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A brief overview of existence results and decay time estimates for a mathematical modeling of scintillating crystals

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Inorganic scintillating crystals can be modelled as continua with microstructure. For rigid and isothermal crystals, the evolution of charge carriers becomes in this way described by a reaction-diffusion-drift equation coupled with the Poisson equation of electrostatic. Here, we give a survey of the available existence and asymptotic decays results for the resulting boundary value problem, the latter being a direct estimate of the scintillation decay time. We also show how to recover various approximated models which encompass also the two most used phenomenological models for scintillators, namely, the kinetic and diffusive ones. Also for these cases, we show, whenever it is possible, which existence and asymptotic decays estimate results are known to date.

KEYWORDS

entropy methods, existence of solutions of PDE, exponential rate of convergence, reaction-diffusion-drift equations, scintillators

MSC CLASSIFICATION

35K57; 35B40; 35B45

1 | INTRODUCTION

A scintillator crystal is a material which converts ionizing radiations into photons in the frequency range of visible light, hence its name. It acts as a true "wavelength shifter" and in such a role is used as radiation sensor into high-energy physics, in medical imaging and in security applications.[1] The physics of scintillation, which is a complex multi-scale phenomenon (see, e.g., Vasil'ev and Getkin[2]) can be described within a continuum approach at three scales: at a *microscopic* scale, the incoming energy E generates a population of charged energy carriers which moves in straight directions for few nanometer[3] and whose density $N = N(E)$ can be found by the means of approximated solutions of the Bethe–Bloch equation.[4–7*] These energy carriers wander and migrate within a greater region either generating other energy carriers or recombining with emission of photons $h\nu$. In the process, some energy is lost, and a scintillator is a material in which

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*In Daví,[8] we show how the density of excitation carriers N induced by an ionizing energy E which hits the crystal at a given point x^* can be obtained by the means of a suitable rescaling to the mesoscopic scale of the approximate solutions of the Bethe–Bloch equation along an elementary cylindrical track: in such a way $N = N(E)$ maintains information on both the initial energy E and the material properties of the crystal.

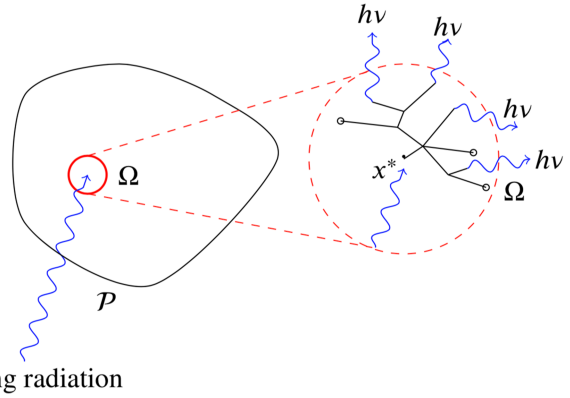

FIGURE 1 The mesoscopic scintillation region Ω [Colour figure can be viewed at wileyonlinelibrary.com]

such a loss reduces the frequency of the incoming ionizing energy to that of visible light. We call this scale the *mesoscopic* scale: for $\mathcal{P}$, the region occupied by the crystal we denote $\Omega \subset \mathcal{P}$ the *mesoscopic volume* in which the recombination of charge carriers into photons takes place (Figure 1). Finally the light rays propagate within the crystal at a *Macroscopic* scale according to the laws of classical optics.

Scintillation is a fast and dissipative phenomenon, and there are two major physical parameters which are to be improved in a scintillating material: (i) the *decay time* τ_d, which is the time required for scintillation emission to decrease to e^{-1} of its maximum and is a measure of the scintillator resolution; (ii) the *light yield LY*, which is the ratio between the collected light energy and the energy of the incoming ionizing radiation and which is a measure of the scintillator efficiency.

Scintillators can be modelled as Continua with microstructure[8–11] to arrive at a reaction-diffusion-drift (RDD) equation for the energy carriers descriptors, coupled with the Poisson equation of electrostatic and both with Neumann-type boundary conditions. Further in Daví,[12] by following the results obtained in previous studies,[13,14] we showed that for these equations, it is possible to proof the global existence of renormalized and weak solutions and also how the decay time can be estimated explicitly in terms of these equations constitutive parameters.

The continuum mechanics approach allows for a coherent thermodynamical treatment of the charge carriers evolution and recombination at the mesoscopic scale. Sources of dissipation, constitutive couplings, the role of entropy, and entropic contributions arises quite naturally within this framework and not as ad hoc assumptions based on physically similar phenomena. There exists indeed an huge literature which describes, at the same scale, either the kinetic of chemical reactions or the pair production and recombination in semiconductors; this is in turn associated to many recent and very important mathematical results mainly concerned with the existence of various kind of solutions and the asymptotic decay estimates, as one can see from the long list of references at the end of this paper. The continuum mechanics approach we choose allows for a correct transposition of all these results into a physically different phenomenon like scintillation is.

Therefore, by using the results obtained into Chen and Jungel,[15] we expand the results of Daví[12] to show the existence of weak-strong renormalized solutions: we further show also (whenever it is possible) existence and asymptotic decay results for the phenomenological models widely used in the literature, which can be obtained from our model by introducing suitable approximations as we did into Daví.[9]

We remark that scintillation is strongly affected by temperature, and the crystal is linearly elastic deformable bodies, but here we limit our analysis to the isothermal case (the temperature being at most a parameter in the constitutive quantities) and to rigid crystals as in Daví.[9] The effects of temperature are dealt with into Daví[8,10] whereas an insight into the deformable case is provided in a previous study.[11] A complete treatment of these electromagnetical, thermal and mechanical interactions will be provided in a forthcoming book.[16]

The paper is organized as follows: in Section 2, we give an overview of the model proposed into previous works,[8–11] which leads to a RDD system: then we study the properties of the stationary solutions, which are important when we deal with the solution asymptotic properties.

In Section 3, we extend, with the help of Chen and Jungel,[15] results presented in Daví[12] and we give results on existence of weak-strong renormalized solutions and on asymptotic decay, this last result being a direct estimate of the decay time. This result allows, for the first time, to estimate the decay time in terms of the parameters describing the physical properties of a given scintillating crystal and which appears explicitly in the equations we obtained.

Finally in Section 4, first of all, we put the coupled RDD and Poisson equations in an adimensional form by introducing characteristic length and time related to the scale of scintillation, as we did into Daví.[9] The resulting boundary value

problem depends on a set of three adimensional parameter related to diffusion, drift, and recombination, respectively. Then by choosing a suitable measure of smallness, we obtain, to within higher order terms in such a measure, different approximated models which represents different physically meaningful regimes for scintillators. In such a way we not only recover the most used phenomenological model in use, namely, the *Kinetic* and *Diffusive* ones, but also show under which hypotheses scintillation could be described either by a reaction-diffusion or a diffusion-drift equation. Also for these approximated boundary value problems, we give a survey of the existence and asymptotic decay results, either by taking them straight from the available literature or by adapting them to the specific context of scintillation as described by our model.

2 | THE CONTINUUM MODEL FOR ISOTHERMAL AND RIGID SCINTILLATORS: THE BOUNDARY VALUE PROBLEM

2.1 | A reaction-diffusion-drift equation for scintillators

The charge carriers with density N, generated at the point x^* and time t^* and whose dimension is $(\text{length})^{-3}$, represent a population of carriers which can differ by their sign (e.g., negative electrons, positive holes, neutral excitons, and so on) and by their recombination mechanism. Accordingly, we may differentiate these carriers by introducing an ordered array n, the *charge carrier densities* vector:

$$n \equiv (n_1, n_2, \dots n_k),\ N = \sum_{j=1}^{k} n_j; \tag{1}$$

clearly, the greater is k, the finer will be our description of the phenomena: the simplest non-trivial choice is $k = 2$ as in Li et al,[17] where n_1 represents the electrons population (which is equal to the holes population) and n_2 represents the population of excitons, which are bounded electron-hole pairs evolving together. In the various phenomenological models for scintillation thus far proposed, we may have $m = 3$ as in Williams et al[18] and Moses et al.,[19] $m = 7$ as in Gridin et al[20] whereas in Vasil'ev,[21] we have $m \geq 11$. The charge carrier densities are related to N by

$$n_j = \alpha_j N,\ \alpha_j \geq 0,\ j = 1, 2, \dots, k,\ \sum_{j=1}^{k} \alpha_j = 1; \tag{2}$$

the set $\{\alpha_1, \alpha_2, \dots, \alpha_k\}$ depends on the specific scintillator, on the initial energy E and on the initial data (cf. the discussion in Bizzarri et al.[22]).

