Phase transitions as catastrophes: The tricritical point

J. A. Gaite
Grupo de Física Teórica and Departamento de Física Aplicada, Universidad de Salamanca, Salamanca 37008, Spain
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The formulation of phase transitions in the framework of catastrophe theory is studied, taking as a reference the tricritical two-fluid system. Adding renormalization-group ideas, a general approach to its classification is explored.

I. INTRODUCTION

In spite of long-standing allusions to catastrophe theory in the extensive literature on critical phenomena there does not appear to be a thorough investigation on the formal equivalence between them (however, see Ref. 1). The usual argument goes as follows. The equilibrium states of a thermodynamical system can be described as the extrema of the thermodynamical potential (grand-canonical potential per unit volume):

$$g(T,\mu_i;x_i)=f(T,x_i)-\sum_i \mu_i x_i \tag{1}$$

where f is the Hemholtz potential depending on the temperature and the generalized displacements (volume or density, concentrations of the various components) and the generalized forces for thermodynamical fields (pressure or chemical potential).

The extremum conditions $\partial g/\partial x_i=0$ give the equations of state relating the values of the generalized displacements to the external fields $x_i(T,\mu_j)$. For some range of these fields there may be several values of the generalized displacements minimizing g, corresponding to distinct phases of the system.

Catastrophe theory provides a general setting for classifying the singularities of families of functions depending on several parameters which appear when the critical point structure of a function changes. This occurs in the so-called bifurcation manifold. When it is applied to g, one is interested only in the manifolds where local minima, corresponding to metastable states, appear or disappear.

The catastrophes having one or two variables (co-rank 2) and at most five control parameters are well studied; they constitute the seven elementary catastrophes of Thom.[2]

The relation between the cusp catastrophe and the usual one-component liquid-vapor phase transition has been analyzed in Refs. 3 and 4. The butterfly catastrophe as a model for the ferromagnetic or ferroelectric phase transition was proposed in Ref. 5. Here, we deal with the general two-fluid mixture (see Ref. 6 for a wide covering of its thermodynamical properties). The Gibbs phase rule allows it to have at most four coexisting phases (quadruple point), two liquid and two vapor, that occur for unique value of the fields. However, our main concern will be the tricritical point (TCP), where three critical lines meet and two phases disappear. It can be described with the next to the cusp even codimension, co-rank-1 catastrophe, the butterfly, or universal unfolding of x^6, as we shall argue below (Sec. II). Nevertheless, some features could demand the use of co-rank-2 catastrophes. This case is briefly commented in Sec. III.

The generalization to many-component systems may be made along the same lines. In Sec. IV some results of renormalization-group (RG) theory are used to conjecture how the catastrophe classification may relate systems in different dimensions and connect to other realms of theoretical physics.

II. THE CONDITIONS FOR THE TRICRITICAL POINT AND LOWER-ORDER SINGULARITIES

Let us first consider the type-1 phase transition in the light of catastrophe theory. The Gibbs potential of the mole of fluid

$$g(T,P;v)\equiv f(T,v)+Pv,\quad v\equiv V/N,\quad g\equiv G/N\ , \tag{2}$$

gives the equilibrium value of the volume $\bar{v}=v(T,P)$ constrained by the external pressure (and temperature), from the minimum conditions

$$\left.\frac{\partial g}{\partial v}\right|_{T,P}=0,\quad \left.\frac{\partial^2 g}{\partial v^2}\right|_{T,P}>0\ , \tag{3}$$

$$g(T,P)\equiv g(T,P;v(T,P))\ . \tag{4}$$

Wherever several minima appear, the stable phase $\bar{v}$ corresponds to the global one while the others (two at most) are metastable. The expansion of g around $\bar{v}$ allows one to classify its critical points for the range of (T,P) where they are close enough to consider such an expansion reliable:

$$g(T,P;v)=g(T,P)+(\tfrac{1}{2})f_{vv}(\bar{v})(v-\bar{v})^2+(1/3!)f_{vvv}(\bar{v})(v-\bar{v})^3+\cdots\ . \tag{5}$$

