SCIENTIFIC REVOLUTION: THERMODYNAMICS AS A GENERAL NONSTATISTICAL SCIENCE OF ANY SYSTEM IN ANY STATE

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GENERAL REMARKS

Ever since Clausius postulated that "the energy of the universe is constant", and "the entropy of the universe strives to attain a maximum value", practically every scientist and engineer shares and promulgates the almost religious beliefs that: (i) thermodynamics is a statistical theory, restricted to phenomena in macroscopic systems in thermodynamic equilibrium states; and (ii) entropy — the concept that distinguishes thermodynamics from mechanics — is a statistical measure of ignorance and ultimate disorder.

These beliefs stem from the conviction that the "known laws" of mechanics (classical or conventional quantum) are the ultimate laws of physics, and from the fact that statistical theories of thermodynamics yield many correct and practical numerical results about thermodynamic equilibrium states.

Notwithstanding the conviction and numerical successes, the almost universal efforts to compel thermodynamics to conform to statistical explanations, and to restrict it only to thermodynamic equilibrium states are puzzling in the light of many accurate, reproducible, and nonstatistical experiences, many phenomena that cannot possibly be described in terms of thermodynamic equilibrium states, and contrary to a long list of theoretical considerations.

Since the advent of thermodynamics, many academics and practitioners have questioned the clarity, unambiguity, and logical consistency of traditional expositions of the subject. Some of the questions raised are: (i) can temperature T and heat Q be defined without specifying all three laws (the first, second, and third) of thermodynamics, and without proving a large number of their theorems?; (ii) how can entropy, Q/T, be defined if Q and T are viewed as undefinable primitive concepts?; (iii) why is thermodynamics restricted to thermodynamic equilibrium states only, given that the universally accepted and practiced statements of energy conservation and entropy nondecrease are demonstrably time dependent?; (iv) why do we insist that it applies to macroscopic systems only, given that even Gibbsian statistics and systems in states with negative temperatures prove beyond a shadow of a doubt that thermodynamics is valid for any system, including systems consisting of one particle or one spin?; (v) how can any statistical expression of entropy be accepted if none of these expressions conforms to all the requirements that the entropy of thermodynamics must satisfy?; (vi) why do we continue to believe that thermodynamic equilibrium is a state of ultimate disorder despite the fact that both

experimental and theoretical evidence indicates that such a state represents ultimate order?; and (vii) why in response to relativistic and quantum phenomena do we accept radical modifications of the "known laws" of classical mechanics, whereas in the case of entropic phenomena we refuse to consider even the possibility that though correct these laws are incomplete?

A NEW EXPOSITION OF THERMODYNAMICS

Over the past three decades, a small group at the Massachusetts Institute of Technology has developed a nonstatistical exposition of the foundations and applications of thermodynamics that applies to all systems (including one particle or one spin systems) and to both thermodynamic equilibrium and not thermodynamic equilibrium states.

In the new exposition, we start with the mechanical concepts of space, time, and inertial mass or force, and express the first law as follows: *Any two states A_1 and A_2 of system A may always be the end states of a process that involves no other effects external to the system except the change in elevation of a weight between z_1 and z_2, that is, solely a purely mechanical effect, and $z_1 - z_2$ depends only on A_1 and A_2.* In contrast to all other expositions, it is noteworthy that this statement does not involve the concepts of energy, temperature, heat, and work, all of which are defined later.

The first law has many implications — gives rise to many rigorously proven theorems. Examples are: (i) at each state of a system there must exist a function E, called energy, such that the change of its value $E_2 - E_1$ from state A_1 to state A_2 is proportional to $z_2 - z_1$; (ii) in the course of spontaneous changes of state (changes in time in an isolated system), E is invariant; and (iii) in the course of interactions, $E_2 - E_1$ must be accounted by the energy exchanged with systems interacting with A.

Next, depending on their evolution in time,

we classify states in the seven categories encountered in mechanics, that is, unsteady, steady, nonequilibrium, equilibrium, unstable equilibrium, metastable equilibrium, and stable equilibrium, and raise the question: For given values of the energy, the volume, and the amounts of constituents of a system, are there any stable equilibrium states?

