Statistical Field Theory: Interfaces of Binary Fluid Systems

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Overview

Introduction

- Interfaces
- Phenomenology of Binary Fluid Systems

Why do we care?

- Phase Transitions and Universality
- From Ising Model to ϕ^4 -Theory

• Formalism: The Effective Potential

- Generating Functionals in Field Theory
- The Effective Potential

• The Formalism applied to Interfaces

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Interfaces





- **Phase A** Since Aristoteles' times scientists have been interested in interfaces
 - These were assumed to be *sharp* and *discontinuous* like it appears to the human eye





- Poisson (1831): Interface is a diffuse, continuous transition; it can be described by a continuous profile φ(x)
- φ(x) is e.g. the density difference of the phases



Interfaces



 \Rightarrow Definition of interface thickness is arbitrary and therefore a problem on its own

Cyclo-Hexane (C_6H_{12}) and Aniline $(C_6H_5NH_2)$



- Experiment by Atack and Rice (1953)
- Below $T_C = 30.9$ °C both fluids seperate into two pure phases
- Above T_C the fluids mix perfectly
- No latent heat is measured \Rightarrow second order phase transition

Cyclo-Hexane (C_6H_{12}) and Aniline $(C_6H_5NH_2)$



• Near T_C the reduced interface tension $\sigma = \tau/k_B T$ obeys a scaling law

$$\sigma \sim \sigma_0 t^\mu \qquad \mu = 1.26 \pm 0.01$$

with reduced temperature $t = \frac{T - T_{\rm C}}{T_{\rm C}}$

• Correlation length ξ^+ diverges at T_C according to

$$\xi^+ = \xi_0^+ t^{-
u}$$
 $u = 0.630 \pm 0.002$



Cyclo-Hexane (C_6H_{12}) and Aniline $(C_6H_5NH_2)$



Universality

- Many other BFS show the same behaviour:
 - Isobutyric acid + Water
 - Triethylamine + Water
 - ...
- Even some binary systems of one fluid and one gas behave similar

Cyclo-Hexane (C_6H_{12}) and Aniline $(C_6H_5NH_2)$



Universality

- While σ_0, ξ_0^{\pm} vary from system to system...
- ... the critical exponents μ, ν agree almost perfectly

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Continuous Phase Transitions

- We only consider phase transitions of second order
- Low temperature phase is usually more orderly than high temperature phase because of

F = U - TS F minimal at equilibrium

- Order parameter $\phi \neq 0$ only for one phase. Examples:
 - spontaneous magnetization $M_S = \frac{1}{V} \sum_i \vec{\mu_i}$ of a magnetical system
 - difference of concentrations $\Delta C = C_A C_B$ of a binary fluid system

Critical Exponents

- Near $T_{\rm C}$ many physical quantities behave according to power laws of the reduced temperature t
- Define *critical exponents* λ, λ' to physical quantity F as follows:

$$\lambda = \lim_{t \searrow 0} \frac{\ln |F(t)|}{\ln t} \qquad \lambda' = \lim_{t \nearrow 0} \frac{\ln |F(t)|}{\ln (-t)}$$
$$\Rightarrow F(t) = t^{\lambda} \quad \text{as} \quad t \to 0^+ \quad \text{and} \quad F(t) = (-t)^{\lambda'} \quad \text{as} \quad t \to 0^-$$

Remember: Phase Transitions and Universality

Critical Exponents

$$\mathsf{specific heat:} \quad \mathcal{C}_{\pi=0} \sim \left\{ egin{array}{cc} (-t)^{-lpha'} & \mathrm{for} & \mathcal{T} < \mathcal{T}_{\mathrm{C}} \ t^{-lpha} & \mathrm{for} & \mathcal{T} > \mathcal{T}_{\mathrm{C}} \end{array}
ight.$$

order parameter:
$$\phi \sim (-t)^{eta} \ (\phi = 0 \ {
m for} \ T \geq T_{
m C})$$

susceptibility:
$$\chi \sim \begin{cases} (-t)^{-\gamma'} & \text{for } T < T_{\rm C} \\ t^{-\gamma} & \text{for } T > T_{\rm C} \end{cases}$$

critical isotherm: $T \sim \phi^{\delta}$ for $T = T_{
m C}$

correlation length: $\xi \sim \left\{ \begin{array}{ll} (-t)^{u'} & {
m for} & {\cal T} < {\cal T}_{
m C} \\ t^{u} & {
m for} & {\cal T} > {\cal T}_{
m C} \end{array}
ight.$

