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High-Frequency Thermometry*

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Recent advances in the technology of fast-response thermal detectors and their application to the measurement of short-time thermal transport effects have made it expedient to make some theoretical estimates with which the experimental results may be compared. A dispersion relationship between the applied frequency of thermal oscillation and its associated wave vector is therefore derived. This relationship, besides involving the diffusion constant of the medium through which the thermal transport is proceeding, depends on three new parameters—two characteristic times and a characteristic length. Formulas are presented for these parameters in terms of the appropriate averages over the phonon energy spectrum of the microscopic interaction rates which govern the transport. Hence, from measurements of the thermal dispersion and the parameters which govern it, further information on these microscopic rates can be deduced.

I. INTRODUCTION

CONSIDERABLE progress has been made recently in the technology of fast-response thermometers. In particular, alloy-film superconducting bolometers[1–3] —or resistance thermometers—have been fabricated which have submicrosecond response times. This technology has been further stimulated by theoretical conjectures on the existence of second sound in solids.[4–7] Fast bolometers were employed to search for this phenomenon.[3]

In the light of this new technology we wish to explore theoretically the question: What new information, if any, can we gather by utilizing the dimension of time in making thermal measurements? Presently, one measures equilibrium or steady-state thermal parameters such as heat capacity or thermal conductivity as functions of temperature. The nature of the microscopic scattering processes obtaining during transport is deduced from the latter through a computer-aided curve-fitting process.[8–10] The usual procedure utilizes formulas for thermal conductivity[11–14] involving the relaxation times for various processes. Computations are made employing expressions for these relaxation times involving adjustable parameters. These parameters are varied to get a best fit to experimental results. Since at low temperatures these times may exceed microseconds, the motivation in the present exposition is to explore theoretically the possibility of utilizing fast thermometry to get more definitive and direct estimates of these times. Several recent computations[5,15] have indicated that there is information obtainable from boundary or size dependence when the scattering mean free path becomes large. Since experimentally, it is far more inconvenient to vary size than to vary rate, it seems expedient to investigate the analogous situation for thermal time effects.

In the following, the transport conditions that obtain when the temperature is allowed to vary rapidly with time will be explored. The scale of thermal time variations characterized by the angular rate $\omega[=i(\partial/\partial t)]$ is assumed to be smaller than most—but perhaps comparable to some—of the inherent relaxation rates which characterize the interactions in the phonon gas. In this connection, it should be pointed out that the case of second sound obtains when ω is greater than some relaxation rates—umklapp ones—and less than others. Here, we explore, not second sound, but simply time-dependent thermometry when the thermal oscillations are less rapid than all collision mechanisms. The term "high-frequency thermometry" indicates fast times technologically, but slow times on the scale of microscopic processes.

The net result of the following will be to derive the differential equation governing the transport of heat in a medium when allowance is made for relatively high-speed thermal fluctuations. This equation, exhibited as (27), involves three new parameters which, like the thermal diffusion constant D, characterize the medium. Expressions for these parameters in terms of the microscopic rates of phonon umklapp and normal relaxation processes are given in Eqs. (24)-(26). In these expressions allowance is made for the dependence of these rates on the phonon energy.

Measurements of these parameters as functions of temperature can yield information on the microscopic relaxation times just as measurements of D or the thermal conductivity do presently. This additional information comes in the form of averages over the

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phonon spectrum of specific combinations of the various phonon relaxation times. This is the same form in which measurements of the thermal conductivity present us with phonon relaxation times.[14] Experimental data on these new parameters, when combined with thermal-conductivity data can, therefore, be made to yield a more complete and consistent picture of phonon collision processes.

