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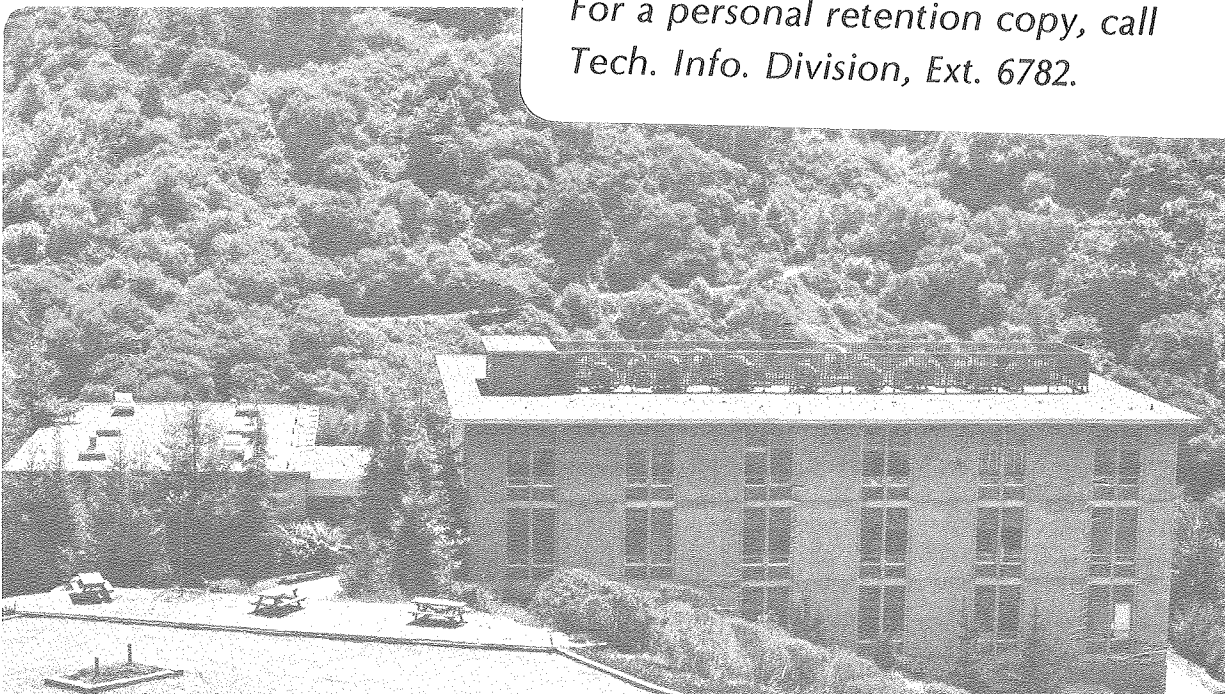
**THERMOELECTRIC GENERATION OF CHARGE IMBALANCE AT
A SUPERCONDUCTOR-NORMAL METAL INTERFACE**

D.J. Van Harlingen

January 1981

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THERMOELECTRIC GENERATION OF CHARGE IMBALANCE
AT A SUPERCONDUCTOR-NORMAL METAL INTERFACE

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The thermoelectric voltage produced across a superconductor-normal metal-superconductor (SNS) sandwich by an applied heat current has been measured in Pb-Cu-PbBi and In-Al-Sn as a function of temperature. The observed divergence of the thermoelectric voltage near T_c is attributed to a charge imbalance region decaying into the superconductor from the NS interface over the quasiparticle diffusion length λ_{Q^*} . The charge imbalance is generated by thermoelectrically driven quasiparticle currents in the superconductor. It contributes a voltage per unit heat power given by $V_S/P = \lambda_{Q^*} S/\kappa A$, where A is the sample cross-sectional area, and S and κ are the thermopower and the thermal conductivity of quasiparticles in the superconductor. For Pb and In, we find the measured thermopower in the superconducting state to be slowly-varying with temperature near T_c and consistent in magnitude with normal state values. This result is in agreement with theoretical predictions of thermoelectric effects in superconductors but contrary to previous experimental results obtained by other methods.

I. INTRODUCTION

The idea that the quasiparticles in a superconductor are driven by a temperature gradient, first proposed by Ginzburg¹, has been widely discussed recently²⁻⁴ and is generally accepted. However, experimental investigations of the thermoelectric effect have yielded conflicting results. Theoretically, it is predicted that the thermoelectric coefficient L_T describing the quasiparticle current $\vec{J}_n = L_T(-\vec{\nabla}T)$ should be continuous with the value of L_T in the normal state at $T=T_C$ and roll-off smoothly to zero as T decreases. Instead, measurements of the thermoelectrically-generated magnetic flux in bimetallic superconducting rings⁴⁻⁶ have suggested that L_T diverges weakly at T_C as $(T_C-T)^{-1/2}$, becoming several orders of magnitude larger than the normal state coefficient. On the other hand, investigations of the polarity asymmetry of the critical current in clean superconducting point contacts with a temperature gradient^{3,7} have observed no thermoelectric current at all.

In this paper, we report the observation of a thermoelectric voltage in excess of the contribution from the normal metal region in superconductor-normal-superconductor (SNS) sandwiches with an applied uniform heat current. We attribute the excess voltage to a charge imbalance region generated by thermoelectric currents in the superconductors. Measurement of the temperature dependence of the excess voltage enables a determination of the thermoelectric coefficient L_T in the superconducting state near the transition temperature. For superconducting Pb and In, we find L_T to be slowly-varying and consistent with expected normal state values at T_C , in agreement with theories of thermoelectricity in superconductors⁸.

II. THEORETICAL DESCRIPTION

We begin by describing the mechanism for the generation of charge imbalance at an NS interface by thermoelectric currents, first suggested by Artemenko and Volkov⁹. A charge imbalance exists in a superconductor whenever the quasiparticle distribution f_k is perturbed from equilibrium in such a way that the net charge per unit volume of the quasiparticles, Q^* is non-zero. In order to conserve total electronic charge, the condensate charge must then compensate by a shift of the pair chemical potential μ_s from its equilibrium value. As discussed by Waldram¹⁰, Pethick and Smith¹¹, and others, the difference in chemical potentials may be related to Q^* by

$$Q^* \equiv \sum_k q_k f_k = -2N(0)e(\mu_s - \mu_n), \quad (1)$$

where $N(0)$ is the single spin electronic density of states and q_k is the charge of the quasiparticle in the state $\vec{k}$. In our notation, q_k varies from an electron charge e for $k \gg k_F$ to a hole charge $-e$ for $k \ll k_F$, where k_F is the Fermi wavevector. The quantity μ_n in Eq. (1) is the quasiparticle chemical potential, which depends only on the number of electrons and the temperature; it should not be confused with the effective potential μ_n of Pethick and Smith¹¹ used to model the non-equilibrium quasiparticle distribution.

