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1972-03-01

Submitted to Physical Review Letters

LBL-817 c.1
Preprint

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AEC Contract No. W-7405-eng-48

DISCLAIMER

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EXPERIMENTAL OBSERVATION OF PAIR-QUASIPARTICLE POTENTIAL DIFFERENCE IN NON-EQUILIBRIUM SUPERCONDUCTORS*

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ABSTRACT

It is shown experimentally that when a quasiparticle current is converted into a pair current in a superconductor, the quasiparticle potential in the non-equilibrium region differs from the pair chemical potential.

In a recent Letter, Rieger, Scalapino, and Mercereau[1] developed a theory of non-equilibrium superconductivity. They considered a current I flowing through a superconductor S of volume Ω so that quasiparticles were injected and pairs extracted, and found that the pair and quasiparticle chemical potentials (μ_p and μ_{qp}) differed by

$$(\mu_{qp} - \mu_p)/e = I\tau_{GL}/24e^2\Omega N(0). \qquad (1)$$

In (1), τ_{GL} is the Ginzburg-Landau relaxation time, and N(0) the density of states at the Fermi level for electrons of one spin.

In this Letter, we show experimentally that the quasiparticle potential in a non-equilibrium superconductor differs from μ_p/e; however, the data do not support (1) in detail. In the following Letter[2], a new theory is presented which is in good agreement with the experimental results. In general, the quasiparticle chemical potential is not a well-defined quantity, and is replaced by a "quasiparticle potential" which arises from the imbalance of electron-like and hole-like excitations. Throughout S, μ_p is constant (as in ref. 1), and the difference between the quasiparticle potential and μ_p/e is shown to be[2]

$$V = I\tau_Q/2e^2\Omega N(0)g_{NS} . \qquad (2)$$

In (2), τ_Q is the relaxation time for the electron-like and hole-like imbalance[2], and g_{NS} is the normalised conductance[3] of an NS tunnel junction in the low-voltage limit. This result should be a good approximation over all temperatures.

To observe these non-equilibrium effects, we require a superconductor, S, of small volume into which quasiparticles are injected from a normal electrode and from which pairs are extracted into a superconducting electrode. The quasiparticle potential is measured by a normal probe which exchanges single electrons with S, while μ_p/e is measured by pair exchange with a superconducting probe. If the normal injection electrode were in good metallic contact with S, electrons incident on the interface from the normal metal with energies less than the energy gap, Δ, would be Andreev[4] reflected as holes, and pairs would be transmitted into the superconductor. Thus significant non-equilibrium effects would be observed only when $\Delta \lesssim kT$. Furthermore, good metallic contact of either normal probe or normal injection electrode with S could substantially depress the condensation amplitude in S by means of the proximity effect[5]. These difficulties may be circumvented by coupling both normal metals to S through tunnel junctions. Ideally[2], one would also like to couple the superconducting probe and electrode to S via Josephson[6] junctions, so that the non-equilibrium processes would be confined to S. In practice, this configuration would be difficult to fabricate, and in the experiment only the normal electrode and probe were coupled via junctions. However, the geometry was such that the quasiparticle-pair conversion did take place in a well-defined volume.

A strip of Al (XX') (Fig.1) of width $d \sim 3$mm and thickness ~ 1500Å, was evaporated on to a glass slide, and oxidized. Next a strip (YY') of Sn [thickness (t) 2000 to 5000 Å] was evaporated across the Al strip to form a tunnel junction. The volume of Sn overlaying the Al was the region where the quasiparticle-pair conversion occurred. The sample was removed from the evaporator, so that the Sn oxidized somewhat, and a layer of varnish applied, leaving a window about 1mm square in the middle of the junction. A diagonal strip (ZZ') of Cu was then evaporated to form the normal probe. To reduce the resistance of this probe and so make possible very low voltage measurements, a 5000 Å film of Pb was deposited over the Cu. The Cu was sufficiently thick (>1μ), and sufficiently dirty, that no Josephson tunneling[6,7] between the Sn and Pb was possible. The resistance of the Al-Sn injection junction, typically 10Ω, was much higher than the resistance of the Al strip forming the junction, so that quasiparticles were uniformly injected into the Sn. The barrier between the Sn and Cu was of much lower resistance, but was sufficiently thick to effectively quench the proximity effect; this result was verified in a separate experiment.

