Generalization of Onsager's reciprocal relations

V. A. Etkin

Integrative Research Institute, Geula 39, Haifa 33197, Israel

E-mail address: v_a_etkin@bezeqint.net

ABSTRACT

Shows, that it is possible to expand the existing theory of irreversible processes to processes of useful transformation of any forms of energy by a finding of motive forces and the speeds of real processes as derivative of energy, but not entropy; that for such processes there are differential reciprocal relations with Onsager's symmetry and Cazimir's anti-symmetry conditions resulting from them; that Maxwell's equations follows from these correlation, confirming their equity for a wide class of processes, satisfying to these equations

Keywords: non-equilibrium thermodynamics; generalization; differential reciprocal relations; substantiation; verification; consequences

1. INTRODUCTION

The rise of engineering interest in phenomena at the interface between various disciplines and perception of their close relation with the phenomena of the energy dissipation led to creating, in the early 20th century, the thermodynamic theory of real process rate (TIP). This field in macroscopic physics of the 20th century was named the theory of irreversible processes [1…10]. It has enriched the theoretical mind of the 20th century with a number of general physics principles and notably contributed to cognition of the in-depth interrelations between different-type phenomena. Its contribution was appreciated by two Nobel prizes awarded in the field (L.Onsager, 1968; I. Prigogine, 1977).

But this theory in its present state does not consider the processes of useful energy transformation of heat into work, that makes the fundamental principles of all thermodynamics. This situation is caused to a great extent by the reason that basic operational values of this theory – thermodynamic forces X_i and fluxes J_i – are determined with the help of the entropy production expression that does not take into consideration reversible component of real processes power and consequently it can not measure their intensity. As a result of it an vast region of processes with relative efficiency above zero fall out of TIP competency.

Meantime kinetics of those processes interests not only power engineering and energy technology for which energy transformation and are basic TIP-method are the most acceptable in principle. The thermodynamic investigation of biological systems is also impossible without useful work against equilibrium supporting the non-equilibrium state of such systems and providing their vital activity. The application of thermodynamics to cosmological objects that develop, according to current ideas, bypassing equilibrium would also be incomplete without work considered as ordered form of exchange.

All that begets a problem of enlarging TIP to include the systems performing some useful work. In this article consistently thermodynamic decision of this problem is offered.

2. THE LIMITATIONS OF THE EXISTING THEORY OF IRREVERSIBLE PROCESSES

More than centenary had passed before it became clear that "thermodynamics unaware of time" (to a Brian's figurative locution) was substantially thermostatics wherein only Fourier's, Navier's, Ohm's, Fick's, Darcy's, Newton's, etc. equations prefigured the coming theory of non-static processes. However, the development in that direction demanded introducing in thermodynamics the transfer concepts intrinsically extraneous for it. One of these belongs to N. Umov (1873), who wrote the law of conservation of energy in terms of the energy flow across the stationary borders of the system [11]. The flow concept in application to entropy (G. Jaumann, 1911) became another stride [12]. Note, that application was quite novel, because of the statistic-mechanical interpretation of entropy as a measure of state probability for which the transfer concept is absolutely senseless. A little bit later De Donder (1927) related the entropy source with the rate of the chemical reactions with its affinity [13]. That was how the concepts of flow and process rate started their introduction into thermodynamics.

The most decisive move in that direction was not, however, made, until 1931 (L. Onsager) [1]. Onsager built his theory of physicochemical process rate based on the expression for the entropy generation rate, having thus emphasized the irreversible part of real phenomena. The entropy S of a closed adiabatic isolated system in equilibrium state is known to be maximal. If the parameters $\Theta_1, \Theta_2, \ldots, \Theta_n$ differ from their equilibrium values by a value of $\alpha_1, \alpha_2, \ldots, \alpha_n$, it is naturally to assume that the difference between the entropies of the current

S and equilibrium S_o states ΔS is a some function of α_i. In this case the reason of the ith scalar process generation (the scalar thermodynamic force X_i) and the generalized rate of relaxation process (named by Onsager the flow J_i) could be found from the expression for the entropy generation rate:

$$dS/dt = \Sigma_i(\partial S/\partial \alpha_i)\, d\alpha_i/dt = \Sigma_i X_i J_i , \qquad (1)$$

where $X_i = (\partial S/\partial \alpha_i)$; $J_i = d\alpha_i/dt$.

