



# Entropy Decrease in Isolated Systems: Theory, Fact and Tests

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## ABSTRACT

First, we emphasize that preconditions of entropy increase are 1) for isolated systems; 2) various internal interactions in system must be neglected; 3) they must be thermal equilibrium processes. We proposed possible entropy decrease due to fluctuation magnified and internal interactions in isolated systems, and research various possible entropy decreases in physics, which include phase transformation from disorder uniformity to order state. Next, the solidification forms spontaneously an order structure, and it may be process of entropy decrease. Third, we propose that entropy decrease exists necessarily in self-assembly as isolated system. Fourth, we discuss the molecular motor and entropy decreases in biology. Fifth, we research entropy decrease in astronomy and propose quantitatively a total formula of entropy change for universal evolution of any natural and social systems. As long as we break through the bondage of the second law of thermodynamics, the rich and complex world is full of examples of entropy decrease.

**Keywords:** Entropy, Internal interaction, Isolated system, Condensed matter, Self-assembly, Solidification, Molecular motor, Biology, Astronomy.

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## INTRODUCTION

There is no doubt that the second law of thermodynamics is a great contribution to the development of science, and applied widely many aspects. However, the basis of thermodynamics is the statistics, in which a basic principle is statistical independence: The state of one subsystem does not affect the probabilities of various states of the other subsystems, because different subsystems may be regarded as weakly interacting (Landau, Lifšic, Lifshitz, & Pitaevskii, 1980). It shows that various interactions among these subsystems are not considered. Such preconditions of entropy increase are: 1) for isolated systems, which are only approximation; 2) various

internal interactions in system must be neglected; 3) they must be thermal equilibrium processes, thereby no phase transition. However, if these premises are regardless and unlimited extension to all the process in spite of its preconditions, it will be metamorphosed into belief divorced from reality and violated science. For example, all systems in Nature will tend to “heat death” (Rifkin, 1980), the second law of thermodynamics prescribes entropy increase in all processes (Hokikian, Planck, & Grant), etc. In particular, it is casual in application of social science, these are all entropy increase either a big group is formed, and a molecule is decomposed (Hokikian et al.).

The entropy increases as a new worldview (Rifkin, 1980) introduce already some disputes, even misunderstand.

If various internal complex mechanism and interactions cannot be neglected, a state with smaller entropy (for example, self-organized structure) will be able to appear under some conditions. We proposed that if interactions, fluctuations and their magnified exist among various subsystems of an isolated system, entropy decrease in the isolated system is possible (Chang, 1994, 2005), which includes physics (Chang, 2013c, 2015b), chemistry (Chang, 2014; Chang et al., 2013), biology (Chang, 2006a, 2013b), astronomy (Chang, 2013a, 2018) and social sciences (Chang, 2013c, 2015b). For attractive process, internal energy, system entropy, and nonlinear interactions, etc., an isolated system may form a self-organized structure with lower entropy. Some possible entropy decreases are calculated quantitatively (Chang, 2005, 2015a). In this paper, we research various possible entropy decreases in general physics, which include phase transformation from disorder uniformity to order state, and in self-assembly entropy, decrease exists necessarily.

### POSSIBLE ENTROPY DECREASE IN PHYSICS

A well-known development of thermodynamics is the theory of dissipative structure proposed by Prigogine. We researched possible entropy decrease in isolated system for various complex systems (Chang, 1994, 2015b), and proposed a universal formula for any isolated system (Chang, 2005),

$$dS = dS^a + dS^i \quad (1)$$

It is symmetry with the formula

$$dS = d_i S + d_e S \quad (2)$$

In the theory of dissipative structure. From this, we derived a complete symmetrical structure on change of entropy

$$\text{Entropy} \rightarrow \begin{cases} \text{increase.} \\ \text{decrease} \rightarrow \begin{cases} dS = d_i S + d_e S. \\ dS = dS^a + dS^i. \end{cases} \end{cases} \quad (3)$$

Here entropy decrease may be the dissipative structure for an open system, or be the internal interactions for an isolated system (Chang, 2015b).

A classic example on entropy increase as belief is negative temperature, which as a measurement of molecular motions, how can it be negative? We studied carefully and found that negative temperature is contradiction with usual meaning of temperature and with some basic concepts of physics and mathematics, and derives necessarily entropy decrease  $dS < 0$  (Berillo, Mattiasson, Galaev, & Kirsebom, 2012) (Fig.1).

For example, “at  $T = 0$ , the system is in its lower quantum state, and its entropy is zero” (Landau et al., 1980). In fact, the temperature  $T = 0$  (absolute zero) is impossibly achieved. “As the temperature increases, the energy and entropy of the system increase monotonically. At  $T = \infty$ , the energy is  $\bar{E}_n$  and the entropy reaches its maximum value  $N \ln g$ ; these values correspond to a distribution with equal probability over all quantum states of the system, which is the limit of the Gibbs distribution as  $T \rightarrow \infty$ .” The statement of original negative

temperature is based on the two premises: entropy of the system increase monotonically, and the Gibbs distribution holds. From this, some strange arguments are obtained:

