HIDDEN DYNAMICS AND THE ORIGIN OF PULSATING WAVES IN SELF-PROPAGATING HIGH TEMPERATURE SYNTHESIS

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ABSTRACT. The present paper contains the announcement and heuristics of results to be published elsewhere.

We derive the precise limit of SHS in the high activation energy scaling suggested by B.J. Matkowkey-G.I. Sivashinsky in 1978 and by A. Bayliss-B.J. Matkowkey-A.P. Aldushin in 2002. In the time-increasing case the limit coincides with the Stefan problem for supercooled water with spatially inhomogeneous coefficients. In general it is a nonlinear forward-backward parabolic equation with discontinuous hysteresis term.

In the first part of our paper we give a complete characterization of the limit problem in the case of one space dimension.

In the second part we construct in any finite dimension a rather large family of pulsating waves for the limit problem.

In the third part, we prove that for constant coefficients the limit problem in any finite dimension does not admit non-trivial pulsating waves.

The combination of all three parts strongly suggests a relation between the pulsating waves constructed in the present paper and the numerically observed pulsating waves for finite activation energy in dimension $n \geq 1$ and therefore provides a possible and surprising explanation for the phenomena observed.

All techniques in the present paper belong to the category far-from-equilibrium-analysis/far-from-bifurcation-point-analysis.

1. INTRODUCTION

The present paper contains the announcement and heuristics of results to be published elsewhere. In particular the present paper does not contain any proofs.

The system
\[
\begin{align*}
\partial_{t}u - \Delta u &= vf(u), \\
\partial_{t}v &= -vf(u),
\end{align*}
\]
where $u$ is the normalized temperature, $v$ is the normalized concentration of the reactant and the non-negative nonlinearity $f$ describes the reaction kinetics, is a simple but widely used model for solid combustion (i.e. the case of the Lewis number being $+\infty$). In particular it is being used to model the industrial process of Self-propagating High temperature Synthesis (SHS). In the case of high activation

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energy interesting phenomena like instability of planar waves, fingering and screw-like spin combustion waves are observed.

In [15] B.J. Matkowsky-G.I. Sivashinsky derived from a special case of (1) a formal singular limit containing a jump condition for the temperature on the interface. Later it has been argued that the problem is for high activation energy related to a Stefan problem describing the freezing of supercooled water (see [15], [8, p. 57]). Subsequently the Stefan problem for supercooled water became the basis for numerous papers focusing on stability analysis of (1), fingering, screw-like spin combustion waves etc. (see for example [2]).

Surprisingly there are few mathematical results on the subject: In [14] E. Logak-V. Loubeau proved existence of a planar wave in one space dimension and gave a rigorous proof for convergence as the activation energy goes to infinity.

Instability of the planar wave for a special linearization (and high activation energy) is due to [4].

The present paper consists of three parts: in the first part we prove rigorously that in the case of one space dimension the SHS system converges to the irreversible Stefan problem for supercooled water (cf. Theorem 5.1). In the time-increasing case we obtain also convergence in higher dimensions (see Theorem 4.1). As the initial data of the reactant concentration enter the equation as the activation energy goes to infinity, our result also seems to provide a possible explanation for the numerically observed pulsating waves (cf. [9], [17] and [2]).

As a matter of fact, in the second part of our paper (Theorem 7.1) we use the spatially inhomogeneous coefficients in order to construct a pulsating wave for each periodic function $v^0$ (or $Y^0$, respectively) on $\mathbb{R}^n$, using the approach in [3]. We also obtain the spin combustion waves (or "helical waves") on the cylinder mantle (see Remark 7.2).

In contrast, we show in the third part (see Theorem 8.1) that for constant $v^0$ in any finite dimension, no non-trivial pulsating waves exist. In addition, formal stability analysis suggests that in one space dimension the planar wave is stable.