Thus L. Onsager endued the force X_i with a meaning quite different from that in Newton's mechanics and construed it as a parameter measuring the deviation of a system from internal equilibrium. At the same time L. Onsager postulated that for minor deviation from thermodynamic equilibrium any of the flows J_i obeyed the law of linear dependence on all the thermodynamic forces X_j active in the system:

$$J_i = \Sigma_j L_{ij} X_j . \quad (i, j = 1, 2, \ldots, n) . \qquad (2)$$

Onsager referred to those laws of relaxation processes, as well as the associated coefficients L_{ij}, as "phenomenological" (i.e. practice-based). Such a (matrix) form of kinetic equations differed from Fourier's, Ohm's, Fick's, Darcy's, etc. laws by the presence of additional terms (with subscripts $i \neq j$). The off-diagonal summands in expression (2) were introduced by Onsager to allow for various "superposition" (interrelation) effects of different-type irreversible processes running simultaneously in the same spatial zones.

The proof of reciprocity relationships between the "off-diagonal" phenomenological coefficients L_{ij} and L_{ji} $(i \neq j)$ was most important in the Onsager's theory:

$$L_{ij} = L_{ji} . \qquad (3)$$

These symmetry conditions are called the reciprocal relations. They reduce the number of the coefficients L_{ij} to be experimentally defined from n (for mere empirical description) down to $n(n+1)/2$ (where n – a number of independent flows) and lead to setting up a before unknown relationship between the rates of different-type irreversible processes. Onsager obtained those relationships based on a known statement of statistical mechanics regarding the reversibility of micro-processes in time under the assumption that the coefficients L_{ij} and L_{ji} were constant, while the subscript-dissimilar flows J_i and J_j were linearly independent and became zero with disappearance of the forces X_i and X_j. These three statements outstep the framework of thermodynamics. Therefore he named it "quasi-thermodynamics".

Onsager was afterward awarded the Nobel Prize (1968) for his studies in that field. Those studies attributed to non-equilibrium thermodynamics just as much as the R. Clausius' studies to the making of classic thermodynamics. They have embodied the odds and ends of concepts and facts representing them in an accessible and understandable form. Their

publication made a good start to the intensive development of the TIP in macroscopic and statistical physics. H. Cazimir (1945) extended the Onsager's theory to cover vector processes [2], having herein proved that in case the α– and β–type forces (even and odd time functions) acted simultaneously, the reciprocity relationships (6) would go over into the anti–symmetry conditions:

$$L_{ij} = - L_{ji} \tag{4}$$

A little bit later (1956-62) I. Prigogine based on Curie's symmetry law showed that in case the scalar and vector processes ran simultaneously, only the processes of the same (or even) tensor range could interact (superimpose) [3]. That allowed further solving a number of problems associated with the evolution of non-equilibrium systems.

The interest in that field of knowledge was caused not only by its general theoretical significance, but rather its important applications having been outlined in the forties and fifties and having involved the thermal diffusion isotope separation, the allowance for additional terms in the hydrodynamic equations for missile art and plasma physics, the development of membrane technique, biophysics, etc. The development of the phenomenological and statistical TIP has advanced the approximation of the heat-mass transfer theory to hydrodynamics, electrodynamics and continuum mechanics. It appeared to have been especially useful to study phenomena at interfaces between those disciplines.

However, those theories never did eliminate the abovementioned line of demarcation between thermodynamics and the heat transfer theory. The reason is that the TIP is restricted to studying the dissipation processes like heat conductivity, electric conductivity, diffusion, as well as effects of their superposition, but does not deal with the processes of useful energy conversion, which are the main object of investigation in thermodynamics. This is the reason why the necessity appeared to call thermodynamics into being as a unified theory of energy transfer and conversion rate and capacity, which, unlike W. Thomson's "pseudo-thermostatics" [13] and L. Onsager's "quasi-thermodynamics" [1], would not "un-file" reversible or irreversible part of real processes, but would cover the entire of its spectrum. For this purpose it is necessary to pass to a finding of forces X_i, X_j and streams J_i, J_j on more general basis supposing both increase and decrease of energy of system [14].

3. GENERALIZATION OF NON-EQUILIBRIUM THERMODYNAMICS

It is known, that in thermodynamics full energy is subdivided on internal U and external E. The first depends on internal variables Θ_i (entropy S, volume V, mass of kth substances M_k, charge 3, etc.),. i.e. $U = U (\Theta_1, \Theta_2, \ldots, \Theta_n)$. The second depends on position of system as the whole concerning an environment, i.e. from radius-vector **r** of the center of these values, i.e. E

$=E(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_m)$. It means, that full energy of system as function of its condition looks like $\Im = \Im(\Theta_i, \mathbf{r}_j)$, and they exact differential be expressed by the following identity [15]:

$$d\Im \equiv \Sigma_i \psi_i \, d\Theta_i - \Sigma_j \mathbf{F}_j \cdot d\mathbf{r}_j, \quad (i = 1,2,\ldots,n; j = 1,2,\ldots,m) \tag{4}$$

where $\psi_i \equiv (\partial\Im/\partial\Theta_i)$ – the generalized potentials such absolute temperature T and absolute pressure p, chemical, electric, etc. potential; $\mathbf{F}_i \equiv -(\partial\Im/\partial\mathbf{r}_i)$ – forces in their usual (newtonian) meaning[1]; n, m – the number of the scalar and vector coordinates describing a condition of system.

