GENERALIZED IRREVERSIBLE THERMODYNAMICS AND ITS APPLICATION TO LASERS. PART I. GENERAL THEORY*

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Dedicated to Professor P. T. Landsberg in appreciation of his excellent selection of "quotations on irreversibility and entropy" (Landsberg 1970).

A general irreversible quantum statistical thermodynamics with many temperatures is formulated. It is called the "generalized irreversible thermodynamics" because of the possibility of occurrence of many temperatures with the same or different dimensions (in particular of temperatures of different orders). The theory is based on a non-Hamiltonian (irreversible) dynamics of density operators described by a semigroup of motions satisfying Kossakowski's axioms (Kossakowski 1972), and on a generalization of the Kossakowski principle of the isoentropic motion (Kossakowski 1969). It is assumed that the system is observed only in an initial moment in the macroscopic sense (*i.e.* a finite number of ensemble mean values of linearly independent and thermodynamically regular observables of the system are measured). The theory describes only the really irreversible effects and neglects temporal fluctuations of entropy (memory cycle effects). The paper gives also a classification of all possible motions of a non-isolated quantum system.

1. Introduction

"...et sic matheseos demonstrationes cum aleae incertitudine jungendo et quae contraria videntur conciliando ab utraque nominationem suam accipiens stupendum hunc titulem jure sibi arrogat: aleae Geometria."

Blaise Pascal[1]

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[1] "...and thus joining mathematical proofs with the uncertainty of chance and reconciling seeming contradictions, lending its name from both its parts, this science is fully worthy of its astonishing name: Geometry of chance." B. Pascal in a letter to the Academy of Paris, *Celeberrimae Matheseos Academiae Parisiensi*, 1654 (Pascal 1954, p. 73, *cf.* also Rényi 1969, pp. 18 and 89).

Pascal, one of the founders of probability theory, expressed in these words the conceptual difficulties which plagued this theory since its beginning almost to the present days, and which plague statistical thermodynamics even now. The problem consists, namely, in "reconciling seeming contradictions" between certainty and uncertainty, and between reversibility and irreversibility, respectively. After centuries of inconclusive discussions the first problem was finally solved by Kolmogorov (1933) by means of a rigorous axiomatic formulation of the concept of probability. We think that the second problem was solved only recently by Kossakowski (1972) also by means of an axiomatic formulation, in this case of the concept of irreversible motion. The irreversible motions of a physical system form a semigroup, while the reversible motions form a group, and the difference lies in the existence oı non-existence of the inverse element (inverse motion) to a given motion. Thus we can never obtain irreversibility from reversibility by any mathematical tricks, such as averaging, coarse-graining, incomplete knowledge, smoothing, *etc.*, in spite of claims of many authors (*cf.* the mentioned selection of quotations by Landsberg (1970) which shows the drastic differences of opinions on the foundations of statistical thermodynamics between many leading contemporary scientists of today). Shortly, neither probability theory nor information theory can produce irreversibility from reversibility of a mechanical system, or more precisely of a closed (isolated) mechanical system, classical or quantum. What these theories can actually produce at most is only seeming irreversibility, "reversible irreversibility", *i. e.* periodic or quasi-periodic oscillations of entropy (information), where information is lost and recovered again, as in memory systems of computers (we shall call such phenomena m e m o r y c y c l e e f f e c t s or m e m o r y l o o p e f f e c t s). Some people claimed that periods of such oscillations may be enormously long (Poincaré periods), but we shall show below by an explicit calculation of an example that, on the contrary, these periods may be also enormously short (as Larmor precession periods). The problem, however, consists not in the shortness or largeness of some periods, but in their existence or non-existence. Therefore, the principal question of irreversibility cannot be solved by any approximative method, such as the perturbation method, for instance. Also finite systems (with a finite number of particles), even very large, cannot be approximated (for this purpose) by infinite systems, as is frequently done (*cf.* Ruelle 1969) simply because the difference between these systems is infinite. Infinite systems are closed and open in the same time, both in the topological and in the physical sense of this word (the latter meaning isolation or non-isolation of a system), and therefore they can have a Hamiltonian and be irreversible, which cannot be true for any finite system. Finite systems are irreversible only if they are open (non-isolated), *i. e.* n o n - H a m i l t o n i a n, anyway on the base of the present quantum (and classical) mechanics; how this will turn out with future mechanics, we cannot judge nor even guess now. Closeness (isolation) means non-existence of any dynamical interaction with particles or degrees of freedom which do not belong to our system, no matter whether they lie inside or outside of the spacial boundary of the system. (The so-called internal friction is an example of the former, and the radiation damping of a laser is an example of the latter.)

