Low-Temperature Magnetoresistance of *n*-Type Germanium Doped with Small and Intermediate Concentrations of Phosphorus*

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(Received 2 February 1967)

The magnetoresistance ratio ρ/ρ_0 of *n*-type, P-doped Ge samples having room-temperature carrier concentrations between 7×10^{15} and 1×10^{17} has been measured at low temperatures. It is found that at 4.1°K and below, ρ/ρ_0 is roughly proportional to exp (γB^2) and depends on the crystallographic orientation of the transverse magnetic field in a manner similar to that found for *n*-type Ge doped with small concentrations of Sb or As. Our results are interpreted in terms of the influence of the magnetic field on the donor wave functions whose overlap controls the conduction process. It is found that γ goes through a minimum as a function of the donor concentration. This minimum is correlated with the transition from phonon-assisted, hopping conduction to "ϵ_2-type" impurity conduction although the concentration dependence of γ at high concentrations is unexplainable. The magnetoresistance is found to be temperature-dependent in many instances. A discussion of these results is given in terms of various hopping and band-conduction mechanisms, the relative importance of each being dependent on impurity concentration and temperature. A novel explanation of some humps in the ln ρ/ρ_0-versus-$1/T$ curves is given which depends on the randomness of the impurity distribution.

I. INTRODUCTION

INVESTIGATIONS of lightly doped ($R>4a$, where R is the average spacing between majority impurities and a is the effective Bohr radius of the wave function of the impurity ground state) and of moderately doped ($2a<R<4a$) germanium at low temperatures have demonstrated that the electrical conductivity is comprised of two or more terms which depend exponentially on temperature.[1] The relative importance of each term depends on temperature and the impurity concentrations. The activation energy of each term arises from a different cause. Thus, when carriers are freezing out of the conduction band, a σ_1 term predominates, and an activation energy ϵ_1 is observed which is interpreted to be the donor ionization energy. We shall not be concerned with this regime.

At lower temperatures, a σ_3 term predominates in lightly doped samples because of the phonon-assisted hopping of electrons between donor sites being responsible for conduction. A detailed and successful theory[2] is available for this type of conduction in zero magnetic field. The activation energy ϵ_3 depends on the compensation (ratio of the concentration of minority to majority impurities) and on the concentration of majority impurities. Investigations of the magnetoresistance of ϵ_3-type conduction have been made previously. Sladek and Keyes[3] (SK) made measurements on Sb-doped and As-doped Ge and proposed a model which explained the field dependence and the anisotropy of ρ/ρ_0 which they observed. Their model is based on the effect of the magnetic field on the impurity wave functions. Specifically, SK considered how each component of a donor wave function which is derived from a given conduction-band valley is shrunk by the magnetic field. Then, assuming that hopping occurs only in directions in which overlap of corresponding components of the wave functions on adjacent donors is large, they obtained a specific formula for the conductivity. Mikoshiba[4] (M) developed a theory to account for the data of SK which also included the difference in the phase of wave functions on adjacent donors produced by the magnetic field. His theory was incomplete in that the shrinkage and phase effects were treated separately and only for a wave function derived from a single valley. By combining these effects for each component of the donor wave function and summing over all components, Chroboczek and Sladek[5] recently were able to fit the anisotropy of ρ/ρ_0 of lightly P-doped Ge in higher magnetic fields than those used in the present work. The temperature dependence of ρ/ρ_0 observed by SK was not explainable by their model nor by the theory of M.

In moderately doped samples another activation energy ϵ_2 is observed at temperatures between the ϵ_1 and ϵ_3 (or ϵ_4)[6] regions. The conduction mechanism in this regime and the origin of ϵ_2 is still in doubt. ϵ_2 does seem to represent the separation between the lowest electronic state on a donor and some higher, more highly conducting states.[7] At one time it seemed that the latter states were a "D band"[8] comprised of overlapping orbitals of negative donor ions. However, recent theo-

* Work supported by the U. S. Army Research Office (Durham) and based on part of a thesis submitted by W. W. Lee to the Department of Physics, Purdue University, in partial fulfillment of the requirements for the degree of Doctor of Philosophy. A preliminary account of this work was given by W. W. Lee and R. J. Sladek, Bull. Am. Phys. Soc. 10, 546 (1965).

