T^{-3} Temperature Dependence and a Length Scale for the Thermal Boundary Resistance between a Saturated Dilute ^{3}He-^{4}He Solution and Sintered Silver

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(Received 1 August 1994)

We have measured the thermal boundary resistance between various configurations of sintered silver and a saturated ^{3}He-^{4}He solution between 8 and 150 mK. We observe that there is a length scale involved in the heat flow between sinter and liquid, which manifests itself as a thickness-dependent crossover from a $T^{-1.5}$ dependence of the resistance at high temperatures to a T^{-3} behavior at low temperatures. This allows us to extract the Kapitza boundary resistance which we find to vary as T^{-3}, as expected from simple theory, and not as T^{-2} as suggested by most recent measurements.

A knowledge of the behavior of the coupling between dilute ^{3}He-^{4}He solutions and sintered metal powders, especially silver, is of central importance to low-temperature physics, owing to the universal use of sinters in dilution refrigerators and for ultralow temperature experimentation in general. At the end of the 1970s it was assumed that the Kapitza resistance between dilute solutions and sinters varied as T^{-3}, being limited by phonons either via the acoustic mismatch or, more likely, via phonon-quasiparticle relaxation processes, see, for example, the review by Harrison [1]. Virtually all subsequent measurements have found the resistance to follow a T^{-2} relation at low temperatures, e.g., [2,3]. This has been a puzzle since there is no satisfactory theory which would suggest such a temperature dependence.

In this Letter we present the results of a series of measurements on the thermal boundary resistance between a range of sinters and saturated dilute mixture in the mixing chamber of a dilution refrigerator. We find that, whereas the microscopic surface area plays only a minor role in determining the resistance, at the higher temperatures the most important factor influencing the behavior is the bulk thickness of the sinter layer. Hence, we have been able to separate unambiguously the effect of the resistance of the liquid within the sinter from the actual interfacial boundary resistance, which we find to vary as T^{-3} as suggested by theory. The results have profound implications for the design of low-temperature experiments involving sinters and dilute solutions.

This work began as a very simple experiment to check the Kapitza resistance of a new silver powder. In the past we have used the nominally 700 Å diameter Ulvac powder, Vacuum Metallurgical Company [4]. Recently, a new source has appeared, Silbest C-8 from the Tokuriki Honten Company [5], prepared by a different process and much cheaper. Various sintering recipes and the resulting microscopic surface areas have already been investigated by Itoh et al. [6], but the Kapitza resistance has not been systematically studied.

The sinters are placed in the dilute phase in the mixing chamber of a dilution refrigerator described previously [7]. Two silver wires attached to each sinter are glued through the mixing chamber wall, and in vacuum, a heater is attached to one wire and a carbon resistance thermometer attached to the other. The resistance thermometers are modified 0.5 W 47 Ω Speer carbon resistors, with most of the original ceramic casing sanded off to improve thermal contact. The resulting 0.3 mm thick slab retained much of the copper leads imbedded in the carbon for making electrical contact. For thermal contact, the resistor slab, insulated by cigarette paper soaked in Stycast 1266 epoxy, was wrapped in a 0.1 mm silver foil, which was spot welded to the appropriate silver wire. The resistors had a value of around 300 Ω at room temperature and about 3 kΩ near 10 mK.

The measurement is very simple. The resistance thermometers on each sinter are calibrated against a vibrating wire resonator in the mixing chamber from 5 to 150 mK, from the dilute solution viscosity data of de Waele et al. [8]. For a measurement, heat is supplied to the heater on one sinter, the temperature of the mixing chamber is stabilized, and the Kapitza resistance is calculated from the (small) temperature rise relative to that of the unheated sinters. The procedure is repeated for each sinter in turn and then the mixing chamber temperature is readjusted for the next set of measurements. Since the temperatures of all the nonheated sinters are monitored, we can check that the measuring process causes no thermal gradients in the surrounding mixture.

In the first experiments, the sinters were made by our usual method. A 99.99% pure 8 mm by 42 mm 0.1 mm thick silver foil was spot welded along the two long sides to two 1 mm diameter 99.99% silver wires for the attachment of the resistance thermometer and heater. The foil was roughened and perforated to help key the sinter in low-pressure oxygen to improve the thermal conductivity, typically yielding a resistance ratio of 1000. A pad of compressed silver powder approximately 1 mm thick was then sintered to the foil, using the conventional procedure of dusting the foil with a small quantity of a larger size flakelike powder [9] which improves adhesion.

