LBL-26231
Preprint

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

RECEIVED
LAWRENCE
BERKELEY LABORATORY

JAN 25 1989

LIBRARY AND
DOCUMENTS SECTION

Submitted to Physics Letters B

Nuclear Spinodal Decomposition

J.A. López and G. Lübeck

November 1988

TWO-WEEK LOAN COPY
This is a Library Circulating Copy
which may be borrowed for two weeks.

NUCLEAR SCIENCE DIVISION

Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098.

LBL-26231 c.2

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

LBL-26231

Nuclear Spinodal Decomposition*

Jorge A. López

Nuclear Science Division, Lawrence Berkeley Laboratory
1 Cyclotron Road, Berkeley, California 94720 U.S.A.

and

Georg Lübeck†

The Niels Bohr Institute
Blegdamsvej 17, DK-2100 Copenhagen Ø, Denmark

November 7, 1988

Abstract:

We use computer simulations to study nuclear fragmentation as expected to occur in heavy ion reactions. Using integral equation techniques and molecular dynamics calculations we first find a potential with an equation of state resembling that of hot and dense nuclear matter. We then use this potential to study the disassembly of two dimensional classical drops. Along the lines of Cahn's theory of spinodal decomposition we calculate the structure factor of the system and extract information about the development of density fluctuations during the breakup. We find isothermal spinodal decomposition to play the dominant role in the breakup. Nucleation of bubbles in the two phase region and adiabatic spinodal decomposition were found not to contribute to the fragment production. Strong density fluctuations were detected in disassemblies crossing the critical point.

*This work was supported in part by the Director, Office of Energy Research, Office of High Energy and Nuclear Physics, Nuclear Physics Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

†Present address: Fred Hutchinson Cancer Research Center, 1124 Columbia St. Seattle, WA 98104

1 Introduction

Heavy ion collisions, with projectile energies from the tens to the hundreds of MeV/A, are expected to yield information about the thermodynamical properties of hot and dense nuclear matter and, in particular, about a possible liquid-gas phase transition. The study of such phase transformations, ordinarily difficult, is further complicated in the nuclear case by the finite size of the system and the short time scales involved in the reaction. Any realistic attempt at understanding the nuclear fragmentation problem must deal simultaneously with many body correlations, finite size and short time effects.

Out of the different techniques currently used to study such reactions [1-5], only molecular dynamics (MD) calculations, which directly solve the classical evolution of an N-body system, can describe a change of phase incorporating the wanted correlations and effects. Besides of taking into account all dissipative and relaxational effects, MD can also go, under the proper circumstances, into any specific limiting behavior such as non-equilibrium dynamics or hydrodynamical flow.

In particular for the study of hot and dense nuclear matter, the large values of the momenta involved in the collisions are expected to reduce the blocking introduced by the Pauli principle and make the mean free path shorter than at lower excitation energies. Under those considerations the quantum effects become less important and a classical treatment is more reasonable. The numerical solution of the classical equations of motion is likely to yield a proper description of the reaction dynamics.

Classical MD calculations with potentials ranging from simple hard spheres to more sophisticated momentum-dependent ones have been used to study different aspects of heavy ion reactions [2-5]. In particular, the nuclear fragmentation process has been simulated by the dissociation of droplets of argon atoms interacting via a Lennard-Jones 6-12 potential by Pandharipande and coworkers [5]. Their findings, which show fragmentation occuring inside a region roughly bounded by the argon adiabatic spinodal, strongly suggest adiabatic spinodal decomposition as the disassembly mechanism, as predicted earlier by Bertsch and Siemens[6].

These results, unfortunately, cannot be directly translated into the nuclear case. The absence of nucleation and other isothermal critical phenomena in the argon case can be tied to the 6-12 potential. Due to the steep rise of the core of this potential, a dense argon droplet will expand very rapidly on thermal diffusion time scales leaving little time for thermal interaction between the two phases. This seems to be in contrast to the nuclear matter case where, according to recent studies of Pethick and Ravenhall [7], the density fluctuations leading to fragmentation are expected to develop isothermally.

With the hope of getting an insight of the physical processes responsible for the nuclear fragmentation, we study in this paper the evolution of density fluctuations during the dissasembly of hot and dense classical droplets. For simplicity we use a two dimensional simulation with a classical potential adapted to simulate nuclear matter. In the next section we introduce such a potential and compare its ther-

modynamical properties with those of nuclear matter using the hypernetted chain and Percus-Yevick equations and MD techniques. We then use this potential in section 3 to follow the expansion of classical two dimensional drops. In section 4 we study the time evolution of the density fluctuations by means of the structure factor in the framework of Cahn's theory of spinodal decomposition. We close this letter with a discussion of our findings and conclusions.