Like the conduction electrons in a normal metal, the quasiparticles in a superconductor respond to external fields according to the transport equation

$$\vec{J}_n = \frac{\sigma}{e} (-\vec{\nabla}\phi_n) + L_T(-\vec{\nabla}T), \quad (2)$$

where σ is the electrical conductivity and ϕ_n the electrochemical potential of the quasiparticles. The electrical driving force on the quasiparticles $(-\vec{\nabla}\phi_n) = e\vec{E} - \vec{\nabla}\mu_n$ depends on the electric field $\vec{E}$ and the quasiparticle chemical potential μ_n . On the other hand, in order that the supercurrent is not accelerated, the London equations require that the electrical driving force on the pairs vanish in steady-state so that $(-\vec{\nabla}\phi_s) = e\vec{E} - \vec{\nabla}\mu_s = 0$, where ϕ_s is the pair electrochemical potential. Thus, we may also write Eq. (2) in the form

$$\vec{J}_n = \frac{\sigma}{e} \vec{\nabla}(\mu_s - \mu_n) + L_T(-\vec{\nabla}T) = \frac{\sigma}{2N(0)e^2} (-\vec{\nabla}Q^*) + L_T(-\vec{\nabla}T). \quad (3)$$

We see that a charge imbalance gradient, as well as a temperature gradient, drives a quasiparticle current in a superconductor. In a configuration for which $\vec{\nabla} \times \vec{J}_n = 0$, the Meissner effect exists in a superconductor even in a non-equilibrium state⁴. The normal current $\vec{J}_n$ is then locally cancelled by a counterflow of supercurrent $\vec{J}_s = -\vec{J}_n$ in order to exclude the magnetic field except within a penetration depth of the surface.

The steady-state charge imbalance is determined from the Boltzmann equation by requiring

$$\frac{dQ^*}{dt} = -\vec{\nabla} \cdot \vec{J}_n - \frac{Q^*}{\tau_{Q^*}} = 0. \quad (4)$$

Charge imbalance is created at the rate $\vec{\nabla} \cdot \vec{J}_n$ and decays with the characteristic relaxation time τ_{Q^*} . Combining Eqs.(3) and (4), we obtain differential equations for J_n and Q^* :

$$\vec{J}_n - (\lambda_{Q^*})^2 \vec{\nabla}(\vec{\nabla} \cdot \vec{J}_n) = L_T(-\vec{\nabla}T), \quad (5)$$

$$Q^* - (\lambda_{Q^*})^2 \nabla^2 Q^* = \tau_{Q^*} L_T \nabla^2 T. \quad (6)$$

Here λ_{Q^*} is the usual quasiparticle diffusion length characterizing the spatial decay of charge imbalance given by

$$\lambda_{Q^*} = \left(\frac{\sigma \tau_{Q^*}}{2N(0)e^2} \right)^{1/2} = \left(\frac{1}{3} v_F \ell \tau_{Q^*} \right)^{1/2}, \quad (7)$$

where ℓ is the mean free path and v_F is the Fermi velocity.

We consider a perfect normal metal-superconductor (NS) interface with an applied uniform heat current density $\vec{U} = U\hat{x}$, as shown in Fig. 1(a). We assume that the superconductor is near its transition temperature T_c so that the dominant heat flow is by quasiparticle conduction; the temperature gradients in N and S are then each uniform (although not necessarily equal). In the normal metal, under open circuit conditions a uniform electrochemical potential gradient $(-\vec{\nabla}\phi) = eS_N(\vec{\nabla}T)_N$ is established preventing any electric current flow. This is the usual thermoelectric voltage which depends on the normal metal thermopower $S_N \equiv (L_T)_N/\sigma_N$, and the temperature gradient $(\vec{\nabla}T)_N$ in N. The currents and fields in the superconductor are obtained by solving Eqs. (5) and (6) in one-dimension, requiring $\vec{J}_n = \vec{J}_s = 0$ at the interface ($x=0$) and noting that in this case $\nabla^2 T = 0$. We find

$$\vec{J}_n(x) = L_T(-\vec{\nabla}T) (1 - e^{-x/\lambda_{Q^*}}), \quad (8)$$

$$Q^*(x) = 2N(0)e^2 \lambda_{Q^*} S \nabla T e^{-x/\lambda_{Q^*}}, \quad (9)$$

where $S = L_T/\sigma$ is the corresponding thermopower of the superconductor. The quasiparticle current, plotted in Fig. 1(b), has the value $L_T(-\vec{\nabla}T)$ well inside the superconductor and falls to zero at the interface over the length λ_{Q^*} . The current reduction is caused by the build-up of charge imbalance near the interface, decaying exponentially into the superconductor with the same length λ_{Q^*} as shown in Fig. 1(c). It should be emphasized again

that the quasiparticle current is accompanied by an equal and opposite supercurrent so that the net current is zero everywhere.

A charge imbalance proportional to $\vec{\nabla}T$ is thus generated near an NS interface by thermoelectric currents when heat flows into the superconductor. By symmetry, heat extraction creates an imbalance of the opposite sign. We should note that the interaction of the superfluid counterflow with the temperature gradient also produces a uniform charge imbalance in the superconductor proportional to $(\nabla T)^2$, as discussed by Sacks¹², Entin-Wohlman and Orbach¹³, and looked for by Falco¹⁴. Because of coupling between charge and energy modes, the charge imbalance created by this mechanism scales with the elastic scattering time τ_{imp} rather than with the charge imbalance relaxation time $\tau_{Q^*} \gg \tau_{imp}$ of Eq. (4), and hence is much smaller than the effect considered here. As is clear from Eq. (6), a non-uniform temperature gradient ($\nabla^2 T \neq 0$) can also create an additional Q^* , an effect which has been commented on by Tinkham¹⁵. In particular, at the interface of a superconductor and an insulator (or another superconductor far below its transition temperature), heat enters the superconductor as phonons and is transferred to the quasiparticle distribution over a phonon diffusion length, λ_{ph} . For $x \lesssim \lambda_{ph}$, $\nabla^2 T \neq 0$ and the expressions for $\vec{J}_n$ and Q^* are modified slightly from those derived for the NS interface. Since $\lambda_{ph} \ll \lambda_{Q^*}$, the difference is negligible unless heat is deliberately injected or extracted along the length of the superconductor.

Previous investigations of thermoelectric effects in superconductors have relied on discriminating between the counterbalancing normal and superfluid components of the current in the bulk of a superconductor. In this experiment, we detect the charge imbalance thermoelectrically generated

near an NS interface. Consider a normal metal sample sandwiched between two superconductors, S and S', as illustrated in Fig. 2. We assume that $T \lesssim T_{CS} \ll T_{CS'}$, so that we may ignore any non-equilibrium effects at the NS' interface.

