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Supersolidity and the Thermodynamics of Solid Helium

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*The specific heat of solid helium in the temperature range below about 1.2 K has been found to contain a term varying as T^7 , in addition to the usual T^3 contribution always found in a crystalline dielectric solid. It has been proposed by Anderson, Brinkman and Huse, (Science **310**, 1164 (2005)) that the existence of this T^7 term supports their theory of supersolidity. However, in this paper we show that corrections to the phonon specific heat arising from phonon dispersion are much larger than expected based on simple order of magnitude estimates and, as a consequence, it is very unlikely that the existence of this T^7 term can be considered as evidence for supersolidity.*

KEY WORDS: solid helium; specific heat; supersolid.

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1. INTRODUCTION

Recently, a number of experiments have reported evidence that suggests that solid helium becomes a superfluid when cooled below a critical temperature T_c . Kim and Chan¹⁻³ have performed torsional oscillator experiments with solid helium over a wide range of pressure and temperature. They found that below T_c the period of the oscillator decreases indicating that a fraction of the mass of the solid helium has become decoupled from the torsional oscillator. This is referred to as non-classical rotational inertia (NCRI). At the time of writing this article, however, the experimental situation is not entirely clear. Rittner and Reppy⁴ have also detected a decoupling of mass in a torsional oscillator experiment but report that the effect can be greatly reduced by annealing the solid helium sample at high temperature. This suggests that the effect is associated with some



33 sort of defect. Initial attempts to detect a steady state superflow, i.e., a dc
34 flow of mass through a solid helium crystal were unsuccessful.^{5,6} However,
35 a recent experiment by Sasaki et al.⁷ was able to detect a flow of mass
36 occurs when a crystal contains grain boundaries. This flow had the char-
37 acteristics of a superflow, i.e., the rate of transport of mass was essentially
38 independent of the driving force and appeared to be limited by a critical
39 velocity.

40 There have been a number of theoretical papers that argue that a su-
41 persolid state does not occur in solid helium that is free of defects.^{8,9} In
42 contrast, Anderson, Brinkman and Huse¹⁰ have presented a theory based
43 on the idea that solid helium is “incommensurate”. In this context, incom-
44 mensurate means that in the ground state the number of lattice sites is
45 greater than the number of atoms. One of the interesting predictions of
46 their theory is that there should be a correction to the low temperature
47 specific heat that varies as T^7 . This would be in addition to the normal T^3
48 term arising from phonons in a dielectric solid at low temperatures. They
49 point out that measurements of the specific heat by Gardner, Hoffer and
50 Phillips¹¹ provide evidence for such a term. Furthermore, they claim that a
51 term with this temperature dependence and magnitude is unlikely to arise
52 from corrections to the low temperature limiting form of the phonon spec-
53 ific heat. Such corrections can arise from two independent mechanisms.
54 Anharmonicity of the phonons does lead to a T^7 contribution but should
55 be very small. The deviation of the phonon dispersion relation from lin-
56 earity gives a contribution which in lowest order goes as T^5 . Anderson
57 et al.¹⁰ point out that such terms are smaller than the T^3 contribution by
58 powers of the parameter $(T/\Theta_D)^2$, where Θ_D is the Debye temperature.
59 Since for solid helium Θ_D is 25 K, they expected that the corrections to
60 the specific heat arising from phonon dispersion should be negligible in
61 the temperature range around 1 K. In this paper we examine this question
62 more closely and show that, in fact, the corrections due to dispersion are
63 very unlikely to be small.

64

2. SPECIFIC HEAT OF PHONONS

65 The specific heat of solid helium has been investigated by a number
66 of authors.¹¹⁻¹⁴ Here, we concentrate attention on the data of Gardner
67 et al.¹¹ which are for pressures close to the melting curve and cover the
68 temperature range 0.35 to around 1.2 K. The data for a molar volume of
69 20.96 cm^3 are shown in Fig. 1. It can be seen from the figure that these
70 data can be fit very well by the sum of the usual T^3 term expected in
71 a dielectric solid at low temperature together with an extra contribution
72 going as T^7 . Can the existence of this T^7 term be understood in terms of

