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SOME REMARKS ABOUT THE WELL-POSEDNESS OF LIFSHITZ–SLYOZOV’S EQUATIONS WITH NUCLEATION KINETICS

JUAN CALVO*, ERWAN HINGANT†, AND ROMAIN YVINEC‡,§

ABSTRACT. The Lifshitz–Slyozov model is a nonlocal transport equation that can describe certain types of phase transitions in terms of the temporal evolution of a mixture of monomers and aggregates. Most applications of this model so far do not require boundary conditions. However, there is a recent interest in situations where a boundary condition might be needed -e.g. in the context of protein polymerization phenomena. Actually the boundary condition may change dynamically in time, depending on an activation threshold for the monomer concentration. This new setting poses a number of mathematical difficulties for which the existing literature is scarce. In this contribution we construct examples of solutions for which the boundary condition becomes activated (resp. deactivated) dynamically in time; we also discuss how to approach the well-posedness problem for such situations.

1. INTRODUCTION

The Lifshitz–Slyozov model describes the temporal evolution of a mixture of monomers and aggregates. This is done in terms of the monomer concentration $u(t)$ and the number density for aggregates, $f(t, x)$, where x is a size variable. The model reads

$$\frac{\partial f(t, x)}{\partial t} + \frac{\partial}{\partial x} [(a(x)u(t) - b(x))f(t, x)] = 0, \quad t > 0, \quad x \in (0, \infty), \quad (1.1)$$

$$u(t) + \int_0^\infty x f(t, x) dx = \rho, \quad t > 0 \quad (1.2)$$

for some given $\rho > 0$, subject to the initial condition

$$f(0, x) = f^{in}(x), \quad x \in (0, \infty). \quad (1.3)$$

Here the transport term in (1.1) describes the changes in size aggregates according to the following interactions: (i) a monomer attaches to a pre-existing aggregate of size x , with a rate $a(x)$, (ii) a monomer detaches from a pre-existing aggregate of size x , with a rate $b(x)$. We interpret $\rho > 0$ as the total mass of the system, hence (1.2) encodes mass conservation during evolution.

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The former model was initially introduced to describe phase transitions in supersaturated solid solutions [12]. In this context we think of x as a volume variable and we take $a(x) \simeq x^{1/3}$, $b(x) \simeq 1$. More general kinetic rates can be considered and in this way we get a general framework that can be applied to describe various problems in the science of materials (concerning e.g. crystal precipitation or metal alloys, see for instance [9, 17]). For those descriptions x usually accounts for aggregate volume and the kinetic rates a, b are such that small aggregates tend to diminish in size. Actually, the standard dynamics in those scenarios resembles the so-called ‘‘Ostwald ripening’’ phenomena, where the growth of large aggregates is promoted thanks to the monomers gained from the shrinkage of small aggregates.

On mathematical grounds, those works analyzing Ostwald ripening phenomena in the literature assume from the start kinetic rates such that the transport term in (1.1) always points outwards at size $x = 0$ [4, 11, 13, 14]. Therefore no boundary condition is needed to render the problem (1.1)–(1.3) well-posed. However, this need not be always the case. We discuss below -Proposition 2.6- a family of kinetic rates such that the transport term becomes characteristic at $x = 0$ during the evolution. This shows that in general we must be prepared for the possibility of having to prescribe a (dynamic) boundary condition in order to have a well-posed problem. The interesting point here is to relate the eventual boundary condition with relevant mechanisms of the process we are trying to describe.

One specific context where these ideas sit in naturally is that of protein polymerization phenomena, where we think of aggregates as linear chains of length x . To trigger the linear growth of a certain aggregate by monomer addition at both ends, a minimal stable structure is needed first to build upon it. We can represent the formation of such seeds (zero-size aggregates) via a nucleation step involving a few monomers, e.g. [15, 10, 16]. Therefore, in this context the following boundary condition seems reasonable:

$$(a(0)u(t) - b(0))f(t, 0)\chi_{\{a(0)u(t)-b(0)\geq 0\}} = \mathbf{n}(u(t))\chi_{\{a(0)u(t)-b(0)\geq 0\}}. \quad (1.4)$$

Here we think of \mathbf{n} as a mass action kinetics, i.e. $\mathbf{n}(u) \simeq u^n$; however, more general choices for \mathbf{n} can be considered, which opens the room for additional applications of this description apart from those to protein polymerization.

