

Characterization of Phase Transition in Heisenberg Fluids from Density Functional Theory*

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Abstract The phase transition of Heisenberg fluid has been investigated with the density functional theory in mean-field approximation (MF). The matrix of the second derivatives of the grand canonical potential Ω with respect to the particle density fluctuations and the magnetization fluctuations has been investigated and diagonalized. The smallest eigenvalue being 0 signals the phase instability and the related eigenvector characterizes this phase transition. We find a Curie line where the order parameter is pure magnetization and a spinodal where the order parameter is a mixture of particle density and magnetization. Along the spinodal, the character of phase instability changes continuously from predominant condensation to predominant ferromagnetic phase transition with the decrease of total density. The spinodal meets the Curie line at the critical endpoint with the reduced density $\rho^* = \rho\sigma^3 = 0.224$ and the reduced temperature $T^* = kT/\epsilon = 1.87$ (σ is the diameter of Heisenberg hard sphere and ϵ is the coupling constant).

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1 Introduction

In the last decade there were many investigations on spin fluids with coupled spin and spatial interactions.^[1,2] The spin fluid could be taken as a simplified model of real system.^[3] Due to the additional spin degrees of freedom and the coupling between them and spatial coordinates, the spin fluids can have a rich phase behavior. Besides gas-liquid (G-L),^[4] such a spin fluid model displays liquid-liquid (L-L),^[5] liquid-solid (L-S),^[6] and paramagnetic-ferromagnetic (P-F) phase transitions, critical end, and tri-critical point behavior. Many models have been proposed such as the discrete Ising,^[7] continuous XY,^[8] and Heisenberg fluid. The more microscopically motivated studies, so far, try to comprehend the picture of phase behavior of these fluids by employing density functional theory (DFT),^[9,10] mean-field theory (MFT),^[11–14] integral equation (IE),^[15–17] as well as Monte Carlo (MC) simulations.^[2,16,18,19]

The recent interest has been stimulated partly by MC simulations (Mryglod *et al.*^[2] and Nijmeijer *et al.*^[18]) of Heisenberg fluid, which strongly hint that the ferromagnetic transition of spin fluid challenge the traditional viewpoint to ferromagnetic transition theory. Tavares and co-workers using both MF and a more refined modified mean field (MMF) density functional theory have found in some regime mixed first-order transition, namely a condensation-ordering transition. Both theoretical works and MC results showed that the first-order transition (i.e., an isotropic vapor phase and a ferromagnetic liquid phase)

are the mixed transition of the ordering fluctuations and density fluctuations.

However, as we shall demonstrate here, knowledge of the complete phase diagram is essential but not enough. In order to understand the existence of the mixed transition, we have to classify the phase transition characters. So the purpose of the present work is to characterize the phase transition along condensation-ordering transition line. Therefore, we investigate this system using the method proposed by Chen *et al.*^[20,21] to characterize phase transition. We address these questions using density functional theory in the so-called simple mean-field theory. As a result, the phase transitions of different character take place in this system, as studies of the phase diagram and of the associated critical phenomena have been shown.

2 Model

We consider a Heisenberg hard-sphere (HHS) fluid with diameter σ and pair potential,^[9]

$$u(\mathbf{r}_1, \omega_1, \mathbf{r}_2, \omega_2) = u^{\text{hs}}(r) + u^{\text{ss}}(r, \omega_1, \omega_2), \quad (1)$$

where $r = |\mathbf{r}_1 - \mathbf{r}_2|$ is the distance between particles at positions \mathbf{r}_1 and \mathbf{r}_2 and $\omega_i = (\theta_i, \phi_i)$ are the orientations of particles with $0 \leq \theta_i \leq \pi$ and $0 \leq \phi_i \leq 2\pi$. The repulsive hard sphere interaction is given by

$$u^{\text{hs}}(r) = \begin{cases} +\infty, & r \leq \sigma, \\ 0, & r > \sigma. \end{cases} \quad (2a)$$

$$(2b)$$

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The spin-spin interaction is given by

$$u^{ss}(r, \omega_1, \omega_2) = \begin{cases} 0, & r \leq \sigma, \\ -J(r) \mathbf{s}_1 \cdot \mathbf{s}_2, & \sigma < r \leq r_c, \\ 0, & r > r_c, \end{cases} \quad (3a)$$

