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MAGNETICALLY SENSITIVE ELECTRICAL RESISTOR MATERIAL

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ABSTRACT

Theory. The theory of Davis and of Seitz on the magneto resistance effect is summarized and its extension in several directions is begun. Formulas are developed for the case where the magnetic field H does not lie along the k_3 axis, and for the case where the energy E and the relaxation time t are not even functions of k_1 , k_2 , and k_3 . The case where the temperature is not close to absolute zero is briefly indicated, and the necessary formalism is also set up to deal with noncubic crystals. A new theorem is found regarding the absence of any magneto-resistance effect, either at a low temperature or at all temperatures.

Experiment. Experiments are described measuring $\Delta\rho/\rho$ under various circumstances. Using a field of the order of 500 Gauss, most of the experiments deal with Bi and its anisotropy effects under various conditions of preparation. The results obtained are not essentially new, but represent our effort to get down to basic quantities from the measurements. A few other materials have also been measured, in view of the temperature requirements stated in the contract, but so far no material with strong and temperature—insensitive MR effect has been discovered.

In neither the experimental nor the theoretical phase did we encounter any unusual difficulties.

FORTHCOMING PUBLICATIONS

Theoretical aspects of the MR effect will be presented in a paper by L. P. Kao and E. Katz at the meeting of the American Physical Society, Detroit-Ann Arbor, Michigan, March 18-19-20, 1954; see the Bulletin of the American Physics Society, 29, 35 (1954), V8. The paper will contain essentially the leading ideas of the theoretical part of this report, augmented by some extensions and elaborated after the closing date of the report. It is intended to publish the theory, but not until it has been developed much further than is the case at present.

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A. INTRODUCTION

As was anticipated at the end of the previous quarterly report (in the section "Plans for the Immediate Future"), work has been undertaken on certain experimental and theoretical developments. Accordingly this report is divided into a theoretical part, and an experimental part.

The theoretical part describes what has been done in extending the Davis theory of the magneto-resistance effect. This work was done by L. P. Kao and E. Katz. (Mr. Kao has been with this project for only two months.) It was necessary to state in quite a few places that this or that problem will be worked out further as the aim during this period was primarily the exploring of the possibilities and directions in which the theory could be extended with a reasonable amount of labor. As a result of this exploration we feel certain at present that a great deal of significant information lies within reach just ahead, as may be estimated by the results obtained in this relatively short period.

The experimental part describes measurements on bismuth. This work was done by Mr. Tantraporn and Mr. Patterson, with Professor E. Katz supervising. This work produced a number of leads, described in this report, which it is hoped to investigate in the near future.

B. THEORETICAL PART

1. Motivation

The motivation for this study was derived primarily from a sentence in A. H. Wilson's book dealing with the magneto-resistance effect of real metals as measured, for example, by Justi and Scheffers. This sentence states:

"It does not seem likely that any model, simple enough to be tractable theoretically would give a magneto-resistance curve of the complexity of those actually observed." This complexity is seen in Fig. 1, which is taken from E. Justi's book. The personnel of this project agree with Wilson that the problem is not simple, but it is felt that the difficulty is one of labor and persistence rather than of principle. This view is supported by the fact, indicated in the previous report, that the theory of Davis, which is closely related to earlier work of Wilson, is limited to a number of restricted conditions which can be generalized without essential difficulty. Such conditions are special orientation and symmetry of the sample, very weak fields for which only the H² term is significant, and very low temperatures.

2. Outline of the Davis Theory

The eigenfunctions of conduction electrons in an unperturbed periodic lattice are of the form

$$\psi_{k} (\underline{r}) = e^{i\underline{k}\cdot\underline{r}} u_{k} (\underline{r}) ,$$

where u has the periodicity of the lattice. The number of electrons whose wave vectors lie in the range \underline{k} and $\underline{k}+d\underline{k}$ is $(1/4\pi^3)$ f (\underline{k}) dk, where dk stands for dk₁ dk₂ dk₃. With perturbing electric and magnetic fields \underline{F} and \underline{H} the equation of Boltzmann which expresses a stationary condition becomes

$$-(\epsilon/\hbar) [F + v_k \times H/c] \cdot \nabla f + (f - f_0)/t (k) = 0$$
 (1)

Here $-\epsilon$ is the charge of the electron, v_k is the group velocity of a wave packet about \underline{k} , and f_0 is the value of f when $\underline{F} = \underline{H} = \underline{O}_{\bullet}$. It is assumed that

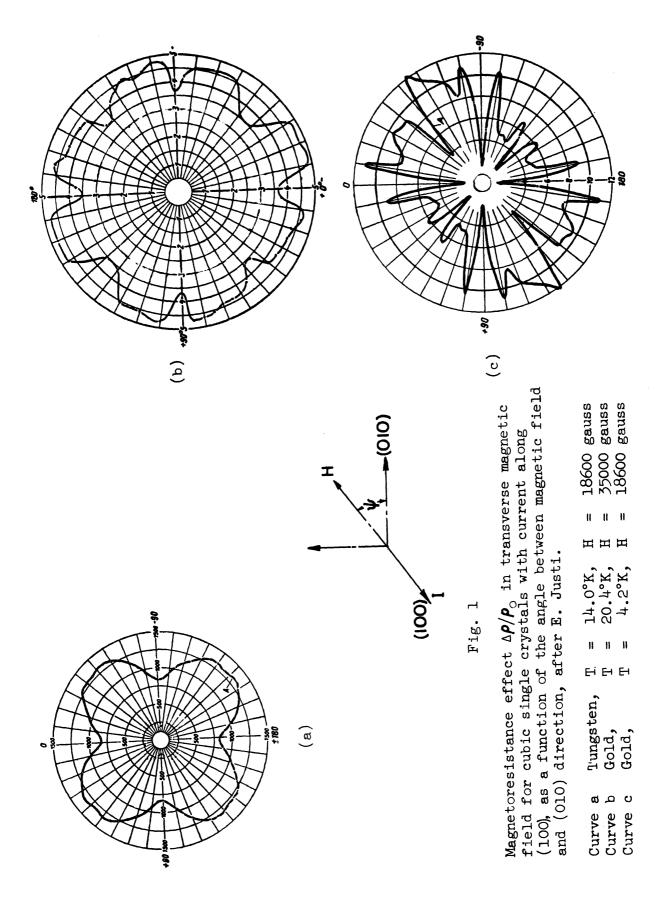
- I. a relaxation time t (k) can be defined, and
- II. interband transitions can be neglected.

The gradient is taken in k-space. A discussion of some conditions for the validity of Equation 1 is found in Wilson's book and in a paper by Jones and Zener⁴. Assumption II is discussed by C. Zener⁵. For \underline{v}_k we can write

$$\underline{v}_{k} = (1/\hbar) \nabla E(\underline{k})$$
, (2)

where E (k) is the energy eigenvalue belonging to k. While Equation 2 is usually credited to Zener, it was not until 1951 that its validity was established in the presence of a magnetic field by Luttinger⁶.