(a) “The temperature  $T = -\infty$  is physically identical with  $T = \infty$ ; the two values give the same distribution and the same values of the thermodynamic quantities for the system” (Landau et al., 1980). According to the general definition, temperature cannot be infinite, since the quantity of heat or molecular motion, not all can be infinite. Negative temperature, even negative infinite temperature is stranger. In the same book (Landau et al., 1980), Landau proved a very important result that the temperature must be positive  $T > 0$ . Moreover,  $T = \infty = -\infty$  is excluded by mathematics, which should be maximum entropy  $N \ln g$ . (b) “A further increase in the energy of the system corresponds to an increase in the temperature from  $T = \infty$ ”, and “the entropy decreases monotonically”, i.e.,  $dS < 0$ . (c) “At  $T = 0$  – the energy reaches its greatest value and the entropy returns to zero, the system then being in its highest quantum state.” This obeys Nernst’s theorem, but according to the third law of thermodynamics  $T = 0$  is always impossible, and in which the quantity of heat is zero at  $T = 0$ . While at  $T = 0$  – it possesses highest quantum state! We do not know whether  $T = 0 = T = 0$  – holds or not. (d) “The region of negative temperature lies not below absolute zero but above infinity”, i.e., “negative temperatures are higher than positive ones”. In fact, “ $T < 0$ ” should be  $dS < 0$ .

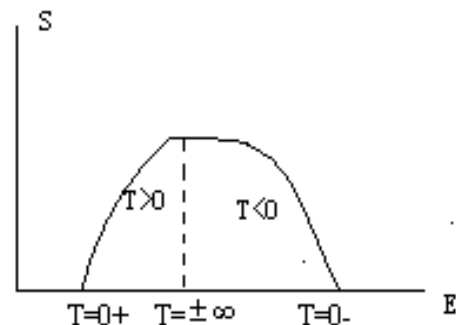


Fig.1. “negative temperature”

(e) Further, according to an efficiency of heat engines

$$\eta = 1 - \frac{T_2}{T_1} \quad (4)$$

Here if either temperature is negative, the efficiency will be greater than unity. “The results arrived at for negative temperatures which are strange to our intuition have no practical significance in the field of energy production.” But, “systems at negative Kelvin temperatures obey the second law and its many corollaries.” Of course, “it would be useless to consume work in order to produce a reservoir at a negative temperature which can be used to operate a very efficient heat engine” (Hatsopoulos, Keenan, & Butler, 1966).

In a word, it is a fallacy in thermodynamics. This seems to imply that negative temperature is introduced only in order to obey the second law of thermodynamics. The above statement of “negative temperature” and Fig.1 prove just that entropy is able to decrease with internal interactions in an isolated system. The experimental study requires that the spin system be well isolated from the lattice system (Ramsey, 1956). This

isolation is possible if the ratio of spin-lattice to spin-spin relaxation times is large (Landau et al., 1980).

This may be described by Fig. 1, which is namely Fig.1 (Ramsey, 1956) and Fig.10 (Landau et al., 1980), in which the finite threshold value  $T_c$  corresponds to only a maximum point  $dS/dE = 0$  (Berillo et al., 2012).

We proposed this possibility exists in transformations of internal energy, complex systems, and some new systems with lower entropy, etc. Self-organization and self-assembly must be entropy decrease. Generally, much structures of Nature all are various self-assembly and self-organizations. We researched some possible tests and predictions on entropy decrease in isolated systems. They include various self-organizations and self-assembly, some critical phenomena, physical surfaces and films, chemical reactions and catalysts, various biological membranes and enzymes, and new synthetic life, etc. They exist possibly in some microscopic regions, nonlinear phenomena, many evolutionary processes and complex systems, etc (Chang, 2019).

The second law of thermodynamics must be thermal equilibrium processes, so, which have not phase transformation. In various phases and phase transformations, only gas diffusion process agrees best with the second law of thermodynamics. For classically isolated gas, entropy is (Reichl, 1980).

$$S(T, V) = S_0 + nR \ln \left[ \left( \frac{T}{T_0} \right)^\alpha \frac{V}{V_0} \right] \quad (5)$$

Therefore, entropy should decrease when the volume becomes smaller or the temperature decreases. Entropy of the ideal gases is

$$S = C_V \ln T + nR \ln V + S_0 \quad (6)$$

For an equal-temperature process  $T = \text{constant}$ ,

$$dS = S_f - S_i = nR \ln(V_f / V_i) \quad (7)$$

This decreases  $dS < 0$ , when  $V_f < V_i$ , i.e., for the attractive process. Conversely, the entropy increases  $dS > 0$ , when  $V_f > V_i$ . The entropy of non-ideal gases is (Landau et al., 1980).

$$S = S_{id} + N \log(1 - Nb/V) \quad (8)$$

This is smaller than one of ideal gases, since  $b$  is four times volume of atom,  $b > 0$ . It corresponds to the existence of interaction of the gas molecules, and average forces between molecules are attractive. The entropy of a solid is (Landau et al., 1980).

$$S = 2\pi^2 VT^3 / 15(\hbar\bar{u})^3 \quad (9)$$

so  $dS < 0$  for  $dT < 0$ . The free energy with the correlation part of plasma is (Landau et al., 1980).

$$F = F_{id} - \frac{2e^3}{3} \frac{\pi^{1/2}}{(VT)^{1/2}} \left( \sum_a N_a z_a^2 \right)^{3/2} \quad (10)$$

Correspondingly, the entropy is

$$dS = S - S_{id} = -\frac{e^3}{3} \frac{\pi^{1/2}}{V^{1/2}} \left( \sum_a N_a z_a^2 \right)^{3/2} T^{-3/2} < 0 \quad (11)$$

There are electric attractive forces between plasma. Generally, entropy decreases for gravitation (Berillo et al., 2012; Chang, 2005). Other, celestial nebula may agglomerate stars, and liquid has the capillary flow.

