

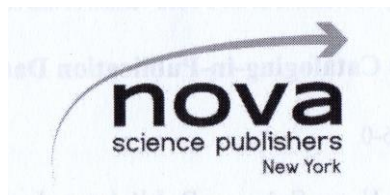
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PHYSICS RESEARCH AND TECHNOLOGY

ADVANCES IN THERMODYNAMICS RESEARCH

JOSEPH A. COBB
EDITOR



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PREFACE

This book consists of three chapters that each describe recent advancements in the field of thermodynamics, which is a branch of physics that deals with heat, work, and temperature. Chapter One provides a calculated confirmation of the theoretical positions of the concept of non-equilibrium and its mathematical apparatus using examples of determining the loss of non-equilibrium of adiabatic and isolated systems. Chapter Two focuses on the thermodynamic behavior of hydrophobic non-electrolyte tetramethylurea (TMU) and well-established hydrophilic non-electrolyte urea (U) in three solvents forming a three-dimensional H-bond network on a supramolecular level viz. water, ethylene glycol (EG) and formamide (FA). Chapter Three introduces the concept of generalized entropy and discusses some of its properties.

Chapter 1 – Most physical laws are quantitative expressions of the philosophical laws of the conservation of matter and its property of motion. The first law of thermodynamics (FLT) is an analytical expression of the law of conservation of motion when its shape changes. As for the second law of thermodynamics (SLT), it has not yet been clarified which property of matter does not change during the course of reversible processes and changes during the course of irreversible processes in an isolated system (IS). From here, a large number of the second law formulations and an abundance of material to clarify these formulations. The author introduces

into the basis of the second law of thermodynamics “the postulate of non-equilibrium,” according to which there is an objective property of matter - “non-equilibrium,” which characterizes the unequal distribution of matter and motion in space. All processes (reversible and irreversible) can occur only in non-equilibrium systems. A new formulation of the SLT is given in relation to the set of locally equilibrium subsystems that make up the non-equilibrium system: when real (irreversible) processes occur, the non-equilibrium of the isolated system (IS) decreases, and in reversible processes the non-equilibrium in the system of locally equilibrium subsystems does not change (the increment of one kind of non-equilibrium completely compensated by a decrease in the disequilibrium of some other kind). As a quantitative characteristic (measure) of the non-equilibrium of the system, the maximum work that can be performed during the transition of the non-equilibrium system to the equilibrium state is considered. To calculate this work, the following quantities are used: “potential difference,” the “entropic difference, and change in exergy. All these quantities decrease during the course of real (irreversible) processes in the IS, and do not change during the course of reversible (ideal) processes. As a result, a generalized expression of the SLT was obtained through the quantitative characteristics of the non-equilibrium of the system in the form of an inequality, including the R. Clausius inequality for the change in the entropy of an isolated system. This chapter provides a calculated confirmation of the theoretical positions of the concept of non-equilibrium and its mathematical apparatus using examples of determining the loss of non-equilibrium of adiabatic and isolated systems.

Chapter 2 – Hydrophobic hydration and hydrophobic interaction play the major role in various important chemical phenomena such as formation of nanoscale micelles and bilayer membranes, protein folding, molecular recognition, *etc.* These processes are often considered to be important steps of the origins of life. There is only little experimental evidence that similar but more weakly pronounced phenomena may take place in H-bonded non-aqueous solvents such as *N*-methylacetamide, monoethanolamine or ethylene glycol. However, the important experimental information for comparative analysis is rather scarce. In this chapter, the authors focus on

the thermodynamic behavior of hydrophobic non-electrolyte tetramethylurea (TMU) and well-established hydrophilic non-electrolyte urea (U) in three solvents forming a three-dimensional H-bond network on a supramolecular level *viz.* water, ethylene glycol (EG) and formamide (FA). The standard enthalpies, volumes and heat capacities of solution and transfer of U and TMU are determined and compared at various temperatures in the first section to highlight common features and fundamental differences in solvation of non-electrolytes in aqueous and non-aqueous media. To explain the heat capacity values in water, the available results of computer simulations are invoked and discussed. The second section contains important information on the solute-solute pair interaction in a liquid phase obtained by the virial expansion technique. The enthalpic, free energy, entropic, heat capacity and volume pair- interaction parameters are computed both in water and non-aqueous solvents. Special attention is paid to the calculation and analysis of the second virial coefficient and its temperature dependence. The authors' comparative analysis highlights several unique features of TMU solvation and pair interactions in non-aqueous media which are very similar to those observed in an aqueous solution of small hydrophobic units. The authors show that the solvophobic interaction between two TMU molecules in EG and, especially, in FA does exist. However, it is weaker compared to hydrophobic interaction in water leading to the formation of large TMU clusters.

