Thermal expansion and magnetostriction of a nearly saturated $^3$He-$^4$He mixture


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(Received 10 November 2008; final version received 28 April 2009)

We have measured the molar volume of a nearly saturated $^3$He-$^4$He mixture, at temperatures below 100 mK and in magnetic fields to 16 T, by immersing capacitive dilatometers in the dilute phase of a top-loading dilution refrigerator. In zero magnetic field the coefficient of volume thermal expansion is negative and shows the linear temperature dependence expected of a Fermi liquid. Subject to experimental assumptions, we find the fractional increase in molar volume with magnetic field at 20 mK and 16 T to be about 0.01%.

Keywords: helium 3-4 mixtures; magnetostriction; thermal expansion; molar volume; Fermi liquid

1. Introduction

Though the physics of liquid $^3$He-$^4$He mixtures is quite rich (for reviews, see Edwards [1] and references therein), dilute mixtures below 100 mK have a relatively simple physical picture: The $^4$He component is almost completely in its superfluid ground state, essentially a ‘mechanical vacuum’ [2] through which the $^3$He atoms interact by exchanging virtual phonons. The $^3$He component is a weakly interacting Fermi gas whose Fermi energy can be tuned by varying the concentration $x = n_3/(n_3 + n_4)$ (where $n_3$ and $n_4$ are the densities of the $^3$He and $^4$He atoms, respectively), the low-energy excitations are quasiparticles with an effective mass $m^* = 2.4 m_3$. Though several theoretical frameworks for $^3$He-$^4$He mixtures can be found in the literature (see [1] and references therein), the approach by Bardeen, Baym and Pines (BBP) [3] is widely used and fits naturally into a discussion of molar volumes [4]. In BBP theory the magnitude of the interaction potential is given by $V_0 = -\alpha^2 m_4 s^2 / m_3$, where $s$ is the speed of first sound in pure $^4$He and $\alpha$, the BBP parameter, is the excess volume of a $^3$He atom in solution relative to that of a $^4$He atom. $\alpha$ is determined from

$$V_m(T, H, x) = V_4(T, H)[1 + \alpha(T, H, x)x]. \tag{1}$$

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where \( V_m(T, H, x) \) is the molar volume of the mixture and \( V_4(T, H) \) is the molar volume of pure \(^4\)He. The value of the BBP parameter in the \( T = H = x = 0 \) limit is about 0.285 [4,5].

The maximum solubility of \(^3\)He in \(^4\)He as \( T \to 0 \) is \( x_s = 6.6\% \), for higher concentrations phase separation will occur in this limit [2]. Since the work of BBP, the temperature-dependent molar volume of a mixture has been measured by several groups [4–7]. Along the phase separation curve the volume thermal expansion \( \beta = \partial(\ln V) / \partial T \) is positive, the molar volume itself tending towards a value of \( V_m(0,0,x_s) = 28.07 \text{ cm}^3/\text{mol} \) [7]. For concentrations below \( x_s \) the thermal expansion is negative indicating [7], as with pure \(^3\)He [8,9], that the effective mass \( m^* \) decreases more strongly with volume than \( V^{2/3} \). In this paper we report measurements of the thermal expansion of a nearly saturated \(^3\)He-\(^4\)He mixture below 100 mK.

The physics of spin-polarized quantum systems, such as dilute \(^3\)He-\(^4\)He mixtures in high magnetic fields, is also quite rich (for reviews see Owens-Bradley [10], Lee [11], Meyerovich [12], and references therein). Polarization effects in dilute mixtures were first observed in the second sound measurements of Greywall et al. [13]. As the polarization increases, the exchange interaction between quasiparticles with parallel nuclear spins produces phenomena that range from novel to spectacular. A spectacular example involves transport phenomena such as viscosity: in this case the exchange interaction suppresses s-wave scattering thereby increasing the mean free path. In the limit of 100\% polarization the mean free path, and hence the viscosity, is predicted to diverge [12]. Such divergent behavior has recently been observed in a dilute mixture by Akimoto et al. [14] who report an increase in viscosity by more than a factor of 500 as the mixture is ‘brute-force’ polarized near 1 mK.

