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# NEGATIVE MAGNETORESISTANCE OF COMPENSATED *n*-TYPE GaAs AT 77°K TEMPERATURE*

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The magnetoresistance of strongly compensated *n*-type GaAs doped with Cr and O has been measured at 77 °K temperature. The electron concentrations of the samples were between $3.8\times10^{17}$ and $1.6\times10^{18}$ $cm^{-3}$ at room temperature. In low magnetic fields negative magnetoresistance was observed at liquid nitrogen temperature. The characteristics of this anomalous negative magnetoresistance were shown to be very similar to those observed at liquid He temperatures in heavily doped semiconductors. In weak fields the negative magnetoresistance was proportional to the square of the magnetic field, and showed saturation in the magnetic field range of 4000—6000 G. To account for the observed effects it was postulated that metal-like impurity band conduction is the dominant mechanism of the charge carrier transport in our samples. The observed features of the negative magnetoresistance are shown to be consistent with the model of scattering of electrons in the impurity band by localized magnetic moments. For the average value of the localized magnetic moments a value of the order of 2—3 Bohr magneton was deduced from the experimental data. It is shown that the strong compensation may play an important role in the formation of impurity band at 77 °K temperature and above it.

## Introduction

Negative magnetoresistance is frequently observed in heavily doped semiconductors at very low (1—10 °K) temperatures. At liquid He temperature (4.2 °K) it was found in various semiconductors, notably in *n*- and *p*-type InSb, in *n*-type Ge doped with As and Sb, in *n*-type GaAs [1], [2], further in *p*-type GaAs and *n*-type CdS [1], in *n*-type Si doped with P, in *p*-type Si doped with B, in Ge doped with Cu, in *n*-type Ge doped with P, in *p*-type GaAs doped with Cd [2], and quite recently in *p*-type CdSb [3].

The existence of negative magnetoresistance in *n*-type GaAs at temperatures below 20—30 °K was established by R. BROOM et al. [4], in 1956. Their crystals were undoped, having room temperature electron concentration of the order of $10^{17}$ $cm^{-3}$; but their samples were much more contaminated judging by the comparatively low mobility values measured by them. Negative magnetoresistance of undoped and of S- and Se-doped *n*-type GaAs at very low temperatures was thoroughly investigated by O. V. EMELYANENKO et al. [5]. Their samples had electron concentrations in the range of $3\times10^{15}$— $2\times$ $\times10^{17}$ $cm^{-3}$. They have shown that the negative magnetoresistance at 4.2 °K

* Dedicated to Prof. P. GOMBÁS on his 60th birthday.

temperature was most pronounced in the electron concentration range of $10^{16}$— $10^{17}$ cm$^{-3}$. J. F. Woods and C. Y. Chen have measured negative magnetoresistance in $n$-type GaAs with electron concentrations between $2\times10^{15}$ and $3\times10^{18}$ cm$^{-3}$ at liquid He temperatures, and in Cd-doped $p$-type GaAs with hole concentrations of the order of $10^{17}$ cm$^{-3}$ [2]. In one of their samples, with an electron concentration of $6\times10^{15}$ cm$^{-3}$, the negative magnetoresistance turned to be positive above 30 °K temperature. Quite recently L. Halbo and R. J. Sladek have measured the negative magnetoresistance at 4.2 °K temperature in undoped $n$-type GaAs of comparatively high purity [6].

The above mentioned results show that the occurrence of negative magnetoresistance at very low temperatures and at moderate and high doping levels is a common phenomenon in various semiconductors. Its main features are the following.

In low magnetic fields the specific resistivity decreases. This decrease is either quadratic in the magnetic field [1], [5], [6], or approximately linear in it [2]. In certain cases anisotropy consistent with the cubic symmetry is seen in these fields. The decrease of the resistivity becomes less steep as the field is increased, and saturates at a certain magnetic field. Further increase of the magnetic field results in a reversal of this trend, and the resistivity begins to increase. In higher fields magnetoresistance turns to be positive and is proportional to the square of the magnetic field.

With increasing temperature the negative magnetoresistance decreases and at a certain temperature it completely disappears. This temperature is usually of the order of 20—30 °K.

It is supposed that the measured total magnetoresistance at low temperatures is composed of two parts. One of them is the usual positive component proportional to the square of the magnetic field, the other is the anomalous and temperature dependent component, which predominates in the lower fields and saturates at intermediate fields [1].

The appearance of negative magnetoresistance is usually accompanied by a characteristic peak in the Hall coefficient versus temperature curve. The presence of this maximum on the Hall curve, and the fact that the negative magnetoresistance is observed in crystals having high impurity concentrations indicates that this anomalous magnetoresistance is connected with the impurity conduction [1], [2], [5]. At high doping levels impurity conduction is manifested through impurity band conduction, and at lower doping levels through hopping conduction. The transition between them is not sharp [2]. According to O. V. Emelyanenko et al. [5], in highly doped $n$-type GaAs at low temperatures the impurity band conduction is the dominant mechanism of charge transport.

Up to recent times negative magnetoresistance was observed only at very low temperatures, usually below 30—50 °K. Quite recently O. V.

