## Investigation of the nuclear phase transition using the Landau free-energy approach

J. Mabiala,<sup>1,\*</sup> A. Bonasera,<sup>1,2</sup> H. Zheng,<sup>1,3</sup> A. B. McIntosh,<sup>1</sup> L. W. May,<sup>1,4</sup> P. Cammarata,<sup>1,4</sup> Z. Kohley,<sup>1,4,†</sup> K. Hagel,<sup>1</sup> L. Heilborn,<sup>1,4</sup> A. Raphelt,<sup>1,4</sup> G. A. Souliotis,<sup>1,5</sup> A. Zarrella,<sup>1,4</sup> and S. J. Yennello<sup>1,4</sup>

<sup>1</sup>Cyclotron Institute, Texas A&M University, College Station, Texas 77843, USA

<sup>2</sup>Laboratori Nazionali del Sud, INFN, via Santa Sofia, 62, 95123 Catania, Italy

<sup>3</sup>Physics Department, Texas A&M University, College Station, Texas 77843, USA

<sup>4</sup>Chemistry Department, Texas A&M University, College Station, Texas 77843, USA

<sup>5</sup>Laboratory of Physical Chemistry, Department of Chemistry, National and Kapodistrian University of Athens, Athens GR-15771, Greece

(Received 26 October 2012; published 31 January 2013)

Fragment yield data have been analyzed using the Landau free energy approach to investigate the critical phenomena 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. Pairing coefficients normalized to the temperature of the fragmenting source  $(a_p/T)$  were extracted from yield data of N = Z fragments in order to correct for odd-even effects in free energy calculations. Plots of fragment free energy (F/T) versus the neutron-proton asymmetry show three minima, indicating the system to be in a first-order phase transition regime. Values of fitting parameters to the Landau free energy equation as well as  $a_p/T$ , which are related to thermodynamic properties of the fragmenting systems, exhibit a dependence on the neutron-proton asymmetry in addition to their dependence on the excitation energy of the fragmenting source.

DOI: 10.1103/PhysRevC.87.017603

PACS number(s): 21.65.Ef, 24.10.Pa, 25.70.Pq, 25.70.Mn

Studying the behavior of nuclear matter under extreme conditions of temperature and density, including possible phase transitions, is one of the most interesting phenomena in heavy-ion collisions in the Fermi energy domain. The existence of a nuclear phase transition is currently the subject of many investigations. Several papers have presented evidence for the observation of critical behavior in experimental data. This was done through caloric curves [1,2], critical exponents [3], negative heat capacities [4], and other observables [5–9].

Nuclei are two-component systems (consisting of protons and neutrons), and the change in nuclear energy associated with changing neutron-proton asymmetry is defined as the asymmetry energy. In earlier studies [10-14], the asymmetry energy has been shown to be the dominant contribution to the Landau free energy of fragments produced in various systems in the vicinity of the critical point. Note that, near the critical point for a liquid-gas phase transition, volume and surface have very small contributions. The effect of the Coulomb term has been observed to be of negligible importance in systems approaching the critical point [11,14].

The nature of the phase transition of nuclear matter is expected to be of the liquid-gas type due to the specific form of the nucleon-nucleon interaction. This nuclear matter phase transition is qualitatively very different from the phase transition phenomena studied in relativistic heavy-ion collisions (RHIC) [15]. Similar to ground-state shape phase transitions [16], the liquid-gas phase transition is sensitive to the proton-neutron interactions.

In this brief report, we have investigated the role of the neutron-proton asymmetry of the fragmenting source within the Landau free energy approach of phase transitions. A detailed analysis of fragment yield data from fragmenting systems formed in the symmetric entrance channels  ${}^{64}$ Zn +  ${}^{64}$ Zn,  $^{64}$ Ni +  $^{64}$ Ni, and  $^{70}$ Zn +  $^{70}$ Zn at 35 MeV/nucleon is presented. A similar work was performed with different systems by Tripathi et al. [12-14]. Increased silicon coverage, greater isotopic resolution and increased statistics have provided the opportunity to carry out a more detailed analysis presented here. It has been shown that a nuclear phase transition is driven by the difference in proton-neutron concentration of the fragments.

The experiment was performed at the Texas A&M University Cyclotron Institute. Charged particles were measured using the NIMROD-ISiS  $4\pi$  detector array, which was housed inside the TAMU Neutron Ball [17]. 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 with the target which will subsequently undergo breakup. The Neutron Ball [17] provided event-by-event multiplicity of the free neutrons emitted during each reaction. Details on the experiment are reported in Refs. [18–20]. In order to minimize the contribution from events that were dominated by nonequilibrium emission, limits were set on the deformation of the QP through a quadrupole cut [21]. The QPs were reconstructed by selecting events with the condition that the longitudinal velocity of fragments with  $Z = 1, 2, \ge 3$  be in the range of  $\pm 65\%$ ,  $\pm 60\%$ , and  $\pm 40\%$ , respectively, of the velocity of the heaviest fragment in the event [21,22]. The QP mass was restricted to be in the range  $54 \le A \le 64$ . Its excitation energy was deduced using the measured free neutron multiplicity, the charged particle kinetic energies, and the Q-value of the breakup [22]. To minimize any contribution from collective effects, the fragment kinetic energy was calculated using only