Verification of the form of the governing differential equation (27) and the determination of the values of the parameters characterizing this equation is relatively easily accomplished if sufficiently fast-response thermal detectors are employed. In principle, one utilizes a long rod of specimen at one end of which is a heat source. This may be, for example, an evaporated thin metal film subject to Ohmic heating due to a sinusoidally varying current through it. Sinusoidal heating at the film end of the specimen will then occur at a frequency twice that at which the current is driven through the film. At several points down the length of the specimen rod are placed thermal detectors to measure the temperature of the specimen at these locations as a function of time. By observing the amplitude and the phase of the temperature variations at these locations in response to the oscillating heat input, one effectively measures both the real and imaginary parts of a thermal-propagation vector $\mathbf{k}$ (cf. Eqs. (14) and (22)]. The imaginary part corresponds to the attenuation and the real part produces a phase shift. By performing this experiment at a number of different input heating frequencies one may, in principle, measure the entire dispersion relationship $\mathbf{k}=\mathbf{k}(\omega)$ thus making an experimental determination to be compared with Eq. (22). This is no more than the operator form for the thermal differential equation (27). In fact, any empirical determination of $\mathbf{k}=\mathbf{k}(\omega)$ effectively defines the proper differential equation governing the process under study. Hence measurements of the type described are definitive in checking the results outlined here. Furthermore, they can ascertain the values of the key parameters arising in those results.

In practice, the ideal experimental situation outlined may not be expedient. Finite boundaries may be present and the geometry may be multidimensional (instead of one-dimensional as in the foregoing). Furthermore, pulsed experiments are more practical than continuous-wave ones. All of these experimental contingencies require that the proper boundary-value problems, regarding (27), be solved to suit the particular experimental configuration utilized so that the theory may legitimately be compared with experiment. These considerations do not, however, affect the results exhibited in (27) and (22) which obtain regardless of the particular geometrical configuration to which they are applied. They are valid in the bulk locally and hence for any boundary condition.

II. MODEL

To discuss the problem at hand we follow the now familiar model utilized by Callaway.[14] This model is applicable to electrically nonconducting crystals at low temperatures. The medium is characterized by the presence of a phonon gas of mean number N with phonon wave vector $\mathbf{q}$ at the "position" $\mathbf{r}$ and at "time" t. This statement must be understood to be tempered by the usual stipulations on the accuracy with which the position $\mathbf{r}$ is defined with respect to the uncertainty in wave vector $\mathbf{q}$. And a similar consideration obtains for the time definition, Δt, and the phonon energy $\epsilon=\epsilon(q)$. With these in mind, the number N follows the Boltzmann equation:

$$\partial N/\partial t+\mathbf{v}\cdot\nabla N=(\partial N/\partial t)_c, \tag{1}$$

where $\mathbf{v}$ is the phonon group velocity for the particular mode (or wave vector) to which N refers, and $(\partial N/\partial t)_c$ represents the increase in N due to collisions. We will assume, following Callaway, an isotropic crystal and sufficiently low temperatures so that those phonons present suffer no dispersion. In this approximation the velocity $\mathbf{v}$ is

$$\mathbf{v}\equiv(1/\hbar)\nabla_q\epsilon=\mathbf{q}s/q, \tag{2}$$

where s is the appropriate speed of sound for the particular branch of ϵ versus q (longitudinal, transverse) for which the calculation is being done.

The right-hand side of (1) represents the net rate at which phonons are added to the mode under consideration by collisions. This term is approximated by

$$(\partial N/\partial t)_c=-(N-N_\lambda)/\tau_N-(N-N_0)/\tau_U, \tag{3}$$

where

$$N_0\equiv\left[\exp\left(\frac{\epsilon}{KT'}\right)-1\right]^{-1}, \tag{4}$$

and

$$N_\lambda\equiv\left[\exp\left(\frac{\epsilon-\hbar\boldsymbol{\lambda}\cdot\mathbf{q}}{KT'}\right)-1\right]^{-1}. \tag{5}$$

The quantity $T'=T'(\mathbf{r},t)$ represents the local temperature at the position $\mathbf{r}$ and at the time t. This may differ from the ambient temperature T. The parameter $\boldsymbol{\lambda}$ is also a function of $\mathbf{r}$ and t and must ultimately be so chosen as to insure the validity of the dissociation exhibited in Eq. (3). This will be accomplished later by setting

$$\int \mathbf{q}(\partial N/\partial t)_c\, d^3q=-\int \mathbf{q}[(N-N_0)/\tau_U]\, d^3q. \tag{6}$$

