Decay time estimates in Scintillators from continuum physics models

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Outline

1. Background

2. Reaction-Diffusion-Drift equations

3. Existence and Asymptotic decays estimates

Aim

- To get a fully-consistent phenomenological theory of Scintillators as it was done for Semiconductors.
- To obtain mathematical estimates for Decay Time and Local Light Yield.
- To perform parametric analysis by means of qualitative properties of PDE.

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Main References

1. F.D.- A Continuum Theory for Scintillating Crystals. European Physics Journal B, *submitted*, 2018.
2. F.D.- Existence, decay time and light yield for a reaction-diffusion-drift equation in the continuum physics of scintillators. Springer INDAM Proceedings "Harnack's inequalities and nonlinear operators" *in print*, 2018.

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State variable: excitation carriers densities

Vector of excitation carriers densities

$$\mathbf{n} \equiv (n_1\,, n_2\,, \dots\,, n_m)\,, \quad n_k : \Omega \times [0\,, \tau) \mapsto \mathcal{M} \equiv [0\,, +\infty)^m\,,$$

where Ω is the scintillation region. We set

$$N = \operatorname{tr}\mathbb{N} = \sum_{k=1}^{m} n_k > 0\,, \quad \mathbb{N}(\mathbf{n}) \equiv \operatorname{diag}[n_1\,, n_2\,, \dots\,, n_m]\,,$$

where N is an approximated solution of the Bethe-Bloch equation renormalized for the elementary track volume going to zero.

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Micromechanical balance laws

Balance equation for Continuum Micro-Mechanics:

$$\begin{aligned} \operatorname{div}\mathbf{T} - \mathbf{k} + \mathbf{b} = \dot{\mathbf{n}} \qquad & \text{in } \Omega \times [0\,,\tau) \\ \mathbf{T}\mathbf{m} = \mathbf{s} \qquad & \text{on } \partial\Omega \times [0\,,\tau) \end{aligned}$$

They represent the conservation of of the electric current (normalized w.r.t. to e); an electric potential is associated to $\mathbf{n}$:

$$-\epsilon_o \Delta\varphi = e\mathbf{q}\cdot\mathbf{n} \qquad \text{in } \Omega \times [0\,,\tau)$$

where $\mathbf{q}$ is the charge number.

Thermodynamics

We describe the scintillation at a scale such that thermodynamics makes sense.

Gibbs free-energy $\psi = \varepsilon + \theta\eta$

$$\begin{aligned} \varepsilon(\mathbf{n}\,,\theta) &= \frac{1}{2}\epsilon_0\|\nabla\varphi\|^2 + u(\theta)\,, \text{ Internal Energy} \\ \eta(\mathbf{n}\,,\theta) &= \eta_e(\mathbf{n}) + \eta_u(\theta)\,, \text{ Entropy} \end{aligned}$$

Dissipation inequality:

$$\dot{\psi} + \eta\dot{\theta} - \boldsymbol{\mu}\cdot\dot{\mathbf{n}} - \mathbf{T}\cdot\nabla\boldsymbol{\mu} - \mathbf{k}\cdot\boldsymbol{\mu} + \theta^{-1}\mathbf{h}\cdot\nabla\theta \leq 0\,,$$

where $\boldsymbol{\mu}$ is the *Scintillation Potential*. (The equivalent of the electrochemical potential in semiconductors).

Constitutive assumptions

If $\psi = \hat{\psi}(\mathbf{n}, \theta)$, then we have a conservative

$$\eta = -\hat{\psi}_\theta(\mathbf{n}, \theta) \quad \boldsymbol{\mu} = \hat{\psi}_{\mathbf{n}}(\mathbf{n}, \theta)$$

and a totally dissipative part:

$$\begin{aligned} \mathbf{T}(\mathbf{n}, \theta, \nabla\boldsymbol{\mu}) &= \mathbb{S}(\mathbf{n}, \theta)[\nabla\boldsymbol{\mu}] \\ \mathbf{k}(\mathbf{n}, \theta, \boldsymbol{\mu}) &= \mathbb{H}(\mathbf{n}, \theta)\boldsymbol{\mu} \end{aligned}$$

$\mathbb{S}$ and $\mathbb{H}$ are two symmetric and positive semi-definite $m \times m$ matrices.

