

Phase transitions and Critical Phenomena

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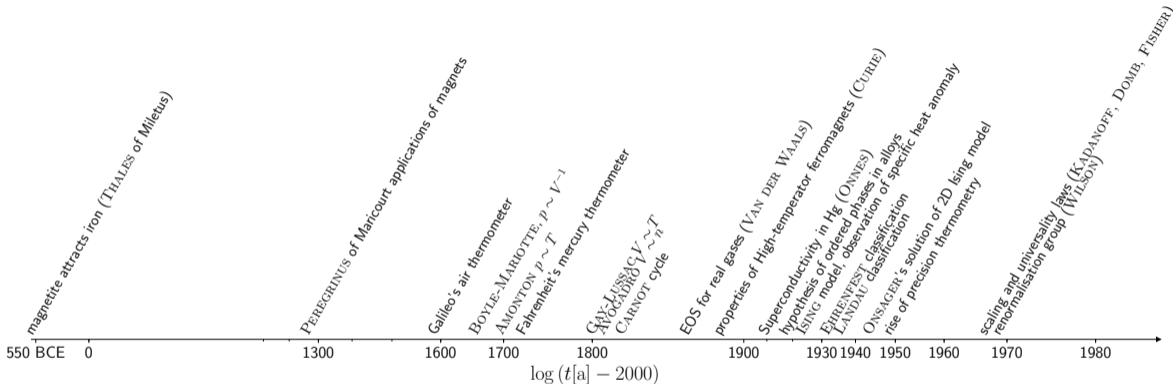


Figure: Timeline of milestones for the theory of phase transitions.^{[1][2]}

[1] Papon, Leblond, and Meijer, *Physics of Phase Transitions*, p. 33ff.

[2] Lindley, *Degrees Kelvin: A Tale of Genius, Invention, and Tragedy*, p. 99f.

Thermodynamics – The Basics

- Recall thermodynamic potentials:

Thermodynamic potential	fluid system	magnetic system
Internal energy $U(S, V)$	$dU = TdS - pdV$	$dU = TdS + HdM$
Enthalpy $H'(S, p)$	$dH' = TdS + Vdp$	$dH' = TdS - MdH$
Gibbs free energy $G(T, p)$	$dG = -SdT + Vdp$	$dG = -SdT - MdH$
Helmholtz free energy $F(T, V)$	$dF = -SdT - pdV$	$dF = -SdT + HdM$

- Switching is obtained via a *Legendre transform* in the state variables.
- Of course, additions like „ $+\mu dN$ “ are possible but for now we will ignore them.

Response Functions

From the thermodynamic potentials and the state variables the response functions can be derived.

- Specific heat

$$C_x \equiv \left(\frac{dQ}{dT} \right)_x = T \left(\frac{\partial S}{\partial T} \right)_x \quad x = p, V, M, H \quad (1)$$

- In the fluid system:
 - Compressibility

$$\kappa_x \equiv -\frac{1}{V} \left(\frac{\partial V}{\partial p} \right)_x \quad x = T, S \quad (2)$$

- Coefficient of thermal expansion

$$\alpha_p \equiv \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_p \quad (3)$$

Response Functions

From the thermodynamic potentials and the state variables the response functions can be derived.

- Specific heat

$$C_x \equiv \left(\frac{dQ}{dT} \right)_x = T \left(\frac{\partial S}{\partial T} \right)_x \quad x = p, V, M, H \quad (1)$$

- In a magnetic system:

- Susceptibility

$$\chi_x \equiv \left(\frac{\partial M}{\partial H} \right)_x \quad x = T, S \quad (2)$$

