TECHNICAL REPORT NO. 2

OFFICE OF NAVAL RESEARCH

Contract No. N0016-66-C0194

Req. No. NR017-467/? 28-66

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Prepared for Publication in

Physical Review

Department of Materials Science

State University of New York at Stony Brook

Stony Brook, N. Y.

November 1967

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ABSTRACT

The galvanomagnetic effects in antimony have been measured at low magnetic fields in the temperature range 50-300°K. It is shown that the behavior of the resistivity and the Hall coefficients at low temperatures is in qualitative agreement with the model of two overlapping bands as proposed by Windmiller and Falicov and Lin. However the initial increase in the Hall coefficient $\rho_{23,1}$ with temperature indicates the possibility of another hole band with the band edge lying close to the Fermi level.

INTRODUCTION

Recently band structure of antimony has been investigated using the de Haas-van Alphen effect [1-3], cyclotron resonance [4], ultrasonic attenuation [5-7], the Schubnikov-de Haas effect [8-10], and the galvanomagnetic effects [11-13]. The cyclotron resonance [4] and the de Haas-van Alphen effect [3] data have been analysed in terms of a Fermi surface consisting of two sets of closed pockets corresponding to electrons and holes respectively. The band structure calculations [14] for antimony are in accordance with the two band model and show that the electrons are located in three equivalent pockets centered at the points L of the Brillouin zone (fig. 1) while the holes are located in six equivalent pockets centered at points on the mirror plane σ near T. From the analysis of the de Haas-van Alphen effect, Windmiller [3] has estimated the carrier concentrations to be $N_e = 5.49 \times 10^{19}$ electrons/cm^3 and $N_h = 5.49 \times 10^{19}$ holes/cm^3 at liquid helium temperatures.

In order to explain the room temperature galvanomagnetic effects in antimony, Hall and Koenig have proposed two sets of holes such that one set provides the Hall conductivity in the trigonal plane required to give + ve Hall coefficient when the magnetic field is along the three-fold axis while the other set has a small effective mass in the trigonal direction thus making the second component of the Hall tensor positive. In the present work the galvanomagnetic coefficients have been measured at lower temperatures in order to arrive at a more conclusive model for the valence band in antimony.

EXPERIMENTAL DETAILS

High purity antimony (69 grade) was purchased from Consolidated Mining and Smelting Company and a large single crystal was grown by pulling the ingot through a horizontal furnace in the helium atmosphere. Several samples in the approximate dimensions of 12 mm x 2.5 mm x 2.5 mm were prepared by first cleaving along the trigonal plane and then reducing them to the final size by grinding. The orientation of each sample was checked with both the Laue pattern and the light figure technique.

The samples were mounted on a lucite holder in either the horizontal or the vertical position. The current and the potential leads were made using No. 38 copper wires soldered directly onto the samples with Cerro low alloy.

The potentials were measured using a Leeds and Northrup potentiometer and a high sensitivity dc null detector. The current passed through the samples was varied from 500 mA to 800 mA and the magnetic field from 100 gauss to 8000 gauss. The overall sensitivity of the electrical set up was such as to give an accuracy of ±0.1μV in the absolute measurement of the potential differences. The temperatures lower than the liquid nitrogen temperature were obtained by pumping on liquid nitrogen while liquid oxygen was used for the higher temperature. Complete galvanomagnetic measurements were thus made at four values of temperature in the range 50-95°K. Using a standard liquid helium cryostat, higher temperatures were obtained by adjusting the helium gas pressure in the inner dewar and filling the outer dewar with liquid nitrogen. A vapor pressure thermometer and an iron-constantan thermometer were used to measure the temperatures in the ranges 50-78°K and 78°K-300°K respectively.

