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MAGNETICALLY SENSITIVE

ELECTRICAL RESISTOR MATERIAL

1 February 1954 to 31 May 1954

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#### ABSTRACT

#### Theory

It is proved that the magnetoresistance is an even function of the magnetic field  ${\rm H}_{\star}$ 

Some features are discussed regarding the dependence of the magnetoresistance on:

- (1) the orientation of the wire with respect to the crystal axes,
- (2) the orientation of the magnetic field with respect to the crystal axes, and
- (3) the crystal symmetry, as it affects the bracket symbols of which the magnetoresistance is composed.

Some progress was made towards determining how many and which measurements are required to furnish fundamental material constants.

#### Experiment

Bismuth electrode contacts were greatly improved by using gallium contact wells.

A linear term in the measured dependence of the magnetoresistance was shown to be due to insufficient maintaining of isothermal conditions. Improved conditions practically eliminate the occurrence of this term and give experimental support to the evenness theorem proved theoretically.

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#### A. INTRODUCTION

The present report is a direct continuation of the previous quarterly report (No. 2) and is again divided into a theoretical and an experimental section.

In the theoretical part, formulas are developed for the magnetoresistance and its dependence on crystal anisotrophy. So far as we know, these
formulas are new and apply to the most general cases of crystal anisotrophy,
of orientation of the current, and of the magnetic field. Probably many
special cases of interest are present in latent form in these general formulas
and will have to be analyzed further in the future. One special detail which
has been worked out, namely the proof that the magnetoresistance effect is an
even function of the magnetic field for all crystal symmetries, has found
interesting confirmation by new experiments (see below) which demonstrate the
spurious nature of odd terms found by other investigators for bismuth.

In the experimental part, measurements are reported on Bi, and improvements in the technique of measurement and in the interpretation of data Formulas will henceforth be numbered with a first number referring to the report, a second number referring to the section, and a third number for designation within each section consecutively.

#### B. THEORETICAL PART

#### 1. Purpose

The purpose of the theory, the development of which was started in the previous report and is continued here, is to develop formulas expressing the magnetoresistance in terms of basic material constants, for all 32 crystallographic groups, for arbitrary orientation of the electric current and the magnetic field. This purpose has essentially been realized in the period covered by this report and all that remains to be done (although this may well turn out to be a major job) is the application of the general formulas obtained to special cases of interest, and the derivation of some of their less obvious consequences.

#### 2. Coordinate System and Notation

Let us take an orthogonal set of coordinates  $(k_1, k_2, k_3)$  in reciprocal space, adapted to the symmetry of the reciprocal lattice. The last phrase means that  $k_1$ ,  $k_2$ ,  $k_3$  are so oriented that they coincide with a maximum number of proper or improper rotation axes. In systems of high symmetry, this requirement essentially defines the orientation of the coordinate axes, whereas for systems of lower symmetry the coordinate axes may be partially or entirely arbitrary. To be more specific, we observe that the reciprocal lattice always possesses a center of symmetry, essentially due to the principle of time reversal, and hence belongs to one of the following eleven groups:

S2, C2h, S6, C4h, D2h, C6h, D3i, D4h, D6h, Th, Oh.

In the first case the coordinate axes are completely arbitrary. In the cases  $C_{2h}$ ,  $S_6$ ,  $C_{4h}$ ,  $C_{6h}$  one coordinate axis coincides with the symmetry axis, the other two coordinate axes being free in the plane normal to the first one. In all other cases at least two coordinate axes coincide with symmetry axes, leaving no freedom for the direction of the third axis. In particular, for bismuth, whose reciprocal lattice belongs to  $D_{3i}$ ,  $k_1$  shall be along the trigonal axis,  $k_2$  along a binary axis, and  $k_3$  accordingly (or permutations of these orientations).

This coordinate system is chosen in order to use the symmetry properties of the energy surface and the relaxation time in reciprocal space to best advantage.

The direction of the current is specified by the three direction cosines  $\ell_1$ ,  $\ell_2$ ,  $\ell_3$  with respect to the coordinate axes. For long thin samples the direction of the current is along the wire. This will always be assumed.

The direction of the magnetic field H is specified by the three direction cosines  $\gamma_1$ ,  $\gamma_2$ ,  $\gamma_3$  with respect to the coordinate axes. We discuss the general case in which the magnetic field makes an arbitrary angle with the current.

The present choice of coordinates and notation is related to that used in the previous report as shown in Table I.

TABLE I

COMPARISON OF NOTATION IN PREVIOUS AND PRESENT REPORT

	Previous	Present
Coordinates	K <sub>1</sub> , K <sub>2</sub> , K <sub>3</sub> (case b)	$k_1, k_2, k_3$
Direction cosines of current density J	l,m,n	$\ell_1,\ell_2,\ell_3$
Direction cosines of the magnetic field	λ,μ,ν	$\gamma_1, \gamma_2, \gamma_3$
Components of the con- ductivity tensor	$\sum_{ ext{ij}}$	$\sigma_{\mathtt{i}\mathtt{j}}(\underline{\mathtt{H}})$
Bracket quantities of equation 21	[p,q,n-p-q] <sub>ij</sub>	[p',p,n-p-p'] <sub>ij</sub>
Measured conductivity.	Measured conductivity $\sum$	Magneto conductance $\sigma(\underline{H})$

# 3. The General Formulas for the Magneto Conductance

In the previous report, section 6, we introduced the "measured conductivity," defined as the ratio of the electric field along the wire to the current along the wire, with the auxilliary condition that the transverse current be zero. Since this quantity depends on the magnetic field we shall call it the magneto conductance  $\sigma(\underline{H})$ .

Equations (25), (26), (27) of the previous report express the magneto conductance in terms of the components of the conductivity tensor  $\sigma_{ij}(H)$  and the direction cosines  $\ell_1$ ,  $\ell_2$ ,  $\ell_3$  of the wire, and the  $\sigma_{ij}$  are in turn expressed by means of equations (20) and (21) in terms of the basic functions

 $E(\underline{k})$  and t(k), which are characteristic for any material, and the direction cosines of the magnetic field.

