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Pair Tunneling as a Probe of Order Parameter Fluctuations in Superconductors: Zero Magnetic Field Effects*

J. T. Anderson+, R. V. Carlson++ and A. M. Goldman
School of Physics and Astronomy
University of Minnesota
Minneapolis, Minnesota 55455

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+ Present address: W. W . Hansen Laboratories of Physics, Stanford University, Stanford, California.

++ NSF Trainee

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ABSTRACT

Thermal fluctuations of the order parameter in a superconductor above its transition temperature result in manifestations of the ordered state which appears below the transition. In a Josephson junction consisting of two metals with different transition temperatures there is associated with the incipient superconductivity an enhanced conductivity across the junction at temperatures between the transition temperatures of the two metals. The enhanced conductivity is observed as a current which flows in excess of the junction quasiparticle current. The excess current has been investigated using $Sn-Sn_xO_y-Pb$ and $Al-Al_xO_y-Pb$ junctions masked to eliminate effects of graded film edges. Details of the variation of the excess current with voltage and temperature are in quantitative agreement with calculations in the $Al-Al_xO_y-Pb$ junctions. The excess current-voltage characteristic is a direct measure of the imaginary part of the generalized susceptibility above T_c. The pair relaxation frequency in aluminum based on the data is consistent with the theoretical value.

I. Introduction

The study of the onset of the superconducting state has attracted considerable experimental and theoretical interest since it was realized that the rounding of the resistive transition in disordered films could be associated with intrinsic features of the transition rather than with sample inhomogeneities of a mechanical or chemical nature. Extensive investigations of precursive electrical conductivity[1], diamagnetic susceptibility[2], and tunneling density of states[3] have now fully confirmed the observability of the effects of order parameter fluctuations in superconductors.[4] The local time-dependent Ginzburg-Landau equation is believed to provide an essentially correct description of local superconductivity above T_c. Where deviations from theory are observed, they involve the quantitative sizes of coefficients which can be ultimately explained by microscopic theory[5], or as in the case of high-temperature precursive diamagnetism can be explained by non-local generalization of the theory.[6]

The previously mentioned experiments, although serving as convincing demonstration of the existence of precursive behavior due to order parameter fluctuations, cannot be used to critically test the theory. The most critical way to probe the fluctuations associated with a phase transition is to measure the wave-number and frequency-dependent generalized susceptibility associated with the transition. This quantity is proportional to the spectrum of the fluctuations and can be calculated directly from theoretical models of the transition. Electrical conductivity and diamagnetic susceptibility involve on the other hand, complicated convolutions of the generalized susceptibility. Because of the quantum mechanical nature of the order parameter in a superconductor and the resultant absence of a classical field thermodynamically conjugate to the order parameter, direct determination of the generalized susceptibility had been thought to be impossible. Recent theoretical[7,8] and experimental work[9] has shown that the generalized susceptibility can indeed be measured by a simple dc tunneling experiment in which the susceptibility is proportional to an excess-current due to pair tunneling in the I-V characteristic of a junction consisting of the metal of interest just above its transition temperature and a second, fully superconducting metal.

Ferrell[7] was the first to suggest that there might be a connection between pair tunneling and fluctuations and that the generalized susceptibility might be experimentally determined. He showed that the frequency-dependent conductivity of a bimetallic junction could be used to determine $\chi(\omega)$, the required field being provided by the non-zero pair amplitude, or order parameter of the superconducting side. Scalapino[8] subsequently showed that the dc I-V characteristic itself was a direct measure of $\chi(q,\omega)$, the wave-number and frequency dependent generalized susceptibility. Equivalent results have been obtained by Takayama.[10] Other theoretical contributions have been made by Kulik[11] and by Tan[12], Kulik having considered the problem of fluctuation pair tunneling between identical normal metals above T_c, and Tan having carried out a calculation of the magnetic field dependence of the excess current.

Measurements in qualitative agreement with Ferrell's calculation have been reported by several groups.[13,14] Semiquantitative studies of $\chi(q,\omega)$ substantially in agreement with Scalapino's calculations have been reported in studies of Pb-Sn_xO_y-Sn junctions.[9]

In the following, we present experimental studies of lead-aluminum junctions which are in quantitative agreement with theory at temperatures above the transition temperature of aluminum. Detailed consideration will be given only to the case of effects in zero magnetic field. An analysis of finite field data will be presented in a later paper. Section II of this paper is devoted to a discussion of the theory of the pair tunneling current based on the time-dependent Ginzburg-Landau equation. Section III contains experimental details. Section IV is devoted to presentation of data and comparison with theory. Discrepancies between experiment and theory are discussed in the last section.

II. Theory

We consider a tunneling junction consisting of two different superconductors at a temperature T such that $T_c < T < T_c'$, where T_c and T_c' are the transition temperatures of the superconductors. Following Ferrell[7] and Scalapino[8], the coupling energy between the superconducting and the normal halves of the junction can be interpreted as an effective Hamiltonian of the form

$$H_I = -\bar{C}e^{-i\omega t} \int d^2r e^{i\vec{q}\cdot\vec{r}} \Delta(\vec{r},t) + h{\cdot}c. \tag{1}$$

In this equation $\Delta(\vec{r},t)$, the pair field operator of the low transition temperature superconductor, is assumed to have zero average value. The corresponding quantity for the high temperature side of the junction is assumed to have a well-defined average value and negligible fluctuations. Its phase is treated as a fixed reference for the phase of the order parameter fluctuations in the normal metal. In equation (1), ω is related to the voltage bias across the junction through the Josephson relation, $\hbar\omega = 2eV$ and q, the wave-vector is determined by the relation

$$q = \frac{2eH}{\hbar c}(\lambda' + d/2), \tag{2}$$

where H is the value of a magnetic field applied parallel to the plane of the junction, d is the thickness of the normal half of the junction and λ' is the magnetic penetration length of the superconducting half. It is assumed that the superconducting half is thicker than λ' and that magnetic fields penetrate the normal metal completely. The constant $\bar{C}$ is of the form

$$\bar{C} = \frac{\hbar}{e^2}(R_N A)^{-1} \ln [4T_c'/T_c] \tag{3}$$

where R_N is the normal state tunneling resistance, and A is the area of the plane of the junction perpendicular to the direction of current flow. The integral in equation (1) is taken over the same plane. An additional assumption is that the normal film thickness is smaller than the temperature dependent coherence length $\xi(T)$.

Equation (1) is formally similar to the Hamiltonian which describes the coupling of a magnetic system to an external field.[15] For magnetic systems the susceptibility is often determined experimentally by inserting a specimen in a resonant microwave or radio frequency structure and measuring the power absorbed by the material. The latter is related to the imaginary part of the susceptibility and the strength of the radio frequency field by an expression of the form

$$P = 2\omega\chi'' H_1^{\,2}, \tag{4}$$

where P is the power absorbed, χ'' is the imaginary part of the susceptibility and H_1 is the strength of the radio frequency magnetic field. The experimental determination of the suceptibility of a superconductor by tunneling is formally identical to the determination of the magnetic susceptibility. Equation (1) can be interpreted as describing the coupling of the order parameter fluctuations in the normal metal with a spatially and temporally varying field originating in the superconducting half of the junction and coupled across the oxide layer by the tunneling interaction. The time rate of change of the internal energy of the normal metal due to this coupling can then be calculated using standard linear response techniques, yielding a result formally identical to (4) with the replacement of the magnetic field by $\bar{C}$ and the magnetic susceptibility by the generalized susceptibility for a superconductor, the pair-field susceptibility. In the magnetic case the power absorbed by the sample is supplied by the radio frequency field and detected by changes in the Q of the resonant circuits. In contrast, the power supplied in the tunneling experiment is provided by the dc current source in the external circuit, and is simply the product of I_1V, where I_1 is the average pair tunneling current and V is the dc voltage across the junction. The total measured tunneling current is the sum of the quasiparticle current and I_1.

To calculate I_1, in analogy with the magnetic case, I_1V is set equal to the time rate of change of the internal energy of the normal metal, where the latter is computed using linear response theory and equation (1). In carrying out the calculation, it is convenient to work with ψ_{op}, an operator corresponding to the order parameter ψ of the Ginzburg-Landau theory.