A great deal remains to be investigated about the precise limits of validity of Equations 1 and 2. However, these equations will be used as



starting assumptions to see what can be derived from them regarding the MR effect. Essentially this approach was also followed by Davis.

In solving Equation 1 Davis introduces:

Restriction 1: The field H lies along k3.

Assumption III: The material is isothermal; \underline{H} and \underline{F} are homogeneous throughout the material.

Davis then defines the function ϕ (<u>k</u>) and the operator Ω_3 (Ω in Davis' notation) by:

$$f = f_0 - \emptyset (k) \partial f_0 / \partial E = f_0 + \emptyset (k) g (E) , \qquad (3)$$

and

$$\Omega_{3} = -(\nabla E \times \nabla)_{3} \quad (4)$$

The function g is a peak function about the Fermi energy level $E_{\rm O}$ and approaches the Dirac delta function for low temperatures; i.e., for any reasonable function P (E) we have

$$\int g (E) P (E) dE \approx P (E_0) . \qquad (5)$$

The operator Ω_3 can be considered as the third component of a vector operator. It has the important property that when it operates on any function of the energy E the result is zero.

Substituting from Equation 2, 3, and 4 into Equation 1, an equation for ϕ (\underline{k}) is obtained

$$\phi (\underline{k})/t (\underline{k}) + (\epsilon/\hbar) \underline{F} \cdot \nabla E - (\epsilon H/\hbar^2 c) \Omega_3 \phi (\underline{k}) = 0$$
, (6)

where the product of F and \emptyset is neglected. This is equivalent to assumption IV. The electric field F is not large enough to cause deviations from Ohm's law; i.e., the relationship between F and the current is linear. This assumption is very well satisfied and we shall not modify it.

The solution to Equation 6 can be written as a power series in H

$$\phi (\underline{\mathbf{k}}) = -(\epsilon/\hbar) \sum_{n=1}^{\infty} (\epsilon \mathbf{H}/\hbar^2 c)^n (t\Omega_3)^n (t\underline{\mathbf{F}} \cdot \nabla \mathbf{E}) . \tag{7}$$

The current density J in relation to the field F is

$$\underline{J} = (\epsilon/4\pi^3) \int \underline{v}_k f(\underline{k}) dk$$

$$= - (\epsilon/4\pi^3 \hbar) \int \nabla E g(E) \phi (\underline{k}) dk .$$
 (8)

The integration is over all of k-space. Substituting from Equation 7 we have

$$\underline{J} = (\epsilon^2/4\pi^3\hbar^2 \sum_{n=0}^{\infty} (\epsilon H/\hbar^2c)^n \int g(E) \nabla E(t\Omega_3)^n (t\nabla E \cdot \underline{F}) dk.$$
 (9)

From this equation all expressions pertaining to magnetic effects on the electric current are derived, in particular for the conductivity, the Hall coefficient, the coefficient of the transverse and the longitudinal quadratic magneto-resistance effect. In deriving these expressions Davis introduces restriction 2: E and t are even functions of k_1 , k_2 , and k_3 . He noted in particular that if E and t were spherical surfaces in k-space then the coefficients of magneto-resistance vanished for restriction 3: very low temperatures.

A remarkable result was that under these restrictions the ratio of the transverse and the longitudinal effect was always > 4.08 for cubic materials whose Fermi energy surface was described by the first cubic harmonic following a sphere.

3. Summary of Davis' Assumptions and Restrictions

The following assumptions were made and are retained in the present work.

- I. A relaxation time t (\underline{k}) can be defined.
- II. Interband transitions can be neglected.
- III. The material is isothermal: H and F are homogenous throughout the material.
 - IV. The current and F are proportional (Ohm's law)

The following restrictions were applied at various stages and will be broadened below.

- 1. H lies along k3.
- 2. E and t are even functions of k1, k2, and k3.
- $3. T \sim 0$ (g (E) is a delta function).

- 4. Only terms up to H² were calculated.
- 5. $E(\underline{k})$ and $t(\underline{k})$ have cubic symmetry and can be adequately described by a sphere and the first higher cubic harmonic.

4. Definition of the Present Problem in Terms of Equation 9

The present considerations will deal only with the transverse MR effect. The most general problem is defined thus: the sample is assumed to be a single crystal in the form of a long thin wire. The orientation of crystallographic symmetry elements is not related to the direction of the wire but the magnetic field H is normal to the wire. The direction of the current J is along the wire and no transverse current is present. The coordinate axes can be chosen in two ways

- (a) k1, k2, and k3 are as in Davis' case, i.e., k3 along H, k1 along J, or
- (b) K1, K2, and K3 are according to crystal symmetries.

Both cases have certain advantages in that certain integrals vanish and the transformations of these integrals from one system to the other one will be important.

The general problem will be approached by discussing progressively more general cases, starting with simple conditions. This is both didactically and historically justified. For practical applications these intermediate cases seem to throw considerable light on the problem.

The current density will be measured in units of $\epsilon^2/4\pi^3h^2$ and the magnetic field in units of \hbar^2c/ϵ .

Thus Equation 9 is reduced to

$$J = \sum_{n=0}^{\infty} H^{n} \int g(E) \nabla E(t\Omega_{3})^{n} (t\nabla E \cdot \underline{F}) dk .$$
 (10)

5. General Case A

Here H is along k3 and hence Equation 10 is applicable.

We derive now a general expression for the conductivity as measured in a wire in a transverse magnetic field. The condition that no transverse current flows is

$$J_{1} = \sigma_{11} F_{1} + \sigma_{12} F_{2} + \sigma_{13} F_{3},$$

$$J_{2} = \sigma_{21} F_{1} + \sigma_{22} F_{2} + \sigma_{23} F_{3} = 0,$$
(11)

and

$$J_3 = \sigma_{31} F_1 + \sigma_{32} F_2 + \sigma_{33} F_3 = 0$$

where σ_{ij} are the components of the conductivity tensor with magnetic field present, referred to the k-axes.

According to Equation 10 $\sigma_{i,i}$ can be written as

$$\sigma_{ij} = \sum_{n=0}^{\infty} [n]_{ij} H^n, \qquad (12)$$

where

$$[n]_{ij} = \int g(E) \nabla E_i(t\Omega_3)^n(t\nabla E_j) dk. \qquad (13)$$

By partial integration it is seen that

$$[n]_{i,j} = (-)^n [n]_{j,i} *$$
 (14)

Eliminating F_2 and F_3 and introducing the measured conductivity σ defined by

$$J_1 = \sigma F_1 , \qquad (15)$$

Equation 11 yields

$$\sigma = \sigma_{11} + (\sigma_{22} \sigma_{33} + \sigma_{23}^2)^{-1} \left\{ \sigma_{12} (\sigma_{12} \sigma_{33} + \sigma_{13} \sigma_{23}) + \sigma_{13} (\sigma_{12} \sigma_{23} + \sigma_{13} \sigma_{22}) \right\}. \quad (16)$$

Additional symmetry conditions simplify Equation 16. (See section 7.)