EMELYANENKO et al. [7], observed negative magnetoresistance and impurity conduction in *n*-type GaAs, compensated with Cu diffusion, up to 200 °K temperatures. At the same time B. S. LISENKER et al. [8], observed impurity band conduction in Fe-doped *n*-type GaAs in the temperature range of 77—200 °K. Preliminary results of observations of negative magnetoresistance in Cr-doped *n*-type GaAs at 77 °K temperature were reported by the author of this paper [9].

This negative magnetoresistance, which was observed at liquid nitrogen or higher temperatures only in strongly compensated *n*-type GaAs [7], [9], is similar to the effect found at very low temperatures. It was suggested by O. V. EMELYANENKO et al. [7] that the negative magnetoresistance at higher temperatures is connected with the impurity conduction, most probably with impurity band conduction.

In this work results of observations of negative magnetoresistance at 77 °K and higher temperatures in compensated, Cr- and O-doped *n*-type GaAs single crystals are presented. In the second part of this paper sample preparation and measurement methods are described. In the third part the experimental results are presented. In the fourth and last part the results of magnetoresistance measurements are discussed and compared with other results and with the existing theoretical models.

## Experimental techniques

*n*-type GaAs crystals were prepared by the horizontal Bridgman method by E. PAPP and his coworkers at the Research Institute for Metallic Industry, Budapest [10]. According to our observations undoped crystals were of *n*-type, with an electron concentration of the order of $10^{17}$ cm$^{-3}$. Some crystal, were slightly doped either with Cr or with O during the growth process, buf remained of low resistivity with *n*-type conduction. Stronger doping with Cr produced semi-insulating crystals of high resistivity. For our measurements samples cut out from Cr- and O-doped as well as undoped *n*-type GaAs crystals of low resistivity were used.

Prism-shaped samples with dimensions of $12 \times 2 \times 0.5$ mm were cut from the crystals. The samples were not oriented. Electrical contacts were made by alloying 0.5 mm diameter In dots to the samples at 450 °C temperature in $H_2$ atmosphere [11], then 0.1 mm diameter gold wires were connected to them by the help of a micro-soldering pin. Two contacts at the ends of the samples served as current leads, and four, two on each face with dimensions of $12 \times 0.5$ mm served as potential probes. The separation between the potential probes was about 5 mm.

While the width and thickness of the samples could be easily determined with high accuracy, the determination of separation between the potential

probes, which is necessary for the evaluation of resistivity, is more uncertain because the contact dimensions are not small in comparison with contact separation, actually the former is about 10 per cent of the latter. Besides, the finite dimensions of contacts cause a distortion of current pattern in the sample resulting in errors in the measurements of the magnetoresistance. The contact separation was taken to be equal to the distance between the centres of contact dots, thus causing a maximum error of about 5 per cent in the determination of the resistivity, and an error of the order of 5—10 per cent in the determination of magnetoresistance.

Conductivity, Hall coefficient, Hall mobility and magnetoresistance were measured by the usual d.c. compensation method at 77° and 300 °K temperatures. On some of our samples these parameters were measured in the function of temperature between 77 °K and 400 °K temperatures. Measurements at 77 °K were made simply by immersing the sample into liquid $N_2$. Temperatures between 80° and 400 °K were produced in a cryostat, similar to the one described in [12]. The maximum magnetic field was about 8000 G, and it was measured with an estimated accuracy of 2 per cent. Four readings were taken to get each value of the Hall coefficient, properly commuting the sample current and the magnetic field. A similar method was used when measuring the magnetoresistance. The zero-field resistance was measured before the first and after the fourth measurement. In this way the accuracy of the measurements of the magnetoresistance was greatly improved. The potential measuring circuit had a sensitivity of 1—2 $\mu$V depending on the resistance of the sample and of the contacts. Magnetoresistances of the order of $5 \times 10^{-5}$ could be determined without causing inadmissible power dissipation in the sample.

## Experimental results

Five samples cut out from four different crystals were measured. One crystal was undoped, two were doped with Cr and the fourth was doped with O. From one of the Cr-doped crystals two samples were cut, from near to both ends of the crystals. The results of Hall and resistivity measurements are summed up in Table I. The geometry of the samples allowed to measure two Hall coefficients and two resistivities on each sample, from these values we could judge the homogeneity of our samples. The differences between the two resistivities measured on each side of the samples did not exceed 10 per cent, and the differences in the measured Hall coefficients did not exceed 10—15 per cent, except samples cut out from crystal No. 178, where they reached 20—30 per cent, showing that these two samples had a considerable inhomogeneity in macroscopic dimensions. Inhomogeneities of the same order were detected by J. F. WOODS and C. Y. CHEN [2], in some of their samples,

**Table I**

| Sample N° | 158/1 | 176 | 178/*a* | 178/*b* | 177 |
|---|---|---|---|---|---|
| Dopant | — | Cr | Cr | Cr | 0 |
| Cond. type | *n* | *n* | *n* | *n* | *n* |
| $R_H$ cm³/Asec $T = 300$ °K | 75.0 | 10.1 | 5.3* | 16.8* | 3.9 |
| $\mu_H$ cm²/Vsec T = 300 °K | 3860 | 3700 | 2200 | 1800 | 1930 |
| $R_H$ cm³/Asec $T = 77$ °K | 93.0 | 10.3 | 5.4* | 17.6* | 4.0 |
| $\mu_H$ cm²/Vsec $T = 77$ °K | 3960 | 3300 | 1900 | 1250 | 1350 |
| $n$ cm$^{-3}$ $T = 300$ °K | $8.3\times10^{16}$ | $6.2\times10^{17}$ | $1.2\times10^{18}$ | $3.8\times10^{17}$ | $1.6\times10^{18}$ |
| $N_d$ cm$^{-3}$ | $3.5\times10^{17}$ | $1.0\times10^{18}$ | $2.4\times10^{18}$ | $2.3\times10^{18}$ | $4.3\times10^{18}$ |
| $N_a$ cm$^{-3}$ | $2.6\times10^{17}$ | $0.4\times10^{18}$ | $1.2\times10^{18}$ | $1.9\times10^{18}$ | $2.7\times10^{18}$ |
| $K_n$ | 0.7 | 0.4 | 0.5 | 0.8 | 0.6 |