The effective Hamiltonian is then

$$H_I = -\int d^2 r F(r,t)\psi_{op}(r,t) + \hbar.c., \tag{5}$$

where

$$F = \bar{C}\, e^{i(\vec{q}\cdot\vec{r}-\omega t)}\, [N(0)/\alpha(T)]^{-1/2}, \tag{6}$$

and

$$\psi_{op}(\vec{r},t) = [N(0)/\alpha]^{1/2}\Delta(r,t) \tag{7}$$

In the above expressions $\alpha(T)=\hbar^2/2m\xi^2(T)$ is the coefficient of the quadratic term of the Ginzburg-Landau free energy functional[16], and N(0) is the single electron density of states. $\xi(T)$ is the usual temperature dependent coherence length.

The time rate of change of the energy in the normal metal is the time derivative of the thermal average of the internal energy in the normal metal. The calculations must be carried out to second order in H_I to obtain non-zero results. The internal energy is then

$$\langle E(t)\rangle = Tr(\rho_2 H_0) + Tr(\rho_1 H_I), \tag{8}$$

where $\rho_1(t)$ and $\rho_2(t)$ are the first and second order terms in the expression of the statistical operator $\rho(t)$ in powers of H_I. H_0 is the full Hamiltonian for the normal metal. A standard linear response[17] calculation yields

$$\frac{d}{dt}\langle E(t)\rangle = 2\omega A \mid F \mid^{2} \chi''(q,\omega), \tag{9}$$

where $\chi''(q,\omega)$ is the imaginary part of the pair-field susceptibility

$$\chi''(q,\omega) =$$

$$\int d^2r_1 dt_1 e^{-i[q\cdot(r_1-r_2)-\omega(t_1-t_2)]}\langle[\psi^+_{op}(r_1t_1),\ \psi_{op}(r_2t_2]\rangle \tag{10}$$

Using a form of the fluctuation-dissipation[17] theorem, $\chi''(q,\omega)$ may be related to the fourier transform of the order-parameter-order parameter correlation function $G(q,\omega)$:

$$\chi''(q,\omega) = \frac{1-\exp(-\hbar\omega/k_BT)}{2\hbar} G(q,\omega) \tag{11}$$

where

$$G(q,\omega) =$$

$$\int d^2r_1 dt_1 e^{-iq\cdot(r_1-r_2) + i\omega(t_1-t_2)}\langle\psi_{op}(r_1t_1)\psi^+_{op}(r_2t_2)\rangle \tag{12}$$

For the experimentally interesting range of voltages $h\omega \ll k_BT$ and $\chi''(q,\omega)$ becomes

$$\chi''(q,\omega) = \frac{\omega}{2k_BT} G(q,\omega) \tag{13}$$

In evaluating (12) it is convenient to introduce the Fourier transform

$$\psi_{op}(\vec{r},t) = \sum_k \psi_{op}(k,t)e^{i\vec{k}\cdot\vec{r}} . \qquad (14)$$

The susceptibility becomes

$$\chi''(q,\omega) =$$

$$\sum_{kk} \frac{\omega}{2k_BT} \int d^2rdte^{-i[\vec{q}\cdot(\vec{r}_1-\vec{r}_2)-\omega(t_1-t_2)-\vec{k}\cdot\vec{r}_1+\vec{k}'\cdot r_2]} \langle\psi_{op}(k,t_1)\ \psi^+_{op}(k',t_2)\rangle \qquad (15)$$

In order to allow for order parameter fluctuations a statistical operator for an unrestricted ensemble must be used to evaluate the thermal average in (15). The standard result[16] is of the form:

$$\langle\psi_{op}(kt_1)\psi^+_{op}(k't_2)\rangle =$$

$$\frac{\prod_{k_i} \int d\{\psi(k_i)\} \exp [-\frac{1}{k_BT} F(\{\psi(k_i)\})]\psi(k,t_1)\psi^*(k',t_2)}{\prod_{k_i} \int d\{\psi(k_i)\} \exp [-\frac{1}{k_BT} F(\{\psi(k_i)\})]} \qquad (16)$$

In the above expression the integral is taken over the values of the complete set of fourier components in the expansion of the order parameter. $F(\{\psi(k_i)\})$ is the free energy functional in zero magnetic field which in linearized form[16] for a two dimensional system of area A and thickness d is

$$F = \frac{\hbar^2}{2m} Ad \sum \{\frac{1}{\xi^2} + k^2\} \mid \psi(k) \mid ^2 \qquad (17)$$

Strictly considered, the free energy given by (17) should contain terms in H_I, however the pair potential decays rapidly in the insulating barrier of the junction and the magnitude of H_I compared to other terms in the free energy is generally small.

The time dependence of $\psi(k,t)$ is obtained from the Ginzburg-Landau equation for the relaxation of the order parameter in zero field;[16]

$$[\frac{\partial}{\partial t} + \frac{\Gamma_0}{\alpha}(\alpha + \frac{1}{2m}(-i\hbar\nabla)^2 + \beta\,|\,\psi\,|^2)]\psi(r,t) = 0. \tag{18}$$

After linearizing and Fourier transforming (18) it becomes

$$[\frac{\partial}{\partial t} + \Gamma_k]\,\psi(k,t) = 0 \tag{19}$$

where

$$\Gamma_k = \Gamma_0(1 + k^2\,\xi^2(T)), \tag{20}$$

and the quantity Γ_0 is the pair relaxation frequency given by:

$$\Gamma_0 = \frac{8}{\pi}\frac{k_B T_c}{\hbar}\,[\frac{T - T_c}{T_c}], \tag{21}$$

The solution to (19) for $\psi(k,t)$ is

$$\psi(k,t) = \psi(k)\,\exp(-\Gamma_k\,|\,t\,|\,). \tag{22}$$

Inserting (22) into (16) and carrying out the integration over $\{\psi(k_i)\}$ yields:

$$<\psi_{op}(kt_1)\psi^+_{op}(k't_2)> =$$

$$\delta_{kk'} \frac{k_B T_c}{Ad} \frac{2m}{\hbar^2} [\frac{1}{\xi^2} + k^2]^{-1} \exp [-\Gamma_k \mid t_1 - t_2 \mid] \qquad (23)$$

Substituting (23) into (15) and evaluating the space and time integrals, the imaginary part of the susceptibility is:

$$\chi''(q,\omega) = \frac{2m}{\hbar^2 d} \frac{2\Gamma_q}{\omega^2 + \Gamma_q^2} [\frac{1}{\xi^2} + q^2]^{-1}. \qquad (24)$$

Combining (24) with (9) and setting $\frac{d}{dt} \langle E \rangle$ equal to $I_1 V$ the following expression for the pair current is obtained after some rearrangement:

$$I_1 = \frac{4e \mid \bar{C} \mid^2 A}{h \; dN(0)} \frac{1}{\varepsilon} \frac{\omega/\Gamma_0}{(1 + q^2\xi^2)^2 + (\omega/\Gamma_0)^2} \qquad (25)$$

Equation (25) is identical to Scalapino's[8] equation for the pair current. In (25), the parameter ω is related to the junction voltage through the Josephson relation and q is found from the value of the magnetic field applied parallel to the plane of the junction as in equation (2).

It must be emphasized that the above treatment is strictly correct only in the limit of a weak magnetic field as an applied field is assumed to determine only the phase of the interaction Hamiltonian. A more rigorous theory must include the magnetic field in the free energy functional used to calculate the various thermal averages. The Fourier decomposition of the order parameter must be made using eigenfunctions appropriate to the problem of a finite thickness superconducting film in a uniform parallel magnetic field. A calculation including these features has been carried out by Tan[12] and is in substantially better agreement with experiment[9] than the quasi-classical model which leads to (25). The magnetic field problem, as was indicated earlier, will be the subject of a future paper.

A second aspect of the calculation of the pair current, omitted in the derivation of (25) is the role of the thermal noise associated with the quasiparticle tunneling channel. In the evaluation of the order-parameter-order parameter correlation function the real and imaginary parts of the order parameter were treated as independent stochastic variables whose thermal properties could be computed using the Ginzburg-Landau free energy functional. To include voltage fluctuations due to quasiparticle noise in the calculation, it is necessary to add an additional independently fluctuating component to the overall phase of the order parameter in the normal metal. This phase fluctuation is implied by the quasiparticle voltage fluctuations through the Gor'kov-Josephson relation $\frac{d\phi}{dt} = \frac{2e\tilde{V}}{\hbar}$. The quantity $\tilde{V}$ is determined from the power spectrum of the quasiparticle noise which in the limit $V \ll \Delta$ may be taken to be that of Johnson noise.[18,19] There are several complications that must be considered in order to carry out a realistic calculation. It is first necessary to take into account the nonlinearity of the dc I-V characteristic and the possibility that the dc dynamic resistance might be negative. It is certain that this is the case for junctions of high quasiparticle resistance close to T_c. Secondly, a realistic calculation must take into account the displacement currents associated with the junction capacitance. In effect, what is required is the extension of the Ambegaokar-Halperin, Ivanchenko-Zil'berman[19] calculation of the effect of thermal noise on the dc Josephson current which applied to a superconductor-superconductor junction, to the present high temperature geometry consisting of a normal metal and superconductor.

