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Book Review[1]

The Kind of Motion We Call Heat: A History of the Kinetic Theory of Gases in the 19th Century, Volume VI of **Studies in Statistical Mechanics.** By Stephen G. Brush. North-Holland Publ. Co., Amsterdam, New York, Oxford, 1976. **Book 1: Physics and the Atomists,** xxxix + 299 pp., $24.75; **Book 2: Statistical Physics and Irreversible Processes,** xxxix + 470 pp., $59.75 (the set $75.00).

Professor Brush, a physicist by training, is a prolific writer in the history of the kinetic theory and statistical mechanics. This is a collection of articles he has published over the past two decades. The introductory chapter and other material have been added, "to provide a reasonably comprehensive picture of the entire subject," to cover the nineteenth century. He revised all chapters in the light of his "own current research and the secondary literature up to June 1975."

Following the Introduction, the rest of Volume 1 (Chapters 2–8) is organized according to personalities. Beginning with the little known precursors Herapath and Waterston, there follow Clausius, Maxwell, Boltzmann, van der Waals, and finally, for the opposition, Mach.

Volume 2 is devoted to *Problems.* Since it is impossible to do justice to all aspects of this work, a list of the chapters should give an impression of its scope. Chapter 9, The wave theory of heat; Chapter 10, Foundations of statistical mechanics 1845–1915; Chapter 11, Interatomic forces and the equation of state; Chapter 12, Viscosity and Maxwell–Boltzmann transport theory; Chapter 13, Heat conduction and the Stefan–Boltzmann law; Chapter 14, Randomness and irreversibility; Chapter 15, Brownian movement.

Each section is followed by a bibliography with many informative comments; at the end there is an extensive bibliography of research publications on the kinetic theory of gases, 1801–1900, along with a detailed index. The book is attractively produced, but is not quite free of misprints.

The student of the Boltzmann equation interested in its (and his) roots will find rich material on the personalities and conflicts of the great founders

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[1] There will be a response from the book's author in a future issue of the journal.

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as well as on lesser figures, whom the author has sought out with a historian's flair. The organization of the work brings about a certain fragmentation. The reader may make the best of it by selecting topics of interest to him without concern for breaking the continuity.

The reviewer believes that a historical study can have more than anecdotal interest for the research physicist. Tracing modern concepts to their origin often brings to light unexpected shades of meaning, and it is instructive to study the turning points of history, the so called scientific revolutions, which have brought about dramatic clarifications of conceptual quandaries.

However, these benefits come about only through a careful analysis of judiciously chosen material. I am afraid that the present work does not qualify by such rigorous standards.

Let us consider an original and not untypical contribution in which Brush traces the origin of the concept of irreversibility back to the speculations of eighteenth and nineteenth century geologists on the cooling of the earth (Section 14.2). This problem is abstruse even by modern standards and its inconclusive discussion is carried on over 16 pages. The feeling of unreality is heightened by the fact that Brush does not let us on to the fact that the problem was insoluble before the discovery of radioactivity.

We have to add that there was no gap in standard history-writing that called for the introduction of this innovation. The irreversible approach to a state of equilibrium is inherent in the method of calorimetry known since the beginning of the eighteenth century. The simple experimental setup involved was of use to Joseph Black in his development of other concepts as well: the intensity and quantity of heat (temperature and caloric), the conservation of heat quantity, and the concepts of specific heat, latent heat, and heat of reactions—in sum the whole conceptual arsenal of early thermophysics and chemistry.

Why are these well-known facts passed over by Brush in favor of a new idea which only generates perplexities? I conjecture that the clue is in his methodological preconceptions, which we have to examine.

Physicists have traditionally prided themselves on the fact that the "scientific method," combining experiment and mathematics with an objective analysis, raises them above the level of verbal squabbling. It is widely accepted at present that this picture is somewhat optimistic. Surrounding the area of solid achievements is a grey area where speculation is indispensable. There can be honest disagreements, and adherents of different schools of thought may well behave as contending parties in a power struggle. In fact, in polarized situations criticism is apt to turn from objectivity to partisanship.

