The density and temperature dependence of the nuclear symmetry free energy is investigated using microscopic two- and three-body nuclear potentials constructed from chiral effective field theory. The nuclear force models and many-body methods are benchmarked to properties of isospin-symmetric nuclear matter in the vicinity of the saturation density as well as the virial expansion of the neutron matter equation of state at low fugacities. The free energy per particle of nuclear matter with varying neutron-to-proton ratio is calculated assuming a quadratic dependence of the interaction contributions on the isospin asymmetry. The thermodynamic equation of state of isospin-asymmetric nuclear matter is examined in detail.

I. INTRODUCTION

Constructing the thermodynamic equation of state (EoS) of dense nucleonic matter is a central objective in modern nuclear theory. Of particular importance is to extract the nuclear symmetry free energy, which characterizes the dependence of the EoS on the ratio of protons to neutrons in the system. The symmetry free energy as a function of density and temperature is essential to understand heavy-ion collision dynamics, the structure of neutron stars, heavy element nucleosynthesis, core-collapse supernovae and the cooling of proto-neutron stars [1]. Next-generation radioactive beam facilities [2, 3] studying the reactions and structure of exotic neutron-rich isotopes provide further motivation to improve our microscopic description of highly isospin-asymmetric matter. The present work is a first step toward the development of a chiral effective field theory thermodynamic equation of state across the temperatures, densities and isospin asymmetries relevant for describing astrophysical phenomena and the matter produced experimentally in heavy-ion collisions at moderate energies.

Chiral effective field theory ($\chi$EFT) provides the basis for the study of strongly-interacting matter at the energy scales characteristic of normal nuclei [4–6]. In $\chi$EFT, microscopic nuclear interactions are organized in a systematic expansion, with many-nucleon forces naturally included. The low-energy constants parametrizing the interactions are generally fixed by high-precision fits to nucleon-nucleon scattering phase shifts and properties of light nuclei. Employing chiral interactions in calculations of nuclear many-body systems then gives pure predictions without additional fine tuning, and theoretical uncertainties can be estimated [6–10] by varying the resolution scale, the fitting procedures applied to fix the low-energy constants, and chiral order of the nuclear potentials. In the case of isospin-symmetric nuclear matter, (semi-)empirical constraints from the zero-temperature saturation energy, density and incompressibility as well as the critical point of the nuclear liquid-gas phase transition have been reproduced with low-momentum chiral nuclear forces in many-body perturbation theory [11, 12]. This motivates a study of isospin-asymmetric matter, which is by comparison much less constrained by experimental data.

In the present work we examine the density and temperature dependence of the symmetry free energy $F_{\text{sym}}(T, \rho)$, defined as the difference between the free energy per particle in homogeneous isospin-symmetric matter (SNM) and pure neutron matter (PNM):

$$F_{\text{sym}}(T, \rho) = F(T, \rho, \delta = 1) - F(T, \rho, \delta = 0),$$  

where $T$ is the temperature, $\rho = \rho_n + \rho_p$ is the total nucleon density, and $\delta = (\rho_n - \rho_p)/(\rho_n + \rho_p)$ is the isospin asymmetry parameter (with $\rho_n/p$ the neutron/proton density). The symmetry free energy obeys the following approximate relation for the free energy per nucleon in homogeneous nuclear matter with proton fraction $Y_p = (1 - \delta)/2$:

$$F(T, \rho, \delta) \approx F(T, \rho, \delta = 0) + F_{\text{sym}}(T, \rho) \delta^2,$$

*Electronic address: corbinian.wellenhofer@tum.de
The approximate quadratic dependence of $\bar{F}(T, \rho, \delta)$ on the isospin asymmetry at zero temperature has been validated in various microscopic many-body calculations (see e.g., Refs. [13–14]). However, at finite temperatures the free Fermi gas contribution to $\bar{F}(T, \rho, \delta)$ already contains large terms with quartic and higher powers of $\delta$, which we quantify explicitly in this work. By comparison, in initial calculations we have found that at the densities and temperatures relevant for the nuclear liquid-gas phase transition the interaction contributions give rise to weaker quartic terms and will therefore be assumed to have a quadratic dependence on $\delta$ in this work. Future research will address the accuracy of this approximation in greater detail.

The liquid-gas phase transition in isospin-asymmetric nuclear matter (a binary thermodynamic system) involves a solubility gap (noncongruent phase transition [15]). If isospin-symmetry breaking effects are neglected the solubility gap and the related distillation effects vanish in SNM where neutrons and protons are thermodynamically indistinguishable and the system behaves like a pure substance. The new features of the phase transition in ANM were discussed in detail in Refs. [15–19] using different phenomenological models of the nuclear force. The transition region is comprised of regions of metastable or unstable single-phase equilibrium, corresponding to different dynamical phase separation mechanisms: nucleation and spinodal decomposition [20, 21]. In this work we focus mostly on the spinodal which delineates the inner region of thermodynamic instability where no metastable state can exist. The evolution of the phase coexistence region with increasing isospin asymmetry is analyzed in terms of the trajectory of the critical temperature $T_c(\delta)$. Moreover, we determine the neutron drip point in cold nuclear matter and the temperature above which no self-bound drop of liquid nuclear matter can exist. We note that an accurate treatment of the liquid-gas coexistence region in (finite) nucleonic matter needs to account for Coulomb interactions and the surface tension of the liquid-gas interfaces. In particular, at low temperatures and densities close to nuclear saturation density the competition between short-range nuclear attraction and long-range Coulomb repulsion of protons entails phase ordering with nontrivial spatial structures, so-called nuclear pasta [22]. The dynamics of pasta formation has been recently studied using molecular dynamics simulations in Ref. [23]. Incorporating these effects as well as the presence of few-nucleon bound-states (nuclei) at very low densities [24–26] represents a future challenge.

The paper is organized as follows. In Sec. II we recall the main results for the EoS of SNM obtained in Ref. [12], and show results for additional derived thermodynamic quantities, i.e., the entropy per nucleon and the internal energy per nucleon. In Sec. III we extend the calculations to pure neutron matter. The zero-temperature results are compared to those from recent quantum Monte Carlo simulations while the finite-temperature EoS at low densities is compared to the virial expansion. In Sec. IV we investigate the thermodynamic EoS of isospin-asymmetric nuclear matter is studied in Sec. V. In particular, we examine in detail the dependence on isospin asymmetry of the EoS of a free nucleon gas. Finally, Sec. VI provides a short summary.

