The Quantum Nature of a Nuclear Phase Transition.

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In their ground states, atomic nuclei are quantum Fermi liquids. At finite temperatures and low densities, these nuclei may undergo a phase change similar to, but substantially different from, a classical liquid gas phase transition. As in the classical case, temperature is the control parameter while density and pressure are the conjugate variables. At variance with the classical case, in the nucleus the difference between the proton and neutron concentrations acts as an additional order parameter, for which the symmetry potential is the conjugate variable. Different ratios of the neutron to proton concentrations lead to different critical points for the phase transition. This is analogous to the phase transitions occurring in 4 He−3 He liquid mixtures. We present experimental results which reveal the N/Z dependence of the phase transition and discuss possible implications of these observations in terms of the Landau Free Energy description of critical phenomena.

In recent times a large body of experimental evidence has been interpreted as demonstrating the occurrence of a phase transition in finite nuclei at temperatures (T) of the order of 10 MeV and at densities, ρ, less than half of the normal ground state nuclear density [1]. Even though strong signals for a first and a second-order phase transition have been found [1, 2], there remain a number of open questions regarding the Equation of State of nuclear matter near the critical point. In particular the roles of Coulomb, symmetry, pairing and shell effects have yet to be clearly delineated. There is a general consensus that in finite atomic nuclei non-equilibrium effects might play an important role, however, statistical models are very successful in reproducing accurately a large variety of experimental data [3]. This is supported by advanced experimental techniques able to isolate an equilibrated region for each collision and to the statistical averages over those events.

A nucleus excited in a collision expands nearly adiabatically until it is close to the instability region thus the expansion is isentropic [3]. At the last stage of the expansion the role of the Coulomb force becomes very important. In fact, without the Coulomb force, the system would require a much larger initial compression and/or temperature in order to enter the instability region and fragment. The Coulomb force acts as an external piston, giving to the system an `extra push' to finally fragment. These features are clearly seen in Classical Molecular Dynamics (CMD) simulations of expanding drops with and without a Coulomb field [3, 4]. The expansion with Coulomb included is very slow in the later stage and nearly isothermal.

Even though the analogy to classical systems is quite useful, it should not be overemphasized as in the (T, ρ) region of interest, the nucleus is still a strongly interacting quantum system, while at high T and small ρ the nucleus behaves as a classical fluid. In particular the ratio of T to the Fermi energy at the (presumed) critical point is still smaller than 1 which suggests that the Equation of State (EOS) of a nuclear system is quite different from the classical one. To date this expected difference has not been well explored [1, 6, 7, 8, 9, 10]. In this paper we will discuss experimental evidence which indicates that the phase transition is strongly influenced by the relative proton and neutron concentrations. We show that near the critical temperature for a second-order phase transition, the quantity I/A=(N-Z)/A behaves as an order parameter and the difference in chemical potential between the neutrons and protons is its conjugate variable. We also note that the phase transition has a strong resemblance to that observed in superfluid mixtures of liquid 4 He−3 He near the λ point. In both systems, changing the concentration of one of the components of the mixture, changes the characteristics of the EOS. Furthermore, at high concentrations there is a first-order [11, 12]. The analogy should not be stressed too much since in our case we have two strongly interacting Fermi liquids while in the He mixtures we have mixed bosons and fermions. However, phase transitions exhibit universal features, which are independent on the details of the forces and of the systems involved.

The experiment was performed at the Texas A&M University Cyclotron Institute using the K500 Superconducting Cyclotron. Beams of 64 Zn, 70 Zn and 64 Ni at 40 A MeV were incident on targets of 58 Ni, 64 Ni, 112 Sn, 124 Sn, 197 Au and 232 Th. Emphasis was placed upon obtaining high quality isotopic identification and high statistics for isotopes with Z between 3 and 16, for all the systems.

