6 is a plot of $\ln(\alpha_s/\alpha_n)$ versus T_c/T for sound propagation in the basal plane. We have included the curves corresponding to the isotropic energy gaps of $2\Delta(0) = 3.1kT_c$ (upper solid) and $3.4kT_c$ (triangles) for comparison with the curves calculated from the two models. We find that the curves based on our models lie between the isotropic energy gap curves of 3.3 and $3.5kT_c$, the best fit being $3.4kT_c$. We note that the anisotropy observed by Lea and Dobbs[13] is approximately $0.3kT_c$, the approximate anisotropy expected from our models.

We have not compared the results of our two models with electronic specific heat and thermal conductivity measurements. However, such measurements on zinc indicate that the anisotropy in the energy gap is expected to be larger than in most superconductors.

IV. CONCLUSION

We find that microwave-absorption measurements in superconducting zinc can be described by a simple model for the anisotropic energy gap. The minimum quasiparticle energy, Δ_p, for an electron of momentum $\mathbf{p}=\mathbf{p}(\theta,\phi)$ is taken to be constant and equal to Δ_c for $0\leq\theta\lesssim\pi/6$. For $\pi/6\lesssim\theta\leq\pi/2$, Δ_p is constant and equal to $\Delta_a(0)=1.55kT_c$. The magnitude of Δ_c depends on whether or not one assumes that photon absorption occurs simultaneously with a diffuse scattering of quasiparticles at the surface of the metal. For the latter case, A, in which the initial- and final-state electron momenta are equal, $\Delta_c(0)=2.00kT_c$. For the former case, B, in which the initial- and final-state electron momenta are unrelated $\Delta_c(0)=2.45kT_c$.

We have compared our models for the anisotropic energy gap with the energy gaps obtained from measurements of various superconducting properties. Reasonable agreement is obtained but we are not able to decide, based on the present measurements, which of our models is correct. Tunneling experiments with single crystals should be decisive in determining the correct model and, therefore, the nature of photon absorption.

We would like to emphasize that our models are undoubtedly an oversimplification of the true anisotropic energy gap. The models are based on microwave-absorption measurements for which the angular averages involved can obscure any complicated variation of the energy gap. In view of this, our models are only descriptive of the gross features for the anisotropic energy gap.

ACKNOWLEDGMENT

I would like to thank Professor M. P. Garfunkel for helpful discussions on this work.

PHYSICAL REVIEW B VOLUME 1, NUMBER 9 1 MAY 1970

Superconducting Tunneling in Single-Crystal and Polycrystal Films of Aluminum*

G. L. Wells,† J. E. Jackson,‡ and E. N. Mitchell
University of North Carolina, Chapel Hill, North Carolina 27514
(Received 15 December 1969)

The energy gap (2Δ) and transition temperature (T_c) of aluminum films in Al-insulator-M tunnel junctions was measured. The orientation and crystallite size of the aluminum film was changed for different tunnel junctions by changing the substrate and/or substrate temperature at evaporation. The second metal (M) was either aluminum or indium. It was found that the strain on the aluminum film due to differential thermal expansion could account for most of the osberved variation in T_c. If M was indium, both 2Δ and T_c were changed. Empirically, it was found that T_c was increased by 3.2% and 2Δ was decreased by 3.3% owing to the indium film. This effect is not understood. The energy gaps of the aluminum films were $3.64kT_c$ for tunneling in the [100] direction, $3.52kT_c$ for tunneling in "isotropic" material, and $3.41kT_c$ for tunneling in the [111] direction. This anisotropy is in approximate agreement with theoretical predictions.

I. INTRODUCTION

ELECTRON tunneling between aluminum films and other metals has been extensively studied for the past several years, since aluminum films easily oxidize to form a tunneling barrier. However, with few exceptions, the characteristics of the aluminum films have not been studied in detail. Blackford and March[1] report the most extensive study of aluminum films and show that the gap ratio $2\Delta_0/kT_c$ for aluminum in Al-Al tunnel junctions on glass and quartz substrates is 3.53, the predicted weak-coupling value. In contrast to that experiment, most other measurements of the gap ratio in aluminum[2–4] have been made on Al-M tunnel junc-

* Work supported by the U. S. Air Force Office of Scientific Research under Grant No. AF-AFOSR-1241-67 and by the Advanced Research Projects Agency of the Department of Defense to the University of North Carolina Materials Research Center under Contract No. SD-100.

† Current address: Physics Department, University of Notre Dame, Notre Dame, Ind. 46556.

‡ Current address: Physics Department, University of Illinois at Chicago Circle, Chicago, Ill.

[1] B. L. Blackford and R. H. March, Can. J. Phys. **46**, 141 (1968).
[2] J. Nicol, S. Shapiro, and P. H. Smith, Phys. Rev. Letters **5**, 461 (1960).
[3] I. Giaever and K. Megerle, Phys. Rev. **122**, 1101 (1961).
[4] D. H. Douglass, Jr. and R. Meservey, Phys. Rev. **135**, A19 (1964).

tions (M not aluminum) and have been significantly less than 3.53. Some measurements on bulk single-crystal aluminum[5,6] have shown considerable energy-gap anisotropy which is not observed in typical polycrystalline aluminum films. It is also known that the transition temperature of aluminum is highly dependent upon stress.[7,8] This dependence may also effect the gap ratio.

The initial direction of this experiment was to measure energy-gap anisotropy in epitaxially grown aluminum films by electron tunneling. All of the tunnel junctions studied were of the form Al-insulator-M with M being aluminum or indium and the insulator most likely Al_2O_3. The crystal structure of the first evaporated aluminum film was changed for different tunnel junctions by changing the substrate and/or the substrate temperature at evaporation. As a consequence of these changes, the effects of stress on the aluminum films due to thermal expansion differences and the effects of indium as the second metal of the tunnel junction were studied as well as energy-gap anisotropy.

II. TUNNEL JUNCTIONS

All aluminum films used in this experiment were prepared by the evaporation of 99.999% pure aluminum from vacuum-degassed tungsten filaments at a pressure of less than 5×10^{-6} Torr. The aluminum film thickness could not be measured by standard optical techniques owing to cleavage steps in the KBr substrate. Transmission electron microscopy indicates that the films were approximately 5000 a.u. thick. When the aluminum was evaporated onto a cleavage face of a KBr crystal at 650°K, a single-crystal aluminum film was formed with the [100] direction normal to the surface. Figure 1 is an electron diffraction pattern from a 25-μ area of a typical film. The presence of Kukuchi[9] lines indicates good orientation parallel to the electron beam (i.e., normal to the surface). Figure 2 is a transmission electron micrograph of a typical film prepared in this manner. The dominant structure is not a defect in the film but is a result of destructive interference between a diffracted electron beam and the main transmitted beam.[10] In many films, these rays could be traced over distances of order of 1 mm without a deviation of the type which is expected at grain boundaries. This indicates that these aluminum films have very large crystallites.