II. ISOSPIN-SYMMETRIC NUCLEAR MATTER

In Ref. [12] we calculated the free energy per nucleon in infinite homogeneous SNM using the Kohn-Luttinger-Ward many-body perturbation series [27–28] including contributions up to second order, i.e.,

$$\bar{F}(T, \mu_0, \delta = 0) = \bar{F}_0(T, \mu_0, 0) + \bar{F}_{el}(T, \mu_0, 0) + \lambda \bar{F}_1(T, \mu_0, 0) + \lambda^2 \bar{F}_2(T, \mu_0, 0) + O(\lambda^3).$$  \hspace{1cm} (3)

Here, $\lambda$ counts the number of interaction insertions, $\bar{F}_0(T, \mu_0)$ corresponds to a nonrelativistic free nucleon gas, and $\bar{F}_{el}(T, \mu_0)$ is a correction term which together with $\bar{F}_0$ reproduces the properties of a relativistic free nucleon gas over a wide range of densities and temperatures [29]. The first- and second-order terms $\bar{F}_1(T, \mu_0, 0)$ and $\bar{F}_2(T, \mu_0, 0)$ receive contributions from both the two-body and the three-body nuclear force. The second-order term $\bar{F}_2(T, \mu_0, 0)$ includes (temperature and density dependent) self-energy corrections, and has been evaluated by approximating the three-nucleon interaction with a temperature and density dependent effective two-body potential [for details see Refs. [12, 30–33]]. Explicit formulas for the different contributions in Eq. (3) are given in Ref. [12]. The effective one-body chemical potential $\mu_0$ is in one-to-one correspondence with the nucleon density via

$$\rho(T, \mu_0, \delta = 0) = \frac{1}{\pi^2} \sum_{\tau} \int_0^{\infty} dk k^2 \left[ 1 + \exp \frac{k^2/2M - \mu_0}{T} \right]^{-1},$$  \hspace{1cm} (4)

where $\tau \in \{-1/2, 1/2\}$ is the isospin projection quantum number and $M \approx 938.9$ MeV is the average nucleon mass. For Eq. (3) to be sufficiently converged at second order in $\lambda$, low-momentum interactions
have to be used, i.e., interactions with restricted resolution in coordinate space (corresponding to an ultraviolet cutoff in momentum space). The various sets of N3LO (i.e., fourth order in the chiral expansion) two-body and N2LO three-body chiral low-momentum interactions used in Ref. [12] correspond to different regularization methods, resolution scales $\Lambda$, and low-energy constants. For interactions constructed at resolution scales $\Lambda \leq 450$ MeV appropriate perturbative behavior was found. The SNM equation of state obtained from the sets of two- and three-body potentials denoted by n3lo414 ($\Lambda = 414$ MeV) and n3lo450 ($\Lambda = 450$ MeV), respectively, (see Refs. [11, 34, 35] for details) agree with empirical constraints from the zero-temperature saturation energy, density and incompressibility [36–39], and with estimates for the critical point of the nuclear liquid-gas phase transition obtained through the analysis of data from multifragmentation, fission and compound nuclear decay experiments [40–43]. The values of these quantities obtained from n3lo414 and n3lo450 in Ref. [12] are displayed in Table I.

\begin{figure*}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{(Color online) Results for the free energy per nucleon $\bar{F}(T, \rho, \delta = 0)$, the pressure $P(\rho, T, \delta = 0)$, the entropy per nucleon $\bar{S}(T, \rho, \delta = 0)$ and the internal energy per nucleon $\bar{E}(\rho, T, \delta = 0)$ in isospin-symmetric nuclear matter. The uncertainty bands correspond to calculations using two different sets of chiral low-momentum two-and three-body interactions, n3lo414 (solid lines) and n3lo450 (dash-dot lines). The spinodal region is colored in yellow. The critical point is shown as a yellow circle (full circle for n3lo414, open circle for n3lo450).}
\end{figure*}

From the free energy per nucleon the pressure and the entropy per nucleon follow via standard thermodynamic relations:

\[ P(T, \rho, \delta = 0) = \rho^2 \frac{\partial \bar{F}(T, \rho, \delta = 0)}{\partial \rho}, \quad \bar{S}(T, \rho, \delta = 0) = -\frac{\partial \bar{F}(T, \rho, \delta = 0)}{\partial T}. \]  

(5)

The internal energy per nucleon is given by $\bar{E} = \bar{F} + T\bar{S}$. The results for these quantities are shown in Fig. 1 for temperatures in the range $T = 0 - 25$ MeV. The spinodal region\(^1\) where the homogeneous system is unstable with respect to infinitesimal density fluctuations is shown explicitly. Note that for

\[ \text{In SNM the unstable spinodal region corresponds to } (\partial P/\partial \rho)_T \leq 0, \text{ with } (\partial P/\partial \rho)_T = 0 \text{ on the spinodal, cf. Sec. V B.} \]

\(^1\)
low temperatures the region of negative pressure extends into the metastable region (cf. also Fig. 13),
which is a generic property of liquids that are self-bound at low temperatures and a well-known feature
of superheated molecular liquids [44].

|         | $E_{\text{sat}}$ (MeV) | $\rho_{\text{sat}}$ (fm$^{-3}$) | $K$ (MeV) | $T_c$ (MeV) | $\rho_c$ (fm$^{-3}$) | $P_c$ (MeV fm$^{-3}$) |
|---------|---------------------|-------------------------------|---------|----------|-----------------|------------------|
| n3lo414 | -15.79              | 0.171                         | 223     | 17.4     | 0.066           | 0.33             |
| n3lo450 | -15.50              | 0.161                         | 244     | 17.2     | 0.064           | 0.32             |

Table I: Zero-temperature saturation energy $E_{\text{sat}}$, density $\rho_{\text{sat}}$ and incompressibility $K$, as well as
the critical temperature $T_c$, density $\rho_c$ and pressure $P_c$ of the liquid-gas phase transition in isospin-
symmetric nuclear matter from the sets of chiral two- and three-body nuclear interactions n3lo414 and n3lo450.

III. PURE NEUTRON MATTER

The free energy per particle in PNM, $\tilde{F}(T, \rho, \delta = 1)$, is obtained by restricting the isospin sum(s) in
Eq. (4) and in the different contributions in Eq. (3). Moreover, in PNM the three-body contributions
proportional to the low-energy constants $c_{E,D}$ and $c_4$ are absent [30]. The results for the free energy per
particle, the pressure, the entropy per particle and the internal energy per particle in PNM are shown
in Fig. 2. Note that the uncertainty bars obtained by varying the resolution scale (n3lo414 vs. n3lo450)
increase with temperature and are significantly reduced as compared to the SNM results in Ref. [12],
which is due to the decreased magnitude of nuclear interactions in PNM. At very low temperatures the
internal energy per particle increases monotonically with increasing density, but otherwise there is a local
minimum at finite density.

Figure 2: (Color online) Results for the free energy per particle $\tilde{F}(T, \rho, \delta = 1)$, the pressure $P(\rho, T, \delta = 1)$, the
entropy per particle $\tilde{S}(T, \rho, \delta = 1)$ and the internal energy per particle $\tilde{E}(\rho, T, \delta = 1)$ in pure neutron matter.
The solid lines show the results from n3lo414, the dash-dot lines the n3lo450 results. The green dashed lines
correspond to the model-independent virial equation of state (VEoS) determined from nucleon-nucleon scattering
phase shifts. The VEoS lines end where the fugacity is $z = 0.5$. 
At very low densities where the neutron-neutron scattering length $a_s \simeq -19 \text{ fm}$ is large compared to the interparticle separation, $k_F|a_s| \gg 1$ (with $k_F$ the Fermi momentum), a perturbative approach to neutron matter is not reliable. The model-independent virial equation of state (VEoS) computed by Horowitz and Schwenk in Ref. [45] from neutron-neutron scattering phase-shifts provides a benchmark for perturbative calculations of low-density neutron matter at nonzero temperature. In the virial expansion, the grand canonical expressions for the pressure and the density are expanded in powers of the fugacity $z = \exp(\mu/T)$, leading to