Hypothesis of Universality (Griffiths, 1970)

- Systems with a phase transition of second order can be sorted in *universality classes*
- Crititical behaviour (i.e. the critical exponents) of systems in the same UC is essentially equal
- Classification depends on
 - a) the system's dimension D
 - b) the dimension *n* of the order parameter
 - c) the symmetries of the system

From Ising Model to ϕ^4 -Theory

How do we describe such Binary Fluid Systems theoretically?

• Values of μ, ν suggest the systems to be in the same universality class as the 3D Ising model

Ising Model

Hamiltonian (ferromagnetic)

$$H = -J\sum_{ij} S_i S_j - \mu B \sum_i S_i$$
$$S_i = \pm 1$$

Order parameter:

$$M = \frac{1}{V} \sum_{i} \left\langle S_i \right\rangle$$



How do we describe such Binary Fluid Systems theoretically?

- Values of μ, ν suggest the systems to be in the same universality class as the 3D Ising model

Ising Model

- Fluid systems: Identify
 - $S_i = 1$ ("up") with a volume cell of fluid component A
 - $S_i = -1$ ("down") with a volume cell of component B



From Ising Model to ϕ^4 -Theory

Interfaces of Ising Systems

• The Ising Model can be used to treat systems with interfaces numerically



Interfaces of Ising Systems

• The Ising Model can be used to treat systems with interfaces numerically



From Ising Model to ϕ^4 -Theory

Analytical Treatment: Landau-Ginzburg Model



- System with interface can be described by double-well potential
- Minima correspond to pure phases of the components

Analytical Treatment: Landau-Ginzburg Model

- Order parameter: Continuous variable ϕ $(-\infty < \phi < \infty)$ where $\langle \phi \rangle = M$
- Continuum: $x_i \to x$, $\phi_i \to \phi(x)$, $a \to 0$, $N \to \infty$
- Landau approximation: Neglect fluctuations $\rightarrow \phi = \langle \phi \rangle$
- Analyticity assumption: Near T_c free energy G(T, φ) is expandable in powers of φ

$$G(T,\phi) = G_0(T) - \pi\phi + \frac{1}{2}r_0(T)\phi^2 + \frac{1}{4!}u_0\phi^4 + \dots$$

(symmetry is assumed \Rightarrow no odd powers of ϕ if no external force π is present)

Analytical Treatment: Landau-Ginzburg Model

 The Ginzburg-Landau Model uses the "Hamiltonian" (valid only in vicinity of T_C)

$$\Rightarrow H_{\rm LG}[\phi] = \int d^D x \, \left[\frac{1}{2} (\nabla \phi)^2 + \frac{1}{2} r_0(T) \phi^2 + \frac{1}{4!} u_0 \phi^4 \right]$$

• Pertubation expansion terms contain statistical fluctuations

From Ising Model to ϕ^4 -Theory

Analytical Treatment: Landau-Ginzburg Model

• Partition function:

$$Z = \sum_{\{S_i\}} e^{-\beta H}$$

• Each configuration $\{S_i\}$ corresponds to a field $\phi(x)$

$$\Rightarrow Z = \int \mathscr{D}\phi(x) e^{-\beta \int d^D x \left[\frac{1}{2} (\nabla \phi)^2 + \frac{1}{2} r_0(T) \phi^2 + \frac{1}{4!} u_0 \phi^4\right]}$$

• Generating functional of correlation functions:

$$Z[B] = \int \mathscr{D}\phi(x) \, e^{-\beta H_{\rm GL} - \int d^D x \, B(x)\phi(x)}$$

From Ising Model to ϕ^4 -Theory

Comparison: Landau-Ginzburg Model vs. ϕ^4 -QFT

• Generating functional in QFT:

$$Z[J] = \int \mathscr{D}\phi \, e^{-\frac{1}{\hbar}S_{\mathrm{E}}[\phi] + \int d^{D}x \, J(x)\phi(x)}$$

