HOW TO HOMOGENIZE A NONLINEAR DIFFUSION EQUATION (E.G.) STEFAN'--ETC(U)
MRC Technical Summary Report #2061

HOW TO HOMOGENIZE A NONLINEAR DIFFUSION EQUATION (e.g. STEFAN'S PROBLEM)

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April 1980

Received December 17, 1979

Approved for public release
Distribution unlimited

Sponsored by
U.S. Army Research Office
P.O. Box 12211
Research Triangle Park
North Carolina 27709

80 7 7 115
ABSTRACT

We study the homogenization of a Stefan problem (i.e., heat conduction with change of phase) when the structure is \( \varepsilon \)-periodic and prove that the constitutive laws of the limit medium do not depend upon the boundary conditions and are those of an anisotropically heat conducting medium which undergoes a change of phase at each temperature of change of phase of the original substances.

AMS(MOS) Subject Classification: 35K55, 65P05, 80.35, 80.49

Key Words: Homogenization, Stefan problem, Heat diffusion, Changes of phase.

Work Unit #1 - Applied Analysis

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Sponsored by the United States Army under Contract Nos. DAAG29-75-C-0024 and DAAG29-80-C-0041.
We study the homogenization of a periodically distributed mixture of two (or more) pure media which can undergo one or more changes of phase. The problem is to find the properties of the idealized homogeneous medium (if it exists) whose behaviour the original mixture approaches when the periodic mesh gets finer and finer.

It is proved under quite general boundary conditions, and under the restriction that for each original medium the heat conductivity is independent of the temperature, that such a limit medium exist and that it has the following properties: its heat diffusion is homogeneous but non-isotropic in general, its specific heat is an average of the specific heats of the original media, and it does undergo change of phase at all the temperatures of change of phase of the original media. This last fact is in strong agreement with everyday experience, as witnessed for example by the freezing of ice cream.
I. Introduction

For many physical studies of composite materials, one is more interested in a global or "macroscopic" behavior of a composite medium rather than in a detailed "microscopic" one. To put it in a different way, and for reasons that can also come from numerical analysis when discretization is considered, one is interested in finding the relevant properties (i.e., the constitutive laws or physical parameters) for an idealized homogeneous medium which would have the limit behavior of the composite material when the size of the periodic mesh goes to zero.

Finding the relevant parameters of this idealized limit (when it exists) is the origin and one of the main contributions of homogenization theory. Since this is not intended to be an introduction, let alone a survey, of homogenization theory, the reader is referred to Bensoussan-Lions-Papanicolaou [1] for a complete set of references.

The model problem we are looking at here is the problem of homogenization of a nonlinear heat equation for a composite material consisting of a periodic mixture of media which can undergo changes of phase, (Stefan's problem). One can also apply the present results to electromagnetic composite materials (see Bossavit-Damlamian [1]).

The plan is as follows

2. The model ϵ-problem; weak formulation.
5. The limit problem and its constitutive laws.

Conclusions

The author is very grateful to Professor J. M. Lasry for raising this interesting question, to Professor C. Tartar and F. Murat for their helpful suggestions.
2. The Modal c-problem.

In short, the problem we are looking at is the following:

Let \( \Omega \) be a given bounded domain (in \( \mathbb{R}^N \) usually \( N = 3 \)) with smooth boundary \( \partial \Omega \).

We restrict ourselves to two media \( M_1 \) and \( M_2 \). Their distribution in \( \Omega \) is given according to a periodic structure of mesh size \( \epsilon \), proportional to a basic period \( Y \) of size 1. The basic period \( Y \) is partitioned into two smooth subsets \( Y_1 \) and \( Y_2 \) corresponding to each medium \( M_1 \) and \( M_2 \), so that correspondingly \( \Omega \) is partitioned into \( \Omega_1, \epsilon \) and \( \Omega_2, \epsilon \). The boundary between \( Y_1 \) and \( Y_2 \) is denoted by \( Z, \epsilon \), and its image in \( \Omega \) is \( \partial \Omega \) which is supposed to be rigid and perfectly heat conducting. As for \( Z, \epsilon \) it is split into \( T_{1, \epsilon} \) and \( T_{2, \epsilon} \) corresponding to each medium.

It is assumed as customary for a Stefan problem that the variations in volume are negligible.

Time will be restricted to an interval \([0,T]\) and it will be shown that the result is independent of \( T \).

