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Multicritical behaviour in magnetic fluids

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The phases of a magnetic fluid in an external field are considered. As model systems we take an ideal (hard core) gas with Ising interaction and a van der Waals gas with additional Heisenberg interaction. In mean field approximation various phases and critical points are identified. For appropriate values of the ratio of the magnetic to the non-magnetic interactions there exist multicritical points like tricritical points and critical end points. For the ideal Ising fluid we calculate the line of wing critical points analytically and prove classical tricritical behaviour. In the van der Waals case wing critical points and the gas-liquid critical point may coexist. The corresponding phase diagrams in (p, t, h) -space are shown.

Key words: *multicriticality, magnetic fluids, tricritical points*

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1. Introduction

Quite recently experimental evidence has been found for the existence of magnetic order in the liquid alloy $Co_{80}Pd_{20}$ [1]. So far liquid ferromagnetism has been observed only in colloidal systems, and Au-Co alloys [2,3]. Thus it seems that in the near future phase diagrams of real systems might be compared with theoretical predictions.

Theoretical models for a magnetic fluid which contain as model parameter the relative strengths of the magnetic interaction to the non-magnetic interaction may form phase diagrams of various topologies containing first and second order phase transition lines ending in critical or tricritical points as well as in critical end points [4,5]. These mean field calculations were performed at zero magnetic field. We have extended these calculations to non-zero field [6].

Several models exhibiting tricritical points are known [7], most of them are lattice models or models for mixtures (e.g. ^{3}He-^{4}He). In many magnetic models it is the competition of ferromagnetic and antiferromagnetic ordering which leads to multicritical behaviour. In magnetic liquids, it is the competition between magnetic and spatial (density) ordering which leads to multicriticality. The phase diagrams in the space of pressure, temperature and magnetic field are symmetric with respect to the magnetic field. Therefore in mean field theory, concerning the tricritical behaviour, classical critical exponents of symmetric type are found.

2. Ising fluid

Microscopically the properties of a magnetic fluid are fixed by its Hamiltonian. In our case we start from

$$\mathcal{H}\left(\{\mathbf{r}\},\{\sigma\}\right)=\sum_{i<j}\left[\varphi\left(\mathbf{r}_{ij}\right)+\sigma_i\sigma_j\varphi_{\mathrm{ex}}\left(\mathbf{r}_{ij}\right)\right]-H\sum_i\sigma_i.$$

In mean field approximation [5,8] one arrives at two equations of state. The magnetic one,

$$m(\rho,T,H)=\tanh\frac{a_{\mathrm{m}}\rho m+H}{k_{\mathrm{B}}T}, \tag{1}$$

renders the magnetization m as function of the particle-density ρ, the temperature T and the magnetic field H; a_{m} is a measure of the strength of the magnetic interaction. The equation of state for the pressure P of the system is the modified van der Waals equation

$$P\left(\rho,T,m^2\right)=\mathcal{MC}\left(k_{\mathrm{B}}T\frac{\rho}{1-b\rho}-\frac{1}{2}a_{\mathrm{m}}m^2\rho^2-\frac{1}{2}a\rho^2\right), \tag{2}$$

where in addition to the attractive interaction of strength a the magnetic interaction appears; $\mathcal{MC}$ advises to perform a Maxwell-construction if necessary. The coefficients a and a_{m} are integral expressions involving the interaction energies φ and φ_{ex} respectively.

The features of a phase diagram of this system for $H=0$ depend on the ratio $R=a_{\mathrm{m}}/a$ of the two types of interaction. In any case there is a line L_λ of critical points of the transition between a magnetic and a nonmagnetic fluid. For a certain range of values of R this line terminates in a tricritical point (TCP). At this point the critical properties differ from those along the critical line L_λ. So the critical exponent β at the TCP is 1/4 instead of 1/2 along L_λ.

More important for our purpose is the fact that a TCP is a point, where 3 critical lines meet. To check this property of the point in question, i.e. being really a TCP, we considered the phases in (T,P,H)-space. In particular we calculate the edges $\mathrm{L}_\pm$ of the so-called wings from the conditions

$$\partial p/\partial x=0,\quad \partial^2 p/\partial x^2=0 \tag{3}$$

for the scaled equations of state

$$m = \tanh\left(\frac{Rm}{xt}+\frac{h}{t}\right), \tag{4}$$

$$p = \mathcal{MC}\left(\frac{t}{x-1}-R\frac{m^2}{2x^2}-\frac{1}{2x^2}\right). \tag{5}$$

These equations, where $x{=}\frac{1}{\rho b},\ t{=}\frac{k_{\mathrm{B}}Tb}{a},\ h{=}Hb/a,\ p{=}p\left(x,t,m^2\right){=}\frac{b^2}{a}P\left(\rho,T,m^2\right)$, depend only on the parameter R.

