CONCERNING THE FORMULATION OF THE IRREVERSIBILITY PRINCIPLE IN LOCALLY NONEQUILIBRIUM THERMODYNAMICS

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It is shown that in the existing locally nonequilibrium thermodynamic theory there are a number of paradoxes that can be interpreted as an indication that the used formulation of the second law of thermodynamics is insufficiently complete. An additional thermodynamic postulate allowing one to remove these paradoxes and having close analogies in other thermodynamic approaches is suggested.

Keywords: *thermodynamics, second law, locally nonequilibrium systems, heat conduction with memory, electromagnetic field, additional thermodynamic postulate.*

In one of his last works devoted to the most urgent problems of heat and mass transfer [1] Aleksei Vasilievich Luikov placed particular emphasis upon the importance and potential for the then new trend of research called "nonlinear thermomechanics." This term is understood to refer to the region of investigations in which, based on new approaches of continuum mechanics and locally nonequilibrium thermodynamics, it was possible to construct thermodynamically consistent models of media with complicated properties such as media with memory, media with internal variables of state, micropolar media, etc. Approximately at that time he pioneered research work in that area at the Heat and Mass Transfer Institute. After the demise of A. V. Luikov, investigations along these lines were continued under the guidance of V. L. Kolpashchikov, by A. A. Baranov, N. A. Migun, S. Yu. Yanovskii, V. T. Borukhov, and by the author of the present paper, in which some recent results of research in this field are given.

In the present article, the problem on the necessity of refining and supplementing the formulation of the second law of thermodynamics in the so-called entropy-free form is investigated. Earlier we have ventured a paradox that there may exist increasing temperature waves in the linear theory of heat conduction with memory that can be interpreted as an indication that the existing formulation of the second law does not exclude physically inadmissible situations [2]. Below it is shown that there is an analogous paradox also for electromagnetic waves in equilibrium dielectrics with memory. To overcome these difficulties of the theory, an additional thermodynamic postulate is suggested which is formulated as the requirement of the satisfiability of the Clausius property (see [3]) relative to a certain additional thermodynamic action. One further motivation of this postulate is that the additional thermodynamic potential, whose existence follows from this postulate, is in many respects analogous to the dissipative potential, whereas the postulate itself is analogous to the universal evolution criterion introduced by Prigogine with coauthors [4]. In the concluding part of the article it is shown that the indicated additional postulate makes it possible to prove the impossibility of the existence of increasing electromagnetic waves in equilibrium dielectrics with memory, as well as excluding the conterexample showing the possibility of increasing temperature waves.

1. Unificaton of Representation of Various Thermodynamic Systems. It will be shown in this section in which way various specific thermodynamic systems can be represented universally and, consequently, embedded in a certain unified formal scheme. Based on this scheme, a generalized abstract thermodynamic theory is then constructed.

The two most widely used (in contemporary thermodynamics) formulations of the second law date back to two classical formulations of the nineteenth century:

a) in each cyclic process the reduced heat integral is nonpositive, i.e.,

$$\oint \frac{dQ}{\theta} \leq 0 \, ; \tag{1.1}$$

715

b) for any process the following inequality holds:

$$\int \frac{dQ}{\theta} \le S_2 - S_1 . \tag{1.2}$$

The thermodynamic approach based on the contemporary variant of the formulation of (1.1) belonging to Clausius is usually called entropy-free. If the thermodynamic system is a continuum, then the reduced heat integral can be represented in a local field formulation, and then inequalities (1.1) and (1.2) are reduced to the following form [5–9]:

$$\oint \left[-\frac{1}{\rho} \operatorname{div}\left[\frac{\mathbf{q}}{\theta}\right] + \frac{r}{\theta} \right] dt \le 0 , \tag{1.3}$$

$$\eta\,(t_2) - \eta\,(t_1) \ge \int_{t_1}^{t_2} \left[-\frac{1}{\rho} \operatorname{div}\left(\frac{\mathbf{q}}{\theta}\right) + \frac{r}{\theta} \right] dt . \tag{1.4}$$

The last inequality can be presented in differential form (the Clausius–Duhem inequality):

$$\dot{\eta}\,(t) \ge -\frac{1}{\rho} \operatorname{div}\left[\frac{\mathbf{q}}{\theta}\right] + \frac{r}{\theta} . \tag{1.5}$$

From here on the dot above the symbol denotes the substantive time derivative.

We will consider several examples of thermodynamic systems for which the first law of thermodynamics (the energy conservation law) is represented as follows:

a) a nondeformable heat-conducting body

$$\dot{e} = -\frac{1}{\rho} \operatorname{div} \mathbf{q} + r ; \tag{1.6}$$

b) a deformable heat-conducting body

$$\dot{e} = -\frac{1}{\rho} \operatorname{div} \mathbf{q} + \operatorname{tr}(\mathbf{S}\dot{\mathbf{F}}) + r ; \tag{1.7}$$

c) a nondeformable nonconducting dielectric body with an electromagnetic field [5]:

$$\dot{e} = \frac{1}{\rho} \dot{\mathbf{D}} \cdot \mathbf{E} + \frac{1}{\rho} \dot{\mathbf{B}} \cdot \mathbf{H} + r . \tag{1.8}$$

We will define r, e.g., from (1.7) and substitute into (1.3). As a result, having introduced the inverse absolute temperature $\vartheta = 1/\theta$ and its gradient in spatial variable $\mathbf{g} = \operatorname{grad} \vartheta$, we obtain

$$\oint \left(\dot{e}\vartheta - \vartheta \operatorname{tr}(\mathbf{S}\dot{\mathbf{F}}) - \frac{1}{\rho} \mathbf{q}\cdot\mathbf{g} \right) dt \le 0 , \tag{1.9}$$

$$\dot{e}\vartheta - \vartheta \operatorname{tr}(\mathbf{S}\dot{\mathbf{F}}) - \frac{1}{\rho} \mathbf{q}\cdot\mathbf{g} \le \dot{\eta} . \tag{1.10}$$

