Diffusion of aerosol particles in a stochastic nonisothermal medium

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The diffusion of highly dispersed aerosol particles in a turbulent medium with low-intensity velocity fluctuations is considered in the presence of a temperature gradient. The velocity-field fluctuations are assumed Gaussian with a uniform pair correlation function. An expression that takes into account thermophoresis and turbulent migration of the particles is obtained for the effective diffusion tensor. It is shown that the effective thermophoresis velocity is always lower than the regular one.

1. INTRODUCTION

Interest in the transport of highly dispersed aerosol particles (AP) in a medium with a random velocity field has increased greatly in recent years. In a number of applied problems, such as controlling combustion in a gas mixture by activators, or trapping AP in variable-temperature tubes and channels, account must be taken of the temperature inhomogeneity of the medium.[1,2]

It is known[3,4] that in the presence of temperature gradient the AP are acted upon by a thermophoretic force due to the uncompensated momentum imparted to the particles by the molecules of the nonuniformly heated gas. Numerous theoretical calculations and experimental investigations have shown that AP drift induced by the thermophoretic force (thermophoresis) can exceed substantially the Brownian diffusion and be the decisive transport mechanism for particles measuring 0.1–1 μm (Ref. 5). Most studies, however, either dealt with AP transport in laminar streams or disregarded the correction for the interference between the thermophoresis and the turbulent migration of the particles.

It was pointed out in Ref. 2 that such an approach is incorrect, and an equation that takes the non-additivity of these phenomena into account was proposed. Nonetheless, the proposed heuristic equation does not reveal the explicit dependence of the effective thermophoresis velocity and of the turbulent-diffusion coefficient on the statistical properties of the medium and on the temperature gradient. We therefore derive here rigorously, for a model with passive impurity, a transport equation for highly dispersed aerosol particles in a turbulent non-isothermal region with small random velocity pulsations at low relative temperature drops. The equation derived accounts explicitly for the interaction of the temperature and particle-density fields.

2. FORMULATION OF THE PROBLEM

Highly dispersed AP are characterized by low values of the inertia index,[2] so that their diffusion can be regarded, just as the heat propagation, in the passive-impurity-model approximation.[6]

Thus, random realization of ensembles of the particle-density and temperature fields, $n(\mathbf{r},t)$ and $T(\mathbf{r},t)$, in an incompressible medium with a specified random velocity field $\mathbf{v}(\mathbf{r},t)$ can be described by the following set of equations:

$$\frac{\partial n}{\partial t}-D_0\nabla^2 n=-\nabla(\mathbf{u}n)-\mathbf{v}\nabla n; \tag{1}$$

$$\frac{\partial T}{\partial t}-\chi\nabla^2 T=-\mathbf{v}\nabla T, \tag{2}$$

where $\mathbf{u}$ is the thermophoresis velocity. D_0 and χ are the coefficients of the Brownian diffusion of the AP and of the thermal diffusivity of the medium, respectively.

The characteristic time of hydrodynamic relaxation of highly dispersed AP can usually be regarded as small, and the thermophoresis velocity can be calculated in a quasi-stationary approximation. We assume in addition that the relative temperature drop over the characteristic scale of the considered medium permits linearization of the expression for the thermophoresis velocity[1)]

$$\mathbf{u}=-f_T\frac{\nu_e}{T_*}\nabla T. \tag{3}$$

Here T_* is the average temperature of the considered region; f_T is the thermophoretic coefficient and depends on the thermophysical properties of the gas and particles, as well as on the particle radius[7]; ν_e is the kinematic viscosity coefficient of the medium.

We represent the hydrodynamic velocity by a sum of a regular and a fluctuating component:

$$\mathbf{v}(\mathbf{r},t)=\langle\mathbf{v}(\mathbf{r},t)\rangle+\delta\mathbf{v}(\mathbf{r},t).$$

The angle brackets $\langle\ldots\rangle$ denote averaging over an ensemble of realizations of the stochastic field $\mathbf{v}(\mathbf{r},t)$. Assuming $\langle\mathbf{v}(\mathbf{r},t)\rangle=\text{const}$, we shall consider Eqs. (1) and (2) in a comoving coordinate frame. With allowance for (3), the system (1)–(2) takes then the form

$$\frac{\partial n}{\partial t}-D_0\nabla^2 n=f_T\frac{\nu_e}{T_*}\nabla(n\nabla T)-\delta\mathbf{v}\nabla n; \tag{4}$$

$$\frac{\partial T}{\partial t}-\chi\nabla^2 T=-\delta\mathbf{v}\nabla T. \tag{5}$$

The system (4)–(5) describes the interaction of the stochastic fields of the AP density and of the temperature of the medium, which result from the thermophoresis.