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[6] G. Sadasiv, Phys. Rev. 128, 1131 (1962).
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retical work[9] finds that the D levels are much nearer the conduction band than the original theories[8] found. This may mean that the conducting states, which are ϵ_2 above the ground state, are a band of excited atomic states.[10] Previous investigators[6,11] have made magnetoresistance measurements on n-type Ge samples in the ϵ_2 conduction region. However, they empolyed Sb- or As-doped samples and concentrated on the dependence of ρ/ρ_0 on temperature and magnetic field strength. The activation energy was found[6] to be given by $\epsilon_2+\alpha B^2$, where B is the magnetic induction. The parameter α was correlated with dependence of the impurity wave function on magnetic field strength by Sadasiv,[6] who, however, pointed out some quantitative difficulties in his interpretation. Yamanouchi[11] attributed the dependence of ϵ_2 on magnetic field strength to the spin degeneracy of the ground state being split by the magnetic field while the conducting D level remained unshifted by the magnetic field. The distance between the lower spin component of the ground state and the D levels was thus increased by the magnetic field. A statistical treatment yielded a rather complicated dependence of ϵ_2 on B. Hall-effect measurements by Yamanouchi[12] seemed to support her model for As-doped but not for Sb-doped Ge.

The present investigation employs Ge samples doped with phosphorus in concentrations appropriate to the ϵ_3 and ϵ_2 conduction regions. The dependence of ρ/ρ_0 on magnetic-field strength and temperature and the anisotropy of ρ/ρ_0 in a transverse magnetic field are studied.

II. EXPERIMENTAL DETAILS

The samples were cut by means of a cavitron from (111) slices of single crystal, n-type, phosphorus-doped, germanium ingots grown in nearly the [111] direction. Slices of such orientation were used in order to minimize concentration gradients. Each sample had a rectangular parallelepiped body of about $10\times1\times1$ mm^3 and two pairs of arms for attaching the potential leads and was lapped with 600-grit SiC powder and etched with CP4. Teflon-insulated copper wires were soldered on to the ends and arms of each sample with 50 In 50 Sn (Cerroreal) solder.

Some characteristics of the samples are given in Table I. The sample number indicates in an abbreviated fashion the room-temperature electron concentration n, deduced from the Hall coefficient R_H, measured at 25 kG (by which field strength R_H had become almost independent of field strength). Any ambiguity remaining in deducing n from R_H was slight enough to be immaterial to us. It was assumed that N_D-N_A, the excess in the concentration of donors over the concentration of acceptors, is just equal to the room-temperature value of n.

TABLE I. Characteristics of the n-type, phosphorus-doped germanium samples. R is the average spacing between donors, R/a is the ratio of the average donor spacing to the effective Bohr radius of a donor wave function, ϵ_3 is the activation energy derived from the resistivity at lowest temperatures, and N_A/N_D is the ratio of the concentration of acceptors N_A to the concentration of donors N_D.

Sample	ρ_0(298°K) (Ω cm)	R (Å)	(R/a)	ϵ_3 (meV)	N_A/N_D	N_D (10^{16} cm^{-3})
6.9–15	0.249	260	7.4	1.70	0.025	0.7
1.4–16	0.134	210	6.0	1.78	0.06	1.5
2.2–16	0.093	180	5.1	1.94	0.08	2.4
3.5–16	0.072	150	4.3	2.00	0.12	4.0
7.1–16	0.045	120	3.4	2.10	0.20	8.9
7.3–16	0.043	120	3.4	1.88	0.30	10
1.2–17	0.0261	100	2.9	2.32	0.26	16

Low-temperature measurements were made with the sample immersed in liquid helium contained in a double Pyrex Dewar flask having liquid nitrogen in the outer chamber. Various temperatures were obtained and measured by controlling and measuring the vapor pressure of the helium bath. Special care was taken to assure that the bath was in thermal equilibrium between 4.2 and 5°K.

Electrical measurements at low temperatures were made by reading the setting of an L and N, type K-2 potentiometer in the feedback loop of an Applied Physics Corp. vibrating reed electrometer used as a null detector and connected across the voltage arms of the sample or across a precision resistor to allow the current to be deduced. At room and liquid-nitrogen temperatures the potentiometer was used with a galvanometer in the usual manner.