Introducing, into (1.10), a new thermodynamic potential Φ:

$$\Phi = e\vartheta - \eta , \tag{1.11}$$

and adding to (1.9) the identity

$$\oint (e^{\cdot}\vartheta)\, dt = 0\,, \tag{1.12}$$

we represent (1.9) and (1.10) in the form

$$\oint \left(e\dot{\vartheta} + \vartheta\, \mathrm{tr}\, (\mathbf{S}\dot{\mathbf{F}}) + \frac{1}{\rho}\, \mathbf{q}\cdot\mathbf{g} \right) dt \geq 0\,; \tag{1.13}$$

$$-\dot{\Phi} + e\dot{\vartheta} + \vartheta\, \mathrm{tr}\, (\mathbf{S}\dot{\mathbf{F}}) + \frac{1}{\rho}\, \mathbf{q}\cdot\mathbf{g} \geq 0\,. \tag{1.14}$$

We will consider the space $\mathbf{S} = \mathscr{D}(\mathscr{E}) \times \mathscr{E} \times \mathscr{R}$. From its definition it follows that the elements of $\mathbf{S}$ are the triplets $\gamma = \{\mathbf{T}, \mathbf{a}, \lambda\}$ consisting of a tensor, vector, and scalar. If $\gamma_1 = \{\mathbf{T}_1, \mathbf{a}_1, \lambda_1\}$ and $\gamma_2 = \{\mathbf{T}_2, \mathbf{a}_2, \lambda_2\}$ are two elements of $\mathbf{S}$, then their scalar product and norm $|\cdot|$ in $\mathbf{S}$ will be defined as follows:

$$\langle \gamma_1, \gamma_2 \rangle = \mathrm{tr}\, (\mathbf{T}_1\mathbf{T}_2) + \mathbf{a}_1\cdot\mathbf{a}_2 + \lambda_1\lambda_2\,, \quad |\gamma| = \langle \gamma, \gamma \rangle^{1/2}\,. \tag{1.15}$$

Then, if for each material point we introduce the time function

$$\sigma = \left\{ \vartheta\mathbf{S}, \frac{1}{\rho}\, \mathbf{q}, e \right\} \quad \text{и} \quad \varepsilon = \{\mathbf{F}, \overline{\mathbf{g}}, \vartheta\}\,, \tag{1.16}$$

where

$$\overline{\mathbf{g}}\,(t) = \int\limits_{-\infty}^{t} \mathbf{g}\,(s)\, ds\,, \tag{1.17}$$

then inequalities (1.13) and (1.14) can be written in the form

$$\oint \langle \sigma, \dot{\varepsilon} \rangle\, dt \geq 0\,, \tag{1.18}$$

$$\langle \sigma, \dot{\varepsilon} \rangle \geq \dot{\Phi}\,. \tag{1.19}$$

In the same way, with the aid of analogous transformations of the thermodynamic systems introduced above under items a) and c) (see (1.6), (1.8)), both formulations of the second law can be presented in the form of (1.18) and (1.19), where

$$\text{a)}\ \mathbf{S} = \mathscr{E} \times \mathscr{R}\,, \quad \sigma = \left\{ \frac{1}{\rho}\, \mathbf{q}, e \right\}, \quad \varepsilon = \{\overline{\mathbf{g}}, \vartheta\}\,; \tag{1.20}$$

$$\text{b)}\ \mathbf{S} = \mathscr{E} \times \mathscr{E} \times \mathscr{R}\,, \quad \sigma = \left\{ \frac{\vartheta}{\rho}\, \mathbf{B}, \frac{\vartheta}{\rho}\, \mathbf{D}, e \right\}, \quad \varepsilon = \{\mathbf{H}, \mathbf{E}, \vartheta\}\,, \tag{1.21}$$

In the last case, the thermodynamic potential is

$$\Phi = e\vartheta - \eta - \vartheta\mathbf{B}\cdot\mathbf{H} - \vartheta\mathbf{D}\cdot\mathbf{E}\,. \tag{1.22}$$

The possibility of such a unified representation of various thermodynamic systems is used as the basis for the formalism employed in what follows. Thus, corresponding to the below-introduced notions of configurational space, configurational trajectory, and trajectory of generalized stress are the above-defined space $\mathbf{S}$, time function $\varepsilon(t)$, and time

function $\sigma(t)$ in each of the three considered specific cases, respectively. At the same time, this formalism can describe a great number of other physical systems.

2. Thermodynamic Theory. Any thermodynamic theory of continua of the type of the above-considered examples, in order to be closed, must be supplemented, in addition to the indicated relations and conservation laws, by the so-called constitutive or material equations that assign the way in which dependent variables are defined in terms of independent ones and thus define the medium properties. In the thermodynamics of irreversible processes [4] these equations are assigned by linear relations between current values of thermodynamic fluxes and forces, whereas the function of state for entropy on the basis of the principle of local equilibrium for nonequilibrium states is taken the same as for equilibrium ones. The thermodynamic approach accepted here does not use the principle of local equilibrium and, as is seen from what follows, the entropy (or any other thermodynamic potential expressed in its terms) does not coincide in equilibrium and nonequilibrium states.

In all of the foregoing cases independent variables are grouped into the variable ε (generalized configuration), whereas dependent variables with account for the normalization conditioned by the form of the reduced heat integral are grouped into the variable σ (generalized stress).

The most universal form of constitutive equations follows from the assumption that the values of dependent variables at the current time instant are defined not only by the values of independent variables at the current instant, but also by their prehistory (the causality principle). This is the so-called model of media with memory, which is the most general means of assigning locally nonequilibrium systems [6, 7]. In this case, since the concrete dependences on prehistories represent functionals of the type of convolutions, then by integration by parts they can be represented in terms of the dependence of their time derivatives on prehistories (differential histories). For reasons of convenience and compactness of basic relations we represent this dependence precisely in terms of differential histories.

