PHYSICAL REVIEW B VOLUME 13, NUMBER 5 1 MARCH 1976

Tomasch-effect-induced negative-resistance phenomena in single-crystal superconducting Pb films[*]

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(Received 23 September 1974)

A negative-resistance region has been observed just above the gap in tunnel junctions fabricated on 〈110〉-oriented single-crystal Pb films. This phenomenon was found to be present only when tunneling into single-crystal Pb films about 1.4 μm thick. When these junctions were raised to temperatures above 3.7°K or placed in a parallel applied magnetic field above 200 G the negative resistance was not present although a deep minimum in conductance persisted. The structure was always accompanied by large Tomasch oscillations and appears to be related to the presence of the Tomasch effect and the details of the effect as they occur in 〈100〉-oriented single-crystal Pb films. The negative resistance and accompanying structure were reproduced in some respects by theoretical calculations using a superconductor density of states modified by the Tomasch term.

I. INTRODUCTION

Tunnel junctions fabricated on high-quality single-crystal Pb films backed by Ag have been found to exhibit large Tomasch oscillations. This effect is sometimes as much as 20% of the total dynamic conductance of the junction. Junctions prepared on a thin single-crystal Pb film with the tunneling direction along the 〈110〉 axis exhibit a sharp minimum in the dI/dV-V characteristics at energies just above the highest gap value in addition to the smooth Tomasch structure (see Fig. 1). This minimum was found to be negative for films 1.2 to 1.6 μm thick. The maximum negative resistance occurred when the film thickness was about 1.4 μm and sometimes exhibited double structure. Raising the film temperature or applying a magnetic field to the film caused the negative resistance to disappear. Junctions on single-crystal Pb films with the tunneling direction along axes other than 〈110〉 did not show a negative-resistance region, although some orientations did have a sharp minimum in dI/dV.

Calculations of the dI/dV characteristics using a superconductor density of states modified by the Tomasch oscillations[1] reproduced the negative-resistance region at the proper bias voltage but did not describe the thickness dependence. Since the Tomasch theories[1–3] do not include magnetic field or temperature effects these effects could not be entered directly in the calculation.

II. EXPERIMENTAL DETAILS

The single-crystal films used in this study were grown by a method developed by Schober.[4] In this method a polished KBr substrate,[5] that had been cut so its surface lies in the desired plane, was rapidly heated to 200 °C. Ag was then vapor-deposited at a rate of 1000 Å/sec directly onto the substrate surface which resulted in the epitaxial growth of the Ag. Immediately Pb was vapor-deposited, also at a rate of 1000 Å/sec, onto the Ag film and this produced the epitaxial growth of the Pb.

Pressures during film deposition were less than 5×10^{-6} Torr. The Ag film thickness was typically of the order of 1000 Å and the purity of both metals was 99.999%. The Pb film surface

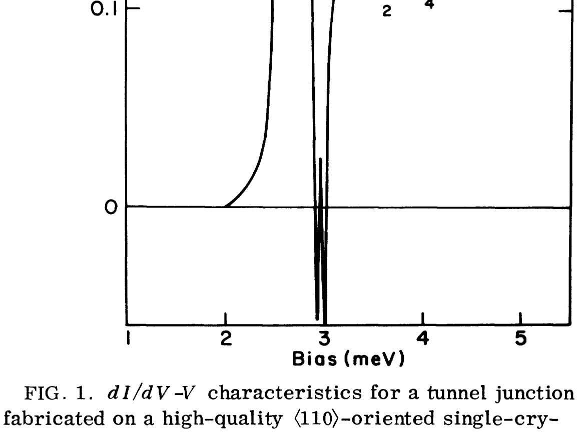

FIG. 1. dI/dV-V characteristics for a tunnel junction fabricated on a high-quality 〈110〉-oriented single-crystal Pb film 1.4 μm thick.