Thermodynamic properties are affected in a less dramatic fashion. The specific heat of a fully polarized mixture, for example, is predicted to be smaller by a factor of 1.59 compared to the unpolarized mixture, the sign of this effect changes with increasing temperature [10]. In this paper we also describe the initial results of an experiment to measure the magnetostriction of a nearly saturated \(^3\)He-\(^4\)He mixture. In this case, the exchange interaction acts like a repulsive pseudo-force [15] to increase the molar volume, \( V_m \), with polarization \( \Delta \). Bedell [16] has shown that the increase in molar volume is proportional to \( \Delta^2 \).

2. Experimental details

We used a capacitive dilatometer originally designed for measurements on solids, a detailed description is given elsewhere [17]. When top-loaded into the dilute phase of a dilution refrigerator liquid helium fills the capacitor gap, its dielectric properties causing the capacitance to increase. The enormous thermal expansion of the helium liquids leads to a temperature-dependent dielectric constant that usually dwarfs the contribution of solids below 1 K. However, when the solid sample is removed from our dilatometer and replaced with an appropriate length of the metal of which the dilatometer is built (either OHFC Cu or commercially pure grade-2 Ti for the work presented here) the result is an adjustable parallel plate capacitor with edge effects and parasitic capacitances less than 1\%.
The gap of the ‘empty’ copper dilatometer is independent of temperature (below about 2 K to at least 300 mK) and magnetic fields at the sub-angstrom level. Measurements of the commercially pure (about 98% pure) titanium used in the construction of the titanium dilatometer are consistent with this temperature dependence but are also consistent with the presence of magnetic impurities that can be quenched by fields less than 0.1 T and with the presence of Pauli paramagnetism (G.M. Schmiedeshoff, unpublished), these effects will be discussed below. The gap \( D \) of the Ti dilatometer below 100 mK is roughly quadratic with field (nominally \( D(H) = D(0) - (0.365A/T^2)H^2 \) to 16 T) and independent of temperature, within experimental resolution, above about 40 mK (where the magnetostriction of the helium itself begins to affect the data, see below).

The capacitance of the dilatometer, nominally 30 pF, was measured with an Andeen–Hagerling capacitance bridge operating at 1 kHz. Our RuO thermometer was calibrated against the melting curve of \(^3\)He and, below 10 mK, against a nuclear orientation thermometer. A capacitance thermometer using kapton as its dielectric [18], calibrated against the RuO thermometer in zero field, was used as a transfer standard for temperature measurements in the field. An extremum in the temperature dependence of this thermometer determines the upper limit of its usefulness, for data taken with the titanium (copper) dilatometer, this upper limit was about 60 mK (100 mK).

Our ‘sample’ is the \(^3\)He–\(^4\)He mixture on the dilute side of the phase boundary in the mixing chamber of a top-loading dilution refrigerator. If this mixture was static the \(^3\)He concentration would be \( x_s \). Since the mixture in an operating dilution refrigerator is not static there must be a gradient in the concentration between the phase boundary and the dilatometer. We do not have an estimate of this gradient available, but we suspect it is small because the \(^3\)He quasiparticles are moving through a column of superfluid \(^4\)He; we therefore use the term ‘nearly saturated mixture’ to characterize our sample and we assume that its concentration is close to \( x_s \) [19].