Initially we measured five sinters made in this way. Two of Ulvac powder and three of Silbest powder produced with various sintering temperatures and times. Surface areas were measured with the nitrogen Brunauer-Emmett-Teller method. The results showed resistances which fell on a curve with gradually increasing slope to lower temperatures with an approximate mean T^{-2} dependence. Surprisingly, all the sinters performed essentially identically, independent of age, type of powder, or microscopic surface area. Although we knew by then that the new powder was satisfactory, we made one further series of measurements of sinters with a range of thicknesses to see the influence on the behavior. The temperature dependences were broadly similar but the variation with thickness somewhat contradictory. Since the packing fraction is not easily controlled in our standard design, we decided to make a final series of measurements with much better controlled sinters.

We redesigned the silver substrate to place the spot welds outside the sinter region. To avoid any possibility of the sinter separating from the substrate, the thin silver foil was replaced by a 1 mm silver slab with sinter on one side only. We produced four sinters of approximately 10 mm by 10 mm centered on a 17 mm square silver slab. Silbest C-8 powder was pressed under 0.5 t for 15 min, then sintered for 30 min under 50 Torr of H_2 at 160°. The only difference between the sinters was the mass of powder used (and therefore the thickness). Masses chosen were 0.111, 0.2666, 0.4822, and 1.007 g yielding, respectively, thicknesses of 0.187, 0.389, 0.743, and 1.685 mm with a packing fraction averaging 56%. Surface areas per gram were constant to within 5% at 2 m^2/g for all sinters. Great care was taken to ensure that the fabrication process was identical for each specimen. For comparison, one further annealed silver slab was measured, identical in all respects to the others but having no sinter. The experimental arrangement is shown in Fig. 1.

The measured boundary resistance is shown in Fig. 2. (We should emphasize that this is the bare measured resistance for each sinter without any normalization.) First, not surprisingly, we see that the silver plate with no sinter shows a much higher boundary resistance than those with sinter. Second, as a result of the more careful uniform sintering treatment, we now clearly see the difference in boundary resistances for the different thicknesses of sinter. Most striking is the fact that at 150 mK the resistance for all four sintered pads is essentially the same. As the temperature falls, the resistance follows an approximate $T^{-1.5}$ variation until at around 50 mK the resistance for the thinnest sinter begins to rise faster to approach a T^{-3} power law. With falling temperature, successively thicker sinters peel off from the common $T^{-1.5}$ curve to follow a T^{-3} dependence.

This is clear evidence of a length scale. At 150 mK the sinters are only active over a depth less than the thickness of the thinnest sinter and thus all return the same resistance. At 50 mK, the active depth becomes greater
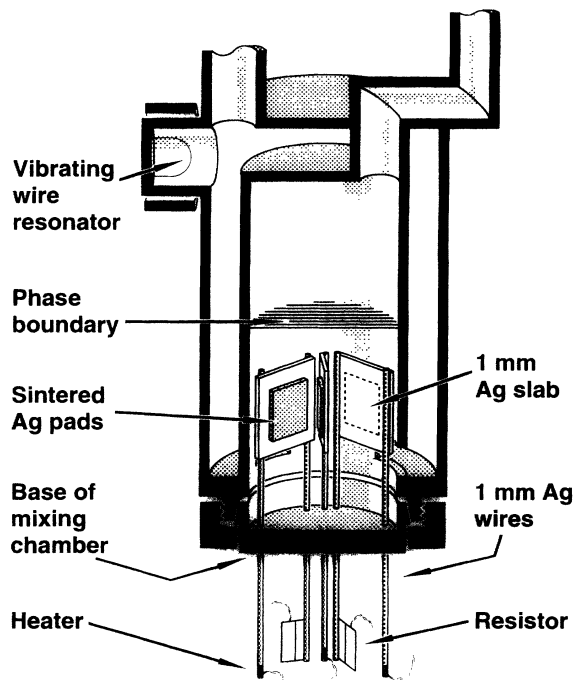

FIG. 1. The arrangement of the sintered silver pads plus silver support structure in the mixing chamber of the dilution refrigerator.