2 The Molecular Dynamics Potential

Ideally, a potential to be used in a classical simulation would have to reproduce all the known static characteristics of nuclear systems as well as the thermodynamical properties of finite nuclear matter. Unfortunately, this just might be too much to ask, and at present no such potential is available. Consequently, one must limit the expectations and try to reproduce those key features most needed for the simulation. Up to now, current potentials under use have been mainly designed to reproduce nuclear binding energies and radii [2-4], with the exception of Ref. [5] where a potential was modeled to give an equation of state similar to that of cold nuclear matter.

As pointed out by Heiselberg, Pethick and Ravenhall[8], the high density stage of the reaction determines the velocity with which matter expands. This velocity, which is dictated by the nuclear compressibility, sets the time scales that will allow thermal interactions to occur and density fluctuations to grow. To have an adequate simulation of the breakup of nuclear droplets we need a potential that reproduces the equation of state of hot and dense nuclear matter correctly.

In this work we choose to use an interparticle potential of the form

$$V(r) = g_r e^{-m_r r}/r - g_a e^{-m_a r}/r,$$

with the coefficients g_r, m_r, g_a, and m_a, adjusted to yield a $T = 20\ MeV$ pressure-density isotherm resembling the one coming from a Skyrme-type interaction as determined by Friedman and Pandharipande[9]. The equation of state of a potential can be determined with the repeated use of MD simulations. This, however, is very demanding computationally and the use of more economical techniques, such as integral equation techniques, is more appropriate.

After making an initial guess for the four parameters, we solve both the hypernetted chain and Percus-Yevick equations[1] to obtain a quick estimate of the critical temperature T_c and density ρ_c corresponding to the selected set of parameters. We then use these values to adjust the energy and length scales to have T_c and ρ_c correspond to the nuclear values. The two remaining dimensionless parameters, say g_r/g_a and m_r/m_a, are then determined by fitting the wanted isotherm with a simple

[1] Integral equations are a useful technique to obtain the pair correlation function of classical fluids $g(r)$. From this function one can then obtain thermodynamical variables such as the pressure or internal energy. For details see *e.g.* Ref. [10].

Newton algorithm. Final verification of the shape of the isotherm is then obtained by performing a series of MD simulations with the selected potential.

Fig. 1 shows a comparison of the $T = 20\ MeV$ $P - \rho$ isotherm of Friedman and Pandharipande with our calculations using the set of values $g_r = 10726\ MeV - fm$, $m_r = 0.3955\ fm^{-1}$, $g_a = 10215\ MeV - fm$, and $m_a = 0.3582\ fm^{-1}$. To obtain this fit, the range of the potential was limited to a cut-off radius of $r_c = 4.17\ fm$ and the potential was shifted by a constant $c = -55.6\ MeV$ for continuity. The saturation density for close packing corresponding to this potential is $\rho_0 = 0.166$ fm^{-2} and at the critical point $\rho_c = 0.065\ fm^{-2}$ and $T_c = 17.5\ MeV$.

To establish the phase diagram corresponding to this potential we have to resort to quenched MD simulations since the integral equations cannot be used in the mixed phase regions. As explained in detail by Abraham[11], one can artificially cool a previously equilibrated droplet of matter to determine the thermodynamical variables of the system at a given temperature and density. After obtaining enough points to draw $P - \rho$ isotherms at $T = 5$, 10 and 15 MeV, we obtain the isothermal phase boundaries by a Maxwell construction. This phase diagram (*cf.* Fig. 2) will be useful in tracking the expansion trajectories in the $T - \rho$ plane in the following section.

3 Simulations

Our main interest is to study the expansion of a drop of liquid and follow the developments of instabilities during the fragmentation of the system. We employ the MD method in two dimensions in much the same way as Pandharipande and coworkers[5] did in three dimensions. After equilibrating an initial area containing 400 particles under periodic boundary conditions at a desired temperature and density, we cut a disk out of it and let the approximately 270 particles inside expand into free space.

We then follow the evolution of the macroscopic properties as in Ref. [5]. The density of the central particles is spatially averaged over the 50% most central particles. In two dimensions we determine the density by

$$\rho \;=\; \frac{3}{2\pi}\,(2\,N_c/3)^3/\,(\sum_c r_i)^2,$$

where N_c stands for the number of central particles and r_i is the distance of the ith particle to the center of mass. The temperature and pressure are determined by the time averages

$$T \;=\; \frac{2}{3}\frac{<E_{kin}>}{N} \quad \text{and} \quad P \;=\; \rho\,T \;-\; \frac{1}{6V}<\sum_{i\neq j} r_{ij}(dV_{ij}/dr_{ij})>,$$

where again, the factors of 2/3 and 1/6 in these expressions are replaced by unity and 1/4, respectively, in two dimensions.