6. General Case B

Here the wire direction cosines are ℓ , m, and n with respect to the coordinates K_1 , K_2 , and K_3 adapted to the crystal symmetry. The equations for vanishing transverse current are

$$\lambda^{-1} \left(\sum_{11} F_{1} + \sum_{12} F_{2} + \sum_{13} F_{3} \right) = m^{-1} \left(\sum_{21} F_{1} + \sum_{22} F_{2} + \sum_{23} F_{3} \right)$$

$$= n^{-1} \left(\sum_{31} F_{1} + \sum_{32} F_{2} + \sum_{33} F_{3} \right) , \qquad (17)$$

where \sum are the components of the conductivity tensor with magnetic field present, and referred to the K-axes.

Let the magnetic field have direction cosines $\lambda,\;\mu_{\nu}$ and ν_{\star} . For a transverse field

$$\lambda \mathcal{L} + \mu m + \nu n = 0. \tag{18}$$

Then the general equation replacing Equation 10 becomes

$$\underline{J} = \sum_{n=0}^{\infty} H^{n} \int g(E) \nabla E(t\{\lambda \Omega_{1} + \mu \Omega_{2} + \nu \Omega_{3}\})^{n} (t \nabla E \cdot \underline{F}) dk.$$
 (19)

Accordingly

$$\sum_{i,j} = \sum_{n=0}^{\infty} H^{n} \sum_{q=0}^{n} \sum_{p=0}^{n-q} [p, q, n-p-q]_{i,j} \lambda^{p} \mu^{q} \nu^{n-p-q}$$
(20)

where

$$[p, q, n-p-q]_{i,j} = \int g(E) \nabla E_i \{ P[(t\Omega_1)^p(t\Omega_2)^q(t\Omega_3)^{n-p-q}] \} (t\nabla E_j) dk, \qquad (21)$$

and P[$(t\Omega_1)^p$ $(t\Omega_2)^q$ $(t\Omega_3)^{n-p-q}$] is the sum of all permutations of p operators $t\Omega_1$, q operators $t\Omega_2$, and n-p-q operators $t\Omega_3$, in different order. This sum consists of n!/p! q! (n-p-q)! terms. It is seen that the integrals in Equation 13 are special cases of integrals of the type in Equation 21, for which p = q = 0.

By partial integration it is seen that

$$[p, q, n-p-q]_{ij} = (-)^n [p, q, n-p-q]_{ij}$$
 (22)

Additional relations among these integrals due to symmetry are given in section 7. The measured electric field F along the wire is given by

$$F = F_1 l + F_2 m + F_3 n$$
 (23)

The measured current density J in the wire is related to F by

$$J = \sum F, \qquad (24)$$

where \sum , the measured conductivity is

$$\sum = \Delta/(mn \Delta'), \qquad (25)$$

$$\sum = \Delta / (mn \Delta'), \qquad (25)$$

$$\Delta = \text{Det} \left| \sum_{i,j} \right|, \qquad (26)$$

We now proceed to consider formulas for special cases of high symmetry.

7. Vanishing MR Effect for Certain Symmetries

Assume, as in Davis' case that E and t are even in k_1 , k_2 and k_3 . This means in most cases that the crystal axes K_1 , K_2 , and K_3 coincide with k_1 , k_2 , and k_3 and that for t (\underline{k}) and E (\underline{k}) there are reflection planes normal to these axes. Remember that H is along k3 and J along k1. According to the principle of time reversal the origin of k-space is always an inversion center. Hence the axes are also twofold rotation axes, and the symmetry of the reciprocal lattice belongs to one of the classes

$$D_{2h}$$
, D_{4h} , T_{h} , O_{h} , or D_{6h}

For D_{2h} and T_h, the k-axes must lie along the crystal axes.

For D_{4h}, k₁ must lie along the fourfold axis and k₂ and k₃ along a pair of twofold axes, or cyclic interchanges of this configuration. There are two pairs of twofold axes at 45° to each other in the plane normal, to the fourfold axis.

For Doh, k₁ must lie along the sixfold axis, k₂ and k₃ along a pair of twofold axes, or cyclic interchanges of this configuration. There are three pairs of twofold axes at 30° to each other in the plane normal to the sixfold axis.

For Oh, k1 must lie along a fourfold axis and k2 and k3 either along other fourfold axes or along twofold axes, or cyclic interchanges of this configuration. There is one pair of twofold axes and one pair of fourfold axes at 45° to each other in the plane normal to each fourfold axis.

The symmetry of the crystal itself may be of any class that yields one of the above five, when augmented by an inversion center, hence, in addition to the above five, we may have

$$D_2$$
, C_{2V} , D_4 , D_{2V} , C_{4V} , D_6 , D_{3h} , C_{6V} , T , T_d , or O ,

i.e., all except triclinic monoclinic trigonal, and C4, S4, C4h, C6h. For these cases in the orientations specified above, the transverse magnetoresistance effect will now be derived.

The integrals in Equation 13, denoted by $[n]_{ij}$ have now the following symmetry properties

$$[\epsilon]_{ij} = 0 \quad i \neq j$$

$$[\omega]_{ij} \neq 0 \quad \text{only if } i + j = 3$$

$$[\omega]_{ij} = 0 \quad \text{for } i + j \neq 3$$
(28)

where ϵ represents any even number and ω any odd number.

According to Equation 12

 $\sigma_{\mbox{\scriptsize i}\,\mbox{\scriptsize i}}$ is an even function of H

$$\sigma_{13} = \sigma_{23} = 0$$

 $\sigma_{12} = -\sigma_{21}$ is an odd function of H

and Equation 16 reduces to

$$\sigma = (\sigma_{11} \ \sigma_{22} + \sigma_{12})/\sigma_{22} . \tag{29}$$

Using Equation 12 after slight rearrangement

$$\sigma = \sigma_0 \left[1 + \sum_{m=1}^{\infty} H^{2m} M_{2m} / \left\{1 + \sum_{m=1}^{\infty} \left(\frac{(0)_{11}}{(0)_{22}}\right)^{1/2} (2m)_{22} H^{2m}\right\}\right], \quad (30)$$

where

$$M_{2m} = \sum_{q=0}^{m-1} \left\{ (2q)_{22} (2m-2q)_{11} + (2q+1)_{12} (2m-2q-1)_{12} \right\}, \qquad (31)$$

$$(n)_{ij} = [n]_{ij}/([0]_{11} [0]_{22})^{1/2},$$
 (32)

and the conductivity with H = 0

$$\sigma_{O} = [O]_{11} \quad (33)$$

Only even powers of H occur in σ . It is clear from Equation 30 that the expression for $\Delta\rho/\rho=(\sigma_0-\sigma)/\sigma_0$ is much simpler than that for $\Delta\rho/\rho_0=(\sigma_0-\sigma)/\sigma$, therefore attention will be confined to the former. Knowing $\Delta\rho/\rho$, $\Delta\rho/\rho_0$ can always be found by the substitution

$$\frac{\Delta \rho}{\rho_{\rm O}} = \frac{\Delta \rho/\rho}{1 - \Delta \rho/\rho} . \tag{34}$$

For resistance changes of a few percent the two quantities are indistinguishable; for larger changes $\Delta\rho/\rho$ approaches 1 as $\Delta\rho/\rho_0$ goes to infinity and both quantities vanish simultaneously. The following theorem can be proved.

