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Complete finite-strain isotropic thermo-elasticity

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Abstract

This paper deals with finite strain isotropic thermo-elasticity without any specific Ansatz regarding the Helmholtz free energy. On the theoretical side, an Eulerian setting of isotropic thermo-elasticity is developed, based on the objective left Cauchy-Green tensor along with the Cauchy stress. The construction of the elastic model relies on a particular invariants choice of the strain measure. These invariants are built so that a succession of elementary experiments, in which the invariants evolve independently, ensures the complete identification of the Helmholtz free energy and thus of the thermo-elastic constitutive law. Expressions idealizing these experimental tests are proposed. A wide range of hyperelastic models are found to be a special case of the model proposed herein.

Keywords: Finite strain thermo-elasticity, experimental identification

Introduction

Elasticity is probably one of the most extensively discussed topics in the analytical mechanics literature over the last few centuries. Since Robert Hooke's linear relationship linking the current Cauchy stress $\boldsymbol{\sigma}$ to the linearized strain $\boldsymbol{\varepsilon}$, many authors studied small strain but also non-linear elasticity, the latter being well suited to study materials, such as polymer, which undergo large deformations. The fundamental ground of this phenomenon, which can precede more complex non linear events, is now a very well assessed subject, both theoretically [3, 9, 25] and experimentally.

However, specific Ansatz are still commonly used concerning the Helmholtz free energy from which the elastic constitutive law is derived. Classical small-strain elasticity, linking the second Piola-Kirchhoff tensor $\boldsymbol{\pi}$ to the right Green-Lagrange strain tensor \boldsymbol{E} relies on a quadratic expansion of the Helmholtz free energy with respect to the small strain \boldsymbol{E} . As regards finite strain elasticity, where such series expansion are no longer valid, the construction is based on a choice of the Helmholtz free energy according to invariants of the deformation tensor. Two main categories are often distinguished, depending on the state variables

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chosen. On the one hand, Neo-Hookean[20], Mooney-Rivlin [22], or Yeoh [36] models postulate a polynomial expansion of the free energy with respect to the fundamental invariants of a strain tensor. The coefficients involved in the polynomial expansion are fitted to reproduce experimental data. On the other hand, stretched based models, such as Ogden's which develop the Helmholtz free energy as a power series, retain the positive eigenvalues of the right Cauchy-Green tensor \mathbf{C} . Ogden models have been found to be very efficient for large strain elasticity of rubber like material [6]. Many articles have offered reviews of these different models and their respective pros and cons [7, 10, 19, 23].

The first objective of this paper is to present a choice of a triplet of kinematically significant invariants, allowing for the rigorous identification of the Helmholtz free energy, through a succession of simple elementary experiments and without *a posteriori* adjustment. These invariants are chosen so that they can evolve independently.

In Section 1, we will recall the main lines of construction of the finite strain isotropic thermo-elastic behavior. In Section 2, we will present the invariants of the left Cauchy-Green deformation tensor used for the construction of the elastic model and the resulting constitutive law. Then, we will study, Section 3, the successive elementary experiments which allow the identification of the Helmholtz free energy. Physically realistic expressions for the experimental data required to build the model will then be proposed in Section 4.

1. Reminder of finite strain elasticity

This section briefly presents the fundamental assumption of finite strain isotropic thermo-elasticity within an Eulerian setting. The formulation of the general theory is given in the thermodynamic framework by introducing the Helmholtz free energy as a function of a finite strain measure, and by exploring both the first and the second law of thermodynamics.

1.1. Framework

Let $\mathcal{D}_0^m \in \mathbb{R}^3$ be a material domain corresponding to the reference configuration of a continuous medium. Particles labeled by $\mathbf{x}_0 \in \mathcal{D}_0^m$ are transported in the current configuration to $\mathbf{x}_t \in \mathcal{D}_t^m$ by a one-to-one mapping $\boldsymbol{\varphi}_t$. Let us note \mathbf{F} the deformation gradient of this mapping.

Let us consider two observers (reference frames) \mathcal{R} and $\tilde{\mathcal{R}}$. We denote \mathbf{Q}_0 the orthogonal transformation linking these two reference frames at the initial time and \mathbf{Q}_t the orthogonal tensor linking these same two observers in the current configuration. A second-rank tensor, \mathbf{Y} for the observer \mathcal{R} , equal to $\tilde{\mathbf{Y}}$ for the observer $\tilde{\mathcal{R}}$, is called invariant (or “objective Lagrangian”) [21] if:

$$\tilde{\mathbf{Y}} = \mathbf{Q}_0 \cdot \mathbf{Y} \cdot \mathbf{Q}_0^\top \quad \mathbf{Q}_0 \in SO(3); \forall t. \quad (1)$$

51 Similarly, we will call “objective” any second-rank tensor \mathbf{X} satisfying the following equality:

$$\tilde{\mathbf{X}} = \mathbf{Q}_t \cdot \mathbf{X} \cdot \mathbf{Q}_t^\top \quad \mathbf{Q}_t \in SO(3). \quad (2)$$

52 The invariance or objectivity property of a tensor is intrinsic and comes either from its mathematical
53 definition or from a physically motivated assumption. The first step in the construction of a constitutive
54 law is the choice of a deformation measure, which cannot be simply \mathbf{F} , among two large families:

- 55 • On the one hand, strain tensors based on the product $\mathbf{C} = \mathbf{F}^\top \cdot \mathbf{F}$ or its related tensors of the form
56 $\mathbf{Y} = \frac{1}{n}(\mathbf{U}^n - \mathbf{1})$ with $\mathbf{U} = \sqrt{\mathbf{C}}$ (pure stretch). These deformation tensors are often called “right
57 strain measure”, “Lagrangian” or “invariant” because they obey the relation (1). These strain measures
58 are used to close the mechanical problem by relating the first or second Piola-Kirchoff stress tensor \mathbf{P} ,
59 $\boldsymbol{\pi}$ (which are also postulate invariant) to the displacement field \mathbf{u} .
- 60 • On the other hand, strain measure, called “left”, “Eulerian” or “objective”, and based on the left Cauchy-
61 Green deformation $\mathbf{B} = \mathbf{F} \cdot \mathbf{F}^\top$ (subsequently called “Finger tensor”) are more rarely considered to
62 build elasticity models. These deformation measurements effectively link the objective Cauchy stress
63 $\boldsymbol{\sigma}$ to the displacement field.

64 1.2. Elastic behavior

65 The finite-strain isotropic elastic behavior is based on the following three assertions:

- 66 1. Temperature and a strain measure are the only state variables.
- 67 2. The stress measure is a state function.
- 68 3. In all evolutions, the intrinsic dissipation is null.

69 The first assumption 1) forbids the presence of additional internal variables which could be the witness of
70 phenomena such as plasticity, damage etc... But also anisotropy through a structural tensor [4, 37]. The
71 second assumption 2 excludes viscous effects and a possible strain rate \mathbf{D} (\mathbf{D} denotes the symmetric part of
72 the spatial velocity gradient) dependency of the Cauchy stress. The last assumption 3 ensures reversibility
73 in the sense that the entropy production other than thermal is zero. To satisfy these three requirements,
74 most treatises devote the so-called “Lagrangian” approaches and relate the second Piola-Kirchoff tensor to
75 the right Green-Lagrange deformation tensor. Eulerian formulations of elasticity are rarely considered (see
76 nevertheless [24, 31]), especially since the Cauchy stress cannot be written as the simple derivative of a
77 potential with respect to a strain measure.

1.3. Eulerian finite strain isotropic elasticity

Here, we choose to catch the motion with the left Cauchy-Green objective strain tensor \mathbf{B} which we relate to the Cauchy stress $\boldsymbol{\sigma}$ whose physical meaning is perfectly clear.

The isotropy of the material $\boldsymbol{\sigma}(T, \mathbf{B}) = \mathbf{Q}_t \cdot \boldsymbol{\sigma} \cdot \mathbf{Q}_t^\top (T, \mathbf{Q}_t \cdot \mathbf{B} \cdot \mathbf{Q}_t^\top)$ and the Rivlin-Ericksen theorem [25] implies that the Cauchy stress necessarily expands as a quadratic polynomial of the strain measure:

$$\boldsymbol{\sigma} = K_0^B \mathbf{1} + K_1^B \mathbf{B} + K_2^B \mathbf{B}^2. \quad (3)$$

The coefficients K_i^B are scalar functions of the temperature, but also of a triplet of \mathbf{B} invariants. Their expressions, as a function of the free energy, can be deduced from the nullity of the intrinsic dissipation (see [35] p.295) written on the current (spatial) configuration:

$$\Phi = -\rho \left(\dot{\psi}^m + \dot{T} s^m \right) + \boldsymbol{\sigma} : \mathbf{D} = 0. \quad (4)$$

For an isotropic material, the Helmholtz massic free energy is an isotropic function [4, 32] $\psi^m(T, \mathbf{B}) = \psi^m(T, \mathbf{Q}_t \cdot \mathbf{B} \cdot \mathbf{Q}_t^\top)$, $\forall \mathbf{Q}_t \in SO(3)$ so that there exists f_ψ^B such that:

$$\psi^m(T, \mathbf{B}) = f_\psi^B(T, B_I, B_{II}, B_{III}), \quad (5)$$

where the fundamental invariants (B_I, B_{II}, B_{III}) denote the coefficients of the characteristic polynomial of \mathbf{B} (*main invariant*). Note that any other triplet of invariants bijectively related to (B_I, B_{II}, B_{III}) , is legitimate. Thus, the “principal” invariants defined by $B_i = \frac{1}{i} \text{Tr} \mathbf{B}^i$ are often used to construct elastic laws as their partial derivatives with respect to the strain measure are straightforward. In the following, we will retain invariants whose kinematic meaning is clear.

According to the fundamental invariants (B_I, B_{II}, B_{III}) , the isotropic finite strain thermo-elastic law reads, (see Appendix A but also [16, 31]):

$$\boldsymbol{\sigma} = \frac{2\rho_0}{\sqrt{B_{III}}} \left(B_{III} \partial_{B_{III}} f_\psi^B \mathbf{1} + (\partial_{B_I} f_\psi^B + B_I \partial_{B_{II}} f_\psi^B) \mathbf{B} - \partial_{B_{II}} f_\psi^B \mathbf{B}^2 \right). \quad (6)$$

1.4. Elastic model based on volumetric split

Most materials exhibit completely different volumetric and isochoric responses so that we make use of the isochoric-volumetric commutative split of the gradient \mathbf{F} [13, 30]:

$$\mathbf{F} = \left(J^{\frac{1}{3}} \mathbf{1} \right) \cdot \left(J^{-\frac{1}{3}} \mathbf{F} \right) = \left(J^{\frac{1}{3}} \mathbf{1} \right) \cdot \bar{\mathbf{F}}, \quad (7)$$

$$\Rightarrow \mathbf{B} = \left(J^{\frac{2}{3}} \mathbf{1} \right) \cdot \left(J^{-\frac{2}{3}} \mathbf{B} \right) = \mathbf{B}^{sph} \cdot \bar{\mathbf{B}}. \quad (8)$$

\mathbf{B}^{sph} is a spherical tensor whose only invariant is the volume expansion J :

$$J = \det \mathbf{F} = \sqrt{\det \mathbf{B}} = \sqrt{B_{III}}. \quad (9)$$

98 The isochoric Finger tensor $\overline{\mathbf{B}}$ satisfies $\det \overline{\mathbf{B}} = 1$ and is therefore represented by two invariants. We
 99 note, without any ambiguity, \overline{B}_I and \overline{B}_{II} the two fundamental invariants of the isochoric Finger tensor.
 100 These invariants are trivially related to those of \mathbf{B} by:

$$\overline{B}_I = J^{-\frac{2}{3}} B_I = B_{III}^{-\frac{1}{3}} B_I \text{ and } \overline{B}_{II} = J^{-\frac{4}{3}} B_{II} = B_{III}^{-\frac{2}{3}} B_{II}. \quad (10)$$

