From CTRW to Fractional Diffusion of pointly defects at the atomic scale

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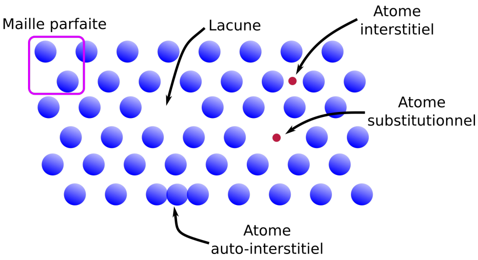
Défauts ponctuels dans les métaux & diffusion

- Il existe différents types de défauts ponctuels dans les métaux.
- La migration de ces défauts ponctuels $\Longrightarrow$ processus de diffusion.
- Cette migration des défauts ponctuels est influencée par leur environnement : le type de matériau, autres défauts présents ou non (dislocations, joints de grains, etc ...), hétérogénéité, ...



Cas des dislocations

- La notion de dislocation fut introduite en physique du solide en 1934 par G.I. Taylor, E. Orowan & M. Polanyi.
- Elle permit de résoudre le désaccord entre la théorie et l'expérience.
- Notion essentielle en physique du solide : comportement des matériaux sous déformation plastique.

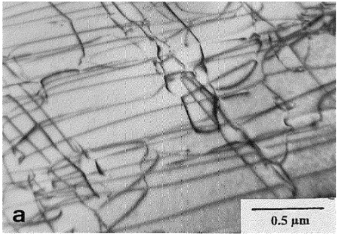


Fig. 1.1 – Image obtenue en microscopie électronique à transmission (MET) par M. Azzaz et al. [57] de lignes de dislocations parfaites dans un échantillon de nitrure d'aluminium déformé à haute température.

Dislocations & circuit de Burgers

- Les dislocations sont des singularités linéaires du réseau cristallin.
- Elles permettent de relaxer localement l'énergie du réseau déformé.
- Ces défauts linéaires propagent la déformation plastique en se déplaçant.

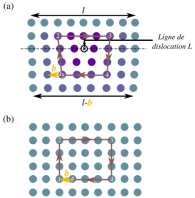


Fig. 1.3 – (a) Circuit de Burgers dans réseau cristallin cubique simple déformé, contenant une ligne de dislocation orientée vers l'arrière, les cercles verts représentent les atomes étant en configuration de cristal parfait, les autres cercles représentent les atomes en configuration métastable. Le sens de rotation du circuit est selon le sens des aiguilles d'une montre (Point 1, puis point 2,...). (b) Circuit de Burgers de (a) transposé à l'identique dans le réseau cristallin parfait.

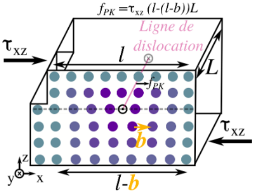
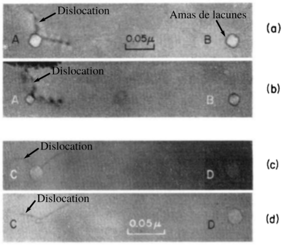


Fig. 1.6 – Schéma d'un cristal, contenant une dislocation coin, soumis à une contrainte de cisaillement τ_{xz}.

Dislocations & circuit de Burgers

- Images de microscopie électronique en transmission provenant de Volin & Balluffi.
- Séquence de recuit de cavités isolées et de cavités connectées à des dislocations.
  - (a) Échantillon recuit à 180°C.
  - (b) Idem que (a) mais 29 min plus tard.
  - La diminution de la taille de l'amas A connecté à une dislocation est plus grande que celle de l'amas isolé B.
  - (c) Échantillon recuit à 80°C. (d) Idem que (c) mais 48,5 h plus tard. La diminution de la taille de l'amas C connecté à une dislocation est plus grande que celle de l'amas isolé D.