Theorem. The necessary and sufficient condition for vanishing $\Delta\rho/\rho$ (for all powers of H) is

$$\Omega_3 (tE_1) = cE_2$$
 (35)

where a subscript i on E means differentiation with respect to \mathbf{k}_{i} and \mathbf{c} is an arbitrary constant.

Proof. First it will be shown Eq 35 is necessary and sufficient for $M_2 = 0$. Second it will be proved if Eq35 holds, $M_{2m} = 0$ for all m. Since $\Delta \rho/\rho = 0$ (for all H) implies $M_2 = 0$ the theorem is then proved.

First Part. Consider

$$M_2 = (0)_{22} (2)_{11} + (1)_{12} = 0.$$
 (36)

If the explicit form of these symbols is substituted in Equations 32 and 13, this is a Schwartz equality, since the integrand of the last term squared equals minus the product of the integrands in the first term. It is well-known that the necessary and sufficient condition for this to vanish is that the integrands of $[0]_{22}$ and $[2]_{11}$ be linearly dependent. This condition is just Equation 35, which establishes the first part of the proof.

Second Part. If Equation 35 holds it is a simple matter to show that

$$(2k + 2)_{11} = -c^2 (2k)_{22}, (37)$$

and

$$(2k+1)_{12} = -c (2k_{,22} *$$
 (38)

Substituting this into Equation 31 it can be seen that the terms between {} yanish for all values of q and m, thus the second part is proved and so is the theorem.

This theorem has a number of interesting consequences:

- (a) If Equation 35 is satisfied there is no resistance change for all H at any temperature.
- (b) If E is of the form $ak_1^2 + bk_2^2 + ck_3^2$ and t is constant Equation 35 is satisfied; thus the MR effect vanishes to all orders. The term ck_3^2 may, in fact, be any even

function of k_3 . The quadratic form is of interest, since it is mentioned in various places in the literature.

(c) An interesting paradox was encountered when it was found that for certain functions $E\left(\underline{k}\right)$ and $t\left(\underline{k}\right)$ the MR effect would vanish, although Equation 35 was not satisfied. This seems to contradict the theorem. The explanation of this paradox is as follows:

The conditions under which the theorem is proved lead to Equation 35 as the necessary and sufficient condition that the MR effect vanish for all temperatures. However, if Equation 35 is not satisfied, the MR effect must not vanish for any temperature $T \neq 0$. It may happen that the discrepancy between the right and left sides of Equation 35 becomes smaller and smaller as $T \rightarrow 0$ and in the limit as T = 0, Equation 35 is satisfied. This seems to be a rather academic case at first, but it is of practical importance, since the temperature is always very low compared to the degeneracy temperature of the electron gas and hence is close to absolute zero. Consequently g (E) is usually taken as a delta function, for example by Davis and by Wilson. Thus, the usual calculation does not reveal the difference between an MR effect that vanishes only for T = 0 and one that vanishes for all T. The conditions given in Equation 35 refers to the cases which vanish for all T, while an extension of the theorem is required for cases that vanish only for T = 0. It is easy to show, along lines similar to those used to prove Equation 35, that the necessary and sufficient condition for vanishing MR effect at T = 0 is

$$\left[\Omega_{3} \left(t E_{1} \right) = c E_{2} \right]_{E=E_{0}}$$
(39)

Here the integrands of Equation 35 are considered in terms of the independent variables E, θ , ϕ , and E_O is the Fermi energy. Relation 39 is then valid for all θ and ϕ . The explicit expressions for E₁ and E₂ in terms of the new variables have not been substituted. If, in addition, t is a function of the energy only, Equation 39 reduces further to

$$\left[\Omega_{3} \ \mathbb{E}_{1} = c\mathbb{E}_{2}\right]_{\mathbb{E}=\mathbb{E}_{0}} \tag{40}$$

Under the conditions specified, this is necessary and sufficient for vanishing MR effect at absolute zero, whereas

at higher temperatures the effect becomes, in general, proportional to T^2 , as may be seen by the familiar expansion of Fermi integrals such as Equation 13. It is interesting to note that behavior of this sort has never been observed; on the contrary, the observed effect usually decreases with rising temperature. Apparently Equation 39 or 40 is not satisfied in all cases studied experimentally so far. Further work on estimating what conditions make this T^2 effect large may be done later.

- (d) In particular, if E is as described in (b) and t = t (E), Equation 40 is satisfied and the low—T MR effect must vanish. Likewise, the two-band model suggested by Wilson for Bi, in the simple form in which it was suggested, leads to zero MR effect.
- (e) While the conditions of Equations 36, 39, and 40 are not of great help in finding materials that have an exceptionally large MR effect, they do serve to discard certain materials a priori. In particular the very low values for the MR effect in good conductors such as Cu, Al, etc., are understood. In these materials the top band is half filled and the free-electron approximation is reasonably good, with the energy surfaces probably not too far from spherical.
- (f) In this connection it is also interesting that Justi's observations on Au (cubic) at low T show very pronounced minima of $\Delta\rho/\rho$ for those angles (n . 45°) which satisfy the symmetry conditions stated at the beginning of the present analysis.
- (g) It may be possible in the future to extend the approach of this section to cases where the difference between Ω_3 (tE₁) and cE₂ is small.
- (h) It is, of course, possible to write Equation 30 as a single power series in ${\rm H}^2$. The coefficients are lengthy expressions in terms of the M and parentheses quantities and will not be given here.
- (i) If the crystal is cubic with its fourfold axes along the current and the magnetic field, we have in addition to the above symmetry conditions

$$(0)_{ij} = (0)_{11}$$
 (41)

8. Some General Expressions for B in the k System

The coefficient B of the quadratic term can be expressed also in terms of the integrals (n); in more general cases than those of Davis by means of Equation 16. The derivation is tedious but straightforward. Using a system of k1, k2, and k3 axes as before with H along k3 and J along k1, we

$$B = \frac{(0)_{22} (2)_{33} + (2)_{22} (0)_{33} - 2 (0)_{23} (2)_{23} + (1)_{23}^{2}}{(0)_{22} (0)_{33} - (0)_{23}^{2}}$$

$$- \frac{1}{|0|} \{ (0)_{11} [(1)_{23}^{2} + 2 (0)_{23} (2)_{23} + (0)_{22} (2)_{33} + (0)_{33} (2)_{22}]$$

$$(42)$$

+
$$(0)_{12}[-2(1)_{13} (1)_{23}-2(0)_{33} (2)_{12} + 2(0)_{23} (2)_{13}-(0)_{12} (2)_{33}-2(0)_{13} (2)_{23}]$$