It is well known that entropy is measurement of disorder in a system. In phase transformation, the crystallization of a supercooling liquid or of a supersaturated solution is surely an ordering process. The cooling principle is entropy reduced. The difference of entropies between the normal state and the superconductive state is

$$dS = S_s - S_n = \frac{H_c V}{4\pi} \frac{\partial H_c}{\partial T}, \quad (12)$$

where  $S_s$  is entropy of the normal state, and  $S_s$  is one of the superconductive state. Such  $dS < 0$  for  $(\partial H_c / \partial T) < 0$ .

A superconducting state is more order than a normal state. This phase transformation from the normal state to the superconducting state is a condensation process. Generally, any condensation process, in which attractive interactions exist, should be one of entropy decrease.

In phase transformation, structures and symmetries are different, corresponding internal energies and entropies are also different. At a critical point of phase transformation, entropy can increase or decrease, i.e., possesses two-direction property. This corresponds to reversibility of transformation between order and disorder. The general phase transformation is an open system. But, if input energy is interrupted at the critical point, it will become an isolated system. Although for any isolated system the gravitational field cannot be screened completely, but the electromagnetic field may be screened completely.

Hwang, et al., investigated the two-dimensional growth of Au on Ru(0001) in the submonolayer range. At room temperature, highly dendritic islands of one layer thickness grow on large Ru terraces. These irregular dendritic islands exhibit a fractal character, and a dimensional analysis yields a fractal dimension  $1.72 \pm 0.07$ , which agrees quantitatively with a two-dimensional diffusion-limited-aggregation (DLA) growth mechanism (Hwang, Schröder, Günther, & Behm, 1991). Brune, et al., studied nucleation on atomic scale for Ag deposition on a Pt(111) surface at low temperature (50-120 K), in which the transition from the initial steps of nucleation to growth and coalescence as a function of temperature (Brune, Röder, Boragno, & Kern, 1994). Röder, et al., studied diffusion-limited aggregation (DLA) of Ag on Pt(111) and Ag(111), and found an extended range of deposition temperature and deposition flux in both metal-on-metal systems fractal growth shapes, and determined the diffusion constants for edge diffusion from a quantitative analysis of the exponential variation of the branch width with temperature

(Röder, Bromann, Brune, & Kern, 1995). Bott, et al., proposed and demonstrated a new approach for the determination of activation energy and attempt frequency for the diffusion of single adatoms on a surface for Pt adatom diffusion on Pt(111). The method involves only a minimum of assumptions and is independent of classical nucleation theory (Bott, Hohage, Morgenstern, Michely, & Comsa, 1996).

Vvedensky, et al., discussed stochastic equations of motion for the surface of a solid that evolves under typical epitaxial growth (Vvedensky, Zangwill, Luse, & Wilby, 1993). For the growth of non-crystalline films, when cooling melt with certain rate from high temperature, its volume  $V$ , entropy  $S$  and enthalpy  $H$  decrease continuously, temperature  $T$  achieves melting point  $T_m$ , and volume, entropy and enthalpy decrease suddenly, material becomes crystal. Then volume, entropy and enthalpy of crystals decrease slowly with temperature. Moreover, in phase transformation there is the supercooling state. In usual physics, the thermodynamic fluctuation do not allow that any two-dimensional crystal exists as stable single under finite temperature. But, in 2004 Novoselov and Geim separated big piece monolayer graphene (Novoselov et al., 2004). It shows a question for thermodynamics. The order structure of graphene may be isolated system. Grapheme has a special interacting Wigner-Seitz radii  $r_s$  (coupling constant).

Experiments show electron interaction in grapheme is weak than one in two dimensional electron gas (Neto, Guinea, Peres, Novoselov, & Geim, 2009). This is an internal interaction. Supersaturated liquid phase aggregates easily film in surface. Liquid after supersaturated should be isolated system, and VdW force is internal interaction.

Phase transformations like chemical reactions are driven by heat fluctuation (Kurz & Fisher, 1998). Lange discussed chemical solution routes to single-crystal thin films (Lange, 1996). It introduces solution with microscopic interaction. Usual chemical ways are also based on different interactions. We researched possible entropy decrease in chemical reactions (Chang, 2014; Chang et al., 2013).

Heat preservation and cooling may be in isolated system at least for a shorter time. Oil mixed water forms sphere (its curvature  $R = 2/r$  is the biggest), and crystal are all ordered. Two interfaces in general physics possess microscopic interactions, in which the simplest tensile stress. Generally, soluble liquid mixed and the dissolved objects in the liquid cannot separate each other, but insoluble matter mixed must be separated after mixing. Special mud water must be clarified because the mass and gravity are different. Various biological solutions are more complex, so biological thermodynamics should be investigated (Chang, 2006a, 2013b).