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Gibbs entropy and Scintillation potential

We assume for **n** a Gibbs entropy

$$\eta(\theta\,,\mathbf{n}) = \lambda\log\theta + \theta\,k_B(\mathbf{n}\cdot\log(\mathbb{N}(\mathbf{n})\mathbf{c}) - \mathrm{tr}\,\mathbb{N}(\mathbf{n}));$$

which leads to a Scintillation potential:

$$\boldsymbol{\mu}(\mathbf{n}\,,\theta) = e\varphi\mathbf{q} + \theta\,k_B\log(\mathbb{N}(\mathbf{n})\mathbf{c})$$

Mobility and Diffusivity

From $\mathbb{S}(\mathbf{n}\,,\theta)$ we define the *Mobility* $m \times m$ matrix

$$\mathbb{M}(\theta) = e^{-1}\mathbb{S}(\mathbf{n}\,,\theta)(\mathbb{N}(\mathbf{n}))^{-1}\,;$$

and the *Diffusivity* $m \times m$ matrix

$$\mathbb{D}(\theta) = k_B\theta\,\mathbb{M}(\theta) \text{ (the Einstein-Smoluchowski relation).}$$

Recombination term

We assume

$$\mathbb{H}(\mathbf{n}\,,\theta) = f(\mathbf{n}\,,\boldsymbol{\mu}\,,\theta)\,\mathbf{a}(\theta)\otimes\mathbf{a}(\theta)$$

with

$$\begin{aligned} f(\mathbf{n}\,,\boldsymbol{\mu}\,,\theta) &= \frac{\mathbf{a}(\theta)\cdot\mathbf{n}}{\boldsymbol{\mu}\cdot\mathbf{a}(\theta)}\sum_{k=0}^{\infty}(\mathbf{c}_k(\theta)\cdot\mathbf{n})^k \geq 0 \\ \lim_{\mathbf{n}\to 0^+} f(\mathbf{n}\,,\boldsymbol{\mu}\,,\theta) &= 0 \end{aligned}$$

to obtain:

$$\mathbb{H}(\mathbf{n}\,,\theta)\boldsymbol{\mu} = \mathbb{K}(\mathbf{n}\,,\theta)\mathbf{n}$$

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Non-isothermal R-D-D for scintillators

$$\text{div}\,(\mathbb{D}(\theta)\nabla\mathbf{n} + \mathbb{M}(\theta)\mathbb{N}(\mathbf{n})\mathbf{q}\otimes\nabla\varphi) - \mathbb{K}(\mathbf{n}\,,\theta)\mathbf{n} = \dot{\mathbf{n}}$$

$$\lambda\dot{\theta} = \text{div}\,\mathbf{C}(\mathbf{n}\,,\theta)[\nabla\theta] - e\varphi\,\mathbf{q}\cdot\dot{\mathbf{n}} \qquad \text{in } \Omega\times[0\,,\tau)$$

$$-\epsilon_o\Delta\varphi = e\mathbf{q}\cdot\mathbf{n}$$

Recovers and generalize known results, *e.g.* Vasil'ev 2008, Bizzarri, Moses, Williams, Vasil'ev et al. 2009-2017.