The aim of the present paper is twofold. In part I we would like to formulate and discuss the principle of irreversible statistical thermodynamics in our sense, including also multitemperature thermodynamics (Ingarden 1963, 1965, 1968, 1969), and called therefore generalized irreversible thermodynamics. In this formulation we shall use and in part generalize important results of Kossakowski (1969, 1972) which were obtained in close contact and collaboration with the present investigation. In Part II we shall develop our investigation of thermodynamics of a simple model of the laser published previously only in a short and imperfect summary (Ingarden 1971), using also some results of Z. Kojro which will be published in his Ph. D. thesis performed under the guidance of the present author. The investigations of Part II will be an illustration and application of the general methods presented in Part I.

2. *A classification of motions of general quantum systems*

By a general quantum system we mean a quantum system which is closed (isolated, Hamiltonian) or open (non-isolated, non-Hamiltonian). By a motion in the most general sense of a general quantum system we mean an arbitrary map

$$\Lambda: W \to W, \tag{2.1}$$

where W is the set of all density operators (states, mixed states) of the system, *i. e.*

$$W = W(H) := \{\varrho \in L(H): \varrho \geqslant 0, \operatorname{Tr} \varrho = 1\}. \tag{2.2}$$

Here: $=$ means "equal by definition", $L(H)$ is the set of all linear operators acting in the Hilbert space H of the system, and $\varrho \geqslant 0$ denotes the positive definiteness of operator ϱ.

Following Kossakowski (1972) we introduce several linear spaces connected with the Hilbert space H. Let $B_1(H)$ be the Banach space (over the field $\boldsymbol{R}$ of real numbers) of self-adjoint trace class linear operators on H — a linear operator is called trace class iff (if and only if) its trace is finite — with the norm

$$||\varrho||_1 := \sup \sum_{n=1}^{\infty} |(x_n, \varrho y_n)|, \tag{2.3}$$

where the supremum is taken over all orthonormal complete bases $\{x_n\}$ and $\{y_n\}$ in H, and $(.,.)$ denotes the scalar product in H, *i. e.*

$$B_1(H) := \{\varrho \in L(H): \varrho = \varrho^*, ||\varrho||_1 < \infty\}. \tag{2.4}$$

The set of all real valued continuous linear functionals on $B_1(H)$ is called the dual space to $B_1(H)$ and will be denoted by $B_\infty(H)$. It is well-known (Dunford and Schwartz 1963) that each real continuous linear functional on $B_1(H)$ has the form

$$\langle A, \varrho \rangle := \operatorname{Tr}(A\varrho), \quad \varrho \in B_1(H), \tag{2.5}$$

where A is a bounded self-adjoint linear operator on H (a bounded observable of our system). The space $B_\infty(H)$ is also a Banach space with the norm

$$||A||_\infty := \sup_{||\varrho||_1 = 1} |\langle A, \varrho \rangle| = \sup_{x \in H} \frac{||Ax||}{||x||},$$

$$A \in B_\infty(H), \; ||x||^2 = (x, x). \tag{2.6}$$

The set of all positive definite operators from $B_1(H)$ is called the positive cone in $B_1(H)$ and will be denoted by $B_1^+(H)$, *i. e.*