RESULTS

According to the phenomenological theory, the relation between the electric field and the current density is given by:

$$E_i = \sum_j \rho_{ij}(\vec{H})J_j$$

where the resistivity tensor components ρ_{ij} are functions of the magnetic field. At low magnetic fields, $\rho_{ij}(\vec{H})$ can be assumed to have the form:

$$\rho_{ij}(\vec{H}) = \rho^{\circ}_{ij} + \rho_{ij,k}H_k + \rho_{ij,kl}\, H_k H_l$$

From the group theoretical analysis of $\bar{3}m$ symmetry crystals, Okada [16] and Juretschke [17] have shown that each of the ρ_{ij} and $\rho_{ij,k}$ has two independent components while $\rho_{ij,kl}$ has eight components. Following Zitter's work on bismuth, it was sufficient to prepare two sets of samples: one set having its length along the trigonal axis and the other set with the length along a binary. Samples with these two orientations were sufficient to measure all the twelve components of the resistivity tensor.

In all the measurements, the magnetic field was varied from 0.1 to 8 KG. Since at low temperatures the Hall and the magneto-resistance voltages showed considerable deviation from the linear and the quadratic dependence on the magnetic field, the coefficients were determined by extrapolation to zero field. Following this procedure it was possible to obtain a consistent set of resistivity tensor components at all temperatures. The results of the analysis are shown graphically in Figures 2 - 5 and in Table I.

INTERPRETATION AND DISCUSSION

As an initial step in interpreting the data, it is assumed that the Fermi surface in antimony consists of two sets of closed pockets corresponding to electrons and holes respectively. The cyclotron resonance as well as de Haas-van Alphen effect show that both sets are tilted in the trigonal-bisectrix plane by approximately 4° and 36° from the trigonal axis. In accordance with the band structure calculations, it is assumed in the present work that the small tilt pockets correspond to electrons having the effective mass tensor components[4] $m_{11} = 0.093$, $m_{22} = 1.14$, $m_{33} = 0.093$, $|m_{23}| = 0.082$ and the large tilt pockets contain holes with the mass components $m_{11} = 0.068$, $m_{22} = 0.63$, $m_{33} = 0.34$, and $|m_{23}| = 0.41$. For an isotropic relaxation time and an equal number of electrons and holes, the two components of the conductivity tensor are given by:

$$\sigma^\circ_{11} = \frac{Ne^2}{2m_0}\left[(\alpha_{11} + \alpha_{22})\tau_e + (\beta_{11} + \beta_{22})\tau_h\right]$$

$$\sigma^\circ_{33} = \frac{Ne^2}{\eta_0}\left[\alpha_{33}\tau_e + \beta_{33}\tau_h\right]$$

where the α's and the β's are inverse effective mass tensors components for electrons and holes respectively referred to the crystal axes coordinate system. The resistivity data in Figure 2 show that $\rho^\circ_{11}/\rho^\circ_{33} \simeq 1.5$. Using the values of the effective masses given above, this ratio would require $\tau_e \simeq \tau_h$. The equality of the relaxation times for electrons and holes would further give following ratio for the two Hall coefficients:

$$\frac{\rho_{12,3}}{\rho_{23,1}} = \frac{2\rho^\circ_{11}}{\rho^\circ_{33}} \frac{(\alpha_{11}\alpha_{22} - \beta_{11}\beta_{22})}{(\alpha_{11} + \alpha_{22})\alpha_{33} - \alpha^2_{23} - (\beta_{11} + \beta_{22})\beta_{33} + \beta^2_{23}}$$