was then oxidized in dry oxygen for one hour, SiO was deposited to mask the film edges, and polycrystalline cross-strips were deposited to complete the tunnel junction. The substrate was immediately mounted on a probe where electrical connections to the junction were made with spring-loaded platinum contacts and the assembly was inserted into a glass Dewar system. If the junction impedence increased slightly on cooling to 77 K the junction was considered good and liquid helium was transferred into the cryostat. Pumping on the helium bath lowered the temperature to about 1.2 K. Temperature measurements were made using a 250-Ω carbon resistor to an absolute accuracy of ± 50 mK with a resolution of about 10 mK. Tunneling measurements yielded I - V, dI/dV- V, and dV/dI - V curves using a bridge network similar to that designed by Adler and Jackson.[6] The ratio of dI/dV measured inside the gap, near zero bias, to that measured outside the gap was less than 10^{-3} for all junctions used in this study indicating that there was little, if any, nontunneling current present.

The magnetic field dependence of the tunneling characteristics was measured with the junction in a magnetic field applied parallel to the plane of the film. The magnetic field was measured to ±1 G with a Rawson-Lush rotating-coil gaussmeter.

III. MEASURED TUNNELING CHARACTERISTICS

The shape of all I - V and dI/dV- V curves taken were similar to that shown in Figs. 1 and 2. There are three important features to note: (1) a negative-resistance region centered at 2.95 mV, (2) the double structure of the negative-resistance region which was not always present, and (3) the unusually large Tomasch oscillations that are a characteristic of junctions fabricated on very high quality single-crystal Pb films backed by a single-crystal Ag film. Tomasch oscillations have been observed as large as (20–30)% of the total dynamic conductance of the junction outside the gap.

It was found that the negative-resistance region only occurred in junctions fabricated on ⟨ 110⟩ - oriented Pb films with a thickness of 1.2 to 1.6 μm. Junctions on single-crystal Pb films of other orientations (⟨ 100⟩ and ⟨111⟩) did exhibit large Tomasch oscillations and sometimes a deep minimum in dI/dV just above the gap but a negative resistance was never produced by varying the film thickness. A deeper minimum was obtained with ⟨ 111⟩-oriented films than with ⟨ 100⟩-oriented films. The properties of the polycrystalline Pb cross-strip did not effect any of the tunneling characteristics.

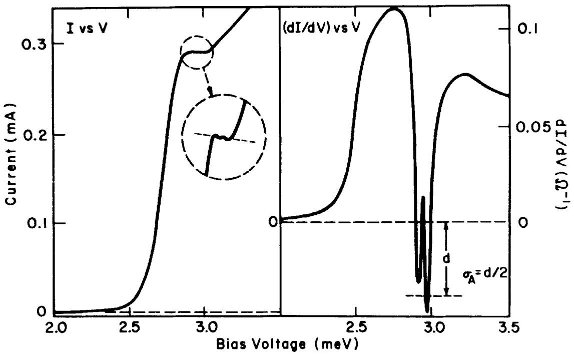

FIG. 2. Experimental I-V and dI/dV-V characteristics of a junction with ⟨110⟩-oriented Pb film. The vertical amplification has been increased by a factor of 30 in the inset and the slope of the dashed line is σ_A. Film thickness is 1.25 μm.

In order to show the thickness dependence of the magnitude of the negative resistance, it is necessary to define an average negative conductance σ_A which can be calculated from the dI/dV-V curves. The average negative conductance is defined as one-half the mean value of the maximum negative conductance in each negative peak of the doublet. Precision was somewhat limited owing to noise in dI/dV. This value, however,