101 One can find a function \tilde{f}_ψ^B of the invariants $(T, \overline{B}_I, \overline{B}_{II}, B_{III})$ coinciding with the Helmholtz massic free
 102 energy:

$$\psi^m(T, \mathbf{B}) = f_\psi^B(T, B_I, B_{II}, B_{III}) = \tilde{f}_\psi^B(T, \overline{B}_I, \overline{B}_{II}, B_{III}). \quad (11)$$

103 The partial derivatives of the new function \tilde{f}_ψ^B are then obtained by simple algebraic calculations. The
 104 elastic law (6) is then transformed to give (see Appendix B):

$$\begin{aligned} \boldsymbol{\sigma} &= \frac{2\rho_0}{\sqrt{B_{III}}} \left(B_{III} \partial_{B_{III}} \tilde{f}_\psi^B \mathbf{1} + \partial_{\overline{B}_I} \tilde{f}_\psi^B \text{Dev} \overline{\mathbf{B}} - \partial_{\overline{B}_{II}} \tilde{f}_\psi^B \text{Dev} \left(\overline{\mathbf{B}}^2 - \text{Tr}(\overline{\mathbf{B}}) \overline{\mathbf{B}} \right) \right), \\ \boldsymbol{\sigma} &= \frac{2\rho_0}{\sqrt{B_{III}}} \left(B_{III} \partial_{B_{III}} \tilde{f}_\psi^B \mathbf{1} + \frac{\partial_{\overline{B}_I} \tilde{f}_\psi^B}{B_{III}^{\frac{1}{3}}} \text{Dev} \mathbf{B} - \frac{\partial_{\overline{B}_{II}} \tilde{f}_\psi^B}{B_{III}^{\frac{2}{3}}} \text{Dev} (\mathbf{B}^2 - \text{Tr}(\mathbf{B}) \mathbf{B}) \right). \end{aligned} \quad (12)$$

105 $\text{Dev} \mathbf{X} = \mathbf{X} - (\frac{1}{3} \text{Tr} \mathbf{X}) \mathbf{1}$ refers to the deviatoric (trace-less) part of a second-order tensor.

106 (12) emphasizes the deep link between the *isochoric strain* and the *deviatoric part of the stress*.

107 2. Construction of the thermo-elastic model

108 The invariants used so far are natural, but inefficient for the identification of the Helmholtz free energy,
 109 as they evolve simultaneously in classical experimental tests. The corner-stone of the subsequent section is
 110 the choice of independent invariants associated with simple elementary experiments, allowing them to evolve
 111 one after the other.

112 2.1. State variables

113 *First invariant of the isochoric Finger tensor.* We introduce a first invariant γ_s of the isochoric Finger tensor
 114 $\overline{\mathbf{B}}$, defined by:

$$\gamma_s = \sqrt{\overline{B}_I - 3} = \sqrt{\frac{B_I}{B_{III}^{\frac{1}{3}}} - 3}, \quad (13)$$

115 γ_s is bijectively related to the maximum distortion δ_{\max} (see [15]):

$$\gamma_s = \sqrt{3} \sqrt{(\delta_{\max})^{\frac{2}{3}} - 1} \text{ with } \delta_{\max} = \max_{\mathbf{Q} \in SO(3)} \frac{1}{|\det(\mathbf{Q} \cdot \mathbf{u}_t, \mathbf{Q} \cdot \mathbf{u}'_t, \mathbf{Q} \cdot \mathbf{u}''_t)|}, \quad (14)$$

116 where $(\mathbf{u}_t, \mathbf{u}'_t, \mathbf{u}''_t)$ denotes an initially orthogonal material direction triplet (at $t = 0$, $\det(\mathbf{u}_0, \mathbf{u}'_0, \mathbf{u}''_0) = 1$).

117 The maximum distortion δ_{\max} reflects the maximum decrease of the solid angle formed by three initially

orthogonal material directions. Moreover, in an isochoric simple shear test, defined by the transformation field (see Figure 1):

$$\mathbf{x}_t = \boldsymbol{\varphi}(\mathbf{x}_0, t) = J^{\frac{1}{3}}(\mathbf{x}_0 + \gamma Y \mathbf{e}_x), \quad (15)$$

simple algebra yield $\gamma_s = \gamma$, so that γ_s equalized the motion magnitude.

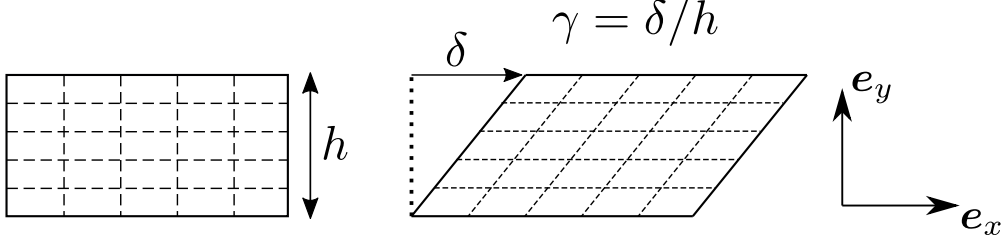


Figure 1: Isochoric planar simple shear kinematics

Second invariant of the isochoric Finger tensor. We propose to construct a second invariant of $\bar{\mathbf{B}}$ which remains null in any isochoric simple shear test. Thus, one may clarify the dependence of the Helmholtz massic free energy with respect to this new invariant. The isochoric Finger tensor associated with the motion (15) reads:

$$\bar{\mathbf{B}} = (1 + \gamma^2)(\mathbf{e}_x \otimes \mathbf{e}_x) + \gamma(\mathbf{e}_x \otimes \mathbf{e}_y + \mathbf{e}_y \otimes \mathbf{e}_x) + \mathbf{e}_y \otimes \mathbf{e}_y + \mathbf{e}_z \otimes \mathbf{e}_z. \quad (16)$$

Set $\gamma_{\perp} = 0$ and $e = 1$ in (D.3) shows that the two fundamentals invariants of (16) stay equal $\bar{B}_{\text{II}} = \bar{B}_{\text{I}}$.

We define the invariant γ_t , identically zero in any spherical evolution and in a simple shear test, by:

$$\gamma_t = \sqrt{\bar{B}_{\text{I}} - 3} - \sqrt{\bar{B}_{\text{II}} - 3} = \gamma_s - \sqrt{\frac{\bar{B}_{\text{II}}}{J^{\frac{4}{3}}} - 3}, \quad (17)$$

γ_t is always well defined (i.e. the terms under the roots are always positive) by concavity of the logarithm. A closed form of its variations' domain is given in Appendix C.

2.2. Constitutive law with the new invariants set

The list of reduced state variables retained is thus $(T, J, \gamma_s, \gamma_t)$ (9), (13), (17). The relations are inversed in:

$$J = B_{\text{III}}^{\frac{1}{2}}; \quad \gamma_s = \sqrt{\bar{B}_{\text{I}} - 3}; \quad \gamma_t = \gamma_s - \sqrt{\bar{B}_{\text{II}} - 3} \Leftrightarrow B_{\text{III}} = J^2; \quad \bar{B}_{\text{I}} = \gamma_s^2 + 3; \quad \bar{B}_{\text{II}} = (\gamma_s - \gamma_t)^2 + 3; \quad (18)$$

The generic elastic law (12), expressed in terms of the principal invariants $(T, \bar{B}_{\text{I}}, \bar{B}_{\text{II}}, B_{\text{III}})$ is transformed by applying the chain rule:

$$\boldsymbol{\sigma} = \rho_0 \left(\partial_J \tilde{f}_{\psi} \mathbf{1} + \frac{1}{\gamma_s J^{\frac{5}{3}}} \left(\partial_{\gamma_s} \tilde{f}_{\psi} + \partial_{\gamma_t} \tilde{f}_{\psi} \right) \text{Dev} \mathbf{B} + \frac{\partial_{\gamma_t} \tilde{f}_{\psi}}{(\gamma_s - \gamma_t) J^{\frac{7}{3}}} \text{Dev} (\mathbf{B}^2 - \text{Tr}(\mathbf{B}) \mathbf{B}) \right). \quad (19)$$

134 This is the most general isotropic thermo-elastic law, formulated with respect to the variables $(T, J, \gamma_s, \gamma_t)$.
 135 Postulating that the free energy does not depend on the γ_t invariant, i.e. $\partial_{\gamma_t} \tilde{f}_\psi = 0$, reduces (19) to the
 136 elastic law used by [24, 26, 29, 31]. It should be noted, however, that in these papers, the expression
 137 of the free energy is presented as a hypothesis and is not deduced from experimentally assessed stresses.
 138 These models, being limited to the triplet (T, J, γ_s) , choose not to distinguish energetically two different
 139 deformations which would share the same maximum distortion δ_{\max} and thus the same invariant γ_s .

140 3. Experimental identification

141 The thermodynamic construction presented herein follows the generic method presented by Guarrigues
 142 [16, 17].

143 Let \tilde{f}_ψ denote the free energy variation within the non-isothermal thermo-elastic process from a reference
 144 configuration, $E_0 = (T_0, 1, 0, 0)$ for which the free energy is set to zero, to an arbitrary configuration given
 145 by the state variables $(T, J, \gamma_s, \gamma_t)$:

$$\psi^m = \tilde{f}_\psi(T, J, \gamma_s, \gamma_t). \quad (20)$$

To define the Helmholtz massic free energy at any point $(T, J, \gamma_s, \gamma_t)$ of the state space, we define a particular path

$$E_0 = (T_0, 1, 0, 0) \xrightarrow{\mathcal{P}^{(1)}} E_1 = (T, 1, 0, 0) \xrightarrow{\mathcal{P}^{(2)}} E_2 = (T, J, 0, 0) \xrightarrow{\mathcal{P}^{(3)}} E_3 = (T, J, \gamma_s, 0) \xrightarrow{\mathcal{P}^{(4)}} E_t = (T, J, \gamma_s, \gamma_t)$$

146 such that \tilde{f}_ψ is additively found, given the energy variation in every path:

$$\psi^m = g^{(1)}(T) + g^{(2)}(T, J) + g^{(3)}(T, J, \gamma_s) + g^{(4)}(T, J, \gamma_s, \gamma_t). \quad (21)$$

147 - $g^{(1)}(T)$ is the Helmholtz massic free energy variation within $\mathcal{P}^{(1)}$ (strain-locked heating):

$$\dot{T} \neq 0; \quad J = 1; \quad \dot{J} = 0; \quad \gamma_s = 0; \quad \dot{\gamma}_s = 0; \quad \gamma_t = 0; \quad \dot{\gamma}_t = 0; \quad (22)$$

148 - $g^{(2)}(T, J)$ is the Helmholtz massic free energy variation within $\mathcal{P}^{(2)}$ (isothermal spherical motion):

$$\dot{T} = 0; \quad \dot{J} \neq 0; \quad \gamma_s = 0; \quad \dot{\gamma}_s = 0; \quad \gamma_t = 0; \quad \dot{\gamma}_t = 0; \quad (23)$$

149 - $g^{(3)}(T, J, \gamma_s)$ is the Helmholtz massic free energy variation within $\mathcal{P}^{(3)}$ (isochoric simple shear test):

$$\dot{T} = 0; \quad \dot{J} = 0; \quad \dot{\gamma}_s \neq 0; \quad \gamma_t = 0; \quad \dot{\gamma}_t = 0; \quad (24)$$