We identify the charge carriers densities at (x^*, t^*) with fields defined on the whole $\Omega \times [0, \tau)$ and whose regularity will shall made precise when needed:

$$\Omega \times [0, \tau) \ni (x, t) \mapsto n_j(x, t) \geq 0,\ j = 1, 2, \dots k; \tag{3}$$

without loss of generality we may assume that the mesoscopic volume Ω is a ball of a still unprescribed radius R, centered at x^*.

We assume accordingly the k-dimensional field, the *excitation carrier densities* vector

$$\Omega \times [0, \tau) \ni (x, t) \mapsto n(x, t) \in \mathcal{M},\ \mathcal{M} = \mathbb{R}^k_+ \cup \{0\}, \tag{4}$$

as the main state variable in our description of scintillation. In view of (1), since $n_j \geq 0$, $j = 1, 2, \dots, k$, we can view the extension of N to a field on Ω as the $L^1(\Omega)$ norm of n:

$$\|n\|_{L^1(\Omega)} = \int_\Omega \sum_{j=1}^{k} |n_j| = \int_\Omega N. \tag{5}$$

Let e be the elementary charge, then the excitation carrier vector induces a free charge density within the scintillation volume Ω

$$\rho_f = ez \cdot n,\ \text{in}\ \Omega, \tag{6}$$

with $z=(z_1,z_2,\ldots z_k), z_j\in\mathbb{Z}, j=1,2,\ldots k$ the *charge vector*. By the Maxwell-Lorentz equations in absence of magnetic fields,[23] these free charges induce a *local electric potential* $(x,t)\mapsto\varphi(x,t)$ which, for a given time $t\in[0,\tau)$, is the solution of the Poisson equation of electrostatic with associated Neumann boundary conditions:

$$\begin{aligned} -\epsilon\Delta\varphi &= \chi|_{\Omega} ez\cdot n, \text{ in } \mathbb{R}^3\times[0,\tau),\\ [\![\nabla\varphi]\!]\cdot\mathbf{m} &= 0, \text{ on } \partial\Omega\times[0,\tau), \end{aligned} \tag{7}$$

where ϵ is the permittivity of the crystal (which at this stage we assume isotropic or at most cubic)[†]

and $\mathbf{m}$ is the outward unit normal to $\partial\Omega$ and

$$\chi|_{\Omega}=\begin{cases} 1, \text{ in } \Omega,\\ \\ 0, \text{ in } (\mathbb{R}^3/\Omega). \end{cases} \tag{9}$$

We notice that in (7), we do not take into account either bound charges or external charges since we are mainly interested into X- or γ-rays which have zero charge, whereas for α- and β-rays, which have, respectively, positive and negative charges, an external charge contribution q^* should be added to (7).

We further remark that an implicit way to select the radius of Ω is indeed that at its boundary, the Neumann condition holds, that is, there is no electric field outflow trough $\partial\Omega$.

The boundary value problem (7) admits an unique (up to a constant) weak solution $\varphi\in H^1(\Omega)$ provided the *total charge* is conserved:

$$Q(t)=e\int_{\Omega} z\cdot n=0,\ \forall t\in[0,\tau), \tag{10}$$

and the constant can be conveniently determined if we set $\bar{\varphi}=0$,[‡] thus making the solution unique.

Following Albinus et al,[24] we assume that the *electrostatic free-energy* associated to the electric potential has, besides the classical conservative term which depends on n by the means of (7), a dissipative term $F(n)$ of entropic nature which also depends on the excitation carrier vector:

$$U(n)=\frac{1}{2}\int_{\mathbb{R}^3}\epsilon\|\nabla\varphi(n)\|^2-\theta\int_{\Omega}F(n), \tag{11}$$

where $\theta>0$ is the (fixed) absolute temperature; in Daví,[10] we showed how to such an energy it can be associated an electrostatic self-power:

$$\dot{U}=\int_{\Omega} s\cdot\dot{n}, \tag{12}$$

where the elements of the array $s\equiv(s_1,s_2,\ldots s_k)$ represent the *scintillation potentials* associated to the various charge carriers:

$$s_j(n)=ez_j\varphi(n)-\theta\frac{\partial F}{\partial n_j}(n),\ j=1,2,\ldots,k. \tag{13}$$

We shall call s, with an abuse of terminology, the *scintillation potentials vector*:

$$s(n)=ez\varphi(n)-\theta\frac{\partial F}{\partial n}(n), \tag{14}$$

[†] For anisotropic crystals equations (7) become:

$$\begin{aligned} -\epsilon_o\mathrm{div}\mathbf{K}[\nabla\varphi] &= \rho^*, \text{ in } \mathbb{R}^3\times[0,\tau),\\ [\![\mathbf{K}\nabla\varphi]\!]\cdot\mathbf{m} &= 0, \text{ on } \partial\Omega\times[0,\tau), \end{aligned} \tag{8}$$

where ϵ_o is the vacuum permittivity and $\mathbf{K}$ is the symmetric and positive definite *permittivity tensor*; relations (11) changes accordingly with $\epsilon_o\mathbf{K}[\nabla\varphi]\cdot\nabla\varphi$ in place of $\epsilon\|\nabla\varphi\|^2$

[‡] For any given integrable function f, we shall denote with $\bar{f}$ its mean value:

$$\bar{f}=\frac{1}{\mathrm{meas}(\Omega)}\int_{\Omega} f.$$

and it is easy to show that

$$s = \mathcal{D}U, \tag{15}$$

where $\mathcal{D}$ denotes the Frechet derivative.

In Daví[8] and the related papers,[9,10] we showed how, by using a continuum with microstructure approach and the classical dissipation inequality, the evolution equation for the charge carriers in scintillators can be represented as

$$\begin{aligned} &\mathrm{div} S(n)[\nabla s] - H(n)s = \dot{n}, \ \text{ in } \ \Omega \times [0,\tau), \\ &S(n)[\nabla s]\cdot \mathbf{m} = 0, \ \text{ on } \ \partial\Omega \times [0,\tau), \end{aligned} \tag{16}$$

where $S(n)$ and $H(n)$ are two definite-positive $k\times k$ matrices and

$$\nabla s = ez \otimes \nabla\varphi - \theta \frac{\partial^2 F}{\partial n^2}\nabla n. \tag{17}$$

The boundary value problem (16) has an equivalent variational formulation which leads to a gradient-flow type problem (vid., e.g., previous studies[25,26] and for more recent results[27]):

$$\dot{n} = -\mathcal{D}\Psi^*(n,s), \ \Psi^*(n,s) = \frac{1}{2}\int\limits_{\Omega} S(n)[\nabla s]\cdot\nabla s + H(n)s\cdot s, \tag{18}$$

where the *conjugate dissipation* functional $\Psi^*(n\,,s)$ is related to the thermodynamical dissipation $\mathfrak{D}$ by

$$\mathfrak{D} = 2\Psi^*. \tag{19}$$

The gradient-flow classical structure for a driving functional U is

$$\mathbb{G}(n)\dot{n} = -\mathcal{D}U; \tag{20}$$

starting with Mielke,[28] provided there exists $\mathbb{K}(n) = \mathbb{G}^{-1}(n)$, the following formulation was proposed and successfully used into, e.g.,[29–36]

$$\dot{n} = -\mathbb{K}(n)[\mathcal{D}U], \tag{21}$$

where the Onsager structure $\mathbb{K}$ can be splitted additively into different contributions. For instance, in our case, we may set:

$$\mathbb{K} = \mathbb{K}_D + \mathbb{K}_R, \ \mathbb{K}_D[\cdot] = -\mathrm{div} S(n)\nabla[\cdot], \ \mathbb{K}_R[\cdot] = H[\cdot]. \tag{22}$$

Furthermore, in Daví,[8] it is shown that, provided we identify the entropic term with the Gibbs entropy,

$$F(n) = -k_B\sum_{j=1}^{k} n_j\left(\log\left(\frac{n_j}{c_j}\right) - 1\right), \tag{23}$$

with c_j, $j = 1, 2, \ldots, k$, normalizing constants and k_B the Boltzmann constant then, by using (23) into (14) and (17), we arrive at a reaction-diffusion-drift equation for the evolution of charge carriers in scintillators with Neumann boundary conditions and initial data:

$$\begin{aligned} &\mathrm{div}(D[\nabla n] + M[N(n)z\otimes\nabla\varphi]) - r(n) = \dot{n}, \ \text{ in } \ \Omega\times[0,\tau), \\ &\quad D[\nabla n]\mathbf{m} = 0, \ \text{ on } \ \partial\Omega\times[0,\tau); \\ &\quad -\epsilon\Delta\varphi = \chi|_{\Omega} ez\cdot n, \ \text{ in } \ \mathbb{R}^3\times[0,\tau), \\ &\quad [[\nabla\varphi]]\cdot\mathbf{m} = 0, \ \text{ on } \ \partial\Omega\times[0,\tau), \\ &\quad n_o(x) = n(x,0), \ \ \varphi_o(x) = \varphi(x,0), \text{ in } \ \Omega. \end{aligned} \tag{24}$$

In (24), the $k\times k$ matrix $N(n)$ is defined as $N(n)=\mathrm{diag}\{n_1,n_2,\ldots n_k\}$, the $k\times k$ symmetric and semi-definite positive *Diffusion* and *Mobility* matrices D and M are correlated by the Einstein–Smoluchowsky relation:

$$D=\frac{\theta k_B}{e}M, \tag{25}$$

and $r(n)$ is the k-dimensional *recombination* array.