In a generalized form the thermodynamic system is defined by a constitutive equation that assigns the generalized stress σ in terms of the trajectories of generalized configuration $\varepsilon(t)$:

$$\sigma(t) = \hat{\sigma}(\varepsilon(t), \dot{\varepsilon}^{t}), \tag{2.1}$$

where $\dot{\varepsilon}^{t}(\cdot)$ is the differential history of generalized configuration defined as $\dot{\varepsilon}^{t}(s) = \dfrac{d\varepsilon(t-s)}{dt}$. Here σ and ε are vectors from the abstract vector configurational space **S** with the scalar product $\langle\cdot,\cdot\rangle$ (1.15) and $\hat{\sigma}$ is an ordinary function of the first argument and a functional of the second. If on a set of differential histories $\mathcal{H}$ we assign the norm $\|\|$, for example, as $\|\dot{\varepsilon}^{t}\| = \int\limits_{0}^{\infty} \dot{\varepsilon}^{t}(s)\chi(s)ds$ (here $\chi(s)$ is the integrable positive monotonically decreasing function of the influence), then for the functional $\hat{\sigma}$ we can define the notions of continuity, differentiability, and of Frechet derivatives [6, 7] over the normalized space $\mathcal{H}$.

Thus, for example, for case a) from the above-listed examples (see (1.6), (1.20)) the constitutive equations of the form of (2.1) will be written as

$$\mathbf{q}(t) = \hat{\mathbf{q}}(\vartheta(t), \dot{\overline{\mathbf{g}}}^{t}, \dot{\vartheta}^{t}), \quad e(t) = \hat{e}(\vartheta(t), \dot{\overline{\mathbf{g}}}^{t}, \dot{\vartheta}^{t}), \tag{2.2}$$

where $\dot{\vartheta}^{t}(s) = \dfrac{d}{dt}\vartheta(t-s)$ is the differential history of the inverse temperature and the remaining histories were determined analogously. In (2.2), $\hat{\mathbf{q}}$ and $\hat{e}$ represent ordinary function of the first argument and functionals of two latter ones. These relations describe the model of heat conduction with memory that possesses hyperbolic properties of the propagation of heat — the model suggested by Gurtin and Pipkin in [8].

Analogous relations can be cited for the remaining two examples. For the constitutive equations of the form of (2.2) in the below-expounded theory of thermodynamic systems with memory there corresponds the constitutive functional (more precisely, vector-functional) of generalized stress (2.1).

We will consider the generalized theory of thermodynamic systems with memory set out in [3, 9] in a somewhat less formalized treatment.

718

In the standard entropy-free formulation ([9] postulate **P1**) in a less formal form the second law can be presented as follows:

P1. For any $\kappa > 0$ there can be such $\sigma > 0$ that for all the trajectories $\varepsilon(\cdot)$ and time instants t_1 and t_2 such that $[|\varepsilon(t_2) - \varepsilon(t_1)|^2 + \|\dot{\varepsilon}^{t_2} - \dot{\varepsilon}^{t_1}\|]^{1/2} \leq \delta$ the following inequality holds:

$$\int_{t_1}^{t_2} \langle \hat{\sigma}(\varepsilon(t), \dot{\varepsilon}^t), \dot{\varepsilon}(t) \rangle \, dt \geq -\kappa . \tag{2.3}$$

This postulate is a certain generalization of formulation (1.18) needed because of the fact that for media with memory there exist very little strictly closed processes; therefore, speaking informally, it is required that for approximately closed processes the reduced heat integral (thermodynamic action) could be approximately nonnegative.

From this postulate follows, as the prerequisite condition of its satisfiability [3, 9], the existence of the thermodynamic potential $\hat{\psi}(\varepsilon(t), \dot{\varepsilon}^t)$ such that

$$\int_{t_1}^{t_2} \langle \hat{\sigma}(\varepsilon(t), \dot{\varepsilon}^t), \dot{\varepsilon} \rangle \, dt \geq \hat{\psi}(\varepsilon(t_2), \dot{\varepsilon}^{t_2}) - \hat{\psi}(\varepsilon(t_1), \dot{\varepsilon}^{t_1}) . \tag{2.4}$$

If $\hat{\psi}$ is differetiable, then (2.4) can be written in a differential form as

$$\langle \sigma, \dot{\varepsilon} \rangle \geq \dot{\hat{\psi}} , \tag{2.5}$$

which proves the equivalence of the formulations of the second law in entropy and entropy-free forms (1.18), (1.19).

Let us define the equilibrium function of the generalized stress as

$$\sigma^0(\varepsilon) := \hat{\sigma}(\varepsilon, 0^+) , \tag{2.6}$$

where 0^+ is the zero differential history ($0^+(s) = 0_{\mathbf{S}}$ for $s \in [0, \infty)$, $0_{\mathbf{S}}$ is the zero element of $\mathbf{S}$).

We will consider the corollaries following from this postulate for the case of semi-linear constitutive equations, i.e., Eq. (4.1) of the form

$$\sigma(t) = \sigma^0(\varepsilon(t)) + \int_0^{\infty} \mathbf{R}(s) \, \dot{\varepsilon}^t(s) \, ds , \tag{2.7}$$

where $\mathbf{R}(\cdot)$ is the relaxation function determined on $[0, \infty)$ whose values are the linear operators from $\mathbf{S}$ into $\mathbf{S}$.

Note that for Eq. (2.7) the equilibrium function of the generalized stress (2.6) is represented by the first term on the right-hand side of (2.7), which is reflected in the notation.

For constitutive equations of the form of (2.7) the necessary and sufficient conditions for the satisfiability of the postulate **P1** are, as shown in [9–11], the following:

a) there exists a scalar function $\varphi(\varepsilon)$ such that

$$\sigma^0(\varepsilon) = \partial_\varepsilon \varphi(\varepsilon) ; \tag{2.8}$$

b) the relaxation function $\mathbf{R}$ satisfies the inequality

$$\langle [\overline{\mathbf{R}}(\omega) + \overline{\mathbf{R}}^{\times}(-\omega)] a, a^* \rangle \geq 0 , \quad \forall a \in \mathscr{C} , \quad \omega \geq 0 , \tag{2.9}$$

where $\overline{\mathbf{R}}(\omega)$ is the Fourier transformation of the function $\mathbf{R}$, $\mathscr{C}$ is a set of complex numbers, a^* means a complex-conjugate number of a, and the superscript $\times$ denotes transposition.