In popular accounts the fight of new ideas against entrenched tradition has always had more dramatic appeal than a pedantic description of longwinded scientific arguments, and in recent years historians of science have given increasing attention to adversary attitudes. The analogy between scientific and political revolutions is used to come to terms with the unwieldy scientific process. Brush clearly adheres to this school of thought, as he explains in Section 1.6.

I believe that this emphasis on adversary methods of argumentation has been carried too far. There is ample evidence that periods of intense partisanship are transient, in the sense that old issues are being clarified, and the controversy shifts to new areas. While we recognize subjective partisanship as a historical fact under certain conditions, we need not accept this as a standard of procedure. In fact, historians and physicists might cooperate to set the record straight once the necessary state of knowledge has been reached.

Let us examine now how Brush applies these general ideas to concrete situations in thermophysics. I shall confine myself to the two main issues of this work: the "overthrow" of the caloric theory and the vindication of the kinetic theory of gases over the critique of the phenomenologists.

The evolution of the concepts of thermodynamics has a huge literature, and the author presumably considers his interpretation superior to the existing ones. Since he fails to make any actual comparison, however, it is in order to give a short sketch of the conventional account.[2]

We have noted already the foundation of caloric thermodynamics by Joseph Black, based on the principle of conservation. The conversion experiments of Rumford, Davy, and Volta around 1800 cast doubt on this idea. The only way to save calorimetry was to reconcile the apparently antithetical concepts of conservation and conversion. This was carried out by Carnot for reversible and by Joule and Mayer for irreversible conversion without regard to consistency. In 1848 William Thomson derived fundamental results from Carnot's principle, and he pointed out in 1849 that the latter conflicts with the experiments of Joule.[3] The clearly stated paradox was resolved the following year by Clausius, and in 1851 Thomson came out with an almost modern formulation of thermodynamics. The confusion was over and the conservation of heat quantity remained as valid as ever under the limiting conditions of calorimetry.

The phenomenological character of the above discussion conforms to the ground rules of thermodynamics. These rules are completely turned around by Brush, who suggests that the only problem is the replacement of the dictum "heat is matter" with "heat is motion" (Section 1.5 and Chapter 9). Thus the issue is shifted to a vague verbal plane, and the real problem

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[2] The following sketch covers well-known ground; for a short survey with documentation, particularly on the link connecting Volta with Joule, see Tisza.(1)

[3] See Ref. 2 for Thomson's papers referred to here.

concerning the scope and validity of conservation is not even mentioned. In fact, "conservation" does not appear in the index of this work on thermophysics, as against more than fifty entries under "radiation." The author's interest in the latter is explained on p. 31:

> My conclusion, based on material presented in detail in Ch. 9, is that the caloric theory of heat was rejected not because of any difficulties in explaining thermal phenomena but because the particle theory of light has been replaced by the wave theory of light. Because of the great interest in *radiant* heat during the period 1800–40, and several key experiments which indicated that radiant heat had the same qualitative properties as light, it was generally accepted that both heat and light must be explained by the same kind of theory...

On p. 32 he writes, "The transition from the caloric was not too difficult, since many of the properties formerly attributed to caloric could now be attributed to the ether."

Chapter 9 contains a survey of opinions that show a preference for the wave theory of heat. However, there is no evidence that this conception had or could have had any real role in the establishment of the principles of thermodynamics. Specifically, Brush notes the total absence of such an influence in the above-mentioned Thomson paper of 1849. However, instead of acknowledging the failure of his working hypothesis, his reaction is (pp. 331–332) that this article, "displays an amazing ignorance of the current state of opinion among physicists on the nature of heat." Thomson's turnabout two years later is also explained: "During this interval someone has told him about the wave theory of heat...."

The two papers of Thomson and that of Clausius are also discussed in more realistic terms on pp. 566–579, but the discrepancy with p. 331 remains unexplained. It is hard to avoid the conclusion that the author's historical reconstruction conflicts even with his own selection of historical facts.

The decisive step dispelling the confusion that blocked the consistent formulation of thermodynamics was not brought about by a shift in public opinion, nor by vigorous attacks on the caloric theory, but by the analysis of Clausius, who had, in the words of Gibbs,⁽³⁾ "this ability to bring order out of confusion, this breadth of view which could apprehend one truth without losing sight of another, this nice discrimination to separate truth from error...."

