Joule heating in Boltzmann theory of metals

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Boltzmann theory deals with the quasiparticle distributions F_K for electrons and N_Q for phonons. It is difficult to reconcile the common situation of "steady state" (where distributions are out of equilibrium but carry steady charge current $\vec{j}$) and Joule heating $\vec{j}\cdot\vec{E}$. This paper offers elementary clarification. The resolution is simplest if a true steady state is abandoned.

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I. INTRODUCTION

Joule heat is so well known that an explanation is needed for this paper. Boltzmann theory is well described in textbooks, the old master being Ziman [1]. The theory uses quasiparticles as defined by Landau. Electron quasiparticles have crystal quantum numbers $K = (\vec{k}, n, \sigma)$ and phonons have quantum numbers $Q = (\vec{q}, j)$. These quasiparticle states are not eigenstates of electron and phonon Hamiltonians. Instead, they have thermal shifts and lifetime broadening, as described by Green's function theories. The wave vectors $\vec{k}$ and $\vec{q}$ are in one-to-one correspondence with those of the eigenstates of effective Hamiltonians, and are defined for finite systems by using periodic boundary conditions. Boltzmann theory gives a very successful theory of how the distributions F_K and N_Q deviate from equilibrium Fermi-Dirac f_K and Bose-Einstein n_Q, and how they evolve in time under the influence of applied external fields. In recent years, computational theory has made important advances in using Boltzmann theory to predict linear transport properties of crystalline matter (see for example, Ref. [2] for phonons and Ref. [3] for electrons) and, less often, nonlinear effects [4].

In this paper the field will be $\vec{E}$. Most often, but not necessarily, it is constant in time and space. When $\vec{E}$ is constant, one expects a steady current $\vec{j}$. But then Joule heat $\vec{j}\cdot\vec{E}$ necessarily accompanies the current. In the rigorous quasiparticle picture, the system is closed, so its energy is increasing in time. This confusion has bothered many of us. It has been correctly addressed in previous literature [5,6]. The main result in this paper is to provide a simple clarifying update. The clarification is that this deviation from steady state does not occur to first order in $\vec{E}$; $\vec{j}\cdot\vec{E}$ is second and higher order. When one wants only first-order effects, Joule heating is omitted, but a nonequilibrium temperature shift ΔT can be included. The electron distribution function can be expanded in powers of E and ΔT: $F_K = \sum_K F_K^{(n)}$ where $F_K^{(0)} = f_K$ and $F_K^{(n)} = \sum_{m=0}^{n} a_{n,m} E^{n-m} \Delta T^m$. The first-order correction obeys the steady-state condition $\partial F_K^{(1)}/\partial t = 0$. To study higher-order effects, the system being closed, it is necessary to have $\partial F_K^{(n)}/\partial t \neq 0$ for $n > 1$. This will be explained in subsequent sections. This is not a new idea. It was mentioned in passing by Greenwood [7] in 1958. Here we provide clarification that Greenwood omitted.

Observable effects of Joule heat often happen in small systems which are coupled to their environment [8,9]. The coupling is a challenge for Boltzmann theory. We believe that there is no rigorous resolution, but satisfactory phenomenological coupling to environment is often available. An example is studies of nonlinear I-V behavior of carbon nanotubes [10]. Another situation where Joule heat needs to be carefully examined is current-driven metal-insulator transitions [11–16]. In some experiments, metal/insulator phase boundaries are observed to move in time, which requires keeping nonzero $\partial F_K^{(n)}/\partial t$ for $n > 1$ [11,13,16].

II. SIMPLEST VERSION

Consider a long metallic wire (length L). Voltage ΔV is applied through electrodes at the two ends. In the interior, uniform current $\vec{j} = \sigma\vec{E}$ flows in response to a uniform internal field $E \approx -\Delta V/L$. The time rate of work done per unit volume by the internal E field is the Joule heat $\vec{j}\cdot\vec{E} = \sigma E^2$. Suppose the wire is suspended in vacuum between the electrodes. Heat can escape by conduction to the electrodes and by radiation to vacuum. A deeper discussion is postponed until the last section. Except at high T, radiation is inefficient. If the wire has low enough thermal conductance that the conductive heat escape can be ignored, then the wire is not in steady state. Its temperature increases in time, $dT/dt = \vec{j}\cdot\vec{E}/C$ where C is the heat capacity. This is not normally included in microscopic linear theories of conductivity, where Joule heat is not included. So where in Boltmann theory does the Joule heat reside?

