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## Exchange and Thermal Conductivity in bcc He<sup>3</sup> Containing He<sup>4</sup> Impurity

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Using recent theory for solid He<sup>3</sup>, prediction of the large increase in exchange constant  $J$  and thermal resistance  $K^{-1}$  due to He<sup>4</sup> impurity is attempted. Using variational methods, a large change in the He<sup>4</sup> function but little relaxation ( $\sim 0.75\%$ ) around the He<sup>4</sup> or change in the neighboring He<sup>3</sup> functions is found. An increase in  $J$  by a factor of  $\sim 1.6$  only is directly predicted. If a large relaxation ( $\sim 5\%$ ) or He<sup>3</sup> wave-function change is assumed, a large increase, but to positive  $J$  only, occurs. Direct evaluation of the change in the self-consistent force constants around the He<sup>4</sup> fails to predict the large defect resistance. It can be predicted using a continuum model, and calculating the local compressibility change which suggests that large relaxation around the He<sup>4</sup> may not be needed to explain  $K$ .

### I. INTRODUCTION

Small concentrations of He<sup>4</sup> have a profound effect on the relaxation characteristics of the nuclear spin system<sup>1</sup> and on the thermal conductivity<sup>2,3</sup>  $K$  of bcc He<sup>3</sup>. The change in the spin system may be crudely expressed as a locally enhanced exchange constant  $|J|$  between two He<sup>3</sup> atoms near an He<sup>4</sup>. For a concentration  $x \approx 0.5\%$   $|J|$  increases by a factor of  $\sim 5-10^1$  depending on how many pairs have an increased  $J$ .

When containing 1% He<sup>4</sup>, the thermal resistance increases to  $\sim 10$  times the pure crystal value.<sup>2</sup> This extra point-defect resistance is usually expressed as phonon relaxation time<sup>4</sup>

$$\tau_{\text{pt}}^{-1} = A\omega^4. \quad (1)$$

If only the mass change  $\delta m$  were effective,

$$A = A_{\text{mass}} = x(1-x)V_a^2 \left(\frac{\delta m}{m}\right)^2 / 4\pi c^3,$$

the Rayleigh formula for scattering of sound waves from a local region of mass change which is the same for both continuous<sup>5</sup> and discrete media.<sup>6</sup> Here  $V_a$  is the volume per atom and  $c$  is the velocity of sound. Analysis of experiment<sup>3</sup> leads to  $A/A_{\text{mass}} \approx 10-30$  so that some defect property other than  $\delta m$  is largely responsible for  $\tau_{\text{pt}}$ . Klemens and Maradudin<sup>7</sup> have considered the effects due to assumed relaxation around the He<sup>4</sup>. Following their expression a spacing change  $\delta R/R \approx 4.5\%$  is needed to explain  $A/A_{\text{mass}}$ .

In this paper we wish to report some first efforts to predict these effects using recent theory of quantum crystals at  $T=0^\circ\text{K}$ . We use a mixture of the single-particle theory due to Nosanow<sup>8</sup> and the concept of the self-consistent harmonic approximation as developed by Koehler.<sup>9</sup> As we are discussing point defects we wish to emphasize the single-particle aspects of the system and therefore follow more closely the work of Nosanow.

In the following section we outline the theory and

apply it to one He<sup>4</sup> in bcc He<sup>3</sup>. The results are used to calculate  $J$  and  $\tau_{\text{pt}}^{-1}$  in Secs. III and IV. These are discussed in Sec. V.

## II. THEORETICAL OUTLINE AND APPLICATION TO 1 He<sup>4</sup> IN He<sup>3</sup>

The exact Hamiltonian for the crystal is, with  $\lambda^2 \equiv \hbar^2/m$ ,

$$H = -\frac{1}{2}\lambda^2 \sum_i \nabla_i^2 + \frac{1}{2} \sum_{i,j} v(\vec{r}_i - \vec{r}_j), \quad (2)$$

where  $v(r_i - r_j)$ , the He-He potential, is taken as the He gas Lennard-Jones 12-6 potential.<sup>8</sup> To describe the displacement of the nuclei from their lattice points, we use the trial ground-state solution of  $H$

$$|O\rangle = (\lambda\pi)^{-\frac{3}{4}N} |G|^{-\frac{1}{4}} \exp[-(1/2\lambda)(\vec{r} - \vec{R})\vec{G}(\vec{r} - \vec{R})]. \quad (3)$$