3. DETERMINATION OF THE AVERAGE TOTAL GREEN FUNCTION OF THE AP DENSITY

Formal solution of (5) leads to an integral equation for $T(\mathbf{r},t)$:

$$T(\mathbf{r},t)=T_0(\mathbf{r},t)+\int d\mathbf{r}'\int dt' G_T(\mathbf{r}-\mathbf{r}';t-t')\times(-\delta\mathbf{v}(\mathbf{r}',t')\nabla')T(\mathbf{r}',t'), \tag{6}$$

where $T_0(\mathbf{r},t)$ is the solution of the homogeneous equation and G_T is the retarded Green function for an infinite medium:

$$G_T(\mathbf{r}-\mathbf{r}';\ t-t')$$

$$=\theta(t-t')\,[4\pi\chi(t-t')]^{-3/2}\exp[-(\mathbf{r}-\mathbf{r}')^2/4\chi(t-t')].$$

Substitution of (6) in (4) gives

$$\left\{\frac{\partial}{\partial t_1}-D_0\nabla_1{}^2-f_T\frac{\nu_e}{T_*}[(\nabla_1T_0(1)\nabla_1+\nabla_1{}^2T_0(1))]\right\}n(1)$$

$$=f_T\frac{\nu_e}{T_*}\nabla_1\left[n(1)\nabla_1\int d2G_T(1;2)(-\delta\mathbf{v}(2)\nabla_2)T(2)\right]$$

$$+(-\delta\mathbf{v}(1)\nabla_1)n(1), \qquad (7)$$

where we use for brevity the notation $(\mathbf{r}_1,t_1)=(1)$; $(\mathbf{r}_1-\mathbf{r}_2;\ t_1-t_2)=(1;2)$.

We define the bare Green function $G_n^{(0)}(1;2)$ by the equation

$$\left\{\frac{\partial}{\partial t_1}-D_0\nabla_1{}^2-f_T\frac{\nu_e}{T_*}\right.$$

$$\left.\times[(\nabla_1T_0(1))\nabla_1+\nabla_1{}^2T_0(1)]\right\}G_n^{(0)}(1;2)=\delta(1;2). \qquad (8)$$

We obtain then for the total Green function of Eq. (7)

$$G_n(1;2)=G_n^{(0)}(1;2)+\int d3G_n^{(0)}(1;3)\Big\{(-\delta\mathbf{v}(3)\nabla_3)G_n(3;2)$$

$$+f_T\frac{\nu_e}{T_*}\nabla_3\Big[G_n(3;2)\int d4\nabla_3G_T(3;4)(-\delta\mathbf{v}(4)\nabla_4T(4))\Big]\Big\}. \qquad (9)$$

Introducing the notation

$$\mathbf{u}_0(1)=-f_T\frac{\nu_e}{T_*}\nabla_1T_0(1),\qquad \mathbf{u}(1)=-f_T\frac{\nu_e}{T_*}\nabla_1T(1),$$

$$\mathbf{G}_u(1;2)=\nabla_1G_T(1;2)$$

and differentiating Eq. (6) we arrive at the following system of Dyson equations for the total Green function of the AP density:

$$G_n(1;2)=G_n^{(0)}(1;2)+\int d3G_n^{(0)}(1;3)[-\delta\mathbf{v}(3)\nabla_3G_n(3;2)]$$