+ $(0)_{13}[2(1)_{12} (1)_{23} + 2(2)_{12} (0)_{23}-2(2)_{13} (0)_{22}-(0)_{13} (2)_{22}]$
+ $(0)_{23}[-2(1)_{12} (1)_{13}-(0)_{23} (2)_{11}]$
+ $(0)_{22}[(1)_{13}^2 + (2)_{11} (0)_{33}]$
+ $(0)_{33} (1)_{12}^2$
where
$$|0| = \det(0)_{1,1}^{2} *$$
(45)

If k_2 occurs even in E and t, $(\epsilon)_{12} = (\epsilon)_{23} = (\omega)_{13} = 0$ and Equation 42 reduces to

$$\frac{2}{B = (0)_{13}[(0)_{22}(2)_{33} + (1)_{23}^{2}] - 2(0)_{13}(0)_{33}[(0)_{22}(2)_{13} - (1)_{12}(1)_{23}] + (0)_{33}[(0)_{22}(2)_{11} + (1)_{12}^{2}]}{(0)_{22}(0)_{33}[(0)_{23}(0)_{13} - (0)_{11}(0)_{33}]}$$
(44)

If, instead, k_3 occurs even in E and t, then $(n)_{13} = (n)_{23} = 0$, leading to

$$B = \frac{(0)_{22}^{2} (2)_{11} + (0)_{22} [(1)_{12} - 2 (0)_{12} (2)_{12}] + (2)_{22} (0)_{12}}{(0)_{22} [(0)_{12} - (0)_{11} (0)_{22}]}$$
(45)

It is seen from these results that a reflection plane normal to k3 is more effective in simplifying than one normal to k_{2*} If both reflection planes are present both Equations 44 and 45 reduce to Davis' expression, which in the notation used here reads

$$B = -(2)_{11} (0)_{22} - (1)_{12}^{2}$$
 (46)

(43)

The effects of a number of other symmetries on Equation 42 remains to be investigated. In the general case Equation 42, B is determined by 15 constants. In Equation 44 there are 9 constants, in Equation 45, 6 or 7 constants (6 if it is taken into account that $(0)_{11}$ $(0)_{22}$ = 1) and in Equation 46, 3 constants. The expressions for B are the same, whether $\Delta\rho/\rho$ or $\Delta\rho/\rho_0$ is evaluated. The purpose of deriving these general expressions is to transform them later to a k-system of certain symmetry which does not coincide with the k system. The resulting expressions for B will have to match corresponding expressions derived along another approach. But besides providing a check for later work, they have some value as new relations.

9. A Remark about the Dependence on the Azimuth

For cubic materials Justi and coworkers have found a strong dependence of $\Delta\rho/\rho$ on the angle \emptyset between H and one fourfold cube axis, while the current flows along another fourfold cube axis. H remains always normal to the current. From the general tensor character of the coefficients in the series

$$\Delta \rho / \rho = BH^2 + CH^4 + DH^6 . . . \qquad (47)$$

it follows that these coefficients must generally be of the form

$$B = B_1 + B_2 \sin 2 (\phi - \phi_b)$$
 (48)

$$C = C_1 + C_2 \sin 2 (\phi - \phi_c') + C_3 \sin 4 (\phi - \phi_c''), \qquad (49)$$

$$D = D_1 + D_2 \sin 2 (\phi - \phi_d) + D_3 \sin 4 (\phi - \phi_d') + D_4 \sin 6 (\phi - \phi_d''),$$
..., etc. (50)

For cubic symmetry with the current flowing along a fourfold axis, only the terms with $\sin 4n \ \phi$ can differ from zero. Thus, B can only be a constant independent of ϕ ; C can consist of a constant and a term in 4ϕ , as can D; E contributes 0, 4, and 8ϕ ; etc. Thus, the complex patterns observed by Justi require the study of high coefficients.

The fact that $\Delta\rho/\rho_0$ is approximately a straight line for high values of H does not imply that these higher coefficients vanish. The straight line does not pass through the origin, so the coefficients, far from being negligible, will converge more or less rapidly to zero, with alternating sign. In order to describe the main features of the observations on Au, it is necessary to go to coefficients of H^{24} , and at present this does not look worthwhile. However, this phenomenon is noted here because it points to what we believe is a correct interpretation of the rule stated by these authors, that planes with low indices

show a strong reduction in $\Delta\rho/\rho$ compared to planes with high indices. We would reformulate this as follows: if a Fourier analysis of $\Delta\rho/\rho$ as a function of \emptyset were made, the contribution of the component with $4n\emptyset$ is primarily due to the coefficients of H^{4n} and H^{4n+2} in Equation 47. It is a general characteristic of Fourier series of this sort that they lead, under the conditions of cubic symmetry, necessarily to rather sharp peaks at places with low indices. Thus, it seems that in the future the problem of the azimuth dependence and that of the behavior for high H will turn out to be two aspects of the same formula.

10. Some Symmetry Relations among the [0, q, n-q]

The measured conductivity \sum is expressible in terms of the quantities $[p, q, n-p-q]_{ij}$ according to Equation 20, 21, 25, 26, and 27. For the case that q=0 omit the first term in the bracket and write $[q, n-q]_{ij}$, instead of $[0, q, n-q]_{ij}$. This case is of special interest and a number of relations have been established among these quantities for certain symmetries. An exhaustive treatment of this topic must, however, be deferred. The symmetries refer to the K system regarding E (K) and t (K).

If a mirror plane normal to K1 is present

$$[q, \omega - q]_{ij} = 0, \qquad (51)$$

$$[q, \epsilon - q]_{12} = 0,$$
 (52)

$$[q, \epsilon - q]_{13} = 0, \qquad (53)$$

and

$$[q, \omega - q]_{23} = 0.$$
 (54)

The same is true if K_1 is a twofold axis. If K_1 is a fourfold axis, in addition

$$[q, \epsilon - q]_{23} = (-)^{q} [\epsilon - q, q]_{23}, \qquad (55)$$

if $2q = \epsilon$ and q = odd this is = 0.

$$[q, \omega - q]_{12} = (-)^{q+1} [\omega - q, q]_{13},$$
 (56)

$$[q, \epsilon - q]_{22} = (-)^q [\epsilon - q, q]_{33}, \qquad (57)$$

and

$$[q, \epsilon - q]_{11} = (-)^{\mathbf{q}} [\epsilon - q, q]_{11}, \qquad (58)$$

if $2q = \epsilon$ and q = odd this is = 0. If a mirror plane is normal to K_2 , in addition to the above,

$$[\omega, \epsilon - \omega]_{ii} = 0, \qquad (59)$$

$$[\omega, \omega' - \omega]_{12} = 0, \qquad (60)$$

$$[\epsilon, \omega - \epsilon]_{13} = 0, \tag{61}$$

and

$$[\epsilon, \epsilon' - \epsilon]_{23} = 0 . \tag{62}$$

If in addition to the above K3 is a fourfold axis

$$[0, \epsilon]_{22} = [\epsilon, 0]_{11} = [0, \epsilon]_{11}, \tag{63}$$

and

$$[0, \epsilon]_{23} = [\epsilon, 0]_{12} = 0 *$$
 (64)

11. Some Expressions for the Azimuth Dependence in a Cubic Case

Consider the case of a cubic crystal with current along $K_1=k_1$. The magnetic field (along $k_3)$ makes an angle \emptyset with K_3 and $90^2\emptyset$ with K_{2*} . The angle \emptyset is referred to as the azimuth, and Justi and coworkers have measured $\Delta\rho/\rho_0$ as a function of \emptyset for just such conditions.