$$B_1^+(H) := \{\varrho \in B_1(H) : \varrho \geqslant 0\}. \tag{2.7}$$

Denoting by $B_\infty^+(H)$ the dual cone to $B_1^+(H)$, *i. e.* the set of all $A \in B_\infty(H)$ such that $\langle A, \varrho \rangle \geqslant 0$ for all $\varrho \in B_1^+(H)$, we see that $B_\infty^+(H)$ consists of all positive definite bounded linear operators on H, *i. e.* is the positive cone in $B_\infty(H)$,

$$B_\infty^+(H) := \{A \in B_\infty(H) : A \geqslant 0\}. \tag{2.8}$$

We have

$$||\varrho||_1 = \mathrm{Tr}\,\varrho, \quad \varrho \in B_1^+(H), \tag{2.9}$$

but this equality is in general not true outside of $B_1^+(H)$ (since trace can be then negative, but norm is always non-negative).

The Banach space $B_1(H)$ is the smallest linear space in which the set $W(H)$ of all states may be embeded, and we have

$$W(H) = \{\varrho \in B_1^+(H) : ||\varrho||_1 = 1\}. \tag{2.10}$$

The set $\mathscr{B}_1(H)$ of all bounded linear operators on $B_1(H)$ (superoperators with respect to H) is also a Banach space, namely, with respect to the norm

$$|||\Lambda|||_1 := \sup_{||\varrho||_1 = 1} ||\Lambda\varrho||_1, \quad \varrho \in B_1(H), \Lambda \in \mathscr{B}_1(H). \tag{2.11}$$

It can be shown that to every $\Lambda \in \mathscr{B}_1(H)$ there corresponds one and only one linear operator Λ^* on $B_\infty(H)$ defined by

$$\langle A, \Lambda\varrho \rangle = \langle \Lambda^* A, \varrho \rangle, A \in B_\infty(H), \varrho \in B_1(H). \tag{2.12}$$

The set of all Λ^* forms also a Banach space $\mathscr{B}_\infty(H)$ (dual to $\mathscr{B}_1(H)$) with the norm

$$|||\Lambda^*|||_\infty := \sup_{||A||_\infty = 1} ||\Lambda^* A||_\infty = |||\Lambda|||_1. \tag{2.13}$$

A linear operator $\Lambda \in \mathscr{B}_1(H)$ is called a positive endomorphism of $B_1(H)$ iff it satisfies the conditions

$$1) \quad \Lambda : B_1^+(H) \to B_1^+(H), \tag{2.14}$$

$$2) \quad ||\Lambda\varrho||_1 = ||\varrho||_1, \quad \varrho \in B_1^+(H). \tag{2.15}$$

A positive endomorphism of $B_1(H)$ maps $W(H)$ into itself and, therefore, is a motion. We call such a motion a linear motion.

The following theorem can be proved (Kossakowski 1972) for positive endomorphisms:

$$||\Lambda\varrho||_1 \leqslant ||\varrho||_1, \quad \varrho \in B_1(H), \tag{2.16}$$

i. e. that Λ is a contracting operator (*cf.*, *e. g.*, Dynkin 1965, Yosida 1965).

We say that a sequence of $\{\varrho_n\}$ of elements of $B_1(H)$ is strongly convergent to an element $\varrho \in B_1(H)$ and write

$$\operatorname*{s-lim}_{n\to\infty} \varrho_n = \varrho \tag{2.17}$$

iff

$$\|\varrho_n - \varrho\|_1 \to 0 \text{ for } n \to \infty. \tag{2.18}$$

It is well-known (*cf.*, *e. g.*, Wichmann 1963) that the set $W(H)$ of all states is convex, and its extremal points (and only they) are pure states, *i. e.* states of the form

$$\varrho = P_x, \quad x \in H, \tag{2.19}$$

where P_x is the projector operator on a vector x.