Nevertheless, as we will show in section IV comparison of experiment with equation (25) appears to succeed over a substantial range of voltages and temperatures suggesting that the noise corrections are usually small.

III. Experimental

The basic data for this experiment are current-voltage characteristics of superconductor-insulator-normal metal tunneling junctions biased from a constant current source. As experimentally interesting effects occur at voltages the order of 1μV, phase sensitive detection techniques were used to plot I-V characteristics. To determine the pair tunneling current, it was necessary to determine the quasiparticle current and subtract it from the total tunneling current. As the quasiparticle current is usually a linear function of voltage in a normal metal-superconductor junction in the vicinity of the origin of the I-V characteristic it could be determined by extrapolation from voltages at which the pair current was insignificant. The required subtraction could then be performed electronically using operational amplifier circuits.

Each junction was prepared by vacuum evaporation onto glazed alumina substrates of two crossed metal films separated by an insulating layer thermally grown in an intermediate step. All evaporation and masking steps were done without breaking the vacuum. To eliminate the effects of edges, the edges of the lower film were covered with a layer of bismuth oxide, a minimum of 1000 Å thick, so that only a small area in the middle of the crossed-film structure was available for tunneling currents.

Temperatures were determined by measuring the resistance of a germaniun thermometer, the calibration of which was based on the NBS He^4 vapor pressure scale. The absolute accuracy of the temperatures determined from this thermometer was within $\pm$ 2 mK with a resolution of 10^{-5} K easily possible with the resistance bridge used in the measurements. A servo-feedback loop was used to stabilize sample temperatures. Short-term stability of better than 0.1 mK could easily be attained.

An additional experimental problem was that of shielding junctions from electromagnetic interference. The complete experimental apparatus was housed in a 10' x 10' x 8' shielded room with a shielding from 10 KHz to 10 GHz better than 120 db. Electromagnetically noisy instruments such as oscilloscopes and digital voltmeters were not used in the room while data was being taken. Magnetic shielding was provided by a mumetal enclosure which reduced the field at the site of the sample to less than 3×10^{-3} G.

On cooling to less than T_c, several qualitative tests were performed on the junctions to determine their quality. The magnetic field dependence was examined for the quasiperiodic Fraunhofer pattern. If the junction coupling were nonuniform, secondary maxima would be reduced in magnitude. Another test was that the temperature dependence of the critical current be correct. Of primary concern was the requirement that junctions be free of filamentary shorts exhibiting supercurrents above T_c which might appear to be the sought after pair tunneling current. Strong shorts are easily detected as they are insensitive to weak magnetic fields and their presence makes it impossible to see the superconductor-insulator-superconductor single particle tunneling characteristic. A test for weak shorts is the presence of zero-field steps in the I-V characteristic which Matisoo[20] has shown can result from metallic whiskers. In unshorted junctions, steps appear in homogeneous junctions only in the presence of a field.

A possible complication is an effect due to the resistance of the normal metal film. If its value is the order of the junction resistance, then the I-V characteristic will be distorted by the nonuniform current flow.[21] This problem does not exist if the junction resistance is large compared to the resistance of the films adjacent to the junction. The ratio of junction resistance to film resistance was greater than 20 for all junctions used in these experiments.

IV. Data and Analysis

Extensive measurements of the properties of tin-lead and aluminum-lead junctions used in the analysis are summarized in Table I. From the Fiske step spacing[22] given in Table I, one can find $\bar{c}$,[23] the wave velocity in the junction, and in turn, calculate from $\bar{c}$ the junction capacitance C. Although C plays no role in the present analysis, it would be important in a future comparison with a theory which included the effects of the ac junction impedance and quasiparticle noise.

In Figure 1 are plotted excess current voltage characteristics obtained at several temperatures for sample Al-6. The various curves are in excellent agreement with equation (25) which predicts the characteristic to have a quasi-Lorentzian shape. The quasi-Lorentzian dependence of the excess current on voltage could be observed qualitatively but not quantitatively at temperatures as high as 1 K above T_c in some samples. This suggests that improvements in signal processing techniques might permit the study of the regime in which nonlinear effects are significant.[6]

In certain junctions with high quasiparticle resistances, the existence of a negative resistance region observable over a relatively large temperature range was suggested by the shape of the I-V characteristic at voltages greater than the voltage corresponding to the peak of the excess current. In the immediate vicinity of T_c, all junctions showed evidence of a negative resistance region. The I-V characteristics in the negative resistance region were not studied because the signal processing technique used required that the junctions be current biased, whereas to avoid switching and oscillation in the negative resistance region voltage biasing is necessary.

We have restricted detailed analyses of data to cases in which the I-V characteristic was truly quasi-Lorentzian. In some junctions a nonlinear quasiparticle current-voltage characteristic was observed. In these cases it was impossible to make a unique subtraction which would lend to a quasi-Lorentzian excess current-voltage characteristic so that detailed analyses were not attempted.

Data analysis is based on comparing various features of the excess current-voltage characteristic with the predictions of equation (25). This is reasonable only when the possible complications introduced by a negative dynamical impedance are not present. We have examined the temperature dpendences of the peak current, peak voltage and conductivity in the limit of zero voltage. The peak current and peak voltage alone are sufficient to scale the quasi-Lorentzian dependence of the excess current on voltage.

The peak voltage V_p is directly related to the pair relaxation frequency by the relation

$$V_p = \frac{\hbar\Gamma_0}{2e} = \frac{4k_BT_c}{\pi e}\left(\frac{T-T_c}{T_c}\right) \qquad (26)$$

Data for samples Al-6 and Al-23 are shown in Figures 2a and 2b. The experimental voltages are in quantitative agreement with theory outside of the immediate vicinity of T_c. Near T_c, the peak voltage apparently drops well below the theoretical curve.

In Figures 3a and 3b the values of I_p^{-1}, where I_p is the current at the peak of the excess current-voltage characteristic, are plotted as a function of temperature for samples Al-6 and Al-23. The solid lines computed from equation (25),are given by

$$I_p = \frac{2e}{\hbar} \frac{1}{H_c^2/4\pi} \frac{E_1^2}{Ad} \left(\frac{T - T_c}{T_c} \right)^{-1} \tag{27}$$

where we have used the relations $1/2\ N(0)\Delta^2 = H_c^2/8\pi$, and $|\bar{C}|^2\Delta^2 = E_1^2/A^2$. E_1 is the zero-temperature Josephson coupling energy and H_c is the zero-temperature critical field. E_1 was calculated from

$$E_1(0) = \frac{\hbar}{2e} \frac{\pi}{R_N} \frac{\Delta(0)\ \Delta'(0)}{\Delta(0) + \Delta'(0)} \tag{28}$$

using measured values of the energy gaps of lead and aluminum and the normal state tunneling resistance. The critical field was obtained by scaling the standard value of the critical field for aluminum, 99 gauss, by the shift in the transition temperature, assuming the film to be a BCS superconductor,[24] and taking the bulk transition temperature to be 1.2 K. Examination of (24) shows that the temperature dependence of the peak in the excess current is determined by the temperature dependence of the coherence length $\xi(T)$. Agreement of experiment and theory is seen to be quantitative and can be taken to be a verification of the temperature dependence of $\xi(T)$.

In Figures 4a and 4b we have plotted $\left(\frac{dI_1}{dV}\right)^{-1/2}$ vs. T for Al-6 and Al-23. The straight line is computed from (25) using the parameters of Figure (3) and the theoretical value for the relaxation frequency Γ_0 . The agreement is again seen to be quantitative.

In Figure 5 the temperature dependence of the zero-voltage current of Al-6 is plotted. The inverses of the data points are plotted as a function of temperature in Figure 6. The triangular points in Figure (5) lie on the straight line segment in Figure (6). The triangular points which are plotted as zero voltage currents were obtained at temperatures above the temperature to which the straight line segments in Figures 2a, 3a, 4a extrapolated to zero. The fact that they lie on the straight line segment in Figure 6 is consistent with their being peak currents in a pair tunneling characteristic above T_c for which the peak voltage is in the noise of the electronics (V_p<100nV). The sharp departure from linearity at the low temperature end of Figure 6 would appear to determine the beginning of the transition from the pair fluctuation regime to the regime characterized by ordinary Josephson tunneling. In our discussion T_c is determined by extrapolation of data taken in regimes where the theory appears to be successful. Determination of the "real" transition temperature and the extent of its rounding will require more detailed experiments.