The relation between the matrix S in (16) and the matrix M in (24) is[§]

$$M=eS(n)N^{-1}(n), \tag{26}$$

where $N^{-1}(n)$ denotes the diagonal $k\times k$ matrix whose entries are n_j^{-1} when $n_j\neq 0$ and 0 when $n_j=0$. The semi-definite positiveness of M accounts for excitation carriers whose mobilities (and hence by (25) the associated diffusivities) are either zero or negligible with respect to those of other charge carriers. However, in Section 3, we shall see that in order to get decay time estimates, we shall require the stronger requirement of positive-definiteness for M.

Finally, the $k\times k$ matrix $H(n)$ and the k-dimensional array $r(n)$ are related by

$$H(n)s=r(n), \tag{27}$$

where, by following Mielke,[28] we assume that

$$H(n)=\sum_{h=1}^{s}k_h\ell\left(\frac{n^{a^h}}{c^{a^h}},\frac{n^{b^h}}{c^{b^h}}\right)\left(a^h-b^h\right)\otimes\left(a^h-b^h\right),\ v^a=\prod_{j=1}^{k}v_j^{a_j}. \tag{28}$$

In (28) $h=1,2,\ldots s$ denote the number of recombination processes with rates k_h with the two k-dimensional arrays $a^h=(a_1^h,a_2^h,\ldots a_k^h)$ and $b^h=(b_1^h,b_2^h,\ldots b_k^h)$ describing the h^{th} recombination mechanism with rate k_h:

$$\mathrm{a^h}\overset{\mathrm{k_h}}{\rightleftharpoons}\mathrm{b^h},\qquad \mathrm{h}=1,2,\ldots \mathrm{s}, \tag{29}$$

and the function $\ell(x,y)$ is the logarithmic mean:

$$\ell(x,y)=\begin{cases}\frac{x-y}{\log x-\log y},\ x\neq y,\\ x,\ x=y.\end{cases} \tag{30}$$

Whenever we assume for $F(n)$ the Gibbs entropy (23), then from (27) and (28), we arrive at a polynomial expression for the recombination term $r(n)$ (vid., e.g., Glitzky and Mielke[29] for the details):

$$r(n)=\sum_{h=1}^{s}k_h\left(\frac{n^{a^h}}{c^{a^h}}-\frac{n^{b^h}}{c^{b^h}}\right)(a^h-b^h), \tag{31}$$

where we used the identity $\log v^a=a\cdot\log v$. In order to arrive to (31), we implicitly assumed that all the recombination mechanisms are *detailed balanced*[28]; that is, there exists a steady recombination state which in our case coincides with c; further, let

$$\mathcal{S}\equiv\mathrm{span}\{a^i-b^i|i=1,2,\ldots s\}\subset\mathbb{R}^k, \tag{32}$$

and

$$\mathcal{S}^{\perp}\equiv\{v\in\mathbb{R}^k|v\cdot u,\ \forall u\in\mathcal{S}\}, \tag{33}$$

[§]We assume that the mobility is independent on n, as pointed out into Khodyuk and Dorenbos[37]: in such a case, $S(n)$ must be restricted to the form

$$S(n)=S_oN(n),$$

where S_o is a $k\times k$ matrix with constant components.

then from (31), we have

$$r(n) \in \mathcal{S}. \tag{34}$$

If we consider now the differential form of the charge conservation (10), namely,

$$\frac{\mathrm{d}}{\mathrm{d}t}Q(t) = e\int_{\Omega} z\cdot \dot{n} = 0,\ \forall t \in [0,\tau), \tag{35}$$

then from (24), we obtain

$$0 = \int_{\Omega} r(n)\cdot z = \overline{r(n)}\cdot z, \tag{36}$$

which in turn, by (31), implies the electrical neutrality of each recombination mechanism:

$$z\cdot(a^h - b^h) = 0,\ h = 1,2,\ \dots\ s, \tag{37}$$

and hence $z \in \mathcal{S}^{\perp}$ in such a way that the local orthogonality condition holds:

$$r(n)\cdot z = 0. \tag{38}$$

In Mielke,[28] it is remarked that the term $r(n)$ can be represented as a polynomial relation in n only if we identify $F(n)$ with the Gibbs entropy (23), whereas for a different entropic term like, e.g., the Fermi–Dirac potentials, this is not possible.

Equation $(24)_1$, which represents the conservation of electric current normalized with respect to e, was first proposed for scintillators in Vasil'ev[21] by following Fok[38] and Antonov-Romanovskiy[39] and with $r(n)$ a polynomial, at most cubic, function of n; it was used for scintillators in previous works.[17,18,37,40–42] Moreover, special cases of this equations were widely used to model scintillation at a phenomenological level in many theoretical and experimental paper, as we shall describe in details in Section 4. We also remark that the boundary value problem (24) is the same obtained, by starting from a different approach and with a different reaction term $r(n)$, in Albinus et al,[24] for semiconductors (vid. also previous works[28,30]).

In the available phenomenological models for scintillation, the recombination term $r(n)$ is generally assumed as a cubic expression in n:

$$r_i(n) = r_i^0 + \sum_{j=1}^{k} A_{ij}n_j + \sum_{h,j=1}^{k} B_{ijh}n_h n_j + \sum_{h,m,j=1}^{k} C_{ijhm}n_h n_m n_j,\ i = 1,2,\ \dots\ ,k. \tag{39}$$

The terms r_i^o describes the excitation carriers creation rate in the crystal under irradiation; the other terms in (39) are further splitted in order to represent different recombination mechanism. The terms A_{ij}, which accounts for linear recombination, can be further decomposed into three terms:

$$A_{ij} = A_{ij}^r + A_{ij}^{nr} + A_{ij}^e, \tag{40}$$

which represent, respectively, radiative recombination with photon emission, non-radiative recombination without photon emission and exchange between excitation carriers. The quadratic recombination is represented by the terms B_{ijh} which can also be splitted into $B_{ijh} = B_{ijh}^r + B_{ijh}^{nr}$ with the same meaning of superscript as in (40), whereas the third-order or Auger totally non-radiative recombination is described by the terms $C_{ijhm} = C_{ijhm}^{nr}$.

In order to reconcile (39) with (31) we consider here the following example for $k = 3$ where $n_1 = n_e$ and $n_2 = n_h$ represent respectively the electrons and holes densities and $n_3 = n_{ex}$ represents the excitons density. By following, for instance, the description provided in section 3.3 of Korzhik et al,[43] then we may have these recombination mechanisms:

1. $n_1 + n_2 = 0,\ a^1 = (1,1,0),\ b^1 = (0,0,0),$
2. $n_1 + n_2 = n_3,\ a^2 = (1,1,0),\ b^2 = (0,0,1),$
3. $n_1 + n_2 = n_1 + n_2 + n_3,\ a^3 = (1,1,0),\ b^3 = (1,1,1),$
4. $n_1 = 2n_1 + n_2,\ a^4 = (1,0,0),\ b^4 = (2,1,0),$
5. $n_1 = n_1 + n_3,\ a^5 = (1,0,0),\ b^5 = (1,0,1),$

6. $n_2 = n_1 + 2n_2,\ a^6 = (0,1,0),\ b^6 = (1,2,0),$
7. $n_2 = n_2 + n_3,\ a^7 = (0,1,0),\ b^7 = (0,1,1),$

and then from (31), we get

$$\begin{aligned} r_1(n) = r_2(n) = &- k_1 - \frac{k_4}{c_1} n_1 - \frac{k_6}{c_2} n_2 - \frac{k_2}{c_3} n_3 \\ &+ \frac{k_1 + k_2}{c_1 c_2} n_1 n_2 + \frac{k_4}{c_1^2 c_2} n_1^2 n_2 + \frac{k_6}{c_1 c_2^2} n_1 n_2^2, \\ r_3(n) = &- \frac{k_3}{c_1} n_1 - \frac{k_7}{c_2} n_2 + \frac{k_2}{c_3} n_3 \\ &+ \frac{k_2 - k_3}{c_1 c_2} n_1 n_2 + \frac{k_5}{c_1 c_3} n_1 n_3 + \frac{k_7}{c_2 c_3} n_2 n_3, \end{aligned} \tag{41}$$

which can be trivially put into the form (39), with the appropriate identification of radiative, non-radiative or exchange terms: we notice that the mechanisms 3, 4 and 6 are Auger recombination, whereas 2, 5 and 7 represents scattering and the mechanism 1 is the simple electron-hole recombination (cf., e.g., the models proposed either into Bizzarri et al[22] or Bizarri et al[44] where $n_e = n_h = n_{eh}$).

