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Optimization of chemical batch reactors using temperature control

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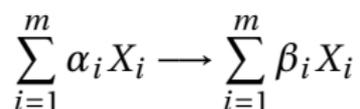
joint work with B. Bonnard (INRIA & UBFC)

Chemical Networks with mass action kinetics

Graph Model :

Species $\{X_1, \dots, X_m\}$.

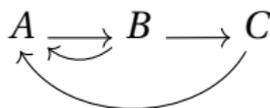
Notations : \mathcal{R} is the set of reactions of the form :



α_i, β_i are stoichiometric coefficients.

Feinberg-Horn-Jackson graph

- Vertices : $\mathbf{y} = (\alpha_1, \dots, \alpha_m)^\top$, $\mathbf{y}' = (\beta_1, \dots, \beta_m)^\top$
- Orientation : $\mathbf{y} \rightarrow \mathbf{y}'$



(1)

Rate dynamics $\mathbf{y} \rightarrow \mathbf{y}'$ (Mass kinetics)

$$K(\mathbf{y} \rightarrow \mathbf{y}') = k(T) c^{\mathbf{y}}$$

- $k(T) = A \exp(-\frac{E}{RT})$: Arrhenius law
 E, A parameters, T temperature and R is the gas constant
- $\mathbf{c} = (\mathbf{c}_1, \dots, c_m)^\top$
 c_i : concentrations of the species X_i with

$$c^{\mathbf{y}} = \mathbf{c}_1^{\alpha_1} \dots c_m^{\alpha_m}$$

$\Rightarrow K(\mathbf{y} \rightarrow \mathbf{y}')$ depends only on \mathbf{y} .

Dynamics for the network

$$\dot{\mathbf{c}}(\mathbf{t}) = F(\mathbf{c}(\mathbf{t}), T) = \sum_{\mathbf{y} \rightarrow \mathbf{y}' \in \mathcal{R}} K(\mathbf{y} \rightarrow \mathbf{y}') (\mathbf{y}' - \mathbf{y})$$

The dynamics is defined by the graph.

More explicit representation of the dynamics.

- **Stoichiometric subspace**

$$S = \text{span} \{ \mathbf{y}' - \mathbf{y}, \mathbf{y} \rightarrow \mathbf{y}' \in \mathcal{R} \}$$

- **Positive class** (strict if > 0)

$$(\mathbf{c}(\mathbf{0}) + S) \cap \mathbb{R}_{\geq 0}^m$$

Lemma

The class $(\mathbf{c}(\mathbf{0}) + S) \cap \mathbb{R}_{> 0}^m$ is **invariant** for the dynamics.

Notations

- **Complex matrix** : $Y = (y_1, \dots, y_n)$ (n : number of complexes).
- **Incidence connectivity matrix** : $A = (a_{ij})_{ij}$
with for instance $a_{21} = k_1$ indicating a reaction with constant k_1 from the first node of the graph to the second.
- **Laplacian matrix** :

$$\tilde{A} = A - \text{diag} \left(\sum_{i=1}^n a_{i1}, \dots, \sum_{i=1}^n a_{in} \right)$$

One has

$$\dot{\mathbf{c}}(\mathbf{t}) = f(\mathbf{c}(\mathbf{t}), T) = Y \tilde{A} \mathbf{c}^Y$$

where $\mathbf{c}^Y = (c^{y_1}, \dots, c^{y_n})^\top$.

Zero deficiency theorem

Definition (Deficiency)

Feinberg and Horn-Jackson : articles in Archive Rational Mechanics

Graph concept : deficiency : $\delta = n - l - s$ where

- n : number of vertices
- l : number of connecting components
- s : dimension of the stoichiometric subspace

Definition

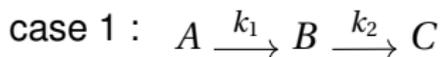
The network is **weakly reversible** if \forall vertices (i, j) such that \exists oriented path joining i to j , there exists an oriented path joining j to i .