Kinetic Monte-Carlo

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Full length article

Efficiency of the vacancy pipe diffusion along an edge dislocation in MgO

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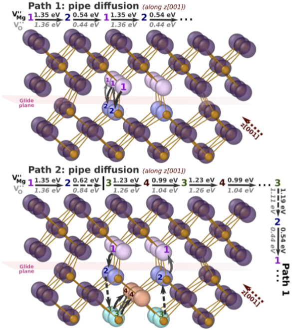
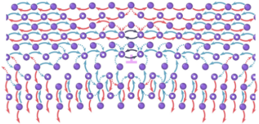
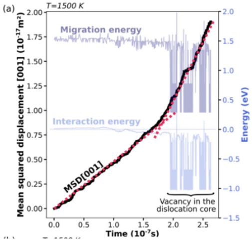






From Continuous Time Random Walk (CTRW) to Fractional Derivatives

$(T_n)_{n\geq 1}$: positive *iid* random waiting times having a *pdf* $\psi(t)$, $t > 0$.
$(X_n)_{n\geq 1}$: *iid* random jumps having a *pdf* $w(x)$, $x \in \mathbb{R}$.

Setting $t_0 := 0$, $t_n := \sum_{k=1}^{n} T_k$.

The wandering particle :

- Starts at point $x = 0$ in instant $t = 0$.
- Makes a jump X_n in instant t_n.
- $x = 0$ for $0 \leq t < T_1 = t_1$.
- $x = \sum_{k=1}^{n} X_k$ for $t_n \leq t < t_{n+1}$.

Hypothesis : $(T_n)_{n\geq 1}$ and $(X_n)_{n\geq 1}$ are independent.

THE MASTER EQUATION OF MONTROLL & WEISS (1965)

Probabilistic arguments $\Longrightarrow$ The master Equation (Montroll & Weiss, 1965)

$$p(x,t) = \delta\,(x)\,\int_t^{+\infty} \psi(\tau)\,d\tau \;+\; \int_0^t \psi(t-\tau)\left(\int_{-\infty}^{+\infty} w(x-\xi)p(\xi,\tau)\,d\xi\right)\,d\tau, \quad (1)$$

where

- $p(x,t)$: the probability that the particle is in position x at time t.
- $\delta\,(x)$ is the Dirac measure.
- $\displaystyle\int_t^{+\infty} \psi(\tau)\,d\tau$: (Survival probability at the origin).
- $\displaystyle\int_t^{+\infty} \psi(\tau)\,d\tau$: probability that the particle is still sitting in $x = 0$ at instant t.
- $p(x, 0^+) = \delta(x)$.

From Master Equation to Fractional Differential Equations

Theorem 1 (R. Gorenflo & F. Mainardi)

Assume that :

- $w(x) \sim c_1 \, |x|^{\alpha-1}$ *as* $|x| \longrightarrow +0,$ *with* $\alpha \in]0,2[.$
- $\psi(t) \sim c_2 \, t^{\beta-1}$ *as* $t \longrightarrow +0,$ *with* $\beta \in]0,1[.$

Then, up to scaling the variables :

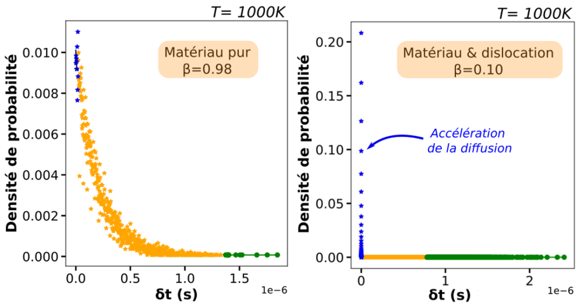
- $x \longleftarrow (\Delta x) \times x,$
- $t \longleftarrow (\Delta t) \times t,$
- *with* $(\Delta x)^\alpha = c_3 \, (\Delta t)^\beta,$

the master equation (1) goes over to the space-time fractional diffusion equation :

$$\begin{cases} D^{\beta}_{+,t} \, p(x,t) \; - \; D^{\alpha}_{+,x} \, p(x,t) = 0, \quad x \in \mathbb{R}, \quad t > 0, \\ u(x,0^+) = \delta(x), \quad x \in \mathbb{R}, \quad t > 0, \end{cases}$$

where the fractional differential operators $D^{\beta}_{+,t}$ *and* $D^{\alpha}_{+,x}$ *will be specified.*