Taken together (cf. section 9), our results strongly suggest a relation between the pulsating waves constructed in the present paper and the numerically observed pulsating waves for finite activation energy in dimension $n \geq 1$ (cf. [9], [17] and [2]) and therefore provide a possible and surprising explanation for the phenomena observed.

In the original setting by B.J. Matkowsky-G.I. Sivashinsky [15, equation (2)], according to our result Theorem 5.1,

\begin{equation}
\begin{aligned}
\partial_t u_N - \Delta u_N &= (1 - \sigma_N) Ne^N u_N \exp(-N/u_N), \\
\partial_t v_N &= -Ne^N u_N \exp(-N/u_N),
\end{aligned}
\end{equation}

each limit $u_\infty$ of $u_N > 0$ as $N \to \infty$ satisfies for $(\sigma_N)_{N \in \mathbb{N}} \subseteq [0, 1)$ (for $\sigma_N \uparrow 1, N \uparrow \infty$ the limit in this scaling is the solution of the heat equation; cf. Section 6.1 and Theorem 5.1)

\begin{equation}
\partial_t u_\infty - v^0 \partial_t \chi = \Delta u_\infty \text{ in } (0, +\infty) \times \Omega,
\end{equation}
where \( u^0 \) are the initial data of \( u_\infty \) and
\[
\chi(t, x) \begin{cases} 
0, & \text{esssup } (0,t)\theta_\infty(\cdot, x) < 1, \\
\in [0,1], & \text{esssup } (0,t)\theta_\infty(\cdot, x) = 1, \\
1, & \text{esssup } (0,t)\theta_\infty(\cdot, x) > 1.
\end{cases}
\]

In the SHS system with another scaling and a temperature threshold (see [2, p. 109-110]),
\[
\begin{align*}
\partial_t \theta_N - \Delta \theta_N &= (1 - \sigma_N)NY_N \exp((N(1 - \sigma_N)(\theta_N - 1))/\sigma_N + (1 - \sigma_N)\theta_N))X_{\{\theta_N > \bar{\theta}\}}, \\
\partial_t Y_N &= -(1 - \sigma_N)NY_N \exp((N(1 - \sigma_N)(\theta_N - 1))/\sigma_N + (1 - \sigma_N)\theta_N))X_{\{\theta_N > \bar{\theta}\}}
\end{align*}
\]
where \( N(1 - \sigma_N) >> 1, \sigma_N \in (0, 1) \) and \( \bar{\theta} \in (0, 1) \), each limit \( \theta_\infty \) of \( \theta_N \) satisfies (cf. Section 6.2 and Theorem 5.1)
\[
\partial_t \theta_\infty - Y^0 \partial_t \chi = \Delta \theta_\infty \text{ in } (0, +\infty) \times \Omega,
\]
where \( Y^0 \) are the initial data of \( Y_\infty \) and
\[
\begin{align*}
\chi(t, x) \begin{cases} 
0, & \text{esssup } (0,t)\theta_\infty(\cdot, x) < 1, \\
\in [0,1], & \text{esssup } (0,t)\theta_\infty(\cdot, x) = 1, \\
1, & \text{esssup } (0,t)\theta_\infty(\cdot, x) > 1.
\end{cases}
\end{align*}
\]

To our knowledge this precise form of the limit problem, i.e. the equation with the discontinuous hysteresis term, has not been known. Even in the time-increasing case it does not coincide with the formal result in [15].

In the case that \( \theta_\infty \) (or \( u_\infty \), respectively) is increasing in time and \( u^0 \) (or \( Y^0 \), respectively) is constant, our limit problem coincides with the Stefan problem for supercooled water, an extensively studied ill-posed problem (for a survey see [5]). As it is a forward-backward parabolic equation it is not clear whether one should expect uniqueness (see [6, Remark 7.2] for an example of non-uniqueness in a related problem).

On the positive side, much more is known about the Stefan problem for supercooled water than the SHS system, e.g. existence of a finger ([10]), instability of the finger ([13]), one-phase solutions ([6]) etc.; those results, when combined with our convergence result, suggest that similar properties should be true for the SHS system.

