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Proximity Effect in Cu-Nb-Sn Composites

by

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Shelly Sit-Ling Shum Eucker

An Abstract of

A Dissertation Submitted to the
Graduate Faculty in Partial Fulfillment of the
Requirements for the Degree of
DOCTOR OF PHILOSOPHY

Approved:

In Charge of Major Work

For the Major Department

For the Graduate College

Iowa State University
Ames, Iowa

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Shelly Sit-Ling Shum Eucker

Under the supervision of D. K. Finnemore
From the Department of Physics
Iowa State University

Heat capacities have been measured in the presence of magnetic fields for Cu-Nb and Cu-Nb₃Sn in situ composites in order to determine the size of the superconducting pair potential which is induced in the normal Cu region as a function of magnetic field. For the Cu-Nb composites the induced pair potential is found to be about 0.35 meV in zero field, in agreement with de Gennes boundary conditions. With the application of a magnetic field, the energy gap declines quickly and above about 17 mT the Cu behaves as a gapless superconductor. A family of life-time broadened density of states which gives the measured C_{es} (Cu) is calculated based on the relation between the entropy S and density of states $N(\omega)$ derived for superconductors doped with paramagnetic impurities. Similar results are found in Cu-Nb₃Sn composites.

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I. INTRODUCTION

A. History of Superconducting Wires

Since the discovery of superconductivity in 1911 by Kamerlingh Onnes (1), a great deal of work has been done on homogeneous materials and there is a good understanding of the basic phenomena involved for both type I and type II superconductors. These homogeneous materials by themselves, however, are not well-suited for most practical applications such as superconducting solenoid magnets. In type I materials the difficulty arises because the superconductivity is "quenched," or suppressed, on exposure to relatively low magnetic fields. The value of the critical field, H_c , is usually only a few hundred gauss.

Type II superconductors are somewhat better in that they can have a very high upper critical field value H_{c2} , on the order of a few hundred kilogauss. Pure type II materials, however, are not well-suited for constructing magnets because there is dissipation of heat in a high magnetic field arising from the motion of quantized vortices. This heating effect arises because magnetic field can penetrate the sample in the form of quantized flux lines and these move in the presence of a transport current under the influence of the Lorentz force,

$$\vec{F} = \vec{J} \times \vec{B} \quad (1)$$

where $\vec{J}$ is the current density vector and $\vec{B}$ is the magnetic field vector. These moving vortices in turn cause time varying magnetic fields which by Faraday's law give electric fields,

$$\nabla \times \vec{E} = - \frac{1}{c} \frac{\partial \vec{B}}{\partial t} \quad (2)$$

In terms of the induced emf ϵ , this becomes

$$\epsilon = - \frac{d\phi_B}{dt} \quad (3)$$

where ϕ_B is the magnetic flux. This induced emf thus causes heating, of magnitude

$$P = \vec{J} \cdot \vec{E} \quad (4)$$

Moreover, as a superconducting wire is wound into a solenoid, good thermal environments do not always occur. If local heating due to flux penetration does not get carried away, it will promote further motion of flux through the material, with the dissipation of still more heat. Through a runaway process the superconductor may eventually revert to its normal resistive state. If the current supply is not turned off, the material might even melt locally.

These problems were encountered in the first few magnets to be constructed (2). It was found that the superconducting

solenoid could pass much less current than the short-sample critical current. To solve these problems, the superconductors were clad with high-purity copper to make a composite. Because of the high thermal and electrical conductivity of copper at such low temperatures, local hot spots can be smoothed out and in emergencies the current can be shunted without dissipating too much power. With this configuration it was found that the coils could be operated at a substantially higher current.

In view of the central role composites play in useful conductors, it is imperative to study these inhomogeneous materials for a better understanding of their properties and the basic physics involved. De Gennes (3) did the first theoretical studies on the boundary conditions and proximity effects in normal-superconducting layered structures, and many experiments have been done on thin film model systems to establish the physical principles involved (4 and references therein). These thin film experiments, however, were done with materials such as Pb-Cd and Pb-Cu which may differ from the materials used in practical magnets such as Cu-Nb₃Sn and Cu-NbTi. Also the boundaries or interfaces for practical superconductors such as Nb₃Sn are difficult to prepare in thin film depositions because there may be oxidation and impurity introduced at the boundary when the sample is prepared. So far very little work has been done on the

proximity effect in bulk wire composites which are used directly in superconducting magnets.

Since Tsuei (5) first suggested that Cu-Nb₃Sn superconducting wire composites could be made from castings of either Cu-Nb or Cu-Nb-Sn alloys, many techniques such as levitation melting by Harbison and Bevk (6), a graphite mold casting technique by Roberge and co-workers (7a,b), and the chill-casting (8) and consumable arc-casting (9) techniques developed here at the Ames Laboratory, have been developed for fabrication of these in situ composites. They are composed of very long, thin superconducting Nb or Nb₃Sn filaments aligned parallel to one another in a Cu matrix. These wire composites are excellent samples for the studies of boundary conditions and proximity coupling between filaments because they have clean and well-defined boundaries, and the filament spacing is such that it is comparable to the decay length of the pair potential in Cu, that is, the distance over which superconductivity can be induced in a normal metal.

The purpose of the work reported here is to study the proximity effect in the Cu-Nb and Cu-Nb₃Sn in situ composites fabricated by the consumable arc-casting technique. More specifically, the goal is to measure the magnitude of the superconducting pair potential induced in the Cu and to determine how this pair potential changes with magnetic field. Specific heat is used as the measuring probe so that bulk

samples of practical materials can be used, as will be discussed further later.

Before the experimental work of this study is discussed, some elementary properties of homogeneous superconductors which relate to this work will be reviewed and the theories of proximity effect will be discussed.

B. Superconductivity in a Homogeneous Medium

1. The pair potential (Δ)

The first evidence for an energy gap existing in superconductors came from specific heat studies, as reviewed in reference 10. It was observed that at very low temperature where $T \ll T_c$, T_c is the critical temperature of the superconductor, the electronic specific heat in the superconducting state could be expressed in the form

$$C_{es} = A e^{-BT_c/T} \quad (5)$$

where A and B are some constants. Here the interpretation is that the C_{es} temperature dependence arises from the thermal excitation of electrons across a gap. This general form was put on a sound theoretical by the BCS theory (11) which gives $C_{es} \approx 8.5 \gamma T_c e^{-1.44T_c/T}$ where γ is the electronic specific heat coefficient. About the same time as the C_p measurements, infrared absorption measurements by Richards and Tinkham (12) gave the first direct measurement of the gap. It was

also confirmed later by electron tunneling measurements (13, 14).

These experiments illustrate a fundamental difference between a superconductor and a normal metal. To create an excitation in a superconductor, the minimum energy required at the Fermi surface is not zero as in a normal metal, but has a finite value Δ . So for the creation of an electron-hole pair excitation, a minimum energy of 2Δ is needed.

This pair potential Δ is temperature dependent (15), as illustrated in Fig. 1. Its value is a maximum at $T = 0$ and decreases to zero at the transition temperature T_c in zero magnetic field. Above T_c the superconductor becomes normal. At $T = 0$, Δ is given by the BCS theory (11) as

$$\Delta(0) = 1.76 kT_c \quad (6)$$

A special feature of the BCS theory is that this energy gap is just equal to the binding energy per pair. As will be discussed later, in superconductors with life-time broadening of the Cooper pairs, the energy gap (Ω) or break in the density of states is no longer equal to the pair potential or binding energy per pair.

2. Calorimetric measurements

Both the photon absorption and electron tunneling are excellent techniques for direct measurements of the pair potential Δ . In the photon absorption process two excitations

are produced (electron-hole pair), so the onset of photon absorption gives the value of 2Δ , as shown in Fig. 2. In electron tunneling no current will flow through the metal-insulator-metal (NIS) junction until the bias voltage has shifted the Fermi level by an amount Δ . The I-V characteristic curve is shown in Fig. 3. Despite the fact that these two methods give direct measurements of Δ , they cannot be used for this work because they only work for samples in the form of thin film. Specific heat measurements, on the other hand, do not give the value of Δ directly, but they do work on any kind of samples and Δ can be deduced from the low temperature data (Eq. 5). The calorimetric method is thus used as the measuring probe.

The normal state specific heat C_N for a normal metal at a temperature much below the Debye temperature can be written in the Debye-Sommerfeld approximation as the sum of electronic and lattice contributions:

$$C_N = C_{eN} + C_{gN} = \gamma T + \alpha T^3 \quad (7)$$

where $C_{eN} = \gamma T$ is the normal electronic part, the coefficient $\gamma = \frac{1}{3} \pi^2 N(\epsilon_F) k^2$ where $N(\epsilon_F)$ is the density of state at the Fermi surface and k is the Boltzmann constant. $C_{gN} = \alpha T^3$ is lattice specific heat, the coefficient $\alpha = 234 N_O k / \theta_D^3$ where N_O is the Avogadro's number and θ_D is the Debye temperature.

The superconducting state specific heat C_s for a superconductor is also assumed to have the form

$$C_s = C_{es} + C_{gs} \quad (8)$$

The lattice specific heat C_{gs} , however, is assumed to be the same in both normal and superconducting states. This is supported experimentally by the absence of any observable changes in the lattice parameters in the transitions (16, 17). Although an exception has been claimed in the case of indium (18, 19), no adequate theoretical explanation of this anomaly has been made.

