Heat Capacity Anomalies of Superfluid ^{4}He under the Influence of a Counterflow near T_λ

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We present a thermodynamic treatment of superfluid helium in the presence of an applied heat current, Q, which produces a counterflow velocity $\vec{W}$. We show that the heat capacity can be expressed in terms of the dependence of the superfluid density on $\vec{W}$. Near T_λ, both mean field theory and renormalization group theory give a divergent heat capacity with an exponent of 0.5 at a depressed transition temperature. In contrast, if $\vec{W}$ rather than Q is held constant, the heat capacity remains finite. [S0031-9007(96)00890-3]

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Owing to the remarkable success of the renormalization group theory (RG), phase transitions at equilibrium are to a large extent well understood. There is still much to be learned, however, concerning nonequilibrium and dynamic phenomena. Near the lambda point of ^{4}He, an applied heat flux Q can have an interesting influence on the nature of the transition. A number of experiments [1] report that the transition temperature is depressed. The depressed transition temperature $T_c(Q)$ scales with Q as $T_\lambda - T_c(Q) \sim Q^x$. Typical values are $T_\lambda - T_c = 1.1\ \mu\text{K}$ at $Q = 10\ \mu\text{W/cm}^2$. Theories [2] predict that $x = 1/(2\nu) = 0.746$, where $\nu = 0.6705$ [3] is the correlation length exponent. Recently, Haussmann and Dohm (HD) [4] have applied RG to this problem and predicted cusp shaped curves [5] for the superfluid density and the heat capacity at constant superfluid velocity, $\vec{v}_s$, for various values of Q near $T_c(Q)$. In this paper we show the surprising result that if Q is held constant instead of $\vec{v}_s$, the heat capacity diverges at $T_c(Q)$, even in mean-field theory. HD have independently discovered this same result [6]. In this paper we present the new discovery and clarify the thermodynamics of this interesting system.

Liquid helium in the presence of a counterflow can be treated as a system that exhibits an extra degree of thermodynamic freedom. This is a unique case in which a dynamic situation may be treated by equilibrium thermodynamic analysis. According to the two-fluid model, the first law of thermodynamics at constant density may be written unambiguously for a unit volume in the superfluid frame as [7]

$$dE^s = Tds + \vec{W} \cdot d\vec{j}_0 \,, \tag{1}$$

where E^s is the energy, $\vec{W} = \vec{v}_n - \vec{v}_s$ is the velocity of the normal fluid in that frame, and $\vec{j}_0 = \rho_n \vec{W}$ is the normal fluid momentum density. The term $\vec{W} \cdot d\vec{j}_0$ is the work per unit volume required to set the normal fluid into motion. Thus the new conjugate variables in the superfluid frame are $(\vec{W}, \vec{j}_0)$. Most phase transition theories, to which we wish to compare our results, assume that the normal fluid is at rest. The internal energy in the normal fluid frame E^n can be obtained using the Galilean transformation [7] $E^n = E^s + \rho \vec{W}^2/2 - \vec{j}_0 \cdot \vec{W}$, giving

$$dE^n = Tds + \vec{P} \cdot d\vec{W} \,, \tag{2}$$

where $\vec{P} = \rho_s \vec{W}$. Thus in the normal fluid frame the new conjugate pair is $(\vec{P}, \vec{W})$. The free energy is $F(T, \vec{W}) = E^n - Ts$ giving

$$\begin{aligned} dF &= -sdT + \vec{P} \cdot d\vec{W} \,, \\ F(T, \vec{W}) &= F(T, 0) + \int_0^{\vec{W}} \rho_s(\vec{W}) \vec{W} \cdot d\vec{W} \,. \end{aligned} \tag{3}$$

We henceforth drop the vector notation because all motions are in the same direction in the case we treat. The term $F(T, 0)$ contains all the characteristics of the phase transition at zero W, which has been well studied both experimentally and theoretically. At finite W the only unknown is the function $\rho_s(W)$. Qualitatively, if ρ_s is a weak function of W, the integral in Eq. (3) can be approximated by $\rho_s(0)W^2/2$. The dashed line in Fig. 1 shows $F(T, W)$ for this case. On the other hand, if ρ_s is significantly depressed [Fig. 1(a)], the integrand in Eq. (3), $\rho_s(W)W$, increases with W at small W, but might decrease at large W [Fig. 1(b)]. As shown by the solid line in Fig. 1(c), a critical counterflow velocity W_c exists when $F(T, W)$ changes from convex to concave [8]. This is also the point where $\rho_s(W)W$ is maximum. If $\rho_s(W)$ is sufficiently depressed to reach this point, superflow breaks down [4].