We plot the spatial variations of the electrochemical potentials in the presence of an applied heat current density U . In the normal region, the potential varies linearly due to the uniform thermoelectric field. In S', far below its transition temperature, $\phi_n = \phi_s$ is constant. However, in S, from Eq. (9), $\phi_n = Q^*/2N(0)e$ decays exponentially from the NS interface with characteristic length λ_{Q^*} while ϕ_s drops to zero within a coherence length of the interface. The difference in potentials results from the charge imbalance in S.

The voltage across the SNS' sandwich is the sum of the thermoelectric voltage of the normal region

$$V_N = L_N S_N (\nabla T)_N, \quad (10)$$

where L_N is the thickness, and a contribution from the charge imbalance region in the superconductor given by Eq. (9):

$$V_S = \frac{Q^*(0)}{2N(0)e} = \lambda_{Q^*} S \nabla T. \quad (11)$$

Since the total heat flow P through the sample is uniform, we may write the voltage in the additive form

$$V = \left[\left(\frac{L_N}{A} \right) \frac{S_N}{\kappa_N} + \left(\frac{\lambda_{Q^*}}{A} \right) \frac{S}{\kappa} \right] P, \quad (12)$$

where A is the cross-sectional area and $\kappa_N = P/A(\nabla T)_N$ and $\kappa = P/A\nabla T$ are the thermal conductivities of N and S respectively. The effect of the charge imbalance is to add to the thermoelectric voltage an amount equivalent to a length λ_{Q^*} of a normal metal characterized by the coefficients S and κ .

In the absence of pair-breaking mechanisms, the charge imbalance relaxation time near T_c is given by ^{11,16}

$$\tau_{Q^*} = \left(\frac{4k_B T_c}{\pi \Delta} \right) \tau_E = \left(\frac{0.73k_B T_c}{\Delta(0)} \right) \tau_E (1-t)^{-1/2}, \quad (13)$$

where τ_E is the inelastic electron-phonon scattering time at the Fermi level for $T=T_c$. The second form is obtained by assuming a BCS superconductor for which $\Delta(t) \sim 1.74\Delta(0)(1-t)^{1/2}$. Thus, according to Eq. (7), λ_{Q^*} diverges weakly as $(1-t)^{-1/2}$ near T_c . This divergence provides a signature for separating out the thermoelectric charge imbalance contribution from the total measured thermoelectric voltage, allowing determination of the transport coefficients in the superconducting state.

It is interesting to compare Eq. (12) with the expression for the voltage generated in the SNS' sandwich by an applied electric current I , derived and experimentally verified by Hsiang and Clarke¹⁷

$$V = \left[\left(\frac{L_N}{A} \right) \rho_N + \left(\frac{\lambda_{Q^*}}{A} \right) \rho Z(t) \right] I. \quad (14)$$

Here ρ_N and ρ are the electrical resistivities of the normal metal and of the quasiparticles in the superconductor respectively, and $Z(t)$ is a universal function of reduced temperature equal to unity at $t=1$ and falling off rapidly as t decreases.¹⁸ Charge flow across the NS interface produces a charge imbalance that decays as in Fig. 1(c) and adds resistance equivalent to a section of length λ_{Q^*} of the superconductor being normal. The factor $Z(t)$ expresses the fraction of the total current I that enters S as quasiparticle current, the rest entering as supercurrent via Andreev reflection processes^{10,19} at the NS interface. In the case considered here of an applied heat current, the excess quasiparticle charge responsible for the charge imbalance is

injected into the region near the NS interface by the thermoelectric currents inside the superconductor rather than by any charge flow across the interface. Ignoring variations of the energy gap of the superconductor with temperature, there is no energy threshold for quasiparticle injection as in Andreev reflection, and the factor $Z(t)$ is absent in the thermoelectric voltage, Eq. (12).

III. EXPERIMENTAL RESULTS

We have measured the thermoelectric voltage across SNS' sandwiches composed of Pb-Cu-PbBi and In-Al-Sn. The component metals are selected to have low mutual solubilities so that sharp NS interfaces are formed instead of broad intermetallic alloy regions. Two different superconductors are used in each sandwich, with $T_{CS} < T_{CS}'$, so that the divergent contribution of only the NS interface to the thermoelectric voltage will be detected. This enables determination of the thermoelectric coefficient of the S components, Pb and In.

The samples were formed by casting 3 mm dia., 2 mm thick discs of superconductor onto both sides of a thin normal metal plate. The Cu plates, 0.2 mm thick, were spark cut from 99.999% pure polycrystalline copper and oxygen-annealed to remove paramagnetic impurities that elevate the normal state thermopower²⁰. Following chemical polishing to remove surface imperfections and oxides, the Pb and PbBi (eutectic) were melted onto opposite sides into a pyrex-bead mold. The In-Al-Sn sandwiches were formed in a similar manner, using 0.5 mm thick Al cut from 99.999% pure, nominally RRR=10,000 aluminum stock.

Figure 3 shows the experimental configuration. The sample was mounted inside a vacuum can, soldered to a copper block that was connected

to the ^4He bath through a weak thermal link. A heater on the block enabled the sample temperature to be elevated above bath temperature and electronically regulated, while a heater at the isolated end provided the heat flow through the sample. The temperature of the NS interface was measured by a Ge thermometer placed on the normal metal; the Andreev thermal boundary resistance¹⁹ between N and S is negligible near T_c . The sample was connected in a potentiometric circuit using an rf SQUID as a null detector. By measuring the current I_s applied to the calibrated standard resistor $R_s \approx 3 \times 10^{-6}$, we determined the thermoelectric voltage across the composite sample with a resolution of 10^{-14}V . The resistance of the SNS' sandwich could also be measured in this configuration by applying a current I through the sample.

In Fig. 4(a) we plot the measured resistance of a Pb-Cu-PbBi sandwich as a function of temperature. At low temperatures, the resistance is slowly varying, but as T is increased, R exhibits a divergence at the Pb transition temperature, $T_c = 7.2\text{K}$. We attribute the divergence to the excess resistance of the charge imbalance region in the Pb. To extract the contribution from the superconductor, R_s , we first fit the resistance to a polynomial below 4K, over which range the charge imbalance contribution is negligible. Since the Cu is expected to be in the residual resistance regime at these temperatures, the appreciable temperature dependence of the resistance indicates that the NS and NS' interfaces are not perfectly metallic²¹, probably containing patches of oxides. The excess resistance above the parameterized fit, extrapolated to higher temperatures as shown in Fig. 4(a), is identified with R_s .

