

H6 Heat transfer between sub-micron silver powder and dilute He³-He⁴ solutions

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The thermal resistance between copper cells lined with silver powder of 0.1–0.2 μm diameter and a dilute stream of He³ in He⁴ was measured between 15 and 250 mK. The results show that this powder can give an order of magnitude lower thermal resistance at 15 mK than an equal volume of copper powder of 1.8 μm diameter. The scientific and engineering significance of such a low resistance in a small volume is discussed. At the higher temperatures R is proportional to T^{-1} , which can be explained by a liquid thermal resistance and a resistance at the joint between the powder and the cell walls.

1 Introduction

Since the thermal boundary resistance (Kapitza resistance) between solid and liquid helium is roughly proportional to T^{-3} , it can be a severe problem in many experiments in the millikelvin range. In our laboratory we have been pursuing a two-fold approach to reduce the Kapitza resistance: finding materials which have a low Kapitza resistivity, $\rho_K = R_K \sigma$, (where R_K is the Kapitza resistance and σ is the surface area); and finding ways of increasing the surface area per unit volume without significantly increasing ρ_K . In this paper we discuss only the second approach.

The use of -325 mesh (44 μm) copper powder for enhanced heat transfer in the millikelvin range became very popular after Wheatley *et al.* [1] first used it in the heat exchangers of a dilution refrigerator. Originally it was believed that the use of powders finer than -325 mesh would not reduce $R_K V_l$ (where V_l is the liquid volume in the pores of the powder) because of the phonon-electron resistance [2–4]. Our measurements of the Kapitza resistance in 1.8 μm diameter copper powder showed this idea to be wrong [5]. We report here Kapitza resistance measurements on 0.15 μm diameter silver powder which show that $R_K V_l$ can still be reduced by an order of magnitude from that for the 1.8 μm copper powder.

2 Sample preparation

The silver powder used in this research was prepared by evaporating silver in a low pressure of argon gas, like that discussed by Kimoto *et al.* [6]. The particle size, as determined from electron micrographs, was about $0.15 \pm 0.05 \mu\text{m}$ which gives an approximate surface area per unit volume of metal $4 \times 10^5 \text{ cm}^{-1}$.

Three Kapitza resistance cells were prepared using this silver powder. The first cell was made by drilling a 2.71 mm hole

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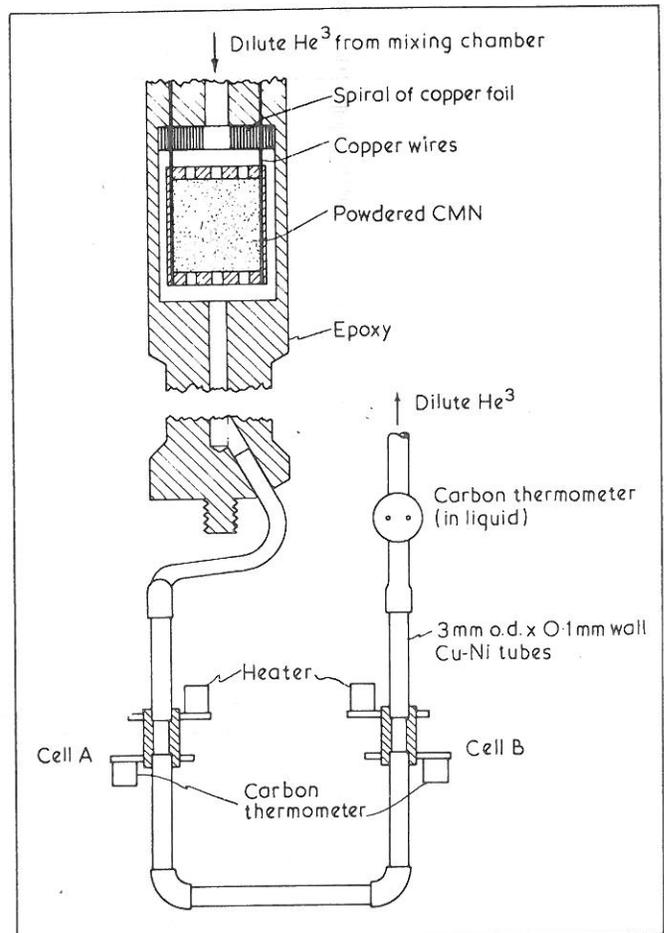


Fig.1 Experimental arrangement of the Kapitza resistance cells in the dilute stream of the dilution refrigerator. The copper foil is coated with copper powder and wound in a spiral with a plastic film between layers to prevent eddy-current heating. The copper wires extend to a gold-plated copper platform where a germanium thermometer is mounted.