Chapter 3 – The concept of generalized entropy is introduced and some of its properties are studied. Irreversible time evolution can be generated by a non-Hermitian superoperator on the states of the system. The case when irreversibility comes about from embedding the system in a thermal reservoir is looked at. The time evolution is found compatible both with equilibrium thermodynamics and entropy production near the final state. Some examples are presented as well as a longer introduction as to how this might play a role in the black hole information loss paradox.

Chapter 1

THE CONCEPT OF NON-EQUILIBRIUM AS THE BASIS OF THE SECOND LAW OF THERMODYNAMICS

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ABSTRACT

Most physical laws are quantitative expressions of the philosophical laws of the conservation of matter and its property of motion. The first law of thermodynamics (FLT) is an analytical expression of the law of conservation of motion when its shape changes. As for the second law of thermodynamics (SLT), it has not yet been clarified which property of matter does not change during the course of reversible processes and changes during the course of irreversible processes in an isolated system (IS). From here, a large number of the second law formulations and an abundance of material to clarify these formulations.

The author introduces into the basis of the second law of thermodynamics “the postulate of non-equilibrium,” according to which

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there is an objective property of matter - “non-equilibrium,” which characterizes the unequal distribution of matter and motion in space. All processes (reversible and irreversible) can occur only in non-equilibrium systems.

A new formulation of the SLT is given in relation to the set of locally equilibrium subsystems that make up the non-equilibrium system: when real (irreversible) processes occur, the non-equilibrium of the isolated system (IS) decreases, and in reversible processes the non-equilibrium in the system of locally equilibrium subsystems does not change (the increment of one kind of non-equilibrium completely compensated by a decrease in the disequilibrium of some other kind).

As a quantitative characteristic (measure) of the non-equilibrium of the system, the maximum work that can be performed during the transition of the non-equilibrium system to the equilibrium state is considered. To calculate this work, the following quantities are used: “potential difference,” the “entropic difference, and change in exergy. All these quantities decrease during the course of real (irreversible) processes in the IS, and do not change during the course of reversible (ideal) processes. As a result, a generalized expression of the SLT was obtained through the quantitative characteristics of the non-equilibrium of the system in the form of an inequality, including the R. Clausius inequality for the change in the entropy of an isolated system.

This chapter provides a calculated confirmation of the theoretical positions of the concept of non-equilibrium and its mathematical apparatus using examples of determining the loss of non-equilibrium of adiabatic and isolated systems.

Keywords: second law of thermodynamics, postulate of non-equilibrium, new formulation second law, new recording second law, calculation of non-equilibrium, entropy

*If we can appreciate non-equilibrium of the entire isolated system,
then we determine the direction of time.*

- F. Rafe

1. INTRODUCTION

All laws of nature are based on the philosophical notions of matter and its property - motion. Based on these concepts, the philosophical laws of conservation of matter and its property of motion were formulated. To

quantitatively characterize the individual sides of matter and its property of motion, various physical quantities (mass, momentum, angular momentum, energy, charge, etc.) were introduced and the corresponding physical conservation laws were formulated. Philosophical laws are more general than physical laws. For example, the law of conservation of motion is confirmed by various physical laws of conservation: impulse, moment of impulse, energy. The first quantitative characteristic of ordered motion (physical quantity, PQ) was introduced by Rene Descartes in the form of the product of mass and speed, which he called “amount of motion.” Then another quantitative characteristic of motion was introduced - energy.

As a result, physicists began to use two different quantities as the stock of motion (amount of motion) of the system: Descartes’ “amount of motion” and energy. Therefore, a new term was proposed for the product of mass and speed - momentum (impulse). Since energy (PQ) quantitatively characterizes both ordered and chaotic motion, this quantity has become universal. Currently, the term “energy” is understood as the physical quantity E , J , and motion itself (an attribute of matter), that is, this term has a categorical ambiguity. Now the term “energy” is often used as a synonym for the term “motion,” which makes it difficult to understand the context.