Again using the Falicov and Lin's assignment and Datar's values of effective masses, the calculated value is $\frac{\rho_{12,3}}{\rho_{23,1}} \gtrsim 3$. The observed ratio in the present work (Figure 3) shows this ratio of the Hall coefficients approaching the calculated value in the lower temperature region. It appears from Steele's data [13] that $\rho_{12,3}$ at 4.2°K has almost the same magnitude as between 50° - 300°K implying thereby that it is only $\rho_{23,1}$ which is varying significantly with temperature. Since both the electrons and holes observed in cyclotron resonance and de Haas-van Alphen effect have Fermi energies of the order of 0.1 e.v., it is difficult to understand the increase in $\rho_{23,1}$ with increasing temperature on the basis of two overlapping bands alone. In order to explain the temperature variation of $\rho_{23,1}$ it becomes necessary to assume the presence of another hole band having the band edge close to the Fermi level and probably located at the point T in the BZ. Following the suggestion of Hall and Koenig, these holes with a low mass in the trigonal direction would provide a large Hall mobility when the magnetic field is along the binary or bisectcix axis. The increase in $\rho_{23,1}$ with increasing temperature would thus be a consequence of thermal excitation of holes from one hole band to another. Contrary to Hall effect, the contributions of electrons and holes to conductivity are additive and a small number of new holes would make very little contribution to the temperature dependence of the electrical conductivity. The data in Figures 2 - 5 indicate that the zero field resistivity coefficients ρ°_{ii} are approximately proportional to $T^{1.9}$ and $T^{1.4}$ below and above 90°K respectively while most of the magnetoresistivity coefficients $\rho_{ijkl} \sim T^{-1.4}$. These variations are also in better agreement with the band structure model of one conduction band overlapping with two valence bands as proposed above.

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#Work supported in part by the U. S. Office of Naval Research.

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TABLE I

Galvanomagnetic coefficients of antimony at room temperature.

ρ°_{ii} in $10^{-6}\Omega$-cm, $\rho_{ij,k}$ in $10^{-10}\Omega$-cm/G; $\rho_{ij,kl}$ in $\frac{10^{-15}\Omega\text{-cm}}{G^2}$

Galvanomagnetic Coefficients	Present Work	Freedman-Juretschke	Epstein-Juretschke
ρ°_{11}	43	43	44.3
ρ°_{33}	30	36	34.6
$-\rho_{23,1}$	2.0	2.2	2.05
$-\rho_{123}$	2.3	2.5	2.34
$\rho_{11,22}$	32	19.9	16.5
$\rho_{11,33}$	6.6	6.4	5.1
1	8.8	13.6	10.8
$\rho_{11,11}$	7.8	7.3	6.6
$\rho_{33,33}$	2.2	5.2	2
$\rho_{23,23}$	-	-2.8	-1.5
$\rho_{11,23}$	5.6	3.5	2.6
$\rho_{23,11}$	-	1.4	2.1

Figure Captions:

Fig. 1: The Brillouin zone for antimony (From Ref. 14).

Fig. 2: Zero-field resistivity tensor components as a function of temperature.

Fig. 3: Small field Hall coefficients as a function of temperature.

Fig. 4: Longitudinal galvanomagnetic coefficients $\rho_{ij,kl}$ as a function of temperature.

Fig. 5: Transverse galvanomagnetic coefficients $\rho_{ij,kl}$ as a function of temperature.

