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Two innovative equivalent statements of the third law of thermodynamics

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It is found from textbooks and literature that there are three different statements for the third law of thermodynamics, i.e., the Nernst theorem, the unattainability statement of absolute zero temperature, and the heat capacity statement. It is pointed out that such three statements correspond to three thermodynamic parameters, which are, respectively, the entropy, temperature, and heat capacity, and can be obtained by extrapolating the experimental results of different parameters at ultra-low temperatures to absolute zero. It is expounded that because there is no need for additional assumptions in the derivation of the Nernst equation, the Nernst theorem should be renamed as the Nernst statement. Moreover, it is proved that both the Nernst statement and the heat capacity statement are mutually deducible and equivalent, while the unattainability of absolute zero temperature is only a corollary of the Nernst statement or the heat capacity statement so that it is unsuitably referred to as one statement of the third law of thermodynamics. The conclusion is that the Nernst statement and the heat capacity statement are two equivalent statements of the third law of thermodynamics.

Keywords: Nernst statement, heat capacity statement, Nernst theorem, absolute zero temperature, the third law of thermodynamics

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1. Introduction

As a fundamental law of thermodynamics, there may be different statements, but the statements must be mutually derivable and equivalent. For example, the second law of thermodynamics can be expressed by many different statements, the two most common of which are the Clausius statement and the Kelvin–Planck statement.[1–4] They have been proved to be equivalent to each other in many textbooks. The third law of thermodynamics can also be expressed by different statements. The Nernst theorem and the unattainability statement of absolute zero temperature are two common statements in textbooks.[2,3,5–7] They have been proved to be mutually equivalent in many textbooks,[2,3,7] but considered not to be equivalent in several textbooks[8–11] and literature.[12–14] Up to now, the equivalence of the two statements has been a controversial issue.[15–20]

In many thermodynamics books, it has further been proved that the Nernst theorem can be directly used to derive an important result that the heat capacities tend to zero as the temperature approaches absolute zero,[2,21] but it has not been proved that the former can be derived from the latter. It is even asserted in several books[22] that the former cannot be derived from the latter. After 1988, some researchers proved that the former can be directly derived from the latter through different methods.[23,24] Those proofs caused some controversies.[16,25,26] Gujrati[27] generalized the Nernst theorem and expounded that the vanishing heat capacity at $T = 0$ is directly related to vanishing susceptibilities so that the generalized Nernst theorem reduces to the original Nernst theorem. Recently, the conclusions in Refs. [23,24] were further generalized to more general situations.[28,29] This shows that the equivalence of the Nernst theorem and the vanishing heat capacity at $T = 0$ is another controversial issue.

Besides the two controversial issues mentioned above, the relationship between the vanishing heat capacity at $T = 0$ and the unattainability statement of absolute zero temperature was rarely discussed. The role of the vanishing heat capacity at $T = 0$ in the equivalence proof on both the Nernst theorem and the unattainability statement of absolute zero temperature has not been elucidated. Thus, clarifying the interrelationships of three statements: the Nernst theorem, the unattainability statement of absolute zero temperature, and the heat capacity statement, will be helpful to the understanding of the essence of the third law of thermodynamics.

2. Three statements

Three different statements of the third law of thermodynamics are listed as follows.

The Nernst theorem. It can be stated as follows.[2] The entropy change ΔS associated with any isothermal reversible process of a condensed system approaches zero as the temperature T approaches absolute zero, i.e.,

$$\lim_{T\to 0}(\Delta S)_T = 0, \tag{1}$$

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which is the Nernst equation. Equation (1) indicates that the Nernst theorem is directly related to the entropy S of a system. Before 1906, many chemists had investigated the chemical reactions of thermodynamic systems at ultra-low temperatures, and measured the changes ΔH and ΔG of the enthalpy H and Gibbs function G of thermodynamic systems at isothermal and isobaric pressures. They found that both ΔH and ΔG get closer and closer as the temperature drops. When the experimental results are extrapolated to absolute zero temperature, one may get $\Delta G = \Delta H$. Using those experimental results and introducing some assumptions, Nernst derived the heat theorem, which was later modified and generalized by Planck, Simon, and others[2] and became known as the Nernst postulate or Nernst theorem. Recently, it was found[30] that the derivation of the Nernst equation does not need those additional assumptions appearing in textbooks. The physical content included by Nernst's equation should not be referred to as Nernst's postulate or Nernst's theorem. It should be renamed as the Nernst statement, i.e., the entropy change associated with any isothermal reversible process of a thermodynamic system approaches zero as the temperature approaches absolute zero. This statement is similar to that in textbooks, but it does not encompass the characteristics of the entropy change of a thermodynamic system during an isothermal process in the region of $T > 0$. Thus, the more perfect statement for the physical connotations of the Nernst equation is listed as follows. The entropy change of a thermodynamic system during an isothermal process diminishes as the temperature decreases, and as the absolute temperature approaches zero, the isothermal process tends to be reversible, with the entropy change of the system approaching zero. This refined statement is characterized by greater accuracy compared to conventional textbook descriptions.