In \( Q = [0,T[ \times \Omega \), each change of phase for each medium will, in the strong formulation, generate a free boundary separating the phases. With our convention that there is at most one change of phase for each medium, two boundaries \( S_{1, \epsilon} \) (i = 1, 2) (not necessarily connected) are generated.

Before writing the strong formulation, we introduce some notations: for each medium \( i (=1,2) \), \( \alpha_i, \beta_i, \gamma_i, k_i \), denote the specific heat (a function of the temperature \( v \)), the temperature of change of phase, the latent heat, the heat conductivity (which is assumed to be strictly positive independent of the temperature \( v \), a limitation for our final result but which has not been lifted so far).

We can now write the strong formulation of the problem. One looks for \( v'(t,x) \) (temperature) and the two surfaces \( S_{1, \epsilon} \) satisfying

\[
\begin{cases}
\text{in } (0,T) \times \Omega_{1, \epsilon} \setminus S_{1, \epsilon} \\
\alpha_i(v^c) \frac{\partial v^c}{\partial t} = \text{div}(k_i \nabla v^c)
\end{cases}
\]

where \( f \) is an internal heating term.
on \((0,T)\) \(\mathbb{R}^d\), continuity of \(v^e\)

\[
(2.2) \begin{cases}
\text{continuity of heat flux} \\
(k_1 - v^e \cdot \hat{n} = k_2 - v^e \cdot \hat{n}) \\
\text{on } S_{1,\varepsilon} \nonumber
\end{cases}
\]

\[
(2.3) \begin{cases}
de_{1} \cos(n_1, \xi) - \int \frac{\partial v^e}{\partial x_j} \cos(n_j, \xi) \right|_{S_{1,\varepsilon}} = 0 \\
\text{where } \hat{n} \text{ is the unit normal to } S_{1,\varepsilon} \text{ in space time, } \left| \right|_{S_{1,\varepsilon}} \text{ indicates the jump across } S_{1,\varepsilon} \text{ along } \hat{n}. \text{ This is the classical Stefan condition on the free boundary.}
\end{cases}
\]

Initial condition: \(v^e(0)\) given in \(\Omega\) together with the initial boundaries \(S_{1,\varepsilon}(0)\); they are assumed to be compatible \((v^e(0) = v_{1,\varepsilon})\) on \(S_{1,\varepsilon}(0)\).

As for the lateral boundary conditions, it is known in the linear homogenization theory that provided they are of variational form, they do not interfere with the limiting process. To be complete we shall take them linear inhomogenous of mixed type. We assume a smooth partition of \(\Gamma\) in \(\Gamma^+\) and \(\Gamma^-\) (\(\Gamma^-\) with non empty interior) and require the following:

\[
(2.5) \begin{cases}
v^e(t,x) = g^-(t,x) \text{ on } \Gamma^- \\
\end{cases}
\]

\[
(2.6) \begin{cases}
k_1 \frac{\partial v^e}{\partial n} + P v^e = g^+(t,x) \text{ on } \Gamma^+ \cap \Gamma_{1,\varepsilon} \\
\end{cases}
\]

here \(P\) is a non-negative smooth function measuring the permeability of the boundary \(\Gamma^+\) to heat flow, \(g^-\) and \(g^+\) are given smooth functions. It is also assumed that the boundary data \(g^-, g^+\) agrees with the initial data \(v^e_0\) at \(t = 0\). It turns out that conditions (2.2) and (2.3) are Rankine-Hugoniot type conditions for the energy balance equation taken in the distribution sense on \(\Omega\). In order to write this equation (which gives a weak formulation) we need some notations:

For each \(i\), let \(Y_i\) denote the maximal monotone graph defined (up to a constant) by

\[
(2.7) Y_i(v) = \int_{\mathbb{R}^+} a_i(s) ds + b_i H(v - a_i) 
\]

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(H is the Heaveside function). This represents the enthalpy as a function of the temperature.

We also put:

(2.8) \[ \gamma(y, v) = \gamma_1(v) \quad \text{for } y \in Y \]

(2.9) \[ k(y) = k_1 \quad \text{for } y \in Y. \]

Then (2.1), (2.2), (2.3) reduce to

(2.10) \[ \frac{3u^c}{\partial t} - \text{div}(k_1 \nabla v^c) = f \quad \text{in } D'(\Omega) \]

(2.11) \[ u^c(t, x) = \gamma_1, v^c(t, x) \]

The initial conditions (2.4), can be expressed in terms of \( u \) alone as an initial condition

(2.12) \[ u^c(0) = u_0, \]

which we can assume independent of \( \epsilon \).