Theoretically, R_s is given by the second term in brackets in Eq. (14). Using Eqs. (7) and (13), we may separate out the explicit temperature dependence in the expression

$$R_s = \frac{1}{A} \left(\frac{0.73 k_B T_c}{2N(0)e^2 \Delta(0)} \right)^{\frac{1}{2}} (\rho \tau_E)^{\frac{1}{2}} [Z(t)(1-t)^{-\frac{1}{4}}]. \quad (15)$$

In this equation, all of the parameters except ρ may be directly measured or obtained from the literature. Hence, in Fig. 4(b) we plot R_s vs. $[Z(t)(1-t)^{-\frac{1}{4}}]$; the inset shows the universal function $Z(t)$. From the slope, we find $\rho = 3.1 \times 10^{-8} \Omega\text{-cm}$, using the calculated value²² of $\tau_E = 2.3 \times 10^{-11}$ sec. This corresponds to a quasiparticle diffusion length given by $\lambda_{Q*} = \lambda_{Q*}(0)(1-t)^{-\frac{1}{4}}$, where $\lambda_{Q*}(0) = 2.7 \mu\text{m}$.

In Fig. 5(a), we plot the thermoelectric voltage per unit power across the Pb-Cu-PbBi sandwich vs. temperature. The negative bump in the thermoelectric voltage centered at 4K is characteristic of the thermopower of Cu²⁰; the bump arises from the Kondo effect caused by magnetic impurities. Near T_c , however, we observe a negative divergence of the measured voltage, evidence for charge imbalance generation by the thermoelectric currents in the Pb. By subtracting the extrapolated curve drawn in Fig. 5(a), we again separate the contribution attributed to the superconductor, V_s , which we identify with the second term of Eq. (12).

In order to isolate the explicit temperature dependence of λ_{Q*} , we may write

$$V_{S/P} = \left(\frac{\lambda_{Q*}(0)}{A} \right) \frac{S}{\kappa} [(1-t)^{-\frac{1}{4}}]. \quad (16)$$

We use the value of $\lambda_{Q*}(0) = 2.7 \mu\text{m}$ obtained from the SNS' resistance, and estimate the thermal conductivity of the Pb from the measured electrical resistivity assuming the Wiedemann-Franz law, $\kappa/\sigma T = L_0$, where L_0 is the Lorenz number ($L_0 = 2.45 \times 10^{-8} \text{V}^2/\text{K}^2$). The only unknown coefficient is the thermoelectric power of the Pb, S , which we determine by plotting V_S/P vs. $(1-t)^{-1/2}$ in Fig. 5(b). It is important to note that the proportionality of V_S/P to the expected temperature variation of λ_{Q*} indicates that S is itself not strongly temperature dependent. A divergence of $S \sim (1-t)^{-1/2}$ in the superconducting state had been suggested by bimetallic ring experiments.⁴ From the slope of the curve in Fig. 5(b), we deduce a value of $S = -7.8 \times 10^{-7} \text{V/K}$ near T_c .

In our configuration, we were unable to measure the normal state thermopower of the Pb section of the sample directly. However, pure Pb in the normal state²³ has a thermopower $S_N = -2.2 \times 10^{-7} \text{V/K}$, within a factor of 4 of our measured value. Since the addition of even small amounts of impurities may change the magnitude of the thermopower by as much as an order of magnitude, the results are consistent with the thermopower of the normal and superconducting states being continuous at T_c . Certainly, the coefficient measured by this technique does not deviate by several orders of magnitude (smaller or larger) from the expected normal state value, as has been observed previously.

Measurements on In-Al-Sn sandwiches near the In transition (3.4K) yield similar results. In Fig. 6, we plot the resistance R and thermoelectric voltage V/P vs. T for an In-Al-Sn sample. From the divergence of R near T_c we obtain $\rho = 9.8 \times 10^{-7} \Omega\text{-cm}$, which gives a diffusion length $\lambda_{Q*}(0) = 1.2 \mu\text{m}$,

assuming the calculated value²² $\tau_E = 1.0 \times 10^{-10}$ sec. for In. Despite τ_E being longer in In than in Pb, we obtain a shorter λ_{Q*} due to the higher resistivity. This we presume to be caused by substantial alloying of the Al into In. The divergence of V/P is evident only very near T_C , arising from the nearly linear normal metal thermopower of the Al. The excess thermoelectric voltage is found to increase roughly as $(1-t)^{-1/4}$, suggesting a slowly-varying superconducting state thermopower $S = -1.1 \times 10^{-7}$ V/K. This is in good agreement with measured values⁴ of the thermopower for normal state indium at T_C ($\sim 4 \times 10^{-7}$ to 3×10^{-8} V/K).

IV. CONCLUSION

We have observed the charge imbalance generated at NS interfaces by thermoelectrically-driven quasiparticle currents in superconducting Pb and In. The charge imbalance, detected as an excess voltage in the presence of an applied heat current through the sample, exhibits the temperature variation of $\lambda_{Q*} \sim (1-t)^{-1/4}$ near T_C . We conclude, therefore, that the thermoelectric coefficient L_T (or S) in the superconducting state is slowly-varying over the range of our measurement, $T = 0.9$ to $0.999 T_C$. The measured magnitudes in Pb and In are in reasonable agreement with typical normal state thermopower values for the metals at their transition temperatures.

Further experiments are being conducted to eliminate some of the uncertainties in the measurements. First of all, it is necessary to fabricate better NS interfaces, eliminating oxide barriers and reducing alloying. Secondly, because of the extreme sensitivity of the thermopower of metals to impurities and defects, it is essential to make a direct measurement of the normal state thermopower in the sample used so that a direct comparison between the normal and superconducting state values can be made.

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FIGURE CAPTIONS

- Fig. 1 (a) At a normal metal-superconductor interface, an applied heat current $\vec{U}$ generates an electrochemical potential gradient $\vec{\nabla}\phi_n$ in the normal metal and counterflowing normal and supercurrents $\vec{J}_n = -\vec{J}_s$ in the superconductor. (b) Spatial variation of quasiparticle current near the NS interface. (c) Spatial decay of the thermoelectrically-generated charge imbalance Q^* .
- Fig. 2 Spatial variations of the pair (ϕ_s) and quasiparticle (ϕ_n) electrochemical potentials in the SNS' sandwich shown at the top. The measured voltage V includes the normal thermoelectric voltage V_N plus the charge imbalance contribution V_S . Here, $T_{CS} \ll T_{CS}'$.
- Fig. 3 Schematic drawing of the experimental configuration used to measure the thermopower and resistance of SNS' sandwiches.
- Fig. 4 (a) The temperature dependence of the resistance R of a Pb-Cu-PbBi sandwich showing the divergence at $T_c = 7.2K$. The solid line is a polynomial fit to the region below 4K. (b) The contribution to the resistance from the charge imbalance region R_S vs. $Z(t)(1-t)^{-1/4}$, where $Z(t)$ is shown in the inset.
- Fig. 5 (a) Plot of the measured thermoelectric voltage per unit power V/P vs. T for a Pb-Cu-PbBi sample. The solid curve is characteristic of the normal thermopower of Cu. (b) Fit of the charge imbalance contribution V_S/P to the temperature dependence of $\lambda_{Q^*} \sim (1-t)^{-1/4}$.
- Fig. 6 (a) The temperature dependence of the resistance R vs. T for an In-Al-Sn sample. (b) Thermoelectric voltage vs. T for the same sample. Both exhibit a divergence at the indium transition temperature $T_c = 3.4K$.

