Experimental Evidence for a Liquid-Gas Phase Transition in Nuclear Systems

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At certain combinations of temperature and density, nuclear matter may exist as a liquid-gas mixture exhibiting phase instabilities, a characteristic signature of which may be found in the emission of intermediate-mass fragments in nuclear collisions. The present analysis of fragment distributions from proton- and heavy-ion-induced reactions, in the framework of a theory of condensation, is suggestive of the occurrence of such phase transitions with a critical exponent $k \sim 1.7$ and a critical temperature $T_c \sim 12$ MeV.

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Through the study of high-energy nuclear collisions, it may become possible to determine the nuclear equation of state,¹ and to create new forms of nuclear matter. Although most attention has been directed towards phase transitions to a pion condensate or a quark-gluon plasma, conjectured to develop at high density and temperature, it is also possible that a liquid-gas phase instability may set in at a critical temperature and density.^{2,3} This instability may be encountered during the expansion of the initially heated zone, formed in a nuclear collision. In this Letter we discuss experimental evidence for such a critical phenomenon from a study of the formation of complex fragments, in a variety of proton- and heavy-ion-induced reactions. Such studies are of intrinsic interest in yielding information on the equation of state at high temperatures and at densities below normal and also for the insight into the hydrodynamical behavior. necessary for the occurrence of more exotic phase transitions, and for the astrophysical ramifications with regard to neutron stars and supernovae.4

The possibility of a phase transition in an equilibrated nuclear system was previously considered theoretically in a number of papers.⁴⁻⁷ Examination⁶ of light-ion production cross sections over a wide range of incident energies has indicated that, to obtain an unambiguous experimental signature of phase transitions, it is necessary to measure intermediate-mass-fragment cross sections. It has already been suggested in a number of recent papers^{2,8} that the powerlaw dependence, $Y(A) \sim A^{-\tau}$, of the fragment distributions may constitute a signature for the occurrence of phase-transition phenomena near a critical point. The widely differing systems which exhibit this characteristic power-law dependence suggest that phase transitions are global in origin, dependent only on the energy imparted to the system. However, the temperatures of the reported systems vary between about 8 MeV,⁸ and 15 MeV.² Clearly, not both of these systems can be near the critical point.

To understand the temperature dependence of the data, we first fitted the available fragment distributions with a power-law dependence of the form $P(A) \sim A^{-\tau}$, where τ , is the "apparent" exponent. In this approximation, the effects of any temperature-dependent factors are absorbed into the power exponent. Therefore, this apparent exponent will vary with temperature.

For some of the analyzed data the temperature of the system has been extracted from movingsource fits to the fragment energy distributions.^{2,10-12} Where the temperature was not given, we determined it either from the slope of the tail of the 90° fragment energy distributions,⁹ or from the ideal Fermi gas, assuming the size of the emitting system to be in the range of 2 to 3 times the largest fragment mass.⁸ The fragment masses used to determine the power-law exponent were in the range of $3 \le Z \le 22$ for all systems; care was taken to avoid contributions from the tail of possible fission fragments. Table I summarizes the data used and the values obtained by the fit.

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System	Energy (GeV)	A or Z Range	Temperature (MeV)	Angular Range (degrees)	Exponent τ^{a} (<u>+</u> 0.2)	Reference
p + Ag	0.21	3 <u>€</u> Z € 8	6.0 <u>+</u> 1.	20, 90, 160	4.1 (4.7)	9
p + Ag	0.30	3 ≤ Z ≤ 8	6.7 <u>+</u> 1.	20, 90, 160	3.7 (4.3)	9
p + Ag	0.48	12 < A <25	8.2 <u>+</u> 1.	20 - 160	3.2	10
p + Ag	4.9	6 ≤ Z ≤18	14.0 <u>+</u> 1.	90 ^b	2.4	11
p + Kr	80-350	12 < A <30	14.5 <u>+</u> 1.	34 ^b	2.9	2
p + Xe	80-350	12 < A <30	15.0 <u>+</u> 1.	34 ^b	2.9	2
p + U	4.9	6 < Z <11	13.5 <u>+</u> 1.	20, 90, 160	1.9 ^C	11
p + U	5.5	3 ≮ Z ≲11	12.6 <u>+</u> 1.	20 - 160	1.7 (1.8)	12
C + Ag	0.36	4 < Z <22	8.3 <u>+</u> 1.	40 - 70	2.6 (2.6)	8
C + Ag	0.18	3 < Z <13	7.2 <u>+</u> 1.	50 - 70	3.0 (3.2)	8
C + Au	0.36	3 ≤ Z ≤11	7.7 <u>+</u> 1.	50 - 120	2.8 (2.8)	8
C + Au	0.18	4 ≤ Z ≤11	6.2 <u>+</u> 1.	50 - 120	3.8 (3.7)	8

TABLE I. Summary of the data used and the values of the apparent exponent obtained from the power-law fit to the fragment distributions.