Aluminum films evaporated onto KBr cleavage faces at 300°K had a dominant [111] texture axis as indicated in Fig. 3. This texture axis was apparent in all aluminum films forming the second member of the tunnel junction. Figure 4 is a transmission electron micrograph of a typical textured film. Note that the magnification is six times more than Fig. 2. The crystallites are of order 1 μ in diameter.

Aluminum films evaporated onto quartz substrates could not be removed for inspection in the electron microscope without severely damaging the film, and, consequently, the orientation and crystallite size are not known. However, the tunneling data are consistent with random orientation and small crystallites. Indium films which were used as the second metal of the tunnel junction were too thick for inspection in the electron microscope.

An aluminum film was always the first evaporated member of the tunnel junction. This film was oxidized, usually in laboratory air, to form a tunneling barrier. The second metal, either aluminum or indium, was always evaporated at room temperature. The resulting tunnel junction size was about $\frac{1}{2}$ mm square and the resistance was approximately 1000 Ω for Al-In tunnel junctions and 100 Ω for Al-Al tunnel junctions.

III. MEASUREMENTS

A ^{3}He cryostat was used to attain temperatures as low as 0.3°K. The temperature was determined using a Speer-type 1002(470-Ω) carbon resistor which was calibrated against the ^{3}He vapor pressure in the range 0.8–1.1°K using the T_{62} ^{3}He temperature scale. The temperatures corresponding to resistance values outside this range were found by extrapolation of a plot of log (^{3}He vapor pressure) versus log (resistance).[11] As the temperature measurements were most concerned with the aluminum film transition temperature, the transition temperature of a bulk aluminum slug was also measured during the course of each experiment. In this manner, the relative accuracy of the film transition temperatures was much better than the estimated 1% error in temperature measurement.

Most of the tunneling data consisted of a family of curves of dI/dV versus V taken with the temperature as a parameter. These curves were taken using circuitry similar to that described by Adler and Jackson.[12] Usually I-versus-V curves were also plotted at the lowest temperatures. These curves were extrapolated in a manner similar to that of Douglass and Meservey[4] to obtain the sum of the energy gaps, and this sum was used to define the correct extrapolation scheme for the derivative curves. The error in measurement of gap sums or differences, including both plotting and calibration errors, was less than $\frac{1}{2}$%.

The transition temperature of the aluminum film was

[5] L. T. Claiborne and R. W. Morse, Phys. Rev. **136**, 893 (1964).
[6] M. A. Biondi, M. P. Garfunkel, and W. A. Thompson, Phys. Rev. **136**, A1471 (1964).
[7] H. Notarys, Appl. Phys. Letters **4**, 79 (1964).
[8] P. N. Chubov, V. V. Eremenko, and Yu. A. Pilipenko, Zh. Eksperim. i Teor. Fiz. **55**, 752 (1969) [Soviet Phys.—JETP **28**, 389 (1969)].
[9] S. Kikuchi, Japan, J. Phys. **5**, 83 (1928).
[10] P. B. Hirsch, A. Howie, R. B. Nicholson, D. W. Pashley, and M. J. Whelan, *Electron Microscopy of Thin Crystals* (Butterworths, Inc., Washington, D. C., 1965), p. 283.
[11] S. Cunsolo, M. Santini, and M. Vicentini-Missoni, Cryogenics **5**, 168 (1965).
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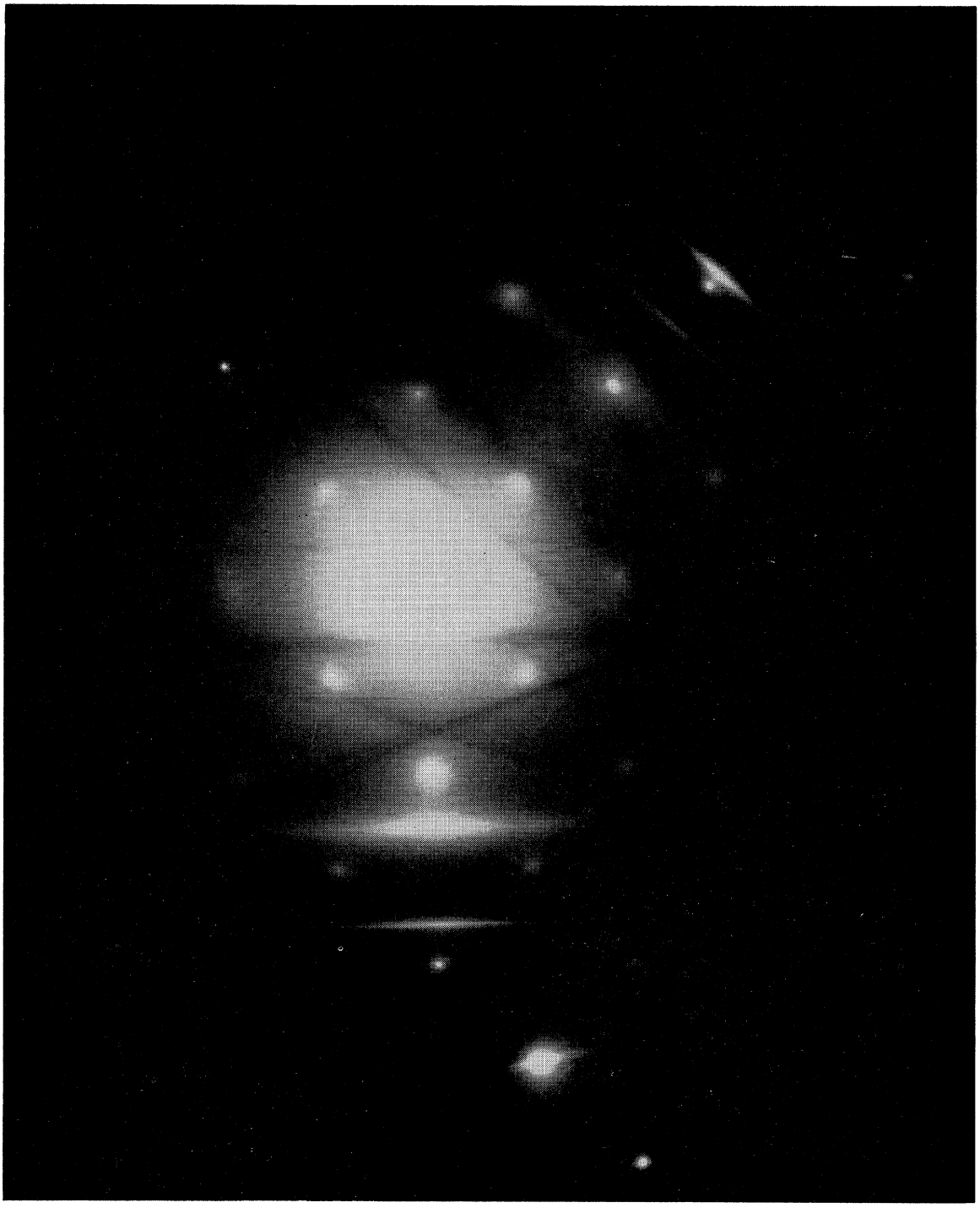

FIG. 1. Selected area electron diffraction pattern from single-crystal aluminum film.

defined by the disappearance of the energy gap. For Al-In tunnel junctions, this was distinctly visible, but for Al-Al tunnel junctions the disappearance was less distinct. Some of these tunnel junctions showed two nearly equal transition temperatures, and the transition temperature reported is therefore an "average" value. Even so, the relative error in transition-temperature measurement was less than 1%.