$$P(T, z, \delta = 1) = \frac{2T}{\lambda^3} \left(z + z^2 b_2(T) + \mathcal{O}(z^3)\right), \quad \rho(T, z, \delta = 1) = \frac{2}{\lambda^3} \left(z + 2z^2 b_2(T) + \mathcal{O}(z^3)\right), \quad (6)$$

where $\mu$ is the chemical potential, and $\lambda = \sqrt{(2\pi)/(MT)}$ is the neutron thermal wavelength. The second virial coefficient is given by

$$b_2(T) = \frac{1}{2^{1/2}\pi T} \int_0^{\infty} dE \exp\left[-E/(2T)\right] \delta_{\text{tot}}(E) - 2^{-5/2}, \quad (7)$$

where $\delta_{\text{tot}}(E)$ is the sum of the isospin-triplet elastic scattering phase shifts at laboratory energy $E$. From the pressure and density as functions of the fugacity the free energy per particle $\bar{F}$, entropy per particle $\bar{S}$, and internal energy per particle $\bar{E}$ follow again from standard thermodynamic relations (see Ref. [45] for details). The results for these quantities are shown as green dashed lines in Fig. 2. One sees that in the case of $\bar{F}$, $P$ and $\bar{S}$ there are almost no visible deviations between the VEoS and the perturbative results. This seemingly perfect agreement is however misleading, because the discrepancies corresponding to the different treatment of the interactions in the virial and the perturbative approach are overpowered by the large size of the (nonrelativistic) free Fermi gas contribution. The deviations are more transparent in the case of the internal energy per particle due to cancellations of the free Fermi gas terms in the free energy and entropy. The virial and perturbative results are closer at larger temperatures, since the EoS is less sensitive to the physics of large scattering lengths at higher momentum scales.

Figure 3: (Color online) Interaction contribution to the internal energy per particle, $\bar{E}_{\text{int}}(\rho, T, \delta = 1)$, in pure neutron matter at $T = 10 \text{ MeV}$ and low densities. The different lines correspond to the results from microscopic chiral nuclear interactions at first order (labeled “HF”) and second order in Eq. (3) as well as the virial expansion truncated at second order (VEoS) and with uncertainty bands obtained by estimating the third-order term. The n3lo414 and n3lo450 results are almost identical.

The differences between the virial and the perturbative results for the internal energy per particle are examined more closely in Fig. 3 for $T = 10 \text{ MeV}$. To depict the deviations more clearly we have

\footnote{Note that we have added the relativistic correction term to the VEoS lines. In particular, the zero-density limit of the internal energy per particle is given by $\bar{E}_V(T, \rho, \delta) \xrightarrow{\rho \to 0} \bar{E}_{\text{rel}}(T, \rho, \delta) |_{\rho \to 0} = 3T/2 + 15T^2/(8M)$. This agrees with the expansion in powers of $T$ of the internal energy per particle of a relativistic classical ideal gas, $\bar{E}_V = 3T + MK_1(M/T)/K_2(M/T)$ [where $K_1, K_2(M/T)$ are modified Bessel functions].}
subtracted the noninteracting contributions, i.e., the quantity shown is $E_{\text{int}} = \bar{E} - \bar{E}_0 - \bar{E}_{\text{rel}}$. The virial results include uncertainty bands obtained from estimating the neglected third virial coefficient as $|b_3(T)| \leq |b_2(T)|/2$. We also show the perturbative results at the Hartree-Fock level [first order in Eq. (3)]. One sees that whereas the Hartree-Fock results deviate visibly from the VEOS bands, the second-order results are much closer to those from the virial expansion and lie within the bands for densities $\rho \gtrsim 0.007$ fm$^{-3}$. The second-order calculation still underpredicts the attractive interaction contributions, in contrast to the pseudopotential approach based on nucleon-nucleon scattering phase shift data that was explored in Ref. [46]. We conclude that while the perturbative approach cannot fully capture the large scattering length physics of low-density neutron matter, the resulting errors are reasonably small when second-order contributions are included.

In recent years, the zero-temperature EoS of PNM from chiral nuclear interactions has been studied by numerous authors within widely different many-body frameworks [8,17,53]. We compare our results to those obtained from perturbative calculations with various chiral interactions by the Darmstadt group [8,17] in Fig. 3. In addition to the N2LO chiral three-nucleon forces, their calculations also include all N3LO three- and four-nucleon interactions. The uncertainty bands in their results were obtained by allowing large variations of the low-energy constants parameterizing the many-nucleon forces. One sees that the (almost overlapping) results from n3lo414 and n3lo450 lie within these bands. In Fig. 3 we also show results obtained from auxiliary-field quantum Monte Carlo simulations with chiral N3LO two-body (AFQMC [2N]) and N3LO two-body plus N2LO three-body forces (AFQMC [3N]) by Wiąziewski et al. [18]. The perturbative and the AFQMC results are similar at densities below 0.025 fm$^{-3}$, and at densities below 0.005 fm$^{-3}$ both are in agreement with the (fixed-node) quantum Monte Carlo calculations (based on the AV18 potential) of Gezerlis and Carlson [54]. However, the EoS predicted by the AFQMC calculations is significantly more repulsive at higher densities, in particular when three-body forces are included. This discrepancy may be (partly) related to systematic errors in the AFQMC treatment.

Figure 4: (Color online) Energy per particle in pure neutron matter at zero temperature, $\bar{E}(T = 0, \rho, \delta = 1)$, obtained from various many-body methods (see text for details). The inset magnifies the behavior at very low densities where quantum Monte Carlo simulations, labeled “QMC [AV18]”, are expected to be most accurate.

IV. SYMMETRY FREE ENERGY, ENTROPY AND INTERNAL ENERGY

From the results for the free energy per particle in SNM and PNM the symmetry free energy $\bar{F}_{\text{sym}}(T, \rho)$ is obtained via Eq. (1). The symmetry entropy and internal energy are related to the symmetry free energy via $\bar{S}_{\text{sym}} = -\partial \bar{F}_{\text{sym}}/\partial T$ and $\bar{E}_{\text{sym}} = \bar{F}_{\text{sym}} + T\bar{S}_{\text{sym}}$. The results for $\bar{F}_{\text{sym}}$, $T\bar{S}_{\text{sym}}$ and $\bar{E}_{\text{sym}}$ are shown as functions of density at different temperatures in the left column of Fig. 5. In the insets we show the noninteracting contribution to these quantities, i.e.,

$$\bar{F}_{\text{nonint,sym}}(T, \rho) = \bar{F}_0(T, \rho, 1) - \bar{F}_0(T, \rho, 0) + \bar{E}_{\text{rel}}(T, \rho, 1) - \bar{E}_{\text{rel}}(T, \rho, 0),$$

in the case of the symmetry free energy. In the right column of Fig. 5 we show $\bar{F}_{\text{sym}}(T, \rho)$, $T\bar{S}_{\text{sym}}(T, \rho)$, and $\bar{E}_{\text{sym}}(T, \rho)$ as functions of temperature at different densities.