In the more simple case of an ideal fluid, $a = 0$ or $R \to \infty$ [4], whose equations of state

$$m = \tanh\left(\frac{m}{x\bar{t}} + \frac{\bar{h}}{\bar{t}}\right), \tag{6}$$

$$\bar{p} = \frac{\bar{t}}{x-1} - \frac{m^2}{2x^2}. \tag{7}$$

are obtained from (4,5) by the rescaling transformation $\bar{p} = p/R, \quad \bar{t} = t/R, \quad \bar{h} = h/R$. We determine $m\left(x\right)$ and $\bar{t}\left(x\right)$ from (3) and thus we find the following parametric form for the wing edge L_+

$$\begin{pmatrix} \bar{t} \\ \bar{h} \\ \bar{p} \end{pmatrix} = \begin{pmatrix} \frac{2(x-1)^2}{9x^2-10x+3} \\ \frac{1}{2}\bar{t}\left(x\right)\ln\frac{1+m(x)}{1-m(x)} - \frac{m(x)}{x} \\ \frac{1}{2}\frac{-2x-1+x^2}{9x^2-10x+3} \end{pmatrix} \tag{8}$$

with $m^2\left(x\right) = \frac{x^2\left(6x-x^2-3\right)}{9x^2-10x+3}$, see figure 1a.

For finite values of R one obtains from (3) only a cubic equation for the temperature t as a function of x. To be more specific we get the equation

$$z^3 - 2z^2\frac{\left(Rx+6x-3\right)}{\left(9x^2-10x+3\right)} + z\frac{\left(R\left(x^2+6x-3\right)+3\left(x^2+2x-1\right)\right)}{x^2\left(9x^2-10x+3\right)} - \frac{2\left(R+1\right)}{x^2\left(9x^2-10x+3\right)} = 0\,. \tag{9}$$

for the variable $z = xt/\left(x-1\right)^2$. If there are wings – this is tantamount to the existence of a tricritical point – equation (9) has at most 2 physical solutions, corresponding to the wing critical point and the gas-liquid critical point. Inserting these

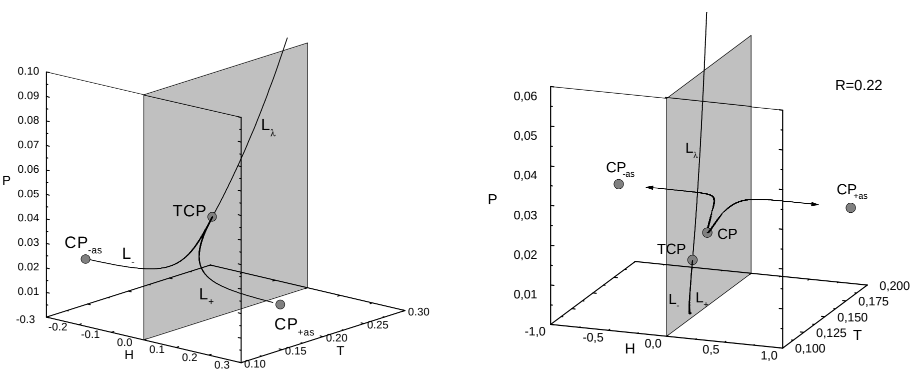

Figure 1. Wing lines in (a) the ideal Ising fluid and (b) the van der Waals Ising fluid. From [6].

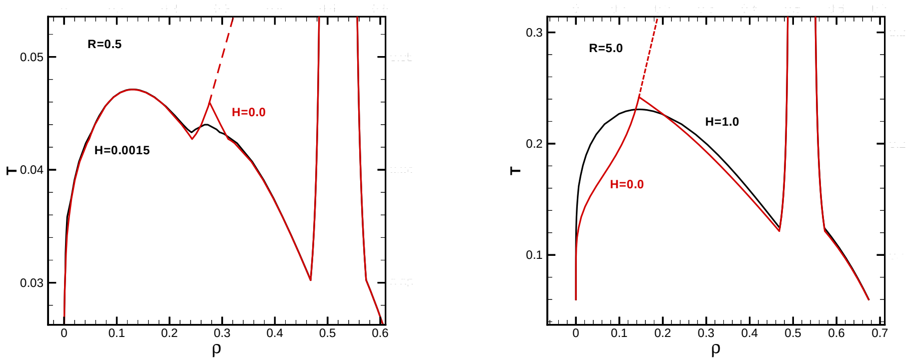

Figure 2. Heisenberg fluid at two different values of R. In both cases the (t,ρ)-diagram without and with a magnetic field is shown.

solutions together with a second condition obtained from (3)

$$m^2 = \frac{(1 - xt/R)\left((x-1)^2 - xt^3\right)}{(x-1)^2 - xt(2x-1)} \tag{10}$$

into the equations of state, we again can construct the wing edges in (p,t,h)-space [6], see figure 1b.