Thus, the problem of expressing the magneto conductance explicitly in terms of basic properties of the material has been solved. However, we realized later that some of the expressions can be greatly simplified by making use of tensor concepts from the start. Moreover, the dependence of the magneto conductance on crystal symmetry, on the orientation of the wire, and on the orientation of the magnetic field, while implied in these formulas, requires further elaboration in order to set in evidence all the particulars that can arise from special symmetry conditions.

We proceed to give now the general formulas in their simplest form. The measured component of the electric field along the wire shall be denoted by E\*. The components of the total electric field E with respect to the adapted cartesian coordinate system are  $E_i$  (i=1,2,3). The direction cosines of the wire are  $\ell_i$ . Then:

$$E^* = E_i \ell_i, \qquad (3.3.1)$$

where summation over the repeating index i is implied.

The resistivity tensor  $\rho_{i,j}$  is defined by

$$E_{i} = \rho_{ij} J_{j}. \tag{3.3.2}$$

The condition that no transverse current flows is

$$J_{j} = J\ell_{j}, \qquad (3.3.3)$$

where J is the total current density and has the direction of the wire. Combining the above formulas, we obtain

$$E^* = J \rho_{i,j} \ell_{i,j}.$$
 (3.3.4)

All the quantities E,E\*, E<sub>i</sub>,  $\rho_{ij}$  are, in general, dependent on the magnetic field  $\underline{H}_*$ . According to equation (3.3.4), the measured resistivity  $\rho(\underline{H})$  is

$$\rho(\underline{\mathbf{H}}) = \rho_{\mathbf{i},\mathbf{j}} \ell_{\mathbf{i}} \ell_{\mathbf{i}} \tag{3.3.5}$$

and the conductance  $\sigma$  is its reciprocal,

$$\sigma(\underline{\mathbf{H}}) = \left[\rho_{\mathbf{i},\mathbf{j}}(\underline{\mathbf{H}})\ell_{\mathbf{i}}\ell_{\mathbf{j}}\right]^{-1}.$$
 (3.3.6)

The connection between  $\rho_{i,j}$  and  $\sigma_{i,j}$  is

$$\rho_{ij} = \Delta_{ji}/\Delta, \qquad (3.3.7)$$

where  $\Delta$  = Det  $\sigma_{ij}$  and  $\Delta_{ji}$  the cofactor of  $\sigma_{ji}$ .

Equation (3.3.6) takes the place of equation (25), (26), (27) in the previous report. It gives the same information in much simpler form.

For the sake of completeness we repeat here the expressions which relate  $\sigma_{i,j}(H)$  to the energy function E(k) and the relaxation time t(k):

$$\sigma_{ij} = \sum_{n=0}^{\infty} H^{n} \sum_{p=0}^{n} \sum_{p'=0}^{n-p} [p', p, n-p-p']_{ij} \gamma_{1}^{p'} \gamma_{2}^{p} \gamma_{3}^{n-p-p'}, \qquad (3.3.8)$$

where

$$[p',p,n-p-p']_{ij} = \int g(E) \nabla E_i \left\{ P[(t\Omega_1)^{p'}(t\Omega_2)^{p}(t\Omega_3)^{n-p-p'}] \right\} (t\nabla E_j) dk \qquad (3*3.9)$$

and  $P[(t\Omega_1)^p'(t\Omega_2)^p(t\Omega_3)^{n-p-p'}]$  is the sum of all permutations of p' operators  $t\Omega_1$ , p operators  $t\Omega_2$ , and n-p-p' operators  $t\Omega_3$  in different order. This sum consists of n!/[p!!p!(n-p-p')!] terms.

All further theory that we plan to work out consists in elaborating on equations (3.3.6, 7, 8, and 9).

It is interesting to note in equation (3.3.5) that the measured resistivity  $\rho(\underline{H})$  can be looked at as the l'l' component of the resistivity tensor in a primed coordinate system whose first axis lies along the wire while the directions of the other axes are irrelevant. Consequently, the dependence of  $\rho(\underline{H})$  on the wire direction can be illustrated by an ellipsoid. To this end we plot in all directions  $\ell_1$ ,  $\ell_2$ ,  $\ell_3$  a length equal to  $\rho^{-1/2} = \sigma^{-1/2}$ . The locus of the endpoints plotted in this way is an ellipsoid which we shall call the magneto resistivity ellipsoid. In general, this ellipsoid will be triaxial and the lengths and directions of its axes will depend on  $\underline{H}$ . The restrictions imposed by crystal symmetry will be discussed later.\*

$$\sigma(\underline{\mathbf{H}}) = \sigma_{\mathbf{i},\mathbf{j}} \ell_{\mathbf{i}} \ell_{\mathbf{j}},$$

and similar considerations for a conductance ellipsoid can be developed. Since all work on magneto resistance that we are aware of is of the type with no transverse current, we shall not follow up further the case of no transverse electric field.

<sup>\*</sup>An experimentally different situation in which transverse currents would be allowed but the transverse electric field would be zero can be treated along very similar lines; in that case the conductance would be given by

The dependence of  $\rho$  or  $\sigma$  on the direction of the magnetic field is much more complicated. Let us assume that the direction of the wire is kept constant with respect to the crystal axes while varying the direction cosines  $\gamma_1$ ,  $\gamma_2$ ,  $\gamma_3$  of  $\underline{H}$  with respect to  $k_1$ ,  $k_2$ ,  $k_3$ . Experimentally, this means varying the direction of  $\underline{H}$  for one given wire sample. Then  $\rho_{i,j}(\underline{H})$  can be expanded in a power series in the components of  $\underline{H}$  as shown in equation (3.3.7), hence also  $\sigma$  can be expanded in such a power series. Actually, it will be shown that only the even powers occur in the expansions of  $\rho$  and of  $\sigma$ :

$$\sigma(\underline{H}) = \sum_{n} \underline{H}^{n} \sum_{\alpha\beta \dots \nu} \sigma_{\alpha\beta \dots \nu}^{(n)} \gamma_{\alpha} \gamma_{\beta} \dots \gamma_{\nu}$$
 (3.3.10)

where n = 0,2,4---. The restrictions imposed by crystal symmetry on the coefficients  $\sigma^{(n)}_{\alpha\beta}$  will be discussed later.