For a homogeneous long wire, the Boltzmann equation is

$$\frac{\partial F_K}{\partial t} = -\dot{\vec{k}}\cdot\frac{\partial F_K}{\partial \vec{k}} + \left(\frac{dF_K}{dt}\right)_{\rm coll}. \tag{1}$$

The electron distribution function F_K deviates from the Fermi-Dirac distribution f_K. The acceleration term uses the

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semiclassical equation $\hbar\dot{\vec{k}} = -e\vec{E}$ where $E = E_{\rm ext}$ is the uniform applied field. The rigorous collision term is a complicated nonlinear function of $F_{K'}$ and N_Q. It is normal, and usually satisfactory, to assume that the phonons are kept in equilibrium by anharmonic scattering, so that the collision term for electrons depends on n_Q but not on $N_Q - n_Q$. This decouples the electron Boltzmann equation (1) from the phonon Boltzmann equation. The next section will discuss the correct coupled equations.

The electron quasiparticle energy is $\Omega u = \sum_K \epsilon_K F_K$, where Ω is the sample volume. The time rate of change of energy density is therefore

$$\frac{\partial u}{\partial t} = \frac{1}{\Omega}\sum_K \epsilon_K \frac{\partial F_K}{\partial t} = \frac{1}{\Omega}\sum_K \epsilon_K \frac{\partial F_K}{\partial \vec{k}}\frac{eE}{\hbar} + \frac{1}{\Omega}\sum_K \epsilon_K \left(\frac{dF_K}{dt}\right)_{\rm coll}. \quad (2)$$

The collision term on the right sums rigorously to zero by energy conservation. The acceleration term can be simplified by integrating by parts,

$$\frac{\partial u}{\partial t} = -\frac{e}{\Omega}\sum_K \vec{v}_K F_K \cdot \vec{E} = \vec{j}\cdot\vec{E}. \quad (3)$$

This uses the quasiparticle (or Boltzmann) definition of current, $\vec{j} = -(e/\Omega)\sum_K \vec{v}_K F_K$. Joule heating is therefore rigorously contained in Boltzmann theory. Importantly, no linear approximation is made; the current $\vec{j}$ is not constrained to the linear form $\sigma\vec{E}$.

III. WITH PHONONS AND NONZERO $\vec{\nabla}T$

Here is an essentially complete version of Boltzmann theory,

$$\frac{\partial F_K}{\partial t} = -\dot{\vec{k}}\cdot\frac{\partial F_K}{\partial \vec{k}} - \dot{\vec{r}}_{\rm el}\cdot\frac{\partial F_K}{\partial \vec{r}} + C_K(\{F,N\}),$$
$$\frac{\partial N_Q}{\partial t} = -\dot{\vec{r}}_{\rm ph}\cdot\frac{\partial N_Q}{\partial \vec{r}} + C_Q(\{F,N\}). \quad (4)$$

There is no acceleration term for phonons. Both distributions can be spatially inhomogeneous, in which case their evolution contains drift at velocity $\dot{\vec{r}}_{\rm el} = \vec{v}_K$ or $\dot{\vec{r}}_{\rm ph} = \vec{v}_Q$, where $\vec{v}_K = \partial\epsilon_K/\partial\vec{k}$ is the electron group velocity, and $\vec{v}_Q = \partial\omega_Q/\partial\vec{q}$ is the phonon group velocity. Factors of $\hbar$ are omitted when confusion is unlikely. The collision terms $(dF_K/dt)_{\rm coll} = C_K(\{F,N\})$ and $(dN_Q/dt)_{\rm coll} = C_Q(\{F,N\})$ contain electron-phonon interactions, which is why they depend on both F and N. They also include electron-electron (Coulomb) and phonon-phonon (anharmonic) interactions, and defect scattering. Boundary scattering can be included in a semiphenomenological way. They conserve charge density n and energy density u,

$$\left(\frac{dn}{dt}\right)_{\rm coll} = -\frac{e}{\Omega}\sum_K C_K(\{F,N\}) = 0, \quad (5)$$

$$\left(\frac{du}{dt}\right)_{\rm coll} = \frac{1}{\Omega}\sum_K \epsilon_K C_K(\{F,N\}) + \frac{1}{\Omega}\sum_Q \omega_Q C_Q(\{F,N\}) = 0. \quad (6)$$