The voltage V between Y and Z was measured with a superconducting galvanometer[8] in series with a resistor, with a null-balancing technique, the whole circuit being immersed in liquid helium. The resistance of the Sn-Ox-Cu-Pb junction was also determined, by applying a current between Y' and Z' and measuring the voltage between Y and Z. This resistance was dominated by the oxide layer, and was typically $10^{-5}\Omega$ at the transition temperature T_c of the Sn. As the temperature was lowered, the resistance of the junction increased at approximately the rate predicted for an SN tunnel junction[3]. At the lower temperatures, the resolution of the voltmeter was seriously reduced by the high resistance thus introduced into the circuit, and relatively high injection currents (I) were required, typically 5 to 20 mA. Near T_c, the currents used were 0.1 - 1 mA. Thus at all temperatures the voltage across the Al-Sn junction was much greater than Δ, and proportional to I.

Data were rejected from samples in which the current-voltage characteristics of the Al-Sn junction indicated that metallic shorts might be present, and the quasiparticle injection therefore highly non-uniform. Average values of V from acceptable samples[9] for each thickness of Sn are shown in Fig.2. For electron injection into the Sn (Al negative relative to Sn), the Cu probe was negative with respect to μ_p. Near T_c, V reversed exactly when I was reversed, and was linear in I. Below about 0.8 T_c, V did not reverse exactly, $|V|$ being larger for electron injection than for electron extraction. This asymmetry is thought to be due to the energy-dependence of N(0) which has been neglected in both theories. The rapid rise in V as the temperature is lowered below 0.8 T_c is explained by the presence of g_{NS} in the denominator of (2). This feature is absent from (1), which is intended to be valid only near T_c, where $g_{NS} \simeq 1$.

To facilitate comparison of the two theories, it is convenient to multiply the right hand side of (1) by g_{NS}^{-1}, and to compute the quantity $\zeta = V\Omega g_{NS} I^{-1}$ from the experimental data. We then compare ζ in turn with the two expressions $\tau_{GL}/24e^2N(0)$ from (1), which varies as Δ^{-2} near T_c, and $\tau_Q/2e^2N(0)$ from (2), which varies[2] as Δ^{-1} near T_c. In Fig.3 we have plotted ζ for data taken from the three thinnest samples averaged over the asymmetry at low temperatures. V appears to be proportional to Ω^{-1}, as predicted by both theories. The temperature dependence of the data reflects the behaviour of the characteristic time. The solid curve represents Δ^{-1}, fitted to the measured T_c (3.81K), and the average value of ζ at low temperatures. The fit is surprisingly good over the whole temperature range. For comparison, the crosses (x) indicate a Δ^{-2} curve, fitted in the same way: acceptable agreement with experiment could be obtained only by choosing $T_c \simeq 4.0$K, a value much higher than that observed experimentally. It appears that a characteristic time proportional to Δ^{-1}, rather than Δ^{-2}, fits the data more adequately, a conclusion that supports the validity of (2) over that of (1).

From (1) and the low-temperature data of Fig. 3, we find that the Ginzburg-Landau time required to fit the data would be approximately 5×10^{-9} sec, about two orders of magnitude higher than any acceptable value. From (2) and the data of Fig. 3, we find that in dirty Sn, $\tau_Q = 4 \times 10^{-10}\ \Delta(0)/\Delta(T)$ sec, where $\Delta(T)/\Delta(0)$ is the normalised gap. This result is in satisfactory agreement with the theoretical estimate[2]. The characteristic length[10] over which quasiparticle-pair conversion occurs, $\lambda = (\ell_o v_F \tau_Q)^{\frac{1}{2}}$, is roughly 5$\mu$ at low temperatures, where we have taken the mean free path, ℓ_o, as 1000 Å.

It might be remarked that an experiment performed by Ginsberg[11] in an attempt to measure the <u>recombination</u> time of injected quasiparticles, in fact would have demonstrated the effects described here if the voltage resolution had been high enough.

Notice also that the configuration of Fig. 1 represents a "superconducting transistor": a current between X' and Y' develops a voltage across Y (or Y') and Z. The device is of course passive, and achieves no power gain, but could possibly be used as an impedance transformer.

Finally, we consider the implications of these experiments for the determination of e/h using the Josephson effect[12]. Electromagnetic radiation of frequency ω induces constant-voltage current steps on the characteristic of a Josephson junction whenever $n\hbar\omega = 2\Delta\mu_p$, where $\Delta\mu_p$ is the difference in <u>pair</u> chemical potential across the junction, and n is an integer. If the current and voltage leads, which are of course normal,[13] on one side of the junction (or on both sides) are within a distance λ (say), the quasiparticle potential difference measured by the voltage leads will differ from $2\Delta\mu_p/e$, and an error in e/h will result. However, in all published determinations of e/h, the current and voltage leads were well separated, and the errors due to non-equilibrium effects utterly negligible.