The electromagnet used to supply the magnetic field was a Pacific Electric Motor Co. model 12C-AT-LIF. Current regulated to 1 part in 10^5 was provided by a PEM solid-state power supply. With the poles $2\frac{1}{2}$ in. apart a maximum induction of 28 kG was obtained. A Rawson rotating coil fluxmeter was used to measure the magnetic field.

III. RESULTS AND DISCUSSION

A. Temperature Dependence of ρ_0

Figure 1 shows the resistivity of each of our P-doped Ge samples in zero magnetic field at temperatures between 5 and 2°K. From Fig. 1 it can be seen that at the lower temperatures ρ_0 for each sample becomes an exponential function of $1/T$, so that a single activated conductivity process predominates and an activation energy can be deduced for each sample. For the lower concentration samples (6.9–15 to 3.5–16) this activation energy is identified with the ϵ_3 of phonon-assisted hop conduction in view of the R/a values of these samples. (See Table I.) For the higher concentration samples this activation energy is identified with ϵ_3 or with an ϵ_4 associated with some other unspecified process.[6]

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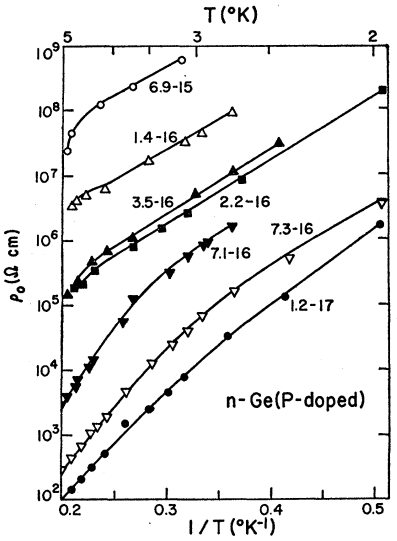

FIG. 1. Resistivity in zero magnetic field versus reciprocal temperature for all of our samples.

The values of the activation energies at lowest temperatures are listed in the ϵ_3 column in Table I. Assuming that they are to be identified with ϵ_3, we used results of Miller and Abrahams[2] (MA) to calculate the compensation, the ratio of the concentration of minority impurities to the concentration of majority impurities (N_A/N_D for n-type material), for each of our samples. In the calculation, we took the average separation between donors, R, to be given by the relation $R=(4\pi N_D/3)^{-1/3}\approx[4\pi(N_D-N_A)/3]^{-1/3}$. From the value of N_A/N_D deduced thus and the value of N_D-N_A taken equal to the room-temperature carrier concentration, we also deduced a value of N_D for each sample. Table I lists the values of R we used and the values of N_A/N_D and N_D which we obtained. Also shown are values of R/a computed using $a=35$ Å.

Figure 1 shows that departures of ρ_0 from a simple exponential dependence on $1/T$ occur which have a different character for the lightly doped and for the moderately doped samples. For the lightly doped samples near 5°K, some conduction is probably due to other processes in addition to the conduction via direct hopping of electrons between the singlet members of the donor ground state on adjacent donors. Part of the additional conduction may be due to processes discussed by MA in which electrons make a singlet to triplet transition on one donor, then a hop from the triplet state of that donor to the triplet state of a second donor, and finally a triplet to singlet transition on the second donor. Using results of MA we estimate that the additional conduction due to these indirect processes is not enough to account completely for the observed fall off of ρ_0 above 4°K, particularly for sample 6.9–15. Another part of the additional conduction must come from residual carriers in the conduction band. Of course, conduction-band conduction is expected to become important at lower temperatures the purer the sample.[1] Comparison of the ρ_0 curves for samples 6.9–15 and 1.4–16 indeed seem to indicate this effect.

For the three moderately doped samples (7.1–16 to 1.2–17), there is clear evidence that two different activation energies occur. The activation energy at the higher temperatures we identify as ϵ_2 on the basis of previous work[1,6,11] on samples of corresponding concentrations (more specifically, corresponding R/a values) since, as can be seen in Table I, for our samples in question $2<R/a<4$.