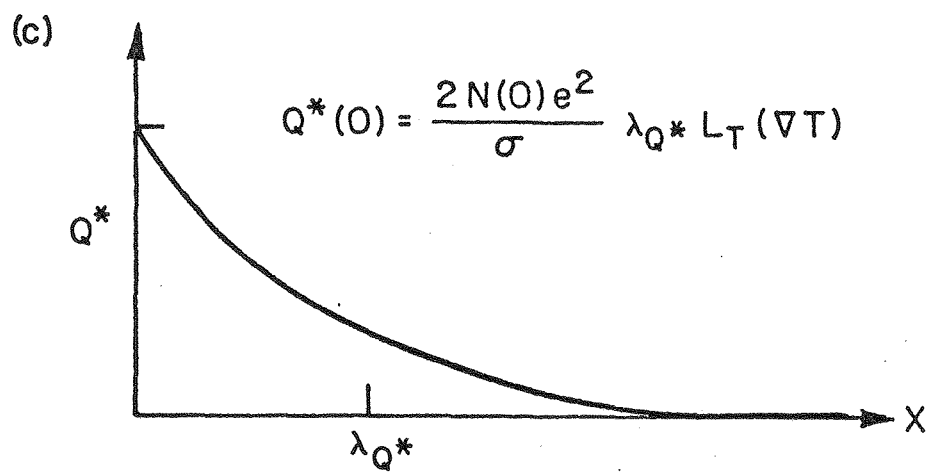
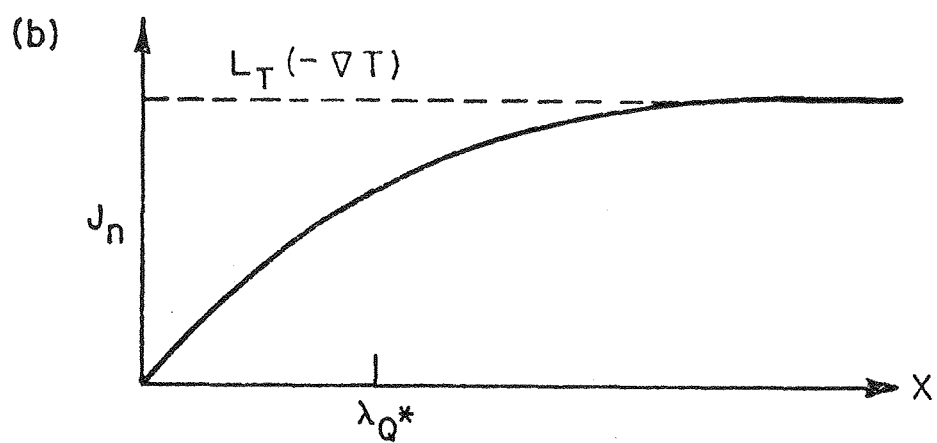
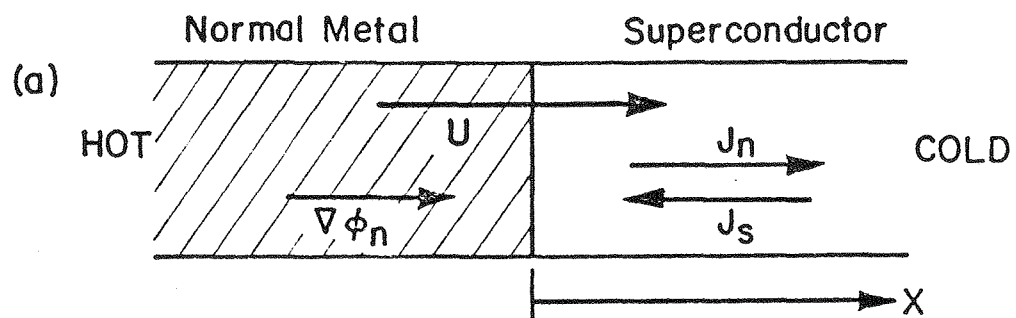


