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RESTRICTIONS OF CONSTITUTIVE EQUATIONS FOR THERMODYNAMIC SYSTEMS WITH MEMORY FOLLOWING FROM THE IRREVERSIBILITY PRINCIPLE*

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Thermodynamic restrictions of the relaxation function matrix, including restrictions imposed on its antisymmetric part, have been obtained for abstract thermodynamic systems with memory. Examples of particular realizations of such systems are presented.

In constructing new phenomenological models of continua, it is very important to include thermodynamic restrictions. First of all, this concerns models of materials with memory [1-3], for which thermodynamics allows predictions in a very wide range. However, models of materials with memory are also of interest for the thermodynamic theory itself as an attractive object for application of its principles and concepts and their perfection by comparing predictions with experiment [3-9]. Of course, the theory should be developed to a level that would ensure experimentally verifiable results.

In [10, 11] the present authors suggested an effective method for investigating the properties of relaxation functions for media with memory following from the second law of thermodynamic theory based on the Clausius-Duhem inequality. In [12, 13] this method was extended to the case of a complete set of relaxation functions, including functions describing both main and cross effects, and corollaries of the second law, governing the interrelation of the cross effects, were obtained. However, the restrictions obtained there only concern the symmetric part of the matrix of relaxation functions.

In this article further development of this method is described and the results include restrictions of the antisymmetric part of the matrix of relaxation functions. Moreover, this method is extended to the case of abstract thermodynamic systems and many cases discussed earlier can be concrete realizations of them. In conclusion some examples of such realizations are given.

1. Notation and Definitions. Let R and R^+ be sets of real and real nonnegative numbers; S be the *configuration space*, which is a finite dimensional linear vector space of elements α, β, γ, ... with the inner product $<\cdot,\cdot>$ and the norm $|\cdot|$; L(S) be the vector space of all linear mappings A, B, C , ... of the space S into itself with the norm $|[\cdot]|$, where

$$|[A]| = \sup\{|A\alpha| : \alpha \in s, \quad |\alpha| = 1\}. \tag{1}$$

For any A∈L(S) the transformation in L(S) conjugate to it (or the transposed A) is denoted by $A^\times$ and defined by

$$\langle \alpha, A\beta \rangle = \langle \beta, A^\times\alpha \rangle \tag{2}$$

for all $\alpha, \beta \in S$.

The history f is a measurable function $f: R^+ \to S$ with the norm

$$\|f\| = \int_0^\infty |f(s)|^2 \xi(s)\, ds, \tag{3}$$

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where $\xi:R^+ \to R^+$ is a continuous monotonically decreasing *influence function* integrable on R^+.

The Hilbert space κ of all f with the finite norm (3) is called a *space of histories*. The history of a particular form, constant for all $s \geq 0$ and equal to an arbitrary $a \in S$, is denoted by α^+ and called a *constant history.*

The set $\varphi = S \times \kappa$ of pairs $\Lambda = (\alpha, f)$ with the norm $\| \Lambda \|_s = (|\alpha|^2 + \| f \|^2)^{1/2}$ will be called a *space of states.*

The two following functions of state will be assumed prescribed:

$\hat{\sigma}:\varphi \to S$ is a constitutive *functional of generalized forces*;

$\hat{h}:\varphi \to R$ is a constitutive *functional of a thermodynamic potential.*

Both functionals are assumed constant and Frechet differentiable in φ, and $\hat{h}$ is twice Frechet differenciable. This implies the existence of the partial derivatives for these functionals, for example, $D\hat{\sigma}:\varphi \to L(S)$ and $\delta\hat{\sigma}:S \to L(\kappa)$, defined for any $\Lambda \in \varphi$, $\beta \in S$, f and $g \in \kappa$ as follows:

$$D\hat{\sigma}(\Lambda)\beta = \frac{d}{d\lambda}\hat{\sigma}(\alpha + \lambda\beta,\ f)|_{\lambda=+0};\quad \delta\hat{\sigma}(\Lambda)(g) = \frac{d}{d\lambda}\hat{\sigma}(\alpha,\ f + \lambda g)|_{\lambda=+0}. \tag{4}$$

Let $C \subset S$ be a cone of admissible configurations in S. The function of time $\varepsilon:R \to C$, bounded with the piecewise continuous derivative, is called a *configurational trajectory of the system.*

At time t each configurational trajectory defines the *configurational history* $\varepsilon^t \in \kappa$ and the *state of the system* $\Lambda^t \in \kappa$ *at the moment* t as follows:

$$\varepsilon^t(s) = \varepsilon(t - s) - \varepsilon(t),\quad \Lambda^t(s) = \{\varepsilon(t),\ \varepsilon^t(\cdot)\}. \tag{5}$$

The state $\Lambda_\varepsilon^0 = \{\varepsilon, 0^+\}$, where 0^+ is the constant history equal to zero everywhere, called an *equilibrium state.* A set of states in the form (5) forms a *subset of admissible states* $\tilde{\varphi}$ in φ.