150 - $g^{(4)}(T, J, \gamma_s, \gamma_t)$ is the Helmholtz massic free energy variation within $\mathcal{P}^{(4)}$ (isothermal isochoric iso-
 151 trace motion):

$$\dot{T} = 0; \quad \dot{J} = 0; \quad \dot{\gamma}_s = 0; \quad \dot{\gamma}_t \neq 0; \quad (25)$$

152 with the initial conditions:

$$g^{(1)}(T_0) = 0; \quad g^{(2)}(T, 1) = 0, \forall T; \quad g^{(3)}(T, J, 0) = 0, \forall (T, J); \quad g^{(4)}(T, J, \gamma_s, 0) = 0, \forall (T, J, \gamma_s); \quad (26)$$

153 which leads to cancel the following partial derivatives:

$$\begin{aligned} \partial_T g^{(2)}(T, 1) &= 0; \quad \partial_T g^{(3)}(T, J, 0) = \partial_J g^{(3)}(T, J, 0) = 0 \quad \forall T, J; \\ \partial_T g^{(4)}(T, J, \gamma_s, 0) &= \partial_J g^{(4)}(T, J, \gamma_s, 0) = \partial_{\gamma_s} g^{(4)}(T, J, \gamma_s, 0) = 0 \quad \forall T, J, \gamma_s. \end{aligned} \quad (27)$$

154 The general form of the massic entropy is given by the Helmholtz relation:

$$s^m = -\partial_T \tilde{f}_\psi = -\partial_T g^{(1)} - \partial_T g^{(2)} - \partial_T g^{(3)} - \partial_T g^{(4)}. \quad (28)$$

155 The massic internal energy is deduced from the definition of the Helmholtz free energy:

$$e^m = \tilde{f}_\psi + T s^m = g^{(1)} + g^{(2)} + g^{(3)} + g^{(4)} - T(\partial_T g^{(1)} + \partial_T g^{(2)} + \partial_T g^{(3)} + \partial_T g^{(4)}). \quad (29)$$

156 3.1. Elementary evolutions

157 The identification of the four unknown functions $g^{(1)}(T)$, $g^{(2)}(T, J)$, $g^{(3)}(T, J, \gamma_s)$ and $g^{(4)}(T, J, \gamma_s, \gamma_t)$
158 comes down to experimental measures in a few experiments carried under the following ideal experimental
159 conditions:

160 1) body forces are negligible;

161 2) state variables $(T, J, \gamma_s, \gamma_t)$ are uniform across the specimen.

162 3) experimental data are evaluated in a quasi-static setting so that the kinetic energy can be neglected;

163 $g^{(1)}(T)$ will be determined by a simple energy balance whereas direct integration of experimental stress data
164 shall clarify the rest of the Helmholtz free energy.

165 3.1.1. Strain-locked pure heating

166 The first evolution $\mathcal{P}^{(1)}$ is a pure heating, strain locked, experiment: $\mathbf{B} = \mathbf{1}$, $J = 1$, $\dot{J} = 0$, $\gamma_s = 0$,
167 $\dot{\gamma}_s = 0$, $\gamma_t = 0$, $\dot{\gamma}_t = 0$. We measure the algebraic massic heat $q_{exp}^{m(1)}(T)$ supplied to the system to go from T_0
168 to T . The conservation of energy between the states E_0 and E_1 is written (quasi-static evolution: $\Delta e_c^m = 0$,
169 no deformation: $w^{m, ext} = 0$):

$$e_1^m - \underbrace{e_0^m}_0 = q_{exp}^{m(1)}(T), \quad (30)$$

which leads to the differential equation in $g^{(1)}$ from (29):

$$\begin{aligned} g^{(1)}(T) - T(\partial_T g^{(1)}(T) + \partial_T g^{(2)}(T, 1) + \partial_T g^{(3)}(T, 1, 0)) + \partial_T g^{(4)}(T, 1, 0, 0) &= q_{exp}^{m(1)}(T), \\ \Leftrightarrow g^{(1)}(T) - T \partial_T g^{(1)}(T) &= q_{exp}^{m(1)}(T) \quad \text{see (27),} \\ \Leftrightarrow g^{(1)}(T) &= -T \int_{T_0}^T \frac{q_{exp}^{m(1)}(\tilde{T})}{\tilde{T}^2} d\tilde{T}. \end{aligned} \quad (31)$$

170 This experimental measurement is very difficult to perform, especially when the samples tend to contract
 171 (usually during cooling). We will see later, Section 3.1.5, that this ideal experiment can be replaced by a
 172 free thermal expansion followed by an isothermal spherical motion.

173 3.1.2. Spherical motion

174 In the second path $\mathcal{P}^{(2)}$, the deformation is purely spherical ($\mathbf{B} = J^{\frac{2}{3}}\mathbf{1}$, $\gamma_s = 0$, $\dot{\gamma}_s = 0$, $\gamma_t = 0$, $\dot{\gamma}_t = 0$)
 175 and isothermal ($\dot{T} = 0$). The elastic law (19) proves that stress tensor must also be spherical:

$$\boldsymbol{\sigma}^{(2)} = \rho_0 \partial_J \tilde{f}_\psi \mathbf{1}, \quad (32)$$

176 and is thus completely characterized by its mean stress.

177 In this experiment, we measure the pressure $p_{exp}^{(2)}(T, J)$ which is the opposite of the uniform normal stress
 178 applied on the boundary if the hypothesis 2) p.8 holds:

$$p_{exp}^{(2)}(T, J) = -\frac{\text{Tr}\boldsymbol{\sigma}^{(2)}}{3} = -\rho_0 \partial_J g^{(2)}(T, J) \quad \text{see (27)}. \quad (33)$$

179 The second contribution to the Helmholtz massic free energy variation then follows naturally.

$$g^{(2)}(T, J) = -\frac{1}{\rho_0} \int_1^J p_{exp}^{(2)}(T, \tilde{J}) d\tilde{J}. \quad (34)$$

180 In the shock-wave mechanics community, the determination of this pressure with respect to the volumetric
 181 change (or equivalently to the mass density ρ) and the temperature, is often referred to as the “equation of
 182 state (EOS)” [1].

183 3.1.3. Isochoric iso- T planar simple shear test

184 To perform an isochoric isothermal planar simple shear [18] in the $(\mathbf{e}_x, \mathbf{e}_y)$ plane with an initial volume
 185 expansion J , we impose the following position field on the particles:

$$\mathbf{x}_t = J^{\frac{1}{3}}(\mathbf{x}_0 + \gamma Y \mathbf{e}_x), \quad (35)$$

186 where $\mathbf{x}_0 = X\mathbf{e}_x + Y\mathbf{e}_y + Z\mathbf{e}_z$ is the reference position of a particle. The Finger tensor $\mathbf{B} = \mathbf{F} \cdot \mathbf{F}^\top$ in the
 187 orthonormal basis $(\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z)$ for this isochoric motion have already been given (16). The strain tensor is
 188 uniform in the specimen $\partial_{\mathbf{x}_0} \mathbf{B} = \mathbf{0}$ so that γ_s is uniform as well and satisfies: $\gamma_s = \sqrt{\overline{B_I} - 3} = \sqrt{\gamma^2} = \pm\gamma$.

189 *Stress tensor in the $\mathcal{P}^{(3)}$ evolution.* The general thermo-elastic law reads:

$$\boldsymbol{\sigma} = \rho_0 \left(\partial_J \tilde{f}_\psi \mathbf{1} + \frac{1}{\gamma_s J^{\frac{5}{3}}} \left(\partial_{\gamma_s} \tilde{f}_\psi + \partial_{\gamma_t} \tilde{f}_\psi \right) \text{Dev} \mathbf{B} + \frac{\partial_{\gamma_t} \tilde{f}_\psi}{(\gamma_s - \gamma_t) J^{\frac{7}{3}}} \text{Dev} (\mathbf{B}^2 - \text{Tr}(\mathbf{B})\mathbf{B}) \right). \quad (36)$$

190 Given the equalities $\gamma_t = 0$, $\partial_{\gamma_s} g^{(4)}(T, J, \gamma_s, 0) = 0$, simple calculations show that the tangential stress
 191 $\tau_{exp}^{(3)}(T, J, \gamma_s) = \sigma_{12}^{(3)}$ within this evolution is directly related to $\partial_{\gamma_s} g^{(3)}$ by:

$$\tau_{exp}^{(3)} = \frac{\rho_0}{J} \partial_{\gamma_s} g^{(3)}(T, J, \gamma_s). \quad (37)$$

In this experiment, we measure the tangential stress $\tau_{exp}^{(3)}$ as a function of γ and we deduce by a simple integration the value of the function $g^{(3)}$:

$$g^{(3)} = \frac{J}{\rho_0} \int_0^{\gamma_s} \tau_{exp}^{(3)}(T, J, \gamma) d\gamma. \quad (38)$$

3.1.4. Iso- γ_s iso- T isochoric double shear test

$\mathcal{P}^{(4)}$ is an isothermal, isochoric, iso- γ_s experiment. Several tests grants this triple condition. For the sake of simplicity, we introduce herein the “double shear test”, eventhough this experiment does not scan the whole domain of variation of γ_t . In the Appendix D, we will present a wider two-parameter test family, which include the present “double shear experiment”, and which browse the whole domain for the variable γ_t .

Isochoric double shear test. Consider the motion defined by the following transformation (see also Figure 2):

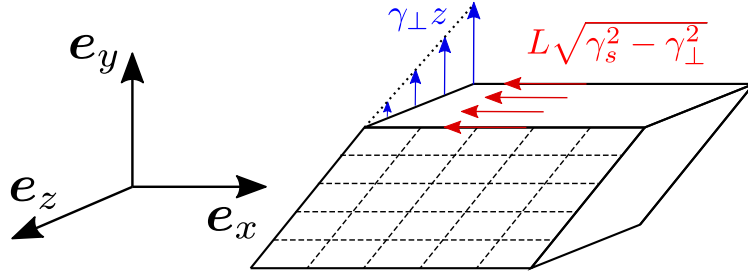


Figure 2: Double shear kinematic

$$\mathbf{x}_t = J^{\frac{1}{3}} (\mathbf{x}_0 + \sqrt{\gamma_s^2 - \gamma_{\perp}^2} Y \mathbf{e}_x + \gamma_{\perp} Z \mathbf{e}_y), \quad (39)$$

where $\mathbf{x}_0 = X \mathbf{e}_x + Y \mathbf{e}_y + Z \mathbf{e}_z$ is the reference position of a particle, J is the volume dilatation prior from path $\mathcal{P}^{(2)}$ and constant in path $\mathcal{P}^{(3)}$ and $\mathcal{P}^{(4)}$, γ_s is the slip prior from path $\mathcal{P}^{(3)}$ and constant in path $\mathcal{P}^{(4)}$. The components of the Finger tensor $\mathbf{B} = \mathbf{F} \cdot \mathbf{F}^T$ in the orthonormal basis $\{\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z\}$ for this isochoric motion are:

$$\mathbf{F} = J^{\frac{1}{3}} \begin{pmatrix} 1 & \sqrt{\gamma_s^2 - \gamma_{\perp}^2} & 0 \\ 0 & 1 & \gamma_{\perp} \\ 0 & 0 & 1 \end{pmatrix} \Rightarrow \overline{\mathbf{B}} = \begin{pmatrix} 1 + \gamma_s^2 - \gamma_{\perp}^2 & \sqrt{\gamma_s^2 - \gamma_{\perp}^2} & 0 \\ \sqrt{\gamma_s^2 - \gamma_{\perp}^2} & 1 + \gamma_{\perp}^2 & \gamma_{\perp} \\ 0 & \gamma_{\perp} & 1 \end{pmatrix} \Rightarrow \overline{B}_I = \gamma_s^2 + 3. \quad (40)$$

This test continuously shifts the deformation from a simple shear test in the $(\mathbf{e}_x, \mathbf{e}_y)$ plane to the same state in the $(\mathbf{e}_y, \mathbf{e}_z)$ plane while keeping the first invariant γ_s constant.