Fig. 2 shows the phase diagram obtained as explained in the previous section. Also displayed are the trajectories followed by expansion runs starting from various initial densities and temperatures. Each run represents an average over 20 events all starting from different microscopic configurations but with equal macroscopic thermodynamical variables. The trajectories in the $T - \rho$ plane belong to one vaporization (I) and three fragmentation processes (II, III and IV). They follow adiabats to a good approximation until the droplet fragments in the region of mechanical instabilities. The production of fragments is accompanied by a generation of surfaces which slows down the rate of expansion of the drop.

4 Spinodal Decomposition

In addition to following T, P and ρ of the central particles, we also studied the density fluctuations arising during the expansion of the drop to understand the mechanism responsible for the disassembly. The advantage of this analysis is that it makes a natural connection with the theory of spinodal decomposition of Cahn[12],[11].

The first step in the study of the kinetics of spinodal decomposition is to consider the behaviour of small density fluctuations via the diffusion equation as first done by Hillert [13],

$$\partial\rho/\partial t \;=\; \nabla\cdot(M\nabla\mu),$$

where M and ρ are the mobility and chemical potential of the medium. In this case the force driving the diffusion comes from a difference between the chemical potential of the disturbance and that of the surrounding medium. The particularization of this equation to the case of small density fluctuations comes through the relation between μ and the Helmholtz free energy.

The free energy density of an inhomogeneous medium can be expressed as a sum of the free energy of a uniform system and the one associated with the generation of incipient surfaces by the inhomogeneities, *i.e.*

$$f(r) \;=\; f_0[\rho(r)] + \frac{1}{2}B(\nabla\rho)^2,$$

where B is the van der Waals constant.[2] Writing μ in terms of the free energy, the diffusion equation can be generalized (after linearizing in ρ) to

$$\partial\rho/\partial t \;=\; M\frac{\partial^2 f}{\partial\rho^2}\nabla^2\rho \;-\; MB\nabla^4\rho.$$

If we now describe an arbitrary fluctuation in terms of its Fourier components, the general solution of this equation for each of these components is an oscillatory

[2] B can be obtained from $B \;=\; -\frac{1}{6}[1 \;+\; \rho\partial/\partial\rho]\;\int d^3r\; r^2V(r)\; g(r;\rho)$, where in two dimensions the factor of $\frac{1}{6}$ is replaced by $\frac{1}{4}$. For symmetric nuclear matter near saturation density $B \sim 80$ MeV fm^5 [14], while for our potential $B \sim 1100$ MeV fm^5 at $\rho = \rho_0$ and $T = 20\ MeV$.

function of the form

$$\delta\rho \;=\; A(\vec{q},t)cos(\vec{q}\cdot\vec{r}),$$

with the amplitude given by

$$A(\vec{q},t) \;=\; A(\vec{q},0)exp\{-Mq^2(\frac{\partial^2 f}{\partial\rho^2} \;+\; Bq^2)t\}.$$

Two kinematical regions are readily seen. If $\partial^2 f/\partial\rho^2 \;+\; Bq^2 \;>\; 0$, the fluctuations are damped away as normally expected, but if $\partial^2 f/\partial\rho^2 \;+\; Bq^2 \;<\; 0$, the inhomogeneities will be amplified and the medium will be unstable with respect to these oscillations. This instability is termed spinodal decomposition. This change of behaviour of the amplitude can be used to indicate the onset of fragmentation.

We follow the amplitude of the fluctuations by means of the structure factor $S(k)$ which is related to the Fourier spectrum of the density fluctuations:

$$S(k) \;=\; \frac{V}{\rho_0} < \rho_k\rho_{-k} >,$$

where ρ_k denotes the Fourier transform of $\delta\rho(r)$. From the pair correlation function $g(r)$ this factor can be obtained by

$$S(k) \;=\; 1 \;+\; \rho \int d^3r \; e^{i\vec{k}\cdot\vec{r}}[g(r)-1].$$

In order to evaluate this function, we sampled the pair distribution function over the central region of the expanding drop and subtract from it the correlation function of a homogenous distribution.

Landau and Lifshitz [15] derive an expression for the mean square fluctuation in thermal equilibrium based on the fact that the probability for a fluctuation, which causes a change of ΔF in free energy, is proportional to $e^{-\Delta F/T}$. In the long wavelength limit the thermal fluctuations in a stable system are given by

$$S_L(k) \;=\; \frac{Tk^2}{m\omega^2(k)},$$

where the frequency of the oscillations is related to the plasma frequency ω_p and speed of sound c_s of the medium via the dispersion relation $\omega(k)^2 = \omega_p^2 + c_s^2k^2$ [7]. To set a scale for comparison, we use this prediction for infinite systems evaluated at the initial T and ρ of each run using $k_1 = 8.65$ and the sound speed of a Fermi gas under those conditions.