We may proceed now to a classification of motions. First of all we shall classify motions with respect to pure or mixed character of initial and final states,

$$\varrho' = \Lambda\varrho, \tag{2.20}$$

where ϱ is an initial, ϱ' — a final state, and $\Lambda \in \Omega$, $\Omega = \Omega(H)$ being the set of all motions of a system with Hilbert space H. For the initial state there are only two possibilities: P — pure, and M — mixed (non-pure). For the final state by a given character of the initial state there exist, however, three possibilities: P, M, and C — combined (pure or mixed, depending on other properties of the initial state). In such a way we obtain the complete classification:

TABLE I

Initial state	Final state ($t = \infty$)								
	1	2	3	4	5	6	7	8	9
P	P	P	M	M	C	C	C	P	M
M	M	P	M	P	C	P	M	C	C

Of the enumerated 9 cases only 3 first ones occur in our investigations: an example of case 1 is any Hamiltonian motion, examples of cases 2 and 3 will be given in Part II (they are examples or ergodic motions). Whether the other cases can occur in nature depends on other properties which should be assumed for real motions. If we assume that each motion in strongly continuous in $B_1(H)$, *i. e.* in the sense of the strong convergence in $B_1(H)$ (2.18), we obtain the result that the motion is linear.

Indeed, if we make a statistical mixture of two initial states ϱ_1 and ϱ_2 with probabilities

$$p_1, p_2 \geqslant 0, \quad p_1 + p_2 = 1, \tag{2.21}$$

i.e.

$$\varrho = p_1\varrho_1 + p_2\varrho_2, \quad \varrho_1, \varrho_2 \in B_1(H), \tag{2.22}$$

this mixture should be preserved also after transformation (because we have an ensemble of physical systems which do not interact physically and which are in the same environment by definition)

$$\Lambda(p_1\varrho_1+p_2\varrho_2) = p_1\Lambda\varrho_1+p_2\Lambda\varrho_2, \tag{2.23}$$

cf. Schlögl, Stahl and Bausch (1965). Eq. (2.23) is a functional equation which among strongly continuous solutions has only a linearly inhomogeneous solution as we may infer by analogy with the Jansen functional equation (*cf.* Aczél 1961, p. 49, we do not go here into details of a rigorous proof). (We may remark that (2.23) is weaker than the condition of linearity because of (2.21), by linearity p_1 and p_2 should be arbitrary real or complex numbers.) But inhomogeneous term can be avoided (Schlögl, Stahl and Bausch 1965) by writing (since Tr $\varrho = 1$)

$$L\varrho+\varrho_0 = (L+\varrho_0\,\mathrm{Tr})\varrho = L'\varrho, \tag{2.24}$$

where L and L' are linear (homogeneous) operators in $W(H)$.

Up to now we did not consider time as a parameter of motion (we are interested here in nonrelativistic quantum physics only). With respect to time we may distinguish three principal types of linear motions: Markov processes (of order 1), *i. e.* described by linear differential equations of order 1 with respect to ϱ and time t, Markov processes of higher order, *i. e.* described by linear differential equations of order $n > 1$ with respect to ϱ and t, and stochastic processes with hysteresis, *i. e.* described by a linear integral equation with respect to ϱ and t, irreducible to linear differential equations of final order (motion non-local in time).

Kossakowski (1972) discussed the first of these three possible cases and for the following we shall confine ourselves only to the case discussed by him. His axiomatic definition goes as follows:

Definition 1. A family $Y(H) := \{\Lambda(t), t \geqslant 0\}$ of endomorphisms of $B_1(H)$ is said to be a dynamical semigroup (quantum Markov process) of a quantum system iff the following conditions are satisfied:

1) $\Lambda(t): B_1^+(H) \to B_1^+(H), \quad t \geqslant 0,$ (2.25)

2) $||\Lambda(t)\varrho||_1 = ||\varrho||_1, \quad \varrho\in B_1^+(H), \quad t \geqslant 0,$ (2.26)

3) $\Lambda(t)$ is strongly continuous with respect to $t \geqslant 0$, (2.27)

4) $\underset{t\to+0}{s\text{-lim}}\, \Lambda(t)\varrho = \varrho, \quad \varrho\in B_1(H),$ (2.28)

5) $\Lambda(t)\,\Lambda(s) = \Lambda(t+s), \; t, s \geqslant 0.$ (2.29)

In other words, we may express this definition by saying that a dynamical semigroup $Y(H)$ is a strongly continuous one-parameter contracting semigroup of positive endomorphisms of $B_1(H)$.