Assumption $\delta = 0$ (Zero deficiency assumption)

Theorem

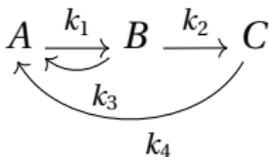
- 1 If the network is **not weakly reversible** then for arbitrary kinetics, the differential equation **cannot have a positive equilibrium nor a positive periodic trajectory**.
- 2 If the network is **weakly reversible**, there exists within each strictly positive compatibility class precisely **one equilibrium** c^* , this equilibrium is locally asymptotically stable with (pseudo-Helmholtz) Lyapunov function $V(c, c^*) = \sum_i [c_i (\ln(c_i) - \ln(c_i^*)) - 1] + c_i^*$.
Moreover there is non trivial periodic orbit.

Application : Test bed cases :



$\delta = 3 - 1 - 2 = 0$: not weakly reversible

case 2 : (**McKeithan network**)



$\delta = 3 - 1 - 2 = 0$: one single equilibrium
globally asymptotically stable

Equilibrium for the McKeithan network

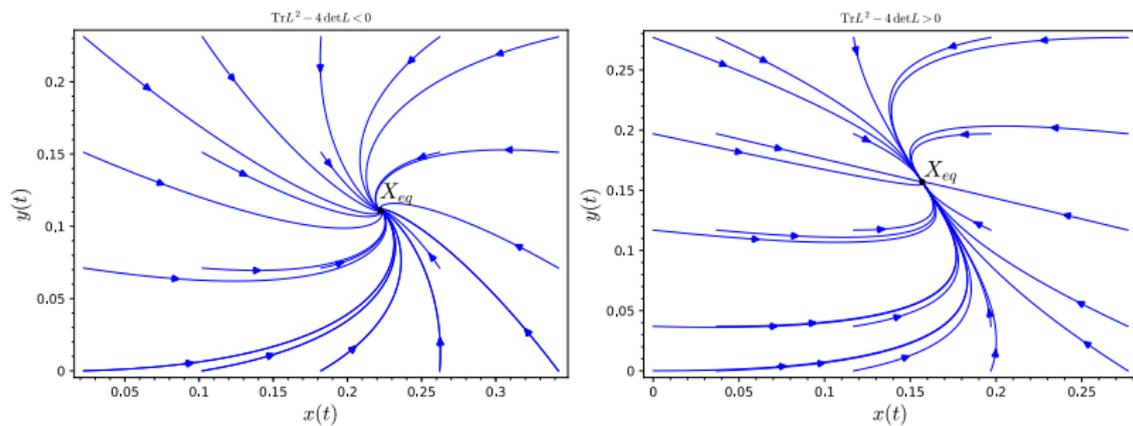


FIGURE – Phase portrait for the McKeithan model. (left) Focus ; (right) Node.

Geometric Optimal Control

Optimal Control Problem

$$\frac{d\mathbf{c}}{dt} = f(\mathbf{c}, T), \quad \frac{dT}{dt} = u, \quad u \in [u_-, u_+]$$

$u(\cdot)$ tracked the derivative of the temperature (related to the Goh Transformation).

Single input C^ω -control system :

$$\begin{cases} \dot{\mathbf{q}} = F(\mathbf{q}) + u G(\mathbf{q}), & |u| \leq 1, \\ \mathbf{q} = (\mathbf{c}, T) \in \mathbb{R}^n \end{cases}$$

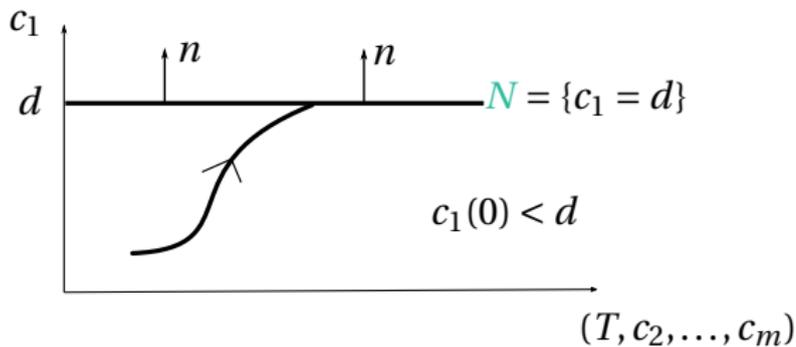
Formulation :

$$\max c_1(t_f) \quad t_f : \text{time batch duration}$$

Formulated as

$$\begin{cases} \min t_f, & |u| \leq 1 \\ c_1(t_f) = d \text{ is a desired quantity} \end{cases}$$

N : terminal manifold of codimension 1.



