Landau Theory, Fluctuations & Break Down of Landau Theory

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ETH Zürich

Spring Term 2007 - LaTeX Slides

1 Introduction
- Second Order Phase Transitions
- Order Parameter
- Bragg-Williams Theory

2 Landau Theory
- Power Series about the critical Point
- External Field
- The Minima
- Critical Behavior

3 Fluctuations
- Fluctuations in the Order Parameter
- The Correlation Length
- The Ginzburg Criterion
- Landau Theory in higher Dimensions

4 Conclusive Words

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Two Types of Phase Transitions

We distinguish:

- First order phase transitions: Transition occurs as an abrupt change in symmetry.

Examples:

- Melting or boiling water

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- First order phase transitions: Transition occurs as an abrupt change in symmetry.
- Second order phase transitions: Symmetry is changed in a continuous way.

Examples:

- Melting or boiling water
- Body center cubic lattice

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Describing the Transition

Second order phase transitions due to a continuous change in symmetry can be described with an order parameter η with the properties:

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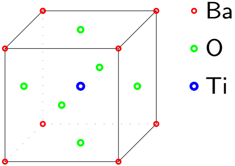
Second order phase transitions due to a continuous change in symmetry can be described with an order parameter η with the properties:

- η is zero for the phase of higher symmetry,
- η takes non-zero values (positive or negative) for the “asymmetric” phase,
- for second order phase transitions, η is a continuous function of temperature.

Examples

Some concrete examples for different types of second order phase transitions:

- $\mathrm{BaTiO_3}$: $\eta = d$



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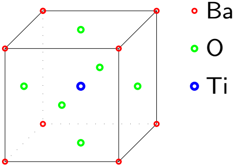

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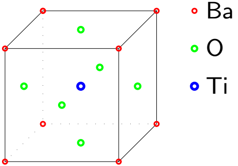

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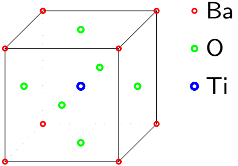

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- Anti-Ferromagnet: $\eta = m_{\mathrm{sublattice}}$

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Simplified Ising Model

Ising model:

$$U = \langle H \rangle = \sum_i H_i S_i - \sum_{i,j} J_{ij} S_i S_j - \sum_{i,j,k} K_{ijk} S_i S_j S_k - \ldots$$

with $S_i = \pm 1$.

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Bragg Williams internal energy:

$$U = -J \sum_{\langle i,j \rangle} \eta^2 = -J \frac{Nz\eta^2}{2}$$

Entropy

For a given $\eta = m = (N_\uparrow - N_\downarrow)/N$, the entropy is the logarithm of the number of configurations with a given number $N_\uparrow$:

$$S = \ln \binom{N}{N_\uparrow} = \ln \left(\frac{N!}{(N(1+\eta)/2)!(N(1-\eta)/2)!} \right)$$

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Sterling's Approximation for large N

$$\ln(N!) \approx N(\ln(N) - 1)$$

leads to

$$S = N \left(\ln(2) - \frac{1+\eta}{2} \ln(1+\eta) - \frac{1-\eta}{2} \ln(1-\eta) \right)$$

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Expanded for small η

$$S = N\left(\ln(2) - \frac{1}{2}\eta^2 - \frac{1}{12}\eta^4 - \ldots \right)$$

Bragg-Williams free Energy

So we have the free energy and the entropy

$$U = -J \sum_{\langle i,j \rangle} \eta^2 = -J \frac{Nz\eta^2}{2}, \qquad \frac{S}{N} = \ln(2) - \frac{1}{2}\eta^2 - \frac{1}{12}\eta^4 - \dots$$

With this we can build:

Bragg-Williams free Energy per Site

$$\frac{F(T,\eta)}{N} = \frac{U - TS}{N} = -T\ln(2) + \frac{1}{2}(T - T_c)\eta^2 + \frac{1}{12}T\eta^4 + \dots$$

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In most cases, $T > T_c$ corresponds to the phase of higher symmetry. Our further discussion will only cover these cases.

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Example for an exception: Seignettesalz $(\mathrm{KNaC_4H_4O_6 \cdot 4H_2O})$

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General Power Series

Assumption:

For small η, the free energy can be expanded in a power series in η

$$F(P, T, \eta) = F_0 + \alpha\eta + A\eta^2 + C\eta^3 + B\eta^4 + \dots$$

where F_0, α, A, C, B are functions of P and T.