than the thickness of the thinnest sinter, and the resistance of this begins to rise relative to the others. By 15 mK even the thickest sinter is beginning to show the same rise, indicating that the active depth is now greater than 1.7 mm.
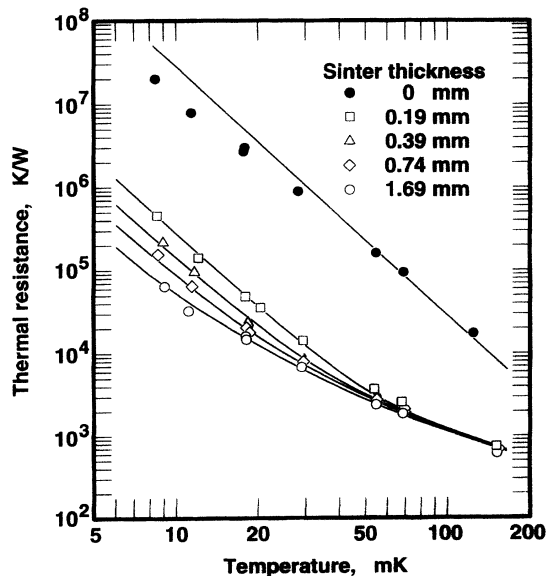

FIG. 2. The measured thermal boundary resistance of the series of sintered silver powder slabs of various thicknesses, along with the resistance of a bare plate, as a function of temperature.
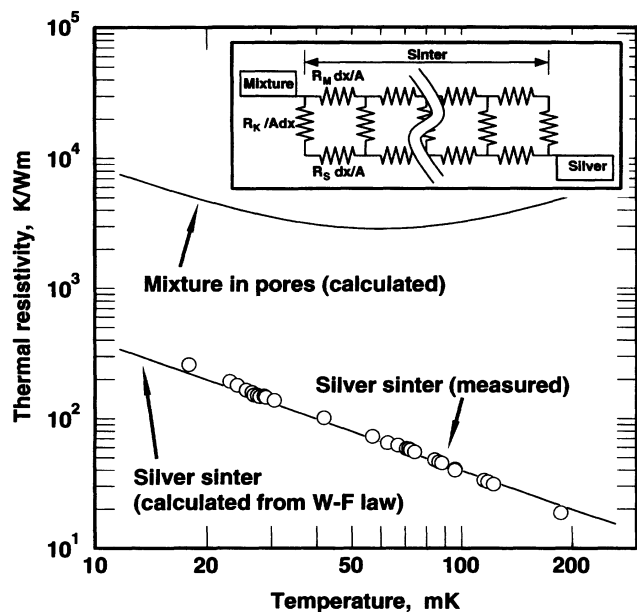
FIG. 3. The calculated thermal resistivity of a saturated dilute mixture confined within the pores of the sinter (see text), along with the measured and calculated resistivity of the silver sinter. Inset is the equivalent circuit used to model the thermal behavior.

The length scale in the sinter arises from competition between the boundary resistance and the thermal resistances within the silver and the mixture. When the thermal resistance is large, compared with the boundary resistance, only a small depth of sinter is active. The simplest model for the behavior of the sinter–dilute-solution composite comprises a ladder of resistance elements where each link of the chain corresponds to a depth dx of sinter of the cross-sectional area A, as shown in the inset of Fig. 3. The thermal resistance of the mixture in the pores is described by $R_M\,dx/A$, the resistance of the sinter by $R_S\,dx/A$ and the true volume boundary resistance between silver and the mixture by $R_K/A\,dx$. For simplicity let us anticipate the case where R_S is negligible. The ladder then reduces to a chain of "T" sections. For an infinite chain, the resistance is given by $Z = \sqrt{R_K R_M}/A$, the thermal current into the mixture decaying exponentially along the chain with characteristic length $\Lambda = \sqrt{R_K/R_M}$. For a sinter of finite thickness l, the full expression for the resistance is $Z = A^{-1}\sqrt{R_K R_M}\,\coth(l/\Lambda)$. However, we can immediately see the two extreme cases. At high-temperatures where Λ is less than the sinter thickness l, the resistance is equal to that for an infinite chain. At low-temperatures where R_K/R_M is large enough that $\Lambda \gg l$, then the resistance is simply given by the boundary resistance alone, R_K/Al. We know that at the higher temperatures, the temperature dependence is approximately $T^{-1.5}$, and at the low temperatures, the temperature dependence is T^{-3}. This would imply that R_K varies as T^{-3}, and R_M is approximately temperature independent.