Stress tensor in the double shear test. Injecting the strain measure deduced from (39) in (36), one can see that the out of plane shear stress $\sigma_{13}^{(4)}$ is directly related to the derivative of the Helmholtz massic free energy with

respect to γ_t . Indeed, \bar{B}_{13}^2 given by (D.2) and (36) proves that the tangential stress $\tau_{exp}^{(4)}(T, J, \gamma_s, \gamma_t) = \sigma_{13}^{(4)}$ in this evolution equalized:

$$\begin{aligned}\tau_{exp}^{(4)}(T, J, \gamma_s, \gamma_\perp) &= \frac{\rho_0}{J(\gamma_s - \gamma_t)} \partial_{\gamma_t} g^{(4)}(T, J, \gamma_s, \gamma_t) \gamma_\perp \sqrt{\gamma_s^2 - \gamma_\perp^2} \text{ with } \gamma_t = \gamma_s - \sqrt{\gamma_s^2 + \gamma_\perp^2 (\gamma_s^2 - \gamma_\perp^2)} \text{ see (D.9),} \\ \tau_{exp}^{(4)}(T, J, \gamma_s, \gamma_t) &= \frac{\rho_0}{J(\gamma_s - \gamma_t)} \partial_{\gamma_t} g^{(4)}(T, J, \gamma_s, \gamma_t) \sqrt{(\gamma_s - \gamma_t)^2 - \gamma_s^2}.\end{aligned}\tag{41}$$

The expression for the massic free energy $g^{(4)}$ is obtained once more by integrating the experimental data:

$$g^{(4)}(T, J, \gamma_s, \gamma_t) = \frac{J}{\rho_0} \int_0^{\gamma_t} \frac{\gamma_s - \tau}{\sqrt{(\gamma_s - \tau)^2 - \gamma_s^2}} \tau_{exp}^{(4)}(T, J, \gamma_s, \tau) d\tau.\tag{42}$$

When γ_\perp evolves in the interval $\left[0, \frac{\gamma_s}{\sqrt{2}}\right]$, the invariant γ_t takes its values in $\gamma_t \in \left[\gamma_s \left(1 - \sqrt{1 + \frac{\gamma_s^2}{4}}\right), 0\right]$ (see Appendix D).

Note that this out-of-plane shear-stress $\sigma_{13}^{(4)}$ is purely “non-linear” in the sense that it arises from the quadratic term of the strain measure, therefore, it cannot be predicted by any “linearized” versions of the elastic law or by any model neglecting the contribution of the second strain invariant \bar{B}_{II} .

The “double shear experiment” is not a standard laboratory test but can be performed using an efficient hexapod machine (Stewart machine [11, 34]).

3.1.5. Free thermal expansion ($\mathcal{P}^{(5)}$)

A direct measurement of the massic heat $q_{exp}^{m(1)}$ in the first path $\mathcal{P}^{(1)}$ (strain-locked heating) is rather difficult. This experiment can be replaced by a free-stress thermal expansion ($p_{exp}^{(2)}(T, J_{exp}^{(5)}(T)) = 0, \forall T$), followed by an isothermal spherical deformation which brings the volume expansion $J_{exp}^{(5)}(T)$ back to 1.

Writing the conservation of energy for this evolution between the initial state $(T_0, 1, 0, 0)$ and the freely expanded state $(T, J_{exp}^{(5)}, 0, 0)$, it follows: $e_5^m = q_{exp}^{m(5)}(T)$. Moreover, the massic internal energy in the freely expanded state $(T, J_{exp}^{(5)}, 0, 0)$ is also given by (29):

$$e_5^m = q_{exp}^{m(1)}(T) - \frac{1}{\rho_0} \int_1^{J_{exp}^{(5)}} p_{exp}^{(2)}(T, \tilde{J}) - T \partial_T p_{exp}^{(2)}(T, \tilde{J}) d\tilde{J},\tag{43}$$

$$\Rightarrow q_{exp}^{m(1)}(T) = q_{exp}^{m(5)}(T) + \frac{1}{\rho_0} \int_1^{J_{exp}^{(5)}} p_{exp}^{(2)}(T, \tilde{J}) - T \partial_T p_{exp}^{(2)}(T, \tilde{J}) d\tilde{J}.\tag{44}$$

(44) proves that given the experimental pressure $p_{exp}^{(2)}(T, J)$ and the free-stress expansion massic heat $q_{exp}^{m(5)}(T)$, one may determine $q_{exp}^{m(1)}(T)$. The function (31) is then finally written:

$$g^{(1)}(T) = -T \int_{T_0}^T \frac{q_{exp}^{m(5)}(\tilde{T})}{\tilde{T}^2} d\tilde{T} - \frac{T}{\rho_0} \int_{T_0}^T \left(\int_1^{J_{exp}^{(5)}(\tilde{T})} p_{exp}^{(2)}(\tilde{T}, \tilde{J}) - \tilde{T} \partial_{\tilde{T}} p_{exp}^{(2)}(\tilde{T}, \tilde{J}) d\tilde{J} \right) d\tilde{T}.\tag{45}$$

228 3.2. Synthesis

229 The complete identification of the finite strain thermo-elastic model relies on four experimental data:

- 230 1) $q_{exp}^{m(1)}(T)$: massic heat exchanged in the first path $\mathcal{P}^{(1)}$ (strain-locked heating or cooling), with the
 231 condition:

$$q_{exp}^{m(1)}(T_0) = 0. \quad (46)$$

- 232 2) $p_{exp}^{(2)}(T, J)$: pressure in the spherical evolution $\mathcal{P}^{(2)}$ (isothermal spherical deformation), with the con-
 233 dition:

$$p_{exp}^{(2)}(T, 1) = p_{exp}^{(1)}(T), \forall T. \quad (47)$$

- 234 3) $\tau_{exp}^{(3)}(T, J, \gamma_s)$: in-plane shear stress in the isochoric planar shear test $\mathcal{P}^{(3)}$, along with the condition:

$$\tau_{exp}^{(3)}(T, J, 0) = 0, \forall T \forall J. \quad (48)$$

- 235 4) $\tau_{exp}^{(4)}(T, J, \gamma_s, \gamma_t)$: out-of-plane tangential stress in the fourth path $\mathcal{P}^{(4)}$, with the condition:

$$\tau_{exp}^{(4)}(T, J, \gamma_s, 0) = 0, \forall T \forall J \forall \gamma_s. \quad (49)$$

$g^{(1)}, g^{(2)}, g^{(3)}, g^{(4)}$ which determine the state functions are expressed in terms of these measurements:

$$g^{(1)}(T) = -T \int_{T_0}^T \frac{q_{exp}^{m(1)}(\tilde{T})}{\tilde{T}^2} d\tilde{T} \text{ or (45),} \quad (50)$$

$$g^{(2)}(T, J) = -\frac{1}{\rho_0} \int_1^J p_{exp}^{(2)}(T, \tilde{J}) d\tilde{J}, \quad (51)$$

$$g^{(3)}(T, J, \gamma_s) = \frac{J}{\rho_0} \int_0^{\gamma_s} \tau_{exp}^{(3)}(T, J, \gamma) d\gamma, \quad (52)$$

$$g^{(4)}(T, J, \gamma_s, \gamma_t) = \frac{J}{\rho_0} \int_0^{\gamma_t} \frac{\gamma_s - \tau}{\sqrt{(\gamma_s - \tau)^2 - \gamma_s^2}} \tau_{exp}^{(4)}(T, J, \gamma_s, \tau) d\tau. \quad (53)$$

236 The internal massic energy is assumed to be a state function, as to ensure (29) takes a simpler expression:

$$e^m = q_{exp}^{m(1)} + g^{(2)} - T \partial_T g^{(2)} + g^{(3)} - T \partial_T g^{(3)} + g^{(4)} - T \partial_T g^{(4)}. \quad (54)$$

237 For the numerical assessment of the model, it is not necessary to compute explicitly $g^{(1)}$ because this function
 238 does not appear neither in the constitutive law, nor in the internal energy.

239 3.3. Volumetric/isochoric splitting assumption

As already mentioned, hyperelastic materials exhibit radically different volume and shear behavior. This explains a well-accepted splitting of the Helmholtz free energy with respect to the isochoric/volumetric part of the strain tensor (see [24, 27]). In this section, we emphasize that this partition comes down to a very

natural assertion: the pressure remains unchanged in isochoric evolutions. With the previous results, the pressure in any state $(T, J, \gamma_s, \gamma_t)$ is:

$$\begin{aligned} -\frac{\text{Tr}\boldsymbol{\sigma}}{3} &= -\rho_0 \partial_J \left(g^{(2)}(T, J) + g^{(3)}(T, J, \gamma_s) + g^{(4)}(T, J, \gamma_s, \gamma_t) \right) \\ &= p_{exp}^{(2)}(T, J) + \int_0^{\gamma_s} \tau_{exp}^{(3)}(T, J, \gamma_s) - J \partial_J \tau_{exp}^{(3)}(T, J, \gamma_s) d\gamma \\ &\quad - \int_0^{\gamma_t} \frac{\gamma_s - \tau}{\sqrt{(\gamma_s - \tau)^2 - \gamma_s^2}} \left(\tau_{exp}^{(4)} + J \partial_J \tau_{exp}^{(4)} \right) (T, J, \gamma_s, \tau) d\tau. \end{aligned} \quad (55)$$

The pressure in any state is a function of the tangential stresses $\tau_{exp}^{(3)}$, $\tau_{exp}^{(4)}$ measured during the paths $\mathcal{P}^{(3)}$, $\mathcal{P}^{(4)}$. If one performs the $\mathcal{P}^{(3)}$ experiment from the initial state $(p_{exp}^{(2)}(T_0, 1) = 0$, no prior volume expansion), the Cauchy stress tensor generated is not deviatoric.

Hypothesis Optional Simplification. *In isochoric deformation, the pressure does not vary. Both conditions $\partial_{\gamma_t} \text{Tr}\boldsymbol{\sigma} = 0$, $\partial_{\gamma_s} \text{Tr}\boldsymbol{\sigma} = 0$ and (55) lead to the differential equations:*

$$\begin{aligned} \tau_{exp}^{(4)} + J \partial_J \tau_{exp}^{(4)} &= 0 \Rightarrow \tau_{exp}^{(4)}(T, J, \gamma_s, \gamma_t) = \frac{\hat{\tau}_{exp}^{(4)}(T, \gamma_s, \gamma_t)}{J}, \\ \tau_{exp}^{(3)} + J \partial_J \tau_{exp}^{(3)} &= 0 \Rightarrow \tau_{exp}^{(3)}(T, J, \gamma_s) = \frac{\hat{\tau}_{exp}^{(3)}(T, \gamma_s)}{J}. \end{aligned} \quad (56)$$

Inserting these expressions into the general formula (52)–(53), (56) obviously split the Helmholtz free energy into an isochoric/volumetric contributions:

$$\psi^m(T, J, \gamma_s, \gamma_t) = g^{(1)}(T) + g^{(2)}(T, J) + g^{(3)}(T, \gamma_s) + g^{(4)}(T, \gamma_s, \gamma_t). \quad (57)$$

This assumption is certainly valid in a wide range of solicitation (e.g is consistent with the classical linearized elasticity), but must be somehow validated experimentally. Note that the complete model (50)–(53) can take into account the volumetric/deviatoric coupling which is relevant for high velocity impact [28]. Indeed, extreme strain-rate experiments can induced finite elastic deformations before plasticity occurs, so that pressure-dependent shear modulus are required (see for instance the SCG model [2, 33]).