It is interesting to observe that even in the time-increasing case our singular limit selects certain solutions of the Stefan problem for supercooled water. For example, \( u(t) = (\kappa - 1)\chi_{\{t<1\}} + \kappa\chi_{\{t>1\}} \) is for each \( \kappa \in (0, 1) \) a perfectly valid solution of the Stefan problem for supercooled water, but, as easily verified, it cannot be obtained from the ODE
\[
\partial_t u_\varepsilon(t) = -\partial_s \exp(-\frac{1}{\varepsilon} \int_{0}^{t} \exp((1 - 1/(u_\varepsilon(s) + 1))/\varepsilon) \, ds) \text{ as } \varepsilon \to 0.
\]

Our approach does not involve stability or bifurcation analysis. For showing the convergence we use standard compactness and topological arguments. We prove that if the measure of the "burnt zone" is small enough in a parabolic cylinder, then in a cylinder of smaller radius there cannot be any burnt part. For the construction of pulsating waves we use the approach in [3] as well as blow-up arguments, Harnack inequality and so on. In order to obtain non-existence of pulsating waves for constant \( v^0 \) we use a Liouville technique.
Let us conclude the introduction with a comparison to blow-up in semilinear heat equations, as the main problem arising in our convergence proof, i.e. excluding "peaking of the solution" or burnt zones with very small measure, resembles the blow-up phenomena in semilinear heat equations. One could therefore hope to apply methods used to exclude blow-up in low dimensions in order to exclude peaking, say in two dimensions. There are however problems: First, here, we are dealing not with a single solution but with the one-parameter family $u_\varepsilon$ concentrating at some "peak" as $\varepsilon$ gets smaller. Second, the $\varepsilon$-problem is not a scalar equation but a degenerate system. Third, in contrast to blow-up, peaking would not necessarily imply $u_\varepsilon$ going to $+\infty$. Fourth, our limit problem is a two-phase problem while most known results for blow-up in semilinear heat equations assume the solution to be non-negative. Fifth, in our problem it does not make much sense studying the onset of burning, say the first time when $u_\varepsilon \geq -\varepsilon$, whereas studying the time of first blow-up can be very reasonable for semilinear heat equations. For the same reason blow-up in our case is always incomplete and there is always the non-trivial blow-up profile of the traveling wave. The last and most important difference is that while semilinear heat equations are parabolic and therefore well-posed in a sense, our limit problem contains a backward component making it ill-posed.

2. Notation

Throughout this article $\mathbb{R}^n$ will be equipped with the Euclidean inner product $x \cdot y$ and the induced norm $|x|$. $B_r(x)$ will denote the open $n$-dimensional ball of center $x$, radius $r$ and volume $r^n \omega_n$. When the center is not specified, it is assumed to be 0.

When considering a set $A$, $\chi_A$ shall stand for the characteristic function of $A$, while $\nu$ shall typically denote the outward normal to a given boundary. We will use the distance $d_{\epsilon}$ with respect to the parabolic metric $d((t,x),(s,y)) = \sqrt{|t-s| + |x-y|^2}$.

The operator $\partial_t$ will mean the partial derivative of a function in the time direction, $\Delta$ the Laplacian in the space variables and $\mathcal{L}^n$ the $n$-dimensional Lebesgue measure. Finally $W^{2,1}_p$ denotes the parabolic Sobolev space as defined in [12].