Fig. 3 shows the structure factor obtained for the expansion runs II and IV of Fig. 2. These two different initial conditions were chosen with the hope of finding significant effects associated with the critical point. In calculating $S(k)$ we have taken into account only the central region of the expanding drop at intervals of 10 fm/c (100 time steps) apart. Like in the case of the $T-\rho$ trajectories, we have averaged the structure factor over 20 independent configurations for each run.

To illustrate the behaviour of $S(k)$ we followed wavenumbers k equal to the zeroes of the Bessel function J_0 as they set the time and length scales for waves resonating in a spherical drop. Fig. 3 also indicates the times when the temperature inside the expanding drop equals the critical temperature T_c, when the system crosses the coexistence curve (CE) and, somewhat later, the isothermal spinodal line (IS).

Finally, to have an alternative picture of the break-up we study the evolution of the total multiplicity of heavy ($A > 10$) fragments for each of these runs. Defining a cluster as a collection of particles "connected" by a chain of steps smaller than r_c, the cut-off radius of the potential, one can uniquely partition the total system into noninteracting clusters. Fig. 4 shows how this multiplicity varies during the expansion for runs II and IV. The restriction to $A > 10$ helps to ignore the unwanted surface evaporation of light clusters.

5 Discussion

A clear picture of the breakup emerges from the study of the evolution of the density fluctuations. Contrary to the argon simulations of Ref. [5], here the isothermal spinodal plays a major role in the fragmentation. As seen in Fig. 2 the onset of fragment formation can be easily identified with an early stage of isothermal spinodal decomposition.

The role of the adiabatic spinodal cannot be determined from this study as we have not calculated this line for our potential. Nevertheless, we checked the adiabatic sound velocity on selected points around the critical density at different temperatures, and the results place the adiabatic spinodal line at $\rho \sim 0.1\, fm^{-2}$ for $T \sim 10 - 12\, MeV$. Our simulations clearly show the fluctuations beginning to grow significantly long before matter crosses this point.

Other interesting observations come afloat. The expansion velocity, as dictated by the initial conditions, appears to be too fast to allow the nucleation of bubbles to occur in the two-phase region before the mechanical instabilities set in. Similarly, the system seems to be able to stay in the unstable region long enough for all of the examined modes to grow, as Fig. 3 demonstrates. No "violent evaporation" or recondensation as seen in the argon simulations of Ref. [5] was observed. The growth rates of the modes under study give the impression to be exponential to a good degree. Different growth rates are observed for different wavenumbers. No apparent connection between pre- and post-fragmentation density fluctuations was observed. The initial inhomogeneities had amplitudes of the order of magnitude expected for thermal fluctuations in an infinite medium as estimated from $S_L(k)$ for the lowest wavenumber shown at the initial conditions of each run. No significant departure from the infinite medium properties was observed.

For run II we find strong effects asociated with the critical point. A significant amplification of the fluctuations was observed even before the system entered the coexistence region. Strong density fluctuations were observed and all modes seem

to grow equally. The precritical growth of the fluctuations and the presence of a number of noninteracting clusters at early stages of this run (see Fig. 4) indicate the strong "delocalization" of the critical point as discussed by Fisher[16] for the droplet model.

6 Conclusion

Although these results are encouraging, we are far from being able to uniquely connect experimental results with thermodynamical properties of nuclear matter. The fine interplay between the initial conditions of the reaction and the transport properties of the medium make the breakup mechanism especially difficult to track down. In addition to this, actual nuclear reactions are expected to have a spectrum of fluctuations induced by the collision and much different than the one studied here. The collision most likely will select specific modes of breakup.

Our findings strongly underline the need for an accurate description of the critical phenomena, this at present is only possible with MD simulations. More realistic computations need to be performed in three dimensions with potentials yielding more appropriate transport properties (*e.g.* better surface tension parameter B, *etc.*). Only then it will possible to simulate collisions and identify the spatial extent of the dominant fluctuations realistically. This will enable us to go from a qualitative understanding of the problem to a quantitative one.

7 Acknowledgements

This work originated from the workshop on nuclear fragmentation organized in NORDITA by A.D. Jackson and C.J. Pethick. We are indebted to A.D. Jackson, C.J. Pethick, H. Schulz and A. Wirzba for their collaboration in the different stages of this project, and to S. Gavin and J. Randrup for a careful reading of the manuscrpt.

*This work was supported in part by the Director, Office of Energy Research, Office of High Energy and Nuclear Physics, Nuclear Physics Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

References

[1] G.F. Bertsch and S. Das Gupta, Phys. Rep., **160**, 189 (1988), and references therein.