Necessary optimality conditions Pontryagin Maximum Principle (1956)

Statement :

$$\begin{cases} \dot{\mathbf{q}} = F(\mathbf{q}) + uG(\mathbf{q}), & |u| \leq 1, \\ \min t_f, & \mathbf{q}(t_f) \in \mathbf{N} \end{cases}$$

- $H(\mathbf{q}, p, u) = p \cdot (F(\mathbf{q}) + uG(\mathbf{q}))$, $p \in \mathbb{R}^n \setminus \{0\}$: adjoint vector
- H : pseudo-Hamiltonian and the maximized Hamiltonian is

$$M(\mathbf{q}, p) = \max_{|u| \leq 1} H(\mathbf{q}, p, u), \quad \mathbf{q}, p \text{ are fixed}$$

Theorem

Assume $(q^*(\cdot), p^*(\cdot))$ is a time minimal solution on $[0, t_f^*]$ then there exists $p^*(\cdot)$ such that a.e. on $[0, t_f^*]$:

$$\dot{q}^*(\cdot) = \frac{\partial H}{\partial p}(q^*(t), p^*(t), u^*(t)), \quad \dot{p}^*(\cdot) = -\frac{\partial H}{\partial q}(q^*(t), p^*(t), u^*(t)) \quad (2)$$

the maximization condition is satisfied

$$H(q^*(t), p^*(t), u^*(t)) = M(q^*(t), p^*(t)).$$

Moreover

- $t \mapsto M(q^*(t), p^*(t))$ is constant and ≥ 0 and at the final time one has the transversality condition :

$$p^*(t_f) \perp T_{q^*(t_f)}\mathbf{N} \quad (3)$$

Extremals are solutions of (2). BC-extremal : transversality condition (4) satisfied.

Maximization condition

- *regular* :

$$u(t) = \text{sign} p(t) \cdot G(\mathbf{q}(t)) \text{ a.e.}$$

Finite number of switches : **Bang-Bang**

- *singular* :

$$p(t) \cdot G(\mathbf{q}(t)) = 0 \quad \forall t$$

Exceptional extremals : $M = 0$.

Moreover

- $t \mapsto M(q^*(t), p^*(t))$ is **constant** and ≥ 0 .
- *Transversality condition* :

$$p^*(t_f) \perp T_{q^*(t_f)}\mathbf{N} \quad (4)$$

Extremals are solutions of (2). BC-extremal : transversality condition (4) satisfied.

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Exceptional extremals : $M = 0$.

Computations of singular extremals and properties

Notation : X, Y : two vector fields on \mathbb{R}^n

Lie bracket :

$$[X, Y](q) = \frac{\partial X}{\partial q} Y(q) - \frac{\partial Y}{\partial q} X(q)$$

$z = (q, p)$ $H_X(z) = p \cdot X(q)$

Poisson bracket :

$$\{H_X, H_Y\}(z) = p \cdot [X, Y](q)$$

Computations $H_G(z) = p \cdot G(q) = 0$

Differentiating twice w.r.t. time gives the two equations

$$\frac{d}{dt} H_G(z) = dH_G \cdot \dot{z} = \{H_G, H_F + u H_G\} = \{\mathbf{H}_G, \mathbf{H}_F\} = \mathbf{0}$$

$$\{\{\mathbf{H}_G, \mathbf{H}_F\}, \mathbf{H}_F\}(z) + u \{\{\mathbf{H}_G, \mathbf{H}_F\}, \mathbf{H}_G\}(z) = \mathbf{0}$$

Then if $\{\{H_G, H_F\}, H_G\}(z) \neq 0$ then we compute \hat{u} and plug it in H to obtain the *true Hamiltonian*.