Using the configurational trajectory and the constitutive functionals, it is possible to define the *trajectory of generalized forces* $\sigma_\varepsilon:R \to S$ and the *trajectory of a thermodynamic potential* $h_\varepsilon:R \to R$ in the following way:

$$\sigma_\varepsilon(t) = \hat{\sigma}(\Lambda^t) = \hat{\sigma}(\varepsilon(t),\ \varepsilon^t(\cdot)),\quad h_\varepsilon(t) = \hat{h}(\Lambda^t) = \hat{h}(\varepsilon(t),\ \varepsilon^t(\cdot)). \tag{6}$$

The triplet of all three trajectories of the system will be called the *thermodynamic trajectory.*

2. Thermodynamic Theory. Thermodynamic consideration within the framework of this approach is based on the following postulate that expresses the second law of thermodynamics:

The thermodynamic postulate (TDP). For any thermodynamic trajectories the Clausius-Duhem inequality is fulfilled:

$$\langle \sigma_\varepsilon,\ \dot{\varepsilon} \rangle \geqslant \dot{h}_\varepsilon. \tag{7}$$

Here the dot superscripts are used to denote time derivatives.

A necessary and sufficient condition for satisfying this postulate is contained in the following theorem (see similar theorems for specific systems in [5]):

Theorem 1. *The TDP is fulfilled if the constitutional functionals satisfy the following relations:*

$$\hat{\sigma}(\Lambda^t) = D\hat{h}(\Lambda^t) - \tilde{D}\hat{h}(\Lambda^t), \tag{8}$$

$$\delta\hat{h}(\varepsilon^t,\ \varepsilon^t - \varepsilon(t)^+)(\dot{\varepsilon}^t) \leqslant 0, \tag{9}$$

where the derivative $\tilde{D}\hat{h}:\varphi \to S$ *is defined by the relation*

$$\langle \tilde{D}\hat{h}(\Lambda),\ \beta \rangle = \delta\hat{h}(\Lambda)(\beta^+) \tag{10}$$

for all $\beta \in S$, $\Lambda \in \varphi$.

Introducing the relaxation function $R:R^+ \to L(S)$, it is possible to use Riss' lemma on the representation of linear potentials to express the historically linear part of the Taylor series of the functional $\hat{\sigma}$ about a certain equilibrium state $\Lambda_{\varepsilon 0}^0 = \{\varepsilon_0, 0^+\} \in \varphi$ as follows:

$$\hat{\sigma}(\Lambda^t) = \hat{\sigma}(\Lambda^0_{\varepsilon_0}) + D\hat{\sigma}(\Lambda^0_{\varepsilon_0})(\varepsilon(t) - \varepsilon_0) + \delta\hat{\sigma}(\Lambda^0_{\varepsilon_0})(\varepsilon^t - \varepsilon(t)^+) + \\ + 0(\|\Lambda^t - \Lambda^0_{\varepsilon_0}\|) = \sigma_0 + E\varepsilon(t) + \int_0^\infty R(s)\varepsilon(t-s)\,ds + 0(\|\Lambda^t - \Lambda^0_{\varepsilon_0}\|), \quad (11)$$

where

$$\sigma_0 = \hat{\sigma}(\Lambda^0_{\varepsilon_0}) - D\hat{\sigma}(\Lambda^0_{\varepsilon_0})\varepsilon_0, \quad E = D\hat{\sigma}(\Lambda^0_{\varepsilon_0}) - \tilde{D}\hat{\sigma}(\Lambda^0_{\varepsilon_0})$$

and R is determined from the relations

$$\delta\hat{\sigma}(\Lambda^0_{\varepsilon_0})(f) = \int_0^\infty \frac{dR(s)}{ds} f(s)\,ds, \quad R(\infty) = 0 \quad (12)$$

for all $f \in \kappa$.

3. Thermodynamic Restrictions of the Relaxation Function. Subsequent investigation of the properties of the relaxation function R following from the requirement of satisfaction of theTDP is based on the following lemma, which is a corollary of Theorem 1. (Here we omit the proof of this lemma based on Taylor functional series expansion of the left-hand side of inequality (9) since it basically does not differ from the proof of similar results in [10, 11].)

L e m m a. *If the TDP is satisfied, the functional* $\hat{h}$ *has the following property: for any equilibrium state* Λ^0_ε *and any* $f \in \kappa$ *such that* $f' = df/ds \in \kappa$ *the inequality*

$$\langle (D\delta\hat{h}(\Lambda^0_\varepsilon)(f') - \tilde{D}\delta\hat{h}(\Lambda^0_\varepsilon)(f')),\ f(0)\rangle + \delta^2\hat{h}(\Lambda^0_\varepsilon)(f',\ f) \geqslant 0. \quad (13)$$

is satisfied.