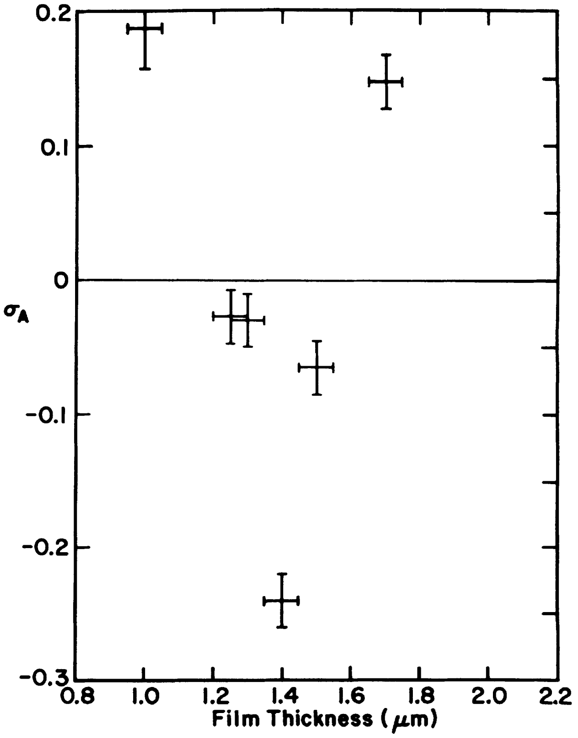

FIG. 3. Thickness dependence of the negative-resistance region.

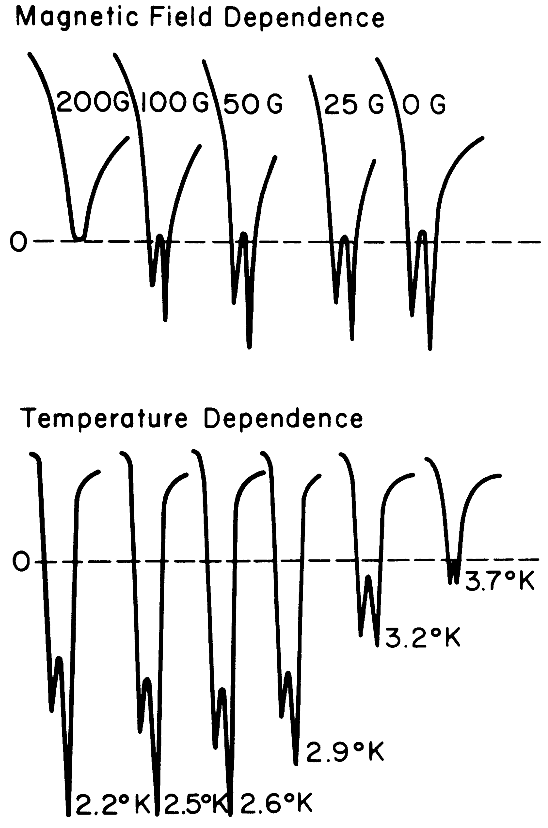

FIG. 4. Magnetic field and temperature dependence of the negative-resistance region.

corresponds quite well with the average slope of the negative-resistance region on the I-V curves in Fig. 2. The value of σ_A as a function of film thickness is plotted in Fig. 3 indicating that the negative resistance vanishes for film thicknesses above about 1.6 μm and below about 1.2 μm for ⟨110⟩-oriented Pb films. Film thicknesses were measured near the junction area to ±0.05 μm with a Rank Precision Talysurf 4. An increasing magnetic field applied parallel to the film, caused the size of the negative resistance to decrease and to finally disappear at fields of about 200 G. All doublet structure when present disappeared with the application of such fields. For fields less than 200 G the smooth Tomasch oscillations remain essentially unchanged, but when the field approaches 200 G the oscillations are reduced in amplitude and some smearing appears in the entire dI/dV curve. It is perhaps noteworthy that the parallel critical field for a 1000-Å-thick Ag film in contact with a Pb film, a few microns thick, has been measured by Valette[7] to be about 200 G.

Increasing the temperature also decreases the size of the negative resistance and the resistance is no longer negative above 3.7 K. There was no noticeable change in dI/dV for temperatures below 2.2 K. The detailed behavior of the negative-resistance region with increasing magnetic field and temperature is shown in Fig. 4.