As we already remarked more complex expressions for (39) can be proposed: for instance, in Gridin et al,[20] for $k = 7$, we had

$$\begin{aligned} r_1(n) &= r_1^0 + A_{14} n_4 + B_{113} n_1 n_3 + B_{115} n_1 n_5 + B_{117} n_1 n_7, \\ r_2(n) &= r_2^0 + A_{23} n_3 + A_{25} n_5 + B_{223} n_2 n_3 + B_{224} n_2 n_4 + B_{227} n_2 n_7, \\ r_3(n) &= A_{33} n_3 + B_{323} n_2 n_3 + B_{336} n_3 n_6, \\ r_4(n) &= A_{44} n_4 + B_{417} n_1 n_7 + B_{424} n_2 n_4, \\ r_5(n) &= A_{55} n_5 + B_{515} n_1 n_5 + B_{527} n_2 n_7, \\ r_6(n) &= r_6^0 + A_{66} n_6 + B_{613} n_1 n_3, \\ r_7(n) &= A_{77} n_7 + B_{715} n_1 n_5 + B_{724} n_2 n_4, \end{aligned} \tag{42}$$

with n_1 and n_2 the electron and holes densities, n_3 and n_6 the self-trapped electrons and excitons, respectively, n_4 and n_5 the electron and holes captured by the activation centers and finally n_7 denotes the excited activator centers: in Gridin et al,[20] a detailed description of the physical motivation of these relations is provided. We notice that in this model the Auger mechanism is missing, and the recombination terms are at most quadratic.

2.2 | Stationary solutions

It is trivial to show that the stationary solutions, that is equilibrium solutions n^∞ for the boundary value problem (24) with $\dot{n} = 0$, can be obtained by setting $s = 0$, which by (27) leads to the equilibrium condition:

$$r(n^\infty) = 0. \tag{43}$$

When $F(n)$ is identified with the Gibbs entropy (23), then from $s = 0$, we have

$$s_j = e z_j \varphi^\infty + \theta k_B \log \frac{n_j^\infty}{c_j} = 0,\ j = 1, 2, \dots, k, \tag{44}$$

where the stationary electric field φ^∞ is the solution of

$$-\epsilon \Delta \varphi^\infty = e \sum_{j=1}^{k} z_j n_j^\infty, \text{ in } \Omega, \tag{45}$$

with Neumann boundary conditions on $\partial\Omega$. From (44), then we have

$$n_j^\infty(x) = c_j \exp\left(-\frac{ez_j\varphi^\infty(x)}{\theta k_B}\right),\ j = 1, 2, \dots, k, \tag{46}$$

and (45), (46) together leads to the semilinear Poisson-Boltzmann equation for the stationary electric field:

$$-\epsilon\Delta\varphi^\infty = e\sum_{j=1}^{k} z_j c_j \exp\left(-\frac{ez_j\varphi^\infty}{\theta k_B}\right), \text{ in } \Omega; \tag{47}$$

in Li,[45] a uniqueness result in $H^1(\Omega)$ for (47) with Dirichlet boundary conditions was obtained.

It is trivial to show that the equilibrium solution n^∞ is also a steady state c for the detailed balance condition, and indeed, in (27), we have

$$\frac{(n^\infty)^{a^h}}{c^{a^h}} - \frac{(n^\infty)^{b^h}}{c^{b^h}} = \exp\left(-\frac{e\varphi}{k_B\theta}(z\cdot a^h - z\cdot b^h)\right) = 0, \tag{48}$$

by (46) and (37), and hence, the equilibrium condition (43) is satisfied.

We finally deal with the problem of the determination of $c = (c_1, c_2, \dots, c_k)$ in (46): if we define the $k\times k$ diagonal matrix $L(x)$

$$L(x) = \text{diag}\left\{\exp\left(-\frac{ez_1\varphi^\infty(x)}{\theta k_B}\right), \exp\left(-\frac{ez_2\varphi^\infty(x)}{\theta k_B}\right), \dots, \exp\left(-\frac{ez_k\varphi^\infty(x)}{\theta k_B}\right)\right\}, \tag{49}$$

then (46) can be written as

$$n^\infty(x) = L(x)c. \tag{50}$$

and then, by (10) $\overline{L(x)}c \in S$.

To obtain the explicit value of c, we may use (43), whereas the uniqueness of c follows instead, as pointed out in Fellner and Kniely,[14] by monotonicity, (10) and the uniqueness of $\varphi^\infty \in H^1(\Omega)$ with $\bar{\varphi}^\infty = 0$.

3 | EXISTENCE AND ASYMPTOTIC DECAY

In this section, we shall give an account of the existing results concerning the existence of solutions and the asymptotic estimates for the boundary value problem (24): we remark that the latter results are important in order to get a scintillation decay time estimate. We shall not enter into the mathematical details which can be found in the references we quote, rather we shall adapt if necessary these results to the specific cases of our boundary value problems.

The problem of finding existence, asymptotic estimates and qualitative bounds for the solutions for the coupled boundary value problem (24) has received a strong attention in the recent years, vid., e.g., [13–15,46–55] and the many references quoted therein: to this regard, it is important to remark that most of these results deal with semiconductors or chemical reactions which differ from scintillators by the structure of the reaction term $r(n)$. Most of these results are based on the so-called *Entropy method*, whose importance is explained in full in these words taken from Di Francesco et al.[56] (vid. also the discussion in Hopf[57]):

> *The entropy method refers to the general idea of a functional inequality relationship between an entropy functional of a system and its monotone change in time, usually called the entropy dissipation. Such an entropy-entropy dissipation inequality entails convergence to an entropy minimizing equilibrium state, at first in entropy and further in L^1 using Cziszár-Kullback-Pinsker-type inequalities. The entropy approach is per se a nonlinear method avoiding any kind of linearization and capable of providing explicitly computable convergence rates. Moreover, being based on functional inequalities rather than particular differential equations, it has the advantage of being quite robust with respect to model variations.*

In the next subsection, we shall show how some of these results can be extended to the RDD equations for scintillators. As far as the decay time is concerned, the available experimental data (vid., e.g., the recent analysis in Swiderski

et al.[58]) and the numerical solution of phenomenological models as in Lu et al,[59] show that the excitation carriers decay exponentially in time to an asymptotic value n_∞, namely:

$$\|n(\cdot,t)-n_\infty(\cdot)\| = A_f \exp(-t/\tau_f) + A_s \exp(-t/\tau_s), \tag{51}$$

where the indices *f* and *s* denotes the so-called *fast* and *slow* components of the excitation, respectively. Accordingly, since by definition the *Decay time* is the time required for scintillation emission to decrease to e^{-1} of its maximum, then we get a Fast Decay Time τ_f and a Slow Decay Time τ_s. In many cases, one of the components is negligible, and the decay obeys a simple exponential law, which can be also used to describe an average decay time.

3.1 | Global existence

The first results concerning existence theory for the boundary value problems like (24) was obtained in previous studies[52,53]: such a result, which relies on the notion of global *renormalized solutions* leaves still open, as pointed out in Fischer,[53] the problem the existence of weak or even smooth global solutions in time: the reason, as pointed out in detail into Fischer[53] is the growth condition on the reaction/recombination terms.

These results were extended into Chen and Jüngel,[13,15] and here, we shall follow the latter. The main strongpoint of these results is that no growth condition is imposed on the reaction/recombination term. In our case, the diffusion is linear: unfortunately, we cannot extend their results to the nonlinear case they study, because for (25), this would imply also a nonlinear mobility, whereas in all these paper, the mobility is implicitly assumed $M = I$ with I the $k\times k$ identity.