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For given P and T, η is to be determined by minimizing F

$$\frac{\partial F(P,T,\eta)}{\partial \eta} = \alpha + 2A\eta + 3C\eta^2 + 4B\eta^3 = 0$$

The Coefficient α

Free energy and derivative:

$$\begin{aligned} F(P,T,\eta) &= F_0 + \alpha\eta + A\eta^2 + C\eta^3 + B\eta^4 \\ \frac{\partial F(P,T,\eta)}{\partial\eta} &= \alpha + 2A\eta + 3C\eta^2 + 4B\eta^3 \end{aligned}$$

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Property 1:

$$\alpha = 0$$

The Coefficient $A(P, T)$

Free energy derivatives:

$$\frac{\partial F(P,T,\eta)}{\partial \eta} = 2A\eta + 3C\eta^2 + 4B\eta^3$$

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Property 2:

$$A(P,T) = 0 \quad \text{at critical point}$$

$B(P,T)$ and $C(P,T)$ at critical Point

Free energy derivatives at critical point:

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Property 2:

$$C(P,T) = 0, \quad B(P,T) > 0 \quad \text{at critical point}$$

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Assumption:

$$C(P,T) \equiv 0$$

The Landau free Energy

The discussion of the coefficients leads to

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Applying an external Field

Applying an external Field h, we have to add a term $-hV\eta$ to the free energy:

Landau free Energy with external Field:

$$F_h(P, T, \eta) \quad = \quad F_0(P, T) + a(P)t\eta^2 + B(P)\eta^4 - hV\eta$$

Where we wrote $t := (T - T_c)$.

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Minima for $h = 0$

Derivative of free energy without external field

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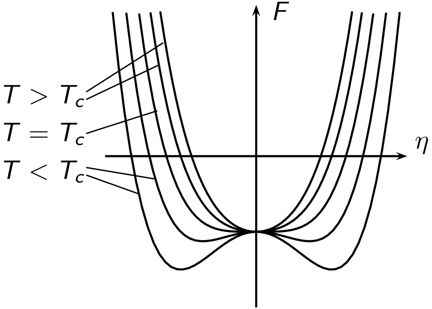
- $t < 0$: Two equivalent minima at

$$\eta = \pm\sqrt{\frac{a}{2B}(T_c - T)}$$

Minima for $h = 0$

Free energy without external field for various temperatures:



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Look at it as

$$(hV)(\eta) = 2at\eta + 4B\eta^3$$

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For $t > 0$, the function $(hV)(\eta) = 2at\eta + 4B\eta^3$ looks like:

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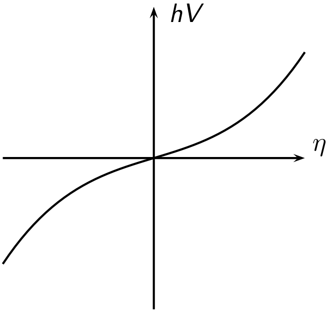

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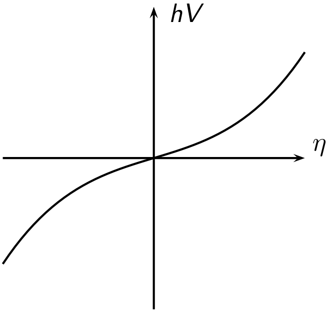

$\Rightarrow$ There is a unique solution $\eta \neq 0$.

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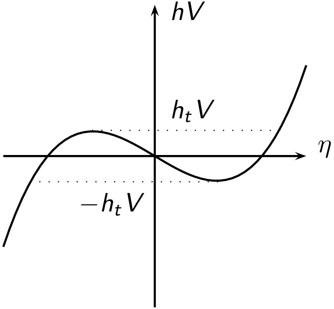

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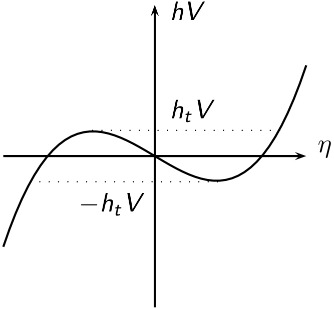

$\Rightarrow$ In a region $-h_t < h < h_t$, there are three solutions $\eta \neq 0$.

