Experimental signals of a nuclear liquid-gas phase transition

J Mabiala$^1$, A Bonasera$^{1,2}$, H Zheng$^{1,3}$, A B McIntosh$^1$, Z Kohley$^{1,4,6}$, P Cammarata$^{1,4}$, K Hagel$^1$, L Heilborn$^{1,4}$, L W May$^{1,4}$, A Raphelt$^{1,4}$, G A Souliotis$^{1,5}$, A Zarrella$^{1,4}$, S J Yennello$^{1,4}$

$^1$Cyclotron Institute, Texas A&M University, College Station, Texas 77843, USA
$^2$Laboratori Nazionali del Sud, INFN, via Santa Sofia, 62, 95123 Catania, Italy
$^3$Physics Department, Texas A&M University, College Station, Texas 77843, USA
$^4$Chemistry Department, Texas A&M University, College Station, Texas 77843, USA
$^5$Laboratory of Physical Chemistry, Department of Chemistry, National and Kapodistrian University of Athens, Athens GR-15771, Greece

E-mail: jmabiala@comp.tamu.edu

Abstract. The critical phenomenon of the nuclear liquid-gas phase transition has been investigated in the reactions $^{64}$Zn+$^{64}$Zn, $^{64}$Ni+$^{64}$Ni and $^{70}$Zn+$^{70}$Zn at beam energy of 35 MeV/nucleon. Yields of fragments arising from fragmenting quasi-projectiles (QPs) with different neutron-proton asymmetries were analyzed within the framework of the Landau free energy approach. Fits to the free energy of fragments as a function of fragment asymmetry showed three minima, indicating the system to be in a regime of a first-order phase transition. The QP temperature estimates were extracted from the analysis of N=Z fragment data. Additionally, we make use of a recent method based on quantum fluctuations of fermions to derive temperatures and densities of selected QPs. Critical scaling of these observables is found for systems which differ in neutron to proton asymmetry. The derived critical exponent $\beta = 0.35 \pm 0.01$, belongs to the liquid-gas universality class.

1. Introduction

Heavy-ion collisions at the Fermi scale are dominated by nuclear fragmentation, the breakup of a nuclear system into several intermediate sized pieces, and often used to explore the phase diagram of nuclear matter. Because of the van der Waals-like nature of the nucleon-nucleon interaction, it is believed that nuclear matter is likely to exhibit a liquid-gas phase transition [1, 2, 3, 4, 5, 6, 7, 8]. However, unlike van der Waals fluids, nuclei are finite and two-component systems. Most of the divergences that are usually linked to a phase transition in macroscopic systems are washed out in these small systems [9]. The additional degree of freedom which is related to proton and neutron concentrations also makes phase transition more complex [10]. The phase transition should however manifest itself through different signals. Several methods explored in order to investigate nuclear phase transition are, among others, caloric curves [2, 11], critical exponents [12] and negative heat capacities [13]. However, in these

$^6$ Present address: National Superconducting Cyclotron Laboratory, Michigan State University, East Lansing, Michigan 48824, USA
works the interpretation of experimental data were based on either constant density or constant pressure.

The asymmetry energy is defined as the energy difference in pure neutron matter and uncharged symmetric nuclear matter. In earlier studies [14, 15, 16, 17, 18], the asymmetry energy has been shown to be the dominant contribution to the free energy in the vicinity of the critical point. The Landau free energy approach is therefore useful for the investigation of the liquid-gas phase transition in nuclear systems. In this approach, the neutron-proton asymmetry of the fragmenting source acts as an external field which can modify the fragment yields.

In this work, fragment yield data of fragmenting quasi-projectiles formed in $^{64}$Zn+$^{64}$Zn, $^{64}$Ni+$^{64}$Ni and $^{70}$Zn+$^{70}$Zn reactions at beam energy of 35 MeV/nucleon have been analyzed within the Landau free energy approach. Normalized pairing coefficients ($a_p/T$) were extracted from yield data of $N=Z$ fragments in order to correct for odd-even effects in free energy calculations. These values are used to estimate temperatures of fragmenting systems which differ in neutron-proton asymmetry. In addition, we extracted temperatures and densities of fragmenting systems using the method presented in Ref. [19, 20] which is based on quantum fluctuations of fermions. Scaling of these observables to their critical values displays universality, i.e. independence from the neutron-proton asymmetry of the source.