B. Magnetic-Field and Concentration Dependence of ρ/ρ_0

Detailed measurements of the dependence of the resistivity on magnetic-field strength were made at 4.1 and 3.3°K. The results can be summarized by saying that $\ln(\rho/\rho_0)$ was found to be nearly proportional to the square of the magnetic field, but that there were some deviations from this simple dependence particularly for the low concentration samples at 4.1°K. These deviations may be due to some hopping processes involving triplet states occurring in an amount which is field-dependent. Some evidence for the occurrence of such processes between 4 and 5°K in the low-concentration samples was mentioned previously and will also be provided later by the temperature dependence of the magnetoresistance of such samples. Figure 2 shows data for some of our samples at 3.3°K which illustrate how nearly $\ln(\rho/\rho_0)$ is proportional to B^2.

For our low-concentration samples, the observed field dependence of ρ is similar to that found by SK for Sb-doped and As-doped Ge samples and is explainable in terms of their model and the theory of M mentioned in the Introduction. Thus we believe it arises due to the magnetic field affecting the donor wave functions and causing a decrease in the rate at which electrons hop between donors.

For our intermediate concentration samples the field dependence of ρ/ρ_0 is similar to that observed for Sb-doped and As-doped Ge samples by other authors[6,11]

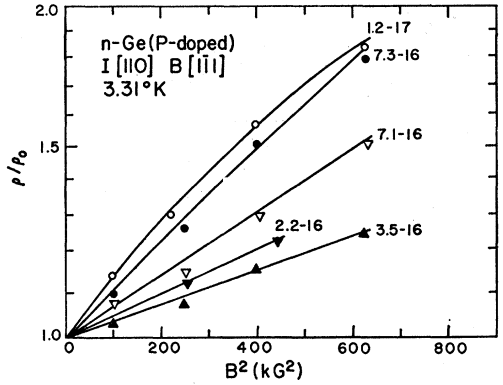

FIG. 2. Magnetoresistance ratio versus the square of the magnetic induction for five of our samples.

and interpreted by one[6] of them in terms of the dependence of the activation energy on magnetic-field strength because this energy depended on R/a, which had an appropriate field dependence according to theory.[2] The actual origin of ϵ_2 was not specified.

In order to display the concentration dependence of ρ/ρ_0 we have plotted the value of $\gamma \equiv [\ln(\rho/\rho_0)]/B^2$ at 25kG for each of our samples at 3.3°K as a function of the majority impurity concentration N_D in Fig. 3. Note that at low concentrations (i.e., $R/a > 4$)γ decreases as N_D increases, and at intermediate concentrations $(2 < R/a < 4)\gamma$ increases as N_D increases.

For our low-concentration samples, the inverse dependence of γ on concentration is explainable qualitatively in terms of a formula from SK, namely,

$$\gamma = K/32 m_t c^2 N_D, \quad (1)$$

where K is the dielectric constant, m_t is the effective mass transverse to a valley axis, c is the velocity of light, and N_D is the donor concentration. However, even for our lowest-concentration samples $\gamma \sim N_D^{-0.8}$ rather than varying like N_D^{-1} as predicted by Eq. (1). Some departure from a N_D^{-1} dependence might be expected from the theory of M because of the phase effect causing a more complicated dependence of the resonance energy on concentration than does the shrinkage effect. However, the departure predicted is opposite to that which we observe. Looking at SK's plot of their γ-versus-N_D data, it can be seen that if a straight line is drawn exactly through their two data points for Sb-doped Ge, it lies above both their data points for As-doped Ge. A separate straight line drawn through the As-doped Ge data points is about parallel to, and below, the line drawn through the Sb-doped Ge data. Separate lines for the Sb-doped and As-doped Ge data may be justified qualitatively if an effective mass which depends directly on the ionization energy of the impurity is used in Eq. (1) rather than m_t of the band valley. The slope of each line is close to -0.7 and is in better agreement with the -0.8 value found for our P-doped samples than in the -1.0 value predicted by Eq. (1). For very strong magnetic fields, previous work[4,13] indicate that $\gamma \sim N_D^{-2/3}$. However, neither SK's, nor our experimental conditions satisfy the strong-field criterion[4] $R/a \ll KB^2R^3/16m^*c^2$ unless R represents impurity spacings much larger than the average separation. This would imply that high resistance jumps control the concentration dependence of the magnetoresistance but not the magnitude of it, since, as will be seen directly below, the magnitude of the magnetoresistance can be accounted for reasonably well using an R characteristic of the average spacing between impurities.