It is apparent that Brush does not have much liking for phenomenological theories, and he has a great deal more to offer on the history of the kinetic theory. He starts with its prehistory, and it is interesting to find out how much thought and effort went into the identification of the appropriate molecular model that we are now apt to take for granted. No sooner was this clarified than the profound problem arose of injecting statistics into deterministic mechanics and to establish a reasonably consistent mathematical formalism.

The problem of identifying the constituents of matter and establishing heir mathematical description continues to be the central issue of physics. Although the frontier has shifted from molecules to quarks, the general spirit is the same, and many will enjoy seeing how old issues have been clarified.

In this process a paramount role was played by criticism, regardless of whether it came from friend or foe. Among the friendly critics, Ehrenfest is right credited with clarifying many obscure points in the kinetic theory. His famous stochastic model (not mentioned by Brush) was felt to be particularly illuminating at the time,(4) and it continues to be used in courses on statistical mechanics. However, Ehrenfest's style of focusing on the weakness of his own theory instead of attacking opponents does not harmonize with the adversary methodology. In one of his critical remarks the author complains (p. 180) that "the reversibility and recurrence paradoxes, popularized by the Ehrenfests, were used by the anti-atomists to attack not only the Statistical Theory but also the Kinetic Theory and Physical Atomism in general."

It is, of course, possible to disagree with some of Ehrenfest's judgements, for instance, his lack of appreciation of the Gibbsian method. Yet the above quotation has a disturbing feature, since Ehrenfest is being reproached for his integrity, for not covering up the weaknesses of the kinetic theory.

The antiatomists mentioned in the same quotation are the author's prime target; they play for him the same role as the caloricists did a half century earlier. The climax of the book is Perrin's verification of Einstein's theory of Brownian motion, which induced the antiatomists to give up their opposition, with the exception of Ernst Mach, "the unrepentant sinner' (Section 8.7). This interpretation of the situation is not peculiar to Brush; it is held also by many physicists. Yet it is unbalanced in two respects.

First, we note that the phenomenological critics of the kinetic theory insisted mainly on the inability of this theory to account for spectroscopy and for chemical binding. The observation of scintillations and of Brownian motion changed nothing in this regard; a solution came about only through the decoding of spectroscopy by means of quantum mechanics.

It is plausible for a history of the classical kinetic theory to end with Brownian motion and not to get involved with the deeper problems of quantum mechanics. Yet it is disingenuous for the author to claim that quantum mechanics removed the equipartition difficulties of the kinetic theory *without changing anything in its conceptual makeup* (pp. 3–5). The fact is that the "atom" of quantum mechanics differs fundamentally from that of the classical theory and it actually satisfies the requirements of the chemically oriented phenomenologists.

The second point to note is that at the beginning of the century there was general agreement on the fact that a phenomenological theory has to be macroscopic. A new era started as Einstein developed his theory of Brownian motion in which statistics and phenomenology were intimately interwoven, as described in detail in Section 15.4. This start was followed by Einstein's theory of critical fluctuations(5): the foundation of statistical thermodynamics. Today the majority of the articles in this Journal deal with applications of statistics to a vast variety of phenomenological models.

It is satisfying that the Brownian motion that convinced many doubters that "atoms exist" also removed the objection to the phenomenological theory as being narrowly macroscopic. This evolutionary dialectic is very different from the confrontation of victors and losers presented by Brush.

Summing up, this huge work has a double aspect. First, there is an impressive amount of historical information, which should be useful for a future synthesis, and second, a methodology based on a priori black-and-white convictions. There is an attempt to cover up the tension between the two by artificial constructions and by stilted logic. Brush refights the battles of the past with a zeal that would be excusable only in the heat of the action. The reviewer believes that the historian-philosopher could better serve the scientist by helping him to defuse rather than to perpetuate the residues of outdated controversies.

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Book Review[1]

Statistical Mechanics, 2nd ed. By J. E. Mayer and M. Goeppert Mayer. John Wiley and Sons, Inc., New York, 1977.