* Averaged values. Measured values showed an inhomogeneity, amounting to 20—30 per cent.

not influencing, however, their measurements of negative magnetoresistance.

The electron concentration as well as the donor and acceptor concentrations can be determined approximately from the room temperature Hall coefficient and the Hall mobility values in the following way. The electron concentration and the Hall coefficient are connected through the equation $n = r/eR_H$, where $r$ is the scattering factor, the value of which was put equal to 1 approximately. The electron concentrations estimated in this way are shown in Table I too.

Taking into account the above mentioned two scattering mechanisms, the measured mobility can be written in the usual approximation as $\mu^{-1} = \mu_I^{-1} + \mu_P^{-1}$, where $\mu_I$ and $\mu_P$ are the electron mobilities, caused by the two scattering mechanisms separately. The best theoretical value for the mobility caused by the scattering on polar optical phonons is $\mu_P = 9300$ cm²/Vs [13], [14]. This was confirmed by recent mobility measurements too [15]. With the help of this value the mobility caused by the scattering on ionized impurity atoms can be deduced for each sample. This mobility depends only on the electron concentration, the Fermi level and the ionized impurity concentration, and thus with the help of the Brooks—Herring formula (see e.g. [16], [17]) the concentration of ionized impurity atoms can be deduced. Because all impurity atoms are ionized at room temperature this gives the total impurity

concentration. Knowing the electron and impurity concentration, the donor and acceptor concentrations are easily obtained. Table I contains these values too with the compensation degree defined as $K_n = N_a/N_d$. All our samples are considerably compensated, but as we shall see later, the undoped sample did not exhibit negative magnetoresistance. Because of the exceptionally low mobilities measured at 77 °K in samples Nos. 178/a, 178/b and 177 we can presume that the compensation is caused by the presence of the Cr and O impurities, respectively. For sample No. 176 the role of the Cr dopant can be inferred only indirectly, by comparing the magnetoresistance results of the Cr-doped samples.

It is well known that Cr gives rise to a deep acceptor level in GaAs [13], at 0.81 eV below the conduction band edge [18]. Strong Cr doping produces semi-insulating GaAs [13], with room temperature resistivities of the order of $10^8$ ohmcm [18]. O-doping also gives rise to high resistivity semi-insulating crystals [13]. As we have mentioned in the second part of this paper, our samples were only slightly doped with Cr and O. Stronger doping with Cr produced high resistivity crystal, with room temperature resistivities of $10^8$ ohmcm. We have measured the temperature dependence of resistivity of two such samples over the temperature range of 290°—400 °K and obtained activation energies of 0.825 and 0.81 eV, respectively, which are in good agreement with the activation energy measured by G. A. ALLEN [18]. This fact confirms our arguments concerning the role of Cr impurity in our magnetoresistance samples.

The conductivity and the Hall coefficient in function of the reciprocal temperature for some of our samples is shown in Fig. 1. The Hall mobility versus temperature curves are presented in Fig. 2. The Hall coefficient versus temperature curves for Cr-doped samples Nos. 178/a and 178/b and for O-doped sample No. 177 are horizontal, not showing the characteristic maximum above 77 °K which was observed in Cu-doped *n*-type GaAs by O. V. EMELYANENKO et al., [7] and in Fe-doped *n*-type GaAs by B. S. LISENKER et al. [8].

Transversal magnetoresistances measured at room temperature in the undoped, in a Cr-doped and in the O-doped samples are shown in Fig. 3. The measurements were performed with the magnetic field perpendicular to the plane with dimensions of $12 \times 2$ mm. In accordance with other experimental results [19], the transversal magnetoresistance had not been changed by rotating the sample by 90° around an axis, parallel to the length of the sample. This agrees with the well-established model of the conduction band of GaAs having a spherical minimum in the middle of the Brillouin zone [13], [14]. It can be seen that the magnetoresistance has a quadratic dependence on the magnetic field, which is a natural consequence of the fact that the measurements were performed in small magnetic field, i.e. $\mu_H B \cong \omega_c \tau \ll 1$, where $\omega_c$ is the cyclotron frequency and $\tau$ is the relaxation time for electrons.