$$-J(r) \mathbf{s}_1 \cdot \mathbf{s}_2, \quad \sigma < r \leq r_c, \quad (3b)$$

$$0, \quad r > r_c, \quad (3c)$$

where

$$J(r) = \varepsilon \frac{e^{-z(r/\sigma-1)}}{r/\sigma}, \quad (4)$$

and \mathbf{s}_i is a unit vector ($|\mathbf{s}_i| = 1$) in the direction of spin with

$$\begin{aligned} \mathbf{s}_1 \cdot \mathbf{s}_2 &= \cos(\omega_1, \omega_2) = \cos \theta_1 \cos \theta_2 \\ &+ \sin \theta_1 \sin \theta_2 \cos(\phi_1 - \phi_2). \end{aligned} \quad (5)$$

In our paper the cutoff distance of the Yukawa potential $r_c = \infty$ and the dimensionless parameter $z = 1$. To have ferromagnetic phase the coupling constant ε here is assumed to be positive.

3 Density Functional and Mean-Field Approximation

The grand canonical free energy Ω of a nonuniform spin liquid is the minimum of the functional^[22,23]

$$\begin{aligned} \Omega[\rho(\mathbf{r}, \omega), T, \mu] &= F[\rho(\mathbf{r}, \omega)] \\ &+ \int d\omega d\mathbf{r} \rho(\mathbf{r}, \omega) (V_{\text{ext}}(\mathbf{r}, \omega) - \mu), \end{aligned} \quad (6)$$

where μ is the chemical potential and the Helmholtz free energy $F[\rho(\mathbf{r}, \omega)]$ is a unique functional of the density $\rho(\mathbf{r}, \omega)$ in the presence of the external potential $V_{\text{ext}}(\mathbf{r}, \omega)$. The Helmholtz free energy can be written as^[24]

$$\begin{aligned} F[\rho(\mathbf{r}, \omega)] &= F^{\text{hs}}[\rho(\mathbf{r}, \omega)] \\ &+ \frac{1}{2} \int_0^1 d\lambda \int d\mathbf{r}_1 d\omega_1 d\mathbf{r}_2 d\omega_2 \rho(\mathbf{r}_1, \omega_1) \\ &\times \rho(\mathbf{r}_2, \omega_2) g(\mathbf{r}_1, \omega_1, \mathbf{r}_2, \omega_2; \lambda) u^{\text{ss}}(r, \omega_1, \omega_2), \end{aligned} \quad (7)$$

where F^{hs} is the Helmholtz free energy of the hard-sphere system and $g(\mathbf{r}_1, \omega_1, \mathbf{r}_2, \omega_2; \lambda)$ is the pair distribution function in a system in which the particles interact via a pairwise potential

$$u_\lambda(r, \omega_1, \omega_2) = u^{\text{hs}}(r) + \lambda u^{\text{ss}}(r, \omega_1, \omega_2). \quad (8)$$

The total number density of particles without specified orientation is given by

$$\rho(\mathbf{r}) = \int d\omega \rho(\mathbf{r}, \omega) = \int_0^{2\pi} d\phi \int_0^\pi d\theta \sin \theta \rho(\mathbf{r}, \theta, \phi). \quad (9)$$

The density $\rho(\mathbf{r}, \omega)$ can be split into the total number density $\rho(\mathbf{r})$ and a normalized factor $\alpha(\mathbf{r}, \omega)$ as

$$\rho(\mathbf{r}, \omega) = \rho(\mathbf{r}) \alpha(\mathbf{r}, \omega), \quad (10)$$

where

$$\int d\omega \alpha(\mathbf{r}, \omega) = 1. \quad (11)$$

In the local-density approximation the Helmholtz free energy of the hard-sphere system^[23,25]

$$\begin{aligned} F^{\text{hs}}[\rho(\mathbf{r}, \omega)] &= \frac{1}{\beta} \int d\mathbf{r} \rho(\mathbf{r}) \left[\ln[\rho(\mathbf{r}) \Lambda^3] - 1 \right. \\ &+ \left. \frac{4\eta(\mathbf{r}) - 3\eta(\mathbf{r})^2}{(1 - \eta(\mathbf{r}))^2} \right] \\ &+ \frac{1}{\beta} \int d\mathbf{r} \rho(\mathbf{r}) \int d\omega \alpha(\mathbf{r}, \omega) \\ &\times \ln[4\pi\alpha(\mathbf{r}, \omega)] \end{aligned} \quad (12)$$

where $\beta = 1/k_B T$ is the inverse temperature, Λ is the thermal de Broglie wavelength and $\eta(\mathbf{r}) = (\pi/6)\rho(\mathbf{r})\sigma^3$ is the packing fraction.