On the one hand, the measured conductivity can be expressed in terms of the quantities $[n]_{ij}$ through Equations 16 and 12, and these can be transformed into $[q, n-q]_{ij}$. In this way the azimuth dependence can be found.

Another way to find the azimuth dependence is by using Equations 25, 20, and 22. This has not yet been done, but will be carried out as a check in the future. The results of the transformations obtained so far are listed in Table I in which S denotes $\sin 4\phi$ and c denotes $\cos 4\phi$. So far, with the help of this transformation table it is possible to express the coefficient B of H² and C of H⁴ in terms of the azimuth ϕ by means of the three quantities $[0,0]_{11}$, $[0,2]_{11}$, and $[0,2]_{33}$ for B, and the eight quantities $[0,0]_{11}$, $[0,2]_{11}$, $[0,2]_{33}$, $[0,3]_{12}$, $[1,2]_{13}$, $[0,4]_{11}$, $[2,2]_{22}$ and $[0,4]_{33}$ for C. For angles that are integral multiples of 90° the quantities in Table I reduce to the untransformed ones $[0,n]_{1j}$ since S = 0 and c = 1.

The quantity B, evaluated according to Equation 42 must be independent of \emptyset and this checks. The value of the constant is then $\{-[0,0]_{11} [0,2]_{11} + [0,1]_{12}\}/[0,0]_{11}$, which is equivalent to Equation 46. The quantity C can best be expressed for 0 and 45°, which determines its dependence on $4\emptyset$. The expressions have been obtained but need rechecking and will be reserved for a future report.

In this way it is possible to express all higher coefficients for cubic crystals as a function of the azimuth in terms of the basic integrals $[q, n-q]_{ij}$. These integrals can, of course, be expressed explicitly only in

TABLE I

AZIMUTH TRANSFORMATION FORMULAS FOR $\begin{bmatrix} n \end{bmatrix}_{1,j}$ FOR THE CUBIC CASE

23	0	0	$\frac{S}{4} \left\{ [0,2]_{11} - [0,2]_{33} \right\}$	0	\$\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\		
13	0	O .	0	10.31-41.01.2	0		
12	0	$[0,1]_{12}$	o *	$[0,5]_{1,2}(\frac{2}{4}+\frac{c}{4})+\frac{c}{8}$	$+ [1,2]_{13} (\frac{c}{h} - \frac{1}{h})^{-h}$ + 0		
55	[0,0]	0	[0,2]33	0	+ \frac{1}{8} [0,4]33(5+3c)+	$+\frac{1}{8}[0,t]_{11}(1-c)+$	$+\frac{1}{8}[2/2]_{22}(1-c)$
22	11[040]	0	[0,2]11	0	$[0, t]_{11}(\frac{2}{t} + \frac{c}{t}) +$	$+\frac{1}{8} [2_y 2]_{22} (1-c)$	
11	[0,0]	Ö	[0,2]	0	$[0_{7}^{4}]_{11}(\frac{2}{4}+\frac{c}{4})+$	$+\frac{1}{9}[0,2]_{11}(1-c)$	
i.	о п	п = 1	n 2	n = 3	т Т	+ + (m)	

terms of the functions E (K) and t (K), which is again a subject for further study. It is hoped that this study may then lead to insight into the reverse problem, which is also theoretically important: can the shape of energy surfaces be determined in practice from magneto-resistance data? We are aware of Shockley's remarks in this connection. T

C* EXPERIMENTAL PART

1. Equipment

The magnet used in this work was a simple electromagnet with a gap of about 5 cm and a cross section of about 5 x 5 cm fed by automobile batteries. The calibration of H in the gap has caused some difficulties and the figure given in the previous quarterly report of 534 Gauss is certainly not quite correct. It was determined by means of a commercial Gauss meter, which was later found to be in error by some 15 percent. The field was actually about 420 Gauss. Our field has been calibrated against those of three permanent beta spectrometer magnets of 795, 330, and 107 Gauss. For many measurements at the present stage the calibration is not now unsatisfactory.

The relation between magnet current and field has been determined and is practically linear above 100 Gauss but not below. The accuracy of absolute values for H is estimated to about 5 percent above 100 Gauss; the relative accuracy is somewhat better. In the course of time it may be necessary to change to an air-coil magnet, but we feel a great deal of information can still be obtained with the present setup, which has the advantage of simplicity, provided more calibration checks are made.

An improved sample holder was constructed to insure as much as possible, isothermal conditions and to permit rotation of the wire in the field about the wire as axis. The holder was sturdier than its predecessor and had a finer scale, permitting more accurate reading of the angle $(1-2^{\circ})$.

In order to study the orientation of the cleavage plane in wires which seem to be single crystals, a reflection microscope is in use. A holder was designed and made by the machine shop which permits the wire to be turned by measurable amounts about two perpendicular axes (accuracy $\sim 2^{\circ}$) while under the microscope, in order to obtain reflection from the cleavage plane at the end of the wire.

A small zone-melting apparatus was built for zone-melting Bi-wires in their glass sheaths. The wire is drawn through an electrically heated Pt-loop by a slowly and smoothly moving carriage at the rate of 6 cm/hour. An

additional permanent magnet, providing a transverse field of 6000 Gauss can be placed across the melted zone, but has not produced any noticeable difference up to the present.

2. Materials

Most of the experiments were done with bismuth from two sources. The cheap supply of granular Bi was from Baker and Adamson, reagent quality, 99.8% pure. The other material was Bi, 99.9992% pure (ductile). Mr. Fitzpatrick, from Muskegon, Michigan came to Ann Arbor in connection with our request for information about the ductile, very pure Bi-wire which he manufactures. He was kind enough to send us four complimentary samples of ductile Bi-wire and also offered to make bismuth alloys for us provided we furnish the alloy materials other than bismuth. Since such alloys would be relatively expensive, and would have to be made in relatively large numbers with varying content of alloying materials and varying parameters of the manufacturing process, we judged that it would be more expedient to study in detail the differences in MR effect of the 99.8% pure wires and the ductile 99.9992% pure wires first.