Note that the limitations imposed on the function $\mathbf{R}$ by condition (2.9) are equivalent to the so-called condition of real positiveness in [12]. The corollary of inequality (2.9) is the following property of the function $\mathbf{R}$ (here the inequality should be understood in the sense of the nonnegative definiteness of the operator):

$$\mathbf{R}(0) \geq 0 . \tag{2.10}$$

For the scalar relaxation function, condition (2.9) is reduced to the requirement

$$\overline{R}_c(\omega) \geq 0 , \quad \forall \omega \geq 0 , \tag{2.11}$$

where $\overline{\mathbf{R}}_c(\omega)$ is the Fourier cosine transformation of R.

3. Paradoxes of the Standard Thermodynamic Theory of the Thermal Conductivity of Solid Bodies with Memory. We will consider a specific physical system, viz., a nondeformable heat-conducting body with memory (case a) of the three cases considered in Sections 1 and 2. The mathematical model of this system is based on the energy balance equations (1.6) and constitutive equations for a heat-conducting medium with memory (2.2).

We recall that since

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

then, according to the definition, the differential history of this variable has the form

$$\dot{\overline{\mathbf{g}}}^{t}(s) = \mathbf{g}(t-s) . \tag{3.1}$$

For the linear and isotropic case, Eqs. (2.2) subject to (3.1) are reduced [8, 10] to

$$e(t) = e_0 - c\vartheta(t) + \int_0^{\infty} \beta(s) \, \dot{\vartheta}(t-s) \, ds , \qquad \mathbf{q} = \int_0^{\infty} \alpha(s) \, \mathbf{g}(t-s) \, ds , \tag{3.2}$$

where β and α are the relaxation functions of the internal energy and of the heat flux, respectively.

The necessary and sufficient conditions (2.11) for the satisfiability of the second law in the form of postulate **P1** for this system can be written in the form

$$\overline{\alpha}_c(\omega) := \int_0^{\infty} \alpha(s) \cos(\omega s) \, ds \geq 0 , \quad \overline{\beta}_c(\omega) := \int_0^{\infty} \beta(s) \cos(\omega s) \, ds \geq 0 , \quad \text{for all} \ \ \omega \geq 0 . \tag{3.3}$$

Substituting constitutive equations (3.3) into energy balance equation (1.6), we obtain the following equation of heat conduction with memory:

$$c\dot{\vartheta}(\mathbf{x}, t) - \int_0^{\infty} \beta(s) \, \ddot{\vartheta}(\mathbf{x}, t-s) \, ds = \int_0^{\infty} \alpha(s) \, \nabla^2 \vartheta(\mathbf{x}, t-s) \, ds . \tag{3.4}$$

It should be noted in passing that due to specially selected independent variables (the inverse absolute temperature and its gradient), this linear equation does not contradict the thermodynamic relations, as it does in the case of the Gurtin–Pipkin original theory [8].

We will seek the solution of Eq. (3.4) in the form of a plane harmonic attenuating wave:

$$\vartheta(\mathbf{x}, t) = \vartheta_0 + A \exp(i(\omega t + k(\mathbf{x} - \mathbf{x}_0) \cdot \mathbf{n}) - \xi(\mathbf{x} - \mathbf{x}_0) \cdot \mathbf{n}) . \tag{3.5}$$

It is shown in [2] that such a solution satisfies Eq. (3.4) if the wave attenuation coefficient is expressed as follows:

$$\xi\left(\omega\right) = \frac{B\left(\omega\right)}{\left(c - \omega\overline{\beta}_s\left(\omega\right)\right)\overline{\alpha}_c\left(\omega\right) + \omega\overline{\beta}_c\left(\omega\right)\overline{\alpha}_s\left(\omega\right)}\,, \tag{3.6}$$

where $\overline{\alpha}_s$ and $\overline{\beta}_s$ are the sine-transformations of functions α and β defined by Eq. (3.3), and $B(\omega) \geq 0$ is the function also expressed in terms of α and β. Thus, the attenuation sign is determined by the sign of the denominator in (3.6). In [2], it was established that there exists a pair of relaxation functions α and β that satisfies the conditions of thermodynamic admissibility (3.3), and at the same time leads to the existence of increasing temperature waves (solutions of Eq. (3.4)) at some frequencies. This is the following pair of functions:

$$\beta\left(s\right) = \beta_0 \exp\left(-s/\tau\right)\,, \quad \alpha\left(s\right) = \frac{\alpha_0 T^2}{s^2}\left[\sin^2\left(\frac{s}{T}\right) + \sin^2\left(\frac{2s}{T}\right) - 2\sin^2\left(\frac{3s}{2T}\right)\right]\,, \tag{3.7}$$

where $\beta_0 > 0$; $\alpha_0 > 0$; T, and $\tau > 0$ are constants. It can be shown that restrictions (3.3) for these functions are fulfilled and at the same time at the frequency $\omega_0 = 1/T$ (as well as in a certain vicinity of it, in view of the continuous dependence of ξ on frequency) the relaxation functions (3.7) lead to a negative expression (3.6), which means an increasing wave solution of Eq. (3.4) at these frequencies. If we take into account the natural viewpoint that increasing wave solutions must be thermodynamically forbidden, then from this fact it follows that the formulation of the second law must contain some additional restrictions. Below, one other example that illustrates the urgency of the problem of additional thermodynamic restrictions will be given.

4. Concerning Increasing Electromagnetic Waves in Dielectrics. We will consider a thermodynamic system with memory, which is a nonconducting dielectric in the presence of an electromagnetic field. The general theory of such systems was developed in [5], and it was shown in the previous sections in which way it can be embedded in the considered generalized thermodynamic formalism. It is based on the energy balance equation (1.9), which accounts for the terms that describe the contribution of the electromagnetic field to the internal energy; the second law is formulated in the form of the Clausius–Duhem inequality, and the constitutive equations for it in the form of Eq. (2.1), when expounded in detail, will have the form

$$e\left(t\right) = \hat{e}\left(\vartheta\left(t\right), \mathbf{E}\left(t\right), \mathbf{H}\left(t\right), \dot{\vartheta}^t\,\dot{\mathbf{E}}^t, \dot{\mathbf{H}}^t\right)\,, \quad \mathbf{D}\left(t\right) = \hat{\mathbf{D}}\left(\vartheta\left(t\right), \mathbf{E}\left(t\right), \mathbf{H}\left(t\right), \dot{\vartheta}^t\,\dot{\mathbf{E}}^t, \dot{\mathbf{H}}^t\right)\,,$$

$$\mathbf{B}\left(t\right) = \hat{\mathbf{B}}\left(\vartheta\left(t\right), \mathbf{E}\left(t\right), \mathbf{H}\left(t\right), \dot{\vartheta}^t\,\dot{\mathbf{E}}^t, \dot{\mathbf{H}}^t\right)\,, \tag{4.1}$$

Here, just as before, a circumflex ($\hat{}$) above the symbol designates constitutive functionals, and the differential histories are defined analogously to the corresponding notions in the previous sections.