This happens near the critical point (CP), defined by

$$f_{vv}=0,\quad f_{vvv}=0,\quad f_{vvvv}>0\ . \tag{6}$$

So, no higher order than the fourth is necessary and (5) reduces to the usual polynomial of the cusp catastrophe.[3-5] Nevertheless, a difference must be noticed: While in catastrophe theory the freedom to choose the origin of the state variable is used to make null the third-order coefficient; here, it is the first-order coefficient which naturally turns out to be null.[7,4] Had we also expanded the fields around the CP (T_c,P_c), we would have obtained the Landau polynomial with $v-v_c$ as the order parameter,[8] suitable for applying directly to it the results of catastrophe theory.

The bifurcation set (the cusp) that separates the instability from the stability zone in the $T-P$ field space is obtained following the standard method,[9] by projection of the fold curve

$$g_{vv}(T,P;\bar{v})=0\ , \tag{7}$$

that is, defined in parametric form, with $\bar{v}$ as the parameter, by

$$f_v(T,\bar{v})=P\ , \tag{8a}$$

$$f_{vv}(T,\bar{v})=0\ . \tag{8b}$$

The second equation gives $T(\bar{v})$, the spinodal curve, which substituted into the first also gives $P(\bar{v})$. Both are the explicit parametric equations of the bifurcation curve. The temperature and pressure play the role of the splitting and normal factors in the terminology of Zeeman.[9]

The cusp CP is found when

$$\frac{dP}{d\bar{v}}=\frac{dT}{d\bar{v}}=0 \tag{9}$$

which can be shown to be equivalent to

$$f_{vvv}(t,\bar{v})=0\ , \tag{10}$$

provided that Eqs. (8) are satisfied.

For the type-3 phase transition of the two-fluid model, the potential to be used is

$$\Omega(T,P,\mu_1;v,x_1)\equiv f(T,v,x_1)+Pv-\mu_1x_1$$
$$=g(T,P;v,x_1)-\mu_1x_1 \tag{11}$$

where x_i, μ_i are the concentrations and chemical potentials of the fluids. Again, the conditions

$$\left.\frac{\partial\Omega}{\partial v}\right|_{T,P,\mu_1,x_1}=0,\quad \left.\frac{\partial^2\Omega}{\partial v^2}\right|_{T,P,\mu_1,x_1}>0 \tag{12}$$

give the mechanical equation of state $\bar{v}=v(T,P,x_1)$, which substituted back into (11) provides a new potential $\Gamma(T,P,\mu_1;x_1)\equiv g(T,P;x_1)-\mu_1x_1$, suitable to study the critical and tricritical behavior of the system in terms of the Gibbs potential $g(T,P;x_1)$.[6] Analogously, the conditions

$$\left.\frac{\partial\Gamma}{\partial x_1}\right|_{T,P,\mu_1}=0,\quad \left.\frac{\partial^2\Gamma}{\partial x_1^2}\right|_{T,P,\mu_1}>0 \tag{13}$$

give the chemical equation of state $\bar{x}_1=x_1(T,P,\mu_1)$, which substituted back in Γ originates the fundamental relation for the system $\mu_2(T,P,\mu_1)$ through the thermodynamical identity.[10]

The expansion around $\bar{x}_1$ up to sixth order,

$$\Gamma(T,P,\mu_1;x_1)=(1-\bar{x}_1)\mu_2(T,P,\mu_1)$$
$$+\tfrac{1}{2}g_{xx}(\bar{x}_1)(x_1-\bar{x}_1)^2+\cdots$$
$$+(1/6!)g_{VI}(\bar{x}_1)(x_1-\bar{x}_1)^6+\cdots\ , \tag{14}$$

is enough to describe the variety of phase behavior of known fluid mixtures.[11] This fact finds its natural explanation if the butterfly catastrophe is able to fit that behavior. We shall show that this is indeed the case.