#### 4. The Magneto Conductance is an Even Function of the Magnetic Field

The question of the evenness of the magneto conductance has been the subject of much controversy during the past twenty years among such investigators as M. Kohler (1934), D. Shoenberg (1935), J. Meixner (1939 and 1941), H.B.G. Casimir and A.N. Gerritsen (1941) and B. Donovan and G.K.T. Conn (1950). Some of the authors mentioned have argued on both sides of the issue within a single paper. Consequently the proof of the evenness of  $\sigma$  as a function of H can be regarded as a result of physical significance. We are not aware of any conclusive proof of the evenness of the magnetoresistance in the literature.

One may, of course, always <u>define</u> a magneto conductance as the even part of the measured conductivity, calling the odd part by some other name such as "longitudinal Hall effect", Umkehreffect, etc. Shoenberg, and independently Casimir, did this. The significance of our theorem is that the odd part is identically zero.

This seems to contradict the experimental facts, reported by Donovan and Conn. These authors find in about ten percent of their bismuth samples that the magneto conductance -- and with it the magnetoresistance -- depends on the magnetic field through the combination of a quadratic and a linear term, for low fields.

Our proof of the evenness of the magneto conductance is so general that we are inclined to ascribe any odd terms, found experimentally, to some spurious effect or systematic experimental error. In the experimental part of this report we shall present evidence obtained by our group, supporting this contention. We can at will produce or eliminate linear terms such as reported by Donovan and Conn by controlling experimental details which affect

the isothermal condition of the sample. Measures that improve conditions of homogeneous temperature throughout the sample tend to eliminate the odd term. Thus the odd term, which is in our case entirely of the same character as that found by Donovan and Conn, is not a genuine magneto conductance effect. (see section C4 for further details).

We proceed now to prove the theorem.

Proof. We know that

$$\sigma_{i,j}(\underline{\mathbf{H}}) = \sigma_{j,i}(-\underline{\mathbf{H}}) , \qquad (3*4*1)$$

which is the famous relation of Onsager, recently proved again on the basis of the principle of microscopic reversibility by P. Mazur and S.R. de Groot (1954). According to (3.3.7) we have also

$$\rho_{i,j}(\underline{\mathbf{H}}) = \rho_{j,i}(-\underline{\mathbf{H}}) . \tag{3.4.2}$$

We use this relation in equation (3.3.6). There we find two types of terms:

a. Terms with equal indices ii. According to (3.4.2) we have

$$\rho_{ii}(\underline{H}) = \rho_{ii}(-\underline{H})$$
,

hence these terms are even functions of H.

b. Terms with unequal indices ij. The coefficient of  $\ell_i \ell_j$  is  $\rho_{i,j}(\underline{H}) + \rho_{j,j}(\underline{H})$ . According to (3.4.2) this is also an even function of H.

Thus  $\sigma(\underline{H})$  is an even function of  $H_*$  Since  $\rho = \sigma^{-1}$ ,  $\rho$  is also an even function of  $H_*$   $\overline{Q}_*E_*D_*$ 

Discussion of the Proof and Consequences. It should be mentioned that in Mazur and de Groot's paper, use was made of the Lorentz force vxH, whereas Kohler and Shoenberg obtain a linear term by using an ad hoc assumption of a "generalized Lorentz force" v<sub>i</sub>H<sub>j</sub>, for which we cannot see any fundamental justification. Equation (3.4.1), on the other hand, is one of the fundamental pillars for the thermodynamics of irreversible processes to which belong the conductance phenomena.

The experimenter is now justified in measuring the magneto conductance by reversing the magnetic field and averaging the two readings, to eliminate spurious effects, or by ac methods.

The evenness of the magneto conductance implies also the evenness of the magnetoresistance  $\Delta\rho/\rho_0$  = -1 +  $\sigma_0/\sigma_*$ 

# 5. The Effect of Orientation of the Wire with Respect to the Crystal Coordinate System

In sections 5, 6, and 7 the discussion centers around this question: What fundamental constants (brackets (3.3.9)) can be derived from magnetoresistance measurements and what measurements are required to derive them? If we knew all the brackets we could predict, according to equations (3.3.6, 7, 8), the magnetoresistance for any orientation of the crystal axes in the sample and of the magnetic field.

As a preliminary approach to answering this question we divide the problem into three parts: The effect of the orientation of the wire with respect to the crystal coordinate system, the effect of the orientation of the magnetic field with respect to the crystal coordinate system, and the effect of crystal symmetry on the bracket symbols. The first part will be discussed presently, the second will receive some attention in section 6, while the third is attacked in section 7.

According to equation (3.3.5), the effect of the direction of the wire with respect to the crystal coordinate system is relatively simple. The dependence of  $\rho$  on the direction cosines  $\ell_1$ ,  $\ell_2$ ,  $\ell_3$  can only be studied experimentally by measuring many samples all having the same  $\rho_{ij}(\underline{\mathbb{H}})$  but different  $\ell_1$ ,  $\ell_2$ ,  $\ell_3$ . (Two samples have the same  $\rho_{ij}(\underline{\mathbb{H}})$  if a magnetic field  $\underline{\mathbb{H}}$  with direction cosines  $\gamma_1$ ,  $\gamma_2$ ,  $\gamma_3$  with respect to a crystal coordinate system in the first sample produces the same  $\rho_{ij}$  as does a magnetic field of the same strength, oriented so as to have the same direction cosines with respect to a crystal coordinate system in the second sample.)