For a linear isotropic case, in the absence of cross effects, the constitutive equations (4.1) are reduced to the form [5, 13]

$$e\left(t\right) = e_0 - c\vartheta\left(t\right) + \int_0^\infty \beta\left(s\right)\dot{\vartheta}\left(t - s\right)ds\,, \quad \mathbf{D}\left(t\right) = \zeta_0\mathbf{E} + \int_0^\infty \zeta\left(s\right)\dot{\mathbf{E}}\left(t - s\right)ds\,,$$

$$\mathbf{B}\left(t\right) = \mu_0\mathbf{H} + \int_0^\infty \mu\left(s\right)\dot{\mathbf{H}}\left(t - s\right)ds\,. \tag{4.2}$$

The necessary and sufficient conditions (2.11) of thermodynamic admissibility of relaxation functions in (4.2) (in the form of the postulate **P1** [9–11, 13]) have the form

$$\overline{\alpha}_c\left(\omega\right) \geq 0\,, \quad \overline{\zeta}_c\left(\omega\right) \geq 0\,, \quad \overline{\mu}_c\left(\omega\right) \geq 0\,, \tag{4.3}$$

where for any function $R(\cdot)$: $\mathbf{R}^+ \to \mathbf{R}$, $\overline{R}_c(\omega)$ means

$$\overline{R}_c(\omega) := \int_0^\infty R(s) \cos \omega s \, ds \,. \tag{4.4}$$

We turn now to the Maxwell equations:

$$\text{rot } \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \,, \quad \text{div } \mathbf{D} = 0 \,; \quad \text{rot } \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} \,, \quad \text{div } \mathbf{B} = 0 \,. \tag{4.5}$$

We supplement this system with two constitutive equations (4.2), which will be written in the following compact form:

$$\mathbf{D} = \zeta_0 \mathbf{E} + \zeta^* \mathbf{E} \,, \quad \mathbf{B} = \mu_0 \mathbf{H} + \mu^* \mathbf{H} \,, \tag{4.6}$$

where for the convolution of the form of $\int_0^\infty \gamma(s)\mathbf{E}(t-s)ds$ the notation $\gamma^*\mathbf{E}$ is used. Equations (4.5) and (4.6) form a closed system that can be reduced by standard transformations to one equation for one of the characteristics of the electromagnetic field, say, $\mathbf{E}$. This equation has the form

$$\Delta \mathbf{E} + \frac{1}{\zeta_0} \zeta^* (\text{grad} (\text{div } \mathbf{E})) = (\zeta_0 + \zeta^*)(\mu_0 + \mu^*) \frac{\partial \mathbf{E}}{\partial t^2} \,. \tag{4.7}$$

We will seek the solution of this equation in the form of a plane attenuating transverse harmonic electromagnetic wave with the attenuation coefficient ξ, wave vector $k\mathbf{n}$, and amplitude $\mathbf{E}_0$ ($|\mathbf{n}| = 1$, $\mathbf{E}_0 \cdot \mathbf{n} = 0$, since the wave is transverse):

$$\mathbf{E}(x, t) = \mathbf{E}_0 \exp(i\omega(t - \mathbf{n} \cdot (\mathbf{x} - \mathbf{x}_0))) \exp(-\xi \mathbf{n} \cdot (\mathbf{x} - \mathbf{x}_0)) \,, \tag{4.8}$$

where

$$\omega \,, \quad k \geq 0 \,. \tag{4.9}$$

The second term on the left-hand side of Eq. (4.7) for transverse waves disappears, since in this case div $\mathbf{E}$ = 0. Substituting (4.8) into (4.7), we obtain

$$(ik + \xi)^2 = -\omega^2 (\zeta_0 + \overline{\xi}_c(\omega) - i\overline{\zeta}_s(\omega))(\mu_0 + \overline{\mu}_c(\omega) - i\overline{\mu}_s(\omega)) \,, \tag{4.10}$$

where $\overline{\mu}_c$, $\overline{\mu}_s$, $\overline{\zeta}_c$, and $\overline{\zeta}_s$ are the Fourier cosine- and sine-transformations of the functions ζ and μ defined similarly to (4.4).

Separating in (4.10) the real and imaginary parts, we obtain expressions for ξ:

$$\xi = \frac{\omega^2}{2k}\left[\overline{\mu}_s(\omega)(\zeta_0 + \overline{\zeta}_c(\omega)) + \overline{\zeta}_s(\omega)(\mu_0 + \mu_c(\omega))\right] \,, \tag{4.11}$$

whence it is seen that in view of (4.9) the wave attenuation sign is determined by the expression within the squared brackets in (4.11).

Just as in Section 3, we will consider a pair of relaxation functions ζ and μ that are analogous to (3.7) and that satisfy the necessary and sufficient conditions of thermodynamic admissibility (4.3):

$$\zeta(s) = \varepsilon_a \exp(-s/\tau) \,, \quad \mu(s) = \mu_a \frac{T^2}{s^2}\left[\sin^2\left(\frac{s}{T}\right) + \sin^2\left(\frac{2s}{T}\right) - 2\sin^2\left(\frac{3s}{2T}\right)\right] \,. \tag{4.12}$$

Conditions (4.3) can be easily checked using the Fourier transformation tables [14]. Equation (4.3) should be supplemented with the inequalities

722

$$\zeta_0 \geq 0, \quad \mu_0 \geq 0, \tag{4.13}$$

which are usually derived in equilibrium thermodynamics from the stability conditions.