• Euclidean action of $\phi^{\rm 4}\text{-}{\rm QFT}$

$$S_{\rm E}[\phi] = \int d^d x \underbrace{\left[\frac{1}{2}(\partial \phi)^2 + \frac{1}{2}m^2\phi^2 + \frac{1}{4!}g\phi^4\right]}_{\text{Lagrangean }\mathscr{L}}$$

is the formal equivalent of

$$H_{\rm GL}[\phi] = \int d^D x \underbrace{\left[\frac{1}{2}(\nabla\phi)^2 + \frac{1}{2}r_0(T)\phi^2 + \frac{1}{4!}u_0\phi^4\right]}_{\text{Hamiltonian density }\mathscr{H}}$$

Comparison: Ginzburg-Landau-Model vs. ϕ^4 -QFT

Order parameter $\phi(x) \quad \leftrightarrow \quad \text{Field } \phi(x)$ Temperatur $k_{\rm B}T = 1/\beta \qquad \leftrightarrow \qquad \text{Planck's } \hbar$ Hamiltonian density $\mathscr{H}(\phi)$ Hamiltonian $H[\phi] = \int d^D x \, \mathscr{H}(\phi(x))$ Landau approximation

Statistical fluctuations

- \leftrightarrow Lagrangean $\mathscr{L}(\phi)$
- \leftrightarrow Euclidean action

$$S_{\rm E}[\phi] = \int d^D x \, \mathscr{L}(\phi(x))$$

- \leftrightarrow Classical limit
- \leftrightarrow Quantum fluctuations

- Binary fluid systems happen to be in the same universality class as $\phi^4\text{-theory!}$
- ϕ^4 -theory is part of the Standard Model of high energy particle physics (e.g. electroweak Higgs field)
- Critical behaviour is universal \Rightarrow details of binary fluid system can be neglected, methods of QFT can be applied to BFS and vice versa
- Interfaces appear in some systems with spontaneously broken symmetry, consider e.g. domain walls in cosmology

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Generating Functional of Correlation Functions

• Addition of a source term to the Lagrangean in the functional integral yields

$$Z[J] = \int \mathscr{D}\phi \exp\left\{-\int d^D x \left[\mathscr{L} + J(x)\phi(x)\right]\right\}$$

$$\langle 0|\phi(x_1)\dots\phi(x_n)|0\rangle = \frac{1}{Z_0} \frac{\delta^n Z[J]}{\delta J(x_1)\dots\delta J(x_n)} Z[J]\Big|_{J=0}$$

mit $Z_0 = Z[J=0]$

Generating Functional of Connected Correlation Functions

• We define another generating functional by $W[J] = \ln Z[J]$

$$\frac{\delta^2 W[J]}{\delta J(x) \delta J(y)} = [\langle \phi(x) \phi(y) \rangle - \langle \phi(x) \rangle \langle \phi(y) \rangle]$$
$$= \left[- \bigcirc - + - \oslash \oslash - - - \oslash \oslash - \right]$$

$$=$$
 — \oslash $=:<\phi(x)\phi(y)>_{\mathrm{conn}}$

• More generally: $\frac{\delta^n W[J]}{\delta J(x_1) \dots \delta J(x_n)} = \langle \phi(x_1) \dots \phi(x_n) \rangle_{\text{conn}}$

The effective Action Γ

• We define the classical field $\phi_{\mathrm{Cl}}(x)$ by

$$rac{\delta W[J]}{\delta J(x)} = \langle \Omega | \phi(x) | \Omega
angle =: \phi_{\mathrm{Cl}}(x)$$

• Analogy to the free energy of statistical mechanics: Legendre transformation of W[J] yields $\Gamma[\phi_{\rm Cl}(x)]$

$$\delta W[J] = \int d^4 x \, \frac{\delta W[J]}{\delta J(x)} \delta J(x) = \int d^4 x \, \phi_{\rm Cl}(x) \delta J(x)$$
$$= \int d^4 x \, \delta(\phi_{\rm Cl}(x)J(x)) - \int d^4 x \, J(x) \delta \phi_{\rm Cl}(x)$$
$$\Rightarrow \underbrace{\delta(W[J] - \int d^4 x \, \phi_{\rm Cl}(x)J(x))}_{\delta \Gamma[\phi_{\rm Cl}]} = -\int d^4 x \, J(x) \delta \phi_{\rm Cl}(x)$$