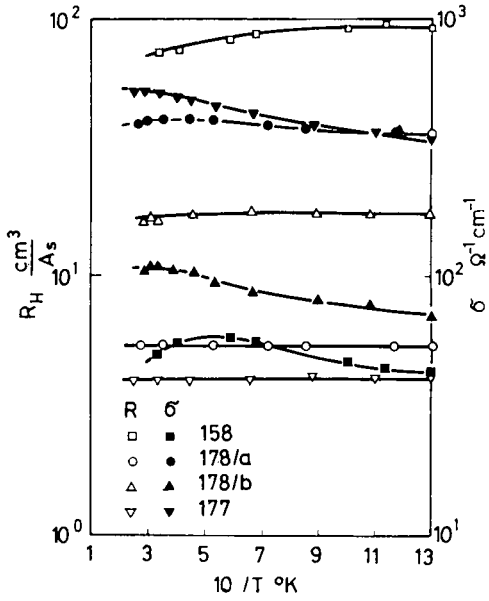


*Fig. 1.* Conductivity and Hall coefficient vs reciprocal temperature for undoped (No. 158), O-doped (No. 177) and Cr-doped (Nos. 178/a and 178/b) samples

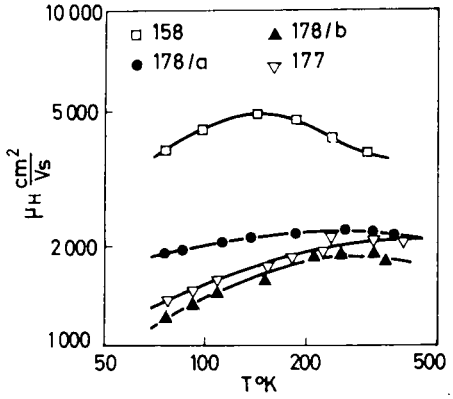


*Fig. 2.* Hall mobility vs temperature for undoped (No. 158), O-doped (No. 177) and Cr-doped (Nos. 178/a and 178/b) samples

Transversal magnetoresistance measured at 77 °K in the Cr-doped samples are shown in Fig. 4, the same measured on the O-doped and undoped samples are presented in Fig. 5. While the undoped sample exhibits normal positive magnetoresistance with quadratic dependence on the magnetic field, the value of which is in accordance with the prediction based on the assumption that at such low temperatures the scattering by ionized impurities determines the electron mobility as was shown in our earlier work [17], the O- and Cr-doped samples show an anomalous negative magnetoresistance.

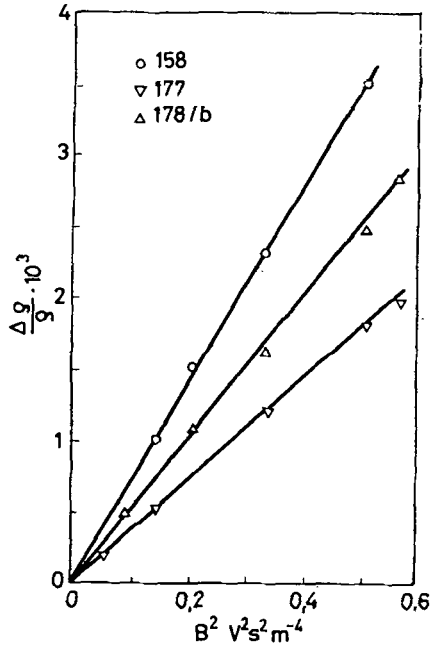


*Fig. 3.* Magnetoresistance vs square of the magnetic field at room temperature for undoped (No. 158), O-doped (No. 177) and Cr-doped (No. 178/b) samples

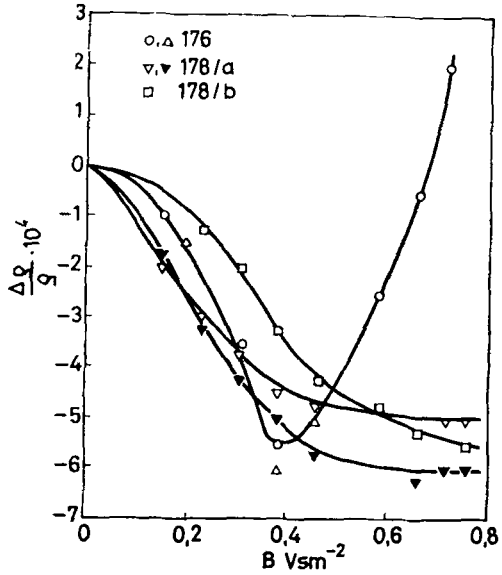


*Fig. 4.* Magnetoresistance of Cr-doped samples at 77 °K temperature

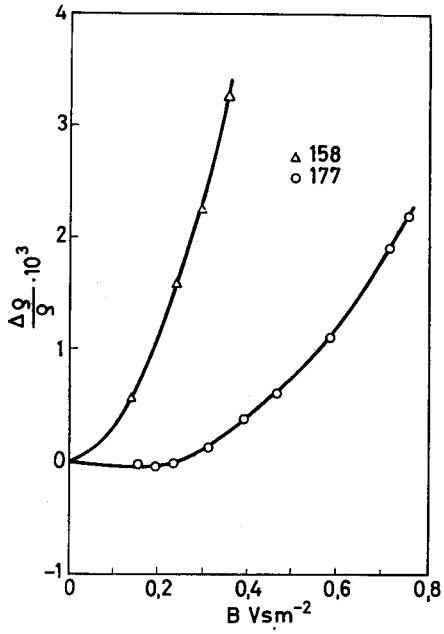


*Fig. 5.* Magnetoresistance of undoped (No. 158) and O-doped (No. 177) samples at 77 °K temperature

In the case of sample No. 177 which was doped with O there is a very small negative magnetoresistance for magnetic fields below 2500 G, the magnitude of which is hardly higher than the detection limit, but its presence was established beyond doubt in several independent runs of the measurement. The results of measurements were independent of the pair of potential contacts on which the magnetoresistance has been detected, or of the rotation of the sample around its length axis with 90°.