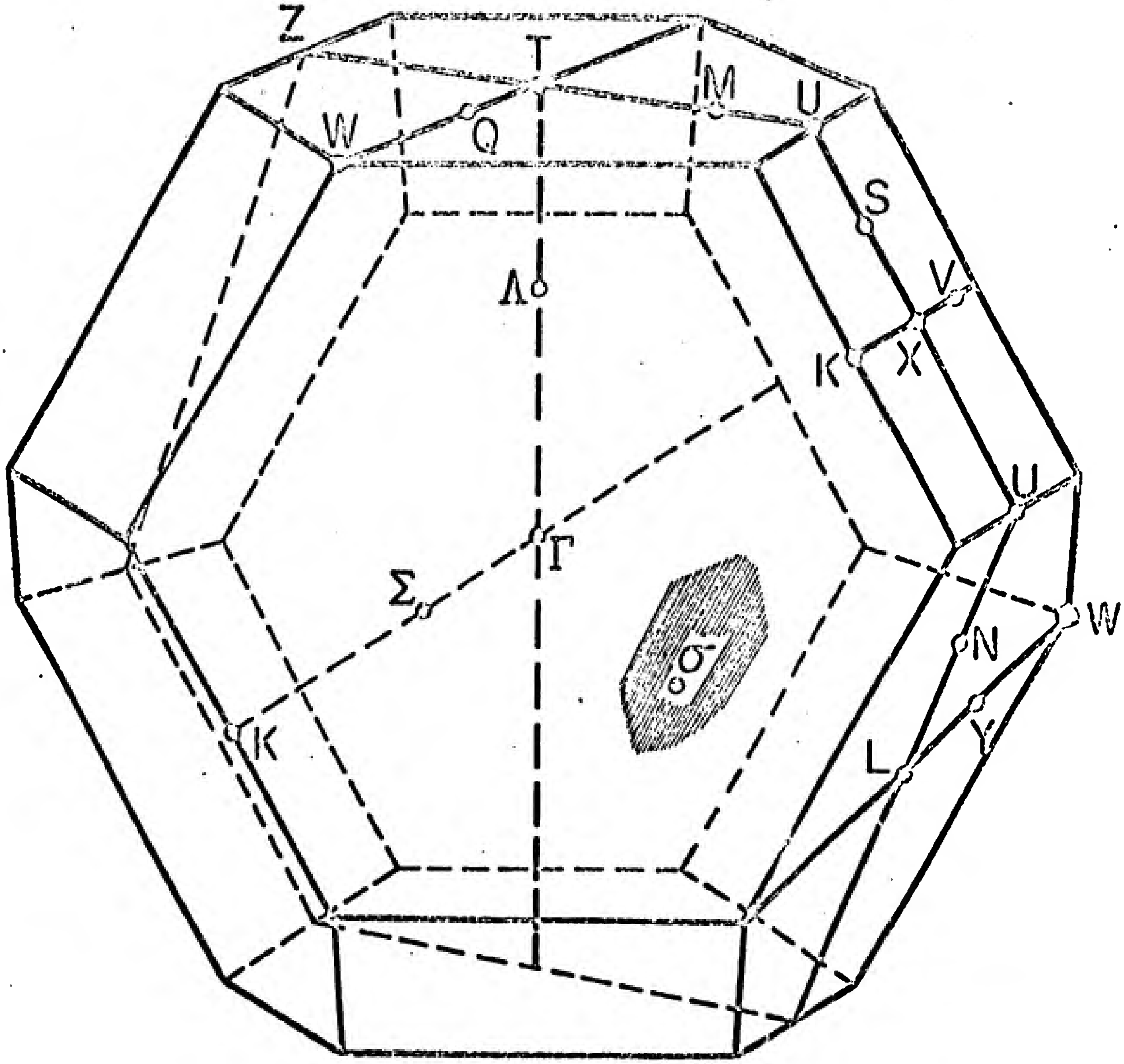