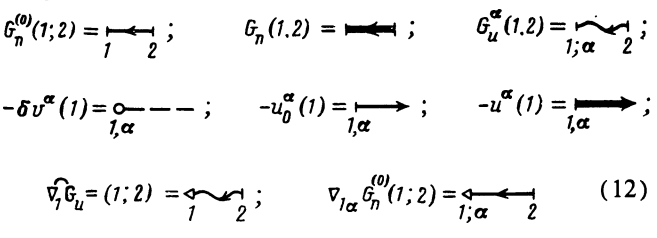
$$+\int d3d4G_n^{(0)}(1;3)\widehat{\nabla_3[G_n(3;2)}\mathbf{G}_u(3;4)(\delta\mathbf{v}(4)\mathbf{u}(4))]; \qquad (10)$$

$$\mathbf{u}(1)=\mathbf{u}_0(1)+\int d2G_u(1;2)(-\delta\mathbf{v}(2)\mathbf{u}(2)). \qquad (11)$$

We have used in (10) the following notation for the convolution:

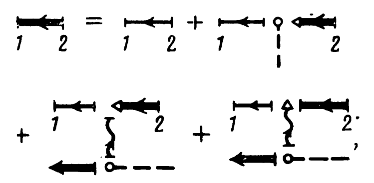
$$\widehat{\nabla_1\mathbf{G}}_u(1;\ 2)=\nabla_{1\alpha}G_u{}^{\alpha}(1;\ 2).$$

For future convenience, it is expedient to transform to diagrammatic notation:

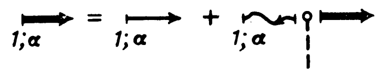


(12)

The system (10) and (11) takes then the form



(13)



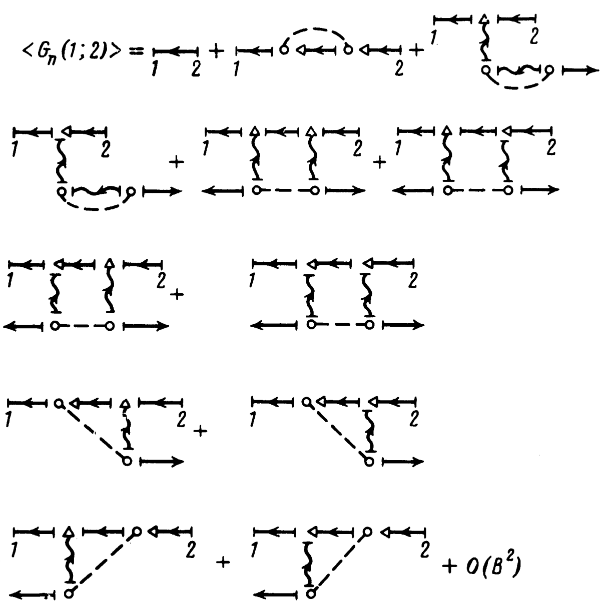
(14)

As usual, in the internal points we integrate over the spatiotemporal arguments and sum over the vector indices.

Assume that the ensemble of the stochastic velocity field is Gaussian with zero mean value and with a homogeneous pair correlation function

$$B_{\alpha\beta}(1;\ 2)=\langle\delta v_\alpha(\mathbf{r}_1,\ t_1)\delta v_\beta(\mathbf{r}_2,\ t_2)\rangle=B_{\alpha\beta}(1-2).$$

Carrying out the iterations and averaging Eq. (13) we obtain, accurate to terms $O(B^2)$, an equation for the average Green function of the AP density:



(15)

The averaging in (15) corresponds to closing of two open dashed lines.

The diagrammatic form identifies clearly the correlations that result from averaging the iteration solution of Eq. (10). For example, the second diagram of (15) corresponds to the contribution of turbulent particle migration without allowance for the fluctuations of the thermophoresis velocity, while the last four diagrams correspond to correlations between terms that describe turbulent migration and fluctuations of the thermophoresis velocity.

4. CALCULATION OF THE EFFECTIVE THERMOPHORESIS VELOCITY AND OF THE COEFFICIENT OF TURBULENT DIFFUSION

For specific calculations we must select the functions $T_0(\mathbf{r},t)$. The simplest and most important for local analysis is the case of a constant temperature gradient,

$$\nabla T_0(\mathbf{r}, t) = \boldsymbol{\tau} = \text{const.}$$

Equation (8) for the bare Green function $G_n^{(0)}$ takes in this case the form

$$\left(\frac{\partial}{\partial t_1} - D_0 \nabla_1{}^2 + \mathbf{u}_0 \nabla_1\right) G_n^{(0)}(1;2) = \delta(1;2), \tag{16}$$

$$\mathbf{u}_0 = -f_T \frac{\nu_e}{T_*} \boldsymbol{\tau}.$$

It will be convenient hereafter to use a Fourier representation in which

$$G_T(p) = \frac{1}{ip^0 + \chi \mathbf{p}^2}; \qquad G_n^{(0)}(p) = \frac{1}{i(p^0 + \mathbf{u}_0\mathbf{p}) + D_0 \mathbf{p}^2}. \tag{17}$$