The characteristic Field h_t

We take the derivative of $(hV)(\eta)$

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This has solutions

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It follows

$$h_t = \frac{2at}{V}\left(\frac{a|t|}{6B}\right)^{1/2} + \frac{4B}{V}\left(\frac{a|t|}{6B}\right)^{3/2} = \left(\frac{2}{3}\right)^{2/3}\frac{a^{3/2}|t|^{3/2}}{VB^{1/2}}$$

The global Minimum for $-h_t < h < h_t$

We use

$$\left.\frac{\partial}{\partial h}\right|_T (2at\eta + 4B\eta^3) = \left(\frac{\partial\eta}{\partial h}\right)_T (2at + 12B\eta^2) = V$$

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to get

$$\left(\frac{\partial \eta}{\partial h}\right)_T \left(\frac{\partial^2 F_h(P,T,\eta)}{\partial \eta^2}\right) = V$$

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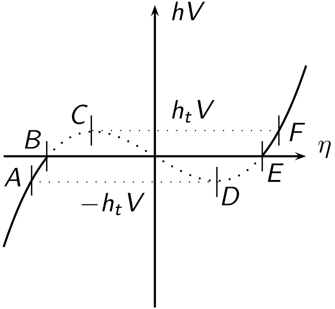

The global Minimum for $-h_t < h < h_t$

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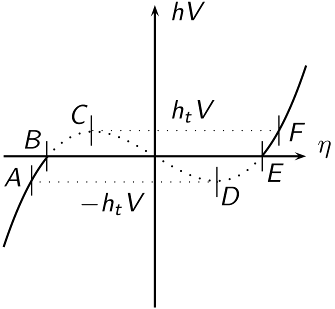

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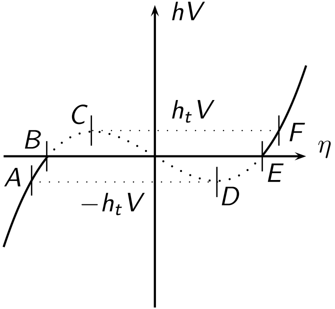

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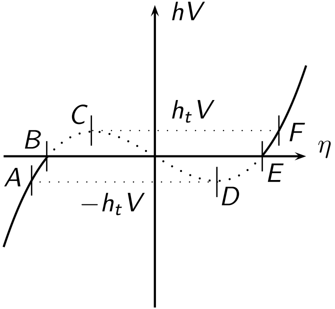

- CD: $(\partial\eta/\partial h)_T < 0 \rightarrow$ Maximum
- BC: $(\partial\eta/\partial h)_T > 0 \rightarrow$ Minimum
- EF: $(\partial\eta/\partial h)_T > 0 \rightarrow$ global Minimum

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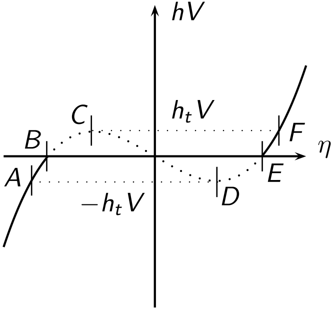
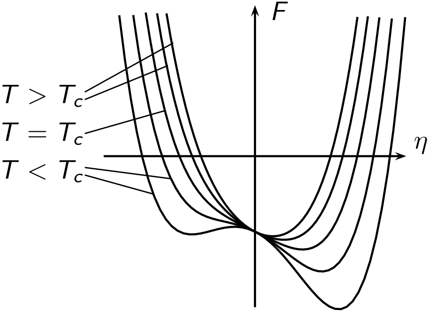


$\Rightarrow$ The global minimum lies on the sections AB or EF.

Minima for $h \neq 0$

Free energy with an external field $h > 0$ for various temperatures:



From this can actually be seen that $\alpha = 0$ also for $t < 0$.

Results

Without external field:

- $t > 0$: Minimum at $\eta = 0$
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Without external field:

- $t > 0$: Minimum at $\eta = 0$
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With an external field $h \neq 0$:

- $t > 0$: Minimum at $\eta \neq 0$
- $t < 0$: Minimum at $\eta \neq 0$

Interpretation:

The field breaks the symmetry and there is no more phase transition!