Charge density conservation for the short version (1) of the Boltzmann equation is just $\partial n/\partial t = 0$, because the acceleration part $(1/\Omega)\sum_K \dot{\vec{k}}\partial F_K/\partial\vec{k}$ integrates to zero. When drift is added, the answer is

$$\frac{\partial n}{\partial t} = -\vec{\nabla}\cdot\vec{j}, \quad (7)$$

because the spatial gradient $\partial/\partial\vec{r}$ in Eq. (4) can be moved outside the K integration. Similarly, energy conservation becomes

$$\frac{du}{dt} = \vec{j}\cdot\vec{E} - \vec{\nabla}\cdot\vec{j}_u, \quad (8)$$

where the energy current density is

$$\vec{j}_u = \frac{1}{\Omega}\sum_K \epsilon_K \vec{v}_K F_K + \frac{1}{\Omega}\sum_Q \omega_Q \vec{v}_Q N_Q. \quad (9)$$

These conservation laws are exact in classical physics, and statistically exact in quantum mechanics. The Boltzmann description is not exact; it uses the quasiparticle approximation for n, u, $\vec{j}$, and $\vec{j}_u$. Nevertheless, it does obey exact conservation laws.

IV. FIRST ORDER IN $\vec{E}$ AND $\vec{\nabla}T$

To study charge current, we focus on electrons, and assume that the scattering term in Eq. (4) for electrons, $C_K(\{F,N\})$, can be approximated by $C_K(\{F,n\})$ because of anharmonic scattering. The distribution function F_K relaxes toward the equilibrium Fermi-Dirac distribution f_K. The deviation from equilibrium is to be computed to first order in E. The linearized version of Eq. (4) is

$$0 = e\vec{E}\cdot\vec{v}_K\frac{\partial f_K}{\partial\epsilon_K} - \vec{v}_K\cdot\frac{\partial f_K}{\partial T}\vec{\nabla}T - \sum_{K'} C^{(1)}_{KK'}F^{(1)}_{K'}, \quad (10)$$

where $-C^{(1)}_{KK'}F^{(1)}_{K'}$ is the linearized version of the scattering operator $C_K(\{F,n\})$. To first order in $a\vec{E} + b\vec{\nabla}T$, the system has a time-independent steady state $\partial F^{(1)}_K/\partial t = 0$. The higher-order pieces of the acceleration and drift terms proportional to $\partial F^{(n)}_K/\partial\vec{k}$ and $\partial F^{(n)}_K/\partial\vec{r}$ are dropped because, containing $n-1$ factors of $a\vec{E} + b\vec{\nabla}T$, they give contributions higher than first order. The linearized collision operator $\hat{C}^{(1)}$ scatters electrons from K to K', conserving particle density and energy density:

$$\sum_{KK'} C^{(1)}_{KK'}F^{(1)}_{K'} = 0 \quad \text{and} \quad \sum_{KK'} \epsilon_K C^{(1)}_{KK'}F^{(1)}_{K'} = 0. \quad (11)$$

These hold for any deviation $F^{(1)}_K$, which requires $\sum_K C^{(1)}_{KK'} = 0$ and $\sum_K \epsilon_K C^{(1)}_{KK'} = 0$. In other words, 1 and ϵ_K are null left eigenvectors of $\hat{C}^{(1)}$. The collision operator is real but not symmetric. All other eigenvalues are greater than 0, except these deriving from conservation laws. In the space orthogonal to 1 and ϵ_K, the matrix $C^{(1)}_{KK'}$ has a well-defined inverse. The solution is

$$F^{(1)}_K = [C^{(1)}]^{-1}_{KK'}\left[e\vec{E}\cdot\vec{v}_{K'}\frac{\partial f_{K'}}{\partial\epsilon_{K'}} - \vec{v}_{K'}\frac{\partial f_{K'}}{\partial T}\vec{\nabla}T\right]. \quad (12)$$