The study of samples along these lines would yield the six functions  $\rho_{11}(\underline{H})$ ,  $\rho_{22}(\underline{H})$ ,  $\rho_{33}(\underline{H})$ ,  $[\rho_{12}(\underline{H}) + \rho_{12}(-\underline{H})]$ ,  $[\rho_{23}(\underline{H}) + \rho_{23}(-\underline{H})]$ ,  $[\rho_{13}(\underline{H}) + \rho_{13}(-\underline{H})]$  according to equation (3.3.6). The study of more than the minimum number of samples would serve to verify the assumption that all samples had the same  $\rho_{ij}(\underline{H})$ . It is intended to look into the feasibility of such a study for the case of bismuth in the near future. On account of the evenness theorem, these measurements alone are not enough to determine the nine functions  $\rho_{ij}(\underline{H})$ . The odd parts of the functions with unequal ij must be determined from different kinds of experiments, for example of the type of Hall-effect measurements, or measurements with zero transverse electric field.

In order to describe the effect of crystal symmetry on the magneto resistivity we refer to the ellipsoid described in section 3. The elements of this ellipsoid depend on the symmetry of the reciprocal lattice of the crystal and on that of the magnetic field. If the magnetic field has any symmetry elements in common with the reciprocal lattice, these symmetry elements must also be possessed by the ellipsoid.

In considering the symmetry elements of the magnetic field we must take into account the evenness theorem, i.e., consider  $\underline{H}$  and  $\underline{-}\underline{H}$  as equivalent. The relevant symmetry elements to be associated with the magnetic field are then as follows:

An inversion center.

A rotation axis of infinite order in the field direction.

A reflection plane normal to the field direction.

An infinite bundle of reflection planes through the axis mentioned above. An infinite set of binary axes in the plane normal to the above axis.

In the case of zero magnetic field the resistivity tensor is symmetric according to equation (3.4.2) and the ellipsoid must have the same symmetry as the reciprocal lattice itself. The higher the symmetry the smaller is the minimum number of samples for measurement required to determine the six conductivity tensor components. Table II surveys the possibilities in this case.

Symmetry	Shape of ellipsoid	Ellipsoid axis directions fixed by crystal symmetry	Relations between components of $ ho_{ extbf{ij}}^*$	Minimum number of samples for measurement
triclinic	triaxial	-	-	6
monoclinic	triaxial	1	$\rho_{12} = \rho_{13} = 0**$	14
rhombic	triaxial	3	same as monoclinic and $\rho_{23} = 0$	3
trigonal tetragonal hexagonal	revolution	axis of revola along crystal axis of highest order	same as rhombic and $\rho_{22} = \rho_{33}$	2
cubic	sphere	<b>-</b>	same as tetragonal and $\rho_{11} = \rho_{22}$	1

<sup>\*</sup> The same relations also hold for the components of  $\sigma_{i,j}$ .

In the monoclinic case, for example, one axis is fixed by symmetry, the other two may depend on the temperature, etc. In the rhombic case all axis directions are independent of the temperature. For bismuth (trigonal) it

<sup>\*\*</sup> Assuming the k<sub>1</sub> axis to have the highest symmetry.

is seen that measurements on two samples of identical material would yield all information of this sort, i.e., six relations determining  $\rho_{i,i}$ 

Table III surveys the situation in the presence of a magnetic field for the various orientations of field with respect to crystal axes that produce special effects.

TABLE III

EFFECT OF CRYSTAL SYMMETRY AND SPECIAL ORIENTATION
OF THE MAGNETIC FIELD ON THE RESISTIVITY ELLIPSOID

		symmetry		
Crystal	Magnetic field	of crystal	Relations between	Minimum number
symmetry	orientation	and magn.	components of	of samples for
	·	field	$ ho_{ t i  t j}(\underline{\mathtt{H}})  imes$	measurement
monocl.	*			
•	// C2	0 1 mamaal	II	14
C <sub>2h</sub>	// 62	C <sub>2</sub> h-monocl.	TT	4
rhombic				
$\mathtt{D}_{2\mathbf{h}}$	// C2	D <sub>2h</sub> -rhombic	III	3
	, .			
trigonal				_
S <sub>6</sub>	// S <sub>6</sub>	S <sub>6</sub> -trig.	IV	2
Dsi	// C <sub>3</sub> // C <sub>2</sub>	D <sub>3i</sub> -trig.	IV	2
D <sub>3</sub> i	// C <sub>2</sub>	C <sub>2</sub> h-monocl.	II	14
tetrag.				
$\mathtt{C}_{4\mathrm{h}}$	// C₄	C <sub>4h</sub> -tetrag.	IV	2
$\mathtt{D}_{\mathtt{4h}}$	// C <sub>4</sub>	D <sub>4h</sub> -tetrag.	IV	2
$\mathtt{D_{4h}}$	// C2	D <sub>2h</sub> -rhombic	III	3
hexagonal				
C <sub>6</sub> h	// C <sub>6</sub>	Ceh-hexag.	IV	2
D <sub>6h</sub>		D <sub>6h</sub> -hexag.	IV	2
Deh	// C <sub>6</sub> // C <sub>2</sub>	D <sub>2h</sub> -rhombic	III	3
cubic <sup>T</sup> h	// c <sub>a</sub>	S <sub>6</sub> -trig.	IV	2
$\mathtt{T}_{\mathbf{h}}^{h}$	// C <sub>3</sub> // C <sub>2</sub>	D <sub>2h</sub> -rhombic	III	3
∸n		-211 - 110 20	<del>-</del>	
O <sub>h</sub>	// C4	D <sub>4h</sub> -tetrag.	IV	2
$o_{\mathbf{h}}^{n}$	// C <sub>3</sub>	D <sub>3i</sub> -trig <sub>*</sub>	·IV	2
$o_{\mathbf{h}}^{-}$	// C <sub>2</sub>	D <sub>2h</sub> -rhombic	III	3

TABLE III (cont)

EFFECT OF CRYSTAL SYMMETRY AND SPECIAL ORIENTATION OF THE MAGNETIC FIELD ON THE RESISTIVITY ELLIPSOID

Crystal symmetry	Magnetic field orientation	symmetry of crystal and magn. field	Relations between components of $p_{ij}$ ( $\underline{\underline{H}}$ )*	Minimum number of samples for measurement
all classes	_ C <sub>2</sub> , C <sub>4</sub> or C <sub>6</sub>	C <sub>2h</sub> -monocl.	II .	14
Classes	⊥ C₃ or S₀	S <sub>2</sub> -triclinio	e I	6
	all other orientations	S <sub>2</sub> -triclinio	e I	6

I means no special relation.