The unattainability statement of absolute zero temperature. It is stated as follows.[2] It is impossible by means of any procedure, no matter how idealized, to reduce the temperature of a system to the absolute zero in a finite number of steps. It can be mathematically expressed as

$$-(\Delta T)/T < 1. \tag{2}$$

Because $+0$ K is the low limit of the absolute temperature,[1,2,31,32] $-(\Delta T)$ is impossibly larger than T for any adiabatic cooling process starting from an arbitrary low temperature. The lower the temperature in any cooling process is, the more difficult to cool it down. At present, spin gradient cooling[33] is permitted to reach the internal kinetic energy below 350 pK. Employing matter-wave lenses based on magnetic[34] and electrostatic[35] forces make it possible to reduce the internal kinetic energy of a Bose–Einstein condensation system to about 50 pK.[36] Although the absolute zero temperature cannot be attained, it does not negate that the absolute zero temperature can be infinitely approached.[37] The above analyses show that the unattainability statement of absolute zero temperature is a direct conclusion of temperature measurement.

The heat capacity statement. It is stated as follows.[28,29] The various heat capacities of a thermodynamic system under different constraints approach to zero as $T \to 0$, i.e.,

$$\lim_{T\to 0} C_x = 0. \tag{3}$$

It is worth pointing out that C_x represents the various heat capacities of the real thermodynamic systems at ultra-low temperatures, but does not include the fictional heat capacities[25,38,39] impossibly appearing in real thermodynamic systems at ultra-low temperatures. C_x has a common characteristic. It decreases with the decrease of T. With the development of cryogenic technology, the heat capacities of many thermodynamic systems at ultra-low temperatures such as quantum Fermi gas, electron gas in metal, quantum Bose gas, superfluid, etc. have been experimentally measured. When those experimental data are simulated and extrapolated to absolute zero temperature, one can obtain Eq. (3).

By looking at the above three statements only from the experiment, they are independent of each other. However, it is very meaning to note that there are certain connections for the three statements because they correspond to the entropy, temperature, and heat capacity of a system, respectively. Using the theory of thermodynamics, we will explain clearly the interrelationships of the three statements mentioned above and answer Blau and Halfpap's question:[15] what is the third law of thermodynamics trying to tell us?

3. Heat capacity and entropy of a thermodynamic system

The internal energy U of a thermodynamic system in many textbooks[5,40–42] is expressed as

$$\mathrm{d}U = T\,\mathrm{d}S + \sum_{i=1}^{n} Y_i\,\mathrm{d}y_i, \tag{4}$$

where y_i and Y_i are the generalized coordinates and corresponding generalized forces,[28] respectively, and n is the number of generalized coordinates. Using Eq. (4) and one thermodynamic relation $H = U - \sum_{j=1}^{l} y_j Y_j$, we can obtain

$$C_x = (\partial H/\partial T)_x = T(\partial S/\partial T)_x, \tag{5}$$

where x represents $(n-l)$ generalized coordinates and l is the generalized forces. C_x is the heat capacity when x remains unchanged. Equation (5) includes the various heat capacities of a thermodynamic system under different constraints. For example, C_x includes C_y as well as C_Y, where C_y is the heat capacity[23] when y is invariant and C_Y is the heat capacity when Y is invariant; the heat capacity at constant volume C_v and the heat capacity at constant pressure C_P often appearing

in textbooks[2–4] and literature[25,43] are two simplest forms of C_x.

Using Eq. (2), one can obtain the expression of the entropy of a thermodynamic system as[28]

$$S = S(T,x) = S(T_0,x) + \int_{T_0}^{T} (C_x/T)\,\mathrm{d}T, \tag{6}$$

where $S(T_0,x)$ is the entropy of the system when $T = T_0$.

4. The equivalence of Eqs. (1) and (3)

Using Eq. (1) and another thermodynamic relation $G = H - TS$, one can directly derive Eq. (3).[28] This shows clearly that Eq. (3) is a direct consequence of the Nernst statement and a universal result of thermodynamics.