V. Discussion

A major discrepancy between experiment and theory does exist for the plot of the temperature dependence of the relaxation frequency in the vicinity of the superconducting transition. There the curve of the peak voltage V_p vs. T drops below the line which is an extrapolation from the high temperature data. The peak current over the same range of temperature does not appear to depart in as drastic a manner from the theory. The width of the region over which the peak voltage drops below the theoretical value is quite substantial in the aluminum-lead junctions. Comparison of data reported here with data reported on tin-lead junctions[9] suggests that the measurements on tin-lead junctions do not cover a sufficiently wide temperature range to permit quantitative conclusions relating to the relaxation frequency.

It is not clear at the present time as to whether the low peak voltages are related to "critical behavior", are some manifestation of the proximity effect or are simply a consequence of negative dynamical resistance near T_c, not taken into account in our analysis. One can probably rule out the proximity effect as there is no correlation between the width of the temperature range over which V_p is less than the theoretical value and the magnitude of the low temperature Josephson current. If the proximity effect were responsible for the observed phenomenon,one would expect its magnitude to depend on the strength of the tunneling interaction which would be proportional to the magnitude of the low temperature Josephson current. It is possible that the observed effects are a manifestation of critical behavior. A reduced value of the relaxation frequency would be observed in a region in temperature over which there was a "critical slowing down" of the fluctuations. The difficulty with this interpretation is that if the Ginzburg criterion[25] is used to estimate the width of the critical region for these films the results are about a factor of ten smaller than the width of the region over which V_p is anomalously low. Nevertheless it is not inconceivable that an observable effect of critical fluctuations such as a "slowing down" might be seen over a range of temperatures several times the width of the critical region as determined by the Ginzburg criterion. Further quantitative experiments and more detailed theoretical investigation are clearly needed before firm conclusions can be made.

We have left discussion of the magnetic field dependence of the fluctuating pair current to a later paper. The results of our early work on tin can be explained qualitatively by the calculations of Tan[12] in which the susceptibility is computed from a free energy functional in which the parallel magnetic field is included explicitly rather than treated quasi-classically. Tan's calculation also incorporates the boundary condition at the oxide-normal metal interface in an explicit manner. The magnetic field data on the aluminum-lead junctions not shown here is also qualitatively consistent with the theory but does not constitute a definitive test as all the aluminum films are in the extreme dirty limit where the magnetic field dependence of the pattern is weak. In our later paper we will discuss the impurity dependence of the magnetic field effects.

In conclusion, the zero-field data appears to be in excellent agreement with the predictions of the Ginzburg-Landau theory over a substantial temperature range. The observation of the quasi-Lorentzian pair current-voltage characteristic in zero magnetic field constitutes a determination of $\chi(0,\omega)$ for a superconductor. In the plot of the temperature dependence of the peak voltage, the numerical value of the slope constitutes a quantitative determination of the $q = 0$ relaxation frequency for order-parameter fluctuations.

In the plot of the temperature dependence of the reciprocal of the peak current, the numerical value of the slope constitutes a quantitative determination of the coherence length above the transition temperature.

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Table I [a]

Junction Properties

Junction: Property	Sn-1 [b]	Sn-2 [b]	Al-5 [c]	Al-6	Al-23
A $(cm)^2$	3.1×10^{-4}	1.04×10^{-4}	7.34×10^{-4}	7.34×10^{-4}	7.34×10^{-4}
R_N (ohms)	5	2.1	0.069	0.10	0.451
R_D (ohms)	70.5	11.4	3.75	8.64	280
$I_1(0)$ (amps)	1.7×10^{-4}	3.4×10^{-4}	------	------	------
$I_1(1.269K)$ (amps)	-------	-------	2.4×10^{-3}	1.7×10^{-3}	1.2×10^{-3}
T_c (Kelvin)	3.712	3.886	1.751	1.765	1.860
d (cm)	1.5×10^{-5}	1.5×10^{-5}	1.28×10^{-5}	1.30×10^{-5}	1.6×10^{-5}
$\rho(N)$ (Ohm-cm)	8×10^{-7}	8×10^{-7}	5.8×10^{-6}	7.5×10^{-6}	3.81×10^{-6}
Fiske Step Spacing (μV)	-------	-------	22.5	15.8	11.4
C(F)	-------	-------	15.6×10^{-9}	31.9×10^{-9}	61.0×10^{-9}

a Notation: A is the area of the plane of the junction, R_N is the normal tunneling resistance, R_D is the quasiparticle resistance, $I_1(0)$ is the zero temperature dc Josephson current, $I_1(1.269K)$ is the measured operation current at 1.269K, T_c is the approximate transition temperature of the normal side of the junction, d is the thickness of the normal side , $\rho(N)$ is the low temperature normal resistivity of the normal side, The Fiske Step Spacing is used to compute the capacitance, with the penetration depth in aluminum taken to be 500 A. The junctions referenced by Sn are Tin-Tin Oxide-Lead and those referenced by Al are Aluminum-Aluminum Oxide-Lead. Linear dimensions and thicknesses are $\pm$ 5%.

b Data for Sn-1 and Sn-2 were published in reference 9.

c Not shown here but almost identical with Al-6

Figure Captions

Figure 1: Excess current-voltage characteristics of junction Al-6 at several temperatures. Curves I, II, III, and IV correspond to T = 1.8420 K, 1.8087 K, 1.7992 K and 1.7809 K respectively. Quasiparticle currents in each case were electronically subtracted from the full tunneling current, assuming a fixed resistance of 8.64 ohms.

Figure 2a: Peak voltage V_p vs. T for junction Al-6. The solid line corresponds to the theoretical value of the relaxation frequency with T_c taken to be 1.762 K.

Figure 2b: Peak voltage V_p vs. T for junction Al-23. T_c is taken to be 1.860 K.

Figure 3a: $(I_p)^{-1}$ vs. T for Al-6. The solid line is computed from equation (27) using parameters in Table I.

Figure 3b: $(I_p)^{-1}$ vs. T for Al-23. The solid line is computed using relevant parameters from Table I.

Figure 4a: $(\frac{dI}{dV})^{-1/2}$ vs. T for Al-6 evaluated at zero bias. The solid curve is computed from equation (25) using the transition temperature from Table I and assuming the aluminum film to be a BCS superconductor.

Figure 4b: $(\frac{dI}{dV})^{-1/2}$ vs. T for A-23, evaluated at zero bias. Relevant parameters are taken from Table I.

Figure 5: Temperature dependence of the zero voltage Josephson current of Al-6. Triangular points may also be interpreted as peak excess currents occurring at anomalously low voltages.

Figure 6: Reciprocals of data of Fig. 5 plotted as a function of temperature. The line correspond to a slope slightly greater than the slope of the line in Figure 3a.