NORMAL *versus* ANOMALOUS DIFFUSION : KMC



Rank $n \in \mathbb{N}^*$ integral of a function

Let $u \in L^1(]a,b[)$. We define the integral operators on $L^1(]a,b[)$:

- $I^1u(x) = \int_a^x f(t)\,dt$
- $I^2u(x) = \int_a^x I^1u(t)\,dt = \int_a^x (x-t)\,u(t)\,dt$ (Fubini)
- $I^3u(x) = \int_a^x I^2u(t)\,dt = \int_a^x \frac{1}{2\,!}(x-t)^2\,u(t)\,dt$ (Fubini)
- $\vdots$
- $I^nu(x) = \int_a^x I^{n-1}u(t)\,dt = \int_a^x \frac{1}{(n-1)\,!}(x-t)^{n-1}\,u(t)\,dt$ (Fubini)

Then,

$$I^nu(x) = \frac{1}{\Gamma(n)}\int_a^x (x-t)^{n-1}\,u(t)\,dt$$

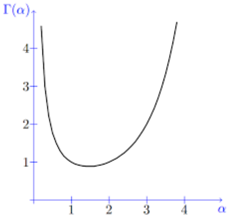
can be extended by analytic continuation to non integer orders.

Definition 2 (Riemann-Liouville's fractional integral and derivative of order $\alpha > 0$)

1. The left and right sided Riemann-Liouville fractional integrals of order α :

$$\begin{cases} I_{+}^{\alpha}\, u(x) = \dfrac{1}{\Gamma(\alpha)} \displaystyle\int_a^x (x-t)^{\alpha-1}\, u(t)\, dt, \\ I_{-}^{\alpha}\, u(x) = \dfrac{1}{\Gamma(\alpha)} \displaystyle\int_x^b (t-x)^{\alpha-1}\, u(t)\, dt. \end{cases}$$

where $\Gamma(\alpha) = \displaystyle\int_0^{+\infty} t^{\alpha-1}\, e^{-t}\, dt$ is the Euler's Gamma function and $n = [\alpha] + 1$.



Non integer order derivation

Definition 3 (Riemann-Liouville's fractional derivatives $\alpha > 0$)

1. The Left and right sided Riemann-Liouville fractional derivatives of order $\alpha > 0$

$$\begin{cases} D_+^\alpha u(x) & = & \dfrac{d^n}{dx^n}\left[I_+^{n-\alpha}u(x)\right], \\ D_-^\alpha u(x) & = & (-1)^n \dfrac{d^n}{dx^n}\left[I_-^{n-\alpha}u(x)\right], \end{cases}$$

where $n = [\alpha] + 1.$

2. $D_+^n\, u = u^{(n)}$ and $D_-^n\, u = (-1)^n\, u^{(n)}, \quad \forall n \in \mathbb{N}.$

Identification of the fractional derivative order at the macroscopic scale

Pourquoi l'identification à l'échelle macroscopique ?

Approche à l'échelle atomique très coûteuse en temps de calcul !

Consider the elliptic boundary value problem :

$$\begin{cases} D_-^{\alpha}\left[k(x)\ D_+^{\alpha} u(x)\right] = f(x),\ x \in \Omega =]a,b[, \\ \\ u(a) = u(b) = 0, \end{cases} \qquad (2)$$

- The source term $f \in L^2(\Omega)$.
- The diffusivity function $k \in C^1(\overline{\Omega})$ is positive.

Question : Can we find α if we have a measure of the solution of (2) ?

Let u_α be the unique solution of Equation (2).