The effective Action Γ

• Definition of $\Gamma[\phi_{\rm Cl}]$ yields

$$\frac{\delta \Gamma[\phi_{\rm Cl}]}{\delta \phi_{\rm Cl}(x)} = -J(x)$$

 \Rightarrow In abscence of external sources Γ becomes stationary

- \Rightarrow equation of motion for VEV $\phi_{\rm Cl}(x) = \langle \phi(x) \rangle_{J=0}$
- It can be shown that

$$\frac{\delta^2 \Gamma[\phi_{\rm Cl}]}{\delta \phi_{\rm Cl}(x) \delta \phi_{\rm Cl}(y)} = -G_{\mathcal{C}}^{-1}(x,y)$$

The effective Action Γ

+ $\Gamma[\phi_{\rm Cl}]$ is the generating functional of the proper vertices

$$\frac{\delta^n \Gamma[\phi_{\rm Cl}]}{\delta \phi_{\rm Cl}(x_1) \dots \delta \phi_{\rm Cl}(x_n)} = \langle \phi(x_1) \dots \phi(x_n) \rangle_{\rm 1PI}$$

• This is proved by induction from *n*-point to (n + 1)-point correlation functions and explicit calculation for the 3-point correlator

$$y = \frac{\delta^3 W[J]}{z} = \langle \phi(x)\phi(y)\phi(z) \rangle_{\text{conn}} = \frac{\delta^3 W[J]}{\delta J_x \delta J_y \delta J_z}$$

$$= \int d^4 w \ G_{zw} \frac{\delta}{\delta \phi_w^{\text{Cl}}} \frac{\delta^2 W[J]}{\delta J_x \delta J_y} = -\int d^4 w \ G_{zw} \frac{\delta}{\delta \phi_w^{\text{Cl}}} \left(\frac{\delta^2 \Gamma[\phi_{\text{Cl}}]}{\delta \phi_x^{\text{Cl}} \delta \phi_y^{\text{Cl}}} \right)^{-1}$$

$$= \int d^4 w \ d^4 u \ d^4 v \ G_{zw} \left(\frac{\delta^2 \Gamma[\phi_{\text{Cl}}]}{\delta \phi_x^{\text{Cl}} \delta \phi_u^{\text{Cl}}} \right)^{-1} \frac{\delta^3 \Gamma[\phi_{\text{Cl}}]}{\delta \phi_w^{\text{Cl}} \delta \phi_v^{\text{Cl}} \delta \phi_y^{\text{Cl}}} \left(\frac{\delta^2 \Gamma[\phi_{\text{Cl}}]}{\delta \phi_v^{\text{Cl}} \delta \phi_y^{\text{Cl}}} \right)^{-1}$$

$$= \int d^4 u \ d^4 v \ d^4 w \ G_{xu} \ G_{vy} \ G_{zw} \frac{\delta^3 \Gamma[\phi_{\text{Cl}}]}{\delta \phi_u^{\text{Cl}} \delta \phi_v^{\text{Cl}} \delta \phi_w^{\text{Cl}}}$$

$$= \bigvee_{y} \qquad z$$

• Induction hypothesis

$$\langle \phi(x_1)...\phi(x_n) \rangle_{\text{conn}} = \frac{\delta^n W[J]}{\delta J(x_1)...\delta J(x_n)}$$

$$= \int d^4 y_1 d^4 y_2...d^4 y_n \left\{ G_{x_1y_1} G_{x_2y_2}...G_{x_ny_n} \frac{\delta^n \Gamma[\phi_{\text{Cl}}]}{\delta \phi_{y_1}^{\text{Cl}} \delta \phi_{y_2}^{\text{Cl}}...\phi_{y_n}^{\text{Cl}}} + ... \right\}$$

where the ... terms consist of some $G_{x_iy_i}$ and some $\frac{\delta^m\Gamma[\phi_{\mathrm{Cl}}]}{\delta\phi_{y_i}^{\mathrm{Cl}}\delta\phi_{y_j}^{\mathrm{Cl}}...}, m < n$ and the derivatives of Γ describe proper vertices