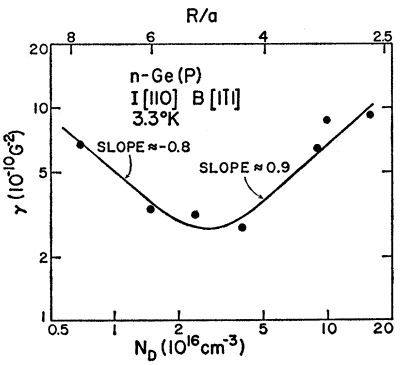

FIG. 3. Concentration dependence of the magnetoresistance parameter γ deduced from $[\ln(\rho/\rho_0)]/B^2$ at 25 kG.

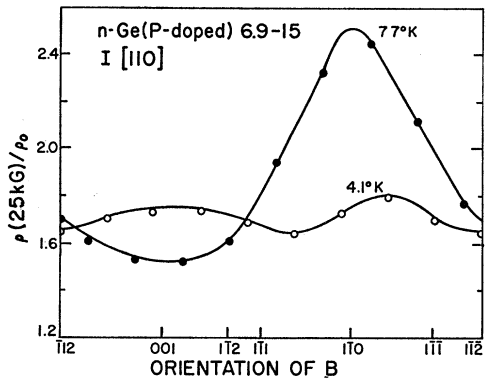

FIG. 4 Comparison of transverse magnetoresistance anisotropy at 77 and 4.1°K for sample 6.9–15.

Using Eq. (1) for our lowest-concentration sample yields a value for γ of $11 \times 10^{-10}\ G^{-2}$ which is about 1.6 times our experimental value. In arriving at Eq. (1), SK set $R^3 = N_D^{-1}$, whereas some evidence[14] favors $R^3 = (4\pi N_D/3)^{-1}$. If we substitute the latter for N_D in Eq. (1), we obtain a value for γ of $2.6 \times 10^{-10}\ G^{-2}$, which is about 2.7 times smaller than our experimental value. In view of the uncertainties involved it seems that the magnitude of the γ values which we observe at low concentrations are in quite good accord with that expected for the type of mechanism thought to be operative.

For our intermediate concentration samples the increase of γ with N_D clearly implies that the conduction mechanism must be different than it is at low concentrations. However, such a dependence of γ on N_D cannot be accounted for in terms of previous explanations of the magnetoresistance in the ϵ_2 regime nor by the theories of Frood[10] or Mycielski.[15] In principle it could be accounted for by the theory of Nishimura[8] for a D-band model if the screening constant in the wave function of the D^- ion increased fast enough with impurity concentration to overcome the explicit direct linear decrease of ϵ_2 on $N_D - N_A$. We regard this as highly unlikely.

In view of the above discussion, the dependence of γ on N_D which we observe in intermediately doped

[13] R. J. Sladek, J. Phys. Chem. Solids **5**, 157 (1958).
[14] See, for example, N. F. Mott and W. D. Twose, Phil. Mag. Suppl. **10**, 107 (1961).
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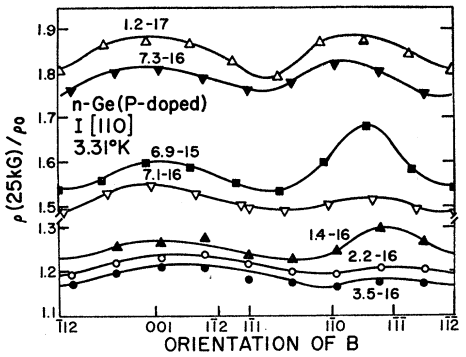

FIG. 5. Transverse magnetoresistance anisotropy for all our samples at 3.31°K.

samples at 3.3°K may be due to the conduction process being more complicated than envisaged by the simple models of ϵ_2-type conduction. Although we have no definite alternative explanation, possible explanations might involve field-dependent amounts of hopping and/or narrow-band conduction processes associated with various types of impurity states.