The first edition of *Statistical Mechanics*, published in 1940, was one of the cornerstones of modern statistical thermodynamics. Mayer and Mayer's book and Fowler and Guggenheim's *Statistical Thermodynamics* were the books from which an entire generation of physicists, chemists, and engineers learned the subject. The success of Mayer and Mayer's book was due in part to the authors' own interest in and concern for the subject. At the time the book was written they had already contributed to many of the areas covered in it, and J. E. Mayer, in particular, was immersed in research on the theory of condensation described in Chapter 14 of the first edition. This personal involvement was communicated to the readers, many of whom were stimulated to make their own contribution to the field.

The second edition of Mayer and Mayer's *Statistical Mechanics* is also a very personal book, for it gives us a clear view of the development of their ideas and interests in statistical mechanics over the years since the first edition was published.[2] In addition to the by now standard topics in statistical thermodynamics, the authors have drawn from their own work, particularly that of J. E. Mayer, to present in this edition chapters on dense gases and the theory of condensation, on liquids and electrolyte solutions, and on the use of density matrices in quantum statistical mechanics.

The book is now divided into three parts. The first part is a short treatment of probability theory and of the kinetic theory of dilute monatomic gases, designed to introduce the student to some of the basic ideas and techniques used in statistical mechanics. It is worth mentioning that the chapter on kinetic theory also contains the book's only quantitative discussion of nonequilibrium processes. The second part of the book consists of four chapters, which contain an exposition of the ensembles and equations of

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[1] There will be a response from the book's author in a future issue of the journal.

[2] Maria Goeppert Mayer died in 1972. However, the second edition was planned and outlined by both of the Mayers.

equilibrium statistical mechanics. The authors adhere strictly to the point of view that the fundamental justification for the equilibrium ensembles should not be based on the study of the dynamical motions of single isolated systems, i.e., on ergodic theory, but rather on the assertion that real, laboratory systems are always subject to random perturbations which have the effect of making every possible quantum state of the system equally probable. This assertion of "equal a priori probabilities," referred to in the book as the ergodic hypothesis, serves to introduce the microcanonical ensemble. The connection with thermodynamics is made through the identification of the entropy S with $k_B \ln \Omega$, where k_B is Boltzmann's constant and Ω is the total number of possible quantum states available to the system with specified values of volume, energy, and number of particles. The properties of the other ensembles are then derived from the microcanonical ensemble. In this section of the book, Mayer and Mayer discuss such points as the magnitude of fluctuations about most probable values, the properties of systems such as solid solutions in which the hypothesis of equal a priori probabilities may not always be correct, the Hahn spin-echo experiments, and the statistical interpretation of entropy.

While the ensemble distributions derived in these chapters are standard, I have some reservations about using the idea of random perturbations at the walls to motivate the microcanonical ensemble. First, it is not at all obvious that all "random" perturbations will lead to equal probabilities of occurrence for all quantum states, so the required properties of the random perturbations need to be specified.

A related objection is that in making this assumption one ignores a number of fundamental questions connected with explaining time-dependent processes in statistical mechanics. Suitably defined random perturbations will no doubt drive the system to equilibrium, but I wonder whether such a picture is really necessary. I prefer to think that a proper statistical mechanical description of the approach to equilibrium involves two major steps: the construction of an ensemble which gives the probability distribution for the system at some *fixed* initial time, and then the use of Liouville's theorem to describe the evolution of this probability distribution in time. It seems to me that only by proceeding along such lines will we be able to understand why the final stages of the system's approach to equilibrium can be described by regular laws, such as the hydrodynamic equations. I doubt that stochastic interactions of the system with the boundaries are necessary before the system exhibits hydrodynamic behavior.

The third part of the book consists of chapters on the statistical thermodynamics of specific systems. Most of the chapters in this part are similar to their counterparts in the first edition. These chapters are the ones discussing the thermodynamic properties of perfect and dense gases, of crystalline solids, of ideal quantum gases, and of systems placed in electric or magnetic fields. They are quite good, but suffer somewhat from a lack of suitable figures and graphs to convey the meaning of the calculations more dramatically. The chapter on liquids, which is a new addition, emphasizes the formal properties of the reduced distribution functions for a fluid, rather than specific properties or detailed models of liquid structure. A discussion of the cell model of liquids, present in the first edition, is not retained in the second edition. There are, however, short discussions of the Percus–Yevick and related equations, of recent computer experiments on fluids, and of the Debye–Hückel theory of electrolyte solutions.