The butterfly catastrophe is usually analyzed through the bifurcation set as a function of the four polynomial coefficients.[9] That procedure may be applied to the sixth-order Landau polynomial, with coefficients g_x, g_{xx}, g_{xxx}, g_{IV}, calculated on the TCP (rather with g_x instead of g_V). The highest codimension singularity, the TCP, occurs when they are all null. Lower codimension singularities occur when certain relations among them are fulfilled, giving singular submanifolds of the bifurcation set. The analysis in terms of thermodynamical fields can be done generalizing what has already been seen for the ordinary CP. Now, the normal factor is the field for $\bar{x}_1$, the chemical potential μ_1. We have to identify the butterfly and bias factors.

The condition $g_{xx}(T,P;\bar{x})=0$ yields the spinodal surface $T(P,\bar{x})$, which is transferred to field space eliminating $\bar{x}$ through the equation of state $g_x(T,P;\bar{x})=\mu$. (Hereafter, the subscript 1 is suppressed without occasioning any confusion). The resulting bifurcation surface $T(P,\mu)$ contains the critical lines, or sets of critical points for every $P(T(P),\mu(P))$ solution of the equations

$$\left.\frac{\partial\mu}{\partial\bar{x}}\right|_P=\left.\frac{\partial T}{\partial\bar{x}}\right|_P=0 \tag{15}$$

or

$$g_{xxx}(T,P;\bar{x})=0\ . \tag{16}$$

These equations, depending on the value of P, can have 1 or 3 solutions. At high pressure, only the liquid phase exists and the critical point marks the end of the inmiscibility zone (Fig. 1).

At lower pressures the vapor phase appears and there

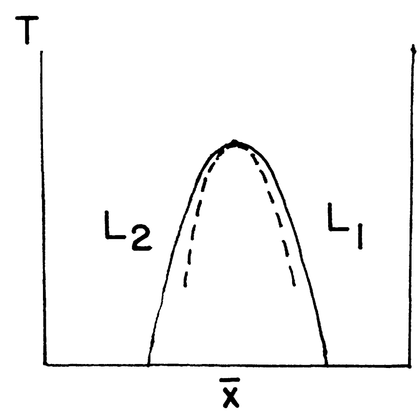

FIG. 1. Phase diagram for high-pressure liquid mixture. The solid and dashed lines represent the coexistence and spinodal curves.

are two liquid-vapor CP's for both liquids, and a third one signaling the extinction of the vapor as a metastable phase, which occurs at temperature below that of the triple point.[8] This is illustrated in Fig. 2, which implies some extrapolation from the usual way to draw the phase diagram.[8]

Both diagrams can be produced by projecting the equilibrium surface of the butterfly catastrophe on the $(T,\bar{x})$ plane, for different values of the butterfly factor.[9] Therefore, we can conclude that P acts as the butterfly factor. For certain value of P, P_C, the lower and one of the upper critical lines join and these two critical points are no more present for $P>P_C$. This is the unstable critical point.[6,12]

In order to obtain P_C, let us consider the critical lines in field space as given by condition (16). It is conveniently rewritten, defining the Gibbs potential restricted to the spinodal $g'(P,\bar{x})$, as

$$g'_{xxx}(P,\bar{x})\equiv g_{xxx}(T(P,\bar{x}),P;\bar{x})=0\ , \tag{17}$$

and yields $P(\bar{x})$, which upon substitution in $T(P,\bar{x})$ and $\mu'(P,\bar{x})\equiv\mu(T(P,\bar{x}),P;\bar{x})$, gives three functions constituting the critical line in parametric form:

$$\begin{aligned}&P(\bar{x})\ ,\\ &\mu''(\bar{x})\equiv\mu'(P(\bar{x}),\bar{x})\ ,\\ &T'(\bar{x})\equiv T(P(\bar{x}),\bar{x})\ .\end{aligned} \tag{18}$$