We have the following

Theorem 1. (Kossakowski 1972). Let $Y(H)$ be a dynamical semigroup in $B_1(H)$. A family $Y^*(H) := \{\Lambda^*(t), t \geqslant 0\}$ of endomorphisms $\Lambda^*(t)$ of $B_\infty(H)$ dual to $\Lambda(t) \in Y(H)$ $t \geqslant 0$, satisfies the conditions

$$1) \quad \Lambda^*(t) : B_\infty^+(H) \to B_\infty^+(H), \qquad t \geqslant 0, \tag{2.30}$$

$$2) \quad \Lambda^*(t)\, I = I,\ t \geqslant 0 \ (I \text{ is the identity operator on } H), \tag{2.31}$$

$$3) \quad ||\Lambda^*(t)A||_\infty \leqslant ||A||_\infty, \qquad A \in B_\infty(H), \tag{2.32}$$

$$4) \quad \Lambda^*(t) \text{ is strongly continuous with respect to } t \geqslant 0, \tag{2.33}$$

$$5) \quad \underset{t \to +0}{s\text{-lim}}\, \Lambda^*(t)A = A, \qquad A \in B_\infty(H), \tag{2.34}$$

$$6) \quad \Lambda^*(t)\Lambda^*(s) = \Lambda^*(t+s), \qquad t, s \geqslant 0. \tag{2.35}$$

The family $Y^*(H)$ dual to $Y(H)$ will be called the dual dynamical semigroup of a quantum system. From (2.12) it follows that

$$\langle A, \Lambda(t)\varrho\rangle = \langle \Lambda^*(t)A, \varrho\rangle, \quad \varrho \in B_1(H),\ A \in B_\infty(H),\ t \geqslant 0. \tag{2.36}$$

Dynamical semigroups $Y(H)$ and $Y^*(H)$ are isomorphic and they describe the time evolution of our physical system in the Schrödinger picture and in the Heisenberg picture, respectively. Eq. (2.36) shows that the mean values do not depend on the picture. This is shown only for the bounded observables, but approximating unbounded observables by the bounded ones we obtain the same result for any observable, if only a mean value exists.

Using a theorem by Hille (Hille and Phillips 1957) and Yosida (1965) it may be easily proved that there exists an infinitesimal operator L of $Y(H)$, and L^* of $Y^*(H)$, defined on dense sets $D(L) \subset B_1(H)$ and $D(L^*) \subset B_\infty(H)$ respectively, such that the following equations are satisfied

$$\frac{d}{dt}(\Lambda(t)\varrho) = L(\Lambda(t)\varrho) = \Lambda(t)\,(L\varrho), \qquad \varrho \in D(L), \tag{2.37}$$

and

$$\frac{d}{dt}(\Lambda^*(t)A) = L^*(\Lambda^*(t)A) = \Lambda^*(t)\,(L^*A), \qquad A \in D(L^*). \tag{2.38}$$

Generators L and L^* may be called generalized Liouville operators. Eqs (2.37) and (2.38) acquire more common forms when we write

$$\varrho(t) := \Lambda(t)\varrho, \quad A(t) := \Lambda^*(t)A. \tag{2.39}$$

Kossakowski (1972) proved the following fundamental

Theorem 2. Let $Y_0(H) = \{\Lambda(t), t \geqslant 0\}$ be a family of endomorphisms of $B_1(H)$ satisfying the conditions