In the new exposition, the answer is given by the second law which avers that (simplified version): *For each set of values of energy E, volume V, and amounts of constituents n, there exists one and only one stable equilibrium state.* It is noteworthy that the mechanical concept of stable equilibrium is what in ordinary expositions is called equilibrium or thermodynamic equilibrium state, and that, in contrast to all other expositions, here the second law does not involve the concepts of heat, temperature, and entropy.

The second law cannot be derived from or explained by the "known laws" of physics either directly or statistically because these laws imply that the state of lowest energy is the only stable equilibrium state, whereas the second law avers that such a state exists for each value of the energy.

Among the many rigorously proven implications of the first and second laws are: (i) at each state of system A there must exist a function S, called entropy, such that its value S_1 in state A_1 is proportional to the difference between the energy E_1 and the energy Ω_1^R exchanged in a reversible process between a weight and a composite consisting of A and a reservoir R; as a result of this process, the state of A would change from A_1 to a state such that the composite of A and R is in a stable equilibrium state; (ii) in the course of spontaneous changes of state, if they are reversible S is invariant, and if they are irreversible S increases; (iii) in the course of interactions that change the state from A_1 to A_2, the difference $S_2 - S_1$ must equal the entropy exchanged with systems interacting with A plus a nonnegative amount generated spontaneously within A; the latter amount is called *entropy generated by irreversibility*;

(iv) the minimum value of entropy is zero; (v) if a system is in a stable equilibrium state, then and only then the entropy is an analytic function of the form $S(E, n, V)$; (vi) for stable equilibrium states only, the concepts of temperature, total potentials, and pressure are defined in terms of partial derivatives of $S(E, n, V)$; for states that are not stable equilibrium, the definitions of temperature, total potentials, and pressure are meaningless and useless; (vii) work, heat, and other interactions are defined in terms of concepts introduced up to this point; for example, work is an interaction that involves only the exchange of energy between the system and other systems in its environment; and heat is an interaction that involves only the exchanges of energy and entropy between a system and one or more reservoirs; (viii) any expression that purports to represent entropy must conform to eight conditions; for example, one of these conditions is implication (ii) in this list; and (ix) all other correct statements of the second law are special cases of the second law given here.

In the new exposition, the third law avers that: *For each given set of values of V and n of system A (without a finite upper limit on energy) there exists one stable equilibrium state with zero temperature.*

Neither the statements of the three laws nor the proofs of any of their theorems — implications — require any considerations about numerical difficulties that prevent us from making explicit calculations, and about statistical measures of ignorance (or lack of information), or any restrictions to systems of specific sizes and specific numbers of degrees of freedom, or any limitations to states of specific types. Exceptions to the last assertion are theorems proven solely for specific classes of states. So a statistical interpretation of thermodynamics is unwarranted and misconceived, and a restriction to specific states totally unjustifiable. In particular, entropy is a nonstatistical property possessed by the constituents of any system in any state in the same sense that iner-

tial mass is a property of any system in any state. Entropy adds a most important dimension to the property space of a system, a dimension that distinguishes the phenomena explained by thermodynamics from the phenomena that correspond to zero entropy and can be encompassed solely by the "known laws" of physics.

AN ENERGY VERSUS ENTROPY GRAPH

At an instant in time, a state can be represented by a point in a multidimensional space with one axis for each amount of constituent, volume, and each independent property. Such a representation, however, is unwieldy because the number of independent properties of any system, even a system consisting of one particle only, is infinite. Nevertheless, useful information can be captured by first cutting the multidimensional state space by a hypersurface corresponding to given values of each amount of constituent and the volume, and then projecting the cut on an energy versus entropy plane. For system A without upper bound on energy, it is proven that the projection must have the shape of the cross-hatched area in Figure 1.
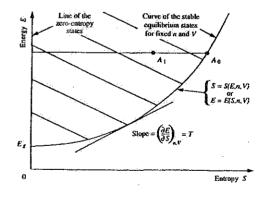

Figure 1: Energy versus entropy graph.

A point either inside the cross-hatched area or on the line $S = 0$ represents the projections of an infinite number of states. Each such state has the same values of amounts of constituents n, volume V, energy E, and entropy S but differing values of other properties, and is not a stable equilibrium state.