Since the differential operators D, $\tilde{D}$ and δ commute with one another, relation (8) can be used to reduce inequality (13) to the form

$$\langle \delta\hat{\sigma}(\Lambda^0_\varepsilon)(f'),\ f(0)\rangle + \delta^2\hat{h}(\Lambda^0_\varepsilon)(f',\ f) \geqslant 0. \quad (14)$$

Let us consider two particular choices of elements f in κ, satisfying the hypothesis of the lemma:

$$f_1(s) = \alpha C_\omega(s) + \beta S_\omega(s), \quad (15)$$

$$f_2(s) = \alpha S_\omega(s) - \beta C_\omega(s), \quad (16)$$

where $S_\omega(s) = \sin(\omega s)$; $C_\omega(s) = \cos(\omega s)$, $\omega \geq 0$.

Since f_1, $f_1' \in \kappa$ and f_2, $f_2' \in \kappa$, they can be substituted into (14) to obtain the following inequalities:

$$\langle \delta\hat{\sigma}(\Lambda^0_\varepsilon)(-\omega\alpha S_\omega + \omega\beta C_\omega),\ \alpha\rangle + \delta^2\hat{h}(\Lambda^0_\varepsilon)((-\omega\alpha S_\omega + \omega\beta C_\omega), \\ (\alpha C_\omega + \beta S_\omega)) \geqslant 0, \quad (17)$$

$$\langle \delta\hat{\sigma}(\Lambda^0_\varepsilon)(\omega\alpha C_\omega + \omega\beta S_\omega),\ -\beta\rangle + \delta^2\hat{h}(\Lambda^0_\varepsilon)((\omega\alpha C_\omega + \omega\beta S_\omega), \\ (\alpha S_\omega - \beta C_\omega)) \geqslant 0. \quad (18)$$

Since for the fixed state Λ^0_ε the second-order Frechet derivative is a bilinear functional symmetric on $\kappa \times \kappa$ (i.e., $\delta^2\hat{h}(\Lambda^0_\varepsilon)(f_1, f_2) = \delta^2\hat{h}(\Lambda^0_\varepsilon)(f_2, f_1)$ for any f_1, $f_2 \in \kappa$ [14]), it can easily be shown that the terms with $\delta^2\hat{h}$ in the inequalities (17) and (18) are equal but have opposite signs. Therefore, in summing these inequalities the mentioned terms are cancelled and as a result we have

$$\langle \delta\hat{\sigma}(\Lambda^0_\varepsilon)(-\omega\alpha S_\omega + \omega\beta C_\omega),\ \alpha\rangle - \langle \delta\hat{\sigma}(\Lambda^0_\varepsilon)(\omega\alpha C_\omega + \omega\beta S_\omega),\ \beta\rangle \geqslant 0. \quad (19)$$

Substitution of the terms into (19) in terms of the relaxation function R according to (12) and division of this inequality by ω ($\omega \geq 0$) give

$$-\left\langle\left(\int_0^\infty R'(s)\,\alpha\sin\omega s\,ds\right),\ \alpha\right\rangle + \left\langle\left(\int_0^\infty R'(s)\,\beta\cos\omega s\,ds\right),\ \alpha\right\rangle -$$

$$-\left\langle\left(\int_0^\infty R'(s)\,\alpha\cos\omega s\,ds\right),\ \beta\right\rangle-\left\langle\left(\int_0^\infty R'(s)\,\beta\sin\omega s\,ds\right),\ \beta\right\rangle\geqslant 0. \tag{20}$$

Integrating by parts here, using (2), and denoting the Fourier cosine and sine transforms of the function R by $\overline{R}_c(\omega)$ and $\overline{R}_s(\omega)$, respectively, reduces the inequality (20) to the form

$$\omega\left[\langle \overline{R}_c(\omega)\alpha,\ \alpha\rangle+\langle \overline{R}_c(\omega)\beta,\ \beta\rangle+\langle(\overline{R}_c(\omega)-\overline{R}_s^{\times}(\omega))\beta,\ \alpha\rangle\right]+ \\ +\langle(R(0)-R^{\times}(0))\beta,\ \alpha\rangle\geqslant 0. \tag{21}$$

Letting $\omega\to 0$, we obtain

$$\langle(R(0)-R^{\times}(0))\beta,\ \alpha\rangle\geqslant 0, \tag{22}$$

hence it follows (because α and β are arbitrary) that

$$R(0)-R^{\times}(0)=0. \tag{23}$$

With the help of this relation inequality (21) reduces to the form

$$\langle \overline{R}_c(\omega)\alpha,\ \alpha\rangle+\langle \overline{R}_c(\omega)\beta,\ \beta\rangle+\langle(\overline{R}_c(\omega)-\overline{R}_s^{\times}(\omega))\beta,\ \alpha\rangle\geqslant 0. \tag{24}$$

In a similar way it can be shown that not only are relation (23) and inequality (24) corollaries of (21), but vice versa, inequality (21) follows from (23) and (24).