Generalized Legendre-Clebsch condition

$$\{\{H_G, H_F\}, H_F\}(z) \geq 0$$

⇒ necessary optimality condition (High Order Maximum Principle, Krener).

Strict Legendre-Clebsch condition

$$\{\{H_G, H_F\}, H_F\}(z) > 0$$

Classification of singular extremals

$M = H_F$: constant value

- $M = 0$: **Exceptional case**
- $M > 0$: $\{\{H_G, H_F\}, H_G\}(z) > 0$: **Hyperbolic case (fast)**
- $M > 0$: $\{\{H_G, H_F\}, H_G\}(z) < 0$: **Elliptic case (slow)**

Classification of regular extremals (Ekeland - IHES, Kupka - TAMS)

Denote :

- σ_+ : bang arc with $u = +1$
- σ_- : bang arc with $u = -1$
- σ_s : singular arc $u = u_s \in]-1, 1[$

$\sigma_1\sigma_2$ is the arc σ_1 followed by σ_2 .

Switching surface :

- $\Sigma : \{p \cdot G(q) = 0\}$

$\Phi(t) = p(t) \cdot G(\mathbf{q}(t))$ is the **switching function**.

$$\dot{\Phi}(t) = p(t) \cdot [G, F](\mathbf{q}(t))$$

$$\ddot{\Phi}(t) = p(t) \cdot ([[G, F], F](\mathbf{q}(t)) + u(t) [[G, F], G](\mathbf{q}(t)))$$

Ordinary Switching time : $t \in]0, t_f[$ such that $\Phi(t) = 0$ and $\dot{\Phi}(t) \neq 0$

Lemma

Near $z(t)$ every extremal solution projects onto $\sigma_+ \sigma_-$ if $\dot{\Phi}(t) < 0$ and $\sigma_- \sigma_+$ if $\dot{\Phi}(t) > 0$

Fold case : If $\Phi(t) = \dot{\Phi}(t) = 0$ then $z(t) \in \Sigma'$

$$\ddot{\Phi}_\varepsilon(z(t)) = p(t) \cdot ([[G, F], F](\mathbf{q}(t)) + \varepsilon [[G, F], G](\mathbf{q}(t))), \quad \varepsilon = \pm 1$$

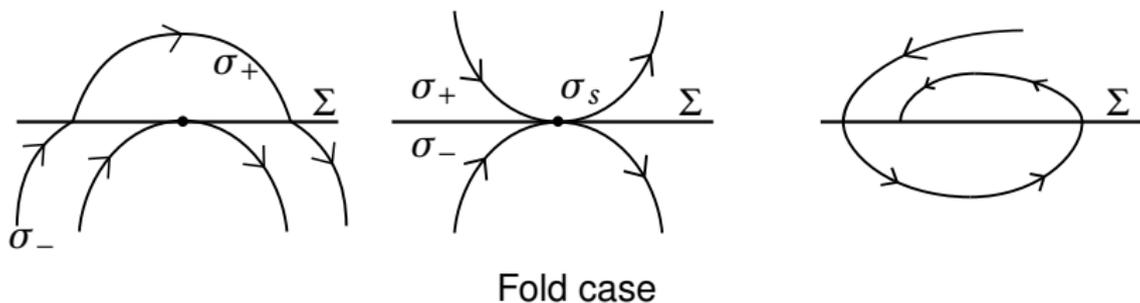
Assumption : Σ' : surface of codimension two, $\ddot{\Phi}_\varepsilon(z(t)) \neq 0$ for $\varepsilon = \pm 1$.