First of all, we recall the notion of renormalized solution, first introduced into literature[60–62] for the Boltzmann and transport equations, as it was given into Fischer.[52]

An excitation density vector n is a renormalized solutions for (24) if for all functions $\xi : \mathcal{M} \to \mathbb{R}$ with compactly supported derivative $\nabla_n\xi$, the function $\xi(n)$ must satisfy the equation derived from (24) by a formal application of the chain rule in a weak sense. As it is pointed out in Villani,[63] the function ξ must belongs to a well-chosen class of admissible solutions. The physical interpretation of these renormalized solution is that they gives a distributional sense to the boundary value problem (71).

More precisely we say that:

- $n = (n_1, n_2, \dots, n_k)$ is a *renormalized solutions* for (24) if $\forall\tau>0$, $n_i \in L^2(H^1(\Omega);[0,\tau))$ and for any $\xi \in C^\infty(\mathcal{M})$, such that $\nabla_n\xi \in C_0^\infty(\mathcal{M};\mathbb{R}^k)$ and $\psi \in C_0^\infty(\bar\Omega\times[0,\tau))$, it holds:

$$\begin{aligned}\int_0^\tau\int_\Omega \xi(n)\dot\psi &= \int_0^\tau\int_\Omega ([D\nabla n + MN(n)z\otimes\nabla\varphi]\cdot\nabla_n\nabla_n\xi[\nabla n] + r(n)\cdot\nabla_n\xi)\psi \\ &+ \int_0^\tau\int_\Omega (D\nabla n + MN(n)z\otimes\nabla\varphi)\cdot\nabla_n\xi\otimes\nabla\psi\,.\end{aligned} \tag{52}$$

We leave out all the details and recall only the main hypotheses and results given in Chen and Jungel,[15] and first of all, we assume that there exist numbers $\pi_i>0$ and $\lambda_i\in\mathbb{R}$, $i=1,2,\dots,k$ such that $\forall n\equiv(n_1,n_2,\dots,n_k)\in(0,\infty)^k$, the following inequality holds:[¶]

$$\sum_{i=1}^k \pi_i r_i(n)\left(\log\frac{n_i}{c_i}+\lambda_i\right)\geq 0, \tag{53}$$

a condition which implies the *quasi-negativity* of $r(n)$, that is (cf. the models in Gridin et al[20] and Bizzarri et al.[22]):

$$r_i(n_1,\dots,n_{i-1},0,n_{i+1},\dots,n_k)\leq 0,\ \forall n\in\mathcal{M},\forall i=1,2,\dots,k, \tag{54}$$

which grants the non-negativity of solutions.

[¶]A certain care is requested when we look at (53) and (54), since the reaction term $f(u)$ in Chen and Jungel[15] is the opposite of our recombination term $r(n)$, say $f(u)=-r(n)$. Hence, the reversed inequalities and different definition than those given into Chen and Jungel.[15]

Condition (53) further ensure the existence of a so-called *total entropy*:

$$\mathcal{H}(n) = \int_\Omega \sum_{i=1}^{k} \pi_i n_i \left(\log \frac{n_i}{c_i} - 1 + \lambda_i \right) + \exp(-\lambda_i), \tag{55}$$

which is a Lyapunov functional for the reaction system (92) if $\pi_i = 1, \forall i$.

Provided these preliminary conditions are satisfied, then the main hypotheses from Chen and Jungel[15] can be rephrased, within the context of our treatment, as

(H1) Drift term: $\nabla\varphi \in L^\infty([0,\tau); L^\infty(\Omega, \mathbb{R}^{k+3}))$;
(H2) i) Recombination term: $r(n) : \mathcal{M} \to \mathbb{R}^k$ is locally Lipschitz continuous, that is there exists a function $K(\cdot): [0,+\infty) \to [0,+\infty)$ non-decreasing and such that a.e. $(x,t) \in \Omega \times [0,\tau)$ and $\forall n, \hat{n} \in \mathcal{M}$:

$$\|r(n) - r(\hat{n})\| \leq K(\max\{\|n\|, \|\hat{n}\|\})\|n - \hat{n}\|, \tag{56}$$

and $\forall \hat{\tau} > 0$, then $r(0) \in L^2(\Omega \times [0,\hat{\tau}))$.
ii) the inequality (53) holds,
iii) there exist $m \in \mathbb{N}$ such that $\forall n \in \mathcal{M}$ with $\sum_{j=1}^k n_j \geq m$, then $\sum_{j=1}^k r_j(n) \geq 0$;
(H3) Initial data: $n_o = (n_1^0, n_2^0, \dots, n_k^0) \in L^\infty(\Omega, \mathbb{R}^k)$, such that $\inf n_j^o > 0$, $j = 1, 2, \dots, k$;
(H4) The mobility matrix M (and hence for (25) the diffusion matrix D) is diagonal and positive-definite.

From a physical point of view, the first three hypotheses requires simply a certain degree of regularity on the initial data, the electric field and the recombination term: as far as the last hypothesis is concerned, from a physical point of view the mobility matrix should be semi-definite positive to allow for charge carriers with no mobility. Hypothesis (H4), which is a truly mathematical one, rules out such possibility and also does not allow for both cross-mobility (and diffusion). It would be an useful generalization to recover the results of Chen and Jungel[15] when it is relaxed into the more physically interesting:

(H4)′ The mobility matrix M (and hence for (25) the diffusion matrix D) is semi-definite positive.

A first consequence of the hypotheses (H1)–(H4) is that there exists

- a global renormalized solution $n = (n_1, n_2, \dots, n_k)$ satisfying $n_i \geq 0$ in Ω, $i = 1, 2, \dots k, \forall t$,
- $\mathcal{H}(n) =< +\infty, \forall t$;

moreover they imply the main result of Chen and Jungel[15] (*Weak-strong uniqueness of the solutions*) which states that the boundary value problem (24):

- admits a weak-strong renormalized solution $n = (n_1, n_2, \dots, n_k)$ satisfying $n_i \geq 0$ in $\Omega \times [0,\tau)$, $i = 1, 2, \dots k, \forall t$,
- $\mathcal{H}(n) =< +\infty$, in $\Omega \times [0,\tau)$,
- $n \in (0,\tau; L^1(\Omega))$, *cf.* (5).

Finally, for n a renormalized solution to (71), $v = (v_1, \dots v_k)$ is a *strong* solution to (71) on some time interval $[0,\tau^\star)$ with $\tau^\star \leq \tau$ in the following sense: there exist $C > c > 0$ such that

- $c \leq v_i(x,t) \leq C$, $(x,t) \in \Omega \times [0,\tau^\star), i = 1, 2, \dots, k$;
- $\|\dot{v}\|_{L^\infty(\Omega\times[0,\tau^\star))} + \|\nabla v\|_{L^\infty(\Omega\times[0,\tau^\star))} \leq C$,

then if, for any $s \in (0,\tau^\star)$, $\phi \in C^\infty(\bar{\Omega} \times [0,\tau^\star))$:

$$\int_0^s \int_\Omega \phi\dot{v} = -\int_0^s \int_\Omega (D[\nabla v] + MN(v)z \otimes \nabla\varphi) \cdot \nabla\phi - \int_0^s \int_\Omega \phi r(v); \tag{57}$$

then $n(s,t) = v(s,t)$, for $x \in \Omega$, $s \in (0,\tau^\star)$.

We remember that, as pointed out into Fischer,[53] we can say nothing about the global existence in time of smooth solutions: however this result is important in order to get a general framework for numerical solutions of (71).

3.2 | Asymptotic estimate for the decay time

In Fellner and Kniely,[14] an explicit estimate of the asymptotic convergence was obtained for the Rosbroeck model for semiconductors with Shockley–Read–Hall potential and $k = 2$: here, we shall show how the results obtained there can be adapted to the case of scintillators in order to obtain an explicit estimate for the decay time. Once again we shall leave out the technicalities and details and we shall give only the main results using our language and notations.