3.4. Time derivative of the massic internal energy

The massic internal energy takes a rather elegant form:

$$\begin{aligned} e^m &= q_{exp}^{m(5)}(T) - \frac{1}{\rho_0} \int_{J_{exp}^{(5)}(T)}^J (p_{exp}^{(2)} - T \partial_T p_{exp}^{(2)})(T, \tilde{J}) d\tilde{J} + \frac{1}{\rho_0} \int_0^{\gamma_s} \left(\hat{\tau}_{exp}^{(3)} - T \partial_T \hat{\tau}_{exp}^{(3)} \right) (T, \tilde{\gamma}_s) d\tilde{\gamma}_s \\ &\quad + \frac{1}{\rho_0} \int_0^{\gamma_t} \frac{\gamma_s - \tau}{\sqrt{(\gamma_s - \tau)^2 - \gamma_s^2}} \left(\hat{\tau}_{exp}^{(4)} - T \partial_T \hat{\tau}_{exp}^{(4)} \right) (T, \gamma_s, \tau) d\tau. \end{aligned} \quad (58)$$

The time derivative of the internal energy, essential for the numerical implementation, is obtained by evaluating the partial derivatives of the massic internal energy with respect to its four variables (see Appendix E for details):

$$\frac{de^m}{dt} = \frac{\partial e^m}{\partial T} \dot{T} + \frac{\partial e^m}{\partial J} \dot{J} + \frac{\partial e^m}{\partial \gamma_s} \dot{\gamma}_s + \frac{\partial e^m}{\partial \gamma_t} \dot{\gamma}_t. \quad (59)$$

4. Idealizations of elementary paths

The expressions (50)–(53) yields the “exact” thermo-elastic behavior of an isotropic material and lies upon four experimental results. In this section, we will propose physically consistent expressions for the experimental stresses. We will see that some rather simple forms reduce our model to a few hyperelastic models classically found in the literature.

4.1. Pressure assumption

Hypothesis Experimental pressure $p_{exp}^{(2)}$. We assume the following form for the pressure $p_{exp}^{(2)}$ in any isothermal spherical deformation:

$$p_{exp}^{(2)}(T, J) = p_{exp}^{(1)}(T) - \kappa(T) \ln J, \quad (60)$$

where $p_{exp}^{(1)}(T)$ is the pressure from the path $\mathcal{P}^{(1)}$

$\kappa(T)$ is a temperature-dependent bulk modulus.

The pressure (60) is reasonable (note for instance $\lim_{J \rightarrow \infty} p_{exp}^{(2)} = -\infty$, $\lim_{J \rightarrow 0} p_{exp}^{(2)} = \infty$), but predicts a finite strain energy under infinite compression $J \rightarrow 0$. For further details about the volumetric part of the Helmholtz free energy from which the pressure is derived, we refer to [12] and references therein. A more reasonable approximation of the experimental stress would be:

$$p_{exp}^{(2)} = p_{exp}^{(1)}(T) - \frac{\kappa(T)}{2} \left(J - \frac{1}{J} \right), \quad (61)$$

$$\Rightarrow g^{(2)}(T, J) = -\frac{(J-1)p_{exp}^{(1)}(T)}{\rho_0} + \frac{\kappa(T)}{2} \left(\frac{1}{2} (J^2 - 1) - \ln J \right). \quad (62)$$

It’s worth noticing that (62) coincides with that proposed by [31] which is a variant of [8].

For the sake of calculus simplicity, we will however keep the pressure given by (60) for the following, noting that the differences between (60) and (61) are very small for any volumic expansion up to 200%.

Both model comes down to the so called linearized elasticity when $J \approx 1$ as $\ln J \approx \frac{1}{2} \left(J - \frac{1}{J} \right) \approx \text{Tr} \epsilon$.

$g^{(2)}$ is obtained using both (60) and (51):

$$g^{(2)}(T, J) = -\frac{(J-1)p_{exp}^{(1)}(T)}{\rho_0} + \frac{\kappa(T)}{\rho_0} (J \ln(J) + 1 - J). \quad (63)$$

If we replace the first path $\mathcal{P}^{(1)}$ by a free expansion $\mathcal{P}^{(5)}$ (in which one save $q_{exp}^{m(5)}(T)$ and $J_{exp}^{(5)}(T)$) followed by an isothermal spherical compression to bring J back to 1, $p_{exp}^{(1)}(T)$ is obtained by evaluating (60) in $(T, J_{exp}^{(5)})$. Remembering that the stress is zero in a freely expanded test, it comes:

$$p_{exp}^{(1)}(T) = \kappa(T) \ln \left(J_{exp}^{(5)}(T) \right). \quad (64)$$

Therefore, by reinjecting this expression (64) into the general formula (60), we deduce the expression of $p_{exp}^{(2)}$:

$$p_{exp}^{(2)}(T, J) = -\kappa(T) \ln \frac{J}{J_{exp}^{(5)}(T)}. \quad (65)$$

Hypothesis Free-stress volume expansion $J_{exp}^{(5)}(T)$. One can set arbitrary for $J_{exp}^{(5)}(T)$:

$$J_{exp}^{(5)}(T) = 1 + \beta(T - T_0), \quad (66)$$

where β refers to a free-stress expansion coefficient.

Inserting both the experimental pressure (64) and the free-stress volume expansion (66) in (63), $g^{(2)}(T, J)$ now yields:

$$g^{(2)}(T, J) = \frac{\kappa(T)}{\rho_0} \left(J \ln \left(\frac{J}{1 + \beta(T - T_0)} \right) + 1 - J + \ln(1 + \beta(T - T_0)) \right). \quad (67)$$

4.2. Massic heat $q_{exp}^{m(1)}$

As a reminder, the first path $\mathcal{P}^{(1)}$ can be substituted by a free thermal expansion followed by isothermal compression. We then obtain:

$$q_{exp}^{m(1)}(T) = q_{exp}^{m(5)}(T) + \frac{1}{\rho_0} \int_1^{J_{exp}^{(5)}} p_{exp}^{(2)}(T, \tilde{J}) - T \partial_T p_{exp}^{(2)}(T, \tilde{J}) d\tilde{J} \quad [see (44)]. \quad (68)$$

The conservation of energy in a free expansion is then written according to (65):

$$q_{exp}^{m(5)}(T) = q_{exp}^{m(1)}(T) + \frac{1}{\rho_0} \left(T \kappa(T) \frac{\partial_T J_{exp}^{(5)}}{J_{exp}^{(5)}} (J_{exp}^{(5)} - 1) + (\kappa(T) - T \kappa'(T)) (1 + \ln(J_{exp}^{(5)}) - J_{exp}^{(5)}) \right). \quad (69)$$

We can choose a simple function of $q_{exp}^{m(5)}(T)$ or $q_{exp}^{m(1)}(T)$ the other being determined by (69).

It is noteworthy that the idealizations of experimental curves presented in this section are only arbitrary examples that have no theoretical justification.

Hypothesis Massic heat $q_{exp}^{m(5)}(T)$. A physically reasonable approximation to $q_{exp}^{m(5)}(T)$ may be, for example:

$$q_{exp}^{m(5)}(T) = C_p(T - T_0), \quad (70)$$

where C_p refers to a specific heat capacity in free expansion.

4.3. Calculation of $g^{(3)}$

Hypothesis In-plane shear stress $\hat{\tau}_{exp}^{(3)}$. We assume that a temperature dependent simple shear modulus $\mu(T)$ linearly links the shear stress with respect to the shear parameter γ :

$$\hat{\tau}_{exp}^{(3)}(T, J, \gamma) = \mu(T) \gamma. \quad (71)$$

297 The shear stress (71) leads to a quadratic massic free energy with respect to γ_s that is also independent of
 298 the volume expansion J :

$$g^{(3)}(T, \gamma_s) = \frac{\mu(T)\gamma_s^2}{2\rho_0}. \quad (72)$$

299 This Helmholtz massic free energy has for example been postulated in [29]. We now choose to enhance the
 300 model considering the contribution of the second invariant \bar{B}_{II} through γ_t .

301 4.4. Calculation of $g^{(4)}$

302 The out-of-plane shear stress is a purely non-linear effect and its interpretation in terms of a “classical”
 303 shear or compressibility coefficient is therefore impossible.

304 **Hypothesis** Out-of-plane shear stress $\hat{\tau}_{exp}^{(4)}$. We postulate that the shear stress $\hat{\tau}_{exp}^{(4)}$ in the fourth path
 305 $\mathcal{P}^{(4)}$ is given by:

$$\hat{\tau}_{exp}^{(4)}(T, \gamma_s, \tau) = \alpha(T)\sqrt{(\gamma_s - \tau)^2 - \gamma_s^2}, \quad (73)$$

306 $\alpha(T)$ refers to a non-linear shear modulus. We further recall that the invariant γ_t is always negative during
 307 the double shear test so that the square root involved in (73) is well-defined. Using the expression (73) $g^{(4)}$
 308 now reads:

$$g^{(4)}(T, \gamma_s, \gamma_t) = \frac{\alpha(T)}{\rho_0} \int_0^{\gamma_t} (\gamma_s - \tau) d\tau = \frac{\alpha(T)}{2\rho_0} (\gamma_s^2 - (\gamma_s - \gamma_t)^2). \quad (74)$$

309 4.5. Idealized elastic law

310 With the idealizations of the experimental stresses (60), (71), (73) the $g^{(\bullet)}$ functions were explicitly
 311 calculated (63), (72), (74). The derivation of the Helmholtz free energy yields the current Cauchy stress:

$$\boldsymbol{\sigma} = \rho_0 \left(\partial_J \tilde{f}_\psi \mathbf{1} + \frac{1}{\gamma_s J^{\frac{5}{3}}} (\mu(T)\gamma_s + \alpha(T)(\gamma_s - \gamma_t)) \mathbf{Dev} \mathbf{B} + \frac{\alpha(T)}{J^{\frac{7}{3}}} \mathbf{Dev} (\mathbf{B}^2 - \text{Tr}(\mathbf{B})\mathbf{B}) \right). \quad (75)$$

312 The invariant γ_t is upper bounded by γ_s whereas the positivity of the second invariant J_2^B implies the lower
 313 bound:

$$J_2^B = \sqrt{\frac{3}{2}} \|\mathbf{Dev} \mathbf{B}\| \geq 0 \Rightarrow \gamma_t \geq \gamma_s \left(1 - \sqrt{\frac{1 + \gamma_s^2}{4}} \right). \quad (76)$$

314 Finally $\lim_{\gamma_s \rightarrow 0} \frac{\gamma_t}{\gamma_s} = 0$ so that (75) does not diverge when $\gamma_s \rightarrow 0$. As we will see in the following section, the
 315 expression (75) is algebraically equivalent to a standard Mooney-Rivlin model.