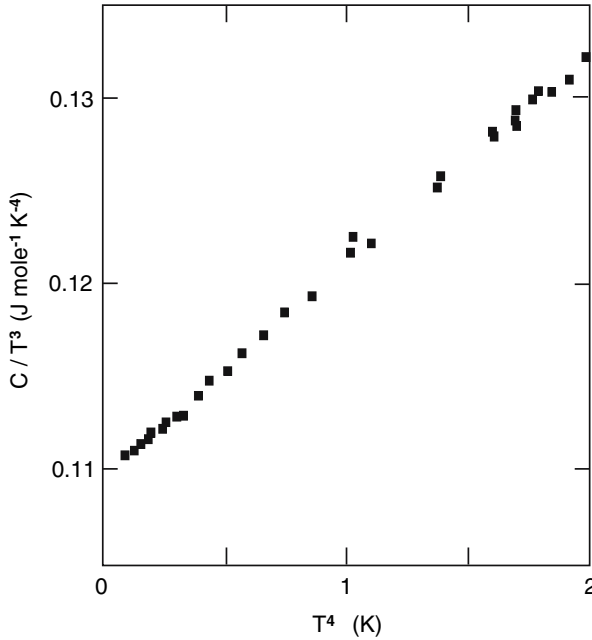


Fig. 1. Specific heat at constant volume of solid helium-4 per mole at a molar volume of $20.96 \text{ cm}^3 \text{ mole}^{-1}$ plotted as C/T^3 as a function of T^4 . This is taken from Fig. 7 of Ref. 11. Some of the data points in the range below $T^4=0.1$ have not been included in this plot.

73 a phonon contribution or does it mean that there must be an extra effect
 74 associated with superfluidity?

75 For a dielectric solid at very low temperatures the specific heat per
 76 unit volume is given by the formula

$$77 \quad C_D(T) \approx \frac{2\pi^2 k^4 T^3}{5v_D^3 \hbar^3} = \frac{12\pi^4}{5} nk \left(\frac{T}{\Theta_D} \right)^3, \quad (1)$$

78 where n is the number of atoms per unit volume and v_D is the Debye
 79 velocity, defined as

$$80 \quad \frac{1}{v_D^3} = \left\langle \frac{1}{v_j(\theta, \phi)^3} \right\rangle, \quad (2)$$

81 with $v_j(\theta, \phi)$ being the velocity of the j -th phonon branch in the direction
 θ, ϕ . The Debye temperature is

82

$$\Theta_D = \left(\frac{6\pi^2}{V_{\text{atom}}} \right)^{1/3} \frac{\hbar v_D}{k}, \quad (3)$$

83 where V_{atom} is the volume per atom. The Debye temperature of solid
84 helium at the melting pressure and as $T \rightarrow 0$ is 26 K.¹¹ This corresponds
85 to a Debye velocity of $2.85 \times 10^4 \text{ cm s}^{-1}$.

86 Here, we investigate whether the effect of phonon dispersion results
87 in an appreciable deviation of the specific heat from the T^3 law in the
88 temperature range of interest, i.e., the range in which Gardner et al. have
89 noticed that there is an excess heat capacity that can be fit by a T^7 term.
90 This range extends from roughly 0.6 K to 1.2 K. At 1.2 K, for example, the
91 specific heat was found to be approximately 20% above the T^3 value.

92 The general expression for the phonon specific heat neglecting anhar-
93 monicity is

$$C(T) = \frac{\hbar^2}{kT^2} \int \frac{\omega^2 g(\omega) \exp(\beta \hbar \omega)}{[\exp(\beta \hbar \omega) - 1]^2} d\omega, \quad (4)$$

95 where $g(\omega)$ is the density of phonon states per unit volume and frequency.
96 The density of states can be calculated from the phonon dispersion curve
97 $\omega_j(\vec{q})$ if this is known for all wave vectors and polarizations. Unfortu-
98 nately, detailed measurements of the phonon dispersion in solid helium
99 have not been made and so it is not possible to determine $g(\omega)$ reliably
100 in this way.¹⁵

101 To get some idea of the corrections to the specific heat from dis-
102 persion consider first a simplified model in which the dispersion relation
103 is isotropic, and the transverse and longitudinal sound velocities are the
104 same and therefore equal to v_D . Suppose that the first correction to linear
105 dispersion is of the form¹⁶

$$\omega = v_D q (1 - \gamma_3 q^2 + \dots). \quad (5)$$

107 Then it is straightforward to show that the lowest order correction to
108 the specific heat is

$$C_3 = \frac{40\pi^4}{7} \gamma_3 \frac{k^6 T^5}{\hbar^5 v_D^5}. \quad (6)$$

110 Thus, the correction relative to the Debye specific heat C_D is

$$\frac{C_3}{C_D} = \frac{100\pi^2}{7} \gamma_3 \left(\frac{kT}{\hbar v_D} \right)^2. \quad (7)$$

111

112 Is this correction appreciable at 1 K? To answer this, an estimate of the
 113 parameter γ_3 is needed. We can get an approximate idea of the magnitude
 114 of γ_3 from the known values of this parameter in liquid helium. Very accu-
 115 rate measurements have been made by Rugar and Foster.¹⁶ They find val-
 116 ues that are of the order of 1 \AA^2 . The value is negative at low pressures
 117 but it is known from other experiments and analysis to become positive at
 118 higher pressures and to have a value of around 2.2 \AA^2 close to the freezing
 119 pressure.¹⁷ If we take 1 \AA^2 as a representative value we obtain the result

$$120 \quad \frac{C_3}{C_D} \approx 0.3T^2 \approx 200 \left(\frac{T}{\Theta_D} \right)^2. \quad (8)$$

121 Thus, based on this estimate at least, two things are clear. The magnitude
 122 of the correction to the specific heat due to non-linear phonon dispersion
 123 is significant at 1 K, i.e., of the order of 30%, and in the same gen-
 124 eral range as found by Gardner et al. Second, the correction to the spe-
 125 cific heat is much larger than the simple order of magnitude estimate of
 126 $(T/\Theta_D)^2$.