[2] A.R. Bodmer and C.N. Panos, Phys. Rev. **C15**, 1342 (1977); L. Wilets, E.M. Henley, M. Kraft and A.D. Mackellar, Nucl. Phys. **A282**, 341 (1977).

[3] D.H. Boal and J.N. Glosli, Phys. Rev. **C37**, 91 (1988); Ann. Rev. Nucl. Part. Sci. **37**, 1 (1987).

[4] C. Dorso, S. Duarte and J. Randrup, Phys. Lett. **B188**, 287 (1987); J. Physique **48**, C2-143 (1987); C. Dorso and J. Randrup, LBL-preprint 25622, submitted to Physics Letters B.

[5] A. Vicentini, G. Jacucci, V.R. Pandharipande, Phys. Rev. **C31**, 1783 (1985); R.J. Lenk and V.R. Pandharipande, Phys. Rev. **C34**, 177 (1986); R.J. Lenk, T.J. Schlagel and V.R. Pandharipande, Univ. of Illinois at Urbana-Champaign preprint, 1987.

[6] G. Bertsch and P.J. Siemens, Phys. Lett. **126B**, 9 (1983).

[7] C.J. Pethick and D.G. Ravenhall, in *Hadronic Matter in Collision*, edited by P. Carruthers and D. Strottman, (World Scientific, Singapore 1986), p277; Nucl. Phys. **A471**, 19c (1987); Ann. Phys. (N.Y.) **183**, 131 (1988).

[8] H. Heiselberg, C.J. Pethick and D.G. Ravenhall, Phys. Rev. Lett. **61**, 818 (1988).

[9] B. Friedman and V.R. Pandharipande, Nucl. Phys. **A361**, 502 (1981).

[10] L.E. Reichl, *A Modern Course in Statistical Physics*, (Univ. of Texas Press, Austin, 1980); D.A. McQuarrie, *Statistical Mechanics*, (Harper & Row, New York, 1976).

[11] F.F. Abraham, Phys. Rep., **53**, 93 (1979).

[12] J.W. Cahn, Acta Metall. **9**, 795 (1961); J.W. Cahn and J.E. Hilliard, J. Chem. Phys. **28**, 258 (1958).

[13] M. Hillert, Acta Metall. **9**, 525 (1961).

[14] D.G. Ravenhall, C.J. Pethick and J.M. Lattimer, Nucl. Phys. **A407**, 571 (1983).

[15] L.D. Landau and E.M. Lifshitz, *Statistical Physics*, Part 1 (3rd. ed. Pergamon, New York, 1980).

[16] M.E. Fisher, Proc. of the Int. School of Physics, Enrico Fermi, Course 51, *Critical Phenomena*, edited by M.S. Green, (Academic Press, New York, 1971), p1.