3. Preliminaries

In what follows, $\Omega$ is a bounded $C^1$-domain in $\mathbb{R}^n$ and

$$u_\varepsilon \in \bigcap_{T \in (0, +\infty)} W^{2,1}_2((0,T) \times \Omega)$$

is a strong solution of the equation

$$\partial_t u_\varepsilon(t,x) - \Delta u_\varepsilon(t,x) = -u_\varepsilon^2(x) \partial_t \exp(-\frac{1}{\varepsilon} \int_0^t g_\varepsilon(u_\varepsilon(s,x)) \, ds), \quad u_\varepsilon(0, \cdot) = u_\varepsilon^0 \text{ in } \Omega, \nabla u_\varepsilon \cdot \nu = 0 \text{ on } (0, +\infty) \times \partial \Omega;$$

where $g_\varepsilon$ is a non-negative function on $\mathbb{R}$ satisfying:

0) $g_\varepsilon$ is for each $\varepsilon \in (0,1)$ piecewise continuous with only one possible jump at $z_0$, $g_\varepsilon(z_0-) = g_\varepsilon(z_0) = 0$ in case of a jump, and $g_\varepsilon$ satisfies for each $\varepsilon \in (0,1)$ and for every $x \in \mathbb{R}$ the bound $g_\varepsilon(x) \leq C_\varepsilon (1 + |x|)$.

1) $g_\varepsilon / \varepsilon \to 0$ as $\varepsilon \to 0$ on each compact subset of $(-\infty, 0)$.

2) for each compact subset $K$ of $(0, +\infty)$ there is $c_K > 0$ such that $\min(g_\varepsilon, c_K) \to c_K$ uniformly on $K$ as $\varepsilon \to 0$. 


The initial data satisfy $0 \leq u_{0}^{\epsilon} \leq C < +\infty$, $u_{0}^{\epsilon}$ converges in $L^{1}(\Omega)$ to $u^{0}$ as $\epsilon \to 0$, $(u_{0}^{\epsilon})_{\epsilon \in (0,1)}$ is bounded in $L^{2}(\Omega)$, it is uniformly bounded from below by a constant $u_{\min}$, and it converges in $L^{1}(\Omega)$ to $u^{0}$ as $\epsilon \to 0$.

**Remark 3.1.** Assumption 0) guarantees existence of a global strong solution for each $\epsilon \in (0,1)$.

4. **The High Activation Energy Limit**

The following theorem has been proved in [16]. Let us repeat the statements and its proof for the sake of completeness.

**Theorem 4.1.** The family $(u_{\epsilon})_{\epsilon \in (0,1)}$ is for each $T \in (0, +\infty)$ precompact in $L^{1}((0,T) \times \Omega)$, and each limit $u$ of $(u_{\epsilon})_{\epsilon \in (0,1)}$ as a sequence $\epsilon_{m} \to 0$, satisfies in the sense of distributions the initial-boundary value problem

\[
\partial_{t}u - v^{0}\partial_{t}\chi = \Delta u \text{ in } (0, +\infty) \times \Omega,
\]

\[
u(0, \cdot) = u^{0} + v^{0}H(u^{0}) \text{ in } \Omega, \quad \nabla u \cdot \nu = 0 \text{ on } (0, +\infty) \times \partial\Omega,
\]

where $\chi(t, x) \begin{array}{ll}
\in [0,1], \quad \text{esssup}_{(0,t)} u(\cdot,x) \leq 0, \\
= 1, \quad \text{esssup}_{(0,t)} u(\cdot,x) > 0,
\end{array}

and $H$ is the maximal monotone graph

\[
H(z) \begin{array}{ll}
= 0, & z < 0, \\
\in [0,1], & z = 0, \\
= 1, & z > 0.
\end{array}
\]

Moreover, $\chi$ is increasing in time and $u$ is a supercaloric function.

If $(u_{\epsilon})_{\epsilon \in (0,1)}$ satisfies $\partial_{t}u_{\epsilon} \geq 0$ in $(0,T) \times \Omega$, then $u$ is a solution of the Stefan problem for supercooled water, i.e.

\[
\partial_{t}u - v^{0}\partial_{t}H(u) = \Delta u \text{ in } (0, +\infty) \times \Omega.
\]

**Remark 4.2.** Note that assumption 1) is only needed to prove the second statement "If ....".

**Remark 4.3.** We also obtain a rigorous convergence result in the case of (higher dimensional) traveling waves with suitable conditions at infinity. In this case our $L^{2}(W^{1,2})$-estimate (Step 2) implies a no-concentration property of the time-derivative.