Fig. I

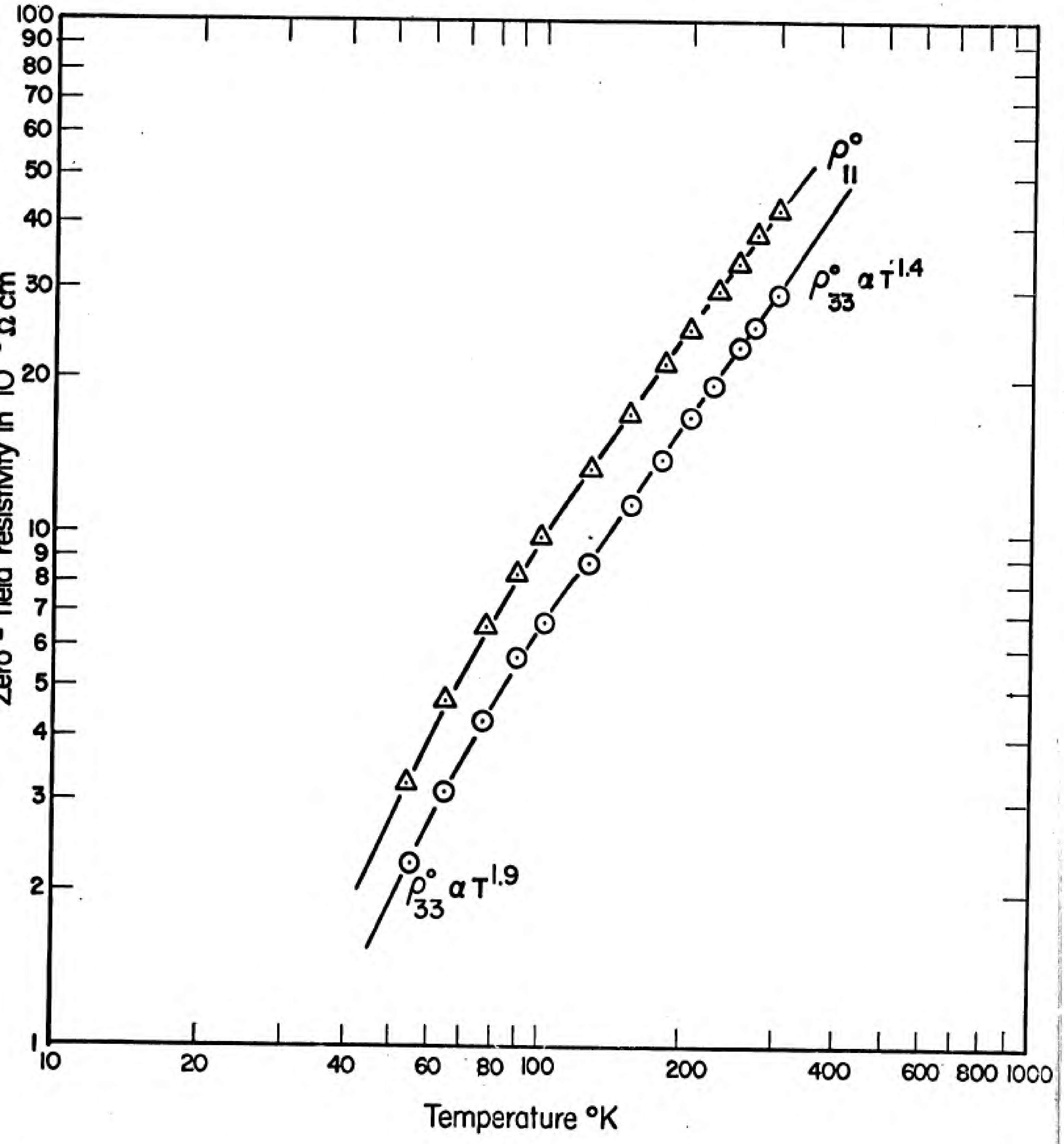