The fundamental identity (4) represents result of joint definitions of parameters ψ_i, Θ_i and $\mathbf{F}_j$, $\mathbf{r}_j$, and consequently is valid regardless of what causes the variation of the parameters Θ_i and $\mathbf{r}_j$ – either the external energy exchange or the internal (including relaxation and anti-relaxation) processes. Last is made against equilibrium in system in processes 'ascending diffusion', active transport in biosystems, polarization of substances, self-organizing of systems, etc. Therefore (1) is applicable to *any processes* (both reversible and irreversible). The work described by second sum of (4) may be external or internal (depending on where the forces arise – either in the system itself or outside); useful or dissipative (depending on what the work involves – either purposeful conversion of energy or its dissipation); long – range or short–range (depending on radius of action); mechanical, thermal, electrical, chemical, etc (depending on nature of the forces to overcome).

Expression (4) can be copied in the form, containing full derivatives on time t from parameters of system:

$$d\Im/dt \equiv \Sigma_i \, \psi_i d\Theta_i/dt - \Sigma_j \mathbf{F}_j \cdot \mathbf{v}_j. \tag{5}$$

were $\mathbf{v}_i \equiv d\mathbf{r}_i/dt$ – the generalized speed of jth process connected with common concept of a vector stream $\mathbf{J}_j$ of value Θ_i by a simple parity

$$\mathbf{J}_j = \Theta_j \mathbf{v}_j. \tag{6}$$

Owing to this generality and the concept of thermodynamic force $\mathbf{X}_j$ gets uniform sense of specific force $\mathbf{X}_j = \mathbf{F}_j/\Theta_i$, and expression $\mathbf{X}_j \cdot \mathbf{J}_j$ - capacityes of jth process $N_i = \mathbf{X}_i \cdot \mathbf{J}_i = \mathbf{F}_j \cdot \mathbf{v}_j$.

4. DIFFERENTIAL RECIPROCAL RELATIONS NON-EQUILIBRIUM PROCESSES

From (4) on the basis of the theorem about independence mixed derivative from the order of differentiation ($\partial^2\Im/\partial\mathbf{r}_i\partial\mathbf{r}_j = \partial^2\Im/\partial\mathbf{r}_j\partial\mathbf{r}_i$) follows:

$$(\partial F_i/\partial\mathbf{r}_j) = (\partial F_j/\partial\mathbf{r}_i). \tag{7}$$

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[1] The sign '−' before 2$^{\text{nd}}$ sums (1) conforms to a rule: useful work is positive, if it is made by system

This differential parity of thermodynamics can be expressed in the generalized speeds of processes, as $d\mathbf{r}_j = \mathbf{v}_j dt$ and $d\mathbf{r}_i = \mathbf{v}_i dt$. Then after reduction dt instead of (7) it is possible to write:

$$(\partial F_i/\partial \mathbf{v}_j) = (\partial F_j/\partial \mathbf{v}_i). \tag{8}$$

Considering (6), this parity can be expressed in terms of thermodynamic forces X_i, X_j and streams J_i, J_i :

$$(\partial X_i/\partial J_j) = (\partial X_j/\partial J_i). \tag{9}$$

These parities have been received by us earlier in more complex way and named by differential reciprocal relations [16]. Risselberghe (1962) was the first who postulated the existence of such-type relationships and proposed to name them the "generalized reciprocity relationships" [17]. It is simple to show, that the Onsager's symmetry conditions ensue directly from these differential relationships for a particular case of linear systems. In fact, applying (9) to equations (2) one can obtain:

$$(\partial J_i/\partial X_j) = L_{ij} = (\partial J_j/\partial X_i) = L_{ji}. \tag{10}$$

Thus the Onsager's reciprocal relations (symmetry conditions) ensue as a corollary of more general differential relationships of thermodynamics and do not need involving whatever statistic-mechanical considerations.