IV. DISCUSSION

Table I is a list of the energy gap $2\Delta_0$ and the transition temperature T_c of aluminum films for all

FIG. 2. Transmission electron micrograph of single-crystal aluminum film.

reliable tunnel junctions studied together with other pertinent data on the films as designated in the caption of the table. It should be noted that the energy gap listed for an Al-Al tunnel junction is not the energy gap for either film. The energy gaps cannot be separated in this case and the value in Table I is therefore the sum of half the energy gap in each film, i.e., $\Delta_1+\Delta_2$.

Figure 5 is a graphical presentation of the data in Table I. The bulk aluminum T_c shown is 1.96°K.[13] It is interesting to note that the T_c of aluminum films on KBr was lower than this value, while the T_c of aluminum films on glass as reported here and by

[13] J. F. Cochran and D. E. Mapother, Phys. Rev. 121, 1688 (1961).

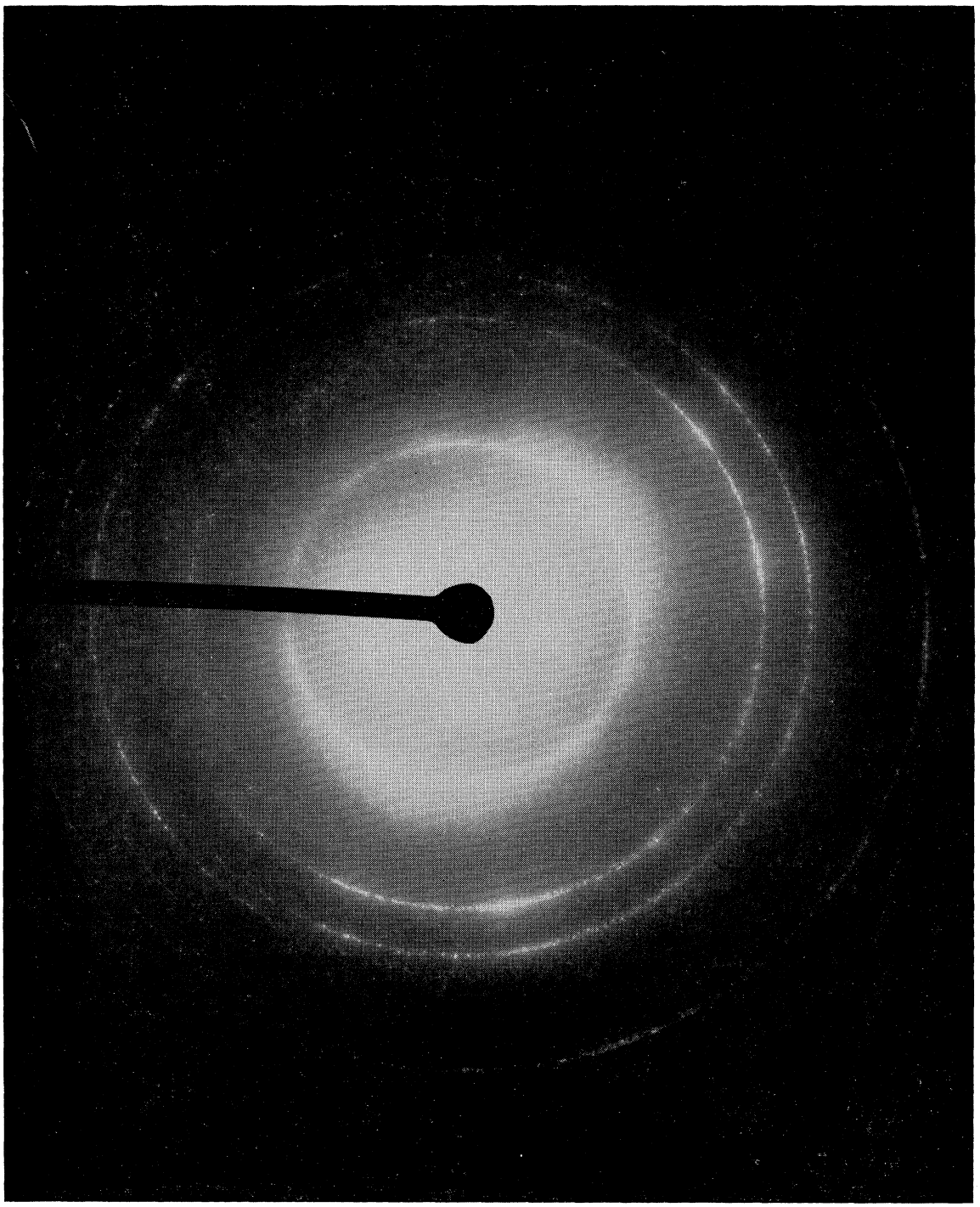

FIG. 3. Selected area electron diffraction pattern from polycrystalline aluminum film.

Blackford and March[1] was higher than this value. Satterthwaite[14] found that the T_c of bulk aluminum decreases as the mean free path decreases, but this cannot explain the low T_c of aluminum on KBr. These films are known to have a relatively long mean free path.[15] It will be shown that the different transition temperatures of the aluminum films can be explained in part by strain due to thermal expansion differences.

[14] C. B. Satterthwaite, Phys. Rev. **125**, 873 (1962).

[15] A. Von Bassewitz and E. N. Mitchell, Phys. Rev. **182**, 712 (1969).