In particular, the line (and more generally the surface) $S = 0$ represents all the states encountered in purely mechanical theories of physical phenomena, that is, the states that are regularized by the "known laws" of physics. The convex curve represents classical thermodynamics. Each point on the curve corresponds to one and only one stable equilibrium state. For any such state, the value of any property is determined solely by the values of the energy, the r amounts of constituents, and the volume of that state. Many theorems of the laws of thermodynamics can be elegantly and simply illustrated on the E versus S diagram. Projections of other cuts of the multidimensional state space on other planes, such as E versus V, or E versus the amount of a constituent, are possible. Each results in a graph that provides visual illustrations of different aspects of the new exposition.

In what follows, we illustrate the generality of the new exposition by providing definitive answers to some questions that have preoccupied thousands and thousands of scientists and engineers over the past century and a half.

A THERMODYNAMIC EXORCISM OF MAXWELL'S DEMON

Maxwell is one of the great scientists who believed that all physical phenomena are mechanical, but numerical difficulties with macroscopic systems force us to abandon the mechanical explanation and resort to the statistical method. He said: "One of the best established facts in thermodynamics is that it is impossible in a system enclosed in an envelope which permits neither change of volume nor passage of heat, and in which both the temperature and the pressure are everywhere the same, to produce any inequality of temperature or of pressure without the expenditure of work. Now let us suppose that such a vessel is divided into two portions B and C by a partition in which there is a small hole, and that a being — a demon — who can see the individual molecules, opens and closes this hole, so as to allow only the swifter molecules to pass from B to C, and only the slower ones to pass from C to B. He will thus, without expenditure of work, raise the temperature of C and lower that of B, in contradiction to the second law of thermodynamics." Inherent in this line of reasoning is the idea that, at the molecular level, entropy is not a physical property.

Hundreds of papers and several books have been written over the past century, all claiming to prove that the demon cannot violate the second law. In our view, none of these publications has proven what is claimed because none addresses the problem posed by Maxwell. In each publication, either the demon or the environment of the vessel, or both experience some effects in sharp contrast to Maxwell's specification that such effects are not needed by an omniscient and omnipotent demon (god?).

In the new exposition, the exorcism satisfies Maxwell's specifications and is definitive. It can be illustrated by means of the energy versus entropy graph shown in Figure 1. This graph can be regarded as representing the states of the air, with entropy being the sum of the entropies of individual molecules in the same sense that inertial mass is the sum of the inertial masses of individual molecules. Starting with stable equilibrium state A_0, the demon is asked to sort the air molecules into swift and slow without any changes in the values of the energy, the amount of the air and the volume, and without any change either of his state or, more generally, of the state of the environment. If this were possible, the final state of A would be A_1, that is a state with the same values of E, n, and V as those of A_0, but less entropy than that of A_0. But we have proven that entropy is a nondestructible, nonstatistical property of every molecule of A and, therefore, of all the molecules of A. Accordingly, the demon cannot reduce the entropy without compensation no matter how "fine his tactile and perceptive organization" is. It is clear that this impossibility has nothing to do with either entropy

generated by irreversibility, shortcomings of the demon's procedures and equipment, or collection and discard of information, that is, with the justifications given in all the publications on the subject to date.

Equivalently, if the demon is regarded as a cyclic perpetual motion machine of the second kind (*PMM2*), then his ultimate task is to extract only energy from system A and, thus, change state A_0 to a state of smaller energy and equal or larger entropy than those of A_0. But under the specified conditions — fixed values of the amount of air and the volume — the graph in Figure 1 shows that each state of energy smaller than that of A_0 has also smaller entropy. And again, because entropy is a nondestructible property of each molecule and, therefore, of the air, the demon cannot accomplish his assignment because if he did, he would have reduced the entropy without compensation.

Some authors claim that the demon is infeasible even if the initial state of A is not stable equilibrium. This claim is also erroneous. If the initial state A_1 is not stable equilibrium and, therefore, lies somewhere within the cross-hatched area in Figure 1, then even an incompetent demon could either extract only energy from A or change the state of A at constant energy without violating the laws of thermodynamics because there exists an infinite number of states with lower or equal energy and equal or larger entropy than the energy and entropy of A_1, respectively.

As we discuss later, another way of exorcising the demon is by proving that, in thermodynamic equilibrium, each molecule is at a standstill and, therefore, there are no fast and slow molecules to be sorted out.