From the assumption that

$$C_{gs} = C_{gN} \quad (9)$$

C_{es} can then be readily obtained experimentally from

$$C_{es} = C_s - (C_N - \gamma T) \quad (10)$$

and to the extent that the data fit the expression in Eq. 5, the energy gap can be deduced.

C. Proximity Effect Theories

A proximity effect arises if electrons have free movement back and forth across the boundary of a normal metal in good electrical contact with a superconductor. In this situation,

Cooper pairs can move freely from the superconductor to the normal metal region and the normal electrons can also spread into the superconductor, tending to make superconductivity weaker near the contact. Hence in this inhomogeneous medium, the pair potential is no longer constant in space and a pair potential is induced in the normal region, as will be discussed here.

The theory of the proximity effect is rather complex and a full description is beyond the scope of this work. In what follows it will be very useful, however, to know the results of the theory, so a basic outline of the theory and the results are given without proof. Further details are given in the literature.

1. Order parameter

Within the BCS theory (11) superconductivity arises because electrons condense into a many electron ground state in which the electron momentum states occur in pairs rather than the random occupation which occurs in normal metal. This transformation from random to pair occupation occurs because the virtual exchange of phonons is an attractive interaction which gives an overall energy of the system which is lower for pair occupation than for random occupation. The matrix element $M_{kk'}$, for the effective electronic interaction parameter $V_{kk'}$, between an electron with momentum k and one with momentum k' is of the form

$$M_{kk'} = \int e^{-i\vec{k}' \cdot \vec{\rho}} H_I e^{i\vec{k} \cdot \vec{\rho}} d\tau \quad (11)$$

where $\vec{\rho} = \vec{r}_1 - \vec{r}_2$ is the relative coordinate between the two electrons 1 and 2, and H_I is the interaction Hamiltonian which scatters a pair of electrons from $(k, -k)$ to $(k', -k')$. To simplify the theory so that one can show the basic ideas of superconducting electron-electron interaction, $V_{kk'}$ is approximated as (Fig. 4)

$$\begin{aligned} V_{kk'} &= \text{constant} = -V \text{ (attractive) for } \epsilon_k < \hbar \omega_c \\ V_{kk'} &= 0 \text{ for } \epsilon_k > \hbar \omega_c \end{aligned} \quad (12)$$

where $\hbar \omega_c$ is some cutoff energy related to the Debye energy. The true electron-electron interaction is replaced by this simplified model to facilitate the calculation.

Morel and Anderson (20) show that V can be regarded as a point interaction, and the electrons are coupled by

$$-V(\vec{r}_i) \delta(\vec{r}_i - \vec{r}_j)$$

In other words, the interaction is short range in space but long range in time, on the order of 10^{-12} sec, the time for an ion to respond. The equation of motion for proximity effect theory can be written as (3)

$$i \frac{\partial \psi_{\alpha}^{+}(\vec{r})}{\partial t} = \left[\frac{p^2}{2m} + u(\vec{r}) \right] \psi_{\alpha}(\vec{r}) - v(\vec{r}) \sum_{\beta} \psi_{\alpha}^{+}(\vec{r}) \psi_{\beta}^{+}(\vec{r}) \psi_{\beta}(\vec{r}) \quad (13)$$

where $\psi_{\alpha}^{+}(\vec{r})$ is the one-electron operator, α and β are the spin indices, and $u(\vec{r})$ is the one-electron potential where the effects of impurities and boundaries are included.

Following the spirit of mean field theory, the last term of the equation can be linearized to

$$v_N \psi_{\alpha}^{+}(\vec{r}) \psi_{\beta}^{+}(\vec{r}) \psi_{\beta}(\vec{r}) \rightarrow v_N \langle \psi_{\alpha}^{+}(\vec{r}) \psi_{\beta}^{+}(\vec{r}) \rangle \psi_{\beta}(\vec{r}) \quad (14)$$

where $\langle \rangle$ denotes a thermal average. The order parameter is thus defined as

$$\Delta(\vec{r}) \equiv v(\vec{r}) \langle \psi_{\alpha}(\vec{r}) \psi_{\beta}(\vec{r}) \rangle \quad (15)$$

where $\Delta(\vec{r})$ is the pair potential and $F(\vec{r}) \equiv \langle \psi_{\alpha}(\vec{r}) \psi_{\beta}(\vec{r}) \rangle$ is the probability amplitude of finding two electrons in the condensed state at point $\vec{r}$. $\Delta(\vec{r})$ is essentially a measure of the ordering in superconductors. In homogeneous superconductors, $\Delta(\vec{r})$ is just the BCS energy gap Δ at the Fermi surface. For an inhomogeneous medium, the pair potential is a function of position.

In the dirty limit and at a temperature near the transition temperature of the normal-superconducting sandwich (NS junction), $\Delta(\vec{r})$ is small at all points in space and, as

shown by Gor'kov (21), this leads to a linear integral equation for $\Delta(\vec{r})$ (for derivation see Ref. 22):

$$\Delta(\vec{r}) = V(\vec{r}) \sum_{\omega} \int d^3\vec{r}' \Delta(\vec{r}') H_{\omega}(\vec{r}, \vec{r}') \quad (16)$$

Hence, pairing at a point $\vec{r}$ is related to that at all other points $\vec{r}'$. The range over which this is true is governed by the kernel $H_{\omega}(\vec{r}, \vec{r}')$. H_{ω} thus acts as a propagator. To calculate this range, one considers the case where $\Delta(\vec{r})$ depends on one coordinate only, that is, $\Delta(\vec{r}) = \Delta(x)$, then it can be shown (3) that, far from the NS interface,

$$H_{\omega}(x, x') = \frac{\pi N}{2|\omega| \xi_{\omega}} e^{-|x-x'|/\xi_{\omega}} \quad (17)$$

where the discrete frequencies $\omega = (2\nu + 1)\pi kT$, $\nu = 0, \pm 1, \pm 2, \dots$ and N is the density of states. The range of H_{ω} is thus

$$\xi_{\omega} = \left(\frac{v_F \ell}{6|\omega|} \right)^{1/2} \quad (18)$$

where v_F is the Fermi velocity and ℓ is the electronic mean free path. The largest range (when $\omega = \omega_0 = \pi kT$) gives the coherence length ξ ,

$$\xi_{\omega_0} = \xi(T) = \left(\frac{\hbar v_F \ell}{6\pi kT} \right)^{1/2} \quad (19)$$

Using the one-frequency approximation, de Gennes (3) derived the shape of the pair potential in closed form for an NS sandwich. He showed that for a superconductor and a normal metal separated by the plane $x = 0$ and $d_s \gg \xi_s$, $d_N \gg \xi_N$, then on the S side

$$\Delta(x) \frac{1-CN_V}{NV} = A \frac{\cos q(x-d_s)}{\cos(q d_s)} \quad x > 0 \quad (20)$$

and on the N side,

$$\Delta(x) \frac{1-CN_V}{NV} = A \frac{\cosh K(x+d_N)}{\cosh(K d_N)} \quad x < 0 \quad (21)$$

where d_s is the width of the superconductor, d_N is the width of the normal metal, A is an arbitrary constant and

$$C = \log \left(\frac{1.14\omega}{T} \right) - 2$$

$$q^2 = \frac{2\pi T}{D_s} \left(-1 + \frac{2(NV)_s}{1-C(NV)_s} \right) > 0$$

$$K^2 = \frac{2\pi T}{D_N} \left(1 - \frac{2(NV)_N}{1-C(NV)_N} \right) > 0$$

$$D = \frac{1}{3} v_F \ell = \text{diffusion coefficient}$$

Hence on the normal side, $\Delta(x)$ decays exponentially as

$$\Delta_N(x) \propto e^{-K_N|x|} \quad (22)$$

and the penetration depth of the pairs into the normal metal has the value

$$\begin{aligned} K_N^{-1} &= (\xi_{NO})^{1/2} \left[1 + \frac{2}{\ln(T/T_{CN})} \right]^{1/2} \\ &= \left(\frac{\hbar v_{FN} \ell_N}{6\pi kT} \right)^{1/2} \left[1 + \frac{2}{\ln(T/T_{CN})} \right]^{1/2} \end{aligned} \quad (23)$$

On the superconducting side the penetration depth is

$$\begin{aligned} K_S^{-1} &\equiv q^{-1} = (\xi_{SO})^{1/2} \left[\frac{2}{\ln(T_{CS}/T)} - 1 \right]^{1/2} \\ &= \left(\frac{\hbar v_{FS} \ell_S}{6\pi kT} \right)^{1/2} \left[\frac{2}{\ln(T_{CS}/T)} - 1 \right]^{1/2} \end{aligned} \quad (24)$$

K_S^{-1} reduces to the Ginzburg-Landau coherence length near T_{CS} . Superconductivity is thus induced in the normal metal region for distances on the order of the coherence length.

Typical variations of $\Delta(x)$ on both the superconductor and normal metal region are shown in Fig. 5. $\Delta(x)$ for an SNS junction is also plotted. The figures are not drawn to scale.