In addition to the two types of Bi-wires described above and some wires made of Bi with 0.4% Pb, a number of other materials were occasionally The motivation for other choices was twofold. In the first place, an inspection of any list of superconducting materials shows that three elements are particularly effective in enhancing superconductivity in other materials that have only very low transition temperatures or no superconductivity at all. These three are Bi, Nb, and N. A compound of Nb and N yields the highest transition temperature known. Since Bi has both this enhancing property for superconductivity and strong magneto-resistance effects, it seemed not altogether impossible that there might exist a relationship between the two effects, We have not as yet been able to obtain Nb wire, but we have ordered powder in order to try to make the wire here, and will continue our efforts to obtain the wire from external sources. In the second place, we have given thought to the technical requirements (PR and C-54-ELS/D-3420, 30 December 1953) especially sections C-2e and B-4. Since a major part of the temperature dependence of $\Delta \rho / \rho$ is due to the temperature dependence of ρ , it is clear that materials with a zero or slightly negative temperature coefficient of ρ are desirable. A few such materials have been measured, constantan, manganin, carbon resistors and wires made of Wood's metal.

3. Measurements

While the ductile wires furnished by Mr. Fitzpatrick were said to be single crystals, x-rays showed that this was not so. Figure 2 is a picture obtained through the courtesy of Dr. S. Krimm of this laboratory, of a small

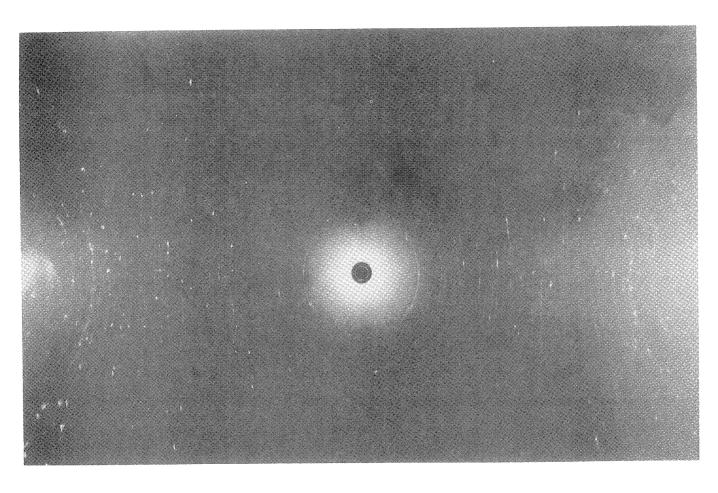


Fig. 2. X-ray photograph of Fitzpatrick ductile Bi wire showing polycrystalline structure

section of the wire and shows some spots with circles of comparable intensity. Thus the sample is far from a single crystal. Since we are not familiar with the details of the manufacture of this wire, we cannot add any further interpretation. We plan to make similar observations with other samples.

The Bi wires drawn in this laboratory showed, in general, a strong anisotropy for rotation about the wire axis through an azimuth angle ϕ_* Theoretically (See sec. B9) this dependence can be represented for weak fields by

$$\Delta \rho/\rho_0 = H^2 (B_1 + B_2 \sin 2\emptyset), \qquad (65)$$

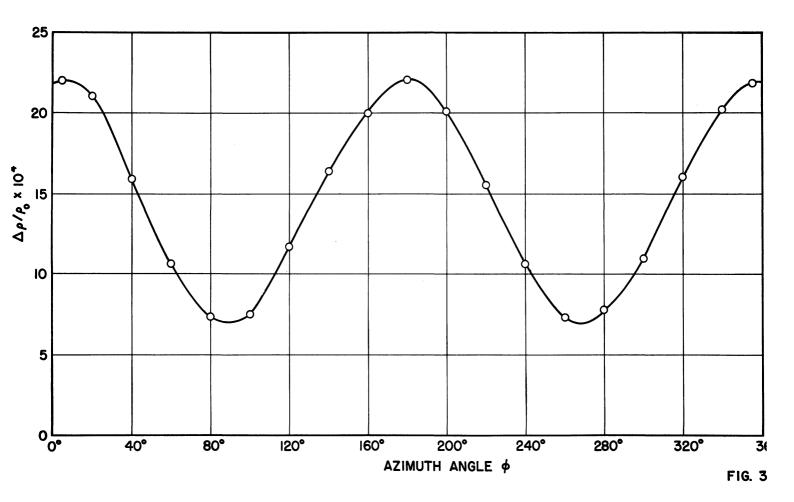
and this fitted the experiments very well; thus, it seems desirable to measure the constants B_1 and B_2 for various samples. They both will depend on the angle between the trigonal Bi axis and the wire. In order to get down to really basic material constants, of which there are about half a dozen in the case of the Bi-symmetry (always ignoring terms of higher powers than H^2), more independent measurements have to be made, but the theory associated with this problem remains to be analyzed later.

The experimental azimuth dependence is illustrated for a representative sample in Fig. 3, showing $\Delta\rho/\rho_O$ versus ϕ , and in Fig. 4 showing $\Delta\rho/\rho_O$ versus sin 2 ($\phi = \phi_O$) which should be a straight line for a suitable choice of ϕ_O .

Table II gives data for a number of Fitzpatrick wires. The absolute accuracy is \pm 5% in H, usually about 1-2% in $\Delta\rho/\rho_0$ and in B₁ and B₂ consequently about 10%. The relative error in B₁ and B₂ for the same sample and for comparison between different samples is considerably less and is estimated to be about 2%.

TABLE II

No.	Bi Sample	H Gauss	$\Delta \rho / \rho_{O} x$ max	104 min	B ₁ x10 ⁹	B ₂ x10 ⁹	r Ohms
1	Fitzpatrick wire 99.9992% pure 19.4Ω/ft	428	16.3	0	8.9	0	1.8
2	Same, melted and drawn	433	22*0	7.22	7.8	3 ∗9	66
3	Another sample same treatment as 2	406	19.5	16	10.8	1.0	24
4	Identical	403	18.9	15*4	10.5	0.75	8
5	Identical	397	18.1	11.5	9.4	4.1	22
6	Identical	282	8.32	6.19	9.1	1.2	45
7	Another sample melted drawn, zone-melted 4x	400	18.1	12.7	9.6	1.7	6.5



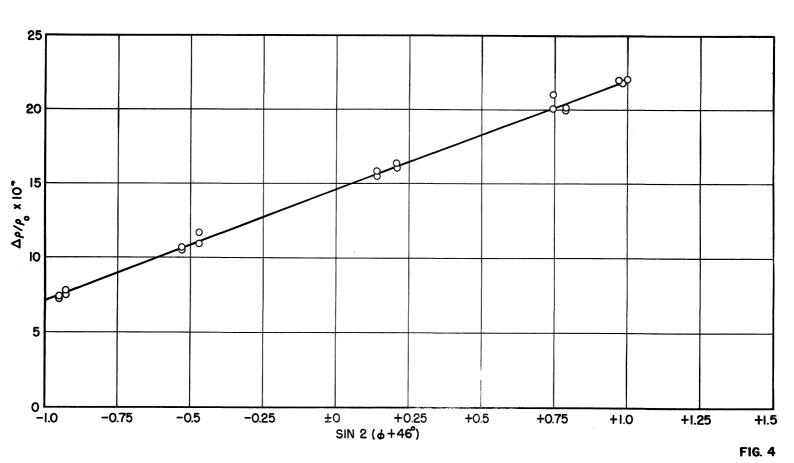


Table III shows deviations from the $\ensuremath{\mathrm{H}^2}$ relation for the Fitzpatrick wires.