On the Cr-doped samples the negative magnetoresistance appeared quite markedly, as shown in Fig. 4. The negative magnetoresistance of samples Nos. 178/a and 176 after the initial increase with the magnetic field reached its maximum or saturated at moderate fields of 4000—6000 G, then its trend reversed and in the case of sample No. 176 it turned to be positive above 6500 G. The negative magnetoresistance of sample No. 178/b saturated at somewhat higher fields. For these samples there were occasionally slight but not significant deviations between the magnetoresistances measured on the same sample but on different pairs of contacts, or after the rotation of the magnetic field by 90°, probably due to the spurious inhomogeneities of the samples already mentioned. Such a case (No. 178/a) is shown in Fig. 4 too.

It is more interesting to note that while the initial increase of negative magnetoresistance of samples Nos. 176 and 178/b follows a quadratic dependence in the magnetic field as demonstrated in Fig. 6, where the initial part

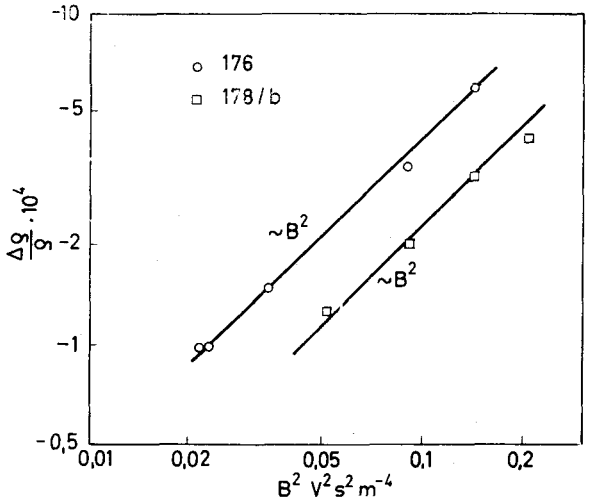


*Fig. 6.* Negative magnetoresistance vs square of magnetic field in Cr-doped samples at 77 °K temperature

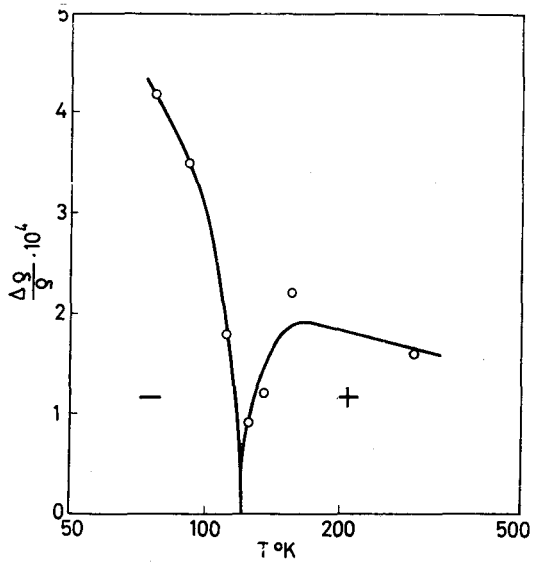


*Fig. 7.* Magnetoresistance of Cr-doped sample No. 176/a at 3000 Gauss vs temperature

of the magnetoresistance curves are shown on a logarithmic scale, at the same time the magnetoresistance of sample No. 178/a exhibits a dependence on the magnetic field which is nearer to the linear. The negative magnetoresistance of O-doped sample No. 177 was too small to draw conclusions concerning its dependence on the magnetic field.

The temperature dependence of magnetoresistance in sample No. 178/a is shown in Fig. 7. At about 120 °K the small field magnetoresistance changes its sign, and above this temperature it turns to be positive. Further, above 150 °K the magnetoresistance begins to be proportional to the square of the magnetic field, and is nearly independent of the temperature. This kind of temperature dependence was observed by several authors in GaAs and in other crystals [2], [4].

## Interpretation of magnetoresistance results

The characteristics of negative magnetoresistance observed in our compensated *n*-type GaAs samples at 77 °K temperature are the same or very similar to the characteristic features of negative magnetoresistance at very low temperature observed by several authors in various materials including GaAs [1], [2], [3], [4], [5], [6], [7]. Only the usually occurring maximum on the Hall coefficient versus temperature curves was not present. We suppose that this is due to the fact that the values of the Hall coefficients are too low to allow to detect a small maximum, the appearance of which is less pronounced for small values of the Hall coefficient [5], [8].

On the basis of this evidence we are justified to suppose that the negative magnetoresistance observed by us in compensated *n*-type GaAs at 77°K temperature and above it is produced by the same cause, namely by the dominant role of impurity conduction in the electron transport of our samples. Now we shall proceed to discuss two interconnected questions. The first of them is the explanation of the existence and characteristic features of negative magnetoresistance observed in our samples on the basis of the various models of impurity conduction put forward in the literature, and the second is how the occurrence of negative magnetoresistance and impurity conduction at 77 °K and higher temperatures, which has been observed up to now only at very low temperatures can be explained.