In this way we have proved the following

Theorem 2. *In order that the TDP be satisfied, it is necessary that the relaxation function R in (12) satisfy conditions (23) and (24) for any $\omega \geq 0$ and $\alpha, \beta \in S$*

A new element in this result is the fact that conditions (23) and (24) contain restrictions of the antisymmetric part of the relaxation function.

4. Applications of the General Theory. One of the possible concrete realizations of the general theory can be obtained if the "mechanistic" terminology used here is understood in a nearly direct sense, i.e., if it is assumed that ε is the strain gradient tensor, σ is the Piola-Kirchhoff stress tensor, h is the Helmholtz specific free energy, and R is the stress relaxation function. Here S is the space of second-order tensors, $C\subset S$ is the subspace of tensors ε satisfying the condition $\det\varepsilon \geq 0$, L(S) is the space of fourth-order tensors. Then the inequality (7) is a particular case of the ordinary Clausius-Duhem inequality, expressing the requirement of nonnegativity of entropy production, and relations (23) and (24) reflect properties of the stress relaxation function following from the second law.

In a similar way, from the general theory it is possible to obtain a model of media with thermal and deformational memories, discussed in Sect. 3 of [13]. This model is a modification of Chen-Gurtin's model [15]. The modification consists in using the inverse absolute temperature and its gradient as independent variables and in introducing the corresponding thermodynamic potential and is necessary for construction of a thermodynamically well-posed linear theory [16]. In this case $\varepsilon = \{\vartheta, F, \overline{g}\}$, where ϑ is the inverse absolute temperature; g is the inverse temperature gradient, and

$$\overline{g}(t)=\int_{-\infty}^{t} g(s)\,ds.$$

Then, $S = R\times L(E^3)\times E^3$, where E^3 is the three-dimensional Euclidean space; $L(E^3)$ is the vector space of second-order tensors. The inner product of the elements $\alpha_1 = \{\lambda_1, H_1, f_1\}$ and $\alpha_2 = \{\lambda_2, H_2, f_2\}$ in S is introduced naturally:

$$\langle\alpha_1,\ \alpha_2\rangle=\lambda_1\lambda_2+\mathrm{tr}\,(H_1H_2)+f_1\cdot f_2.$$

Then, in this case C is a set of the elements $\{e, \vartheta S, q/\rho\}$, where e is the specific internal energy; S is the Piola-Kirchhoff stress tensor; q is the heat flux; ρ is the medium density. Here h is the following thermodynamic potential: $h = e\vartheta - \eta$, where η is the specific entropy. The standard requirement of nonnegative entropy production in the form of the Clausius-Duhem inequality reduces to the form (7) in the present notation. In this case the relaxation function is a matrix of, generally speaking, tensor-valued functions, composed of relaxation functions describing all the main and cross effects, and the properties of these functions, including the interrelation of cross effects, are contained in Theorem 2. The first two terms in inequality (24), following from this theorem, are quadratic forms and only depend on the symmetric part of the matrix of relaxation functions, while the last term depends on the antisymmetric part of this matrix, imposing thermodynamic restrictions on it.

One more particular realization of the general theory is obtained if S is a Cartesian product of three-dimensional Euclidean spaces: $S = E^3 \times E^3$ and ε is a pair of three-dimensional vectors {**E**, **H**}, where **E** and **H** are the magnetic and electric intensities. Then, σ = {**D**, **H**}, where **D** and **B** are the electric and magnetic inductions and h is the free enthalpy defined as in [17]. In this interpretation equality (7) is equivalent to the Clausius-Duhem inequality (4.4) in [16], and relations (23) and (24) express properties of the material equations of the electrodynamics of continua with time variance, following from the thermodynamic restrictions, including the requirement of a definite interrelation of cross effects. As a particular case, they contain the properties obtained in [16], and, moreover, they contain additional restrictions concerning the antisymmetric part of the matrix of relaxation functions.

In conclusion, we note that relation (23), which expresses the property of symmetry of the matrix of instantaneous responses, is a certain analogy of the Onsager reciprocal relation in ordinary nonequilibrium thermodynamics [18]; however, unlike the latter, it is obtained here as a corollary of the requirement of nonnegativity of entropy production (the irreversibility principle) and does not need a special postulate.

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