract No. AT(11-1)-2203.

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FIGURE CAPTIONS

Fig. 1

Plot of the viscosity, obtained from Eq. (1) as a function of temperature. The viscosity is in centipoise and the temperature is in degrees deviation from the critical temperature observed by the peak in the viscosity. The solid line is a logarithmic fit to the data, $\Delta\eta/\eta_0 = -.134 \log_{10} .083(T-T_c)$. The dotted line is an extrapolation of the background viscosity from temperatures greater than $T-T_c = 7^\circ\text{C}$, by an Arrhenius equation, $\log_{10} \eta_0 = -1.175 + 286/T$.

Fig. 2

Plots of the fractional anomalous viscosity $(\eta-\eta_0)/\eta_0$ (top) and its log (bottom) versus $\log (T-T_c)$, where T_c is that used in Fig. 1. The semilog fit and the background viscosity are as in Fig. 1, and the log-log fit is $\Delta\eta/\eta_0 = (T-T_c)^{-.28/6.86}$ (dashed line). Both fits are performed for $T-T_c < 7^\circ\text{C}$.