2. Boundary conditions

To understand the boundary conditions on the wave function, it is helpful to consider again a normal metal and a superconductor separated by the plane $x = 0$. De Gennes (3) showed that the first boundary condition has the form

$$\left[D(x') \frac{d}{dx'} H_{\omega}(xx') \right]_{x'=0^-}^{x'=0^+} = 0 \quad (25)$$

In terms of the pair potential Δ , this becomes

$$\left. \frac{D_N}{V_N} \frac{d\Delta_N}{dx} \right|_0 = \left. \frac{D_S}{V_S} \frac{d\Delta_S}{dx} \right|_0 \quad (26)$$

or

$$\left. \frac{\xi_N^2}{V_N} \frac{d\Delta_N}{dx} \right|_0 = \left. \frac{\xi_S^2}{V_S} \frac{d\Delta_S}{dx} \right|_0 \quad (27)$$

That is, the first boundary condition says that $\frac{D}{V} \frac{d\Delta}{dx}$ is continuous across the boundary. The second boundary condition has the form

$$H_{\omega}(x, 0^{+}) = \lambda H_{\omega}(x, 0^{-}) \quad (28)$$

where $\lambda = N_S/N_N$, the ratio of the superconducting to normal density of states. In a more useful form, this becomes

$$\frac{\Delta_N(0)}{(NV)_N(0)} = \frac{\Delta_S(0)}{(NV)_S(0)} \quad (29)$$

That is, Δ/NV is continuous across the boundary. In other words, the pair wave function is continuous across the boundary, but the pair potential changes at the boundary, as sketched in Fig. 5. One must be cautioned that the de Gennes boundary conditions are valid only in the dirty limit and at a temperature very close to the transition temperature of the NS sandwich.

For more complete discussion of proximity effect theories, see References 3, 22, 23 and 24.

II. EXPERIMENTAL PROCEDURE

A. Sample Preparation

As mentioned earlier, the Cu-Nb and Cu-Nb₃Sn samples were prepared at the Ames Laboratory by a casting and drawing technique called the in situ process (8, 9). Detailed discussion of this method is given in References 8 and 9, so only a brief outline of the preparation process of the composites is given here.

To prepare these alloys, an electrode composed of Cu and Nb is consumably arc melted, as shown in Fig. 6. By striking an arc, copper and niobium can be melted together and the liquid drips to the bottom of the copper container. By controlling the heat flow so that cooling comes primarily from the bottom, the solidified alloy consists of a uniform dispersion of Nb in a Cu matrix. The Nb precipitates are called dendrites. By repeating the process two or at most three times, homogeneity of $\pm 1\%$ can be achieved. The alloy is then extruded and drawn through a series of dies of decreasing size. This process aligns the dendrites in the direction of the wire and also reduces the wire to the desired size.

Typically the size of the Nb filaments is on the order of 60 nm in diameter and several cm long. The Cu barrier separating the filaments is on the order of 100 nm. This

cast copper-niobium alloy is very ductile, so the reduction process usually do not require heat treatment. In some instances, however, heat treatment is used because it enables one to control impurities such as carbon and to design the final filament configuration.

To convert the Nb dendrites to Nb_3Sn dendrites, tin can be introduced in two different ways. In one process, the alloy is extruded into a hollow cylinder and tin is inserted as a rod into the core of the cylinder. The wire is then drawn and heated at 550°C for several days. The heat treatment causes Sn to diffuse through the matrix and form Nb_3Sn . The end product would have a bronze core and Nb_3Sn dendrites in a copper matrix and hence is called "cored" material.

In the other process, the alloy is drawn to wire first, tin-plated and heat-treated. The end product has a bronze jacket instead. This in a sense is better because the ductility of the wire is improved. This process, however, is harder to achieve because surface tension causes the Sn to move along the wire when it melts rather than diffuse in.

The two $\text{Cu-Nb}_3\text{Sn}$ samples used in this study were both made from the first process, so they were cored material with different Cu to Nb_3Sn ratios.

B. Heat Capacity Apparatus

The ^3He cryostat used in this work was essentially the same as was used by L. F. Goodrich. Detailed discussion of the apparatus was discussed in his Doctor of Philosophy thesis (25), and general discussion of a standard ^3He cryostat can be found in Ref. 26, so only a brief summary of the apparatus will be mentioned here.

The ^3He cryostat was placed inside a pair of concentric glass dewars - a liquid nitrogen outer dewar and a liquid helium inner dewar, separated by a vacuum space. All pumping lines of the cryostat were made out of thin wall stainless steel tubing so as to minimize heat exchange with the outside. All electrical wires were #30 AWG manganin or copper wires in the helium bath and #30 or #36 AWG manganin wires in the vacuum can. These wires were anchored at terminal strips in the helium bath and also inside the sample vacuum can.

The ^3He pot and ^4He pot were constructed inside the sample vacuum can. Liquid ^4He was condensed by pressurizing the gas. The ^3He pumping lines was thermally anchored to the top of the ^4He pot so that pumping on the ^4He would allow ^3He to condense and reflux between the thermal anchor and the ^3He pot. The ^3He pumping line extended through the ^4He pot with the ^3He pot connected underneath. The length of this stainless steel tube decoupled the two pots enough

so that pumping on ^3He allowed a lower temperature to be reached. If the diffusion pump was used, a temperature of 0.29 K could be reached.

All samples were wound around a 3-inch long, 1/2-inch diameter sapphire tube and glued down with GE-7031 varnish. Sapphire has a very high thermal conductivity and low heat capacity, as needed. At 4 K, the thermal conductivity of sapphire is about 380 mJ/sec-cm-K, and the specific heat is about 0.1 mJ/gm.

The heat leak wire consisted of several 1/2-inch long sections of Pt-8% W wire, 0.002 inch in diameter. The number of sections used depended on the size of the sample measured such that it would cool the sample from 77 K to 4 K in a day or two and still maintain a slow enough cooling rate for data taking. This will be discussed in more detail in a later section.

The sample heater was a 31-inch long Pt-8% W wire, 0.001 inch in diameter. The resistance of this wire is 400 Ω per foot at 20°C, so the resistance of the heater wire is about 1010 Ω at room temperature. The heater wire was twisted bifilarly and glued onto a piece of kapton polyimide film with GE-7031 varnish (see Fig. 7). The kapton film was then glued to the inside of the sapphire tube, with the heater side touching the sapphire tube.

The thermometer holder was a copper sheet, about 0.002 inch thick. The copper sheet was bent into an omega shape

(Ω), as shown in Fig. 8. The length of the holder was just long enough to cover the thermometer and a small piece of copper sheet was placed on the bottom to hold the thermometer. The thermometer holder was glued to the inside of the sapphire tube using GE-7031 varnish. It was placed on the side where there was no heater wire and centered at the middle. The thermometer used was GRT 4645. Discussion of its calibration can be found in Goodrich's thesis (25). The thermometer was thermally anchored to the sapphire tube with N-grease (Apiezon).

Eight copper terminal strips were mechanically anchored to one end of the sample holder with GE-7031 varnish. Each thermometer lead from the terminal strips to the resistor consisted of a 4-inch long, 0.002 inch diameter Pt-8% W wire. The voltage lead to the heater was a 3-1/2-inch long Pt-8% W wire and the heater current leads were only 1/2-inch long sections of Pt-8% W wire in order to keep their resistance values low. Their resistance values were about 4.4 and 4.2 ohms at room temperature, a difference of about 5%. This gave the total heater lead resistance only about 0.85% of the total heater resistance, so the error from the power of the heat pulse should be less than 0.1%.

In this experiment, the sample was cooled by a heat leak device composed of several 1/2-inch long Pt-8% W wires

soldered together at the ends to two pieces of #36 copper wire, as shown in Fig. 9. One end of the heat leak wire was soldered to one of the terminal strips and the other end was bolted to the supporting frame of the ^3He stage. The sample holder was mounted onto the supporting frame by cotton thread for isolation. The thread was tied to the sapphire tube lengthwise. The whole assembly is sketched in Fig. 9.

Excluding the sample, all the parts shown in Fig. 9 plus the amount of GE-7031 varnish used to glue the sample is called the addenda. The total weight of the addenda was $10.1221 \text{ gm} \pm 0.0001 \text{ gm}$, of which 0.0350 gm was the amount of varnish used. The same addenda was used for all the samples measured.

C. Heat Capacity Measurement

The heat pulse method was used in this study. The thermal conductance of the heat leak wire was about 0.2 mK/sec at 1 K and 0.5 mK/sec at 4 K , with the shield temperature kept at about 0.35 K , so the heat pulses were adjusted to give temperature rises about 25 times larger, as will be discussed here. This permitted the acquisition of 0.1% C_p data.

In a typical heat pulse, the change in temperature ΔT was about 2% of T , so at $T = 1 \text{ K}$, $\Delta T \approx 20 \text{ mK}$ and at $T = 4 \text{ K}$, $\Delta T \approx 80 \text{ mK}$. The duration of the heat pulse was 4 seconds for

temperatures below 8 or 9 K and 10 seconds for higher temperatures. Therefore, the heating rate per heat pulse was about 5 mK/sec at 1 K and 20 mK/sec at 4 K. They were at least 25 times larger than the cooling rate of the sample through the heat leak wire. The ^3He shield temperature was kept at the lowest possible temperature. This kept the heat leak through the connecting wire nearly constant and it made the measurements much easier because the ^3He shield temperature needed not to be controlled at all. It also avoided the possibility of outgassing from the shield if it were to be kept up with the sample temperature.

If magnetic fields were applied, fields were applied with the sample temperature above the critical temperature and the sample was cooled in fields to the lowest possible temperature. This allowed the maximum amount of magnetic flux to penetrate the sample as it was cooled.