As already noted, all fundamental laws of physics are based on the philosophical law of conservation of matter and its property of motion. Thus, the first law of thermodynamics (FLT) is an analytical (quantitative) expression of the law of conservation of motion when its form changes, written through a physical quantity – energy. As for the second law of thermodynamics (SLT), it has not been determined so far which property of matter (motion, form of motion or any other property) is quantitatively conserved in the isolated system (IS) when reversible (ideal) processes occur and changed when irreversible (real) processes occur.

The analytical expression of the second law of thermodynamics is the inequality of Rudolf Clausius (1850)

$$\Delta S_{TC} > 0.$$

noneq.IS •

(1)

According to this inequality, the entropy of a non-equilibrium isolated system S_{noneq} in irreversible processes increases, while in reversible processes it preserves. The question arises, which property of matter (motion, form of motion, or other property) is quantitatively preserved in the isolate system during the course of reversible (ideal) processes. The answer to this question modern thermodynamics does not.

Lack of knowledge (misunderstanding) of the essence of the SLT led to various formulations (about twenty formulations of the SLT were proposed (Putilov 1971)) and an enormous amount of researches on explaining the meaning of these formulations (Bejan 2016; Ben-Naim 2010; Chen et al. 2017; D'Alessio et al. 2016; de Blasio 2019; Marboeuf et al. 2019; Martyushev and Seleznev 2006; Ptaszynski and Esposito 2019; Rakopoulos and Giakoumis 2006; Reyf 1972; Sellitto and Di Domenico 2019). As Moran notes, neither wording fully reflects all aspects of the SLT given by other formulations. Many authors (Moran, et al. 2014) see the reason for the lack of a uniform formulation of the SLT as the diversity of thematic areas covered by the second law.

When explaining the SLT formulations, contradictions arise both within separate textbooks and textbooks of different authors: Andryushchenko 1975; Baehr H. D. 1973; Borgnakke and Sonntag 2009; Calabrese 2018; Callen 1985; Fermi 1937; Hoiyst and Poniewierski 2012; Kirillin et al. 1983; Kittel 1993; Kondepudi and Prigogine 2015; Krichevsky 1970; Krutov et al. 1991; Lavenda 2010; Morales-Rodriguez 2016; Moran et al. 2014; Novikov 1984; Srivastava et al. 2007; Vukalovich and Novikov 1968. These formulations indirectly describe the consequences arising from the SLT, or are redundant (for example, the approval of heat removal in the cycle). We need a single formulation (as is customary in other laws) from which all other formulations should follow as a consequence. If there is a unified the SLT formulation, the need for many formulations will become redundant.

Certain objections raise statements of the type: “a reversible process can proceed only in fully equilibrium systems,” that is, when there are no processes; “The irreversible process cannot be represented graphically,” although the same authors use the corresponding diagrams to depict the

change in the state of the gas as it flows with friction in the channel; “Heat cannot be completely transformed into work, but work can,” although it is known that in the isothermal process of expanding an ideal gas, the heat supplied is equal to the work done; “Equilibrium heat transfer in a thermally non-equilibrium system using an ideal heat engine” and many others.

The “existence” of entropy is proved (postulated) as the first part of the second law. Since entropy is a physical quantity (a named number), it must be derived through basic quantities using the appropriate coupling equations, and not postulated or proven.

The question arises: is the entropy the only characteristic of an irreversible change in the state of a non-equilibrium isolated system, that is, characterizes the direction of the processes in a non-equilibrium isolated system? Along with the entropy in setting forth the second law of thermodynamics, such quantities as thermodynamic potentials, maximum system work, exergy are introduced. These quantities have so far not been united by the general idea of the second law of thermodynamics. There is a need to develop a unified concept of presentation of the second law of thermodynamics, which could combine these quantities in to a single theory. Consider how modern thermodynamics answers this and other questions. This chapter is based on the work of the author Ryndin 2019.