The chapter on dense gases (Chapter 3) will probably be of the greatest scientific and historical interest to people working in statistical mechanics, who will wish to compare the discussion of the gas–liquid phase transition in the new edition with that in the old. In view of this interest it seems worthwhile to summarize some of the salient features of the Mayer theory of condensation. This theory was based on the physical idea that as one increased the density of a gaseous system, large clusters of particles would begin to form, and when the density was sufficiently large, these large clusters would begin to contribute significantly to the thermodynamic properties of the system. Eventually, the larger clusters would become so important that the gas would begin to condense and a liquid state would start to form. Thus J. E. Mayer was the first to suggest a mechanism by which one homogeneous phase—the gas—could change into two phases—gas and liquid. To make a theory from this physical picture, Mayer used the virial expansions—both the fugacity expansions and the density expansions—of the thermodynamic properties of the gas, since these expansions directly expressed the properties of interest in terms of the contributions from clusters of interacting particles. Consider, for example, the Mayer expansion for the density n in powers of the fugacity z, which is $n = \sum lb_l z^l$, where the b_l are cluster integrals defined for a system of l interacting particles. This equation has a simple interpretation in terms of clusters, if one uses the fact that $Vb_l z^l$ is the most probable number of clusters with l particles. Thus, "the total average number of particles in the system, $\langle N \rangle = Vn$, is equal to the sum over all cluster sizes of the number of molecules in each cluster, l, times the most probable number of clusters of that size." Mayer argued that for asymptotically large l, b_l should be proportional to $b_0{}^l$, where b_0 is supposed to be independent of l and has the dimensions of a volume. Now, for large l, the contribution of the large clusters to the density is proportional to $(zb_0)^l$. For $|zb_0| \ll 1$ the large clusters do not make any appreciable contribution to the density, but if for some range of temperature the cluster integrals b_l and b_0 are positive, then the large clusters make an increasing contribution to the density as z increases. There then will be a sharp change in the contributions of the large clusters to n as z passes through the value b_0^{-1} from below. This sharp change signals the onset of condensation of the gas. For an infinitely large system, Mayer expected that the virial expansion of the density would have a mathematical singularity at $z = b_0^{-1}$.

By means of this and other, similar arguments, Mayer was able to map out a description of condensation phenomena. He predicted that the critical temperature T_c, defined as the temperature where the gas and liquid phases have the same density, should, in general, be greater than the temperature T_m below which the meniscus separating the two phases appears. According to the Mayer theory, the so-called "derby hat" region between T_m and T_c is such that the gas-phase isotherms approach the coexistence curve in the PV plane with zero slope, i.e., the isothermal compressibility becomes infinite as the coexistence curve is approached from the gas phase. The prediction of the existence of a derby hat region stimulated a number of experimental searches for it. When the first edition of Mayer and Mayer was written, there was in fact some experimental evidence that T_c and T_m could differ by as much as 10–15 K.

At the time of active research on the Mayer theory of condensation, it was generally realized that the theory had a number of difficulties. First of all, one had yet to prove that the requirements on the cluster integrals b_l for condensation were satisfied at low enough temperatures for any system with a realistic intermolecular potential. For the ideal Bose–Einstein gas, Uhlenbeck and Kahn(1) showed that the b_l were all positive, and that they did behave as $b_0{}^l$ for large l. Thus the Mayer theory seemed to be successful in describing the ideal Bose–Einstein condensation, but no other cases were known. Another difficulty was that Mayer's arguments indicated that once the isotherms became horizontal in the two-phase region they would remain horizontal even in the pure liquid phase. However, it was argued that this deficiency in the theory was due to the fact that Mayer had replaced the volume-dependent cluster integrals $b_l(V)$ in the virial expansion by their values at infinite volume $b_l(\infty) = b_l$, thus ignoring a subtle double limiting process as the system's size becomes large.(1)