In the mean field approximation where the pair distribution function takes its large-distance limit $g(\mathbf{r}_1, \omega_1, \mathbf{r}_2, \omega_2; \lambda) = 1$,^[6] the part of the free energy related to the spin-spin interactions in Eq. (7) becomes

$$\begin{aligned} F_{\text{MF}}^{\text{ss}}[\rho(\mathbf{r}, \omega)] &= \frac{1}{2} \int d\mathbf{r}_1 d\omega_1 d\mathbf{r}_2 d\omega_2 \\ &\times \rho(\mathbf{r}_1, \omega_1) \rho(\mathbf{r}_2, \omega_2) u^{\text{ss}}(r, \omega_1, \omega_2). \end{aligned} \quad (13)$$

After the decomposition $\rho(\mathbf{r}, \omega) = \rho(\mathbf{r})\alpha(\mathbf{r}, \omega)$, the minimum condition of the functional $\Omega[\rho(\mathbf{r}, \omega)]$ is equivalent to the simultaneous minimization of the grand canonical functional with respect to the total number density

$$\frac{\delta \Omega[\rho(\mathbf{r}, \omega), T, \mu]}{\delta \rho(\mathbf{r})} = 0, \quad (14)$$

and the orientational configuration

$$\frac{\delta \Omega[\rho(\mathbf{r}, \omega), T, \mu]}{\delta \alpha(\mathbf{r}, \omega)} = 0. \quad (15)$$

In the absence of the external field ($V_{\text{ext}}(\mathbf{r}, \omega) = 0$) the system is homogeneous in position, but could be ordered in orientation. So we have $\rho(\mathbf{r}) = \rho$, $\alpha(\mathbf{r}, \omega) = \alpha(\omega)$ and the one-particle density becomes

$$\rho(\mathbf{r}, \omega) = \rho \alpha(\omega), \quad (16)$$

with

$$\int d\omega \alpha(\omega) = 1. \quad (17)$$

In the mean field approximation we obtain the bulk expression of the grand-canonical free energy density

$$\begin{aligned} \frac{1}{V} \Omega_{\text{MF}}[\alpha(\omega), \rho, T, \mu] &= f_{\text{CS}}^{\text{hs}}(\rho) - \frac{1}{2} J^{\text{int}} \rho^2 |\bar{\mathbf{s}}|^2 \\ &+ \frac{\rho}{\beta} \int d\omega \alpha(\omega) \ln[4\pi\alpha(\omega)] - \mu \rho, \end{aligned} \quad (18)$$

where the Helmholtz free energy density of hard-sphere system $f_{\text{CS}}^{\text{hs}}(\rho)$ is given by Carnahan and Starling^[25]

$$f_{\text{CS}}^{\text{hs}}(\rho) = \frac{\rho}{\beta} \left[\ln(\rho \Lambda^3) - 1 + \frac{4\eta - 3\eta^2}{(1 - \eta)^2} \right] \quad (19)$$

and

$$\bar{\mathbf{s}} = \int d\omega \alpha(\omega) \mathbf{s}(\omega), \quad (20)$$

$$J^{\text{int}} = \int_\sigma^\infty dr 4\pi r^2 J(r) = 8\pi \varepsilon \sigma^3. \quad (21)$$

From the equilibrium condition (15) and the mean field grand-canonical free energy (18) we can obtain the equilibrium orientational distribution function

$$\alpha(\omega) = \frac{e^{\beta\rho J^{\text{int}} \mathbf{s} \cdot \bar{\mathbf{s}}}}{\int d\omega e^{\beta\rho J^{\text{int}} \mathbf{s} \cdot \bar{\mathbf{s}}}}. \quad (22)$$