2. REVIEW OF THE FORMULATIONS (POSTULATES) OF THE SECOND LAW OF THERMODYNAMICS

2.1. Clausius’ Postulate

The second law of thermodynamics is perhaps the most complicated and confusing, and in any case, the discussion issue of thermodynamics, to which much attention is paid both in textbooks on thermodynamics and in special articles (Avellaneda, J. M., Bataille, F., Toutant, A. 2019; Ben-Naim 2010; Birjukov, Jahnke, Mahler 2008; Darrigol and Renn 2017;

Since the non-equilibrium of the IS does not change, the change in the entropy of the IS is also equal to zero when a reversible heat transfer process occur

$$\Delta S_{\text{NIS}} = \Delta S_{\text{HB}} + \Delta S_{\text{CB}} + \Delta S_{\text{WB}} + \Delta S_{\text{WR}} = Q_{\text{HB}}/T_1 + Q_{\text{CB}}/T_2 = -Q_1/T_1 + Q_2/T_2 = -Q_1/T_1 + (Q_1 - W_{\text{CC}})/T_2 = -Q_1/T_1 + (Q_1 \frac{T_2}{T_1}) \frac{1}{T_2} = 0$$

where the change in the entropy of the work body ΔS_{WB} and the work receiver ΔS_{WR} are equal to zero, since the work body makes a cycle, and the work receiver does not contain chaotic motion, characterized by temperature, but only ordered motion, which is not characterized by temperature and entropy.

CONCLUSION

The main provisions of the non-equilibrium concept underlying the second law of thermodynamics can be formulated in the following theses:

1. The concept of “non-equilibrium of the system” is introduced as a property, a state of matter, due to the unequal distribution of motion in space, as well as the ability (property) of the system to do work (create ordered motion).
2. Depending on the form, type of motion (type of energy, as a synonym for motion), the following types of non-equilibrium can be distinguished: mechanical (kinetic and potential, the latter can be gravitational and elastic); thermal, baric, electrical, chemical, diffusion (concentration), etc.
3. The reason for the flow of processes (the transfer of motion in space) is the non-equilibrium of the system (the inhomogeneity of space), consisting of a set of locally-equilibrium subsystems

(infinitesimally small or finite sizes). In an equilibrium isolated system, no processes (including reversible ones) can occur.

4. All spontaneous (natural) processes proceed in the direction of reducing the non-equilibrium of a given type, that is, the nonequilibrium due to which this process is carried out. For example, heat exchange (it should not be confused with carryover of heat (of chaotic motion) using a refrigeration machine, which is not a natural process) occurs due to thermal non-equilibrium, which decreases in a spontaneous (natural) heat transfer process: a hot body cools down, and a cold body heats up.
5. A spontaneous growth of non-equilibrium of any kind in the absence of any other kind of non-equilibrium in the system or when they remain unchanged is impossible. For example, the growth of thermal non-equilibrium between HB and CB is impossible by only heat exchange between these bodies, since heat exchange is a natural process, and the natural process can proceed only in the direction of reducing the non-equilibrium that caused this process (in this case, thermal non-equilibrium).
6. The *amount of non-equilibrium (non-equilibrium)* is understood as the maximum work that the adiabatic system could transfer to the environment during its transition to an equilibrium state $\Lambda \equiv \Lambda_{\text{NAS}} \equiv W_{\text{noneq.st.} \rightarrow \text{eq.st.}}^{\text{max}}$, or the maximum lost (possible, available) work inside the isolated system during its transition to an equilibrium state $\Lambda \equiv \Lambda_{\text{NIS}} \equiv W_{\text{noneq.st.} \rightarrow \text{eq.ct.}}^{\text{max}} = W_{\text{lost}}^{\text{max}}$.

The maximum lost work can be determined through the maximum change in the entropy of the IS during its transition to an equilibrium state, or through the entropically difference according to the Guy-Stodola formula

$$\Lambda \equiv \Lambda_{\text{NIS}} \equiv W_{\text{noneq.st.} \rightarrow \text{eq.st.}}^{\text{max}} = W_{\text{lost}}^{\text{max}} = T_{\text{min}} \Delta S_{\text{noneq.st.} \rightarrow \text{eq.st.}}^{\text{max}} = T_{\text{min}} \Delta S^*$$

where $\Delta S^* = S_{\text{eq.st.IS}} - S_{\text{NIS}}$ is the entropically difference, defined as the difference between the entropies of the IS in equilibrium and non-equilibrium states; T_{\min} is the minimum constant temperature of a part of the IS (liquid AM is an ocean, atmosphere, or a massive body).