- Coefficient of thermal expansion

$$\alpha_H \equiv \left(\frac{\partial M}{\partial T} \right)_H \quad (3)$$

Response Functions as Second Derivatives

- Upon further inspection one can identify the previous quantities as second derivatives of the thermodynamic potentials

$$C_V = -T \left(\frac{\partial^2 F}{\partial T^2} \right)_V \qquad C_p = -T \left(\frac{\partial^2 G}{\partial T^2} \right)_p \qquad (4)$$

$$\kappa_S = -\frac{1}{T} \left(\frac{\partial^2 H'}{\partial p^2} \right)_S \qquad \kappa_T = -\frac{1}{T} \left(\frac{\partial^2 G}{\partial p^2} \right)_T \qquad (5)$$

$$\chi_S = -\left(\frac{\partial^2 H'}{\partial H^2} \right)_S \qquad \chi_T = -\left(\frac{\partial^2 G}{\partial H^2} \right)_T \qquad (6)$$

Intrinsic Relations

- Due to their relation via the potentials and the equation(s) of state, the response functions are not linearly independent and obey a variety of relations.

$$\kappa_T (C_p - C_V) = TV\alpha_p^2 \quad (7)$$

$$C_P (\kappa_T - \kappa_S) = TV\alpha_p^2 \quad (8)$$

$$\frac{C_p}{C_V} = \frac{\kappa_T}{\kappa_S} \quad (9)$$

- Taking stability and physicality into account, this implies

$$C_p \geq C_V \quad (10)$$

$$\kappa_T \geq \kappa_S \quad (11)$$

- Similar relations hold for the magnetic system

Convexity Relations

- With these relations and response functions at hand, claims on the convexity of the potentials can be made
 - $G(T, p)$ is *concave* in both its arguments,
 - $F(T, V)$ is *concave* with respect to T and *convex* with respect to V .
- For a magnetic system the claims $\chi_x, C_x \geq 0$ can not be made to show convexity.
- Convexity (concavity) follows nevertheless for systems with Hamiltonian (GRIFFITH 1964)

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 - H\hat{M} \quad (12)$$

- Under this condition
 - $G(T, H)$ is *concave* in both its arguments,
 - $F(T, M)$ is *concave* with respect to T and *convex* with respect to M .

Definition – Phase transitions

- Substances of fixed (chemical) composition exist in homogenous forms.
- The properties of those phases are distinguishable (e. g. solid, liquid, gas).
 - e. g. Several varieties of ice corresponding to different crystalline forms.
- Phase transitions (abb. PT) are marked by rapid changes in the properties of the described system.
- They are induced by changes of the intensive thermodynamic variables fueled from the outside.
- Schematically, there are PTs *with* and *without* „latent heat“.

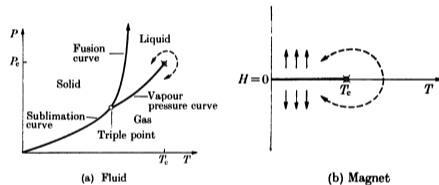


Figure: Phase transitions for (a) a fluid system (b) a magnetic system.^[3]

[3] Stanley and Ahlers, *Introduction to phase transitions and critical phenomena*, p. 2

Example – The Phase Diagram of Water

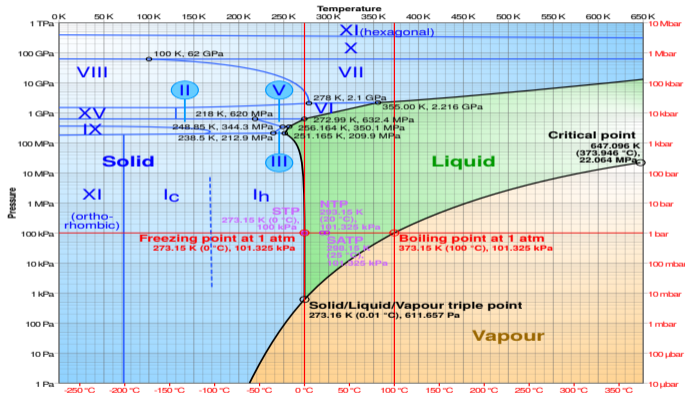


Figure: CMG Lee, *Phase diagram of water as a log-lin chart with pressure from 1 Pa to 1 TPa and temperature from 0 K to 650 K.*