5. **Complete Characterization of the Limit Equation in the Case of One Space Dimension**

**Theorem 5.1.** Suppose in addition to the assumptions at the beginning of Section 4 that the space dimension $n = 1$ and that the initial data $u_{0}^{\epsilon}$ converge in $C^{1}$ to a function $u^{0}$ satisfying $\nabla u^{0} \neq 0$ on $\{u^{0} = 0\}$. Then the family $(u_{\epsilon})_{\epsilon \in (0,1)}$ is for each $T \in (0, +\infty)$ precompact in $L^{1}((0,T) \times \Omega)$, and each limit $u$ of $(u_{\epsilon})_{\epsilon \in (0,1)}$ as a sequence $\epsilon_{m} \to 0$, satisfies in the sense of distributions the initial-boundary value problem

\[
\partial_{t}u - v^{0}\partial_{t}\chi = \Delta u \text{ in } (0, +\infty) \times \Omega,
\]

\[
u(0, \cdot) = u^{0} + v^{0}H(u^{0}) \text{ in } \Omega, \quad \nabla u \cdot \nu = 0 \text{ on } (0, +\infty) \times \partial\Omega,
\]
where $H$ is the maximal monotone graph

$$
H(z) \begin{cases} 
= 0, & z < 0, \\
\in [0, 1], & z = 0, \\
= 1, & z > 0 
\end{cases}
$$

and $\chi(t, x) = H(\text{ess sup}_{(0,t)} u(\cdot, x))$ \begin{cases} 
= 0, & \text{ess sup}_{(0,t)} u(\cdot, x) < 0, \\
\in [0, 1], & \text{ess sup}_{(0,t)} u(\cdot, x) = 0, \\
= 1, & \text{ess sup}_{(0,t)} u(\cdot, x) > 0,
\end{cases}

6. APPLICATIONS

Although the limit equation is an ill-posed problem, the convergence to the limit seems to be robust with respect to perturbations of the $\epsilon$-system and the scaling: here we mention two examples of different systems leading to the same limit. Other examples can be found in mathematical biology (see [11] and [19]).

For the convergence results below we assume that the space dimension is 1.

6.1. The Matkowsky-Sivashinsky scaling. We apply our result to the scaling in [15, equation (2)], i.e.

$$
\begin{align*}
\partial_t u_N - \Delta u_N &= (1 - \sigma_N) N v_N \exp(N(1 - 1/u_N)), \\
\partial_t v_N &= -N v_N \exp(N(1 - 1/u_N)),
\end{align*}
$$

where the normalized temperature $u_N$ and the normalized concentration $v_N$ are non-negative, $(\sigma_N)_{N \in \mathbb{N}} \subseteq [0, 1)$ (in the case $\sigma_N \uparrow 1, N \uparrow \infty$ the limit equation in the scaling is as would be the heat equation, but we could still apply our result to $u_N/(1 - \sigma_N)$ and the activation energy $N \to \infty$.

Setting $u_{\min} := -1, \epsilon := 1/N, u_{\epsilon} := u_N - 1$ and

$$
ge_{\epsilon}(z) := \begin{cases} 
\exp((1 - 1/(z + 1))/\epsilon), & z > -1 \\
0, & z \leq -1
\end{cases}
$$

and integrating the equation for $v_N$ in time, we see that the assumptions of Theorem 5.1 are satisfied and we obtain that each limit $u_{\infty}, \sigma_{\infty}$ of $u_N, \sigma_N$ satisfies

$$
\begin{align*}
\partial_t u_{\infty} - (1 - \sigma_{\infty})v_0 \partial_t H(\text{ess sup}_{(0,t)} u_{\infty}) &= \Delta u_{\infty} \text{ in } (0, +\infty) \times \Omega, \\
u_{\infty}(0, \cdot) &= u^0 + v^0 H(u^0) \text{ in } \Omega, \\
\nabla u_{\infty} \cdot \nu &= 0 \text{ on } (0, +\infty) \times \partial \Omega,
\end{align*}
$$

where $v_0$ are the initial data of $v_{\infty}$. Moreover, $\chi$ is increasing in time and $u_{\infty}$ is a supercaloric function.