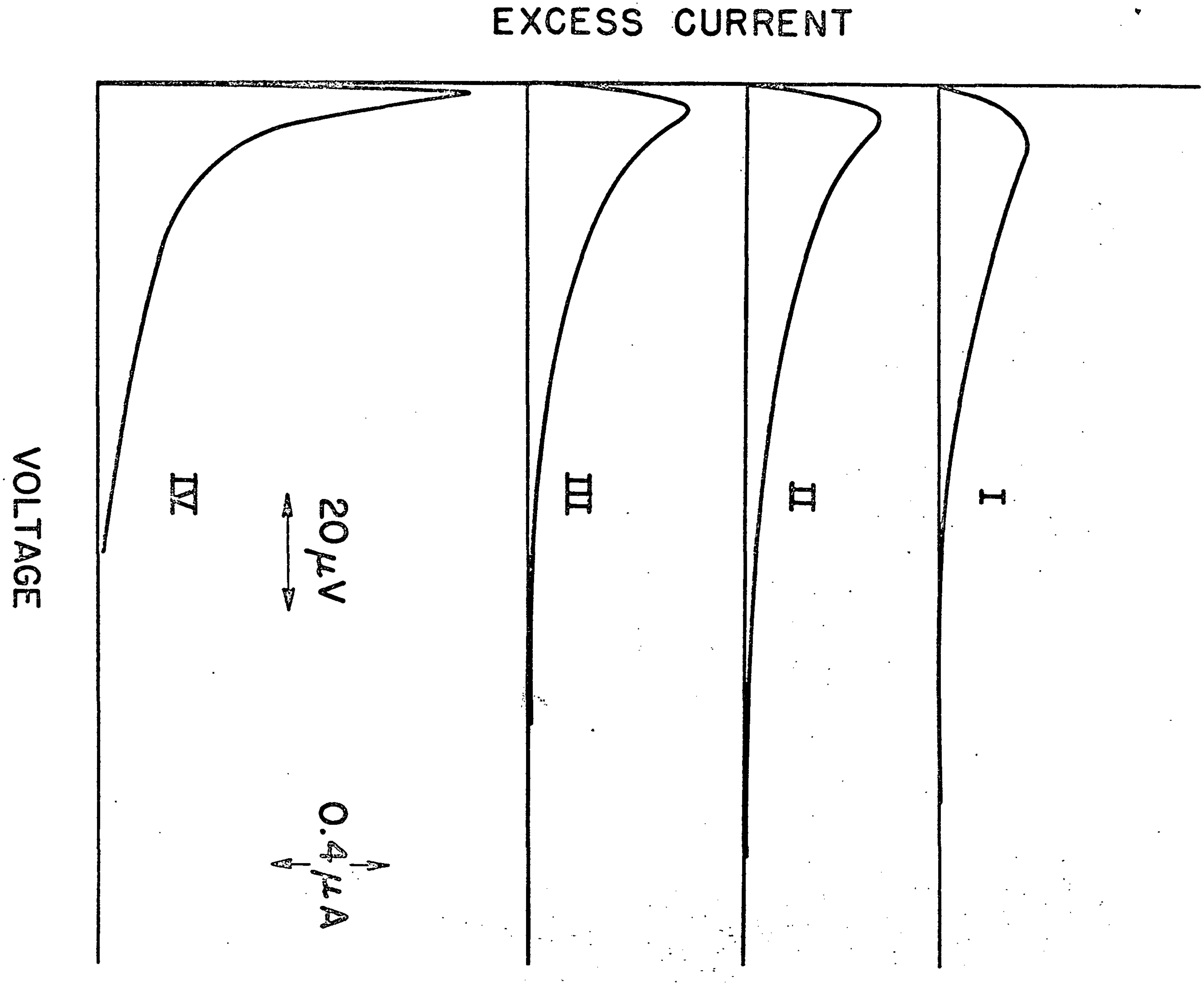


Fig. 1

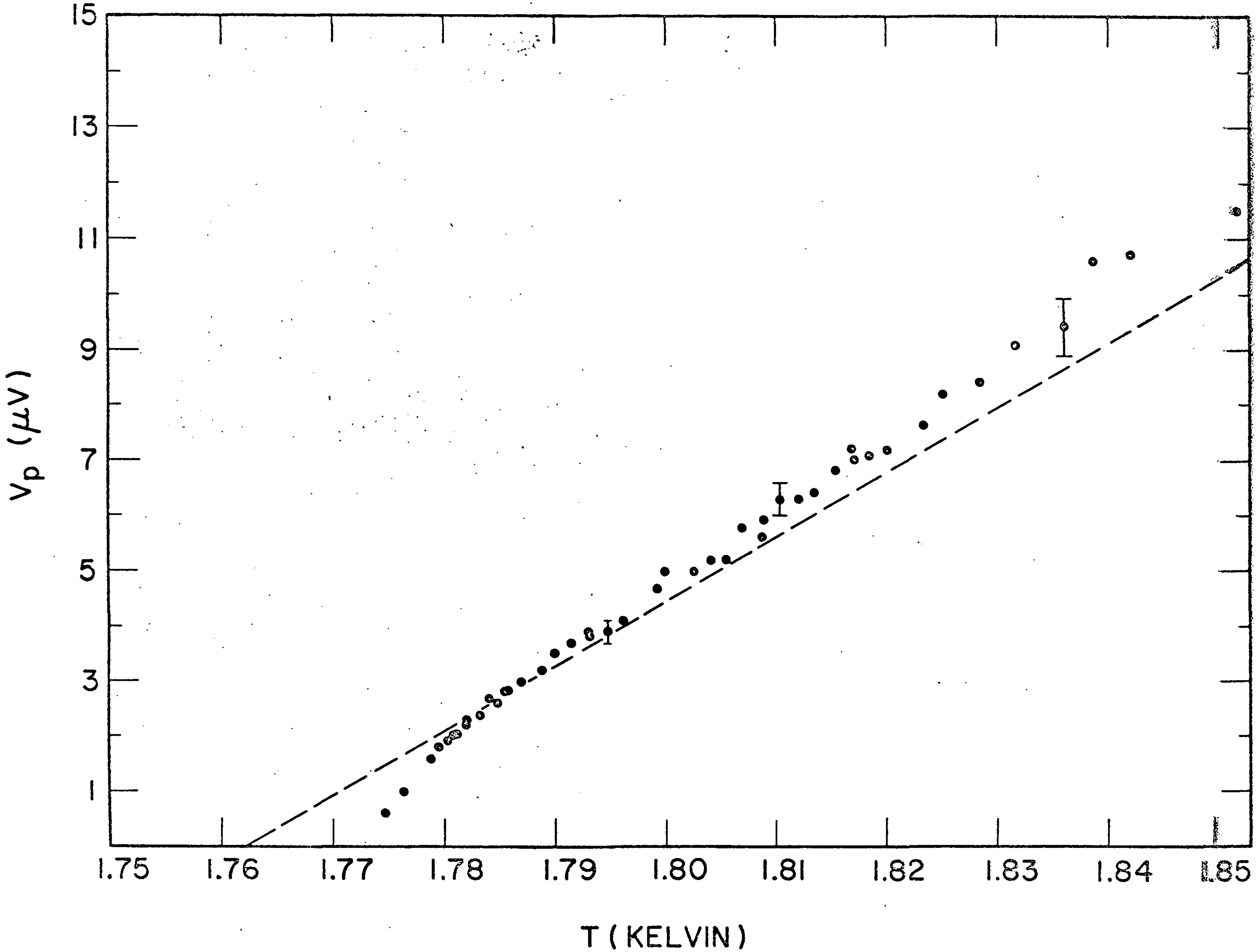

15
13
11
9
7
5
3
1
V_p (μV)
1.75
1.76
1.77
1.78
1.79
1.80
1.81
1.82
1.83
1.84
1.85
T (KELVIN)