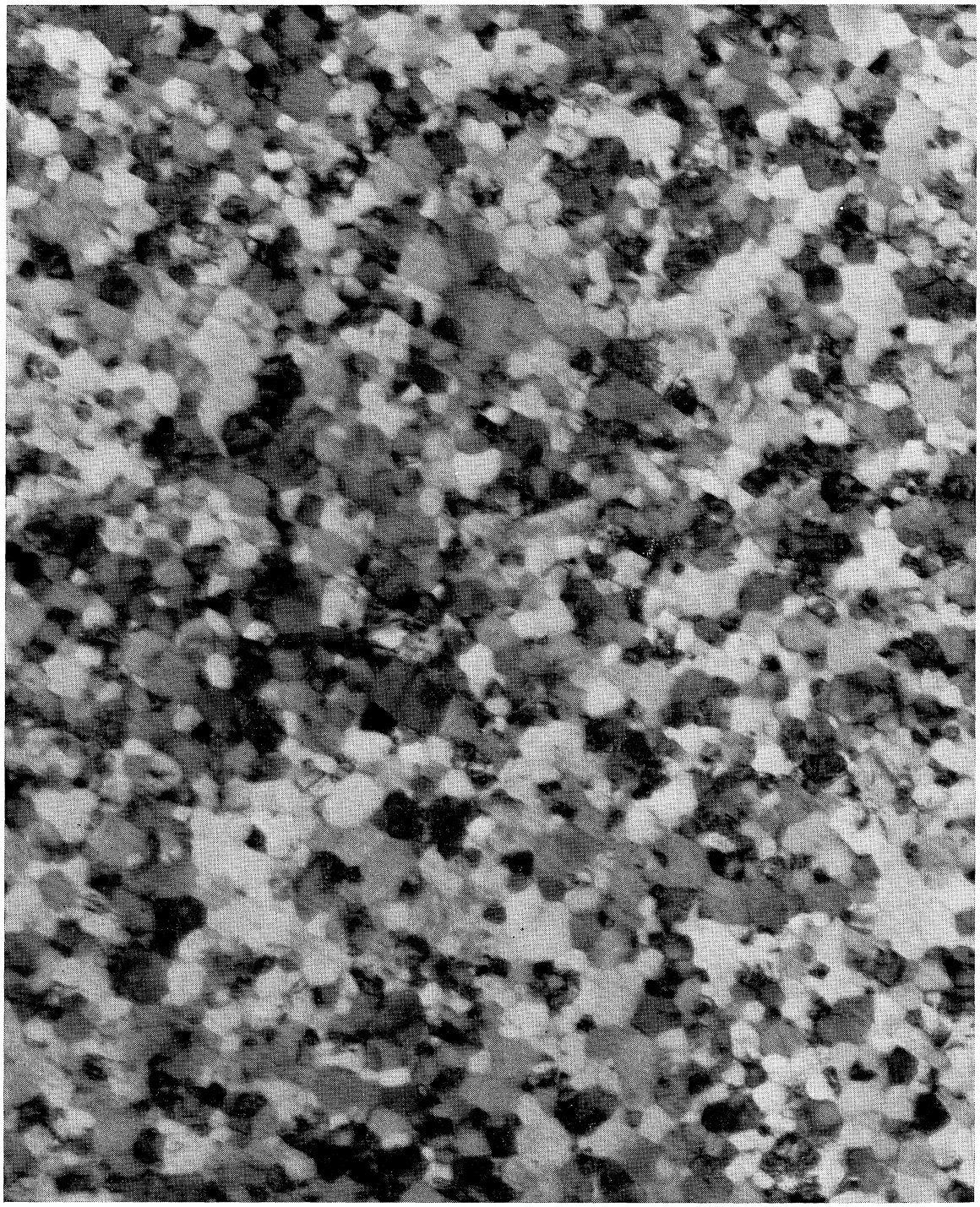

FIG. 4. Transmission electron micrograph of polycrystalline aluminum film.

Notarys[7] measured the transition temperature of aluminium films on Mylar substrates as a function of one-dimensional tensile strain. His results showed that the transition temperature of aluminum increases at the rate of 7°K per unit strain $[\Delta T_c = 7(\Delta l/l)°\mathrm{K}]$. The aluminum films in this experiment were not subjected to a strain of this type, but because of thermal-expansion differences they were subjected to a two-dimensional strain, either compression or extension. Chubov *et al.*[8] found that the change in transition temperature of aluminum as a function of strain due to thermal expansion differences between aluminum and a glass or

TABLE I. Measured data for each reliable tunnel junction studied. T_s: substrate temperature at evaporation of first film; O_1: orientation of first evaporated Al film; O_2: orientation of second evaporated film; T_c: transition temperature of Al film or Al-Al tunnel junction; 2Δ: energy gap at 0.3°K of Al in Al-In tunnel junctions or sum of one-half the energy gap in each film in an Al-Al tunnel junction.

Type of tunnel junction	Substrate	T_s (°K)	O_1	O_2	T_c (°K)	2Δ (meV)	$2\Delta/kT_c$
Al-In	Quartz	300	?	?	1.277	0.367	3.34
Al-In	Quartz	300	?	?	1.199	0.337	3.26
Al-In	Quartz	300	?	?	1.199	0.335	3.24
Al-In	KBr	300	[111]	?	1.130	0.318	3.27
Al-In	KBr	300	[111]	?	1.128	0.315	3.24
Al-In	KBr	300	[111[	?	1.161	0.320	3.20
Al-In	KBr	650	[100]	?	1.119	0.325	3.37
Al-In	KBr	650	[100]	?	1.128	0.333	3.43
Al-In	KBr	650	[100]	?	1.141	0.330	3.36
Al-In	KBr	650	[100]	?	1.110	0.328	3.43
Al-Al	Quartz	300	?	?	1.180	0.358	3.52
Al-Al	KBr	300	[111]	[111]	1.111	0.325	3.41
Al-Al	KBr	650	[100]	[111]	1.098	0.334	3.53
Al-Al	KBr	650	[100]	[111]	1.066	0.325	3.54
Al-Al	KBr	650	[100]	[111]	1.087	0.327	3.49
Al-Al	KBr	650	[100]	[111]	1.112	0.342	3.57

quartz substrate had a coefficient of 8°K per unit strain. Although their data are not as complete as Notarys's[7] results, the apparently different types of strain still produce the same change in T_c.

In order to estimate the net effective strain on an aluminum film (not necessarily part of a tunnel junction), the thermal-expansion coefficients of aluminum and the substrates were approximated as in Table II for the listed temperature intervals.[16] For convenience, the transition temperature was taken to be 1°K for all films. The net strains listed in Table III were calculated and the transition temperatures were computed using Notarys's coefficient[7] and 1.196°K as the unstrained bulk aluminum transition temperature.[17] The average value of the transition temperature for all types of Al-Al tunnel junctions as measured is also given in Table III for comparison. Transition temperatures of aluminum in Al-In tunnel junctions are not used since the presence of the indium film alters the transition temperature of the aluminum film, as will be discussed later. The agreement is remarkably good considering that the numbers used in estimating the strain and its effect are only approximate. The change in the gap ratio as a function of strain is considerably less than the change in the transition temperature. Our data cannot be used to show that it does not change, because the crystalline orientation of the films changes relative to the substrate and this alters the gap ratio.