Fig. 1

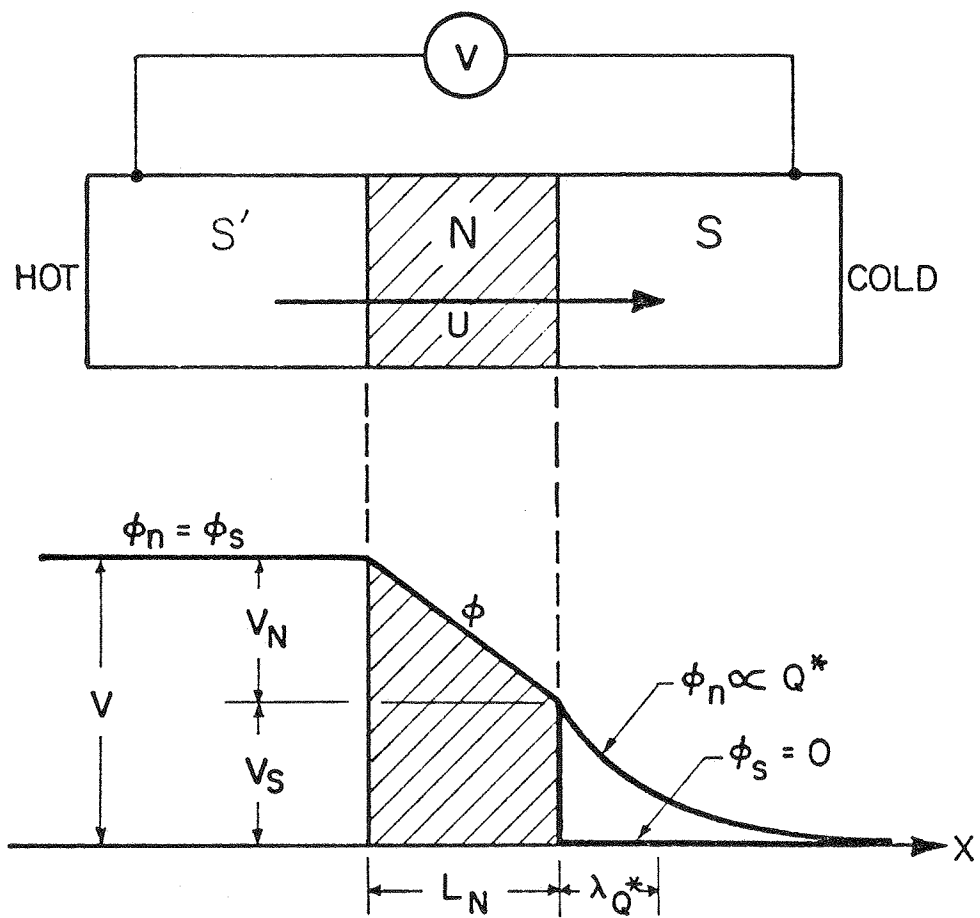


Fig. 2

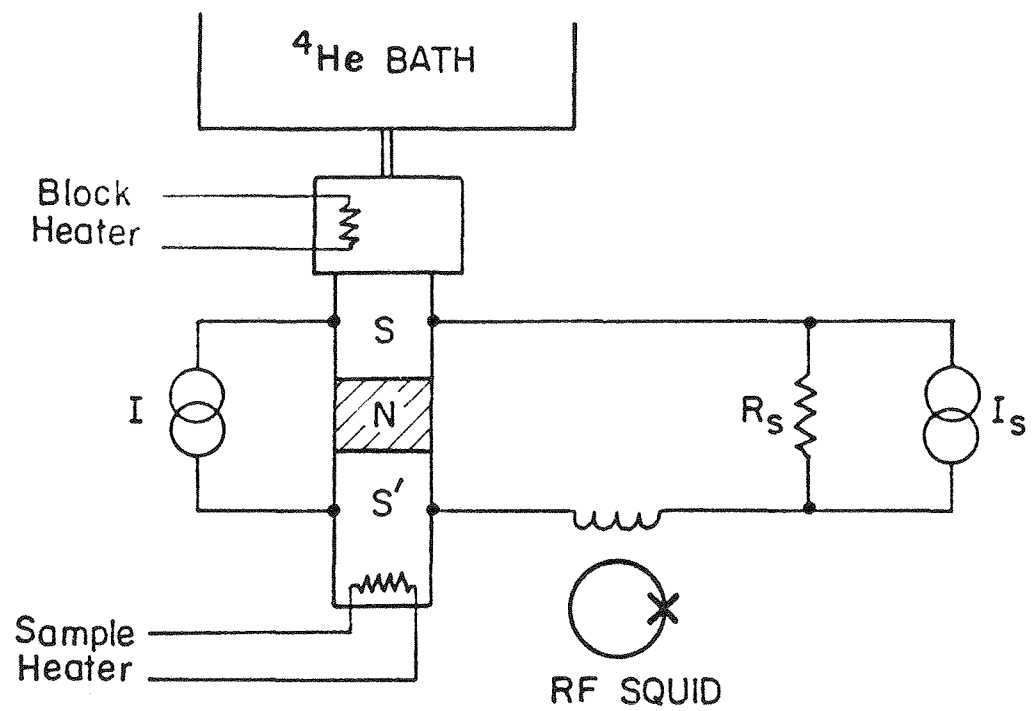


Fig. 3

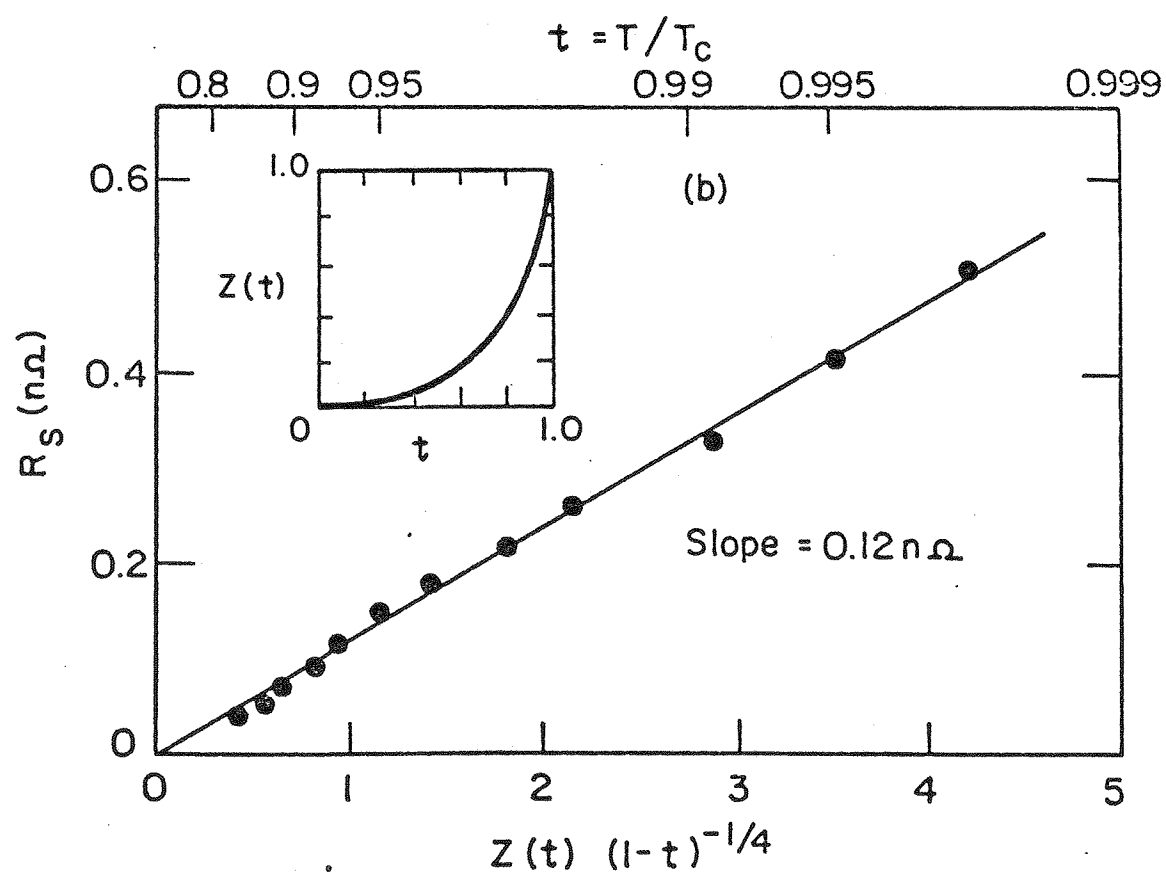
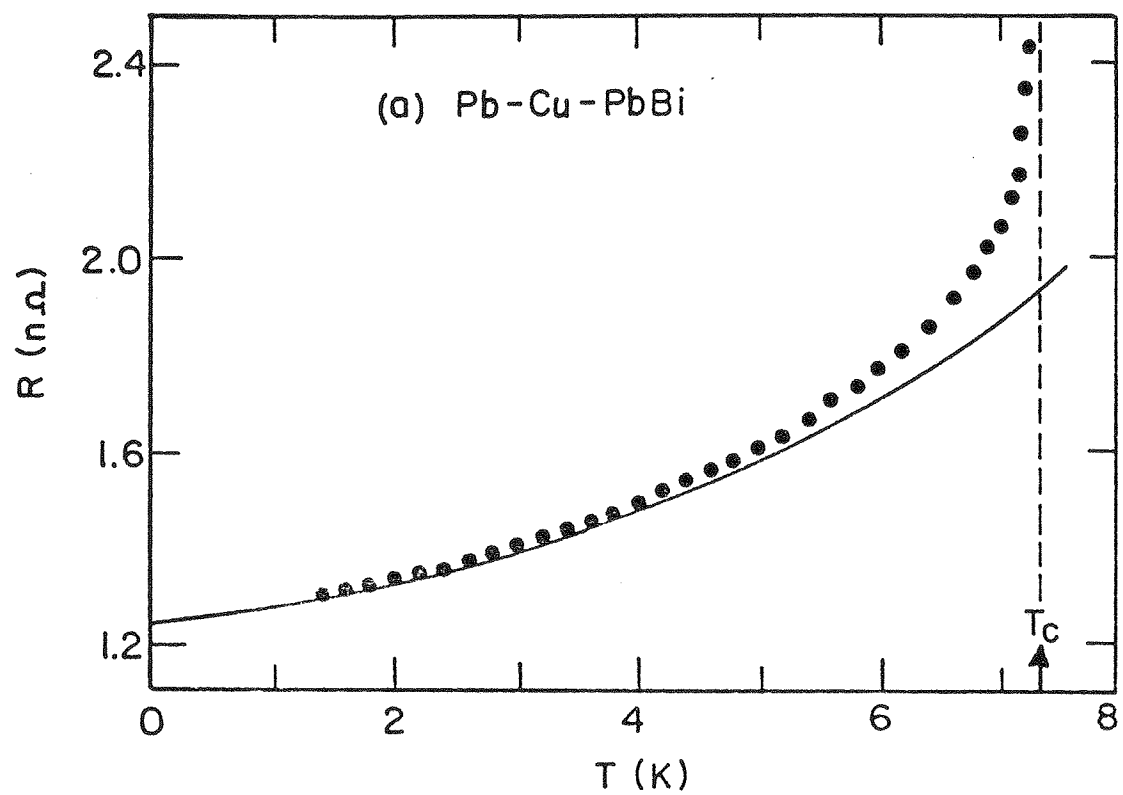