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In order to achieve this goal, a Si telescope, which consisted of four layers of 5cm x 5cm Si quadrant detectors of 129, 300, 1000 and 1000 μm thicknesses was centered at 20°. In this telescope four Li isotopes and six to seven isotopes of each element from Z = 4 up to Z = 16 were clearly identified with an energy threshold ranging from 5 A MeV for Li isotopes to 15 A MeV for Si isotopes. In addition 16 CsI light charged particle detectors and 16 neutron detectors were also placed around the target to detect coincident light particles. In this paper, however, only results for isotopes measured inclusively in the telescope are presented. In order to get the angle integrated yields of isotopes, the energy spectra observed at 17.5° and 22.5° were fit using a moving source parameterization. In the following we will show evidence that, when scaled properly, the yields for all the systems studied behave in a very similar fashion, a necessary condition for systems undergoing phase transitions.

The key factor of our analysis is the value \( I \), proportional to the third component of isospin, of the detected fragments. For instance, a plot of the yield versus mass number when \( I = 1 \) displays a power law with exponent \( \tau = 2.4 \). This is shown in fig. (1) for the \( ^{64}\text{Ni} + ^{232}\text{Th} \) case at 40 MeV/nucleon. Similar plots for \( I = 3 \) give a much flatter distribution. In Figure 1 we have made separate fits for each case sorting odd Z(open symbols) or even Z(filled symbols) separately. As we see we obtain four different curves which suggests that pairing is playing a role in the dynamics. Notice that the plotted yields are for odd \( A \) nuclei for which we expect pairing to be zero\[13\]. This implies that the observed fragments have emitted at least one neutron before cooling down. In that case the parent nuclei would be even-even and odd-odd nuclei\[14\]. This could explain the shifts of the distributions, in particular with the even-even yields being flatter than the odd-odd cases since those fragments are more bound. In general, the mass distributions for \(-2 \leq I \leq 4\), when fitted with a power law, give exponents ranging from 2.5 to 0. For our data, using yields of fragments with \( I = 0 \) or \( I = 1 \) we find an average value of \( \tau = 2.3 \pm 0.1 \).

The observance of this power law suggests that the mass distributions may be discussed in terms of a modified Fisher model\[2]\:

\[
Y = y_0 A^{-\tau} e^{-\beta \Delta \mu A}, \tag{1}
\]

where \( y_0 \) is a normalization constant, \( \beta \) is the inverse temperature and \( \Delta \mu = F(I/A) \) is the difference in chemical potential between neutrons and protons, i.e., the Gibbs free energy per particle, \( F \), near the critical point. If we accept that \( F \) is dominated by the symmetry energy and make the approximation that \( F(I/A) = 25(I/A)^2 \) MeV/A, i.e. the symmetry energy of a nucleus in its ground state\[12\]. We will use this relationship in order to infer an approximate value of the temperature of the system. However, we stress that in actuality, \( F(I/A) \) is a function of density, temperature and all other relevant quantities near the critical point. In the literature\[1\] ground state values of the symmetry energy coefficient or of binding energies of nuclei are often employed to derive the temperatures reached by the nuclei in the collisions. This is not generally correct since the values appropriate to the density and temperature sampled should be used. Since the ratio of the chemical potential to the temperature enters equation(1) or other similar equations obtained from statistical models\[1], deriving the true value of the temperature from yields or yield ratios alone is possible only when the chemical potential is known.

According to the Fisher equation given above, we can compare all systems on the same basis by normalizing the yields and factoring out the power law term. For this purpose we have chosen to normalize the yield data to the \( ^{12}\text{C} \) yield, i.e. we define a ratio:

\[
R = \frac{Y A^7}{(^{12}\text{C})12^7}, \tag{2}
\]