## POSSIBLE ENTROPY DECREASE IN SOLIDIFICATION

The solidification process forms spontaneously an order structure (Langer & Müller-Krumbhaar, 1978). Under certain conditions, it may be isolated system of non-equilibrium process, which is possibly entropy decrease. Configuration entropy  $S_c$  of liquid decreases along with temperature  $T$ , and describes by Adam-Gibbs relation

$$t = t_0 \exp(A / TS_c), \text{ i.e., } S_c = \frac{A}{T \ln(t/t_0)} \quad (13)$$

For solidification there are mixed entropy, and high entropy alloy (Chang, 2006b; Zhou, Zhang, Wang, & Chen, 2007), etc. Non-crystal is metastable matter, which tends spontaneously to stable crystal under certain conditions, for example, high temperature cooling or supercooling. This is spontaneous entropy decrease. Two processes of alloy crystal on liquid are nucleation and growth (Gandin & Rappaz, 1994), and both are all ordering. Phase-field model may be based on the free energy function or entropy function (Karma & Rappel, 1996; Kobayashi, Warren, & Carter, 2000).

In cooling solid phase or liquid phase, enthalpy  $\Delta H$  decreases, and  $\Delta S = \Delta H / T_m$  (Vvedensky et al., 1993). When melting point  $T_m$  invariant, so  $\Delta H < 0$ , and  $\Delta S = \Delta H / T_m < 0$ .

For microscopic structure of solidification, no matter what form of equiaxed growth or preferred crystallographic directions (Kurz & Fisher, 1998) is all ordered.

The formation of spheres (the curvature  $R = 2/r$  is maximum) and crystals are also ordered. Further, clusters, embryos, metastable and supercooling (undercooling), etc., should all be entropy decrease. Solidification theory discusses kinetics of atom attachment, interface kinetics and nucleation kinetics, and introduced melting entropy, crystallography of entropy and dimensionless entropy of fusion  $\alpha = \Delta S_f / R$ . If  $R$  is invariant, when  $\alpha$  from small to bigger, entropy increases; contrarily, when  $\alpha$  from big to smaller, entropy decreases. It is known that definition of classical thermodynamics cannot determine the interfacial stability in growth. Therefore, the pseudo-thermodynamics indeed is introduced (Kurz & Fisher, 1998).

The rapid solidification processing (RSP) may be realized by deep supercooling of melt, bulk supercooling, or power fabrication. In the equilibrium distribution of crystal nucleus of supercooling melt, the mixed system is composed of  $N$  atoms of liquid phase and  $M$  atomic clusters of crystal (each atomic cluster includes  $n$  atoms). By comparison of system with only atoms but without crystal nucleus at the same temperature, change of free energy is

$$\Delta G = M \Delta G_n - T \Delta S_n. \quad (14)$$

Here  $\Delta G_n$  is change of free energy in order to form a crystal nucleus with  $n$  atoms, and  $\Delta S_n$  is mixed entropy with  $M$  atomic clusters and  $N$  atoms of liquid phase (Kurz & Fisher, 1998). From this

$$\Delta S_n = (M \Delta G_n - \Delta G) / T \quad (15)$$

If  $M \Delta G_n > \Delta G$ , there will be  $\Delta S_n > 0$ ; if  $M \Delta G_n < \Delta G$ , so  $\Delta S_n < 0$ .

Usually fluctuations can derive order, and fluctuations may be origin of internal interactions in isolated system. The magnetization process is more ordered, so it should be entropy decrease (Chang, 2005; Landau et al., 1980). A spontaneous magnetization is spontaneous entropy decrease in isolated system, which is also internal interaction. More case that is general is the thermodynamics in magnetism and electrodynamics. We should calculate their change of entropy.

## NECESSITY OF ENTROPY DECREASE IN SELF-ASSEMBLY

Self-assembly (SA) (Konieczny, 2020) is a process in which a disordered system of pre-existing components forms an organized structure or pattern as a consequence of specific, local interactions among the components themselves, without external direction. Therefore, it must be an isolated system, and be entropy decrease (Chang, 2014).

When the constitutive components are molecules, the process is termed molecular self-assembly. Molecular self-assembly is an autonomous process that forms molecules or polymer under non-external influence is nanostructure technology. Crane (1950) proposed two basic principles of molecular self-assembly (Crane, 1950).

Self-assembly can be classified by static or dynamic SA. In static self-assembly, the ordered state forms as a system approaches equilibrium reducing its free energy. In dynamic self-assembly, patterns of pre-existing components organized by specific local interactions are not commonly described as “self-assembled” (Konieczny, 2020).