Since the thermodynamics does not impose any couplings between μ_0 and μ or between ζ_0 and ζ, we can select constants in (4.12) that satisfy the condition

$$\mu_a > \frac{1}{2} \frac{\zeta_a \mu_0 \tau}{\zeta_0 + \zeta_a c_1 T}, \tag{4.14}$$

where

$$c_1 = \frac{1}{4} \ln(3) + \frac{1}{4} \ln(15) - \frac{1}{2} \ln(8) + \text{arctanh}\left(\frac{1}{2}\right) + 2\, \text{arctanh}\left(\frac{1}{2}\right) = 3\, \text{arctanh}\left(\frac{1}{2}\right) = -0.068. \tag{4.15}$$

Calculating, with the aid of the Fourier transformation tables [14], the attenuation coefficient (4.11) for functions (4.12) at the frequency $\omega_0 = \frac{1}{T}$, we find

$$\xi = \frac{1}{2kT^2}\left(-\mu_a(\zeta_0 + \zeta_a T c_1) + \frac{1}{2}\tau\zeta_a\mu_0\right). \tag{4.16}$$

Taking into account (4.14) and (4.9), we conclude that $\xi < 0$. This means that at the given frequency (and in its vicinity) increasing electromagnetic waves propagate. Such a situation, just as in the case with temperature waves, seems to be physically unreal, since here we are dealing with the wave propagating in an initially equilibrium medium. Moreover, in contrast to the case of the media excited out of equilibrium (that enhance electromagnetic radiation), here there is no increase in the electromagnetic energy flux, and we are dealing only with the increase in the wave amplitude. The possibility of an increase of electromagnetic waves in an equilibrium medium at some frequencies would have meant electromagnetic instability of an equilibrium state, since part of the spectrum of equilibrium thermal radiation would have started to increase.

This paradox is another argument stimulating the search for additional thermodynamic postulates allowing one to exclude nonphysical situations.

5. On Additional Thermodynamic Restrictions. If we accept the natural viewpoint that the paradoxes described in the previous sections must be forbidden thermodynamically, then it follows from such results that a standard set of thermodynamic restrictions is insufficient. Consequently, it becomes urgent to seek additional thermodynamic postulates ensuring new restrictions of constitutive equations to exclude nonphysical situations.

In this Section we consider a possible variant of such a postulate making it possible to advance in the solution of the above-considered problems.

We will require that the considered thermodynamic system, apart from the postulate **P1**, satisfy an additional postulate formulated as follows:

P2. For any $\kappa < 0$ one can find $\delta > 0$ such that for all trajectories $\varepsilon(\cdot)$ and time moments t_1, t_2 such that

$$\left[\left|\varepsilon(t_2) - \varepsilon(t_1)\right|^2 + \left\|\dot{\varepsilon}^{t_2} - \dot{\varepsilon}^{t_1}\right\|^2\right]^{1/2} \leq \delta, \tag{5.1}$$

the following inequality holds:

$$\int_{t_1}^{t_2} \left\langle \frac{d}{dt}[\hat{\sigma}(\varepsilon(t), \dot{\varepsilon}^t) - \sigma^0(\varepsilon(t))], \dot{\varepsilon}(t) \right\rangle dt \geq -\kappa. \tag{5.2}$$

To study the corollaries following from this postulate, we will restrict ourselves, as above, to consideration of the case of semi-linear constitutive equation, i.e., equation of the form of (2.7).

We will consider what additional restrictions are introduced by postulate **P2**. Substituting (2.7) into (5.2), we have

$$\int_{t_1}^{t_2} \left\langle \int_0^\infty \mathbf{R}(s)\,\ddot{\boldsymbol{\varepsilon}}^{\,t}(s)\,ds,\,\dot{\boldsymbol{\varepsilon}}(t) \right\rangle dt \ge -\kappa\,. \tag{5.3}$$

With the aid of integration by parts, (5.3) can be represented as

$$\int_{t_1}^{t_2} \left\langle \left(\mathbf{R}(0\,\dot{\boldsymbol{\varepsilon}}(t) + \int_0^\infty \mathbf{R}'(s)\,\dot{\boldsymbol{\varepsilon}}^{\,t}(s)\,ds \right),\,\dot{\boldsymbol{\varepsilon}}(t) \right\rangle dt > -\kappa\,, \tag{5.4}$$

which corresponds to inequality (5.2) in postulate **P2** for the case considered (we recall that the prime here means derivative with respect to *s*, and a dot above a symbol means derivative with respect to *t*).

We can prove the following necessary and efficient condition of the satisfiability of postulate **P2** for semilinear systems.

Theorem 1. *In order that postulate* **P2** *be fulfilled for systems given by constitutive equation (2.7), it is necessary and sufficient that for any restricted integrable functions u:* $\Re \to \mathbf{S}$ *with a carrier limited on the left, the function* $\mathbf{R}(\cdot)$ *satisfy the inequality*

$$\int_{-\infty}^{t} \langle \mathbf{R}(0)\,u(\tau),\,u(\tau)\rangle\,d\tau + \int_{-\infty}^{t}\int_{-\infty}^{\tau} \langle \mathbf{R}'u(\tau-s)\,u(s),\,u(\tau)\rangle\,ds\,d\tau \ge 0\,. \tag{5.5}$$

The proof of this theorem turns out to be an obvious modification of the proof of the basic theorem in [11] with account for properties (2.10) satisfied by virtue of the postulate **P1**.

As is seen from the results of [11, 12], property (5.5) is equivalent to

$$\langle [\overline{\mathbf{R}}'(\omega) + \overline{\mathbf{R}}'^{\times}(-\omega) + \mathbf{R}(0)]\,a,\,a^*\rangle \ge 0\,, \quad \forall a \in \mathscr{C}\,, \quad \omega \ge 0\,. \tag{5.6}$$

Here $\overline{\mathbf{R}}'(\omega)$ means the Fourier-transformation of $\mathbf{R}'(s)$, $\overline{\mathbf{R}}'(\omega) = \overline{\mathbf{R}}'_c(\omega) + i\overline{\mathbf{R}}'_s(\omega)$, where $\overline{\mathbf{R}}'_c(\omega)$ and $\overline{\mathbf{R}}'_s(\omega)$ are the cosine- and sine-Fourier transformations of this function, respectively.