6.2. SHS in another scaling with temperature threshold. Here we consider (cf. [2, p. 109-110]), i.e.

$$
\begin{align*}
\partial_t \theta_N - \Delta \theta_N &= (1 - \sigma_N)NY_N \exp((N(1 - \sigma_N)(\theta_N - 1))/\sigma_N + (1 - \sigma_N)\theta_N)\chi_{(\theta_N > \tilde{\theta})}, \\
\partial_t Y_N &= -(1 - \sigma_N)NY_N \exp((N(1 - \sigma_N)(\theta_N - 1))/\sigma_N + (1 - \sigma_N)\theta_N)\chi_{(\theta_N > \tilde{\theta})}
\end{align*}
$$

where $N(1 - \sigma_N) >> 1, \sigma_N \in (0, 1)$ and the constant $\tilde{\theta} \in (0, 1)$ is a threshold parameter at which the reaction sets in.

Setting $u_{\min} = -1, \epsilon := 1/(N(1 - \sigma_N)), \kappa(\epsilon) := 1 - \sigma_N, u_{\epsilon} := \theta_N - 1,

$$
ge_{\epsilon}(z) := \begin{cases} 
\exp((z/(\kappa(\epsilon)z + 1))/\epsilon), & z > \tilde{\theta} - 1 \\
0, & z \leq \tilde{\theta} - 1
\end{cases}
$$
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and integrating the equation for $Y_N$ in time, we see that the assumptions of Theorem 5.1 are satisfied and we obtain that each limit $u_\infty$ of $u_N$ satisfies

\begin{equation}
\partial_t u_\infty - v^0 \partial_t H(\text{ess sup}_{(0,t)} u_\infty) = \Delta u_\infty \quad \text{in} \quad (0, +\infty) \times \Omega,
\end{equation}

where $v^0$ are the initial data of $v_\infty$. Moreover, $\chi$ is increasing in time and $u_\infty$ is a supercaloric function.

7. Existence of Pulsating Waves

The aim of this section is to construct pulsating waves for the limit problem. For the sake of clarity we have chosen not to present the most general result in the following theorem. Moreover we confine ourselves to the one-phase case.

**Theorem 7.1. (Existence of pulsating waves)**

Let us consider a Hölder continuous function $v^0$ defined on $\mathbb{R}^n$ that satisfies

$v^0(x) \geq 1$ and $v^0(x + k) = v^0(x)$ for every $k \in \mathbb{Z}^n$, $x \in \mathbb{R}^n$.

Given a unit vector $e \in \mathbb{R}^n$ and a velocity $c > 0$, there exists a solution $u(t, x)$ of the one-phase problem

\begin{equation}
\begin{cases}
\partial_t u - v^0 \partial_t \chi_{\{u \geq 0\}} = \Delta u & \text{on} \ R \times \mathbb{R}^n \\
\partial_t u \geq 0 & \text{and} \ -\mu_0 := -\int_{[0,1)^n} v^0 \leq u \leq 0,
\end{cases}
\end{equation}

which satisfies

\begin{equation}
\begin{cases}
u(t, x + k) = u(t - \frac{e \cdot k}{c}, x) & \text{for every} \ k \in \mathbb{Z}^n, \ (t, x) \in R \times \mathbb{R}^n \\
u(t, x) = 0 & \text{for} \ x \cdot e - ct \leq 0 \ \text{and} \ \limsup_{x \cdot e - ct \to +\infty} u(t, x) = -\mu_0,
\end{cases}
\end{equation}

where the last limit is uniform as $x \cdot e - ct$ tends to $+\infty$.