Self-assembly in the classic sense can be defined as the spontaneous and reversible organization of molecular units into ordered structures by non-covalent interactions. The first property of a self-assembled system that this definition suggests is the spontaneity of the self-assembly process: the interactions responsible for the formation of the self-assembled system act on a strictly local level, i.e., nanostructure. This arises in the strong non-equilibrium conditions. The most famous example of self-assembly is the occurrence of the life on Earth. Another example is the phenomenon of electrostatic trapping, in which an electric field is applied between two metallic nano-electrodes. The particles present in the environment are polarized by the applied electric field. Due to dipole interaction with the electric field gradient, the particles are attracted to the gap between the electrodes (Bezryadin, Westervelt, & Tinkham, 1999). Self-assembly of crystals, works well (Stephenson & Hubler, 2015). Self-assembled monolayer and molecular self-assembled film are molecules pass through chemical bond, and interact spontaneously to form stable order film with the lowest energy (Ulman, 1998).

Any chemical reaction drives atoms and molecules to assemble into larger structures. An important feature of SA is the key role of slack interactions. Another distinctive feature of SA is that the building blocks are not only atoms and molecules, but span a wide range of nano- and mesoscopic structures, with different chemical compositions, shapes and functionalities. Recent examples of novel building blocks include polyhedral and patchy particles, and include micro particles with complex geometries, such as hemispherical (Hosein & Liddell, 2007b), dimer (Hosein & Liddell, 2007a), discs (Lee, Meng, Norris, Scriven, & Tsapatsis, 2006), rods, molecules (Garcia, Justo, Machado, & Assali, 2009), and multimers. These nanoscale building blocks (NBBs) can in turn be synthesized through conventional chemical routes or by other SA strategies.

Some important examples of SA in materials science include the formation of molecular crystals, colloids, lipid bilayers, phase-separated polymers and self-assembled monolayers (Roldán et al., 2011; Whitesides & Boncheva, 2002). The folding of polypeptide chains into proteins and the folding of nucleic acids into their functional forms are examples of self-

assembled biological structures. Adleman (Adleman, 1994), Winfree, et al. (Winfree, Liu, Wenzler, & Seeman, 1998) and Ignatova, et al. (Ignatova, Martínez-Pérez, & Zimmermann, 2008) discussed the self-assembly of DNA structures by the molecular and DNA computation. Recently, the three-dimensional macroporous structure was prepared via self-assembly of diphenylalanine derivative under cryoconditions, the obtained material can find the application in the field of regenerative medicine or drug delivery system (Berillo et al., 2012).

Chen, et al., demonstrated a microscale self-assembly method using the air-liquid interface established by Faraday wave as a template. This self-assembly method can be used for generation of diverse sets of symmetrical and periodic patterns from microscale materials such as hydrogels, cells, and cell spheroids (Chen et al., 2014).

Another characteristic common to nearly all self-assembled systems is their thermodynamic stability. SA is to take place without intervention of external forces, the process must lead to a lower Gibbs free energy, and thus self-assembled structures are thermodynamically more stable than the single, unassembled components. The driving force is capillary interaction, which originates from the deformation of the surface of a liquid caused by the presence of floating or submerged particles (Denkov et al., 1992). Uskoković researched that every self-assembly process in reality presents a co-assembly, which makes the former term a misnomer of a kind (Uskoković, 2008). The thesis is built of the concept of mutual ordering of the self-assembling system and its environment. Further, we discuss the nonlinear self-assembled theory.

Self-assembly processes can be observed in systems of macroscopic building blocks, which can be externally propelled (Hosokawa, Shimoyama, & Miura, 1994) or self-propelled (Groß & Dorigo, 2008). Groß, et al., discussed self-assembly at the macroscopic scale (Groß & Dorigo, 2008). Self-assembly is related closely with self-organization. Halley and Winkler discussed consistent concepts of self-organization and self-assembly (Halley & Winkler, 2008). Self-organization is the non-equilibrium process where self-assembly as a spontaneous process leads toward equilibrium. Self-assembly requires components to remain essentially unchanged throughout the process. Moreover, self-organization is related with the memory alloy.

## MOLECULAR MOTOR AND ENTROPY DECREASE IN BIOLOGY

Great scientist Eddington said: “The law that entropy always increases holds, I think, the supreme position among the laws of Nature.” However, science is a developing process. We should research other preconditions of entropy increase. Carnot law describes efficiency of heat engine, which is based temperature. Now some new engines, for example, efficiency of computer and molecular motor, etc., are not dependent of temperature. It is macroscopic concept that entropy  $S = Q/T$  is dependent of temperature. Usual second law of thermodynamics is mainly applied to macroscopic regions. This is probably unsuitable for Brownian movement, which seems to be analogy with a perpetual motion machine, and when microscopic internal energy (for example, chemical energy, nuclear energy, etc.) is transformed to macroscopic energy, the entropy decrease is all possible.

Present the second law of thermodynamics uses the entropy  $S$  to identify the spontaneous changes.  $S$  is a measure of the molecular disorder of a system. Nevertheless, when internal interaction in an isolated system exists, i.e., the kinetic energy is transformed to the potential energy, then the order increases, the kinetic energy and entropy decrease.

The life energy originates mainly from the photosynthesis, which compounds adenosine triphosphates (ATP) by light, and translates into the chemical energy. ATP hydrolyzed is an exergonic reaction, and the glycolysis may again produce ATP, which is an endothermic process. The both opposite processes must include entropy decrease.