IV. CALCULATED TUNNELING CHARACTERISTICS

Several aspects of the phenomenon suggest that it is related to the Tomasch effect. These are (i) The negative resistance seems to develop out of a sharp minimum in the Tomasch structure. (ii) The only thickness-dependent parameter in the expression for the tunneling current at 0 K appears in the Tomasch term. (iii) The anisotropy in $V_F/Z(0)$, the renormalized Fermi velocity, is greater than that of any other parameter in the expression for the tunneling current.[8,9] (iv) Inspection of the phases of the Tomasch oscillations in the density of states shows that ⟨110⟩ is unique in that all three oscillations (resolved using the technique of Haywood[9]) are nearly in phase and each is in the most extreme minimum at a junction bias of about 2.95 mV when the film thickness is 1.4 μm.

Calculation of the tunneling current was made using the Wolfram[1] model of the Tomasch effect at 0 K. The amplitude of the calculated oscillations in the 3–5 mV bias range was adjusted to that of the experiment by using the value of the gap in the Ag backing (Δ_{Ag}) as a fitting parameter. In order to estimate the size of Δ_{Ag}, the gap measurements of Marcus,[10] made by tunneling into thin Ag films backed by a Pb film, were used to derive the following expression:

$$\Delta_{Ag} = \Delta_{Pb}[1 - \exp(-0.14\, d_{Pb}/d_{Ag})],$$

where d_{Pb} and d_{Ag} are the thicknesses of the Pb and Ag films, respectively. This expression when evaluated for $d_{Pb} = 1.4$ μm and $d_{Ag} = 0.1$ μm predicts a value of Δ_{Ag} which is approximately 85% of the Pb gap, Δ_{Pb}. Though this value seems quite large a calculation of dI/dV predicted a 20% Tomasch effect which is in agreement with that observed experimentally.

The values of the single-crystal Pb gaps and associated Fermi velocities, V_F, and the contributions to the tunneling current are listed in Table I. $\Delta(0)$ is the zero-energy gap determined by using the energy dependence of the polycrystalline Pb gap[11] and extrapolating to zero energy. In this work $Z(0)$ was taken as 2.41. The contribution to the tunneling current for each gap was estimated from Blackford's[12] tunneling data on bulk single-crystal Pb. In Blackford's measurements two gaps were resolved for directions near ⟨110⟩ and the ratio of the tunneling currents

TABLE I. Parameters used in calculating *dI/dV* characteristics.

	$\Delta(0)$ (meV)	V_F (10^8 cm/sec) (Refs. 8, 9)	Tunneling current contribution
Δ_1	1.2007	1.98	0.4
Δ_2	1.3204	1.52	0.3
Δ_3	1.3386	1.28	0.3

TABLE II. Experimental and calculated peak positions in meV (see Fig. 1).

	1	2	3	4	5
Expt.	3.25	3.56	3.81	4.00	4.20
Calc.	3.29	3.50	3.70	3.98	4.33

was found to be about $I_2/I_1 = 1.5$ where I_2 is the tunneling current associated with the higher gap and I_1 the lower value. Since according to Bennett,[13] three gaps should be resolved (consistent with the observation of three Tomasch oscillations here) and since the two highest values are close these probably appear as one in Blackford's measurements. It is assumed that the contribution from each is equal and that $(I_2' + I_3')/I_1' = 1.5$ where I_1', I_2', and I_3' are the tunneling currents associated with the three resolved gaps.