The molar volume of the mixture, \( V_m \), is deduced from its dielectric constant, \( \varepsilon \), using the Clausius–Mosotti relation

\[
V_m = \frac{4}{3} \pi a \frac{\varepsilon + 2}{\varepsilon - 1},
\]

where \( a = 0.1232 \text{ cm}^3/\text{mol} \) is the molar polarizability which, within experimental uncertainty, is identical for \(^3\)He and \(^4\)He [20]. To determine \( \varepsilon \) from the capacitance measurement, the value of the ‘empty’ capacitor \( C_0 \) is required. We estimate this value by monitoring the capacitance as the dilatometer is top-loaded into the mixing chamber. For absolute measurements of the molar volume, one must correct for a pressure dependence to \( C_0 \) and to the density of the helium itself (both associated with the hydrostatic pressure of the helium column above the dilatometer) [9,21].

For the data presented below we did not apply these corrections, instead we adjusted the value of \( C_0 \) until the zero field data extrapolated to \( V_m(0,0,x) = 28.07 \text{ cm}^3/\text{mol} \) [7]. Given the sensitivity of \( C_0 \) to the mechanical shocks associated with top-loading, this adjustment is within experimental uncertainty (and is very close, in both sign and magnitude, to that observed when the dilatometer is immersed in the mixture). These adjustments to \( C_0 \), leading to changes in \( V_m \) less than 0.1 cm\(^3\)/mol, did not affect the temperature dependence of \( V_m \) (in other words, small changes in \( C_0 \) simply
shift the curve up or down). These shifts do not affect the isobaric coefficient of volume thermal expansion, \( \beta \), or the isothermal coefficient of volume magnetostriction, \( \omega \), that are evaluated from

\[
\beta = \frac{1}{V_m} \left. \frac{\partial V_m}{\partial T} \right|_{p,H}, \quad \text{and} \quad \omega = \frac{1}{V_m} \left. \frac{\partial V_m}{\partial H} \right|_{p,T}.
\]  

(3)

Operating a heater in the mixing chamber affects the capacitance of the dilatometer a few centimeters away. We suspect this is due to the counterflowing superfluid and normal components of the \(^4\)He in the presence of a heat source [22] and/or convective instabilities in the \(^3\)He affecting the local concentration. For example, taking temperature-dependent data (with the magnetic field fixed) by stabilizing the temperature above 100 mK and then ramping the heater to zero over several hours results in reproducible data that do not exhibit the expected \( T^2 \) behavior as \( T \rightarrow 0 \), whereas taking data by simply shutting the heaters off after the system is stabilized above 100 mK results in reproducible data (after a transient response) that do show the expected \( T^2 \) behavior as discussed below. All of the data used to determine the thermal expansion and magnetostriction of the mixture were taken using the later protocol where the rate of cooling decreases with decreasing temperature and is never greater than 2 mK/min. (Near 20 mK, the rate is about 0.2 mK/min.)

3. Results

The temperature dependence of the molar volume, at zero field and in a 4.0 T field, taken with both Cu and Ti dilatometers, is shown in Figure 1. The two zero field data sets are in reasonably good agreement, exhibiting the expected \( T^2 \) behavior of a

![Figure 1.](image_url)  

Figure 1. (Color online). The molar volume of a nearly saturated \(^3\)He-\(^4\)He mixture plotted against \( T^2 \). Data taken with both Ti and Cu dilatometers at zero field and in a 4.0 T field are shown (see text). The labels indicate the field at which the data were acquired and the dilatometer used.
Fermi liquid. The two 4.0 T data sets illustrate experimental challenges: Compared to the zero field data, the 4.0 T data taken using the Cu dilatometer exhibits a significant increase in noise. The 4.0 T data taken using the Ti dilatometer, though still varying like $T^2$, exhibit a significantly different slope.