1) (2.25), 3) (2.27), 4) (2.28), 5) (2.29) and instead of the condition 2) (2.26), the stronger condition

$$2')\quad ||\Lambda(t)\varrho||_1 = ||\varrho||_1, \quad \varrho \in B_1(H), t \geqslant 0. \tag{2.40}$$

Then there exists a strongly continuous one-parameter group $G_0(H){:} = \{U(t), t \in \boldsymbol{R}\}$ of unitary operators on H such that

$$\Lambda(t)\varrho = U(t)\varrho\, U^{-1}(t), \quad t \geqslant 0, \varrho \in B_1(H), \tag{2.41}$$

i. e. the semigroup $Y_0(H)$ can be extended to the group $G_0(H)$. Such a motion is a Hamiltonian motion and is reversible. We see, therefore, that irreversibility of motion lies in the difference in the conditions 2) and 2′), in particular, in the difference between the Banach space $B_1(H)$ and its positive cone $B_1^+(H)$ as the domain of preservation the norm (2.3) being in $B_1^+(H)$ (and only there) equivalent to the trace of density operators. Preservation of this trace means preservation of probability which has a sense only inside of the cone. We see how closely mathematics is here connected with its physical interpretation.

Examples of non-Hamiltonian motions and their application to the theory of lasers will be given in Part II.

3. *Irreversible statistical thermodynamics*

What has been presented up to now is not yet statistical thermodynamics, it may be called at most statistical mechanics (we carefully distinguish between these two concepts). Statistical mechanics is mechanics, Hamiltonian or non-Hamiltonian, reversible or non-reversible, of statistical operators (density operators), while the usual (Hamiltonian) mechanics is the same theory concerning only pure states (state vectors). (Both theories can be distinguished only in the case 1 of Table I). Generalized Kossakowski's equations (2.37), (2.38) with substitution (2.39) can be called also the quantum master equations or the generalized quantum Fokker-Planck equations (a more special form of the Fokker-Planck equation will be considered in Part II) concerning (in contradistinction to the preceding theories) not only the diagonal part of density operators, but its full shape. However, in order to integrate these equations we have not only to know how to integrate operational equations, but also to know the initial conditions for these equations, *i.e.* either ϱ from $W(H)$ or A from $B_\infty(H)$. The latter knowledge requires in general infinite number of measurements and is mostly unavailable, anyway not by a macroscopic measurement. Macroscopic measurement is by definition incomplete and contains only a finite number of numerical values.

Our idea of statistical thermodynamics is based on the concept of a macrostate which is closely connected with that of macroscopic measurement. The origin of the idea of a macrostate may be traced to Boltzmann, but its precise formulation by means of information-theoretical methods was apparently given first by Ingarden and Urbanik (1962). In this paper this concept was defined only for one moment of time and with respect to one observable (A-thermodynamics), and without explicit use of density operators. Only in later papers (Ingarden 1963, 1965, 1968, 1969) many observables and density operators were introduced. On the other hand, Urbanik (1964) gave the first definition of the time evolution of macrostate (without repeating measurements) of a Hamiltonian system, and his definition was modificated and essentially improved by Kossakowski (1969, 1970). In the two latter papers Kossakowski gave also closed equations of motions for mean values of observables in question which may be justly called thermodynamical equations since they contain only macroscopical information and its time evolution. Kossakowski used for this the so-called isoentropic approximation which simplified very the much solution by making equations local in time. Independently, Robertson (1966, 1967) found similar equations, but without the concept of macrostate and the method of isoentropic motion, although using information-theoretical methods of Jaynes (1957, 1957a). In the present paper we generalize the Kossakowski isoentropic method for the non-Hamiltonian motion and give the physical interpretation of this method. This inteıpretation seems to be important since both Robertson and Kossakowski considered their methods as showing irreversibility even by Hamiltonian motion, which according to the present author is impossible. The present author distinguishes between "real" irreversibility (monotonic in time change of entropy) and "apparent", or "quasi" irreversibility (periodic or quasi-periodic change of entropy). In Part II it will be shown how this generalized method works in practice, and how reasonable and useful are the results it gives in the laser theory.