C. Anisotropy of ρ/ρ_0

The dependence of ρ/ρ_0 on the orientation of the magnetic field as it is aligned parallel to various crystalline directions in a plane perpendicular to the length of the sample (and current direction therein) is shown for sample 6.9–15 in Fig. 4 for 77 and 4.1°K. From Fig. 4 it can be seen that ρ/ρ_0 exhibits a different type of anisotropy at 4.1 than at 77°K. The 77°K results are characteristic of band conduction in n-type Ge.[16] Anisotropy curves similar to the 4.1°K results were obtained for all of our samples at 4.1 and 3.3°K, data for 3.3°K being shown in Fig. 5. These liquid-helium anisotropy curves are similar to those observed by SK for lightly doped Sb-doped Ge and can be accounted for by using their formula. Since as mentioned in the Introduction, this formula is based on the influence of a magnetic field on the donor wave functions, we attribute the anisotropy of our ρ/ρ_0 results to the character of the donor wave functions. Such an explanation is also consistent with the theory of M. Note that we have obtained anisotropy curves for more heavily doped samples than those of SK as well as for samples having concentrations comparable to theirs. The similarity of the anisotropy curves of our intermediately doped samples to those of our lightly doped samples we interpret to mean that conduction is controlled by wave-function overlap in the intermediate-concentration samples as well as in the low-concentration ones. Thus, samples in which only ϵ_2-type conduction is expected to be important must actually have some non-negligible component due to hopping conduction.

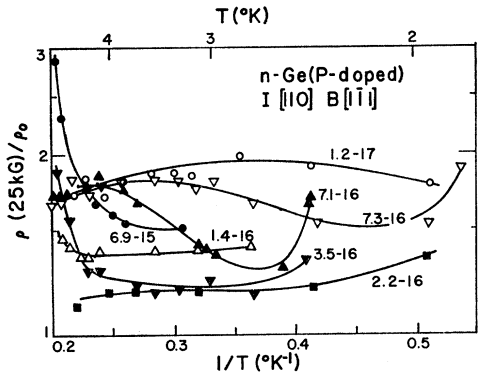

FIG. 6. Temperature dependence of the magnetoresistance for all our samples.

D. Temperature Dependence of ρ/ρ_0

The dependence of the ratio of ρ at 25kG to ρ_0 is shown as a function of $1/T$ in a semilog plot in Fig. 6. Note that for three of the low-concentration samples ρ/ρ_0 exhibits a rather strong temperature dependence between 5 and 4°K. This behavior seems to be correlated with the departure of $\ln\rho_0$ from a $1/T$ law (discussed in Sec. III A.) Another pertinent fact to be noted is that between 4 and 5°K our p-type, Ga-doped Ge samples[17] exhibit neither this strong temperature dependence of ρ/ρ_0 nor a fall of ρ_0 below the low-temperature $1/T$ law. The absence of these effects seems to indicate that neither hopping conduction involving excited states nor valence band conduction has become important in Ga-doped Ge at the temperatures in question. Since the ionization energy of phosphorus[18] in Ge is even greater than that of gallium[19] in Ge, it seems likely that at least part of the temperature dependence of ρ/ρ_0 of our low-concentration samples between 5 and 4°K must be due to the fact that the donor ground state is split into a singlet and a triplet,[20] while the acceptor ground state remains fourfold degenerate.[20]

We have not attempted to account for the observed temperature dependence of ρ/ρ_0 between 5 and 4°K by adapting the MA theory of the singlet-triplet-singlet hopping process to the case of a magnetic field because of the complexity of such a procedure. Instead we derived an expression for ρ/ρ_0 by considering conduction in triplet states to exhibit larger magnetoresistance than parallel conduction in singlet states, with the concentration of electrons in the triplet relative to that in the singlet being $3\exp(-4\Delta_c/kT)$, where $4\Delta_c$ is the distance between the singlet and the triplet and is equal to 2.8 meV for phosphorus[18] in Ge. Upon inserting reasonable

[16] G. L. Pearson and H. Suhl, Phys. Rev. **83**, 768 (1951); L. Gold and L. Roth, *ibid.* **103**, 61 (1956).

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[18] J. H. Reuszer and P. Fisher, Phys. Rev. **135**, A1125 (1964).

[19] R. L. Jones and P. Fisher, J. Phys. Chem. Solids **26**, 1125 (1965).