The noise associated with this Cu dilatometer worsens as the field increases – by 16 T the noise completely dominates the temperature dependence (the noise in the Ti data is much more typical for this dilatometer design). We observed a field-dependent noise in one or two earlier assemblies of these copper dilatometers and attributed it to magnetic impurities in the non-copper assembly materials and/or some unidentified problem with the assembly itself. This dilatometer will be rebuilt; but the agreement of its zero field behavior with that of the Ti dilatometer demonstrates that, in this limit, the data reflect the behavior of the helium mixture under study. Despite the increased noise, the agreement between the temperature dependences of the 4.0 T data and the zero field data suggests that the magnetostriction of the mixture is less than our (compromised) experimental resolution with this dilatometer. This latter result is not surprising given the relatively large concentration and the temperature range of our measurements (in other words, the size of the magnetostriction for these concentrations, temperatures, and fields is expected to be rather small).

For reasons discussed above a magnetic contribution to the behavior of our Ti dilatometer is expected, both from magnetic impurities (that should be quenched by modest magnetic fields) and from the intrinsic Pauli paramagnetism of the Ti which is linear in field along with a small finite-temperature component that goes like $T^2$. We believe that both effects exist in our finite field Ti data: the dc offset due to the linear field dependence is removed from the 4.0 T Ti data in Figure 1 as a result of being plotted as $V_m(T) - V_m(0)$, while the small $T^2$ contribution of the Pauli paramagnetism manifests as a smaller slope for these data.

To remove the magnetic effects of our Ti dilatometer from the high field data we make the following assumptions (in addition to assumption (1) discussed above, that the concentration of our mixture is nearly $x_c$): (2) that the 4.0 T data do not show the effects of helium polarization (consistent with the results from our Cu dilatometer, and estimates described below), (3) that 4.0 T is sufficient to quench the effects of magnetic impurities (consistent with our measurements on commercially pure Ti described above), (4) that the $T^2$ component of the Pauli paramagnetism of the Ti is not field dependent (at and above 4.0 T, we do not have experimental confirmation of this assumption at this time), and (5) that the field dependence of the Pauli paramagnetism of the Ti does not depend upon temperature (consistent with measurements of the isothermal field dependence of the Ti dilatometer at and above 50 mK). We therefore shift our Ti dilatometer data, taken with $H > 4.0$ T to match the 4.0 T data at 50.0 mK (which incorporates the assumption (6) that the magnetostriction of the helium mixture is negligible at and above 50 mK, at least to 16 T). The results, shown in Figure 2, indicate a small increase of molar volume with field, an increase that is qualitatively consistent with that expected for the magnetostriction of the mixture in that it is small, positive, and increases in magnitude as the temperature falls.
4. Discussion

We first note that the concentration of our nearly saturated mixture appears to be relatively stable, both in the short term (as evidenced by the quadratic temperature dependence of the zero-field molar volume) and in the relatively long term (the zero-field measurements taken by the Ti and Cu dilatometers, that agree within experimental resolution, were made about six months apart in the same dilution refrigerator). Given the idiosyncratic nature of dilution refrigerator operation, this may be a useful result for further studies of nearly saturated mixtures.

The coefficient of volume thermal expansion of our nearly saturated \(^3\text{He}\)-\(^4\text{He}\) mixture at low temperatures, as determined from a linear fit to the \((T \leq 60\,\text{mK})\) zero field data shown in Figure 1, can be written as \(\beta/T = (-0.56 \pm 0.01)\,\text{K}^{-2}\). This value is quite a bit larger than estimates from tabulated molar volumes in [4] for concentrations in the vicinity of \(\chi_s\), however, these tabulated values do not quite reach the \(T \ll T_F\) limit so it is not clear that such estimates are suitable for comparison. We also note that our result is larger than that reported for liquid \(^3\text{He}\) (where \(\beta/T\) is about \(-0.17\,\text{K}^{-2}\) as \(T \to 0\) [9,21]), as one might expect for a liquid and its ‘gas phase’.