The significance of Eq. (6) is as follows: Of the two terms in (3) the first one refers to collisions which conserve the phonon pseudo-momentum $\mathbf{q}$, whereas the second one refers to collisions which do not. But it is well known that an integration over phase space, performed on a collision term multiplied by a weighting factor which is microscopically conserved in collisions, will always be zero.[16] Hence, a zero contribution must arise from the integration with the weighting factor $\mathbf{q}$ when applied to the $\mathbf{q}$-conserving collision term and the remainder is that part exhibited in (6). It is due to the non-$\mathbf{q}$-conserving term. Since both collision terms must conserve energy, we also have

$$\int \epsilon(\partial N/\partial t)_c\, d^3q = 0. \qquad (7)$$

These points together with the motivations and justifications for the approximation of Eq. (3) have all been discussed extensively.[13,14] The basis for Eq. (3) lies in the distinction between normal collision processes and umklapp processes. The former are momentum-conserving and are characterized by the relaxation time $\tau_N = \tau_N(\epsilon)$. The latter may occur through a number of different mechanisms which together are characterized by the rate $1/\tau_U$. This umklapp collision rate is a function of phonon energy.

With this model of the transport of a phonon gas as our microscopic picture of the thermal disturbance, we wish to deduce what parameters determine how the temperature fluctuation $\Delta T(\mathbf{r},t) \equiv T' - T$ and the heat current $\mathbf{Q}(\mathbf{r},t)$ vary with position and time. The high-frequency thermometry experiments may then be interpreted in terms of those parameters.

The calculation will proceed on the basis of several approximations and assumptions in addition to those already inherent in the model. If we call n the deviation from the local equilibrium number of phonons present at $\mathbf{r}$ and t then

$$n \equiv N - N_0(T'). \qquad (8)$$

And further, it is convenient to denote the heat capacity per phonon mode as

$$C(\epsilon) = \epsilon \partial N_0/\partial T' \,|_{T=T'} \equiv \epsilon \partial N_0(T)/\partial T. \qquad (9)$$

With these definitions in mind the calculation assumes

(1) isotropy of the medium,

(2) dispersionless medium for phonons; i.e., $\epsilon_\eta = \hbar s_\eta q$, where η denotes the phonon branch—longitudinal, transverse, acoustic,

(3) that the following dimensionless "amplitudes" are small:

$$\Delta T/T, \quad \lambda/s, \quad \epsilon n/TC(\epsilon) \ll 1; \qquad (10)$$

(4) that the combined relaxation time $t_c(\epsilon)$ satisfies the condition

$$\omega\tau_c(\epsilon) < 1 \qquad (11)$$

for all of the thermal phonon energies involved (energies greater than about 1°K), where by definition

$$1/\tau_c \equiv 1/\tau_U + 1/\tau_N, \qquad (12)$$

but that the quantity $\omega\tau_c$ is not completely negligible and is amenable to experimental investigation.

III. CALCULATION

Equations (1) and (3) may be solved by standard methods[5,7,14] to obtain an expression for n in terms of the amplitudes ΔT and $\boldsymbol{\lambda}$. The solution is obtained by a straightforward substitution of the definition (8) into the combination of (1) and (3). All of the time and space variations are contained within the parameters ΔT and $\boldsymbol{\lambda}$. Employing the operators

$$\omega = i\partial/\partial t, \qquad (13)$$

and

$$\mathbf{k} = -i\boldsymbol{\nabla}, \qquad (14)$$

and incorporating the assumptions mentioned, we find

$$n = \frac{TC(\epsilon)}{\epsilon}(1 - i\tau_c\omega + i\tau_c\mathbf{v}\cdot\mathbf{k})^{-1}\left(\frac{\tau_c}{\tau_N}\frac{\mathbf{q}\cdot\boldsymbol{\lambda}}{qs} + i\tau_c(\omega - \mathbf{v}\cdot\mathbf{k})\frac{\Delta T}{T}\right). \qquad (15)$$

To utilize this result we must determine two things: The relationship between the amplitudes $\boldsymbol{\lambda}$ and ΔT and the "dispersion" relationship ω versus $\mathbf{k}$ for the thermal disturbance. These are obtained by inserting the result (15) back into Eqs. (1) and (3) and integrating first with the weighting factor $\mathbf{q}$ and then with the weighting factor ϵ. These are the significant weighting factors with which to integrate because with them the separation of the collision term into normal and umklapp processes is brought into self-consistency. This procedure yields a macroscopic pair of equations which utilize the information contained in the conditions (6) and (7). This new pair of equations involves both the amplitudes ΔT and $\boldsymbol{\lambda}$ homogeneously. They, therefore, have simultaneous solutions only for a unique ω-versus-$\mathbf{k}$ relationship. A determinant expresses this relationship. Once the dispersion ω versus $\mathbf{k}$ is known either one of the two equations can be used to ascertain the dependence of $\boldsymbol{\lambda}$ on ΔT. Thus the two essential connections $\boldsymbol{\lambda}$ versus ΔT and ω versus $\mathbf{k}$ are made.