We shall carry out the calculations for a pair correlation function of the form[8]

$$B_{\alpha\beta}(p) = 6\pi^2 B_0 l_0^{-4} \tau_0^{-1} \delta(|\mathbf{p}| - l_0)(\delta_{\alpha\beta}\mathbf{p}^2 - p_\alpha p_\beta)[(p^0)^2 + \tau_0^{-2}]^{-1}. \tag{18}$$

Here $l_0^{-1} = R_0$; $B_0 = 1/3 v_0^2$; $v_0 = (\langle \delta \mathbf{v}^2 \rangle)^{1/2}$ is the characteristic rate of the velocity pulsations of the medium; R_0 and τ_0 are the characteristic spatial and temporal scales of the velocity-field correlation. The quantity B_0 determines therefore the intensity of the velocity fluctuations.

We write down the averaged Dyson equation (13) in the Fourier representation:

$$\langle G_n(p) \rangle = G_n^{(0)}(p) + G_n^{(0)}(p) K(p) G_n^{(0)}(p). \tag{19}$$

This equation was derived by using the relation

$$\langle G_n(1;2) \rangle = \langle G_n(1-2) \rangle = \frac{1}{(2\pi)^4} \int d^4p\, e^{ip(1-2)} \langle G_n(p) \rangle.$$

Obviously, the spatial and temporal homogeneity of the average Green function is due to the corresponding homogeneity of the correlation function B.

The operator $K(p)$ is defined by the following series:

$$K(p) = \Sigma(p) + \Sigma(p) G_n^{(0)}(p) \Sigma(p) + \Sigma(p) G_n^{(0)}(p) \Sigma(p) G_n^{(0)}(p) \Sigma(p) + \ldots \tag{20}$$

The operators $\Sigma(p)$ in (20) are described by single-particle-irreducible diagrams, i.e., by diagrams that do not break up into the simplest one when one $G_n^{(0)}$ line is broken.

Summing formally the series (20) and substituting the resultant expression for $K(p)$ in Eq. (19) we obtain after simple transformations

$$\langle G_n(p) \rangle^{-1} = [G_n^{(0)}(p)]^{-1} - \Sigma(p). \tag{21}$$

Equation (21) is another form of the averaged Dyson equation (13). It will be used to interpret the results of the calculation of $\Sigma(p)$ as contributions to the effective diffusion coefficient and to the effective thermophoresis velocity.

Accurate to $O(B^2)$, the integrals that enter in $\Sigma(p)$ are contained in Eq. (15) from which the lines $G_n^{(0)}$ at the edges are removed. Note that in the case of a correlation function B that is homogeneous in the spatial variable, the third and fourth diagrams of (15) are zero.

Accurate to $O(B^2)$, if the functions $G_n^{(0)}(p)$, $G_T(p)$, and $B_{\alpha\beta}(p)$ are specified by Eqs. (17) and (18), all the integrals in $\Sigma(p)$ can be evaluated analytically. Let us calculate, for example, the ninth diagram $I_9(p)$

$I_9(p) =$ 

$$= \frac{3}{8} i \frac{B_0}{\pi^2 l_0^4 \tau_0} \int dq^0 \int d\mathbf{q}\, [(\mathbf{u}_0\mathbf{p})\mathbf{q}^2 - (\mathbf{u}_0\mathbf{q})(\mathbf{p}\mathbf{q})]\mathbf{q}^2$$
$$\times \delta(|\mathbf{q}| - l_0)\{[(q^0)^2 + \tau_0^{-2}](-iq^0 + \chi\mathbf{q}^2)[i(p^0 - q^0) + i\mathbf{u}_0(\mathbf{p} - \mathbf{q}) + D_0(\mathbf{p} - \mathbf{q})^2]\}^{-1}. \tag{22}$$