$z(t)$: fold point

- Case 1 : **parabolic case** $\ddot{\Phi}_+(t) \ddot{\Phi}_-(t) > 0$
- Case 2 : **hyperbolic case** $\ddot{\Phi}_+(t) > 0$ and $\ddot{\Phi}_-(t) < 0$
- Case 3 : **elliptic case** $\ddot{\Phi}_+(t) < 0$ and $\ddot{\Phi}_-(t) > 0$

u_s is the singular control defined by

$$p(t) \cdot ([[G, F], F](\mathbf{q}(t)) + u_s(t) [[G, F], G](\mathbf{q}(t))) = 0$$



In the parabolic case $|u_0| > 1$ and the singular arc is not admissible.

Theorem

Kupka TAMS In the neighborhood of $z(t)$ every extremal projects onto :

- *Parabolic case* : $\sigma_+ \sigma_- \sigma_+$ or $\sigma_- \sigma_+ \sigma_-$
- *Hyperbolic case* : $\sigma_{\pm} \sigma_s \sigma_{\pm}$
- *Elliptic case* : every extremal is of the form $\sigma_+ \sigma_- \sigma_+ \sigma_- \dots$ (Bang-Bang) but the number of switches is not uniformly bounded.

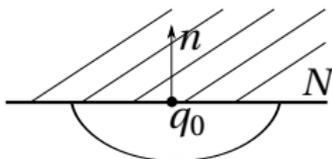
Application to Chemical Networks

Time minimal synthesis for chemical systems

$$\begin{cases} \min t_f & |u| \leq 1 \\ \dot{\mathbf{q}} = F(\mathbf{q}) + uG(\mathbf{q}) \\ \mathbf{c}_1(t_f) \in \mathbf{N} = \{\mathbf{c}_1 = d\} \end{cases}$$

Methods : Two steps :

- 1 Calculation of the time minimal syntheses near the terminal manifold
- 2 Bounds on the number of switches



Step 1: Take $q_0 \in \mathbf{N}$, $z_0 = (q_0, n(q_0))$ where $n(q_0)$ is the normal vector of \mathbf{N} at q_0 .

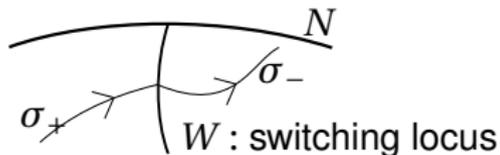
Find, in a small neighborhood U of q_0 , the time minimal closed loop control $u^*(q)$ to reach \mathbf{N} starting from \mathbf{q} in minimal time.

Computations : $\dot{\mathbf{q}} = F(\mathbf{q}) + uG(\mathbf{q})$, $q(t_f) \in \mathbf{N}$

Synthesis : $u^*(\mathbf{q})$ means

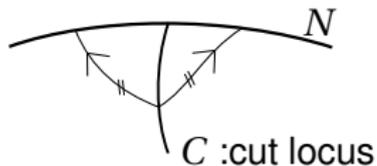
- determine the **switching locus**

Ex. :



- determine the **splitting locus** or the **cut locus** C where two distinct optimal trajectories occur.

Ex. :



Tools : Singularity theory $\mathbf{N} = \{f^{-1}(0)\}$

- *expand* at q_0 with Taylor series : jet spaces.
- *compute* : Normal form to estimate W, C near q_0 . The tools are simple but the classification is complicated.

Tools : Singularity theory $\mathbf{N} = \{f^{-1}(0)\}$

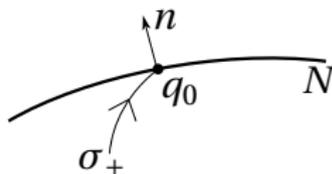
- *expand* at q_0 with Taylor series : jet spaces.
- *compute* : Normal form to estimate W, C near q_0 . The tools are simple but the classification is complicated.

Ex. : *Two reactions only.* $(C, T) \in \mathbb{R}^3$ $\dot{\mathbf{q}} = F + uG$ and $\mathbf{N} = f^{-1}(0)$.

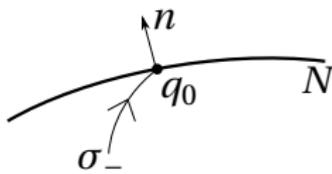
Generic case $z_0 = (q_0, n(q_0))$.