5. ANTI-SYMMETRY OF RECIPROCAL RELATIONS FOR PROCESSES OF INTER-CONVERSION OF ENERGY

Let's apply differential parities (7) to the system which is carrying out transformation by some i^{th} form of energy in j^{th}. In that case 2nd sum (5) addresses in zero. It means, that useful work W_i and W_j on the equation (5) get an opposite sign, and differential relationship (9) become anti-symmetric:

$$(\partial X_i/\partial J_j) = - (\partial X_j/\partial J_i). \tag{11}$$

The opposite sign of the works W_i and W_j means, that phenomenological laws (2) of process of transformation of energy taking the form [18]:

$$J_i = L_{ij} X_i - L_{ji} X_j , \tag{12}$$

$$J_j = L_{ji} X_i - L_{jj} X_j . \tag{13}$$

These kinetic equations can be recorded in more compact form (2), having added with their condition $X_i J_i + X_j J_j = 0$:

$$J_i = \Sigma_j\, L_{ij} X_j\ ;\quad (X_i J_i = -\, X_j J_j). \tag{14}$$

The kinetic equations of such character (with their terms having different signs) correspond to the concept of phenomenological (experience-based) laws than initial equations (2). The primary energy carrier flow J_i (e.g. the current in the primary winding of a transformer) is commonly known to decrease as the forces X_j being overcome increase (with approach to "no-load" operation) or, on the contrary, to increase as these forces decrease (with approach to "short circuit" operation). Similarly, the secondary energy carrier flow (e.g. the current in the secondary winding of a transformer) is commonly known to increase as the supply voltage X_i increases and to decrease as the secondary circuit resistance and the X_j decrease.

This circumstance throws fresh light on the origin of the Cazimir's reciprocity relationships $L_{ij} = -\, L_{ji}$ exposing the underneath meaning of the requirements for different parity of forces with respect to time inversion. In fact, for the dissipation forces not changing their signs with time inversion (i.e. for the so-called "α-type forces") the Onsager's symmetry conditions $L_{ij} = L_{ji}$, as shown above, are valid. Whenever a part of these forces have the reversible character (i.e. refer to the "β-type forces"), the Onsager's reciprocity relationships give place to anti-symmetry conditions $L_{ij} = -\, L_{ji}$. At the same time the consideration endeavored here shows that the applicability of the Cazimir's relationships is not actually restricted to the different-parity forces (α and β-type) case. In fact, let us assume dealing with energy conversion processes of purely dissipative character. Such are, in particular, thermal conductivity, electric conductivity, diffusion and viscous friction described by Fourier's, Ohm's, Fick's and Newton's laws and resulting in only the substance and energy transfer. In this case all terms of kinetic equation (2) describing vector phenomena have the same sign $\mathbf{X}_i \cdot \mathbf{J}_i > 0$ defined by their contribution to dissipative function. In this case reciprocity relationships (11) defining value and sign of the phenomenological coefficients L_{ij} in linear kinetic equations (2) give invariably positive values of the phenomenological coefficients $L_{ij} > 0$ in these equations and result in Onsager's reciprocity relationships $L_{ij} = L_{ji}$.

So for linear transfer processes of a purely dissipative character the matrix of phenomenological coefficients is always symmetrical. However, if in a transfer process useful (reversible) energy conversions occur, i.e. work is done against whatever forces other than dissipation ones, the reciprocity relationships acquire other character. In this case phenomena of the "ascending diffusion" type (transfer of components toward their concentration increase), system ordering, etc. are observed. These processes lead to gradients or differences of temperature, pressure, concentration, electric potential, etc., i.e. to deviation of the system from the internal equilibrium state for some of its degrees of freedom, whereas the system in whole is tending toward equilibrium. As a matter of fact, this is the nature of all the so-called

"superposition effects" the theory of irreversible processes deals with. As we will make sure hereafter, the effects of such a kind bear anti-dissipative character. The processes of useful conversion of the ith form of energy into the jth one in various machines apply to these effects, too. This substantially extends the applicability of the anti-symmetrical reciprocal relations [18].