I should like to thank Professor D. J. Scalapino for helpful discussions during the earlier stages of the experiments, and the Cavendish Laboratory for its hospitality during the writing of this paper. I am grateful to Professor M. Tinkham for numerous helpful comments, and for a critical reading of the manuscript.

References and Footnotes

* Work supported by the U.S. Atomic Energy Commission

† Alfred P. Sloan Foundation Fellow. Presently on leave at Royal Society Mond Laboratory, Cambridge, England.

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7) J. Clarke, Proc. Roy. Soc. A308, 447 (1969).

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9) Usually, 2 out of 4 samples of a given thickness of Sn were acceptable, and were reproducible to perhaps 20%. Only one 5260 Å sample exhibited a good Al-Sn junction characteristic, which was of such low resistance that the injection was undoubtedly very non-uniform; these data were not used in Fig.3.

10) Since $t \ll \lambda \ll d$, the quasiparticle-pair conversion was uniform throughout the region where the Sn overlapped the Al.

11) D. M. Ginsberg, Phys. Rev. Letters 8, 204 (1962).

12) W. H. Parker, D. N. Langenberg, A. Denenstein, and B. N. Taylor, Phys. Rev. 177, 639 (1969).

13) Depending on the nature of the junction, the leads may form tunnel junctions or metallic contacts with the superconductors. In the latter case, the non-equilibrium effects will be greatly reduced, except near T_c, as pointed out in the text.

Figure Captions

Fig. 1 Sample configuration. In order of deposition, the films are: Al(XX'), Sn(YY'), varnish, Cu(ZZ'), and Pb(ZZ'). Galvanometer G and resistor R measure the potential difference V between Y and Z.

Fig. 2 Potential difference V between Y and Z for 4 thicknesses of Sn, against temperature. V normalised to injection current (I) of 1mA. Sample area 0.1 cm^2.

Fig. 3 Plot of $\zeta = V\Omega g_{NS} I^{-1}$ against temperature for three thinnest samples.