Here we restrict to the case $\alpha(\omega) = \alpha(\theta)$ and have

$$\alpha(\omega) = \alpha(\theta) = \frac{1}{4\pi} e^{k(x) + x \cos \theta}, \quad (23)$$

where

$$k(x) = \ln\left(\frac{x}{\sinh x}\right), \quad (24)$$

$$x = \beta\rho J^{\text{int}} \xi, \quad (25)$$

with the magnetization $\xi = \int d\omega \alpha(\theta) \cos \theta$ determined by

$$\xi = \coth(\beta\rho J^{\text{int}} \xi) - \frac{1}{\beta\rho J^{\text{int}} \xi}. \quad (26)$$

Then we obtain

$$\frac{1}{V} \Omega_{\text{MF}} = f_{\text{CS}}^{\text{hs}}(\rho) + \frac{1}{2} J^{\text{int}} \rho^2 \xi^2 + \frac{\rho}{\beta} k(x) - \mu\rho. \quad (27)$$

From the equilibrium condition (14) we can obtain the chemical potential in the mean field approximation

$$\begin{aligned} \mu_{\text{MF}}(\rho, \xi) &= \frac{\partial f_{\text{CS}}^{\text{hs}}(\rho)}{\partial \rho} + \rho J^{\text{int}} \xi^2 + \frac{1}{\beta} k(x) \\ &+ \rho J^{\text{int}} \xi \frac{\partial k(x)}{\partial x}. \end{aligned} \quad (28)$$

4 Spinodal of Heisenberg Liquid

At a stable equilibrium state the grand potential has its minimum and its variation with respect to the changes of total number density and magnetization should be positive,^[20]

$$\frac{\delta\Omega}{V} = \frac{1}{2V} \left[\frac{\partial^2 \Omega}{\partial \rho^2} (\delta\rho)^2 + 2 \frac{\partial^2 \Omega}{\partial \rho \partial \xi} \delta\rho \delta\xi + \frac{\partial^2 \Omega}{\partial \xi^2} (\delta\xi)^2 \right] > 0. \quad (29)$$

The variation of the grand potential can be rewritten in a matrix form

$$\frac{\delta\Omega}{V} = \frac{J^{\text{int}} \rho^2}{2} \begin{pmatrix} \delta\rho & \delta\xi \end{pmatrix} \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix} \begin{pmatrix} \delta\rho/\rho \\ \delta\xi \end{pmatrix}, \quad (30)$$

where the matrix elements

$$\begin{aligned} M_{11} &= \frac{x^2}{\beta J^{\text{int}} \rho} \frac{\partial^2 k(x)}{\partial x^2} - \left(\frac{x}{\beta J^{\text{int}} \rho} \right)^2 \\ &+ \frac{1}{\beta J^{\text{int}} \rho} + \frac{\partial^2 \frac{\rho}{\beta J^{\text{int}} (1-\eta)^2}}{\partial \rho^2}, \end{aligned} \quad (31)$$

$$M_{21} = M_{12} = x \frac{\partial^2 k(x)}{\partial x^2}, \quad (32)$$

$$M_{22} = 1 + \beta J^{\text{int}} \rho \frac{\partial^2 k(x)}{\partial x^2}. \quad (33)$$

The matrix \mathbf{M} has two eigenvalues

$$\lambda_{1,2} = \frac{1}{2} (M_{22} + M_{11} \mp \sqrt{(M_{22} - M_{11})^2 + 4M_{12}^2}), \quad (34)$$

with the corresponding eigenvectors

$$\mathbf{x}_1 = (\cos \varphi, \sin \varphi), \quad (35)$$

$$\mathbf{x}_2 = (-\sin \varphi, \cos \varphi). \quad (36)$$

The angle φ above is determined by

$$\cos \varphi = \frac{a}{\sqrt{1+a^2}}, \quad (37)$$

$$a = -\frac{1}{2M_{12}} (M_{22} - M_{11} + \sqrt{(M_{22} - M_{11})^2 + 4M_{12}^2}). \quad (38)$$

With the eigenvalues and eigenvectors we can rewrite Eq. (32) as

$$\frac{\delta\Omega}{V} = \frac{J^{\text{int}} \rho^2}{2} (\lambda_1 \delta\rho_{\mathbf{x}_1}^2 + \lambda_2 \delta\rho_{\mathbf{x}_2}^2), \quad (39)$$

where

$$\delta\rho_{\mathbf{x}_1} = \cos \varphi \frac{\delta\rho}{\rho} + \sin \varphi \delta\xi, \quad (40)$$

$$\delta\rho_{\mathbf{x}_2} = -\sin \varphi \frac{\delta\rho}{\rho} + \cos \varphi \delta\xi. \quad (41)$$