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The specific Heat c_P

The Entropy near the critical point is given by

$$S = -\frac{\partial F(P, T, \eta)}{\partial T} = S_0 - \frac{\partial A(P, T)}{\partial T}\eta^2 \qquad \frac{\partial F}{\partial \eta} = 0$$

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$$S = \begin{cases} S_0 + \frac{a^2}{2B}(T - T_c), & T < T_c \\ S_0, & T > T_c \end{cases}$$

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It follows

$$C_P = T\left(\frac{\partial S}{\partial T}\right)_P = \begin{cases} C_{P0} + \frac{a^2}{2B}T, & T < T_c \\ C_{P0}, & T > T_c \end{cases}$$

The Susceptibility χ

The susceptibility is defined as

$$\chi \;:=\; \left(\frac{\partial\eta}{\partial h}\right)_{T;h\to 0} \;=\; \lim_{h\to 0}\frac{V}{2at+12B\eta^2}$$

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Using $\lim_{h\to 0} \eta^2 = 0$ for $t > 0$ and $\eta^2 = \frac{a}{2B}(T_c - T)$ for $t < 0$, we get

$$\chi \ = \ \begin{cases} \frac{V}{4a(T_c - T)}, & T < T_c \\ \frac{V}{2a(T - T_c)}, & T > T_c \end{cases}$$

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Mean Square Fluctuation

Consider $\eta \equiv \eta(\mathbf{r}) = \overline{\eta} + \Delta\eta(\mathbf{r})$.

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For small deviations from $\overline{\eta}$

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We get for the mean square fluctuation

$$\omega \sim e^{-\frac{(\eta - \overline{\eta})^2 V}{2\chi k_B T_c}} \Rightarrow \left\langle (\Delta\eta)^2 \right\rangle = \frac{k_B T_c \chi}{V}$$

Landau free Energy Density

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$$F = \int \mathcal{F}(P, T, \eta(\mathbf{r}))\, \mathrm{d}\mathbf{r}$$

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Assuming that $g_{ik} = g\delta_{ik}$ with $g > 0$, we get the Landau free energy density

$$\mathcal{F}(P, T, \eta) = \mathcal{F}_0(P, T) + \alpha t\eta^2 + b\eta^4 + g\left(\frac{\partial\eta}{\partial\mathbf{r}}\right)^2 - \eta h$$

where $\alpha = a/V$ and $b = B/V$.

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Concepts from Statistical Mechanics

The Helmholtz free energy ist given by

$$A = -k_B T \ln\left[Z(h(\mathbf{r}))\right]$$

where $Z(h(\mathbf{r}))$ is the partition function, given by

$$Z(h(\mathbf{r})) = \mathrm{Tr}\,\exp\left[-\frac{1}{k_B T}\left(H(\eta(\mathbf{r})) - \int \mathrm{d}^d\mathbf{r}\, h(\mathbf{r})\eta(\mathbf{r})\right)\right]$$

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The expectation value of $\eta(\mathbf{r})$ then can be obtained by

$$\langle\eta(\mathbf{r})\rangle = -\frac{\delta A}{\delta h(\mathbf{r}')} = \lim_{\varepsilon\to 0} \frac{A(h(\mathbf{r}) + \varepsilon\delta(\mathbf{r}-\mathbf{r}')) - A(h(\mathbf{r}))}{\varepsilon}$$

Concepts from Statistical Mechanics

Defining the generalized isothermal susceptibility by

$$\chi_T(\mathbf{r},\mathbf{r}') = \frac{\delta \langle \eta(\mathbf{r}) \rangle}{\delta h(\mathbf{r}')}$$

we calculate the relation

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\chi_T(\mathbf{r}, \mathbf{r}') &= -\frac{\delta^2 A}{\delta h(\mathbf{r}) \delta h(\mathbf{r}')} \\
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&= \frac{1}{k_B T} \left(\langle \eta(\mathbf{r}) \eta(\mathbf{r}') \rangle - \langle \eta(\mathbf{r}) \rangle \langle \eta(\mathbf{r}') \rangle \right)
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&= \frac{1}{k_B T} \left(\langle \eta(\mathbf{r}) \eta(\mathbf{r}') \rangle - \langle \eta(\mathbf{r}) \rangle \langle \eta(\mathbf{r}') \rangle \right) \\
&= \frac{1}{k_B T} G(\mathbf{r}, \mathbf{r}')
\end{aligned}$$

with $G(\mathbf{r}, \mathbf{r}')$, the correlation function.