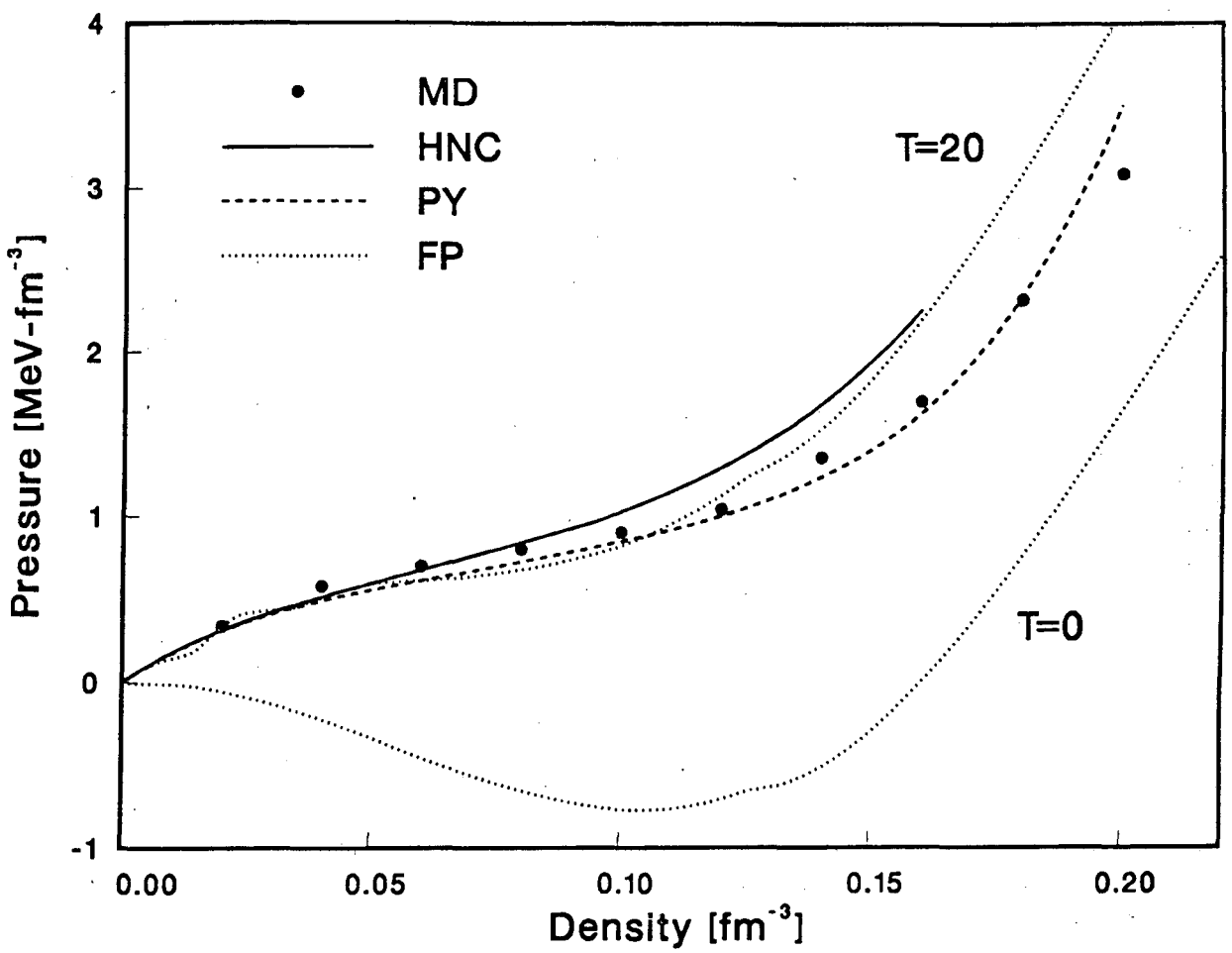

Figure 1

Comparison of the $T = 20\ MeV$ $P - \rho$ isotherms of Friedman and Pandharipande (FP) with those corresponding to the potential of section 2 as obtained from the hypernetted chain (HNC) and Percus-Yevick (PY) equations and MD simulations (MD). Also shown to set the scale is the $T = 0$ isotherm of Friedman and Pandharipande.