The electrical current, to first order in $\vec{E}$, is then

$$\vec{j} = \sigma\vec{E} + \sigma S\vec{\nabla}T. \quad (13)$$

The conductivity $\sigma = (\partial \vec{j}/\partial \vec{E})_{E=0}$ is

$$\sigma = \frac{e^2}{\Omega} \sum_{KK'} \vec{v}_K [C^{(1)}]^{-1}_{KK'} \vec{v}_{K'} \left(-\frac{\partial f_{K'}}{\partial \epsilon_{K'}} \right). \tag{14}$$

Under open circuit conditions, no current flows, and $\vec{E} = -S\vec{\nabla}T$, where S is the Seebeck coefficient,

$$S = \frac{e}{\sigma\Omega} \sum_{KK'} \vec{v}_K [C^{(1)}]^{-1}_{KK'} \vec{v}_{K'} \frac{\epsilon_{K'} - \mu}{T} \left(-\frac{\partial f_{K'}}{\partial \epsilon_{K'}} \right). \tag{15}$$

V. SECOND ORDER IN $\vec{E}$ AND $\vec{\nabla}T$

Here is the equation for $F^{(2)}$:

$$\frac{\partial F^{(2)}_K}{\partial t} = \frac{e\vec{E}}{\hbar} \cdot \frac{\partial F^{(1)}_K}{\partial \vec{k}} - \vec{v}_K \cdot \frac{\partial F^{(1)}_K}{\partial T} \vec{\nabla} T - \sum_{K'} C^{(1)}_{KK'} F^{(2)}_{K'} - \sum_{K',K''} C^{(2)}_{KK'K''} F^{(1)}_{K'} F^{(1)}_{K''}, \tag{16}$$

where $\hat{C}^{(2)}$ is the part of the collision term that is quadratic in $F^{(1)}_K = F_K - f_K$. A referee pointed out correctly that Joule heat causes $\Delta T \propto E^2$ which causes a shift $\propto \partial n_Q/\partial T \times E^2$ in the equilibrium Bose distribution. This introduces a corrected electron-phonon scattering term $-\sum_{K'} \Delta C^{(1)}_{\mathrm{ep},KK'} F^{(1)}_{K'}$ in Eq. (16). In the next section, estimates of ΔT are given. It is unlikely that this additional term plays a significant role in metals.

Equation (16) is a linear equation for the unknown function $F^{(2)}_K$. It is now necessary to invert the operator $\hat{1}\partial/\partial t + \hat{C}^{(1)}$, which is trickier than just inverting $\hat{C}^{(1)}$, as was done for $F^{(1)}$. However,we can calculate the Joule heat without solving Eq. (16), using energy conservation,

$$\frac{du^{(2)}}{dt} = \frac{1}{\Omega} \sum_K \epsilon_K \frac{\partial F^{(2)}_K}{\partial t}. \tag{17}$$

Now insert the right-hand side of Eq. (16) in place of $\partial F^{(2)}_K/\partial t$. Only the first term contributes. The two collision terms vanish by energy conservation, and the drift term vanishes by time-reversal invariance. The $\vec{k}$ derivative of $F^{(2)}_K$ can be avoided by integration by parts, which gives

$$\frac{du^{(2)}}{dt} = -\frac{e\vec{E}}{\Omega} \cdot \sum_K \vec{v}_K F^{(1)}_K = \vec{E} \cdot \sigma \cdot \vec{E} = \vec{E} \cdot \vec{j}^{(1)}. \tag{18}$$

This is the leading term in the Joule heat formula. This made use of Eqs. (12) and (14). Note that it is not a completely trivial extension of the usual first-order Boltzmann result. It involves the $\vec{k}$ derivative of the nonequilibrium distribution $F^{(1)}_K$. Third- and higher-order generalizations of Eq. (16) yield by the same arguments the higher-order terms in the Joule heat.

VI. HEAT FLOW TO THE ENVIRONMENT

If there is no heat dissipation, temperature will increase at a rate $\vec{j} \cdot \vec{E}/C$. For a copper wire of length 1 cm and applied voltage $\Delta V = 1$ mV, using the heat capacity C at 300 K (fairly constant up to melting), the rate of increase is 0.017 K/s.