When $T = 0$, combining Eq. (6) with Eq. (3) can be written as[2,3,28,44]

$$S = S(0,x) + \int_{0}^{T} C_x/T\,\mathrm{d}T, \tag{7}$$

where $S(0,x)$ is the entropy of a thermodynamic system at $T = 0$. If Eq. (3) is invalid, the integral $\int_0^T C_x/T\,\mathrm{d}T$ in Eq. (7) is divergent and Eq. (7) is physically unmeaning. In order to discuss the relationship between the Nernst statement and the vanishing heat capacity at $T = 0$, Landsberg took the heat capacities of ideal gases as an example. Even assuming[25] that the internal energy of ideal gases is a function of temperature, the heat capacities given in Ref. [25] still do not meet Eq. (3). Although the heat capacity not satisfying Eq. (3) may be taken as the expression of a thermodynamic system at other temperature regions, it is impossible at ultra-low temperatures. It should also be pointed out that some fictional heat capacities cannot appear in real thermodynamic systems at ultra-low temperatures, they may satisfy Eq. (3), but they cannot ensure the integral $\int_0^T C_x/T\,\mathrm{d}T$ in Eq. (7) to be convergent.[39] Such fictional heat capacities[38] are unmeaning and cannot be used to discuss the performance of thermodynamic systems at ultra-low temperatures. For example, the heat capacity $C(T) = c_0/[\ln(1+T_0/T)]$ given in a textbook[38] is typically fictional, where c_0 is a positive constant. It has been found by lots of experiments and theories that the expressions of the heat capacity appearing in ultra-low temperature systems not only satisfy Eq. (3) but also ensure the integral $\int_0^T (C_x/T)\,\mathrm{d}T$ in Eq. (7) to be a finite value.

Equation (7) has been used to prove[28] that Eq. (1) can be directly derived from Eq. (3). So far it has proved that Eqs. (1) and (3) are mutually deducible and equivalent.

Below, it will be further proved through a simple method that the Nernst statement and the heat capacity statement are equivalent. When a thermodynamic system starts from the state $\{T,x\}$ and attains the state $\{T+\Delta T, x+\Delta x\}$, the entropy change of the system in such a process is given by $\Delta S = S(T+\Delta T, x+\Delta x) - S(T,x)$, i.e.,

$$\Delta S = (\Delta S)_x + (\Delta S)_T, \tag{8}$$

where $(\Delta S)_x$ is the entropy change caused by a change of T when x remains constant and $(\Delta S)_T$ is the entropy change caused by a change of x when T remains constant. When the process is reversible adiabatic, $\Delta S = 0$ and the temperature change of the system is $(\Delta T)_S$. When $T \to 0$, Eq. (8) can be expressed as

$$\lim_{T\to 0} \frac{-(\Delta T)_S}{T} = \lim_{T\to 0} \frac{(\Delta S)_T}{C_x}. \tag{9}$$

By using Eqs. (1), (3), and (9), the equivalence of both the Nernst statement and the heat capacity statement can be easily proved.[29] It is worth pointing out that the above proof process does not involve the unattainability statement of the absolute zero temperature.

5. Equation (2) is only a corollary of Eq. (1) or Eq. (3)

For a thermodynamic system evolving along a quasistatic reversible adiabatic process, T changes from T_1 to T_2, and x changes from x' to x'', whereas the entropy S remains unchanged. Using Eq. (7), we can obtain

$$S(0,x'') - S(0,x') = F(x',T_1) - F(x'',T_2), \tag{10}$$

where $F(x,T) = \int_0^T (C_x/T)\,\mathrm{d}T$. It is meaning to note that Eq. (10) only includes Eq. (3) but does not include the Nernst equation.

According to the Nernst equation, Eq. (10) can be simplified as[45]

$$F(x',T_1) = F(x'',T_2). \tag{11}$$

It is seen from Eq. (11) that when $T_1 > 0$, $F(x',T_1) > 0$ and T_2 cannot attain zero, i.e., the unattainability statement of absolute zero temperature. It should be pointed out that in the whole derivative process, Eqs. (1) and (3) have been applied. This shows clearly that the unattainability statement of absolute zero temperature can be directly derived from Eq. (1) or Eq. (3) because both Eqs. (1) and (3) are mutually deducible.