Integration yields

$$I_9(p) = |\mathbf{u}_0||\mathbf{p}| \frac{N}{M} \Big\{ \ln\left(\frac{\zeta+1}{\zeta-1}\right) \Big[(1-\zeta^2)(\mathbf{n}_a\mathbf{n}_b) + i\frac{|\mathbf{a}||\mathbf{b}|}{2M^2}(1-3\zeta^2)(1-(\mathbf{n}_a\mathbf{n}_b)^2) \Big]$$
$$+ \zeta[2(\mathbf{n}_a\mathbf{n}_b) + 3i|\mathbf{a}||\mathbf{b}|(1-(\mathbf{n}_a\mathbf{n}_b)^2)] \Big\}, \tag{23}$$

where

$$N = {}^3/_4 i B_0 l_0^2 (\tau_0^{-1} + \chi l_0^2)^{-1}, \quad M = [(\mathbf{a} + i\mathbf{b})^2]^{1/2},$$

$$\zeta = z_0/M, \ \mathbf{n}_a = \mathbf{a}/|\mathbf{a}|, \ \mathbf{n}_b = \mathbf{b}/|\mathbf{b}|, \ \mathbf{a} = 2 l_0 D_0 \mathbf{p},$$
$$\mathbf{b} = l_0 \mathbf{u}_0, \ z_0 = ip^0 + \tau_0^{-1} + i\mathbf{u}_0\mathbf{p} + D_0(\mathbf{p}^2 + l_0^2).$$

The expression for $I_9(p)$ turned out to be unwieldy and difficult to interpret. It can be simplified, however, by taking into consideration the actual values of the characteristic parameters of the problem.

A turbulent medium is determined primarily by the following parameters:

$$\eta = R_0 (4\chi\tau_0)^{-1/2}, \quad \xi = v_0 \tau_0 / 3R_0.$$

We shall assume that in our case $\eta \gg 1$ and $\xi \ll 1$. The first condition means that the characteristic scale of the spatial correlation is much larger than the length of temperature equalization by the molecular thermal conductivity mechanism during the correlation time τ_0. This assumption is valid for practically all real media. The second condition restricts the problem to low-intensity velocity fluctuations. Beside these parameters, a non-isothermal medium acquires a new parameter $\mu = R_0/|\mathbf{u}_0|\tau_0$, defined by the ratio of the correlation scale R_0 to the characteristic length of equalization of the particle density by thermophoresis within a time τ_0, with $\mu \gg 1$ almost always.

Lastly, it was assumed from the very beginning that the radius and correlation time of the turbulent fluctuations is much less than, respectively, the characteristic length and the characteristic time of variation of the average fields, i.e., we have for the pulses $p^0 \ll \tau_0^{-1}$ and $|\mathbf{p}| \ll l_0$.

Taking all the foregoing into account and recognizing the obvious relation $\chi \gg D_0$ we obtain from (23)

$$I_9(p) \approx i(\mathbf{u}_0\mathbf{p}) B_0 \tau_0^2 l_0^2. \tag{24}$$

Similar calculations yield the remaining diagrams contained in the operator $\Sigma(p)$ which takes after the indicated simplifications the form

$$\Sigma(p)\approx i(\mathbf{u}_0\mathbf{p})B_0\tau_0[\tau_0 l_0{}^2+\chi^{-1}\Phi_1(\omega)]$$
$$-\mathbf{p}^2B_0\tau_0[\Phi_2(\omega)+(\mathbf{n}_u\mathbf{n}_p)\Phi_3(\omega)]. \tag{25}$$

Here

$$\Phi_1(\omega)=1+{}^3/_2\omega^{-3}[\omega-\operatorname{arctg}(\omega)(1+\omega^2)],$$
$$\Phi_2(\omega)=-{}^5/_4+{}^3/_4\omega^{-3}[-3\omega+\operatorname{arctg}(\omega)(3+4\omega^2+\omega^4)],$$
$$\Phi_3(\omega)={}^{21}/_4+{}^3/_4\omega^{-3}[9\omega-\operatorname{arctg}(\omega)(9+10\omega^2+\omega^4)],$$
$$\mathbf{n}_u=\mathbf{u}_0/|\mathbf{u}_0|,\ \mathbf{n}_p=\mathbf{p}/|\mathbf{p}|.$$

This gives rise to one more important physical parameter

$$\omega=R_0|\mathbf{u}_0|/\chi, \tag{26}$$

which is the ratio of the thermophoresis velocity to the rate of equalization of the temperature by the molecular thermal conductivity over the correlation scale R_0.

