Doctoral Dissertation (Summary)

博士論文(要約)

Maximum efficiency of heat engines revisited from stochastic thermodynamics

(熱機関の最大効率再訪)

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December 2016

Abstract

The efficiency of heat engines is one of the central topics in thermodynamics. Around 200 years ago, Carnot [1] revealed that the efficiency η of a cyclic process with two heat baths with temperatures T_{H} and T_{L} ($T_{\mathrm{H}} > T_{\mathrm{L}}$) is bounded by a universal function depending only on the two temperatures. Using this fact, Joule and Thomson (Load Kelvin) [2] introduced the absolute temperature and expressed the universal bound for efficiency as

$$\eta \leq \eta_{\mathrm{C}} := 1 - \frac{T_{\mathrm{L}}}{T_{\mathrm{H}}}. \tag{1}$$

The maximum efficiency η_{C} is called *Carnot efficiency.*

Carnot also demonstrated that the upper bound is attainable with quasistatic external operations. However, Carnot did not study the attainability of the maximum efficiency with other types of operations. Therefore, the following two questions naturally arise:

- Suppose a system not operated externally but working automatically (i.e., stationary state). Does such a system attain the Carnot efficiency? and if the answer is yes, when?
- Suppose a system not in quasistatic limit but with finite power. Does such a system attain the Carnot efficiency?

In this Thesis, we answer these two questions with help of the stochastic thermodynamics, which has developed in the last two decades and mainly treats small stochastic systems. To say a result, the answer of the first question is positive, while the necessary condition for autonomous engines to attain the Carnot efficiency is very hard to satisfy in physical setups. We clarify this necessary condition for both small stochastic systems and macroscopic systems, which are quite different from each other. On the second question, we settle in negative the problem by deriving a universal trade-off relation between efficiency and power. Our result is applicable to almost all engines including both small stochastic ones and macroscopic ones. These two results are notable products of stochastic thermodynamics to macroscopic phenomenology, and will lead to qualitative improvement of thermodynamics.

Chapter 1

Background

1.1 Aim of stochastic thermodynamics

Stochastic thermodynamics is an extended form of thermodynamics to small fluctuating systems such as Brownian particles, molecular motors in living systems, and mesoscopic quantum dots. One important achievement of stochastic thermodynamics is to establish how to define thermodynamic quantities (e.g., heat, work, and entropy) in stochastic systems and what relations (e.g., first and second law of thermodynamics) hold among them, which is highly nontrivial tasks from the standpoint of conventional macroscopic thermodynamics and statistical mechanics. Stochastic thermodynamics also provides novel relations including the celebrated fluctuation theorem (FT) and related relations, which reveal a beautiful symmetric structure in nonequilibrium fluctuations. For this reason, the stochastic thermodynamics has attracted interest of many physicist, in particular in nonequilibrium statistical mechanics, in the last two decades.

It is noteworthy that the proof of the FT is very simple and straightforward, which is both good and bad news. The simplicity has facilitated many researchers to derive novel nonequilibrium FT-type relations, which is a good news. In contrast, most of such relations are products of mathematical calculation and lacking physical meaning, which is a bad news. Some researchers even criticize that the FT (and many FT-type equalities) is only a mathematical equality and physically almost useless. Although I think this criticism overclaiming, it is pertinent on the aspect of disrespect of our basic motivation from physics. We should recall what is the aim of stochastic thermodynamics.

Stochastic thermodynamics lies on at least three aims and motivations.

First is based on observations of biochemical systems such as molecular motors. From the viewpoint of thermodynamics and statistical mechanics, molecular motors can be regarded as engines converting chemical potential of such as ATP to mechanical force. However, molecular motors apparently differ from conventional heat engines with many aspects: Molecular motors are so small that they suffice thermal fluctuation, which sometimes disturbs their motion but sometimes plays a crucial role in their motion. In contrast, conventional heat engines do not fluctuate. Molecular motors work automatically under fluctuation, while conventional heat engines are operated externally and deterministically. In addition, scrutinized experiments discovered the high efficiency of many molecular motors as F1-ATPase [3, 4], kinesins [5], and myosins [6] compared to our electric plant[1]. Therefore, why and how molecular motors achieve high efficiency and what is the significance of molecular motors different from conventional heat engines are important problems for nonequilibrium statistical mechanics.