One sees that in the considered range of densities and temperatures, $\bar{F}_{\text{sym}}$ is a monotonic increasing function of density and temperature. The density and temperature dependence of $T\bar{S}_{\text{sym}}$ is more involved.
At low densities $T \bar{S}_{\text{sym}}$ decreases monotonically with $T$, but for densities $\rho \gtrsim 0.2 \text{ fm}^{-3}$ a local minimum is found at $T \sim 10 \text{ MeV}$. The results from n3lo414 and n3lo450 are very similar for densities well below nuclear saturation density, but at higher densities the dependence on the resolution scale becomes significant. In particular, the decrease in the slope of $\bar{F}_{\text{sym}}$ with increasing density is more pronounced in the n3lo450 results. The $T$ dependence of $T S_{\text{sym}}(T, \rho)$ approximately balances that of $\bar{F}_{\text{sym}}(T, \rho)$, and as a result their sum, the symmetry internal energy $\bar{E}_{\text{sym}}$, increases with density but varies only very little with temperature. At densities near nuclear saturation density the deviations of $\bar{E}_{\text{sym}}(T, \rho \approx \rho_{\text{sat}})$ from its value at zero temperature are below 0.5 MeV.

Figure 5: (Color online) Left column: results for the symmetry free energy $\bar{F}_{\text{sym}}(T, \rho)$, the symmetry entropy times temperature $T S_{\text{sym}}(T, \rho)$, and the symmetry internal energy $E_{\text{sym}}(T, \rho)$, plotted as functions of density. The insets show the noninteracting (free Fermi gas) contribution to the different symmetry quantities. Right column: $\bar{F}_{\text{sym}}(T, \rho)$, $T S_{\text{sym}}(T, \rho)$, and $E_{\text{sym}}(T, \rho)$ as functions of temperature at different densities. The lines are interpolated, with calculated data points at $T/\text{MeV} = 0, 3, 5, 8, 10, 12, 15, 20, 25$. 

---

$T = 0 \text{ MeV}$ 
$T = 8 \text{ MeV}$ 
$T = 15 \text{ MeV}$ 
$T = 25 \text{ MeV}$ 
$n_{\text{3lo414}}$ 

$T = 0 \text{ MeV}$ 
$T = 8 \text{ MeV}$ 
$T = 15 \text{ MeV}$ 
$T = 25 \text{ MeV}$ 
$n_{\text{3lo450}}$ 

$0$ 
$5$ 
$10$ 
$15$ 
$20$ 
$25$ 
$0$ 
$5$ 
$10$ 
$15$ 
$20$ 
$25$ 

$E_{\text{sym}} \text{ [MeV]}$

$\rho \text{ [fm}^{-3}\text{]}$

$T \text{ [MeV]}$

$\rho = 0.025 \text{ fm}^{-3}$ 
$\rho = 0.05 \text{ fm}^{-3}$ 
$\rho = 0.085 \text{ fm}^{-3}$ 
$\rho = 0.12 \text{ fm}^{-3}$ 
$\rho = 0.17 \text{ fm}^{-3}$ 
$\rho = 0.22 \text{ fm}^{-3}$

$\bar{F}_{\text{sym}} \text{ [MeV]}$

$F_{\text{sym}} \text{ [MeV]}$

$T \text{ [MeV]}$

$\rho = 0.025 \text{ fm}^{-3}$ 
$\rho = 0.05 \text{ fm}^{-3}$ 
$\rho = 0.085 \text{ fm}^{-3}$ 
$\rho = 0.12 \text{ fm}^{-3}$ 
$\rho = 0.17 \text{ fm}^{-3}$ 
$\rho = 0.22 \text{ fm}^{-3}$
In Fig. 6 we show the symmetry quantities with the noninteracting contributions subtracted, i.e.,

\[ \bar{F}_{\text{int, sym}}(T, \rho) = \bar{F}_{\text{sym}}(T, \rho) - \bar{F}_{\text{nonint, sym}}(T, \rho), \]

(9)
as functions of temperature at different densities. In both cases the interacting contributions tend to counteract the temperature dependence of noninteracting contributions, cf. the insets in Fig. 5. In the case of \( \bar{F}_{\text{sym}} \) (and also \( T \bar{S}_{\text{sym}} \)) the noninteracting contributions dominate, but in the case of \( \bar{E}_{\text{sym}} \) the size of the noninteracting contributions and the ones from chiral nuclear interactions is more balanced, and the \( T \) dependence of both contributions approximately cancels each other, leading to the observed approximate temperature independence at densities near nuclear saturation density.

Figure 6: (Color online) Temperature dependence of the interacting contributions to the symmetry free energy, \( \bar{F}_{\text{sym, int}}(T, \rho) \), and the symmetry internal energy, \( \bar{E}_{\text{sym, int}}(T, \rho) \) at different densities. The lines are interpolated, with calculated data points at \( T/\text{MeV} = 0, 3, 5, 8, 10, 12, 15, 20, 25 \).

In Fig. 7 we compare our results for the symmetry (free) energy at zero temperature to the results obtained by Drischler et al. \[14\] from calculations of the EoS of neutron-rich matter using several renormalization group–evolved chiral nuclear interactions. For comparison we also show the results from microscopic calculations within a variational approach by Akmal et al. \[55\]. While the results of Drischler et al. are compatible with our results, the calculations by Akmal et al. predict a symmetry energy that is somewhat stiffer at densities \( \rho \gtrsim \rho_{\text{sat}} \). In Fig. 7 we also show recent empirical constraints obtained from the analysis of isobaric analog states and neutron skins (IAS+NS) \[56\]. One sees that the n3lo414 and n3lo450 results lie in the IAS+NS bands in the entire constrained density region \( 0.04 \lesssim \rho/\text{fm}^{-3} \lesssim 0.16 \).

Figure 7: (Color online) Symmetry (free) energy as a function of density at zero temperature, \( \bar{F}_{\text{sym}}(T = 0, \rho) \). The results from the chiral nuclear interactions n3lo414 and n3lo450 are compared to those of Drischler et al. \[14\] and Akmal et al. \[55\]. Also shown are empirical constraints from the analysis of isobaric analog states and neutron skins (IAS+NS).
For densities close to nuclear saturation density the symmetry (free) energy at zero temperature is usually expanded around \( J = \tilde{F}_{\text{sym}}(T = 0, \rho_{\text{sat}}) \) in terms of \( x = (\rho/\rho_{\text{sat}} - 1)/3 \):

\[
\tilde{F}_{\text{sym}}(T = 0, \rho) = J + Lx + \frac{1}{2} K_{\text{sym}} x^2 + O(x^3),
\]

where \( L = \partial \tilde{F}_{\text{sym}}(T = 0, \rho)/\partial \rho |_{\rho = \rho_{\text{sat}}} \) is called the slope parameter, and \( K_{\text{sym}} = \partial^2 \tilde{F}_{\text{sym}}(T = 0, \rho)/\partial \rho^2 |_{\rho = \rho_{\text{sat}}} \) the symmetry incompressibility. The zero-temperature saturation density in isospin-asymmetric nuclear matter is related to the parameters in the above expansion via \( \rho_{\text{sat}}(\delta) = \rho_{\text{sat}}[1 - 3L\delta^2/K] + O(\delta^4) \) (cf. Ref. [57]).\(^3\) The corresponding incompressibility \( K(\delta) \) can be expressed as

\[
K(\delta) = K + K_\rho \delta^2 + O(\delta^4), \quad K_\rho = K_{\text{sym}} - 6L,
\]

where \( K_\rho \) is usually called the isobaric incompressibility. In recent years, much effort has been invested in determining the parameters in Eqs. [10]. The empirical values of \( J = 29.0 - 32.7 \text{ MeV} \) and to a lesser degree also \( L = 40.5 - 61.9 \text{ MeV} \) are now relatively well constrained (values from [58], see also [8, 49, 59–61]), whereas experimental determinations of \( K_\rho \) suffer from large uncertainties. For instance, from measurements of neutron skin thicknesses [62] the value \( K_\rho = -500^{+125}_{-100} \text{ MeV} \) was obtained, which is compatible with the giant monopole resonance measured in Sn isotopes [63] giving \( K_\rho = -550 \pm 100 \text{ MeV} \). Theoretical studies using a selection of Skyrme interactions however led to an estimate of \( K_\rho = -370 \pm 120 \text{ MeV} \) [57].\(^4\) Our results for \( J, L \) and \( K_\rho \) are given in Table II. They are in agreement with the mentioned constraints.