316 5. Analogy with the Mooney-Rivlin model

317 If we retain the idealizations (71), (73), the dependence of the massic free energy with respect to the
 318 isochoric invariants is written:

$$\begin{aligned} g^{(3)}(T, \gamma_s) + g^{(4)}(T, \gamma_s, \gamma_t) &= \frac{1}{2\rho_0} (\mu(T)\gamma_s^2 + \alpha(T)(\gamma_s^2 - (\gamma_s - \gamma_t)^2)) \\ &= \frac{1}{2\rho_0} (\mu(T)(\bar{B}_I - 3) + \alpha(T)((\bar{B}_I - 3) - (\bar{B}_{II} - 3))). \end{aligned} \quad (77)$$

319 The right and left Cauchy-Green strain tensors \mathbf{C} and \mathbf{B} share their invariants $B_i = C_i$, as \mathbf{C} and \mathbf{B} are
 320 related through the rotation tensor \mathbf{R} arising from the polar decomposition of the transformation gradient
 321 $\mathbf{F} = \mathbf{R} \cdot \mathbf{U} = \mathbf{V} \cdot \mathbf{R}$. Therefore the free energy (77) coincides exactly with that of a Mooney-Rivlin model
 322 [22]:

$$\psi_{M-R}^m = g^1(T) + g^2(T, J) + \frac{C_{01}}{\rho_0} (\bar{B}_I - 3) + \frac{C_{10}}{\rho_0} (\bar{B}_{II} - 3), \quad (78)$$

323 with the parameters $C_{01} = \frac{1}{2}(\mu(T) + \alpha(T))$, $C_{10} = -\frac{\alpha(T)}{2}$.

324 If we assume that the shear stress along the $\mathcal{P}^{(4)}$ path is identically zero, the massic free energy be-
 325 comes independent of the γ_t invariant and we then observe that the Helmholtz massic free energy evolves
 326 linearly with $\bar{B}_I - 3$, which corresponds to the Neo-Hookean model $\psi_{N-H}^m = g^1(T) + g^2(T, J) + \frac{\mu}{2\rho_0} (\bar{B}_I - 3)$.
 327 Consequently our thermodynamic approach includes these two models which result from the experimental
 328 idealizations (71) (73).

329
 330 More generally, if one looks at the in-plane shear stress curve $\tau_{exp}^{(3)}$ as a function of the slip parameter
 331 γ , it is certain that a polynomial expansion fitting experimental data can be found. By symmetry, this
 332 polynomial expansion must necessarily contain only even powers:

$$\hat{\tau}_{exp}^{(3)}(T, \gamma) = \sum_i \mu_i \gamma^{2i+1}. \quad (79)$$

333 (79) then generates a generalized Neo-Hookean model (power series of the free energy with respect to the
 334 “ $\bar{B}_I - 3$ ” invariant).

335 Moreover, if we postulate by the same reasoning that the out-of-plane shear stress $\hat{\tau}_{exp}^{(4)}$ develops in the
 336 form:

$$\hat{\tau}_{exp}^{(3)}(T, \gamma) = \sum_i \mu_i \gamma^{2i+1} \text{ and } \hat{\tau}_{exp}^{(4)}(T, \gamma_s, \tau) = \sum_i \alpha_i \sqrt{(\gamma_s - \tau)^2 - \gamma_s^2} (\gamma_s - \tau)^{2i}. \quad (80)$$

337 Then one found the following Helmholtz massic free energy:

$$\begin{aligned} \psi^m &= \frac{1}{\rho_0} \sum_i \frac{\mu_i(T)}{2i+2} (\bar{B}_I - 3)^i + \frac{\alpha_i(T)}{2i+2} [(\bar{B}_I - 3)^i - (\bar{B}_{II} - 3)^i] + g^{(1)}(T) + g^{(2)}(T, J), \\ \psi^m &= \frac{1}{\rho_0} \sum_i C_{i0} (\bar{B}_I - 3)^i + C_{0i} (\bar{B}_{II} - 3)^i + g^{(1)}(T) + g^{(2)}(T, J) \quad C_{ij} = 0 \text{ if } i \neq j. \end{aligned} \quad (81)$$

338 That is, a generalized Mooney-Rivlin material [22].

339
 340 In the general case, the integral expressions given in section 3.2 yield the exact finite strain isotropic
 341 thermo-elastic constitutive law.

342 6. Conclusion

343 We have developed a complete finite-strain isotropic thermo-elastic model without any assumption con-
 344 cerning the form of the Helmholtz free energy. The identification of the latter relies on the successive

realization of four elementary experiments, and more precisely the measurement of: a massic heat, a pressure, and two shear stresses. Experiments must be carried many times in order to scan all the accessible values of the state variables e.g. the double shear test $\mathcal{P}^{(4)}$ must be performed for a whole collection of temperature T , volume expansion J , and slip γ_s .

Some idealizations of the experimental stresses have been proposed. They can be replaced by any realistic curves, or even be given by the upscaling results of a large scale molecular dynamic simulation.

The well known Mooney-Rivlin model, and consequently the Neo-Hookean, are special cases of our methodology. They are derived from an assumption concerning the shear stress along the $\mathcal{P}^{(3)}$ and $\mathcal{P}^{(4)}$ paths.

The present work could admit several natural extensions among which:

- The description of an anisotropic medium by the introduction of structural tensors $\{\mathbf{N}_t^\bullet\}$ representing the actual directions of anisotropy. Enhancing the Helmholtz free energy with these new unknowns necessarily yields cross-invariant effects, reflecting the current orientation of the deformation with respect to the anisotropic directions.
- A finite strain plasticity model with the addition of an objective plastic strain tensor \mathbf{B}^p in the arguments' list of the Helmholtz free energy. Taking this tensor into account adds 6 state variables. Three of them are specific to the plastic strain \mathbf{B}^p (2 if one further assumes that the plastic evolution is isochoric), three others orient the principal directions orthogonal trihedron of \mathbf{B} and \mathbf{B}^p .
- The study of large-amplitude shock waves and more particularly shock tails. Indeed, after a large plastic phase which is out of the scope of this study, the elastic release wave on the shock tail can be considered as large strain. A proper coupling between both, an equation of state driving the pressure, and the deviatoric elastic law presented herein is a very interesting perspective.

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Appendix A. Justification of the finite strain isotropic elastic law

The definition of the Finger tensor is $\mathbf{B} = \mathbf{F} \cdot \mathbf{F}^\top$, hence its time derivative reads:

$$\begin{aligned}\dot{\mathbf{B}} &= \dot{\mathbf{F}} \cdot \mathbf{F}^\top + \mathbf{F} \cdot \dot{\mathbf{F}}^\top = (\dot{\mathbf{F}} \cdot \mathbf{F}^{-1}) \cdot (\mathbf{F} \cdot \mathbf{F}^\top) + (\mathbf{F} \cdot \mathbf{F}^\top) \cdot (\mathbf{F}^{-\top} \cdot \dot{\mathbf{F}}^\top), \\ \dot{\mathbf{B}} &= (\mathbf{D} + \mathbf{W}) \cdot \mathbf{B} + \mathbf{B} \cdot (\mathbf{D} - \mathbf{W}).\end{aligned}\tag{A.1}$$

where \mathbf{D} (resp \mathbf{W}) denotes the symmetric (resp skew-symmetric) part of the spatial velocity gradient.

373 Using the combined product $\mathbf{A}_1 : (\mathbf{A}_2 \cdot \mathbf{A}_3) = \mathbf{A}_2 : (\mathbf{A}_1 \cdot \mathbf{A}_3^\top) = \mathbf{A}_3 : (\mathbf{A}_2^\top \cdot \mathbf{A}_1)$, we get successively,
 374 $\forall n \in \mathbb{Z}$:

$$\mathbf{B}^n : \dot{\mathbf{B}} = \mathbf{B}^n : ((\mathbf{D} + \mathbf{W}) \cdot \mathbf{B} + \mathbf{B} \cdot (\mathbf{D} - \mathbf{W})) = 2\mathbf{B}^{n+1} : \mathbf{D}. \quad (\text{A.2})$$

375 Orthogonality of the antisymmetric and symmetric tensors for the doubly contracted product has been used
 376 to simplify (A.2).

377 The time derivatives of the coefficients of the characteristic polynomials of \mathbf{B} are (see also [14, 15]):

$$\begin{aligned} \frac{d\mathbf{B}_I}{dt} &= \text{Tr} \frac{d\mathbf{B}}{dt} = \mathbf{1} : \dot{\mathbf{B}}; & \frac{d\mathbf{B}_{II}}{dt} &= (\mathbf{B}_I \mathbf{1} - \mathbf{B}^\top) : \frac{d\mathbf{B}}{dt}; & \frac{d\mathbf{B}_{III}}{dt} &= (\mathbf{B}_{II} \mathbf{1} - \mathbf{B}_I \mathbf{B}^\top + \mathbf{B}^{2\top}) : \frac{d\mathbf{B}}{dt} = \mathbf{B}_{III} \mathbf{B}^{-\top} : \frac{d\mathbf{B}}{dt}; \\ \dot{\mathbf{B}}_I &= 2\mathbf{B} : \mathbf{D}; & \dot{\mathbf{B}}_{II} &= 2(\mathbf{B}_I \mathbf{B} - \mathbf{B}^2) : \mathbf{D}; & \dot{\mathbf{B}}_{III} &= \mathbf{B}_{III} \mathbf{1} : \mathbf{D}; \end{aligned} \quad (\text{A.3})$$

378 The nullity of intrinsic dissipation (4), is written using the coincident isotropic function (5):

$$-\rho \left(\partial_T f_\psi^B \dot{T} + \partial_{B_I} f_\psi^B \dot{\mathbf{B}}_I + \partial_{B_{II}} f_\psi^B \dot{\mathbf{B}}_{II} + \partial_{B_{III}} f_\psi^B \dot{\mathbf{B}}_{III} + \dot{T} s^m \right) + \boldsymbol{\sigma} : \mathbf{D} = 0. \quad (\text{A.4})$$

379 Injecting both (A.3), (A.2) in (A.4) and using the fact that $\boldsymbol{\sigma}$ and f_ψ^B are state functions so that they do
 380 not depend on \mathbf{D} , the nullity of (A.4) $\forall T \forall \mathbf{D}$ yields the elastic law (6).

381 Appendix B. Thermo-elastic law with respect to isochoric invariants

382 The partial derivatives of the new function \tilde{f}_ψ^B defined by (11) are simple algebraic calculations:

$$\begin{aligned} \partial_{B_I} f_\psi^B &= \partial_{\bar{B}_I} \tilde{f}_\psi^B \partial_{B_I} \bar{B}_I = B_{III}^{-\frac{1}{3}} \partial_{\bar{B}_I} \tilde{f}_\psi^B, \\ \partial_{B_{II}} f_\psi^B &= \partial_{\bar{B}_{II}} \tilde{f}_\psi^B \partial_{B_{II}} \bar{B}_{II} = B_{III}^{-\frac{2}{3}} \partial_{\bar{B}_{II}} \tilde{f}_\psi^B, \\ \partial_{B_{III}} f_\psi^B &= \partial_{\bar{B}_I} \tilde{f}_\psi^B \partial_{B_{III}} \bar{B}_I + \partial_{\bar{B}_{II}} \tilde{f}_\psi^B \partial_{B_{III}} \bar{B}_{II} + \partial_{B_{III}} \tilde{f}_\psi^B, \\ \partial_{B_{III}} f_\psi^B &= -\partial_{\bar{B}_I} \tilde{f}_\psi^B \frac{\bar{B}_I}{3B_{III}} - \partial_{\bar{B}_{II}} \tilde{f}_\psi^B \frac{2\bar{B}_{II}}{3B_{III}} + \partial_{B_{III}} \tilde{f}_\psi^B. \end{aligned} \quad (\text{B.1})$$