Second is motivated by a theoretical interest to understand what structure exists behind thermodynamics. One of the central problems in nonequilibrium statistical mechanics is the foundation of thermodynamic irreversibility from microscopic viewpoint. The problem of how the arrow of time appears from microscopic reversible dynamics[2] has been intensively debated since Boltzmann. Aside from this, several relations in nonequilibrium statistical mechanics employ both thermodynamics and microscopic dynamics. For example, the fluctuation-dissipation relation is first derived by using the consistency with the second law of thermodynamics [7], and later derived from microscopic dynamics [8, 9]. The original derivation of the Onsager reciprocity theorem [10] consists of the combination of thermodynamic phenomenology and reversibility of microscopic dynamics. Although all these relations are established ones, it is a fruitful task to clarify what aspect of the structure of thermodynamics and nonequilibrium statistical mechanics is reflected in these relations.

Third is to provide novel relations or laws on macroscopic quantities. One of the most important role of physics is to establish novel universal laws. The aforementioned three relations—the second law of thermodynamics, fluctuation-dissipation theorem, and the Onsager reciprocity theorem—are established macroscopic laws. However, known macroscopic laws are only these three[3]. It is strongly desired to find and establish novel macroscopic laws.

These aims are summarized as follows[4]:

1. Understanding the significance of molecular motors.
2. Clarifying the structure of thermodynamics.
3. Establishing novel macroscopic laws.

Existing stochastic thermodynamics has partially achieved these three aims.

1. At the first stage, some people even consider that small stochastic systems may escape from the restriction of conventional macroscopic thermodynamics, which is shown in many proposals of the perpetual motion machine of the second type. However, Sekimoto [11] formulated stochastic energetics, that is, a thermodynamic framework for small stochastic systems, which ensures that molecular motors are not different from conventional heat engines on

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[1] The efficiencies of molecular motors are around 0.7∼0.9, while those of electric plants are usually less than 0.5.

[2] Both Hamilton dynamics and unitary evolution are reversible time differential equation.

[3] We here exclude hydrodynamic relations.

[4] We remark that the second and third aims partially treat common problems.

this aspect. In addition, many ratchet models [12] and the Curie principle[5] [11, 13] confirm that one directional motion of molecular motors in stochastic environment is not so surprising.

Some researchers including biophysicists argued that the speciality of molecular motors is seen in the usage of information [14]. Molecular motors consist of several subsystems, and experimental observation [14] suggests that one of subsystem behaves as if it measures another subsystem and changes its motion depending on the measurement outcome, which is a kind of information processes[6]. This interpretation matches the intuitive picture that molecular motors work by utilizing thermal fluctuation. Information thermodynamics, which hybrids stochastic thermodynamics and information theory, is expected to describe molecular motors in terms of information theory. However unfortunately, the Sagawa-Ueda relation [15–17], which is a prominent result of information thermodynamics explaining the role of information at the stochastic level, is not applicable to molecular motors.

2. The fluctuation theorem (FT) [18–22] provides a clear understanding of thermodynamic irreversibility. It sheds light on the importance of the detailed-balance condition and microscopic reversibility. In addition,existing macroscopic laws are reproduced by the fluctuation theorem, which clarified the relation among them [23, 24].

   It has been well known that the second law of thermodynamics and other relations are violated with information processes [25, 26], which is called Maxwell's demon problem. Information thermodynamics established how to construct thermodynamics for each subsystem with information processes by introducing the notion of additive decomposition. This reveals that the fundamental building block of thermodynamics is not a system but a subsystem or a more microscopic building block.

3. Few novel macroscopic laws were obtained from stochastic thermodynamics. One exception is the higher-order fluctuation-dissipation theorem [27], which connects the higher-order response and the response of the correlation function[7]. This relation is experimentally tested with a mesoscopic conductor[8] [28].

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[5] The Curie principle claims that a one-directional motion naturally occurs without any special symmetry or restrictions.

[6] In the case of F1-ATPase, the $\alpha^3\beta^3$ unit surrounds the γ shaft. The $\alpha^3\beta^3$ unit has three stable states, which play as three different potential landscapes of the γ shaft. The experiment [14] reports that the $\alpha^3\beta^3$ unit changes its state as if it measures the state of the γ shaft and performs feedback control depending on the measurement outcome.

[7] We however, note that these quantities are too small to measure in macroscopic objects.

[8] While the theoretical prediction is qualitatively verified, the experimental result quantitatively differs from theoretical prediction.

	Aim 1 (molecular motors)	Aim 2 (thermodynamic structure)	Aim 3 (novel macroscopic relations)
A (Part.II)	Clarifying the role of information	Establishing fundamental building blocks	
B (Part.III)	Clarifying the role of autonomy		Deriving necessary condition to attain the Carnot efficiency
C (Part.IV)		Elucidating the role of speed of processes	Deriving trade-off relation between efficiency and power

Table 1.1: Structure of this Thesis. Three rows represent three parts of this Thesis, in which the topics in the row are treated. Three columns represent three problems or aims of stochastic thermodynamics.