III. RESULTS AND DISCUSSION

A. Addenda Heat Capacity

The addenda for this experiment had a rather complex heat capacity below 1 K, as shown in Fig. 10. There was a distinct bump near 0.9 K which probably arises from the effect of ^3He in the thermometer condensing on the wall. Over most of the temperature range, however, the temperature dependence was close to the sum of electronic (γT) and lattice (αT^3) terms. The addenda heat capacity, C_{add} , was fit to a power series in temperature of the form

$$C_{\text{add}} = \sum_n a_n T^{(2n-1)} \quad (30)$$

where $n = 1, 2, \dots$

To accommodate the irregular shape of C_p vs. T the addenda C_p were divided into temperature intervals and where needed, a divergent term was added to the series to give

$$C_{\text{add}} = \sum_n a_n T^{(2n-3)} \quad (31)$$

To obtain a general form, the fit equations are rewritten in the form

$$\frac{C_{\text{add}}}{T} = \sum_{n=1}^L a_n T^{(2n-2)} \quad (32)$$

and

$$C_{\text{add}} T = \sum_{n=1}^L a_n T^{(2n-2)} \quad (33)$$

and the general form of the fit equations is then of the form

$$Y = \sum_{n=0}^{L-1} a_n X^{2n} \quad (34)$$

A least-squares-fitting technique was used to minimize $(Y - Y_{\text{calc}})^2 / Y_{\text{calc}}^2$, where the subscript calc denotes the calculated value. To calculate the fit constants a_n 's, Eqs. 32 and 33 were used to fit the zero field addenda data. As shown in Fig. 10, the structure of the addenda heat capacity in magnetic fields was very similar to that of zero field, except that the magnitude was higher at the low temperature region. At higher temperatures, the heat capacities approached the same value for all fields. Because the magnetic field dependence of the addenda was weak, the difference between the magnetic field data and the zero field data was fit to

$$C_{\text{add},H} = \sum_{n=0}^L a_n T^n \quad (35)$$

and the magnetic field heat capacity of the addenda was then the sum of the zero field heat capacity and the corresponding

difference. Within experimental error of 1% or less, the addenda heat capacity for fields less than 0.1 Tesla was essentially the same as that of the zero field. This leads to an uncertainty in the sample C_p of less than 0.3%. A table of the addenda fit constants for fields of 0, 0.1, 0.2, 0.4 and 1 Tesla is listed in Appendix A. It is presented in the form of a subroutine used in the computer programming. Seven temperature regions were used to fit the zero field data.

B. Cu-Nb System

The sample was Cu - 20 w/o Nb wire, untwisted and had a Cu shell which comprised 11% of the sample by volume. The wire diameter was 0.0113 inch and the total weight was 7.9798 gm $\pm$ 0.0001 gm. The weight distribution of the components is listed in Table 1.

Table 1. Weight distribution of the Cu-20 w/o Nb sample
(total weight of sample is 7.9798 gm)

	Cu matrix	Nb filaments	Cu shell
Weight (gm)	5.7375	1.4364	0.8060
Molecular weight (mole)	0.09030	0.01546	0.01268

The sample was measured in magnetic fields of 0, 0.01, 0.02, 0.05, 0.1, 0.2, 0.4 and 1 Tesla, with sample always cooling from above T_c in the presence of the measuring field. A list of the zero field heat capacity per gm-atom of the sample is tabulated in Appendix B. The percentage of the addenda heat capacity to that of the total sample is also listed. This ratio typically was about 30% in zero field and decreased to about 10% in 1 T field. A plot of the specific heat of the sample for all measured magnetic fields is shown in Fig. 11, plotting C/T vs. T^2 up to $T = 8.3$ K. As it is shown, the sample showed traces of superconductivity at $H = 0.4$ T, even through T_c was highly suppressed. This can be seen more readily by plotting the difference between the normal state heat capacity C_N of the composite and the superconducting state value $C_S(H)$, as shown in Fig. 12. The 1 T data was used as the normal state value. The magnitude of the specific heat jump, $\Delta C = C_S - C_N$, as indicated in Fig. 12, is determined by drawing a straight line through the start of the jump and a vertical line through the experimentally determined T_c (will be discussed later) and the intercept gives the magnitude of the jump. ΔC as a function of applied field is plotted in Fig. 13, and $\Delta C/\gamma_{Nb} T_c$ as a function of magnetic field is plotted in Fig. 14. For pure Nb, $\Delta C/\gamma_{Nb} T_c \approx 1.44$. At zero field $\Delta C \approx 1.52$ mJ/K for the

sample which is only about 5% smaller than the literature value of pure Nb (27, 28) of 1.6 mJ/K. This indicates that most of the jump was due to Nb filaments.

To calculate T_c , the percentage difference between C_S and C_N , $[C_S(H) - C_N]/C_N$ (in %), was plotted in Fig. 15. As indicated in the figure, T_c is defined (29) as the midpoint between the intercept formed by the two straight lines through the start and the end of the jump and the intercept formed by the line through the end of the jump and the horizontal zero line. Values of T_c as a function of magnetic field, that is, the critical field curve, are plotted in Fig. 16. Plotted also are the critical field curves for pure Nb determined from magnetization data (30) and for Th-Nb composites determined from heat capacity data (25). It shows that T_c for zero field is suppressed much more in the Cu-Nb case than in the Th-Nb case, but H_{C2} rises much more steeply as the temperature is lowered. This can be due to many factors. The presence of dissolved Cu in the Nb will suppress T_c and increase kappa (κ) through the mean free path effect. This indicates that more Cu got dissolved in the Nb than Th, hence shortened the coherence length.

To confirm the observation that the Nb is in the dirty regime, $\kappa_1(T)$ of the Nb filaments was calculated. κ_1 is related to the Ginzburg-Landau parameter κ by the expression

$$\kappa = \lim_{T \rightarrow T_c} \kappa_1(T) = \lim_{T \rightarrow T_c} \frac{H_{c2}}{\sqrt{2} H_c} \quad (36)$$

Using the assumption that the electronic specific heat coefficient γ does not change from the pure Nb value, and the fact that $H_c \propto \gamma T_c$, the so-called law of corresponding states is obtained:

$$\frac{H_0 \text{ (Nb filament)}}{T_0 \text{ (Nb filament)}} = \frac{H_0 \text{ (Nb)}}{T_0 \text{ (Nb)}} \quad (37)$$

where H_0 denotes the critical field H_c at $T = 0$ and T_0 is the critical temperature T_c at $H = 0$. From T. Stromberg's thesis (31), one has $H_0 \text{ (Nb)} = 1993$ gauss and $T_0 \text{ (Nb)} = 9.25$ K. Since $T_0 \text{ (Nb filament)} = 7.6$ K (Fig. 16), one then has $H_0 \text{ (filament)} = 1637.5$ gauss. Using the critical field equation

$$H_c = H_0 (1 - t^2) \quad (38)$$

where $t = T/T_c$, one obtains

$$H_c \text{ (filament)} = 1637.5 (1 - t^2) \text{ gauss} \quad (39)$$

Using the critical field curve from Fig. 16, κ_1 thus can be calculated from Eq. 36. The calculated κ_1 values as a function of applied magnetic field are listed in Table 2. Using Eq. 36, κ was found to be about 3.2. This gave a mean free

Table 2. Summary of the calculated values for κ_1

T (K)	$1-t^2$	H_c (gauss)	H_{c2} (gauss)	κ_1
5.09	0.551	902	4000	3.136
6.51	0.266	436	2000	3.244
7.13	0.121	198	1000	3.571
7.35	0.065	106	500	3.335
7.50	0.026	43	200	3.289

path of approximately 1×10^{-6} cm. The coherence length ξ_0 for pure Nb is 3.8×10^{-6} cm. Hence, the Nb filament is in the dirty regime.

In addition, κ can also be expressed as the sum of the two terms (32)

$$\kappa = \kappa_0 + \kappa_e \quad (40)$$

κ_0 involves only the electronic structure of the metal, independent of electronic scattering, and is given by

$$\kappa_0 = 0.96 \lambda_L(0)/\xi_0 \quad (41)$$

where $\lambda_L(0)$ is the London penetration depth at $T = 0$ and ξ_0 is the BCS coherence length.

The κ_e term involves the electronic mean free path and is given by

$$\kappa_e = 7.53 \times 10^3 \rho_N \gamma^{1/2} \quad (42)$$

where ρ_N is the normal state resistivity.

For pure Nb, $\kappa = 0.78$, $\rho_N(4.2 \text{ K}) = 1.04 \times 10^{-8} \text{ ohm-cm}$ and $\gamma = 7.0 \times 10^4 \text{ ergs/cm}^3 \text{ K}^2$. Using these values, one obtains

$$\kappa_e (\text{Nb}) = 0.021$$

and

$$\kappa_o (\text{Nb}) = \kappa - \kappa_e = 0.76$$

The values for κ_e and ρ_N of the filaments thus can be calculated from

$$\kappa_e(\text{filament}) = \kappa(\text{filament}) - 0.76 \quad (43)$$

$$\text{and } \rho_N(\text{filament}) = \kappa_e(\text{filament}) / 7.5 \times 10^3 \gamma^{1/2} \quad (44)$$

and one obtained that $\kappa_e \approx 2.5$ and $\rho_N \approx 1.27 \times 10^{-6} \text{ ohm-cm}$.