6. VERIFICATION OF THE RECIPROCAL RELATIONS

It is a matter of interest to confirm the anti-symmetrical reciprocity relationships (4) in reversible processes on a wide class of the processes, submitting to the Maxwell's equations. For this let us consider a system consisting of a closed electric circuit with an arbitrary length ℓ_e and variable (in general case) cross section f_e, which comprises an as well closed magnetic circuit with a length ℓ_m and cross section f_m variable thru the length. In this case in the equation (14) X_i and X_j represent accordingly electromotive $X_e = \int\mathbf{E}\cdot d\boldsymbol{\ell}_e$ and magnetomotive $X_M = \int\mathbf{H}\cdot d\boldsymbol{\ell}_M$ forces [19], while $J_e = \int(d\mathbf{D}/dt)d\mathbf{f}_e$; $J_M = \int(d\mathbf{B}/dt)d\mathbf{f}_M$ – total fluxes of electric and magnetic displacements, respectively, sometimes named the "linkage fluxes" and traditionally represented by the number of the lines of force linking the cross section of the electric and magnetic circuits, respectively. Here $\mathbf{E}$, $\mathbf{H}$ – electric and magnetic fields; $\mathbf{D}$, $\mathbf{B}$ – vectors of electric and magnetic induction.

Let us now change, based on the Stokes theorem, in the force equation $X_e = \int\mathbf{E}\cdot d\boldsymbol{\ell}_e$ from the curvilinear integral taken over the closed electric circuit with a length of ℓ_e to the integral $\int\mathrm{rot}\mathbf{E}\cdot d\mathbf{f}_M$ over the magnetic circuit cross section f_M. In the similar way one can change in the force equation $X_M = \int\mathbf{H}\cdot d\boldsymbol{\ell}_M$ from the curvilinear integral over the closed magnetic circuit with a length of ℓ_M to the integral $\int\mathrm{rot}\mathbf{H}\cdot d\mathbf{f}_e$ over the surface f_e covering the electric circuit. Then $\partial X_e/\partial\mathbf{f}_M = \mathrm{rot}\mathbf{E}$; $\partial X_M/\partial\mathbf{f}_e = \mathrm{rot}\mathbf{H}$; $\partial^2 Z_e/\partial\mathbf{f}_e\partial t = \partial J_e/\partial\mathbf{f}_e = d\mathbf{D}/dt$, and $\partial^2 Z_M/\partial\mathbf{f}_M\partial t = \partial J_M/\partial\mathbf{f}_e = d\mathbf{B}/dt$. Substituting these expressions in reciprocity relations (11), we shall find:

$$\mathrm{rot}\ \mathbf{E} = - \ d\mathbf{B}/dt, \tag{15}$$

$$\mathrm{rot}\ \mathbf{H} = d\mathbf{D}/dt. \tag{16}$$

These equations differ from the corresponding Maxwell's equations in that they contain the total time derivatives of electric and magnetic induction vectors. This is not a surprise since the primary equations of thermodynamics (4) contain the exact differentials of vectors $\mathbf{r}_i$ [18]. To form equations (15) and (16) into a more habitual type, we shall consider, that $d\mathbf{D}/dt = \mathbf{j}_e + (\partial\mathbf{D}/\partial t)$, where $\mathbf{j}_e = \varrho_e\mathbf{v} = (\mathbf{v}\cdot\nabla)\mathbf{D}$ – conduction current caused by the free charge transfer, whereas magnetic analogs to the free charge ρ_e do not exist ($\nabla\mathbf{B} = 0$) and $d\mathbf{B}/dt = (\partial\mathbf{B}/\partial t)$. Thus finally:

$$\mathrm{rot}\ \mathbf{H} = \mathbf{j}_e + (\partial\mathbf{D}/\partial t); \tag{17}$$

$$\text{rot } \mathbf{E} = - \left(\partial \mathbf{B}/\partial t \right); \tag{18}$$

$$\text{div } \mathbf{D} = \rho_e ; \tag{19}$$

$$\text{div } \mathbf{B} = 0 . \tag{20}$$

Thus, Maxwel's equations in their modern type can be received as consequence of non-equilibrium thermodynamics without the assumption of existence «currents of displacement» and «vortical electromagnetic fields» in vacuum, or any other postulates. Except for that, the differential relationships between the state parameters and their functions enable solution of also other problems. In particular, they allow calculating by experimental data the fundamental state functions of a system, impose the restricting conditions that should be met for each of models. This always clarifies the models because allows neglecting redundant or non-existent constraints. Therefore, the differential relationships are a very effective tooling of mathematical analysis conducted on an object of investigation [20].

7. CONCLUSIONS

As we have seen, introduction of parameters of spatial heterogeneity allows to distribute a thermodynamic method of the phenomena of transfer to processes of useful transformation of various forms of energy. The mathematical properties of full differential of such parameters allow to give consistently thermodynamic background of differential reciprocal relations, more general, rather than a Onsager's symmetry condition. It explains, why they are justified and outside those restrictions which were imposed by their statistic-mechanical background. Differential reciprocal relations applicable to processes of useful transformation of energy that proves to be true by a conclusion to their basis of Maxwell's equations to which the wide class of converters of energy submits.

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