The depression of ρ_s cannot be derived by thermodynamic arguments. It must be measured experimentally, calculated from microscopic theory, or obtained from phase transition theory near T_λ. Experimentally, not much is known about $\rho_s(W)$. The only experimental evidence to date is the observation by Hess [9] far from T_λ, which agrees with roton theory. Near T_λ, only theoretical predictions exist. The three existing theories are the mean-field theory [10] which we modify by using empirical exponents, the ψ theory [11], and the RG theory of HD [4]. Since we will use the $\rho_s(W)$ expression from these

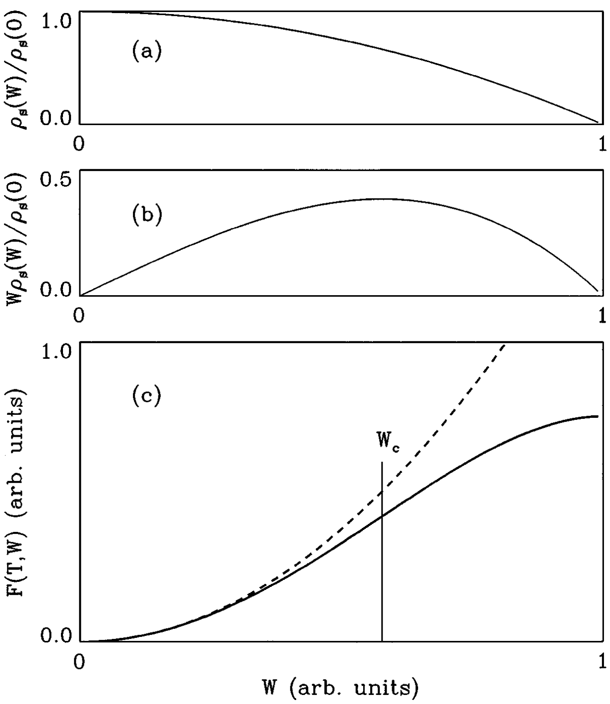

FIG. 1. This illustration discussed in the text is calculated using the mean-field theory.

theories to compute the heat capacity, it is desirable to show that the theories are consistent with thermodynamics. These theories all start from a mean-field expansion,

$$F_{\mathrm{mf}} = \alpha|\psi|^2 + \beta|\psi|^4 + (\hbar^2/2m)|\nabla\psi|^2 + M|\psi|^6. \quad (4)$$

It is not clear at this point how F_{mf} is related to $F(T,W)$ in Eq. (3). Here α, β, and M are expansion coefficients, M is zero except in the ψ theory, the macroscopic wave function is given by $\psi = \eta e^{i\phi}$, where $\rho_s = m|\psi|^2$ and $v_s = (\hbar/m)\nabla\phi$, and m is the mass of a helium atom. In terms of ρ_s and v_s,

$$F_{\mathrm{mf}} = \frac{\alpha\rho_s}{m} + \frac{\beta\rho_s^2}{m^2} + \frac{\rho_s v_s^2}{2} + \frac{\hbar^2(\nabla\rho_s)^2}{8m^2\rho_s} + \frac{M\rho_s^3}{m^3}. \quad (5)$$

A controversy exists in the literature concerning the proper procedure for minimizing F_{mf} with respect to ψ (or ρ_s). Pitaevskii [12] minimizes $F^L_{\mathrm{mf}} = F_{\mathrm{mf}} + \rho_n v_n^2/2$ while holding the momentum $j = P + \rho v_n$ constant. Here, F^L_{mf} is a free energy in the laboratory frame. Khalatnikov [13] uses a Galilean transformation to obtain a free energy in the normal fluid frame,

$$F^n_{\mathrm{mf}} = \frac{\alpha\rho_s}{m} + \frac{\beta\rho_s^2}{m^2} + \frac{\rho_s W^2}{2} + \frac{\hbar^2(\nabla\rho_s)^2}{8m^2\rho_s} + \frac{M\rho_s^3}{m^3}. \quad (6)$$