This is the exact ground-state solution of the Schrödinger equation for a harmonic crystal with Hamiltonian

$$H_h = -\frac{1}{2}\lambda^2 \sum_i \nabla_i^2 + \frac{1}{2}(\vec{r} - \vec{R})\vec{\Phi}(\vec{r} - \vec{R}). \quad (4)$$

The choice of a harmonic trial solution (3) is equivalent to restricting our description to one of harmonic form. Treating  $\vec{G}$  as a variational parameter we minimize  $E_0 = \langle O | H | O \rangle$  to find the best possible harmonic description  $|O\rangle$  of the crystal. This minimization gives the best force constants as<sup>9</sup>

$$\Phi_{ij}^{\alpha\beta} = (G^2)_{ij}^{\alpha\beta} = \left\langle O \left| \frac{\partial^2 v}{\partial r_i^\alpha \partial r_j^\beta} \right| O \right\rangle. \quad (5)$$

To emphasize the single-particle aspects we restrict  $\vec{G}$  to diagonal form so that  $|O\rangle$  is a product of single nuclei functions

$$\phi_i = (A_i/\pi)^{3/4} \exp[-\frac{1}{2}A_i(\vec{r}_i - \vec{R}_i)^2].$$

As  $\phi_i$  has bad form at large  $(\vec{r}_i - \vec{R}_i)$ , we multiply by a correction

$$f(\vec{r}_i - \vec{r}_j) = \exp[-Cv(\vec{r}_i - \vec{r}_j)]$$

to account for short-range correlations between pairs of nuclei. Then

$$|O\rangle = \prod_i \phi_i \prod_{i < j} f_{ij}$$

and  $E_0$  becomes, keeping up to two-body terms in an energy cluster expansion,<sup>8</sup>

$$E_0 \approx \frac{3\hbar^2}{4} \sum_i \left( \frac{A_i}{m_i} \right) + \frac{1}{2} \sum_{i \neq j} \langle f_{ij}^2 \rangle^{-1} \times \langle \phi_i \phi_j | w(r_i - r_j) | \phi_i \phi_j \rangle, \quad (6)$$

where

$$w(\vec{r}) = f^2[v(\vec{r}) - (\hbar^2/2m)\nabla^2 \ln f].$$

In this approximation the self-consistent force constants become

$$\Phi_{ij}^{\alpha\beta} = \left\langle \phi_i \phi_j \left| \frac{\partial^2}{\partial r_i^\alpha \partial r_j^\beta} w(\vec{r}_i - \vec{r}_j) \right| \phi_i \phi_j \right\rangle. \quad (7)$$

In the pure lattice with translational symmetry each atom is identical so we may equate each  $A_i$  to a common value  $A_i = A_p$ . Near a substitutional He<sup>4</sup> impurity, however, this symmetry is lost. If we assume, as seems reasonable for a point defect, that the He<sup>4</sup> causes a spherically symmetric distortion, then all atoms at a given distance from the He<sup>4</sup> should have similar distributions (identical  $A_i$ ) and should be displaced in the same way. To describe this new spherical symmetry we assign a distinct  $A_i = A_0$  to the central He<sup>4</sup>, another  $A_i = A_1$  to all its first neighbors,  $A_i = A_2$  to its second neighbors, and so on out to its fifth neighbors. Beyond this we assume the effect of the He<sup>4</sup> negligible and again set all other  $A_i = A_p$ . The first to fifth neighbor mean positions are also allowed to relax radially inward or outward with the He<sup>4</sup> as center. As  $C$  varies little with volume or isotope considered in the pure lattice,<sup>8</sup> it was held fixed at  $C = C_p = 4.35 \times 10^{-3} \text{K}^{-1}$ .

To find the best  $A_i$ , spacings  $R_{0i}$ , and an upper bound to  $E_0$ , in the presence of a He<sup>4</sup>, we minimize  $E_0$  with respect to the six quantities  $A_i = A_0 \cdots A_5$  and the five spacings  $R_{0i} \cdots R_{05}$ . As a check, and to specifically note the effect of the He<sup>4</sup>, this was done first with He<sup>3</sup> at the central defect site and then with He<sup>4</sup>.