The unstable critical point (UCP), regarded as a turning point (two lines meet), is found for the value of $\bar{x}$ satisfying

$$\frac{dP(\bar{x})}{d\bar{x}}=0\ . \tag{19}$$

It is a cusp point, because this condition also implies (still following the methods of Ref. 9)

$$\frac{d\mu''(\bar{x})}{d\bar{x}}=\left.\frac{\partial\mu'}{\partial\bar{x}}\right|_P+\left.\frac{\partial\mu}{\partial P}\right|_x\frac{dP(\bar{x})}{d\bar{x}}=0\ , \tag{20}$$

$$\frac{dT'(\bar{x})}{d\bar{x}}=\left.\frac{\partial T}{\partial\bar{x}}\right|_P+\left.\frac{\partial T}{\partial P}\right|_x\frac{dP(\bar{x})}{d\bar{x}}=0 \tag{21}$$

where it has been taken into account that, on the critical lines, Eqs. (15) are fulfilled. Hence, the general aspect of the critical lines in the vicinity of the UCP has the aspect shown in Fig. 3.

Not surprisingly, P_C can also be calculated from the vanishing of the fourth derivative of the Gibbs potential, as is done in Ref. 12. The equivalence to the former is established from the fact that on the critical lines

$$\begin{aligned}0=\frac{dg'_{xxx}}{d\bar{x}}&=\left.\frac{\partial g'_{xxx}}{\partial\bar{x}}\right|_P+\left.\frac{\partial g'_{xxx}}{\partial P}\right|_x\frac{dP(\bar{x})}{d\bar{x}}\\ &=g_{IV}+\left.\frac{\partial g_{xxx}}{\partial T}\right|_{P,x}\left.\frac{\partial T}{\partial x}\right|_P\\ &\quad+\left[\left.\frac{\partial g_{xxx}}{\partial P}\right|_{T,x}+\left.\frac{\partial g_{xxx}}{\partial T}\right|_{P,x}\left.\frac{\partial T}{\partial P}\right|_x\right]\frac{dP(\bar{x})}{d\bar{x}}\ .\end{aligned} \tag{22}$$

The identification of the bias factor is more involved. As pointed out by Ref. 5, there does not seem to be any apparent thermodynamical field associated with it. However, the existence of a TCP requires a relation among the interaction parameters defining a concrete two-fluid model. This is shown, for instance, in Ref. 12. Whether that relation defines a thermodynamical field or not amounts to determining its possible experimental meaning, and is immaterial for our purposes. However, we can tell that the necessary field should be able to change those interaction parameters, perhaps through electric or magnetic means.

Let us call w the interaction parameter controlling the appearance of tricritical behavior. In the mean field model of Refs. 12 and 13 it could be defined as w_1-w_2, the difference between specific energies of both fluids, or more generally, the eigenvalues of the Hessian matrix.

Equation (17), giving the critical lines, should now include w:

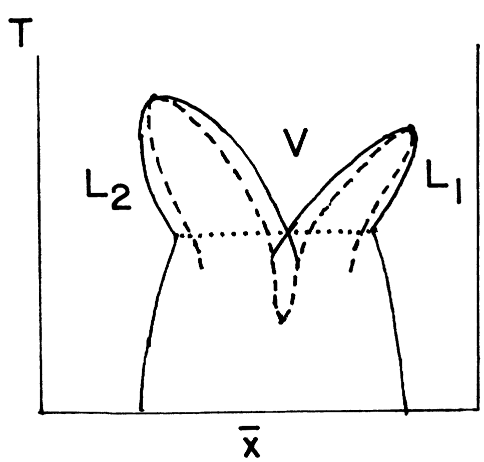

FIG. 2. Phase diagram with three phases. The solid and dashed lines represent the coexistence and spinodal curves. The dotted line corresponds to the triple point.