Proceeding with this progaam the energy condition (7) in combination with (1) becomes

$$\frac{\partial}{\partial t}\left(\sum_\eta \frac{1}{(2\pi)^3}\int \epsilon N\, d^3q\right) + \boldsymbol{\nabla}\cdot\mathbf{Q} = 0. \qquad (16)$$

This is no more than a statement of the conservation of energy. The first term is simply the time rate of change of the internal energy per unit volume. The summation is taken over the three different modes of propagation—two transverse and one longitudinal—labeled by the index η. In terms of the amplitudes ΔT and $\boldsymbol{\lambda}$, Eq. (16) takes the form shown below. In this expression the solution exhibited in Eq. (15) for the

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[16] F. Reif, *Statistical and Thermal Physics* (McGraw-Hill Book Company, Inc., New York, 1965), Chap. 14.4.

phonon distribution function has been employed with terms kept only out to the appropriate order. This order is determined by the condition that in first approximation k^2 goes as ω and λ goes at $k\Delta T$. This notion implies for example, that the terms $k^3\Delta T$, $k^2\lambda$, $\omega k\Delta T$ and $\omega\lambda$ are all of about the same order. And, in fact, it is to just this order that one must calculate. With these features in mind, and after considerable algebraic manipulation, Eq. (16) becomes

$$\tfrac{1}{3}\langle R\rangle i\mathbf{k}\cdot\boldsymbol{\lambda}-[i\omega+\tfrac{1}{3}(ik)^2\langle s^2\rangle_{\text{av}}\langle\tau_c\rangle]\Delta T/T=0. \tag{17}$$

In this equation, and the ones following, it is useful both for notational reasons and for physical interpretation to use a shorthand notation for averages of the various relaxation times and for averages over the modes of propagation—transverse, at a speed s_t, and longitudinal, at a speed s_l. For any function of phonon energy $A(\epsilon)$ the average of this function, denoted by $\langle A\rangle$, means

$$\langle A\rangle\equiv\int C(\epsilon)A(\epsilon)\,d^3q\Big/\int C(\epsilon)\,d^3q. \tag{18}$$

The symbol R is just a shorthand notation for the ratio (always less than unity)

$$R\equiv\tau_U(\epsilon)[\tau_U(\epsilon)+\tau_N(\epsilon)]^{-1}. \tag{19}$$

This ratio indicates to what extent normal processes dominate. The ratio R is unity if only normal processes obtain and zero if there are no normal processes.

By the speed average $\langle s^m\rangle_{\text{av}}$ is meant

$$\langle s^m\rangle_{\text{av}}\equiv\left(\frac{2}{s_t^{3-m}}+\frac{1}{s_l^{3-m}}\right)\left(\frac{2}{s_t^3}+\frac{1}{s_l^3}\right)^{-1}. \tag{20}$$

The momentum equation, obtained by multiplying (1) through by $\mathbf{q}$, integrating and utilizing the condition (6), yields the result in Eq. (21). There is a considerable amount of algebra in this step.

$$\begin{aligned}\tfrac{1}{3}\langle s^{-2}\rangle_{\text{av}}(\langle R/\tau_U\rangle-i\omega\langle R^2\rangle)\boldsymbol{\lambda}&-(1/15)\langle R^2\tau_c\rangle\\ \times[(ik)^2\boldsymbol{\lambda}+2i\mathbf{k}(i\mathbf{k}\cdot\boldsymbol{\lambda})]&+\{\tfrac{1}{3}\langle R\rangle+\tfrac{2}{3}i\omega\langle R\tau_c\rangle\\ &+\tfrac{1}{5}\langle s^2\rangle_{\text{av}}(ik)^2\langle R\tau_c^2\rangle\}i\mathbf{k}\Delta T/T=0.\end{aligned} \tag{21}$$

This equation is, of course, valid to the same order of approximation as (17). And the pair of equations (17) and (21) are the homogeneous relations in $\boldsymbol{\lambda}$ and ΔT mentioned formerly.