<sup>&</sup>lt;sup>\*</sup>jmabiala@comp.tamu.edu

<sup>&</sup>lt;sup>†</sup>Present address: National Superconducting Cyclotron Laboratory, Michigan State University, East Lansing, Michigan 48824, USA.

the fragment transverse velocity. Data were sorted into four different bins in QP source asymmetry  $[m_s = (N_s - Z_s)/A_s]$ ranging from 0.04 to 0.24 with bin width of 0.05. The source asymmetry  $m_s$  was calculated on an event-by-event basis from A and Z of the detected fragments. This value was corrected for free neutrons emitted by the QP [21,23]. The central  $m_s$ 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 central values in the rest of the discussion. Effects of QP excitation energies on the thermodynamic quantities were investigated by gating the data into six bins of 1 MeV in the range of 3–9 MeV/nucleon.

According to the modified Fisher model, the fragment yield near the critical point is given by [14]

$$Y = y_0 A^{-\tau} e^{-\frac{F}{T}A} , (1)$$

where F/T is the free energy per nucleon of the fragment normalized with respect to the system temperature T,  $y_0$ is a constant, A is the fragment mass number and  $\tau$  is a critical exponent. In the Landau free energy approach, the key assumption is that in the vicinity of the critical point, the free energy (F/T) can be expanded in a power series in the order parameter m. This is given by the following equation [14]:

$$\frac{F}{T} = \frac{1}{2}am^2 + \frac{1}{4}bm^4 + \frac{1}{6}cm^6 - \frac{H}{T}m, \qquad (2)$$

where  $m = (N_f - Z_f)/A_f$  and  $N_f$ ,  $Z_f$  and  $A_f$  are the neutron, proton, and mass numbers of the fragment, respectively. The quantity H is the conjugate variable of m and a, b, and c are fitting parameters which depend on the temperature, density or pressure of the fragmenting system. Note that in the absence of H, the free energy F/T is symmetric in the exchange of m to -m indicating that nuclear forces are invariant when exchanging  $N_f$  and  $Z_f$ . This symmetry is violated in the presence of the H which arises when the source is asymmetric in the chemical composition. The quantities m and H are related to each other through the relation  $m = -\partial F / \partial H$  [24,25]. In the absence of any external field (H/T = 0), the condition for the minimum at m = 0 is a > 0while the condition for two additional minima at |m| > 0 is  $b \leq -2\sqrt{ac}$ . The curve defined by  $b = -2\sqrt{ac}$  is the phase coexistence boundary which corresponds to superheating [24]. The condition for three minima at m = 0 and |m| > 0 to be degenerate with F/T = 0 is  $b = -4\sqrt{ac/3}$  [the line of first-order phase transition] [24]. Non-zero external fields lead to shifts of these boundaries.

The free energy F/T values are usually obtained by normalizing fragment yields with respect to <sup>12</sup>C yields to eliminate the constant  $y_0$ . However, for N = Z nuclei, where pairing effects are more easily seen, the F/T values 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 to take into account pairing effects:



FIG. 1. (Color online)  $a_p/T$  values as a function of QP excitation energy. Different symbols correspond to data of different  $m_s$  bins. Errors bars represent fitting errors.

where  $a_p$  is the pairing coefficient and  $\delta = -1$ , 0 and +1 for odd-odd, odd-even and even-even nuclei, respectively. 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 modified at different densities and temperatures.

As F/T = 0 for N = Z fragments, taking the logarithm of Eq. (3) gives the following linear equation:

$$\ln(YA^{\tau}) = \ln(y_0) + \frac{a_p}{T} \frac{\delta}{A^{1/2}}, \qquad (4)$$

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  $y_0$ .

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  $a_p/T$  values systematically decrease with increasing QP excitation energy. The same trend has been observed in previous work [14]. 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 fragmenting source. The  $a_p/T$  values obtained are used to estimate the temperature of the QP for each  $m_s$  bin. Using  $a_p = 12$  MeV, the ground-state pairing energy coefficient, we obtained the QP temperatures of 5.16 MeV for  $m_s = 0.065$  and 5.15 MeV for  $m_s = 0.215$  at  $E^*/A = 5.5$  MeV. As the effect of temperature, density and the neutron-proton asymmetry on the pairing coefficient is not known, the QP temperature values extracted here are only reasonable estimates. The determination of temperatures and densities of the selected QPs in the gas phase is discussed elsewhere [26].