To date, the literature on (1.1)–(1.3) with an inflow boundary condition like (1.4) is scarce. Some instances of this boundary condition have been deduced in [6, 8]. The well-posedness for (1.1)–(1.4) has been treated in [2], where it is shown that only local-in-time solutions can be expected in the general case. This again is due to the fact that the transport field may change its character at $x = 0$ in a dynamic fashion, in such a way that a boundary condition may be needed at earlier times but not at later times.

Therefore, a global analysis of (1.1)–(1.4) seems necessary but is still lacking. The aim of this contribution is to give some partial results about its global well-posedness. Some of these results were announced without proof in [2].

2. KINETIC RATES AND LOCAL-IN-TIME SOLUTIONS

It is handy to discuss the properties of (1.1)–(1.3) in terms of the ratio between polymerization and depolymerization rates, which we now introduce.

Definition 2.1. Assume that $a(x) > 0$ for a.e. $x \in \mathbb{R}^+$. We introduce

$$\Phi : \mathbb{R}^+ \rightarrow \mathbb{R}^+, \quad \Phi(x) := \frac{b(x)}{a(x)}.$$

Let $\Phi^{sup} := \sup_{x \geq 0} \Phi(x)$ and $\Phi_{inf} := \inf_{x \geq 0} \Phi(x)$. We also define

$$\Phi_0 := \lim_{x \rightarrow 0} \Phi(x) \geq 0.$$

We have that $0 \leq \Phi_{inf} \leq \Phi_0 \leq \Phi^{sup}$; note that Φ_0, Φ^{sup} need not be finite.

Well-posedness for (1.1)–(1.3) can be expected to be true if the boundary condition is always “on” (by which we mean $a(0)u(t) - b(0) > 0$ for every time) or always “off”. However, temporal dynamics for which $u(t)$ switches between these two regimes imply a change in the properties of the system. On modeling grounds, the physical meaning of this need not be clear a priori, depending on the specific features of the case under study. Besides, this switching may induce purely mathematical effects whose impact on the well-posedness of the model is not clean cut at first sight. Our investigation here tries to shed some light on this problem. In that regard, the quantity Φ_0 will constitute an important threshold as it will separate two different dynamic regimes.

To state our results we should make clear what do we mean by solutions to (1.1)–(1.3). We need to consider several notions of (weak) solutions. Those are readapted from [2].

Definition 2.2 (Inflow and outflow solutions). Let $T \in (0, \infty]$. We say that a function f is an *inflow solution with nucleation kinetics* (resp. *outflow solution*) to the Lifshitz–Slyozov equation (1.1)–(1.2) on $[0, T]$ with mass $\rho > 0$, kinetic rates $\{a, b, \mathbf{n}\}$ (resp. $\{a, b\}$) and initial value $f^{in} \in L^1(0, \infty)^+$ if the following statements are satisfied:

- (1) The function f belongs to $\mathcal{C}([0, T], w-L^1((0, \infty), (1+x) dx))$, is nonnegative and for each $T^* < T$, it also belongs to $L^\infty((0, T^*), L^1((0, \infty), (1+x) dx))$.
- (2) For every $t \in [0, T)$,

$$u(t) := \rho - \int_0^\infty x f(t, x) dx > \Phi_0. \quad (2.1)$$

(Resp. with $< \Phi_0$.)

- (3) For all $\varphi \in \mathcal{C}_c^1([0, T] \times [0, \infty))$, there holds that

$$\begin{aligned} \int_0^T \int_0^\infty (\partial_t \varphi(t, x) + (a(x)u(t) - b(x))\partial_x \varphi(t, x)) f(t, x) dx dt \\ + \int_0^T \varphi(t, 0) \mathbf{n}(u(t)) dt + \int_0^\infty \varphi(0, x) f^{in}(x) dx = 0. \end{aligned} \quad (2.2)$$

(Resp. for all $\varphi \in \mathcal{C}_c^1([0, T] \times (0, \infty))$, in which case the term in \mathbf{n} is not present.)

One of our aims is to generalize these notions to the case in which an admissible solution can be constructed by merging outflow solutions with inflow solutions. We introduce the following concept.