1.2 Problem setting

On the basis of the above three aims, we investigate the following problems in this Thesis.

A. From the aims of 1 and 2, we shall determine the most fundamental building block of thermodynamics. We establish a decomposition of the entropy increase of a total system into a single transition with keeping thermodynamic properties individually. Employing this, we accomplish information thermodynamics for general information processes including those in molecular motors.

B. From the aims of 1 and 3, we shall elucidate autonomous engines or stationary free energy transducers. One significance of molecular motors is their autonomy, which may impose novel constraints on them. We derive the necessary condition for autonomous engines to attain the Carnot efficiency, which covers general stationary systems from molecular motors to macroscopic thermoelectric conductors. The necessary condition suggests a new insight into the property of molecular motors.

C. From the aim of 2 and 3, we shall establish the trade-off relation between power and efficiency of engines. Conventional macroscopic thermodynamics fails to take the notion of speed into account. We prove a general trade-off relation between power and efficiency which holds for both small and macroscopic engines. As its corollary, we show a no-go theorem that a finite power engine never attains the Carnot efficiency, which settles in negative a longstanding open problem since Carnot.

1.3 Outline of Thesis

This Thesis consists of three main parts. In Part.II, we study the foundation of stochastic thermodynamics from basic results (e.g., stochastic processes, Ito-Stratonovich dilemma, fluctuation theorem) to information thermodynamics and beyond.

In Chap. 2, we review basic notions of stochastic processes and stochastic differential equations, which serve as a language for describing stochastic thermodynamics. Jump processes and stochastic differential equations are explained in Sec. 2.2 and 2.3, respectively.

In Chap. 3, we review the achievements of stochastic thermodynamics in the last two decades. We first explain stochastic energetics in Sec. 3.1, which establishes how thermodynamic structure is constructed on Langevin dynamics. In Sec. 3.2, we explain in detail the celebrated fluctuation theorem, which is the most important result in stochastic thermodynamics. We also demonstrate how the fluctuation theorem reproduces existing thermodynamic relations in Sec. 3.3. We then describe three important results of stochastic thermodynamics (except information thermodynamics, which is explained in the next chapter). In Sec. 3.4, we explain the Hatano-Sasa relation, which can be regarded as a generalized form of the fluctuation theorem and also serves as a pioneering work for steady state thermodynamics. In Sec. 3.5, we introduce the path integral method for Langevin dynamics, which reproduces the fluctuation theorem and the Hatano-Sasa relation, and also provides a novel relation, the Harada-Sasa relation. The Harada-Sasa relation relates stationary heat dissipation to the violation of the fluctuation-dissipation theorem in a stationary system. In Sec. 3.6, we treat systems with coarse-graining of quick variables. While the entropy production is preserved through coarse-graining in equilibrium case, it generally decreases in non-equilibrium case.

In Chap. 4, we review information thermodynamics. Before its explanation, we first see the history of debates on Maxwell's demon problem in Sec. 4.1. We then introduce the Sagawa-Ueda relation in Sec. 4.2, which clarifies the role of information in stochastic thermodynamics and solves the problem of Maxwell's demon. Theoretically speaking, the Sagawa-Ueda relation provides the additive decomposition of entropy production into subsystems, which we discuss in Sec. 4.3. From the perspective of information thermodynamics, we again overview the previous studies on Maxwell's demon in Sec. 4.4. Different from common beliefs, we argue that the insight by Szilard is in the most correct way and that the memory erasure is not crucial to understand the Maxwell's demon. In closing this chapter, in Sec. 4.5 we remark the limitation of the Sagawa-Ueda relation, where it is not applicable to general information processes including molecular motors.

In Chap. 5, we describe my first original result on partial entropy production. We further investigate the idea of additive decomposition and establish the most fundamental bulding block of stochastic thermodynamics. In Sec. 5.1, we introduce the fundamental bulding block named *partial entropy production* and confirm its thermodynamic properties. In Sec. 5.2, by applying the method of partial entropy production, we solve some problems including the characterization of general information processes.

In Part. III, we consider autonomous engines, or stationary free energy transducers, which cover from molecular motors, Feynman's ratchet to thermoelectric transport. We elucidate the attainability of the Carnot efficiency (CE) for autonomous engines. A cyclic process in conventional thermodynamics with an external deterministic operation attains the CE in the quasistatic limit. In contrast, all variables in autonomous engines inevitably fluctuate, which plays a crucial role in particular in the quasistatic limit. Thus, the attainability of the CE for autonomous engines is highly nontrivial, and in fact we reveal that autonomous engines attain the CE only by satisfying severe conditions.