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 not affected by the uncertainty in measuring free neutrons since most of these neutrons are bound in the fragments. The value of  $\tau = 2.3 \pm 0.1$ , obtained in earlier works from the power law dependence of mass yields [10,11], was used in this work. Solid lines (Landau Fit 1) are fits to the data using the complete Landau free energy [Eq. (2)] with *a*, *b*, *c*, and H/T as free parameters. Dashed lines (Landau Fit 2) are fits to data using only the first and last terms of Eq. (2), a case corresponding



FIG. 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. Different 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 = Zfragments. Solid lines (Landau Fit 1) are fits to the data using the complete Landau free energy [Eq. (2)]. Dashed lines (Landau Fit 2) are fits to data using only the first and last terms of Eq. (2). Error bars corresponding to statistical errors are smaller than the points.

to single phase systems. It is seen in the figure, however, that the complete form of Eq. (2) provides a better fit to the free energy data. Three minima are seen 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 is a signature of a first-order phase transition [24].

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



FIG. 3. (Color online) Fitting parameters a, b, c, and H/T obtained from fits to the free energy data as a function of the QP excitation energy for different bins in  $m_s$ . Error bars represent fitting errors.



FIG. 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 long-dashed line corresponds to the line of first-order transition  $(b = -4\sqrt{ac/3})$  while the short-dashed line represents the phase coexistence boundary  $(b = -2\sqrt{ac})$ .

uncertainties, the parameter c is almost constant over the entire range of QP excitation energy. The values of H/Tshow a systematic increase with increasing  $m_s$ , except for the lowest  $m_s$  bin which shows a relatively flat behavior. This linear dependence qualitatively verifies that H is the conjugate variable of  $m_s$ , as shown in Refs. [10–13,27]. A decrease of H/T with increasing QP excitation energy is also observed. The parameters a, b, and c very closely satisfy the condition for a first-order phase transition  $b = -4\sqrt{ac/3}$ , as evidenced in Fig. 4. Clearly, one sees that even considering the error bars the data lie inside the phase coexistence region which is above the short-dashed line  $(b = -2\sqrt{ac})$ . Below this line the system can only exist in a single phase. However, 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, band c.

In conclusion, fragment yield data from fragmenting sources with different proton and neutron concentrations 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 indicate the system to be in a regime of a first-order phase transition. Extracted pairing coefficients from N = Z nuclei give reasonable estimates of the temperatures of 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 and neutron concentration and its excitation energy. This suggests that the nuclear equation of state depends on the proton and neutron concentration of the fragmenting system. The present results lend further support to the validity of conclusions drawn in previous works on the Landau free energy approach. Hence, we regard these results as important from the perspective of using the Landau free energy approach for the description of the critical behavior in nuclei.

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

- [1] J. Pochodzalla et al., Phys. Rev. Lett. 75, 1040 (1995).
- [2] J. B. Natowitz et al., Phys. Rev. C 65, 034618 (2002).
- [3] J. E. Finn *et al.*, Phys. Rev. Lett. **49**, 1321 (1982).
- [4] M. D'Agostino et al., Phys. Lett. B 473, 219 (2000).
- [5] J. B. Elliott et al., Phys. Rev. Lett. 88, 042701 (2002).
- [6] M. Kleine Berkenbusch *et al.*, Phys. Rev. Lett. 88, 022701 (2001).
- [7] J. B. Elliott et al., Phys. Rev. C 67, 024609 (2003).
- [8] B. Borderie et al., Phys. Rev. Lett. 86, 3252 (2001).
- [9] M. Pichon et al., Nucl. Phys. A 779, 267 (2006).
- [10] A. Bonasera et al., Phys. Rev. Lett. 101, 122702 (2008).
- [11] M. Huang et al., Phys. Rev. C 81, 044618 (2010).
- [12] R. Tripathi et al., Phys. Rev. C 83, 054609 (2011).
- [13] R. Tripathi et al., J. Phys. Conf. Ser. 312, 082043 (2011).
- [14] R. Tripathi et al., Int. J. Mod. Phys. E 21, 1250019 (2012).
- [15] C. Spieles, H. Stöcker, and C. Greiner, Phys. Rev. C 57, 908 (1998).

- PHYSICAL REVIEW C 87, 017603 (2013)
- [16] P. Cejnar, J. Jolie, and R. F. Casten, Rev. Mod. Phys. 82, 2155 (2010).
- [17] R. Schmitt *et al.*, Nucl. Instrum. Methods Phys. Res. A **354**, 487 (1995).
- [18] Z. Kohley, Ph.D. thesis, Texas A&M University (2010).
- [19] Z. Kohley et al., Phys. Rev. C 83, 044601 (2011).
- [20] Z. Kohley et al., Phys. Rev. C 86, 044605 (2012).
- [21] S. Wuenschel, Ph.D. thesis, Texas A&M University (2009).
- [22] S. Wuenschel et al., Nucl. Phys. A 843, 1 (2010).
- [23] P. Marini *et al.*, arXiv:1211.2134 (2012), Nucl. Instrum. Methods Phys. Res. A (to be published).
- [24] K. Huang, *Statistical Mechanics* (Wiley & Sons, New York, 1987).
- [25] L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Pergamon, New York, 1980).
- [26] J. Mabiala et al., arXiv:1208.3480 (2012).
- [27] M. Huang et al., Nucl. Phys. A 847, 233 (2010).