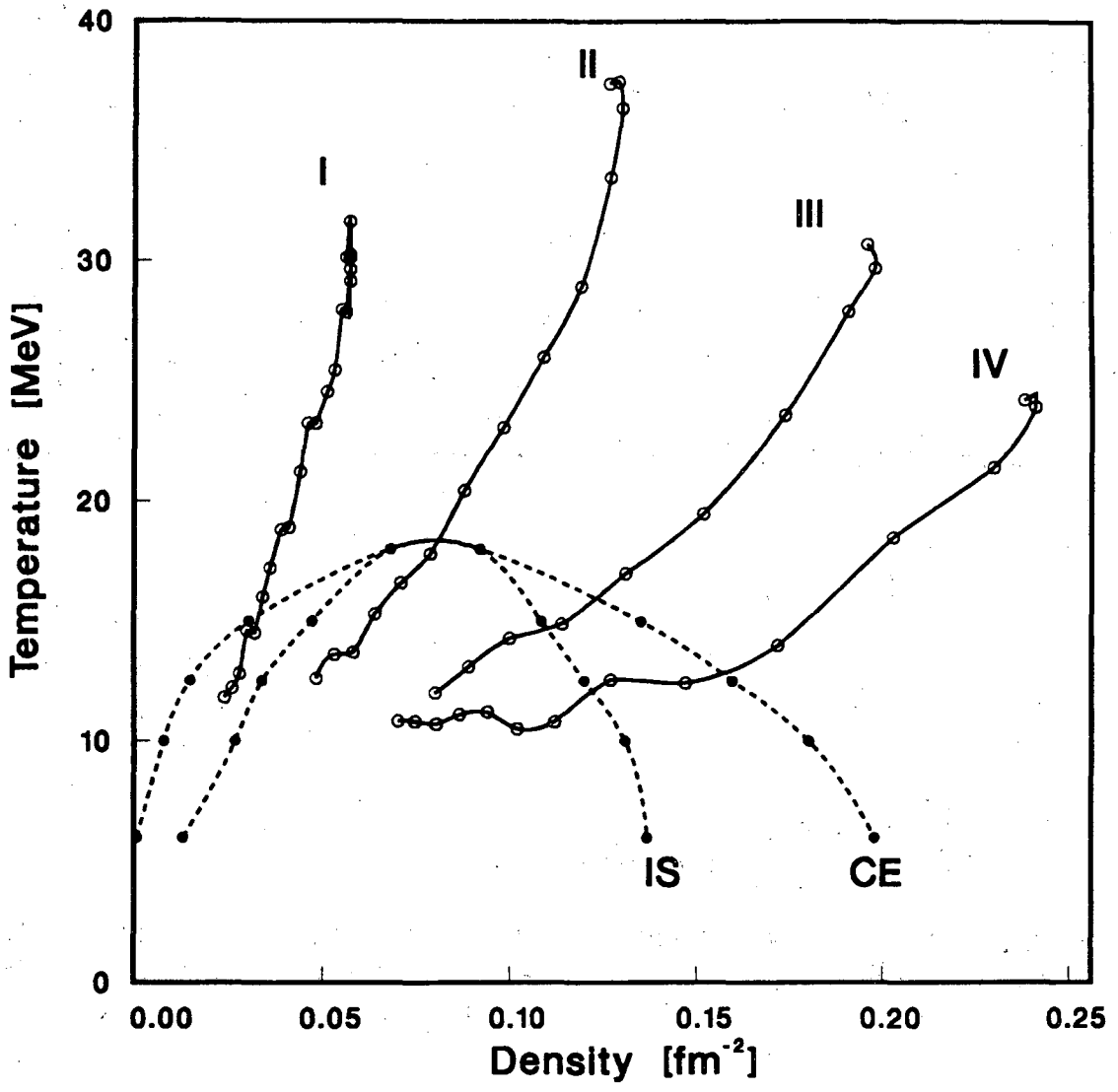

Figure 2

Phase diagram and expansion trajectories in the $T - \rho$ plane for four different macroscopic initial conditions. Points on trajectories are 10 fm/c apart.