In Chap. 6, we review some previous results of the attainability of the CE. Most of previous papers treat specific models. In Sec. 6.1, we see the debate on Feynman's ratchet and the Büttiker-Landauer model, which were first considered to attain the CE but are now revealed not to. In Sec. 6.2, we briefly review other autonomous models which attain the CE. We then explain the tight-coupling condition in Sec. 6.3, which is a scarce general result on the attainability of the CE.

In Chap. 7, we describe my second original result on the attainability of the CE in autonomous engines. Before going to general analyses, in Sec. 7.1, we first introduce a model of a macroscopic autonomous engine converting chemical potential difference (e.g., difference of density) into mechanical work, which exhibits the significance of autonomous engines. We demonstrate that while the engine does not attain the CE with finite chemical potential difference in normal setup, it indeed attains the CE with singular transition rates. In Sec. 7.2, we derive the general necessary condition for autonomous engines to attain the CE. We prove that a certain type of singularity is inevitable to attain the CE as suggested in the previous section. We then clarify the difference between engines of finite size and that in thermodynamic limit by introducing the viewpoint of *tight-coupling window*. We also connect the tight-couping condition to the attainability of the CE beyond the linear response regime.

In Part. IV, we consider finite power engines or processes with finite speed. While all real engines work with finite speed, conventional thermodynamics does not take the notion of speed into account and existing nonequilibrium statistical mechanics provides few general results on finite power engines. In intuition, an engine with large power inevitably accompanies much loss, which implies less efficient. However, previous studies treat only specific situations (e.g., linear response regime) or models (e.g., overdamped Langevin equation) and with additional assumptions (e.g., without parity-odd variables and fields[9]), and general results have been lacked. In contrast, we derive the trade-off relation between power and efficiency for general engines, employing a method recently developed in stochastic thermodynamics.

In Chap. 8, we review some existing results on the relation between power and efficiency. In Sec. 8.1, we explain the studies of the efficiency at maximum power,

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[9] Momentum and magnetic field are well-known example.

which is known to take the half of the CE in the linear response regime. We show this result in three different setups. In Sec. 8.2. we see the longstanding open problem whether a finite power engine attains the CE. We first ensure that the problem is not solved with established general frameworks, that is, conventional thermodynamics and linear irreversible thermodynamics. We then briefly demonstrate that all known concrete models mainly in the linear response regime deny this possibility.

In Chap. 9, we describe my third original result on the trade-off relation between power and efficiency for Markovian engines. In Sec. 9.2, we derive our main inequality on entropy production and heat current between the system and a bath. This leads to a trade-odd relation between efficiency and power of cyclic heat engines, which prohibits a finite power heat engine with the CE as its corollary. The inequality contains a newly introduced quantity Θ, whose properties we discuss in Sec. 9.3. Although our result generally prohibits a finite power heat engine with the CE as long as they are classical and Markovian, some proposals claim the possibility of the realization. In Sec. 9.4, we clarify why these proposals are indeed not counterexamples of our result. We finally remark its extensions to the case with other kinds of current (Sec. 9.5.2) and the quantum case (Sec. 9.5.3).

In Chap. 10, we describe my fourth original result on the trade-off relation between speed of the process and efficiency for quantum non-Markovian engines. Before stating our result, we first give some definitions on a lattice and a Hamiltonian in Sec. 10.1. In Sec. 10.2, we introduce a key relation, the Lieb-Robinson bound, which evaluates the correlation between two different space-time points in quantum non-relativistic systems. Applying its time-dependent version to locally operated systems, we find an indishtinguishability inequality which describes the following fact: One cannot distinguish what operation is done by observables acting on a region far from the operated region within short time interval. In Sec. 10.3, applying this inequality to a cyclic process with two heat baths, we derive an inequality on efficiency in which the efficiency is strictly less than the CE with finite cyclic time interval. In Sec. 10.4, we show other applications of the indistinguishability inequality; relations on transient processes (Sec. 10.4.1) and adiabatic processes (Sec. 10.4.2).

This Thesis is based on my original papers [29–36]. Although some of my papers and partially [37–39] are related, I do not mainly treat these results. My other papers [40,41] are not treated in this Thesis because these topics are different. My original results appear in Chap, 5, 7, 9, and 10 [10].

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[10] We note that the results in Chap. 5 except Sec. 5.2.2, 5.3.1, 5.3.2 have already appeared in my Master thesis, while the results in Sec. 5.2.2, 5.3.1, 5.3.2 first appear in this Thesis.

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