**Remark 7.2.** By modifications of the following proofs and of the theory in [3], it is possible to replace $\mathbb{R}^n$ in Theorem 7.1 by a smooth source manifold. Taking for example $S^1 \times \mathbb{R}$, we obtain the screw-like pulsating waves observed in spin combustion (also called "helical waves"; see for example [9], [17], [2],[9]).

Let us transform the problem by the so-called Duvaut transform (see [18]), setting $w(t, x) = -\int_t^{t+\infty} u(s, x) \, ds$. In this section we will prove the existence of a pulsating wave $w$. More precisely, Theorem 7.1 is a corollary of of the following result which will be proved later.

**Theorem 7.3. (Pulsating waves for the obstacle problem)**

Under the assumptions of Theorem 7.1, there exists a function $w(t, x)$ solving the obstacle problem

\begin{equation}
\begin{cases}
\partial_t w = \Delta w - v^0 \chi_{\{w > 0\}} & \text{on} \ R \times \mathbb{R}^n, \\
w \geq 0, \ -\mu_0 \leq \partial_t w \leq 0, \ \partial_{tt} w \geq 0,
\end{cases}
\end{equation}

7
with the conditions
\begin{equation}
\begin{cases}
w(t, x + k) = w(t - \frac{e \cdot k}{c}, x) & \text{for every } k \in \mathbb{Z}^n, \quad (t, x) \in \mathbb{R} \times \mathbb{R}^n, \\
w(t, x) = 0 & \text{for } x \cdot e - ct \leq 0 \text{ and } \partial_t w(t, x) \to -\mu_0 \text{ as } x \cdot e - ct \to +\infty.
\end{cases}
\end{equation}

The convergence is uniform as $x \cdot e - ct$ tends to $+\infty$.

Sketch of Proof of Theorem 7.3
We will prove the existence of an unbounded solution $w$ in six steps, approximating $w$ by bounded solutions of a truncated equation, for which we can apply the existence of pulsating fronts due to Berestycki and Hamel [3].

Step 1: Approximation by bounded solutions and estimates of the velocity
In this case it is possible to apply Theorem 1.13 of Berestycki, Hamel [3], which states the existence (and uniqueness up to translation in time) of bounded pulsating solutions traveling at a unique velocity.

Step 2: Space-time transformation and first estimates on time derivatives
It turns out to be a good idea to "turn" space-time so that in the new coordinates, the interface is contained in a bounded time-strip.

Step 3: Uniform bound of the solution from above
Step 4: Passing to the limit
Step 5: Non-degeneracy property of the solution and bound from below
Step 6: Estimates on the time derivative of the limit solution

8. NON-EXISTENCE OF PULSATING WAVES IN THE CASE OF CONSTANT INITIAL CONCENTRATION

We consider solutions $u$ of the one-phase limit problem with constant initial concentration in any finite dimension, i.e.
\begin{equation}
\partial_t u - \partial_t \chi_{\{u \geq 0\}} = \Delta u \quad \text{in } \mathbb{R} \times \mathbb{R}^n,
\end{equation}
and prove that $u$ cannot be a non-trivial pulsating wave in the sense of (13), (14). More precisely:

**Theorem 8.1. (Non-existence of pulsating waves for constant initial concentration)**

Let $u$ be a solution of (13), (14) in dimension $n \geq 1$ with $v^0 = \text{constant}$. Then $u(t, x) = u(t - e \cdot x/c, 0)$, i.e. $u$ is a planar wave.

The proof is based on a Liouville argument.