383 And the elastic law (6) now reads:

$$\begin{aligned} \boldsymbol{\sigma} &= \frac{2\rho_0}{\sqrt{B_{III}}} \left(\left(B_{III} \partial_{B_{III}} \tilde{f}_\psi^B - \partial_{\bar{B}_I} \tilde{f}_\psi^B \frac{\bar{B}_I}{3} - \partial_{\bar{B}_{II}} \tilde{f}_\psi^B \frac{2\bar{B}_{II}}{3} \right) \mathbf{1} \right. \\ &\quad \left. + \left(B_{III}^{-\frac{1}{3}} \partial_{\bar{B}_I} \tilde{f}_\psi^B + B_{III}^{\frac{1}{3}} \bar{B}_I B_{III}^{-\frac{2}{3}} \partial_{\bar{B}_{II}} \tilde{f}_\psi^B \right) \mathbf{B} - B_{III}^{-\frac{2}{3}} \partial_{\bar{B}_{II}} \tilde{f}_\psi^B \mathbf{B}^2 \right), \\ \boldsymbol{\sigma} &= \frac{2\rho_0}{\sqrt{B_{III}}} \left(\left(B_{III} \partial_{B_{III}} \tilde{f}_\psi^B - \partial_{\bar{B}_I} \tilde{f}_\psi^B \frac{\bar{B}_I}{3} - \partial_{\bar{B}_{II}} \tilde{f}_\psi^B \frac{2\bar{B}_{II}}{3} \right) \mathbf{1} \right. \\ &\quad \left. + \left(\partial_{\bar{B}_I} \tilde{f}_\psi^B + \bar{B}_I \partial_{\bar{B}_{II}} \tilde{f}_\psi^B \right) \bar{\mathbf{B}} - \partial_{\bar{B}_{II}} \tilde{f}_\psi^B \bar{\mathbf{B}}^2 \right). \end{aligned} \quad (\text{B.2})$$

384 This formula is more elegantly written using the following relations, which hold for any tensor of order 2:

$$\begin{aligned} X_{II} = \frac{1}{2} (X_I^2 - \text{Tr} \mathbf{X}^2) &\Rightarrow -\frac{2}{3} X_{II} \mathbf{1} + X_I \mathbf{X} - \mathbf{X}^2 = -\frac{1}{3} (X_I^2 - \text{Tr} \mathbf{X}^2) \mathbf{1} + X_I \mathbf{X} - \mathbf{X}^2, \\ &= -\text{Dev} (\mathbf{X}^2 - \text{Tr} (\mathbf{X}) \mathbf{X}). \end{aligned} \quad (\text{B.3})$$

385 So that (B.2) is finally written:

$$\begin{aligned}\sigma &= \frac{2\rho_0}{\sqrt{B_{\text{III}}}} \left(B_{\text{III}} \partial_{B_{\text{III}}} \tilde{f}_\psi^B \mathbf{1} + \partial_{\bar{B}_I} \tilde{f}_\psi^B \text{Dev} \bar{\mathbf{B}} - \partial_{\bar{B}_{\text{II}}} \tilde{f}_\psi^B \text{Dev} \left(\bar{\mathbf{B}}^2 - \text{Tr}(\bar{\mathbf{B}}) \bar{\mathbf{B}} \right) \right), \\ \sigma &= \frac{2\rho_0}{\sqrt{B_{\text{III}}}} \left(B_{\text{III}} \partial_{B_{\text{III}}} \tilde{f}_\psi^B \mathbf{1} + \frac{\partial_{\bar{B}_I} \tilde{f}_\psi^B}{B_{\text{III}}^{\frac{1}{3}}} \text{Dev} \mathbf{B} - \frac{\partial_{\bar{B}_{\text{II}}} \tilde{f}_\psi^B}{B_{\text{III}}^{\frac{2}{3}}} \text{Dev} (\mathbf{B}^2 - \text{Tr}(\mathbf{B}) \mathbf{B}) \right).\end{aligned}\quad (\text{B.4})$$

386 Appendix C. Evolution domain of γ_t

Let $(\bar{\lambda}_i)_{i \in \llbracket 1,3 \rrbracket}$ be the principal stretches of the isochoric Finger tensor $\bar{\mathbf{B}}$. We further assume that the invariant γ_s remains fixed. The following system is necessarily verified by the triplet $(\bar{\lambda}_i)_{i \in \llbracket 1,3 \rrbracket}$:

$$\bar{\lambda}_1^2 + \bar{\lambda}_2^2 + \bar{\lambda}_3^2 = \gamma_s^2 + 3 = \bar{B}_I, \quad (\text{C.1})$$

$$\bar{\lambda}_1^2 \bar{\lambda}_2^2 + \bar{\lambda}_2^2 \bar{\lambda}_3^2 + \bar{\lambda}_1^2 \bar{\lambda}_3^2 = \bar{B}_{\text{II}}, \quad (\text{C.2})$$

$$\bar{\lambda}_1 \bar{\lambda}_2 \bar{\lambda}_3 = 1. \quad (\text{C.3})$$

387 By combining the equations (C.1) and (C.3), we deduce:

$$\bar{\lambda}_2^4 + \left(\bar{\lambda}_1^2 - \bar{B}_I \right) \bar{\lambda}_2^2 + \frac{1}{\bar{\lambda}_1^2} = 0. \quad (\text{C.4})$$

388 (C.4) has a positive or zero discriminant:

$$\Delta = \left(\bar{\lambda}_1^2 - \bar{B}_I \right)^2 - \frac{4}{\bar{\lambda}_1^2} \geq 0 \Rightarrow \bar{\lambda}_{2\pm}^2 = \frac{\bar{B}_I - \bar{\lambda}_1^2 \pm \sqrt{\left(\bar{\lambda}_1^2 - \bar{B}_I - \frac{2}{\bar{\lambda}_1} \right) \left(\bar{\lambda}_1^2 - \bar{B}_I + \frac{2}{\bar{\lambda}_1} \right)}}{2}. \quad (\text{C.5})$$

389 The positivity of the term under the square root implies that (C.4) has real solutions if and only if the
390 eigenvalue $\bar{\lambda}_1$ evolves in the interval:

$$\bar{\lambda} \in [X_2, X_0] \text{ with } X_k = 3\sqrt{\tilde{\alpha}} \cos \left(\frac{1}{3} \arccos \left(-(\tilde{\alpha})^{-\frac{3}{2}} \right) + \frac{2k\pi}{3} \right) \text{ with } \tilde{\alpha} = \frac{\bar{B}_I}{3} \geq 0. \quad (\text{C.6})$$

391 We can then express γ_t as a function of the unique variable $\bar{\lambda}_1$:

$$\bar{B}_{\text{II}}(\bar{\lambda}_1) = \bar{\lambda}_1^2 \bar{\lambda}_{2\pm}^2 + \frac{1}{\bar{\lambda}_1^2} + \frac{1}{\bar{\lambda}_{2\pm}^2} = \bar{\lambda}_1^2 \bar{B}_I - \bar{\lambda}_1^4 + \frac{1}{\bar{\lambda}_1^2} \Rightarrow \gamma_t(\bar{\lambda}_1) = \gamma_s - \sqrt{\bar{B}_{\text{II}}(\bar{\lambda}_1) - 3} \quad (\text{C.7})$$

392 Fixing γ_s successively, we plot the evolution domain of γ_t (see Figure C.3a) and compare it to the set that
393 can be scanned by the double shear test:

$$\gamma_t \in \left[\gamma_s \left(1 - \sqrt{1 + \frac{\gamma_s^2}{4}} \right), 0 \right]. \quad (\text{C.8})$$

394 Some values of the shear parameter γ_t are not accessible through the double shear test (see Figure C.3b).
395 Therefore, we present in the following Appendix D a more general motion which scan the whole domain of
396 variation for γ_t .

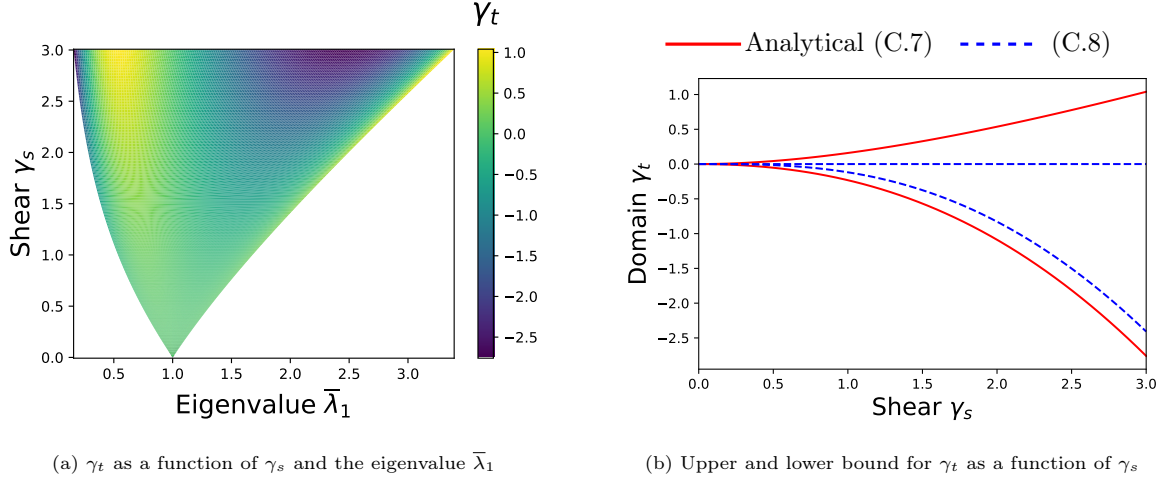


Figure C.3: Value of the second invariant γ_t for various γ_s

397 Appendix D. Double shear traction

398 We define as “isochoric double shear-traction test” the motion given by the following gradient:

$$\mathbf{F} = J^{\frac{1}{3}} \begin{pmatrix} \frac{1}{\sqrt{e}} & \gamma & 0 \\ 0 & \frac{1}{\sqrt{e}} & \gamma_{\perp} \\ 0 & 0 & e \end{pmatrix} \Rightarrow \bar{\mathbf{B}} = \begin{pmatrix} \frac{1}{e} + \gamma^2 & \frac{\gamma}{\sqrt{e}} & 0 \\ & \frac{1}{e} + \gamma_{\perp}^2 & \gamma_{\perp} e \\ Sym & & e^2 \end{pmatrix}. \quad (\text{D.1})$$

399 The “double shear test” mentioned in Section 3.1.4 corresponds to $e = 1$. We also denote as “simple tensile-
400 shear test” the case $\gamma_{\perp} = 0$.

401 The square of the Finger isochoric strain tensor is equal to:

$$\bar{\mathbf{B}}^2 = \begin{pmatrix} \left(\frac{1}{e} + \gamma^2\right)^2 + \frac{\gamma^2}{e} & \frac{\gamma}{\sqrt{e}} \left(\frac{2}{e} + \gamma^2 + \gamma_{\perp}^2\right) & \gamma \gamma_{\perp} \sqrt{e} \\ & \frac{\gamma^2}{e} + (\gamma_{\perp} e)^2 + \left(\frac{1}{e} + \gamma_{\perp}^2\right)^2 & \gamma_{\perp} e \left(\frac{1}{e} + \gamma_{\perp}^2 + e^2\right) \\ Sym & & (\gamma_{\perp} e)^2 + e^4 \end{pmatrix}. \quad (\text{D.2})$$

402 It’s worth noticing a non-zero \bar{B}_{13}^2 deformation which induces a purely non-linear stress. This out-of plane
403 shear stress is relevant for identifying the dependence of the free energy on the second invariant γ_t .