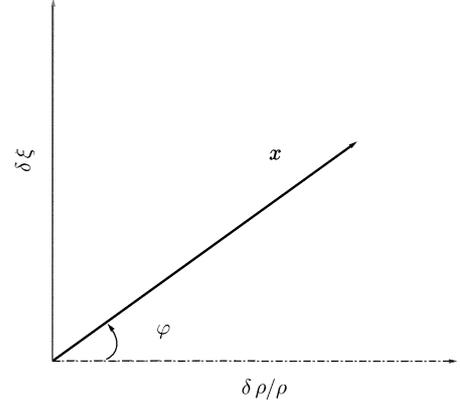


Fig. 1 A schematic plot of the eigenvector, which characterizes the phase transition. In the plane $(\delta\xi, \delta\rho/\rho)$ the eigenvector can be described by an angle φ , which is in the range $0 \leq \varphi < 180^\circ$. When $\varphi = 0$ the phase transition is a pure condensation. The phase transition is pure ferromagnetic when $\varphi = 90^\circ$.

The positive eigenvalues λ_1 and λ_2 of a stable state guarantee that the grand free energy will increase with respect to any variation of total number density and magnetization. If the smaller eigenvalue λ_1 vanishes, the system can deviate away from the original state without any increase of the grand free energy and becomes unstable. Now there is a phase transition. The eigenvector \mathbf{x}_1 corresponding to the zero eigenvalue characterizes this phase transition precisely^[21] and $\delta\rho_{\mathbf{x}_1}$ is the order parameter. In Fig. 1 we show a schematic plot of the eigenvector \mathbf{x}_1 in the plane $(\delta\rho/\rho, \delta\xi)$. The angle φ describes the portion of the total number density and the magnetization in the phase transition and is defined in the range $0 \leq \varphi < 180^\circ$. In general, the phase transition of the Heisenberg fluid is a combination of condensation and ferromagnetic phase transition. If $\varphi = 0$, we have then a pure condensation

phase transition. For φ near zero, we have a condensation dominant phase transition accompanied by a weak ferromagnetic phase transition. If $\varphi = 90^\circ$, we have a pure ferromagnetic phase transition. For φ near 90° , we have a ferromagnetic dominant phase transition accompanied by a weak condensation.

Now we investigate the phase behavior of the Heisenberg fluid by determining the border of stable region from the zero point of the smaller eigenvalue λ_1 . What we will obtain is the spinodal.

5 Results and Discussion

5.1 Phase Transition from Isotropic Phase with $\xi = 0$

At the isotropic phase of the Heisenberg fluid, there is no total magnetization and $x = 0$. In this case we have

$$\lim_{x \rightarrow 0} \frac{\partial^2 k(x)}{\partial x^2} = -\frac{1}{3}, \quad (42)$$

and the elements of the matrix M become

$$M_{11} = \frac{1}{\beta J^{\text{int}} \rho} + \frac{\partial^2 \frac{\rho}{\beta J^{\text{int}}} \frac{4\eta - 3\eta^2}{(1-\eta)^2}}{\partial \rho^2}, \quad (43)$$

$$M_{21} = M_{12} = 0, \quad (44)$$

$$M_{22} = 1 - \beta J^{\text{int}} \frac{\rho}{3}. \quad (45)$$

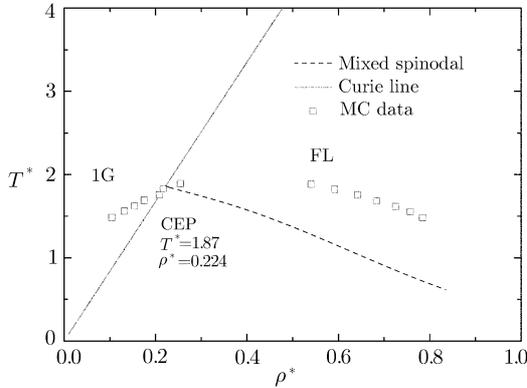


Fig. 2 The spinodal phase diagram of the Heisenberg liquid. There are two phases: the isotropic gas (IG) and the ferromagnetic liquid (FL). The CEP here is the abbreviation of the critical endpoint.