BOLTZMANN'S MISCONCEPTIONS ABOUT ENTROPY AND DISORDER

Boltzmann is another great scientist who believed that all physical phenomena are mechanical. As a consequence he interpreted entropy as a measure of disorder, and the tendency toward larger values of entropy as a tendency toward greater disorder. He specified the entropy expression as the logarithm of the number of microstates that correspond to a given macroscopic state, an expression that is incised on his gravestone. However, both our current theoretical understanding, and the experimental evidence indicate that Boltzmann's views are questionable.

In the new exposition, we prove that each stable equilibrium state is not one of many microstates but a unique state because this uniqueness is required by the second law of thermodynamics. So, even if we use Boltzmann's criterion of disorder, we must conclude that such a state represents perfect order. Moreover, as we discuss later, the expression for entropy proposed by Boltzmann does not conform to all the conditions that it must satisfy.

An empirical contradiction of the idea that a spontaneous entropy increase implies an increase of disorder is the spontaneous crystallization of an agitated and turbulent fluid. It is hard to argue that an agitated and turbulent fluid is more orderly than a perfect crystal!

Another empirical contradiction of the idea of increase of disorder is the spontaneous crystallization of a supercooled liquid. Here again it is difficult to claim that the supercooled liquid is more ordered than the crystal. Nevertheless, Denbigh argues that: "...though there is a decrease of configurational entropy, consequent on the more orderly arrangement of the lattice as compared to the liquid, there is a more than compensating increase in *thermal energy*, due to the randomization of the liberated potential energy over the vibrational motions of the atoms in the crystal."

There are several objections to Denbigh's insistence to explain the spontaneous entropy increase by invoking a connection between thermodynamic equilibrium and disorder. For example, there is only one entropy. Its partition into several different kinds is a numerical convenience and not a fundamen-

tal result. Again, thermal energy — heat — is not a property of a system. Thermal energy refers to the entities that are exchanged between two systems in the course of a heat interaction. Again, as we discuss later, in any stable equilibrium state, the value of the velocity of any individual particle equals zero. So, if a crystal is in a stable equilibrium state, there are no vibrational motions.

THE QUANTUM-THEORETIC CONNECTION

Albert Einstein said: "A theory is the more impressive the greater the simplicity of its premises, the more different kinds of things it relates, and the more extended its area of applicability. Therefore the deep impression which classical thermodynamics made upon me. It is the only physical theory of universal content concerning which I am convinced that, within the realm of the applicability of its basic concepts, it will never be overthrown." This beautiful remark by the "Man of the 20th Century" begs the question: Is it possible for a theory that will never be overthrown to be so devoid of all other theories of natural phenomena?

More than three decades ago, Hatsopoulos and Gyftopoulos puzzled over this question and, after several false starts, discovered that the answer is a very emphatic and productive NO. Specifically, they recognized that: (i) quantum theory admits probability distributions more general than those represented by wave functions; (ii) the more general probability distribution functions provide the link between quantum theory and thermodynamics without resort to statistics; this link extends the realm of quantum theory to states encountered in thermodynamics, and thermodynamic principles to quantum phenomena; (iii) Schoedinger's equation of motion — the equation that describes the evolution in time of concepts encountered in conventional quantum mechanics — is correct but incomplete; it is incomplete because it covers neither all reversible processes nor any irreversible processes; and

(iv) by adding the second law of thermodynamics to the laws of quantum theory, one can establish results for both stable equilibrium and not stable equilibrium states.

Next, Beretta in his doctoral research project discovered an equation of motion that describes both all reversible processes, and irreversible processes; this equation makes the statements of the three laws of thermodynamics superfluous because they become theorems of the complete equation of motion in the same sense that, in classical mechanics, conservation of kinetic energy and of momentum in the course of elastic collisions are theorems of Newton's equation $F = ma$.

In contrast to the special theory of relativity which becomes numerically important for very high energies, the new thermodynamics is numerically important for relatively low energies because then and only then the effects of entropy on available energy (exergy) are numerically sizeable. The reason is that at high energies and in the limit of stable equilibrium, entropy is proportional to the logarithm of the energy and, therefore, numerically ineffectual in comparison to the value of the energy. It is clear that, for the same energy but states that are not stable equilibrium, entropy is even more ineffectual because then its value is smaller than that of the entropy of the stable equilibrium state. The unification of quantum theory and thermodynamics opens new vistas at the frontier of science and engineering, creates new opportunities for further theoretical and applied work, and resolves many controversial issues. Examples of these issues are discussed below.