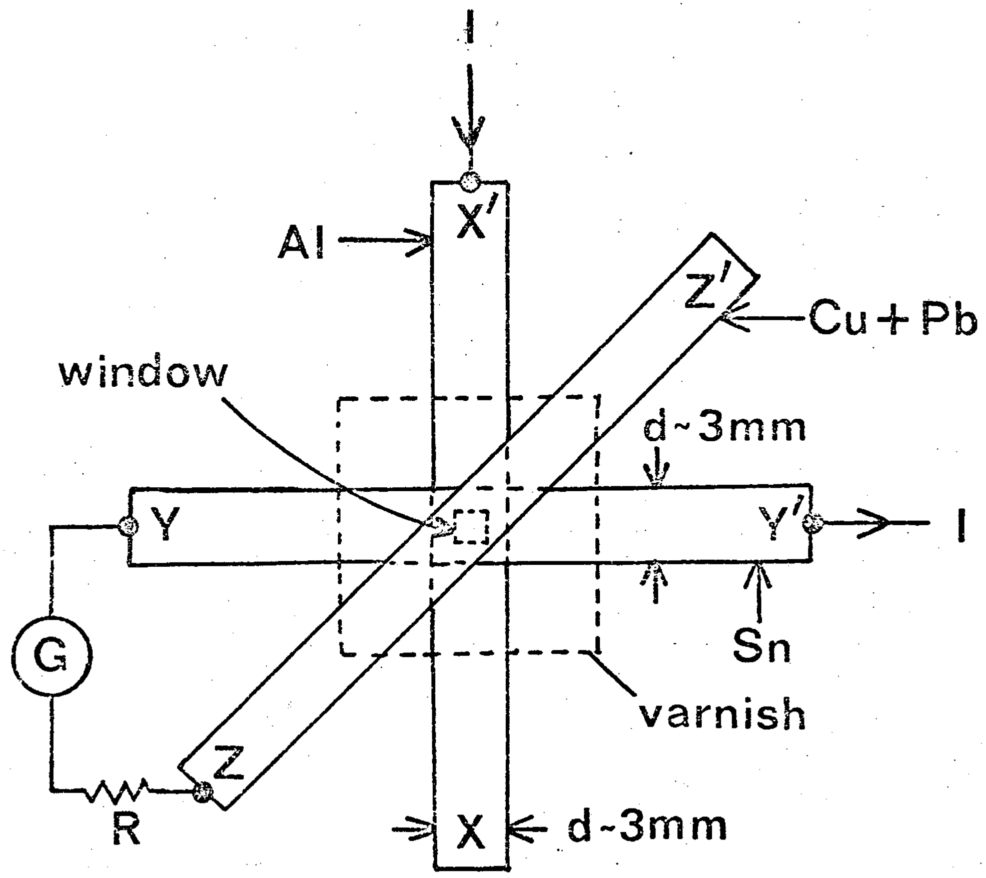


FIG 1

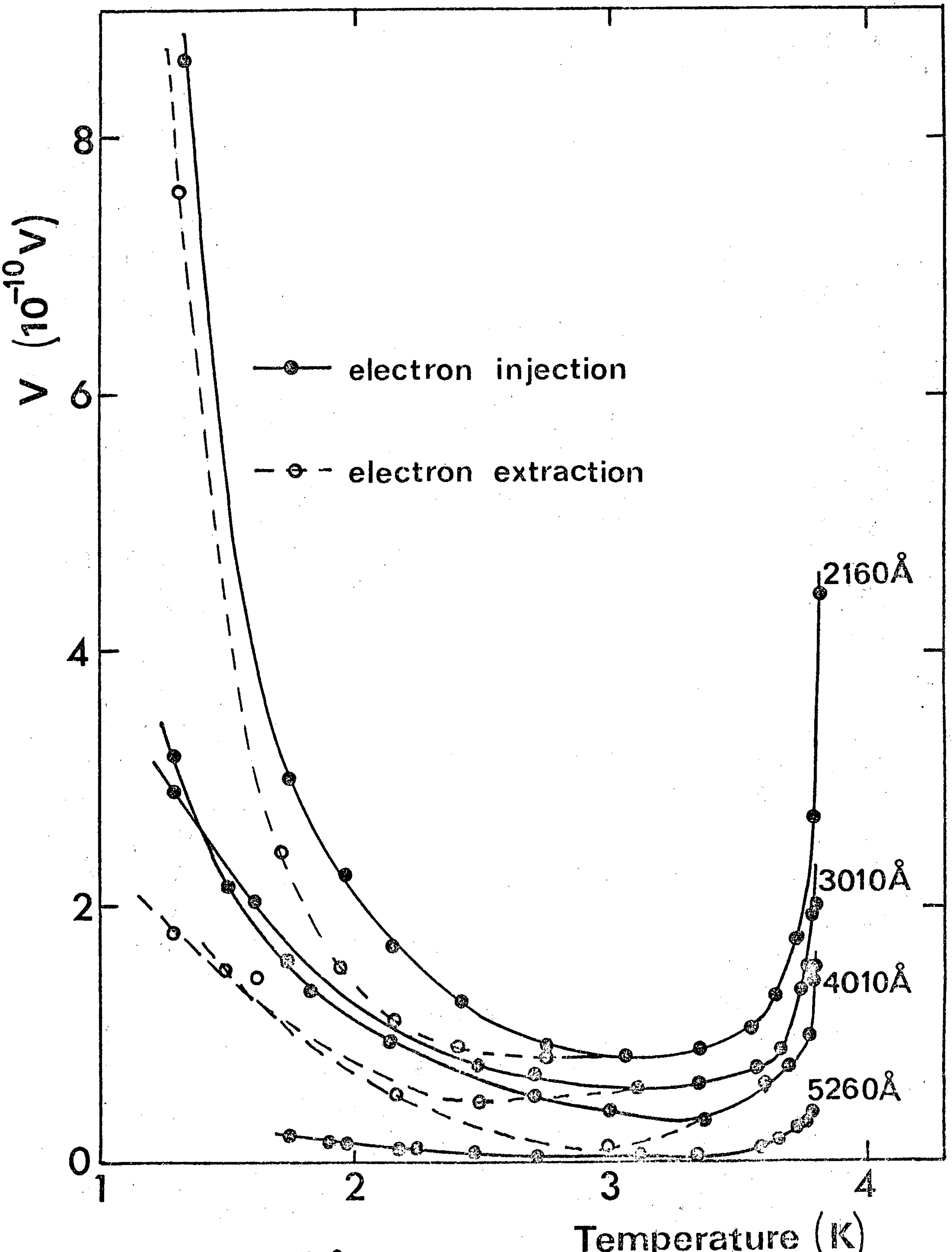
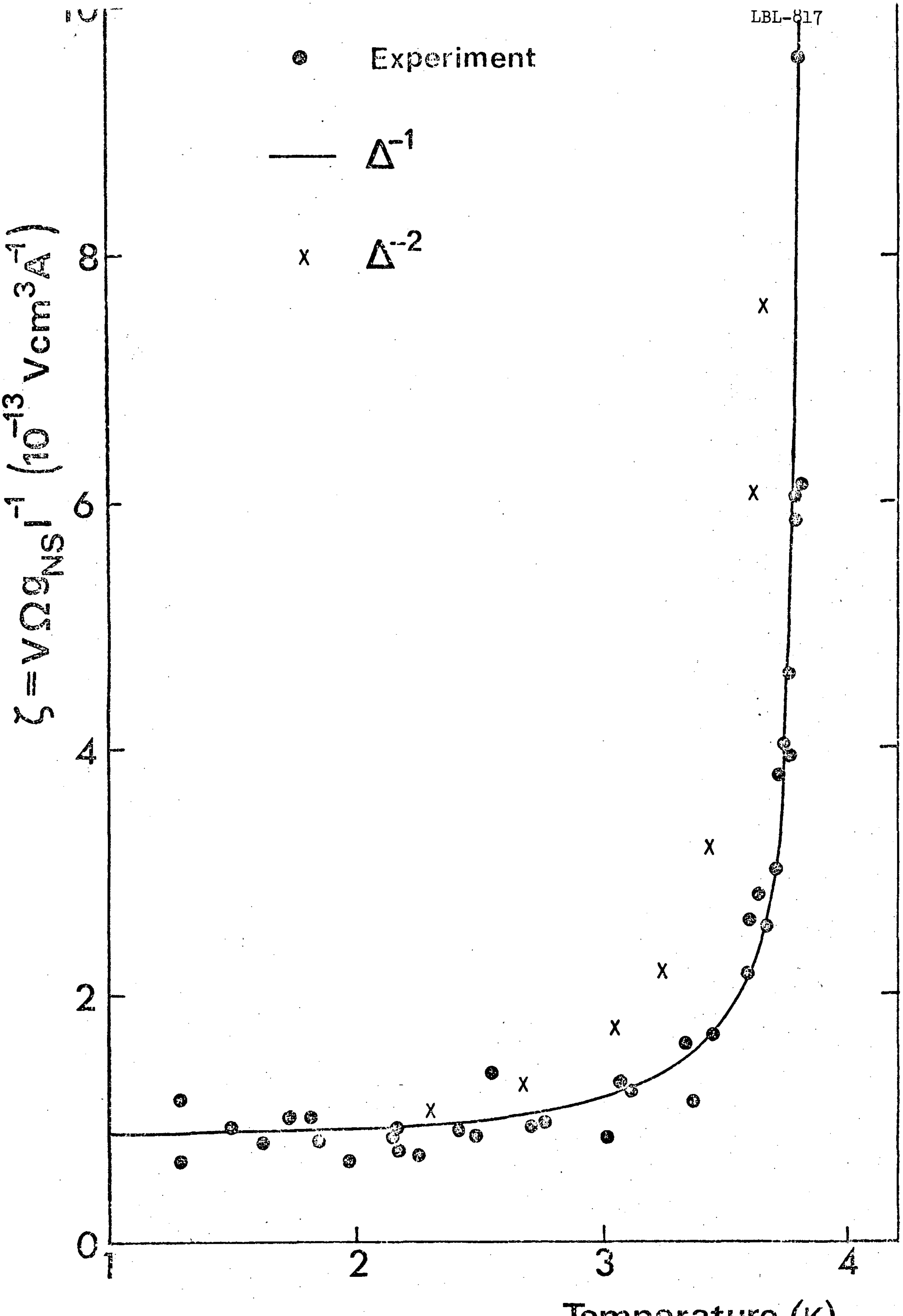

V (10^-10 V)
8
6
4
2
0
1
2
3
4
Temperature (K)
electron injection
electron extraction
2160Å
3010Å
4010Å
5260Å

FIG. 2



Experiment
Δ^{-1}
Δ^{-2}
$\zeta = V\Omega g_{NS} I^{-1}$ $(10^{-13}\ Vcm^3A^{-1})$
0
2
4
6
8
1
2
3
4
Temperature (K)

FIG. 3

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