Substituting (25) in (21) we get

$$\langle G_n(p)\rangle^{-1}=ip^0+i(\mathbf{u}_0\mathbf{p})+D_0\mathbf{p}^2-\Sigma(p)$$
$$\approx ip^0+i(\mathbf{u}_0\mathbf{p})\{1-B_0\tau_0[\tau_0l_0{}^2+\chi^{-1}\Phi_1(\omega)]\}$$
$$+\mathbf{p}^2\{D_0+B_0\tau_0[\Phi_2(\omega)+(\mathbf{n}_u\mathbf{n}_p)^2\Phi_3(\omega)]\}. \tag{27}$$

In the coordinate representation this relation corresponds to a differential operator

$$\frac{\partial}{\partial t}+\{1-B_0\tau_0[\tau_0l_0{}^2+\chi^{-1}\Phi_1(\omega)]\}u_0{}^\alpha\nabla_\alpha$$
$$-\nabla^\alpha\{[D_0+B_0\tau_0\Phi_2(\omega)]\delta_\alpha{}^\beta+n_{(u)\alpha}n^\beta_{(u)}[B_0\tau_0\Phi_3(\omega)]\}\nabla_\beta. \tag{28}$$

Thus, allowance for the turbulent pulsations causes the thermophoresis velocity and the particle diffusion coefficient to take on new effective values. Note that, in contrast to Ref. 8, the transition to asymptotes was made in the calculation of the effective diffusion coefficient in the last stage, in the exact expressions for the operator $\Sigma(p)$.

5. CONCLUSION

It is easily seen that in the presence of a temperature gradient the effective diffusion coefficient of AP in a turbulent medium is a tensor. Choosing the z axis of a Cartesian coordinate frame along the vector $\mathbf{n}_{(u)}$, the diffusion tensor can be described by introducing the longitudinal (in a direction parallel to the temperature gradient) and transverse diffusion coefficients:

$$D_\parallel(\omega)=D_0+B_0\tau_0[1+3\Phi_1(\omega)],$$
$$D_\perp(\omega)=D_0+B_0\tau_0\Phi_2(\omega). \tag{29}$$

The dependence of the effective diffusion coefficient on the parameter ω is obviously due to fluctuations of the thermophoresis velocity, which are in turn due to fluctuations of the temperature field. It is easily seen that the functions $D_\parallel(\omega)$ and $D_\perp(\omega)$ increase monotonically, with the longitudinal diffusion coefficients increasing more rapidly than the transverse one.

For the average thermophoresis velocity we obtain also the effective value

$$\mathbf{u}_{\text{eff}}=\mathbf{u}_0\{1-B_0\tau_0[\tau_0l_0{}^2+\chi^{-1}\Phi_1(\omega)]\}. \tag{30}$$

It follows hence that turbulent pulsations always lower the regular thermophoresis velocity.

Thus, the sought equation for the average AP density takes in the Cartesian frame chosen by us the form

$$\frac{\partial n}{\partial t}+u_{\text{eff}}\frac{\partial}{\partial z}n=D_\perp(\omega)\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right)n+D_\parallel(\omega)\frac{\partial^2}{\partial z^2}n. \tag{31}$$

Note that the condition

$$\omega\ll1, \tag{32}$$

is met for small relative temperature drops, and allowance for it simplifies the obtained expressions.

Omitting the terms $O(\omega^4)$ of the functions contained in (28)–(30), we get

$$\Phi_1(\omega)={}^1/_5\omega^2;\quad \Phi_2(\omega)=1+{}^1/_5\omega^2;\quad \Phi_3(\omega)={}^2/_5\omega^2. \tag{33}$$

In addition to the condition (32), it can be assumed that

$$(\omega/\xi)^2\gg1.$$

This condition makes it possible to disregard in the expressions for the effective diffusion coefficient and for the effective thermophoresis velocity the fourth-order correlations that lead to the terms $O(\xi^2)$ (Ref. 8).

[1] Not being of principal significance, this constraint simplifies the calculations substantially. Note also that the ensuing results can also be used as estimates for the case of larger temperature drops.

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