Table IV contains the data obtained by averaging T_c, 2Δ, and $2\Delta/kT_c$ for each type of tunnel junction. So that the effects of the indium overlayer and the orientation of the first evaporated aluminum film can be directly observed, 2Δ for [100] Al-Al tunnel junctions refers to 2Δ for the [100] direction only. In order to

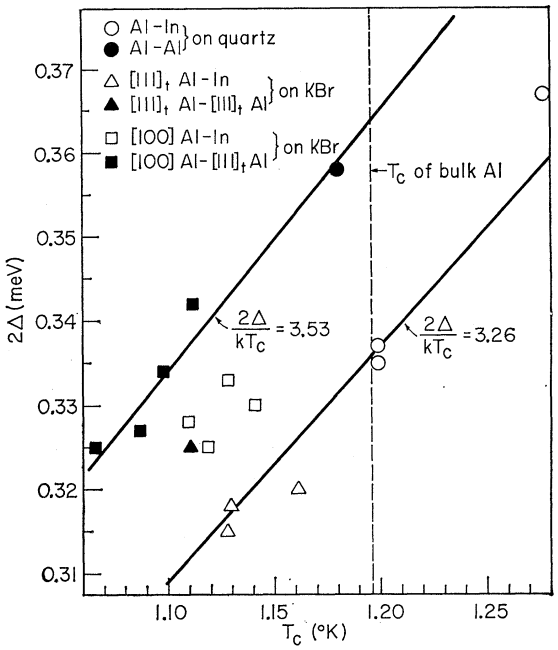

FIG. 5. Energy gap at 0.3°K of aluminum versus transition temperature. (a) Evaporated onto quartz at 300°K; (b) evaporated onto KBr at 300°K, aluminum film has [111] texture axis (designated $[111]_t$) (c) evaporated onto KBr, first aluminum film at 650°K ([100] single crystal), second film at 300°K ([111] texture axis if aluminum).

TABLE II. Approximate thermal-expansion coefficients.

Material	Temperature range (°K)	Expansion coefficient
Al	650–1	18×10^{-6}
KBr	650–1	40×10^{-6}
Quartz	300–1	13×10^{-6}
Glass	300–1	4×10^{-6}

TABLE III. Calculated strains on Al, the calculated transition temperature of Al films on various substrates using Notarys's expression, and experimental transition temperatures.

Evaporation temperature (°C)	Substrate	Strain	T_c(°K) Calculated	T_c(°K) Measured
650	KBr	−0.0143	1.096	1.091
300	KBr	−0.0066	1.150	1.111
300	Quartz	+0.0015	1.207	1.180
300	Glass	+0.0042	1.225	1.228[a]

[a] Measured by Blackford and March (Ref. 1).

[16] *Handbook of Chemistry and Physics* (Chemical Rubber Publishing Company, Cleveland, Ohio, 1957), 39th ed., pp. 2059–2063.

[17] R. David, Philips Res. Rept. 19, 524 (1964).

TABLE IV. Average energy gap and transition temperature of Al films for each type of tunnel junction. The entry for [100] Al-Al tunnel junctions has been corrected to be only the single-crystal film data. See text.

Type of tunnel junction	Substrate	First evaporated aluminum orientation	$\bar{T}_c$(°K)	$2\bar{\Delta}$(meV)	$2\bar{\Delta}/k\bar{T}_c$
Al-In	Quartz	?	1.225	0.346	3.28
Al-In	KBr	[111]	1.140	0.318	3.24
Al-In	KBr	[100]	1.125	0.329	3.40
Al-Al	Quartz	?	1.180	0.358	3.52
Al-Al	KBr	[111]	1.111	0.325	3.41
Al-Al[a]	KBr	[100]	1.091	0.343	3.65
Al-Al[a]	Glass	?	1.228	0.373	3.53

[a] Data for [100] orientation only. See text.
[b] Measured by Blackford and March (Ref. 1).

obtain this value of 2Δ, it is noted that the energy gap measured for a [100] Al-Al tunnel junction is the average of the energy gaps for the [100] direction and the [111] direction. The [111] direction energy gap was measured directly as $3.41kT_c$ and the average of the [111] and [100] gaps was measured as $3.53kT_c$. Therefore, the energy gap in the [100] direction only was taken to be $3.65kT_c$ (0.343 meV if $T_c=1.091$°K). The data in Table IV are presented graphically in Fig. 6.

The gap ratio for aluminum measured from an Al-In junction was less than the gap ratio from an Al-Al junction. This is believed to be a real effect; some caution is necessary, however, because the way T_c and Δ are measured differ for these two types of junctions. The manner in which the indium influences the aluminum is not understood. It is unlikely that this is a proximity effect. One reason is that the energy gap should be increased, not decreased, by the presence of the indium. The strain effects mentioned above do not seem to change the ratio $2\Delta_0/kT_c$, so strain caused by the indium seems to be an unlikely source of the effect observed. Although the explanation of the effect is not known, we feel that it is clear that the presence of the indium film changed both the energy gap and the transition temperature of the aluminum film. With this assumption, the data taken for aluminum films in Al-In tunnel junctions were empirically adjusted by comparison with the data for aluminum films in Al-Al tunnel junctions. The effects of the indium were assumed to be independent of substrate and orientation of the aluminum. A factor of 1.033 was used to adjust the values of 2Δ for Al-In films and a factor of 0.968 was used in the adjustment of the T_c for Al-In films. These corrected data are listed in Table V and shown in Fig. 6.

It appears from Fig. 6 that there is an energy-gap anisotropy in aluminum as determined by these experiments. The two most significant points, the [100] Al-In and the $[111]_t$ Al-In, are averages of data from at least three junctions including the corrections for the In overlay effect mentioned above. It should be noted

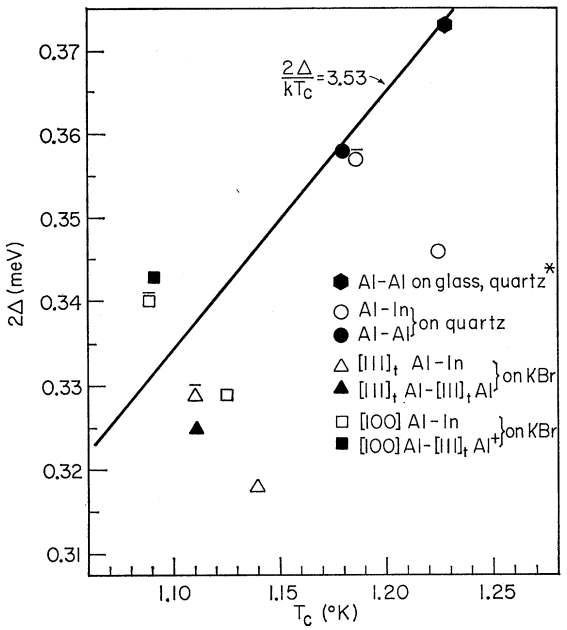

FIG. 6. Average energy gap at 0.3°K of aluminum versus transition temperature. Symbols are as in Fig. 5. The bar over the symbol indicates that the correction for the presence of indium has been applied (see text). The point (†) refers to [100] single-crystal aluminum only (see text). The data designated by (*) are taken from the literature (see Ref. 1).

that this correction does not change the relative locations of the two points.