2. Experimental details

The experiment was performed at the Texas A&M University Cyclotron Institute. Beams of $^{64}$Zn, $^{64}$Ni and $^{70}$Zn at 35 MeV/nucleon were incident on targets of $^{64}$Zn, $^{64}$Ni and $^{70}$Zn respectively [21, 22]. The charged particles and free neutrons produced in the reactions were measured with the NIMROD-ISiS 4π detector array [23]. The granularity and excellent isotopic resolution provided by the array enabled the reconstruction of the quasi-projectile (QP) in mass, charge and excitation energy. The QP is the large excited primary fragment of the projectile following a non-central collision which will subsequently undergo breakup. The Neutron Ball [24] provided event-by-event multiplicity of the free neutrons emitted during each reaction. The QP source was selected by means of event-by-event cuts on the experimental data similar to those used in Refs. [25, 26] with its mass ($A$) restricted to the range $54 \leq A \leq 64$. The excitation energy was deduced using the measured free neutron multiplicity, the charged particle kinetic energies, and the energy needed for the breakup (Q-value).

Recently, the neutron-proton asymmetry of the source was identified as an additional order parameter in the nuclear phase transition [14, 15, 16]. Therefore, to investigate such a dependence, the data were sorted into four different source asymmetry ($m_s = (N - Z)/A$) bins ranging from 0.04 to 0.24 with a bin width of 0.05. The mean $m_s$ values corresponding to these $m_s$ bins are 0.065, 0.115, 0.165 and 0.215. The four $m_s$ bins will be referred to with their mean values in the rest of the discussion. The effects of QP excitation energies on the thermodynamic quantities were investigated by further gating the data into six bins in excitation energy, each 1 MeV wide, in the range of 3-9 MeV/nucleon.

3. The Landau free energy approach

In the modified Fisher model, the experimental fragment yield near the critical point of the emitting source can be written as [18]

$$ Y = y_0 A^{-\tau} e^{-F/T} A, $$

where $y_0$ is a constant, $F/T$ is the fragment free energy normalized to the temperature of the emitting source (QP), $A$ is the fragment mass number and $\tau$ is a critical exponent. In the Landau approach, $\langle F/T \rangle$ can be written in terms of an order parameter $m$ as given by [18]
The parameters \(a, b\) and \(c\) are fitting parameters which depend in general on temperature, and/or density of the fragmenting source. The neutron-proton asymmetry \(m\) of the fragment can be defined as an order parameter if \(m = -\partial F/\partial H\), where \(H\) is its conjugate variable \([3, 5]\). The quantity \(H\) acts as an external field arising from the neutron-proton asymmetry of the QP. In the absence of any external field \((H/T = 0)\), Eq. 2 has three minima with \(m = 0\) being the central minimum. The presence of an external field shifts the positions of all three minima.

The free energy data are usually obtained by normalizing fragment yields with respect to \(^{12}\)C yields to eliminate the constant \(y_0\). However, the free energy \(F/T\) values of \(N = Z\) nuclei were observed to significantly deviate from the regular behavior of the \(N \neq Z\) fragments. These so-called odd-even effects were corrected by a pairing coefficient extracted from an analysis of \(N = Z\) fragment yields. The following equation was used for the fragment yield corrected for pairing effects

\[
Y = y_0 A^{-\tau} e^{(-\frac{F}{T} - \frac{a_p}{T} \delta / A^{1/2}) A},
\]

where \(a_p\) is the pairing coefficient and \(\delta = -1, 0 \) and \(+1\) for odd-odd, odd-even and even-even nuclei, respectively. We note that the pairing free energy used here corresponds to the ground-state energy \((E_p = a_p \delta / A^{1/2})\). This term is expected to be different at extreme conditions of densities and temperatures.

As \(F/T = 0\) for \(N = Z\) fragments, taking the logarithm of Eq. 3 gives a linear equation where \(a_p/T\) is the slope and \(ln(y_0)\) the intercept. A linear fit to the \(N = Z\) data allows the extraction of the values of \(a_p/T\) and \(y_0\).

![Figure 1.](color online) \(a_p/T\) values as a function of the QP excitation energy. Different symbols correspond to data of different \(m_s\) bins. Errors bars represent fitting errors.

In Fig. 1, the \(a_p/T\) values obtained as slopes of the linear fit to the \(N = Z\) data for different \(m_s\) bins are plotted as a function of the QP excitation energy. It is observed that \(a_p/T\) systematically decreases with increasing QP excitation energy. The same trend has been observed in previous work \([18]\). A small dependence on the QP \(m_s\) bin is observed. This suggests a dependence of \(T\) and/or \(a_p\) on the neutron-proton asymmetry of the QP. The \(a_p/T\) values obtained are used to estimate the QP temperatures. Using \(a_p = 12\) MeV, the ground-state pairing energy coefficient, we obtained the QP temperature values of 5.16 MeV for \(m_s = 0.065\) and 5.15
MeV for \( m_s = 0.215 \) at \( E^*/A = 5.5 \) MeV. However, as the effect of temperature, density and the neutron-proton asymmetry on the pairing coefficient is not well understood, the QP temperature values extracted here are only reasonable estimates.