Formally, we have

$$\frac{d}{d\alpha}\left[D_+^\alpha u_\alpha\right] = \widehat{D}_+^\alpha u_\alpha + \frac{\Gamma'(1-\alpha)}{\Gamma(1-\alpha)}\, D_+^\alpha u_\alpha(x) + \; D_+^\alpha \frac{du_\alpha}{d\alpha}, \tag{3}$$

where

$$\widehat{D}_+^\alpha u(x) = \frac{d}{dx}\left(\int_a^x \frac{(x-t)^{-\alpha}}{\Gamma(1-\alpha)} \ln\left(\frac{1}{x-t}\right) u(t)dt\right). \tag{4}$$

In the same way, we get :

$$\frac{d}{d\alpha}\left[D_-^\alpha u_\alpha\right] = \widehat{D}_-^\alpha u_\alpha + \frac{\Gamma'(1-\alpha)}{\Gamma(1-\alpha)}\, D_-^\alpha u_\alpha + \; D_-^\alpha \frac{du_\alpha}{d\alpha}, \tag{5}$$

where

$$\widehat{D}_-^\alpha u(x) = -\frac{d}{dx}\left(\int_x^b \frac{(t-x)^{-\alpha}}{\Gamma(1-\alpha)} \ln\left(\frac{1}{t-x}\right) u(t)dt\right). \tag{6}$$

Let

$$\mathbb{L}_\alpha \, u(x) \quad = \quad D_-^\alpha \left[k(x) \, D_+^\alpha u(x) \right].$$

The problem satisfied by the function $\dfrac{du_\alpha}{d\alpha}$ is :

$$\begin{cases} \mathbb{L}_\alpha \, w(x) = -\mathbb{S}_\alpha u_\alpha - 2 \dfrac{\Gamma'(1-\alpha)}{\Gamma(1-\alpha)} \mathbb{L}_\alpha u_\alpha(x) \quad x \in \Omega =]a, b[, \\ \\ w(a) \; = \; w(b) \; = \; 0, \end{cases} \tag{7}$$

where

$$\mathbb{S}_\alpha u_\alpha = \widehat{D}_-^\alpha \left[k(x) D_+^\alpha \, u_\alpha(x) \right] + D_-^\alpha \left[k(x) D_+ \widehat{D}^\alpha \, u_\alpha(x) \right].$$

Now if

$$J(\alpha) = \frac{1}{2} \int_a^b \left(u_\alpha(x) - u_\alpha^{\text{obs}}(x) \right)^2 dx$$

then (formally)

$$J'(\alpha) = \int_a^b \left(u_\alpha(x) - u_\alpha^{\text{obs}}(x) \right) \, w_\alpha(x) \, dx,$$

where w_α is the solution of (7).

Functional Framework

Consider the problem

$$\begin{cases} \mathbb{L}_\alpha \, u(x) = f(x), \quad x \in \Omega =]a, b[, \\ \\ u(a) = u(b) = 0, \end{cases} \tag{8}$$

Our aim:

- To define the functional framework to study the variational problem (8).
- Study the regularity of the solution of (8) with respect to α.

Definition 4

Let $0 < \alpha \leq 1$ and u, v in $L^1(]a,b[)$. The function v is called the left weak fractional derivative of order α of u if

$$\int_a^b u(t)\, D_-^{\alpha}\varphi(x)\, dx = \int_a^b v(t)\varphi(x)\, dx, \qquad \forall\, \varphi \in \mathscr{C}_0^{\infty}(]a,b[).$$

The function v will be denoted by $\nabla_+^{\alpha}\, u$.

The right weak fractional derivative of order α is defined in a similar way.

1. This definition is motivated by the identity (Fubini) :

$$\int_a^b u(t)\, D_-^{\alpha}\varphi(x)\, dx = \int_a^b \varphi(x) D_+^{\alpha} u(x)\, dx, \qquad \forall\, \varphi \in \mathscr{C}_0^{\infty}(]a,b[).$$

2. $\nabla_+^1\, u = \nabla u$ and $\nabla_-^1\, u = -\nabla u$, where ∇u is the classical weak derivative of u.

Riemann-Liouville Sobolev spaces

Definition 5

Let $0 < \alpha \leq 1$ and $1 \leq p < +\infty$.