Fig. 2a

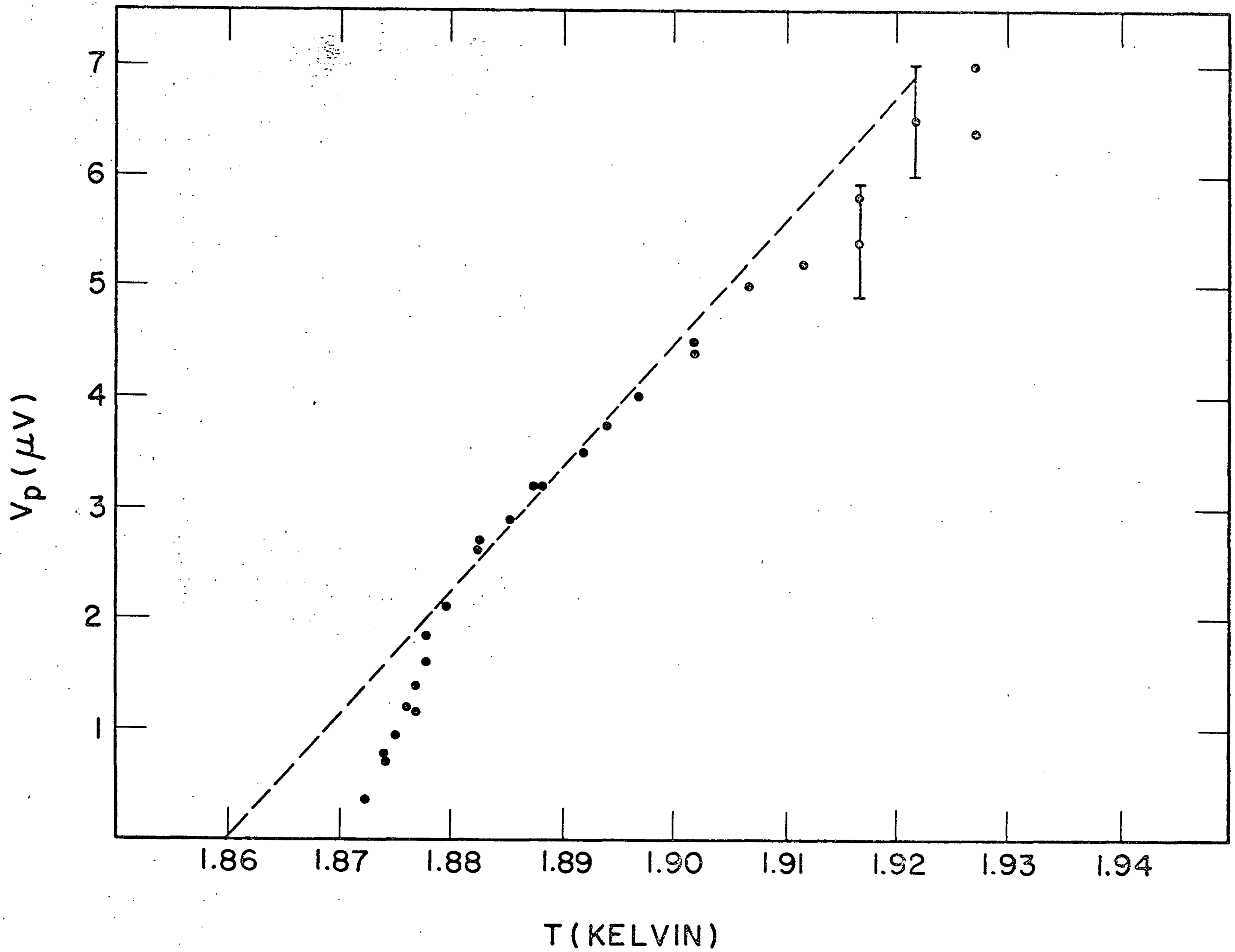
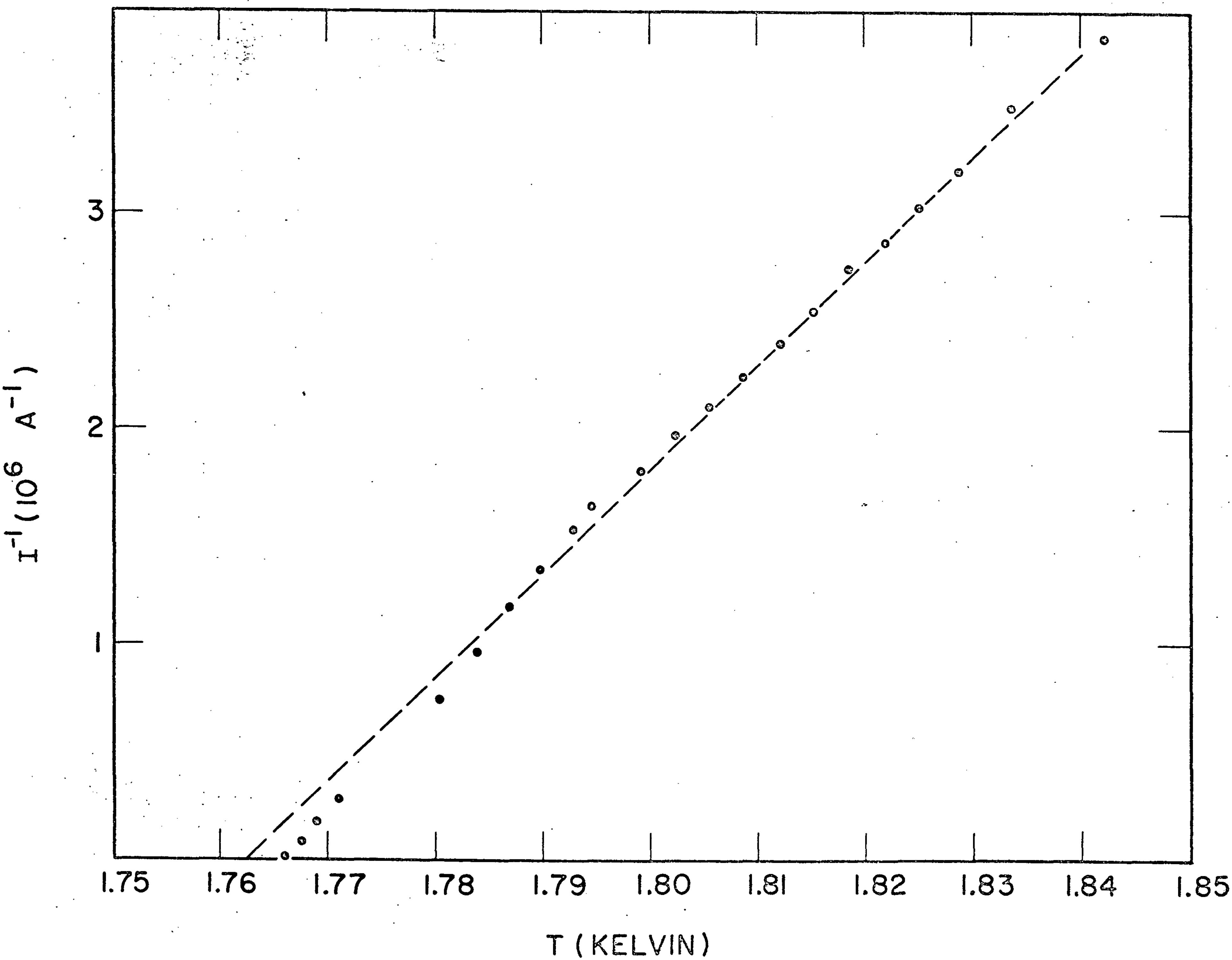


Fig. 2b



3
2
1
I⁻¹ (10⁶ A⁻¹)
1.75
1.76
1.77
1.78
1.79
1.80
1.81
1.82
1.83
1.84
1.85
T (KELVIN)