In Table I are listed the  $A_i$  and  $R_{0i}$  for the central atom and its first and second neighbors with (a) He<sup>3</sup> and (b) He<sup>4</sup> at the center. Only the 1st neighbor equilibrium position changes, relaxing inward by  $\sim 0.75\%$  at each volume, and  $A_1$  increases somewhat. The third to fifth neighbor  $A_i$  and  $R_{0i}$  were negligibly changed (and are omitted here) so that the He<sup>4</sup> does not produce an extended distortion in this approximation.

Using the  $A$  values of Table I we find that the effective He<sup>4</sup>-He<sup>3</sup> potential well

$$(V(\vec{R}_{ij})) = \langle \phi_3 \phi_4 | w(\vec{r}_i' - \vec{r}_j' + \vec{R}_{ij}) | \phi_3 \phi_4 \rangle$$

is deeper and has its minimum at closer approach than the He<sup>3</sup>-He<sup>3</sup> effective potential (see Fig. 1). This means that the first neighbors have moved inward and narrowed their densities in order to rest, on average, more near the minimum of this self-consistent potential. If we had not restricted the  $\phi$  to be spherically symmetric, this suggests that the first neighbors would like to further narrow their density in the direction of the He<sup>4</sup> but, to prevent a large kinetic-energy increase,  $\phi$  would spread perpendicular to the He<sup>3</sup>-He<sup>4</sup> line in the direction of a neighboring He<sup>3</sup>. This effect, so important for exchange, is prevented by the imposition of spherical symmetry on  $\phi$  so that here  $\phi$  has simply narrowed slightly all round.

## III. EXCHANGE NEAR AN He<sup>4</sup>

Generalizing an expression for  $J$  due to Nosanow

TABLE I. Parameters  $A_i$  and distances  $R_{0i}$  of central atom, its first and second neighbors with (a)  $\text{He}^3$  and (b)  $\text{He}^4$  as central atom.

$V$ (cc/mole)	$R_p$ (Å)	$A_p$ (Å <sup>-2</sup> )	Central atom (Defect)		First neighbor		Second neighbor	
				$A_0$	$A_1$	$R_{01}$	$A_1$	$R_{02}$
24.45	3.750	1.290	(a)	1.280	1.280	3.750	1.280	3.985
			(b)	1.505	1.295	3.722	1.285	3.985
21.65	3.600	1.480	(a)	1.480	1.480	3.600	1.480	4.158
			(b)	1.745	1.490	3.575	1.485	4.158
19.05	3.450	1.665	(a)	1.665	1.665	3.450	1.665	4.331
			(b)	1.955 (± 0.005)	1.680 (± 0.002)	3.422 (± 0.002)	1.675	4.331

and Mullin<sup>10</sup> (to  $\text{He}^3$  atoms having nonidentical  $A$  values), we have calculated  $J$  for  $\text{He}^3$  pairs around the  $\text{He}^4$  having the various values of  $A_i, A_j$  and separations  $R_{ij}$  occurring in Table I. Only small increases in  $J$  above the pure lattice  $J_p$  were found with the largest being a factor of  $\sim 1.6$  occurring between the first and fifth neighbors to the  $\text{He}^4$  at  $V = 24.45$  cc/mole.

To investigate whether the observed increase could be predicted using similar wave functions, we have calculated  $J(A, R_{ij})$  as a function of the distance  $R_{ij}$  between two  $\text{He}^3$  nuclei for four different values  $A_p$  (Fig. 2). The four  $A_p$  values used are those which are found on minimizing the pure lattice energy at  $R_p = 3.45, 3.55, 3.65,$  and  $3.75$  Å, respectively. The cross on each  $A_p$  curve marks where  $R_{ij} = R_p$  and therefore marks the pure lattice  $J_p$ . From Fig. 2 we see that a locally decreased  $R_{ij}$  by  $\sim 5\%$  or a decreased  $A$  with  $R_{ij} \approx R_p$  would provide a large increase in  $|J|$ , but only to positive, ferromagnetic  $J$ . However, using the larger  $A$  values obtained for the pure lattice when three-body terms are included<sup>11</sup> no increase to large or positive  $J$  for reasonable  $R_{ij}$  or  $A$  is obtained.