through the centre of a 6.3 mm diameter OFHC copper rod 9 mm long. This was redrilled to 3.0 mm for a distance of 2 mm on each end so that 3 mm o.d. \times 2.8 mm i.d. Cu-Ni tubes could be inserted. The 2.71 mm hole was dusted lightly with 1.8 μm copper powder and the cell heated to 800°C in hydrogen for 10 min. The Cu-Ni tubes were soldered in place with a Cu-Ag eutectic during the same heating step. A Teflon plug with the end portion machined to a diameter of 2.32 mm was inserted in the cell, and silver powder was packed into the annulus between the Teflon and the copper cell with a packing fraction of 33.8%. The Teflon plug remained in place while the cell was sintered for 40 min at 150°C in a hydrogen atmosphere and its thermal expansion forced the silver powder to remain in contact

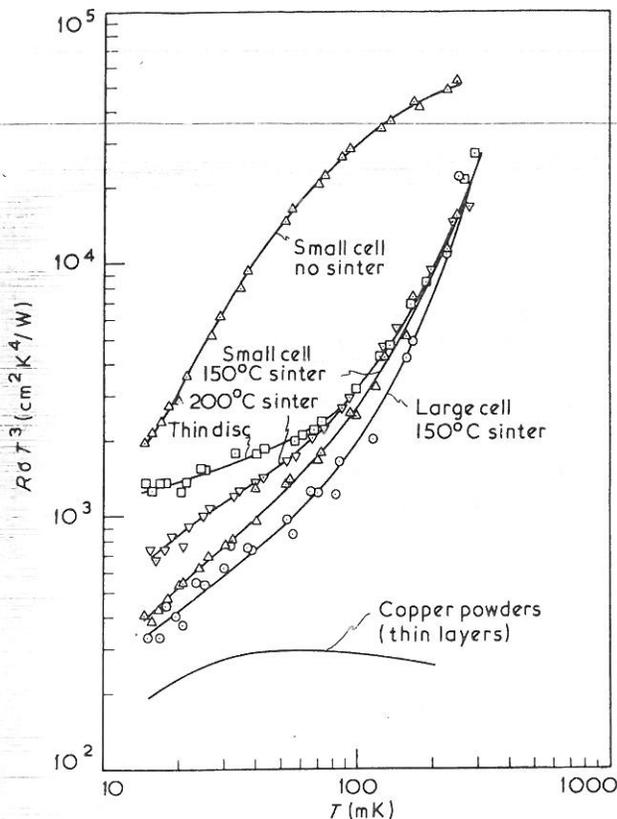


Fig.2 The behaviour of the thermal boundary resistivity between $0.15 \mu\text{m}$ silver powder and dilute $\text{He}^3\text{-He}^4$ as a function of temperature.

with the copper walls of the cell during the sintering process. (A disc of this pressed silver powder with a packing fraction of 37% shrank by 2% after similar sinterconditions.) The surface of this cell was calculated to be 1030 cm^2 with a liquid volume of 0.00505 cm^3 in the pores of the powder and 0.0211 cm^3 in the hole through the powder.

The second cell was prepared in the same way as the first except the diameters of the powder annulus were reduced to 1.59 mm o.d. and 1.15 cm i.d., and the Cu-Ni tubes were 2 mm o.d. instead of 3 mm. The packing fraction in this cell was 48.3% and the surface area estimated from the particle size was 915 cm^2 . The liquid volumes were 0.00244 cm^3 in the pores of powder and 0.00520 cm^3 in the hole.

The third cell was made by packing the powder in the shape of a thin disc or washer against the face of a OFHC copper rod with a 3.0 mm hole drilled through the centre. A 3 mm dia. Cu-Ni tube had been hard-soldered previously in the other end of the 3 mm hole. The powder was sintered at 150°C in hydrogen for 30 minutes. A cap with a 3 mm dia. Cu-Ni tube on one end was soldered with Wood's metal over the copper rod which had the thin layer of silver powder on the face. The geometry of the silver powder washer was 7 mm o.d. \times 3 mm i.d. \times 0.045 mm thick with a packing fraction of 46%. The estimated surface area was 259 cm^2 with liquid volumes of 0.00076 cm^3 in the pores and 0.12 cm^3 in the cell.

3 Experimental procedure

The Kapitza resistance cells were put in series into the dilute side of our dilution refrigerator between a CMN thermo-

meter and the lowest heat exchanger by soldering to the ends of the Cu-Ni tubes. Figure 1 shows the position of the Kapitza cells in the dilution refrigerator. A heater and carbon thermometer were attached to each cell in the following manner. The heater and thermometer elements were each glued on separate gold-plated copper holders. These holders were then bolted to separate copper clamps which were clamped around the Kapitza cells. This arrangement allows the heaters and thermometers to be used over again for each new Kapitza cell. Another carbon thermometer was placed in the dilute liquid downstream from the second cell to measure the change in stream temperature during heating. Details of the method for determining the Kapitza resistance, which includes necessary corrections, have been published [4].