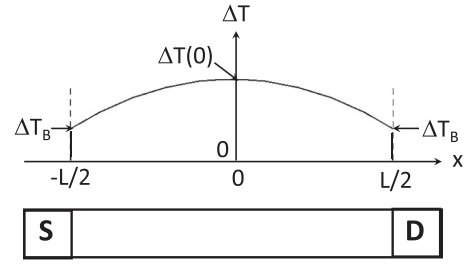

FIG. 1. Schematic of a suspended metal wire of length L, with source and drain electrodes held at temperature T_0. The temperature shift $\Delta T(x) = T(x) - T_0$ is plotted versus position. Diffusive conduction of Joule heat out of the wire causes the parabolic curve. Thermal contact resistance causes the shifts ΔT_B at the boundaries between sample and source and drain.

How does Joule heat dissipate, and what is the steady-state temperature increase ΔT? Consider the model of an ideal metal wire, of length L and cross-sectional area A, suspended between two electrodes, as in Fig. 1. The electrodes are assumed large and serve as heat baths at the environmental temperature T_0. Given volumetric heating $\dot{Q}_E = \vec{j} \cdot \vec{E}$, what is the steady-state temperature difference $\Delta T(x) = T(x) - T_0$ between the wire and the environment (vacuum and electrodes)?

There are two mechanisms, conduction to electrodes, and radiation, that compensate for $\dot{Q}_E$ to allow a steady temperature. Incorporating these in rigorous Boltzmann theory is difficult. The former requires a detailed knowledge of heat-carrying quasiparticles of both sample and electrodes and their coupling at the contacts. The latter requires a similar understanding of the photons in vacuum, the electrons and phonons in the finite-size sample, and their couplings at the surface. This section abandons Boltzmann theory and uses simpler and more approximate formulas.

Conductive heat loss has been studied experimentally [17,18] in nanotubes and nanowires, using scanning thermal microscopy and spatially resolved Raman spectroscopy. Ignoring radiation and assuming diffusive heat transport, the local heat current is $j_{\mathrm{th}}(x) = -\kappa dT(x)/dx$, for $-L/2 < x < L/2$. This current is assumed uniform throughout the cylindrical cross section of area $A = \pi D^2/4$. It ($|j_{\mathrm{th}}|$) is zero in the middle of the wire ($x = 0$ as in Fig. 1), and increases as position $|x|$ increases, as required by energy conservation $dj_{\mathrm{th}}(x)/dx = \dot{Q}_E$. Then, in steady state, the total rate of Joule heat generation $AL\dot{Q}$ balances the heat current $A[|j_{\mathrm{th}}(-\frac{1}{2}L)| + |j_{\mathrm{th}}(\frac{1}{2}L)|]$ leaving the ends of the sample. The result is simpler than that of Shi *et al.* [17] because here the metal sample is assumed to be suspended. The temperature varies quadratically,

$$T(x) = T_0 + \Delta T_C + \frac{\dot{Q}_E}{2\kappa} \left[\left(\frac{1}{2}L \right)^2 - x^2 \right]. \tag{19}$$

Here ΔT_C is the temperature jump caused by the thermal contact resistance, $\Delta T_C = |j_{\mathrm{th}}(\pm\frac{1}{2}L)|/\kappa_C = \dot{Q}L/2\kappa_C$, where κ_C is the thermal contact conductance between sample and source or drain. Then the average temperature $\bar{T}$ of the wire

sample is

$$\overline{\Delta T} = \bar{T} - T_0 = \dot{Q}\left[\frac{L}{2\kappa_C} + \frac{L^2}{12\kappa}\right]. \tag{20}$$

As an example, consider a copper wire at room temperature. The electrical resistivity is $\rho = 1.7\ \mu\Omega\,\mathrm{cm}$ and the thermal conductivity is $\kappa = 385\ \mathrm{W/m\,K}$. These agree to 10% with the Wiedemann-Franz "law" (actually an approximate formula) $\rho\kappa = L_0 T$, where the Lorenz number is $L_0 = 2.44 \times 10^{-8}\ \mathrm{W\,\Omega/K^2}$. For an intimate, clean contact to aluminum (chosen because a reliable thermal contact study [19] is available), the thermal contact conductivity is $\kappa_C = 2.5 \times 10^9\ \mathrm{W/m^2\,K}$ [19]. The bulk and contact contributions to $\overline{\Delta T}$ are equal when $L = 6\kappa/\kappa_C = 0.92\ \mu\mathrm{m}$. Therefore, let us ignore the thermal contact resistance. Then, surprisingly, the temperature increase [using $\dot{Q} = \sigma E^2 = (\Delta V/L)^2/\rho$] is independent of the length and cross-sectional area,

$$(\Delta T)_{\mathrm{cond}} = \frac{(\Delta V)^2}{12\rho\kappa} \approx \frac{(\Delta V)^2}{12 L_0 T}. \tag{21}$$