Inductive step: Addition of an external point

$$\langle \phi(\mathbf{x}_1) \dots \phi(\mathbf{x}_{n+1}) \rangle_{\text{conn}} = \frac{\delta}{\delta J_{\mathbf{x}_{n+1}}} \left(\frac{\delta^n W[J]}{\delta J_{\mathbf{x}_1} \dots \delta J_{\mathbf{x}_n}} \right)$$

• This yields two types of terms:



• Terms of type I:

- In each of the type I terms one external line is replaced by a three-point function
- Terms of type II:

$$\frac{\delta}{\delta J_{x_k}} \left(\frac{\delta^m \Gamma[\phi_{\rm Cl}]}{\delta \phi_{y_i}^{\rm Cl} \phi_{y_j}^{\rm Cl} \dots} \right) = \int d^4 y_k \ G_{x_k y_k} \frac{\delta^{m+1} \Gamma[\phi_{\rm Cl}]}{\delta \phi_{y_i}^{\rm Cl} \phi_{y_j}^{\rm Cl} \dots \phi_{y_k}^{\rm Cl}}$$

 \Rightarrow a new external line is added to what used to be a $\mathit{m}\text{-point}$ proper vertex



Graphical representation of step $n \rightarrow (n+1)$:





The effective Action Γ

- Γ is the generating functional of the proper vertices!
- Its second functional derivative is the inverse propagator
- Effective action Γ contains all the physics of the corresponding theory!

The Effective Potential

Definition of the Effective Potential

- Definition of Γ yields $\frac{\delta \Gamma[\phi_{\rm Cl}]}{\delta \phi_{\rm Cl}} = 0$ for J = 0
- Solutions are the stable VEV: $\phi_{\mathrm{Cl}}(x) = \langle \phi(x) \rangle$
- Expansion of Γ in ϕ_{Cl} and derivatives:

$$-\Gamma[\phi_{\rm Cl}] = \int d^4x \, \left[V_{\rm eff}(\phi_{\rm Cl}) + \frac{1}{2} (\partial_\mu \phi_{\rm Cl})^2 Z(\phi_{\rm Cl}(x)) + \dots \right]$$

• We consider systems with Lorentz invariance and conservation of momentum

$$\Rightarrow \phi_{\rm Cl}(x) = \phi_0 = {\rm const.}$$

• From above expansion only the first term remains:

$$\Gamma[\phi_0] = -\int d^4x \, V_{\mathrm{eff}}(\phi_0) = -V_{\mathrm{eff}}(\phi_0) \int d^4x = -V_{\mathrm{eff}}(\phi_0) \Omega$$

The Effective Potential

What is the Physical Meaning of $V_{\rm eff}$?

• Variation of above expansion yields

$$-\delta\Gamma[\phi_{\rm Cl}] = \int d^4x \, \left[\frac{\partial V_{\rm eff}}{\partial \phi_{\rm Cl}} \delta \phi_{\rm Cl} + \delta(\text{terms containing derivatives}) \right]$$

$$\Rightarrow \frac{\delta \Gamma[\phi_{\rm Cl}]}{\delta \phi_{\rm Cl}} = -\frac{\partial V_{\rm eff}}{\partial \phi_{\rm Cl}} \bigg|_{\phi_0}$$

• It can then be shown that

$$\left. \frac{\partial^2 V_{\text{eff}}}{\partial \phi_{\text{Cl}}^2} \right|_{\phi_0} = m_{\text{phys}}^2 \quad \text{and} \quad \left. \frac{\partial^4 V_{\text{eff}}}{\partial \phi_{\text{Cl}}^4} \right|_{\phi_0} = g$$

Advantages of the Effective Potential Formalism

- If we replace the classical potential by $V_{\rm eff}$ we can apply methods of classical SSB theory to quantum systems
- It can be shown that the divergence structure of a renormalisable theory is not affected by the occurrence of SSB
- Coleman: "Secret symmetry buys us secret renormalizability."