Dynes and Carbotte[18] have calculated the energy gaps for several orientations of aluminum. Their value for the gap associated with the [100] direction is larger than the isotropic gap; that for the [111] direction is smaller. This is in qualitative agreement with the results reported here. Surface resistance measurements reported by Biondi *et al.*[6] on bulk single-crystal aluminium can be interpreted to agree with these general results, although a unique direction cannot be assigned to values of the multiple gaps they found.

Table VI compares the data reported here and the results of Dynes and Carbotte. The values for the energy gaps were calculated from the ratios $2\Delta_0/kT_c$ using the T_c for bulk aluminum.

The ratios for the [100] and [111] directions given in Table VI are the averages of the ratios determined from

TABLE V. Empirically adjusted values of the measured transition temperature and energy gap of Al in Al-In tunnel junstions.

Type of aluminum film and substrate	Adjusted average 2Δ	Adjusted average T_c
Al on Quartz	0.358	1.183
[111] Al on KBr	0.329	1.110
[100] Al on KBr	0.340	1.089

[18] R. C. Dynes and J. P. Carbotte, in Proceedings of the Conference on Superconductivity at Stanford, 1969 (unpublished).

TABLE VI. The theoretical and experiment values for the energy gap of Al as a function orientation.

Direction	Energy Gap Theory (Δ_0)meV	Experiment $2\Delta_0/kT_c$	Experiment (Δ_0)meV $(T_c=1.196)$
Isotropic	(0.18)		
[100]	0.192	3.64	0.187
[110]	0.187		
[111]	0.172	3.42	0.176

Al-Al junctions and Al-In junctions. These data are shown as the two points associated with each direction (using the shifted values of Δ and T_c for the Al-In junctions) in Fig. 6. The gap ratios are not appreciably changed by strain effects, and so from the ratios the energy gap can be calculated and compared to theory. The energy gap of the aluminum films on quartz substrates was found to be 0.182 meV. This result is not inconsistent with results predicted for isotropic specimens.

Both experimental values are too close to the isotropic energy gap. It is possible that this is due to the fact that these aluminum films are dirty superconductors ($l<\xi_0$) because the electron mean free path l was reduced by scattering from the film boundary to be less than the coherence length ξ_0 in a pure bulk superconductor. The mean free path (at least for pure lead films) is $l\sim d$,[19] where d is the film thickness. This probably means that the mean free path in 5000 Å films[20] is slightly greater than $\frac{1}{2}\xi_0$.

A remanent anisotropy, which we must be observing, has also been seen in thick PbBi alloy films where the mean free path of electrons in lead were limited by the bismuth impurity.[21] For the case of this lead system when l was approximately $\frac{1}{2}\xi_0$ the anisotropy was about $\frac{1}{2}$ of the full expected anisotropy of 15% for lead. The results reported here when compared to theory are in basic agreement with this interpretation of a remanent anisotropy.

[19] G. I. Rochlin, Phys. Rev. **153**, 513 (1967).
[20] Film thickness deduced from observations in electron microscope.
[21] C. K. Campbell, R. C. Dynes, and D. G. Walmsley, Can. J. Phys. **44**, 2601 (1966).

It is interesting to note that the dI/dV curves taken from all these junctions did not show structure in the conductance peak which is often seen in the case of junctions consisting of thick polycrystalline films[22] or bulk single crystals.[23,24] This structure is often associated with anisotropy. It is not clear why we should not see some evidence of this structure. For lead, it appears that the condition $l\sim 4\xi_0$ must hold to see structure associated with multiple gaps.[19]

Dowman *et al.*[25] have calculated that the selection criteria for predicting the wave vector for the tunneling electrons is a function of the nature of the insulating barrier. In particular, they claim that the selection rules for single-crystal oxides would be very different from those for amorphous oxides. No information is available on the nature of the insulator in these tunnel junctions except that they were grown near room temperature in either room air or oxygen and hence nothing definitive can be said regarding the structure of the oxide and its influence on the anisotropy studies reported here.

The work reported here was begun to investigate the anisotropy of the superconducting energy gap in aluminum. Anisotropy was observed and agrees very well with the predictions of Dynes and Carbotte.[18] The additional effects of strain on aluminum and the apparent influence of indium in Al-insulator-In junctions on aluminum were also observed and produce large changes in Δ and T_c compared to the effects of anisotropy on Δ. Assuming that the gap ratio remains constant under the influence of these effects, we are seeing genuine anisotropy effects.

ACKNOWLEDGMENTS

The authors would like to thank J. D. Landry and C. V. Briscoe for their helpful suggestions and aid in the performance of this experiment.

[22] C. K. Campbell and D. G. Walmsley, Can. J. Phys. **45**, 159 (1967).
[23] N. V. Zavaritskii, Zh. Eksperim. i Teor. Fiz. **45**, 1839 (1963) [Soviet Phys.—JETP **18**, 1260 (1964)].
[24] B. L. Blackford, Phys. Rev. (to be published).
[25] J. E. Dowman, M. L. A. MacVicar, and J. R. Waldrum, Phys. Rev. **186**, 452 (1969).