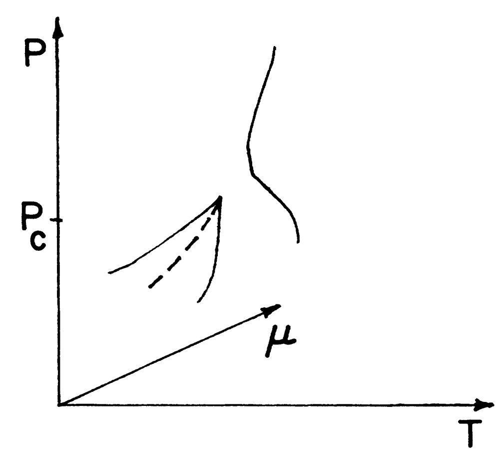

FIG. 3. Neighborhood of the UCP in field space. The critical lines are drawn solid and the triple-point line dashed.

$$g'_{xxx}(w,P,\bar{x})=0 \ . \tag{23}$$

This equation, having two control parameters and one or three solutions, depending on their value, is analogous to (8a) and is also described by the cusp catastrophe. Its bifurcation set is the curve formed by the unstable critical points, as a function of g_{iv}, w, $P_C(w)$, the solution of (23) and $g'_{IV}=0$. The point on it for where the three solutions of (23) simultaneously meet is its cusp CP, given by the further condition $g'_V=0$. Since these solutions represent the critical lines, this is the condition for the TCP, or, equivalently, $g_V=0$, provided that the former derivatives are also null. Again, it coincides with Ref. 12.

III. CO-RANK-2 CATASTROPHES: THE DOUBLE CUSP

Now we want to consider critical points with a vanishing Hessian matrix. They are physically realized in case of critical azeotropy (Ref. 6, p. 210). Then, the Legendre transform eliminating one state variable (v) is no more possible and we have to expand the potential (11).

$$\begin{aligned}\Omega(T,P,\mu_1;v,x_1) &\equiv f(T,v,x_1)+Pv-\mu_1x_1\\ &=(1-\bar{x}_1)\mu_2(T,P,\mu_1)+\tfrac{1}{2}f_{vv}(\bar{v})(v-\bar{v})^2+\tfrac{1}{2}f_{xx}(\bar{x}_1)(x_1-\bar{x}_1)^2+f_{vx}(v-\bar{v})(x_1-\bar{x}_1)\\ &\quad+(1/3!)f_{vvv}(\bar{v})(v-\bar{v})^3+(1/3!)f_{xxx}(\bar{x}_1)(x_1-\bar{x}_1)^3+\cdots\end{aligned} \tag{24}$$

and deal with co-rank-2 catastrophes. Again, they will not appear in the standard form, which does not have terms $\partial\Omega/\partial v$, $\partial\Omega/\partial x$, but rather in one without linear terms, already centered in the minimum (the codimension is not altered).

This case was considered by Griffiths and coworkers[13,14] using as independent state variables instead of (v,x_1) the densities (ρ_1,ρ_2), which give a more symmetrical description. However, the possibility of codimension less than 7 was dismissed. This is the codimension of the double cusp catastrophe X_9, allowing for four-phase coexistence.[14,1] Its utility, specially that of the compact type x^4+y^4, has been emphasized by Ref. 15 where its universal unfolding is also analyzed, giving the possible four co-rank-2 strata (abutments in Ref. 1): the three umbilic uses and one exceptional stratum x^3, noted X. The corresponding phase structure is shown in Ref. 1. It is noticeable that the lowest codimension catastrophe in that list being abutted by A_5 (butterfly) is the exceptional E_6.

In order to obtain the bifurcation set, a procedure similar to that used for the butterfly catastrophe can be followed: Using the vanishing Hessian condition $H=0$ both state variables (v,x_1) are put as functions of unique parameter that gives through the equation of state the bifurcation curve in the (P,μ_1) plane for constant values of the remaining fields. Nevertheless, the general methods quoted in Ref. 16 (p. 43) are more appropriate.