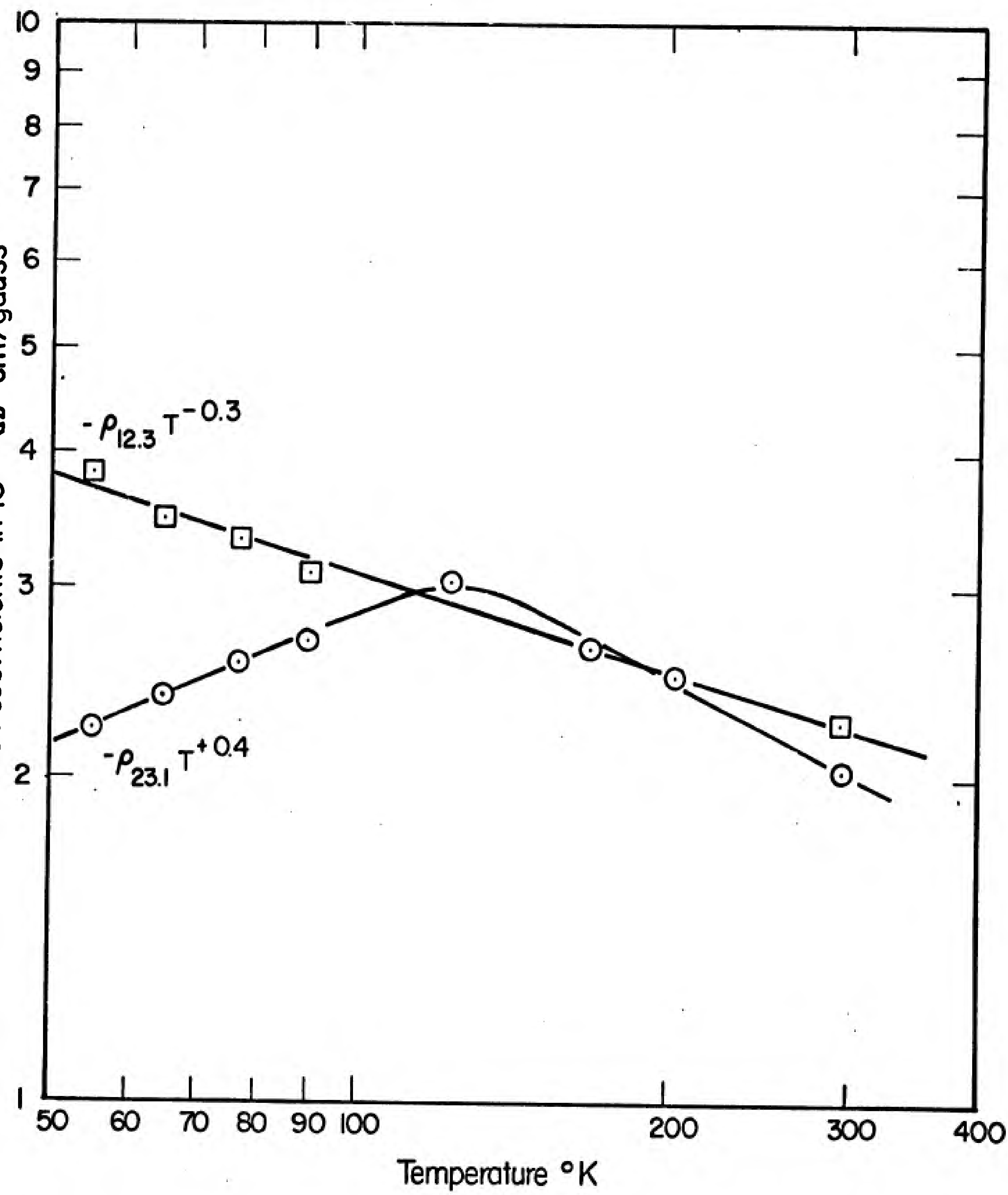