The data shown in Figure 2 indicate that the fractional increase in molar volume, \(\Delta V_m/V_m\), at 20 mK and 16 T is about 0.01%. Bedell [16] has shown that the volume magnetostriction of a normal Fermi liquid can be expressed as

\[
n(\Delta) = n(0) \left(1 - \frac{\Gamma}{1 + F_0} \frac{\Delta^2}{2}\right),
\]

where

\[
\Gamma = 1 + F_0^\dagger + a - \frac{1}{3} \left[ b_0 F_0^\dagger \right] = \frac{n}{E_F} \frac{\partial \Delta}{\partial n},
\]

\(n=1/V_m\) is the molar density, \(\Delta = (n^+ - n^-)/(n^+ + n^-)\) is the polarization, \(T_{sf} = (1 + F_0^\dagger)T_F\) is the spin fluctuation temperature, \(F_0, F_0^\dagger\) and \(F_0^\dagger\) are Landau...
parameters, $a$ characterizes the polarization dependence of the effective mass, $b_0$ characterizes the polarization dependence of the $\uparrow\uparrow$ interaction, and the other symbols have their usual interpretations. Since some of these quantities have not yet been measured, we adapt this result to an ideal Fermi gas by setting the Landau parameters, $a$, and $b_0$ equal to zero. In this limit $\Gamma = 2/3$ is the Grüneisen parameter of an ideal Fermi gas, and the fractional change in molar volume with polarization becomes

$$\frac{\Delta V_m}{V_m} = \frac{1}{3} \Delta^2.$$  \hfill (6)

The polarization at $T = 0$ and in low fields is given by

$$\Delta = \frac{3}{2} \frac{\mu H}{k_B T_F},$$  \hfill (7)

where $\mu$ is the magnetic moment of a $^3$He quasiparticle and the other symbols have their usual meanings (calculations of $\Delta$ at finite temperatures and fields in the quantum limit are discussed by Mullin and Miyake [23]). Using $\mu/k_B = \hbar \gamma / 2k_B = 0.778 \text{mK/T}$ and $T_F = 420 \text{mK}$, we find $\Delta = 4.5\%$ and $\Delta V_m/V_m = 0.07\%$ for a nearly saturated mixture at $T = 0$ and $H = 16 \text{T}$. Since $\Delta$ will continue to increase as the temperature falls, our experimental result of $0.01\%$ at $20 \text{mK}$ does not seem unreasonable. At $T = 0$ and $H = 4 \text{T}$, $\Delta V_m/V_m = 0.004\%$, so assumption (2) above seems reasonable near $20 \text{mK}$.

The uncertainty in our magnetostriction result is dominated by the assumptions we have made about the magnetic behavior of the Ti dilatometer. Nevertheless, we take the results embodied in Figure 2, including the signal-to-noise ratio of the Ti data (characteristic of this dilatometer design), as strong evidence that the magnetostriction of this mixture is large enough to clearly characterize with this experimental approach. This experiment will be improved by using the Cu dilatometer in high fields and by studying a static mixture, isolated from that of the dilution refrigerator.

5. Conclusions
We have measured the molar volume of a nearly saturated $^3$He-$^4$He mixture to temperatures of about 14 mK and in magnetic fields to about 16 T using capacitive dilatometers. Below 100 mK the thermal expansion is negative and proportional to $T$ as expected for a Fermi liquid, we find $\beta/T = (-0.56 \pm 0.01) \text{ K}^{-2}$. Subject to several assumptions about the magnetic behavior of our Ti dilatometer at low temperatures and in high magnetic fields, the fractional increase in molar volume, $\Delta V_m/V_m$, at 20 mK and 16 T is about 0.01%.

Acknowledgements
It is a pleasure to dedicate this work to James L. Smith on the occasion of his 65th birthday. The authors thank him for his advice, his integrity, and his company, often shared under the influence of fine cuisines and vintages: Cheers Jim! We also thank A. Kearns for her assistance.
during the early stages of this work. This work was supported by the National Science Foundation under DMR-0704406. The experiment was performed at the National High Magnetic Field Laboratory which is supported by NSF Cooperative Agreement No. DMR-0654118 and by the State of Florida.

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