For the lateral boundary conditions, we introduce an auxiliary problem, where the time \( t \) is a mere parameter:

Let \( g_\epsilon(t, x) \) be the solution of

\[
\begin{cases}
- \text{div}(k_1 \nabla g_\epsilon(t)) = 0 & \text{in } \Omega \\
g_\epsilon(t) = g_\epsilon^+(t) & \text{on } \Gamma^-
\end{cases}
\]

(2.13) \[ k_1 \left( \frac{3g_\epsilon^-(t)}{\partial n} + p g_\epsilon(t) = g_\epsilon^+(t) \quad \text{on } \Gamma^+. \]

Clearly \( g_\epsilon \) is bounded in \( H^1(\Omega) \) uniformly in \( \epsilon \). Now (2.1)-(2.6) has the following weak formulation

(2.14) \[ \int - \varphi, u_\epsilon + \int_0^T a_\epsilon(v_\epsilon - q_\epsilon, \varphi) = \int \varphi + \int \varphi(0)u_0 \]

for all \( \varphi \in C^1(\Omega), \varphi(T) = 0 \) and \( \varphi = 0 \) on \( (0, T) \times \Gamma^- \)

(2.15) \[ v_\epsilon - q_\epsilon = 0 \quad \text{on } (0, T) \times \Gamma^- \]

In (2.14) \( a_\epsilon \) is the bilinear Dirichlet form given by

(2.16) \[ a_\epsilon(w, \varphi) = \int k_1 \left( \frac{\chi}{\epsilon} \right)(\nabla w, \nabla \varphi) + \int_\Gamma p w \cdot \varphi. \]

The smoothness assumptions made for \( \Gamma^-, \Gamma^+ \) allow for (2.14) to take \( \varphi \) in a larger class namely \( \varphi \in W^{1,2}(0, T; V) \) where \( V \) is the variational space: \( \{ \psi \in H^1(\Omega), \psi|_{\Gamma^-} = 0 \} \).

(see Damlamian [1] for a detailed study).
(2.17) **Definition**: \((u^c,v^c)\) is a weak solution for problem (2.1)-(2.6) if and only if they satisfy (2.11), (2.14), (2.15).

(2.18) **Theorem** (see Damlamian [1] and [2]).

Under the hypothesis that the \(a_i\)'s are bounded above and below away from zero, there exists a unique solution \((u^c,v^c)\) for problems (2.11), (2.14), (2.15) which satisfies:

\[
\begin{align*}
    &u^c \in W^{1,2}(0,T;V^*) \cap L^{\infty}(0,T;L^2(\Omega)) \\
    &v^c - g^c \in W^{1,2}(0,T;L^2(\Omega)) \cap L^{\infty}(0,T;V).
\end{align*}
\]

Instead of giving a detailed proof we will only give the idea of how to obtain uniform estimates in the following paragraph.
3. The uniform estimates.

(3.1) Proposition. The solutions $(u^\varepsilon, v^\varepsilon)$ with $\varepsilon > 0$ satisfy the following:

- $u^\varepsilon$ is bounded in $W^{1,2}(0,T;V^*) \cap L^\infty(0,T;L^2(\Omega))$ uniformly in $\varepsilon > 0$.
- $v^\varepsilon - g^\varepsilon$ is bounded in $W^{1,2}(0,T;L^2(\Omega)) \cap L^\infty(0,T;V)$ uniformly in $\varepsilon > 0$.

Proof: To obtain these estimates, it is enough to show them in the case of smooth $\varepsilon$.

Then $u^\varepsilon$ and $v^\varepsilon$ are smooth enough to replace (2.14) by

$$
\int_{0}^{T} a^\varepsilon(v^\varepsilon - g^\varepsilon, \varphi) = \int_{0}^{T} \varphi(0) u^\varepsilon_0 - \varphi(T) u^\varepsilon(T)
$$

for all $\varphi$ in $W^{1,2}(0,T;V)$.