From the explicit equation of the wing lines, we find that the wing lines approach the tricritical point $(h \to 0)$ with a power law $t_{\mathrm{w}}(h) - t_{\mathrm{t}}$ and $p_{\mathrm{w}}(h) - p_{\mathrm{t}} \propto h^{2/5}$. The magnetization on the wing lines goes to zero in this limit with an exponent $\delta_{\mathrm{t}} = 5$ instead of $\delta = 3$ at the magnetic phase transition.

3. Heisenberg fluid

In the study of mean field models for a Heisenberg fluid we restrict ourselves to a numerical determination of the various phase diagrams; therefore we can now use an improved hard sphere contribution to the pressure more appropriate to the three dimensional case than the van der Waals expression $k_{\mathrm{B}}T/(1-b\rho)$ in equation 2. We use for the hard sphere pressure the equations given by Hall [9] in the whole region of densities,

$$p_{\mathrm{hs}}(\rho,T) = \begin{cases} T\rho\frac{1+\rho^2-0.67825\rho^3-\rho^4-0.5\rho^5-1.7\rho^6}{1-3\rho+3\rho^2-1.04305\rho^3}, & \rho \leqslant 0.4927 \\ p_{\mathrm{hs}}(0.4927,T), & 0.4927 < \rho \leqslant 0.54447 \\ T\rho\frac{1+\rho+\rho^2-0.67825\rho^3-\rho^4-0.5\rho^5-6.028\mathrm{e}^{\xi(7.9-3.9\xi)}\rho^6}{1-3\rho+3\rho^2-1.04305\rho^3}, & \rho > 0.54447, \end{cases} \tag{11}$$

For $H = 0$ this leads to the same results as in [5] where a different hard sphere equation of state is used.

Since the magnetic interaction is now of Heisenberg type, the magnetic equation of state reads

$$m(\rho,T,H) = L\left(\frac{R\rho m}{T} + \frac{H}{T}\right) \tag{12}$$

with the Langevin function $L(z) = \coth(z) - 1/z$. ρ, T, p and H are measured in the units $\pi b^3/6$, $\pi b^3 k/(6a)$, $(\pi b^3/6)^2/a$ and $\pi b^3/a$ respectively.

The phase diagrams include now the liquid solid transition and, depending on R, several topologies exist. Two examples in zero and finite field are shown in figure 2, one with a gas liquid and wing critical point, and one where no gas-liquid critical point appears.

4. Outlook

Fluctuations change the topology of a phase diagram as well as critical exponents for the second order phase transitions [10–12]. At the tricritical point classical exponents are correct apart from logarithmic corrections to the power laws. However the existence of the tricritical point is still under discussion since Monte Carlo calculations in three dimensions [13–15] indicate a critical end point instead.

It seems to be worthwhile to look for the critical exponents also in the case of the gas-liquid transitions in finite magnetic field and to extend the Monte Carlo simulations to finite magnetic field (this has been performed quite recently [16]) to look for the wing critical point.

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Мультикритична поведінка у магнітних флюїдах

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Для моделей ідеального (тверді кульки) газу із взаємодією Ізінга та газу Ван дер Ваальса із взаємодією типу Гайзенберга досліджуються фази магнітного флюїду у зовнішньому полі. У наближенні середнього поля ідентифіковані різні фази та критичні точки. Показано, що для певних значень відношення констант магнітної та немагнітної взаємодій існують мультикритичні точки типу трикритичних і критичних кінцевих точок. Для моделі ідеального ізінгівського флюїду аналітично розраховано бокові лінії критичних точок та знайдено класичну трикритичну поведінку. У випадку моделі Ван дер Ваальса бокові критичні лінії та критична точка можуть співіснувати. Наведені відповідні фазові діаграми у змінних (p, t, h) .

Ключові слова: *мультикритичність, магнітні флюїди, трикритичні точки*

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