- The left Riemann-Liouville Sobolev space $W_{+}^{\alpha,p}(]a,b[)$ is defined by :

$$W_{+}^{\alpha,p}(]a,b[) = \left\{u \in L^p(]a,b[), \quad \nabla_{+}^{\alpha} u \in L^p(]a,b[)\right\}.$$

  The norm in $W_{+}^{\alpha,p}(]a,b[)$ is

$$\|u\|_{W_{+}^{\alpha,p}(]a,b[)} = \left(\|u\|_p^p + \left\|\nabla_{+}^{\alpha} u\right\|_p^p\right)^{\frac{1}{p}}.$$

- The right Riemann-Liouville Sobolev space is defined in a similar way.

For $\alpha = 1$, we find the classical Sobolev space :

$$W_{+}^{1,p}(]a,b[) = W_{-}^{1,p}(]a,b[) = W^{1,p}(]a,b[).$$

Riemann-Liouville Sobolev spaces

Theorem 6

Let $0 < \alpha < 1$*. Then, the Riemann-Liouville Sobolev spaces* $W_{+}^{1,p}(]a,b[)$ *and* $W_{-}^{1,p}(]a,b[)$ *are :*

1. *Separable Banach spaces for* $1 \leq p < +\infty$*.*
2. *Reflexive for* $1 < p < +\infty$*.*

For the boundary conditions, we define :

Definition 7

For $0 < \alpha \leq 1$ and $1 \leq p < +\infty$, the fractional Riemann-Liouville Sobolev space is defined by

$$W_{0,+}^{1,p}(]a,b[) := \overline{\mathscr{C}_0^{\infty}(]a,b[)}^{W_{+}^{1,p}(]a,b[)}$$

Riemann-Liouville Sobolev spaces

For $\alpha = 1$, we find the classical Sobolev space :

$$W_{0,+}^{1,p}(]a,b[) = W_{0,-}^{1,p}(]a,b[) = W_0^{1,p}(]a,b[).$$

Theorem 8

Let $0 < \alpha < 1$. Then, the Riemann-Liouville Sobolev spaces $W_{0,+}^{\alpha,p}(]a,b[)$ and $W_{0,-}^{\alpha,p}(]a,b[)$ are :

1. *Separable Banach spaces for $1 \leq p < +\infty$.*
2. *Reflexive for $1 < p < +\infty$.*

Bounded Integral fractional operators

Lemma 9

1. *The linear fractional operator* $I_+^\alpha \; : \; L^p(]a,b[) \longrightarrow W_+^{\alpha,p}(]a,b[)$ *bounded.*
2. *The linear fractional operator* $I_-^\alpha \; : \; L^p(]a,b[) \longrightarrow W_-^{\alpha,p}(]a,b[)$ *bounded.*

Proof. Let $u \in L^p(]a,b[)$ and $w = I_+^\alpha u$.

- I_+^α is a bounded operator from $L^p(]a,b[)$ to $L^p(]a,b[)$.

  (Riez-Thorin's interpolation Theorem : $p_1 = 1, p_2 = \infty$ and $\theta \in]0,1[$).
- $I_+^{1-\alpha} w = I_+^{1-\alpha} I_+^\alpha u = I_+^1 u = u \in L^p(]a,b[)$. So $D_+^\alpha w = u = \nabla_+^\alpha w$.
- Then $w \in W_+^{\alpha,p}(]a,b[)$.
- $\|I_+^\alpha u\|^p_{W_+^{\alpha,p}(]a,b[)} = \|I_+^\alpha u\|_p^p + \|\nabla_+^\alpha I_+^\alpha u\|_p^p \leq C_{\alpha,p} \|u\|_p^p + \|u\|_p^p$ □

Bounded Derivation fractional operators

Lemma 10

1. *The linear fractional operator* $\nabla_+^\alpha \ : \ W_+^{\alpha,p}(]a,b[) \longrightarrow L^p(]a,b[)$ *is bounded.*
2. *The linear fractional operator* $\nabla_-^\alpha \ : \ W_-^{\alpha,p}(]a,b[) \longrightarrow L^p(]a,b[)$ *bounded.*

Proof. Immediate.