For the case of scalar functions **R** (5.6) is reduced to

$$\overline{R}'_c(\omega) + R(0) \ge 0\,, \quad \forall \omega \ge 0\,. \tag{5.7}$$

With the aid of integration by parts, the latter inequality can be reduced to the form

$$\overline{R}_s \ge 0\,. \tag{5.8}$$

The next assertion demonstrates that the postulates **P1** and **P2** are not contradictory, i.e., there exist objects satisfying both postulates. Such objects are represented, for example, by relaxation thermodynamic systems. We recall that a thermodynamic system with memory is called a relaxation one (analogously to a dynamic system (see [12])) if its relaxation function has the form

$$\mathbf{R}(s) = \sum_{i=1}^{n} \mathbf{R}_i \exp(\lambda_i s)\,, \tag{5.9}$$

where $\lambda_n < \lambda_{n-1} < \ldots < \lambda_1 \le 0$, $\mathbf{R}_i = \mathbf{R}_i^{\times} = \mathbf{R}_i > 0$, $\forall i = 1, 2, \ldots, n$.

Assertion 1. *Semilinear relaxation thermodynamic systems* with σ^0, *determined according to (5.10), satisfy the postulates* **P1** *and* **P2**.

This assertion is proved by direct checking with the aid of Fourier transformation tables [14] and of inequalities (2.11) and (5.7) for a relaxation function of the form of (5.9).

The obtained additional restrictions following from postulate **P2**, together with the standard restrictions of postulate **P1**, allow one, as will be seen below, to advance in the solution of the problems of increasing waves. This is one of the justifications of the new postulate. A further justification is that the given assumption has, as we will show now, direct analogs within the framework of the thermodynamics of irreversible processes developed by I. Prigogine's school [4].

Note that from postulate **P2**, analogously to the case of **P1**, there follows the existence of thermodynamic potential Φ such that, by analogy with (5.5), the following inequality holds:

$$\langle \dot\sigma, \dot\varepsilon \rangle - \langle \dot\sigma^0, \dot\varepsilon \rangle \geq \dot\Phi . \tag{5.10}$$

Since the potentials ψ and Φ are determined by the same thermodynamic system, it is evident that there must exist a coupling between them. We will establish it for the case of the so-called central potential for relaxation systems (5.9), which in concrete applications corresponds to the standard thermodynamic potential.

According to relation (3.24) from [15] (see also [16]), the central thermodynamic potential ψ in our case is expressed as follows:

$$\psi(\varepsilon(t), \dot\varepsilon(t)) = \varphi(\varepsilon(t)) + \frac{1}{2} \int_0^\infty \int_0^\infty \langle \mathbf{R}(\tau+s)\, \dot\varepsilon^t(\tau), \dot\varepsilon^t(s) \rangle \, d\tau ds . \tag{5.11}$$

The central potential in the family of potentials Φ is constructed analogously:

$$\Phi(\dot\varepsilon^t) = \frac{1}{2} \int_0^\infty \int_0^\infty \langle \mathbf{R}'(\tau+s)\, \dot\varepsilon^t(\tau), \dot\varepsilon^t(s) \rangle \, d\tau ds . \tag{5.12}$$

Based on expression (5.11) and inequality (5.5), we now calculate the production Σ of the potential ψ (which in concrete applications is usually the entropy production). Since the production of a potential is a source that supplements inequality (2.5) up to a balance, then Σ is defined as

$$\Sigma := \langle \sigma, \dot\varepsilon^t \rangle - \dot\psi = \left\langle \left[\sigma^0(\varepsilon(t)) + \int_0^\infty \mathbf{R}(s)\, \dot\varepsilon(t-s)\, ds \right], \dot\varepsilon(t) \right\rangle$$

$$- \frac{d}{dt}\left[\varphi(\varepsilon(t)) + \int_0^\infty \int_0^\infty \langle \mathbf{R}(\tau+s)\, \dot\varepsilon^t(\tau), \dot\varepsilon^t(s) \rangle \, d\tau ds \right]$$

$$= \langle \sigma^0(\varepsilon(t)), \dot\varepsilon \rangle + \left\langle \int_0^\infty \mathbf{R}(s)\, \dot\varepsilon(t-s)\, ds, \dot\varepsilon(t) \right\rangle - \langle \partial_\varepsilon \varphi(\varepsilon(t)), \dot\varepsilon(t) \rangle$$

$$- \frac{1}{2} \int_0^\infty \int_0^\infty \langle \mathbf{R}(\tau+s)\, \ddot\varepsilon^t(\tau), \dot\varepsilon^t(s) \rangle \, d\tau ds - \frac{1}{2} \int_0^\infty \int_0^\infty \langle \mathbf{R}(\tau+s)\, \dot\varepsilon^t(\tau), \ddot\varepsilon^t(s) \rangle \, d\tau ds . \tag{5.13}$$

Taking into account relation (5.10) on the right-hand side of (5.13), using the symmetry of the function R (see (5.9)), and integrating by parts, we reduce (5.13) to the form

$$\Sigma = \frac{1}{2} \int\limits_{0}^{\infty} \int\limits_{0}^{\infty} \langle \mathbf{R}^{'}(\tau + s)\, \dot{\varepsilon}^{t}(\tau),\, \dot{\varepsilon}^{t}(s) \rangle \, dt ds \,. \tag{5.14}$$

Comparing (5.12) and (5.14), we come to the conclusion that the additional thermodynamic potential, whose existence follows from the supplementary postulate **P2**, is nothing else but the production of the classical thermodynamic potential (for typical cases this is the entropy production). This result correlates well with the ideas developed by I. Prigogine with co-authors in the works where he introduces the so-called dissipation potential [4].