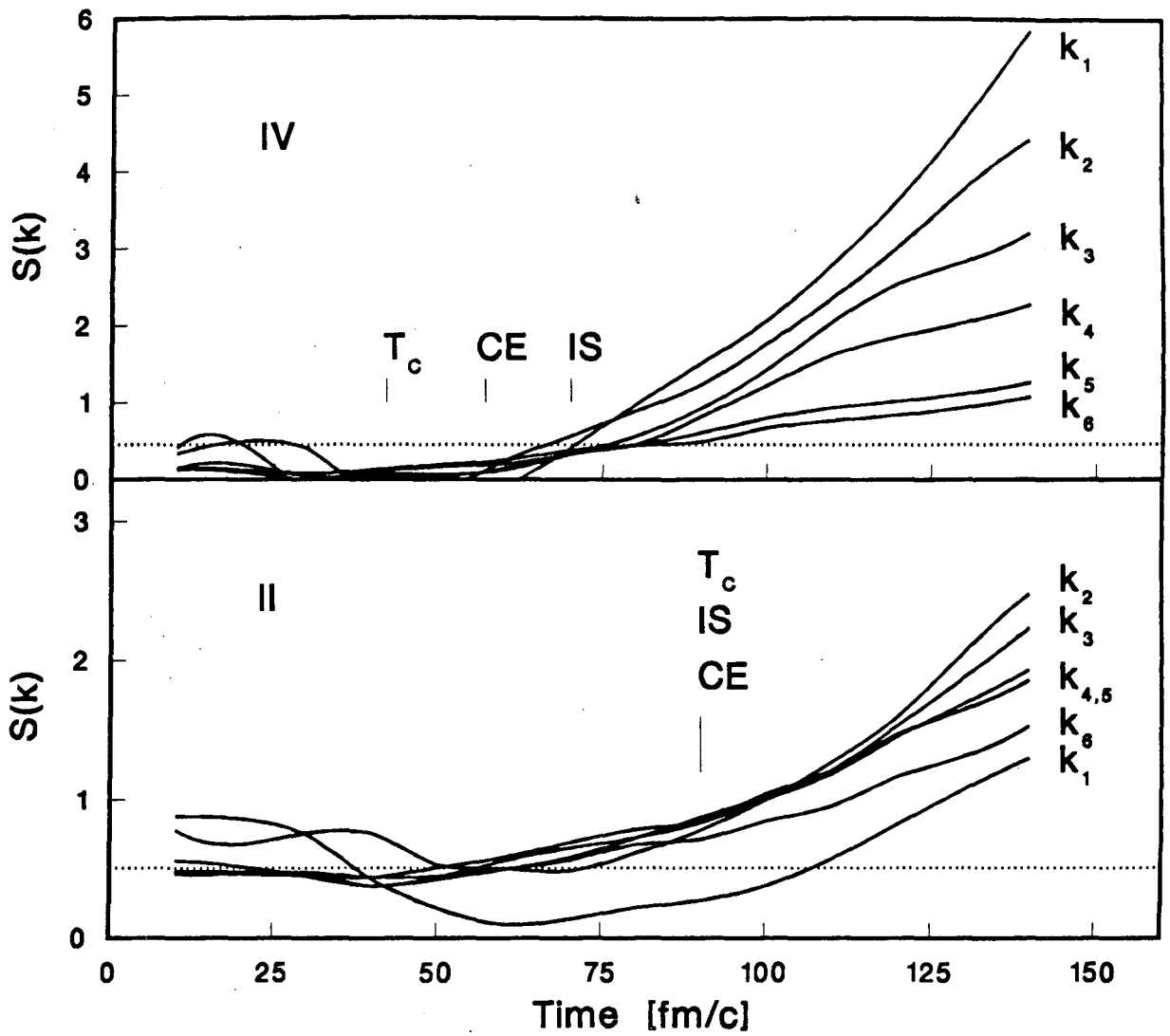

Figure 3

Components of the structure factor $S(k)$ as a function of time for runs IV (top) and II (bottom). T_c, CE and IS point to the times when the system crosses the critical temperature, coexistance curve and isothermal spinodal line, respectively. The wavenumbers k_i correspond to the zeroes of the Bessel function J_0: 8.65, 11.79 $\cdots$. The dotted line represents the background fluctuations expected from $S_L(k)$ for k_1.

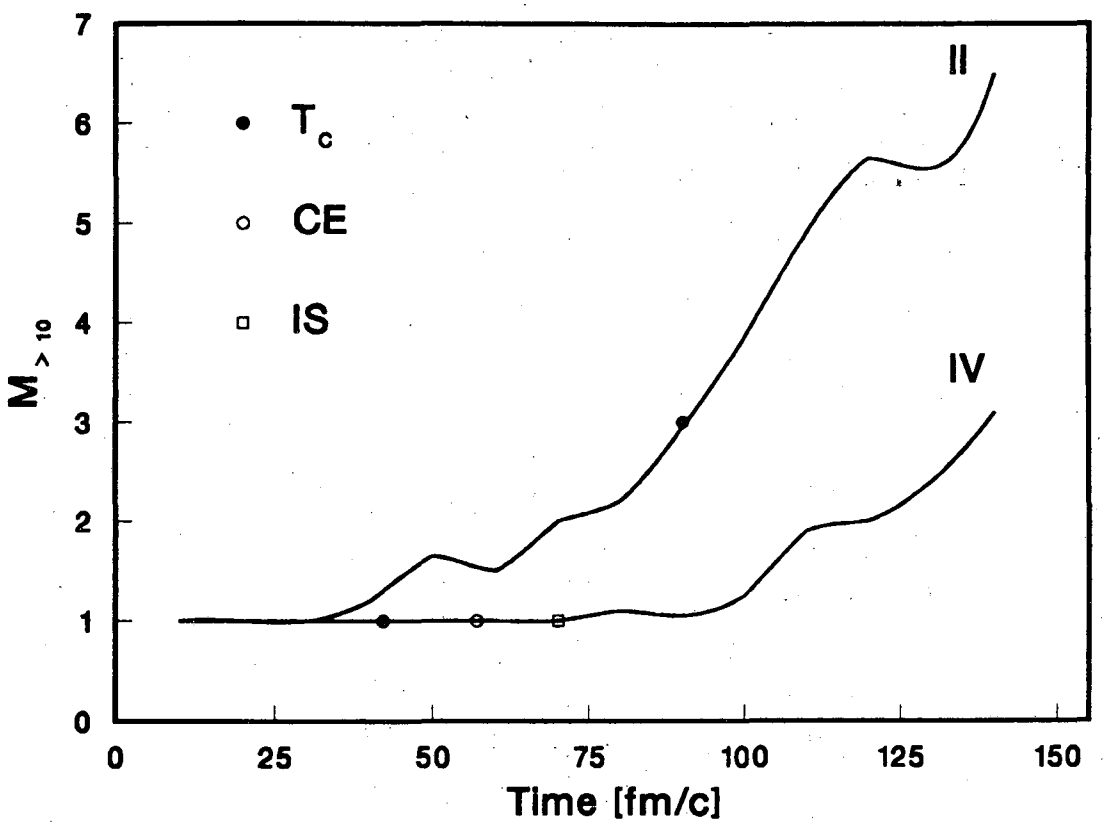

Figure 4

Total multiplicity for clusters with $A > 10$ as a function of time for runs II and IV. T_c, CE and IS are as in Fig. 3.

LAWRENCE BERKELEY LABORATORY
TECHNICAL INFORMATION DEPARTMENT
1 CYCLOTRON ROAD
BERKELEY, CALIFORNIA 94720