A QUANTUM-THEORETIC EXORCISM OF MAXWELL'S DEMON

In the context of the unified quantum theory of mechanics and thermodynamics, in order to establish the velocity of each molecule of Maxwell's air the demon must perform velocity measurements on an ensemble of identical systems prepared so that each system is in the same stable equilibrium state. As a

result of such measurements, we prove that he finds that the quantum-theoretic value of the velocity of each molecule — not the average of the velocities of all the molecules — equals zero. Thus, he concludes that there are no swift and slow molecules to be sorted out, and regrets the 140-year long efforts to solve a problem that does not exist!

This conclusion is puzzling to and unacceptable by almost all scientists and engineers familiar with statistical classical mechanics, and accustomed to thinking of temperature as a statistical ensemble average of kinetic energies of the molecules, and of pressure as a statistical time average of impulses of molecules bouncing off the wall of the container. Both thoughts, however, are misconceived for the following reasons.

In both the unified theory and statistical quantum mechanics, the temperature of a stable equilibrium state is determined exclusively by the expectation value of the energy, and the expectation value depends on the energy eigenvalues of the stationary energy eigenstates of the system. If the system behaves as an ideal gas confined in a box with zero potential energy, then almost all scientists and engineers express each energy eigenvalue as a classical kinetic energy. But this apparently innocuous substitution is not warranted because the expression for kinetic energy is not universal — not common to all paradigms of physics. For example, there is one expression in classical mechanics, another in special relativity, and none in general relativity. What is a common criterion of motion in classical mechanics, special relatively, and quantum theory is a nonzero value of velocity of each constituent — not an average of velocities of many constituents — and then an appropriate measure for kinetic energy. So, for ideal gas behavior, the value of the velocity of each molecule is zero and, therefore, the kinetic energy of each molecule is zero. It follows that a nonzero temperature cannot be a statistical average of zero velocities.

In addition, like entropy, temperature has a

dimension which is not among those of mechanics — space, time, and inertial mass. So the relation in statistical quantum mechanics which associates temperature with the average kinetic energy of the molecules of an ideal gas is numerically correct but theoretically wrong. A simple illustration of the theoretical error is provided by an example in classical mechanics. If the position of a particle varies exponentially in time, then also its velocity varies exponentially in time. Despite the identity of the two time dependencies, no one argues that, in general, velocity is equivalent to position.

In all fundamental theories of physical phenomena, a property is defined as a characteristic of a system that has a definite value independent of the history of the system, and that is measurable at each instant in time. Pressure is a property of a system in a stable equilibrium state. If a system is in such a state, we can rigorously prove that pressure is the force per unit area exerted by the walls of a container on the immobile molecules or, equivalently, the force per unit area that the immobile molecules exert on the walls of the container. This result involves no averaging of impulses on the walls over an infinite period of time, a fact consistent with the definition of a property. So the prevailing impressions about the relations between temperature and pressure and moving particles cannot withstand close scientific scrutiny.

REVERSIBILITY AND THE AGE OF THE UNIVERSE

Some scientists believe that we can expect to see unusual events such as gases unmixing themselves, only if we wait for times inconceivably long compared with the age of the universe. This belief has its origin in a calculation made by Boltzmann in response to objections to the statistical interpretation of irreversibility raised by Poincaré. It perpetuates a fallacy that has plagued the debate about the validity of statistical mechanics for over a century because it overlooks both the thermodynamic definition of reversiblity, and

fundamental results of quantum theory.

Even if valid, the inconceivably long time calculated by Boltzmann is premised on the interpretation of entropy as an informational, statistical measure of disorder, and only one process, the process of spontaneous return to the initial state via velocity reversals and intermolecular collisions. Three experiments and a theoretical argument are typical contradictions of the premises just cited.

A well insulated bucket of water consists of half hot and half cold water. Upon interacting only with each other, the two halves become lukewarm and, of course, the process is irreversible because the entropy of the lukewarm water is larger than the sum of the entropies of the hot and cold parts. However, we can always restore the hot and cold parts over a very short period of time — a time that has no relation to the age of the universe — by means of cyclic machinery which leaves the energy of the environment intact but increases its entropy even if the machinery is thermodynamically perfect — reversible. Moreover, the restoration of the initial state of the water is independent of the speed with which it is achieved, and involves neither velocity reversals nor any special information.