Fig. 3a

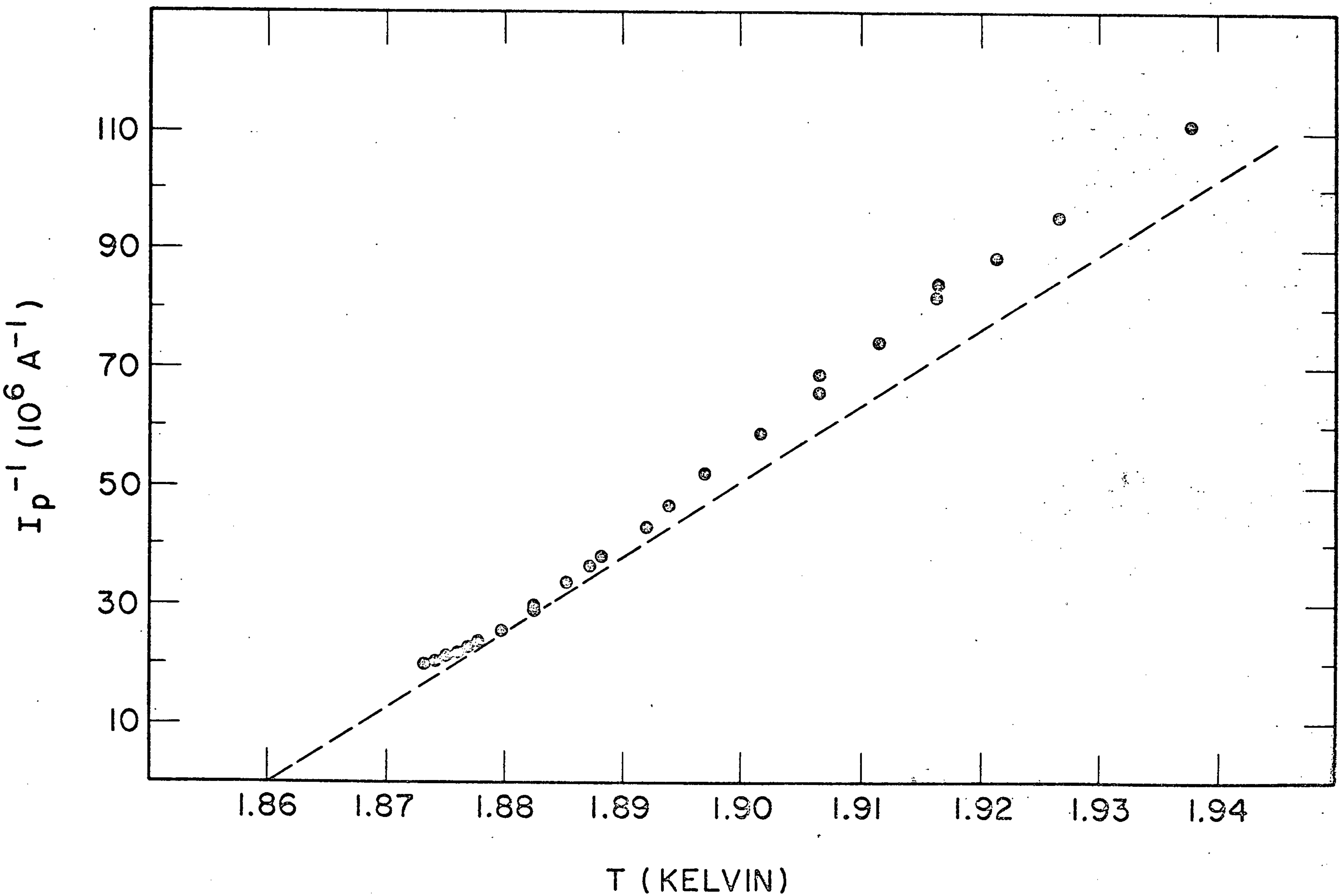
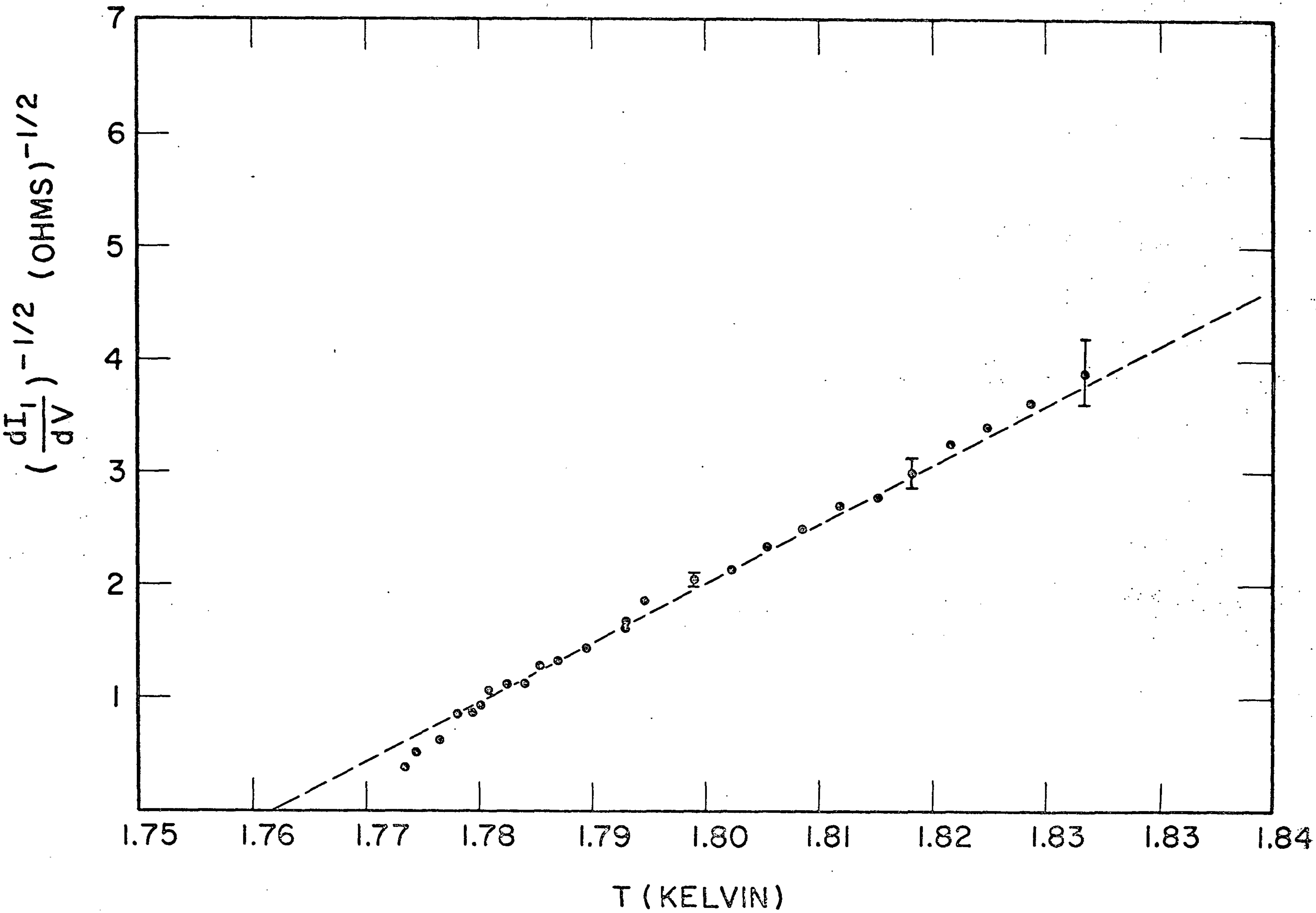
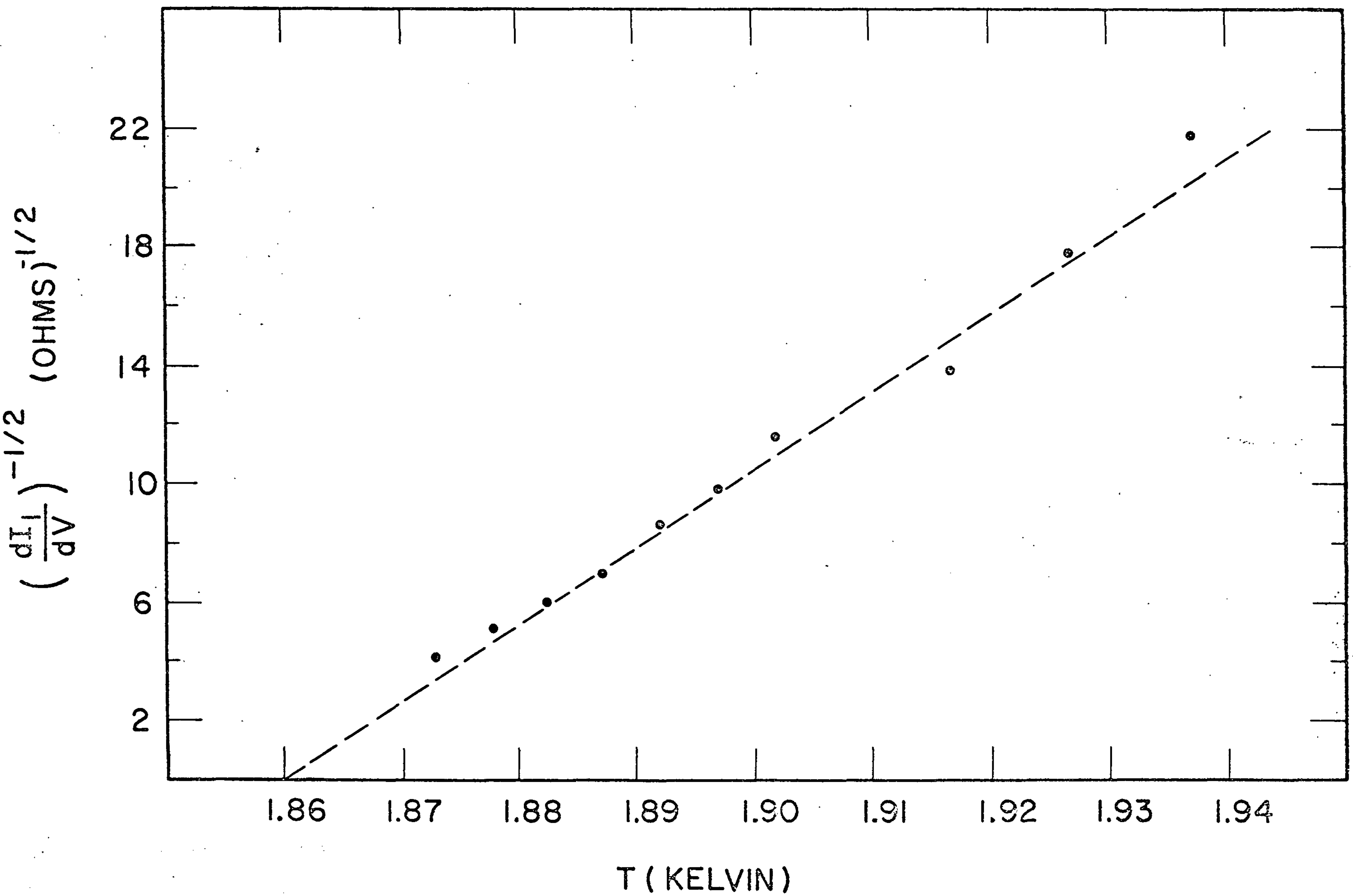