Differential Equation for $G(\mathbf{r}, \mathbf{r}')$

Now we take the functional derivative of the Landau free energy

$$\frac{\delta F}{\delta \eta(\mathbf{r})} = 2\alpha t \eta(\mathbf{r}) + 4b\eta^3(\mathbf{r}) + 2g\nabla^2\eta(\mathbf{r}) - h(\mathbf{r}) = 0$$

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This has to be satisfied by $\langle \eta(\mathbf{r}) \rangle$, so

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With the relation from before, this leads to

$$\frac{1}{k_B T} \left(2\alpha t + 12b\eta^2(\mathbf{r}) - 2g\nabla^2\right) G(\mathbf{r} - \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}')$$

Differential Equation for $G(\mathbf{r}, \mathbf{r}')$

We have

$$\frac{1}{k_B T}\left(2\alpha t + 12 b\eta^2(\mathbf{r}) - 2g\nabla^2\right) G(\mathbf{r}-\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}')$$

and go back to the case $\eta(\mathbf{r}) \equiv \eta$ and use the equilibrium values $\eta^2 = 0$ for $t > 0$ and $\eta^2 = -at/2b$ for $t < 0$ to get

$$\left(\frac{1}{\xi^2(t)} - \nabla^2\right) G(\mathbf{r}-\mathbf{r}') = \frac{k_B T}{2g}\delta(\mathbf{r}-\mathbf{r}')$$

with

$$\xi(t) = \begin{cases} \left(\frac{g}{\alpha t}\right)^{1/2} & T > T_c \\ \left(\frac{g}{2\alpha(-t)}\right)^{1/2} & T < T_c \end{cases}$$

The correlation length $\xi(t)$

For

$$\left(\frac{1}{\xi^2(t)} - \nabla^2\right) G(\mathbf{r} - \mathbf{r}') = \frac{k_B T}{2g} \delta(\mathbf{r} - \mathbf{r}')$$

we use the Fourier Transform to get

$$\hat{G}(k) = \frac{k_B T}{2g} \frac{1}{k^2 + \xi^{-2}}$$

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$$\hat{G}(k) = \frac{k_B T}{2g}\,\frac{1}{k^2 + \xi^{-2}}$$

This is itself the Fourier Transform of the Yukawa Potential, multiplied by $k_B T/8\pi g$. So

$$G(\mathbf{r}) = \frac{k_B T}{8\pi g}\,\frac{e^{-\frac{r}{\xi}}}{r}$$

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That's why we call $\xi(t)$ the correlation length.

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The Ginzburg Criterion for $d = 3$

The criterion for Landau Theory to be valid is

$$\left\langle (\Delta\eta)^2 \right\rangle_{\xi^3} = \frac{k_B T_c \chi}{\xi^3} \ll \frac{\alpha |t|}{2b} = \eta_0^{t<0}$$

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Using the results $\chi = 1/4\alpha|t|$ and $\xi = (g/2\alpha|t|)^{1/2}$ for $t < 0$ from before, we get

$$\alpha|t| \gg \frac{k_B T_c^2 b^2}{g^3}$$

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$$\alpha|t| \gg \frac{k_B T_c^2 b^2}{g^3}$$

Requiring $t \ll T_c$, this leads to

$$\frac{k_B^2 T_c b^2}{\alpha g^3} \ll 1$$

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The Ginzburg Criterion for $d < 3$

We could also formulate the criterion for Landau Theory to be valid for an arbitrary dimension d

$$\frac{T_c \chi}{\xi^3} \ll \frac{\alpha |t|}{2b} \quad \rightarrow \quad \frac{T_c \chi}{\xi^d} \ll \frac{\alpha |t|}{2b}$$

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$$\frac{b}{g^{d/2}} k_B T_c \left(\alpha|t|\right)^{d/2-2} \ll 1$$

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$$\frac{b}{g^{d/2}} \, k_B T_c \left(\alpha |t|\right)^{d/2-2} \ll 1$$

So we get:

- $d > 4$: The criterion can always be satisfied near the critical point,
- $d = 4$: The criterion is either satisfied or not for any temperature.

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- Quantitatively wrong predictions, but good qualitative description

Conclusive Words

- Bragg-Williams Theory
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- Quantitatively wrong predictions, but good qualitative description
- $d \geq 4$: Ginzburg criterion satisfied