Definition 2.3 (Matching solutions). We say that a pair (u, f) is a matching solution of (1.1)–(1.3) in $[0, t^*)$ with nucleation boundary condition (1.4) and initial data (u^{in}, f^{in}) if the following conditions hold:

- (1) The set $\mathcal{S} := \{t \in [0, t^*)/u(t) = \Phi_0\}$ is denumerable. Let $(0, t^*) \setminus \mathcal{S} = \cup_{n \in \mathbb{N}} (t_n, t_{n+1})$.
- (2) If $u(t) > \Phi_0$ in (t_n, t_{n+1}) then f is an inflow solution of (1.1)–(1.4) in $[t_n, t_{n+1})$ with initial data $(f(t_n^-), u(t_n^-))$ -that is, we take f^{in} when $n = 0$, otherwise we take $f(t_n^-)$ as the limit when $t \nearrow t_n$ in the weak topology of $L^1((0, \infty), (1+x) dx)$.
- (3) If $u(t) < \Phi_0$ in (t_n, t_{n+1}) then f is an outflow solution of (1.1)–(1.3) in $[t_n, t_{n+1})$ with initial data $(f(t_n^-), u(t_n^-))$.
- (4) $u(t)$ is continuous at each point of \mathcal{S} and $f(t)$ is also continuous at each point of \mathcal{S} , with respect to the weak topology of $L^1((0, \infty), (1+x) dx)$.

Next we state our hypotheses on the initial datum and the kinetic rates. We will not be aiming at full generality in this contribution. Therefore, we shall adopt the following *running assumptions*:

- (1) $f^{in} \in L^1((0, \infty), (1+x)dx)^+$ and $u^{in} := \rho - \int_0^\infty x f^{in}(x) dx \in (0, \rho]$,
- (2) The function $\Phi(x) := a(x)/b(x)$ is defined for a.e. $x > 0$ and has a limit $\Phi_0 \in [0, \infty]$ as $x \rightarrow 0^+$.
- (3) Attachment and detachment rates are globally Lipschitz and positive for $x > 0$. As a consequence, there holds that

$$a(x) \leq A(1+x) \quad \text{for some } A > 0. \quad (2.3)$$

- (4) $\mathbf{n} \in L^\infty([\Phi_0, \rho])^+$.

A number of additional assumptions will be required at several points of the document in order to construct specific examples:

$$\text{There exist } x_L, C > 0 \text{ such that } a(x)(\rho - \Phi(x)) \leq C \text{ for } x \geq x_L, \quad (2.4)$$

$$a(x) \geq \underline{a} > 0 \quad \text{for every } x \geq 0. \quad (2.5)$$

Here (2.4) is a way to state that for large sizes attachment does not overwhelm detachment, whereas (2.5) guarantees that we have a net attachment rate even for small aggregates.

A suitable adaptation of the results and proofs in [2, 4] provides us the following local existence result.

Proposition 2.4. *Under our running assumptions, the following statements hold true:*

- (1) Let $u^{in} > \Phi_0$. Then, there exists some $t^* > 0$ such that there is a unique inflow solution with nucleation kinetics of (1.1)–(1.3) on $[0, t^*)$.
- (2) Let $u^{in} < \Phi_0$ (or $u^{in} = \Phi_0$ with $(u^{in})' < 0$). Then, there exists some $t^* > 0$ such that there is a unique outflow solution of (1.1)–(1.3) on $[0, t^*)$.

Furthermore, the following statements are fulfilled in both cases:

- (1) $u \in C^1([0, t^*))$. Actually, the time derivative is given by the following expression:

$$\frac{du}{dt} = \int_0^\infty b(x)f(t, x) dx - u(t) \int_0^\infty a(x)f(t, x) dx.$$

- (2) Either $t^* = +\infty$ or $t^* < \infty$ and $\lim_{t \nearrow t^*} u(t) = \Phi_0$.
- (3) Provided that $t^* < \infty$, then $u'(t)$ has a limit as $t \nearrow t^*$ and $f(t)$ has a limit as $t \nearrow t^*$ in the weak topology of $L^1((0, \infty), (1+x) dx)$.