He then minimizes F^n_{mf} holding W constant. To show that this is the correct approach, we note that F^n_{mf} varies with W through $\rho_s(W)$ and the term $\rho_s W^2/2$. Thus

$$\frac{dF^n_{\mathrm{mf}}}{dW} = \left.\frac{\partial F^n_{\mathrm{mf}}}{\partial\rho_s}\right|_W \frac{d\rho_s(W)}{dW} + \left.\frac{\partial F^n_{\mathrm{mf}}}{\partial W}\right|_{\rho_s}. \quad (7)$$

From Eq. (6), $(\partial F^n_{\mathrm{mf}}/\partial W)_{\rho_s} = \rho_s(W)W$. The optimization condition is $(\partial F^n_{\mathrm{mf}}/\partial\rho_s)_W = 0$. Thus Eq. (7) and Eq. (3) become the same, proving consistency with thermodynamics.

In uniform flow, $\nabla\rho_s = 0$. The expression for $\rho_s(W)$ is obtained by optimizing F^n_{mf}. All three theories give $\rho_s(W)$ of the form

$$\rho_s(W) = \rho_s(0)f(\kappa), \quad (8)$$

where $\kappa = W/W_t$ and W_t is a characteristic velocity given by $W_t = \hbar/m\xi$. Below T_λ, $\xi = \xi_0(2t)^{-\nu}$, where $\xi_0 = 1.43 \times 10^{-8}$ cm [14]. The characteristic velocity W_t can be expressed as $W_t = W_0 t^\nu$, where $W_0 = \hbar 2^\nu/m\xi_0 = 175.4$ m/sec. For the mean-field theory, $f(\kappa) = 1 - 2\kappa^2$. For the ψ theory,

$$f(\kappa) = -\frac{1-M}{2M} + \frac{1}{2}\sqrt{\left(\frac{1-M}{M}\right)^2 + \frac{4}{M}\left(1 - \frac{6+2M}{3}\kappa^2\right)}.$$

For HD, $f(\kappa)$ is given by Eqs. 5.12, C11, and C3 in Ref. [4]. All three theories predict that ρ_s is sufficiently depressed to cause superflow to break down.

Next we compute the heat capacity using $\rho_s(W)$ from these theories. We first treat the case where W is held constant. Experimentally, this might be the case of a persistent superfluid current flowing around a loop, similar to the superfluid gyroscope experiment demonstrated by Clow and Reppy [15]. From Eq. (3) above

$$\Delta F(T,W) = F(T,W) - F(T,0) = \rho_s(0)W_t^2 \int_0^\kappa xf(x)\,dx. \quad (9)$$

The heat capacity is changed by $\Delta C_W = -TV \times \partial^2\Delta F(T,W)/\partial T^2|_W$, where $V = 27.38$ cm^3/mole [16] is the molar volume. Using $\rho_s(0) = \rho_0 t^\zeta$, where $\rho_0 = 0.370$ g/cm^3 [17], together with the scaling relation $\zeta = \nu = (2-\alpha)/3$, we obtain

$$\Delta C_W t^\alpha = -C_0\nu\left[3(3\nu - 1)\int_0^\kappa xf(x)\,dx - (4\nu - 1)\kappa^2 f(\kappa) + \nu\kappa^3\frac{\partial f(\kappa)}{\partial\kappa}\right], \quad (10)$$

where $C_0 = V\rho_0 W_0^2/T_\lambda = 143$ J/mole K. For the mean-field theory, this reduces to

$$\Delta C_W t^\alpha = C_0\nu\kappa^2[(1-\nu) + (1+\nu)\kappa^2]/2. \quad (11)$$

For the ψ theory and for HD, Eq. (10) is evaluated using numerical differentiation and integration. These results are shown in Fig. 2(a). C_W approaches a finite constant at $\kappa_c = W_c/W_t$ in all three theories. As discussed above,