Example – The Phase Diagram of a Pure Substance

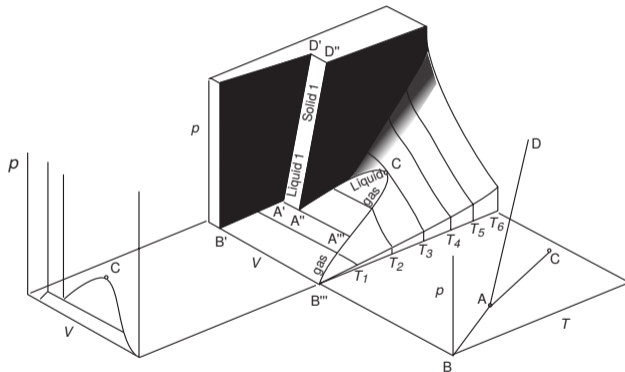


Figure: Phase diagram of a pure substance.^[4]

[4] Papon, Leblond, and Meijer, *Physics of Phase Transitions*, p. 126.

Definition – Phase Transitions

- Characterised by a critical quantity (mostly T_C)
- Classification by *latent heat* (EHRENFEST 1933)

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Phase transitions of first order

Phase transitions of first order are characterised by a discontinuity in the thermodynamic quantities of the system, e. g. entropy, volume. These correspond to discontinuous first-order derivatives of the thermodynamic potential(s).

Schematically speaking, the discontinuity corresponds to the latent heat being released or absorbed during the transition.

Definition – Phase Transitions

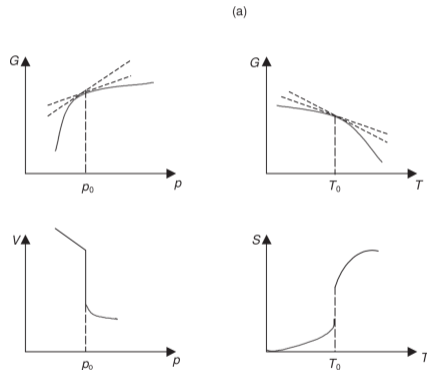


Figure: First-order phase transition.^[5]

[5] Papon, Leblond, and Meijer, *Physics of Phase Transitions*, p. 16.

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Schematically speaking, the discontinuity corresponds to the latent heat being released or absorbed during the transition.

Phase transitions of second order

Phase transitions of second order are characterised by the behaviour of the response functions of the system. The potentials and their first-order derivatives are continuous, while some of the second derivatives approach zero or infinity.

PTs of second order correspond to transitions without latent heat. The transition between the phases is continuous and no real distinction between them can be made.

Definition – Phase Transitions

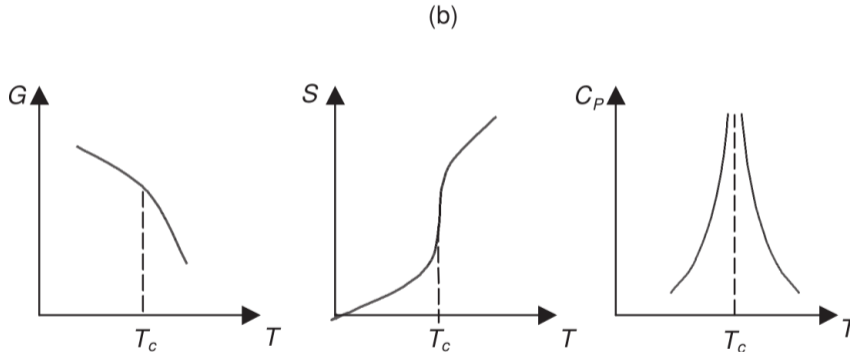


Figure: Second-order phase transition.^[5]

[5] Papon, Leblond, and Meijer, *Physics of Phase Transitions*, p. 16.

Definition – Phase Transitions

- Characterised by a critical quantity (mostly T_c)
- Classification by *latent heat* (EHRENFEST 1933)
- Classification by symmetry (LANDAU 1937)
 - Not limited to thermodynamic view
 - Occurrence of *symmetry breaking*
 - Notion of an *order parameter*, often denoted m (e. g. the magnetisation, or difference to critical density)
 - Distinction between *ordered* and *disordered* phase

Definition – Phase Transitions

PTs without order parameter

The symmetry groups of both phases are such that non is strictly included in the other. Such PTs are always first-order in Ehrenfest's sense.