Then taking $\varphi = [A^{-1}u^\varepsilon]^{-1}(A^\varepsilon$ being the operator associated to $a^\varepsilon$ on $V$) one gets:

$$
\|u^\varepsilon(t)\|_{V^*}^2 + \int_{0}^{t} \|v^\varepsilon u^\varepsilon\|_{V^*}^2 \leq \int_{0}^{t} \|u^\varepsilon_0\|_{V^*} + \int_{0}^{t} \|u^\varepsilon\|_{V^*} + \int_{0}^{t} \|u^\varepsilon\|_{V^*} + \int_{0}^{t} \|u^\varepsilon\|_{V^*}
$$

where $\|.,.\|_{V^*}$ is the dual norm of $(a^\varepsilon(.,.))$ on $V$, the latter being uniformly equivalent to the standard norm on $V$.

Also taking $\varphi = [A^{-1}u^\varepsilon]^{-1} \frac{du^\varepsilon}{dt}$ one gets

$$
\frac{1}{2} \int_{0}^{T} \frac{\|d\varepsilon\|_{V^*}^2}{dt} \leq C_2 \text{ (a constant which depends upon } f, V_0, g^+, g^- \text{ ... )}.
$$

From (3.3) and (3.4) one gets (because $\varepsilon u^\varepsilon$ can be assumed non-negative), that $v^\varepsilon$ is bounded in $L^\infty(0,T;L^2(\Omega))$.

$$
u^\varepsilon \text{ in } W^{1,2}(0,T;V^*) .
$$

Then one takes $e = \frac{d(v^\varepsilon - g^\varepsilon)}{dt}$ to get

$$
\int_{0}^{t} \frac{\|d\varepsilon\|_{V^*}^2}{dt} \leq c_4 \text{ (a constant which depends upon } f, g^+, g^- \text{ ... )} .
$$

From (3.5) one infers that $v^\varepsilon$ stays bounded in $W^{1,2}(0,T;L^2(\Omega))$ and $v^\varepsilon - g^\varepsilon$ stays bounded in $L^\infty(0,T;V)$.

A detailed proof of the above can be found in A. Damlamian [1] and [2], and in much simpler cases in Brezis [4] and Lions [1].
It is worth noticing that given the estimates of (3.1) (even not uniform in \( r \)) equation (2.14) can be replaced by (3.2) or even by

\[
\int_0^T a^\varepsilon(v^\varepsilon - q^\varepsilon, v) = \int_0^T f \text{ } + \int_0^T \varphi(u_0^\varepsilon - u^\varepsilon(t)) \\
\text{for all } v \text{ in } V \text{ (independent of } t \).
\]

This remark (cf. Damlamian [1]) shows that \((v^\varepsilon, q^\varepsilon)\) is the solution of a simpler variational inequality (of the type studied by G. Duvaut [1]).

Another way of looking at (3.6) is the following, since the operator does not depend upon time

\[
\begin{cases}
\alpha^\varepsilon(v^\varepsilon(t), v) = \int_0^T \{ f(t) + u_0^\varepsilon - u^\varepsilon(t) \} v \\
v^\varepsilon(t) = 0 \text{ on } \Gamma
\end{cases}
\]

for all \( t \in (0, T) \), all \( v \) in \( V \), where

\[
v^\varepsilon(t,x) = \int_0^t (v^\varepsilon(s,x) - q^\varepsilon(s,x))ds
\]

\[
f(t,x) = \int_0^t f(s,x)ds.
\]
4. A short review of elliptic homogenization

The purpose of this paragraph is to show how elliptic homogenization works and how it can be applied in the present problem. See Bensoussan-Lions-Papanicolaou [1] (Also L. Tartar [1]). With the same notations as above, we consider the operator

\[ A^\varepsilon = -\text{div}(k \left( \frac{X}{\varepsilon} \right)V) \] on \( \Omega \). Let \( w^\varepsilon \) be the variational solution of

\[
\begin{align*}
A^\varepsilon w^\varepsilon &= f^\varepsilon \quad \text{in} \quad \Omega \\
w^\varepsilon &= g^- \quad \text{on} \quad \Gamma^-\\
k\left( \frac{X}{\varepsilon} \right) \frac{\partial w^\varepsilon}{\partial n} + p w^\varepsilon &= g^+ \quad \text{on} \quad \Gamma^+
\end{align*}
\]

that is

\[ a^\varepsilon(w^\varepsilon, \varphi) = \int_\Omega f^\varepsilon \varphi + \int_{\Gamma^+} g^+ \varphi \]

for all \( \varphi \) in \( V \), \( w^\varepsilon = g^- \) on \( \Gamma^- \).