To investigate prolongability and matching solutions we shall first rule out situations where inflow solutions will surely not take place. The following provides a partial generalization of similar results in [1, 5]:

Lemma 2.5. *Assume that the initial pair (u^{in}, f^{in}) is such that $\rho < \Phi_{inf}$. Then any outflow solution of the Lifshitz–Slyozov model is global in time. Furthermore, under any of the following conditions the outflow solution verifies that $u(t) \nearrow \rho$ as $t \rightarrow \infty$:*

- (1) $a(x) \geq a_0 x$ for some $a_0 > 0$ and for every $x \in [0, \infty)$.
- (2) f^{in} is compactly supported and (2.5) holds.

Proof. Solutions are global in time as $u(t)$ cannot reach Φ_0 . Let us discuss now the long time behavior. For the first case we use that

$$u' = \int_0^\infty a(x)(\Phi(x) - u(t))f(t, x) dx \geq (\Phi_{inf} - \rho) \int_0^\infty a(x)f(t, x) dx.$$

This provides both the time integrability of the first moment and the monotonicity of u . Thanks to the time integrability we can find a divergent sequence of times along which the first moment converges to zero. Therefore u converges to ρ as $t \rightarrow \infty$ thanks to (1.2).

For the second case we use the representation of outflow solutions in terms of characteristics, see for instance [4]. Under our hypotheses, the time derivative of the characteristic curve with data (t, x) can be estimated from above as

$$\begin{aligned} \frac{d}{ds} X(s; t, x) &= a(X(s; t, x))(u(s) - \Phi(X(s; t, x))) \\ &\leq (\rho - \Phi_{inf})a(X(s; t, x)) < \underline{a}(\rho - \Phi_{inf}) < 0. \end{aligned}$$

Then for a given datum (t, x) the associated characteristic curve leaves $[0, +\infty)$ at some finite instant $t' > t$. Therefore the aggregate distribution f vanishes, actually in finite time. \square

For more general sets of kinetic rates, we cannot presently rule out concentration behavior at zero size when $\rho < \Phi_{inf}$ —think for instance in $a(x) = x^\alpha$ with $\alpha \in (0, 1)$. See [3] for a more detailed investigation of this pathological phenomenon. At any rate, we will impose the condition $\Phi_0 < \rho$ in the sequel, in order to allow for inflow solutions with nucleation kinetics.

Next we state and prove a result concerning local solutions of the Lifshitz–Slyozov model in the outflow case. We provide a family of kinetic rates for which such local solutions cannot be extended to a global outflow solution. Thus, not every initial configuration that starts to evolve as an outflow solution will undergo an Ostwald ripening dynamics right away. Even in those cases in which that turns out to be true, a transient with inflow dynamics might take place before that. A restricted instance of this result was announced in [2].

Proposition 2.6. *Assume that $\liminf_{x \rightarrow \infty} \Phi(x) > \Phi_0 = \Phi_{inf} > 0$ and that Φ_{inf} is only attained at $x = 0$. Assume also that $\Phi'(0) > 0$. If f^{in} is compactly supported, (2.4) and (2.5) hold and $u^{in} < \Phi_0 < \rho$ then every outflow solution of (1.1)–(1.3) is local in time.*

Proof. Thanks to the hypotheses we can find a concave, strictly increasing profile Φ_c such that $\Phi(x) \geq \Phi_c(x)$ for every $x \in [0, \infty)$ and $\Phi_c(0) = \Phi_0$. Then, notice that

$$u' \geq \int_0^\infty a(x)(\Phi_c(x) - u(t))f(t, x) dx. \quad (2.6)$$

Since Φ_c is concave we have

$$\Phi_c(x) \geq \Phi(0) + \frac{\Phi_c(z) - \Phi(0)}{z}x, \quad x \in [0, z] \quad (2.7)$$

-recall that $\Phi(0) = \Phi^{inf} = \Phi_0$. Now we shall choose z taking into account that we can control the spatial support of the solution. Namely, if $\text{supp } f^{in} \subset [0, \bar{f}^{in})$ then thanks to (2.4) there exists $C > 0$ such that $\text{supp } f(t) \subset [0, \bar{f}^{in} + tC]$ for every $t \geq 0$ in the life span of the solution. That enables us to use (2.7) with $z(t) = \bar{f}^{in} + tC$. It follows that

$$u' \geq (\Phi_0 - u(t))\underline{a} \int_0^{z(t)} f(t, x) dx + \underline{a} \frac{\Phi_c(z(t)) - \Phi_0}{z(t)}(\rho - u(t)). \quad (2.8)$$