PTs with order parameter

An order parameter can be defined for the system and the symmetry group of the least symmetric phase is a subgroup of the symmetry group of the most symmetric phase. At the transition the order parameter might be *discontinuous*.

This continuity distinguishes PTs of first and second order in Ehrenfest's sense.

The Ornstein-Zernike Approximation

- To account for transitions characterised by different order m the *correlation function* is of essential importance.

$$G(\mathbf{r} - \mathbf{r}') = \langle (m(\mathbf{r}) - \langle m \rangle) (m(\mathbf{r}') - \langle m \rangle) \rangle \quad (13)$$

$$= \langle m(\mathbf{r})m(\mathbf{r}') \rangle - \langle m \rangle^2 \quad (14)$$

$$= n\delta(\mathbf{r} - \mathbf{r}') + n^2\Gamma(\mathbf{r} - \mathbf{r}') \quad (15)$$

- From the correlation function the *structure function* $S(\mathbf{q})$ can be defined as its Fourier transform.

$$S(\mathbf{q}) = n \left(1 + n\hat{\Gamma}(\mathbf{q}) \right) \quad (16)$$

The Ornstein-Zernike Approximation

- The direct correlation function is indirectly defined via its Fourier transform

$$\hat{C}(\mathbf{q}) = \frac{\hat{\Gamma}(\mathbf{q})}{1 + n\hat{\Gamma}(\mathbf{q})} \quad (13)$$

- With this at hand, $S(\mathbf{q})$ becomes

$$S(\mathbf{q}) = \frac{n}{1 - n\hat{C}(\mathbf{q})} \quad (14)$$

- By Taylor expansion up to quadratic order in \mathbf{q} the correlation function can be brought in the asymptotic form for large r

$$S(q) \propto \frac{1}{\kappa^2 + q^2} \quad (15)$$

- By inverse Fourier transform the ORNSTEIN-ZERNIKE form of the correlator is obtained:

$$G(r) \propto \frac{e^{-\kappa r}}{r^{d-2}} \quad (16)$$

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- PTs are characterised by a divergence of the *correlation length* ξ and thus a loss of any relevant scale.
- This will lead to universal behaviour.
- Only scaling dimensions of the thermodynamic quantities in $(T - T_c)$ are needed for description of the system.

Critical-point Exponent

- Indices describing behaviour near critical point
- Define a dimensionless temperature variable (with $t \xrightarrow{T \rightarrow T_c} 0$)

$$t \equiv \frac{T}{T_c} - 1 \quad (14)$$

- Exponent is defined by

$$f(t) \sim t^\lambda \Leftrightarrow \lambda \equiv \lim_{t \rightarrow 0} \frac{\ln |f(t)|}{\ln |t|} \quad (15)$$

- Careful! $f \sim t^\lambda \not\Rightarrow f(t) = At^\lambda$, actually $f(t) = At^\lambda (1 + Bt^x + \dots)$
- Behaviour from below is denoted by a dash

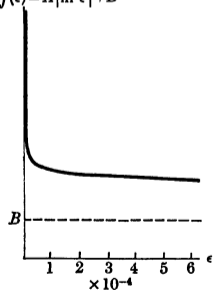
$$f(t) \sim (-t)^{\lambda'} \Leftrightarrow \lambda' = \lim_{t \rightarrow 0} \frac{\ln |f(-t)|}{\ln |t|} \quad (16)$$

Ambiguities

- The case $\lambda = 0$ can correspond to three different cases:^[6]

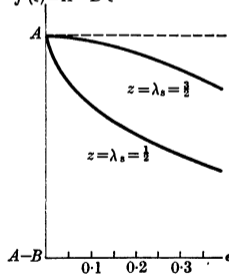
A logarithmic divergence
 ($\lambda_s = 0$)

$$f(\epsilon) = A |\ln \epsilon| + B$$



a cusp-like divergence
 ($\lambda = z$)

$$f(\epsilon) = A - B \epsilon^z$$



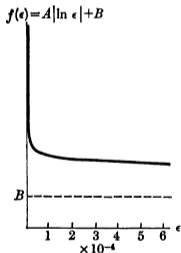
no anomalous behaviour
 (up to jump discontinuity)

[6] Stanley and Ahlers, *Introduction to phase transitions and critical phenomena*, p. 41.