G is tangent to N : Then $p \cdot G = 0$ so p is normal to \mathbf{N} .

Using classification of extremals at a point such that $p \cdot G(\mathbf{q}) = 0$,
 $p \cdot [G, F](\mathbf{q}) \neq 0$:



or



depending on the sign of $p \cdot [G, F](q_0)$.

... but there are more complicated situations

Define :

\mathcal{S} the singular locus : $\{\mathbf{q} \in \mathbf{N}; n \cdot [G, F](\mathbf{q}) = 0\}$

\mathcal{E} the exceptional locus : $\{\mathbf{q} \in \mathbf{N}; n \cdot F(\mathbf{q}) = 0\}$

For \mathcal{S} : from the classification near a fold point one has :

- *Hyperbolic* case
- *Elliptic* case
- *Parabolic* case

To make the analysis we construct a semi-normal form : $\mathbf{q} = (x, y, z)$ near 0

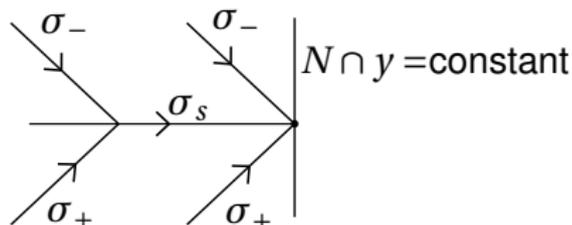
$$\begin{cases} \dot{x} = 1 + a(x) z^2 + 2b(x) yz + c(x) y^2 + \dots \\ \dot{y} = d(x) y + e(0) + \dots \\ \dot{z} = (u - \hat{u}(x)) + f(x)y + g(0)z + \dots \end{cases}$$

with

- \mathbf{N} is identified to $x = 0$
- the singular arc is identified to $\sigma_s : t \rightarrow (t, 0, 0)$ with singular control \hat{u} .
- $a(0) < 0$ hyperbolic if $|\hat{u}| < 1$.
- $a(0) > 0$: elliptic if $|\hat{u}| < 1$.
- parabolic if $|\hat{u}| > 1$.

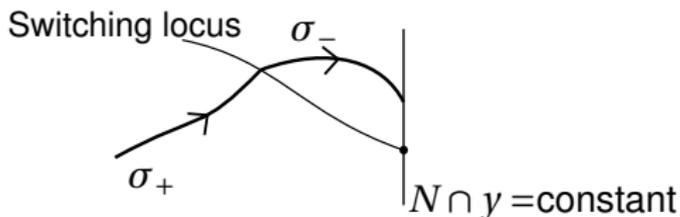
Synthesis : There exists a C^0 -foliation by planes such that in each plane the synthesis is :

Case : Hyperbolic.

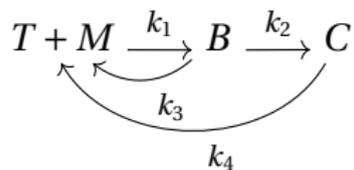


Note that the synthesis is $\sigma_+ \sigma_s \sigma_-$ hence the temperature is not constant.

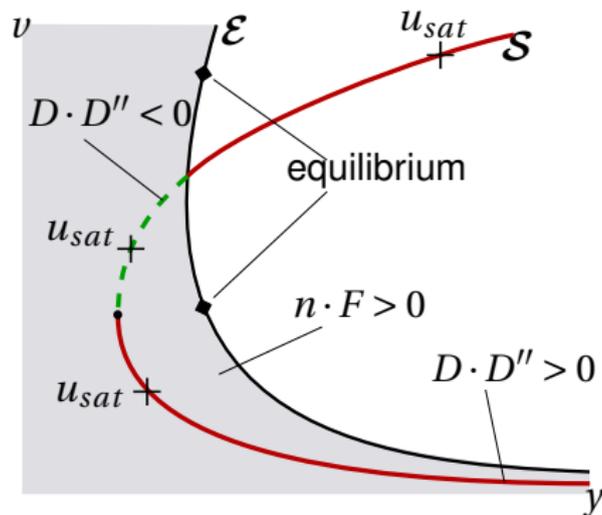
Case : Parabolic. For instance, a synthesis is