Fig. 4

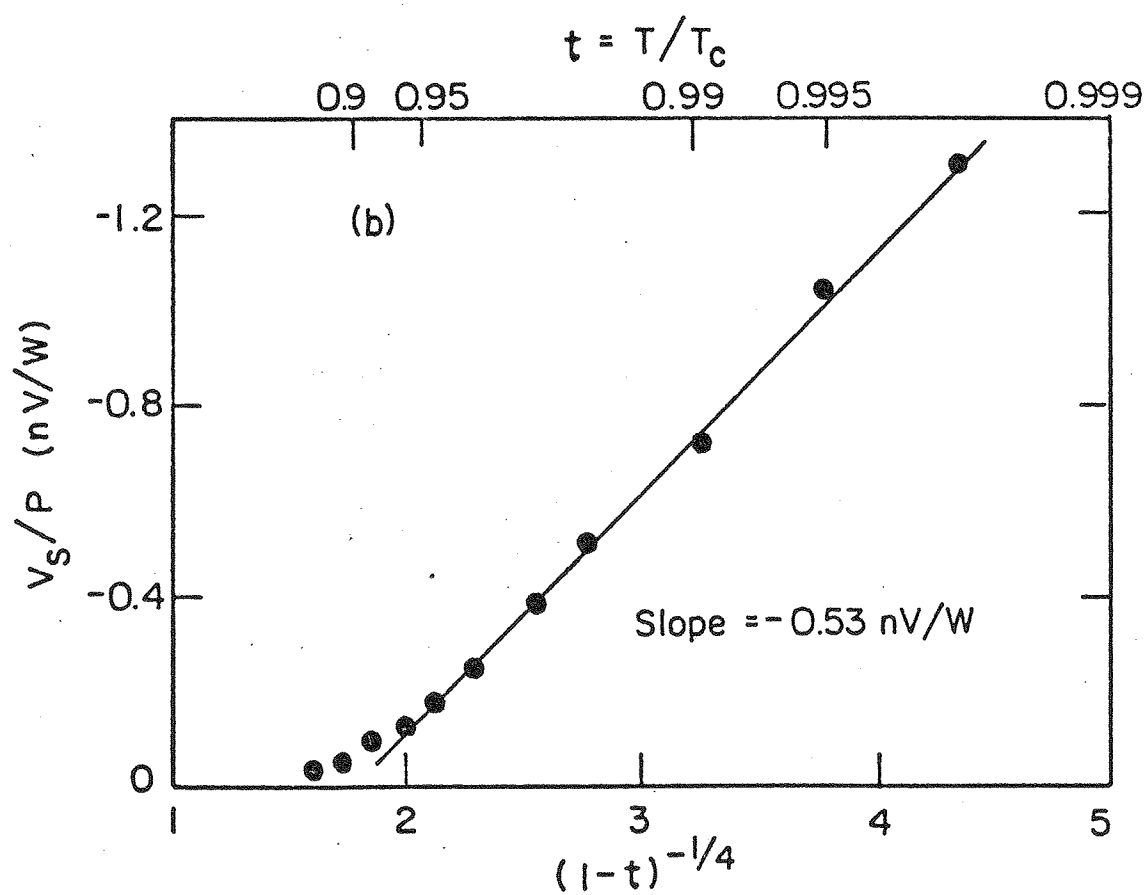
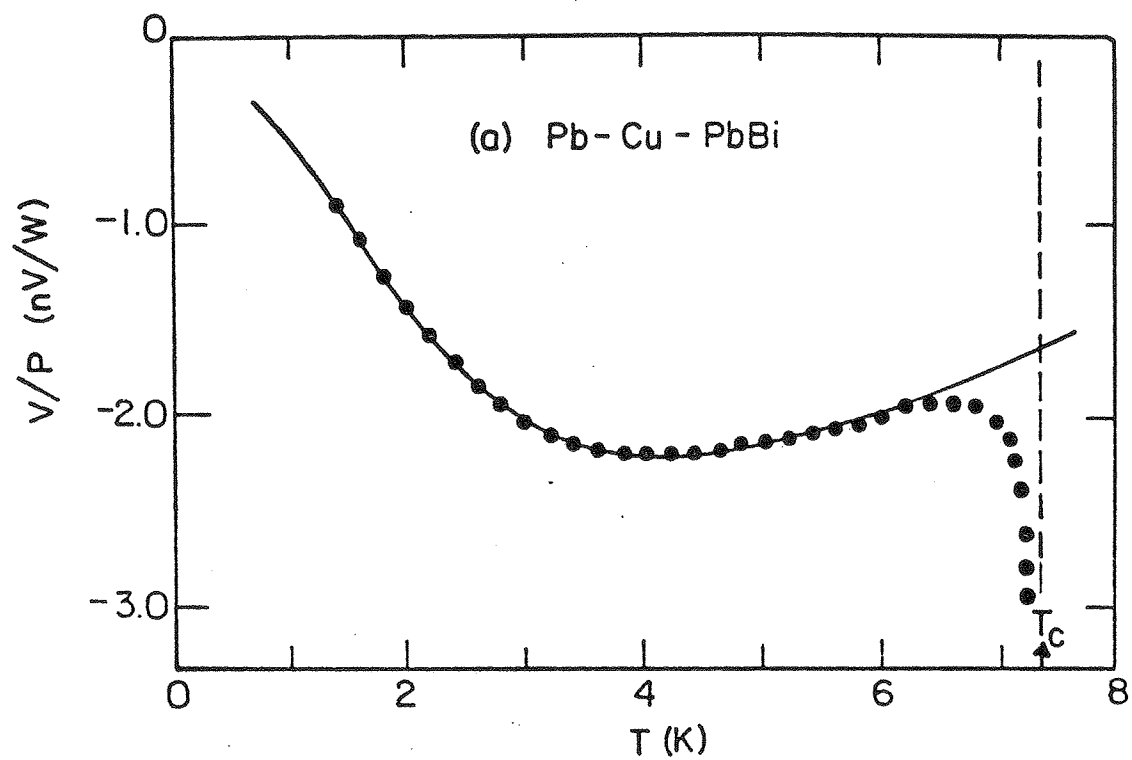