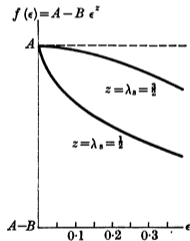
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A logarithmic divergence
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a cusp-like divergence
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no anomalous behaviour
(up to jump discontinuity)

- Correct for this by modified exponent

$$\lambda_s \equiv j + \lim_{t \rightarrow 0} \frac{\ln |f^{(j)}(t)|}{\ln t}, \quad j = \min_{k \in \mathbb{N}} \left\{ k : \left| f^{(k)}(t) \right| \xrightarrow{t \rightarrow 0} \infty \right\} \quad (17)$$

Common Exponents

Exponent	General Definition	
$\alpha^{(t)}$	$C_V \sim \begin{cases} (-t)^{-\alpha'} & t < 0 \\ t^{-\alpha} & t > 0 \end{cases}$	specific heat at V_c, p_c
β	$m \sim (-t)^\beta$	scaling of order parameter
$\gamma^{(t)}$	$\frac{\partial m}{\partial J} \sim \begin{cases} (-t)^{-\gamma'} & t < 0 \\ t^{-\gamma} & t > 0 \end{cases}$	response to a source J
$\nu^{(t)}$	$\xi \sim \begin{cases} (-t)^{-\nu'} & t < 0 \\ t^{-\nu} & t > 0 \end{cases}$	correlation length at p_c
δ	$m \propto J^{1/\delta}$	$t = 0$
η	$G(r) \sim r ^{-(d-2+\eta)}$	correlation function at $t = 0$

Common Exponents

Exponent	Definitions for a fluid system	Quantity
$\alpha^{(l)}$	$C_V \sim \begin{cases} (-t)^{-\alpha'} & t < 0 \\ t^{-\alpha} & t > 0 \end{cases}$	specific heat at V_c, p_c
β	$\rho_l - \rho_g \sim (-t)^\beta$	liquid-gas density difference at p_c
$\gamma^{(l)}$	$\kappa_T \sim \begin{cases} (-t)^{-\gamma'} & t < 0 \\ t^{-\gamma} & t > 0 \end{cases}$	isothermal compressibility at p_c
δ	$p - p_c \sim \text{sgn}(\rho_l - \rho_g) \rho_l - \rho_g ^\delta$	behaviour of the critical isotherm
$\nu^{(l)}$	$\xi \sim \begin{cases} (-t)^{-\nu'} & t < 0 \\ t^{-\nu} & t > 0 \end{cases}$	correlation length at p_c
η	$G(r) \sim r ^{-(d-2+\eta)}$	pair correlation function at $t = 0$

Universality

- Is a characteristic of second order PTs.
- Experiment and theory show that different phenomena obey the same critical behaviour and thus have the same critical exponents.
- This property is called *universality* and gives rise to *universality classes*:
 - A universality class is characterised by a set of critical exponents and contains all physical systems underlying the critical behaviour corresponding to these exponents.
 - E. g. the liquid-gas transition of CO₂ and Xe as well as the 3D Ising model share a universality class^[7] (HOCKEN/MOLDOVER 1976)
 - A VAN DER WAALS-fluid and the mean field Ising model lie in the same class

[7] Baxter, *Exactly Solved Models in Statistical Mechanics*, p. 8.