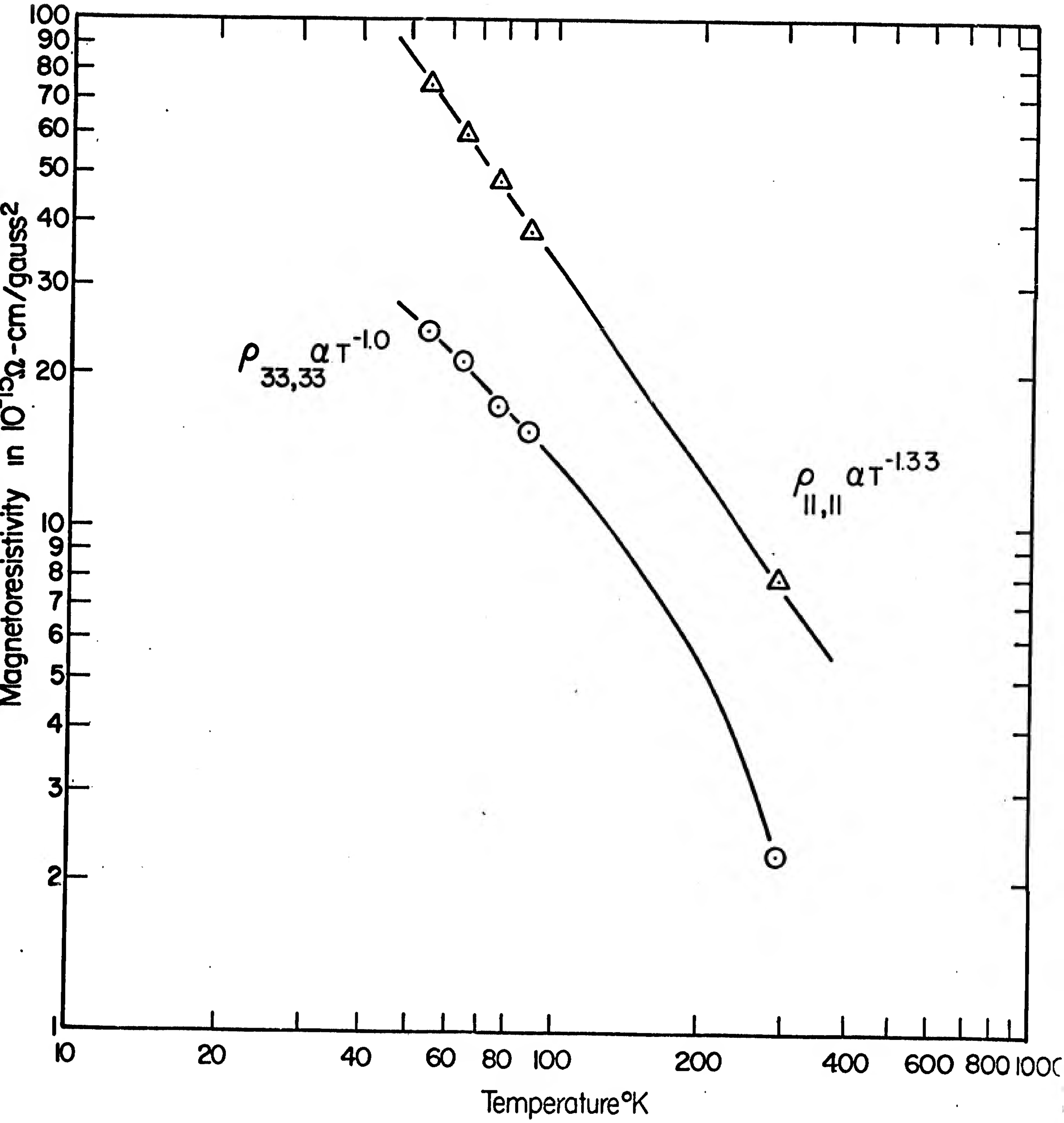