We assume that \( f^\varepsilon \) converges to \( f^0 \) weakly in \( L^2(\Omega) \).

(4.3) Proposition: As \( \varepsilon \) goes to zero, \( w^\varepsilon \) converges weakly in \( H^1(\Omega) \) to the solution \( w^0 \) of the following problem

\[ a^0(w^0, \varphi) = \int_\Omega f^0 \varphi + \int_{\Gamma^+} g^+ \varphi \]

for all \( \varphi \) in \( V \), \( w^0 = g^- \) on \( \Gamma^- \), where \( a^0 \) is the bilinear form given by

\[ a^0(w, \varphi) = \int_\Omega \sum_{j,k} q_{j,k} \frac{\partial w}{\partial x_j} \frac{\partial \varphi}{\partial x_k} + \int_{\Gamma^+} P w \varphi \]

with constant coefficients \( q_{j,k} \) given by

\[ q_{j,k} = \frac{1}{\text{mes}(y)} \int_y k(y) V (x_j^3, y) \nabla (x_j^3 - y) \]

where \( x_j^3 \) is the solution (defined uniquely up to a constant) of

\[ -\text{div}(k(y) V x_j^3) = -\text{div}(k(y)e_j), \quad x_j^3 \quad \text{periodic in} \quad Y. \]

\( e_j \) is the \( j \)th unit vector in \( \mathbb{R}^N \), \( y_j \) being the coordinate on \( e_j \).

Proof: It is clear that \( w^\varepsilon \) being bounded in \( H^1(\Omega) \) (by coerciveness of \( a^\varepsilon \) with the Dirichlet boundary condition). So we can assume (via uniqueness of the solution for the limit problem-to-be) that \( w^\varepsilon - w^0 \). Then by a result of Tartar [1], one can see that

\[ k\left( \frac{X}{\varepsilon} \right) \frac{\partial w^\varepsilon}{\partial x_j} \text{converges weakly in} \quad L^2_{\text{loc}}(\Omega) \text{ (hence in} \quad L^2(\Omega)) \text{ to} \quad \sum_j q_j \frac{\partial w^0}{\partial x_j}, \quad (q_j \text{ given by} \quad (4.5), \quad (4.6)), \quad \text{so that} \quad (4.2) \quad \text{goes to the limit to} \quad (4.4), \quad \text{which is the weak formulation of} \]

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Here \( \frac{\partial}{\partial y} A^0 \) is the conormal derivative for \( A^0 \), which one should notice is not diagonal, but still symmetric, and with constant coefficients

\[
A^0 = - \sum_{j,k} q_{j,k} \frac{\partial^2}{\partial x_j \partial x_k}.
\]

Here we have also used the compactness of the trace operator from \( H^1(\Omega) \) into \( L^2(\Gamma) \).
5. The limit problem and its constitutive laws.

Making use of the results of paragraph 4, one sees that $g^\epsilon(t)$ converges weakly in $W^1(\Omega)$ to the solution $g^0(t)$ of

\[
\begin{align*}
\begin{cases}
0^0 g^0(t) & = 0 \\
g^0(t) & = g^\epsilon(t) \quad \text{on } \Gamma^- \\
\frac{\partial g^0(t)}{\partial n^0} + p^0 g^0(t) & = q^+ \quad \text{on } \Gamma^+ .
\end{cases}
\end{align*}
\]

(5.1)

Also, by the estimates of (3.1) one can extract a sequence of values of $\epsilon$ going to zero such that

- $u^\epsilon - u^0 \text{ in } W^{1,2}(0,T;V^*) \cap L^\infty(0,T;L^2(\Omega))$
- $v^\epsilon - v^0 - v^0 \text{ in } W^{1,2}(0,T;L^2(\Omega)) \cap L^\infty(0,T;V)$.

Hence $u$ converges strongly in $C([0,T];V^*)$ and for all $t \in [0,T]$, $u^\epsilon(t)$ converges weakly to $u^0(t)$ in $L^2(\Omega)$.