Scaling Hypothesis

- Formulated by DOMB, KADANOFF, WIDOM
- Near the critical point the thermodynamic quantities obey *scaling laws* and only the correlation length characterises the system:
 - Equation of state is left invariant under rescalings of the form $x \mapsto \lambda^\Delta x$
- The thermodynamic potential can be split in a singular part (denoted by index S) and an analytical part.
- The scaling hypothesis now states

$$\lambda G_S(t, H) = G_S(\lambda^n t, \lambda^m H) \quad (18)$$

- The scaling hypothesis and the concept of universality are not strictly coupled.

Relations between critical exponents

- The critical exponents are not independent. Their relations can be derived from the scaling laws and the intrinsic relations of the state variables through dimensional analysis.^[8]

$$\alpha' + 2\beta + \gamma' = 2 \quad (19)$$

$$\alpha' + \beta(1 + \delta) = 2 \quad (20)$$

$$\beta(\delta - 1) = \gamma \quad (21)$$

$$\nu = \nu' \quad (22)$$

$$(2 - \eta)\nu = \gamma \quad (23)$$

$$2 - \alpha = d\nu \quad (24)$$

- Only two independent exponents remain to be determined.
- Using renormalisation group technics, the critical exponents can be calculated from the dimensionality of the system d and the *degree of isotropy* (WILSON 1972, FISHER 1974).

[8] RUSHBROOKE 1963, GRIFFITH 1965, WIDOM 1965, FISHER 1967, KADANOFF 1967

Classical Critical Exponents for the Ising model in the Mean Field Approach

- Have Hamiltonian

$$\mathcal{H}[\Omega] = -\mu H \sum_i \sigma_i - J \sum_{\langle i,j \rangle} \sigma_i \sigma_j \quad (25)$$

- Expansion of the spins around their mean value $\sigma_i = m + \delta\sigma_i$ leads to:

$$\mathcal{H} = -[zJm + \mu_0 H] \sum_i \sigma_i \quad (26)$$

- From this the magnetisation can be deduced to be:

$$m = \tanh \frac{\mu_0 H + zJm}{kT} \xrightarrow{H \rightarrow 0} \tanh \frac{zJm}{kT} \quad (27)$$

- The critical temperature now is recognised as $T_c = zJ/k$.

Classical Critical Exponents for the Ising model in the Mean Field Approach

- The magnetisation in absence of an external field becomes:

$$m = \tanh \frac{zJm}{kT} \quad (25)$$

- The critical temperature now is recognised as $T_c = zJ/k$.
- Near the critical temperature, we can expand^[9]

$$m = (1+t) \operatorname{artanh} m \simeq (1+t) \left(m + \frac{1}{3}m^3 \right) \quad (26)$$

$$\Rightarrow m = (-3t)^{1/2} (1 + \mathcal{O}(t)) \quad (27)$$

- From this we conclude $\beta = \frac{1}{2}$.

^[9] Baxter, *Exactly Solved Models in Statistical Mechanics*, p. 44f.

Classical Critical Exponents for the Ising model in the Mean Field Approach

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$$m = \tanh \frac{zJm}{kT} \quad (25)$$

- The critical temperature now is recognised as $T_c = zJ/k$.
- From this we conclude $\beta = \frac{1}{2}$.
- From Eq. (25) we can deduce the CURIE-WEISS law for the susceptibility near criticality:

$$\chi \propto \left(1 - \frac{T_c}{T}\right)^{-1} \propto t^{-1} \quad (26)$$

- leading to $\gamma' = 1$.

Classical Critical Exponents for the Ising model in the Mean Field Approach

- The magnetisation in absence of an external field becomes:

$$m = \tanh \frac{zJm}{kT} \quad (25)$$

- The critical temperature now is recognised as $T_c = zJ/k$.
- We conclude $\beta = \frac{1}{2}$ and $\gamma' = 1$.
- Now from the other equalities we have $\alpha' = 0$ and $\delta = 3$.
- These values are typical for the mean field approach.
- In ONSAGER's case, the exponents are $\alpha = \alpha' = 0$, $\beta = \frac{1}{8}$, $\nu = \nu' = 1$, $\delta = 15$ and $\gamma = \frac{7}{4}$ (ABRAHAM 1973).