The first point to note about (21) and (17) is that, as one expects on physical grounds, $\mathbf{k}$ and $\boldsymbol{\lambda}$ are collinear. The physical grounds rest on the realization that the medium is assumed to be isotropic and hence there is no other direction characteristic of the problem but that of $\mathbf{k}$. The mathematical proof follows from forming the cross product of (21) with $\mathbf{k}$ (or $\boldsymbol{\lambda}$) and noting that a nonzero constant multiplied by $\mathbf{k}\times\boldsymbol{\lambda}$ is zero only if $\mathbf{k}\times\boldsymbol{\lambda}=0$. The fact that the numerical coefficient of $\mathbf{k}\times\boldsymbol{\lambda}$ is indeed not zero follows from noting the inconsistencies obtained in comparing the results of the cross products of Eq. (21) with $\boldsymbol{\lambda}$ and with $\mathbf{k}$ on the assumption that $\mathbf{k}\times\boldsymbol{\lambda}$ is not zero. Utilizing this result plus the fact that simultaneous solutions for ΔT and $\boldsymbol{\lambda}$ exist only if the proper relation between their coefficients obtains one may find this relation. It is an equation which is quadratic in the quantity $(ik)^2$ and of the form

$$\alpha(ik)^4+(ik)^2(1-i\omega\tau_1)+(i\omega/D)(1-i\omega\tau_2)=0, \tag{22}$$

where the parameters called α, D, τ_1, and τ_2 are symbols for combinations of relaxation-time averages and speed-average values:

$$D=\tfrac{1}{3}[\langle R\rangle^2/\langle s^{-2}\rangle_{\text{av}}\langle R/\tau_U\rangle]+\tfrac{1}{3}\langle s^2\rangle_{\text{av}}\langle\tau_c\rangle, \tag{23}$$

$$\begin{aligned}\tau_1=(\langle s^{-2}\rangle_{\text{av}}\langle R/\tau_U\rangle D)^{-1}(\tfrac{1}{3}\langle s^2\rangle_{\text{av}}\langle s^{-2}\rangle_{\text{av}}\langle\tau_c\rangle\langle R^2\rangle&\\ +\tfrac{3}{5}\langle R^2\tau_c\rangle-\tfrac{2}{3}\langle R\rangle\langle R\tau_c\rangle),&\end{aligned} \tag{24}$$

$$\tau_2=\langle R^2\rangle\langle R/\tau_U\rangle^{-1}, \tag{25}$$

$$\begin{aligned}\alpha=\tfrac{1}{5}\langle s^2\rangle_{\text{av}}(\langle s^{-2}\rangle_{\text{av}}\langle R/\tau_U\rangle D)^{-1}(\langle R\rangle\langle R\tau_c^2\rangle&\\ -\langle R^2\tau_c\rangle\langle\tau_c\rangle).&\end{aligned} \tag{26}$$

IV. DISCUSSION

It is clear from the form of (22) that the symbols D, τ_1, τ_2, and α may be interpreted respectively as the thermal diffusion constant, two characteristic times and the square of a characteristic length for the process of thermal transport. Furthermore, by the meaning of the operators $i\mathbf{k}$ and $-i\omega$ Eq. (22) implies that thermal disturbances follow the form

$$\begin{aligned}\alpha\nabla^4T'+\nabla^2T'+\tau_1\nabla^2\partial T'/\partial t-(1/D)\partial T'/\partial t&\\ -(\tau_2/D)\partial^2T'/\partial t^2=0.&\end{aligned} \tag{27}$$

In the limit that the new parameters α, τ_1 and τ_2—which have arisen here because of the higher order to which the calculation has been extended—are taken to be zero, then (27) reduces to the familiar diffusion equation as it should. However, as the frequency ω at which the temperature is caused to oscillate gets large, the other terms in the dispersion formula begin to play a role.