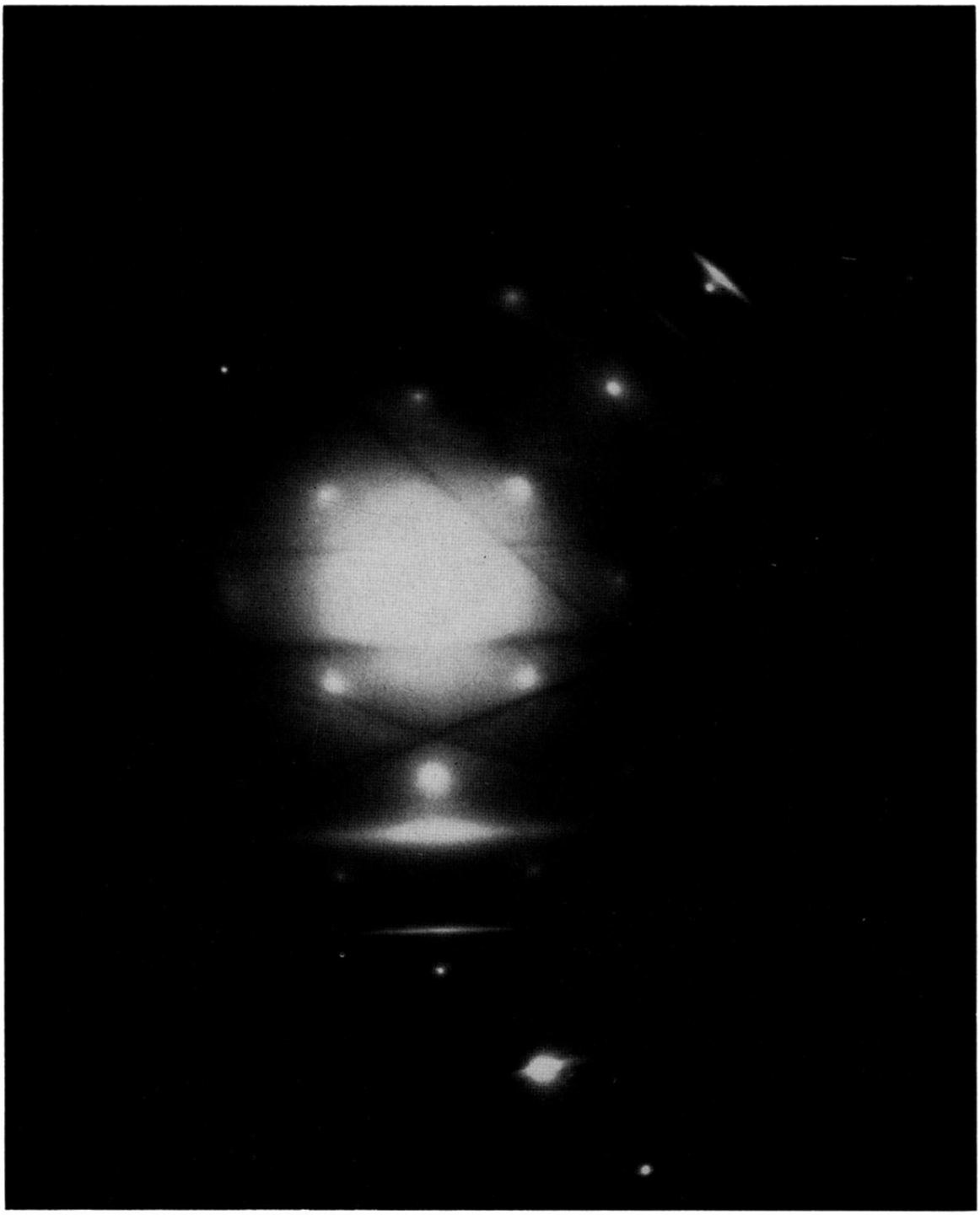

FIG. 1. Selected area electron diffraction pattern from single-crystal aluminum film.

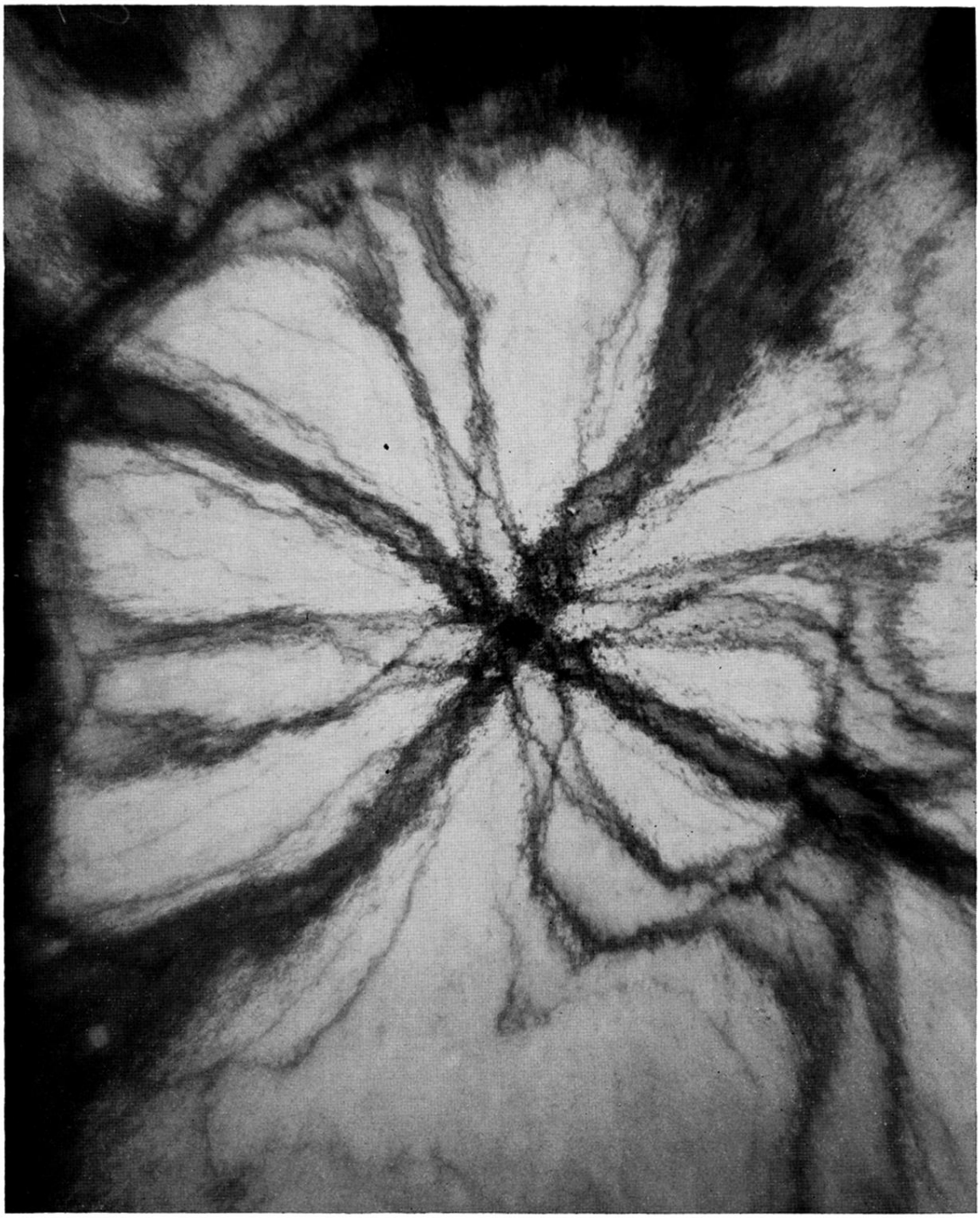

FIG. 2. Transmission electron micrograph of single-crystal aluminum film.