[20] W. Kohn, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1957), Vol. 5, p. 274.

values for γ of the singlet and γ of the triplet, it is found that ρ/ρ_0 is much less temperature-dependent than we have observed. Thus, we conclude that a conduction-band effect must also be involved to explain the temperature dependence of ρ/ρ_0 although the preceding comparison with data for p-type Ge seemed to make this unlikely. A simple model for the temperature dependence of $\Delta\rho/\rho_0$ which involved freezeout of carriers from the conduction band was given by Fritzche[1] for low magnetic fields. We shall not pursue this issue further in view of the complexities and ambiguities involved.

From Fig. 6 it can be seen that in part of the temperature range, ρ/ρ_0 of our low-concentration samples is almost independent of temperature as expected for ϵ_3-type conduction.

Our intermediate-concentration samples show various behaviors. Some of the complexity is probably due to more than one conduction mechanism being important. Our 1.2–17 sample, which, however, should be in the ϵ_2 regime at least between about 4.1 and 3°K, exhibits a ρ/ρ_0 which does not increase with decreasing temperature in the simple exponential manner found to be the case for Sb-doped and As-doped germanium.[6] We cannot explain this other than to suggest that pure ϵ_2-type conduction may not be occurring. The general falloff of ρ/ρ_0 at low or intermediate temperatures may be indications of a transition to ϵ_3-type (or ϵ_4-type) conduction. The rather sharp rises in ρ/ρ_0 of samples 3.5–16, 7.1–16, and 7.3–16 at low temperatures are quite puzzling.

Some of the undulations in the $\ln(\rho/\rho_0)$-versus-$1/T$ curves may arise from the randomness of the impurity distribution. To check on this idea, an approximate relation for ρ/ρ_0 was derived for the case of ϵ_3-type conduction by making an obvious modification of an expression by MA for the probability of transition of electrons between donors. The relation we obtained is

$$\rho/\rho_0=\int n(\Delta)n_q\Delta d\Delta \Big/ \int n(\Delta)n_q \exp(-\gamma B^2)d\Delta, \quad (2)$$

where the integrals are over the various values of Δ which occur due to the randomness of the impurity distributions[2], Δ is the difference in energy between adjacent donors, $n(\Delta)$ is the distribution of Δ's and is taken to be that for a Gaussian distribution of impurities, and n_q is the density of phonons.

Using Eq. (2) for a majority impurity concentration of 10^{16} cm^{-3} and $B=30$ kG, we obtained the result shown in Fig. 7. Thus, a single hump could occur for phonon-assisted hop conduction involving singlet-singlet transitions. For higher impurity concentration or larger γ, the hump shifts toward lower temperature. Note that for different samples, humps in ρ/ρ_0 do occur at different temperatures. More than one hump for a given sample could also be explained in terms of randomness of the impurities if parallel conduction in various types of states are involved in the hopping process.

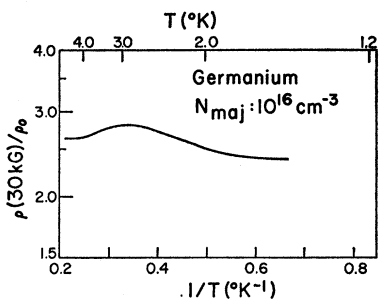

FIG. 7. Magnetoresistance versus reciprocal temperature calculated for a Gaussian impurity distribution using Eq. (2).

Our data and the results of the simple calculation above seem to indicate that the randomness of the impurity distribution may be observable in dc hop conduction via the magnetoresistance, at least for concentrations on the border line between ϵ_3-type and ϵ_2-type conduction. Theory for the effect of randomness on the magnetoresistance of ϵ_2-type conduction should be developed and a more rigorous treatment of the ϵ_3 regime be made before any more detailed conclusions can be drawn.

IV. CONCLUSION

Magnetoresistance measurements at low temperatures on n-type Ge samples doped with small or intermediate concentrations of phosphorus help to characterize some of the conduction mechanisms and electronic energy states involved in impurity conduction but indicate that more theory is needed for a complete understanding of impurity conduction, especially at intermediate concentrations of impurities. Some information about the impurity distributions is deducible from the temperature dependence of the magnetoresistance.

ACKNOWLEDGMENTS

We wish to acknowledge very helpful and stimulating discussions and correspondence with Professor A. Miller of Syracuse University. Thanks are due to Louise Roth for growing the crystals and to Professor H. J. Yearian for x-ray orientation of them.