The bifurcation set [spinodal surface in the (T,v,x) space] is characterized by the vanishing Hessian

$$H=\begin{vmatrix}\dfrac{\partial P}{\partial v} & \dfrac{\partial P}{\partial x}\\[2mm] \dfrac{\partial \mu}{\partial v} & \dfrac{\partial \mu}{\partial x}\end{vmatrix}=0 \tag{25}$$

so that the rank of the map $(v,x)\rightarrow(P,\mu)$ given by the equations of state reduces to 1 or what is the same, and has a nontrivial kernel whose equation is

$$\delta P=\frac{\partial P}{\partial v}\delta v+\frac{\partial P}{\partial x}\delta x=0 \tag{26}$$

(or the one with μ instead of P).

The critical point is a secondary singularity, defined by the fact tha the map restricted to the spinodal, which has, in principle, rank 2, degenerates again on it. Equivalently, there, the tangent plane to the spinodal includes the former kernel, so that the equations

$$\delta P=\frac{\partial P}{\partial v}\delta v+\frac{\partial P}{\partial x}\delta x=0 \ , \tag{27}$$

$$\delta H=\frac{\partial H}{\partial v}\delta v+\frac{\partial H}{\partial x}\delta x=0 \tag{28}$$

are equivalent and

$$\Delta\equiv\begin{vmatrix}\dfrac{\partial P}{\partial v} & \dfrac{\partial P}{\partial x}\\[2mm] \dfrac{\partial H}{\partial v} & \dfrac{\partial H}{\partial x}\end{vmatrix}=0 \ . \tag{29}$$

This form of the equation for the critical lines is due to Gibbs according to Ref. 12.

IV. HOMOGENEITY PROPERTIES AND SYMMETRY OF THE LANDAU POLYNOMIAL

It has been known for some time that the Landau theory of multicritical phenomena is exact in dimension $d_n=2n/n-1$ where $n=2,3,\ldots$ is the criticality index.[17,18] In said dimension, the potential is

$$\Gamma=\mu_1x+\cdots+\mu_{2n-2}x^{2n-2}/(2n-2)!+x^{2n}/(2n)! \ . \tag{30}$$

For every value d_n a bifurcation occurs and when $d_{n+1}<d<d_n$, the nontrivial critical exponents can be calculated by perturbative field theory methods.[19]

From the point of view of the scaling theory of critical phenomena,[20] multicritical points in every dimension are described by a thermodynamical potential which is a generalized homogeneous function of its arguments.[21] Therefore, it is tempting to assume that the Landau polynomial is still valid for nontrivial fixed points of the renormalization group, although its arguments no longer represent the physical order parameter and fields but rather some powers of them. This is admissible in catastrophe theory, where the potential is defined up to diffeomorphisms.[5] In other words, one can put forward the hypothesis that the bifurcation branches at d_n are prolongable down to $d=2$. In this special dimension, there are powerful methods to classify the possible types of critical behavior[22] based on the infinite two-dimensional conformal symmetry.[23] Some authors[24] have conjectured that this classification can indeed be identified with the Landau models (30). However, a study of these models with high criticality index, which should have a reliable ϵ expansion near $d=2$, has found some discrepancies concerning anomalous dimensions, yet to be explained.[25]

The generalization to the Landau models with several order parameters has been proposed in Refs. 26 and 27. The key to the classification is the external symmetry group for the multicritical point.[8] For every group there exists a series of models labelled by its criticality index, realizing that symmetry.[28] The $\mathbb{Z}_2$ series gives the one-order-parameter models of Zamolodchikov. These are the A models of the ADE classification,[27] which, at a time, is the simplest case, for it has modality zero, that is, no marginal couplings.[26]

The class of quasihomogeneous singularities[16] provides the most general Landau polynomials representing the expansion of a generalized homogeneous function near its top criticality point.

The two-dimensional conformal group and its extensions, Kac-Moody, W_N symmetry (see Ref. 29 for a review), shed new light on some old problems. For instance, given the Landau polynomial, one can find its symmetry group. Also, they allow one to make contact with string theory,[30] concretely with the ground-state problem, which is currently formulated in the renormalization-group language in the space of two-dimensional statistical models.[31]

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