The other proof method commonly used in thermodynamics,[2] i.e., a proof by contradiction, is given as follows. If the Nernst statement is assumed not to be true, the first case is to assume $S(0,x'') > S(0,x')$ in Eq. (10), so that one can find a positive T_1 to ensure that $F(x',T_1)$ is small enough, $S(0,x'') - S(0,x') = F(x',T_1)$, and $T_2 = 0$; the second case is to assume $S(0,x'') < S(0,x')$ in Eq. (10), the reversible adiabatic process goes in reverse, and a positive T_2 can be found to ensure that $F(x',T_2)$ is small enough, so that $S(0,x') - S(0,x'') = F(x'',T_2)$ and $T_1 = 0$. The above proofs show that only if the Nernst statement is not true, can the absolute zero temperature attain. This proves once again that

Eq. (1) or Eq. (3) can be used to derive the unattainability statement of absolute zero temperature, while no any additional assumptions are required.

The third derivation method is also briefed. Using the Nernst statement and Eq. (7), one can plot the temperature–entropy diagram,[2,3,37] which is directly used to prove that the unattainability statement of absolute zero temperature can be derived from the Nernst equation.

Although the unattainability statement of absolute zero temperature does not imply the Nernst theorem has long ago been pointed out,[12] there has been no consensus on this view. For example, in some textbooks[2,7] and articles,[44] Eq. (10) is directly used to prove that the Nernst theorem can be derived from the unattainability statement of absolute zero temperature. It is natural because Eq. (3) has been applied to Eq. (10) and the Nernst equation can be directly derived from Eq. (3). This issue has not been noted in most textbooks. Without the aid of Eq. (3), no mathematical connection can be established between the Nernst equation and the unattainability statement of absolute zero temperature, let alone that the former can be derived from the latter.

The above analyses show that only when the substantive role of the heat capacity statement of the third law of thermodynamics is clearly expounded, can the issue of whether both the Nernst statement and the unattainability statement of absolute zero temperature are equivalent or not be deeply recognized.

In fact, it is intuitively seen from the mathematical structures of Eqs. (1) and (2) that both the Nernst statement and the unattainability statement of absolute zero temperature cannot be equivalent. It is also seen from the physical contents of Eqs. (1) and (2) that the two statements mentioned above cannot be equivalent, because the Nernst equation may be used to discuss the performance of thermodynamic systems at $T = 0$, while the unattainability statement of absolute zero temperature has never been involved in the performance of thermodynamic systems at $T = 0$.

So far it has been expounded that neither the Nernst statement nor the heat capacity statement can be independently derived from the unattainability of absolute zero temperature. It shows clearly that the unattainability statement of absolute zero temperature is only a corollary of the Nernst statement or the heat capacity statement. Both the unattainability statement of absolute zero temperature and the Nernst statement are not equivalent. Of course, both the unattainability statement of absolute zero temperature and the heat capacity statement are also not equivalent.

6. Two equivalent statements of the third law of thermodynamics

Like the Clausius statement and the Kelvin–Planck statement[4,7,45] as two statements of the second law of thermodynamics, the Nernst statement and the heat capacity statement can be used as two statements of the third law of thermodynamics so that the core contents of the third law of thermodynamics are a true reflection of the objective world without any artificial additional assumptions. The two innovative equivalent statements proved above will play an important role in improving the theoretical framework of thermodynamics. This discovery can effectively prevent some artificial assumptions into the third law of thermodynamics, making which be a true reflection of the objective world. It solves the awkward problem caused by using the Nernst theorem and/or the unattainability statement of absolute zero temperature as the statements of the third law of thermodynamics for over one hundred years.

7. Conclusions

Three different statements of the third law of thermodynamics corresponding to the entropy, temperature, and heat capacity can be summed up by measuring the different parameters of a system at ultra-low temperatures and extrapolating the experimental results to absolute zero temperature.

The derivation process of the Nernst equation in textbooks should be rewritten because no artificial additional assumptions are required. The Nernst postulate or Nernst theorem in textbooks should be renamed as the Nernst statement.

It has been proved through the theory of thermodynamics that among the three statements of the third law of thermodynamics appearing in textbooks and literature, the Nernst statement and the heat capacity statement are equivalent to each other, while the unattainability statement of absolute zero temperature is only a corollary of the Nernst statement or the heat capacity statement. Strictly speaking, only the Nernst statement and the heat capacity statement can be used as the statements of the third law of thermodynamics.

For the heat capacity statement of the third law of thermodynamics, the corresponding heat capacity must be the real heat capacity of thermodynamic systems at ultra-low temperatures rather than the fictional heat capacity impossibly appearing in ultra-low temperature systems. It is unmeaning to use the fictional heat capacity to discuss the performance of thermodynamic systems.

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