We have already seen that we can safely assume uniqueness for c: given this in Fellner and Kniely[14] some preliminary bounds are necessary, namely, the following for $c = (c_1, c_2)$ and $n^\infty = (n_1^\infty, n_2^\infty)$:

$$c_j \le e^{\Phi^\infty}, \; n_j^\infty \le e^{2\Phi^\infty}, \; (n_j^\infty)^{-1} \le e^{2\Phi^\infty}, \; j = 1, 2, \tag{58}$$

with

$$\Phi^\infty = \left\| \frac{ez\varphi^\infty}{\theta k_B} \right\|_{L^\infty(\Omega)}. \tag{59}$$

Moreover, for the recombination $r(n)$ given by (39), there exists a constant k_o such that

$$0 < k_o \le \|r(n)\|_{L^\infty(\Omega)}. \tag{60}$$

If we consider the free-energy $U(n)$ defined by (11) with the choice of the Gibbs entropy (23) for the term $F(n)$, then the main results of Fellner and Kniely[14] are based on the derived notion of *Relative Gibbs free-energy* (or *relative entropy* according to Fellner and Kniely[14]):

$$\mathcal{G}(u|v) = U(u) - U(v) - \mathcal{D}U(v)(u - v); \tag{61}$$

by an explicit calculation it can be show that

$$\mathcal{G}(n|n^\infty) = \int_\Omega \sum_{i=1}^{k} n_i \log\left(\frac{n_i}{n_i^\infty}\right) - (n_i - n_i^\infty) + \frac{1}{2}\epsilon\|\nabla\varphi - \nabla\varphi^\infty\|^2, \tag{62}$$

and then, by an easy calculation, it can be shown that the Dissipation $\mathcal{D}$ defined by (19) is given by

$$\mathfrak{D}(n, \varphi) = -\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{G}(n|n^\infty). \tag{63}$$

In Fellner and Kniely,[14] by starting from (63) and by the means of a repeated use of Csiszár–Kullback–Pinsker type inequalities, provided (58) holds, the two following estimates were obtained:

$$\begin{aligned} &\mathfrak{D}(n, \varphi) \ge C_1 \mathcal{G}(n, \varphi), \\ &\|n - n_\infty\|_{L^1(\Omega)}^2 + \|\varphi - \varphi_\infty\|_{H^1(\Omega)}^2 \le C_2 \mathcal{G}(n_o, \varphi_o) \exp(-C_1 t), \end{aligned} \tag{64}$$

where φ_o is the unique solution of (7) for the initial data n_o.

The most remarkable feature of this result is that both C_1 and C_2 have an explicit dependence on the parameters of (71):

$$\begin{aligned} C_1^{-1} =& \frac{1}{2}\exp(2\Phi^\infty)\max\left\{\frac{T}{m}\exp(2\Phi^\infty), \frac{1}{k_o}\right\} \cdot (1 + \mathcal{L}(\Omega)\exp(2\Phi^\infty)), \\ C_2 =& 3\exp(2\Phi^\infty) + \frac{1}{2}\mathcal{G}(n_o, \varphi_o) + 2(1 + \mathcal{L}(\Omega)); \end{aligned} \tag{65}$$

in (65) $\mathcal{L}(\Omega) > 0$ is the constant in the Poincaré inequality:

$$\|\Psi\|_{L^p(\Omega)}^2 \le \mathcal{L}(\Omega)\|\nabla\Psi\|_{L^p(\Omega)}^2, \; \forall \Psi \in H^1(\Omega), \bar{\Psi} = 0, \tag{66}$$

which in $L^2(\Omega)$ reduces to $\mathcal{L}(\Omega) = \lambda_1^{-1}$ with λ_1 the first eigenvalue of:

$$\begin{aligned} \Delta\Psi &= \lambda\Psi, \text{ in } \Omega, \\ \Psi &= 0, \text{ on } \partial\Omega. \end{aligned} \tag{67}$$

We notice that, for Ω a sphere of radius R it is $\mathcal{L}(\Omega) \leq (2R/\pi)^2$[64]: for $L = 2R$ then we have $\mathcal{L}(\Omega) \leq 0.1$. Such a value is consistent with the value $\mathcal{L}(\Omega) \approx 0.07$ which was obtained into Daví[9] where the λ_1 was calculated as the square root of the reciprocal of the first zero of the Bessel function Jo'.

The expression for the decay time $\tau = C_1^{-1}$ depends, by $(65)_1$, in an explicit manner on the mobility parameter m, the reaction term $r(0)$, the initial data n_o, and the scintillation volume Ω: as far as we know it is the first explicit estimate of scintillator decay time which depends on the (measurable) constitutive parameters of the model, albeit limited to the case $k = 2$. In Daví,[9] we showed how for four different scintillators these results gave a very good estimate for the experimentally measured fast decay time.

4 | THE APPROXIMATED MODELS

4.1 | The adimensionalized boundary value problem

In order to put the boundary value problem (24) in an adimensionalized form, we begin with the choice of characteristic length and time pair $(L\,, T)$ related to the space and time scintillation scales, in order to define the dimensionless coordinates $(\zeta\,, \tau)$:

$$\zeta = \frac{x}{L}, \ \tau = \frac{t}{T}, \tag{68}$$

and the dimensionless excitation carriers density $u = u(\zeta\,, \tau)$ and electric potential $\psi = \psi(\zeta\,, \tau)$:

$$u = nL^3, \ \psi = \varphi\frac{\epsilon}{eL}. \tag{69}$$

If we further set:

$$M^* = \mu^{-1}M, \ r^* = \kappa^{-1}r, \tag{70}$$

where μ is the greatest eigenvalues of M and and κ the greater component of r or, as in Daví,[9] the value of their norm, then equations $(24)_{1,3}$ can be rendered dimensionless

$$\begin{aligned} &\mathrm{div}_\zeta(dD^*[\nabla_\zeta u] + mM^*[ez \otimes \nabla_\zeta\psi]) - kr^*(u) = u_\tau\,, \\ &-\Delta_\zeta\psi = z\cdot u\,, \end{aligned} \tag{71}$$

and dependent on the three adimensional parameters:

$$d = \frac{Tk_B\theta}{eL^2}\mu, \ m = \frac{eT}{\epsilon L^3}\mu, \ k = \kappa T. \tag{72}$$

We remark that the parameter k depends on the incoming energy by the means of $r(n)$, whereas m depends only on constituive or scaling parameters: accordingly, depending on the energy of ionizing radiation we may have different physically meaningful regimes which are described in the following subsections. Further, for

$$\delta = \frac{k_B\theta}{e}\mu, \tag{73}$$

the greatest eigenvalue of D and provided we define the *diffusion length* $L_D = \sqrt{\delta\, T}$, then we have

$$d = \left(\frac{L_D}{L}\right)^2; \tag{74}$$

we notice that an experimental estimate of L_D is given *e.g.* into.[22]

The set of dimensionless parameters $\{d\,,m\,,k\}$ which appears into $(71)_1$ describes three main regimes for the boundary value problem, depending on the crystal constitutive properties and on the initial ionizing energy. In the next subsections we shall show how many of the most used phenomenological models for scintillators, can be encompassed within $(71)_1$ by an appropriate choice of these parameters. For these models, which we define approximated because some terms of $(71)_1$ can be neglected, we shall also briefly describe the results concerning existence and decay time which are available in the literature.

4.2 | The reaction-diffusion approximation

Let $d \approx k = O(1)$, that is,

$$\kappa \approx \frac{\theta k_B}{eL^2}\mu = O(1), \tag{75}$$

and let

$$\frac{m}{k} = \frac{e}{\epsilon L^3}\frac{\mu}{\kappa} = o(\varepsilon),\ \frac{m}{d} = \frac{e^2}{\epsilon L\theta k_B} = o(\varepsilon), \tag{76}$$

then from $(71)_1$ we recover, to within higher order terms, the reaction-diffusion equation

$$\mathrm{div} D[\nabla n] - r(n) = \dot{n}, \tag{77}$$

and $(71)_2$ is uncoupled, the excitation density vector being a data; this phenomenological model, which traces back its ancestry to the analogous models for chemical reactions,[65,66] is used in many papers dealing with experimental identification of scintillator properties, as in previous works[59,67–72] and did not take into account the drift contribution induced by the electric field φ.

Reaction-diffusion equations were studied with a great interest starting from the kinetics of chemical reactions[66] and there are treatises and textbook devoted to various aspects of them like, for example, in literature[73–75] and many others. However, problems like the existence of solutions and their asymptotic decay have attracted a growing number of studies in recent years, mainly because the difficulties posed by the lack of control of the recombination terms as pointed out in details in previous works.[51–53] Most of the recent results dealing with asymptotic decay deals indeed with reaction terms with quadratic growth for chemical reactions, vid. literature,[76–80] and also, for a different point of view, see previous studies.[28,46,81–83] A first result concerning the existence of renormalized solutions was presented into Desvillettes et al,[84] whereas the most general result at the present available is Fellner et al,[85] where however the nonlinear case of diffusion in porous media is treated. See also the references into Fischer et al,[35] where these results were extended to the non-isothermal case.