On quite general grounds we expect the pair ground-state function to be symmetric giving  $J < 0$ . Although the above  $J > 0$  cannot be taken too

seriously as  $R_{ij}$  and  $A$  have been arbitrarily varied, Fig. 2 suggests that a large  $J < 0$  will be difficult to obtain with this form of wave function. The picture may be changed somewhat when non-spherical Gaussian functions are used, but it is likely that again  $J > 0$  would be obtained unless a new form of  $f_{ij}$  is used. This further points to the need for an improved correlation function, and much work seems needed to consistently predict  $J$  near an  $\text{He}^4$ .

#### IV. THERMAL RESISTANCE DUE TO $\text{He}^4$

As the functions  $\phi_i$  and spacings  $R_{ij}$  around the  $\text{He}^4$  change, the self-consistent force constants are locally changed. With only  $\phi_4$  and  $R_{34}$  changed significantly, most of this change in our effective harmonic description may be expressed as a small perturbation on  $H_h$  of the form

$$H' = \sum_{j < 0} \delta\Phi(\tau_j) u_{0j} u_j, \quad (8)$$

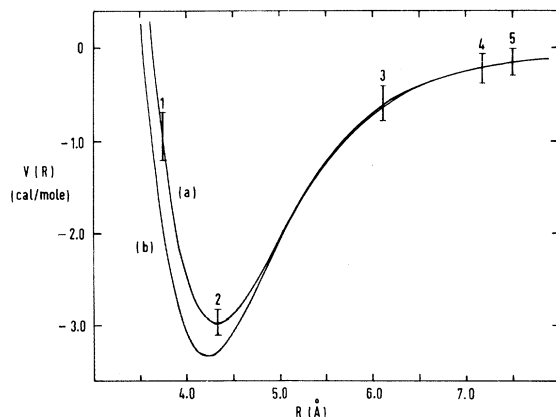


FIG. 1. The effective potential  $\langle O|w|O\rangle$  between (a)  $\text{He}^3$ - $\text{He}^3$  and (b)  $\text{He}^3$ - $\text{He}^4$  atoms for the wave-function parameters in Table I at  $V = 24.45$  cc/mole. The bars mark the distance of the first to fifth neighbors.

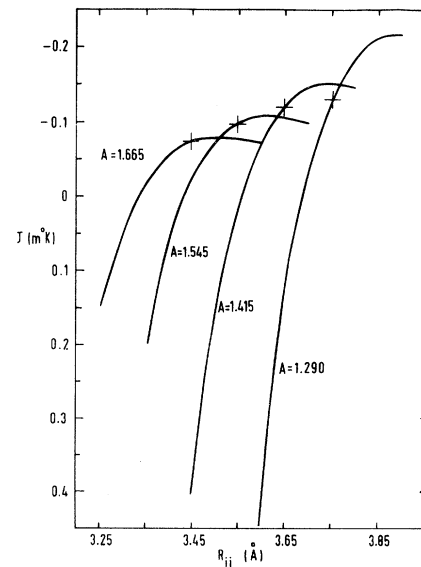


FIG. 2. The exchange constant  $J$  ( $\text{m}^\circ\text{K}$ ) between two  $\text{He}^3$  nuclei as a function of the mean separation  $R_{ij}$  for four pure lattice Gaussian wave-function parameters  $A_p$ . The cross marks where  $R_{ij} = R_p$ , the pure lattice interatomic spacing corresponding to the  $A_p$  value used.

where  $\delta\Phi(\tau_j)$  is the force-constant change between the  $j$ th He<sup>3</sup> and the central atom on inserting the He<sup>4</sup>,  $\vec{\tau}_j \equiv \vec{R}_{0j}$  and  $\vec{u}_j \equiv \vec{r}_j - \vec{R}_j$ . This  $H'$  causes elastic collisions between the self-consistent phonon states  $k$ .