II means  $\rho_{12}(H) + \rho_{12}(-H) = \rho_{13}(H) + \rho_{13}(-H) = 0$  for all H (k<sub>1</sub> along H).

III means  $\rho_{i,j}(H) + \rho_{i,j}(-H) = 0$  for all unequal ij for all H.

IV means  $\rho_{ij}(H) + \rho_{ij}(-H) = 0$  for all unequal ij and  $\rho_{22}(H) = \rho_{33}(H)$  for all  $H(k_1 \text{ along } H)$ .

It is seen from this table that cubic symmetry is always destroyed in the presence of a magnetic field, and that special importance should be given to measurements in which the magnetic field lies along a crystal axis of higher order than two.

It is further evident that we need from 2 to 6 samples of equal  $\rho_{ij}(\underline{H})$  in order to determine the ellipsoid that corresponds to a particular  $\underline{H}$ . The number of measurements required for determining the dependence of the  $\rho_{ij}$  on the magnitude of  $\underline{H}$  for fixed direction of  $\underline{H}$  can be derived from Tables II and III. For example, for a cubic crystal with  $\underline{H}$  direction along  $C_4$  we need 2n+1 measurements in order to determine  $\rho_{ij}$  up to terms with  $\underline{H}^{2n}$  inclusive, namely n+1 measurements on one sample and n on another, with varying  $|\underline{H}|$ .

# 6. The Effect of the Orientation of H with Respect to the Crystal Coordinate System

The dependence on the direction of H is much more complicated. The direction cosines  $\gamma_1$ ,  $\gamma_2$ ,  $\gamma_3$  of H occur in equation (3.3.8). In order to see the influence of special values of these direction cosines we plan in the future to analyze three cases:

<sup>\*</sup>The same relations hold also between the components  $\sigma_{i,i}(\mathbb{H})$ .

- a. The general case.
- b. H normal to one crystal axis.
- c. H normal to two crystal axes.

If  $\gamma_1$ ,  $\gamma_2$ , or  $\gamma_3$  vanishes, we need to consider in (3.3.8) only terms with p' = 0, p = 0, or n-p-p' = 0, respectively. An analysis of what this implies in the various crystal symmetries and for different values of n has not yet been carried out to a point suitable for reporting.

It seems that while the effects of the direction of  $\underline{J}$  and of  $\underline{H}$  are both tractable in relatively simple forms, no simplification appears if special angles between  $\underline{J}$  and  $\underline{H}$  are investigated. This leads to the important conclusion that in those crystals which have sufficiently high symmetry to fix the coordinate axes, more easily interpreted results are obtained from experiments with simple orientations of  $\underline{J}$  with respect to  $\underline{k}$  and of  $\underline{H}$  with respect to  $\underline{k}$ , than of  $\underline{H}$  with respect to  $\underline{J}$ . The conventionally measured transverse and longitudinal effects are not, in general, the quantities best suited for obtaining theoretically significant information.

# 7. The Effect of Crystal Symmetry on the Bracket Symbols

As a consequence of the symmetry of the reciprocal lattice, a number of relations are satisfied by the bracket symbols defined by equation (3.3.9). The functions  $E(\underline{k})$  and  $t(\underline{k})$  must have the same symmetry as the reciprocal lattice, and this works itself out in certain relations involving the brackets which it will be our aim to find.

The goal would be reached if we could present all the relations imposed on the brackets for all eleven symmetry groups of the reciprocal lattice. This task requires more time than was available before the close of this report, so that we present here some results of a preliminary attack, whereby we have only investigated the effect of reflection planes.

According to the principle of time reversal, the origin of k-space is always an inversion center of the energy surfaces  $E(\underline{k})$ . We do not know of any general proof that the same is true for the relaxation time t. If the relaxation time is a function of E only then the origin of k-space is obviously also an inversion center for t. If t is a more general function of  $\underline{k}$  we shall assume that the same is true.

Thus the symmetry of E and of t belongs to one of the eleven crystallographic point groups having a center of symmetry listed in section 2. The most important simplifications arise from the presence of symmetry planes or, what amounts to the same thing, binary axes. In order to simplify our problem we shall therefore not investigate the eleven groups separately but rather class them into three categories:

- I. E and t are even in none of  $(k_1, k_2, k_3)$  individually.
- II. E and t are even in one of the  $(k_1, k_2, k_3)$  individually.
- III. E and t are even in  $k_1$ , in  $k_2$ , and in  $k_3$ .

Table IV will give us the 32 classes and the proper category number:

TABLE IV

DISTRIBUTION OF THE 32 CRYSTALLOGRAPHIC POINT GROUPS OVER THE THREE CATEGORIES

Triclinic	C <sub>1</sub> , S <sub>2</sub> *	I
Monoclinic	Cz, C <sub>s</sub> , C <sub>zh</sub>	II
Rhombic	D <sub>2</sub> , C <sub>2V</sub> , D <sub>2h</sub>	III
Trigonal	C <sub>3</sub> , S <sub>e</sub> * D <sub>3</sub> , C <sub>3V</sub> , C <sub>3h</sub> , D <sub>3i</sub> * D <sub>3h</sub>	II II
Tetragonal	$C_4$ , $S_4$ , $C_{4h}$ * $C_{4V}$ , $D_4$ , $D_{2d}$ , $D_{4h}$	II III
Hexagonal	C <sub>6</sub> , C <sub>6</sub> h * C <sub>6</sub> V, D <sub>6</sub> , D <sub>6</sub> h	II
Cubic	$T$ , $T_d$ $O$ , $T_h^*$ , $O_h^*$	III

<sup>\*</sup>The ll point groups marked with an asterisk have a center of inversion.