Here we show how these results can be used in the context of our problem, described by the equation (77). There are, at the best of our knowledge, no results for a general recombination terms: however, if we neglect the cubic Auger effect, we can use the existence results of Desvillettes et al[84] and decay estimates obtained in Fellner and Tang.[80]

We shall give only a brief survey of both these results and how their hypotheses fit within the physics underlying scintillation: we leave out all the technical details contained in the cited works. We remark that a complete existence and decay estimate results for the scintillation models has yet to be done and the task, given the nature of the reactive term, is a far from an easy one. A further result presented in Fellner and Tang[80] is concerned with the existence of renormalized solutions for (77), whose structure is the same as in (52) when we neglect the drift terms.

4.2.1 | Existence of global weak solutions

The main result of Desvillettes et al[84] concerns the existence of *global weak solutions* for a reaction/recombination term which is at most quadratic in n. The relevant hypotheses are that the recombination term is locally Lipschitz continuous and quasi-negative (the hypotheses (H2)-i) and (54) of Section 3: then, provided $D = \mathrm{diag}\{D_1\,,D_2\,,\ \dots\ ,D_k\}$ with $D_j > D_0 > 0$, $j = 1\,,2\,,\ \dots\ k$ and provided there exist a function $\Theta(x\,,t) \in L^2(\Omega \times [0\,,\tau))$, a scalar $\mu \in (0\,,+\infty)$ and a function $K(\cdot)$ defined as in (56) such that:

- $K(\| r \|) \leq C(1 + \| r \|)$,
- $\forall n \in (1\,,+\infty)^k$ and a.e. $(x\,,t)$:

$$-\sum_{j=1}^{k} \log(n_j) r_j(n) \leq \Theta + \mu \sum_{j=1}^{k} n_j, \tag{78}$$

then (77) admits a global weak solution for any non-negative initial data such that:

$$\|n_o\| \log(\|n_o\|) \in L^2(\Omega). \tag{79}$$

4.2.2 | Asymptotic decay estimates

The result obtained into Fellner and Tang[80] is tailored on chemical reactions, which implies mass conservation and detailed or complex balance of reactive terms, conditions which have no correspondence in the physics of scintillation. In this paper, by using arguments and tools which are unsurprisingly related to those used into Fellner and Kniely,[14] it is shown that, for n^∞, a detailed balanced equilibrium solution corresponding to $r(n^\infty) = 0$, then:

$$\|n - n^\infty\|^2_{L^1(\Omega)} \leq C_1 \mathcal{G}(n_o|n^\infty) \exp(-C_2 t), \tag{80}$$

where the relative entropy $\mathcal{G}$ is defined as (cf. (62)):

$$\mathcal{G}(n_o|n^\infty) = \int_\Omega \sum_{i=1}^{k} n_{oi} \log\left(\frac{n_{oi}}{n_i^\infty}\right) - (n_{oi} - n_i^\infty), \tag{81}$$

$C_1 = C_{CKP}^{-1}$ is the constant in a Csiszár–Kullback–Pinsker type inequality, whereas the constant C_2, whose inverse is the estimate for the decay time, is given by

$$C_2 = \frac{1}{2} \min\left\{\lambda_1, \frac{K_2 H_1(t)}{K_1}\right\}; \tag{82}$$

in this relation $\lambda_1 = C_{LSI} \min\{D_i\}$, with $D = \text{diag}\{D_1, D_2, \dots D_k\}$ and C_{LSI} is the constant in the logarithmic Sobolev inequality, whereas the two constants $K_{1,2}$ depends explicitly on Ω, D, the set k_h, a^h, b^h, n^∞ and on the constant K such that

$$\mathcal{G}(n|n^\infty) \leq K, \tag{83}$$

all the details being given in full in Feller and Kniely.[80] Finally, the term $H_1(t)$ in (82) is given by

$$\sum_{j=1}^{s}\left[\left(\sqrt{\frac{n}{n^\infty}}\right)^{a^h} - \left(\sqrt{\frac{n}{n^\infty}}\right)^{b^h}\right]^2 \geq H_1(t) \sum_{i=1}^{k}\left(\sqrt{\frac{n_i}{n_i^\infty}} - 1\right)^2. \tag{84}$$

4.3 | The diffusion-drift approximation

Let $d \approx m = O(1)$, that is

$$\frac{k_B\theta}{e^2} \approx \frac{1}{\epsilon L}, \tag{85}$$

and

$$\frac{k}{d} = \frac{eL^2}{k_B\theta}\frac{\kappa}{\mu} = o(\varepsilon), \quad \frac{k}{m} = \frac{\epsilon L^3}{e}\frac{\kappa}{\mu} = o(\varepsilon), \tag{86}$$

then we recover, to within higher order terms, the Poisson-Nernst-Planck system, used to model ion fluxes, cell biology and other electrically driven evolution phenomena:

$$\begin{aligned} \text{div}(D[\nabla n] + MN[ez \otimes \nabla\varphi]) &= \dot{n}, \\ &\text{in } \Omega \times [0,\tau), \\ -\epsilon\Delta\varphi &= ez \cdot n, \end{aligned} \tag{87}$$

with Neumann boundary conditions. This model was used into Li et al[17,40] to describe the initial stage of the scintillation, before the recombination takes place. The system (87) is well-studied, and there are many results, dealing with existence,

asymptotic decay, equilibrium solutions and even explicit analytical solutions for $k = 2$ (e.g., among the many[86–89] and for more recent advances[90–93]).

Equation $(87)_1$ can be put in the equivalent gradient flow formulation (21) with $\mathbb{K} = \mathbb{K}_D$. Its stationary solutions $(n^\infty, \varphi^\infty)$ are characterized by s^∞ =const., and therefore, (46) is replaced by

$$n_j^\infty(x) = c_j \exp\left(\frac{s^\infty - ez_j\varphi^\infty(x)}{\theta k_B}\right), \ j = 1, 2, \dots, k, \tag{88}$$

and the Poisson-Boltzmann equation (47) changes accordingly.

4.4 | The reaction-drift approximation

In this case $k \approx m = O(1)$, and

$$\frac{d}{k} = \frac{\theta k_B}{eL^2}\frac{\mu}{\kappa} = o(\varepsilon), \ \frac{d}{m} = \frac{\epsilon L \theta k_B}{e^2} = o(\varepsilon). \tag{89}$$

The boundary value problem (71) reduces in this case to

$$\operatorname{div}(M[N(n)z \otimes \nabla\varphi]) - r(n) = \dot{n}, \ \text{in } \Omega \times [0, \tau), \tag{90}$$

and (7). At the best of our knowledge this equation was never used for scintillators and indeed it can be found in the theory of dopant diffusion in semiconductors (vid., e.g., Höfler et al.[94]): however, we list it together with those used for scintillators for the sake of completeness. For the boundary value problem (90) and (7) global existence, uniqueness and asymptotic decay results were obtained into Glitzky and Hünlich[95] (vid. also Glitzky and Hünlich[96] and Glitzky[97]).

4.5 | The kinetic approximation

Let ε a small parameter, then for $k = O(1)$ and

$$\frac{d}{k} = \frac{\theta k_B}{eL^2}\frac{\mu}{\kappa} = o(\varepsilon), \ \frac{m}{k} = \frac{e}{\epsilon L^3}\frac{\mu}{\kappa} = o(\varepsilon), \tag{91}$$

then $(71)_2$ reduces, to within higher order terms in ε, to the rate ODE equation:

$$-r(n) = \dot{n}, \ \text{in } [0, \tau), \tag{92}$$

with initial condition $n(0) = n_o$; we remark that by (92) and (38), then

$$C_z = z \cdot n, \ C_z \in \mathbb{R}, \tag{93}$$

is a first integral of (92). Also in this case $(71)_2$ is uncoupled, the solution of (92) becoming a charge density supply.

With such approximation, which describes phenomena mainly driven by recombination, we recover the so-called *Kinetic model*, the oldest and most used phenomenological model for scintillators,[20,22,98–103] which is borrowed from the kinetic of chemical reactions; see, e.g., previous works.[104,105]

In a series of recent papers,[32–36] dealing with the gradient structure of (24) it is shown that also (92) admits a gradient structure with $\mathbb{K} = \mathbb{K}_R$ for detailed balanced recombination mechanisms and hence, the energy dissipation methods can be used with success to get asymptotic decay estimates.