Calculation of the Effective Potential

- For non-trivial models $V_{
 m eff}$ cannot be calculated exactly
- It can be approximated by loop expansion
- The zero-loop term is the classical potential \boldsymbol{U}

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• Action functional of ϕ^4 -Theory

$$S[\phi] = \int d^{D}x \left\{ \frac{1}{2} (\partial \phi)^{2} + U(\phi) \right\} = \int d^{D}x \left\{ -\frac{1}{2} \phi \partial^{2} \phi + U(\phi) \right\}$$
$$U(\phi) = \frac{m^{2}}{2} \phi^{2} + \frac{g}{4!} \phi^{4}$$

• Classical interface profiles obey

$$rac{\delta \mathcal{S}[\phi]}{\delta \phi} = -\partial^2 \phi + U'(\phi) = 0$$

$$U'(\phi) = m^2 \phi + \frac{g}{3!} \phi^3$$

• Assumption:
$$\phi = \phi(x_3)$$

• Multiply equation of motion by $\partial \phi$, integrate over x_3

$$-\frac{1}{2}(\partial\phi)^2 - \frac{m^2}{4}\phi^2 + \frac{g}{4!}\phi^4 = C$$

• Boundary conditions:

$$\phi(x_3) = \begin{cases} v_0, & \text{as} \quad x_3 \to +\infty \\ -v_0, & \text{as} \quad x_3 \to -\infty \end{cases}$$



Classical "kink" solution $\phi_{\rm Cl}(x_3) = v_0 \tanh\left(rac{m}{2}(x_3-a)
ight)$

• Replacing U by $V_{\rm eff}$ yields the equation for interface profiles with loop corrections

$$-\frac{1}{Z_3}\partial^2\phi+V_{\rm eff}'(\phi)=0$$

- Assumption: $\phi = \phi(x_3)$
- Multiply by $\partial \phi$, integrate over x_3

$$\Rightarrow -rac{1}{2Z_3}(\partial\phi)^2 + V_{\mathrm{eff}}(\phi) = C$$

• $V_{\rm eff}$ can be normalized to get C=0

$$(\partial \phi)^2 = 2Z_3 V_{\text{eff}}(\phi)$$

 $\partial \phi = \sqrt{2Z_3 V_{\text{eff}}(\phi)}$

We use ∂φ = +√... in our calculations and retrieve the solution with negative sign simply by substituting x₃ by −x₃ in our results

Results to second order (Küster, 2001)



Outlook

- The interface profile is to be calculated
- Küster for example approximated the profile by using the effective potential $V_{\rm eff}$, which is defined for constant $\phi_{\rm Cl}$ to get a differential equation for a non-constant profile ϕ
- Instead of $V_{\rm eff}$ we are now going to use $\Gamma[\phi_{\rm Cl}]$ with the classical kink solution $\phi_{\rm Cl}$
- The first orders of the loop expansion of Γ will be used to retrieve a new differential equation for the profile ϕ
- The resulting profile will hopefully respect long-wavelength fluctuations a phenomenon known as interface roughening which are ignored by approximations using $V_{\rm eff}$

Summary

- Interfaces in binary fluid systems can be described by a continuous order parameter field
- BFS belong to the same universality class as the 3D Ising Model and the Ginzburg-Landau Model ($\phi^4\text{-Theory})$
- The statistical $\phi^4\text{-}\mathsf{Theory}$ is formally equal to the $\mathit{euclidean}$ $\phi^4\text{-}\mathsf{QFT}$
- The effective action Γ is the Legendre-Transform of the generating functional of connected correlation functions
- For a constant ϕ_{Cl} an effective potential V_{eff} is defined

Summary

- The transition from classical to quantum field theory can be done by subsituting the classical potential U with the effective potential $V_{\rm eff}$
- This equals the transition from statistical field theory in Landau approximation to a theory with fluctuations
- + $V_{\rm eff}$ can be used to formulate a differential equation for the interface profile
- Instead we will use $\mathrm{F}[\phi_{\mathrm{Cl}}]$ with the classical kink solution ϕ_{Cl} to calculate the profile