We can find R_S directly by measuring the thermal resistance of a piece of sinter in vacuum, shown in Fig. 3, along with the curve calculated by the Wiedemann-Franz law from the measured 4.2 K electrical resistance. We must estimate R_M, which we do as follows. In bulk, there are contributions to the conductivity from ^{3}He quasiparticles, dominant below 10 mK, and from phonons, important in the 30–100 mK region. The conductivity of both components is quite high in the bulk, owing to the long mean free paths. However, within the sinter, mean free paths are restricted to the typical pore size. Consequently, at all temperatures of interest here, the phonon contribution is entirely negligible and we need consider only the quasiparticle contribution.

The quasiparticle mean free path in bulk, λ_b, varies as $1/T^2$ in the Fermi liquid regime. Hence, the conductivity K, proportional to the product of heat capacity, Fermi velocity, and mean free path $Cv_F\lambda$, varies as $1/T$. Measurements of the bulk conductivity show that $\lambda_b \sim 5\ \mu$m at 10 mK in the saturated solution. This is considerably larger than the sinter pore size. We estimate the effective helium conductivity by a simple model, in which we treat the sinter as a series of parallel channels with a range of submicron diameters. In a particular channel of size a, we take the effective mean free path to be given by $1/\lambda = 1/\lambda_b + 1/a$. At low temperatures this gives a channel conductance proportional to T, switching to the bulk $1/T$ at a temperature high enough that $\lambda_b \ll a$. We have modeled our sinter as a series of channels with a Gaussian distribution of pore sizes centered on 800 Å with a spread of 1200 Å, coupled with an obliquity factor which we estimate to be around 3 to take into account the tortuosity and possible noncommunication of the pores. The resistivity so calculated, shown in Fig. 3, turns out to be almost temperature independent over the range 30 to 100 mK, consistent with the present measurement. From the figure, it is clear that the resistivity of the metal may be neglected in comparison with that of the helium.

We now have all the information needed to fit the data. We take the thermal boundary resistance R_K to have T^{-3} dependence, and we assume that the thermal resistance of the solution R_M has the form calculated above. The fitted result is shown by the solid curves in Fig. 2. The parameters are fitted for the thinnest sinter and the other three curves are calculated using the same parameters and the appropriate sinter thickness. The fitted value of the Kapitza resistance for our sinters is found to be $5.6 \times 10^{-9}T^{-3}$ K m^3/W. (This figure corresponds to unit *volume* of the sinter-helium composite.)

There are two conclusions we can draw from these results. The first concerns the cryogenic application of sinters which are universally used for thermal contact to mixtures at low temperatures. It is clear from the figure that at 50 mK nothing is gained by making sinters any thicker than 0.1 mm. Not until 20 mK does a thickness of 1 mm become the optimum. At the lowest dilution refrigerator temperatures currently accessible,

these measurements suggest that a thickness of at least 4 mm is needed for optimum coupling.

Second, the true boundary resistance follows a T^{-3} temperature dependence. Radebaugh and Siegwarth [10] in an early set of measurements also saw a change from $T^{-1.5}$ to T^{-3} for sinters of small copper particles in various thicknesses, but as the results were analyzed in terms of the microscopic surface area, the length scale conclusion was missed. However, Harrison does hint at the significance in a comment in Ref. [1]. Fortuitously, all recent measurements of the boundary resistance to dilute mixtures have been made with sinter thickness such that the temperature range measured spanned the transition region giving an apparent T^{-2} behavior.