Bounded Derivation fractional operators

Lemma 11

1. $\nabla_+^\alpha \circ I_+^\alpha = id$ *on* $L^p(]a,b[)$, *(precedent Lamma)*
2. $I_+^\alpha \circ \nabla_+^\alpha = id$ *on* $W_{0,+}^{\alpha,p}(]a,b[)$.

Proof.

2. Idea : For any $\varphi \in \mathscr{C}_0^\infty(]a,b[)$ and any $x \in]a,b[$ one has :

$$I_+^\alpha \nabla_+^\alpha \varphi(x) = I_+^\alpha D_+^\alpha \varphi(x) = \varphi(x) - \frac{I_+^{1-\alpha}\varphi(a)}{\Gamma(\alpha)}(x-a)^{\alpha-1} = \varphi(x).$$

– Fix $u \in W_{0,+}^{\alpha,p}(]a,b[)$ and a sequence $(\varphi_n)_n \subset \mathscr{C}_0^\infty(]a,b[)$ such that

$$\varphi_n \longrightarrow u \quad \text{in} \quad W_{0,+}^{\alpha,p}(]a,b[).$$

– Use the fact that

$$I_+^\alpha \nabla_+^\alpha u - u = \left(I_+^\alpha \nabla_+^\alpha u - I_+^\alpha \nabla_+^\alpha \varphi_n\right) + (\varphi_n - u).$$

– Conclude. □

Poincaré inequality

Lemma 12

We have the Poincaré inequality on $W^{\alpha,p}_{0,+}(]a,b[)$ *:*

$$\|u\|_p \leq \frac{(b-a)^\alpha}{\Gamma(\alpha+1)} \left\|\nabla^\alpha_+ u\right\|_p , \quad \forall\, u \in W^{\alpha,p}_{0,+}(]a,b[).$$

Proof. Use the continuity of the operator ∇^α_+ on $L^p(]a,b[)$. □

→ In what follows, $W^{\alpha,p}_{0,+}(]a,b[)$ will be endowed with the equivalent norm

$$\|u\|_{W^{\alpha,p}_{0,+}(]a,b[)} = \left\|\nabla^\alpha_+ u\right\|_p .$$

Lemma 13 (Continuous representative in $W_{0,+}^{\alpha,p}$)

For $\alpha > \frac{1}{p}$, we have :

$$\forall\, u \in W_{0,+}^{\alpha,p}(]a,b[), \quad \exists\, \widetilde{u} \in \mathscr{C}^0([a,b]), \quad : \quad u \;=\; \widetilde{u} \quad a.e \quad \text{sur} \quad [a,b].$$

Proof. Let $u \in W_{0,+}^{\alpha,p}(]a,b[)$ and set

$$\widetilde{u} := I_+^\alpha \, \nabla_+^\alpha \, u.$$

- $|\widetilde{u}(x)| \quad \leq \quad C(a,b,\alpha)\, \|\nabla_+^\alpha \, u\|_p \qquad \text{since} \quad p'(1-\alpha) > -1.$
- Let x and y in $[a,b]$. Then

$$\begin{aligned} |\widetilde{u}(x) - \widetilde{u}(y)| &= |I_+^\alpha \nabla_+^\alpha u(x) - I_+^\alpha \nabla_+^\alpha u(y)| \\ &\leq C|x-y|^{\alpha - \frac{1}{p}} \, \|\nabla_+^\alpha \, u\|_p \end{aligned}$$

□

Compactness of the embedding $W^{\alpha,p}_{0,+}(]a,b[) \subset \mathscr{C}^0([a,b])$

Lemma 14 (Compactness of the embedding $W^{\alpha,p}_{0,+}(]a,b[) \subset \mathscr{C}^0([a,b])$)

For $\alpha > \frac{1}{p}$, the embedding of $W^{\alpha,p}_{0,+}(]a,b[)$ in $\mathscr{C}^0([a,b])$ is compact.