| \( J \) (MeV) | \( L \) (MeV) | \( K_\rho \) (MeV) |
|---------------|---------------|-----------------|
| n3lo414       | 32.51         | -424            |
| n3lo450       | 31.20         | -434            |

Table II: Value of the (zero-temperature) symmetry energy at saturation density \( J \), the slope parameter \( L \), and the isobaric incompressibility \( K_\rho \), extracted from the results obtained from the sets of chiral nuclear two and three-body interactions n3lo414 and n3lo450.

V. THERMODYNAMICS OF ISOSPIN-ASYMMETRIC NUCLEAR MATTER

In this section we examine the thermodynamic equation of state of isospin-asymmetric nuclear matter (ANM). The free energy per nucleon in ANM is calculated as follows. The dependence of the nonrelativistic free Fermi gas contributions on the isospin asymmetry \( \delta \) is treated exactly, while the relativistic correction term and the interaction contributions are assumed to have quadratic dependence on isospin asymmetry:

\[
\tilde{F}(T, \rho, \delta) \approx \tilde{F}_0(T, \rho, \delta) + \tilde{F}_{\text{rel}}(T, \rho, 0) + \tilde{F}_{\text{sym,rel}}(T, \rho) \delta^2 + \tilde{F}_{\text{int}}(T, \rho, 0) + \tilde{F}_{\text{sym, int}}(T, \rho) \delta^2.
\]

This approach is motivated in Sec. VA where we investigate in detail the dependence on isospin asymmetry of the noninteracting contributions \( \tilde{F}_0(T, \rho, \delta) \) and \( \tilde{F}_{\text{rel}}(T, \rho, \delta) \).

In Sec. VB we then discuss the construction of the spinodal in ANM and present our results for the critical temperature. The dependence of the neutron and proton chemical potentials on isospin asymmetry is examined in Sec. VC and we determine the neutron drip point in cold nuclear matter. Finally, in Sec. VD we show results for the free energy per nucleon and pressure in isospin-asymmetric nuclear matter and determine the values of \( T \) and \( \delta \) where an isolated drop of liquid nuclear matter becomes unstable.

\(^3\) The densities \( \rho_{\text{sat}}(\delta) \) correspond to stable self-bound states only for isospin asymmetries up to the neutron drip point, \( \delta \leq \delta_{\text{dp}}, \) cf. Secs. VC and VD.

\(^4\) However, in each case a slightly different definition of \( K_\rho \) is used, with the differences corresponding to higher-order terms in Eq. [11] and finite-size effects [57].
A. Isospin dependence of free nucleon gas

Here we examine the $\delta$ dependence of the noninteracting contributions to the free energy per nucleon.

We compute the four leading terms in an expansion of $\bar{F}_0(T, \rho, \delta)$ and $\bar{F}_{rel}(T, \rho, \delta)$ in powers of $\delta^2$. The results show that the accuracy of the quadratic approximation for $\bar{F}_0(T, \rho, \delta)$ decreases significantly with increasing temperature, which necessitates the exact calculation of this contribution. The relativistic correction term on the other hand can be safely approximated via $\bar{F}_{rel}(T, \rho, \delta) \approx \bar{F}_{rel}(T, \rho, 0) + \bar{F}_{sym,rel}(T, \rho) \delta^2$.

Assuming analyticity, the free energy per nucleon as a function of temperature $T$, nucleon density $\rho$ and isospin asymmetry $\delta$ can be expanded in powers of $\delta^2$ around the free energy per nucleon in SNM:

$$\bar{F}(T, \rho, \delta) = \bar{F}(T, \rho, 0) + \sum_{n=1}^{\infty} B_{2n}(T, \rho) \delta^{2n}. \quad (13)$$

The various expansion coefficients $B_{2n}$ are given by

$$\bar{B}_{2n}(T, \rho) = \frac{1}{(2n)!} \left. \frac{\partial^{2n} \bar{F}(T, \rho, \delta)}{\partial \delta^{2n}} \right|_{\delta=0}. \quad (14)$$

Setting $\delta = 1$ in Eq. $\bar{F}_{sym}(T, \rho)$ we obtain the symmetry free energy as the sum of the above coefficients, i.e.,

$$\bar{F}_{sym}(T, \rho) = \sum_{n=1}^{\infty} \bar{B}_{2n}(T, \rho) \bar{F}_{sym}(T, \rho) \sum_{n=1}^{\infty} \beta_{2n}(T, \rho), \quad \text{where } \beta_{2n}(T, \rho) = \frac{\bar{B}_{2n}(T, \rho)}{\bar{F}_{sym}(T, \rho)}. \quad (15)$$

Here, we have introduced the weight factors $\beta_{2n}(T, \rho)$ as a means to specify the relative size of the different expansion coefficients.

The noninteracting contributions to the free energy density, $F_0 = F_0^{n/p} + F_0^{p/n}$ and $F_{rel} = F_{rel}^{n/p} + F_{rel}^{p/n}$, can be expressed in terms of polylogarithmic functions $\text{Li}_\nu(x) = \sum_{k=1}^{\infty} x^k k^{-\nu}$, i.e.,

$$F_0^{n/p}(T, \mu_0^{n/p}) = -\alpha T^{5/2} \left( \ln(x_{n/p}) \text{Li}_{3/2}(x_{n/p}) - \text{Li}_{5/2}(x_{n/p}) \right), \quad (16)$$

$$F_{rel}^{n/p}(T, \mu_0^{n/p}) = \frac{150 T^{7/2}}{8M} \text{Li}_{7/2}(x_{n/p}) \quad (17)$$

where $\mu_0^{n/p}$ are the neutron and proton effective one-body chemical potentials, $x_{n/p} = -\exp(\mu_0^{n/p}/T)$ and $\alpha = 2^{-1/2}(M/\pi)^{3/2}$. For given values of $T$, $\rho$ and $\delta$ the effective one-body chemical potentials $\mu_0^{n/p}$ are uniquely determined by $\rho_{n/p} = -\alpha T^{3/2} \text{Li}_{3/2}(x_{n/p})$.