When the sinter thicknesses are properly taken into account, all reported results are consistent with the picture presented here. One convincing confirmation comes from the measurements of Osheroff and Corrucini [2], with a completely different sinter configuration and a very different temperature range (0.8 to 4 mK). The sinter was an annulus 25 mm high with outside diameter 25 mm and inside diameter 12 mm with embedded silver posts. We can compare these measurements with those of this work by assuming the same T^{-3} Kapitza resistance used to fit our data, simply scaled by the volume of the Osheroff and Corrucini sinter and a reasonable estimate of the high-temperature T^{-2} term expected from their geometry using our mixture conductance. (The high-temperature term goes as T^{-2} here, since we are in the T^{-1} region of the mixture thermal resistivity of Fig. 3.) If our geometrical assumptions are correct, then this is a no-free-parameters fit. On this basis we would predict a resistivity within a factor of 3 of the data but having the correct temperature dependence. The data in fact refer to a 8% ^{3}He-^{4}He mixture at 20 bar with a sinter of lower packing fraction (40%), which would yield even better agreement if allowed for. The data were originally taken as evidence of a T^{-2} dependent boundary resistance. However, for such a thick sinter, the transition from T^{-2} to T^{-3} on our model only occurs at around 2 mK, and the total resistance indeed varies as T^{-2} over a large part of the range.

We should comment briefly on the behavior of the plate with no sinter. This plate was measured after an extremely good anneal which gave large visible crystallites. Subsequent unnecessary handling was avoided. The effective area of the plate is about 5 times that of the macroscopic active surface area (10 mm × 10 mm) of the sinter pads. As can be seen in Fig. 2, at high temperatures the resistance follows a T^{-3} behavior which droops towards lower temperatures. The solid line in the figure represents the resistance calculated from acoustic mismatch theory [11], using values of the F parameters, $F_1 + F_2 = 1.54$. The agreement is surprisingly good at the higher temperatures, despite the fact that most measurements by other workers have yielded resistances substantially smaller than that predicted by the theory. The deviation at lower temperatures may arise from coupling to surface modes but is

more likely a result of a fraction of the heat generated in the heater being lost to the epoxy where the silver wires pass through the mixing chamber wall.

Knowing that the true boundary resistance follows a T^{-3} dependence, we may ask what is the mechanism. While it is hard to believe that acoustic mismatch is applicable to sinters when the bulk thermal phonon wavelengths in both the silver and helium are much greater than the pore size, for the parameters relevant to the sinters measured here, simple acoustic mismatch theory does in fact predict a resistance only a factor of 3 lower than the value of R_K we measure. If we follow Harrison and assume that the T^{-3} dependence arises from the ^{3}He quasiparticle-phonon coupling, which is enhanced in sinters as a result of the replacement of the bulk quasiparticle mean free path by the pore-size-limited value, then we calculate a value for R_K (again assuming our Gaussian distribution) which is also a factor of 3 lower than the measured value. Clearly more work on the concentration dependence would be worthwhile.

Apart from the obvious application to dilution refrigerators, the present measurement is a blow to those of us contemplating experiments on dilute solutions to the lowest possible temperatures, for example, in the search for superfluidity in the dilute ^{3}He component. That the boundary resistance varies as T^{-3} rather than T^{-2} means that the cosmic-ray background heating is unlikely to allow us to cool anywhere near the superfluid transition without recourse to underground experiments.

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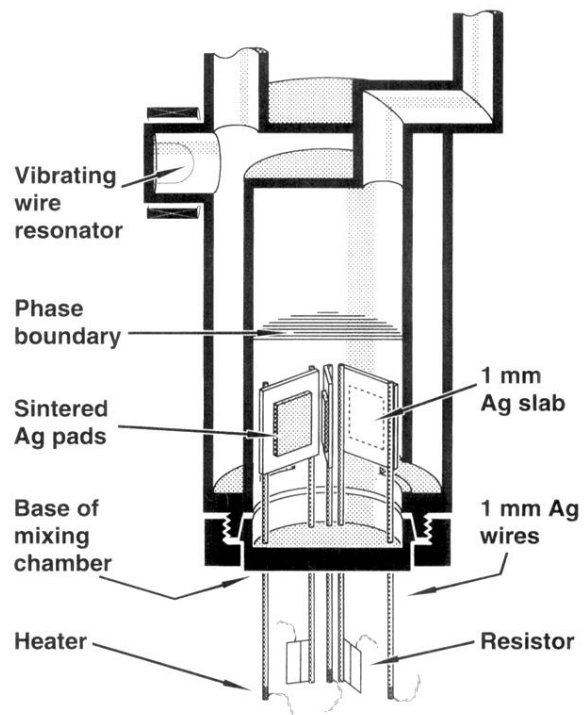
FIG. 1. The arrangement of the sintered silver pads plus silver support structure in the mixing chamber of the dilution refrigerator.