To obtain information about the electronic excitation spectrum in the Cu matrix, one way to accomplish this is by computing the entropy S_s using the equation

$$S_s(T') = \int_0^{T'} \frac{C_{es}}{T} dT \quad (45)$$

The measured C_{es} data of the composite were fit to polynomials for such a calculation. The calculated entropy values for all measured fields are plotted in Fig. 17 for temperatures below 1.5 K. The normal state entropy values for Cu and for Cu plus Nb are also included. As shown, the zero field entropy curve falls far below the Cu normal state value, indicating that the Cu does indeed show the effects of an induced pair potential. As the magnetic field strength increases, the entropy increases rather rapidly and at 0.7 K, it becomes equal to $\gamma_{Cu} T$ at about 17 mT. Hence, rather small magnetic fields of a few mT destroy the energy gap in the Cu which then behaves as a gapless superconductor. This finding is similar to that found in Pb-Cd-Pb triple layer junctions (4).

It has been pointed out by Maki (33) that there is a very close correspondence between all forms of gapless superconductivity whether it occurs in the core of a vortex, in a paramagnetic impurity doped superconductor or in a proximity junction. This arises because life-time broadening has similar effects on the excitation spectrum in all of these cases. If one takes seriously this premise that all life-time broadened superconductors have the same excitation spectrum, then it may possibly use the energy dependence of the density of states derived for paramagnetic impurity scattering by Abrikosov and Gor'kov (34) and by Skalski et al. (35).

To obtain some idea about the excitation spectrum in the Cu matrix, one can calculate a whole family of life-time broadened density of states $N(\omega)$ which gives the measured entropy values of the Cu and Nb calculated from Eq. 45. The association between life-time broadening due to proximity effect and paramagnetic impurity scattering probably is valid only at very low temperatures where Δ is essentially uniform across the Cu regions. At higher temperatures where Δ has a significant space dependence, a more complex theory would be required.

As discussed by Skalski et al. (35), there is a general relation between S and $N(\omega)$ given by

$$S = 4k \int_0^{\omega_D} d\omega N(\omega) \left[\ln(1 + e^{-\beta\omega}) + \beta\omega(e^{\beta\omega} + 1)^{-1} \right] \quad (46)$$

where $\beta = (kT)^{-1}$ and ω_D is the BCS cutoff energy. $N(\omega)$ is determined numerically by trial and error and the procedure used here is only an approximation.

One guesses a value for $\Gamma/\Delta^p(0)$, where Γ is the inverse collision time for exchange scattering and $\Delta^p(0)$ is the order parameter at $T = 0$. The superscript p refers to the value when $\Gamma = 0$ (BCS like behavior). The sample is in the normal state when $\Gamma = 1/2$. From Fig. 18a (reproduced from Fig. 2

of Ref. 35), the corresponding values for T_c/T_c^P and $\Delta(0)/\Delta^P(0)$ are obtained. $\Delta(T)/\Delta^P(0)$ as a function of T/T_c^P is thus generated, similar to those shown in Fig. 18b. For a particular temperature T' , the corresponding $\Delta(T')/\Delta^P(0)$ can then be obtained. From these values, one then obtains

$$\frac{\Gamma}{\Delta(T', \Gamma)} = \frac{\Gamma/\Delta^P(0)}{\Delta(T', \Gamma)/\Delta^P(0)} \quad (47)$$

Using this value, $N(\omega)/N_0$ as a function of $\omega/\Delta(T)$ can be calculated, similar to those shown in Fig. 18c, where N_0 is the density of states at the Fermi surface. A family of life-time broadened density of states curves which gives the measured entropy for $T < 1$ K was calculated based on this procedure and plotted in Fig. 19. One must be cautioned that the curves give only the Skalski et al. $N(\omega)$ which fits the C_e data of Cu plus Nb. The spatial resolution of $N(\omega)$ is not shown here because the Skalski et al. calculation works only in a homogeneous medium. Hence, the curves give only the changes in $N(\omega)$ for the composite system.

A rather different way to obtain information about the excitation spectrum in the Cu matrix is to perform a model calculation in which we assume that the electronic specific heat of the Cu matrix is of the form

$$C_e(\text{Cu}) = \gamma'T + Ae^{-\Delta/kT} \quad (48)$$

The first term here is essentially normal-like term and the second term is similar to a BCS superconductor. If one subtracts the contributions from the Cu jacket, the lattice terms of the Cu matrix and Nb filaments, and the superconducting electronic term of the Nb (from references 27 and 28), one would get C_e of Cu plus possibly the normal electronic term of Nb if the magnetic field is high enough to drive part of the Nb normal. This C_e term is plotted in Fig. 20. Using the fact that at low temperatures (below 1 K), the exponential term in Eq. 48 is much smaller than the $\gamma'T$ term, so γ' can be obtained by extrapolating the data to $T = 0$ in Fig. 20. γ' as a function of magnetic field is plotted in Fig. 21.

For $H = 1$ Tesla, the sample is in the normal state and γ' should be of the normal state value. The γ' value for the 1 T data, however, is about 0.217 mJ/K^2 which is about 15% higher than the sum of the bulk γ_{Cu} plus γ_{Nb} . This rather high discrepancy is probably not all due to experimental uncertainty. It is possible that these $1000 \text{ \AA}$ filamentary Cu barriers do not have the same properties as the bulk Cu. As discussed by Bevk and Massalski (36), the γ value for a CuSn alloy is higher than that of pure Cu. If 8.71 at % Sn is added to the Cu, the γ value is higher by as much as 9%.

This same effect could be similar in the Cu-Nb alloy. Further investigation of this effect is needed.

Using these γ' values in Eq. 48, the superconducting part of the electronic specific heat, C_{es} , of the Cu matrix can be calculated from the relation

$$C_{es}(\text{Cu}) = C_e(\text{Cu}) - \gamma' T \quad (49)$$

Since C_{es} is of the form

$$C_{es} = A e^{-\Delta/kT} \quad (50)$$

or
$$\ln C_{es} = \ln A - \frac{\Delta}{kT} \quad (51)$$

the pair potential Δ can thus be readily deduced from the measured specific heat data.

$\ln C_{es}$ ($H = 0$) as a function of T_c/T is plotted in Fig. 22. The data is very linear with T_c/T up to $T \approx 3$ K. By fitting the data in the temperature region below 3 K where Δ is essentially constant, the thermal average of Δ is calculated, of value

$$\Delta_{\text{Cu}} \approx 0.35 \text{ meV}$$

To see how well this obeys the de Gennes boundary condition (Eq. 26), one starts with the expression

$$kT_c = \frac{k\theta_D}{1.45} e^{-1/NV} \quad (52)$$

for Nb, and using $\theta_D(\text{Nb}) = 275 \text{ K}$, one finds $(\text{NV})_{\text{Nb}} \cong 0.33$.

Using $\Delta_{\text{Nb}} = 1.56 \text{ meV}$ (37), one obtains

$$\begin{aligned} (\text{NV})_{\text{Cu}} &= (\text{NV})_{\text{Nb}} \cdot \frac{\Delta_{\text{Cu}}}{\Delta_{\text{Nb}}} \\ &= 0.074 \end{aligned}$$

This is within the experimentally determined range of 0.05 to 0.12 (24)! Hence, the proximity effect existing in this composite obeys the theory rather well and the proximity coupling between the filaments is very effective.

C. Cu-Nb₃Sn System

Two samples of this in situ composite were measured and most of the results were similar to those presented for the Cu-Nb sample. The first sample was prepared from arc cast #4 as cored wire, 0.0115 inch in diameter. The wire was untwisted and reacted at 550°C for 6 days. The total weight of the sample was 4.7467 gm $\pm$ 0.0001 gm. The weight distribution of the components is listed in Table 3. The second sample was also a cored wire, cast #24. It was twisted and had a diameter of 0.0126 inch. The total sample weight was 9.9449 gm $\pm$ 0.0001 gm. The weight distribution of the components is listed in Table 4 and a photograph of this sample is shown in Fig. 23.

Table 3. Weight distribution of arc cast #4 cored Cu-Nb₃Sn sample (total weight of sample is 4.7467 gm)

	Cu matrix	Nb ₃ Sn filaments	Bronze core
Percentage weight	52%	20%	28%
Weight (gm)	2.4683	0.9493	1.3291

Table 4. Weight distribution of arc cast #24 cored Cu-Nb₃Sn sample (total weight of sample is 9.9449 gm)

	Cu matrix	Nb ₃ Sn filaments	Bronze core
Percentage weight	57%	20.5%	22.5%
Weight (gm)	5.6686	2.0387	2.2376

Both samples were measured in magnetic fields of 0, 0.05, 0.1, 0.2, 0.4 and 1.0 Tesla. Their specific heat values were essentially the same, so only the results for the arc cast #24 sample will be presented.

A list of the zero field heat capacity per gm-atom of sample is tabulated in Appendix C. A plot of C/T vs. T^2 is

shown in Fig. 24 for temperatures below 1.3 K. Fig. 25 shows a plot of the electronic specific heat per Cu mole. Since H_{c2} of pure Nb_3Sn is about 20 Tesla, it is thus assumed here that Nb_3Sn does not go normal in these measured fields. The figure indicates again that filamentary Cu probably does not have the same heat capacity as bulk Cu because at $H = 0.05$ T, γ' of the composite is bigger than the γ value for bulk Cu, although some $\gamma'T$ behavior in the Nb_3Sn is expected for fields above H_{c1} of Nb_3Sn which is about 0.15 Tesla.