One may insert the result (22) back into (17) or (21) to discover how $\boldsymbol{\lambda}$ depends upon ΔT. Utilizing this together with (22), the proper definition of $\mathbf{Q}$, the heat current density, and keeping only first-order terms, one finds the familiar result

$$\mathbf{Q}=-\kappa\nabla T'+\text{higher order terms}. \tag{28}$$

The parameter κ, which obviously must be interpreted as the thermal conductivity, is found by this procedure to be given by

$$\kappa=CD, \tag{29}$$

where C is the specific heat and D is the diffusion constant given in Eq. (23). This result for κ in terms of relaxation-time averages reduces to just that of Callaway[14] in the limit $\langle s^{2m}\rangle_{\text{av}}=\langle s^2\rangle_{\text{av}}{}^m$. This assumption was inherent in Callaway's calculation but not in the present calculation. At most, something approaching a 25% error results from Callaway's simplifying approximation.

Contact is made with a calculation by Sussmann and Thellung[5] by taking the limiting case of our result where no umklapp processes may occur, $\tau_U \to \infty$, $R \to 1$. They considered a phonon gas where only normal processes obtained and where the phonon collision relaxation time was independent of phonon energy. They allowed for different transverse and longitudinal velocities, however, as we have done.

Equation (22) represents an expression for $(ik)^2$ as a function of applied thermal frequency ω. Its validity is restricted to the regime $\omega\tau_1$, $\omega\tau_2 < 1$. This condition is a more relaxed one than the original one upon which the calculation was based. It is the macroscopic analog of the microscopic condition $\omega\tau_c < 1$. The latter is harder to justify since, in fact, τ_c is not a number. It is a function and takes on a vast range of values depending upon the phonon energy to which it refers. The mean macroscopic thermal times, τ_1 and τ_2 are simply numbers. They depend only on the ambient background temperature T through the Eqs. (24) and (25).

The diffusion constant D (or the thermal conductivity κ) measures some complicated phonon energy average of microscopic collision times. The particular combination of relaxation time averages which κ measures was originally pointed out by Callaway. It is exhibited in Eq. (23). Theoretical estimates of these relaxation times inserted into (23) yield D (or κ) as a function of temperature, and this result may be compared with experimental measurements of $\kappa(T)$.

The same process may be employed for the characteristic times $\tau_1 = \tau_1(T)$ and $\tau_2(T)$. Theoretical estimates of $\tau_2(T)$, $\tau_1(T)$, and $\alpha(T)$ would utilize the same information as that employed for $\kappa(T)$ since the same microscopic relaxation times are involved. The characteristic parameters α, τ_1 and τ_2 are simply different combinations of the averages of these times. Hence, both a check and further information on phonon relaxation rates is provided by measuring these parameters as well as $D(T)$ [or $\kappa(T)$].

As might be expected, on physical grounds the characteristic time τ_2 reflects the dominance of normal or umklapp processes. Large positive values of τ_2 produce thermal propagation which is less diffusive and more wavelike. This regime obtains, as might be expected, when non-umklapp or normal processes dominate, i.e., $R \to 1$. On the other hand, if umklapp processes dominate ($R \to 0$), τ_2 becomes very small (as does D) so that the transport process is highly diffusive.

When the applied thermal frequency ω gets very large so that it exceeds some of the microscopic relaxation rates, then (22) is no longer valid.

V. CONCLUSION

It has been the purpose of the foregoing to find what extra information regarding phonon interactions in solids may be obtained by utilizing high-frequency thermometry. This information is contained in the dispersion relation (22), in combination with Eqs. (24), (25), and (26) for the values of the parameters $\tau_1(T)$, $\tau_2(T)$, and $\alpha(T)$. This study was motivated by the appearance of fast thermal detectors as technological tools. Measurements utilizing such detectors are easily capable of directly extracting the dispersion relation between applied thermal frequency and wave vector. Such measurements can evaluate both the form of the dispersion and the amplitudes of the appropriate parameters characterizing the relationship. Thus, the experimental results may be compared with the deduction (22) where the parameters τ_1, τ_2, α, and $D(T)$ have been evaluated and are given in Eqs. (23)–(26). The measurements of these parameters may then be compared with theoretical predictions regarding the form of the relaxation times $\tau_N(\epsilon)$ and $\tau_U(\epsilon)$.

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