FIG. 2



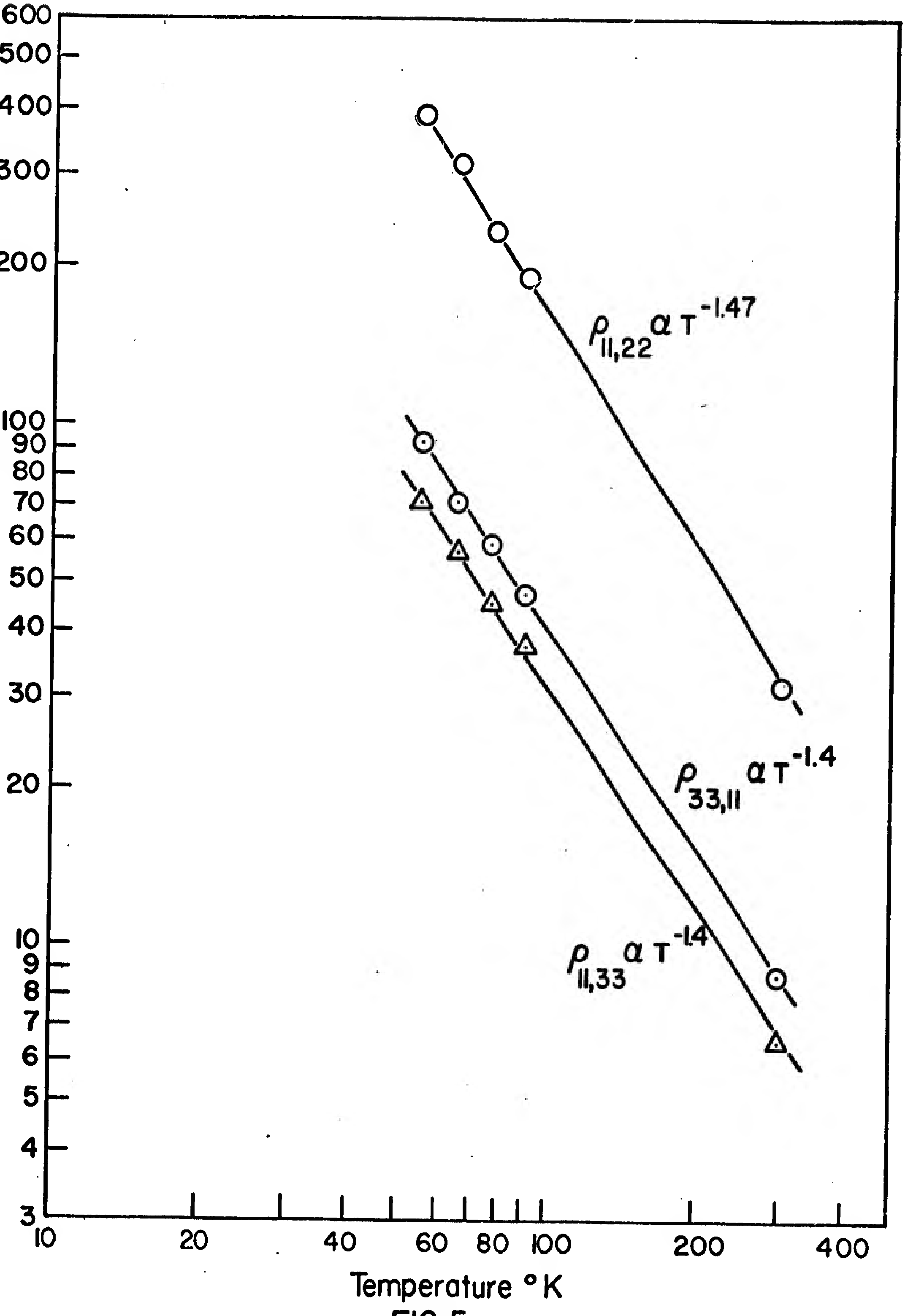
10
9
8
7
6
5
4
3
2
1
$-\rho_{12.3}\,T^{-0.3}$
$-\rho_{23.1}\,T^{+0.4}$
50 60 70 80 90 100 200 300 400
Temperature °K

FIG. 3



Magnetoresistivity in 10⁻¹⁵Ω-cm/gauss²
100
90
80
70
60
50
40
30
20
10
9
8
7
6
5
4
3
2
1
$\rho_{33,33} \alpha T^{-1.0}$
$\rho_{11,11} \alpha T^{-1.33}$
10
20
40
60
80
100
200
400
600
800
1000
Temperature°K

FIG. 4


600
500
400
300
200
100
90
80
70
60
50
40
30
20
10
9
8
7
6
5
4
3
10
20
40
60
80
100
200
400
ρ11,22 α T-1.47
ρ33,11 α T-1.4
ρ11,33 α T-1.4
Temperature °K
FIG. 5

Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author) Department of Materials Science State University of New York Stony Brook, New York	2a. REPORT SECURITY CLASSIFICATION Unclassified 2b. GROUP

3. REPORT TITLE
Galvanomagnetic Effects in Antimony

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)
Technical Report No. 2, November 1967

5. AUTHOR(S) (First name, middle initial, last name)
K. Tanaka, S. K. Suri and A. L. Jain

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
November 1967		18

8a. CONTRACT OR GRANT NO. N00016-66-C0194 b. PROJECT NO. c. NR017-467/2-28-66 d.	9a. ORIGINATOR'S REPORT NUMBER(S) 2 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report) none

10. DISTRIBUTION STATEMENT
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11. SUPPLEMENTARY NOTES none	12. SPONSORING MILITARY ACTIVITY Department of the Navy Office of Naval Research Physics Branch

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