I_p^{-1} (10^6 A^{-1})
110
90
70
50
30
10
1.86
1.87
1.88
1.89
1.90
1.91
1.92
1.93
1.94
T (KELVIN)

Fig. 3b



$(\frac{dI_1}{dV})^{-1/2}$ (OHMS)$^{-1/2}$
7
6
5
4
3
2
1
1.75
1.76
1.77
1.78
1.79
1.80
1.81
1.82
1.83
1.83
1.84
T (KELVIN)

Fig. 4a



$\left(\frac{dI_1}{dV}\right)^{-1/2}$ (OHMS)$^{-1/2}$
22
18
14
10
6
2
1.86
1.87
1.88
1.89
1.90
1.91
1.92
1.93
1.94
T (KELVIN)

Fig. 4b

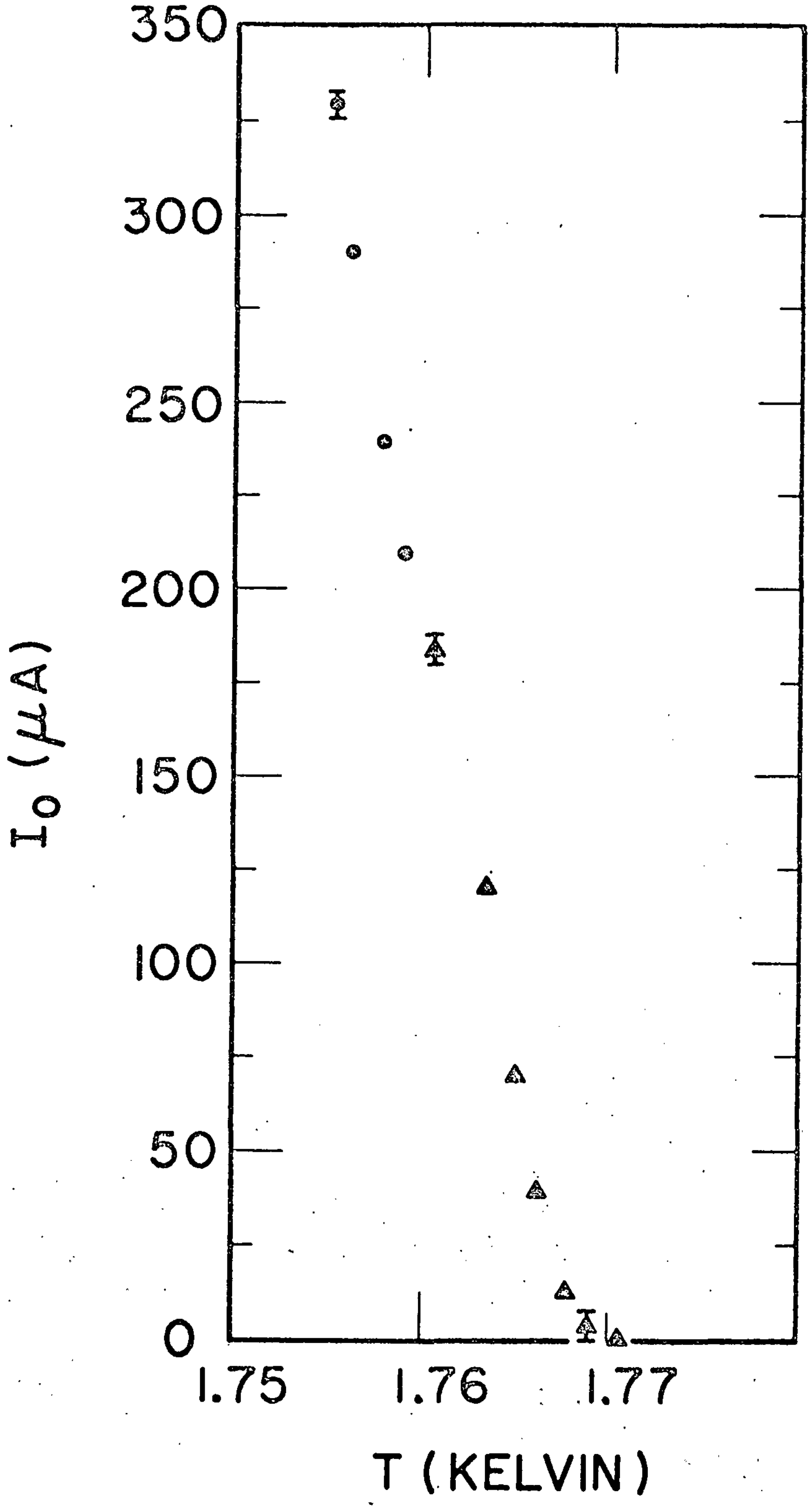
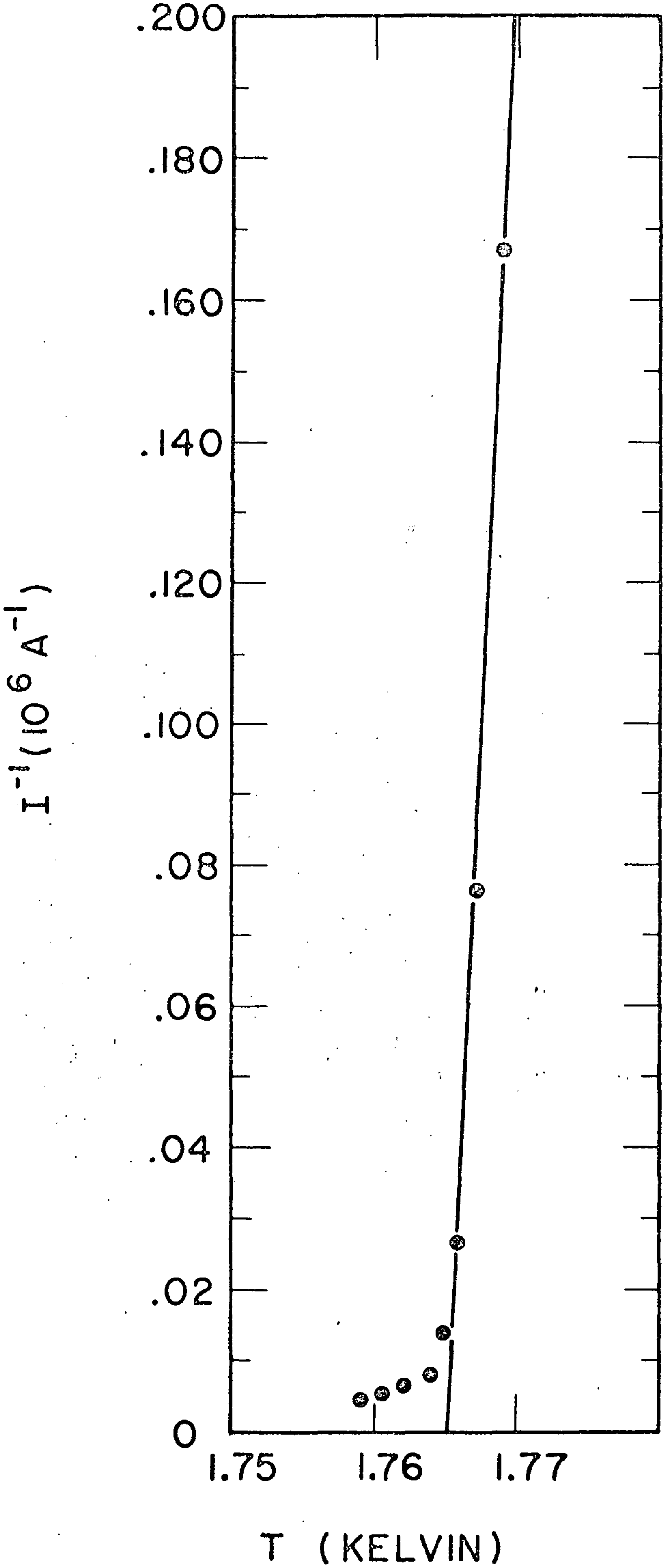


Fig. 5



.200
.180
.160
.140
.120
.100
.08
.06
.04
.02
0
$I^{-1}(10^6 A^{-1})$
1.75
1.76
1.77
T (KELVIN)

Fig. 6