The choice to normalize to \( ^{12}\text{C} \) is not arbitrary. We want to scale to the power law dependence in the equation (1) and for this reason we choose a nucleus, which belongs to a power law distribution \( I = 0, 1 \) cases discussed above). Using different nuclei belonging to the same groups gives results similar to the ones discussed below. On the other hand using nuclei from groups (e.g. \( I = 4 \)) exhibiting smaller power law \( \tau \) parameters introduces a spurious constant/A dependence that destroys the scaling discussed below. The normalized ratios for the system \( ^{64}\text{Ni} + ^{64}\text{Ni} \) at 40 MeV/nucleon are plotted as a function of the (ground state) symmetry energy in figure (2). The data display an exponential decrease with...
increasing symmetry energy, except for the isotopes for which \( I = 0 \). The yields of these \( I = 0 \) isotopes are clearly not sensitive to the symmetry energy but rather to the Coulomb and pairing energies and possibly to shell effects. The appearance of two exponential curves with the same exponent will become clear from the discussion below. A fit using eqs. (1) and (2) gives an 'apparent temperature' \( \Theta \) of 6.0 MeV. This value of \( \Theta \) would be the real one if \( \mu = 25 \text{ MeV} \) (the g.s. symmetry energy coefficient value) and if secondary decay effects are negligible. In general we expect that the symmetry energy coefficient is density and temperature dependent, as we will discuss in the framework of the Landau free energy approach below. We stress that the appearance of two branches in fig. 2, clearly indicates that the free energy must contain an odd power term in \((I/A)\) at variance with the ground state symmetry energy.

![Figure 2](image_url)

**FIG. 2:** Ratio versus symmetry energy for the \(^{64}\text{Ni} + ^{64}\text{Ni}\) case at 40 MeV/nucleon. The dashed lines are fits using a ground state symmetry energy, eq. 1, and an apparent 'temperature' of 6 MeV. The \( I < 0 \) and \( I > 0 \) isotopes are indicated by the open and full circles respectively. The \( I = 0 \) cases by the full squares.

To further explore the role of the relative nucleon concentrations we plot in figure (3) the quantity \( F = -\frac{\ln(ratio)}{A} \) versus \((I/A)\) i.e. the neutron to proton concentration. As expected the normalized yield ratios depend strongly on \((I/A)\). Pursuing the question of phase transition we can perform a fit to these data within the Landau description. In this approach the ratio of the free energy to the temperature is written in terms of an expansion:

\[
\frac{F}{T} = \frac{1}{2} a m^2 + \frac{1}{4} b m^4 + \frac{1}{6} c m^6 - m \frac{H}{T} \tag{3}
\]

where \( m = (I/A) \) is an order parameter, \( H \) is its conjugate variable and \( a - c \) are fitting parameters. The introduction of the conjugate variable \( H \) is necessary since the order parameter can be obtained from the derivative of the Gibbs free energy with respect to \( H \). A practical consequence of this is that we expect that the ratio plotted in fig. 2 should display two branches the lower one referring essentially to proton-rich and the higher one to neutron-rich isotopes. In fig. 2 only a few isotopes are seen in the lower yield branch since other proton-rich nuclei either have a short lifetime or evaporate some particle before reaching the detector. Microscopic Anisymmetrized Molecular Dynamics (AMD) calculations, which are dynamical calculations taking into account the Pauli principle as well as a realistic nuclear mean field, allow the identification of the primary excited fragments and clearly indicate the existence of such a lower branch in the first stages of the reaction. The existence of the two branches also clearly appears from the quantities plotted in fig. 3.

![Figure 3](image_url)

**FIG. 3:** (Minus) free energy versus symmetry term for the case \(^{64}\text{Ni} + ^{58}\text{Ni} \) (upper panel) and \(^{64}\text{Ni} + ^{232}\text{Th} \) at 40 MeV/nucleon (lower panel). The dashed lines are fits based on Landau \(O(m^6)\) free energy either for a second-order(long-dashed) or first-order (short-dashed) phase transition (see text).

We stress that the use of the Landau approach is for guidance only and now proceed to discuss some possible scenarios that should be tested in future studies. If we force the parameter \( c = 0 \) in eq. 3, i.e. we reduce the Landau free energy to fourth order (which implies a second-order phase transition with given critical exponents) and we fit the data of fig. 3 we obtain \( a = 19.2 \) and \( b = -130.73 \). This result is unphysical since it implies that the free energy is negative for large \( m \). A fit using eq. 3 gives the following values for the \(^{64}\text{Ni} + ^{58}\text{Ni} \) (\(^{64}\text{Ni} + ^{232}\text{Th} \) : \( a = 23.5(18.86), b = -413.8(-260.3), c = 2848.3(1408.1) \) and \( H/T = 0.79(1.06) \) and is displayed in fig. 3(long-dashed line). This case is discussed in detail in [11] and corresponds to a ‘classical’ second-order phase transition. However it also indicates that a tricritical behaviour is
possible for different temperatures and/or densities and could be obtained in different experimental conditions. There are some features that it is important to emphasize:

1) In Landau’s approach a line of first-order phase transitions is given by the condition

\[ b = -4\sqrt{ca^3} \]  

Using the values of \( a \) and \( c \) given above we get \( b = -597.5(-376.3) \) which is close to the fitted values given above. In particular if we substitute eq.(4) into eq.(3) and perform the fit of the experimental data given in fig. 3, we obtain a result (given by the short-dashed line) of a similar quality to that seen for the case of a ‘classical’ second-order phase transition. In the case of a first-order transition the fit bends up for large (positive) \( m \), however the data do not distinguish between the two cases. In fact, to explore this possibility requires high precision data for very asymmetric isotopes even for small mass numbers. This is beyond the capability of the present data.

2) The position of the maximum in fig. 3 (minimum of the free energy) is displaced from \( I = 0 \), and depends on the proton/neutron ratio of the emitting source. For \( N = Z \) sources we would expect the maximum at \( I = 0 \). Nevertheless a discontinuity remains since the fragments that have \( I = 0 \) have different yields.

These features are reminiscent of a superfluid \( \lambda \) transition observed as some \(^3\)He is added to \(^4\)He\(^{10}\). Starting from pure \(^4\)He which has a critical temperature of 2.18 K, the critical temperature for the second-order transitions decreases with increasing \(^3\)He concentration until at a lower temperature, \( T = 0.867\) K, a first-order transition appears. This point is known as the tricritical point for this system. Similarly, a nucleus, which should undergo a liquid-gas phase transition, is influenced by the different neutron to proton concentrations. Thus the discontinuity observed in fig. 3 could be a signature for a tricritical point as in the \(^4\)He\(^{10}\) case. We believe that our data, together with the use of the Landau \( O(m^4) \) free energy, suggest such a feature but are not sufficient to clearly demonstrate it. Some other works\(^{17, 18}\), also suggest that a line of critical points might be found away from its ‘canonical’ position, i.e. at the end of a first-order phase transition and, for small systems, even extending into the coexistence region.

3) Our choice of the free energy given by eqs. 3 and 4, if analytically extended (\( H/T = 0 \)) for large asymmetries displays three equal minima. However we would like to stress that such minima might not be found experimentally, because secondary evaporation effects can strongly influence the yields of isotopes far from symmetry.

4) As expected the position of the maximum, i.e. of the critical value of \((I/A)_c\) depends on the \((I/A)\) of the source. This is shown in fig. 4 where such a critical value is plotted as a function of the symmetry divided by the Coulomb energy of the compound system. We note this dependence on the compound nucleus asymmetry but recognize that the actual value of the peak could be shifted because of secondary evaporation effects. We have explored this using the AMD code of A. Ono\(^{10}\) to determine yields of both the primary (300 fm/c) hot fragments and secondary cold fragments remaining after fragment de-excitation. In fig. 4 we display these calculated yields. As we see from the calculations there is indeed a shift even though the qualitative features of the phase transition are preserved. Notice that in the calculations the critical value \((I/A)\) is not zero even for collisions of initially symmetric \( N = Z \) nuclei. However, we would predict that experimental data for such a system would exactly give zero since the calculations are systematically higher than data for the systems studied here. We stress that if the critical \( m = (I/A) \) is zero (i.e. \( H = 0 \)) it implies that the order parameter is zero above the critical point\(^{11, 12}\). In the cases studied here, since \( H \neq 0 \) in all cases, in agreement with the results of phase transitions in presence of an external field\(^{12}\), we suggest that the order parameter \( m \) is never zero even above the critical temperature. This implies that above the critical point the system has a lower degree of symmetry than in the case with \( H = 0 \). An experimental investigation of this feature would be very interesting and this could be accomplished in collisions of \( N = Z \) nuclei (eg. \(^{40}\)Ca + \(^{40}\)Ca) as function of beam energy in the same conditions as the present measurements.