127 As a next step, we consider higher order terms in the dispersion.
 128 At first sight one might expect that the frequency ω would be a power
 129 series containing only odd powers of q . However, in a remarkable paper
 130 Kemoklidze and Pitaevski (KP)¹⁸ have shown that because of the long
 131 range part of the van der Waals force the series contains a q^4 term, i.e.,

$$132 \quad \omega = v_D q (1 - \gamma_3 q^2 - \gamma_4 q^3 + \dots). \quad (9)$$

133 They derive an explicit result for the coefficient γ_4

$$134 \quad \gamma_4 = \frac{\pi^2}{24} \frac{\rho \phi}{v_L^2 m^2}, \quad (10)$$

135 where ρ is the mass density, v_L is the longitudinal sound velocity and m
 136 is the atomic mass. ϕ is the coefficient of the van der Waals force at long
 137 distance, i.e., the potential energy associated with the van der Waals force
 138 between two atoms is $V(r) = -\phi/r^6$. As far as we can see, the KP effect
 139 should only apply to longitudinal phonons. Putting in values appropriate
 140 for solid helium at the melting pressure¹⁹ ($\rho = 0.1908 \text{ g cm}^{-3}$, $v_L = 4.88 \times$
 141 10^4 cm s^{-1} , and $\phi = 1.57 \times 10^{-60} \text{ erg cm}^6$) gives $\gamma_4 = 1.2 \text{ \AA}^3$. If this were the
 142 only correction term in the dispersion relation, i.e., if $\gamma_3 = \gamma_5 = \dots = 0$, then
 143 the lowest order correction to can be calculated to be

$$144 \quad C_4 = \frac{15, 120 \zeta(7)}{\pi^2} \gamma_4 \frac{k^7 T^6}{\hbar^6 v_L^6}. \quad (11)$$

145 To compare this with the T^3 specific heat, note that in an isotropic
146 approximation the Debye velocity is related to the longitudinal v_L and
147 transverse v_T sound velocities by the formula

$$148 \quad v_D^{-3} = (v_L^{-3} + 2v_T^{-3})/3. \quad (12)$$

149 Then we find that

$$150 \quad \frac{C_4}{C_1} = \frac{113,400\zeta(7)}{\pi^4} \gamma_4 \frac{1}{1 + 2v_L^3/v_T^3} \frac{k^3 T^3}{\hbar^3 v_L^3} = 1.6 \times 10^{-3} T^3. \quad (13)$$

151 Thus in the temperature range up to 1.2 K, this amounts to a small cor-
152 rection. Consequently we will not further consider effects arising from the
153 KP term γ_4 .

154 Now let us consider how a phonon contribution to the specific heat
155 that goes as T^7 might arise. There appear to be two possibilities. The first
156 is to propose that the corrections to the phonon dispersion relation are of
157 the form

$$158 \quad \omega = v_D q (1 - \gamma_3 q^2 - \gamma_5 q^4), \quad (14)$$

159 with negligible higher order terms, and with the coefficient γ_3 much
160 smaller than expected. If γ_3 is zero, it is straightforward to show that the
161 correction to the specific heat arising from γ_3 is C_5 where

$$162 \quad \frac{C_5}{C_1} = 112\pi^4 \gamma_5 \frac{k^4 T^4}{\hbar^4 v_D^4}. \quad (15)$$

163 To get a T^7 term of the magnitude found by Gardner *et al* would require
164 that $\gamma_5 = 2.0 \text{ \AA}^4$. This is not unreasonably large since it is of the order of
165 the square of the values of γ_3 that have been measured in liquid helium.

166 A second possibility that we consider as more likely is that the cubic
167 dispersion term γ_3 is positive for some propagation directions and polariza-
168 tions and negative for others. We note that calculations of the phonon
169 dispersion in solid helium give anomalous dispersion (i.e., negative γ_3) for
170 some directions and polarizations.²⁰ Then when considering all polariza-
171 tions and directions, the T^5 term could happen to be small. A T^7 term
172 could arise either from the γ_5 term already discussed or from the γ_3 term
173 in second order. For a single branch with velocity v_D , the correction to the
174 specific heat arising from the γ_3 term in second order is

$$175 \quad C_{3,2} = \frac{896\pi^6}{15} \gamma_3^2 \frac{k^8 T^7}{\hbar^7 v_D^7}. \quad (16)$$

176 The total correction coming from all polarizations and directions would
 177 then be given by the same expression but with an average value $\langle \gamma_3^2 \rangle$
 178 replacing γ_3^2 . To get agreement with the data of Gardner *et al* requires
 179 $\langle \gamma_3^2 \rangle^{1/2} = 0.7 \text{ \AA}^2$, which is a reasonable value.