The non-equilibrium of the AS (maximum work) can be determined by the maximum loss of the total thermodynamic potential or by the potential difference

$$\Lambda_{\text{NAS}} \equiv W_{\text{NAS} \rightarrow \text{eq.st.}}^{\max} = -\Delta \Pi_{\text{NAS} \rightarrow \text{eq.st.}}^{\max} = \Delta \Pi^*$$

where $\Delta \Pi^* = \Pi_{\text{noneq.st.}} - \Pi_{\text{eq.st.}} = W_{\text{noneq.st.} \rightarrow \text{eq.st.}}^{\max}$ is the potential difference, defined as the potential difference of the AS in non-equilibrium and equilibrium states.

The loss of thermal non-equilibrium during transfer of heat in the amount of Q_1 in the temperature range from T_1 to $T_{\text{AM}} = T_0 = \text{const}$ is determined through the exergy of heat, which is understood as the work of an ideal Carnot cycle

$$-\Delta \Lambda_{\text{therm}} = W_{\text{CC}} = Q_1 (1 - T_{\text{AM}}/T_1) = Q_1 - T_{\text{AM}} \Delta S_{\text{AM}}^0 = E_Q$$

The non-equilibrium of a system consisting of a flow element of a unit mass and an ambient medium of constant parameters (AM) is determined by the exergy of the flow, which is understood as the maximum specific work obtained from a flow element in a turbine upon expansion to the AM parameters

$$\lambda_{\text{WB-AM}} = e_x = h - h_0 + T_0 (s_0 - s) = w_{\text{turb}}^{\max}$$

7. Processes in which an increase in the non-equilibrium of a given type is compensated by a decrease in the non-equilibrium of some

other type so that in a system of interacting bodies the complete (total) non-equilibrium is preserved are called ideal or reversible.

For example, a decrease in thermal non-equilibrium in the case of reversible heat transfer from the HB to the CB is compensated by an increase in mechanical (or electrical, chemical) non-equilibrium due to an increase in the KE of the flywheel of the engine performing the CC (or due to an increase in the voltage of the battery charged by the generator driven by the engine shaft).

In the case of a reversible process of falling of a body in a vacuum, a decrease in the potential non-equilibrium of the body-Earth system, characterized by the potential energy of the body relative to the Earth, is compensated by an increase in kinetic disequilibrium, characterized by the kinetic energy of the body relative to the Earth.

Thus, in the presence of special devices, reversible processes can also occur in purely non-equilibrium systems, and not only in systems close to the state of equilibrium, as is commonly believed.

8. In a reversible process, complete non-equilibrium is maintained for any combination of partial non-equilibrium constituents of complete non-equilibrium. For example, in a ball-spring system, total non-equilibrium can be equal to kinetic or potential nonequilibrium, or it may consist in part of the kinetic non-equilibrium of the ball and the potential non-equilibrium of the spring.

The law of conservation of complete non-equilibrium in a reversible process for any combination of partial non-equilibrium agrees with the law of conservation of total mechanical energy for any combination of kinetic and potential energies included in the total mechanical energy.

In other words, the law of conservation of mechanical energy is an analytical (quantitative) expression of the law of conservation of complete mechanical non-equilibrium (philosophical category) in a system of interacting bodies.

9. The growth of the non-equilibrium of a given species is possible only due to a decrease in the non-equilibrium of any other species.

For example, an increase in thermal non-equilibrium (transfer of heat (chaotic motion) from HB to CB) is possible only due to an equivalent (or greater) decrease in mechanical non-equilibrium - the kinetic energy of the flywheel that drives the refrigeration machine.

10. The second law of thermodynamics is the law of a decrease in the complete non-equilibrium of an isolated system: all real processes proceed in such a way that the complete non-equilibrium in a system of several interacting bodies decreases (while individual non-equilibriums may increase) and upon reaching full equilibrium (zero non-equilibrium), all real processes cease - the reverse transition of an isolated system from an equilibrium state to a nonequilibrium state is impossible.
11. The following types of inequalities are analytical expressions of the principle (postulate) of non-equilibrium (complete nonequilibrium of an isolated system cannot increase), which is the basis of the second law of thermodynamics:

$$d\Lambda_{IS} = \sum d\Lambda_i \leq 0, \quad d(\Delta S^*) = -dS_{NIS} \leq 0 \quad \text{or} \quad dS_{NIS} = -d(\Delta S^*) \geq 0.$$

According to the SLT, as the isolated system approaches the equilibrium state, its entropy increases (the heat of dissipation increases) in irreversible processes and is preserved in reversible processes.

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