The $\delta$ derivative of order $n$ of $F_i^{n/p}$, $i \in \{0, \text{rel}\}$, at fixed density and temperature is given by

$$\left( \frac{\partial^n F_i^{n/p}}{\partial \delta^n} \right)_{T, \rho} = \frac{(\pm 1)^n \alpha T^{5/2}(n-1)!}{(1 + \delta)^n} \mathcal{Y}_i^{(n)}(x_{n/p}). \quad (18)$$

where the functions $\mathcal{Y}_i^{(n)}$ are defined recursively as

$$\mathcal{Y}_i^{(n)}(x_{n/p}) = \frac{x_{n/p}}{n-1} \frac{\text{Li}_{3/2}(x_{n/p})}{\text{Li}_{1/2}(x_{n/p})} \frac{\partial}{\partial x_{n/p}} \mathcal{Y}_i^{(n-1)}(x_{n/p}) - (1 - \delta_{1,n}) \mathcal{Y}_i^{(n-1)}(x_{n/p}), \quad n \geq 1, \quad (19)$$

with $\delta_{k,l}$ the Kronecker delta. The expressions to start the recursion are

$$\mathcal{Y}_0^{(0)}(x_{n/p}) = \ln(-x_{n/p}) \text{Li}_{3/2}(x_{n/p}) - \text{Li}_{5/2}(x_{n/p}), \quad \mathcal{Y}_\text{rel}^{(0)}(x_{n/p}) = -\frac{150 T}{8M} \text{Li}_{7/2}(x_{n/p}). \quad (20)$$

---

5 As noted in the introduction, in initial calculations we have found that at the Hartree-Fock level the contribution from two-nucleon interactions is quadratic in $\delta$ to very high accuracy. We also examined the exact isospin asymmetry dependence of the second-order contribution from two-nucleon interactions, and three-nucleon contributions at the Hartree-Fock level; in both cases we have found that higher-order terms in $\delta$ are more sizeable, but still significantly small compared to those that emerge from $F_0(T, \rho, \delta)$. Future research will quantify the isospin asymmetry dependence of the interaction contributions in more detail.

6 Odd-order terms in $\delta$ arise only from isospin-symmetry breaking effects, which we neglect here.
One then obtains for $\bar{B}_{\text{nonint},2n}$ the expression

$$\bar{B}_{\text{nonint},2n}(T, x) = \frac{T}{2n \text{Li}_{3/2}(x)} \left( \Phi_0^{(2n)}(x) + \Phi_{\text{rel}}^{(2n)}(x) \right),$$

(21)

where $x = -\exp(\mu_0/T)$, with $\mu_0$ the nucleon effective one-body chemical potential, which is uniquely determined by $\rho = -2\alpha T^{3/2}\text{Li}_{3/2}(x)$.

The results for the first weight factor $\beta_{\text{nonint},2} = \bar{B}_{\text{nonint},2}/\bar{F}_{\text{nonint,sym}}$ and the ratios $\beta_{\text{nonint},2n}/\beta_{\text{nonint},2(n+1)}$ for $n = 1, 2, 3$ are displayed in Fig. 8. Note that the limits $\rho \to 0$ and $T \to 0$ do not commute. The $\rho \to 0$ limit of the symmetry coefficients at finite temperature is given by

$$\bar{B}_{\text{nonint},2n}(T \neq 0, \rho \to 0) = \frac{T}{(2n)!} \left( \frac{\partial^{2n}}{\partial \delta^{2n}} \left( (1 + \delta) \ln(1 + \delta) + (1 - \delta) \ln(1 - \delta) \right) \right)_{\delta = 0} = \frac{T}{n(2n - 1)},$$

(22)

which comes entirely from the logarithmic terms, $\sim \ln(-x_{n/p}) = \mu_0^{n/p}/T$, in the expression for $\bar{F}_0 = (F_0^n + F_0^n)/(\rho_0 + \rho_p)$. The $\rho \to 0$ and $T \to \infty$ limits of the weight factors $\beta_{\text{nonint},2n} = \bar{B}_{\text{nonint},2n}/\bar{F}_{\text{nonint,sym}}$ coincide, causing the convergence rate of the quadratic expansion of $\bar{F}_{\text{nonint}}$ to decrease significantly with temperature. This is entirely caused by the presence of the logarithmic terms in the nonrelativistic contribution, i.e., by the first term proportional to the sum of the effective one-body chemical potentials, $\sim \mu_0^n + \mu_0^n$. The $\rho \to 0$ limit of the second term in $\bar{F}_0$ proportional to $[\text{Li}_{5/2}(x_n) + \text{Li}_{5/2}(x_p)]/[\text{Li}_{3/2}(x_n) + \text{Li}_{3/2}(x_p)]$ equals $-T$, which is independent of $\delta$; the convergence rate of the expansion in powers of $\delta^2$ of this term alone increases very strongly with increasing temperature. Similarly, the $\rho \to 0$ limit of $\bar{F}_{\text{rel}}$ equals $-15T^{5/2}/(8M)$, and the quadratic approximation of the relativistic correction term becomes increasingly accurate with increasing temperature.

![Figure 8](image_url)

Figure 8: (Color online) Temperature dependence of the first weight factor $\beta_{\text{nonint},2}$ and the ratios $\beta_{\text{nonint},2n}/\beta_{\text{nonint},2(n+1)}$ for $n = 1, 2, 3$ at different densities, corresponding to the expansion of $\bar{F}_{\text{nonint}}(T, \rho, \delta)$ in powers of $\delta^2$. The full lines show the results with the relativistic correction term included, the dotted lines the nonrelativistic results.
B. Spinodal and critical temperature

Thermodynamic stability is associated with a number of equivalent stability criteria \[14, 64, 65\], which are derived from the fundamental principle of maximum entropy \[66\]. In the canonical ensemble the corresponding requirement is that the free energy density at fixed temperature \(T\) is a convex function of the component densities \(\rho_1\) and \(\rho_2\), implying that the Hessian matrix \(\mathcal{F}_{ij}\) has no negative eigenvalues. In the unstable region delineated by the spinodal the free energy density is concave; in the metastable region between the spinodal and binodal \(F(T, \rho_1, \rho_2)\) is locally convex and the system is protected against phase separation by a nucleation barrier. The Hessian matrix \(\mathcal{F}_{ij}\) is given by

\[
\mathcal{F}_{ij}(T, \rho_1, \rho_2) = \left[ \frac{\partial^2 F(T, \rho_1, \rho_2)}{\partial \rho_i \partial \rho_j} \right] = \left[ \frac{\partial \mu_i(T, \rho_1, \rho_2)}{\partial \rho_j} \right], \quad i, j \in \{1, 2\},
\]

Its eigenvalues are given by

\[
\xi_\pm(T, \rho_1, \rho_2) = \frac{1}{2} \left[ \text{tr}[\mathcal{F}_{ij}] \pm (\text{tr}[\mathcal{F}_{ij}]^2 - 4 \det[\mathcal{F}_{ij}])^{1/2} \right]
\]

\[
= \frac{1}{2} \left[ \mathcal{F}_{11} + \mathcal{F}_{22} \pm ((\mathcal{F}_{11} - \mathcal{F}_{22})^2 + 4\mathcal{F}_{12}^2)^{1/2} \right].
\]

The eigenvalues are invariant under (linear) basis transformations. From the data \(F(T, \rho, \delta)\) they are readily evaluated using as independent density parameters \(\rho_1 = \rho_n + \rho_p = \rho\) (nucleon density) and \(\rho_2 = \rho_n - \rho_p = \rho \delta\) (isospin asymmetry density). In this basis the Hessian matrix becomes diagonal at \(\delta = 0\) with eigenvalues \(\xi_\pm = \partial^2 F / \partial \rho^2_\delta > 0\) and \(\xi_- = \partial^2 F / \partial \rho_1^2\), therefore in SNM the region inside the spinodal corresponds to a negative isothermal compressibility \(\kappa_T = \rho^{-1}(\partial \rho / \partial P)_{T, \delta}\), where \(\kappa_T^{-1} > 0\) is a stability criterion for a pure substance.