In spite of the fact that Mayer's theory is a very suggestive one, it is no longer actively pursued, because it is plagued by a number of shortcomings. To begin with, there are no reliable experimental data suggesting that T_m and T_c are different. All the experiments that purported to do so were shown to suffer from impurity and/or gravity effects.[3] Although Mayer's arguments for the derby hat region were strong, they were not at all rigorous, nor could they predict the size of this region. It might have been possible to reconcile Mayer's

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[3] A historical survey of the experimental literature on this subject has been prepared by Levelt Sengers.(2) I would like to thank Dr. Levelt Sengers for her helpful remarks on this subject.

theory with the experimental failure to detect the derby hat, but in 1952 Yang and Lee presented their rigorous theory of phase transitions which showed that Mayer's theory could not be a general description of the liquid–gas phase transition. Yang and Lee considered the analytic properties of the grand canonical partition function as a function of the fugacity z and showed that the phenomena of phase transition can be associated with the distribution of zeros of the grand canonical partition function in the complex z plane. As consequences of the Yang and Lee description, it was possible to show that:

(a) If Mayer's fugacity expansions have a singularity on the z axis, this singularity need not coincide with the actual value of z where the condensation takes place, but may be larger than the actual value. Here the difference is due to the improper $V \to \infty$ limit procedure alluded to earlier.

(b) The fugacity and density expansions and their analytic continuations, if such are possible, have in general no meaning outside the gas-phase region. In fact, a study of these expansions and their analytic continuations for a system that does undergo a phase transition may not even indicate that a condensation occurs. Ford(3) has constructed a mathematical model of a grand canonical partition function which has many features of a real gas partition function. In this model one can sum the fugacity series for the density and study its analytic continuation outside the small-z region. The fugacity series for the density and its analytic continuation have *no* singularities on the positive z axis, while in actuality there is a singularity in $n(z)$ at $z = 1$. For $z < 1$, the fugacity expansion of n and the actual value of n coincide, but not for $z \geqslant 1$.

There have been a number of attempts to redo the Mayer theory more carefully, and to find conditions on the potential energy of the system such that the Mayer expansions would predict the correct location of the singularity on the positive z axis.[4] These attempts have not been very successful, and the Mayer theory is no longer an active area of research in statistical mechanics.

The second edition provides an exposition of those features of Mayer's theory that have held up over the intervening years. That is, the virial expansions are derived, and the volume dependence of the cluster integrals is discussed, as is the relation between the cluster integrals and actual physical clusters. Then Mayer gives a careful treatment of the analytic properties of the virial expansion in light of the Yang–Lee theory. The presentation is along the lines developed in the first edition and anyone who is familiar with the Mayer theory can find here and there some echoes of the derby hat region. Nevertheless, this chapter is sufficiently clear and self-contained that it is worth studying on its own merits. The chapter on dense gases would be of

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[4] For a useful summary of the Mayer theory and of later attempts to improve it, see Münster.(4)

more general interest to the student, though, if some more modern aspects of the theory of phase transitions were presented. No mention is made of critical exponents, scaling laws, and exactly-soluble models, which presently play an important role in this area.

Finally, I am sorry that the authors did not choose to discuss nonequilibrium statistical mechanics. This area of statistical mechanics has become an active field of research over the past few decades. Theories of transport phenomena in fluids and solids are certainly of interest to the students for whom the book is addressed, and some discussion of them in a text on statistical mechanics would not have been inappropriate.

Readers who are considering using the second edition of Mayer and Mayer as a textbook should be aware that no problems or exercises are provided. In addition, some of the sections in Part 2 which deal with the foundations of the theory are not very lucidly written and may be quite difficult for a beginning student to understand. Mayer and Mayer suggest that such readers skip the more difficult sections and return to them after the main outlines of the theory have been assimilated. In addition, there is a noticeable lack of illustrations as well as of discussions of recent applications of the theory. Considering the fact that the first edition appeared in 1940, it is regrettable that a more modern discussion of such topics as virial coefficients, or equations of state, for example, do not appear in this new edition. Nevertheless, those teaching statistical mechanics and those working in the field will appreciate the Mayers' facility in performing the calculations and will benefit from the discussions of interesting details of the process. For this reason, and for its careful presentation of many areas of statistical thermodynamics, the second edition of *Statistical Mechanics* can be recommended as a supplementary text for students and as a resource text for teachers of statistical thermodynamics.

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