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Adimensionalized equation

In the Isothermal case, with $\theta = \theta_o$, for (L^*, T^*) a characteristic length/time pair, let:

$$z = \frac{x}{L^*}, \quad \tau = \frac{t}{T^*}, \quad \mathbf{n}^* = (L^*)^3 \mathbf{n}, \quad \varphi^* = \frac{\epsilon_o L^*}{e} \varphi,$$

then we have:

$$d \, \mathrm{div}_z \bar{\mathbb{D}} \nabla_z \mathbf{n}^* + m \, \mathrm{div}_z (\bar{\mathbb{M}} \mathbb{N}(\mathbf{n}^*) \mathbf{q} \otimes \nabla_z \varphi^*) - h \, \bar{\mathbb{K}} \mathbf{n}^* = \mathbf{n}^*_\tau ,$$

where

$$d = \frac{T^* k_B \theta_o}{e (L^*)^2} \|\mathbb{M}\|, \quad m = \frac{e T^*}{\epsilon_o (L^*)^3} \|\mathbb{M}\|, \quad h = T^* \|\mathbb{K}\| .$$

Notice that h depends on the incoming energy.

then:

- for $h >> \max\{d\,, m\}$ we obtain the *kinetic* model (pure reaction without diffusion and drift), widely used by physicists,

$$-h\,\bar{\mathbb{K}}\mathbf{n}^* = \mathbf{n}^*_{\tau}\,,$$

- whereas for and $d >> \max\{h\,, m\}$ we recover the *diffusive* model (pure diffusion without drift):

$$d\,\mathrm{div}_{\,z}\bar{\mathbb{D}}\nabla_{z}\mathbf{n}^* = \mathbf{n}^*_{\tau}\,,$$

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- Many recent papers on this kind of equation, mainly by L. Desvillettes, K. Fellner, A. Jüngel, J. Fischer and co-workers, 2006-2018;
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Existence: J. Fischer, 2016, X. Chen and A. Jüngel, 2017

Main result: Global existence of *Weak Solutions*. If:

(H1) Drift term: $\nabla\varphi \in L^\infty([0\,,\tau)\,;L^\infty(\Omega))$;

(H2) Reaction term: $\mathbb{K}(\mathbf{n})\mathbf{n} = \mathbb{H}\boldsymbol{\mu} \in L^1([0\,,\tau)^m;\mathcal{M})$;

(H3) Initial data: $\mathbf{n}_o \in \mathcal{M}$ is measurable and the total initial entropy is finite;

(H4) $\exists \pi_i > 0\,, \exists \lambda_i \in \mathbb{R}$, $i = 1, 2, \ldots, k$: $\forall \mathbf{n} \in \mathcal{M}$

$$\sum_{i=1}^{m} \pi_i (\mathbb{K}(\mathbf{n})\mathbf{n})_i (c_i \log n_i + \lambda_i) \leq 0\,;$$

(H5) The mobility matrix $\mathbb{M}$ is symmetric and positive-definite;

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…then, for any $\mathbf{v} \equiv (v_1, \dots v_k) \in C^\infty([0, \tau)^k, \mathcal{M})$, $\mathbf{n}$ is a *Global Weak Solution* if:

$$\int_\Omega (\mathbf{v} \cdot \mathbf{n}) \Big|_0^\tau - \int_0^\tau \int_\Omega \mathbf{n} \cdot \dot{\mathbf{v}} = - \int_0^\tau \int_\Omega \mathbb{S}[\nabla \boldsymbol{\mu}] \cdot \nabla \mathbf{v} + \mathbb{H} \boldsymbol{\mu} \cdot \mathbf{v} \, .$$

The existence of a weak solution is the starting point to obtain numerical solutions (FEM-like) for this R-D-D equation.

Asymptotic Decay: K. Fellner, 2017

Main result: explicit estimate of asymptotic decay for $k = 2$, *i.e.* $\mathbf{n} \equiv (n_1\,, n_2)$.