Figure 2. (Color online) \( F/T \) values as a function of fragment neutron-proton asymmetry \( m \) for an excitation energy of 6.5 MeV of the QP. Panels correspond to different neutron-proton asymmetry \( (m_s) \) bins of the QP. The values of \( a_p/T \) and \( y_0 \) used in the calculation of \( F/T \) were obtained from the analysis of \( N = Z \) fragments. Solid lines (Landau Fit1) are fit to the data using the complete Landau free energy (Eq. 2). Dashed lines (Landau Fit2) are fit to data using only the first and last terms of Eq. 2. Error bars corresponding to statistical errors are smaller than the points.

Figure 2 shows free energy \( (F/T) \) values for different \( m_s \) bins at an excitation energy of 6.5 MeV of the QP. Only charged particle yields are used in the Landau fitting since the efficiency for measuring neutrons differs from the efficiency for measuring charged particles. The calculated \( m_s \) values are, however, not affected by the uncertainty in measuring free neutrons as these partly contribute to the neutron excess. The value of \( \tau = 2.3 \pm 0.1 \), obtained in earlier experimental works from the power law dependence of mass yields [14, 15], was used in this study. Solid lines (Landau Fit1) are fit to the data using the complete Landau free energy (Eq. 2) with \( a, b, c \) and \( H/T \) as free parameters. Dashed lines (Landau Fit2) are fit to data using only the first and last terms of Eq. 2. It is clearly seen from the figure that the Landau equation provides a better fit to the free energy data. Three minima are shown in the free energy plot where the central minimum is shifted from zero, indicating the presence of an external field. The appearance of three minima might be a signature of a first-order phase transition.

The values of the fitting parameters \( a, b, c \) and \( H/T \) extracted for each \( m_s \) bin are plotted versus the QP excitation energy in Fig. 3. The parameters \( a, b \) and \( c \), which are related to thermodynamic properties of the fragmenting system show a dependence on \( m_s \). The parameter \( a \) decreases with increasing QP excitation energy, while \( b \) generally increases with increasing...
QP excitation energy. Within experimental uncertainties, the parameter \( c \) is almost constant over the entire range of QP excitation energy. A decrease of \( H/T \) is observed with an increase in the excitation energy. The values of \( H/T \) also show a systematic increase with increasing \( m_s \). This linear dependence qualitatively verifies that \( H \) is the conjugate variable of \( m_s \). The parameters \( a, b \) and \( c \) are very close to satisfying the condition for a first-order phase transition \( b = -\frac{4}{3}\sqrt{ac} \) \([5]\) within the error bars, as evidenced in Fig. 4. This interpretation would only be regarded as conclusive with data points at large \( m \) values that give a better constraint on the parameters \( a, b \) and \( c \). The dependence of the Landau parameters and \( a_p/T \) on the neutron-proton asymmetry of the fragmenting source decidedly indicates the dependence of the nuclear equation of state on the neutron-proton asymmetry of the source.

4. Quantum fluctuations of fermions

4.1. Temperatures and densities

The temperatures of the selected QPs which differ in neutron-proton asymmetry are calculated with the momentum quadrupole fluctuation method reported in Refs. \([19, 20]\). The momentum quadrupole is defined as \( Q_{xy} = p_x^2 - p_y^2 \) where \( p_x \) and \( p_y \) are the transverse components of a given fragment’s momentum. This quantity is zero on average in the center of mass of the equilibrated QP. The longitudinal component, \( p_z \), is excluded to minimize contributions from the collision dynamics, which manifest in the beam direction. In this work, we use protons as the probe particle.

Assuming a Fermi-Dirac distribution, the fluctuation of the momentum quadrupole \( \sigma_{xy}^2 \) is connected to the temperature \( T \) by the relation

\[
\sigma_{xy}^2 = 4m^2\bar{N}T^2F_{QC},
\]

where \( \bar{N} \) and \( m \) are the average multiplicity per event and the proton mass, respectively. \( F_{QC} \) is the quantum correction factor which converges to one at high \( T \) (classical limit) and is expressed
Figure 4. (Color online) Plot of $-4\sqrt{ac/3}$ as a function of $b$ for different bins in $m_s$. Error bars represent fitting errors. The dot-dashed line corresponds to $b = -4\sqrt{ac/3}$.

\begin{equation}
F_{QC} = 0.2 \left( \frac{T}{\varepsilon_f} \right)^{-1.71} + 1 ,
\end{equation}

where $\varepsilon_f = \varepsilon_{f0}(\rho/\rho_0)^{2/3}$ is the Fermi energy of the nuclear matter at density $\rho$. The quantities $\rho_0$ and $\varepsilon_{f0}$ denote the normal nuclear density and the corresponding Fermi energy, respectively. The values of $\rho_0 = 0.15$ fm$^{-3}$ and $\varepsilon_{f0} = 36$ MeV are used in the calculations.