A great number of theoretical [20], [21], [22], [23], [24], [25] and experimental [1], [2] investigations have shown that there are different types of impurity conduction in semiconductors depending on the concentration of impurities. At low impurity concentrations the electrons are localized at the impurity centres and electrical conduction can take place through the occasional jumps of the localized electrons to neighbouring centres (hopping), [21], with the assistance of phonons [26], if there are nearby vacant centres caused by impurity compensation. For instance, in *n*-type Ge hopping conduction can take place below donor concentrations of $10^{15}$— $10^{16}$ $cm^{-3}$ [27].

Increasing the impurity concentration the electron wave functions localized on different impurity centres begin to overlap with each other, and the localization of electrons becomes obscured. The energy level of impurities

splits, and the activation energy of charge carriers decreases with increasing impurity concentration as observed in $n$-type Ge by J. F. LE HIR [27], by P. DEBYE and E. CONWELL [28], and in $n$-type GaAs by O. V. EMELYANENKO et al. [5] and by D. V. EDDOLS et al. [29]. At the same time, by the overlap of impurity wave functions an impurity band is formed in which the electrons become completely delocalized and can move freely in it. The impurity band is separated from the conduction (or valence) band by an energy gap of the order of the ionization energy of impurities, but this separation decreases with increasing impurity concentration. The formation of impurity band takes place in $n$-type Ge between $1\times10^{16}$ and $2\times10^{17}$ cm$^{-3}$ donor concentrations [27].

Above a certain critical concentration which is about $2\times10^{17}$ cm$^{-3}$ for $n$-type Ge [27], [28] and $(1—2)\times10^{16}$ cm$^{-3}$ for $n$-type GaAs [5], [29], the activation energy of the impurity atoms completely disappears, and at higher concentrations the electrical resistivity is approximately independent of the temperature. At such high impurity concentrations the impurity band and the conduction band (or valence band) merge, and a new type of charge carrier conduction, the metal-like conduction of degenerate electron gas dominates the charge carrier transport mechanism [1], [27].

The transition to this metal-like electron conduction in the impurity-band is observed at impurity concentrations where the interatomic distance between majority impurities is about 2.5 time the effective Bohr radius of the hydrogen-like impurity atoms [1]. This observation is in good agreement with the estimations of N. F. MOTT [20], [30].

According to W. SASAKI [1], and to J. F. WOODS and C. Y. CHEN [2], the various observations of negative magnetoresistance at 4.2 °K were all made in the impurity conduction range of dopings and among the various materials all the three ranges of impurity conduction are represented. Notably in the case of GaAs all the three ranges were observed in [1], [2], [5] and [6].

The various possible mechanisms giving rise to negative magnetoresistance were briefly analyzed by J. F. WOODS and C. Y. CHEN [2]. According to them it seems to be only two possible kinds of mechanisms which could account for negative magnetoresistance in the impurity conduction range. Either the magnetic field gives rise to a redistribution of charge carriers among different energy states having different carrier mobility, or the magnetic field increases the mobility or hopping probability of electrons. It is not necessary for these mechanisms to be mutually exclusive, but it is probable that in the different ranges of impurity conduction (i.e. hopping conduction, transition range and impurity band conduction) different mechanisms should play the leading role.

As was shown by R. J. SLADEK and R. W. KEYES [31], and by N. MIKOSHIBA [32], the magnetic field reduces the mobility or hopping probability in the hopping (or transition) region, due to the shrinkage of wave functions

of impurities thus reducing the orbital overlap. To interpret the results of measurements in this range it seems inevitable to postulate a model with energy states having different mobilities in order to account for the observed negative magnetoresistance, at least in the absence of impurity band conduction [2]. According to J. F. WOODS and C. Y. CHEN [2], the general character of the magnetoresistance observations is consonant with a two energy state model of impurity conduction, in which the energy difference between the two states is reduced by the application of a magnetic field thus increasing the occupation probability of the higher energy states which presumably have a higher mobility than the ground states due to the greater orbital overlap of wave functions. A competing process would be the restriction of orbital overlap as shown by R. J. SLADEK and R. W. KEYES [31] thus the resistance change would be the outcome of the results of the influence of two competing processes. This model was put forward by J. F. WOODS and C. Y. CHEN [2], when interpreting their measurements of negative magnetoresistance of *n*- and *p*-type GaAs at liquid He temperatures, which in low magnetic field was approximately linear in the field, in contrast with other observations, where a quadratic dependence was found in a wide range of impurity concentrations [1], [5], [6].

A two-state model would, in general, predict a variation of the Hall coefficient with the magnetic field. However, since the Hall effect was not measured in the hopping region so far this prediction could not be verified [2]. The two-state model in principle could be applied to the impurity band conduction region too but in this range in *n*-type GaAs no significant variation of the Hall coefficient with the magnetic field could be found [2], [33]. We should like to note that in our measurements too a variation of the Hall coefficient with the magnetic field was not detected.

The only successful model of mobility increase mechanism was put forward by Y. TOYOZAWA [22], [23], which can account for the negative magnetoresistance in the impurity band and metallic conduction region [1], but it is incapable to explain the features of negative magnetoresistance in the hopping conduction region. According to this model there exist localized magnetic moments in the crystal, which obey a Curie—Weiss-type law, and the magnetic scattering of the electrons moving in the impurity band by these localized magnetic moments results in the negative magnetoresistance. For highly doped (*n*-type) semiconductors it is believed that the magnetic moments causing this behaviour are the spins of electrons semilocalized around the donor impurities [22], [23].