Consequently we can apply the result of (4.3) to (3.7) so that $v^\epsilon(t)$ which obviously converges to $v^0(t) = \int_0^t (v^0(s) - q^0(s))ds$ satisfies:

\[
\begin{align*}
\begin{cases}
a^0(v^0(t),\psi) & = \int_\Omega (\tau(t) + u^0(t) - u^0(t))\psi \\
v^0(t) & = 0 \quad \text{on } \Gamma^- .
\end{cases}
\end{align*}
\]

(5.2)

Using the equivalence with the weak formulation of type (2.14) we get

\[
\begin{align*}
\begin{cases}
v^0 - q^0 & = 0 \quad \text{on } (0,T) \times \Gamma^- \\
\int_0^t u^0 + \int_\Omega a^0(v^0 - q^0,\varphi) & = \int_\Omega f + \int_\Omega u_0\varphi(0) \\
\end{cases}
\end{align*}
\]

for all $\varphi$ in $W^{1,2}(0,T;V)$ $\varphi(T) = \tau_1(0T)_{x_1} = 0$.

We now turn to (2.11), that is

$u^\epsilon(t,x) \in \gamma(x, \epsilon, v^\epsilon(t,x))$.

Assuming $\epsilon$ is chosen so that $g^\epsilon$ converges to $g^0$ weakly in $H^1(\Omega)$, then $v^\epsilon$ converges to $v^0$ weakly in $W^{1,2}(0,T;L^2(\Omega)) \cap L^\infty(0,T;H^1(\Omega))$ so that the convergence is

\[ (*) \]

The limit problem having a unique solution, it will be clear by the end of the proof that the whole sequence converges.
uniform in $C([0,T], L^2(\cdot))$ for example.

Let $c$ be a real number, different from $\gamma_{t'}$, put $w_c^t(x) = \gamma(\frac{x}{c}, c)$. Clearly, $w_c^t$ converges weakly in $L^2$ to a constant

$$
\gamma(c) = \frac{1}{\text{mes}(Y)} \int_Y \gamma(y, c) \, dy
$$

Using the monotonicity of $\gamma(\frac{\cdot}{c}, \cdot)$ we have

$$
\gamma(t, x) = (w_c^t(t, x) - w_c^t(x)) (v^t(t, x) - c) \geq 0 \quad \text{a.e.}
$$

Hence, using the proper convergences, we get that $u(t, x)$ converges weakly in the sense of measures on $\Omega$ for all $t$ to $(u^0 - \gamma(c)) (v^0 - c)$ which has to be non-negative.

Hence $u^0(t, x)$ belongs to the unique maximal extension of the monotone graph $\gamma$, which we denote by $\tilde{\gamma}$. Consequently (2.11) goes to

$$
u^0(t, x), \quad \tilde{\gamma}(v^0(t, x)) \quad \text{a.e. in } x, \text{ for all } t.
$$
6. Conclusion

We conclude that the limit equations correspond to the weak formulation of the following strong problem:

\[
\begin{align*}
\frac{du}{dt} + A^0 v &= f \\
u(t,x) &
\in \tilde{\gamma}(v(t,x)) \\
u(0,x) &= u^0(x) \\
v(t,x) &= g^-(t,x) & x &\in \Gamma^- \\
\frac{\partial v}{\partial v^0}(t,x) + p v(t,x) &= g^+(t,x) & x &\in \Gamma^+ 
\end{align*}
\]

or to look at it from the Stefan problem point of view, a non isotropic Stefan problem.

One can notice that we recover the heat diffusion operator of the linear case, that is a homogeneous but anisotropic heat diffusion.

One also gets an "averaging" phenomenon for the graphs \( \gamma_i \)'s over \( \gamma \), which is the only averaging consistent with the fact that both \( \gamma_i \)'s are defined up to an additive constant and so is \( \tilde{\gamma} \). Both temperatures of change of phases appear for discontinuities of \( \tilde{\gamma} \), which is in agreement with daily experience (any other averaging of \( \gamma_1 \) and \( \gamma_2 \) would have yielded no discontinuity in the average, hence no change of phase). It is easy to see that the specific heat and latent heat of the limit medium are averages over \( \gamma \) of the corresponding terms.

Finally, on the theoretical side of things, it is of interest to realize that the isotropic diffusion laws are not stable under homogenization of Stefan problems, but anisotropic ones are stable.

It remains to prove that the above can be extended to the case of temperature-dependent heat conductivity for each medium, which is already more complicated but solved in the non Stefan case (see Tartar [1]).
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L. Tartar
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We study the homogenization of a Stefan problem (i.e., heat conduction with change of phase) when the structure is \( r \)-periodic and we prove that the constitutive laws of the limit medium do not depend upon the boundary conditions and are those of an anisotropically heat conducting medium which undergoes a change of phase at each temperature of change of phase of the original substances.