The molecular motor takes a very important role for keeping high order in biologic systems. We think, the molecular motor corresponds to  $dS < 0$ , in which the chemical energy of cell translates into mechanical energy, whose efficiency is almost 100%. In the microtubule, the motor proteins have kinesin and dynein. Their moving way is hand-over-hand (Yildiz et al., 2003). The kinesin moves matter of cell nucleus to cell membrane, and dynein moves matter of cell membrane to cell nucleus. Their transport direction is just opposite, but both are not competition (Karp, 2002). Moreover, many motors may work together, and produce speed with 10-time unit motor. It is namely order cooperative action (Jülicher & Prost, 1995). The rotary motor is composed of biologic macromolecule, whose volume is small, and efficiency is very high almost 100%, and they may converse rotate. Its type is ATPase, which is a core enzyme for biologic energy translation in organism. The entire process of cell up growth and metabolism need energy, which is obtained from the chemical energy hydrolyzed by ATP under the most cases and ATP is synthesized from ATPase. The molecular motor of ATPase may hydrolyze ATP, and may synthesize ATP. This is similar with membrane and Maxwell demon. The endothermic reactions and opposite exergonic reaction in biology should correspond to entropy increase and opposite entropy decrease (Chang, 2006a; Yi-Fang, 1997).

The complex biological systems provide some modes on entropy decrease in an isolated system. This is known that any organism all is a typical self-organized system, and must be an order process of entropy decrease. As long as this process is isolated at least in a certain time, it all is a violation for the second law of the classical thermodynamics. Simultaneity, the cell membrane and ferment show a control for direction, which is a similar with the Maxwell demon.

Brain, consciousness, neuroscience, the permeable membrane, the molecular motor, etc., are all some internal interactions. They even possible take a key role for decrease entropy in isolated system. Dormancy of living body is an order state, whose entropy pass through adjustment and decrease to smaller. For the typical instance, the hibernation of animal, and the dormancy of Madagascar's lemur and of various hexapods all show obviously the entropy decrease in isolated system. Qigong and various practices are often related to these order states with entropy decrease (Chang, 2013b).

Ashby pointed out that two substances such as ammonia and hydrogen in a gaseous state can be mixed to form a solid (Ashby, 1961). Similarly, about twenty different types of amino acids present in microorganisms can gather to form a new reproductive process. It is commonly understood that solids are more orderly than gases, such that the entropy of a solid is less than the entropy of the same material in its gaseous

state. Microorganisms should likewise represent a more orderly state than the amino acids from which they are formed. In a biological self-organizing process, some isolated systems may spontaneously proceed toward the orderly states. Prigogine and Stengers have discussed such a case (Stengers, 1984): Under particular circumstance, such as when *Dictyostelium discoideum* experiences a lack of nutrition, solitary cells will spontaneously unite to form a larger cell cluster. In such a case, the cells and nutrition-liquid together may be regarded as an isolated system. Jantsch (Jantsch, 1979) pointed out that when different types of sponge and water are mixed within a uniform suspension, they rest for a few hours and then automatically separate into different types. More interestingly, when a small hydra is cut into its individual cells, the individual cells spontaneously evolve to form cell-clusters. Some cell clusters are malformations, but other cell clusters will eventually become a normal hydra.

The auto-control mechanism in an isolated system may produce a degree of order. If it does not need the input energy, at least in a given time interval, the auto-control will act like a type of Maxwell demon, which is just a type of internal interactions. Ordering is the formation of structure through the self-organization from a disordered state. The emergence and self-organization of biology and human all depend mainly on self-interaction, because only sunlight cannot produce spontaneously biology on Earth, at least, they cannot evolve some higher living body.

In an evolutionary process with long time, life forms a nonlinear complex and complete system with multi-levels: gene, cell, tissue, organ, system, individual, population, community, ecosystem, and biosphere. For various levels in isolated systems usual entropy increase, but entropy possibly decrease under some conditions with internal interactions, which has possibly different levels in biological systems, for example, membrane, enzyme, ATP and molecular motor, etc.

Membrane is one of basic biologic framework. The biologic membranes may choose a direction self-motion. Samal and Geckeler (Samal & Geckeler, 2001) investigated an unexpected solute aggregation for DNA, etc., in water on dilution, which violates the second law of thermodynamics. The cell membrane is a barrier with selectivity, on which the ion channel exists. For cell, it inputs continuously the metabolized matter, and removes the metabolized outcome. ATP provides energy, and lead that living body shows the macroscopic order. A permeable membrane is namely the Maxwell demon, which may derive entropy decrease.

In physiology, increased metabolism and an emotional state of being upset should be characterized by larger entropy. Conversely, decreased metabolism and easy conscience or a calm and good-natured emotional state should be characterized by smaller entropy. The immunity of an organism increases for positive emotions, but metabolism and body-temperature show a remarkable increase for a nervous state and negative emotions increase an organism's susceptibility to various diseases.

Anomalous cognition (AC) is defined as a form of information transfer in which all known sensorial stimuli are absent. Lantz et al., have reported testing sender condition and target types in AC experiments (Lantz, Luke, & May, 1994). There is a difference between static and dynamic target material. Entropy is defined as a measure of uncertainty or lack of information about a system. The data from both of these studies were analyzed with regard to the gradient of Shannon's entropy of

the targets. May, et al., were able to compute the entropy and its mathematical gradient for each target in these experiments (May, Spottiswoode, & James, 1994). AC was more pronounced when targets underwent massive changes in energy or entropy in a very short period. In addition, dynamic targets produced better results in the Ganzfeld than did static targets, a result that is suggestive of changes of entropy.