According to this model some impurity atoms (donors in *n*-type semiconductors) are close enough to each other so that the electronic states form an impurity band. Conduction is via electrons in this band and is limited by the scattering of electrons. Due to the random distribution of impurities, however, some impurities are far enough from each other for the electrons

to be localized around them most of the time, giving rise to localized magnetic moments around the impurity atoms, which interact with the other electrons, thus causing a scattering of charge carriers in the impurity band. In magnetic fields these magnetic moments become aligned and the amount of scattering caused by them will be reduced, and the resistivity will decrease. The resistivity change is approximately proportional to the square of the average magnetization of these impurities, i.e.

$$\frac{\Delta\varrho}{\varrho_0} = \frac{\varrho(B) - \varrho(0)}{\varrho(0)} \sim -\langle m\rangle^2,$$

where $\langle m\rangle$ is the average magnetization [22], [23]. For small and large magnetic fields, respectively, this yields

$$\frac{\varrho(B) - \varrho(0)}{\varrho(0)} \sim -\left(\frac{\mu B}{kT}\right)^2 \text{ (for small } B),$$

$$\frac{\varrho(B) - \varrho(0)}{\varrho(0)} = \text{const.} \qquad \text{(for large } B),$$

where $\mu$ is the magnetic moment localized in the impurities [22], [23]. The basic features of this model, i.e. the quadratic dependence of the negative magnetoresistance at the lowest fields, saturation in larger fields, and the increase of the negative magnetoresistance with decreasing temperature are all in good agreement with the bulk of the experimental data [1], [5], [6], [22], [23]. Because the localized magnetic moments obey a Curie—Weiss-type law, the temperature dependence of the negative magnetoresistance in this model at low temperatures is the following, [22], [23]

$$\frac{\varrho(B) - \varrho(0)}{\varrho(0)} \sim -\frac{1}{(T+\Theta)^2}.$$

The constant $\Theta$ appears due to the coupling between the magnetic moments, and its experimental values are positive, of the order of 1—2 °K, showing an antiferromagnetic interaction between the localized magnetic moments [1], [6], [22], [23].

In our samples the donor concentrations of Cr- and O-doped samples were in the range of $N_d = (1\text{—}2.4)\times 10^{18}\ \text{cm}^{-3}$, so the average distance between the donor impurities is $r_d = (3/4\pi N_d)^{1.3} = (0.45\text{—}0.65)\times 10^{-6}$ cm, while the effective Bohr radius is $r_B = a_0\varepsilon_r m_0/m_* = 0.85\times 10^{-6}$ cm with $a_0 = \hbar^2/m_0e^2$ and with $\varepsilon_r = 11.5$ and $m_*/m_0 = 0.072$ [14]. Thus the average distance between the donor impurities is of the order of the effective Bohr radius, therefore our samples are all in the metal-like impurity band conduction range [1], [20], where the model of the localized magnetic moments by Y. TOYOZAWA [22], [23] can be applied [1].

The results of our measurements of negative magnetoresistance, i.e. the quadratic field dependence observed in the Cr-doped samples, the saturation at moderate fields, the steep decrease of the negative component with increasing magnetic field, all agree well with the predictions of Y. TOYOZAWA's model. This interpretation of the results for the Cr-doped samples has been already put forward by us in [9] without detailed analysis.

On the basis of the formalism developed by Y. TOYOZAWA in [23] we can estimate the average magnitude of the magnetic moments localized on the impurities, using the measured values of the weak field negative magnetoresistance, and of the saturation value of the magnetoresistance. We can make only a very crude estimation because the saturation value of the negative magnetoresistance is not very well determined, due to the presence of the common positive component of the magnetoresistance. On the other hand, the impurity concentration in our samples is somewhat greater than the impurity concentrations for which Y. TOYOZAWA's model was originally applied [23], and the strong compensation of our samples can reduce the reliability of this analysis.

For the localized magnetic moments of the impurity centres we got values of the order of 2—3 Bohr magneton. These values seem to be in good agreement with other results [23]. This analysis applied to other cases gave values for the magnetic moments of the order of 5—10 Bohr magneton [22], [23]. In his model this great value was explained by Y. TOYOZAWA [23], as being the result of the statistical distribution of the magnetic moments of localized spins due to their collective nature. According to Y. TOYOZAWA on the one hand, they are caused by the electrons which are constantly entering and leaving the magnetic sites, and on the other hand, the surrounding non-magnetic sites make an additional contribution to the magnetic moments. In our samples it is possible that the magnetic moments of the Cr impurities too play a role in determining the interaction of electrons with the localized magnetic moments.