TABLE III

No.	Bi Sample	H Gauss	$\Delta \rho / \rho_{O} x 10^{4}$ max	(B ₁ +B ₂)x10 ⁹
1	Same as No. 1	428	16.3	8.9
	Table II	261	7.6	11.2
		119	2.5	17.6
2	Same as No. 2 Table II	433	22.0	11.47
	lable II	264	9*6	13.9
		125	2*9	18.3
3	Same as No. 3	406	19,5	11.8
	Table II	270	8.7	11.8
		135	2,2	12.1

Table IV gives data for a number of wires made from Baker and Adamson 99*8% pure Bi.

TABLE IV

No.	Bi Sample and Treatment	H Gauss		o ^{x10⁴} min	B ₁ xl0 ⁹	B ₂ x10 ⁹	r Ohms
1	Melted and drawn	386	20.2	11.4	9∗5	2.7	20
2	Another sample same as No. 1	397	22.7	8.6	10.6	4.5	12.7
3	Another sample same as No. 1	406	23.1	22.1	13*7	0,.3	26.2
4	Melted, drawn, zone melted once	415	21.8	14.6	10.6	2.1	7*4

Table V shows deviations from the ${\rm H}^2$ relation for various wires made from Baker and Adamson Bi.

TABLE V

No.	Bi Sample	H Gauss	Δρ/ρ _Ο χ10 ⁴ max	(B ₁ +B ₂)x10 ⁹
1	Same as No. 1	386	20.2	12.2
	Table IV	367	18.3	13.6
2	Same as No. 4 Table IV	415	21.8	12.7
'l'a	Table IA	277	10.4	13.6
		140	2.7	14.0
	Alloyed with 0.4% Pb, melted and drawn	432	7.4	4.0
	merced and drawn	287	3∗5	4*3
		138	0.83	4*3
14	Another sample Same as No. 3 Table V	432	13.0	7.0
	pomic an ida) table A	287	6.0	7.4
		127	1.4	8.7

Table VI compares data for wires that were solidified in or outside a magnetic field of 6000 Gauss normal to the wire during zone melting. All wires were 99.8% pure Bi.

TABLE VI

No.	Bi Sample	H Gauss	$\Delta \rho / \rho_{\rm C}$ max	x10 ⁴ min	B ₁ x10	B ₂ x10 ⁹	r Ohms
1	Same as No. 3 Table IV Untreated	406	23.1	22.1	13.7	0.3	26.2
2	Part of same wire as sample 1, zone melted in H=6000 Gauss	392	20.9	5∗3	8.5	5.1	23.6
3	Another wire zone melted in H=6000 Gauss	397	18.8	6.4	8.0	4.0	21.8
4	Another wire zone melted in H=6000 Gauss	394	21.1	17.3	12.4	1,2	11.5

Measurements on wires of manganin, constantan, and carbon resistors yielded no detectable $\Delta\rho/\rho_O$, which means that $\Delta\rho/\rho_O<3.10^{-5}$, or no more than about 1-2% of the corresponding values for Bi.

4. Discussion and Conclusions

The data presented in Tables II to VI suggest the following conclusions:

Table II. The original ductile polycrystalline wire seemed to have no azimuth dependence (B_2 = 0); this may be connected with the random orientation of the grains (Fig. 2). Apart from some scattering from one sample to another, it seems that melting and drawing this material in the usual manner in glass tubing does not materially affect the magnitude of the MR effect; if anything, there is a slight improvement. The wires obtained in this way are single crystals, as was occasionally checked by X-rays, and have reverted to the brittle state. They exhibit an azimuth dependence varying in strength from sample to sample. Also, additional zone melting does not affect the results. It seems at present that there is little connection, if any, between the ductility or brittleness of the wire and its MR behavior at room temperature.

Table III. The wires from Fitzpatrick show, in general, a more than quadratic increase of $\Delta\rho/\rho_0$ with H when untreated. When melted and drawn this effect sometimes disappears. Further work along this line is necessary to establish this conclusion definitely and to trace its origin. It may be due to the presence of a small negative term, linear in H, as found by Donovan and Conn⁹. It seems strange, though, that such a large linear effect should be found for polycrystalline material.

Table IV. The Baker and Adamson wire seems to have at least as good or somewhat better MR properties than the Fitzpatrick wire. It shows, like all wires that are made by melting and drawing in a glass tube, strong azimuth anisotropy which seems to scatter more or less randomly. Either this material is actually much purer than the tolerance given (99.8%) or the impurities are of a nature affecting the MR effect very little, or the effect is rather insensitive to impurities in this range of concentrations. This must be investigated further.

Table V_* The Baker and Adamson wires show a much smaller deviation from the quadratic H relation than the Fitzpatrick wires of Table III although there is a systematic trend. This trend is also present if 0.4% Pb is added, but the H-calibration will have to be checked once more to make sure that this trend is real. If it is real, it may again be due to a linear term and should probably be absent for samples with very low B_2 . This remains a subject for further study and clarification.

Table $\overline{\text{VI}}$. The data show that the MR effect is not enhanced by zone melting in a transverse magnetic field of 6000 Gauss. If anything, it is slightly reduced.

General Remarks. The preparation, mounting, and measuring of a sample is at present still rather time-consuming and may possibly be somewhat accelerated.

D. PROGRAM FOR NEXT INTERVAL

Theoretical

Further development of the theory will follow present lines.

Experimental

We feel that there are still a number of factors in the study of Bi to be cleared up: for example, the relation of the axial anisotropy with the orientation of crystal axes relative to the wire, the question of the existence of a linear effect and a more accurate study of the influence of impurities starting from the very pure Bi.

We also plan to work on a number of alloys suggested by their superconductivity; this depends on their availability or the problems encountered in making and shaping them.

E. IDENTIFICATION OF PERSONNEL

In addition to the persons identified in the previous report, Mr. H. Patterson has worked on the experimental aspects of the project since November 1, 1953 and Mr. L. P. Kao on the theoretical aspect since December 1, 1953. Both are experienced graduate students in Physics above the M. S. level. Altogether the following man-hours were devoted to the work during the period covered by the present report.

	Hrs
W. Tantraporn	245
H. Patterson	74
L. P. Kao	352
E. Katz	94
Total	765

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