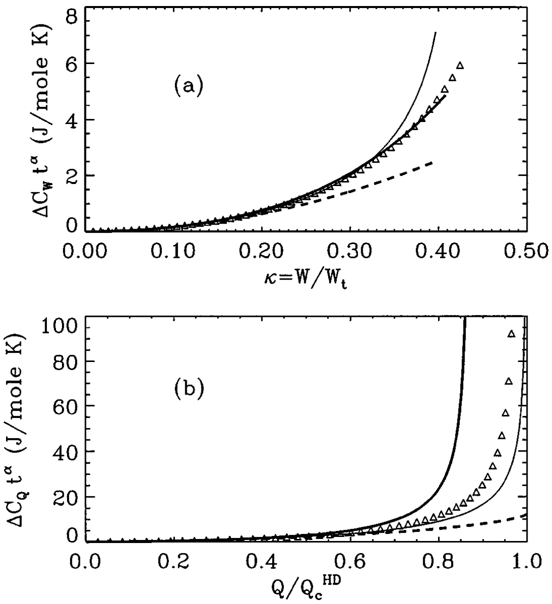

FIG. 2. Change in the heat capacity times t^α at (a) constant W, and (b) constant Q. Thin line, HD theory; thick line, mean-field theory; triangles, ψ theory with $M = 1$; dashed line, ρ_s not depressed by W as discussed in Ref. [6].

superflow also produces a small shift in the transition temperature in all three theories.

It is experimentally feasible to measure the heat capacity in a thermal conductivity cell while passing a constant heat flux Q through it, where

$$Q = \rho_s(W)WST, \tag{12}$$

and $S = 1.58$ J/g K [18] is the entropy per gram. Therefore, keeping Q constant is the same as keeping $P = \rho_s(W)W$ constant. At constant P, it is convenient to define $\Phi(T,P) = F(T,W) - WP$, giving $d\Phi = -sdT - WdP$ and $\Delta\Phi(T,P) = \Phi(T,P) - \Phi(T,0) = -\int_0^W Wd(\rho_s W)$. The heat capacity can be computed from

$$\Delta C_Q = -TV[\partial^2\Delta\Phi(T,P)/\partial T^2]_Q. \tag{13}$$

Although ΔC_W is finite, ΔC_Q diverges at $T_c(Q)$. The reason may be seen directly from thermodynamics. Starting from the entropy density $s(T,W)$, we obtained the relations

$$ds = (\partial s/\partial T)_W dT + (\partial s/\partial W)_T dW, \tag{14}$$

$$\begin{aligned} C_Q &= TV(\partial s/\partial T)_Q \\ &= C_W + TV(\partial s/\partial W)_T(\partial W/\partial T)_Q. \end{aligned} \tag{15}$$

From Eq. (3), $dF = -sdT + PdW$, we obtained a Maxwell relation $(\partial P/\partial T)_W = -(\partial s/\partial W)_T$. Thus,

$$C_Q = C_W - TV(\partial P/\partial T)_W(\partial W/\partial T)_P. \tag{16}$$

Here we have made use of Eq. (12) to obtain the relation $(\partial W/\partial T)_Q = (\partial W/\partial T)_P$. Using the chain rule

$$(\partial P/\partial T)_W(\partial T/\partial W)_P(\partial W/\partial P)_T = -1, \tag{17}$$

$$C_Q = C_W + TV(\partial P/\partial T)_W^2/(\partial P/\partial W)_T. \tag{18}$$

Superflow breaks down when $(\partial^2 F/\partial W^2)_T = (\partial P/\partial W)_T = 0$. Thus C_Q diverges at this point.

This result must be true for any theory that depresses ρ_s enough to reach $(\partial P/\partial W)_T = 0$, including all three theories discussed here. Equation (18) gives

$$\begin{aligned} \Delta C_Q = \Delta C_W &+ C_0 t^{-\alpha}\nu^2\kappa^2 \\ &\times \left[\frac{\kappa\partial f(\kappa)}{\partial\kappa} - f(\kappa)\right]^2 \Big/ \frac{\partial\kappa f(\kappa)}{\partial\kappa}. \end{aligned} \tag{19}$$