From the measured C_{es} , the entropy was calculated using Eq. 45 and is plotted in Fig. 26 for all measured magnetic fields. As shown in Fig. 26, the entropy value does not rise as quickly as for the Cu-Nb sample when the magnetic field strength increases, and at $T = 0.7$ K, it becomes equal to $\gamma_{Cu}T$ at a much higher magnetic field, on the order of about 45 mT. However, this is still a rather small magnetic field above which the Cu behaves as a gapless superconductor. A family of life-time broadened density of states curves which gives the measured entropy for $T < 1$ K is shown in Fig. 27. One must be cautioned again that they give only the $N(\omega)$ curves for the composite system.

It is more difficult to deduce the thermal average of the pair potential Δ for the Cu- Nb_3Sn sample because in this sample, there is probably still about 1% Sn left in the Cu

matrix due to incomplete reaction process; thus, the mean free path is shortened and the pair potential decay length in the Cu, $K_N^{-1} = (\hbar v_F \ell / 6\pi kT)^{1/2}$, is much shorter than in the Cu-Nb sample. The pair potential thus has a substantial space dependence, not uniform across the Cu barrier as it was assumed in the Cu-Nb composite.

If one uses the assumption that Δ is constant across the Cu barrier in this sample and calculates the pair potential that would be induced in the Cu, one obtains

$$\Delta_{\text{Cu}} \cong 0.28 \text{ meV}$$

Using de Gennes boundary condition (Eq. 26) and $(NV)_{\text{Nb}_3\text{Sn}} = 0.4$, $\Delta_{\text{Nb}_3\text{Sn}} = 3 \text{ meV}$, one finds that

$$(NV)_{\text{Cu}} \cong 0.04$$

which is fairly close to the literature lower limit of value 0.05. One thus would expect that at the interface, the induced pair potential has a much higher value than at the center of the barrier.

IV. SUMMARY

Two superconductor-normal metal in situ composites, Cu-Nb and Cu-Nb₃Sn, prepared at the Ames Laboratory by the consumable arc casting technique have been found to have an induced pair potential Δ in the Cu regions. For the Cu-Nb composites specific heat measurements indicate that the induced pair potential has a value of about 0.35 meV in zero applied field. Based on de Gennes boundary conditions, this gives a value for NV , the product of the density of states and interaction parameter, of Cu to be about 0.07 which is within the literature limits of values 0.05 and 0.1.

A family of life-time broadened density of states $N(\omega)$ which gives the measured entropy S (and thus C_{es}) for the composite is calculated based on the general relation between S and $N(\omega)$ derived for life-time broadened superconductors, as discussed by Skalski et al. S is calculated from the measured C_p using Eq. 45. The entropy is found to rise quickly as the magnetic field increases and at 0.7°K it becomes equal to $\gamma_{Cu}T$ at about 17 mT. The Cu thus behaves as a gapless superconductor in rather small magnetic fields. The family of density of states curves shows roughly the changes in $N(\omega)$ which are required to give the measured S and thus C_p .

In zero field a specific heat jump occurring at 7.6°K has a magnitude of about 1.52 mJ/K which is expected from the amount of Nb present. This jump decreases rather quickly as the magnetic field strength increases and is down to about 20% of the zero field value at 0.4 Tesla. The measured critical field curve gives a H_{c2} value for the composite of about 0.75 Tesla and a kappa value of about 3.3 for Nb, indicating that Nb is indeed in the dirty regime.

For the Cu-Nb₃Sn sample the analysis is more complicated because about 1% Sn is still left in the Cu matrix. Hence the decay length K_N^{-1} in the Cu is much shorter and the pair potential thus has a substantial space dependence. In other respects the results are similar to those presented for the Cu-Nb sample.

Figure 1. The pair potential Δ as a function of temperature (from Ref. 15), T_c is the critical temperature of the superconductor

Figure 2. A typical photon absorption plot where the ratio of the absorption coefficients for the superconducting state (α_S) and normal state (α_N) is plotted as a function of the photon energy $\hbar\omega$ and the onset of absorption gives the value 2Δ

Figure 3. A typical normal to superconducting I-V plot where the dashed line indicates the normal state value

Figure 4. Approximation for the interaction parameter $V_{kk'}$

Figure 5. Typical variations of the pair potential $\Delta(x)$ as a function of position (not drawn to scale)

Figure 6. Copper-Niobium ingot produced by consumable arc casting technique

Position of heater on sample holder

Figure 7. Sample heater configuration

Figure 8. Thermometer holder configuration

Figure 9. Sample holder arrangement (not drawn to scale)

Figure 10. Addenda heat capacity illustrating the low temperature structures in zero and 1 T fields

Figure 11. Specific heat of Cu-Nb sample

Figure 12. Plot of the difference of Cu-Nb sample heat capacity data from the normal state data as a function of temperature to illustrate the specific heat jump $\Delta C = C_S - C_N$ (the solid lines indicate the magnitude of the jump in zero field)

Figure 13. The specific heat jump ΔC of Cu-Nb as a function of applied magnetic field

Figure 14. Plot of $\Delta C/\gamma_{Nb}T_c$ of Cu-Nb as a function of magnetic field ($\Delta C/\gamma_{Nb}T_c = 1.44$ for pure Nb)

Figure 15. Percent difference of Cu-Nb heat capacity data from the normal state data (the solid and dashed lines illustrate how to determine T_c)

Figure 16. Critical field curve of Cu-Nb compared with those of Th-Nb and pure Nb

Figure 17. Electronic entropy of the Cu-20 w/o Nb sample

FIG. 5. The order parameter plotted as a function of temperature for several values of the inverse collision time. The superscript P refers to the value when $\Gamma=0$. The dashed curve is a boundary curve separating the gapless region from the region with a gap.

FIG. 4. The density of states in energy plotted as a function of the reduced energy ω/Δ for several values of the reduced inverse collision time Γ/Δ . $\Delta(T)$ is to be understood as $\Delta(T, \Gamma)$.

Figure 19. A family of life-time broadened density of states which gives the measured entropy for the Cu-Nb sample

Figure 20. The electronic specific heat C_e of the Cu-Nb sample where $C_e = C_p (\text{total sample}) - C_p (\text{jacket}) - \alpha_{\text{Cu}} T^3 - \alpha_{\text{Nb}} T^3 - C_{\text{es}} (\text{Nb})$

Figure 21. Values of γ' extrapolated from Figure 20 as a function of magnetic field

Figure 22. Plot of $\ln C_{es}(\text{Cu})$ as a function of T_c/T for the Cu-Nb sample

Figure 23. A photograph of the arc cast #24 cored Cu-Nb₃Sn sample with a bronze core

Figure 24. Heat capacity of the arc cast #24 cored Cu-Nb₃Sn sample for $T < 1.3^\circ K$

Figure 25. The electronic specific heat C_e of the arc cast #24 cored Cu-Nb₃Sn sample

Figure 26. Electronic entropy of the arc cast #24 cored Cu-Nb₃Sn sample

Figure 27. A family of life-time broadened density of states which gives the measured entropy for the arc cast #24 cored Cu-Nb₃Sn sample

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VIII. APPENDIX A: SUBROUTINE FOR THE
ADDENDA HEAT CAPACITY

```

SUBROUTINE CADD (I,T,CAD,H)
DOUBLE PRECISION A(8),T(1),CAD,CADH,B(4)
DIMENSION H(1)
CADH = 0.0D0

      IF (T(I)- 0.570) 10,1,1
1  IF (T(I)- 0.915) 11,2,2
2  IF (T(I)- 1.005) 12,3,3
3  IF (T(I)- 1.620) 13,4,4
4  IF (T(I)- 3.70 ) 14,5,5
5  IF (T(I)- 8.60 ) 15,16,16
10 A(1) = 7.72016867478695D-01
   A(2) = -1.20102417802552D 01
   A(3) = 7.73128481154535D 01
   A(4) = -2.62231132900120D 02
   A(5) = 4.94516173851537D 02
   A(6) = -4.91424279868275D 02
   A(7) = 2.01138976859432D 02
   A(8) = 0.0                      D 00
   NFIT=-1
   GO TO 20
11 A(1) = -1.35848994334069D-01
   A(2) = 1.71774604516966D 00
   A(3) = -8.65416566035422D 00
   A(4) = 2.28834612002572D 01
   A(5) = -3.33019550416266D 01
   A(6) = 2.54320369604124D 01
   A(7) = -7.93739363951927D 00
   A(8) = 0.0                      D 00
   NFIT=-1
   GO TO 20
12 A(1) = 1.44400461886015D 03
   A(2) = -9.78398387233431D 03
   A(3) = 2.74563544481122D 04
   A(4) = -4.08655900159329D 04
   A(5) = 3.40383094136652D 04
   A(6) = -1.50492509384202D 04
   A(7) = 2.76017814836902D 03
   A(8) = 0.0                      D 00
   NFIT=-1
   GO TO 20