![FIG. 4: Critical I/A (upper panel solid circles) and apparent temperature (lower panel solid circles) versus ratio of the compound nucleus symmetry to Coulomb energies for all the experimentally investigated systems. The same quantities are plotted for AMD calculations (open symbols). The lines are drawn to guide the eye.](image-url)
Thus we could expect the critical ‘T’ to increase because of a reduction of the surface term with respect to the volume term. However, at the same time we increase the Coulomb and the symmetry term contributions which would normally act to reduce the critical temperature. Because the system expands, it is apparently able to rearrange in such a way as to reduce the effect of the Coulomb force as much as possible. For this reason we have chosen in fig. 4 to plot the displayed quantities as a function of the ratio of the (g.s.) symmetry energy to the corresponding Coulomb energy. We expect that when this ratio approaches 1 the ‘T’ value should start to saturate and eventually decrease.

In conclusion, in this paper we have presented and discussed experimental evidence for the observation of a quantum phase transition in nuclei, driven by the neutron/proton asymmetry. Using the Landau approach, we have derived the free energy for our systems and found that it is consistent with a line of first-order phase transitions terminating at a point where the system undergoes a second-order transition. The properties of the critical point depend on the symmetry and the critical temperature increases for increasing asymmetry (and source size). This is analogous to the well known superfluid $\lambda$ transition in $^3$He-$^4$He mixtures. We suggest that a tricritical point, observed in $^3$He-$^4$He systems may also be observable in fragmenting nuclei. These features call for further vigorous experimental investigations using high performance detector systems with excellent isotopic identification capabilities. Extension of these investigations to much larger asymmetries should be feasible as more exotic radioactive beams become available in the appropriate energy range.

Exploration of quantum phase transitions in nuclei is important to our understanding of the nuclear equation of state and can have a significant impact in nuclear astrophysics, helping to clarify the evolution of massive stars, supernovae explosions and neutron star formation.

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[1] WCI proceedings, Ph. Chomaz et al., eds., EPJ A30 (2006), numb.1.
[2] A. Bonasera et al., Rivista Nuovo Cimento, 23 (2000) 1.
[3] H. Muller and B. D. Serot, Phys. Rev. C52 (1995) 2072; P. Siemens and G. Bertsch, Phys. Lett. B126 (1983) 9; P. J. Siemens, Nature 336 (1988) 109.
[4] M. Belkacem et al., Phys. Rev. C52 (1995) 271; A. Bonasera et al., Phys. Lett. B244 (1990) 169.
[5] C. Dorso et al., Phys. Rev. C60 (1999) 034604; M. Belkacem et al., Phys Rev. C54 (1996) 2435.
[6] L. Moretto et al., Phys. Rev. Lett. 88 (2002) 042701 and Phys. Rev. Lett. 94 (2005) 202701.
[7] M. D'Agostino et al., Nucl. Phys. A650 (1999) 329.
[8] P. F. Mastinu et al., Phys. Rev. Lett. 76 (1996) 2646.
[9] K. Hagel et al., Phys. Rev. C62 (2000) 034607.
[10] J. Pochodzalla et al., Phys. Rev. Lett. 75 (1995) 1040.
[11] K. Huang, Statistical Mechanics, second edition, ch.16-17, J. Wiley and Sons, New York, 1987; A. Hosaka and H. Toki, Quarks, Baryons and Chiral Symmetry, ch. , World Sci.,Singapore (2001).
[12] L. D. Landau, E. M. Lifshitz, Statistical Physics, 3rd edition, Pergamon press, New York,1989.
[13] M. A. Preston, Physics of the nucleus, Addison-Wesley pub., Reading-Mass. (1962).
[14] N. Marie et al., Phys. Rev. C58 256 (1998).
[15] M. E. Fisher, Rep. Prog. Phys. 30 (1967) 615.
[16] A. Ono and H. Horiiuchi, Progr. Part. Nucl. Phys. 53 (1996) 2958; A. Ono, Phys. Rev. C59 (1999) 853; A. Ono and H. Horiiuchi, Phys. Rev. C53 (1996) 2958.
[17] X. Campi, H. Krivine and N. Sator, Nucl. Phys. A681 (2001)458.
[18] F. Gulminelli et al., Phys. Rev. C65 (2002) 051601.