For $(\mathbf{n}_\infty\,, \varphi_\infty)$ an asymptotic stationary solution (*i.e.* a solution with $\dot{\mathbf{n}} = 0$):

$$\mathbf{n}_\infty = \frac{1}{k_B \theta} (e^{-e\varphi_\infty \mathbf{q}}) \mathbf{c}\,,$$

Provided:

- $0 < K_1 \leq \|\mathbb{K}(\mathbf{n})\|_{L^\infty(\Omega)}\,;$
- $\|\mathbf{n}_\infty\|_{L^1(\Omega)} \leq e^{2\Phi_\infty}$, with $\Phi_\infty = e\|\mathbf{q}\varphi_\infty\|_{L^\infty(\Omega)}$
- $L(\Omega)$ is the Poincaré constant of Ω

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then we have the estimate:

$$\|\mathbf{n} - \mathbf{n}_\infty\|^2_{L^1(\Omega)} + \|\varphi - \varphi_\infty\|^2_{H^1(\Omega)} \leq C_2(\int_\Omega \psi(\mathbf{n}_o\,, \varphi_o))e^{-C_1 t}\,,$$

with C_1 and C_2 that depend explicitly on the R-D-D equation parameters. C_2 gives an estimate of the maximum number of excitation carriers in terms of the initial Gibbs free-energy:

$$C_2 = (3e^{2\Phi_\infty} + \frac{1}{2}(\int_\Omega \psi(\mathbf{n}_o\,, \varphi_o)) + \frac{2}{\varepsilon_o}(1 + L(\Omega))$$

Decay Time

The constant $C_1^{-1} = \tau_d$ is a mathematical estimate of the Decay Time:

$$C_1^{-1} = \frac{1}{2} e^{2\Phi_\infty} \max\{\frac{\epsilon_o}{M^*} e^{2\Phi_\infty}, \frac{1}{K_1}\} \cdot (1 + \frac{L(\Omega)}{\epsilon_o} e^{2\Phi_\infty}),$$

where M^* is the smallest eigenvalue of M ($\approx$ lowest excitation carrier mobility).

Important remark I: mechanical stress

It is:

$$\tau_d \propto \epsilon_o \,,$$

in a stressed crystal (applied loads, residual stresses) by photo elasticity we have $\epsilon_o = \epsilon_o(\sigma)$, hence

$$\tau_d = \tau_d(\sigma)\,.$$

The Decay Time may depend on the mechanical stress σ.

Important remark II: anisotropic crystal

In an anisotropic crystal

$$\epsilon_o = \inf\{\epsilon_1\,, \epsilon_2\,, \epsilon_3\}\,,$$

and hence the decay time depends on the crystal *highest* refraction index

$$\tau_d = \tau_d(n_{max})\,, \quad n_{max} = \sup\{n_1\,, n_2\,, n_3\}.$$

Example. NaI:Tl

We have $n_1 = n_{ex}$ and $n_2 = n_{eh}$ with $q_1 = q_2 = 0$ and $\varphi_\infty = 0\,, \mathbf{n}_\infty = 0$.

- $L^* = 1$ nm;
- $T^* = 1$ ns;
- $K_1 = 6.61 \cdot 10^6$ sec^{-1};
- $\Phi_\infty = 0$
- $L(\Omega) = 0,068$ (unit sphere);
- $M^* = 8 \cdot 10^4$ m^2/V sec;
- $\epsilon_o = 7.29\,\epsilon_{vac}$;

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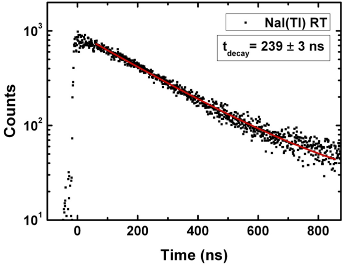
Example. NaI:Tl

$$\tau_d = C_1^{-1} = \frac{1}{2}\max\{\frac{T^*}{m}, \frac{T^*}{h}\}\cdot(1+L(\Omega)) = 249\ \text{ns}\,.$$



(from Sibczynski *et al.*, 2012)

The error is 3%.

Further developments

- Search for the internal length an time scale L^* and T^* (Onsager radius?)
- Obtain asymptotic decay estimate for $k > 2$;
- Give a mathematically consistent definition of local Light Yield based on the previous estimates;
- Extend to stress, defects and radiation damage.

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Es gibt nichts mehr praktisch als eine gute theorie

(L. Prandtl 1875-1953)

Thanks for the attention!