The resistivity in the metal-like impurity band was estimated by N. F. MOTT and W. D. TWOSE [20]. The conduction electrons form a degenerate electron gas with the same density of states effective mass as that of the conduction band electrons [1]. For a degenerate Fermi gas the conductivity is

$$\sigma = \frac{ne^2 l}{vm_*},$$

where $l$ and $v$ are the mean free path and the electron velocity at the surface of Fermi distribution. For degenerate electron gas we have the formula

$$\frac{vm_*}{\hbar} = 2\pi \left(\frac{3n}{8\pi}\right)^{1/3}.$$

Supposing that the mean free path can be written as

$$l = \frac{p}{n^{1/3}},$$

where $p$ is the number of interatomic distances on a mean free path. Combining these expressions we get for the conductivity

$$\sigma = \frac{n^{1/3} e^2 p}{\hbar \pi} \left(\frac{\pi}{3}\right)^{1/2}.$$

These formulas were applied by N. F. MOTT and W. D. TWOSE [20] to the case of $n$-type Ge with donor concentrations between $2 \times 10^{17}$ and $4 \times 10^{19}$ cm$^{-3}$ at liquid He temperatures, and they have got an electron mean free path which was nearly constant in the whole range of impurity concentrations.

We have applied this model to our Cr- and O-doped samples and the results are shown in Table II. The reduced Fermi levels $\zeta = E_F/kT$ which are also shown in Table II were evaluated with the assumption that the density of states in the metal-like impurity band is the same as in the conduction band [1], to show that in our samples at 77 °K the electron gas is really degenerate. Except for sample No. 178/b we can see that the mean free path does not depend strongly on the electron donor concentrations. Apparently the assumption concerning the degeneracy does not hold for sample No. 178/b.

Our last task is to explain the occurrence of metal-like impurity band conduction and of the negative magnetoresistance in our samples at relatively high temperatures (77 °K and above). The fact that our samples were strongly compensated, presumably plays a significant role in the formation of impurity band at higher temperature than in other cases. According to O. V. EMELYANENKO et al., [7], in their $n$-type GaAs samples which were doped with Cu diffusion, the strong compensation produced relatively high concentrations of impurities with lower concentrations of charge carriers, thus giving rise to the formation of impurity band at higher temperatures than it is usual. The same is the case with our Cr- and O-doped and strongly compensated samples, where the metal-like conduction mechanism shows that the conduction and impurity bands merged, and most probably with the samples of B. S. LISENKER et al., [8], having Fe as the compensating impurity.

It is very interesting to note that until now the formation of impurity band and the presence of negative magnetoresistance at relatively higher temperatures (77 °K and above) have been observed only in highly compensated $n$-type GaAs, with compensating impurities having deep energy levels. As we have mentioned Cr creates deep acceptor levels in GaAs with energy level of 0.81 eV below the conduction band edge, Fe creates deep acceptors too with

energy level most probably at 0.52 eV but at the same time another level of 0.37 eV was observed too [18]. It seems that there are more than one deep acceptor levels associated with the Cu impurity [34], and the acceptor level connected with the O impurity lies about in the middle of the forbidden band at 0.63 eV above the valence band edge [35].

**Table II**

| Sample No | $\sigma$ $\Omega^{-1}$ cm$^{-1}$ | $n$ cm$^{-3}$ | $P$ | $l$ cm | $\zeta$ |
|---|---|---|---|---|---|
| 176 | 323 | $6.1\times10^{17}$ | 48.8 | $58\times10^{-6}$ | 5.3 |
| 178/*a* | 358 | $1.2\times10^{18}$ | 43.3 | $41\times10^{-6}$ | 8.3 |
| 178/*b* | 71 | $3.6\times10^{17}$ | 12.9 | $18\times10^{-6}$ | 3.6 |
| 177 | 337 | $1.6\times10^{18}$ | 37.1 | $32\times10^{-6}$ | 10.2 |

Finally we would like to point out that in the compensated samples the mobility at 77 °K is usually exceptionally low as observed in our samples Nos. 177, 178/a and 178/b and therefore the normal positive component of the magnetoresistance is very low too, and cannot mask the not too strong negative magnetoresistance.

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## ОТРИЦАТЕЛЬНОЕ МАГНЕТОСОПРОТИВЛЕНИЕ В КОМПЕНСИРОВАННОМ АРСЕНИДЕ ГАЛЛИЯ *n*-ТИПА ПРИ ТЕМПЕРАТУРЕ 77 °К

Б. ПЭДЭР

### Резюме

Магнетосопротивление в сильно компенсированном арсениде галлия *n*-типа с примесью хрома и кислорода было измерено при температуре 77 °К. Концентрация электронов при комнатной температуре находилась в пределах от $3{,}8\times10^{17}$ до $1{,}6\times10^{18}$ $см^{-3}$. При слабых магнитных полях отрицательное магнетосопротивление было обнаружено при температуре жидкого азота. Было показано, что поведение этого аномального отрицательного магнетосопротивления очень похоже на то, которое было обнаружено в сильно дегивованных полупроводниках при гелиевых температурах. Отрицательное магнетосопротивление было пропорционально квадрату магнитного поля при слабых полях, и показало насыщение при полях 4000—6000 гаусс. Для объяснения обнаруженного эффекта было предположено, что главным механизмом переноса носителей тока в наших образцах является металлическая проводимость в примесной зоне.

Было показано, что обнаруженные характерные черты отрицательного магнетосопротивления совместимы с моделью рассеяния электронов на локализированных магнитных моментах в примесной зоне. Из экспериментальных данных среднее значение локализированных моментов составляет порядка 2—3 магнетона Бора.

Было высказано предположение, что сильная компенсация может играть существенную роль в формировании примесной зоны при температурах 77 °К и выше.