The exact expressions for the nonrelativistic free Fermi gas contribution to the Hessian matrix components are given by\footnote{Note that \(\mathcal{F}_{0,11} = \mathcal{F}_{0,22}\), but \(\mathcal{F}_{\text{int},11} \neq \mathcal{F}_{\text{int},22}\).}

\[
\mathcal{F}_{0,11}(T, \rho_0^n, \mu_0^n) = -\frac{T^{-1/2}}{4\alpha} \left( \frac{1}{\text{Li}_1(x_n)} + \frac{1}{\text{Li}_1(x_p)} \right) = \mathcal{F}_{0,22}(T, \mu_0^n, \mu_0^n),
\]

\[
\mathcal{F}_{0,12}(T, \rho_0^n, \mu_0^n) = -\frac{T^{-1/2}}{4\alpha} \left( \frac{1}{\text{Li}_1(x_n)} - \frac{1}{\text{Li}_1(x_p)} \right),
\]

where again \(x_n/p = -\exp(\mu_0^n/p / T)\) and \(\alpha = 2^{-1/2}(M/\pi)^{3/2}\). In the limit \(\delta \to 1\) the proton density vanishes, \(\rho_p \to 0\), and the proton effective one-body chemical potential diverges (at finite \(T\)), \(\mu_0^p \to -\infty\), thus \(\text{Li}_1(x_p) \to 0\). Hence, the exact calculation of the nonrelativistic free Fermi gas contribution \(F_0(T, \rho, \delta)\) leads to divergent behavior of the Hessian components in the limit of vanishing proton concentration. The same divergent behavior is obtained in the exact calculation of \(\mathcal{F}_{0,ij}\) at zero temperature. The instability region then vanishes at a value \(\delta < 1\) for all values of \(T\). This constraint is lost if the free Fermi gas contribution is approximated by truncating the expansion in powers of \(\delta^2\) of \(F_0(T, \rho, \delta)\) at a finite order, e.g., at first order as in Eq. \[1\].

The evolution of the critical temperature \(T_c(\delta)\) where (for a given value of \(\delta\)) the concave region vanishes is depicted in Fig. \[9\]. The results from \(n3lo414\) and \(n3lo450\) are very similar; in both cases the critical lines end approximately at an isospin asymmetry \(\delta_{\text{end}} \simeq 0.9996\) or a proton concentration \(y_{p,\text{end}} \simeq 2 \cdot 10^{-4}\). A comparably large critical line endpoint \(\delta_{\text{end}}\) has been obtained also in Ref. \[16\] in calculations with effective Skyrme forces. We note that in ANM the coexistence region does not vanish at the critical temperature \(T_c(\delta)\) but at a higher temperature \(T_{\text{max}}(\delta)\). Although for temperatures in the range \(T_c(\delta) < T < T_{\text{max}}(\delta)\) there is no first-order phase transition from a dense liquidlike to a dilute gaslike phase, for \(\delta \neq 0\) the system can still undergo a retrograde condensation \[15, 17\].
C. Neutron drip point

From the data $F(T, \rho, \delta)$ the neutron and proton chemical potentials are obtained via

$$\mu_{n/p}(T, \rho, \delta) = \frac{\partial F(T, \rho, \delta)}{\partial \rho} \pm \frac{1 \mp \delta}{\rho} \frac{\partial F(T, \rho, \delta)}{\partial \delta}. \quad (27)$$

The results for $\mu_n(T, \rho, \delta)$ and $\mu_p(T, \rho, \delta)$ are displayed in Fig. 10 for temperatures $T/\text{MeV} = 0, 15$. One sees that $\mu_n(T, \rho, \delta)$ increases and $\mu_p(T, \rho, \delta)$ decreases with $\delta$, as is obvious from the positivity of the symmetry free energy and the different signs in Eq. (27). The chemical potentials at finite $T$ diverge as $\rho \to 0$, but at zero temperature $\mu_{n/p} \to 0$ for $\rho \to 0$. The origin of this feature is the collapse of Fermi-Dirac distribution functions into Heaviside step functions at $T = 0$, which eliminates the divergence of the free energy per nucleon at vanishing density [12, 16].

The Gibbs conditions for the coexistence of the two phases I (liquid) and II (gas) in mutual thermodynamic equilibrium are

$$T^I = T^{II}, \quad P^I = P^{II}, \quad \mu_n^I = \mu_n^{II}, \quad \mu_p^I = \mu_p^{II}. \quad (28)$$

Whereas at finite $T$ liquid-gas equilibrium corresponds to finite values of density and isospin asymmetry in both phases, where for $\delta \neq 0$ the gas is more neutron-rich and the liquid is more proton-rich relative to the global value of $\delta$ [13][18], the vanishing of $\mu_{n/p}(T = 0, \rho, \delta)$ at vanishing density entails that at $T = 0$ the Gibbs conditions for the neutron and proton chemical potentials can in most cases not be satisfied, leading to a gas phase that is either empty (vacuum) or contains only neutrons [10]. The neutron drip point, $\delta_{ND}$, is given by the value of $\delta$ where the neutron chemical potential at vanishing temperature and pressure becomes positive. For isospin asymmetries $\delta \leq \delta_{ND}$ an isolated drop of cold liquid nuclear matter is stable (in equilibrium with the vacuum), defining a stable self-bound state. As seen from Fig. 10, in our equation of state neutron drip occurs at an isospin asymmetry $\delta_{ND} \simeq 0.30$ or a proton concentration $Y_{p,ND} = (1 - \delta_{ND})/2 \simeq 0.35$.  

Figure 9: Trajectories of the critical temperature $T_c(\delta)$ determined from n3lo450 and n3lo414. The trajectories end at $\delta \simeq 0.9996$. The calculated data points are shown explicitly. The results from n3lo414 and n3lo450 almost overlap.
Figure 10: (Color online) Neutron and proton chemical potentials, $\mu_n(T, \rho, \delta)$ and $\mu_p(T, \rho, \delta)$, in isospin-asymmetric nuclear matter at temperatures $T/\text{MeV} = 0, 15$, calculated using n3lo414 (solid lines) and n3lo450 (dash-dot lines). Stable self-bound states are shown as gold-colored lines with circles (full circles for n3lo414, open circles for n3lo450); the lines end at the neutron drip point $\delta_{ND} \approx 0.30$.

D. Metastable self-bound liquid

The zero-temperature results for the (ground state) energy per nucleon $\tilde{E} = \tilde{F}$ and the pressure $P$ are displayed in Fig. 11 as functions of the nucleon density $\rho$ for different values of $\delta$. The trajectory of the saturation points\(^8\) where the energy per nucleon has a local minimum and the pressure is zero is shown explicitly. These points correspond to the properties of a drop of cold liquid nuclear matter surrounded by vacuum. For $\delta \leq \delta_{ND} \approx 0.30$ the cold drop is stable (the saturation point lies on the binodal, cf. Sec. V C), and for $\delta_{ND} < \delta < \delta_{FP}$ the saturation point lies in the metastable region between the binodal and the spinodal. We refer to the point $\delta_{FP} \approx 0.66$ where the saturation point trajectory encounters the spinodal as the fragmentation point (FP). The energy per nucleon at neutron drip and at the fragmentation point is $E_{ND} \approx -13.0 \text{MeV}$ and $E_{FP} \approx -3.1 \text{MeV}$, respectively. For comparison we follow the local energy minima also into the unstable spinodal region; i.e., we show also the point where both derivatives of the analytic energy per nucleon vanish (saddle point, SP) at $\delta_{SP} \approx 0.80$ (0.81 for n3lo450) as well as the local energy minimum at $\delta \approx 0.76$. For $\delta > 0.76$ the analytic energy per nucleon is positive at all (finite) densities, and for $\delta > \delta_{SP}$ the pressure is a semipositive definite function of density.