Let us consider n observables $A_1, A_2, \ldots, A_n$ (not necessarily bounded) of a physical system (for simplicity we work in the Schrödinger picture, but all the theory can be rewritten also in the Heisenberg picture, *cf.* Section 2), such that

1) if $a_0 I + a_1 A_1 + a_2 A_2 + \ldots + a_n A_n = 0,\ a_0, a_1, \ldots, a_n \in \boldsymbol{R}$,

$$\text{then } a_0 = a_1 = \ldots = a_n = 0 \tag{3.1}$$

(we say then that $I, A_1, \ldots, A_n$ are linearly independent),

2) there exists $a_1, \ldots, a_n \in \boldsymbol{R}$ such that

$$\operatorname{Tr} \exp\left(-\sum_{j=1}^{n} a_j A_j\right) < \infty \tag{3.2}$$

(we say then that $A_1, \ldots, A_n$ are thermodynamically regular).

Now we assume that in time $t = 0$ we measure the ensemble mean values of observables $A_1, \ldots, A_n$

$$\langle A_1, \varrho \rangle = \operatorname{Tr}(A_1 \varrho) = U_1, \ldots, \langle A_n, \varrho \rangle = \operatorname{Tr}(A_n \varrho) = U_n, \quad \varrho \in W(H). \tag{3.3}$$

The macrostate M in time $t = 0$ is defined as

$$M := \{\varrho \in W(H) : \operatorname{Tr}(A_j \varrho) = U_j, j = 1, \ldots, n\}. \tag{3.4}$$

The mean values of macrostate M are by definition U_j ($j = 1, \ldots, n$) while the entropy (information) of M is defined as

$$S = S(M) := \sup_{\varrho \in M} (-\operatorname{Tr}(\varrho \ln \varrho)). \tag{3.5}$$

It may be shown (Ingarden and Urbanik, 1962, Wichmann 1963, Kossakowski 1970) that there exists the state $\varrho_M \in M$ (called the representative state of M) such that

$$-\mathrm{Tr}\,(\varrho_M \ln \varrho_M) = S(M), \tag{3.6}$$

i.e. the state of the maximum entropy in M.

To construct the macrostate in time $t \geqslant 0$ we assume that all $\varrho \in W(H)$ develop in time according to equations (2.37) and (2.39)

$$\frac{d\varrho(t)}{dt} = L\varrho(t), \qquad \varrho(t) = \Lambda(t)\varrho. \tag{3.7}$$

In particular, (3.7) defines the time evolution of ϱ_M:

$$\varrho_M(t) := \Lambda(t)\varrho_M, \qquad t \geqslant 0. \tag{3.8}$$

By means of $\varrho_M(t)$ we define in turn

$$U_j(t) := \mathrm{Tr}\,(A_j \varrho_M(t)), \qquad j = 1, \ldots, n, \qquad t \geqslant 0, \tag{3.9}$$

and

$$M(t) := \{\varrho \in W(H) : \mathrm{Tr}\,(A_j \varrho) = U_j(t), \quad j = 1, \ldots, n\}, \qquad t \geqslant 0, \tag{3.10}$$

called just the macrostate in time $t \geqslant 0$. The state $\varrho_M(t) \in M(t)$, but not necessarily has the maximum value of entropy over $M(t)$ (such a situation occurs, in general, when A_j do not commute among themselves and $L^*A_j \neq 0$, so for $t > 0$ it is not necessarily the representative state of $M(t)$). The representative state of $M(t)$ will be denoted by

$$\sigma_M(t) := \varrho_{M(t)} \tag{3.11}$$

and may be easily calculated by means of $U_j(t)$, $j = 1, \ldots, n$. Namely (*cf.* Ingarden 1965, 1968, Kossakowski 1969, 1970), we obtain