Landsberg defined “disorder” as the entropy normalized to the maximum entropy, which corresponding to a completely random system. A living being as a whole represents an extremely orderly state of being and must be an open system for long-time. A living being’s death represents a transformation to a state of total disorder, while sickness is a state of local disorder and a state of recovering from sickness is marked by a return to the higher order of health. The order parameters are thus health targets. However, for a short time Qigong and some states attained during religious practices, for example, Buddhist and Taoist meditation may be considered isolated systems that are characterized by entropy decreases.

In Buddhist practice, everyone is expected to face every day in a happy mood with thanks to Nature. A harmonious unification of these two activities of the human mind (body) and spirit can only consist completely of a normal sound activity. Qigong requires that one must be calm and good-natured, which is a more orderly state for a person. Clinical practices show that the practice of Qigong causes a reduction in human metabolism. This reduction seems to the possibility that human ideas and similar thought forms also entropy decrease, and achieves an ordering within a living system that could cure sickness and increase internal immune system strength. In the Chinese traditional practices the Inducing into Tranquilization and Qigong, and the “Chan-Ding” in Buddhism are all an ordering state. In these cases, the metabolism is entropy decrease in isolated system. These practice methods all are to benefit the control of consciousness. Probably, some ways are propitious to the cure of cancer.

Various developments in Universe are not always disorder. General biologic systems never are more disorder. The declining process of life is from order to disorder, and entropy increases. The origin, birth and developed process of life must be from disorder to order, and information increases. It should be entropy decrease with internal interactions. Both middle processes are the dissipation structure, and entropy is invariant. This is a total principle, which shows the relation between entropy decrease and life science. The self-organized order of any organism in isolated system is inevitably a process of entropy decrease, it may hold at least in a certain time. Various biological systems possess very rich and colorful internal interactions, we proposed entropy decrease as an index of therapeutics in biophysics, and believe that the bio thermodynamics will make with great contribution for the test of development on the thermodynamics of entropy decrease in isolated system. Biology will be a wide region for research of entropy decrease in various isolated systems.

## ENTROPY DECREASE IN ASTRONOMY AND SOME DISCUSSIONS

The material structure and energy have different levels. Gravity is between celestial bodies; classical macroscopic objects are mainly kinetic and potential energy; condensed matter and molecules are thermal energy; molecules and atoms are chemical energy; charged bodies are electrical energy;

organisms have biological energy; particles are strong and weak interactions. Thermodynamics began because of heat energy, and then extended to all levels indefinitely. But, we must consider internal energy transformation and entropy decrease (Chang, 2014).

If entropy is a measurement of disorder, some processes of the celestial evolutions must be order, and entropy decrease. For instance, a disorder nebula becomes an order star due to the gravitational interaction and so on. If the cosmological expansion and general hot movement and diffusion correspond to entropy increase, so the contraction of part region will be entropy decrease. Generally, entropy increase for repulsion, and entropy decrease for gravitation (Chang, 2013a, 2018). If entropy increases for black hole, entropy will decrease for white hole.

The galaxy may be regarded as an isolated system (Schmidt, 1974). Some scientists think that the total entropy is conserved in the evolution of the universe, and the entropy of each component is conserved (Yagi, Hatsuda, & Miake, 2005). But, usual assumption is “the entropy of the Universe increases in all natural processes”, then “isolated systems tend toward greater disorder, and entropy is a measure of that disorder” (Serway & Faughn, 2004), and even “heat death”. World is not pessimistic always (Yi-Fang, 1997). In some cases, internal interactions are very important. The gravitational interactions produce various ordered stable stars and celestial bodies. Various stable objects and their formations from particles to stars are accompanied with internal interactions, which have implied a possibility of entropy decrease. Different attractive and repulsive interactions are symmetrical, in which attractive interactions correspond to entropy decrease. These great symmetries and their breaking produce all of Universe.

The initial Universe is origin of quantum fluctuations, which do not obey the second law of thermodynamics. If the primeval matter (universe) was already a complete disorder state of maximum entropy (Davies & Davies, 1984), further evolution must be entropy decrease. In general, case, entropy decreases  $dS < 0$  for attraction force, and cooperation leads to order. Entropy increases  $dS > 0$  for repulsive force, and struggle leads to disorder. Gravity derives various celestial bodies and structures. Solar luminescence is derived from nuclear fusion and strong interaction. Boyanovsky discussed the imprint of entanglement entropy in the power spectrum of inflationary fluctuations (Boyanovsky, 2018).

Black hole should be entropy decrease, is opposite to gas diffusion with entropy increase, and is the biggest internal interaction. Moreover, black hole cannot be an isolated system, and thermodynamics of black hole should be the theory of dissipation structure, whose entropy decreases possibly (Chang, 2013a). This is impossible that both contrary collection and evaporation of black hole, or the Big Bang Universe and the Circulating Universe are all entropy increase. For opposite black hole and white hole, one is entropy increase, so another must be entropy decrease. We think some studies on entropy of black hole are only based on the belief of the second law of thermodynamics (Chang, 2018).