The McKeithan network



Stratification of the terminal manifold :



Bridge phenomenon

Local (planar) simplified model (inspired from the saturation problem in Magnetic Resonance Imaging) :

$$\min_{u(\cdot)} t_f \quad \dot{q}(t) = F(q(t)) + u G(q(t)), \quad t \in [0, t_f]$$

where

$$q = (x, y), \quad F = (1 - x^2 y) \frac{\partial}{\partial y}, \quad G = -(y - 1) \frac{\partial}{\partial x} + x \frac{\partial}{\partial y}.$$

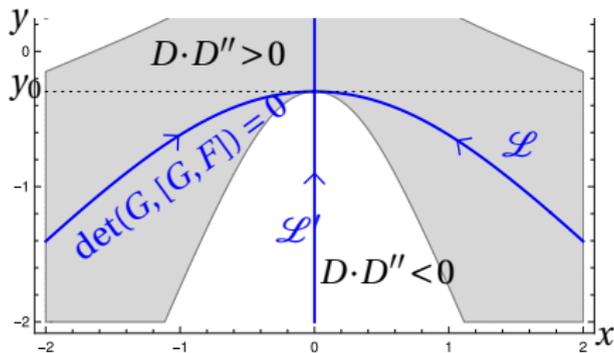
Singular lines $\mathcal{L} : \det(G, [G, F])(q) = 0$

- **fast** if $D(q) \cdot D''(q) > 0$
- **slow** if $D(q) \cdot D''(q) < 0$

where $D(q) = \det(G(q), [G, F](q), [[G, F], G](q))$

and $D'(q) = \det(G(q), [G, F](q), [[G, F], F](q))$

Singular sets

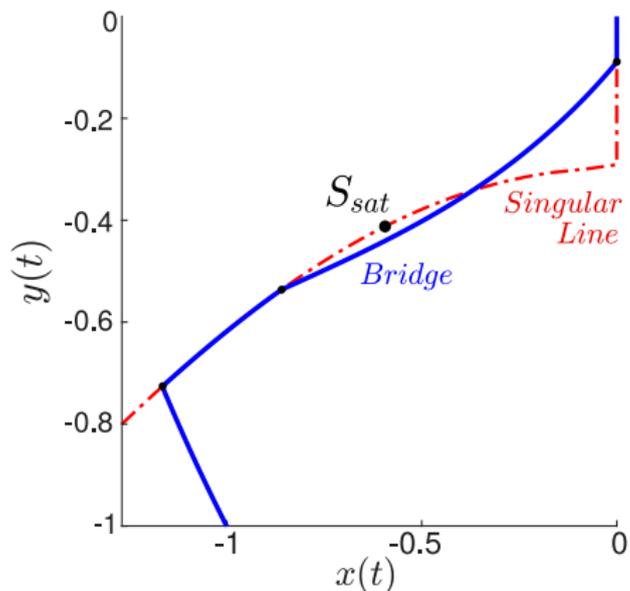


The **Singular control** along \mathcal{L} is

$$u_s(q) = -\frac{D'(q)}{D(q)}$$

and is not bounded.

Trajectories : bridge



Bridge connecting two switching points of the singular set.

Conclusion

General techniques to handle complicated networks :

Geometric approach : Find coordinates to analyze the syntheses

→ applicable to general networks

Details :

T. Bakir, B. Bonnard, J. Rouot *Geometric Optimal Control Techniques to Optimize the Production of Chemical Reactors using Temperature Control* (submitted 2019)

B. Bonnard, G. Launay, M. Pelletier,

Classification générique de synthèses temps minimales avec cible de codimension un et applications,

Annales de l'I.H.P. Analyse non linéaire **14** no.1 (1997) 55–102.

Even a simple network $A \rightarrow B \rightarrow C$ can give complex optimal solution : work in progress on the *McKeithan network*.