For copper at $T = 300$ K the temperature rise is $\Delta T = 0.013$ K for a source-drain voltage $\Delta V = 1$ mV.

For a cylindrical wire of length L and diameter D, the Stefan-Boltzmann law for radiative heat loss is

$$\dot{Q}_{\mathrm{rad}}(x) = \frac{4\bar{\epsilon}}{D}\sigma_{\text{S-B}}\left[(T_0 + \Delta T_{\mathrm{rad}})^4 - T_0^4\right] = \frac{(\Delta V)^2}{\rho L^2}. \tag{22}$$

Here the factor $4/D$ is the ratio of surface area to volume of the wire, $\sigma_{\text{S-B}} = 5.67 \times 10^{-8}\ \mathrm{W/m^2\,K^4}$ is the Stefan-Boltzmann constant, and $\bar{\epsilon}$ is an effective emissivity. The emissivity of IR radiation by metals is small, anisotropic, frequency dependent, and temperature dependent. The small factor $\omega/\omega_p \sim 0.008$ enters, evaluated for ω at the peak of the Planck spectrum at room temperature, using the plasma frequency ω_p for copper. The effective emissivity is complicated. A sensible guess is $\bar{\epsilon} \sim 0.01$ [20]. The ratio of temperature increases ΔT caused by radiation and conduction is then

$$\frac{(\Delta T)_{\mathrm{rad}}}{(\Delta T)_{\mathrm{cond}}} = \left[\frac{3\kappa}{4\bar{\epsilon}\sigma_{\mathrm{SB}}}\right]\frac{D}{L^2 T_0^3} \sim (1.9 \times 10^4\ \mathrm{m})\frac{D}{L^2}\left(\frac{300\ \mathrm{K}}{T_0}\right)^3, \tag{23}$$

where it is assumed that $(\Delta T)_{\mathrm{rad}} \ll T_0$.

Consider a Cu wire of length $L = 1$ cm and diameter $D = 1\ \mu\mathrm{m}$ ($D/L^2 = 1\ \mathrm{m^{-1}}$), with 1 mV applied voltage ΔV and $T = 300$ K. Its resistance is 2.2 Ω, and it carries 0.46 mA of current. The temperature rise, when only radiative heat loss is considered, is $\Delta T \sim 246$ K, not small compared with $T_0 = 300$ K. It is $\Delta T = 130$ K if the Taylor expansion of $(T_0 + \Delta T)^4$ is not used. This is greater by 10^4 than the temperature increase allowed by conductive cooling. Conduction is more efficient for dissipating Joule heat in Cu wires at 300 K when $L^2/D \leqslant 2 \times 10^4$ m. Radiative efficiency improves rapidly with T, and conductive efficiency worsens as $1/T$. For Cu with $L^2/D \sim 1$ m, to make radiation as effective as conduction requires $T_0 \sim 3000$ K (∼8000 K, except that emissivity $\bar{\epsilon}$ increases as the peak of the Planck spectrum shifts upward with rising T_0.)

VII. SUMMARY

Joule heating arises naturally when a nonvanishing $\partial F/\partial t$ term is included in the Boltzmann equation. To include Joule heating requires solving the Boltzmann equation to at least second order in $\vec{E}$. For idealized systems best described by Boltzmann theory, if electrical current is not too high, the system is in a "quasi-steady state," but not a true steady state. Energy density slowly increases at the rate $\vec{j}\cdot\vec{E}$. This is true to all orders in $\vec{E}$ even though $\vec{j}$ will have parts nonlinear in $\vec{E}$. If analysis in a true steady state is desired, then there should be an external heat bath coupled to the system. This is not easy in Boltzmann theory, without simplified approximations.

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