Following a similar derivation, the normalized multiplicity fluctuation ($\sigma_N^2/N$) of fermions emitted from the QP is shown to depend on $T/\varepsilon_f$ [19, 20]. This quantity is parametrized in terms of $\sigma_N^2/N$, and given as

\begin{equation}
\frac{T}{\varepsilon_f} = -0.442 + \frac{0.442}{(1 - \sigma_N^2/N)^{0.656}} + 0.345 \left( \frac{\sigma_N^2}{N} \right) - 0.12 \left( \frac{\sigma_N^2}{N} \right)^2 .
\end{equation}

Once the normalized multiplicity fluctuation is experimentally determined for a given excitation energy, using Eq. 5 and Eq. 6 the quantum correction factor $F_{QC}$ can be obtained. From Eq. 4, one can easily calculate the temperature $T$ and, from the fact that $\varepsilon_f = \varepsilon_{f0}(\rho/\rho_0)^{2/3}$, the density $\rho$ can also be obtained. More details can be found in Refs. [19, 20].

Temperatures and densities of the QPs are plotted as a function of excitation energy per nucleon in Fig. 5. The extracted densities for the four source asymmetries (bottom panel of Fig. 5) show a dependence on the value of $m_s$. An overall ordering in the density with $m_s$ is observed for each excitation energy: the larger the asymmetry, the lower the density. A previous analysis has shown an ordering in $m_s$ bins of the temperatures within a classical treatment [27]. In the present treatment, the correlation between temperatures and densities indicates that the dependence on $m_s$ is manifest in the densities. However, the spacing between the density values for different $m_s$ bins increases as the excitation energy increases. Caloric curves, i.e. temperature as a function of excitation energy (top panel of Fig. 5), show a monotonic rising behavior. Within statistical errors, a small dependence on $m_s$ is observed.
4.2. Critical exponent $\beta$

The critical exponent $\beta$ is determined from the relation [5]

$$1 - \frac{\rho}{\rho_c} \propto \left(1 - \frac{T}{T_c}\right)^\beta . \quad (7)$$

This parameter defines the universality class of the system and, therefore, systems with similar $\beta$ values have similar underlying physics. Experimental data of reduced density $\rho/\rho_c$ versus reduced temperature $T/T_c$ for different $m_s$ bins must fall on the same line if they belong to the same universality class. This allows us to determine the critical temperature $T_c$ and the critical density $\rho_c$ for each $m_s$ bin. The values for $T_c$ and $\rho_c$ for each $m_s$ bin are reported in Table 1. The critical temperatures $T_c$ are observed to increase when increasing $m_s$. This trend is consistent with the results reported in Ref. [14]. The critical densities $\rho_c$, however, decrease with increasing $m_s$. Figure 6 shows an excellent fit to the data points which results in a slope $\beta = 0.35 \pm 0.01$. This value is in the range of that expected for the liquid-gas universality class [5, 28]. The fact that we are getting a value of $\beta$ consistent with a liquid-gas phase transition supports our strategy for calculating densities and temperatures from the Fermi gas assumption.

5. Summary and conclusion

Fragment yield data from QPs with different proton and neutron compositions have been analyzed using the Landau free energy approach. The Landau equation was used to fit free energies obtained from fragment yields. The fitted curves have shown three minima that may indicate a first-order phase transition. Extracted pairing coefficients from $N = Z$ nuclei gave
reasonable estimates of the temperatures of the QP sources. The parameters of the Landau equation as well as extracted pairing coefficients which are related to the state variables of the QP have been observed to depend on its proton-neutron composition and its excitation energy.

In addition, by means of a new quantum method based on quantum fluctuations of fermions, temperatures and densities of selected QP sources were derived. These state parameters and their corresponding critical values have shown a dependence on the QP neutron/proton concentration. The extracted critical exponent \( \beta \) has been found to belong to the liquid-gas universality class. These results provide a means to establish the proton-fraction dependence of the equation of state (EOS) in systems with large neutron excess, such as neutron stars.

**Acknowledgments**

This work was supported by the Robert A. Welch Foundation (A-1266) and the U. S. Department of Energy (DE-FG03-93ER-40773).

**References**

[1] Jaqaman H, Mekjian A Z and Zamick L 1983 *Phys. Rev. C* 27(6) 2782–2791
[2] Pochodzalla J, et al. 1995 *Phys. Rev. Lett.* 75(6) 1040–1043
[3] Landau L D and Lifshitz E M 1980 *Statistical Physics* (Pergamon, New York)
[4] Guggenheim E A 1945 *J. Chem. Phys.* 13 253–261
[5] Huang K 1987 *Statistical Mechanics* (Wiley & Sons, New York)
[6] Borderie B and Rivet M 2008 *Prog. Part. Nucl. Phys.* 61 551–601
[7] Pichon M et al. 2006 *Nucl. Phys. A* 779 267 – 296