```



```

13  A(1) = 1.05672387987485D-01
    A(2) = -2.72073661004616D-01
    A(3) = 3.06779546788998D-01
    A(4) = -1.53834967375202D-01
    A(5) = 3.89043488022101D-02
    A(6) = -3.82455203318527D-03
    A(7) = 0.0                D 00
    A(8) = 0.0                D 00
    NFIT=-1
    GO TO 20
14  A(1) = 6.03051638864103D-03
    A(2) = 2.20826090082400D-03
    A(3) = 1.20148092908512D-02
    A(4) = -7.04146924498042D-05
    A(5) = 0.0                D 00
    A(6) = 0.0                D 00
    A(7) = 0.0                D 00
    A(8) = 0.0                D 00
    NFIT=-1
    GO TO 20
15  A(1) = 2.92038168084529D-03
    A(2) = 1.21931232915553D-02
    A(3) = -9.25650241996797D-05
    A(4) = 5.76062164323431D-07
    A(5) = -1.62440740654423D-09
    A(6) = 0.0                D 00
    A(7) = 0.0                D 00
    A(8) = 0.0                D 00
    NIFT= 1
    GO TO 20
16  A(1) = 1.16908357895119D-01
    A(2) = 7.59495110331486D-03
    A(3) = -1.98348445035447D-05
    A(4) = 3.29582180057882D-08
    A(5) = -2.07991770214705D-11
    A(6) = 0.0                D 00
    A(7) = 0.0                D 00
    A(8) = 0.0                D 00
    NFIT= 1
20  IF (H(I).EQ.10.0) GO TO 30
    IF (H(I).EQ.4.0)  GO TO 40
    IF (H(I).EQ.2.0)  GO TO 50
    IF (H(I).EQ.1.0)  GO TO 60
    GO TO 100

```

```

30  IF (T(I).GT. 2.40) GO TO 100
    IF (T(I).GT. 1.20) GO TO 31
    B(1) = 4.64463091753007D-03
    B(2) = -1.31832617194992D-03
    B(3) = 0.0 D 00
    B(4) = 0.0 D 00
    GO TO 35
31  B(1) = -5.27112443645171D-03
    B(2) = 1.80359440378494D-02
    B(3) = -1.18615967778683D-02
    B(4) = 2.26041340000567D-03
35  DO 36 K = 1,4
36  CADH = CADH+B(5-K)*(T(I)**(4-K))
    GO TO 100

40  IF (T(I).GT. 1.98) GO TO 100
    IF (T(I).GT. 1.20) GO TO 41
    B(1) = 2.01345969285529D-03
    B(2) = -5.68647311170471D-04
    B(3) = 0.0 D 00
    B(4) = 0.0 D 00
    GO TO 45
41  B(1) = -2.19557098015695D-02
    B(2) = 4.68198553979928D-02
    B(3) = -3.03314875520771D-02
    B(4) = 6.22508590918281D-03
45  DO 46 K = 1,4
46  CADH = CADH+B(5-K)*(T(I)**(4-K))
    GO TO 100

50  IF (T(I).GT. 1.60) GO TO 100
    IF (T(I).GT. 1.20) GO TO 51
    B(1) = 1.01345959285529D-03
    B(2) = -5.68647311170471D-04
    B(3) = 0.0 D 00
    B(4) = 0.0 D 00
    GO TO 55
51  B(1) = 1.19557098015695D-03
    B(2) = -1.48212918472513D-03
    B(3) = 0.0 D 00
    B(4) = 0.0 D 00
55  DO 56 K = 1,4
56  CADH = CADH+B(5-K)*(T(I)**(4-K))
    GO TO 100

```

```

60  IF (T(I).GT. 1.20) GO TO 100
    B(1) = 7.18502695101517D-04
    B(2) = -3.58037943237940D-04
    B(3) = 0.0                      D 00
    B(4) = 0.0                      D 00
    DO 66 K = 1,4
66  CADH = CADH+B(5-K)*(T(I)**(4-K))
100  CAD=0.0 DOO
    DO 101 K=1,8
101  CAD=CAD+A(9-K)*(T(I)**2)**(8-K)
    IF (NFIT.EQ.-1) GO TO 110
    CAD=CAD*T(I)
    GO TO 200
110  CAD=CAD/T(I)
200  CAD = CAD+CADH
    RETURN
    END

```

IX. APPENDIX B: ZERO FIELD SPECIFIC HEAT DATA
OF Cu-Nb SAMPLE

T (K)	C _p (mJ/gm-atom K)	C _{add} /C _{total} (percent)
0.57484	0.12359	38.565
0.59064	0.11971	39.996
0.60283	0.11284	41.951
0.61421	0.13971	37.372
0.62220	0.13830	38.010
0.63007	0.14721	36.973
0.64750	0.15141	37.382
0.66155	0.17535	34.963
0.67416	0.18929	34.165
0.68746	0.18099	36.247
0.69859	0.18726	36.405
0.71247	0.19173	37.089
0.72322	0.19715	37.430
0.73376	0.20703	37.283
0.74536	0.20700	38.420
0.75612	0.23105	36.915
0.76869	0.23662	37.649
0.78101	0.23611	39.025
0.79144	0.25053	38.781
0.80156	0.26011	39.057
0.81382	0.27530	39.170
0.82609	0.28075	40.221
0.83504	0.28532	40.973
0.84514	0.28498	42.316
0.85938	0.31905	41.240
0.87196	0.32317	42.704
0.88627	0.33205	43.751
0.89938	0.35388	43.563
0.91090	0.36293	43.910
0.92468	0.35942	44.267
0.92671	0.37451	43.006
0.94391	0.39159	38.052
0.96067	0.42093	32.307
0.97561	0.44312	29.465
0.99210	0.44577	29.209
1.00860	0.45675	28.660
1.02020	0.46767	28.351
1.03370	0.50150	27.192
1.04735	0.51739	26.859

T (K)	C _p (mJ/gm-atom K)	C _{add} /C _{total} (percent)
1.06209	0.53572	26.522
1.07041	0.55139	26.171
1.07814	0.56073	26.049
1.07844	0.56260	25.993
1.08793	0.57210	25.931
1.09582	0.57059	26.205
1.10828	0.60041	25.594
1.12494	0.62482	25.343
1.14233	0.64122	25.398
1.15984	0.68116	24.821
1.17727	0.68948	25.159
1.19208	0.71950	24.844
1.20964	0.75409	24.543
1.22798	0.77150	24.716
1.24873	0.80287	24.648
1.26433	0.84365	24.232
1.28544	0.86596	24.419
1.30725	0.89042	24.589
1.33137	0.94746	24.192
1.35104	0.98733	24.037
1.36525	1.01006	24.050
1.38666	1.03407	24.264
1.40953	1.09160	23.958
1.43056	1.13304	23.904
1.45448	1.17776	23.909
1.47108	1.20169	24.037
1.48930	1.23862	24.029
1.49958	1.27643	23.789
1.52420	1.32566	23.838
1.54507	1.35676	24.045
1.56675	1.41141	23.981
1.58731	1.44681	24.153
1.61051	1.49033	24.315
1.63520	1.54925	24.333
1.66038	1.60384	24.429
1.67428	1.65517	24.251
1.69706	1.70485	24.361
1.72143	1.76498	24.416
1.71548	1.73427	24.572
1.74737	1.81815	24.602
1.77783	1.86902	24.947
1.80811	1.96104	24.895
1.84289	2.05183	24.009
1.87902	2.14123	25.196
1.91961	2.23751	25.463

T (K)	C _p (mJ/gm-atom K)	C _{add} /C _{total} (percent)
1.96010	2.37340	25.427
1.99655	2.46180	24.693
2.03796	2.59152	25.792
2.10063	2.76682	26.144
1.93486	2.30085	25.344
1.97031	2.39068	25.560
2.01541	2.51413	25.785
2.05682	2.64347	25.899
2.09524	2.74415	26.165
2.12620	2.83577	26.317
2.16433	2.93416	26.614
2.20580	3.08534	26.666
2.24614	3.20534	26.910
2.28997	3.34458	27.137
2.32930	3.45828	27.421
2.37517	3.62782	27.558
2.44769	3.84824	28.071
2.49489	4.07422	28.005
2.54806	4.26420	28.285
2.59491	4.43200	28.546
2.65684	4.70600	28.680
2.71776	4.98432	28.815
2.78210	6.16403	29.449
2.84714	5.47135	29.605
2.91429	5.68851	30.170
2.98765	6.11123	30.139
3.06224	6.48080	30.370
3.14178	6.81641	30.834
3.21409	7.20417	31.022
3.28835	7.59307	31.274
3.37882	8.14691	31.401
3.43156	8.33575	31.846
3.51351	8.86060	31.958
3.60385	9.37569	32.275
3.69426	10.04599	32.278
3.80262	10.68545	32.678
3.91424	11.40338	33.005
4.02278	12.14945	33.268
4.12526	12.97965	33.332
4.21863	13.42478	33.945
4.33503	14.48321	33.906
4.44056	15.32973	34.095
4.50723	15.79314	34.333
4.55428	16.11242	34.514
4.60095	16.56503	34.509

T (K)	C _p (mJ/gm-atom K)	C _{add} /C _{total} (percent)
4.65175	16.80734	34.849
4.70531	17.49386	34.639
4.75997	17.98109	34.718
4.82125	18.63850	34.679
4.88325	19.16510	34.821
4.94424	19.65798	34.995
5.00592	20.39683	34.917
5.07502	21.01908	35.047
5.14465	21.65564	35.188
5.21492	22.49429	35.137
5.28581	23.13863	35.313
5.35773	23.87854	35.393
5.42714	24.79880	35.298
5.50180	25.75052	35.251
5.57901	26.40825	35.503
5.65193	27.48818	35.356
5.73388	27.75032	35.992
5.81850	29.44098	35.500
5.90428	30.48004	35.566
5.98473	31.44561	35.645
6.07138	32.46626	35.753
6.16231	33.83189	35.675
6.25702	34.73139	35.960
6.34723	35.94862	35.998
6.44311	37.32440	36.002
6.54130	38.42515	36.205
6.63708	40.05169	36.086
6.73809	40.96776	36.432
6.83889	42.49440	36.437
6.93879	44.14825	36.383
7.05080	45.43659	36.629
7.17018	46.42173	37.088
7.28746	48.35310	37.062
7.42236	49.40629	37.601
7.56075	49.82373	38.457
7.64475	50.46334	38.787
7.79527	51.36089	39.484
7.94735	51.72604	40.425
8.08921	52.53739	41.068
8.23231	53.91908	41.449
8.37256	55.35909	41.781
8.51155	56.76465	42.117
8.64834	59.06074	42.059
8.78054	61.57966	41.896
8.91586	63.53923	41.993
9.04980	65.84301	41.963