4 Results and discussion

Figure 2 shows the behaviour of the product $R\sigma T^3$ as a function of temperature for the three cells of silver powder. In this case R is the total thermal resistance between the cell and a nearly saturated $\text{He}^3\text{-He}^4$ solution, and σ is the surface area. The highest curve is for the smaller cell before it was sintered (it had been kept for 30 hours at room temperature). After this cell was sintered at 150°C for 30 min in hydrogen, its $R\sigma T^3$ values agreed well with those for the larger cell sintered under the same conditions. Therefore, most of the thermal resistance of the unsintered cell must be in the silver powder or at the junction between the powder and the copper cell. This resistance may still dominate the Kapitza resistance after the 150°C sinter. Sintering the small cell at 200°C for 45 minutes in hydrogen (Teflon plug inserted) increased the Kapitza resistance and suggests that surface area decreases with such sintering conditions. The somewhat high value of $R\sigma T^3$ for the thin disc of silver powder is not understood at present. Results on the Kapitza resistance of bulk 99.9% silver indicate a roughly constant value of $R\sigma T^3$ which varies from about $16 \text{ cm}^2 \text{ K}^4/\text{W}$ to $55 \text{ cm}^2 \text{ K}^4/\text{W}$ with different surface conditions. One measurement on silver powder of about $6 \mu\text{m}$ diameter yielded about $115 \text{ cm}^2 \text{ K}^4/\text{W}$ for $R\sigma T^3$.

The $R\sigma T^3$ behaviour of thin layers of copper powder [5] are shown in Fig.2 for comparison. Thick layers of copper

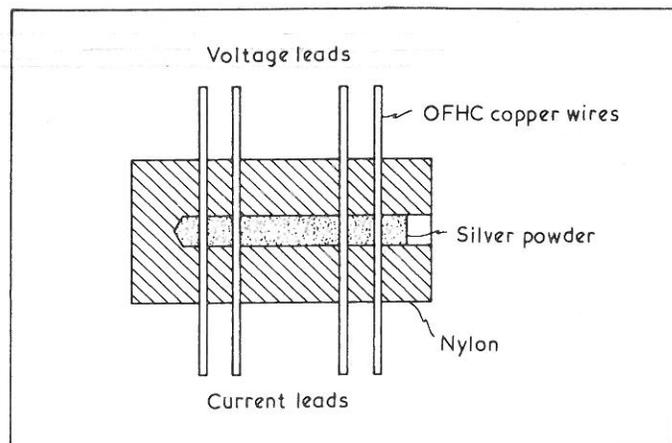


Fig.3 The arrangement used to measure the electrical resistivity of the silver powder and of the joints between the powder and OFHC copper wires.

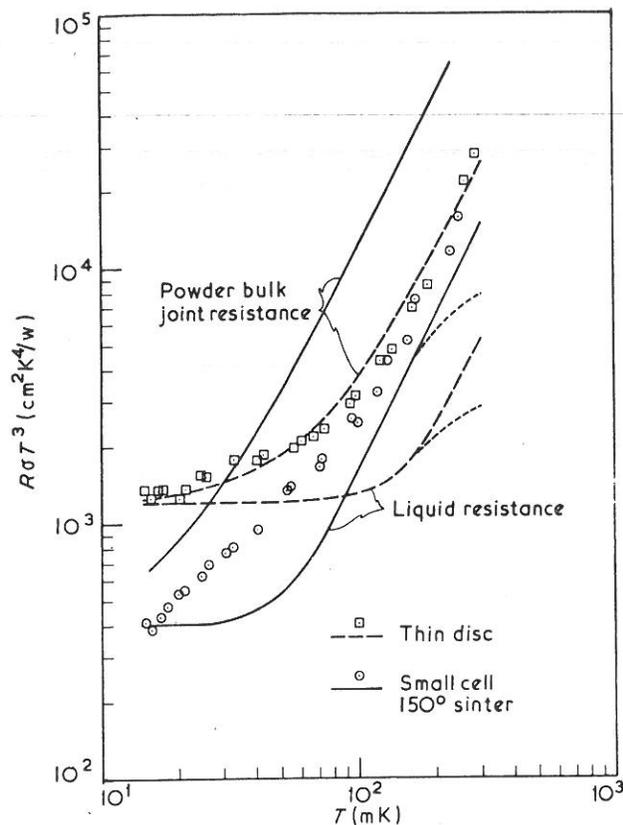


Fig.4 A comparison between measured and calculated values of the thermal resistance including resistance at the joint between the silver powder and the bulk copper cell, and resistance through the liquid in the pores of the powder.

powder also show the rapid increase of $R\sigma T^3$ with increasing temperature. Such an increase in $R\sigma T^3$ at higher temperatures previously was attributed to the thermal resistance of the liquid in the pores [5], but after a more accurate calculation we now feel that such an effect cannot account for all of the increase. Some of the increase must be due to resistance in the powder or at the junction between the powder and the copper walls. Such a resistance would behave as $R \propto T^{-1}$, which approximates the behaviour actually seen.