For all densities ρ the element M_{11} is positive. Only the element M_{22} can become zero, which corresponds to a pure ferromagnetic phase transition with $\varphi = 90^\circ$. From $M_{22} = 0$ we can get the Curie line of the Heisenberg fluid. With the reduced density $\rho^* = \rho\sigma^3$ and the reduced temperature $T^* = 1/\beta\varepsilon$ we can express the Curie line as

$$T^* = \frac{8\pi}{3} \rho^* \quad (46)$$

which agrees with the result of Tavares *et al.*^[9] and is shown in Fig. 2. It is the mean field approximation that

makes the phase transition be pure ferromagnetic. In a more accurate theory the phase transition is not pure ferromagnetic and should be accompanied by a weak condensation.

5.2 Ferromagnetic Phases $\xi \neq 0$

When $\xi \neq 0$ the matrix element $M_{12} = M_{21}$ is nonzero. The condition of $\lambda_1 = 0$ becomes

$$M_{11} \cdot M_{22} = M_{12}^2, \quad (47)$$

which can be solved numerically.

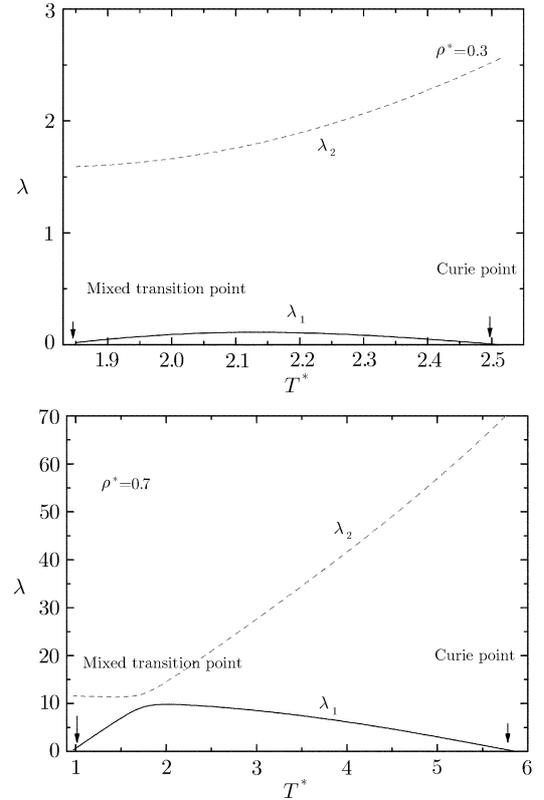


Fig. 3 Plot of the eigenvalues λ_1 and λ_2 vs T^* for the Heisenberg fluid at the densities $\rho^* = 0.3$ (up figure) and $\rho^* = 0.7$ (down figure). The Curie point and the mixed point here are on the Curies line and the mixed spinodal of Fig. 3 respectively.

In Fig. 3 the eigenvalues λ_1 and λ_2 are shown as a function of the reduced temperature T^* for densities $\rho = 0.3$ and 0.7 . The eigenvalue λ_2 keeps to be positive, but the eigenvalue λ_1 approaches zero when decreasing or increasing the temperature. The instability at higher temperature is actually on the Curie line where $\varphi = 90^\circ$. With the zero points of λ_1 for different densities ρ we can get the spinodal phase diagram of the Heisenberg liquid, which is shown in Fig. 2. Our spinodal here is inside the coexistence curve of isotropic vapor and ferromagnetic liquid which was obtained with Monte Carlo simulation.^[17] We find that the critical endpoint is at $\rho^* = 0.224$ and