Note that both terms at the right hand side are positive. We drop the first one; to handle the second we argue by contradiction and assume that $u(t) < \Phi_0$ for every time. In that way we get to

$$u(t) \geq u^{in} + (\rho - \Phi_0)\underline{a} \int_0^t \frac{\Phi_c(z(\tau)) - \Phi_0}{z(\tau)} d\tau.$$

The former integral is strictly positive, and so is $\rho - \Phi_0$ by assumption. Since $|\Phi_c(z(t)) - \Phi_0| \geq |\Phi_c(z(0)) - \Phi_0|$ the time integral diverges logarithmically. Then $u(t)$ reaches Φ_0 in finite time, say $u(t^*) = \Phi_0$. Moreover, from (2.8) we can also deduce that $\lim_{t \nearrow t^*} u'(t) > 0$. \square

3. PROLONGABILITY AND GLOBAL SOLUTIONS

Our next result gives a sufficient condition to merge inflow solutions with nucleation kinetics and outflow solutions.

Proposition 3.1. *Given $u^{in} = \Phi_0$, assume that $u'(0) < 0$ (resp. $u'(0) > 0$). Then there is some $\delta > 0$ such that (1.1)–(1.3) has an outflow solution (resp. (1.1)–(1.4) has an inflow solution with nucleation kinetics) for $t \in (0, \delta)$, agreeing continuously with the initial data and such that $u(t) < \Phi_0$ (resp. $u(t) > \Phi_0$) for $t \in (0, \delta)$.*

Proof. Let us deal first with the case of negative starting derivative. We consider the auxiliary problem given by (1.3) coupled with

$$\frac{\partial f}{\partial t} + \frac{\partial}{\partial x} [((\Phi_0 - |u(t) - \Phi_0|)a - b)f] = 0, \quad (3.1)$$

in $[0, 1) \times \mathbb{R}^+$, say. Note that for this problem the characteristics at $x = 0$ are never pointing inwards, so there is no need for a boundary condition. Having Lipschitz rates grants uniqueness of the characteristic curves; moreover, it enables us to apply a suitable modification of Proposition 2.4 for the above flow in the specific case $u^{in} = \Phi_0$. This provides us with some $\delta > 0$ –which can be estimated from f, a and b – such that: (i) there is a pair $u \in C^1(0, \delta)$ and $f \in \mathcal{C}([0, T], w - L^1((0, \infty), (1+x)dx))$ which matches continuously the initial data, (ii) the pair solves (3.1) coupled with (1.2) and (1.3), (iii) the given solution satisfies $u(t) < \Phi_0$ on $(0, \delta)$. Then, we notice that on $(0, \delta)$ problems (1.1)–(1.2)–(1.3) and (3.1)–(1.2)–(1.3) are equivalent for this solution, thus proving our first claim.

The proof for the second claim is nearly the same as for the first one. This time we consider the following auxiliary problem:

$$\frac{\partial f}{\partial t} + \frac{\partial}{\partial x} [((\Phi_0 + |u(t) - \Phi_0|)a(x) - b(x))f(t, x)] = 0, \quad (3.2)$$

for which characteristics at zero are never pointing outwards. This is complemented with (1.2) and the following boundary condition replacing (1.4):

$$((\Phi_0 + |u(t) - \Phi_0|)a(0) - b(0))f(t, 0) = \mathbf{n}(u(t)).$$

The remaining steps can be performed as before. \square

There are some particular cases in which we can show *a priori* that $t^* = +\infty$ in Proposition 2.4 based on the information we have about the temporal evolution of the monomer concentration. The existing mathematical results for Ostwald ripening dynamics [4, 11, 13, 14] are based on a set of assumptions on the rates that guarantee that the transport field will point outwards for every positive time. Here we provide another result in the same line but with a different way of formulating the outflow assumptions.