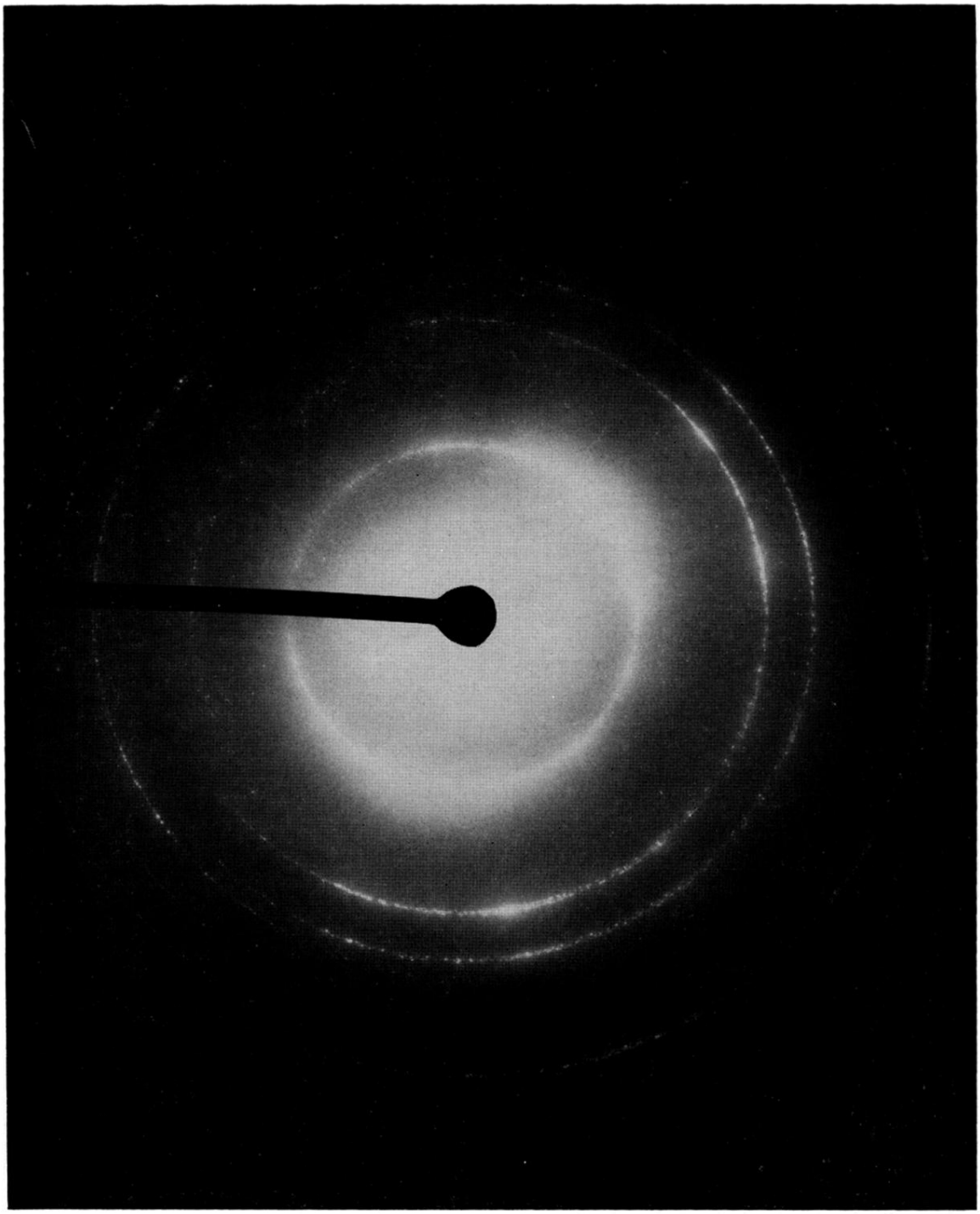

FIG. 3. Selected area electron diffraction pattern from polycrystalline aluminum film.

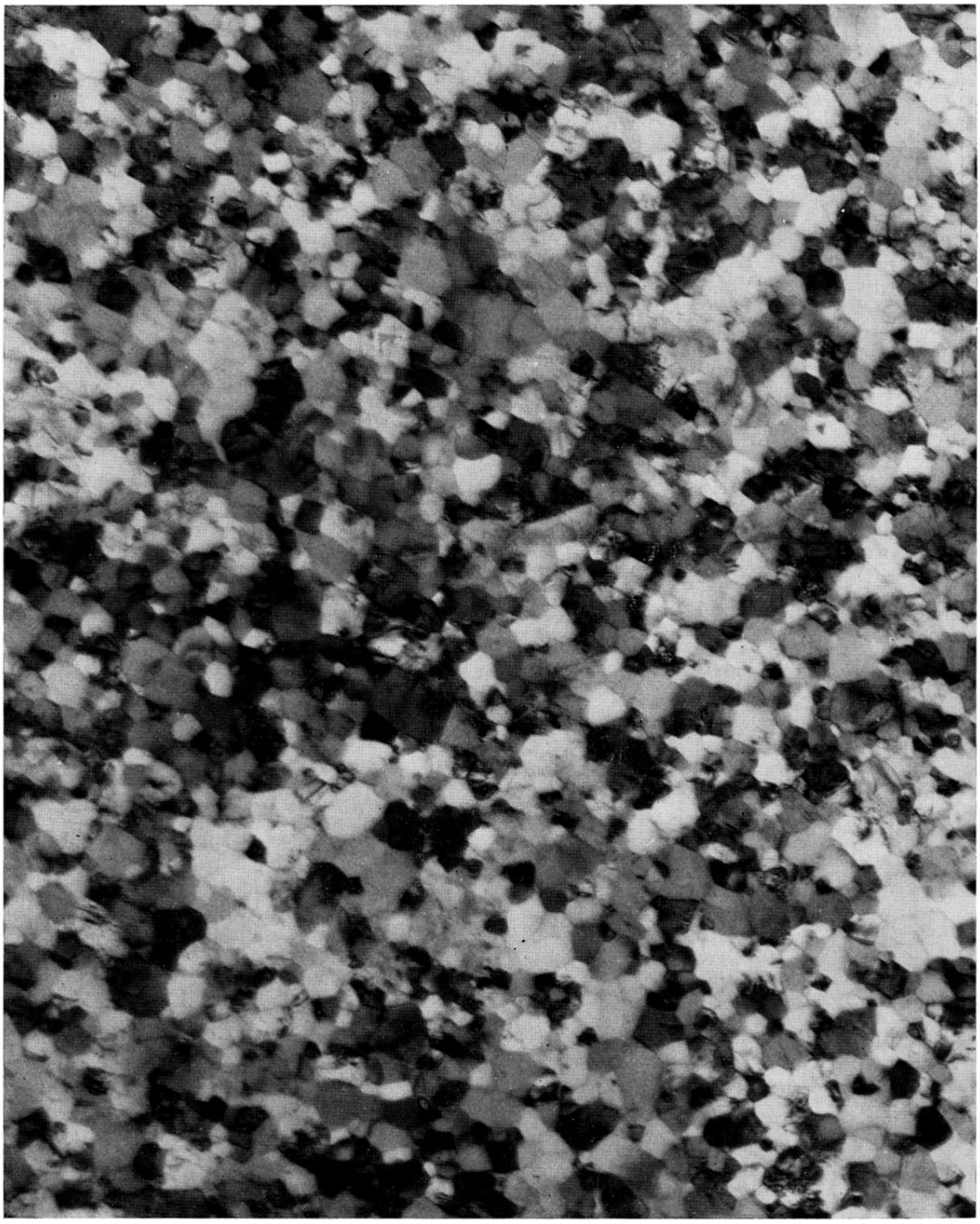

FIG. 4. Transmission electron micrograph of polycrystalline aluminum film.