404 A simple computation of the characteristic polynomial of the isochoric Finger tensor shows that the
405 fundamental invariants are equal to:

$$\bar{B}_I = \gamma^2 + \gamma_{\perp}^2 + e^2 + \frac{2}{e} \text{ and } \bar{B}_{II} = \gamma^2 (\gamma_{\perp}^2 + e^2) + 2e + \frac{1}{e^2} + \frac{\gamma_{\perp}^2}{e^2}. \quad (\text{D.3})$$

406 The conservation of the parameter γ_s is ensured if at each instant, the following equality holds:

$$\bar{B}_I = \gamma^2 + \gamma_{\perp}^2 + e^2 + \frac{2}{e} = \gamma_s^2 + 3 \text{ with } e(0) = 1, \gamma(0) = \gamma_s, \gamma_{\perp}(0) = 0. \quad (\text{D.4})$$

407 The motion has two independent parameters: a shear γ_\perp and a traction magnitude e . The value of γ is
 408 adjusted so as to guarantee at each instant the equality (D.4).

409 We then deduce, by simple calculations, the value of the invariant γ_t :

$$\gamma_t(e, \gamma_\perp) = \gamma_s - \sqrt{(\gamma_\perp^2 + e^2) \left(\bar{B}_I - \gamma_\perp^2 - e^2 - \frac{2}{e} \right) + \frac{1}{e^2} + 2e + \frac{\gamma_\perp^2}{e} - 3}, \quad (\text{D.5})$$

410 with the necessary conditions of existence:

$$\bar{B}_I - \gamma_\perp^2 - e^2 - \frac{2}{e} \geq 0 \text{ and } e > 0. \quad (\text{D.6})$$

411 The existence conditions (D.6) define a sub-domain of \mathbb{R}^2 . At $\gamma_\perp \leq \gamma_s$ fixed, the range of variation of e
 412 is analytic and simply given by $e \in [e_2, e_0]$ with:

$$e_k = 2\sqrt{\tilde{\alpha}} \cos \left(\frac{1}{3} \arccos \left(-(\tilde{\alpha})^{-\frac{3}{2}} \right) + \frac{2k\pi}{3} \right) \text{ with } \tilde{\alpha} = \frac{\bar{B}_I - \gamma_\perp^2}{3} \geq 0. \quad (\text{D.7})$$

413 Hence, the values of the parameter γ_t that can be scanned through the simple shear tensile test ((D.9) with
 414 $\gamma_\perp = 0$) are:

$$\begin{aligned} \gamma_t(e, 0) &= \gamma_s - \sqrt{e^2 \left(\bar{B}_I - e^2 - \frac{2}{e} \right) + \frac{1}{e^2} + 2e} \\ \gamma_t(e, 0) &= \gamma_s - \sqrt{e^2 \bar{B}_I - e^4 + \frac{1}{e^2}} \end{aligned} \quad e \in [e_2(0), e_0(0)], \quad e_k(0) \text{ given by (D.7)}. \quad (\text{D.8})$$

415 Whereas for the double-shear test ((D.9) with $e = 1$):

$$\gamma_t(1, \gamma_\perp) = \gamma_s - \sqrt{(\gamma_\perp^2 + 1) (\bar{B}_I - \gamma_\perp^2 - 3) + \gamma_\perp^2} = \gamma_s - \sqrt{\gamma_s^2 + \gamma_\perp^2 (\gamma_s^2 - \gamma_\perp^2)}. \quad (\text{D.9})$$

416 We represent in figure D.4 the values of γ_t covered by the double shear tensile test with $\gamma_s = 3$. The red line
 417 corresponds to the single tensile-shear test ($\gamma_\perp = 0$), while the orange line corresponds to the double shear
 418 test ($e = 1$). The widest γ_t interval is scanned when $\gamma_\perp = 0$ (see D.4), i.e., during a single tensile-shear test.

419
 420 We further represent the γ_t that can be scanned by both the tensile-shear (Figure (D.5b)) and double
 421 shear (Figure D.5a) tests for various γ_s . We note that the double-shear test only explores negative values of
 422 γ_t while the tensile-shear test scans the whole domain (see Figure D.6 or more simply the equality between
 423 (C.7) and (D.8) as $e = \bar{\lambda}_1$).

424
 425 The identification of the constitutive law with respect to the second invariant γ_t Section 3.1.4 was built
 426 upon the double shear test. The reason is quite simple: the single shear tensile test, although scanning the
 427 whole domain for γ_t , does not allow to simply identify the dependence on γ_t because the non-linear stress σ_{13}
 428 vanishes. Therefore, measuring either σ_{12} or σ_{13} will couple $g^{(4)}$ to the other functions. The most efficient
 429 test, but also the hardest to perform experimentally, shall set a small shear strain γ_\perp and then perform a
 430 tensile test by varying e . Hence σ_{13} would be non-zero and one could characterize \tilde{f}_ψ for $\gamma_t \geq 0$.

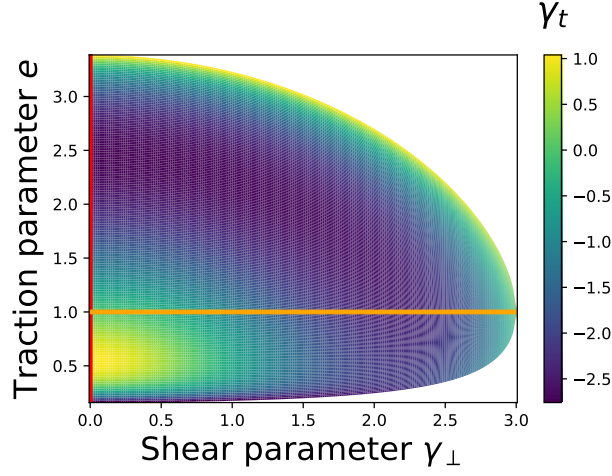


Figure D.4: Value of γ_t , $\gamma_s = 3$ fixed, for different values of e and γ_\perp

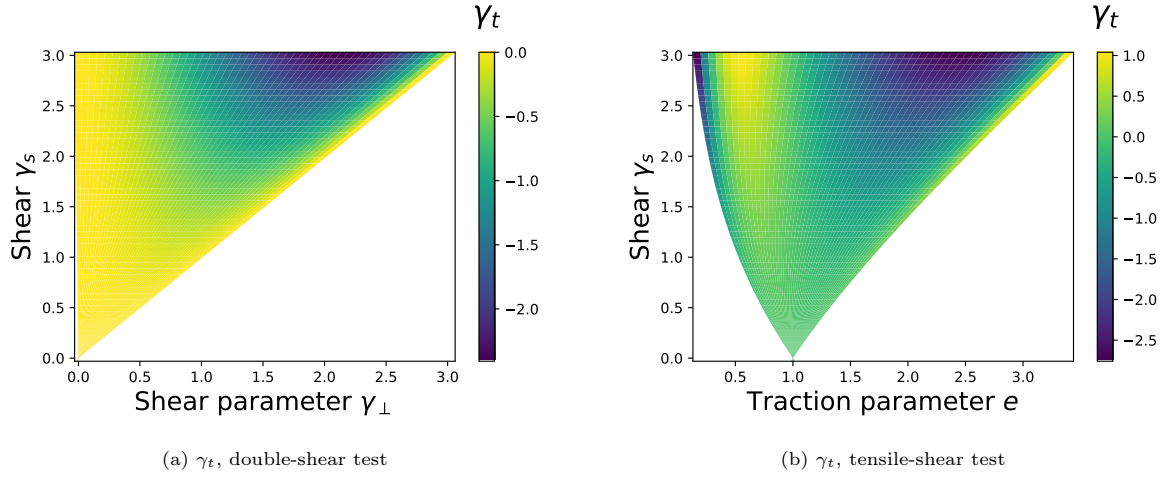


Figure D.5: γ_t for various γ_s

431 Appendix E. Generic partial derivatives of the internal energy

432 The partial derivative with respect to temperature is certainly the most cumbersome:

$$\begin{aligned}
 \frac{\partial e^m}{\partial T} &= \frac{dq_{exp}^{(5)}}{dT} - \frac{1}{\rho_0} \frac{dJ_{exp}^{(5)}(T)}{dT} T \partial_T p_{exp}^{(2)}(T, J_{exp}^{(5)}(T)) \\
 &\quad + \frac{1}{\rho_0} \int_1^J T \partial_T^2 p_{exp}^{(2)}(T, \tilde{J}) d\tilde{J} - \frac{1}{\rho_0} \int_0^{\gamma_s} T \partial_T^2 \hat{\tau}_{exp}^{(3)}(T, \tilde{\gamma}_s) d\tilde{\gamma}_s \\
 &\quad - \frac{1}{\rho_0} \int_0^{\gamma_t} \frac{\gamma_s - \tau}{\sqrt{(\gamma_s - \tau)^2 - \gamma_s^2}} T \partial_T^2 \hat{\tau}_{exp}^{(4)}(T, \gamma_s, \tau) d\tau.
 \end{aligned} \tag{E.1}$$

433 The partial derivative with respect to the volume expansion J thanks to the volumetric/isochoric split:

$$\frac{\partial e^m}{\partial J} = -\frac{1}{\rho_0} \left(p_{exp}^{(2)}(T, J) - T \partial_T p_{exp}^{(2)}(T, J) \right). \tag{E.2}$$

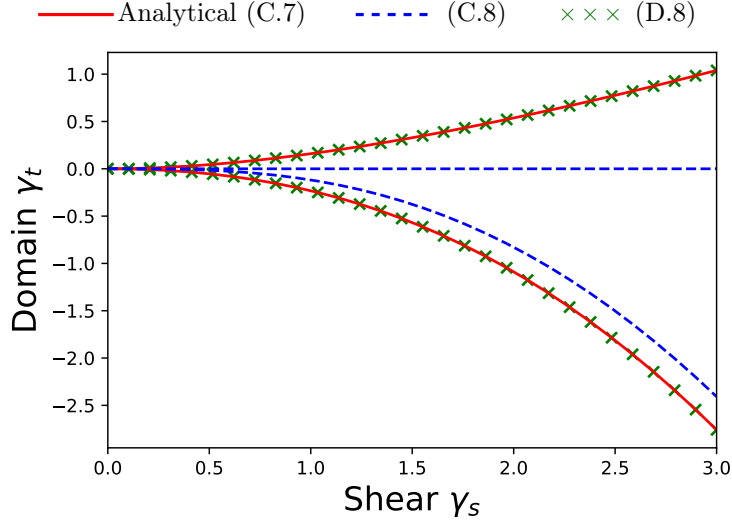


Figure D.6: Upper and lower bound of γ_t

434 The partial derivative with respect to γ_s and γ_t are written as:

$$\begin{aligned} \frac{\partial e^m}{\partial \gamma_s} &= \frac{1}{\rho_0} \left(\hat{\tau}_{exp}^{(3)}(T, \gamma_s) - T \partial_T \hat{\tau}_{exp}^{(3)}(T, \gamma_s) \right) + \frac{1}{\rho_0} \int_0^{\gamma_t} \partial_{\gamma_s} \left(\frac{\gamma_s - \tau}{\sqrt{(\gamma_s - \tau)^2 - \gamma_s^2}} \left(\hat{\tau}_{exp}^{(4)} - T \partial_T \hat{\tau}_{exp}^{(4)} \right) (T, \gamma_s, \tau) \right) d\tau, \\ \frac{\partial e^m}{\partial \gamma_t} &= \frac{1}{\rho_0} \frac{\gamma_s - \gamma_t}{\sqrt{(\gamma_s - \gamma_t)^2 - \gamma_s^2}} \left(\hat{\tau}_{exp}^{(4)} - T \partial_T \hat{\tau}_{exp}^{(4)} \right) (T, \gamma_s, \gamma_t). \end{aligned} \quad (E.3)$$

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