Thus the 32 point groups break up into

- 4 classes satisfying condition I,
- 12 classes satisfying condition II,
- 16 classes satisfying condition III.

Table V shows the effect of the three categories in regard to the brackets. Some of the results coincide with those obtained in section 10 of the previous report, but more general cases are now included. As was stated before, we hope to extend this table later to include all relations for the eleven symmetry groups of the reciprocal lattice.

We have indicated in Table V an arbitrary even number by  $\epsilon$  and an arbitrary odd number by w, as before. Symmetry makes the brackets listed in this table vanish.

TABLE V THE BRACKETS [p', p, n-p-p'] VANISH FOR THE FOLLOWING SYMMETRIES OF  $E(\underline{k})$  and  $t(\underline{k})$  WITH RESPECT TO CRYSTAL COORDINATES

	Category I		Category II		Category III		
2.5	200	no symmetry	symmetry with respect to		symmetry with respect to		
ij	n		k <sub>1</sub>	k <sub>2</sub>	kg	k <sub>1</sub> and k	and kg.
			p'=	p=	n-p-p' =	p' = or p =	
	$\epsilon$	-	W.I	W 1	w¹	W	W <sup>11</sup>
11, 22, 33						•	
	W	all	all	all	all	all	all
_	ε	=	€¹	€ †	w ¹	€¹	$\epsilon_{\mu}$
12			•	<b>T</b>	· <b>*</b>	<b>Y</b>	11
	W	<del>-</del>	W t	W t	€ †	M,	M <sub>tt</sub>
	ε		w¹	€¹	€ †	w <sup>†</sup>	€'
23	C	_	w	•		<b>w</b>	E
<b>-</b>	w	_	۠	w ¹	w ¹	€1	W <sup>†</sup>
			Č	•	••	C	W.
	€		۠	w '	۠	€'	w '
31							
-	W	_	w *	€ 1	W 1	w <sup>1</sup>	€ 1
	W	-	W	E	W	W	·E

#### C. EXPERIMENTAL PART

#### 1. Purpose

The purpose of the measurements reported is the obtaining of fundamental data on the magnetoresistance of bismuth. In order to overcome a number of technical difficulties in the measurements we have limited ourselves mainly to this one material. The technical difficulties referred to are of two sorts. First, the difficulty of mounting our wire samples satisfactorily, with good electrical contact to supports, and sufficiently rigidly to allow angular measurements. The second difficulty stems from certain sources of error that were found, studied, and eliminated in this period.

A few measurements beyond these main lines were also performed on other materials.

#### 2. Equipment

Previously, it took sometimes several hours before a good soldering contact between a bismuth wire and the copper or brass supports was made and aged so as to give stable resistance readings in various orientations. Following a suggestion of Mr. Tantraporn we have now substituted gallium wells for the contacts, which resulted in remarkable improvement of stability and ease of operation. The melting point of gallium is  $29.6^{\circ}$ C but once melted it can remain a liquid, supercooled at room temperature. Another ideal property of gallium is that it wets glass, bismuth, copper, and other materials we use. With this discovery we can now exchange bismuth wires in and out of the holder in a matter of minutes and the stability of the connection is excellent.

A new sample holder was constructed to incorporate this feature. It was also equipped with a watertight jacket for temperature-control work. The mounting electrodes underwent several changes in connection with errors to be described below. It turned out to be of extreme importance that the two electrodes have the same temperature. We ended up with electrodes of massive copper, making isothermal contact across a thin sheet of mica that provides electrical insulation.

An evaporating chamber was also added to the equipment in an attempt to evaporate bismuth films on the outside of thin glass tubing. The bismuth does not form an even film without further precautions. It was learned later that the support must not be at room temperature. Control of its temperature must still be incorporated into this part of the equipment.

The microscope sample holder, described in the second quarterly report, has been used successfully for studying the orientation of cleavage planes.

#### 3. Materials

Bismuth was used from the same sources as described in the previous report.

Carbon resistors of 20, 75, and 200 ohms were tested for MR-effect (Ohmite Mfg. Co, Chicago, Ill.).

Niobium wire was purchased from Fansteel Metallurgical Corp., North Chicago, Ill.

#### 4. Measurements

a. Axis Direction. The orientation of the cleavage plane of several Bi wire samples was determined with the microscope by means of the special holder described earlier. The same samples were subjected to X rays (this work was kindly done by Prof. Krimm of this laboratory) and the orientation of the trigonal axis determined from the diffraction spots. The same samples were finally measured for magnetoresistance as a function of the azimuth angle, that is, the angle between the transverse magnetic field and the plane through the wire and the trigonal axis.

The results were as follows. The microscope measurements of the cleavage plane agreed in all four cases to within the error of measurement (2-5°) with the X rays. In some samples the X rays indicated two crystals of nearly the same orientation. All samples were melted in glass and drawn. This result justifies omitting in the future the X-ray diffraction and determining the orientation of the trigonal axis by means of the cleavage plane only. Further work is planned to determine whether the cleavage plane remains parallel throughout a wire sample, that is, whether it is usually a single crystal under the conditions of preparation used. Indications from the azimuth measurements so far are in the affirmative, but this must be checked more systematically.

The azimuthal measurements of  $\Delta \rho/\rho$  agreed with the cleavage plane and X-ray diffraction measurements in that it was found that  $\Delta \rho/\rho$  is a maximum when the trigonal axis lies in the plane of the wire and the magnetic field, and a minimum when it is perpendicular to that plane. This result strongly supports the single-crystal nature of the samples, since the X rays covered only a relatively small fraction of the sample of the order of 1 mm, and the cleavage plane, strictly speaking, only an infinitesimal fraction, whereas the MR measurement averages over the entire length of the sample, about one inch.

b. The Linear Term of  $\Delta\rho/\rho$  as a function of H, for Weak Fields. Donovan and Conn report that in about ten percent of their samples the dependence of  $\Delta\rho/\rho$  on H for weak fields was represented by a shifted parabola, and hence can be represented by the sum of a linear and a quadratic term. Another way of stating these results is  $\Delta\rho/\rho(H) \neq \Delta\rho/\rho(-H)$ .