The results can be expressed in terms of the variable $q = Q/Q_c$ using the relation $q = \kappa f(\kappa)/[\kappa_c f(\kappa_c)]$ obtained from Eq. (12). The values for κ_c, $f(\kappa_c)$, and $Q_c/t^{2\nu}$ are listed in Table I. For the mean-field theory

$$\begin{aligned} t^\alpha \Delta C_Q & \\ &= C_0\nu\kappa^2\left[\frac{(\nu + 1) + 5(3\nu - 1)\kappa^2 + 2(\nu - 3)\kappa^4}{2(1 - 6\kappa^2)}\right] \\ &= (C_0/2)\nu(\nu + 1)\kappa_c^2 f^2(\kappa_c)q^2[1 + 0.965q^2 + \cdots], \end{aligned} \tag{20}$$

at small q. Figure 2(b) shows that all three theories give a divergent C_Q. Again the results for the ψ theory and the HD theory are obtained numerically. Because Q_c is different for the three theories, we have used Q/Q_c^{HD} as the x axis, so that all three theories can be plotted on the same scale. Here Q_c^{HD} is the critical heat current given by HD. Near Q_c, Eq. (18) gives $C_Q \sim 1/(\partial P/\partial W)_T$. We can expand P about P_c, the superfluid momentum at the phase transition:

$$\begin{aligned} P = P_c &+ \left(\frac{\partial P}{\partial W}\right)_{W_c}(W - W_c) \\ &+ \frac{1}{2}\left(\frac{\partial^2 P}{\partial W^2}\right)_{W_c}(W - W_c)^2 + \cdots. \end{aligned} \tag{21}$$

Since $(\partial P/\partial W)_{W_c} = 0$, and $(\partial^2 P/\partial W^2)_{W_c} < 0$, $P_c - P \sim (W_c - W)^2$, and $(\partial P/\partial W)_T \sim W_c - W$. Thus,

$$C_Q \sim 1/(W_c - W) \sim 1/\sqrt{P_c - P} \sim (Q_c - Q)^{-u}, \tag{22}$$

where the exponent $u = 0.5$. We have verified numerically that all three theories are consistent with this prediction. It is easy to show that if we define $\theta = [T_c(Q) - T]/T_c(Q)$, then

$$C_Q \sim \theta^{-u}. \tag{23}$$

TABLE I. A summary of κ_c, $f(\kappa_c)$ for the three theories ($M = 1$ for the ψ theory).

	Mean field	ψ theory	HD theory
κ_c	$1/\sqrt{6}$	0.433	0.397
$f(\kappa_c)$	2/3	0.707	0.790
$Q_c/t^{2\nu}$ (W/cm^2)	6082	6842	7007

In conclusion, our analysis has lead to a number of surprising results. There exists near T_λ, in the T-Q plane, a curve $T_c(Q)$ at which superflow ceases and the heat capacity, C_Q, diverges according to Eq. (23). Unlike other familiar phase transitions, the heat capacity divergence in this case is predicted by mean-field theory, and, indeed, the arguments leading to Eq. (22) show that $u = 1/2$ in any theory in which P is an analytic function of W [19]. Experimental measurements of C_Q near $T_c(Q)$ are urgently needed. As our arguments have shown, they would constitute the first information concerning how ρ_s depends on W near T_λ. Existing experiments [1] show that dissipation due to vortex formation [20] tends to set in at $Q/Q_c^{HD} \sim 0.4$, except perhaps very close to T_λ, where Q_c is very small. However, according to the data displayed in Fig. 2(b) a large effect ($\Delta C_Q \sim 1.5$ J/mole K) may be expected even at $Q/Q_c^{HD} \sim 0.4$.

The phenomenon that occurs at $T_c(Q)$ has been compared to a spinodal [4,5]. We would like to point out that it also bears some resemblance to a phase transition, even though there does not exist a normal phase of finite Q on the other side of the transition. For one reason, all other heat capacity divergences we know of do signal phase transitions. Second, when a system is characterized by a pair of conjugate variables (pressure-volume, concentration-chemical potential, magnetization-magnetic field), a phase transition occurs when the generalized susceptibility diverges (gas-liquid critical point, binary mixture phase separation, Curie point). In the present case, P and W are a new conjugate pair characterizing superflow whose susceptibility, $(\partial W/\partial P)_T$, diverges at $T_c(Q)$. This is not the ordinary superfluid transition, since ρ_s is not zero. By analogy to the other cases, W (not ρ_s) may be the order parameter and P the conjugate field. Seen in this light, the lambda transition at $Q = 0$ is rather like a tricritical point. If the transition is approached along this unique thermodynamic path, the coefficient of the θ^{-u} term vanishes, leaving only the familiar, near logarithmic divergence in the heat capacity, due to the disappearance of ρ_s [21].