T (K)	C _p (mJ/gm-atom K)	C _{add} /C _{total} (percent)
9.18459	67.95150	42.025
9.32368	70.56487	41.948
9.46141	73.68258	41.715
9.60810	75.98131	41.826
9.76018	79.58678	41.573
9.93126	82.59482	41.636
10.00206	84.04867	41.606
10.19246	91.49053	40.591
10.38820	92.41685	41.391
10.57403	96.74723	41.263

X. APPENDIX C: ZERO FIELD SPECIFIC HEAT DATA
OF ARC CAST #24 CORED Cu-Nb₃Sn SAMPLE

T (K)	C _p (mJ/gm-atom K)	C _{add} /C _{total} (percent)
0.48763	0.27694	22.414
0.49296	0.28100	21.029
0.50612	0.29093	18.594
0.51204	0.29549	17.864
0.52261	0.30256	17.022
0.53135	0.31025	16.525
0.54125	0.31592	16.293
0.54988	0.32314	16.091
0.54094	0.31568	16.301
0.55312	0.32509	16.060
0.56403	0.33315	15.925
0.57102	0.33876	15.662
0.58058	0.34577	15.625
0.58943	0.35285	15.563
0.59770	0.35768	15.576
0.60667	0.36393	15.565
0.61748	0.37343	15.512
0.61364	0.37177	15.466
0.62141	0.37694	15.501
0.63201	0.38622	15.505
0.63616	0.38770	15.591
0.64689	0.39673	15.667
0.65766	0.40610	15.777
0.66815	0.41518	15.925
0.67762	0.42340	16.089
0.68599	0.43047	16.263
0.69660	0.43727	16.580
0.70536	0.44555	16.779
0.71475	0.45388	17.029
0.72456	0.46190	17.333
0.73463	0.47064	17.652
0.74301	0.47806	17.931
0.75347	0.48538	18.363
0.76010	0.49391	18.532
0.77273	0.50394	19.082
0.78195	0.50976	19.561
0.79158	0.52093	19.937
0.80610	0.53253	20.729
0.81366	0.54056	21.112
0.82552	0.54915	21.855
0.93605	0.56068	22.486
0.84558	0.56604	23.217

T (K)	C _p (mJ/gm-atom K)	C _{add} /C _{total} (percent)
0.85781	0.57809	24.017
0.87119	0.59013	24.929
0.88211	0.59972	25.647
0.89481	0.61440	26.281
0.90685	0.62257	26.906
0.92326	0.62735	27.205
0.93469	0.64894	25.057
0.93273	0.64441	25.528
0.94561	0.65236	22.796
0.95949	0.66215	20.025
0.97177	0.67793	18.414
0.98154	0.68663	17.924
0.99510	0.69548	17.779
1.00622	0.70821	17.444
1.01755	0.71543	17.412
1.02774	0.72987	17.248
1.03767	0.73425	17.299
1.04596	0.74459	17.225
1.05557	0.75269	17.229
1.06414	0.75813	17.278
1.06775	0.76942	17.134
1.08294	0.77951	17.243
1.09414	0.78995	17.286
1.10984	0.80647	17.333
1.11896	0.81799	17.338
1.13891	0.83442	17.523
1.14612	0.84455	17.524
1.15276	0.84691	17.647
1.17810	0.87545	17.801
1.20347	0.89688	18.098
1.23066	0.92682	18.318
1.25753	0.95757	18.530
1.27971	0.98423	18.693
1.30420	1.01876	18.803
1.32999	1.03540	19.222
1.34400	1.05822	19.246
1.37721	1.08740	19.679
1.40112	1.10197	20.091
1.43097	1.15394	20.133
1.46213	1.19309	20.416
1.49040	1.21825	20.832
1.52522	1.27061	21.084
1.55557	1.30756	21.446
1.57836	1.33122	21.784
1.60975	1.37786	22.086
1.64153	1.42779	22.353
1.67820	1.48125	22.720
1.71678	1.42741	23.237

T (K)	C _p (mJ/gm-atom K)	C _{add} /C _{total} (percent)
1.66870	1.47460	22.538
1.67429	1.48598	22.557
1.70644	1.53108	22.911
1.75164	1.58643	23.514
1.78474	1.63677	23.853
1.82993	1.70784	24.304
1.86841	1.76915	24.692
1.90614	1.82449	25.132
1.95484	1.89244	25.745
1.98581	1.98414	25.664
2.01394	1.99647	26.289
2.05629	2.06866	26.715
2.10778	2.16061	27.206
2.14379	2.23409	27.476
2.19784	2.32972	28.025
2.24678	2.45407	28.217
2.27420	2.51573	28.402
2.32797	2.61076	28.982
2.37108	2.72754	29.134
2.43613	2.79514	30.218
2.48490	2.93336	30.377
2.41771	2.98041	30.824
2.55256	3.08070	30.945
2.59776	3.26054	30.791
2.64710	3.33287	31.460
2.69792	3.44863	31.883
2.74147	3.59111	31.986
2.80025	3.77694	32.193
2.84914	3.84439	32.878
2.90096	3.07171	32.735
2.96148	4.35247	32.551
3.03108	4.48720	33.327
3.09640	4.68464	33.709
3.14683	4.85628	33.923
3.20003	5.10576	33.860
3.25852	5.35012	33.956
3.31219	5.42247	34.689
3.36632	5.61636	34.926
3.41088	5.79401	35.058
3.28294	5.48759	33.858
3.35691	5.70140	34.408
3.43534	6.05063	34.530
3.51900	6.40448	34.771
3.58431	6.68771	34.957
3.62799	6.85361	35.170
3.66498	6.99972	35.335
3.72542	7.27654	35.486

T (K)	C _p (mJ/gm-atom K)	C _{add} /C _{total} (percent)
3.78726	7.66717	35.333
3.85217	8.01635	35.388
3.91131	8.35289	35.409
3.97743	8.47337	36.140
4.04921	9.05270	35.749
4.14378	9.43880	36.244
4.21598	9.87122	36.299
4.29225	10.74670	35.469
4.36575	10.93316	36.135
4.44387	12.04955	35.009
4.51875	12.11231	35.924
4.54345	12.18877	36.118
4.61772	12.45042	36.638
4.68070	13.50972	35.595
4.73967	13.66871	36.229
4.82531	14.35797	36.069
4.90338	15.05853	35.957
4.99014	15.74388	36.007
5.08586	16.61322	35.928
5.17967	16.97255	36.549
5.27975	18.15245	36.159
5.36282	19.08424	35.949
5.46617	20.18946	35.801
5.58197	20.75400	36.422
5.70275	22.66725	35.667
5.83316	24.24354	35.461
5.93598	25.32451	35.489
6.06768	26.74126	35.525
6.20851	28.58524	35.332
6.26257	29.22352	35.329
6.41972	31.33009	35.166
6.57284	33.93970	34.683
6.73778	36.43037	34.484
6.93886	39.75163	34.158
7.16976	42.39820	34.518
7.41963	47.52108	33.830
7.57011	50.26939	33.660
7.78894	54.58012	33.356
8.02892	57.75258	33.715
8.28409	65.06228	32.721
8.78668	75.12834	32.592
9.00979	79.39002	32.651
9.19700	81.27144	33.180
9.36195	98.80472	32.402
9.53202	92.04109	32.251
9.69195	95.88052	32.191
9.84075	100.35210	31.957
9.99313	105.22605	31.688

T (K)	C _p (mJ/gm-atom K)	C _{add} /C _{total} (percent)
10.14991	108.94838	31.701
10.30458	116.15750	31.063
10.47187	119.27375	31.277
10.63398	123.75342	31.231
10.79506	132.41300	30.510
10.97002	136.70631	30.598
11.14600	143.47172	30.329
11.33406	145.63709	30.803
11.52038	154.93990	30.258
11.73118	165.36864	29.732
11.96196	172.79008	29.711
12.18290	182.61662	29.394
12.41015	191.49462	29.245
12.64571	205.63628	28.619
12.89785	210.22103	29.040
13.13836	225.55263	28.411
13.39122	235.82549	28.333
13.63604	245.85198	28.269
13.87160	258.75055	27.968
14.12254	276.67620	27.375
14.40976	284.27503	27.614
14.67992	295.96518	27.653
14.94238	310.45994	27.434
15.19959	331.10357	26.860
15.46978	344.30786	26.806
15.73666	360.08150	26.619
16.01487	374.80130	26.541
16.30951	384.28230	26.784
16.60239	402.61363	26.587
16.90399	423.62921	26.316
17.21689	444.06190	26.134
17.53790	464.12225	26.015
17.85277	486.56481	25.813
18.16592	514.61614	25.431
18.48308	538.11134	25.267
18.79154	576.35757	24.629
19.13107	606.66065	24.377
19.47787	639.87765	24.096
19.82290	686.43560	23.499