$$\sigma_M(t) = Z^{-1}(\beta_1(t), \ldots, \beta_n(t)) \exp\,(-\sum_{j=1}^{n} \beta_j(t) A_j), \qquad t \geqslant 0, \tag{3.12}$$

$$Z(\beta_1(t), \ldots, \beta_n(t)) := \mathrm{Tr}\,\exp\,(-\sum_{j=1}^{n} \beta_j(t) A_j), \tag{3.13}$$

where the inverse temperatures $\beta_j(t)$, $j = 1, \ldots, n$, can be calculated from the equations

$$U_j(t) = -\frac{\delta}{\delta\beta_j(t)} \ln Z(\beta_1(t), \ldots, \beta_n(t)), \qquad j = 1, \ldots, n, \; t \geqslant 0. \tag{3.14}$$

We have, of course,

$$\sigma_M(0) = \varrho_M(0) = \varrho_M. \tag{3.15}$$

All this calculation is unique and in principle can be performed in any case. Practically, however, such a calculation can be carried out only in very exceptional cases, even if the motion is Hamiltonian (which we do not assume). The difficulty consists in integration of equations of motion (2.37) or (3.7) and finding $\varrho_M(t)$ (3.8). Therefore, we shall try to find $U_j(t)$ directly from Kossakowski's master equations (2.37) and initial values $U_j(0) = U_j$, $j = 1, \ldots, n$, by using information-theoretical estimation consisting in maximizing entropy (the Jaynes principle, Jaynes 1957, 1957a) in any time $t \geqslant 0$. We obtain

$$\frac{dU_j(t)}{dt} = \mathrm{Tr}\left(A_j \frac{d\varrho_M(t)}{dt}\right) = \mathrm{Tr}\,(A_j L \varrho_M(t)), \quad j = 1, \ldots, n. \tag{3.16}$$

Writing

$$\varrho_M(t) = \sigma_M(t) + (\varrho_M(t) - \sigma_M(t)) \tag{3.17}$$

and neglecting the difference $\varrho_M(t) - \sigma_M(t)$ in comparison with $\sigma_M(t)$ we get

$$\frac{dU_j(t)}{dt} = \mathrm{Tr}\,(A_j L \sigma_M(t)) = F_j(U_1(t), \ldots, U_n(t)),$$

$$j = 1, \ldots, n, \quad t \geqslant 0 \tag{3.18}$$

since $\sigma_M(t)$ is a function of $U_1(t), \ldots, U_n(t)$ in the same time $t \geqslant 0$ defined by (3.12), (3.13), and (3.14). In such a way we obtain a closed system of equation of motion for the thermodynamic quantities $U_j(t)$ from which we calculate the inverse temperatures $\beta_j(t)$ and the thermodynamic entropy

$$S(t) := \sup_{\varrho \in M(t)} (\mathrm{Tr}\,(-\varrho \ln \varrho)) = \mathrm{Tr}\,(-\sigma_M(t) \ln \sigma_M(t)), \quad t \geqslant 0. \tag{3.19}$$

Eqs (3.18) can be now integrated independently of integration of operator equation (2.37) with initial conditions

$$U_j(0) = U_j, \quad j = 1, \ldots, n. \tag{3.20}$$

We may call (3.18) the thermodynamical equations of motion of our system observed macroscopically by means of observables $A_1, \ldots, A_n$ (of $(A_1, \ldots, A_n)$-thermodynamics, as may be said more precisely). Our method is a direct generalization of the Kossakowski izoentropic method (Kossakowski 1969, 1970) for a non-Hamiltonian motions. Now $S(t)$ is no more constant in time, we obtain in general a monotonic increase of entropy (if we have no pumping of information into the system). In Part II we shall study this behaviour on particular simple examples, and only after this discussion we shall come back to the problem of general physical interpretation of our method which may be called quasi-isoentropic or of minimal entropy increase.

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