Proposition 3.2. *Assume that $\Phi_0 = \Phi^{sup}$. If $u^{in} < \Phi_0$ then the outflow solution of (1.1)–(1.3) satisfies $t^* = \infty$ and thus it is globally defined.*

Proof. This is clearly true in the case $\Phi_0 = \Phi^{sup} > \rho$, since mass conservation implies that $\lim_{t \nearrow t^*} u(t) = \Phi_0$ will never happen. Therefore, we focus on the case $\Phi_0 = \Phi^{sup} \leq \rho$. Let (f, u) be the solution pair provided by Proposition 2.4. For any $t < t^*$ we have that

$$\frac{d}{dt}(u(t) - \Phi^{sup}) = u'(t) \leq (\Phi^{sup} - u(t)) \int_0^\infty a(x)f(t, x) dx.$$

This implies that

$$\begin{aligned} u(t) - \Phi_0 &\leq (u^{in} - \Phi_0) \exp \left\{ - \int_0^t \int_0^\infty a(x)f(\tau, x) dx d\tau \right\} \\ &\leq (u^{in} - \Phi_0) \exp \left\{ -At \left(\rho + \int_0^\infty f^{in}(x) dx \right) \right\} < 0. \end{aligned}$$

Thus, if we assume $t^* < \infty$ we arrive to the fact that $\lim_{t \nearrow t^*} u(t) < \Phi_0$, which is a contradiction. Therefore we must have $t^* = \infty$. \square

The proof of the former result is very similar to that of the dual statement for inflow solutions with nucleation kinetics given in [2], Theorem 1.9; we recall it here for the reader's convenience:

Proposition 3.3. *Assume that $\Phi_0 = \Phi^{inf}$. If $u^{in} > \Phi_0$ then the inflow solution of (1.1)–(1.3) with nucleation kinetics (1.4) satisfies $t^* = \infty$ and thus it is globally defined.*

3.1. Hints on finite-time crossings. The purpose of this paragraph is to analyze some situations for which local-in time solutions can be extended to global matching solutions. Intuitively speaking, a sufficient condition for prolongability would be as follows: $u(t)$ should reach Φ_0 with non-vanishing derivative. Let us show that this is enough to extend those local solutions constructed in Proposition 2.6, which exhibit a change from outflow to inflow behavior. This was announced in [2] and the proof follows similar ideas to those used to prove Theorem 1.9 in that reference.

Proposition 3.4. *Under the same assumptions of Proposition 2.6, there is a matching solution in the sense of Definition 2.3 which is globally defined and such that \mathcal{S} is a singleton and $\liminf_{t \rightarrow \infty} u(t) \geq \Phi_0$.*

Proof. Proposition 2.4 ensures that there is an outflow solution in some nonempty time interval $[0, t^*)$ in the sense of Definition 2.2, with initial pair (u^{in}, f^{in}) . Actually, $t^* < \infty$. In fact, Proposition 2.6 shows that

$$\lim_{t \nearrow (t^*)^-} u(t) = \Phi_0, \quad \lim_{t \nearrow (t^*)^-} u'(t) < 0.$$

Next we use Proposition 3.1 with the initial pair $(u((t^*)^-), f((t^*)^-))$. This produces an inflow solution with nucleation kinetics in some nonempty interval $[t^*, T)$. Here T can be made arbitrary large after Proposition 3.3. Therefore, the concatenation of the outflow solution in $[0, t^*)$ and the inflow solution with nucleation kinetics in $[t^*, \infty)$ produces a globally defined matching solution. \square

Remark 3.5. The matching solution constructed above is actually unique. This is due to the fact that matching solutions as introduced in Definition 2.3 are actually such that $u(t)$ is a C^1 function of time, check Proposition 2.4. However, this does not suffice to disregard nonuniqueness phenomena in a general scenario, where we have to consider the possibility that $u'(t)$ might vanish as we approach t^* .

For completeness we state a dual result, which covers the case of some solutions whose behavior changes from inflow to outflow. This can be proved using the same techniques as the previous proposition and extends the scope of the result already given in [2].

Corollary 3.6. *Assume that $\limsup_{x \rightarrow \infty} \Phi(x) < \Phi_0 = \Phi^{sup} < \infty$ and that Φ^{sup} is only attained at $x = 0$. Assume that (2.4) is fulfilled. If f^{in} is compactly supported and $u^{in} > \Phi_0$ then there is a matching solution of (1.1)–(1.4) which is globally defined and such that \mathcal{S} is a singleton and $\limsup_{t \rightarrow \infty} u(t) \leq \Phi_0$.*

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