Proof. Arzelà–Ascoli's Theorem : any bounded sequence in $W^{\alpha,p}_{0,+}(]a,b[)$ is equicontinuous. □

Lemma 15 (Boundary conditions)

For $\alpha > \frac{1}{p}$, the affirmation

$$u \in W^{\alpha,p}_{0,+}(]a,b[) \quad \Longrightarrow u(a) = u(b) = 0$$

holds true.

Proof.

- Use the precedent Lemma.

Existence and uniqueness of solution to the identification problem

Consider the variational problem

$$\begin{cases} \mathbb{L}_\alpha \, w(x) = F(u_\alpha) \quad x \in \Omega =]a, b[, \\ \\ w(a) = w(b) = 0, \end{cases} \tag{9}$$

where

$$F(u_\alpha) = -\mathbb{S}_\alpha u_\alpha - 2 \frac{\Gamma'(1-\alpha)}{\Gamma(1-\alpha)} \mathbb{L}_\alpha u_\alpha .$$

Definition 16

A weak solution to (9) is a function $u \in W^{\alpha,p}_{0,+}(]a, b[)$ satisfying

$$\int_a^b k(x) \, \nabla^\alpha_+ \, u(x) D^\alpha_+ \, \varphi(x) \, dx = \int_a^b F(u_\alpha)(x) \varphi(x) \, dx, \quad \forall \, \varphi \in \mathscr{C}^\infty_0([a, b]).$$

A classical solution to (9) is a function $u \in W^{\alpha,p}_{0,+}(]a, b[)$ satisfying

$I^\alpha_- \, D^\alpha_+ \, u$ is derivable on $]a, b[$, that is $D^\alpha_- \, D^\alpha_- \, u$ is well defined on $]a, b[$.

Euler-Lagrange functional

- The Euler-Lagrange functional associated to (9) is given by :

$$J_\alpha(u) = \frac{1}{2}\int_a^b k(x)\,|\nabla_+^\alpha u(x)|^2 dx - \int_a^b F(u_\alpha)(x)\,u(x)\,dx.$$

- J_α is $\mathscr{C}^1(W_{0,+}^{\alpha,p}(]a,b[))$.
- $J'_\alpha(u)\cdot v = \int_a^b k(x)\,\nabla_+^\alpha u(x)\,\nabla_+^\alpha v(x)\,dx - \int_a^b F(u_\alpha)(x)\,v(x)\,dx,$
  $\forall\, v \in W_{0,+}^{\alpha,p}(]a,b[)$.
- Moreover, the critical points of J_α are weak solutions of Problem (9).

Existence of a weak or a classical solution

Theorem 17

Let $1/2 < \alpha < 1$.

1. *If* $f \in L^2(]a,b[)$ *then Problem (9) has a unique weak solution.*
2. *If* $f \in \mathscr{C}^0(]a,b[)$ *then Problem (9) has a unique classical solution.*

Proof.

1. If $f \in L^2(]a,b[)$: Use Lax-Milgram's Theorem.
2. If $f \in \mathscr{C}^0(]a,b[)$: Show that $I_-^{\alpha-1}\left(D_+^\alpha u - I_-^\alpha f\right) = \text{const}$ in]a,b[and compose with $\dfrac{d}{dx}$.

Regularity w. r. t. α

Theorem 18

Let $p \in [1, +\infty]$, *then for any* $\alpha_0 \geq 0$ *on has :*

$$\lim_{\alpha \to \alpha_0} \| I_+^{\alpha} - I_+^{\alpha_0} \|_{\star, p} = 0.$$

Moreover, it holds :

$$\lim_{\alpha \to 0^+} \frac{I_+^{\alpha} u(x) - u(x)}{\alpha} = -\Gamma'(1) u(x) + \frac{d}{dx} \int_a^x \ln(x - t) u(t)\, dt.$$

Proof. Very technical !

Merci pour votre attention !