Since the mean square fluctuations in W, $\langle \Delta W^2 \rangle$ [22], diverge at $T_c(Q)$, the real issue becomes not whether we call this strange new phenomenon a spinodal or a phase transition, but rather whether the velocity fluctuations renormalize $\rho_s(W)$ and thereby change the critical point exponent from its mean-field value of 0.5, and whether the phenomenon belongs to a different universality class from the lambda transition. The answers to these questions are not yet known.

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[1] P. Leiderer and F. Pobell, Z. Phys. **223**, 378 (1969); S. M. Bhagat and R. A. Lasken, Phys. Rev. A **3**, 340 (1971); R. V. Duncan, G. Ahlers, and V. Steinberg, Phys. Rev. Lett. **60**, 1522 (1988); F.-C. Liu and G. Ahlers, Phys. Rev. Lett. **76**, 1300 (1996).

[2] H. J. Mikeska, Phys. Rev. **179**, 166 (1969); A. Onuki, J. Low Temp. Phys. **55**, 309 (1984).

[3] D. R. Swanson, T. C. P. Chui, and J. A. Lipa, Phys. Rev. B **46**, 9043 (1992); L. S. Goldner, N. Mulders, and G. Ahlers, J. Low Temp. Phys. **93**, 131 (1992).

[4] R. Haussmann and V. Dohm, Phys. Rev. B **46**, 6361 (1992).

[5] R. Haussmann and V. Dohm, Phys. Rev. Lett. **72**, 3060 (1994).

[6] Private communication. Related comments: D. Goodstein, T. Chui, and A. Harter, Phys. Rev. Lett. **77**, 979 (1996); R. Haussmann and V. Dohm, Phys. Rev. Lett. **77**, 980 (1996).

[7] D. L. Goodstein, *States of Matter* (Dover, New York, 1985), p. 336.

[8] Mikeska (Ref. [2]) gave a similar description based on the mean-field theory.

[9] G. B. Hess, Phys. Rev. Lett. **40**, 119 (1978).

[10] V. L. Ginzburg and L. P. Pitaevskii, Zh. Eksp. Teor. Fiz. **34**, 1240 (1958) [Sov. Phys. JETP **7**, 858 (1958)].

[11] V. L. Ginzburg and A. A. Sobyanin, J. Low Temp. Phys. **49**, 507 (1982).

[12] L. P. Pitaevskii, Zh. Eksp. Teor. Fiz. **35**, 408 (1959) [Sov. Phys. JETP **8**, 282 (1959)].

[13] I. M. Khalatnikov, Zh. Eksp. Teor. Fiz. **57**, 489 (1969) [Sov. Phys. JETP **30**, 268 (1970)].

[14] W. Y. Tam and G. Ahlers, Phys. Rev. B **32**, 5932 (1985).

[15] J. R. Clow and J. D. Reppy, Phys. Rev. A **5**, 424 (1972).

[16] H. A. Kierstead, Phys. Rev. **162**, 153 (1967).

[17] We used the value given by D. S. Greywall and G. Ahlers, Phys. Rev. A **7**, 2145 (1973). The value of ρ_0 needed to make our result consistent with HD is 0.406 g/cm^3.

[18] A. Singsaas and G. Ahlers, Phys. Rev. B **29**, 4951 (1984).

[19] In HD theory, P is analytic at all values of κ up to $\kappa_c = 0.397$. It is nonanalytic at $\kappa = 1/\sqrt{6} = 0.408$. If this nonanalyticity occurred at κ_c, the exponent u would be changed to $1/3$.

[20] T. C. P. Chui, U. E. Israelsson, and M. J. Lysek, Physica (Amsterdam) **194-196B**, 601 (1994).

[21] For similar behavior near the tricritical point, see G. Ahlers, *The Physics of Liquid and Solid Helium Part I*, edited by K. H. Bennemann and J. B. Ketterson (Wiley, New York, 1976), p. 143.

[22] It can be shown that $\langle \Delta W^2 \rangle = k_B T (\partial W/\partial P)_T$, where k_B is the Boltzmann constant.