We have prepared a number of samples and measured their H dependence in the hope of encountering one that would show this anomaly. We also measured the azimuthal dependence of these samples since the linear term was said to be strongest for the azimuth which makes  $\Delta\rho/\rho$  a minimum. We were fortunate enough to get several samples that exhibited the linear term. However, in the course of the measurements it became clear that the linear term is not a magnetoresistance effect at all.

Figure 1 shows the results of a typical one of these samples. The shift of the parabola is clearly visible. These data were obtained with a

measuring current of 4.2 mA. Then the measurements were repeated on the same sample, keeping the absolute value of H constant, and varying the measuring current in the Wheatstone bridge. Figure 2 shows the results. For measuring currents of more than 2 mA, there is a marked difference between  $\Delta\rho(H)$  and  $\Delta\rho(-H)$ , indicating that the coefficient of the linear term depends on the measuring current.

For sufficiently small measuring current, the measurements for opposite fields are indistinguishable, and represent the true magnetoresistance effect.

The relation between the difference  $\Delta d$  in galvanometer deflection of the bridge for +H and -H, and the measuring current i, for various values of H, is now under study for possible interpretation as a thermomagnetic phenomenon.

The dependence of the linear term on the measuring current suggested that this term would disappear if strictly isothermal conditions prevailed. In examining our sample holder we noticed that the measurements shown in Figs. 1 and 2 were performed with brass electrode blocks that were separated by an air gap of about one cm. A new sample holder was made with copper electrode blocks separated only by a thin sheet of mica, in order to improve isothermal conditions.

It was found that samples showing a behavior similar to that shown in Figs. 1 and 2 when mounted in the former sample holder, showed practically no linear term (difference between +H and -H) when mounted in the latter. More precisely, the measuring current had to be raised to ten times its old value to produce in the new sample holder the same effect as was measured in the old one. Thus the coefficient of the linear term is found to depend essentially on the electrode arrangement, and this is certainly proof that it is not a genuine magnetoresistance effect, whatever its nature may be.

These results are in agreement with the theorem of the evenness of the magnetoresistance proved in section 4.

We assume that the linear term in the results of Donovan and Conn was of a similar spurious origin.

Another manifestation of the linear term was encountered in surveying the data obtained with the old sample holder. In Table III and Table V of the previous report there was a trend for  $B_1+B_2$  to decrease with increasing magnetic field in the range of 0-500 gauss. Its possible relation to the presence of a linear term was mentioned on page 26. However, in view of our evenness theorem it could also be due to an  $H^4$ -term.

The following discussion will show that the above mentioned trend is indeed due to a linear term and how its influence, once recognized, can be eliminated.

If the observed relation is of the form

$$\Delta \rho / \rho = A H + B H^2$$
,

then a plot of  $\Delta\rho/\rho H$  versus H should be a straight line. If the observed relation is

$$\Delta \rho / \rho = B H^2 + CH^4,$$

then a plot of  $\Delta\rho/\rho H^2$  should be a straight line. The former is shown for a typical sample in Fig. 3, and the latter for the same sample in Fig. 4. It is clear that Fig. 3 is a straight line within the error of measurement whereas Fig. 4 is not, showing conclusively that the trend of B is due to the linear term whose spurious nature we have recognized.

Thus, in order to find true values for B, one should take the slope of a plot of the type of Fig. 3. The data for the sample illustrated here are given in Table VI. The values  $A = 0.14 \times 10^{-6}$  and  $B = 5.48 \times 10^{-9}$  are derived from slope and intercept of Fig. 3.

TABLE VI

		_	$(AH + BH^2) \times 10^4$	
Magn. field	$(\Delta \rho/\rho) \times 10^4$	B x 10 <sup>9</sup>	calculated with	Difference of
$\mathbf{H}_{m{y}}$	observed	uncorrected	$A = 0.14 \times 10^{-6}$	columns 2 and
gauss			$B = 5.48 \times 10^{-9}$	4 in percent.
480	13.30	5.77	13.30	0.0
464.4	12.61	5 <b>.</b> 85	12.47	1.1
440.8	11.24	5.79	11.27	0.3
421.2	10.45	5 <b>.</b> 89	10.31	1.3
401.6	9.60	5.95	9 • 40	2,1
382.0	8.72	5.497	8.53	2.2
362.4	7.76	5.91	7.71	0.6
342.8	6.88	5.86	6.92	0.6
323.2	6.03	5.77	6.17	2.3
<b>30</b> 3 <b>,</b> 6	5*35	5.80	5,48	2.4
284.0	4.69	5 <b>.</b> 81	4.82	2, 8
264.4	4.11	5.88	4.20	2.2
244.8	3.73	6.22	3 <sub>*</sub> 62	2.5
225.2	3.10	6.11	3.10	0.0
205,6	2.69	6.36	2.61	30
186.0	2.25	6.50	2.16	4.0
			1,11	

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c. Other Measurements. Some 15 samples of bismuth have been measured for various azimuth and various H values, partly to substantiate the evidence reported, partly to provide material for the determination of fundamental constants. For the latter, the interpretation awaits progress in the theory, and so we postpone reporting details.

Also a large number of carbon resistors of 20, 75, and 200 ohms were measured. No magnetoresistance effect was found in our operating range, setting an upper limit for B of < 0.1 x 10<sup>-9</sup>. Also Nb wire gave no measurable  $\Delta\rho/\rho$  for the fields employed.

# 5. Conclusions

The problem of mounting bismuth wire samples was solved by using gallium wells.

A spurious linear term in the magnetoresistance was found to be due to insufficiently homogeneous temperature conditions. It vanishes for sufficiently low bridge currents and electrodes designed for best isothermal conditions.

This conclusion brings experiments in agreement with the evenness theorem reported in the theoretical part.