The electrical resistances of a packed rod of silver powder and of the joint between the powder and embedded OFHC copper wires were measured to determine their thermal resistances via the Wiedemann-Franz law. Figure 3 shows the arrangement used to measure these resistances. The resistivity of the powder decreased with time at room temperature after packing of the powder. After 100 hours the resistivity had decreased by 12% from the value of $30.5 \mu\Omega$ cm obtained immediately after packing. Such a decrease indicates that some sintering occurs even at room temperature. After sintering at 150°C for 30 min in hydrogen, the resistivity of the silver powder dropped to $16.8 \mu\Omega$ cm. The ratio of room temperature to liquid helium temperature resistance was 3.0 before sintering and 4.1 after sintering. The thermal conductivity coefficient, κ/T , as deduced from the electrical resistivities, was $2.9 \text{ mW cm}^{-1} \text{ K}^{-2}$ before sintering and $6.0 \text{ mW cm}^{-1} \text{ K}^{-2}$ after the 150°C sinter. These figures are over an order of magnitude too high to account for the large increase of $R\sigma T^3$ with temperature. The electrical resistance at the contact between the powder

and the copper wires (contact area A) yielded a thermal resistivity coefficient, $R_C A T$, of about $320 \text{ cm}^2 \text{ K}^2/\text{W}$ after the 150°C sinter both for wires precoated with $1.8 \mu\text{m}$ copper powder and for uncoated wires. This number is not very accurate since the joint resistance was quite susceptible to mechanical disturbances of the copper wires, particularly before the sintering process. The total thermal resistance R in the Kapitza cells would be the sum of the actual Kapitza resistance and the contact resistance R_C of the joint. Figure 4 shows the calculated behaviour of $R\sigma T^3$ for the 150°C sinter of the small cell and the thin disc sample. In the calculations we have taken $R_K \sigma T^3 = 400 \text{ cm}^2 \text{ K}^4/\text{W}$ for the small cell and $R_K \sigma T^3 = 1200 \text{ cm}^2 \text{ K}^4/\text{W}$ for the thin disc. The agreement between the experimental points and the calculated curves is certainly within the uncertainty of R_C .

Also shown in Fig.4 is the calculated behaviour of the total thermal resistance if only a liquid resistance is added to the Kapitza resistance. With a thermal resistance through the liquid in the pores of the powder, the total resistance will be given by

$$R\sigma T^3 = R_K \sigma T^3 [mL \tanh(mL)], \quad (1)$$

where L is the powder depth and

$$m = [(\sigma/V_l)T^3/(R_K \sigma T^3 \kappa_l)]^{1/2}. \quad (2)$$

The liquid thermal conductivity κ_l in the fine pores is determined mainly by the He^3 quasiparticle contribution, which $\kappa_l \approx 3 \times 10^{-6} \text{ T}^{-1} \text{ W/cm K}$. If the phonons could absorb heat in the pores, then the curves would be lowered to that of the dotted lines due to phonon conduction. As the figure shows, the liquid resistance cannot account for all of the observed increase in $R\sigma T^3$ at higher temperatures.

The low value of $R V_l T^3$ is of considerable importance both from a scientific and an engineering standpoint. At 15 mK, $R V_l T^3 = 1.7 \times 10^{-3} \text{ cm}^3 \text{ K}^4/\text{W}$, which is about an order of magnitude less than that observed for the $1.8 \mu\text{m}$ copper powder [5]. It also is more than an order of magnitude less than one might expect due to the phonon- He^3 quasiparticle resistance in the liquid [5] and slightly less than that caused by the phonon-electron resistance in the metal [7]. This is then further evidence that phonons are not responsible for the major part of the heat transfer at temperatures below about 30 mK. Heat transfer by a direct interacting between conduction electron spins and the He^3 quasiparticle spins had been proposed earlier [5, 8] but recent calculations [9] show this to be small. An interaction between electrons and He^3 quasiparticles via paramagnetic local moments [9-11] might explain the large heat transfer possible with this fine powder.

From an engineering standpoint the low value of $R V_l T^3$ means that much smaller liquid volumes can be used to provide good thermal contact between a solid and dilute He^3 - He^4 . Good thermal contact is required for thermometry, for cooling of samples, and for heat exchangers of a dilution refrigerator.

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