^aThe apparent exponent, τ , obtained with the restricted range of masses, $5 \le Z$, is shown in parentheses.

 b An almost isotropic (to about 20%) angular distribution was assumed in obtaining the integrated total cross section (Refs. 2 and 11).

^cCross sections for Z = 6, 7, 8, taken from Ref. 12, were normalized to 4.9 GeV.

In Fig. 1 we plot the apparent exponent, τ , determined from a least-squares fit to the fragment distributions as a function of the temperature of the emitting system. We observe a dramatic temperature dependence of the apparent exponent, which decreases as the temperature increases up to about 11-12 MeV, after which the trend is reversed. The exponent appears to reach the minimum value of about 1.7 at a temperature of about 11-12 MeV. If we assign a critical temperature, T_c , corresponding to the minimum value of the apparent exponent, i.e., corresponding to the maximum probability for fragment emission, then from Fig. 1 we may conclude that $T_c \sim 11-12$ MeV. This indication is in disagreement with the result $T_c = 3.3$ MeV,³ obtained from the isotope distributions, but it is in agreement with calculations for finite nuclear systems,⁷ which predict T_c in the range of 9-13 MeV.

Since the deexcitation processes of the emitted fragments may make it difficult to obtain accurate information from lighter fragments, we restricted the range of the fragment mass to $Z \ge 5$ to obtain the power-law apparent exponent (see Table I). This new set of data produced the



FIG. 1. The apparent exponent, τ , of the power-law fit to the fragment distributions as a function of the temperature, *T*. The systems are, : circles, p + Ag(0.21-4.9 GeV); crosses, p + U (4.9,5.5 GeV); squares, p + Xe, p + Kr (80,350 GeV), triangles and inverted triangles, C + Ag, C + Au (180,360 MeV). The inset shows a typical power-law fit to a fragment distribution.

same dramatic features as the full set shown in Fig. 1.

This behavior of the temperature dependence of the fragment yield prompted us to fit the fragment distributions with expressions obtained from a theory of condensation in excited nuclear matter,^{4, 13} in which the probability for fragment formation of size A is given by

$$P(A) \sim A^{-k} \exp\{\left[-a_{s}'(T)A^{2/3} - a_{v}'(T)A + \mu(T)A\right]/T\},\$$

where k is the critical exponent, $a_s'(T) = a_s(T)$ -TS_s is the surface free energy per particle, $a_v'(T) = a_v(T) - TS_v$ is the volume free energy per particle, and $\mu(T)$ is the chemical potential per particle. The above relation can be written in the following form:

$$P(A) \sim A^{-k} X^{A^{2/3}} Y^{A}$$

where

$$X = \exp[-a_{s}'(T)/T],$$

$$Y = \exp\{-[a_{n}'(T) - \mu(T)]/T\}.$$

In the region $T < T_c$, where gas and liquid phases coexist, the sum of the volume energy per particle in the liquid phase and the Gibbs free energy per particle in the gaseous phase is equal to zero, $a_v'(T) - \mu(T) = 0$. Therefore, the exponential factors are Y = 1 and X < 1, and the probability P(A) can be written as

$$P(A) \sim A^{-k} \exp[-a_{s'}(T)A^{2/3}/T].$$
 (1)

At the critical point, determined by $T = T_c$, the surface free energy term is equal to zero, $a_s'(T) = 0$, and in addition $a_v'(T) - \mu(T) = 0$. Therefore, both exponential factors are X = 1 and Y = 1, and the probability assumes the pure power-law form

$$P(A) \sim A^{-k} \tag{2}$$

Finally for $T > T_c$, corresponding to a gas phase, we assume that the surface free energy is very small, $a_{s'}(T) \sim 0$, while $a_{v'}(T) - \mu(T) > 0$. Therefore, the exponential factors are $X \sim 1$ and Y < 1, and the probability P(A) assumes the form

$$P(A) \sim A^{-k} \exp\{-[a_{\nu}'(T) - \mu(T)]A/T\}.$$
 (3)