A calculation of dI/dV made using the preceding assumptions contains a negative-resistance region at the proper bias voltage of 2.95 mV. However, the structure due to the presence of three gaps is extremely sharp and the conductance peak is too high. If an additional imaginary part of the Pb gap of − 0.01 mV is added, the structure in the region of the gaps is made less sharp and the height of the conductance peak corresponds better with the values observed in the experiments (Fig. 5). The depth of the negative resistance in the dI/dV curve is decreased by a factor of 2 and agrees with experiment. The miminum remains at 2.95 mV. Seah and Dy[14] in their theoretical studies of Tomasch oscillations find that such a contribution is important. Perhaps this can be partially justified by the fact that while these experiments were done at a temperature of 1.2 K, 0 K was assumed for the calculation. Positions of the maxima and minima of the other Tomasch oscillations in the calculated dI/dV curve are compared to those measured (Fig. 1) in Table II. The agreement, while not exact, is acceptable. It must be remembered that by changing the film thickness the oscillation frequency changes and any error in the measurement of film thickness will contribute to the above disagreement.

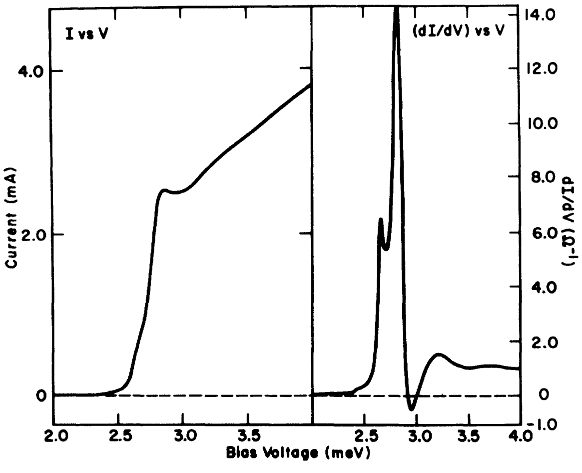

FIG. 5. Calculated tunneling characteristics for a junction with a ⟨110⟩-oriented Pb film 1.4 μm thick.

This model did not, however, predict the observed thickness dependence correctly or the existence of double structure in the negative-resistance region. The calculations predicted that there should be a negative resistance for single-crystal Pb films about 1.2 to 2.8 μm thick with the maximum negative resistance occurring at a thickness of about 1.9 μm and extremely sharp structure in the conductance peak was predicted.

The negative-resistance phenomena is only seen in films of the very highest quality presumably because a high degree of coherence must be preserved in the Tomasch oscillations if they are to interfere to exhibit the negative resistance. As the films are made thicker, coherence may not be sufficient even in the best films to preserve the negative-resistance phenomena. This would explain why our experimental data agree rather well with theoretical predictions near the minimum thickness for the phenomena but show a maximum in the effect and a disappearance of the effect at film thicknesses less than those predicted. Since the theories on the Tomasch effect do not include temperature or magnetic-field effects, these are not considered in our calculations.

V. CONCLUSIONS

Pb-*I*-Pb-Ag tunnel junctions with the single-crystal Pb film having a thickness in the range 1.2 to 1.6 μm and the ⟨110⟩ axis along the tunneling direction exhibit a negative-resistance region in their dI/dV-V characteristics. This phenomenon does not occur if the tunneling direction is along a principal crystallographic direction other than ⟨110⟩. The negative resistance region is always just above the highest measured gap which corresponds to a bias voltage of 2.95 mV. A theoretical calculation of the dI/dV-V characteristics using a superconductor density of states modified only by the Tomasch term reproduces the magnitude of the negative-resistance region at the proper bias voltage. The orientation dependence can also be accounted for using this model. The structure observed in the negative-resistance region, the temperature dependence, and the magnetic-field dependence cannot be accounted for on the basis of the Tomasch effect alone. Correlation of the magnetic field dependence with the reported critical field of the Ag layer suggests that the particular structure of the negative resistance may be tied to properties of the entire normal-metal backing and not just the superconductor–normal-metal interface. These proximity-effect properties do not seem to effect the general Tomasch oscillations.

ACKNOWLEDGMENTS

The authors would like to acknowledge K. K. Seah and K. S. Dy for helpful discussions of the theories.

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*This work was supported in part by the Materials Research Center, University of North Carolina at Chapel Hill under Grant No. GH-33632 from the National Science Foundation.

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