Using first-order time-dependent perturbation theory we may calculate the rate of change of state occupation  $n_k$  due<sup>12</sup> to  $H'$ . Assuming (1) that the distribution of He<sup>4</sup> is random, (2) that all branches may be treated as equivalent, and (3) that it is meaningful to define a separate collision time due to defects for each state  $k$ , then  $H'$  gives

$$\frac{1}{\tau_{pt}} = x(1-x) \frac{3V_a^2}{\pi c^3} \left( \frac{\sum_{\tau \neq 0} \tau_{\alpha}^2 \delta\Phi^{\alpha\alpha}(\tau)}{\sum_{\tau \neq 0} \tau_{\alpha}^2 \Phi^{\alpha\alpha}(\tau)} \right)^2 \omega^4 \equiv A_{fc} \omega^4 \quad (9)$$

for long wave phonons along the [100] direction.

Using (7) and the wave-function parameters of Table I to calculate the expression in the large brackets in (9), we find  $A_{fc} \approx A_{mass}$  (see Table II).  $A_{fc}$  increases slightly with volume but is always  $\sim A_{mass}$ . This approach fails to predict the large  $A$  – most probably because expression (8) is too simple and restrictive to account for the complicated vibrational changes that occur around the He<sup>4</sup>. These would be considerably altered if the first neighbor He<sup>3</sup> atoms adopt non-spherical  $\phi$  as is suggested by the calculation in Sec. II. Also the pure lattice sound velocity  $c$  does not change rapidly enough with volume suggesting that the  $\Phi$  are not sufficiently sensitive to changes in  $A$ . Although (9) is crudely derived, we do not expect it to be in error by an order of magnitude.

To account for these changes in a broad way, we go back to a continuum model of the lattice. This is not unreasonable as the rms vibrational amplitude is  $\sim \frac{1}{3}R$ . Also the continuum expression  $c^2 = (\kappa\rho)^{-1}$ , where  $\kappa$  is the compressibility and  $\rho$  is the density, gives reasonable values for the sound velocity. (560 m/sec compared with the observed 680 m/sec<sup>13</sup> at  $V=20.2$  cc/mole and about 100 m/sec below the observed value at all volumes.) The original Rayleigh formula<sup>5</sup> for scattering of sound waves from a spherical region of mass and compressibility change in a continuum then gives, written as a long-wave relaxation time,

TABLE II. Calculated and observed values of  $A/A_{mass}$ , with  $\tau_{pt}^{-1} = A\omega^4 = (A_{mass} + A_{other})\omega^4$ .

$V$ (cc/mole)	$A/A_{mass}$ calculated fc Eq. (9)	$A/A_{mass}$ calculated $\delta\kappa$ Eq. (10)	$A/A_{mass}$ observed (Refs. 2, 3)
19.5	1.5	9	9–27
20.2	1.6	15	~23
21.0	1.7	20	
22.0	1.9	33	

$$\begin{aligned} \tau_{pt}^{-1} &= A\omega^4 \\ &= \frac{x(1-x)}{4\pi c^3} \left[ V_m^2 \left( \frac{\delta m}{m} \right)^2 + V_K^2 \left( \frac{\delta\kappa}{\kappa} \right)^2 \right] \\ &= A_{mass} \left[ 1 + \left( \frac{V_K (\delta\kappa/\kappa)}{V_m (\delta m/m)} \right)^2 \right] \omega^4. \end{aligned} \quad (10)$$

Here  $V_m$  and  $V_K$  are the volumes occupied by the mass and compressibility changes and for one isotope  $V_m = V_a$ .

To evaluate the local compressibility change around the He<sup>4</sup>, a 50-50 He<sup>3</sup>-He<sup>4</sup> crystal was made up using the wave-function parameters  $A_0$  and  $A_1$  for the He<sup>4</sup> and He<sup>3</sup> and the corresponding spacing  $R_{01}$ . In such a 50-50 bcc mixture, each He<sup>4</sup> is surrounded entirely by He<sup>3</sup> (and vice versa) so that in this way we make up a bulk crystal of units having the same geometry, spacings, and wave functions as found locally around the He<sup>4</sup>. By evaluating the compressibility of this mixture using  $\kappa^{-1} = V\partial^2 E_0 / \partial V^2$  and comparing it with a similar calculation for pure He<sup>3</sup> (see Fig. 3), we may determine  $(\delta\kappa/\kappa)$  between the He<sup>4</sup> impurity and its first neighbors. With the first neighbors included in the local change  $V_K = 5.7V_a$  and the resulting values of  $A/A_{mass}$  are listed in Table II. This procedure should give a realistic estimate of  $\tau_{pt}^{-1}$  but only an estimate.