6. Corollaries from the Supplementary Thermodynamic Postulate. We turn again to the thermodynamic system considered in Section 4. According to (4.11), the attenuation of harmonic electromagnetic waves in such a system is expressed as

$$\xi(\omega) = \frac{\omega^2}{2k} \left[\bar{\mu}_{\mathrm{s}}(\omega)\,(\zeta_0 + \bar{\zeta}_{\mathrm{c}}(\omega)) + \bar{\zeta}_{\mathrm{s}}(\omega)\,(\mu_0 + \mu_{\mathrm{c}}(\omega)) \right]. \tag{6.1}$$

The restrictions following from postulate **P1** (4.13) for relaxation functions are

$$\bar{\zeta}_{\mathrm{c}}(\omega) \geq 0 \,, \quad \bar{\mu}_{\mathrm{c}}(\omega) \geq 0 \,. \tag{6.2}$$

To this, we add the inequalities that are substantiated in equilibrium thermodynamics from the requirement of stability of the equilibrium state:

$$\mu_0 \geq 0 \,, \quad \zeta_0 \geq 0 \,. \tag{6.3}$$

If we require the fulfillment of postulate **P2** relative to this system, we will obtain the following additional restrictions of relation functions (compare with (6.2)):

$$\bar{\zeta}_{\mathrm{s}}(\omega) \geq 0 \,, \quad \bar{\mu}_{\mathrm{s}}(\omega) \geq 0 \,. \tag{6.4}$$

Allowing for inequalities (6.2)–(6.4) in relation (6.1), we obtain the inequality

$$\xi(\omega) \geq 0 \,. \tag{6.5}$$

Consequently, the following assertion 2 has been proved.

Assertion 2. *If a thermodynamic system representing a medium given by constitutive equations (4.2) satisfies postulates **P1** and **P2**, then the increasing electromagnetic waves are forbidden in an equilibrium medium.*

Thus, postulate **P2**, in addition to the arguments given at the end of the previous section, can be motivated also by the fact that one can also eliminate the paradox of increasing electromagnetic waves. This postulate also excludes the counterexample leading to increasing temperature waves considered in Section 3, since it can be easily checked [14] that the relaxation function $\alpha(s)$ in (3.7) does not satisfy the requirement (5.8) following from **P2**.

Conclusions. Thus, the results of the present work allow us to make the conclusion that in the modern thermodynamic theory of locally nonequilibrium systems there are difficulties connected with the fact that it admits the existence of increasing temperature and electromagnetic waves. At the same time, these difficulties can be overcome if along with the well-known thermodynamic postulate on the property of having a fixed sign for the heat integral reduced by the inverse absolute temperature, in any cyclic process of thermodynamic action one postulates the requirement of a fixed sign in any cyclic process of a new thermodynamic action. The physical sense of this new thermodynamic action can be interpreted as the difference of integrals of heat reduced by the rate of change of the inverse temperature in nonequilibrium and equilibrium processes with an identical trajectory. The new postulate also entails the existence of a new thermodynamic potential in many respects analogous to the Prigogine dissipation potential.

NOTATION

A, ϑ_0, amplitude and reference temperature of a thermal wave; a, arbitrary complex number; $\mathbf{a}_1$, $\mathbf{a}_2$, arbitrary vectors; $\mathscr{C}$, set of complex numbers; c_1, constant defined in (4.15); $\mathbf{D}$, $\mathbf{B}$, electric and magnetic induction, respectively; $\mathbf{E}$, $\mathbf{H}$, strength of electric and magnetic field, respectively; $\mathbf{E}_0$, amplitude of electromagnetic wave; $\mathscr{E}$, three-dimensional Euclidean vector space; e, specific internal energy; e_0, reference value of internal energy; $\mathbf{F}$, tensor of strain gradient; $\mathbf{g}$, gradient of inverse temperature; $\bar{\mathbf{g}}$, integral gradient of inverse temperature; $\mathscr{H}$, set of differential histories; k, wave number of thermal and electromagnetic waves; $\mathscr{D}(\mathscr{E})$, space of three-dimensional tensors of second rank; $\mathbf{n}$, unit vector; Q, quantity of heat received by the system by the current moment; $\mathbf{q}$, heat flux vector; $\hat{\mathbf{q}}$, $\hat{e}$, constitutive functionals of the heat flux and internal energy; r, internal heat release per unit mass; $\mathbf{R}$, generalized relaxation function; R, scalar relaxation function; $\mathscr{R}$, set of real numbers; S_1, S_2, entropy of the system in the initial and final state; $\mathbf{S}$, tensor of the Piola–Kirchhoff stress tensor; $\mathbf{S}$, configurational space; s, moments related to the past relative to the current moment; $\mathbf{T}_1$, $\mathbf{T}_2$, arbitrary tensors; t, t_1, t_2, arbitrary time instants; $\mathbf{x}_0$, fixed point; α, β, relaxation functions of internal energy and of heat flux, respectively; $\bar{\alpha}_s$, $\bar{\beta}_s$, sine-transformations of functions α and β; α_0, β_0, T, and τ, constants determining relaxation functions (3.7); γ, γ_1, γ_2, arbitrary elements from $\mathbf{S}$; δ, κ, scalar parameters; ε, generalized configuration; η, specific entropy; θ, absolute temperature; ϑ, inverse absolute temperature; ϑ^t, differential history of inverse temperature; λ_i, $\mathbf{R}_i$, parameters determining the relaxation function (5.9); μ_0, ζ_0, equilibrium magnetic and electric permeabilities, respectively, with μ and ζ being the relaxation functions expressing the relaxation part of permeabilities; $\bar{\mu}_c$, $\bar{\mu}_s$, $\bar{\zeta}_c$, $\bar{\zeta}_s$, cosine- and sine Fourier transforms of functions μ and ζ; ξ, attenuation of thermal and electromagnetic waves; ρ, density of a medium; ζ_a, μ_a, T, τ, constants determining relaxation functions (4.12); σ, generalized stress; σ^0, equilibrium generalized stress; χ, influence function; ψ, Φ, thermodynamic potentials; $\hat{\psi}$, constitutive functional for thermodynamic potential ψ; ω, frequency of thermal and electromagnetic waves; ω_0, characteristic frequency. Subscripts and superscripts: 0, characteristic value; 1, initial state; 2, final state; t, history up to moment t; a, amplitude value.

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