\(^8\) In thermodynamics saturation refers to the properties of the two phases on the binodal. In this work we use saturation point to denote a stable or metastable self-bound state.
Figure 11: (Color online) Energy per nucleon $E = F$ and pressure $P$ in isospin-asymmetric nuclear matter at zero temperature, calculated using n3lo414 (solid lines) and n3lo450 (dash-dot lines). The saturation point trajectories are shown as gray-blue (gold-colored below neutron drip) lines with circles (full circles for n3lo414, open circles for n3lo450). The trajectories end at the fragmentation point $\delta_{FP} \approx 0.66$ (red circle). The inset magnifies the behavior of the pressure at low densities.

Figure 12: (Color online) Free energy per particle $\tilde{F}(T, \rho, \delta)$ and pressure $P(T, \rho, \delta)$ at temperatures $T = 5, 15$ MeV. The solid lines show the n3lo414 results, the dash-dot lines the n3lo450 results. The gray-blue lines with circles depict the trajectories of the saturation points, up to the point where they encounter the spinodal (red circle). The insets magnify the behavior of the pressure at low densities.
The finite-temperature results for the free energy per nucleon $\bar{F}(T, \rho, \delta)$ and the pressure $P(T, \rho, \delta)$ are shown in Fig. [12] for temperatures $T/\text{MeV} = 5, 15$. At $T = 5 \text{ MeV}$ the saturation point trajectory ends at $\delta_{FP} \simeq 0.61$, defining the fragmentation temperature for nuclear matter with proton concentration $Y_p \simeq 0.195$. At finite $T$ the saturation point trajectories lie entirely in the metastable region; an isolated drop of hot liquid nuclear matter has to be stabilized by a surrounding nucleon gas. For temperatures $T \gtrsim 13.5 \text{ MeV}$ the analytic free energy per nucleon is a monotonic increasing function of density for all values of $\delta$.

The relation between the spinodal, the binodal, and the saturation point trajectories is illustrated in Fig. 13. The two plots in Fig. 13 represent isochemical ($\delta = \text{const}$) and isothermal cross sections of the respective surfaces (spinodal, binodal, saturation point manifold) in $(T, \rho, \delta)$ space (cf. Refs. [16, 17]). In the second plot we also show the surface with divergent isothermal compressibility $\kappa_T = \rho^{-1}(\partial \rho/\partial P)_{T, \delta}$, which corresponds to the violation of the stability criterion $\kappa_T^{-1} > 0$ for a one-component system. Hence, the saddle point (SP) where both derivatives of the analytic free energy per nucleon with respect to the nucleon density vanish coincides with the fragmentation point (FP) where a liquid drop becomes unstable only for $\delta = 0$ where nuclear matter behaves like a pure substance. For $\delta \neq 0$ the more restrictive two-component stability criteria are needed [64] ($\kappa_T^{-1} > 0$ is not a relevant stability criterion in that case) and the SP is located in the interior of the spinodal.

The trajectory of the fragmentation temperatures $T_{FP}(\delta)$ is shown in Fig. 14 [for comparison we also show the saddle point temperatures $T_{SP}(\delta)$]. Liquid nuclear matter with temperature $T_{FP}(\delta) \leq T < T_c(\delta)$ can exist only in coexistence with a nucleon gas, or if an external pressure is applied.

![Figure 13](image1.png)

**Figure 13:** (Color online) Left plot: binodal, spinodal, and saturation point trajectory (“self-bound liquid”) in SNM ($\delta = 0$). Right plot: $T = 10 \text{ MeV}$ cross sections of the spinodal, the $\kappa_T = \infty$ boundary, and the saturation point manifold (“self-bound liquid”); only the $\delta = 0$ endpoints of the binodal are shown (we did not construct the binodal for $\delta \neq 0$). The critical points (CP), fragmentation points (FP) and saddle points (SP) are shown explicitly in both plots (in SNM the FP and the SP coincide).

![Figure 14](image2.png)

**Figure 14:** (Color online) Trajectory of the fragmentation temperature $T_{FP}(\delta)$ above which no metastable self-bound state can exist (red line). For comparison we also show the trajectory of the points where the analytic free energy per nucleon has a saddle point, $T_{SP}(\delta)$ (blue line). The calculated data points are shown explicitly.
VI. SUMMARY

In this work, we have investigated in detail the temperature and density dependence of the symmetry free energy \( F_{\text{sym}}(T, \rho) = F(T, \rho, \delta = 1) - F(T, \rho, \delta = 0) \) in homogeneous nuclear matter using chiral effective field theory interactions constructed at resolution scales \( \Lambda = 414, \text{450 MeV} \). The free energy per nucleon of isospin-symmetric nuclear matter, \( F(T, \rho, \delta = 0) \), and pure neutron matter, \( F(T, \rho, \delta = 1) \), have been calculated within the framework of many-body perturbation theory. Empirical constraints from the nuclear saturation point, the critical point of the liquid-gas phase transition, and the density dependence of \( F_{\text{sym}} \) at zero temperature are reproduced, and our results are consistent with the virial expansion and with perturbative calculations using chiral nuclear interactions by the Darmstadt group.

The four leading coefficients in an expansion of the noninteracting contributions \( F_0 \) and \( F_{\text{rel}} \) in terms of \( \delta^2 \) have been examined, and we have found that the convergence rate of the expansion of \( F_0 \) decreases significantly with temperature. Therefore we have used the exact expressions for \( F_0 \) in computing the free energy per nucleon \( F(T, \rho, \delta) \) in isospin-asymmetric nuclear matter (ANM). The many-body contributions from nuclear interactions on the other hand have been assumed to have a quadratic dependence on the isospin asymmetry \( \delta \).

From the results for \( F(T, \rho, \delta) \) we have computed the pressure \( P(T, \rho, \delta) \) and the neutron and proton chemical potentials \( \mu_{n/p}(T, \rho, \delta) \), and we have constructed the trajectory of the critical temperature \( T_c(\delta) \). The critical line ends at a proton fraction \( Y_{p,\text{end}} \approx 2 \cdot 10^{-4} \). The neutron drip point in infinite nuclear matter at zero temperature has been located at \( Y_{p,\text{ND}} \approx 0.35 \). Furthermore, we have determined the trajectory of the fragmentation temperature \( T_{\text{FP}}(\delta) \) above which no metastable self-bound state exists.

Future work will be aimed at improving the description of the isospin asymmetry dependence of the interaction contributions to the thermodynamic equation of state, a more detailed description of matter at low densities, and the extrapolation to higher temperatures and densities required for simulations of core-collapse supernovae and binary neutron star mergers.

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