As the kinetic energy changes more rapidly than the potential with volume,  $\kappa$  is predominately fixed by the kinetic energy.  $\delta\kappa$  arises here then from the local kinetic energy change due to  $A_j - A_0$  when the He<sup>4</sup> is inserted. The effect is not directly connected with relaxation and so represents a different approach from that of Klemens and Maradudin.<sup>7</sup>

## V. DISCUSSION

To predict an increased  $J$  near He<sup>4</sup> directly by variational methods, nonspherical functions for

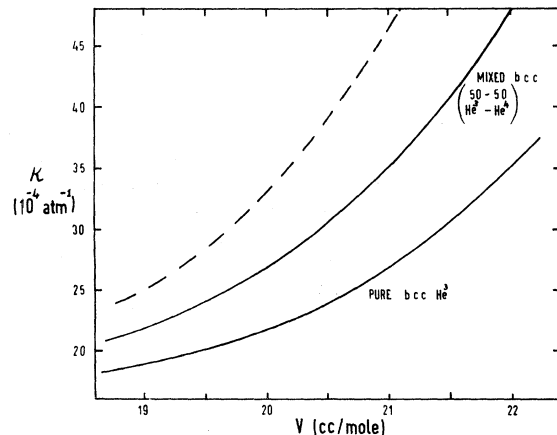


FIG. 3. The compressibility  $\kappa$  for pure bcc He<sup>3</sup> and 50-50 He<sup>3</sup>-He<sup>4</sup> bcc ordered mixture. The dashed line is the calculated  $\kappa$  for hcp He<sup>4</sup> (Ref. 8).

the neighboring  $\text{He}^3$  appear necessary. If these functions assume the form anticipated in Sec. II, then significant relaxation about the  $\text{He}^4$  will not be necessary. To get  $J$  near the observed value an improved form for the correlation function  $f(r_{ij})$  is also needed, and this represents a difficult task. One of the chief difficulties is that those parts of  $|O\rangle$  which are important to  $J$  have little influence on  $E_0$  and so are not well determined by an energy variational method.

Simply evaluating the change in the self-consistent harmonic force constants around the  $\text{He}^4$  does not explain the large observed  $\tau_{\text{pt}}^{-1}$  suggesting that this model is too restrictive. On the other

hand, although  $\tau_{\text{pt}}^{-1}$  can be predicted directly using a continuum model, this model is so crude that it tells us little in detail about the defect.<sup>14</sup> It does suggest, however, that division of defect properties into mass and force constant changes may be artificial and that large relaxation around the  $\text{He}^4$  is not necessarily needed to explain  $\tau_{\text{pt}}^{-1}$ .

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<sup>14</sup>The model is reasonable in this case as  $T \lesssim \theta/10$  and long wave phonons are unaffected by changes in detail.

## Experimental Heat Capacity of Pure Liquid $\text{He}^3$ †

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Measurements of the heat capacity of pure  $\text{He}^3$  at 0.24 atm obtained by a differencing method are presented for the temperature range from 20–150 m°K. The temperature scale  $T^*$  used is that valid for powdered cerium magnesium nitrate in the form of a right circular cylinder with diameter equal to height. There is excellent agreement between the present and earlier measurements. Considering all low-pressure difference data we find that from 6–125 m°K the ratio of heat capacity to magnetic temperature decreases linearly with increasing temperature. The relationship of the measurements to the temperature scale and to theories of spin fluctuations in  $\text{He}^3$  is also discussed.

### I. INTRODUCTION

In recent years the heat capacity of pure  $\text{He}^3$  has been of considerable interest. It is related to the Landau theory of Fermi liquids.<sup>1</sup> It bears directly on the question of superfluidity in  $\text{He}^3$ .<sup>2,3</sup> What is known of its temperature dependence was

not predicted by theory and has been the subject of theoretical speculation.<sup>4,5</sup> Owing to the possibility of a superfluid transition, most recent measurements have emphasized the properties of  $\text{He}^3$  at the lowest achievable temperatures.<sup>3,6</sup> As a result, in no single experiment have accurate measurements been made in the temperature range,