Fig. 5

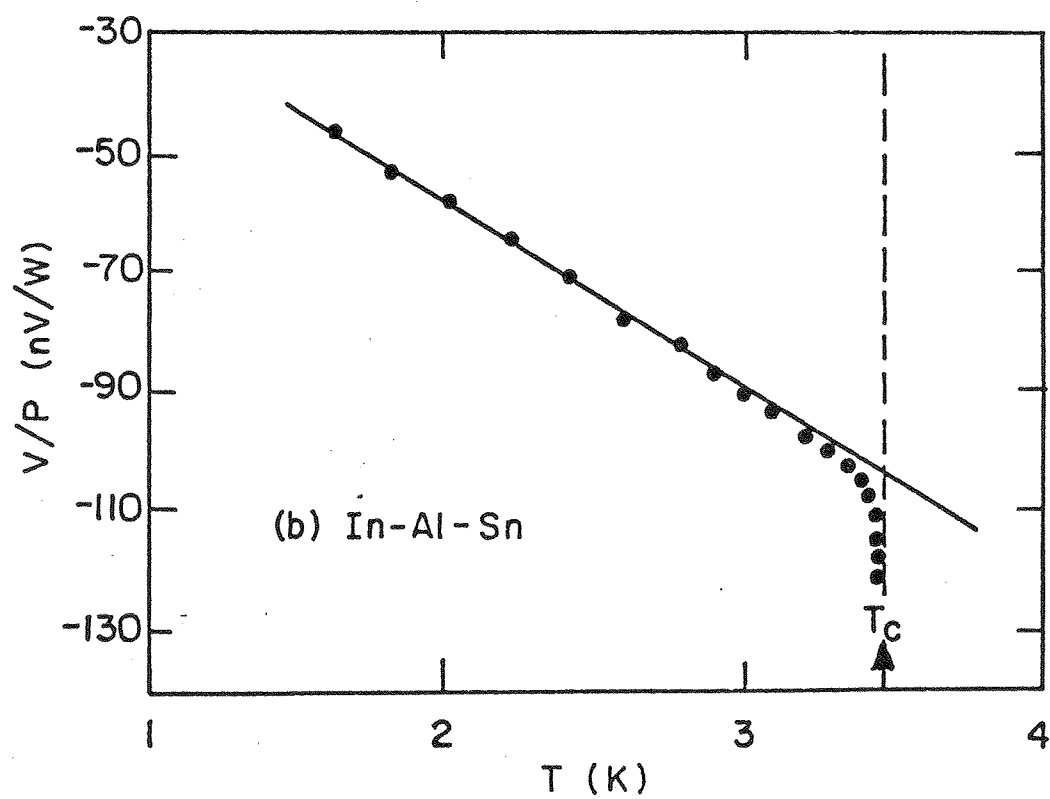
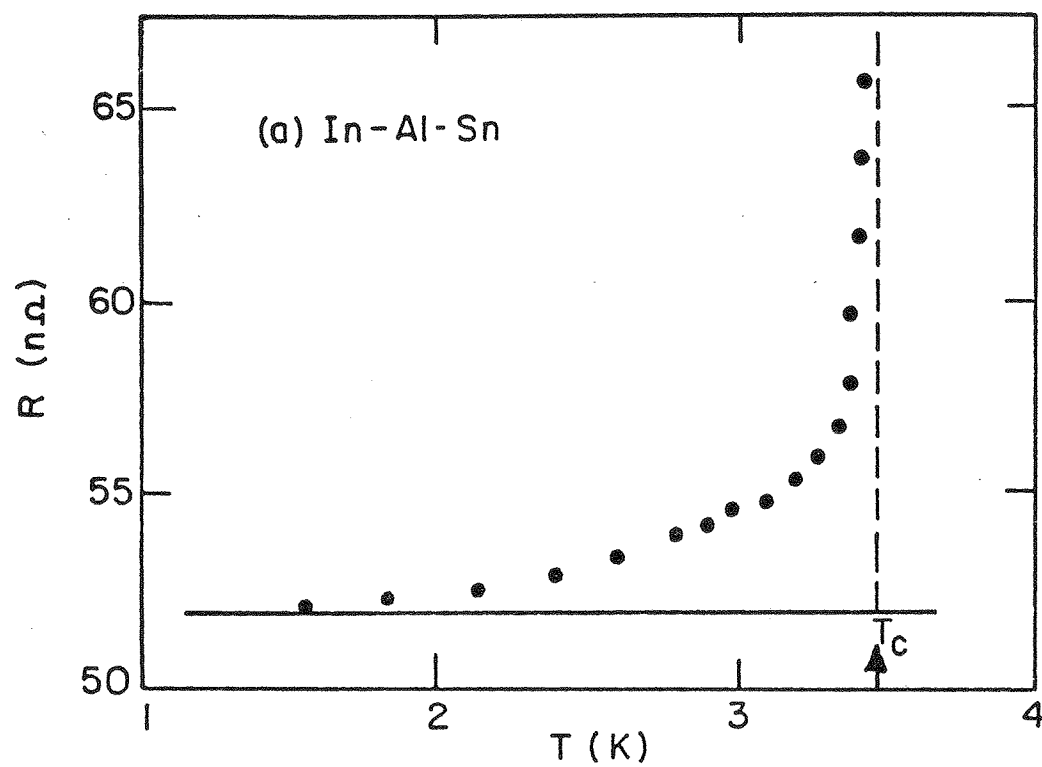


Fig. 6