This is amazing that more generally various opposite processes, such as attraction and repulsion, endothermic and exothermic all are entropy increases.

Any stable objects and their formations from particles to stars are accompanied with internal interactions inside these objects, which imply a possibility of entropy decrease. Moreover, entropy decrease exists widely in social sciences

and society (Chang, 2013c, 2015b), for example, from troubled times to rule, from war to peace. A certain population can be used as an isolated system. If they communicate only, their information will be reduced; but, if they discuss, argue and interaction each other, many new ideas may be generated, and information will increase, and entropy will decrease. Therefore, the democratic and equal society is more conducive to the development of science and the social progress. Based on the complete structure (3) on entropy change, we propose quantitatively a total formula of entropy change for universal evolution of any natural and social systems, which may include physics, chemistry, biology, astronomy and social sciences, etc.

The total formula of entropy change is

$$dS = dS^a + dS_+^i - dS_-^i + dS_i + dS_e^+ - dS_e^- \quad (16)$$

When

$$dS^a + dS_+^i + dS_i + dS_e^+ > dS_-^i + dS_e^-, \quad (17)$$

Entropy increase  $dS > 0$ , the system tends to disorder. When

$$dS^a + dS_+^i + dS_i + dS_e^+ < dS_-^i + dS_e^-, \quad (18)$$

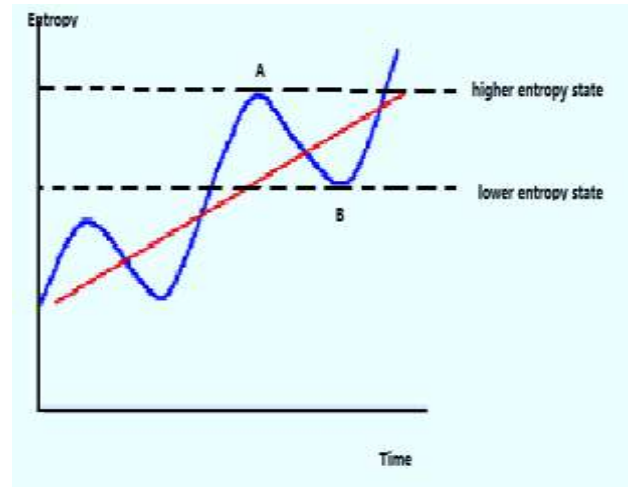
Entropy decrease  $dS < 0$ , the system tends to order. Both differences are determined by the input negative entropy flow in open system and the internal attractive interactions in isolated system  $dS_e^- + dS_-^i$ .

Entropy decrease is a change of entropy  $dS$  extended from the positive number to the entire real axis included negative number,  $dS(+R, 0) \rightarrow (+R, -R)$ . Further, we suppose that entropy can extend to the plane of complex number. It corresponds to  $dS$  is a complex number, whose pure form may be  $dS + id\tilde{S}$ , whose meaning is possibly that  $dS$  represents quantity of change, and  $d\tilde{S}$  represents undulate of change. They can correspond to vectors and the life index on heartbeat, breath, blood pressure and so on. The metric of the symplectic geometry is antisymmetric, so the electromagnetic field should be the symplectic geometry. The unification of gravitational and electromagnetic fields will be a unification of symplectic geometry and Riemannian geometry.

## CONCLUSION

At present, some physicists already identified with our viewpoint, for example, Tabti, et al., discussed melting of argon cluster (Tabti, Eddahbi, Ouaskit, & Elarroum, 2012)[76], and Quarati, et al., researched negentropy in the many-body quantum systems and energy from negentropy of

non-Cahotic systems (Quarati, Lissia, & Scarfone, 2016; Quarati, Scarfone, & Kaniadakis, 2018)[77,78]. The entropy of the Universe increases in all natural processes, and isolated systems tend to greater disorder (Serway & Faughn, 2004).



**Fig.2.** Transformation processes between states with higher entropy and lower entropy

Although the total entropy for whole system is positive and increase possibly, but, so long as different entropy states for any systems exist, entropy should decrease in transformation process from a higher entropy state to a lower entropy state (in Fig. 2 from A to B), for example, from chaos to order, from war to peace and so on (Chang, 2015a, 2015b). If this system is isolated, it will correct and develop the second law of thermodynamics.

Generally, the entropy increase law possesses relativity and approximation, because it must be an isolated system, but which always dependent of some conditions. Complete exact isolated system does not exist in essence, since the gravitational interaction cannot be screened completely. Change of entropy must consider relations, structures, time and evolution, etc.

In a word, in various phases and phase transitions, the most suitable for entropy increase is only the special gas diffusion process, and the nebula may be condensed into stars, liquid has also the capillary phenomenon. When internal interactions exist, entropy decrease is possible. Entropy increase or entropy decrease should be symmetrical and beautiful. This can derive the complexity of natural evolution and social phenomena. If the world is always entropy increasing, it is not only pessimistic, but also lack of basic beauty and complexity.

The change of entropy should be a testable science. At least some extensions of the second law of thermodynamics are not universal. As long as we break through the bondage of the second law of thermodynamics, so the rich and complex world is full of examples of entropy decrease.

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