$T^* = 1.87$, which is in agreement with Ref. [9]. Below the temperature of the critical endpoint there is a first-order phase transition between isotropic vapor and ferromagnetic liquid. For the spinodal near the ferromagnetic fluid the phase instability is a combination of ferromagnetic phase transition and condensation. To characterize the phase instability precisely we investigate the angle φ of the eigenvector \mathbf{x}_1 and the magnetization ξ along the spinodal, which are shown in Fig. 4. For densities larger than 0.6 the angle φ is very small and positive. The phase instability is predominantly related to the condensation. The positivity of φ means that an increase of the total number density will be accompanied by a small ordering. This

result is plausible, because the increase of total density will enhance the average attraction of the system which results in more ordering. The magnetization ξ for the density larger than 0.6 is larger than 0.9. With the decrease of the density the angle φ increases continuously. The phase instability has more and more portion of the ferromagnetic phase transition. At the density $\rho^* = 0.228$, there is $\varphi = 45^\circ$ and the phase instability is related equally to condensation and ferromagnetic phase transition. When approaching the critical endpoint $\rho^* = 0.224$ the angle φ approaches 90° . The phase transition at critical endpoint is then a pure ferromagnetic phase transition in the mean field approximation.

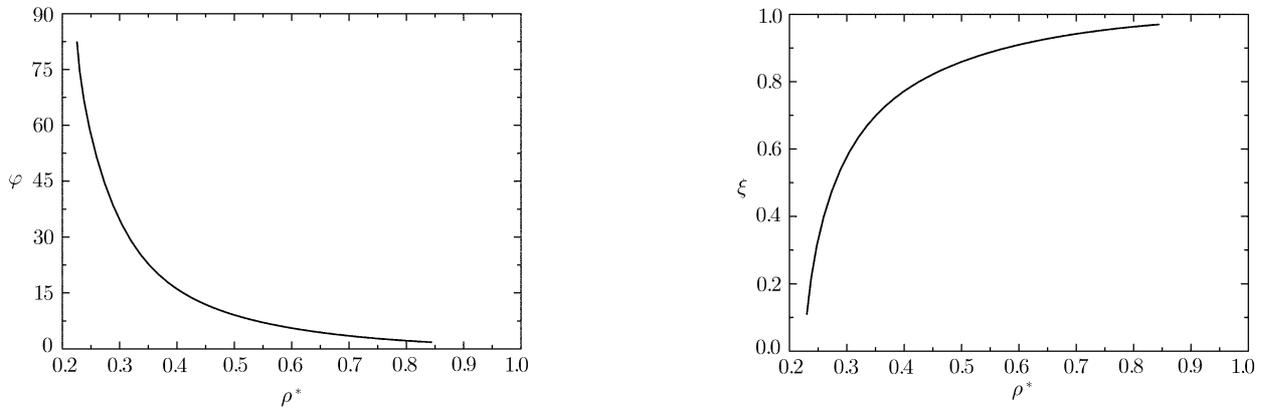


Fig. 4 Plot of the angle φ and the magnetization ξ with respect to density along the spinodal (dashed line in Fig. 3). φ is shown in the up figure and ξ is shown in the down figure.

6 Conclusions

Using the density functional theory in the mean-field approximation we have investigated the phase transition of Heisenberg liquid. The phase instability of the system is discussed with the method developed in Ref. [21]. From the matrix of the second derivatives of the grand canonical free energy Ω with respect to the particle density and the magnetization, we can determine the thermodynamic stability of the system. When the smaller eigenvalue of the matrix becomes zero, the system becomes unstable. The eigenvector corresponding to the zero eigenvalue can characterize the phase instability precisely. In the magnetization-density plane $(\delta\xi, \delta\rho/\rho)$ the eigenvector can be described by an angle φ which is in the range $0 \leq \varphi < 180^\circ$. If φ is very small the phase instability is predominantly a condensation. The phase instability is predominantly ferromagnetic when φ is near 90° .

For temperature above $T^* = 1.87$ of the critical endpoint, the angle $\varphi = 90^\circ$ and the phase transition is pure

ferromagnetic. This is the result in the mean-field approximation. With a more accurate approximation the angle φ does not equal exactly to 90° and is just near 90° . Then the particle density will be related to the phase transition, which is predominantly ferromagnetic and accompanied by a weak condensation. Below $T^* = 1.87$ there is a first-order phase transition between isotropic gas and ferromagnetic liquid. The spinodal we obtain is inside the coexistence curve of the Monte Carlo simulation and so in consistency with the MC data.^[17] The phase instability along the spinodal near the ferromagnetic liquid is a combination of condensation and ferromagnetic phase transition. With the decrease of temperature from the critical endpoint the angle φ decreases from 90° to nearly zero degree. So the phase instability changes from predominant ferromagnetic phase transition to predominant gas-liquid phase transition.

Acknowledgments

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