We note that the exponential factors X and Y, at temperatures $T < T_c$ and $T > T_c$, respectively, modulate the power-law dependence of the fragment distribution. Furthermore, the form of expression (3) has an A dependence similar to the coalescence formula, which is applicable to composite-fragment emission in high-energy collisions.¹⁴

We parametrized the temperature dependence of the surface free energy as

$$a_{s}'(T) = 18.4(1 - T/T_{c})^{2},$$
 (4)

since $a_{s}'(T=0) = 18.4$ MeV, the cold-nuclearmatter surface energy, and $a_{s}'(T=T_{c}) = 0$. For the volume and Gibbs free energies we take

$$a_{v}'(T) - \mu(T) = b(1 - T/T_{c})^{2}, \qquad (5)$$

where b is a coefficient not known a priori; a value in the range 8–10 MeV was determined by the fitting routine.¹⁵ A least-squares fit to the fragment distributions was undertaken by using expressions (1) and (3) with the parametrizations (4) and (5), respectively, and letting the exponent, k, take successively the values 1.6, 1.7, 1.8, 2.0, and 2.33 to yield corresponding T_c values. For the temperature of each system we used $T = T' \pm 1$ MeV, where T' is the temperature over all emitted fragments used in each fit. This variance in T, resulting from the uncertainty in establishing the temperature of the source, produced a variance in the calculated T_c .

Figure 2 shows the extracted critical temperature as a function of the critical exponent, k. We observe that for k between 1.7 and 1.8 the calculated T_c values for both $T < T_c$ and $T > T_c$ coincide, while for both lower and higher k values they diverge. We are, therefore, inclined to accept $k \sim 1.7$ as the critical exponent and T_c = 12.0 ± 0.2 MeV as the critical temperature. The uncertainty in the critical temperature allows the possibility that the critical exponent kmay take on a value between 1.7 and 1.8, which is lower than the theoretically minimum value 2.0 for an infinite system. This lower value may reflect the finite systems considered and needs further theoretical investigation. We note, however, that this choice of parametrization reproduces the essential features of Fig. 1. Calculations of thermal properties of nuclei,¹⁶ using the thermal Hartree-Fock approximation and the Skyrme III interaction, yield a critical temperature $T_c = 12.58$ MeV for the liquid-drop surface free energy, close to the temperature extracted by our analysis.

The similarity of the power-law dependence $(2.5 < \tau < 2.7)$ of the fragment distributions^{2,8} at very different temperatures $(8 \le T \le 15 \text{ MeV})$ can now be understood in our picture; these points lie on either side of the critical temperature and

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FIG. 2. The extracted critical temperature, T_c , as a function of the critical exponent, k, for $T < T_c$ and $T > T_c$.

the power law is about equally modified by the temperature-dependent exponential factors X and Y.

In this work it has been essential to assume the formation of thermalized hot matter of nuclear dimensions in the temperature range of 5 to 20 MeV. Furthermore, we have to rely on the freezeout concept, which ensures that the experimentally observed mass distribution reflects the configuration at the freezeout density of matter. Although both of the above concepts are generally accepted at very high energies (E/A > 500 MeV), they have to be confirmed by careful examination of experimental data. It is very encouraging, however, to find that the simple Ansatz, taken from the theory of condensation, works very well in this wide energy range and for very different systems.

Our analysis made use of fragment distributions from widely different systems and with appreciable uncertainty in the temperature of the source. To circumvent this inherent difficulty one should choose one system, such as Ne or Ar +Ag, and measure an excitation function of the fragment distributions, spanning the region of critical temperature of about 9 to 14 MeV. From such a study one may also be able to infer more accurately the form of the T dependence of the free energies very close to T_c , providing information concerning the equation of state of nuclear systems.

In conclusion, the evidence presented by our analysis indicates that critical phenomena, associated with phase transitions and manifested by the formation of intermediate-mass fragments, occur in equilibrated nuclear systems, as suggested by Refs. 2 and 3 but with quantitatively different results.

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