#### D. PROGRAM FOR NEXT INTERVAL

#### Theoretical

Further development of the theory will follow present lines.

#### Experimental

Further work on bismuth is planned, allowing complete freedom of orientation in a Cardan suspension.

Work will be done also towards obtaining evaporated Bi films and measuring their magnetoresistance.

The range of field strength will be somewhat extended.

#### E. IDENTIFICATION OF PERSONNEL

In addition to the persons identified in the previous report, the project has hired the following persons on a part time basis, all graduate students or former graduate students: Mrs. Ruth Suits, Mr. A. H. Alsaqqar, Miss K. Hanchon, Mr. B. Crane. In the period February, March, April, May, 1954, the following manhours were devoted to the project.

	Hours
Alsaqqar	
Crane	66.5
Hanchon	
Katz	158.0
Kao	704.0
Patterson	290.0
Suits	
Tantraporn	340.0
Total	***************************************

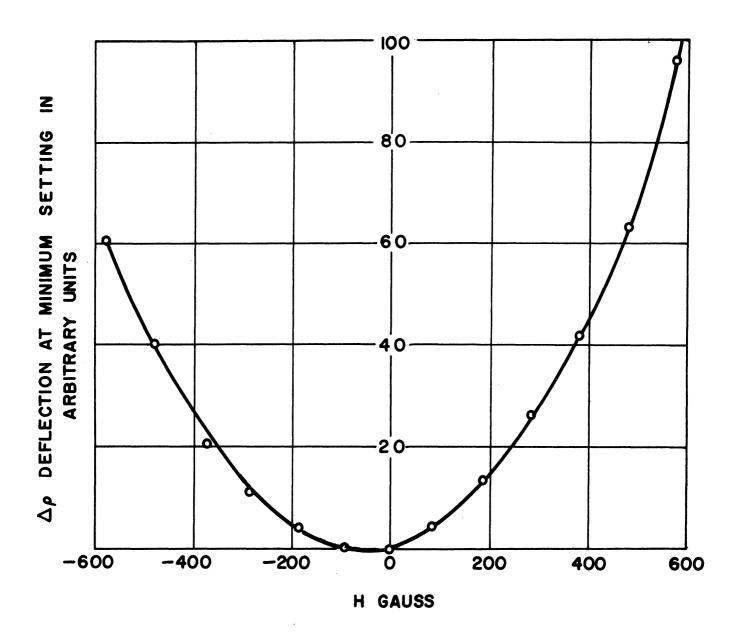


Fig. 1. The Dependence of the Magnetoresistance of a Sample on the Magnetic Field, Exhibiting the Linear Term. Measuring Current i = 4.2 mA.

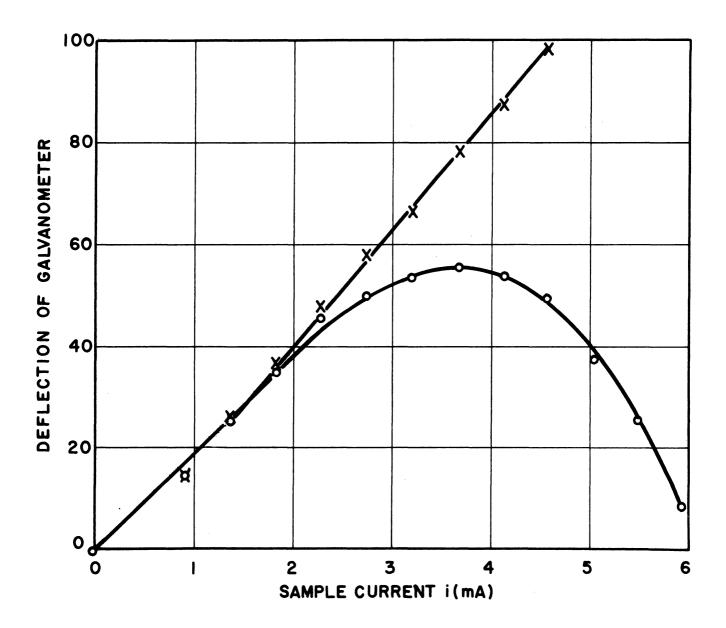


Fig. 2. The Dependence of the Galvanometer Deflection, Measuring  $i\Delta\rho$ , on the Measuring Current i, for |H|=578 gauss. Upper Curve + H, Lower Curve - H. Same Sample as in Fig. 1.

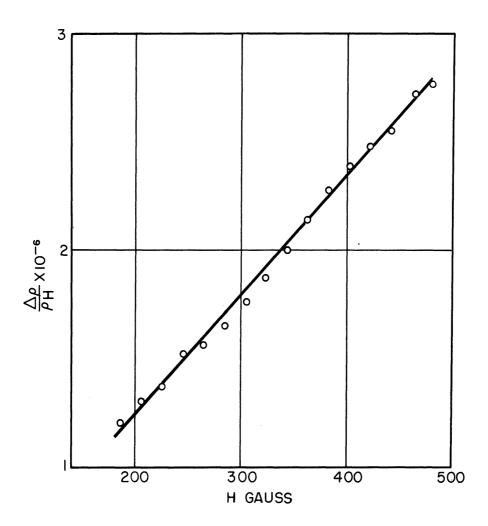


Fig. 3.  $\Delta\rho/\rho H$  Versus H Showing Straight-Line Relation. From Intercept with H = 0, A = 0.14 x 10<sup>-6</sup>. From Slope, B = 5.48 x  $10^{-9}$ 

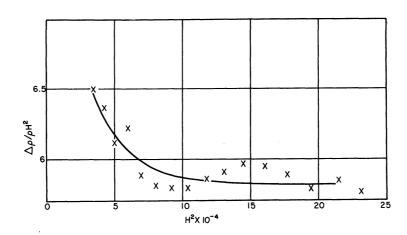


Fig. 4.  $\Delta\rho/\rho H^2$  Versus  $H^2$  Showing Nonlinear Trend for Low H. Same Data as in Fig. 3.