In detail, into Maas and Mielke,[36] the well-posedness of (92) was obtained, in the sense that for all initial data $n_o \in \mathcal{M}$ there exists an unique global solution $n : [0, \tau) \to \mathcal{M}$. Moreover it was proved that since the recombination term satisfies the detailed balance conditions for the equilibrium state $n^\infty = c$, then (92) admits the gradient structure:

$$\dot{n} = -H\mathcal{D}U(n), \tag{94}$$

with H given by (28).

Finally, into Mielke and Mittnenzweig,[33] an estimate of the kind of (64) was provided:

$$\mathcal{D}^* \geq C_R \mathcal{G}(n|n^\infty), \tag{95}$$

where the simplified dissipation $\mathcal{D}^*$, such that $\mathcal{D} \geq k_o \mathcal{D}^*$ where k_o is given by, e.g., (60), is defined as

$$\mathcal{D}^* = \sum_{h=1}^{s} \frac{k_h}{k_o} \ell \left(\frac{n^{a^h}}{c^{a^h}}, \frac{n^{a^h}}{c^{b^h}} \right). \tag{96}$$

We can however assume that the boundary value problem (24) in the kinetic approximation admits non-homogeneous solutions: in such a case $(24)_1$ is replaced by

$$-r(n(x,t)) = \dot{n}(x,t), \text{ in } \Omega \times [0,\tau), \tag{97}$$

and it becomes a straightforward exercise to follow, for $k = 2$, the developments of Fellner and Kniely[14] in order to recover the estimates (64) with

$$\begin{aligned} C_1^{-1} =& \frac{1}{2k_o} \exp(2\Phi^\infty) \cdot (1 + \mathcal{L}(\Omega) \exp(2\Phi^\infty)), \\ C_2 =& 3 \exp(2\Phi^\infty) + \frac{1}{2} \mathcal{G}(n_o, \varphi_o) + 2(1 + \mathcal{L}(\Omega)); \end{aligned} \tag{98}$$

in place of (65), the former being the decay time estimate.

4.6 | The diffusive approximation

For $d = O(1)$ (which, by (74) means that $L \approx L_D$), and

$$\frac{m}{d} = \frac{e^2}{\epsilon L \theta k_B} = o(\varepsilon), \ \frac{k}{d} = \frac{eL^2}{\theta k_B} \frac{\kappa}{\mu} = o(\varepsilon), \tag{99}$$

then from $(71)_1$, we obtain a classical anisotropic parabolic equation:

$$\mathrm{div} D[\nabla n] = \dot{n}. \tag{100}$$

In this case, which describes the phenomena when the diffusion within the track is the driving mechanism of scintillation, we recover the *Diffusive model*, which describes the diffusion of excitation carrier within the track, vid. previous study,[19] and is rarely used alone as in, e.g., literature.[37] Once again, the equation of electrostatic $(71)_2$ is uncoupled. We notice that the ratio k/d is known as the *Thiele modulus* in the kinetics of chemical reactions.

A sharp estimate of the decay time for (100) can be easily found by following[56#]: first of all, we notice that at the equilibrium $\dot{n} = 0$ and, by the Neumann boundary conditions, $\nabla n = 0$, hence $n^\infty = \bar{n}^\infty = \text{const}$ and by the charge conservation (10):

$$z \cdot \bar{n}(t) = z \cdot \bar{n}_o = z \cdot n^\infty = 0. \tag{101}$$

Further, by multiplying both sides of (100) by $n(x,t) - n^\infty$, integrating by parts with the Neumann boundary conditions and by the Poincaré inequality, then we get

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_\Omega |n(x,t) - n^\infty|^2 = -2 \int_\Omega D[\nabla n] \cdot \nabla n \leq -\frac{2\delta}{\mathcal{L}(\Omega)} \int_\Omega |n(x,t) - n^\infty|^2. \tag{102}$$

#We notice that in Haskovec et al,[32] this problem was instead formulated in terms of gradient structure with the relative entropy as driving functional, as it was done for the general reaction-diffusion-drift systems.

The first and last terms of (102) yield a first order equation which can be integrated to obtain at an expression alike (64):

$$\|n(x,t)-n^{\infty}\|^2_{L^1(\Omega)} \leq \mathcal{G}(n_o,n^{\infty})\exp\left(-\frac{2\delta}{\mathcal{L}(\Omega)}t\right), \tag{103}$$

with

$$\mathcal{G}(n_o,n^{\infty}) = \int_{\Omega} |n_o(x)-n^{\infty}|^2. \tag{104}$$

Accordingly the decay time τ_d can be estimated as

$$\tau_d \leq \frac{\mathcal{L}(\Omega)}{2\delta} = \frac{e\mathcal{L}(\Omega)}{2k_B\theta\mu}. \tag{105}$$

Whenever $D = \mathrm{diag}\{D_1, D_2, \dots, D_k\}$, equation (100) reduces to the k independent classical diffusion equations:

$$D_j\Delta n_j = \dot{n}_j, \text{ in } \Omega\times[0,\tau),\ D_j>0, j=1,2,\dots,k, \tag{106}$$

with Neumann boundary conditions, whose a complete mathematical treatment can be found into many books, like, e.g., Di Benedetto.[106]

5 | CONCLUSIONS

For the reaction-diffusion-drift equation which describes the evolution and recombination processes of charge carriers in scintillators, we gave an overview of the existence and asymptotic decay estimate which are known to date, at the best of our knowledge. Despite the fact that the topics is a well-studied one, as the non-exhaustive list of references shows, there are still many unanswered questions which deserves further investigations: here, we shall give a concise list of some them. Such a list is of course far from be exhaustive and its items are those which seem more interesting for the physicists.

First of all, the mathematical treatment of the RDD system is based on a precise choice of the entropic term $F(n)$, namely, the Gibbs entropy: however, the physics of scintillation processes suggests that such a choice can be appropriate into describing the behavior over a limited range of energy. The choice of Fermi–Dirac potential in place of the Gibbs entropy will be not only more general but also more related to the true physical nature of the phenomena. Clearly, this leads to a different formulation for the recombination term which requires a different mathematical treatment.

A second point of interest is the decay time estimate obtained for the RDD system: here, there are two major directions for further investigations. The first one is to extend the results obtained into Fellner and Kniely[14] for a system of two charge carriers, $k = 2$ to systems with general k as most of the scintillator phenomenological studies require. The second and more intriguing aspect is the following: the decay time estimate which follows from the result of Fellner and Kniely[14] gives as an upper bound of the decay time the maximum between two values, one which depends on the carrier mobility and the other which depends on the recombination time. When into Daví,[9] we applied these results to four scintillating crystals, not only we obtained a very good estimate of the *fast* decay time: we also notice that the *minimum* between these two values looked like a *lower bound* for the slow decay time. Clearly, the results of Fellner and Kniely[14] tells nothing about this but it would be tempting to prove such an assertion. In such a case, we should have two bounds, an upper one on the fast decay time and a lower one on the slow decay time, a result that would be appreciable in terms of material science.

A third remark is that the *Entropic methods* used to get these estimate should be applied also to the approximated phenomenological models, in particular to the *kinetic* one: we need to remark that a very useful aspects of the results in Fellner and Kniely[14] is the explicit dependence of the estimates on the constitutive parameters of the RDD systems. This should allow for a predictive use of these results, not only for an *a-posteriori* check with the available experimental data.

As far as the questions related to the existence issues for very general form of recombination term is concerned, the necessity of further generalization does not need to be justified, since they are the foundation stone of any numerical procedure we need to implement in order to obtain numerical solutions.

Finally, there are two side-aspects related to those we looked at in this paper and which we left out. The first one concerns the solution of the Bethe–Bloch equation, which is a necessary requirement to bridge the microscopic world with the phenomenological treatment we want to give to scintillation. To date there are many results which concern mainly the

radiation decay length or the Bragg peak: a treatment which conveys in a straightforward way the relevant parameters of the phenomena at the mesoscopic scale would be welcomed.

The second and in some sense more important problem we left out is the *Ligth yield*: such a parameter is indeed a measure of scintillator effectiveness and tells us about the minimum energy we can detect. To date, there is not a coherent mathematical definition for this parameter, apart a descriptive one:

$$LY = \frac{\text{Number of charge carriers recombined into photons}}{\text{Total number of charge carriers generated}},$$

which in turns can be applied either locally or globally, whatever this means. Once again, a formal definition and its consequent mathematical treatment are, in our opinion, still missing, and it would benefit from the huge amount of experimental works which give a precise evaluation of Light Yield.

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CONFLICT OF INTEREST

This work does not have any conflicts of interest.

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