180 We can also investigate the corrections to the T^3 law in a simple
 181 model crystal. Consider as an example a face-centered cubic crystal with
 182 central forces between nearest neighbors described by a spring constant K .
 183 Let the atomic mass be M . For this model the Debye temperature is ²¹

$$184 \quad \Theta_D = \frac{2.965\hbar}{k} \sqrt{\frac{K}{M}}. \quad (17)$$

185 A straightforward numerical calculation of the phonon density of states
 186 followed by an evaluation of the specific heat gives the result

$$187 \quad \frac{C}{C_D} = 1 + 60 \left(\frac{T}{\Theta_D} \right)^2 + \dots \quad (18)$$

188 Thus, again the correction to the specific heat is much larger than the
 189 rough estimate of $(T/\Theta_D)^2$. In Fig. 2 we show the specific heat of the fcc
 190 crystal as a function of T/Θ_D . The measurements of Gardner *et al.* cover

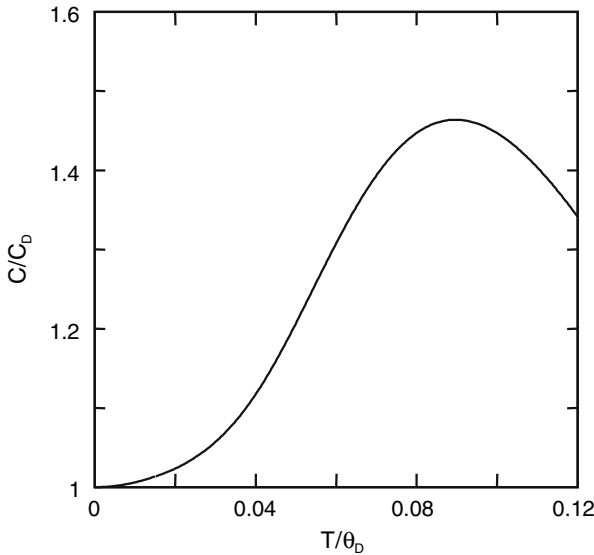


Fig. 2. Specific heat $C(T)$ of a fcc crystal plotted as $C(T)/C_D$ versus T/Θ_D . C_D is the low temperature Debye specific heat proportional to T^3 .

191 the temperature range up to $T/\Theta_D : 0.05$. It can be seen that for the fcc
192 model C/C_D varies by 20 % over this temperature range.

193

3. DISCUSSION

194 The above estimates show that one cannot conclude that the excess
195 specific heat seen by Gardner et al.¹¹ is evidence that there is a T^7 con-
196 tribution to the specific heat that results from the zero-point vacancies in
197 an incommensurate solid. Phonon dispersion should lead to a contribu-
198 tion to the specific heat that has a magnitude roughly the same as the T^7
199 term that has been found experimentally, but at the present time there is
200 no way to calculate the precise magnitude and temperature dependence of
201 this contribution. For example, while the simple model of the fcc crystal
202 that we have considered gives a correction to the phonon specific heat that
203 is positive, if the phonon dispersion is anomalous as it is in liquid helium
204 at low pressure,^{16,17} the correction will be negative. An accurate measure-
205 ment of the phonon dispersion curves by neutron scattering, for example,
206 would make it possible to estimate the phonon contribution.

207 Finally, we emphasize that in this paper we have addressed only the
208 phonon contribution to the specific heat. There may also be a significant
209 contribution from thermally- excited vacancies. Through X-ray studies,²²
210 the lattice parameter of solid helium has been measured as a function of
211 temperature for samples maintained under conditions of nearly constant
212 volume. The lattice parameter is found to decrease as the temperature is
213 raised and it is assumed that this decrease occurs because vacancies are
214 thermally excited. From the experimental data, an activation energy E_v for
215 vacancy formation can be found. It is then possible to use this value of
216 E_v to calculate the vacancy contribution C_{vac} to the specific heat. Surpris-
217 ingly, the predicted value of C_{vac} appears to be too large to be consis-
218 tent with the measured specific heat.²² One possible explanation for this
219 discrepancy is that the vacancies are not localized and that as a result
220 they form a band of energies.²³⁻²⁵ It is also possible that in fact the pho-
221 non specific heat is less than has been believed and that as a consequence
222 more of the specific heat that is experimentally measured arises from
223 vacancies.

224

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