9. CONCLUSIONS

In this section we try to take a conclusion of the previous sections.
Let us consider a sequence of solutions $(u_\varepsilon, v_\varepsilon)$ of the $\varepsilon$-problem (6) satisfying for example the assumptions in the time-increasing case of Theorem 4.1, and suppose
that they are getting closer and closer to one-phase pulsating waves as \( \varepsilon \to 0 \), i.e. for some \( t_{\varepsilon} \to +\infty \),

\[
\begin{align*}
  u_{\varepsilon}(t, x + k) &= o(1) + u_{\varepsilon}(t - \frac{e \cdot k}{c}, x) \quad \text{for every } k \in \mathbb{Z}^n, \quad (t, x) \in (t_{\varepsilon}, +\infty) \times \mathbb{R}^n \\
  u_{\varepsilon}(t, x) &= o(1) \quad \text{for } x \cdot e - ct \leq -t_{\varepsilon} \quad \text{and } \lim_{\varepsilon \to 0} \limsup_{x \cdot e - ct + t_{\varepsilon} \to +\infty} u_{\varepsilon}(t, x) = -\mu_0.
\end{align*}
\]

Translating \( t_{\varepsilon} \) to 0, we obtain by Theorem 4.1 a sequence \((u_{\varepsilon}(t_{\varepsilon} + t, x), u_{\varepsilon}(t_{\varepsilon} + t, x))\) that is locally compact in \( L^1 \) and \( u_{\varepsilon} \) converges to \( u \) satisfying all assumptions in Theorem 7.1 except the assumptions for \( v^0(x) \). Let us assume \( v^0 \geq c > 0 \), i.e. there has been enough fuel everywhere initially, and let us show that \( v^0 \) is \( \mathbb{Z}^n \)-periodic: for the integrated function \( w \) as in section 7 and \( k \in \mathbb{Z}^n \),

\[
0 = (\partial_t - \Delta)(w(t, x + k) - w(t - \frac{e \cdot k}{c}, x))
\]

\[
= v^0(x)\chi_{\{w(t - \frac{e \cdot k}{c}, x) > 0\}} - v^0(x + k)\chi_{\{w(t, x + k) > 0\}} = (v^0(x) - v^0(x + k))\chi_{\{w(t, x + k) > 0\}}
\]

and therefore, choosing \(-t \) large, \( v^0(x) - v^0(x + k) = 0 \).

But then Theorem 8.1 tells us that \( u \) must either be one of the pulsating waves constructed in section 7, or a planar wave.

So our results when combined, strongly suggest a relation of the numerically observed pulsating waves for dimensions \( n \geq 1 \) and the pulsating waves constructed in section 7. Of course there remain possible alternatives, for examples our results do not exclude the non-existence of pulsating waves for small \( \varepsilon \), or the strong convergence of pulsating waves to planar waves from far away, or the existence of non-trivial true two-phase pulsating waves in the case of constant \( v^0 \). We leave these possibilities open to mathematical discussion. However in communication with mathematicians doing numerics for pulsating waves of the very systems mentioned in the introduction the above alternatives have been considered unlikely.

Let us also mention that the chaos reported in [1] for finite activation energy presents no contradiction to our results: we never claimed that the dynamics around the pulsating waves is simple.

10. Open Questions

The most pressing task is of course to study for space dimension \( n \geq 2 \) the existence or non-existence of "peaking" of the solution in the negative phase. A related question is whether \((u_{\varepsilon})_{\varepsilon \in (0, 1)}\) is bounded in \( L^\infty \) in the case of uniformly bounded initial data. Although this seems obvious, it is not clear how to prevent concentration close to the interface.

Another challenge is to use the information on the limit problem gained in the present paper to construct pulsating waves for the \( \varepsilon \)-problem.

Uniqueness for the limit problem (the irreversible Stefan problem for supercooled water) in general seems unlikely. One might however ask whether time-global uniqueness holds in the case that \( u \) is strictly increasing in the \( x_1 \)-direction. By the result in [7] for the ill-posed Hele-Shaw problem, time-local uniqueness is likely to be true here, too.

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