Outlook

- The treatment of a system near criticality as *conformal field theory* is possible
 - The critical exponents then translate to the scaling dimensions of the corresponding operators.
- The theory of PTs is not restricted to physics and chemistry.
 - E. g. models in sociology (player strategies, swing-voting)
- There are still unsolved Problems.
 - Analytical solution to the 3D Ising model
 - Mystery of the He- λ -transition exponent $\alpha = -0.0127(3)$.^[10]

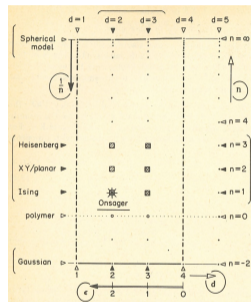


Figure: Diagram of the (d, n) -plane with position of various models.^[11]

[10] Lipa et al., “Specific heat of liquid helium in zero gravity very near the lambda point”

[11] Fisher, “The renormalization group in the theory of critical behavior”, Fig. 1

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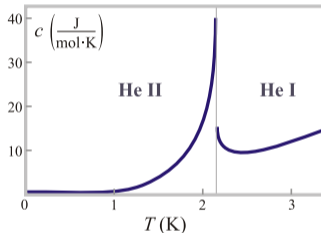







Figure: Specific heat capacity of helium.^[12]




[10] Lipa et al., “Specific heat of liquid helium in zero gravity very near the lambda point”

[12] Mpfiz, *The lambda plot[t] of the specific heat capacity of helium with lambda point*

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-  CMG Lee. *Phase diagram of water as a log-lin chart with pressure from 1 Pa to 1 TPa and temperature from 0 K to 650 K, compiled from data in <http://www.lsbu.ac.uk/water/phase.html> and <http://ergodic.ugr.es/termo/lecciones/water1.html> by CMG Lee. Note that the phases of Ice X and XI (hexagonal) differ from the diagram in <https://chem331001fall09.pbworks.com/w/page/15647320/LECTURE-10>. 2018. URL: https://upload.wikimedia.org/wikipedia/commons/0/08/Phase_diagram_of_water.svg.*

Bonus Panel - Critical Exponents from Renormalisation Group

TABLE I. Third order ϵ expansion^a for the exponent γ .

$$\begin{aligned} \gamma = 1 &+ \frac{(n+2)}{2(n+8)} \epsilon + \frac{(n+2)(n^2+22n+52)}{4(n+8)^3} \epsilon^2 \\ &+ \frac{(n+2)}{8(n+8)^3} \left[(n+2)^2 \right. \\ &\quad \left. + 24 \frac{(n+2)(n+3) - (10n+44) \zeta(3)}{(n+8)} \right. \\ &\quad \left. + 4 \frac{55n^2 + 268n + 424}{(n+8)^2} \right] \epsilon^3 + O(\epsilon^4). \end{aligned}$$

^a From E. Brézin, J. C. LeGuillou, J. Zinn-Justin, and B. G. Nickel (1973).

TABLE II. Fourth order expansion^a for the exponent η .

$$\begin{aligned} \eta = \frac{n+2}{2(n+8)^2} \epsilon^2 &+ \frac{n+2}{8(n+8)^2} \left[\frac{24(3n+14)}{(n+8)^2} - 1 \right] \epsilon^3 \\ &+ \frac{n+2}{2(n+8)^2} \left[\frac{-5n^2 + 234n + 1076}{16(n+8)^2} \right. \\ &\quad \left. - 8 \frac{3n^2 + 53n + 160 + 3(5n+22) \zeta(3)}{(n+8)^3} \right. \\ &\quad \left. + 45 \frac{(3n+14)^2}{(n+8)^4} \right] \epsilon^4 + O(\epsilon^5). \end{aligned}$$

^a From E. Brézin, J. C. LeGuillou, J. Zinn-Justin, and Nickel (1973).

Figure: First- to third-order corrections to the critical exponents from the renormalisation group. ϵ corresponds to the dimensionality of the system via $\epsilon = 4 - d$.^[13]

[13] Fisher, "The renormalization group in the theory of critical behavior"