A high quality charged battery is wrapped in excellent insulation and left idle on a shelf. After a few years, the battery is found to be dead because of internal discharge at constant energy. At that time, we can restore the initial state of the battery over a period of time much shorter than the time required for the completion of the spontaneous internal discharge, without velocity reversals and complicated instructions. The spontaneous discharge is irreversible. Upon completing the recharging process, the energy of the environment is unchanged but its entropy increases even if the recharging is perfect, and occurs over a short or long period of time.

The chemical reaction $2H_2 + O_2 = 2H_2O$ in an isolated oxidation chamber is irreversible. However, the electrolytic reformation of H_2 and O_2 can be accomplished very quickly, without velocity reversals. At the end of the reformation, the energy of the environment has not changed but its entropy has increased at least by a minimum amount of entropy.

Finally, there is a fundamental theoretical objection to both the validity of the inconceivably long time calculated by Boltzmann and the recurrence of the initial state via velocity reversals. The objection arises from a basic result of the unified quantum theory, has no classical analogue, and is almost always overlooked. It asserts that, in a stable equilibrium state, the value of the velocity of each and every molecule is zero. Accordingly, neither the collisions invoked by Boltzmann nor velocity reversals can restore the initial state over either a short or a long period of time because every molecule is at a standstill, and there are no velocities to be reversed.

VISUALIZATION OF THE ENTROPY OF THERMODYNAMICS

A large number of analytical expressions have been proposed for the entropy of thermodynamics but none satisfies all eight conditions that have been established in the new exposition. It is proven that the only acceptable expression is one in terms of the general probability distributions identified in the unified quantum theory of mechanics and thermodynamics. In addition to satisfying the eight conditions, the new expression has an interesting interpretation. It is a measure of the quantum-theoretic shape of the constituents of the system in any particular state. Moreover, it is shown that the spontaneous generation of entropy — the sole source of irreversibility — occurs as the constituents of a system adjust their shapes in order to conform to both the externally applied forces, such as forces exerted by the walls of a container, and the interparticle forces in effect in each system. This adjustment can continue until no further reshaping is possible, that is, until the appropriate, unique stable equilibrium state is reached.

It is noteworthy that a change of shape of

molecules is inconceivable in classical mechanics and, therefore, statistical classical mechanics cannot represent the phenomena regularized by thermodynamics. In addition, though shapes and shape changes exist in conventional quantum mechanics, they too cannot account for the phenomena addressed by thermodynamics because they correspond to zero entropy and, therefore, statistical quantum mechanics cannot represent states with nonzero entropy.

The resolution of these difficulties is achieved by the introduction of the more general probability distribution functions of the unified quantum theory of mechanics and thermodynamics. This theory includes shapes and shape changes that correspond to both zero and nonzero entropy states.

CONCLUDING REMARKS

Maxwell's and Boltzmann's contributions to the statistical interpretation of thermodynamics have had and continue to have profound, stimulating, and productive influences on science and engineering, but they are colored by the conviction that, in principle, all physical phenomena obey only the "known laws" of physics, that is, the laws of classical mechanics and the laws of conventional quantum mechanics.

Throughout the past century and a half, many eminent scientists and engineers, and serious students of thermodynamics have raised objections to the validity and completeness of the statistical interpretation but almost always fell short of providing answers that are radically different from the Maxwell-Boltzmann paradigm.

Over the past 30 years, the group at MIT has recognized that the rules of quantum theory and the second law of thermodynamics admit broader interpretations than those accorded them in the literature. As a result, the group developed a unified quantum theory of mechanics and thermodynamics, discovered a complete equation of motion, and composed a new exposition of thermo-dynamics without reference to quantum theory. These developments result in the elimination of statistical and information theory arguments, and recognize the universal — without exception — existence of entropy as a bona fide property of any system in any state in the same sense that inertial mass and energy are bona fide properties of any system in any state. The new paradigm provides definitive answers to questions that have been intensely and inconclusively debated over the past century and a half, and creates novel opportunities for expanding the frontiers of both mechanics and thermodynamics. Though it differs radically from that of Maxwell and Boltzmann, the newer understanding does not diminish the greatness and importance of the contributions of these scientific giants. In fact, it makes their contributions even more valuable because without them no progress would have been made.